Atmos. Chem. Phys. Discuss., 9, C11037–C11043, 2010
www.atmos-chem-phys-discuss.net/9/C11037/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sources of uncertainties in modelling Black Carbon at the global scale" *by* E. Vignati et al.

E. Vignati et al.

elisabetta.vignati@jrc.ec.europa.eu

Received and published: 22 February 2010

The paper evaluates BC concentrations at various surface measurement stations around the world, with two different aerosol schemes within the TM4 model, a simple bulk scheme and a more complex dynamical scheme. I find this study interesting and valuable, but to simplified in its current form. Therefore I suggest major revisions.

We thank the reviewer for the comments and suggestions which improved the manuscript. We address the specific points below.

1) Observational datasets: Constraining a coarse model by surface measurements only is highly questionable, more data sets, e.g. vertical BC profiles from campaign measurements, such as ARCTAS, AVE, CARB, ARCPAC etc. should be included in

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

this study. Surface deposition rates of soluble species can as well be evaluated.

The suggested vertical profiles were used for model evaluation and results are reported in Figure 7. The following text has been added to paragraph 3.2: “To compare with the observed aircraft measurements vertical profiles of modelled BC are constructed averaging monthly mean fields (standard model output) at the locations correspondent to the flight tracks (see Koch et al. 2009 for the details of flight and tracks). It should be noted that the observed profiles are collected during a few days of flight and in different years that the modelled fields. Figure 9 shows the observed and modelled profiles over mid-latitude regions (a,d), in the tropics (b,c) and at high latitudes (e-i). The two model approaches behave in similar ways agreeing with the observations in one case (a) and underestimating the concentrations in (d) at mid-latitudes and overestimating the concentrations in the tropics at high levels in the tropics. The BULK model already estimates lower concentrations compared to the DYNA case, although the differences are small compared to the measurements. At high latitudes both the models underestimate the concentrations and the differences between the models is in some cases larger. Looking at the comparison of model results in the BC evaluation done in Koch et al (2009) the results of our work fall in the same range, having the tendency to overestimate in the mi-latitude and tropic cases, except for case (d) where all models underestimate the concentrations at lower levels. The same models generally underestimate BC in the high latitude cases. “

2) What is the data source of the longterm campaign measurements? Please provide a citation or website.

The following references were added to the text in paragraph 2.3: “(Sharma et al. 2004, 2006; Wolff and Cachier, 1998; Bodhaine, 1995; Echalar et al, 1998)”

3) Better description of aerosol processes. This paper focuses on the differences in wet removal of bulk versus microphysical aerosol scheme, therefore all removal processes should be explained in more detail. Furthermore a better description is needed

ACPD

9, C11037–C11043, 2010

to understand the internally mixing assumption as used in the BULK model. How is solubility calculated? The model results show that BULK aerosols have a shorter lifetime than the DYNA aerosols. This is surprising as better treatment of aerosol mixing state would make BC more soluble. Therefore it is critical to understand the aerosol characterization within both models. The found differences are not only associated to the wet removal schemes. A more detailed diagnostic is necessary to understand those differences.

In the BULK approach BC is externally mixed, considered only by mass and assumed hydrophobic. In case of rain only 70% in-cloud BC is considered highly soluble and in-cloud scavenged. Therefore in the BULK case independently on the ambient concentrations of other particles or gases, BC behaves in the same way and the scavenging is dependent only on the amount of rain and not on the BC actual status. Only in TM5/M7 the solubility of the particle containing BC is dynamically calculated, it is dependent on the presence of soluble particles with which freshly emitted BC can coagulate or be coated by soluble H2SO4 and become hygroscopic. Figure 8 shows the annual and zonal mean of the interstitial fraction in TM5/M7 and it is clear that the interstitial fraction for this case is in general larger than 30% that is supposed in the BULK case. It means that in the bulk case the model removes almost everywhere more BC than in the TM5/M7 case, and this is the reason why BC lifetime in the BULK case is shorter. Also the second case where the 60% interstitial aerosol is applied removes much more BC in the areas far away from the sources (Figure 8) than TM5/M7. The text of paragraph 2.1.3 has been partially rephrased.

4) The paper focuses on wet removal, therefore an evaluation of the used cloud product, convective versus stratiform clouds and precipitation rates, should be discussed.

The ERA-40 precipitations and cloud properties have been already evaluated and some of the resulting studies are reported. The following text has been added to paragraph 4.1: “The precipitation is taken from the ECMWF ERA-40 data, which have been evaluated using satellite and gauge measurements and reported in Hagermann et al.

ACPD

9, C11037–C11043, 2010

(2005) and in more recent works. The precipitations over the oceans are overestimated compared to measurements, which are very uncertain. Over land the precipitations are much closer to the observations although slightly overestimated. Betts et al (2009) compared the ERA-40 data with observations in the Amazon Basis and found that the model underestimates the precipitations during the wet season and overestimates in the dry season. Another precipitation evaluation was done for China and ERA-40 resulted in underestimating precipitations in most years (Ma et al., 2009). Betts et al. evaluated also the cloud cover using satellite measurements and showed that it has the same seasonal cycle of the retrieved data underestimating it in the second part of the year."

5) BC in remote regions strongly depends on biomass burning. As this study is limited to the years 2002 – 2003, the biomass burning patterns of those years and their impacts on the involved stations should be considered.

We have available as actual BC concentration measurements of 2002 and 2003 from the remote stations only data from Alert and Zeppelin, which are influenced mainly by the anthropogenic sources. All other measurements from remote sites are climatological data, averages of former years, therefore they do not show characteristics that can be associated to the biomass burning patterns of the year 2002 and 2003. Also the biomass burning emission inventories used in the model are climatological data. Therefore it is not possible from the model output, nor from the BC concentration measurements to spot the actual patters that could be related to the 2002-2003 emission inventories. We have anyway added a text in paragraph 3.2 to underline the influencing sources at remote sites: "The stations in the Arctic regions are influenced by anthropogenic sources located in Europe and in Russia, which impact mainly in late winter-spring time in the Arctic Haze (Sharma et al. 2004; Bodhaine, 1995; Eleftheriadis et al. 2009). During the winter-spring period the Antarctic sites are influenced by BC biomass burning emissions taken place in the South Hemisphere and efficiently transported to Antarctica (Wolff and Cachier, 1998; Pereira et al., 2006)."

ACPD
9, C11037–C11043, 2010

6) My main concern is that this study is too simple. I believe this study could be strengthened by expanding the evaluation datasets and than more systematically testing model processes. It would be interesting to learn how the model behaves when BC is treated with more microphysical details. Therefore one could start from an externally mixed case and then gradually assign the BC particles more microphysical properties, and expand the transport and removal processes and test how this could improve the simulation. I'm lacking in this study a real understanding why the results are different. We have included the comparison to the vertical profiles as previously suggested.

The differences between the two model results resides in the availability of the hygroscopic properties and actual size of the particles, the lack of this information in the BULK case determines a constant removal of the in-cloud particles, only as function of rain. In the DYNA case the removal is dependent on the BC characteristics, and the BC surviving the removal over source areas and reaching for the remote regions is in small and hydrophobic particles, the vertical profile analysis has also lead to that. We have added the following sentence to paragraph 4.1: "The analysis of the vertical profiles in the case of 60% interstitial shows that at mid-latitudes and tropical regions the resulting BC falls between the DYNA and the BULK case. At high latitudes the 60% interstitial profile gets closer to the BULK case profile. Looking again at Figure 8 it is clear that closer to high latitudes the DYNA approach estimates an interstitial fraction larger than 60-70% up to more than 90% therefore the 60% hypothesis leads again in those areas to an overestimation of the wet removal. Therefore the major difference between the DYNA and BULK results in remote regions is due to the variable interstitial fraction and the fixed fraction, respectively."

7) The study could be extended using the AeroCom models.

The model results were commented also in comparison of the AEROCOM models in general regarding the burden and lifetime, and in particular for the application done in the article of Koch et al., 2009 in the paragraph 3.2: "Looking at the comparison of model results in the BC evaluation done in Koch et al (2009) the results of our work fall

ACPD

9, C11037–C11043, 2010

in the same range, having the tendency to overestimate in the mi-latitude and tropic cases, except for case d) where all models underestimate the concentrations at lower levels. The same models generally underestimate BC in the high latitude cases. “

Minor comments: What is the impact of the zoom over Europe and the extremely coarse resolution out-side of Europe?

In Europe the comparison to the EMEP stations gives better results when the finer resolution is taken into account, because the stations are not always representative of the regional background. For the IMPROVE stations a finer resolution of 1°x1° could better estimate the concentrations unless the stations are representative of such a large region as 6°x4°, which is not always the case.

Page24325 Line 2 - Give number for efficiency of sulphate scavenging Line 5 – How sensitive are results to the 30% BC interstitial mass assumption.

The in-cloud scavenging for BC, as for the other aerosols in BULK, is coupled to local precipitation (LSP). Therefore it is not possible to give one number since the scavenging efficiency depends on surface precipitation, cloud occurrence, (that determines where the rain is produced in the column) and the rain droplet radius (assumed 20 micron), as in Jueken et al. (2001).

The sensitivity of the model to the assumption of 30% BC interstitial fraction is discussed in paragraph 4.1

Page 24326: Below cloud removal the same for BULK and DYNA?

Yes it is the same, this has been clarified in the text in paragraph 2.1.3

Page 24327: What are particle sizes in BULK case. Where does BC radii for fossil/bio fuel come from?

The particle in BULK case are assumed to be 0.14 um, for the removal processes. The radii are from Stier et al. (2005)

ACPD

9, C11037–C11043, 2010

Page 24329: Why is the lifetime of BULK BC shorter? Shouldn't that be the other way around, as Aging makes BC more soluble?

We have answered to this point under point 3

Page 243300: give daily correlation coefficients.

Table 3 and 4 containing the daily correlation coefficients have been added.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 24317, 2009.

ACPD

9, C11037–C11043, 2010