Estimating historical anthropogenic global sulfur emission patterns for the period 1850–1990

Allen S. Lefohn\textsuperscript{a,*}, Janja D. Husar\textsuperscript{b}, Rudolf B. Husar\textsuperscript{b}

\textsuperscript{a}A.S.L. and Associates, 111 North Last Chance Gulch, Helena, Montana 59601, USA
\textsuperscript{b}Center for Air Pollution Impact and Trend Analysis (CAPITA), Washington University, St. Louis, Missouri 63130, USA

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Abstract

It is important to establish a reliable regional emission inventory of sulfur as a function of time when assessing the possible effects of global change and acid rain. This study developed a database of annual estimates of national sulfur emissions from 1850 to 1990. A common methodology was applied across all years and countries allowing for global totals to be produced by adding estimates from all countries. The consistent approach facilitates the modification of the database and the observation of changes at national, regional, or global levels. The emission estimates were based on net production (i.e., production plus imports minus exports), sulfur content, and sulfur retention for each country’s production activities. Because the emission estimates were based on the above considerations, our database offers an opportunity to independently compare our results with those estimates based on individual country estimates. Fine temporal resolution clearly shows emission changes associated with specific historical events (e.g., wars, depressions, etc.) on a regional, national, or global basis. The spatial pattern of emissions shows that the US, the USSR, and China were the main sulfur emitters (i.e., approximately 50\% of the total) in the world in 1990. The USSR and the US appear to have stabilized their sulfur emissions over the past 20 yr, and the recent increases in global sulfur emissions are linked to the rapid increases in emissions from China. Sulfur emissions have been reduced in some cases by switching from high- to low-sulfur coals. Flue gas desulfurization (FGD) has apparently made important contributions to emission reductions in only a few countries, such as Germany. © 1999 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Concerns about (1) anthropogenic aerosols affecting possible global change (Charlson et al., 1992) and (2) acid rain effects on the environment (Legge and Krupa, 1990) have prompted interest in the transformation and fate of sulfur in the atmosphere. Aerosols theoretically have the ability to cool the atmosphere and thus, anthropogenic sulfate particles may be an important factor in the climate system (Charlson et al., 1992). Their influence might be relatively small regionally, or perhaps counter the entire effect of greenhouse warming in the Northern Hemisphere (Charlson et al., 1992). The Intergovernmental Panel on Climate Change (IPCC) report (IPCC, 1994) reconfirmed the ability of aerosols to affect climate by changing the radiative balance of the atmosphere.

The development of a reliable regional emission inventory of sulfur as a function of time is an important first step in assessing the potential impact of sulfate aerosols on climate. A global sulfur emission inventory provides data for (1) global pollution models; (2) comparisons of sulfur emission patterns and atmospheric sulfur concentrations; and (3) emission projections. Estimations of sulfur dioxide emissions since the mid 1800s have been made by Bettelheim and Littler (1979), Dignon and Hameed (1989), Husar and Husar (1990), Örn et al.
(1996), and Mylona (1996), while others have estimated global sulfur emissions for shorter periods (e.g., Kellogg et al., 1972; Cullis and Hirschl, 1980; Möller, 1984; Varhelyi, 1985; Dignon, 1992; Spiro et al., 1992). Large discrepancies among the approximations probably reflect the many uncertainties associated with estimating sulfur emissions. In addition to the above, there are also continental and national estimates of sulfur emissions (e.g., Gschwandtner et al., 1986; Placet et al., 1990; Fujita et al., 1991; Kato and Akimoto, 1992).

This paper describes the development of a database that provides annual estimates of global emissions of sulfur from 1850 to 1990. The period was selected because it included the onset of 19th century industrialization with rapid growth in production of fuel and minerals and the more complex transitions of the 20th century. A common methodology was applied across all years and countries; the estimation of emissions was based on net production (i.e., production plus imports minus exports), sulfur content, and sulfur retention information associated with that country’s activities (Husar, 1986). Previous studies (e.g., Benkovitz et al., 1996) and estimates using alternative methodologies, (e.g., US emissions estimated by the Environmental Protection Agency) provided an important comparison with our own results. Our database differs from other global sulfur emission inventories by (1) detailing the production activities that lead to sulfur emissions by country and (2) calculating emission for each year. The application of a common methodology across all years and countries allows one to improve upon our estimates, as additional data become available. In addition, our study provides the opportunity to independently compare our historic global sulfur emission estimates with those of other investigators. Because the focus of this study was on annual, national sulfur emissions, no attempt was made to estimate the contribution of sulfur emissions from ships. Corbett and Fischbeck (1997) estimate that approximately 5% of sulfur emitted by all fuel combustion sources is associated with sulfur emitted from ships.

2. Approach

The trend in global sulfur emissions was estimated from production figures (Cullis and Hirschl, 1980; Husar, 1986; Spiro et al., 1992), relying on national statistics for the extraction of sulfur bearing fuels and metals. Because there is a significant international export-import trade in fossil fuels that physically separates the locations of production and consumption, emission computations require net production figures. The net production includes the extraction within the country, plus the imports, and minus the exports; provided there is no long-term accumulation (bunkering), consumption will equal net production. Because metallurgical production, rather than mining, gives rise to sulfur release, emissions were associated with this process.

The emission factors were estimated from sulfur content derived from (1) fuel and mineral analyses and (2) retention indices. These are spot, not continuous analyses, but sufficient because individual fuel and ore reservoirs have a reasonably constant sulfur content. Fuel combustion and metal smelting liberate some fraction of the solid sulfur into gaseous sulfur dioxide, the remainder being retained in mineral residues or ash. Additionally, some may be recovered or scrubbed from the effluent gases. Sulfur retention or release is estimated from process analysis at the combustion site (US EPA, 1995a).

Annual sulfur emission rate for a country’s fuel use or metal extraction can be calculated as a product of (1) net production rate of fuels and metals, (2) sulfur content, and (3) the release factor. The sulfur emission calculations require these three parameters for each country, year, and fuel/metal type (i.e., coal, oil, zinc, lead, copper, and nickel). With the exception of the US, it was assumed that the sulfur content depended on the country and fuel/metal, but not on time. The switch to low-sulfur coal in the US in the early 1970s resulted in varying sulfur content of fuels. The sulfur release factor was assumed to vary with time and extractive process but remain constant worldwide.

Production figures, as well as import/export data, were taken from Mitchell (1981–1983) which covers 1850–1975, although some early figures are fragmentary. Some of this missing data was estimated by linearly extrapolating backwards to a time when major use was assumed to begin: coal and metals, 1850 and oil, 1900. Gaps in the data (e.g., war times) were bridged by linear interpolation. Metal extraction figures were not interpolated because the gaps had little impact on emission estimates. Fortunately little data were absent for major producers; in all, 234 countries were included in the database. Production data include anthracite coal, bituminous hard coal, bituminous brown coal, crude petroleum, and natural gas, as well as copper, zinc, lead, and nickel. There are some problems of nomenclature of coals in statistical sources, but our data set labeled anthracite and bituminous hard coal as hard coal (with other coals such as lignite and bituminous brown coals labeled as brown coal).

Mitchell’s collection was supplemented by the fuel data set (Marland and Boden, 1994), prepared by the Carbon Dioxide Information Analysis Center (CDIAC) from the United Nations Energy database for 1950–1990. Marland and Rotty (1994) estimated that the uncertainty of the global fuel energy data is about 10%, but the uncertainty for individual countries is likely to be greater. The CDIAC category coal production was identified as hard coal and lignite-brown coal was identified as brown coal. These assignments produce fairly good agreement
between Mitchell and CDIAC coal figures for overlapping years. Additional mining data came from the Institute of Geological Sciences (1981,1982) and the US Bureau of Mines Statistical Yearbook (1986,1993), with some gaps filled from UN Statistical Yearbooks (1952,1971,1985) and the League of Nations (1936). Based on the above data sets, the production database is a homogeneous trend data set of 16 variables, (i.e., import and export for hard coal, brown coal, crude petroleum, and natural gas, as well as production data for the metals, copper, zinc, lead, and nickel). Lefohn et al. (1996) describe the consolidation and integration of the various data sets.

The sulfur content of the coals (summarized in Lefohn et al., 1996) used in this study have been estimated from sources such as the World Energy Conference (1986) and World Energy Council (1992) with additional information from Husar (1986), Afnogenova and Ryaboshapko (1988), Spiro et al. (1992), Dianwu (1995), and Kato (1996). The sulfur content of oil is largely based on the Bartlesville Project Office (1987) that includes virtually all oil producing regions. This compilation provides information on the sulfur content of gas diesel, jet fuel, motor gas, residual oil, and kerosene in the US for the period 1955–1990 (summarized in Lefohn et al., 1996). Not all refined products in the CDIAC database could be assigned sulfur contents from the Bartlesville compilation. The sulfur content for refined products in the United States was used for all countries for the time period defined, except for Asia, where the data from Kato (1996) were used. We assumed that the refining technology in practice defined the sulfur content of the refined products. Sulfur from natural gas was set to zero because its low sulfur content results in relatively small sulfur emissions. A summary of the sulfur content percentages used in the study can be found on the Internet (http://ourworld.compuserve.com/homepages/ASL_ASSOCIATES/).

Detailed sulfur content for metals was not available for individual countries, so US Environmental Protection Agency (EPA) emission factors for uncontrolled emissions, without sulfur recovery, were adopted (US EPA, 1995a). The emission factors expressed as ton of sulfur per ton of the metal produced are: copper smelting 1.2, zinc 0.5, lead 0.14, and nickel 1.20.

The sulfur release factor represents the fraction of the sulfur in fuels or metals released to the atmosphere, such that complete volatilization would correspond to a release factor of unity. In general, coal combustion in modern plants has a release factor close to one, although a small fraction of the sulfur is retained during coal preparation and in fly ash. If flue gas desulfurization (FGD) is installed, release factors are substantially reduced. For oil products, the refining tends to concentrate the sulfur in the heavy residues, such as residual fuel oil. Metal refining, where a fraction of the ore sulfur is recovered as a useful byproduct, has a release factor well below unity. The sulfur release factors depend largely on the technologies used in the processing of fuels and metals. The sulfur release factor for individual countries was not available, so we have used the same factors for all countries. The aggregated release factors were used to reflect broad historical trends (e.g., Darmstadter et al., 1987). Charts of aggregate release factors that depict the historical trend of these factors since 1850 were used. The uncertainties of these factors are a major contribution to the uncertainty of sulfur emission estimates. This is particularly true for sulfur emission estimates associated with the oils and metals. In our study, the release factor was decreased from unity in 1850 to 0.47 in 1990.

FGD controls began to be implemented around 1972. Using the cumulative total of thermal generating capacity information by year supplied by the International Energy Agency (IEA) in Paris (personal communication) and the cumulative total of FGD capacity (IEA London, personal communication), it was possible to determine the percentage of the FGD installed capacity by country by year. Based on information received from the IEA London (personal communication), the average efficiency of FGD controls by year was calculated (Table 1). Using information obtained from OECD (1973–1991), the fraction of coal used in power plants was obtained by country and year; this information was then combined with the above data and used in the final emission calculations.

3. Results and discussion

3.1. Sulfur production

The worldwide production of anthracite and bituminous hard coal exceeds brown coal production by approximately a factor of three (Fig. 1). Production rose between 1880 and 1910, followed by a plateau until about 1950 and a further rise between 1950 and 1990. World coal production has doubled since 1950, with brown coal becoming increasingly important. In 1900, the largest coal producing country was Great Britain, followed by the US and Germany. By 1960, the three major coal-producing countries were China, the US, and the USSR (Fig. 2), with China dominating by 1990 at 1.1 Gtonne/annum.

The world oil production grew exponentially from 1900 to the early 1970s (Fig. 1), but fluctuated about 10% over the past 20 yr. Import and export figures indicate that prior to World War II, international oil trade was insignificant, but by the 1960s represented about 50% of the global production. Since 1850, metal production from sulfur containing ores (i.e., copper, zinc, lead, and nickel) exhibits a rather varied behavior. Using the data developed from this
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study, Lefohn et al. (1996) found that copper, zinc, and lead have roughly comparable production rates from 1850 to about 1950. From 1950, copper and zinc production increased by a factor of 3-4, while lead production only doubled. Between 1970 and 1990, the global lead production actually declined. Nickel production grew from 1930 to 1990 but more slowly than the other three metals.

3.2. Comparison of global sulfur emission estimates

Möller (1984) estimated the global anthropogenic sulfur emissions from 1860 to 2000, and although applying a consistent methodology, the author used average emission factors rather than country- or source-specific information. Similarly, Dignon and Hameed (1989) used statistical models that relate sulfur dioxide emission to fuel consumption to estimate global emissions at 10-yr intervals from 1860 to 1980. Using data from multiple sources, Örn et al. (1996) published global sulfur emission estimates from 1860 to 1985. At 10-yr increments, Fig. 3 compares the global emission estimates by Möller (1984), Dignon and Hameed (1989), and Örn et al. (1996) with this study. Möller’s (1984) figures are consistently higher than the other studies and may reflect the importance of fuel switching and, in some cases, the use of FGD controls. The results of Dignon and Hameed (1989) agree closely with our study. However, the authors did not account for sulfur emissions associated with metal production. Thus, in effect, their reported results represent higher emission estimates than our own. In comparing our estimates with those of Örn et al. (1996), we find close agreement. From 1860 through 1930, our estimates are higher. In the two decades, 1970 and 1980, the estimates by Örn et al. (1996) were slightly higher. Differences between the sulfur emission estimates made by other investigators and our study are probably associated with different assumptions, such as fuel sulfur content and process emissions.

Spiro et al. (1992) determined sulfur emissions for all countries. Applying their methodology for all countries, the authors reported 88.4 Mtonne of anthropogenic sulfur emissions for 1980. The authors substituted their estimates for Europe, the USSR, North America, and Japan with existing inventories developed by the individual countries and recalculated global anthropogenic sulfur emissions at approximately 78 Mtonne. Our estimate for 1980 was approximately 66 Mtonne. The estimates by Spiro et al. (1992) were used by Örn et al. (1996). The values reported for 1985 and 1990 by Benkovitz et al. (1996), using an entirely different protocol than the one used in our study, were almost identical to ours (i.e., approximately 65 and 71.5 Mtonne, respectively).

In 1985, the European Council of Ministers made a decision to form a Commission for gathering,
coordinating, and ensuring the consistency of information on the state of the environment and natural resources in the European Community. One of the components of the program was the CORINE AIR emission inventory (CORINAIR). Estimates of sulfur emissions in Europe were made for 1985 and 1990 by CORINAIR (US EPA, 1995b), and Mylona (1996). For example, in 1990, except for a few countries (e.g., Germany, Bulgaria, and Italy), the European estimates from this study were close to CORINAIR (Fig. 4). Differences between the sulfur emission estimates may be associated with differences in the published production levels that we used and modified government statistics.

One geographic area where our estimates disagreed considerably was Japan. Sulfur emission estimates made by Kato (1996) for Japan ranged from 37 to 66% of estimates made in this study. Our study considered FGD, but not coal washing, which may account for part of the lower values published by Kato (1996). We do not have a full explanation for the difference between our results and those of Kato (1996).

The EPA (1995b) summarized trends for the US for the period 1900–1994, identifying a downward trend from the mid-70s. However, our study suggests level emissions from the mid-70s to early 80s, a downward trend until the mid-80s, and then a gradual increase into the 90s.
We estimated US emissions at 10.8 and 12.5 Mtonne for 1985 and 1990, while the EPA estimated 10.6 and 10.2 Mtonne. Applying the data from our study, Lefohn et al. (1996) suggest some of the difference that occurs after 1985 between our estimates and those of EPAs may be associated with the consumption of residual fuel oil. In addition, the EPA (1994) assumed sulfur dioxide and \( \text{PM}_{10} \) controls to be totally effective, while our study rated sulfur controls only 85% efficient.

3.3. Estimated global sulfur emissions

Since 1850, the global anthropogenic sulfur emission trend for 1850–1990 shows that there has been a general
increase of sulfur emissions (Fig. 5). We estimate that emissions in 1850 (approximately 1.2 Mtonne) were about 1.7% of the current values (71.5 Mtonne). There was some leveling off, beginning in 1913, with a decline during World War I. The great depression (1930–1932) led to a marked decrease in global emissions with increases in 1933 through 1944, partly associated with World War II. The post war years saw a continuous increase, with a drop in 1981–1983, resulting primarily from declining oil demand during the global recession. Fig. 5 compares emissions from North America, Europe, and Asia. While North American, and in some cases, European emissions have been leveling off, rapid increases are occurring in Asia.

The effect of sulfur emissions in the US as a function of applying different corrections is shown in Fig. 6. The
corrections used in our study were (1) FGD controls, (2) the switch to low-sulfur coal (2.3–1.3%, from 1973 to 1990), and (3) increased retention in oil refining on US emissions. The top line shows the level of sulfur emissions that would have occurred if corrections were not applied. The bottom line reflects estimated emissions using the corrections implemented in our study. Note that the bottom line and the line representing no FGD controls are close to one another. Alternatively, the line representing the effects of applying no FGD controls and fuel switching is much higher than the line representing no FGD controls. This observation implies that FGD controls had much less impact on sulfur emissions than switching to low-sulfur coal. Reviewing the data from our study, we found that globally, flue gas desulfurization (FGD) has made important contributions to emission reductions in only a few countries, such as Germany (Lefohn et al., 1996).

Fig. 7 shows that the US, the USSR, and China were the main sulfur emitters in the world in 1990. The USSR and US appear to have stabilized their emissions over the past 20 years, such that recent increases can be linked to industrialization in China. In 1990, these three countries accounted for 53% of the global sulfur emissions.

Using our data, Lefohn et al. (1996) discuss the changes in sulfur emissions over the years. The authors note that the combustion of coal is the dominant anthropogenic source of sulfur. The contribution from oil products rose rapidly between 1950 and 1980, but has fluctuated from 1980 to the present. Sulfur emissions associated with copper smelting peaked in 1974. Therefore, the steady increase in emissions since the 1970s is attributable to increased coal combustion. The US shows substantial sulfur emissions from hard coal prior to 1940; China and the USSR emitted insignificant quantities in the first half of the 20th Century. Sulfur emissions from brown coal are mainly from Europe.

Major emissions from oil consumption occur in the US, the USSR, and Saudi Arabia, with significant contributions from Mexico, China, and Japan. Historically we see an early dominance by the US and the subsequent emergence of the other two major emitters since the 1950s (Lefohn et al., 1996).

The sulfur emissions from copper show an identical pattern to copper production, because both sulfur content of ores and the release factor were assumed to be geographically constant. Since 1910, the copper-related emissions from the US have fluctuated, while there have been steady increases from Chile since 1920 (see Lefohn et al., 1996). In 1990, the primary emitter was Chile, where the emissions of 0.9 Mtonne were a major part of its contribution. The USSR and Canada dominate emissions associated with nickel production, while major zinc related emissions arise from Canada and Australia. Lead derived emissions are relatively small (i.e., less than 0.04 Mtonne), with the largest in 1990 from Australia, the US and the USSR. In many smaller countries, where mining makes a big contribution to the economy, metallurgical emissions can be the dominant source of anthropogenic sulfur.

4. Conclusion

This study estimated annual national sulfur emissions from 1850 to 1990, based on net production, sulfur
content and sulfur release factors for each country's production activities, using a common methodology applied across all years and countries. In 1990, the spatial pattern of emission shows that the US, the USSR, and China were the main sulfur emitters (i.e., approximately 50% of the total). While the USSR and the US appear to have stabilized their sulfur emissions over the past 20 yr, recent increases in global emissions are linked to the rapid industrialization in China. Combustion of coal remains the dominant anthropogenic source of sulfur. The US and Europe, unlike China and the USSR, showed substantial sulfur emissions from hard coal prior to 1940. Although the contribution from oil products rose rapidly from 1950, growth has fluctuated since 1980. Major emissions from oil consumption occur in the US, the USSR, and Saudi Arabia, with significant contributions from Mexico, China, and Japan. Historically, we see an early 20th Century dominance by the US and the subsequent emergence of the other two major emitters since the 1950s. Sulfur emissions have been reduced in some cases by switching from high- to low-sulfur coals. Flue gas desulfurization (FGD) has made important contributions to emission reductions in only a few countries, such as Germany.

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