Global and regional trends of atmospheric sulfur

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The profound changes in global SO2 emissions over the last decades have affected atmospheric composition on a regional and global scale with large impact on air quality, atmospheric deposition and the radiative forcing of sulfate aerosols. Reproduction of historical atmospheric pollution levels based on global aerosol models and emission changes is crucial to prove that such models are able to predict future scenarios. Here, we analyze consistency of trends in observations of sulfur components in air and precipitation from major regional networks and estimates from six different global aerosol models from 1990 until 2015. There are large interregional differences in the sulfur trends consistently captured by the models and observations, especially for North America and Europe. Europe had the largest reductions in sulfur emissions in the first part of the period while the highest reduction came later in North America and East Asia. The uncertainties in both the emissions and the representativity of the observations are larger in Asia. However, emissions from East Asia clearly increased from 2000 to 2005 followed by a decrease, while in India a steady increase over the whole period has been observed and modelled. The agreement between a bottom-up approach, which uses emissions and process-based chemical transport models, with independent observations gives an improved confidence in the understanding of the atmospheric sulfur budget.

There have been large changes in the global and regional SO2 emissions over the last decades. After a steady increase in emissions of SO2 since the beginning of the twentieth century1, the growing awareness of the negative effects of air pollution on environment and human health, gave rise to international and national legislation on emission-reductions2–4. European and North American SO2 emissions were reduced by 70–80% since 19902,3,5, with the largest emission reductions in North America occurring in the last part of the period1,3,6–8, while in Europe the reductions were largest in the first part of the period1,2,5,9–12. There were also substantial reductions earlier, from 1980–19901. These large regional SO2 emission reductions resulted in a global decrease from around 1980 until 20005, after which the global emissions increased due to a sharp rise in the Chinese emissions up to around 2006, followed by a declining global trend, mainly due to stricter emission controls in China13–20 and trends in Europe and North America1,4,12,14. Not all countries have implemented effective emission controls. In India, the emissions continue to increase1,21,22, and India is now the world’s second largest SO2 emitting country after China23.
The large changes in SO$_2$ emissions have also influenced the radiative forcing of aerosols. Sulfate aerosols have an impact on climate directly by scattering solar radiation and thus cooling the earth’s surface, and also an indirect effect on the formation of clouds and precipitation$^{24}$. The global mean radiative forcing, due to aerosol changes over the 1990–2015 period, increased by about +0.1 W m$^{-2}$, but the uncertainty is large$^{25}$. The main reason for the increased positive radiative forcing of aerosols over this period is the substantial reduction of global mean SO$_2$ emissions coupled with higher black carbon emissions$^{25}$. Furthermore, the reductions of SO$_2$ emissions over Europe are simulated to exert a local radiative forcing of 3–4 W m$^{-2}$ for the same period$^{25}$.

The trends calculated by the global aerosol models have seldom been compared to trends in observations. Trend assessments have mostly been done on regional and national observations$^{6–8,11,26,27}$, though there are also studies which combine modeled and observed trends$^{9,10,28–30}$. Global or hemispheric assessments have on the other hand, been done for short time periods or selected years only$^{31–33}$, or on somewhat limited dataset$^{36}$ and few models$^{39}$. In this study, we have compiled monthly average mean concentrations of SO$_2$ sulfate in aerosols and wet deposition of sulfate from major regional networks from 1980 onward until 2015 when available (See Fig. S1 in Supplementary Material (SM)). These trends have been compared to trends estimated by different global models for the period 1990–2015.

We address the question whether global climate models are able to reproduce the recent observed changes in regional amplitudes of atmospheric sulfur and its inhomogeneous spatial distribution. Reproduction of historical atmospheric pollution levels is crucial to demonstrate that such models are capable to predict the impact of air pollution on climate and air quality in future scenarios. In turn, the consistency allows to clearly attribute the observed concentration changes to the emission changes. This is particularly important since a central objective of the long-term regional monitoring programmes is to document changes in atmospheric composition and test the effectiveness of environmental policies. This work may demonstrate the usefulness of strengthening the international cooperation among regional measurement networks.

Results and Discussions

How have the changes in SO$_2$ emissions affected average regional sulfate concentrations?

The regional contributions to the global trends in sulfur emissions from 1990 to 2015 are illustrated with five year intervals in Fig. 1. The regional evolution of emissions are compared to annual average observed and modeled sulfate concentrations at the sites with measurements of sulfate in aerosols since 1990 (North America and Europe) or 2000 (East Asia). The model results are given as an ensemble mean from six global models. A complete statistical trend analysis of all the measured and modeled data of sulfate in aerosols, for several (sub-)periods is
given in Table 1. The observations, emissions, and the model results show a consistent and substantial change in the regional sulfur budgets.

| Time period | Region | Nr. of stations | Average annual trend (STD) %/year | Absolute annual trend (STD) µgS/m³/year |
|-------------|--------|----------------|-----------------------------------|----------------------------------------|
| 1980–1990   | Europe | 16             | −2.56 (3.10)                      | −0.048 (0.094)                         |
| 1990–2000   | Europe | 41             | −5.16 (2.11)                      | −0.073 (0.052)                         |
| 1990–2000   | North America | 101   | −2.08 (1.44)                      | −0.024 (0.025)                         |
| 1990–2015   | Europe | 33             | −2.93 (0.69)                      | −0.031 (0.015)                         |
| 1990–2015   | North America | 124   | −2.14 (0.67)                      | −0.026 (0.024)                         |
| 2000–2010   | East Asia | 13    | 0.44 (4.25)                       | 0.003 (0.034)                          |
| 2000–2010   | Europe | 43             | −2.86 (2.20)                      | −0.029 (0.041)                         |
| 2000–2010   | North America | 227   | −3.03 (1.72)                      | −0.029 (0.029)                         |
| 2000–2015   | East Asia | 13    | 2.68 (9.41)                       | 0.003 (0.037)                          |
| 2000–2015   | Europe | 36             | −2.67 (2.03)                      | −0.025 (0.028)                         |
| 2000–2015   | North America | 218   | −3.15 (1.30)                      | −0.028 (0.029)                         |

Table 1. Average absolute and per cent trends in sulfate in aerosols. Data in bold indicate trends not consistent with the observations within the standard deviation (STD).

Globally the SO₂ emissions were reduced by 55 TgS (31%) from 1990 to 2015. Individual regions have had different contributions to the global emission budget throughout this period (see Fig. 1) as also documented in other studies. The largest decrease in global SO₂ emissions occurred in the first decade, from 1990–2000 and was mainly due to a large reductions in Europe (−42 TgS/−54%). There was a smaller decrease in North America (−7 TgS/−21%) during this time, and an increase in East Asia (+10 TgS/32%). In comparison, in the following period 2000–2015, emissions in Europe and the US decreased by a similar total amount (−14 and −13 TgS) or in relative terms, respectively by −40% and −50%. In Eastern Asia, there was an increase of the emissions up to 2005 by more than +20 TgS (70%), while in the last ten years from 2005 to 2015 there has been a reduction, we have used emission inventories with a decrease of −6 TgS (−13%). For the whole 25 year period from 1990 to 2015, India’s emissions increased from 4.5 to 15 TgS, while in Africa only small changes occurred, +1 TgS (8%).

The emission inventories in China, which is the main contributor to the SO₂ emissions in East Asia, have been extensively studied the last years, and the most recent estimates indicate that the negative trends in the inventory used in our study is most likely underestimated. Zheng et al. estimate SO₂ reduction in China of about 62% in the period 2010–2017, with the largest decrease after 2013 when the China’s Clean Air Action was implemented. This rapid decline in the recent years appears to be confirmed with satellite observations. The main reason for these differences is that achieved effectiveness of policies implemented following China's Clean Air Action plans was not anticipated in earlier inventories. To illustrate the difference between the emissions used by the models in this study and the most recent estimates for the last ten year period in East Asia, the new inventories are included in Fig. 1, showing a decrease of −18 TgS (−34%) between 2005–2015 (shown as ‘Alternative emissions’).

The aerosol sulfate trends in observations and ensemble model results at the measurement sites with long term trends compare well with the trends of SO₂ emissions for all regions, especially North America and Europe (See Fig. 1). The ensemble model results and observations give comparable annual reductions of sulfate in aerosols by around 5.2%/y and 2.0%/y in Europe and North America, respectively, for the period 1990–2000 (Table 1). For the 2000–2015 period, the ensemble model and observations agree with 3.1%/y reduction in North America, while in Europe the ensemble model mean shows a higher relative reduction of 3.3%/y, compared to 2.7%/y in the observations, although the absolute changes are similar (−0.025 µgS/y, see Table 1), though the modelled and observed trends are comparable within the uncertainties for both relative and absolute changes per year.

The temporal development in East Asia was different with an increase in SO₂ emissions up to 2005, and a decline thereafter. The observations from this region do not capture the complete period, starting only in 2000, with relatively few sites. However they indicate the same tendency for an increase from 2000 to 2005 followed by a decline from 2005 to 2010. For the 2000–2010 period, both the ensemble model mean and the observations show a small average increase of 0.4%/y, although with quite high uncertainty, Table 1. There is an observed increase from 2010 to 2015, though it should be emphasized that the variability between the sites is high, and when considering that the emissions most probably have decreased more in the last five-year period, the increase in the observed sulfate concentrations seems to be non-representative. One should note that none of the sites with aerosols measurements used in this study are located in China. The large positive trend is mainly caused by two sites in Indonesia with 20–30% increases, which were not captured by the models. Whether this was due to local influence or long-range transport is difficult to say. It could be due to influences from volcanic activities in the region, but there does not seem to be an increase in eruptions the latter years nor do we see the same signal for the measured SO₂.

Comparability between sulfur trends in gaseous phase, aerosols and wet deposition. The trends of SO₂, sulfate in aerosols and sulfate in precipitation were compared using the EMEP MSC-W model result and the global observation set. It is important to note that the EMEP MSC-W model results are close to the ensemble mean for aerosol sulfate, as discussed in the next section. In addition to the measurement sites that provided long term observations of sulfate in aerosols (discussed above), observations of sulfate in precipitation
Average absolute and per cent trends in SO2. Data in bold indicate trends not consistent with the observations within the standard deviation (STD).

Table 2. Average absolute and per cent trends in wet deposition of sulfate. Data in bold indicate trends not consistent with the observations within the standard deviation (STD).

| Time period | Region         | Nr. of stations | Average annual trend (STD) %/year | Absolute annual trend (STD) kgS/ha year |
|-------------|----------------|-----------------|-----------------------------------|----------------------------------------|
| 1980–1990   | Europe         | 23              | −2.37 (2.33)                      | −0.30 (0.36)                           |
| 1980–1990   | India          | 10              | −13.7 (25.6)                      | 0.00 (0.18)                            |
| 1980–1990   | North America  | 78              | −1.80 (4.09)                      | −0.06 (0.18)                           |
| 1990–2000   | East Asia (China) | 3              | −1.27 (2.67)                      | 5.07 (0.36)                            |
| 1990–2000   | Europe         | 60              | −4.02 (4.44)                      | −0.29 (0.29)                           |
| 1990–2000   | India          | 10              | 22.6 (16.0)                       | 4.31 (1.53)                            |
| 1990–2000   | North America  | 186             | −1.84 (2.33)                      | −1.93 (0.48)                           |
| 1990–2015   | East Asia (China) | 3              | 1.15 (0.74)                       | 2.84 (0.23)                            |
| 1990–2015   | Europe         | 55              | −3.03 (0.93)                      | −3.74 (0.49)                           |
| 1990–2015   | North America  | 189             | −2.17 (0.65)                      | −2.41 (0.26)                           |
| 2000–2010   | Africa (Lamto) | 1               | 4.15                             | 2.69                                   |
| 2000–2010   | East Asia      | 30              | 0.49 (4.14)                       | 0.85 (2.01)                            |
| 2000–2010   | Europe         | 73              | −3.85 (2.83)                      | −4.36 (1.22)                           |
| 2000–2010   | India          | 10              | 2.27 (8.48)                       | 5.64 (2.21)                            |
| 2000–2010   | North America  | 226             | −2.30 (2.74)                      | −3.79 (0.68)                           |
| 2000–2015   | East Asia      | 30              | −0.98 (2.48)                      | 0.16 (1.33)                            |
| 2000–2015   | Europe         | 67              | −3.40 (1.37)                      | −3.94 (0.96)                           |
| 2000–2015   | North America  | 215             | −2.78 (2.02)                      | −3.75 (0.65)                           |

Table 3. Average absolute and per cent trends in SO2. Data in bold indicate trends not consistent with the observations within the standard deviation (STD).

| Time period | Region         | Nr. of stations | Average annual trend (STD) %/year | Absolute annual trend (STD) µgS/m²/year |
|-------------|----------------|-----------------|-----------------------------------|----------------------------------------|
| 1980–1990   | Europe         | 20              | −5.03 (2.04)                      | −0.211 (0.168)                         |
| 1990–2000   | Europe         | 43              | −7.56 (1.81)                      | −8.54 (1.40)                           |
| 1990–2000   | North America  | 53              | −3.27 (1.69)                      | −2.63 (0.30)                           |
| 1990–2015   | Europe         | 40              | −4.43 (0.88)                      | −4.63 (0.43)                           |
| 1990–2015   | North America  | 71              | −3.14 (0.75)                      | −2.83 (0.30)                           |
| 2000–2010   | Africa         | 8               | 17.6 (13.9)                       | 1.86 (1.19)                            |
| 2000–2010   | East Asia      | 19              | 5.84 (9.40)                       | 0.35 (2.22)                            |
| 2000–2010   | Europe         | 51              | −4.23 (3.17)                      | −5.31 (1.61)                           |
| 2000–2010   | North America  | 78              | −4.55 (1.68)                      | −4.44 (0.95)                           |
| 2000–2015   | Africa         | 8               | 4.97 (2.70)                       | 1.11 (0.63)                            |
| 2000–2015   | East Asia      | 19              | −0.14 (5.32)                      | −0.41 (9.2)                            |
| 2000–2015   | Europe         | 47              | −3.89 (2.16)                      | −4.86 (1.31)                           |
| 2000–2015   | North America  | 77              | −4.69 (1.35)                      | −4.40 (0.93)                           |

were available from sites in Africa, India, and additional Chinese sites (Table 2). For SO2, there was additional observational data from Africa (Table 3). Figure 2 shows the modeled global distribution of absolute and relative trends of concentrations and deposition. Comparing the maps with relative and absolute changes, the former visualize the trends better in areas where the concentrations or depositions are relatively low. This is especially visible in India where the relative changes are particularly large while the absolute changes are not, further note considerable positive trends from ship traffic, which has increased during the last decades4344.

Concentrations of the primary (directly) emitted compound, SO2, show greater decreases than the secondary (formed via atmospheric chemistry) sulfate in aerosols and in precipitation (Tables 1–3 and Figs S3–S5 in the SM). This non-linear relationship is seen in both model results and observations in North America and in Europe. This may partly be explained by the increasing oxidation capacity of the atmosphere during these twenty years4546. In the early 1990's, SO2 emissions were still high, and the oxidation of SO2 to sulfate was limited to some extent by the availability of the oxidants H2O2, OH and O3. As the emissions decreased, more oxidants became available, and SO2 was more efficiently oxidized to sulfate. Furthermore, a very important factor is that the decrease in SO2 emissions (and only slightly decreasing ammonia emissions) has led to less acidic cloud droplets, which increased
the oxidation rate of SO2 to sulfate via the ozone pathway\(^9,30,45\). In addition, less acidity in the environment probably leads to more efficient dry deposition of SO2\(^42,43,46\), which would also contribute to a larger reduction in SO2 concentrations with respect to sulfate. The trends in wet deposition of sulfur are lesser than the trends in SO2, but larger than those of sulfate in aerosols, because both SO2 and sulfate are efficiently scavenged by rain, and the wet deposition trend therefore represents a mix of SO2 and sulfate in aerosols.

The somewhat puzzling increase in the observed sulfate concentration in aerosol seen in East Asia for 2010 to 2015 is not observed for SO2 nor in sulfate in precipitation (Tables 1–3 and Figs S3–S5 in the SM). These show a steady decline from 2005–2015 and are more in line to the more recent inventories with a larger decrease in SO2 emissions since 2010 period than the emissions used in the model calculations.

Comparability between the global models. The spatial variability in the aerosol sulfate trends calculated by the individual models for the whole period 1990–2015 is shown in Fig. 3, including also the ensemble model mean and the standard deviation between the models. The general pattern is quite well represented by all the models. Quantitatively, the models show very similar trends in North America and Europe, while in East Asia the spatial differences are larger. Notably some of the largest differences are found over the Himalayas, which is also the boundary between areas with upward and downward trends. Some of the differences between the models might be explained by how fast the models oxidize SO2 and the lifetime of sulfate in aerosols\(^47\).

When comparing the average relative trends of sulfate based on the observations and the models for different periods in Fig. 4 (the statistical information is found in Table S1 and S2 in the SM), the different models give similar trends as the observations, but there are systematic differences between them. NorESM gives the lowest relative reductions, while Sprintars and ECHAM6 generally give the highest reductions. The differences between the models are larger for Europe compared to North America, maybe due to fewer sites in Europe representing the region. For the modeled average relative trends for the regions defining the largest emission areas in North

Figure 2. Absolute (left) and relative (right) trends of air concentrations and wet deposition calculated by the EMEP MSC-W model with observed trends superimposed (open circles), of SO2 (a) and sulfate in aerosol (b) and wet deposition of sulfate (c) over the 1990–2015 period.
America and Europe, the systematic differences in the relative trends between the models are smaller (Fig. S6 in SM). When comparing the average concentrations for the regions, there is a large spread between the models, but the differences depend on how the regions are defined since the models show somewhat different spatial patterns.

**Spatial representativeness of the regional trends.** The number of sites needed to quantify and validate emission changes depends on both the spatial and temporal variability of the trends. This is true for both models and observations, but the variability in the measurements is higher (see box plot in Fig. S5 in SM), thus more difficult to detect significant trends. For example, in Europe, the models seem to simulate somewhat higher trend from 2000 onwards compared to the observations (see Fig. 4, Tables 1–3), although the differences are within the standard deviation. These variations can partly be explained by some sites with quite low concentration levels and high relative changes (though not necessarily significant trends), influencing the average relative trends. The models compare well with the trends at those sites with large significant reductions, which can be seen for the trends for the 25th percentile of the sites with the highest rate, with a change of $-4.1\%/y$ and $-3.8\%/y$ for the period from 2000 to 2015 for the ensemble model mean and observations for sulfate in aerosols, respectively.

In East Asia there are positive and negative trends depending on periods and components, while in India and Africa there are positive trends, but the variability in the observations are high and there are relatively few sites for all these three regions. The sites do not necessarily represent the whole regions. In Africa, $SO_2$ increases at a rate of $17 \pm 14%/y$ from 2000 to 2010, but the increase occurred only in South Africa due to the influence of increased coal burning. At West- and Central African sites the concentrations are low and no significant trends are apparent, as reported earlier by Adon et al. In China, there are three sites with precipitation measurements from 1990 on but, except for one site, these do not show the increase indicated by the models and by the emission trends for the 1990–2000 period. However, by extending the period to 2005, the sites indicate an increase, illustrating the sensitivity of the results to the choice of time period. In India the sites are quite well spread over

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**Figure 3.** Relative trends in sulfate concentrations in aerosol for 1990–2015, calculated by the individual and ensemble models, with the observed trends superimposed (open circles). The differences between the models are shown in the inter-model variability map.
The good agreement also enhances confidence in the emission inventories in North America and in Europe, and, Still, the work here reveals consistent trends on regional and global scales between the two independent methods. The need for more sulfur observations in air, aerosols and precipitation to enable more homogeneous global coverage.

For normalized trends (Fig. S4 in SM), the trends in the bootstrap averages are consistent with the modeled trends in North America for all the species, while in Europe the modeled trends at the sites are larger than the bootstrap average. This suggests that the site density in North America is representative for trend analysis for the whole region while in Europe the number of sites is apparently too low for a representative estimate. The normalized trends in emissions are most consistent with the trends in sulfate in aerosols, indicating that this is a relevant parameter for compliance monitoring. In East Asia, the normalized modeled trends from 1990–2005 show smaller increase than the bootstrap average, especially for SO2, which indicates that the sites are not located where the largest changes in the region have taken place, nevertheless the observations show larger changes for SO2 and aerosol sulfate, though as discussed there are quite large variabilities and scattering in the time series.

A bootstrap approach has been used to assess how well the number of sites available represents each region. This is a possible method of assessing the representativeness of the trends, which, however, has its deficiencies. For example, changes in the absolute concentrations in North America, where most areas are remote, one is much more likely to pick a point in low rather than high concentration areas, while a large fraction of the observational sites are located closer to the higher emission and concentration regions. Thus the average observed concentrations are higher than in the bootstrap analysis results (Fig. S3 in SM). However, the standard deviation around the bootstrap mean is an indicator of the uncertainty caused by the geographical location of the given number of sites within each region. Figure 1 shows a high standard deviation of the sulfate concentrations in East Asia due to the relatively small number of sites and the inhomogeneous emission changes in this region.

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Conclusions
The results give confidence in the global aerosol models’ abilities to calculate historical sulfur trends and, thus, their scenario analyses of their future impact on climate and air quality. The fact that trends of emitted sulfur, sulfur in the aerosol phase after chemical transformation in the atmosphere, and sulfur in precipitation after wet removal agree between the observations and the model further implies that the relevant processes are realistically represented in the models.

However, one needs to be cautious when drawing conclusions from trends in regions with poor measurement site coverage, like India and China, and to some degree Africa, Australia and South America. There is a strong need for more sulfur observations in these regions to enable more homogeneous global coverage. Still, the work here reveals consistent trends on regional and global scales between the two independent methods. The good agreement also enhances confidence in the emission inventories in North America and in Europe, and, in turn, confirms that the concentration changes are attributable to SO2 emission changes. However, the bottom up emission inventory in East Asia used in this study has not captured the most likely large decrease in emissions in China after 2013. Further studies in this region require in depth comparison between updated emission inventories, atmospheric transport models, in situ- as well as satellite observations to better describe the significant changes in sulfur the last ten years for this region. The large ongoing increase in sulfur emissions in India also needs further investigations and should be more closely monitored.

This work has illustrated the strength of close co-operation between the regional networks to assemble a harmonized comparable global dataset. Future work on global trend analysis combining models and observations should include other species, in particular nitrogen compounds, organic and absorbing carbon are of high interest.

Figure 4. Comparison of the relative trends in sulfate concentrations in aerosol calculated by the different global models at sites with observed trends in the selected periods. The error bars indicate the standard deviation between the sites.
Methods

Global dataset of sulfur observations. Measurement data were collected from different regional and global networks, in total 365 sites. A map showing the sites is found in Supplementary Fig. S1. More information on the networks, methods and access to original data can be found in the references and links given in Table 4.

Precipitation and wet deposition sulfate measurements are mainly from wet-only samplers or bulk samplers if proven comparable to wet-only and from similar sites as was described elsewhere. The sulfate aerosol measurements are done using the aerosol filters, either PM$_{10}$ or no size cut off like the filterpack method, except for the IMPROVE network, which uses PM$_{2.5}$ filters. For SO$_2$, the filterpack method is commonly used in North America and Europe, while in Africa passive samplers dominate. In East Asia, both these methods in addition to continuous monitors are used. The sampling frequency is mostly daily and sometimes weekly, except for African data for which precipitation is sampled by event and SO$_2$ with monthly passive samplers. Wet deposition and volume-weighted precipitation data are based on the standard rain gauge depth if that is measured in parallel. At other sites without rain gauge, the sample precipitation amount is used. Monthly means are calculated for months with 70% or better data coverage. Urban sites are not included, and sites where the surroundings have changed considerably over the years have been excluded when known. The sites have been screened to be regionally representative with 70% or better data coverage. The sites have been screened to be regionally representative with 70% or better data coverage.

Global modelling and emission estimates. The global models involved in the present study are described in Table 5. The model setup to simulate the sulfur changes varies between the models; from any fixed meteorology, to that of one particular meteorological year, to just fixed sea surface temperatures. All models simulated and diagnosed sulfate in aerosol, while only one model (EMEP/MSC-W) simulated SO$_2$ and wet deposition of sulfate for this study. In addition to sulfate, all the models simulated black carbon (BC) and primary organic aerosols (POA), and several also included secondary organic aerosols, nitrate, sea salt and dust. Details are found in Myhre et al. and in the references in Table 5. All models use identical anthropogenic emission data from the EU project ECLIPSE for the 1990 to 2015 period; several updates and improvements compared to earlier emission data sets were included in this inventory. A direct link to the respective dataset is found here: http://www.iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv5a.html.

Natural emissions from i.e. volcanos are included but do vary somewhat between the models. See the references in Table 5 for details.

Statistical calculations. For each of the periods considered, the sites were selected based on the criteria that number of available years should be at least 75% of the total number of years in the period. The statistics are done from both yearly and seasonal averages, though only yearly are included. At least 10 measurements are required per season for being used in the statistics. The yearly averages are computed from the seasonally averages if the four seasonal averages are available (with more than 10 measurements) per year. Data from the same sites and periods have been extracted from the model output and trends have been calculated in similar way. Note that for most of the models (except NorESM and ECHAM6), there are only modelled data for each 5 year period. For the statistical analysis, the non-parametric Mann–Kendall test has been used on annual means for detecting and estimating trends; this is a common method when missing values occur and when data are not normally distributed. In parallel, the Theil–Sen’s slope estimator has been used to quantify the magnitude of potential trends. A 90% confidence interval (p-value below 0.10) was chosen as the criterion for determining the significance of the trend. The Scientific Library for Python (scipy-0.14.0) has been used for calculating the trends. The average percent changes are calculated for all the sites, not only for those with a significant trend. The robustness in the trends in modelled data for shorter periods (10–15 years) is hampered by having only few data point, i.e. 3 points for a ten year period. The Mann Kendall test allows for calculation even down to 3 points, but with pval < 0.1 it is necessary with 4 points of more to estimate significance or not.

Table 4. List of networks contributing with sites and data. The original data can be accessed from the given web pages.
Table 5. Model descriptions.

| Models and references | Resolution | Fixed-met or fixed-SST |
|-----------------------|------------|------------------------|
| ECHAM6-HAM2

 85 | T63 1.8° × 1.8° L31 | Climatological monthly varying fixed-SST and sea ice extent averaged for the period 1979 to 2008. |
| EMEP/MSC-W

 94 | 0.5° × 0.5° L20 | 2010 meteorology, 3 hourly ECMWF based |
| GISS

 66 | 2.0° × 2.5° L40 | 2000 climatological monthly varying fixed-SSTs and sea-ice |
| NorESM1

 59 | 1.9° × 2.5° L26 | Climatological monthly varying fixed SSTs and sea ice extent over the 1990–2013 period |
| OsloCTM2

 84 | T42 2.8° × 2.8° L60 | 2010 meteorology, 3 hourly ECMWF based |
| SPRINTARS

 65 | 1.125° × 1.125° L56 | Climatological monthly varying fixed SSTs and sea ice extent over the 1988–1992 period |

Data Availability

The aggregated observational global data set with monthly mean concentrations and total wet deposition values, together with the model results at the same sites have, been associated with a specific DOI and can be downloaded from https://doi.org/10.21336/gen.2. The statistical results from all the individual time series are also available. A web tool, http://aerocom.met.no/trends/S-trends/ maps the trends in SO2 and sulfate in aerosol and wet deposition.

References

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The document contains a list of authors and references. It appears to be a scientific reference list, typically found at the end of a scientific paper. The references are cited in the text, indicating that the information is sourced from these works. The references span a variety of topics within atmospheric science and environmental studies, including aerosol-climate interactions, model evaluations, and emissions trends.
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Author Contributions
W.A. has coordinated the study and prepared the observational data, while A.M. has been responsible for the statistical calculations, mapping and developing the web interface. G.M., S.T., H.F., M.S. and C.L.M. have been part of the core writing team. W.A., V.B., J.H., C.G.-L. P.S.P.R., A.S., R.V., C.M.N.L., K.S. and X.X. have provided data from the various networks, Z.K. has provided emission data and G.M., S.T., R.C., G.F., H.F., D.O., J.Q., D.S., R.B.S. and T.T. have provided model output.

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Supplement for:

Global and regional trends of atmospheric sulfur

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Sites used
The site selection is largely based on trend analyses done in regional assessments. For EMEP, the same sites were used in Tørseth et al.\textsuperscript{1,2} and Collette et al.\textsuperscript{1,2}, as well as the IPCC AR5\textsuperscript{3}, though we added the years after 2012. For EANET, NADP, CAPMoN the same precipitation sites are used as in Vet et al.\textsuperscript{4} with an extended time period. The Indian precipitation data used have recently been presented with details on sampling procedures and site descriptions\textsuperscript{5}. The IMPROVE data are similar as presented by Hand et al.\textsuperscript{6} though extended to 2015.

![Map of sites](image)

**Figure S1.** *Location of the sites and measurements used.*

There has been some minor manual selection of sites and correction in the data used in some of the Figures and in the analysis:

- For the 1990-2015 trends (Figures 1, S3 and S4), the 2000 data for many EANET sites have been estimated by using 2001 for several sites which started their program then.
- Some European sites, which lacked measurements for 2015, data for 2014 were used for the 1990-2015 trends (Figures 1, S3 and S4).
- Three French sites were deleted in the statistical calculation for 2000-2010 because they ended in 2007, which in addition showed suspiciously high concentrations affecting the average trend for this period.
Figure S2. Number of sites with significant and insignificant trends, observed and modeled (EMEP/MSC-W) for different time periods. NA=North America, EA=East Asia, IN=India, AF=Africa. Note that for the ten year periods the model only have three points (five year interval) and cannot detect significance or not.
Figure S3. Trends in SO$_2$, sulfate in aerosols and in wet deposition in East Asia, North America and Europe from observations and the EMEP/MSC-W model (and Ensemble model for sulfate in aerosols) at the selected number of sites, and the average bootstrap trends and standard deviations from 1000 iterations. Observed and modelled concentrations are given on the y-axis on the left, while emissions on the right. For East Asia an alternative emission development the last ten years are included, based on more recent inventories$^{7,8}$. 

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**East Asia**
- 13 sites
- SO$_2$
- Sulfate in aerosols
- Sulfate wet deposition

**North America**
- 42 sites
- SO$_2$
- Sulfate in aerosols
- Sulfate wet deposition

**Europe**
- 25 sites
- SO$_2$
- Sulfate in aerosols
- Sulfate wet deposition
The normalised trends in Figure S4 below have been calculated by setting the 1990 concentration to 0.0 and then adding the relative change for each five year interval. For East Asia the normalised trends were calculated from 2000 for the observations but from 1990 for the emissions and model results. The standard deviation of the bootstrap averages was calculated using the ratio between the bootstrap concentration and standard deviation for the different years.

**Figure S4.** Normalised trends of SO$_2$, sulfate in aerosols and in wet deposition in East Asia, North America and Europe in observation and EMEP/MSC-W model (and Ensemble model mean for sulfate in aerosols) at the selected number of sites, and the average bootstrap trends and standard deviations from 1000 iterations. For East Asia an alternative emission development the last ten years are included, based on more recent inventories.$^7,8$. 
**Figure S5.** Box plot indicating 10, 25, 50, 75 and 90 percentiles of the seasonal and annual observed and modelled trends (EMEP/MSC-W model for SO\textsubscript{2} and wet deposition of sulfate; ensemble models for sulfate in aerosols) for different time periods.
Figure S6. Comparison of the average concentrations and total relative change of modelled sulfate concentrations in aerosols for the period 1990-2015, calculated by the different global models for the different (sub)regions indicated in the bottom right map.
### Table S1. Trends in observed and modelled SO$_2^-$ in aerosols, % change per year

| Time period | Region       | Number of stations | Obs.     | ECHAM6       | EMEP/MSC-W | GISS       | NorESM     | OsloCTM2    | SPRINTARS   |
|-------------|--------------|--------------------|----------|--------------|------------|------------|------------|-------------|-------------|
| 1980-1990   | Europe       | 16                 | -2.56 (3.10) | -5.80 (1.26) | -5.35 (1.20) | -3.68 (1.63) | -5.02 (0.94) | -5.76 (1.17) |
| 1990-2000   | Europe       | 41                 | -5.16 (2.11) | -5.80 (0.82) | -5.35 (1.20) | -3.68 (1.63) | -5.02 (0.94) | -5.76 (1.17) |
| 1990-2000   | North America| 101                | -2.08 (1.44) | -2.52 (0.62) | -1.91 (0.30) | -1.92 (0.32) | -1.62 (0.70) | -1.71 (0.32) | -1.95 (0.30) |
| 1990-2015   | Europe       | 33                 | -2.93 (0.69) | -3.52 (0.36) | -3.22 (0.37) | -2.96 (0.55) | -2.47 (0.60) | -2.95 (0.45) | -3.45 (0.53) |
| 1990-2015   | North America| 124                | -2.14 (0.67) | -2.40 (0.26) | -2.23 (0.22) | -2.19 (0.36) | -1.95 (0.31) | -2.06 (0.37) | -2.32 (0.23) |
| 2000-2010   | East Asia    | 13                 | 0.44 (4.25)  | 0.50 (1.43) | 0.40 (1.01)  | 0.63 (0.91) | 0.20 (1.16) | 0.34 (1.04) | 0.44 (1.35) |
| 2000-2010   | Europe       | 43                 | -2.86 (2.20) | -4.34 (1.06) | -3.64 (1.35) | -3.25 (0.96) | -2.69 (1.05) | -3.29 (0.86) | -3.96 (1.07) |
| 2000-2010   | North America| 227                | -3.03 (1.72) | -3.70 (0.61) | -3.29 (0.72) | -3.16 (0.83) | -2.75 (0.97) | -2.97 (0.86) | -3.46 (0.63) |
| 2000-2015   | East Asia    | 13                 | 2.68 (9.41)  | -0.09 (0.86) | 0.02 (0.82)  | 0.11 (0.89) | 0.08 (0.97) | 0.02 (0.91) | -0.02 (0.99) |
| 2000-2015   | Europe       | 36                 | -2.67 (2.03) | -3.58 (1.01) | -3.25 (1.16) | -3.01 (0.79) | -2.79 (0.66) | -3.06 (0.71) | -3.85 (0.79) |
| 2000-2015   | North America| 218                | -3.15 (1.30) | -3.43 (0.60) | -3.26 (0.60) | -3.09 (0.79) | -2.86 (0.72) | -2.95 (0.75) | -3.47 (0.52) |

### Table S2. Trends in observed and modelled SO$_2^-$ in aerosols, change in μgS/m3 per year

| Time period | Region       | Number of stations | Obs.     | ECHAM6       | EMEP/MSC-W | GISS       | NorESM       | OsloCTM2     | SPRINTARS |
|-------------|--------------|--------------------|----------|--------------|------------|------------|--------------|--------------|-----------|
| 1980-1990   | Europe       | 16                 | -0.048 (0.094) | -0.082 (0.056) | -0.082 (0.055) | -0.051 (0.037) | -0.036 (0.028) | -0.034 (0.021) | -0.135 (0.110) |
| 1990-2000   | Europe       | 41                 | -0.073 (0.052) | -0.028 (0.014) | -0.020 (0.013) | -0.017 (0.011) | -0.021 (0.014) | -0.011 (0.009) | -0.029 (0.022) |
| 1990-2000   | North America| 101                | -0.024 (0.025) | -0.042 (0.027) | -0.038 (0.025) | -0.026 (0.018) | -0.022 (0.013) | -0.017 (0.011) | -0.068 (0.046) |
| 1990-2015   | Europe       | 33                 | -0.031 (0.015) | -0.028 (0.016) | -0.022 (0.017) | -0.020 (0.015) | -0.027 (0.019) | -0.015 (0.011) | -0.033 (0.028) |
| 1990-2015   | North America| 124                | -0.026 (0.024) | -0.028 (0.016) | -0.022 (0.017) | -0.020 (0.015) | -0.027 (0.019) | -0.015 (0.011) | -0.033 (0.028) |
| 2000-2010   | East Asia    | 13                 | 0.003 (0.034) | 0.007 (0.018) | 0.005 (0.011) | 0.008 (0.008) | 0.005 (0.020) | 0.003 (0.006) | 0.005 (0.017) |
| 2000-2010   | Europe       | 43                 | 0.003 (0.034) | 0.007 (0.018) | 0.005 (0.011) | 0.008 (0.008) | 0.005 (0.020) | 0.003 (0.006) | 0.005 (0.017) |
| 2000-2010   | North America| 227                | -0.029 (0.029) | -0.032 (0.018) | -0.025 (0.020) | -0.022 (0.018) | -0.028 (0.020) | -0.016 (0.013) | -0.038 (0.033) |
| 2000-2015   | East Asia    | 13                 | 0.003 (0.037) | 0.000 (0.010) | 0.001 (0.006) | 0.002 (0.006) | 0.003 (0.012) | 0.002 (0.005) | -0.001 (0.015) |
| 2000-2015   | Europe       | 36                 | 0.003 (0.037) | 0.000 (0.010) | 0.001 (0.006) | 0.002 (0.006) | 0.003 (0.012) | 0.002 (0.005) | -0.001 (0.015) |
| 2000-2015   | North America| 218                | 0.003 (0.037) | 0.000 (0.010) | 0.001 (0.006) | 0.002 (0.006) | 0.003 (0.012) | 0.002 (0.005) | -0.001 (0.015) |
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