

Response to comments for Hand et al.

We appreciate the Referee's time and comments; they serve to improve the paper. Our responses follow comments below.

Referee #1

The authors analyze trends in particulate sulfate concentrations measured in the United States over a twenty-year time period, 1990-2010. The authors also present SO₂ emission trends from electricity generating units over the same time period, and examine relationships between changes in SO₂ emissions and changes in air quality. Relative to an earlier paper by Malm et al. (JGR 2002), this paper covers a longer time period, and takes advantage of a large increase in the number of both urban (CSN) and rural (IMPROVE) measurement sites since 2000.

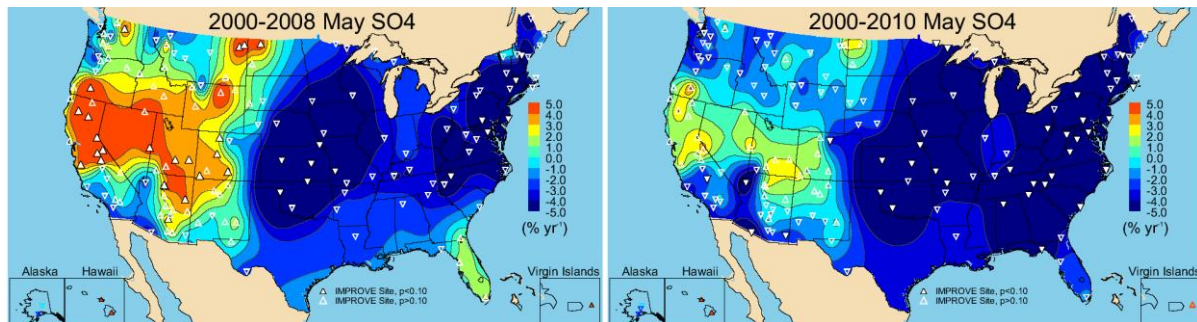
Major Comments

1. On page 19313, the authors misconstrue the paper by Pope and Dockery, which C5886 raises concerns about health effects of fine particulate matter in general, not sulfate specifically. It could be that water-soluble particles such as nitrate and sulfate are not the toxic agents. The statements about health effects of sulfate particles are not adequately supported. Figuring out which components of PM_{2.5} are truly harmful to health is a very important but unresolved research question.

We appreciate the referee's point regarding the uncertainty surrounding the health effects of sulfate. While sulfate is a major contributor to PM_{2.5}, it may not be associated with the most harmful components. A recent review paper by Rohr and Wyzga (2012, Atmos Environ) reports on several studies where sulfate may be implicated. We will cite this paper rather than Pope and Dockery as it does provide some evidence for the role of sulfate in health effects.

2. The monthly analysis of ambient concentration trends shown in Figures 2, 3, 7, and 8 is weakly supported by the underlying dataset, and appears susceptible to spurious findings and poor statistical power in detecting trends. The IMPROVE and CSN measurements are made at best on a once every third day schedule. Therefore subdividing the dataset into monthly averages means at most 10 samples per year at each site. Missing or invalid data will decrease the number of samples available to even lower numbers. These problems are clearly apparent for example in the large and meaningless oscillations that appear in Figures 7-8. I recommend analysis and reporting of quarterly average (DJF, MAM, JJA, SON) concentrations rather than monthly averages, with corresponding revisions to Figure 2. Much of the discussion on pages 19321-22 is already written in terms of quarterly rather than monthly averages. It would be clearer if Figure 2 was constructed analogously. Also Figures 3, 7, and 8 seem to show minor curiosities in the data and distract attention from other more important findings. I recommend omitting them.

We will substitute trends of seasonal mean concentrations for trends in monthly mean concentrations in Figure 2. However, in doing so some information may be lost because there is clearly different monthly mean behavior at many sites. In fact, much of the poor statistical power for May monthly mean trends is due not to sampling issues but to behavior in the timelines. Concentrations fell rather dramatically in 2009 and 2010 at many sites. Below are maps of trends in May monthly mean concentrations for only IMPROVE sites from (a) 2000-2008 compared to (b) 2000-2010. The statistical power for many trends in the West decreased significantly when including the last two years of data.



The timelines we showed in Figure 3 demonstrated the drop in concentrations at both an urban and rural sites in California. However, we will omit Figure 3 based on the referee's comments and because these data are included in the regional trend in Figure 7. We argue to include Figures 7 and 8 because they demonstrate important findings. These timelines show the divergence of concentrations and emissions in the West and the central Great Plains, implying power plant SO₂ emissions cannot account for the behavior observed in the concentrations. We do not expect these data to suffer from sampling issues because in the western region, data from 15 to nearly 40 aggregated sites were used, and in the Great Plains, 3 to roughly 20 sites were used, depending on network and time period. The tracking of urban and rural concentrations in both of these figures suggests the variability is real.

3. Figure 1 is important and interesting, but it conveys only relative changes over time, and this does not convey important spatial differences in absolute concentrations. I suggest showing national maps for a couple of years (maybe 1990, 2000, and 2010?) with absolute sulfate concentrations in ug/m³, as well as the current Figure with the percentage changes. Figure 6 shows the absolute concentrations, but the y-axis scales vary and it is hard to see the national picture from this figure.

We will include two maps of annual mean sulfate concentrations from 2000-2002 and 2008-2010.

Minor Comments

1. Check very low reported values on page 19323: only 12.3 and 3 grams per year of SO₂ emissions. There may be a sign or other error in the exponents. Figure 4 suggests SO₂ emissions in Idaho (one of the numbers cited above) are much higher than stated in the text.

Yes, thank you for catching our typographical error in the units. The Idaho and Rhode Island emissions have been corrected (3.04 t yr^{-1} and 1.23 t yr^{-1} , respectively). Figure 4 is replotted using a log scale to demonstrate this range more clearly.

2. Page 19323, line 27, less that at other regions should be in other regions

Changed.

3. Page 19330, line 14, given should be some

Changed.