

## Responses to Reviewer #1

(1) The paper reports the comparison of PMCAMx AOD predictions with AERONET and MODIS observations. The comparison is made for limited time (1 month) and presented only for monthly average values. Periods with high dust contributions have been screened out, so that the evaluation is expected to reflect mainly the model's ability to reproduce the organic and secondary inorganic aerosols. In addition to the PM concentration, AOD depends also on the PM composition, size distribution, mixing state and interaction with atmospheric humidity. Thus, there are many possible reasons for the model measurement discrepancies and additional information is necessary to apportion the error. The paper heavily relies on a previous publication by Fountoukis et al. (2011), who compared the PMCAMx predictions of aerosol composition for the same period with ground based and airborne AMS observations. However, that comparison was fairly limited, including the data from only 4 stations for OA and secondary inorganic aerosols in PM<sub>1</sub>. The paper is clearly written and the methodology is presented in an understandable way. The authors have adequately answered majority of the previous reviewer requests with a few exceptions. The screening of the high coarse particle concentrations periods is one of the original characteristics of this work. This allows us to focus on the ability of the model to reproduce the organic and secondary inorganic aerosols as well as their water uptake. The use of one period with high photochemical activity also enables us to test if the model can reproduce the high variability (in space and time) of the corresponding concentrations. The use of detailed AMS measurements from ground stations and an airplane adds more detail to this comparison. In the revised paper we have added the evaluation of the model against additional ground measurements that were available during this period. Detailed responses to the comments of the reviewer can be found below.

(2) I agree with the previous reviewers, that more extensive comparison with the observations of e.g. the EMEP network would give higher confidence in the models ability to reproduce the near-surface concentrations of the aerosol components in Europe. Currently no evaluation is presented for instance for the modelled sea salt and the paper does not discuss how it could influence the AOD predictions over the sea.

We have compared the predictions of PM<sub>2.5</sub> and PM<sub>1</sub> of PMCAMx against the corresponding EMEP daily measurements in 27 stations. A total of 795 data points have been used for the evaluation. The model showed very little bias (fractional bias equal to -0.07) and reasonable scatter (fractional error equal to 0.49). The average predicted PM<sub>2.5</sub> concentration was 9.07  $\mu\text{g m}^{-3}$  while the average observed was 9.82  $\mu\text{g m}^{-3}$ . This performance is quite similar to the one reported by Fountoukis et al. (2011) for the EUCAARI stations and airborne campaign for the same period. A summary of this intercomparison has been added to Section 4 and the detailed results have been added to the Supplementary Information (Tables S4 and S5). Please note that the high sea-salt periods are also screened together with the dust. Therefore this paper does not address the ability of PMCAMx to reproduce the sea-salt concentrations. We have added this point in several points including the abstract and conclusions in the revised manuscript.

(3) The temporal and spatial correlations of the model with the MODIS and AERONET observations should also be reported, as was requested by referee #2 - the

current set of model scores does not provide the information on how well the model can reproduce the AOD patterns and variations.

The ability of the model to reproduce the spatial pattern of the measured AODs is shown in Figure 5 of the original paper in which the station-average AODs are compared. The maps of Figure 3 also illustrate this comparison in the form of a map. We have added in the revised paper the corresponding correlation coefficients for completeness. Comparisons of the predicted and measured diurnal average AOD profiles in 24 AERONET stations are shown now in the Supplementary Information. A lot of the diurnal profiles are relatively flat during the period of the AERONET measurements so quantitative correlation analysis is rather misleading in this case.

**(4)** The authors have presented a variety of model scores for a number of regions, however, they have not adequately discussed the discrepancies in the modelled and observed AOD patterns over Europe. What is the reason for underestimating the high AODs, while the low AODs are overestimated? How does the AOD underestimation in sea areas compare with PMCAMx ability to model sea salt?

This is an interesting point that clearly needs additional discussion. Based on the monthly average AERONET observations (see Figure 5b) there is no indication that the model underestimates the high AODs and overestimates the low ones. On average, it does a reasonable job in both. However, when one examines the individual measurements (Figure 6) the range of the measurements exceeds that of the predictions. There are a number of possible reasons for this behavior that is often encountered in chemical transport models. The use of the same anthropogenic emissions inventory every day (with the exception of weekends) is one reason. These emissions do vary from day to day, however the model uses their average missing in the process both ends of the actual air pollution distribution. Measurement uncertainty is a second reason. This will also tend to extend the range of the measured AOD distribution compared to the predicted one. Errors in meteorological fields can also contribute to this discrepancy. Please note that the omission of the high coarse PM periods from the evaluation data set has also eliminated the high sea-salt concentration periods. As a result, the present work offers little insight about the ability (or lack there-of) of PMCAMx to model sea salt (see also our response to Comment 2 above). We have added this discussion to the revised paper.

Specific remarks:

**(5)** Page 4, l 17 - change EUCAARI intensive to EUCAARI intensive campaign.

We have made the correction.

**(6)** Page 5, lines 9-12 "The major new methodological improvement in this effort is the screening of the satellite retrievals for periods with high dust (or coarse particles in general) concentrations as well as the combination of the MODIS and AERONET datasets so that the conclusions can be more robust." - Unclear, please restate.

We have rephrased this complicated sentence.

**(7)** Page 6, lines 22-24. The authors say that they use gas phase anthropogenic emissions from GEMS inventory and carbonaceous aerosols from EUCAARI. However, the anthropogenic emissions also have a non-carbonaceous component, and based on Kuenen et al. (2014) this is not negligible for PM<sub>2.5</sub> in Europe. Was the non-

carbonaceous fraction of primary anthropogenic PM ignored in the model computations?

This is a good point. The non-carbonaceous anthropogenic emissions used in this study have been taken from the GEMS inventory so they are not ignored. This information has been added to the model description section.

**(8)** Page 7, line 6. IS4FIRES provides PM<sub>10</sub> emissions. How was this speciated to PMCAMx species and sizes?

The size and composition distribution used is based on Andreae and Merlet (Global Biogeochem. Cycles, 15, 955–966, 2001). We assume that approximately 75% of the emissions are in the PM<sub>2.5</sub> fraction and the rest in the coarse fraction. The average composition (the particles consist mostly organic aerosol and BC) is also based on Andreae and Merlet (2001). This information has been added to the revised paper.

**(9)** Page 8, lines 16-18. How is the aerosol size used in the Mie computations? Is the mean bin diameter used or is there an integration over the size bin?

The mean bin diameter is used for the Mie computations. We have evaluated the accuracy of this simplification by performing detailed calculations over the full diameter range assuming uniform mass or number distributions in each bin. In all cases examined, the differences in the estimated AOD were at most a few percent justifying our simplification. This information has been added to the paper.

**(10)** Page 11, line 2 - What is the explanation, why only 0.4% of the MODIS AODs get discarded over water due to dust influence, while for AERONET this fraction was much higher and very similar to land?

This is mostly due to the fact that a lot of the periods with high dust levels are also accompanied by cloud cover over water. As a result there are no MODIS AOD retrievals during these periods thus lowering the corresponding fraction that needs to be discarded due to dust influence. The location of the AERONET stations also contributes to this discrepancy. A sentence making this point has been added to the manuscript.

**(11)** Page 11, line 23 - Page 12, line 4. Replace the number of data points with the number of stations and flights.

We have added the information about the number of stations (four) and number of flights (sixteen) to the paper. We have kept the number of points to stress that the evaluation was performed based on hourly measurements (for the ground stations) and 5 min averages (for the flights).

**(12)** How does the model bias at ground stations compare with the bias at higher altitudes and how this is expected to influence the modelled AOD?

The model biases for organics and sulfate, the two major PM<sub>1</sub> components, were quite similar at the ground and higher altitudes (Fountoukis et al., 2011). For example for organic aerosol the mean bias was  $-0.4 \mu\text{g m}^{-3}$  in both cases while for sulfate it was  $+0.1 \mu\text{g m}^{-3}$  at the ground and  $-0.1 \mu\text{g m}^{-3}$  aloft. The model reproduced well the almost zero ground PM<sub>1</sub> nitrate levels in the Eastern Mediterranean (Finokalia) (mean bias  $0.02 \mu\text{g m}^{-3}$ ) and the moderate levels in Central Europe (Melpitz) (mean bias  $-0.1 \mu\text{g m}^{-3}$ ). In the high ammonium nitrate region based on the Cabauw measurements the nitrate bias at the ground was  $+0.8 \mu\text{g m}^{-3}$ . The mean bias at higher altitudes was  $-0.2 \mu\text{g m}^{-3}$ . This information has been added to the paper. In general, we do not

expect any complications because of this issue given the relatively small biases in most cases.

**(13)** The authors should discuss the discrepancies in the modelled and observed AOD patterns. Currently major differences are visible between the model and MODIS maps on Figure 3, which are not adequately addressed in the paper. What is the reason for underestimating the high AODs in England, Northern Italy, Balkans region and southern part of the Eastern Europe region, while the low AODs are overestimated in Scandinavia, Northern Russia and Central Europe? Does the AOD underestimation in Atlantic, Mediterranean and Black Sea reflect underestimations of sea salt? The temporal and spatial correlations of the model with the MODIS and AERONET observations should also be reported, as was already requested by referee #2.

We have followed the reviewer's suggestion and extended the discussion of the discrepancies between the predicted and observed values. We have included the results of the comparisons of the EMEP measurements in the corresponding discussion. The ability of the model to reproduce the spatial pattern of the measured AODs is shown in Figure 5 of the original paper in which the station-average AODs are compared. The maps of Figure 3 also illustrate this comparison in the form of a map. We have added in the revised paper the corresponding correlation coefficients for completeness. Comparisons of the diurnal average AOD profiles in several AERONET stations are shown in the Supplementary Information.

**(14)** Figure 4 - if possible, redraw with colour scale which would clearly distinguish between the over- and underestimations and accurate predictions (e.g. white around zero).

We have redrawn this figure following the reviewer's suggestion.

**(15)** Page 14, lines 16-17, Figure 5 - are the error envelopes computed for the monthly average values?

We now clarify that the error envelopes shown in Figure 5 are those discussed in the end of Section 3.

**(16)** Page 16, lines 22-24. Fountoukis et al. (2011) reports about as large negative absolute bias for organic aerosol in Finokalia as for sulphates. That should also contribute to the AOD underestimation.

The concentration of sulphate in Finokalia was more than twice that of organic aerosol. Given the difference in hygroscopicity between the two, most of the AOD (more than 80%) is due according to PMCAMx to sulfates. However, the reviewer is right that the organics are probably making a contribution to the AOD underestimation. We have added this point to the discussion.

**(17)** Page 18, line 14. As AOD at all other seas is also underestimated, could sea salt be underestimated in PMCAMx?

This underestimation is clearly possible. Unfortunately the predicted concentrations over the western Atlantic are heavily influenced by the boundary conditions used for the left side of our modeling domain. Any underestimation of these boundary conditions could also explain the small underestimation of the AOD in this area. As a result, we would prefer not to speculate on the accuracy of the sea-salt predictions of the model.

(18) Page 18, lines 16-17. Model absolute bias is a better statistic than the fractional bias when explaining the AOD underestimation on monthly level.

We agree with this point and we have added the mean bias ( $-0.5 \mu\text{g m}^{-3}$ ) for sulfate which does help explain the AOD underestimation in this area.

(19) Page 19, line 12. Please change to "Sulfates were the major fine PM components predicted in the Black Sea region during the simulation period."

We have rephrased this sentence following the reviewer's suggestion.

(20) Page 20, lines 12-14. When increasing the particle size, was the particle number kept constant? I would expect that when mass concentrations would be kept constant, AOD would decrease noticeably.

We now clarify that the number of particles was not kept constant during this sensitivity test. The lack of sensitivity to particle size for typical ambient size distributions is discussed in detail by Pilinis et al. (Sensitivity of direct climate forcing by atmospheric aerosols to aerosol size and composition, J. Geophys. Res., 100, 18739-18754,1995). The reference and a brief discussion have been added to the paper.

(21) Page 20, lines 15-18. Usually when discussing the BC mixing state, the BC core is assumed to get coated with a scattering layer, which scatters the solar radiation towards the absorbing core, thus increasing both the particle absorption and scattering. Neither of the cases presented here (internal volume averaged mixture and external mixture) take into account that option.

Our initial test of the sensitivity of the AOD in this area to the BC mixing state considering the two models (external mixture and homogeneous internal mixture) showed that the AOD was not sensitive to this assumption. These represent relative extreme cases. The results are reasonable given the relatively low levels of BC and the high levels of secondary inorganic and organic aerosol during this period. The reviewer is right that one can add the case of the core-shell model to the discussion. However, given the low sensitivity the expected changes will be of the order of 1% or so. We have added this point to the revised paper.

(22) Page 21, line 10. I would strongly discourage the authors from using the word "excellent" when describing their results, while discrepancies between the model predictions and observations are clearly visible on Figure 3 for majority of the regions they mention.

We have defined the performance characterization "excellent" in parenthesis as absolute fractional bias less than 15% and fractional error less than 35%. To avoid confusion we have placed the term in quotes. We have also defined "good" and "average" and we believe that under these conditions these terms can be used. Please also note that this performance only applies to specific regions in the domain and not in the regions where major differences exist.

#### Extra references

Kuenen, J.J.P., Visschedijk, A. J.H., Jozwicka, M., Denier van der Gon, H. A. C., 2014. TNO-MACCII emission inventory: a multi-year (2003-2009) consistent high-

resolution European emission inventory for air quality modelling. *Atmos. Chem. Phys. Discuss.* 14, 5837–5869. doi:10.5194/acpd-14-5837-2014.

## Responses to Reviewer #2

(1) In the interests of keeping the peer review process moving forwards, I have decided to submit a review of this paper while also continuing as the Handling Topical Editor. Unfortunately the second anonymous referee for this round of review has not been able to submit their review in a timely manner.

We do appreciate the extra effort by the editor to expedite the review of this manuscript.

(2) The manuscript of Panagiotopoulou et al. is currently in its second round of review. The anonymous reviewers of the original submission suggested several revisions, many of which the authors have implemented, and which have improved the paper. As the anonymous reviewer in this second round points out, however, not all of the recommendations from the first round have been implemented in the revised version of the manuscript. The main outstanding points are 1) the limited nature of the AOD comparison, focusing on monthly averages with no assessment of spatial or temporal correlation, and 2) the speculative nature of the explanations offered for model/measurement agreement and discrepancies.

We have done our best to address both of these points as explained below, under the specific comments.

(3) Regarding the first point, the addition of a paragraph at the bottom of page 19 is a welcome step towards evaluation of the temporal performance of the model. It is not clear exactly what is shown in Figures S3 and S4 though. I presume that these are average diurnal cycles over the whole month of May 2008, right? If so this should be explicitly stated. In any case this extra analysis falls short of the request of the anonymous reviewer in the first round of review to see a spatial and temporal correlation analysis of the comparison between the simulated and measured AOD. This request has been repeated by the anonymous referee from the second round of review.

We now explicitly state that these are the diurnal averages of the measured and predicted AODs. The ability of the model to reproduce the spatial pattern of the measured AODs is shown in Figure 5 of the original paper in which the station-average AODs are compared. The maps of Figure 3 also illustrate this comparison in the form of a map. We have added in the revised paper the corresponding correlation coefficients for completeness. Additional comparisons of the diurnal average AOD profiles in 24 AERONET stations are now shown in the revised Supplementary Information. Given the fact that a lot of the average profiles are relatively flat the correlation coefficient do not convey much useful information and we believe that the actual figures are a lot more informative about the ability of PMCAMx to reproduce the average diurnal variation (or lack thereof).

(4) Regarding the second point, it is not enough to simply state that the model has been evaluated against surface measurements in a previous publication, then leave it at that. Anonymous referees from both rounds of review have called for a more integrated analysis involving comparison of the modelled aerosol composition at the surface with surface measurements from monitoring stations, such as those operated by EMEP. I also think that such an analysis would help alleviate the speculative nature of the discussion of modelled aerosol composition in this paper.

We have compared the predictions of PM<sub>2.5</sub> and PM<sub>1</sub> of PMCAMx against the corresponding EMEP daily measurements in 27 stations. A total of 795 data points have been used for the evaluation. The model showed very little bias (fractional bias equal to -0.07) and reasonable scatter (fractional error equal to 0.49). The average predicted PM<sub>2.5</sub> concentration was 9.07 µg m<sup>-3</sup> while the average observed was 9.82 µg m<sup>-3</sup>. This performance is quite similar to the one reported by Fountoukis et al. (2011) for the EUCAARI stations and airborne campaign for the same period. A summary of this intercomparison has been added to Section 4 and the detailed results have been added to the Supplementary Information (Tables S4 and S5). Unfortunately, there are very few additional EMEP fine PM composition measurements during the simulation period (almost all of them have been already used in the evaluation), so we have to restrict the comparison to the numerous fine PM concentration values.

(5) This is particularly important for Section 5.2. To start with, when discussing the "major components" (eg. line 17, page 15) of PM, please be explicitly clear in each case whether you are referring to the major components as simulated by the model, or measured, and if this refers to measurements, please give a reference. If your discussion of aerosol composition in each region does not currently include any comparison with measured aerosol composition, please compare your modelled composition with surface measurements wherever they are available.

We have rephrased, when necessary, Section 5.2 clarifying that we are referring to the simulated components. We also mention the corresponding ground measurements of composition and mass in the discussion of each region.

(6) There are only two references made in Section 5.2 to the previous evaluations of Fountoukis et al. It would help a great deal if results from these previous evaluations could be integrated better into your analysis. Please describe in the introduction to the paper in more detail what was evaluated and where in these previous studies, and refer back to this explicitly from Section 5.2 when discussing simulated aerosol composition in relevant regions.

We have added details about the previous evaluation in the introduction and also added the corresponding additional information in the discussion of each section.



# 1 Comparison of PMCAMx aerosol optical depth 2 predictions over Europe with AERONET and MODIS 3 measurements 4

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15 **Abstract.** The ability of the chemical transport model (CTM) PMCAMx to reproduce aerosol optical  
16 depth (AOD) measurements by the Aerosol Robotic Network (AERONET) and the Moderate  
17 Resolution Imaging Spectroradiometer (MODIS) over Europe during the photochemically active period  
18 of May 2008 (EUCAARI campaign) is evaluated. Periods with high ~~dust-dust~~ dust-dust or sea-salt levels are  
19 excluded so the analysis focuses on the ability of the model to simulate the mostly secondary aerosol  
20 and its interactions with water. PMCAMx reproduces the monthly mean MODIS and AERONET AOD  
21 values over the Iberian Peninsula, the British Isles, central Europe, and Russia with fractional bias less  
22 than 15% and fractional error less than 30%. However, the model overestimates the AOD over northern  
23 Europe most probably due to an overestimation of organic aerosol and sulfates. On the other end,  
24 PMCAMx underestimates the monthly mean MODIS AOD over the Balkans, the Mediterranean, and  
25 the South Atlantic. These errors appear to be related to an underestimation of sulfates. Sensitivity tests  
26 indicate that the evaluation results of the monthly mean AODs are quite sensitive to the relative  
27 humidity (RH) fields used by PMCAMx, but are not sensitive to the simulated size distribution and the

1 black carbon mixing state. The screening of the satellite retrievals for periods with high dust (or coarse  
2 particles in general) concentrations as well as the combination of the MODIS and AERONET datasets  
3 leads to more robust conclusions about the ability of the model to simulate the secondary aerosol  
4 components that dominate the AOD during this period.

5

## 6 **1 Introduction**

7 Atmospheric aerosols are suspensions of solid and/or liquid particles in air that scatter and  
8 absorb light. The aerosol optical depth (AOD) is defined as the integrated extinction coefficient over the  
9 entire atmospheric column and is a measure of the total aerosol loading (King et al., 1999;  
10 Kokhanovsky, 2008; Vijayarachavan et al., 2008; Hidy et al., 2009). Calculations of AOD require  
11 knowledge of the aerosol vertical profile, including the particulate matter size distribution, chemical  
12 composition, and microphysical state (Seinfeld and Pandis, 2006).

13 Aerosol properties can be retrieved from ground-based measurements as well as from satellite  
14 earth observations (Holben et al., 1998; Levy et al., 2007a, b; Kokhanovsky, 2008; Levy et al., 2010;  
15 Duncan et al., 2014; Hu et al., 2014). Global observations of high spatial coverage are provided by  
16 satellites (King et al., 1999; Vijayarachavan et al., 2008; Hidy et al., 2009) and more limited spatial  
17 coverage by ground-based stations. Regarding temporal coverage, satellite observations are sparse when  
18 compared against ground measurements. Ground-based measurements of AOD are direct measurements  
19 while satellite AOD measurements are indirect, resulting from inversion procedures and exhibiting  
20 larger uncertainties. The magnitude of the satellite AOD uncertainties is higher over land where the  
21 surface reflectance cannot be neglected and it must be retrieved simultaneously with the aerosol  
22 properties (Levy et al., 2007a, 2007b, 2010). The satellite inversion procedure is simpler over water  
23 since the surface contribution is small and the detected signal is mostly due to aerosol reflectance (Shi et  
24 al., 2011; Anderson et al., 2013; Schutgens et al., 2013).

1 Chemical transport models (CTMs) are valuable tools for the study of the impact of pollutant  
2 emissions, the development of air quality improvement strategies, studies of aerosol radiative forcing,  
3 visibility, and global climate change. Uncertainties of the CTM's input data, including meteorological  
4 fields, emission inventories, and boundary conditions as well as weaknesses in representation of  
5 atmospheric processes may lead to weak model performance (Kinne et al., 2003, 2006). CTMs have  
6 been used in the past to provide AOD predictions either globally (Chin et al., 2002, 2004; Lee et al.,  
7 2010; Johnson et al., 2012; De Meij et al., 2012; Pozzer et al., 2012; Yu et al., 2012) or over specific  
8 regions like Asia (Han et al., 2010; Park et al., 2011), United States (Roy et al., 2007), and Europe  
9 (Jeuken et al., 2001; Hodzic et al., 2006; Meij et al., 2007; Tombette et al., 2008; Myhre et al., 2009;  
10 Carnevale et al., 2011; Im et al., 2014). Model evaluation often relies on in-situ ground measurements  
11 but also measurements from airborne platforms. These in-situ measurements cover by necessity a  
12 limited part of the modeling domain. Comparisons against remote sensing data have been used to close  
13 that gap.

14 Jeuken et al. (2001) compared the TM3 CTM AOD predictions with the ATSR-2 radiometer  
15 AOD retrievals during a 1997 summer episode over Europe. Model errors (neglecting organics and  
16 mineral aerosol) in the vertical distributions of sulfate, ammonium, and nitrate, in the hygroscopic  
17 growth, and in the optical parameters led to an average AOD (at 550 nm) underestimation by 0.17-0.19.  
18 Hodzic et al. (2006) used the CHIMERE model to simulate AOD at 865 nm over Europe during August  
19 2003. The model generally reproduced AOD within a factor of 2 and with correlation coefficients  
20 ranging from 0.4 to 0.6 in comparison with POLDER and AERONET. Sporadic aerosol emissions due  
21 to forest fires or dust events led to regional AOD underestimations. De Meij et al. (2007) used the  
22 mesoscale TAPOM model to investigate AOD over Milan, Italy during June 2001. Simulated and  
23 observed AODs by AERONET, MODIS and MISR (Multi-angle Imaging Spectroradiometer) differed  
24 by a factor of 2 or 3 in days with cirrus clouds and Saharan dust but showed good agreement in clear

1 sky days. A finer model resolution gave a more detailed AOD distribution pattern and improved by 15%  
2 the agreement with the AOD observations. Tombette et al. (2008) compared the Polair3D estimated  
3 AOD against AERONET measurements over Europe for 2001. The black carbon (BC) mixing state had  
4 almost no effect on the estimated single scattering albedo (SSA) but the aerosol water content  
5 influenced significantly both the SSA and the AOD. Myhre et al. (2009) used the global Oslo CTM2 to  
6 predict AODs at 550 nm focusing on specific European regions (Adriatic Sea, Black Sea, and Po  
7 Valley). Comparisons against AOD measurements from AERONET, MODIS, and MISR were  
8 presented for a short period during late summer-early autumn of 2004. The model underestimated AOD  
9 around Venice against AERONET because of organic carbon underestimation. Carnevale et al. (2011)  
10 implemented the TCAM CTM to simulate AODs during 2004 over Italy. In general, TCAM was found  
11 to underestimate MODIS AODs. Analysis of the extinction coefficient showed that the submicron  
12 inorganic aerosol played a key role. Im et al. (2014) simulated air pollution over Europe using the  
13 WRF-CMAQ modeling system for 2008. The model underestimated AERONET AOD measurements  
14 by 3-22% on average. AOD underestimations were attributed to underestimation of either the  
15 anthropogenic emissions or the natural and re-suspended dust emissions.

16 The PM<sub>1</sub> composition predictions of PMCAMx have been evaluated over Europe for the May  
17 2008 EUCAARI intensive [campaign](#) (Fountoukis et al., 2011). The model performance was evaluated  
18 against ground measurements which were taken at stations located in the Netherlands, Greece, Ireland,  
19 and Germany as well as against airborne measurements from 15 flights in North-Western Europe. More  
20 than 94% of the organic aerosol (OA) hourly values and more than 82% of the sulfate ones were  
21 reproduced within a factor of 2. PMCAMx performance against airborne measurements was as good as  
22 its performance against the hourly ground measurements.

23 One of the limitations of the previous AOD-based CTM evaluation exercises is that errors in  
24 dust emissions, transport, and removal often dominate the overall results. In the present work MODIS

1 | and AERONET AODs are filtered to exclude periods with high dust or sea-salt levels and to focus on  
2 | the rest of the anthropogenic and biogenic aerosol components. A period with high photochemical  
3 | activity is selected so that the emphasis is on secondary aerosol components. In this work we exclude  
4 | for each locations periods characterized by high coarse particle concentrations, so PM<sub>1</sub> is the  
5 | appropriate metric for aerosol composition evaluation.

6 | In the present study we provide a first time evaluation of the ability of PMCAMx (Murphy and  
7 | Pandis, 2009; Fountoukis et al., 2011) to reproduce AOD observations over Europe. The objective of  
8 | this work is to identify weaknesses and strengths of PMCAMx and its inputs, by taking advantage of the  
9 | wide spatial coverage of MODIS and the temporal coverage of AERONET. The major new  
10 | methodological improvement in this effort is the screening of the satellite retrievals for periods with  
11 | high dust (or coarse particles in general) concentrations as well as the combination of the MODIS and  
12 | AERONET datasets. This combined with the high photochemical activity of the period allows us to  
13 | focus on the ability of the model to predict secondary inorganic and organic aerosols and their  
14 | interactions with water. so that the conclusions can be more robust. The May 2008 period was chosen  
15 | for two reasons. First it coincides with the EUCAARI campaign focusing on a photochemically active  
16 | period with summertime-like conditions. Detailed continuous measurements of PM<sub>1</sub> composition both at  
17 | the ground and aloft as well as a corresponding emission inventory (prepared by TNO) exist for that  
18 | period. The second reason was that the ability of PMCAMx to reproduce these detailed PM<sub>1</sub>  
19 | composition measurements has already been evaluated in previous work (Fountoukis et al., 2011; 2014)  
20 | and therefore we can focus on the optical properties of the fine particulate matter in this paper. The  
21 | exact dates simulated here were the same as in the previous publications for consistency.

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## 1 **2 PMCAMx description**

2 PMCAMx is a three-dimensional CTM that employs the framework of CAMx (Environ, 2003)  
3 simulating the processes of horizontal and vertical advection, horizontal and vertical dispersion, wet and  
4 dry deposition as well as gas, aqueous, and aerosol-phase chemistry. Three detailed aerosol models are  
5 employed: inorganic aerosol growth (Gaydos et al., 2003; Koo et al., 2003), aqueous-phase chemistry  
6 (Fahey and Pandis, 2001) as well as OA formation and chemical aging (Murphy and Pandis, 2009). The  
7 specific modules utilize a sectional approach that dynamically models the evolution of the aerosol size  
8 distribution. Ten size sections covering particle diameters from 40 nm to 40  $\mu\text{m}$  are used. The model  
9 simulates the composition of each size section and therefore predicts the size-resolved PM composition  
10 using in this application 10 size bins. PMCAMx calculates the aerosol number from the corresponding  
11 mass distribution while its sister model, PMCAMx-UF, simulates both the aerosol number and mass  
12 distributions explicitly. Both primary and secondary organic PM is treated as semivolatile and  
13 photochemically reactive employing the volatility basis set (Murphy and Pandis, 2009). Additional  
14 details about the model can be found in Fountoukis et al. (2014), Tsimpidi et al. (2011), and Fountoukis  
15 et al. (2011).

16 The PMCAMx European modeling domain in this application is a region of 5,400 x 5,832  $\text{km}^2$   
17 with 36x36  $\text{km}^2$  grid resolution and 14 vertical layers extending up to approximately 6 km. The  
18 considered period is May 2008 (EUCAARI campaign). Simulations were performed on a polar  
19 stereographic map projection. Horizontal wind components, vertical diffusivity, temperature, pressure,  
20 water vapor, clouds, and rainfall were provided by the Weather Research and Forecasting  
21 meteorological model (WRF) (Skamarock et al., 2008). We used hourly meteorological data from WRF  
22 as input to PMCAMx. WRF was driven by static geographical data as well as dynamic meteorological  
23 data (near real time and historical data that were generated by the Global Forecast System at  $1 \times 1^\circ$ ). In  
24 the vertical dimension 27 sigma-p layers up to 0.1 bars were employed. Each PMCAMx layer is aligned

1 with the WRF layers. WRF was periodically (every 3 days) reinitialized in order to increase the  
2 accuracy of the meteorological input fields to PMCAMx. Anthropogenic gas ~~emissions and inorganic~~  
3 ~~aerosol emissions~~ are from the GEMS European ~~emissions~~ ~~missions~~-database, while elemental carbon  
4 and organic carbon emissions are from the EUCAARI Pan European Carbonaceous Aerosol Inventory  
5 (Kulmala et al., 2011). This carbonaceous aerosol inventory was derived from the IIASA's GAINS  
6 inventory (Klimont et al., 2002; Kupiainen and Klimont, 2004) by application of source specific  
7 elemental carbon and organic carbon fractions. Details about the development of the EUCAARI TNO  
8 emissions can be found in Visschedijk et al. (2007) and Kulmala et al. (2011). Biogenic emissions were  
9 calculated by the MEGAN v2.04 model (Guenther et al., 2006). The marine aerosol emission model  
10 developed by O'Dowd et al. (2008) was employed for the estimation of mass fluxes for both  
11 accumulation and coarse mode, including the organic aerosol fraction. Emissions from wildfires are  
12 taken from IS4FIRES (Sofiev et al., 2009) and the size and composition distribution used is based on  
13 Andreae and Merlet (2001). Approximately 75% of the emissions are in the PM<sub>2.5</sub> fraction and the rest  
14 in the coarse fraction. Details about the development of the EUCAARI TNO emissions can be found in  
15 Visschedijk et al. (2007) and Kulmala et al. (2011).

16 One baseline model simulation for May 2008 was performed together with a number of  
17 additional sensitivity tests described in subsequent sections. Given that the initial conditions are quite  
18 uncertain and dominate the model predictions during the first few days, we have excluded the  
19 corresponding "start-up" period (first six days) from the model evaluation. Concentrations of the major  
20 PM<sub>2.5</sub> aerosol components at the boundaries of the domain (Table S1 in Supplementary Information) are  
21 based on measurements of typical background concentrations in sites close to the domain boundaries  
22 (Zhang et al., 2007; Seinfeld and Pandis, 2006). All concentrations given here are under ambient  
23 temperature and pressure conditions.

24

## 25 **2.1 AOD prediction by PMCAMx**

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1 The size and chemically resolved concentrations of aerosol particles are simulated by PMCAMx  
 2 for every computational cell. Inorganic aerosol water concentration is calculated online by the  
 3 thermodynamic equilibrium model ISORROPIA (Nenes et al., 1998). Taking into account all the  
 4 vertical layers, we calculate the PMCAMx AOD at 550 nm as the sum of the extinction coefficients at  
 5 each layer:

$$6 \quad \text{AOD} = \sum_{i=1}^{14} b_{ext,i} \Delta z_i \quad (1)$$

8 where  $b_{ext,i}$  is the extinction coefficient of layer  $i$  and  $\Delta z_i$  is the corresponding layer thickness. Assuming  
 9 that the particles are homogeneous spheres and that all particles in each size bin have the same  
 10 composition (internal mixture), the aerosol extinction coefficient ( $b_{ext,i}$ ) for layer  $i$  is:

$$11 \quad b_{ext,i} = \sum_{j=1}^{10} \frac{\pi D_j^2}{4} N_j Q_{ext,j}(m_j, D_j) \quad (2)$$

12 where  $D_j$  is the mean diameter of size bin  $j$  and  $Q_{ext,j}$  is the extinction efficiency of a single particle  
 13 having a complex refractive index  $m_j$ .  $N_j$  is the aerosol number concentration for bin  $j$  calculated  
 14 according to:

$$15 \quad N_j = \frac{6c_j}{\pi p_j D_j^3} \quad (3)$$

16 where  $c_j$  is total concentration of all aerosol chemical components and  $p_j$  is the aerosol average density  
 17 at size bin  $j$ . The extinction efficiency for bin  $j$  is estimated as the sum of the scattering,  $Q_{scat,j}$ , and  
 18 absorption,  $Q_{abs,j}$  efficiencies:



$$Q_{ext,j} = Q_{scat,j} + Q_{abs,j} \quad (4)$$

Aerosol scattering and absorption efficiencies ( $Q_{scat,j}$ ,  $Q_{abs,j}$ ) are calculated using Mie theory (Seinfeld and Pandis, 2006) and mass concentrations provided for each size bin by PMCAMx, including the concentrations of particulate water. The mean bin diameter is used for the Mie computations. We have evaluated the accuracy of this simplification by performing detailed calculations over the full diameter range assuming uniform mass or number distributions in each bin. In all cases examined, the differences in the estimated AOD were at most a few percent justifying our simplification. The complex refractive,  $m_j$ , index of a homogeneous sphere is estimated using the volume weighted average of the individual refractive indices (Piliinis and Pandis, 1995). Sulfate and ammonium are assumed to have a real refractive index of 1.53, which is the value of ammonium sulfate (GEISA, 2011; NASA, 2006). Nitrate is assumed to have a real refractive index of 1.56, similar to the value of ammonium nitrate (NASA, 2006). Sodium and chloride have a real refractive index of 1.5 (GEISA, 2011; NASA, 2006). Dust is assumed to have a complex refractive index of 1.53-0.0055i (GEISA, 2011). OA is assumed to be non-absorbing with a refractive index of 1.5 (Nessler et al., 2005; Fierz Schmidhauser et al., 2010). Biomass burning was minimal during the period of interest (Crippa et al., 2014), so this simplifying assumption regarding the OA absorptivity has little effect on the predicted AOD. The black carbon refractive index has the largest uncertainties (Bond and Bergstrom, 2005) and we use a value of 1.75-0.44i (GEISA, 2011). In the base case BC is assumed to be internally mixed with the other components in each size range. The sensitivity of the model predictions to this assumption is discussed in a subsequent section. Biomass burning emissions in Europe during the simulated period were low and therefore any effects from biomass burning related brown carbon are also expected to be small.

### 3 MODIS and AERONET data

The cloud screened and quality assured Level 2 AERONET direct AOD measurements are used for the PMCAMx evaluation. AERONET applies the Beer Lambert Bouguer law to measure AOD from

1 direct sun observations (Holben et al., 1998) therefore it is considered to be the ground truth with AOD  
2 uncertainties of 0.01 – 0.02 (Eck et al., 1999). The AERONET measurements have a variable temporal  
3 resolution which is on average around 15 min. Measurements start after sunrise when the sun is  
4 approximately 7.5 degrees above the horizon and end a little before sunset when the sun is once more at  
5 approximately 7.5 degrees. We use here the AERONET AOD at 550 nm. In this work only the AOD  
6 values corresponding to Angstrom Exponent values greater than 0.9 are employed in an effort to  
7 exclude periods with high dust or high sea-salt levels (Schuster et al., 2006). This filter rejected 29% of  
8 AODs over land and 28% over water. The geographical distribution of the corresponding AERONET  
9 stations is depicted in Fig. 1 and the number of stations in each region is shown in Table 1. Some  
10 AERONET stations in the domain of interest did not have available Level 2 AOD data for the period of  
11 interest while all data from three stations (OHP\_OBSERVATOIRE in South France, FORTH\_CRETE  
12 in Crete, Greece, and ATHENS\_NOA in Athens, Greece) have been excluded after the ~~dust~~-coarse  
13 particle (dust or sea-salt) rejection filtering.

14 The polar-orbiting MODIS monitors global aerosol properties from two satellites: Terra and  
15 Aqua (Salomonson et al., 1989). MODIS employs 36 channels from 0.412 to 14.2  $\mu\text{m}$ , has a wide swath  
16 of 2,330 km, and observes every location of the globe at least once daily. The default resolution for  
17 aerosol retrieval is  $10 \times 10 \text{ km}^2$  (Levy et al., 2009). Each data set retrieved by MODIS is associated with  
18 a Quality Assurance Confidence (QAC) flag which ranges from 0 (no confidence) to 3 (highest  
19 confidence). For increased spatial coverage we use both the Terra and Aqua MODIS AOD retrievals  
20 with  $\text{QAC} \geq 1$ . We employ the MODIS Level 2 Collection 5.1 aerosol datasets. The Dark-Target  
21 algorithm products were used. We did not alter the values of the data records and we did not apply any  
22 additional transformations. The MODIS AOD values, retrieved with spatial resolution  $10 \times 10 \text{ km}^2$ , were  
23 assigned to the corresponding computational cells of the PMCAMx modeling domain. AOD retrievals  
24 are provided at seven wavelengths (470, 550, 660, 870, 1,200, 1,600, 2,100 nm) over water surface and

1 four wavelengths (470, 550, 660, 2,100 nm) over land. In this study we focus on the 550 nm values.  
2 Figure 2 presents the geographical distribution of the available MODIS AOD measurements during the  
3 period of interest 1-29 May 2008 (EUCAARI campaign) over Europe. The average number of retrievals  
4 is  $12 \pm 9$ . The maximum number of retrievals is 65 in areas in the North Atlantic.

5 Dust emissions from the Sahara are not included in the PMCAMx emissions used here and the  
6 focus of this study is on periods and regions in which Saharan dust or other coarse particles like sea-salt  
7 ~~does~~ not contribute significantly to the AOD. To exclude periods with high coarse particledust levels  
8 and to focus on the rest of the anthropogenic and biogenic aerosol components, MODIS AODs are  
9 filtered. Over water we employ the ~~dust~~ coarse particle rejection filter of Barnaba and Gobbi (2004).  
10 According to this filter, AOD values greater than 0.3 also corresponding to coarse mode fraction higher  
11 than 0.3 are assumed to be coarse particledust-influenced periods. Over land we only use the AOD  
12 values which correspond to Angstrom Exponent values exceeding 0.9 (Schuster et al., 2006). The above  
13 filters discard 16% of MODIS AOD values over land and 0.4% over water. This is mostly due to the  
14 fact that a lot of the periods with high dust levels are also accompanied by cloud cover over water. As a  
15 result there are no MODIS AOD retrievals during these periods thus lowering the corresponding  
16 fraction that needs to be discarded due to dust influence. The location of the AERONET stations also  
17 contributes to this difference. ———▲

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18 The evaluation of the MODIS AODs at 550 nm for the land algorithm was performed following  
19 the approach of Remer et al. (2005) and Levy et al. (2007b). Fig. S1 presents a comparison of the  
20 corresponding AERONET observations with the MODIS AOD retrievals AERONET measurements  
21 were spatially and temporally collocated with MODIS retrievals, similar to the scheme proposed by  
22 Ichoku et al. (2002). The collocated data were sorted according to the AERONET AOD observations.  
23 The resulting data were partitioned into groups of 100 AOD points and then averaged. At higher optical  
24 depths since the data became sparser we used 25 points for each bin. The regression line of the

1 collocated AODs, prior to partitioning, had a slope of 1.05. 73% of the 8,331 collocated points fall  
2 within the expected error envelope. These results indicate that the mean MODIS AOD over land in the  
3 region and period of interest was retrieved with the expected accuracy. The highest quality flag QAC =  
4 3 provides the closest match, but including the QAC = 2 and 1 retrievals results in only a minor  
5 reduction of accuracy while increasing significantly the size of the dataset (Table S2).

6 Previous studies have shown that MODIS AOD retrievals have an expected error of  $\pm(0.05 +$   
7  $0.15AOD_{AERONET})$  over land and  $\pm(0.03 + 0.05AOD_{AERONET})$  over water (Chu et al., 2002; Remer et al.,  
8 2005; Levy et al., 2007a,b, 2010; Anderson et al., 2013). Table S3 summarizes the values of the  
9 expected MODIS AOD uncertainties for the various regions in our modeling domain during May 2008,  
10 based on the monthly mean values of AERONET AOD. The MODIS-AERONET AOD differences for  
11 this period are consistent with the expected uncertainty of the MODIS retrievals (Fig. S1).

12

#### 13 **4 Evaluation of PMCAMx fine PM composition and mass predictions**

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14 Fountoukis et al. (2011) evaluated the ability of PMCAMx to simulate the chemical  
15 composition of PM<sub>1</sub> components during the same period simulated in this study (May 2008) using the  
16 measurements of the intensive campaign of European Aerosol Cloud Climate and Air Quality  
17 Interactions (EUCAARI) project (Kulmala et al., 2011). The model predictions were compared with  
18 hourly averaged AMS ground measurements as well as airborne measurements over Europe (Morgan et  
19 al., 2010). The measurements covered Central Europe, England and Ireland, North Atlantic and the  
20 Mediterranean. Approximately 8500 measurements (data points) from four ground stations and sixteen  
21 flights were used in this evaluation.

22 PMCAMx predictions were in close agreement with the AMS measurements at Cabauw for all  
23 species. The predicted monthly average concentrations for OA, nitrate, sulfate and ammonium were 4.0,  
24 3.2, 2.2 and 1.9  $\mu\text{g m}^{-3}$  respectively compared to the measured average of 4.1, 2.5, 1.5 and 1.7  $\mu\text{g m}^{-3}$ .

1 The model reproduced approximately 90% of the hourly PM<sub>1</sub> OA data within a factor of 2. At Finokalia  
2 the average predicted concentration was 2.1 µg m<sup>-3</sup> for OA, 4.7 µg m<sup>-3</sup> for sulfate, 0.09 µg m<sup>-3</sup> for  
3 nitrate and 1.3 µg m<sup>-3</sup> for ammonium in comparison with the AMS measurements of 2.5, 5.2, 0.08, and  
4 1.5 µg m<sup>-3</sup>, respectively. At Mace Head PMCAMx reproduced 79% and 74% of the hourly PM<sub>1</sub> OA and  
5 sulfate hourly measurements within a factor of 2. However, greater errors were seen for PM<sub>1</sub> nitrate and  
6 ammonium, because of the bulk equilibrium assumption used in that PMCAMx application. In Melpitz  
7 the model reproduced more than 80% of the hourly PM<sub>1</sub> OA data within a factor of 2. Overall,  
8 PMCAMx agreement with the AMS ground measurements for all stations was encouraging. More than  
9 70% of the hourly data points for PM<sub>1</sub> sulfate and 87% for PM<sub>1</sub> OA lay within the 2:1 and 1:2 error  
10 lines. As expected, the model performance based on daily averaged values was even better, reproducing  
11 94% and 82% of the hourly data within a factor of 2 for OA and sulfate, respectively. Overall the model  
12 fractional bias for the ground stations was -0.1 for OA, 0.1 for sulfate, 0.2 for ammonium and 0.4 for  
13 nitrate. For the airborne measurements, the PMCAMx fractional bias was -0.2 for OA, 0.2 for sulfate, -  
14 0.3 for nitrate and -0.08 for ammonium.

15 PMCAMx predictions of the vertical distribution of sub-micron aerosol chemical composition  
16 were evaluated against the airborne AMS data. Both PMCAMx and LONGREX airborne observations  
17 showed low OA concentrations in the 2-6 km altitude range over Europe during the simulation period.  
18 The ability of the model to reproduce the high time resolution airborne measurements at various  
19 altitudes and locations was similar to its ability to simulate the ground level concentrations. PMCAMx  
20 reproduced almost 70% of the sulfate and OA concentrations within a factor of 2. For measured sulfate  
21 and OA higher than 1 µg m<sup>-3</sup>, the model reproduced 77% and 75% of the corresponding measurements,  
22 respectively, within a factor of 2.

23 A detailed evaluation of the ability of PMCAMx to reproduce observations of the organic  
24 aerosol composition for the same May 2008 period has been presented by Fountoukis et al. (2014). The

1 PMCAMx predictions using the Volatility Basis Set approach were compared against AMS positive  
 2 matrix factorization results. The model correctly predicted the low concentrations of fresh primary  
 3 transportation-related OA ( $<0.3 \mu\text{g m}^{-3}$ ) at Melpitz and Finokalia. At Mace Head it showed a small  
 4 tendency towards underprediction of the same component with a mean error of  $-0.25 \mu\text{g m}^{-3}$ . Overall, in  
 5 the comparison of PMCAMx against AMS hydrocarbon-like OA (HOA) measurements from all  
 6 stations, the mean error was  $0.26 \mu\text{g m}^{-3}$  while the mean bias was less than  $1 \mu\text{g m}^{-3}$ . Regarding  
 7 oxygenated OA (the major component of OA according to the measurements in all stations) the model  
 8 reproduced 83% of the measured values within a factor of 2. The model biases for organics and sulfate,  
 9 the two major PM<sub>1</sub> components, were quite similar at the ground and higher altitudes (Fountoukis et al.,  
 10 2011). For example for organic aerosol the mean bias was  $-0.4 \mu\text{g m}^{-3}$  in both cases while for sulfate it  
 11 was  $+0.1 \mu\text{g m}^{-3}$  at the ground and  $-0.1 \mu\text{g m}^{-3}$  aloft. The model reproduced well the almost zero ground  
 12 PM<sub>1</sub> nitrate levels in the Eastern Mediterranean (Finokalia) (mean bias  $0.02 \mu\text{g m}^{-3}$ ) and the moderate  
 13 levels in Central Europe (Melpitz) (mean bias  $-0.1 \mu\text{g m}^{-3}$ ). In the high ammonium nitrate region based  
 14 on the Cabauw measurements the nitrate bias at the ground was  $+0.8 \mu\text{g m}^{-3}$ . The mean bias at higher  
 15 altitudes was  $-0.2 \mu\text{g m}^{-3}$ .

16 The predictions of PM<sub>2.5</sub> and PM<sub>1</sub> of PMCAMx can also be compared against the corresponding  
 17 EMEP daily average mass concentration measurements. A total of 795 data points in 26 ground stations  
 18 are available during the simulated period and have been used for the evaluation. The model showed  
 19 very little bias (fractional bias equal to  $-0.07$ ) and reasonable scatter (fractional error equal to  $0.49$ ). The  
 20 average predicted PM<sub>2.5</sub> concentration was  $9.07 \mu\text{g m}^{-3}$  while the average observed was  $9.82 \mu\text{g m}^{-3}$ .  
 21 This performance is quite similar to the one reported by Fountoukis et al. (2011) for the EUCAARI  
 22 stations and airborne campaign for the same period. Details about this intercomparison can be found in  
 23 the Supplementary Information.

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## 5 Evaluation of PMCAMx AOD predictions

The ~~coarse particledust~~-screened monthly mean AODs for Europe during May 2008 retrieved by MODIS and predicted by PMCAMx are shown in Fig. 3. The PMCAMx AODs have been calculated for exactly the same periods as the MODIS retrievals to allow the direct comparison of the two. The comparisons with the MODIS AOD retrievals correspond exactly in space and time, so the times coincide with the satellites' overpasses.

The MODIS retrievals show high AOD values ( $> 0.25$ ) over England, South Ireland, North Italy, South Poland, East Romania, Greece, and North Atlantic. Low AOD values ( $< 0.1$ ) were retrieved over East France, Belgium, Sweden, and North Russia. PMCAMx predicts high AODs over England, South Ireland, North Italy, and central Atlantic and low AODs over North Sweden, East Russia, North and South Atlantic. The data sample size is small over North Africa due to the high levels of dust in these areas during the whole simulation period. As a result the corresponding ~~coarse particledust~~-screened AOD comparisons provide little information about the ability of PMCAMx to simulate fine PM in this region.

### 5.1 Overall evaluation

The difference between PMCAMx and MODIS monthly mean AODs is depicted in Fig. 4. PMCAMx AODs are higher than those of MODIS over England, Ireland, France, Germany, central and South Italy, North and East Europe, central, North and West Russia, West Balkans, and central Atlantic. On the other hand PMCAMx predicts lower AODs than MODIS over parts of Russia, North Italy, central and South Balkans, South Poland, North and South Atlantic, and the African coast of the

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1 Mediterranean. On a domain average basis PMCAMx predicts an AOD equal to 0.14 while MODIS  
2 retrieved 0.16. Detailed comparisons for each region can be found in Table 2. 94% of the monthly mean  
3 AOD values fall inside the expected MODIS error envelope over land (Fig. 5a). Over the whole domain  
4 the PMCAMx monthly mean AODs have a mean error of 0.05 and a fractional bias of -16% compared  
5 to the MODIS monthly mean AODs (Tables 2 and S4). The correlation coefficient  $R$  between the  
6 MODIS monthly-average AODs and the PMCAMx predicted AODs was 0.51.

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7 PMCAMx AODs were also compared with the AERONET values for the simulation period.  
8 Once more the comparisons were done for the grid cells of the AERONET stations and corresponding  
9 measurement periods. The PMCAMx monthly mean AODs had a mean error of 0.03 and a fractional  
10 bias of 4% compared to the AERONET monthly mean AODs (Table 1). The comparison of the  
11 PMCAMx with AERONET monthly mean AODs is summarized in Fig. 5b for the 50 AERONET  
12 stations which are employed in the present study. The correlation coefficient  $R$  between the AERONET  
13 monthly-average AODs and the PMCAMx predicted AODs was 0.57.

## 15 5.2 Regional evaluation

16 The performance of the model for AOD combined with its performance for composition in the sites  
17 where there are ground and airborne PM composition measurements, can be used to reach some  
18 tentative conclusions about its performance in reproducing the fine PM levels and composition. These  
19 are clearly limited to the components dominating the AOD in each area and either suggest problems or  
20 lack of major errors. These are discussed for each region below.

21  
22 *Spain and Portugal:* The relatively low AOD levels (0.11 for the 8 AERONET stations and 0.14 for  
23 MODIS) are reproduced well by PMCAMx (0.12 for the AERONET sites and 0.12 for the periods of  
24 the MODIS retrievals). The monthly mean PMCAMx AOD predictions have a mean error of 0.02



1 (AERONET) and 0.04 (MODIS) (Tables 1 and 2). The model shows little bias (5%) compared to the  
2 AERONET stations and a small tendency towards underprediction (-15%) compared to MODIS. 83%  
3 of the monthly mean PMCAMx AODs are within the expected MODIS error envelope. These results  
4 are consistent with the evaluation of PMCAMx against the daily ground PM<sub>2.5</sub> mass measurements in  
5 eight stations in Spain (Niembro, Campisabalos, Cabo de Creus, Barcarotta, Zarra, Penausende, Els  
6 Torms, O Savinao) in which the fractional bias of the model is -0.04 and the fractional error is 0.52  
7 (Table S6). Sulfate and organic aerosol are the predicted major components of dry fine PM in Spain  
8 and Portugal (Table 3).

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9  
10 *Russia, Belarus, and Ukraine:* PMCAMx reproduces well (0.14 predicted versus 0.15 measured) the  
11 average AOD observations at the 5 AERONET stations in this region (2 in West Russia, 1 in Belarus, 1  
12 in Ukraine, and 1 in Crimea) (Table 1). The model has a similar good performance against the MODIS  
13 retrievals (0.12 predicted versus 0.13 retrieved) (Table 2). As a result, the monthly mean PMCAMx  
14 AOD predictions have a low mean error of 0.02 (AERONET) and 0.04 (MODIS). PMCAMx shows a  
15 slight tendency towards underprediction (-11%) compared to AERONET and no bias (<1%) compared  
16 to MODIS. 92% of the monthly mean PMCAMx AODs are within the expected MODIS error envelope.  
17 Sulfates and organic aerosol are predicted to predominate in this region and it appears that PMCAMx  
18 performs reasonably well in this ground-level measurement poor region. Significant discrepancies  
19 between predicted and observed AOD over Russia were expected given the uncertainty in the  
20 corresponding emissions. However, the agreement was quite good with both AERONET and MODIS.  
21 This rather surprising result clearly requires additional investigation and could be due to offsetting  
22 errors.

23  
24 *United Kingdom (UK) and Ireland:* This area was relatively polluted during the simulation period with  
25 high levels of nitrates, sulfates, and organic aerosol based on both PMCAMx predictions and the

1 | airborne measurements (Table 3). PMCAMx reproduces the relatively high average MODIS (0.23  
2 | predicted versus 0.21 retrieved) and AERONET (0.24 predicted versus 0.25 measured in the station of  
3 | Chibolton). The monthly mean PMCAMx AODs have a mean error of 0.04 compared to MODIS with a  
4 | small tendency towards overprediction (14%). 90% of the monthly mean PMCAMx AODs fall within  
5 | the expected MODIS error envelope. The encouraging agreement of PMCAMx retrievals over this  
6 | region is consistent with its good performance when compared against the EUCAARI airborne and  
7 | ground measurements in this region (Fountoukis et al., 2011; 2014) for the major fine PM components.

8 |  
9 | *Balkans:* The Balkans according to PMCAMx had some of the highest sulfate levels in the domain  
10 | during the simulation period (Table 3). The model underpredicts the AOD both against MODIS (0.14  
11 | predicted versus 0.19 retrieved) and the two AERONET stations (0.15 predicted versus 0.21 measured).  
12 | The corresponding fractional biases are -24% against MODIS and -33% against AERONET. However,  
13 | 80% of the monthly mean PMCAMx AODs fall within the expected MODIS error envelope. These  
14 | results are consistent with the PM<sub>2.5</sub> mass concentration underprediction (fractional bias -24%) in the  
15 | station of Iskrba in Slovenia (Table S6). Given that most of the predicted AOD is due to the sulfate  
16 | these results suggest that the PMCAMx underprediction is probably due to their underestimation.

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17 |  
18 | *Central Europe:* PMCAMx showed a small tendency towards overprediction of the moderate AODs in  
19 | this region compared to both AERONET (12%) and MODIS (13%). For example, overpredictions were  
20 | evident over France and Germany (Fig. 4). The corresponding fractional errors on a monthly average  
21 | basis were 22% against AERONET and 30% against MODIS. Organic aerosol, sulfate, and nitrate and  
22 | organic aerosol were the major predicted fine PM components in central Europe during this period  
23 | consistent with the measurements in Melpitz and Cabauw (Fountoukis et al., 2011). PMCAMx  
24 | overpredicted sulfate levels (fractional bias equal to 0.3) and nitrate levels (fractional bias of 0.3) in  
25 | Cabauw, but showed little bias (equal to 0.01) for OA. These results appear consistent with the AOD

1 overpredictions. However, in Melpitz the model slightly underpredicted sulfate (fractional bias= -0.1)  
2 and underpredicted OA (fractional bias= -0.3). Using the airborne measurements over this region,  
3 sulfate was overpredicted (fractional bias of 0.2) and OA was underpredicted (fractional bias of -0.2).  
4 These results are also consistent with the ground PM<sub>2.5</sub> mass concentration measurements in six stations  
5 in the area (Illmitz, Payerne, Rigi, Weldoff, Schaunisland, and Ispra) in which the absolute fractional  
6 biases are less than 35% and the fractional errors less than approximately 50% (Table 6). There higher  
7 PM<sub>2.5</sub> errors in the site of Montelibretti in Rome, but these are mostly due to the coarse resolution of the  
8 model. These results suggest that the sulfate overpredictions can probably explain to some extent the  
9 AOD overpredictions. Errors in the relative humidity fields could also explain parts of these AOD  
10 discrepancies.

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12 *East Europe:* PMCAMx slightly overpredicted the AODs in this region compared to both AERONET  
13 (24%) and MODIS (25%). 82% of the monthly mean PMCAMx AODs fell within the expected MODIS  
14 error envelope. PMCAMx predicts more frequently AODs > 0.1 than measured by AERONET during  
15 the corresponding period of measurements probably because of an overestimation of sulfates and  
16 organic aerosol. The ground PM<sub>2.5</sub> measurements in the two sites in Latvia (Rucava and Zoseni) are the  
17 highest in the domain and are seriously underpredicted by PMCAMx (Table S6) and inconsistent with  
18 the low to moderate MODIS AODs in the same area (Figure 3). The reasons for these high fine PM  
19 measurements are not clear and need additional investigation.

Μορφοποιήθηκε: Δείκτης

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21 *North Europe:* PMCAMx reproduces the low pollution levels in this area with a mean AOD of 0.12  
22 compared to 0.08 by the 4 AERONET stations. The absolute monthly mean errors are low: 0.04  
23 (AERONET) and 0.06 (MODIS). However, there is significant fractional positive bias compared to  
24 both AERONET (36%) and MODIS (47%). 54% of the monthly mean PMCAMx AODs fall outside the  
25 expected MODIS error envelope. 53% of the hourly PMCAMx AODs are greater than 0.1 while only

18% of the AERONET values are greater than 0.1. A similar overprediction of the PM<sub>2.5</sub> concentrations can be seen in several stations in the area (Asprveten, Hyytiala, Lille Valby) while in some other stations (Rao and Vavihill) there is little or no overprediction (Table S6). Sulfates and organic aerosol are the dominant predicted fine PM components in this region and the model probably overestimates at least one of them in this region.

*Turkey and Northern Africa:* There are only two AERONET stations in this area and PMCAMx underpredicts by 30% the corresponding moderate AOD measurements. However, the model performance appears to be much better against the MODIS retrievals covering a much bigger area and the underprediction drops to 13%. 79% of the PMCAMx AODs fall inside the expected MODIS error envelope. Sulfates and organic aerosol are the major fine PM components according to PMCAMx in these regions and they are probably slightly underestimated by the model.

*Mediterranean Sea:* PMCAMx exhibits a tendency towards underprediction (-24%) against MODIS and 58% of the monthly mean PMCAMx AODs fall inside the expected MODIS error envelope. The major discrepancies are evident in the southern part of the Mediterranean especially close to the African coast. These suggest that dust may be partially responsible for the errors even after the filtering of the data. The model performance is better in the eastern Mediterranean (Fig. 4). Sulfates dominated the AOD in the Mediterranean during the simulation period according to PMCAMx. ~~PMCAMx predicted PM<sub>1</sub> sulfate with practically zero fractional bias in Finokalia in Crete in this region (Fountoukis et al., 2011).~~ Both the organic aerosol and the sulfate were underpredicted in the site of Finokalia in Crete with a mean bias of approximately -0.5  $\mu\text{g m}^{-3}$  (Fountoukis et al., 2011). However, the concentration of sulfate in Finokalia was more than twice that of organic aerosol. Given the difference in hygroscopicity between the two, most of the AOD (more than 80%) is due according to PMCAMx to sulfates. The

1 omission of the high coarse PM periods from the evaluation data set has also eliminated the high sea-  
2 salt concentration periods from areas over water. As a result, the present work offers little insight about  
3 the ability (or lack there-of) of PMCAMx to model sea salt in the various marine environments  
4 examined here.

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6 *South Atlantic:* The PMCAMx AOD predictions are significantly lower (-45%) compared to the  
7 MODIS retrievals in this region with 74% of the monthly mean PMCAMx AODs falling outside the  
8 expected MODIS error envelope. Sulfate and sea-salt dominated the predicted AOD in this region in  
9 May 2008 and there is evidence that they may be underpredicted. However, errors in relative humidity  
10 or cloud contamination could be also responsible for these discrepancies (Anderson et al., 2013). The  
11 predicted concentrations over the western Atlantic are heavily influenced by the boundary conditions  
12 used for the left side of our modeling domain. Any underestimation of these boundary conditions could  
13 also explain the underestimation of the AOD in this area.

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16 *North Atlantic:* The model performance is much better in the North than in the South Atlantic. The  
17 mean AOD error is 0.04 compared to MODIS with a tendency towards underprediction (-21%). 53% of  
18 the monthly mean PMCAMx AODs fall inside the expected MODIS error envelope. There is one  
19 AERONET station in this area (in Helgoland around 50 km from the coast of Germany) and PMCAMx  
20 predicts an average AOD equal to 0.16 compared to the 0.11 measured. Sulfates, organic aerosol, and  
21 sea-salt were the major predicted fine PM components in North Atlantic during May 2008 (Table 3).  
22 PMCAMx performed relatively well (absolute fractional bias less than 0.2 for both sulfate and OA)  
23 when compared with the EUCAARI airborne measurements in this region.

1 *Black Sea*: PMCAMx exhibits a tendency towards underprediction (-18%) versus MODIS in this  
2 relatively polluted region. 66% of the PMCAMx AODs fall within the expected MODIS error envelope.  
3 Sulfates were the major predicted fine PM component ~~s~~ in the Black Sea during the simulation period.

4  
5 The results of the PMCAMx-MODIS comparison for the various regions are summarized in Fig.  
6 6. These results suggest that the variability of the MODIS retrievals exceeds that of the PMCAMx  
7 predictions for almost all areas. Based on the monthly average AERONET observations (see Figure 5b)  
8 there is no indication that the model underestimates the high AODs and overestimates the low ones. On  
9 average, it does a reasonable job in both. This could be due to However, when one examines the  
10 individual measurements (Figure 6) the range of the measurements exceeds that of the predictions.  
11 There are a number of possible reasons for this behavior that is often encountered in chemical transport  
12 models. The use of the same anthropogenic emissions inventory every day (with the exception of  
13 weekends) is one reason. These emissions do vary from day to day, however the model uses their  
14 average missing in the process both ends of the actual air pollution distribution. Measurement  
15 uncertainty is a second reason. This will also tend to extend the range of the measured AOD distribution  
16 compared to the predicted one. Errors in meteorological fields can also contribute to this discrepancy.  
17 Other potential contributors to this discrepancy is the relatively coarse ~~the~~ spatial resolution of the  
18 model inputs, ~~including the emissions, but also to other reasons including~~ missing short-term air  
19 pollution sources in the inventory, potential cloud contamination of the retrievals, etc.

20 The AERONET measurements provide an additional opportunity to test the ability of the model  
21 to reproduce the observed average diurnal AOD variation, at least for the approximately 12 hours for  
22 which measurements are available. Some of these comparisons are shown in Figures S3 ~~and~~ S84 in the  
23 Supplementary Material. Overall, for over 90% of the hourly averaged AOD measurements the

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1 PMCAMx error was less than 50%, indicating that the agreement of the average AODs was not due to  
2 offsetting temporal errors.

### 3 4 **6 Sensitivity analysis of the predicted AODs**

5 There are various possible sources of bias in the PMCAMx predictions of AOD other than the  
6 concentration and composition of aerosol. We explore here the role of the relative humidity calculated  
7 by the WRF model, the role of the mixing state of BC, as well as that of the predicted aerosol size  
8 distribution.

9 In the first test the absolute humidity was increased uniformly by 5%, while maintaining the  
10 maximum relative humidity in cloud-free regions at 99%. The PMCAMx monthly mean AOD increased  
11 on average by 13% (Fig. S2). The increases ranged from 7% in Turkey and Northern Africa to 31% in  
12 the North Atlantic. This AOD change can explain a significant part of the base case discrepancies which  
13 cause a fractional error of PMCAMx 22% versus AERONET and 33% versus MODIS.

14 In another test the diameter of all particles was increased by 20% without keeping the particle  
15 number constant. 72% of the PMCAMx monthly mean AOD values changed by less than 0.01. The  
16 average increase of the monthly mean AOD was 1% (ranging from 0.3% in the Black Sea to 4% in the  
17 UK and Ireland). This small sensitivity of the aerosol forcing to moderate changes of aerosol size has  
18 been discussed in detail by Pilinis et al. (1995).

19 In a third sensitivity test we assumed that BC was always externally mixed with the other  
20 components in each size range, forming pure BC spheres. 73% of the PMCAMx monthly mean AOD  
21 values changed by less than 0.01 in this test. The average change of the monthly mean AOD was  
22 negligible (< 0.5%). –These two cases represent relative extreme cases of mixing. The results are  
23 reasonable given the relatively low levels of BC and the high levels of secondary inorganic and organic

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1 aerosol during this period. A similarly low sensitivity of the order of 1% or so is expected in the case of  
2 a core-shell model which was not examined in detail.  
3  
4

## 5 **7 Conclusions**

6 Previous evaluations of the ability of the 3-D CTM PMCAMx to reproduce the aerosol levels in  
7 Europe, the US, and Mexico City have been based on comprehensive chemical composition  
8 measurements at a few ground sites and limited data from a few flights. In this study we expand these  
9 efforts by using the MODIS and AERONET retrievals of AOD over Europe during a photochemically  
10 active period (May 2008). We exclude periods during which the different areas are strongly affected by  
11 dust (mainly from the Sahara) or other coarse particles like sea-salt in an effort to focus on the other  
12 primary and secondary anthropogenic and biogenic aerosol components.

13 PMCAMx can reproduce the observed AODs for this period with little bias (-16% for MODIS  
14 and +4% for AERONET). The corresponding fractional errors are 33% against MODIS and 22%  
15 against AERONET. These results are consistent with those of Fountoukis et al. (2011; 2014) who  
16 compared the PMCAMx predictions for the same period against ground measurements of fine PM  
17 composition in four sites and airborne measurements from several flights over central and northern  
18 Europe.

19 The AOD performance of PMCAMx against the MODIS retrievals is ~~excellent~~ “excellent”  
20 (absolute fractional bias less than 15% and fractional error less than 35%) in the Iberian Peninsula,  
21 UK/Ireland, central Europe, Russia-Belarus-Ukraine, Turkey-northern Africa. We were expecting  
22 significant discrepancies between predicted and observed AOD over Russia given the uncertainty in the  
23 corresponding emissions. However, the agreement was quite good with both AERONET and MODIS.  
24 This rather surprising result clearly requires additional investigation and could be due to offsetting



1 | errors. The model performance is “good” (absolute fractional bias less than 25% and fractional error  
2 | less than 35%) in East Europe, the Balkans, and over the Mediterranean, the North Atlantic, and the  
3 | Black Sea. Finally, its performance is “average” (absolute fractional bias less than 50% and fractional  
4 | error less than 55%) in the relatively clean area of North Europe and the South Atlantic. The  
5 | performance is more or less similar against AERONET with the exception of a few areas with only one  
6 | or two AERONET stations. The average performance against the AERONET measurements is  
7 | considered using the above criteria “excellent” and against MODIS it is on the borderline between  
8 | “good” and “excellent”.

9 |       The above results suggest that the major weaknesses of PMCAMx appear to be overpredictions  
10 | of sulfate and/or organics over North and East Europe, underprediction of sulfate over the Balkans, and  
11 | underprediction of fine sodium chloride, sulfates, or organics in the southern Mediterranean and South  
12 | Atlantic. However, these discrepancies are quite sensitive to the relative humidity fields predicted by  
13 | WRF. In a sensitivity test the average predicted AOD increased by 13% (ranging from 7 to 31%  
14 | depending on the area) for a uniform 5% change in RH. On the other hand, the details of the fine PM  
15 | size distribution and the black carbon mixing state have a very small effect on the AOD predictions.

16 |       Comparison of the predicted AOD with the MODIS and AERONET results can shed only  
17 | limited light on the ability of a CTM to reproduce the composition of the aerosol. The performance of  
18 | the model for AOD, combined with its performance for composition in the sites where there are ground  
19 | and airborne PM composition measurements, can be used to strengthen these tentative conclusions  
20 | about its composition performance. These are clearly limited to the components dominating the AOD in  
21 | each area, as discussed above, and either suggest problems or lack of major errors.

22 |

23 | **Code availability**

1 PMCAMx is the research version of the publicly available CAMx (www.camx.org). The Fortran source  
2 code of CAMx (Version 6.20 was posted on March 23, 2015) and a User's Guide both prepared by  
3 ENVIRON can be downloaded through the above website. The PMCAMx code is used as testbed for  
4 testing of different hypotheses, algorithms, etc. The version used in this paper as well as the most  
5 current version can be obtained upon request by contacting Prof. S. Pandis  
6 (spyros@chemeng.upatras.gr).

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γραμματοσειράς; Αυτόματο,  
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Μορφοποιήθηκε:  
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**Table 1.** Error metrics for the evaluation of PMCAMx against AERONET monthly mean AODs.

Region	Number of AERONET stations	Mean AERONET AOD	Mean PMCAMx AOD	Mean Error	Mean Bias	Fractional Error	Fractional Bias
UK/Ireland	1	0.25	0.24	0.01	-0.01	0.04	-0.04
Central Europe	25	0.16	0.17	0.03	0.02	0.22	0.12
North Europe	4	0.08	0.12	0.04	0.04	0.36	0.36
Spain and Portugal	8	0.11	0.12	0.02	0.01	0.20	0.05
East Europe	2	0.11	0.14	0.03	0.03	0.24	0.24
Balkans	2	0.21	0.15	0.06	-0.06	0.33	-0.33
Russia, Belarus, and Ukraine	5	0.15	0.14	0.02	-0.02	0.16	-0.11
Turkey and	2	0.17	0.12	0.05	-0.05	0.30	-0.30

Northern Africa							
Mediterranean Sea	-	-	-	-	-	-	-
North Atlantic Ocean	1	0.11	0.16	0.05	0.05	0.37	0.37
South Atlantic Ocean	-	-	-	-	-	-	-
Black Sea	-	-	-	-	-	-	-
Domain	50	0.15	0.15	0.03	0.001	0.22	0.04

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$$\text{Mean Error} = \frac{1}{N} \sum_{i=1}^N |P_i - O_i| \quad \text{Mean Bias} = \frac{1}{N} \sum_{i=1}^N (P_i - O_i)$$

$$\text{Fractional Error} = \frac{2}{N} \sum_{i=1}^N \frac{|P_i - O_i|}{P_i + O_i} \quad \text{Fractional bias} = \frac{2}{N} \sum_{i=1}^N \left( \frac{P_i - O_i}{P_i + O_i} \right)$$

where  $P_i$  are predicted values by PMCAMx,  $O_i$  the AERONET retrievals and  $N$  the number of stations.

**Table 2.** Error metrics for the evaluation of PMCAMx against MODIS monthly mean AODs.

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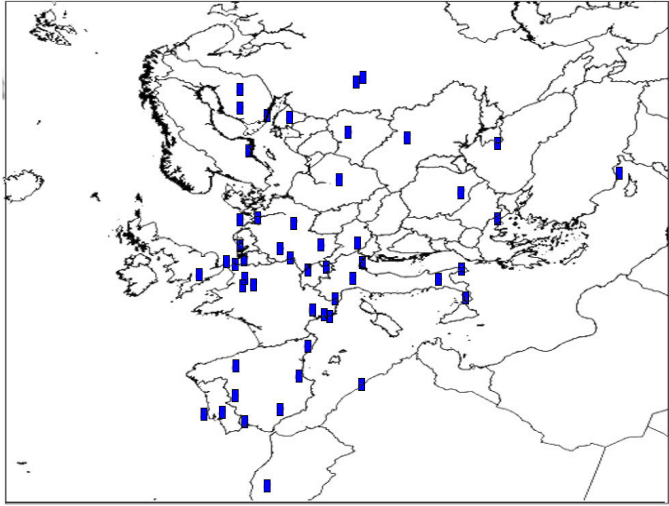
Region	Mean MODIS AOD	Mean PMCAMx AOD	Mean Error	Mean Bias	Fractional Error	Fractional Bias
UK and Ireland	0.21	0.23	0.04	0.02	0.22	0.14
Central Europe	0.16	0.17	0.05	0.01	0.30	0.13
North Europe	0.09	0.14	0.06	0.05	0.53	0.47
Spain and Portugal	0.14	0.12	0.04	-0.03	0.28	-0.15
East Europe	0.13	0.15	0.05	0.03	0.35	0.25
Balkans	0.19	0.14	0.05	-0.04	0.28	-0.24
Russia, Belarus, and Ukraine	0.13	0.12	0.04	0.01	0.30	-0.01
Turkey and Northern Africa	0.16	0.14	0.05	-0.03	0.31	-0.13
Mediterranean Sea	0.18	0.14	0.04	-0.04	0.25	-0.24
North Atlantic Ocean	0.17	0.14	0.04	-0.03	0.30	-0.21
South Atlantic Ocean	0.16	0.10	0.06	-0.06	0.45	-0.45
Black Sea	0.17	0.14	0.04	-0.03	0.22	-0.18
Domain	0.16	0.14	0.05	-0.02	0.33	-0.16

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**Table 3.** Monthly predicted mean ground-level concentration in  $\mu\text{g m}^{-3}$  of the major  $\text{PM}_{2.5}$  components

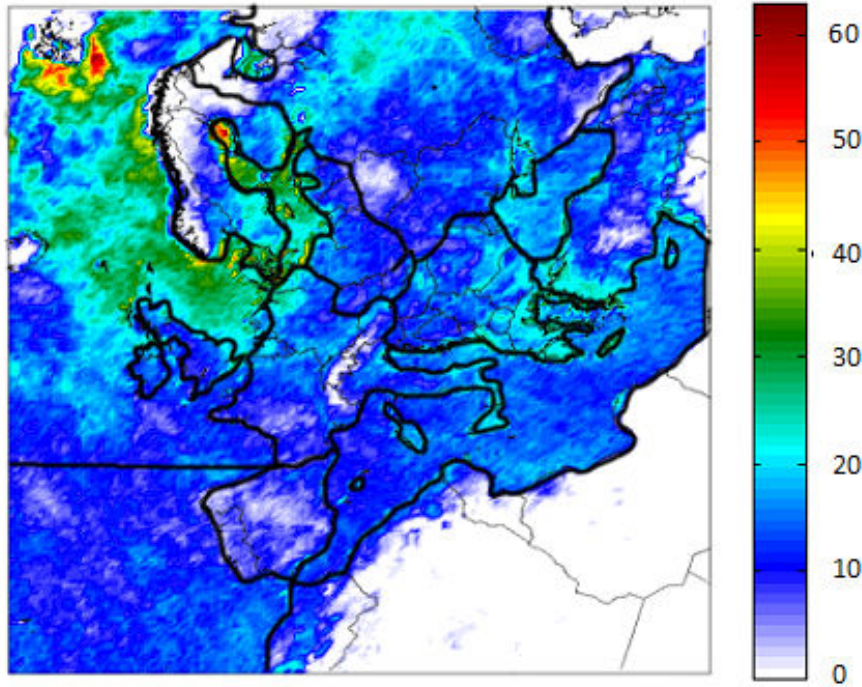
Region	$\text{SO}_4^{2-}$	OA	EC	$\text{Cl}^-$	$\text{Na}^+$	$\text{NH}_4^+$	$\text{NO}_3^-$	$\text{H}_2\text{O}$	Crustal
UK and Ireland	3.6	3.4	0.5	0.6	0.8	2.4	3.8	33.3	0.7
Central Europe	3.0	3.4	0.5	0.2	0.4	1.4	1.3	10.1	0.6
North Europe	2.2	2.3	0.2	0.2	0.4	0.9	0.6	5.1	0.4
Spain and Portugal	1.6	1.2	0.2	0.1	0.2	0.7	0.5	11.7	0.3
East Europe	2.9	3.1	0.4	0.1	0.4	1.2	0.8	9.2	0.6
Balkans	3.9	2.7	0.3	0.1	0.3	1.4	0.3	4.9	0.6
Russia, Belarus, and Ukraine	2.5	2.1	0.3	0.03	0.2	0.9	0.2	3.9	0.5
Turkey and Northern Africa	2.7	2.2	0.2	0.2	0.4	1.0	0.6	8.2	0.5
Mediterranean Sea	4.2	2.4	0.3	1.3	1.4	1.2	0.3	11.3	0.7
North Atlantic Ocean	2.1	1.8	0.2	1.0	1.0	1.0	1.0	24.2	0.4
South Atlantic Ocean	1.5	1.1	0.06	0.8	0.8	0.5	0.3	8.8	0.3
Black Sea	3.8	2.8	0.3	0.6	0.7	1.3	0.4	9.7	0.7
Domain	2.4	1.9	0.2	0.5	0.6	0.9	0.5	10	0.5

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**Figure 1.** Geographical distribution of the 50 AERONET stations used in the present study.

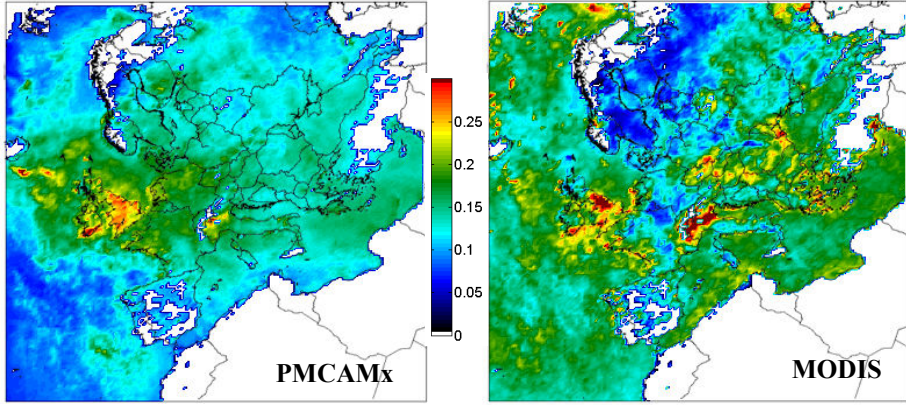
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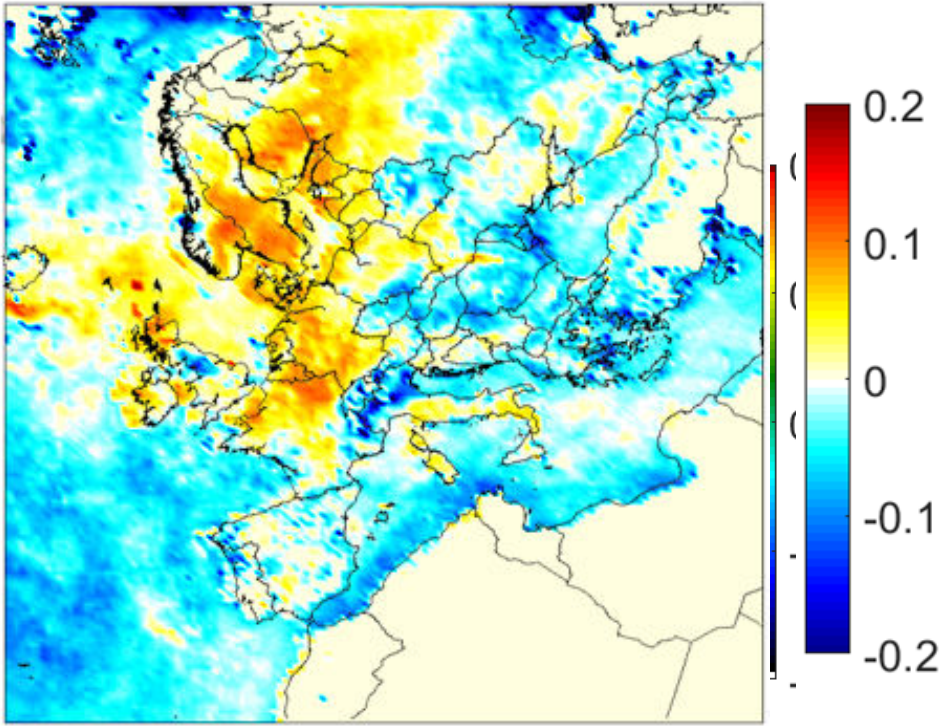
**Figure 2.** Geographical distribution of the number of available AOD retrievals from MODIS over Europe during May 2008. White color denotes no retrievals. Land is partitioned into 8 regions including the United Kingdom and Ireland, central Europe, North Europe, Spain and Portugal, East Europe, Balkans, Russia/Belarus/Ukraine, Turkey, and Northern Africa. The sea is partitioned into 4 regions: the Mediterranean, North Atlantic, South Atlantic, and Black Sea.

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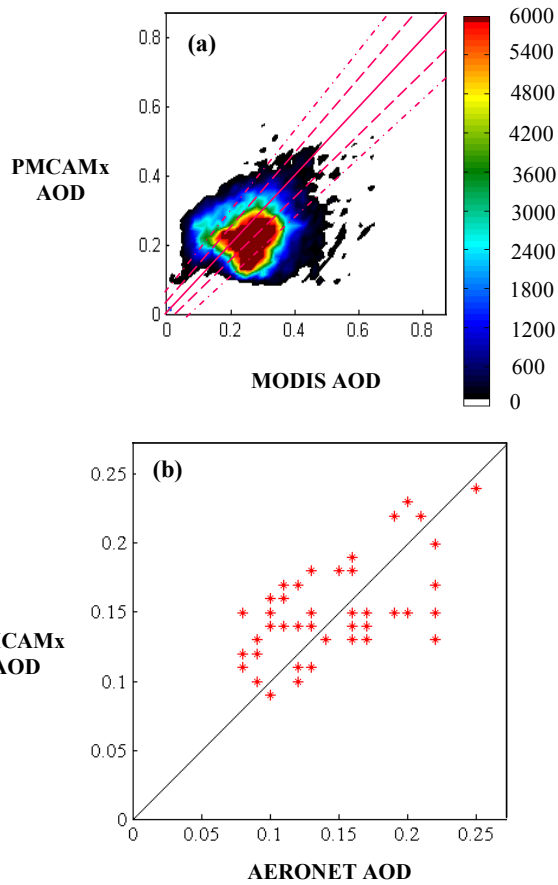
**Figure 3.** Monthly mean AODs from PMCAMx and MODIS ( $QAC \geq 1$ ) during May 2008. White color denotes no AOD retrieval. A coarse particle rejection filter has been employed. The PMCAMx AODs correspond to the periods of the MODIS retrievals.

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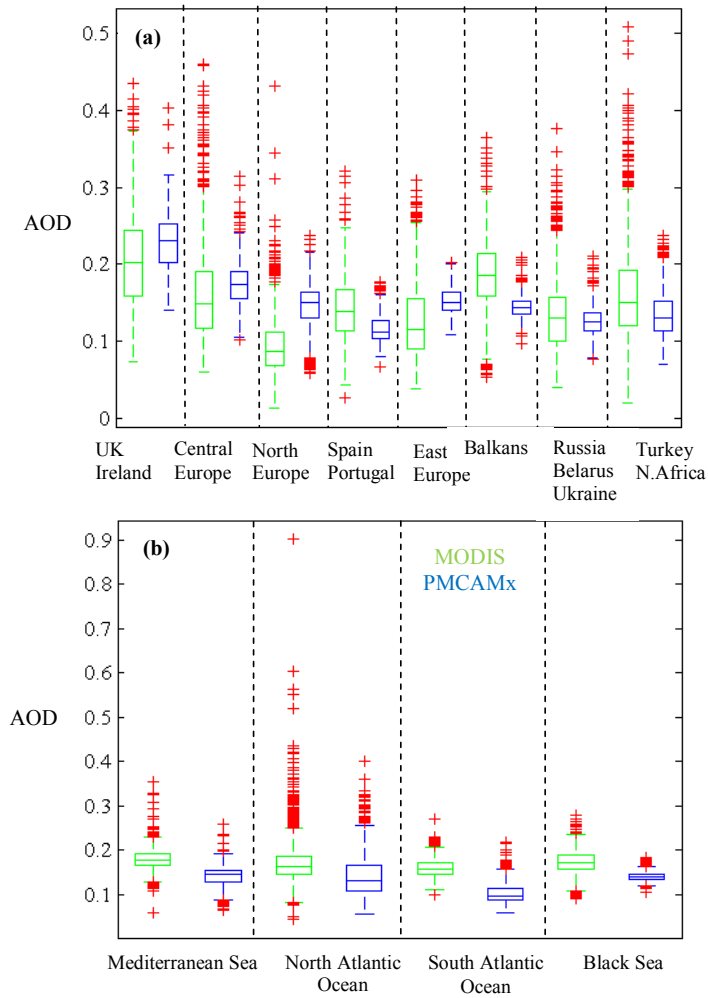


**Figure 4.** Difference of the PMCAMx from MODIS (QAC  $\geq 1$ ) monthly mean AODs during May 2008. Positive means that PMCAMx overpredicts AOD compared to MODIS. There were not enough dust-screened AOD retrievals for the model evaluation in the white areas in Northern Africa and in the Middle East.





**Figure 5. a)** Comparison of the PMCAMx predictions with MODIS ( $QAC \geq 1$ ) monthly mean AODs. The different colors indicate density. The dashed red lines denote the ocean expected error envelope and the dotted lines denote the land envelope which describes MODIS AOD uncertainties with respect to AERONET (see Section 3). The solid red line is the 1:1 line. **b)** Comparison of the PMCAMx with AERONET monthly mean AODs. The PMCAMx values correspond to the periods of measurement for the 50 AERONET stations.



**Figure 6. a)** Box plots of the PMCAMx and MODIS ( $QAC \geq 1$ ) monthly mean AODs for land. The central mark is the median, the edges of the box are the 25th and 75th percentiles, the whiskers extend to the extreme data points considered to be not outliers, and the outliers are plotted individually by the

- 1 red marks. Points are drawn as outliers if they are larger than  $Q3+1.5(Q3-Q1)$  or smaller than  $Q1-$
- 2  $1.5(Q3-Q1)$ , where  $Q1$  and  $Q3$  are the first and third quartiles, respectively. **b)** Box plots of the
- 3 PMCAMx and MODIS ( $QAC \geq 1$ ) monthly mean AODs for water.