Toward a new chemical mechanism in WRF/Chem for direct and indirect aerosol effects: A focus on the carbonaceus aerosols

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Abstract An update version of WRF/Chem with a new parameterization for the secondary organic aerosol (SOA) based on the volatile basis set (VBS), is evaluated over Europe in May-June 2003. The model results are compared against surface observations issued from EC/OC 2002-2003 EMEP campaign. WRF/Chem underestimates the elemental carbon (EC) and organic aerosol mass (OA) by -17% and -38%. The analysis of EC:OA ratio reveals that the underprediction may be explained by a misrepresentation of anthropogenic emissions of carbonaceous aerosols due to the coarse resolution of the inventory and by the missing of wildfire emissions. The modeled concentration of OA constituent is near constant during all the day. The predicted SOA/OA ratio has a value of 80%. The biogenic SOA (BSOA) are the 30% of the total OA mass.

Introduction

Although the organic aerosols constitute a highly uncertain effect on global radiative forcing and represent a large fraction of sub-micron aerosol mass (20-90%), their sources and evolution are still not well characterized [1]. As consequece, the chemical transport models (CTM) generally tend to underestimate the OA mass observed in the atmosphere, because the processes involved in the production and evolution of secondary organic aerosol are missed [2]. In this paper we report on a evaluation over Europe of an update SOA yields parameterization implemented in WRF/Chem model. The work is the first step toward a new chemistry option available in WRF/Chem with the and microphysics processes fully coupled with the aerosols (i.e, direct and indirect effects).

Table 1 WRF/Chem configuration.

PHYSICAL PROCESS	WRF/Chem OPTION
Microphysics	Morrison
Long-wave radiation	RRTM
Short-wave radiation	Goddard
Surface layer	Monin-Obukhov
Land-surface model	Noah LSM
Boundary layer scheme	Mellor-Yamada Nakanishi and Niino
Cumulus	New Grell scheme (G3)
Photolysis	Madronich
Chemistry model	New RACM-ESRL [2]
Aerosol model	MADE and VBS scheme for SOA [2]
Aerosol feedback	No

WRF/Chem model

In this paper we use a pre-release version 3.4 of WRF/Chem. WRF/Chem is a fully coupled "online" meteorology-chemical model, a complete description is given by *Grell et al.* [3]. A simulation is carried out in May and June 2003 over Europe on a grid extent from 35°N to 57°N in latitude and from 15°W to 27°E in longitude. The horizontal resolution is of 30 Km with 28 vertical levels extent up to 50 hPa. The initial and boundary meteorological conditions are provided by NCEP analyses every six hours. The chemical initial and boundary conditions consist of invariant-time climatological profiles. The "traditional" configuration of WRF/Chem [3] uses the SORGAM scheme to estimate the SOA formation. Several studies over USA and Europe [3-5] demonstrated that this parameterization produces very little SOA. In this work we use an update parameterization based on the volatility basis set approach, as describe by *Ahmadov et al.* [2]. The model configuration is shown in Table 1.

The anthropogenic emissions (NO_x, NMVOCs, CO, SO_x, NH_3 and unspeciated aerosol mass) are taken from EMEP inventory (www.emep.int). Elemental carbon and organic carbon emissions are provided by the Laboratoire d'Aerologie (www.aero.obsmip.fr). The emissions are adapted to chemical mechanism used in this study as described by *Tuccella et al.* [5]. Biogenic emissions are based on the MEGAN model.

Model evaluation May-June 2003

WRF/Chem simulations are compared to surface measurements of EC/OC issued from EMEP 2002-2003 campaign [7]. The observations are constituted by a daily sample per week. To take into the account the OA mass, we multiply the observed OC by a factor 1.6 [6]. Figure 1 shows the comparison among the observed mean concentrations of EC and OA at every EMEP stations and the model results. Figure 1

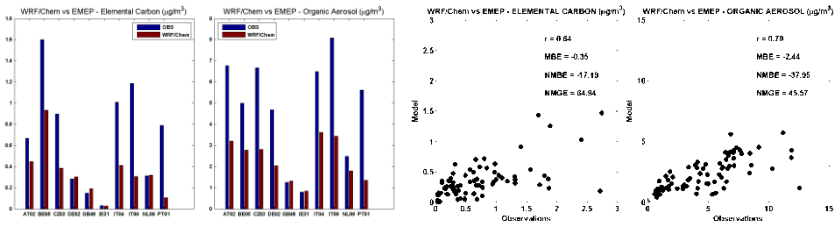

Fig. 1. (left) Intercomparison among the mean observations of EC/OA and WRF/Chem simulations at each EMEP stations in the domain. (right) Scatter plots of observed and simulated EC and OA, with the 1:1, 2:1 and 4:1 lines and some statistical indices (see the main text) are also shown.

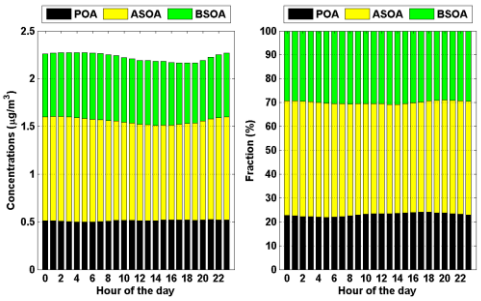

Fig. 2. Diurnal cycle of modeled OA composition (see the main text) averaged over all EMEP sites and over all days during May-June 2003. (left) Average diurnal variations in OA composition. (right) Mean diurnal variation in constituent fraction of OA.

presents also the scatter plots of observed and simulated EC and OA with the correlation coefficient (r), mean bias error (MBE), normalized mean bias error (NMBE) and normalized mean gross error (NMGE). WRF/Chem reproduces the observations of EC and OA with a r of 0.64 and 0.70 respectively. Although the model is in agreement with the lower end values of the observed distribution, tends generally to underestimate the measurements. EC is simulated with a negative MBE of -0.35 μg/m3 (-17%), while the modeled OA mass is low biased of -2.44 μg/m3 (-38%). One possible reason of the underprediction of modeled carbonaceous aerosols could be attributed to the coarse resolution of anthropogenic emissions and to the absence of wildfire emissions, which in spring time contribute to 5-35% of the European aerosol optical depth [8]. Indeed the analysis of EC:OA ratio reveals that WRF/Chem underestimates the observed correlation and slope of EC:OA ratio. The modeled values are 0.60 and 2.69 respectively, against 0.77 and 3.63 measured. Moreover the reader should also consider that the configuration of the model used here treats the inorganic and organic aerosols only in the nuclei and accumulation mode, while the data EMEP are referred to PM_{10} mass. *Putaud et al.* [9] shown that EC and OA over Europe contribute to 1-10% and 5-15% respectively to PM_{coarse} mass.

Figure 2 shows the mean diurnal cycle of modeled OA composition at EMEP station. The concentrations of primary OA (POA), anthropogenic SOA (ASOA) and biogenic SOA (BSOA) are near constant throughout the day. This can be attributed to the errors in the boundary layer dynamic due to the coarse vertical resolution of the model. The simulated SOA/OA ratio is about 80%, against 50-80% observed over Europe [1]. Moreover the average predicted BSOA/SOA ratio is about 30%, but it is about 50% at the stations in South Europe.
Future work will consist to include the direct and indirect aerosol effects in the new chemical option used in this work and validate it over Europe.

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