Atmos. Chem. Phys. Discuss., 9, C11028–C11036, 2010
www.atmos-chem-phys-discuss.net/9/C11028/2010/

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

This paper examines uncertainties in model processes affected the black carbon aging and its removal rate. Using a chemical transport model. The paper tests two treatments of aging, a fixed removal rate and one based on some interactions in the model. The two treatments give a significantly different result from each other in the global average, but close to source regions, concentrations are found to be similar in both cases. The authors find a better agreement with observations with the more complex approach. The authors also test scaling wet removal rates to estimate resulting effects and find a difference. Overall, I find that this study uses treatments of particle aging and wet removal that are much less rigorous than the state of the art in modeling, so cannot give a reliable answer to the questions raised. Aside from the physical processes rep-

resented/numerical methods used, further evidence of the problems with the treatment is the fact that results are significantly different from that of a more detailed model and from what we would expect physically. Also, the study uses a chemical transport model that cannot account for feedbacks of processes, further raising questions about the accuracy of the conclusions since feedbacks would alter the rates of precipitation and internal mixing. As I believe a publication should encompass not only new information but information that represents an advance over what has been published previously, I recommend against publication of the present results, which will certainly change upon modification of the model, possibly drastically. More detailed comments are given below.

We think that there is a misunderstanding regarding the interpretation of the manuscript results, which are thought to be much different that with previous studies and we address this in point 2. Chemistry Transport Models as well as GCMs have been used for the evaluation of black carbon estimations in a recent paper by Koch et al. 2009 (ACP, 9, 9001-9026). Although the feedbacks influence the BC concentrations, the CTM models perform in the range of the GCMs. Furthermore, a version of TM5 participated in this evaluation, using the same wet removal parameterisations although a different BC description.

Papers have been already published in peer-reviewed journals on TM5:

- Krol, M., Houweling, S., Bregman, B., van den Broek, M., Segers, A., van Velthoven, P., Peters, W., Dentener, F., and Bergamaschi, P.: The two-way nested global chemistry-transport zoom model TM5: Algorithm and applications, Atmos. Chem. Phys., 5, 417-432, 2005. - Peters, W., Krol, M. C., Dlugokencky, E. J., Dentener, F. J., Bergamaschi, P., Dutton, G., Velthoven, P. v., Miller, J. B., Bruhwiler, L., and Tans, P. P.: Toward regional-scale modeling using the two-way nested global model TM5: Characterization of transport using SF6, J. Geophys. Res. - Atmos., 109, D19314, doi:10.1029/2004JD005020, 2004.

on using TM5 addressing aerosol related issues: - De Meij, A., Krol, M., Dentener, F., Vignati, E., Cuvelier, C., and Thunis, P.: The sensitivity of aerosol in Europe to two different emission inventories and temporal distribution of emissions, Atmos. Chem. Phys., 6, 4287-4309, 2006. - M. Karl, K. Tsigaridis, E. Vignati and F. Dentener: Formation of secondary organic aerosol from isoprene oxidation over Europe, ACP, 9, 7003-7030, 2009 - E. Marmer, F. Dentener, J. v. Aardenne, F.Cavalli, E. Vignati, K. Velchev, J. Hjorth, F. Boersma, G. Vinken, N. Mihalopoulos, and F. Raes: What can we learn about ship emission inventories from measurements of air pollutants over the Mediterranean Sea? ACP, 9, 6815-6831, 2009

on participation to model intercomparisons: - Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, T., Kloster, S., Koch, D., Kirkeva?g, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 6, 1777-1813, 2006 - Fiore A, Dentener F, Wild O, Cuvelier C, Schultz M, Hess P, Textor C, Schulz M, Doherty R, Horowitz L, Mackenzie I, Sanderson M, Shindell D, Stevenson D, Szopa S, Van Dingenen R, Zeng G, Atherton C, Bergmann D, Bey I, Carmichael G, Collins W, Duncan B, Faluvegi G, Folberth G, Gauss M, Gong S, Hauglustaine D, Holloway T, Isaksen I, Jacob D, Jonson J, Kaminski J, Keating T, Lupu A, Marmer E, Montanaro V, Park R, Pitari G, Pringle K, Pyle J, Schroeder S, Vivanco M, Wind P, Wojcik G, Wu S, Zuber A. Multi-Model Estimates of Intercontinental Source-Receptor Relationships for Ozone Pollution. JOURNAL OF GEOPHYSICAL RESEARCH 114; p. D04301-1 D04301-21. JRC46808, 2009 - Koch, D., Schulz M., Kinne S., McNaughton C., Spackman J.R., Balkanski T., Bauer S., Berntsen T., Bond T. C., Boucher O., Chin M., Clarke A., De Luca N., Dentener F., Diehl T., Dubovik O., Easter R., Fahey D. W., Feichter J., Fillmore D., Freitag S., Ghan S.,. Ginoux P, Gong S., Horowitz L., Iversen T., Kirkevåg A., Klimont Z., Kondo

Y., Krol M., Liu X., Miller R., Montanaro V., Moteki N., Myhre G., Penner J.E., Perlwitz J., Pitari G., Reddy S., Sahu L., Sakamoto H., Schuster G., Schwarz J. P., Seland Ø., Stier P., Takegawa N., Takemura T., Textor C., van Aardenne J. A., and Zhao Y.: Evaluation of black carbon estimations in global aerosol models, Atmos. Chem. Phys., 9, 9001–9026, 2009

and using TM5/M7: - Vignati, E., Facchini, M.C., Rinaldi, M., Scannell, C., Ceburnis, D., Sciare, J., Kanakidou, M., Myriokefalitakis, S., Dentener F., and O'Dowd C.D.: Global scale emission and distribution of sea spray aerosol: sea-salt and organic enrichment, Atmospheric Environment, 44, 670-677, 2010

We address the specific points below.

1) First, the empirical equation for scavenging efficiency (Equation A1, Appendix) is arbitrary, as it depends only on the precipitation rate and does not consider the hygroscopicity of particles or the probability of collision of particles of one size versus those of another with precipitation particles of different size.

In the convective storm, we assume that all aerosols are removed, both in the bulk and in the TM5/M7 model, and the hygroscopicity does not play a role. For the Large Scale Precipitation removal, in the bulk approach the hygroscopic properties are not taken into account, so that 70% of all in-cloud BC is removed. In the TM5/M7 model the wet removal is dependent on the particle hygroscopicity and size. In fact only soluble accumulation and coarse modes are removed. Below cloud scavenging is dependent on the particle size in both models. In the paper we investigate the differences between the two approaches: a very simplified one (bulk) where BC is represented without taking into account hygroscopicity and size and TM5/M7 where BC has the two properties and all processes, including wet removal, are dependent on them.

The coagulation between particles and cloud droplet has not been included because Wilson et al (2001) demonstrated that the process makes the scavenging of nucleation mode particles more efficient but it has a very limited effects on the accumulation mode

number.

Wilson, J., Cuvelier, C., and Raes, F.: A modeling study of global mixed aerosol fields, J. Geophys. Res. - Atmos., 106, 34081-34108, 2001.

As at least some global models treat the size and composition dependence of aerosol particle removal by precipitation particles of different size (e.g., Jacobson, 2004, JGR 109, D21201, doi:10.1029/2004JD004945 using the methodology in JGR 108, doi:10.1029/2002JD002691, 2003), it is difficult to see how the use of an empirical scavenging efficiency represents an advance over previous work.

We describe the formulation in the paper because it has not been done before, but we are fully aware that the formulation is simple. The scavenging parameterisation is not the advance over previous work. The scope of the article is to look into the different aspects of the BC modelling and understand the impact of certain choices of formulations or used observations.

2) The difference in methodology (both the use of a CTM versus a coupled climate-CTM and the use of empirical removal treatment) most likely contributes to the significant difference in result between the present study and the Jacobson (2004) result, wherin the author found that wet deposition removed > 90% of black carbon. Here, the result using an empirical treatment was 40-70%. It seems implausible that dry deposition could account for the remainder (60-30%) of BC removal from the atmosphere as most BC particles are too small to sediment noticeably. There is little discussion of how dry deposition is actually calculated in the model and no discussion to how it is physically plausible for up to 60% of BC to be removed by dry deposition.

There is a misunderstanding of the text regarding the amount of BC which is wet removed. In the abstract and in the results for “interstitial aerosols” it is meant the fraction available in clouds to be wet removed and not the fraction available in the system to be removed. So the text of the abstract has been changed accordingly: “The schemes for the atmospheric processing of black carbon that have been tested with the model

are (i) a simple approach considering black carbon as bulk aerosol and a simple treatment of the removal with fixed 70% of in-cloud black carbon concentrations scavenged by clouds and removed when rain is present and (ii) a more complete description of microphysical aging within an aerosol dynamics model, where removal is coupled to the microphysical properties of the aerosol, which results in a global average of 40% in-cloud black carbon that is scavenged in clouds and subsequently removed by rain, thus resulting in a longer atmospheric lifetime.”

In fact the predominant removal mechanism is wet deposition and the following line has been added to the text in paragraph 3.1: “TM5 estimates dry and wet deposition around 0.16 and 8 TgC y-1 for both approaches, as expected wet deposition is by far the predominant mechanism of removal.”

3) Further, for the model to predict wet removal accurately, it is necessary for the precipitation rate to be accurate. The authors need to show the global distribution of precipitation and compare this with observed precipitation.

The ERA-40 precipitations 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, (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)”

4) With respect to aerosol aging, the present study uses the model of Vignati et al. (2005). This represents an improvement over the schemes used in many global models

but is still an over simplification, as it treats modes instead of discrete size bins, which some other models treat (e.g., Gong et al., 2003, JGR 108, doi:1029/2001JD002002; Jacobson 2002 cited in the authors' manuscript). Gong et al. (2003) found that a minimum number of sections is needed to represent aerosol microphysical processes reasonably, more than the few modes treated here.

In Vignati et al (2004) M7 is compared to a detailed sectional model having 46 classes for the externally mixed species and 46x46 for the mixed particles. The result of the comparison was that M7 captures the size distribution and median of the aerosol parameters more than adequately.

5) The present model assumes a single coagulation kernel when coagulation between modes is considered, whereas in reality, modes consist of particles of different composition and size, where the coagulation kernel varies as a function of size and composition. Vignati et al. (2005) shows a comparison of the coagulation scheme against an analytical solution for total number, but in that scheme, the coagulation kernel is constant and the actual size distribution is not shown. The authors should at a minimum show a realistic size distribution and compare the modal treatment of coagulation with a sectional and/or analytical solution when the coagulation kernel in the sectional treatment varies as a function of particle size (thus, the modal coagulation kernel must represent some integrated value over the section value).

In Vignati et al (2004) figure 3 shows a comparison of the evolution of the size distribution and particle concentration from M7 and the sectional model using a calculated coagulation coefficient as function of the particle size both for M7 and the sectional model.

6) Treatment of condensation with a modal method is also a problem, as condensation varies as a function of particle size and composition. A sectional method can account for Raoult's law and the Kelvin effect, but a modal method cannot. The authors need to discuss and quantify to the best extent they can the inherent errors in treating con-

densation with a modal scheme versus a sectional scheme in cases other than ideal cases (e.g., Vignati et al., 2005).

For condensation we have calculated the mass transfer to and from atmospheric particles depending on their size relative to the mean free path of the vapour molecule: for relative large particle the mass transfer takes place in the continuum regime and for small particle in the kinetic regime (Seinfeld and Pandis, 1998). For matching the continuum and kinetic regimes the formulation of Fuchs (1964) was used. In Vignati et al (2004) Figure 4 shows the comparison between M7 and the sectional model; size distributions and particle concentrations are compared in case of condensation of sulphuric acid on BC particles. In Figure 5 of the same article the comparison is done between M7 and the sectional model allowing both condensation and coagulation (with coagulation coefficients depending on the particle size) to take place.

7) The paper states that "The model transport has been extensively validated using Rn and SF6." However, there is no discussion of the actual advection scheme used nor whether it has been evaluated even in one dimension against peak-preserving schemes. If no reference for such a comparison exists, the authors should perform a comparison and present the results. Transport schemes in global CTMs are notoriously diffusive, often losing 30% of their peaks in 50 grid cells of advection. Rn and SF6 tests are not sufficient for determining diffusivity of such schemes. The authors should also show the modeled globally-averaged vertical distribution of black carbon to illustrate whether the vertical profile is diffusive or not.

The scheme used in TM5 is the slope scheme in Russell et al (1981). The scheme has been evaluated also in Petersen et al. (1998). TM5 has been evaluated also in the paper of Krol et al (2005). The following text has been added to the paragraph 2.1.1: "TM5 uses the slope scheme for the advection calculations (Russel et al. 1981), scheme that was evaluated also in Petersen et al. (1998)."

Russell, G. L. and Lerner, J. A.: A new finite-differencing scheme for the tracer transport

equation. J. Appl. Meteorol., 20, 1483-1498, 1981

Petersen, A. C., Spee, E. J., Dop, H. v., and Hundsdorfer, W.: An evaluation and intercomparison of four new advection schemes for use in global chemistry models, J. Geophys. Res., 103, 19 253–19 269, 1998

Krol M., Houweling, S., Bregman, B., van den Broek, M., Segers, A., van Velthoven, P., Peters, W., Dentener, F., and Bergamaschi, P. (2005). The two-way nested global chemistry-transport zoom model TM5: algorithm and applications. Atmos. Chem. Phys., 5(2):417-432.

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