On the interplay between upper and ground levels dynamics and chemistry in determining the surface aerosol budget

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Abstract We use the WRF/Chem model to interpret observations of the aerosol concentration and its chemical composition both at surface level and along vertical profiles performed during an intensive campaign in July 2007 in Milan urban area. The model is added with a new diagnostic for aerosol budget analysis, building on that available for gas species, in order to study the contribution of upper levels processes on the aerosol formation at ground level. The analysis illustrates a quite variegated evolution of budget terms, which we found to depend strongly on the hour of the day, the vertical level, the aerosol compound, and the aerosol size. Primary components are generally emitted near the ground and rapidly transported by turbulent motions to the upper levels, where they gradually disperse and age. For some secondary components, such as nitrate, we calculate a net chemical destruction in the bottom layers, as opposed to a net chemical production higher in the boundary layer, which supply new material to ground level aerosol through turbulent mixing.

Keywords: aerosol, vertical, boundary layer, budget analysis.

Model Setup

WRF/Chem is a fully coupled “online” meteorology-radiation-chemistry model [1]. We use three nested domains at 30, 10, and 2 km horizontal resolution covering Europe, Northern Italy, and Po Valley, respectively, and 32 vertical levels extending 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 time-invariant climatological profiles. We use an updated parameterization for the generation of secondary organic aerosols (SOA) based on the volatility basis set approach, as described in Ahmadov et al. [2]. More details on model configuration are reported in Table 1.

The anthropogenic emissions (NO_x, NMVOCs, CO, SO_x, NH_3 and unspeciated aerosol, elemental carbon and organic carbon) are taken from the TNO inventory at 7 x 10 km over Europe. The emissions are adapted to the chemical mechanism used in this study as described by Tuccella et al. [3]. Biogenic emissions are based on the MEGAN model. The chemidiag diagnostic option for gases [4] is added with aerosol species and processes, and used to build the budget analysis.

Table 1 WRF/Chem main options.

PHYSICAL PROCESS	WRF/Chem OPTION
Microphysics	Morrison
Long-wave radiation	RRTMG
Short-wave radiation	RRTMG
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 feedbacks	No

Results

In Table 2 we show statistical indices [5] of comparison of simulation on the inner domain at 2 km resolution with meteorological and chemical variables at the measurements site, located near downtown Milan, in the University campus of

Milano Bicocca (45°31’19”N, 9°12’46”E). The meteorological bias is in the range of expected values for an urban environment. Temperature is underestimated by 2.4°C, most probably because of misrepresentation of urban landuse and related energy fluxes with the default USGS database. Indeed, improvements were shown in a preliminary test employing CORINE database [6]. Relative humidity high bias is a consequence of that of temperature. Wind speed is overestimated by 0.55 m/s (39%). The bias is not systematic as for T and RH, but it concentrates nighttime (not shown), most probably because of difficulties in representing the transition from mixed to stable boundary layer in evening hours. Wind also shows larger fluctuations than observed, as denoted by an enhanced RMSE at 1.3 m/s and the null correlation.

Nitrogen oxides are underestimated by 40% during the day, but the NO morning peak is well captured, indicating a potential important role of dynamics against emissions. Ozone is overestimated by about 10%, especially nighttime and during the afternoon (not shown). The significant difference between bias and RMSE denotes the presence of both systematic and unsystematic errors. PM10 and PM2.5 display a bias of -36% and +17% respectively, and are quite well correlated with measurements (r ~ 0.6). The low bias of PM10 may indicate a missing source from road dust resuspension.

Table 2. Comparison of WRF/Chem with ground based hourly measurements for simulation period 5-18 July 2007 at University campus Milano Bicocca site (45°31’19”N, 9°12’46”E).

Variable	Bias	NMB (%)	RMSE	r
T (°C)	-2.4	-10	2.7	0.98
RH (%)	14	35	17	0.76
WS (m/s)	0.55	39	1.3	0.02
WD (°)	33	19	130	0.25
NO (ppb)	-3.0	-42	6.9	0.59
NO2 (ppb)	-9.7	-39	14	0.43
O3 (ppb)	3.2	9.8	12	0.64
PM10 (μg/m^3)	-8.8	-36	13	0.60
PM2.5 (μg/m^3)	1.9	17	5.7	0.57

In Fig. 1 we show a sample of the information derived from the budget analysis over Milan. Only the net chemical and vertical mixing terms for aerosol sulfate and nitrate are shown at noon. Sulfate is almost homogeneously produced throughout the PBL, it is deposited at the ground and mixed upward. On the other hand, nitrate is mostly produce above 800 m and destroyed below, so the nitrate concentrations we observe at the ground are actually those transported by turbulence from the upper levels.

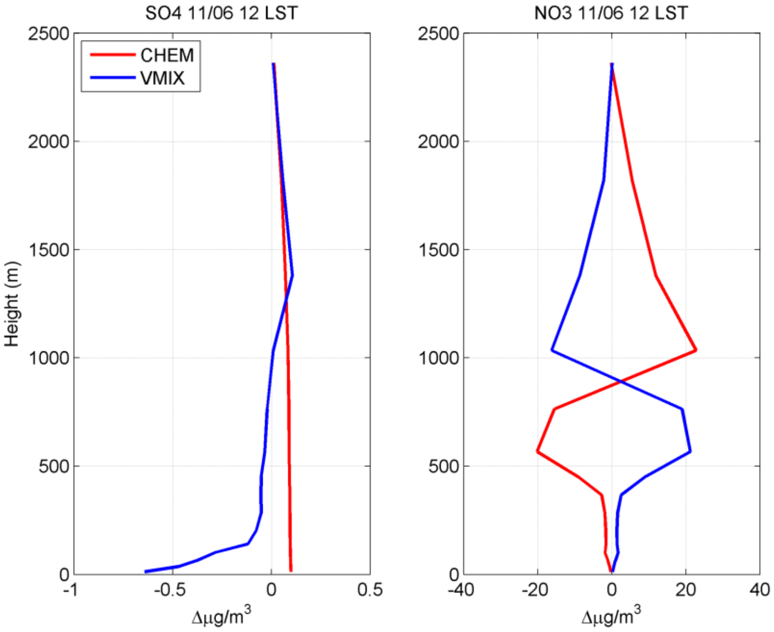

Fig. 1. Sample budget profile of particulate sulfate (left) and nitrate (right) over Milan on June 11th 2007 at 12 local solar time. CHEM is the net production due to all aerosol processes, while VMIX is the vertical mixing plus dry deposition.

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