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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "The role of sea-salt emissions and heterogeneous chemistry in theair quality of polluted coastal areas" *by* E. Athanasopoulou et al.

**E. Athanasopoulou et al.**

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(1) "The main flaw of the paper is the comparison with measurements: for the period that was selected to be simulated no measurements are available, which led the authors to compare their results with measurements with other time periods. Although this can be acceptable, no arguments exist on why that period was selected for modelling, since the selection of another year and/or season could coincide with measurements and would had made the comparison straightforward. There are some intensive campaigns through Greece, why they didn't select one of them?"

This is a valid concern (see also the first comment of the first referee). Unfortunately, we have been unable to find a period with the appropriate meteorological conditions,



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available information for anthropogenic and biogenic emissions, and suitable measurements for the detailed evaluation of the model. Our main concern was to select a simulation period with: -representative wind speed values over the area of interest, as wind speed controls SSA emissions -wind direction favorable for the transfer of sea-salt towards the Athens basin. -elevated pollution over Athens To address the above data limitation we collected as much of the available observational data and restricted our comparison of the model predictions to the averages and ranges of these measurements. While we do use the limited observational studies as much as practical for model evaluation, it should be kept in mind, however, that the point of the paper is not the model evaluation itself, but assessing the potential role of sea-salt aerosol. This gap highlights the need for further aerosol observational studies in Greece or similar area, and a more complete routine monitoring effort as well, at areas affected by both natural (marine) and anthropogenic emissions. Measurement sites could be indicated by this and other relevant studies. These points are now explained in the revised manuscript.

(2) " "urban characteristics" should be specified; is it emissions, elevation, meteorology, others? (p. 3813, l. 21)"

The sentence: "The applied version involves improved descriptions of the urban characteristics (Dandou et al., 2005)." has been changed to: "The applied MM5 version includes recent advances in the description of the urban boundary layer, such as incorporation of the anthropogenic heat and heat storage fluxes, as well as modifications in surface stress and heat flux (Dandou et al., 2005)."

(3) "How about domestic emissions? Further, were there any biomass burning events during the studied period? (p. 3815, l. 3-5)"

The available emission inventory includes central heating emissions for the winter. These emissions are zero during the simulated summer period. There were no major biomass burning events (fires) during the investigated period. We have used the



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typical summer weekday emission rates without fire emissions. We have added this information in the section describing the emission sources.

(4) "This is a real result, or it is just the effect of the grid-size? If there was no nesting over that domain, would that result remained the same or not? (p. 3817, l. 1-3)"

This is a real result for areas with extended coastlines like Attica and is independent of the grid-size used. This, of course, assumes that the correct coastline length is used in all scales. Surf-zone production is a more intense aerosol production path than the open-ocean for the typical meteorological conditions and plays an important role especially for Attica. We have modified this paragraph as: "Over Attica (in the nested domain), the surf-zone mechanism produces most of the total SSA mass, although the surface from which it originates is small. This is due to the higher intensity of surf-zone compared to the open-ocean aerosol production, combined with a relatively limited open-ocean area inside the nested domain, compared to that of the parent domain."

(5) "There are HNO3 measurements around Greece that the authors can compare their model with. As an indication, see references 1-5. (p. 3818, l. 23 -24)"

We appreciate this contribution. The suggested studies all show lower nitric acid levels than predicted by the model. This is consistent with our explanation regarding the under prediction of ammonium nitrate. The inventory used appears to underestimate ammonia emissions, leading to lower ammonium nitrate levels in the particulate phase and therefore higher nitric acid levels in the gas phase. A discussion of these results and their implication has been added to the paper.

(6) "Why not making a simulation with double or triple ammonia emissions and see how this affects the results? This will be a good indication on the author&'s conclusion on the ammonia emissions, and it would be interesting to see how the nitrate partitioning will change, already mentioned on p. 3818, l. 24. (p. 3819, l. 7-8)"

This is a good suggestion. We have performed such a simulation increasing signif-



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icantly the emission rates of ammonia. The following paragraph has been added to the paper describing the results of this test: "To study the sensitivity of the model predictions to NH3, an additional simulation was conducted increasing the agricultural ammonia emissions by a factor of 10 for the parent domain. This adjustment substantially improves the model performance in terms of NH3, aerosol ammonium and nitrate. Predicted ammonia concentrations approach observed values (about 2 ug m-3) over most of the sites of interest. Sulfate concentrations are slightly affected, as (NH4)2SO4 production is limited by available H2SO4. Nitrate and ammonium predictions improve at downwind areas of the high NH3 emissions, stated also in a similar study (Wu et al., 2008)."

(7) "It would be interesting to have a figure with the daily variability of aerosol composition over certain sites (p. 3820, l. 17 - 28)."

We have added a new figure (Figure 8) showing the daily variation of the PM10 concentrations of the major aerosol components in two sites: a marine location to the north of Crete and a polluted area in Attica. We have also added the corresponding discussion of these predicted diurnal profiles.

(8) "p. 3822, sect. 5.4: This is a scary result, it means that in order to have a correct sea-salt representation in models we need to have very high resolution?"

We are afraid that the implications of this result may have been misunderstood. This section discusses the comparison of a model trying to simulate the urban polluted area of interest (e.g. Attica) using a domain including only that area and fixed boundary conditions for sea-salt with one nesting the domain of interest inside a larger domain. Given the dependence of sea-salt transport into the city to sea-salt production outside its immediate vicinity the use of the nesting makes a significant difference. In other words, this section addresses mostly the importance of the outside (lower resolution) domain for the inside (higher resolution) domain. The correct sea-salt representation in low resolution models (e.g., regional scales) is possible provided that the coast-line



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effects are described appropriately (please also see our response to comment 4). To avoid misinterpretation of this result we have rewritten the beginning of this section. It now reads: "The use of a greater parent domain in which the area of interest is nested is found necessary during this event, as the transport of SSA from its boundaries is temporally and spatially variable. A significant fraction of the sea-salt over the urban area is produced in marine areas relatively far from it."

(9) "How this will affect models with not so high resolution? A comment about it would be very interesting."

Assuming that the medium or low resolution Chemical Transport Models are used to describe sea-salt production, transport and chemistry in an area where the anthropogenic emissions are more spatially homogeneous this would not have a significant effect. In our case it is the existence of the quite different scales (small length-scale for the description of Athens together with the larger scale for transport of sea-salt from the Aegean to Athens) and that makes necessary the nested description. If the appropriate coastline length is used in each cell together with the appropriate meteorological fields, then the SSA representation would be relatively independent of the resolution (at least in the 1-10 km scales investigated here). A short comment has been added to the manuscript.

(10) "Led to grater PM10 levels compared to what? A simulation without sea-salt? A simulation with "traditional" sea-salt sources? A simulation without gas-to-particle conversion? I guess the first, but it has to be clear in the text (p. 3822, l. 10-12)"

The above sentence has been changed to: "Use of SSA emissions in a simulation of atmospheric chemical dynamics over Greece led to greater PM10 levels than these predicted when excluding such emissions and..."

(11) "At Thrakomakedones, the concentration dropped to 1ug/m3 of the xxx ug/m3: xxx should be added here. (p. 3822, l. 22)"



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We have rephrased this sentence. It now reads: "For example, marine-related aerosol (2.5 ug m-3) comprises 15% of PM10 mass 9 km inland in the Athens basin. This contribution drops to 1 ug m-3 representing 7% of PM10 at Thrakomakedones."

(12) "It is not possible to say that the finer grid performs better than the coarse one, since no proper comparison with measurements was made (see general comments) in order to validate whether this is an improvement or not. (p. 3823, l. 3-5)"

We agree. The original sentence was not comparing the performance of the modeling approaches. To avoid misunderstandings it has been rephrased to: "A nested approach was used to simulate both the finer-scale processes (e.g., surf-zone SSA production and near-coastal gas-aerosol interactions), and the effects of sea-salt production, chemical transformation and transport from the Aegean to the urban area of interest (Athens)."

(13) "Figure 5: CL06 and G-M03 are very discontinuous (this is a log-scale), how does this affect the results, especially close to the discontinuity? Further, how did the authors selected the size threshold of the two parameterisations used?"

We have conducted sensitivity runs for each of these 2 parameterizations for the common size range and we found CL06 application predicting slightly higher but similar aerosol concentrations (hourly concentration differences up to 20% above land). We judge that the combination of these 2 parameterizations is necessary despite the discontinuity, because CL06 reaches the smallest particle size (down to about 0.005 um radius), but is applicable for particles up to 8 um in diameter (threshold depending on RH). Thus, we have used this parameterization in its range of application and then used G-M03 for larger particles. G-M03 has been found to agree reasonably well with observations up to 40 um in diameter at 80% RH (Gong, 2003 p. 8-6).

(14) Technical corrections: "p. 3809, l. 5: "life" should be "lifetime"."

Corrected.



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"p. 3809, l. 22-23: "describes" should be "describe"."

The verb "describe" refers to the word "generation", so no change is necessary.

"p. 3816, l. 2: "combines" should be "combine"."

Corrected.

"p. 3822, l. 19 and 23: "on land" would be "above land"."

Corrected.

"Table 1 legend: RH is in percentage or fraction from 0 to 1?"

This information has been added (RH fraction from 0 to 1).

"Figure 4 legend: SSA emissions are in strange units, why per cell instead of a more comprehensive per surface area?"

Corrected.

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 3807, 2008.