Anthropogenic Sulfur Dioxide Emissions: 1850-2005

Supplementary Material

S. J. Smith[1], J. van Aardenne[2], Z. Klimont[3], R. J. Andres[4], A. Volke[1] , S. Delgado Arias[1]

[1] Joint Global Change Research Institute, Pacific Northwest National Laboratory, 5825 University Research Court, Suite 3500, College Park, MD 20740.

[2] European Commission Joint Research Centre, European Commission, Via E. Fermi 2749, TP 290, I - 21027 Ispra (VA), Italy (current address: European Environment Agency, Kongens Nytorv 6, 1050 Copenhagen K, Denmark).

[3] International Institute for Applied Systems Analysis, Schlossplatz 1, A-2361, Laxenburg, Austria.

[4] Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6335, USA.

Correspondence to: S. J. Smith (ssmith@pnl.gov)

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S.1 Region Definitions

Unless specified otherwise, the summary 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).

The correspondence between country and summary region name is given in table S-1 below:

IEA Name	Country Name	Summary Region Name		IEA Name	Country Name	Summary Region Name
ALBANIA	Albania	Central Europe		KUWAIT	Kuwait	Middle East
ALGERIA	Algeria	Africa		KYRGYZSTAN	Kyrgyzstan	FSU
ANGOLA	Angola	Africa		LATVIA	Latvia	FSU
ARGENTINA	Argentina	Argentina		LEBANON	Lebanon	Middle East
ARMENIA	Armenia	FSU		LIBYA	Libya	Africa
AUSTRALI	Australia	Aus & NZ		LITHUANIA	Lithuania	FSU
AUSTRIA	Austria	Western Europe		LUXEMBOU	Luxembourg	Western Europe
AZERBAIJAN	Azerbaijan	FSU		MALAYSIA	Malaysia	South & East Asia
BAHRAIN	Bahrain	Middle East		MALTA	Malta	Western Europe
BANGLADESH	Bangladesh	South & East Asia		MEXICO	Mexico	Mexico
BELARUS	Belarus	FSU		MOLDOVA	Republic of Moldova	FSU
BELGIUM	Belgium	Western Europe		MONGOLIA	Mongolia	China+
BENIN	Benin	Africa		MOROCCO	Morocco	Africa
BOLIVIA	Bolivia	Central America		MOZAMBIQUE	Mozambique	Africa
BOSNIAHERZ	Bosnia and Herzegovina	Central Europe		MYANMAR	Myanmar	South & East Asia
BOTSWANA	Botswana	Africa		NAMIBIA	Namibia	Africa
BRAZIL	Brazil	Brazil		NANTILLES	Netherlands Antilles	Central America
BRUNEI	Brunei Darussalam	South & East Asia		NEPAL	Nepal	South & East Asia
BULGARIA	Bulgaria	Central Europe		NETHLAND	Netherlands	Western Europe
CAMBODIA	Cambodia	China+		NICARAGUA	Nicaragua	Central America
CAMEROON	Cameroon	Africa		NIGERIA	Nigeria	Africa
CANADA	Canada	Canada		NORWAY	Norway	Western Europe
CHILE	Chile	South America		NZ	New Zealand	Aus & NZ
CHINA	China	China+		OMAN	Oman	Middle East
COLOMBIA	Colombia	Central America		OTHERAFRIC	Other Africa	Africa
CONGO	Congo	Africa		OTHERASIA	Other Asia	South & East Asia
CONGOREP	Democratic Republic of Congo	Africa		OTHERLATIN	Other Latin America	Central America
COSTARICA	Costa Rica	Central America		PAKISTAN	Pakistan	South & East Asia
COTEIVOIRE	Cote d'Ivoire	Africa		PANAMA	Panama	Central America
CROATIA	Croatia	Central Europe		PARAGUAY	Paraguay	South America
CUBA	Cuba	Central America		PERU	Peru	South America
CYPRUS	Cyprus	Western Europe		PHILIPPINE	Philippines	South & East Asia
CZECH	Czech Republic	Central Europe		POLAND	Poland	Central Europe
DENMARK	Denmark	Western Europe		PORTUGAL	Portugal	Western Europe
DOMINICANR	Dominican Republic	Central America		QATAR	Qatar	Middle East
ECUADOR	Ecuador	South America		ROMANIA	Romania	Central Europe
EGYPT	Egypt	Africa		RUSSIA	Russia	FSU
ELSALVADOR	El Salvador	Central America		SAUDIARABI	Saudi Arabia	Middle East
ERITREA	Eritrea	Africa		SENEGAL	Senegal	Africa
ESTONIA	Estonia	FSU		SERBMONT	Serbia and Montenegro	Central Europe
ETHIOPIA	Ethiopia	Africa		SINGAPORE	Singapore	South & East Asia
FINLAND	Finland	Western Europe		SLOVAKIA	Slovak Republic	Central Europe
FRANCE	France	Western Europe		SLOVENIA	Slovenia	Central Europe
FYROM	Former Yugoslav Republic of Macedonia	Central Europe		SOUTHAFRIC	South Africa	Africa
GABON	Gabon	Africa		SPAIN	Spain	Western Europe
GEORGIA	Georgia	FSU		SRILANKA	Sri Lanka	South & East Asia
GERMANY	Germany	Western Europe		SUDAN	Sudan	Africa
GHANA	Ghana	Africa		SWEDEN	Sweden	Western Europe
GIBRALTAR	Gibraltar	Western Europe		SWITLAND	Switzerland	Western Europe
GREECE	Greece	Western Europe		SYRIA	Syria	Middle East
GUATEMALA	Guatemala	Central America		TAIPEI	Chinese Taipei	South & East Asia
HAITI	Haiti	Central America		TAJIKISTAN	Tajikistan	FSU
HONDURAS	Honduras	Central America		TANZANIA	United Republic of Tanzania	Africa
HONGKONG	Hong Kong, China	China+		THAILAND	Thailand	South & East Asia
HUNGARY	Hungary	Central Europe		TOGO	Togo	Africa
ICELAND	Iceland	Western Europe		TRINIDAD	Trinidad and Tobago	Central America
INDIA	India	India		TUNISIA	Tunisia	Africa
INDONESIA	Indonesia	South & East Asia		TURKEY	Turkey	Western Europe
IRAN	Islamic Republic of Iran	Middle East		TURKMENIST	Turkmenistan	FSU
IRAQ	Iraq	Middle East		UAE	United Arab Emirates	Middle East
IRELAND	Ireland	Western Europe		UK	United Kingdom	Western Europe
ISRAEL	Israel	Middle East		UKRAINE	Ukraine	FSU
ITALY	Italy	Western Europe		URUGUAY	Uruguay	South America
JAMAICA	Jamaica	Central America		USA	United States	USA
JAPAN	Japan	Japan		UZBEKISTAN	Uzbekistan	FSU
JORDAN	Jordan	Middle East		VENEZUELA	Venezuela	South America
KAZAKHSTAN	Kazakhstan	FSU		VIETNAM	Vietnam	China+
KENYA	Kenya	Africa		YEMEN	Yemen	Middle East
KOREA	Korea	Korea		ZAMBIA	Zambia	Africa
KOREADPR	Korea, DPR	China+		ZIMBABWE	Zimbabwe	Africa

Table S-1 – Region assignment used in this work.

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 were generally available from 1960 for OECD countries and from 1971 for other countries. The following categories of end-use were not included, as emissions from these non-combustion activities were 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 of Andres et al. (1999), who used data, in turn, from Etemad et al. (1991) and Mitchell (1998a, 1998b, 1998c) and other sources. 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, creating 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 were included as process emissions. Sulfur from coking coal used in earlier "beehive" ovens were 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 was assumed to decline linearly between a starting year and end-year as indicated in Table S-2, drawing from Bond et al. (2007). Data on coking coal consumption before 1950 were not available, so combusted coal prior to 1950 was scaled by the ratio of combusted over total coal in 1950.

Region	Begin Year	End Year
Western Europe	1900	1950
Eastern Europe	1910	1950
Other OECD90	1910	1950
FSU	1910	1950
China+	1910	1950
Middle East	1950	1970
Africa	1950	1970
Central & South America	1950	1970
Other Asia	1950	1970

Table S-2 – 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).

Estimating of the emissions from petroleum products is complicated by refinery transformations and trade in multiple grades of petroleum products, grades often defined in terms of sulfur content. Emissions from petroleum products were determined using IEA data as described below (§S.3, §S.5) and in the main text. Where country-level IEA energy data were not available, the earliest emissions estimate was scaled back in time using liquid fuel-related carbon dioxide emissions estimate by country from CDIAC (Marland et al. 2008),

between 1970 and 1950. Emissions before 1971 were scaled using the petroleum consumption estimate of Andres et al. (1999). While this procedure is approximate, particularly for early years, detailed global data were not available and petroleum emissions, not including shipping, are a small portion of total emissions before 1950. An explicit estimate of emissions from petroleum use was calculated for the United States. The resulting global emissions are a good match with the independent mass balance estimate (main text, and § S-8 below).

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 for 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 Hurtt 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.

IEA energy consumption data were processed into end-use sectors for use in the key-year (1900, 2000, 2005) emissions calculations, default emissions estimates, and for use in estimating emissions by end-use sector (see below). The mapping between end-use sectors and IEA data categories followed the categories used in Lamarque et al. (2010), and is given in the table below. Fuel consumption in categories considered process emissions were excluded from the combustion emissions calculations in order to avoid double counting.

End-Use Sector	FLOW_CODE	FLOW
aviation	DOMESAIR	Domestic Aviation
aviation	INTLAIR	International Aviation
domestic	AGRICULT	Agriculture/Forestry
domestic	COMMPUB	Commercial and Public Services
domestic	ONONSPEC	Non-specified (Other)
domestic	RESIDENT	Residential
domestic	THEAT	Heat Pumps
energy	DISTLOSS	Distribution Losses
energy	EBIOGAS	Gasification Plants for Biogas
energy	EBKB	BKB Plants
energy	EBLASTFUR	Blast Furnaces
energy	ECHARCOAL	Charcoal Production Plants
energy	ECOALLIQ	Coal Liquefaction Plants
energy	ECOKEOVS	Coke Ovens
energy	EGASWKS	Gas Works
energy	EGTL	Gas-to-Liquids (GTL) Plants
energy	ELNG	Liquefaction (LNG) / Regasification Plants
energy	EMINES	Coal Mines
energy	ENONSPEC	Non-specified (Energy)
energy	ENUC	Nuclear Industry
energy	EOILGASEX	Oil and Gas Extraction
energy	EPATFUEL	Patent Fuel Plants
energy	EPOWERPLT	Own Use in Electricity, CHP and Heat Plants

(Table continues next page)

End-Use Sector	FLOW_CODE	FLOW
energy	EPUMPST	Used for Pumped Storage
energy	EREFINER	Petroleum Refineries
energy	TCHARCOAL	Charcoal Production Plants
energy	TGASWKS	Gas Works
energy	TNONSPEC	Non-specified (Transformation)
energy	TPETCHEM	Petrochemical Industry
energy	TREFINER	Petroleum Refineries
industry	CHEMICAL	Chemical and Petrochemical
industry	CONSTRUC	Construction
industry	FOODPRO	Food and Tobacco
industry	INONSPEC	Non-specified (Industry)
industry	IRONSTL	Iron and Steel
industry	MACHINE	Machinery
industry	MINING	Mining and Quarrying
industry	PAPERPRO	Paper, Pulp and Print
industry	TEXTILES	Textile and Leather
industry	TRANSEQ	Transport Equipment
industry	WOODPRO	Wood and Wood Products
not-used	BUNKERS	International Marine Bunkers
process-emission	NONFERR	Non-Ferrous Metals
process-emission	NONENUSE	Non-Energy Use
process-emission	NONMET	Non-Metallic Minerals
process-emission	TBKB	BKB Plants
process-emission	TBLASTFUR	Blast Furnaces
process-emission	TCOALLIQ	Coal Liquefaction Plants
process-emission	TCOKEOVS	Coke Ovens
process-emission	TGTL	Gas-to-Liquids (GTL) Plants
process-emission	TPATFUEL	Patent Fuel Plants
transportation	DOMESNAV	Domestic Navigation
transportation	FISHING	Fishing
transportation	PIPELINE	Pipeline Transport
transportation	RAIL	Rail
transportation	ROAD	Road
transportation	TRNONSPE	Non-specified (Transport)

Table S-3 – Mapping between the end-use sectors used here and IEA data categories.

S.3 Methodology Overview

Constructing a global estimate of emissions over the time period considered here must take into account changing patterns of fuel consumption and emissions controls, while also recognizing the limited availability of detailed data for earlier years. The methodology used for this estimate, as outlined in Figure 1 in the main text, begins with the development of an inventory by fuel and sector (domestic, industrial, transport, and industrial combustion, and specific industrial processes) for three key years: 1990, 2000, and 2005. The estimates for these years were given the most scrutiny and compared with inventory data and other estimates. For countries with detailed emission inventory data (§ S.4) these data could be

calibrated exactly to emissions by sector. Emissions from Asia were compared with other inventories as described in S.4 below.

A default set of emissions with annual resolution was constructed by linearly interpolating emissions factors by sector and fuel between the key years 1990, 2000, and 2005. Emissions factors before 1990 were initially scaled regionally by trends from Smith et al. (2004), back to 1970-1975 and generally held constant before this point. This was done at the sectoral level where fuel consumption information was available, which is back to 1960 for OECD countries, and 1971 for most non-OECD countries. Earlier IEA data for coal consumption in the Former Soviet Union were used to scale country level sectoral coal consumption from 1990 to 1975.

The regional scaling for petroleum emissions was adjusted at the country level to eliminate outlier values in the final result for countries with inventory data. This default scaling of petroleum emissions ranges from ~20% increases from 1990 to 1970 in some developing regions (Middle East, Central & South America, and Africa) to 400% increases in Canada, Europe, and Japan. The assumptions for petroleum sulfur content are most important in the 1970s, where petroleum emissions (not including shipping) were a large portion of total emissions. Before this time assumptions for coal sulfur content begin to dominate the emissions estimate.

For earlier years, where sectoral energy consumption data were not available, default emissions from coal and petroleum were separately scaled by total consumption from the composite dataset described above (§ S.2).

The result at the end of this calculation is an estimate of emissions by country and source (petroleum combustion, coal combustion, biomass combustion, smelters, pulp and paper, oil processing, natural gas processing, and other process). Data on fuel use and sulfur properties by sector were not available for earlier years, so the calibration to inventory values is performed at this more aggregate level instead of by end-use sector. Emissions from agricultural waste burning were also estimated, but not included in calibration with inventory values.

The final emissions estimate was determined by scaling emissions from petroleum and coal combustion so that total emissions from fossil fuels and processes are equal to emissions estimates from the inventories described in the next section (§ S.4). Because petroleum emissions become a relatively small component before 1970, petroleum emissions were generally not scaled before this point in time. Before 1990, inventory data were generally available only for countries in Europe, the USA, and Japan. Where aggregate coal and petroleum emissions factors appeared to be unreasonable, the default emissions estimate was altered to improve consistency between regions. The result of this calculation is an estimate of total sulfur emissions by fuel (coal, petroleum, biomass) and process (smelting, etc.) by country.

The ratio of inventory to default values in 1975 for countries in Europe ranged from 0.3 to 2, indicating that emissions factors changed at different rates in different countries (See Figure S-1). The scaling changed total European emissions little (~ 1%), which is expected since the inventory data used for calibration are nearly the same as the data used to develop the regional estimate of Smith et al. (2004). This does, however, give some indication of the differing rates at which emissions factors change in different countries, due to differences in the use of end-

of-pipe controls and changes in sulfur content. The ratios of default to inventory values in 1960 have a similar range. Differences in 1960 will largely reflect country-level differences in coal sulfur content, while differences in 1975 will reflect a combination of differences in coal properties and differences in petroleum product consumption. There are groups of countries with small adjustment ratios in 1975 but with large adjustments in 1960, and also a group of countries with a range of adjustment ratios in 1975 but similar ratios in 1960 (Figure S-1).

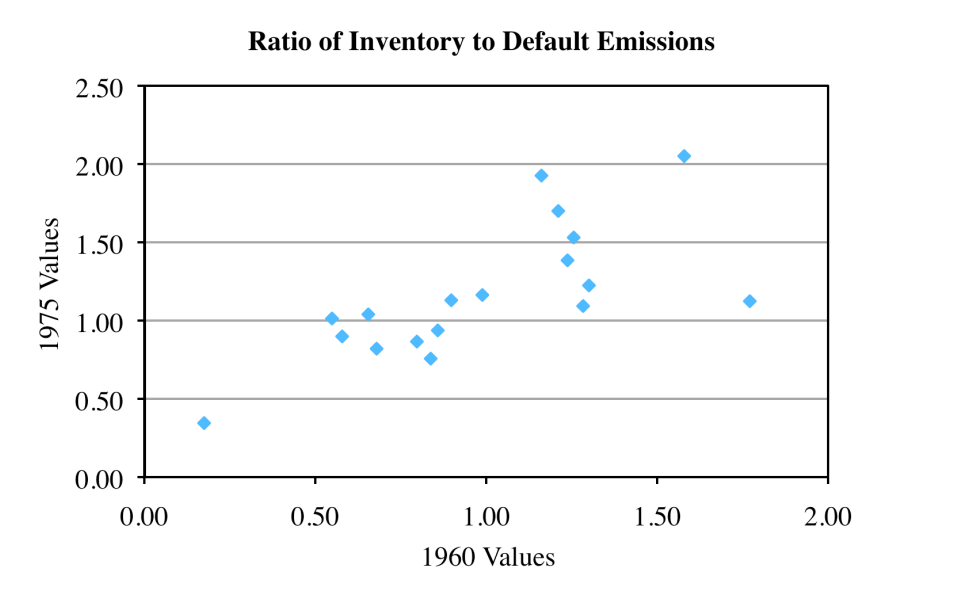

Figure S-1 – Ratio of inventory to default values for 18 European countries where adjustments were made in the indicated year. Iceland and Ireland are not included because emissions from these countries are small.

The Mylona (1996) inventory, to which all of the European values were calibrated, relied on country-specific information by year for sulfur contents and properties of coal and petroleum products, including detail on fuel type for coal (hard and brown coal) and petroleum products (petrol, medium distillate, and residual). This level of detail provides for a more reliable estimate than regional values, which is why the Mylona (1996) inventory was used as a benchmark.

The final step is to estimate emissions by end-use sector. The sectoral split for 1990, 2000, and 2005 was available from the first step of this methodology. For previous years, emissions from coal and petroleum were split into end-use sectors (domestic, industrial, energy, and transportation) for each decadal year from 1850-1980. Where sectoral energy consumption data by country were available from IEA, these were used to perform this split. Where these data were not available, generally before 1970 or 1960 (§ S.2), the trend in sectoral distribution of NO_x emissions from the EDGAR-HYDE historical inventory (van Aardenne et al. 2001) was used to scale the sector-specific estimates developed here back to 1890. Emissions factors in the EDGAR-HYDE estimate were held constant before 1970. While, in principle, any of the emissions species could have been used, NO_x emissions were the most appropriate for this purpose since they arise almost entirely from combustion processes for the sectors considered. The sectoral split was linearly interpolated from 1890 to 1850 to match the 1850 distribution of fuel use estimated by Bond et al. (2007).

This is an approximate procedure, however; sector-specific activity data were not available for these earlier years. Such data could be used in the future to improve the accuracy of the sectoral split, although for SO_2 emissions, this approach was only used to estimate sectoral emissions and had no impact on total emissions.

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.

S.4 Emissions Inventories Used

Table S-4 details the inventory data used for calibration by region and specific countries. There were several sources of data for European countries, although a consistent, composite data series for all years 1980-2005 was not available for all countries.

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 through 1980	Mylona (1996)
Turkey	1990, 2000, 2005	GAINS [c]
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).

[c] Baseline scenario in the European GAINS application; available from http://gains.iiasa.ac.at

Table S- 4 – Inventory data used for calibration of total country emissions.

Emissions estimates were calibrated to annual inventory data from 1980-2005 where available, and to values every five years before 1980. The calibration adjustments were interpolated for intermediate years where inventory data were not 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.

A number of additional emissions estimates were 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), GAINS Asia (Klimont et al. 2009; for China and India), and recent GAINS implementation of the World Energy Outlook 2009 data (IEA, 2009) available from the 'UNEP_IEA09_Ref' scenario in the online application (http://gains.iiasa.ac.at). In many cases, these estimates were 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-5.

	2000					2005		
Country	**Current Estimate**	**REAS**	**Trace-P**	**GAINS**		**Current Estimate**	**Intex-B***	**GAINS**
China	21,393	27,590	20,385	23,176		32,673	31,020	34,364
Taiwan	311	266	376	494		246	189	402
North Korea	345	298	227	265		248	233	263
South Korea	943	987	829	771		395	497	555
Thailand	967	998	961	1,017		542	1,299	548
Indonesia	1,012	1,078	884	833		1,535	1,451	1,077
SE Asia (Rest of)	1,248	1,577	1,475	1,583		1,717	2,729	1,423
India	5,363	6,141	5,536	5,128		6,275	5,596	6,413
Pakistan	971	1,097	1,416	788		771	2,882	878

* The Intex-B estimate is for 2006.

Table S- 5 – Comparison with recent inventory estimates for Asia. Note that the "SE Asia (rest of)" region in this table excludes all the other countries individually included in this table. The GAINS column includes estimates from GAINS-Asia project for China and India (Klimont et al., 2009), where national data provided by local experts was used, with remaining data from the GAINS UNEP/IEA baseline scenario.

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-5.

S.5 Methodological Detail: Fossil Combustion Emissions

For most European countries and the United States, emissions estimates by country were available from at least 1900. Emissions from coal and petroleum were scaled up or down in order to match total country emissions over time where inventory data were available. While total emissions were constrained by inventory values, emissions by fuel were less well constrained by the available data. These emissions estimates often imply changes in aggregate coal sulfur content over time.

Following an assumption similar to that of Mylona (1996), we linearly increased the assumed sulfur retained in ash for coal combustion to 20% in 1920, starting from regional values in 1960, to account for changing coal combustion technologies. Coal used for the production of coke or as process heat for cement manufacture was not included in the calculation of combustion emissions in recent years; any residual emissions from these uses were assumed to be included in the process emissions estimate. Emissions from coal used for coke production were included in earlier years before the introduction of by-product coke plants. Assumptions for sulfur emissions from coal combustion in China and the countries of the Former Soviet Union were drawn from Foell et al. (1995), Klimont et al. (2009), Xu et al. (2009) and Ryaboshapko et al. (1996) as discussed below.

The sulfur content of coal within each country is assumed to be constant over time except as indicated above. This assumption can be problematic for countries with significant imports unless detailed country-level inventory data are available. To examine the potential importance of this assumption, we examined coal imports and emissions in 1990 (the situation is similar in 2005). There were only three countries outside of Europe and Japan, where we have calibrated to emission inventories, with imports larger than 10,000 tonnes per year: Brazil, South Korea, and Taiwan. While there are other countries with large net coal imports relative to consumption, the fraction of sulfur emissions from coal in those countries was small. We estimate coal to have produced 11%, 25%, and 24% of SO_2 emissions in these three countries in 1990. Information on the source of coal imports over time would be helpful to refine emissions estimates from these countries, although the impact is relatively small in absolute terms. A more refined estimate is complicated by the need to know not only the properties of imported coal but also its use. For example, the emissions impact of coal used to produce coal coke can be very different than coal used for electricity generation.

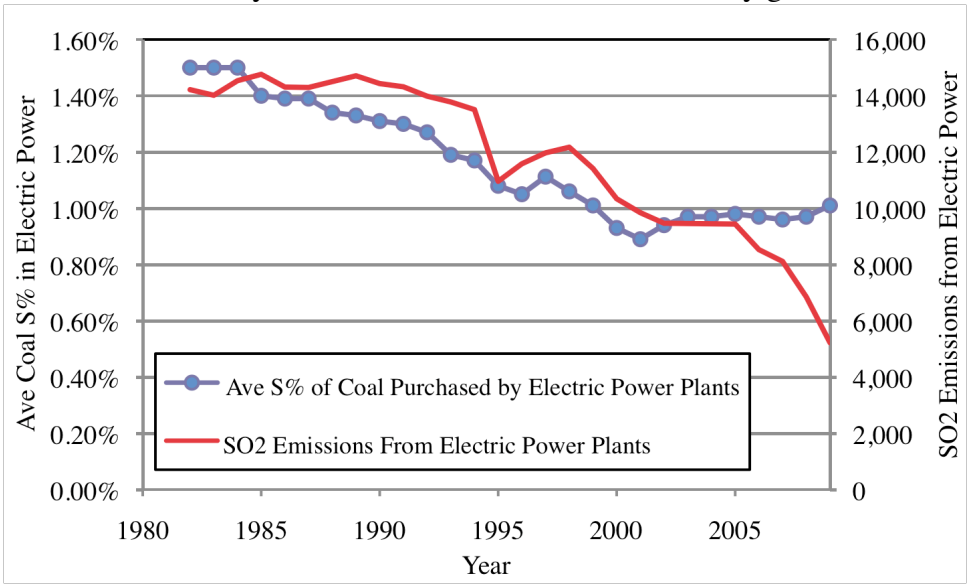

Figure S-2 – Emissions from electric power plants (US EPA 1996a, 2008) and the average sulfur content of coal purchased by electric power plants (US DoE 2009 and previous versions). Emissions for 2009 estimated using DoE emissions figures to scale EPA emissions estimates for 2008 (US DoE 2008).

An example of the impact of changing sulfur content is shown in Figure S-2, for emissions from electric power generation in the United States. Until 2000-2005, emissions dropped roughly in parallel with coal sulfur content. After 2004, however, emissions have continued to

decrease while coal sulfur content actually has increased slightly. Presumably, stronger emissions limits could not be met only with low-sulfur coal, and flue-gas desulfurization units now play an increasing role in driving lower emissions.

S.6 Methodological Detail: 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, were available only for recent years, or were incomplete. 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.10, 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 very 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.7 Methodological Detail: 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 were used for coal consumption. There were 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). 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 were 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

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

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, which is also the rate of decrease assumed by Ohara et al (2007). We applied the same decrease rate 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 lacked 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 assumed that the fraction of low sulfur coal supplied for consumption decreased over this time. To account for this, we assumed 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 used in Smith et al. (2004). Klimont et al. (2009) assumed an ash retention fraction of 25% for industrial boilers. Given that is much larger than the value used in other regions (5-10%), we used 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 accounted 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 flue-gas desulfurization (FGD) units 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 (Akimoto et al 2006). 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 Methodological Detail: 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 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 off California 1.1%. For a few states without sulfur content information, a default value of 0.8% was used, although this had 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 (1992 - 2006) 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 for some regions and years were reported only as totals that included sulfur recovered from natural gas processing. The amount of sulfur removed from natural gas processing was 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 natural gas plants for the earliest available year, extrapolated in time using natural gas production. For several countries and regions (Middle East, Germany, Kazakhstan, and Uzbekistan) only the total extraction from natural gas and petroleum was reported. The amount removed from natural gas was estimated using extraction ratios from other regions and was subtracted from these totals. These estimated quantities accounted for a significant portion of the total estimated sulfur recovery, increasing from 30% in 1990 to 40% in 2005, which results in significant uncertainty in the global mass-balance estimate.

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

[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

sulfur removal from petroleum was 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 for 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 were 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 above. 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 were 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 were from refineries (in which case they should be included in the mass balance), and what portion were from sulfur gases associated with crude oil extraction (which should not be included in the mass balance estimate).

As a check of the assumptions used in the crude oil mass balance calculations, we compared the aggregate sulfur content of crude oil refined in the United States as estimated by this mass-balance estimate with reported values. The estimate constructed here for the mass balance calculation averages 7% higher than the average sulfur content of crude input to refineries as reported by EIA from 1981-2002, and within 1% of the value reported by Bingham et al. (1973) for 1970. This lends confidence to our conclusion that emissions from petroleum combustion in the United States were underestimated in earlier years.

S.9 Methodological Detail: Shipping Emissions

We constructed a composite global estimate of shipping fuel consumption following Eyring et al. (2010) by using values from Eyring et al. (2010) for 1896 – 2007 and Endresen et al. (2007) from 1925 – 2007. As discussed by these authors, these estimates include both international and domestic shipping. For years prior to 1925 we used values reported in Fletcher (1997) from 1870. The split between coal and petroleum fuels in early years is estimated by combining data reported in Endresen et al. (2007) and Fletcher (1997). Values were linearly interpolated for years where no data were available. Estimates of fuel consumption for 1860 and 1850 were obtained by scaling with total ship tonnage as reported in Mitchell (1998a, 1998b, 1998c), using the tabulation of Bond et al. (2007).

In order to estimate sulfur emissions, we require a time series of bunker fuel consumption by fuel type (e.g., coal, residual, distillate, and other). We estimated these data by combining reported bunker fuel sales from EIA International Energy Annual (EIA 2008b, and previous years) with UN data for earlier years. These data represent reported bunker fuel sales. The bottom-up global fuel estimates above also include domestic shipping fuel consumption, so in order to produce a consistent comparison we added reported values from IEA for domestic

shipping and fishing (available from 1971 forward) to our composite consumption time series. The difference between the bottom-up consumption time series and the composite reported fuel consumption time series is remarkably small. The 5-year running average difference between the total regional time series and the global estimate is 10% or less over the period 1971-2005.

We find, therefore, that reported fuel consumption over this period appears to be only slightly lower than that inferred by bottom-up analysis. Thus, we used the composite-derived fuel consumption time series as the basis for determining the split between residual and distillate fuel consumption for purposes of estimating sulfur emissions. We estimate that the fraction of residual fuel used in shipping has fallen steadily over time, from an estimated value of 78% in 1971 to 59% in 2005. The residual fraction is kept constant prior to 1971 due to a lack of data.

The most authoritative data on marine fuel sulfur content for bunker fuels is from the sulfur-monitoring program of the International Marine Organization's Marine Environment Protection Committee. The data are supplied by "providers of sampling and testing services" with average sulfur content is based on the "number of samples tested and not the actual quantity of fuel oil bunkered" (IMO 2007). It does not appear that the fraction of bunker fuel tested is known, nor is the magnitude of potential bases if sulfur contents were weighted by bunker amount. The uncertainty in reported sulfur content is, therefore, is difficult to estimate.

	Shipping Emissions		
Year	International	Internal Nav + Fishing	Total
1850	30	5	36
1860	60	11	70
1870	104	18	122
1880	182	32	215
1890	294	52	345
1900	446	79	525
1910	725	128	852
1920	963	170	1,133
1930	1,137	200	1,337
1940	1,116	196	1,313
1950	1,624	286	1,909
1960	2,340	412	2,752
1970	3,234	569	3,804
1980	3,304	619	3,923
1990	3,520	590	4,110
2000	4,889	650	5,539
2005	6,039	653	6,692

Table S- 6 – Estimated shipping emissions. The column labeled Internal Nav + Fishing emissions are estimated emissions from IEA fuel use reported in these categories. These emissions were subtracted from the totals to give the international estimate, which is used in the tables and figures in this work to avoid double counting.

As discussed in the main text, emissions from fuel-use reported as internal navigation and fishing in the IEA data are included in the surface transportation sector of the inventory. The figures and tables presented elsewhere in this paper do not include these emissions in order to avoid double counting. Many estimates of shipping, however, include all shipping emissions.

In order to allow comparison, both components are given in Table S-6. Note that the emissions estimate here for domestic and fishing emissions is almost certainly an underestimate, since these data are likely underreported in the IEA dataset as is international shipping fuels. The total emissions category below is, therefore, the most reliable figure since this corresponds to our estimate of total shipping fuel consumption, not just reported IEA data.

Emissions from fuels reported in the internal navigation and fishing categories were calculated using the same emissions factors as for overall shipping. Reported fuel use for internal navigation and fishing had a lower fraction of residual oil, about 40% in the 1970s decreasing to around 30% from the mid-1980s onward. The fraction of emissions from fuel reported used for internal navigation and fishing category was 15% of total emissions in 1971, the earliest year for which these data are available. This fraction was held constant for earlier years.

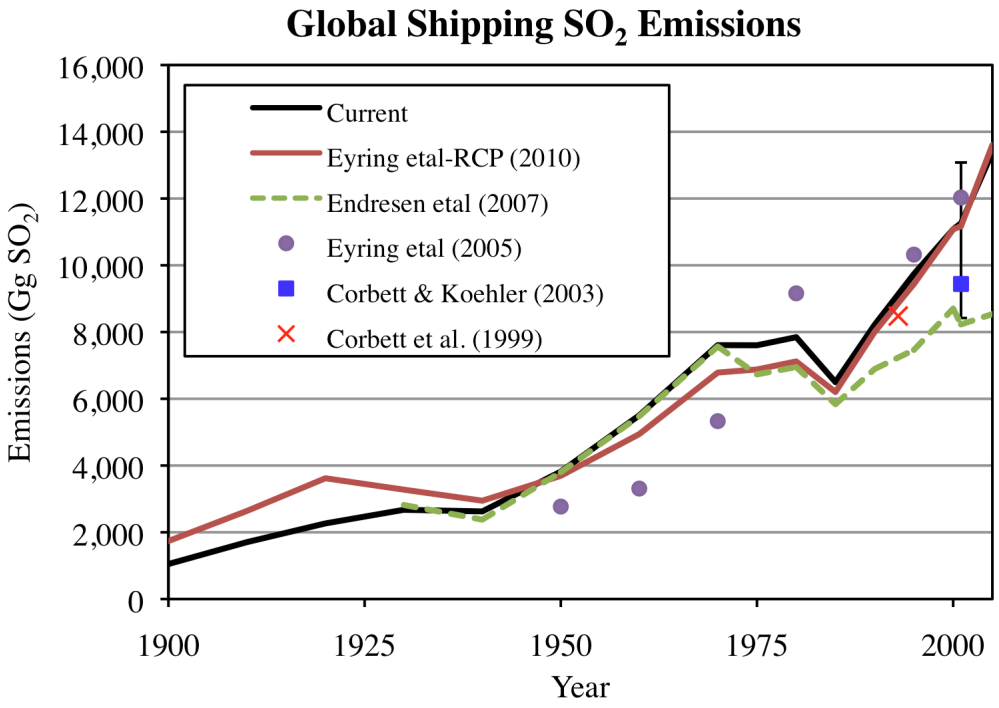

Figure S-3 – Global shipping emissions from this work compared to previous work.

Figure S-3 shows emissions from this work as compared to previous estimates. The estimate here is close to the Eyring et al. (2010) estimate from 1985 – 2005, which is expected since the estimate here was calibrated to the year 2000 value from Eyring et al. (2010). Differences result from our assumption that aggregate sulfur content changes over time due to the changing mix of residual and other fuels.

The estimate here is also close to that of Endresen et al. (2007) prior to 1970. The fuel consumption used here is identical to the Endresen et al. (2007) values before 1970 and the sulfur assumptions used here are similar to those in Endresen et al. (2007).

The Eyring et al. (2010) estimate was extended to earlier years in Lamarque et al. (2010) by scaling with CO_2 emissions. The estimate here is below the RCP extension before 1950 as

coal becomes more important as a shipping fuel because the sulfur content of coal, relative to its CO_2 content, is lower than that of petroleum.

The current estimate, as well as that of Eyring et al. (2010), is within the uncertainty range estimated by Corbett and Köhler (2003) of 8,400 – 13,100 Gg in 2001, which was determined through a detailed uncertainty analysis. The approximate uncertainty range used in this work (Table 3 main text) of ±28% corresponds to a similar range of 8,100 – 14,400 Gg SO_2, although the central value used here is higher than the central value in Corbett and Köhler (2003).

S.10 Methodological Detail: 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.

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, and extrapolated to zero in 1925 when the first recovery plant was reported to be opened. Sulfur recovery data for other countries were not

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

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 lacked detailed data on when sulfur recovery plants were implemented in Europe. We had 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 were available for a few recent years while removal data were available for some years before 1990, in which cases removal fractions were interpolated across the data gaps.

S.11 Methodological Detail: Other Emissions

Data for natural gas production emissions were found only for the United States (USEPA 1996a) and for the Former Soviet Union (Ryaboshapko et al. 1996). In order to construct estimates for other natural gas producing regions, we assumed a default emissions factor of 0.29 kt SO_2/Mtoe (toe=tonne of oil equivalent) for natural gas production, derived from US EPA inventory estimates. For the countries of the Former Soviet Union, we find a slightly higher emissions factor of 0.59 kt SO_2/Mtoe, based on the emissions reported in Ryaboshapko et al. (1996), which is expected since petroleum deposits in the Former Soviet Union also have higher than average sulfur content. The default emissions factor was scaled in regions with aggregate petroleum sulfur contents significantly different from the United States. The emissions factor for the Middle East was scaled up by 1.9 and in Africa scaled down by 0.4.

Petroleum production emissions were taken either from country level inventories or, where those were not available, from the EDGAR 3.2 (Olivier and Berdowski 2001) and 3.2 FT inventories (Olivier et al. 2005). Emissions were scaled with petroleum production over time where inventory data were not available.

Emissions from pulp and paper operations were estimated using emissions factors from Mylona (1996) for early years, scaled down to match UNFCCC and EDGAR inventory estimates by 1990. The emissions factors derived from USEPA emissions data for the mid 20th century were far lower than those from Mylona, so an intermediate emissions factor was adopted for US emissions previous to 1960. Wood pulp production was taken from FAO forest product statistics for 1961–2005 (FAOSTAT 2009), supplemented by data from Mitchell (1998a,b,c), McKeever (1987), and Statistics Canada (Leacy 1983).

Biomass emissions factors for sulfur dioxide range over an order of magnitude depending on the fuel source. Biomass properties are not uniform and it is, therefore, difficult to estimate sulfur emissions from biomass combustion without source- and location-specific data. The default value from EPA's AP-42 document is 0.2 kg/Mg (USEPA 1996b). Streets et al. (2003) use values ranging from 0.18–4.1 kg/Mg. Recent analysis conducted as part of the GAINS Asia project[5] resulted in estimates of 1.1 kg/Mg for China and 0.4 kg/Mg for India (assuming an energy density of 16 GJ/tonne). For this estimate, we use a default value of 0.25 kg/Mg for most regions, 0.4 kg/Mg for countries in South Asia, and 0.8 kg/Mg for East Asia. Although there is a large relative uncertainty in sulfur emissions from biomass combustion, biomass emissions are a relatively small contributor to total sulfur emissions in most regions. Biomass emissions were assumed to be included in the country level inventories used for calibration in recent years, in order to avoid double counting, and were added prior to 1980 in those countries with recent inventory data.

Remaining process emissions originate from a variety of sources, with sulfuric acid production one of the largest sources, particularly in earlier years. Process emissions were taken from the above sources and scaled over time prior to 1990 where inventory data were not available by the regional HYDE estimate (van Aardenne et al. 2001). Where updated 2005 data were not available, year 2000 values were used.

S.12 Methodological Detail: Gridding Details

The EDGARv4.0 energy sector emissions (JRC/PBL, 2009) were distributed on a 0.1° grid based on a combined allocation grid using information from Carma[6], IEA Coal Power (IEA 2008), and Platts (2006), supplemented with population data (CIESIN and CIAT 2005) where no power plant information was available. Emissions from industrial combustion were allocated using urban population density. Transport emissions where allocated based on a road network density distribution based on VMAP (NIMA 1997). Emissions from agricultural waste burning were allocated using cropland maps derived from HYDE (Goldewijk et al. 2007). The EDGAR 4.0 emissions grids were then aggregated to a 0.5° grid for use in this project.

Emissions from smelting and fuel processing were distributed using EDGAR 3.2 1990 and EDGAR FT 2000 gridded distributions, since updated spatial information on these sources was not available. In order to capture the concentrated nature of smelting emissions, the EDGAR 3.2 and FT grids were mapped to 0.5° resolution by placing the source in the lower left quadrant of each 1° grid cell. For several countries, no smelting emissions were present in the EDGAR data set and additional smelter locations were added using a USGS copper smelter database (USGS 2003) and literature sources.

S.13 Comparison With Previous Version

Much of the data, assumptions, and methodology used here are similar to those in Smith et al. (2004). Analysis of the differences between these two inventories can provide guidance to the magnitude and sources of uncertainties in estimating sulfur dioxide emissions. Figure S-4 shows the difference between the current estimate and the Smith et al. (2004) estimate by

[5] http://gains.iiasa.ac.at/index.php/gains-asia
[6] www.CARMA.org

source. The current estimate is lower largely due to lower emissions from coal and petroleum. Lower assumed emissions from terrestrial petroleum combustion are offset slightly by higher emissions from ocean shipping.

Other process emissions are higher in this estimate, largely due to a more comprehensive treatment of these sources. In countries where emissions were calibrated to total values, however, this change did not increase total emissions but, instead, just changed distribution of emissions by source. For example, about one half of the increase in other process emissions for 1970 was from Europe and Japan, which was offset by lower assumed fossil emissions.

Figure S-5 shows the difference by region. The largest absolute changes are in the estimated emissions from China and from the Former Soviet Union. Emissions averaged for 1950-2000 are lower by 30% and 25% respectively in this estimate. Emissions from South and East Asia are also substantially lower in recent years. Smaller differences are seen in most regions.

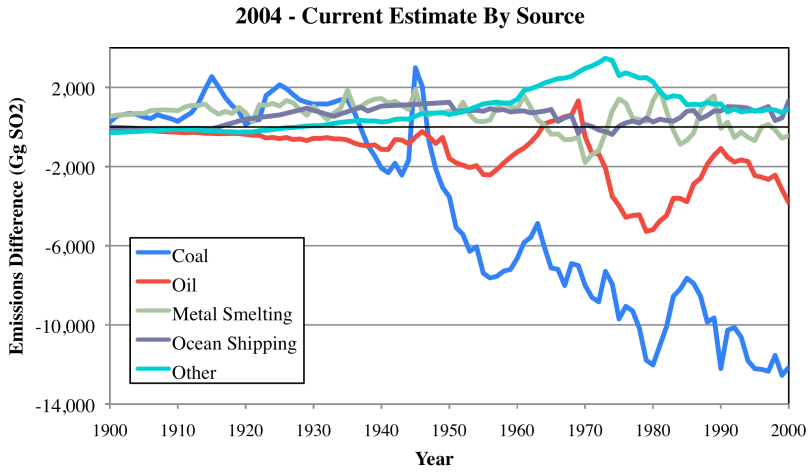

Figure S-4 – Emissions from Smith et al. (2004) minus current values by source.

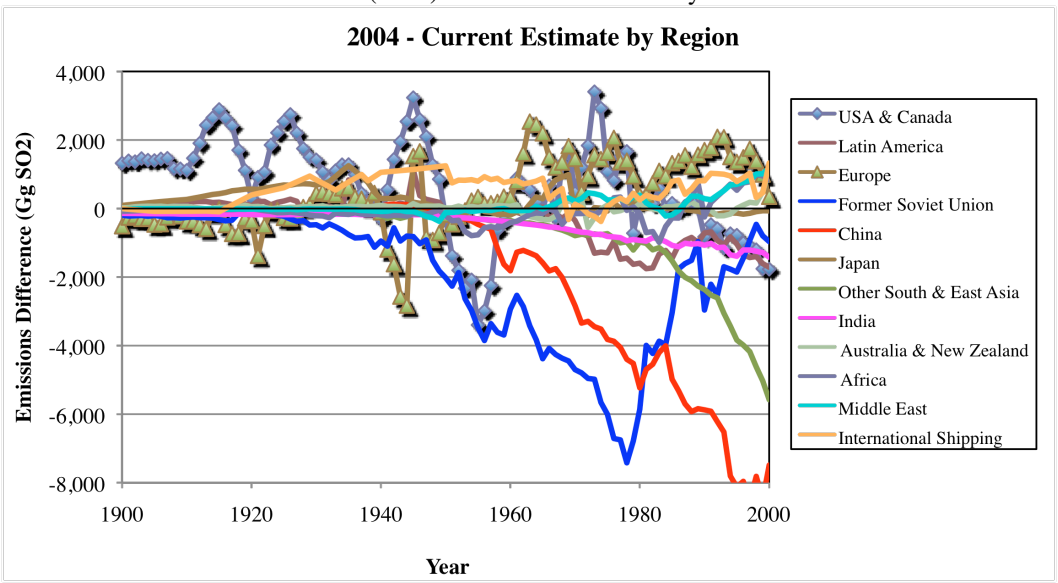

Figure S-5 – Emissions from Smith et al. (2004) minus current values by region. Open burning emissions (forest and grassland fires, agricultural waste burning on fields) are not included.

The reasons for these differences vary by region. For China, the difference is due largely to three factors: different assumed energy to weight conversion in recent years, different treatment of coking coal, and larger values assumed for sulfur retained in ash. Taken together, these three changes resulted in lower emissions in all years.

Where IEA data are used, the current estimate uses energy to weight conversion factors reported by IEA. The resulting total coal consumption by weight is lower than that used in Smith et al. (2004) by about 15% (averaged over 1985-2000). The IEA total by weight is also lower than the BP (2009) estimate by 9% and lower than that reported by EIA by 4%. Reported figures, therefore, vary for reasons that are not clear.

A second reason for lower emissions from coal in China and in other developing regions is the assumption that coking coal is currently used in by-product plants such that the majority of the sulfur dioxide contained in coal is captured as acid (S.2). This has a particularly large effect on China, where the fraction of coal used in coking plants has increased from 8% in 1985 to over 10% in recent years (IEA 2006). The resulting emissions estimate was, therefore, decreased by nearly this fraction. This change in methodology also is the reason for a portion of the decrease in coal emissions in other developing regions.

An increase in the assumed amount of sulfur retained in ash for China, particularly for industrial coal combustion (20-25% in the current version compared to 14% in Smith et al. 2004), also reduced emissions. This results in a 5-10% decrease in estimated emissions. Considerable quantitative uncertainties are present in each of the factors discussed: China coal consumption by weight, emissions factors for China coking coal operations, and the fraction of sulfur retained in ash.

This estimate also assumes an increase in emissions control efforts from 1990 to 2000 resulting in a 4% aggregate decrease in emissions factor over this period. The earlier Smith et al. (2004) estimate had a constant emissions factor for coal over this period.

The second largest change is in the estimate of emissions from the countries of the Former Soviet Union. The primary reason for this difference was a change in base-year data. For a small portion of the FSU inventory data could be used to calibrate recent emissions, which changed base-year emissions. A change in interpretation of the data source used for the remainder of the FSU (§ S.6) also decreased emissions. Because emissions factors now differ between the different countries that comprised the FSU, the amount of fuel used in each of these countries over time can have a large impact on the results over time. This is particularly important for emissions from coal combustion, where regional emissions factors derived in Ryaboshapko et al. (1996) vary substantially for major regions. Coal from Ukraine has twice the emissions factor of the FSU average, for example. Note that a portion of this variation is due to differences in assumed retention in ash. Changes over time prior to 1990 in the distribution of coal production and consumption over the countries of the FSU, and its associated properties, is a substantial source of uncertainty for historical emissions in this region.

Emissions in South and East Asia (exclusive of mainland China and Japan) are lower in recent years due to lower fuel sulfur standards overall and emissions controls on coal power plants, particularly in South Korea, Taiwan, and Thailand.

Emissions in Eastern Europe are slightly higher than in the previous estimate due to the use of more complete inventory data.

Overall, estimated emissions from petroleum combustion from countries constrained by inventories (Europe, USA, Canada, Japan, Australia, and New Zealand) increased in this inventory, although this did not alter the total emissions estimate except for Eastern Europe. The change in source attribution was apparently due to improved calibration at the country and source level that was facilitated by the availability of newer and more detailed inventory data.

Estimated emissions from petroleum products from the rest of the world decreased significantly as compared to the Smith et al (2004) estimate. This change, in aggregate, is one of the larger changes between the two inventories and contributes to the lower overall emissions estimate. The difference in petroleum emissions for developing and reforming economies, was about half the total difference in FSU, China, and Asia coal emissions discussed above, averaged from 1950 – 2000s. Uncertainties in the sulfur content of petroleum products contributes emissions estimates for these countries rely on assumptions about fuel sulfur is a major source of these differences.

While the congruence between the petroleum emissions estimate produced here and the global mass balance estimate for petroleum (main text, and §S.8 above) provides support for the emissions estimate, substantial uncertainties exist in both calculations.

S.14 EPA Inventory Analysis

The U.S. Environmental Protection Agency has been releasing inventory estimates for many years. Some insight into uncertainties in inventory estimates can be gained from examining changes in estimated emissions values over time. Earlier EPA emissions data has been released with detailed sulfur dioxide emissions by fuel and sector. This allows an informative analysis of the sources of changes in inventory values over time. Note that data at this level is not available after 2002, so the analysis below examines changes up to this point.

Table S-7 shows different patterns of changes in inventory values over time. Several points are evident. Reported emissions from sectors where a large portion of emissions are measured, such as those from coal-fired power plants, do not change much over time.

Petroleum emissions have changed substantially at times as assumptions have been refined. Petroleum emissions arise from a variety of sectors, many not regulated during the time period shown, making data collection difficult. From 1985 – 2000, 48% - 36% of sulfur emissions from petroleum combustion in the United States is from residual fuel. The sulfur content of residual fuel is variable and subject to different regulations in different geographic areas and sectors, likely making precise estimates difficult.

The most recent couple years of data are generally more uncertain than previous years. The most recent estimates contain provisional information on both activity rates and emissions factors and are subject to change as more complete information becomes available.

It is important to note that uncertainties in emissions are likely larger than indicated by the changes below. No change in the estimate does not mean there is no uncertainty, but simply that no new information was available to alter emission values. Emission values for 1980 presumably do not change because no new information was available. (It is also possible that earlier inventory values are not re-estimated as new information becomes available.)

Inventory	Data Year 1980	1985	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000
Coal – Electric Plants														
Trends 1995	0%	0%	0%	0%	0%	0%	0%	-1%	0%					
Trends 1996	0%	0%	1%	0%	0%	0%	0%	0%	0%	-1%				
Trends 1998	0%	0%	0%	0%	0%	0%	0%	0%	0%	-1%	-1%	0%		
Trends 2000	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	1%	1%
Coal – Industrial Combustion														
Trends 1995	0%	0%	0%	-4%	**-14%**	-3%	-6%	-1%	1%					
Trends 1996	0%	0%	0%	0%	0%	0%	0%	0%	0%	**34%**				
Trends 1998	0%	0%	0%	0%	0%	0%	0%	0%	0%	**12%**	**11%**	**11%**		
Trends 2000	0%	0%	0%	0%	0%	0%	0%	0%	0%	1%	0%	0%	**24%**	**29%**
Petroleum (all sectors)														
Trends 1995	-1%	-3%	-1%	**-11%**	0%	-3%	-4%	-9%	**-13%**					
Trends 1996	-1%	-3%	-1%	2%	4%	6%	4%	-3%	-3%	6%				
Trends 1998	-1%	**13%**	**17%**	**19%**	**24%**	**27%**	**25%**	**20%**	**22%**	**28%**	**28%**	**27%**		
Trends 2000	-1%	**13%**	**17%**	**20%**	**24%**	**27%**	**26%**	**20%**	**22%**	**44%**	**44%**	**42%**	**58%**	**64%**
Metals Processing														
Trends 1995	0%	0%	0%	-9%	3%	6%	**11%**	**23%**	**36%**					
Trends 1996	0%	0%	0%	0%	0%	0%	0%	0%	0%	**36%**				
Trends 1998	0%	0%	0%	0%	0%	0%	0%	0%	0%	**10%**	**11%**	10%		
Trends 2000	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	**32%**	**31%**
Total														
Trends 1995	0%	0%	0%	-3%	-1%	-1%	-1%	-1%	-2%					
Trends 1996	0%	0%	0%	0%	1%	1%	0%	0%	0%	4%				
Trends 1998	0%	2%	2%	3%	3%	3%	3%	2%	3%	4%	4%	4%		
Trends 2000	0%	2%	2%	3%	3%	3%	3%	2%	3%	6%	6%	6%	**10%**	**11%**

Table S- 7 – Comparison of different releases of EPA inventory estimates of SO_2 emissions with the 2002 release of the inventory. The amount, as a percentage, the given release differs from the 2002 release is shown. Note that not all years are shown. A positive value indicates a value that is larger than in the 2002 release.

S.15 Uncertainty Assumptions

The uncertainty values used are shown in Table S-8. In order to produce an uncertainty analysis without too many assumptions, uncertainty is assigned in broad source categories by country. Confidence intervals were set by specifying values for uncertainty in emissions factor and uncertainty in driving forces and combining the two in quadrature. Uncertainty was calculated for the following source categories: petroleum combustion, coal combustion, smelting, fuel processing, other process, and biomass combustion.

Uncertainty values for coal combustion assume increasing uncertainty in both activity levels and emissions factors in earlier years and in developing countries, similar to values in the literature (Schöpp et al. 2005, Gregg et al. 2008). As discussed above (§S.13) emissions from coal combustion in the countries of the FSU is particularly uncertain as coal consumption and aggregate sulfur content by country is not available for earlier years. To account for this, uncertainty from coal combustion in these countries was increased by a factor of 2 in 1970 and before (and by 1.5 in 1980).

We note that the assumed uncertainty bounds could overestimate uncertainty in regions where emissions from coal in sources such as electric power plants are subject to emissions limits and are directly measured with continuous emission monitoring systems (CEMS). This has a somewhat limited impact on total uncertainty since emissions from such sources, have been decreasing in recent years. In 2000, for example, about 40% of sulfur emissions from coal were from countries where monitoring is likely to be common (European Union, United States, Canada, Japan, Australia & NZ).

The two analyses above (§S.13, §S.14) indicate that uncertainty in petroleum emissions is often larger than for coal. Note also our finding that petroleum emissions in the United States appear to be underestimated in previous inventories (§S.8). A large portion of sulfur emissions from petroleum combustion in many countries is from residual fuel, where the sulfur content can vary with crude oil properties and also with the applicability of regulation given that use of heavy oil in industrial operations do not always fall under strict standards. In addition, the sulfur content of petroleum products will depend on refinery operations, crude oil characteristics, sulfur standards (and any exemptions to those standards), and the enforcement of standards. We, therefore, assume a larger uncertainty for the sulfur content of petroleum products.

As discussed above regarding coal assumptions, this might overestimate uncertainty in regions with comprehensive and well-enforced fuel sulfur content standards that include residual fuel use. Emissions from countries with strong and comprehensive sulfur standards are also likely to be declining, which will decrease the absolute magnitude of any such overestimate of uncertainty.

Uncertainty for other process emissions and biomass were also assumed to be larger, although these two sources globally comprise only a relatively small portion of emissions.

Changes in methodology, such as the treatment of coking coal, result in systemic error, and an overall offset in emissions. Coking coal comprised about 12% of total coal consumption in 1970, declining to 8% in 2005. Removal of coking coal from the emissions calculation in recent years decreased the estimate of emissions for developing countries, where total emissions were not calibrated to inventory values. Emissions from this sector could be either over or under estimated globally, depending on the accuracy of the emissions factor used. The systemic uncertainty component discussed in the main text was added to account for such issues.

	Coal			Petroleum		
Category	**S Content**	**Driver**	**Total**	**S Content**	**Driver**	**Total**
I. Recent-Country-Inventory	±10%	±5%	±11%	±20%	±5%	±21%
II. Older Inventory	±15%	±10%	±18%	±25%	±10%	±27%
IIa. OECD (pre inventory)	±20%	±15%	±25%	±40%	±15%	±43%
III. Other Countries	±20%	±20%	±28%	±40%	±20%	±45%
IV. Int Shipping	±20%	±20%	±28%			
IV. Int Shipping (earlier)	±30%	±30%	±42%			

	Smelting			Other Process, Biomass		
Category	**S Content**	**Driver**	**Total**	**S Content**	**Driver**	**Total**
I. Recent-Country-Inventory	±10%	±10%	±14%	±20%	±10%	±22%
II. Older Inventory	±20%	±15%	±25%	±35%	±15%	±38%
IIa. OECD (pre inventory)	±20%	±15%	±25%	±50%	±15%	±52%
III. Other Countries	±30%	±20%	±36%	±50%	±20%	±54%

Table S- 8 – Uncertainty values by country category.

The regions used for the uncertainty calculation are: Canada, United States, Western Europe, Eastern Europe, Japan, Australia & New Zealand, Former Soviet Union, China+, Middle East, Africa, South Korea, India, South and East Asia, and Central/South America. Region definitions are discussed in § S.1. The random uncertainty component was summed for each source within each of these regions, assuming, in effect, that uncertainties with a sector are perfectly correlated within each region, since similar regional assumptions were used (Equation 1 main text). Uncertainties are combined in quadrature between regions and between source.

An alternative calculation assuming no correlation between values at the country level, where the random component of uncertainty was combined in quadrature at the country level instead of the regional level, results a lower global uncertainty estimate (lower by 5-25%), depending on the year, due to greater statistical cancellation between countries.

Table S-9 shows the classification of uncertainty by country. The inventory codes 1–6 correspond to the numerical values given in Table 3 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- 9 – Uncertainty categories by country and time period (see Table 1 of main text). Values for 1990 are used for all years 1990 through 2005.

The lowest level of uncertainty (category I) is applied to countries with sectoral-level inventory values available, largely from UNFCCC submissions, where these appear to be complete such as the United States, and Western Europe. Some UNFCCC submissions were not complete, so the overall uncertainty was judged to be higher in these regions (Category II). This larger uncertainty value (Category II) was also assigned to years calibrated to older inventory data. Before the period where inventory data are available, uncertainty is assumed to be even higher still (Category IIa). Countries with no inventory data, or where inventories are inconsistent, were assigned the highest uncertainty level (Category IV).

For Western Europe countries not otherwise specified below, for example, the random uncertainty in emissions from petroleum combustion ($CI\_random_s^r$ in Equation 1 in the main text) were taken to be 38% in 1990 and beyond and 52% in 1970, with values linearly interpolated between these years.

Figure S-6 shows the total uncertainty bounds estimated here in comparison with a number of other global inventories. While estimates for many years are within the uncertainty bounds estimated here, there are still some significant differences, particularly for the years around 1990. The estimate of Stern (2006) is generally near the upper limit estimated here, and slightly higher than the upper limit in the 1940s and 1950s and the late 1980s. The estimate of Lefohn et al. (1999) is below the lower limit over the 1960s and 1970s, but slightly above the upper bound before 1920. The EDGAR 3.2 estimate is consistently above the upper limit as is the earlier estimate of Spiro et al. (1992) for 1980.

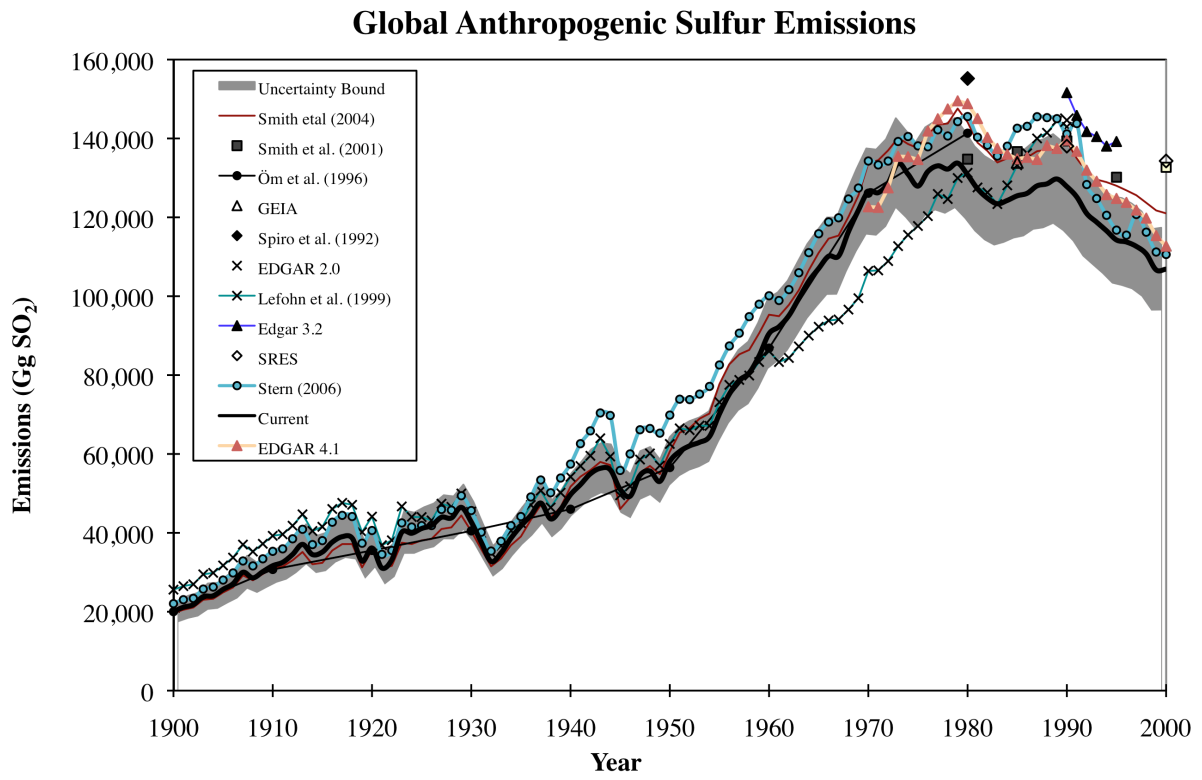

Figure S-6 – Figure 8 of the main text with the addition of the uncertainty bounds as estimated from this work.

Table S-10 shows uncertainty by source category as a fraction of emissions by that source. The relative uncertainty depends on the distribution of emissions between regions and the

uncertainty assumptions. Emissions from coal generally have the lowest relative uncertainty, with uncertainty from other categories generally higher. The highest relative uncertainty in earlier years is from shipping since both emissions factors and fuel consumption are uncertain.

Uncertainty as fraction of source

	1900 - 2005			
	Max	**Min**	**1900**	**2005**
Coal	19%	12%	17%	19%
Petroleum	29%	16%	28%	18%
Smelting	20%	15%	20%	18%
Other Process	29%	13%	29%	19%
Shipping	42%	28%	42%	28%
Biomass	36%	26%	26%	34%
Total	14%	8%	14%	11%

Table S- 10 – Uncertainty as fraction of emissions from each source in 1900, 2005, and the maximum and minimum over this period.

S.16 EDGAR 4.1 Inventory

Table S-11 shows the difference between the current estimate and the EDGAR 4.1 SO_2 estimate (JRC/PBL 2010). While global emissions are generally within about 10% of the current estimate, except for around 1980, the regional differences are much larger. The EDGAR 4.1 estimate was developed with a consistent methodology in all regions, but without calibrating to regional inventory data. This results in significant regional differences with the data presented here.

The largest differences between the two estimates occur around 1980, where EDGAR 4.1 emissions from the USA, Japan, and most developing and transitional countries are larger than the estimate here. For the USA, Europe, and Japan, where the current estimate is calibrated to inventory estimate, the EDGAR 4.1 estimate is substantially smaller by 1970.

The EDGAR values in the FSU are significantly larger than the estimate here, and also larger than the estimates of Ryaboshapko et al. (1996) for 1990 and 1985. While the two estimate are similar for 2005, the difference between the two estimates increases to 40% by 1985. The reason for this difference in trend is not known.

Emissions in Asia, Central & South America, and the Middle East are generally larger in the EDGAR 4.1 inventory. Emissions are generally lower in Africa and Mexico.

Emissions from international shipping are lower, perhaps because reported IEA energy data may have been used instead of the larger bottom-up estimates used here.

Relative differences between the two estimates are consistently large (greater than 30%) in Japan, the FSU, South & East Asia, South Korea, India, Argentina, and international shipping (Table S-11). These differences are likely due to a combination of different assumptions for sulfur content, sulfur retention in ash, and abatement measures.

The largest differences by sector are from energy and industry, indicating that the differences may largely stem from different assumptions from fossil fuel combustion. By 2005, there is also a significant difference in emissions from the domestic sector.

EDGAR 4.1 - this work	1970	1975	1980	1985	1990	1995	2000	2005
USA	-1,756	1,076	4,377	3,046	2,296	632	-1,308	-2,325
Canada	-850	-328	-341	-561	-180	-324	-70	-21
OECD90 Europe	-2,365	-2,024	-323	-2,235	-1,294	2,245	2,576	2,403
Japan	-2,386	759	1,707	1,408	1,547	1,395	909	654
Aus & NZ	364	380	391	255	219	25	51	44
FSU	4,709	8,495	10,069	6,017	7,774	2,800	1,075	49
China+	1,089	506	2,267	2,103	3,505	3,845	2,320	4,492
Middle East	59	-138	539	687	554	947	954	1,339
Africa	-672	-792	-640	-213	272	-67	-392	687
Central & South America	55	371	528	423	240	372	591	1,643
S&E Asia	433	463	548	368	451	1,052	739	862
Eastern Europe	-2,587	-2,021	-1,151	-2,412	-3,108	-1,463	-1,076	-1,655
Korea	456	430	569	723	678	878	1,383	1,152
India	458	502	534	629	692	827	957	1,098
Argentina	633	565	514	290	251	124	113	75
Brazil	158	195	237	15	-56	134	212	129
Mexico	-126	-125	-212	-99	-308	-291	-329	-251
Int. Shipping	-1,603	-1,640	-1,654	-1,154	-1,844	-2,717	-3,014	-4,206
Total	-3,928	6,675	17,958	9,291	11,688	10,413	5,691	6,167

Ratio: EDGAR 4.1/this work	1970	1975	1980	1985	1990	1995	2000	2005
USA	0.9	1.0	1.2	1.1	1.1	1.0	0.9	0.8
Canada	0.8	0.9	0.9	0.8	0.9	0.9	1.0	1.0
OECD90 Europe	0.9	0.9	1.0	0.9	0.9	1.2	1.3	1.4
Japan	0.6	1.3	2.3	2.5	2.6	2.5	2.0	1.8
Aus & NZ	1.2	1.2	1.2	1.2	1.1	1.0	1.0	1.0
FSU	1.3	1.5	1.5	1.3	1.4	1.2	1.1	1.0
China/CPA	1.1	1.0	1.2	1.1	1.2	1.2	1.1	1.1
Middle East	1.0	0.9	1.2	1.2	1.2	1.2	1.2	1.2
Africa	0.9	0.8	0.9	1.0	1.0	1.0	0.9	1.1
Latin America	1.0	1.1	1.1	1.1	1.1	1.1	1.2	1.5
S&E Asia	1.5	1.4	1.3	1.2	1.1	1.3	1.2	1.2
Eastern Europe	0.8	0.8	0.9	0.8	0.7	0.8	0.8	0.7
Korea	1.8	1.5	1.4	1.6	1.5	1.5	2.5	3.9
India	1.4	1.4	1.3	1.3	1.2	1.2	1.2	1.2
Argentina	4.8	4.6	4.5	3.4	3.2	1.9	1.8	1.6
Brazil	1.2	1.2	1.2	1.0	1.0	1.1	1.1	1.1
Mexico	0.9	0.9	0.9	0.7	0.6	0.7	0.6	0.7
Int. Shipping	0.8	0.7	0.7	0.8	0.7	0.7	0.7	0.7

Regional Difference	1970	1975	1980	1985	1990	1995	2000	2005
USA, Europe, Japan	-9,093	-2,210	4,609	-192	-559	2,809	1,101	-924
China + FSU	5,799	9,002	12,335	8,119	11,279	6,645	3,395	4,541
Rest of World	968	1,523	2,668	2,517	2,812	3,676	4,208	6,756
Int. Shipping	-1,603	-1,640	-1,654	-1,154	-1,844	-2,717	-3,014	-4,206
Total	-3,928	6,675	17,958	9,291	11,688	10,413	5,691	6,167
Diff as %	-3%	5%	14%	7%	9%	9%	5%	5%

Sectoral Differences	1970	1975	1980	1985	1990	1995	2000	2005
Energy & Fuel Proc	-2,944	2,037	6,213	2,109	7,277	8,521	5,066	4,761
Smelting	-806	-773	-384	-451	-104	-54	-485	-502
Other Process	-1,893	-998	-414	-165	-174	-1,015	-911	-375
Industry-Comb	5,312	8,711	13,472	9,282	8,043	8,139	7,716	9,660
Transport	-1,572	-1,075	-164	87	-739	-349	-35	-451
Domestic	-556	297	739	-571	-933	-2,274	-2,817	-2,884
International Shipping	-1,603	-1,640	-1,654	-1,154	-1,844	-2,717	-3,014	-4,206
Total	-4,060	6,562	17,811	9,138	11,529	10,253	5,521	6,003

Table S- 11 – Difference between the emissions estimate developed here and the EDGAR 4.1 emissions estimate in absolute values (Gg SO_2) and also as ratios. Emissions from open burning and aviation are not included. Positive values indicate emissions that are larger in the EDGAR 4.1 estimate than in this work.

S.17 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-7 and Table S-12 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.

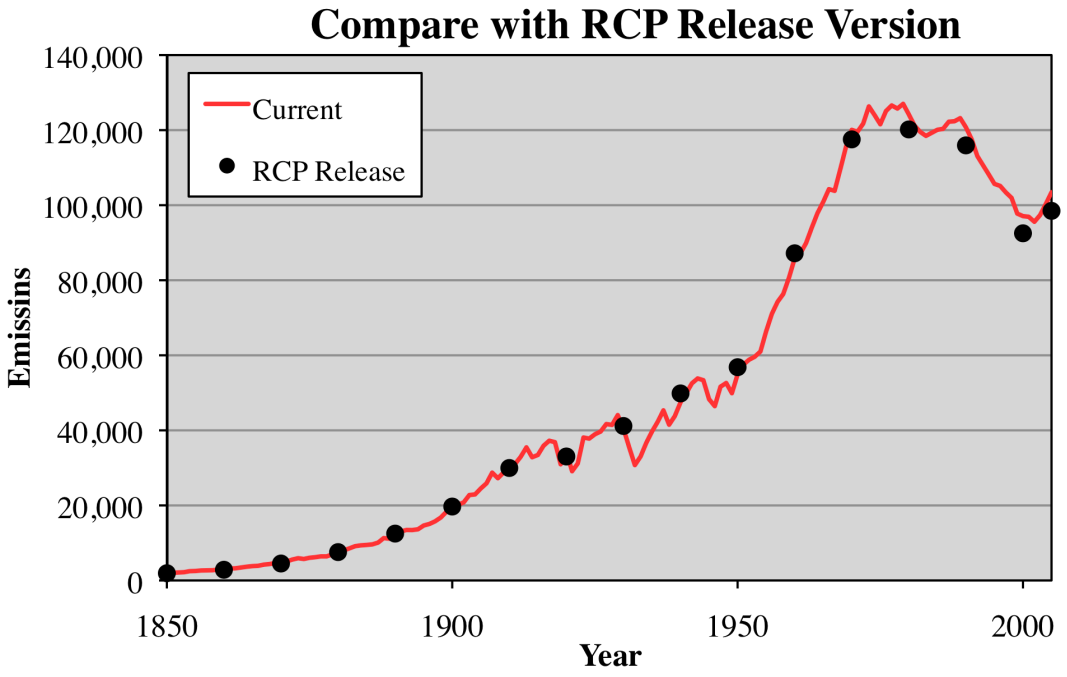

Figure S-7 – Current version (v 2.85) of the inventory as compared to the RCP inventory release for global emissions from fossil fuel combustion and process emissions, excluding international shipping.

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

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