Impact of Grid Resolution on Aerosol Predictions: A Case Study over Italy

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ABSTRACT

This study investigates the effect of grid resolution on the particulate matter (PM_{10}, PM_{2.5}) mass concentrations and its chemical composition simulated with the AMS-MINNI modelling system. The air pollution was simulated over Italy with grid resolutions of 20 and 4 km, for a whole year. The gridded emissions were produced performing speciation and space-time disaggregation of aggregated inventory data, using both land use information and anthropogenic activity-based profiles. Often, the fine grid simulations, based on high resolution gridded emissions, improved the agreement between model and measurements. In particular, the use of a fine grid improved predictions of primary species such as elemental carbon (EC), PM_{10} and PM_{2.5} mainly at urban stations. An improvement of predicted PM components and mass concentration at high altitudes sites was also observed, especially during winter. However, a general overestimation of nitrate (NO_{3}⁻) and of secondary inorganic species, more evident at night than during the day, was increased by employing a finer grid. Organic carbon (OC) was more affected by the grid resolution than the other species. At urban and kerbside stations, the use of a finer grid resulted in an overestimation of primary organic carbon aerosol (POC) but had a negligible effect on secondary organic carbon aerosol (SOC). The overestimation of carbonaceous aerosol (defined as the sum of EC, POC and SOC), at an urban station, opposite to general underestimation of this component by air quality (AQ) models, indicates that the anthropogenic emissions can contribute as much as organic model formulation at the success of simulation in reproducing experimental data.

The modelling results obtained under stable meteorological conditions characterised by weak winds, which are often encountered in the Po Valley, did not improve substantially by the increase of the modelling system resolution.

Keywords: Horizontal grid resolution; Aerosol chemical composition; PM_{10}; PM_{2.5}; Air quality modelling.

INTRODUCTION

Over the last decade, the role of air quality (AQ) models in support of air quality regulatory actions and policy (EC, 2008) has significantly increased. Models are low-cost, effective tools for identifying and estimating the source contributions to air pollution and a unique tool for developing effective plans to reduce air pollution at local and regional scales. The role of air quality forecasts for short-term mitigation actions and public information/warning is also increasing. The combined use of air quality monitoring data from networks and state-of-the-art dispersion models provides a more realistic representation of the spatial distribution (at census-block group resolution) and of the temporal evolution (annual and seasonal) of pollutants, leading to reductions in the uncertainties involved in the assessment of exposure in epidemiological studies (Physick et al., 2007; Silibello et al., 2014). For the above reasons, the scientific community
is continually searching to improve the AQ models and simulations. One of the remaining major challenges is the prediction of particulate matter (PM) concentrations that generally tends to be biased towards values that are lower than the observations. The multitude of factors that are involved in simulating PM over a certain area such as input data (meteorology, emissions and boundary conditions) and process formulation (transport and diffusion, gas-phase chemistry, aerosol chemistry and microphysics, aerosol-gas interaction, aerosol dry and wet removal, etc.) makes this very difficult. To further complicate matters, the horizontal grid spacing used over the simulated area/domain affects all the above mentioned factors.

The increase of spatial resolution often improves the meteorological simulations (Bricheno et al., 2013), but this does not always lead to significant improvement of pollutants simulations (D’Isidoro et al., 2013).

Fountoukis et al. (2013) provided a synthesis of the grid spacing effect reported in different air quality studies over United States (US) and Europe. On average, the increase of grid resolution from several tens of km to few km resulted in changes within 5% for PM mass concentrations and within 10% for organic aerosol concentrations during summer (Stroud et al., 2011). Fountoukis et al. (2013) also tested the use of two approaches to estimate the emissions of the fine grid simulation: through interpolation from the coarse grid emissions over US and by nesting high resolution (4 × 4 km) emissions with megacity emission inventories within the European coarser grid of 36 × 36 km. Both cases showed similar improvements of simulated primary PM more significant during the winter than the summer season.

The grid resolution effect on PM mass concentrations and its composition over long periods is far from being quantified by the few studies available, especially because their results depend on the simulated area and period as well as on the model formulation. The availability of measurements and emissions inventories is a limiting factor for such studies.

In this work, the impact of two grid resolutions on the simulated PM mass concentrations and PM compositions is examined over a full year over the orographically complex Italian Peninsula. The emissions over the domain were distributed using an approach similar to the ones of Fountoukis et al. (2013), see next section. Differently from Fountoukis et al. (2013), nesting with local high resolution inventories was not applied here. Nevertheless, the procedure works coherently in the whole domain with a high detail in the activity classification thereby producing a uniform emission input at 4 km spatial resolution. Data from several field campaigns published in literature and public databases were used to assess the effect of fine grid simulation on daily/nightly, seasonal and annual concentrations.

DESCRIPTION OF MODELLING SYSTEM AND SETUP SIMULATIONS

The simulations were performed with the AMS-MINNI modelling system (Mircea et al., 2014) configured with the SAPRC-90 gas-phase chemical mechanism (Carter, 1990) and the AERO3 aerosol module (Binkowski and Roselle, 2003). In order to save computation time and storage, the fine grid simulation (4 × 4 km) used five computational domains covering the Italian territory (Fig. 1) (Vitali et al., 2010, Mircea et al., 2010) while the coarse resolution simulation (20 × 20 km) covers both sea and land as delimited by the biggest rectangle. The simulations were carried out for the year 2007, with 16 terrain-following levels irregularly spaced from the ground to 10000 m above surface level, with the first four layers placed at 20, 75, 150 and 250 m. The initial and boundary conditions (IC/BC) of chemical species for coarse grid simulation were derived from the EMEP MSC-W model output (50 × 50 km, every 3 h) while the fine grid simulation was nested in the coarse grid simulation.

The meteorological fields were produced using the prognostic non-hydrostatic model RAMS (Regional Atmospheric Modeling System) (Cotton et al., 2003), which allows to perform high resolution simulations by means of two-way nested domains. The model was employed to produce both 20 km and 4 km resolution fields. The simulations were carried out in the nudging mode, i.e., assimilating reanalysis fields during the model run with a forcing term added to the dynamical equations. This drives the model to follow more closely the meteorological observations. Reanalysis fields for the assimilation were produced by means of the RAMS pre-processor ISAN (IsentropicANalysis) (Tremback, 1990), which implements an optimal interpolation method. ECMWF (European Centre For Medium - Range Weather Forecast) analyses fields together with measured surface data from the WMO (World Meteorological Organization) network were used as input data for ISAN. Reanalysis fields were used also to re-initialize simulations every ten days, in order to avoid shortcomings due to possible errors with simulation time.

A combination of the national emission inventory for Italy (ISPRA, 2005) and the EMEP (http://www.ceip.at/ms/ceip_home1/ceip_home/webdab_emepdatabase/) inventory for the surrounding countries was used for anthropogenic emissions. Biogenic emissions were assigned using the ISPR2005 dataset for Italy and the Guenther et al. (2005) global database for the surrounding countries. Emissions from port areas and shipping lanes were also included, using the data from the national and EMEP inventories respectively for national and international maritime activities. The diffuse emissions were distributed in the lowest model layers (below 50 m) with 80% in the first 20 m above ground, while the most significant point sources (including volcanoes) were treated individually by considering the plume rise effect estimated on the basis of the hourly meteorology. The ISPR2005 inventory provides the annual total emissions of aggregated species (PM$_{10}$, PMcoarse, SO$_2$, NO$_x$, CO, NMVOC and NH$_3$) at NUTS2 (http://simap.europa.eu/codes-and-nomenclatures/codes-nuts/index_en.htm) administrative level (province). A top-down spatial disaggregation of the emission inventory in the whole domain starting from a mass consistent scaling to NUTS3 administrative levels (http://simap.europa.eu/codes-and-nomenclatures/codes-nuts/index_en.htm) using quantitative proxies followed by a spatial distribution in the grid through thematic layers was
applied. This procedure works coherently in the whole domain with a high detail in the activity classification thereby producing a uniform emission field with 4 km spatial resolution. These emissions were then speciated and modulated in time according to speciation and time profiles (daily, weekly and monthly) depending on the anthropogenic activity. Diffusive emissions were disaggregated on the grids by means of gridded-based spatial proxies. Emissions from vehicle traffic, including re-suspension, were disaggregated according to road layouts and population in the absence of a coherent description of traffic flow on the whole road network. PM emissions from biomass burning were also present in the inventory. The choice of the year was determined by the amount of available information on the chemical composition of PM$_{10}$ and of PM$_{2.5}$ measurements. Fig. 1 shows as bullets the location of experimental data on the chemical composition of PM obtained during different field campaigns (Carbone et al., 2010 (PM$_{2.5}$); Perrone et al., 2012 (PM$_{2.5}$), Larsen et al., 2012 (PM$_{2.5}$, PM$_{10}$) and of the two EMEP stations (Ispra (PM$_{2.5}$, http://ebas.nilu.no/) and Montelibretti (PM$_{2.5}$, http://ebas.nilu.no/; PM$_{10}$, http://95.110.213.190/medparticles/index.php?lang=en)). More details about these stations are given in Table 1 and Table S1 in Supplementary Material. Some Italian cities (urban areas) are also shown in Fig. 1 by flags as reference points. The location of PM$_{10}$ and PM$_{2.5}$ measurements available from monitoring stations, gathered from Italian air quality database BRACE (Caricchia et al., 2003) is shown in Fig. 8(a).
RESULTS AND DISCUSSION

The analysis is limited by the impact of grid resolution on primary versus secondary aerosol species (PM components) on diurnal, seasonal and annual variations at stations located in areas with different altitudes of pollution and at different altitudes is demonstrated.

The effects of changing grid resolution on meteorological parameters and on spatial distribution of anthropogenic emissions are shown in Section 1 and Section 2 of Supplementary Material.

**Chemical Composition of Aerosol**

Mass concentrations of aerosol species (PM components) such as sulphate ($\text{SO}_4^{2-}$), nitrate ($\text{NO}_3^-$), ammonium ($\text{NH}_4^+$), organic (OC) and elemental carbon (EC) in the fine fraction ($\text{PM}_{2.5}$) were examined against literature data from Perrone et al. (2012) and Carbone et al. (2010) and against data from the EMEP database for the stations Ispra and Montelibretti. Mass concentrations of carbonaceous aerosol and contribution of primary and secondary sources to organic aerosol were compared to the data shown in Larsen et al. (2012). The grid resolution effect on chemical composition is shown using Perrone et al. (2012) and EMEP data for different seasons, using Carbone et al. (2012) data for day and night during summer, and using Larsen et al. (2012) data for primary and secondary OC sources during winter.

Fig. 2 (Table S2) compares the chemical composition of the aerosol fine fraction for simulations with low and high spatial resolution using the data shown in Fig. 2 by Perrone et al. (2012). The sites used in this comparison are an urban (MI), a plain rural characteristic of Po Valley (OB) and a high altitude typical remote site in the Alps (MI). In this graph, Ca, Mg, Fe, Al, etc. shown by Perrone et al. (2012) are included in the unknown aerosol fraction since these species are not considered by the model.

A generally very good agreement between the two simulations and measurements is observed for all the stations, with differences in the order of a few µg m$^{-3}$ in the total mass concentrations. However, there are some interesting discrepancies, which will be discussed in the following.

During summer, the fine resolution simulation, compared to coarse one, produces the same or lower concentrations of inorganic aerosol species at all stations and higher EC and OC concentrations at the urban station (MI). Moreover, during winter the fine resolution simulation produces higher concentrations of NO$_3^-$, OC and EC at the OB and MI stations and higher SO$_4^{2-}$ concentrations at the urban station (MI), only. At urban station, this enhancement of concentrations is the result of receptor proximity to the anthropogenic sources of air pollution in atmospheric conditions characterised by low dispersivity. Overall, the fine grid simulations estimate lower total PM mass concentrations than the coarse grid simulations at all stations during summer and at the high altitude station during winter and an opposite behaviour at urban and rural stations during winter. The similar response of urban and rural stations to grid change can be explained by the fact that in the Po Valley, with its particular topography, the air masses, strongly influenced by anthropogenic sources, are often entrapped over longer time periods and mixed

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**Table 1. Description of 19 sites with data on aerosol chemical composition (Fig. 1).**

<table>
<thead>
<tr>
<th>Site number</th>
<th>Name of site</th>
<th>Acronym</th>
<th>Network/Reference</th>
<th>Colour</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>High Altitude Site</td>
<td>ASC</td>
<td>Perrone et al. (2012)</td>
<td>pink</td>
</tr>
<tr>
<td>2</td>
<td>Oasi Le Bine</td>
<td>OB</td>
<td>Perrone et al. (2012)</td>
<td>pink</td>
</tr>
<tr>
<td>3</td>
<td>Milano (Torre Sarca)</td>
<td>MI</td>
<td>Carbone et al.(2010), Perrone et al. (2012)</td>
<td>pink</td>
</tr>
<tr>
<td>4</td>
<td>Ispra</td>
<td>IT04</td>
<td>EMEP</td>
<td>yellow</td>
</tr>
<tr>
<td>5</td>
<td>Montelibretti</td>
<td>MTL or IT01</td>
<td>Carbone et al.(2010), EMEP</td>
<td>yellow</td>
</tr>
<tr>
<td>6</td>
<td>Rende</td>
<td>REN</td>
<td>Carbone et al.(2010)</td>
<td>red</td>
</tr>
<tr>
<td>7</td>
<td>San Pietro Capofiume</td>
<td>SPC</td>
<td>Carbone et al.(2010)</td>
<td>red</td>
</tr>
<tr>
<td>8</td>
<td>Monte Cimone</td>
<td>CMN</td>
<td>Carbone et al.(2010)</td>
<td>red</td>
</tr>
<tr>
<td>9</td>
<td>Pavia Samnazzaro</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
<tr>
<td>10</td>
<td>Brescia</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
<tr>
<td>11</td>
<td>Cantù</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
<tr>
<td>12</td>
<td>Lodi Abbadia</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
<tr>
<td>13</td>
<td>Mantova</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
<tr>
<td>14</td>
<td>Milan-ks</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
<tr>
<td>15</td>
<td>Milan-ub</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
<tr>
<td>16</td>
<td>Milan-S</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
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<tr>
<td>17</td>
<td>Saronne</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
<tr>
<td>18</td>
<td>Sondrio</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
<tr>
<td>19</td>
<td>Bergamo</td>
<td></td>
<td>Larsen et al. (2012)</td>
<td>green</td>
</tr>
</tbody>
</table>
Fig. 2. Chemical composition of fine fraction aerosol during summer and winter, for 4 km and 20 km simulations: comparison with Perrone et al. (2012).

Throughout the valley as recently demonstrated with back trajectories and chemical characterisation of PM (Belis et al., 2011, Larsen et al., 2012, Perrino et al., 2014). At the high altitude site (ASC), the fine grid reduces the bias for NO$_3^-$ and SO$_4^{2-}$ aerosols, but not for OC and unknown aerosols (for details see Fig. S4). NO$_3^-$ overestimation and OC underestimation observed at ASC site in both seasons are also noted at the rural station, in particular during the summer, suggesting that during this season most of the aerosol at ASC site is transported from the valley. Coherently, the higher total mass concentration observed during summer than during winter can be also explained by more transport of PM from the valley favoured by more intensive convective transport and higher mixing heights during summer.

As evidenced also by other studies (Fountoukis et al., 2013), a significant improvement occurs when using finer grid to simulate EC which is emitted directly in atmosphere by anthropogenic sources, at the urban site in both seasons.

Fig. 3 (Table S3) compares the chemical composition of fine fraction aerosol (PM$_{2.5}$), for 4 km and 20 km grid
simulations, with the data shown in Fig. 4 by Carbone et al. (2010) for July 2007, from 14 to 18. This period was characterised by stagnant conditions determined by the presence of an anticyclonic circulation in the central Mediterranean area. The sites included in this study cover the whole Italy from North to South: MI, SPC, CMN (high altitude 2165 m a.s.l), MTL, REN (Fig. 1, Table 1) and they are characterized by different pollution regimes: MI is an urban station, the same as in Fig. 2, SPC is a rural station in Po Valley, CMN is a high altitude site in the Apennine mountains, MTL is a peri-urban site and REN is a rural station close to Tyrrhenian Sea. The sum of WSOM (water soluble organic matter), WINCM (water insoluble carbonaceous matter) and EC (Carbone et al., 2010) is here represented as the sum of OM and EC, carbonaceous aerosol. Fig. 3 compares the concentrations of main species contained in PM$_{2.5}$ as follows: Aitken and accumulation modes of the model against the first three impactor stages at MI, SPC and CMN, and to the first two impactor stages at MTL and REN.

![Chemical composition of fine fraction aerosol during summer time, day and night: comparison with Carbone et al. (2010).](image-url)
At all stations, except MI, the fine grid simulation reduces the overestimation of SO$_4^{2-}$ concentrations during the day. Practically no grid effect was observed for NH$_4^+$ and carbonaceous aerosol at all stations, except at the MI station, while the 4 km grid simulation estimated higher NO$_3^-$ concentrations at the MTL and MI stations, both of which are more influenced by anthropogenic emissions. During the day at the MI station, the fine grid simulation of the carbonaceous aerosol concentration shows a doubling with respect to coarse grid and to observations. The overestimation of SO$_4^{2-}$ and underestimation of carbonaceous aerosol is also observed during the night at all stations. The fine grid simulation estimates a lower total mass concentration than the coarse grid simulation at all stations, except MI and MTL, during day and night. At urban (MI) and suburban (MTL) stations, the fine grid simulation overestimates the total mass concentrations, more at MI than at MTL, both during day and night, mainly due to the overestimation of secondary inorganic aerosol species. This overestimation is higher during the night than during the day and varies by a factor 3 and 2, respectively, at MTL and by a factor 6 and 3, respectively, at MI.

The aerosol concentrations are higher at night than during the day at all the stations, apart from the CMN station. This is explained by a lower vertical pollutant transport at night, which is better reproduced by the fine grid simulation. On the contrary, at the high altitude station CMN, the observed mass concentration of fine aerosol fraction has lower values at night than during the day. This may be explained by the fact that the height of the boundary layer increases during the day, thereby transporting pollutants from the valley to mountain summit due to enhanced vertical diffusion. Only the fine grid simulation reproduces this trend, pointing out that it performs better in depicting local meteorological features. Moreover, at night, the fine grid simulates better CMN data than the coarse grid while the concentrations of NO$_3^-$ and in particular of carbonaceous PM components are underestimated during day.

At all other sites than the CMN station, the aerosol concentrations are higher at night than during the day, mainly due to the lower vertical pollutant transport at night. This feature is better reproduced by the fine grid simulation.

It is interesting to note the different response of aerosol concentrations to grid resolution at the same station, MI, and in the same period, summer (Figs. 2(a) and 3(a)). The fine grid simulation produces higher mass concentration of aerosol and of carbonaceous aerosol (OC + EC) than coarse grid simulation for a week characterised by Saharan dust transport (Fig. 3(a)) but not for the whole summer (Fig. 2(a)). This comparison clearly shows the dependence of model performances on evaluation period and the difficulties of model to reproduce the rebuild-up of anthropogenic aerosol during periods of stable meteorological conditions such as that shown in Fig. 3(a). Moreover, since Po Valley is often characterised by long periods of stagnant conditions during the winter, Figs. 2(b) and 3(b) show similar behaviour of grid resolution effect such as in Fig. 3(a).

Fig. 4 shows mass concentrations of primary and secondary organic aerosols (POC and SOC), and elemental carbon at eleven stations (Fig. 1, Table 1) located in Northern Italy during winter, as calculated by 4 km and 20 km simulations, against average source contributions to carbonaceous aerosol estimated by positive matrix factorisation (PMF) from Fig. 1 shown in Larsen et al. (2012). An extensive description of
stations and sampling periods can be found in Belis et al. (2011). Both simulations are in better agreement with the results of PMF than with the macro-tracers model (not shown here) obtained by Larsen et al. (2012). The fine grid simulation yields higher carbonaceous concentrations than the coarse grid one at all the stations, except for Pavia-Sannazzaro, as a consequence of the increases of EC and POC. At the Milano-ks and Milano-ub stations, the high resolution simulation overestimates the POC concentrations by a factor of 2. This overestimation may be the result of an erroneous spatial distribution of anthropogenic emissions inventory in the area. The largest difference between the fine grid simulation and the receptor-model results obtained by Larsen et al. (2012) is seen for the Sondrio station, located in a narrow and deep mountain valley, greatly influenced by residential heating and by wood burning fraction contributions whose emissions are not captured by the AMS-MINNI model. SOC is underestimated at all stations and has similar values for both simulations. The underestimation of SOC at rural sites such as Lodi, representative for typical contributions whose emissions are not captured by the AMS-MINNI model. SOC is underestimated at all stations and has similar values for both simulations. The differences between fine and coarse grid simulations is station-dependent, the highest difference is obtained for Milano where the fine grid simulation estimates SOA concentrations 2 times higher than the coarse grid simulation (much more than the 10% shown by Stroud et al. (2011)).

Fig. 5 compares the results of the two simulations with the measurement data at the EMEP stations Montelibretti (MTL above and in Carbone et al. (2012) and IT01 in EMEP database) and Ispra (IT04), (Fig. 1, Table 1). Montelibretti is situated in central Italy at an altitude of 48 m while Ispra of the EU - Joint Research Centre is situated in northern Italy at an altitude of 209 m. At the Montelibretti site, the daily PM sampling was carried out by means of two techniques: PM$_{10}$ single filter and diffusion line. The single-filter technique consists in the sampling of PM$_{10}$ on two independent sampling lines, one equipped with a Teflon filter, the other one equipped with a quartz fiber filter. The Teflon filter is devoted to the measurement of PM mass concentration by the beta attenuation method (and, occasionally, by gravimetry), to the subsequent determination of elements by X-ray fluorescence and, after extraction in deionized water, to the measurement of the ionic content by ion chromatography. The quartz filter is analyzed for its elemental and organic carbon content by thermo-optical analysis (NIOSH-QUARTZ temperature protocol). Details about the overall procedure are reported in Perrino et al. (2009) and Perrino et al. (2014). Diffusion lines are used in the framework of the EMEP program and consist in five annular diffusion demuders set in series for the determination of nitric acid, sulphur dioxide and ammonia, a cyclone for the determination of coarse particles (> 2.5 µm) and a filter-pack for the determination of sulphate, nitrate and ammonium in PM$_{2.5}$ (Perrino et al., 2001). This set-up allows a reliable distinction between the gaseous and the particulate phase and, in particular, the determination of the evolved fraction of ammonium salts that are lost when the sampling is carried out on single filters.

Fig. 5 shows the simulated (20 and 4 km grid resolutions) and measured concentrations of NH$_4^+$, NO$_3^-$, SO$_4^{2-}$, EC and OC for data averaged over the whole year and seasonally over winter, spring, summer and autumn. The data refer to IT04 (PM$_{2.5}$ sampled by filter-pack) and to MTL (IT01) (PM$_{10}$ sampled by the single-filter technique). NH$_4^+$, SO$_4^{2-}$ and NO$_3^-$ data measured at MTL by the diffusion line and the single-filter technique together with simulated concentrations are shown in Fig. 6. Here, for summertime, for a given spatial resolution, the averaged simulated concentrations have different values in correspondence of the two measurement techniques since the calculations were performed only when the daily measurements were available and there was a lack of single-filter data.

Fig. 5 shows that the simulated ammonium concentrations are generally higher than the one measured. The use of fine grid improves substantially the agreement with measurements at IT04 station during summertime. For the others seasons, it tends to worsen the agreement with measurements at IT04 and practically it has no effect at MTL, except for summertime. Considering the MTL measurements made by the diffusion line, the agreement between model and measurements is considerably improved (Fig. 6) since this technique is able to measure also the evolved fraction of ammonium salts.

Simulated nitrate concentrations are much higher than the measured concentrations at both stations in all seasons (Fig. 5), except MTL during summertime. As for ammonium, the fine grid simulation has no impact on the agreement with measurements at MTL but it worsens at IT04, especially during the winter. This model behavior at stations located in the Po Valley, other than the urban ones, was already discussed above. At MTL, Fig. 6 shows a better agreement between the model simulations and the measurements with diffusion line. The sampling carried out on single filters (Fig. 5) leads to the volatilization of ammonium nitrate contained in the fine fraction of PM and to a decrease of the measured values, which may be very relevant with respect to the real atmospheric concentration. This can be observed in Fig. 6, during summertime and wintertime, at low temperatures, by comparing the measurements from single-filter and diffusion lines. On the contrary, during the summer, NO$_3^-$ measurements from the single-filter technique are higher than those from the diffusion line because of the contribution of coarse nitrate salts. The comparison of model predictions with measurements from the diffusion line is more adequate since the model does not include uptake/chemistry of coarse nitrate in the present formulation.

Fig. 5 shows that the model predictions of sulphate concentrations are much higher than the observations at IT04 but show a good performance, with slightly better results for the fine grid simulation at MTL. SO$_4^{2-}$ (mostly in the form of ammonium sulphate) is not affected by the measurement technique used, PM$_{10}$ single-filter or diffusion line, (Fig. 6) since it is a thermally stable specie contained in the fine fraction of PM.
Fig. 5. Fine (4 km) and coarse (20 km) simulated concentrations vs. the observed ones at EMEP stations: Montelibretti (IT01) (PM$_{10}$ single filter technique) and Ispra (IT04), for NH$_4^+$, NO$_3^-$, SO$_4^{2-}$, EC and OC. Annual and seasonal averages in µg m$^{-3}$. 

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EC is relatively well simulated at MTL in all seasons, while it is highly underestimated during autumn and winter at IT04 (Fig. 5). EC is confined to the fine fraction in both model and real atmosphere; therefore the model comparison with PM$_{10}$ and PM$_{2.5}$ values is appropriate and justifies the well reproduction of EC in PM$_{10}$ at MTL.

The OC model predictions are much lower than the measured concentrations at both stations, in all the seasons, with the highest difference during winter (Fig. 5). At IT04, the difference may be due to low emission estimations from residential heating, wood burning fraction in particular, and due to model difficulty in reproducing stable meteorological conditions that favour the accumulation of air pollutants at surface as discussed previously. These justify also the recurrent overestimations of NH$_4^+$, NO$_3^-$, SO$_4^{2-}$ and underestimations of EC and OC at this station.

The impact of grid resolution on model predictions is high for inorganic secondary volatile species such as NH$_4^+$, NO$_3^-$; its magnitude varies with season and depends on station location. For both stations, it has a small effect on both EC and OC model predictions in all seasons, and on annual averages of all species.

Overall, both simulations at both EMEP stations capture well the seasonal variability of all aerosol species, in particular the significant differences between summer and winter concentrations and the transition during spring and autumn periods, but significant overall improvement by using a fine grid is not always observed. Moreover, the model performances depend on the measurements used as it is shown in Fig. 6 where the simulations are compared to two measurement techniques based on different size and volatility principles.

**Mass Concentrations: PM$_{2.5}$ and PM$_{10}$**

The evaluation of model performances in reproducing PM$_{2.5}$ and PM$_{10}$ mass concentrations was also carried out using the data routinely collected for stations of regional and local monitoring networks and contained in BRACE national database. Only monitoring stations with time coverage of the measured data in compliance with the Directive 50 (EC, 2008) were included in the comparison and no correction factors were applied to the concentrations.
simulated by the model. Figs. 7, 8(a) and 8(b) show the fine grid resolution effect on PM$_{10}$ and PM$_{2.5}$ mass concentrations for different type of stations: urban, suburban and rural. As expected, the fine grid simulation yields higher concentrations than the coarse one at many of the stations, in particular at the urban and suburban ones. This generally improves the agreement with the measured concentrations, but leads to overestimations at some urban stations. On the contrary, at some rural stations, the fine grid simulation yields lower PM concentrations than the coarse grid simulation, which can be explained by the redistribution of emissions, in particular emissions from domestic heating. The different response to the increase in the grid resolution may be the result of a non-linear combination between emissions and process formulation in the model that actually controls the chemical composition of aerosol: primary versus secondary species, anthropogenic versus biogenic species. Fig. 8(b) zooms in on Northern Italy for the fine grid simulation in order to better evidence the differences with respect to the coarse simulation. The simulation over the whole domain can be seen in Fig. 8(a).

Fig. 9 shows the frequency distribution of annual statistical indicators for PM$_{10}$ simulated with fine and coarse horizontal resolutions at urban BRACE stations. This statistical approach requires a minimum number of stations and thus, cannot be applied to the other stations types and to PM$_{2.5}$ stations. The mathematical formulation of statistical indicators is shown in SM (Section 3 Statistical indexes). It can be observed that refining the grid resolution generally reduces the number of stations with a high negative bias and increases the number of stations with a bias between $-5$ and $5$ µg m$^{-3}$. Thus, the increase of resolution reduces model underestimations. Moreover, refining the grid resolution also increases the number of stations with high correlations (0.6–0.8) and the number of stations with an agreement between simulated and measured concentrations within a factor 2 (fac2).

The other indicators (mage, mange and rmse) also show an improvement in model performance when the refined grid
resolution is used but there are still stations that are not well captured by the model. As showed by Righini et al. (2014) using the emissions fields, the spatial representativeness of urban sites is limited to few square kilometers, less than 4 × 4 km² used in this study. However, the use of the fine grid simulation is probably not sufficient only for a dozen of stations that have correlations (corr) and factor 2 (fac2) below 0.5, rate above 20 µg m⁻³, range above of 55% and very high rmse.

CONCLUSIONS

The state-of-the-art in air pollution modelling generally expects that an increase of the grid resolution improves simulations. The present study investigated the impact of increasing the horizontal grid resolution from 20 × 20 km² to 4 × 4 km² on the simulation of the chemical composition and mass concentration of aerosol over the Italian territory for a full year with the AMS-MINNI modelling system. The results confirmed a general improvement of the comparison of predicted vs measured data by increasing the resolution. However, an improvement was not always observed for specific sites, aerosol components, and seasons, which indicates that the AMS-MINNI modelling system can still to be ameliorated, in particular, for vertical transport and diffusion of pollutants and the parameterization of the mixing layer and predictions of transported aerosol compounds from Po Valley.

The most consistent improvement of the comparison of predicted vs. measured data was observed for the primary aerosol component of anthropogenic origin, EC. Nevertheless, the results obtained at stations located in highly polluted areas with a complex orography during the cold season indicate that the local EC emissions and meteorology can still be further improved.

Carbonaceous aerosol was generally underestimated by the AMS-MINNI modelling system compared to experimental data and overestimated only at two of the included urban and kerbside stations strongly influenced by local emissions. The overestimation is a consequence of higher POC concentrations predicted with fine grid while SOC concentrations remained unchanged. This stresses the role of emissions with respect to primary organic aerosol in highly polluted areas such as Po Valley, in particular during winter.

The impact of the grid resolution on the compounds with a dominating secondary component varied with season,
station and compound and was not always positive for the comparison of predicted vs measured data. Independent of the grid resolution used, the concentration of secondary inorganic species was generally overestimated by the AMS-MINNI modelling system especially during night.

This study shows that the annual averages of model predictions for all species (\(\text{NH}_4^+, \text{NO}_3^-, \text{SO}_4^{2-}, \text{EC} \text{ and } \text{OC}\)) are little affected by the change of grid resolution while significant changes exist for data averaged seasonally. Apart from the necessity of having an extended temporal coverage of the measurements representative for different meteorological and pollution conditions, it also point out that the use of measurement techniques based on different size and volatility principles plays a key role in the evaluation of model performances.

Overall, the use of the fine grid improved the annual predictions of PM\(_{2.5}\) and PM\(_{10}\) but not at all stations. Several factors may explain that, such as an inadequate spatial representativeness of stations according to the grid resolution of simulation, sub-grid scale effects, and the emission inventory and/or the approach used for producing gridded emissions. In addition some model aspects may also play a role, such as the parameterizations of the planetary boundary layer, and of the inorganic and organic aerosol chemistry.

The results of this study confirm that the use of high-resolution simulations based on high resolution emission inventories generally improves the AMS-MINNI modelling system predictions but also point out that further research is needed to improve the model. The required input for such improvements must come from experimental studies of the chemical composition of tropospheric aerosol distributed diurnally and seasonally in areas with different meteorological and pollution conditions.

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SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

REFERENCES


