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Use of aerosol optical properties for chemistry-transport model evaluation

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This study presents an aerosol optical scheme developed in the chemistry-transport model CHIMERE dedicated to calculate optical properties of particles. Such developments are very helpful as they complement the usual validation with PM (Particulate Matter) ground-based measurements by using surface (AERONET/PHOTONS network) and satellite (MODIS) remote sensing observations. To reach this goal, Aerosol Optical Thickness (AOT), column-averaged Single Scattering Albedo (SSA) and asymmetry parameter (g) are calculated at 440 nm, 675 nm, 870 nm and 1020 nm (AERONET wavelengths) under three hypotheses on the particle mixing state (external, internally homogeneous and core-shell). Furthermore and in addition to optical calculations, an original development has been made to estimate column volume size distributions in CHIMERE, directly comparable with AERONET retrievals. Comparisons between simulations and observations are made over Western Europe for the year 2003 but also for two specific cases focused on ammonium nitrate and on secondary organic aerosols.

Results indicate that modeled SSA is fairly constant with increasing wavelength while it is very sensitive to particle mixing. Indeed, the core-shell SSA exhibits close match (at the annual mean) with AERONET values compared to external and internally homogeneous mixings. Concerning the asymmetry parameter, discrepancies between observations and simulations for the three mixings remain small at the four wavelengths with up to 10–15 % differences (at the annual mean), which could indicate good consistencies between observed and modeled particle type. Concerning the aerosol optical thickness, comparisons indicate that the seasonal cycle of modeled AOT (few sensitive to particle mixing) agrees well with AERONET observations (correlations in the range 0.50–0.74). However, modeling results reveal also some biases according to seasons. In fall, winter and early spring, AOT_{tot} and AOT_{fine} AERONET values are well reproduced by the model (with small negative biases) compared to late spring and summer periods. Results obtained for a pollution episode of ammonium nitrate (March 2003) reveal that the CHIMERE model rather well estimates AERONET fine mode volume size distribution, leading to good agreements between modeled and observed AOT. In parallel, the overestimation of hydrophilic ammonium nitrate concentrations during this episode leads to important discrepancies between AERONET and core-shell SSA, due to the thickness of the aerosol shell.

In summertime, uncertainties in modeling secondary organic aerosol formation during warm days cause an inconsistent simulated aerosol volume size distribution (for both the internal and external mixings). This leads to a weak spectral dependence of modeled AOT_{fine} in contradiction with observations and to an important underestimation of the total and fine AERONET AOT. Due to elevated temperatures over Western Europe during late spring 2003, this discrepancy in modeled volume size distribution could also explain the underestimation of AOT during this period.

Adsorption of SOA gaseous precursors onto background coarse particles over rural areas could be a reason of the discrepancies observed on the volume size distribution. In parallel, other processes not yet taken into account in CHIMERE, such as homogeneous nucleation of gaseous VOC or their nucleation with sulfuric acid, could form finest SOA with higher visible light extinction efficiency. In addition, underpredicted modeled concentrations of gaseous VOC likely to nucleate over existing fine aerosols could also explain the underestimation of AOT.