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# MULTI-MODEL ASSESSMENT OF PM TRENDS IN EUROPE DURING TWO DECADES (1990-2010)

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**Abstract**: The model trend analysis for PM<sub>10</sub> and PM<sub>2.5</sub>, performed within the Eurodelta-Trends experiment, covers 21 year, from 1990 through 2010, with particular focus on the period 2001-2010 for which appropriate amount of PM observations is available. Eight chemical transport models (CTM) participated in the multi-modal trend analysis: EMEP/MSC-W, CHIMERE, LOTOS-EUROS, MINNI, MATCH, WRF-Chem, CMAQ and Polyphemus (out of which six models performed trend runs for the 21-year period). The average modelled trends are somewhat smaller than the observed, though the models identify significant PM trends at more sites in the period 2001-2010. There are considerable difference in the PM trends between the regions/countries and in different seasons. Investigation of the changes in PM chemical composition during the investigated period shows that the models differ in terms of relative contribution of the individual PM components to the PM trends. For the 2001-2010 period, the effect of inter-annual meteorological variability appears more important relative to emission changes. Finally, we look at PM trends/changes during the 1990-2010 period.

Key words: PM10 and PM2.5 trends, multi-modal assessment, observed trends.

#### INTRODUCTION

The Eurodelta-Trends multi-model chemistry-transport experiment was designed in order to better understand the evolution of air pollution and its drivers since the early 1990s. The main objective of the experiment is to assess the efficiency of air pollutant emissions mitigation measures in improving regional scale air quality in Europe. The presented multi-model trend analysis is a contribution to the assessment of the evolution of air pollution in the EMEP region over the 1990-2012 period coordinated by the Task Force on Monitoring and Modelling of EMEP (Cooperative Programme for Monitoring and

Evaluation of the Long-range Transmission of Air Pollutants in Europe) and presented Colette et al., 2016).

#### **EXPERIMENT DESIGN**

The set-up and input information for model runs were largely harmonized, thus allowing investigation of the uncertainties in calculation results due to the differences in model formulations. Six of the chemical transport models (CTM) participated in the multi-modal trend analysis, i.e. EMEP MSC-W, CHIMERE, LOTOS-EUROS, MINNI, MATCH, WRF-Chem, performed a 21-year run series over Europe for the 1990-2010 period. In addition, those six models and also CMAQ and Polyphemus made calculations for the years 1990, 2000 and 2010. In order to study the effect of meteorological variability, additional runs with constant 2010 emission were performed for the same years. For detailed description of the experiment see Colette et al. (2017).

The model trend analysis for  $PM_{10}$  and  $PM_{2.5}$  covers 21 years, from 1990 through 2010. For the 2001-2010 period, for which enough PM observations are available from the EMEP monitoring network, the modelled trends of  $PM_{10}$  and  $PM_{2.5}$  have been compared with measurement data. For both model results and observations, the Mann Kendall (MK) method has been used to identify significant trends, and Sen's slopes have been calculated to quantify PM trends.

#### RESULTS

#### Model performance for PM

Comparison with EMEP observations for the years 2001 to 2010 shows that the models underestimate  $PM_{10}$  by 14% and  $PM_{2.5}$  by 11% on average. The inter-annual bias variation is mostly within 5% (up to 10%), and a certain consistency can be noticed between the models' performance (e.g. smaller underestimation of  $PM_{10}$  for 2001, 2005, 2006 and 2009 and greater underestimation for 2002, 2007, 2008, 2010). The average spatial correlation coefficients are 0.58  $PM_{10}$  and 0.67 for  $PM_{2.5}$  and in general vary only moderately between the years and the models.

#### **Trends for 2001-2010**

Figure 1 presents for the period 2001-2010 the observed and modelled changes in mean  $PM_{10}$  and  $PM_{2.5}$  levels and the trend slopes at the selected set of trend-sites (Colette et al., 2016), showing the mean from 5-model ensemble and the individual models' results.



Figure 1. PM10 and PM2.5 mean level change and Sen's slopes observed and according to 6-model ensemble for the period 2001-2010; bottom figures also show results from the individual models.

The PM levels from the 5-model ensemble are lower than observed, with negative biases being larger for  $PM_{10}$  than for  $PM_{2.5}$ , but the discrepancies somewhat decrease towards the end of the period. The model calculated year-to-year PM changes follow quite well the observations, except from the end of the period. There, we see a decrease in calculated PM levels, while the measured PM increases. The resulting average modelled trends are somewhat smaller than the observed ones. For  $PM_{10}$ : calculated mean -0.31  $\mu$ g m<sup>-3</sup> yr<sup>-1</sup> (or -2.0 % per yr), observed -0.42  $\mu$ g m<sup>-3</sup> yr<sup>-1</sup> (or -2.3 % per yr); and for  $PM_{2.5}$ : calculated mean -0.28  $\mu$ g m<sup>-3</sup> yr<sup>-1</sup> (or -2.5 % per yr), observed -0.41  $\mu$ g m<sup>-3</sup> yr<sup>-1</sup> (or -3.2 % per yr). For the individual models, the correlations between annual mean modelled and measured  $PM_{10}$  are: 0.85 for CHIM, 0.88 for EMEP, 0.62 for LOTO, 0.80 for MINNI and 0.84 for MATCH. For  $PM_{2.5}$ , the correlations between the model results and observations are even higher, with the corresponding values being 0.95, 0.94, 0.83, 0.90 and 0.95.

The calculations and observations show considerable difference in regional PM trends. Figure 2 shows the mean values of Sen's slop for the period of 2001-2010 calculated by the ensemble of six models and measured at EMEP sites. The model mean and observed annual levels of  $PM_{10}$  and  $PM_{2.5}$  were in general decreasing overall in Europe during the 2001-2010. Compared to observations, the models identified significant trends in  $PM_{10}$  and  $PM_{2.5}$  for larger geographical regions and more locations (though underestimating the trend slopes).



**Figure 2.** Mean Sen's slopes for PM10 and PM2.5 trends in 2001-2010 (μg m-3 year<sup>-1</sup>) calculated by 6-model ensemble (described in Colette et al. (2016), Appendix A3.) Shown are modelled trends – coloured contours (grey means insignificant trends) and observed trends - coloured triangles (significant) and circles (insignificant).

The study reveals quite a large variability of observed mean trends at the individual sites, ranging from - 0.93 to 0.003  $\mu$ g m-3 yr-1 (from -0.83 to -0.308  $\mu$ g m-3 yr-1 for significant trends). Due to large meteorological variability and the relatively short studied period, the observations do not indicate significant PM10 trends at 14 out of 29 sites; and only at 8 out of 18 PM2.5 trend-sites, significant trends were observed. The model calculated mean trends at different sites vary less compared to the observations between the sites. In general, the models tend to under-predict significant observed trends, whereas the modelled trend are closer to, or overestimate observations at the sites with insignificant measred trends.



Figure 3. Seasonal mean concentrations of PM<sub>10</sub> averaged over 29 (blue) and PM<sub>2.5</sub> averaged over 18 sites (red). Solid lines - observations, dotted lines - 5-model ensemble; grey lines - Sen slopes.

Analysis of seasonal profiles shows considerable variability, with the largest trends obesrved and calculated for summer and the smallest for winter (Figure 3).

The models agree that the chemical composition of European  $PM_{10}$  was changing in the course of 2001-2010 decade, with  $NO_3$  becoming increasingly more important constituent compared to  $SO_4$ ,  $NH_4$  and carbonaceous particles from primary PM. However, the models differ in terms of relative contribution of the individual PM components to the PM trends.

#### Trends for 1990-2010

PM trends for the period of 1990-2010 could only be studied based on model calculations, which show downward trends over the whole of Europe. The mean estimated trends are  $-0.37 \ \mu g \ m^{-3} \ yr^{-1}$  for PM<sub>10</sub> and  $-0.32 \ \mu g \ m^{-3} \ yr^{-1}$  for PM<sub>2.5</sub>, though the slope values vary considerably between the countries. All models calculate a larger decrease of PM levels from 1990 to 2000 compared to the 2000-2010 period, except from Northern Europe (as example from the EMEP model in Figure 4).



**Figure 4.** Mean PM<sub>10</sub> trends (µg m<sup>-3</sup> yr<sup>-1</sup>) for the individual countries for 1990-2000 and 2000-2010 periods (calculated with the EMEP model).

Finally, our study shows considerable effect of the inter-annual changes in meteorological conditions on the modelled 1990-2010 trends, and for the shorter period of 2001-2010 with smaller emission changes the meteorology appears more important than the emission changes.

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