Atmos. Chem. Phys. Discuss., 15, C3479–C3487, 2015
www.atmos-chem-phys-discuss.net/15/C3479/2015/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source contributions to 2012 summertime aerosols in the Euro-Mediterranean region" *by* G. Rea et al.

G. Rea et al.

geraldine.rea@lmd.polytechnique.fr

Received and published: 11 June 2015

We thank the reviewer for the valuable comments to improve the quality of the manuscript. Please find below our point-by-point replies.

The manuscript by Rea et al. address a very important and urgent matter: the quantification of the contribution of different sources to the aerosol budget in Europe and the related contribution of natural events to PM_{10} exceedances. Model evaluation presented for summer 2012 illustrates good model performance, in line with the best state-of-the-art models. However, in this reviewer opinion, the manuscript may be enriched with more data from the Central and Eastern Mediterranean region and the final analysis relative to PM_{10} exceedances needs revision.

My two main concerns are: 1. In the manuscipt only data from the AirBase database for North and Western Europe are used. However, I accessed the AirBase portal on 14th April 2015 and PM_{10}/PM2.5 data for e.g. Italy, Greece and Turkey are apparently there for the year 2012: http://www.eea.europa.eu/themes/air/interactive/PM_{10}. The inclusion of these data is of overwhelming importance, since they include the regions where the greates part of the monitoring stations apparently report PM_{10} exceedances, and highest levels in general. Probably at the beginning of their work the authors couldn't find those data in the database, but now they're there (I am not sure when they were uploaded), so I would seriously encourage their use in the revised work. I understand that this will put significant efforts on the authors, but I feel this is determinant for the quality of the proposed analysis. 2. In this reviewer opinion the analysis on the contribution of natural events to PM_{10} exceedances is not robust and may carry misleading final messages. The weakest point is the lack of the quantification of the uncertainty associated to the percentages of naturally-occurring exceedances. In the model validation sections, the general bias of the model in terms of average and peak levels of PM_{10}, PM2.5 and AOD is well characterized (for those regions covered by AirBase data), and some model limitations are discussed but not quantified, such as dust positive bias. This complicates the interpretation of the final results. Let's, for example, consider the MED-We region (basically the Iberian Peninsula). There is systematic low bias in the PM_{10} and PM2.5 simulations, say -24% for PM_{10} and -33% for PM2.5 (Table 3). This is apparently related to some missing regional source, since the background levels are constantly underestimated (Figure 5). However, many peak values of PM during episodes are overestimated (Figure 3 and 5), and the model predicts that most of those peaks are mainly due to dust (Figure 10). It results that 92.5% of the exceedances are attributed to natural events (dust). This number, however, could be much less with a better simulation of PM_{10}: the -24% bias may entirely due to anthropogenic emissions (we don't know it actually) and the dust concentration may be overestimated by a factor of 2 during some episodes. This would drastically change the number of exceedances

ACPD
15, C3479–C3487, 2015

explained by dust. In conclusion, I thus strongly suggest to entirely revise the analysis presented in section 6 and discuss further (and perhaps attempt to quantify) the uncertainties associated to the percentage of exceedances attributable to natural sources.

1. The reviewer mentioned missing data though available on the airbase website, particularly in the Mediterranean region. The study began as supposed when those data were not present on the website. It is important for the study to have all the stations available; we thus downloaded and analysed the new database for the new version of the manuscript. It adds 167 rural background stations for PM_{10} concentrations, and 155 for $PM_{2.5}$ concentrations. Most of the added stations are located in Med-We, NEU-Ce, and Med-Ea (MED-Ea only for PM_{10}). The statistics do not significantly change for most of the regions, except in MED-ea for PM_{10} where the model underestimates strongly the observations (Section 4.1): "In the regional comparisons, highest differences are obtained in the MED-Ea region (26.9% of the stations meet the performance criteria and only 3.2% the performance goal for PM_{10}). A strong underestimation is noticed in this region with a MFB=-73.18%." The corresponding statistics are added in Tables 2 and 3, and the time series in Figure 6 has been update, as well as the entire section 4.1.
2. The reviewer suggests that the analysis of air quality exceedances of PM_{10} must be revised, as the uncertainty on natural source events, and particularly dust outbreaks, is not clearly quantified. In the new version, the contribution of each source on exceedances only seen by the model is analysed. The bias associated with the background concentration and to concentrations peaks is estimated and subtracted at each station. The section 6 is thus entirely re-written, with a new paragraph for the method used: "First, the bias on background levels is evaluated at each measurement station. The background is defined as the baseline concentration, on days when no significant peak is measured. It is generally associated with anthropogenic and biogenic sources, which have relatively low variability during the summer compared to dust and fire emissions. The average "background bias" is estimated to -6.3 μg m^{-3} for PM_{10} and -1.9 μg m^{-3} for $PM_{2.5}$ in MED-We, -0.93 μg m^{-3} and 1.3 μg m^{-3} in NEU-We, and

ACPD
15, C3479–C3487, 2015

-3.2 μg m^{-3} and -0.2 μg m^{-3} in NEU-Ce, on average over all stations. Then for the sporadic sources, i.e. for the dust and fire, the resulting bias at each point of a peak is subtracted only for concentrations from the corresponding source. This processing is performed only when the model overestimates the concentrations with respect to observations, but not when a peak is detected only by the observations."

The contributions are then analysed on this basis, making the results more robust (the total number of exceedances is 549 for PM_{10} (instead of 1964 initially), all of them detected also in the observations). The proportion of exceedances that happened exclusively from a single source is quantified, and also the proportion for the other exceedances: "In total, the only sources that result exclusively (i.e. when their contribution alone on PM concentration is more that the threshold of 50 μg m^{-3}) in an exceedance are dust (in 59% of the cases, i.e. 294 exceedances on the 498 observed and simulated), fires (in 1% of the cases, i.e. 5 exceedances), and anthropogenic sources (2 exceedances). The other 197 exceedances are due to mixing between several sources: anthropogenic sources contribute to 9.2% (MED-We) to 27.9% (NEU-Ce) of the concentrations, fires from 14.2% to 17.8%, and dust from 49.5 to 67.1%."

Other minor comments/suggestions:

P8192, L5: "if yes" would probably better be "if so"

corrected

P8192, L9: perhaps is worth adding a quick information on the horizontal resolution of CHIMERE.

"to perform a sensitivity analysis on a 50 km resolution domain"

ACPD

15, C3479–C3487, 2015

P8192, L13-14: "The sensitivity simulations are . . .". This sentence is redundant and may be omitted.

corrected

P8192, L15: Since the region defined in parentheses is the whole domain, I suggest to move its definition above, near L9.

"to perform a sensitivity analysis on a 50 km resolution domain (from -10W to 40E and from 30N to 55 N)"

P8192, L29: ". . .particularly when diverse sources are mixed". Why should be that? There is no clear evidence of that in the paper.

This sentence has been removed.

P8193, L1-4: The meaning of the last sentence of the abstract is completely obscure, must be completely revised.

This part of the abstract has been modified according to the new results from the optimized exceedances.

P8194, L18: ". . . because of mineral dust in addition to local anthropogenic pollutants". Perhaps rephrase as "because of a combination of mineral dust and anthropogenic pollutants from local sources"

corrected

ACPD
15, C3479–C3487, 2015

P8194, L28: "PM_{10} pollution" is referred to sea salts: can this natural source be termed as pollution?? I would prefer to see words such as "levels", "concentrations" when referring to natural sources, not "pollution".

corrected

P8196, L9: the underestimation of PM_{10} at the European scale was also recently confirmed by Im et al. (2014), Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2. Part II: Particulate Matter, Atmos. Environ., doi:10.1016/j.atmosenv.2014.08.072

This study has been added in the bibliography concerning the underestimation of PM_{10}

P8196, L14: An additional uncertainty in the model to obs comparison is the conversion of aerosol concentrations to optical depths, a process that is subject to its own assumptions and uncertainty. See e.g. : Péré et al. (2010), Evaluation of an aerosol optical scheme in the chemistry-transport model CHIMERE, Atmos. Environ., doi:10.1016/j.atmosenv.2010.06.034 Curci et al. (2014), Uncertainties of simulated aerosol optical properties induced by assumptions on aerosol physical and chemical properties: an AQMEII-2 perspective, Atmos. Environ., doi:10.1016/j.atmosenv.2014.09.009

These two studies have been added in the bibliography with a sentence added in the manuscript (Introduction): "The uncertainty on PM concentrations leads to uncertainties on the simulated aerosol optical properties and AOD, in addition to uncertainties in the calculation of optical properties (refractive indices, assumptions on aerosol mixing, etc) (Péré et al., 2010; Curci et al., 2014)."

P8198, L5: suggest not to skip the units here and throughout the text, e.g. add ug/m3

ACPD
15, C3479–C3487, 2015

after 80 here.

corrected

P8108, L11: "(80 of the daily mean values)", perhaps the authors meant "daily mean value of 80 ug/m3"? Not clear.

corrected

P8202, L28: please specify what assumption on the mixing state of aerosol is used for AOD calculations (external, internal, . . .)

The mixing state of aerosol assumed to be internal (Section 3.2): "The total AOD is also obtained from these calculations, with an internal mixing state assumption for aerosols, at five different wavelengths: 200, 300, 400, 600 and 1000 nm".

P8203, L4: ". . .linear interpolation. . ." this sounds strange, since usually AOD is scaled with wavelength using a log-scale interpolation (Angstrom exponent), please correct and clarify.

We agree that the calculation of the AOD using a linear interpolation was not accurate. It has been re-computed using a log-scale interpolation with the Angstrom exponent: "In this study, the simulated AOD from CHIMERE at 500 and 550 nm are used and extracted using the Angström coefficient from the output wavelengths." (end of Section 3.2) The results of the manuscript (evaluation of AOD and contributions) have been modified, but the differences are not significant.

P8203, L16-19: references to the procedures used to disaggregate model species and establish hourly profiles seem appropriate here.

ACPD

15, C3479–C3487, 2015

the article describing the procedure has been added in the bibliography and in the corresponding sentence: "Hourly emissions are estimated by applying seasonal, daily and weekly factors depending on the SNAP (Selected Nomenclature for Air Pollution) sectors (Menut et al., 2012)."

P8205, L17: "(zone of influence. . .)", the zone of influence cannot be detected removing diffuse sources, I would remove this comment in parenthesis.

comment removed

P8206, L13-14: please define the mathematical formulas used for MFE and MBE (and maybe also for other statistical indices used in the paper).

The formulas has been added for MFE and MFB, and RMSE :

$$MFB = \frac{1}{N}\sum_{i=1}^{N}\frac{(C_{mod} - C_{obs})}{(C_{mod} + C_{obs})/2} \quad (1)$$

$$MFE = \frac{1}{N}\sum_{i=1}^{N}\frac{|C_{mod} - C_{obs}|}{(C_{mod} + C_{obs})/2} \quad (2)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{N}(C_{mod} - C_{obs})^2}{N}} \quad (3)$$

P8207, L10: Add "%" after -21.2

corrected

ACPD
15, C3479–C3487, 2015

P8207, L16: change "adapted" to "appropriate for"

corrected

P8207, L25: "(but lower)" it is not clear what is referring to, the model?

corrected, it referred to the other high values observed at AirBase stations in MED-We : "High values are also observed for this date on some of the Mediterranean stations such as Miramas".

P8208, L10: add "s" to "dataset".

corrected

Figure 1: the top panels are very difficult to read. Please redraw, e.g. removing the black circles around each point.

Figure 1 (now Figure 2) has been redraw by removing the black circles

Figure 13: What is N in the titles? The total number of stations/points exceeding PM_{10} limits? Please clarify

N is the number of stations considered in the analysis in each sub-regions. This precision has been added in the legend.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 8191, 2015.