

Dear Editor

Please find below the answers to both referees which list all relevant changes.

Sincerely

Klaus Klingmüller

Answer to referee #1

We are grateful for the referee's valuable comments which we address individually in the following:

1. We have changed the title to

„Sensitivity of aerosol radiative effects to different mixing assumptions in the AEROPT 1.0 submodel of the EMAC atmospheric chemistry-climate model“

(Please note that we now also provide explicitly the version number of the AEROPT submodel which was previously only implied by the MESSy version. This version number is most relevant since AEROPT is not linked to a particular EMAC version and in the present work it is mostly used independently of EMAC.)

2. We are referring to the absorption cross section per geometric cross section which decreases as the absorbing components get diluted by less absorbing components. For clarification, we changed (p. 3389, l. 28)

„... the decreased absorption efficiency.“

to

„... the decreased absorption per cross section.“

In addition, in line 12, page 3371, we have added

„For given extinction coefficient σ , the fraction of absorbed radiation $1 - \omega$ is proportional to the absorption efficiency.“

3. We have changed the notation to „asymmetry parameter g “.
4. We have included a reference to Chylek et al. 1988 (p. 3372, l. 2).
5. We have added the following as section 2.2.3 „Non-spherical particles“:

„Even though multilayered particle models can represent atmospheric aerosol particles more realistically than homogeneous spheres, common implementations still assume spherical symmetry. While this approximation is well defined as changing the order of averaging over many particles and computing the optical properties, it is generally not well motivated physically but rather used for computational simplicity.

In reality, dust particles can have irregular shapes which may be better represented by

spheroids, ellipsoids or even more complex structures possibly yielding, for the same particle volume, a higher optical thickness and single scattering albedo (Kalashnikova and Sokolik 2004; Colarco et al. 2014). Soot is known to form a variety of shapes by aggregating into fractal-like chains (Adachi et al. 2010; Scarnato et al. 2013). Even if liquid components lead to a spherical particle surface, embedded solid components are not necessarily distributed concentrically within the particle, and the exact position affects the optical properties (Fuller et al. 1999; Adachi et al. 2010).

The implementation of more detailed particle models becomes increasingly important as the overall precision of global climate models is improving. Introducing non-spherical particles into EMAC remains subject of future work.“

6. We have replaced „lensing“ by „amplification“ (p. 3374, l. 19, 20).

7. On page 3376, line 25, we have added the sentence

„The smallest radii plotted are just below the validity limit of the analytic approximation which does not reproduce the drop in the single scattering albedo for very small radii.“

8. In the supplement, we have included the data plotted in Fig. S1 as tables (Tab. S1-S2) and specified the wavelengths for which the Sokolik and Kirchstetter data is used by replacing/extending

„The mineral dust and organic carbon values have been complemented by data from I. N. Sokolik (unpublished data, 2005) and Kirchstetter et al. (2004), respectively.“

by

„The mineral dust values have been complemented by data for $\lambda > 2.5$ micro-m from I. N. Sokolik (unpublished data, 2005), the organic carbon values by data for $\lambda < 0.7$ micro-m from Kirchstetter et al. (2004). The numerical values of the refractive indices can be found in Tab. S1 (real part n) and Tab. S2 (imaginary part k).“

In the main document we have extended the last sentence on page 3379 to

„The indices are plotted in Fig. S1 in the Supplement, the numerical values can be found in Tab. S1 (real part n) and Tab. S2 (imaginary part k).“

Answer to referee #2

We thank the referee for pointing out the important value of our study as indeed we quantify the difference between two common treatments in climate models, which is a major result of our study. In fact, most climate models consider external or homogeneous internal mixing (and possibly a combination of both). A listing of models and their aerosol mixing (internal, external or internal + external) can be found in, e.g., Easter et al. 2004, which we have now included as reference. The exact mixing scheme we compare to external mixing – namely homogeneous internal mixing within externally mixed modes – is used similarly by various models like ECHAM-HAM (Zhang et al.

2012), MIRAGE (Easter et al. 2004) and EMAC (Joeckel et al. 2005, 2006). Hence, our comparison is most valuable for the development of these and comparable models and in particular advancing the EMAC model is our main concern. We have added after line 25 on page 3369:

„..., possibly allowing a combination of both. Easter et al. (2004) list 16 models, five using external mixing, two internal mixing and the remaining models a combination of both. Also EMAC uses a combination by assuming internal mixing within externally mixed modes, similarly to, e.g., ECHAM-HAM (Zhang et al. 2012) and MIRAGE (Easter et al. 2004).“

For further clarification, we have changed line 19, page 3370, from

„A key aspect of this work is the comparison of internal and external mixing ...“

to

„A key aspect of this work is the comparison of internal and external mixing within the aerosol modes...“

In the context of the present study the comparison has the additional purpose of allowing the assessment of the results for our newly implemented core-shell treatment. As argued in the article, this treatment can be a more realistic representation of coated particles (even though in our case studies it yields results quite close to the results for homogeneous internal mixing). Thus, the core-shell column results are another major finding of our study introducing a new level of detail to the modelling of aerosol optical properties in EMAC.