

## ***Interactive comment on “Description and evaluation of GMXe: a new aerosol submodel for global simulations (v1)” by K. J. Pringle et al.***

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We thank referee two for the useful comments. We respond to the comments in the following text in which referee comments are in *italic* and our response is in regular text.

*Major comments:*

*Comparability aspects of the model and the observations have to be considered in more detail. In particular the following aspects have to be addressed:*

*1. If measurements during specific episodes are compared to the model output, the specific episodes should be extracted from the simulations. Comparing annual*

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*means from two specific years to data from shorter time periods (of possibly other years), as presented in Figure 3, could lead to a systematic bias since the respective episodes might show specific dynamical or chemical features which might not be consistent with the two-year mean. The authors argue that the comparison is only qualitative in such cases but that does not lead to a robust evaluation and any interpretation of the results might be meaningless. The authors should therefore think about presenting only those comparisons which can be interpreted from a quantitative point of view.*

We chose to include the comparison of simulated CN to the CN data collated by Andreae (2009) as it is the only available compilation that gives good geographical coverage and thus is able to corroborate the strong spatial gradients between polluted and clean environments, as calculated by the model. If possible we would like to keep this section in the final paper as we feel we have clearly explained the limitations of this comparison and it is our opinion that the comparison is still useful as it gives a global overview of the simulated aerosol number compared to observations around the globe.

To give a more quantitative comparison of simulated aerosol number to observations we have extended this section to compare to output from four Global Aerosol Watch stations that provide data for 2001 and 2002 (see the new Figure 5 (also shown at end of this comment)). These are the only four boundary layer stations that give coverage of this time period.

The following addition text has been added: “The Global Atmospheric Watch ([http://wdca.jrc.it/data/aerosol\\_program.html](http://wdca.jrc.it/data/aerosol_program.html)) program has collated a range of aerosol data from a number of observation stations. In Figure 5 we compare simulated aerosol number concentrations against the four boundary layer GAW sites which have data for both 2001 and 2002. GMXe simulates the aerosol number concentration in the

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Southern Great Plains (North America) site with good agreement, although the model tends to underestimate compared to observations (simulated annual average of  $4316 \text{ cm}^{-3}$  cf  $5614 \text{ cm}^{-3}$ ). Both model and observations show no distinct seasonal cycle. Aerosol number, however, is overestimated (simulated annual mean of  $7149 \text{ cm}^{-3}$  cf  $5045 \text{ cm}^{-3}$ ) in the nearby Bondville site. In the remote continental sites of Barrow (Alaska) and Pallas (Finland) the model systematically overestimates number (Barrow simulated annual mean of  $515 \text{ cm}^{-3}$  cf  $248 \text{ cm}^{-3}$ ; Pallas simulated annual mean of  $1818 \text{ cm}^{-3}$  cf  $825 \text{ cm}^{-3}$ ). Additionally GMXe fails to capture the observed seasonal cycle in Pallas with observations showing a decrease in aerosol number in winter months but the model showing much less seasonal variation.”

*2. Also the comparison to the mass spectrometer data (section 4.4) suffers from comparing with observational data from years which are not covered by the simulations. The authors could extend their simulations either to cover the times of observation or generate climatological information from the model showing a representative state of the aerosol. Means and variability gained from a longer term simulation (e.g. over 10 years) could be compared even to observations from time periods not covered by the model. In such a case, the observed values should be covered by the model variability. Only in cases where a low inter-annual variability of the considered quantities can be expected, the comparison as presented in the paper might be justified, but this has to be discussed in the paper and the authors should explain why only the two years were simulated. It should also be taken into account that changing emissions could cause discrepancies when different time periods are compared. This can be particularly important also in the case of the AEROCE data comparison where different decades are considered (section 4.5).*

We understand the reviewers concern that comparing model data simulated for two years to data collected over a wider range of years is not ideal, but we would like to clarify a few points:

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The simulations presented cover two full years of simulation time. Although this is a short time compared to the decadal timescale mentioned by the reviewer we would like stress that by considering two model years we include some inter-annual variability which is not captured when just one model year is evaluated (e.g. Stier et al., 2005; Spracklen et al., 2007; Trivitayanurak et al., 2008; Mann et al., 2010) or when evaluation is done just for selected months (e.g. Feng and Penner, 2007, who evaluate the model compared to one year of January and July data only).

Furthermore, the inter-annual variability in the model is only small (see e.g. figure 5), so that adding subsequent years in the comparison adds only limited information. The simulated inter-annual variability will increase when inter-annually varying emission estimates become available. This is in progress, as we are testing the latest version of GFED biomass burning emissions and the new EDGAR4 data.

Additionally, in the paper the main model evaluation for the sulfate / nitrate / ammonium aerosol is done in comparison to the EMEP / CASTNET and EANET measurements sites where both model and observations are for the same years (see Figures 9 to 12).

We chose to extend the evaluation to consider additional data, but due to the paucity of available measurements it was not possible to find measurements with a good geographical and seasonal distribution that are taken just in the years 2001 and 2002. To maximise the amount of data used to evaluate the model we extended the comparison to include the data collated by Zhang et al. (2007) which ranges from 2000 to 2006. We then present comparison of model with observations to this data. In Figure 13 we plot both the 2001 (triangle) and 2002 (star) data to give an estimate of the inter-annual variability.

To clarify the potential biases we have included the statement: “We note that as

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the observations span a wider range of years than the simulations the simulated meteorology will not represent the conditions at the time of measurement, thus there may be compensating errors that can not be identified without extending the simulation to span the full measurement time period. In future work GMXe will be used in a longer term simulation where evaluation of this kind will be possible.”

3. *Sea-salt and dust emissions are chosen according to offline emission data sets. Are these data consistent with the specific observation periods considered? The authors should discuss in section 4.2.1 whether this could be a source of systematic bias. If so, alternative simulations over a longer time period using the on-line emission approaches might help to evaluate the uncertainties inherent in the dust and sea salt simulations.*

The EMAC model has the option to simulate dust and sea salt emissions either online or offline. Both options have a range of advantages and disadvantages. Online emissions are wind speed dependent and thus are strongly sensitive to the resolution of the model. Emission of mineral dust aerosol can be model dependent as it also depends on e.g. soil moisture. The online emission of mineral dust needs thorough evaluation for every model setup.

The simulations presented here are run at a resolution of T42 but the work is intended to form a baseline evaluation case for a suite of higher resolution (T106) simulations that are planned in the future. The use of online emission of dust and sea salt would limit the applicability of the baseline evaluation to other resolutions.

Additionally, the offline emissions used are reliable and have been thoroughly tested in a range of models (Textor et al., 2007), as the focus of the paper is to evaluate the model in terms of the aerosol processing (not the emission fields) it is advantageous to use emission fields that are so well established.

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The drawback of the offline emission fields is, as the reviewer comments, is that the emission data is representative of the year 2000, thus it may induce biases when used from other years. We accept that this is a limitation of the study, but we do not expect that the bias is systematic or should strongly hinder the evaluation of the GMXe model.

To clarify we have added a number of caveats throughout the text:

Added to Section 2.5.1: “Offline in emission fields are used in this study given their extensive use and evaluation in a number of aerosol model frameworks (e.g. Textor et al., 2007). In taking this approach we acknowledge that the model will likely underestimate the inter-seasonal variability of dust and sea spray aerosol.”

Added to Section 4.2.1: “Dust emissions have a large inter-annual variability thus it is advantageous to compare to datasets that span a number of years (as done here), but in these simulations we use offline dust emissions representative of the year 2000 thus we will underestimate the inter-annual variability in dust concentrations.”

*Minor comments:*

1. *Model name: What is the meaning of the ?e? in the acronym GMXe? The model name would rather suggest the acronym GMeX.*

The "e" comes from the second e in the word eXtension.

2. *The use of the term ‘sea spray’ instead of the commonly used ?sea salt? has to be explained.*

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We choose to use the more general sea spray term as in GMXe the sea spray aerosol can comprise a mix of different components in addition to the NaCl, which is implied by the term 'sea salt'. The term is already widely used in literature, particularly in more recent publications.

3. *Introduction section: The authors highlight the comprehensive representation of nitrate in their model. It could also be mentioned in the introduction, that ammonium is considered explicitly, since this is another advantage over other models which only consider sulfate and refrain from explicit considerations of nitrate and ammonium.*

We agree that this is an other advantage of the model. We have added the text: "Including the partitioning of nitrate between the gas and aerosol phases in models has an additional benefit in that it also allows the more detailed treatment of ammonium aerosol. Models that do not consider the partitioning typically assume that all sulfate is in the form of ammonium sulfate, thus the concentration of ammonium in the aerosol is simply implied from the concentration of sulfate. Treatment of the partition of the sulfate / nitrate / ammonium system allows the on-line calculation of ammonium concentrations, rather than assuming a fixed sulfate to ammonium ratio."

4. *Section 2.1: The years which have been simulated should be specified here.*

Done

5. *Page 573, line 24: Write '(initiated by both...)' instead of '(both...)', since in-cloud scavenging leads to wet deposition only when precipitation occurs.*

Done

6. *Section 2.4.1: The authors should explain why they refrain from calculating*

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*dust and sea salt emissions on-line. Are the uncertainties of such on-line calculations larger than those of the applied emission climatologies?*

The choice to use the AeroCom offline climatologies was made to maximise comparability with the previous aerosol modelling work that has been done. We accept that using offline emissions we will underestimate inter-seasonal variability of dust and sea spray aerosol. As GMXe has the option of both online and offline emissions the user is free to choose the most appropriate for the experiment concerned. To clarify we have added the text:

"Offline in emission fields are used in this study given their extensive use and evaluation in a number of aerosol model frameworks (e.g. Textor et al., 2007). In taking this approach we acknowledge that the model will likely underestimate the inter-seasonal variability of dust and sea spray aerosol."

7. *Page 576, line 5: Provide reference for this kind of splitting.*

Added

8. *Page 576, line 16: refer to section 3.2.1 and Table 2.*

Done

9. *Section 3.2.1: The mathematical definition of the lognormal distribution should be provided here since it represents the basic concept of the model, at least a reference should be included.*

Equation has been included.

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10. Section 3.2.2: *The relative humidity might also be a driving parameter, in addition to temperature and sulfate concentration.*

Added the term “relative humidity” to the description.

11. Page 577, line 8: *The authors should discuss why sigma can be fixed. Is it justified since sigma shows a much lower variability than number and mass?*

Fixing sigma is a standard feature of the M7 microphysics scheme (Vignati et al., 2004; Stier et al., 2005) and other modal schemes e.g. (Mann et al., 2010). The standard deviation does tend to have lower variability than number and mass and by fixing this parameter calculation is simplified which makes the M7 model suitable for use in longer term simulations.

We have added the references to Vignati et al. (2004); Stier et al. (2005).

12. Page 579, line 8: *Replace ?aqueous species? by ?aqueous solutions??*

Done

13. Page 579, line 14: *Provide example for an aerosol metastable, inverse/forward problem?.*

We have re-written to:

“In ISORROPIA-II, the aerosol can be in either a thermodynamically stable state (where salts precipitate once the aqueous phase becomes saturated) or in a metastable state (where the aerosol is composed only of a supersaturated aqueous phase). The model can solve for either; (i) “forward” problems where the total (i.e.

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gas+aerosol) concentrations are known and the gas / aerosol concentrations are predicted, or (ii) “reverse” problems where the aerosol concentration is known and the gas concentrations are predicted. In this work we use ISORROPIA-II in the “forward” mode.”

14. Page 579, lines 17,18: *Specify which of the studies (references) deal with global, regional, and urban-scale modelling.*

Changed to: “Since its release, ISORROPIA-II has been used in a number of global (Pye et al., 2009) and urban-scale (Fountoukis et al., 2009; Karydis et al., 2010) model studies.”

15. Page 581, line 24: *A quantitative example for the coarse mode fraction of total nitrate should be given as a motivation.*

Added: “For example in a field study in two polluted coastal regions Yeatman et al. (2001) found that between 40 to 81 % of the total nitrate present in the aerosol phase was found in the coarse mode.”

16. *A reference for the n-monolayer thresholds should be provided. Are there any laboratory studies which would support such assumptions?*

In Section 3.4 we have re-written the discussion of the use of the monolayer threshold:

“The use of a monolayer approach was introduced by Vignati et al. (2004) and used by Stier et al. (2005) to express the ageing of hydrophobic particles into hydrophilic modes. Vignati et al. (2004) chose to define this ageing based on a monolayer approach: when sufficient hydrophilic material is added to the hydrophobic modes that “n” monolayers of hydrophilic material could be created then the material

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is transferred between modes. Vignati et al. (2004) varied “n” in a box model and found that n equal to 1 gave the best agreement to a detailed sectional model. In GMXe n equal to 5 is chosen, as in GMXe more material is available for condensation compared to Vignati et al. (2004, who treat condensation of H<sub>2</sub>SO<sub>4</sub> only). In GMXe the larger monolayer threshold gave better comparison of BC and dust aerosol to observations. The value of n is an adjustable parameter in the model (and in other models that take the monolayer approach, for example the GLOMAP-mode model which uses a monolayer threshold of 10 (Mann et al., 2010)). This larger threshold is in line with the finding of Granat et al. (2010) who examined aerosol concentrations in precipitation in the Maldives and found that soot aerosol could remain hydrophobic for many days after emission. Modal aerosol models (including GMXe) would benefit from additional laboratory and field studies into particle ageing which could better constrain how much hydrophilic material is required to make a hydrophobic particle hydrophilic.”

17. Page 585, line 4: mention that also EQSAM was applied as an alternative.

We would rather not add this here as it is explained later and we think that referring to EQSAM in this section may make it a little confusing.

18. Page 585, line 15: ?zonal mean annual average?.

Changed

19. Section 4.1: it should be discussed in more detail why the evaluated number concentrations by Stier et al. can be used as a reference point here. What are the differences to Stier et al.? Can they be neglected? The authors only quantify the largest differences (coarse mode). For evaluation purposes it might be useful to quantify the maximum deviations in the other modes.

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Changed text to: “As a first evaluation step we present simulated fields of number concentration in the same format as those presented by Stier et al. (2005) using the ECHAM-HAM model. Comparison of GMXe fields with ECHAM-HAM fields is useful as ECHAM-HAM is a well established and widely used aerosol model (e.g. Stier et al., 2006; Lohmann et al., 2007) and inter-model comparison gives a global overview of the number concentration (per mode) that is not easily achieved from field observations. The zonal mean annual average aerosol number concentrations simulated by GMXe are shown in Fig. ???. For evaluation, our Figures 2 and 3 are comparable to the number concentrations shown by Stier et al. (2005) in their Fig. 4.”

Added: “The other large difference is in the accumulation hydrophilic mode where the strong minima ( $<0.05 \text{ cm}^{-3}$ ) simulated by ECHAM-HAM at the poles at 500 hPa is not simulated in GMXe (number concentrations of 20-50  $\text{cm}^{-3}$  are simulated instead), however observations in this region are limited so it is difficult to determine the bias of either model.”

20. Section 4.2: Explain why using different metrics (GMR, ARM, . . .). Which specific conclusions can be drawn from the specific metrics?

Added: “The use of a range of different metrics is advantageous as each metric is susceptible to different biases, for example PF2 is not biased by data points that are very far from the median and is a useful metric for giving an overview of performance, but it gives no indication if the deviation between model and observations is systematic (e.g. constant underestimation) or random, which can be seen from GMR and AMR. By combining the three metrics we gain an overview of model performance.”

21. Page 587, line 1: ?percentage of model point where the respective species concentrations deviate from the observations by less than a factor of two?.

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Changed

22. Section 4.2.1, discussion of Fig. 6, Tab. 3: The modelled BC concentrations over North America show nearly perfect agreement with the IMPROVE observations. This is remarkable regarding the comparatively high uncertainty inherent in current BC emission data. It is also remarkable since many IMPROVE stations are located in rural areas. Hence deviations from the modelled mean concentrations (including also concentrations in highly polluted areas) must be expected. The authors therefore should discuss that the perfect agreement might not indicate highest achievable model quality.

Added: "As the IMPROVE sites are all rural locations this comparison does not cover the full range of observed concentrations in different regions of North America, thus the very good agreement between model and observations will be investigated further in the future, with comparison of a higher resolution model to data taken from a wider range of environments."

23. The presentation of comparisons with large-scale observation network data in section 4.2.1 is inconsistent with the title of section 4.3. Hence the title should be changed.

Changed to: "Comparison of simulated sulfate, nitrate and ammonium concentrations to large-scale observation networks"

24. Section 4.2.2, last paragraph: discrepancies can also arise from different representation of precursor chemistry and different NO<sub>x</sub> emissions.

We agree this is a good point. The additional text has been added.

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25. Page 589, line 19: Provide examples for such problems and/or a reference.

I have deleted the statement: "although we acknowledge the well documented problems associated with comparing point measurements to large scale model data."

26. Page 589, last paragraph: Add sentence "The details of the evaluation are discussed in the following."

Added

27. Section 4.3: Information on the time periods covered by the observations from the specific networks and the time periods considered for model evaluation should be included.

Added: "In the following section both the observations and the model results are from the years 2001 and 2002."

28. Page 590, line 2: insert ?concentration? behind ?sulfate?.

Done

29. Page 591, line 7: explain why dust is important here.

Re-phrased to: "As ammonium sulfate is formed preferentially over ammonium nitrate, the latter only forms in inland regions if there is excess of cations e.g. ammonia or the cations present in mineral dust available after all the sulfate has been neutralised."

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30. *Section 4.3.3: It should be discussed whether uncertainties in nitrate could also result from uncertain gas phase precursor chemistry or NO<sub>x</sub> emissions.*

Added: “An additional possible cause of bias in the simulated nitrate concentrations comes from the concentrations of the gas phase precursors. Estimates of global NO<sub>x</sub> emissions are uncertain (e.g. Konovalov et al., 2008; Han et al., 2009) and these uncertainties (along with uncertainties in the deposition rate of gas phase precursors) add to the potential for biases in the gas phase precursor fields. The simulated fields of HNO<sub>3</sub> and NO in ECHAM/MESSy were evaluated in Jöckel et al. (2006) and Tost et al. (2007) examined the wet deposition of nitrate and found the model simulated deposition fluxes to be in good agreement with observations.”

31. *Page 592, lines 19, 20: Even a perfect agreement of model and observations might not attest high model quality since differences might have to be expected, regarding the differences in meteorology.*

Added: “We note that as the observations span a wider range of years than the simulations the simulated meteorology will not represent the conditions at the time of measurement, thus there may be compensating errors that can not be identified without extending the simulation to span the full measurement time period. In future work GMXe will be used in a longer term simulation where evaluation of this kind will be possible.”

32. *Page 596, lines 16, 17: Is this in agreement with the changes in sulfur emissions which have been large during the last decades?*

Added: “As the geographical distribution of sulfate precursor emissions has changed significantly since the 1980s, a perfect agreement between model and observations is not expected, however, this change in the distribution of emissions

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is less important for remote marine locations where the AEROCE data is concentrated.”

33. *Section 4.7: Since the results from EQSAM and ISORROPIA mostly agree, it would be worthwhile to discuss also technical aspects like the computational expenses or technical advantages/disadvantages of the modules.*

We would rather not detail the differences in the technical aspects of EQSAM and ISORROPIA-II in this section as both models are detailed in the literature and some description of each is given in Section 3.3.1 and 3.3.2. What is important for this paper is simply that both models are implemented in GMXe and give similar results.

However, we have expanded the discussion a little to include (Section 3.3.2):

“EQSAM3 is developed and maintained as part of the ECHAM/MESSy group, and will be further developed to include additional aerosol compounds e.g. sugars.”

Added in Section 4.7: “Finally, it is worth noting that as implemented within GMXe, both partitioning models have similar CPU times.”

*Editorial changes:*

All the editorial changes suggested have been implemented.

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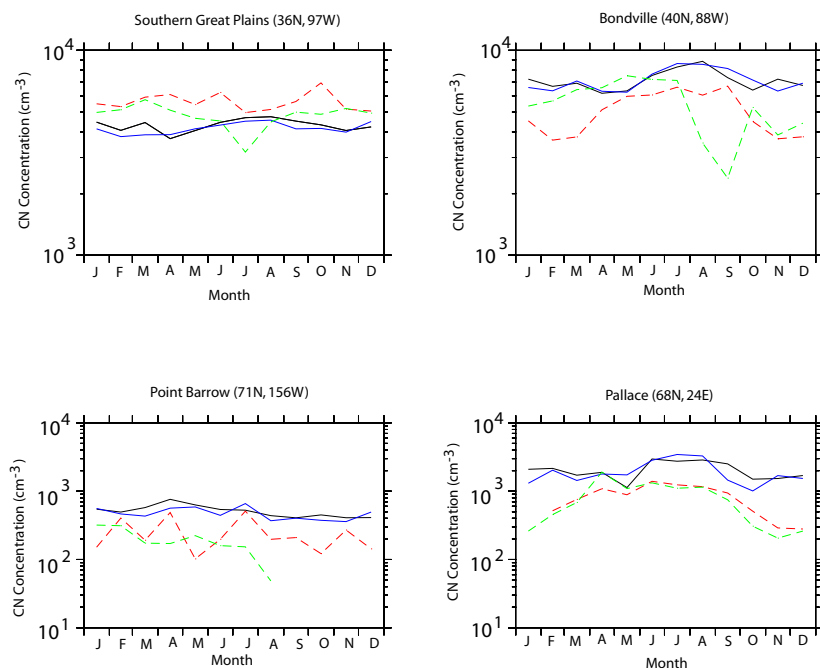
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**Fig. 1.** Seasonal cycle of aerosol number concentration ( $\text{cm}^{-3}$ ) in four locations as measured by the Global Atmospheric Watch network. Lines show modelled values for 2001 (black) and 2002 (blue) and obser

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