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Geoscientific Model Development Discussions
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Interactive comment on "High resolution air quality simulation over Europe with the chemistry transport model CHIMERE" *by* E. Terrenoire et al.

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General update

In the submitted version of the paper, the model resolution is referred as 7 km. This estimate was based on the geographical degree coordinates of the grid cells size (0.125° x 0.0625°) which are actually closer to 8 km at the centre of the domain. This new estimate is now used throughout the revised version of the paper.

First, we would like to thanks the Referee #3 for its constructive remarks and suggestions. Here are the different answers regarding the Referee #3 comments:

GENERAL COMMENTS

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Interactive Discussion
Discussion Paper

The present paper shows the results of an operational evaluation of an improve version of the CHIMERE chemical-transport model (version 2009) for the year 2009 over Europe. The CHIMERE CTM (7 km x 7 km horizontal resolution) is compared against rural and urban background stations from the EMEP and AIRBASE monitoring networks based on O3, NO2, PM10 and PM2.5 and PM components. The present work could be of interest to the readers of GMD because it tries to contribute to increase the scientific knowledge about air quality modelling. From my point of view, the novelty of this paper lies in the following facts: (1) the annual modelling of air quality in Europe at urban scale (7 km x 7 km), (2) new methodologies in emission modelling, and (3) testing new parameterization and new meteorological data. However, the implication of these facts has not been explicitly quantified in each case. Furthermore, I miss a detail description of the used methodology and a discussion about that with the objective that other users could reproduce it. Even more, the analysis of the evaluation of the results is very descriptive, and does not show anything new about deficiencies in the model. From the format point of view, author should make efforts to highly improve the format of the article (see large list of technical correction above). In this sense, I recommend the publication of this work after the authors provide a more detailed methodology and a deep discussion of the results, and they improve the format of the paper (typos, acronyms, figures, etc.).

The method section have been detailed and now includes a description of the CHIMERE version used for the study, the meteorology data used (including the urban correction), the methodology for the preparation of the anthropogenic emissions (including the SNAP2 temporal modulation), the observation data used and a description of the data analysis methodology.

In order to make it clearer we address the main results in a more concise way. The description of model performance is done by species and if the agreement is poor, we address explanations for this behaviour.

A separate discussion section has been added. It discusses, firstly about the model

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developments including the urban meteorology correction. Then, more explanations are proposed to explain the underestimation NO2. Finally, it discusses about the performance of the model for PM and the related SIA, dust, sea salt and SOA.

As suggested by the Referee, the format of the article has been improved following the list of technical corrections suggested by the referee.

SPECIFIC COMMENTS

Abstract

P4138, line 1: The abstract should be rewritten. It is too generic. It should indicate the name and version of the model and include the novelties of the work.

The abstract has been rewritten.

Introduction

P4138, line 20: Introduction. Since you are running a model at urban scale, your introduction should put in context your resolution with other studies already done over Europe and explain the important of increasing resolution. P4138, line 22: when you say “coarse horizontal”, which rage of resolution are you talking about? Provide some references. P4139, line 6: when you talk about GMES and GMES-MACC, are they used as examples of forecast applications, or are they other application? Rephrase this paragraph accordingly. GMES and GMES-MACC are acronyms and they should be described. P4140, line 9: replace “assessment” by “evaluation”.

The introduction has been rewritten and the motivations for high resolution simulations are clearly identified. We also provide references about the performance of other studies when increasing resolution and we mention recent comparisons studies that can compared to our work.

Model description

P4140, line 23: 20 m is mean level or full level? Could you clarify this, please? In this

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sense, how many vertical levels are you using? Which is the top of your configuration?

Clarifications about the description are done in the revised version of the paper. We use 8 vertical levels from the ground to 500hPa. 20 m is the full level.

P4140, line 26: which version of the MEGAN model are you using? Are you assimilating all the species which come from MEGAN? How do you perform speciation for MEGAN species to MELCHIOR chemical mechanism?

MEGAN v2.04 is used to calculate biogenic emission of the six species (isoprene, α-pinene, β-pinene, limonene, ocimene, and NO) that are then used by the chemical mechanism (MELCHIOR2) of CHIMERE. For more detail the reader is referred to the Menut et al. (2013) paper.

Meteorology

P4141: in the section of meteorology authors describe a sensitivity test using IFS vs. WRF, but you do not specify the version of WRF, the number of stations used for the test nor quantified the improvement of using IFS instead of WRF. Also author only focus on wind direction, what about other meteo variable such asT2M and PBL performance?

This section was modified. The motivations for the meteorological driver are now textual and we added some more references.

P4141, line10-11: how does the urban correction for Kz is applied? Which is the variability of Kz between urban and rural areas? You evaluate this correction in terms of air quality, but what about in terms of wind speed? Why this correction for Kz has more impact for PM than for gases? Have you see any different between PM10 and PM2.5?

The urban correction is described in the meteorology section. The discussion part described the impact of urban correction for Kz and the wind speed for four species (O3, NO2, PM10 and PM25) but some suggestions done by the Referee would need further investigations that should be done in another paper that would look at in more

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details the impact of the urban correction.

Anthropogenic emissions

P4142, line 25: Which is the reference year for the emission database used? Please, used EMEP reference corresponding to the report describing the emission you used (usually they have a report for each emission database). P4143, line 1-11: I think it is well known. Maybe, you could merge this information with table 1. I miss some information about speciation of emission to the chemical mechanism used. If you used MEGAN for biogenic emission, are you double counting emission if you take into account SNAP 11? P4144, line 3: which version of CHIMERE emission pre-processor are you using or based on? Please, include some information about that in the model configuration.

The section related to the spatial re-griding of the anthropogenic emission has been rewrite to make it clearer. SNAP11 are indeed replaced by the MEGAN model.

Chemical speciation

P4145, line 10-20: Which chemical mechanism are you using? I guess it is MELCHIOR, but which version? In this section and in the same paragraph authors mix information about temporal disaggregation, it is a little bit confusing.

Tropospheric gas phase chemistry is represented using the reduced MELCHIOR chemical mechanism (120 reactions and 44 gaseous species).

SNAP 2 emission temporal modulation

P4145, line 22-26: I do not like the way these two new concepts, degree day indicator (Dj) and the daily modulation factor (Fj), are introduced in the text. It is confusing and hard to follow. Some units are missing and some constants appear without any justification. For example, what about A coefficient? How do you fix this value? Please, number equations.

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The definition of degree days and the corresponding modulation factor have been rewritten.

Observation data

P4147, line 2-17: What about measurement for SO2? Why do authors omit this pollutant in the evaluation of CHIMERE?

The SO2 boxplot time series were added to the revised version of the paper and analysed in the model evaluation section.

P4147, line 2-17: In the same line, if the authors evaluate PM components, why organic carbon or elemental carbon is not evaluate? At least there is four stations providing measurement for black carbon in the EMEP network.

We agree with the referee comment, unfortunately organic carbon or elemental carbon were not available as single species in the CHIMERE version used for the study.

P4147, line 2-17: Looking at Table 3, any EMEP ozone station is used in your evaluation. However, you sed NO2 data from this network. Any explanation for that?

In the study we did not used NO2 data from the EMEP network and NO2 was removed from table 3.

Model results

P4148, line 6-13: When you compare chemical components for PM, which range of diameter are you using? PM10 or PM2.5? Some measurements do not take into account the size of the aerosol, just chemistry.

The range of diameter used for the EMEP measurement is PM10 as explain the section 2.4.

Nitrogen dioxide

P4148, line21-25: Could you provide more reasons to justify that NO2 underestimation

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in UB is due to emission lack? Could you provide reference to support that? Furthermore, later in P4149, line 10-13 you say that the reasons are unclear. So, in the end everything is confusing. I guess to go more in detail with NO2 urban underestimation author should study NO2 daily cycle to see when the underestimations take place, maybe it could be a problem of PBL, and mixing ratio. Please, try to verify your hypothesis.

NO2 underestimation ranges between -34% and -54% at RB and UB Airbase stations. Considering that: 1) BIAS in NO2 mean values is generally higher than the corresponding bias in the standard deviation 2) Ozone is overestimated, implying that NOX titration is not a limiting effect 3) PM2.5 and PM10 show a less relevant underestimation, stating that meteorology can not be considered the only reason for model discrepancies 4) BIAS at urban stations is clearly stronger than rural sites we can reasonably assume that NOX are proabably underestimated. Of course, we agree that the possible emission underestimation is not the only reason explaining NO2 discrepancies.

Ozone

P4149, line 22: to be consistent, use RB for rural as you use UB site.

This correction was done in the revised version of the paper.

P4150, line 1-2: the highest overestimations are found for autumn at RB and UB sites,in relative (30.6 and 36.9%, respectively) and absolute values, not in winter. Any explanation for that?

We agree with the referee comment. In order to explain this overestimation of O3 in the autumn, we used the timeseries from the remote station of Valentia. It shows at this station that the overestimation is maximal during the autumn thus indicating that the overestimation of ozone during this season is very likely caused by the overestimation of O3 at the lateral boundaries of the domain.

P4150, line 3: replace “maxima” by “maximum concentration”. Instead of winter, you

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could say the colder seasons.

This correction was done in the revised version of the paper.

P4150, line 5-7: CHIMERE overestimates concentration at rural background sites. Has the O3 boundary condition from LMDz any impact? Have any explanation for that overestimation. Did you check how the O3 daily cycle? Any idea about how the model performance during the day/night?

The daily cycle of O3 was analysed and shows a higher positive bias (9 ppb) during the morning (7 am) than during the afternoon ozone peak (5 ppb). This tendency is likely to be also related to the lack of O3 titration by NO2 due to the previously described underestimation of NO2.

PM10 and PM2.5

PM10 and PM2.5: this section should be rewritten. Evoid adjective as “good”, “worse”. Have you think about measurements uncertainty? Please discuss more in detail the source of uncertainty in your evaluation.

This section was substantially modified and the source of the underestimation of PM was analysed and discussed. We also add a paragraph about the uncertainty of PM measurement in the section 2.4.

P4150, lines 26 - P4150, lines 6 make reference to Table 6, corresponding to EMEP data. But in the text you say they are airbase station. The message is confusing, you are using two networks but what are they telling you? Also units at P4151, line 3-4 should be ugm-3.

The sentence was rewritten to make it clearer and the unit at P4151, line 3-4 was corrected.

P4151, lines10-11: this is not true, because FBs for PM2.5 at EMEP sites are bigger than PM10.

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Yes, correct. PM2.5 overestimation is probably due to the corresponding overestimation of sulphate. Conversely, sulphate overestimation is less visible in PM10 scores, due to the compensating underestimation of the PM coarse.

Sulphate

P4152, line 16: replace “Valencia” by “Valentia”

The correction was done in the revised version of the paper.

Particulate and total nitrate

P4152, line 23: replace “Nitrogen dioxide (NO2)” by “Ammonia (NH3)”

The correction was done in the revised version of the paper.

P4154, line 1-2: this sentence means that nitrate underestimation is due to NOx emission underestimation. NO2 Emission underestimation is not well demonstrated in previous section. Please, go more in detail with the reason why NO3 is underestimated.

TNO3 bias is comparable to NO2 bias, at least at UB stations (that represent about 70% of the available sites). This indicates that the chemical pathway of oxidised nitrogen, from NO to HNO3 is correctly balanced with respect to observed values, suggesting that the limiting factor in nitrate (HNO3 + NO3-) production is the availability of NO2 and in turn of NOX emissions Finally, it is worth noting that NO3 and TNO3 observed mean values are more similar than the corresponding modelled concentrations. This means that in the observed values the equilibrium between HNO3 and nitrate is more shifted toward the aerosol phase than the modelled one. This is probably related to the higher availability of modelled sulphate (overestimated by CHIMERE) limiting the conversion of HNO3 into aerosol phase, hence explaining the worsening in model performance when aerosol nitrate alone is considered.

Spatio-temporal variability of the modelled concentration fields

This section (3.1) is now located before the model evaluation section (3.2).

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P4155, line 26-28: O3 shows high concentration over the sea because the low dry deposition over the oceans. Have a look at O3 dry deposition in CHIMERE.

We agree and this point was added in the revised version of the paper.

P4156, line 1-2: also along shipping routes along northern Europe and Strait of Gibraltar.

This point was added in the revised version of the paper.

P4156, line 6: please quantify “nicely modelled” and “slight underestimation”.

The sentence was rewritten in order to quantify the performance of the model

P4156, line 12-13: the mean concentrations of PM10 in the maps do not reach 40 ug/m3. How desert dust are implemented in the boundaries? Could you say anything about the contribution of sea salt in PM10?

Figure 7 shows that PM10 reach 36 μg/m3 over the south part of the domain. Desert dust are conditions of the mother domain are monthly mean climatology taken from the GOCART model (see section 2.1). Sea salt winter and summer mean maps were added to the revised version of the paper. For the sea salt, the maxima are met during the winter and are located over the North Sea (up to 13 μg/m3) and represent the major part of PM10 mass. Over land a zonal gradient is observed the maxima modelled over Western Europe (5 μg/m3 over Ireland) and the minima over eastern Europe (<1 μg/m3) where the influence of oceanic winds caring sea salt particles is lower than over the west of Europe.

P4156, line 20-24: the complex topography of the Po valley together with a complex emission pattern are reasons of the high concentration of PM10 (and PM2.5). These conditions generate a complex situation where the models present difficulties to reproduce correctly the air quality dynamic there.

We agree with the Referee.

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P4157, line 5-11: how is the level of confidence in SO2 pattern? You have not evaluated the concentration with available EMEP measurements.

The SO2 yearly boxplot time series were added to the revised version of the paper and analysed in the model evaluation section.

P4157, line 8-9: any explanation for this maximum along African coast? shipping?

Yes, we detailed in the text that SO2 emissions from shipping, often confined in the shallow marine boundary layer can favour the formation of sulphate during summer through radical oxidation of SO2 to H2SO4.

P4157, line 9-11: also due to the meteorological condition in summer which favour high temperature and accumulation and recirculation of pollutant in the Mediterranean basin.

Yes, we agree with the referee. This point was added in the revised version of the paper.

Conclusions

P4157, line 21: replace "(7 x 7 km)" by "(7 km x 7 km)". Check it in the whole manuscript.

In the submitted version of the paper, the model resolution is referred as 7 km. This estimate was based on the geographical degree coordinates of the grid cells size (0.125° x 0.0625°) which are actually closer to 8 km at the centre of the domain. This new estimate is now used throughout the revised version of the paper.

P4158, line 24-26 and P4158, line 1-2: the implication of each of the improvements implements should be discuss more in detail. Overall, conclusions are poor. In terms of CHIMERE community, which are the benefits respect previous CHIMERE versions? Which are your recommendations about horizontal resolution?

The conclusion has been rewritten in order to motivate the points for further research.

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The main degree of improvement is seen using the urban correction and is describe in the discussion section as well as other model improvements.

TECHNICAL CORRECTIONS

Used correlation coefficient instead of correlation index. P4139, line 9: replace “assessment” by “evaluation”. P4139, line 21: replace “Particles” by “particulate matter”. The same for line 22. P4140, line 8: replace: “module; Vautard et al.” by “module and Vautard et al.”. P4140, line 18: replace: “whole of Europe” by “whole Europe”. P4140, line 20: replace: “0.125 x 0.0625_” by “0.125 x 0.0625_” (7 km x 7 km). P4140, line 22: replace: “Data for comparison” by “Modelled concentration for comparison”. P4141, line 4: describe the ECMWF-IFS acronym. P4142, line 1: replace “(Kz coefficient)” by “(dispersion coefficient, Kz)”. Consequently, replace: “dispersion coefficient (Kz)” by “Kz”. P4142, line 17: insert a “,” after “However” P4142, line 22: the tonne the International System of Units (SI) is used as “t”. P4143, line 20: replace: “US Geological Surve (USGS) Land Uses database” by “US Geological Surve (USGS) land-use database”. P4143, line 21: replace “Twenty height” by “Twenty eight” P4144, line 5: replace “SNAP 2 emissions sector” by “SNAP 2 emissions”. P4144, line 7: replace “0.08 3 x 0.083_” by “0.083_ x 0.083_” P4144, line 8: PPM acronym is not defined P4144, line 21-23: the sentence “The picture show the . . .” is repeat (see line 20-21). P4145, line 16-20: check the punctuation. P4147, line 2 and line 6: be careful with “Airbase” or “AIRBASE”. Be consistent in the whole document, also in figures and tables. P4147, line 8-9: I suggest: “The spatial distribution of the stations” instead of “The stations spatial repartition” P4147, line 16: remove “s” in “different measurements techniques” References omitted in the Reference section: P4147, line 19: Dennis et al. (2010) is omitted in Reference section. P4147, line 23: replace “, agreement” by “index of agreement”. P4147, line 23: use correlation coefficient instead of correlation index in the whole document. P4159 and P4160: This section “Appendix A” is not relevant since description of statistic is well known. As you use AEMET tool for calculating you statistics, I recommend you just reference AEMET software. However,

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you are using thresholds in the observation when you calculate the statistics. I think it is worth mentioning this fact in the section 2.5 Data analysis methodology, and even explain how these thresholds have been chosen. Table 1: To be consistent whit the text, use SNAP 1, SNAP 2, etc instead of S1, S2, etc. Table 2: To be consistent with the text, use SNAP 1, SNAP 2, etc instead of S1, S2, etc. I guess these values are in %. Table 3: Replace “TNH3” by “TNH4“. Describe what “UB”,“RB”, and “X” mean. Be consistent with “AIRBASE” name along the manuscript. Table 4: Please indicate the units for standard deviation of the observation and modelled concentration. The same for RMSE. Replace “Automn” by “Autumn”. Be consistent along the test with “Root Mean Square Error”, capital letter or lowercase? Replace “PM25” by “PM2.5” Table 5: Replace “PM25” by “PM2.5” Table 6. In Table 3 you have said you evaluate NO2 from EMEP, but in this table this pollutant does not appear. I guess you use EMEP just for chemical composition, in that case remove NO2 for Table 3. However, in my opinion, it is hard evaluate PM components without evaluate the gas precursors. This could help you in the analysis of your results. Figure 1: Indicate the number of station, the date, if the evaluation is hourly or daily. Figure 2: remover “for four main pollutants” Figure 3: Be careful with subscripts for NOx, SOx, and NH3. Replace “profils” by “profiles”. Tonnes in the International System of Units is “t”. Please, when you use EMEP in this case, use as “EMEP emission database” because you have also EMEP measurements. In this flowchart, where the vertical disaggregation is? Figure 4: be consistent between units in y-axis "kg/inhab.” (in the graphic) or “kginb-1 yr-1” (in the caption). Figure 5: units are reference to the year 2009. Please indicate that. Figure 6: replace in the caption “emission” by “emissions”. Use subscript for Fj. Figure 7: Move the title of the figure to the caption. Figure 12: Use subscript for pollution in figure titles. Figure 13: For O3, the summer is right and winter is the left. Please correct the figure or the caption. Figure 14: For PM10 and PM2.5 the summer pattern is the right column. Please correct the figures of the caption.

All the technical corrections proposed by the Referee were taken into consideration in the revised version of the paper. An Appendix was put as online supplementary

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material. The thresholds have been derived empirically having a look at observed time series. They have been selected in order to preserve as much as possible the original observed data.

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