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

Adani Mario<sup>1</sup>, Mircea Mihaela<sup>1</sup>, D'Isidoro Massimo<sup>1</sup>, Gualtieri Maurizio<sup>1</sup>

<sup>1</sup> ENEA-Italian National Agency for New Technologies, Energies and Sustainable Economic Development, via Martiri di Monte Sole 4,40129 Bologna, Italy

## Abstract

A dynamical evaluation of Flexible Air quality Regional Model (FARM) modelling system was conducted to evaluate the models skills to predict changes in heavy metals (HMs) and polycyclic aromatic hydrocarbons (PAHs) concentrations between the years 2005 and 2010 over Italy. Several simulations were performed to relate the variations of PAHs and HMs concentrations to the emission inventories, meteorology or grid resolution changes. The results generally show that the HMs and PAHs 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 for all chemicals while the meteorology determined important changes of reactive species concentrations such as benzo[a]pyrene (BaP) in high-resolution simulations, especially over complex topography. The analyses also show that the total emitted mass of pollutant is important but its geographical distribution is fundamental for estimating the variations in concentration.

*Keywords:* heavy metals; polycyclic aromatic hydrocarbons; air quality modelling; dynamical assessment; sensitivity experiments.

Corresponding author. Tel: +39-051-6098916; Fax: +39-051-6098675 E-mail address: mario.adani@enea.it Fig. S1 shows the locations of monitoring stations used for model validations. White dots correspond to monitoring stations that measure all the chemicals, the only gray dot corresponds to a station that measures all the compounds except Ni. While black dots indicate the stations that measure only BaP.

Figs. S2 and S3 show the percentage differences in annual mean concentrations of HMs (As, Cd, Ni and Pb) and PAHs (BaP, BbF,BkF and IP). Column 1, 2 and 3 are: (E10M10-E05M05)/ E05M05, (E10M10-E05M10)/ E05M10 and (E05M10-E05M05)/ E05M05, respectively.

Fig. S4 for Italian domain and Fig. S5 for North Italy domain show the differences between meteorological fields for 2010 and 2005. The left column shows the annual average of wind speed, of accumulated precipitation and of annual average of temperature for 2010 while the right column shows the differences with respect to 2005. It is evident that 2010 was more windy, rainy and cold. The first two factors do not favor the accumulation of pollutants in atmosphere while the third control the formation of some secondary species. The presences of intense winds favor the transport of pollutants far away from the source preventing the pollutant accumulation. Rain washes out the atmosphere increasing the wet deposition but decreasing the atmospheric concentrations while low temperature inhibits the gas phase chemical reactions and aerosol condensation reducing the formation of some pollutants. These results are consistent with the fact that 2010 meteorology decreases the annual mean concentration of both HMs and PAHs as discussed in the article. The difference between the high (4 km) and low (20 km) resolution experiments is important for both meteorological variables and concentrations of chemicals in both investigated years, especially in complex orographic areas.

Fig. S6 shows As and BaP difference between high and low resolution on top and bottom respectively. The differences for year 2005,  $(E05B05M05)_{4}$ - $(E05B05M05)_{20}$ , and for 2010,  $(E10B10M10)_{4}$ - $(E10B10M10)_{20}$ , are shown in the first and second column respectively. Third column shows ( $(E10B10M10)_{4}$ - $(E10B10M10)_{20}$ - $((E05B05M05)_{4}$ - $(E05B05M05)_{20}$ ) which can be written as ( $(E10B10M10)_{4}$ - $(E05B05M05)_{4}$ )- $((E10B10M10)_{20}$ - $(E05B05M05)_{20}$ )=B<sub>4</sub>-B<sub>20</sub> where B<sub>4</sub> indicate the difference in modelled concentration between 2010 and 2005 at high resolution while B<sub>20</sub> represent the difference in modelled concentration between 2010 and 2005 at low resolution.

## **Figure Captions**

**Fig. S1.** Locations of the monitoring stations used for model validations. White dots indicate the stations that measure all the chemicals (BaP, As, Cd, Ni, Pb). The only gray dot corresponds to a monitoring station that measures all the chemicals except Ni. Black dots correspond to stations that measure only BaP.

**Fig. S2.** Percentage differences in annual mean concentrations of As, Cd, Ni [%] and Pb [%], from top to bottom. Column 1, 2 and 3 are: (E10M10-E05M05)/ E05M05, (E10M10-E05M10)/ E05M10 and (E05M10-E05M05)/ E05M05, respectively.

**Fig. S3.** Percentage differences in annual mean concentrations of BaP, BbF,BkF and IP [%], from top to bottom. Column 1, 2 and 3 are: E10M10-E05M05, E10M10-E05M10 and E05M10-E05M05, respectively. Column 1, 2 and 3 are: (E10M10-E05M05)/ E05M05, (E10M10-E05M10)/ E05M10 and (E05M10-E05M05)/ E05M05, respectively.

**Fig. S4.** Left column shows the annual mean wind speed, accumulated precipitation and temperature for 2010 (from top to bottom). Right column shows, for the same quantities, the differences with respect to 2005.

**Fig. S5.** Left column shows the annual mean wind speed, accumulated precipitation and temperature for 2010. Right column shows, for the same quantities, the difference with respect to 2005. The differences were computed for the low-resolution grid.

**Fig. S6.** First and second columns show the annual mean difference between high and low resolution for 2005 and 2010 respectively. Third column is the difference between the second and first column. Upper and lower panels show the results for the As and BaP respectively.



**Fig. S1.** Locations of the monitoring stations used for model validations. White dots indicate the stations that measure all the chemicals (BaP, As, Cd, Ni, Pb). The only gray dot corresponds to a monitoring station that measures all the chemicals except Ni. Black dots correspond to stations that measure only BaP.



**Fig. S2.** Percentage differences in annual mean concentrations of As, Cd, Ni and Pb, from top to bottom. Column 1, 2 and 3 are: (E10M10-E05M05)/ E05M05, (E10M10-E05M10)/ E05M10 and (E05M10-E05M05)/ E05M05, respectively.



**Fig. S3.** Percentage differences in annual mean concentrations of BaP, BbF,BkF and IP, from top to bottom. Column 1, 2 and 3 are: (E10M10-E05M05)/ E05M05, (E10M10-E05M10)/ E05M10 and (E05M10-E05M05)/ E05M05, respectively.



**Fig. S4.** Left column shows the annual mean wind speed, accumulated precipitation and temperature for 2010 (from top to bottom). Right column shows, for the same parameters, the differences between 2010 and 2005.



**Fig. S5.** Left column shows the annual mean wind speed, accumulated precipitation and temperature for 2010. Right column shows, for the same parameters, the difference between 2010 and 2005. The differences were computed for the high-resolution grid.



**Fig. S6.** First and second columns show the annual mean difference between high and low resolution for 2005 and 2010 respectively. Third column is the difference between the second and first column. Upper and lower panels show the results for the As and BaP respectively.