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Geoscientific
Model Development
Discussions

Interactive comment on "Air quality modelling using the Met Office Unified Model: model description and initial evaluation" *by* N. H. Savage et al.

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We would like to thank referee 1 for their comments which have helped improved the paper. Below we reply to each of the comments in turn.

1 General comments

It would be good to include rate constants and references to this list.

This has now been added in the revised version of the Supplement.

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It is unclear whether dry deposition is calculated online using actual meteorology, or whether a climatology is used.

We have clarified the text to make it clear that we calculate the deposition online.

The model seems to extend into the stratosphere (39 km altitude), which is rather high for an air quality model and possibly needs some explanation. I wonder what is the performance of stratospheric ozone, and how this may affect the surface concentrations.

As indicated now in Sect 2.1 this is an on-line air quality model and the top of the model was chosen in order to deliver the best performance in the meteorological modelling. We agree that it would be interesting to evaluate the performance for stratospheric ozone and the possibility of using a climatology instead. However, such analyses and how the modelling of stratospheric ozone affects the predictions of surface ozone are beyond the scope of this paper.

Additional information of the general model performance (and possible biases) of the chemistry scheme as well as the aerosol would help to assess reasons of specific performance of the model for RAQ purposes, and to assess which discrepancies can be related to issues regarding emissions and which to (specific) model assumptions. It would put the current model performance into perspective of the global chemistry modelling activities that have been performed with the same chemistry / aerosol scheme.

We agree that this would be an interesting study but it is beyond the scope of this paper to do comparisons with global models. Note that the objective of this paper is to present the description of a new model and a first evaluation.

With respect to the evaluation, I find the assessment of the model performance rather rudimentary. Partially this is a consequence of the nature of this paper. Nevertheless, I think the paper would become more valuable if some more spe-

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cific information, on, e.g., seasonal cycle, and/or more spatially discriminated performance would be presented. This can relatively simply be achieved.

We thank the referee for this comment. We have tried to address this without making the manuscript unnecessarily lengthy. We have included a new table (Table 3) to document the seasonal performance of the year-long forecast for surface ozone. In addition, we also show the diurnal cycle of the mean bias for this species (Fig. 2) and put some of the results (e.g. strongest positive bias at around 0500Z) in the context of the limitations of the model to represent some processes (see Section 4.1.2).

Also an attempt to explain specific model performance in Sect. 4 and all subsections would be helpful. The evaluation would further benefit from additional evaluation of other quantities and/or trace gases (e.g. AOD, ozone sondes, CO, aircraft observations, NO2 columns, . . .).

These additional observations would be very useful in helping to understand the reasons for specific errors. However, as the title of the paper indicates, this is only an initial evaluation of a new model and the validation we have conducted is limited to comparison against surface observations; we have added statements to this effect in the abstract and introduction

Finally, to my opinion the authors should be more critical in selecting appropriate figures to support their messages. I miss spatial maps of, e.g., O3, while there are a couple of soccer plots that do not yield helpful information. Time series are very helpful and clarifying, but not if one cannot see the data (as in Fig. 15).

We have reviewed the figures included in the manuscript and made some changes. We have removed two soccer plots for ozone during pollution episodes (former Figures 6 and 11), keeping only one soccer plot for the year-long forecast (Fig. 1). A new figure (Fig. 2) has been added to support some additional analyses in the paper. The previous Fig. 15 (hourly ozone from AQUM, MACC ensemble and observations) has been changed to show only daily ozone maxima (new Figure 17), which improves the

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visualisation.

2 Specific comments

P3134, l 8: "(NAE)": Please note the boundaries of the domain (E/W/N/S), and refer to fig. 9 for graphical interpretation

We still refer to Fig. 9 at the beginning of Section 2.1 to illustrate the domain of AQUM, but have added some additional text explaining that the model domain is a rotated grid coordinate system (hence it is does not have boundaries defined by simple E/W/N/S coordinates).

P3135 l 4: "40 transported species": Please provide a table with all species as used in the model, and additional tables which species are subject to wet/dry deposition respectively.

We have added this to the Supplement (see Table S1).

P3137 l 16:"Indirect effects are as described by Jones et al. (2001)." Do I understand correctly that aerosol composition is influencing meteorology? Is this assessed anywhere, and how does this relate to possible biases in the aerosol module?

The impacts of the feedbacks between aerosol and meteorology are not assessed in this paper and are a key topic for future research with this model.

P. 3138, l 5: GEMS/MACC data are used as LBC in AQUM. Biases in the LBC have been reported elsewhere (Schere et al., 2012), and could potentially influence the system. What is the impact of the use of these (aerosol/gas-phase) boundary conditions for the AQUM model performance?

We now discuss this issue in a new section (4.3) dedicated to the 'Sensitivity to chem-

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ical LBCs'; we include a reference to Schere et al. (2012) which is broadly consistent with our findings.

Section 2.6: Emissions. It would be good to state explicitly that no diurnal or seasonally varying emissions are applied as yet, if I understand this correctly. Would this possibly contribute to differences in ozone biases for urban and rural stations?

We have now modified Sect. 2.6 to make it clear that we include seasonal and diurnal variations in our emissions.

P3141, L4: The use of GFED aerosol emissions for the year 2000 seems rather arbitrary: It might be better to refrain from using such emissions, or otherwise use actual emission estimates. Also CO and NOx fire emissions are missing.

We recognise that this is a weakeness of the emissions data we have chosen. However, in the current model domain, biomass burning emissions have a relatively small impact.

P3141 L5: Biogenic isoprene emissions: What about soil NOx emissions? Furthermore, would the use of climatological emissions be a reason for model biases during specific events.

We do not include soil NOx emissions and have made this clear in Sect. 2.6, explaining that the impact in our domain is negligible. We do include seasonal as well as diurnal variation of isoprene emissions, as indicated now at the end of Sect. 2.6. We agree that the use of a climatology in the case of isoprene emissions might be partly responsible for biases during specific ozone events; that is the reason why we have recently developed interactive emissions of biogenic VOCs in the model and will use them in the future (see comment on emissions in Sect. 5).

Section 3. I miss a discussion of the temporal and/or spatial aggregation of model data. Why not include an assessment of the seasonal cycle? For ozone it

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could additionally be interesting to assess the diurnal cycle, and analyse daytime and nighttime biases separately.

We have expanded the information in Section 4.1.2 to include more discussion on this subject for ozone. We have added a new figure (Figure 2, diurnal cycle of bias) and a new table (Table 3, seasonal variation of model performance metrics)

Section 4.1.2: Ozone: it is good to see the general performance statistics as presented in Table 2 and the soccer plot, which are essential for operational air quality model purposes. Nevertheless, I miss a seasonal evaluation, a spatial map of the model performance and/or an assessment of daytime and nighttime O3 biases separately. Such additional information help to appreciate better why the model is performing as it is.

As indicated above: we have expanded the information in Section 4.1.2 to include more discussion on this subject, and have added the new Figure 2 and Table 3 to better illustrate the model's performance for ozone on a diurnal and seasonal basis, respectively.

Table 2 shows a relatively low hit rate for ozone on annual basis (0.57), while the model is biased high by about 10%. Contrary to the conclusions of the authors, this suggests that the dynamical range in model is not very large. In other words, the model ozone variability is relatively flat. Could you comment on this?

Actually this hit rate is relatively high; in Sect 4.4 and Table 6 we describe a comparison with the MACC ensemble which has a hit rate of 0.27. Even when the high hit rate of AQUM is partly related to the positive bias of the model for ozone, we also show that the variability in ozone from AQUM is close to that of the observations (the standard deviation of modelled and observed ozone are compared for this purpose). We have added a comparison of observation and model variability to Tables 2 to 7, plus a discussion of this at various places in the text (e.g. Sect 4.1.2, 4.2.1, 4.2.2, 4.4).

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P. 3147, l17. "molybdenum converters may overestimate the true concentration". Would it be possible to estimate a correction factor for this, based on (modelled) NOy concentrations, see, e.g., Lamsal et al., (2008) Additionally it would be interesting to evaluate the model against satellite NO2 observations.

Lamsal et al. (2008) quantified the interference of NO2 measurements using chemiluminescence analysers equipped with molybdenum converters at the time of the OMI overpass. To estimate the corrected NO2 concentrations they used the following correction factor (CF) calculated from simulated concentrations of NO2, sum of all alkyl nitrates (AN), peroxyacetyl nitrate (PAN) and nitric acid (HNO3):

$$CF = NO2/(NO2 + AN + 0.95PAN + 0.35HNO3)$$

This formulation is based on the high conversion efficiencies of molybdenum converter analysers to both AN and PAN found by previous laboratory experiments. However the quantification of the conversion for HNO3 is difficult since it is known to be deposited and evaporated on the inlet, processes which are temperature and humidity dependent. As a consequence, HNO3 interferences depend on the specific design and implementation of the inlet system and are subject to memory effects. Note that while Lamsal et al. (2008) found that a 35% conversion efficiency for HNO3 best resolved the discrepancies at the OMI overpass time, in a more recent study Lamsal et al. (2010) found that a 15% conversion efficiency for HNO3 yielded the best agreement with correction factors derived from observations in all seasons except in winter.

We appreciate the referee's suggestion and will consider it for future comparisons of modelled NO2 with surface observations from the AURN network. However, given the uncertainties in the quantification of the interference of molybdenum converters to HNO3, we believe that at this stage such kind of estimate would only bring uncertainties to the first evaluation of the model. We have made some changes to that part of the text and cited the mentioned analyses by Lamsal.

Regarding the evaluation of the model against satellite observations, that is the subject

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of ongoing work and we have added text in the abstract and introduction to clarify that this paper only concerns model validations against surface observations.

Section 4.1.4: The authors blame the biases in the model basically to uncertainties in the emissions. What are the reasons for this? There will be biases in the model too. Can this be quantified to some respect? How did the aerosol model perform in previous studies? Additionally it would be interesting to look at aerosol optical depth.

We have added an analysis of PM2.5 model performance to the paper (see Sect. 4.1.4, Table 2). The comparison of PM2.5 against PM10 performance provides evidence that the coarse component of PM (i.e. PM2.5 to10) is under-represented in the model. As we indicate in the revised version of the text, this is most probably due to insufficient emissions, although other processes in the model (e.g. dry deposition or transport) which act on particulates in this size range might also play a role. We agree that it would be interesting to have a look at AOD, but as stated previously, our comparisons in this paper are limited to the routine surface air quality network.

P. 3150, l. 4: "Figure 6 summarizes the good model performance. . ." I believe for this assessment it is more critical whether the model has desired hit and false alarm rates, and captures the event to a decent degree, as presented in Fig. 5. Statistics in Figure 6 is dominated by daily "background" performance rather than the event Therefore I don't think this figure adds necessary information to this situation, and could be removed.

We have removed the soccer plot previously shown in Figure 6.

P.3150, l. 6: "bias is particularly low": but the rms remains rather high, so this result is only low by chance.

We agree and have added a comment on the fact that the RMSE remains high.

P3150, l 26: "The model captures this first episode well": Is this not just by

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chance? In Fig. 7, other (smaller) events are not well captured, except for 26-27 July. What are differences?

We have added some text to acknowledge that the model did not capture all the peaks well. An analysis of why the model predicted some peaks and not others would require a level of detail which is neither possible nor appropriate here. Note that the aim of this manuscript is not to analyse a single pollution episode in detail.

P 3153, l. 1: "over-predicts values": Are there any reasons for this? How to align this with the results that the model in general is biased low?

The analysis we have added of PM2.5 model performance (see Sect. 4.1.4, Table 2) helps to clarify this point. We explain that this episode is dominated by secondary PM2.5, whereas the general low bias the reviewer refers to is in the coarse component of PM2.5-10.

P 3154, l. 2: Interesting results that the hit-rate in AQUM is significantly better than MACC, while the false alarm rate is not significantly worse. Is it possible to find a reason for this? Does this for instance suggest that the ensemble approach is mostly suitable for background conditions?

We have added some comments in the text discussing the reasons for this.

P. 3155, l. 14: "Likely due to emissions": I'm not convinced here. Are you sure that there are no model biases?

Repeating our comment above: We have added an analysis of PM2.5 model performance to the paper (see Sect. 4.1.4, Table 2). The comparison of PM2.5 against PM10 performance provides evidence that the coarse component of PM (i.e. PM2.5 to10) is under-represented in the model. As explained above, this is most probably due to insufficient emissions, although other physical processes in our model such as dry deposition or transport might also play a role.

In the outlook you mention the implementation of a new aerosol model. What do

you expect from this upgrade?

We have expanded the discussion in Section 5 to include more explanations of our aims with this upgrade and how we think it will improve performance.

Figure 11 may be removed, as this does not yield any relevant information that helps the reader.

We agree and have removed the figure.

Figure 13/14: Very interesting to see this type of evaluation. Why not merge the two figures into one, so that an assessment of the model performance is more easy?

A merged figure would have too many lines to read easily; we feel this is best left as two separate figures.

Figure 15: Please modify this figure, to improve readability. E.g. show daily max. O3 only.

We have followed this advice and modified the figure.

Interactive comment on Geosci. Model Dev. Discuss., 5, 3131, 2012.

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