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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud condensation nuclei activity at Jeju Island, Korea in spring 2005" *by* M. Kuwata et al.

M. Kuwata et al.

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We thank the referee for the useful comments and positive remarks. Our responses to the comments are described follows.

Comment:1. Lines 209-214: D50 is defined as threshold diameter (CCN/CN = 50%), which is similar to effective cut-off size given by Dusek et al. (Science, 2006, 312:1376). Since the authors use "threshold diameters" (line 79) to represent D50, and frequently compare D50 with Dcrit (obtained from theoretical calculation based on Kohler theory), more explanations beyond lines 211-214 (which need to be rephrased) are needed to validate the definition of D50 and comparability between D50 and Dcrit.

Reply: We used D50 (observed) and Dcrit (calculated) diameters as the threshold diameters for CCN activation in the manuscript. The comparisons of these diameters

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have been performed in various laboratory experiments, and they agree very well in laboratory experiments (e.g., Corrigan and Novakov, 1999; Kumar et al., 2003). These results demonstrate the validity of the comparison of D50 and Dcrit. A description of this point is added in the revised manuscript.

Comment: 2. Since the long DMA used in this work most likely underestimates particles with a mobility diameter smaller than 40 nm, it is necessary to discuss how such artifacts affect the reported data and interpretations.

Reply: We assumed Knutson-type DMA transfer function to perform the multiple charge correction of size-resolved CCN data. Particle loss in the DMA does not affect the quality of the correction because both CN and CCN concentrations are measured downstream of the DMA, and the fractions of multiply charged particles are negligible for particles smaller than 40 nm. In addition, because CCN number concentration was measured without using the DMA, the characteristics of the DMA do not affect the CCN number concentration reported in the manuscript.

Comment: 3. Lines 244-254: (a) The authors equate D50 with Dcrit and then attribute temporal variation in D50 to different chemical composition (lines 244-248). How valid is such comparison and interpretation? Differing from the interpretation (lines 244-248), in the following paragraph (lines 250-254), theoretical calculations are a basis to conclude that field monitored particles, in average, have "rather uniform" chemical composition. Approaches employed in these paragraphs appear to be less than consistent. In the manuscript, at times, D50 is taken as Dcrit to provide interpretations, while a substantial amount of discussions is later given to examine discrepancies between D50 and Dcrit. Approaches and/or reorganization in this manuscript are worth re-considered to provide integrated discussions and understanding.

Reply: We use D50 as the observed activation diameter, and Dcrit as the calculated value. The average values of B show that the chemical compositions averaged over the observation period were not significantly dependent on particle size. However, this

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does not necessary mean that the temporal variations of D50 (or B) at four different supersaturations always correlate. Thus, we do not think that the description in the manuscript is inconsistent.

(b) Since Dusek et al. (2006) have shown that size, rather than chemical composition, of particles is the major factor affecting cloud nucleating activity based on both calculations and field monitoring data, comparing observations in this manuscript with the study of Dusek et al. will yield interesting discussions.

Reply: The objective of Dusek et al. (2006) was to clarify the important factors affecting the CCN number concentration. However, the purpose of our research is the closure of the activation diameters. For this reason, our analysis is quite different from that of Dusek et al. (2006). Thus, we regard the comparison with Dusek et al. (2006) as beyond the scope of the study.

(c) Lines 252-254: Why is the statement given based on selected size range of 30-160 nm? Didn’t ambient particles monitored with a size range of 10-300 nm (or up to 1 um?) as given in line 164?

Reply: The diameter range of D50 is 30 – 160 nm. Thus, we refer to the size range here.

Comment: 4. Lines 274-275: Is "... particles appeared and began to grow." meant to describe growth of small particles in "size" or "number"? Other descriptions using a similar phrase of "particle growth" or "new particle formation" should be rephrased or clarified. For example, does the sentence in lines 285-287 use "particle formation" to suggest appearance of more particles monitored? Since concentration trends in CO suggest that anthropogenic emissions enhanced CCN (lines 221-225), how could transported pollutants (e.g., combustion emitted particulates) contribute to the presence/addition of "new particles", and affect the current interpretation of hygroscopic properties (or D50) of aerosols monitored? This question also applies to interpretation of "newly formed particles" in lines 484-487.

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Reply: The term “particle growth” is used to describe the particle growth in size. This point is clarified in the revised manuscript. Regarding the relationship between enhanced CO concentration and CCN number concentration, please see the reply to the referee #2.

Comment: 5. Lines 355-361: It is unclear how correlation among D50 at various SSs leads to an interpretation that "... temporal variation of mass fraction of PM2.5 is reflected in D50 at higher SSs". How valid is such a claim since D50 is derived based on particle size of 10-300 nm (or a smaller size range)? Is temporal variation of mass fraction of PM2.5 available in the manuscript?

Reply: We did not derive D50 based on the size distribution measured by SMPS. We determined D50 using size-resolved CCN measurement. Thus, there is no concern regarding the validity of the data in this sense. As mentioned in the manuscript, D50 depends on A (Kelvin effect) and B (Raoult’s effect), which are determined by the chemical composition of the particle. The statement that "... temporal variation of mass fraction of PM2.5 is reflected in D50 at higher SSs" is based on the comparison of D50 (SS = 0.097%) and the chemical composition of PM2.5 as shown in Figure 8. It is true that there is gap in size ranges between PM2.5 and the CCN particles described here. But as you can see in Figure 8, the correspondence is remarkable.

Comment: 6. Sections 5.2-5.4: The major concerns on these sections are employed directions and approaches of theoretical calculations as well as discussions focusing on potential factors affecting chemical composition and resultant CCN properties of particulates for the following reasons: (a) Estimated "B" of Kohler equation in this study is claimed to vary little under individual SSs (lines 249-254), suggesting that chemical composition insignificantly affected hygroscopic properties of particles monitored in this study. Justification is needed for, later in the manuscript, a substantial amount of attempt to evaluate effects of chemical composition on hygroscopic effects of particulates. The justification should be consistent with data and interpretation presented in this study; (b) "B" is also claimed to mainly depend on inorganic components, whose

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concentrations substantially dominate over water soluble organic compounds (lines 349-353 & Fig. 8). Since water soluble organic compounds play a minor role of affecting hygroscopic properties of particulates as mentioned in the manuscript, three scenarios using oxalic acids, adipic acids, and HULIS deviate from real particles monitored in this study. In particular, laboratory and theoretical studies (e.g., Raymond and Pandis, JGR, 2002, 107:4787) have shown that while hygroscopic behaviors of NaCl and ammonium sulfate agree well with predictions using Kohler theory, various compounds, including adipic acid, significantly differ from theoretical calculated results. This suggests foreseen deviation if one adopts adipic acid as a model compound for calculations. Instead, employing fractions (e.g., upper and lower bound) of water soluble inorganics for theoretical calculations will yield more meaningful data and discussions.

Reply: Although the variation of B was small, it was not constant. Thus, the variation can be compared with chemical composition, which was simultaneously measured. We used the adipic acid approximation to simulate the case where the average properties (molecular weight, density, and elemental composition) of water-soluble organic carbon were close to that of adipic acid. This does not mean that we think that all water-soluble organic carbon came from adipic acid. We changed the text to clarify the point.

Comment: (c) Validity of adopting chemical composition of PM2.5 (in mass concentration) to particulates smaller than 200 nm is questionable (lines 369-370). Chemical composition of particulates is known to be size dependent according to published articles and statement given in this manuscript (Topping et al., 2004) (lines 460-463). Hence, rather than superimposing chemical composition of PM2.5 to submicron particulates, and then discussing inappropriateness of such an assumption/application (section 5.3.1), it is more valid to employ a range of fractions of water soluble compounds (i) available in literature along with postulated fractions of water insoluble compounds (if such data are unavailable in published literature), or (ii) experimentally analyze fractions of water soluble vs. insoluble compounds in particulates of size consistent with

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monitored aerosols. This will yield more realistic theoretical calculations/predictions and meaningful assessment of potential factors (e.g., density and surface tension of particulates) influencing hygroscopicity of particulates monitored in this study. Following comments 3(b) above, it is worthwhile to compare results obtained in this study with data of Dusek et al. (2006) to demonstrate contribution of this work.

Reply: We agree that if the size distribution of chemical compounds is available, it is better to use such data. However, it is not available. For this reason, we employed the PM2.5 chemical composition for the calculation. The reviewer suggests that we use the chemical composition available in the literature. We have already performed the comparison using the data described in Mochida et al. (2007). To clarify this point, we change Table 6 to a figure. We do not think that it is useful to derive the “soluble fraction” because some assumptions (e.g., complete insolubility in organic compounds) are required. Please see our reply to comment 3(b) for the comparison with Dusek et al. (2006).

Comment:7. Lines 121-122: Is the sentence "Concentration of CCN (SS=0.97%) ..." based on data given in Table 2, Fig. 4(b), or any source?

Reply: It is based on Figure 4(b), as D50 (SS = 0.97%) is about 30 nm. This point is added to the revised manuscript.

Comment: 8. Lines 260-261: According to Figure 4b, why are the data on March 27 (relative to the dates listed) excluded from time periods showing "new particle formation"?

Reply: We could not study the number size distribution of March 27 because SMPS data were missing on that day.

Comment: 9. Lines 263-270: Is the sentence "Concentration of CCN (SS= 0.097%) ..." (lines 263- 264) given based on Table 2, Figure 4, or ? What are reasons that March 29 and 30, instead of March 25 and 27, were selected for discussion of occurring events?

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Reply: It is based on Figure 5b. We selected this event because it was the most significant particle formation event during the observation period.

Comment: 10. Lines 277-291 and Fig. 5: (a) This paragraph is difficult to follow. Specify figures or tables for individual relevant sentences. (b) Do the red lines in Fig. 5 represent peak diameters of D50, CN, or CCN? If they were shown for D50 or CCN, specify corresponding SS. If they were shown for CN, why are the red lines absent from the period prior to 12:00 on Mar 29? (c) What are the main messages of this paragraph?

Reply: The paragraph describes the new particle formation that has affected D50 and CCN number concentration. We simplify the description so that the readers will be able to follows the manuscript more easily. In addition, Figure 5 is also modified.

Comment:11. Figures 4 and 5 contain complicated information. It will help the readers to follow relevant discussion if it is possible to rearrange their presentation.

Reply: We modify it so that the readers will be able to follow the manuscript more easily.

Comment: 12. Lines 306-312: What are the main messages?

Reply: We put Figure 6 so that readers can get the image of the size distributions more easily.

Comment: 13. Lines 341-346: Could the "good" correlation between D50 and water soluble fraction be expressed quantitatively? The qualitative description needs more evidential support or to be revised.

Reply: If the relationship were linear, we would be able to show the correlation co-efficient. However, according to Köhler theory, it is not expected to have the linear relationship. Thus, we did not show the goodness of the correlation quantitatively. This explanation is added to the manuscript.

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Comment: 14. Line 568: It should be "6", instead of "5".

Reply: It is corrected in the revised manuscript.

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