Atmos. Chem. Phys., 11, 1101–1116, 2011
www.atmos-chem-phys.net/11/1101/2011/
doi:10.5194/acp-11-1101-2011

Atmospheric Chemistry and Physics

Anthropogenic sulfur dioxide emissions: 1850–2005

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

[1]Joint Global Change Research Institute, Pacific Northwest National Laboratory, 5825 University Research Court, Suite 3500, College Park, MD 20740, USA
[2]European Commission Joint Research Centre, European Commission, Via E. Fermi 2749, TP 290, 21027 Ispra (VA), Italy
[3]International Institute for Applied Systems Analysis, Schlossplatz 1, 2361, Laxenburg, Austria
[4]Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6335, USA
[*]current address: European Environment Agency, Kongens Nytorv 6, 1050 Copenhagen K, Denmark

Received: 28 May 2010 – Published in Atmos. Chem. Phys. Discuss.: 30 June 2010
Revised: 28 January 2011 – Accepted: 1 February – Published: 9 February 2011

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. Sulfuric 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 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, causing a net cooling. 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 substantially 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. This paper presents a new estimate of global and country-level sulfur dioxide anthropogenic emissions over the 1850–2005 period. This work represents a substantial update of previous work (Smith et al., 2001; Smith et al., 2004) with newer data and improved methodologies, and was the basis for the sulfur emissions in Lamarque et al. (2010). The emissions reconstruction presented here accounts for regional differences in the pace and extent of emission control programs, has annual resolution, includes all anthropogenic sources, and provides global coverage. Fuel-based and activity-based (Eyring et al., 2010) 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.

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

Published by Copernicus Publications on behalf of the European Geosciences Union.

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 uncertain since sulfur contents vary and information on sulfur removals is not always reported. 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 with the available country inventory data, contains complete coverage of all relevant emissions sources (assuming the country inventory data are complete), and is consistent across years.

Emissions were estimated annually by country for the following sources: coal combustion, petroleum combustion, natural gas processing and combustion, petroleum processing, biomass combustion, shipping bunker fuels, metal smelting, pulp and paper processing, other industrial processes, and agricultural waste burning (AWB). The approach is summarized in Fig. 1, and further detailed in the Supplement (S.2, S.3). The first step was to 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. Initial estimates for other years were constructed by interpolating emissions factors, with final estimates by source and country found after calibrating to country-level emissions inventories where those were available (S.3). This methodology accounts for changing patterns of fuel consumption and emission controls in a context of limited detail for earlier years.

Emissions by end-use sector for decadal years were estimated for 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.

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 Supplement S.2) using data from IEA/OECD (2006), UN energy statistics (1996), and Andres et al. (1999). 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),

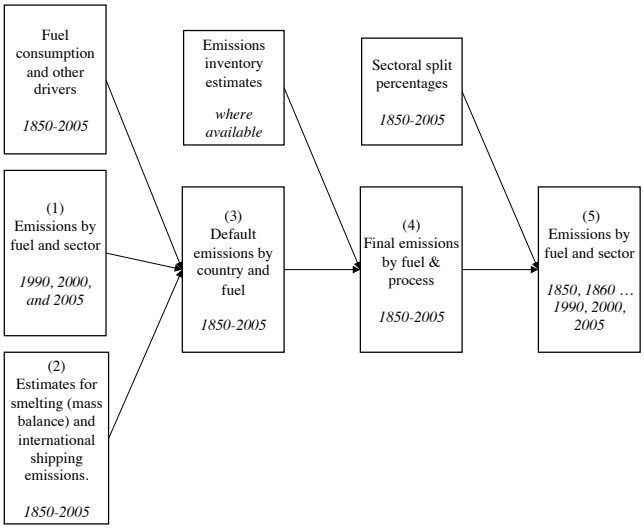

Fig. 1. Calculation summary. The key steps in the calculation are: **(1)** development of an inventory by sector and fuel for three key years, **(2)** development of detailed estimates for smelting and international shipping, **(3)** calculation of a default set of emissions by interpolating emissions factors from the key years, **(4)** calculation of final annual emissions values by fuel that match inventory values, and **(5)** estimate sectoral emissions.

Mylona (1996), Gschwandtner et al. (1986), UK National Atmospheric Emissions Inventory (2009), USEPA (1996), and Vestreng et al. (2007) as detailed in the Supplement (S.4, S.5). Fossil-fuel emissions factors were scaled such that total emissions matched inventory values for those years where inventory values are available. While total emissions for these countries are, therefore, constrained by inventory values, emissions by fuel are not as well constrained by the available data.

The ratio of default to inventory emissions in 1975 ranged from 0.3 to 2 for countries in Europe, for example, giving an indication of the differing rates at which emissions factors change in different countries. These changes can be due to different fuel sulfur contents and emission control policies. A similar range was found when comparing 1975 to 1960 values (see Supplement S.3).

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; Klimont et al., 2009) and the initial estimates here were adjusted where necessary to better match these studies (see Supplement S.4). These estimates differ from each other in some countries and sectors, highlighting a need for improved inventories in this rapidly changing region. Specific assumptions for China and the Former Soviet Union are described in the Supplement (S.6, S.7), including the operation of SO_2 scrubbers in China (Xu et al., 2009).

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 for petroleum were adjusted downward in recent years to account for increased stringency of sulfur content standards as reported in UNEP (2008).

Petroleum mass-balance

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 S.8). The mass balance estimate for global sulfur emissions from petroleum as compared to the inventory estimate is shown in Fig. 2 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. The global mass balance estimate is also subject to significant uncertainties, particularly in regard to the estimates of the amount of sulfur removed at refineries, which is not always reported separately from sulfur removed from natural gas (S.8).

As noted in Smith et al. (2004), the aggregate emissions factor from petroleum products in the US as implied by current estimates (Gschwandtner et al., 1986; USEPA, 1996) 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 Supplement S.8). The increased emissions are, in part, due to imports of residual fuel and a decrease prior to 1980 in reported

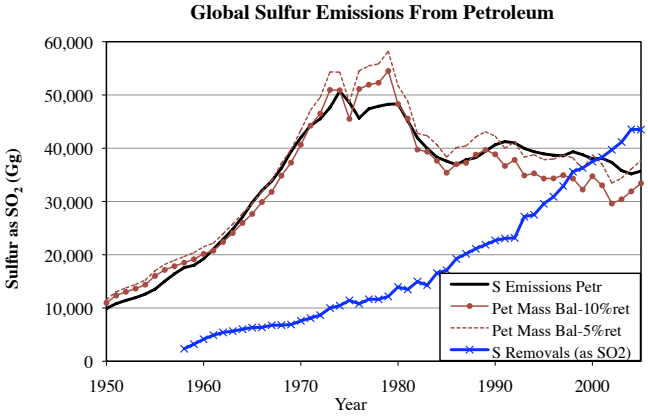

Fig. 2. Global petroleum emissions from the present inventory compared to estimates from a global crude oil mass balance (see text) using two different assumptions for the fraction of total sulfur retained in non-combusted products. The inventory data include terrestrial and international shipping combustion and process emissions from refineries. Also shown is the estimated global amount of sulfur removed at refineries (as Gg SO_2).

bunker fuel use, which is, in effect, an export of sulfur from the continent. 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. This increased US emissions from petroleum combustion by around 2000 Gg SO_2 in 1972, for example.

2.2 International Shipping

Shipping emissions have been estimated using different methodologies, some focusing on reported fuel consumption while others use bottom-up estimates using shipping statistics (Corbett et al., 1999; Corbett and Köhler, 2003, 2004; Endresen et al., 2007; Eyring et al., 2010). The estimate here uses both reported data and bottom-up estimates of shipping fuel consumption. A composite global estimate of shipping fuel consumption by fuel type (e.g., coal, residual, distillate, and other) was constructed by combining data from Eyring et al. (2010), EIA (2008), Endresen et al. (2007), Fletcher (1997), and UN (1996) with additional assumptions as described in the Supplement (S.9). We find 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. This has a substantial impact on emissions as the sulfur content of residual oil can be much higher than that of distillate fuels.

These are global values for fuel consumption, which would ideally need to be reconciled with regional energy data. As noted by Eyring et al. (2010) 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 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.

In order to estimate sulfur dioxide emissions, we calibrate to the year 2000 value from Eyring et al. (2010) of 11 080 Gg SO_2/year, which is the mean of estimates from Corbett and Köhler (2003), Eyring et al. (2005) and Endresen et al. (2007). Using the global division between fuels derived above, we match this value using the following sulfur contents: residual: 2.9% and distillate/other: 1.3%. The average value for sulfur in bunker coal is assumed to be 1.1%. For simplicity these values are kept constant over time. These values, and their time trends, are not known with precision.

The most authoritative data on marine fuel sulfur contents is from the IMO (2007), however the uncertainty in these values difficult to estimate (see Supplement S.9). While the estimate here is similar to other estimates in the literature (Corbett and Köhler, 2003; Eyring et al., 2005, 2010; Endresen et al., 2007), as presented in the Supplement, these emissions are particularly uncertain. The estimate here falls within the 2001 uncertainty of 8400–13 100 Gg as estimated by Corbett and Köhler (2003, 2004).

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 discussed 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 (see Supplement S.2, S.9). 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.

2.3 Metal amelting

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. Smelter production of copper, zinc, lead, and nickel was tabulated by country, primarily using USGS mineral yearbooks and predecessor publications for 1930–2005 and a variety of supplemental sources, particularly for earlier years. 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 Supplement (S.10).

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 readily 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 the sulfur content of ore appears to be larger than default values, which could also be the case in regions without inventory data, potentially 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. Natural gas production emissions were estimated over time for the United States, the Former Soviet Union, and other regions without inventory data (see Supplement S.11).

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 Gg 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) and inventory data combined with wood pulp production statistics (see Supplement S.11).

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.

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 Supplement S.2).

Emissions from agricultural waste burning on fields were from EDGAR v4.0 (JRC/PBL, 2009). Production statistics for 24 crop types from FAO (FAOSTAT Crop Production) were combined with information on the fraction burned on the fields (Yevich and Logan, 2003; Eggleston et al., 2006; UNFCCC, 2008) and emission factors from Andreae and Merlet (2001). Emissions from waste burning were calculated in a similar manner, although these are small and the data for this sector are incomplete. EDGARv4.0 emissions from agricultural waste burning and waste were included in the Representative Concentration Pathway (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 from 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, and additional information from van Aardenne et al. (2001) and Bond et al. (2007), see Supplement for details (S.12).

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 and other industrial, transportation combustion, and agricultural waste burning on fields within each country (Supplement S.12).

For 1850–1900, emissions from combustion and other industrial 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.

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 downscaling these emissions estimates to a spatial grid (van Vuuren et al., 2010), including incorporation of emissions measurements, could 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 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.

We consider two sources of uncertainty, random and systemic uncertainties, as summarized in Eq. (1)

Table 1. Uncertainty bounds (as 95% confidence interval) by country category and emissions type. The uncertainty bounds shown in the table are used for random effects in Eq. (1). An additional systemic uncertainty was added (see text) with a magnitude of 2.5% for countries with category I inventories, and 5% for all other countries (and all countries prior to 1970).

Category	Coal	Petroleum	Smelting	Other Process, Biomass
I. Recent-Country-Inventory	±11%	±21%	±14%	±22%
II. Older Inventory	±18%	±27%	±25%	±38%
IIa. OECD (pre inventory)	±25%	±43%	±25%	±52%
III. Other Countries	±28%	±45%	±36%	±54%
IV. Int Shipping	±28%			
IV. Int Shipping (earlier)	±42%			

$$\text{uncertainty} = \sqrt{\sum_r \sum_s \left(\text{Emissions}_s^r \cdot \text{CI\_random}_s^r\right)^2} + \sum_r \sqrt{\sum_s \left(\text{Emissions}_s^r \cdot \text{CI\_systemic}_s^r\right)^2}, \quad (1)$$

where CI is the assumed 5–95% confidence level, in percent from Table 1, for a given region and category. The sums s and r are conducted over the source categories and regions listed in the Supplement (S.1, S.15).

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 1 are applied to countries categorized depending on the estimated quality of the data used to construct the inventory values (see Supplement S.15). Uncertainties 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), such as shown in the Supplement (S.15). Only the total values, however, are used in this calculation.

The values in Table 1 are based on the authors' judgment, informed by previous work in the literature (Schöpp et al., 2005; Gregg et al., 2008; Eyring et al., 2010), comparisons with previous versions of this work, and changes over time in EPA inventories (see Supplement, S.13, S.14). These sources suggest that, overall, uncertainty is smallest where emissions are directly measured, such as in coal-fired power plants, and is relatively larger for emissions from petroleum products (except for countries with well-enforced and comprehensive sulfur standards), and process emissions.

In recent decades, sulfur emissions in most high-income countries have come under increasingly stringent control regimes. In earlier years, information on sulfur emissions was less complete, and we, therefore, assume that uncertainties are larger at these times. For similar reasons, we also assume that emissions are more uncertain in countries without comprehensive control regimes, or where such regimes have only been implemented recently. In addition, information on activity levels are also more uncertain in the past and in developing countries generally. Because common assumptions and data sources are used for large portions of the world, we assume that uncertainties with each source category are perfectly correlated within 14 world regions.

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: 6–10% over the 20th century. This low value is due to cancellation between source categories 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 removal, and other driver data, methodological assumptions, and the use of common default assumptions for sources 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 emissions estimates over multiple world regions (see Supplement S.13).

To include the impact of such systemic effects, we add to the uncertainty estimate for each source category 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 source categories. This latter assumption is made since there is little overlap in assumptions between sulfur contents, emissions controls, or driver data between the broad categories used in this calculation. The uncertainty estimates are, again, author's judgments, but are sufficient to increase the overall uncertainty estimate enough to encompass a larger fraction of the existing global estimates (S.13).

The global value of the systemic uncertainty component is less than 3% since 1960, due to statistical cancellation across sectors, increasing to 4% by 1920 as emissions from coal combustion become more dominant. 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 1.3 to 1.5, depending on the year. Even with this component, however, the global uncertainty in sulfur dioxide emissions over the 20th century is still only 8–14%. The global emissions estimate of Smith et al. (2004) now largely falls within this expanded uncertainty 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 25% of total emissions to 29% of total emissions).

The combined uncertainty bounds for global emissions are shown in Fig. 3. 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 1970s, with a significant decrease over the 1990's, and likely increasing slightly in recent years. In 2000, the estimated uncertainty in global emissions is $\pm 9\,600$ Gg SO_2. While a number of previous estimates lie within the uncertainty bounds estimated here, there are some significant differences. In particular, a number of estimates are larger than even the upper uncertainty bound in 1990 (see Supplement S.15). We return to this point in the discussion.

The estimated uncertainty bounds for the United States and China are also shown in Fig. 3. The uncertainty range for the United States is smaller, even in earlier years, than the estimate for China. In recent years, a large portion of this difference is due to the assumed lower uncertainty by source (Table 1). 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. Examples of such effects are discussed for European emissions estimates in Schöpp et al. (2005). Offsetting this somewhat is a similar relative impact of systematic uncertainties. Uncertainty in emissions from China is large enough that significant differences in trends over recent decades are possible if trends in uncertain parameters also change over time.

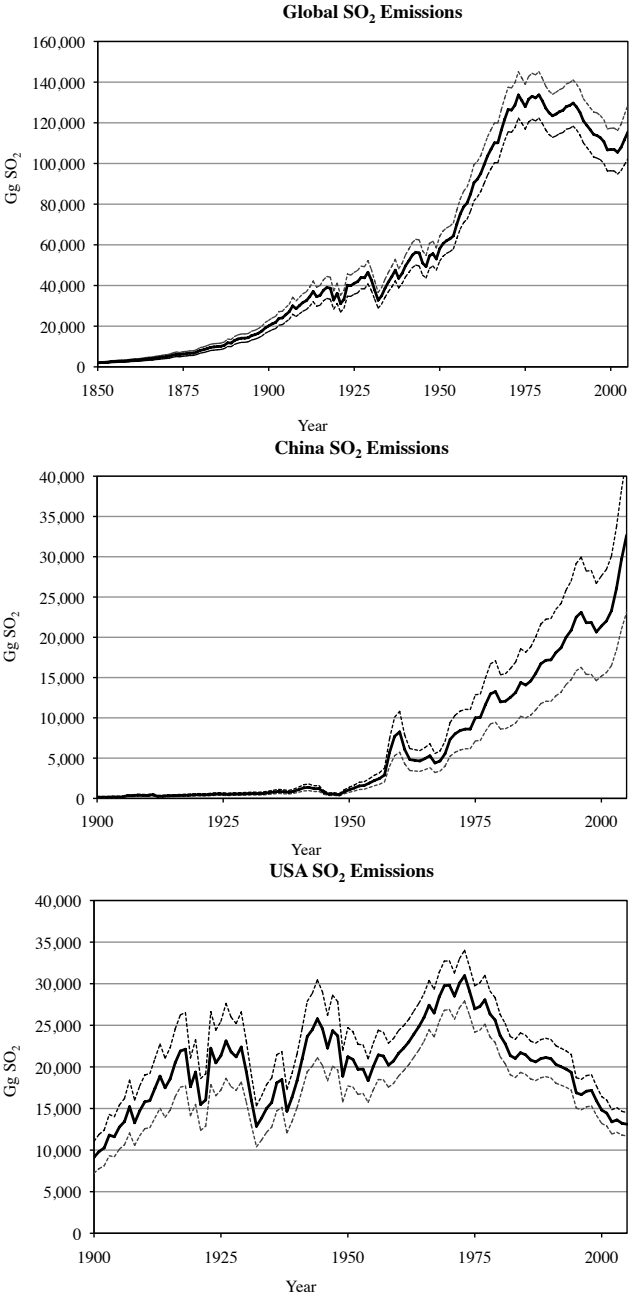

Fig. 3. Sulfur dioxide emissions from fuel combustion and process emissions with central value (solid line) and upper and lower uncertainty bounds (dotted lines). **(a)** Global,**(b)** China, and **(c)** USA. China and USA graphs exclude shipping emissions.

The regional uncertainty values, before combination to global values as described above, are shown in Fig. 4. China is the largest single contributor to emissions uncertainty

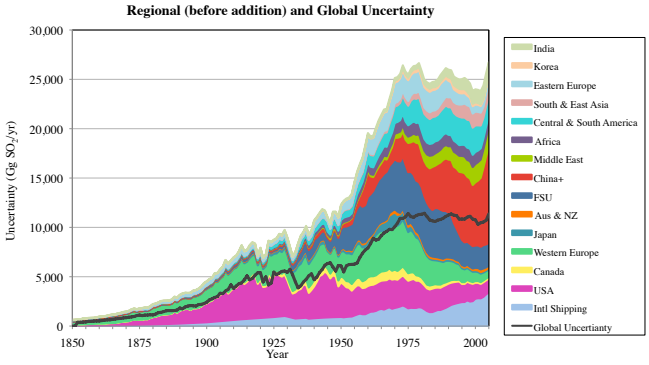

Fig. 4. Regional uncertainty in sulfur dioxide emissions including both uncorrelated and correlated components before addition (see text). The global uncertainty after addition is also shown (bold line).

since about 1980, with an estimated uncertainty in 2000 of ±6 200 Gg SO_2, a value comparable to our estimated emissions from India or the Middle East. The second largest source of uncertainty since the mid 1990s 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 TgC, with an uncertainty range of 2.0–7.4 TgC, 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.

4 Results and discussion

4.1 Emission trends

Sulfur dioxide emissions since 1850 have shown substantial sectoral and regional shifts that reflect economic and technological trends over this period. Global SO_2 emissions from 1850–2005 by source, including emissions from forest and grassland fires, and by end-use sector are shown in Fig. 5. Table 2 presents a summary of total emissions by region for decadal years, with global emissions from open burning also shown. Uncertainty bounds for the same regions are shown in Table 3.

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. Global emissions peaked in the 1970s, and have declined overall since 1990, with an increase between 2002 and 2005, largely due to strong growth of emissions in China.

The fraction of emissions from coal and petroleum have remained remarkably constant since 1980, with around 50% of emissions from coal and 30% from petroleum products. While terrestrial petroleum emissions have declined, emissions from international shipping have increased. This is a sector where high sulfur bunker fuel could, until recently, be used without significant 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.

From an end-use perspective (Fig. 5), emissions from energy conversion, largely from electricity generation, have steadily increased over the 20th century to now comprise nearly 50% of SO_2 emissions. Global emissions from domestic and industrial combustion peaked around 1970 as these sectors switched to electricity and natural gas. While energy (conversion) now comprises the largest emission sector, the remaining emissions are distributed over a range of end-uses, notably smelting, industrial combustion, ocean shipping, domestic combustion, and other process emissions. Surface transportation contributes a relatively small portion of total sulfur dioxide emissions. The largest portion of emissions from petroleum combustion arise from use of heavy fuel oils, including residual fuel, which are not often used in surface transportation other than for domestic shipping. The only end-use sectors that have increased emissions between 1970 and 2005 are the energy sector and international shipping. Total process emissions (smelting, fuel processing and extraction, pulp and paper, etc.) appear to have exceeded emissions from industrial combustion since 1990. These trends point to the central role of energy-sector emissions in policies to reduce sulfur emissions overall, but also the increasing importance that other sectors, such as international shipping, can play once energy sector emissions come under emission control regimes and begin to decrease (Miola et al., 2010; Eyring et al., 2010).

Table 2. Total emissions estimates by decade (for the indicated year) and region ($Gg\,SO_2$), with global emissions shown from sectors not included in the country-level estimates (open biomass burning from forest, grassland, and agricultural waste burning on fields, see text).

Region	1850	1860	1870	1880	1890	1900	1910	1920	1930	1940	1950	1960	1970	1980	1990	2000	2005
USA & Canada	311	707	1.380	2.701	5.443	9.345	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	1.003	2.003	2.729	2.991	2.145
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	1.101	1.346	2.740	3.648	5.328	5.314	4.719	4.432
Western Europe	1.292	1.929	2.754	3.972	5.295	7.213	9.323	9.035	11.649	12.519	13.974	20.599	29.329	26.759	18.206	7.998	6.242
Central Europe	34	75	219	518	903	1.323	1.756	1.448	2.280	3.334	3.942	7.235	11.462	13.589	12.316	5.704	4.832
Russia	40	39	54	85	125	278	389	120	517	1.897	2.565	5.744	7.926	10.143	10.632	6.352	5.975
Ukraine	7	9	15	33	55	140	192	76	260	907	1.710	3.249	4.320	5.505	4.921	1.548	1.470
Other Former Soviet Union	6	6	7	10	19	39	39	21	125	525	695	2.864	3.684	3.777	4.116	2.516	3.338
China	162	147	138	140	142	148	357	458	581	1.089	1.070	8.261	7.327	11.981	17.194	21.393	32.673
Japan	25	26	27	54	119	261	533	1.081	1.347	1.641	1.099	2.188	5.337	1.317	975	885	834
Other South & East Asia	25	27	29	32	35	49	70	107	213	325	241	780	1.796	3.654	5.489	6.330	6.111
India	35	39	43	48	64	102	161	221	281	338	425	702	1.114	1.674	3.302	5.363	6.275
Australia & New Zealand	9	24	32	53	61	149	264	213	339	466	486	927	1.553	1.692	1.648	2.438	2.606
South Africa	1	2	3	11	20	29	170	271	252	399	588	833	1.491	1.860	2.283	2.392	2.477
Africa	11	12	14	16	18	20	26	50	220	823	1.087	2.070	3.168	3.784	3.350	3.322	2.690
Middle East	3	3	4	4	5	5	5	11	20	136	324	572	1.179	2.490	3.436	5.218	5.490
International Shipping	61	120	208	365	587	892	1.450	1.927	2.274	2.233	3.247	4.680	6.469	6.607	7.041	9.779	12.078
Global Total (Comb + Process)	2.063	3.224	4.996	8.120	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
Additional Emissions (Global)																	
Forest & Grassland Burning	2.447	2.447	2.447	2.447	2.447	2.447	2.439	2.244	2.035	1.958	1.859	1.920	2.198	2.649	3.357	3.836	3.836
Agricultural Waste Burning	83	88	94	100	106	110	115	120	126	134	141	153	155	180	211	205	205

Table 3. Estimated emissions uncertainty, as 5–95% confidence interval, for the indicated year and region ($Gg\,SO_2$).

Region	1850	1860	1870	1880	1890	1900	1910	1920	1930	1940	1950	1960	1970	1980	1990	2000	2005
USA & Canada	82	191	384	748	1.310	1.922	3.250	3.931	3.734	3.404	3.658	2.998	3.212	2.664	2.357	1.650	1.440
Mexico	1	1	1	1	6	17	71	77	102	124	210	257	370	857	1.219	1.301	822
Central America	3	0	0	0	0	0	3	6	55	38	59	123	184	187	184	172	127
South America	11	20	24	29	24	20	53	96	199	310	329	587	774	1.181	1.166	836	764
Western Europe	272	385	536	661	761	981	1.275	1.359	1.514	1.699	1.577	2.172	2.771	2.097	1.197	654	521
Central Europe	7	15	42	110	189	267	341	277	348	512	571	995	1.418	1.600	1.532	559	604
Russia	14	14	17	24	34	82	116	39	165	703	1.044	1.917	2.337	2.633	2.443	1.382	1.280
Ukraine	3	3	5	13	20	56	84	29	115	449	884	1.501	1.705	1.581	1.106	427	394
Other Former Soviet Union	2	2	2	2	3	7	7	4	26	137	192	701	843	770	825	590	738
China	95	87	81	82	83	86	113	139	170	318	310	2.542	2.094	3.367	5.139	6.229	9.579
Japan	11	12	12	13	20	45	94	205	249	299	185	356	1.178	229	186	160	142
Other South & East Asia	6	6	7	7	8	9	11	15	33	45	34	94	265	522	689	831	713
India	21	21	22	23	25	31	43	58	75	94	108	173	263	399	791	1.334	1.496
Australia & New Zealand	3	7	9	12	13	32	57	41	70	100	92	183	315	245	144	226	238
South Africa	0	1	1	4	7	7	47	73	76	111	167	230	378	485	587	669	779
Africa	3	3	3	4	4	5	5	9	52	254	273	562	739	758	582	555	423
Middle East	1	1	1	2	2	2	2	3	5	47	82	123	245	525	701	954	1.010
International Shipping	26	51	88	155	249	379	615	817	965	789	918	1.324	1.830	1.869	1.991	2.766	3.416
Global Total	334	493	755	1.166	1.781	2.551	4.079	4.808	4.893	5.009	5.515	7.519	8.906	9.284	9.975	9.857	12.827

Regional emissions trends are shown in Fig 6. As seen in previous work (Smith et al., 2001, 2004; Stern, 2006), 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, with approximately 40% of global emissions originating from Asia by 2005. Emissions from other regions (Africa, Middle-East, Central and South America) have also increased, but to a lesser extent. Emissions from China have increased to 28% of the estimated global total in 2005.

For most of the historical period, sulfur dioxide emissions increased in rough proportion to activity levels. This began to change 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. 2) 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 globally. In the United States, where detailed information on coal sulfur content and emissions are available, the shift

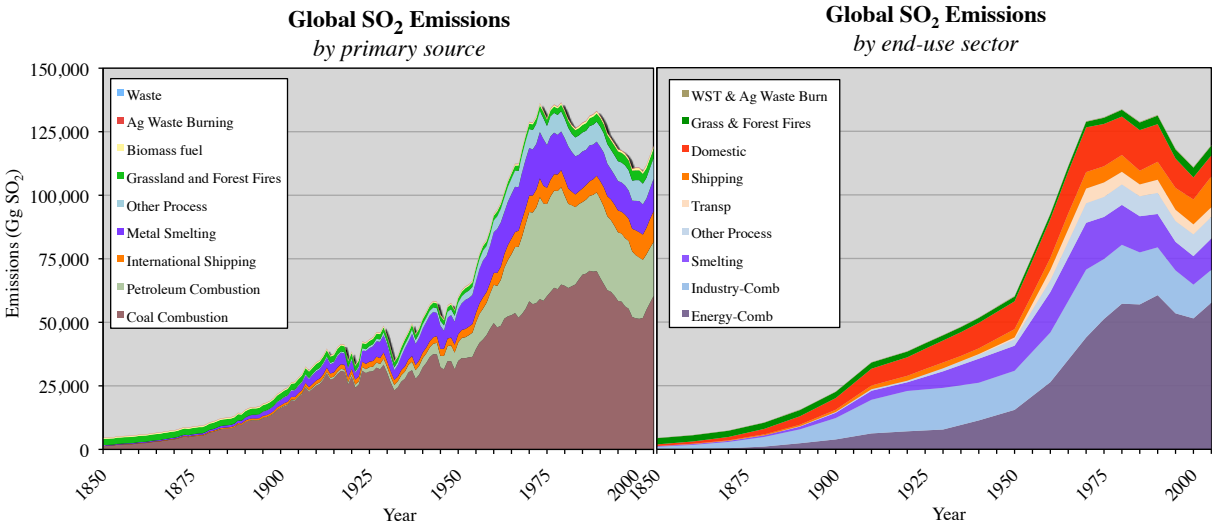

Fig. 5. Global sulfur dioxide emissions by **(a)** source and **(b)** end-use sector. Emissions by source are the primary inventory result from this work. Emissions were then mapped to sector. Included are emissions from forest and grassland fires from van der Werf et al. (2006), Schultz et al. (2008), and Mieville et al. (2010) as used in the RCP historical inventory exercise (Lamarque et al., 2010).

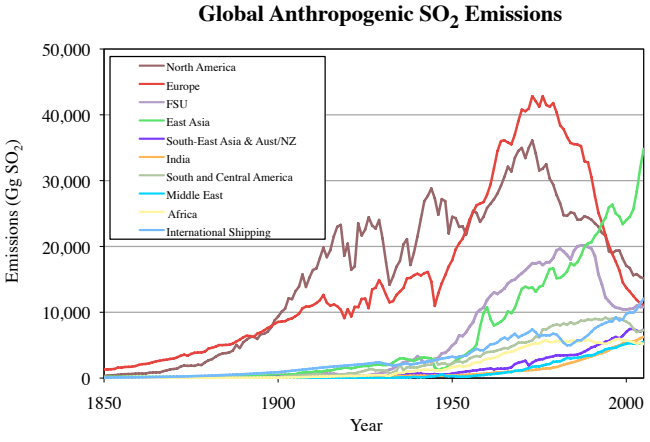

Fig. 6. Global sulfur dioxide emissions by region (North America = USA + Canada; East Asia = Japan, China+, and South Korea).

to low-sulfur coal was a major driver of emissions reductions until recent years. Flue-gas desulfurization has played an increasing role in recent years, when the coal sulfur content actually increased, but emissions continued to decrease (see Supplement, Fig. S-2).

The impact of these changes can be seen in Fig. 7, which shows the aggregate emissions factor (emissions over fuel combusted) by region. Note that the split between coal and petroleum emissions is approximate in some regions, which means that there is uncertainty in these emissions factors. The aggregate emissions factor shown in Fig. 7 includes the impact of changes in sulfur content, sectoral shifts, and emissions controls.

The change in coal emissions factor varies greatly by region, with some regions showing little change, while emissions factors in many regions, including the Japan, Europe, United States, Canada, and South Korea, have decreased substantially since 1970. As a result, the global average emissions factor for combusted coal has decreased by 2005 to 60% of the 1970 value. Shifts to lower sulfur coal and flue-gas desulfurization have contributed to lower relative emissions from coal combustion, although emissions data alone is not sufficient to quantify these effects individually.

The emissions factor for petroleum has decreased in all world regions. The global average emissions factor for petroleum products in 2005 is about half of the 1970 value. From a top-down perspective, this is due to an increase in the fraction of sulfur removed from crude oil at oil refineries. From a bottom-up perspective, the decrease is due to limits on the sulfur content of end-use fuels and a reduction in the fraction of residual oil consumed. Countries such as Mexico and South Korea had particularly high percentages of residual oil in their consumption mix until around 1990, and a decrease in this fraction since then has contributed to the decline in the aggregate petroleum emissions factor.

4.2 Comparison with other estimates

The methodology used here, whereby regional inventories are used to calibrate a bottom-up emissions estimate, was used in order to produce an estimate that uses what was judged to be the best data from various regions. Use of such inventory information automatically takes into account emissions control efforts, providing the inventory data used accurately takes these factors into account. Uncertainty

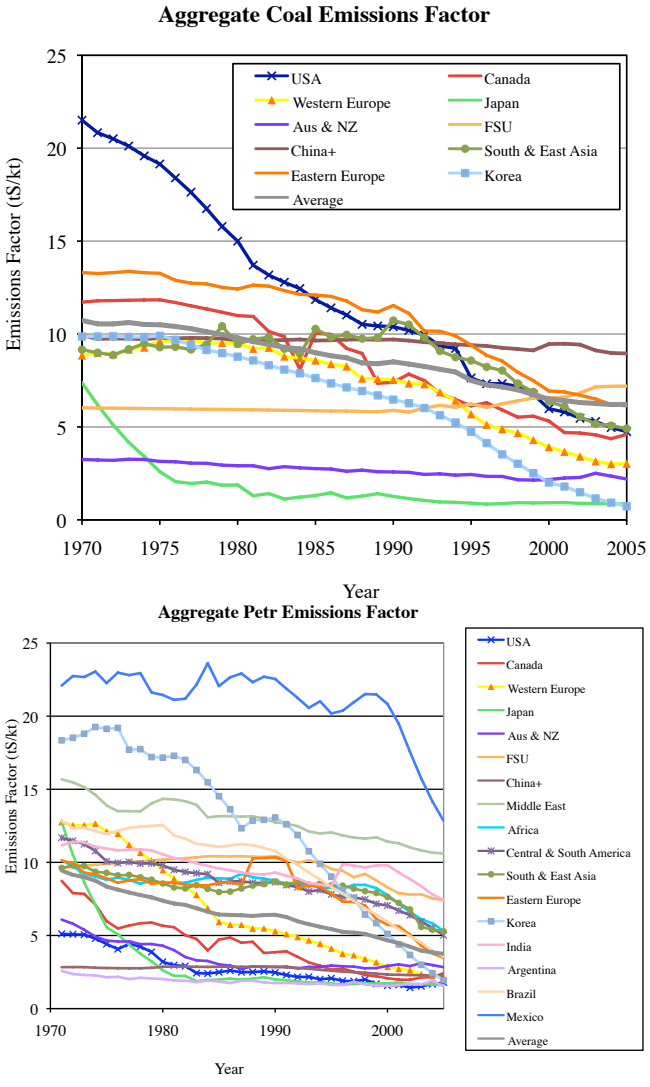

Fig. 7. Aggregate emissions factors for **(a)** coal and **(b)** petroleum combustion by region.

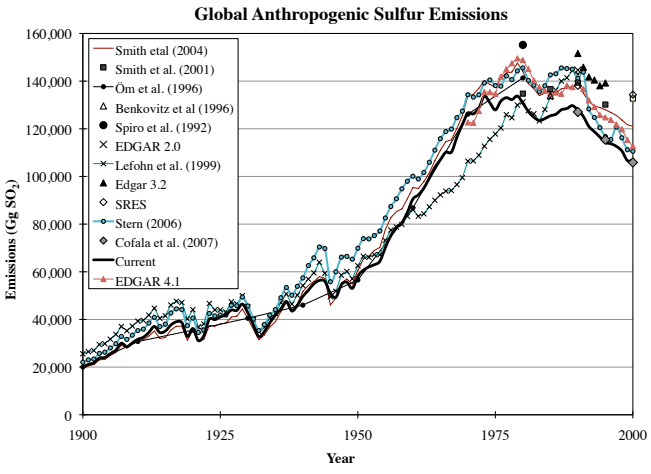

Fig. 8. Global sulfur emissions from this and previous studies: Cofala et al., 2007; GEIA (Benkovitz et al., 1996); Lefohn et al., 1999; Öm et al., 1996; Smith et al., 2001, 2004; Spiro et al., 1992; SRES (Nakicenovic and Swart, 2001); Stern, 2006, EDGAR 2.0 (Olivier et al., 1996), EDGAR 3.2 (Olivier and Berdowski, 2001), and EDGAR 4.1 (JRC/PBL 2010). 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.

assumptions were coupled with the use of inventory data, with emissions estimates assumed to be more uncertain in years before inventory data were available.

The annual global emissions estimate in this work is similar before 1970 to the previous estimate using similar methodologies (Smith et al., 2004), with an average absolute difference of 4% from 1900–1970. After 1970, the current global emissions estimate is consistently lower than Smith et al. (2004), and the difference increases to 13% over 1995–2000. The current estimate is lower for the Former Soviet Union, China, and South and East Asia, but larger for international shipping. The largest difference is lower emissions from coal, with lower emissions also from petroleum (aside from shipping). The reasons for the lower emissions vary by region, and include different treatment of coking coal, assumptions for coal emissions in China, different calibration data for the Former Soviet Union, and inclusion of additional emissions control measures in Asia, and lower sulfur standards in most regions. Other differences include updated information on sulfur removals from refineries and smelters, and improved methodologies. See the Supplement (S.13) for further discussion and analysis.

The current estimate is compared to other estimates of global sulfur dioxide emissions in Fig. 8. The current estimate is somewhat below many recent estimates, particularly in the 1970s and 1980s. 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 country inventory data for many OECD countries. The uncertainty bounds estimated in section 4 encompass many of the other estimates (see also S.15), 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 recent estimates from the EDGAR 4.1 (JRC/PBL, 2010) inventory are, on a global basis, somewhat higher than the estimates here (see Supplement S.16). While the global emissions value is similar in 1970 and 2005, there are large regional differences in all years. The EDGAR inventory uses a consistent approach for all regions and gases, but does not calibrate to country-level inventories. Some differences are, therefore, expected since not all region- and country-specific

data can be included. There are particularly large differences in the two estimates in the Former Soviet Union, an area where comprehensive emissions information is still relatively sparse. Large relative differences are also seen for Japan, the FSU, South & East Asia, South Korea, India, Argentina, and international shipping. The reasons for these differences should be examined once methodological details from the EDGAR estimate are available. The global totals from the EDGAR-HTAP inventory for 2000–2005, which substitute regional inventory values for the EDGAR 4.1 values where available, are within 1–2% of the values reported here.

It is also useful to compare 2005 emissions with previous estimates from global scenario projections. The 2005 emissions, estimated here as $115\,510 \pm 12\,830$ 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 Supplement S.17).

Emissions in Asia are of particular interest due to their recent increase, and it is useful to compare emissions here with previous estimates. As shown in the Supplement, even recent estimates for SO_2 emissions in Asia (Ohara et al., 2007; Streets et al., 2003; Zhang et al., 2009, and Klimont et al., 2009) sometimes differ substantially (S.4). 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 (Ohara et al., 2007) estimate for 2000 consistently larger than the other estimates quoted here, but within the estimated uncertainty range. 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 (see Supplement S.4).

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 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 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.

4.3 Uncertainty

The uncertainty methodology used here uses a relatively simple procedure whereby confidence intervals, based largely on the authors' judgment, but also informed by analysis of inventory differences, are applied to broad emissions sectors and regions. The relatively small resulting 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 analysis of high-emitting regions, and the countries of the Former Soviet Union in particular, may be useful to better bound current and past environmental impacts of sulfur dioxide emissions.

Because the simple methodology used here does not incorporate correlations between uncertainty assumptions in different regions and sectors (parameters that can be difficult to estimate in any event), a systemic uncertainty component was added. Systematic errors and biases are difficult to quantify, but are particularly important for emissions such as sulfur dioxide, where most input values are only weakly correlated between regions, which results in relatively small global uncertainties as random errors tend to statistically cancel across regions.

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 shown by an analysis of inventory values from the USA (§S.14), however, significant changes can occur over time in national inventory values. Substantial changes have also occurred for inventory estimates for Europe (e.g., see Konovalov et al., 2008 for NO_x emissions). Analysis of the sources of such changes would be valuable for both improving both inventory methodologies and uncertainty estimates.

5 Future work

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 subnational 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.

Improved and updated emissions estimates for countries without national inventories will depend, in part, on accurate representations of changing sulfur content standards. The sectors and specific fuel types subject to standards, and the level of enforcement, will all impact the resulting emissions. The increase in international shipping emissions should slow and ultimately decline once recent agreements on sulfur standards for bunker fuels are implemented.[1] Regionally, any expansion of Sulfur Emission Control Areas, where lower sulfur fuel limits will apply, will further reduce shipping emissions.

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 9 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 satellite estimate indicates a slight increase in concentrations from 1996/1997 to 2000/2001 while the inventory estimate is nearly flat, taking the average over the indicated years. There is, of course,

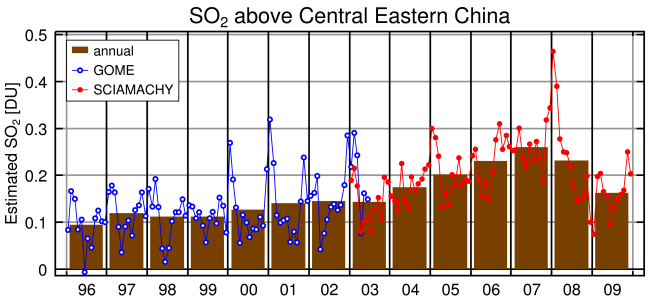

Fig. 9. Monthly (lines) and annual average (bars) estimated sulfur dioxide column over Eastern China as estimated from satellite measurements (Gottwaldov et al., 2010).

substantial uncertainty in the emissions estimate, and a different trend over this period would be within the uncertainty range. In addition, a 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 the 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 in satellite estimates 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.[2]

Supplementary material related to this article is available online at: http://www.atmos-chem-phys.net/11/1101/2011/acp-11-1101-2011-supplement.pdf.

Acknowledgements. Work at PNNL on this project was supported by the Office of Science (BER), US Department of Energy. Z. Klimont and GAINS work was supported by ACCENT and EUCAARI. RJA was sponsored by US Department of Energy, Office of Science, Biological and Environmental Research (BER)

[1] Agreements conducted under the International Convention for the Prevention of Pollution From Ships, commonly refered to as MARPOL. Marine Environment Protection Committee (MEPC) – 57th session: 31 March–4 April 2008.

[2] http://www.iiasa.ac.at/web-apps/tnt/RcpDb/

programs and performed at Oak Ridge National Laboratory (ORNL) under US Department of Energy contract DE-AC05-00OR22725. The authors would like to thank Elaine Chapman for a very useful internal review, Andreas Richter for Figure 9 and helpful comments, and the many colleagues and organizations that shared data used in this project. We also thank Mayda Nathan, Terri Weber, and Andrew Mizrahi for assistance with proofreading and formatting. The authors would like to thank the two anonymous referees for their detailed comments, which were of great value in improving the paper.

Edited by: R. Harley

References

Andreae, M. O. and Merlet, P.: Emissions of trace gases and aerosols from biomass burning: Global Biogeochem. Cy., 15, 955–966, doi:10.1029/2000GB001382, 2001.

Andres, R. J., Fielding, D. J., Marland, G., Boden, T. A., and Kumar, N.: Carbon dioxide emissions from fossil-fuel use, 1751–1950, Tellus, 51, 759–765, 1999.

Arndt, R. L., Carmichael, G. R., Streets, D. G., and Bhatti, N.: Sulfur dioxide emissions and sectorial contributions to sulfur deposition in Asia, Atmos Environ., 31, 1553–2310, doi:10.1016/S1352-2310(96)00236-1, 1997.

Benkovitz, C. M., Scholtz, M. T., Pacyna, J., Tarrason, L., Dignon, E. C., Voldner, P. A., Spiro, Logan, J. A., and Graedel, T. A.: Global gridded inventories of anthropogenic emissions of sulphur and nitrogen, J. Geophys. Res., 101(D22), 29239–29253, doi:10.1029/96JD00126, 1996.

Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J.-H., and Klimont, Z.: A technology-based global inventory of black and organic carbon emissions from combustion, J. Geophys. Res., 109, D14203, doi:10.1029/2003JD003697, 2004.

Bond, T. C., Bhardwaj, E., Dong, R., Jogani, R., Jung, S., Roden, C., Streets, D. G., and Trautmann, N. M.: Historical emissions of black and organic carbon aerosol from energy-related combustion, Global Biogeochem. Cy., 21, GB2018, doi:10.1029/2006GB002840, 2007.

Center for International Earth Science Information Network (CIESIN), Columbia University; and Centro Internacional de Agricultura Tropical (CIAT), Gridded Population of the World Version 3 (GPWv3): Population Density Grids. Palisades, NY: Socioeconomic Data and Applications Center (SEDAC), Columbia University, available at: http://sedac.ciesin.columbia.edu/gpw, 2005.

Cofala, J., Amann, M., Klimont, Z., Kupiainen, K., and Höglund-Isaksson, L.,: Scenarios of Global Anthropogenic Emissions of Air Pollutants and Methane Until 2030, Atmos. Environ., 38, 4769–4778, doi:10.1016/j.atmosenv.2007.07.010, 2007.

Corbett, J. J. and Köhler, H. W.: Updated emissions from ocean shipping, J. Geophys. Res., 108(D20), 4650, doi:10.1029/2003JD003751, 2003.

Corbett, J. J. and Köhler, H. W.: Considering alternative input parameters in an activity-based ship fuel consumption and emissions model: Reply to comment by Øyvind Endresen et al., on "Updated emissions from ocean shipping", J. Geophys. Res., 109, D23303, doi:10.1029/2004JD005030, 2004, 2004.

Corbett, J. J., Fischbeck, P. S., and Pandis, S. N.: Global nitrogen and sulfur inventories for oceangoing ships, J. Geophys. Res., 104(D3), 3457–3470, doi:10.1029/1998JD100040, 1999.

Eggleston, S., Buendia, L., Miwa, K., Ngara, T., and Tanabe, K. (Eds): 2006 IPCC Guidelines for National Greenhouse Gas Inventories, Intergovernmental Panel on Climate Change, IGES, Japan, 2006.

Endresen, Ø., Sørgard, E., Behrens, H.L., Brett, P.O., and Isaksen, I.S.A.: A historical reconstruction of ships' fuel consumption and emissions, J. Geophys. Res., 112, D12301, doi:10.1029/2006JD007630, 2007.

Energy Information Administration (EIA, 2008): International Energy Annual 2006, Washington, DC, available at: www.eia.doe.gov), 2004.

Energy Information Administration (EIA, 2009): Annual Energy Review 2008, Washington, DC, DOE/EIA-0384, 2009.

Environment Canada 1985–2006: Historical SO_x emissions for Canada (version 2, 8 April 2008), available at: http://www.ec.gc.ca/, 2008.

European Environment Agency (EEA, 2002): Anthropogenic emissions of sulphur (1980–2010) in the ECE region (September 2000).

Eyring, V., Köhler, H. W., van Aardenne, J., and Lauer, A.: Emissions from international shipping: 1. The last 50 years, J. Geophys. Res., 110, D17305, doi:10.1029/2004JD005619, 2005.

Eyring, V., Isaksen, I. S. A., Berntsen, T., Collins, W. J., Corbett, J. J., Endresen, O., Grainger, R. G., Moldanova, J., Schlager, H., and Stevenson, D. S.: Transport impacts on atmosphere and climate: Shipping, Atmos. Environ., 44, 4735–4771, 2010.

Fernandes, S. D., Trautmann, N. M., Streets, D. G., Roden, C. A., and Bond, T. C.: Global biofuel use, 1850–2000, Global Biogeochem. Cy., 21, GB2019, doi:10.1029/2006GB002836, 2007.

Fletcher, M. E.: "From coal to oil in British shipping" in: Williams, David, M., The World of Shipping, by Aldershot, Hants, England, Ashgate, Brookfield VT, 1997.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., and van Dorland, R.: Changes in Atmospheric Constituents and in Radiative Forcing, In: Climate Change 2007: The Physical Science Basis, Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B. M., Tignor, M., and Miller, H. L., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 129–234, 2007.

Fujita, S.: Sanseiu kenkyu 100 nen no rekishi to sono hensen, J. Res. Environ., 29, 82–88, 1993.

Garg, A., Shukla, P. R., and Kapshe, M.: The sectoral trends of multigas emissions inventory of India, Atmos. Environ., 40, 4608–4620, 2006.

Goldewijk, K. K: Three Centuries of Global Population Growth: A Spatial Referenced Population (Density) Database for 1700–2000 Population and Environment, 26, doi:10.1007/s11111-005-3346-7, 2005.

Gottwald, M. and Bovensmann, H. (Eds): SCIAMACHY, Exploring the Changing Earth's Atmosphere; 1st edition, Springer, ISBN 978-90-481-9895-5., 2011.

Gregg, J. S., Andres, R. J., and Marland, G.: China: the emis-

sions pattern of the world leader in CO_2 emissions from fossil fuel consumption and cement production Geophys. Res. Lett., 35, L08806, doi:10.1029/2007GL032887, 2008.

Gschwandtner, G., Gschwandtner, K., Eldridge, K., Mann, C., and Mobley, D.: Historic emissions of sulfur and nitrogen oxides in the United States from 1900 to 1980, Journal of Air Pollution Control Association, 36, 139–149, 1986.

International Energy Agency/Organization for Economic Cooperation and Development (IEA/OECD) Energy Statistics and Balances, International Energy Agency, Paris, 2006.

International Maritime Organization (IMO): Prevention of Air Pollution From Ships. Sulphur monitoring for 2007 (Marine Environment Protection Committee, 57th session, MEPC 57/4/24), 2007.

European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL) Emission Database for Global Atmospheric Research (EDGAR) release version 4.1., available at: http://edgar.jrc.ec.europa.eu, 2010.

Klimont, Z., Cofala, J., Schöpp W., Amann, M., Streets, D. G., Ichikawa, Y., and Fujita, S.: Projections of SO_2, NO_x, NH3 and VOC Emissions in East Asia up to 2030, J. Water Air Soil Poll., 130, 193–198, 2001.

Klimont, Z., Cofala, J., Xing, J., Wei, W., Zhang, C., Wang, S., Kejun, J., Bhandari, P., Mathur, R., Purohit, P., Rafaj, P., Chambers, A., Amann, M., and Hao, J.: Projections of SO_2, NO_x and carbonaceous aerosols emissions in Asia, Tellus B, 61, 602–617, 2009.

Konovalov, I. B., Beekmann, M., Burrows, J. P., and Richter, A.: Satellite measurement based estimates of decadal changes in European nitrogen oxides emissions, Atmos. Chem. Phys., 8, 2623–2641, doi:10.5194/acp-8-2623-2008, 2008.

Krotkov, N. A., McClure, B., Dickerson, R. R., Carn, S. A., Li, C., Bhartia, P. K., Yang, K., Krueger, A. J., Li, Z., Levelt, P. F., Chen, H., Wang, P., and Lu, D.: Validation of SO_2 retrievals from the Ozone Monitoring Instrument over NE China, J. Geophys. Res., 113, D16S40, doi:10.1029/2007JD008818, 2008.

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmos. Chem. Phys., 10, 7017–7039, doi:10.5194/acp-10-7017-2010, 2010.

Lefohn, A. S., Husar, J. D., and Husar, R. B.: Estimating historical anthropogenic global sulfur emission patterns for the period 1850–1990, Atmos. Environ., 33, 3435–3444, 1999.

Li, C., Zhang, Q. N., Krotkov, D. G., Streets, K., He, S.-C., Tsay, and Gleason, J. F.: Recent large reduction in sulfur dioxide emissions from Chinese power plants observed by the Ozone Monitoring Instrument GRL 37, L08807, doi:10.1029/2010GL042594, 2010.

Logan, J: Diverging energy and economic growth in China: where has all the coal gone?, Pacific and Asian Journal of Energy, 11, 1–13, 2001.

Lu, Z., Streets, D. G., Zhang, Q., Wang, S., Carmichael, G. R., Cheng, Y. F., Wei, C., Chin, M., Diehl, T., and Tan, Q.: Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000, Atmos. Chem. Phys., 10, 6311–6331, doi:10.5194/acp-10-6311-2010, 2010.

Mieville, A., Granier, C., Liousse, C., B. Guillaume, Mouillot, F., Lamarque, J.-F.,. Gregoire, J.-M, and Petron, G.: Emissions of gases and particles from biomass burning using satellite data and an historical reconstruction, Atmos Environ, 10.1016/j.atmosenv.2010.01.011, 2010.

Miola, A. Ciuffo, B., Giovine, E., and Marra, M.: The State of the Art on Methodologies, Technologies and Policy Options, Publications Office of the European Union, Luxembourg, 2010.

Moss, R. H., Edmonds, J. A., Hibbard, K., Carter, T., Emori, S., Kainuma, M., Kram, T., Manning, M., Meehl, J., Mitchell, J., Nakicenovic, N., Riahi, K., Rose, S., Smith, S. J., Stouffer, R., Thomson, A. M., van Vuuren, D., Weyant, J., and Wilbanks, T.: Representative Concentration Pathways: A New Approach to Scenario Development for the IPCC Fifth Assessment Report, Nature, 463, 747–756, 2010.

Mylona, S.: Sulphur dioxide emissions in Europe 1880–1991 and their effect on sulphur concentrations and depositions, Tellus B, 48, 662–689, 1996.

Nakicenovic, N. and Swart, R. (Eds.): Emissions Scenarios 2000: Special Report of the Intergovernmental Panel on Climate Change, Cambridge, UK: Cambridge University Press, 2001.

National Institute of Environmental Research-Korea (NIER), available at: http://airemiss.nier.go.kr/ Downloaded 09-11-2008, 2008.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980-2020, Atmos. Chem. Phys., 7, 4419–4444, doi:10.5194/acp-7-4419-2007, 2007.

Olivier, J. G. J. and Berdowski, J. J. M.: Global emissions sources and sinks, in: The Climate System, edited by: Berdowski, J., Guicherit, R., and Heij, B. J., 33–78, A. A. Balkema Publishers/Swets & Zeitlinger Publishers, Lisse, The Netherlands, ISBN: 90 5809 255 0, 2001.

Olivier, J. G. J., Bouwman, A. F., van der Maas, C. W. M., Berdowski, J. J. M., Veldt, C., Bloos, J. P. J., Visschedijk, A. J. H., Zandveld, P. Y. J., and Haverlag, J. L.: Description of EDGAR Version 2.0: A set of global emission inventories of greenhouse gases and ozone-depleting substances for all anthropogenic and most natural sources on a per country basis and on 1×1 degree grid. National Institute of Public Health and the Environment (RIVM) report no. 771060 002/TNO-MEP report no. R96/119, 1996.

Olivier, J. G. J., van Aardenne, J. A., Dentener, F., Pagliari, V., Ganzeveld, L. N., and J. A. H. W., Peters: Recent trends in global greenhouse gas emissions: regional trends 1970–2000 and spatial distribution of key sources in 2000, Environ. Sci., 2(2–3), 81–99, doi:10.1080/15693430500400345, 2005.

Öm, G., Hansson U., and Rodhe, H.: Historical worldwide emissions of anthropogenic sulfur: 1860–1985, Department of Meteorology, Stockholm University, Report CM-91, 1996.

Schöpp, W., Klimont, Z., Suutari, R., and Cofala, J.: Uncertainty analysis of emission estimates in the RAINS integrated assessment model, Environmental Science and Policy, 8, 601–613, 2005.

Schultz, M. G., Heil, A., Hoelzemann, J. J., Spessa, A., Thonicke, K., Goldammer, J., Held, A .C., Pereira, J. M.,

and van het Bolscher, M.: Global Wildland Fire Emissions from 1960 to 2000, Global Biogeochem. Cy., 22, GB2002, doi:10.1029/2007GB003031, 2008.

Smith, S. J., Pitcher, H., and Wigley, T. M. L: Global and Regional Anthropogenic Sulfur Dioxide Emissions, Global Planet. Change, 29(1–2), 99–119, 2001.

Smith, S. J., Andres, R., Conception, E., and Lurz, J.: Sulfur Dioxide Emissions: 1850–2000 (JGCRI Report. PNNL-14537), 2004.

Smith, S. J., Pitcher, H., and Wigley, T.M.L.: Future Sulfur Dioxide Emissions, Clim. Change, 73(3), 267–318, 2005.

Spiro, P. A., Jacob, D. J., and Logan, J. A.: Global inventory of sulfur emissions with a $1^\circ \times 1^\circ$ resolution, J. Geophys. Res, 97, 6023–6036, 1992.

Stern, D.I .: Reversal of the trend in global anthropogenic sulfur emissions, Global Environ. Change, 16, 207–220, 2006.

Streets, D. G. and Waldhoff, S. T.: Present and future emissions of air pollutants in China: SO_2, NO_x, and CO Atmos. Environ., 34, 363–374, 2000.

Streets, D. G., Tsai, N. Y., Akimoto, H., and Oka, K.: Sulfur dioxide emissions in Asia in the period 1985–1997, Atmos. Environ., 34, 4413–4424, 2000.

Streets, D., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J.-H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res., 108, 8809, doi:10.1029/2002JD003093, 2003.

US Environmental Protection Agency (USEPA, 1996), National Air Pollutant Emission Trends, 1900–1995, EPA-454/R-96-007, Washington, DC, 1996.

UK National Atmospheric Emissions Inventory, available at: http://www.naei.org.uk/, 2009.

UNFCCC: National Inventory Submissions of Annex I Parties to the UNFCCC, available at: http://unfccc.int/, 2008.

United Nations: The United Nations Energy Statistics Database (UN, 1996) (United Nations Statistics Division, New York, June, 1996.

United Nations Environmental Program (UNEP): Sulphur Levels in Diesel Fuel Matrix tables, various regions, Partnership for Clean Fuel and Vehicles, downloaded 10/9/2008, 2008.

United Nations Framework Convention on Climate Change, UNFCCC, 2008 Country submissions, Downloaded 1/2/2009, available at: http://unfccc.int/, 2009.

van Aardenne, J. A., Dentener, F. J., Olivier, J. G. J., Klein Goldewijk, C. G. M., and Lelieveld, J.: A 1×1 degree resolution dataset of historical anthropogenic trace gas emissions for the period 1890–1990, Global Biogeochem. Cy., 15, 909–928, 2001.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Kasibhatla, P. S., and Arellano Jr., A. F.: Interannual variability in global biomass burning emissions from 1997 to 2004, Atmos. Chem. Phys., 6, 3423–3441, doi:10.5194/acp-6-3423-2006, 2006.

van Vuuren, D., Smith, S. J., and Riahi, K.: Downscaling Drivers of Global Environmental Scenarios, Wiley Interdisciplinary Reviews, Climate Change, 1(3), 393–404, doi:10.1002/wcc.50, 2010.

van Vuuren, D. P., Meinshausen, M., Plattner, G.-K., Joos, F., Strassmann, K. M., Smith, S. J., Wigley, T. M. L., Raper, S. C. B., Riahi, K., de la Chesnaye, F., den Elzen, M., Fujino, J., Jiang, K., Nakicenovic, N., Paltsev, S., and Reilly, J. M.: Temperature increase of 21st century mitigation scenarios. P. Natl. Acad. Sci., 105, 15258–15262, 2008.

Vestreng, V., Myhre, G., Fagerli, H., Reis, S., and Tarrasón, L.: Twenty-five years of continuous sulphur dioxide emission reduction in Europe, Atmos. Chem. Phys., 7, 3663–3681, doi:10.5194/acp-7-3663-2007, 2007.

Xu, Y., Williams, R. H., and Socolow, R. H.: China's rapid deployment of SO_2 scrubbers, Energy Environ. Sci., 2, 459–465,2009.

Yevich, R. and Logan, J.: An assessment of biofuel use and burning of agricultural waste in the developing world, Global Biogeochem. Cy., 17, 1095–1034, 2003.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131–5153, doi:10.5194/acp-9-5131-2009, 2009.