

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

Anonymous Referee #4

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This manuscript reports data on observed D50 at Jeju Island. This work also discusses potential factors affecting discrepancies between experimentally monitored D50 and theoretically estimated Dcrit under selected supersaturation conditions (SS = 0.097%). The topic is interesting and many data are provided. However, the article is somewhat difficult to follow; often, it is unclear which tables or figures should be read along with a description. Specific comments on technical approaches and organization, as well as questions for various clarifications are given below.

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),

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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.

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.

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. (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. (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 μm ?) as given in line 164?

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

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"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.

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 PM_{2.5} 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 PM_{2.5} available in the manuscript?

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 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

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inorganics for theoretical calculations will yield more meaningful data and discussions. (c) Validity of adopting chemical composition of PM_{2.5} (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 PM_{2.5} 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 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.

Other comments & questions:

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?
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"?
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?
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

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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?

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.

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

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.

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

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