April 19, 2013

Dear Editor,

Please find enclosed the review of the paper formerly entitled:

Impact of the vertical emission profiles on ground-level gas-phase pollution simulated from the EMEP emissions over Europe
by Sylvain Mailler, Dmitry Khvorostiyanov and Laurent Menut.

We wish to thank Anonymous Referees #1 and #2 for their useful reviews, which helped greatly to improve the manuscript by finding the points where more discussion was necessary to make the interpretation of the results clearer. All suggestions of the Reviewers have been taken into account, and significant parts of the paper have been re-written accordingly. Mainly, the main changes are:

- As suggested by the reviewer #2, the title was changed and is now *Impact of the vertical emission profiles on* background *gas-phase pollution simulated from the EMEP emissions over Europe*.
- A complete discussion about the choice of the stations is added in this letter and in the manuscript, showing the selected stations are the more convenient to highlight the results presented in this paper,
- Another new paragraph was added to explain that results can not be due to error compensations as asked by the two reviewers.

Detailed answers to both Reviewers' comments and recommandations are given in the following pages, as well as the explications of the changes that have been made following these comments and recommendations. The reviewers comments are in blue and the parts of the article that have been modified substantially are in green in the revised version.

Reviewer #1

Answer to specific comments

The use of a relatively coarse horizontal grid resolution with a rather refined vertical resolution seems to be not optimal, furthermore while more refined emission data than by EMEP are available with the MACC-TNO emission data base.

Yes, this study could very well have been performed with a higher resolution, which would have permitted to analyze the results for non-background stations. The MACC-TNO emission data base, for example, is provided at 7x7km for Europe, which permits high resolution simulations. However, five simulations have been performed for a 1-year period of time, and running the full modelling system (WRF and CHIMERE) with an enhanced resolution for a full one-year period would have represented a very heavy burden both in terms of computer time and of disk-space. It is true, however, that the use of a higher horizontal resolution would certainly have permitted to analyze the results not only for rural background stations, as it is currently the case.

The revised version includes a paragraph stating that the coarse resolution does not permit the intrpretation of model results for stations other than "background rural" type (section 3.3, first paragraph)

Regarding the correspondance between the horizontal and the vertical resolution, the vertical resolution used consists of 8 vertical hybrid sigma-p layers between the 0.997 sigma-level and 500 hPa.

The configuration with 8 levels is the most common configuration of CHIMERE. We think that, even when the horizontal resolution is degraded, this does not change the need for a relatively fine vertical resolution, in order to take into account adequately the concentration gradients within and above the Planetary Boundary Layer.

A similar vertical horizontal and vertical resolution has been used with CHIMERE for the AQMEII model intercomparison project, which is mentioned in the revised version (Section 2.1, second paragraph).

The improvement in model performance going from the default EMEP vertical distribution to the more realistic Bieser distribution is attributed to the improved vertical distribution only. However, this assumes that the quantity of the emissions is basically correct, which does not have to be the case.

This is completely true and also joins a concern of Referee #2.

The discussion of this point has been considerably extended in the revised version, see section 3.2.1, last paragraph, and the second paragraph of Section 4.

technical comments

Page 3664, line 18, needs instead of need, page 3671, line 8, were instead of was, Page 3674 detail instead of datail, page 3680 line 4, during instead of suring

Yes, this is done in the revised version, thank you.

Reviewer #2

General answer

I do not feel to have learnt anything new by reading this manuscript. The authors have to clarify what's new in this contribution and why the paper deserved publication in Atmospheric Chemistry and Physics.

Even though a couple of papers have been published in the last decade about the present topic, we do not feel that the results presented in the discussion paper are already known. The main articles published in this sense are those of [De Meij et al.(2006)], [Pregger and Friedrich(2009)] and [Bieser et al.(2011)] at continental scale (for Europe), and the work of [Pozzer et al.(2009)] at global scale. Following this comment, and as no specific bibliographic source is provided by Reviewer #2, we have looked for more bibliography but failed to find any recent result of significance the well-known journals of the area. It is true that a specialist knowledgeable about this topic can expect that the revision of the vertical layering of the emissions from the standard EMEP recommandation to alternative vertical profiles such as the ones from [Bieser et al.(2011)] will have an impact on pollutant concentrations simulated at ground level. However, the present paper analyzes the impact of the vertical layering of emissions by performing various model simulations varying the vertical layering of emissions, all else being equal, and compares the model outputs from these simulations to station observations, which has not be done in any of the previously mentioned studies except by [Pozzer et al.(2009)] in a very different context and with very different goals (at global scale with T42 spectral resolution and for tropospheric trace gases not including NO_2, O_3 or SO_2).

In the case of CHIMERE, at continental scale, with $0.5°\times0.5°$ resolution and 8 vertical levels, it is shown that switching from the EMEP recommandations to the profiles from [Bieser et al.(2011)] yields a very significant improvement of model results when compared to observations.

The message passed in the conclusion is that the modellers using a variety of Chemistry-transport models in Europe should revise carefully their policy regarding the vertical layering of emissions because the potential impact of different choices in the vertical layering of emissions is very strong (of the same order of magnitude as the simulated concentrations for SO_2, and of the same order of

magnitude as the model biases for NO_2 and O_3), which is a conclusion of importance for the modelling community.

We feel that these results are not known at present and represent a significant progress in the topic of the vertical layering of emissions (as also mentioned by Reviewer #1), complementing the papers of [Pregger and Friedrich(2009)] and [Bieser et al.(2011)] by an assessment of the impact of the new data and methods provided by these studies on modelled air quality. The scope of the manuscript is recorded in the last paragraph of the Introduction as follows:

"Despite this renewed interest during the recent years in the estimation of effective emission heights however, to the authors' knowledge, no study has systematically investigated the impact of updating the EMEP emissions heights towards other vertical profiles in a study validated through comparison with real-world data. The purpose of the present paper is to examine several strategies for revising the EMEP vertical disaggregation, either performing manual adjustments from the EMEP profiles or vertical profiles adapted from the [Bieser et al.(2011)] study, and evaluate the impact of these updated vertical profiles on CHIMERE performance relative to Airbase measurements over Europe."

Major comments

1. One of my major concerns is about the resolution of WRF-CHIMERE simulations. The resolution used is $0.5° \times 0.5°$ [...] so I guess that the results presented here are only valid for background concentration of pollutants, because, as stated by the authors, this resolution does not allow an assessment over urban and industrial areas [...] So the assessment presented here is only valid for background areas, and this has to be clarified both in the title and in the discussion results

Yes, the fact that the assessment provided here is only valid for background concentrations needed to be clarified.

The title is changed from **Impact of the vertical emission profiles on ground-level gas-phase pollution simulated from the EMEP emissions over Europe** to **Impact of the vertical emission profiles on *background* gas-phase pollution simulated from the EMEP emissions over Europe**.

Regarding the inclusion of this point in the Discussion, the fact that the simulation will be compared to observations mostly for stations of the "Background rural" type is now more clearly stated in the first paragraph of section 2.3.

2. Why not focus the statistical comparison just in [the rural background] type of stations ?

Focusing the comparison just to the *Rural background* would have been a possible choice. In fact, the results for stations other than the *Rural Background* stations are only presented in Table. 3 (in a very condensed form), and briefly discussed (p. 3674, l. 2-6) mostly to justify the fact that the stations other than *rural background* are not further analyzed. We think it is useful to actually show by providing the statistics (not only state) that the simulation performed can not be compared usefully to observations for other types of stations.

This point is clarified in Section 3.2.2, first and second paragraph.

3. Moreover, after reading the design of the experimental setup and the vertical profiles used, I would have expected a much more realistic approach to the vertical disaggregation and layering. Here, the authors do not trust realistic information on vertical profiles (but the work of Bieser et al., 2011) but perform a sensitivity analysis [...] That could lead to right results because of wrong reasons :

This point is of course very important. However, we have the feeling that the vertical profiles by Bieser et al., 2011 are the best available profiles at present (and this seems to be also the opinion

of Reviewer #1 who notes that the [Bieser et al.(2011)] are "As far as possible realistic"). More realistic approaches could include the realisation of simulations with a plume-in-grid approach in order to simulate the injection of industrial emissions into the atmosphere depending on the simulated meteorological conditions. However, there is a lack of actual information about industrial sources at the European level (as the EPER database does not include any information about stack-height, exit temperature and velocity), so that the profiles simulated by [Bieser et al.(2011)] represent the current state of the art. These profiles are based on statistics from several hundreds of individual sources and on a precise and published algorithm for the evaluation of the injection heights, which is not the case of the EMEP recommandations). Therefore, there is substantial evidence, provided by [Bieser et al.(2011)], to think that the profiles they provide are more realistic than the EMEP recommantations, and the results presented in the present discussion paper do not aim at establishing this, but at showing that the update from standard EMEP vertical emission profiles to the [Bieser et al.(2011)] emission profiles yields very substantial difference on the simulated background gas-phase pollution and, in the case of CHIMERE, substantial improvement. More realistic approaches need to be developed and used regarding the simulation of the injection heights and realistic plume simulation for each individual source, but this is not within the scope of the present study.

As this point is, in our opinion, very important, a new substantial discussion on this point is now included in the first and second paragraphs of subsection 3.2.1, as well as in the last paragraph of the same subsection.

4. Section 3.2.2 deals with the analysis of the simulations in two stations: DENW081 and PL0243. I personally find this analysis is very biased. [...] Why have these stations been selected ? In there any reason for that ? Why not selecting stations in other areas (southern Europe Mediterranean areas)?

The two stations for which time series are shown have been chosen because they are background rural type, but exhibit relatively high concentrations of pollutants compared to most stations of this type (Fig. 2). This is explained because they are influenced by two important industrial areas, the Rhur and Silesia. As such, they represent the effect of these two important industrial regions on regional background pollution. They also depict a different effect of the change in effective emission heights, due to different model behaviour for these two locations: as discussed, CHIMERE tends to overestimate the NO_2 concentrations at the PL0243 station, but underestimate them at the DENW081 station, so that comparing model behaviour between these two stations is, in our opinion interesting. The will to actually show and discuss the time series for the selected stations naturally limits the number of stations that can be selected. We think it is instructive for the reader to be able to appreciate on a couple of stations the effect a change of effective emission heights on the modelled concentrations.

The first paragraph of the section titled "Individual stations" has been rewritten to explain the choice of these two stations, and explicitly state that these stations are *not necessarily representative of all the "rural background" stations*.

Finally, it would maybe be possible to replace this section, as suggested, by a more detailed analysis where all stations have been considered, probably grouped by latitude, or station type, in order to have a more complete scope of the results.

The possibility to present an analysis grouping stations by type does not seem to be a real possibility, since as discussed above and in the manuscript, the results are significant only for stations of the "rural background" type. The concept of "grouping by latitude" is not scientifically sound because location of indistrial sources has no relation with the latitude. These emissions are sparse over a large domain (the Europe) and the meteorology and transport between sites lead to different behaviour from one site to another one. To group informations by latitude may be useful for meteorology and climate studies, but not in the case of industrial emissions. To prove this, Fig. 3 shows that the impact of the change in the vertical layering of emissions is basically a function of the distance to major industrial

centers. Therefore, we think that it is interesting to select two stations close to industrial centers and examine directly the time series.

5. no analysis on O3 concentrations is done in the manuscript, despite in the abstract the authors say they are going to do so (also, the analysis of NO2 is very limited in the manuscript

We do not feel that this comment is accurate. Results for O_3 are discussed for general statistics (p. 3674 l. 11-18, p. 3675 l. 11-18), for the DENW081 station (p. 3676 l. 21-23, l.26, continuing until line 10 of p. 3677), and for the PL0243 station (p. 3677, l. 28-29, continuing until l. 6 of p. 3678). In the Discussion, O_3 is discussed p. 3679, l. 8-11, l. 24-26 and p. 3680, l. 4. In the same way, results about NO_2 are discussed in similar length as for O_3. The main results presented and discussed about O_3 and NO_2 are that lowering the effective emission heights for industrial emissions tends to increase the background NO_2 concentrations at ground level, and correspondingly decrease the O_3 concentrations at ground level which, in the case of CHIMERE with the discussed configuration, tends to reduce model biases by about considerably for NO_2 and for O_3 (as mentioned in the Discussion, P. 3678, l. 25).

Minor comments

The page and line numbers mentioned by the Referee do not seem to correspond with those of the document as available on the ACPD website. However the comments are detailed enaugh so that it has been easy to find the corresponding place for each of the 4 minor comments.

Introduction: First two paragraphs must be supported by corresponding references

Yes, this is done in the revised version for submission to ACP, with reference to the articles of Zhang et al. (2012) and Menut and Bessagnet (2010).

Line 18, Page 5: I know the authors wanted to cover a full annual cycle, but I do not know the point of starting precisely on Feb. 20, 2008. In there any reason for that ? - I assume this corresponds to l. 11, p. 3668. We found that there was good data availability during this period. There is no other particular scientifical reason for this choice.

Line 21, Page 6: Since this article is still in press, some information on this topic should be included in this manuscript, either in this section or as supplementary material. I guess this information is essential to understand how temporal and spatial dissagregation is done in this work" (I assume this refers to l. 18, p. 3669) As mentioned above (p. 3669, l. 2-5), no horizontal disaggregation was performed since the model was run on the EMEP $0.5° \times 0.5°$ grid. The corresponding passage - second paragraph of section 2.2 - has been rewritten substantially in order to mention again explicitly the fact that no horizontal disaggregation has been performed, avoid the misleading mention of horizontal disaggregation on l. 17), and to state explicitly the strategy used for temporal disaggregation. The title of the corresponding section has been changed from "Downscaling of the emissions" to "Temporal and vertical downscaling of the emissions"

Line 2 and 11, Page 13: Is the code station DEN081 or DENW081? Please correct. (this refers to l. 15 and 20, P. 3676).

Yes, the correct code is DENW081, this is modified in the revised version, thanks.

Best regards,

Sylvain MAILLER

References

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[De Meij et al.(2006)] De Meij, A., Krol, M., Dentener, F., Vignati, E., Cuvelier, C., and Thunis, P.: The Sensitivity of Aerosol in Europe to two different Emission Inventories and Temporal Distribution of Emissions, Atmospheric Chemistry and Physics, 6, 4287–4309, 2006.

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