

Interactive comment on “New Particle Formation in the South Aegean Sea during the Etesians: importance for CCN production and cloud droplet number” by P. Kalkavouras et al.

Anonymous Referee #1

Received and published: 8 August 2016

General Comments:

Kalkavouras and co-authors present results from an intriguing experiment in the Aegean Sea. The nature of pollution arriving at the long-standing Finokalia measurement platform is investigated directly with observations at Santorini, a site strategically located along the trajectory from the European mainland. The observations target aerosol size distributions, relevant primary and secondary pollutant concentrations, and detection of new particle formation events. The authors identify two events representing characteristic flow from the north, albeit of two distinct types. The Etesian flow example is marked with NPF events at both sites, although the events are stronger at the Santorini site. The authors extend their observations of particle number and

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composition to predictions of CCN at various supersaturations. They also go further to predict the total effect of the NPF events on cloud droplet number, taking into account the impact of constrained available water vapor. The paper starts with a nice scientific idea and goes into good detail into the results. What I see lacking is a little more connective tissue linking the observations at each site with each other and the mainland into a cohesive story. The material is already there, but it is somewhat buried and could be highlighted with a figure, for instance. I would like to see the following points addressed before recommending publication:

Specific Comments:

1. I recommend the authors adopt two shorthand names for the distinct periods (22/7-24/7 and 25/7-27/7). They could be referred to as “Etesian Flow” and “Moderate Surface Flow,” for example. Small changes like this could help the readability of the paper. A useful addition to this paper would be a two-panel cartoon, each overlaid on the Greek domain map in Fig. 1, for example, that describes the factors at play in these two periods. They could identify generally where they expect emissions, mixing, oxidation, NPF, and aging of new particles to be happening.

2. The WRF-Chem aerosol module configuration, as documented by the authors, is problematic for this particular application. It is quite likely that NPF events and subsequent processing are not captured realistically at all by the model. The sulfuric acid/water pathway parameterized by Kulmala et al. (1998) is likely not strong enough to enhance particles near the surface and lower troposphere to the levels observed at the Santorini site. It is now well-documented that other reagents play important roles in this process (e.g. NH₃ and organics), and these pollutants have been identified by the authors to be present and significant components of the aerosol. My guess is that most of the Aitken-mode particles in the model originate from direct emissions, not from secondary generation. A related issue is the lack of a dedicated nucleation mode in the model. Without this mode, any NPF events will artificially broaden the Aitken mode distribution and give unrealistic lifetimes against deposition and coagulation. It

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will also affect the growth rates predicted by the model.

The authors astutely sidestep relying on the model to predict size distributions and use their own observations when possible for calculating CCN and cloud drop number concentrations. However, since they include an entire section (2.2) detailing the regional modeling they performed, it is a good idea to explicitly state the limitations of this analysis for particle size distributions, and remind the reader that they are using the WRF-Chem output for its knowledge of advection flows and chemical composition, not microphysical processing.

3. The authors conclude that the NPF events observed at Santorini are regional in nature with a spatial scale of 250 km and characteristic transport time of 4.5 hours. They also assert that Finokalia does not see the bursts because it is 3 hours away and particle populations age before they arrive there. The authors do note that this second observation demonstrates how site-to-site variability can be important during a regional event. I am not sure that this totally addresses the issue though. Why are the events sort of regional and sort of not-regional? Is this an issue of using up the NPF precursors before the air mass gets to Santorini and then shifting to chemical conditions that favor condensation to available surface area? Or is something else at play here?

4. The paragraph beginning line 13 on page 9 describes an interesting hypothesis for how pollutants are transported to the middle of the Aegean Sea with limited aging. However, I'm not convinced there is enough evidence to warrant the detailed discussion that is given to this possibility or the certainty with which it is treated in the conclusions section. As described in my first comment, any model data related to the size distribution of Aitken-mode particles probably cannot be trusted in this case. If I understand correctly, the main assertion here is that the particles were formed over the Turkish mainland and transported quickly before they have a chance to be significantly coagulated away. Why could the enhanced number concentrations not come instead from oxidation and NPF over the water during transport, where there may be enhanced pho-

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tochemistry, complex interactions with clouds, interesting boundary layer phenomena, etc? If I'm not understanding the meat of the argument correctly, please explain it to me and consider rephrasing it in the text to be clearer. What insight do the model CO concentrations help to provide regarding the stratification and mixing of distinct layers downwind of the continent?

5. I recommend separating the paragraphs detailing the Nd calculations (starting on Page 12) into their own section, perhaps called "Impact of NPF events on cloud droplet number." Then section 3.5 would be called "Impact of NPF events on CCN production."

6. How is the partial sensitivity of cloud droplet number to chemical composition and vertical velocity determined? Can the equations be provided? What is the uncertainty associated with this? Please document it if possible.

Minor Changes/Typos:

Pg 2, Ln 25: The phrase "without any particular seasonal preference about their occurrence" is difficult to understand. Can the authors please reword this to be more specific?

Pg 3, Ln 4: "prior to reaching"

Pg 5, Ln 11-13: This is technically not a sentence.

Fig. 3 and caption: "open circles" not "cycles". Also, please indicate on the figure that the solid lines describe wind speed and the circles describe direction. It is hinted at in the figure and explained in the caption, but it would be quicker for the reader to have it identified visually, with an arrow or something.

Pg 6, Ln 16-17: The "less pronounced" diurnal cycle at Finokalia for ozone is not obvious to me from Fig. S4. Please include a plot of the actual diurnally averaged profiles or report the daily minima and maxima to demonstrate this point.

Pg 6, Ln 28-29: I would not characterize -21% or -15% under-prediction as "small".

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Either establish what they are small compared to, or please get rid of this qualification.

Pg 6, Ln27-30: Please break this sentence up. It is long and confusing.

Pg 6, Ln 31-32: Is there a more recent or relevant reference than McKeen et al. (1991)? Anything that specifically identifies this model scenario or modern European scenarios in general as suffering from ozone boundary condition issues?

Pg 7, Ln 5-6: In what way did the inorganic and organic mass concentrations show “similar behavior” to that of ozone? Are the authors just identifying them all as secondary pollutants? Please provide an estimate of the correlation coefficient or index of agreement for a statement like this.

Pg 7, Ln 9: Please remove the comma after the parentheses.

Pg 7, Ln 20: Please refer to some quantitative statistics to back up this claim.

Pg 10, Ln 6-8: This sentence is worded in a confusing way.

Pg 10, Ln 25 – Pg 11, Ln 19: Most of this material would be better-placed in the methods section (2.3 maybe). This goes for the second paragraph a page 12 as well.

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