**Atmospheric
Chemistry
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Discussions**

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

Anonymous Referee #1

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The manuscript describes measurements of aerosol size distribution at three sites: one urban in New York City and two rural sites in New York State. General characteristics of size distributions and absolute concentrations are discussed. The effect of diffusion loss correction on size distribution and aerosol number concentration is given significant attention. I agree with the authors that diffusion loss correction should be applied to the aerosol size distribution data, but feel that the manuscript does not provide sufficient evidence for the accuracy of the method of correcting the data. While the correlation with the CPC is improved when applying the correction, there are indications that the spectral shapes of the nano-SMPS and long-SMPS in the overlap region do not match after the correction. The paper is rather tedious to read and should be substantially shorted. For example, section 3.2 can be entirely removed, because it has little value: the formulas for the TSI diffusion loss correction can be found in the

literature (Reineking and Porstendörfer, 1986; Birmili et al., 1997; Chen et al., 1998).

Specific comments:

Abstract, lines 14 – 17. The high number concentration events in the urban sites are attributed to primary emissions, while in the rural sites are attributed to nucleation events. Nucleation events have been observed in urban areas. On what basis are the high number concentration events are attributed to primary emissions alone?

Section 2.2. The CPC counts were diffusion corrected (Section 3.4). This needs to be mentioned in this section.

Section 2.3.2. The inclusion of the activation and counting efficiency of the CPC into the total diffusion loss correction is confusing, because these processes have no common physical basis. Why then is the efficiency of bipolar charging not included into the "total penetration"?

Section 2.4. When discussing the condensation sink, the authors correct the mean free path for the temperature and pressure effects (Eq.6). Were these effects substantial? If so, were the calculations of aerosol size distributions corrected for these effects or were the standard settings of the TSI software used?

Sections 3.2. I do not understand the purpose of this section. Was it to determine the diffusion loss correction by the TSI software? If so, the easiest way would have been to compare the aerosol size distributions obtained with the new and old versions of the TSI software. The formulas for the TSI diffusion loss correction can also be found in the literature (Reineking and Porstendörfer, 1986; Birmili et al., 1997; Chen et al., 1998). What is done, however, is rather confusing and shows a lack of understanding of the SMPS data inversion, which is more complex than a simple factor connecting the CPC counts to the actual size distribution.

For example, the authors use "transfer function" to describe a relationship between the raw counts collected by the SMPS and the size distribution reported by the TSI

software. This term could be very confusing, because it is usually used to describe the efficiency of particle transfer through the DMA as a function of particle electrical mobility (see Knutson and Whitby 1975, Wang and FLagan 1990, etc.).

It is also not clear what exactly the "empirical transfer function" incorporates, because the authors do not specify how the "raw" counts per size bin were obtained. The TSI SMPS operates by collecting CPC raw counts at 10 Hz, i.e. the CPC counts are equally distributed in time. On the other hand, the size bins produced by the TSI software are equally distributed in log-size (usually 64 bins per decade of particle size). It is obvious that the raw CPC counts could not correspond to the size bins: different scan rates produce different number of time increments, but the number of size bins remains practically the same. My guess is that the authors used "raw" counts exported by the TSI software, which re-bins the CPC raw counts into size bins, corresponding to the size bins in the inverted size distribution. If my guess is correct, then the "empirical transfer function" includes not only the diffusion correction, but also the charging efficiency correction and all the flow rate effects in the data inversion. Basically, these "empirical transfer functions" are applicable only for the current flow setting (via the ratio of the sample, sheath and CPC flows, and the diffusion broadening of the DMA transfer function). Therefore, these transfer functions are of little use and should be removed from the paper. Another important question is: was multiple charge correction used? Depending on size distribution, it may have a significant effect on what the authors call "empirical transfer function".

Page 78, lines 9–18. The noise at small sizes is common even when no diffusion correction is made. Yet, the authors present it as if the diffusion correction was the only reason why they have neglected the data below 8 nm. In fact, the diffusion correction is still minor relatively to the other corrections during the inversion, which is apparent from Fig.2. I do not think the noise justifies the removal of data below 8 nm.

Section 3.3. The difference of agreement between Nano-SMPS and LDMA at different sizes in the overlap range raises a question of how accurate/valid the diffusion correc-

tion is. Even though no examples of size distributions measured with the Nano-SMPS and LDMA are given, the size dependence of r^2 suggests that the spectra have different shapes. I also doubt that r^2 is suitable for judging the agreement. Smaller size bins usually have higher noise due to the weaker counting statistics amplified by the charging and diffusion loss corrections. This would lead to lower r^2 at smaller sizes even if the agreement between the two instruments is perfect on average. Would the ratio of concentrations be a better measure? Also, the use of a single size bin for merging seems not very robust statistically; why not compare an average of several bins?

Page 79, lines 7–9. A different merge point for the PSP04 campaign is explained by "different operation conditions". What are these conditions? If the flow rates are meant, then this raises a question of the accuracy of the diffusion loss correction, because this is probably the only reason why the results would be different.

Page 79, lines 10–11 and Table 3. Why would the bins be different in width and size? Does the TSI software not export the data in a standard way?

Section 3.4. Why was the size-dependent counting efficiency of the CPC not taken into account when calculating the CPC-proxy data from the Nano-SMPS distributions? How would the particles smaller than 8 nm, some of which would still be counted by the CPC, affect the comparison? To support the statement about statistical significance of the slope increase, the uncertainty in their values should be provided. How was the statistical significance of the increase in r^2 determined?

Page 81, lines 8–9. What is "a much more dynamic situation"?

Page 81, lines 17–18. I would recommend applying the diffusion correction, provided it is accurate, not only to urban, but to any data set.

Page 81, lines 18–20. How the findings presented in this paper can be used to evaluate whether or not reprocessing (any reprocessing?) "extremely large data sets . . . will yield important additional information"? Will small data sets be any different in this

respect?

Page 82, lines 1–4. Why would the slower oxidation of intermediate volatility compounds lead to higher aerosol number concentrations, would it not be the other way around? A more probable reason would be lower mixing heights.

Page 83, lines 1–2. The statistical criteria for an outlier need to be specified.

Page 83, second paragraph. The selection of bin boundaries is rather arbitrary and probably is not needed, because practically no critical information is extracted from this binning. Showing average size distributions would have provided all the information (such as the smaller particles dominate the number concentration, for example). In the bin definition, the emissions are attributed to the second and third bins, yet in the following section (p.84, 5–10) the high number concentration events, which are dominated by the fist two bins, are attributed to traffic emissions. The definition of "ultrafine mode" given on line 14 contradicts the preceding attribution of the second and third bins to the ultrafine mode, because part of bin one and four also fall into this mode. The lower boundary of the accumulation mode is usually attributed to 100 nm, not 67.3 nm.

Page 84, lines 10–12. "There is a strong association between CPC and the SMPS size range of 8.35–19.8..." This is one of the examples of trivial statements in the paper. The CPC measures the total number concentration (or a close proxy of it), the SMPS measures another proxy of the total number concentration, the two correlate (see section 3.4), the size distribution is dominated by the smallest size range. Is it then surprising that the CPC and the lowest size bins of the SMPS correlate? Of course, not. What new information does this sentence provide?

Sections 3.8 and 3.9. I do not understand why the relationship between the ion balance and the number concentration is investigated. Is there any physical or chemical process that would link the two? The discussion of ion balance and chemical composition seems irrelevant to this paper. Unless the authors present a compelling reason

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why this needs to be discussed, the parts dealing with chemical composition should be removed from the paper.

Page 86, line 24. What is "the small particle limit"?

Page 87, lines 7–13. I do not agree with these two sentences: a strong local source, such as a busy roadway, may influence both the number concentration and the PM2.5 mass; regional scale nucleation events may influence urban number concentrations, but not their PM2.5 mass.

Page 87, line 25. Indicate the units of CS values.

Page 88, lines 7–10. The sentence "The stronger relationship..." is confusing: what does "dynamic condensation processes from primary particles" mean? If it means that the primary particles contribute to CS, and CS correlates with PM2.5, then it contradicts the statement on p.87, l.7–13 that the primary sources do not contribute significantly to PM2.5.

Table 1. ASRC and DEC need to be defined. I would also suggest adding time periods for each campaign.

Figure 1. Why are the points not equally spaced along the x axis in the diffusion loss graphs? The SMPS usually exports size distribution data in equally spaced log-diameter size bins.

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