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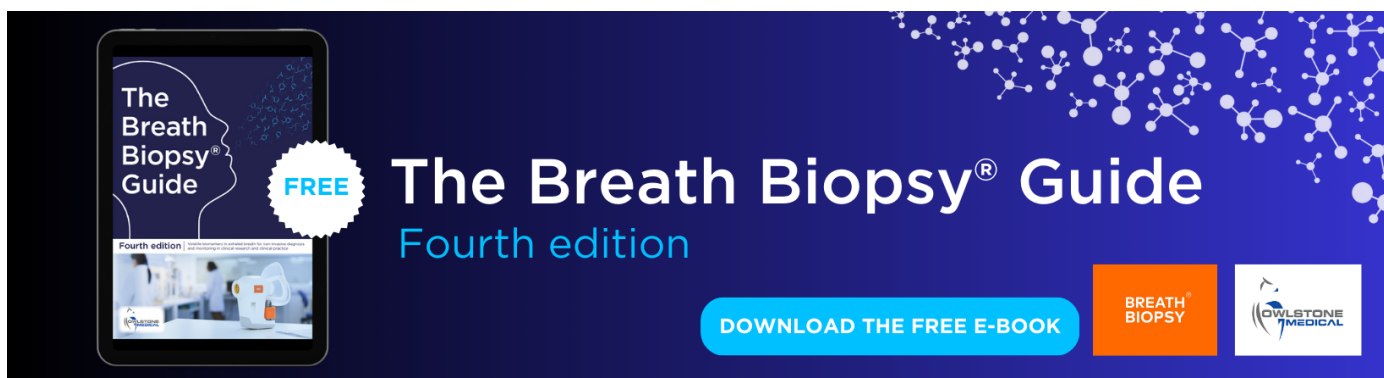
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The last decade of global anthropogenic sulfur dioxide: 2000–2011 emissions

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Abstract

The evolution of global and regional anthropogenic SO₂ emissions in the last decade has been estimated through a bottom-up calculation. After increasing until about 2006, we estimate a declining trend continuing until 2011. However, there is strong spatial variability, with North America and Europe continuing to reduce emissions, with an increasing role of Asia and international shipping. China remains a key contributor, but the introduction of stricter emission limits followed by an ambitious program of installing flue gas desulfurization on power plants resulted in a significant decline in emissions from the energy sector and stabilization of total Chinese SO₂ emissions. Comparable mitigation strategies are not yet present in several other Asian countries and industrial sectors in general, while emissions from international shipping are expected to start declining soon following an international agreement to reduce the sulfur content of fuel oil. The estimated trends in global SO₂ emissions are within the range of representative concentration pathway (RCP) projections and the uncertainty previously estimated for the year 2005.

Keywords: sulfur dioxide, global emissions, RCP, anthropogenic

 Online supplementary data available from stacks.iop.org/ERL/8/014003/mmedia

1. Introduction

Sulfur dioxide emissions have substantial impacts on human health (e.g., Pope *et al* 2007), terrestrial and aquatic ecosystems (e.g., Likens and Bormann 1974) and have come under increasing regulation world-wide (Smith *et al* 2011). Sulfur dioxide is also a principal precursor of anthropogenic aerosols in the atmosphere, acting as a cooling agent (e.g., Seinfeld and Pandis 1998). Updated estimates of SO₂ emissions are a central input needed to evaluate regional and global trends of aerosol optical depth (AOD) (e.g., Hsu *et al* 2012, Zhang and Reid 2010). These are also useful to evaluate the evolution of emissions over 2000–2010 in

the RCP (representative concentration pathways) scenarios, which are greenhouse gas and air pollutant emission pathways developed for the climate modeling community (Moss *et al* 2010, van Vuuren *et al* 2011). The RCPs are being used in the latest global climate model inter-comparison CMIP5 (Taylor *et al* 2011, 2012), which will be evaluated as part of the IPCC AR5 process.

Previous assessments of the global emissions of sulfur dioxide (SO₂) included estimates until the year 2005 (Smith *et al* 2011) and showed substantial regional changes in emissions patterns in recent decades, along with a slight increase in global emissions from 2000 to 2005. Several regional studies have compiled inventories for more recent years (e.g., Tørseth *et al* 2012, EEA 2011, Lu *et al* 2011, Wang and Hao 2012, US EPA 2012) and a number of countries submit emissions data to international bodies under various international treaties (e.g., UNFCCC: www.unfccc.org, UNECE CLRTAP: www.ceip.at). However, a global



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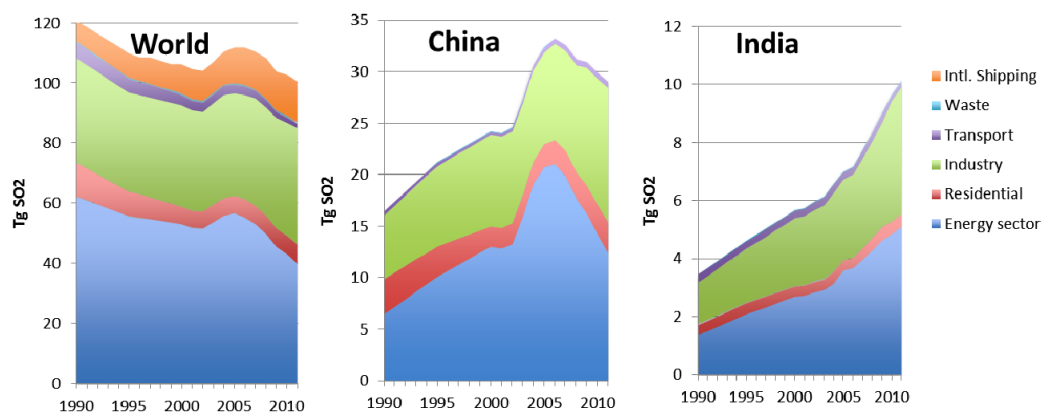


Figure 1. Sectorial trend in global, China, and Indian SO₂ emissions since 1990, Tg SO₂. Note different scales: i.e., India about 1/3 of China and the latter 1/3 of the world emissions.

assessment covering recent years has been missing and we, therefore, present here an estimate of global SO₂ emissions up to 2011.

This work makes use of the statistical data on energy consumption until 2011 (BP 2012, IEA 2012), and recent reporting under several international treaties. The results are compared with previous global and regional assessments.

2. Methodology

This work relies on existing methods and tools to estimate sulfur emissions as described in Smith *et al* (2011), Cofala *et al* (2007) and Amann *et al* (2011a). In particular, the GAINS (Greenhouse gas–air pollution Interactions and Synergies) model (Amann *et al* 2011a) was used to calculate land based anthropogenic emissions for 2000, 2005 and 2010, while annual energy consumption data (BP 2012) was used to scale these estimates for intermediate years and extending to produce a preliminary estimate for 2011 (see supplemental material (SM) available at stacks.iop.org/ERL/8/014003/mmedia).

Emissions were calculated at a detailed country, regional and sectoral level and are presented here for aggregated regions (see figure 1, tables S-1 and S-3 in SM available at stacks.iop.org/ERL/8/014003/mmedia) and key sectors including power plants (shown in energy sector in figures and tables), industrial combustion and process (industry), domestic combustion (residential), road and non-road transport, and waste burning (see table S-2 available at stacks.iop.org/ERL/8/014003/mmedia). Sulfur dioxide emissions from international shipping were estimated starting with the work of Eyring *et al* (2010) and extending this using energy and shipping activity data as described in Smith *et al* (2011) and in the SM (available at stacks.iop.org/ERL/8/014003/mmedia). Gridded emissions ($0.5^\circ \times 0.5^\circ$) were calculated for anthropogenic land based sources (see figure S-3 in the SM available at stacks.iop.org/ERL/8/014003/mmedia) using the set of proxies developed within the Global Energy Assessment (GEA 2012); these are consistent with proxies applied within the RCP projections as described in Lamarque *et al* (2010) and were modified to accommodate

for more recent information where available, e.g., population distribution, open biomass burning; see the SM (available at stacks.iop.org/ERL/8/014003/mmedia) for more details.

2.1. Current legislation

Calculation of emissions of air pollutants has been performed by assuming current policies in each country, i.e., measures that were in force by 2010. In particular, for Europe all emission limit values and fuel quality standards have been included, as used in the analysis for the revision of the Gothenburg Protocol to the UN Convention of Long-Range Transboundary Air Pollution (CLRTAP) (Amann *et al* 2011b) and recent work on the revision of the EU thematic strategy on air pollution (Amann 2012). For large combustion plants, this assessment includes sulfur emission limit values according to the large combustion plants directive (LCPD). The LCPD has been recently replaced with the more stringent industrial emissions directive (IED), which entered into force in 2010 and therefore has no or only very limited impact by 2011.

For non-European countries policies have been assessed based on available literature (Cofala *et al* 2007) and more recent studies (Klimont *et al* 2009, Lu *et al* 2011, Xing *et al* 2011, Zhang *et al* 2012, Wang and Hao 2012, Xu *et al* 2009, Xu 2011). Assumptions about emission controls in the power sector have been cross-checked with detailed information from the database on world coal fired power plants (IEA CCC 2012).

GAINS estimates for 2005 and 2000 have been updated, taking into account recent emission inventories as submitted by members of the CLRTAP to the Convention. Similarly, updates of emission inventories and assessments for other countries have been taken into account (e.g., Granier *et al* 2011, Wang and Hao 2012, Zhang *et al* 2012). For instance, in China installation and increased operation of flue gases desulfurization (FGD) for existing and new plants (Xu 2011, Zhang *et al* 2012) has caused a reduction of emissions compared with previous expectations (e.g., Zhang *et al* 2012, Xu *et al* 2009).

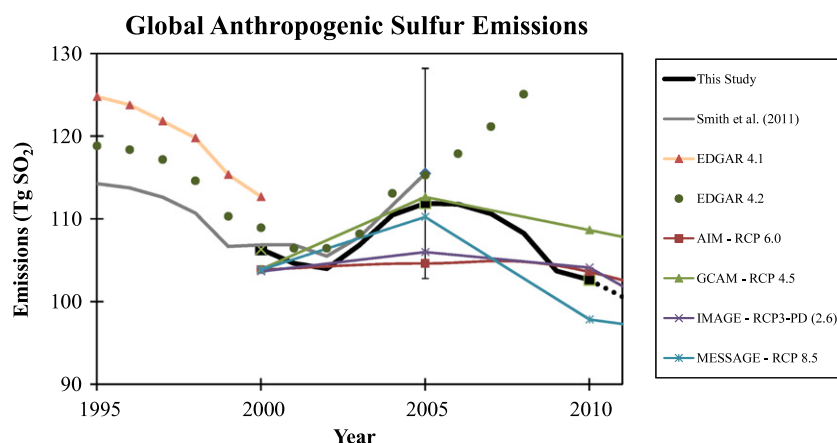


Figure 2. Global sulfur dioxide emissions from the current study as compared to several previous inventories, including EDGAR 4.1, EDGAR 4.2, and Smith *et al* (2011). A bar for the uncertainty estimate for the Smith *et al* (2011), 2005 emissions estimate is shown. Also shown are the global SO₂ estimate for the four RCP scenarios. All emission estimates exclude open burning from grasslands, savannahs, forests, and deforestation.

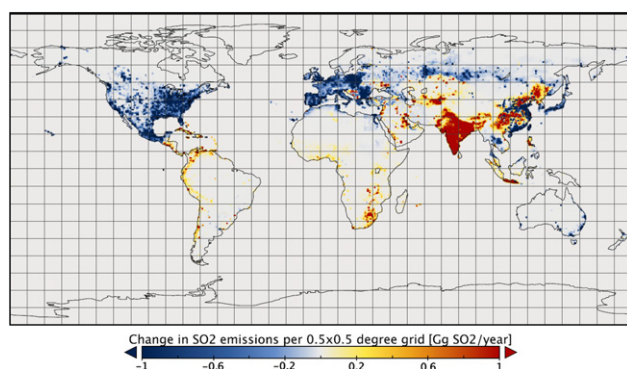


Figure 3. Change in regional distribution of anthropogenic land based SO₂ emissions. Changes indicated as a difference between 2010 and 2005 emissions in 0.5° × 0.5° grids.

3. Results

Overall, global emissions in the GAINS estimate are similar to those reported in previous versions of GAINS, also matching well the increase from 2000 to 2005 reported in Smith *et al* (2011) (figure 2). This trend has changed since 2005, however, with global emissions in 2010 estimated to be lower than in 2000 by about 3%. While there is a net increase over 2000–2010 period from the EECCA (Eastern Europe, Caucasus and Central Asia), China, India, and international shipping, these increases were smaller than emission reductions in North America (US and Canada) and Europe, leading to a net decrease in global emissions (figure 1, tables S-1 and S-2 available at stacks.iop.org/ERL/8/014003/mmedia). Since 2000, the contribution of Asia increased from about 41% to 52% while that of North America and Europe (including Russia and EECCA countries) declined from 38% to 25% (table S-1 available at stacks.iop.org/ERL/8/014003/mmedia). The energy sector is the main contributor to the recent overall reduction, with its share dropping from about 50% in 2000 to below 42% in 2010. At the same time, emissions from industry and international shipping continued

Table 1. Change in emissions for broad world regions from the GAINS model estimate.

Region	Change in emissions (Gg SO ₂)	
	2000–2005	2005–2010
Africa	471	450
China	8203	–2559
India	1178	2655
Middle East	240	381
Other Asia and Pacific	–632	–295
Europe ^a	–2792	–3621
EECCA ^b	–229	628
Russian Federation	278	–1556
Latin America and Caribbean	–1946	–310
US and Canada	–1447	–6660
International shipping	2260	1605
Global	5584	–9281

^a Excluding EECCA countries and Russian Federation.

^b Eastern Europe, Caucasus and Central Asia (EECCA) includes countries of Former Soviet Union (FSU) excluding Russian Federation and Baltic States (Lithuania, Latvia, Estonia) which joined the European Union.

to increase from 32% and 9% in 2000 to 38% and 13% in 2010 (table S-2 available at stacks.iop.org/ERL/8/014003/mmedia).

The varying regional trends in SO₂ emissions since 2005 are illustrated in figure 3 and table 1. Reductions in the United States were particularly large over this period, estimated to be over 6000 Gg SO₂ lower in 2010 than in 2005 (US EPA 2012); consistent with our assessment. We estimate that emissions in the European Union (EU) decreased since 2005 by about 3700 Gg SO₂. Compared to the officially reported emissions to the CLRTAP (www.ceip.at), GAINS model underestimates EU SO₂ emissions in 2010 by about 3% primarily due to more optimistic assumptions on abatement penetration. According to the most recent officially submitted information, emissions in power sector, refineries and industrial combustion were somewhat higher owing to slightly delayed compliance or accepted derogations. Emissions in China were estimated to

have peaked about 2006, and then declined towards 2010 as operation of emission controls increased in the electric power generation sector. In contrast, SO₂ emissions from India continue to increase sharply.

As a check on the emission estimate, total emissions from petroleum from 2000 to 2010 from this estimate match well a top-down estimate of global petroleum mass balance (see the SM available at stacks.iop.org/ERL/8/014003/mmedia). The petroleum mass balance indicates a slightly larger absolute total, but the difference is well within the error of these estimates.

We estimate that sulfur dioxide from international shipping increased from 9800 Gg SO₂ in 2000 to 13 600 in 2010, closely following a 40% increase in goods loaded over this period, consistent with available sales data (see the SM available at stacks.iop.org/ERL/8/014003/mmedia). Uncertainties in this estimate are substantial, and include poorly reported bunker fuel sales, uncertain sulfur content of bunker fuels, and any changes in shipping operations that may have occurred in 2009 and 2010 that could not be accounted for in data used here.

3.1. Emissions in Asia

China continues to be the largest single contributor to global SO₂ emissions representing nearly 30% of the global total in 2010. We estimate that, while emissions continued to grow rapidly since 2000, emissions peaked in about 2006 at 33 Tg SO₂ and declined towards 2010 primarily because of increasing capacity (and operation) of FGD installations (Xu 2011). A similar development of emissions in China in the last decade has been also estimated by Lu *et al* (2011), Zhang *et al* (2012), Wang and Hao (2012) as well as indicated by the analysis of the remote sensing data (Gottwald and Bovensmann 2011). The latter study has shown that analysis of GOME and SCIAMACHY data over East China pointed to a decline in the SO₂ column after 2007. Our results show a very similar evolution of emissions in China to that of Zhang *et al* (2012) where power sector is the key contributor to stabilization and then reduction of emissions (figure 1). At the same time, similar controls are lacking, or lagging behind, in industry, which results in significant growth of this sector; we estimate that industrial emissions of SO₂ grew from 2005 by about 40% by 2010 (figure 1). These developments contribute to the rapidly changing landscape of SO₂ emissions in China. These trends have resulted in a decrease in the fraction of China emissions from the power sector from 60% in 2005, down to 40% of the total in 2010; a reduction of about 30%. A similar result is found by Lu *et al* (2011).

The second largest emitter in Asia is India where we see no sign of slowing growth of emissions. This is largely due to an expansion of coal consumption in the power sector where current legislation does not require installation of flue gas desulfurization (FGD). Emissions from industry follow a similar path; in fact growing even faster (figure 1). Although, total Indian SO₂ emissions are not as large as that of China, they exceeded in 2010 emissions of US and have grown by over 40% since 2005 (a slightly slower growth rate than found

by Lu *et al* 2011). If these trends continue, India will become an increasingly important contributor to global sulfur dioxide emissions, although emissions in 2011 were estimated to be only a third of China's total.

3.2. Comparison with other studies

The estimates developed within this study compare well with Smith *et al* (2011) and EDGAR 4.2 for the period prior to 2005. A more detailed discussion of several studies has been presented in Smith *et al* (2011) and Granier *et al* (2011). For more recent years, the trend calculated in this work is different from EDGAR 4.2 but is within the range of representative concentration pathways (RCP) projections (figure 2). The trend estimated by EDGAR 4.2 is similar to the coal consumption growth in China and a hypothetical trajectory of SO₂ emissions shown by Zhang *et al* (2012) if the further FGD controls in power sector were not introduced. The current estimate and the RCP scenarios are within the estimated uncertainty in 2005 (Smith *et al* 2011). The annualized GAINS estimates are similar to most country-level inventory estimates; a more detailed discussion and comparison of GAINS against UNFCCC submissions is presented in the SM (available at stacks.iop.org/ERL/8/014003/mmedia). Tørseth *et al* (2012) presented a trend analysis of European emissions up to 2009 and shown a reduction of about 6090 Gg SO₂ (−41%) since 2000 for the European territory excluding Russia. We estimated for the same time period and area a reduction of 6144 Gg SO₂ which represents a decline of about 41% since 2000 (compare table S-3 in the SM available at stacks.iop.org/ERL/8/014003/mmedia). Also for EU-15, very similar reductions are shown in Tørseth *et al* (2012) and GAINS, i.e., decline by 58 and 57%, respectively (table S-3 in the SM available at stacks.iop.org/ERL/8/014003/mmedia).

4. Discussion

While global sulfur dioxide emissions have generally declined since the mid 1970s, the upturn in emissions from 2000 to 2005 (Smith *et al* 2011) raised the possibility that increasing aerosol forcing might be offsetting recent warming from increasing greenhouse gas concentrations. We find here that this increase was short lived: global SO₂ emissions have decreased since about 2006, with a change in trend in China due to implementation of sulfur emission controls in energy sector and further large reductions in more affluent regions, particularly the United States and Europe (table 1 and table S-1 available at stacks.iop.org/ERL/8/014003/mmedia). While the current reductions in emissions from China are from the implementation of end-of-pipe emission controls on electric power plants, continued reductions in emissions from China, however, will ultimately require reductions from the much more numerous and varied industrial sources, including boilers, smelters, coke ovens, and brick kilns.

One sector with increasing emissions is international shipping (Eyring *et al* 2010, Smith *et al* 2011), but emissions increases here will slow and ultimately fall if the

MARPOL limits on sulfur in shipping fuels are successfully implemented³.

Smith *et al* (2011) estimated a global uncertainty in 2005 SO₂ emissions of $\pm 11\%$. While we have not repeated this calculation here, uncertainty in current (2010/2011) emissions is likely larger due to the larger contribution of particularly uncertain emissions from developing countries and international shipping. Reducing this uncertainty will require more robust estimates of both the sulfur content of coal and heavy oils (particularly for shipping) and more detailed tracking of the operation of emission controls.

Zhang and Reid (2010) find a 'statistically negligible' global trend in aerosol optical depth (AOD) over the oceans for 2000–2009. This would appear to be consistent with the global emissions trends estimated in this work, where global emissions (terrestrial plus ocean) increased and then decreased over this time period, but with an increase in international shipping emissions. The global AOD response to emissions will depend on differing regional responses to emissions in a context of significant variability. The global decline in emissions that we find for the past several years may be over too short of a period to provide a detectable signal at this time.

The Zhang and Reid finding of a regional increase over the Indian Bay of Bengal, the east coast of Asia, and the Arabian Sea is generally consistent with our regional emission estimates. Different factors will likely be impacting AOD trends near China in the coming years. We estimate that terrestrial SO₂ emissions in China are currently falling, while shipping emissions still appear to be increasing. Emissions of primary carbonaceous aerosol may also be increasing as they are impacted much less by electric power plant emission controls.

Declining emissions from both China, still the world's largest source of sulfur dioxide, the planned decrease in shipping emissions, and continued decreases industrialized countries are likely to lead to a further net decrease in global sulfur dioxide emissions in the future. This will have regional and global consequences, decreasing the net negative radiative forcing from sulfur dioxide emissions (Forster *et al* 2007) that is 'masking' some of the impact of increasing greenhouse gases. A continued decline in global SO₂ emissions will likely result in an increase in the rate of future climate change.

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