

Black carbon ageing over Europe during LONGREX-EUCAARI campaign

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Black Carbon (BC), a byproduct of incomplete combustion of fossil fuels and biomass, is a strong absorber of solar radiation and is considered to make important contributions to the radiative forcing of the atmosphere. BC particles may exist as either insoluble particles, or in mixtures that are considered partly soluble. BC in a soluble mixture is more efficiently removed by dry and wet deposition processes, and has a shorter atmospheric residence time (Zuberi et al., 2005). Following emissions, BC is subject to atmospheric processes, which include oxidation, condensation and coagulation. These processes, termed the ageing processes, act to change the solubility of the BC aerosol. Therefore, parameterization of the BC ageing processes in global models is a key issue to the determination of the atmospheric abundance of this aerosol.

Also the radiative properties of BC depend strongly on the mixing state (Bond et al., 2006). Currently in most large-scale models, primary BC particles are emitted as separate particles and are assumed to transfer into the internally mixed mode dependent on the amount of secondary products that have condensed. Jacobson (2000) conducted a model simulation and reported that the radiative forcing of BC is +0.27W/m² when it is externally mixed, but this is enhanced to +0.54 W/m² when internally mixed.

The GLObal Model of Aerosol Processes (GLOMAP, Spracklen et al., 2005) is a global model that combines a complete tropospheric chemistry scheme with an advanced aerosol microphysics module. It has a comprehensive sectional scheme (20 bins ranging from 1 nm to 25 micron) and includes sea spray, sulphate, elemental and organic carbon, dust and a simple scheme for secondary organic carbon into two distributions (soluble and insoluble). Aerosol can be transferred from the insoluble to the soluble distribution via “ageing” through coagulation and condensation of sulphuric acid on the insoluble aerosol when coated by 1 molecule of sulphuric acid (monolayer).

The focus of this study is on the conversion of insoluble BC to soluble/mixed BC. This conversion is crucial since soluble particles can act as cloud condensation or ice nuclei.

The analysed period occurred in May 2008 during the ADIENT/EUCAARI-LONGREX intensive aircraft field campaign where high aerosol

loading was sampled during air mass transport over Europe. Size distribution, mass concentration and the mixing state of BC were measured using a single-particle soot photometer (SP2) flown on board the UK FAAM aircraft. The age of BC was estimated through the time series of the BC agespectrum, a product of the Lagrangian particle dispersion model FLEXPART (Stohl et al., 1998), used to determine the origin of aerosols. Backward simulations were done along the flight-path with a time resolution less than 1 minute. For each backward simulation, the relative contribution of the BC mass in 20 age bins (ranging from 1 day to 10 days) is calculated and then “fresh” and “aged” BC is estimated by a statistical analysis.

Comparisons with observations show that the parameterization of the ageing in GLOMAP is too fast (underestimation of the model in case of “aged” BC). Therefore, sensitivity runs with 10 molecules have been done (10 layer runs) and showed a better agreement with observations. A new model setup was also done to track the amount of sulfate in the soluble distribution.

Since aerosol optical properties like the single scattering albedo (SSA) strongly depend on the representation of the mixing state, the dependence of SSA on the BC ageing is also investigated.

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