Anthropogenic Sulfur Dioxide Emissions: 1850-2005

Supplementary Material

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S.1 Region Definitions

Unless specified otherwise, the region definitions used in this work, where some ambiguity might exist, are: Western Europe (OECD Europe as of 1990, including Turkey), Eastern Europe (including Albania and the countries of the former Yugoslavia), the Former Soviet Union (including Moldova, Estonia, Latvia, and Lithuania), and China+ (includes Cambodia, Hong Kong, North Korea, Mongolia, and Vietnam).

S.2 Composite energy demand

A composite time series of fossil fuels used for combustion was constructed by combining data from a number of sources. For the most recent years, IEA (2006) detailed energy balance tables were used to provide fossil fuel combustion quantities by weight for each country. These data generally extend from 1960 forward for OECD countries and from 1971 forward for other countries. The following categories of end-use were not included, as emissions from these non-combustion activities are included elsewhere: feedstock use, coal liquefaction, coal transformation, and non-metallic minerals (largely cement). The international bunkers consumption category was also not included since, as described in the main text, reporting here is incomplete and alternative data sources were used. Total consumption for China for 2000-2002 is described in section S.7 below.

Two primary data sets were used to supply data for earlier years: UN energy statistics (UN 1996), and the estimates from Etemad et al. (1991) as used by Andres et al. (1999). Gaps in the resulting annual time series were filled by interpolation. Consumption of coal for the production of coke, a value taken from the UN data set, was subtracted to estimate combusted coal. Data on other non-combustion uses of coal were not available, which represents a small discontinuity in the combustion coal time series; however, the major non-combustion use of coal is coking coal. Sulfur emissions from modern by-product coking plants are relatively low and are included as process emissions. Sulfur from coking coal used in earlier "beehive" ovens are assumed to be emitted at the same rate as other coal combustion (including some retention in ash). The fraction of coal used in modern coking plants is assumed to decline linearly between a starting year and end-year as indicated in Table S-1, drawing from Bond et al. (2007). Data on coking coal consumption before 1950 were not available, so combusted coal previous to 1950 was scaled by the ratio of combusted over total coal in 1950.

Region	Begin Year	End Year
OECD90 Europe	1900	1950
Eastern Europe	1910	1950
Other OECD90	1910	1950
FSU	1910	1950
China/CPA	1910	1950
Middle East	1950	1970
Africa	1950	1970
Latin America	1950	1970
Other Asia	1950	1970

Table S- 1 – Assumptions for the introduction of modern by-produce coking plants. Country-specific values were used for Germany (1880 – 1950) and UK (1890-1950). Based on Bond et al. (2007).

IEA data for petroleum consumption were scaled back to 1960 (where necessary) by using the liquid fuel-related carbon dioxide emissions estimate by country from CDIAC (Marland et al. 2008), scaled between 1960 and 1950 to match the petroleum consumption estimate of Andres et al. (1999) by 1950.

No explicit consideration of country boundary changes was made during the emissions calculation beyond that contained in the historical data sets used. During calibration with historical country-inventory values in Europe, emissions factors were frozen for previous years where large changes in area occurred in order to avoid spurious changes in emissions.

Biomass consumption for recent years was taken from the IEA energy balances, which were similar to the estimate of Fernandes et. al. (2007). The IEA values were converted to per-capita values and these were used from 1960-2005 where IEA data were lacking. Country-level per-capita estimates were developed for 1850-1900 using the regional values from Figure 7 of Fernandes et. al. (2007), adjusting individual country estimates by the wood consumption estimates of Hurt et al. (2006), while approximately matching the regional totals from Fernandes et. al. (2007). Per-capita consumption values were linearly interpolated between 1900 and 1960. The resulting total biomass consumption estimates average 7% lower than Fernandes et. al. (2007) between 1900 and 2000.

S.3 Emissions Inventories Used

Table S-2 below details the inventory data used for calibration by region and specific countries. There are several sources of data for European countries, although a consistent, composite data series for all years 1980-2005 was not available for all countries. Calibration was performed to annual inventory data from 1980-2005 where available, and to values every five years before 1980. The calibration adjustments were interpolated in-between years where inventory data were available. Emissions factors were held constant for years before the first calibration data. As described in the text, the first calibration year was chosen to minimize inconsistencies due to historical boundary changes. Consistent emissions estimates for countries in Eastern Europe, particularly Albania and the countries of the Former Yugoslavia, were developed for the key years of 1990, 2000, and 2005 by comparing existing inventory data and data for neighboring countries, and adding estimates for missing sectors where necessary. The emissions estimates of Vestreng et al. (2007) were used for countries where reported data appeared to be incomplete, particularly prior to 1990.

Region/Country	Years	Inventory
OECD Europe, Eastern/Central Europe, Poland (2004-2005), Japan, Australia, New Zealand, Lithuania, Estonia, Latvia, Ukraine (1998-2005), United States	1990 – 2005 (annual)	UNFCCC (2009)
OECD Europe, Eastern/Central Europe[a]	1980 – 1990 (annual)	EEA (2002)
Bulgaria, Cyprus, Czech republic, Greece, Italy, Luxembourg, Malta, Moldova, Norway, Romania, Slovakia, Slovenia,	Various years – 1980 through 2002	Vestreng et al. (2007)
OECD Europe, Eastern/Central Europe[b]	Various years – 1980	Mylona (1996)
Turkey	1990, 2000, 2005	GAINS (Klimont et al. 2009).
United States (see main text)	1970, 1975, 1980-1990 (annual)	US EPA (1996a)
United States (see main text)	1900-1965	Gschwandtner et al. (1986)
Canada	1985-2005	Environment Canada (2008)
Japan	1905-1990	Fujita (1993)
United Kingdom	1970-1990	UK National Atmospheric Emissions Inventory (2009)

[a] In some cases emissions estimate submitted to the UNFCCC data differed from previous EEA estimates, in which cases EEA estimates were scaled between 1985 and 1990 to match the UNFCCC data in 1990.

[b] Calibration to the Mylona (1996) estimate was used where the data appeared to be consistent with the fuel consumption and other data used, considering potential changes in country/reporting boundaries. The earliest Mylona estimate was used except for the following: Germany (1950), Austria (1920), Bulgaria (1925), Denmark (1905), Finland (1930), Greece (1885), Ireland (1935), Norway (1910), Portugal (1915), Romania (1915), Switzerland (1895).

Table S- 2 – Inventory data used for calibration of total country emissions.

A number of additional emissions estimates are available in the literature and, while these were not used for an exact calibration, these were compared to the estimates here, and emissions factors were adjusted to achieve some measure of overall consistency. For East Asia, we examined estimates from Ohara et al. (2007), Streets et al. (2003), Zhang et al. (2009), and GAINS Asia (Klimont et al. 2009). In many cases, these estimates are not consistent with each other, and determining the source of differences was beyond the scope of the present project. A comparison between the present estimate and these inventories is provided in Table S-3.

	2000					2005		
Country	Current Estimate	REAS	Trace-P	GAINS		Current Estimate	Intex-B*	GAINS
China	19,947	27,590	20,385	23,192		32,517	31,020	31,557
Taiwan	311	266	376	494		246	189	402
North Korea	390	298	227	167		301	233	121
South Korea	943	987	829	604		395	497	434
Thailand	1,004	998	961	1,016		1,272	1,299	547
Indonesia	1,012	1,078	884	825		1,535	1,451	1,071
SE Asia (Rest of)	1,244	1,577	1,475	1,346		1,716	2,729	1,058
India	5,363	6,141	5,536	5,064		6,275	5,596	5,929
Pakistan	862	1,097	1,416	928		684	2,882	1,043

* The Intex-B estimate is for 2006.

Table S- 3 – Comparison with recent inventory estimates for Asia.

We can also compare South Korea's estimates with an estimate by the National Institute of Environmental Research-Korea (NIER 2008). The 2005 estimate here was adjusted to be almost identical to the NIER estimate. The 2000 estimate here, however, is nearly twice that from NIER, but similar to estimates in Table S-3.

S.4 FSU Assumptions

Emissions from the countries of the Former Soviet Union[1] are particularly uncertain. Comprehensive country-based inventories are not produced by many of these national governments, or are available only for recent years. We used the estimate from Ryaboshapko et al. (1996) to calibrate overall emissions from FSU. Emissions from metal smelting were calculated as described in section S.6, calibrated to the Ryaboshapko et al. value for 1990. The Ryaboshapko et al. (1996) estimate for iron working, however, was not used, as it was high compared to other regions.

UNFCCC submissions from Ukraine, Estonia, Latvia, and Lithuania were used to calibrate the emissions estimate for these countries. The end result was a lower overall estimate for the FSU countries than in Smith et al. (2004).

Fossil fuel use and other driver information are generally available only for these countries after about 1990. In previous years, most compilations report values only for the Soviet Union. In order to approximate the historical distribution of emissions by country, FSU totals were distributed by country using historical population estimates of Goldewijk (2005). Overall, the emissions here are within 10% of recent GAINS estimates for this region, considering the sum of emissions where country-level data is available in both data sets.

S.5 Petroleum Mass Balance Estimate

The global petroleum mass balance was constructed by calculating the total amount of sulfur in crude oil and subtracting the amount of sulfur removed in refineries. Crude oil production was taken from the HYDE database (HYDE 2002), as compiled from Etemad et al. (1991) and Mitchell (1998a, 1998b, 1998c), supplemented with more

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[1] Armenia, Azerbaijan, Belarus, Estonia, Georgia, Kazakhstan, Kyrgyzstan, Latvia, Lithuania, Moldova, Russia, Tajikistan, Turkmenistan, Ukraine, and Uzbekistan.

recent data from EIA (2008a). Crude oil production of a given country was multiplied by the average sulfur content of crude oil from that country to estimate the total amount of sulfur in crude oil. Sulfur contents of crude oil production by country were estimated from a variety of sources (Carrales and Martin 1975; OGJ 1990; NIPER 1995; PIW 1997). In cases where a sulfur content value was not available for a country, an average value for the region was used. Values estimated for 1971 (Carrales and Martin 1975) and data used here (collected for later years) were comparable for most countries except for the United States and Canada, due to the large variety of petroleum fields and properties in these two countries. Except for the United States and Canada (see below), shifts in production within countries resulting in a change in the average sulfur content of the crude oil produced were not captured.

For the United States and Canada, the aggregate sulfur content of crude oil production was estimated by year. Crude oil production in the United States by state from 1859 through 2005 was compiled from API (1999) and EIA[2]. The sulfur content of crude oil was estimated by using an average sulfur content for production from each state, estimated using Carrales and Martin (1975). The sulfur content of offshore production from Alaskan North Slope was assumed to be 1.1%, Gulf of Mexico 1.8% (Platts 2010), and federal waters offshore California 1.1%. For a few states without sulfur content information, a default value of 0.8% was used, although this has little impact on the results.

For Canada, we estimate the time series of the aggregate crude oil sulfur content by using the split between heavy and light crude over time from the Canadian Petroleum Producers Statistical Handbook (2008). We assume that the average sulfur content of light crude is 0.4% and that of heavy crude is 2%. These values match the aggregate value derived from the more detailed data of Carrales and Martin (1975). We also assume that synthetic crude derived from tar sands is upgraded to a crude specification of 0.2%.

Data on the amount of sulfur removed at refineries from 1972 through 2005 for the US and Canada is taken from USGS Sulfur Yearbooks and US Bureau of Mines Mineral Yearbooks (1974 through 1991).[3] An additional data point for the United States for 1970 was taken from Bingham et al. (1973).

Recovered sulfur before 1970 and 1972 for the US and Canada, respectively, and before 1990 for most other regions, were reported only as totals that include sulfur recovered from natural gas processing. The amount of sulfur removed from natural gas processing is a large fraction of total sulfur removals for the United States, Canada, Western Europe, the Former Soviet Union, and the Middle East. Petroleum removals before 1970 (1972 for Canada, and 1975 for the FSU) were estimated by scaling with crude oil consumption in United States and crude oil production in Canada and the FSU. For other regions, the amount of sulfur recovered from oil refineries was estimated by subtracting the estimated natural gas sulfur recovery from the total recovered sulfur amount. An estimate of natural gas sulfur recovery was found by scaling the reported removal from

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[2] Crude Oil Production (by state), state offshore production, U.S. Field Production of Crude Oil, Crude oil and oil product imports and exports. Release date 6/29/2009. www.eia.doe.gov.

[3] minerals.usgs.gov

natural gas plants for the earliest available year, extrapolated back in time using natural gas production.

No data on sulfur removals were available before 1958, so removals were assumed to linearly go to zero by 1950, when sulfur recovery units began to be commercialized. The estimated sulfur removal from petroleum is only 10% of the global sulfur content in 1958, so this assumption has only a small impact on the total estimated sulfur balance for petroleum.

For the United States, we also constructed a national petroleum sulfur mass balance estimate from 1900-2005 that accounts for inputs and exports of petroleum products and crude oil, and net consumption of domestic production. Net consumption of domestic crude oil and net imports in tonnes was taken from EIA.[2] The sulfur content of net imports of crude oil was estimated by combining data on net sulfur imports by country for 1970, 1975, and 1979-2005 (US Census 2009; with values linearly extrapolated where data were not available) with sulfur contents of crude production from each country as discussed in the text. The sulfur content of net crude imports before 1970 was scaled with the total weight of net imports of crude oil. The sulfur content of net imports of refined petroleum products was estimated by assigning a default value for each product to import and export data covering 1949-2008 from EIA (2009). Data on product imports and exports were not available before 1949, but this component was less than 10% of total sulfur content at that time. Sulfur in bunker fuels sold in US ports was determined as described in the main text. Data on sulfur removal in refineries is taken from the sources discussed above.

Note that additional uncertainty arises in evaluating the overall petroleum mass balance because it is unclear what fraction of the emissions reported as fugitive emissions in the inventory data are from refineries (in which case they should be included in the mass balance), and what portion are from sulfur gases associated with crude oil extraction (which should not be included in the mass balance estimate).

S.6 Smelting Mass Balance Estimate

The default estimate for metal smelting emissions was calculated using a mass balance approach using data on weight of metal produced by country from a variety of sources. The primary data source was USGS Minerals Yearbooks and Bureau of Mines Minerals Yearbooks from 1932-1993 (Courtesy of the University of Wisconsin Ecology and Natural Resources Collection[4]), which provide data as early as 1926. Data from UN was also used (kindly provided by D. Stern) where these values represented smelting output. Production data for the United States were generally available from 1900 from USGS and production data for Canada from 1886 to 1890 from Statistics Canada (Leacy 1983).

Data for earlier years were compiled from a variety of sources (Adams 1900, Of 1912, Of 1913, Ingalls 1902, Mulhall 1892, Read 1914, Butts 1922, Stevens 1907, Stevens 1904, Weed 1918). Where annual data were not available, estimates were linearly interpolated between years with data.

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[4] http://digicoll.library.wisc.edu/EcoNatRes/

Default emissions factors were assumed to be: 1.06 (Copper), 0.49 (Zinc), 1.0 (Nickel), and 0.15 (Lead) ktS/kt metal (USEPA 1996b). Default emission factors for Europe were slightly higher, following Mylona (1996). The sulfur content of ores can vary widely and some of these emissions factors are lower limits based on stoichiometry of common ores. Sulfur emissions can be much higher than indicated by these emissions factors due to the presence of additional sulfur compounds in ore, particularly if these are not separated before smelting. Adjustments to these values were made for Canada, the United States, Australia, and the Former Soviet Union to better match emissions inventory data, increasing emissions factors substantially in some instances. In some cases emissions factors were adjusted to assure that the gross sulfur content of smelted ore was larger than reported smelter sulfur removals. Emissions from aluminum manufacture were also included with a coefficient of 0.02 ktS/kt metal (derived from USEPA 1996a), although these emissions are relatively small.

Sulfur removed at smelters from 1972 through 2005 was tabulated at the country level using data from USGS sulfur yearbooks and earlier U.S. Bureau of Mines Mineral Yearbooks. Earlier removals data for the United States were obtained from these same sources from 1928, with two additional data points from Smith (1918) for sulfuric acid produced from zinc smelters in 1914 and 1917 and Weed (1918) for copper smelters. Sulfur recovery in the United States was extrapolated to zero in 1909 when the first recovery plant was reported constructed in Tennessee (Weed 1918). Canadian sulfur recovery data were obtained from the Canadian Minerals Yearbook for 1947-1974, extrapolated to zero in 1925 when the first recovery plant was reported to be opened. Sulfur recovery data for other countries were not available before 1972. Recovery values were converted to a fraction of total sulfur recovered from metals and this fraction was extrapolated to zero in 1950. This date is somewhat arbitrary, as we lack detailed data on when sulfur recovery plants were implemented in Europe. We have one report from Mäkinen (2006) that the acid recovery plant at Harjavalta in Finland was started in 1947 (recovery in Finland was extrapolated to this date). Errors due to these extrapolations are likely to be small by 1950. We estimated sulfur recovery in Canada and the USA to be about 15% of total sulfur in ore by this time.

Sulfur removal in tonnes is converted to a fraction of sulfur removed in order to estimate net sulfur emissions from all smelting operations within each country. Emissions can only be calculated for the sector as a whole because sulfur removal data are generally only available in aggregate and not by metal. Sulfur removal data are also often reported in round numbers, for example based on reports of sulfur removal capacity or sales over various periods. For this reason the sulfur removal percentage was smoothed with a three-year smoother before emissions were calculated. This eliminated spurious variability due to reporting issues.

Where inventory data were available for this sector, generally after 1989, the smelting emissions estimate was calibrated to the inventory value by adjusting the sulfur removal percentage. In a few countries, as discussed in the text, this resulted in a removal percentage that is different than implied by the sulfur removal data. In these cases, the removal fraction was interpolated between the value derived from sulfur removal data and that implied by the inventory data, generally back to 1980. In a few cases, inventory data was available for a few recent years while removal data was available for some years before 1990, in which cases removal fractions were interpolated across the data gaps.

S.7 China coal emissions assumptions

Emissions from coal use in China comprise a large portion of global emissions, but are also particularly uncertain. We discuss in this section assumptions for coal consumption, sulfur content, ash retention, and emission controls. IEA (2006) data are used for coal consumption. There are substantial differences in reported data on coal consumption in China. IEA (2006), BP (2008), and EIA (2008b) data for coal consumption in China are consistently different, with the BP data larger than EIA, and EIA larger than IEA. The source of this difference is not clear.

For 1990 and previous years we assumed an average coal sulfur content of 1.23% for industrial combustion (Foell et al. 1995). A sulfur content of 1.58% was assumed for the residential sector (but with a higher ash retention assumption, see below). For the year 2000, we drew on recent work on the GAINS Asia project (Klimont et al. 2009), using the WEO_2009 scenario (downloaded March 2010, 2008). These authors calibrated their 2000 emissions estimate to an emissions inventory developed for China. Using an energy content of 20.7 GJ/tonne, total coal consumption was estimated to be 1,447 Mt, which implies a sulfur content of 0.97% for hard coal. This energy content value is smaller than the value assumed in the modified IEA data used here, as is the total consumption by weight of 1,167 Mt using IEA assumptions. Total coal consumption in China from BP (2009) in 2000 is 1,320 Mt, also higher than the IEA estimate. The source of these discrepancies is not clear, but we have increased the IEA values to match the trends of the BP annual time series for 2000, 2001, and 2002. This results in a total consumption value that is smaller than the GAINS value but larger than IEA.

The GAINS China estimates for sulfur emissions from coal incorporate a 15% decrease in the total sulfur content of coal from 1990 to 2000. We applied the same decrease here. This resulted in an emissions estimate from coal that is 8% lower than the GAINS Asia value, largely due to the lower coal consumption estimated here. We lack consistent data on coal sulfur content in the year 2005. Overall coal consumption increased by 60-80% over this five-year period, depending on the data source used. With such a large increase in coal consumption, we assume that the fraction of low sulfur coal supplied for consumption decreased over this time. To account for this, we assume a 10% increase in aggregate coal sulfur content from 2000 to 2005.

The sulfur retained in ash (ash retention) was assumed to be 10% for power plants, 20% for industrial combustion, and 30% in the residential sector, similar to values in the GAINS project, but larger than those assumed in the previous RAINS estimates. Klimont et al. (2009) assumed an ash retention fraction of 25% for industrial boilers. Given this is much larger than the value used in other regions (5-10%), we use a slightly lower value here for consistency. Ash retention depends on technology (older, less efficient technologies are thought to have higher ash retention fractions) and coal properties. Industrial combustion accounts for about 20% of coal combustion in 2000, although this fraction is decreasing due to increased coal consumption for electricity generation.

Additional reductions due to coal washing and lime injection were assumed to be 15% in industrial combustion and 10% in electric power plants in 2005, and 5% for these sources in 2000. Following Xu et al. (2009), reduction by FGD operation was assumed to be 3% in 2005 for electric power plants.

The resulting emissions factor combining sulfur content, ash retention, and additional reduction activities was linearly interpolated by sector between the years 1990, 2000, and 2005.

The substantial uncertainties in consumption and fuel properties in China may be due to some combination of incomplete data and inconsistent reporting. Some coal in China is washed or otherwise treated, which means that coal properties at the mine mouth may differ from properties at the point of consumption. A time series of coal production by mine (or mining district), along with measured properties at the mine mouth, is one method by which changes in aggregate coal properties over time could be established.

S.8 Uncertainty Assumptions

Table S-4 shows the classification of uncertainty by country. The inventory codes 1–6 correspond to the numerical values given in Table 2 of the main text as indicated below. Uncertainty is linearly interpolated in between the years shown in the table below. A blank indicates that the fractional uncertainty bounds are linearly interpolated through that two-decade period.

Uncertainty Category by Country													
Country/Region	1870	1880	1890	1900	1910	1920	1930	1940	1950	1960	1970	1980	1990
USA	3	3		2	2	2	2	2	2		1	1	1
CANADA	3	3	3	3	3	3	3	3	3		2		1
AUSTRIA	3	3	3	3		2	2	2	2	2	2		1
GERMANY	3	3	3	3	3	3	3		2	2	2		1
IRELAND	3	3	3	3	3		2	2	2	2	2		1
LUXEMBOU	3	3	3	3	3	3	3	3	3		2		1
TURKEY	4	4	4	4	4	4	4	4	4	4	4	4	4
Western Europe (not otherwise specified)	3		2	2	2	2	2	2	2	2	2		1
LATVIA	3	3	3	3	3	3	3	3	3		2		1
BULGARIA	3		2	2	2	2	2	2	2	2	2	2	2
HUNGARY	3	3	3	3		2	2	2	2	2	2		1
POLAND	3	3	3	3		2	2	2	2	2	2	2	2
ROMANIA	3		2	2	2	2	2	2	2	2	2	2	2
JAPAN	3		2	2	2	2	2	2	2	2		1	1
KOREA	3	3	3	3	3	3	3	3	3	3	3		2
AUSTRALI	3	3	3	3	3	3	3	3	3		2		1
NZ	3	3	3	3	3	3	3	3	3		2		1
International Shipping	6	6	6	6	6	6	6		5	5	5	5	5
Rest of World	4	4	4	4	4	4	4	4	4	4	4	4	4

Category	Code
I. Recent-Country-Inventory	1
II. Older Inventory	2
IIa. OECD (pre inventory)	3
III. Other Countries	4
IV. Int Shipping	5
IV. Int Shipping (earlier)	6

Table S- 4 – Uncertainty categories by country and time period (see Table 2 of main text).

S.9 RCP Inventory

An earlier version (2.50) of this inventory was distributed for decadal years (1850 – 2000) for use in the RCP scenario exercise (Lamarque et al. 2010). The inventory reported here (ver 2.85) contains additional data and revised methodologies to improve the estimate. Figure S-1 and Table S-5 summarize the differences between the current inventory and the RCP release. Compared to the RCP release, the current inventory is slightly larger in 1990, 2000, and 2005 (by 4-5%) and slightly smaller for some early years.

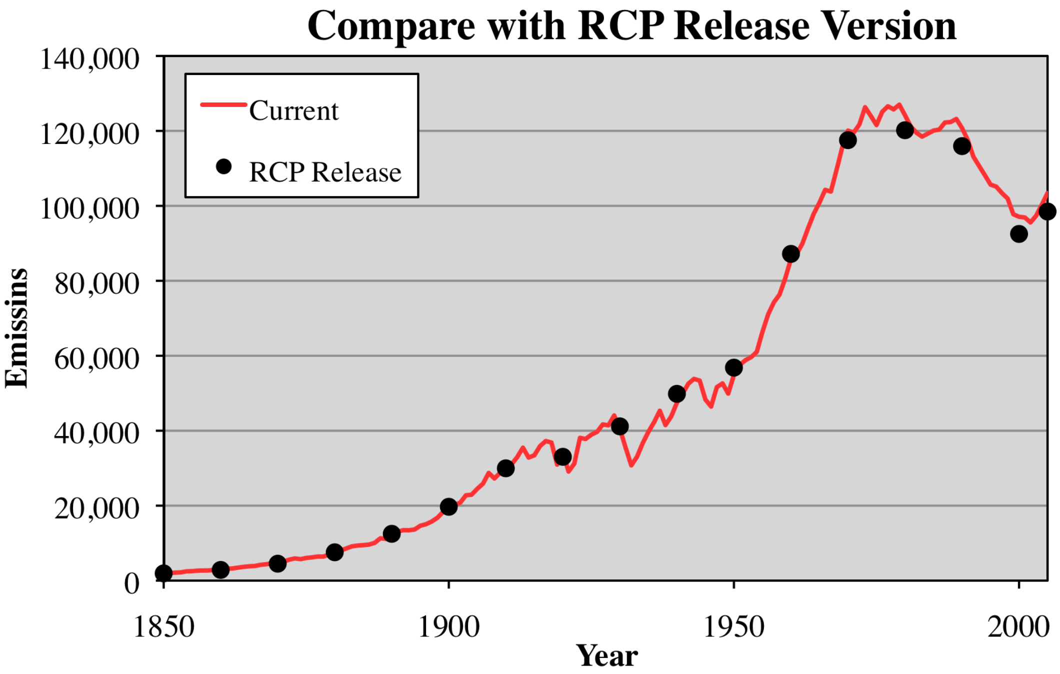

Figure S-1 – Current version (v 2.85) of the inventory as compared to the RCP inventory release for global emissions from fossil fuel combustion and process emissions, excluding international shipping.

Current release as compared to RCP release version

Year	Difference (%)	Difference (Gg SO_2)
1850	4%	72
1860	8%	249
1870	6%	272
1880	3%	195
1890	-1%	-94
1900	-2%	-469
1910	1%	247
1920	3%	1,189
1930	-2%	-682
1940	-5%	-2,376
1950	-3%	-1,917
1960	-2%	-1,362
1970	2%	2,539
1980	3%	4,000
1990	4%	4,814
2000	5%	4,580

Table S- 5 – Current version (v 2.85) of the inventory as compared to the RCP inventory release.

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