

Interactive comment on “Towards an online-coupled chemistry-climate model: evaluation of COSMO-ART” by C. Knote et al.

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R1.0) This paper describes the performance of a regional weather prediction model, COSMO, when it has been coupled on-line with a version of the RADM photochemical mechanism and the MADE / SORGAM treatment for aerosols. Four cases studies, each for a different season, are evaluated with operational surface meteorological, trace gases, and particulate data. The performance of the model is over-stated several places in the manuscript. The title of this manuscript is apt, since there are clearly missing pieces to the model that the authors acknowledge, such as wet removal of gases and aqueous phase chemistry. Aerosol indirect effects are not included as well. While, the title is apt, some of the conclusions regarding the ability of the model to be

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used as a climate model need to be changed since climate-relevant calculations have not been evaluated. In addition, the text does not describe whether COSMO-ART will be coupled with ocean / sea-ice / ecosystem models which are also important components of climate models. While the paper contains a useful evaluation useful for publication in GMD, there first needs to be significant changes to the text as described in the comments below.

We thank the reviewer for the efforts and constructive comments regarding the quality of our manuscript. We have responded to all comments below.

Major Comments:

R1.1) Many of the conclusions regarding the performance of the model are over-stated. Examples are included the specific comments below. Many of these statements are too broad and lack specificity. The authors need to rephrase the text so that better reflects the performance as depicted in the figures. On a positive note, the authors do provide sufficient material so that the reader can judge for themselves how well the model performs.

We have altered the text sections describing the figures to better reflect their content. On multiple occasions we now avoid too generous statements regarding model performance.

R1.2) While the title of the paper is apt, the model has a long way to go before it has all the necessary components needed as part of a climate model. The model does include aerosol direct effects but there is insufficient description on how those processes are treated and an initial evaluation of those processes. The model is only compared to AOD, which is not the same as aerosol direct effect. Aerosol indirect effects apparently have been incorporated but not used in this study. Since this study is trying to demonstrate the utility of using COSMO-ART as a chemistry-climate model, some discussion of the findings of Bangert et al. (2011) is warranted in the present work. The omission of wet removal of trace gases and aqueous processes is problematic for some other

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variables, such as sulfate. It would suggest that the next paper written using COSMOART should essentially repeat the present work, but include the processes that are currently missing in the model.

We have chosen the title with consideration. The goal of this work has not been to evaluate direct / indirect aerosol effects. Rather it is a preparatory study to understand the model's ability to simulate gas and aerosols characteristics which are indispensable parameters to accurately simulate climate feedbacks. We have therefore adapted the title, which now reads: "Towards an online coupled regional chemistry climate model: Evaluation of trace gases and aerosols in COSMO-ART". We have further altered the text of the introduction to better state the fact that this is no evaluation of aerosol climate feedbacks. The sentence "Particular focus is laid on the accuracy of the representation of climate relevant parameters of aerosols, i.e. their optical properties and ability to act as cloud condensation nuclei." was changed to "Through a detailed analysis of aerosol size distributions and chemical composition we set the basis for subsequent analyses of aerosol-climate interactions in COSMO-ART."

We agree with the reviewer that aq.-phase chemistry is a major deficiency and we are working on including this process. A sentence has been added to the abstract to inform the reader that this is still missing (penultimate sentence in abstract): "Suggestions for further improvement of the modeling system consist of the inclusion of a revised secondary organic aerosols scheme, aqueous-phase chemistry and improved aerosol boundary conditions."

Several important modules relevant for the simulation of climate-chemistry interactions are currently being added to the model, including aerosol-cloud interactions (both impact of aerosols on cloud formation and impact of cloud processing on aerosol properties and removal) and coupling to the NCAR Community-Land-Model (CLM) for a more interactive simulation of biogenic VOC emissions but also for a better representation of land-atmosphere processes (a first coupled version was already developed in a master thesis). Coupling to other models like e.g. a sea-ice or ocean model is

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an ongoing effort within the community using COSMO in climate mode (CCLM). Also, several activities are ongoing to make the model computationally more efficient.

Specific Comments:

R1.3) Page 1811, line 8: I am not convinced that the present work is the first regional chemistry model application in Europe. Perhaps I am getting hung up on the word “comprehensive” which could imply that most other studies employ more limited amounts of data. But then where is the line between “limited” and “comprehensive”?

“comprehensive” here refers to the measurements of aerosol chemical composition. The EUCAARI campaign was one of the first campaigns in Europe where measurements with Aerosol Mass Spectrometer have been taken in a coordinated manner at several stations throughout Europe. This is the first time this data is published, so we are the first ones to use it. The sentence in page 1811, line 8, does not imply that our efforts were the “first regional chemistry model application in Europe”.

R1.4) Page 1811, lines 12 and 13: This sentence is self-contradictory. First it says the model reproduces the bulk properties well, but then states there is a clear tendency to underestimate PM mass (itself a bulk property). The first part of this sentence is also vague. It would be useful to be more specific and also be specific on the bias in the PM mass.

The sentence now reads: “Mean, variability and spatial distribution of the concentrations of O₃ and NO_x are well reproduced. SO₂ is found to be overestimated, simulated PM_{2.5} and PM₁₀ levels are on average underestimated, as is AOD.”

R1.5) Page 1811, line 13: It is consistent that an underestimation of PM mass will lead to an underestimation in AOD. However, errors in aerosol composition and size distribution also affect AOD. Simple statements like this tend to mislead the reader into thinking that if predicted PM mass was correct then AOD would be correct for the right reasons.

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We fully agree with this statement since AOD is determined by particles smaller than those dominating PM. The rewritten sentence (R1.4) does not imply that AOD is underestimated due to PM₁₀ / PM_{2.5} mass underestimation.

R1.6) Page 1811, line 21: Please be more specific as to what “simulated well” means. Adding some numbers would help.

These sentences have been rewritten, and now read: “Aerosol number concentrations compare well with measurements for larger size ranges, but overestimations of particle number concentrations by factors of 2 - 5 are found for particles smaller than 50 nm. Size distribution characteristics are often close to measurements, but show discrepancies at polluted sites.”

R1.7) Page 1815, line 24: Since the purpose this manuscript is to demonstrate the ability of the model to simulate climate processes, it would seem very important to present some description of how aerosols affect radiation. For example, how are the aerosol optical properties handled? Has this been done in a previous study and just not cited here? Why not show a plot depicting the impact of aerosols on radiation when compared to AERONET data over Europe? Presumably including aerosols would improve the radiation calculations?

We did not intend to analyse aerosol-(radiation/climate) feedbacks but want to evaluate gas & aerosol characteristics. However, there have also been earlier studies focussing on aerosol direct effects of certain aerosol types: Vogel et al. (2009) and Stanelle et al. (2010). Also the methods to calculate the aerosol optical properties are described in these studies.

R1.8) Page 1816, lines 1-14: The text discusses that MOZART is used to provide the boundary conditions for trace gases, but not aerosols. It is not surprising that aerosol species between MOZART and the present model are different, but it would seem that some sort of mapping could have been made. This would not have been a difficult task.

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It is true that such a mapping could be made, and we actually tested this. However it is not as straightforward as it seems. Delivered are bulk PM mass for several aerosol components but neither size distribution information nor number concentrations are available. Therefore, the aerosol mass below 2.5 μm would have to be estimated somehow. Thereafter individual species need to be mapped and the total mass below 2.5 needs to be distributed to the two different modes (Aitken, Accumulation) and finally number concentrations have to be guessed assuming some diameter. We agree that mapping the species is not very problematic but assumption on the amount of PM_{2.5} mass out of total PM and the calculation of number concentrations are problematic. We have in fact done this mapping, but found e.g. for sulfate aerosols a strong overestimation in the resulting boundary data which lead to unrealistic values. We could not determine the source of this error and did not settle on a reasonable procedure up to now - but it is on the list of future developments.

R1.9) Page 1821, section 3: I found some of the discussion relating to the figures a bit confusing. The problem is that some figures show meteorology, trace gases, and aerosols, but the text first describes the meteorology, trace gases, and aerosols in that order. So the reader needs to go back to previous figures – not a natural progression. This is just an organizational issue and I am not sure the best way to fix this. It would be useful at the beginning of this section to state that some of the figures contain multiple parameters that are not all described at once. That way the reader knows up-front that discussion of the material will come later. Similarly, I was expecting some elaboration on the model results in Section 3, but some of this material was covered later in Section 4. Again some introductory material to Section 3 to explain how the results are presented and discussed would be very useful.

We have added an introductory paragraph to section 3 to better guide the reader: “The following section contains a description of the results of our evaluation efforts, starting with meteorology, then trace gases and finally aerosol characteristics. Each section is accompanied by a figure / table summarizing the results for the species discussed.

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Section 4 then further elaborates on the results for aerosol characteristics.”

R1.10) Page 1821, lines 14 – 22: The authors explain that the free running model simulation performed as well as cases when data assimilation or reinitialization is used. Given the simulation periods (multiple weeks) and large mesoscale modeling domain size, this is hard to believe without some proof. I believe that with only lateral boundary condition forcing, the model can perform qualitatively well but there is likely room for improvement. Significant research and development has been conducted on data assimilation and operational design. The operational community has been arguing for years that data assimilation improves forecasts, so the statements here seem to contradict how operational models are run.

Our intention was not to state the the model performs equally well in a free run as in a nudged run with continuous data assimilation at the accuracy NWP modellers strive to have, and we are well aware of the efforts and evaluations of the operational modeling community. From evaluation against measurements we see however no significant increase in temperature / wind-speed / wind-direction bias over the integration time - something that is not visible from the mean bias figures in the manuscript. Our comparison methods (mean biases / timelines over a raw ensemble of SYNOP station data) are certainly not as elaborate as the detailed and accurate analyses of scientists occupied with NWP forecasts. Another reason why the meteorological variables were in a rather good agreement with the observations although we simulated quite a long period was that we used the reanalysis of the IFS model as boundary conditions. From that perspective we did not perform free forecasts for a timespan of four weeks. Therefore the statement we want to make is: meteorological quality is reasonable for AQ-simulation purposes, but can be improved using data assimilation. This is also stated in the conclusions on page 1843, line 18-19). We also mentioned this in section 3.1, page 1821 lines 15-18. We have slightly adapted the text in section 3.1 to highlight that our comparison is not as sophisticated as for operational verification. The start of section 3.1 now reads: “COSMO-ART is in its meteorological core code identical

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with the NWP model COSMO, and its performance is continuously verified by several European weather services. Meteorological evaluation has therefore been limited in this work to surface parameters. In all periods, the comparison [...]”

R1.11) Page 1821, line 23: A reference seems in order for this statement.

We have added the 2 references also mentioned in the previous paragraph on the same topic: Schluenzen and Katzfey, 2003; Heinemann and Kerschgens, 2005.

R1.12) Page 1821, section 3.1: The meteorological evaluation presented in this study employs only surface stations. Where comparisons with upper-air measurements, such as radiosondes, made? It would seem that boundary layer depth is another critical meteorological parameter needs to be evaluated since this will affect vertical mixing of trace gases and aerosols. I suggest adding some additional analyses using upper-air data. If not, please add text somewhere in the manuscript describing that the model has not been evaluated with data aloft how that would affect conclusions regarding trace gases and aerosols.

No, we have not evaluated aircraft campaigns / radiosonde data. We have added a sentence in the conclusions, end of first paragraph: “Not included in our work has been an evaluation of upper-air variables with aircraft campaigns or radiosonde measurements.” Also we have added a reference to a recent field campaign (COPS) where the meteorological capabilities of COSMO have been extensively evaluated (Barthlott et al. (2011)).

R1.13) Page 1822, line 16: Are the results in Table 2 for the entire period (day and night periods). It would be useful to be specific.

We thank the reviewer for spotting this one. AIRBASE data is 12 - 18 LT, everything else is the entire period. We have added the information to the table.

R1.14) Page 1823, section 3.2.1: Are there any black carbon measurements that can be used to evaluate the model? For direct radiative forcing relevant for climate applica-

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tions, this is a critical component. Even a small amount can have a profound effect on the relative contribution of scattering and absorption in the atmosphere.

We did not compare black carbon as it has not been available to us in the AMS datasets (for almost all of the stations). COSMO-ART contains an explicit description of soot aging (Riemer et al, 2003) and has been evaluated for this component during the implementation of this method. We concede that this evaluation is not extensive and could be added. We will try to find measurement data and include this in a separate reply.

R1.15) Page 1823, lines 25-29: No VOCs measurements are used to evaluate the model in the present study. VOCs are not normally measured routinely, but are often measured during field campaigns. Where there any VOC measurements available during the campaigns mentioned in Section 2.3 for some of the simulation periods? A lack of VOCs affects the interpretation of simulated, but also simulated SOA.

Yes, but VOC data is really limited. Actually we compared measurements in the Swiss midlands (Dübendorf) and total NVMOC is OK there. (This is also stated in the manuscript, page 1823 lines 27 - 29): “A preliminary comparison with total NMVOC measured at Duebendorf (CH) showed good agreement (not shown), which gave confidence that our NMVOC levels are in the correct range, but we could not assess the spatial distribution”) 3 stations (Hohenpeissenberg, Rigi, Jungfrauoch) have “routine” data on single components - but unfortunately all of them are mountain tops / hillsides, and it is not straightforward to compare a mountain top / hillside station to model output. Some VOC measurements have been made during EUCAARI by PTRMS but going deep into a comparison against these observations would require a separate study.

R1.16) Page 1824, section 3.2.2: It is true that the model does qualitatively resemble the spatial distribution in the measurements; however, the number of stations is rather limited in many regions so it is difficult to draw conclusions in portions of the modeling domain, especially over water. The evidence that the ship emissions are too high is

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very slim. The simulated trace gases and aerosols in along the ship tracks may seem unreasonable, but there are no measurements to confirm that conclusion. One would also need to know how well the model simulates mixing within the marine boundary layer.

We agree it is somewhat debatable if this really is the case, but there are two indications supporting this, i) the satellite data (see also table 2 which shows the highest (positive) biases for NO₂ over sea grid points), and measurements at coastal stations (also here - strongest overestimation of NO₂ and SO₂ at coastal stations, i.e. near ship tracks).

R1.17) Page 1825, lines 24-29: I assume the results described here are not shown, and it should be stated somewhere they are not shown.

A note has been added. The sentence now reads: “While NH₃ mean concentrations were comparable to measurements, the diurnal cycles were not (both not shown).”

R1.18) Page 1829, line 1-7: The authors should compare their results with diurnal cycles shown from other PM models, such as McKeen et al, (2007).

We thank the reviewer for this reference. 2 sentences have been added: “McKeen et al. (2007) compared PM_{2.5} measurements with several air quality forecast models in North America and concluded that, while most of the models are able to accurately represent daily average PM_{2.5} concentrations, there are substantial inconsistencies in representing the diurnal cycle. Most models show a negative bias and exaggerate diurnal variability, something we observe also for the (single) rural PM_{2.5} station in our comparison (Fig.5).

R1.19) Page 1829, line 13: Some specificity in the “better performance” would be useful for the reader.

The sentence has been revised and now reads: “Stern et al. (2008) saw better agreement with measurements (i.e. less underestimation) for PM_{2.5} [...]”

R1.20) Page 1830, line 10: I assume the boundary conditions for dust is a large fraction

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here, but I assume this is discussed in Section 4?

We have added a sentence: “At the boundaries only low dust concentrations are prescribed due to the way aerosol boundaries are treated (see Section 2.1).”

R1.21) Page 1831, line 12-13: What about simulated aerosol water and errors in simulated composition that will also affect simulated AOD. AOD is tricky because a model can simulate the values well but for the wrong reasons that are difficult to assess fully.

Thanks to the reviewer for adding this. The role of aerosol water was already discussed, we have added a sentence regarding chemical composition at the end of this paragraph: “Secondly, differences in simulated and real aerosol chemical composition will also have an effect on AOD. The next section addresses a comparison of aerosol chemical composition.”

R1.22) Page 1832, line 11: This sentence states that the simulated PM is reasonable, but then goes on to show that nitrate is too high and OA is too low. Therefore, the model produces the “right answer for the wrong reasons”. This fact needs to be included somehow in this sentence.

We have added this information and the sentence now reads: “At all stations the time evolution of NR-PM1 is represented well by our simulations, sometimes however for the wrong reasons due to a mismatch in chemical composition.”

R1.23) Page 1833: line 4: Need another word for “bursts” – not sure this terminology is correct.

We have changed that to “isolated peaks”.

R1.24) Page 1836, line 20-21: This statement is overly optimistic. There are some stations with relatively large differences. Even subtle differences in the size distribution will have an impact on direct radiative forcing through radiation. Indirect radiative forcing via CCN activation also depends on size distribution. Since this paper is describing a model that ultimately will be used to simulate climate effects, this is pretty important.

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Simulating size distribution is perhaps more difficult for models than simulating total particulate mass, which is a critical parameter for air quality models. Same comment for line 17 on the next page.

This is correct, but the reviewer should take into account that we are comparing 3 weeks of data with a statistical distribution over 3 months (mentioned in the text). Therefore we would not expect a 1:1 agreement. We agree however that the statement should be measured more regarding model performance. We have modified the sentence on page 1836: “Overall, the modelled size distributions are at several stations close to the observed ones.” Further, the first sentence of the mentioned paragraph on line 17, page 1837 now reads: “In general the model has an acceptable representation of the variability of number concentrations between stations [...]”.

R1.25) Page 1839, line 11: Here it is stated that OA is underestimated by a factor of 2, but the difference between observed and simulated OA in the figures looks less than that at first glance.

We state the discrepancy can “reach factors of 2” (page 1839, line 11), which does not presume that this is always the case. The given differences (pie charts / table) are average values and do not show extremes.

R1.26) Page 1839, line 27: Have the authors looked at satellite-derived fire emission products? They could establish whether a significant number of fires occurred within the modeling domain for their simulation periods. This could establish how important this source could have been for the simulated trace gases and aerosols.

We did not analyse that. Inclusion of fire emissions from the GFED products is underway and will be addressed in a future study.

R1.27) Page 1840, line 4: The authors discuss work on emissions that will be included in the future. What about work to better represent the mix of POA and SOA that is also described in this paragraph?

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This will also be topic of the further developments of the volatility basis set SOA module (currently in the works at KIT Karlsruhe).

R1.28) Page 1842: line 6: Here it is stated that the nucleation rates are too high, but earlier on page 1835 the authors show aerosol number too high and attribute that to primary emissions of small particles and discuss that the Kerminen and Wexler (1994) parameterization produces a low fraction of new particles. Most nucleation parameterizations produce too few particles, and this parameterization would be consistent with previous studies. So, I am a bit confused by the statement on page 1842 when compared to earlier discussion.

The current approach in COSMO-ART will probably (almost surely) underestimate new particle formation in the atmosphere, due to the Kerminen and Wexler parameterization. However, the used primary emission scheme, where SO₄ mass is instead emitted in Aitken mode, will move the same amount of SO₄ directly to approximately one order of magnitude higher diameters (although with lower overall number concentration). This has two effects: 1) the particles, which would have been sourced by nucleation, are instead now appearing in the higher sizes, with more possibility to survive coagulation losses (primarily to accumulation mode), and 2) the coagulation sink will actually increase further removing any change of nucleation and moving the mass even more strongly into Aitken mode. As the SO₄ formed particles do not have to survive the 1-nm to 10-nm growth period with significant coagulation losses, the primary emission-derived scheme can actually in the end create more particles in the 30-50 nm range – as seems to be the case in the COSMO-ART. We have added the following sentences to the end of the first paragraph in section 4.4 (page 1842): “We consider the nucleation scheme of Kerminen and Wexler (1994) used to contribute to these differences. No explicit nucleation mode exists in COSMO-ART, hence secondary generated sulfate particles are transferred directly into the Aitken mode. A fixed factor is applied here to reasonably reduce number concentrations that get lost through coagulation during the growth from freshly nucleated clusters to Aitken-mode-size particles. In

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case the number of existing particles (e.g. in strongly polluted regions) does not match the assumptions made for this conversion factor, formation rates of Aitken-mode-sized particles through nucleation of SO₄2- are under-/overestimated.”

R1.29) Page 1843, line 17: Not much meteorological evaluation is presented in this study, so to say it was “very well simulated” is a bit of a stretch. It was well simulated in terms of the surface meteorological parameters examined. An evaluation of the upper-air conditions, boundary layer structure, cloud distribution, precipitation amounts, etc. is needed to make such a broad statement.

We agree that this would also be necessary. However, COSMO-ART is in its meteorological core identical to the COSMO NWP used operationally by several European Weather services. They are continuously evaluating the model. Our setup closely follows the setup of the German Weather Service (DWD) and we are therefore confident that our model exhibits similar performance properties as are found in operational verification by DWD. We have added two sentences at the beginning of the meteorological evaluation section mentioning this fact: “COSMO-ART is in its meteorological core code identical to the NWP model COSMO, and its performance is continuously verified by several European weather services. Meteorological evaluation has therefore been limited in this work to surface parameters [...]”. Further we have also restricted our conclusion to “Surface meteorological parameters”, so sentence 1 in the second paragraph of the conclusions now reads: “Surface meteorological conditions are very well simulated in all periods investigated without any need for tuning.”

R1.30) Page 1843, section 4: At the end of this section, a discussion on a lack of chemical data aloft is needed. Surface quantities, used exclusively in this study, depend on mixing from above. Why not pick a simulation period that includes field campaign data aloft?

Of course this would have been a further possibility of validating our model, although we would be limited to certain days only. Moreover, our paper is already quite long.

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Nevertheless, we have now mentioned this deficiency in the conclusions section (first paragraph, last sentence): “Not included in our work has been an evaluation of vertical profiles and upper-air variables of chemical and meteorological parameters with aircraft or radiosonde measurements.”

R1.31) Figure 1843, section 4: The number of stations is rather limited. Are these stations the only ones that contain data that is readily available to scientists? From a North American perspective, I would have expected more data to be collected routinely. However, it may not be readily available from some countries. Please elaborate.

There are more routinely measuring stations, however the dataset of Asmi et al. (2011) is one of the few homogenized ones. For comparison with such rather complex measurements careful consideration and correction for measurement conditions and setup are necessary which we could not do for this study and therefore settled on the Asmi et al. (2011) dataset which already did this work for us.

R1.32) Page 1844, line 21: The authors state that the model is suitable to simulate climate interactions. However, they have only shown predictions of AOD which is only one component contributing to of aerosol radiative forcing calculations. Other factors include single scattering albedo and angstrom component, but neither is evaluated in this study. Aerosol indirect effects are likely to be larger than direct effects, but the authors have not yet included those calculations in the model. Therefore, I do not think it is appropriate to state that the model is suitable for climate applications until the direct effect is evaluated in more detail with additional aerosol optical properties and when the aerosol indirect effects have been included and evaluated. Perhaps the authors mean to state that the basic model framework is now suitable to begin implementing and evaluating feedback processes?

The interactions of aerosols with radiation (Vogel et al., 2009, Stanelle et al., 2010) and most recently with clouds (Bangert et al., 2011) are already realized within the model system. However, within this study we switched of these interactions. The

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sentence has been changed and now reads: “We conclude that the model is suitable for air-quality assessments and the framework is set to evaluate the accuracy of aerosol-climate interactions.” Furthermore, the sentence “Particular focus is laid on the accuracy of the representation of climate relevant parameters of aerosols, i.e. their optical properties and ability to act as cloud condensation nuclei.” in the introduction section has been changed to “Through a detailed analysis of aerosol size distributions and chemical composition we set the basis for subsequent analyses of aerosol-climate interactions.” to inform the reader right from the beginning about the scope of this work.

R1.33) Table 1: Units of the quantities needs to be included for all variables. Only temperature units are included.

All units are normalized mean biases in %, the temperature units were erroneously put there to indicate that T biases were calculated from Kelvin (absolute temperature) values. We have removed the (K).

R1.34) Table 2: I assume that the N50, N150, and N250 are measurements made for a size range. So does N50 mean N50-100, etc? Please clarify.

This was given in the text (paragraph crossing pages 1834 and 1835), we have added it to the table caption as well, which now reads: “Comparison of number concentrations in different size ranges after Asmi et al. (2011) for number size distributions during the autumn 2008 simulation. N30_50: 30 to 50 nm, N50: above 50 nm, N100: above 100 nm, N250: above 250 nm. Note that the N250 parameter has a larger uncertainty than the others due to very low sampling rates.”

R1.35) Figure 2: The labels indicating the number of stations used are unreadable and covered over by other material. The authors should change the scale to make all of the text legible. The model and measurement label below the wind direction plot is meaningless, since only the biases are shown for that panel.

We have revised Figure 2 and 3 of the manuscript to make the number of stations

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readable and we have removed the model /meas. labels for the bias plot. Figures 1 and 2 show the revised versions.

R1.36) Figure 7: It might be useful to include a non-linear scale to better depict the differences between the simulated MODIS values when AOD is low.

We have settled for a scaling that better highlights the differences and enhance visibility of differences in low AOD regions. Figure 7 was revised, the updated version is shown in Fig. 3.

R1.37) Figure 8: I appreciate the authors showing many time series plots since they are more enlightening than just statistics. But for this figure it might be better to reduce the number of panels. I suggest focusing on a few (4-6) stations and then show the simulated and observed values for all of the cases, rather than for one case in the main part of the text. The rest of the material can be moved to the supplemental material. This just means exchanging portions of Figure 8 with the equivalent plots in the supplemental material.

We have changed the figure accordingly, choosing 6 stations of different regions in Europe. The rest has been moved to the supplement. The new figure in the manuscript now looks like Figure 4. Figure 18 from the supplement now looks like Figure 5.

R1.38) Table 6, supplemental material: I suggest moving this table to the main part of the paper.

The table has been moved accordingly.

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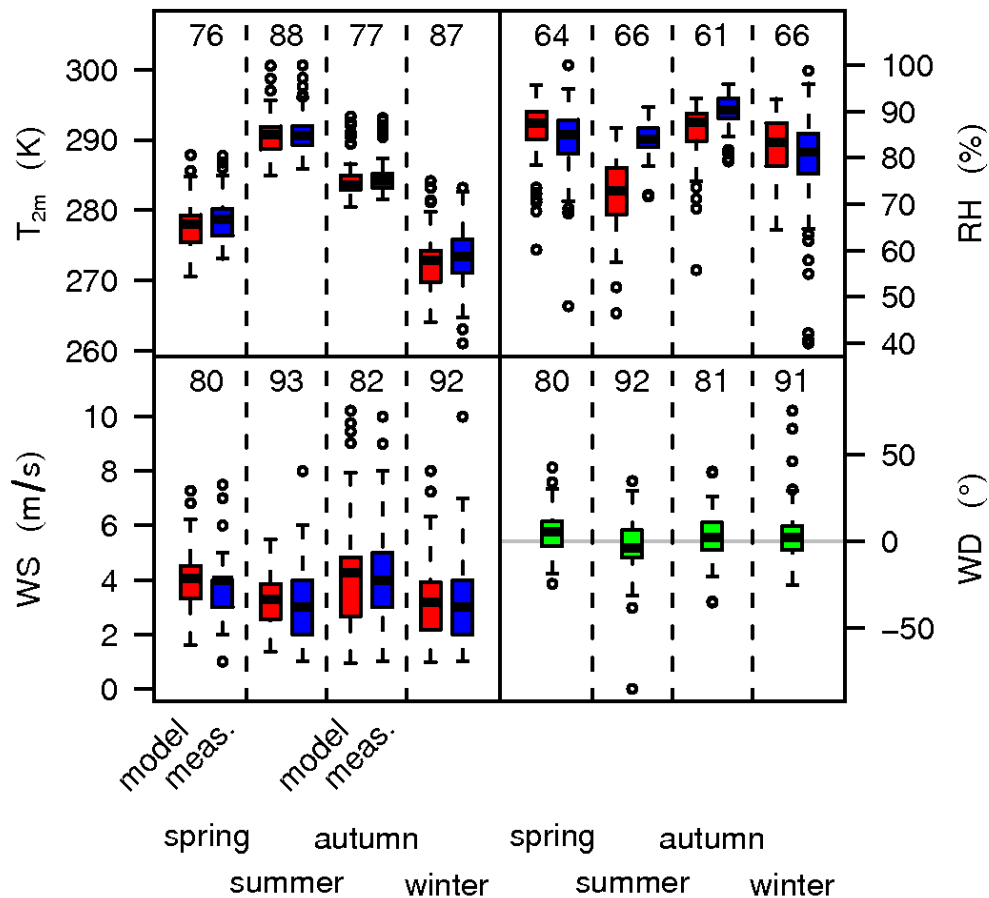


Fig. 1. revised version of Figure 2 in the manuscript

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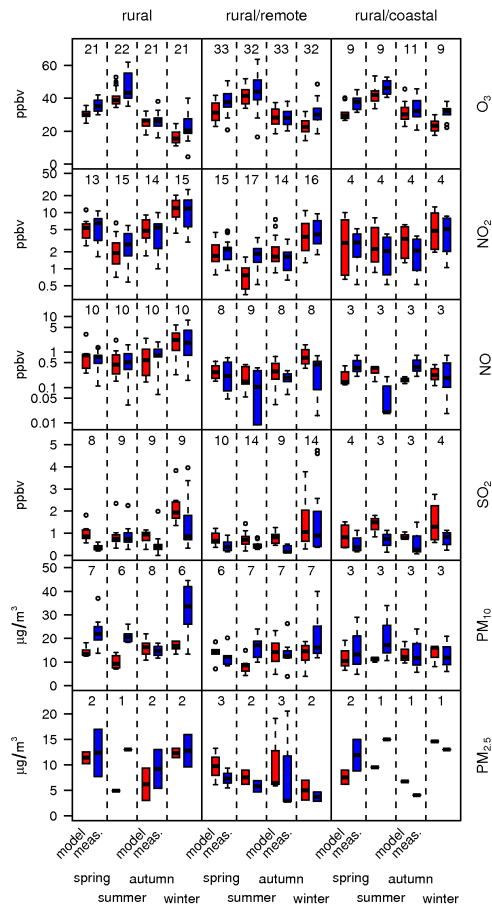


Fig. 2. revised version of Figure 3 in the manuscript

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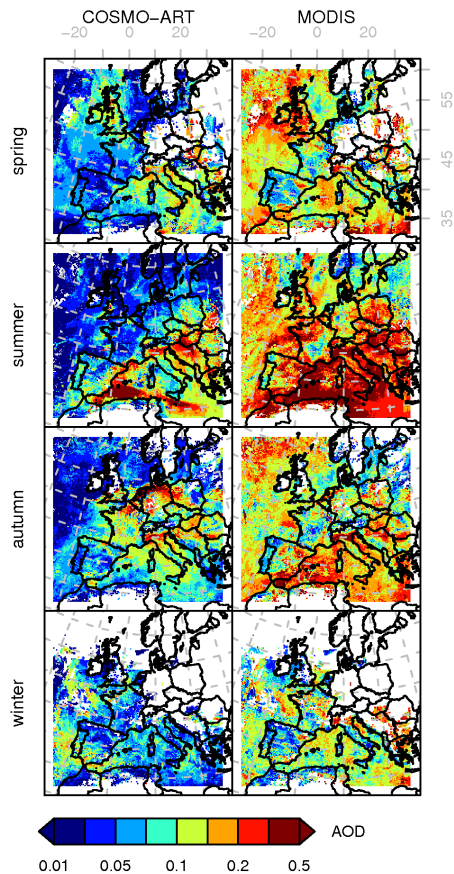


Fig. 3. revised version of Figure 7 in the manuscript

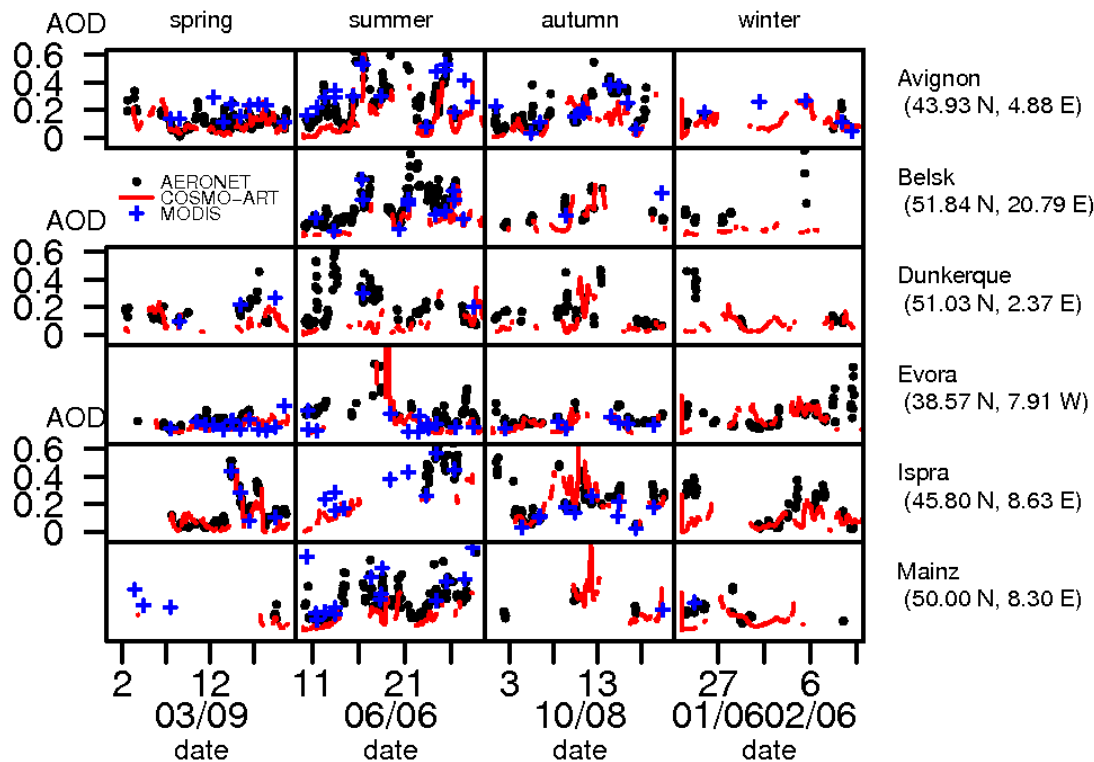


Fig. 4. revised version of Figure 8 in the manuscript

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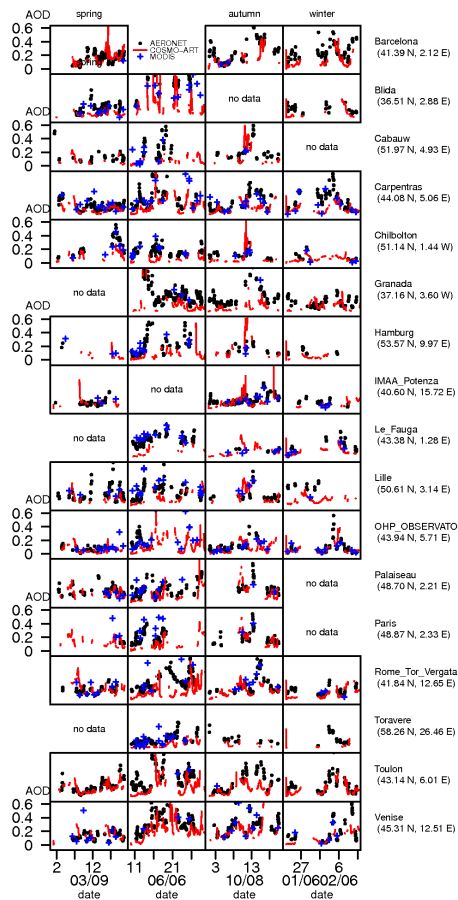


Fig. 5. revised version of Figure 18 in the supplement