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EVALUATION OF THE CHIMERE MODEL AT HIGH RESOLUTION OVER EUROPE: FOCUS ON URBAN AREA

E.Terreiroire (1), B.Bessagnet (1), G.Pirovano (2), P.Thunis (3), A.Colette (1), A.Ung (1), L.Letinois (1), L. Rouil (1)

(1) INERIS –Parc Technologique Alata – BP 2 – 60550 Verneuil-en-Halatte – France

(2) RSE – Via Rubattino 54 – 20134 Milano – Italia

(3) JRC – Via Enrico Fermi 2749 – 21027 Ispra – Italia

Presenting author email: etienne.terreiroire@ineris.fr

ABSTRACT

A comprehensive evaluation of a CHIMERE model run at high resolution was conducted for the year 2009 over Europe. The performance of the model is systematically assessed according to the type/location of the stations and time of year. Along the year, CHIMERE reproduces nicely the daily NO₂ and O₃ daily variability. Over the year, the model shows negative bias for NO₂ and a positive one for O₃ which is higher during winter and summer respectively. CHIMERE gives an overall satisfactory performance concerning the simulation of PM₁₀ concentrations (FB=-14%) and a good performance for prediction of PM_{2,5} concentrations (FB=4.9%). For sulfate, the model performs rather well during the summer (FB=7.1%) but overestimates the concentrations at spring time (FB=41.0%). Over the year, the total nitrate concentrations are underestimated (FB=-57.8%) but are better reproduced than particulate nitrate. Finally, the total ammonia is better simulated by the model during the spring (FB=4.0%) and the autumn (FB=-1.5%) seasons than during the summer (FB=-21.2). Along with work on the aerosol module in order to improve the urban modelling of SIA, the next steps will include calculation of the modelled PM_{2,5} and NO₂ urban increments for European cities and the integration of a standard methodology for the urban increment calculation to be implement in integrated assessment models.

1. INTRODUCTION

Over the last decades, numerous papers intended to evaluate the skills of CTMs to correctly estimate tropospheric air pollutants concentrations. Both constant evolution of model parameterisations and the increased quality of input data including meteorology and emissions should foster frequent CTMs assessments. A list of European model evaluation studies that took place during the last decade can be found in [Pay et al. \(2011\)](#). The CHIMERE model itself had undergone several time extensive evaluations ([Vautard et al. \(2007a\)](#); [Van Loon et al. \(2007\)](#)). However, we note that previous studies are generally coarser in terms of horizontal resolution and use fewer stations to achieve the validation part. The aim of the study is to comprehensively evaluate a fine resolution (0.0625x0.125°) CHIMERE runs throughout the Europe using the largest set of monitoring stations available in 2009. The analysis is performed for ozone (O₃), nitrogen dioxide (NO₂), PM₁₀, PM_{2,5} and PM compounds such as sulfate (SO₄²⁻), nitrate (NO₃⁻), total nitrate (HNO₃+NO₃⁻), ammonium (NH₄⁺), total ammonia (NH₃+NH₄⁺).

2. METHODOLOGY

2.1. CTM description

The purpose of CHIMERE is to calculate the concentrations of usual chemical species that are involved in the physico-chemistry of the low troposphere. CHIMERE has been described in detail in several papers: [Schmidt et al. \(2001\)](#) for the dynamics and the gas phase module; [Bessagnet et al. \(2008, 2009\)](#) for the aerosol module; [Vautard et al. \(2005, 2007b\)](#) for the latest substantial model improvements. The aerosol model species are the primary particle material, sulfates, nitrates, ammonium, organic aerosols, sea-salts and dust. For more detail on the latest developments one can refer to the online documentation (<http://www.lmd.polytechnique.fr/chimere>). For the study, we defined a nested fine resolution domain (324x410 grid boxes) that covers the whole of Europe from 10.4375°W to 29.9375°E in longitude and 35.9062°N to 61.4687°N in latitude with a resolution of 0.0625x0.125° (Figure 1). Boundary conditions are monthly mean climatology taken from the LMDz-INCA model for gaseous species ([Hauglustaine et al. \(2004\)](#)) and from the GOCART model for aerosols ([Ginoux et al. \(2001\)](#)). Data for comparison with observations are extracted from the lowest vertical level (20m on average). A complete and high resolution set of both biogenic and anthropogenic emissions are needed in order to perform CHIMERE computations. Six biogenic species (isoprene, α -pinene, β -pinene, limonene, ocimene, and NO) are calculated using the MEGAN model ([Guenther et al. \(2006\)](#)). We also account for wildfire emissions issue from the GFED3 ([Kaiser et al. \(2011\)](#)).

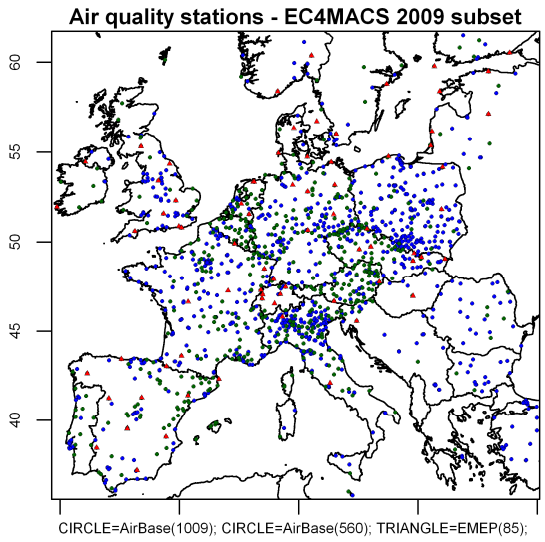


Figure 1: Airbase rural background (green), Airbase urban background (blue) and EMEP rural stations (red) projected on the domain used for the evaluation.

2.2. Meteorology

As CHIMERE is an off-line model, we had to select a set of meteorological data for the entire 2009 year. For this study, we shifted from the usual WRF limited area models data to the ECMWF Integrated Forecast System (IFS) data (<http://www.ecmwf.int/research/ifsdocs>).

Motivations include the systematic over wind speed estimation by WRF (Jimenez and Dudhia) and the save in time allows when using IFS data as no meteorological pre-runs are needed. The IFS model has a $0.25 \times 0.25^\circ$ horizontal grid spacing (T799) from surface to 0.1 hPa (91 levels in total). It delivers typical meteorological variables (temperature, wind components, specific humidity, pressure, sensible and latent heat fluxes) that are vertically and horizontally interpolated onto the CHIMERE grid (8 levels).

2.3. Anthropogenic emission pre-processing

The emission pre-processor transforms raw anthropogenic emissions in ton/year/cells to CHIMERE compliant spatialised emissions dataset. VOC, NO_x, CO, SO₂, NH₃, PPM (Primary Particle Material) annual emissions come from the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air pollutant EMEP (Vestreng (2003)). Two main steps can be identified in the anthropogenic emission pre-processing: the spatialisation and regridding of raw emission on the CHIMERE grid and the speciation/aggregation step. The first step consists in regridding national anthropogenic emission inventories onto the CHIMERE computational grid using the USGS database (<http://www.usgs.gov>). Annual NO_x emissions were speciated into NO, NO₂ and HNO₂ using the coefficients recommended by IIASA (personal communication). For NMVOC, the speciation was performed over 32 NMVOC NAPAP classes (Middleton et al. (1990)) and for the aggregation step the lumping of NMVOCs into model species is performed following Middleton et al. (1990). Time disaggregation was done on the basis of GENEMIS data using monthly, weekly and hourly coefficients depending on the activity sector (Society et al. (1994)). For SNAP2, we also propose a new temporal profile according to the daily ambient temperature (degree day concept).

2.4. Observation data

Observed data come from two different databases. The first one is Airbase (<http://acm.eionet.europa.eu/databases>) gathering regulatory data reported by Member States according to the air quality directives. For this study, we used two different Airbase types of station: the Rural Background stations (RB) and Urban Background stations (UB). We also used data from the EMEP network (<http://www.emep.int/>) that provides observations of SIA at remote Rural Background sites (RB) over a more extended area of the European Union. Stations with an altitude under 750 m were selected if 75% or more data are available over the year. Figure 1 displays the spatial distribution of the AIRBASE (green for RB and blue for UB) and the EMEP (red) stations used for the evaluation. Three key statistical indicators are selected for their ability to diagnose the model performance: the correlation index (R), the root mean square error (RMSE) and the fractional bias (FB). Details about the calculation of the statistics using Atmospheric Model Evaluation Tool software (AMET) can be found in Appel et al. (2011). The following section is based on yearly and seasonal mean of the statistical indicators.

3. RESULTS AND DISCUSSION

Figure 2 shows daily box-whisker plots time series of the NO₂, O₃, PM₁₀ and PM_{2.5} observed and calculated concentration averaged over all UB Airbase stations. Table 1 presents a few yearly mean statistical scores for gas and particulate pollutants. Along the year, CHIMERE catches nicely the temporal variability of NO₂ both at RB (R=0.65) and UB (R=0.61) sites but underestimates significantly the concentration especially during the winter season (FB=-65.9). This behaviour could be due to the general underestimation of NO_x urban emissions whose the impact is magnified by the winter time stagnant conditions that increase the NO₂

observed concentration at low level. Overall, daily temporal variability of O_3 concentrations is very well simulated both at rural ($R=0.78$) and urban background sites ($R=0.77$). The model reaches its lowest FB in the summer (15.8%) at UB sites. Over the year, the modelled values present a systematic positive bias which is higher at urban ($FB=28.5\%$) than at rural sites (19.8%). This tendency could be link to the underestimation of the NO_2 previously mention especially during the winter at urban sites. For the PM_{10} , at UB stations, R gets highest values during the summer and the autumn (0.46 and 0.48 respectively) and its lowest at spring (0.41). Conversely, the FB is lower in warm season (-1.4% at spring time) than in the winter (-30.5%). At RB site, the model performs better in terms of correlation (0.60 in the winter). Over the year, in opposition to the UB sites ($FB=-17\%$), CHIMERE overestimates the concentration ($FB=8.1\%$) at RB sites. For $PM_{2.5}$, the behaviour is similar to PM_{10} . However, we note a higher R value over the year both at UB sites (0.54) and RB sites (0.61) than for PM_{10} . At UB sites, CHIMERE performs better in autumn (0.56) than in summer (0.40). The highest R is observed at RB sites during the winter (0.69). The FB is very low during the summer at UB stations (1.8%) and negative during the winter (-11.9%). At RB sites the FB is always positive and the maximum is observed at spring time (28.9%) and the minimum at winter time (6.5%). We underline that CHIMERE gives, over the year, a good performance concerning the reproduction $PM_{2.5}$ concentrations ($FB=4.9\%$). PM_{10} , $PM_{2.5}$ and PM speciation data were available on several EMEP sites. As previously observed with the Airbase RB stations, CHIMERE overestimates the PM_{10} ($FB=19.9\%$) and $PM_{2.5}$ ($FB=20.1\%$) concentrations at the EMEP stations. A strong inter-seasonal variability is observed with a minimum FB during the winter (7.1% for PM_{10} and 0.6% for $PM_{2.5}$). The highest correlation coefficient is found during the winter (0.55 for PM_{10}) and is good for $PM_{2.5}$ (0.69). Concerning the secondary inorganic aerosol (SIA), an inter-seasonal variation is also noted. For the sulfate, the model performs rather well during the summer ($FB=7.1\%$) but strongly overestimates the concentration at spring time ($FB=41.0$). The nitrates are strongly underestimated along the year but rather high R value is noted during the winter (0.68). This underestimate is mainly due to the slight overestimate of sulfate and the missing coarse nitrate chemistry in CHIMERE. The total nitrate concentration is much better reproduced with $R>0.6$ over the year except during the summer (0.17). The order of magnitude of the FB indicates an underestimation of a factor 2. Along with sulfate, ammonium appears to be the best SIA compound reproduced by CHIMERE. The FB is rather low and indicates a slight overestimation during the winter (7.7%) and an underestimation during the summer (-5.9%). The total ammonia is nicely reproduced by CHIMERE with some low bias observed during the spring ($FB=4.0\%$) and autumn ($FB=-1.5\%$) seasons. The performance is worse during the summer where the model is underestimating the most ($FB=-21.2\%$).

Table 1: Yearly mean selected statistical indexes calculated using Airbase UB and RB (values in brackets) stations and using EMEP RB stations (SIA only). For observation mean, modelled mean and RMSE the units are: in ppb for NO_2 and O_3 , in $\mu g/m^3$ for PM_{10} and $PM_{2.5}$; in $\mu gS/m^3$ for sulfate, in $\mu gN/m^3$ for total nitrate and total ammonia. FB in %.

Pollutant	Nb stations	Observation mean	Modelled mean	R	RMSE	FB
NO_2	770 (300)	13.2 (6.6)	8.2 (4.9)	0.61 (0.65)	8.4 (4.7)	-54.4 (-26.8)
O_3	586(361)	23.6 (27.9)	29.5 (32.6)	0.77 (0.78)	9.8 (8.8)	28.5 (19.8)
PM_{10}	677 (238)	29.3 (20.6)	22.8 (20.2)	0.42 (0.48)	21.9 (13.2)	-14.0 (8.1)
$PM_{2.5}$	267 (92)	17.5 (13.6)	16.4 (14.1)	0.54 (0.61)	12.6 (10.0)	4.9 (20.5)
Sulfate	37	0.7	0.9	0.48	0.6	27.6
Total Nitrate	26	0.6	0.4	0.57	0.6	-57.8
Total Ammonia	14	1.5	1.4	0.60	1.1	-7.3

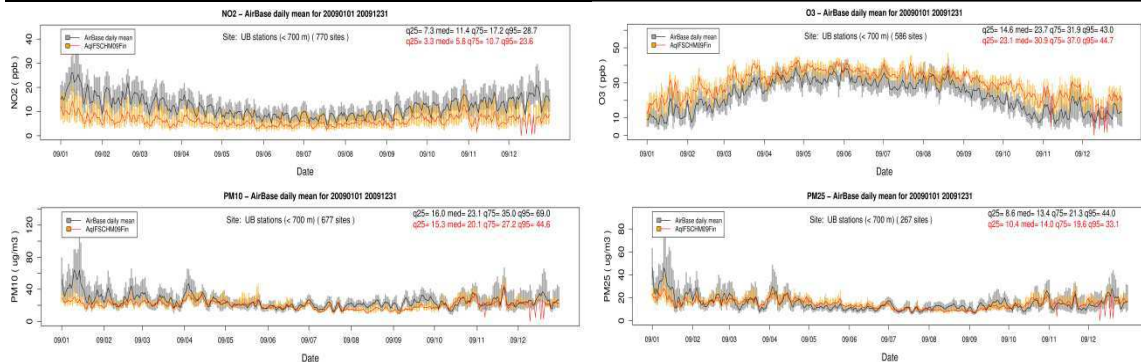


Figure 2: Daily box-whisker plots time series of the NO_2 , O_3 , PM_{10} and $PM_{2.5}$ observed and calculated concentration averaged over all UB Airbase stations. The continuous lines represent the medians and the bars show the 25th -75th quantile interval. The yearly 25th, 50th, 75th, and 95th quantiles are reported on the top right corner of the plots (in the legend AqIFSCHM09Fin refers to the CHIMERE simulation).

CONCLUSIONS

A comprehensive evaluation of the CHIMERE model was conducted for the year 2009. The performance of the model is systematically assessed according to the type/location of the stations and time of year. It reproduces correctly daily NO₂ variability along the year but underestimates significantly the concentration especially during the cold season. It simulates nicely the day to day O₃ variation similarly at urban and rural sites with an overestimation which is higher during the winter at urban sites. CHIMERE gives an overall satisfactory performance concerning the prediction of PM₁₀ concentrations (FB=-14%) and a good performance for PM_{2.5} concentrations (FB=4.9%). For the sulfate, the model performs rather well during the summer (FB=7.1%) but strongly overestimates the concentration at spring time (FB=41.0%). The total nitrate concentration is much better reproduced than nitrate stand alone with high R over the year (R>0.6). For total nitrate, the model underestimates the observation. Finally, the total ammonia is better reproduced by the model during spring (FB=4.0%) and autumn (FB=-1.5%) whereas the model performance is lower during the summer (FB=-21.2%). Along with work on the aerosol module in order to improve the urban modelling of SIA, the next steps will include calculation of the modelled PM_{2.5} and NO₂ urban increments for European cities and the integration of a standard methodology for the urban increment calculation to be implemented in integrated assessment models.

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