Modeling global secondary organic aerosol formation and growth: Integrating the volatility basis set into the EMAC chemistry climate model

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Organic compounds are an important constituent of atmospheric particulates. Depending on the region it contributes 20 to 90% to the total submicron particulate mass (Zhang et al., 2007). However, organic aerosol sources, atmospheric processing, and removal are very uncertain. This study aims to improve the description of organic aerosols suitable for large-scale modelling, making use of recent developments based on laboratory and field measurements. The volatility basis set, VBS (Tsimpidi et al., 2010;Lane et al., 2008), a computationally efficient framework for the description of organic aerosol partitioning and chemical aging, is implemented in the ECHAM/MESSy Atmospheric Chemistry (EMAC) model (Jöckel et al., 2006) to simulate secondary organic aerosol (SOA) formation. SOA formation from monoterpenes, sesquiterpenes, isoprene, and anthropogenic precursors will be estimated, respectively. The new OA module suggests that the semi-volatile products of the VOC oxidation reactions are grouped by their effective saturation concentration into four aerosol species. The aerosol yields used for the production of the four SOA species were measured during recent smog chamber experiments and are also given in Table 1. This four-product set was suggested by Lane et al. (2008) as being satisfactory for the representation of biogenic and anthropogenic organic matter for urban and regional pollution modeling. The new OA module also assumes that the semi-volatile organics in the gas phase continue to react with the OH radical. These chemical reactions can lead to continued SOA formation even after complete precursor consumption and may become a major source of SOA (Murphy and Pandis, 2009;Tsimpidi et al., 2010). Different assumptions regarding the water solubility of secondary organic gas, the temperature sensitivity of SOA, and the chemical aging of anthropogenic SOA by gas-phase reaction with the hydroxyl radical, have been explored. The new organic aerosol module is tested in long-term simulations covering the years 2005-2008. Model predictions with and without SOA aging are compared with data from the European Monitoring and Evaluation Programme (EMEP), the Clean Air Status and Trends Network (CASTNet), the Acid Deposition Monitoring Network in East Asia (EANET), as well as with results from the global aerosol mass spectrometer (AMS) dataset (Zhang et al., 2007). The possibility of high anthropogenic SOA production rates, that may challenge the assumption that anthropogenic volatile organic compounds are SOA precursors of minor importance on a global scale, will be investigated.

Table 1. SOA yield scenarios using a four-product basis set with saturation concentrations of 1, 10, 100, and 1000µg m^{-3} at 298 K.

SOA precursors	SOA Yields (µg m^{-3}/µg m^{-3})			
	1	10	100	1000
Higher Alkenes	-	-	0.3	-
Terminal Alkenes	0.005	0.009	0.060	0.225
Internal Alkenes	0.023	0.044	0.129	0.375
Toluene	0.011	0.257	0.750	0.964
Xylene	0.075	0.3	0.375	0.525
Monoteprenes	0.107	0.092	0.359	0.600
Sesquiterpenes	0.075	0.150	0.750	0.900
Isoprene	0.009	0.030	0.015	0.000

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