

Interactive comment on “Aerosol and precipitation chemistry in the southwestern United States: spatiotemporal trends and interrelationships” by A. Sorooshian et al.

Anonymous Referee #2

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This paper gives is well written, and it gives a nice overview of the aerosol and precipitation chemistry in southwestern US. I find it quite interesting, even though it is somewhat speculative, that you maybe can distinguish the difference in sulfate and dust when it comes to their potential as cloud condensation nuclei versus ice nuclei. The conclusion is not surprising as such since it is well known that sulfate act as CCN and that more crystalline particles act as IN, but that you may interpret this from the statistics in the monitoring data is quite neat.

Specific comments to the text:

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Figure 5 and the text in general: Why is not ammonium included in the IMPROVE data? Is ammonium only measured at selected IMPROVE sites? It is certainly of interest to know the relative contribution of ammonium. The properties of the aerosols are dependent on whether all the ammonium which is available is bound to sulfate or if it is sufficient ammonium to make ammonium nitrate. Ammonium nitrate is not stable and you can have biased nitrate measurements due to evaporation from the filters. From the precipitation data it seems like it is relatively small contribution of ammonium in the region. Would be nice to have some more discussion on this issue.

Page 8624 line 18-22. An additional explanation of higher particulate nitrate concentration in winter is that particulate nitrate winter due to more stable particulate face. NH_4NO_3 dissociate to NH_3 and HNO_3 when warm conditions and it is quite common to measure lower concentration in summer especially in warm areas.

It would have been nice to include some discussion of the potential artifact problem. It is well know that nitrate (and ammonium) aerosols measurements can be biased due to the gas/particle exchange. However, IMPROVE use a denuder system and may be that the method gives good/accurate measure of nitrate. Are there any studies which has looked at the comparability of the nitrate measurements from IMPROVE with other methods?

Page 8626, line 4-8. There are high pHs other places than those mentioned here. Typically in areas in where the it is low emission of SO_2 and NO_x and/or high emissions of dust. Typically in the northern Mediterranean, central Africa, south Asia, Mongolia. You may refer to the regional networks like EANET, IDAF and EMEP. As well as the NADP and CAPMoN of course. You also have regions in US and Canada with lower pH. The publication on Asian data are quite old, and the pH may have changed quite a lot since the nineties due to more sulfuric acid. You may rather to EANET, see data report here: <http://www.eanet.asia/product/index.html> For European data, you may look at the EMEP database: <http://ebas.nilu.no/> African, IDAF data is given in various publications, I, .:

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Galy-Lacaux, C., Laouali, D., Descroix, L., Gobron, N., Liousse, C., 2009. Long term precipitation chemistry and wet deposition in a remote dry savanna site in Africa (Niger). *Atmospheric Chemistry and Physics* 9, 1579-1595

Line 8627 line 17-20. Here again I find the selection of studies used for comparison a bit strange and arbitrary selection. China and Tibet has certainly different source influence than US. Why not just look at US, at least compare to other North American studies in addition. Globally you have a wide range of various relative distributions of ions depending on the main sources in the regions and meteorology.

Page 8629, line 13. "...dust is the dominant source of SO₄" Source? The fact that SO₄ comes together with mineral dust is not the same as saying SO₄ is from dust. In large part of Asia the main source of sulfate is anthropogenic. Also in the paper by Zhao et al 2011 it is stated that SO₄ is at least the latter decades mainly is from anthropogenic sources, i.e. sulfuric acid which attach to mineral dust particles.

Page 8630, line 2: "As SO₄ and fine soil represent the most abundant PM_{2.5} constituents". Well, only if you neglect the carbonaceous fraction. . . Page 8632, line 12. The reference to Spain is an old study, maybe use an updated reference from the same group:

Izquierdo, R., Avila, A., Alarcón, M. (2012) Trajectory statistical analysis of atmospheric transport patterns and trends in precipitation chemistry of a rural site in NE Spain in 1984-2009. *Atmospheric Environment* 61, pp. 400-408

At this site as well as many places in Europe, nitrate is getting relatively more important (even though the absolute concentration do decrease many places) to sulfur. At this Spanish site the NO₃:SO₄ is 1:1 the latter period. Also here it would be nice to include more North American (NADP+ CAPMoN) studies, and not somewhat arbitrarily global studies. The regional differences in NA are also large.

Page 8635, line 12. Include reference to NTN/NADP trends done by others, i.e:

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Christopher M. B. Lehmann and David A. Gay (2011) Monitoring Long-Term Trends of Acidic Wet Deposition in US Precipitation. *PowerPlant Chemistry* 2011, 13 (7). 2011 Her it seems like nitrate actually has a had significant increase in the South-Midwest

Page 8637, line 8. "Other regions" is here very undefined and too general. Surely there are other regions with lower nitrate to sulfate that observed in this study, especially in Asia where sulfate is dominating. But you have many examples of the opposite. Several places in Europe and Africa nitrate is the dominating anion

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