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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characteristics of size distributions at urban and rural locations in New York" *by* M.-S. Bae et al.

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Characteristics of Size Distributions at Urban and Rural Locations in New York Min-Suk Bae1, James J. Schwab1, Olga Hogrefe1, Brian P. Frank2, G. Garland Lala1, Kenneth L. Demerjian1 1 Atmospheric Sciences Research Center, University at Albany, State University of New York, Albany, New York, USA, 2 Division of Air Resources, New York State Department of Environmental Conservation, Albany, NY, USA

Interactive comment on "Characteristics of size distributions at urban and rural locations in New York" by M.-S. Bae et al. Anonymous Referee #2 Received and published: 3 March 2010

This manuscript describes aerosol number size distribution measurements conducted

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Interactive Discussion

Discussion Paper

with twin-SMPS systems at three different sites during four intensive field campaigns in New York state. The authors conclude that diffusional losses in the sampling lines of the SMPS have a significant effect on the data and should be accounted for. They also give some general characteristics of the size distributions at the studied sites. The results are intersting, and the data sets are unique. However, in its' present form, I think this manuscript is not scientifically significant enough to be published in ACP, and requires quite significant additional analysis and re-structuring. My comments are below:

General comments: 1) First of all, I strongly agree with Reviewer 1's comments, which I think the authors should carefully consider.

Reviewer 1 submitted many thoughtful and constructive comments. We have carefully considered and fully responded to those comments.

2) The authors now use a lot of space to describe the diffusional loss calculation that was done based on a basic tube loss correction scheme. Based on their calculations, the authors conclude that diffusional losses indeed need to be accounted for in their sampling lines. I think this is general knowledge in aerosol science (e.g. described in basic text book of aerosol measurements such as Hinds(1999)), and most aerosol scientists are already correcting their SMPS and CPC data for diffusion (or at least they should be!). Besides this, as pointed out by Reviewer 1, the standard loss correction even does not seem to be working optimally for the data. Potential reasons for this could e.g. turbulence introduced by connectors, inlets and curvature of the tubing. Since the authors are not providing any new improved loss correction scheme, I do not think that making the point that diffusional losses in sampling lines are important is worth the space that it is given now. On the other hand, I think it is nice to see particle number size distribution data from New York state - the data sets from these three sites are unique. However, I think that these data sets have not been analyzed deeply enough in this manuscript to provide significant scientific insight into the characteristics of the studied environments. Therefore, I suggest that the authors con-

ACPD

10, C1647–C1656, 2010

centrate on either a) quantitatively characterizing the diffusion losses and improving loss correction they are applying now - maybe resulting in an improved loss correction scheme that others could use too; or b) settle with doing the standard correction they are doing now, acknowledge its uncertainties, and concentrate on doing a more detailed analysis and reporting of the characteristics of the size distributions and number concentrations. This would perhaps mean combining the current manuscript with the "companion study" that the authors refer to on p. 85-86.

There are two aspects to this comment; one addressing the diffusion correction, and the other addressing the level of analysis the data has been subjected to. As reveiewer 2 notes, the diffusion loss issue was prominently raised by reviewer 1, and we refer to our responses to those comments rather than repeat them here. In short, we respect the concerns of the reviewers about the space given to diffusion loss correction, and we have moved all material related to the diffusion corrections, outside of a brief description, to the supplementary material. The second point about how we should “concentrate on doing a more detailed analysis and reporting of the characteristics of the size distributions and number concentrations” has also been addressed in the revised manuscript. We added a more detailed comparison and discussion about (1) Particle Growth Events at the Rural Site with growth rate, (2) Particle Growth Events at the Urban Site. 3) In particular, I would like to see a more detailed comparison and discussion of 1) the sources of particle number and mass at the different sites (for instance what are the relative contributions of traffic, long range transport or nucleation at different sitest); 2) the characteristics of the nucleation events at the different sites (as described by e.g. Dal Maso et al., 2005); 3) comparison of the results to similar studies that have been conducted around the world in other environments (see e.g. Kulmala et al., 2004 for a review). Now the authors are reporting very general results from the sites and not really putting them into a context where they would be useful for the scientific community.

We have added additional analysis and discussion of the features of the size distribu-

ACPD

10, C1647–C1656, 2010

tions measured at the various sites as suggested by the reviewer. It does not seem feasible nor desirable to analyze all the aspects suggested by us and by the reviewer in this manuscript – it would result in a very long paper! We have added analysis to section 3.3 and 3.4, and added two new short sections on particle growth events at rural and urban site. In section 3.12 we added the following explanation for one limiting case of the relationship between median diameter and PM2.5 mass: "The small particle limit is generally accompanied by observations of low PM2.5 mass, high number concentration, and low diameter caused by the occurrence of "small fresh particles" associated with two possible processes; 1) recent primary emission (as observed for the QC01 and QC04 campaigns) associated with Black Carbon (BC) or hydrocarbon-like organic aerosol (HOA), 2) particle nucleation and growth associated with sulfate (Jung et al 2006) (as observed for the WFM02 and PSP04 campaigns)." Figure 1 shows an example the relationship between number concentrations and EC. This figure provides evidence for process 1) above, but is not included in the manuscript.

In addition, we added a more detailed comparison and discussion about (1) Particle Growth Events at the Rural Site with growth rate, (2) Particle Growth Events at the Urban Site. 4) To gain more insight to the analysis, the authors might want to look at air mass trajectories on the studied periods (calculated with e.g. HYSPLIT trajectory model http://ready.arl.noaa.gov/HYSPLIT.php). This would probably help in analyzing the contribution and origin of different particle sources, and also give knowledge whether some air masses prevail when particle formation events or intense pollution episodes take place.

Thanks for your comments. We have, in fact, done some of the suggested analysis, but did not include it in the original manuscript so that the paper would not become too long and unfocused. We looked at air mass trajectories at the time of the strong growth event (The hybrid single-particle Lagrangian integrated trajectories (HYSPLIT 4.5) model at the sampling sites on July 24 at 10:00 AM in 2004 and on July 24 at 8:00 AM in 2002 (24 h backward trajectories with elevations of 2, 267, 517 m (above

ACPD

10, C1647–C1656, 2010

the sampling site level) indicated by every 6 h. Based on the trajectories, it seems that the clean air mass from the north areas could have been transported to the PSP and WFM sampling sites, implying that transport could be responsible for the condition of ambient particle events at the sampling sites. Please see Supplementary Material Fig S-4. 5) Were there meteorological data (such as ambient RH, temperature, wind direction, solar radiation, pressure or trace gas concentrations) available at these sites? A comparison to meteorological data recorded at the measurement sites might also give insight on e.g. the effect of boundary layer dynamics on particle concentrations.

Thanks for your comments. As seen in Figure 2 below, we explored that the relationship between time series number concentrations (#/cm3) from Nano SMPS and vector plots of wind direction and speed, temperature (Celsius), & Relative Humidity. One might note that the very strong particle growth events during the summer at the rural sites were associated with large drops in temperature. But, not all drops in temperature were associated with bursts of particles, so a simple cause and effect seems not to hold. One point that does seem to come from an inspection of figure 2 is that in the urban location, local sources seem to dominate over meteorology – with essentially daily bursts of particles; while in the rural locations, the periods of large particle concentrations are more likely to be linked to meteorological conditions.

6) Regarding the characteristics of the nucleation events, it would be good if the authors would report particle formation and growth rates - this would help putting the results to a larger context.

In response to this suggestion we have added two new sections to the paper: 3.11 Particle Growth Events at the Rural Site and 3.12 Periods of high particle concentration at the Urban Site. In addition to the added text in the main paper, there are figures and additional explanation of the analysis in support of these sections in the supplementary material.

Detailed comments: 7) p. 74, Eq. 2: Was the diffusion coefficient corrected for temper-

ACPD

10, C1647–C1656, 2010

ature? What about slip-correction? The authors should refer to a source where they have taken the diffusion coefficient formulation.

Diffusion coefficients and slip correction values were calculated at 293 K, which was the temperature (within a few degrees) of the shelters where the instruments were located at each site. The diffusion coefficient formulations was taken from Hinds (1982), which is cited directly above equation 2. Source: Hinds, W. C.: Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles, John Wiley & Sons, NJ, 1982

8) p. 76, Eq. 6: The authors are calculating the mean free path of air. However, in the condensational sink calculation, they should use the mean free path of the condensing vapor (often assumed to have the properties of sulfuric acid) - so in this case Eq. 6 and its application for 0.0664μ m does not apply. There is thus probably an error in the condensational sink calculations.

Thank you, this is a good point and, we agree with the reviewer. We recalculated the condensation sink using the mean free path of H2SO4 and found values for the condensation sink roughly 50 percent higher. To clarify this point to the reader, we have changed the following sentences to the text: "At 293 K and 1 atmospheric pressure, the mean free path is 0.039 μm for H2SO4." "The averages (+ standard deviation) of the CS (8.35 – 283.9 nm) yielded 2.3 (+ 0.9) * 10ˆ-2, 8.7 (+ 4.4) * 10ˆ-3, 3.5 (+ 1.3) * 10ˆ-2, and 9.2 (+ 4.7) * 10ˆ-3 (1/s) for the QC 01, WFM02, QC 04 and PSP04 campaigns, respectively. The CS in the urban areas is roughly two to three times higher than rural areas due to differences in number concentrations and size distributions."

9) Generally, the authors often give far to many decimals in their numbers, considering the accuracy of the experiments. For instance, in Table 1, the size ranges are given with the accuracy of 0.1 nm. I find it hard to believe that this was the precision of the instrument.

The reviewer raises a good point. The size ranges, which were reported here, are

ACPD

10, C1647–C1656, 2010

the output values reported by the TSI instruments and software. Rather than adjust every reported number, then making sure they were self-consistent, we simply used the values reported by the instruments.

10) p. 79, line 24: The authors state that they do not understand the reasons behind the differences in the DMA merging points. On the other hand they imply that it might be related to the fact that one campaing was in winter and the other in summer. Could the authors elaborate on this. For instance, what were the ambient temperatures or relative humidities? What about the temperatures / humidities in the SMPS-systems? Was the sample dried? These issues might have an effect on the interpretation of the measurements.

Thanks for your comments. All SMPSs had dried samples with driers in sheath flow and collected samples in the shelter under room temperature. We really do not understand why only one campaign had a merge point different than the other three. Three of the four campaigns were during summer, and only one of those had the lower merge point. We have removed any speculation about the reason for the difference from the revised manuscript.

11) The urban site was the only site that had measurements for both summer and winter. In section 3.8. the authors should make a clearer point of comparing the characteristics of particle size distributions in summer and winter. One can, for instace clearly see that there are a lot more particles in the winter than in the summer. This is naturally probably related to 1) combustion 2) lower atmospheric boundary layer during the winter.

The reviewer raises a good point that will help add clarity to the manuscript. To clarify this point to the reader, we have changed the following sentences to the text: “Possible reasons for this seasonal difference are changes in vehicular emissions caused by cold starts during cooler parts of the year or increased residential heating & burning and/or lower atmospheric boundary layer during the winter.”

ACPD

10, C1647–C1656, 2010

12) I suggest that the authors plot the size distributions (Figs. 11) in logarithmic concentration scale. This would reveal some of the general features of the data better.

Thanks for your comments. We have inserted the logarithmic concentration scale for all four campaigns.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 69, 2010.

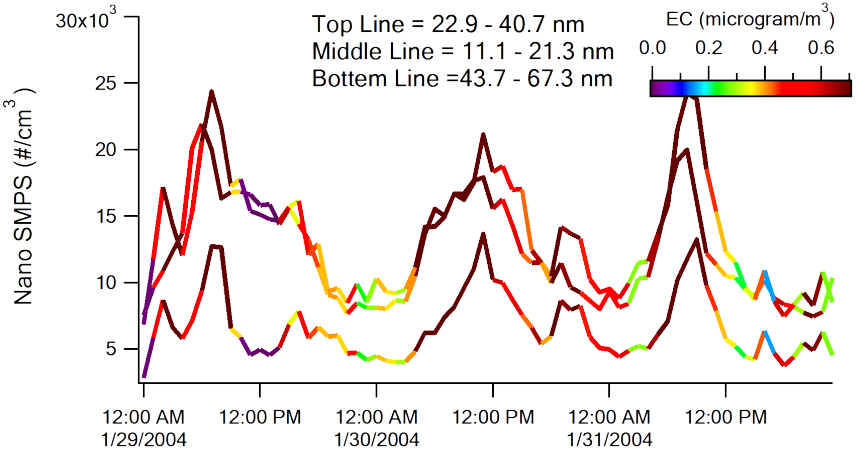

Fig. 1. Time series number concentrations (#/cm3) from Nano SMPS colored by EC concentrations (microgram/m3)

ACPD
10, C1647–C1656, 2010

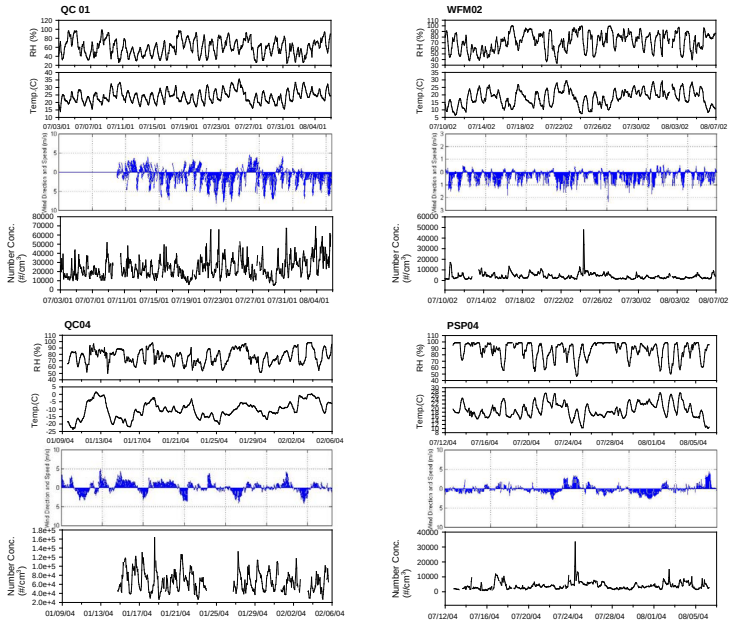

Fig. 2. Time series number concentrations (#/cm3) from Nano SMPS, vector plots of wind direction and speed, temperature (Celsius), and Relative Humidity (%).

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10, C1647–C1656, 2010