**Atmospheric
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Anthropogenic sulfur dioxide emissions: 1850–2005

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Abstract

Sulfur aerosols impact human health, ecosystems, agriculture, and global and regional climate. A new annual estimate of anthropogenic global and regional sulfur dioxide emissions has been constructed spanning the period 1850–2005 using a bottom-up mass balance method, calibrated to country-level inventory data. Global emissions peaked in the early 1970s and decreased until 2000, with an increase in recent years due to increased emissions in China, international shipping, and developing countries in general. An uncertainty analysis was conducted including both random and systemic uncertainties. The overall global uncertainty in sulfur dioxide emissions is relatively small, but regional uncertainties ranged up to 30%. The largest contributors to uncertainty at present are emissions from China and international shipping. Emissions were distributed on a 0.5° grid by sector for use in coordinated climate model experiments.

1 Introduction

Anthropogenic emissions have resulted in greatly increased sulfur deposition and atmospheric sulfate loadings near most industrialized areas. Sulfate acid deposition can be detrimental to ecosystems, harming aquatic animals and plants, and damaging to a wide range of terrestrial plant life. Sulfur dioxide forms sulfate aerosols that are thought to have a significant effect on global and regional climate. Sulfate aerosols reflect sunlight into space and also act as condensation nuclei, which tend to make clouds more reflective and change their lifetimes. The radiative forcing change wrought by sulfate aerosols may be second only to that caused by carbon dioxide, albeit in the opposite direction (Forster et al., 2007).

Sulfur is ubiquitous in the biosphere and often occurs in relatively high concentrations in fossil fuels, with coal and crude oil deposits commonly containing 1–2% sulfur by weight. The widespread combustion of fossil fuels has, therefore, greatly increased sulfur emissions into the atmosphere, with the anthropogenic component now substan-

tially greater than natural emissions on a global basis (Smith et al., 2001).

Historical reconstructions of sulfur dioxide emissions are necessary to access the past influence of sulfur dioxide on the earth system and as base-year information for future projections (Lamarque et al., 2010). This paper presents a new estimate of

5 global and regional sulfur dioxide anthropogenic emissions over the period from 1850 to 2005. This work represents a substantial update of previous work (Smith et al., 2001; Smith et al., 2004) with newer data and improved methodologies. The emissions reconstruction presented here accounts for regional differences in the pace and extent of emission control programs, has annual resolution, includes all anthropogenic

10 sources, and provides global coverage. Fuel-based and activity-based (Eyring et al., 2009) estimates of shipping emissions were reconciled for recent decades and then extrapolated to 1850. A global mass balance for sulfur in crude oil was calculated as an independent estimate of petroleum emissions. Finally, a regional and global uncertainty analysis was conducted.

15 **2   Methodology**

Sulfur emissions from combustion and metal smelting can, in principle, be estimated using a bottom-up mass balance approach where emissions are equal to the sulfur content of the fuel (or ore) minus the amount of sulfur removed or retained in bottom ash or in products. Data limitations, however, make the bottom-up approach uncer-

20 tain. Countries with air pollution policies in place generally require detailed reporting of emissions, including direct measurement of emissions for many large sources. These data are likely to be more accurate than bottom-up estimates; therefore, we constrain our calculations to match country-level inventories where these data are available and judged to be reliable. This method produces an emissions estimate that is consistent

25 with the available country inventory data, contains complete coverage of all relevant emissions sectors (assuming the country inventory data are complete, see below), and is consistent across years.

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Emissions were estimated annually by country for the following sectors: coal combustion, petroleum combustion, biomass combustion, shipping bunker fuels, metal smelting, natural gas processing and combustion, petroleum processing, pulp and paper processing, other industrial processes, and agricultural waste burning (AWB). The approach was to first develop a detailed inventory estimate by sector and country for three key years: 1990, 2000, and 2005. These years were chosen due to data availability and because these years span a time period of significant change in emissions controls. Estimates for other years were constructed by interpolating emissions factors, while calibrating to country-level emissions inventories where those were available.

These sectoral emissions were then transformed to a set of standard reporting sectors (energy, industry, transportation, domestic, AWB) before downscaling to a 0.5° spatial grid. Descriptions of each element in this calculation are given below, with further details in the supplementary material.

2.1 Fossil fuel combustion

Emissions from coal and petroleum combustion were estimated starting with the regional emissions factors from Smith et al. (2001). A composite fossil fuel consumption time series was constructed (see supplementary material) using data from IEA (2006), UN energy statistics (1996), and Andres et al. (1999), who compiled historical estimates from Etemad et al. (1991). Country-level emissions estimates for Europe, North America, Japan, Australia, and New Zealand were compiled from: UNFCCC (2009) for 1990–2005; Environment Canada (2008), the EEA (2002), Fujita (1993), Mylona (1996), Gschwandtner et al. (1986), UK National Atmospheric Emissions Inventory (2009), USEPA (1996a), and Vestreng et al. (2007) as detailed in the supplementary material. Fossil-fuel emissions factors were adjusted such that total emissions matched inventory values for those years where inventory values are available.

For most European countries and the United States, emissions estimates by country are available from at least 1900. 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 is not included in the calculation of combustion emissions in recent years; any residual emissions from these uses are assumed to be included in the process emissions estimate. Emissions from coal used for coke production are included in earlier years before the introduction of by-product coke plants (see supplementary material). The assumptions for sulfur emissions from coal combustion in China and the countries of the Former Soviet Union are drawn from (Foell et al., 1995; Klimont et al., 2009; Xu et al., 2009) and (Ryaboshapko et al., 1996), respectively as discussed in the supplementary material. 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 is 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.

The emission estimates for other Asian countries developed here were compared to a number of regional estimates (Ohara et al., 2007; Streets et al., 2003; Zhang et al., 2009; NIER 2008) and the estimates here were adjusted to better match these studies

(see supplementary material). These estimates differ from each other in some countries and sectors, highlighting a need for improved inventories in this rapidly changing region.

Petroleum emissions are particularly difficult to estimate in the absence of detailed, country-level inventory data. For regions without detailed emission inventory data, default values for sulfur contents were used. Emissions factors were adjusted downward in recent years to account for increased stringency of sulfur content standards as reported in UNEP (2008).

As an overall check on the petroleum assumptions used, a 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 (see supplemental information). The final mass balance estimate for global sulfur emissions from petroleum as compared to the inventory estimate is shown in Fig. 1 for the period 1950–2005, where petroleum emissions are a substantial fraction of the global total. We estimate that in the early portion of the 21st century the world passed a threshold where slightly over half of the sulfur contained in crude oil is removed at the refinery. Thus, global emissions from petroleum have been relatively flat since 1985 despite increases in petroleum consumption. Mass balance estimates using two assumptions for the amount of sulfur that is retained in non-energy use of petroleum products and non-combusted products, such as asphalt, are shown in order to reflect uncertainty in this parameter.

The resulting estimate of global petroleum sulfur is consistent with the global inventory value for petroleum emissions for most years. The mass-balance estimate indicates that there may be an underestimate on the order of 5000 Gg SO_2 in the inventory data during the late 1970s. The 10% retention mass-balance case results in a lower global estimate than the inventory data past 1990. The 5% retention case matches well with the inventory during this time, perhaps indicating the impact of increased refinery efficiency. While this comparison validates the overall approach, significant errors could still be present at the country level.

As noted in Smith et al. (2004), the aggregate emissions factor from petroleum prod-

ucts in the US as implied by current estimates (Gschwandtner et al., 1986; USEPA, 1996a) before about 1980 appeared to be lower than those for other world regions. In order to better estimate petroleum-related emissions in the United States, a mass balance estimate for the United States was constructed, accounting for imports and exports of crude oil by region and imports and exports of petroleum products (EIA 2009). This mass balance indicated that sulfur emissions from petroleum were substantially underestimated prior to 1980 by 30–50% (see Supplimentary material). The increased emissions are, in part, due to imports of residual fuel, and a decrease prior to 1980 in reported bunker fuel use, which is, in effect, an export of sulfur from the continental areas. Emissions from petroleum prior to 1980 were increased to be within 20% of the total sulfur as indicated in the mass balance estimate, allowing, conservatively, for retention of sulfur in non-combustion uses.

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.

2.2 International shipping

Emissions from international shipping were estimated using a combination of reported data and bottom-up estimates of shipping fuel consumption. We constructed a composite global estimate of shipping fuel consumption following Eyring et al. (2009) by using values from Eyring et al. (2009) 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 es-

timated 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, b, c), 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 2008, 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. 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. This has a substantial impact on emissions as the sulfur content of residual oil can be much higher than that of distillate fuels. The residual fraction is kept constant prior to 1971 due to a lack of data.

These are global values for fuel consumption, which would ideally need to be reconciled with regional energy data. As noted by Eyring et al. (2009) for 1896–2007 and Endresen et al. (2007), the IEA energy data used for terrestrial coal and oil consumption here substantially underreport shipping fuel consumption. Comparison of the country and regional level time series developed here with IEA data finds that the difference is often within the IEA "statistical error" category. There is generally no other

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consumption category in the IEA data that is large enough to include the difference between our regional fuel consumption estimate and the IEA reported bunker fuel use. We presume, therefore, that the difference is unreported consumption and no adjustment to the IEA consumption data has been made.

5   In order to estimate sulfur dioxide emissions, we calibrate to the year 2000 value from Eyring et al. (2009) of 11 080 kt SO_2/year. Using the global division between fuels derived above, we match this value using the following sulfur contents: residual: 2.86% and distillate: 1.25%. Further, the average value for sulfur in bunker coal is assumed to be 1.1%. For simplicity these values are kept constant over time. This is a significant

10   source of uncertainty in international shipping emissions, which is considered further in Sect. 4.

The fossil fuel consumption data from IEA used to estimate combustion emissions (Sect. 2.1) were processed to exclude fuels reported as international bunkers but included fuels used for domestic shipping and fishing, using sectoral definitions as dis-

15   cussed further in Sect. 2.5. In parallel with this assumption, domestic shipping and fishing emissions in the inventory data were included in the surface transportation sector. Emissions due to domestic shipping and fishing emissions from inventory data were subtracted from the total shipping estimate in the tables and figures reported below to avoid double counting.

20   **2.3   Metal smelting**

Smelting emissions were estimated using a mass balance approach where emissions are equal to the gross sulfur content of ore minus reported smelter sulfuric acid production, with estimates adjusted to match inventory data where available (see supplementary material). Smelter production of copper, zinc, lead, and nickel was tabulated

25   by country, primarily using USGS mineral yearbooks and predecessor publications for 1930–2005 and a variety of supplemental sources, particularly for earlier years (see supplementary material). Some mineral sources and production technologies emit minimal sulfur and these were accounted for where they could be identified. Further

details are available in the supplementary material.

A number of data biases can affect the estimate of sulfur emissions from metal smelting. To evaluate this possibility, we can compare emissions from inventories to emissions estimated purely using the mass balance approach. For the period 1990–2005, emissions by sector were available for a number of countries. When compared to emissions from the mass balance approach, the inventory values for fifteen countries in 1990 appear to be significantly lower than those estimated by the mass balance approach, although the opposite was the case in the United States. This indicates that either the sulfur content of ore was overestimated or that the amount of sulfur removal was underestimated. The latter may be the more likely possibility, given that the sulfur recovery tabulations from USGS are not necessarily complete, since data will be more available for commodities, such as ores, which are internationally traded than for sulfuric acid, which might be used locally. While these changes impact estimates of emissions from smelting, this has only a small impact on total emissions for recent years since total emissions for most of these countries are constrained by inventory values.

These comparisons indicate that emissions from areas without inventory data could be overestimated if sulfur removal data are underreported. In contrast, there are several regions with inventory data where sulfur content of ore appears to be larger than default values, which could also be the case in regions without inventory data, leading to an underestimate. These comparisons emphasize the importance of site-specific information in order to accurately estimate metal smelting emissions.

2.4 Other emissions

Natural gas deposits often contain significant amounts of sulfur compounds, particularly hydrogen sulfide, that are either flared, thus producing sulfur dioxide, or removed and converted to a salable product. Data for natural gas production emissions were found only for the United States (US EPA 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 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.

While natural gas distributed for general use has minimal sulfur, natural gas containing larger amounts of sulfur, known as sour gas, can be combusted for industrial applications, resulting in sulfur emissions. The US EPA inventory contains an estimate for emissions of 376 kt SO_2 in 2000, and the US inventory estimates were used for US emissions from this source. This is the only explicit inclusion of this source in our estimate. Any sour gas emissions in industrialized countries were presumed to be included in country inventories, but no data were available to explicitly account for these emissions, which would be included, indirectly, though our calibration procedure.

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 US EPA 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 from 1961–2005 (FAOSTAT, 2009), supplemented by data from Mitchell (1998a, b, c), McKeever (1987), and Statistics Canada (Leacy, 1983).

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).

5 Where updated 2005 data were not available, year 2000 values were used.

Emissions from biomass combustion (exclusive of open burning) were estimated using a historical reconstruction of biomass consumption based on the estimate of Fernandes et al. (2007) combined with IEA data (see supplementary material). Biomass emissions factors for sulfur dioxide range over an order of magnitude depending on the

10 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 (US EPA, 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[1] resulted in estimates of 1.1 kg/Mg for China and

15 0.4 kg/Mg for India (assuming an energy density of 16 GJ/t). 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

20 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.

Emissions from agricultural waste burning on fields were from EDGAR 4.0. Production statistics for 24 crop types (FAO, 2007) were combined with information on the

25 fraction burned on the fields (UNFCCC, 2008; IPCC, 2006; Yevich and Logan, 2003) and emission factors from Andreae and Merlet (2004). Emissions from waste burning were calculated in a similar manner, although these are small and the data for this sector are incomplete. Emissions from agricultural waste burning and waste were included

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[1]http://gains.iiasa.ac.at/index.php/gains-asia

in the RCP emissions release described in Lamarque et al. (2010).

2.5 Emissions by sector and grid

The emissions estimates developed here were mapped to a standard set of reporting sectors and then downscaled to a 0.5° spatial grid as part of the production of historical data for the new RCP scenarios (Moss et al., 2010; Lamarque et al., 2010). The reporting sectors for the RCP historical data were the following: energy transformation, residential/commercial, industry, surface transportation, agricultural waste burning on fields, waste burning, solvent use, and agricultural activities (non-combustion). There are no appreciable SO_2 emissions from the last two sectors.

Emissions smelting and other industrial processes were mapped to the industrial sector, biomass fuel emissions to the domestic sector, and fossil fuel extraction and processing emissions to the energy sector. Emissions from coal and petroleum combustion were split into the first four sectors above by using inventory data, sector-specific emissions factors, and IEA fuel use data, where these data were available (see supplementary material). For historical periods (generally before 1970 or 1960, see supplementary material), 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 for earlier years. 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.

The emissions estimate was distributed onto a 0.5° resolution global grid for each decade from 1850 to 2000. The sub-national split within a grid cell was estimated by using the 2.5 min national boundary information from the Gridded Population of the World dataset (CIESIN and CIAT, 2005). From 1960 through 2000, emissions were distributed using a preliminary version of the year 2000 emissions distribution from the EDGAR 4.0 project, separated into energy sector combustion, industrial combustion

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and other industrial, transportation combustion, and agricultural waste burning on fields within each country.

The EDGARv4.0 energy sector emissions were distributed on a 0.1° grid based on a combined allocation grid using information from Carma[2], 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 crop land 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.

For 1850–1900, emissions from combustion and other IND sectors for each country were distributed using the HYDE gridded population distribution (Goldewijk, 2005). The emissions distribution for each country was interpolated from the "modern" grid in 1960 to the population-based grid in 1900 by linearly increasing the weighting for the population-based distribution in each year 1950 to 1910 and decreasing the weighting for the "modern" grid, until a pure population-based grid is used in 1900.

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.

The emissions grids were produced to facilitate the use of these data in global modeling experiments. In most cases, the distribution of emissions within each country is determined by proxy data, not by actual emissions data. Alternative methods of down-

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[2]http://www.CARMA.org

scaling these emissions estimates to a spatial grid (van Vuuren et al., 2010), including incorporation of emissions measurements, would likely produce improved emissions distributions. No consideration of country boundary changes was made during the emissions gridding procedure. Incorporation of these changes over time was beyond the scope of this project.

3 Results

Global SO_2 emissions from 1850–2005 by sector are shown in Fig. 2. To obtain total anthropogenic emissions, extrapolated emissions from forest and grassland fires (Van der Werf et al., 2006; Schultz et al., 2008; Mieville et al., 2010) as used in the RCP historical inventory exercise (Lamarque et al., 2010) are also included. Table 1 presents a summary of total emissions, exclusive of open burning, by region for decadal years. Global emissions peaked in the 1970s after a nearly monotonic increase over two decades. Emissions have declined overall since 1990, with an increase between 2002–2005 due largely to strong growth of emissions in China.

Sectoral and regional emissions trends are shown in Figs. 2 and 3. Emissions from petroleum combustion have become a smaller portion of total emissions, with a large portion of estimated petroleum SO_2 emissions now originating from international shipping, a sector where high sulfur bunker fuel could, until recently, be used without restriction in most of the world. Trends in emissions from coal combustion include a steady decline since the 1970s in Europe and North America combined with large changes in coal combustion in the states of the Former Soviet Union after 1990, and a large increase in coal combustion in China in recent years. Emissions from metal smelting have decreased since their peak around 1970 due to increased sulfur removal, in part due to the widespread introduction of flash smelter technologies.

As seen in previous work (Smith et al., 2001, 2004), we find that there has also been a shift in the regional distribution of sulfur emissions, with an increasing proportion of emissions from Asia. Until the middle of the 20th century, emissions were dominated

by Europe and North America. Since that time, emissions from other regions, Asia in particular, have increased. Since the mid-1970's, an increasing fraction of global emissions have originated from Asia. Emissions from other regions (Africa, Middle-East, South America and Latin America) have also increased, but to a lesser extent. Emissions from China have increased to 28% of the estimated global total in 2005.

The annual global emissions estimate is similar before 1970 to the previous estimate using similar methodologies (Smith et al., 2001), albeit with some larger regional differences. Past 1970, the current estimate is lower, due in large part to lower emissions estimates for China and the Former Soviet Union, although emissions in these regions are particularly uncertain.

4 Uncertainty

It is useful to examine uncertainty in emissions by source and region. To our knowledge, this is the first consistent estimate of global and regional uncertainty in sulfur dioxide emissions. For this estimate, we apply a relatively simple approach to uncertainty analysis whereby a set of uncertainty bounds are applied to broad classes of countries. This is warranted in large part since, as noted by Schöpp et al. (2005), limited data are available to specify parameter uncertainty bounds, leading to bounds that are generally specified through expert judgment. This is particularly true for developing countries. In addition, sulfur dioxide emissions are principally determined by fuel sulfur content and not technology-specific emissions factors, at least in the absence of emissions controls. Data on fuel sulfur content are sparse in general, and those that contain uncertainty information even rarer. These considerations make a more complex assessment of global uncertainty unwarranted at this time.

The first component of the uncertainty analysis considers errors in the individual components of the emissions calculation. The set of uncertainty bounds given in Table 2 are applied to countries categorized depending on the estimated quality of the data used to construct the inventory values (see supplementary material). Uncertain-

ties are applied separately in each country to emissions from the following sources: coal, petroleum, biomass, fuel processing, smelting, and other process. Uncertainties in each of these categories are assumed to be independent and are combined in quadrature. Conceptually, aggregate uncertainty can be divided into uncertainty in driving forces, such as fuel consumption or smelter metal output, and uncertainty in sulfur content (and assumptions such as ash retention) that as shown in Table 2. However, only the total values are used in this calculation. The values in Table 2 are based on the author's judgment and are similar to other estimates in the literature (Schöpp et al., 2005; Gregg et al., 2008; Eyring et al., 2009).

Because common assumptions and data sources are used for large portions of the world, we assume for simplicity that uncertainties with each sector are perfectly correlated within 14 world regions. An alternative calculation assuming no correlation between values at the country level results in lower uncertainty at a global level by 3–27%, depending on the year.

This procedure assumes uncertainties are symmetric. This is likely not strictly true since, for example, sulfur removal (for petroleum and metal smelting) is bounded above, sulfur retained in ash is bounded below, and some emissions drivers have potential biases in one direction – for example, underreporting of consumption (Logan, 2001). It is not clear, however, if a more nuanced calculation is warranted given the number of assumptions that would need to be made.

The uncertainty estimate calculated as described above results in uncertainty bounds on annual global total SO_2 emissions that are relatively small: 4–8% over the 20th century. This low value is due to cancellation between sectors and regions. This uncertainty level would appear to be unrealistically low given that a number of previous global sulfur dioxide emissions estimates do not fall within this estimated uncertainty bound (Smith et al., 2001; Öm et al., 1996; Van Aardenne et al., 2001; Lefohn et al., 1999; Spiro et al., 1992; Stern, 2006). The reason is that additional, essentially correlated uncertainties are present that add to the uncertainty value estimated above. Examples include reporting or other biases in global data sets for energy, sulfur re-

moval, and other driver data, methodological assumptions, and the use of common default assumptions for sectors where little data exists. Comparing the present inventory with that of Smith et al. (2004), for example, indicates that the differences between these two estimates involve several methodological and data changes that impacted
5 emissions estimates over multiple world regions.

To include the potential impact of such correlated effects, we add to the uncertainty estimate for each sector an additional uncertainty amounting to 5% of total emissions (half this value for countries with well-specified inventories), with the additional uncertainty combined again in quadrature between sectors. This latter assumption is made
10 since there is little overlap in assumptions between either sulfur contents, emissions controls, or driver data between the broad sectors used in this calculation. The global value of the correlated uncertainty component is less than 3% since 1960, due to a cancellation across sectors, increasing to 4% by 1920 as emissions from coal combustion become more dominant.

15 The addition of this correlated component to uncertainty has a large impact on the final uncertainty value. The uncertainty range is increased by a factor of 1.4 to 1.7, depending on the year. Even with this component, however, the global uncertainty in sulfur dioxide emissions over the 20th century is still only 6–12%. The global emissions estimate of Smith et al. (2004) now largely falls within this expanded uncertainty
20 range. While this component has a large relative impact on the global uncertainty, the impact of this assumption on regional uncertainties is somewhat smaller. For example, the largest uncertainty in recent years is in emissions from China. The addition of this correlated component increases the magnitude of the total estimated uncertainty for China emissions by a factor of 1.2 (from 24% of total emissions to 28% of total
25 emissions).

The combined uncertainty bounds for global emissions and emissions from China are shown in Fig. 4. The uncertainty bounds are not wide enough on a global level to change the overall character of emissions over time, with global emissions peaking in the early 1970s, decreasing until 2000, and increasing slightly in recent years. In 2000,

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the estimated uncertainty in global emissions is $\pm 9\,000$ Gg SO_2.

The estimated uncertainty bounds for the United States are also shown in Fig. 4. The uncertainty range for the United States is smaller, even in earlier years, than the estimate for China. A portion of this difference is due to the assumed lower uncertainty bounds (Table 2). Also contributing to a lower overall uncertainty estimate is that emissions from the United States throughout the 20th century are from a wider variety of sources as compared to China, where emissions are dominated by coal consumption in all periods. This results in a larger statistical cancelation of uncertainties between sectors in the United States. Offsetting this somewhat is a similar relative impact of systematic uncertainties.

The regional uncertainty values, before combination to global values as described above, are shown in Fig. 5. China is the largest single contributor to emissions uncertainty since about 1980, with an estimated uncertainty in 2000 of ± 5200 Gg SO_2. The second largest source of uncertainty in 2005 is emissions from international shipping. The largest contributors to uncertainty have changed over the years, with countries of the Former Soviet Union and Western Europe dominant during the 1960s and 1970s, and the United States dominant during the early to mid-20th century.

Sulfur emissions are less uncertain than emissions of most other air pollutants because emissions depend largely on sulfur contents rather than combustion conditions. There is a large contrast with emissions of another important aerosol, black carbon. Bond et al. (2004) estimated global fossil black carbon emissions in 1996 as 3.0 Tg C, with an uncertainty range of 2.0–7.4 Tg C, or +150% and −30%, with the upper range an order of magnitude larger than the uncertainty in sulfur dioxide emissions estimated here.

Note that uncertainty in the spatial allocation was not assessed, but will likely be much higher than the uncertainty in national or sectoral emissions totals due to the simple downscaling methods used here.

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In 1850, global sulfur dioxide emissions over land areas were split roughly evenly between emissions from open burning and anthropogenic industrial activities. Over the next 50 years this changed dramatically as anthropogenic sulfur dioxide emissions increased by an order of magnitude, driven by increased use of coal. In the early 20th century the steady growth of emissions was slowed by a global depression and the second world war followed by the post war economic expansion, resulting in an unprecedented absolute rate of emissions growth averaging 3400 Gg/decade from 1950 to 1970.

For most of this period, sulfur dioxide emissions increased in proportion to activity levels. This began to change dramatically after 1970 due to concern over the impacts of these emissions on regional scales. Precursors to this change are evident in smelting and petroleum refining sectors. The amount of sulfur removed from crude oil has been steadily increasing since 1958 (Fig. 1) and a similar pattern exists for metal smelting, where around half the sulfur contained in metal ores was removed at the smelter by 1980. Removal of sulfur from coal combustion by means of post-combustion scrubbers began somewhat later, but is now a major driver of sulfur emission reductions. Shifts in coal supply to favor lower sulfur coal have also been a factor, although it is difficult to estimate the magnitude of this effect.

The current estimate is compared to other estimates of global sulfur dioxide emissions in Fig. 6. The current estimate is somewhat below many recent estimates, particularly in the 1970s and 1980s, including the previous estimate by Smith et al. (2004) using a similar methodology, but at a more aggregate scale. The major differences between these two datasets over this time period are lower emissions estimates for China and the countries of the Former Soviet Union. Other differences include updated information on sulfur removals from refineries and smelters, and improved methodologies. The estimate here for 1990, 1995, and 2000 agrees very closely with that of Cofala et al. (2007), in part due to the use of similar assumptions for China and calibration to

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country inventory data for many OECD countries.

The uncertainty bounds estimated in Sect. 4 encompass many of the other estimates, although the Spiro et al. (1992) and EDGAR 3.2 (Olivier and Berdowski, 2001) estimates lie above the uncertainty range, as does the estimated year 2000 emissions data point used in the SRES emissions scenarios. The uncertainty analysis conducted here highlights the potential importance of systematic errors and biases in emissions estimates, particularly for emissions such as sulfur dioxide, where most input values are only weakly correlated between regions. While the uncertainty estimates here are based on the authors' judgment, the relatively small global uncertainty that results indicates that a more complex global uncertainty analysis may not be warranted. Regional uncertainty can be far higher than global uncertainty, however, and more detailed uncertainty analysis of high-emitting regions may be useful to better bound current and past environmental impacts of sulfur dioxide emissions.

When comparing data sets, it should be noted that most of these estimates rely on similar, if not identical, data sets for historical fossil fuel use and for historical emissions from Europe and the United States. Errors or biases in these data, such as the apparent underestimate of SO_2 emissions from petroleum products in the United States prior to 1980, are likely to be common to most of these estimates. As demonstrated in Sect. 4, methodological biases and systematic errors can make a large contribution to uncertainty, particularly for SO_2 emissions, which depend on characteristics that are largely uncorrelated between regions.

It is also useful to compare 2005 emissions with previous estimates from global scenario projections. The 2005 emissions, estimated here as $115\,510\pm12\,850$ Gg, largely overlap with the range of future SO_2 emissions scenarios from Smith et al. (2005). This range is significantly lower than the SRES scenario projections for 2005, but does overlap with many of the scenarios in the post-SRES literature (Van Vuuren et al., 2008). The recent RCP emissions scenarios (Moss et al., 2010) closely match the range estimated here since they were calibrated to an earlier version of this inventory (see supplementary material).

Emissions in Asia are of particular interest and it is useful to compare emissions here with previous estimates. As shown in the supplementary material, even recent estimates for SO_2 emissions in Asia (Ohara et al., 2007; Streets et al., 2003; Zhang et al., 2009; Klimont et al., 2009) sometimes differ substantially. Focusing on China and India, the two countries with the largest emissions, these recent estimates all lie within the uncertainty range estimated here, with the REAS estimate for 2000 consistently larger than the other estimates quoted here, but near the upper bound of our uncertainty range for both China and India (although 1% over our uncertainty bound for India). The estimate here for China is within a few percent of the estimate in Lu et al. (2010) for 2000–2005. The current estimate for India is close to the estimate from Garg et al. (2006) in 1985 but diverges to be 30% higher than the Garg et al. value by 2005, but similar to other recent inventories (supplementary material).

The current estimate for China is 20–30% lower than earlier estimates based on the RAINS Asia project (Arndt et al., 1997; Streets et al., 2000; Streets and Waldhoff, 2000; Klimont et al., 2001) for 1895–1990. This difference is within the estimated uncertainty range. While the current estimate for India for 2000 and 2005 is similar to a number of other regional estimates for these years (although higher than Garg et al., 2006), the current estimate is 40% lower than the circa 1987 estimates from Arndt et al. (1997) and Streets et al. (2000). Note that such historical differences are not limited to developing countries, as indicated above (Sect. 2.1) for past emissions from petroleum in the United States.

It is not clear if such historical differences are due to variation in energy consumption data, parameter assumptions such as sulfur content or ash retention fraction, or other methodological differences, such as treatment of shifts in petroleum product consumption and sulfur contents. Analysis of these differences is beyond the scope of the current project. Given that the current estimate is similar to recent literature for later years, there may be have been changes in assumptions and methodology in recent estimates that would imply changes in these earlier emissions estimates as well. Because emissions in China are dominated by coal consumption, any uncertainty in coal

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emission parameters will translate directly into uncertainty in total emissions. A historical analysis of coal production and consumption in China, particularly data on coal production by source in order to track changing sulfur contents, would be especially useful in better determining historical emissions.

Future estimates of historical sulfur dioxide emissions could be improved through improved regional data sets for fossil fuel consumption, fuel properties, and industrial process drivers (including metal smelting amounts, ore properties, sulfuric acid production, and pulp and paper production). Limited information was available for sulfur dioxide emissions resulting from petroleum and natural gas extraction operations and the use of sour gas, even though these sources could be regionally significant. Many historical data series are only readily available at the country level. Improved spatial estimates of historical emissions would require sub-national consumption and activity information, particularly for geographically large countries such as the United States, Russia, and China.

Differences among estimates of both current and past emissions point to the need for further research to identify current and historical fuel sulfur contents and the characteristics of fuel-emissions technology combinations (such as sulfur retention in ash). While the bottom-up estimation methods used, in part, in this work are appropriate for countries with few emissions controls, direct emissions monitoring of large sources will become even more important for accurate emission estimates as the use of sulfur control technologies becomes more widespread.

The use of satellite observations (Krotkov et al., 2008; Li et al., 2010; Lu et al., 2010; Gottwald et al., 2010) to verify trends shows promise, although estimation of absolute emission values through use of satellite data is more difficult. Figure 7 shows the SO_2 column concentration above Eastern China as estimated by Gottwald et al. (2010) using satellite data. The increase in estimated SO_2 column from 2000 to 2005 is about 1.5, which is the same change as in total emissions over China from this period. The comparison for the earlier period is less clear. The change from 1996/1997 to 2000/2001 in the satellite estimate indicates a slight increase in concentrations while

the inventory estimate decreases over this period, taking the average over the indicated years. Part of this difference could be due to shifts in SO_2 source distribution over this time period. Concentrated emissions from large sources, which are also lofted higher into the atmosphere, are more likely to be detected by satellite instruments. From 1995 to 2000, we estimate that fraction of total emissions from power plants in China increased from 46% to 60%, which is in the right direction to help explain the difference in trends. Meteorological and atmospheric chemistry effects on SO_2 transport and lifetime will also affect the relationship between SO_2 column measurements and emissions. Additional uncertainty arises from changes in aerosol loading leading to changes in satellite sensitivity. See Gottwald et al. (2010) and the papers cited above for further discussion of these effects. It is not yet clear how these uncertainties on the detection side compare to the uncertainty in the inventory-based emissions estimate as quantified here.

Historical estimates of sulfur dioxide emissions are necessary for estimating past trends in acid deposition, the associated impacts, and past climate forcing. The current estimate represents a consistent global data set with annual resolution that can be used for historical modeling studies. The annual emissions data described in this paper are available from the corresponding author. The 0.5˚ gridded emissions data released for the RCP project are available at the RCP web site (see also the supplementary material).[3]

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/10/16111/2010/ acpd-10-16111-2010-supplement.pdf.

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Table 1. Total emissions estimates by decade (for the indicated year) and region (Gg SO$_2$), not including open biomass burning (forest, grassland, and agricultural waste burning) or emissions from waste.

Region	1850	1860	1870	1880	1890	1900	1910	1920	1930	1940	1950	1960	1970	1980	1990	2000	2005
USA and Canada	311	707	1380	2701	5443	9345	16 424	20 436	21 193	21 475	24 513	25 849	34 980	27 809	24 066	17 054	15 131
Mexico	1	2	2	2	17	52	218	254	331	404	658	766	1003	2003	2729	2991	2145
Central America	7	1	1	1	1	1	9	17	115	84	187	444	760	816	778	867	708
South America	33	56	68	76	75	79	278	408	757	1101	1346	2740	3648	5328	5314	4719	4432
Western Europe	1292	1929	2754	3972	5295	7213	9323	9035	11 649	12 519	13 974	20 599	29 329	26 759	18 206	7998	6242
Central Europe	34	75	219	518	903	1323	1756	1448	2280	3334	3942	7235	11 462	13 589	12 316	5704	4832
Russia	40	39	54	85	125	278	389	120	517	1897	2565	5744	7926	10 143	10 632	6352	5975
Ukraine	7	9	15	33	55	140	192	76	260	907	1710	3249	4320	5505	4921	1548	1470
Other Former Soviet Union	6	6	7	10	19	39	39	21	125	525	695	2864	3684	3777	4116	2516	3338
China	162	147	138	140	142	148	357	458	581	1089	1070	8261	7327	11 981	17 194	21 393	32 673
Japan	25	26	27	54	119	261	533	1081	1347	1641	1099	2188	5337	1317	975	885	834
Other South and East Asia	25	27	29	32	35	49	70	107	213	325	241	780	1796	3654	5489	6330	6111
India	35	39	43	48	64	102	161	221	281	338	425	702	1114	1674	3302	5363	6275
Australia and New Zealand	9	24	32	53	61	149	264	213	339	466	486	927	1553	1692	1648	2438	2606
South Africa	1	2	3	11	20	29	170	271	252	399	588	833	1491	1860	2283	2392	2477
Africa	11	12	14	16	18	20	26	50	220	823	1087	2070	3168	3784	3350	3322	2,690
Middle East	3	3	4	4	5	5	5	11	20	136	324	572	1179	2490	3436	5218	5490
International Shipping	61	120	208	365	587	892	1450	1927	2274	2233	3247	4680	6469	6607	7041	9779	12 078
Global Total	2063	3224	4996	8120	12 983	20 126	31 665	36 155	42 756	49 694	58 158	90 502	126 544	130 788	127 795	106 869	115 507

Table 2. Uncertainty bounds by country category and emissions type. The energy category includes coal, petroleum, and biomass combustion and fuel processing.

Category	Energy			Industrial Processes		
	S Content	Driver	Total	S Content	Driver	Total
I. Recent-Country-Inventory	–	–	5%	–	–	10%
II. Older Inventory	10%	10%	14%	20%	15%	25%
IIa. OECD (pre inventory)	15%	15%	21%	20%	15%	25%
III. Other Countries	20%	20%	28%	30%	20%	36%
IV. Int Shipping	20%	20%	28%			–
IV. Int Shipping (earlier)	30%	30%	42%			–

Global Sulfur Emissions From Petroleum

Legend:
- S Emissions Petr
- Pet Mass Bal-10%ret
- Pet Mass Bal-5%ret
- S Removals (as SO2)
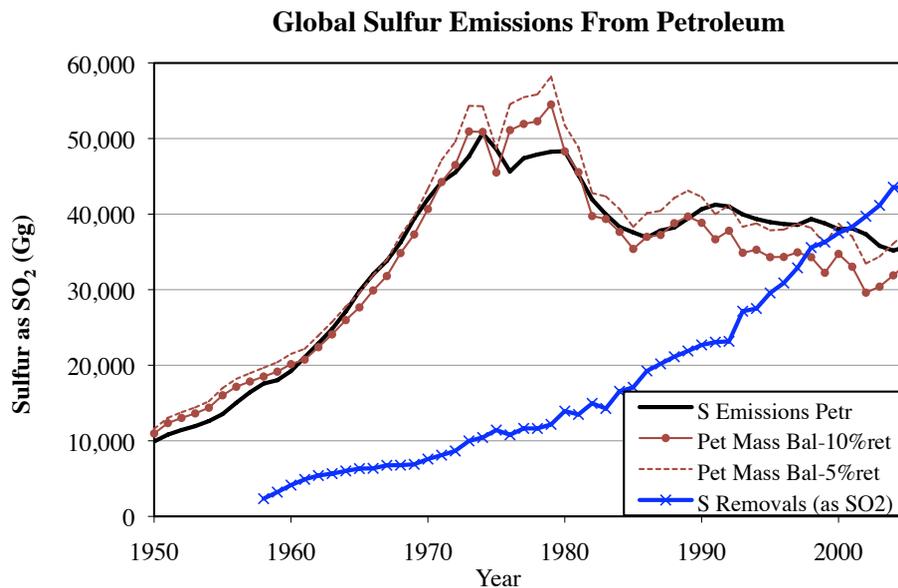
Fig. 1. Global petroleum emissions from the present inventory compared to estimates from a global crude oil mass balance (see text) using two different assumptions for the fraction of total sulfur retained in non-combusted products. The inventory data include terrestrial and international shipping combustion and process emissions from refineries. Also shown is the estimated global amount of sulfur removed at refineries as SO_2.
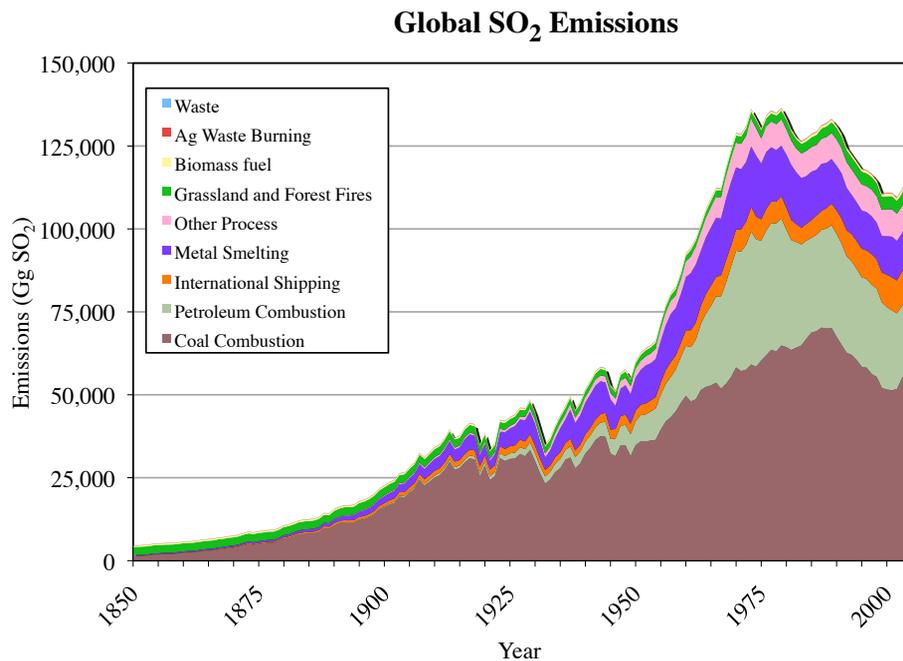
Fig. 2. Global sulfur dioxide emissions by sector.

Global Anthropogenic SO$_2$ Emissions

Legend:
- North America
- Europe
- FSU
- East Asia
- South-East Asia & Aust/NZ
- India
- South and Central America
- Middle East
- Africa
- International Shipping

X-axis: Year (1850–2000)
Y-axis: Emissions (Gg SO$_2$), 0 to 50,000

Fig. 3. Global sulfur dioxide emissions by region.

(a)

(b)

(c)
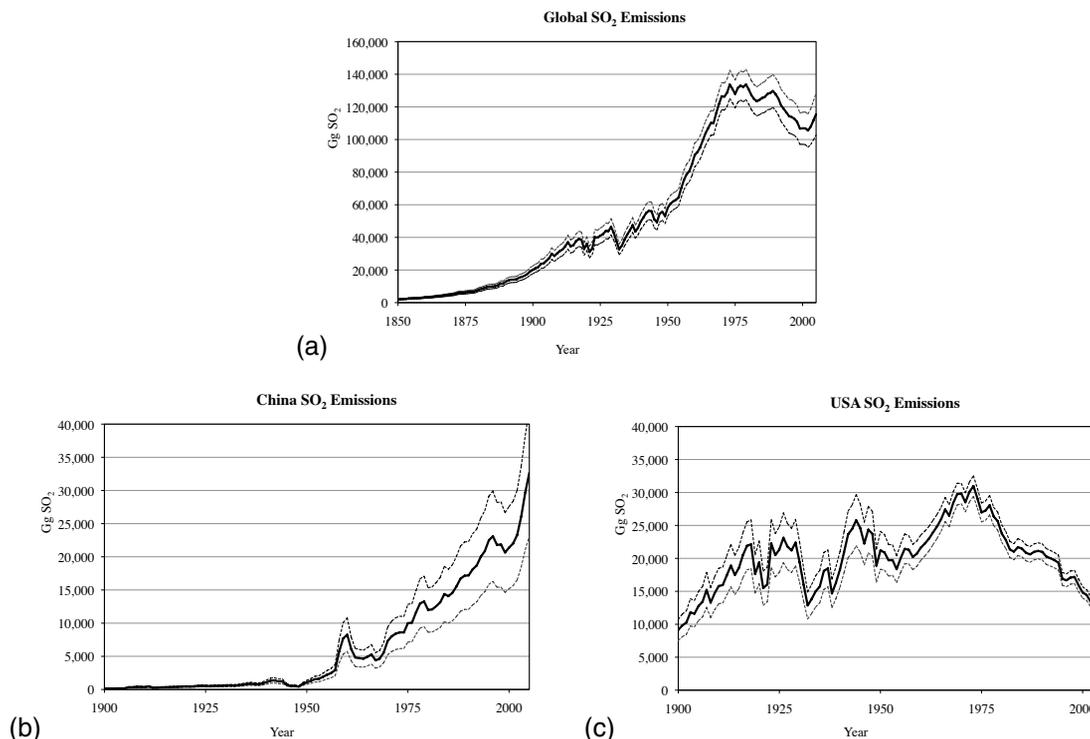
Fig. 4. (a) Global sulfur dioxide emissions from fuel combustion and process emissions with upper and lower uncertainty bounds. **(b)** Emissions from China with upper and lower uncertainty bounds. **(c)** Emissions from the USA with upper and lower uncertainty bounds. China and USA graphs exclude shipping emissions.
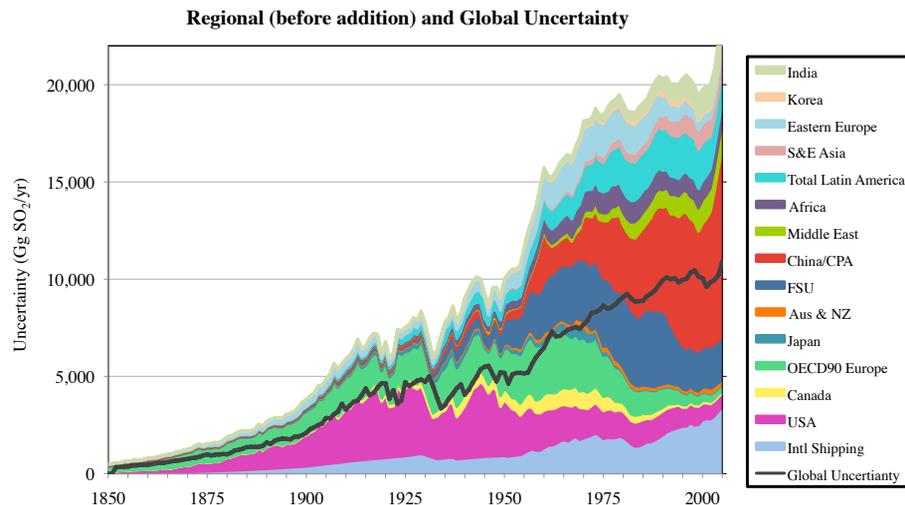
Fig. 5. Regional uncertainty in sulfur dioxide emissions including both uncorrelated and correlated components before addition (see text). The global uncertainty after addition is also shown (bold line).
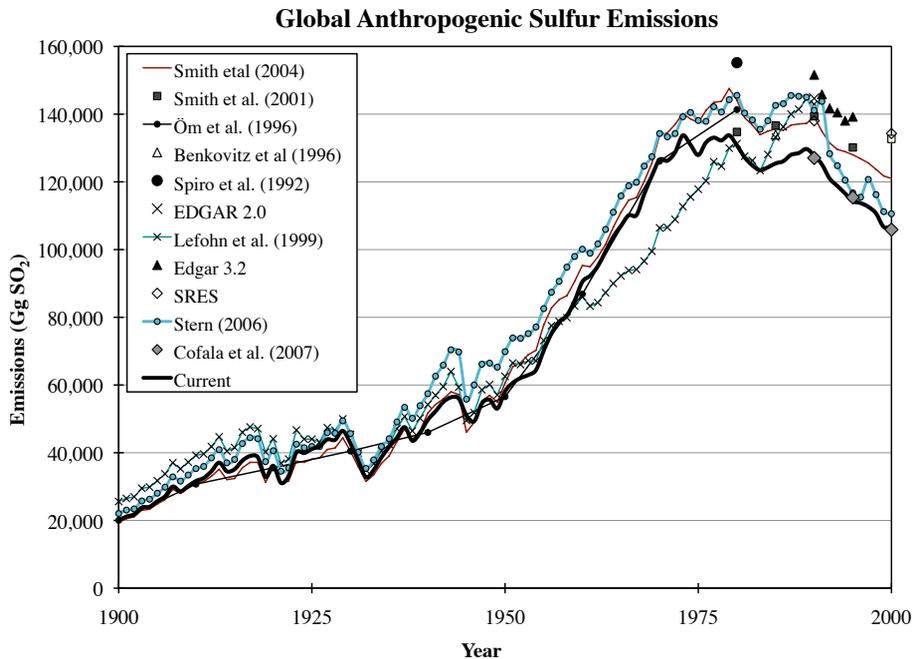
Fig. 6. Global sulfur emissions from this and previous studies: Cofala et al. (2007); GEIA (Benkovitz et al., 1996); Lefohn et al. (1999); Öm et al. (1996); Smith et al. (2001, 2004); Spiro et al. (1992); SRES (Nakicenovic and Swart, 2001); Stern (2006); EDGAR 2.0 (Olivier et al., 1996); and EDGAR 3.2 (Olivier and Berdowski, 2001). Open biomass burning emissions, where available, were not included in the totals shown. Cofala et al. (2007) estimates adjusted to include international shipping as estimated here. The 2000 points from SRES and Smith et al. (2001) are shown as open symbols as these were projected emissions estimates made before year 2000 data were available.

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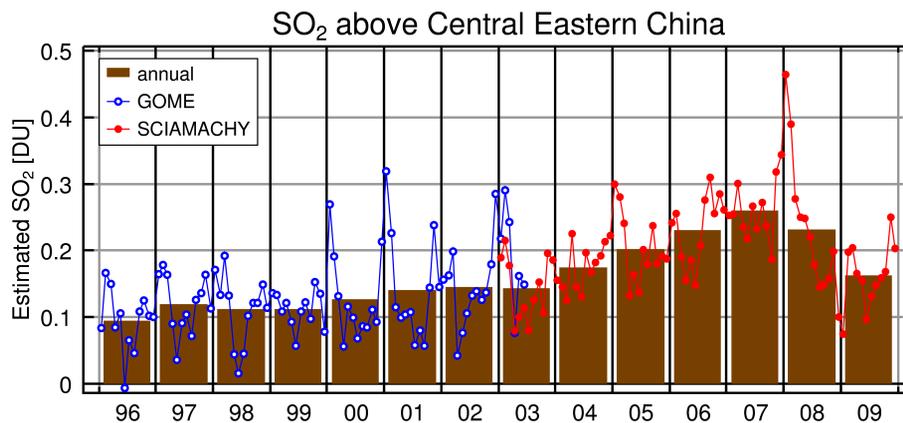
Fig. 7. Monthly (lines) and annual average (bars) estimated sulfur dioxide column over Eastern China as estimated from satellite measurements (Gottwald et al., 2010).

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