



Global scale emission and distribution of sea-spray aerosol: Sea-salt and organic enrichment

E. Vignati^{a,*}, M.C. Facchini^b, M. Rinaldi^b, C. Scannell^c, D. Ceburnis^c, J. Sciare^d, M. Kanakidou^e, S. Myriokefalitakis^e, F. Dentener^a, C.D. O'Dowd^c

^aEuropean Commission, Joint Research Centre, Institute of Environment and Sustainability, Ispra, Italy

^bInstituto di Scienze dell'Atmosfera e del Clima, CNR, Bologna, Italy

^cSchool of Physics and Centre for Climate and Air Pollution Studies, Environmental Change Institute, National University of Ireland, Galway, Ireland

^dLab. des Sciences du Climat et de l'Environnement, Gif-sur-Yvette, France

^eEnvironmental Chemical Processes Laboratory, Department of Chemistry, University of Crete, Heraklion, Greece

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ABSTRACT

The chemical composition of marine aerosols as a function of their size is an important parameter for the evaluation of their impact on the global climate system. In this work we model fine particle organic matter emitted by sea spray processes and its influence on the aerosol chemical properties at the global scale using the off-line global Chemistry-Transport Model TM5. TM5 is coupled to a microphysical aerosol dynamics model providing size resolved information on particle masses and numbers. The mass of the emitted sea spray particles is partitioned between water insoluble organic matter (WIOM) and sea salt components in the accumulation mode using a function that relates the emitted organic fraction to the surface ocean chlorophyll-*a* concentrations. The global emission in the sub-micron size range of organic matter by sea spray process is 8.2 Tg yr⁻¹, compared to 24 Tg fine yr⁻¹ sea-salt emissions. When the marine sources are included, the concentrations of modelled primary particulate organic matter (POM) increase mainly over the oceans. The model predictions of WIOM and sea salt are evaluated against measurements carried out at Mace Head (Northern Hemisphere) and Amsterdam Island (Southern Hemisphere), showing that in clean marine conditions WIOM marine emissions contribute significantly to POM values.

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1. Introduction

Marine aerosols contribute significantly to the global aerosol load and consequently influence the Earth's radiative budget, scattering the incoming solar radiation and modifying cloud properties and their interaction with the solar radiation. Sea spray particles and sulphate produced by the oxidation of dimethylsulphide (DMS) have been considered for many years to dominate the mass of marine aerosols respectively in coarse and fine size fractions, but more recently, several studies carried out in the North Atlantic and Austral Ocean have revealed that organic matter concentration shows a strong seasonal variation, dominating the chemical composition in the sub-micron size range during high biological activity periods (Cavalli et al., 2004; O'Dowd et al., 2004; Yoon et al., 2007; Sciare et al., 2008). The high percentage of water insoluble organic matter (WIOM) observed in the fine aerosol

fraction during periods of high biological activity have been associated to sea spray processes (Keene et al., 2007; Ceburnis et al., 2008; Facchini et al., 2008) and correlated to satellite derived chlorophyll-*a* concentrations (Yoon et al., 2007). Field observations have also shown that the size of the emitted sea spray particles increases during the high biological activity periods when particles have the highest WIOM content (Yoon et al., 2007).

Based on these findings, O'Dowd et al. (2008) have developed an approach that relates sub-micron WIOM, as a function of chlorophyll-*a* concentrations derived from satellite observations, to the sea-spray source function and implemented the combined organic–inorganic sea-spray source function in a regional model focusing on the North Atlantic Ocean. This formulation has been slightly revised and applied to the global scale (Langmann et al., 2008) and using monthly averaged wind speed fields the global Water Insoluble Organic Carbon (WIOC) marine sources was estimated to be 2.3–2.8 TgC yr⁻¹.

Roelofs (2008) estimated global annual emission of primary organics of 75 TgC yr⁻¹, with comparable contributions of soluble and insoluble fractions, with a sensitivity study by assuming the

* Corresponding author. Tel.: +39 0332 789414; fax: +39 0332 785837.
E-mail address: elisabetta.vignati@jrc.ec.europa.eu (E. Vignati).

geographical distribution and seasonality of organic emissions to be similar to the oceanic emission of DMS. Using satellite retrieved chlorophyll-*a*, back-trajectories and measured Organic Carbon (OC) Spracklen et al. (2008) calculated that global marine organic carbon emissions from primary and secondary sources are about 8 TgC yr⁻¹, of which the sub-micron source is of about 5.5 TgC yr⁻¹. The latter study did not include any mechanistic parameterisation for sea-spray aerosol and simply correlated OC mass to chlorophyll-*a* fields. Gantt et al. (2009) applying empirical relationships between fluxes of water soluble and insoluble organic carbon and chlorophyll-*a* estimated the total primary source of marine sub- and super-micron WIOC as 2.9 and 19.4 TgC yr⁻¹, respectively. They used parameterisations suggested by Ceburnis et al. (2008), and Smith and Harrison (1998) for sub- and super-micron sea spray sizes, respectively, and Langmann et al. (2008), for the organic fraction enrichment. The method was not evaluated against WIOM observations.

To estimate the global marine sources of WIOM the present study uses a revised combined organic–inorganic sub-micron sea-spray source function derived from O'Dowd et al. (2008) in the global Chemistry-Transport Model TM5. The size of the emitted sea spray particles is calculated using a new formulation that relates the particle dimension to the chlorophyll-*a* concentration. The estimate is done mainly for the sub-micron range for which this method can be applied. To evaluate the approach, modelled concentrations are for the first time compared with the measurements of WIOM carried out in the Northern and Southern Hemispheres, while Spracklen et al. (2008), following a different approach, compared the total OC. A qualitative estimation of the contribution of the super-micron range to the primary organic fraction is derived from the measurements.

2. Methodology

2.1. Model set-up

The TM5 model is an off-line global transport chemistry model (Krol et al., 2005) that uses in this work ECMWF Integrated Forecast System meteorology for the year 2002–2003 and can run a resolution ranging from 6° × 4° to 1° × 1°. For computational reasons the calculations were performed at 6° × 4°. However surface processes relating to for example the quantification of sea spray were calculated at a higher global resolution of 1° × 1°. In the current version, the model has a vertical resolution of 25 layers up to 0 hPa, defined in a hybrid sigma-pressure coordinate system with a higher resolution in the boundary layer and around the tropopause. The height of the first layer is approximately 50 m.

The model transport has been extensively validated using ²²²Rn and SF₆ (Peters et al., 2004; Krol et al., 2005).

Gas phase chemistry is calculated using the CBM-IV mechanism modified by Houweling et al. (1998) and solved using the EBI method (Hertel et al., 1993). Dry deposition is calculated using the ECMWF surface characteristics at 1° × 1° resolution and the resistance method (Ganzeveld and Lelieveld, 1995). In-cloud as well as below-cloud wet removal are parameterised differently for convective and stratiform precipitation, building on the work of Guelle et al. (1998), and Jeuken et al. (2001). Aerosol below-cloud scavenging is parameterised accordingly to Dana and Hales (1976).

In this work TM5 is coupled to the microphysical aerosol dynamics model M7 (Vignati et al., 2004). In M7 the particles are represented by seven internally mixed modes: four are for soluble mixed particles representing nucleation, Aitken, accumulation, and coarse mode, and three are for the hydrophobic particle (Aitken, accumulation, and coarse mode). The aerosol components are organic carbon, mineral dust, black carbon (BC), sulphate, and sea

salt. A conversion factor of 1.4 between OC and POM (Particulate Organic Matter) was used. The structure, boundaries and chemical compounds of the modes are reported in Table 1. In this application no Secondary Organic Aerosols are included. An accompanying paper (Myriokefalitakis, 2009) analyses the secondary organic component.

Nucleation, condensation of sulphuric acid and coagulation between the particles are included. The OC and BC ageing from the hydrophobic to the hydrophilic form is accomplished by considering condensation of H₂SO₄ and coagulation with soluble particles, which form a soluble shell around the hydrophobic core and the particles are moved from the hydrophobic modes to the soluble/mixed.

The hydrophobic modes of M7 do not uptake water. The change of ability to uptake water is due to the physical properties of the particle. We compare model results to measurements of water insoluble organic matter (WIOM) defined as organic matter non extractable in water solution (Cavalli et al., 2004).

In the presence of clouds the soluble accumulation and coarse modes are assumed to form cloud droplets and are thus processed within the clouds. The oxidation of SO₂ by O₃ and H₂O₂ takes place and the resulting sulphate is partitioned between these two modes in proportion to the number of particles present in the modes (Stier et al., 2005). Particles from these two modes are scavenged by rain, while the remaining modes are assumed to form interstitial aerosols, which are not removed by in-cloud scavenging.

2.2. Emission inventories

The primary emission fields of POM, BC and sulphate that are available are for mass only; it is therefore necessary to assume the emitted particle size distribution in order to derive the emitted particle numbers (see following sessions). The only gaseous compound interacting with the particles through aerosol dynamics is sulphuric acid; therefore only details on sulphur emissions are reported in the following paragraph. Dust and the remaining gaseous emission inventories as well as emission heights are from the AEROCOM model inter-comparison exercise (Dentener et al., 2006) (<http://nansen.ipsl.jussieu.fr/AEROCOM/>). Global annual emissions are reported in Table 2.

2.2.1. POM and BC emissions

The anthropogenic contributions of POM and black carbon from fossil and bio-fuel sources are from Bond et al. (2004). Large scale biomass burning emissions are from van der Werf et al. (2004) as described in Dentener et al. (2006). The number of emitted particles is calculated assuming emission number median radii of 0.015 and 0.04 μm, for fossil/bio fuel and biomass burning, respectively, and a standard deviation of $\sigma = 1.59$ (adapted from Dentener et al., 2006). POM from fossil fuel sources is considered to be insoluble and mainly originating from unburned fuel and lubricant oil, hence having a hydrophobic hydrocarbon nature (e.g., Kleeman et al., 2008). Such

Table 1

Boundaries (dry radii, r), standard deviations (σ), particle number (N) and mass (M) tracers of modes in M7.

Mode	Soluble/mixed	Hydrophobic
Nucleation $r \leq 0.005 \mu\text{m}$, $\sigma = 1.59$	N_{nuc} , M_{SO_4}	
Aitken $0.005 < r \leq 0.05 \mu\text{m}$, $\sigma = 1.59$	N_{aiti} , M_{SO_4} , M_{BC} , M_{OC}	N_{aiti} , M_{BC} , M_{OC}
Accumulation $0.05 < r \leq 0.5 \mu\text{m}$, $\sigma = 1.59$	N_{accs} , M_{SO_4} , M_{BC} , M_{OC} , M_{SS} , M_{DU}	N_{acci} , M_{DU}
Coarse $r \geq 0.5 \mu\text{m}$, $\sigma = 2.0$	N_{coas} , M_{SO_4} , M_{BC} , M_{OC} , M_{SS} , M_{DU}	N_{coai} , M_{DU}

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