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Interactive comment on “Size-resolved CCN distributions and activation kinetics of aged continental and marine aerosol” by A. Bougiatioti et al.

Anonymous Referee #1

Received and published: 16 June 2011

General Comments:

Bougiatioti et al. present results of size-resolved CCN measurements made in the summer 2007 in Finokalia, Crete. Detailed information is provided about the CCN properties of the aerosol; however, the manuscript reads as a report and could be strengthened in several ways, as outlined below. In particular, given that the aerosol impacting Crete is generally high aged, it would strengthen the manuscript if the authors could connect the size-resolved CCN activities with likely aging processes. While aerosol chemistry measurements are described in the methods section, these data were not utilized in detail to further understand the CCN characteristics of the aerosol;

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therefore, aerosol chemistry should be integrated further. In addition, the influence of different air masses should be discussed. Previously, Bougiatioti et al 2009 (ACP) reported non-size-resolved CCN activities in fall 2007 in Finokalia, Crete; therefore, in this manuscript, the advantages and new findings from the size-resolved measurements should be clarified. Further, it should be stated that the two FAME07 campaigns were not completed at the same times; this is not currently clear in the manuscript.

Specific Comments:

Abstract: From the abstract alone, it is unclear what is different from Bougiatioti et al 2009 (ACP); the goal of this new manuscript should be made clear. What is the suggested origin of the ~ 30 nm particles and ~ 100 nm particles? The CCN activities should be tied more directly to suggested aging processes; while photochemical aging and volatilization of less hygroscopic material from the aerosol are noted in the abstract, evidence supporting these hypotheses do not appear to be presented in the manuscript.

Introduction: The introduction is very long and cumbersome; it should be shortened and revised to emphasize only what the reader needs to know to understand the main points of the manuscript. If the goal of the manuscript is to examine the impact of aging on CCN properties, then this should be discussed.

Page 12610: This paragraph starts with the sentence “Size-resolved CCN activity measurements can quantify the role of composition on CCN activity.” However, this is not investigated in detail in this manuscript. In addition, nucleation and biomass burning events are discussed; however, these events are not discussed in terms of the FAME07 data.

Page 12613, Lines 9-11: It is noted that at the field site anthropogenic pollution can mix with strong dust events; however, this phenomenon is not discussed in the manuscript with respect to the FAME07 data.

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Section 2.3: This section is awkwardly placed between two CCN methods sections, and it also contains non-aerosol methods information, despite the label “Aerosol chemical composition and size distribution”. Consider reorganizing. Also, for example, lines 15-21 would seem to belong in Section 2.2. Further, the gas-phase and aerosol chemistry measurements are mostly not used in the manuscript and should be utilized to further understand the sources and aging processes of the aerosol.

Page 12619, Lines 22-25: What are the standard deviations (or confidence intervals) associated with these averages? Are they significantly different?

Page 12620, Lines 2-4: Was the aerosol chemistry observed to be nearly the same in both studies? This is not, and should be, discussed.

Page 12621, Line 6: State the diameter associated with this maxima here for clarity; also, what is the suggested sources of these particles?

Section 3.2, Paragraph 1: General aspects of aerosol chemistry are noted; however, this information should be integrated into the discussion of the CCN properties.

Section 3.2, Paragraph 2: How was K derived from the filter analysis? This did not appear to be discussed in the methods section. Did the previously reported FAME07 study experience influences from similar air masses? The statement that “the particles exhibit a small (but detectable) chemical heterogeneity” is vague and confusing; specifics should be discussed with respect to the chemical data.

Page 12623, Lines 12-17: Lower concentrations do not necessarily mean less variability in aerosol chemistry or properties.

Page 12623, Lines 25-26: Evidence should be provided for the suggested photochemical oxidation. Perhaps comparisons with the O₃ and NO_x data would be useful.

Page 12624, Lines 19-21: Why was the WSOC data from the summer FAME07 study not used?

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Page 12624, Lines 22-24 & Page 12625, Lines 12-14: Why wasn't a variable K utilized?

Page 12626, Lines 18-19: Was the variability in average droplet diameter correlated with air mass?

Page 12628, Lines 10-11: It is stated that "analysis of the droplet sizes using asymptotic and threshold droplet growth analysis suggests that the activation kinetics of ambient CCN does not vary..."; however, this finding may only apply to aged aerosol air masses and should be noted as such. It should not be generalized, as it likely depends on the extent of aerosol aging.

Page 12628, 13-17: This result is important and should be discussed further. Has this been observed previously? What in particular is unique about this study?

Fig. 1: Very high altitudes were utilized for the HYPSLIT air mass backward trajectories for this ground-based study. Are these representative of the boundary layer air? Do 100, 300, and 500 m backward trajectories appear similarly?

Technical Comments:

Page 12609, Line 4: Fix spelling.

Page 12610: For clarity, define size ranges of Aitken and accumulation mode particles.

Page 12610, Line 12: Where was FACE-2005?

Page 12610, Lines 17-20: Please clarify sentence.

Page 12611, Line 5: "ammoniated" should be "ammonium"

Page 12613, Line 15: Should this state "aerosol source region" rather than implying an exact "aerosol source"?

Section 2.2 Label: Consider renaming "CN & CCN Instrumentation" or similar, as there is "Instrumentation" described in other sections as well, making this label misleading.

Page 12615, Lines 27-28: This sentence is too vague; for example, what was the

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frequency of CCN calibrations?

Page 12618, Line 11: Fix typo.

Page 12619, Line 4: Briefly describe TDGA.

Page 12620, Line 7: “An example” should be “Examples”

Page 12620, Line 10: “diameter” should be “diameters”

Page 12620, Line 19: Fix typo.

Page 12621, Lines 3-5: Clarify this sentence.

Page 12621, Line 13: Fix typo.

Page 12624, Lines 4-5: What study does this statement correspond to?

Page 12624, Line 6: Fig. 7 does not show CCN activity as implied.

Page 12624, Line 7: Is this decrease statistically significant?

Page 12626, Line 24: Fix grammar.

Page 12627, Line 7: The extent of organic aerosol oxidation is not presented in this manuscript as implied here.

Page 12627, Line 23: Are these averages statistically different?

Page 12628, Line 3: Why is this value different than that reported on page 12625?

Fig. 5 caption: “Example” should be “Examples”

Fig. 8 caption: Please clarify what is meant by “chemical and mixing state parameters” as this is confusing.

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