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## ***Interactive comment on* “Particulate sulfate ion concentration and SO<sub>2</sub> emission trends in the United States from the early 1990s through 2010” by J. L. Hand et al.**

### **Anonymous Referee #1**

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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 SO<sub>2</sub> emission trends from electricity generating units over the same time period, and examine relationships between changes in SO<sub>2</sub> 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

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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<sub>2.5</sub> are truly harmful to health is a very important but unresolved research question.

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.

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  $\mu\text{g}/\text{m}^3$ , 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.

## Minor Comments

1. Check very low reported values on page 19323: only 12.3 and 3 grams per year

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of SO<sub>2</sub> emissions. There may be a sign or other error in the exponents. Figure 4 suggests SO<sub>2</sub> emissions in Idaho (one of the numbers cited above) are much higher than stated in the text.

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

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

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 19311, 2012.

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