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 #2

This paper is a thorough analysis of trends in sulfate and SO2 power generation emissions in the United States over the last decade(s), providing useful information on the link between decreasing emissions and the response in observed sulfate levels. While this is a fine analysis, I found the paper somewhat lacking in insight into the origins of the observed behavior. I've detailed this a bit more below with some specific examples.

Major Comments:
1. Often the trend for a specific site or region is given with little discussion of the potential causes. While it may not always be clear to the authors, some discussion of possible causes would provide some useful context to the results. Examples as follows:

We very much appreciate the questions raised by the referee regarding the causes of these trends. The intent of this paper is to evaluate changes in sulfate concentrations in urban and rural sites across the country and to investigate whether the changes in power plant SO2 emissions could account for the observed behavior in sulfate concentrations. In doing so we uncovered important regions and seasons where the two diverge and point these out to the community as important considerations. In our earlier paper (Hand et al., 2012b, Atmos Environ), we pointed out winter trends in sulfate and nitrate, and performed some analyses using back trajectories to investigate causes. These investigations were inconclusive and suggest additional tools may be necessary to tease out the causes. Therefore, in this work, we speculate as to possible causes, such as transpacific transport for the spring trends, and perhaps contributions from oil and gas development or Canadian transport influences on winter trends. To definitively state the causes will require work beyond the scope of this paper.

a. Page 19320, lines 4-6: Why is sulfate increasing at these 3 sites? How does this correspond with the local trend in emissions?

The trends at Hawaii and Alaska are very interesting. We do not know why the concentrations are increasing. Fort Peck, MT, is a site that is experiencing significant increased concentrations in December (nearly 18% yr^{-1}) and analysis in our previous paper on winter trends suggested it may be influenced by transport from oil sands in Canada. However, it is also in a region that may be influenced by local oil and gas development as well.

b. Page 19321: Discussion of Dec trends in N/Central Great Plains: Why are such large increases apparent?

This is a very interesting region and we do not fully understand what is happening there.

c. Page 19321: Discussion of Dec trends in central-eastern US: again, why do these show winter time increases?

See previous comments.

d. Page 19321/19322: Springtime trends possible associated with transpacific transport? Does this affect the California sites?

Possibly. We speculate as to this cause, especially given the trends in Chinese emissions, however we cannot conclude this decisively from our data. Other tools, such as global modeling, will be helpful.

e. Page 19325, line 14-15: So if not urban emissions, what is the cause of this difference?

The difference in the scales largely reflects geographical differences. Sulfate is highest in the East where the majority of the CSN sites are located. IMPROVE site density is highest in the West where concentrations are lower. Urban excess estimates based on annual mean data from 2005-2008 for nearby urban and rural sites suggest that the difference in annual mean ammonium sulfate was 1.3 ± 0.9 μg m^{-3} (Hand et al., 2011).

f. Page 19325-19326: There is an increase in sulfate concentrations in 2005 across the entire US which is inconsistent with the emissions. What is the possible cause of this? What about the similar, although more modest, enhancement in 2007?

Good questions and not understood.

g. Page 19327, lines 14-15: Why? (see #4 above)
h. Page 19327, lines 15-22: Why? (see #2 above)

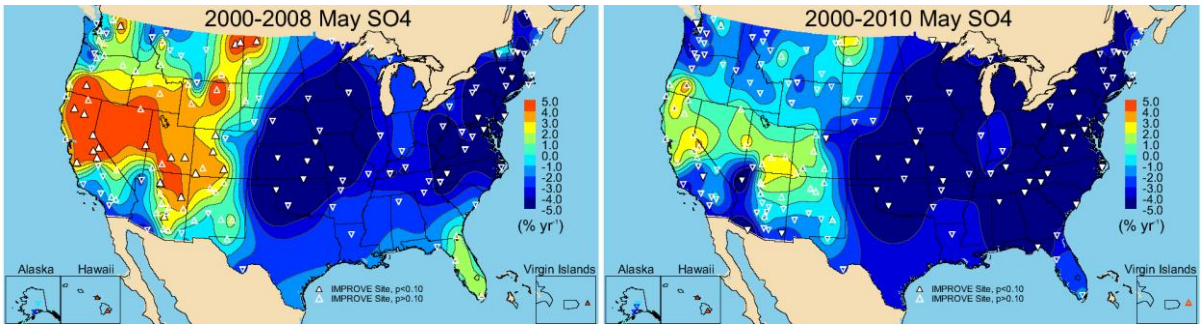
Again, good questions and not understood.

i. Page 19328, lines 7-12: This is the first discussion of transpacific transport, but you mention long range transport as a factor in sulfate concentrations trends in the Introduction, so this is on the reader's mind as they go through Section 3 (see #4 and #7 above). Why wait to say this?
j. Page 19329, lines 2-5: ditto.

We did mention transpacific transport and possible causes of winter trends in the Introduction (pg 19315, line 16, 21). However, the organization of the paper was such that discussions and speculations came at the end, and because at this point it is just speculation, we did not develop it more thoroughly.

2. Are the monthly trends shown in Figures 2 statistically significant (given few samples each month)? Is the lack of data for each year a potential cause for some of the spurious looking "hotspots"? Perhaps it would be better to look at seasonal trends?

We will substitute trends in 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.



Minor Comments:

1. Abstract, line 11-12: The text is ambiguous as to whether the 2 different trends given are for urban vs. rural or simply the two together over two different time horizons. Clarify text.

Fixed.

2. Abstract: It would read better if trends in emissions and concentrations were given for the same time horizon (so as to be directly compared) in the abstract. The differences in record lengths is discussed in the manuscript, so perhaps these summary trend numbers should all be provided for 2001-2010.

We will report the IMPROVE and SO_2 emission trends for the entire US for 2001-2010. However, when computing trends for aggregated sites, not individual sites, we found that using aggregated CSN data for the total US in 2001 actually biased the trends because few sites were on line in 2001 and those that were operating were predominantly in the West. In 2002 many more eastern sites came on line and the geographical distribution of those sites were more representative of CSN than the western sites. Therefore, unfortunately, we can only report CSN aggregated US trends from 2002-2010.

3. Page 19313, line 3: 40-60% is a narrow range, is this for the annual mean contribution? Clarify in the text.

If we look only at IMPROVE sites, than 40-60% is accurate for eastern regions (Hand et al, 2012 JGR). Including CSN sites widens the range to 30-60% due to the role of nitrates in winter. We will include the wider range in the paper.

4. Page 19313, line 8: why has the NW max shifted to the spring?

We believe the shift has occurred due to the increase in sulfate concentrations in the spring, however we do not know the cause of the increase.

5. Page 19314, line 17: What does "decreased concentrations were largest" mean? It implies that the concentrations were "largest" when I think the authors mean that the decrease was largest.

Fixed in the text.

6. Page 19317, line 10: What does "(3*sulfur)" mean? Please expand or remove text.

3*Sulfur converts sulfur mass to sulfate mass based on their molecular weights (32.06/(32.06+4*16). We clarified this in the text.

7. Page 19319, line 6-7: Was the increasing trend one of the significant sites?

No, the increased trend at Big Bend, TX was not significant.

8. Page 19325, lines 19-20: This last sentence should be given in the figure caption.

Agreed.

9. Page 19329, lines 6-15: This discussion would be more appropriate in Section 3.4 when discussing the correspondence of the emissions & concentrations. Also, when reading 3.4 I was thinking that looking at the correlation coefficients for each region by season would be a good summary of when concentration trends are predominantly related to emission trends or not.

We have moved this paragraph to the end of Section 3.4. We will report the correlation coefficients for the two time periods we discussed.