

**Response to reviewer comments on
“Hindcast experiments of tropospheric composition during the summer 2010 fires
over western Russia”
by V. Huijnen et al.**

First of all we would like to thank the reviewer for his careful comments.

Major concerns

1. The scientific goals of this study are not defined sufficiently clearly. It is said in the abstract that the "extreme event is used to evaluate the ability of the global MACC (Monitoring Atmospheric Composition and Climate) atmospheric composition forecasting system to analyze large-scale pollution episodes and to test the respective influence of a priori emission information and data assimilation on the results". These goals and questions formulated in Introduction mostly concern a specific modeling system and have technical character.

I think that the authors should try to put this study into a broader scientific context and to emphasize its importance. In particular, the following questions could be addressed in Introduction:

- (i) What is the place of the MACC system among other similar modeling systems (if they exist)?
- (ii) Has the impact of fire emission estimates on atmospheric composition forecasting been already addressed in any other studies,
- (iii) Have any other modeling systems been evaluated against simultaneous satellite measurements of several species during an extreme air pollution event caused by wildfires? It would also be useful to provide a brief overview of previous studies attempting validation of fire emissions by comparing model results with measurements;
- (iv) What are possible practical applications of forecasting air pollution on the global scale? Are such forecasts needed to provide better boundary conditions for regional models and/or to improve meteorological forecasts? Are there any examples of such applications?

Response:

First of all we would like to thank the reviewer for his careful comments.

In response to the reviewer's comments we have revisited the introduction, giving attention to the place of MACC among other systems, providing a selection of relevant studies that assess the impact of fire emissions, using a range of satellite instruments in several cases. Also the practical applications are mentioned.

Revised text:

Several systems exist that aim to combine various observational data sources to obtain a complete and consistent view of the atmospheric composition. In the United States, an assimilation system for atmospheric composition is developed at the National Aeronautics and Space Administration (NASA)/Global Modeling and Assimilation Office (GMAO; see <http://gmao.gsfc.nasa.gov>). Also the National Oceanic and Atmospheric Administration (NOAA)/National Weather Service (NWS) in collaboration with the U.S. Environmental Protection Agency (EPA) developed a data assimilation system that is

used for operational air quality forecasting. Other examples of chemical data assimilation strategies are described in Sandu and Chai (2011).

In this context the MACC (Monitoring Atmospheric Composition and Climate, <http://www.gmes-atmosphere.eu>) project is a European initiative to achieve a complete and consistent view of the atmospheric composition, and to establish connections between chemical composition and weather forecasts, which in future may improve weather forecasts (Zhang, 2008, Grell and Baklanov, 2011).

In MACC, analyses and forecasts of atmospheric composition are routinely produced based on the coupled system CTM-IFS (Chemistry Transport Model – Integrated Forecast System, Flemming et al., 2009), extended with an aerosol model within the IFS (Morcrette et al., 2009, Benedetti et al., 2009). This data assimilation system makes use of analyses of both meteorology and chemical composition (Hollingsworth et al., 2008). Thus, it is able to monitor variations in chemical composition due to varying meteorology, such as episodes of increased ozone levels caused by heat waves (Ordóñez et al., 2010), or the onset of the ozone hole (Flemming et al., 2011).

The impact of fire emissions on atmospheric composition is long recognized (e.g., Duncan et al., 2003). Various systems for the generation of up-to-date, or even NRT global fire emission estimates have been reported (van der Werf et al., 2010, Wiedinmyer et al., 2010). Chemical composition in fire plumes is assessed using a range of satellite instruments as well as model results from various systems, some of them including chemical data assimilation (Verma et al., 2009, Dupont et al., 2012, Val Martin et al., 2006, Real et al., 2007). Uncertainties in fire inventories on the modeling of atmospheric composition, e.g., Williams et al. (2012), and long-range transport (Miller et al., 2011, McMillan et al., 2010, Elguindi et al., 2010) have previously been quantified.

Within MACC, a NRT daily fire emission estimate based on FRP observations from MODIS was developed: the Global Fire Assimilation System (GFAS, Kaiser et al., 2012). In this study we assess the capability of the MACC system using NRT fire emission estimates to forecast chemical composition a few days in advance. These forecasts are relevant to serve as boundary conditions to regional air quality models, as distant emissions can potentially influence air quality (Hodzic et al., 2007, Pfister et al., 2011). For instance, for the 2010 Russian fires smoke particles have been reported over Finland (Kaiser et al., 2012).

The summer 2010 wildfires over western Russia provide an opportunity for a comprehensive assessment of the MACC assimilation/forecast system for this type of extreme pollution event. In this paper, we focus on the following questions: What is the relative importance of (1) the chemical data assimilation and (2) the NRT fire emission estimates on the accuracy of forecasts of tropospheric composition? What are direct effects, and which effects can be attributed to chemical interaction within the system, e.g. by persistent changes in the oxidative capacity? These questions may help to identify

causes of variations in forecast accuracy from the perspective of both the modeling and the observing framework and can provide guidelines to improve them.

2. In most cases, the difference between the error statistics of the Assim and Assim-GFAS runs is very small. This is a rather puzzling result, which may mean that the model does not add any important information to the forecasted characteristics in comparison with corresponding contribution of measurements. It also seems possible that the assimilation system is not well optimized. Since one of the goals of this study is to evaluate the MACC system, it would be important to demonstrate that the information from the model is combined with observations in an optimal way. My suggestion is to perform two more runs with the error covariances increased or decreased (globally) by, e.g. 50 percent. Such experiments would clarify the respective contributions of the model and observations to the forecasts and could help in identifying possible ways to further improve the MACC system performance.

The reviewer notes there is little difference in the error statistics between runs Assim and Assim-GFAS. We do not fully agree with his comments, considering the statistics given in Tables 4 – 10, where differences are generally not negligible between those runs, and furthermore mostly increase with increasing hindcast length. The smallest difference was found for assessment of the RMSE of NO₂ with respect to OMI data (Table 9), which for all hindcast days and all systems led to similar results. This seems therefore more related with issues that are common for all model versions, e.g. the spatial distribution and magnitude of anthropogenic and soil NO_x emissions, rather than issues with the assimilation. Relatively little difference is also found for both the bias and RMSE of D+0 hindcasts of CO. This can partly be explained by the fact that run 'Assim' implicitly does include fire emissions through the assimilation of high CO columns from the IASI product. As discussed in the manuscript, the vertical distribution of CO concentrations is much more realistic in run Assim-GFAS, see also Fig. 7. However, at low altitudes the satellite instrument has little sensitivity and this can therefore not be discriminated through validation with the MOPITT satellite retrieval. Therefore we now include an evaluation of ground-based CO total columns, see also our response to the other reviewer. Furthermore, the impact of fire emissions in run Assim-GFAS becomes clear from the statistics for hindcast days D+1 to D+3. This all shows that the model does add important information to the hindcasts.

Nevertheless, we acknowledge that the assimilation procedure in the MACC system can be further optimized. For instance, the NMC method (Parish and Derber, 1992), which is used to derive initial background error statistics for the reactive gases, does not account for errors in the emissions. But it is beyond the scope of the current work to assess how background errors should be improved.

But we think the current experiments with/without assimilation are suitable to assess the impact of initialization by assimilation on hindcasts of chemical composition, showing its merits (reduction of model biases) as well as its limitations (e.g. the little sensitivity to the surface, no aerosol composition information, possible biases for CO, and the lack of long-term influence for NO₂).

To accommodate the concerns of the reviewer we add the following statement to section 2.1:

Background error statistics for reactive gases have been derived using the NMC method (Parish and Derber, 1992), which currently does not account for high variability in emissions or the correlation between different trace gases.

3. The IASI measurements show high ozone columns over Kazakhstan which are not reproduced by the model. The authors discuss several possible reasons but recognize that the true reasons are not fully understood. It also remains unknown whether this feature is due to uncertainties in the IASI retrieval or due to some deficiencies in the model. I believe that the authors should put some more efforts in elucidating the origin of this puzzling ozone high. In particular, the respective OMI data for the upper troposphere (which are anyway used in the assimilation) could be considered for this purpose.

The reviewer is correct that the puzzling low model bias in tropospheric ozone over Kazakhstan was not satisfactory. To constrain further uncertainties in both the model and the retrieval we have analyzed this aspect in more detail.

As mentioned in the manuscript, local ozone production may suffer from uncertainties related to soil and biogenic emissions, which are dominating over anthropogenic and fire emissions locally. An assessment of the applied emissions (Lathiere et al., 2006) compared to another inventory (MEGANv2, Guenther et al., 2006) shows that the currently used emissions are actually on the high side. So updating this would probably lead to reductions in ozone production. A sensitivity study was done using globally half the soil NO_x emissions. This showed a change in tropospheric O₃ concentrations on the order of 5%, which gives an upper limit of the regional impact of the uncertainty in these emissions. As written in the manuscript, this uncertainty on regionally averaged tropospheric ozone concentrations can well contribute to the bias that is observed between the model and IASI observations. However, we do not believe that this can explain the high O₃ columns over Kazakhstan, with a discrepancy between the model and observations of the order of 30%, considering its relatively local scale.

An analysis of the IASI O₃ retrieval shows that the quality of the IASI spectra fit is less good in the region of Kazakhstan. We suspect that interference of ozone band with water vapor lines could perturb the ozone retrieval, as was written in the manuscript. This still needs further investigation. For our purposes we now add an additional filter on the product, to discard the data with too poor fit quality. To account for other sources of error than radiometric noise (due to uncertainties on temperature, spectroscopy etc), a noise level of about 70 nW/(cm² cm⁻¹ sr) on the radiance measured by IASI seems more realistic. When we base our additional quality filter on this criterion the area with high tropospheric ozone columns is significantly reduced, while it did not change ozone columns elsewhere. With this filter we are more confident in the IASI data.

As suggested by the reviewer, we have further looked at a tropospheric ozone product derived from OMI observations (Ziemke et al., 2006, http://acd-ext.gsfc.nasa.gov/Data_services/cloud_slice/). During August 2010 over Kazakhstan the tropospheric O₃ product does show increased columns, with a magnitude of ~45 DU, i.e. substantially lower than the IASI 0-12 km product for this month (~55 DU), but this is not conclusive, because the two products are too different. For instance, the tropospheric column from OMI is derived indirectly, subtracting the stratospheric contribution from the total column, thereby introducing uncertainties on the tropopause height. Clearly also the vertical extend of the two products is different. Therefore a quantitative assessment on the agreement between the IASI and OMI tropospheric O₃

products is beyond the scope of this work, and cannot be used to assess biases in one or the other product.

Summarizing, in the manuscript we now use the IASI product with a more strict filter for uncertain retrievals, and change the discussion accordingly, also mentioning the assessment of the emissions applied in the model.

4. The accuracy of the hindcasts is evaluated in terms of the bias and RMSE. Another important metric which is used in most of forecasting studies is the correlation coefficient (for time series). Values of the correlation coefficient should be provided along with the bias and RMSE in order to facilitate using the results of this study for future references.

We considered the use of temporal correlation for assessing our hindcast results. However, we decided not to apply this metric to evaluation against satellite observations because of several issues.

Temporal correlation statistics is misleading when calculated from satellite data: this data does not have a uniform daily coverage, e.g. due to varying cloud cover, or different overpass times and variable availability of measurements. This is especially important for short-lived trace gases such as HCHO and NO₂. Furthermore, individual observations at the same location but at different days are significantly influenced by scatter in the retrieval. This all makes the interpretation of the numbers unclear and easily misleading.

Temporal correlation statistics is most meaningful when based on in-situ observations having a good temporal data coverage. This is currently the case for the ground-based AOD observations; for the ground-based CO total columns observations the time series is less suitable, as it has some data gaps. We now provide temporal correlation statistics for those two observational time series and provide an interpretation to that.

5. Time series are shown for CO and ozone but not for NO₂ and HCHO. For HCHO, even the biases and RMSE are not reported. The missing figures and tables should be provided. My opinion is that scientific results should be presented in an objective way, even if some of them may not look "nice". The same comment also concerns Fig. 13, where an unspecified spatial smoothing is applied to the SCIAMACHY measurements. The original measurements should also be shown.

We did not show NO₂ time series previously because they convey limited additional information on model performance additional to what can be perceived from the maps, the scatter plots and the tables. Reasons for that are similar as mentioned above with respect to the temporal correlation: variations in magnitude between different days (or the lack of them) are not generally caused by differences in local daily concentrations, but also by differences in the spatial distribution of retrieved pixels.

Nevertheless, some variability of retrieval data is visible from area-average NO₂ time series. We agree with the reviewer that this aspect, as well as daily differences between the various model runs, helps to better assess the overall NO₂ evaluation. Therefore we now provide time series of NO₂ against both SCIAMACHY and OMI retrievals.

For HCHO an evaluation of time series on a daily basis is problematic, considering the very sparse amount of individual retrieval pixels per day, in addition combined with the large scatter of the measured values and high uncertainty associated to single measurements. Therefore we decide not to present this data, as these individual daily average observations are not representative for a larger region and hence misleading.

Also the RMSE and mean bias statistics for HCHO are seriously affected by retrieval issues, as we have written in the manuscript, and hence do not yield any significant information. For the HCHO evaluation the scatter plots are most relevant, illustrating key performance issues of the model with respect to the observations.

In accordance to the reviewer's comments we now present unsmoothed HCHO columns, which also illustrates scatter in the data. We did not show this before because we argued that for a qualitative comparison of fields it would not be useful to see the scatter as a consequence of retrieval uncertainties. Note that model evaluation performed in terms of regression slope and spatial correlation was done using unsmoothed figures.

Yet we agree with the reviewer that for a fair understanding of the observational data it is better to show the unsmoothed HCHO fields, so that the reader can better assess aspects related to the HCHO evaluation. Furthermore, we have upgraded the evaluation methodology for the HCHO SCIAMACHY (e.g., a better re-gridding of satellite data into the model grid), which has lead to changes in the HCHO values for the regression slope and spatial correlation.

Other comments

Abstract, l.7.: "analyze": In my understanding, the paper discusses the ability of MACC to forecast rather than to analyze large-scale pollution episodes.

The reviewer is correct that "analyze" is misleading in this context. It should be understood as a reference to the assimilation system, which provides an "analysis" of the atmospheric composition.

We now change this to:

This extreme event is used to evaluate the ability of the global MACC (...) atmospheric composition forecasting system to provide analyses of large-scale pollution episodes...

Introduction, p. 31853. The statement: "a range of observations were used in various studies to characterize the tropospheric composition during this episode : : ." may be understood such that all of the four mentioned studies are purely observational (in contrast to the study by Huijnen et al.), what is not true. The respective paragraph should be revised and extended to allow a reader to get a better and more accurate idea about the previous studies of the same episode.

We have revised the introduction accordingly:

A range of observations were used in various studies to characterize the tropospheric composition during this episode, including in-situ data for fine and coarse aerosol mass ($PM_{2.5}$ and PM_{10} , van Donkelaar et al., 2011), aerosol optical thickness (Chubarova et al., 2011) ozone (O_3) and carbon monoxide (CO) (Konovalov et al., 2011, Elansky et al., 2011), CO total columns (Yurganov et al., 2011), as well as space-based information of CO total columns from Atmospheric Infrared Sounder (AIRS), and aerosol optical depth (AOD) and single-scattering albedo from Ozone Monitoring Instrument (OMI) (Witte et al., 2011, Mei et al., 2011).

In addition, it had been shown before that fire episodes can be analyzed by the Infrared Atmospheric Sounding Interferometer (IASI) CO (Turquety et al., 2009) as well as nitrogen dioxide (NO_2) and formaldehyde (HCHO) observations from the OMI and Scanning Imaging Absorption SpectroMeter for Atmospheric Chartography (SCIAMACHY) instruments (e.g., Mebust et al., 2011; Stavrakou et al., 2009). Model studies assessing estimates of emissions for CO (Konovalov et al., 2011, Yurganov et al., 2011, Fokeeva et al., 2011), and aerosol (van Donkelaar et al., 2011, Kaiser et al., 2012) have been reported for the 2010 Russian fires.

p. 31857, l. 4,5: " It applies the same 60 level vertical discretization as the IFS, but the horizontal resolution is 3 lon x 2 lat, globally". Is the horizontal resolution of IFS different?

Yes, as specified on p. 31856, l 23, IFS runs on T159, which is approx. 1.2 deg.

p. 31859, l. 20. "23 percent": Where this critical value is taken from (reference)?

This value was based on the assumption that water stress occurs below $0.5 \theta_{sat}$, the volumetric soil moisture at saturation. Here $\theta_{sat} = 0.472 \text{ m}^3/\text{m}^3$ for loamy soils, see, e.g., the IFS manual, <http://www.ecmwf.int/research/ifsdocs/CY37r2/IFSPart4.pdf>, and references therein. We acknowledge that the actual value of $0.5 \theta_{sat}$ is rather arbitrary, and change the text accordingly to:

... the soil wetness was below the critical level of 23%, i.e. half the volumetric soil moisture at saturation, where water stress in vegetation is assumed to take place.

p. 31860, l. 18-21: "Furthermore, the CO and HCHO emissions are much higher than the monthly-mean GFEDv3.1 emissions. This is mainly caused by the different predominant soil type maps used in GFEDv3.1 and GFASv1.0". Can the authors justify the last statement, or is it simply a guess?

GFEDv3.1 represents organic soil content in parts of Russia but only for the calculation of the dry matter combustion rate; the emission factors for peat are not applied outside tropical peat regions (van der Werf et al., 2010). GFASv1.0 applies emission factors for peat across all tropical and Russian peat land cover types. The finding of larger CO and

HCHO emissions and lower NO_x and BC emissions in GFAS than in GFED is consistent with the differences in respective emission factors for peat and agricultural fires.

We have updated the manuscript accordingly.

p. 31860: "the CO and HCHO emissions are much higher than the monthly-mean GFEDv3.1 emissions": It is mentioned on the page 31858 that the conversion factor was derived with a linear regression between the observed fire radiative energy and the dry matter burned in the GFEDv3.1 inventory. Do these facts imply that GFAS estimates are much smaller than the GFEDv3.1 data in some other regions. Are so large differences between GFAS and GFEDv3.1 typical, or these region and event are exceptional? Please comment.

If the conversion factor had been as a ratio of global budgets then compensations in other regions and events could be expected. However, the approach of a linear regression has been chosen for the very reason of minimising such compensations. This leads, for example, to overall slightly higher emission because of the lower detection threshold of GFAS, which is not compensated in any of the more active fire regions. Further differences occur because the land cover map of GFED is finer than the one of GFAS (500m vs 0.5deg). On a global multi-annual average, the CO emission of GFAS are 6% larger than the ones of GFED. On a continental scale it may be larger or smaller by typically 20% and in the region east of Moscow with variable fires and the dedicated peat emission factors of GFAS, it is typically twice as large in GFAS than in GFED. For more details, see Tab. 5 and Fig. 7 of Kaiser et al. BG 2012.

For clarity we now include the following comment in the description of GFAS:

...on a global, multi-annual average, the CO emissions of GFASv1.0 are 6% larger than GFEDv3.1, on a continental scale differences are typically 20%.

p. 31860, l. 28: "The CO emissions are ~25% higher than Konovalov et al. (2011)" According to Table 3 of the reviewed paper, CO emissions from fires in the considered regions during July and August are ~ 13.3 Tg, while according to Table 4 in Konovalov et al. (2011) the fires in European Russia emitted about 12.8 Tg of CO in the same period. That is, the difference is actually less than 4 percent. This is an encouraging agreement. However, the considered regions are not quite identical. Please correct the discussion accordingly (including the conclusions). *ibid*: ": : higher than Konovalov et al. (2011)" => ": : higher than in Konovalov et al. (2011)"

We interpret Table 4 of Konovalov et al. (2011) as reporting total fire emissions over European Russia of 9.7 Tg CO, of which 3.2 Tg is attributed to peat fires. This appears to be different to the reviewer's interpretation, who added the two numbers. So we think the statement of ~25% higher emissions as found in the current study compared to Konovalov et al. is correct.

p. 31862, l. 28: "The RMSE for D+3 in run Assim-GFAS is slightly worse than the one in run Assim": I would not say that the difference between 0.77 and 0.52 (that is, almost 50 percent) is really a "slight" difference.

The reviewer is correct. We remove 'slightly'

p. 31866, l. 16, 17.: "key spatial patterns are captured by the model, such as the north-south gradient in O₃ columns over Western Russia". What are the other "key spatial patterns" captured by the model?

We change this to:

... the large-scale spatial patterns are captured by the model, including the north-south gradient in O₃ columns over western Russia.

p. 31866, l. 23: "This is larges for run CNT" => "It is largest for the run CNT"

Replaced.

p. 31869: I do not think that the large difference in RMSE obtained with SCIAMACHY and OMI data is explained sufficiently; the corresponding discussion should be extended. In particular, can these differences relate to any differences in the retrieval algorithm? What is the potential impact of optically dense aerosols on the different satellite NO₂ data products (see, e.g., Leitao et al., 2010)?

When looking more into the difference between results obtained with SCIAMACHY and OMI, we took the opportunity to update the analysis with the new DOMINO v2.0 product, version 2.0 (Boersma et al., 2011). With the use of DOMINO v2.0 model biases remain practically identical. Also we found a bug in the post-processing script for OMI data, neglecting the 'root' operation in the calculation of the RMSE. This resulted in an increase of RMSE values, which are now in the order of 0.4×10^{15} molec/cm². We apologize for this error.

There are significant differences between the DOMINO v2 and Bremen SCIAMACHY NO₂ retrieval products, e.g. for the comparison to OMI NO₂ we apply averaging kernels to the model, while for the comparison to SCIAMACHY these are not used. Furthermore, the two instruments are onboard satellites with different overpass time: SCIAMACHY measures in the morning and OMI in the afternoon. Also, differences in the coverage (better for OMI) and resolution (better for OMI) result in more observations per model gridbox for OMI. Therefore the daily mean SCIAMACHY fields, and hence the daily RMSE over western Russia, is more influenced by retrieval errors than with OMI. We believe this is the largest contributor to the differences in the RMSE between OMI and SCIAMACHY.

Leitao et al. (2010) show that the sensitivity of the satellite measurements for a particular trace gas can be increased (albedo effect) or decreased (screening), depending on the amount and optical properties of the aerosols, and its vertical distribution relative to that of the trace gas. Because of this complexity of effects one cannot easily estimate how the vertical columns values would change if a different aerosol correction had been applied.

Regarding the OMI product, Boersma et al., (2011) note that in the DOMINO retrieval, most of the required aerosol correction proceeds implicitly through the cloud retrieval approach, which accounts for the combined effect of clouds and aerosols.

We now include a comment on the uncertainty due to aerosol in the manuscript, as well as a more detailed description of differences between the OMI and SCIAMACHY products.

p. 31872, l. 22: "The increase in CO lifetime illustrates a reduction of the hydroxyl radical (OH) concentration". Would not it be better to say that "the increase in CO lifetime is due to a reduction of the hydroxyl radical (OH) concentration"?

We agree with the reviewer that it is more clear to formulate the reason for the CO increase as he suggests. We change the text accordingly.

p. 31876, l. 8: "An important factor for the accuracy of the fire emissions was the development of a detailed soil map". I do not think that the importance of this factor is really demonstrated in this study. Did authors try using emissions obtained with some other soil map?

A map that did not contain peat soils was tested previously, and showed significant under-estimations of CO emissions and hence modeled CO concentrations wrt MOPITT observations. Because of the relatively trivial findings that without peat soils the emissions were highly underestimated this simulation was not included in the manuscript. We will now include the following comment to Section 2.2:

We note that an earlier attempt to generate emissions during the 2010 Russian fires episode yielded far lower CO emissions, partly because the biome distribution originally did not contain peat soils in the area around Moscow.

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