

Interactive comment on “Measurement of ambient aerosol hydration state at Great Smoky Mountains National Park in the Southeastern United States” by N. F. Taylor et al.

Anonymous Referee #1

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The paper presents results of two deployments of a tandem differential mobility analyzer configuration capable of size-resolved detection of hysteresis in hydration behavior and determination of ambient hydration state (AS-TDMA). The AS-TDMA was installed at a site in eastern Tennessee on the border of Great Smoky Mountains National Park during the summer of 2006 and winter of 2007–2008. During the summer, 12% of ambient aerosol particles displayed hysteresis and were found in a more hydrated state. None existed in a less hydrated state. Winter measurements indicated 32% and 17% of aerosol particles in more and less hydrated states, respectively.

To a large degree, the manuscript is well written and the detailed description of DF

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and EF cycles is helpful. It seems, however, that the characterization of more and less hydrated states is somewhat confusing. In some of the discussion there seems to be an implication that the particles passing through the AS-TDMA are of one type of particle as opposed to an ensemble of particles, some that do not show hysteretic behavior, some that at some time in their past history have deliquesced, and some that have not. For instance, see on page 9, line 289, the sentence “. . .that the particle is hysteretic and. . .” The AS-TDMA does not measure particle characteristics but rather characteristics of ensembles of particles with similar characteristics. This may seem minor but is confusing to the reader when trying to understand how the instrument was ultimately used under ambient conditions. In that context, I think Figure 2 and the associated discussion could be modified, discussing an ensemble of particles with these three characteristics and relating it to the size distributions shown in the lower right. Remove the size distribution graphs directly adjacent to the D/Do curves – it seems that this “ideal” aerosol never occurs, or if the authors chose to keep the curves, it should be made clear that they are associated with a distribution of particles with the same hysteretic characteristics.

Page 2, line 34: Figure 1 can be eliminated. The discussion on this page can be referred to Figure 2, which shows the same information.

Page 16, paragraph starting with line 544: Estimating dissolution RH using the AIM model, which only applies to inorganics when organics are present and when most of the summer aerosol did not even show hysteretic effects, seems to be inappropriate at best. Furthermore, using ammonium data from the IMPROVE network will result in a substantial underestimation of true ambient ammonium concentration. It has been shown that, especially in the warm summer months, much of the ammonium collected on a nylon filter will volatilize. Because of this volatilization issue, ammonium measurements have been discontinued in the IMPROVE program. An underestimation of ammonium will result in an estimated dissolution RH that is substantially lower than what it would be for the ambient aerosol. Therefore any discussion and model calcula-

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tions using IMPROVE ammonium should be removed from the manuscript.

Page 16, section 3.1.2: I believe this section, along with Figure 5, can be removed from the manuscript. The focus of the manuscript is on the AS-TDMA results, and section 3.1.2 adds very little to the main focus of the paper. Figure 5 merely shows an example scan that can be found in many other places in the literature. The deliquescence/efflorescence summary would be of interest if it represented the same scans or time periods associated with the AS-TDMA, which it apparently does not because of intermittent instrument issues.

Figures 6 and 7 represent nice summaries of the AS-TDMA data!

Figures 8 and 9 contain many data points that are overlaid on each other and can't really be differentiated. They add very little if anything to the main essence of the manuscript in that they only show data collected in two sampling periods. I suggest that these figures along with the associated discussion be removed from the manuscript.

Figure 10 is a nice summary of the AS-TDMA results.

Figure 13 and associated discussion should be removed from the manuscript, especially in light of the error in the bulk IMPROVE ammonium measurements. Also, the AIM model does not account for organic/inorganic interactions, and therefore dissolution calculations using this model are not expected to be representative of typical ambient aerosols.

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