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Geoscientific
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Discussions

Interactive comment on "Development of an online radiative module for the computation of aerosol optical properties in 3-D atmospheric models: validation during the EUCAARI campaign" *by* B. Aouizerats et al.

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To Reviewer#1,

The authors appreciate the constructive and helpful comments provided by Reviewer#1. Its comments helped to improve our manuscript. The paper has thus been modified to take into account the recommendations given. Below, we have copied

the referee comments in italics and inserted our responses in standard font where appropriate.

Regards,

Benjamin Aouizerats

1 Specific comments

1. *My main comments and suggestions are directed to the clarity of the presentation.*

   An effort has been made in order to give more clarity for the readers. A detailed description of improvements will be given under the specific comments.

2. *Also, I am very surprised by the reported in this study single scattering albedo reaching the values of 0.6 and even 0.5 at the wavelength of 550 nm. These values are obtained by measurements and reproduced by the radiative module for the Cabauw site in the Netherlands. I hardly believe that such low values can be observed for the ambient aerosols, even in an industrial region. The authors certainly have to check it carefully before possible publication.*

   We understand that SSA values below 0.6 are unusual and may be surprising. However, we would like to insist that most of the observed and modelled values of SSA reported in this work are around 0.8 (with a mean value of 0.815 for the modelled values and 0.818 for the observed values during the period studied) and only a few limited observations reach values lower than 0.6 (Figure 10). Moreover, it should be noted that several studies (Marley et al., 2009, Gomes et al., 2008, Babu et al. 2002, Singh et al. 2005, Ganguly et al., 2006) have reported chronic low SSA values for continental/urban polluted atmosphere. In addition, it has to be noted that the SSA observed and modelled are both under dry conditions, leading to lower SSA values compared to wet conditions. In that sense, we have now modified the text (we added some references and insisted on the dry conditions) to clarify this specific point.

3. *Coming back to the clarity of the presentation, I found that some statements are limited in justifications and some parts of the manuscript can be better formulated. In particular, the validation part of the paper (section 4) has to be improved. The authors go too fast into the detailed explanation of the measurements and comparison of the number. It has to be clearly stated in which conditions the validation is conducted and what are the restrictions. In addition, I would suggest providing a schematic diagram in the first part of the manuscript illustrating logistics of the developed radiative module. Second schematic diagram can illustrate logistics of the validation part. I leave to the authors to decide how to improve the presentation clarity, but I believe that this improvement will be appreciated by the readers. Nevertheless, I think that this is a useful study which worth of publication and certainly appropriate to Geoscientific Model Development.*

   We understand that some parts of the paper were not very clear. We made a special efforts to improve the presentation clarity. As suggested by Reviewer#1, we added a shematic diagram for the validation methodoly. We agree that this

figure and the comments now associated within the text were necessary for a correct understanding of the steps leading to the validation. In that way, section 4 has been particulary improved. We also clarified the text concerning section 3, and in that way we don't think that a schematic diagram would be more helpful.

4. *Regarding the limitations of the presented validation: As far as I understand this module is not validated for dust particles. The measured chemical composition does not include mineral dust and the effect of non-sphericity is not accounted for. By the way, how it was found that 29 May represents dust?*

   This is effectively right and this optical scheme has been not evaluated for dust particles. In that sense, the effects of non-sphericity on aerosol optical properties is not accounted for. This assumption is supported by the fact that fresh BC aerosols (not spherical) are generally coated with secondary hydrophilic aerosols (Giawaly et al., 2009) in urban atmosphere, allowing to use the Mie theory. Furthermore, this optical scheme is treating aerosols in the internal mixing way (not external) using the core/shell representation (to calculate associated refractive indexes) that allows to take into account the coating of primary BC by secondary aerosols. Finally, it should be noted that an associated aerosol scheme dedicated to dust particles is already existing (Grini et al. 2006). This specific development has been evaluated in the frame of the AMMA experiment (Tulet et al., 2008, Mallet et al., 2009). This last point was not so clear in the article and we have now modified it. For example, we have removed the sentence Dust were observed.... in order to clarify the text. Indeed, we did not study this dust event in our work due to the absence of experimental observations and the validation part ends on 28 May.

5. *Is a constant value of density (2.5 gcm-3) used for all type of particles? Could*

references or a discussion be provided for this value? What are the expected uncertainties?

A constant value of density is used for the whole aerosol population (to get the mass concentration from the observed number concentration) We understand that this value may not be the most appropriate. According to bibliography (Lee and Adams, 2010), aerosol density values are between 1.4 for Organic Matter and 2.65 for dust particles. We assume that the choice of a constant density, and particulary a value of 2.5 may be the main source of error in the validation part. However, the comparisons of mass extinction efficiencies and single scattering albedo are not affected by this error. Indeed, the density is beeing used only to compute the total mass concentration. Thus, the mass extinction efficiency is not dependant of the total mass concentration, neither is the single scattering albedo.

6. *Are only dry conditions considered in the validation procedure?*

   Indeed, the measurements were done only under dry conditions. We have now modified it in the text to gain more clarity.

7. *The absorption Angstrom exponent is used for the assessment of the absorption coefficient at 550 nm. The absorption Angstrom exponent is derived from the aethalometer measurements and I think that presentation of these values could be interesting. Also how they agree with the literature?*

   We have now reported the values of the absorption Angstrom exponent (AAE) obtained during the period studied and compared them with values reported by Russell et al. (2010) for different aerosol types. In our case, the comparisons

between referenced and simulated AAE show consistent results.

8. *The section 4.1.2 is not very clear to me. Could you please provide a justification for 60- 40% split of POM? How do you explain the factor used for multiplication of composition concentration? Are these non measurable remaining chemical elements? If yes, what are the assumptions regarding optical properties of these elements?*

   We have worked on the clarity of this part.
   We used the 60-40 % split for POM according to Dentener et al., 2006. The inventory for Western Europe show values of emissions of 0.33 Tg/year for SOA and 0.81 Tg/year for total POM. However we performed sensitivity tests by changing the split values for POM, and the optical properties resulting are very close. This is certainly due to the fact that the refractive index for primary and secondary are the same in this study.
   The multiplication applied to the composition concentration results from the fact that the AMS has a cut-off diameter of 0.5 μm. Then, we multiply the mass of each aerosol compound by the factor found between the total mass concentration given by the SMPS+APS and the mass observed by the AMS+MAAP.

9. *I think that the word assumption appropriates better than hypothesis in sections 4.1.1 and 4.1.2.*

   We agree and replaced the word hypothesis by assumption .

10. *The next sentence is not clear to me The median diameter evolves as the geometric standard deviation. Also, there is a typo in second after this sentence:*

size description replace by size distribution .

We meant Median diameter and geometric standard deviation both evolve.. The sentence has been corrected. The typo has been corrected.

11. *Section 4.2: The correlation coefficients for the modeled and the measured mass extinction efficiency and single scattering albedo are reported. The authors can present also the biases.*

    The bias values have been added.

12. *Explanation of EUCAARI abbreviation is missing. Please provide it already in the abstract and in the introduction. Introduction, row 14: What do the authors mean by and so ? Please avoid. Introduction, row 29: There is a typo elvolving .*

    The abbreviation has been added in abstract and introduction, and the typo has been corrected.