Geophysical Research Abstracts
Vol. 13, EGU2011-6162, 2011
EGU General Assembly 2011

Tracer-based source-apportionment from the EUCAARI project and comparison with the EMEP model

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Particulate carbonaceous matter (PCM) is found to constitute 10-40% (mean 30%) of PM10 levels at rural and natural background sites in Europe (Yttri et al., 2007; Putaud et al., 2004). Recent reviews have highlighted the complexity of the carbonaceous aerosol both in terms composition and formation mechanisms Hallquist et al. (2009), and until recently there have been very few direct measurements which allow a determination of how much of PCM is from anthropogenic versus biogenic sources, or from primary emissions versus from secondary organic aerosol (SOA) formation. However, over the last few years a number measurement results using tracer methods have become available which have started to shed light on the important sources of PCM in Europe (Gelencsér et al., 2007; Saarikoski et al., 2008; Szidat et al., 2006, 2007, 2009).

Within the EUCAARI project, data on 14C, EC, OC and GC-MS analyses are available from four sites, K-Puszta (Hungary), Hyytiälä (Finland), Melpitz (Germany), and San Pietro Capofiume (Italy). Additional data and analyses (AMS, NMR, other) are available from a number of other sites, including for example Barcelona (Spain), or Vavihill (Sweden). The link between tracers and their associated organic carbon amounts are of course very uncertain. Following Gelencsér et al. (2007) we define both a central best-estimate value for each factor and a plausible range of uncertainty. In order to tackle the multitude of possible combinations of these uncertain parameters, we have made use of an effective statistical approach known as Latinhypercube sampling (LHS) (Iman et al., 1981). LHS approaches are somewhat similar to Monte Carlo calculations, and allow vast numbers of combinations of input variables to be computed. A Monte-Carlo simulation would involve testing all possible combinations of input parameters. LHS provides a much more effective way of sampling the data, and for our purposes provides essentially the same results as a full Monte-Carlo analysis.All valid combinations of parameters (i.e. excluding those producing negative contributions) are condensed in frequency distributions of possible solutions.

This talk focuses on the use of such tracers to shed light on the sources of PCM in Europe. We make use of methodologies similar to those used by Szidat et al. (2006, 2009), and within the EU CARBOSOL project (Gelencsér et al., 2007), in an effort to calculate the relative contributions of the primary/secondary and anthropogenic/natural sources of the carbonaceous aerosol.

Model results for SOA are also extremely sensitive to a wide range of assumptions, with little bases for choosing between different approaches. We illustrate how source-apportionment analysis can be used to discriminate between different SOA schemes, and to constrain model possibilities.