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**Atmospheric Chemistry and Physics Discussions**

# *Interactive comment on* "3-D evaluation of tropospheric ozone simulations by an ensemble of regional Chemistry Transport Model" *by* D. Zyryanov et al.

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We thank reviewer #2 for his important remarks, which led to clarifications of several points in the revised manuscript. In the following, the same exercise has been done to answer remarks and comments of reviewer #2.

A. Major comments:

1. The authors put forward a few quite speculative reasons such as coarse horizontal resolution, uncertainties in long-range transport and emissions, and limitations of chemistry schemes to explain why RMSEs grow with altitude and why correlation appears to be the smallest near 8 km. I think that this is not enough. The authors need



to do more diagnostics. One thing that the authors can do is to examine the model meteorology at one or two sites within the domain using atmospheric soundings to see how good or bad the model temperature and winds compare with the observations.

Here the referee raises important questions that led us to conduct additional sensitivity tests. Concerning meteorology and especially winds that drive pollutant transport, it is likely that it have impact on RCTM behaviour. If it does not explain differences between models themselves (they almost all are using the same IFS meteorological forcing), it can explain part of the errors when compared to observations. The error diagnostics for the IFS system during the 2008 period are available. It is calculated by comparing 24h forecast to analysis.We present here some of these RMS maps. They show that errors on winds (u and v components) can be significant at northern midlatitudes with maximum errors between 200 and 400hPa in general ranging from 1 to 3.6 m.s-1 for the period of summer 2008. This corresponds also to altitudes where model errors can be high.

Comparing a limited number of radio soundings with one of the models will only give a very partial view of model errors. Probably, more systematic comparisons between models and soundings associated to sensitivity test to evaluate the impact on ozone fields will be necessary but this is a topic in itself and it is probably far beyond the scope of this paper. An important issue for such an analysis would be that the 4D var analysis system assures an optimal compromise between use of observations and the consistency of a well balanced wind field. This makes differences between the analysed wind fields and local observations unavoidable. We propose to add the following sentence in the section (4.1) were results are analysed. We mention results of the ECMWF verification procedure (available from the ECMWF teams directly) without adding figures to avoid lengthening of the text:

"Another explanation of increasing RMSE with altitude is related to the performances of the IFS itself. Indeed, systematic verifications of IFS performances are proposed at ECMWF. They show for this period at European latitude that RMSE (calculated by comparing 24h forecast to analysis) of both wind components are increasing with altitude with maximum errors occurring between 200 and 300hPa (in the jet stream region) and are ranging from 1 to 3.6 m.s-1. These errors in wind amplitude and direction can impact on ozone advection simulated by RCTMs. A detailed analysis of this issue goes beyond the scope of this paper. Note that IFS meteorology is input for all simulations in this study (directly or as boundary conditions for mesoscale models, thus it is expected that the impact on model errors is similar)"

Another thing that the authors can do is to perform some sensitivity experiments by changing the emissions rates of ozone precursors using one CTM. Since the ensemble of CTMs were run with various resolution and chemistry schemes but still showed more or less similar distributions in RMSE and correlations, deficiencies in model meteorology and uncertainties in emissions did appear to play important roles. I think that the suggested diagnostics may be able to sort those things out.

Emissions (of anthropogenic or biogenic origin) of ozone precursors (NOx and VOC) are mostly emitted at ground and do not affect too much the free tropospheric composition at the continental scale. This hypothesis is probably no longer valid when considering inter-hemispheric domains that allows more efficient vertical transport of pollutants. To confirm this, we first have made simulations with the CHIMERE model (for the whole 2008 summer) with 20% reduction and 20% increase of all emissions. These two sensitivity simulations shows clearly that discrepancies with the reference run remain mostly in the planetary boundary layer with differences always lesser than 10% at 2 kilometers height. This differences are rapidly decreasing with increasing altitude (<5% at 5km height). Also, in order to test a more realistic impact due to emission variations, we also have re-run the CHIMERE model with EMEP anthropogenic emissions instead of the TNO inventory used in the framework of GEMS and MACC projects. The use of this 2 alternative (and both well recognized) inventory allows to better catch the real uncertainty associated to anthropogenic emissions. For example mean discrepancies for NOx emissions are about 15% but with sometimes larger differences at specific locations (B. Bessagnet Personnal Comm.). Results shows same patterns than the previous sensitivity tests with differences in the ozone field concentrations lower than 10% at 2 km height and even about 80% of discrepancies lower than 5% at this altitude. Nevertheless, and as mentionned in the answer to reviewer 1, altitude emission associated to aircraft and deep convection are not directly taken into account by regional models. They are only represented in the IFS-MOZART system, probably with high uncertainty level. Now these aspects are discussed in the text in the following sentence of section 4.1:



"Concerning model errors it should be added that uncertainty in surface emissions inside the modelling probably domain does not play a significant role above the planetary boundary layer height. This is confirmed by sensitivity tests made with the CHIMERE model using either 20% increased/decreased surface emissions or the EMEP inventory (Vestreng et al, 2005) instead the TNO inventory. Indeed, corresponding changes in ozone concentrations were always below 10% within the first 2 km height, and below 5% above this altitude. On the other hand, we could imagine that altitude emissions produced either by lightning or aircraft could explain a part of model error. The regional models of this study do not represent these emissions; they are only taken into account in IFS-MOZART and it is also well-known that these processes are still not well characterised. Nevertheless, Due to the low residence time of air masses in the free troposphere within the model domain (of the order of several days) and small ozone production rates there, lightning NOx and aircraft emission over Europe are not expected to significantly impact European free tropospheric ozone levels."

2. The authors suggest that the somehow poor performance of CTMs at some of the western stations may be related to the boundary conditions. Though it is a logic way to think this way, I am not convinced that the boundary conditions impact the western stations much more than the interior stations. I am not sure how far away the western stations are from the model boundaries in grid numbers but I think that nowadays both CTMs and meteorological models are pretty good at dealing with boundary discontinu-

ities and noises. Even though the models produce some unwanted waves along the boundaries, the waves would eventually impact the interior stations as much as they impact the stations near the boundaries. I think that the relatively poor performance at some of the western stations may be related to the land-sea contrast and mountainous terrain that induce mesoscale circulations. These circulations would control the transport and dispersion of the pollutants but the models may not resolve the circulations well as the authors briefly mentioned. I would like the authors to discuss more along this line.

We agree with the fact that local meteorology can impact the ozone fields and we now the limitation of models to represent some of these features such as land/sea breeze or mountain venting. We agree to mention this in the text when analysing results at "western" station in the following sentence:

"However, in the case of Valentia and Lerwick that are coastal stations, we can not exclude a systematic misrepresentation of local meteorological patterns such as land/sea breeze by the models."

Nevertheless, it is also true that the distance of the stations from the domain boundary is an important factor for the impact of boundary conditions of chemical species. Szopa et al (2009) have clearly shown that the stations close to the western and northern model boundaries were much more influenced by boundary conditions than more continental stations due to the distance to boundaries but also due to the impact of surroundings continental emissions and dry deposition controlling the ozone concentration at these latter sites much more than large scale-advection. The consequence is that these regional models generally perform better for these continental stations due to a better characterisation of emissions and their impact than the ozone loading in Atlantic air masses.

Since Frankfurt had nearly two vertical profiles per day during the study period, the authors may be able to compare the simulations and observations at different times of



the day and see if the CTMs perform differently, which may indicate some model issues with representing day/night or early morning/late afternoon circulations.

Such approach could be interesting to inspect the behaviour of model in the boundary layer especially to evaluate the vertical distribution of pollutants (as well as associated chemistry) and its evolution during the day. Evaluate these aspects using only MOZAIC data is difficult because of the specificity of airport and their direct environment (very high punctual emission sources). Probably such work should be extended to available surface stations. If interesting, it is also beyond the scope of this paper.

3. Summertime corresponds to strong forcing at surface that tends to generate convective clouds and circulations. I am wondering if the authors ever looked at the satellites images and atmospheric soundings during the months to see how high the convective clouds might be (i.e., use satellite "measured" cloud top temperature and compare with the soundings to determine the cloud top). If the top of the clouds were located around 8 km, then we might be able to explain the C-shaped structure of correlation since numerical models still can't handle clouds good enough.

Indeed, these aspects need also some comments in the paper. Once more this aspect is a topic in itself and the quick inspection of satellite imagery does not reveal clear features. Moreover, considering the generally sparse character (at continental scale) of deep convection at mid-latitude, it is difficult to catch its signature on radiosounding without a really close inspection of all available data. Another difficult point is the real impact on ozone fields of deep convective cloud, it is a mix of vertical transport of air masses that can be richer or poorer (compared to free troposphere air masses) in ozone concentrations, of reduced/or enhanced photochemistry, vertical transport of precursor, production of NOx by lightning. This makes difficult the identification of a clear case of deep convective impact and even though it is difficult to draw real conclusion from such a potential single case. Authors choose to mention the potential impact of deep convection with the following sentence:

“Besides producing NOx via lightning activity, deep convection can also alter the redistribution of ozone and its precursors (Lawrence et al, 2005). Colette et al (2005) have shown that 10% of ozone rich-layer in the European free troposphere could have been uplift by convection from PBL. Nevertheless, if taken into account in models the parameterisation of such processes and their impact still remains highly uncertain. ”

Moreover, we have conducted a sensitivity test by comparing two summer simulations with and without the deep convection module activated in the CHIMERE model. If a difference occurs, it is not significant representing less than 5% compared to the reference run and almost no impact on comparisons with observations (whatever the altitudes are). It shows that differences in convective parameterizations can not explain discrepancies between models themselves. It shows that activating the convective paramerization has almost no impact on ozone fields. Knowing the difficulty to simulate deep convection it does not mean that the real deep convective processes has no impact on ozone fields and then on model errors. A sentence mentioning this sensitivity test is also added in the text (section 4.1):

“It should be noted that results of CHIMERE ozone simulations (made over the whole 2008 summer) where deep convection has been by switch off does not show significant differences (always less than few percent) compared to the results obtained with the parameterisation included.”

B. Minor comments:

1. The authors use “planetary boundary layer”, “free troposphere”, and “upper troposphere”to denote the three layers between 0-2 km, 2-8 km and 8-10 km in the vertical. I would suggest the authors to use “middle troposphere” instead of “free troposphere” since in meteorology, “free troposphere” refers to the layer above the planetary boundary layer which includes the upper troposphere.

We agree with this. The text has been updated in this way.



2. Along this line, I am wondering what is the reason behind the classification of these three layers in the vertical. Is it due to changes in ozone concentrations or different dynamic and chemical processes? The authors need to make this clear in the beginning of the paper.

We agree with this. The following sentence has been added in the beginning of section 4.1:

“In the following, results are presented as a function of tree ranges of altitude: 1) the Planetary Boundary layer (PBL, 0-2 km height); 2) the middle troposphere (MT, 2-8 km height); the upper troposphere (UT, 8-10 km height). It allows being more synthetic to present these results and to take into account the main differences in processes driving ozone concentrations as a function of altitude (i.e surface emissions, “fast” chemistry and dry deposition associated to turbulent transport in the PBL, horizontal transport and “slow” chemistry in the MT and UTLS exchange processes in the upper troposphere).”

3. I am wondering how the model runs were set up, i.e., daily restart or continuous runs from June 1 through August 31, 2008? How often the boundary (meteorological and chemical) conditions were updated?

In this case, because the exercise has been set-up in hindcast mode (It is a resimulation of this period), all runs has been made in a continuous mode but some specific unwanted stop maybe happened. We are not aware of this potential “accident”. In this case it is likely that model have been restarted using previous simulated state of the atmosphere to avoid any “spin-up effect”. The meteorological boundary conditions are available every 3 hours in the IFS system (in fact every 6 hours for analysis and every 3 hours for forecasts). This information is now added in the text (section of model description). The chemical boundary conditions are available every hours either from IFS-MOZART either from MOCAGE. This information is already present in the text.

4. The authors used MOZART-IFS at times and IFS-MOZART at other times. Please be consistent throughout the paper. If the authors do need to use MOZART-IFS and IFS-MOZART separately, please explain the differences.

We now use IFS-MOZART homogeneously in the text.

5 Acronyms should be defined upfront. I would suggest the authors to have a table in the introduction section with all the acronyms defined.

A table with acronyms has been added upfront as suggested. We have also check in the text that all abbreviations has been defined.

6. I would suggest to change ”associated to” to “associated with”, and “participate to” to “participate in” throughout the paper. 7. Please change “through” on Pages 3, 24, 25, and 26 to “trough” (i.e., ridge and trough not ridge and through). 8. Please improve the writing of the paper by paying attention to details. For example, on Page 3 Line 18, “during summer is well catched” should be “during summer is well captured”; on Page 5 Line 18, “It is proposed here is to conduct” should be “What is proposed here is to conduct”; on Page 16 Line 16, the second “either as” should be “or as”; on Page 21, Line 7, “on Fig. 4” should “in Fig. 4”; on Page 27 Line 1, “to catch to a full extent” should better be “to capture to a full extent”.

All these suggestions (6 to 8) have been taken into account in the new version of the text.

Figure Captions

Figure 1. Zonal mean forecast errors (RMS) at D+1 term (calculated against corresponding analysis) calculated over the whole summer (JJA) 2008 for meridian wind component simulated by the IFS system. Vertical axe represents pressure (hPa). To access values or at least ranges of error’s values multiply the boundary of the painted scale by the unit (0.1 m.s-1).

Figure 2. Zonal mean forecast errors (RMS) at D+1 term (calculated against corre-



sponding analysis) calculated over the whole summer (JJA) 2008 for zonal wind component simulated by the IFS system. Vertical axe represents pressure (hPa). To access values or at least ranges of error’s values multiply the boundary of the painted scale by the unit (0.1 m.s-1).

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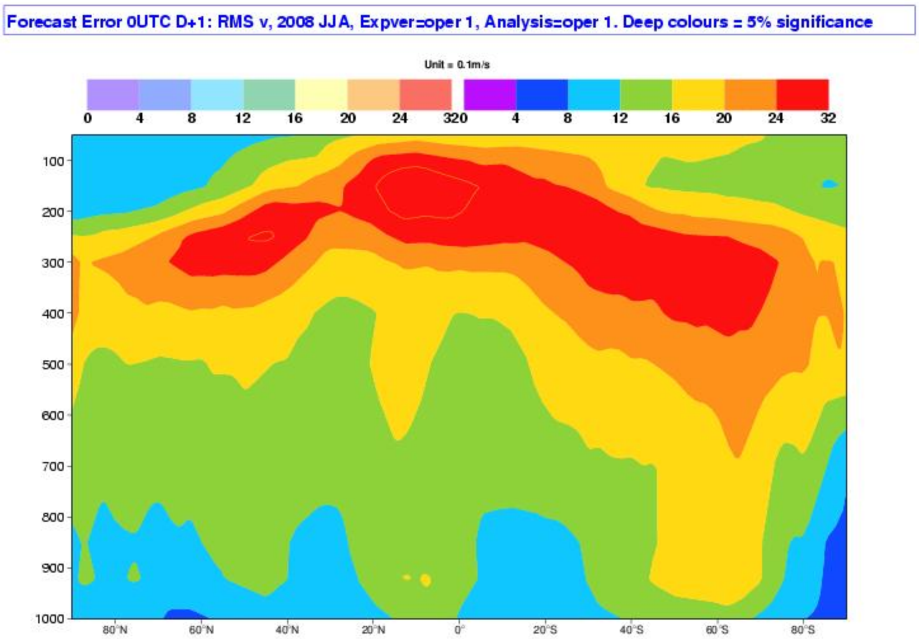


**Fig. 1.** cf Figure captions



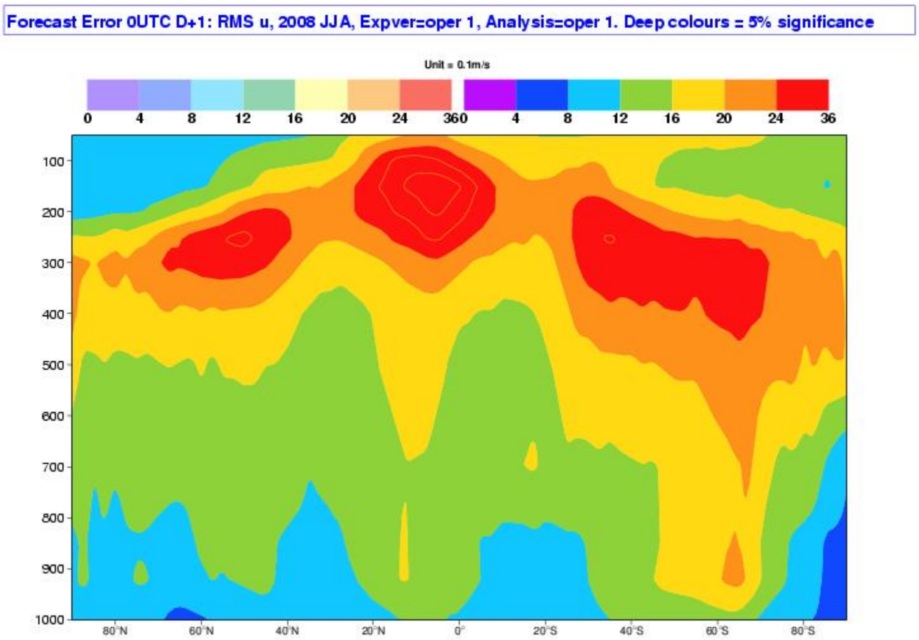


**Fig. 2.** cf Figure captions