UDC 550.34.01+66.01

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SIMULATIONS OF PM10, PM2.5 AND OTHER POLLUTANTS DURING WINTER 2003 IN GERMANY: A MODEL EXPERIMENT WITH MM5-CMAQ AND WRF/CHEM MODELS

Abstract. *We have applied the MM5-CMAQ model to simulate the high concentrations in PM10 and PM2.5 during a winter episode (2003) in Central Europe. The selected period is January, 15 -April, 6, 2003. Values of daily mean concentrations up to 75 μgm^{-3} are found on average of several monitoring stations in Northern Germany. This model evaluation shows that there is an increasing underestimation of primary and secondary species with increasing observed PM10. The high PM levels were observed under stagnant weather conditions, that are difficult to simulate. The MM5 is the PSU/NCAR non-hydrostatic meteorological model and CMAQ is the chemical dispersion model developed by EPA (US) used in this simulation with CBM-V. The TNO emission inventory was used to simulate the PM10 and PM2.5 concentrations with the MM5-CMAQ model. The results show a substantial underestimation of the elevated values in February and March, 2003. An increase on the PM2.5 emissions (five times) produces the expected results and the correlation coefficient increases slightly. The WRF/CHEM model results show an excellent performance with correct emission database. The main difference between MM5-CMAQ simulations and WRF/CHEM is the MOSAIC particle models and the "classical" MADE/SORGAM particle model used in WRF/CHEM and CMAQ respectively. MOSAIC seems to make a better job than MADE particle model for this particular episode.*

Keywords: *emissions, PM10 and PM2.5, air quality models, air particles.*

1 Introduction

Simulations of elevated PM10 and PM2.5 concentrations have been always underestimated by modern three dimensional air quality modelling tools. This fact has focused much more attention between researchers during last years. Three dimensional air quality models have been developed during the last 15-20 years and substantial progress has occurred in this research area. These models are composed by a meteorological driver and a chemical and transport module. Examples of meteorological drivers are: MM5 (PSU.NCAR, USA), [9], RSM (NOAA, USA), ECMWF (Redding, U.K.), HIRLAM (Finnish Meteorological Institute, Finland), WRF [12] and examples of dispersion and chemical transport modules are EURAD (University of Cologne, Germany), [22], EUROS (RIVM, The Netherlands), [13], EMEP Eulerian (DNMI, Oslo, Norway), MATCH (SMHI, Norrkoping, Sweden), [6], REM3 (Free University of Berlin, Germany), [24], CHIMERE (ISPL, Paris, France), [19], NILU-CTM (NILU, Kjeller, Norway), [7], LOTOS (TNO, Apeldoorm, The Netherlands), [15], DEM (NERI, Roskilde, Denmark), [8], OPANA model [16,17,18] based on MEMO and MM5 mesoscale meteorological models and with the chemistry on-line solved by [11], STOCHEM (UK Met. Office, Bracknell, U.K.), [5] and CMAQ (Community Multiscale Air Quality modelling system) [3], developed by EPA (USA). In USA, CAMx Environ Inc., STEM-III (University of Iowa) and CMAQ model are the most up-to-date air quality dispersion chemical models. In this application we have used the CMAQ model (EPA, U.S.) which is one of the most complete models and includes aerosol, cloud and aerosol chemistry.

2 PM10 and PM2.5 episode

During the period January, 15, 2003 to April, 5, 2003, in central Europe (mainly northern part of Germany), we observe three high peaks on PM10 and PM2.5 values in several monitoring stations located in the area of North-East of Germany. The daily averages of PM10 concentrations were close to 80 μgm^{-3} and higher than 70 μgm^{-3} for PM2.5 concentrations. These values are about 4-5 times higher than those registered as "normal" values. The first peak on PM10 and PM2.5 concentrations was developed after Feb. 1 until Feb. 15. During this period of time, Central Europe was under the influence of a high-pressure system coming from Russia through Poland and Souther Scandinavia. In Northern part of Germany, we found southeasterly winds and stable conditions with low winds. These meteorological conditions brought daily PM10 concentrations at about 40 μgm^{-3}. The second peak was characterized by a sharp gradient on PM10 concentrations after Feb. 15 and until March, 7. These episode reached daily PM10 concentrations up to 70 μgm^{-3}. The meteorological conditions on March, 2 (peak values) was characterized by a wind rotation composed by Southwesterly winds from Poland over the North of Germany and Northwesterly and Western winds in the Central part of Germany. Finally a third peak with values of about 65 μgm^{-3} on March, 27 starts on March, 20 ending on April, 5. 2003. was having a similar structure and causes than the second one.

3 Emission data

In both models, we have applied the TNO emissions [23] as area and point sources with a geographical resolution of 0.125º latitude by 0.25º longitude and covering all Europe. The emission totals by SNAP activity sectors and countries agree with the baseline scenario for the Clean Air For Europe (CAFE) program [1]. This database gives the PM10 and PM2.5 emission for the primary particle emissions. We also took from CAFE the PM splitting sub-groups, height distribution and the breakdown of the annual emissions into hourly emissions. The PM2.5 fraction of the particle emissions was split into an unspecified fraction, elemental carbon (EC) and primary organic carbon (OC). The EC fraction of the PM2.5 emissions for the different SNAP sectors were taken from [20]. For the OC fraction, the method proposed by [2] is applied as follows: an average OC/EC emission ratio of two was used for all sectors, i.e. the OC fraction were set as twice the EC fractions, except if the sum of the two fractions exceed the unity. In this case (fEC>0.33), fOC was set as: fOC = 1 – fEC. With this prepared input, the WRF/CHEM and CMAQ took the information as it is. The hourly emissions are derived using sector-dependent, monthly, daily and hourly emission factors as used in the EURODELTA (http://aqm.jrc.it/eurodelta/) exercise.

4 Observational data

Eighteen PM10 stations were selected for the comparison with the model results. Seventeen stations represent the rural background and one station represent the urban background in Berlin. All stations are located in flat or moderate hill terrain. Most of the stations are operated by the respective Federal State agencies. At four stations (Neuglobsow, Zingst, Westerland and Deuselbach, which are EMEP background stations run by the German Environmental Protection Agency, Umweltbundesamt), the observed concentrations of particulate sulphate, total nitrate (HNO_3+NO_3^-) and total ammonia (NH_3+NH_4^+) were available. Deuselbach, in the southwest of Germany, is located outside of the high PM10 concentration region. In addition, at the research station Melpitz [21] the concentrations of the components of secondary inorganic aerosols SO_4^{--}, NO_3^-, NH_4^+, as well as the concentrations

of EC, OC and NH_3 were available. The SO_2 and NO_2 concentrations at these five stations were also taken into account in the model comparison. PM2.5 observations were available at four stations: Melpitz, Waldhof, Deuselbach and Hannover. All PM10 and PM2.5 observations are based on gravimetric measurements, and the concentrations of the inorganic species in aerosol particles on ion chromotography. The chemical composition data at Melpitz result from the PM2.5 fraction, whereas the composition data from the other stations were analyzed from the PM10 particle concentrations. OC data were corrected by a factor of 1.4 to account for the non-C atoms in the particulate organic matter (OM) concentrations, which are currently not measured [14].

5 MM5-CMAQ and WRF-CHEM architectures and configurations

MM5 was set up with two domains: a mother domain with 60x60 grid cells with 90 km spatial resolution and 23 vertical layers and 61x61 grid cells with 30 km spatial resolution with 23 vertical layers. The central point is set at 50.0 N and 10.0 E. The model is run with Lambert Conformal Conical projection. The CMAQ domain is slightly smaller following the CMAQ architecture rules. We use reanalysis T62 (209 km) datasets as 6-hour boundary conditions for MM5 with 28 vertical sigma levels and nudging with meteorological observations for the mother domain. We run MM5 with two-way nesting capability. We use the Kain-Fritsch 2 cumulus parameterization scheme, the MRF PBL scheme, Schultz microphysics scheme and Noah land-surface model. In CMAQ we use clean boundary profiles for initial conditions, Yamartino advection scheme, ACM2 for vertical diffusion, EBI solver and the aqueous/cloud chemistry with CB05 chemical scheme. Since our mother domain includes significant areas outside of Europe (North of Africa), we have used EDGAR emission inventory with EMIMO 2.0 emission model approach to fill those grid cells with hourly emission data. The VOC emissions are treated by SPECIATE Version 4.0 (EPA, USA) and for the lumping of the chemical species, we have used the [4] procedure, for 16 different groups. We use our BIOEMI scheme for biogenic emission modeling. The classical, Atkin, Accumulation and Coarse modes are used (MADE/SORGAM modal approach).

In WRF/CHEM simulation we have used only one domain with 30 km spatial resolution similar to the MM5. We have used the Lin et al. (1983) scheme for the microphysics, Yamartino scheme for the boundary layer parameterization and [10] for the biogenic emissions. The MOSAIC sectional approach is used with 4 modes for particle modeling.

6 Model results

The comparison between daily average values (averaged over all monitoring stations) of PM10 concentrations and modeled values has been performed with several statistical tools such as: Calculated mean/Observed mean; Calculated STD/Observed STD; bias; squared correlation coefficient (R2); RMSE/Obsrved mean (Root Mean Squared Error); percentage within +/-50% and number of data sets. Figure 1 shows the comparison between PM10 observed averaged daily values and the modeled values by MM5-CMAQ. The results show that MM5-CMAQ underestimates about 4 times the observed peak values and particularly the highest one on March, 2, 2003. The R2 coefficient is 0.69. Figure 2 shows similar information but for the WRF/CHEM results. In this case WRF/CHEM captures quite well the magnitude of the peaks, particularly the first one. For the second and third peak, the model undertimates about 20 % the peak values. The R2 coefficient is 0.61. In case of PM2.5 the Figures 3 and 4 shows similar results than Fig. 1 and 2. The R2 coefficients are 0.41 and 0.58. The squared correlation coefficient goes from 0.69 to 0.61 in the case of PM10 but increases substantially in case of PM2.5, from 0.41 to 0.58. In WRF/CHEM both R2 coefficients (for PM10

and PM2.5) are quite close (0.58 and 0.61) but in case of MM5–CMAQ, PM2.5 R2 coefficient is substantially lower than in case of PM10.

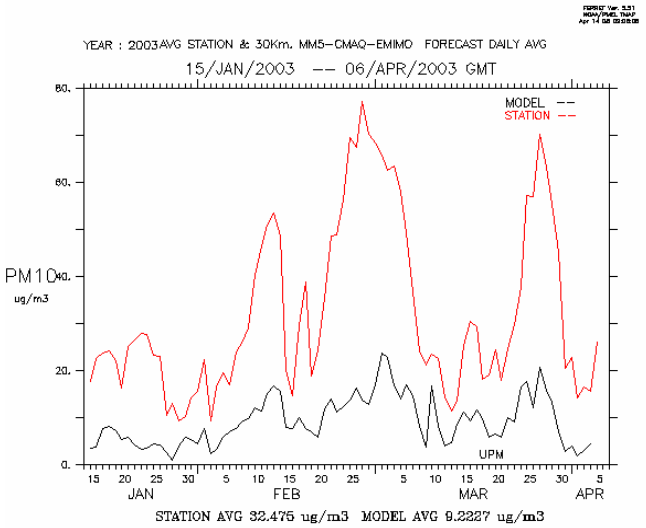

Fig. 1 – Comparison between daily average observed PM10 concentrations and model results produced by MM5-CMAQ. The model does not capture the magnitude of the PM10 peaks.

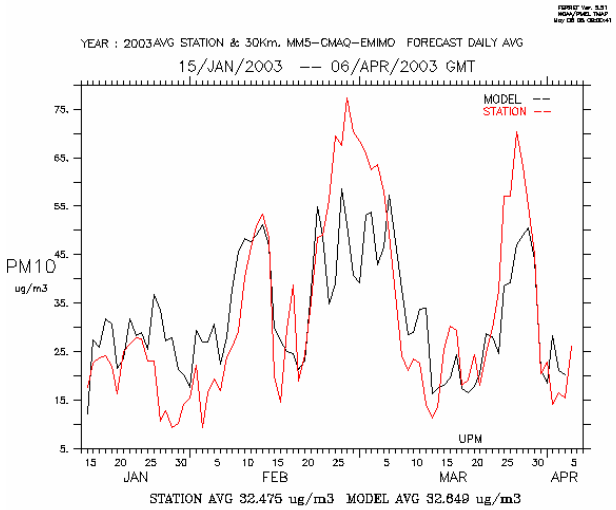

Fig. 2 – Comparison between daily average observed PM10 concentrations and model results produced by WRF/CHEM. The model captures quite well the magnitude of the PM10 peaks, particularly the first one.

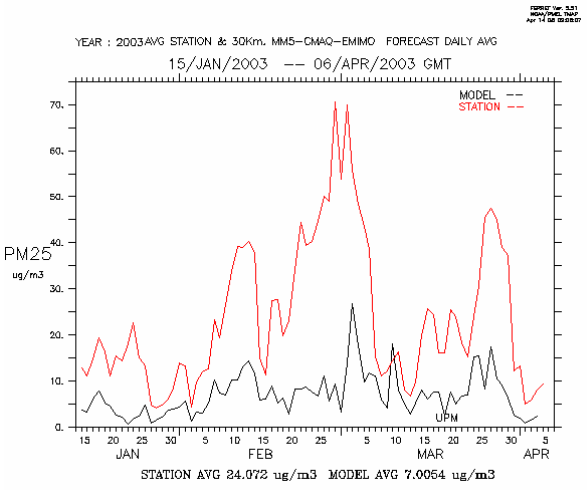

Fig. 3 – Comparison between daily average observed PM2.5 concentrations and model results produced by MM5-CMAQ. The model does not capture the magnitude of the PM2.5 peaks.

We performed another full experiment with MM5–CMAQ. We multiply by 5 the PM2.5 emissions provided by TNO in the whole domain. The results are shown in Figures 5 and 6. The results are surprisingly good for both species. The R2 coefficient is 0.70 and 0.48 for PM10 and PM2.5 respectively. In both cases the correlation is improved and particularly for PM2.5 although just slightly. It is difficult to explain these results but it is a fact.

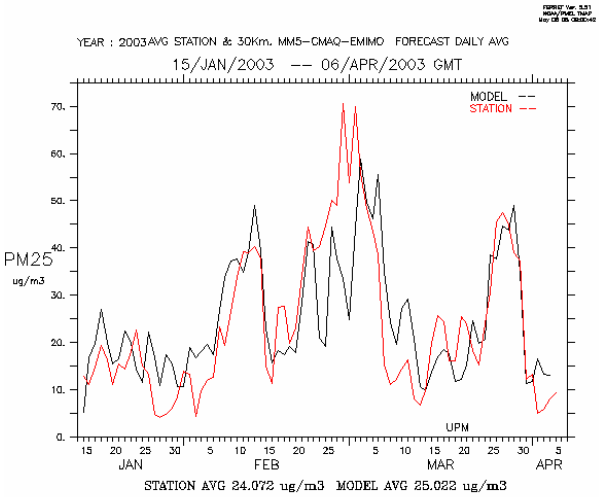

Fig. 4 – Comparison between daily average observed PM2.5 concentrations and model results produced by WRF/CHEM. The model captures quite well the magnitude of the PM10 peaks, particularly the last one.

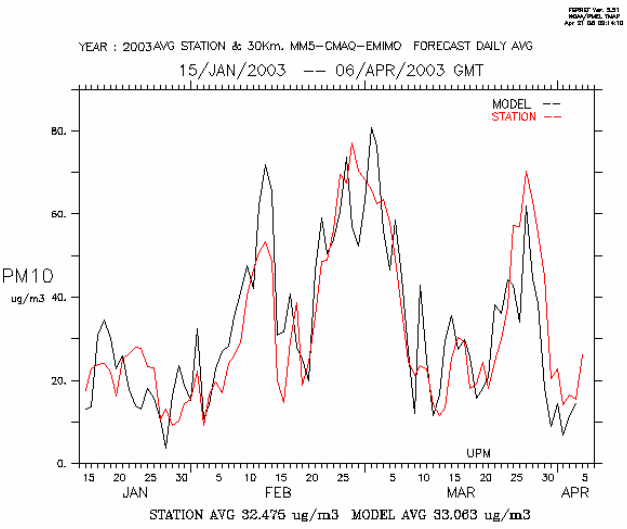

Fig. 5 – Comparison between daily average observed PM10 concentrations and model results produced by MM5-CMAQ with PM2.5 emissions multiplied by 5. The model captures quite well the magnitude of the PM10 peaks, particularly the second one.

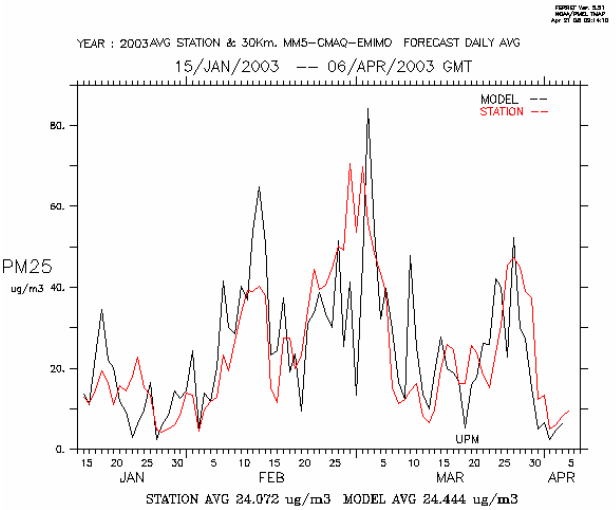

Fig. 6 – Comparison between daily average observed PM2.5 concentrations and model results produced by MM5-CMAQ with PM2.5 emissions multiplied by 5. The model captures quite well the magnitude of the PM10 peaks, particularly the third one.

7 Conclusions

We have implemented and run two different models (MM5-CMAQ and WRF-CHEM) for the same episode over Northern part of Germany during the winter period of 2003 (Jan. 15 -Apr. 5, 2003). WRF-CHEM made a better job than MM5-CMAQ, not only the patterns reproduce the peak values quite well but also the statistical parameters are good. The calculated mean values divided by thye observed mean value os exactly 1.0 for PM10 and WRF/CHEM on-line model. For the MM5-CMAQ this ratio is 0.28 and when we multiply the PM2.5 emissions by 5, the ratio is 1.02 which is also excellent. The bias values for WRF/CHEM, MM5-CMAQ and MM5-CMAQ (x5) are 0.09, -23.33 and 0.51 which are excellent values for WRF/CHEM and MM5-CMAQ (x5). No realistic explanation is found for the exercise related to multiply by 5 the PM2.5 emissions from TNO emission inventory. The main apparent reason why WRF/CHEM is doing much better job than normal MM5-CMAQ is the use of MOSAIC particle model based on sectional modal approach instead the "classical" approach based on MADE/SORGAM modal approach.

Acknowledgements. We would like to thank Dr. Peter Builtjes for then initial guidance and suggestion for this experiment and also COST 728 project (EU) where the inter-comparison experiment was proposed.

References

1. *Amann, M., I. Bertok, J. Cofala, F. Gyarfas, C. Heyes, Z. Klimon* (2005) Baseline Scenarios for the Clean Air for Europe (CAFE) Programme. Final Report, International Institute for Applied Systems Analysis, Schlossplatz 1, A–2361 Laxenburg, Austria.
2. *Beekmann, M., A. Kerschbaumer, E. Reimer, R. Stern, D. Möller* (2007) PM Measurement Campaign HOVERT in the Greater Berlin area: model evaluation with chemically specified observations for a one year period. Atmos. Chem. Phys., 7, pp. 55–68.
3. *Byun, D.W., J. Young, G. Gipson, J. Godowitch, F. Binkowsky, S. Roselle, B. Benjey, J. Pleim, J.K.S. Ching, J. Novak, C. Coats, T. Odman, A. Hanna, K. Alapaty, R. Mathur, J. McHenry, U. Shankar, S. Fine, A. Xiu, and C. Lang.* (1998) Description of the Models-3 Community Multiscale Air Quality (CMAQ) model. Proceedings of the American Meteorological Society, 78th Annual Meeting Phoenix, AZ, Jan. 11–16, pp. 264–268.
4. *Carter, W.P.L.* (2007) Development of the SAPRC-07 Chemical Mechanism and Updated Ozone Reactivity Scales. Final report to the California Air Resources Board Contract, No. 03-318, August. Available at http://www.cert.ucr.edu/~carter/SAPRC.
5. *Collins, W.J., D.S. Stevenson, C.E. Johnson and R.G. Derwent* (1997) Tropospheric ozone in a global scale 3D Lagrangian model and its response to NOx emission controls. J. Atmos. Chem., 86, pp. 223–274.
6. *Derwent, R., and M. Jenkin* (1991) Hydrocarbons and the long-range transport of ozone and PAN across Europe. Atmospheric Environment 8, pp. 1661–1678.
7. *Gardner, R.K., K. Adams, T. Cook, F. Deidewig, S. Ernedal, R. Falk, E. Fleuti, E. Herms, C. Johnson, M. Lecht, D. Lee, M. Leech, D. Lister, B. Masse, M. Metcalfe, P. Newton, A. Schmidt, C Vandenberg. and R. van Drimmelen* (1997) The ANCAT/EC global inventory of NOx emissions from aircraft. Atmospheric Environment 31, pp. 1751–1766.
8. *Gery M.W., G.Z. Whitten, J.P. Killus and M.C. Dodge* (1989) A photochemical kinetics mechanism for urban and regional scale computer modelling. Journal of Geophysical Research 94, D10, pp. 12925–12956.
9. *Grell, G.A., J. Dudhia and D.R. Stauffer* (1994) A description of the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5). NCAR/TN–398+STR, NCAR Technical Note.

10. *Guenther A., C.N. Hewitt, D. Erickson, R. Fall, C. Geron, T. Graedel, P. Harley, L. Klinger, M. Lerdau, W.A. McKay, T. Pierce, B. Scholes, R. Steinbrecher, R. Tallamraju, J. Taylor and P. Zimmerman* (1995) A global model of natural volatile organic compound emissions, Journal of Geophysical Research 100, pp. 8873–8892.
11. *Jacobson M.Z. and R.P. Turco* (1994) SMVGEAR: A sparse-matrix, vectorized GEAR code for atmospheric models. Atmospheric Environment 28, 2, pp. 273–284.
12. *Janjic, Z. I., J. P. Gerrity, Jr. S. Nickovic* (2001) An Alternative Approach to Nonhydrostatic Modeling. Monthly Weather Review, Vol. 129, pp. 1164-1178
13. *Langner J., R. Bergstrom and K. Pleijel* (1998) European scale modeling of sulfur, oxidized nitrogen and photochemical oxidants. Model development and evaluation for the 1994 growing season. SMHI report RMK No. 82, Swedish Met. And Hydrol. Inst., SE–601 76 Norrkoping, Sweden.
14. *Putaud, J., F. Raesa, R. Van Dingenen, E. Bruggemann, M. Facchini, S. Decesari, S. Fuzzi, R. Gehrig, C. Hueglin, P. Laj, G. Lorbeer, W. Maenhaut, N. Mihalopoulos, K. Mueller, X. Querol, S. Rodriguez, J. Schneider, G. Spindler, H. ten Brink, K. Torseth, A. Wiedensohler* (2004) A European aerosol phenomenology - 2: chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe. Atmospheric Environment 38, pp. 2579–2595.
15. *Roemer M., G. Boersen, P. Builtjes and P. Esser* (1996) The Budget of Ozone and Precursors over Europe Calculated with the LOTOS Model. TNO publication P96/004, Apeldoorn, The Netherlands.
16. *San José R., L. Rodriguez, J. Moreno, M. Palacios, M.A. Sanz and M. Delgado* (1994) Eulerian and photochemical modelling over Madrid area in a mesoscale context. Air Pollution II, Vol I., Computer Simulation, Computational Mechanics Publications, Ed. Baldasano, Brebbia, Power and Zannetti, pp. 209–217.
17. *San José R., J. Cortés, J. Moreno, J.F. Prieto and R.M. González* (1996) Ozone modelling over a large city by using a mesoscale Eulerian model: Madrid case study, Development and Application of Computer Techniques to Environmental Studies, Computational Mechanics Publications, Ed. Zannetti and Brebbia, pp. 309–319.
18. *San José, R., J.F. Prieto, N. Castellanos and J.M. Arranz* (1997) Sensitivity study of dry deposition fluxes in ANA air quality model over Madrid mesoscale area, Measurements and Modelling in Environmental Pollution, Ed. San José and Brebbia, pp. 119–130.
19. *Schmidt H., C. Derognat, R. Vautard and M. Beekmann* (2001) A comparison of simulated and observed ozone mixing ratios for the summer 1998 in Western Europe, Atmospheric Environment 35, pp. 6277–6297.
20. *Schaap, M., H. Denier van der Gon, A. Visschedijk, M. van Loon, H. ten Brink, F. Dentener, J. Putaud, B. Guillaume, C. Liousse, P. Builtjes* (2004a) Anthropogenic Black Carbon and Fine Aerosol Distribution over Europe, J. Geophys. Res., 109, D18207, DOI:10.1029/2003JD004330.
21. *Spindler, G., K. Mueller, E. Brueggemann, T. Gnauk, H. Herrmann* (2004) Long-term size-segregated characterization of PM10, PM2.5, and PM1 at the IfT research station Melpitz downwind of Leipzig (Germany) using high and low-volume filter samplers. Atmospheric Environment 38, pp. 5333–5347.
22. *Stockwell W., F. Kirchner, M. Kuhn and S. Seefeld* (1977) A new mechanism for regional atmospheric chemistry modeling, J. Geophys. Res. 102, pp. 25847–25879.
23. *Visscherdijk, A. and H. Denier van der Gon* (2005) Gridded European anthropogenic emission data for NOx, SO2, NMVOC, NH3, CO, PM10, PM2.5 and CH4 for the year 2000. TNO-report B&O-AR, 2005/106.
24. *Walcek C.* (2000) Minor flux adjustment near mixing ration extremes for simplified yet highly accurate monotonic calculation of tracer advection. J. Geophys. Res. 105, pp. 9335–9348.

Моделирование концентраций фракций PM10, PM2.5 и других загрязняющих веществ в течение зимнего периода 2003 в Германии: численные эксперименты с моделями MM5-CMAQ и WRF/CHEM

Аннотация. *Авторы статьи применили модель MM5-CMAQ для моделирования высоких концентраций загрязняющих веществ с диаметрами менее 10.0 и 2.5 мкм (фракции PM10 и PM2.5, соответственно) в Центральной Европе на протяжении зимнего периода 2003 г. Выбран период с 15 января по 6 апреля 2003 г. Значения среднесуточных концентраций, достигающие 75 μgm^{-3}, были выявлены в основном на некоторых станциях мониторинга в северной Германии. Оценки модели показали, что при увеличении измеренных концентраций PM10 имеет место увеличение недооценки первичного и вторичного типов загрязняющих веществ. Высокие концентрации фракций PM наблюдались при застойных погодных условиях, которые трудно смоделировать. MM5 является негидростатической моделью PSU/NCAR, а CMAQ – это химическая дисперсионная модель, разработанная в EPA (US) и используемая в этом эпизоде совместно с моделью CBM–V. Для того чтобы в модели MM5–CMAQ смоделировать концентрации фракций PM10 и PM2.5 использовался кадастр вредных выбросов, составленный Netherlands Organization for Applied Scientific Research. Результаты показали существенную недооценку значений концентраций на высотах в феврале и марте 2003 г. Увеличение выбросов PM2.5 (в пять раз) дает ожидаемый результат, и коэффициент корреляции увеличивается незначительно. Результаты модели WRF/CHEM показали хорошую согласованность с стандартной базой данных выбросов. Главное отличие между моделированием с помощью MM5–CMAQ и WRF/CHEM являются модели MOSAIC и MADE/SORGAM используемые в WRF/CHEM и CMAQ, соответственно. Представляется, что модель MOSAIC дает лучшие результаты для рассмотренных эпизодов, чем модель MADE.*

Ключевые слова: *выбросы, фракции PM10 и PM2.5, модели качества воздуха, атмосферные частицы.*