



Impact of Emissions, Meteorology and Grid Resolution on Changes in HMs and PAHs Concentrations between 2005 and 2010 in Italy

Adani Mario^{*}, Mircea Mihaela, D'Isidoro Massimo, Gualtieri Maurizio

ENEA-Italian National Agency for New Technologies, Energies and Sustainable Economic Development, 40129 Bologna, Italy

ABSTRACT

A dynamical evaluation of the Flexible Air quality Regional Model (FARM) system was conducted to assess its ability to predict changes in heavy metals (HMs) and polycyclic aromatic hydrocarbons (PAHs) concentrations over Italy between the years of 2005 and 2010. Several simulations were performed to relate variations in the PAHs and HMs concentrations to changes in the emission inventories, meteorology or grid resolution. The results generally indicate that these concentrations have decreased through the years and the magnitude of decrease is dependent on the specific chemical species. The changes in emissions have determined the major variations of all chemicals, whereas the meteorology has caused significant changes in the concentrations of reactive species, such as benzo[a]pyrene (BaP), in high-resolution simulations, especially over complex topography. The analyses also show that while the total emitted mass of pollutants is important, its geographical distribution is fundamental to estimating changes in concentration.

Keywords: Heavy metals; Polycyclic aromatic hydrocarbons; Air quality modelling; Dynamical assessment; Sensitivity experiments.

INTRODUCTION

Micro-pollutants such as heavy metals (HMs) and polycyclic aromatic hydrocarbons (PAHs) may be a burden of risk for human and ecosystem health since some of them are toxic even at low concentrations (Lenntech, 2004), persistent and/or bio-accumulate over a period of time (UNEP/GPA, 2004). The main anthropogenic sources of these pollutants are domestic combustion, vehicle exhaust (traffic emissions), industry (production sites of metals, coke and asphalt) and power stations (UNEP/GPA, 2004). These activities have increased dramatically the air concentrations of HMs and PAHs since pre-industrial times. Therefore much effort has been made to reduce their emissions since the early 1990s, in particular for cadmium and lead (<http://www.eea.europa.eu/data-and-maps/indicators/eea32-heavy-metal-hm-emissions-1/assessment-3>). However, in urban areas and in areas close to highways and industrial regions, the concentrations of HMs and PAHs still pose a risk for human health and environment requiring more detailed analysis on national and local scales for developing effective abatement strategies.

Air quality (AQ) models are fundamental instruments for air quality management allowing the understanding of the contribution of different sources to air pollution and the effect of future emission policies and/or technologies in pollutants reduction. The reliability of AQ models for these tasks is ascertained through simulations of air quality changes for given changes in emissions (dynamic evaluation) and comprehensive model evaluation by comparing model outputs with observations (operational evaluation). At present, HMs physics is included in the EMEP/MSCE regional model of heavy metals airborne pollution (MSCE-HM) (Travnikov and Ilyin, 2005) and in CHIMERE model (Gonzalez *et al.*, 2012), while PAHs physics and chemistry are taken into account in MSCE-POP model (Gusev *et al.*, 2005) and in WRF-CMAQ-BaP (Matthias *et al.*, 2009; San Jose *et al.*, 2013). AMS-MINNI modelling system (Adani *et al.*, 2015) is the only three-dimensional Eulerian multicomponent chemistry transport model that includes both HMs and PAHs therefore representing a unique among AQS.

Very few studies employed the dynamic evaluation approach at present. Gilliland *et al.* (2008) and Napelenok *et al.* (2011) investigated the changes of ozone (O₃) levels in response to the reduction of NO_x emission in U.S. while Thunis *et al.* (2015) investigated the effects of different emissions scenarios on daily averaged PM₁₀ and daily 8-h maximum O₃ changes at three European regions: Po Valley, Benelux and south of Poland.

^{*} Corresponding author.

Tel.: +39-051-6098916; Fax: +39-051-6098675
E-mail address: mario.adani@enea.it

This work shows for the first time a dynamic evaluation for HMs and PAHs at national level. The results are based on simulations performed with the AMS-MINNI modelling system over Italy for the years 2005 and 2010. Specifically, the changes in daily modelled ground level concentrations are evaluated against the measured arsenic (As), cadmium (Cd), nickel (Ni), lead (Pb) and benzo[a]pyrene (B[a]P) concentrations at air quality monitoring stations. Additional simulations were performed in order to disentangle the effect of emission changes from meteorological ones. Finally, the model's capability to dynamically respond to emissions changes is investigated at different spatial grid resolutions.

DESCRIPTION OF ATMOSPHERIC MODELLING SYSTEM AND OF SIMULATIONS SET-UP

Air quality simulations were performed using AMS-MINNI modelling system (Mircea *et al.*, 2014, 2016) composed of the air quality model FARM (Flexible Air quality Regional Model) (Gariazzo *et al.*, 2007; Silibello *et al.*, 2008; Kukkonen *et al.*, 2012; Silibello *et al.*, 2012; Adani *et al.*, 2015), the meteorological model RAMS (Regional Atmospheric Modelling System) (Cotton *et al.*, 2003), the pre-processor SURFPro (SURFace-atmosphere interface PROcessor) (Arianet, 2011) and the emission processor EMMA (EMission MANager) (Arianet, 2014). FARM is a three-dimensional Eulerian model that accounts for pollutant transformations by advection and turbulent diffusion, gas phase and in-cloud chemistry, wet (in-cloud and below-cloud) and dry removal, and aerosol dynamics and chemistry.

The natural HM emissions associated to soil dust and sea-salt aerosols have been simulated as described in Adani *et al.* (2015). Briefly, over the land the estimated HM emission fluxes are based on the wind fields and HM concentrations measured in topsoil (Geochemical Atlas of Europe developed under the auspices of the Forum of European Geological Surveys—FOREGS [www.gtk.fi/publ/foresatlas/]).

The above mentioned system was used to perform three simulations: one for the year 2005 (E05B05M05); one for the year 2010 (E10B10M10), validated in Ciancarella *et al.* (2016); and a cross simulation using meteorology, boundary conditions and natural emissions for 2010 and anthropogenic emissions for the year 2005 (E05B10M10). For both years, air quality boundary conditions were derived from the output of EMEP/MS-C-E and EMEP/MS-C-W models while meteorological boundary conditions from ECMWF operational analysis. The anthropogenic emissions over the Italian territory were provided by the National Emissions Inventory developed by ISPRA: in particular, ISPRA (2009) was considered as representative of 2005 emissions and used for E05B05M05 and E05B10M10 simulations while ISPRA (2012) was representative of 2010 emissions and applied for E10B10M10 simulation. The emissions of the other countries in the domain were derived from the EMEP inventory, elaborated by CEIP (Centre on Emission Inventories and Projections) based on 2010 data provided by CLRTAP (Convention on Long-

range Transboundary Air Pollution) member states. In all simulations, the natural emissions of HMs were associated to the meteorological conditions since the wind is their main driver of this contribution as previously reported (Adani *et al.*, 2015).

The three simulations covered the whole Italian territory at $20 \times 20 \text{ km}^2$ horizontal spatial resolution and only the north of Italy at $4 \times 4 \text{ km}^2$. This part of the Italian domain is one of the most polluted areas in Europe and the Piedmont region, in north-west part of the domain, has few measured observations of HMs and PAHs in both years (2005, 2010) that can be used for the dynamical evaluation of model's skill. The model was configured with 16 unevenly spaced vertical levels (first level is at 20 m above the ground surface and the last at ca. 10,000 m), for both horizontal resolutions.

RESULTS AND DISCUSSION

This section shows the dynamical evaluation of the model for the years 2005 and 2010, the impact of changing meteorology and anthropogenic emissions, and the impact of grid resolution. Brief description of the differences in meteorological conditions can be found in the supplementary material.

Model Evaluation

Fig. 1 summarises some statistics of the percentage differences in HMs and PAHs air concentrations between 2010 and 2005 with respect to 2005 estimated from observations ("obs") and from simulations (E05B05M05, E10B10M10) with low ($20 \text{ km} \times 20 \text{ km}$) and high ($4 \text{ km} \times 4 \text{ km}$) grid resolutions. Black bars and dots indicate the median and the average values respectively; the grey box shows the 25th and 75th percentile and the black segments the 1st and 99th percentile. The evaluation regards As, Cd, Pb (16 stations), Ni (15 stations) and BaP (19 stations), the only PAHs monitored at the selected stations for both years. For these chemical species, the model predicts a lowering of concentrations in 2010 with respect to 2005 in agreement with the measurements. On average, the predicted decrease is lower than that estimated from observations for HM and higher for BaP, up to a factor of 4. For Cd, Ni and Pb, the increase of spatial resolution improves the modelled results; the median and average values of predicted percentage differences are closer to the observed ones while no improvements were observed for As and BaP at the change of scale resolution. The scarcity of data prevented a more comprehensive validation of the model's performance (Fig. S1). However, it is important to note that the concentrations simulated using national emission inventories are able to capture the trends of measured airborne pollutant concentrations for all simulated chemical species in spite of the simplifications applied to perform temporal and spatial distribution of emissions and of those currently used in formulation of chemical transport models. The significant reduction of emissions in 2010 with respect to 2005 for Ni and BaP leads to an evident reduction of their atmospheric concentrations, while the negligible reduction

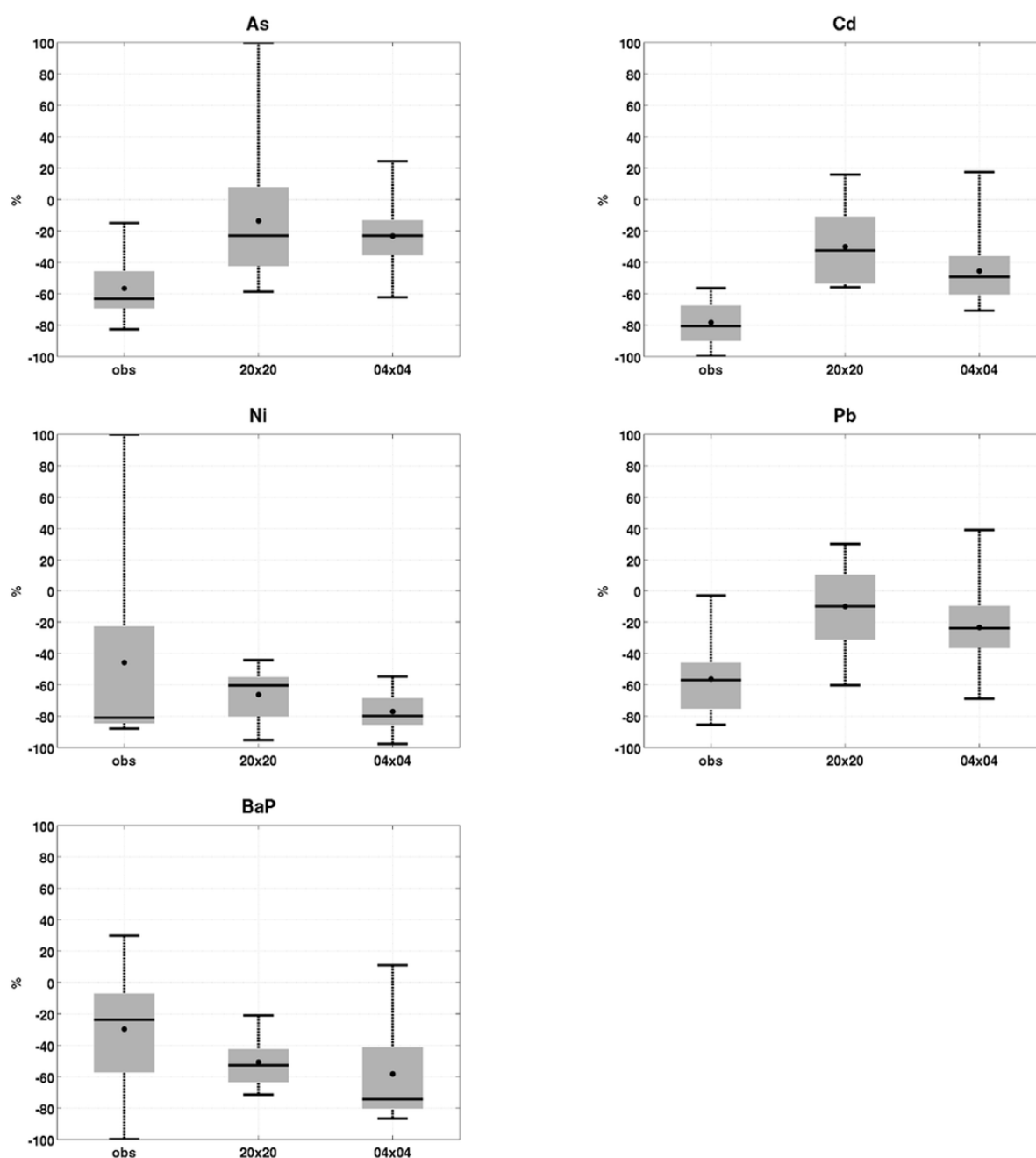


Fig. 1. Observed and simulated percentage differences concentrations between years 2010 and 2005 with respect to year 2005 of As, Cd, Ni, Pb and BaP. The simulated differences correspond to low (20×20 km) and high (4×4 km) grid resolutions. The boxes indicate the 25th, 50th and 75th percentiles of the data, while the ends of the whiskers represent the 1st and 99th ones. Black dots correspond to the averaged difference.

of Cd and Pb emissions produce only moderate changes in their concentrations. In the Piedmont region, the emissions are reduced by 60% for Ni, 28% for Cd, 24% for BaP, 2% for Pb leading to a reduction in concentration of ca. 80%, 50%, 70% and 20%, respectively (Fig. 1). On the contrary the As emission increases by 15% and the concentration decreases on average by 20%, this discrepancy may be due to meteorological conditions also considering the negligible transboundary transport of chemicals to Piedmont (Adani *et al.*, 2015).

Effect of Changing Anthropogenic Emissions and Meteorology

Figs. 2 and 3 show the difference in HMs and PAHs annual mean surface concentrations between the three simulations, E05B05M05, E05B10M10 and E10B10M10 (Section 2). In both figures, the first column illustrates the differences in concentrations between the two years: 2010 and 2005 (E10B10M10–E05B05M05)—base case (B20); the second column shows the differences due to changes in emissions (E10B10M10–E05B10M10) for 2010 meteorology

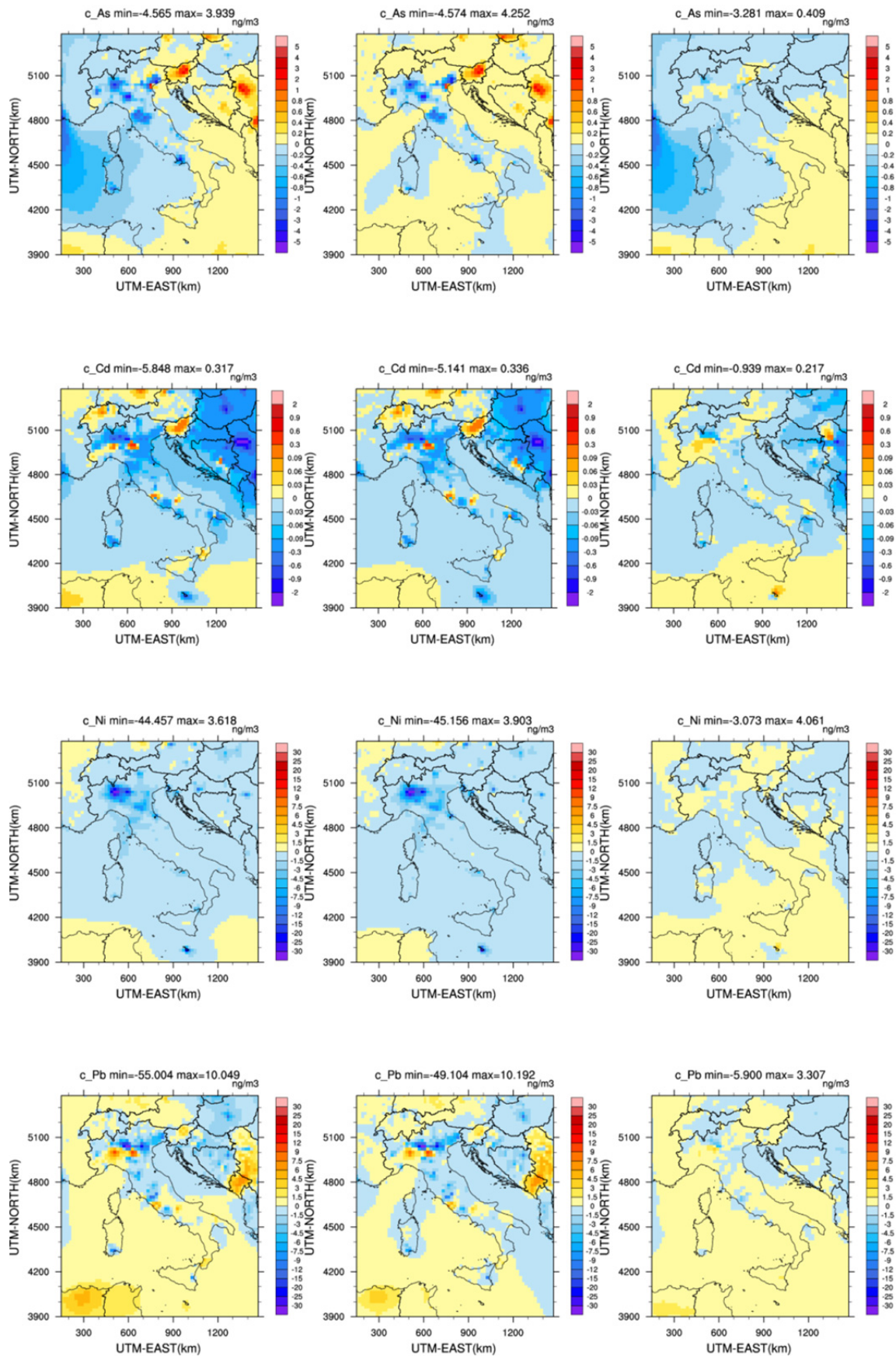


Fig. 2. Differences in annual mean concentrations of As, Cd, Ni and Pb (ng m⁻³), from top to bottom. Column 1, 2 and 3 are: E10B10M10–E05B05M05, E10B10M10–E05B10M10 and E05B10M10–E05B05M05, respectively.

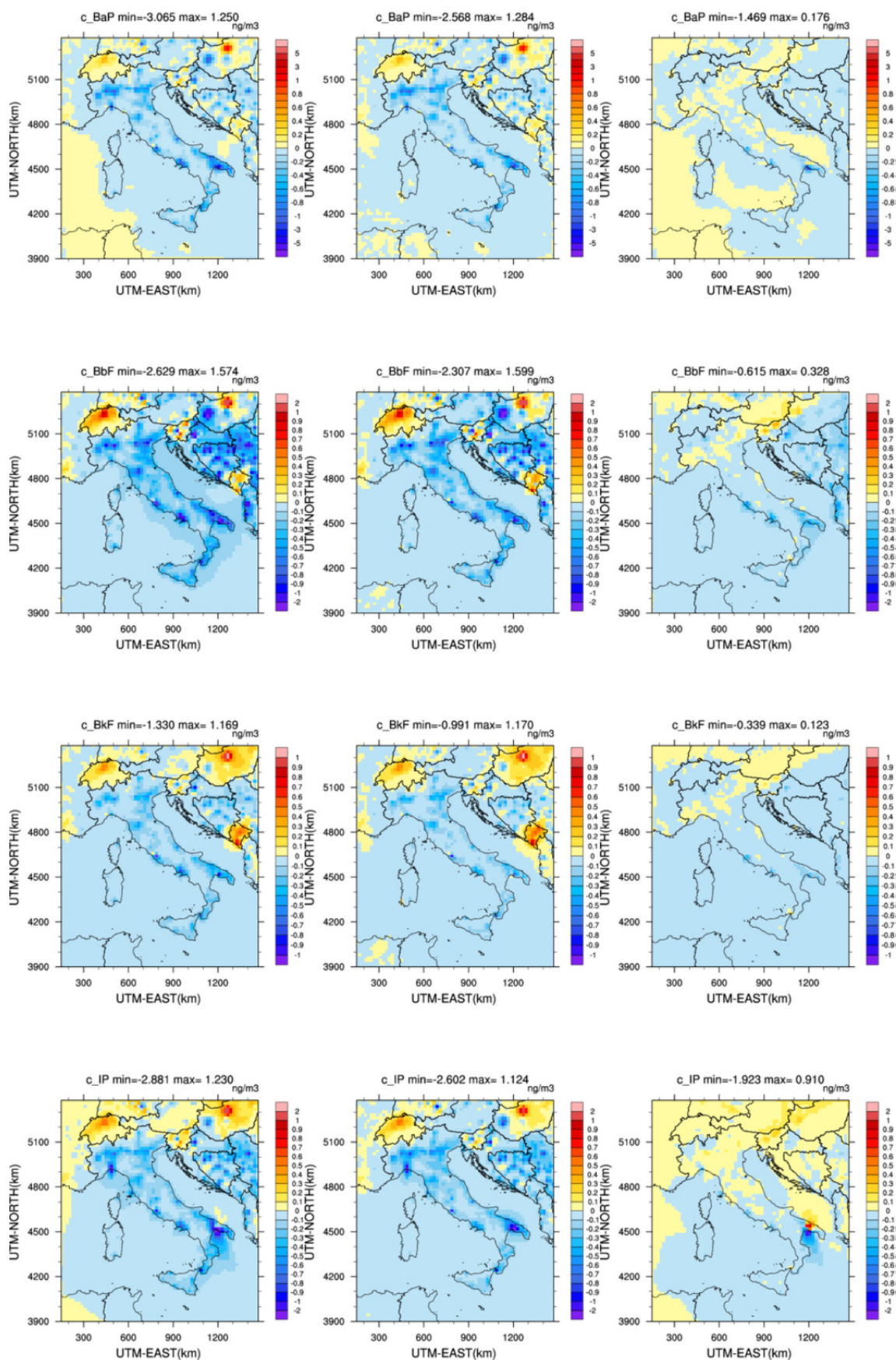


Fig. 3. Differences in annual mean concentrations of BaP, BbF, BkF and IP (ng m⁻³), from top to bottom. Column 1, 2 and 3 are: E10B10M10–E05B05M05, E10B10M10–E05B10M10 and E05B10M10–E05B05M05, respectively.

and boundary conditions—emissions case (E20) and the last column shows the differences due to changes in meteorology (E05B10M10–E05B05M05) for 2005 emissions—meteorology case (M20).

In general, over the Italian territory, the concentrations of HMs and PAHs were lower in 2010 with respect to 2005 (first column in Figs. 2 and 3). This decline is partly explained by the reduction of anthropogenic emissions (Table 1) and partly by the meteorological conditions less favourable to accumulation of air pollutants in 2010 with respect to 2005 (Figs. S4 and S5). Both low (Fig. S4) and high resolution (Fig. S5) simulations show that the accumulated precipitation was higher in 2010 with respect to 2005: the former over the whole Italy, apart Sicily and small area in the central-east part of peninsula (Abruzzo region) and the latter over the Po Valley (Northern Italy). This increase of precipitation enhances the wet removal of air pollutants leading thus to lower concentrations. In highly polluted areas such as Po Valley, central west coast of peninsula and the islands (Sicily and Sardinia), the wind speed is also higher in 2010 than in 2005 while the temperature is lower. Both changes produce further lowering of air pollutants concentrations: wind speed by causing more intense dispersive conditions and temperature by decreasing the reaction rates responsible for PAHs formation. Apart for As and to a lesser extent for Pb, the differences in concentrations in proximity of the domain borders do not reach the central part of the domain suggesting limited influence of boundary conditions on the modelled concentrations in agreement with results shown by Adani *et al.* (2015).

The percentage change in concentrations is highly variable for HMs: from a decrease of 67% for Pb up to 95% for Ni over an extended area in north of Italy. For PAHs, the highest reduction is observed for BaP and the lowest for indeno[1,2,3-cd]pyrene (IP) (see also Figs. S2 and S3).

As and Pb shows slight increases of concentrations in the south of Italy and Sicily, and over the sea, this is caused by a combination of “production” factors such as boundary conditions and natural emissions with “transport/dispersion/removal” factors driven by meteorological conditions. The increase of HMs concentration in some cities is higher than 100% due to increases in local emissions (Fig. S2). The increase for arsenic in Venice (red spot in Fig. 1) is almost entirely due to an increase of the glass manufacturing sector emissions recorded in 2010 with respect to 2005 (03-03-17 sector according to the Selected Nomenclature for sources of Air Pollution, SNAP). The increase of Cd and Pb concentration in

Pavia, Mantua, Isernia and Rome (main red spots in Fig. S2) is related increased emissions in commercial and institutional combustion plant with thermic power less than 50 Mw (SNAP sector 02-01-03). However, the increase of Pb concentrations in Rome (Central Italy, west coast) is not exclusively due to SNAP sector 02-01-03 that account for the 60% of the emission increase, but also due to air international cruises (SNAP sector 08-05-04) emissions that account for more than 20% and road transport emissions that account for ca. 10%.

Generally, over the Italian territory, the emissions play a more major role than meteorology for both HMs (Fig. 2) and PAHs (Fig. 3) since the magnitude of the differences in concentrations is usually greater in the second column with the respect to the third one. This behaviour is strongly related to local emissions: the areas affected by the changes of emissions are much lower than those influenced by the changes of meteorology. Therefore, the former are located around big cities and industrial areas, while the latter extend over wider areas including in the proximity of the domain borders, which are mainly influenced by transport of pollutant from outside of the domain.

The general reduction of emissions in 2010 with respect to 2005 over the Italian territory (Table 1) is also confirmed by the negative values of the differences in concentrations shown in the second column of Figs. 2 and 3 (E10B10M10–E05B10M10) for As, Ni and all PAHs, although there are reported increases in Cd and Pb concentrations in few cities such as Mantua, Pavia, Rome and Isernia. Whereas PAHs show a substantial emission decrease that range between 33% and 42%, HM emission decrease is dependent on the chemicals species. Cd and Ni total emissions decrease more than 10% and 60% respectively; Pb emission remains constant throughout the years while As has an increase of more than 20% in total emissions. Such increase is mainly attributable to an increase from diffused sources such as contact combustion processes for glass production. The provinces that are responsible for such increase are Padua, which accounts for one third of the total increment followed by Treviso, Venice, Vicenza Verona and Lucca. Nevertheless, the model estimates an average decrease of surface concentrations, except in Venice area, as mentioned above. Since the meteorological and boundary conditions used by the model were the same, this result clearly shows that the reduction of total emissions alone does not guarantee a positive effect on air pollution; the final effect on airborne concentrations of reduction emissions depends mainly on the characteristics of the source, such as its spatial distribution, and the temporal occurrence of the emission beside the total amount emitted.

Table 1. Total amount of emitted mass per year, from point and diffusive anthropogenic sources in year 2005 and 2010 over the Italian territory.

Chemical	As		Cd		Ni		Pb		BaP		BbF		BkF		IP	
	2005	2010	2005	2010	2005	2010	2005	2010	2005	2010	2005	2010	2005	2010	2005	2010
Total (t)	36	42	8	7	96	33	254	258	51	32	26	15	14	9	33	22
Diffuse (t)	30	38	6	5	78	15	201	211	20	14	25	12	14	8	17	9
Point (t)	6	4	2	2	18	18	53	47	31	18	1	3	0	1	16	13

For all chemicals, the changes of foreign emissions do not show a significant impact on the concentrations over the Italian territory. The lack of a gradient across the Italian boundaries in the differences of concentrations due to emission changes (graphs in the second column of Figs. 2 and 3, 2S and 3S) clearly evidences no supplementary transport of air pollutants due to foreign emission changes; the variations of concentrations due to changes of emissions are confined within the national boundaries as it can be observed for As and Cd for Slovenia, Pb, BbF, BkF for Albania and PAHs for Switzerland.

The decrease of PAHs concentrations over the Italian territory (Column 1 and 2 of Figs. 2 and 3) is more evident than that for HMs which are also influenced by natural emissions from soil and water surfaces under the action of wind.

Third column of Figs. 2, 3, S2 and S3 shows that the effect of meteorology is quite uniform over Italy, since the

percentage differences in concentrations vary between –10 and 10% for all chemical species. High increases in PM concentrations exist at the borders of the simulated domain, in particular in the south-west, south and south-east, mainly due to change in BCs. South of Italy and Sicily also show an increase of As and Pb concentrations in 2010, while in Sardinian island an increase of Pb is also observed.

Overall, without considering the cities, the increase of concentrations over the Italian territory is less than 10% in areas with low concentrations (Adani *et al.*, 2015).

The impact of anthropogenic emission reduction on the lowering of concentrations is clearly shown in Figs. 4 and 5 for HMs and PAHs, respectively. The differences in concentrations (DC/C) are plotted as a function of differences in emissions (DE/E) normalized by the concentrations and emissions used for 2005 (E05B05M04). Only grid points with total annual emissions higher than the median for the year 2005 and located over the Italian

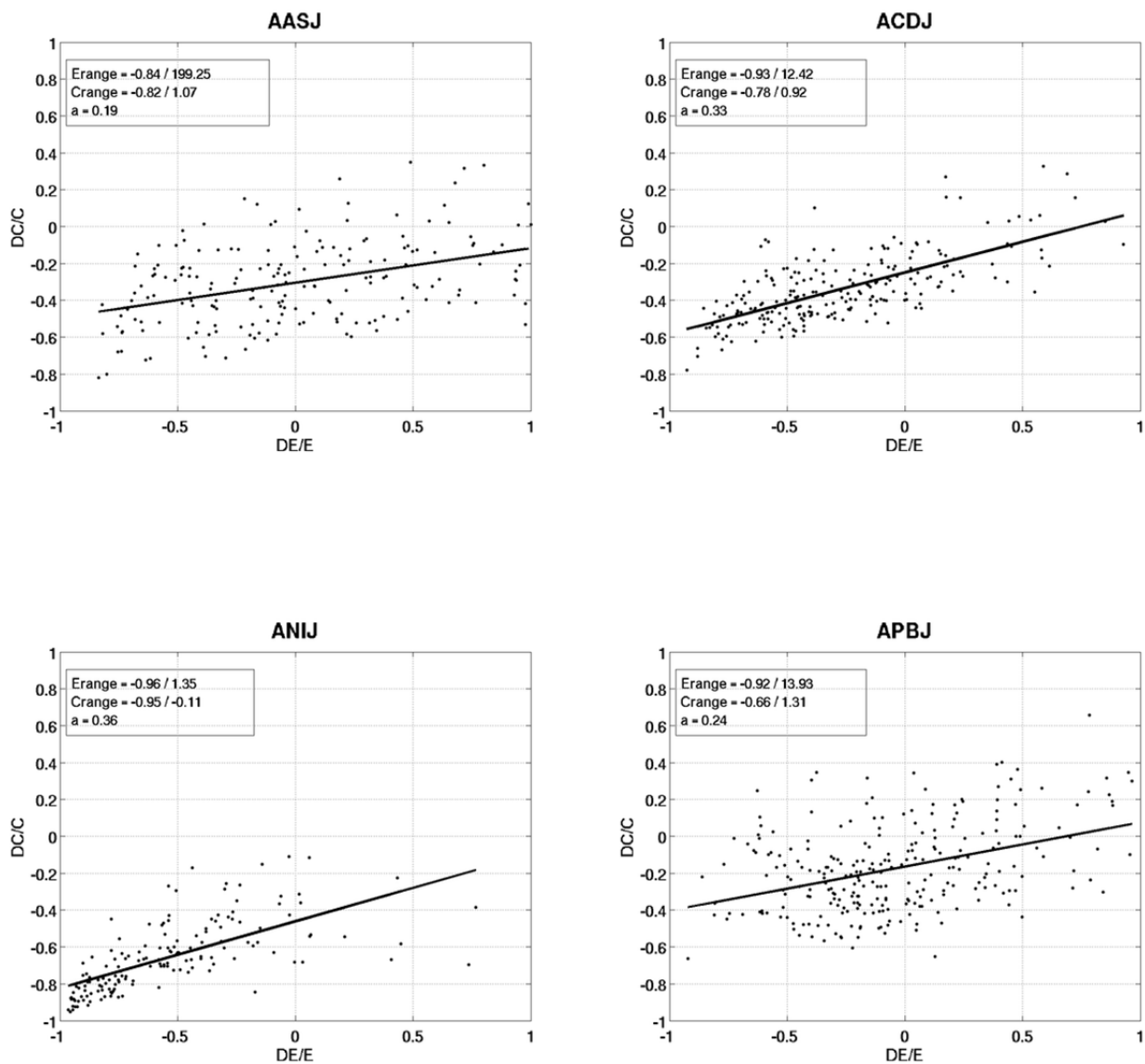


Fig. 4. Normalized variation of concentrations with respect to emissions for HMs. Top-right panel correspond to As; top-left, Cd; bottom-right, Ni, and bottom-left, Pb.

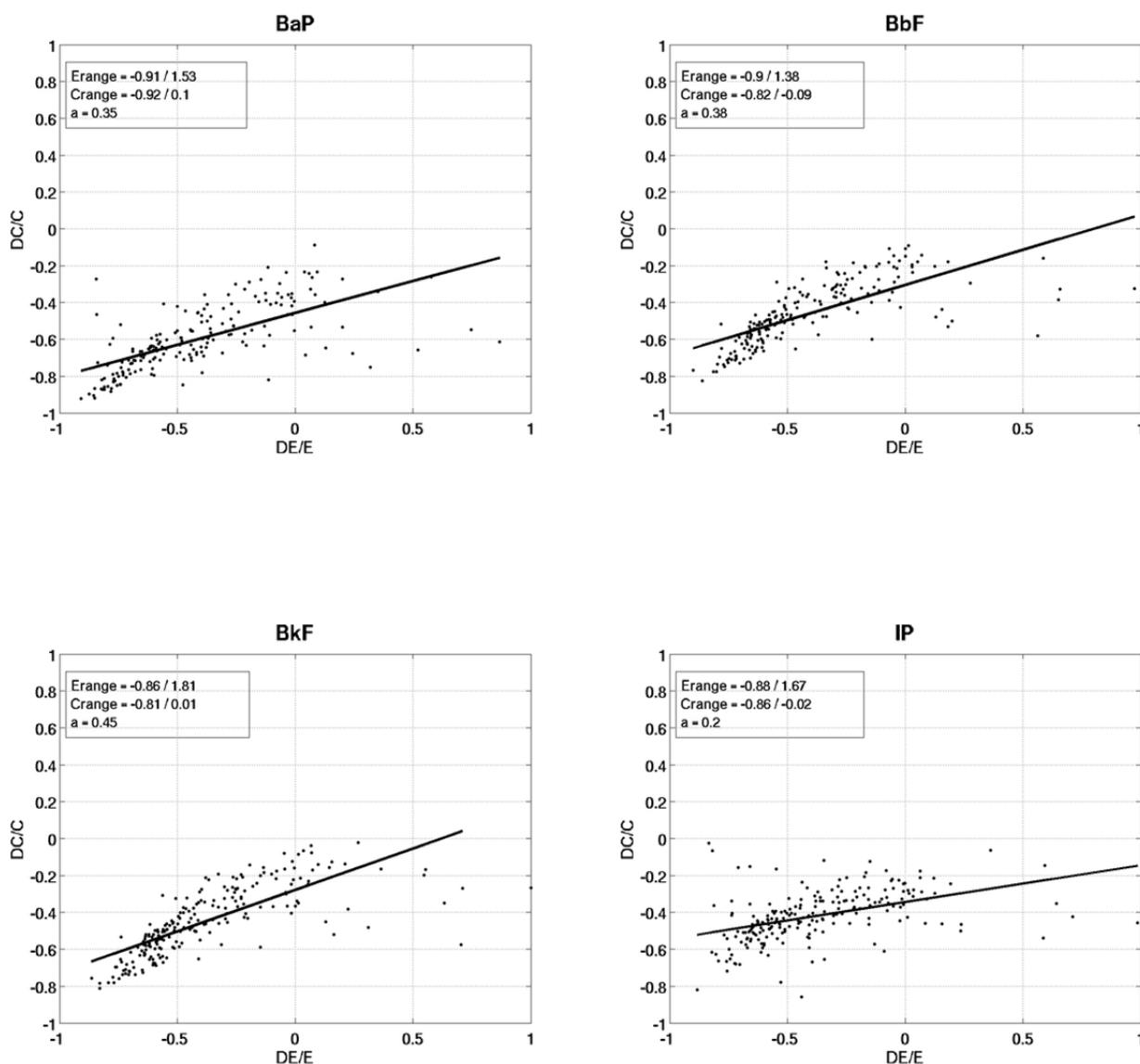


Fig. 5. Normalized variation of concentrations with respect to emissions for PAHs. Top-right panel correspond to BaP; top-left, BbF; bottom-right, BkF, and bottom-left, IP.

territory were considered. Apart for Pb and As, a decrease of local emissions lead to a decrease in concentrations (negative changes in concentrations correspond negative changes in emissions). For the other chemical species, Ni, BaP, benzo[b]fluoranthene (BbF) and benzo[k]fluoranthene (BkF), the linear relationship between concentration and emission changes has different slopes: from 0.45 for BkF to 0.2 for IP, which indicates different sensitivity of concentrations reduction to emissions changes.

As, Cd and Pb show a wide range of relative change concentrations in response to a given reduction of emissions: for example, a reduction of emissions of ca. 60% in Cd can lead to reduction of concentrations from 0 to 75%. This result is observed only for HMs: PAHs show less variability in concentrations for a given reduction of emission. Since all pollutants experience the same meteorological conditions, their different response to emission changes may be explained by the emission

sources characteristics and/or model formulation. Both for HMs and PAHs, grid points where concentrations decrease even if the local emissions increase, exist. This demonstrates that advection of pollutants emitted elsewhere in the model grid may determine local increases of concentrations.

Over the whole domain, the slope of the linear regression for each HMs and PAHs range from a minimum of 0.19 for As to a maximum of 0.37 for Ni, and from 0.19 for IP to 0.45 for BkF.

These relationships also include the differences between the meteorological conditions of 2010 and 2005, but their impact is minor with respect to the emissions as shown in Figs. 2 and 3.

Overall, it can be concluded that the air pollutants concentrations investigated are considerable diminished only in correspondence of significant reduction of local emissions. The decrease of concentrations in 2010 with respect to 2005 is mainly caused by the decrease of the

total emissions but the spatial and temporal distribution of the emission is important as well. For example, in case of As, the total emissions increase but the air concentrations decrease (Fig. 2). This reduction of concentrations may be explained only by the cleaning effect due to meteorological conditions since emissions are distributed in time and space in the same way for both years. For all chemicals, the temporal and spatial distributions of the emissions follow the same methodology used for PM_{2.5} and PM₁₀, at emission sector level.

Effect of Grid Resolution

Figs. 6 and 7 show the impact of higher horizontal spatial resolution simulations (4 km) on HMs and PAHs concentrations over the north of Italy for base case (B4–

B20)—first column, emissions case (E4–E20)—second column and meteorological case (M4–M20)—third column. The differences between the concentrations were computed on high-resolution grid after the re-gridding of the low-resolution simulations by linear interpolation. The concentration differences between 2010 and 2005 were computed as:

$$B4-B20 = (E10B10M10-E05B05M05)_4 - (E10B10M10-E05B05M05)_{20} \tag{1}$$

Fig. 6S shows in the first column the differences in concentrations for 2005 and in the second column for 2010 computed as $(E05B05M05)_4 - (E05B05M05)_{20}$ and $(E10B10M10)_4 - (E10B10M10)_{20}$. The positive areas

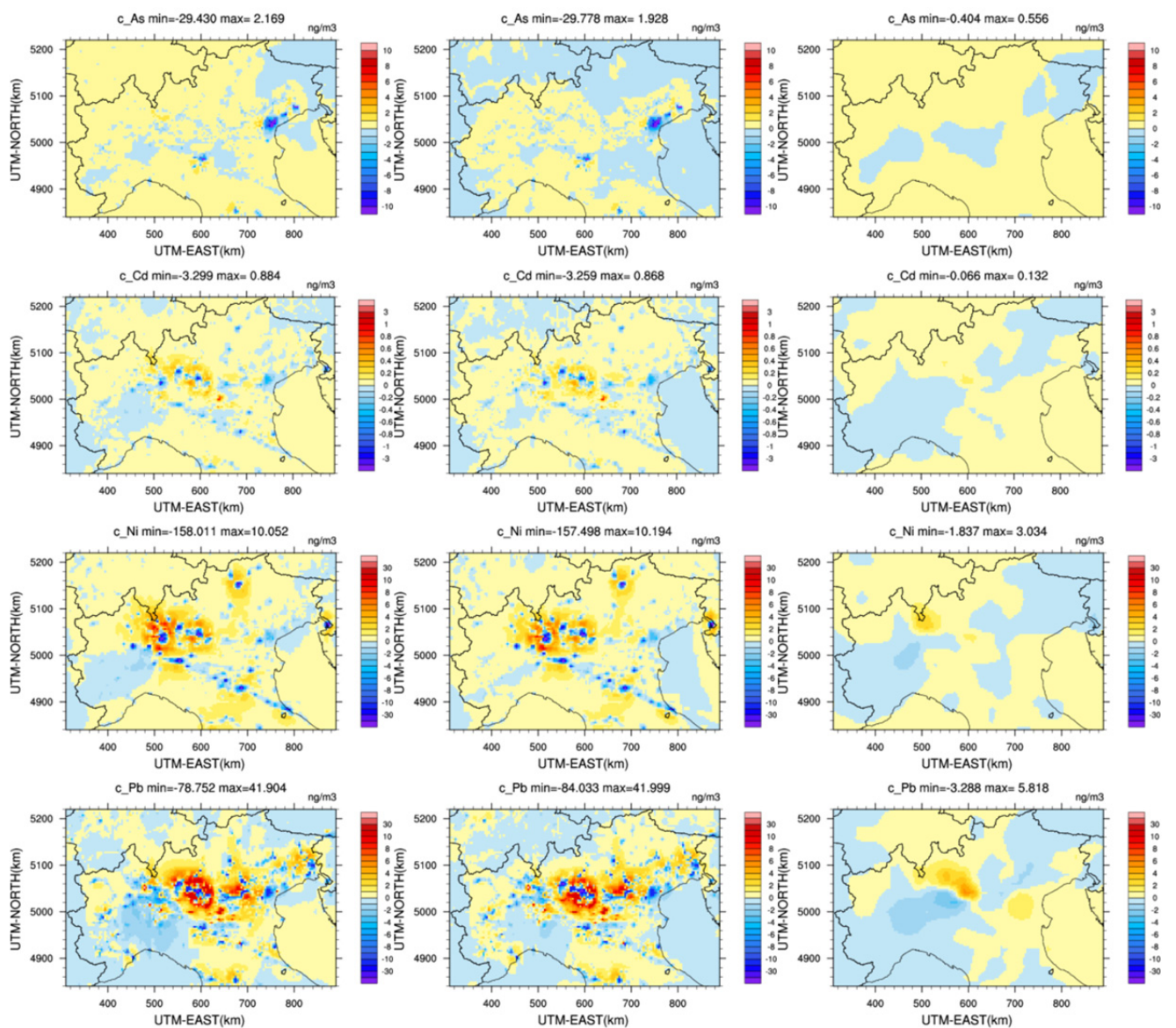


Fig. 6. Differences between high-low resolutions experiments computed as in Fig. 3. In particular: first column is E10B10M10–E05B05M05, second column is E10B10M10–E05B10M10; eventually the last column is E05B10M10–E05B05M05. From the above mentioned anomalies, the differences between high and low resolution has been computed for As, Cd, Ni and Pb from top to bottom.

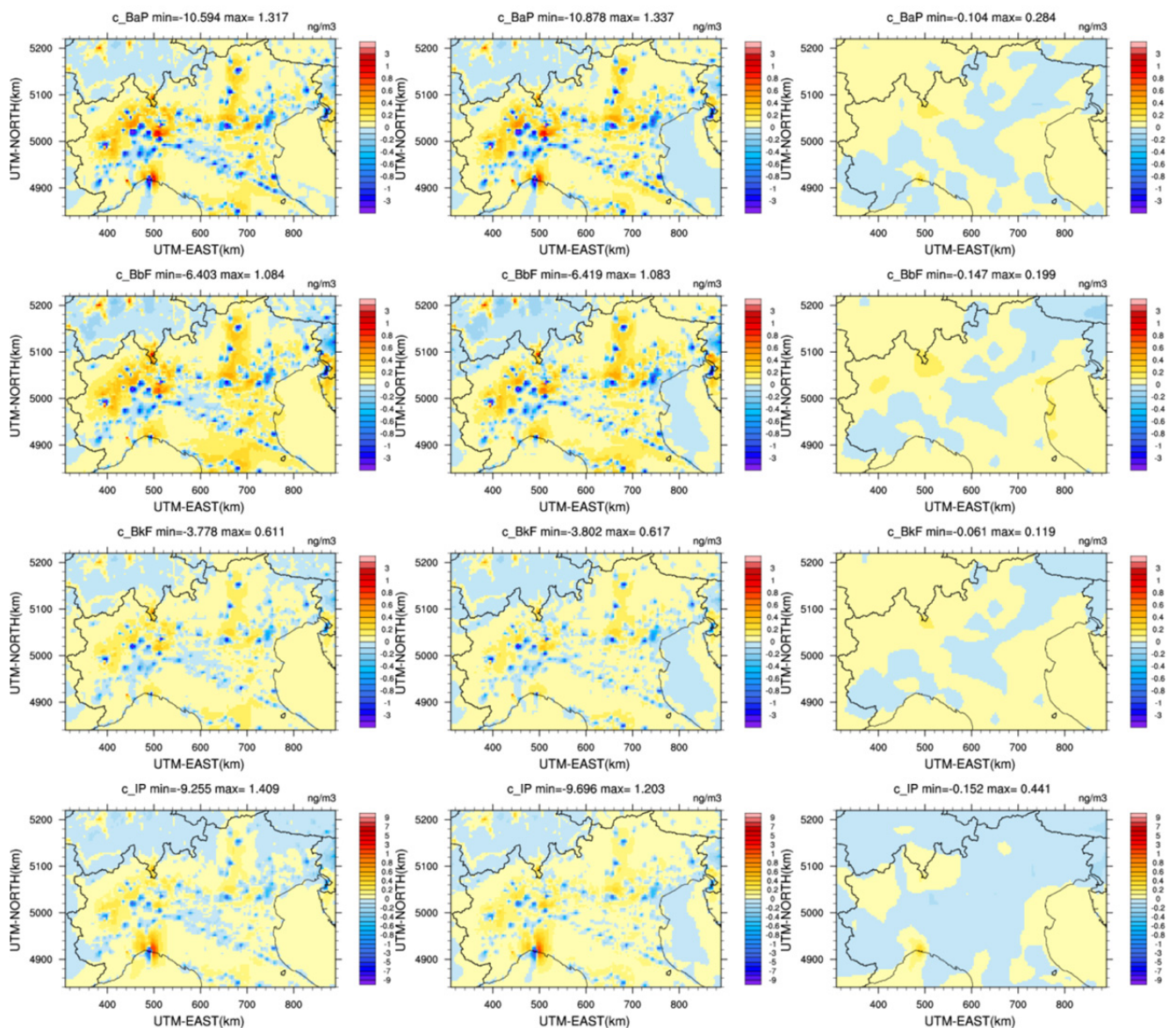


Fig. 7. Differences between high-low resolutions experiments computed as in Fig. 3. In particular: first column is E10B10M10–E05B05M05, second column is E10B10M10–E05B10M10; eventually the last column is E05B10M10–E05B05M05. From the above mentioned anomalies, the differences between high and low resolution has been computed for BaP, BbF, BkF and IP from top to bottom.

correspond to higher concentrations on high grid resolution, opposite is valid for the negative areas. Third column of Fig. 6S is the difference between first and second columns (B4–B20) that correspond also to Column 1 of Figs. 6 and 7. The differences between (E4–E20) and (M4–M20) are computed in similar way. Positive values of the first column of Figs. 6 and 7 show lower differences between high and low resolution for less polluted year (2010) with respect to more polluted year (2005).

In meteorological case (third column of Figs. 6 and 7), for all pollutants, the increase of grid resolution gives very limited impact except for Pb in some polluted and complex orography areas such as Brescia (on maps at coordinates: about 600 km, 5030 km). For all pollutants, the spatial distribution of the concentration differences is very similar

for base (first column of Figs. 6 and 7) and emission cases (second column of Figs. 6 and 7), the positive areas are dominant. This result showing extended areas with higher concentrations differences for low than for high grid resolution, is opposed to the well-known behaviour of concentrations in highly polluted areas in response to the increase of grid resolution. As shown in Adani *et al.* (2015), a high grid resolution leads to an increase of HM concentrations close to the anthropogenic sources with respect to low-resolution simulations, but it does not produce always an increase of the differences in concentrations (Figs. 6 and 7). The negative values located in urban areas and along the main roads, hotspots of high pollution, may be interpreted either as higher differences between resolutions for most polluted year (2005) compared to less polluted

one (2010) or as higher differences between the two years for the simulations at lower resolution than those at higher resolution. The response of these to the interplay between meteorology and emissions is not straightforward but their pattern, strongly related to the distribution of main emission sources and orography, and magnitude suggest the need to use high resolution simulations for assessing the changes in pollution conditions.

CONCLUSIONS

This study provides a dynamical assessment of the AMS-MINNI modelling system's ability to predict changes in the HMs and PAHs concentrations. The impact of emissions, meteorology and grid resolution on these concentrations for the years 2005 and 2010 was also analysed.

The modelling results are consistent with the available measurements, reproducing the observed decrease in concentrations from 2005 to 2010. For some chemicals, such as Cd, Ni and Pb, the use of a high grid resolution improves the model's agreement with the measurements.

Emissions have a greater impact than meteorology on the concentrations of HMs and PAHs: the differences in concentration are due to emission reduction rather than to changes in the meteorological conditions. In addition to reduction, the spatial and temporal distribution of emission fluxes plays a key role in predicting the concentration, as shown for As, which exhibited a decrease in concentration from 2005 to 2010 despite increasing in overall amount.

Furthermore, concentration patterns due to various emissions differ from those due to meteorology; whereas emissions cause a substantial impact on a local scale (cities, highways and highly industrialised areas), meteorology brings about changes on a synoptic scale. On a local scale, according to the chemical analysis, a decrease in emissions produces different effects on the concentrations, mainly owing to the varying characteristics of emission sources, as the same physical and chemical processes apply to all HMs and PAHs. For the same reason, the pattern and the magnitude of the changes in concentration when the emissions vary depend on the chemicals.

Increasing the grid resolution does not reveal a systematic behaviour that accounts for the differences in concentration (positive and negative values co-exist), but it allows us to better quantify the changes on a local level.

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SUPPLEMENTARY MATERIAL

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

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