SPECIAL PROJECT FINAL REPORT

All the following mandatory information needs to be provided.

Project Title:	Global atmospheric chemistry modelling
Computer Project Account:	spdeacm
Start Year - End Year :	2012 - 2014
Principal Investigator(s)	O. Stein, M. G. Schultz
Affiliation/Address:	Forschungszentrum Jülich GmbH IEK-8: Troposphäre 52425 Jülich Germany
Other Researchers (Name/Affiliation):	S. Schröder, Forschungszentrum Jülich A. Heil, Forschungszentrum Jülich S. Rast, MPI Hamburg M. Decker, Forschungszentrum Jülich S. Waychal, Forschungszentrum Jülich

The following should cover the entire project duration.

Summary of project objectives

(10 lines max)

- Development of a chemistry module for IFS (CIFS-MOZ)
- Maintenance and improvements of the quasi-operational coupled MACC system MOZ-IFS
- Evaluation of the MOZ-IFS and CIFS-MOZ model for the troposphere and stratosphere
- Evaluation of MACC NRT forecasts and reanalysis
- investigate global budgets of trace gases in the atmosphere
- scientific model development of gas-phase chemistry in MOZART3, MOZ-IFS and CIFS-MOZ
- development and processing of global emission inventories

Summary of problems encountered

(If you encountered any problems of a more technical nature, please describe them here.)

Experience with the Special Project framework

(Please let us know about your experience with administrative aspects like the application procedure, progress reporting etc.)

No problems, flexible responses

Summary of results

(This section should comprise up to 10 pages and can be replaced by a short summary plus an existing scientific report on the project.)

See attached doc-file

List of publications/reports from the project with complete references

Peer-reviewed:

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Future plans

(Please let us know of any imminent plans regarding a continuation of this research activity, in particular if they are linked to another/new Special Project.)

A new special project spdeacm has been approved for the years 2015-2017, which can be seen as a continuation of the project described here. This project phase will cover our group activities in the EU project MACC-III and eventual evolvement in the upcoming CAMS project administered by ECMWF. Main aims will be the further development of CIFS, where our group is contributing continuously with the scientific updates, operation and evaluation of CIFS-MOZ. New directions in the modelling of gas-phase chemistry and coupling strategies to aerosol processes will be followed on to keep CIFS up to the state of the art.

Global modelling of atmospheric chemistry
special project SPDEACM 2012-2014 final report
June 2015

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Forschungszentrum Jülich

During the project phase 2012-2014 the special project had been closely related to the FZ Jülich engagement in the EU projects MACC (2009-2012), MACC-II (2012-2014) and MACC-III (2014-2015). Since 2012, the MACC project partners published 10 papers in peer-reviewed journals, which were directly relying on the model developments performed in spdeacm (plus three discussion papers and two submitted). These publications cover the description and validation of the MACC reanalysis for global reactive gases, the description and near-realtime evaluation of the MACC forecasts with the MOZ-IFS system, development of CIFS, the use of MACC boundary conditions for regional model studies, emission products, and global trace gas budgets. The major results will be highlighted in the following sections.

MACC reanalysis

The MACC system for reactive gases was developed during the years 2009-2014 and made use of the coupled system MOZART-IFS which was developed inside the spdeacm project (Stein et al., 2013). It has mainly been used for the quasi-operational global MACC forecasts from 2010 to September 2014 and for the MACC reanalysis (Stein et al., 2012).
The MACC reanalysis has been performed at ECMWF for the years 2003-2012 as a major output of the MACC project and is described in detail in Inness et al. (2013). Its main aim was to provide global estimates of atmospheric composition based on satellite observations and chemical transport modelling. This reanalysis provides fields of chemically reactive gases, namely carbon monoxide, ozone, nitrogen oxides, and formaldehyde, as well as aerosols and greenhouse gases globally at a horizontal resolution of about 80 km for both the troposphere and the stratosphere.
It could be shown that the gas-phase-species reproduce the observations relatively well, except for a general low bias in Northern Hemisphere carbon monoxide (Fig. 1) and an underestimation of wintertime NO2 maxima over anthropogenic pollution regions and an overestimation of NO2 in northern and southern Africa during the tropical biomass burning seasons. Spdeacm contributed largely to the MACC reanalysis by the development work for the coupled model MOZ-IFS, which was used as the MACC system for reactive gases. Furthermore we processed and applied the novel MACCity anthropogenic emission inventory (Granier et al., 2011) as well as the GFAS emission inventory for biomass burning (Kaiser et al., 2012) for use in the reanalysis system.
In addition to the evaluation of the MACC reanalysis presented in Inness et al. (2013) several aspects of the MACC reanalysis have been examined in more detail. Sheel et al. (2014) compared MACC reanalysis results for CO profiles over an urban site in India with MOZAIC aircraft profiles and other model calculations. They showed that mean biases with respect to the observed CO profiles were lower for the MACC reanalysis than for model simulations with MOZART and MRI-CCM2. The CO in the PBL region was consistently underestimated by MACC reanalysis during all the seasons, while MOZART and MRI-CCM2 show both positive and negative biases depending on the season.

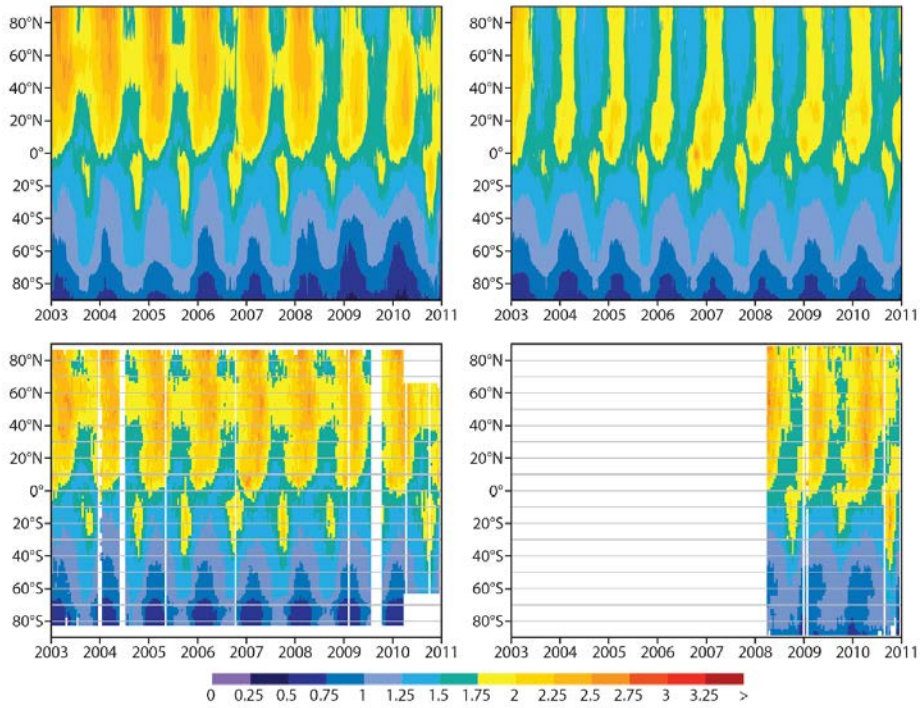

Figure 1: Time series of zonal mean total column CO [10^{18} molecules cm^{-2}] from the MACC reanalysis (top left), a control run without data assimilation (top right), MOPITT (bottom left) and IASI observations (bottom right).

Katragkou et al. (2015) evaluate the MACC reanalysis with respect to near surface ozone for specific European subregions. Measurements at rural locations from the European Monitoring and Evaluation Program (EMEP) and the European Air Quality Database (AirBase) are used for this evaluation assessment. The annual overall error of near surface ozone reanalysis is on average 24% over Europe, the highest found over Scandinavia (27%) and the lowest over the Mediterranean marine stations (21%). Near surface ozone shows mostly a negative bias in winter and a positive bias during warm months. Assimilation reduces the bias in near surface ozone and its impact is mostly notable in winter. With respect to the seasonal cycle, the MACC reanalysis reproduces the photochemically driven broad spring-summer maximum of surface ozone of central and south Europe. However, it does not capture adequately the early spring peak and the shape of the seasonality at northern and north-eastern Europe (Fig. 2). The diurnal range of surface ozone, which is an indication of the local photochemical production processes, is reproduced fairly well, with a tendency for a small overestimation during the warm months.

The performance of the MACC reanalysis and a control run without data assimilation in the extratropical upper troposphere/lower stratosphere (UTLS) over Europe is assessed in Gaudel et al. (2015) with MOZAIC/IAGOS in-flight data for ozone and CO. On average over the period, the reanalysis underestimates O3 by 60 ppbv in the lower stratosphere (LS), whilst CO is overestimated by 20 ppbv. In the upper troposphere (UT), O3 is overestimated by 50 ppbv, but CO is partly over or underestimated by up to 20 ppbv (Fig. 3). As expected, assimilation generally improves model results. Additionally, the observations clearly show a general negative trend of CO in the upper troposphere, which is rather well reproduced by the reanalysis. However, the reanalysis misses the full observed inter-annual variability in summer.

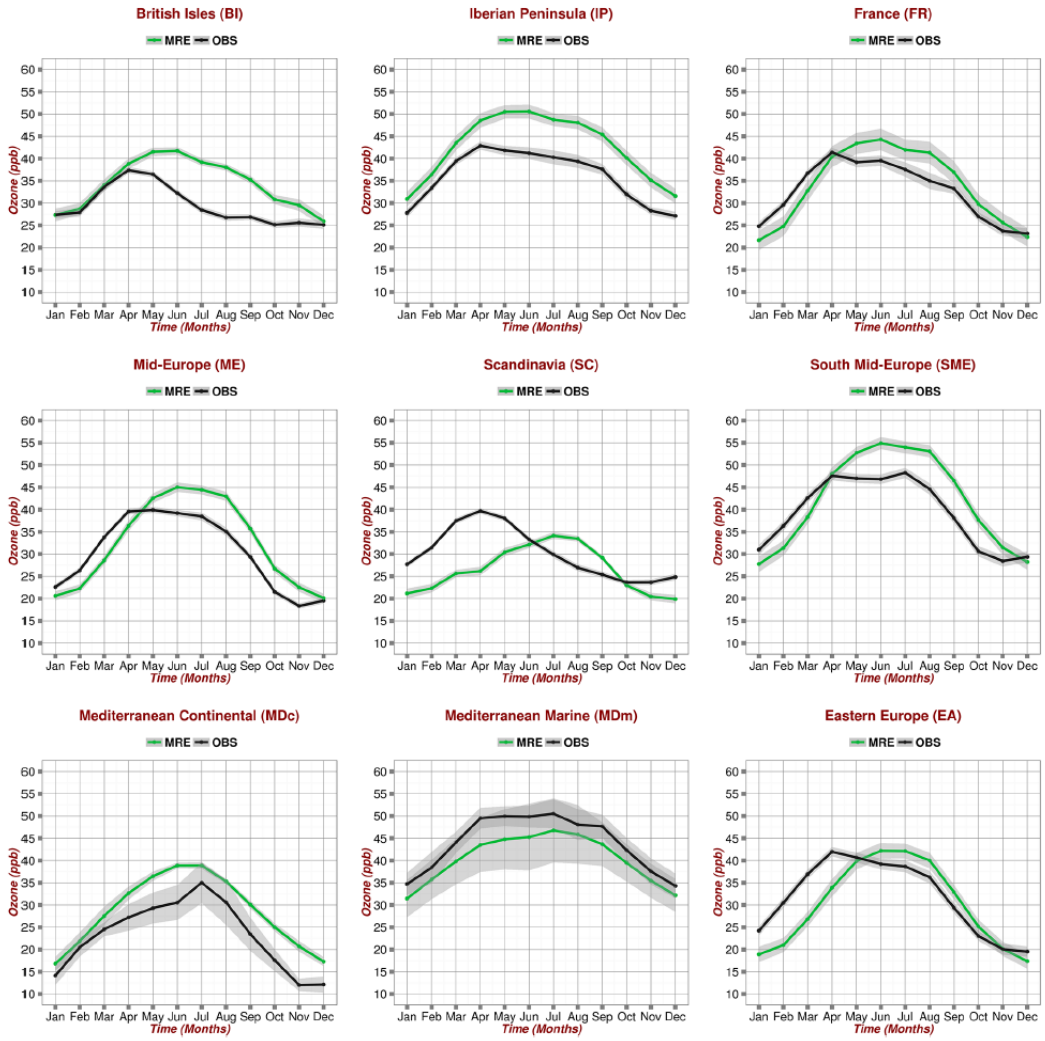

Figure 2: Mean 2003–2012 annual cycle of near surface ozone for the different European subregions of the MACC reanalysis and observations. The shading areas denote 95% confidence interval of the mean values.

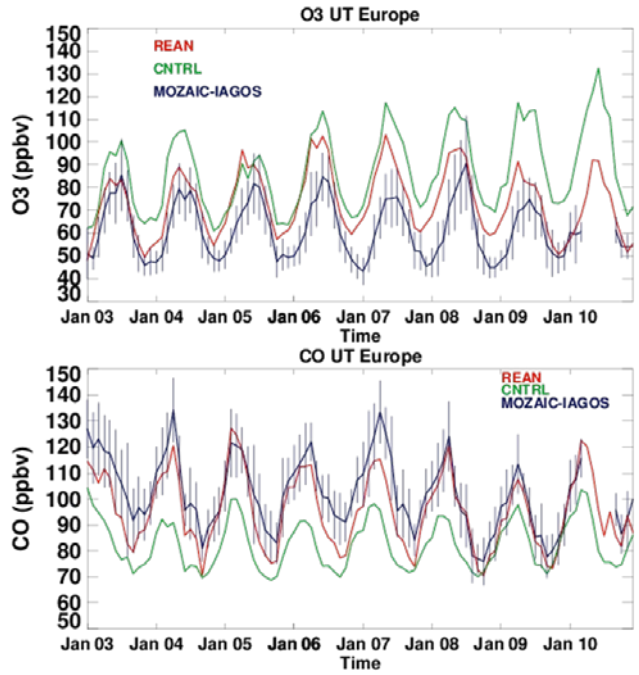

Figure 3: Time series of monthly mean ozone (top) and CO (bottom) in the UT for REAN (red), CNTRL (green) and MOZAIC-IAGOS observations (blue).

MACC quasi-operational forecast and analysis system

The MACC quasi-operational forecasts including data assimilation of chemical species are continuously evaluated in near-real-time from the VAL group in the MACC project. Besides the regular MACC VAL online reports (https://www.gmes-atmosphere.eu/services/aqac/global_verification/validation_reports/) three scientific papers summarize the validation efforts: Lefever et al. (2015) evaluate and discuss the quality of the stratospheric ozone analyses during the 3-year period between September 2009 and September 2012. The MOZART-IFS chemical data assimilation system is compared to the Belgian Assimilation System for Chemical ObsErvations (BASCOE), the Synoptic Analysis of Chemical Constituents by Advanced Data Assimilation (SACADA), and the Data Assimilation Model based on Transport Model version 3 (TM3DAM). The MACC system delivered total column values that agree well with ground-based observations (biases < 5%) and have a realistic seasonal cycle. Vertically alternating positive and negative biases are found in the MOZART-IFS analyses as well as an overestimation of 30 to 60% in the polar lower stratosphere during polar ozone depletion events (Fig. 4).

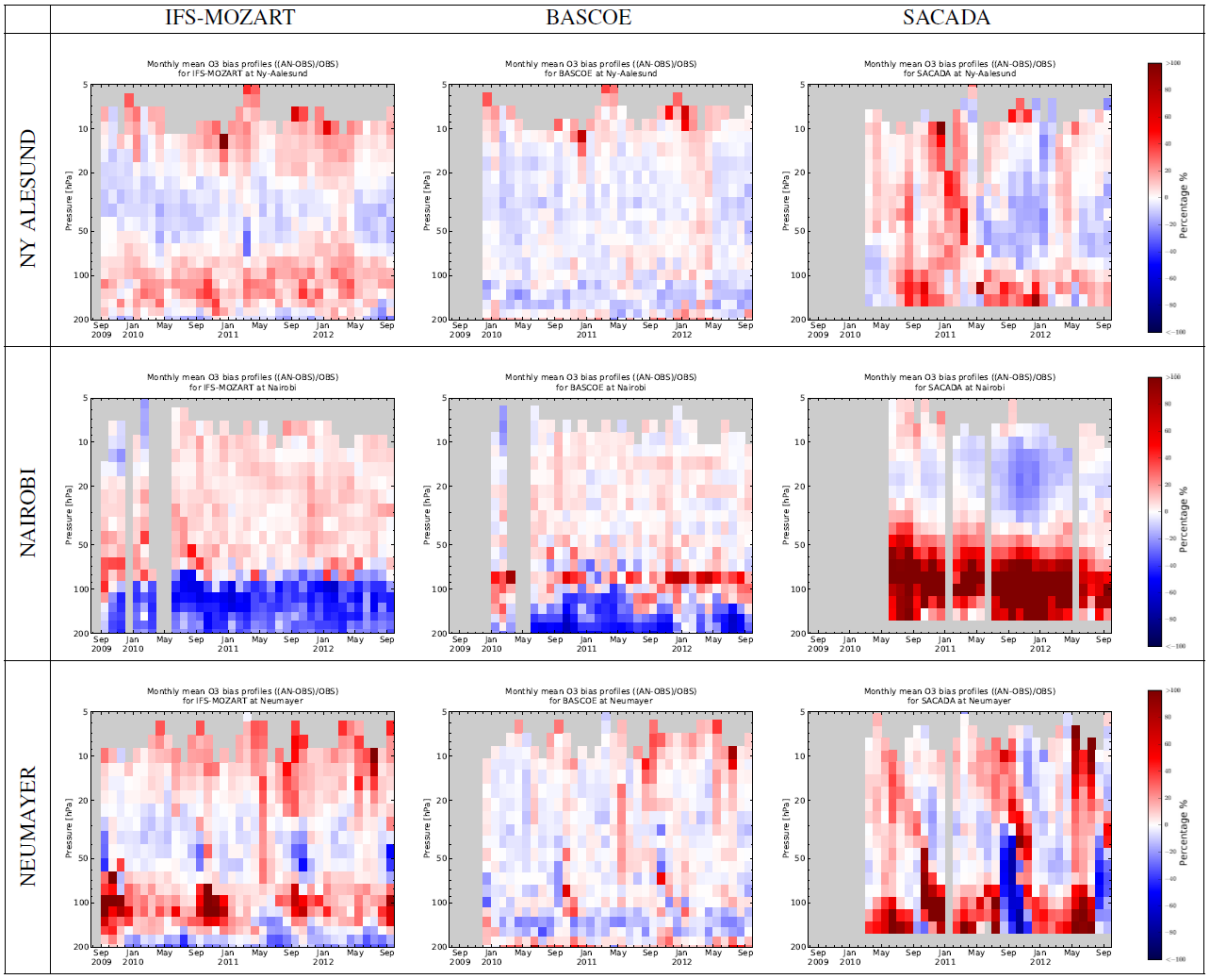

Figure 4: Time series of monthly mean ozone biases (analysis minus observations) with respect to ozone sondes at Ny-A° lesund, Nairobi and Neumayer in %. Left: IFS-MOZART, middle: BASCOE, right: SACADA.

Offline MOZART sensitivity tests were performed by spdeacm for spring 2011 which indicate that the differences between the forward models or the assimilation algorithms are much less important than the characteristics of the assimilated data sets. They also show that MOZART-IFS, despite some deficiencies in stratospheric chemistry and dynamics, is able to deliver realistic analyses of ozone both in the troposphere and in the stratosphere, but this

requires the assimilation of observations from nadir-looking instruments as well as the assimilation of profiles, which are well resolved vertically and extend into the lowermost stratosphere.

Reactive gases (O_3, CO, NO_2) in the troposphere from the MACC system are evaluated by Wagner et al. (2015). The validation was performed based on CO and O_3 surface observations from the Global Atmosphere Watch (GAW) network, O_3 surface observations from the European Monitoring and Evaluation Programme (EMEP), NO_2 tropospheric columns derived from the satellite sensors SCIAMACHY and GOME-2, and CO total columns derived from MOPITT. The MACC system proved capable of reproducing reactive gas concentrations in consistent quality, however, with a seasonally dependent bias compared to surface and satellite observations: for northern hemispheric surface O_3 mixing ratios, positive biases appear during the warm seasons and negative biases during the cold parts of the years, with monthly Modified Normalised Mean Biases (MNMBs) ranging between -30 and 30 % at the surface. Model biases are likely to result from difficulties in the simulation of vertical mixing at night and deficiencies in the model's dry deposition parameterization. Observed tropospheric columns of NO_2 and CO could be reproduced correctly during the warm seasons, but are mostly underestimated by the model during the cold seasons, when anthropogenic emissions are at a highest, especially over the US, Europe and Asia (Fig. 5).

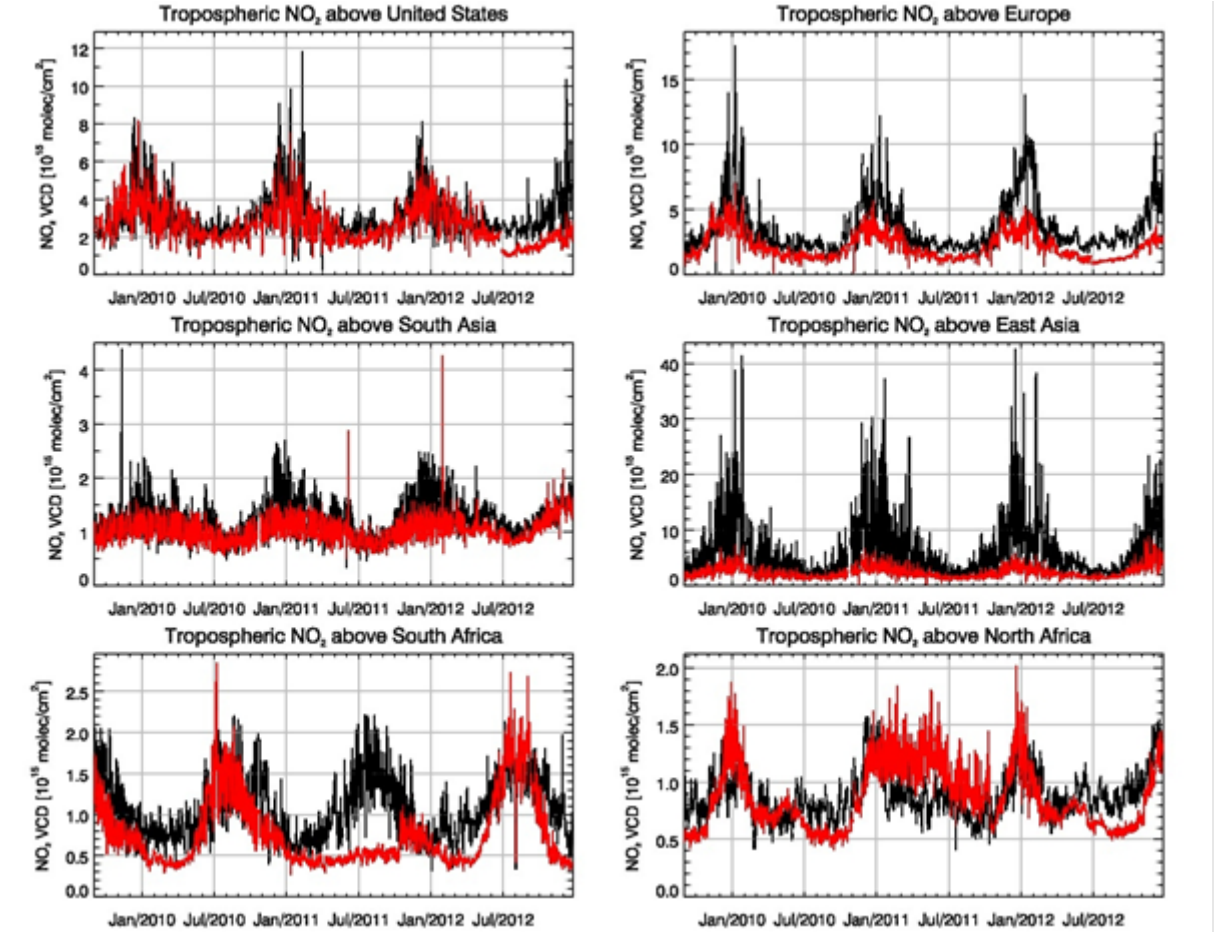

Figure 5: Time series of daily tropospheric NO_2 column densities [10^{15} molec cm^{-2}] averaged over different regions. Black lines show satellite observations (SCIAMACHY up to March 2012, GOME-2 from April 2012 onwards), red lines correspond to the MACC operational analysis simulations.

Monthly MNMBs of the satellite data evaluation range between -110 and 40 % for NO_2 and at most -20 % for CO over the investigated regions. The underestimation is likely to result from a combination of errors concerning the dry deposition parameterization and certain

limitations in the current emission inventories, together with an insufficiently established seasonality in the emissions.
The MACC validation process itself is presented in Eskes et al. (2015). They discuss the approach to validation that has been developed over the past three years. Topics discussed are the validation requirements, the operational aspects, the measurement data sets used, the structure of the validation reports, the models and assimilation systems validated, the procedure to introduce new upgrades, and the scoring methods. One specific target of the MACC system concerns forecasting special events with high pollution concentrations. Such events receive extra attention in the validation process. All of these aspects have been supported by model runs and analysis tools performed in spdeacm.

CIFS-MOZ

A major achievement of the spdeacm project in 2012-2014 was the establishment of a stable and reliable version of Composition-IFS with MOZART chemistry (CIFS-MOZ) in 2014. From the beginning of the MACC project, an approach was pursued to equip the IFS system with an integrated chemistry module (C-IFS). Major aims of this effort were: i) achieve a consistent setup for meteorology and chemistry processes, ii) to overcome the computational overhead induced by the coupler framework, iii) facilitate the use of data assimilation, archiving and postprocessing chains at ECMWF. A first CIFS version could be established for the quasi-operational MACC-III forecasts since October 2014. This version uses the CBO5 chemistry scheme adopted from the CTM TM5 (Flemming et al., 2015) but no explicit stratospheric chemistry scheme is included so far. Chemistry schemes from two other CTMs, MOCAGE and MOZART, have also been integrated into IFS, both of them including a detailed description of stratospheric chemistry. A detailed description of the gas-phase mechanism, photolysis rates and stratospheric heterogeneous reactions for CIFS-MOZ is given in Stein et al., 2015. There also the MOZ specific implementation of the chemistry interface in IFS is presented and the current status of the implementation is evaluated shortly. In many aspects the model shows results comparable to the other C-IFS realizations (with CB05/BASCOE or MOCAGE) as shown for surface ozone in Fig. 6, but some high biases for tropospheric ozone and poor vertical ozone distributions in the stratosphere are under consideration for model modification. In this regard development of C-IFS-MOZ is ongoing after the end of the MACC-III project.

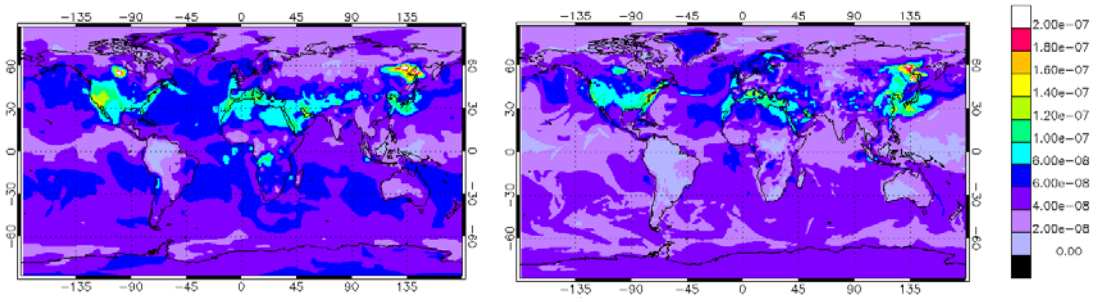

Figure 6: Ozone surface layer mass mixing ratios [kg kg^{-1}] at 20080701, 0UTC from a MOZART standalone simulation driven by ERA Interim and from C-IFS-MOZ (exp gasu)

For the integration of the MOZ chemistry into the IFS framework, major changes to the original MOZART CTM setup needed to be done. A chemical interface was developed and the MOZART code needed to be adapted to the CIFS coding standards. All output related code from the MOZART CTM needed to be removed as well as the physical parameterization routines for wet deposition, cloud processes and boundary fluxes, which were taken over by the IFS model and are described in Flemming et al. (2015). The transported species in C-IFS

are defined from a CTM specific species input table together with settings for emission, deposition or fixed boundary conditions. For emissions and deposition processes, new IFS interfaces common to the CTMs in use have been developed. The connections from MOZ to these interfaces also needed to be designed carefully. Spdeacm contributed also to other CIFS versions, namely CIFS-CBO5 based on the CTM TM5 (Huijnen et al., 2012; Flemming et al., 2015; Inness et al., 2015).
The northern hemisphere wintertime low bias of modelled carbon monoxide is examined and discussed in Stein et al. (2014) with a series of standalone MOZART simulations for the year 2008, which were partly performed at ECMWF under spdeacm. Such a bias has been observed by the MACC system (Inness et al., 2015; Wagner et al., 2015), even with CO satellite data assimilation, but also by most other state-of-the-art global models. The model bias can in principle originate from either an underestimation of CO sources or an overestimation of its sinks. The model sensitivity simulations are compared to observational data from ground-based stations, satellite observations, and vertical profiles from measurements on passenger aircraft. In the base case simulation using MACCity anthropogenic emissions, the near-surface CO mixing ratios are underestimated in the Northern Hemisphere by more than 20 ppb from December to April, with the largest bias of up to 75 ppb over Europe in January. An increase in global biomass burning or biogenic emissions of CO or volatile organic compounds (VOCs) was not able to reduce the annual course of the model bias and yields concentrations over the Southern Hemisphere which are too high. Raising global annual anthropogenic emissions with a simple scaling factor results in overestimations of surface mixing ratios in most regions all year round. Instead, our results indicate that anthropogenic CO and, possibly, VOC emissions in the MACCity inventory are too low for the industrialized countries only during winter and spring. Reasonable agreement with observations can only be achieved if the CO emissions are adjusted seasonally with regionally varying scaling factors. A part of the model bias could also be eliminated by exchanging the original resistance-type dry deposition scheme with a parameterization for CO uptake by oxidation from soil bacteria and microbes, which reduces the boreal winter dry deposition fluxes. The best match to surface observations, satellite retrievals, and aircraft observations was achieved when the modified dry deposition scheme was combined with increased wintertime road traffic emissions over Europe and North America (factors up to 4.5 and 2, respectively, see Fig. 7). One reason for the apparent underestimation of emissions may be an exaggerated downward trend in the Representative Concentration Pathway (RCP) 8.5 scenario in these regions between 2000 and 2010, as this scenario was used to extrapolate the MACCity emissions from their base year 2000. This factor is potentially amplified by a lack of knowledge about the seasonality of emissions. A methane lifetime of 9.7 yr for our basic model and 9.8 yr for the optimized simulation agrees well with current estimates of global OH, but we cannot fully exclude a potential effect from errors in the geographical and seasonal distribution of OH concentrations on the modelled CO.

Jülich OWS Interface JOIN

An important service from the global MACC component is the provision of chemical boundary conditions for regional air quality (RAQ) models. The Jülich OWS Interface (JOIN) provides interoperable web services for modeling and emission data sets allowing for easy download and visualization of multi-dimensional atmospheric composition and emission data via the internet (Waychal et al., 2013). MACC and other data are made available in the form of different catalogs either stored locally on the Jülich WCS server or accessed from other WCS servers from European and international partners. JOIN provides a user friendly interface for flexible selection of data sets delivered from WCS servers. The user can select a geographical region, time range and different variables from the selected dataset and then can

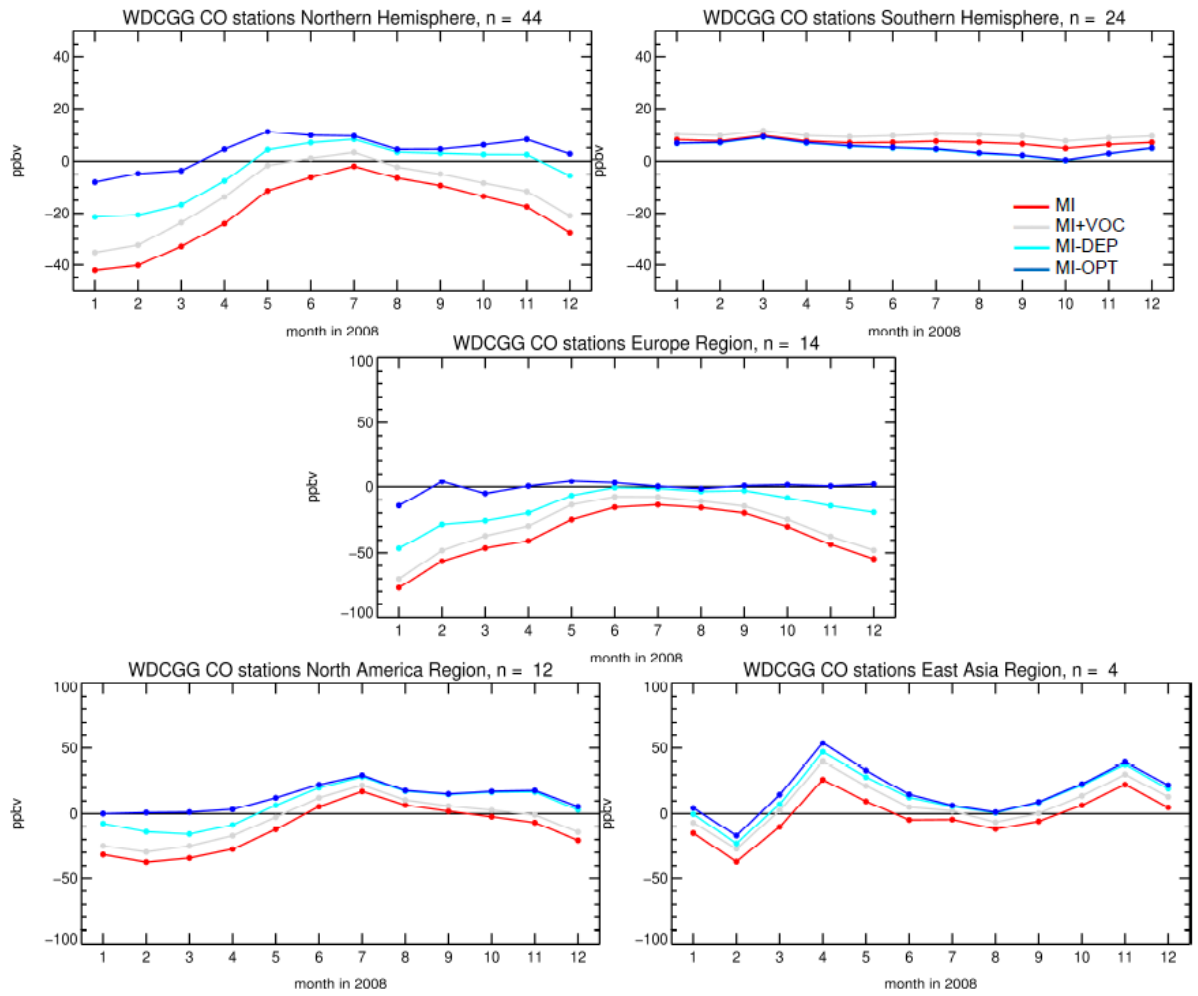

Figure 7: Bias of modelled 2008 monthly mean surface level mixing ratios from MOZART simulations MI (MACCity emissions), MI+VOC (doubling of anthropogenic VOC emissions), MI-DEP (alternative dry deposition), and MI-OPT (optimized emissions and dry deposition) compared to observations from WDCGG surface stations. n denotes the number of stations used for each region.

download or visualize the data in the form of maps, vertical cross sections or time series. A special feature is the comparison of model results with observational data in near real-time, which means that the data from the actual MACC forecasts stored at ECMWF is updated daily in an automated manner. JOIN uses standards like WCS, CF-netCDF and INSPIRE to test in a real-life environment. The JOIN server (http://join.iek.fz-juelich.de/) has been established in 2011 and has been continuously improved since then.

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