

Interactive comment on “Changes in the domestic heating fuel in Greece: effects on atmospheric chemistry and radiation” by Eleni Athanasopoulou et al.

Eleni Athanasopoulou et al.

eathana@meteo.noa.gr

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1. In section 2.2 and table 1 you should better explain the model setup. It is not clear to me which areas were modeled with COSMO-ART, if there is a coarser grid around Athens that was modeled with a CTM as well. It looks like the area you ran COSMOART on is about 30x40 grid points. This is rather small, in particular when you use boundaries for the gaseous species which are on a 2.5 x 1.9 grid. In this case one grid cell of the outer grid would be bigger than your entire model domain. In addition, the horizontal resolution of your emissions is much coarser than the model resolution, by a factor of 2.5 x 5. The question arises why you did not make any attempts to use

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spatial surrogates like population density to redistribute the emission data and provide them in a higher resolution as model input. Author's response: There is a single area modeled with COSMO-ART, "the extended area of Greece, centered on Athens" (p.5 line 8). The covered area is 18 to 30 E° and 33 to 42 N°. The horizontal spatial resolution is 0.025 deg, thus the whole domain is composed by 384 x 330 grid points. The reviewer though is correct, the horizontal extend of the domain was not explicitly mentioned in the initial version of the paper. It is now added in Table 1. With respect to the grid differences between emission data and model runs, first please note that the size factor between the TNO/MACC inventory and the COSMO-ART grid is smaller than a factor 2.5 x 5, since COSMO-ART ran on a rotated grid with equator placed over Greece. At the latitude of Greece, the size factor is in the order of 2.5 x 3.9. Still, emission downscaling could be beneficial, but this has not yet been implemented in the official COSMO-ART emission pre-processor. A proper downscaling is a quite complex issue (see e.g. Lopez-Aparicio et al., Atmos. Environ. 154, 285-296, 2017). For instance, population data alone is not sufficient for this purpose, since it is a poor proxy for industrial sources or power plant emissions and only a moderately reliable proxy for traffic. With respect to the suitable proxy(-ies) for residential wood combustion, see Comments no. 7 and 37.

2. page 2, line 21 and in the following: You use BC and EC as it is the same. You need to explain this a bit, e.g. provide definitions of both of them and their relation to what you call "soot". This is of particular importance because the radiative effects of the aerosol is in close connection to the BC/EC concentrations. Author's response: The reviewer is of course correct about the different definitions of black carbon, elemental carbon and soot. Apart from this specific statement in the Introductory section, we only deal with BC data throughout the current study. Therefore, we see no actual need for the provision of the definitions and inter-relation, rather than providing a relevant reference and a statement with respect to their erroneous usage as synonyms (p. 2 line 22).

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3. page 5, line5: “The atmospheric pressure and precipitation parameters were optimized with respect to the high spatial resolution of the current application (0.025°). This could be anything, therefore you need to explain what you did with the data. Author’s response: In the COSMO-ART (and pre-processor) configuration, there are certain parameters/flags that are spatially-sensitive. Among others, these settings correspond to a better balancing of the pressure fields and to a tuning in precipitation, in accordance to the horizontal spatial resolution of 0.025 deg. Therefore, the input data remained intact, the microphysics scheme was not altered, but e.g. the mask for smoothing of steep orography is adjusted, the critical value for normalized over-saturation is adjusted and so on. It should be noted that these changes are imposed either by using ‘true’ or ‘false’ or by applying certain code numbering in specific flags of the model (and pre-processor) job scripts. In order to avoid a similar confusion by the readers, we suggest deleting the respective phrase from the document.

4. page 5, line 16: If I understand it correctly, you model 2013/14 but you used 2009 emissions for all species and sectors, except for wood combustion. This can be well justified if no big changes in the emissions from year to year can be expected. However, you argue that “non-solid fuels” became very expensive (page 2, line 2/3) and that the financial crisis changed the emission pattern quite drastically. Shouldn’t this be considered in the emissions of other sectors than wood burning as well? Author’s response: In principle, the reviewer is correct, the latest available data relevant to emission rates should be considered for atmospheric modelling. Nevertheless, the latest available, official emission data from the TNO-MACC_II database refer to year 2009, which we were able to update according to recent specialized measurements of aerosol species during recent, intensive RWB wintertime periods. A recent study on air pollution emissions in Greece based on actual data (Fameli et al., 2016) shows that with respect to residential heating, fuel consumption is more or less stable for all fuels from 2009 to 2012 (latest available data), except for wood which is increased. With respect to the road sector, we cannot estimate the impact of crisis on transport, since available data end in 2010. This is now added in the text (p. 5 lines 23-26)

5. page 5, line 21: “For Greece, it is calculated that almost all mass (i.e. 98%) of the total PM₁₀ emissions from this source category reflects wood combustion (i.e. open fireplaces)”: Is this given in the TNO emission inventory? Or is this given in the IIASA publication which you cite later (line 25)? You should state this more clearly. Author’s response: Yes, this is a finding based on the TNO emission data for Greece, by comparing PM₁₀ total residential combustion to PM₁₀ residential wood burning (mass) emissions (personal communication with Hugo Denier van der Gon). It is true though that the TNO_MACC-II methodology incorporates model approximations, when data for certain sectors/countries are unavailable. In particular, reported data for PM₁₀ and PM_{2.5} emissions were not available for Greece, thus emissions were taken from the GAINS model (IIASA, 2012). This is now more clearly stated in the document (p. 6 line 6).

6. page 5, line 31-page6, line 11: This change in the temporal profile is an important modification of the main PM emission pattern. I think it would be good to explain in some more detail how you did that, e.g. by showing the long term BC measurements you mention. Can you be sure that the BC observations are dominated by wood combustion and not heavily influenced by traffic? Author’s response: It is true that the revised temporal cycles led to crucial changes in the daily and weekly cycles of the PM concentrations (e.g. Figure 5). This revision was based solely on the wood burning fraction of BC (BC_{wb}; method of separation of BC to BC_{wb} and BC_{ff} -fossil fuel- as already described in Sect. 2.1), thus not influenced by traffic or other sources. Second, both temporal cycles correspond to the mean values of the long-term measurements (2013-2015) of BC_{wb} in Athens, normalized with respect to the average value of each cycle. So actually Figure 1 reflects directly the long-term (normalized) measurements of BC_{wb}, as suggested by the reviewer’s comment. This is analytically described in Sect. 2.3, but now also added in the caption of Figure 1.

7. page 6, line 21: Although you modified the temporal profile of the RWB emissions you did not improve the spatial distribution, e.g. by using higher resolved population

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density maps. Why not? Shouldn't this make a significant difference? Did you make an attempt to couple the day-by-day variation of the emissions to the ambient temperature? Author's response: As already stated in Comment no. 1, emission downscaling has not yet been implemented in the official COSMO-ART emission pre-processor. Using proxies for wood burning is even more elusive. Discussions are under way at the European level (e.g. FAIRMODE forum) for the best approach on distributing wood burning emissions. The geographical spread of fire places/wood stoves use, would probably be a better proxy than the population density for the estimation of RWB emissions, which are two parameters not necessarily inter-correlated. Such a coupling went beyond the scope of this study, but is a currently ongoing effort of our group, so that the downscaling of residential combustion emissions in the Athens basin to the intra-urban scale is performed, based on construction and socio-economic criteria. With respect to the correlation of air pollution to temperature, this is already performed for PM concentrations, shown in Figure 4 (cold nights are highlighted) and Figure 8 (OA vs. Tmin). It is explicitly written in the text that this correlation reflects the coupling of the day-by-day variation of the emissions to the ambient temperature (so called 'heating demand'; p. 12 lines 30-32). We have concluded that the performance of their numerical coupling online can be an added value for the model system, when long-term winter-time model outputs are necessary, during the current period (intense use of fireplaces). Nevertheless, if one wishes to focus on RWB smog events, as in our case, coupling is not crucial, as model outputs capture the measured aerosol peaks.

8. page 6, line 26-31: I didn't get what the assumptions are concerning the magnitude of the emissions. If I understand you correctly, you took the emitted PM10 mass from the TNO inventory without further changes, although you explained before that the assumptions about the fuel split made in this inventory are not valid for the time period you investigate here. In the end, you argue that the magnitude of the emissions is ok because the agreement between model results and observations indicates that. This seems to be inconsistent. Author's response: Indeed, the total PM10 mass emitted from the residential combustion section, provided by the TNO database, was

not altered, as on a daily basis there was good agreement between model results and measurements of total PM₁₀, especially during cold days/smog events. However, the carbonaceous PM components (OA and BC) were poorly captured. Thus, our improvements involved a different chemical profile to the residential combustion aerosol emissions (different emitted mass of OA and BC component of PM₁₀) than the TNO (central heating profile), driven by the respective, specialized (RWB) measurements (p. 6 line 27- page 7 line 2).

9. page 6, line 15: What is the reason for the differences between your Mie calculations and the values used in COSMO-ART? Is the conclusion that the optical properties for soot are incorrect in COSMO-ART? Author's response: No, the optical properties for aerosol particles incorporated in COSMO-ART are correct. Nevertheless, they have been defined for typical air pollution conditions in an area of Germany. Our calculations are based on local conditions with respect to the aerosol chemical profile for the event/area of interest, as well as for the local, representative RH and PBL height (cf. Sect. 2.4). Thus, they differ from the ones in COSMO-ART, but they are similar both to other findings in the Mediterranean (Mishra et al., 2014; 2015), as well as to respective model inputs (Takemura et al., 2002). The relevant phrase is now enriched as follows: "These values differ from the ones used in Vogel et al. (2009; black and red lines in Figure 3), which is expected due to the different geographical areas and periods of interest between the two studies." (p.7 lines 31-32).

10. page 6, line 20: "The period studied can be characterized as a relatively mild winter period.": Why did you choose this period when you can expect that the heating activities will be comparably low? Author's response: Indeed, the winter of 2013-14 was mild. Nevertheless, the period we have focused our research on the RWB implications on air pollution issues (19 Dec – 5 Jan) can be characterised as an intense smog period, as explained in p. 8 lines 18-23). We have rephrased the initial statement (p. 8 line 5), so that the reader is not confused with respect to the whole winter and the selected smog period.

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11. page 8, line 24/25: Is the intense smog period the entire period from 19 Dec to 21 Jan? The mean concentration values for case 2 agree better to the observations than those from the baseline although the total emission amount was the same. Is that correct? You should make this clear. Author's response: No, the smog period is not the entire modelled period. Taken from the text (p. 8 lines 18-20): "19 December 2013 – 05 January 2014 ... excluding the rainy days mentioned above, constitutes the intense smog period of the current modeling study". Yes, the reviewer is correct, a clarification and explanation is needed for this finding. In fact, although the total PM10 mass emitted by residential combustion is unchanged within a typical week, the weekday mass amounts are reduced due to the revised weekly cycles, which are instead emitted during the weekends (cf. Fig. 1b). Given the fact that the intense smog period (13 days) includes only 4.5 weekend days, the differences found by the two scenarios in the average PM10 concentrations are expected. This is now explained in the revised text (p.9 lines 13-16).

12. page 9, line 12 and line 15: You should avoid descriptions like "satisfactorily represented" and "nicely captured". Each reader might think differently about what is satisfying or nice. Obviously, for case 2 the temporal profile of the concentrations fits better to the observations than for case 1 and you should just describe this. You could give a correlation coefficient to show this in numbers. Author's response: 'satisfactorily represented' is replaced by 'reproduced' and 'nicely' has been removed. Correlation coefficients are added for the daily cycles of carbonaceous species, and mean hourly bias is added in the peak comparisons (p. 10 lines 3-4, lines 6-8).

13. page 10, line 33: "The peaks observed at the urban core, as revealed from this model application, are somewhat displaced from the exact location of the site. This finding is in line with the characterization...": This is a bit over-interpreted, having the coarse emission map in mind. The grid cells with the highest emissions are simply north of Thissio. Author's response: We agree over possible over-interpretation, which is why we have calculated the mean, minimum and maximum values of a greater urban

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area (118 km², 15 cells; SW corner: 37.94°E, 23.67°N) and compared them to the model results at the grid point of Thissio. The results, which are given in p. 11 line 30 – p. 12 line 5), further support the argument already posed by Gratsea et al. (2016), on the characterization of Thissio as an urban background site, not intensively affected by local traffic and representative of the average background pollution conditions in Athens.

14. page 12, line 9/10: "... for a pre-crisis period or significant change in dominant heating fuels, the TNO-MACC_II emissions from residential heating should be further adjusted.": Why for a pre-crisis period? Didn't you say that the fuels changed significantly during the crisis? Why is the TNO inventory then wrong in a pre-crisis period? Author's response: It is true that during the pre-crisis period, Greek households did not use wood as the primary heating fuel, but central heating installations, which emit far less PM₁₀ mass than wood combustion. As evident from our study, TNO-MACC_II PM₁₀ emission rates for Greece, better fit to this RWB increase, rather than the central heating emission conditions. Thus, caution should be placed when the simulated periods reflect non-wood residential combustion, but the usage of other fuels, such as gas. The reason behind these findings might be that for the case of PM₁₀ and PM_{2.5} in Greece, TNO had no data available, thus applied model approximations for the calculation of emissions (cf. reply to Comment no 5).

15. page 14: I do not agree with the last part of the conclusions. In particular: - "Thus, human health implications, as well as policy making, when the fireplaces are in use to cope with high heating demand conditions (HD > 7.5 C), can be satisfactorily estimated and planned with the aid of such a tool.": You did not estimate any human health effects with your model system. How will help to estimate them? What can be planned with the help of the model results? - "For mild winter conditions (T_{min} > 8-9 C), a post-processing of model results according to the linear regression between HD and model bias, can further improve the quality of the model system.": Wouldn't it be better to improve the emission estimates than to correct the model? - "Alternatively, an inter-

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active treatment of RWB emissions, i.e. their online adjustment according to the actual temperature conditions of the simulation period, is proposed as a means to further enhance the reliability of operational forecasts of online-coupled atmospheric models.” I agree to this, but how would you know how much wood is used in comparison to oil or other fuels? Author’s response: The reviewer is correct; we are not estimating health effects within this study. Nevertheless, these or similar model results can be used to provide maps with exceedances of the EU limits of PM, thus to estimate the differential exposure of population to air pollution threats. Such information could be of help to policy makers as well, with respect to warnings and measures against RWB emissions and population exposure during wintertime. Yes, we agree that the best approximation would be to improve emission estimates (online) than correct the model outputs (offline). In other words, a hybrid model of RWB treatment of emissions during the model runs, could be beneficial, depending on the scope of the study. The proposed methodology does not need the information with respect to the use of wood in comparison to other fuels, because it will adjust the given TNO mass emissions according to the ambient temperature (timestep or hourly model values), based on actual measurements (PM vs. HD) that already include this behavioural response to the use of the different heating fuels. Such a modification is out of the scope of the current study, but valuable in future studies, targeting long-term wintertime periods or operational forecasts rather than intense and specific smog episodes. A supporting statement to this, is now added in p. 12 line 30).

Minor comments 16. page 1, line 17: “: : : accurately predicts : : :”: Qualifiers like “accurately” are always a bit difficult if do not give numbers for the deviations. Author’s response: ‘accurately predicts’ is replaced by ‘reproduces the measured’

17. page 2, line 2: “exorbitant” should be replaced by “very high”. Author’s response: done

18. page 3, line 7: Denier van der Gon Author’s response: done

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19. page 2, line 21 and in the following: You use BC and EC as it is the same. You need to explain this a bit, e.g. provide definitions of both of them and their relation to what you call “soot” Author’s response: The reviewer is of course correct about the different definitions of black carbon, elemental carbon and soot. Apart from this specific statement in the Introductory section, we only deal with BC data throughout the current study. Therefore, we see no actual need for the provision of the definitions and interrelation, rather than providing a relevant reference and a statement with respect to their erroneous usage as synonyms (p. 2 line 22).

20. page 3, line 32: “ : : : which was, however, nonlinear.”: In which way nonlinear? Author’s response: The ratio of ΔF (net radiative flux in W/m^2) and AOD (at 450 nm), has a linear relation for the net shortwave flux at the surface, i.e. the points in a scatter plot (y-axis ΔF , x-axis AOD) form a line with a negative slope (Stanelle et al., 2010). For the longwave the graph is not a line and resembles more a logarithmic graph. The text is now written as: “Stanelle et. al. (2010) concentrated on dust episodes over West Africa and found an average increase of $70 Wm^{-2}$ for the long-wave radiative effect. Comparing the AOD (at 450nm) with the surface long-wave flux, he found the relationship to be nonlinear in contrast to the shortwave case.” (p. 33 - p. 4 line 2)

21. page 8, line2: “mean maximum nighttime PM10”: over which period was the mean taken? Was this hourly? Author’s response: the period is 19 December 2013 – 05 January 2014. Yes, this is an hourly value. The whole phrase now is “The meteorological conditions favored the accumulation of smog over Athens during the period 19 December 2013 – 05 January 2014 (mean maximum nighttime hourly PM10 measured concentrations of $103.7 \mu g m^{-3}$).” (p. 8 lines 18-19).

22. page 9, line 1: “Overall, the revised run improved more than the 70% of the day and nighttime PM10 peaks during the intense smog period.”: This is not well formulated and therefore unclear. What exactly is improved? Author’s response: The reviewer is correct. The phrase is now formulated as: “Overall, the revised run shows improvements in the calculated PPEA values for more than the 70% of the day and nighttime

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PM10 peaks during the intense smog period.” (p. 9 lines 25-26).

page 9, line 10/11: “which leads to the improvement of the half PM1OA and of all PM1BC the daytime peaks during the intense smog period.” This sentence is obscure. Author’s response: it is again about better PPEA values. The phrase is formulated as “The PPEA values for both carbonaceous species are significantly lower for case 2, i.e. for the half PM1OA and of all PM1BC the daytime peaks during the intense smog period.” (p. 9 line 33 – p. 10 line 2).

23. page 9, line 28-30: “Thus, the aerosol chemical composition during the economic crisis is completely altered with respect to the chemical profile of wintertime aerosols beforehand.” What was the chemical profile before the economic crisis? Author’s response: This is given in p. 6 lines 27-28: “. . .the original chemical profile of aerosol emissions from non-industrial combustion (20% BC, 40% OA and 40% others) was modified. . .”

24. page 10, line 2/3: “The comparison with available measurements during increased wood burning in the Alpine area (Szidat et al., 2009) reveals similarities”: Where are these measurements shown? Are they given as the observations in Table 3? Please add the values they found somewhere. Author’s response: The values are now added in Table 5.

25. page 10, line 27: “PM1BC is found below 8”: The unit is missing. Author’s response: The unit “ $\mu\text{g m}^{-3}$ ” is now added.

26. page 12, line 2/3: “Inversely, during the cold days (T_{min} below 8 C) that the HD is increased (>7.5 C), and consequently no model overestimation occurs.” Omit the word “that”. In addition: It is true, that no overestimation occurs. Instead, you see an underestimation. Author’s response: “that” is deleted from the text. We kept the rest of the sentence as was, since our main point in this figure and paragraph is model overestimation, i.e. that the model does not overestimate PM concentrations during smog events (high HD conditions) as during the milder winter days, without

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actually implying that it systematically underestimates (model bias below -25% occurs in 4 cases).

27. Table 3 is really hard to read. It looks unstructured I would suggest to split into two or three different tables. One suggestion could be to have the ratios in the bottom (OC/BC,...) in separate table and also the peak analysis in another separate one. For the ratios in the bottom, it is unclear how they are given. OC/BC = 2.8 makes some sense, but what about the other values (BC/TC,...)? Are they given in %? BC/TC = 28 makes no sense. In addition, it seems that you derived them from case2/case 3 differences but you do not say this in the caption or anywhere else in the table itself. Author's response: The table is now split in 3, as suggested by the reviewer. All ratios are now dimensionless (pure numbers). RWB fraction predictions are the outcome of case 2 – case 3 differences. This is now explained in Table 3.

28. Figure 7: The caption is wrong. It contains parts that belong to Fig. 8. Author's response: The caption is now corrected: "The mean daily cycle of PM10 concentrations during the mild winter period, from the observations (black dots) and model predictions by case 3 (green line). All data refer to the Thissio site (Athens)."

Please also note the supplement to this comment:

<https://www.atmos-chem-phys-discuss.net/acp-2017-139/acp-2017-139-AC2-supplement.pdf>

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-139>, 2017.

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