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EVALUATION OF SEVEN CHEMISTRY TRANSPORT MODELS IN THE FRAMEWORK OF EURODELTA III INTERCOMPARISON EXERCISE

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Abstract: The EURODELTA III (ED-III) exercise aimed to perform a comprehensive chemistry transport model inter-comparison study exploiting the data from four intensive measurement campaigns carried out by EMEP. The campaigns were held in different seasons (1–30 June 2006; 8 January–4 February 2007; 17 September–15 October 2008; 25 February–26 March 2009) thus allowing to test the influence of different meteorological conditions on models' results. Seven models simulated the air quality over the whole Europe: CHIM (CHIMERE; version chim2013), EMEP (rv 4.1.3), LOTO (LOTOSEUROS, V1.8), CAMX (CAMx, v5.41 VBS), MINNI (version 4.7), CMAQ (V5.0.1) and RCG (v.2.1). Except CMAQ, all the models performed simulations over the same domain with the same horizontal spatial resolution. They also used the same input data (emissions, meteorology and boundary

conditions) as much as possible. This work presents and discusses the behaviour of the models with regard to the criteria defined in the EU Directive on Air Quality 2008/50/EC for the air concentrations of PM10, PM2.5, O_3, NO_2 and SO_2 and to the meteorological conditions. The wet deposition of sulphate (S) (WSOx), of oxidized and reduced nitrogen (N) (WNOx and WNHx, respectively) and the air concentrations of the deposited species were also investigated. Furthermore, a comparison of the capacities of air quality models to simulate carbonaceous aerosols (elemental (EC) and organic carbon (OC)) in Europe was conducted, given the diversity in modelling natural precursor emissions and formation and evolution of organic species, both natural and anthropogenic.
In addition to EMEP data, the evaluation of models' output included AirBase data and meteorological data from more than 2000 synoptic stations. The simulated concentrations of organic aerosol (OA) were compared to measurements available from two intensive measurement field campaigns carried out in a joint framework of EMEP and EUCAARI (the European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions) project in 2008 and 2009.

Key words: *model intercomparison, air quality, atmospheric pollutant concentration, N and S atmospheric deposition*

INTRODUCTION

The inter-comparison and evaluation of regional chemical transport models (CTMs) for different air pollutants is more necessary than ever due to increasing use of more and more complex air quality models and due to Directive 2008/50/EC on Ambient Air Quality and Cleaner Air for Europe which encourages modelling as a valid tool for activities such as air quality assessment, forecasting and planning.
The ED-III exercise established an unique framework in terms of input data, models' configuration and data for models' evaluation over Europe. This allowed to improve the understanding of CTMs performances and behaviour with respect to criteria pollutants defined in Directive 2008/50/EC.

MODELS AND INPUT DATA

All models participating in this intercomparison exercise (CHIM, EMEP, LOTO, CAMX, MINNI, CMAQ and RCG) are offline CTMs which implies that they are driven by external meteorological data. Apart CMAQ runs which were based on meteorological variables from the COSMO model (CLimate Mode, COSMO-CLM, version 4.8 clm 11), all runs were based on meteorological fields provided by ECMWF IFS (Integrated Forecast System) at a spatial resolution of 0.2°. The hourly meteorological data were used as input to meteorological pre-processors of CTMs which prepared the actual data for the chemistry models in two ways: 1) calculating the dispersion parameters required by CTMs and/or 2) mapping meteorological variables on 3D the domain used by CTMs. Due to the fact that the pre-processor modules of CTMs used different surface and boundary-layer parameterisations and that the vertical resolution of CTMs was different, the meteorological data actually used for runs were different. In particular, for CMAQ which performed the simulations on a Lambert-conformal conic projection with the standard parallels at 30 and 60 and a grid of 112x106 cells of size 24 km x 24 km while the rest of the models used the same domain at 0.25° x 0.25° spatial resolution.
The 2D anthropogenic emissions at 0.25° x 0.25° spatial resolution were provided by INERIS. The data were obtained by merging several databases: 1) TNO 0.125° x 0.0625° for 2007 from MACC (Kuenen et al., 2011), 2) EMEP 0.5° x 0.5° emission inventory for 2009 (Vestreng et al., 2007) and 3) emission data from the GAINS database (http://gains.iiasa.ac.at/gains). Only for the 2006 campaign, fire emissions were provided based on the GFASv1.0 database (Kaiser et al., 2012). Each CTM used its own processor to distribute the emissions of NH_3, NOx, PM2.5, PMcoarse and NMVOC to aerosol and gas species according to its aerosol and gas phase chemistry models. Seasonal, weekly and hourly time profiles and vertical injection profile of emissions were also provided. The natural emissions such as biogenic VOC emissions from vegetation, soil nitrogen monoxide emissions, sea salt and dust emissions were calculated according with their own schemes by CTMs. CAMx, CHIMERE and CMAQ have no dust production in this exercise.
MACC reanalyses were used as input data for the boundary conditions (Inness et al., 2013).
More details on the input data and models' description can be found in Bessagnet et al. (2016) and Vivanco et al. (2017).

RESULTS

Figures 1 and 2 show the normalised mean bias (NMB) defined as in Bessagnet et al. (2016) for all models and all campaigns:

$$NMB = (\overline{M} - \overline{O}/\overline{O}) \quad (1)$$

where $\overline{M}$ is model average and $\overline{O}$ is observed average.

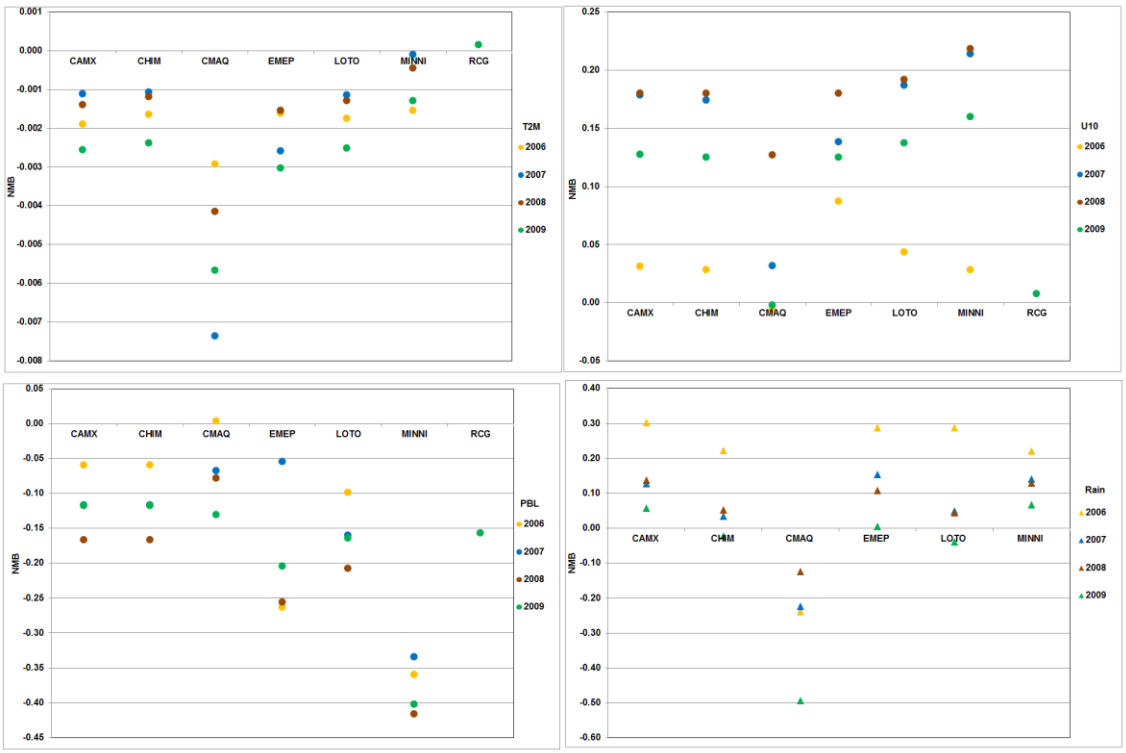

Figure 1. NMB of models for temperature at 2m (T2M) and wind speed at 10m (U10) height above ground, planetary boundary layer (PBL) height and rain for all campaigns.

The NMBs of meteorological variables are shown in Figure 1 while those of the chemical variables are shown in Figure 2. For all models and all campaigns, the differences in temperature at 2m (T2M) height above ground considered by CTMs are the lowest (below 1%) with respect to those of wind velocity at 10m (U10), planetary boundary layer (PBL) height and rain. For all campaigns, in case of rain and temperature, the NMBs of CMAQ model show different behaviour with respect to the other models: underestimates the rain while the others models overestimate and shows much variability in temperature with season (0.5% variability from summer-2006 to autumn-2008 than a maximum of ca.0.2% for the others models).

All models overestimate the wind velocity (U10) and underestimate the PBL heights. For these variables, the best agreement with measurements is achieved in summer (2006) and the worst in autumn (2008). MINNI model shows the highest underestimation of PBL height for all campaigns.

The NMBs of rain shown in Figure 1 consider only the sites with WNOx and WNHx measurements which corresponding NMBs are shown in Figure 2. CMAQ coherently shows underestimation of NOx wet deposition associated to underestimation of rain (negative values of NMB of WNOx & WNHx and rain). The other models show mostly underestimations of WNOx & WNHx in spite of rain overestimation which may be explained by two factors: lower NOx & NHx atmospheric concentrations and/or the wet deposition schemes used. According to Vivanco et al. (2017) who investigated in detail the models' behaviour, the underestimation of the gas scavenging efficiency may play the key role in obtaining low wet deposition loads.

Elemental carbon (EC) contained in PM10 is a primary aerosol, directly emitted in atmosphere by anthropogenic sources, it is subject to physical processes mainly driven by meteorological conditions but less affected by atmospheric chemistry. Therefore, Figure 2 shows different behaviour of NMBs during warm (summer -2006 and early autumn - 2008) and cold seasons (winter - 2007 and early spring -2009). During summer, EC concentrations are underestimated by all models, with an average up to ca. 20% and maximum of 40%, while during winter all the models overestimate the measured concentrations. These similarities in models' response can be explained only by the EC emissions which may be overestimated during cold seasons and the opposite during warm seasons. The differences in meteorological variables and models' parametrizations may explain the diversity between the models' results and their dependence on campaign/season.

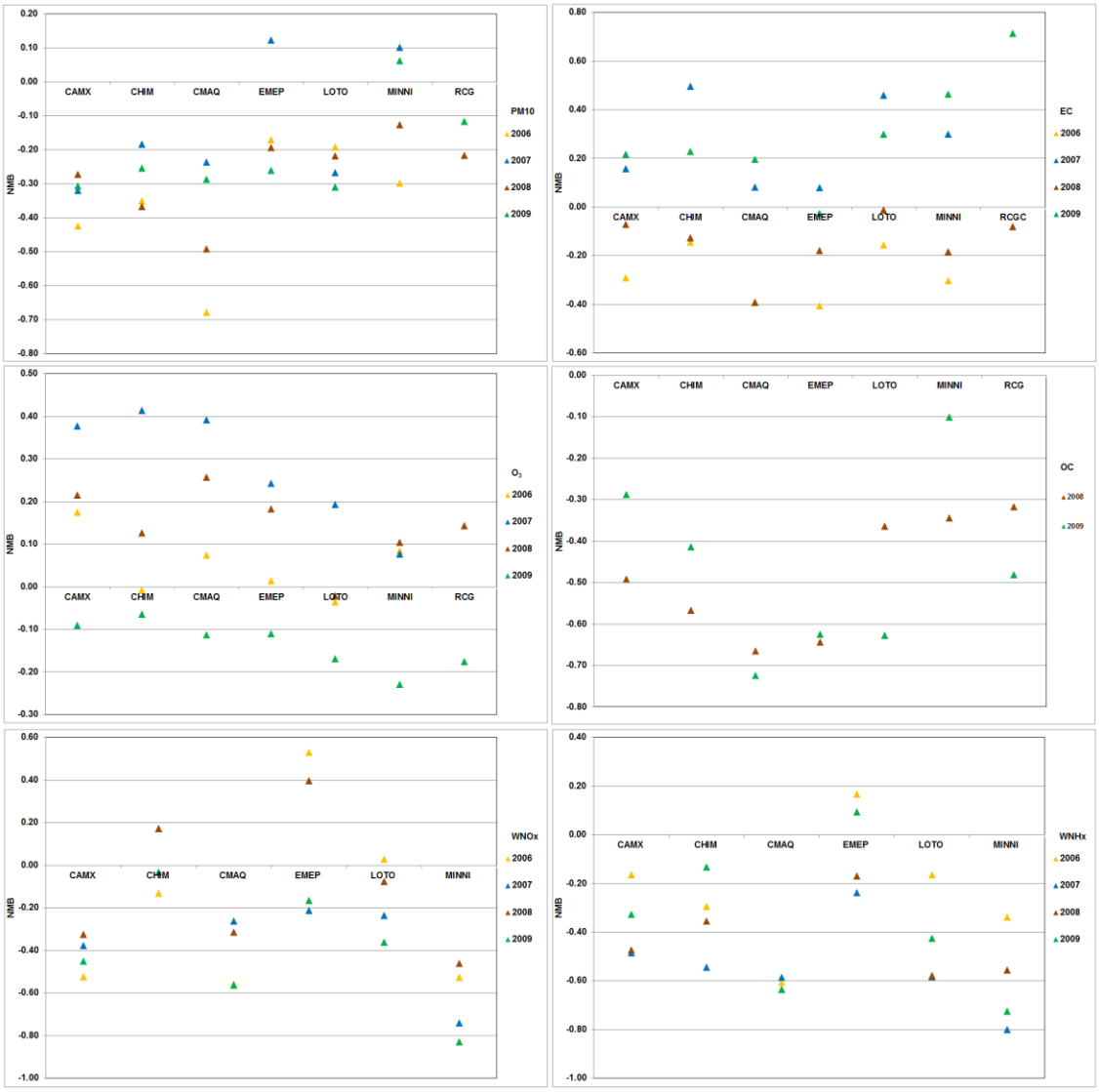

Figure 2. NMB of models for O_3, PM10, EC and OC atmospheric concentrations and WNOx, WNHx deposition for all campaigns.

The organic carbon (OC) contained in PM10 does not show such a well-defined behaviour for early autumn (2008) and spring (2009) campaigns. This may be due to the fact that an important fraction of OC aerosol is secondary, produced in atmosphere from non-methane volatile organic carbon (NMVOC) anthropogenic emissions (the same for all models) and from biogenic volatile organic carbon (BVOC) emissions from vegetation which varied among the CTMs. In addition to this, the different gas-phase chemistry mechanisms, aerosol models and deposition schemes used off-set the seasonal signature of

meteorological variables and of primary OC aerosol emissions. All the models underestimate the OC concentrations, CMAQ has the highest values while MINNI the lowest probably due to the high underestimation of PBL heights. Data for evaluating the models' performance for OC were available only from the field measurements carried out during EUCAARI (the European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions) project in 2008 and 2009.
Apart from MINNI in 2007 & 2009 and EMEP in 2007, all models underestimate PM10 concentrations to different degrees. The highest underestimation, up to 70%, is observed for CMAQ during warm seasons (2006 and 2008) while the rest of models shows underestimations below 30%. CAMX, CHIM and LOTO show seasonal variability below 20%, EMEP and MINNI is ca. 40 % and CMAQ around 50%. For CHIM, CMAQ and MINNI, PM10 is better predicted during cold season (2007 & 2009) than during the warm season (2006 & 2008). Since both primary and secondary aerosol compounds, natural and anthropogenic, contribute to PM10 concentrations by complex nonlinear processes, the main responsible in terms of input data or model formulation has still to be identified.
In case of O_3, the models underestimate the air concentrations in early spring (2009) but overestimate the measurements during the other campaigns. The highest overestimations are observed during winter 2007 for CAMX, CHIM and CMAQ. As for OC and PM10, the O_3 variation cannot be attributed to a single factor since its production and destruction depend on natural and anthropogenic emissions, meteorological conditions, gas-phase chemistry mechanisms and deposition schemes in a complex way. The lack of wood burning emissions could explain the high underestimation of organic matter in the models.

CONCLUSIONS

The results of ED-III show that the CTM models have similar performances; generally, they underestimate OC, PM10 and WNOx, and show EC and O_3 dependence on season. However, the differences between the models' results and with respect to measurements is generally higher than 10% and cannot be explained only by the differences meteorological variables estimated with surface and boundary-layer parameterisations.

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