Atmos. Chem. Phys. Discuss., 14, C10621–C10624, 2014
www.atmos-chem-phys-discuss.net/14/C10621/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Particle water and pH in the southeastern United States" *by* H. Guo et al.

Anonymous Referee #1

Received and published: 31 December 2014

The investigators present results from modeling and field observations in southeastern USA and focus on particle liquid water content (LWC) and pH. They bring together a lot of tools and go into length about uncertainties. Selected final results include: (i) particle water and pH are closely related; (ii) particle water is impacted by water uptake of ionic and organic species; (iii) particle ion balances don't necessarily agree with particle pH; (iv) and a number of specific values of pH and LWC are reported for the Southeast including diurnal trends and the point is made that pH levels are low and this can impact a number of relevant physical processes. The topic of the paper is of interest to the journal and the paper is organized and written well. The title and abstract adequately reflect the contents of the manuscript.

Major Comments:

This reviewer has a number of comments ranging from minor to major. One such issue

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Discussion Paper

that puzzles this reviewer is that another paper was just published in ACP ("Trends in particle-phase liquid water during the Southern Oxidant and Aerosol Study") discussing the same general topic, region, and campaign (SOAS) with figures that at times are showing similar things as in this paper. Based on the current form of this paper in review, I did not get a sense that it was sufficiently different to be comfortable. The methods will differ, but the general scientific nuggets presented after going through the details of the methods and uncertainties are aligned and at times redundant. For example, compare the current Figure 7 (for this paper in review) with their Figures 6-7. At times during my reading I felt that this paper was better suited for a methods-oriented venue such as AMTD. With all this being said, with significant revisions the authors need to make a much stronger effort to articulate and show how this manuscript is different than the other one and to make a case for why the science presented is of sufficient impact beyond those of the other paper. Together with this past comment, the authors should consider strongly that the current paper in review is quite heavy in discussion of methods and uncertainties and it is hard to extract the key scientific advancements due partly to the great effort required to digest and comprehend all of the content up until Section 4.4.

One thing that was challenging upon reading the manuscript was how to keep track of all the uncertainties and assumptions that were being made. By the time I reached Sections 4.4-4.6, I lost track about how much the numbers would be affected by these issues, some of which were discussed, but some of which were not. Several assumptions required more discussion such as one discussed below about the nephelometer data. More details are needed about the different sizes examined by the various instruments, potential volatilization issues in the lines of some instruments, and how size distributions varied during the sampling which can affect the validity of some of the methods employed.

What are the broader implications of this study to other regions and studies since it seems to be highly specific to the instrumentation/methods and sites examined by the

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investigators?

Specific Comments:

Section 2.2: can the authors clarify more about whether they measured PM2.5 or PM1 and what the sampling strategy was. This seems like a key point since PILS is being compared to the AMS. Clarity is needed throughout the paper about the different size resolutions being compared between instruments and how differences impact results.

AMS description: Clarify assumptions about the collection efficiency and how results can be impacted by the uncertainty in this value.

Page 27153: write out what TEOM is.

Figure S4: I would recommend starting the y-axis at a more reasonable lower limit than zero to show the variations better in the plotted points.

Supplement Section 3 about nephelometers and in Section 2.3: A major assumption that seems highly problematic is that Q_wet and Q-dry are equivalent. Just looking at Figure S3 shows that some significant changes can be expected in Q based on particle size. For example, if a dry particle at 250 nm grows in a humid environment to 450 nm, the ratio of the Q values will be much larger than 1. Couldn't refractive index change?, and would this affect this Q_wet=Q_dry assumption? Careful discussion/analysis is needed here since this was not given adequate attention based on my reading.

Same section: Since measurements are done for ambient RH, how would the RH change in the sampling lines up to the point of measurement? Also, how would the presence of coarse aerosol affect the nephelometer measurements, and overall study results, and what length did the authors go through the remove cases of coarse aerosol in their study using size distribution data? Presumably in the region there may be sources of such larger particles. Discussion/analysis to address these points is needed.

The authors should discuss (via analysis) if volatilization of vulnerable species (e.g.

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ammonium) in instrumentation (including sampling lines) affects the results in way and how this was accounted for.

Pg 27159, Line 22: change “resulted” to “resulting”

The authors focus quite a lot on sulfate and ammonium in terms of PILS species in their calculation of pH uncertainty, but what about other species that the PILS is capable of examining?

Figure 3-5: A general comment that these figures may confuse other readers unless the captions become more descriptive of what is being shown.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 27143, 2014.