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Combining external and internal mixing representation of atmospheric aerosol for optical properties calculations: focus on absorption properties over Europe and North America using AERONET observations and AQMEII simulations

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The calculation of optical properties from knowledge of the composition and abundance of atmospheric aerosol implies a certain number of assumptions. First and if not known or explicitly simulated, a size distribution must be assigned to each aerosol component (e.g. sulfate-like inorganic ions, organic and back carbon, soil dust, sea salt). Second, physical-chemical properties such as the shape, density, complex refractive index, and hygroscopic factors must be associated to each aerosol species. Third, a representation of how the aerosol species combine together must be made: among those, the most popular are the assumptions of external mixing, in which each particle is assumed to be formed of a single compound and the optical properties may be calculated separately for each species, or of internal core-shell arrangement, in which each particle consists of a water-insoluble core coated with a water-soluble shell and that requires more elaborate calculations for optical properties.

Previous work found that the assumption on the mixing state (external or core-shell internal) is the one that introduces the highest uncertainty, quantified in about 30% uncertainty on the calculation of monthly mean aerosol optical depth (AOD) and single-scattering albedo (SSA). The external mixing assumption is generally more reasonable for freshly emitted aerosol, while the internal mixing case is associated with aged aerosol that had the time to form the coating around the core. Both approximations are thus regarded as valid, but in general a combination of the two mixing states may be expected in a given air mass. In this work, we test a simple empirical parameterization of the fraction of internally mixed particles (F_{in}) in a generic air mass. The F_{in} fraction is calculated in two alternative ways, one exploiting the NO_z to NO_x ratio (proxy of the photochemical aging), and the other using the relative abundance of black carbon with respect to other aerosol components (proxy of the coating formation).

We compare sunphotometer observations from the AERosol RObotic NETwork (AERONET, http://aeronet.gsfc.nasa.gov/) across Europe and North America for the year 2010 with simulations from the Air Quality Modeling Evaluation International Initiative (AQMEII, http://aqmeii.jrc.ec.europa.eu/). The calculation of optical properties from simulated aerosol profiles is carried out using a single post-processing tool (FlexAOD, http://pumpkin.aquila.infn.it/flexaod/) that allows explicit and flexible assignment of the underlying assumptions mentioned above. We found that the combination of externally and internally mixed particles weighted through the F_in fraction gives the best agreement between models and observations, in particular regarding the single-scattering albedo.