SPECIAL PROJECT FINAL REPORT

Project Title:	Monitoring Atmospheric Climate and Composition – Interim Implementation (MACC-II)
Computer Project Account:	SP DEFRIU
Start Year - End Year :	June 2011 – June 2014
Principal Investigator(s)	Hendrik Elbern
Affiliation/Address:	Rhenish Institute for Environmental Research at the University of Cologne, RIU (RIUUK) Aachener Str. 209 D-50931 Köln Germany Email: he@eurad.uni-koeln.de
Other Researchers (Name/Affiliation):	Elmar Friese Rhenish Institute for Environmental Research at the University of Cologne

Summary of project objectives

MACC-II (Monitoring Atmospheric Composition and Climate – Interim Implementation) is an interim stage in the development of the Copernicus Atmosphere Service. Its objective is to function as the bridge between the developmental precursor project MACC and the Atmosphere Service envisaged to form part of Copernicus Operations for 2014-2020.
The continental-to-local scale chemical data assimilation EURAD-IM system with both its full 4d-var and 3d-var assimilation options is implemented. The system operates to perform the tasks invoked by MACC-II for RIUUK. These tasks include development and delivery of improved data assimilation methods to assure optimal exploitation of observations, assimilation of available data from both in situ and satellite devices, provision of pre-operational NRT daily air quality forecasts, and performance of regional air quality analyses for the years 2010-2012.

Summary of problems encountered

None

Experience with the Special Project framework

We are satisfied with the application procedure and with the requirements on project reporting.

Summary of results

1 Data Assimilation for European Air Quality (EDA)

The principal objective of sub-project EDA is to assure increasing and skilful use of new trace gas and aerosol measurements or retrievals, within a scenario of changing earth observation data compositions, retrieval versions and model configurations. These developments will be provided for prototype operational use in MACC-II sub-projects ENS and EVA.
Furthermore this sub-project aims to provide for validity of the data assimilation in case of extraordinary geophysical or atmospheric events and to investigate the capacity of four dimensional variational emission inversion to improve the skill of AQ forecasts.

1.1 Assimilation of SYNAER AOT retrievals

The impact of assimilation of SYNAER AOT retrievals on forecast improvement has been investigated for the time span from July 1 to 15, 2003 (Nieradzik, 2011). For this purpose a reference run without assimilation, analysis runs with assimilation of EEA surface in situ measurements only, assimilation of SYNAER AOT retrievals only, and combined assimilation of EEA PM_{10} measurements and SYNAER retrievals, have been performed. First guess runs have been initialised with the final state of the according analysis run for the previous day. An influence of both PM_{10} in situ measurements and SYNAER retrievals on the results of the combined assimilation run is clearly visible (see Figure 1.1).

To estimate the influence of the assimilation of SYNAER AOT retrievals on the forecast performance, quotients of first guess bias and reference run bias for PM_{10} in situ measurements are shown in the left panel of Figure 1.2. The right panel exhibits the respective quotients of the RMSE. For each day of the time period from July 1 to 15, 2003 a small to moderate bias as well as RMSE reduction due to assimilation of SYNAER retrievals could be achieved.

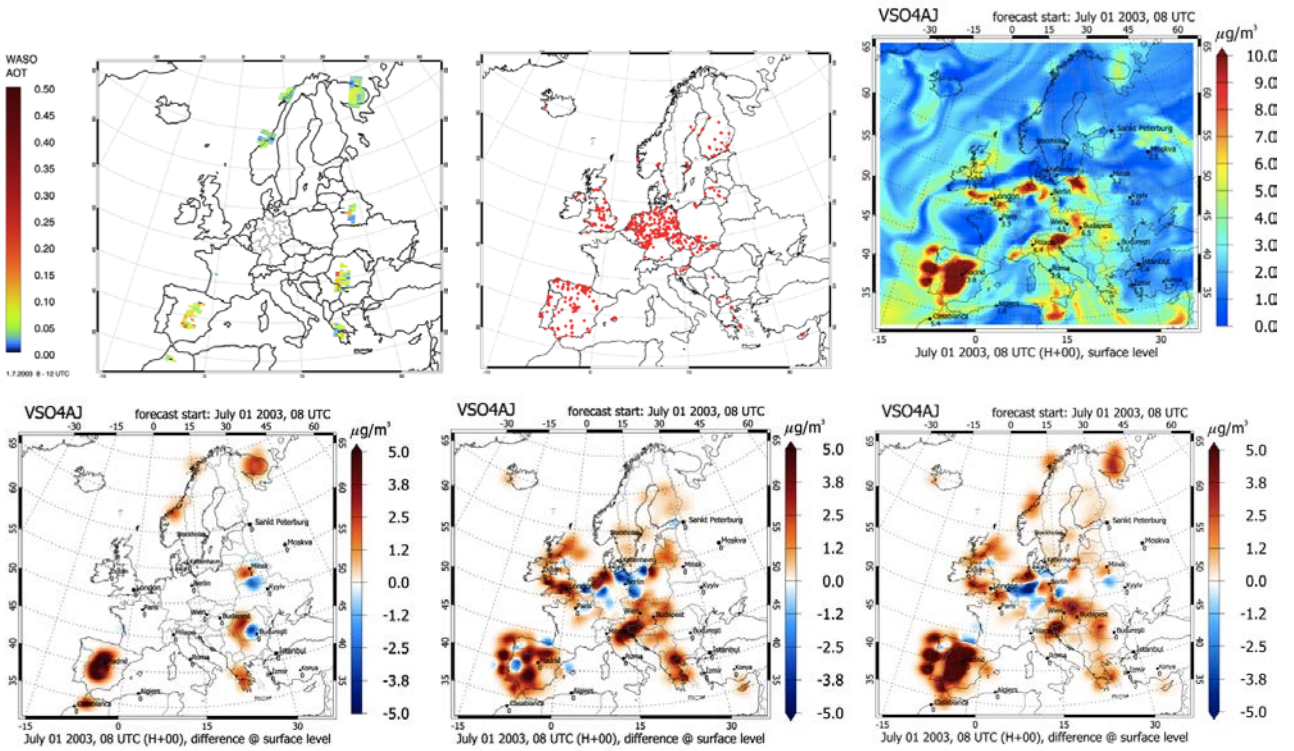


Figure 1.1: Footprints of SYNAER AOT retrievals for July 1, 2003 at 08:00 UTC exemplary shown for the water soluble aerosol class (upper left), locations of EEA PM_{10} in situ measurement sites (upper middle), accumulation mode SO_4^{2-} concentrations for July 1, 2003 at 08:00 in the near surface layer from an analysis run with combined assimilation of SYNAER AOT retrievals and PM_{10} surface in situ measurements (upper right), and differences in near surface layer accumulation mode SO_4^{2-} concentrations between an assimilation run and the reference run for assimilation of SYNAER retrievals only (lower left), EEA PM_{10} measurements only (lower middle), and combined assimilation of SYNAER retrievals and PM_{10} in situ data (lower right).

The bias reduction by means of combined assimilation of PM_{10} in situ measurements and SYNAER retrievals is comparable to the reduction by means of assimilation of PM_{10} in situ data only. An average bias reduction of 0.96, 0.67, and 0.67 could be achieved due to assimilation of SYNAER data only, EEA PM_{10} measurements only, and combined assimilation of SYNAER retrievals and EEA measurements. A RMSE reduction of 0.99, 0.92, and 0.93, respectively, could be achieved.

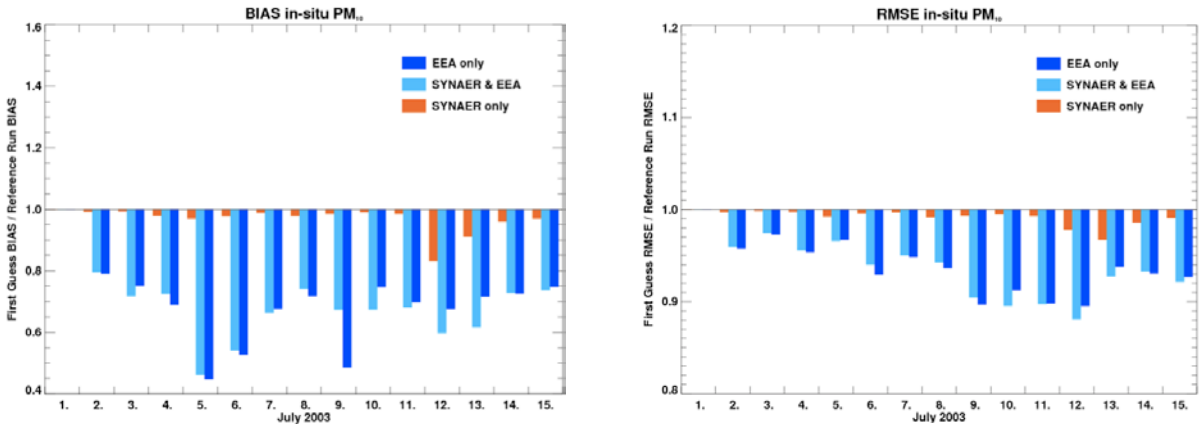


Figure 1.2: Quotient of first guess bias and reference run bias for in situ PM_{10} measurements for each day of the time span from July 1 to 15, 2003. Quotients are shown for the initialisation of the first guess with an analysis of EEA PM_{10} in situ measurements only (dark blue), combined in situ PM_{10} measurements and SYNAER AOT retrievals (light blue), and an analysis of SYNAER retrievals only (orange).

Taking into account that SYNAER measurements may fill spatial gaps between the locations of EEA PM_{10} measurement sites, the combined assimilation of PM_{10} surface in situ measurements and SYNAER AOT retrievals is able to improve the forecast performance.

1.2 Assimilation of MOPITT CO profiles

MOPITT (Measurements Of Pollution In The Troposphere) is particularly sensitive to the abundance of CO in the middle troposphere near 500hPa. Data covers all longitudes but is restricted to latitudes from 65°S to 65°N because of the difficulty eliminating cloud contamination. MOPITT pixels are square 22x22km. The swath width is approximately 640km, so it takes about three days to obtain approximately complete global coverage. The vertical resolution is defined by several broad and overlapping weighting functions. CO is output at seven pressure levels. The lowest level is at 900hPa. Figure 1.3 shows a comparison between a pure forecast without data assimilation and an analysis with 3d-var assimilation of MOPITT CO profile retrievals for June 15, 2012. Model results for the lowest MOPITT data level at 900hPa are depicted in the upper panel. Differences up to about 50ppbV between the forecast and the analysis could be obtained. Within EURAD-IM the background error covariance matrix is implemented following Elbern et al. (2007) using the diffusion approach by Weaver and Courtier (2001). Due to the fact that the vertical diffusion coefficient is calculated depending on the current meteorological conditions, this approach allows the vertical propagation of observation minus forecast differences from the lowest MOPITT data level to the surface. The lower panel of Figure 1.3 shows that the assimilation of MOPITT CO profiles can also improve model skill at the surface.

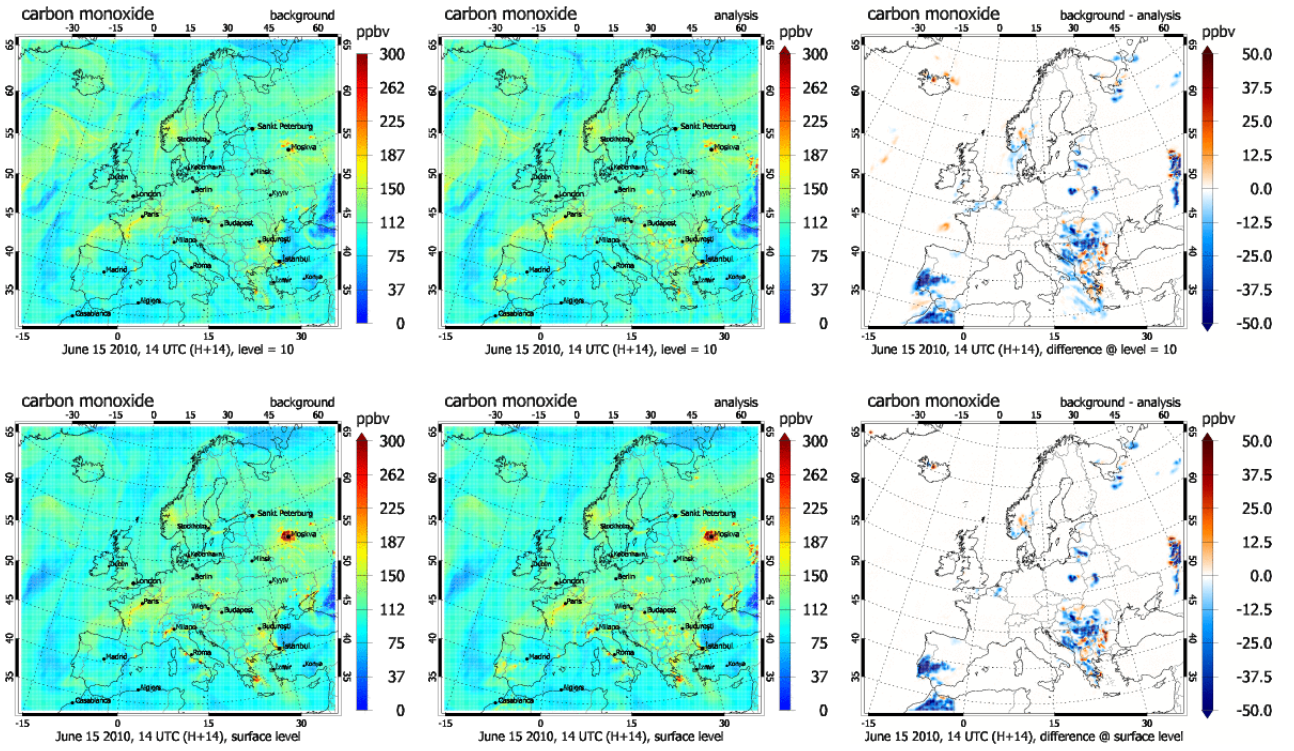


Figure 1.3: CO mixing ratio at 900hPa (upper panels) and in the near surface layer (lower panels) for June 15, 2012 as simulated with the EUTAD-IM system. Left: pure forecast without assimilation, middle: 3d-var assimilation of MOPITT CO profiles, right: differences between forecast and assimilation run.

1.4 Assimilation of IASI Ozone Columns

IASI is an infrared Fourier transform spectrometer developed jointly by CNES (the French spatial agency) with support of the scientific community, and by EUMETSAT. IASI is mounted on-board the European polar-orbiting MetOp satellite with the primary objective to improve numerical weather predictions, by measuring tropospheric temperature and humidity with high horizontal resolution and sampling, and with 1 km vertical resolution. As a second priority IASI contributes to

atmospheric composition measurements for climate and chemistry applications (Clerbaux et al., 2009). To reach these two objectives, IASI measures the infrared radiation of the Earths surface and of the atmosphere between 645 and 2760 cm^{-1} at nadir and along a 2200 km swath perpendicular to the satellite track. A total of 120 views are collected over the swath, divided as 30 arrays of 4 individual Field-of-views varying in size from 36 × π km^2 at nadir (circular 12 km diameter pixel) to 10 × 20 × π km2 at the larger viewing angle (ellipse-shaped Field-of-view at the end of the swath). IASI offers in this standard observing mode global coverage twice daily, with overpass times at around 9:30 and 21:30 mean local solar time.

An observations operator for IASI partial Ozone columns and its adjoint has been developed and was integrated in the EURAD-IM assimilation system. Figure 1.4 shows results from an assimilation of IASI O_3 columns in the free troposphere.

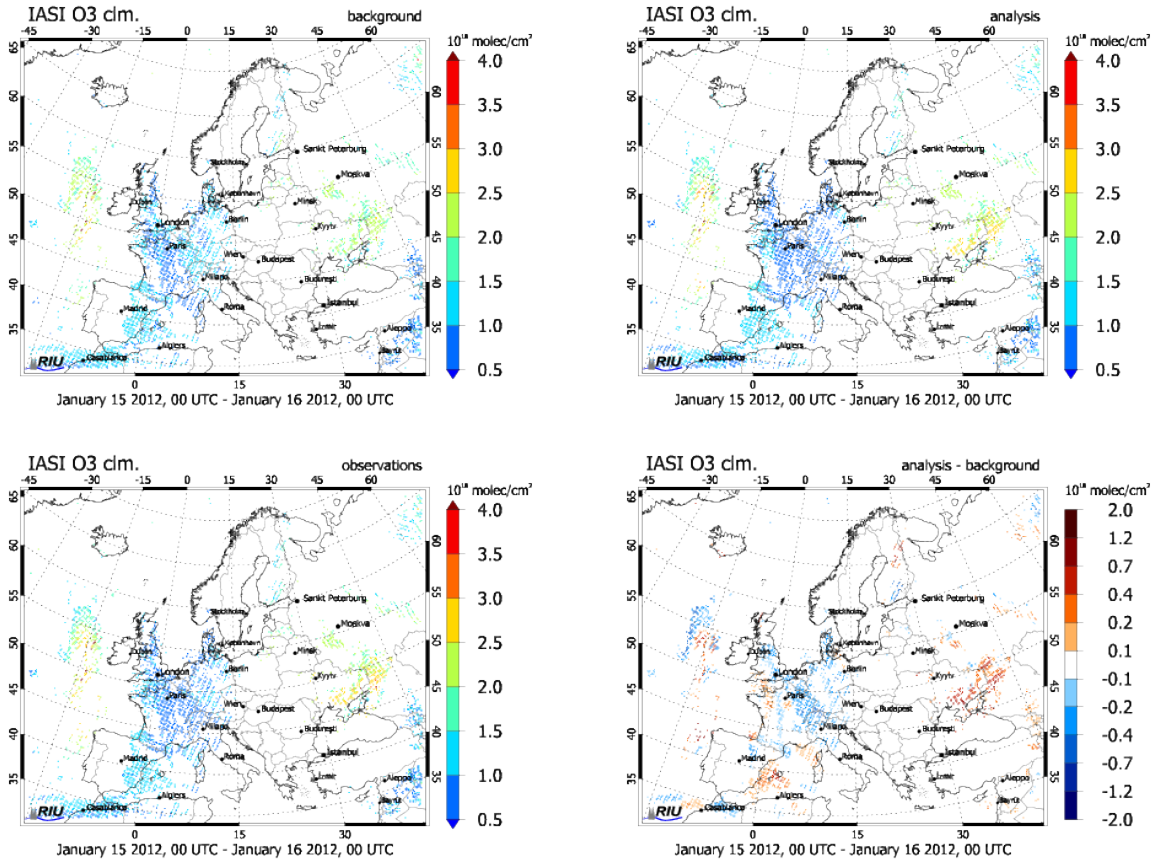


Figure 1.4: IASI partial Ozone columns above 800 mb for January 15, 2012 interpolated on the EURAD-IM grid with 15 km resolution. Upper left: background, upper right: analysis, lower left: observations, lower right: difference between analysis and background.

1.5 4d-var Emission Inversion

The data assimilation algorithm concerning optimisation of emission factors was extended compared to its implementation described by Elbern et al. (2007). Within this scope the adjoint code for the emission estimation has been adapted to the recently developed EURAD-IM online emission model. Applying the online emission model in the EURAD-IM makes the assimilation more accurate due to the detailed distribution of emissions in time and height per source category (SNAP code) and per grid box. The 4D-var formulation implements the estimation of a proper emission factor error covariance matrix $\boldsymbol{K}$, factorised as:

$$\boldsymbol{K} = \boldsymbol{\Gamma D}^{1/2}\boldsymbol{D}^{T/2}\boldsymbol{\Gamma}$$

where $\boldsymbol{\Gamma}$ is a diagonal matrix containing the standard deviations of the emission factors and $\boldsymbol{D}$ is the background error correlation matrix. The matrix $\boldsymbol{\Gamma}$ is based on the state of the art, whereas the matrix $\boldsymbol{D}$ has been expanded: At the 19 emitted species that were used to be, 5 more (coming from

biogenic sources) have been added in the EURAD-IM model. Thus a better and more accurate estimation of the correlation between these 24 anthropogenic and biogenic emitted species has been done. So the EURAD-IM model with 4D-var using the online emission model has been applied. Sample model runs for optimisation of the emission factors, using ground observation data of CO and SO_2, have been performed (See figures 1.5 and 1.6). The diffusion approach according to Weaver and Courtier (2001) was applied to the gradient of the cost function with respect to emission factors due to numerical reasons.

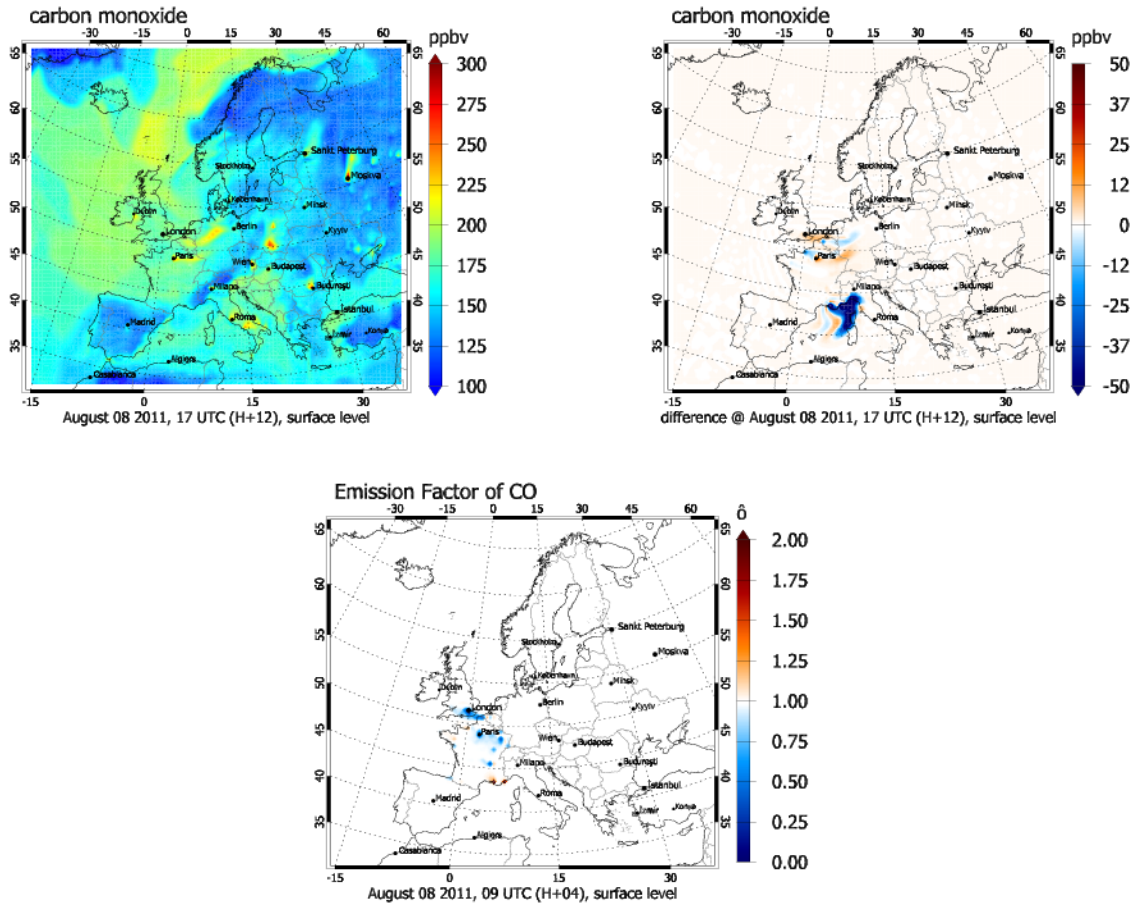


Figure 1.5: 4d-var optimisation of CO emission factors for August 8, 2011. Upper left: background, upper right: background minus analysis, lower panel: optimised CO emission factors

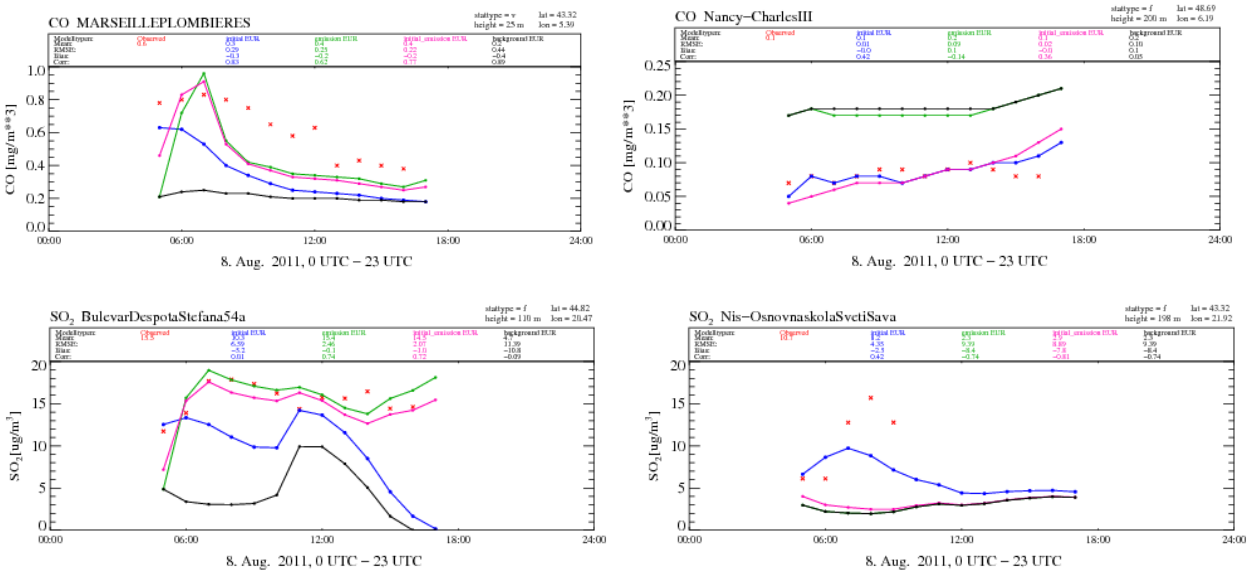


Figure 1.6: time-series of CO concentrations at two EEA measurement sites in France (above) and of SO_2 concentrations at two EEA measurement sites in Serbia(below). Black: background, green: 4d-var optimisation of emission factors, blue: 4d-var optimisation of initial values, pink: joined optimisation of emission factors and initial values, red: observations.

1.4 Wild fire data assimilation

A Wild fire data assimilation module based on the GFAS wild fire emission data (Kaiser et al., 2012) has been implemented in EURAD-IM and tested with the 2010 Russian peat fires. This is in preparation of the development of a NRT wild fire data assimilation module. Figure 1.6 shows PM_{10} time-series averaged over all available Finnish AIRBASE measurement sites during the 2010 Russian peat fires. On days with long-range transport of air pollutants originating from Russian peat fires, the strong negative bias of EURAD.IM results has been significantly reduced.

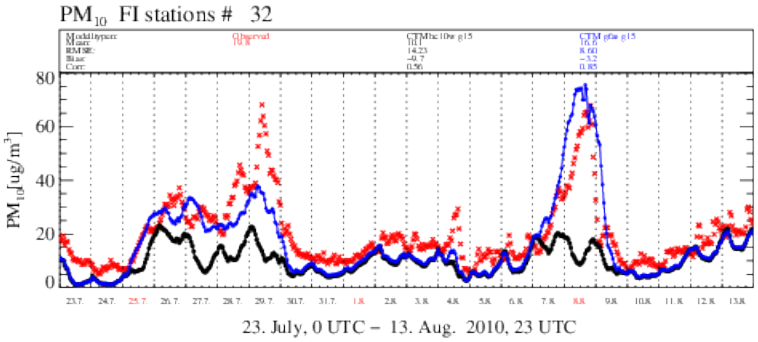


Figure 1.6: PM_{10} time-series averaged over all available Finnish measurement sites from July 23 to August 13, 2010. Black: EURAD-IM background, blue: predicted by EURAD-IM with GFAS wild fire emission data included, red: observations.

2. Ensemble Air-Quality Forecasts for Europe (ENS)

2.1 Air-quality forecasts

This task corresponds to the pre-operational provision of forecasts for key air-quality compounds with EURAD-IM, which is developed and constantly updated within MACC-II. Forecasts with the EURAD-IM system, up to 96h are provided for a range of molecules and vertical levels in GRIB2 format. Core products are forecasts of O_3, NO, NO_2, SO_2, CO, NH_3, NMVOC, total PAN, $PM_{2.5}$, and PM_{10} at the surface level and at 50, 250, 500, 1000, 2000, 3000, and 5000 m height. These core products are verified at the surface on a daily NRT basis. The number of provided air pollutants and vertical levels has been increased in response to the requirements of MACC-II users.

2.2 Air-quality analyses

This task corresponds to the prototype operational provision of analyses for key air-quality compounds (O_3, NO, NO_2, SO_2, CO, and PM_{10}) with the EURAD-IM model. Hourly analyses for the previous day are regularly delivered. At first, this is done once per day in the early morning with a reduced set of observations to provide improved initial values for the subsequent forecast. A second analysis of the previous day is set up in the early afternoon, which includes as many as possible observations from all available NRT instruments.

2.3 Model development

An important additional task of MACC-II sub-project ENS is the continued further development of the participating air quality models. In this context parameterisations already included in the EURAD-IM have been improved and new parameterisations were added.

2.3.1 Modelling of secondary organic aerosol

The Secondary Organic Aerosol Model (SORGAM) (Schell et al., 2001) has been further developed (Li et al., 2013). A temperature dependent parameterization of thermodynamic properties of the

secondary organic aerosol (SOA) was introduced and parameterisations of additional chemical and physical processes were included:

- Recent chamber experiments on the oxidation of Isoprene, Limonene and m-Xylene have been used to introduce a temperature dependent parameterisation of stoichiometric and partitioning coefficients.
- The formation of SOA due to Isoprene oxidation by O_3 and OH has been introduced.
- SOA formation pathways due to oxidation of biogenic precursors (α-Pinene, Limonene, and Isoprene) by the NO_3 radical were added.
- The influence of acidity on stoichiometric coefficients was taken into account.
- A humidity dependent parameterisation of the saturation concentration of organic condensable vapours was introduced.

Figure 2.1 shows a time-series of observed and modelled SOA concentrations for the IMPACT measurement campaign during May 2008. The strong under estimation of SOA by SORGAM without the extensions by Li et al. could be significantly reduced.

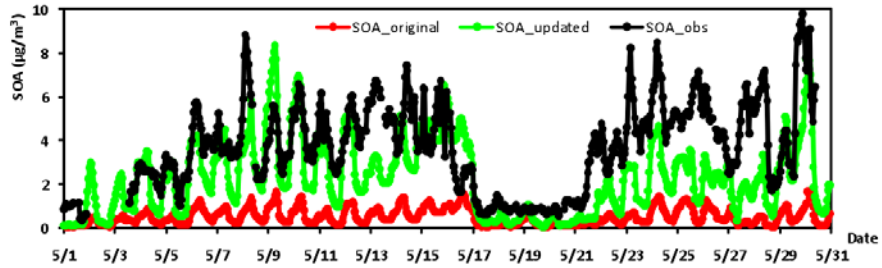


Figure 2.1: Time-series of SOA concentrations at Cabauw during the IMPACT measurement campaign in May 2008. Red: original SORGAM module, green: extended SORGAM module, black: observations.

2.3.2 Biogenic emissions

MEGAN 2.1 (Guenther et al., 2012) has been implemented into EURAD-IM and was used for the calculation of biogenic emissions in a sensitivity study. Figure 2.2 shows time-series of observed and modeled concentrations of SOA from NO_3 oxidation at two Finnish measurement sites. Due to the application of MEGAN the over estimation of SOA from NO_3 formation in Northern Europe could be significantly reduced.

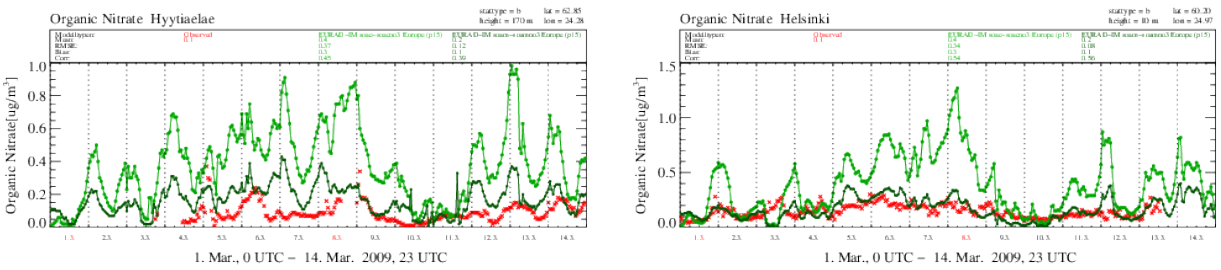


Figure 2.2: Time-series of SOA concentrations from NO_3 oxidation at Hyytiälä (left) and Helsinki (right) from 1st to 14th of March 2008. Red: observations, light green: EURAD-IM simulation with standard biogenic emissions, dark green: EURAD-IM simulation with biogenic emissions from MEGAN.

2.3.3 Wild fire emissions

EURAD-IM CTM has been extended to include GFASv1.0 wild fire emission data with 0.1° horizontal resolution. The implementation was tested on the Russian peat fires in summer 2010. High CO and PM_{10} concentrations observed at Finnish AIRBASE measurement sites were well

described by an EURAD-IM simulation with GFASv1.0 emissions included.

2.3.4 Birch pollen forecast and analysis

Biogenic particulate matter e.g. pollen grains contribute significantly to primary particulate matter mass and affect its size distribution. Due to their seasonal character, they affect also the temporal variation of primary PM in the atmosphere. The introduction of bio-aerosols as intrinsic components of particulate matter in Europe is expected to improve significantly the forecast of PM in the region. Also, it is recognized that it is necessary to consider exposure to both pollutants and bio-aerosols in health impact studies.

Traditionally, pollen forecasts have been based solely on local observations and do not consider transport of pollens from other regions, while measurements have shown evidences of transport of some bio-aerosols over thousands of kilometres. A modeling component of primary biogenic aerosol particles (PBAP) has been developed within MACC and MACC-II. A flexible emission subroutine has been developed as part of the SILAM model and has been coupled to ENS forecasting systems.

ENS modeling teams, in co-operation with national pollen monitoring networks of Austria, Germany, France, and Finland, members of European Aeroallergen Network, performed trial operational forecasting of birch pollen emission and dispersion over Europe in 2013. Prior to entering the forecasting mode, the re-analysis for 2010 was calculated to verify the implementation of the pollen module. Figure 2.3 shows mean pollen concentrations over Europe.

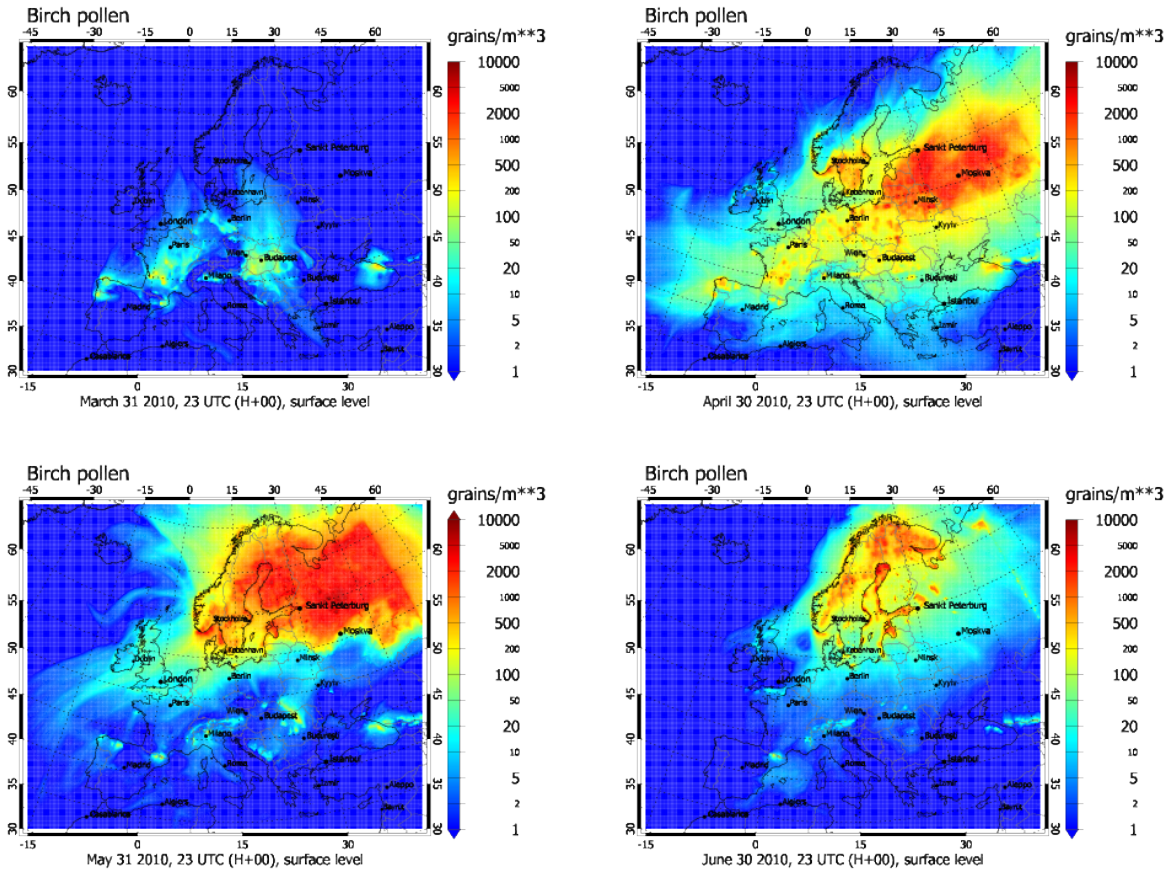


Figure 2.3: Mean pollen concentration in the near surface layer for March (upper left), April (upper right), Mai (lower left), and June (lower right) 2010 calculated by the EURAD-IM model.

The forecast was provided on a daily basis during the birch pollination season from 1.3 - 30.6.2013 with spatial resolution of 15km. The results were provided with hourly temporal resolution and a forecast horizon of 4 days. From aerobiological point of view, 2013 was a very late and extremely low year. In some parts of Europe, the season started two weeks later than usually. The seasonal integral of pollen concentrations was more than 10 times lower than long-term average and about

100 times lower than previous season of 2012.
Comparison with observations shows that start of the season 2013 is well captured by the models. The season ending caused more problems due to cold and humid weather, which impact seems to be over-estimated by the source term. The absolute concentrations were strongly over-estimated. Figure 2.4 shows time-series of birch pollen concentrations.

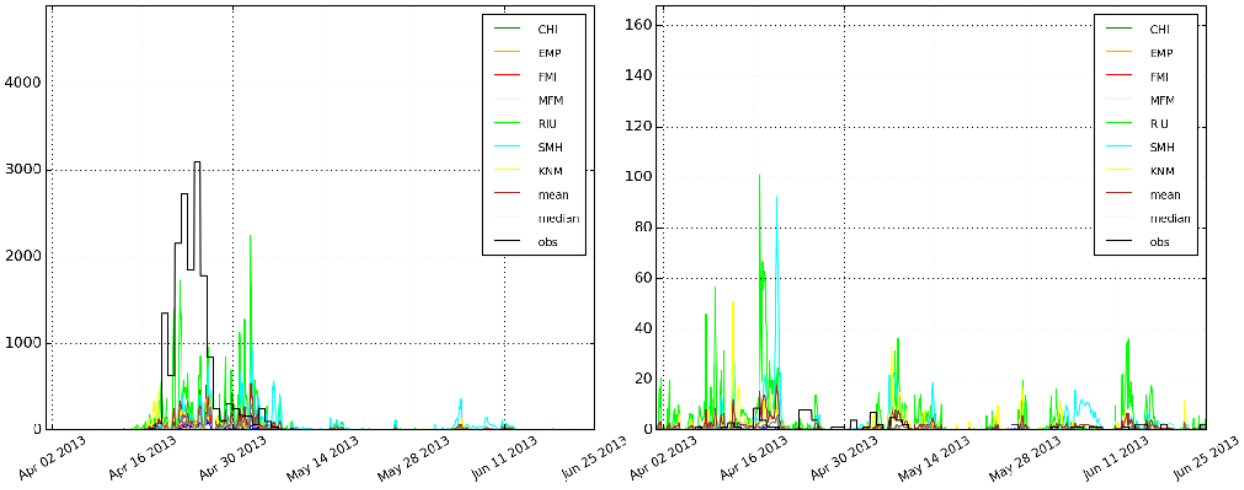
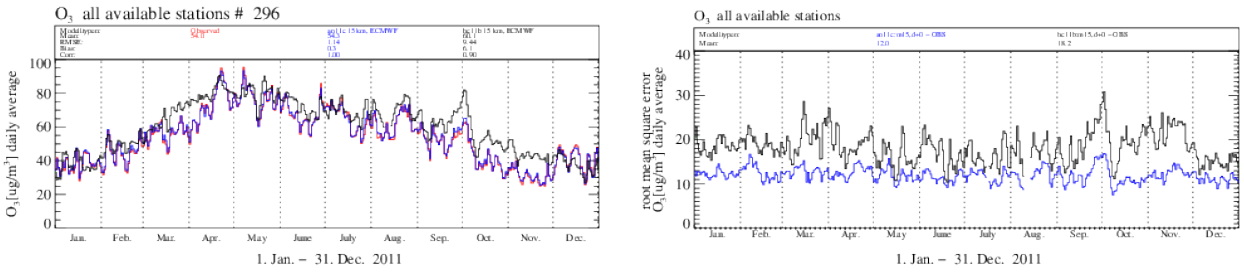


Figure 2.4: Time-series of birch pollen concentrations at Berlin (left) and Nice (right) during the pollen season 2013. Black: observations (European Aeroallergen Network), colored lines: results from individual models of the MACC-II ENS ensemble.

3. Validated assessment of air quality in Europe (EVA)

MACC-II sub-project EVA is focused on the evaluation of air quality in Europe, established on validated observational data, and also on operational air quality model results. The overall objective of this sub-project is an operational chain using models and data assimilation methodologies for simulating realistic air quality patterns in Europe. The final products of the sub-project are annual assessment reports, describing the state and the evolution of background concentrations of air pollutants in European countries. Special attention is given to pollutants characterized by the influence of long range transport, correctly caught by European scale modelling: O_3, NO_2, $PM_{2.5}$, and PM_{10}. An ensemble approach is used to provide the best possible representation of these pollutants. The EURAD-IM 3d-var assimilation system contributes to the ensemble air quality re-analyses for the years 2010-2013. Results from the re-analyses for 2011 are shown in Figure 3.1. In situ data from the Airbase measurement network and NO_2 column retrievals from OMI and GOME-2 are assimilated every hour using the intermittent 3d-var technique. About 30% of surface in situ background stations are held back from assimilation for validation purposes.



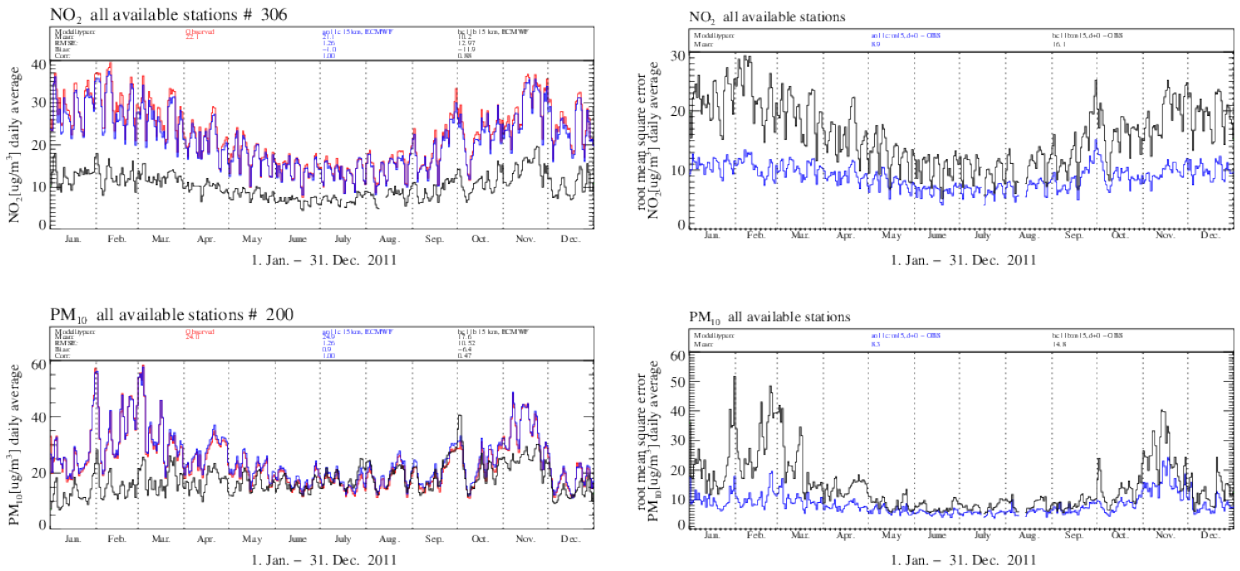


Figure 3.1: Daily averaged concentration (left) and its root mean square error (right) of O3 (above), NO2 (middle), and PM10 (below) averaged over all Airbase measurement sites, which were held back from assimilation for the year 2011. Red: observations, blue: EURAD-IM 3d-var re-analysis, 30% of stations held back from assimilation, black: control run (no data assimilation at all).

References

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

Clerbaux, C., A. Boynard, L. Clarisse, M. George, J. Hadji-Lazaro, H. Herbin, D. Hurtmans, M. Pommier, A. Razavi, S. Turquety, C. Wespes, P.-F. Coheur: Monitoring of atmospheric composition using the thermal infrared IASI/MetOp sounder, *Atmos. Chem. Phys.*, **9,** 6041-6054, 2009, doi: 10.5194/acp-9-6041-2009

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.

Nieradzik, L., Four-dimensional Variational Assimilation of Aerosol Data from In-situ and Remote Sensing Platforms, Ph.D. thessis, University of Cologne, Cologne, 2011.

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.

Schell, B., I.J. Ackermann, H. Hass, F.S. Binkowski, and A. Ebel, Modeling the formation of secondary organic aerosol within a comprehensive air quality model system, *J. Geophy. Res.*, **106**, D22, 28275-28293, 2001.

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

List of publications/reports from the project with complete references

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.

Goris, N. and H. Elbern, Singular vector decomposition for sensitivity analyses of troposheric chemical scenarios, Atmos. Chem. Phys.,**13**, 5063-5087, 2013, doi:10.5194/acp-13-5063-2013.

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.

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.

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.

Lahoz, W.A., V.-H. Peuch, J. Orphal, J.-L. Atti, K. Chance, X. Liu, D. Edwards, H. Elbern, J.-M. Flaud and M. Claeyman, Monitoring air quality from space: The case for the geostationary platform, Bull. Amer. Meteo. Soc., 93, 221-233, 2012, doi:10.1175/BAMS-D11-00045.1

Goris, N. and H. Elbern, Singular vector decomposition for sensitivity analyses of tropospheric chemical scenarios, doi: 10.5194/acpd-11-16745-2011, Atmos. Chem. Phys. Discuss., 11, 16745-16799, 2011.

Strunk, A., A. Ebel, H. Elbern, A nested application of four-dimensional variational assimilation of tropospheric chemical data. Int. J. Environment and Pollution, 46, pp. 43/60, 2011.

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.

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.

Future plans

MACC-III (Monitoring Atmospheric Composition and Climate – Phase 3) will function as the bridge between MACC-II and the Atmosphere Service envisaged to form part of Copernicus Operations and expected to start in 2015. MACC-III will sustain the current pre-operational atmosphere activities until March 2015 in order to avoid any interruption in the critical handover phase between the pre-operational and fully operational service.

RIUUK will again play an active role in the MACC-III sub-projects EDA, ENS, and EVA providing further developments of data assimilation algorithms, air quality forecasts and air quality analyses. Furthermore it is planned that RIUUK contributes to the proposed operational part of the Copernicus Atmosphere Service.