

## ***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 one for the useful comments, we have responded to all the comments and made appropriate changes to the text. Referee comments are in *italic*, our response in regular text.

*This manuscript describes a new or, more precisely, an updated aerosol sub-module for global modeling purposes. The model has been adequately described and evaluated in a sufficient detail against atmospheric observations. The paper is scientifically sound and well written. There are a few issues that should be addressed before acceptance for publication in GMD.*

*Section 2.2: From the description it remains unclear whether EMAC produces the*

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*concentration fields of gas phase species, or their production rate fields. I suppose it should be production rate because many of the gaseous species are lost irreversibly to the aerosol phase, or are partitioned between the two phases.*

The MECCA submodel in EMAC Sander et al. (2005) calculates the production rate of gas phase species, from which a concentration is inferred. The loss of gas phase species to the aerosol through heterogeneous reactions (e.g.  $\text{N}_2\text{O}_5$  to form  $\text{HNO}_3$ ) is treated using the HETCHEM submodel (Jöckel et al., 2006). In the simulation presented here a climatological aerosol distribution is used by HETCHEM, but it is also possible to use the online distribution as simulated by GMXe. Other gas / aerosol partitioning is treated through GMXe.

We have altered the text in Section 2.2 to:

“The EMAC model calculates fields of gas phase species online through the MECCA submodel (via the Module Efficiently Calculating the Chemistry of the Atmosphere Sander et al., 2005). MECCA calculates the concentration of a range of gas phase species, including aerosol precursor species such as  $\text{SO}_2$ ,  $\text{NH}_3$ ,  $\text{HNO}_3$ , DMS,  $\text{H}_2\text{SO}_4$  and DMSO. The concentrations of the major oxidant species ( $\text{OH}$ ,  $\text{H}_2\text{O}_2$ ,  $\text{NO}_2$ , and  $\text{O}_3$ ) are also calculated online (see Sander et al., 2005; Jöckel et al., 2006).

In GMXe the loss of gas phase species to the aerosol through heterogeneous reactions (e.g.  $\text{N}_2\text{O}_5$  to form  $\text{HNO}_3$ ) is treated using the HETCHEM submodel (e.g. Jöckel et al., 2006).”

*Section 3.2.2: Is the model capable of dealing with nucleation schemes other than the binary one?*

At present this is not possible, but the model design is flexible and additional

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schemes will be implemented in the future.

We have clarified by adding the word “binary”:

“Two binary nucleation schemes are available in GMXe”

*Section 3.3: The author should mention how they treat the cloud droplet activation of aerosol particles, and how this is coupled to aqueous-phase chemistry described in section 2.3.*

Added the following new Section 2.4:

“Wet removal of aerosol particles occurs via both nucleation and impaction scavenging. Whereas the impaction scavenging is caused by the physical process of falling droplets and crystals which affects both hydrophobic and hydrophilic particles, the nucleation scavenging (representing the activation of aerosol particles) is only calculated for the hydrophilic modes. For determining the scavenged fraction of the particles per mode an empirical formula (see Tost et al., 2006) is applied.

The material incorporated in cloud droplets via nucleation scavenging can be removed from the atmosphere based on the precipitation formation rate or released back into the aerosol phase after cloud evaporation. Furthermore, it can participate in chemical reactions in the aqueous phase (see above). Due to the assumed internal mixing of the particles within the hydrophilic modes also the coated hydrophobic cores, e.g. OC, BC or dust can undergo nucleation scavenging if these compounds exist in the hydrophilic modes. However, the information of the nucleation scavenged particles is not used for determining the cloud droplet number concentration, which is in the current simulations a climatological value only. ”

*Section 3.3.3: The authors assume that organic aerosol species do not uptake water, which is contrary to observations. This has probably little effect on the aerosol liquid*

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*water content, but it might influence the aerosol cloud droplet nucleation activity (see also the comment above).*

We agree that this is a limitation of the present model, work is currently in progress to calculate the water uptake of the organic aerosol using the  $\kappa$  parameter (Petters and Kreidenweis, 2007), but this is still undergoing testing. Once complete, we will examine the importance of treating water uptake on organics on the aerosol distribution and lifetime.

Additionally at present the model wet scavenges the aerosol in the hydrophilic modes without an explicit dependence on aerosol composition (instead an empirical parameterisation (Tost et al., 2006) is used). See answer above.

*Section 3.3.4: This is a well-documented and proper approach. However, where are the assumed values of accommodation coefficients (page 582, line 9) taken from?*

Added references.

*Section 3.4: Physically, the cloud droplet nucleating activity of an aerosol particle has little to do with the number of monolayers of soluble material on it. The authors should be justify their approach and explain why they have chosen 5 monolayers instead of 1 used in M7.*

The use of a monolayer approach was justified in (Vignati et al., 2004; Stier et al., 2005). However, we agree with the reviewer that the monolayer approach is a simple parameterisation and not a representation of the true ageing process, which remains unresolved in most global aerosol models; models typically use either (i) the monolayer approach, (ii) a e-folding lifetime that is not dependent on the concentration of the condensible species or (iii) no treatment of ageing.

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The one monolayer threshold used by Vignati et al. (2004); Stier et al. (2005) was chosen as it gave the best agreement of a 1D version of a modal aerosol model to a sectional aerosol model, for the setups considered. As it is a very difficult concept to evaluate with regards to observations there is considerable uncertainty in the the choice of the value. For example, the GLOMAP-mode aerosol model uses a 10 monolayer threshold (Mann et al., 2010).

We found in GMXe that using a threshold value of just one monolayer lead to unrealistically short lifetimes of BC and dust aerosol, this understandable as GMXe treats more condensable species than ECHAM-HAM thus the ageing is faster, for this reason we increased the threshold to 5 monolayers.

We have altered the paragraph in the text to clarify this point. New text:

“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 is transfered 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

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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.”

*Section 4.2.1: The authors explain the modelled bias in sea spray concentrations as an artifact due to strong concentration gradients in coastal areas. How about elsewhere? It is well known that current sea spray emission modules have an uncertainty of at least a factor of 2-3.*

We agree that there is considerable uncertainty in sea spray emissions which could explain some of the bias in the model, but in Figure 6 we show that the model gives good comparison to sea spray observations when sea spray concentrations are high (close to production regions) but when sea spray concentrations are lower (typically sites further from the ocean) the model overestimates sea spray concentrations. This is a feature also noted by Stier et al. (2005) and Kerkweg et al. (2008) in different implementations of the M7 modal aerosol model. In these papers the authors suggest that it is due to a failure of a model with relatively coarse resolution to capture strong horizontal gradients, which most often occur at coasts. From the simulations done in GMXe, we agree with their analysis of the problem.

Work is currently underway to run GMXe at a higher horizontal resolution (T106), in this new version we will examine the rate of decay of sea spray with distance from the coast, which will allow us to better understand (and hopefully solve) the bias, but this work is still underdevelopment and will be presented in a future publication.

*Section 4.6 and Figure 13: Although most of the modelled values of AOD are within a factor 2 from those measured by AERONET, the scatter appears to be relatively high. Maybe the authors could comment this feature a bit.*

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Added the text:

The scatter plot shows that most (92%) simulated AOD values are within a factor of two of the AERONET observations, however despite the generally good agreement with observations there is still a lot of scatter in the comparison, particularly in the monthly mean data, this implies that the seasonal cycle of the AOD may not always be well captured. This will be investigated further with the AEROPT submodel, when the submodel development is complete.

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