Atmos. Chem. Phys. Discuss., 14, C1262–C1265, 2014
www.atmos-chem-phys-discuss.net/14/C1262/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Hygroscopic properties of newly formed ultrafine particles at an urban site surrounded by a deciduous forest in northern Japan during the summer of 2011" *by* J. Jung and K. Kawamura

Anonymous Referee #1

Received and published: 9 April 2014

There are a number of shortcomings in the paper. My biggest concern is that the authors attempt to conclude on the compositions of the particles based on HTDMA measurements. I appreciate the difficulty in analyzing the compositions of these nanoparticles but the tone used throughout the manuscript is not appropriate. At best, we can use the HTDMA measurements to infer the compositions or the HTDMA measurements can be explained by some proposed compositions. There are a number of places that I feel the authors have stretched too much in the implications/conclusions from their measurements.

1. Abstract is a bit focused too much on reporting the observations without enough science. The conclusion that "the hygroscopic property of large Aitken and small accumulation mode particles is highly influenced by the long range atmospheric transport of particles and their precursors" is of no surprise and is hardly an advancement of our understanding of atmospheric aerosols.

2. Pg 8259, line 20. . . "the hygroscopic growth factor of nucleated particles can be used to estimate their chemical compositions." I can understand that the authors perhaps used "estimate" to soften their tone but in my opinion, it is still an assertion that is not well substantiated. This is too strong a statement for field measurements.

3. The authors need to define the term nucleated particles more clearly especially when freshly nucleated particles and grown Aitken mode particles were discussed too. It is not clear if the discussions of "nucleated particles" of Ehn et al. and Ristovski et al. mean freshly nucleated particles or grown nucleated particles.

4. Page 8262, line 14: I cannot understand the sentence "Since a mode peak diameter of the Gaussian curve fit was used in this study, broadening effects caused by a transfer function between he first and second DMAs are negligible". The broadening effects are related to the spread of the measurements. It is not clear to me how the fitting would reduce or eliminate the broadening. Do you mean that broadening is not an issue since you are only concerned with the mode diameter?

5. Page 8264, line 2-4: "Increases in the number concentrations of humidified particles at dry Dp = 40nm were consistent with those at dry Dp = 20nm with and without a time gap". Pls rewrite this sentence. Meaning is not clear.

6. Page 8264, line 15: would some estimates of growth rates be useful to see if the increase in larger particle concentrations is due to condensation or inflow of different air mass?

7. The paper calculates equivalent g(85%) at dry Dp=100nm to correct for the Kelvin

effect. Kelvin effect depends on surface tension, which in turn depends on chemical compositions. The authors discussed the variation of compositions (as inferred from GF trends) of particles of different sizes, e.g. freshly nucleated particles and grown Aitken particles. Has this size dependence of surface tension been incorporated in the calculation? Would the uncertainty of the different surface tension be sufficiently large to affect the conclusion of the difference in GF of 20nm and 40nm particles and their inferred compositions?

8. Page 8265, line 3: "Water soluble organic aerosols are assumed in the conversion to obtain Equivalent g at dry Dp=100nm using the Kohler equation." This does not seem to be consistent with the compositions of particles as discussed later in the ms. For example, see the quoted sentence below.

9. Page 8265, line 26: "These results imply that water-insoluble particles emitted from traffic may contribute a large portion of particles at the dry Dp range of 40–120nm during the NPF event periods compared to the non-NPF periods." By reading Figures 3a and 3b, the difference is really not that great, less than 0.1 in g(85%), especially that the data for non –NPF (Figure 3b) fluctuate much more than those in Figure 3a. The oscillations of the data in Figure 3b are likely noise, probably because of lower particle concentrations. Hence the conclusion in the quoted sentence is a bit far-fetched to me.

10. Page 8266, section 3.3, why a bimodal distribution is presented in a typical highly hygroscopic particle distribution? Was the less hygroscopic mode always observed when the highly hygroscopic fraction appeared? If so, was it due to merely mixing of different air mass?

11. Page 8269, line 10-15, "The different behaviors of the hygroscopic properties indicate that different growth mechanisms for freshly formed nucleation mode particles may exist between the boreal coniferous forest in southern Finland and the present urban site adjacent to a deciduous forest in northern Japan." The authors argue that the growth mechanisms are different between this study and the Finland study show

different trends. However, is it possible that the freshly nucleated particles are different but the growth mechanisms are the same, which could also lead to different trends in terms of changes of hygroscopic properties between the nucleation and the Aitken mode particles?

12. Page 8269, line 20-25: "Increased number fractions of intermediately-hygroscopic particles at dry Dp = 20nm were observed when the burst of nucleation mode particles occurred (Fig. 5a and d), indicating that the hygroscopic property of freshly formed nucleation mode particles is intermediate." I notice that the number concentration shows a huge difference (5d) during NPF but the increases of the highly-hygroscopic fractions are moderate. I would imagine that if the nucleation mode particles were mainly highly hygroscopic particles, its fraction will overwhelm the rest. Figure 5a does not suggest that though.

13. The authors put the discussions of different air masses and the diel variations of particle concentrations almost at the end of the paper. I would think that it is more logical to put it up front as an overall discussion and then focus on those observations that are due to NPF. As is, these discussions appear almost like an after-thought. I also noticed that these issues are discussed first in the beginning of the conclusion.

14. Related to the above, I found (Page 8270, line 10-13) "Thus, local wind direction was an important factor controlling the growth of newly formed particles and their hygroscopic properties" very strange. The discussion was on different air masses. I would agree that local wind direction affects the OBSERVED growth factor of the particles but I have strong reservations that there is evidence to show that they affect the growth of the newly formed particles their hygroscopic properties.

15. Page 8271, line 10-15: "Significantly higher g(85 %)_total values at dry Dp = 120nm were obtained during the polluted periods (1.27 ± 0.05) than the clean period (1.19 ± 0.06)." I am curious what the PM compositions over the clean period are.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 8257, 2014.