MODELLING ATMOSPHERIC PARTICLES IN THE IBERIAN PENINSULA AND BALEARIC ISLANDS

Marta G. Vivanco, Inmaculada Palomino, Fernando Martín and Magdalena Palacios.
Air Pollution Unit. CIEMAT. Madrid, Spain

INTRODUCTION

Besides ozone and nitrogen oxides, exceedances of air quality standards for other pollutants, such as particulate matter, are being recorded in Spain. Studies and measurements during the last years indicate that PM10 is the pollutant with a higher number of air quality standard exceedances in Spain. Many of the exceedances are caused by natural events such as Saharan dust transport.

Models have become a useful tool in air quality management, since European legislation determine the obligation to evaluate air quality and improve air quality in polluted areas. For this purpose, models must be able to reproduce all the physical and chemical processes affecting pollutant behaviour in the atmosphere.

In this paper we include the results of the application of the CHIMERE model to simulate PM10 and PM2.5 in Spain for 2004. Results were compared with measurements of Spanish stations.

MODELLING SETUP

Simulations were carried out using a regional version of the CHIMERE chemistry-transport model. This version (V200603par-rc1) calculates the concentration of 44 gaseous species and both inorganic and organic aerosols of primary and secondary origin, including primary particulate matter, mineral dust, sulphate, nitrate, ammonium, secondary organic species and water. A detailed description of the model configuration and performances over Europe are presented in previous studies (Bessagnet et al., 2004; Vautard et al., 2003).

Because of the possible influence of long range pollutant transport the model system was first used at European scale over a domain ranging from 10.5W to 22.5E and from 35N to 57.5 N with a 0.5 degree horizontal resolution and 14 vertical sigma-pressure levels extending up to 500 hPa. Downscaling was done for the Iberian Peninsula using a fine-scale domain with a 0.2 degree resolution and a one-way nesting procedure where coarse-grid simulations force the fine-grid ones at the boundaries without feedback.

The MM5 model (Grell et al., 1995) was used to obtain meteorological input fields. The simulations were carried out for a coarse domain and a finer one, with respective resolutions of 36 Km and 19 Km. MM5 simulations are forced by the National Centres for Environmental Prediction model (GFS) analyses at both scales, using a nudging procedure.

For both domains emissions were derived from the annual totals of the EMEP database for 2004 (Vestreng et al., 2005). Original EMEP emissions were disaggregated taking into account the land use information, in order to get higher resolution emission data. Boundary conditions for the coarse domain were provided from monthly climatology from LMDz-INCA model (Hauglustaine et al., 2004) for gases concentrations and from GOCART model (Chin et al., 2002) for particulate species, as described in Vautard et al. (2005).

These observations correspond to PM10 and PM2.5 daily averaged concentrations recorded at rural, suburban and urban background stations.

MODEL VALIDATION METHODOLOGY

Model results were compared with daily concentrations data of a set of Spanish stations. Data of these stations were selected and provided by Querol and Spanish Ministry of Environment (private communication) (see figure 1)

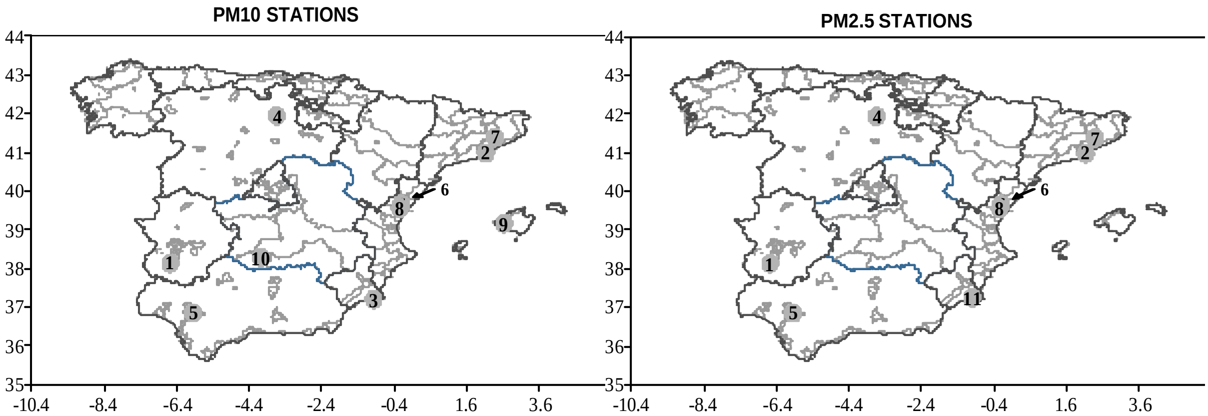

Fig. 1; Maps showing the locations of the PM10 and PM2.5 stations..

Model validation was done using graphical and statistical techniques. The used statistics were the following:

1. Normalized Mean Bias.

$$NMB = \frac{\sum_{1}^{N}\left(C_p - C_o\right)}{\sum_{1}^{N} C_o} \qquad (1)$$

2. Normalized absolute error.

$$NMSE = \frac{\sum_{1}^{N}\left|\left(C_p - C_o\right)\right|}{\sum_{1}^{N} C_o} \qquad (2)$$

where C_0, C_p and N are the observed and predicted concentrations and the number of cases, respectively.

Cut-offs of 10 and 5 $\mu gr/m^3$ were used to compute the above mentioned statistics for PM10 and PM2.5, respectively. They were computed for the whole year (Table 1) and also for the summer (April 1 – September 30) and winter (October 1 – March 30) periods in order to analyse possible seasonal variation (Table 2 and 3).

Table 1. Statistics for PM10.and PM2.5

station	PM10 NMB	PM10 NMSE	PM2.5 NMB	PM2.5 NMSE
1	21	53.1	-31.4	41.5
2	21.4	46.9	-9.2	41.8
3	-15.1	40.3	-57.4	57.6
4	-26.1	40.5		
5	10.5	47.1	-23.5	39.4
6	-3	41.8	-49	49.4
7	16.9	39.9	-19	39.2
8	9.3	40.1	-33.9	43.9
9	27.2	47.4		
10	-32.9	44.3		
11			-14	42.1
average	2.9	44.1	-29.7	44.4

Table 2. Statistics for PM10.and PM2.5 in winter time

station	PM10 NMB	PM10 NMSE	PM2.5 NMB	PM2.5 NMSE
1	9.3	57.4	-45.6	52.4
2	50.5	71.2	7.7	51.5
3	-3.3	48	-61.5	61.8
4	-22.8	46.9		
5	28.5	59.5	-26.5	48.7
6	11.1	57.6	-49.8	50.6
7	30	52.3	-11.7	42
8	14.7	55.5	-11.6	40.6
9	33.1	56.5		
10	-41.9	53.2		
11			-2	50.5
average	10.9	55.8	-25.1	49.8

Table 3. Statistics for PM10.and PM2.5 in summer time.

station	PM10 NMB	PM10 NMSE	PM2.5 NMB	PM2.5 NMSE
1	33.1	48.6	-17.4	30.9
2	-4.8	25.1	-24.6	33
3	-26.7	32.8	-53.3	53.3
4	-29.5	34.1		
5	-7.9	34.5	-21	31.7
6	-16.1	27.2	-48	48
7	5.8	29.4	-26.4	36.3
8	6.1	30.9	-45.7	45.7
9	22.4	39.9		
10	-26.7	38.2		
11			-25.1	34.4
average	-4.4	34	-32.7	39.1

RESULTS AND DISCUSSION

In the case of PM10, the CHIMERE model does not seem to show a clear trend to over or underprediction in average as shown by the low values of NMB in Table 1. However, there is underprediction for stations 3, 4 and 10, while overprediction is observed specially at stations 1, 2, 7 and 9. the NMSE values are relatively high in all the stations. A notable seasonal variation is noted (see table 2 and 3) in several stations with higher values of NMSE in wintertime than in summertime. In addition, the model tends to overpredict slightly in wintertime. In some cases as the station 2 (Barcelona), the seasonal variation is very remarkable in the statistics. The figure 2 shows clearly this behaviour. This fact could be due to a non-adequate time disaggregation of the EMEP emission data for some sector related to urban activities. In this figure, the effect of Saharan dust event is noted during some days of February with daily concentrations of almost 180 $\mu gr/m^3$. This event is also noted at the PM2.5 in some less extend (Figure 3). The model predictions in this event do not fit well the observations (underprediction) showing that the CHIMERE model or, more precisely, the inputs of the CHIMERE model must be improved to take into account correctly the long range transport of dust from Sahara Desert.

In the case of PM2.5, the model underpredicts but less in wintertime. It must be related to the emission data besides modelling causes. The values of NMSE are quite similar to those for PM10 showing the same seasonal behaviour.

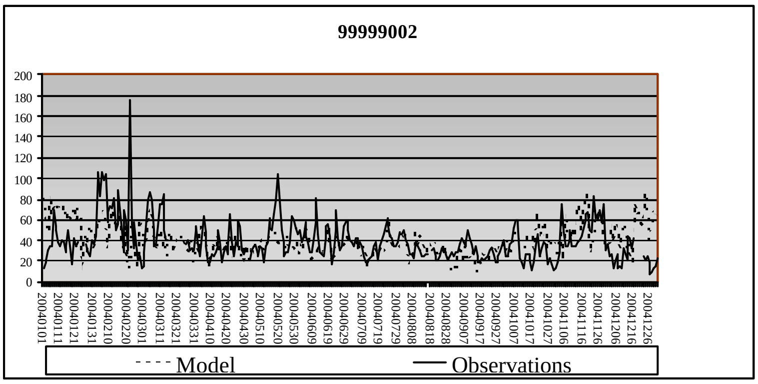

Fig. 2; Time series of daily PM10 concentration at station 2.

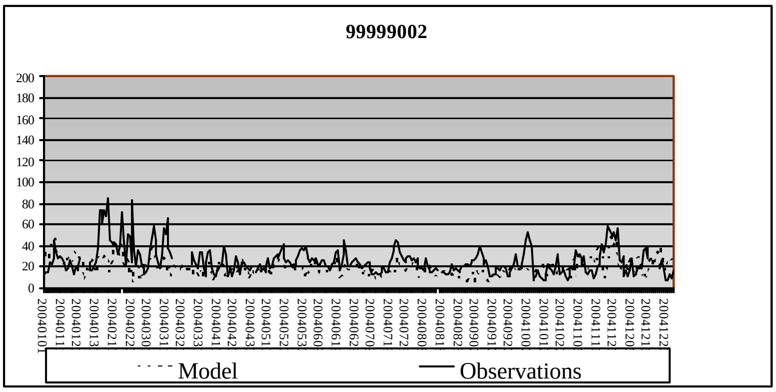

Fig. 3; Time series of daily PM2.5 concentration at station 2.

This work corresponds to the activities of model validation previous to the operational running of the CHIMERE model as a part of the CALIOPE model system, which is a model system for air quality forecasting in the Iberian Peninsula and Balearic Islands. CALIOPE project is being leaded by Barcelona Supercomputing Center and several institutions, besides CIEMAT, such as CSIC-Instituto Jaume Almera and CEAM are participating. Other models involved in this system are WRF and CMAQ. Validations for other pollutants can be seen in (Vivanco et al, 2007 and Jiménez-Guerrero et al, 2007). In addition, a higher spatial resolution emission data will be used in CALIOPE and then, it expected the model performance will be much better.

ACKNOWLEGDEMENTS

This work was financially supported by the Spanish Ministry of Environment by means of the project CALIOPE (ref. 441/2006/3-12.1). Authors thank to Dr. Xavier Querol for providing data of PM in the selected stations, and to Dr. Bertrand Bessagnet for his support in using the CHIMERE model.

REFERENCES

Bessagnet, B., A. Hodzic, R. Vautard, M. Beekmann, L. Rouil, R. Rosset, 2004: Aerosol modelling with CHIMERE –first evaluation at continental scale. Atmospheric Environment **38**, 2803-2817.

Chin, M., P. Ginoux, S. Kinne, B. N. Holben, B. N. Duncan, R. V. Martin, J. A. Logan, A. Higurashi, and T. Nakajima, 2002: Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sunphotometer measurements, J. Atmos. Sci. **59**, 461-483.

Grell G.A., Dudhia J., Stauffer D.R., 1995: A Description of the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5).NCAR/TN-398 + STR. NCAR TECHNICAL NOTE.

Hauglustaine, D. A., Hourdin, F., Jourdain, L., Filiberti, M.-A., Walters, S., Lamarque, J.-F., and Holland, E. A., 2004: In www.atmos-chem-phys.net/6/3487/2006/ Atmos. Chem. Phys., 6, 3487–3503, 2006 3502 A. Colette et al.: Impact of transport and transformation on ozone variability teractive chemistry in the Laboratoire de Meteorologie Dynamique general circulation model: Description and background tropospheric chemistry evaluation, J. Geophys. Res., **109**, doi:10.1029/2003JD003957.

Jimenez-Guerrero P., Vivanco M.G., Jorba O.; Palomino I., Pérez C., Martín F. and Baldasano J.M. , 2007: An intercomparison and evaluation study of two chemistry transport models in southwestern europe for an annual cycle. Presentation in the 2nd ACCENT Symposium, Urbino, Italy, July 23-27.

Tesche T.W., P. Georgopoulos, J.H. Seinfeld , G. Cass, F.W. Lurmann, and P.M. Roth, 1990: Improvement of procedures for evaluating photochemical models. Draft final report prepared for California Air Resources Board, Sacramento, CA by Radian.

Vautard. R., Bessagnet, B., Chin, M. and Menut, L., 2005: On the contribution of natural Aeolian sources to particulate matter concentrations in Europe: Testing hypotheses with a modelling approach, Atmospheric Environment., **39**, 3291-3303.

Vautard, R. D. Martin, M. Beekmann, P. Drobinski, R. Friedrich, A. Jaubertie, D. Kley, M. Lattuati, P. Moral, B. Neininger, J. Theloke, 2003: Paris emission inventory diagnostics from ESQUIF airborne measurements and a chemistry transport model. Journal of Geophysics Research **108** (D17), 8564-8585.

Vestreng, V., K. Breivik, M. Adams, A. Wagener, J. Goodwin, O. Rozovskkaya, J. M. Pacyna 2005, Inventory Review 2005, Emission Data reported to LRTAP Convention and NEC Directive, Initial review of HMs and POPs, Technical report MSC-W 1/2005, ISSN 0804-2446

Vivanco M.G., Palomino I.; Vautard, R.; Bessagnet, B.; Martín, F.; Menut, L.; Jiménez S., 2007: Multi-year assessment of photochemical air quality simulation over Spain. Submitted to Science of the Total Environment.