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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol chemistry above an extended Archipelago of the Eastern Mediterranean basin during strong northern winds" *by* E. Athanasopoulou et al.

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Comment 1: The authors state (page 9359, line 10) that the relation between meteorology and aerosol load "is not extensively studied" over the Eastern Mediterranean. I must somehow disagree. The authors cite at least four works covering this topic, but there is a number of modeling works covering the topic. So, I would rephrase the sentence, but if the authors want to keep the sentence as it is, they must provide stronger arguments for this fact.

Reply: This and the former paragraph of the introduction, focus on the aerosol mod-

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elling works over the Eastern Mediterranean (EM). The first of these two paragraphs presents the findings of pure aerosol studies (e.g. chemical composition, sources), which are indeed many in number (to the best of our knowledge they are at least nine). On the other hand, studies discussing the relation between meteorology and aerosol load are fewer (we have found only four). This was the reason the phrase 'is not extensively studied' was used. Having said that, we understand the concern and the argument of the reviewer, and therefore we will now use the sentence: "The relation between meteorology and aerosol load over the EM is less understood, and it has been only until recently that people started studying it using atmospheric models".

Comment 2: Also, they state (Page 9360, last paragraph): "[...] providing the first extensive evaluation of aerosol simulation performance over a wide region of the Mediterranean". I also believe that this sentence is not correct; again, a number of works have provided extensive evaluations of modeling performance over the Western and Eastern Mediterranean. This sentence is completely unnecessary in this context and should probably be removed from the manuscript.

Reply: Prior to this statement in the Introduction, all (to the best of our knowledge) aerosol modelling studies conducted for the region over the EM are cited. Most of those include comparisons with ground observations, while two of them are compared against flight measurements. Here, we should point out that the temporal and spatial (3D) coverage of the aerosol model evaluation in our study ($\sim$ 1500-2250 samples per species), is more extended than that of the previously performed comparisons. This was the reason we originally mentioned that this is the 'first extensive' study for aerosol predictions over the EM. Nevertheless, we can understand possible misinterpretations of the word 'first', which will be replaced by the word 'most'.

Comment 3: In Page 9363, line 22, the authors comment: "All air quality predictions presented [...]", which leads to think that the modeling results are forecasted outputs. However, the WRF model is driven by a reanalysis form NCEP, and therefore regional results are not forecasted, but driven by a reanalysis. Probably the authors should

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change the word "predictions" in order to avoid further confusion.

Reply: The aim of this phrase was to make clear that all gaseous and aerosol model values presented in the results are the output of the regional model PMCAMx, and not from the global model GEOS-CHEM. To avoid confusion that can be caused by the word 'predictions', we will replace it with 'model results'.

Comment 4: For the type of evaluation done in this work, having a detailed resolution for the vertical layering is an essential issue. I have doubts about the vertical resolution used. The authors indicate that they use just 14 model layers up to 5.8 km to cover the troposphere with the 1st layer at 21 meters (Page 9365, line 18), but do not provide further information on the vertical coordinates of the model. Further discussion should be devoted to this issue in the manuscript.

Reply: We performed some preliminary tests before deciding about the vertical resolution. In these tests we found that the vertical configuration of 14 layers with the domain-averaged layer top at 21, 29, 70, 129, 169, 228, 531, 869, 1256, 1696, 2166, 3252, 4496 and 5758 m above ground level, gives reasonable profiles, covers the boundary layer height, excludes higher altitudes with minimum aerosol concentration values, and therefore does not unjustifiably increase the computational time. It should be noted that mass transfer from higher altitudes is incorporated into PMCAMx simulations through the coupling of GEOS-CHEM to PMCAMx (top conditions). The same applies for the meteorological phenomena through WRF-to-PMCAMx coupling (more information on the configuration of the three models lies in Table 1 of the main manuscript).

Comment 5: In Page 9372, line 26, the authors indicate that "sulfate model performance is not consistent throughout the troposphere". The authors should elaborate on the causes of this discrepancy. Is it just a consequence of the low concentrations leading to a high value of the statistical figure considered, or is there any other cause for this vertical gradient of the error?

Reply: Yes, we support that this inconsistency is mostly related to lower values. About

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40% of the SO4 observations at altitudes > 2.2 km asl are low (below 1μg m-3), and it is known that a few large deviations between low concentration values can have a significant impact on the overall performance assessment. As mentioned in the text (page 9372, line 26 and onwards), when we confine the results above Turkey (25% of the total number of data pairs over 27 to 29 °E, correspond to altitudes from 4 to 7 km asl), then this difference becomes even more pronounced. Of course, since we focus on higher altitudes, thus air masses with non-local origin, the GEOS-CHEM model performance could have also interfered to the aloft discrepancies. Yet, a first evaluation of the model, performed for CO and O3 has shown that its performance regarding the physical and chemical processes that affect the gaseous species is adequate (Protonotariou et al., 2013; cf. list of reference in the manuscript). Another hypothesis for this discrepancy is inaccuracies in the injection heights of point sources (industries), which could pose some differential uncertainty per altitude. However, there is no data to support this hypothesis, thus it remains as a theoretical allegation.

Comment 6: In Page 9376, line 10, the manuscript claims that "fire activity is the main deficiency in the current model application with respect to organic aerosols". This sentence has to be carefully considered. First, the authors should provide evidence for the fire activity (e.g. satellite images) in the target region during the August-September 2011 episode. Moreover, if the fires were causing under predictions in particles levels, a substantial underestimation should be also observed for black carbon (which is not observed in this study). The authors should also explore the VBS mechanism for the formation of SOA, which could be causing the underprediction of organic aerosols.

Reply: In general, biomass burning plumes are known contributors to the organic aerosol load over the EM during summer. During the period we investigated, the aerosol load over the Aegean Sea is affected by the fire activity in the eastern Balkans and the western coastline of Black Sea, as indicated by the fire spots (cf. Fig. S2) and back-trajectory analysis from a previous study (Bezantakos et al., 2013; list of references in the main manuscript). For additional information and on the point regarding

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the possible errors introduced by the use of the VBS mechanism, please refer to our reply to the Comment 1 of Reviewer#2. Although we agree with the reviewer that no model underestimation is observed with respect to the average concentration of ground Elemental Carbon (and Organic Carbon), at the same time we cannot ignore the evidence mentioned above, which strongly indicates that the aerosol over the Aegean Sea is affected by the wildfires. Moreover, we want to stress the following points regarding EC: 1. Long-range transport of pollutants is more efficient in higher altitudes due to a lack of surface deposition and stronger winds. This also occurs in our case for the fire plumes: air masses originating or passing from the fire spots shown in Fig. S2 reach the atmosphere over the Aegean Sea in layers from 400 to 4500 m (Bezantakos et al., 2013). This creates an extra OA mass being measured aloft. On the other hand, the good performance for PM10 EC is found at the ground sites (filter samples). This is consistent to the PM10 OC measurements at the ground, which are also somewhat underestimated by the model system (Table S4), but perform much better than PM1 OA in and above the PBL (as indicated by the AMS airborne measurements). 2. As shown in Fig. 6, the 6-hour averaged values of ground observations and predictions are low ($\leq 1\mu$g m-3), with $\sim$40% of their differences being above 0.2 μg m-3. Similar is the average amount of the biomass burning fraction of EC measurements at Finokalia. The signal of fires on EC is most probably within the difference between observations and predictions over the Aegean Sea during summertime, as has been previously reported by Sciare et al. (2008).

It is commonly known that the accurate representation of emissions –including wildfires– in models strongly affects the quality of aerosol predictions (e.g., Heald et al., 2011). The fact that wildfires are not treated by the current modelling application, can explain the observed PM1 OA underestimation within PBL. Regarding the other types of sources, as well as the uncertainty of OA treatment within models, please refer to our reply to the Comment 1 of Reviewer#2. The main contributor to the PM1 OA underestimation (ca. 55%) of our current modelling application is the missing fire activity upwind the region of the Aegean Sea. This is a quite realistic hypothesis, given the

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observational evidence reported by Sciare et al. (2008) and Bougiatioti et al. (2014), as well as the modelling outputs of Bossioli et al. (2014). For more information on this issue, as well as our changes in order to address the points made by both reviewers, please refer to our reply to the Comment 1 of Reviewer#2.

Minor comments:

Abstract, line 1; Page 9360, line 26: What is exactly a "carefully designed model system"? How can a model system be designed "carefully"?

Reply: Our model system is composed of: a) the regional model PMCAMx, which provides the aerosol mass concentrations over the greater area of the Aegean Archipelago, b) the WRF model, which provides the meteorology to the PMCAMx applications and c) the global model GEOS-CHEM, which provides the initial and boundary pollution conditions to the PMCAMx application. The coupling between the models is characterised –"carefully designed"– because certain settings are set after thorough investigation: 1. The YSU PBL scheme for the WRF application is selected in the frame of this study because it is non-local and has a better performance than other schemes as far as water vapor mixing ratio predictions are concerned (Dandou et al. 2014), which is an important input parameter for air quality studies (Tombrou et al. 2015)." 2. Given that the PMCAMx model is not provided with pre-calculated PBL depths but with fields of diffusion coefficients (Kv), it is expected that the results are strongly dependent on the latter. For that reason, the WRF code has been modified so that it directly provides Kv outputs. These fields are regarded more accurate than Kv derived by the default procedure (offline calculation of Kv considering the calculated temperature, wind speed, friction velocity etc from WRF). Moreover, Kv values are then adjusted for the heights under 100 m, which is found to benefit air quality predictions (Environ, 2011b). This information is briefly provided in the ACPD version of the paper (pp. 9365, lines 11-14). 3. The default pre-processor provided by the environ.corp in order to adjust WRF output to PMCAMx inputs has been extended to treat frictional velocity, monthly vegetation fraction, fractional land use / vegetation type, gravimetric

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soil moisture and soil texture type, so as to provide hourly sea-salt and dust emission fields, as analytically described in Athanasopoulou et al (2008, 2009), which are cited in Table 1 of the manuscript. 4. The previous study which coupled GEOS-CHEM to a regional air quality model (UAM-V) over our area of interest (Tombrou et al., 2009) has shown the importance of a nested GEOS-CHEM application in order to provide initial and boundary gaseous concentrations to UAM-V. This method is also applied in the current study (Table 1). 5. "Detailed coupling of gases and aerosols between the PMCAMx and GEOS-CHEM chemical transport models (CTM) is carefully designed and applied here for the first time in order to capture more efficiently the airflows over the Aegean basin" (pp. 9360, lines 25-27). In particular, it involves 63 chemical species. Modifications to the model were necessary not only for the chemical speciation of some species (e.g., sea-salt mass of GEOS-CHEM should be broken down to Na, Cl, Br etc), but also for their size distribution (e.g., total sulfates of GEOS-CHEM should be broken down to the size bins of PMCAMx). Sea-salt and dust species treated by GEOS-CHEM are chemically resolved to the species treated in PMCAMx, following Athanasopoulou et al. (2008) and Kandler et al. (2007), respectively (cf. Table 2). Moreover, primary and secondary organic mass of GEOS-CHEM (e.g., Ketones, Limonene, Terpenes, and their oxidation & aerosol products) were processed so as to match with different gaseous species (e.g., monoterpenes, sesquiterpenes) and with the VBS treatment of PMCAMx (anthropogenic and biogenic oxidized and aerosol species of different volatilities). Also, in order to assess the relative contribution of the different OA precursors to the total SOA transported from outside the PMCAMx modeling domain (GEOS-CHEM BCs), each of the five lumped SOA species treated by the volatility basis set (VBS) scheme in PMCAMx is coupled to each unique oxidative product treated by GEOS-CHEM (instead to that of the uniform distribution of their mixture, Sect. 4.3). After this comment, we will include the above information within the main text or supplement, while the phrase 'carefully designed' will be replaced by 'comprehensive', in order not to confuse the reader.

Page 9363, line 14: please change "nitrogen oxide" to "nitrogen oxides"

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Reply: The change has been made.

Figures 2 and 6 are hard to read

Reply: We have improved the quality of these and the rest of the figures shown in the manuscript. Those will be separately uploaded together with the final version, following the rules of ACP.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 9355, 2015.