Black carbon aerosol optics in chemical data assimilation and climate forcing studies

M. Kahnert*

Swedish Meteorological and Hydrological Institute, Folkborgsvägen 1, 601 76 Norrköping, Sweden.

Optical properties of freshly emitted black carbon aerosols are computed with a numerically exact method for fractal aggregates. Computations are performed for 14 bands in the wavelength range 0.2−12.195 µm. The results are integrated into the aerosol optical observation operator of the Multiple-scale Atmospheric Transport and CHemistry modelling system MATCH. 3D-fields of ensemble-averaged aerosol optical properties computed with MATCH are coupled to radiative transfer calculations to assess the regional radiative forcing effect of black carbon over Europe. The results indicate that the forcing estimates obtained with the aggregate model can be up to a factor of 2 higher than corresponding estimates obtained with the homogeneous sphere approximation, which is still commonly employed in chemical transport and climate modelling.

INTRODUCTION

The authors of the most recent IPCC report have identified the direct interaction of electromagnetic radiation with aerosols as one of the major sources of uncertainty in assessing the radiative energy balance of the atmosphere [1]. Most aerosol species counteract the positive forcing of greenhouse gases. However, black carbon (BC) originating from soot and biomass burning emissions has been identified as the second most important primary cause for global warming after CO_2 [2]. BC global forcing estimates range between 0.4 to 1.2 W/m^2, which is as much as 55 % of the forcing of CO_2, and more than the forcing caused by all other anthropogenic greenhouse gases combined [1]. BC aerosols increase the energy in the atmosphere, reduce the radiative flux at the surface, and increase the radiative forcing at the top-of-the-atmosphere. BC is a short-lived warming agent, so emission reductions would have an immediate effect to slow down global warming. Therefore the radiative impact of this aerosol species is of considerable interest for both researchers and policy makers.

The trend in modern climate modelling is to build Earth system models in which aerosol fields are treated dynamically by coupling a chemical transport model (CTM) to an atmosphere-ocean circulation model. The treatment of aerosol optical properties in such a model needs to be geared to the output of the CTM. This usually requires that all computations need to be based on volume-equivalent size comparisons.

Elevated concentrations of BC in ambient air are also a public health concern, as they can give rise to increased morbidity and mortality. Air pollution modelling relies on the use of a CTM, which one usually constrains by use of chemical data assimilation of both in situ and satellite observations of aerosols. Assimilation of remote sensing observations relies,

*Michael Kahnert (michael.kahnert@smhi.se)

again, on coupling an aerosol optics model to the output of the CTM. This paper deals with computations of BC optical properties suitable for chemical data assimilation and Earth system climate modelling.

PHYSICAL PROPERTIES OF FRESHLY EMITTED BC

According to a recent critical review of available measurements, the refractive index and mass density of BC aerosols encountered in the atmosphere vary within a range that is considerably smaller than previously assumed [3]. The variation of the refractive index of atmospheric BC can be mainly explained by varying amounts of void fractions in the carbonaceous material, and to a much lesser extent by a variation in the sp^2/sp^3 electronic structure. The measurements in Ref. [4] are reasonably representative for atmospheric BC and agree well with void-fraction simulations of the refractive index [3]. The mass density is around 1.8 g/cm^3. Both measurements and modelling studies agree in that the fractal dimension of the BC aggregates lies around 1.82, and the fractal prefactor can be assumed to be around 1.27 [5]. Fractal dimensions larger than 2 are not representative for fresh BC aerosols. They are more typical for aged BC aggregates. The radius of the primary monomers, of which the aggregates are composed, varies between 10-25 nm. In this range the mass absorption cross section of BC aggregates is not sensitive to the monomer size. However, our sensitivity studies indicate that a monomer radius of 25 nm yields the most reasonable results for the single-scattering albedo.

Model geometries were computed by the random cluster generation algorithm written by Mackowski [6]. Figure 1 shows a model aggregate consisting of 1000 monomers.

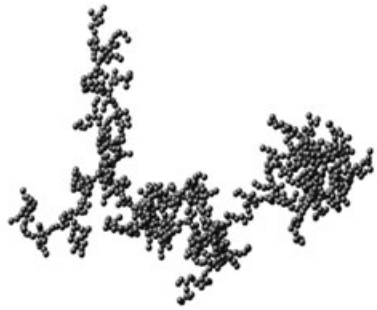

Figure 1. Black carbon model aggregate consisting of 1000 monomers.

OPTICAL PROPERTIES OF BC AGGREGATES

We tested two superposition T-matrix codes, one written by Mackowski and Mishchenko [7] and another written by Xu and Gustafson [8]. The latter offers, in addition to an exact solver, a fast approximate solution method based on limiting the interaction among the monomers. However, we found that the approximate method was not sufficiently accurate for our purposes. The exact method in Ref. [8] gave results that agreed well with those obtained with the exact method described in Ref. [7], and the computation times were

comparable. For the broadband computations, we employed the code by Mackowski and Mishchenko [7].

Computations were performed for aggregates consisting of up to 1000 monomers, and for 16 wavelengths in the range between 0.2 µm−12.195 µm. The spectral range and division points were tailored to the most recent version of the radiation model employed in the Integrated Forecasting System (IFS), which is the global numerical weather prediction model operated at the European Centre for Medium Range Weather Forecast (ECMWF). The new global Earth system climate model EC-EARTH currently developed at ECMWF is based on IFS. As an example, Fig. 2 shows results for a wavelength of 440 nm. Corresponding results for volume-equivalent spheres are also shown.

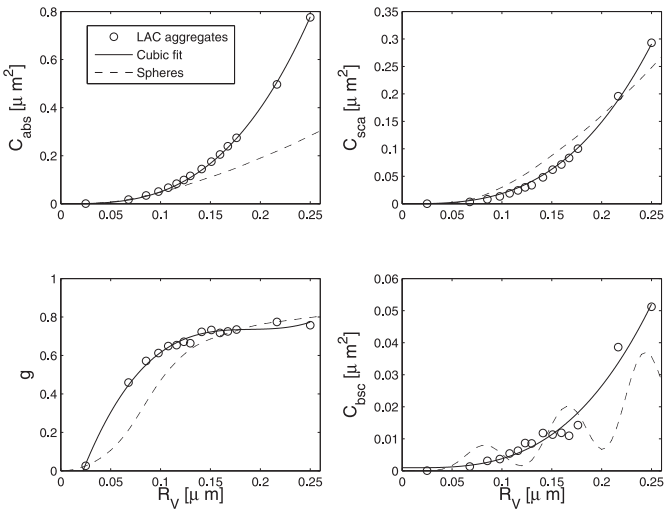

Figure 2. Optical properties as a function of volume-equivalent radius R_V at λ=440 nm.

The use of the homogeneous sphere approximation (HSA) is still common practice in climate research and chemical transport modelling. As can be seen in Fig. 2, over a large part of the size range the HSA strongly underestimates absorption, overestimates total scattering, and underestimates the asymmetry parameter (overestimated side-scattering).

We included the computed optical properties into the optical observation operator of the Multiple-scale Atmospheric Transport and CHemistry modelling system (MATCH) [9], which is a regional CTM. The optical properties of all aerosol components other than BC are computed with the HSA. The optical properties computed with MATCH are coupled to the radiative transfer model DISORT [10] to compute spectral radiative net flux F_λ. By repeating the computations in the absence of BC, one obtains a spectral reference net flux F_λ^0, from which one obtains the spectral radiative forcing effect $\Delta F_\lambda = F_\lambda - F_\lambda^0$. As an example, Fig. 3 shows ΔF_{440} at a wavelength of 440 nm. In this case, we have an aerosol layer at altitudes up to 2−3 km. The BC aerosols give rise to a positive radiative forcing above the aerosol layer, and a negative forcing at the surface, as expected. At the top of the atmosphere, the forcing effect estimated with the aggregate model is twice as high as that computed with the HSA.

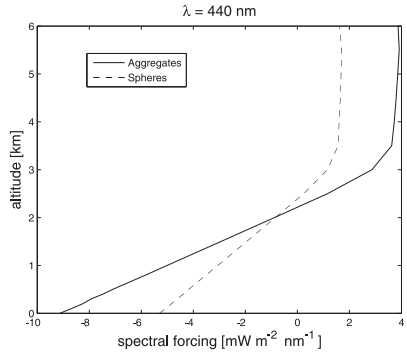

Figure 3. BC radiative forcing as a function of altitude at a latitude of 48.2° N and a longitude of 2.3° E, computed with MATCH and DISORT for 25 July 2006. The solid and dashed lines represent results obtained with the aggregate model and the HSA, respectively.

Acknowledgments: The author is grateful to D. Mackowski for providing his program for aggregated-particle generation. B. Å. S. Gustafson, D. Mackowski, M. Mishchenko, and Y. Xu are acknowledged for making their electromagnetic scattering programs publicly available.

REFERENCES

[1] P. Forster et al. Changes in atmospheric constituents and in radiative forcing. In: *Climate Change 2007: The Physical Science Basis*. S. Solomon et al. (eds.). Cambridge University Press, Cambridge (2007).

[2] V. Ramanathan and G. Carmichael. Global and regional climate changes due to black carbon. Nature Geoscience **1** (2008).

[3] T. C. Bond and R. W. Bergstrom. Light absorption by carbonaceous particles: An investigative review. Aerosol Sci. Technol. **40** (2006).

[4] H. Chang and T. T. Charalampopoulos. Determination of the wavelength dependence of refractive indices of flame soot. Proc. R. Soc. Lond. A **430** (1990).

[5] C. M. Sorensen and G. M. Roberts. The prefactor of fractal aggregates. J. Colloid. Interface Sci. **186** (1997).

[6] D. W. Mackowski. Calculation of total cross sections of multiple-sphere clusters. J. Opt. Soc. Am. A **11** (1994).

[7] D. W. Mackowski and M. I. Mishchenko. Calculation of the T matrix and the scattering matrix for ensembles of spheres. J. Opt. Soc. Am. A **13** (1996).

[8] Y. Xu and B. Å. S. Gustafson. A generalized multiparticle Mie solution: further experimental verification. J. Quant. Spectrosc. Radiat. Transfer **70** (2001).

[9] C. Andersson, J. Langner, and R. Bergström. Interannual variation and trends in air pollution over Europe due to climate variability during 1958-2001 simulated with a regional CTM coupled to the ERA40 reanalysis. Tellus **59B** (2007).

[10] K. Stamnes, S.-C. Tsay, W. Wiscombe, and K. Jayaweera. Numerically stable algorithm for discrete-ordinate-method radiative transfer in multiple scattering and emitting layered media. Appl. Opt. **27** (1988).