

## General Answer

*The authors of this manuscript would like to thank both referees for their comments and questions. It has improved the readability of the manuscript as well as the quality of the analysis. We have further developed the methods. These developments touch two main points.*

*The first such point is the calculation of the diameter uncertainty. The calculation has been improved to include the DMA transfer function, the voltage applied to the DMA and the uncertainty on the flows inside the DMA. In the previous work, we calculated the uncertainty on both sides of the measured diameter but kept only the biggest of the two, that we used on both sides. Now we use each uncertainty measure on their respective sides. Hence, the referees will notice that the boxes in Figure 3 are no longer symmetrical.*

*The second point that has been modified is the generation of the randomized points within the uncertainty boxes during the fitting procedure. Those randomly generated points are now more constrained within the uncertainty boxes than they previously were. We believe that this is more representative of the measured data. This has also raised the standards for the evaluation of the fits' quality.*

*Finally, we would like to point out that, because of these changes, the average fraction of ion-induced nucleation is now  $1.3 \pm 0.4\%$  compared to  $0.8 \pm 0.9\%$ . Note that these results overlap.*

## Anonymous Referee #2

### Overall comments:

This manuscript describes experimental aerosol data from a mildly polluted urban site in Finland. Size distributions of the new ultrafine particles have been measured both by a typical, assumedly stepwise SMPS-method and a comparable method with no neutralizer. From the field data, the actual charging state of the particles compared to their steady charging state in the neutralizer, can be derived and the result may give additional information from the initial nucleation mechanism. Also two kinds of growth rates can be obtained from the spectra: charged and steady state. New results are derived, mostly confirming the previous findings that pre-existing particles seem to scavenge free small ions and therefore decrease the fraction of ion-induced nucleation and thereby the extent of overcharging to be observed. The value of 0.8 % for IIN fraction is consistent with previous data obtained elsewhere.

I think that the title does not describe sufficiently what has been done in the paper. The title should reveal the fact that both a) measurements have been performed and b) new type of data treatment has been applied and more or less quantified.

*We have rephrased the title quite substantially, also taking into account concerns of Referee #1 into consistency in the choice of words used. The new title is: "Aerosol charging state at an urban site: new analytical approach and implications for ion-induced nucleation".*

Secondly, there seems to be a parallel numerously cited not-yet-published manuscript Leppä et al., 2011. I feel slightly uneasy about the fact that some of the important actions in

this manuscript are justified by citing that parallel paper Leppä et al., not yet available to the reader.

*The authors understand the referee's concern. Since the paper by Leppä et al. is not yet ready for publication, a full derivation of the equations is presented in Appendix A.*

### **Specific comments:**

Page 15884 Lines 12-13. Two thousand fits per day? Please modify the unclear sentence.

*This paragraph has been changed substantially and the phrasing has been changed: “Two thousand fits were made for each event day and polarity, the median  $S_0$  value and its corresponding  $K$  value were taken as the representative values, along with the median absolute deviation (MAD) as an error estimate. The MAD is a value reflecting how much the charging state varies from one fit to another due to measurement uncertainty (Gagné et al., 2008).”.*

Page 15885 Line 12. How is the factor  $f_{eq}$  calculated? Please describe. Is it same for both polarities?

*This was a very unfortunate omission: We are using  $f_{eq}$  as the described by Wiedensohler (1988) . We realize that this reference was missing. We have now added these sentences in section 2.3.4 that mentions Wiedensohler (1988):*

*“To calculate the ion-induced nucleation fraction, we multiply the charging state  $S_0$  of the event, obtained by fittings, by the equilibrium charged fraction  $f_{eq}$ . This gives the fraction of particles involved in nucleation that were charged at the diameter  $d_0$ . The equilibrium charged fraction used in this work is that given by Wiedensohler (1988). In the asymmetric case, the asymmetric charged fraction is used instead.”*

*Wiedensohler, A.: An approximation of the bipolar charge distribution for particles in the submicron range, J. Aerosol Sci., 19, 387-389, 1988.*

Page 15887 Lines 15-16. Unclear sentence. Please modify. Which are the four categories?

*Yes, it seems that there was a major problem in the text at this point. It is now fixed: “We classified the days into four categories: event, non-event, undefined and no measurements. The results are shown in Fig. 4.”.*

Page 15888 Lines 2-3. “This may be an indication. . .” Please clarify. What is that fact which may be the mentioned indication?

*This was indeed unclear. We changed the sentence to “The dominance of undercharged days in Helsinki indicates that the chemical or dynamical processes taking place in Helsinki may be different from those observed at the SMEAR II rural station where most days are classified overcharged.”.*

Page 15887 Line 2. First author's own name is misspelled.

*This has now been corrected. Thank you for pointing it out.*

Page 15887 Line 32. Initial "T" is missing for Dr. Petäjä.

*This has now been corrected. Thank you for pointing it out.*

Page 15915 Figure 1. Please indicate the row for 3.9 nm in the plot.

*This is done. We added an arrow pointing to the relevant row, and describe it in the figure caption.*

Page 15916. Figure 2. Please include units in the graph.

*This has been corrected.*