

Total	4,227	22,943	30,985	38,924	54,739	62,272	74,519
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^a See Annex 3.2, Table A-88 for additional information on transportation consumption of these fuels.

Methodology

Woody biomass emissions were estimated by applying two EIA gross heat contents (Lindstrom 2006) to U.S. consumption data (see Table 3-58), provided in energy units. This year woody biomass consumption data for the industrial, residential, and commercial sectors were obtained from EIA 2011, while woody biomass consumption data for the electricity generation sector was estimated from EPA's Clean Air Market Acid Rain Program dataset (EPA 2011). The bottom-up analysis of woody biomass consumption based on EPA's Acid Rain Program dataset indicated that the amount of woody biomass consumption allocated in the EIA statistics should be adjusted. Therefore, for these estimates, the electricity generation sector's woody biomass consumption was adjusted downward to match the value obtained from the bottom-up analysis based on EPA's Acid Rain Program dataset. As the total woody biomass consumption estimate from EIA is considered to be accurate at the national level, the woody biomass consumption totals for the industrial, residential, and commercial sectors were adjusted upward proportionately.

One heat content (16.95 MMBtu/MT wood and wood waste) was applied to the industrial sector's consumption, while the other heat content (15.43 MMBtu/MT wood and wood waste) was applied to the consumption data for the other sectors. An EIA emission factor of 0.434 MT C/MT wood (Lindstrom 2006) was then applied to the resulting quantities of woody biomass to obtain CO₂ emission estimates. It was assumed that the woody biomass contains black liquor and other wood wastes, has a moisture content of 12 percent, and is converted into CO₂ with 100 percent efficiency. The emissions from ethanol consumption were calculated by applying an emission factor of 18.67 Tg C/QBtu (EPA 2010) to U.S. ethanol consumption estimates that were provided in energy units (EIA 2011) (see Table 3-59).

Table 3-58: Woody Biomass Consumption by Sector (Trillion Btu)

End-Use Sector	1990	2005	2006	2007	2008	2009	2010
Industrial	1,525.8	1,580.8	1,598.4	1,533.3	1,452.3	1,308.8	1,426.2
Residential	613.7	468.2	423.4	466.6	486.3	469.8	458.3
Commercial	69.6	75.8	70.3	75.3	78.9	79.1	76.7
Electricity Generation	7.1	11.5	16.9	23.2	26.7	22.8	24.7
Total	2,216.2	2,136.4	2,108.9	2,098.5	2,044.2	1,880.5	1,985.9

Table 3-59: Ethanol Consumption by Sector (Trillion Btu)

End-Use Sector	1990	2005	2006	2007	2008	2009	2010
Transportation	60.4	327.4	441.7	556.8	785.8	893.9	1,069.7
Industrial	0.8	6.8	9.7	9.8	11.6	13.0	15.5
Commercial	0.5	0.9	1.3	2.0	2.1	2.8	3.4
Total	61.7	335.1	452.6	568.6	799.6	909.7	1,088.6

Uncertainty and Time-Series Consistency

It is assumed that the combustion efficiency for woody biomass is 100 percent, which is believed to be an overestimate of the efficiency of wood combustion processes in the United States. Decreasing the combustion efficiency would decrease emission estimates. Additionally, the heat content applied to the consumption of woody biomass in the residential, commercial, and electric power sectors is unlikely to be a completely accurate representation of the heat content for all the different types of woody biomass consumed within these sectors. Emission estimates from ethanol production are more certain than estimates from woody biomass consumption due to better activity data collection methods and uniform combustion techniques.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2009. Details on the emission trends through time are described in more detail in the Methodology section,

above.

Recalculations Discussion

Wood and ethanol consumption values were revised relative to the previous Inventory for 2009 based on updated information from EIA's Annual Energy Review (EIA 2011). Additionally, the change in methodology for calculating emissions from woody biomass led a decrease in emissions from the electricity generation sector and an increase in emissions for the other sectors over the time series. This adjustment of historical data for wood biomass consumption resulted in an average annual decrease in emissions from wood biomass consumption of about 1.0 Tg CO₂ Eq. (0.5 percent) from 1990 through 2009. Slight adjustments were made to ethanol consumption based on updated information from EIA (2011), which slightly increased estimates for ethanol consumed. As a result of adjustments to historical EIA data, average annual emissions from ethanol consumption increased by less than 0.05 Tg CO₂ Eq. (less than 0.05 percent) relative to the previous Inventory.

Planned Improvements

The availability of facility-level combustion emissions through EPA's GHGRP will be examined to help better characterize the industrial sector's energy consumption in the United States, and further classify business establishments according to industrial economic activity type. Most methodologies used in EPA's GHGRP are consistent with IPCC, though for EPA's GHGRP, facilities collect detailed information specific to their operations according to detailed measurement standards, which may differ with the more aggregated data collected for the Inventory to estimate total, national U.S. emissions. In addition, and unlike the reporting requirements for this chapter under the UNFCCC reporting guidelines,¹⁰⁹ some facility-level fuel combustion emissions reported under the GHGRP may also include industrial process emissions. In line with UNFCCC reporting guidelines, fuel combustion emissions are included in this chapter, while process emissions are included in the Industrial Processes chapter of this report. In examining data from EPA's GHGRP that would be useful to improve the emission estimates for the CO₂ from biomass combustion category, particular attention will also be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all inventory years as reported in this inventory. Additionally, analyses will focus on aligning reported facility-level fuel types and IPCC fuel types per the national energy statistics, ensuring CO₂ emissions from biomass are separated in the facility-level reported data, and maintaining consistency with national energy statistics provided by EIA. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹¹⁰

¹⁰⁹ See <<http://unfccc.int/resource/docs/2006/sbsta/eng/09.pdf>>

¹¹⁰ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

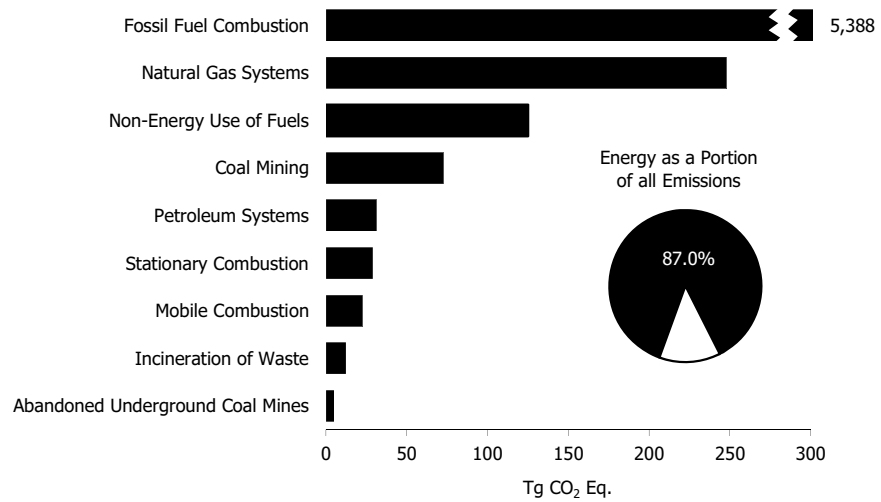


Figure 3-1: 2010 Energy Chapter Greenhouse Gas Sources

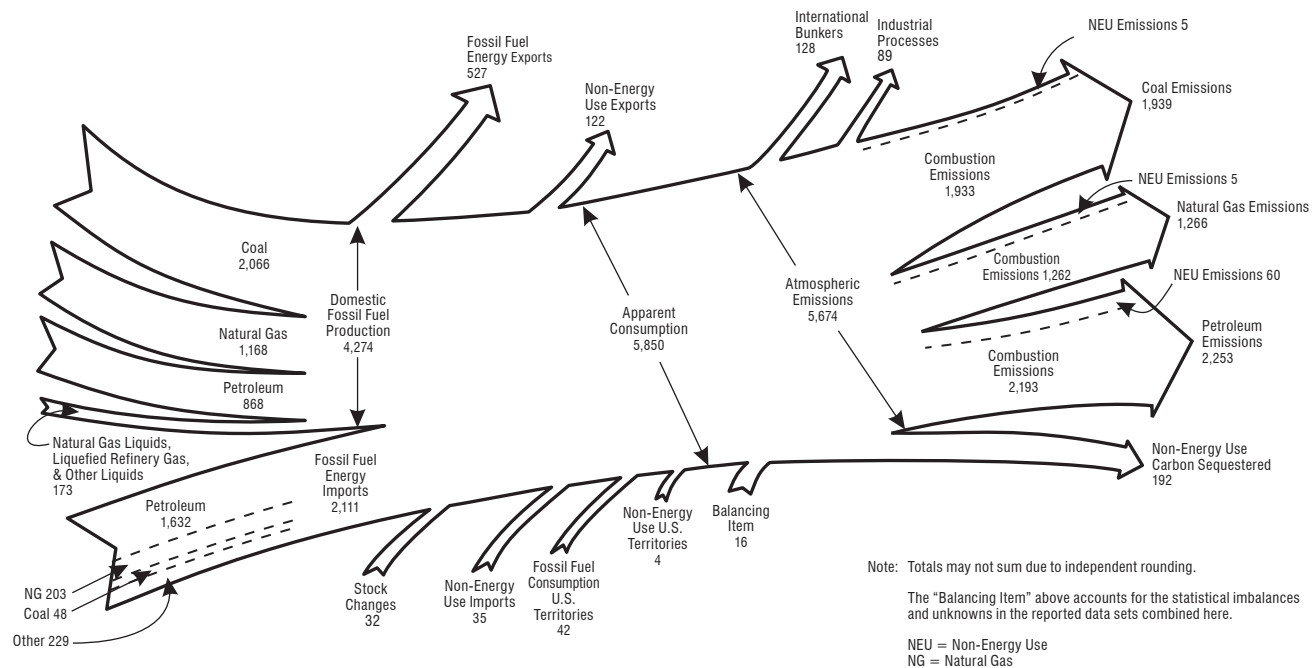


Figure 3-2 2010 U.S. Fossil Carbon Flows (Tg CO₂ Eq.)

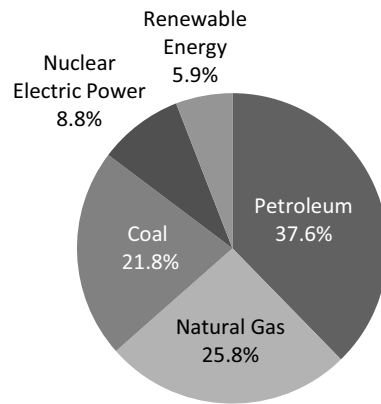


Figure 3-3: 2010 U.S. Energy Consumption by Energy Source

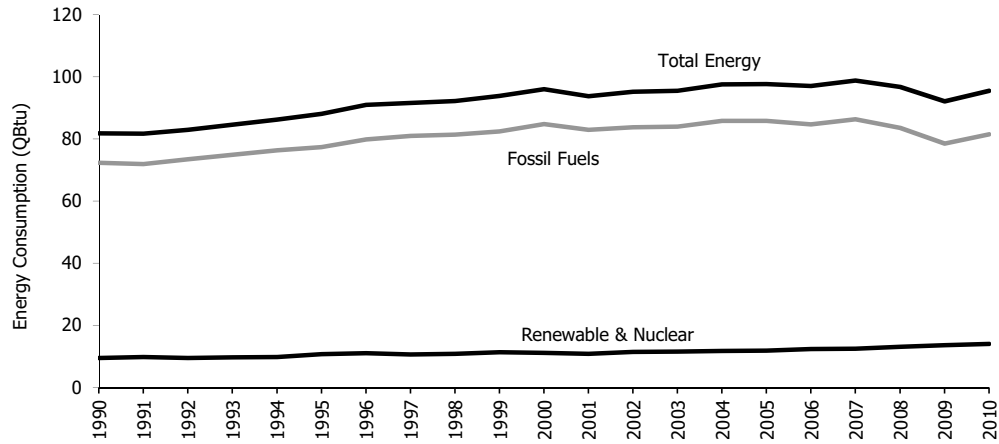


Figure 3-4: U.S. Energy Consumption (Quadrillion Btu)

Note: Expressed as gross calorific values.

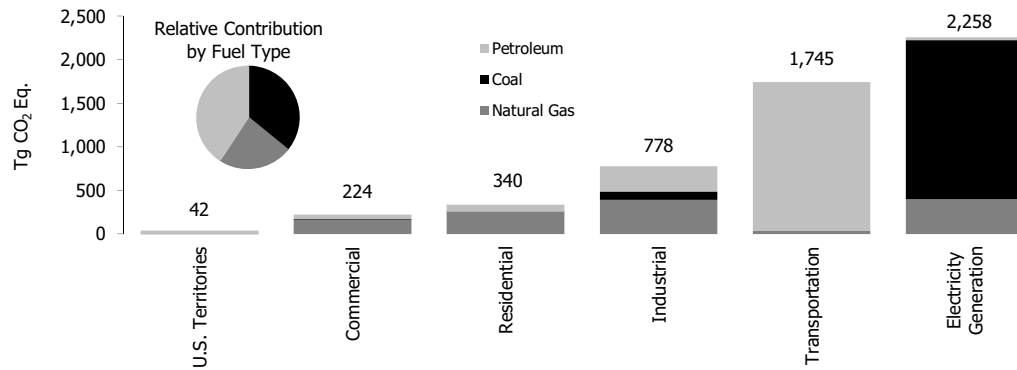


Figure 3-5: 2010 CO₂ Emissions from Fossil Fuel Combustion by Sector and Fuel Type

Note: The electricity generation sector also includes emissions of less than 0.5 Tg CO₂ Eq. from geothermal-based electricity generation.

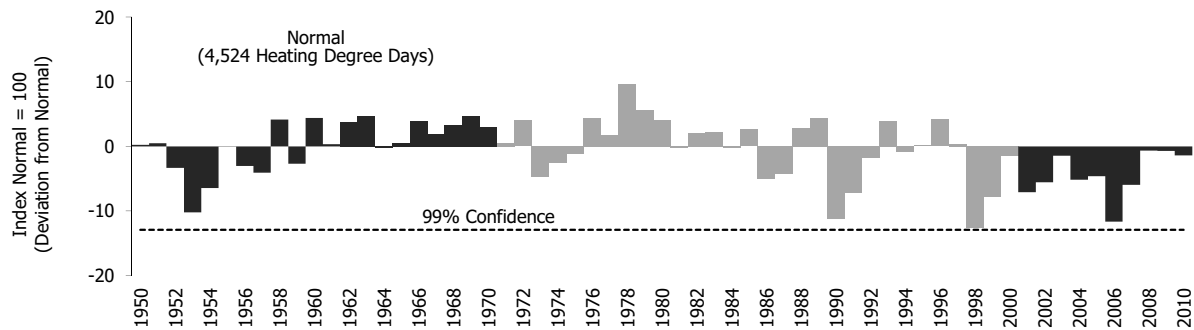


Figure 3-6: Annual Deviations from Normal Heating Degree Days for the United States (1950-2010)

Note: Climatological normal data are highlighted.

Statistical confidence interval for "normal" climatology period of 1971 through 2000.

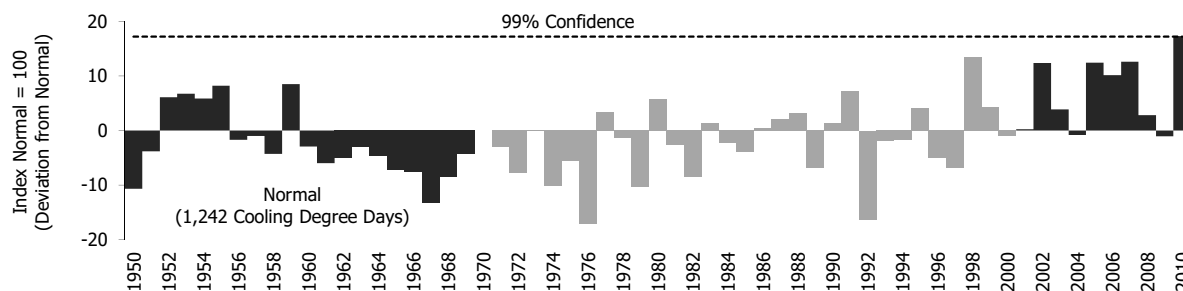


Figure 3-7: Annual Deviations from Normal Cooling Degree Days for the United States (1950-2010)

Note: Climatological normal data are highlighted.

Statistical confidence interval for "normal" climatology period of 1971 through 2000.

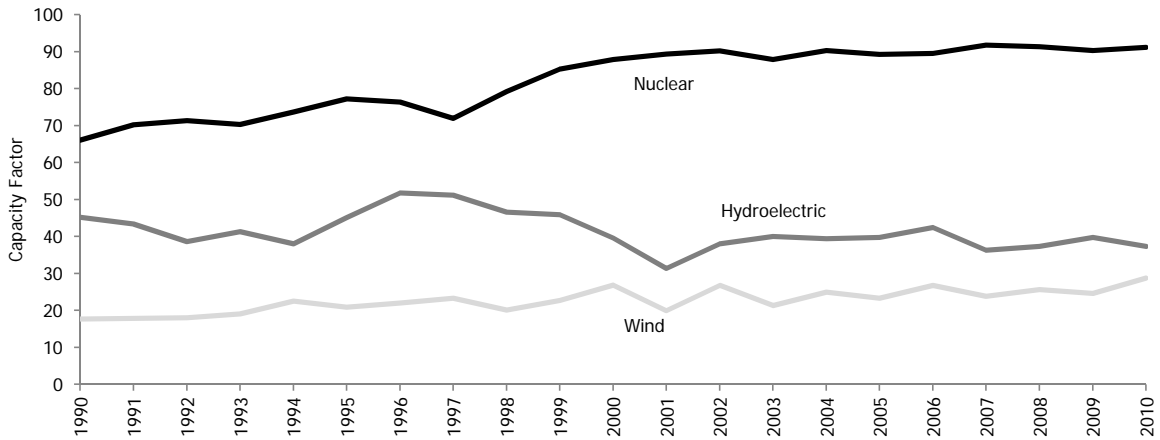


Figure 3-8: Nuclear, Hydroelectric, and Wind Power Plant Capacity Factors in the United States (1990-2010)

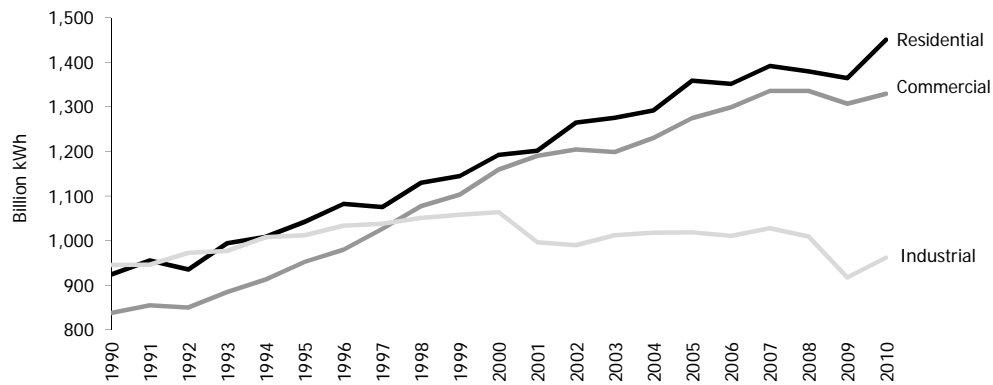


Figure 3-9: Electric Generation Retail Sales by End-Use Sector
 Note: The transportation end-use sector consumes minor quantities of electricity.

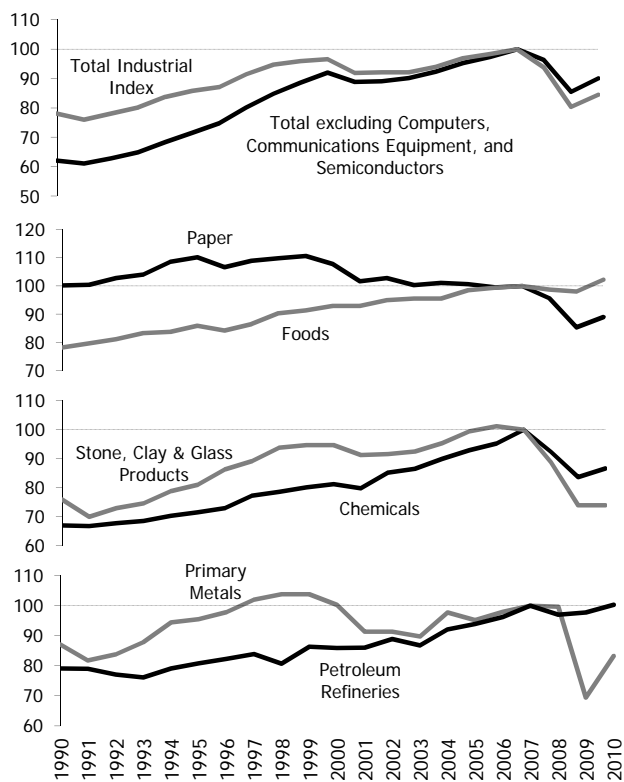


Figure 3-10: Industrial Production Indexes (Index 2007=100)

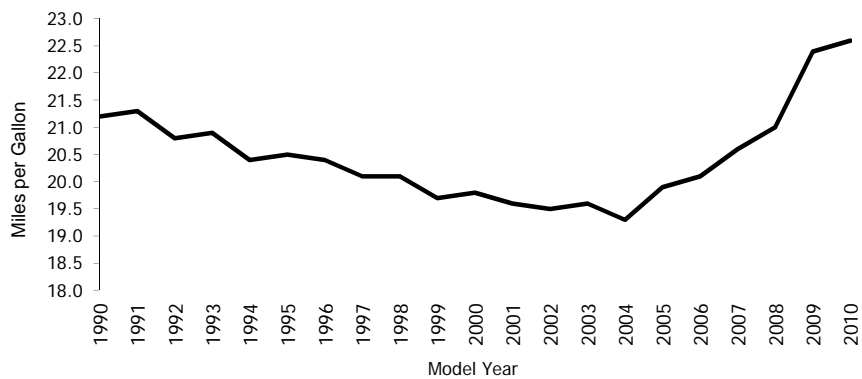


Figure 3-11: Sales-Weighted Fuel Economy of New Passenger Cars and Light-Duty Trucks, 1990-2010

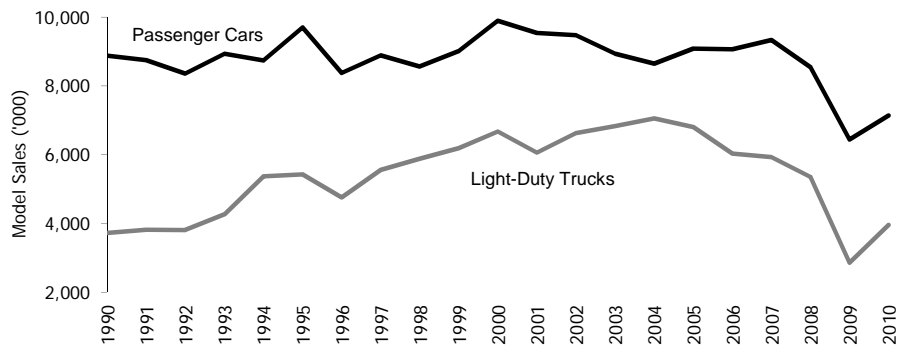


Figure 3-12: Sales of New Passenger Cars and Light-Duty Trucks, 1990-2010

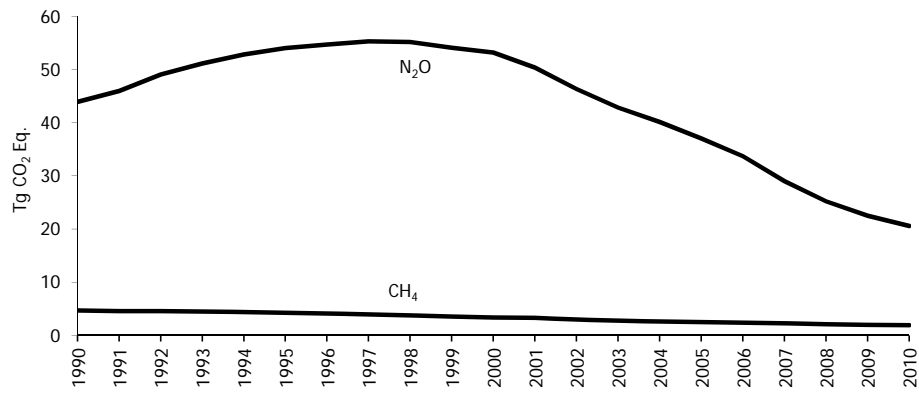


Figure 3-13: Mobile Source CH₄ and N₂O Emissions

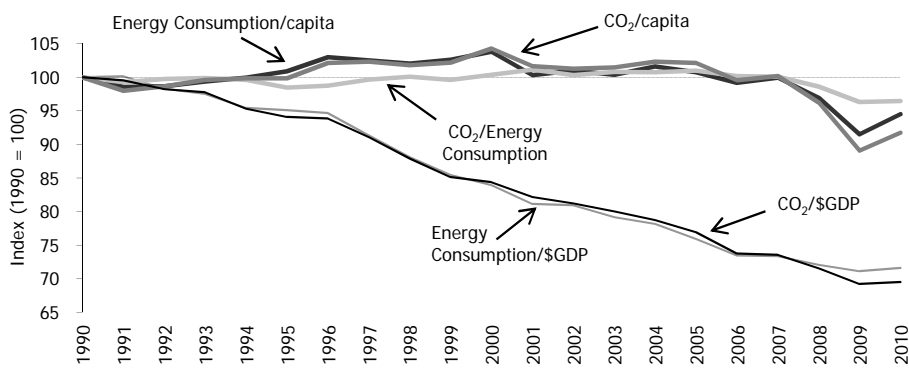


Figure 3-14: U.S. Energy Consumption and Energy-Related CO₂ Emissions Per Capita and Per Dollar GDP

4. Industrial Processes

Greenhouse gas emissions are produced as the by-products of various non-energy-related industrial activities. That is, these emissions are produced from an industrial process itself and are not directly a result of energy consumed during the process. For example, raw materials can be chemically transformed from one state to another. This transformation can result in the release of greenhouse gases such as carbon dioxide (CO₂), methane (CH₄), and nitrous oxide (N₂O). The processes addressed in this chapter include iron and steel production and metallurgical coke production, cement production, lime production, ammonia production and urea consumption, limestone and dolomite consumption (e.g., flux stone, flue gas desulfurization, and glass manufacturing), soda ash production and use, aluminum production, titanium dioxide production, CO₂ consumption, ferroalloy production, phosphoric acid production, zinc production, lead production, petrochemical production, silicon carbide production and consumption, nitric acid production, and adipic acid production (see Figure 4-1).

Figure 4-1: 2010 Industrial Processes Chapter Greenhouse Gas Sources

In addition to the three greenhouse gases listed above, there are also industrial sources of man-made fluorinated compounds called hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), and sulfur hexafluoride (SF₆). The present contribution of these gases to the radiative forcing effect of all anthropogenic greenhouse gases is small; however, because of their extremely long lifetimes, many of them will continue to accumulate in the atmosphere as long as emissions continue. In addition, many of these gases have high global warming potentials; SF₆ is the most potent greenhouse gas the Intergovernmental Panel on Climate Change (IPCC) has evaluated. Usage of HFCs is growing rapidly since they are the primary substitutes for ozone depleting substances (ODSs), which are being phased-out under the Montreal Protocol on Substances that Deplete the Ozone Layer. In addition to their use as ODS substitutes, HFCs, PFCs, and SF₆ are employed and emitted by a number of other industrial sources in the United States. These industries include aluminum production, HCFC-22 production, semiconductor manufacture, electric power transmission and distribution, and magnesium metal production and processing.

In 2010, industrial processes generated emissions of 303.4 teragrams of CO₂ equivalent (Tg CO₂ Eq.), or 4.4 percent of total U.S. greenhouse gas emissions. Carbon dioxide emissions from all industrial processes were 139.7 Tg CO₂ Eq. (139,726 Gg) in 2010, or 2.4 percent of total U.S. CO₂ emissions. Methane emissions from industrial processes resulted in emissions of approximately 1.5 Tg CO₂ Eq. (69 Gg) in 2010, which was less than 1 percent of U.S. CH₄ emissions. N₂O emissions from adipic acid and nitric acid production were 19.5 Tg CO₂ Eq. (63 Gg) in 2010, or 6.4 percent of total U.S. N₂O emissions. In 2010 combined emissions of HFCs, PFCs, and SF₆ totaled 142.7 Tg CO₂ Eq. Total emissions from Industrial Processes in 2010 were 3.4 percent less than 1990 emissions.

The slight decrease in overall Industrial Processes emissions since 1990 reflects a range of emission trends among the industrial process emission sources. Emissions resulting from most types of metal production have declined significantly since 1990 but largely due to production shifting to other countries, but also due to transitions to less-emissive methods of production (in the case of iron and steel) and to improved practices (in the case of PFC emissions from aluminum production). Emissions from mineral sources have either increased or not changed significantly since 1990 but largely track economic cycles, while CO₂ and CH₄ emissions from chemical sources have either decreased or not changed significantly. HFC emissions from the substitution of ozone depleting substances have increased drastically since 1990, while the emission trends of HFCs, PFCs, and SF₆ from other sources are mixed. Trends are explained further within each emission category throughout the chapter.

Table 4-1 summarizes emissions for the Industrial Processes chapter in Tg CO₂ Eq., while unweighted native gas emissions in Gg are provided in Table 4-2. The source descriptions that follow in the chapter are presented in the order as reported to the UNFCCC in the common reporting format tables, corresponding generally to: mineral products, chemical production, metal production, and emissions from the uses of HFCs, PFCs, and SF₆.

Table 4-1: Emissions from Industrial Processes (Tg CO₂ Eq.)

Gas/Source	1990	2005	2006	2007	2008	2009	2010
CO₂	188.5	165.4	169.9	172.6	159.5	118.1	139.7
Iron and Steel Production and Metallurgical Coke Production	99.6	66.0	68.9	71.1	66.1	42.1	54.3
<i>Iron and Steel Production</i>	97.1	64.0	66.9	69.1	63.8	41.2	52.2
<i>Metallurgical Coke Production</i>	2.5	2.0	1.9	2.1	2.3	1.0	2.1
Cement Production	33.3	45.2	45.8	44.5	40.5	29.0	30.5
Lime Production	11.5	14.4	15.1	14.6	14.3	11.2	13.2
Limestone and Dolomite Use	5.1	6.8	8.0	7.7	6.3	7.6	10.0
Ammonia Production	13.0	9.2	8.8	9.1	7.9	7.9	8.7
Urea Consumption for Non-Agricultural Purposes	3.8	3.7	3.5	4.9	4.1	3.4	4.4
Soda Ash Production and Consumption	4.1	4.2	4.2	4.1	4.1	3.6	3.7
Petrochemical Production	3.3	4.2	3.8	3.9	3.4	2.7	3.3
Aluminum Production	6.8	4.1	3.8	4.3	4.5	3.0	3.0
Carbon Dioxide Consumption	1.4	1.3	1.7	1.9	1.8	1.8	2.2
Titanium Dioxide Production	1.2	1.8	1.8	1.9	1.8	1.6	1.9
Ferroalloy Production	2.2	1.4	1.5	1.6	1.6	1.5	1.7
Zinc Production	0.6	1.0	1.0	1.0	1.2	0.9	1.2
Phosphoric Acid Production	1.5	1.4	1.2	1.2	1.2	1.0	1.0
Lead Production	0.5	0.6	0.6	0.6	0.5	0.5	0.5
Silicon Carbide Production and Consumption	0.4	0.2	0.2	0.2	0.2	0.1	0.2
CH₄	1.9	1.8	1.7	1.7	1.6	1.2	1.5
Petrochemical Production	0.9	1.1	1.0	1.0	0.9	0.8	0.9
Iron and Steel Production and Metallurgical Coke Production	1.0	0.7	0.7	0.7	0.6	0.4	0.5
<i>Iron and Steel Production</i>	1.0	0.7	0.7	0.7	0.6	0.4	0.5
<i>Metallurgical Coke Production</i>	+	+	+	+	+	+	+
Ferroalloy Production	+	+	+	+	+	+	+
Silicon Carbide Production and Consumption	+	+	+	+	+	+	+
N₂O	33.4	23.9	25.0	29.8	18.9	17.3	19.5
Nitric Acid Production	17.6	16.4	16.1	19.2	16.4	14.5	16.7
Adipic Acid Production	15.8	7.4	8.9	10.7	2.6	2.8	2.8
HFCs	36.9	115.0	116.0	120.0	117.5	112.0	123.0
Substitution of Ozone Depleting Substances ^a	0.3	99.0	101.9	102.7	103.6	106.3	114.6
HCFC-22 Production	36.4	15.8	13.8	17.0	13.6	5.4	8.1
Semiconductor Manufacturing HFCs	0.2	0.2	0.3	0.3	0.3	0.3	0.3
PFCs	20.6	6.2	6.0	7.5	6.7	5.6	5.7
Semiconductor Manufacturing PFCs	2.2	3.2	3.5	3.7	4.0	4.0	4.1
Aluminum Production	18.4	3.0	2.5	3.8	2.7	1.6	1.6
SF₆	32.6	17.8	16.8	15.6	15.0	13.9	14.0
Electrical Transmission and Distribution	26.7	13.9	13.0	12.2	12.2	11.8	11.8
Magnesium Production and Processing	5.4	2.9	2.9	2.6	1.9	1.1	1.3
Semiconductor Manufacturing SF ₆	0.5	1.0	1.0	0.8	0.9	1.0	0.9
Total	313.9	330.1	335.5	347.3	319.1	268.2	303.4

+ Does not exceed 0.05 Tg CO₂ Eq.

Note: Totals may not sum due to independent rounding.

^a Small amounts of PFC emissions also result from this source.

Table 4-2: Emissions from Industrial Processes (Gg)

Gas/Source	1990	2005	2006	2007	2008	2009	2010
CO₂	188,460	165,402	169,895	172,609	159,457	118,105	139,726
Iron and Steel Production and Metallurgical Coke Production	99,593	66,000	68,854	71,138	66,092	42,113	54,276
<i>Iron and Steel Production</i>	97,123	63,957	66,934	69,083	63,758	41,157	52,192
<i>Metallurgical Coke Production</i>	2,470	2,043	1,919	2,055	2,334	956	2,084
Cement Production	33,278	45,197	45,792	44,538	40,531	29,018	30,509
Lime Production	11,533	14,379	15,100	14,595	14,330	11,225	13,151
Limestone and Dolomite Use	5,127	6,768	8,035	7,702	6,276	7,649	10,017
Ammonia Production	13,047	9,196	8,781	9,074	7,883	7,855	8,678
Urea Consumption for Non-Agricultural Purposes	3,784	3,653	3,519	4,944	4,065	3,415	4,365
Soda Ash Production and Consumption	4,141	4,228	4,162	4,140	4,099	3,554	3,735
Petrochemical Production	3,311	4,181	3,837	3,931	3,449	2,735	3,336
Aluminum Production	6,831	4,142	3,801	4,251	4,477	3,009	3,009
Carbon Dioxide Consumption	1,416	1,321	1,709	1,867	1,780	1,784	2,203
Titanium Dioxide Production	1,195	1,755	1,836	1,930	1,809	1,648	1,876
Ferroalloy Production	2,152	1,392	1,505	1,552	1,599	1,469	1,663
Zinc Production	632	1,030	1,030	1,025	1,159	943	1,168
Phosphoric Acid Production	1,529	1,386	1,167	1,166	1,187	1,018	1,017
Lead Production	516	553	560	562	547	525	542
Silicon Carbide Production and Consumption	375	219	207	196	175	145	181
CH₄	88	86	83	82	74	58	69
Petrochemical Production	41	51	48	48	43	39	44
Iron and Steel Production and Metallurgical Coke Production	46	34	35	33	31	17	25
<i>Iron and Steel Production</i>	46	34	35	33	31	17	25
<i>Metallurgical Coke Production</i>	+	+	+	+	+	+	+
Ferroalloy Production	1	+	+	+	+	+	+
Silicon Carbide Production and Consumption	1	+	+	+	+	+	+
N₂O	108	77	81	96	61	56	63
Nitric Acid Production	57	53	52	62	53	47	54
Adipic Acid Production	51	24	29	34	8	9	9
HFCs	M	M	M	M	M	M	M
Substitution of Ozone Depleting Substances ^a	M	M	M	M	M	M	M
HCFC-22 Production	3	1	1	1	1	+	1
Semiconductor Manufacturing HFCs	+	+	+	+	+	+	+
PFCs	M	M	M	M	M	M	M
Semiconductor Manufacturing PFCs	M	M	M	M	M	M	M
Aluminum Production	M	M	M	M	M	M	M
SF₆	1	1	1	1	1	1	1
Electrical Transmission and Distribution	1	1	1	1	1	+	+
Magnesium Production and Processing	M	M	M	M	M	M	M
Semiconductor Manufacturing SF ₆	+	+	+	+	+	+	+

+ Does not exceed 0.5 Gg

M (Mixture of gases)

Note: Totals may not sum due to independent rounding.

^a Small amounts of PFC emissions also result from this source.

[BEGIN BOX]

Box 4-1: Industrial Processes Data from EPA's Greenhouse Gas Reporting Program

On October 30, 2009, the U.S. EPA published a rule for the mandatory reporting of greenhouse gases from large GHG emissions sources in the United States. Implementation of 40 CFR Part 98 is referred to as EPA's Greenhouse Gas Reporting Program (GHGRP). 40 CFR part 98 applies to direct greenhouse gas emitters, fossil fuel suppliers, industrial gas suppliers, and facilities that inject CO₂ underground for sequestration or other reasons and requires reporting by 41 industrial categories. Reporting is at the facility level, except for certain suppliers of fossil fuels and industrial greenhouse gases. In general, the threshold for reporting is 25,000 metric tons or more of CO₂ Eq. per year. For calendar year 2010, the first year in which data were reported, facilities in 29 categories provided in 40 CFR part 98 were required to report their 2010 emissions by the September 30, 2011 reporting deadline.

EPA's GHGRP dataset and the data presented in this inventory report are complementary and, as indicated in the respective planned improvements sections for source categories in this chapter, EPA is analyzing how to use facility-level GHGRP data to improve the national estimates presented in this inventory. Most methodologies used in EPA's GHGRP are consistent with IPCC, though for EPA's GHGRP, facilities collect detailed information specific to their operations according to detailed measurement standards. This may differ with the more aggregated data collected for the inventory to estimate total, national U.S. emissions. In addition, it should be noted that the definitions and provisions for reporting fuel types in EPA's GHGRP may differ from those used in the national inventory in meeting the UNFCCC reporting guidelines. In line with the UNFCCC reporting guidelines¹¹¹, the inventory report is a comprehensive accounting of all emissions from fuel types identified in the IPCC guidelines and provides a separate reporting of emissions from biomass. Further information on the reporting categorizations in EPA's GHGRP and specific data caveats associated with monitoring methods in EPA's GHGRP has been provided on the EPA's GHGRP website.¹¹²

EPA presents the data collected by EPA's GHGRP through a data publication tool¹¹³ that allows data to be viewed in several formats including maps, tables, charts and graphs for individual facilities or groups of facilities.

[END BOX]

4.1. Cement Production (IPCC Source Category 2A1)

Cement production is an energy- and raw-material-intensive process that results in the generation of CO₂ from both the energy consumed in making the cement and the chemical process itself.¹¹⁴ CO₂ emitted from the chemical process of cement production is the second largest source of industrial CO₂ emissions in the United States. Cement is produced in 36 states and Puerto Rico. Texas, California, Missouri, Pennsylvania, Alabama, and Michigan were the six largest (in descending order) cement-producing states in 2011 and accounted for approximately half of U.S. production (USGS 2011).

During the cement production process, calcium carbonate (CaCO₃) is heated in a cement kiln at a temperature of about 1,450°C (2,400°F) to form lime (i.e., calcium oxide or CaO) and CO₂ in a process known as calcination or

¹¹¹ See <http://unfccc.int/resource/docs/2006/sbsta/eng/09.pdf>.

¹¹² See

<<http://www.ccdsupport.com/confluence/display/ghgp/Detailed+Description+of+Data+for+Certain+Sources+and+Processes>>.

¹¹³ See <<http://ghgdata.epa.gov>>.

¹¹⁴ The CO₂ emissions related to the consumption of energy for cement manufacture are accounted for under CO₂ from Fossil Fuel Combustion in the Energy chapter.

calcining. Next, the lime is combined with silica-containing materials to produce clinker (an intermediate product), with the earlier byproduct CO₂ being released to the atmosphere. The clinker is then allowed to cool, mixed with a small amount of gypsum and potentially other materials (e.g., slag), and used to make portland cement.¹¹⁵

In 2010, U.S. clinker production—including Puerto Rico—totaled 59,000 thousand metric tons (USGS 2011). The resulting CO₂ emissions were estimated to be 30.5 Tg CO₂ Eq. (30,509 Gg) (see Table 4-3).

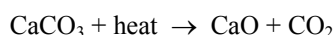
Table 4-3: CO₂ Emissions from Cement Production (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	33.3	33,278
2005	45.2	45,197
2006	45.8	45,792
2007	44.5	44,538
2008	40.5	40,531
2009	29.0	29,018
2010	30.5	30,509

Greenhouse gas emissions from cement production grew every year from 1991 through 2006, but have decreased since. Emissions since 1990 have decreased by eight percent. Emissions decreased significantly between 2008 and 2009, due to the economic recession and associated decrease in demand for construction materials. Although emissions increased slightly from 2009 levels in 2010, they remain 25 percent below 2008 levels, again due to the ongoing contraction of the housing market. Cement continues to be a critical component of the construction industry; therefore, the availability of public and private construction funding, as well as overall economic conditions, have considerable influence on cement production.

Methodology

CO₂ emissions from cement production are created by the chemical reaction of carbon-containing minerals (i.e., calcining limestone) in the cement kiln. While in the kiln, limestone is broken down into CO₂ and lime, with the CO₂ released to the atmosphere. The quantity of CO₂ emitted during cement production is directly proportional to the lime content of the clinker. During calcination, each mole of CaCO₃ (i.e., limestone) heated in the clinker kiln forms one mole of lime (CaO) and one mole of CO₂:



CO₂ emissions were estimated by applying an emission factor, in tons of CO₂ released per ton of clinker produced, to the total amount of clinker produced. The emission factor used in this analysis is the product of the average lime fraction for clinker of 65 percent and a constant reflecting the mass of CO₂ released per unit of lime (van Oss 2008). This calculation yields an emission factor of 0.51 tons of CO₂ per ton of clinker produced, which was determined as follows:

$$EF_{\text{Clinker}} = 0.6460 \text{ CaO} \times \left[\frac{44.01 \text{ g/mole CO}_2}{56.08 \text{ g/mole CaO}} \right] = 0.5070 \text{ tons CO}_2/\text{ton clinker}$$

During clinker production, some of the clinker precursor materials remain in the kiln as non-calcinated, partially calcinated, or fully calcinated cement kiln dust (CKD). The emissions attributable to the calcinated portion of the CKD are not accounted for by the clinker emission factor. The IPCC recommends that these additional CKD CO₂

¹¹⁵ Approximately three percent of total clinker production is used to produce masonry cement, which is produced using plasticizers (e.g., ground limestone, lime) and portland cement (USGS 2011). Carbon dioxide emissions that result from the production of lime used to create masonry cement are included in the Lime Manufacture source category.

emissions should be estimated as two percent of the CO₂ emissions calculated from clinker production (when data on CKD generation are not available).¹¹⁶ Total cement production emissions were calculated by adding the emissions from clinker production to the emissions assigned to CKD (IPCC 2006).

Furthermore, small amounts of impurities (i.e., not calcium carbonate) may exist in the raw limestone used to produce clinker. The proportion of these impurities is generally minimal, although a small (one to two percent) amount of magnesium oxide (MgO) may be desirable as a flux. Per the IPCC Tier 2 methodology, a correction for magnesium oxide is not used, since the amount of magnesium oxide from carbonate is likely very small and the assumption of a 100 percent carbonate source of CaO already yields an overestimation of emissions (IPCC 2006). The 1990 through 2010 activity data for clinker production (see Table 4-4) were obtained from USGS (US Bureau of Mines 1990 through 1993, USGS 1995 through 2011). The data were compiled by USGS through questionnaires sent to domestic clinker and cement manufacturing plants.

Table 4-4: Clinker Production (Gg)

Year	Clinker
1990	64,355
2005	87,405
2006	88,555
2007	86,130
2008	78,382
2009	56,116
2010	59,000

Uncertainty and Time-Series Consistency

The uncertainties contained in these estimates are primarily due to uncertainties in the lime content of clinker and in the percentage of CKD recycled inside the cement kiln. Uncertainty is also associated with the assumption that all calcium-containing raw materials are CaCO₃, when a small percentage likely consists of other carbonate and non-carbonate raw materials. The lime content of clinker varies from 60 to 67 percent; 65 percent is used as a representative value (van Oss 2008). CKD loss can range from 1.5 to 8 percent depending upon plant specifications. Additionally, some amount of CO₂ is reabsorbed when the cement is used for construction. As cement reacts with water, alkaline substances such as calcium hydroxide are formed. During this curing process, these compounds may react with CO₂ in the atmosphere to create calcium carbonate. This reaction only occurs in roughly the outer 0.2 inches of surface area. Because the amount of CO₂ reabsorbed is thought to be minimal, it was not estimated.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-5. Based on the uncertainties associated with total U.S. clinker production, the CO₂ emission factor for clinker production, and the emission factor for additional CO₂ emissions from CKD, 2010 CO₂ emissions from cement production were estimated to be between 26.5 and 34.7 Tg CO₂ Eq. at the 95 percent confidence level. This confidence level indicates a range of approximately 13 percent below and 14 percent above the emission estimate of 30.5 Tg CO₂ Eq.

Table 4-5: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Cement Production (Tg CO₂ Eq. and Percent)

¹¹⁶ Default IPCC clinker and CKD emission factors were verified through expert consultation with the Portland Cement Association (PCA 2008) and van Oss (2008).

Source	Gas	2010 Emission Estimate	Uncertainty Range Relative to Emission Estimate ^a			
		(Tg CO ₂ Eq.)	(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Cement Production	CO ₂	30.5	26.5	34.7	-13%	+14%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Activity data for the time series was revised for the current Inventory. Specifically, clinker production data for 2006 through 2009 were revised to reflect updated USGS data. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Cement Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹¹⁷

4.2. Lime Production (IPCC Source Category 2A2)

Lime is an important manufactured product with many industrial, chemical, and environmental applications. Its major uses are in steel making, flue gas desulfurization systems at coal-fired electric power plants, construction, and water purification. Lime is also used as a CO₂ scrubber, and there has been experimentation on the use of lime to capture CO₂ from electric power plants. For U.S. operations, the term "lime" actually refers to a variety of chemical compounds. These include calcium oxide (CaO), or high-calcium quicklime; calcium hydroxide (Ca(OH)₂), or hydrated lime; dolomitic quicklime ([CaO•MgO]); and dolomitic hydrate ([Ca(OH)₂•MgO] or [Ca(OH)₂•Mg(OH)₂]).

Lime production involves three main processes: stone preparation, calcination, and hydration. Carbon dioxide is generated during the calcination stage, when limestone—mostly calcium carbonate (CaCO₃)—is roasted at high temperatures in a kiln to produce CaO and CO₂. The CO₂ is given off as a gas and is normally emitted to the atmosphere. Some of the CO₂ generated during the production process, however, is recovered at some facilities for use in sugar refining and precipitated calcium carbonate (PCC) production.¹¹⁸

Lime production in the United States—including Puerto Rico—was reported to be 18,259 thousand metric tons in 2010 (USGS 2011). This production resulted in estimated CO₂ emissions of 13.2 Tg CO₂ Eq. (13,151 Gg) (see Table 4-6 and Table 4-7).

Table 4-6: CO₂ Emissions from Lime Production (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	11.5	11,533

¹¹⁷ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

¹¹⁸ PCC is obtained from the reaction of CO₂ with calcium hydroxide. It is used as a filler and/or coating in the paper, food, and plastic industries.

2005	14.4	14,379
2006	15.1	15,100
2007	14.6	14,595
2008	14.3	14,330
2009	11.2	11,225
2010	13.2	13,151

Table 4-7: Potential, Recovered, and Net CO₂ Emissions from Lime Production (Gg)

Year	Potential	Recovered*	Net Emissions
1990	12,004	471	11,533
2005	15,131	752	14,379
2006	15,825	725	15,100
2007	15,264	669	14,595
2008	14,977	647	14,330
2009	11,913	688	11,225
2010	13,795	644	13,151

* For sugar refining and PCC production.

Note: Totals may not sum due to rounding

Lime production in 2010 rebounded from a 21 percent decline in 2009 to 18,259 thousand metric tons, which is still eight percent below 2008 levels. Lime production declined in 2009 mostly due to the economic recession and the associated significant downturn in major markets such as construction and steel. The surprising rebound in 2010 is primarily due to increased consumption in steelmaking, chemical and industrial uses, and in flue gas desulfurization. The contemporary lime market is approximately distributed across five end-use categories as follows: metallurgical uses, 35 percent; environmental uses, 32 percent; chemical and industrial uses, 23 percent; construction uses, nine percent; and refractory dolomite, one percent. Consumption for metallurgical uses, which accounted for 57 percent of the overall decrease in lime consumption in 2009, recorded the most significant (62 percent) gains of 2010 (USGS 2011).

Methodology

During the calcination stage of lime production, CO₂ is given off as a gas and normally exits the system with the stack gas. To calculate emissions, the amounts of high-calcium and dolomitic lime produced were multiplied by their respective emission factors using the Tier 2 approach from the 2006 IPCC Guidelines (IPCC 2006). The emission factor is the product of a constant reflecting the mass of CO₂ released per unit of lime and the average calcium plus magnesium oxide (CaO + MgO) content for lime (95 percent for both types of lime) (IPCC 2006). The emission factors were calculated as follows:

For high-calcium lime:

$$[(44.01 \text{ g/mole CO}_2) \div (56.08 \text{ g/mole CaO})] \times (0.9500 \text{ CaO/lime}) = 0.7455 \text{ g CO}_2/\text{g lime}$$

For dolomitic lime:

$$[(88.02 \text{ g/mole CO}_2) \div (96.39 \text{ g/mole CaO})] \times (0.9500 \text{ CaO/lime}) = 0.8675 \text{ g CO}_2/\text{g lime}$$

Production was adjusted to remove the mass of chemically combined water found in hydrated lime, determined according to the molecular weight ratios of H₂O to (Ca(OH)₂ and [Ca(OH)₂•Mg(OH)₂]) (IPCC 2000). These factors set the chemically combined water content to 24.3 percent for high-calcium hydrated lime, and 27.2 percent for dolomitic hydrated lime.

Lime emission estimates were multiplied by a factor of 1.02 to account for lime kiln dust (LKD), which is produced as a byproduct during the production of lime (IPCC 2006).

Lime emission estimates were further adjusted to account for PCC producers and sugar refineries that recover CO₂ emitted by lime production facilities for use as an input into production or refining processes. For CO₂ recovery by sugar refineries, lime consumption estimates from USGS were multiplied by a CO₂ recovery factor to determine the total amount of CO₂ recovered from lime production facilities. According to industry outreach by state agencies, sugar refineries use captured CO₂ for 100 percent of their CO₂ input (Lutter 2009). Carbon dioxide recovery by PCC producers was determined by multiplying estimates for the percentage CO₂ of production weight for PCC production at lime plants by a CO₂ recovery factor based on the amount of purchased CO₂ by PCC manufacturers (Prillaman 2008 through 2010). As data were only available starting in 2007, CO₂ recovery for the period 1990 through 2006 was extrapolated by determining a ratio of PCC production at lime facilities to lime consumption for PCC (USGS 1992 through 2008).

Lime production data (high-calcium- and dolomitic-quicklime, high-calcium- and dolomitic-hydrated, and dead-burned dolomite) for 1990 through 2010 (see Table 4-8) were obtained from USGS (1992 through 2011). Natural hydraulic lime, which is produced from CaO and hydraulic calcium silicates, is not produced in the United States (USGS 2010). Total lime production was adjusted to account for the water content of hydrated lime by converting hydrate to oxide equivalent based on recommendations from the IPCC, and is presented in Table 4-9 (IPCC 2000). The CaO and CaO•MgO contents of lime were obtained from the IPCC (IPCC 2006). Since data for the individual lime types (high calcium and dolomitic) was not provided prior to 1997, total lime production for 1990 through 1996 was calculated according to the three year distribution from 1997 to 1999.

Table 4-8: High-Calcium- and Dolomitic-Quicklime, High-Calcium- and Dolomitic-Hydrated, and Dead-Burned-Dolomite Lime Production (Gg)

Year	High-Calcium Quicklime	Dolomitic Quicklime	High-Calcium Hydrated	Dolomitic Hydrated	Dead-Burned Dolomite
1990	11,166	2,234	1,781	319	342
2005	14,100	2,990	2,220	474	200
2006	15,000	2,950	2,370	409	200
2007	14,700	2,700	2,240	352	200
2008	14,900	2,310	2,070	358	200
2009	11,800	1,830	1,690	261	200
2010	13,800	2,110	1,910	239	200

Table 4-9: Adjusted Lime Production^a (Gg)

Year	High-Calcium	Dolomitic
1990	12,514	2,809
2005	15,781	3,535
2006	16,794	3,448
2007	16,396	3,156
2008	16,467	2,771
2009	13,079	2,220
2010	15,246	2,484

^a Minus water content of hydrated lime

Uncertainty and Time-Series Consistency

The uncertainties contained in these estimates can be attributed to slight differences in the chemical composition of these products and recovery rates for sugar refineries and PCC manufacturers located at lime plants. Although the methodology accounts for various formulations of lime, it does not account for the trace impurities found in lime,

such as iron oxide, alumina, and silica. Due to differences in the limestone used as a raw material, a rigid specification of lime material is impossible. As a result, few plants produce lime with exactly the same properties.

In addition, a portion of the CO₂ emitted during lime production will actually be reabsorbed when the lime is consumed. As noted above, lime has many different chemical, industrial, environmental, and construction applications. In many processes, CO₂ reacts with the lime to create calcium carbonate (e.g., water softening). Carbon dioxide reabsorption rates vary, however, depending on the application. For example, 100 percent of the lime used to produce precipitated calcium carbonate reacts with CO₂; whereas most of the lime used in steel making reacts with impurities such as silica, sulfur, and aluminum compounds. Quantifying the amount of CO₂ that is reabsorbed would require a detailed accounting of lime use in the United States and additional information about the associated processes where both the lime and byproduct CO₂ are “reused” are required to quantify the amount of CO₂ that is reabsorbed. Research conducted thus far has not yielded the necessary information to quantify CO₂ reabsorption rates.¹¹⁹

In some cases, lime is generated from calcium carbonate byproducts at pulp mills and water treatment plants.¹²⁰ The lime generated by these processes is not included in the USGS data for commercial lime consumption. In the pulping industry, mostly using the Kraft (sulfate) pulping process, lime is consumed in order to causticize a process liquor (green liquor) composed of sodium carbonate and sodium sulfide. The green liquor results from the dilution of the smelt created by combustion of the black liquor where biogenic C is present from the wood. Kraft mills recover the calcium carbonate “mud” after the causticizing operation and calcine it back into lime—thereby generating CO₂—for reuse in the pulping process. Although this re-generation of lime could be considered a lime manufacturing process, the CO₂ emitted during this process is mostly biogenic in origin, and therefore is not included in the industrial processes totals (Miner and Upton 2002). In accordance with IPCC methodological guidelines, any such emissions are calculated by accounting for net carbon (C) fluxes from changes in biogenic C reservoirs in wooded or crop lands (see Chapter 7).

In the case of water treatment plants, lime is used in the softening process. Some large water treatment plants may recover their waste calcium carbonate and calcine it into quicklime for reuse in the softening process. Further research is necessary to determine the degree to which lime recycling is practiced by water treatment plants in the United States.

Uncertainties also remain surrounding recovery rates used for sugar refining and PCC production. The recovery rate for sugar refineries is based on two sugar beet processing and refining facilities located in California that use 100 percent recovered CO₂ from lime plants (Lutter 2010). This analysis assumes that all sugar refineries located on-site at lime plants also use 100 percent recovered CO₂. The recovery rate for PCC producers located on-site at lime plants is based on the 2009 value for PCC manufactured at commercial lime plants, given by the National Lime Association (Prillaman 2010).

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-10. Lime CO₂ emissions were estimated to be between 12.1 and 14.4 Tg CO₂ Eq. at the 95 percent confidence level. This confidence level indicates a range of approximately 8 percent below and 9 percent above the emission estimate of 13.2 Tg CO₂ Eq.

¹¹⁹ Representatives of the National Lime Association estimate that CO₂ reabsorption that occurs from the use of lime may offset as much as a quarter of the CO₂ emissions from calcination (Males 2003).

¹²⁰ Some carbide producers may also regenerate lime from their calcium hydroxide byproducts, which does not result in emissions of CO₂. In making calcium carbide, quicklime is mixed with coke and heated in electric furnaces. The regeneration of lime in this process is done using a waste calcium hydroxide (hydrated lime) [$\text{CaC}_2 + 2\text{H}_2\text{O} \rightarrow \text{C}_2\text{H}_2 + \text{Ca}(\text{OH})_2$], not calcium carbonate [CaCO_3]. Thus, the calcium hydroxide is heated in the kiln to simply expel the water [$\text{Ca}(\text{OH})_2 + \text{heat} \rightarrow \text{CaO} + \text{H}_2\text{O}$] and no CO₂ is released.

Table 4-10: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Lime Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate	Uncertainty Range Relative to Emission Estimate ^a			
		(Tg CO ₂ Eq.)	(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Lime Production	CO ₂	13.2	12.1	14.4	-8%	+9%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Data on lime consumption for sugar refining in 2009 was revised by USGS from 733 to 731 metric tons. This revision resulted in an increase of emissions from 11,223 to 11,225 Gg CO₂ Eq., an increase of 0.02 percent.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Lime Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹²¹

Future improvements to the lime source category will also involve continued research into CO₂ recovery associated with lime use during sugar refining and precipitate calcium carbonate (PCC) production. Currently, two sugar refining facilities in California have been identified to capture CO₂ produced in lime kilns located on the same site as the sugar refinery (Lutter 2010). Data on CO₂ production by these lime facilities is unavailable. Future work will include research to determine the number of sugar refineries that employ the carbonation technique, the percentage of these that use captured CO₂ from lime production facilities, and the amount of CO₂ recovered per unit of lime production. Future research will also aim to improve estimates of CO₂ recovered as part of the PCC production process using estimates of PCC production and CO₂ inputs rather than lime consumption by PCC facilities.

4.3. Limestone and Dolomite Use (IPCC Source Category 2A3)

Limestone (CaCO₃) and dolomite (CaCO₃MgCO₃)¹²² are basic raw materials used by a wide variety of industries, including construction, agriculture, chemical, metallurgy, glass production, and environmental pollution control. Limestone is widely distributed throughout the world in deposits of varying sizes and degrees of purity. Large deposits of limestone occur in nearly every state in the United States, and significant quantities are extracted for industrial applications. For some of these applications, limestone is heated sufficiently enough to calcine the material and generate CO₂ as a byproduct. Examples of such applications include limestone used as a flux or purifier in metallurgical furnaces, as a sorbent in flue gas desulfurization (FGD) systems for utility and industrial plants, and as a raw material for the production of glass, lime, and cement.

In 2010, 21,004 thousand metric tons of limestone and 2,624 thousand metric tons of dolomite were consumed for these emissive applications (USGS 2011a). Usage of limestone and dolomite resulted in aggregate CO₂ emissions of 10.0 Tg CO₂ Eq. (10,017 Gg) (see Table 4-11 and Table 4-12). Overall, emissions have increased 95 percent from 1990 through 2010.

¹²¹ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

¹²² Limestone and dolomite are collectively referred to as limestone by the industry, and intermediate varieties are seldom distinguished.

Table 4-11: CO₂ Emissions from Limestone & Dolomite Use (Tg CO₂ Eq.)

Year	Flux Stone	Glass Making	FGD	Magnesium Production	Other Miscellaneous Uses	Total
1990	2.6	0.2	1.4	0.1	0.8	5.1
2005	2.7	0.4	3.0	+	0.7	6.8
2006	4.5	0.7	2.1	+	0.7	8.0
2007	2.0	0.3	3.2	+	2.2	7.7
2008	1.0	0.4	3.8	+	1.1	6.3
2009	1.8	0.1	5.4	+	0.4	7.6
2010	1.6	0.4	7.1	+	0.9	10.0

Notes: Totals may not sum due to independent rounding. "Other miscellaneous uses" include chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining.

+ Emissions are less than 0.1 Tg CO₂ Eq.

Table 4-12: CO₂ Emissions from Limestone & Dolomite Use (Gg)

Year	Flux Stone	Glass Making	FGD	Magnesium Production	Other Miscellaneous Uses	Total
1990	2,593	217	1,433	64	819	5,127
2005	2,650	425	2,975	+	718	6,768
2006	4,492	747	2,061	+	735	8,035
2007	1,959	333	3,179	+	2,231	7,702
2008	974	387	3,801	+	1,114	6,276
2009	1,785	61	5,406	+	396	7,649
2010	1,572	440	7,068	+	938	10,017

+ Emissions are less than 0.1 Tg CO₂ Eq.

Methodology

CO₂ emissions were calculated based on the IPCC 2006 Guidelines Tier 2 method by multiplying the quantity of limestone or dolomite consumed by the average C content, 12.0 percent for limestone and 13.0 percent for dolomite (based on stoichiometry), and converting this value to CO₂. This methodology was used for flux stone, glass manufacturing, flue gas desulfurization systems, chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining and then converting to CO₂ using a molecular weight ratio. Flux stone used during the production of iron and steel was deducted from the Limestone and Dolomite Use estimate and attributed to the Iron and Steel Production estimate.

Traditionally, the production of magnesium metal was the only other significant use of limestone and dolomite that produced CO₂ emissions. At the start of 2001, there were two magnesium production plants operating in the United States and they used different production methods. One plant produced magnesium metal using a dolomitic process that resulted in the release of CO₂ emissions, while the other plant produced magnesium from magnesium chloride using a CO₂-emissions-free process called electrolytic reduction. However, the plant utilizing the dolomitic process ceased its operations prior to the end of 2001, so beginning in 2002 there were no emissions from this particular sub-use (USGS 2011b).

Consumption data for 1990 through 2010 of limestone and dolomite used for flux stone, glass manufacturing, flue gas desulfurization systems, chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining (see Table 4-13) were obtained from the USGS *Minerals Yearbook: Crushed Stone Annual Report* (1995 through 2011a) and the U.S. Bureau of Mines (1991 and 1993a). The production capacity data for 1990 through 2010 of dolomitic magnesium metal also came from the USGS (1995 through 2011b) and the U.S. Bureau of Mines (1990 through 1993b). During 1990 and 1992, the USGS did not conduct a detailed survey of limestone and dolomite consumption by end-use. Consumption for 1990 was estimated by applying the 1991 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to 1990 total use. Similarly, the 1992 consumption figures were approximated by applying an average of the 1991 and 1993 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to the 1992 total.

Additionally, each year the USGS withholds data on certain limestone and dolomite end-uses due to confidentiality agreements regarding company proprietary data. For the purposes of this analysis, emissive end-uses that contained withheld data were estimated using one of the following techniques: (1) the value for all the withheld data points for limestone or dolomite use was distributed evenly to all withheld end-uses; (2) the average percent of total limestone or dolomite for the withheld end-use in the preceding and succeeding years; or (3) the average fraction of total limestone or dolomite for the end-use over the entire time period.

There is a large quantity of crushed stone reported to the USGS under the category “unspecified uses.” A portion of this consumption is believed to be limestone or dolomite used for emissive end uses. The quantity listed for “unspecified uses” was, therefore, allocated to each reported end use according to each end uses fraction of total consumption in that year.¹²³

Table 4-13: Limestone and Dolomite Consumption (Thousand Metric Tons)

Activity	1990	2005	2006	2007	2008	2009	2010
Flux Stone	6,737	7,022	11,030	5,305	3,253	4,623	4,441
Limestone	5,804	3,165	5,208	3,477	1,970	1,631	1,921
Dolomite	933	3,857	5,822	1,827	1,283	2,992	2,520
Glass Making	489	962	1,693	757	879	139	1,000
Limestone	430	920	1,629	757	879	139	1,000
Dolomite	59	43	64	+	+	+	+
FGD	3,258	6,761	4,683	7,225	8,639	12,288	16,064
Other Miscellaneous Uses	1,835	1,632	1,671	5,057	2,531	898	2,122
Total	12,319	16,377	19,078	18,344	15,302	17,948	23,628

+ Emissions are less than 0.1 Tg CO₂ Eq.

Notes: "Other miscellaneous uses" includes chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining. Zero values for limestone and dolomite consumption for glass making result during years when the USGS reports that no limestone or dolomite are consumed for this use.

Uncertainty and Time-Series Consistency

The uncertainty levels presented in this section arise in part due to variations in the chemical composition of limestone. In addition to calcium carbonate, limestone may contain smaller amounts of magnesia, silica, and sulfur, among other minerals. The exact specifications for limestone or dolomite used as flux stone vary with the pyrometallurgical process and the kind of ore processed. Similarly, the quality of the limestone used for glass manufacturing will depend on the type of glass being manufactured.

The estimates below also account for uncertainty associated with activity data. Large fluctuations in reported consumption exist, reflecting year-to-year changes in the number of survey responders. The uncertainty resulting from a shifting survey population is exacerbated by the gaps in the time series of reports. The accuracy of distribution by end use is also uncertain because this value is reported by the manufacturer and not the end user. Additionally, there is significant inherent uncertainty associated with estimating withheld data points for specific end uses of limestone and dolomite. The uncertainty of the estimates for limestone used in glass making is especially high; however, since glass making accounts for a small percent of consumption, its contribution to the overall emissions estimate is low. Lastly, much of the limestone consumed in the United States is reported as “other unspecified uses.” Therefore, it is difficult to accurately allocate this unspecified quantity to the correct end-uses.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-14. Limestone and Dolomite Use CO₂ emissions were estimated to be between 8.7 and 11.8 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 13 percent below and 18 percent above the emission estimate of 10.0 Tg CO₂ Eq.

Table 4-14: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Limestone and Dolomite Use (Tg CO₂ Eq. and Percent)

¹²³This approach was recommended by USGS.

Source	Gas	2010 Emission Estimate	Uncertainty Range Relative to Emission Estimate ^a			
		(Tg CO ₂ Eq.)	(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Limestone and Dolomite Use	CO ₂	10.0	8.7	11.8	-13%	+18%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Limestone and Dolomite Use source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹²⁴ Additionally, future improvements include revisiting the methodology to distribute withheld data across emissive end-uses for all years to improve consistency of calculations.

4.4. Soda Ash Production and Consumption (IPCC Source Category 2A4)

Soda ash (sodium carbonate, Na₂CO₃) is a white crystalline solid that is readily soluble in water and strongly alkaline. Commercial soda ash is used as a raw material in a variety of industrial processes and in many familiar consumer products such as glass, soap and detergents, paper, textiles, and food. It is used primarily as an alkali, either in glass manufacturing or simply as a material that reacts with and neutralizes acids or acidic substances. Internationally, two types of soda ash are produced, natural and synthetic. The United States produces only natural soda ash and is second only to China in total soda ash production. Trona is the principal ore from which natural soda ash is made.

Only two states produce natural soda ash: Wyoming and California. Of these two states, only net emissions of CO₂ from Wyoming were calculated due to specifics regarding the production processes employed in the state.¹²⁵ During the production process used in Wyoming, trona ore is calcined to produce crude soda ash. Carbon dioxide is generated as a byproduct of this reaction, and is eventually emitted into the atmosphere. In addition, CO₂ may also be released when soda ash is consumed.

In 2010, CO₂ emissions from the production of soda ash from trona were approximately 1.5 Tg CO₂ Eq. (1,548 Gg). Soda ash consumption in the United States generated 2.2 Tg CO₂ Eq. (2,187 Gg) in 2010. Total emissions from soda ash production and consumption in 2010 were 3.7 Tg CO₂ Eq. (3,735 Gg) (see Table 4-15 and Table 4-16). Total emissions in 2010 increased by approximately 5 percent from emissions in 2009, and have decreased overall by approximately 9.8 percent since 1990.

¹²⁴ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

¹²⁵ In California, soda ash is manufactured using sodium carbonate-bearing brines instead of trona ore. To extract the sodium carbonate, the complex brines are first treated with CO₂ in carbonation towers to convert the sodium carbonate into sodium bicarbonate, which then precipitates from the brine solution. The precipitated sodium bicarbonate is then calcined back into sodium carbonate. Although CO₂ is generated as a byproduct, the CO₂ is recovered and recycled for use in the carbonation stage and is not emitted. A third state, Colorado, produced soda ash until the plant was idled in 2004. The lone producer of sodium bicarbonate no longer mines trona in the state. For a brief time, NaHCO₃ was produced using soda ash feedstocks mined in Wyoming and shipped to Colorado. Because the trona is mined in Wyoming, the production numbers given by the USGS included the feedstocks mined in Wyoming and shipped to Colorado. In this way, the sodium bicarbonate production that took place in Colorado was accounted for in the Wyoming numbers.

Emissions have remained relatively constant over the time series with some fluctuations since 1990. In general, these fluctuations were related to the behavior of the export market and the U.S. economy. Specifically, the extended downturn in residential and commercial construction and automotive industries between 2008 and 2010 resulted in reduced consumption of glass products, causing a drop in global demand for soda ash and a corresponding decrease in emissions. Furthermore, the glass container sector is one of the leading soda ash consuming sectors in the United States. Some commercial food and beverage package manufacturers are shifting from glass containers towards lighter and more cost effective polyethylene terephthalate (PET) based containers, putting downward pressure on domestic consumption of soda ash (USGS 2010 and 2011).

Table 4-15: CO₂ Emissions from Soda Ash Production and Consumption (Tg CO₂ Eq.)

Year	Production	Consumption	Total
1990	1.4	2.7	4.1
2005	1.7	2.6	4.2
2006	1.6	2.5	4.2
2007	1.7	2.5	4.1
2008	1.7	2.4	4.1
2009	1.5	2.1	3.6
2010	1.5	2.2	3.7

Note: Totals may not sum due to independent rounding.

Table 4-16: CO₂ Emissions from Soda Ash Production and Consumption (Gg)

Year	Production	Consumption	Total
1990	1,431	2,710	4,141
2005	1,655	2,573	4,228
2006	1,626	2,536	4,162
2007	1,675	2,465	4,140
2008	1,733	2,366	4,099
2009	1,470	2,083	3,554
2010	1,548	2,187	3,735

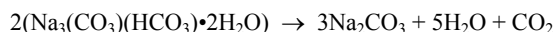
Note: Totals may not sum due to independent rounding.

The United States represents about one-fourth of total world soda ash output. Based on final 2010 reported data, the estimated distribution of soda ash by end-use in 2010 was glass making, 48 percent; chemical production, 29 percent; soap and detergent manufacturing, 10 percent; distributors, 5 percent; flue gas desulfurization, 4 percent; other uses and pulp and paper production, 2 percent each; and water treatment, less than 1 percent (USGS 2011).

Although the United States continues to be a major supplier of world soda ash, China, which surpassed the United States in soda ash production in 2003, is the world's leading producer. While Chinese soda ash production appears to be stabilizing, U.S. competition in Asian markets is expected to continue. Despite this competition, U.S. soda ash production is expected to increase by about 0.5 percent annually (USGS 2008).

Methodology

During the production process, trona ore is calcined in a rotary kiln and chemically transformed into a crude soda ash that requires further processing. Carbon dioxide and water are generated as byproducts of the calcination process. Carbon dioxide emissions from the calcination of trona can be estimated based on the following chemical reaction:



[trona]

[soda ash]

Based on this formula, which is consistent with an IPCC Tier 1 approach, approximately 10.27 metric tons of trona are required to generate one metric ton of CO₂, or an emission factor of 0.097 metric tons CO₂ per metric ton trona (IPCC 2006). Thus, the 15.9 million metric tons of trona mined in 2010 for soda ash production (USGS 2011) resulted in CO₂ emissions of approximately 1.5 Tg CO₂ Eq. (1,548 Gg).

Once produced, most soda ash is consumed in glass and chemical production, with minor amounts in soap and detergents, pulp and paper, flue gas desulfurization and water treatment. As soda ash is consumed for these purposes, additional CO₂ is usually emitted. In these applications, it is assumed that one mole of C is released for every mole of soda ash used. Thus, approximately 0.113 metric tons of C (or 0.415 metric tons of CO₂) are released for every metric ton of soda ash consumed.

The activity data for trona production and soda ash consumption (see Table 4-17) between 1990 and 2010 were taken from USGS Minerals Yearbook for Soda Ash (1994 through 2011). Soda ash production and consumption data were collected by the USGS from voluntary surveys of the U.S. soda ash industry.

Table 4-17: Soda Ash Production and Consumption (Gg)

Year	Production*	Consumption
1990	14,700	6,530
2005	17,000	6,200
2006	16,700	6,110
2007	17,200	5,940
2008	17,800	5,700
2009	15,100	5,020
2010	15,900	5,270

* Soda ash produced from trona ore only.

Uncertainty and Time-Series Consistency

Emission estimates from soda ash production have relatively low associated uncertainty levels in that reliable and accurate data sources are available for the emission factor and activity data. The primary source of uncertainty, however, results from the fact that emissions from soda ash consumption are dependent upon the type of processing employed by each end-use. Specific emission factors for each end-use are not available, so a Tier 1 default emission factor is used for all end uses. Therefore, there is uncertainty surrounding the emission factors from the consumption of soda ash.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-18. Soda Ash Production and Consumption CO₂ emissions were estimated to be between 3.5 and 4.0 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 7 percent below and 7 percent above the emission estimate of 3.7 Tg CO₂ Eq.

Table 4-18: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Soda Ash Production and Consumption (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)		Uncertainty Range Relative to Emission Estimate ^a (Tg CO ₂ Eq.)		(%)	
		Lower Bound	Upper Bound	Lower Bound	Upper Bound	Lower Bound	Upper Bound
Soda Ash Production and Consumption	CO ₂	3.7	3.5	4.0	-7%	+7%	

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations

Trona production data was updated for 2009 and soda ash consumption data was updated for 2008 and 2009 based on newly available data from the USGS Minerals Yearbook Soda Ash 2010 (USGS 2011). This resulted in a decrease of total emissions from soda ash production and consumption for 2008 and 2009 by approximately 0.3 percent and 17 percent, respectively.

Planned Improvements

Future inventories are anticipated to estimate emissions from glass production and other use of carbonates. These inventories will extract soda ash consumed for glass production and other use of carbonates from the current soda ash consumption emission estimates and include them under those sources.

In examining data from EPA's GHGRP that would be useful to improve the emission estimates for Soda Ash and

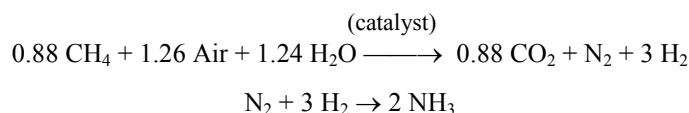
Consumption category, particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all inventory years as reported in this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹²⁶

4.5. Ammonia Production (IPCC Source Category 2B1)

Emissions of CO₂ occur during the production of synthetic ammonia, primarily through the use of natural gas, petroleum coke, or naphtha as a feedstock. The natural gas-based, naphtha-based, and petroleum coke-based processes produce CO₂ and hydrogen (H₂), the latter of which is used in the production of ammonia. One synthetic ammonia production plant located in Kansas is producing ammonia from petroleum coke feedstock; other synthetic ammonia production plants in the U.S. are using natural gas feedstock. In some plants some of the CO₂ produced by the process is captured and used to produce urea rather than being emitted to the atmosphere. The brine electrolysis process for production of ammonia does not lead to process-based CO₂ emissions.

There are five principal process steps in synthetic ammonia production from natural gas feedstock. The primary reforming step converts CH₄ to CO₂, carbon monoxide (CO), and H₂ in the presence of a catalyst. Only 30 to 40 percent of the CH₄ feedstock to the primary reformer is converted to CO and CO₂ in this step of the process. The secondary reforming step converts the remaining CH₄ feedstock to CO and CO₂. The CO in the process gas from the secondary reforming step (representing approximately 15 percent of the process gas) is converted to CO₂ in the presence of a catalyst, water, and air in the shift conversion step. Carbon dioxide is removed from the process gas by the shift conversion process, and the hydrogen gas is combined with the nitrogen (N₂) gas in the process gas during the ammonia synthesis step to produce ammonia. The CO₂ is included in a waste gas stream with other process impurities and is absorbed by a scrubber solution. In regenerating the scrubber solution, CO₂ is released from the solution.

The conversion process for conventional steam reforming of CH₄, including the primary and secondary reforming and the shift conversion processes, is approximately as follows:



To produce synthetic ammonia from petroleum coke, the petroleum coke is gasified and converted to CO₂ and H₂. These gases are separated, and the H₂ is used as a feedstock to the ammonia production process, where it is reacted with N₂ to form ammonia.

Not all of the CO₂ produced during the production of ammonia is emitted directly to the atmosphere. Some of the ammonia and some of the CO₂ produced by the synthetic ammonia process are used as raw materials in the production of urea [CO(NH₂)₂], which has a variety of agricultural and industrial applications.

The chemical reaction that produces urea is:



Only the CO₂ emitted directly to the atmosphere from the synthetic ammonia production process are accounted for in determining emissions from ammonia production. The CO₂ that is captured during the ammonia production process and used to produce urea does not contribute to the CO₂ emission estimates for ammonia production presented in this section. Instead, CO₂ emissions resulting from the consumption of urea are attributed to the urea consumption or urea application category (under the assumption that the C stored in the urea during its manufacture is released into the environment during its consumption or application). Emissions of CO₂ resulting from agricultural applications of urea are accounted for in the Cropland Remaining Cropland section of the Land-use, Land-use Change, and Forestry chapter. Emissions of CO₂ resulting from non-agricultural applications of urea (e.g., use as a feedstock in chemical production processes) are accounted for in the Urea Consumption for Non-Agricultural Purposes section of the Industrial Process chapter.

Total emissions of CO₂ from ammonia production in 2010 were 8.7 Tg CO₂ Eq. (8,678 Gg), and are summarized in

¹²⁶ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

Table 4-19 and Table 4-20. The observed decrease in ammonia production and associated CO₂ emissions between 2007 and 2009 is due to several factors, including market fluctuations and high natural gas prices. Ammonia production relies on natural gas as both a feedstock and a fuel, and as such, domestic producers are competing with imports from countries with lower natural gas prices (EEA 2004). The 2010 increase in ammonia production (and associated CO₂ emissions) is largely attributable to dramatically lower natural gas prices in the U.S. after 2009 (EIA 2011).

Table 4-19: CO₂ Emissions from Ammonia Production (Tg CO₂ Eq.)

Source	1990		2005	2006	2007	2008	2009	2010
Ammonia Production	13.0		9.2	8.8	9.1	7.9	7.9	8.7
Total	13.0		9.2	8.8	9.1	7.9	7.9	8.7

Table 4-20: CO₂ Emissions from Ammonia Production (Gg)

Source	1990		2005	2006	2007	2008	2009	2010
Ammonia Production	13,047		9,196	8,781	9,074	7,883	7,855	8,678
Total	13,047		9,196	8,781	9,074	7,883	7,855	8,678

Methodology

The calculation methodology for non-combustion CO₂ emissions from production of synthetic ammonia from natural gas feedstock is based on the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006). The method utilizes a CO₂ emission factor published by the European Fertilizer Manufacturers Association (EFMA) that is based on natural gas-based ammonia production technologies that are similar to those employed in the United States. The CO₂ emission factor (1.2 metric tons CO₂/metric ton NH₃) is applied to the percent of total annual domestic ammonia production from natural gas feedstock. Emissions from fuels consumed for energy purposes during the production of ammonia are accounted for in the Energy chapter.

Emissions of CO₂ from ammonia production are then adjusted to account for the use of some of the CO₂ produced from ammonia production as a raw material in the production of urea. The CO₂ emissions reported for ammonia production are reduced by a factor of 0.733 multiplied by total annual domestic urea production. This corresponds to a stoichiometric CO₂/urea factor of 44/60, assuming complete conversion of NH₃ and CO₂ to urea (IPCC 2006, EFMA 2000).

All synthetic ammonia production and subsequent urea production are assumed to be from the same process—conventional catalytic reforming of natural gas feedstock, with the exception of ammonia production from petroleum coke feedstock at one plant located in Kansas. The CO₂ emission factor for production of ammonia from petroleum coke is based on plant specific data, wherein all C contained in the petroleum coke feedstock that is not used for urea production is assumed to be emitted to the atmosphere as CO₂ (Bark 2004). Ammonia and urea are assumed to be manufactured in the same manufacturing complex, as both the raw materials needed for urea production are produced by the ammonia production process. The CO₂ emission factor for the petroleum coke feedstock process (3.57 metric tons CO₂/metric ton NH₃) is applied to the percent of total annual domestic ammonia production from petroleum coke feedstock.

The emission factor of 1.2 metric ton CO₂/metric ton NH₃ for production of ammonia from natural gas feedstock was taken from the EFMA Best Available Techniques publication, *Production of Ammonia* (EFMA 1995). The EFMA reported an emission factor range of 1.15 to 1.30 metric ton CO₂/metric ton NH₃, with 1.2 metric ton CO₂/metric ton NH₃ as a typical value. Technologies (e.g., catalytic reforming process) associated with this factor are found to closely resemble those employed in the U.S. for use of natural gas as a feedstock. The EFMA reference also indicates that more than 99 percent of the CH₄ feedstock to the catalytic reforming process is ultimately converted to CO₂. The emission factor of 3.57 metric ton CO₂/metric ton NH₃ for production of ammonia from

petroleum coke feedstock was developed from plant-specific ammonia production data and petroleum coke feedstock utilization data for the ammonia plant located in Kansas (Bark 2004). As noted earlier, emissions from fuels consumed for energy purposes during the production of ammonia are accounted for in the Energy chapter. Ammonia production data (see Table 4-21) was obtained from Coffeyville Resources (Coffeyville 2005, 2006, 2007a, 2007b, 2009, 2010, and 2011) and the Census Bureau of the U.S. Department of Commerce (U.S. Census Bureau 1991 through 1994, 1998 through 2011) as reported in Current Industrial Reports Fertilizer Materials and Related Products annual and quarterly reports. Urea-ammonia nitrate production was obtained from Coffeyville Resources (Coffeyville 2005, 2006, 2007a, 2007b, 2009, 2010, 2011). Urea production data for 1990 through 2008 were obtained from the Minerals Yearbook: Nitrogen (USGS 1994 through 2009). Urea production data for 2009 through 2010 were obtained from the U.S. Bureau of the Census (2011).

Table 4-21: Ammonia Production and Urea Production (Gg)

Year	Ammonia Production	Urea Production
1990	15,425	7,450
2005	10,143	5,270
2006	9,962	5,410
2007	10,393	5,590
2008	9,570	5,240
2009	9,372	5,084
2010	10,084	5,122

Uncertainty and Time-Series Consistency

The uncertainties presented in this section are primarily due to how accurately the emission factor used represents an average across all ammonia plants using natural gas feedstock. Uncertainties are also associated with natural gas feedstock consumption data for the U.S. ammonia industry as a whole, the assumption that all ammonia production and subsequent urea production was from the same process—conventional catalytic reforming of natural gas feedstock, with the exception of one ammonia production plant located in Kansas that is manufacturing ammonia from petroleum coke feedstock. Uncertainty is also associated with the representativeness of the emission factor used for the petroleum coke-based ammonia process. It is also assumed that ammonia and urea are produced at collocated plants from the same natural gas raw material.

Recovery of CO₂ from ammonia production plants for purposes other than urea production (e.g., commercial sale) has not been considered in estimating the CO₂ emissions from ammonia production, as data concerning the disposition of recovered CO₂ are not available. Such recovery may or may not affect the overall estimate of CO₂ emissions depending upon the end use to which the recovered CO₂ is applied. Further research is required to determine whether byproduct CO₂ is being recovered from other ammonia production plants for application to end uses that are not accounted for elsewhere.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-22. Ammonia Production CO₂ emissions were estimated to be between 7.8 and 10.9 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 10 percent below and 25 percent above the emission estimate of 8.7 Tg CO₂ Eq.

Table 4-22: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Ammonia Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate		Uncertainty Range Relative to Emission Estimate ^a		
		(Tg CO ₂ Eq.)		(Tg CO ₂ Eq.)	(%)	
		Lower Bound	Upper Bound	Lower Bound	Upper Bound	
Ammonia Production	CO ₂	8.7	7.8	10.9	-10%	+25%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

For the current Inventory, emissions resulting from non-agricultural urea consumption have been transferred from the Ammonia Production section to a new section within the Industrial Process chapter titled Urea Consumption for Non-Agricultural Purposes. From 1990 to 2009, urea consumption for non-agricultural purposes accounted for an average of 27 percent of the combined emissions from ammonia production and non-agricultural urea consumption each year.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Ammonia Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹²⁷ Specifically, the planned improvements include assessing data to update the emission factors to include both fuel and feedstock CO₂ emissions and incorporate CO₂ capture and storage. Methodologies will also be updated if additional ammonia-production plants are found to use hydrocarbons other than natural gas for ammonia production.

4.6. Urea Consumption for Non-Agricultural Purposes

Urea is used as a nitrogenous fertilizer for agricultural applications and also in a variety of industrial applications. Urea's industrial applications include its use as adhesives, binders, sealants, resins, fillers, analytical reagents, catalysts, intermediates, solvents, dyestuffs, fragrances, deodorizers, flavoring agents, humectants and dehydrating agents, formulation components, monomers, paint and coating additives, photosensitive agents, and surface treatments agents. In addition, urea is used for abating nitrous oxide emissions from coal-fired power plants and diesel transportation motors.

Urea is produced using ammonia and CO₂ as raw materials. All urea produced in the U.S. is assumed to be produced at ammonia production facilities where both ammonia and CO₂ are generated. The chemical reaction that produces urea is:



This section accounts for CO₂ emissions associated with urea consumed exclusively for non-agricultural purposes. CO₂ emissions associated with urea consumed for fertilizer are accounted for in the Cropland Remaining Cropland section of the Land Use, Land-Use Change, and Forestry chapter.

Emissions of CO₂ from urea consumed for non-agricultural purposes in 2010 were estimated to be 4.4 Tg CO₂ Eq. (4,365 Gg), and are summarized in Table 4-23 and Table 4-24.

¹²⁷ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

Table 4-23: CO₂ Emissions from Urea Consumption for Non-Agricultural Purposes (Tg CO₂ Eq.)

Source	1990		2005	2006	2007	2008	2009	2010
Urea Consumption	3.8		3.7	3.5	4.9	4.1	3.4	4.4
Total	3.8		3.7	3.5	4.9	4.1	3.4	4.4

Table 4-24: CO₂ Emissions from Urea Consumption for Non-Agricultural Purposes (Gg)

Source	1990		2005	2006	2007	2008	2009	2010
Urea Consumption	3,784		3,653	3,519	4,944	4,065	3,415	4,365
Total	3,784		3,653	3,519	4,944	4,065	3,415	4,365

Methodology

Emissions of CO₂ resulting from urea consumption for non-agricultural purposes are estimated by multiplying the amount of urea consumed in the U.S. for non-agricultural purposes by a factor representing the amount of CO₂ used as a raw material to produce the urea. This method is based on the assumption that all of the C in urea is released into the environment as CO₂ during use.

The amount of urea consumed for non-agricultural purposes in the U.S. is estimated by deducting the quantity of urea fertilizer applied to agricultural lands, which is obtained directly from the Land Use, Land-Use Change, and Forestry chapter and is reported in Table 4-25, from the total domestic supply of urea. The domestic supply of urea is estimated based on the amount of urea produced plus the sum of net urea imports and exports. A factor of 0.73 tons of CO₂ per ton of urea consumed is then applied the resulting supply of urea for non-agricultural purposes to estimate CO₂ emissions from the amount of urea consumed for non-agricultural purposes. The 0.733 tons of CO₂ per ton of urea emission factor is based on the stoichiometry of producing urea from ammonia and CO₂. This corresponds to a stoichiometric CO₂/urea factor of 44/60, assuming complete conversion of NH₃ and CO₂ to urea (IPCC 2006, EFMA 2000).

Urea production data for 1990 through 2008 were obtained from the Minerals Yearbook: Nitrogen (USGS 1994 through 2009). Urea production data for 2009 through 2010 were obtained from the U.S. Bureau of the Census (2011). Import data for urea were obtained from the U.S. Census Bureau Current Industrial Reports Fertilizer Materials and Related Products annual and quarterly reports for 1997 through 2010 (U.S. Census Bureau 1998 through 2011), The Fertilizer Institute (TFI 2002) for 1993 through 1996, and the United States International Trade Commission Interactive Tariff and Trade DataWeb (U.S. ITC 2002) for 1990 through 1992 (see Table 4-25). Urea export data for 1990 through 2010 were taken from U.S. Fertilizer Import/Exports from USDA Economic Research Service Data Sets (U.S. Department of Agriculture 2011).

Table 4-25: Urea Production, Urea Applied as Fertilizer, Urea Imports, and Urea Exports (Gg)

Year	Urea Production	Urea Applied as Fertilizer	Urea Imports	Urea Exports
1990	7,450	3,296	1,860	854
2005	5,270	4,779	5,026	536
2006	5,410	4,985	5,029	656
2007	5,590	5,097	6,546	271
2008	5,240	4,925	5,459	230
2009	5,084	4,925	4,727	289
2010	5,122	4,925	6,631	152

Uncertainty and Time-Series Consistency

The amount of urea used for non-agricultural purposes is estimated based on estimates of urea production, urea imports, urea exports, and the amount of urea used as fertilizer. The primary uncertainties associated with this source category are associated with the accuracy of these estimates as well as the fact that each estimate is obtained from a different data source. There is also uncertainty associated with the assumption that all of the C in urea is released into the environment as CO₂ during use.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-26. CO₂ emissions associated with urea consumption for non-agricultural purposes were estimated to be between 2.3 and 5.0 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 47 percent below and 15 percent above the emission estimate of 4.4 Tg CO₂ Eq.

Table 4-26: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Urea Consumption for Non-Agricultural Purposes (Tg CO₂ Eq. and Percent)

Source	2010 Emission Estimate		Uncertainty Range Relative to Emission Estimate ^a			
	Gas	(Tg CO ₂ Eq.)	(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Urea Consumption for Non-Agricultural Purposes	CO ₂	4.4	2.3	5.0	-47%	+15%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements to the urea consumption for non-agricultural purposes source category involve continuing to research obtaining data on how much urea is consumed for specific applications in the United States and whether C is released to the environment fully during each application.

4.7. Nitric Acid Production (IPCC Source Category 2B2)

Nitric acid (HNO₃) is an inorganic compound used primarily to make synthetic commercial fertilizers. It is also a major component in the production of adipic acid—a feedstock for nylon—and explosives. Virtually all of the nitric acid produced in the United States is manufactured by the catalytic oxidation of ammonia (EPA 1997). During this reaction, N₂O is formed as a byproduct and is released from reactor vents into the atmosphere.

Currently, the nitric acid industry controls for emissions of NO and NO₂ (i.e., NO_x). As such, the industry in the US uses a combination of non-selective catalytic reduction (NSCR) and selective catalytic reduction (SCR) technologies. In the process of destroying NO_x, NSCR systems are also very effective at destroying N₂O. However, NSCR units are generally not preferred in modern plants because of high energy costs and associated high gas temperatures. NSCRs were widely installed in nitric plants built between 1971 and 1977. As of 2010, approximately 32 percent of nitric acid plants use NSCR, representing 17.3 percent of estimated national production (EPA 2010). The remaining 82.7 percent of production occurs using SCR or extended absorption, neither of which is known to reduce N₂O emissions.

N₂O emissions from this source were estimated to be 16.7 Tg CO₂ Eq. (54 Gg) in 2010 (see Table 4-27). Emissions from nitric acid production have decreased by 5.1 percent since 1990, with the trend in the time series closely tracking the changes in production. Emissions increased 15.3 percent between 2009 and 2010. Emissions have decreased by 20 percent since 1997, the highest year of production in the time series.

Table 4-27: N₂O Emissions from Nitric Acid Production (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	17.6	57
2005	16.4	53
2006	16.1	52
2007	19.2	62
2008	16.4	53
2009	14.5	47
2010	16.7	54

Methodology

N₂O emissions were calculated by multiplying nitric acid production by the amount of N₂O emitted per unit of nitric acid produced. The emission factor was determined as a weighted average of two known emission factors: 2 kg N₂O/metric ton HNO₃ produced at plants using non-selective catalytic reduction (NSCR) systems and 9 kg N₂O/metric ton HNO₃ produced at plants not equipped with NSCR (IPCC 2006). In the process of destroying NO_x, NSCR systems destroy 80 to 90 percent of the N₂O, which is accounted for in the emission factor of 2 kg N₂O/metric ton HNO₃.

In 2009, several nitric acid production facilities that did not have NSCR abatement systems installed were closed (Desai 2012). As a result, as of 2010 approximately 32 percent of HNO₃ plants in the United States are equipped with NSCR representing 17.3 percent of estimated national production (EPA 2010).

Hence, the emission factor used for 2010 is equal to the production-weighted emission factor based on the production at facilities with and without NSCR and the associated emission factors, equating to:

$(2 \text{ kg N}_2\text{O/metric ton HNO}_3 \text{ produced} \times 0.173) + (9 \text{ kg N}_2\text{O/metric ton HNO}_3 \text{ produced} \times 0.827) = 7.8 \text{ kg N}_2\text{O per metric ton HNO}_3.$

The emission factor used for years prior to the plant closures (i.e., 1990-2009) is equal to:

$(2 \text{ kg N}_2\text{O/metric ton HNO}_3 \text{ produced} \times 0.153) + (9 \text{ kg N}_2\text{O/metric ton HNO}_3 \text{ produced} \times 0.847) = 7.9 \text{ kg N}_2\text{O per metric ton HNO}_3.$

Nitric acid production data for the U.S. for 1990 through 2002 were obtained from the U.S. Census Bureau (2010b); 2003 production data were obtained from the U.S. Census Bureau (2008); 2004 through 2007 production data were obtained from the U.S. Census Bureau (2009); 2008 and 2009 production data were obtained from the U.S. Census Bureau (2010a); and 2010 production data were obtained from the U.S. Census Bureau (2011) (see Table 4-28).

Table 4-28: Nitric Acid Production (Gg)

Year	Gg
1990	7,195
2005	6,711
2006	6,572
2007	7,827
2008	6,686
2009	5,924
2010	6,931

Uncertainty and Time-Series Consistency

Uncertainty associated with the parameters used to estimate N₂O emissions includes that of production data, the share of U.S. nitric acid production attributable to each emission abatement technology over the time series, and the emission factors applied to each abatement technology type. While some information has been obtained through outreach with industry associations, limited information is readily available over the time series for a variety of facility level variables, including plant specific production levels, abatement technology type and installation date and accurate destruction and removal efficiency rates. Some information will be available through EPA's reporting program, but this data is not available over the time series.

The results of this Tier 2 quantitative uncertainty analysis are summarized in Table 4-29. N₂O emissions from nitric acid production were estimated to be between 10.1 and 23.9 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 39 percent below to 40 percent above the 2010 emissions estimate of 16.7 Tg CO₂ Eq.

Table 4-29: Tier 2 Quantitative Uncertainty Estimates for N₂O Emissions from Nitric Acid Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO₂ Eq.)	Uncertainty Range Relative to Emission Estimate^a			
			(Tg CO₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Nitric Acid Production	N ₂ O	16.7	10.1	23.9	-39%	+40%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements to the Nitric Acid Production category involve research into the availability of facility level nitric acid production data, abatement technology type and installation dates, more accurate destruction and removal efficiency percentages, the current and past share of nitric acid production attributable to various abatement technologies, as well as efforts to analyze data reported under EPA's GHGRP. These research efforts are especially important given the cancellation of the U.S. Census Bureau's Current Industrial Reports data series, from which national Nitric Acid production data are derived. Furthermore, in examining data from EPA's GHGRP that would be useful to improve the emission estimates for nitric acid production category, particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all inventory years as reported in this inventory. In implementing improvements and integration of data from EPA's

GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹²⁸

4.8. Adipic Acid Production (IPCC Source Category 2B3)

Adipic acid production is an anthropogenic source of N₂O emissions. Worldwide, few adipic acid plants exist. The United States and Europe are the major producers. In 2010, the United States had two companies with a total of three adipic acid production facilities, two of which were operational (CW 2007; Desai 2010; VA DEQ 2009). The United States accounts for the largest share of global adipic acid production capacity (30 percent), followed by the European Union (29 percent) and China (22 percent) (SEI 2010). Adipic acid is a white crystalline solid used in the manufacture of synthetic fibers, plastics, coatings, urethane foams, elastomers, and synthetic lubricants. Commercially, it is the most important of the aliphatic dicarboxylic acids, which are used to manufacture polyesters. 84 percent of all adipic acid produced in the United States is used in the production of nylon 6,6; nine percent is used in the production of polyester polyols; four percent is used in the production of plasticizers; and the remaining four percent is accounted for by other uses, including unsaturated polyester resins and food applications (ICIS 2007). Food grade adipic acid is used to provide some foods with a “tangy” flavor (Thiemens and Trogler 1991).

Adipic acid is produced through a two-stage process during which N₂O is generated in the second stage. The first stage of manufacturing usually involves the oxidation of cyclohexane to form a cyclohexanone/cyclohexanol mixture. The second stage involves oxidizing this mixture with nitric acid to produce adipic acid. N₂O is generated as a byproduct of the nitric acid oxidation stage and is emitted in the waste gas stream (Thiemens and Trogler 1991). Process emissions from the production of adipic acid vary with the types of technologies and level of emission controls employed by a facility. In 1990, two of the three major adipic acid-producing plants had N₂O abatement technologies in place and, as of 1998, the three major adipic acid production facilities had control systems in place (Reimer et al. 1999). One small plant, which last operated in April 2006 and represented approximately two percent of production, did not control for N₂O (VA DEQ 2009; ICIS 2007; VA DEQ 2006).

Very little information on annual trends in the activity data exist for adipic acid. Primary production data is derived from the American Chemistry Council (ACC) *Guide to the Business of Chemistry*, which does not provide source specific trend information. The USGS does not currently publish a Minerals Yearbook for adipic acid, and it is not included in the general USGS Minerals Commodity Summary.

N₂O emissions from adipic acid production were estimated to be 2.8 Tg CO₂ Eq. (9.1 Gg) in 2010 (see Table 4-30). National adipic acid production has increased by approximately 4 percent over the period of 1990 through 2010, to roughly 760,000 metric tons. Over the same period, emissions have been reduced by 82 percent due to both the widespread installation of pollution control measures in the late 1990s and plant idling in the late 2000s. In April 2006, the smallest of the four facilities ceased production of adipic acid (VA DEQ 2009); furthermore, one of the major adipic acid production facilities was not operational in 2009 or 2010 (Desai 2010). Due to a lack of data, 2010 emissions from adipic acid production were assumed to be equal to 2009 estimates.

Table 4-30: N₂O Emissions from Adipic Acid Production (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	15.8	51
2005	7.4	24
2006	8.9	28.7
2007	10.7	34.4
2008	2.6	8.3
2009	2.8	9.1
2010	2.8	9.1

¹²⁸ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

Methodology

Due to confidential business information, plant names are not provided in this section. The four adipic acid-producing plants will henceforth be referred to as Plants 1 through 4.

For Plants 1 and 2, 1990 to 2009 emission estimates were obtained directly from the plant engineer and account for reductions due to control systems in place at these plants during the time series (Desai 2010). These estimates were based on continuous process monitoring equipment installed at the two facilities. In 2009 and 2010, no adipic acid production occurred at Plant 1. For Plant 4, N₂O emissions were calculated by multiplying adipic acid production by an emission factor (i.e., N₂O emitted per unit of adipic acid produced) and adjusting for the percentage of N₂O released as a result of plant-specific emission controls. On the basis of experiments, the overall reaction stoichiometry for N₂O production in the preparation of adipic acid was estimated at approximately 0.3 metric tons of N₂O per metric ton of product (IPCC 2006). Emissions for plants lacking emissions monitoring data were estimated using the following equation:

$$\text{N}_2\text{O emissions} = (\text{production of adipic acid [metric tons \{MT\} of adipic acid]}) \times (0.3 \text{ MT N}_2\text{O} / \text{MT adipic acid}) \times (1 - [\text{N}_2\text{O destruction factor} \times \text{abatement system utility factor}])$$

The “N₂O destruction factor” represents the percentage of N₂O emissions that are destroyed by the installed abatement technology. The “abatement system utility factor” represents the percentage of time that the abatement equipment operates during the annual production period. Overall, in the United States, two of the plants employ catalytic destruction (Plants 1 and 2), one plant employs thermal destruction (Plant 3), and the smallest plant that closed in 2006 used no N₂O abatement equipment (Plant 4).

For Plant 3, 2005 through 2009 emissions were obtained directly from the plant engineer (Desai 2012). For 1990 through 2004, emissions were estimated using plant-specific production data and IPCC factors as described above for Plant 4. Production data for 1990 through 2003 was estimated by allocating national adipic acid production data to the plant level using the ratio of known plant capacity to total national capacity for all U.S. plants. For 2004, actual plant production data were obtained and used for emission calculations (CW 2005).

Plant capacities for 1990 through 1994 were obtained from Chemical and Engineering News, “Facts and Figures” and “Production of Top 50 Chemicals” (C&EN 1992 through 1995). Plant capacities for 1995 and 1996 were kept the same as 1994 data. The 1997 plant capacities were taken from Chemical Market Reporter “Chemical Profile: Adipic Acid” (CMR 1998). The 1998 plant capacities for all four plants and 1999 plant capacities for three of the plants were obtained from Chemical Week, Product Focus: Adipic Acid/Adiponitrile (CW 1999). Plant capacities for 2000 for three of the plants were updated using Chemical Market Reporter, “Chemical Profile: Adipic Acid” (CMR 2001). For 2001 through 2003, the plant capacities for three plants were kept the same as the year 2000 capacities. Plant capacity for 1999 to 2003 for the one remaining plant was kept the same as 1998. For Plant 4, which last operated in April 2006 (VA DEQ 2009), plant-specific production data were obtained across the time series from 1990 through 2008 (VA DEQ 2010). Since the plant has not operated since 2006, production through 2010 was assumed to be zero. The plant-specific production data were then used for calculating emissions as described above.

National adipic acid production data (see Table 4-31) from 1990 through 2010 were obtained from the American Chemistry Council (ACC 2011).

Table 4-31: Adipic Acid Production (Gg)

Year	Gg
1990	735
2005	903
2006	964
2007	930
2008	869
2009	819
2010	764

Uncertainty and Time-Series Consistency

Uncertainty associated with N₂O emission estimates included that of the methods used by companies to monitor and estimate emissions and the use of 2009 emissions data as a proxy for 2010.

The results of this Tier 2 quantitative uncertainty analysis are summarized in Table 4-32. N₂O emissions from adipic acid production for 2010 were estimated to be between 2.6 and 3.1 Tg CO₂ Eq. at the 95 percent confidence level. These values indicate a range of approximately 9 percent below to 9 percent above the 2010 emission estimate of 2.8 Tg CO₂ Eq.

Table 4-32: Tier 2 Quantitative Uncertainty Estimates for N₂O Emissions from Adipic Acid Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate		Uncertainty Range Relative to Emission Estimate ^a		
		(Tg CO ₂ Eq.)		(Tg CO ₂ Eq.) (%)		
		Lower Bound	Upper Bound	Lower Bound	Upper Bound	
Adipic Acid Production	N ₂ O	2.8	2.6	3.1	-9%	+9%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Adipic Acid Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹²⁹ Specifically, the planned improvements include assessing data to update the N₂O emission factors and update abatement utility and destruction factors based on actual performance of the latest catalytic and thermal abatement equipment at plants with continuous process and emission monitoring equipment.

Recalculations

For the current Inventory, plant specific N₂O emissions data for Plant 3 were obtained directly from the plant engineer for 2005 through 2009. In the previous Inventory, 2005 through 2009 estimates of N₂O emissions from adipic acid production at Plant 3 were developed using plant production data. For the 1990 through 2009 inventory, Plant 3 emissions for, which uses thermal destruction, the N₂O abatement system destruction factor was assumed to be 98.5 percent, and the abatement system utility factor was assumed to be 97 percent (IPCC 2006). This recalculation resulted in an 84 percent increase in average annual estimated N₂O emissions from adipic acid production between 2005 and 2009, relative to the previous report.

4.9. Silicon Carbide Production (IPCC Source Category 2B4) and Consumption

Carbon dioxide and CH₄ are emitted from the production¹³⁰ of silicon carbide (SiC), a material used as an industrial abrasive. To make SiC, quartz (SiO₂) is reacted with C in the form of petroleum coke. A portion (about 35 percent)

¹²⁹ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

¹³⁰ Silicon carbide is produced for both abrasive and metallurgical applications in the United States. Production for metallurgical applications is not available and therefore both CH₄ and CO₂ estimates are based solely upon production estimates of silicon carbide for abrasive applications.

of the C contained in the petroleum coke is retained in the SiC. The remaining C is emitted as CO₂, CH₄, or CO.

Carbon dioxide is also emitted from the consumption of SiC for metallurgical and other non-abrasive applications. The USGS reports that a portion (approximately 50 percent) of SiC is used in metallurgical and other non-abrasive applications, primarily in iron and steel production (USGS 2006a). Markets for manufactured abrasives, including SiC, are heavily influenced by activity in the U.S. manufacturing sector, especially in the aerospace, automotive, furniture, housing, and steel manufacturing sectors. As a result of the economic downturn in 2008 and 2009, demand for SiC decreased in those years. Low cost imports, particularly from China, combined with high relative operating costs for domestic producers, continue to put downward pressure on the production of SiC in the United States. However, demand for SiC consumption in the United States has recovered somewhat from its lows in 2009 (USGS 2011a).

Carbon dioxide emissions from SiC production and consumption in 2010 were 0.18 Tg CO₂ Eq. (181 Gg). Approximately 51 percent of these emissions resulted from SiC production while the remainder resulted from SiC consumption. Methane emissions from SiC production in 2010 were 0.01 Tg CO₂ Eq. CH₄ (0.4 Gg) (see Table 4-33 and Table 4-34).

Table 4-33: CO₂ and CH₄ Emissions from Silicon Carbide Production and Consumption (Tg CO₂ Eq.)

Year	1990		2005	2006	2007	2008	2009	2010
CO ₂	0.4		0.2	0.2	0.2	0.2	0.1	0.2
CH ₄	+		+	+	+	+	+	+
Total	0.4		0.2	0.2	0.2	0.2	0.2	0.2

+ Does not exceed 0.05 Tg CO₂ Eq.

Note: Totals may not sum due to independent rounding.

Table 4-34: CO₂ and CH₄ Emissions from Silicon Carbide Production and Consumption (Gg)

Year	1990		2005	2006	2007	2008	2009	2010
CO ₂	375		219	207	196	175	145	181
CH ₄	1		+	+	+	+	+	+

+ Does not exceed 0.5 Gg.

Methodology

Emissions of CO₂ and CH₄ from the production of SiC were calculated by multiplying annual SiC production by the emission factors (2.62 metric tons CO₂/metric ton SiC for CO₂ and 11.6 kg CH₄/metric ton SiC for CH₄) provided by the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC 2006).

Emissions of CO₂ from silicon carbide consumption for metallurgical uses were calculated by multiplying the annual utilization of SiC for metallurgical uses (reported annually in the USGS Minerals Yearbook for Silicon) by the C content of SiC (31.5 percent), which was determined according to the molecular weight ratio of SiC.

Emissions of CO₂ from silicon carbide consumption for other non-abrasive uses were calculated by multiplying the remaining annual SiC consumption (total SiC consumption minus SiC utilization for metallurgical uses) by the emissive utilization percentage for SiC utilization for other non-abrasive uses (50 percent) (USGS 2009).

Production data for 1990 through 2009 were obtained from the Minerals Yearbook: Manufactured Abrasives (USGS 1991a through 2010a and 2011b). Production data for 2010 was taken from the Minerals Commodity Summary: Abrasives (Manufactured) (2011a). Silicon carbide consumption by major end use was obtained from the Minerals Yearbook: Silicon (USGS 1991b through 2010b and 2011c) (see Table 4-35) for years 1990 through 2009. Silicon carbide consumption for 2010 is proxied using 2009 data due to unavailability of data at time of publication. Net imports for the entire time series were obtained from the U.S. Census Bureau (2005 through 2011).

Table 4-35: Production and Consumption of Silicon Carbide (Metric Tons)

Year	Production	Consumption
1990	105,000	172,465
2005	35,000	220,149
2006	35,000	199,937
2007	35,000	179,741
2008	35,000	144,928
2009	35,000	92,280
2010	35,000	154,540

Uncertainty and Time-Series Consistency

There is uncertainty associated with the emission factors used because they are based on stoichiometry as opposed to monitoring of actual SiC production plants. An alternative would be to calculate emissions based on the quantity of petroleum coke used during the production process rather than on the amount of silicon carbide produced. However, these data were not available. For CH₄, there is also uncertainty associated with the hydrogen-containing volatile compounds in the petroleum coke (IPCC 2006). There is also uncertainty associated with the use or destruction of methane generated from the process in addition to uncertainty associated with levels of production, net imports, consumption levels, and the percent of total consumption that is attributed to metallurgical and other non-abrasive uses.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-36. Silicon carbide production and consumption CO₂ emissions were estimated to be between 9 percent below and 10 percent above the emission estimate of 0.2 Tg CO₂ Eq. at the 95 percent confidence level. Silicon carbide production CH₄ emissions were estimated to be between 9 percent below and 9 percent above the emission estimate of 0.01 Tg CO₂ Eq. at the 95 percent confidence level.

Table 4-36: Tier 2 Quantitative Uncertainty Estimates for CH₄ and CO₂ Emissions from Silicon Carbide Production and Consumption (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate		Uncertainty Range Relative to Emission Estimate ^a	
		(Tg CO ₂ Eq.)	(Tg CO ₂ Eq.)	(%)	(%)
		Lower Bound	Upper Bound	Lower Bound	Upper Bound
Silicon Carbide Production and Consumption	CO ₂	0.2	0.2	-9%	+10%
Silicon Carbide Production	CH ₄	+	+	-9%	+9%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

+ Does not exceed 0.05 Tg CO₂ Eq. or 0.5 Gg.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Silicon Carbide Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's

GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹³¹ In addition, improvements will involve continued research to determine if calcium carbide production and consumption data are available for the United States. If these data are available, calcium carbide emission estimates will be included in this source category. Additionally, as future improvement to the silicon carbide uncertainty analysis, USGS Mineral Commodity Specialists will be contacted to verify the uncertainty range associated with silicon carbide emissive utilization.

4.10. Petrochemical Production (IPCC Source Category 2B5)

The production of some petrochemicals results in the release of small amounts of CH₄ and CO₂ emissions. Petrochemicals are chemicals isolated or derived from petroleum or natural gas. Methane emissions are presented here from the production of carbon black, ethylene, ethylene dichloride, and methanol, while CO₂ emissions are presented here for only carbon black production. The CO₂ emissions from petrochemical processes other than carbon black are currently included in the Carbon Stored in Products from Non-Energy Uses of Fossil Fuels Section of the Energy chapter. The CO₂ from carbon black production is included here to allow for the direct reporting of CO₂ emissions from the process and direct accounting of the feedstocks used in the process.

Carbon black is an intense black powder generated by the incomplete combustion of an aromatic petroleum or coal-based feedstock. Most carbon black produced in the United States is added to rubber to impart strength and abrasion resistance, and the tire industry is by far the largest consumer. Ethylene is consumed in the production processes of the plastics industry including polymers such as high, low, and linear low density polyethylene (HDPE, LDPE, LLDPE), polyvinyl chloride (PVC), ethylene dichloride, ethylene oxide, and ethylbenzene. Ethylene dichloride is one of the first manufactured chlorinated hydrocarbons with reported production as early as 1795. In addition to being an important intermediate in the synthesis of chlorinated hydrocarbons, ethylene dichloride is used as an industrial solvent and as a fuel additive. Methanol is an alternative transportation fuel as well as a principle ingredient in windshield wiper fluid, paints, solvents, refrigerants, and disinfectants. In addition, methanol-based acetic acid is used in making PET plastics and polyester fibers.

Emissions of CO₂ and CH₄ from petrochemical production in 2010 were 3.3 Tg CO₂ Eq. (3,336 Gg) and 0.9 Tg CH₄ Eq. (44 Gg), respectively (see Table 4-37 and Table 4-38), totaling 4.3 Tg CO₂ Eq. There has been an overall increase in CO₂ emissions from carbon black production of one percent since 1990. Methane emissions from petrochemical production have increased by approximately seven percent since 1990.

Table 4-37: CO₂ and CH₄ Emissions from Petrochemical Production (Tg CO₂ Eq.)

Year	1990		2005	2006	2007	2008	2009	2010
CO ₂	3.3		4.2	3.8	3.9	3.4	2.7	3.3
CH ₄	0.9		1.1	1.0	1.0	0.9	0.8	0.9
Total	4.2		5.3	4.8	4.9	4.3	3.6	4.3

Note: Totals may not sum due to independent rounding.

Table 4-38: CO₂ and CH₄ Emissions from Petrochemical Production (Gg)

Year	1990		2005	2006	2007	2008	2009	2010
CO ₂	3,311		4,181	3,837	3,931	3,449	2,735	3,336
CH ₄	41		51	48	48	43	39	44

Methodology

Emissions of CH₄ were calculated by multiplying annual estimates of chemical production by the appropriate emission factor, as follows: 11 kg CH₄/metric ton carbon black, 1 kg CH₄/metric ton ethylene, 0.4 kg CH₄/metric ton

¹³¹ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

ethylene dichloride,¹³² and 2 kg CH₄/metric ton methanol. Although the production of other chemicals may also result in CH₄ emissions, insufficient data were available to estimate their emissions.

Emission factors were taken from the Revised 1996 IPCC Guidelines (IPCC/UNEP/OECD/IEA 1997). Annual production data (see Table 4-39) were obtained from the American Chemistry Council's Guide to the Business of Chemistry (ACC 2002, 2003, 2005 through 2011) and the International Carbon Black Association (Johnson 2003 and 2005 through 2011). Methanol production data for 1990 through 2007 were obtained from the ACC Guide to the Business of Chemistry (ACC 2002, 2003, 2005 through 2011). The ACC discontinued its data series for Methanol after 2007, so methanol production data for 2008 through 2010 was obtained through the Methanol Institute (Jordan 2011a and 2011b).

Table 4-39: Production of Selected Petrochemicals (Thousand Metric Tons)

Chemical	1990	2005	2006	2007	2008	2009	2010
Carbon Black	1,307	1,651	1,515	1,552	1,362	1,080	1,317
Ethylene	16,541	23,954	25,000	25,392	22,539	22,596	23,961
Ethylene Dichloride	6,282	11,260	9,736	9,566	8,981	8,131	8,820
Methanol	3,785	2,336	1,123	1,068	810	810	903

Almost all carbon black in the United States is produced from petroleum-based or coal-based feedstocks using the “furnace black” process (European IPPC Bureau 2004). The furnace black process is a partial combustion process in which a portion of the carbon black feedstock is combusted to provide energy to the process. Carbon black is also produced in the United States by the thermal cracking of acetylene-containing feedstocks (“acetylene black process”) and by the thermal cracking of other hydrocarbons (“thermal black process”). One U.S. carbon black plant produces carbon black using the thermal black process, and one U.S. carbon black plant produces carbon black using the acetylene black process (The Innovation Group 2004).

The furnace black process produces carbon black from “carbon black feedstock” (also referred to as “carbon black oil”), which is a heavy aromatic oil that may be derived as a byproduct of either the petroleum refining process or the metallurgical (coal) coke production process. For the production of both petroleum-derived and coal-derived carbon black, the “primary feedstock” (i.e., carbon black feedstock) is injected into a furnace that is heated by a “secondary feedstock” (generally natural gas). Both the natural gas secondary feedstock and a portion of the carbon black feedstock are oxidized to provide heat to the production process and pyrolyze the remaining carbon black feedstock to carbon black. The “tail gas” from the furnace black process contains CO₂, carbon monoxide, sulfur compounds, CH₄, and non-CH₄ volatile organic compounds. A portion of the tail gas is generally burned for energy recovery to heat the downstream carbon black product dryers. The remaining tail gas may also be burned for energy recovery, flared, or vented uncontrolled to the atmosphere.

The calculation of the C lost during the production process is the basis for determining the amount of CO₂ released during the process. The C content of national carbon black production is subtracted from the total amount of C contained in primary and secondary carbon black feedstock to find the amount of C lost during the production process. It is assumed that the C lost in this process is emitted to the atmosphere as either CH₄ or CO₂. The C content of the CH₄ emissions, estimated as described above, is subtracted from the total C lost in the process to calculate the amount of C emitted as CO₂. The total amount of primary and secondary carbon black feedstock consumed in the process (see Table 4-40) is estimated using a primary feedstock consumption factor and a secondary feedstock consumption factor estimated from U.S. Census Bureau (1999, 2004, and 2007) data. The average carbon black feedstock consumption factor for U.S. carbon black production is 1.69 metric tons of carbon black feedstock consumed per metric ton of carbon black produced. The average natural gas consumption factor for U.S. carbon black production is 321 normal cubic meters of natural gas consumed per metric ton of carbon black produced. The amount of C contained in the primary and secondary feedstocks is calculated by applying the respective C contents of the feedstocks to the respective levels of feedstock consumption (EIA 2003, 2004).

¹³² The emission factor obtained from IPCC/UNEP/OECD/IEA (1997), page 2.23 is assumed to have a misprint; the chemical identified should be ethylene dichloride (C₂H₄Cl₂) rather than dichloroethylene (C₂H₂Cl₂).

Table 4-40: Carbon Black Feedstock (Primary Feedstock) and Natural Gas Feedstock (Secondary Feedstock) Consumption (Thousand Metric Tons)

Activity	1990	2005	2006	2007	2008	2009	2010
Primary Feedstock	2,213	2,794	2,564	2,627	2,305	1,828	2,230
Secondary Feedstock	284	359	329	337	296	235	286

For the purposes of emissions estimation, 100 percent of the primary carbon black feedstock is assumed to be derived from petroleum refining byproducts. Carbon black feedstock derived from metallurgical (coal) coke production (e.g., creosote oil) is also used for carbon black production; however, no data are available concerning the annual consumption of coal-derived carbon black feedstock. Carbon black feedstock derived from petroleum refining byproducts is assumed to be 89 percent elemental C (Srivastava et al. 1999). It is assumed that 100 percent of the tail gas produced from the carbon black production process is combusted and that none of the tail gas is vented to the atmosphere uncontrolled. The furnace black process is assumed to be the only process used for the production of carbon black because of the lack of data concerning the relatively small amount of carbon black produced using the acetylene black and thermal black processes. The carbon black produced from the furnace black process is assumed to be 97 percent elemental C (Othmer et al. 1992).

Uncertainty and Time-Series Consistency

The CH₄ emission factors used for petrochemical production are based on a limited number of studies. Using plant-specific factors instead of average factors could increase the accuracy of the emission estimates; however, such data were not available. There may also be other significant sources of CH₄ arising from petrochemical production activities that have not been included in these estimates.

The results of the quantitative uncertainty analysis for the CO₂ emissions from carbon black production calculation are based on feedstock consumption, import and export data, and carbon black production data. The composition of carbon black feedstock varies depending upon the specific refinery production process, and therefore the assumption that carbon black feedstock is 89 percent C gives rise to uncertainty. Also, no data are available concerning the consumption of coal-derived carbon black feedstock, so CO₂ emissions from the utilization of coal-based feedstock are not included in the emission estimate. In addition, other data sources indicate that the amount of petroleum-based feedstock used in carbon black production may be underreported by the U.S. Census Bureau. Finally, the amount of carbon black produced from the thermal black process and acetylene black process, although estimated to be a small percentage of the total production, is not known. Therefore, there is some uncertainty associated with the assumption that all of the carbon black is produced using the furnace black process.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-41. Petrochemical production CO₂ emissions were estimated to be between 2.5 and 4.3 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 26 percent below to 29 percent above the emission estimate of 3.3 Tg CO₂ Eq. Petrochemical production CH₄ emissions were estimated to be between 0.7 and 1.2 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 29 percent below to 30 percent above the emission estimate of 0.9 Tg CO₂ Eq.

Table 4-41: Tier 2 Quantitative Uncertainty Estimates for CH₄ Emissions from Petrochemical Production and CO₂ Emissions from Carbon Black Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^a			
			(Tg CO ₂ Eq.)	(Tg CO ₂ Eq.)	(%)	(%)
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Petrochemical Production	CO ₂	3.3	2.5	4.3	-26%	+29%
Petrochemical Production	CH ₄	0.9	0.7	1.2	-29%	+30%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations

Methanol production data for 2008 and 2009 was updated relative to the previous report based on correspondence with Jim Jordan of Jordan Associates (Jordan 2011a and 2011b). This resulted in a decrease of total CO₂ and CH₄ emissions from petrochemical production of less than 1 percent.

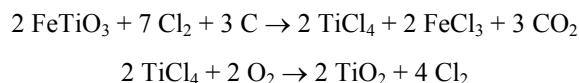
Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Petrochemical Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹³³ Additional future improvements involve updating the methodology to use CH₄ emission factors for petrochemical production from the IPCC 2006 guidelines rather than the IPCC 1996 guidelines and assessing the data EPA obtains to update data sources for acrylonitrile production in the United States.

4.11. Titanium Dioxide Production (IPCC Source Category 2B5)

Titanium dioxide (TiO₂) is a metal oxide manufactured from titanium ore, and is principally used as a pigment. Titanium dioxide is a principal ingredient in white paint, and is also used as a pigment in the manufacture of white paper, foods, and other products. There are two processes for making TiO₂: the chloride process and the sulfate process. The chloride process uses petroleum coke and chlorine as raw materials and emits process-related CO₂. The sulfate process does not use petroleum coke or other forms of C as a raw material and does not emit CO₂.

The chloride process is based on the following chemical reactions:



The C in the first chemical reaction is provided by petroleum coke, which is oxidized in the presence of the chlorine and FeTiO₃ (the Ti-containing ore) to form CO₂. The majority of U.S. TiO₂ was produced in the United States through the chloride process, and a special grade of "calcined" petroleum coke is manufactured specifically for this purpose.

Emissions of CO₂ in 2010 were 1.9 Tg CO₂ Eq. (1,876 Gg), which represents an increase of 57 percent since 1990 (see Table 4-42).

Table 4-42: CO₂ Emissions from Titanium Dioxide (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	1.2	1,195
2005	1.8	1,755
2006	1.8	1,836
2007	1.9	1,930
2008	1.8	1,809
2009	1.6	1,648
2010	1.9	1,876

¹³³ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

Methodology

Emissions of CO₂ from TiO₂ production were calculated by multiplying annual TiO₂ production by chloride-process-specific emission factors.

Data were obtained for the total amount of TiO₂ produced each year. For years previous to 2004, it was assumed that TiO₂ was produced using the chloride process and the sulfate process in the same ratio as the ratio of the total U.S. production capacity for each process. As of 2004, the last remaining sulfate-process plant in the United States had closed; therefore, 100 percent of post-2004 production uses the chloride process (USGS 2005). An emission factor of 0.4 metric tons C/metric ton TiO₂ was applied to the estimated chloride-process production. It was assumed that all TiO₂ produced using the chloride process was produced using petroleum coke, although some TiO₂ may have been produced with graphite or other C inputs. The amount of petroleum coke consumed annually in TiO₂ production was calculated based on the assumption that the calcined petroleum coke used in the process is 98.4 percent C and 1.6 percent inert materials (Nelson 1969).

The emission factor for the TiO₂ chloride process was taken from the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006). Titanium dioxide production data and the percentage of total TiO₂ production capacity that is chloride process for 1990 through 2009 (see Table 4-43) were obtained through the Minerals Yearbook: Titanium Annual Report (USGS 1991 through 2011b). Production data for 2010 was obtained from the Minerals Commodity Summary: Titanium and Titanium Dioxide (USGS 2011a). Due to lack of available 2010 capacity data at the time of publication, the 2009 capacity estimate is used as a proxy for 2010. Percentage chloride-process data were not available for 1990 through 1993, so data from the 1994 USGS Minerals Yearbook were used for these years. Because a sulfate-process plant closed in September 2001, the chloride-process percentage for 2001 was estimated based on a discussion with Joseph Gambogi (2002). By 2002, only one sulfate plant remained online in the United States and this plant closed in 2004 (USGS 2005).

Table 4-43: Titanium Dioxide Production (Gg)

Year	Gg
1990	979
2005	1,310
2006	1,370
2007	1,440
2008	1,350
2009	1,230
2010	1,400

Uncertainty and Time-Series Consistency

Although some TiO₂ may be produced using graphite or other C inputs, information and data regarding these practices were not available. Titanium dioxide produced using graphite inputs, for example, may generate differing amounts of CO₂ per unit of TiO₂ produced as compared to that generated through the use of petroleum coke in production. While the most accurate method to estimate emissions would be to base calculations on the amount of reducing agent used in each process rather than on the amount of TiO₂ produced, sufficient data were not available to do so.

Also, annual TiO₂ is not reported by USGS by the type of production process used (chloride or sulfate). Only the percentage of total production capacity by process is reported. The percent of total TiO₂ production capacity that was attributed to the chloride process was multiplied by total TiO₂ production to estimate the amount of TiO₂ produced using the chloride process (since, as of 2004, the last remaining sulfate-process plant in the United States closed). This assumes that the chloride-process plants and sulfate-process plants operate at the same level of utilization. Finally, the emission factor was applied uniformly to all chloride-process production, and no data were available to account for differences in production efficiency among chloride-process plants. In calculating the amount of petroleum coke consumed in chloride-process TiO₂ production, literature data were used for petroleum coke composition. Certain grades of petroleum coke are manufactured specifically for use in the TiO₂ chloride

process; however, this composition information was not available.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-44. Titanium dioxide consumption CO₂ emissions were estimated to be between 1.6 and 2.1 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 13 percent below and 13 percent above the emission estimate of 1.9 Tg CO₂ Eq.

Table 4-44: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Titanium Dioxide Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)		Uncertainty Range Relative to Emission Estimate ^a (Tg CO ₂ Eq.)		Range Relative to Emission Estimate ^a (%)	
		Lower Bound	Upper Bound	Lower Bound	Upper Bound	Lower Bound	Upper Bound
Titanium Dioxide Production	CO ₂	1.9	1.6	2.1		-13%	+13%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations

Production data for 2009 were updated relative to the previous Inventory based on recently published data in the USGS Minerals Yearbook: Titanium 2009 (USGS 2011). This resulted in a 7 percent decrease in 2009 CO₂ emissions from TiO₂ production relative to the previous report.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Titanium Dioxide source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹³⁴ In addition, the planned improvements include researching the significance of titanium-slag production in electric furnaces and synthetic-rutile production using the Becher process in the United States. Significant use of these production processes will be included in future estimates.

4.12. Carbon Dioxide Consumption (IPCC Source Category 2B5)

CO₂ is used for a variety of commercial applications, including food processing, chemical production, carbonated beverage production, and refrigeration, and is also used in petroleum production for enhanced oil recovery (EOR). Carbon dioxide used for EOR is injected into the underground reservoirs to increase the reservoir pressure to enable additional petroleum to be produced.

For the most part, CO₂ used in non-EOR applications will eventually be released to the atmosphere, and for the purposes of this analysis CO₂ used in commercial applications other than EOR is assumed to be emitted to the atmosphere. Carbon dioxide used in EOR applications is discussed in the Energy Chapter under "Carbon Capture and Storage, including Enhanced Oil Recovery" and is not discussed in this section.

CO₂ is produced from naturally occurring CO₂ reservoirs, as a byproduct from the energy and industrial production processes (e.g., ammonia production, fossil fuel combustion, ethanol production), and as a byproduct from the production of crude oil and natural gas, which contain naturally occurring CO₂ as a component. Only CO₂ produced from naturally occurring CO₂ reservoirs and used in industrial applications other than EOR is included in this

¹³⁴ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

analysis. Neither byproduct CO₂ generated from energy nor industrial production processes nor CO₂ separated from crude oil and natural gas are included in this analysis for a number of reasons. Carbon dioxide captured from biogenic sources (e.g., ethanol production plants) is not included in the inventory. Carbon dioxide captured from crude oil and gas production is used in EOR applications and is therefore reported in the Energy Chapter. Any CO₂ captured from industrial or energy production processes (e.g., ammonia plants, fossil fuel combustion) and used in non-EOR applications is assumed to be emitted to the atmosphere. The CO₂ emissions from such capture and use are therefore accounted for under Ammonia Production, Fossil Fuel Combustion, or other appropriate source category.¹³⁵

CO₂ is produced as a byproduct of crude oil and natural gas production. This CO₂ is separated from the crude oil and natural gas using gas processing equipment, and may be emitted directly to the atmosphere, or captured and reinjected into underground formations, used for EOR, or sold for other commercial uses. A further discussion of CO₂ used in EOR is described in the Energy Chapter under the text box titled “Carbon Dioxide Transport, Injection, and Geological Storage.” The only CO₂ consumption that is accounted for in this analysis is CO₂ produced from naturally-occurring CO₂ reservoirs that is used in commercial applications other than EOR.

There are currently three facilities (one in Mississippi and two in New Mexico) producing CO₂ from naturally occurring CO₂ reservoirs for use in both EOR and in other commercial applications (e.g., chemical manufacturing, food production). A fourth facility in Colorado is producing CO₂ from naturally occurring CO₂ reservoirs for commercial applications only. There are other naturally occurring CO₂ reservoirs, mostly located in the western United States, that produce CO₂ but they are only producing CO₂ for EOR applications, not for other commercial applications (Allis et al. 2000). Carbon dioxide production from these facilities is discussed in the Energy Chapter.

In 2010, the amount of CO₂ produced by the Colorado, Mississippi, and New Mexico facilities for commercial applications and subsequently emitted to the atmosphere was 2.2 Tg CO₂ Eq. (2,203 Gg) (see Table 4-45). This is an increase of 23 percent from the previous year and an increase of 56 percent since 1990. This increase was largely due to an increase in production at the Mississippi facility, despite the low percentage (13 percent) of the facility’s total reported production that was used for commercial applications in 2010.

Table 4-45: CO₂ Emissions from CO₂ Consumption (Tg CO₂ Eq. and Gg)

Year	Tg CO₂ Eq.	Gg
1990	1.4	1,416
2005	1.3	1,321
2006	1.7	1,709
2007	1.9	1,867
2008	1.8	1,780
2009	1.8	1,784
2010	2.2	2,203

Methodology

CO₂ emission estimates for 1990 through 2010 were based on production data for the four facilities currently producing CO₂ from naturally-occurring CO₂ reservoirs for use in non-EOR applications. Some of the CO₂ produced by these facilities is used for EOR and some is used in other commercial applications (e.g., chemical manufacturing, food production). It is assumed that 100 percent of the CO₂ production used in commercial applications other than EOR is eventually released into the atmosphere.

CO₂ production data for the Jackson Dome, Mississippi facility and the percentage of production that was used for non-EOR applications were obtained from Advanced Resources International (ARI 2006, 2007) for 1990 to 2000

¹³⁵ There are currently four known electric power plants operating in the U.S. that capture CO₂ for use as food-grade CO₂ or other industrial processes; however, insufficient data prevents estimating emissions from these activities as part of CO₂ Consumption.

and from the Annual Reports of Denbury Resources (Denbury Resources 2002 through 2011) for 2001 to 2010 (see Table 4-46). Denbury Resources reported the average CO₂ production in units of MMCF CO₂ per day for 2001 through 2010 and reported the percentage of the total average annual production that was used for EOR. Production from 1990 to 2000 was set equal to 2001 production. Carbon dioxide production data for the Bravo Dome, New Mexico facilities were obtained from ARI (ARI 1990 through 2011). Data for the West Bravo Dome facility was only available for 2009 and 2010. The percentage of total production that was used for non-EOR applications were obtained from the New Mexico Bureau of Geology and Mineral Resources (Broadhead 2003 and New Mexico Bureau of Geology and Mineral Resources 2006). Production data for the McCallum Dome, Colorado facility were obtained from the Colorado Oil and Gas Conservation Commission (COGCC) for 1999 through 2010 (COGCC 2011). Production data for 1990 to 1998 and percentage of production used for EOR were assumed to be the same as for 1999.

Table 4-46: CO₂ Production (Gg CO₂) and the Percent Used for Non-EOR Applications

Year	Jackson Dome CO ₂ Production (Gg) (% Non-EOR)	Bravo Dome CO ₂ Production (Gg) (% Non-EOR)	West Bravo Dome CO ₂ Production (Gg) (% Non- EOR)	McCallum Dome CO ₂ Production (Gg) (% Non- EOR)
1990	1,353 (100%)	6,301 (1%)	-	0.07 (100%)
2005	4,677 (27%)	5,798 (1%)	-	0.06(100%)
2006	6,610 (25%)	5,605 (1%)	-	0.06(100%)
2007	9,529 (19%)	5,605 (1%)	-	0.07(100%)
2008	12,312 (14%)	5,605 (1%)	-	0.07(100%)
2009	13,201 (13%)	4,639 (1%)	2,126 (1%)	0.02(100%)
2010	16,487 (13%)	4,832 (1%)	870 (1%)	0.05(100%)

Uncertainty and Time-Series Consistency

Uncertainty is associated with the number of facilities that are currently producing CO₂ from naturally occurring CO₂ reservoirs for commercial uses other than EOR, and for which the CO₂ emissions are not accounted for elsewhere. Research indicates that there are only two such facilities, which are in New Mexico and Mississippi; however, additional facilities may exist that have not been identified. In addition, it is possible that CO₂ recovery exists in particular production and end-use sectors that are not accounted for elsewhere. Such recovery may or may not affect the overall estimate of CO₂ emissions from that sector depending upon the end use to which the recovered CO₂ is applied. Further research is required to determine whether CO₂ is being recovered from other facilities for application to end uses that are not accounted for elsewhere.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-47. Carbon dioxide consumption CO₂ emissions were estimated to be between 1.6 and 2.9 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 25 percent below to 30 percent above the emission estimate of 2.2 Tg CO₂ Eq.

Table 4-47: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from CO₂ Consumption (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^a			
			(Tg CO ₂ Eq.)		Lower Bound	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
CO ₂ Consumption	CO ₂	2.2	1.6	2.9	-25%	+30%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section,

above.

Recalculations Discussion

For the current Inventory, two new facilities, the West Bravo and McCallum domes, were added to the time series. The impact of these facilities upon emission estimates for the time series, relative to the previous report, is negligible.

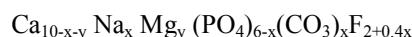
Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Carbon Dioxide Consumption source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹³⁶

4.13. Phosphoric Acid Production (IPCC Source Category 2B5)

Phosphoric acid (H_3PO_4) is a basic raw material in the production of phosphate-based fertilizers. Phosphate rock is mined in Florida, North Carolina, Idaho, Utah, and other areas of the United States and is used primarily as a raw material for phosphoric acid production. The production of phosphoric acid from phosphate rock produces byproduct gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), referred to as phosphogypsum.

The composition of natural phosphate rock varies depending upon the location where it is mined. Natural phosphate rock mined in the United States generally contains inorganic C in the form of calcium carbonate (limestone) and also may contain organic C. The chemical composition of phosphate rock (francolite) mined in Florida is:

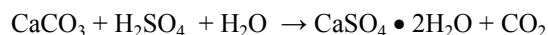


The calcium carbonate component of the phosphate rock is integral to the phosphate rock chemistry. Phosphate rock can also contain organic C that is physically incorporated into the mined rock but is not an integral component of the phosphate rock chemistry. Phosphoric acid production from natural phosphate rock is a source of CO_2 emissions, due to the chemical reaction of the inorganic C (calcium carbonate) component of the phosphate rock.

The phosphoric acid production process involves chemical reaction of the calcium phosphate ($\text{Ca}_3(\text{PO}_4)_2$) component of the phosphate rock with sulfuric acid (H_2SO_4) and recirculated phosphoric acid (H_3PO_4) (EFMA 2000). The primary chemical reactions for the production of phosphoric acid from phosphate rock are:



The limestone (CaCO_3) component of the phosphate rock reacts with the sulfuric acid in the phosphoric acid production process to produce calcium sulfate (phosphogypsum) and CO_2 . The chemical reaction for the limestone-sulfuric acid reaction is:



Total marketable phosphate rock production in 2010 was 25.8 million metric tons (USGS 2011). Approximately 87 percent of domestic phosphate rock production was mined in Florida and North Carolina, while approximately 13 percent of production was mined in Idaho and Utah. Total imports of phosphate rock in 2010 were 2.4 million metric tons (USGS 2011). The vast majority, 99 percent, of imported phosphate rock is sourced from Morocco (USGS 2005). Marketable phosphate rock production, including domestic production and imports for consumption stayed relatively flat between 2009 and 2010, decreasing by 2.3 percent between 2009 and 2010. Over the 1990 to 2010 period, domestic production has decreased by nearly 48 percent. Total CO_2 emissions from phosphoric acid production were 1.0 Tg CO_2 Eq. (1,017 Gg) in 2010 (see Table 4-48). After experiencing weak market conditions

¹³⁶ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

due to the global economic downturn in 2008 and 2009, demand for and trade in phosphate rock increased in 2010 (USGS 2011).

Table 4-48: CO₂ Emissions from Phosphoric Acid Production (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	1.5	1,529
2005	1.4	1,386
2006	1.2	1,167
2007	1.2	1,166
2008	1.2	1,187
2009	1.0	1,018
2010	1.0	1,017

Methodology

CO₂ emissions from production of phosphoric acid from phosphate rock are calculated by multiplying the average amount of calcium carbonate contained in the natural phosphate rock by the amount of phosphate rock that is used annually to produce phosphoric acid, accounting for domestic production and net imports for consumption.

The CO₂ emissions calculation methodology is based on the assumption that all of the inorganic C (calcium carbonate) content of the phosphate rock reacts to CO₂ in the phosphoric acid production process and is emitted with the stack gas. The methodology also assumes that none of the organic C content of the phosphate rock is converted to CO₂ and that all of the organic C content remains in the phosphoric acid product.

From 1993 to 2004, the *USGS Mineral Yearbook: Phosphate Rock* disaggregated phosphate rock mined annually in Florida and North Carolina from phosphate rock mined annually in Idaho and Utah, and reported the annual amounts of phosphate rock exported and imported for consumption (see Table 4-49). For the years 1990, 1991, 1992, and 2005 through 2010, only nationally aggregated mining data was reported by USGS. For these years, the breakdown of phosphate rock mined in Florida and North Carolina, and the amount mined in Idaho and Utah, are approximated using average share of U.S. production in those states from 1993 to 2004 data. Data for domestic production of phosphate rock, exports of phosphate rock (primarily from Florida and North Carolina), and imports of phosphate rock for consumption for 1990 through 2010 were obtained from *USGS Minerals Yearbook: Phosphate Rock* (USGS 1994 through 2011). From 2004 through 2010, the USGS reported no exports of phosphate rock from U.S. producers (USGS 2005 through 2011).

The carbonate content of phosphate rock varies depending upon where the material is mined. Composition data for domestically mined and imported phosphate rock were provided by the Florida Institute of Phosphate Research (FIPR 2003). Phosphate rock mined in Florida contains approximately 1 percent inorganic C, and phosphate rock imported from Morocco contains approximately 1.46 percent inorganic C. Calcined phosphate rock mined in North Carolina and Idaho contains approximately 0.41 percent and 0.27 percent inorganic C, respectively (see Table 4-50).

Carbonate content data for phosphate rock mined in Florida are used to calculate the CO₂ emissions from consumption of phosphate rock mined in Florida and North Carolina (87 percent of domestic production) and carbonate content data for phosphate rock mined in Morocco are used to calculate CO₂ emissions from consumption of imported phosphate rock. The CO₂ emissions calculation is based on the assumption that all of the domestic production of phosphate rock is used in uncalcined form. As of 2006, the USGS noted that one phosphate rock producer in Idaho produces calcined phosphate rock; however, no production data were available for this single producer (USGS 2006). Carbonate content data for uncalcined phosphate rock mined in Idaho and Utah (13 percent of domestic production) were not available, and carbonate content was therefore estimated from the carbonate content data for calcined phosphate rock mined in Idaho.

Table 4-49: Phosphate Rock Domestic Production, Exports, and Imports (Gg)

Location/Year	1990	2005	2006	2007	2008	2009	2010
U.S. Production ^a	49,800	36,100	30,100	29,700	30,200	26,400	25,800
<i>FL & NC</i>	42,494	31,227	26,037	25,691	26,123	22,836	22,317
<i>ID & UT</i>	7,306	4,874	4,064	4,010	4,077	3,564	3,483
Exports—FL & NC	6,240	-	-	-	-	-	-
Imports—Morocco	451	2,630	2,420	2,670	2,750	2,000	2,400
Total U.S. Consumption	44,011	38,730	32,520	32,370	32,950	28,400	28,200

^a USGS does not disaggregate production data regionally (FL & NC and ID & UT) for 1990 and 2005 through 2010. Data for those years are estimated based on the remaining time series distribution.

- Assumed equal to zero.

Table 4-50: Chemical Composition of Phosphate Rock (percent by weight)

Composition	Central Florida	North Florida	North Carolina (calcined)	Idaho (calcined)	Morocco
Total Carbon (as C)	1.60	1.76	0.76	0.60	1.56
Inorganic Carbon (as C)	1.00	0.93	0.41	0.27	1.46
Organic Carbon (as C)	0.60	0.83	0.35	-	0.10
Inorganic Carbon (as CO ₂)	3.67	3.43	1.50	1.00	5.00

Source: FIPR 2003

- Assumed equal to zero.

Uncertainty and Time-Series Consistency

Phosphate rock production data used in the emission calculations were developed by the USGS through monthly and semiannual voluntary surveys of the active phosphate rock mines during 2010. For previous years in the time series, USGS provided the data disaggregated regionally; however, beginning in 2006 only total U.S. phosphate rock production were reported. Regional production for 2010 was estimated based on regional production data from previous years and multiplied by regionally-specific emission factors. There is uncertainty associated with the degree to which the estimated 2010 regional production data represents actual production in those regions. Total U.S. phosphate rock production data are not considered to be a significant source of uncertainty because all the domestic phosphate rock producers report their annual production to the USGS. Data for exports of phosphate rock used in the emission calculation are reported by phosphate rock producers and are not considered to be a significant source of uncertainty. Data for imports for consumption are based on international trade data collected by the U.S. Census Bureau. These U.S. government economic data are not considered to be a significant source of uncertainty.

An additional source of uncertainty in the calculation of CO₂ emissions from phosphoric acid production is the carbonate composition of phosphate rock; the composition of phosphate rock varies depending upon where the material is mined, and may also vary over time. Another source of uncertainty is the disposition of the organic C content of the phosphate rock. A representative of the FIPR indicated that in the phosphoric acid production process, the organic C content of the mined phosphate rock generally remains in the phosphoric acid product, which is what produces the color of the phosphoric acid product (FIPR 2003a). Organic C is therefore not included in the calculation of CO₂ emissions from phosphoric acid production.

A third source of uncertainty is the assumption that all domestically-produced phosphate rock is used in phosphoric acid production and used without first being calcined. Calcination of the phosphate rock would result in conversion of some of the organic C in the phosphate rock into CO₂. However, according to the USGS, only one producer in Idaho is currently calcining phosphate rock, and no data were available concerning the annual production of this single producer (USGS 2005). For available years, total production of phosphate rock in Utah and Idaho combined amounts to approximately 13 percent of total domestic production on average (USGS 1994 through 2005).

Finally, USGS indicated that approximately 7 percent of domestically-produced phosphate rock is used to manufacture elemental phosphorus and other phosphorus-based chemicals, rather than phosphoric acid (USGS

2006). According to USGS, there is only one domestic producer of elemental phosphorus, in Idaho, and no data were available concerning the annual production of this single producer. Elemental phosphorus is produced by reducing phosphate rock with coal coke, and it is therefore assumed that 100 percent of the carbonate content of the phosphate rock will be converted to CO₂ in the elemental phosphorus production process. The calculation for CO₂ emissions is based on the assumption that phosphate rock consumption, for purposes other than phosphoric acid production, results in CO₂ emissions from 100 percent of the inorganic C content in phosphate rock, but none from the organic C content.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-51. Phosphoric acid production CO₂ emissions were estimated to be between 0.8 and 1.2 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 18 percent below and 18 percent above the emission estimate of 1.0 Tg CO₂ Eq.

Table 4-51: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Phosphoric Acid Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate	Uncertainty Range Relative to Emission Estimate ^a			
		(Tg CO ₂ Eq.)	(Tg CO ₂ Eq.)	(%)		
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Phosphoric Acid Production	CO ₂	1.0	0.8	1.2	-18%	+18%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations

Phosphate rock import and production values for 2008 and 2009 were updated relative to the previous Inventory based on recently published data (USGS 2011). This resulted in a decrease in 2008 and 2009 emissions by less than 1 percent and approximately 2 percent, respectively, relative to the previous report.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Phosphoric Acid Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹³⁷ Additionally, as a future improvement to the phosphoric acid uncertainty analysis, USGS Mineral Commodity Specialists will be contacted to verify uncertainty ranges associated with phosphate rock imports and exports.

4.14. Iron and Steel Production (IPCC Source Category 2C1) and Metallurgical Coke Production

The production of iron and steel is an energy-intensive activity that also generates process-related emissions of CO₂ and CH₄. Process emissions occur at each step of steel production from the production of raw materials to the refinement of iron to the making of crude steel. In the United States, steel is produced through both primary and secondary processes. Historically, primary production—using a basic oxygen furnace (BOF) with pig iron as the primary feedstock—has been the dominant method. But secondary production through the use scrap steel and electric arc furnaces (EAFs) has increased significantly in recent years due to the economic advantages of steel recycling, which has been driven by the increased availability of scrap steel. Total production of crude steel in the

¹³⁷ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

United States in the time period between 2000 and 2008 ranged from a low of 99,320,000 tons to a high of 109,879,000 tons (2001 and 2004, respectively). Due to the decrease in demand caused by the global economic downturn, especially from the automotive industry, crude steel production in the United States decreased to 65,460,000 tons in 2009. In 2010, crude steel production rebounded to 88,730,000 tons as economic conditions improved (AISI 2011a).

Metallurgical coke is an important input in the production of iron and steel. Coke is used to produce iron or pig iron feedstock from raw iron ore. The production of metallurgical coke from coking coal occurs both on-site at “integrated” iron and steel plants and off-site at “merchant” coke plants. Metallurgical coke is produced by heating coking coal in a coke oven in a low-oxygen environment. The process drives off the volatile components of the coking coal and produces coal (metallurgical) coke. Carbon containing byproducts of the metallurgical coke manufacturing process include coke oven gas, coal tar, coke breeze (small-grade coke oven coke with particle size <5mm) and light oil. Coke oven gas is recovered and used as fuel for underfiring the coke ovens and as process gas and fuel within the iron and steel mill. Small amounts of coke oven gas are also sold as synthetic natural gas outside of iron and steel mills (and are accounted for in the Energy chapter). Coal tar is used as a raw material to produce anodes used for primary aluminum production, electric arc furnace (EAF) steel production, and other electrolytic processes, and also is used in the production of other coal tar products. Light oil is sold to petroleum refiners who use the material as an additive for gasoline. The metallurgical coke production process produces CO₂ emissions and fugitive CH₄ emissions.

Iron is produced by first reducing iron oxide (iron ore) with metallurgical coke in a blast furnace. Iron can be introduced into the blast furnace in the form of raw iron ore, taconite pellets (9-16mm iron-containing spheres), briquettes, or sinter. In addition to metallurgical coke and iron, other inputs to the blast furnace include natural gas, fuel oil, and coke oven gas. The carbon in the metallurgical coke used in the blast furnace combines with oxides in the iron ore in a reducing atmosphere to produce blast furnace gas containing carbon monoxide (CO) and CO₂. The CO is then converted and emitted as CO₂ when combusted to either pre-heat the blast air used in the blast furnace or for other purposes at the steel mill. This pig iron or crude iron that is produced from this process contains about 3 to 5 percent carbon by weight. The pig iron production process in a blast furnace produces CO₂ emissions and fugitive CH₄ emissions.

Iron can also be produced through the direct reduction process; wherein, iron ore is reduced to metallic iron in the solid state at process temperatures less than 1000°C. Direct reduced iron production results in process emissions of CO₂ and emissions of CH₄ through the consumption of natural gas used during the reduction process.

Sintering is a thermal process by which fine iron-bearing particles, such as air emission control system dust, are baked, which causes the material to agglomerate into roughly one-inch pellets that are then recharged into the blast furnace for pig iron production. Iron ore particles may also be formed into larger pellets or briquettes by mechanical means, and then agglomerated by heating. The agglomerate is then crushed and screened to produce an iron-bearing feed that is charged into the blast furnace. The sintering process produces CO₂ and fugitive CH₄ emissions through the consumption of carbonaceous inputs (e.g., coke breeze) during the sintering process.

Steel is produced from varying levels of pig iron and scrap steel in specialized BOF and EAF steel-making furnaces. Carbon inputs to BOF steel-making furnaces include pig iron and scrap steel as well as natural gas, fuel oil, and fluxes (e.g., limestone, dolomite). In a BOF, the carbon in iron and scrap steel combines with high-purity oxygen to reduce the carbon content of the metal to the amount desired for the specified grade of steel. EAFs use carbon electrodes, charge carbon and other materials (e.g., natural gas) to aid in melting metal inputs (primarily recycled scrap steel), which are refined and alloyed to produce the desired grade of steel. Carbon dioxide emissions occur in BOFs through the reduction process. In EAFs, CO₂ emissions result primarily from the consumption of carbon electrodes and also from the consumption of supplemental materials used to augment the melting process.

In addition to the production processes mentioned above, CO₂ is also generated at iron and steel mills through the consumption of process byproducts (e.g., blast furnace gas, coke oven gas) used for various purposes including heating, annealing, and electricity generation. Process byproducts sold for use as synthetic natural gas are deducted and reported in the Energy chapter (emissions associated with natural gas and fuel oil consumption for these purposes are reported in the Energy chapter).

The majority of CO₂ emissions from the iron and steel production process come from the use of metallurgical coke in the production of pig iron and from the consumption of other process byproducts at the iron and steel mill, with lesser amounts emitted from the use of flux and from the removal of carbon from pig iron used to produce steel.

Some carbon is also stored in the finished iron and steel products.

According to the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006), the production of metallurgical coke from coking coal is considered to be an energy use of fossil fuel and the use of coke in iron and steel production is considered to be an industrial process source. Therefore, the Guidelines suggest that emissions from the production of metallurgical coke should be reported separately in the Energy source, while emissions from coke consumption in iron and steel production should be reported in the industrial process source. However, the approaches and emission estimates for both metallurgical coke production and iron and steel production are both presented here because the activity data used to estimate emissions from metallurgical coke production have significant overlap with activity data used to estimate iron and steel production emissions. Further, some byproducts (e.g., coke oven gas) of the metallurgical coke production process are consumed during iron and steel production, and some byproducts of the iron and steel production process (e.g., blast furnace gas) are consumed during metallurgical coke production. Emissions associated with the consumption of these byproducts are attributed to point of consumption. As an example, CO₂ emissions associated with the combustion of coke oven gas in the blast furnace during pig iron production are attributed to pig iron production. Emissions associated with the use of conventional fuels (e.g., natural gas and fuel oil) for electricity generation, heating and annealing, or other miscellaneous purposes downstream of the iron and steelmaking furnaces are reported in the Energy chapter.

Metallurgical Coke Production

Emissions of CO₂ and CH₄ from metallurgical coke production in 2010 were 2.1 Tg CO₂ Eq. (2,084 Gg) and less than 0.00003 Tg CO₂ Eq. (less than 0.001 Gg), respectively (see Table 4-52 and Table 4-53), totaling 2.1 Tg CO₂ Eq. Emissions increased in 2010 yet have decreased overall since 1990. In 2010, domestic coke production increased by 35 percent but has decreased overall since 1990. Coke production in 2010 was 28 percent lower than in 2000 and 46 percent below 1990. Overall, emissions from metallurgical coke production have declined by 16 percent (0.4 Tg CO₂ Eq.) from 1990 to 2010.

Table 4-52: CO₂ and CH₄ Emissions from Metallurgical Coke Production (Tg CO₂ Eq.)

Year	1990	2005	2006	2007	2008	2009	2010
CO ₂	2.5	2.0	1.9	2.1	2.3	1.0	2.1
CH ₄	+	+	+	+	+	+	+
Total	2.5	2.0	1.9	2.1	2.3	1.0	2.1

+ Does not exceed 0.05 Tg CO₂ Eq.

Table 4-53: CO₂ and CH₄ Emissions from Metallurgical Coke Production (Gg)

Year	1990	2005	2006	2007	2008	2009	2010
CO ₂	2,470	2,043	1,919	2,055	2,334	956	2,084
CH ₄	+	+	+	+	+	+	+

+ Does not exceed 0.5 Gg

Iron and Steel Production

Emissions of CO₂ and CH₄ from iron and steel production in 2010 were 52.2 Tg CO₂ Eq. (52,192 Gg) and 0.5 Tg CO₂ Eq. (24.5 Gg), respectively (see Table 4-54 through Table 4-57), totaling approximately 52.7 Tg CO₂ Eq. Emissions increased in 2010—largely due to increased steel production associated with improved economic conditions—but have decreased overall since 1990 due to restructuring of the industry, technological improvements, and increased scrap steel utilization. Carbon dioxide emission estimates include emissions from the consumption of carbonaceous materials in the blast furnace, EAF, and BOF as well as blast furnace gas and coke oven gas consumption for other activities at the steel mill.

In 2010, domestic production of pig iron increased by 41 percent from 2009 levels. Overall, domestic pig iron production has declined since the 1990s. Pig iron production in 2010 was 44 percent lower than in 2000 and 46 percent below 1990. Carbon dioxide emissions from steel production have increased by 5 percent (0.4 Tg CO₂ Eq.)

since 1990, while overall CO₂ emissions from iron and steel production have declined by 46 percent (44.9 Tg CO₂ Eq.) from 1990 to 2010.

Table 4-54: CO₂ Emissions from Iron and Steel Production (Tg CO₂ Eq.)

Year	1990	2005	2006	2007	2008	2009	2010
Sinter Production	2.4	1.7	1.4	1.4	1.3	0.8	1.0
Iron Production	47.9	19.6	24.0	27.3	25.8	16.0	19.0
Steel Production	7.5	8.5	8.9	9.4	7.5	6.6	7.8
Other Activities ^a	39.3	34.2	32.6	31.0	29.1	17.8	24.3
Total	97.1	64.0	66.9	69.1	63.8	41.2	52.2

Note: Totals may not sum due to independent rounding.

^a Includes emissions from blast furnace gas and coke oven gas combustion for activities at the steel mill other than consumption in blast furnace, EAFs, or BOFs.

Table 4-55: CO₂ Emissions from Iron and Steel Production (Gg)

Year	1990	2005	2006	2007	2008	2009	2010
Sinter Production	2,448	1,663	1,418	1,383	1,299	763	1,045
Iron Production	47,944	19,645	24,010	27,353	25,773	15,995	19,042
Steel Production	7,476	8,489	8,924	9,384	7,540	6,585	7,844
Other Activities ^a	39,256	34,160	32,583	30,964	29,146	17,815	24,260
Total	97,123	63,957	66,934	69,083	63,758	41,157	52,192

Note: Totals may not sum due to independent rounding.

^a Includes emissions from blast furnace gas and coke oven gas combustion for activities at the steel mill other than consumption in blast furnace, EAFs, or BOFs.

Table 4-56: CH₄ Emissions from Iron and Steel Production (Tg CO₂ Eq.)

Year	1990	2005	2006	2007	2008	2009	2010
Sinter Production	+	+	+	+	+	+	+
Iron Production	0.9	0.7	0.7	0.7	0.6	0.4	0.5
Total	1.0	0.7	0.7	0.7	0.6	0.4	0.5

+ Does not exceed 0.05 Tg CO₂ Eq.

Note: Totals may not sum due to independent rounding.

Table 4-57: CH₄ Emissions from Iron and Steel Production (Gg)

Year	1990	2005	2006	2007	2008	2009	2010
Sinter Production	0.9	0.6	0.5	0.5	0.4	0.3	0.4
Iron Production	44.7	33.5	34.1	32.7	30.4	17.1	24.2
Total	45.6	34.1	34.6	33.2	30.8	17.4	24.5

Note: Totals may not sum due to independent rounding.

Methodology

Emission estimates presented in this chapter are largely based on Tier 2 methodologies provided by the 2006 IPCC *Guidelines for National Greenhouse Gas Inventories* (IPCC 2006). These Tier 2 methodologies call for a mass balance accounting of the carbonaceous inputs and outputs during the iron and steel production process and the metallurgical coke production process. Tier 1 methods are used for certain iron and steel production processes (e.g. DRI production) for which available data are insufficient for utilizing a Tier 2 method.

Metallurgical Coke Production

Coking coal is used to manufacture metallurgical (coal) coke that is used primarily as a reducing agent in the production of iron and steel, but is also used in the production of other metals including lead and zinc (see Lead Production and Zinc Production in this chapter). Emissions associated with producing metallurgical coke from coking coal are estimated and reported separately from emissions that result from the iron and steel production process. To estimate emission from metallurgical coke production, a Tier 2 method provided by the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006) was utilized. The amount of carbon contained in materials produced during the metallurgical coke production process (i.e., coke, coke breeze, coke oven gas, and coal tar) is deducted from the amount of carbon contained in materials consumed during the metallurgical coke production process (i.e., natural gas, blast furnace gas, coking coal). Light oil, which is produced during the metallurgical coke production process, is excluded from the deductions due to data limitations. The amount of carbon contained in these materials is calculated by multiplying the material-specific carbon content by the amount of material consumed or produced (see Table 4-58). The amount of coal tar produced was approximated using a production factor of 0.03 tons of coal tar per ton of coking coal consumed. The amount of coke breeze produced was approximated using a production factor of 0.075 tons of coke breeze per ton of coking coal consumed. Data on the consumption of carbonaceous materials (other than coking coal) as well as coke oven gas production were available for integrated steel mills only (i.e., steel mills with co-located coke plants). Therefore, carbonaceous material (other than coking coal) consumption and coke oven gas production were excluded from emission estimates for merchant coke plants. Carbon contained in coke oven gas used for coke-oven underfiring was not included in the deductions to avoid double-counting.

Table 4-58: Material Carbon Contents for Metallurgical Coke Production

Material	kg C/kg
Coal Tar	0.62
Coke	0.83
Coke Breeze	0.83
Coking Coal	0.73
Material	kg C/GJ
Coke Oven Gas	12.1
Blast Furnace Gas	70.8

Source: IPCC 2006, Table 4.3. Coke Oven Gas and Blast Furnace Gas, Table 1.3.

The production processes for metallurgical coke production results in fugitive emissions of CH₄, which are emitted via leaks in the production equipment rather than through the emission stacks or vents of the production plants. The fugitive emissions were calculated by applying Tier 1 emission factors (0.1 g CH₄ per metric ton) taken from the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006) for metallurgical coke production.

Data relating to the mass of coking coal consumed at metallurgical coke plants and the mass of metallurgical coke produced at coke plants were taken from the Energy Information Administration (EIA), Quarterly Coal Report October through December (EIA 1998 through 2011d) (see Table 4-59). Data on the volume of natural gas consumption, blast furnace gas consumption, and coke oven gas production for metallurgical coke production at integrated steel mills were obtained from the American Iron and Steel Institute (AISI), *Annual Statistical Report* (AISI 2004 through 2011a) and through personal communications with AISI (2008b) (see Table 4-60). The factor for the quantity of coal tar produced per ton of coking coal consumed was provided by AISI (2008b). The factor for the quantity of coke breeze produced per ton of coking coal consumed was obtained through Table 2-1 of the report *Energy and Environmental Profile of the U.S. Iron and Steel Industry* (DOE 2000). Data on natural gas consumption and coke oven gas production at merchant coke plants were not available and were excluded from the emission estimate. Carbon contents for coking coal, metallurgical coke, coal tar, coke oven gas, and blast furnace gas were provided by the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006). The C content for coke breeze was assumed to equal the C content of coke.

Table 4-59: Production and Consumption Data for the Calculation of CO₂ and CH₄ Emissions from Metallurgical Coke Production (Thousand Metric Tons)

Source/Activity Data	1990	2005	2006	2007	2008	2009	2010
Metallurgical Coke Production							
Coking Coal Consumption at Coke Plants	35,269	21,259	20,827	20,607	20,022	13,904	19,135
Coke Production at Coke Plants	25,054	15,167	14,882	14,698	14,194	10,109	13,628
Coal Breeze Production	2,645	1,594	1,562	1,546	1,502	1,043	1,435
Coal Tar Production	1,058	638	625	618	601	417	574

Table 4-60: Production and Consumption Data for the Calculation of CO₂ Emissions from Metallurgical Coke Production (million ft³)

Source/Activity Data	1990	2005	2006	2007	2008	2009	2010
Metallurgical Coke Production							
Coke Oven Gas Production ^a	250,767	114,213	114,386	109,912	103,191	66,155	95,405
Natural Gas Consumption	599	2,996	3,277	3,309	3,134	2,121	3,108
Blast Furnace Gas Consumption	24,602	4,460	5,505	5,144	4,829	2,435	3,181

^a Includes coke oven gas used for purposes other than coke oven underfiring only.

Iron and Steel Production

Emissions of CO₂ from sinter production and direct reduced iron production were estimated by multiplying total national sinter production and the total national direct reduced iron production by Tier 1 CO₂ emission factors (see Table 4-61). Because estimates of sinter production and direct reduced iron production were not available, production was assumed to equal consumption.

Table 4-61: CO₂ Emission Factors for Sinter Production and Direct Reduced Iron Production

Material Produced	Metric Ton CO ₂ /Metric Ton
Sinter	0.2
Direct Reduced Iron	0.7

Source: IPCC 2006, Table 4.1.

To estimate emissions from pig iron production in the blast furnace, the amount of C contained in the produced pig iron and blast furnace gas were deducted from the amount of C contained in inputs (i.e., metallurgical coke, sinter, natural ore, pellets, natural gas, fuel oil, coke oven gas, direct coal injection). The C contained in the pig iron, blast furnace gas, and blast furnace inputs was estimated by multiplying the material-specific C content by each material type (see Table 4-62). Carbon in blast furnace gas used to pre-heat the blast furnace air is combusted to form CO₂ during this process.

Emissions from steel production in EAFs were estimated by deducting the C contained in the steel produced from the carbon contained in the EAF anode, charge carbon, and scrap steel added to the EAF. Small amounts of C from direct reduced iron, pig iron, and flux additions to the EAFs were also included in the EAF calculation. For BOFs, estimates of C contained in BOF steel were deducted from carbon contained in inputs such as natural gas, coke oven gas, fluxes, and pig iron. In each case, the C was calculated by multiplying material-specific carbon contents by each material type (see Table 4-62). For EAFs, the amount of EAF anode consumed was approximated by multiplying total EAF steel production by the amount of EAF anode consumed per metric ton of steel produced (0.002 metric tons EAF anode per metric ton steel produced (AISI 2008b)). The amount of flux (e.g., limestone and dolomite) used during steel manufacture was deducted from the Limestone and Dolomite Use source category to avoid double-counting.

CO₂ emissions from the consumption of blast furnace gas and coke oven gas for other activities occurring at the steel mill were estimated by multiplying the amount of these materials consumed for these purposes by the material-specific C content (see Table 4-62).

CO₂ emissions associated with the sinter production, direct reduced iron production, pig iron production, steel production, and other steel mill activities were summed to calculate the total CO₂ emissions from iron and steel production (see Table 4-54 and Table 4-55).

Table 4-62: Material Carbon Contents for Iron and Steel Production

Material	kg C/kg
Coke	0.83
Direct Reduced Iron	0.02
Dolomite	0.13
EAF Carbon Electrodes	0.82
EAF Charge Carbon	0.83
Limestone	0.12
Pig Iron	0.04
Steel	0.01
Material	kg C/GJ
Coke Oven Gas	12.1
Blast Furnace Gas	70.8

Source: IPCC 2006, Table 4.3. Coke Oven Gas and Blast Furnace Gas, Table 1.3.

The production processes for sinter and pig iron result in fugitive emissions of CH₄, which are emitted via leaks in the production equipment rather than through the emission stacks or vents of the production plants. The fugitive emissions were calculated by applying Tier 1 emission factors taken from the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006) for sinter production and the 1995 IPCC Guidelines (IPCC/UNEP/OECD/IEA 1995) (see Table 4-63) for pig iron production. The production of direct reduced iron also results in emissions of CH₄ through the consumption of fossil fuels (e.g., natural gas); however, these emissions estimates are excluded due to data limitations.

Table 4-63: CH₄ Emission Factors for Sinter and Pig Iron Production

Material Produced	Factor	Unit
Pig Iron	0.9	g CH ₄ /kg
Sinter	0.07	kg CH ₄ /metric ton

Source: Sinter (IPCC 2006, Table 4.2), Pig Iron (IPCC/UNEP/OECD/IEA 1995, Table 2.2)

Sinter consumption and direct reduced iron consumption data were obtained from AISI's Annual Statistical Report (AISI 2004 through 2011a) and through personal communications with AISI (2008b) (see Table 4-64). Data on direct reduced iron consumed in EAFs were not available for the years 1990, 1991, 1999, 2006, 2007, 2008, 2009, and 2010. EAF direct reduced iron consumption in 1990 and 1991 were assumed to equal consumption in 1992, and consumption in 1999 was assumed to equal the average of 1998 and 2000. EAF consumption in 2006, 2007, 2008, 2009, and 2010 were calculated by multiplying the total DRI consumption for all furnaces as provided in the 2010 AISI Annual Statistical Report by the EAF share of total DRI consumption in 2005 (the most recent year that data was available for EAF vs. BOF consumption of DRI). Data on direct reduced iron consumed in BOFs were not available for the years 1990 through 1994, 1999, 2006, 2007, 2008, 2009, and 2010. BOF direct reduced iron consumption in 1990 through 1994 was assumed to equal consumption in 1995, and consumption in 1999 was assumed to equal the average of 1998 and 2000. BOF consumption in 2006, 2007, 2008, 2009, and 2010 were calculated by multiplying the total DRI consumption for all furnaces as provided in the USGS Mineral Industry Survey: Iron and Steel Scrap in December 2010 (USGS 2011) by the BOF share of total DRI consumption in 2005 (the most recent year that data was available from the AISI Annual Statistical Reports for EAF vs. BOF consumption of DRI).¹³⁸

The Tier 1 CO₂ emission factors for sinter production and direct reduced iron production were obtained through the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC 2006). Data for pig iron production, coke, natural gas, fuel oil, sinter, and pellets consumed in the blast furnace; pig iron production; and blast furnace gas produced at the iron and steel mill and used in the metallurgical coke ovens and other steel mill activities were obtained from AISI's Annual Statistical Report (AISI 2004 through 2011a) and through personal communications with AISI (2008b) (see Table 4-65).

Data for EAF steel production, flux, EAF charge carbon, direct reduced iron, pig iron, scrap steel, and natural gas consumption were obtained from AISI's Annual Statistical Report (AISI 2004 through 2011a) and through personal communications with AISI (2011b and 2008b). The factor for the quantity of EAF anode consumed per ton of EAF steel produced was provided by AISI (AISI 2008b). Data for BOF steel production, flux, direct reduced iron, pig iron, scrap steel, natural gas, natural ore, pellet sinter consumption as well as BOF steel production were obtained from AISI's Annual Statistical Report (AISI 2004 through 2011a) and through personal communications with AISI (2008b). Because data on pig iron consumption in BOFs and EAFs were not available for 2006, 2007, and 2008 while scrap steel consumption data in BOFs and EAFs were not available for 2006 and 2007, values for these years were calculated by multiplying the total pig iron and scrap steel consumption for all furnaces as provided in the USGS Minerals Survey: Iron and Steel Scrap in December 2010 (USGS 2011) by the BOF and EAF shares of total pig iron and scrap consumption in 2005 (the most recent year that data was available from the AISI Annual Statistical Reports for EAF vs. BOF consumption of pig iron and scrap steel).¹³⁹ Because data for pig iron consumption in EAFs was also not available in 2003 and 2004, the average of 2002 and 2005 pig iron consumption data were used. Data on coke oven gas and blast furnace gas consumed at the iron and steel mill other than in the EAF, BOF, or blast furnace were obtained from AISI's Annual Statistical Report (AISI 2004 through 2011a) and through personal communications with AISI (2008b).

Data on blast furnace gas and coke oven gas sold for use as synthetic natural gas were obtained from EIA's Natural Gas Annual 2010 (EIA 2011b). C contents for direct reduced iron, EAF carbon electrodes, EAF charge carbon,

¹³⁸ 2010 DRI consumption values were not yet available when the 2010 AISI Annual Statistical Report was published, so the USGS Minerals Survey was used as a proxy.

¹³⁹ 2010 pig iron and scrap steel consumption values were not yet available when the 2010 AISI Annual Statistical Report was published, so the USGS Minerals Survey was used as a proxy.

limestone, dolomite, pig iron, and steel were provided by the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC 2006). The C contents for natural gas, fuel oil, and direct injection coal were obtained from EIA 2011c and EPA 2010. Heat contents for the same fuels were obtained from EIA (1992, 2011a). Heat contents for coke oven gas and blast furnace gas were provided in Table 2-2 of the report Energy and Environmental Profile of the U.S. Iron and Steel Industry (DOE 2000).

Table 4-64: Production and Consumption Data for the Calculation of CO₂ and CH₄ Emissions from Iron and Steel Production (Thousand Metric Tons)

Source/Activity Data	1990	2005	2006	2007	2008	2009	2010
Sinter Production							
Sinter Production	12,239	8,315	7,088	6,914	6,497	3,814	5,225
Direct Reduced Iron Production							
Direct Reduced Iron Production	936	1,633	1,497	2,087	1,769	1,243	1,343
Pig Iron Production							
Coke Consumption	24,946	13,832	14,684	15,039	14,251	8,572	10,883
Pig Iron Production	49,669	37,222	37,904	36,337	33,730	19,019	26,844
Direct Injection Coal Consumption	1,485	2,573	2,526	2,734	2,578	1,674	2,279
EAF Steel Production							
EAF Anode and Charge Carbon Consumption	67	1,127	1,245	1,214	1,109	845	1,189
Scrap Steel Consumption	35,743	37,558	38,033	40,845	40,824	35,472	36,560
Flux Consumption	319	695	671	567	680	476	640
EAF Steel Production	33,511	52,194	56,071	57,004	52,791	36,725	49,339
BOF Steel Production							
Pig Iron Consumption	46,564	32,115	32,638	33,773	29,322	24,404	28,214
Scrap Steel Consumption	14,548	11,612	11,759	12,628	8,029	6,641	8,881
Flux Consumption	576	582	610	408	431	318	408
BOF Steel Production	43,973	42,705	42,119	41,099	39,105	22,659	31,158

Table 4-65: Production and Consumption Data for the Calculation of CO₂ Emissions from Iron and Steel Production (million ft³ unless otherwise specified)

Source/Activity Data	1990	2005	2006	2007	2008	2009	2010
Pig Iron Production							
Natural Gas Consumption	56,273	59,844	58,344	56,112	53,349	35,933	47,814
Fuel Oil Consumption (thousand gallons)	163,397	16,170	87,702	84,498	55,552	23,179	27,505
Coke Oven Gas Consumption	22,033	16,557	16,649	16,239	15,336	9,951	14,233
Blast Furnace Gas Production	1,439,380	1,299,980	1,236,526	1,173,588	1,104,674	672,486	911,180
EAF Steel Production							
Natural Gas Consumption	15,905	19,985	21,897	28,077	10,826	7,848	10,403
BOF Steel Production							
Coke Oven Gas Consumption	3,851	524	559	525	528	373	546
Other Activities							
Coke Oven Gas Consumption	224,883	97,132	97,178	93,148	87,327	55,831	80,626
Blast Furnace Gas Consumption	1,414,778	1,295,520	1,231,021	1,168,444	1,099,845	670,051	907,999

Uncertainty and Time-Series Consistency

The estimates of CO₂ and CH₄ emissions from metallurgical coke production are based on material production and consumption data and average carbon contents. Uncertainty is associated with the total U.S. coking coal consumption, total U.S. coke production and materials consumed during this process. Data for coking coal consumption and metallurgical coke production are from different data sources (EIA) than data for other carbonaceous materials consumed at coke plants (AISI), which does not include data for merchant coke plants. There is uncertainty associated with the fact that coal tar and coke breeze production were estimated based on coke production because coal tar and coke breeze production data were not available. Since merchant coke plant data is not included in the estimate of other carbonaceous materials consumed at coke plants, the mass balance equation for CO₂ from metallurgical coke production cannot be reasonably completed. Therefore, for the purpose of this analysis, uncertainty parameters are applied to primary data inputs to the calculation (i.e., coking coal consumption and metallurgical coke production) only.

The estimates of CO₂ emissions from iron and steel production are based on material production and consumption data and average C contents. There is uncertainty associated with the assumption that direct reduced iron and sinter consumption are equal to production. There is uncertainty associated with the assumption that all coal used for purposes other than coking coal is for direct injection coal. Some of this coal may be used for electricity generation. There is also uncertainty associated with the C contents for pellets, sinter, and natural ore, which are assumed to equal the C contents of direct reduced iron. For EAF steel production there is uncertainty associated with the amount of EAF anode and charge C consumed due to inconsistent data throughout the time series. Also for EAF steel production, there is uncertainty associated with the assumption that 100 percent of the natural gas attributed to “steelmaking furnaces” by AISI is process-related and nothing is combusted for energy purposes. Uncertainty is also associated with the use of process gases such as blast furnace gas and coke oven gas. Data are not available to differentiate between the use of these gases for processes at the steel mill versus for energy generation (e.g., electricity and steam generation); therefore, all consumption is attributed to iron and steel production. These data and C contents produce a relatively accurate estimate of CO₂ emissions. However, there are uncertainties associated with each.

For the purposes of the CH₄ calculation from iron and steel production it is assumed that all of the CH₄ escapes as fugitive emissions and that none of the CH₄ is captured in stacks or vents. Additionally, the CO₂ emissions calculation is not corrected by subtracting the C content of the CH₄, which means there may be a slight double counting of C as both CO₂ and CH₄.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-66 for metallurgical coke production and iron and steel production. Total CO₂ emissions from metallurgical coke production and iron and steel production were estimated to be between 45.5 and 63.2 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 16 percent below and 17 percent above the emission estimate of 54.3 Tg CO₂ Eq. Total CH₄ emissions from metallurgical coke production and iron and steel production were estimated to be 0.5 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 21 percent below and 22 percent above the emission estimate of 0.5 Tg CO₂ Eq.

Table 4-66: Tier 2 Quantitative Uncertainty Estimates for CO₂ and CH₄ Emissions from Iron and Steel Production and Metallurgical Coke Production (Tg. CO₂ Eq. and Percent)

Source	Gas	Uncertainty Range Relative to Emission Estimate ^a			
		2010 Emission Estimate (Tg CO ₂ Eq.)		(Tg CO ₂ Eq.)	
				Lower Bound	Upper Bound
Metallurgical Coke & Iron and Steel Production	CO ₂	54.3	45.5	63.2	-16%
Metallurgical Coke & Iron and Steel Production	CH ₄	0.5	0.4	0.6	+22%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section,

above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Iron and Steel Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹⁴⁰

Additional improvements include accounting for emissions estimates for the production of metallurgical coke to the Energy chapter as well as identifying the amount of carbonaceous materials, other than coking coal, consumed at merchant coke plants. Other potential improvements include identifying the amount of coal used for direct injection and the amount of coke breeze, coal tar, and light oil produced during coke production. Efforts will also be made to identify inputs for preparing Tier 2 estimates for sinter and direct reduced iron production, as well as identifying information to better characterize emissions from the use of process gases and fuels within the Energy and Industrial Processes chapters.

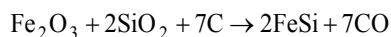
Recalculations Discussion

The average heat content of natural gas consumed in the United States is obtained directly from EIA and varies slightly each year (from 1024 to 1030 MMBTU/million cubic feet). In the previous Inventory, the 2009 heat content of natural gas was incorrectly applied to all historical years, so the year-to-year variation in the heat content of natural gas was not captured. This issue has been corrected for years 1990 through 2009 and decreased emissions for iron and steel production by less than 0.2 percent each year relative to the previous report.

4.15. Ferroalloy Production (IPCC Source Category 2C2)

Carbon dioxide and CH₄ are emitted from the production of several ferroalloys. Ferroalloys are composites of iron and other elements such as silicon, manganese, and chromium. When incorporated in alloy steels, ferroalloys are used to alter the material properties of the steel. Estimates from two types of ferrosilicon (25 to 55 percent and 56 to 95 percent silicon), silicon metal (about 98 percent silicon), and miscellaneous alloys (36 to 65 percent silicon) have been calculated. Emissions from the production of ferrochromium and ferromanganese are not included here because of the small number of manufacturers of these materials in the United States. Subsequently, government information disclosure rules prevent the publication of production data for these production facilities.

Similar to emissions from the production of iron and steel, CO₂ is emitted when metallurgical coke is oxidized during a high-temperature reaction with iron and the selected alloying element. Due to the strong reducing environment, CO is initially produced, and eventually oxidized to CO₂. A representative reaction equation for the production of 50 percent ferrosilicon is given below:



While most of the C contained in the process materials is released to the atmosphere as CO₂, a percentage is also released as CH₄ and other volatiles. The amount of CH₄ that is released is dependent on furnace efficiency, operation technique, and control technology.

Emissions of CO₂ from ferroalloy production in 2010 were 1.7 Tg CO₂ Eq. (1,663 Gg) (see Table 4-67 and Table 4-68), which is a 23 percent reduction since 1990. Emissions of CH₄ from ferroalloy production in 2010 were 0.01 Tg CO₂ Eq. (0.466 Gg), which is a 31 percent decrease since 1990.

Table 4-67: CO₂ and CH₄ Emissions from Ferroalloy Production (Tg CO₂ Eq.)

Year	1990	2005	2006	2007	2008	2009	2010
------	------	------	------	------	------	------	------

¹⁴⁰ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

CO ₂	2.2		1.4	1.5	1.6	1.6	1.5	1.7
CH ₄	+		+	+	+	+	+	+
Total	2.2		1.4	1.5	1.6	1.6	1.5	1.7

+ Does not exceed 0.05 Tg CO₂ Eq.

Note: Totals may not sum due to independent rounding.

Table 4-68: CO₂ and CH₄ Emissions from Ferroalloy Production (Gg)

Year	1990		2005	2006	2007	2008	2009	2010
CO ₂	2,152		1,392	1,505	1,552	1,599	1,469	1,663
CH ₄	1		+	+	+	+	+	+

+ Does not exceed 0.5 Gg.

Methodology

Emissions of CO₂ and CH₄ from ferroalloy production were calculated using a Tier 1 method from the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC 2006), specifically by multiplying annual ferroalloy production by material-specific default emission factors provided by IPCC (2006). For ferrosilicon alloys containing 25 to 55 percent silicon and miscellaneous alloys (including primarily magnesium-ferrosilicon, but also including other silicon alloys) containing 32 to 65 percent silicon, an emission factor for 45 percent silicon was applied for CO₂ (2.5 metric tons CO₂/metric ton of alloy produced) and an emission factor for 65 percent silicon was applied for CH₄ (1 kg CH₄/metric ton of alloy produced). Additionally, for ferrosilicon alloys containing 56 to 95 percent silicon, an emission factor for 75 percent silicon ferrosilicon was applied for both CO₂ and CH₄ (4 metric tons CO₂/metric ton alloy produced and 1 kg CH₄/metric ton of alloy produced, respectively). The emission factors for silicon metal equaled 5 metric tons CO₂/metric ton metal produced and 1.2 kg CH₄/metric ton metal produced. It was assumed that 100 percent of the ferroalloy production was produced using petroleum coke using an electric arc furnace process (IPCC 2006), although some ferroalloys may have been produced with coking coal, wood, other biomass, or graphite C inputs. The amount of petroleum coke consumed in ferroalloy production was calculated assuming that the petroleum coke used is 90 percent C and 10 percent inert material (Onder and Bagdoyan 1993).

Ferroalloy production data for 1990 through 2010 (see Table 4-69) were obtained from the USGS through personal communications with the USGS Silicon Commodity Specialist (Corathers 2011, Corathers 2012) and through the *Minerals Yearbook: Silicon Annual Report* (USGS 1991 through 2010). Because USGS does not provide estimates of silicon metal production for 2006-2010, 2005 production data are used. Until 1999, the USGS reported production of ferrosilicon containing 25 to 55 percent silicon separately from production of miscellaneous alloys containing 32 to 65 percent silicon; beginning in 1999, the USGS reported these as a single category (see Table 4-69). The composition data for petroleum coke was obtained from Onder and Bagdoyan (1993).

Table 4-69: Production of Ferroalloys (Metric Tons)

Year	Ferrosilicon 25%-55%	Ferrosilicon 56%-95%	Silicon Metal	Misc. Alloys 32-65%
1990	321,385	109,566	145,744	72,442
2005	123,000	86,100	148,000	NA
2006	164,000	88,700	148,000	NA
2007	180,000	90,600	148,000	NA
2008	193,000	94,000	148,000	NA
2009	123,932	104,855	148,000	NA
2010	153,000	135,000	148,000	NA

NA (Not Available)

Uncertainty and Time-Series Consistency

Although some ferroalloys may be produced using wood or other biomass as a C source, information and data

regarding these practices were not available. Emissions from ferroalloys produced with wood or other biomass would not be counted under this source because wood-based C is of biogenic origin.¹⁴¹ Even though emissions from ferroalloys produced with coking coal or graphite inputs would be counted in national trends, they may be generated with varying amounts of CO₂ per unit of ferroalloy produced. The most accurate method for these estimates would be to base calculations on the amount of reducing agent used in the process, rather than the amount of ferroalloys produced. These data, however, were not available, and are also often considered confidential business information.

Emissions of CH₄ from ferroalloy production will vary depending on furnace specifics, such as type, operation technique, and control technology. Higher heating temperatures and techniques such as sprinkle charging will reduce CH₄ emissions; however, specific furnace information was not available or included in the CH₄ emission estimates.

Also, annual ferroalloy production is now reported by the USGS in three broad categories: ferroalloys containing 25 to 55 percent silicon (including miscellaneous alloys), ferroalloys containing 56 to 95 percent silicon, and silicon metal. It was assumed that the IPCC emission factors apply to all of the ferroalloy production processes, including miscellaneous alloys. Finally, production data for silvery pig iron (alloys containing less than 25 percent silicon) are not reported by the USGS to avoid disclosing company proprietary data. Emissions from this production category, therefore, were not estimated.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-70. Ferroalloy production CO₂ emissions were estimated to be between 1.3 and 1.7 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 12 percent below and 12 percent above the emission estimate of 1.5 Tg CO₂ Eq. Ferroalloy production CH₄ emissions were estimated to be between a range of approximately 12 percent below and 12 percent above the emission estimate of 0.01 Tg CO₂ Eq.

Table 4-70: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Ferroalloy Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)		Uncertainty Range Relative to Emission Estimate ^a (Tg CO ₂ Eq.)			
				Lower Bound		Upper Bound	
Ferroalloy Production	CO ₂	1.7	1.5	1.3	1.9	-12%	+12%
Ferroalloy Production	CH ₄	+	+	+	+	-12%	+12%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

+ Does not exceed 0.05 Tg CO₂ Eq.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Ferroalloy Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹⁴² Additionally, research will be conducted to determine whether data are available concerning raw material consumption (e.g., coal coke, limestone and dolomite flux, etc.) for inclusion in ferroalloy production emission estimates.

¹⁴¹ Emissions and sinks of biogenic carbon are accounted for in the Land Use, Land-Use Change, and Forestry chapter.

¹⁴² See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

4.16. Aluminum Production (IPCC Source Category 2C3)

Aluminum is a light-weight, malleable, and corrosion-resistant metal that is used in many manufactured products, including aircraft, automobiles, bicycles, and kitchen utensils. As of last reporting, the United States was the fifth largest producer of primary aluminum, with approximately four percent of the world total (USGS 2011). The United States was also a major importer of primary aluminum. The production of primary aluminum—in addition to consuming large quantities of electricity—results in process-related emissions of CO₂ and two perfluorocarbons (PFCs): perfluoromethane (CF₄) and perfluoroethane (C₂F₆).

CO₂ is emitted during the aluminum smelting process when alumina (aluminum oxide, Al₂O₃) is reduced to aluminum using the Hall-Heroult reduction process. The reduction of the alumina occurs through electrolysis in a molten bath of natural or synthetic cryolite (Na₃AlF₆). The reduction cells contain a carbon lining that serves as the cathode. Carbon is also contained in the anode, which can be a carbon mass of paste, coke briquettes, or prebaked carbon blocks from petroleum coke. During reduction, most of this carbon is oxidized and released to the atmosphere as CO₂.

Process emissions of CO₂ from aluminum production were estimated to be 3.0 Tg CO₂ Eq. (3,009 Gg) in 2010 (see Table 4-71). The carbon anodes consumed during aluminum production consist of petroleum coke and, to a minor extent, coal tar pitch. The petroleum coke portion of the total CO₂ process emissions from aluminum production is considered to be a non-energy use of petroleum coke, and is accounted for here and not under the CO₂ from Fossil Fuel Combustion source category of the Energy sector. Similarly, the coal tar pitch portion of these CO₂ process emissions is accounted for here.

Table 4-71: CO₂ Emissions from Aluminum Production (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	6.8	6,831
2005	4.1	4,142
2006	3.8	3,801
2007	4.3	4,251
2008	4.5	4,477
2009	3.0	3,009
2010	3.0	3,009

In addition to CO₂ emissions, the aluminum production industry is also a source of PFC emissions. During the smelting process, when the alumina ore content of the electrolytic bath falls below critical levels required for electrolysis, rapid voltage increases occur, which are termed “anode effects.” These anode effects cause carbon from the anode and fluorine from the dissociated molten cryolite bath to combine, thereby producing fugitive emissions of CF₄ and C₂F₆. In general, the magnitude of emissions for a given smelter and level of production depends on the frequency and duration of these anode effects. As the frequency and duration of the anode effects increase, emissions increase.

Since 1990, emissions of CF₄ and C₂F₆ have declined by 92 percent and 89 percent, respectively, to 1.3 Tg CO₂ Eq. of CF₄ (0.19 Gg) and 0.3 Tg CO₂ Eq. of C₂F₆ (0.033 Gg) in 2010, as shown in Table 4-72 and Table 4-73. This decline is due both to reductions in domestic aluminum production and to actions taken by aluminum smelting companies to reduce the frequency and duration of anode effects. Since 1990, aluminum production has declined by 57 percent, while the combined CF₄ and C₂F₆ emission rate (per metric ton of aluminum produced) has been reduced by 80 percent.

Table 4-72: PFC Emissions from Aluminum Production (Tg CO₂ Eq.)

Year	CF ₄	C ₂ F ₆	Total
1990	15.8	2.7	18.4
2005	2.5	0.4	3.0
2006	2.1	0.4	2.5
2007	3.2	0.6	3.8
2008	2.2	0.5	2.7

2009	1.3	0.3	1.6
2010	1.3	0.3	1.6

Note: Totals may not sum due to independent rounding.

Table 4-73: PFC Emissions from Aluminum Production (Gg)

Year	CF ₄	C ₂ F ₆
1990	2.4	0.3
2005	0.4	+
2006	0.3	+
2007	0.5	0.1
2008	0.3	0.1
2009	0.2	+
2010	0.2	+

+ Does not exceed 0.05 Gg.

In 2010, U.S. primary aluminum production totaled approximately 1.7 million metric tons, less than half a percent increase from 2009 production levels (USAA 2011a). In 2010, five companies managed production at nine operational primary aluminum smelters. Two smelters were permanently closed in 2010. An additional five smelters were temporarily idled in 2010 (USGS 2011). During 2010, monthly U.S. primary aluminum production was less in January through April, and greater from June through December when compared to the corresponding month in 2009 (USAA 2011a).

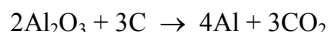
For 2011, total production was approximately 2.0 million metric tons compared to 1.7 million metric tons for the same period in 2010, a 15 percent increase (USAA 2012). Based on the increase in production, process CO₂ and PFC emissions are likely to be greater in 2011 compared to 2010 given no significant changes in process controls at operational facilities.

Methodology

Process CO₂ Emissions from Anode Consumption and Anode Baking

CO₂ emissions released during aluminum production were estimated by combining individual facility reported data with process-specific emissions modeling. These estimates are based on information gathered from EPA's Voluntary Aluminum Industrial Partnership (VAIP) program, U.S. Geological Survey (USGS) Mineral Commodity reviews, and The Aluminum Association (USAA) statistics, among other sources.

Most of the CO₂ emissions released during aluminum production occur during the electrolysis reaction of the carbon anode, as described by the following reaction:



For prebake smelter technologies, CO₂ is also emitted during the anode baking process. These emissions can account for approximately 10 percent of total process CO₂ emissions from prebake smelters.

Depending on the availability of smelter-specific data, the CO₂ emitted from electrolysis at each smelter was estimated from: (1) the smelter's annual anode consumption, (2) the smelter's annual aluminum production and rate of anode consumption (per ton of aluminum produced) for previous and/or following years, or, (3) the smelter's annual aluminum production and IPCC default CO₂ emission factors. The first approach tracks the consumption and carbon content of the anode, assuming that all carbon in the anode is converted to CO₂. Sulfur, ash, and other impurities in the anode are subtracted from the anode consumption to arrive at a C consumption figure. This approach corresponds to either the IPCC Tier 2 or Tier 3 method, depending on whether smelter-specific data on anode impurities are used. The second approach interpolates smelter-specific anode consumption rates to estimate emissions during years for which anode consumption data are not available. This approach avoids substantial errors and discontinuities that could be introduced by reverting to Tier 1 methods for those years. The last approach corresponds to the IPCC Tier 1 method (2006) and is used in the absence of present or historic anode consumption

data.

The equations used to estimate CO₂ emissions in the Tier 2 and 3 methods vary depending on smelter type (IPCC 2006). For Prebake cells, the process formula accounts for various parameters, including net anode consumption, and the sulfur, ash, and impurity content of the baked anode. For anode baking emissions, the formula accounts for packing coke consumption, the sulfur and ash content of the packing coke, as well as the pitch content and weight of baked anodes produced. For Söderberg cells, the process formula accounts for the weight of paste consumed per metric ton of aluminum produced, and pitch properties, including sulfur, hydrogen, and ash content.

Through the VAIP, anode consumption (and some anode impurity) data have been reported for 1990, 2000, 2003, 2004, 2005, 2006, 2007, 2008, and 2009. Where available, smelter-specific process data reported under the VAIP were used; however, if the data were incomplete or unavailable, information was supplemented using industry average values recommended by IPCC (2006). Smelter-specific CO₂ process data were provided by 18 of the 23 operating smelters in 1990 and 2000, by 14 out of 16 operating smelters in 2003 and 2004, 14 out of 15 operating smelters in 2005, 13 out of 14 operating smelters in 2006, 5 out of 14 operating smelters in 2007 and 2008, and 3 out of 13 operating smelters in 2009. For years where CO₂ process data were not reported by these companies, estimates were developed through linear interpolation, and/or assuming representative (e.g., previously reported or industry default) values.

In the absence of any previous historical smelter specific process data (i.e., 1 out of 9 smelters in 2010, 1 out of 13 smelters in 2009, 1 out of 14 smelters in 2006, 2007, and 2008, 1 out of 15 smelters in 2005, and 5 out of 23 smelters between 1990 and 2003), CO₂ emission estimates were estimated using Tier 1 Söderberg and/or Prebake emission factors (metric ton of CO₂ per metric ton of aluminum produced) from IPCC (2006).

Process PFC Emissions from Anode Effects

PFC emissions from aluminum production were estimated using a per-unit production emission factor that is expressed as a function of operating parameters (anode effect frequency and duration), as follows:

$$\text{PFC (CF}_4 \text{ or C}_2\text{F}_6\text{) kg/metric ton Al} = S \times (\text{Anode Effect Minutes/Cell-Day})$$

where,

$$S = \text{Slope coefficient ((kg PFC/metric ton Al)/(Anode Effect Minutes/Cell-Day))} \\ (\text{Anode Effect Minutes/Cell-Day}) = (\text{Anode Effect Frequency/Cell-Day}) \times \text{Anode Effect Duration (minutes)}$$

This approach corresponds to either the Tier 3 or the Tier 2 approach in the 2006 IPCC Guidelines, depending upon whether the slope-coefficient is smelter-specific (Tier 3) or technology-specific (Tier 2). For 1990 through 2010, smelter-specific slope coefficients were available and were used for smelters representing between 30 and 94 percent of U.S. primary aluminum production. The percentage changed from year to year as some smelters closed or changed hands and as the production at remaining smelters fluctuated. For smelters that did not report smelter-specific slope coefficients, IPCC technology-specific slope coefficients were applied (IPCC 2000, 2006). The slope coefficients were combined with smelter-specific anode effect data collected by aluminum companies and reported under the VAIP, to estimate emission factors over time. For 1990 through 2009, smelter-specific anode effect data were available for smelters representing between 80 and 100 percent of U.S. primary aluminum production. For 2010, no reported smelter-specific anode effect data were available. Where smelter-specific anode effect data were not available, representative values (e.g., previously reported or industry averages) were used.

For all smelters, emission factors were multiplied by annual production to estimate annual emissions at the smelter level. For 1990 through 2009, smelter-specific production data were available for smelters representing between 30 and 100 percent of U.S. primary aluminum production. (For the years after 2000, this percentage was near the high end of the range.) For 2010, no reported smelter-specific production data was available. Production at non-reporting smelters was estimated by calculating the difference between the production reported under VAIP and the total U.S. production supplied by USGS or USAA, and then allocating this difference to non-reporting smelters in proportion to their production capacity. Emissions were then aggregated across smelters to estimate national emissions.

Between 1990 and 2009, production data were provided under the VAIP by 21 of the 23 U.S. smelters that operated during at least part of that period. For the non-reporting smelters, production was estimated based on the difference between reporting smelters and national aluminum production levels (USAA 2011a), with allocation to specific

smelters based on reported production capacities (USGS 2011).

National primary aluminum production data for 2010 were obtained via The Aluminum Association (USAA 2011a). For 1990 through 2001, and 2006 (see Table 4-74) data were obtained from USGS, Mineral Industry Surveys: Aluminum Annual Report (USGS 1995, 1998, 2000, 2001, 2002, 2007). For 2002 through 2005, and 2007 through 2009 national aluminum production data were obtained from the USAA's Primary Aluminum Statistics (USAA 2004, 2005, 2006, 2008, 2009, 2010).

Table 4-74: Production of Primary Aluminum (Gg)

Year	Gg
1990	4,048
2005	2,478
2006	2,284
2007	2,560
2008	2,659
2009	1,727
2010	1,727

Uncertainty and Time Series Consistency

For CO₂, uncertainty was assigned to each of the parameters used to estimate CO₂ emissions. Uncertainty surrounding estimated production data was assumed to have a triangular distribution with a minimum value of zero and a maximum value corresponding to the reported production capacity (USGS 2011). For additional variables, such as net C consumption, and sulfur and ash content in baked anodes, estimates for uncertainties associated with reported and default data were obtained from IPCC (2006). A Monte Carlo analysis was applied to estimate the overall uncertainty of the CO₂ emission estimate for the U.S. aluminum industry as a whole, and the results are provided below.

To estimate the uncertainty associated with emissions of CF₄ and C₂F₆, the uncertainties associated with three variables were estimated for each smelter: (1) the quantity of aluminum produced, (2) the anode effect minutes per cell day (which may be reported directly or calculated as the product of anode effect frequency and anode effect duration), and, (3) the smelter- or technology-specific slope coefficient. A Monte Carlo analysis was then applied to estimate the overall uncertainty of the emission estimate for each smelter and for the U.S. aluminum industry as a whole.

The results of this Tier 2 quantitative uncertainty analysis are summarized in Table 4-75. Aluminum production-related CO₂ emissions were estimated to be between 1.5 and 3.1 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 49 percent below to 2 percent above the emission estimate of 3.0 Tg CO₂ Eq. Also, production-related CF₄ emissions were estimated to be between 0.6 and 1.3 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 50 percent below to 6 percent above the emission estimate of 1.3 Tg CO₂ Eq. Finally, aluminum production-related C₂F₆ emissions were estimated to be between 0.1 and 0.4 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 62 percent below to 15 percent above the emission estimate of 0.3 Tg CO₂ Eq.

Table 4-75: Tier 2 Quantitative Uncertainty Estimates for CO₂ and PFC Emissions from Aluminum Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to 2010 Emission Estimate ^a			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Aluminum Production	CO ₂	3.0	1.5	3.1	-49%	+2%
Aluminum Production	CF ₄	1.3	0.6	1.3	-50%	+6%
Aluminum Production	C ₂ F ₆	0.3	0.1	0.4	-62%	+15%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

The 2010 emission estimate was developed using either company-wide or site-specific PFC slope coefficients for all

but 1 of the 9 operating smelters where default IPCC (2006) slope data was used. In some cases, where smelters are owned by one company, data have been reported on a company-wide basis as totals or weighted averages. Consequently, in the Monte Carlo analysis, uncertainties in anode effect minutes per cell day, slope coefficients, and aluminum production have been applied to the company as a whole and not to each smelter. This probably overestimates the uncertainty associated with the cumulative emissions from these smelters, because errors that were in fact independent were treated as if they were correlated. It is therefore likely that the uncertainties calculated above for the total U.S. 2010 emission estimates for CF₄ and C₂F₆ are also overestimated.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations

In 2010, reported production data for one smelter was updated for the years 1990, 2000, and 2003. These data were used to recalculate emissions, and revised total PFC emissions by less than one percent for each of those years.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Aluminum Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹⁴³

4.17. Magnesium Production and Processing (IPCC Source Category 2C4)

The magnesium metal production and casting industry uses sulfur hexafluoride (SF₆) as a cover gas to prevent the rapid oxidation of molten magnesium in the presence of air. Sulfur hexafluoride has been used in this application around the world for more than twenty-five years. A dilute gaseous mixture of SF₆ with dry air and/or CO₂ is blown over molten magnesium metal to induce and stabilize the formation of a protective crust. A small portion of the SF₆ reacts with the magnesium to form a thin molecular film of mostly magnesium oxide and magnesium fluoride. The amount of SF₆ reacting in magnesium production and processing is considered to be negligible and thus all SF₆ used is assumed to be emitted into the atmosphere. Although alternative cover gases, such as AM-coverTM (containing HFC-134a), NovecTM 612 and dilute SO₂ systems can be used, many facilities in the United States are still using traditional SF₆ cover gas systems.

The magnesium industry emitted 1.3Tg CO₂ Eq. (0.05 Gg) of SF₆ in 2010, representing an increase of approximately 21 percent from 2009 emissions (See Table 4-76). The increase can be attributed to: increased demand for magnesium for use in iron and steel desulfurization as U.S. steel production recovered from the economic downturn (USGS 2011b); increased production and processing due to improving economic conditions and increased demand from the automotive industry (USGS 2011b). The increase was mitigated in part by continuing industry efforts to utilize SF₆ alternatives, such as NovecTM612 and sulfur dioxide, as part of the EPA's SF₆ Emission Reduction Partnership for the Magnesium Industry.

Table 4-76: SF₆ Emissions from Magnesium Production and Processing (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	5.4	0.2
2005	2.9	0.01
2006	2.9	0.1
2007	2.6	0.1
2008	1.9	0.1
2009	1.1	0.04

¹⁴³ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

Methodology

Emission estimates for the magnesium industry incorporate information provided by industry participants in EPA's SF₆ Emission Reduction Partnership for the Magnesium Industry. The Partnership started in 1999 and, currently, participating companies represent 100 percent of U.S. primary and secondary production and 90 percent of the casting sector production (i.e., die, sand, permanent mold, wrought, and anode casting). Absolute emissions for 1999 through 2010 from primary production, secondary production (i.e., recycling), and die casting were generally reported by Partnership participants. Partners reported their SF₆ consumption, which was assumed to be equivalent to emissions. When a partner did not report emissions, they were estimated based on the metal processed and emission rate reported by that partner in previous and (if available) subsequent years. Where data for subsequent years was not available, metal production and emissions rates were extrapolated based on the trend shown by partners reporting in the current and previous years. When it was determined a Partner is no longer in production, their metal production and emissions rates were set to zero if no activity information was available.

Emission factors for 2002 to 2006 for sand casting activities were also acquired through the Partnership. For 2007, 2008, 2009 and 2010, the sand casting partner did not report and the reported emission factor from 2005 was utilized as being representative of the industry. The 1999 through 2010 emissions from casting operations (other than die) were estimated by multiplying emission factors (kg SF₆ per metric ton of metal produced or processed) by the amount of metal produced or consumed. The emission factors for casting activities are provided below in Table 4-77. The emission factors for primary production, secondary production and sand casting are withheld to protect company-specific production information. However, the emission factor for primary production has not risen above the average 1995 partner value of 1.1 kg SF₆ per metric ton.

Die casting emissions for 1999 through 2010, which accounted for 15 to 52 percent of all SF₆ emissions from the U.S. magnesium industry during this period, were estimated based on information supplied by industry partners. From 2000 to 2010, partners accounted for all U.S. die casting that was tracked by USGS. In 1999, partners did not account for all die casting tracked by USGS, and, therefore, it was necessary to estimate the emissions of die casters who were not partners. Die casters who were not partners were assumed to be similar to partners who cast small parts. Due to process requirements, these casters consume larger quantities of SF₆ per metric ton of processed magnesium than casters that process large parts. Consequently, emission estimates from this group of die casters were developed using an average emission factor of 5.2 kg SF₆ per metric ton of magnesium. The emission factors for the other industry sectors (i.e., permanent mold, wrought, and anode casting) were based on discussions with industry representatives.

Table 4-77: SF₆ Emission Factors (kg SF₆ per metric ton of magnesium)

Year	Die Casting	Permanent Mold	Wrought	Anodes
1999	2.14 ^a	2	1	1
2000	0.72	2	1	1
2001	0.72	2	1	1
2002	0.71	2	1	1
2003	0.81	2	1	1
2004	0.81	2	1	1
2005	0.79	2	1	1
2006	0.86	2	1	1
2007	0.67	2	1	1
2008	1.15	2	1	1
2009	1.77	2	1	1
2010	2.51	2	1	1

^a Weighted average that includes an estimated emission factor of 5.2 kg SF₆ per metric ton of magnesium for die casters that do not participate in the Partnership.

Data used to develop SF₆ emission estimates were provided by the Magnesium Partnership participants and the USGS. U.S. magnesium consumption (casting) data from 1990 through 2010 were available from the USGS (USGS 2002, 2003, 2005, 2006, 2007, 2008, 2010, 2011). Emission factors from 1990 through 1998 were based on a

number of sources. Emission factors for primary production were available from U.S. primary producers for 1994 and 1995, and an emission factor for die casting of 4.1 kg per metric ton was available for the mid-1990s from an international survey (Gjestland & Magers 1996).

To estimate emissions for 1990 through 1998, industry emission factors were multiplied by the corresponding metal production and consumption (casting) statistics from USGS. The primary production emission factors were 1.2 kg per metric ton for 1990 through 1993, and 1.1 kg per metric ton for 1994 through 1997. For die casting, an emission factor of 4.1 kg per metric ton was used for the period 1990 through 1996. For 1996 through 1998, the emission factors for primary production and die casting were assumed to decline linearly to the level estimated based on partner reports in 1999. This assumption is consistent with the trend in SF₆ sales to the magnesium sector that is reported in the RAND survey of major SF₆ manufacturers, which shows a decline of 70 percent from 1996 to 1999 (RAND 2002). Sand casting emission factors for 2002 through 2010 were provided by the Magnesium Partnership participants, and 1990 through 2001 emission factors for this process were assumed to have been the same as the 2002 emission factor. The emission factor for secondary production from 1990 through 1998 was assumed to be constant at the 1999 average partner value. The emission factors for the other processes (i.e., permanent mold, wrought, and anode casting), about which less is known, were assumed to remain constant at levels defined in Table 4-77.

Uncertainty

To estimate the uncertainty surrounding the estimated 2009 SF₆ emissions from magnesium production and processing, the uncertainties associated with three variables were estimated (1) emissions reported by magnesium producers and processors that participate in the Magnesium Partnership, (2) emissions estimated for magnesium producers and processors that participate in the Partnership but did not report this year, and (3) emissions estimated for magnesium producers and processors that do not participate in the Partnership. An uncertainty of 5 percent was assigned to the data reported by each participant in the Partnership. If partners did not report emissions data during the current reporting year, SF₆ emissions data were estimated using available emission factor and production information reported in prior years; the extrapolation was based on the average trend for partners reporting in the current reporting year and the year prior. The uncertainty associated with the SF₆ usage estimate generated from the extrapolated emission factor and production information was estimated to be 30 percent for each year of extrapolation. The lone sand casting partner did not report in the past two reporting years and its activity and emission factor were held constant at 2005 levels due to a reporting anomaly in 2006 because of malfunctions at the facility. The uncertainty associated with the SF₆ usage for the sand casting partner was 52 percent. For those industry processes that are not represented in Partnership, such as permanent mold and wrought casting, SF₆ emissions were estimated using production and consumption statistics reported by USGS and estimated process-specific emission factors (see Table 4-77). The uncertainties associated with the emission factors and USGS-reported statistics were assumed to be 75 percent and 25 percent, respectively. Emissions associated with sand casting activities utilized a partner-reported emission factor with an uncertainty of 75 percent. In general, where precise quantitative information was not available on the uncertainty of a parameter, a conservative (upper-bound) value was used.

Additional uncertainties exist in these estimates that are not addressed in this methodology, such as the basic assumption that SF₆ neither reacts nor decomposes during use. The melt surface reactions and high temperatures associated with molten magnesium could potentially cause some gas degradation. Recent measurement studies have identified SF₆ cover gas degradation in die casting applications on the order of 20 percent (Bartos et al. 2007). Sulfur hexafluoride may also be used as a cover gas for the casting of molten aluminum with high magnesium content; however, the extent to which this technique is used in the United States is unknown.

The results of this Tier 2 quantitative uncertainty analysis are summarized in Table 4-78. SF₆ emissions associated with magnesium production and processing were estimated to be between 1.01 and 1.10 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 6 percent below to 5 percent above the 2008 emission estimate of 1.05 Tg CO₂ Eq. The uncertainty estimates for 2009 are lower relative to the 2008 reporting year which is likely due to the fact that emission estimates for this year are based more on actual reported data than last year with two emission sources using projected (highly uncertain) estimates.

Table 4-78: Tier 2 Quantitative Uncertainty Estimates for SF₆ Emissions from Magnesium Production and Processing (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^a			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Magnesium Production	SF ₆	1.3	1.23	1.33	-4%	+4%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Recalculations Discussion

The USGS 2010 Mineral Yearbook for Magnesium showed a revision in its estimate of sand casting production of magnesium for 2009 in the United States, revising its previous estimate of 44 metric tons in 2009 to 107 metric tons.

Planned Improvements

Cover gas research conducted over the last decade has found that SF₆ used for magnesium melt protection can have degradation rates on the order of 20 percent in die casting applications (Bartos et al. 2007). Current emission estimates assume (per the 2006 IPCC Guidelines, IPCC 2006) that all SF₆ utilized is emitted to the atmosphere. Additional research may lead to a revision of IPCC Guidelines to reflect this phenomenon and until such time, developments in this sector will be monitored for possible application to the inventory methodology. Another issue that will be addressed in future inventories is the likely adoption of alternate cover gases by U.S. magnesium producers and processors. These cover gases, which include AM-cover™ (containing HFC-134a) and Novec™ 612, have lower GWPs than SF₆, and tend to quickly degrade during their exposure to the molten metal. Magnesium producers and processors have already begun using these cover gases for 2006 through 2010 in a limited fashion; because the amounts being used by companies on the whole are low enough that they have a minor effect on the overall emissions from the industry, these emissions are only being monitored and recorded at this time.

4.18. Zinc Production (IPCC Source Category 2C5)

Zinc production in the United States consists of both primary and secondary processes. Primary production in the United States is conducted through the electrolytic process while secondary techniques include the electrothermic and Waelz kiln processes as well as a range of other metallurgical, hydrometallurgical, and pyrometallurgical processes. Worldwide primary zinc production also employs a pyrometallurgical process using the Imperial Smelting Furnace process; however, this process is not used in the United States (Sjardin 2003). Of the primary and secondary processes used in the United States, only the electrothermic and Waelz kiln secondary processes result in non-energy CO₂ emissions (Viklund-White 2000).

During one secondary technique, the electrothermic process, roasted zinc concentrate and secondary zinc products enter a sinter feed where they are burned to remove impurities before entering an electric retort furnace. Metallurgical coke added to the electric retort furnace reduces the zinc oxides and produces vaporized zinc, which is then captured in a vacuum condenser.

In the other secondary technique or Waelz kiln process, EAF dust, which is captured during the recycling of galvanized steel, enters a kiln along with a reducing agent—often metallurgical coke. When kiln temperatures reach approximately 1100–1200°C, zinc fumes are produced, which are combusted with air entering the kiln. This combustion forms zinc oxide, which is collected in a baghouse or electrostatic precipitator, and is then leached to remove chloride and fluoride. Through this process, approximately 0.33 metric ton of zinc is produced for every metric ton of EAF dust treated (Viklund-White 2000).

In 2010, U.S. primary and secondary refined zinc production were estimated to total 249,000 metric tons (USGS 2012), which was larger than 2009 levels, likely due to the general improvement in the U.S. economy in 2010 (see Table 4-75). This was despite an explosion at one of the biggest secondary refined zinc facilities in the United States (Horsehead Corporation's Monaca facility), which resulted in a temporary shutdown at the facility (Horsehead Corp.

2010b).

Emissions of CO₂ from zinc production in 2010 were estimated to be 1.2 Tg CO₂ Eq. (1,168 Gg) (see Table 4-80). All 2010 CO₂ emissions resulted from secondary zinc production processes. Emissions from zinc production in the U.S. have increased overall since 1990 due to a gradual shift from non-emissive primary production to emissive secondary production. In 2010, emissions were estimated to be 85 percent higher than they were in 1990.

Table 4-79: Zinc Production (Metric Tons)

Year	Primary	Secondary
1990	262,704	95,708
2005	191,120	156,000
2006	113,000	156,000
2007	121,000	157,000
2008	125,000	161,000
2009	94,000	109,000
2010	120,000	129,000

Table 4-76: CO₂ Emissions from Zinc Production (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	0.6	632
2005	1.0	1,030
2006	1.0	1,030
2007	1.0	1,025
2008	1.2	1,159
2009	0.9	943
2010	1.2	1,168

Methodology

Non-energy CO₂ emissions from zinc production result from the electrothermic and Waelz kiln secondary production processes, which both use metallurgical coke or other C-based materials as reductants. The methods used to estimate emissions from these processes are based on Tier 1 methods from the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006). The Tier 1 emission factors provided by IPCC (2006) for Waelz kiln-based secondary production were derived from coke consumption factors and other data presented in Viklund-White (2000). These coke consumption factors as well as other inputs used to develop the Waelz kiln emission factors are shown below. IPCC (2006) does not provide an emission factor for electrothermic processes due to limited information; therefore, the Waelz kiln-specific emission factors were applied to zinc produced from electrothermic processes.

For Waelz kiln-based production, IPCC (2006) recommends the use of emission factors based on EAF dust consumption if possible rather than the amount of zinc produced since the amount of reduction materials used is more directly dependent on the amount of EAF dust consumed. Since only a portion of emissive zinc production facilities consume EAF dust, the emission factor based on zinc production is applied to the non-EAF dust consuming facilities while the emission factor based on EAF dust consumption is applied to EAF dust consuming facilities.

The Waelz kiln emission factor based on the amount of zinc produced was developed based on the amount of metallurgical coke consumed for non-energy purposes per ton of zinc produced, 1.19 metric tons coke/metric ton zinc produced (Viklund-White 2000), and the following equation:

$$EF_{\text{Waelz Kiln}} = \frac{1.19 \text{ metric tons coke}}{\text{metric tons zinc}} \times \frac{0.85 \text{ metric tons C}}{\text{metric tons coke}} \times \frac{3.67 \text{ metric tons CO}_2}{\text{metric tons C}} = \frac{3.70 \text{ metric tons CO}_2}{\text{metric tons zinc}}$$

The Waelz kiln emission factor based on the amount of EAF dust consumed was developed based on the amount of metallurgical coke consumed per ton of EAF dust consumed, 0.4 metric tons coke/metric ton EAF dust consumed (Viklund-White 2000), and the following equation:

$$EF_{\text{EAF Dust}} = \frac{0.4 \text{ metric tons coke}}{\text{metric tons EAF dust}} \times \frac{0.85 \text{ metric tons C}}{\text{metric tons coke}} \times \frac{3.67 \text{ metric tons CO}_2}{\text{metric tons C}} = \frac{1.24 \text{ metric tons CO}_2}{\text{metric tons EAF Dust}}$$

The only companies in the United States that use emissive technology to produce secondary zinc products are Horsehead, PIZO, and Steel Dust Recycling. For Horsehead, EAF dust is recycled in Waelz kilns at their Beaumont, TX; Calumet, IL; Palmerton, PA; Rockwood, TN; and Barnwell, SC facilities. These Waelz kiln facilities produce intermediate zinc products (crude zinc oxide or calcine), most of which is transported to their Monaca, PA facility where the products are smelted into refined zinc using electrothermic technology. Some of Horsehead's intermediate zinc products that are not smelted at Monaca are instead exported to other countries around the world (Horsehead Corp. 2010a). PIZO and Steel Dust Recycling recycle EAF dust into intermediate zinc products using Waelz kilns, and then sell the intermediate products to companies who smelt it into refined products.

The total amount of EAF dust consumed by Horsehead at their Waelz kilns was available from Horsehead financial reports for years 2006 through 2010 (Horsehead Corp. 2008, 2010a, and 2011). Consumption levels for 1990 through 2005 were extrapolated using the percentage change in annual refined zinc production at secondary smelters in the United States as provided by USGS Minerals Yearbook: Zinc (USGS 1994 through 2011). The EAF dust consumption values for each year were then multiplied by the 1.24 metric tons CO₂/metric ton EAF dust consumed emission factor to develop CO₂ emission estimates for Horsehead's Waelz kiln facilities.

The amount of EAF dust consumed by PIZO's facility in 2009 and 2010 and Steel Dust Recycling's facility for 2008, 2009, and 2010 (the only years these facilities have been in operation) was not publically available. Therefore, these consumption values were estimated by calculating the 2008, 2009, and 2010 capacity utilization of Horsehead's Waelz kilns and multiplying this utilization ratio by the capacities of the PIZO and Steel Dust Recycling facilities, which were available from the companies (PIZO 2011 and Steel Dust Recycling LLC 2011). The 1.24 metric tons CO₂/metric ton EAF dust consumed emission factor was then applied to PIZO's and Steel Dust Recycling's estimated EAF dust consumption to develop CO₂ emission estimates for those Waelz kiln facilities.

Refined zinc production levels for Horsehead's Monaca, PA facility (utilizing electrothermic technology) were available from the company for years 2005 through 2010 (Horsehead Corp. 2008 and 2011). Production levels for 1990 through 2004 were extrapolated using the percentage changes in annual refined zinc production at secondary smelters in the United States as provided by USGS Minerals Yearbook: Zinc (USGS 1994 through 2011). The 3.70 metric tons CO₂/metric ton zinc emission factor was then applied to the Monaca facility's production levels to estimate CO₂ emissions for the facility. The Waelz kiln production emission factor was applied in this case rather than the EAF dust consumption emission factor since Horsehead's Monaca facility did not consume EAF dust.

Uncertainty and Time-Series Consistency

The uncertainties contained in these estimates are two-fold, relating to activity data and emission factors used.

First, there is uncertainty associated with the amount of EAF dust consumed in the United States to produce secondary zinc using emission-intensive Waelz kilns. The estimate for the total amount of EAF dust consumed in Waelz kilns is based on (1) an EAF dust consumption value reported annually by Horsehead Corporation as part of its financial reporting to the Securities and Exchange Commission (SEC), and (2) an estimate of the amount of EAF dust consumed at a Waelz kiln facility operated in Alabama by Steel Dust Recycling LLC. Since actual EAF dust consumption information is not available for the Steel Dust Recycling LLC facility, the amount is estimated by multiplying the EAF dust recycling capacity of the facility (available from the company's Web site) by the capacity utilization factor for Horsehead Corporation (which is available from Horsehead's financial reports). Therefore, there is uncertainty associated with the assumption that the capacity utilization of Steel Dust Recycling LLC's Waelz kiln facility is equal to the capacity utilization of Horsehead's Waelz kiln facility.

Second, there are uncertainties associated with the emission factors used to estimate CO₂ emissions from secondary zinc production processes. The Waelz kiln emission factors are based on materials balances for metallurgical coke and EAF dust consumed as provided by Viklund-White (2000). Therefore, the accuracy of these emission factors depend upon the accuracy of these materials balances. Data limitations prevented the development of emission factors for the electrothermic process. Therefore, emission factors for the Waelz kiln process were applied to both electrothermic and Waelz kiln production processes. The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-80. Zinc production CO₂ emissions were estimated to be between 1.0 and 1.4 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 17 percent below and 19 percent above the emission estimate of 1.2 Tg CO₂ Eq.

Table 4-80: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Zinc Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^a			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Zinc Production	CO ₂	1.2	1.0	1.4	-17%	+19%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Zinc Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all Inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹⁴⁴

Recalculations Discussion

In 2009, PIZO Technologies LLC commissioned an EAF dust consuming secondary production facility. The 2009 EAF dust consumption from this facility was not captured in the previous inventory. In addition, the EAF dust consumption data provided from Horsehead Corp for years 2006 through 2009 were incorrectly considered to be in metric tons in the previous Inventory when the data were actually provided in short tons (this also impacted 1990 through 2005 EAF dust consumption data that are estimated based on the 2006 estimate). Both of these issues have been corrected and decreased 1990 through 2009 emissions from zinc production by an average of 5.2 percent per year.

4.19. Lead Production (IPCC Source Category 2C5)

Lead production in the United States consists of both primary and secondary processes—both of which emit CO₂ (Sjardin 2003). Primary lead production, in the form of direct smelting, occurs at a just a single plant in Missouri. Secondary production largely involves the recycling of lead acid batteries at approximately 20 separate smelters in the United States. Fourteen of those secondary smelters have annual capacities of 15,000 tons or more and were collectively responsible for 99 percent of secondary lead production in 2011 (USGS 2012a). Secondary lead production has increased in the United States over the past decade while primary lead production has decreased. In 2009, secondary lead production accounted for approximately 92 percent of total lead production (USGS 2012b).

Primary production of lead through the direct smelting of lead concentrate produces CO₂ emissions as the lead concentrates are reduced in a furnace using metallurgical coke (Sjardin 2003). U.S. primary lead production

¹⁴⁴ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

increased by 24 percent from 2009 to 2010, and has decreased by 72 percent since 1990 (USGS 1995, 2012b).

Similar to primary lead production, CO₂ emissions from secondary production result when a reducing agent, usually metallurgical coke, is added to the smelter to aid in the reduction process. Carbon dioxide emissions from secondary production also occur through the treatment of secondary raw materials (Sjardin 2003). U.S. secondary lead production increased from 2009 to 2010 by 3 percent, and has increased by 24 percent since 1990 (USGS 1995 and 2012b).

In 2010, U.S. primary and secondary lead production totaled 1,255,000 metric tons (USGS 2012b). The resulting emissions of CO₂ from 2010 production were estimated to be 0.5 Tg CO₂ Eq. (542 Gg) (see Table 4-81). The majority of 2010 lead production is from secondary processes, which accounted for 95 percent of total 2010 CO₂ emissions. At last reporting, the United States was the third largest mine producer of lead in the world, behind China and Australia, accounting for 9 percent of world production in 2010 (USGS 2012b).

Table 4-81: CO₂ Emissions from Lead Production (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	0.5	516
2005	0.6	553
2006	0.6	560
2007	0.6	562
2008	0.5	547
2009	0.5	525
2010	0.5	542

After a steady increase in total emissions from 1995 to 2000, total emissions have gradually decreased since 2000 but were still 5 percent greater in 2010 than in 1990. Although primary production has decreased significantly (75 percent since 1990), secondary production has increased by about 20 percent over the same time period. Since secondary production is more emissions-intensive, the increase in secondary production since 1990 has resulted in a net increase in emissions despite the sharp decrease in primary production (USGS 1994 and 2012b).

Methodology

Non-energy CO₂ emissions from lead production result from primary and secondary production processes that use metallurgical coke or other C-based materials as reductants. The methods used to estimate emissions for lead production are based on Tier 1 methods from the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006). For primary lead production using direct smelting, Sjardin (2003) and the IPCC (2006) provide an emission factor of 0.25 metric tons CO₂/metric ton lead. For secondary lead production, Sjardin (2003) and IPCC (2006) provide an emission factor of 0.25 metric tons CO₂/metric ton lead for direct smelting as well as an emission factor of 0.2 metric tons CO₂/metric ton lead produced for the treatment of secondary raw materials (i.e., pretreatment of lead acid batteries). The direct smelting factor (0.25) and the sum of the direct smelting and pretreatment emission factors (0.45) are multiplied by total U.S. primary and secondary lead production, respectively, to estimate CO₂ emissions.

The 1990 through 2010 activity data for primary and secondary lead production (see Table 4-82) were obtained through the USGS Mineral Yearbook: Lead (USGS 1994 through 2012b).

Table 4-82: Lead Production (Metric Tons)

Year	Primary	Secondary
1990	404,000	922,000
2005	143,000	1,150,000
2006	153,000	1,160,000
2007	123,000	1,180,000

2008	135,000	1,140,000
2009	103,000	1,110,000
2010	115,000	1,140,000

Uncertainty and Time-Series Consistency

Uncertainty associated with lead production relates to the emission factors and activity data used. The direct smelting emission factor used in primary production is taken from Sjardin (2003) who averages the values provided by three other studies (Dutrizac et al. 2000, Morris et al. 1983, Ullman 1997). For secondary production, Sjardin (2003) adds a CO₂ emission factor associated with battery treatment. The applicability of these emission factors to plants in the United States is uncertain. There is also a smaller level of uncertainty associated with the accuracy of primary and secondary production data provided by the USGS.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-83. Lead production CO₂ emissions were estimated to be between 0.5 and 0.6 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 15 percent below and 15 percent above the emission estimate of 0.5 Tg CO₂ Eq.

Table 4-83: Tier 2 Quantitative Uncertainty Estimates for CO₂ Emissions from Lead Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)		Uncertainty Range Relative to Emission Estimate ^a (Tg CO ₂ Eq.)		Uncertainty Range Relative to Emission Estimate ^a (%)	
		Lower Bound	Upper Bound	Lower Bound	Upper Bound	Lower Bound	Upper Bound
Lead Production	CO ₂	0.5	0.5	0.6		-15%	+15%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Lead Production source category. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all inventory years as required for this inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.¹⁴⁵

Recalculations Discussion

Activity data for the time series was revised for the current Inventory. Specifically, secondary production data for 2008 were revised to reflect updated USGS data, which resulted in a lowering of the emission estimate for 2008 by less than one percent. Details on the emission trends through time are described in more detail in the Methodology section, above.

4.20. HCFC-22 Production (IPCC Source Category 2E1)

Trifluoromethane (HFC-23 or CHF₃) is generated as a byproduct during the manufacture of chlorodifluoromethane (HCFC-22), which is primarily employed in refrigeration and air conditioning systems and as a chemical feedstock for manufacturing synthetic polymers. Between 1990 and 2000, U.S. production of HCFC-22 increased significantly as HCFC-22 replaced chlorofluorocarbons (CFCs) in many applications. Between 2000 and 2007, U.S.

¹⁴⁵ See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

production fluctuated but generally remained above 1990 levels. In 2008 and 2009, U.S. production declined markedly before increasing slightly in 2010. Because HCFC-22 depletes stratospheric ozone, its production for non-feedstock uses is scheduled to be phased out by 2020 under the U.S. Clean Air Act.¹⁴⁶ Feedstock production, however, is permitted to continue indefinitely.

HCFC-22 is produced by the reaction of chloroform (CHCl_3) and hydrogen fluoride (HF) in the presence of a catalyst, SbCl_5 . The reaction of the catalyst and HF produces SbCl_xF_y , (where $x + y = 5$), which reacts with chlorinated hydrocarbons to replace chlorine atoms with fluorine. The HF and chloroform are introduced by submerged piping into a continuous-flow reactor that contains the catalyst in a hydrocarbon mixture of chloroform and partially fluorinated intermediates. The vapors leaving the reactor contain HCFC-21 (CHCl_2F), HCFC-22 (CHClF_2), HFC-23 (CHF_3), HCl, chloroform, and HF. The under-fluorinated intermediates (HCFC-21) and chloroform are then condensed and returned to the reactor, along with residual catalyst, to undergo further fluorination. The final vapors leaving the condenser are primarily HCFC-22, HFC-23, HCl and residual HF. The HCl is recovered as a useful byproduct, and the HF is removed. Once separated from HCFC-22, the HFC-23 may be released to the atmosphere, recaptured for use in a limited number of applications, or destroyed.

Three facilities produced HCFC-22 in the U.S. in 2010. Emissions of HFC-23 in 2010 were estimated to be 8.1 Tg CO_2 Eq. (0.7 Gg) (see Table 4-84). This quantity represents a 50 percent increase from 2009 emissions but a 78 percent decline from 1990 emissions. The increase from 2009 emissions was caused by a 10 percent increase in HCFC-22 production and a 36 percent increase in the HFC-23 emission rate. The decline from 1990 emissions is due to a 27 percent decrease in HCFC-22 production and a 69 percent decrease in the HFC-23 emission rate since 1990. The decrease in the emission rate is primarily attributable to five factors: (a) five plants that did not capture and destroy the HFC-23 generated have ceased production of HCFC-22 since 1990, (b) one plant that captures and destroys the HFC-23 generated began to produce HCFC-22, (c) one plant implemented and documented a process change that reduced the amount of HFC-23 generated, and (d) the same plant began recovering HFC-23, primarily for destruction and secondarily for sale, and (e) another plant began destroying HFC-23.

Table 4-84: HFC-23 Emissions from HCFC-22 Production (Tg CO_2 Eq. and Gg)

Year	Tg CO_2 Eq.	Gg
1990	36.4	3
2005	15.8	1
2006	13.8	1
2007	17.0	1
2008	13.6	1
2009	5.4	0.4
2010	8.1	1

Methodology

To estimate HFC-23 emissions for five of the eight HCFC-22 plants that have operated in the United States since 1990, methods comparable to the Tier 3 methods in the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC 2006) were used. For the other three plants, the last of which closed in 1993, methods comparable to the Tier 1 method in the 2006 IPCC Guidelines were used. Emissions from these three plants have been calculated using the recommended emission factor for unoptimized plants operating before 1995 (0.04 kg HCFC-23/kg HCFC-22 produced).

The five plants that have operated since 1994 measured concentrations of HFC-23 to estimate their emissions of HFC-23. Plants using thermal oxidation to abate their HFC-23 emissions monitor the performance of their oxidizers to verify that the HFC-23 is almost completely destroyed. Plants that release (or historically have released) some of their byproduct HFC-23 periodically measure HFC-23 concentrations in the output stream using gas chromatography. This information is combined with information on quantities of products (e.g., HCFC-22) to

¹⁴⁶ As construed, interpreted, and applied in the terms and conditions of the *Montreal Protocol on Substances that Deplete the Ozone Layer*. [42 U.S.C. §7671m(b), CAA §614]

estimate HFC-23 emissions.

To estimate 1990 through 2009 emissions, EPA relied on reports from an industry association that aggregated HCFC-22 production and HFC-23 emissions from all U.S. HCFC-22 producers and reported them to EPA (ARAP 1997, 1999, 2000, 2001, 2002, 2003, 2004, 2005, 2006, 2007, 2008, 2009, 2010). To estimate 2010 emissions, EPA analyzed facility-level data (including both HCFC-22 production and HFC-23 emissions) reported through the Greenhouse Gas Reporting Program (ICF, 2012). In 1997 and 2008, EPA (through a contractor) performed comprehensive reviews of plant-level estimates of HFC-23 emissions and HCFC-22 production (RTI 1997; RTI 2008). The 1997 and 2008 reviews enabled EPA to review, update, and where necessary, correct U.S. totals, and also to perform plant-level uncertainty analyses (Monte-Carlo simulations) for 1990, 1995, 2000, 2005, and 2006. Estimates of annual U.S. HCFC-22 production are presented in Table 4-85.

Table 4-85: HCFC-22 Production (Gg)

Year	Gg
1990	139
2005	156
2006	154
2007	162
2008	126
2009	91
2010	101

Uncertainty and Time Series Consistency

The uncertainty analysis presented in this section was based on a plant-level Monte Carlo Stochastic Simulation for 2006. The Monte Carlo analysis used estimates of the uncertainties in the individual variables in each plant's estimating procedure. This analysis was based on the generation of 10,000 random samples of model inputs from the probability density functions for each input. A normal probability density function was assumed for all measurements and biases except the equipment leak estimates for one plant; a log-normal probability density function was used for this plant's equipment leak estimates. The simulation for 2006 yielded a 95-percent confidence interval for U.S. emissions of 6.8 percent below to 9.6 percent above the reported total.

The relative errors yielded by the Monte Carlo Stochastic Simulation for 2006 were applied to the U.S. emission estimate for 2010. The resulting estimates of absolute uncertainty are likely to be reasonably accurate because (1) the methods used by the three plants to estimate their emissions are not believed to have changed significantly since 2006, and (2) although the distribution of emissions among the plants may have changed between 2006 and 2010 (because both HCFC-22 production and the HFC-23 emission rate declined significantly), the two plants that contribute significantly to emissions were estimated to have similar relative uncertainties in their 2006 (as well as 2005) emission estimates. Thus, changes in the relative contributions of these two plants to total emissions are not likely to have a large impact on the uncertainty of the national emission estimate.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-86. HFC-23 emissions from HCFC-22 production were estimated to be between 7.5 and 8.9 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 7 percent below and 10 percent above the emission estimate of 8.1 Tg CO₂ Eq.

Table 4-86: Quantitative Uncertainty Estimates for HFC-23 Emissions from HCFC-22 Production (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^a			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
HCFC-22 Production	HFC-23	8.1	7.5	8.9	-7%	+10%

^a Range of emissions reflects a 95 percent confidence interval.

Details on the emission trends through time are described in more detail in the Methodology section, above.

4.21. Substitution of Ozone Depleting Substances (IPCC Source Category 2F)

Hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) are used as alternatives to several classes of ozone-depleting substances (ODSs) that are being phased out under the terms of the *Montreal Protocol* and the Clean Air Act Amendments of 1990.¹⁴⁷ Ozone depleting substances—chlorofluorocarbons (CFCs), halons, carbon tetrachloride, methyl chloroform, and hydrochlorofluorocarbons (HCFCs)—are used in a variety of industrial applications including refrigeration and air conditioning equipment, solvent cleaning, foam production, sterilization, fire extinguishing, and aerosols. Although HFCs and PFCs are not harmful to the stratospheric ozone layer, they are potent greenhouse gases. Emission estimates for HFCs and PFCs used as substitutes for ODSs are provided in Table 4-87 and Table 4-88.

Table 4-87: Emissions of HFCs and PFCs from ODS Substitutes (Tg CO₂ Eq.)

Gas	1990	2005	2006	2007	2008	2009	2010
HFC-23	+	+	+	+	+	+	+
HFC-32	+	0.3	0.6	1.0	1.3	1.7	2.5
HFC-125	+	8.5	10.0	12.0	14.3	17.3	22.2
HFC-134a	+	74.9	74.8	72.2	69.3	66.7	66.8
HFC-143a	+	8.7	9.5	10.3	11.1	12.6	14.7
HFC-236fa	+	0.8	0.8	0.9	0.9	0.9	0.9
CF ₄	+	+	+	+	+	+	+
Others*	0.3	5.6	6.0	6.3	6.7	7.0	7.4
Total	0.3	99.0	101.9	102.7	103.6	106.3	114.6

+ Does not exceed 0.05 Tg CO₂ Eq.

* Others include HFC-152a, HFC-227ea, HFC-245fa, HFC-4310mee, C₄F₁₀, and PFC/PFPEs, the latter being a proxy for a diverse collection of PFCs and perfluoropolyethers (PFPEs) employed for solvent applications. For estimating purposes, the GWP value used for PFC/PFPEs was based upon C₆F₁₄.

Note: Totals may not sum due to independent rounding.

Table 4-88: Emissions of HFCs and PFCs from ODS Substitution (Mg)

Gas	1990	2005	2006	2007	2008	2009	2010
HFC-23	+	1	1	1	2	2	2
HFC-32	+	505	970	1,489	2,025	2,609	3,845
HFC-125	+	3,053	3,584	4,297	5,119	6,175	7,920
HFC-134a	+	57,637	57,572	55,517	53,274	51,333	51,423
HFC-143a	+	2,290	2,511	2,718	2,911	3,325	3,861
HFC-236fa	+	125	131	136	141	144	146
CF ₄	+	2	2	2	2	2	3
Others*	M	M	M	M	M	M	M

M (Mixture of Gases)

+ Does not exceed 0.5 Mg

* Others include HFC-152a, HFC-227ea, HFC-245fa, HFC-4310mee, C₄F₁₀, and PFC/PFPEs, the latter being a proxy for a diverse collection of PFCs and perfluoropolyethers (PFPEs) employed for solvent applications.

In 1990 and 1991, the only significant emissions of HFCs and PFCs as substitutes to ODSs were relatively small amounts of HFC-152a—used as an aerosol propellant and also a component of the refrigerant blend R-500 used in chillers—and HFC-134a in refrigeration end-uses. Beginning in 1992, HFC-134a was used in growing amounts as a refrigerant in motor vehicle air-conditioners and in refrigerant blends such as R-404A.¹⁴⁸ In 1993, the use of HFCs

¹⁴⁷ [42 U.S.C § 7671, CAA § 601]

¹⁴⁸ R-404A contains HFC-125, HFC-143a, and HFC-134a.

in foam production began, and in 1994 ODS substitutes for halons entered widespread use in the United States as halon production was phased-out. In 1995, these compounds also found applications as solvents.

The use and subsequent emissions of HFCs and PFCs as ODS substitutes has been increasing from small amounts in 1990 to 114.6 Tg CO₂ Eq. in 2010. This increase was in large part the result of efforts to phase out CFCs and other ODSs in the United States. In the short term, this trend is expected to continue, and will likely continue over the next decade as HCFCs, which are interim substitutes in many applications, are themselves phased-out under the provisions of the Copenhagen Amendments to the *Montreal Protocol*. Improvements in the technologies associated with the use of these gases and the introduction of alternative gases and technologies, however, may help to offset this anticipated increase in emissions.

Table 4-89 presents emissions of HFCs and PFCs as ODS substitutes by end-use sector for 1990 through 2010. The end-use sectors that contributed the most toward emissions of HFCs and PFCs as ODS substitutes in 2010 include refrigeration and air-conditioning (97.6 Tg CO₂ Eq., or approximately 85 percent), aerosols (9.3 Tg CO₂ Eq., or approximately 8 percent), and foams (5.4 Tg CO₂ Eq., or approximately 5 percent). Within the refrigeration and air-conditioning end-use sector, motor vehicle air-conditioning was the highest emitting end-use (44.1 Tg CO₂ Eq.), followed by refrigerated retail food and transport. Each of the end-use sectors is described in more detail below.

Table 4-89: Emissions of HFCs and PFCs from ODS Substitutes (Tg CO₂ Eq.) by Sector

Gas	1990		2005	2006	2007	2008	2009	2010
Refrigeration/Air Conditioning	+		87.9	90.1	90.3	90.4	91.3	97.6
Aerosols	0.3		7.3	7.7	8.2	8.6	9.1	9.3
Foams	+		1.9	2.1	2.3	2.5	3.9	5.4
Solvents	+		1.3	1.3	1.3	1.3	1.3	1.3
Fire Protection	+		0.5	0.6	0.7	0.7	0.8	0.9
Total	0.3		99.0	101.9	102.7	103.6	106.3	114.6

Refrigeration/Air Conditioning

The refrigeration and air-conditioning sector includes a wide variety of equipment types that have historically used CFCs or HCFCs. End-uses within this sector include motor vehicle air-conditioning, retail food refrigeration, refrigerated transport (e.g., ship holds, truck trailers, railway freight cars), household refrigeration, residential and small commercial air-conditioning and heat pumps, chillers (large comfort cooling), cold storage facilities, and industrial process refrigeration (e.g., systems used in food processing, chemical, petrochemical, pharmaceutical, oil and gas, and metallurgical industries). As the ODS phaseout is taking effect, most equipment is being or will eventually be retrofitted or replaced to use HFC-based substitutes. Common HFCs in use today in refrigeration/air-conditioning equipment are HFC-134a, R-410A¹⁴⁹, R-404A, and R-507A.¹⁵⁰ These HFCs are emitted to the atmosphere during equipment manufacture and operation (as a result of component failure, leaks, and purges), as well as at servicing and disposal events.

Aerosols

Aerosol propellants are used in metered dose inhalers (MDIs) and a variety of personal care products and technical/specialty products (e.g., duster sprays and safety horns). Many pharmaceutical companies that produce MDIs—a type of inhaled therapy used to treat asthma and chronic obstructive pulmonary disease—have replaced the use of CFCs with HFC-propellant alternatives. The earliest ozone-friendly MDIs were produced with HFC-134a, but the industry has started to use HFC-227ea as well. Conversely, since the use of CFC propellants was banned in 1978, most non-medical consumer aerosol products have not transitioned to HFCs, but to “not-in-kind” technologies, such as solid roll-on deodorants and finger-pump sprays. The transition away from ODS in specialty aerosol products has also led to the introduction of non-fluorocarbon alternatives (e.g., hydrocarbon propellants) in certain applications, in addition to HFC-134a or HFC-152a. These propellants are released into the atmosphere as the aerosol products are used.

¹⁴⁹ R-410A contains HFC-32 and HFC-125.

¹⁵⁰ R-507A, also called R-507, contains HFC-125 and HFC-143a.

Foams

CFCs and HCFCs have traditionally been used as foam blowing agents to produce polyurethane (PU), polystyrene, polyolefin, and phenolic foams, which are used in a wide variety of products and applications. Since the *Montreal Protocol*, flexible PU foams as well as other types of foam, such as polystyrene sheet, polyolefin, and phenolic foam, have transitioned almost completely away from fluorocompounds, into alternatives such as CO₂, methylene chloride, and hydrocarbons. The majority of rigid PU foams have transitioned to HFCs—primarily HFC-134a and HFC-245fa. Today, these HFCs are used to produce polyurethane appliance, PU commercial refrigeration, PU spray, and PU panel foams—used in refrigerators, vending machines, roofing, wall insulation, garage doors, and cold storage applications. In addition, HFC-152a, HFC-134a and CO₂ are used to produce polystyrene sheet/board foam, which is used in food packaging and building insulation. Emissions of blowing agents occur when the foam is manufactured as well as during the foam lifetime and at foam disposal, depending on the particular foam type.

Solvents

CFCs, methyl chloroform (1,1,1-trichloroethane or TCA), and to a lesser extent carbon tetrachloride (CCl₄) were historically used as solvents in a wide range of cleaning applications, including precision, electronics, and metal cleaning. Since their phaseout, metal cleaning end-use applications have primarily transitioned to non-fluorocarbon solvents and not-in-kind processes. The precision and electronics cleaning end-uses have transitioned in part to high-GWP gases, due to their high reliability, excellent compatibility, good stability, low toxicity, and selective solvency. These applications rely on HFC-4310mee, HFC-365mfc, HFC-245fa, and to a lesser extent, PFCs. Electronics cleaning involves removing flux residue that remains after a soldering operation for printed circuit boards and other contamination-sensitive electronics applications. Precision cleaning may apply to either electronic components or to metal surfaces, and is characterized by products, such as disk drives, gyroscopes, and optical components, that require a high level of cleanliness and generally have complex shapes, small clearances, and other cleaning challenges. The use of solvents yields fugitive emissions of these HFCs and PFCs.

Fire Protection

Fire protection applications include portable fire extinguishers (“streaming” applications) that originally used halon 1211, and total flooding applications that originally used halon 1301, as well as some halon 2402. Since the production and sale of halons were banned in the United States in 1994, the halon replacement agent of choice in the streaming sector has been dry chemical, although HFC-236fa is also used to a limited extent. In the total flooding sector, HFC-227ea has emerged as the primary replacement for halon 1301 in applications that require clean agents. Other HFCs, such as HFC-23 and HFC-125, are used in smaller amounts. The majority of HFC-227ea in total flooding systems is used to protect essential electronics, as well as in civil aviation, military mobile weapons systems, oil/gas/other process industries, and merchant shipping. As fire protection equipment is tested or deployed, emissions of these HFCs occur.

Methodology

A detailed Vintaging Model of ODS-containing equipment and products was used to estimate the actual—versus potential—emissions of various ODS substitutes, including HFCs and PFCs. The name of the model refers to the fact that it tracks the use and emissions of various compounds for the annual “vintages” of new equipment that enter service in each end-use. The Vintaging Model predicts ODS and ODS substitute use in the United States based on modeled estimates of the quantity of equipment or products sold each year containing these chemicals and the amount of the chemical required to manufacture and/or maintain equipment and products over time. Emissions for each end-use were estimated by applying annual leak rates and release profiles, which account for the lag in emissions from equipment as they leak over time. By aggregating the data for nearly 60 different end-uses, the model produces estimates of annual use and emissions of each compound. Further information on the Vintaging Model is contained in Annex 3.8.

Uncertainty

Given that emissions of ODS substitutes occur from thousands of different kinds of equipment and from millions of point and mobile sources throughout the United States, emission estimates must be made using analytical tools such as the Vintaging Model or the methods outlined in IPCC (2006). Though the model is more comprehensive than the IPCC default methodology, significant uncertainties still exist with regard to the levels of equipment sales,

equipment characteristics, and end-use emissions profiles that were used to estimate annual emissions for the various compounds.

The Vintaging Model estimates emissions from 60 end-uses. The uncertainty analysis, however, quantifies the level of uncertainty associated with the aggregate emissions resulting from the top 21 end-uses, comprising over 95 percent of the total emissions, and 5 other end-uses. These 26 end-uses comprise 97 percent of the total emissions. In an effort to improve the uncertainty analysis, additional end-uses are added annually, with the intention that over time uncertainty for all emissions from the Vintaging Model will be fully characterized. Any end-uses included in previous years' uncertainty analysis were included in the current uncertainty analysis, whether or not those end-uses were included in the top 95 percent of emissions from ODS Substitutes.

In order to calculate uncertainty, functional forms were developed to simplify some of the complex "vintaging" aspects of some end-use sectors, especially with respect to refrigeration and air-conditioning, and to a lesser degree, fire extinguishing. These sectors calculate emissions based on the entire lifetime of equipment, not just equipment put into commission in the current year, thereby necessitating simplifying equations. The functional forms used variables that included growth rates, emission factors, transition from ODSs, change in charge size as a result of the transition, disposal quantities, disposal emission rates, and either stock for the current year or original ODS consumption. Uncertainty was estimated around each variable within the functional forms based on expert judgment, and a Monte Carlo analysis was performed. The most significant sources of uncertainty for this source category include the emission factors for retail food equipment and refrigerated transport, as well as the percent of non-MDI aerosol propellant that is HFC-152a.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-90. Substitution of ozone depleting substances HFC and PFC emissions were estimated to be between 111.8 and 129.3 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 7 percent below to 8 percent above the emission estimate of 120.0 Tg CO₂ Eq.

Table 4-90: Tier 2 Quantitative Uncertainty Estimates for HFC and PFC Emissions from ODS Substitutes (Tg CO₂ Eq. and Percent)

Source	Gases	2010 Emission Estimate (Tg CO ₂ Eq.) ^a	Uncertainty Range Relative to Emission Estimate ^b			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Substitution of Ozone Depleting Substances	HFCs and PFCs	112.2	110.5	127.1	-1.5%	+13.3%

^a 2010 emission estimates and the uncertainty range presented in this table correspond to selected end-uses within the aerosols, foams, solvents, fire extinguishing agents, and refrigerants sectors, but not for other remaining categories. Therefore, because the uncertainty associated with emissions from "other" ODS substitutes was not estimated, they were excluded in the estimates reported in this table.

^b Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Recalculations Discussion

A review of the window units and residential unitary air conditioning end-uses led to minor revisions in the assumed transition scenarios. Overall, these changes to the Vintaging Model had negligible effects on estimates of greenhouse gas emissions across the time series. An update to the retail food refrigeration end-uses resulted in the replacement of the medium retail food end-use with small condensing units and large condensing units. In addition, updates were made to the charge sizes, leak rates, and equipment transitions for each end-use. These changes to the Vintaging Model had a significant impact on the estimates of greenhouse gas emissions for the retail food refrigeration sector.

4.22. Semiconductor Manufacture (IPCC Source Category 2F6)

The semiconductor industry uses multiple long-lived fluorinated gases in plasma etching and plasma enhanced chemical vapor deposition (PECVD) processes to produce semiconductor products. The gases most commonly employed are trifluoromethane (HFC-23 or CHF₃), perfluoromethane (CF₄), perfluoroethane (C₂F₆), nitrogen trifluoride (NF₃), and sulfur hexafluoride (SF₆), although other compounds such as perfluoropropane (C₃F₈) and perfluorocyclobutane (c-C₄F₈) are also used. The exact combination of compounds is specific to the process employed.

A single 300 mm silicon wafer that yields between 400 to 500 semiconductor products (devices or chips) may require as many as, or more than 100 distinct fluorinated-gas-using process steps, principally to deposit and pattern dielectric films. Plasma etching (or patterning) of dielectric films, such as silicon dioxide and silicon nitride, is performed to provide pathways for conducting material to connect individual circuit components in each device. The patterning process uses plasma-generated fluorine atoms, which chemically react with exposed dielectric film to selectively remove the desired portions of the film. The material removed as well as undissociated fluorinated gases flow into waste streams and, unless emission abatement systems are employed, into the atmosphere. PECVD chambers, used for depositing dielectric films, are cleaned periodically using fluorinated and other gases. During the cleaning cycle the gas is converted to fluorine atoms in plasma, which etches away residual material from chamber walls, electrodes, and chamber hardware. Undissociated fluorinated gases and other products pass from the chamber to waste streams and, unless abatement systems are employed, into the atmosphere. In addition to emissions of unreacted gases, some fluorinated compounds can also be transformed in the plasma processes into different fluorinated compounds which are then exhausted, unless abated, into the atmosphere. For example, when C₂F₆ is used in cleaning or etching, CF₄ is generated and emitted as a process by-product. Besides dielectric film etching and PECVD chamber cleaning, much smaller quantities of fluorinated gases are used to etch polysilicon films and refractory metal films like tungsten.

For 2010, total weighted emissions of all fluorinated greenhouse gases by the U.S. semiconductor industry were estimated to be 5.4 Tg CO₂ Eq. Combined emissions of all fluorinated greenhouse gases are presented in Table 4-91 and Table 4-92 below for years 1990 and the period 2005 to 2010. The rapid growth of this industry and the increasing complexity (growing number of layers)¹⁵¹ of semiconductor products led to an increase in emissions of 148 percent between 1990 and 1999, when emissions peaked at 7.2 Tg CO₂ Eq. The emissions growth rate began to slow after 1998, and emissions declined by 26 percent between 1999 and 2010. Together, industrial growth and adoption of emissions reduction technologies, including but not limited to abatement technologies, resulted in a net increase in emissions of 85 percent between 1990 and 2010.

Table 4-91: PFC, HFC, and SF₆ Emissions from Semiconductor Manufacture (Tg CO₂ Eq.)

Year	1990	2005	2006	2007	2008	2009	2010
CF ₄	0.7	1.1	1.2	1.3	1.4	1.5	1.6
C ₂ F ₆	1.5	2.0	2.2	2.3	2.4	2.5	2.4
C ₃ F ₈	0.0	0.0	0.0	0.0	0.1	0.0	0.0
C ₄ F ₈	0.0	0.1	0.1	0.1	0.1	0.0	0.0
HFC-23	0.2	0.2	0.3	0.3	0.3	0.3	0.3
SF ₆	0.5	1.0	1.0	0.8	0.9	1.0	0.9
NF ₃ *	0.0	0.4	0.7	0.5	0.6	0.5	0.5
Total	2.9	4.4	4.7	4.8	5.1	5.3	5.4

Note: Totals may not sum due to independent rounding.

* NF₃ emissions are presented for informational purposes, using the AR4 GWP of 17,200, and are not included in totals.

Table 4-92: PFC, HFC, and SF₆ Emissions from Semiconductor Manufacture (Mg)

Year	1990	2005	2006	2007	2008	2009	2010
------	------	------	------	------	------	------	------

¹⁵¹ Complexity is a term denoting the circuit required to connect the active circuit elements (transistors) on a chip. Increasing miniaturization, for the same chip size, leads to increasing transistor density, which, in turn, requires more complex interconnections between those transistors. This increasing complexity is manifested by increasing the levels (i.e., layers) of wiring, with each wiring layer requiring fluorinated gas usage for its manufacture.

CF ₄	115	168	181	198	216	226	245
C ₂ F ₆	160	216	240	249	261	271	264
C ₃ F ₈	0	5	5	6	13	5	5
C ₄ F ₈	0	13	13	7	7	4	4
HFC-23	15	18	22	23	25	28	29
SF ₆	22	40	40	34	36	40	40
NF ₃	3	26	40	30	33	29	31

Methodology

Emissions are based on Partner reported emissions data received through the EPA's PFC Reduction/Climate Partnership and the EPA's PFC Emissions Vintage Model (PEVM), a model which estimates industry emissions in the absence of emission control strategies (Burton and Beizaie 2001).¹⁵² The availability and applicability of Partner data differs across the 1990 through 2010 time series. Consequently, emissions from semiconductor manufacturing were estimated using four distinct methods, one each for the periods 1990 through 1994, 1995 through 1999, 2000 through 2006, and 2007 through 2010.

1990 through 1994

From 1990 through 1994, Partnership data was unavailable and emissions were modeled using the PEVM (Burton and Beizaie 2001).¹⁵³ 1990 to 1994 emissions are assumed to be uncontrolled, since reduction strategies such as chemical substitution and abatement were yet to be developed.

PEVM is based on the recognition that PFC emissions from semiconductor manufacturing vary with: (1) the number of layers that comprise different kinds of semiconductor devices, including both silicon wafer and metal interconnect layers, and (2) silicon consumption (i.e., the area of semiconductors produced) for each kind of device. The product of these two quantities, Total Manufactured Layer Area (TMLA), constitutes the activity data for semiconductor manufacturing. PEVM also incorporates an emission factor that expresses emissions per unit of layer-area. Emissions are estimated by multiplying TMLA by this emission factor.

PEVM incorporates information on the two attributes of semiconductor devices that affect the number of layers: (1) linewidth technology (the smallest manufactured feature size),¹⁵⁴ and (2) product type (discrete, memory or logic).¹⁵⁵ For each linewidth technology, a weighted average number of layers is estimated using VLSI product-specific worldwide silicon demand data in conjunction with complexity factors (i.e., the number of layers per Integrated Circuit (IC)) specific to product type (Burton and Beizaie 2001, ITRS 2007). PEVM derives historical consumption of silicon (i.e., square inches) by linewidth technology from published data on annual wafer starts and average wafer size (VLSI Research, Inc. 2010).

The emission factor in PEVM is the average of four historical emission factors, each derived by dividing the total

¹⁵² A Partner refers to a participant in the U.S. EPA PFC Reduction/Climate Partnership for the Semiconductor Industry. Through a Memorandum of Understanding (MoU) with the EPA, Partners voluntarily report their PFC emissions to the EPA by way of a third party, which aggregates the emissions.

¹⁵³ Various versions of the PEVM exist to reflect changing industrial practices. From 1990 to 1994 emissions estimates are from PEVM v1.0, completed in September 1998. The emission factor used to estimate 1990 to 1994 emissions is an average of the 1995 and 1996 emissions factors, which were derived from Partner reported data for those years.

¹⁵⁴ By decreasing features of Integrated Circuit components, more components can be manufactured per device, which increases its functionality. However, as those individual components shrink it requires more layers to interconnect them to achieve the functionality. For example, a microprocessor manufactured with the smallest feature sizes (65 nm) might contain as many as 1 billion transistors and require as many as 11 layers of component interconnects to achieve functionality while a device manufactured with 130 nm feature size might contain a few hundred million transistors and require 8 layers of component interconnects (ITRS 2007).

¹⁵⁵ Memory devices manufactured with the same feature sizes as microprocessors (a logic device) require approximately one-half the number of interconnect layers, whereas discrete devices require only a silicon base layer and no interconnect layers (ITRS 2007). Since discrete devices did not start using PFCs appreciably until 2004, they are only accounted for in the PEVM emissions estimates from 2004 onwards.

annual emissions reported by the Partners for each of the four years between 1996 and 1999 by the total TMLA estimated for the Partners in each of those years. Over this period, the emission factors varied relatively little (i.e., the relative standard deviation for the average was 5 percent). Since Partners are believed not to have applied significant emission reduction measures before 2000, the resulting average emission factor reflects uncontrolled emissions. The emission factor is used to estimate world uncontrolled emissions using publicly available data on world silicon consumption.

1995 through 1999

For 1995 through 1999, total U.S. emissions were extrapolated from the total annual emissions reported by the Partners (1995 through 1999). Partner-reported emissions are considered more representative (e.g., in terms of capacity utilization in a given year) than PEVM estimated emissions, and are used to generate total U.S. emissions when applicable. The emissions reported by the Partners were divided by the ratio of the total capacity of the plants operated by the Partners and the total capacity of all of the semiconductor plants in the United States; this ratio represents the share of capacity attributable to the Partnership. This method assumes that Partners and non-Partners have identical capacity utilizations and distributions of manufacturing technologies. Plant capacity data is contained in the World Fab Forecast (WFF) database and its predecessors, which is updated quarterly (Semiconductor Equipment and Materials Industry 2011).

2000 through 2006

The emission estimate for the years 2000 through 2006—the period during which Partners began the consequential application of PFC-reduction measures—was estimated using a combination of Partner reported emissions and PEVM modeled emissions. The emissions reported by Partners for each year were accepted as the quantity emitted from the share of the industry represented by those Partners. Remaining emissions, those from non-Partners, were estimated using PEVM and the method described above. This is because non-Partners are assumed not to have implemented any PFC-reduction measures, and PEVM models emissions without such measures. The portion of the U.S. total attributed to non-Partners is obtained by multiplying PEVM's total U.S. emissions figure by the non-Partner share of U. S. total silicon capacity for each year as described above.¹⁵⁶⁻¹⁵⁷ Annual updates to PEVM reflect published figures for actual silicon consumption from VLSI Research, Inc., revisions and additions to the world population of semiconductor manufacturing plants, and changes in IC fabrication practices within the semiconductor industry (see ITRS 2008 and Semiconductor Equipment and Materials Industry 2011).¹⁵⁸⁻¹⁵⁹⁻¹⁶⁰

¹⁵⁶ This approach assumes that the distribution of linewidth technologies is the same between Partners and non-Partners. As discussed in the description of the method used to estimate 2007 emissions, this is not always the case.

¹⁵⁷ Generally 5 percent or less of the fields needed to estimate TMLA shares are missing values in the World Fab Watch databases. In the 2007 World Fab Watch database used to generate the 2006 non-Partner TMLA capacity share, these missing values were replaced with the corresponding mean TMLA across fabs manufacturing similar classes of products. However, the impact of replacing missing values on the non-Partner TMLA capacity share was inconsequential.

¹⁵⁸ Special attention was given to the manufacturing capacity of plants that use wafers with 300 mm diameters because the actual capacity of these plants is ramped up to design capacity, typically over a 2–3 year period. To prevent overstating estimates of partner-capacity shares from plants using 300 mm wafers, *design* capacities contained in WFW were replaced with estimates of *actual installed* capacities for 2004 published by Citigroup Smith Barney (2005). Without this correction, the partner share of capacity would be overstated, by approximately 5 percent. For perspective, approximately 95 percent of all new capacity additions in 2004 used 300 mm wafers, and by year-end those plants, on average, could operate at approximately 70 percent of the design capacity. For 2005, actual installed capacities were estimated using an entry in the World Fab Watch database (April 2006 Edition) called “wafers/month, 8-inch equivalent,” which denoted the actual installed capacity instead of the fully-ramped capacity. For 2006, actual installed capacities of new fabs were estimated using an average monthly ramp rate of 1100 wafer starts per month (wspm) derived from various sources such as semiconductor fabtech, industry analysts, and articles in the trade press. The monthly ramp rate was applied from the first-quarter of silicon volume (QSV) to determine the average design capacity over the 2006 period.

¹⁵⁹ In 2006, the industry trend in co-ownership of manufacturing facilities continued. Several manufacturers, who are Partners, now operate fabs with other manufacturers, who in some cases are also Partners and in other cases are not Partners. Special attention was given to this occurrence when estimating the Partner and non-Partner shares of U.S. manufacturing capacity.

¹⁶⁰ Two versions of PEVM are used to model non-Partner emissions during this period. For the years 2000 to 2003 PEVM v3.2.0506.0507 was used to estimate non-Partner emissions. During this time, discrete devices did not use PFCs during

2007 through 2010

For the years 2007 through 2010, emissions were also estimated using a combination of Partner reported emissions and PEVM modeled emissions; however, two improvements were made to the estimation method employed for the previous years in the time series. First, the 2007 through 2010 emission estimates account for the fact that Partners and non-Partners employ different distributions of manufacturing technologies, with the Partners using manufacturing technologies with greater transistor densities and therefore greater numbers of layers.¹⁶¹ Second, the scope of the 2007 through 2010 estimates is expanded relative to the estimates for the years 2000 through 2006 to include emissions from Research and Development (R&D) fabs. This was feasible through the use of more detailed data published in the World Fab Forecast. PEVM databases are updated annually as described above. The published world average capacity utilization for 2007 through 2010 was used for production fabs while for R&D fabs a 20 percent figure was assumed (SIA 2009).

In addition, publicly available actual utilization data was used to account for differences in fab utilization for manufacturers of discrete and IC products for the emissions in 2010 for non-partners. PEVM estimates were adjusted using technology weighted capacity shares that reflect relative influence of different utilization.

Gas-Specific Emissions

Two different approaches were also used to estimate the distribution of emissions of specific fluorinated gases. Before 1999, when there was no consequential adoption of fluorinated-gas-reducing measures, a fixed distribution of fluorinated-gas use was assumed to apply to the entire U.S. industry. This distribution was based upon the average fluorinated-gas purchases made by semiconductor manufacturers during this period and the application of IPCC default emission factors for each gas (Burton and Beizaie 2001). For the 2000 through 2010 period, the 1990 through 1999 distribution was assumed to apply to the non-Partners. Partners, however, began reporting gas-specific emissions during this period. Thus, gas-specific emissions for 2000 through 2010 were estimated by adding the emissions reported by the Partners to those estimated for the non-Partners.

Data Sources

Partners estimate their emissions using a range of methods. For 2010, it is assumed that most Partners used a method at least as accurate as the IPCC's Tier 2a Methodology, recommended in the IPCC Guidelines for National Greenhouse Inventories (2006). Data used to develop emission estimates are attributed in part to estimates provided by the members of the Partnership, and in part from data obtained from PEVM estimates. Estimates of operating plant capacities and characteristics for Partners and non-Partners were derived from the Semiconductor Equipment and Materials Industry (SEMI) World Fab Forecast (formerly World Fab Watch) database (1996 through 2010) (e.g., Semiconductor Materials and Equipment Industry, 2011). Actual world capacity utilizations for 2010 were obtained from Semiconductor International Capacity Statistics (SICAS) (SIA, 2010). Estimates of silicon consumed by linewidth from 1990 through 2010 were derived from information from VLSI Research, Inc. (2010), and the number of layers per linewidth was obtained from International Technology Roadmap for Semiconductors: 2006 Update (Burton and Beizaie 2001, ITRS 2007, ITRS 2008).

Uncertainty and Time Series Consistency

A quantitative uncertainty analysis of this source category was performed using the IPCC-recommended Tier 2 uncertainty estimation methodology, the Monte Carlo Stochastic Simulation technique. The equation used to estimate uncertainty is:

$$\text{U.S. emissions} = \sum \text{Partnership gas-specific submittals} + [(\text{non-Partner share of World TMLA}) \times (\text{PEVM Emission})]$$

manufacturing and therefore only memory and logic devices were modeled in the PEVM v3.2.0506.0507. From 2004 onwards, discrete device fabrication started to use PFCs, hence PEVM v4.0.0701.0701, the first version of PEVM to account for PFC emissions from discrete devices, was used to estimate non-Partner emissions for this time period.

¹⁶¹ EPA considered applying this change to years before 2007, but found that it would be difficult due to the large amount of data (i.e., technology-specific global and non-Partner TMLA) that would have to be examined and manipulated for each year. This effort did not appear to be justified given the relatively small impact of the improvement on the total estimate for 2007 and the fact that the impact of the improvement would likely be lower for earlier years because the estimated share of emissions accounted for by non-Partners is growing as Partners continue to implement emission-reduction efforts.

Factor × World TMLA)]

The Monte Carlo analysis results presented below relied on estimates of uncertainty attributed to the four quantities on the right side of the equation. Estimates of uncertainty for the four quantities were in turn developed using the estimated uncertainties associated with the individual inputs to each quantity, error propagation analysis, Monte Carlo simulation, and expert judgment. The relative uncertainty associated with World TMLA estimate in 2010 is about ±10 percent, based on the uncertainty estimate obtained from discussions with VLSI, Inc. For the share of World layer-weighted silicon capacity accounted for by non-Partners, a relative uncertainty of ±8 percent was estimated based on a separate Monte Carlo simulation to account for the random occurrence of missing data in the World Fab Watch database. For the aggregate PFC emissions data supplied to the partnership, a relative uncertainty of ±50 percent was estimated for each gas-specific PFC emissions value reported by an individual Partner, and error propagation techniques were used to estimate uncertainty for total Partnership gas-specific submittals.¹⁶² A relative uncertainty of approximately ±10 percent was estimated for the PEVM emission factor, based on the standard deviation of the 1996 to 1999 emission factors.¹⁶³ All estimates of uncertainties are given at 95-percent confidence intervals.

In developing estimates of uncertainty, consideration was also given to the nature and magnitude of the potential bias that World activity data (i.e., World TMLA) might have in its estimates of the number of layers associated with devices manufactured at each technology node. The result of a brief analysis indicated that U.S. TMLA overstates the average number of layers across all product categories and all manufacturing technologies by 0.12 layers or 2.9 percent.¹⁶⁴ The same upward bias is assumed for World TMLA, and is represented in the uncertainty analysis by deducting the absolute bias value from the World activity estimate when it is incorporated into the Monte Carlo analysis.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-4-93. The emissions estimate for total U.S. PFC emissions from semiconductor manufacturing were estimated to be between 4.8 and 5.9 Tg CO₂ Eq. at a 95 percent confidence level. This range represents 10 percent below to 10 percent above the 2010 emission estimate of 5.4 Tg CO₂ Eq. This range and the associated percentages apply to the estimate of total emissions rather than those of individual gases. Uncertainties associated with individual gases will be somewhat higher than the aggregate, but were not explicitly modeled.

Table 4-4-93: Tier 2 Quantitative Uncertainty Estimates for HFC, PFC, and SF₆ Emissions from Semiconductor Manufacture (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate ^a (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^b			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound ^c	Upper Bound ^c	Lower Bound	Upper Bound
Semiconductor Manufacture	HFC, PFC, and SF ₆	5.4	4.8	5.9	-10%	10%

^a Because the uncertainty analysis covered all emissions (including NF₃), the emission estimate presented here does not match that shown in Table 4-91.

^b Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

^c Absolute lower and upper bounds were calculated using the corresponding lower and upper bounds in percentages.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2009. Details on the emission trends through time are described in more detail in the Methodology section, above.

¹⁶² Error propagation resulted in Partnership gas-specific uncertainties ranging from 17 to 27 percent.

¹⁶³ The average of 1996 to 1999 emission factor is used to derive the PEVM emission factor.

¹⁶⁴ This is based on an analysis of 2004 data.

Planned Improvements

One point of consideration for future national emissions estimates is the inclusion of PFC emissions from heat transfer fluid (HTF) loss to the atmosphere. Heat transfer fluids, of which some are liquid perfluorinated compounds, are used during testing of semiconductor devices and, increasingly, are used to manage heat during the manufacture of semiconductor devices. Evaporation of these fluids is a source of fluorinated emissions (EPA 2006).

4.23. Electrical Transmission and Distribution (IPCC Source Category 2F7)

The largest use of SF₆, both in the United States and internationally, is as an electrical insulator and interrupter in equipment that transmits and distributes electricity (RAND 2004). The gas has been employed by the electric power industry in the United States since the 1950s because of its dielectric strength and arc-quenching characteristics. It is used in gas-insulated substations, circuit breakers, and other switchgear. Sulfur hexafluoride has replaced flammable insulating oils in many applications and allows for more compact substations in dense urban areas.

Fugitive emissions of SF₆ can escape from gas-insulated substations and switchgear through seals, especially from older equipment. The gas can also be released during equipment manufacturing, installation, servicing, and disposal. Emissions of SF₆ from equipment manufacturing and from electrical transmission and distribution systems were estimated to be 11.8 Tg CO₂ Eq. (0.5 Gg) in 2010. This quantity represents a 56 percent decrease from the estimate for 1990 (see Table 4-94 and Table 4-95). Two trends contributed to this decrease: a sharp increase in the price of SF₆ during the 1990s and a growing awareness of the environmental impact of SF₆ emissions through programs such as EPA's SF₆ Emission Reduction Partnership for Electric Power Systems.

Table 4-94: SF₆ Emissions from Electric Power Systems and Electrical Equipment Manufacturers (Tg CO₂ Eq.)

Year	Electric Power Systems	Electrical Equipment Manufacturers	Total
1990	26.3	0.3	26.7
2005	13.1	0.8	13.9
2006	12.2	0.8	13.0
2007	11.5	0.7	12.2
2008	11.1	1.1	12.2
2009	11.3	0.6	11.8
2010	11.0	0.8	11.8

Note: Totals may not sum due to independent rounding.

Table 4-95: SF₆ Emissions from Electric Power Systems and Electrical Equipment Manufacturers (Gg)

Year	Emissions
1990	1.1
2005	0.6
2006	0.5
2007	0.5
2008	0.5
2009	0.5
2010	0.5

Methodology

The estimates of emissions from Electrical Transmission and Distribution are comprised of emissions from electric power systems and emissions from the manufacture of electrical equipment. The methodologies for estimating both sets of emissions are described below.

1999 through 2010 Emissions from Electric Power Systems

Emissions from electric power systems from 1999 to 2010 were estimated based on: (1) reporting from utilities

participating in EPA's SF₆ Emission Reduction Partnership for Electric Power Systems (Partners), which began in 1999; and, (2) the relationship between emissions and utilities' transmission miles as reported in the 2001, 2004, 2007, and 2010 Utility Data Institute (UDI) Directories of Electric Power Producers and Distributors (UDI 2001, 2004, 2007, 2010). (Transmission miles are defined as the miles of lines carrying voltages above 34.5 kV.) Over the period from 1999 to 2010, Partner utilities, which for inventory purposes are defined as utilities that either currently are or previously have been part of the Partnership, represented between 43 percent and 48 percent of total U.S. transmission miles. For each year, the emissions reported by or estimated for Partner utilities were added to the emissions estimated for utilities that have never participated in the Partnership (i.e., non-Partners).¹⁶⁵

Partner utilities estimated their emissions using a Tier 3 utility-level mass balance approach (IPCC 2006). If a Partner utility did not provide data for a particular year, emissions were interpolated between years for which data were available or extrapolated based on Partner-specific transmission mile growth rates. In 2010, non-reporting Partners accounted for approximately 16 percent of the total emissions attributed to Partner utilities.

Emissions from non-Partners in every year since 1999 were estimated using the results of a regression analysis that indicated that the emissions from reporting utilities were most strongly correlated with their transmission miles. The results of this analysis are not surprising given that, in the United States, SF₆ is contained primarily in transmission equipment rated above 34.5 kV. The equations were developed based on the 1999 SF₆ emissions reported by a subset of 42 Partner utilities (representing approximately 23 percent of U.S. transmission miles) and 2000 transmission mileage data obtained from the 2001 UDI Directory of Electric Power Producers and Distributors (UDI 2001). Two equations were developed, one for small and one for large utilities (i.e., with fewer or more than 10,000 transmission miles, respectively). The distinction between utility sizes was made because the regression analysis showed that the relationship between emissions and transmission miles differed for small and large transmission networks. The same equations were used to estimate non-Partner emissions in 1999 and every year thereafter because non-Partners were assumed not to have implemented any changes that would have resulted in reduced emissions since 1999.

The regression equations are:

Non-Partner small utilities (fewer than 10,000 transmission miles, in kilograms):

$$\text{Emissions (kg)} = 0.89 \times \text{Transmission Miles}$$

Non-Partner large utilities (more than 10,000 transmission miles, in kilograms):

$$\text{Emissions (kg)} = 0.58 \times \text{Transmission Miles}$$

Data on transmission miles for each non-Partner utility for the years 2000, 2003, 2006, and 2009 were obtained from the 2001, 2004, 2007, and 2010 UDI Directories of Electric Power Producers and Distributors, respectively (UDI 2001, 2004, 2007, 2010). The U.S. transmission system grew by over 25,000 miles between 2000 and 2003 and by over 52,000 miles between 2003 and 2006. These periodic increases are assumed to have occurred gradually. Therefore, transmission mileage was assumed to increase at an annual rate of 1.3 percent between 2000 and 2003 and 2.6 percent between 2003 and 2006. This growth rate slowed to 0.2% from 2006 to 2009 as transmission miles increased by just 4,400 miles (approximately).

As a final step, total electric power system emissions were determined for each year by summing the Partner reported and estimated emissions (reported data was available through the EPA's SF₆ Emission Reduction Partnership for Electric Power Systems) and the non-Partner emissions (determined using the 1999 regression equations).

1990 through 1998 Emissions from Electric Power Systems

Because most utilities participating in the Partnership reported emissions only for 1999 through 2010, modeling was used to estimate SF₆ emissions from electric power systems for the years 1990 through 1998. To perform this modeling, U.S. emissions were assumed to follow the same trajectory as global emissions from this source during the 1990 to 1999 period. To estimate global emissions, the RAND survey of global SF₆ sales were used, together with the following equation for estimating emissions, which is derived from the mass-balance equation for chemical

¹⁶⁵ Partners in EPA's SF₆ Emission Reduction Partnership reduced their emissions by approximately 62% from 1999 to 2010.

emissions (Volume 3, Equation 7.3) in the IPCC Guidelines for National Greenhouse Gas Inventories (IPCC 2006).¹⁶⁶ (Although equation 7.3 of the IPCC Guidelines appears in the discussion of substitutes for ozone-depleting substances, it is applicable to emissions from any long-lived pressurized equipment that is periodically serviced during its lifetime.)

$$\text{Emissions (kilograms SF}_6\text{)} = \text{SF}_6 \text{ purchased to refill existing equipment (kilograms)} + \text{nameplate capacity of retiring equipment (kilograms)}^{167}$$

Note that the above equation holds whether the gas from retiring equipment is released or recaptured; if the gas is recaptured, it is used to refill existing equipment, thereby lowering the amount of SF₆ purchased by utilities for this purpose.

Gas purchases by utilities and equipment manufacturers from 1961 through 2003 are available from the RAND (2004) survey. To estimate the quantity of SF₆ released or recovered from retiring equipment, the nameplate capacity of retiring equipment in a given year was assumed to equal 81.2 percent of the amount of gas purchased by electrical equipment manufacturers 40 years previous (e.g., in 2000, the nameplate capacity of retiring equipment was assumed to equal 81.2 percent of the gas purchased in 1960). The remaining 18.8 percent was assumed to have been emitted at the time of manufacture. The 18.8 percent emission factor is an average of IPCC default SF₆ emission rates for Europe and Japan for 1995 (IPCC 2006). The 40-year lifetime for electrical equipment is also based on IPCC (2006). The results of the two components of the above equation were then summed to yield estimates of global SF₆ emissions from 1990 through 1999.

U.S. emissions between 1990 and 1999 are assumed to follow the same trajectory as global emissions during this period. To estimate U.S. emissions, global emissions for each year from 1990 through 1998 were divided by the estimated global emissions from 1999. The result was a time series of factors that express each year's global emissions as a multiple of 1999 global emissions. Historical U.S. emissions were estimated by multiplying the factor for each respective year by the estimated U.S. emissions of SF₆ from electric power systems in 1999 (estimated to be 15.0 Tg CO₂ Eq.).

Two factors may affect the relationship between the RAND sales trends and actual global emission trends. One is utilities' inventories of SF₆ in storage containers. When SF₆ prices rise, utilities are likely to deplete internal inventories before purchasing new SF₆ at the higher price, in which case SF₆ sales will fall more quickly than emissions. On the other hand, when SF₆ prices fall, utilities are likely to purchase more SF₆ to rebuild inventories, in which case sales will rise more quickly than emissions. This effect was accounted for by applying 3-year averaging to utility SF₆ sales data. The other factor that may affect the relationship between the RAND sales trends and actual global emissions is the level of imports from and exports to Russia and China. SF₆ production in these countries is not included in the RAND survey and is not accounted for in any another manner by RAND. However, atmospheric studies confirm that the downward trend in estimated global emissions between 1995 and 1998 was real (see the Uncertainty discussion below).

1990 through 2010 Emissions from Manufacture of Electrical Equipment

The 1990 to 2010 emission estimates for original equipment manufacturers (OEMs) were derived by assuming that manufacturing emissions equal 10 percent of the quantity of SF₆ provided with new equipment. The quantity of SF₆ provided with new equipment was estimated based on statistics compiled by the National Electrical Manufacturers Association (NEMA). These statistics were provided for 1990 to 2000; the quantities of SF₆ provided with new equipment for 2001 to 2010 were estimated using Partner reported data and the total industry SF₆ nameplate capacity estimate (141.1 Tg CO₂ Eq. in 2010). Specifically, the ratio of new nameplate capacity to total nameplate capacity of a subset of Partners for which new nameplate capacity data was available from 1999 to 2010 was calculated. This ratio was then multiplied by the total industry nameplate capacity estimate to derive the amount of SF₆ provided with new equipment for the entire industry. The 10 percent emission rate is the average of the "ideal" and "realistic" manufacturing emission rates (4 percent and 17 percent, respectively) identified in a paper prepared

¹⁶⁶ Ideally, sales to utilities in the U.S. between 1990 and 1999 would be used as a model. However, this information was not available. Two U.S. manufacturers of SF₆ were operating during this time period, consequently, concealing sensitive sales information by aggregation was not feasible.

¹⁶⁷ Nameplate capacity is defined as the amount of SF₆ within fully charged electrical equipment.

under the auspices of the International Council on Large Electric Systems (CIGRE) in February 2002 (O’Connell et al. 2002).

Uncertainty

To estimate the uncertainty associated with emissions of SF₆ from Electric Transmission and Distribution, uncertainties associated with three quantities were estimated: (1) emissions from Partners, (2) emissions from non-Partners, and (3) emissions from manufacturers of electrical equipment. A Monte Carlo analysis was then applied to estimate the overall uncertainty of the emissions estimate.

Total emissions from the SF₆ Emission Reduction Partnership include emissions from both reporting and non-reporting Partners. For reporting Partners, individual Partner-reported SF₆ data was assumed to have an uncertainty of 10 percent. Based on a Monte Carlo analysis, the cumulative uncertainty of all Partner reported data was estimated to be 5.3 percent. The uncertainty associated with extrapolated or interpolated emissions from non-reporting Partners was assumed to be 20 percent.

There are two sources of uncertainty associated with the regression equations used to estimate emissions in 2010 from non-Partners: 1) uncertainty in the coefficients (as defined by the regression standard error estimate), and 2) the uncertainty in total transmission miles for non-Partners. In addition, there is uncertainty associated with the assumption that the emission factor used for non-Partner utilities (which accounted for approximately 57 percent of U.S. transmission miles in 2010) will remain at levels defined by Partners who reported in 1999. However, the last source of uncertainty was not modeled.

Uncertainties were also estimated regarding (1) the quantity of SF₆ supplied with equipment by equipment manufacturers, which is projected from Partner provided nameplate capacity data and industry SF₆ nameplate capacity estimates, and (2) the manufacturers’ SF₆ emissions rate.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 4-96. Electrical Transmission and Distribution SF₆ emissions were estimated to be between 9.2 and 14.7 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 22 percent below and 25 percent above the emission estimate of 11.8 Tg CO₂ Eq.

Table 4-96: Tier 2 Quantitative Uncertainty Estimates for SF₆ Emissions from Electrical Transmission and Distribution (Tg CO₂ Eq. and percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to 2010 Emission Estimate ^a			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Electrical Transmission and Distribution	SF ₆	11.8	9.2	14.7	-22%	+25%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

In addition to the uncertainty quantified above, there is uncertainty associated with using global SF₆ sales data to estimate U.S. emission trends from 1990 through 1999. However, the trend in global emissions implied by sales of SF₆ is similar to the trend in global emissions implied by changing SF₆ concentrations in the atmosphere. That is, emissions based on global sales declined by 29 percent between 1995 and 1998 (RAND 2004), and emissions based on atmospheric measurements declined by 17 percent over the same period (Levin et al. 2010).

Several pieces of evidence indicate that U.S. SF₆ emissions were reduced as global emissions were reduced. First, the decreases in sales and emissions coincided with a sharp increase in the price of SF₆ that occurred in the mid-1990s and that affected the United States as well as the rest of the world. A representative from DILO, a major manufacturer of SF₆ recycling equipment, stated that most U.S. utilities began recycling rather than venting SF₆ within two years of the price rise. Finally, the emissions reported by the one U.S. utility that reported 1990 through 1999 emissions to EPA showed a downward trend beginning in the mid-1990s.

Recalculations Discussion

SF₆ emission estimates for the period 1990 through 2009 were updated based on 1) new data from EPA’s SF₆

Emission Reduction Partnership; 2) revisions to interpolated and extrapolated non-reported Partner data; and 3) a correction made to 1999-2001 reported emissions data for a Partner. Correcting the reported emissions not only directly impacted overall emissions for 1999-2001, but also impacted the regression coefficient used to estimate emissions for non-Partners, which is based on the relationship between transmission miles and emissions for Partners that reported emissions in 1999. Specifically, the regression coefficient for utilities with fewer than 10,000 transmission miles decreased from 1.001 kg of emissions per transmission mile to 0.89 kg of emissions per transmission mile. Based on the revisions listed above, SF₆ emissions from electrical transmission and distribution decreased between 6 and 9 percent for each year from 1990 through 2009.

4.24. Industrial Sources of Indirect Greenhouse Gases

In addition to the main greenhouse gases addressed above, many industrial processes generate emissions of indirect greenhouse gases. Total emissions of nitrogen oxides (NO_x), carbon monoxide (CO), and non-CH₄ volatile organic compounds (NMVOCs) from non-energy industrial processes from 1990 to 2010 are reported in Table 4-97.

Table 4-97: NO_x, CO, and NMVOC Emissions from Industrial Processes (Gg)

Gas/Source	1990	2005	2006	2007	2008	2009	2010
NO_x	591	569	553	537	520	568	568
Other Industrial Processes	343	437	418	398	379	436	436
Chemical & Allied Product							
Manufacturing	152	55	57	59	61	55	55
Metals Processing	88	60	61	62	62	60	60
Storage and Transport	3	15	15	16	16	15	15
Miscellaneous*	5	2	2	2	2	2	2
CO	4,125	1,555	1,597	1,640	1,682	1,549	1,549
Metals Processing	2,395	752	788	824	859	752	752
Other Industrial Processes	487	484	474	464	454	484	484
Chemical & Allied Product							
Manufacturing	1,073	189	206	223	240	187	187
Storage and Transport	69	97	100	103	104	97	97
Miscellaneous*	101	32	30	27	25	29	29
NMVOCs	2,422	1,997	1,933	1,869	1,804	1,322	1,322
Storage and Transport	1,352	1,308	1,266	1,224	1,182	662	662
Other Industrial Processes	364	415	398	383	367	395	395
Chemical & Allied Product							
Manufacturing	575	213	211	210	207	206	206
Metals Processing	111	44	44	43	42	44	44
Miscellaneous*	20	17	14	10	7	15	15

* Miscellaneous includes the following categories: catastrophic/accidental release, other combustion, health services, cooling towers, and fugitive dust. It does not include agricultural fires or slash/prescribed burning, which are accounted for under the Field Burning of Agricultural Residues source.

Note: Totals may not sum due to independent rounding.

Methodology

Due to the lack of data available at the time of publication, emission estimates for 2010 rely on 2009 data as a proxy. Emission estimates for 2009 were obtained from preliminary data (EPA 2010, EPA 2009), and disaggregated based on EPA (2003), which, in its final iteration, will be published on the National Emission Inventory (NEI) Air Pollutant Emission Trends web site. Emissions were calculated either for individual categories or for many categories combined, using basic activity data (e.g., the amount of raw material processed) as an indicator of emissions. National activity data were collected for individual categories from various agencies. Depending on the category, these basic activity data may include data on production, fuel deliveries, raw material processed, etc.

Activity data were used in conjunction with emission factors, which together relate the quantity of emissions to the activity. Emission factors are generally available from the EPA's Compilation of Air Pollutant Emission Factors, AP-42 (EPA 1997). The EPA currently derives the overall emission control efficiency of a source category from a variety of information sources, including published reports, the 1985 National Acid Precipitation and Assessment Program emissions inventory, and other EPA databases.

Uncertainty and Time-Series Consistency

Uncertainties in these estimates are partly due to the accuracy of the emission factors and activity data used. A quantitative uncertainty analysis was not performed.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

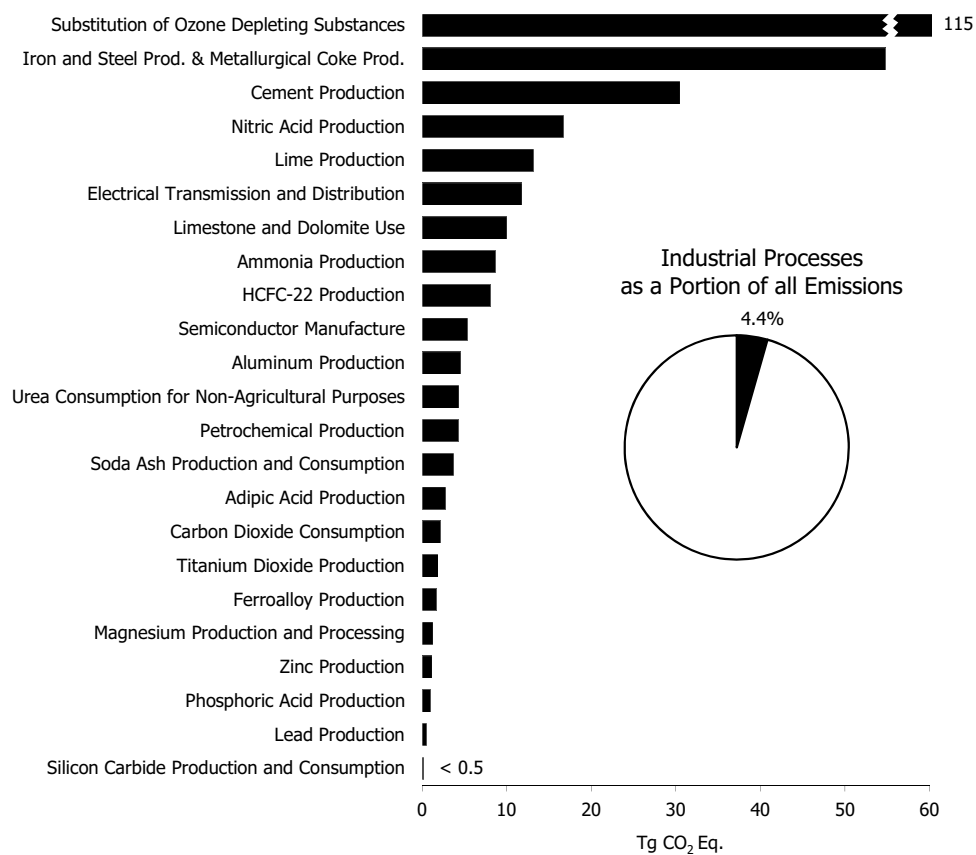


Figure 4-1: 2010 Industrial Processes Chapter Greenhouse Gas Sources

5. Solvent and Other Product Use

Greenhouse gas emissions are produced as a by-product of various solvent and other product uses. In the United States, emissions from Nitrous Oxide (N₂O) Product Uses, the only source of greenhouse gas emissions from this sector, accounted for less than 0.1 percent of total U.S. anthropogenic greenhouse gas emissions on a CO₂ equivalent basis in 2010 (see Table 5-1). Indirect greenhouse gas emissions also result from solvent and other product use, and are presented in Table 5-5 in gigagrams (Gg).

Table 5-1: N₂O Emissions from Solvent and Other Product Use (Tg CO₂ Eq. and Gg)

Gas/Source	1990	2005	2006	2007	2008	2009	2010
N ₂ O from Product Uses							
Tg CO ₂ Eq.	4.4	4.4	4.4	4.4	4.4	4.4	4.4
Gg	14	14	14	14	14	14	14

5.1. Nitrous Oxide from Product Uses (IPCC Source Category 3D)

N₂O is a clear, colorless, oxidizing liquefied gas, with a slightly sweet odor. Two companies operate a total of five N₂O production facilities in the United States (Airgas 2007; FTC 2001). N₂O is primarily used in carrier gases with oxygen to administer more potent inhalation anesthetics for general anesthesia, and as an anesthetic in various dental and veterinary applications. As such, it is used to treat short-term pain, for sedation in minor elective surgeries, and as an induction anesthetic. The second main use of N₂O is as a propellant in pressure and aerosol products, the largest application being pressure-packaged whipped cream. Small quantities of N₂O also are used in the following applications:

- Oxidizing agent and etchant used in semiconductor manufacturing;
- Oxidizing agent used, with acetylene, in atomic absorption spectrometry;
- Production of sodium azide, which is used to inflate airbags;
- Fuel oxidant in auto racing; and
- Oxidizing agent in blowtorches used by jewelers and others (Heydorn 1997).

Production of N₂O in 2010 was approximately 15 Gg (Table 5-2).

Table 5-2: N₂O Production (Gg)

Year	Gg
1990	16
2005	15
2006	15
2007	15
2008	15
2009	15
2010	15

N₂O emissions were 4.4 Tg CO₂ Eq. (14 Gg) in 2010 (Table 5-3). Production of N₂O stabilized during the 1990s because medical markets had found other substitutes for anesthetics, and more medical procedures were being performed on an outpatient basis using local anesthetics that do not require N₂O. The use of N₂O as a propellant for whipped cream has also stabilized due to the increased popularity of cream products packaged in reusable plastic tubs (Heydorn 1997).

Table 5-3: N₂O Emissions from N₂O Product Usage (Tg CO₂ Eq. and Gg)

Year	Tg CO ₂ Eq.	Gg
1990	4.4	14
2005	4.4	14
2006	4.4	14
2007	4.4	14
2008	4.4	14
2009	4.4	14
2010	4.4	14

Methodology

Emissions from N₂O product usage were calculated by first multiplying the total amount of N₂O produced in the United States by the share of the total quantity of N₂O attributed to each end use. This value was then multiplied by the associated emission rate for each end use. After the emissions were calculated for each end use, they were added together to obtain a total estimate of N₂O product usage emissions. Emissions were determined using the following equation:

$$\text{N}_2\text{O Product Usage Emissions} = \sum_i [\text{Total U.S. Production of N}_2\text{O}] \times [\text{Share of Total Quantity of N}_2\text{O Usage by Sector } i] \times [\text{Emissions Rate for Sector } i]$$

where,

i = Sector.

The share of total quantity of N₂O usage by end use represents the share of national N₂O produced that is used by the specific subcategory (i.e., anesthesia, food processing, etc.). In 2010, the medical/dental industry used an estimated 89.5 percent of total N₂O produced, followed by food processing propellants at 6.5 percent. All other categories combined used the remainder of the N₂O produced. This subcategory breakdown has changed only slightly over the past decade. For instance, the small share of N₂O usage in the production of sodium azide has declined significantly during the 1990s. Due to the lack of information on the specific time period of the phase-out in this market subcategory, most of the N₂O usage for sodium azide production is assumed to have ceased after 1996, with the majority of its small share of the market assigned to the larger medical/dental consumption subcategory (Heydorn 1997). The N₂O was allocated across the following categories: medical applications, food processing propellant, and sodium azide production (pre-1996). A usage emissions rate was then applied for each sector to estimate the amount of N₂O emitted.

Only the medical/dental and food propellant subcategories were estimated to release emissions into the atmosphere, and therefore these subcategories were the only usage subcategories with emission rates. For the medical/dental subcategory, due to the poor solubility of N₂O in blood and other tissues, none of the N₂O is assumed to be metabolized during anesthesia and quickly leaves the body in exhaled breath. Therefore, an emission factor of 100 percent was used for this subcategory (IPCC 2006). For N₂O used as a propellant in pressurized and aerosol food products, none of the N₂O is reacted during the process and all of the N₂O is emitted to the atmosphere, resulting in an emission factor of 100 percent for this subcategory (IPCC 2006). For the remaining subcategories, all of the N₂O is consumed/reacted during the process, and therefore the emission rate was considered to be zero percent (Tupman 2002).

The 1990 through 1992 N₂O production data were obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997). N₂O production data for 1993 through 1995 were not available. Production data for 1996 was specified as a range in two data sources (Heydorn 1997, Tupman 2002). In particular, for 1996, Heydorn (1997) estimates N₂O production to range between 13.6 and 18.1 thousand metric tons. Tupman (2003) provided a narrower range (15.9 to 18.1 thousand metric tons) for 1996 that falls within the production bounds described by Heydorn (1997). Tupman (2003) data are considered more industry-specific and current. Therefore, the midpoint of the narrower production range was used to estimate N₂O emissions for years 1993 through 2001 (Tupman 2003). The 2002 and 2003 N₂O production data were obtained from the Compressed Gas Association Nitrous Oxide Fact Sheet and Nitrous Oxide Abuse Hotline (CGA 2002, 2003). These data were also provided as a range. For

example, in 2003, CGA (2003) estimates N₂O production to range between 13.6 and 15.9 thousand metric tons. Due to unavailable data, production estimates for years 2004 through 2010 were held at the 2003 value.

The 1996 share of the total quantity of N₂O used by each subcategory was obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997). The 1990 through 1995 share of total quantity of N₂O used by each subcategory was kept the same as the 1996 number provided by SRI Consulting. The 1997 through 2001 share of total quantity of N₂O usage by sector was obtained from communication with a N₂O industry expert (Tupman 2002). The 2002 and 2003 share of total quantity of N₂O usage by sector was obtained from CGA (2002, 2003). Due to unavailable data, the share of total quantity of N₂O usage data for years 2004 through 2010 was assumed to equal the 2003 value. The emissions rate for the food processing propellant industry was obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997), and confirmed by a N₂O industry expert (Tupman 2002). The emissions rate for all other subcategories was obtained from communication with a N₂O industry expert (Tupman 2002). The emissions rate for the medical/dental subcategory was obtained from the 2006 IPCC Guidelines.

Uncertainty and Time-Series Consistency

The overall uncertainty associated with the 2010 N₂O emission estimate from N₂O product usage was calculated using the IPCC Guidelines for National Greenhouse Gas Inventories (2006) Tier 2 methodology. Uncertainty associated with the parameters used to estimate N₂O emissions include production data, total market share of each end use, and the emission factors applied to each end use, respectively.

The results of this Tier 2 quantitative uncertainty analysis are summarized in Table 5-4. N₂O emissions from N₂O product usage were estimated to be between 4.1 and 4.7 Tg CO₂ Eq. at the 95 percent confidence level. This indicates a range of approximately 8 percent below to 8 percent above the emissions estimate of 4.4 Tg CO₂ Eq.

Table 5-4: Tier 2 Quantitative Uncertainty Estimates for N₂O Emissions from N₂O Product Usage (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^a			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
N ₂ O Product Usage	N ₂ O	4.4	4.1	4.7	-8%	+8%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Furthermore, methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time-series are described in more detail in the Methodology section, above.

Planned Improvements

Planned improvements include a continued evaluation of alternative production statistics for cross verification, a reassessment of N₂O product use subcategories to accurately represent trends, investigation of production and use cycles, and the potential need to incorporate a time lag between production and ultimate product use and resulting release of N₂O. Additionally, planned improvements include considering imports and exports of N₂O for product uses.

5.2. Indirect Greenhouse Gas Emissions from Solvent Use

The use of solvents and other chemical products can result in emissions of various ozone precursors (i.e., indirect greenhouse gases).¹⁶⁸ Non-CH₄ volatile organic compounds (NMVOCs), commonly referred to as “hydrocarbons,”

¹⁶⁸ Solvent usage in the United States also results in the emission of small amounts of hydrofluorocarbons (HFCs) and

are the primary gases emitted from most processes employing organic or petroleum based solvents. As some of industrial applications also employ thermal incineration as a control technology, combustion by-products, such as carbon monoxide (CO) and nitrogen oxides (NO_x), are also reported with this source category. In the United States, emissions from solvents are primarily the result of solvent evaporation, whereby the lighter hydrocarbon molecules in the solvents escape into the atmosphere. The evaporation process varies depending on different solvent uses and solvent types. The major categories of solvent uses include: degreasing, graphic arts, surface coating, other industrial uses of solvents (i.e., electronics, etc.), dry cleaning, and non-industrial uses (i.e., uses of paint thinner, etc.).

Total emissions of NO_x, NMVOCs, and CO from 1990 to 2010 are reported in Table 5-5.

Table 5-5: Emissions of NO_x, CO, and NMVOC from Solvent Use (Gg)

Activity	1990	2005	2006	2007	2008	2009	2010
NO_x	1	3	4	4	4	3	3
Surface Coating	1	3	4	4	4	3	3
Graphic Arts	+	+	+	+	+	+	+
Degreasing	+	+	+	+	+	+	+
Dry Cleaning	+	+	+	+	+	+	+
Other Industrial Processes ^a	+	+	+	+	+	+	+
Non-Industrial Processes ^b	+	+	+	+	+	+	+
Other	NA	+	+	+	+	+	+
CO	5	2	2	2	2	2	2
Surface Coating	+	2	2	2	2	2	2
Other Industrial Processes ^a	4	+	+	+	+	+	+
Dry Cleaning	+	+	+	+	+	+	+
Degreasing	+	+	+	+	+	+	+
Graphic Arts	+	+	+	+	+	+	+
Non-Industrial Processes ^b	+	+	+	+	+	+	+
Other	NA	+	+	+	+	+	+
NMVOCs	5,216	3,851	3,846	3,839	3,834	2,583	2,583
Surface Coating	2,289	1,578	1,575	1,573	1,571	1,058	1,058
Non-Industrial Processes ^b	1,724	1,446	1,444	1,441	1,439	970	970
Degreasing	675	280	280	280	279	188	188
Dry Cleaning	195	230	230	229	229	154	154
Graphic Arts	249	194	193	193	193	130	130
Other Industrial Processes ^a	85	88	88	87	87	59	59
Other	+	36	36	36	36	24	24

^a Includes rubber and plastics manufacturing, and other miscellaneous applications.

^b Includes cutback asphalt, pesticide application adhesives, consumer solvents, and other miscellaneous applications.

Note: Totals may not sum due to independent rounding.

+ Does not exceed 0.5 Gg.

Methodology

Emissions were calculated by aggregating solvent use data based on information relating to solvent uses from different applications such as degreasing, graphic arts, etc. Emission factors for each consumption category were

hydrofluoroethers (HFEs), which are included under Substitution of Ozone Depleting Substances in the Industrial Processes chapter.

then applied to the data to estimate emissions. For example, emissions from surface coatings were mostly due to solvent evaporation as the coatings solidify. By applying the appropriate solvent-specific emission factors to the amount of solvents used for surface coatings, an estimate of emissions was obtained. Emissions of CO and NO_x result primarily from thermal and catalytic incineration of solvent-laden gas streams from painting booths, printing operations, and oven exhaust.

Due to the lack of data available at the time of publication, emission estimates for 2010 rely on 2009 data as a proxy. Emission estimates for 2009 were obtained from preliminary data (EPA 2010, EPA 2009), and disaggregated based on EPA (2003), which, in its final iteration, will be published on the National Emission Inventory (NEI) Air Pollutant Emission Trends web site. Emissions were calculated either for individual categories or for many categories combined, using basic activity data (e.g., the amount of solvent purchased) as an indicator of emissions. National activity data were collected for individual applications from various agencies.

Activity data were used in conjunction with emission factors, which together relate the quantity of emissions to the activity. Emission factors are generally available from the EPA's Compilation of Air Pollutant Emission Factors, AP-42 (EPA 1997). The EPA currently derives the overall emission control efficiency of a source category from a variety of information sources, including published reports, the 1985 National Acid Precipitation and Assessment Program emissions inventory, and other EPA databases.

Uncertainty and Time-Series Consistency

Uncertainties in these estimates are partly due to the accuracy of the emission factors used and the reliability of correlations between activity data and actual emissions.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

6. Agriculture

Agricultural activities contribute directly to emissions of greenhouse gases through a variety of processes. This chapter provides an assessment of non-carbon-dioxide emissions from the following source categories: enteric fermentation in domestic livestock, livestock manure management, rice cultivation, agricultural soil management, and field burning of agricultural residues (see Figure 6-1). Carbon dioxide (CO₂) emissions and removals from agriculture-related land-use activities, such as liming of agricultural soils and conversion of grassland to cultivated land, are presented in the Land Use, Land-Use Change, and Forestry chapter. Carbon dioxide emissions from on-farm energy use are accounted for in the Energy chapter.

Figure 6-1: 2010 Agriculture Chapter Greenhouse Gas Emission Sources

In 2010, the Agriculture sector was responsible for emissions of 428.4 teragrams of CO₂ equivalents (Tg CO₂ Eq.), or 6.3 percent of total U.S. greenhouse gas emissions. Methane (CH₄) and nitrous oxide (N₂O) were the primary greenhouse gases emitted by agricultural activities. Methane emissions from enteric fermentation and manure management represent about 21 percent and 8 percent of total CH₄ emissions from anthropogenic activities, respectively. Of all domestic animal types, beef and dairy cattle were by far the largest emitters of CH₄. Rice cultivation and field burning of agricultural residues were minor sources of CH₄. Agricultural soil management activities such as fertilizer application and other cropping practices were the largest source of U.S. N₂O emissions, accounting for 68 percent. Manure management and field burning of agricultural residues were also small sources of N₂O emissions.

Table 6-1 and Table 6-2 present emission estimates for the Agriculture sector. Between 1990 and 2010, CH₄ emissions from agricultural activities increased by 14.5 percent, while N₂O emissions fluctuated from year to year, but overall increased by 5.0 percent.

Table 6-1: Emissions from Agriculture (Tg CO₂ Eq.)

Gas/Source	1990	2005	2006	2007	2008	2009	2010
CH₄	172.9	193.9	195.9	202.9	202.6	200.8	202.2
Enteric Fermentation	133.8	139.0	141.4	143.8	143.4	142.6	141.3
Manure Management	31.7	47.9	48.4	52.7	51.8	50.7	52.0
Rice Cultivation	7.1	6.8	5.9	6.2	7.2	7.3	8.6
Field Burning of Agricultural Residues	0.2	0.2	0.2	0.2	0.2	0.2	0.2
N₂O	214.9	230.7	229.6	229.7	231.3	225.6	226.2
Agricultural Soil Management	200.0	213.1	211.1	211.1	212.9	207.3	207.8
Manure Management	14.8	17.6	18.4	18.5	18.3	18.2	18.3
Field Burning of Agricultural Residues	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Total	387.8	424.6	425.4	432.6	433.8	426.4	428.4

Note: Totals may not sum due to independent rounding.

Table 6-2: Emissions from Agriculture (Gg)

Gas/Source	1990	2005	2006	2007	2008	2009	2010
CH₄	8,234	9,232	9,327	9,663	9,647	9,564	9,627
Enteric Fermentation	6,373	6,618	6,731	6,850	6,829	6,788	6,728
Manure Management	1,511	2,280	2,303	2,508	2,465	2,416	2,478
Rice Cultivation	339	326	282	295	343	349	410
Field Burning of Agricultural Residues	10	8	11	11	11	11	11
N₂O	693	744	741	741	746	728	730
Agricultural Soil Management	645	687	681	681	687	669	670
Manure Management	48	57	59	60	59	59	59

Field Burning of Agricultural Residues	+		+	+	+	+	+	+
+ Less than 0.5 Gg.								
Note: Totals may not sum due to independent rounding.								

6.1. Enteric Fermentation (IPCC Source Category 4A)

Methane is produced as part of normal digestive processes in animals. During digestion, microbes resident in an animal's digestive system ferment food consumed by the animal. This microbial fermentation process, referred to as enteric fermentation, produces CH₄ as a byproduct, which can be exhaled or eructated by the animal. The amount of CH₄ produced and emitted by an individual animal depends primarily upon the animal's digestive system, and the amount and type of feed it consumes.

Ruminant animals (e.g., cattle, buffalo, sheep, goats, and camels) are the major emitters of CH₄ because of their unique digestive system. Ruminants possess a rumen, or large "fore-stomach," in which microbial fermentation breaks down the feed they consume into products that can be absorbed and metabolized. The microbial fermentation that occurs in the rumen enables them to digest coarse plant material that non-ruminant animals cannot. Ruminant animals, consequently, have the highest CH₄ emissions among all animal types.

Non-ruminant animals (e.g., swine, horses, and mules) also produce CH₄ emissions through enteric fermentation, although this microbial fermentation occurs in the large intestine. These non-ruminants emit significantly less CH₄ on a per-animal basis than ruminants because the capacity of the large intestine to produce CH₄ is lower.

In addition to the type of digestive system, an animal's feed quality and feed intake also affect CH₄ emissions. In general, lower feed quality and/or higher feed intake leads to higher CH₄ emissions. Feed intake is positively correlated to animal size, growth rate, and production (e.g., milk production, wool growth, pregnancy, or work). Therefore, feed intake varies among animal types as well as among different management practices for individual animal types (e.g., animals in feedlots or grazing on pasture).

Methane emission estimates from enteric fermentation are provided in Table 6-3 and Table 6-4.

Total livestock CH₄ emissions in 2010 were 141.3 Tg CO₂ Eq. (6,728 Gg). Beef cattle remain the largest contributor of CH₄ emissions from enteric fermentation, accounting for 72 percent in 2010. Emissions from dairy cattle in 2010 accounted for 23 percent, and the remaining emissions were from horses, sheep, swine, goats, American bison, mules, burros, and donkeys.

From 1990 to 2010, emissions from enteric fermentation have increased by 5.6 percent. Generally, emissions decreased from 1996 to 2003, though with a slight increase in 2002. This trend was mainly due to decreasing populations of both beef and dairy cattle and increased digestibility of feed for feedlot cattle. Emissions increased from 2004 through 2007, as both dairy and beef populations have undergone increases and the literature for dairy cow diets indicated a trend toward a decrease in feed digestibility for those years. Emissions decreased again in 2008 to 2010 as beef cattle populations again decreased. During the timeframe of this analysis, populations of sheep have decreased 51 percent while horse populations have increased over 87 percent, mostly between 2001 and 2006. Goat and swine populations have increased 25 percent and 20 percent, respectively, during this timeframe, though with slight decreases from 2009 to 2010, while the populations of American bison and mules, burros, and donkeys have more than quadrupled.

Table 6-3: CH₄ Emissions from Enteric Fermentation (Tg CO₂ Eq.)

Livestock Type	1990	2005	2006	2007	2008	2009	2010
Beef Cattle	96.2	101.4	103.0	104.0	103.1	102.0	101.1
Dairy Cattle	31.8	30.4	31.1	32.4	32.9	33.2	33.0
Horses	1.9	3.5	3.6	3.6	3.6	3.6	3.6
Swine	1.7	1.9	1.0	2.1	2.1	2.1	2.0
Sheep	1.9	1.0	1.9	1.0	1.0	1.0	0.9
Goats	0.3	0.3	0.3	0.3	0.3	0.3	0.3
American Bison	0.1	0.4	0.4	0.3	0.4	0.3	0.3
Mules, Burros, and Donkeys	+	+	0.1	0.1	0.1	0.1	0.1
Total	133.8	139.0	141.4	143.8	143.4	142.6	141.3

Notes: + Does not exceed 0.05 Tg CO₂ Eq. Totals may not sum due to independent rounding.

Table 6-4: CH₄ Emissions from Enteric Fermentation (Gg)

Livestock Type	1990	2005	2006	2007	2008	2009	2010
Beef Cattle	4,581	4,829	4,904	4,953	4,909	4,857	4,812
Dairy Cattle	1,513	1,449	1,479	1,544	1,564	1,581	1,569
Horses	91	166	171	171	171	171	171
Swine	81	92	93	98	101	99	97
Sheep	91	49	50	49	48	46	45
Goats	13	14	15	16	16	16	16
American Bison	4	17	17	16	17	17	16
Mules, Burros, and Donkeys	1	2	2	3	3	3	3
Total	6,373	6,618	6,731	6,850	6,829	6,788	6,728

Note: Totals may not sum due to independent rounding.

Methodology

Livestock emission estimate methodologies fall into two categories: cattle and other domesticated animals. Cattle, due to their large population, large size, and particular digestive characteristics, account for the majority of CH₄ emissions from livestock in the United States. A more detailed methodology (i.e., IPCC Tier 2) was therefore applied to estimate emissions for all cattle. Emission estimates for other domesticated animals (horses, sheep, swine, goats, American bison, and mules, burrow, and donkeys) were handled using a less detailed approach (i.e., IPCC Tier 1).

While the large diversity of animal management practices cannot be precisely characterized and evaluated, significant scientific literature exists that provides the necessary data to estimate cattle emissions using the IPCC Tier 2 approach. The Cattle Enteric Fermentation Model (CEFM), developed by EPA and used to estimate cattle CH₄ emissions from enteric fermentation, incorporates this information and other analyses of livestock population, feeding practices, and production characteristics.

National cattle population statistics were disaggregated into the following cattle sub-populations:

- Dairy Cattle
 - Calves
 - Heifer Replacements
 - Cows
- Beef Cattle
 - Calves
 - Heifer Replacements
 - Heifer and Steer Stockers
 - Animals in Feedlots (Heifers and Steers)
 - Cows
 - Bulls

Calf birth rates, end-of-year population statistics, detailed feedlot placement information, and slaughter weight data were used to create a transition matrix that models cohorts of individual animal types and their specific emission profiles. The key variables tracked for each of the cattle population categories are described in Annex 3.9. These variables include performance factors such as pregnancy and lactation as well as average weights and weight gain. Annual cattle population data were obtained from the U.S. Department of Agriculture's (USDA) National

Agricultural Statistics Service (NASS) QuickStats database (USDA 2011).

Diet characteristics were estimated by region for U.S. dairy, foraging beef, and feedlot beef cattle. These estimates were used to calculate Digestible Energy (DE) values (expressed as the percent of gross energy intake digested by the animal) and CH₄ conversion rates (Y_m) (expressed as the fraction of gross energy converted to CH₄) for each population category. The IPCC recommends Y_m ranges of 3.0±1.0 percent for feedlot cattle and 6.5±1.0 percent for other well-fed cattle consuming temperate-climate feed types (IPCC 2006). Given the availability of detailed diet information for different regions and animal types in the United States, DE and Y_m values unique to the United States were developed. The diet characterizations and estimation of DE and Y_m values were based on information from state agricultural extension specialists, a review of published forage quality studies and scientific literature, expert opinion, and modeling of animal physiology.

The diet characteristics for dairy cattle were based on Donovan (1999) and an extensive review of nearly 20 years of literature from 1990 through 2009. Estimates of DE were national averages based on the feed components of the diets observed in the literature for the following year groupings: 1990-1993, 1994-1998, 1999-2002, 2003, 2004-2006, 2007, and 2008 onwards. Base year Y_m values by region were estimated using Donovan (1999). A ruminant digestion model (COWPOLL, as selected in Kebreab et al. 2008) was used to evaluate Y_m for each diet evaluated from the literature, and a function was developed to adjust regional values over time based on the national trend. Dairy replacement heifer diet assumptions were based on the observed relationship in the literature between dairy cow and dairy heifer diet characteristics.

For feedlot animals, the DE and Y_m values used for 1990 were recommended by Johnson (1999). Values for DE and Y_m for 1991 through 1999 were linearly extrapolated based on the 1990 and 2000 data. DE and Y_m values for 2000 onwards were based on survey data in Galyean and Gleghorn (2001) and Vasconcelos and Galyean (2007).

For grazing beef cattle, Y_m values were based on Johnson (2002), DE values for 1990 through 2006 were based on specific diet components estimated from Donovan (1999), and DE values from 2007 onwards were developed from an analysis by Archibeque (2011), based on diet information in USDA (2010). Weight and weight gains for cattle were estimated from Holstein (2010), Doren et al. (1989), Enns (2008), Lippke et al. (2000), Pinchack et al. (2004), Platter et al. (2003), Skogerboe et al. (2000), and expert opinion. See Annex 3.9 for more details on the method used to characterize cattle diets and weights in the United States.

To estimate CH₄ emissions from all cattle types except calves younger than 7 months,¹⁶⁹ the population was divided into state, age, sub-type (i.e., dairy cows and replacements, beef cows and replacements, heifer and steer stockers, heifers and steers in feedlots, and bulls), and production (i.e., pregnant, lactating) groupings to more fully capture differences in CH₄ emissions from these animal types. The transition matrix was used to simulate the age and weight structure of each sub-type on a monthly basis, to more accurately reflect the fluctuations that occur throughout the year. Cattle diet characteristics were then used in conjunction with Tier 2 equations from IPCC (2006) to produce CH₄ emission factors for the following cattle types: dairy cows, beef cows, dairy replacements, beef replacements, steer stockers, heifer stockers, steer feedlot animals, heifer feedlot animals, and bulls. To estimate emissions from cattle, population data from the transition matrix were multiplied by the calculated emission factor for each cattle type. More details are provided in Annex 3.9.

Emission estimates for other animal types were based on average emission factors representative of entire populations of each animal type. Methane emissions from these animals accounted for a minor portion of total CH₄ emissions from livestock in the United States from 1990 through 2010. Also, the variability in emission factors for each of these other animal types (e.g., variability by age, production system, and feeding practice within each animal type) is less than that for cattle. Annual livestock population data for sheep and swine were obtained for all years from USDA NASS (USDA 2011). Horse population data were obtained from the Food and Agriculture Organization of the United Nations (FAO) FAOSTAT database (FAO 2011), because USDA does not estimate U.S. horse populations annually. Goat and mule, burro, and donkey population data were available for 1987, 1992, 1997, 2002, and 2007 (USDA 1992, 1997, 2011); the remaining years between 1990 and 2010 were interpolated and extrapolated from the available estimates. American bison population estimates were available from USDA for 2002 and 2007 (USDA 2011) and from the National Bison Association (1999) for 1997 through 1999. Additional years were based on observed trends from the National Bison Association (1999), interpolation between known data

¹⁶⁹ Because calves younger than 7 months consume mainly milk and the IPCC recommends the use of a methane conversion factor of zero for all juveniles consuming only milk, this results in no methane emissions from this subcategory of cattle.

points, and ratios of population to slaughter statistics (USDA 2011), as described in more detail in Annex 3.9. Methane emissions from sheep, goats, swine, horses, American bison, and mules, burros, and donkeys were estimated by using emission factors utilized in Crutzen et al. (1986, cited in IPCC 2006). These emission factors are representative of typical animal sizes, feed intakes, and feed characteristics in developed countries. For American bison the emission factor for buffalo was used and adjusted based on the ratio of live weights to the 0.75 power. The methodology is the same as that recommended by IPCC (2006).

See Annex 3.9 for more detailed information on the methodology and data used to calculate CH₄ emissions from enteric fermentation.

Uncertainty and Time-Series Consistency

A quantitative uncertainty analysis for this source category was performed using the IPCC-recommended Tier 2 uncertainty estimation methodology, Monte Carlo Stochastic Simulation technique as described in ICF (2003). These uncertainty estimates were developed for the 1990 through 2001 Inventory report. There have been no significant changes to the methodology, although the source of some input variables have been updated, at this time there are not better estimates available for the uncertainty ranges around the 2010 activity data and emission factor input variables used in the current submission. Consequently, these uncertainty estimates were directly applied to the 2010 emission estimates.

A total of 185 primary input variables (177 for cattle and 8 for non-cattle) were identified as key input variables for the uncertainty analysis. A normal distribution was assumed for almost all activity- and emission factor-related input variables. Triangular distributions were assigned to three input variables (specifically, cow-birth ratios for the three most recent years included in the 2001 model run) to ensure only positive values would be simulated. For some key input variables, the uncertainty ranges around their estimates (used for inventory estimation) were collected from published documents and other public sources; others were based on expert opinion and best estimates. In addition, both endogenous and exogenous correlations between selected primary input variables were modeled. The exogenous correlation coefficients between the probability distributions of selected activity-related variables were developed through expert judgment.

The uncertainty ranges associated with the activity data-related input variables were plus or minus 10 percent or lower. However, for many emission factor-related input variables, the lower- and/or the upper-bound uncertainty estimates were over 20 percent. The results of the quantitative uncertainty analysis are summarized in Table 6-5. Based on this analysis, enteric fermentation CH₄ emissions in 2010 were estimated to be between 125.8 and 166.7 Tg CO₂ Eq. at a 95 percent confidence level, which indicates a range of 11 percent below to 18 percent above the 2010 emission estimate of 141.3 Tg CO₂ Eq. Among the individual cattle sub-source categories, beef cattle account for the largest amount of CH₄ emissions as well as the largest degree of uncertainty in the emission estimates. Among non-cattle, horses account for the largest degree of uncertainty in the emission estimates because there is a higher degree of uncertainty among the FAO population estimates used for horses than for the USDA population estimates used for swine, goats, and sheep. American bison, mules, burros, and donkeys were excluded from the initial uncertainty estimate because they were not included in the estimate of emissions at that time, although because of their small populations they would not significantly increase the uncertainty estimate ranges of the overall emissions from enteric fermentation.

Table 6-5: Quantitative Uncertainty Estimates for CH₄ Emissions from Enteric Fermentation (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^{a, b, c}			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Enteric Fermentation	CH ₄	141.3	125.8	166.7	-11%	+18%

^a Range of emissions estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

^b Note that the relative uncertainty range was estimated with respect to the 2001 emission estimates submitted in 2003 and applied to the 2010 estimates.

^c The overall uncertainty calculated in 2003, and applied to the 2010 emission estimate, did not include uncertainty estimates for American bison, mules, burros, and donkeys, and was based on the Tier 1 methodology for bulls. Consequently, there was more uncertainty with bull emissions than with other cattle types.

Methodological recalculations were applied to the entire time series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section.

QA/QC and Verification

In order to ensure the quality of the emission estimates from enteric fermentation, the IPCC Tier 1 and Tier 2 Quality Assurance/Quality Control (QA/QC) procedures were implemented consistent with the U.S. QA/QC plan. Tier 2 QA procedures included independent peer review of emission estimates. Recent updates to the foraging portion of the diet values for cattle made this the area of emphasis for QA/QC this year, with specific attention to the data sources and comparisons of the current estimates with previous estimates.

In addition, over the past few years, particular importance has been placed on harmonizing the data exchange between the enteric fermentation and manure management source categories. The current inventory submission now utilizes the transition matrix from the CEFM for estimating cattle populations and weights for both source categories, and the CEFM is used to output volatile solids and nitrogen (N) excretion estimates using the diet assumptions in the model in conjunction with the energy balance equations from the IPCC (2006). This approach facilitates the QA/QC process for both of these source categories.

Recalculations Discussion

There were several modifications to the methodology that had an effect on emission estimates, including the following:

- Emissions from bulls were estimated using Tier 2 methodology. This resulted in an increase of emissions from bulls by an average of approximately 79 percent per year compared to the previous Inventory estimates which used a Tier 1 methodology, such that bulls represent 3.4 percent of total enteric fermentation emissions from cattle.
- Revisions to the DE values for foraging cattle diets were applied to 1990-2010, resulting in an average change of less than 0.1 percent for foraging beef cattle emissions estimates for 1990 through 2006 and an average increase of 0.4 percent for 2007 through 2009. Details on the current dietary assumptions are discussed in Annex 3.9.
- During the QA/QC process, it was realized that the one data point from 1988 (total births) had been revised by USDA since its original download. Therefore, the data point was corrected from 39,318.0 to 39,317.9 thousand births. This is a very minor change, but it is noted in detail specifically because it affects 1990 base year emissions by trickling through the transition matrix in the growing populations for 1989 and 1990.
- The equations used to distribute end-of-year remaining populations for feedlot cattle to the individual state populations were updated so that the population proportions reflect the current year rather than the following year populations. This did not affect total populations, but there were minor changes to the populations by state for feedlot cattle for all years.
- Previously, American bison and mules, burros, and donkeys were excluded from this source category. Emission estimates are now included for these animal types for all years, and contribute an average of 0.2 percent of total emissions from enteric fermentation across the time series.
- The USDA published revised estimates in several categories that affected historical emissions estimated for cattle, including slight revisions in 2009 cattle on feed population estimates for “other states” (aggregated data for states with small populations of cattle on feed), dairy cow milk production for several states, and steer and heifer placement and slaughter statistics. Additionally, calf births were revised for both the 2008 and 2009 estimates. These changes had an insignificant impact on the overall results.
- There were additional population changes for goats from 2003 through 2006, sheep for 2004, 2006, and 2009, and swine in 2009, as discussed in the recalculations discussion for manure management. Historical emission estimates for goats increased an average of 12.1 percent per year compared to the previous emission estimates for the years mentioned above. All other population changes resulted in a decrease in emissions of less than 1 percent for the animal type and year noted.

As a result of these changes, overall methane emissions from enteric fermentation increased an average of 111 Gg (1.7 percent) per year for 1990 through 2009.

Planned Improvements

Continued research and regular updates are necessary to maintain an emissions inventory that reflects the current base of knowledge. Ongoing revisions for enteric fermentation could include some of the following options:

- Updating input variables that are from older data sources, such as beef births by month and beef cow lactation rates;
- Investigation of the availability of annual data for the DE and crude protein values of specific diet and feed components for foraging and feedlot animals;
- Given the many challenges in characterizing dairy diets, further investigation will be conducted on additional sources or methodologies for estimating DE for dairy. For example, the current method causes some significant shifts in data between years that may not mimic actual feeding conditions. Regional trend lines may be used to smooth the transition.
- The possible breakout of other animal types (i.e., sheep, swine, goats, horses) from national estimates to state-level estimates or updating to Tier 2 methodology; and
- The investigation of methodologies for including enteric fermentation emission estimates from poultry.

In addition, recent changes that have been implemented to the CEFM warrant an assessment of the current uncertainty analysis; therefore, a revision of the quantitative uncertainty surrounding emission estimates from this source category will be initiated.

6.2. Manure Management (IPCC Source Category 4B)

The management of livestock manure can produce anthropogenic CH₄ and N₂O emissions. Methane is produced by the anaerobic decomposition of manure. Direct N₂O emissions are produced as part of the N cycle through the nitrification and denitrification of the organic N in livestock dung and urine.¹⁷⁰ Indirect N₂O emissions are produced as result of the volatilization of N as NH₃ and NO_x and runoff and leaching of N during treatment, storage and transportation.

When livestock or poultry manure are stored or treated in systems that promote anaerobic conditions (e.g., as a liquid/slurry in lagoons, ponds, tanks, or pits), the decomposition of materials in the manure tends to produce CH₄. When manure is handled as a solid (e.g., in stacks or drylots) or deposited on pasture, range, or paddock lands, it tends to decompose aerobically and produce little or no CH₄. Ambient temperature, moisture, and manure storage or residency time affect the amount of CH₄ produced because they influence the growth of the bacteria responsible for CH₄ formation. For non-liquid-based manure systems, moist conditions (which are a function of rainfall and humidity) can promote CH₄ production. Manure composition, which varies by animal diet, growth rate, and type, including the animal's digestive system, also affects the amount of CH₄ produced. In general, the greater the energy content of the feed, the greater the potential for CH₄ emissions. However, some higher-energy feeds also are more digestible than lower quality forages, which can result in less overall waste excreted from the animal.

The production of direct N₂O emissions from livestock manure depends on the composition of the manure and urine, the type of bacteria involved in the process, and the amount of oxygen and liquid in the manure system. For direct N₂O emissions to occur, the manure must first be handled aerobically where ammonia (NH₃) or organic N is converted to nitrates and nitrites (nitrification), and then handled anaerobically where the nitrates and nitrites are reduced to dinitrogen gas (N₂), with intermediate production of N₂O and nitric oxide (NO) (denitrification) (Groffman et al. 2000). These emissions are most likely to occur in dry manure handling systems that have aerobic conditions, but that also contain pockets of anaerobic conditions due to saturation. A very small portion of the total

¹⁷⁰ Direct and indirect N₂O emissions from dung and urine spread onto fields either directly as daily spread or after it is removed from manure management systems (e.g., lagoon, pit, etc.) and from livestock dung and urine deposited on pasture, range, or paddock lands are accounted for and discussed in the Agricultural Soil Management source category within the Agriculture sector.

N excreted is expected to convert to N₂O in the waste management system (WMS). Indirect N₂O emissions are produced when nitrogen is lost from the system through volatilization (as NH₃ or NO_x) or through runoff and leaching. The vast majority of volatilization losses from these operations are NH₃. Although there are also some small losses of NO_x, there are no quantified estimates available for use, so losses due to volatilization are only based on NH₃ loss factors. Runoff losses would be expected from operations that house animals or store manure in a manner that is exposed to weather. Runoff losses are also specific to the type of animal housed on the operation due to differences in manure characteristics. Little information is known about leaching from manure management systems as most research focuses on leaching from land application systems. Since leaching losses are expected to be minimal, leaching losses are coupled with runoff losses and the runoff/leaching estimate does not include any leaching losses.

Estimates of CH₄ emissions in 2010 were 52.0 Tg CO₂ Eq. (2,478 Gg), 64 percent higher than in 1990. Emissions increased on average by 1.0 Tg CO₂ Eq. (3.0 percent) annually over this period. The majority of this increase was from swine and dairy cow manure, where emissions increased 20 and 107 percent, respectively. Although the majority of manure in the United States is handled as a solid, producing little CH₄, the general trend in manure management, particularly for dairy and swine (which are both shifting towards larger facilities), is one of increasing use of liquid systems. Also, new regulations limiting the application of manure nutrients have shifted manure management practices at smaller dairies from daily spread to manure managed and stored on site. Although national dairy animal populations have been generally decreasing, some states have seen increases in their dairy populations as the industry becomes more concentrated in certain areas of the country. These areas of concentration, such as California, New Mexico, and Idaho, tend to utilize more liquid-based systems to manage (flush or scrape) and store manure. Thus the shift toward larger facilities is translated into an increasing use of liquid manure management systems, which have higher potential CH₄ emissions than dry systems. This shift was accounted for by incorporating state and WMS-specific CH₄ conversion factor (MCF) values in combination with the 1992, 1997, 2002, and 2007 farm-size distribution data reported in the *Census of Agriculture* (USDA 2009a). Methane emissions from sheep have decreased significantly since 1990 (a 56 percent decrease from 1990 to 2010); however, this is mainly due to population changes. Overall, sheep contribute less than one percent of CH₄ emissions from animal manure management. From 2009 to 2010, there was a 2.6 percent increase in total CH₄ emissions, mainly due to minor shifts in the animal populations and the resultant effects on manure management system allocations.

In 2010, total N₂O emissions were estimated to be 18.3 Tg CO₂ Eq. (59 Gg); in 1990, emissions were 14.8 Tg CO₂ Eq. (48 Gg). These values include both direct and indirect N₂O emissions from manure management. Nitrous oxide emissions have remained fairly steady since 1990. Small changes in N₂O emissions from individual animal groups exhibit the same trends as the animal group populations, with the overall net effect that N₂O emissions showed a 24 percent increase from 1990 to 2010 and a less than 1 percent increase from 2009 through 2010.

Table 6-6 and Table 6-7 provide estimates of CH₄ and N₂O emissions from manure management by animal category.

Table 6-6: CH₄ and N₂O Emissions from Manure Management (Tg CO₂ Eq.)

Gas/Animal Type	1990	2005	2006	2007	2008	2009	2010
CH₄^a	31.7	47.9	48.4	52.7	51.8	50.7	52.0
Dairy Cattle	12.6	22.4	23.1	25.7	26.0	25.9	26.0
Beef Cattle	2.7	2.8	2.9	2.9	2.8	2.7	2.8
Swine	13.1	19.2	18.9	20.6	19.7	18.8	19.9
Sheep	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Goats	+	+	+	+	+	+	+
Poultry	2.8	2.7	2.7	2.8	2.7	2.7	2.7
Horses	0.5	0.6	0.6	0.6	0.5	0.5	0.5
N₂O^b	14.8	17.6	18.4	18.5	18.3	18.2	18.3
Dairy Cattle	5.3	5.7	5.8	5.9	5.8	5.8	5.9
Beef Cattle	6.5	7.8	8.3	8.2	8.1	8.1	8.2
Swine	1.2	1.8	1.8	2.0	2.0	2.0	1.9
Sheep	0.1	0.4	0.4	0.4	0.4	0.3	0.3
Goats	+	+	+	+	+	+	+
Poultry	1.5	1.7	1.7	1.7	1.7	1.6	1.6
Horses	0.2	0.3	0.3	0.3	0.3	0.3	0.3

Total	46.5	65.5	66.7	71.1	70.0	68.9	70.4
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+ Less than 0.05 Tg CO₂ Eq.

^aAccounts for CH₄ reductions due to capture and destruction of CH₄ at facilities using anaerobic digesters.

^bIncludes both direct and indirect N₂O emissions.

Note: Totals may not sum due to independent rounding.

Table 6-7: CH₄ and N₂O Emissions from Manure Management (Gg)

Gas/Animal Type	1990	2005	2006	2007	2008	2009	2010
CH₄^a	1,511	2,280	2,303	2,508	2,465	2,416	2,478
Dairy Cattle	599	1069	1101	1224	1238	1233	1239
Beef Cattle	128	135	138	136	132	131	134
Swine	624	914	901	982	938	896	948
Sheep	7	3	3	3	3	3	3
Goats	1	1	1	1	1	1	1
Poultry	131	129	131	134	129	128	129
Horses	22	28	27	27	24	24	24
N₂O^b	48	57	59	60	59	59	59
Dairy Cattle	17	18	19	19	19	19	19
Beef Cattle	21	25	27	27	26	26	27
Swine	4	6	6	6	6	6	6
Sheep	+	1	1	1	1	1	1
Goats	+	+	+	+	+	+	+
Poultry	5	5	5	5	5	5	5
Horses	1	1	1	1	1	1	1

+ Less than 0.5 Gg.

^aAccounts for CH₄ reductions due to capture and destruction of CH₄ at facilities using anaerobic digesters.

^bIncludes both direct and indirect N₂O emissions.

Note: Totals may not sum due to independent rounding.

Methodology

The methodologies presented in IPCC (2006) form the basis of the CH₄ and N₂O emission estimates for each animal type. This section presents a summary of the methodologies used to estimate CH₄ and N₂O emissions from manure management. See Annex 3.10 for more detailed information on the methodology and data used to calculate CH₄ and N₂O emissions from manure management.

Methane Calculation Methods

The following inputs were used in the calculation of CH₄ emissions:

- Animal population data (by animal type and state);
- Typical animal mass (TAM) data (by animal type);
- Portion of manure managed in each waste management system (WMS), by state and animal type;
- Volatile solids (VS) production rate (by animal type and state or United States);
- Methane producing potential (B₀) of the volatile solids (by animal type); and
- Methane conversion factors (MCF), the extent to which the CH₄ producing potential is realized for each type of WMS (by state and manure management system, including the impacts of any biogas collection efforts).

Methane emissions were estimated by first determining activity data, including animal population, TAM, WMS usage, and waste characteristics. The activity data sources are described below:

- Annual animal population data for 1990 through 2010 for all livestock types, except horses and goats were obtained from USDA NASS. For cattle, the USDA populations were utilized in conjunction with birth rates, detailed feedlot placement information, and slaughter weight data to create the transition matrix in the

CEFM that models cohorts of individual animal types and their specific emission profiles. The key variables tracked for each of the cattle population categories are described in Section 6.1 and in more detail in Annex 3.9. Horse population data were obtained from the FAOSTAT database (FAO 2010). Goat population data for 1992, 1997, 2002, and 2007 were obtained from the *Census of Agriculture* (USDA 2009a).

- The TAM is an annual average weight which was obtained for animal types other than cattle from information in USDA's *Agricultural Waste Management Field Handbook* (USDA 1996a), the American Society of Agricultural Engineers, Standard D384.1 (ASAE 1999) and others (EPA 1992, Safley 2000, ERG 2010a). For a description of the TAM used for cattle, please see section 6.1, Enteric Fermentation.
- WMS usage was estimated for swine and dairy cattle for different farm size categories using data from USDA (USDA 1996b, 1998b, 2000a, 2009a) and EPA (ERG 2000a, EPA 2002a, 2002b). For beef cattle and poultry, manure management system usage data were not tied to farm size but were based on other data sources (ERG 2000a, USDA 2000b, UEP 1999). For other animal types, manure management system usage was based on previous estimates (EPA 1992).
- VS production rates for all cattle except for bulls and calves were calculated by head for each state and animal type in the CEFM. VS production rates by animal mass for all other animals were determined using data from USDA's *Agricultural Waste Management Field Handbook* (USDA 1996a, 2008 and ERG 2010b and 2010c) and data from the American Society of Agricultural Engineers, Standard D384.1 (ASAE 1998).
- The maximum CH₄ producing capacity of the VS (B₀) was determined for each animal type based on literature values (Morris 1976, Bryant et al, 1976, Hashimoto 1981, Hashimoto 1984, EPA 1992, Hill 1982, and Hill 1984).
- MCFs for dry systems were set equal to default IPCC factors based on state climate for each year (IPCC 2006). MCFs for liquid/slurry, anaerobic lagoon, and deep pit systems were calculated based on the forecast performance of biological systems relative to temperature changes as predicted in the van't Hoff-Arrhenius equation which is consistent with IPCC (2006) Tier 2 methodology.
- Anaerobic digestion system data were obtained from the EPA AgSTAR Program, including information presented in the *AgSTAR Digest* (EPA 2000, 2003, 2006) and the AgSTAR project database (EPA 2011). Anaerobic digester emissions were calculated based on estimated methane production and collection and destruction efficiency assumptions (ERG 2008).

To estimate CH₄ emissions for cattle, the estimated amount of VS (kg per animal-year) managed in each WMS for each animal type, state, and year were taken from the CEFM. For animals other than cattle, the annual amount of VS (kg per year) from manure excreted in each WMS was calculated for each animal type, state, and year. This calculation multiplied the animal population (head) by the VS excretion rate (kg VS per 1,000 kg animal mass per day), the TAM (kg animal mass per head) divided by 1,000, the WMS distribution (percent), and the number of days per year (365.25).

The estimated amount of VS managed in each WMS was used to estimate the CH₄ emissions (kg CH₄ per year) from each WMS. The amount of VS (kg per year) were multiplied by the maximum CH₄ producing capacity of the VS (B₀) (m³ CH₄ per kg VS), the MCF for that WMS (percent), and the density of CH₄ (kg CH₄ per m³ CH₄). The CH₄ emissions for each WMS, state, and animal type were summed to determine the total U.S. CH₄ emissions.

Nitrous Oxide Calculation Methods

The following inputs were used in the calculation of direct and indirect N₂O emissions:

- Animal population data (by animal type and state);
- TAM data (by animal type);
- Portion of manure managed in each WMS (by state and animal type);
- Total Kjeldahl N excretion rate (N_{ex});
- Direct N₂O emission factor (EF_{WMS});
- Indirect N₂O emission factor for volatilization (EF_{volatilization});
- Indirect N₂O emission factor for runoff and leaching (EF_{runoff/leach});
- Fraction of N loss from volatilization of NH₃ and NO_x (Frac_{gas}); and
- Fraction of N loss from runoff and leaching (Frac_{runoff/leach}).

N₂O emissions were estimated by first determining activity data, including animal population, TAM, WMS usage, and waste characteristics. The activity data sources (except for population, TAM, and WMS, which were described above) are described below:

- Nex rates for all cattle except for bulls and calves were calculated by head for each state and animal type in the CEFM. Nex rates by animal mass for all other animals were determined using data from USDA's *Agricultural Waste Management Field Handbook* (USDA 1996a, 2008 and ERG 2010b and 2010c) and data from the American Society of Agricultural Engineers, Standard D384.1 (ASAE 1998).
- All N₂O emission factors (direct and indirect) were taken from IPCC (2006).
- Country-specific estimates for the fraction of N loss from volatilization (Frac_{gas}) and runoff and leaching ($\text{Frac}_{\text{runoff/leach}}$) were developed. Frac_{gas} values were based on WMS-specific volatilization values as estimated from EPA's *National Emission Inventory - Ammonia Emissions from Animal Agriculture Operations* (EPA 2005). $\text{Frac}_{\text{runoff/leach}}$ values were based on regional cattle runoff data from EPA's Office of Water (EPA 2002b; see Annex 3.1).

To estimate N₂O emissions for cattle, the estimated amount of N excreted (kg per animal-year) managed in each WMS for each animal type, state, and year were taken from the CEFM. For animals other than cattle, the amount of N excreted (kg per year) in manure in each WMS for each animal type, state, and year was calculated. The population (head) for each state and animal was multiplied by TAM (kg animal mass per head) divided by 1,000, the nitrogen excretion rate (Nex, in kg N per 1000 kg animal mass per day), WMS distribution (percent), and the number of days per year.

Direct N₂O emissions were calculated by multiplying the amount of N excreted (kg per year) in each WMS by the N₂O direct emission factor for that WMS (EF_{WMS} , in kg N₂O-N per kg N) and the conversion factor of N₂O-N to N₂O. These emissions were summed over state, animal, and WMS to determine the total direct N₂O emissions (kg of N₂O per year).

Next, indirect N₂O emissions from volatilization (kg N₂O per year) were calculated by multiplying the amount of N excreted (kg per year) in each WMS by the fraction of N lost through volatilization (Frac_{tas}) divided by 100, and the emission factor for volatilization ($\text{EF}_{\text{volatilization}}$, in kg N₂O per kg N), and the conversion factor of N₂O-N to N₂O. Indirect N₂O emissions from runoff and leaching (kg N₂O per year) were then calculated by multiplying the amount of N excreted (kg per year) in each WMS by the fraction of N lost through runoff and leaching ($\text{Frac}_{\text{runoff/leach}}$) divided by 100, and the emission factor for runoff and leaching ($\text{EF}_{\text{runoff/leach}}$, in kg N₂O per kg N), and the conversion factor of N₂O-N to N₂O. The indirect N₂O emissions from volatilization and runoff and leaching were summed to determine the total indirect N₂O emissions.

The direct and indirect N₂O emissions were summed to determine total N₂O emissions (kg N₂O per year).

Uncertainty and Time-Series Consistency

An analysis (ERG 2003) was conducted for the manure management emission estimates presented in the 1990 through 2001 Inventory report to determine the uncertainty associated with estimating CH₄ and N₂O emissions from livestock manure management. The quantitative uncertainty analysis for this source category was performed in 2002 through the IPCC-recommended Tier 2 uncertainty estimation methodology, the Monte Carlo Stochastic Simulation technique. The uncertainty analysis was developed based on the methods used to estimate CH₄ and N₂O emissions from manure management systems. A normal probability distribution was assumed for each source data category. The series of equations used were condensed into a single equation for each animal type and state. The equations for each animal group contained four to five variables around which the uncertainty analysis was performed for each state. These uncertainty estimates were directly applied to the 2010 emission estimates.

The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 6-8. Manure management CH₄ emissions in 2010 were estimated to be between 42.7 and 62.5 Tg CO₂ Eq. at a 95 percent confidence level, which indicates a range of 18 percent below to 20 percent above the actual 2010 emission estimate of 52.0 Tg CO₂ Eq. At the 95 percent confidence level, N₂O emissions were estimated to be between 15.4 and 22.7 Tg CO₂ Eq. (or approximately 16 percent below and 24 percent above the actual 2010 emission estimate of 18.3 Tg CO₂ Eq.).

Table 6-8: Tier 2 Quantitative Uncertainty Estimates for CH₄ and N₂O (Direct and Indirect) Emissions from Manure Management (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^a			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Manure Management	CH ₄	52.0	42.7	62.5	-18%	+20%
Manure Management	N ₂ O	18.3	15.4	22.7	-16%	+24%

^aRange of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

QA/QC and Verification

Tier 1 and Tier 2 QA/QC activities were conducted consistent with the U.S. QA/QC plan. Tier 2 activities focused on comparing estimates for the previous and current inventories for N₂O emissions from managed systems and CH₄ emissions from livestock manure. All errors identified were corrected. Order of magnitude checks were also conducted, and corrections made where needed. Manure N data were checked by comparing state-level data with bottom up estimates derived at the county level and summed to the state level. Similarly, a comparison was made by animal and WMS type for the full time series, between national level estimates for N excreted and the sum of county estimates for the full time series.

The U.S. specific values for TAM, Nex, VS, B₀, MCF, and the resulting implied emission factors were also compared to the IPCC default values. Although significant differences exist in some instances, these differences are due to the use of U.S. specific data and the differences in U.S. agriculture as compared to other countries. The U.S. manure management emission estimates use the most reliable country-specific data, which are more representative of U.S. animals and systems than the IPCC default values. For example, the U.S. implied CH₄ emission factor for dairy cattle is significantly higher than the IPCC default implied CH₄ emission factor. This is because U.S. dairy manure is most commonly managed in liquid systems, which produce more CH₄.

Recalculations Discussion

The CEFM produces population, VS and Nex data for cattle that are used in the manure management inventory. As a result, all changes to the CEFM described in Section 6.1 Enteric Fermentation contributed to changes in the population, VS and Nex data used for calculating CH₄ and N₂O cattle emissions from manure management.

Data from the 2007 *Census of Agriculture* were incorporated into the inventory. Census farm size distribution data were used to update the WMS distributions for dairy and swine in 2007. The dairy and swine WMS distributions between 2002 and 2007 were extrapolated based on the 2002 and 2007 data; WMS distributions after 2007 were assumed to be equal to 2007 values. The dairy and swine WMS update caused changes in dairy and swine emission estimates from 2003 on.

In addition, census county-level population data were used to update the county-level population estimates. These estimates are used as input to the Agricultural Soil Management calculations, and to determine population-weighted state temperatures which are used to calculate MCFs for liquid systems. The county-level population update caused minor changes in methane emissions for all animals throughout the timeseries.

State animal populations were updated to reflect updated USDA NASS datasets. Population changes occurred for all animals in 2009. Sheep populations experienced changes in 2004 to 2006 and 2008 estimates due to a change in the “other states” reported by USDA NASS.

Temperature data were updated to incorporate the most recent available data. The temperature data are used to estimate MCFs for liquid systems; this update caused minor changes in CH₄ emission estimates from dairy, swine, beef, and poultry from 2007 to 2009.

Planned Improvements

Tier 1 emission estimates for mules, donkeys, burros, and American bison will be incorporated into future

inventories. Although these animal groups are considered very minor sources of emissions and will not contribute significantly to the overall U.S. emissions from manure management, they will be included for completeness and consistency across source categories.

The uncertainty analysis will be updated in the future to more accurately assess uncertainty of emission calculations. This update is necessary due to the extensive changes in emission calculation methodology, including estimation of emissions at the WMS level and the use of new calculations and variables for indirect N₂O emissions.

6.3. Rice Cultivation (IPCC Source Category 4C)

Most of the world's rice, and all rice in the United States, is grown on flooded fields. When fields are flooded, aerobic decomposition of organic material gradually depletes most of the oxygen present in the soil, causing anaerobic soil conditions. Once the environment becomes anaerobic, CH₄ is produced through anaerobic decomposition of soil organic matter by methanogenic bacteria. As much as 60 to 90 percent of the CH₄ produced is oxidized by aerobic methanotrophic bacteria in the soil (some oxygen remains at the interfaces of soil and water, and soil and root system) (Holzapfel-Pschorn et al. 1985, Sass et al. 1990). Some of the CH₄ is also leached away as dissolved CH₄ in floodwater that percolates from the field. The remaining un-oxidized CH₄ is transported from the submerged soil to the atmosphere primarily by diffusive transport through the rice plants. Minor amounts of CH₄ also escape from the soil via diffusion and bubbling through floodwaters.

The water management system under which rice is grown is one of the most important factors affecting CH₄ emissions. Upland rice fields are not flooded, and therefore are not believed to produce CH₄. In deepwater rice fields (i.e., fields with flooding depths greater than one meter), the lower stems and roots of the rice plants are dead, so the primary CH₄ transport pathway to the atmosphere is blocked. The quantities of CH₄ released from deepwater fields, therefore, are believed to be significantly less than the quantities released from areas with shallower flooding depths. Some flooded fields are drained periodically during the growing season, either intentionally or accidentally. If water is drained and soils are allowed to dry sufficiently, CH₄ emissions decrease or stop entirely. This is due to soil aeration, which not only causes existing soil CH₄ to oxidize but also inhibits further CH₄ production in soils. All rice in the United States is grown under continuously flooded conditions; none is grown under deepwater conditions. Mid-season drainage does not occur except by accident (e.g., due to levee breach).

Other factors that influence CH₄ emissions from flooded rice fields include fertilization practices (especially the use of organic fertilizers), soil temperature, soil type, rice variety, and cultivation practices (e.g., tillage, seeding, and weeding practices). The factors that determine the amount of organic material available to decompose (i.e., organic fertilizer use, soil type, rice variety,¹⁷¹ and cultivation practices) are the most important variables influencing the amount of CH₄ emitted over the growing season; the total amount of CH₄ released depends primarily on the amount of organic substrate available. Soil temperature is known to be an important factor regulating the activity of methanogenic bacteria, and therefore the rate of CH₄ production. However, although temperature controls the amount of time it takes to convert a given amount of organic material to CH₄, that time is short relative to a growing season, so the dependence of total emissions over an entire growing season on soil temperature is weak. The application of synthetic fertilizers has also been found to influence CH₄ emissions; in particular, both nitrate and sulfate fertilizers (e.g., ammonium nitrate and ammonium sulfate) appear to inhibit CH₄ formation.

Rice is cultivated in eight states: Arkansas, California, Florida, Louisiana, Mississippi, Missouri, Oklahoma, and Texas.¹⁷² Soil types, rice varieties, and cultivation practices for rice vary from state to state, and even from farm to farm. However, most rice farmers apply organic fertilizers in the form of residue from the previous rice crop, which is left standing, disked, or rolled into the fields. Most farmers also apply synthetic fertilizer to their fields, usually urea. Nitrate and sulfate fertilizers are not commonly used in rice cultivation in the United States. In addition, the climatic conditions of southwest Louisiana, Texas, and Florida often allow for a second, or ratoon, rice crop. Ratoon crops are much less common or non-existent in Arkansas, California, Mississippi, Missouri, Oklahoma, and northern areas of Louisiana. Methane emissions from ratoon crops have been found to be considerably higher than those from the primary crop. This second rice crop is produced from regrowth of the stubble after the first crop has been

¹⁷¹ The roots of rice plants shed organic material, which is referred to as "root exudate." The amount of root exudate produced by a rice plant over a growing season varies among rice varieties.

¹⁷² A very small amount of rice is grown on about 20 acres in South Carolina; however, this amount was determined to be too insignificant to warrant inclusion in national emission estimates.

harvested. Because the first crop's stubble is left behind in ratooned fields, and there is no time delay between cropping seasons (which would allow the stubble to decay aerobically), the amount of organic material that is available for anaerobic decomposition is considerably higher than with the first (i.e., primary) crop.

Rice cultivation is a small source of CH₄ in the United States (Table 6-9 and Table 6-10). In 2010, CH₄ emissions from rice cultivation were 8.6 Tg CO₂ Eq. (410 Gg). Annual emissions fluctuated unevenly between the years 1990 and 2010, ranging from an annual decrease of 14 percent to an annual increase of 17 percent. There was an overall decrease of 17 percent between 1990 and 2006, due to an overall decrease in primary crop area.¹⁷³ However, emission levels increased again by 45 percent between 2006 and 2010 due to an increase in rice crop area in all states except Oklahoma, which reported no rice production in 2009 and 2010. The factors that affect the rice acreage in any year vary from state to state, although the price of rice relative to competing crops is the primary controlling variable in most states.

Table 6-9: CH₄ Emissions from Rice Cultivation (Tg CO₂ Eq.)

State	1990		2005	2006	2007	2008	2009	2010
Primary	5.1		6.0	5.1	4.9	5.3	5.6	6.5
Arkansas	2.1		2.9	2.5	2.4	2.5	2.6	3.2
California	0.7		0.9	0.9	1.0	0.9	1.0	1.0
Florida	+		+	+	+	+	+	+
Louisiana	1.0		0.9	0.6	0.7	0.8	0.8	1.0
Mississippi	0.4		0.5	0.3	0.3	0.4	0.4	0.5
Missouri	0.1		0.4	0.4	0.3	0.4	0.4	0.4
Oklahoma	+		+	+	+	+	+	+
Texas	0.6		0.4	0.3	0.3	0.3	0.3	0.3
Ratoon	2.1		0.8	0.9	1.3	1.9	1.8	2.1
Arkansas	+		+	+	+	+	+	+
Florida	+		+	+	+	+	+	+
Louisiana	1.1		0.5	0.5	0.9	1.2	1.1	1.4
Texas	0.9		0.4	0.4	0.3	0.6	0.7	0.7
Total	7.1		6.8	5.9	6.2	7.2	7.3	8.6

+ Less than 0.05 Tg CO₂ Eq.

Note: Totals may not sum due to independent rounding.

Table 6-10: CH₄ Emissions from Rice Cultivation (Gg)

State	1990		2005	2006	2007	2008	2009	2010
Primary	241		287	241	235	254	265	308
Arkansas	102		139	119	113	119	125	152
California	34		45	44	45	44	47	47
Florida	1		1	1	1	1	1	1
Louisiana	46		45	29	32	39	39	45
Mississippi	21		22	16	16	19	21	26
Missouri	7		18	18	15	17	17	21
Oklahoma	+		+	+	+	+	+	+
Texas	30		17	13	12	15	14	16
Ratoon	98		39	41	60	89	84	101
Arkansas	+		1	+	+	+	+	+
Florida	2		+	1	1	1	2	2
Louisiana	52		22	22	42	59	51	68

¹⁷³ The 14 percent decrease occurred between 2005 and 2006; the 17 percent increase happened between 1993 and 1994.

Texas	45		17	18	16	29	31	32
Total	339		326	282	295	343	349	410

+ Less than 0.5 Gg

Note: Totals may not sum due to independent rounding.

Methodology

IPCC (2006) recommends using harvested rice areas, area-based daily emission factors (i.e., amount of CH₄ emitted per day per unit harvested area), and length of growing season to estimate annual CH₄ emissions from rice cultivation. To that end, the recommended methodology and Tier 2 U.S.-specific emission factors derived from rice field measurements were used. Average U.S. seasonal emission factors were applied since state-specific and daily emission factors were not available. Seasonal emissions have been found to be much higher for ratooned crops than for primary crops, so emissions from ratooned and primary areas are estimated separately using emission factors that are representative of the particular growing season. This approach is consistent with IPCC (2006).

The harvested rice areas for the primary and ratoon crops in each state are presented in Table 6-11, and the area of ratoon crop area as a percent of primary crop area is shown in Table 6-12. Primary crop areas for 1990 through 2010 for all states except Florida and Oklahoma were taken from U.S. Department of Agriculture's *Field Crops Final Estimates 1987–1992* (USDA 1994), *Field Crops Final Estimates 1992–1997* (USDA 1998), *Field Crops Final Estimates 1997–2002* (USDA 2003), and *Crop Production Summary* (USDA 2005 through 2011). Source data for non-USDA sources of primary and ratoon harvest areas are shown in Table 6-13. California, Mississippi, Missouri, and Oklahoma have not ratooned rice over the period 1990 through 2010 (Guethle 1999 through 2010; Lee 2003 through 2007; Mutters 2002 through 2005; Street 1999 through 2003; Walker 2005, 2007 through 2008; Buehring 2009 through 2011).

Table 6-11: Rice Areas Harvested (Hectares)

State/Crop	1990	2005	2006	2007	2008	2009	2010
Arkansas							
Primary	485,633	661,675	566,572	536,220	564,549	594,901	722,380
Ratoon ^a	-	662	6	5	6	6	7
California	159,854	212,869	211,655	215,702	209,227	225,010	223,796
Florida							
Primary	4,978	4,565	4,575	6,242	5,463	5,664	5,330
Ratoon	2,489	+	1,295	1,873	1,639	2,266	2,275
Louisiana							
Primary	220,558	212,465	139,620	152,975	187,778	187,778	216,512
Ratoon	66,168	27,620	27,924	53,541	75,111	65,722	86,605
Mississippi	101,174	106,435	76,487	76,487	92,675	98,341	122,622
Missouri	32,376	86,605	86,605	72,036	80,534	80,939	101,578
Oklahoma	617	271	17	+	77	+	+
Texas							
Primary	142,857	81,344	60,704	58,681	69,607	68,798	76,083
Ratoon	57,143	21,963	23,675	21,125	36,892	39,903	41,085
Total Primary	1,148,047	1,366,228	1,146,235	1,118,343	1,209,911	1,261,431	1,468,300
Total Ratoon	125,799	50,245	52,899	76,544	113,648	107,897	129,971
Total	1,273,847	1,416,473	1,199,135	1,194,887	1,323,559	1,369,328	1,598,271

^a Arkansas ratooning occurred only in 1998, 1999, and 2005 through 2010.

+ Emissions are less than 0.1 Tg CO₂ Eq.

- No reported value

Note: Totals may not sum due to independent rounding.

Table 6-12: Ratooned Area as Percent of Primary Growth Area

State	1990	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Arkansas	0%		+	+			0%			0.1%	+	+	+	+	+
Florida	50%			65%	41%	60%	54%	100%	77%	0%	28%	30%	30%	40%	43%
Louisiana	30%			40%	30%	15%	35%	30%	13%	20%	35%	40%	35%	40%	
Texas	40%			50%	40%	37%	38%	35%	27%	39%	36%	53%	58%	54%	

+ Indicates ratooning rate less than 0.1 percent.

Table 6-13: Non-USDA Data Sources for Rice Harvest Information

State/Crop	1990		2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Arkansas													
Ratoon	Wilson (2002 – 2007, 2009 – 2011)												
Florida													
Primary	Scheuneman (1999 – 2001)		Deren (2002)	Kirstein (2003, 2006)				Gonzales (2006 – 2011)					
Ratoon	Scheuneman (1999)		Deren (2002)	Kirstein (2003-2004)		Cantens (2005)		Gonzales (2006 – 2011)					
Louisiana													
Ratoon	Bollich (2000)		Linscombe (1999, 2001 – 2011)										
Oklahoma													
Primary	Lee (2003-2007)									Anderson (2008 – 2011)			
Texas													
Ratoon	Klosterboer (1999 – 2003)				Stansel (2004 – 2005)			Texas Ag Experiment Station (2006 – 2011)					

To determine what CH₄ emission factors should be used for the primary and ratoon crops, CH₄ flux information from rice field measurements in the United States was collected. Experiments that involved atypical or nonrepresentative management practices (e.g., the application of nitrate or sulfate fertilizers, or other substances believed to suppress CH₄ formation), as well as experiments in which measurements were not made over an entire flooding season or floodwaters were drained mid-season, were excluded from the analysis. The remaining experimental results¹⁷⁴ were then sorted by season (i.e., primary and ratoon) and type of fertilizer amendment (i.e., no fertilizer added, organic fertilizer added, and synthetic and organic fertilizer added). The experimental results from primary crops with added synthetic and organic fertilizer (Bossio et al. 1999; Cicerone et al. 1992; Sass et al. 1991a, 1991b) were averaged to derive an emission factor for the primary crop, and the experimental results from ratoon crops with added synthetic fertilizer (Lindau and Bollich 1993, Lindau et al. 1995) were averaged to derive an emission factor for the ratoon crop. The resultant emission factor for the primary crop is 210 kg CH₄/hectare-season, and the resultant emission factor for the ratoon crop is 780 kg CH₄/hectare-season.

Uncertainty and Time-Series Consistency

The largest uncertainty in the calculation of CH₄ emissions from rice cultivation is associated with the emission factors. Seasonal emissions, derived from field measurements in the United States, vary by more than one order of magnitude. This inherent variability is due to differences in cultivation practices, particularly fertilizer type, amount, and mode of application; differences in cultivar type; and differences in soil and climatic conditions. A portion of this variability is accounted for by separating primary from ratooned areas. However, even within a cropping season or a given management regime, measured emissions may vary significantly. Of the experiments

¹⁷⁴ In some of these remaining experiments, measurements from individual plots were excluded from the analysis because of the aforementioned reasons. In addition, one measurement from the ratooned fields (i.e., the flux of 1,490 kg CH₄/hectare-season in Lindau and Bollich 1993) was excluded, because this emission rate is unusually high compared to other flux measurements in the United States, as well as IPCC (2006) default emission factors.

used to derive the emission factors applied here, primary emissions ranged from 22 to 479 kg CH₄/hectare-season and ratoon emissions ranged from 481 to 1,490 kg CH₄/hectare-season. The uncertainty distributions around the primary and ratoon emission factors were derived using the distributions of the relevant primary or ratoon emission factors available in the literature and described above. Variability about the rice emission factor means was not normally distributed for either primary or ratooned crops, but rather skewed, with a tail trailing to the right of the mean. A lognormal statistical distribution was, therefore, applied in the Tier 2 Monte Carlo analysis.

Other sources of uncertainty include the primary rice-cropped area for each state, percent of rice-cropped area that is ratooned, and the extent to which flooding outside of the normal rice season is practiced. Expert judgment was used to estimate the uncertainty associated with primary rice-cropped area for each state at 1 to 5 percent, and a normal distribution was assumed. Uncertainties were applied to ratooned area by state, based on the level of reporting performed by the state. No uncertainty estimates were calculated for the practice of flooding outside of the normal rice season because CH₄ flux measurements have not been undertaken over a sufficient geographic range or under a broad enough range of representative conditions to account for this source in the emission estimates or its associated uncertainty.

To quantify the uncertainties for emissions from rice cultivation, a Monte Carlo (Tier 2) uncertainty analysis was performed using the information provided above. The results of the Tier 2 quantitative uncertainty analysis are summarized in Table 6-14. Rice cultivation CH₄ emissions in 2010 were estimated to be between 3.0 and 21.8 Tg CO₂ Eq. at a 95 percent confidence level, which indicates a range of 65 percent below to 153 percent above the actual 2010 emission estimate of 8.6 Tg CO₂ Eq.

Table 6-14: Tier 2 Quantitative Uncertainty Estimates for CH₄ Emissions from Rice Cultivation (Tg CO₂ Eq. and Percent)

Source	Gas	2010 Emission Estimate (Tg CO ₂ Eq.)	Uncertainty Range Relative to Emission Estimate ^a			
			(Tg CO ₂ Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Rice Cultivation	CH ₄	8.6	3.0	21.8	-65%	+153%

^a Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure time-series consistency from 1990 through 2010. Details on the emission trends through time are described in more detail in the Methodology section, above.

QA/QC and Verification

A source-specific QA/QC plan for rice cultivation was developed and implemented. This effort included a Tier 1 analysis, as well as portions of a Tier 2 analysis. The Tier 2 procedures focused on comparing trends across years, states, and cropping seasons to attempt to identify any outliers or inconsistencies. No problems were found.

Planned Improvements

A possible future improvement is to create region-specific emission factors for rice cultivation. The current methodology uses a nationwide average emission factor, derived from several studies done in a number of states. The prospective improvement would take the same studies and average them by region, presumably resulting in more spatially specific emission factors.

6.4. Agricultural Soil Management (IPCC Source Category 4D)

Nitrous oxide is produced naturally in soils through the microbial processes of nitrification and denitrification.¹⁷⁵ A

¹⁷⁵ Nitrification and denitrification are driven by the activity of microorganisms in soils. Nitrification is the aerobic microbial oxidation of ammonium (NH₄⁺) to nitrate (NO₃⁻), and denitrification is the anaerobic microbial reduction of nitrate to N₂. Nitrous

number of agricultural activities increase mineral N availability in soils, thereby increasing the amount available for nitrification and denitrification, and ultimately the amount of N₂O emitted. These activities increase soil mineral N either directly or indirectly (see Figure 6-2). Direct increases occur through a variety of management practices that add or lead to greater release of mineral N to the soil, including fertilization; application of managed livestock manure and other organic materials such as sewage sludge; deposition of manure on soils by domesticated animals in pastures, rangelands, and paddocks (PRP) (i.e., by grazing animals and other animals whose manure is not managed); production of N-fixing crops and forages; retention of crop residues; and drainage and cultivation of organic cropland soils (i.e., soils with a high organic matter content, otherwise known as histosols).¹⁷⁶ Other agricultural soil management activities, including irrigation, drainage, tillage practices, and fallowing of land, can influence N mineralization in soils and thereby affect direct emissions. Mineral N is also made available in soils through decomposition of soil organic matter and plant litter, as well as asymbiotic fixation of N from the atmosphere,¹⁷⁷ and these processes are influenced by agricultural management through impacts on moisture and temperature regimes in soils. These additional sources of mineral N are included at the recommendation of IPCC (2006) for complete accounting of management impacts on greenhouse gas emissions, as discussed in the Methodology section. Indirect emissions of N₂O occur through two pathways: (1) volatilization and subsequent atmospheric deposition of applied/mineralized N,¹⁷⁸ and (2) surface runoff and leaching of applied/mineralized N into groundwater and surface water. Direct emissions from agricultural lands (i.e., cropland and grassland) are included in this section, while direct emissions from forest lands and settlements are presented in the Land Use, Land-Use Change, and Forestry chapter. However, indirect N₂O emissions from all land-uses (cropland, grassland, forest lands, and settlements) are reported in this section.

Figure 6-2: Sources and Pathways of N that Result in N₂O Emissions from Agricultural Soil Management

Agricultural soils produce the majority of N₂O emissions in the United States. Estimated emissions from this source in 2010 were 207.8 Tg CO₂ Eq. (670 Gg N₂O) (see Table 6-15 and Table 6-16). Annual N₂O emissions from agricultural soils fluctuated between 1990 and 2010, although overall emissions were almost 4 percent higher in 2010 than in 1990. Year-to-year fluctuations are largely a reflection of annual variation in weather patterns, synthetic fertilizer use, and crop production. On average, cropland accounted for approximately 70 percent of total direct emissions, while grassland accounted for approximately 30 percent. These percentages are about the same for indirect emissions since forest lands and settlements account for such a small percentage of total indirect emissions. Estimated direct and indirect N₂O emissions by sub-source category are shown in Table 6-17 and Table 6-18.

Table 6-15: N₂O Emissions from Agricultural Soils (Tg CO₂ Eq.)

Activity	1990	2005	2006	2007	2008	2009	2010
Direct	155.8	169.1	165.6	166.8	168.5	162.2	162.3
Cropland	103.0	118.0	115.7	117.8	118.0	112.4	112.4
Grassland	52.8	51.1	49.9	49.0	50.5	49.9	49.9
Indirect (All Land- Use Types)	44.1	43.9	45.5	44.3	44.4	45.0	45.5
Cropland	37.4	36.7	38.7	37.5	37.6	37.9	38.5
Grassland	6.4	6.5	6.0	6.1	6.1	6.4	6.4
Forest	+	0.1	0.1	0.1	0.1	0.1	0.1

oxide is a gaseous intermediate product in the reaction sequence of denitrification, which leaks from microbial cells into the soil and then into the atmosphere. Nitrous oxide is also produced during nitrification, although by a less well-understood mechanism (Nevison 2000).

¹⁷⁶ Drainage and cultivation of organic soils in former wetlands enhances mineralization of N-rich organic matter, thereby increasing N₂O emissions from these soils.

¹⁷⁷ Asymbiotic N fixation is the fixation of atmospheric N₂ by bacteria living in soils that do not have a direct relationship with plants.

¹⁷⁸ These processes entail volatilization of applied or mineralized N as NH₃ and NO_x, transformation of these gases within the atmosphere (or upon deposition), and deposition of the N primarily in the form of particulate NH₄⁺, nitric acid (HNO₃), and NO_x.