**EURAD-IM forecast and analysis system**

EURAD-IM is an Eulerian meso-scale chemistry transport model involving advection, diffusion, chemical transformation, wet and dry deposition and sedimentation of tropospheric trace gases and aerosols (Hass et al., 1995, Memmesheimer et al., 2004). It includes 3d-var and 4d-var chemical data assimilation (Elbern et al., 2007) and is able to run in nesting mode. As meteorological driver the Weather Research and Forecasting Model (WRF) is applied. EURAD-IM has been applied on several recent air pollution studies (Monteiro et al., 2013; Zyryanov et al., 2012; Monteiro et al, 2012; Elbern et al. 2011; Kanakidou et al., 2011).

The EURAD-IM assimilation system includes (i) the EURAD-IM CTM and its adjoint, (ii) the formulation of both background error covariance matrices for the initial states and the emission, and their treatment to precondition the minimisation problem, (iii) the observational basis and its related error covariance matrix, and (iv) the minimisation including the transformation for preconditioning. The quasi-Newton limited memory L-BFGS algorithm described in Nocedal (1980) and Liu and Nocedal (1989) is applied for the minimization. Following Weaver and Courtier (2001) with the promise of a high flexibility in designing anisotropic and heterogeneous influence radii, a diffusion approach for providing the background error covariance matrices is implemented.

The positive definite advection scheme of Bott (1989) is used to solve the advective transport. An Eddy diffusion approach is used to parameterize the vertical sub-grid-scale turbulent transport. The calculation of vertical Eddy diffusion coefficients is based on the specific turbulent structure in the individual regimes of the planetary boundary layer (PBL) according to the PBL height and the Monin-Obukhov length (Holtslag and Nieuwstadt, 1986). A semi-implicit (Crank-Nicholson) scheme is used to solve the diffusion equation.

Gas phase chemistry is represented by the Regional Atmospheric Chemistry Mechanism (RACM) (Stockwell et al., 1997) and an extension based on the Mainz Isoprene Mechanism (MIM) (Geiger et al., 2003). A two-step Rosenbrock method is used to solve the set of stiff ordinary differentials equations (Sandu et al., 2003, Sandu and Sander, 2006). Photolysis frequencies are derived using the FTUV model according to Tie et al. (2003). The radiative transfer model therein is based on the Tropospheric Ultraviolet-Visible Model (TUV) developed by Madronich and Weller (1990). The modal aerosol dynamics model MADE (Ackermann et al., 1998) is used to provide information on the aerosol size distribution and chemical composition. To solve for the concentrations of the secondary inorganic aerosol components, a FEOM (fully equivalent operational model) version, using the HDMR (high dimensional model representation) technique (Rabitz et al., 1999, Nieradzik, 2005), of an accurate mole fraction based thermodynamic model (Friese and Ebel, 2010) is used. The updated SORGAM module (Li et al., 2013) simulates secondary organic aerosol formation.

The MACC-II inventory for the period 2009 with 7 km x 7 km resolution is used for anthropogenic emissions (Kuenen et al., 2014). Biogenic emissions are calculated in the EURAD-IM CTM with the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2012). Additionally, emissions from fires are taken into account using the GFASv1.1 product (Kaiser et al., 2012) available daily at 0.1°x0.1° resolution.

The gas phase dry deposition modelling follows the method proposed by Zhang et al. (2003). Dry deposition of aerosol species is treated size dependent using the resistance model of Petroff and Zhang (2010). Wet deposition of gases and aerosols is derived from the cloud model in the EPA Models-3 Community Multiscale Air Quality (CMAQ) modelling system (Roselle and Binkowski, 1999).

**References**

Ackermann, I.J., H. Hass, M. Memmesheimer, A. Ebel, F.S. Binkowski, and U. Shankar, Modal aerosol dynamics model for Europe: Development and first applications, *Atmos. Environ.*, **32**, 2981-2999, 1998.

Bott, A., A positive definite advection scheme obtained by non-linear re-normalization of the advection fluxes, *Mon. Wea. Rev*., **117** (5), 1006-1015, 1989.

Binkowski, F.S., Aerosols in Models-3 CMAQ, in: *Science algorithms of the EPA Models-3 Community multiscale air quality (CMAQ) modeling system*, EPA 600/R-99-030, EPA, 1999.

Elbern, H.,Strunk, **A**., Schmidt and Talagrand, O., Emmision rate and chemical state estimation by 4-dimensional variational inversion, *Atmos. Chem. Phys*., **7**, 3749-3769, 2007

Elbern, H., A. Strunk, E. Friese, and L. Nieradzik, Assessment of Source/Receptor Relations by Inverse Modelling and Chemical Data Assimilation, in Persistent Pollution Past, Present and Future School of Environmental Research - Helmholtz-Zentrum Geesthacht, Quante, M.; Ebinghaus, R.; Flöser, G. (Eds.) 1st Edition, ISBN 978-3-642-17420-9, 2011.

Friese, E. and A. Ebel, Temperature dependent thermodynamic model of the system H+-NH₄+-Na+-SO₄²⁻-NO₃⁻-Cl⁻-H₂O, *J. Phys. Chem. A*, **114**, 11595-11631, 2010.

Geiger, H., I. Barnes, I. Bejan, T. Benter, and M. Spttler, The tropospheric degradation of isoprene: an updated module fort he regional atmospheric chemistry nechanism, *Atmos. Environ*., **37**, 1503-1519, 2003.

Guenther, A. B., X. Jiang, C.L. Heald, T. Sakulyanontvittaya, T. Duhl, L.K. Emmons, X. Wang, The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1). An extended and updated framework for modeling biogenic emissions. *Geosci. Model Dev.,* **5**, 1471-1492, 2012.

Hass, H., H.J. Jakobs, and M. Memmesheimer, Analysis of a regional model (EURAD) near surface gas concentration predictions using observations from networks, *Met. Atmos. Phys.*, 57, 173-200, 1995.

Holtslag, A.A.M. and F.T.M. Nieuwstadt, Scaling the atmospheric boundary layer, *Boundary-Layer Met*., **36**, 201-209, 1986.

Kaiser, J.W., A. Heil, M.O. Andreae, A. Benedetti, N. Chubarova, L. Jones, J.-J. Morcrette, M. Razinger, M.G. Schultz, M. Suttie, and G.R. van der Werf, Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power, *Biogeosciences*, **9**, 527-554, 2012.

Kanakidou, M., M. Dameris, H. Elbern, M. Beekmann, I. Konovalov, L. Nieradzik, A. Strunk, M. Krol, Synergistic use of retrieved trace constituents distributions and numerical modelling, in "The remote sensing of tropospheric composition from space", J. Burrows, U. Platt, P. Borrell (Eds.), Springer, doi 10.1007/978-3-642-14791-3, 2011.

Kuenen, J. J. P., A.J.H. Visschedijk, M. Jozwicka, and H.A.C. Denier van der Gon: TNO-MACC\_II emission inventory: a multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling, Atmos. Chem. Phys. Discuss., 14, 5837-5869, 2014.

Li, Y.P. H. Elbern, K.D. Liu, E. Friese, A. Kiendler-Scharr, Th.F. Mentel, X.S. Wang, A. Wahner, and Y.H. Zhang, Updated aerosol module and its application to simulate secondary organic aerosols during IMPACT campaign May 2008, *Atmos. Chem. Phys.*, **13**, 6289 - 6304, 2013, doi:10.5194/acp-13-6289-2013.

Liu, D.C. and J. Nocedal, On the limited memory BFGS method for large scale optimization, *Math. Programming*, **45**, 503-528, 1989.

Madronich, S. and G. Weller, Numerical integration errors in calculated tropospheric photodissociation rate coefficients, *J. Atmos. Chem*., **10**, 289-300, 1990.

Memmesheimer, M., E. Friese, A. Ebel, H. J. Jakobs, H. Feldmann, C. Kessler and G. Piekorz, Long-term simulations of particulate matter in Europe on different scales using sequential nesting of a regional model, *Int. J. Environm. and Pollution*, **22**, (1-2), 108-132, 2004.

Monteiro, A., I. Ribeiro, O. Tchepel, E. Sá, J. Ferreira, A. Carvalho, V. Martins, A. Strunk, S. Galmarini, H. Elbern, M. Schaap, P. Builtjes, A. I. Miranda, and C. Borrego, Bias Correction Techniques to Improve Air Quality Ensemble Predictions: Focus on O3 and PM Over Portugal, Environ. Model. Assess., **18**, 533-546, 2013,doi:10.1007/s10666-013-9358-2.

Monteiro, A., A. Strunk, A. Carvalho, O. Tchepel, A. I. Miranda, C. Borrego, S. Saavedra, A. Rodriguez;, J. Souto, J. Casares, E. Friese, H. Elbern, Investigating a very high ozone episode in a rural mountain site, Env. Pol., 162, 176-189, 2012.

Nieradzik, L.P., *Application of a high dimensional model representation on the atmospheric aerosol module MADE of the EURAD-CTM*, Master Thesis, Institut für Geophysik und Meteorologie der Universität zu Köln, 2005.

Nocedal, J., Updating quasi-Newton matrices with limited storage, *Math. Comput*., **35** (151), 773-782, 1980.

Petroff, A. and L. Zhang, Development and application of a size-resolved particle dry deposition scheme for application in aerosol transport models, *Geosci. Model Dev*., **3**, 753-769, doi: 10.5197/gmd-3-753-2010.

Rabitz, H., Ö.F.. Alis, General foundations of high-dimensional model representations, *J. Math. Chem*., **25**, 197-233, 1999.

Roselle, S.J. and F.S. Binkowski, Cloud Dynamics and Chemistry, in: *Science algorithms of the EPA Models-3 Community multiscale air quality (CMAQ) modeling system*, EPA 600/R-99-030, EPA, 1999.

Sandu, A., D. N. Daescu, and G.R. Carmichael, Direct and adjoint sensitivity analysis of chemical kinetic systems with KPP : part I – theory and software Tools, *Atmos. Environ*., **37**, 5083-5096, 2003.

Sandu, A. and R. Sander, Technical node: Simulating chemical systems in Fortran90 and Matlab with the Jinetic PreProcessor KPP-2.1, *Atmos. Chem. Phys*., **6**, 187-195, 2006.

Stockwell, W.R., F. Kirchner, M. Kuhn, and S. Seefeld, A new mechanism for regional atmospheric chemistry modeling, *J. Geophys. Res*., **102** (D22), 25847-25879, 1997, doi: 10.1029/97JD00849.

Tie, X., S. Madronich, S. Walters, R. Zhang, P. Rasch, and W. Collins, Effect of clouds on photolysis and oxidants in the troposphere, *J. Geophys. Res*., **108** (D20), 4642, doi : 10.1029/2003JD003659, 2003.

Weaver, A. and P. Courtier, Correlation modeling on the sphere using a generalized diffusion equation, *Q. J. R. Meteorol. Soc*., **127**, 1815-1846, 2001.

Zhang, L., J.R. Brook, and R. Vet, A revised parameterization for gaseous dry deposition in air-quality models, *Atmos. Chem. Phys*., 3, 2067-2082, 2003.

Zyryanov, D., G. Foret, M. Eremenko, M. Beekmann, J.-P. Cammas, M. D'Isidoro, H. Elbern, J. Flemming, E. Friese, I. Kioutsioutkis, A. Maurizi, D. Melas, F. Meleux, L. Menut, P. Moinat, V.-H. Peuch, A. Poupkou9, M. Razinger, M. Schultz, O. Stein, A. M. Suttie, A. Valdebenito, C. Zerefos, G. Dufour, G. Bergametti, and J.-M. Flaud, 3-D evaluation of tropospheric ozone simulations by an ensemble of regional Chemistry Transport Models, Atmos. Chem. Phys., 12, 3219-3240, 2012, doi:10.5194/acp-12-3219-2012.