

General Comment

The authors implements previously-developed or still-developing methods for analyzing recent measurement data on the particle size distributions of electrically charged and total (charged + neutral) particles in 3-11 nm particle diameter range to learn about the contribution of ion-induced nucleation and also to verify whether it is possible to obtain diameter growth rates from measured electrically charged fraction of freshly nucleated particles in these size range. It is very meaningful that authors emphasize the effect of measurement uncertainty on the output of their theory-supported analysis. Their rural sampling site seems to be an ideal atmospheric environment for studying the dynamics of particle growth and particle-ion interaction occurring simultaneously under relatively small effect of scavenging by pre-existing larger particles and self-coagulation of nucleated particles and relatively high concentration of atmospheric ions. It is encouraged that authors continue accumulating data on this site since it is essential to have such data to verify any novel analysis technique suggested in this field. On the other hand, it is recommended that authors check the consistency of specific technical terms used in the paper before final submission.

Specific Comments

Is there a general agreement in the research field for the starting particle diameter (or critical size) of the IIN is 2 nm? Why not 1.5 nm or 1 nm? How significant the effect of chosen size on the conclusion made in this paper?

Page 15879, line 10. The method used to obtain GR from charged fraction in this paper is simpler than those used by Iida et al (2008). It may be inappropriate to state that the method was adapted from Iida et al (2008).

Page 15880, Line 1. The system used by Iida (2006) is based on a scanning mobility particle sizer (SMPS). It is not clear whether the DMPS operate under stepping mode or scanning mode.

Page 15882, Line 1 to 14.

The third simple method a reader can easily think of is to calculate a representative charged fraction at a given size range by taking the ratio of average concentration of charged particles and total particles during an event period. This method integrates all the particle counts during the event period; therefore, it is the most noise-reducing method. In addition, a systematic uncertainty of the representative charged fraction can be calculated from the number

of counts under ambient and neutralized modes. I imagine that authors chose not to take this simplest approach after long years of experience in the measurement of charged fractions, and the readers of this paper would like to know the reason.

Page 15882, Line 15 to 23

It is not clear whether the uncertainty mentioned in this paragraph is used as the value for the uncertainty box in the fitting procedure or not because descriptions here are not quantitative. In addition, the algorithm used to calculate the uncertainty of each method should be described more in detail. Readers would imagine that the uncertainty of the charged fraction of the first method is calculated from one standard deviation of the measured charged fraction. However, calculating in the uncertainty of the charged fraction of the second method is more involved. If authors have utilizing some statistical functions within a software to calculate the uncertainty of the slope they can refer to the function in the software. The uncertainty of the slope is most likely equivalent to the uncertainty of time-averaged charged fraction, which is different from the standard deviation among measured charged fraction by a factor of $1/(\# \text{ of data})^{0.5}$. It is also helpful to mention here why MAD is chosen as a parameter to describe the scattering of the data.

Page 15884, Line 6-16

It is likely that most reader would find this paragraph too technical to understand. Which method (1 or 2) is being referred to by stating “charging ratio” in the first sentence? The last two sentences are especially unclear. What is the meaning of “median fit”?

Section 2.3.3

I believe that most reader find the contents in this section confusing. It is recommended that author refine this paragraph to deliver important messages more concisely. Here are some points.

- Ion-DMPS only measures the f^{\pm} . I believe that f_{eq} , which is needed to calculate S, is calculated using charging theory by assuming ion mobility, mass, and concentration are same between positive and negative ions.
- The term f^{\pm}_{asy} is not well explained in words. I believe that f^{\pm}_{asy} are steady state charged fraction accounting for the difference in ion mobility, mass, and concentration between positive and negative ions.
- The term f^{\pm}_{eq} is a confusing term because it accounts for difference in ion mobility and mass between positive and negative ions but not concentration. I think that

author can explain about the analysis by using only f^{\pm} and f^{\pm}_{asy} .

- In Section 2.3.2. K and S are free parameter why ion concentrations are needed.
- I believe that one need to assume ion mass because ions masses are usually not measured at sampling site. However, I guess that ion mobility and concentration are measured by ion mobility spectrometer during most of the event days analyzed in this paper. It is not clear why ion concentration are taken from annual average values of BSMA .

Page 15885-15886, Section 2.3.4.

Some reader may find it confusing that the value of S_0 and K are separated into positive and negative in Table B1 & B3 since Section 2.3.2 says S_0 and K are the two free parameters in the polarity-symmetry model. It may be more consistent with the assumption of the model if input charged fraction, steady state charged fraction, ion concentration, ion mobility and ion mass are all average values between positive and negative to make all the input parameter entirely symmetrical. Then solve equation (1) and obtain S_0 and K . If S_0 turned out to be close to average of S_{asy^+} and S_{asy^-} the polarity-symmetry assumption is a good shortcut. Authors are free to decide either using current or above suggested approach.

Page 15886, Line 16

It is more appropriate to state that “agreed well with GR_{PSD} ” not “very well”.

Page 15886-7, Section 2.3.5.

Steady state charged fraction is non-linear function of D_p . If the measured f in some event days look non-linear function of D_p the author might have thought about applying a linear fit on a logarithmic scale, then express df/dD_p in Equation 6 or 7 as $(f/D_p) \cdot (d\log f/d\log D_p)$. It is not clear why authors chose to consider f is always a linear function of D_p .

In addition, Iida et al (2008) solved for f vs D_p by integrating the growth trajectory (D_p vs t) multiple times by adjusting GR_f and until the solution fit with the measured f vs D_p ; therefore, they did not obtain any slope by fitting because derivative taken from real data are sensitive to noises in the data. However, the approach taken by them was very time consuming.

Ion concentration directly affects the solution of GR_f , therefore, the ion concentration needs to be taken from the same NPF period during which Ion-DMPS was operating. It seems important to state in this paragraph that how the ion

concentrations were selected as an input of this method.

In the polarity-asymmetry case (Eqn. 6) the difference in the values of ion-particle combination and recombination coefficients between positive and negative are not accounted.

In Eqn 5 & 6 the ion-particle recombination coefficient is approximated by ion-ion recombination coefficient. Eqn 5 & 6 are still in the form of original rate expression under steady state; therefore, it does not seem necessary to apply any approximation in this equation. For example, ion-particle recombination coefficient in Eqn 5 can be calculated from the average ion property at each size.

Minor Comment

Page 15887, Line 15-16

The message of Figure 4 is very clear but the associated description in the text is not clear.

15888 Line 15 & 16

The difference between Class-I(a) and Class-I(b) are not described.

Figure 2. Units are missing. The axis title should be corrected to “concentration of charged particles”.