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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 for its constructive remarks and suggestions. Here are the different answers regarding the Referee #1 comments.

Chemistry transport models play a key role in the definition emission control strategies. It is therefore of great importance to have well validated models that operate at

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the appropriate scale for its purpose. In air quality modeling a tendency to operate models at increasing resolution is observed. The study at hand describes the results of a CHIMERE model simulation at 7 Km resolution across Europe. Although validation studies are very useful and deserve publication, I have a number of concerns that needs to be addressed before this paper is acceptable for publication. First, the paper lacks a firm discussion of the results. Moreover, at many locations the explanation of the model behaviour could be formulated more concise. I would recommend to include a separate discussion section in which the (at least) the following topics are discussed: a) Is the model performance of PM as robust as suggested? b) Shortcomings in SIA modelling c) Modelling of dust d) Modelling of (S)OA e) Choice of emission data, shortcomings, impact of downscaling approach f) impact of measurement location on performance assessment g) Does the increase of resolution provide a better performance? h) Do the model developments from this study improve the performance? i) Impact of urban meteorology correction (move from method section) j) Are there other studies that indicate that NOx emissions in Europe are underestimated? Can there be other explanations for the NO2 underestimation? Following the suggestion of the Referee, we have reinforced the discussion of the results throughout the paper and in order to make it clearer, in the result section, we address the main results in a more concise way. A separate discussion section has been added. It discusses, firstly about the model 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.

In this way the three main areas for further improvement identified in the conclusions can be motivated (and maybe extended?). The topics are discussed in more detail below. Second, the paper shows that the CHIMERE model performs quite well for total particulate mass, whereas obvious shortcomings in the modeling of secondary inorganic aerosols are shown. Moreover, I would suggest to provide and discuss the modelled distributions for dust, sea salt and (secondary) organic aerosol. These components explain a large part of modeled mass but model parameterizations or source

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terms are very uncertain. I wonder if the statements on the model performance for PM should not be weakened considering these issues. Also, I would advise to put chapter 4 before chapter 3 to present the distributions before the validation. The modelled spatial distributions for dust, sea salt and secondary organic aerosol are now provided, described and discussed. We agree with the fact that they are shortcomings in the modelling of secondary inorganic aerosol however we wish to underline the good performance of the model for total particulate mass especially for PM2.5. As suggested the spatio-temporal variability of the modelled concentrations section was put before the model evaluation.

Third, the introduction is quite sparse. I think the research questions can be better motivated. Moreover, the citation of other studies (excluding those of the CHIMERE team) throughout the paper is quite thin.

The introduction has been rewritten to better address the research questions and more references have been added throughout the paper.

Finally, the emission gridding performed in this study is not really new. Please explain why the MACC data were not used as input to this study. This database was available and avoids some of the problems that are now included in the runs here. The emission gridding section has been modified to better explain the methodology used in the study. The emission data used for this study included MACC and EMEP data indeed.

Detailed remarks:

Introduction:

Please introduce why CTMs are going to higher resolutions. Do other studies see improvements in performance when increasing resolution?

P4139, line 4: the Collette paper deals with regional trends P4139, Line 5: GEMS = GMES P4139, line 12. Please mention the comparison studies themselves. The introduction has been rewritten and the motivations for high resolution simulations are

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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:

Please specify the version number, methodology for wet and dry deposition, vertical extend, and upper boundary conditions. The model description now include the version number, vertical extend and the upper boundary conditions used. For a detail description of the CHIMERE parameterisations the reader is reported to the Menut et al. 2013 paper.

P4140, line 13: the wording suggest that you have a nest in a larger CHIMERE simulation? True? Yes, it is right. However as the boundaries of the mother domain are close to the nested domain, its boundary conditions are strongly influenced by the climatology dataset used at the boundary of the mother domain.

P4140, line 18: the specified domain does not cover the whole Europe. It is right and we now used “most of Europe” in the revised version of the paper.

Meteorology:

First paragraph: why is the WRF and ECMWF data compared in this paper? It was not one of the model developments listed in the conclusions. Maybe a textual description is enough. We agree with the Referee, the motivations for the meteorological driver are now textual and we added some more references.

P4142. Line 8-19: here a description of results is given. Maybe this could be moved to the discussion section? This part describing the impact of the urban correction has been moved to the discussion part.

P4143, line 11: SNAP 11 also includes sinks? Isn’t snap 11 normally used for natural emissions? It is correct, the SNAP 11 used for the study correspond to the natural emission that are produced using the MEGAN model.

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P4143: The spatial regridding is performed keeping the 0.5x0.5 degree emission total fixed? This would mean that also uncertainties present in the EMEP gridding are introduced in the high resolution emissions used here. Does this mean that all anthropogenic emissions (except agriculture) is put on urban areas? So not on road segments, rivers, canals, industrial locations, etc? The section related to the spatial re-griding of the anthropogenic emission has been rewrite to make it clearer.

P4144: line 1. Do I understand correctly that population is not used? Or? Yes, it is correct. Population data were used just for SNAP2

Line 9.Please state more clearly if you used the function representative for France to distribute SNAP2 emissions outside the large urban areas in all countries? I am confused by the text. Please discuss how representative the function may be. . . The underlying hypothesis in applying over the whole Europe such function, derived just from French data, supposes that the relationship between the “use of wood” (i.e the fraction of wood in fuel mixture) and the population density is pretty constant across Europe. This is not surely true, but we think it can be considered as a first guess approach valid for the whole domain. It is worth noting that we did not use this function to estimate the emission load, but just to distribute it in each cell.

If you use the function, what happens when you keep the total of the EMEP cell fixed? Wouldn’t the Paris SNAP2 emissions return back to Paris as there is no other city on that 0.5x0.5 box? Yes, if in the EMEP cell, that is the provided emission we must split over a set of smaller computational cells, includes just the Paris city, all the emissions accounted for that EMEP cell will go just over the computational cells covering the Paris area. This is correct, because, in this case, the Paris EMEP emissions just contribute to the fine scale Paris emissions.

Shouldn’t you redistribute the SNAP2 emissions from the country total and allow emissions to shift across the country? Or did you do that? No, we did not do that. Of course this is a possible solution, but we preferred starting from an emission inventory already

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–partially- disaggregated over a spatial grid (the EMEP one) to keep our emissions more consistent to the official EMEP data.

From the discussion on figure 5 I get the feeling that all SNAP2 emissions are going into the urban areas with the largest population density. With the weighing of the data with the wood combustion function I would assume Paris would be gone, but it is not. . . The SNAP2 emission distribution in Poland, with strong regional differences, may deserve an explanation. The final spatial distribution of the PM emissions from SNAP2 relies on two elements: 1) the emission burden in each EMEP cell; 2) the population density through function 2. Fact 1 drives the large scale spatial distribution of the emissions (e.g. the differences between the emission over Poland, with respect to Germany, etc). Factor 2 drives just the spatial distribution within each EMEP cell. Concerning this, it is worth noting that function in figure 2 does not state that emissions decreases with increasing population density, but that emission per inhabitant decreases with population density. This means, at the end, that SNAP2 emissions still increase with population density but at a growth rate less than linear.

P4145, line 6: lowest layer is 20m. It is correct. The lowest layer is 20 m deep.

P4146. Line 15-25: is this explanation really completely correct? I would guess that a colder climate would have a lower seasonal variability in the temporal profiles by definition. A warmer climate has fewer days that count in the heating degree day sum, so fewer days require heating. Thus smaller emissions but with a higher variability in the temporal multiplication factor (as it is normalized to 1). We agree, and the same is stated in the text and showed in the figure

P4151, Line 11 : Somewhere the challenges for the coarse mode need to be discussed more thoroughly. Please explain why the PM10 goes less well than PM2.5 (a part of PM10). Is this because the dust and sea salt are not captured well, also in terms of timing? Other studies have shown that desert dust and sea salt events can be modelled in time quite well. Sea salt emissions computation is included as well as a contribution

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of long range transport of dust through boundary conditions. The degradation of the model performance moving from PM2.5 to PM10 can be surely partially explained by an imprecise reconstruction of dust and sea salt, but we think it could be also related to some missing or underestimated processes such as road dust resuspension, non-exhaust transport emissions a PM coarse chemistry. This statement and a reference showing the same behaviour has been added to the revised version of the paper.

P4152, line 4: How is the cloud chemistry described? For this kind of details about CHIMERE parameterisation the reader is reported to the Menut et al. (2013) paper.

P4152, Line 17: This statement can be stronger by looking at the background at Valencia stations. Is it the low values in the time series that are overestimated? Looking at the background at Valentia station indicated that the lowest value was overestimated.

P5152, Line 23: Nitrogen dioxide should be ammonia. This point has been corrected in the revised version of the paper.

P5153, Line 15-23: What are the typical levels of coarse nitrate in Europe? Are those levels sufficient to close the gap between observed and modeled nitrate? A coarse nitrate formation scheme was implemented in CHIMERE as part of a research project by Hodzic et al. (2006). The study showed that it can increase the nitrate model concentrations up to 3 μg/m3 especially the south part of Europe where coarse nitrates can represent the major part of the nitrate total mass. Hence, the introduction of a coarse nitrate formation scheme into CHIMERE could certainly help to reduced the bias between observed and modelled nitrate.

Line 26: is total nitrate really much better than nitrate? Table 6 states that most of the performance indicators (e.g. RMSE, FB and FE) clearly improve moving from NO3 to TNO3.

P5154, Line 1-2: The underestimation of NO2 is much less than the underestimation of (total) nitrate. The implication to the NOx emissions may be too easy, as the underesti-

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mation of NO2 is not really discussed in the paper. Moreover, the NO2 underestimation reported here is not in line with the TFMM model comparison results provided in a recent EMEP-report. The nitrate underestimation seems to be present in that study,so I wonder if the NOx underestimate is not due to other reasons. Please go into some more depth on the NO2, NO3 underestimation. 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 cannot 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.

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.

NO2 hourly modelled concentrations are also considered in order to go in some more

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depth on NO2 underestimation. The performance reported in TFMM report is done considering a different and much smaller set of UB stations (main cities only) thus explaining the better performance reported in that report.

Line 6: It is very likely that the overestimation is related to SO4, as nitrate is underestimated. Yes, we agree

P4155, line 22. Ozone also peaks above the Mediterranean because of a lack of ozone dry deposition above sea. Yes, we agree

P4156, Line 7: Looking at the distributions of the PM stations and the underestimation it seems that the areas in southwestern and southeastern Europe are largely underestimated. Is the comparison at the monitoring stations and the interpretation of those representative for the whole of Europe? The sentence has been rewritten to make the comment clearer.

Line 12: the sentence on dust is important and the role of dust and possible improvements should be discussed. Indeed, using the summer mean modelled concentrations map, we underline in the paper that dust contribute significantly in the south of the domain to the PM10 total mass.

Line 21: Strong power sector contributions are found there too. The power sector has been added in the text.

P4157: Could you also show dust and Organics as a distribution? The revised version of the paper now includes dust and SOA aerosol 2D mean concentration maps.

Conclusions section:

I would have been interested to see the impact and degree of improvement by the mentioned developments. The main degree of improvement is seen using the urban correction and is described in the discussion section as well as other model improvements.

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P4158, line 5: here an implicit comparison to coarser resolutions is made. It says that increasing resolution provides a better temporal correlation. This is not shown in the paper, but would be interesting. Did you compare the performance to earlier runs and the previous model version? Can you say something on that? The sentence has been changed. The aim of the paper was not to compare the performance of different resolution. However, such work is related also using CHIMERE in Colette et al., 2014.

The points for further research are not nicely motivated. Why these three? A discussion is needed. The conclusion has been rewritten in order to motivate the points for further research.

Line 25. The last point is a strange sentence. How can you introduce the French database, which is probably also top-down, for eastern European cities? Please rephrase. Note that this work has been done for some areas already, which could be mentioned. One of those is MEGAPOLI, in which the French teams were very much involved. The sentence has been rewritten to make this point clearer. We agree with the Referee that local inventory such as the one developed in the frame of MEGAPOLI can be of great interest for high resolution simulation such as the one described in the paper (INS for France and HERMES for Spain) and should be use for future simulations.

Figure 3. Step 2 should be daily disaggregation's as hourly is done in step 4. In step 4 the degree day approach is listed, whereas this provides a daily multiplication factor, not hour of the day. In step 2 it should be profiles instead of profils. Figure 3 has been removed from the revised version of the paper and the reader is referred to the section 2.3 for the description the anthropogenic emissions used.

Figure 13: NO2 pictures for summer and winter seem to be reversed. Same for PM10 in Figure 14 The order of the picture have been corrected in the revised version of the paper.

Figure 16. The ammonium distribution for the winter shows the nitrate pattern in east-

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ern Europe, whereas I expected to see that of sulfate. Why does ammonium concentrations minimize in the SO2, hotspots? I guess the ammonium and (2xSulphate + nitrate) are in agreement with each other, or? Please explain! Ammonium winter concentration shows a wide and rather homogenous pattern, covering both western and eastern Europe, together with a hot spot covering the whole Po valley. Ammonium concentration in western Europe is mainly driven by nitrate availability), whereas in Eastern Europe, ammonium spatial pattern is closely related to sulphate.

Interactive comment on Geosci. Model Dev. Discuss., 6, 4137, 2013.