

1 **Evaluation of the MACC operational forecast system-**  
2 **potential and challenges of global near-real-time modelling**  
3 **with respect to reactive gases in the troposphere**

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#### 4 **Abstract**

5 Monitoring Atmospheric Composition and Climate (MACC) currently represents the  
6 European Union's Copernicus Atmosphere Monitoring Service (CAMS)  
7 (<http://www.copernicus.eu/>), which will become fully operational in the course of 2015. The  
8 global near-real-time MACC model production run for aerosol and reactive gases provides  
9 daily analyses and 5-day forecasts of atmospheric composition fields. It is the only  
10 assimilation system world-wide that is operational to produce global analyses and forecasts of  
11 reactive gases and aerosol fields. We have investigated the ability of the MACC analysis  
12 system to simulate tropospheric concentrations of reactive gases (CO, O<sub>3</sub>, and NO<sub>2</sub>) covering  
13 the period between 2009 and 2012. A validation was performed based on CO and O<sub>3</sub> surface  
14 observations from the Global Atmosphere Watch (GAW) network, O<sub>3</sub> surface observations  
15 from the European Monitoring and Evaluation Programme (EMEP) and furthermore, NO<sub>2</sub>  
16 tropospheric columns derived from the satellite sensors SCIAMACHY and GOME-2, and CO  
17 total columns derived from the satellite sensor MOPITT. The MACC system proved capable  
18 of reproducing reactive gas concentrations in consistent quality, however, with a seasonally  
19 dependent bias compared to surface and satellite observations: For northern hemisphere  
20 surface O<sub>3</sub> mixing ratios, positive biases appear during the warm seasons and negative biases  
21 during the cold parts of the years, with monthly Modified Normalised Mean Biases (MNMBs)  
22 ranging between -30% and 30% at the surface. Model biases are likely to result from  
23 difficulties in the simulation of vertical mixing at night and deficiencies in the model's dry  
24 deposition parameterization. Observed tropospheric columns of NO<sub>2</sub> and CO could be  
25 reproduced correctly during the warm seasons, but are mostly underestimated by the model  
26 during the cold seasons, when anthropogenic emissions are at a highest, especially over the  
27 US, Europe and Asia. Monthly MNMBs of the satellite data evaluation range between -110%  
28 and 40% for NO<sub>2</sub> and at most -20% for CO, over the investigated regions. The  
29 underestimation is likely to result from a combination of errors concerning the dry deposition  
30 parameterization and certain limitations in the current emission inventories, together with an  
31 insufficiently established seasonality in the emissions.

## 1 **1 Introduction**

2 The impact of reactive gases on climate, human health and environment has gained increasing  
3 public and scientific interest in the last decade (Bell et al., 2006, Cape 2008, Mohnen et al.,  
4 2013, Seinfeld and Pandis 2006, Selin et al., 2009). As air pollutants, carbon monoxide (CO),  
5 nitrogen oxides (NO<sub>x</sub>) and ozone (O<sub>3</sub>) are known to have acute and chronic effects on human  
6 health, ranging from minor upper respiratory irritation to chronic respiratory and heart  
7 disease, lung cancer, acute respiratory infections in children and chronic bronchitis in adults  
8 (Bell et al., 2006, Kampa and Castanas 2006). Tropospheric ozone, even in small  
9 concentrations, is also known to cause plant damage in reducing plant primary productivity  
10 and crop yields (e.g. Ashmore 2005). It is also contributing to global warming by direct and  
11 indirect radiative forcing (Forster et al., 2007, Sitch et al., 2007). Pollution events can be  
12 caused by local sources and processes but are also influenced by continental and  
13 intercontinental transport of air masses. Global models can provide the transport patterns of  
14 air masses and deliver the boundary conditions for regional models, facilitating the forecast  
15 and investigation of air pollutants.

16 The EU-funded research project MACC - Monitoring Atmospheric Composition and Climate,  
17 (consisting of a series of European projects, MACC to MACC-III), provides the preparatory  
18 work that will form the basis of the Copernicus Atmosphere Monitoring Service (CAMS).  
19 This service is established by the EU to provide a range of products of societal and  
20 environmental value with the aim to help European governments respond to climate change  
21 and air quality problems. MACC provides reanalysis, monitoring products of atmospheric key  
22 constituents (e.g. Inness et al., 2013), as well as operational daily forecasting of greenhouse  
23 gases, aerosols and reactive gases (Benedetti et al., 2011, Stein et al., 2012) on a global and  
24 on European-scale level, and derived products such as solar radiation. An important aim of  
25 the MACC system is to describe the occurrence, magnitude and transport pathways of  
26 disruptive events, e.g., volcanoes (Flemming and Inness, 2013), major fires (Huijnen et al.,  
27 2012, Kaiser et al., 2012) and dust storms (Cuevas et al., 2015). The product catalogue can be  
28 found on the MACC website, <http://copernicus-atmosphere.eu>. For the generation of  
29 atmospheric products, state-of-the-art atmospheric modelling is combined with assimilated  
30 satellite data (Hollingsworth et al., 2008, Inness et al., 2013, 2015, more general information  
31 about data assimilation can be found in e.g. Ballabrera-Poy et al., 2009 or Kalnay 2003).  
32 Within the MACC project there is a dedicated validation activity to provide up-to-date

1 information on the quality of the reanalysis, daily analyses and forecasts. Validation reports  
2 are updated regularly and are available on the MACC websites.

3 The MACC global near-real-time (NRT) production model for reactive gases and aerosol has  
4 operated with data assimilation from September 2009 onwards, providing boundary  
5 conditions for the MACC regional air quality products (RAQ), and other downstream users.  
6 The model simulations also provide input for the stratospheric ozone analyses delivered in  
7 near-real-time by the MACC stratospheric ozone system (Lefever et al., 2014).

8 In this paper we describe the investigation of the potential and challenges of near-real-time  
9 modelling with the MACC analysis system between 2009 and 2012. We concentrate on this  
10 period because of the availability of validated independent observations (namely surface  
11 observations from the Global Atmosphere Watch Programme GAW, the European  
12 Monitoring and Evaluation Programme EMEP, as well as total column satellite data from the  
13 MOPITT, SCIAMACHY and GOME-2 sensors) that are used for comparison. In particular,  
14 we study the model's ability to reproduce the seasonality and absolute values of CO and NO<sub>2</sub>  
15 in the troposphere as well as O<sub>3</sub> and CO at the surface. The impact of changes in model  
16 version, data assimilation and emission inventories on the model performance is examined  
17 and discussed. The paper is structured in the following way: Section 2 contains a description  
18 of the model and the validation data sets as well as the applied validation metrics. Section 3  
19 presents the validation results for CO, NO<sub>2</sub> and O<sub>3</sub>. Section 4 provides the discussion and  
20 section 5 the conclusions of the paper.

## 21 **2 Data and Methods**

### 22 **2.1 The MACC model system in the 2009-2012 period**

23 The MACC global products for reactive gases consist of a reanalysis performed for the years  
24 2003-2012 (Inness et al., 2013) and the near-real-time analysis and forecast, largely based on  
25 the same assimilation and forecasting system, but targeting different user groups. The  
26 MOZART chemical transport model (CTM) is coupled to the Integrated Forecast System  
27 (IFS) of the European Centre for Medium-Range Weather forecast (ECMWF), which together  
28 represent the MOZART-IFS model system (Flemming et al., 2009 and Stein et al. 2012). An  
29 alternative analysis system has been set up based on the global CTM TM5 (Huijnen et al.,  
30 2010). Details of the MOZART version used in the MACC global products can be found in  
31 Kinnison et al., 2007 and Stein et al. (2011, 2012). In the simulation, the IFS and the

1 MOZART model run in parallel and exchange several two- and three-dimensional fields  
2 every model hour using the OASIS4 coupling software (Valcke and Redler 2006), thereby  
3 producing three-dimensional IFS fields for O<sub>3</sub>, CO, SO<sub>2</sub>, NO<sub>x</sub>, HCHO, sea salt aerosol, desert  
4 dust, black carbon, organic matter, and total aerosol. The IFS provides meteorological data to  
5 MOZART. Data assimilation and transport of the MACC species takes place in IFS, while the  
6 whole chemical reaction system is calculated in MOZART.

7 The MACC\_osuite (operational suite) is the global near-real-time MACC model production  
8 run for aerosol and reactive gases. Here, we have investigated only the MACC analysis. In  
9 contrast to the reanalysis, the MACC\_osuite is a near-real-time run, which implies that it is  
10 only run once in near-real-time and may thus contain inconsistencies in e.g. the assimilated  
11 data. The MACC\_osuite was based on the IFS cycle CY36R1 with IFS model resolution of  
12 approximately 100 km by 100 km at 60 levels (T159L60) from September 2009 until July  
13 2012. The gas-phase chemistry module in this cycle is based on MOZART-3 (Kinnison et al.,  
14 2007). The model has been upgraded, following updates of the ECMWF meteorological  
15 model and MACC-specific updates, i.e. in chemical data assimilation and with respect to the  
16 chemical model itself. Thus, from July 2012 onwards, the MACC\_osuite has run with a  
17 change of the meteorological model to a new IFS cycle (version CY37R3), with an IFS model  
18 resolution of approximately 80 km at 60 levels (T255L60) and an upgrade of the MOZART  
19 version 3.5 (Kinnison et al., 2007; Emmons et al., 2011, Stein et al. 2013). This includes,  
20 amongst others, updated velocity fields for the dry deposition of O<sub>3</sub> over ice, as described in  
21 Stein et al. (2013). A detailed documentation of system changes can be found at:

22 [http://www.copernicus-atmosphere.eu/oper\\_info/nrt\\_info\\_for\\_users/](http://www.copernicus-atmosphere.eu/oper_info/nrt_info_for_users/)

### 23 **2.1.1 Emission inventories and assimilated data sets**

24 In the MACC\_osuite, anthropogenic emissions are based on emissions out of the EU project  
25 RETRO merged with updated emissions for East Asia from the REAS inventory, (Schultz et  
26 al. 2007) in the following referred to as RETRO-REAS. The horizontal resolution is 0.5° in  
27 latitude and longitude and it contains a monthly temporal resolution. Biogenic emissions are  
28 taken from GEIA, fire emissions are based on a climatology derived from GFEDv2 (van der  
29 Werf et al., 2006) until April 2010, when fire emissions change to GFAS fire emissions  
30 (Kaiser et al., 2012). Between January 2011 and October 2011 there has been a fire emission

1 reading error in the model, where, instead of adjusting emissions to the appropriate month, the  
2 same set of emissions have been read throughout this period.

3 After the model upgrade to the new cycle version CY37R3, in July 2012, the emission  
4 inventories changed from the merged RETRO-REAS and GEIA inventories, used in the  
5 previous cycle, to the MACCcity anthropogenic and biogenic emissions (Granier et al., 2011)  
6 and (climatological) MEGAN-v2 (Guenther et al., 2006) emission inventories. Wintertime  
7 anthropogenic CO emissions are scaled up over Europe and North America (see Stein et al.,  
8 2014). Near-real-time fire emissions are taken from GFASv1.0 (Kaiser et al. 2012), for both  
9 gas-phase and aerosol.

10 In the MACC\_osuite, the initial conditions for some of the chemical species are provided by  
11 data assimilation of atmospheric composition observations from satellites (see Benedetti et  
12 al., 2009; Inness et al., 2009, 2013; Massart et al., 2014). Table 1 lists the assimilated data  
13 products. From September 2009 to June 2012, O<sub>3</sub> total columns of the MLS and SBUV-2  
14 instruments are assimilated, as well as OMI and SCIAMACHY total columns (the latter only  
15 until March 2012, when the European Space Agency lost contact with the ENVironmental  
16 SATellite ENVISAT). CO total columns are assimilated from the IASI sensor and aerosol  
17 total optical depth is assimilated from the MODIS instrument. After the model cycle update in  
18 July 2012, data assimilation also contains OMI tropospheric columns of NO<sub>2</sub> and SO<sub>2</sub>, as well  
19 as CO MOPITT total columns. The CO total columns retrieved by MOPITT and IASI  
20 instruments have a relatively similar seasonality, but there is a systematic difference with  
21 MOPITT CO being higher over most regions in the northern hemisphere, especially during  
22 winter and spring. George et al. (“An examination of the long-term CO records from  
23 MOPITT and IASI and comparison of retrieval methodology”, AMTD, 2015 submitted)  
24 investigated the differences between MOPITT and IASI, and showed the impact of a priori  
25 information on the retrieved measurements.

26 Table 1 and 2 summarize the data assimilation and setup of the MACC\_osuite.

## 27 **2.2 Validation data and methodology**

28 In this study, mainly the same evaluation data sets have been used as during the MACC near-  
29 real-time validation exercise. This implies some discontinuities in the evaluations, e.g. the  
30 substitution of SCIAMACHY data with GOME-2 data after the loss of the ENVISAT sensor  
31 or an exclusion of MOPITT satellite data after the start of its assimilation into the model. The

1 continuous process of updating and complementation of data sets in databases requires the  
2 selection and definition of an evaluation data set at some point. The comparatively small  
3 inconsistencies between our data sets are considered to have a negligible impact on the overall  
4 evaluation results.

### 5 **2.2.1 GAW Surface O<sub>3</sub> and CO Observations**

6 The Global Atmosphere Watch (GAW) programme of the World Meteorological  
7 Organization (WMO) has been established to provide reliable long-term observations of the  
8 chemical composition and physical properties of the atmosphere, which are relevant for  
9 understanding atmospheric chemistry and climate change (WMO, 2013). GAW tropospheric  
10 O<sub>3</sub> measurements are performed in a way to be suitable for the detection of long-term regional  
11 and global changes. Furthermore, the GAW measurement programme focuses on  
12 observations, which are regionally representative and should be free from influence of  
13 significant local pollution sources and suited for the validation of global chemistry climate  
14 models (WMO 2007). Detailed information on GAW and GAW related O<sub>3</sub> and CO  
15 measurements can be found in WMO (2010, 2013).

16 Hourly O<sub>3</sub> and CO data have been downloaded from the WMO/GAW World Data Centre for  
17 Greenhouse Gases (WDCGG) for the period between 09/2009 and 12/2012 (status of  
18 download: 07/2013). Our evaluation includes 29 stations with surface observations for CO  
19 and 50 stations with surface observations for O<sub>3</sub>. Table 3 lists the geographic coordinates and  
20 altitudes of the individual stations. Being a long-term data network, the data in the database is  
21 provided with a temporal delay of approximately 2 years. As the data in the database becomes  
22 sparse towards the end of the validation period, near-real-time observations, as used in the  
23 MACC-project for near-real-time validation, presented on the MACC website, have been  
24 included to complement the validation data sets. For the detection of long-term trends and  
25 year-to-year variability, the data quality objectives (DQOs) for CO in GAW measurements  
26 are set to a maximum uncertainty of  $\pm 2$  ppb and to  $\pm 5$  ppb for marine boundary layer sites  
27 and continental sites that are influenced by regional pollution and to  $\pm 1$  ppb for ozone  
28 (WMO, 2012, 2013).

29 For the evaluation with GAW station data, 6-hourly values (0, 6, 12, 18 UTC) of the analysis  
30 mode have been extracted from the model and are matched with hourly observational GAW  
31 station data. Model mixing ratios at the stations' location have been linearly interpolated from  
32 the model data in the horizontal. In the vertical, modelled gas mixing ratios have been

1 extracted at the model level, which is closest to the GAW stations' altitude. Validation scores  
2 (see section 2.3) have been calculated for each station between the 6-hourly model analysis  
3 data and the corresponding observational data for the entire period (09/2009- 12/2012) and as  
4 monthly averages.

## 5 **2.2.2 EMEP Surface O<sub>3</sub> Observations**

6 The European Monitoring and Evaluation Programme (EMEP) is a scientifically based and  
7 policy driven programme under the Convention on Long-Range Transboundary Air Pollution  
8 (CLRTAP) for international co-operation to solve transboundary air pollution problems.  
9 Measurements of air quality in Europe have been carried out under the EMEP since 1977.

10 A detailed description of the EMEP measurement programme can be found in Tørseth et al.  
11 (2012). The surface hourly ozone data between 09/2009 and 12/2012 have been downloaded  
12 from the EMEP data web-page (<http://www.nilu.no/projects/ccc/emepdata.html>). For the  
13 validation, only stations meeting the 75% availability threshold per day and per month are  
14 taken into account. The precision is close to 1.5 ppb for a 10s measurement. More  
15 information about the ozone data quality, calibration and maintenance procedures can be  
16 found in Aas et al. (2000).

17 For comparison with EMEP data, 3-hourly model values (0, 3, 6, 12, 15, 18, 21 UTC) of the  
18 analysis mode have been chosen, in order to be able to evaluate day and night time  
19 performance of the model separately. Gas mixing ratios have been extracted from the model  
20 and are matched with hourly observational surface ozone data at 124 EMEP stations in the  
21 same way as for the GAW station data. The EMEP surface ozone values and the interpolated  
22 surface modeled values are compared on a monthly basis for the latitude bands of 30°N –  
23 40°N (southern Europe), 40°N – 50°N (central Europe) and 50°N – 70°N (northern Europe).  
24 For the identification of differences in the MACC\_osuite performance between day and night  
25 time, the MACC\_osuite simulations and the EMEP observations for the three latitude bands  
26 have been additionally separated into day-time (12:00–15:00 Local Time LT) and night-time  
27 (00:00–03:00 LT) intervals.

## 28 **2.2.3 MOPITT CO total column retrievals**

29 The MOPITT (Measurement Of Pollution In The Troposphere) instrument is mounted on  
30 board the NASA EOS Terra satellite and provides CO distributions at the global scale (Deeter  
31 et al., 2004). MOPITT has a horizontal resolution of 22 km x 22 km and allows global

1 coverage within 3 days. The data used in this study corresponds to CO total columns from  
2 version 5 (V5) of the MOPITT thermal infrared (TIR) product level 3. This product is  
3 available via the following web server: <http://www2.acd.ucar.edu/mopitt/products>. Validation  
4 of the MOPITT V5 product against in-situ CO observations showed a mean bias of  $0.06 \times 10^{18}$   
5 molecules  $\text{cm}^{-2}$  (Deeter et al., 2013). Following the recommendation in the users' guide,  
6 ([www.acd.ucar.edu/mopitt/v5\\_users\\_guide\\_beta.pdf](http://www.acd.ucar.edu/mopitt/v5_users_guide_beta.pdf)), the MOPITT data were averaged by  
7 taking into account their relative errors provided by the Observation Quality Index (OQI).

8 Also, in order for better data quality we used only daytime CO data since retrieval sensitivity  
9 is greater for daytime rather than nighttime overpasses. A further description of the V5 data is  
10 presented in Deeter et al. (2013) and Worden et al. (2014).

11 For the validation, the model CO profiles (X) were transformed by applying the MOPITT  
12 averaging kernels (A) and the a priori CO profile ( $X_a$ ) according to the following equation  
13 (Rodgers, 2000) to derive the smoothed profiles  $X^*$  appropriate for comparison with  
14 MOPITT data:

$$15 \quad X^* = X_a + A(X - X_a)$$

16 Details on the method of calculation are referred to in Deeter et al. (2004) and Rodgers  
17 (2000). The averaging kernels indicate the sensitivity of the MOPITT measurement and  
18 retrieval system to the true CO profile, with the remainder of the information set by the a  
19 priori profile and retrieval constraints (Emmons, 2009; Deeter et al., 2010). The model CO  
20 total columns used in the comparison with MOPITT observations, have been calculated using  
21 the averaging kernel smoothed profiles  $X^*$  which have the same vertical resolution and a  
22 priori dependence as the MOPITT retrievals. For the evaluation, 8 regions are defined (see  
23 Fig. 1): Europe, Alaska, Siberia, North Africa, South Africa, South Asia, East Asia and the  
24 United States.

25 The model update in July 2012 includes an integration of MOPITT CO total columns in the  
26 model's data assimilation system. With this, the MOPITT validation data has lost its  
27 independency for the rest of the validation period and MOPITT validation data has thus only  
28 been used until June 2012 for validation purposes.

#### 29 **2.2.4 SCIAMACHY and GOME-2 NO<sub>2</sub> Satellite Observations**

30 The SCanning Imaging Absorption spectroMeter for Atmospheric CHartographyY  
31 (SCIAMACHY; Bovensmann et al., 1999) onboard the ENVISAT and the Global Ozone

1 Monitoring Experiment-2 (GOME-2; Callies et al., 2000) onboard the Meteorological  
2 Operational Satellite-A (MetOp-A) comprise UV-VIS and NIR sensors designed to provide  
3 global observations of atmospheric trace gases.

4 In this study, the tropospheric NO<sub>2</sub> column data set described in Hilboll et al. (2013a) has  
5 been used. In short, the measured radiances are analysed using Differential Optical  
6 Absorption Spectroscopy (DOAS), (Platt and Stutz, 2008) in the 425–450 nm wavelength  
7 window (Richter and Burrows, 2002). The influence of stratospheric NO<sub>2</sub> air masses has been  
8 accounted for using the algorithm detailed by Hilboll et al. (2013b), using stratospheric NO<sub>2</sub>  
9 fields from the B3dCTM model (Sinnhuber et al., 2003a; Sinnhuber et al., 2003b; Winkler et  
10 al., 2008). Tropospheric air mass factors have been calculated with the radiative transfer  
11 model SCIATRAN (Rozañov et al., 2005). Only measurements with FRESCO+ algorithm  
12 (Wang et al., 2008) cloud fractions of less than 20% are used.

13 Tropospheric NO<sub>2</sub> vertical column density (VCD) from the MACC\_osuite is compared to  
14 tropospheric NO<sub>2</sub> VCD from GOME-2 and SCIAMACHY. As the European Space Agency  
15 lost contact with ENVISAT in April 2012, GOME-2 data is used for model validation from 1  
16 April 2012 onwards, while SCIAMACHY data is used for the remaining time period  
17 (September 2009 to March 2012). Satellite observations are gridded to the horizontal model  
18 resolution, i.e. 1.875° for IFS cycle CY36R1 (09/2009 -06/2012) and 1.125° for cycle  
19 CY37R3 (07/2012- 12/2012).

20 A few processing steps are applied to the MACC\_osuite data to account for differences to the  
21 satellite data such as observation time. Firstly, model data are vertically integrated to  
22 tropospheric NO<sub>2</sub> VCDs by applying National Centers for Environmental Prediction (NCEP)  
23 reanalysis (Kalnay et al., 1996) climatological tropopause pressure shown in Fig.1 of Santer et  
24 al. (2003). Secondly, simulations are interpolated linearly in time to the SCIAMACHY  
25 equator crossing time (roughly 10:00 LT). This most likely leads to some minor  
26 overestimation of model NO<sub>2</sub> VCDs compared to GOME-2 data, as the equator crossing time  
27 for GOME-2 is about 9:30 LT. Moreover, only model data for which corresponding satellite  
28 observations exist are considered. For the evaluation, the same regions have been used as for  
29 MOPITT (Fig.1), except for Siberia and Alaska. In contrast to MOPITT data, no averaging  
30 kernel is applied.

31 Satellite observations of tropospheric NO<sub>2</sub> columns have relatively large uncertainties, mainly  
32 linked to incomplete stratospheric correction (important over clean regions and at high

1 latitudes in winter and spring) and to uncertainties in air mass factors (mainly over polluted  
 2 regions) (e.g. Boersma et al., 2004 and Richter et al., 2005). The uncertainty varies with  
 3 geolocation and time but in first approximation can be separated into an absolute error of  
 4  $5 \times 10^{14}$  molec  $\text{cm}^{-2}$  and a relative error of about 30%, whichever is larger. As some of the  
 5 contributions to this uncertainty are systematic, averaging over longer time periods does not  
 6 reduce the errors as much as one would expect for random errors. Over polluted regions, the  
 7 uncertainty from random noise in the spectra is small in comparison to other error sources, in  
 8 particular for monthly averages.

### 9 **2.3 Validation metrics**

10 A comprehensive model evaluation requires the selection of validation metrics that provide  
 11 complementary aspects of model performance. The following metrics have been used in the  
 12 evaluation:

#### 13 **Modified Normalized Mean Bias MNMB**

$$14 \quad MNMB = \frac{2}{N} \sum_i \frac{f_i - o_i}{f_i + o_i} \quad (1)$$

#### 15 **Root Mean Square Error RMSE**

$$16 \quad RMSE = \sqrt{\frac{1}{N} \sum_i (f_i - o_i)^2} \quad (2)$$

#### 17 **Correlation Coefficient**

$$18 \quad R = \frac{\frac{1}{N} \sum_i (f_i - \bar{f})(o_i - \bar{o})}{\sigma_f \sigma_o} \quad (3)$$

19 where:  $N$  is the number of observations,  $f$  are the modelled analysis and  $o$  the observed  
 20 values,  $\bar{f}$  and  $\bar{o}$  are the mean values of the analysis and observed values and  $\sigma_f$  and  $\sigma_o$   
 21 are the corresponding standard deviations.

22 The validation metrics above have been chosen to provide complementary aspects of model  
 23 performance. The modified normalized mean bias is a normalization based on the mean of the  
 24 observed and forecast value (e.g. Elguindi et al. 2010). It ranges between  
 25 -2 and 2 and is very useful to check whether there is a negative or positive deviation between

1 model and observations. When multiplied by 100%, it can be interpreted as a percentage bias.  
2 The advantage of the MNMB is that it varies symmetrically with respect to under- and  
3 overestimation and is robust with respect to outliers. However, when calculated over longer  
4 time periods, a balance in model error, with model over- and underestimation compensating  
5 each other, can lead to a small MNMB for the overall period. For this reason, it is important  
6 to additionally consider an absolute measure, such as the RMSE. However, it has to be noted  
7 that the RMSE is strongly influenced by larger values and outliers, due to squaring. The  
8 correlation coefficient R can vary between 1 (perfect correlation) and -1 (negative correlation)  
9 and is an important measure to check the linearity between model and observations.

## 10 **3 Results**

### 11 **3.1 Evaluation of Ozone**

12 The evaluation of the MACC\_osuite run with O<sub>3</sub> from GAW surface observations (described  
13 in section 2.2.1) demonstrates good agreement in absolute values and seasonality for most  
14 regions. Figure 2 shows maps with Modified Normalized Mean Bias (MNMB, see section  
15 2.3) evaluations for 50 GAW stations globally (top) and in Europe (below). Figure 3 presents  
16 selected time series plots representing the results for high latitudes, low latitudes and Europe.  
17 Large negative MNMBs over the whole period 09/2009 to 12/2012 (-30 to -82%) are  
18 observed for stations located in Antarctica (Neumayer-NEU, South Pole-SPO, Syowa-SYO  
19 and Concordia- CON) whereby O<sub>3</sub> surface mixing ratios are strongly underestimated by the  
20 model. For stations located in high latitudes in the northern hemisphere (Barrow-BAR, Alaska  
21 and Summit-SUM, Denmark), the MACC\_osuite exhibits similar underestimated values of up  
22 to -35% for the whole evaluation period. The time series plots for Arctic and Antarctic  
23 stations (e.g. Summit-SUM, Neumayer-NEU and South Pole-SPO) in Fig. 3 show that an  
24 underestimation visible in these regions has been remedied and model performance improved  
25 with an updated dry deposition parameterization over ice, which has been introduced with the  
26 new model cycle in July 2012 (see section 2.1).

27 Large positive MNMBs (up to 50 to 70%, Fig. 2) are observed for stations that are located in  
28 or nearby cities and thus exposed to regional sources of contamination (Iskrba-ISK Slovenia,  
29 Tsukuba- TSU, Japan, Cairo-CAI, Egypt). In tropical and subtropical regions, O<sub>3</sub> surface  
30 mixing ratios are systematically overestimated (by about 20% on average) during the  
31 evaluation period. The time series plots for tropical and subtropical stations (e.g. for Ragged

1 Point-RAG, Barbados and Cape Verde Observatory, Cape Verde –CVO, Fig. 3) reveal a  
2 slight systematic positive offset throughout the year, however with high correlation  
3 coefficients (0.6 on average).

4 For GAW stations in Europe, the evaluation of the MACC\_osuite for the whole period shows  
5 MNMBs between -80 and 67%. Large biases appear only for 2 GAW stations located in  
6 Europe: Rigi- RIG, Switzerland (-80%), located near mountainous terrain and Iskrba- ISK,  
7 Slovenia (67%). For the rest of the stations MNMBs lie between 22 and -30%. Root Mean  
8 Square Errors (RMSEs, see section 2.3) range between 7 and 35 ppb (15 ppb on average).  
9 Again, results for Iskrba-ISK and Rigi-RIG show the largest errors. All other stations show  
10 RMSEs between 7 and 20 ppb. Correlation coefficients here range between 0.1 and 0.7 (with  
11 0.5 on average). Table 4 summarizes the results for all stations individually.

12 Monthly MNMBs (see Fig. 4) show a seasonally varying bias, with positive MNMBs  
13 occurring during the northern summer months (with global average ranging between 5 and  
14 29% during the months June and October), and negative MNMBs during the northern winter  
15 months (between -2 and -33% during the months December to March). These deviations  
16 partly cancel each other out in MNMB for the whole evaluation period. For the RMSEs, (Fig.  
17 5) maximum values also occur during the northern summer months with global average  
18 ranging between 11 and 16 ppb for June to September. The smallest errors appear during the  
19 northern hemisphere winter months (global average falling between 8 and 10 ppb for  
20 December and January). The correlation does not show a distinct seasonal behaviour (see Fig.  
21 6).

22 The time series plots in Fig. 3 show that the seasonal cycle of O<sub>3</sub> mixing ratios with maximum  
23 concentrations during the summer months and minimum values occurring during winter times  
24 for European stations (e.g. Monte Cimone-MCI, Italy, Kosetice-KOS, Czech Republic, and  
25 Kovk- KOV, Slovenia), could well be reproduced by the model, although there is some  
26 overestimation in summer resulting mostly from observed minimum concentrations that are  
27 not captured correctly by the MACC\_osuite, (Kosetice-KOS, Czech Republic, and Kovk-  
28 KOV, Slovenia).

29 The validation with EMEP surface ozone observations (described in section 2.2.2) in three  
30 different regions in Europe for the period 09/2009 to 12/2012 likewise confirms the behaviour  
31 of the model to overestimate O<sub>3</sub> mixing ratios during the warm period and underestimate O<sub>3</sub>  
32 concentrations during the cold period of the year (see Fig. 7). The positive bias (May-

1 November) is between -9 and 56% for northern Europe and Central Europe and between 8%  
2 and 48% for Southern Europe. Negative MNMBs appear, in accordance with GAW validation  
3 results, during the winter-spring period (December-April) ranging between -48 and -7% for  
4 EMEP stations in northern Europe (exception: December 2012 with 25%), between -1  
5 and -39% in central Europe (exception: December 2012 with 31%), whereas in southern  
6 Europe, deviations are smaller and remain mostly positive (between -8 and 9%) in winter  
7 (exception: December 2012 with 37%). The different behaviour for December 2012 likely  
8 results from the limited availability of observations towards the end of the validation period.  
9 The separate evaluation of day and night-time O<sub>3</sub> mixing ratios (Fig. 8) shows that for  
10 northern Europe larger biases appear during night time. For central Europe and southern  
11 Europe night-time biases are larger during cold periods (December-April), whereas during  
12 warm periods (May–November) larger biases appear during day time.

### 13 **3.2 Evaluation of Carbon Monoxide**

14 The evaluation of the MACC\_osuite with surface observations of 29 GAW stations (described  
15 in section 2.2.1) shows that over the whole period September 2009 to December 2012, CO  
16 mixing ratios could be reproduced with an average MNMB of -10%. The MNMBs for all  
17 stations range between -50 and +30%. Results are listed in Table 5, a selection of time series  
18 plots shows the results for stations in Europe, Asia and Canada in Fig. 9. MNMBs exceeding  
19  $\pm 30\%$  appear for stations that are either located in or nearby cities and thus exposed to  
20 regional sources of contamination (Kosetice- KOS, Czech Republic) or are located in or near  
21 complex mountainous terrain (Rigi-RIG, Switzerland, BEO Moussala- BEO, Bulgaria) which  
22 is not resolved by the topography of the global model. RMSEs fall between 12 and 143 ppb  
23 (on average 48 ppb) for all stations during the validation period, but for only four stations  
24 (Rigi-RIG, Kosetice- KOS, Payerne-PAY, Switzerland and BEO Moussala-BEO, all located  
25 in Europe) do the RMSEs exceed 70 ppb. Correlation coefficients from the comparison with  
26 GAW station data calculated over the whole time period range between 0 and 0.8 (on average  
27 0.4), with only four stations showing values smaller than 0.2 (Rigi-RIG, Moussala-BEO, East  
28 Trout Lake-ETL and Lac la Biche-LAC (the latter two located in Canada).

29 Considering the monthly MNMBs and RMSEs, it can be seen that during the northern  
30 hemisphere summer months, June to September, both are small (absolute differences less than  
31 5%), see Fig. 10 and Fig. 11. Negative MNMBs (up to -35%) and larger RMSEs (up to 72  
32 ppb) appear during the northern hemisphere winter months, November to March, when

1 anthropogenic emissions are at a highest, especially for the US, northern latitudes and Europe.  
2 Monthly correlation coefficients are between 0.1 and 0.5 and do not show a distinct seasonal  
3 behaviour (see Fig. 12), the low values of 0.1 during the period January 2011 to October 2011  
4 result from the reading error in the fire emissions (see section 2.1.1). The generally only  
5 moderate correlation coefficient is related to mismatches in the strong short-term variability  
6 seen in both the model and the measurements.

7 The time series plots for stations in Europe, Asia and Canada in Fig. 9 demonstrate that the  
8 annual CO cycle could to a large degree be reproduced correctly by the model with maximum  
9 values occurring during the winter period and minimum values appearing during the summer  
10 season. However, the model shows a negative offset during the winter period. Seasonal air  
11 mass transport patterns that lead to regular annual re-occurring CO variations could be  
12 reproduced for GAW stations in East Asia: The time series plots for Yonagunijima- YON and  
13 Minamitorishima- MNM station, Japan (Fig. 9) show that the drop of CO, associated with the  
14 air mass change from continental to cleaner marine air masses after the onset of the monsoon  
15 season during the early summer months, is captured by the MACC\_osuite. Deterioration in all  
16 scores is visible during December 2010 in the time series plots of several stations (e.g.  
17 Jungfraujoch-JFJ, and Sonnblick-SBL, Fig. 9). This is likely a result of changes in the  
18 processing of the L2 IASI data and a temporary blacklisting of IASI data (to avoid model  
19 failure) in the assimilation.

20 The comparison with MOPITT satellite CO total columns between October 2009 and June  
21 2012 (described in section 2.2.3) shows a good qualitative agreement of spatial patterns and  
22 seasonality, see Table 6. The MNMBs for 8 regions are listed in Fig. 13 and range between  
23 14% and -22%. The seasonality of the satellite observations is captured well by the  
24 MACC\_osuite over Asia and Africa, with MNMBs between -6% and 9% (North Africa),  
25 -12% and 8% (South Africa), -11% and 12% (East Asia), and -3% and 14% (South Asia). The  
26 largest negative MNMBs appear during the winter periods, especially from December 2010 to  
27 May 2011 and from September 2011 to April 2012, for Alaska and Siberia and for the US and  
28 Europe (MNMBs up to -22%), which coincides with large differences between MOPITT and  
29 IASI satellite data (see Fig. 14). On the global scale the average difference between the IASI  
30 and MOPITT total columns is less than 10% (George et al., 2009), and there is a close  
31 agreement of MOPITT and IASI for S. Asia and Africa (see Fig. 14). However, larger  
32 differences between MOPITT and IASI data appear during the northern winter months over

1 Alaska, Siberia, Europe and the US, which result in lower CO concentrations in the model,  
2 due to the assimilation of IASI CO data in the MACC\_osuite. The differences between  
3 MOPITT and IASI data can be mainly explained by the use of different a priori assumptions  
4 in the IASI and MOPITT retrieval algorithms (George et al., 2015 submitted). Indeed, the  
5 Fast Optimal Retrievals on Layers for IASI (FORLI) software (IASI) is using a single a priori  
6 CO profile (with an associated variance-covariance matrix) whereas the MOPITT retrieval  
7 algorithm is using a variable a priori, depending on time and location. George et al., 2015  
8 (submitted) show that differences above Europe and the US in January and December (for a 5  
9 year study) decrease by a factor of 2 when comparing IASI with a modified MOPITT product  
10 using the IASI single a priori. Between January 2011 and October 2011 there has also been a  
11 reading error in the fire emissions that contributes to larger MNMBs during this period (see  
12 section 2.1.1).

### 13 **3.3 Evaluation of Tropospheric Nitrogen Dioxide**

14 Figure 15 shows global maps of daily tropospheric NO<sub>2</sub> VCD averaged from September 2009  
15 to March 2012. Overall, spatial distribution and magnitude of tropospheric NO<sub>2</sub> observed by  
16 SCIAMACHY are well reproduced by the model. This indicates that emission patterns and  
17 NO<sub>x</sub> photochemistry are reasonably well represented by the model. However, the model  
18 underestimates tropospheric NO<sub>2</sub> VCDs over industrial areas in Europe, East China, Russia,  
19 and South East Africa compared to satellite data. This could imply that anthropogenic  
20 emissions from RETRO-REAS are underestimated in these regions, or that the lifetime in the  
21 model is too short. The model simulates larger NO<sub>2</sub> VCD maxima over Central Africa, which  
22 mainly originate from wild fires. It remains unclear if GFEDv2/GFAS fire emissions are too  
23 high here or if NO<sub>2</sub> fire plumes closer to the ground cannot be seen by the satellites due to  
24 light scattering by biomass burning aerosols (Leitao et al., 2010). In the northern hemisphere,  
25 background values of NO<sub>2</sub> VCD over the ocean are lower in the simulations than in the  
26 satellite data. The same is true for the South Atlantic Ocean to the west of Africa (see Fig.15).  
27 This might suggest a model underestimation of NO<sub>2</sub> export from continental sources or too  
28 rapid conversion of NO<sub>2</sub> into its reservoirs. However, as the NO<sub>2</sub> columns over the oceans are  
29 close to the uncertainties in the satellite data, care needs to be taken when interpreting these  
30 differences.

31 Time series of daily tropospheric NO<sub>2</sub> VCD averaged over different regions and  
32 corresponding monthly means are presented in Figs. 16 and 17, respectively. Time series of

1 the MNMB and RMSE are shown in Figs. 18 and 19, respectively. Table 7 summarizes the  
2 statistical values derived over the whole time period. High anthropogenic emissions occur  
3 over the United States, Europe, South Asia and East Asia compared to other regions on the  
4 globe (e.g., Richter et al., 2005). In principle, the MACC\_osuite catches the pattern of  
5 satellite NO<sub>2</sub> VCD over these regions. However, the model tends to underestimate NO<sub>2</sub> VCDs  
6 throughout the whole time period investigated here. The negative bias is most pronounced  
7 over East Asia with a modelled mean NO<sub>2</sub> VCD for September 2009 to December 2012 of  
8 about  $3.8 \times 10^{15}$  molec cm<sup>-2</sup> lower than that derived from satellite measurements (see Table  
9 7).

10 Considering monthly values, the MACC\_osuite strongly underestimates magnitude and  
11 seasonal variation of satellite NO<sub>2</sub> VCD over East Asia (MNMBs between about -40 % and  
12 -110 % and RMSE between  $1 \times 10^{15}$  molec cm<sup>-2</sup> and  $14 \times 10^{15}$  molec cm<sup>-2</sup> throughout the  
13 whole time period). A change in the modelled NO<sub>2</sub> values is apparent in July 2012 when the  
14 emission inventories changed and the agreement with the satellite data improved for South  
15 and East Asia but deteriorated for the US and Europe. This results in a drop of MNMBs (Fig.  
16 18) for Europe and the US with values approaching around -70% by the end of 2012.  
17 Nevertheless, correlations between daily satellite and model data derived for the whole time  
18 period (see Table 7) are high for East Asia (0.8), South Asia (0.8), Europe (0.8), and lower,  
19 but still rather high, for the US (0.6).

20 The North African and South African regions are strongly affected by biomass burning  
21 (Schreier et al., 2013). Magnitude and seasonality of daily and monthly tropospheric NO<sub>2</sub>  
22 VCDs (Figs. 16 and 17, respectively) are rather well represented by the model, apart from  
23 January 2011 to October 2011, due to difficulties in reading fire emissions for this time period  
24 (see section 2.1.1). The latter results in large absolute values of the MNMB (Fig. 18) and  
25 large RMSEs (Fig. 19) between January 2011 and October 2011 compared to the rest of the  
26 time period. As for other regions investigated in this section, mean values of simulated daily  
27 tropospheric NO<sub>2</sub> VCDs over North Africa and South Africa between September 2009 and  
28 December 2012 tend to be lower than the corresponding satellite mean values (see Table 7).  
29 The correlation between daily model and satellite data over the whole time period is about 0.6  
30 for South Africa and 0.5 for North Africa. It should be investigated in future studies, if this  
31 difference in model performance for the African regions is due to meteorology, chemistry or  
32 emissions.

## 1 **4 Discussion**

2 The validation of global O<sub>3</sub> mixing ratios with GAW observations at the surface levels  
3 showed that the MACC\_osuite could generally reproduce the observed annual cycle of ozone  
4 mixing ratios. Model validation with surface data shows global average monthly MNMBs  
5 between -30% and 30% (GAW) and for Europe between -50% and 60% (EMEP). The bias  
6 between measured O<sub>3</sub> surface mixing ratios and the MACC\_osuite is seasonally dependent,  
7 with an underestimation of the observed O<sub>3</sub> mixing ratios during the northern winter season  
8 and an overestimation during the summer months.

9 The validation of day-time versus night-time concentrations for Northern and Central Europe  
10 shows larger negative MNMBs in the winter months during night time than day time (Fig. 8),  
11 so that the negative bias in winter could be attributed to the simulation of vertical mixing at  
12 night, also described by Ordoñez (2010) and Schaap (2008), which remains a challenge in the  
13 model. The systematic underestimation of O<sub>3</sub> mixing ratios throughout the year for high  
14 latitude northern regions and Antarctica has its origin in an overestimation of the O<sub>3</sub> dry  
15 deposition velocities over ice. With the implementation of the new model cycle and  
16 MOZART model version, which includes updated velocity fields for the dry deposition of O<sub>3</sub>,  
17 as described in Stein et al. (2013), the negative offset in the MACC\_osuite model has been  
18 remedied for high latitude regions from July 2012 onwards (see the time series plots for the  
19 South Pole station- SPO and Neumayer- NEU in Fig. 3). The overestimation of O<sub>3</sub> mixing  
20 ratios for the northern hemisphere summer months is a well-known issue and has been  
21 described by various model validation studies (e.g., Brunner et al., 2003, Schaap et al., 2008,  
22 Ordoñez et al., 2010, Val Martin et al., 2014). Inadequate ozone precursor concentrations and  
23 aerosol induced radiative effects (photolysis) have been frequently identified as being the  
24 main factors. The time series plots in Fig. 3, however, demonstrate that the minimum  
25 concentrations in particular are not captured by the model during summer. Possible  
26 explanations include a general underestimation of NO titration which especially applies to  
27 stations with urban surroundings and strong sub-grid scale emissions (e.g. Tsukuba-TSU Fig.  
28 3), including difficulties by the global model to resolve NO titration in urban plumes. It also  
29 seems likely that dry deposition at wet surfaces in combination with the large surface sink  
30 gradient due to nocturnal stability cannot be resolved with the model's vertical resolution. In  
31 regions such as Central and Southern Europe (Fig. 8) where day time biases exceed night time  
32 biases, the overestimation of O<sub>3</sub> might be related to an underestimation of day-time dry

1 deposition velocities: Val Martin et al., (2014) describe a reduction of the summertime O<sub>3</sub>  
2 model bias for surface ozone after the implementation of adjustments in stomatal resistances  
3 in the MOZART model's dry deposition parameterization.

4 The MACC\_osuite model realistically reproduces CO total columns over most of the  
5 evaluated regions with monthly MNMBs falling between 10% and -20% (Table 6). There is a  
6 close agreement of modelled CO total columns and satellite observations for Africa and South  
7 Asia throughout the evaluation period. However, there is a negative offset compared to the  
8 observational CO data over Europe and North America. The largest deviations occur during  
9 the winter season when the observed CO concentrations are at a highest. The evaluation with  
10 GAW surface CO data accordingly shows a wintertime negative bias of up to -35% at the  
11 surface for stations in Europe and the US. A general underestimation of CO from global  
12 models in the northern hemisphere has been described by various authors (e.g., Shindell et al.,  
13 2006, Naik et al., 2013). According to Stein et al. (2014) this underestimation likely results  
14 from a combination of errors in the dry deposition parameterization and certain limitations in  
15 the current emission inventories. The latter include too low anthropogenic CO emissions from  
16 traffic or other combustion processes and missing anthropogenic VOC emissions in the  
17 inventories together with an insufficiently established seasonality in the emissions. An  
18 additional reason for the apparent underestimation of emissions in MACCcity may be an  
19 exaggerated downward trend in the RCP8.5 (Representative Concentration Pathways)  
20 scenario in North America and Europe between 2000 and 2010, as this scenario was used to  
21 extrapolate the MACCcity emissions from their bench mark year, i.e. 2000. For CO,  
22 uncertainties in the evaluation also include the retrieved amount of CO total columns between  
23 IASI and MOPITT. These vary with region, with IASI showing lower CO concentrations in  
24 several regions (Alaska, Siberia, Europe and the US) during the northern winter months,  
25 which possibly contribute to the deviations observed between the modelled data and MOPITT  
26 satellite data, as only IASI data has been assimilated in the model. The differences can  
27 primarily be explained by the use of different a priori assumptions in the IASI and MOPITT  
28 retrieval algorithms (George et al., 2015 submitted). On a global scale however, the average  
29 difference between the IASI and MOPITT total columns is less than 10% (George et al.,  
30 2009). From July 2012 onwards, MOPITT CO total columns are also assimilated in the  
31 MACC\_osuite.

1 Modelled NO<sub>2</sub> total columns agree well with satellite observations over the United States,  
2 South Asia and North Africa. However, there is also a negative offset for NO<sub>2</sub> over Europe  
3 and East Asia. Again, the largest deviations are occurring during the winter season. The  
4 quality of the emission inventory is even more crucial for short lived reactive species such as  
5 NO<sub>2</sub>, where model results depend to a large extent on emission inventories incorporated in the  
6 simulations. This is highlighted by the deterioration of agreement between model results and  
7 satellite data for the US in July 2012 when anthropogenic emissions were changed from  
8 RETRO-REAS to MACCity. This change led to an increasing negative bias in NO<sub>2</sub> over  
9 Europe and North America and to an improvement for South and East Asia (see Fig. 18). A  
10 deterioration in MNMBs associated with the fire emissions is visible between January 2011  
11 and October 2011 over regions with heavy fire activity (Africa and East Asia), and goes back  
12 to a temporary error in the model regarding the reading of fire emissions (see Figs. 17 and  
13 18). Particular challenges for an operational forecast system are regions with rapid changes in  
14 emissions such as China, where inventories need to be extrapolated to obtain reasonable  
15 trends. A large underestimation of NO<sub>2</sub> in China especially in winter has been reported for  
16 other CTMs in previous publications (He et al., 2007, Itahashi et al., 2014). The latter has  
17 been linked to an underestimation of NO<sub>x</sub> and VOC emissions, unresolved seasonality in the  
18 emissions and expected non-linearity of NO<sub>x</sub> chemistry. The change in validation data sets  
19 from SCIAMACHY to GOME-2 has shown to have negligible impact on the validation  
20 results and conclusions.

21

## 22 **5 Conclusion**

23 The MACC\_osuite is the global near-real-time MACC model analysis run for aerosol and  
24 reactive gases. The model has been evaluated with surface observations and satellite data  
25 concerning its ability to simulate reactive gases in the troposphere. Results showed that the  
26 model proved capable of a realistic reproduction of the observed annual cycle for CO and O<sub>3</sub>  
27 mixing ratios at the surface, however, with seasonally dependent biases. For ozone, these  
28 seasonal biases likely result from difficulties in the simulation of vertical mixing at night and  
29 deficiencies in the model's dry deposition parameterization. For CO, a negative offset in the  
30 model during the winter season is attributed to limitations in the emission inventories together  
31 with an insufficiently established seasonality in the emissions.

1 CO and NO<sub>2</sub> total columns derived from satellite sensors could be reproduced over most of  
2 the evaluated regions, but showed a negative offset compared to the observational data over  
3 Europe and North America (CO) and over Europe and East Asia (NO<sub>2</sub>). It has become clear,  
4 that that the emission inventories play a crucial role for the quality of model results and  
5 remain a challenge for near-real-time modeling, especially over regions with rapid changes in  
6 emissions. Inconsistencies in the assimilated satellite data and fire emissions showed only a  
7 temporary impact on the quality of model results.

8 The MACC NRT system is constantly evolving. A promising step in model development is  
9 the on-line integration of modules for atmospheric chemistry in the IFS, currently being tested  
10 for implementation in the MACC\_osuite. In contrast to the coupled model configuration as  
11 used in this paper, the on-line integration in the Composition IFS (C-IFS) provides major  
12 advantages; apart from an enhanced computational efficiency, C-IFS promises an  
13 optimization of the implementation of feedback processes between gas-phase/aerosol  
14 chemical processes and atmospheric composition and meteorology, which is expected to  
15 improve the modeling results for reactive gases. Additionally, C-IFS will be available in  
16 combination with different CTMs, (MOZART and TM5), which will help to explain whether  
17 deviations between model and observations go back to deficiencies in the chemistry scheme  
18 of a model.

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1 Table 1: List of assimilated data in the MACC\_osuite

<b>Instrument</b>	<b>Satellite</b>	<b>Provider</b>	<b>Version</b>	<b>Type</b>	<b>Status</b>
MLS	AURA	NASA	V02	O <sub>3</sub> Profiles	20090901 - 20121231
OMI	AURA	NASA	V883	O <sub>3</sub> Total column	20090901 - 20121231
SBUV-2	NOAA	NOAA	V8	O <sub>3</sub> 6 layer profiles	20090901 - 20121231
SCIAMACHY	Envisat	KNMI		O <sub>3</sub> total column	20090916 - 20120408
IASI	MetOp-A	LATMOS/ULB		CO Total column	20090901 - 20121231
MOPITT	TERRA	NCAR	V4	CO Total column	20120705 - 20121231
OMI	AURA	KNMI	DOMINO V2.0	NO <sub>2</sub> Tropospheric column	20120705 - 20121231
OMI	AURA	NASA	v003	SO <sub>2</sub> Tropospheric column	20120705 - 20121231
MODIS	AQUA / TERRA	NASA	Col. 5	Aerosol total optical depth	20090901 - 20121231

2

1

2 Table 2: Description of the set-up of the MACC\_osuite between 9/2009 and 12/2012. Details  
3 on the assimilated data are provided in Table 1. A description of the emissions is given in  
4 section 2.1.1 in the text.

<b>Model Cycle</b>	<b>CTM</b>	<b>Assimilated Data</b>	<b>Emissions</b>
CY36R1	MOZART v3.0	O <sub>3</sub> (MLS, OMI, SBUV-2 SCIAMACHY), CO (IASI)	RETRO / REAS / GEIA / GFEDv2/GFAS
CY37R3	MOZART v3.5	O <sub>3</sub> (MLS, OMI, SBUV-2), CO (IASI, MOPITT), NO <sub>2</sub> (OMI), SO <sub>2</sub> (OMI)	MACCity / MEGAN / GFASv1.0 daily

5

1 Table 3: List of GAW and EMEP stations used in the evaluation (GAW listed by label, EMEP  
 2 listed by region: Northern Europe NE, Central Europe CE and Southern Europe SE).

Station	Label/Region	Programme	Lat	Lon	Alt [m a.s.l.]	Station	Label/Region	Programme	Lat	Lon	Alt [m a.s.l.]
Ähtäri II	NE	EMEP	62.58	24.18	180	Masenberg	CE	EMEP	47.35	15.88	1170
Alert	ALT	GAW	82.45	-62.52	210	Mauna Loa	MAU	GAW	19.54	-155.58	3397
Arrival Heights	ARH	GAW	-77.80	166.67	184	Minamitorishima	MNM	GAW	24.29	153.98	8
Aspvreten	NE	EMEP	58.80	17.38	20	Montandon	CE	EMEP	47.30	6.83	836
Assekrem	ASS	GAW	23.27	5.63	2710	Monte Cimone	MCI	GAW	44.18	10.70	2165
Aston Hill	NE	EMEP	52.50	-3.03	370	Monte Velho	SE	EMEP	38.08	-8.80	43
Auchencorth	NE	EMEP	55.79	-3.24	260	Montelibretti	CE	EMEP	42.10	12.63	48
Ayia Marina	SE	EMEP	35.04	33.06	532	Montfranc	CE	EMEP	45.80	2.07	810
Barcarola	SE	EMEP	38.47	-6.92	393	Morvan	CE	EMEP	47.27	4.08	620
Baring Head	BAH	GAW	-41.41	174.87	85	Narberth	NE	EMEP	51.23	-4.70	160
Barrow	BAR	GAW	71.32	-156.60	11	Neuglobsow	NGW/NE	GAW/EMEP	53.17	13.03	62
BEO Moussala	BEO	GAW	42.18	23.59	2925	Neumayer	NEU	GAW	-70.65	-8.25	42
Birkenes	NE	EMEP	58.38	8.25	190	Niembro	CE	EMEP	43.44	-4.85	134
Bredkälen	NE	EMEP	63.85	15.33	404	Norra-Kvill	NE	EMEP	57.81	15.56	261
Bush	NE	EMEP	55.86	-3.21	180	O Saviñao	CE	EMEP	43.23	-7.70	506
Cabauw	NE	EMEP	51.97	4.92	60	Offagne	CE	EMEP	49.88	5.20	430
Cabo de Creus	CE	EMEP	42.32	3.32	23	Oulanka	NE	EMEP	66.32	29.40	310
Cairo	CAI	GAW	30.08	31.28	35	Pallas	NE	EMEP	68.00	24.15	340
Campisabalos	CE	EMEP	41.28	-3.14	1360	Payerne	PAY/CE	GAW/EMEP	46.81	6.94	510
Cape Grim	CAG	GAW	-40.68	144.68	94	Penausende	CE	EMEP	41.28	-5.86	985
Cape Point	CAP	GAW	-34.35	18.48	230	Peyrusse Vieille	CE	EMEP	43.62	0.18	200
Cape Verde	CVO	GAW	16.85	-24.87	10	Pic du Midi	PIC/CE	GAW/EMEP	42.94	0.14	2877
Charlton Mackrell	NE	EMEP	51.06	-2.68	54	Pillersdor	CE	EMEP	48.72	15.94	315
Chaumont	CE	EMEP	47.05	6.98	1130	Preila	NE	EMEP	55.35	21.07	5
Chibougamau	CHI	GAW	49.68	-74.34	393	Prestebakke	NE	EMEP	59.00	11.53	160
Chopok	CE	EMEP	48.93	19.58	2008	Puy de Dôme	PUY/CE	GAW/EMEP	45.77	2.95	1465
Concordia	CON	GAW	-75.10	123.33	3233	Ragged Point	RAG	GAW	13.17	-59.43	45
De Zilk	NE	EMEP	52.30	4.50	4	Rao	NE	EMEP	57.39	11.91	10
Diabla Gora	NE	EMEP	54.15	22.07	157	Revin	CE	EMEP	49.90	4.63	390
Dobele	DOB	GAW	56.37	23.19	42	Rigi	RIG/CE	GAW/EMEP	47.07	8.46	1030
Doñana	SE	EMEP	37.03	-6.33	5	Rojen Peak	CE	EMEP	41.70	24.74	1750
Donon	CE	EMEP	48.50	7.13	775	Rucava	RUC/NE	GAW/EMEP	56.10	21.10	18
Dunkelsteinerwald	CE	EMEP	48.37	15.55	320	Ryori	RYO	GAW	39.03	141.82	260
East Trout Lake	ETL	GAW	54.35	-104.98	492	Sable Island	SAB	GAW	43.93	-60.02	5
Egbert	EGB	GAW	44.23	-79.78	253	San Pablo de los Montes	SE	EMEP	39.55	-4.35	917
Eibergen	NE	EMEP	52.08	6.57	20	Sandve	NE	EMEP	59.20	5.20	15
Els Torms	CE	EMEP	41.40	0.72	470	Schauinsland	SCH/CE	GAW/EMEP	47.92	7.92	1205
Eskdalemuir	NE	EMEP	55.31	-3.20	243	Schmücke	NE	EMEP	50.65	10.77	937
Estrange	NE	EMEP	67.88	21.07	475	Sibton	NE	EMEP	52.29	1.46	46
Estevan Point	ESP	GAW	49.38	-126.55	39	Śnieżka	NE	EMEP	50.73	15.73	1603
Eupen	NE	EMEP	51.46	6.00	295	Sonnblick	SBL/CE	GAW/EMEP	47.05	12.96	3106

Everest - Pyramid	EVP	GAW	27.96	86.82	5079	South Pole	SPO	GAW	-89.98	-24.80	2810
Finokalia	SE	EMEP	35.32	25.67	250	Spitsbergen	NE	EMEP	78.90	11.88	474
Forsthof	CE	EMEP	48.10	15.91	581	St. Osyth	NE	EMEP	51.78	1.08	8
Fraserdale	FRA	GAW	49.88	-81.57	210	Stará Lesná	CE	EMEP	49.15	20.28	808
Gänserndorf	CE	EMEP	48.33	16.73	161	Starina	CE	EMEP	49.05	22.27	345
Gerlitz	CE	EMEP	46.69	13.92	1895	Stixneusiedl	CE	EMEP	48.05	16.68	240
Graz Platte	CE	EMEP	47.11	15.47	651	Strath Vaich Dam	NE	EMEP	57.73	-4.77	270
Great Dun Fell	NE	EMEP	54.68	-2.45	847	Summit	SUM	GAW	72.58	-38.48	3238
Grebenzen	CE	EMEP	47.04	14.33	1648	Svratouch	CE	EMEP	49.73	16.05	737
Grimsoe	NE	EMEP	59.73	15.47	132	Syowa Station	SYO	GAW	-69.00	39.58	16
Harwell	NE	EMEP	51.57	-1.32	137	Tänikon	CE	EMEP	47.48	8.90	540
Haunsberg	CE	EMEP	47.97	13.02	730	Topolniky	CE	EMEP	47.96	17.86	113
Heidenreichstein	CE	EMEP	48.88	15.05	570	Trinidad Head	TRI	GAW	41.05	-124.15	120
High Muffles	NE	EMEP	54.33	-0.80	267	Tsukuba	TSU	GAW	36.05	140.13	25
Hurdal	NE	EMEP	60.37	11.08	300	Tudor Hill	TUD	GAW	32.27	-64.87	30
Illmitz	CE	EMEP	47.77	16.77	117	Tustervatn	NE	EMEP	65.83	13.92	439
Iskrba	ISK/CE	GAW/EMEP	45.56	14.86	520	Tutuila	TUT	GAW	-14.24	-170.57	42
Izaña (Tenerife)	IZO	GAW	28.30	-16.50	2367	Ushuaia	USH	GAW	-54.85	-68.32	18
Jarczew	NE	EMEP	51.82	21.98	180	Utö	NE	EMEP	59.78	21.38	7
Jungfrauoch	JFJ/CE	GAW/EMEP	46.55	7.99	3578	Vavihill	NE	EMEP	56.01	13.15	175
Karasjok	NE	EMEP	69.47	25.22	333	Vezin	NE	EMEP	50.50	4.99	160
Keldsnor	NE	EMEP	54.73	10.73	10	Vilsandi	NE	EMEP	58.38	21.82	6
Kollumerwaard	KOW/NE	GAW/EMEP	53.33	6.28	1	Vindeln	VIN/NE	GAW/EMEP	64.25	19.77	225
Košetice	KOS/CE	GAW/EMEP	49.58	15.08	534	Virolahti II	NE	EMEP	60.53	27.69	4
Kovk	KOV/CE	GAW/EMEP	46.12	15.11	600	Vorhegg	CE	EMEP	46.68	12.97	1020
K-pusztá	CE	EMEP	46.97	19.58	125	Vredepeel	NE	EMEP	51.54	5.85	28
Krvavec	CE	EMEP	46.30	14.54	1740	Waldhof	WAL/NE	GAW/EMEP	52.80	10.77	74
La Coulonche	CE	EMEP	48.63	-0.45	309	Westerland	WES/NE	GAW/EMEP	54.93	8.32	12
La Tardière	CE	EMEP	46.65	-0.75	143	Weybourne	NE	EMEP	52.95	1.12	16
Lac La Biche	LAC	GAW	54.95	-112.45	540	Wicken Fen	NE	EMEP	52.30	-0.29	5
Ladybower Res.	NE	EMEP	53.40	-1.75	420	Yarner Wood	NE	EMEP	50.59	-3.71	119
Lahemaa	NE	EMEP	59.50	25.90	32	Yonagunijima	YON	GAW	24.47	123.02	30
Lauder	LAU	GAW	-45.03	169.67	370	Zarodnje	CE	EMEP	46.42	15.00	770
Le Casset	CE	EMEP	45.00	6.47	750	Zarra	SE	EMEP	39.09	-1.10	885
Leba	NE	EMEP	54.75	17.53	2	Zavodnje	ZAV	GAW	46.43	15.00	770
Lerwick	NE	EMEP	60.13	-1.18	85	Zillertaler Alpen	CE	EMEP	47.14	11.87	1970
Lille Valby	NE	EMEP	55.69	12.13	10	Zingst	ZIN/NE	GAW/EMEP	54.43	12.73	1
Lough Navar	NE	EMEP	54.44	-7.87	126	Zoebelboden	CE	EMEP	47.83	14.44	899
Lullington Heath	NE	EMEP	50.79	0.17	120	Zoseni	ZOS/NE	GAW/EMEP	57.13	25.90	188
Mace Head	NE	EMEP	53.17	-9.50	15	Zugspitze	SFH	GAW	47.42	10.98	2656
Market Harborough	NE	EMEP	52.55	-0.77	145						

1

2

1 Table 4: Modified normalized mean bias (MNMB) [%], correlation coefficient (R), and root  
 2 mean square error (RMSE) [ppb] derived from the evaluation of the MACC\_osuite with  
 3 Global Atmosphere Watch (GAW) O<sub>3</sub> surface observations during the period 09/2009 to  
 4 12/2012.

Station	ARH	ASS	BAH	BAR	BEO	CAI	CAG	CAP	CVO	CON	DOB	EVP	ISK	IZO	JFJ	KOW	KOS
<b>MNMB</b>	-39.8	-6.3	-8.6	-35.1	-21.4	70.1	-12.7	13.7	15.2	-81.6	6.3	18.4	67.2	10.4	1.9	5.8	-5.9
<b>R</b>	0.6	0.7	0.5	0.3	0.4	-0.1	0.4	0.6	0.6	0.3	0.3	0.7	0.1	0.5	0.7	0.6	0.6
<b>RMSE</b>	10.6	6.5	8.0	13.8	20.4	29.2	8.9	7.6	8.0	17.2	14.3	12.0	34.5	10.8	7.4	12.0	16.3

5

Station	KOV	KRV	LAU	MAU	MNM	MCI	NGW	NEU	PAY	PIC	PUY	RAG	RIG	RUC	RYO	SCH	SBL
<b>MNMB</b>	21.2	9.5	-5.5	13.7	38.6	2.3	-11.4	-45.2	-28.8	5.5	12.8	38.6	-80.3	-0.1	10.5	8.5	8.1
<b>R</b>	0.6	0.6	0.5	0.6	0.8	0.7	0.5	0.5	0.7	0.6	0.6	0.6	0.3	0.3	0.1	0.7	0.6
<b>RMSE</b>	19.5	11.1	9.0	11.5	13.0	8.2	14.3	11.4	15.6	7.7	10.6	10.6	28.4	15.0	14.4	12.2	9.3

6

Station	SFH	SPO	SUM	SYO	TRI	TSU	TUD	TUT	USH	VIN	WAL	WES	YON	ZAV	ZIN	ZOS
<b>MNMB</b>	10.1	-70.6	-24.4	-31.2	3.2	55.1	45.3	40.2	-7.0	4.6	-18.0	-12.3	22.0	19.7	-17.5	22.3
<b>R</b>	0.6	0.4	0.5	0.7	0.3	0.0	0.5	0.8	0.5	0.4	0.6	0.6	0.7	0.6	0.4	0.2
<b>RMSE</b>	9.3	16.3	11.7	8.9	13.3	27.6	18.2	8.0	7.6	11.2	13.6	11.6	13.6	18.6	13.9	17.0

7

1 Table 5: Modified normalized mean bias (MNMB) [%], correlation coefficient (R), and root  
 2 mean square error (RMSE) [ppb] derived from the evaluation of the MACC\_osuite with  
 3 Global Atmospheric Watch (GAW) CO surface observations during the period 09/2009 to  
 4 12/2012.

Station	ALT	BEO	CAP	CHI	CVO	EGB	ESP	ETL	FRA	IZO	JFJ	KOS	KOW	KRV	LAC	MCI	MNM
<b>MNMB</b>	-6.9	-36.1	29.7	-7.3	-0.6	4.5	-1.7	-19.9	-12.0	-6.8	-15.1	-50.1	-5.9	-30.4	-24.2	-19.0	6.4
<b>R</b>	0.5	0.0	0.6	0.4	0.7	0.3	0.5	0.1	0.3	0.7	0.6	0.2	0.4	0.4	0.0	0.6	0.8
<b>RMSE</b>	23.4	90.3	20.4	31.1	14.2	60.1	25.7	53.9	35.9	15.3	25.8	131.1	70.1	49.1	58.5	32.0	22.0

5

Station	NGW	PAY	PIC	PUY	RIG	RYO	SAB	SBL	SCH	SFH	USH	YON
<b>MNMB</b>	-1.7	-7.3	-9.3	-10.4	28.2	-4.8	-8.1	-25.1	-15.8	-25.7	-9.1	-1.6
<b>R</b>	0.4	0.3	0.7	0.6	0.0	0.4	0.4	0.5	0.5	0.4	0.6	0.7
<b>RMSE</b>	61.6	99.2	18.4	30.6	143.5	44.5	31.6	36.8	39.8	45.0	12.3	62.3

6

1 Table 6: Modified normalized mean bias (MNMB) [%] derived from CO satellite  
 2 observations (MOPITT) and the MACC\_osuite simulations of CO total columns from  
 3 10/2009 until 06/2012 averaged over different regions.

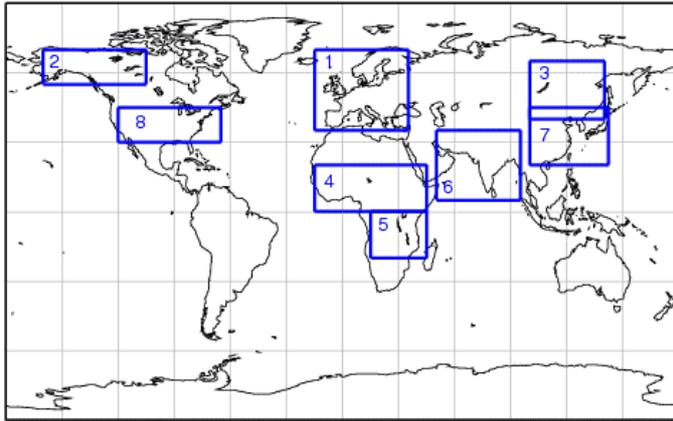
	<b>Oct 09</b>	<b>Nov 09</b>	<b>Dec 09</b>	<b>Jan 10</b>	<b>Feb 10</b>	<b>Mar 10</b>	<b>Apr 10</b>	<b>May 10</b>	<b>Jun 10</b>	<b>Jul 10</b>	<b>Aug 10</b>
<b>Europe</b>	4.17	1.35	-7.02	-7.17	-7.84	-8.56	-5.20	-2.15	-2.96	0.75	-2.88
<b>Alaska</b>	0.31	-3.16	-6.71	-8.85	-6.39	-3.13	-4.49	-3.85	-8.69	-6.18	-3.94
<b>Siberia</b>	2.02	1.62	-1.44	-2.75	-1.36	-2.27	-3.58	-2.93	-5.30	4.21	-8.43
<b>N. Africa</b>	6.53	9.17	5.82	7.05	3.45	-2.96	-3.53	-1.75	-3.40	-1.21	-3.58
<b>S. Africa</b>	-12.45	-9.44	3.10	6.53	8.27	6.63	3.57	2.33	7.34	0.57	-2.75
<b>S. Asia</b>	9.20	13.73	6.95	6.41	6.69	1.12	3.18	1.26	-3.01	1.98	2.15
<b>E. Asia</b>	8.04	12.33	-5.86	-9.18	-6.64	-4.49	-5.12	-5.61	-7.72	-4.34	-2.80
<b>US</b>	9.73	6.71	-5.42	-7.75	-10.88	-6.26	-3.80	-2.04	1.58	2.54	2.98
	<b>Sep 10</b>	<b>Oct 10</b>	<b>Nov 10</b>	<b>Dec 10</b>	<b>Jan 11</b>	<b>Feb 11</b>	<b>Mar 11</b>	<b>Apr 11</b>	<b>May 11</b>	<b>Jun 11</b>	<b>Jul 11</b>
<b>Europe</b>	-1.97	-0.92	-2.94	-7.78	-15.41	-17.22	-18.78	-17.34	-13.34	-6.62	-3.91
<b>Alaska</b>	-5.00	-1.89	-4.87	-7.51	-14.54	-9.90	-9.29	-12.54	-11.95	-10.04	-4.73
<b>Siberia</b>	-2.94	-1.93	-1.73	-3.02	-7.71	-7.78	-12.09	-21.99	-17.23	-11.59	-4.97
<b>N. Africa</b>	-1.22	3.33	5.98	7.03	-0.53	4.31	2.66	1.37	4.23	4.71	4.37
<b>S. Africa</b>	-5.13	2.84	7.39	4.37	1.41	3.39	3.80	0.99	5.71	3.45	-2.75
<b>S. Asia</b>	5.05	6.72	9.63	10.30	2.19	2.91	1.48	-1.76	1.68	1.62	2.90
<b>E. Asia</b>	6.13	6.93	2.44	3.23	-11.25	-9.18	-9.63	-8.58	-4.73	-1.62	5.00
<b>US</b>	0.08	-0.71	1.20	-8.06	-18.30	-16.98	-14.33	-13.52	-8.10	-4.72	-0.64
	<b>Aug 11</b>	<b>Sep 11</b>	<b>Oct 11</b>	<b>Nov 11</b>	<b>Dec 11</b>	<b>Jan 12</b>	<b>Feb 12</b>	<b>Mar 12</b>	<b>Apr 12</b>	<b>May 12</b>	<b>Jun 12</b>
<b>Europe</b>	-2.57	-7.28	-10.80	-11.85	-14.79	-13.50	-14.16	-15.30	-11.49	-7.00	-3.65
<b>Alaska</b>	-5.69	-11.86	-18.05	-14.33	-12.29	-11.50	-11.24	-11.92	-9.42	-8.71	-4.74
<b>Siberia</b>	-6.05	-15.16	-16.50	-10.32	-11.59	-10.15	-8.45	-13.14	-12.18	-11.08	-4.45
<b>N. Africa</b>	6.15	5.35	6.27	-0.93	3.37	2.04	1.11	-5.90	-3.40	-3.59	-0.95
<b>S. Africa</b>	-6.70	-4.43	-0.58	3.64	4.66	4.25	2.91	0.91	3.41	1.33	-1.23
<b>S. Asia</b>	3.80	2.27	4.24	4.76	7.00	3.24	1.72	-1.23	-0.90	0.49	-0.61
<b>E. Asia</b>	3.05	1.60	-2.60	-2.48	-5.15	-5.56	-4.63	-0.85	-0.36	-2.63	0.68
<b>US</b>	-1.17	-2.40	-4.23	-6.14	-10.84	-13.30	-14.87	-9.19	-6.94	-2.88	-2.55

1 Table 7: Statistics derived from satellite observations (SCIAMACHY from 09/2009 until  
 2 03/2012, GOME-2 from 04/2012 to 12/2012) and the MACC\_osuite simulations of daily  
 3 tropospheric NO<sub>2</sub> VCD [ $10^{15}$  molec cm<sup>-2</sup>] averaged over different regions for September 2009  
 4 to December 2012.

Region	United States	Europe	South Asia	East Asia	South Africa	North Africa
Model mean NO <sub>2</sub> VCD [ $10^{15}$ molec cm <sup>2</sup> ]	2.6	2.1	1.0	2.4	0.8	0.9
Satellite mean NO <sub>2</sub> VCD [ $10^{15}$ molec cm <sup>2</sup> ]	3.1	3.6	1.2	6.2	1.1	0.9
Modified normalized mean bias (MNMB) [%]	-17.3	-49.0	-13.4	-70.7	-36.8	-0.4
Root mean square error (RMSE) [ $10^{15}$ molec cm <sup>2</sup> ]	1.2	2.0	0.3	6.0	0.5	0.3
Correlation coefficient (R) [dimensionless]	0.6	0.8	0.8	0.8	0.6	0.5

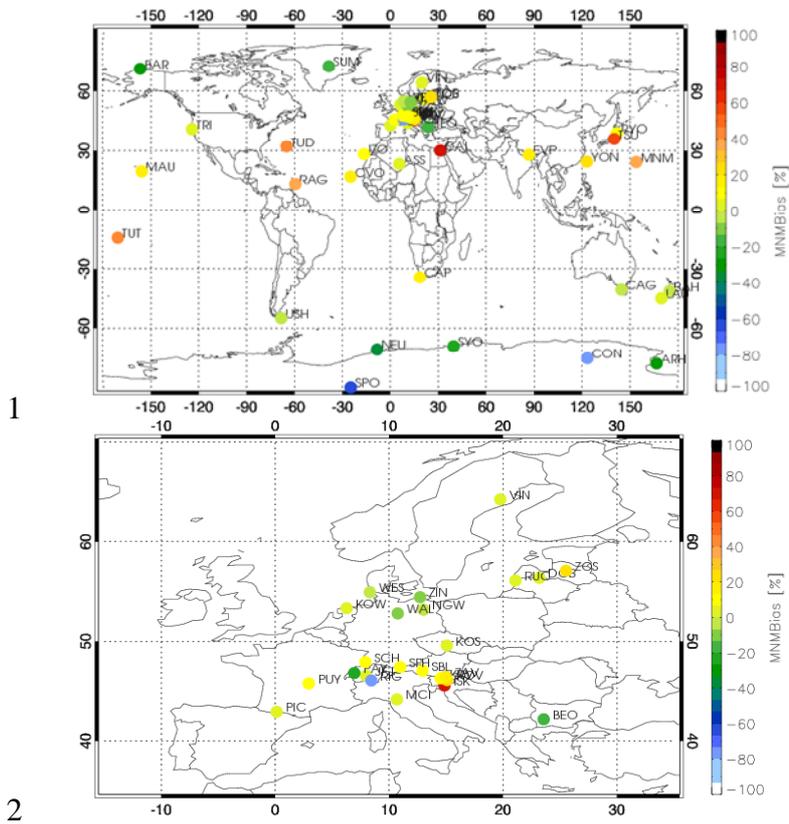
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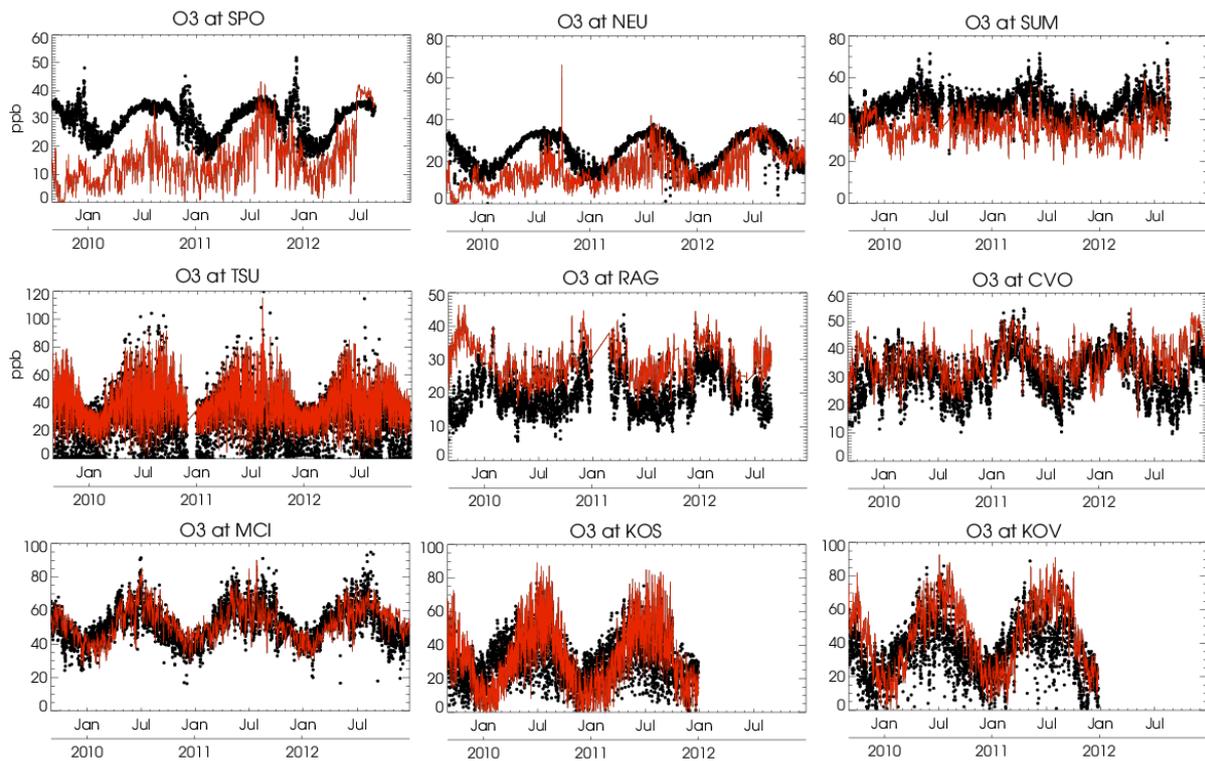


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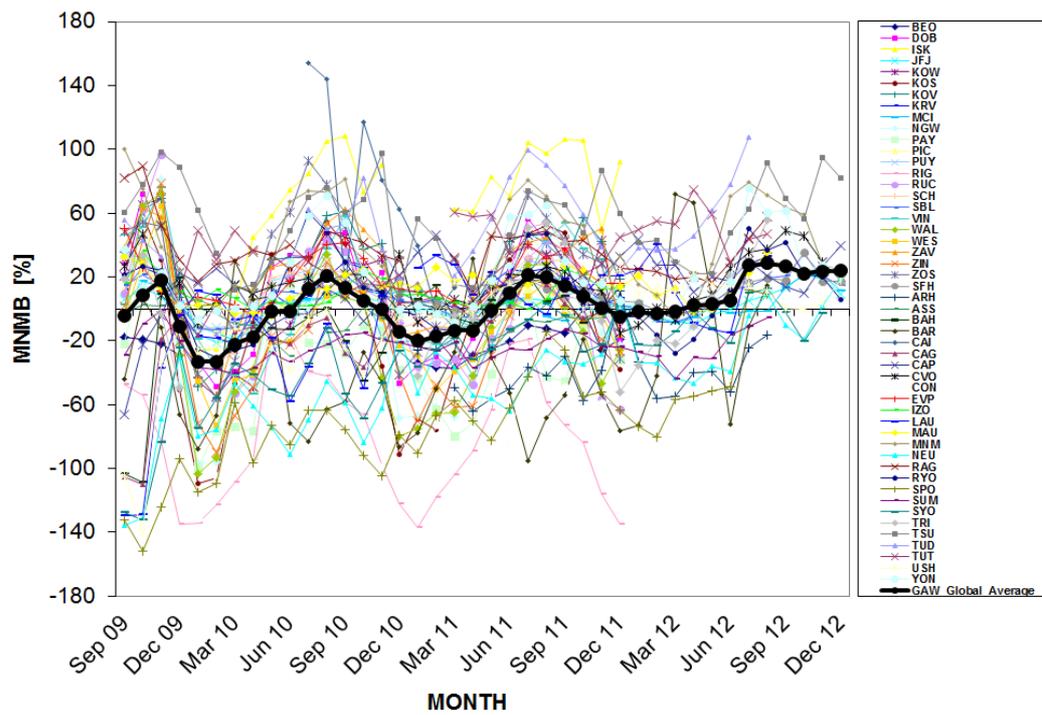
3 Figure 1: Regions used for regional data-stratification in the troposphere for the comparison  
4 with satellite data. The following regions are defined: **1** Europe (15W– 35E, 35N–70N), **2**  
5 Alaska (150W–105W, 55N–70N), **3** Siberia (100E–140E, 40N–65N), **4** North Africa (15W–  
6 45E, 0N–20N), **5** South Africa (15E–45E, 20S–0S), **6** South Asia (50E–95E, 5N–35N), **7** East  
7 Asia (100E–142E, 20N–45N), **8** United States (120W–65W, 30N–45N).



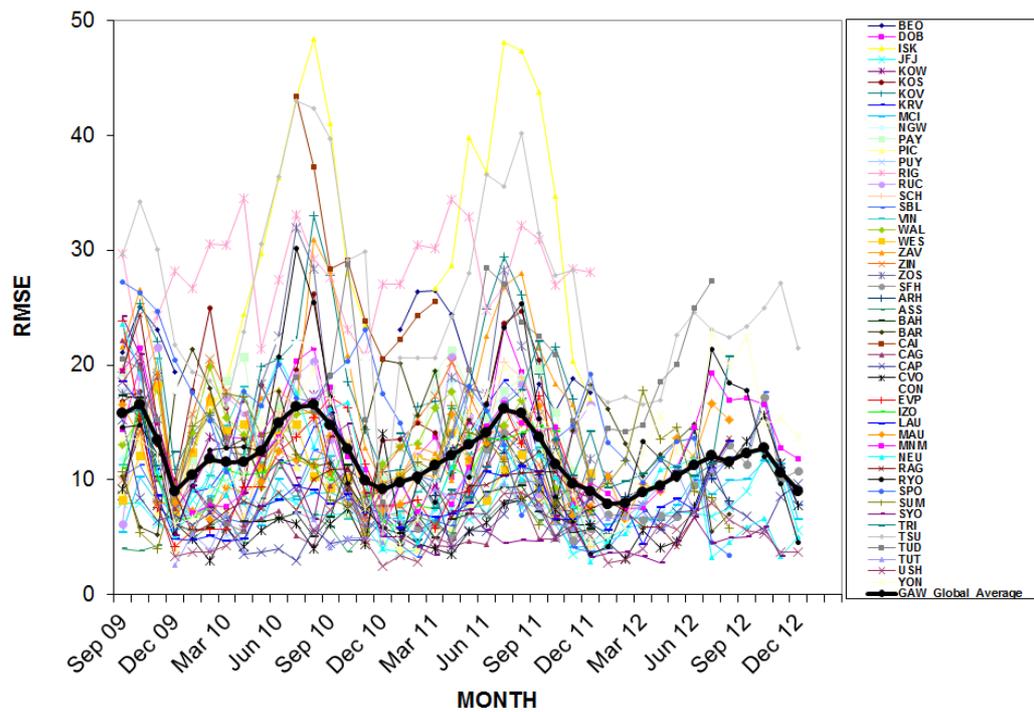
1  
2  
3 Figure 2: Modified normalized mean biases (MNMBs) [%] derived from the evaluation of the  
4 MACC\_osuite with GAW O<sub>3</sub> surface observations during the period 09/2009 to 12/2012  
5 globally (top), and for Europe (below). Blue colours represent large negative values;  
6 red/brown colours represent large positive values.



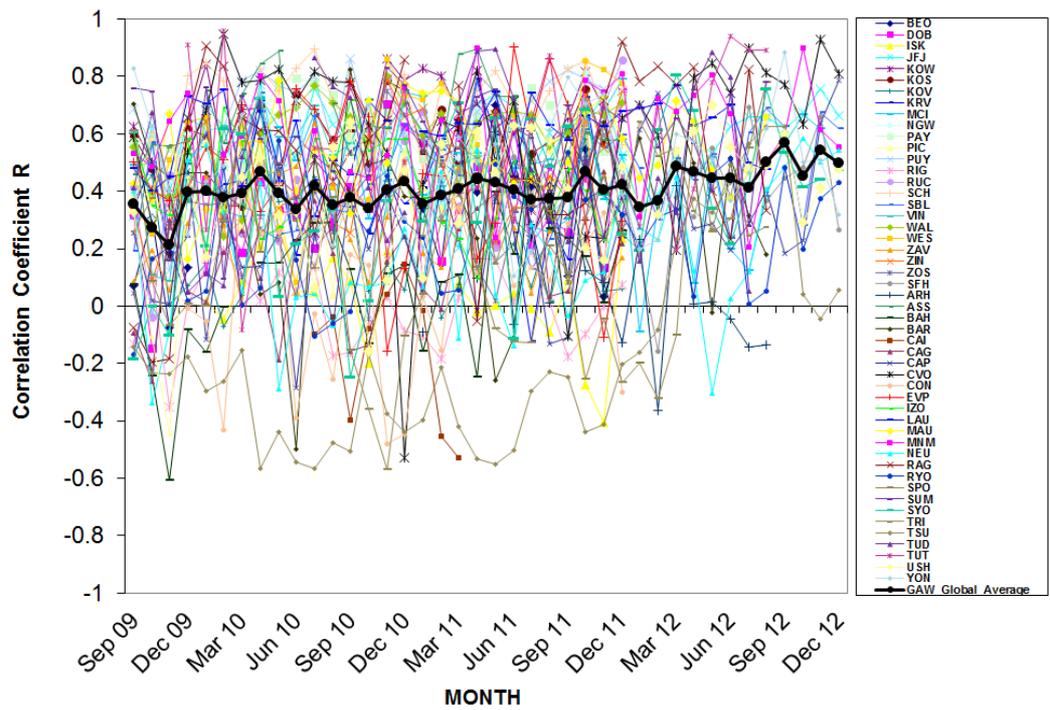
1  
 2 Figure 3: Time series plots of the MACC\_osuite 6-hourly O<sub>3</sub> mixing ratios (red) and GAW  
 3 surface observations (black) for South Pole-SPO (Antarctica), Neumayer-NEU (Antarctica),  
 4 Summit-SUM (Denmark), Tsukuba-TSU (Japan), Ragged Point-RAG, (Barbados), Cape  
 5 Verde Observatory-CVO (Cape Verde), Monte Cimone-MCI (Italy), Kosetice-KOS (Czech  
 6 Republic), Kovk- KOV(Slovenia) during the period 09/2009 to 12/2012. Unit: ppb



1  
 2 Figure 4: Modified normalized mean bias (MNMB) in % derived from the evaluation of the  
 3 MACC\_osuite with GAW O<sub>3</sub> surface observations during the period September 2009 to  
 4 December 2012 (black line: global average of 50 GAW stations. Multi-coloured lines:  
 5 individual station results, see legend to the right).

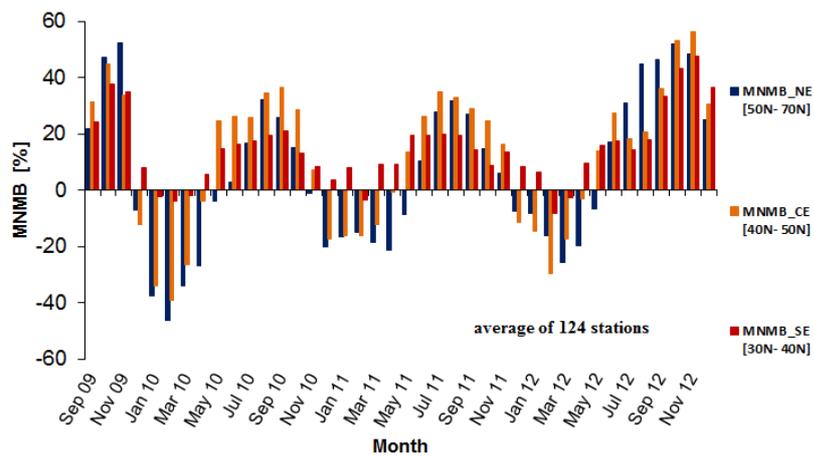


1  
 2 Figure 5: Root mean square error (RMSE) in ppb derived from the evaluation of the  
 3 MACC\_osuite with GAW O<sub>3</sub> surface observations during the period September 2009 to  
 4 December 2012 (black line: global average of 50 GAW stations. Multi-coloured lines:  
 5 individual station results, see legend to the right).

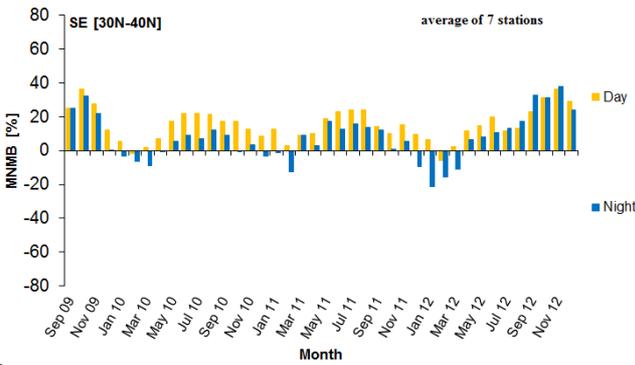
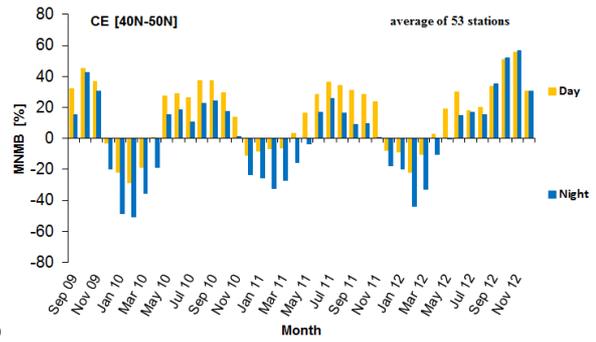
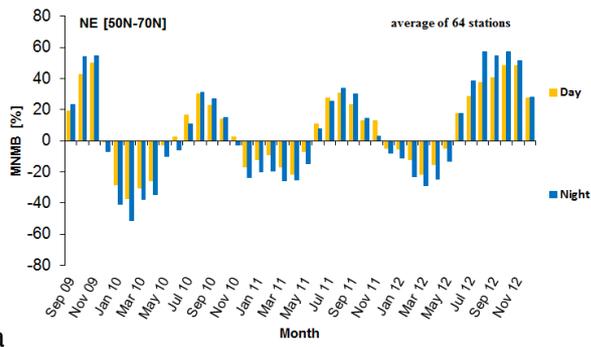


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2 Figure 6: Correlation coefficient (R), derived from the evaluation of the MACC\_osuite with  
 3 GAW O<sub>3</sub> surface observations during the period September 2009 to December 2012 (black  
 4 line: global average of 50 GAW stations. Multi-coloured lines: individual station results, see  
 5 legend to the right).



1  
 2 Figure 7: Modified normalized mean biases (MNMBs) derived from the evaluation of the  
 3 MACC\_osuite with EMEP O<sub>3</sub> surface observations in three different parts in Europe (blue:  
 4 Northern Europe, orange: Central Europe, red: Southern Europe) during the period September  
 5 2009 to December 2012.

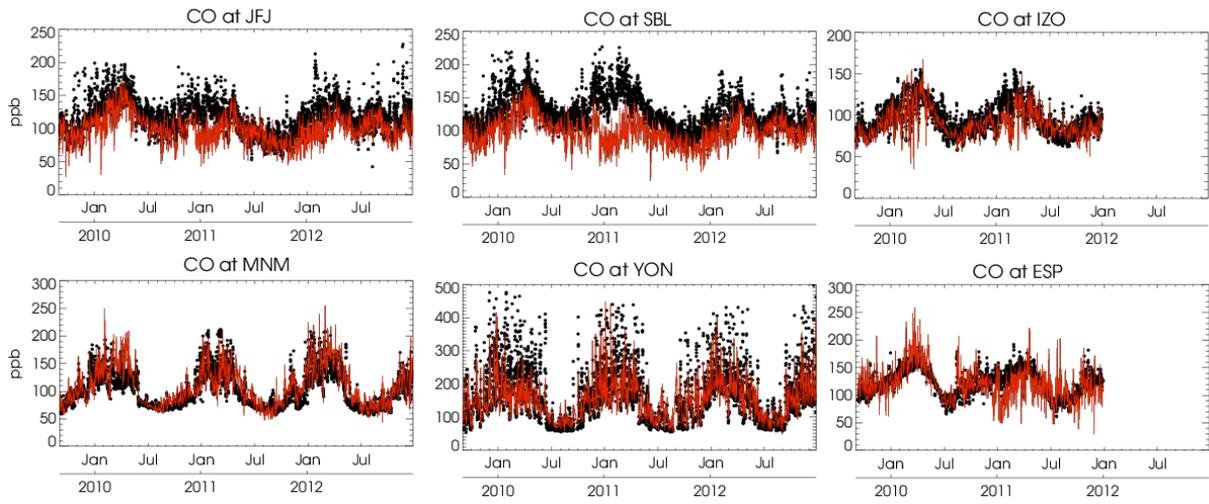


1 a

b

2 c

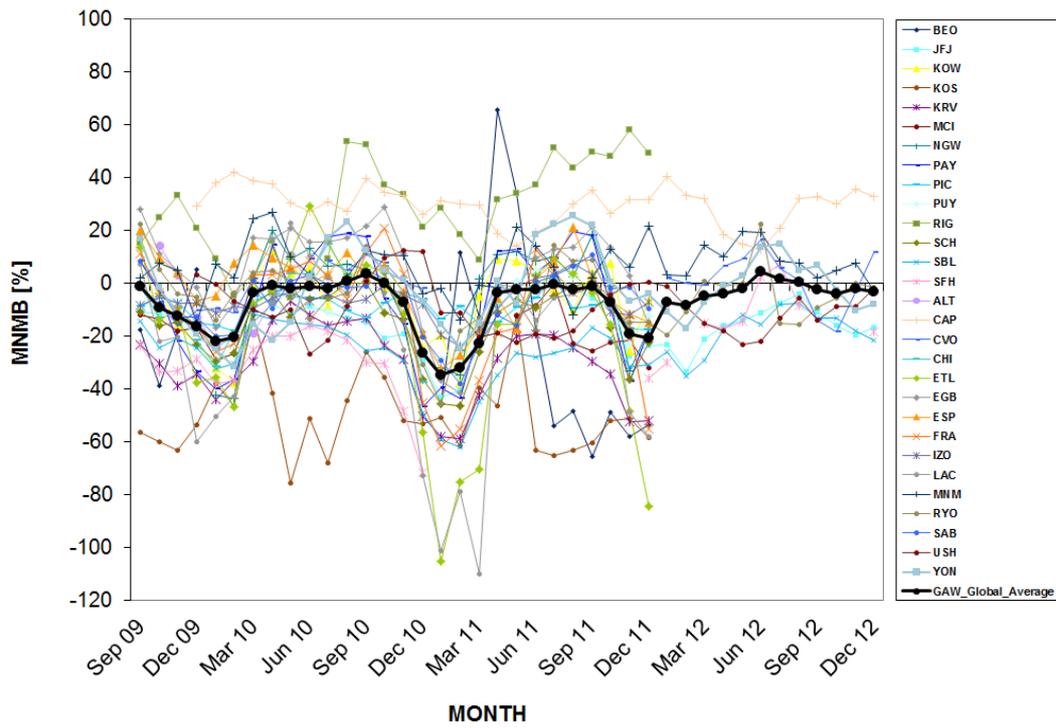
3 Figure 8: Modified normalized mean biases (MNMBs) derived from the evaluation of the  
 4 MACC\_osuite with EMEP O<sub>3</sub> surface observations during day-time (yellow color), and night-  
 5 time (blue color) over northern Europe (a), central Europe (b) and southern Europe (c) during  
 6 the period September 2009 to December 2012.



1

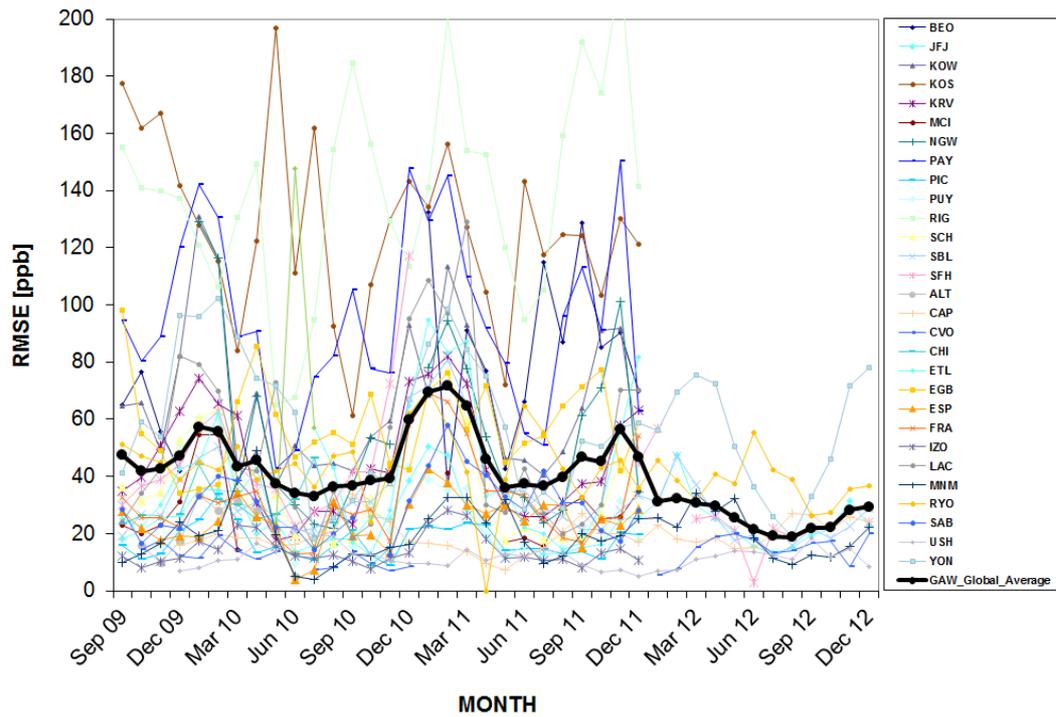
2 Figure 9: Time series plots of the MACC\_osuite 6-hourly CO mixing ratios (red) and GAW  
 3 surface observations (black) for Jungfrau-joch- JFJ (Switzerland), Sonnblick- SBL (Austria),  
 4 Izana Observatory- IZO (Tenerife), Minamitorishima- MNM (Japan), Yonagunijima- YON  
 5 (Japan), Estevan Point- EVP (Canada) during the period 09/2009 to 12/2012. Unit: ppb.

1

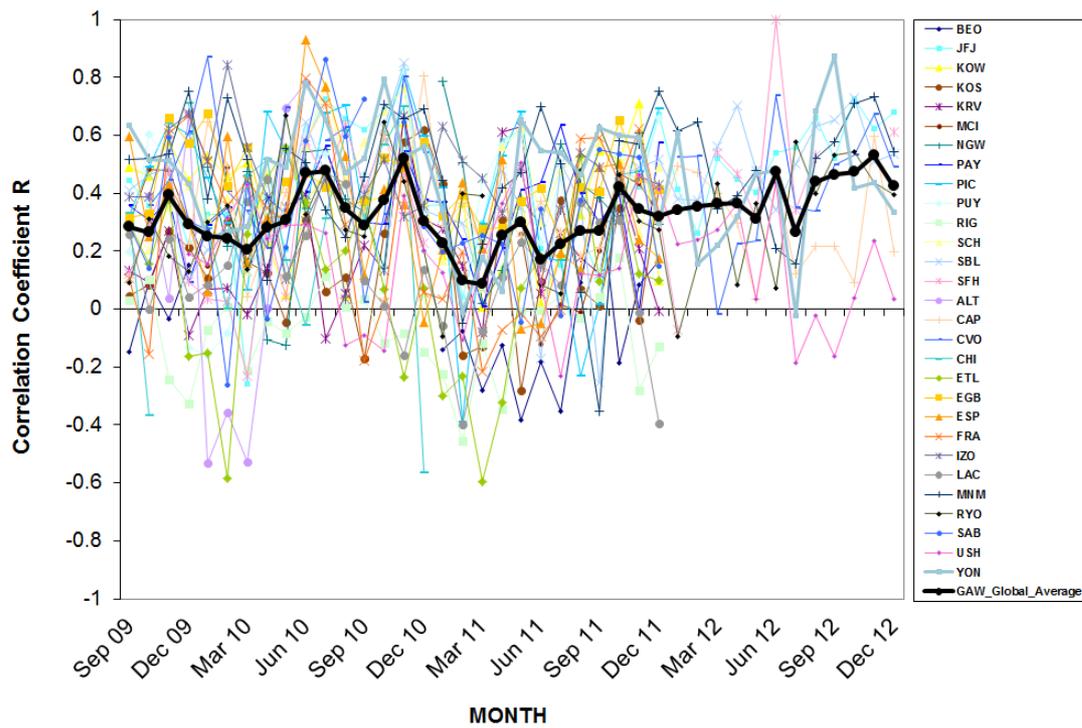


2

3 Figure 10: Modified normalized mean bias (MNMB) in % derived from the evaluation of the  
4 MACC\_osuite with GAW CO surface observations over the period September 2009 to  
5 December 2012 (black line: global average of 29 GAW stations. Multi-coloured lines:  
6 individual station results, see legend to the right).

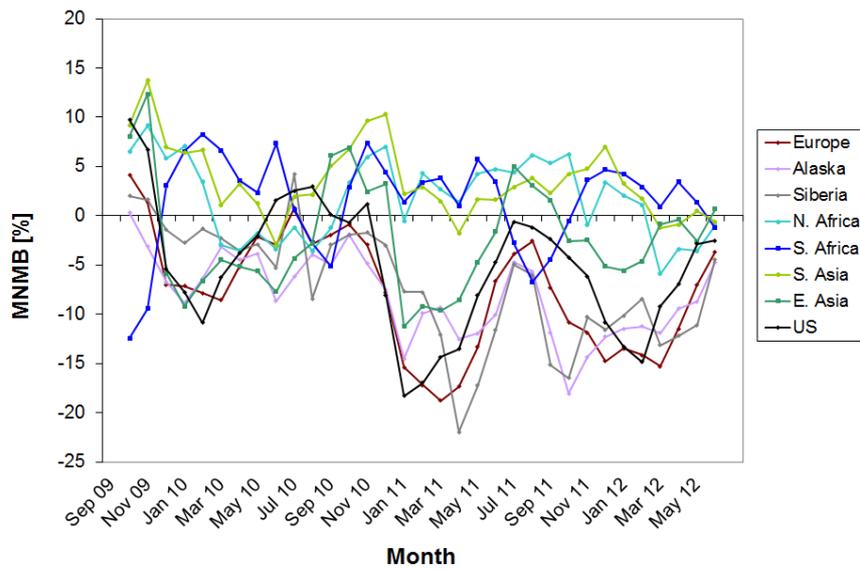


1  
 2 Figure 11: Root mean square error (RMSE) in ppb derived from the evaluation of the  
 3 MACC\_osuite with GAW CO surface observations over the period September 2009 to  
 4 December 2012 (black line: global average of 29 GAW stations multi-coloured lines:  
 5 individual station results, see legend to the right).



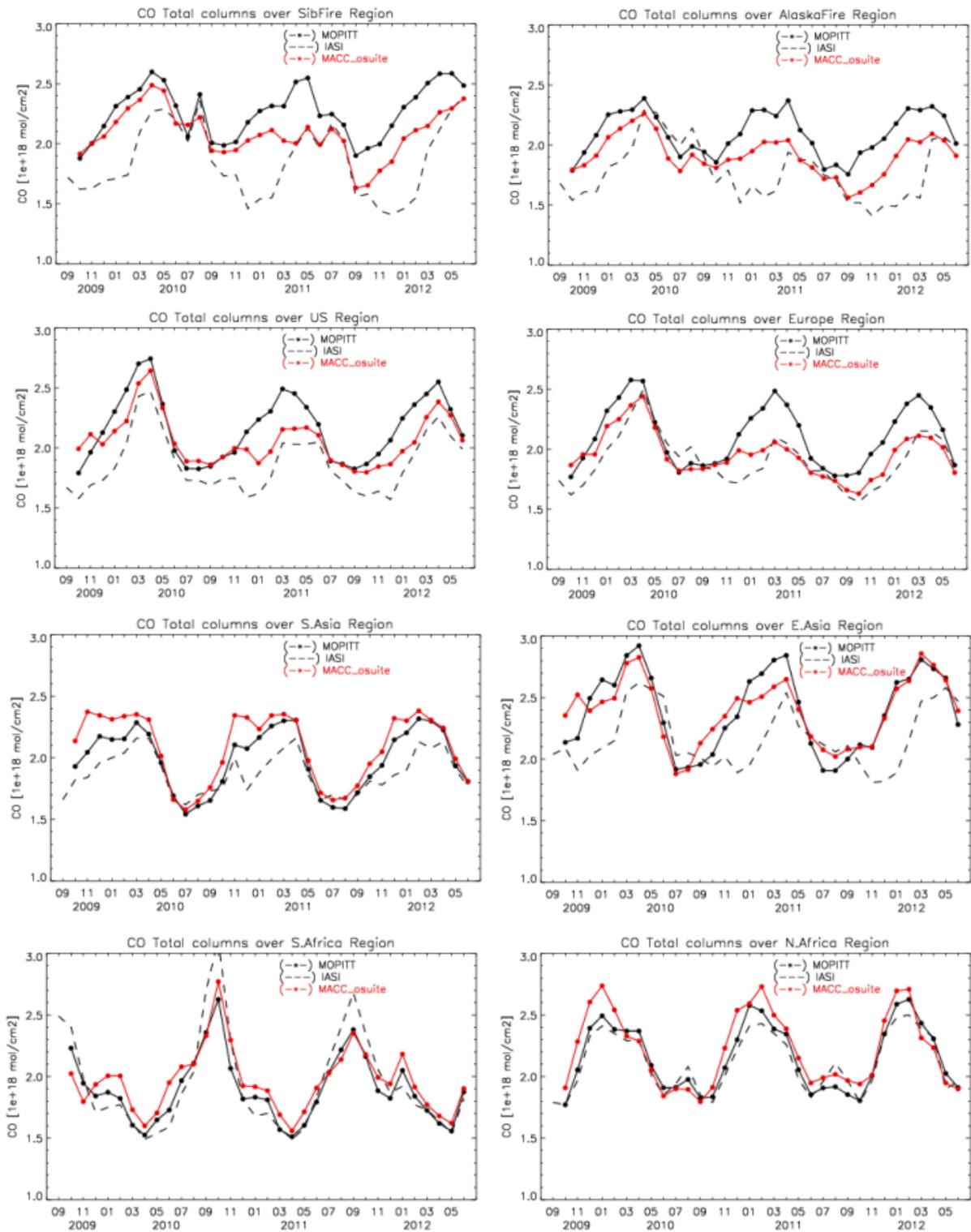
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 2 Figure 12: Correlation coefficient (R), derived from the evaluation of the MACC\_osuite with  
 3 GAW CO surface observations over the period September 2009 to December 2012 (black  
 4 line: global average of 29 GAW stations. Multi-coloured lines: individual station results, see  
 5 legend to the right).

6



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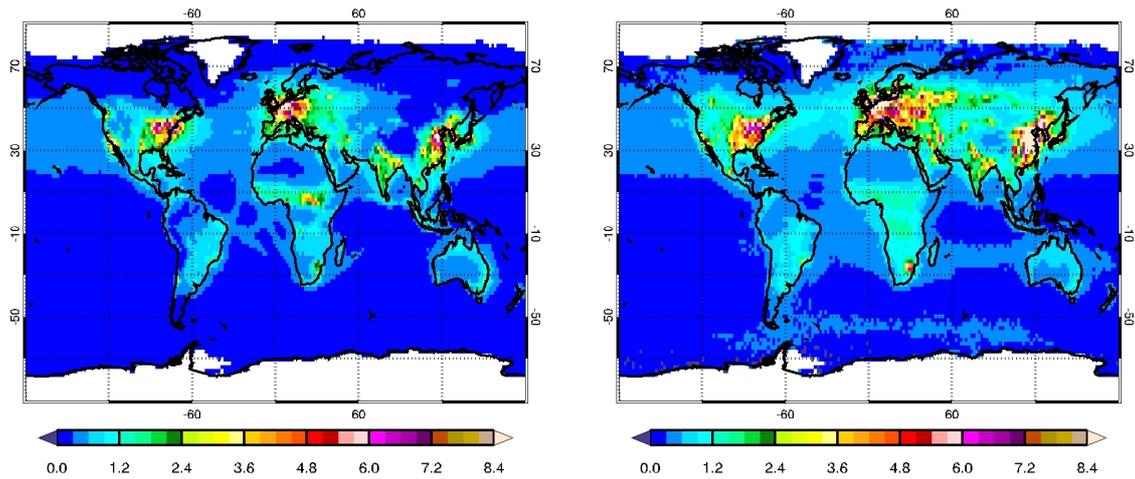
2 Figure 13: Monthly average of modified normalized mean biases (MNMBs) derived from the  
 3 comparison of the MACC\_osuite with MOPITT CO total columns for 8 different regions  
 4 during the period 09/2009 to 06/2012 (see legend on the right).



1

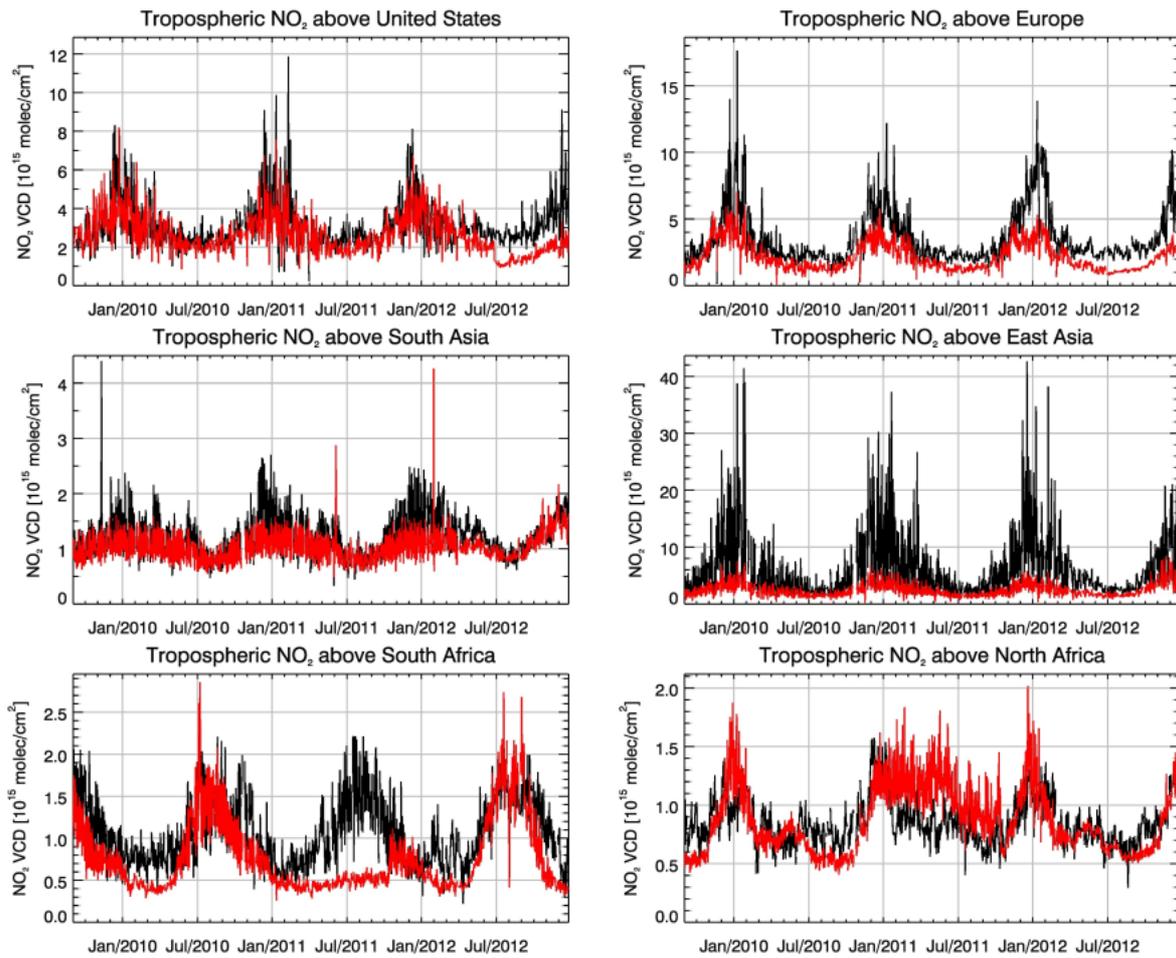
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3 Figure 14: Time series plots of MOPITT CO total columns (black line) compared to IASI CO  
 4 total columns (black dashed line) and the MACC\_osuite CO total columns (red line) for 8  
 5 different regions (defined in Figure 1) during the period 09/2009 to 06/2012. Top: Siberia  
 6 (left), Alaska (right), second row: United States (left), Europe (right), third row: South Asia  
 7 (left), East Asia (right) bottom: South Africa (left), North Africa (right).



1  
 2 Figure 15: Long-term average of daily tropospheric NO<sub>2</sub> VCD [ $10^{15}$  molec cm<sup>-2</sup>] from  
 3 September 2009 to March 2012 for (left) MACC\_osuite simulations and (right)  
 4 SCIAMACHY satellite observations. Blue colours represent low values; red/brown colours  
 5 represent high values.

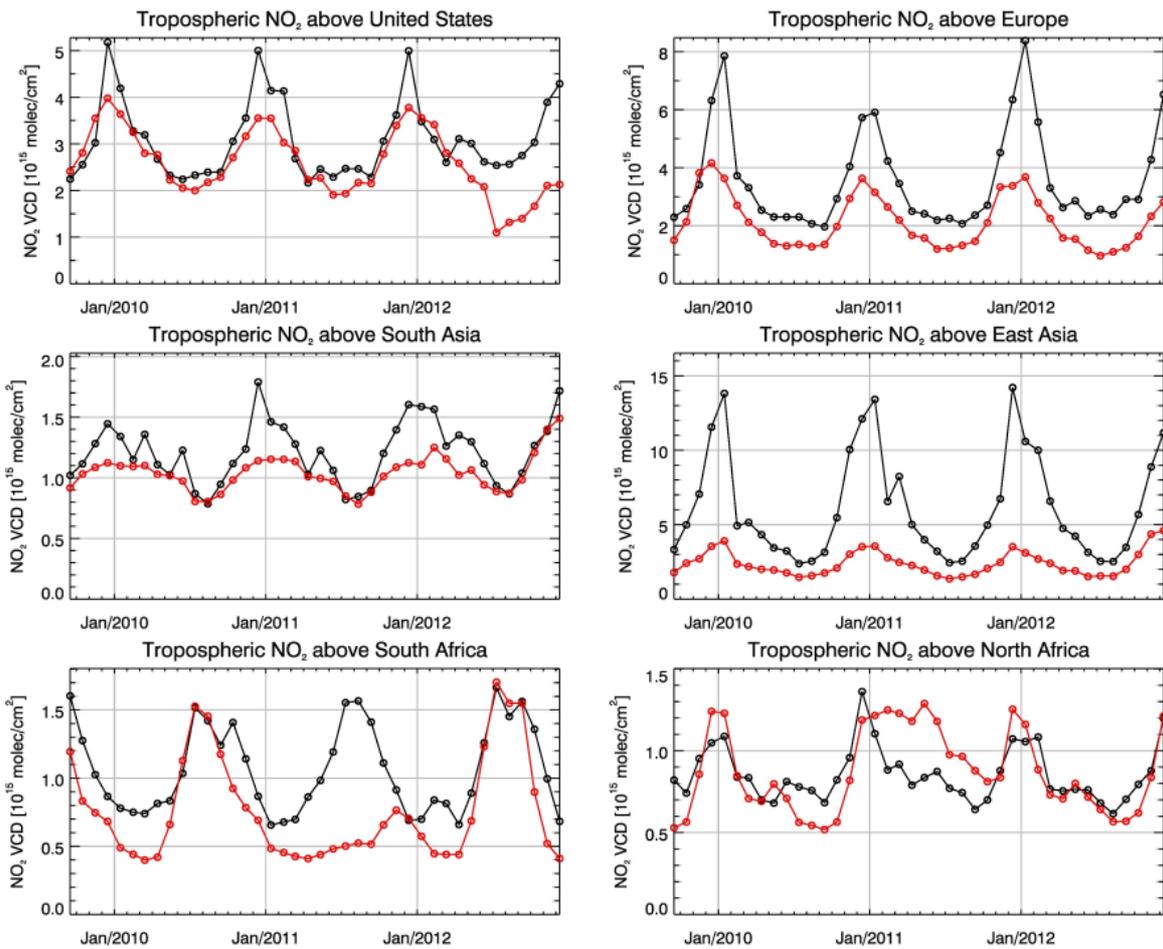
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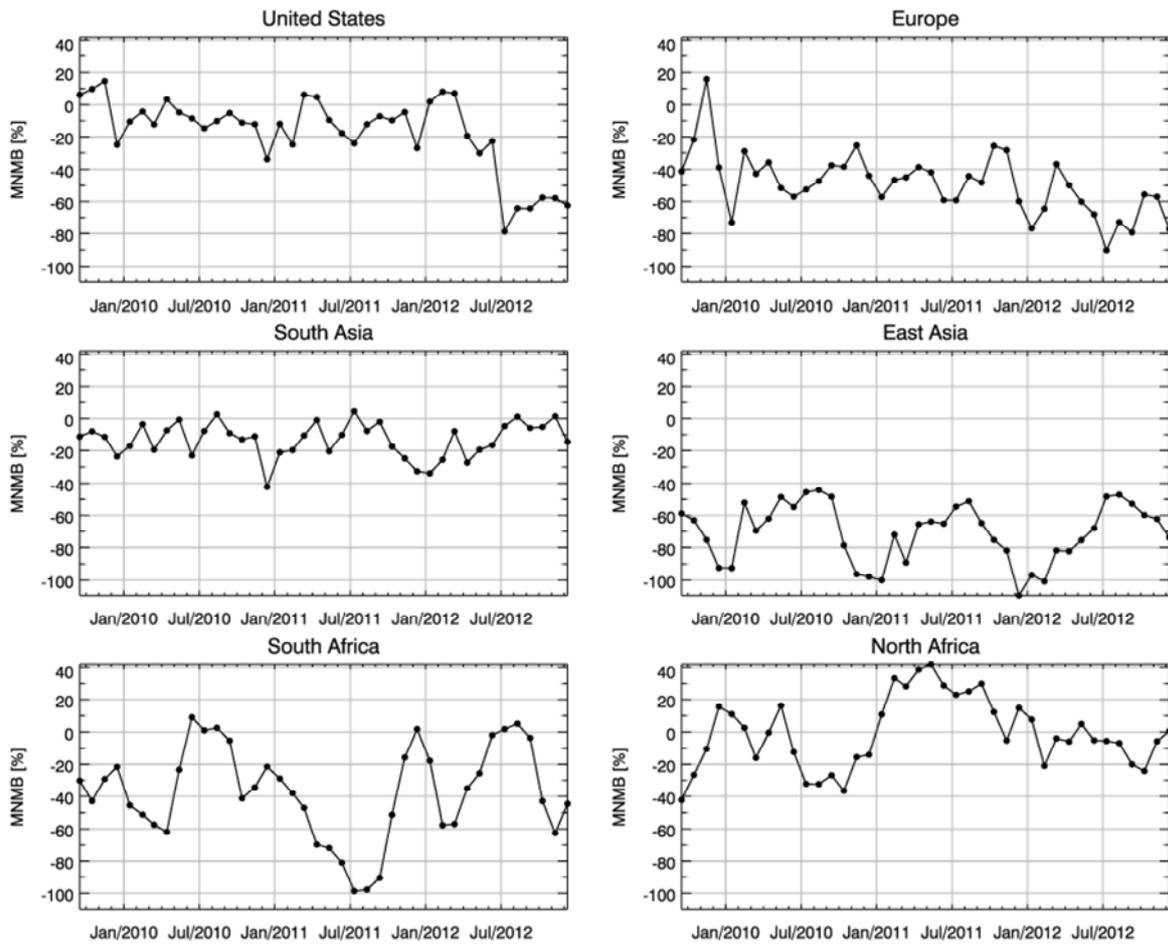
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3 Figure 16: Time series of daily tropospheric NO<sub>2</sub> VCD [10<sup>15</sup> molec cm<sup>-2</sup>] averaged over  
4 different regions. Top: United States (left), Europe (right), second row: South Asia (left), East  
5 Asia (right), bottom: South Africa (left), North Africa (right). Black lines show satellite  
6 observations (SCIAMACHY up to 03/2012, GOME-2 from 04/2012 to 12/2012), red lines  
7 correspond to the MACC\_osite simulations.

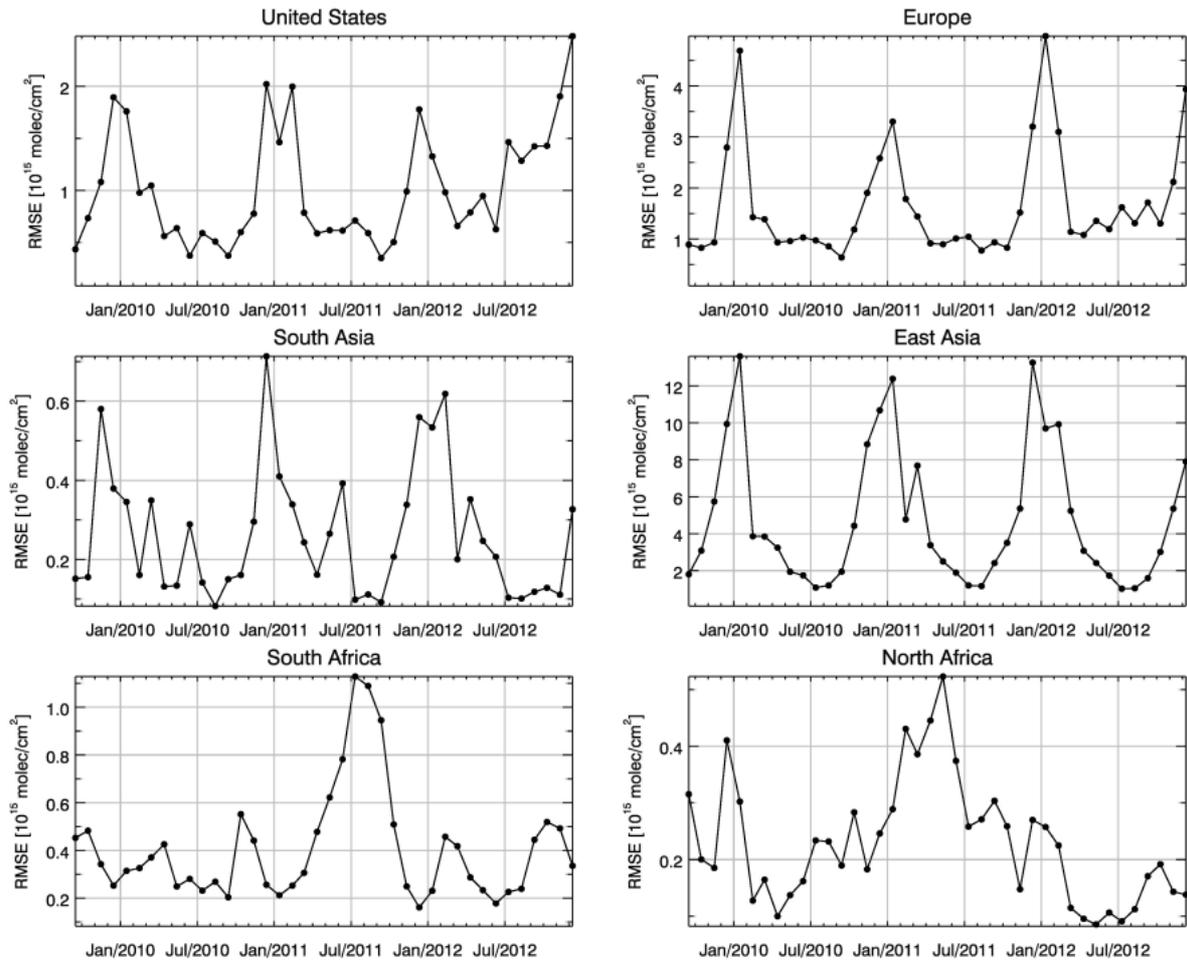
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1  
 2 Figure 17: As in Fig. 16 but for monthly means of daily tropospheric NO<sub>2</sub> VCD [10<sup>15</sup> molec  
 3 cm<sup>-2</sup>] averaged over different regions. Top: United States (left), Europe (right), second row:  
 4 South Asia (left), East Asia (right), bottom: South Africa (left), North Africa (right).



1  
2 Figure 18: Modified normalized mean bias [%] for monthly means of daily tropospheric NO<sub>2</sub>  
3 VCD averaged over different regions (see Fig.1 for latitudinal and longitudinal boundaries)  
4 derived from the MACC\_osuite simulations and satellite observations (SCIAMACHY up to  
5 03/2012, GOME-2 from 04/2012 to 12/2012). Top: United States (left), Europe (right),  
6 second row: South Asia (left), East Asia (right), bottom: South Africa (left), North Africa  
7 (right). Values have been calculated separately for each month.



1

2 Figure 19: As in Fig. 18 but for the root mean square error [ $10^{15}$  molec cm<sup>-2</sup>].