

Below, the final author comments on behalf of all co-authors are published. The response to the Referees is structured as: (1) comments from Referees (in italics), (2) author's response after each comment (normal fonts), (3) author's changes in manuscript, marked with track changes.

5 **Comments from Anonymous Referee #1:**

This is an interesting and well written paper. In general, the approach that was followed is sound and the assumptions can be followed. There are some points that the authors need to explain in more detail. They need to better justify the approach and the model setup they have chosen. I also suggest some modifications w.r.t. the wording as well as to figures and tables.

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Major comments:

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 spatial surrogates like population density to redistribute the emission data and provide them in a higher resolution as model input.*

20 **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.

25 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.

35 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).

45 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 PM2.5 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 (BCwb; method of separation of BC to BCwb and BCff -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 BCwb in Athens, normalized with respect to the average value of each cycle. So actually Figure 1 reflects directly the long-term (normalized) measurements of BCwb, 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 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.

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 area (118 km², 15 cells; SW corner: 37.94°, 23.67°) 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 PM2.5 in Greece, TNO had no data available, thus applied model approximations for the calculation of emissions (cf. reply to Comment no 5).

- 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 ($HD > 7.5\text{ C}$), 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 ($T_{min} > 8-9\text{ C}$), 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 interactive 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?

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15 **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.

20 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).

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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.*

35 **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*

40 **Author’s response:** done

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”*

45 **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).

20. *page 3, line 32: “ : : which was, however, nonlinear.”: In which way nonlinear?*

5 **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.

10 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?*

15 **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 PM_{10} measured concentrations of $103.7 \mu g m^{-3}$).” (p. 8 lines 18-19).

20 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?*

25 **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 PM_{10} 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.

30 **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 PM_{1OA} and of all PM_{1BC} the daytime peaks during the intense smog period.” (p. 9 line 33 – p. 10 line 2).

35 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...”

40 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.*

45 **Author's response:** The unit “ $\mu g m^{-3}$ ” 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.

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

10 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.

15 **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.

20 **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).”

Comments from Anonymous Referee #2: The manuscript concerns the impact of increasing burning for residential heating in Athens, Greece. Even if the Greek situation is peculiar due to the impact of the economic crisis, this subject has great importance over the whole Europe, where the use of wood burning for house heating is increasing in a number of areas of different states. Even if the scientific knowledge of the dangers associated to biomass burning is growing, the air quality impact of biomass burning in domestic devices is not yet perceived and understood by the population and decision makers.

The manuscript is properly organized and well written. It points out that biomass burning for house heating has a major impact on the air quality and on atmospheric composition, while it has a minor influence on local radiative forcing. The authors describe their approach to improve wood burning description within the emission modelling that could be useful for model development and application in different areas and contexts.

29. Because the manuscript is proposed for publication in a special issue on coupled chemistry-meteorology modelling, a discussion on the relevance of the on-line coupling approach on the presented results concerning atmospheric composition and aerosol chemistry is presently missing. The on-line coupling advantage is evident only for the studied aerosols feedback on meteorology, while it should be specified is any difference could be noticed on pollutants concentrations when feedback was switched off (Scenario 4).

Author's response: We thank the reviewer for the positive comments and the interesting suggestion. We have calculated the differences on PM10 concentrations (and their chemical composition) between Case2 and Case4. The feedback of the online coupling approach on aerosol is found very small, which was expected given the small negative effect of particles on radiation in our case study.

The section 3.2 is now named after "Impact of RWB smog on radiation and feedbacks on atmospheric composition", and a relevant discussion is now added therein (p. 13 lines 21-26), plus some additions in abstract (p. 1 line 24) and conclusions (p. 14 lines 32-33).

The present form of the manuscript needs a revision including: clarifications, figures improvement, extension of feedback effects discussion.

Specific comments:

2.2 Model framework and setup

Page 5

Line 3

30. *The model domain is defined to be "the extended area of Greece" (the same definition is repeated in Table 1). This definition is quite generic and should be made more specific adding a Figure or a better definition of the domain boundaries.*

Author's response: the definition of the horizontal domain in degrees is now added in Table 1.

Lines 5-6

31. *The sentence "The atmospheric pressure...." is not understandable in this form. How where pressure and precipitation optimized? Do the authors refer to the choice of the microphysics scheme? What is the mentioned optimization?*

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 further puzzling of readers, we have decided to remove the respective phrase from the document.

5

Line 9

32. *Does “constant initial conditions” mean uniform initial conditions?*

Author's response: Combining this with the next comment (no. 33), the phrase (p. 5 line 14) is now replaced by: “...the uniform (in space) and constant (in time) initial conditions (e.g. for SO₂ and aerosol species).”

10

Table 1

33. *It is not clear how the initial and boundary conditions for aerosols are defined. Are the values included in Table 1 uniform in space? Are those values kept constant at boundaries?*

Author's response: Yes to both. This is now better explained in text (cf. response to Comment no. 32).

15

2.3 Modifications of the aerosol emissions

34. *Figure 2 is hardly understandable. Its quality should be improved.*

Author's response: all figures will be uploaded separately in high quality, following the ACP guidelines.

20

Page 6

Lines 21-23

35. *Does the sentence “Combined with the temporal...” refer to Figure 2?*

Author's response: After this comment, the phrase (p. 7 lines 3-4) is modified for clarity as: “...in Figure 2. These rates, combined with the temporal profiles (Figure 1),...”

25

36. *Does Figure 2 describe average emissions or do plotted values refer to a specific hour?*

Author's response: These are hourly emissions “for a night hour of a weekday (Tuesday, 21.00 UTC)”, as indicated in the caption of the figure.

30

Lines 23-24

37. *Maximum wood burning emissions are said to be located at the urban core, while it would be reasonable to expect to have maximum emissions over peripheral areas, where the access to wood should be easier than in the center.*

35

Author's response: In principle, population density is used as a spatial proxy in order to distribute wood combustion emissions ([TNO report, 2010](#)). The TNO-MACC_II emission database we have used for this study (Kuenen et al., 2014), uses a population map at high resolution, and a special proxy for the distribution of residential wood combustion. The latter takes into account both the population density, but also the proximity to wood (for more information on proxies for RWB, cf. Comment no. 7). Despite this combination for the distribution of residential wood combustion, an overallocation of the emissions in urbanized centres is possible, as the reviewer also comments, which is already described in previous studies (Denier van der Gon et al., 2014 and Timmermans et al., 2013). We have now included this info in the text. (p. 7 lines 5-10).

40

2.4 The aerosol optical properties

45

Page 7

Line 9

38. *Concerning aerosols composition, it is not clear how the concentration values reported in brackets should be interpreted.*

Author's response: These are RWB smog period-averages of the respective hourly values measured at Thissio during the RWB periods of winter 2013-14. This is explained in the text, in the previous sentence: "...is based on observational data collected in Greece during the 2013-2014 RWB smog episodes. In particular, the average surface chemical composition of ultrafine aerosols in Athens (pure soot: $2.8 \mu\text{g m}^{-3}$, water soluble mixture of sulfate, nitrate, ammonium and organics: $22.2 \mu\text{g m}^{-3}$), local relative humidity (50-70 %) and an average mixing layer height (600 m a.g.l., Gerasopoulos et al., 2017), were used to feed the OPAC software (Hess et al., 1998), which then - by applying the Mie theory - provides the respective optical properties for 61 wavelengths between 0.25 and $40 \mu\text{m}$ ". Thus, no interpretation should be extracted from these values; we provide information on the representative (average) values we used to calculate the aerosol optical properties for our case study.

Lines 15-18

39. *The values used for Athens differ from those used by Vogel et al. (2009). Is the difference due to the geographic area of application or to any other understandable reason?*

Author's response: Yes, the difference is related both to the geographic area and to the selected events. The phrase (p. 7 lines 31-32) 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."

40. *To which geographic region do values reported by Takemura et al. refer?*

Author's response: They do not correspond to a specific area, but are used as model inputs for the model study of Takemura et al. (2002). Thus, they are directly comparable to our findings. This is now more clearly explained in p. 8 line 2).

3.1 Impacts of residential wood burning (RWB) on atmospheric aerosol mass and chemistry

3.1.1 Aerosol model performance under smog influence

Page 8

Line 23

41. *Do values reported in Table 3 as "daily mean" refer to the whole period mean as understood from the manuscript text?*

Author's response: Yes, these are averaged daily mean values for the RWB smog sub-period of the simulation period. The phrase 'averaged daily mean' is now used instead of 'daily mean'.

Page 9

Lines 1-2

42. *How (on the basis of what parameters) is it evaluated the mentioned 70% improvement?*

Author's response: The relevant phrase is now written as "Overall, the revised run shows improvements in the calculated PPEA values for more than the 70% of the day and nighttime PM_{10} peaks during the intense smog period.", so that this question is answered in the manuscript (p. 9 lines 25-26).

Lines 10-11

43. *The meaning of the sentence "which leads to the improvement of the half PM_1 OA and of all PM_1 BC the daytime peaks during the intense smog period" is not clear.*

Author's response: The relevant phrase is now written as “The PPEA values for both carbonaceous species are significantly lower for case 2, i.e. for the half PM_{10A} and of all PM_{10BC} the daytime peaks during the intense smog period”, so that its meaning is clear (p. 9 line 33 – p. 10 line 2).

5 3.1.2 Representative spatial aerosol fields

Page 10

Lines 16-17

44. *The reference to PM10 EU alarm threshold is not clear, please include proper references to EU directives.*

10 **Author's response:** The reviewer is correct. The PM10 alarm value we refer to, is not suggested by any EU directive, but rather by the National Legislation (Joint Ministerial Decision by FEK 3272B/23-12-2013, in Greek), which supplements the respective EU directive with respect to public information and emission reduction thresholds.

15 The acronym ‘EU’ is now replaced with ‘National’ within text and a proper reference is added (p. 11 line 9). The same changes are now done for the alert threshold for the whole population (p. 6 line 29).

3.2 Impact of RWB smog on radiation

Page 12

Lines 25-27

20 45. *The sentence concerning removal of absorbing BC is not very clear and should be rephrased to be more clearly understandable.*

Author's response: This sentence (p. 13 lines 18-20) is now rephrased as: “...while the daytime mean was 98 $\mu\text{g m}^{-3}$, 50% of which corresponding to RWB particles. By further subtracting the absorbing BC aerosols (14 $\mu\text{g m}^{-3}$) from the RWB mean PM₁₀ concentration value, it is found that...”

25 |

Changes in the domestic heating fuel in Greece: effects on atmospheric chemistry and radiation

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Abstract. For the past 8 years, Greece has been experiencing a major financial crisis which, among other side effects, led to a shift in the fuel used for residential heating from fossil fuel towards bio-fuels, primarily wood. This study simulates the fate of the residential wood burning aerosol plume (RWB smog) and implications on atmospheric chemistry and radiation, with the support of detailed aerosol characterization from measurements during the winter 2013-2014 in Athens. The applied model system (TNO-MACC_II emissions / COSMO-ART model) and configuration used, ~~accurately predicts~~reproduces the measured frequent nighttime aerosol spikes (hourly $PM_{10} > 75 \mu g m^{-3}$) and their chemical profile (carbonaceous components and ratios). Updated temporal and chemical RWB emission profiles, derived from measurements, were used, while the level of model performance was tested for different heating demand conditions, resulting to better agreement with measurements for $T_{min} < 9 \text{ }^{\circ}C$. Half of the aerosol mass over the Athens basin is organic in the submicron range, 80% of which corresponds to RWB (average values during the smog period). Although organic particles are important light scatterers, the direct radiative cooling of the aerosol plume during ~~the~~-wintertime is found low (monthly average forcing of -0.4 W m^{-2} at the surface), followed by a minor feedback to the concentration levels of aerosol species. The low radiative cooling of such a strong air pollution event ~~is~~ is attributed to the timing of the smog plume appearance, both directly ~~-(important interactions with the~~-long-wave radiation increases during ~~the~~-nighttime)-emission- and indirectly, ~~(i.e.~~-the mild effect of the residual plume on solar radiation during the next day, due to ~~the~~-removal and dispersion processes).

1 Introduction

30 Biomass has been traditionally used as a fuel for residential heating in several regions with cold climates, such as the Alpine mountain range and Scandinavia (Yttri et al., 2005; Molnar et al., 2005; Puxbaum et al., 2007; Herich et al., 2014). The European incentives to use biomass fuels, under the assumption of biomass carbon neutrality, have caused a further increase of wood burning for heating purposes in several European cities (Bari et al., 2010; Gu et al., 2013; Waked et al., 2014;

Hovorka et al., 2015). Along with this trend, the financial crisis in Europe during the past nine years has about a significant increase in the market price of conventional fuels, primarily due to ~~exorbitant~~very high taxation. In Greece, the respective increase on the non-solid fuel price was in the range of 40–60%. The high market price together with the limited financial capacity of many households has resulted in an excessive use of biomass for domestic heating (Sarigiannis et al., 2014, 5 Fourtziou et al., 2017).

Several studies have focused on the impacts of residential wood burning (RWB) emissions on air quality. Fuller et al. (2013) reported on aerosol (PM₁₀) contributions from residential wood burning (hereafter also called ‘RWB smog’) in three major European cities: London, Paris and Berlin and found that, during winter, RWB makes an important contribution to breaches of the daily mean EU limit and PM₁₀ contribution may surpass that of road traffic. In Portugal, wood combustion is 10 estimated to comprise 60% of residential energy use, but to account for almost 99% of domestic PM₁₀ emissions (Borrego et al., 2010). An emission inventory constructed for Greece by Faneli and Assimakopoulos (2016) showed that regarding residential emissions, 67% of the emitted CO in Greece originates from fireplaces. At the European level (EU-28), fuel combustion in the residential combustion sector (commercial, institutional and household) is the major source of primary 15 PM_{2.5} and PM₁₀, as well as black carbon (BC) emissions contributing 56, 40 and 46 % respectively (EEA, 2016). Residential combustion (mainly wood) accounts for 3-20% (2-30%) of ambient annual mean PM₁₀ (PM_{2.5}) levels in different European regions, with maximum contributions of up to almost 30% (30%) for the winter period mean value (Viana et al, 2016).

The type of areas examined regarding wood burning impacts on air quality, may range from a specific residential area or a suburb (Hellen et. al., 2008 and Yli-Tuomi et. al., 2015), to a large city such as London (Fuller et. al., 2014, Young D.E. et. al., 2015) and to a regional level, such as Flanders (Maenhaut et. al., 2012). They all agree on a substantial contribution of 20 RWB to PM mass, even when the use is for secondary heating. Wood combustion in open fireplaces emits a mixture of soot (often used interchangeably with BC and/or EC; e.g. Maricq, 2014) and organic carbon (OC), with the latter being the major component of RWB aerosol (Bolling et al., 2009). The contribution of wood combustion to OC is found up to 41% of OC in the PM₁₀ fraction during the wintertime in Zurich (Szidat et al., 2007) and up to 50% in Lombardy (Piazzalunga et al., 2011). In Switzerland, during winter, BC from RWB contributes on average 24–33% to measured BC levels, a noticeable high 25 fraction as the contribution of wood burning to the total final energy consumption is less than 4 % (Herich et al., 2014). Saffari et al. (2013) showed the connection between wood burning and increased levels of PM during the winter in Thessaloniki, while Sarigiannis et al. (2014) further quantified an average increase in PM₁₀ (and PM_{2.5}) due to biomass burning of up to 43µg m⁻³. In Athens, the intense use of open fireplaces and woodstoves for domestic heating during the last 30 years has caused notable increases in the ambient PM concentrations, with the contribution from biomass combustion reaching up to 70% of the PM mass concentration during extreme pollution episodes. The relevant five-year measurements study showed also an increase of 80% in BC due to increased wood burning (Paraskevopoulou et. al., 2014). The chemical footprint of these smog events has been well established via specialized aerosol measurements, e.g. significant correlations between PM_{2.5} and levoglucosan, or high fine mode particulate potassium, demonstrating that, during wintertime, wood

burning in Athens could be responsible for $PM_{2.5}$ levels higher than $45 \mu g m^{-3}$ (Fourtziou et al., 2017; Paraskevopoulou et al., 2014).

Modeling applications devoted to RWB are less numerous and primarily focused on source apportionment and sensitivity to emissions. Simpson et. al. (2007) pointed out that the missing wood-burning contributions may explain the discrepancies between simulations and observations for wintertime OC. Fountoukis et. al. (2014) estimated a decrease in fine organic aerosol (up to 60% in urban and suburban areas during winter), elemental carbon (30-50% in large parts of Europe) and $PM_{2.5}$ mass (15-40% during winter in continental Europe), by replacing current residential wood combustion technologies with pellet stoves, underlining the high sensitivity to emissions. This increasing importance of RWB is also recognized by the revised higher estimates of the latest version of the TNO emission database ([Denier van der Gon et al., 2015](#)), as far as particulate matter is concerned. The inventory indicated that about half of the total $PM_{2.5}$ emissions in Europe are carbonaceous aerosol and identified RWB as the largest organic aerosol source. Moreover, the authors note that while emissions of particulate matter or carbonaceous aerosols are notoriously uncertain, a revised bottom-up inventory was constructed with emphasis on residential wood combustion, which was previously significantly underestimated.

There can be a multitude of impacts of the increasing carbonaceous aerosol contribution to PM as a result of increasing importance of RWB. Exposure to ambient particulate matter has been associated with a range of negative health effects, including increased morbidity and mortality from pulmonary and cardiovascular diseases (Bolling et al., 2009). Recent epidemiologic studies have displayed the risks of exposure to increased levels of carbonaceous aerosols, a major constituent of wood smoke, revealing notable associations with the aforementioned diseases (Ostro et al., 2009; Lipsett et al., 2011; Krall et al., 2013). In a systematic review from short-term epidemiological and cohort studies, it was found that BC is a better indicator of harmful particulate substances from combustion sources than undifferentiated particulate matter mass and it may operate as a universal carrier of a wide variety of chemicals of varying toxicity (Janssen et al., 2012).

Another important aspect of wood burning particles and other aerosols is their direct and indirect effect on atmospheric physics and dynamics. Carbonaceous species, composed of both light-absorbing BC and light-scattering OC, are well recognized contributors to radiative forcing (RF) (Novakov et al., 2005), and the fraction of OC/EC (or OC/BC) becomes decisive with respect to the sign of contribution (Baumer et al., 2007). BC has been considered as the second most important climate forcing agent from human sources in the present-day atmosphere, behind carbon dioxide (Bond et al., 2013). Several studies concentrated on the direct radiative effect of aerosols and their impact on ground temperature (Vogel et al., 2009; Stanelle et al., 2010; Bangert et al., 2012; Lungren et al., 2012; Athanasopoulou et al., 2013). Severe winter haze events over the heavily polluted North China Plain and over Eastern China were found to have a significant negative RF ranging from 20 to $140 W m^{-2}$ (Gao et al., 2015; Zhang et al., 2015). Focusing on a carbonaceous-rich aerosol loading induced by wildfires in Greece, Athanasopoulou et al. (2014) revealed a negative impact on the surface radiative budget in the order of $10 W m^{-2}$ (three day average) and a reduction of surface temperature by 0.5 K over land. The radiative effect of aerosols on [longwave long-wave](#) radiation is less examined. Stanelle et. al. (2010) concentrated on dust episodes over West Africa and found an average increase of $70 W m^{-2}$ for the [longwave 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 short-wave case, which was, however, nonlinear. Panicker et al. (2008) found a positive surface forcing of $6-9 \text{ Wm}^{-2}$ over an urban environment in Prune, India.

Finally, aerosols have an indirect effect on meteorology and climate by acting as cloud condensation nuclei (CCN) and ice nuclei (IN) in aerosol-cloud interactions (Kanakidou et al., 2005, Bangert et al., 2011). To the best of our knowledge, there has been no study so far addressing the direct radiative effects of RWB smog.

The current study focuses on the winter 2013-2014 in Athens, when several severe RWB smog events were recorded. The comprehensive, online-coupled, modelling system COSMO-ART was used to quantify the effect of wood burning on aerosol levels and aerosol chemistry, as well as on radiation during this period. Detailed chemical measurements of concentrations, speciation and fractions related strictly to wood burning, were available from the longest winter measuring campaign of RWB smog to date (e.g. Paraskevopoulou et al., 2014; Fourtziou et al., 2017). Within this paper we want to address the following topics: 1. Assessment of the emission data from RWB, 2. Contribution of RWB to the total aerosol load and chemistry, 3. Calculation of the optical properties of RWB aerosol and 4. Impact of RWB smog on radiation.

2 Data and Methods

2.1 Experimental data

Since winter 2012, several intensive campaigns have been designed and carried out at the official aerosol monitoring site of the National Observatory of Athens (NOA), at its central premises at Thissio (37.97°N , 23.72°E), in Athens' city center. Thissio is an urban background site, representative of Athens' average atmospheric load. Details about the site are provided in Paraskevopoulou et al. (2015) and Fourtziou et al., (2017). For the needs of this modeling study, measurements were taken from the campaign in winter 2013-2014 (16 December, 2013 to 21 February, 2014), during which unique datasets of wood burning indicators were acquired (Fourtziou et al., 2017).

In particular, a time series of hourly PM_{10} is used here taken from a PM_{10} beta-attenuation analyzer. PM_1 chemical composition of non-refractory aerosol particles (organics, sulfate, nitrate, ammonium and chloride) was obtained by an Aerosol Chemical Speciation Monitor – ACSM. PM_1OC was obtained via thermal optical transmission technique, using a Sunset Laboratory Inc. (Oregon) carbon analyzer (see Paraskevopoulou et al., 2014) and PM_1BC by a portable aethalometer (AE-42; Magee Scientific, at 7 wavelengths: 370, 470, 520, 590, 660, 880 and 950 nm). Details about the aforementioned measurements and techniques for the specific campaign can be found in Fourtziou et al., (2017). Among the results described there (and references therein) is the decomposition of the BC time series into BC associated with fossil fuel (BC_{ff}) and wood burning (BC_{wb}), enabled by the wavelength dependence of the BC measurements. Standard meteorological parameters (relative humidity, temperature, rain, wind speed and direction) are measured on a routine basis at the station.

2.2 Model framework and setup

COSMO-ART is a regional atmospheric model which couples online meteorology, chemistry, and aerosol dynamics. COSMO is the operational numerical weather prediction model of the German and other European weather services (Baldauf et al., 2011) and is used as a regional climate model in a modified version CCLM (Rockel et al., 2008). ART (Aerosols and Reactive Trace gases) is the chemistry extension of COSMO. Detailed descriptions of the model, the physic-chemical characteristics of the aerosol modes and the parameterizations of feedbacks of aerosols on radiation, temperature, cloud and ice condensation nuclei (CCN and IN), are given in Vogel et al. (2009), Bangert et al. (2011; 2012), and Rieger et al. (2014). The model domain chosen in this study is the extended area of Greece, centered on Athens, which is the region of interest. The setup of the current model application is largely based on the pilot study of the COSMO-ART application over the same domain (Athanasopoulou et al., 2014) and is given in Table 1. ~~The atmospheric pressure and precipitation parameters were optimized with respect to the high spatial resolution of the current application (0.025°).~~ The simulations were carried out for the period 17 December 2013 till 22 January 2014, providing hourly outputs directly comparable with the aforementioned in-situ, surface level measurements (Section 2.1). The first two days of all model runs (Table 2) were used as spin-up to dampen the effect of the ~~constant-uniform (in space) and constant (in time)~~ initial conditions (e.g. for SO₂ and aerosol species).

Hourly anthropogenic emissions of gases (CO, NH₃, NMVOC, NO_x and SO₂) and aerosols (PM₁₀, PM_{2.5}) are based on the TNO-MACC_II emission inventory for Europe (van der Gon et al., 2010; Kuenen et al., 2011; 2014; Denier van der Gon et al., 2011). Data are provided at a high-resolution (7×7 km²) and reflect emissions from industrial sources, road (and off-road), rail, air and other transport, waste treatment, agriculture and residential combustion.

The processing of this data set for COSMO-ART applications includes a source-specific speciation of NMVOCs and PM (BC, OA, SO₄, rest), the application of time profiles (for diurnal, day-of-week and seasonal variability), and a mapping onto the simulation grid (Knote et al., 2011). For the present study, emissions representative of 2009 (latest year available in the inventory) were used with no further scaling to the simulation period. ~~A recent study on air pollution emissions in Greece based on actual data (Fameli et al., 2016) shows that with respect to residential heating, no clear trend from 2009 to 2012 (latest available data) is evident for the consumption of different type of fuels, except for wood which is increased. With respect to the road sector, similar estimations cannot be performed, since the latest available data refer to 2010.~~ Thus, ~~in~~ in the frame of the current study, the prescribed wood combustion emissions and aerosol optical properties (Vogel et al., 2009) have been ~~processed and~~ revised, according to observational findings representative for the area ~~of interest and period of interest~~ and the wintertime periods of the ongoing crisis (Sect. 2.3 and 2.4).

2.3 Modifications of the aerosol emissions from residential combustion

Non-industrial (residential and agricultural) combustion is spatially distributed according to population density and the proximity to wood (Kuenen et al., 2014). 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).

5 | Unlike the emission data for the gaseous species, which are officially submitted by Greece (EEA, 2011), [the PM₁₀ \(and PM_{2.5}\) emission rates provided by the TNO-MACC II emission inventories](#) are not based on real data, but solely on model approximations (IIASA, 2012), thus significant improvements are feasible upon real data availability. For instance, changes in the temporal emission profiles can have a great impact on the air quality model results for Europe, especially with respect to residential combustion (Denier van der Gon et al., 2011). More specifically, the diurnal profiles suggested therein for the residential combustion sector, have a peak in the morning (black line in Figure 1a), while the common practice in Greece is to use fireplaces mainly in the evening. Furthermore, space heating is more intense on weekends, opposite to the respective suggested weekly variation (black line in Figure 1b).

Therefore, both the hourly and weekly cycles applied to wood combustion emissions were revised to better match common practices in Greece. In particular, assuming that the temporal variation of BC_{wb} closely follows the temporal profile of the corresponding emissions during winter when the planetary boundary layer (PBL) dynamics is moderate, hourly observations of this well-defined RWB index, normalized with respect to the average value, were used for the revision (green line in Fig. 1a). The average profile was derived from the long term monitoring of BC, during the 2013–2015 winter periods. In this way, the two primary peaks in the standard profile were replaced by a much more realistic profile, with an intense peak in the evening hours. In order to revise the original weekly cycle for residential wood combustion, the same data were used, but only during the peak activity period of the fireplaces (20.00-02.00 LST). A representative day-of-week cycle was derived from the observations, based on the median values (to minimize the influence of possible outliers e.g. holiday weekdays). Again, the weekly cycle (green line in Figure 1b) is now consistent with the increased residential wood combustion in Athens during the weekends. It should be noted that the revised temporal cycles were applied only to the aerosol fraction of wood combustion (98% of total combustion aerosol emissions), while gases were assumed to be emitted in equal amounts by wood (revised profiles) and non-wood fuel combustion (original profiles). Thus, the final profiles were calculated from the respective weighted means.

Furthermore, the original chemical profile of aerosol emissions from non-industrial combustion (20% BC, 40% OA and 40% others) was modified based on the chemical composition of aerosol spikes ($> 75 \mu\text{g m}^{-3}$, i.e. [EU-National alert value for the whole population: Joint Ministerial Decision by FEK 3272B/23-12-2013](#)) during night time. Measurements show that the aerosol mass originating from fireplaces is primarily composed of OA (80%), while the rest is equally partitioned into BC and other species. This doubling of the OA mass is in line with a recent study on particulate emissions, based on the latest rise in residential wood combustion in Europe (Denier van der Gon et al., 2015) and is related to the semi-volatile organic matter that is instantaneously formed after emissions are exposed to the cooler ambient temperature. Furthermore, the

currently applied emission ratio of BC/OC (0.12), derived from the measurements at Thissio, is close to the average of 11 studies (0.16 ± 0.05) on emission factors for RWB (fireplaces), presented in Szidat et al. (2007).

A representative map of the modified emission rates for the area of interest is shown in Figure 2. These rates, combined with the temporal profiles (Figure 1), can be transformed to the hourly emission rates from wood burning for any hour and day of a typical winter week in Athens, as represented in the revised TNO-MACC_II emissions database. As expected Given the basic proxy used for the spatial allocation of emission totals (population density), emission values have a maximum at the urban core, where aerosol emissions from RWB represent around 90% of the total value. In particular, the hourly emission rate of aerosol during a smog episode in the Athens city center is in the range of $3.5 - 4.5 \text{ kg hr}^{-1} \text{ km}^{-2}$. It is possible, though, that an over-allocation of emissions in the city centre occurs, as already described in previous studies (Timmermans et al., 2013; Denier van der Gon et al., 2014).

The revised TNO database for Athens was used for the revised run of the current study (case 2 in Table 2). A comparative run using the original TNO-MACC_II database was also performed (case 1 or baseline) to evaluate the level of improvements achieved. It should be noted that an artificial increase in the mass of residential aerosol emissions due to the switch from residential heating from fossil fuels to wood burning was not attempted, as the comparison among model outputs and observations during the smog period (Sect. 3.1.1.) confirmed the magnitude of the original daily PM_{10} emission rates for residential combustion.

2.4 The aerosol optical properties

The properties of atmospheric particles (chemical composition, liquid water content, optical properties) determine their effects on short- and long-wave radiation (scattering and absorption). COSMO-ART incorporates prescribed values for the single scattering albedo, extinction coefficient (in $\text{m}^2 \text{ g}^{-1}$) and asymmetry factor for the five ultrafine aerosol mixtures it handles (pure soot, aged soot coated with soluble material in the nucleation and in the accumulation mode and purely soluble mixture in the nucleation and in the accumulation mode).

For the purpose of the present study, the calculation of the aerosol optical properties is based on observational data collected in Greece during the 2013-2014 RWB smog episodes. In particular, the average surface chemical composition of ultrafine aerosols in Athens (pure soot: $2.8 \mu\text{g m}^{-3}$, water soluble mixture of sulfate, nitrate, ammonium and organics: $22.2 \mu\text{g m}^{-3}$), the local relative humidity (50-70 %) and an average mixing layer height (600 m a.g.l., Gerasopoulos et al., 2017), were used to feed the OPAC software (Hess et al., 1998), which then - by applying the Mie theory - provides the respective optical properties for 61 wavelengths between 0.25 and 40 μm (grey lines in Figure 3). These values have been applied to the eight wavelength bands simulated by COSMO, which cover the spectral range from 0.25 to 104.5 μm (green lines in Figure 3) and were used for the case 2.

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. On the contrary, the smooth reduction with increasing wavelength in the single scattering albedo (SSA) of pure soot, as well as the abrupt decrease from 1 to 3 μm in the

water soluble aerosol modes, are consistent with the findings of Mishra et al. (2014; 2015) for the polluted air masses over the Mediterranean, as well as ~~by~~ with the respective model values used in the study by Takemura et al. (2002) for carbonaceous and sulfate aerosol.

3 Results

5 | The ~~winter 2013-'14 period studied~~ can be characterized as a relatively mild ~~winter period~~. December 2013 was only the 34th coldest December of the period 1897-2014 (NOA records for Athens), with a daily mean temperature of 10.7°C and minimum temperatures (T_{\min}) from 4.9 to 11.5 °C. January 2014 was the warmest January since 1897. The mean daily temperature was 12.5°C. Daytime peaks reached values above 16°C, while nighttime temperatures were greater than 10°C for 75% of the days. The coldest period of this month was 3-5 January, with T_{\min} from 7.3 to 8.2 °C. The relative humidity
10 | during the period was generally above 50% reaching values above 90% during several precipitation events, which took place on 26-27 December 2013, 31 December 2013 – 2 January 2014, 05-06 and 15-16 January 2014 (Figure 4a).

The wind speeds during this period were rather low (usually from the northern sector), preventing the dispersion of pollution. The average wind speed was 2.7 ms⁻¹, with stronger winds (5 - 8 ms⁻¹) from the north direction on 17, 27, 31 December 2013 and 07, 15 and 19-22 January, 2014. An exception with strong (up to 8 ms⁻¹) ~~southeastern~~ south-eastern winds occurred
15 | on 21 January, when a Saharan dust intrusion took place. This event is well documented by satellite retrievals of Aerosol Optical Depth (<https://giovanni.sci.gsfc.nasa.gov/giovanni/>), as well as BSC-DREAM8b model outputs (http://www.bsc.es/ESS/nmmb_bsc-dust).

The meteorological conditions favored the accumulation of smog over Athens during the period 19 December 2013 – 05
20 | January 2014 (mean maximum nighttime hourly PM₁₀ measured concentrations of 103.7 µg m⁻³). This time frame, excluding the rainy days mentioned above, constitutes the intense smog period of the current modeling study, analyzed in Sect. 3.1.1. An interesting case to study aerosol spatial fields (Sect. 3.1.2) is 4-5 January (from noon to noon), as this was the most polluted smog event of the whole period (the maximum nighttime PM₁₀ hourly measured value was 167.7 µg m⁻³) and showed a good model performance (the maximum modeled nighttime PM₁₀ value was 202.2 µg m⁻³). A mild winter day (January, 7 noon – 8 noon, T_{\min} = 10.6 °C) with low smog influence (BC_{wb} equals 44% of BC_{tot}) according to measurements
25 | and predictions (Figure 4c), is also examined for comparison purposes, using results from the case 3.

The mean radiative aerosol effect under smog conditions is discussed in Sect. 3.2, together with the maximum impact during the intense smog event defined above. Cloudless conditions during this day with no interferences by cloud-radiation interactions as well as the good representation of aerosol observations gives credibility to the model results with respect to the direct radiative effects of wood burning smog.

3.1 Impacts of residential wood burning (RWB) on atmospheric aerosol mass and chemistry

3.1.1 Aerosol model performance under smog influence

The applied model system as well as its configuration and methodology for the smog period simulation is here examined and evaluated through: (a) the reproduction of the mean and peak values of mass concentrations of PM₁₀, PM₁OA and PM₁BC measurements, (b) the degree of correlation with the respective observations, (c) the values of representative aerosol ratios (RWB fraction, OC/BC, BC/TC, TC/PM₁₀, OA/PM₁₀ and BC/PM₁₀) and (d) the reproduction of the observed diurnal cycles of PM₁₀, PM₁OA and PM₁BC. The model's ability to reproduce the mean values of mass concentrations is quantified by calculating the Mean Bias (MB), the correlation coefficient (r^2) and the Mean Absolute Normalized Gross Error (MANGE), for cases 1 and 2 (baseline and revised simulation). For the peak values, the Paired Peak Estimation Accuracy (PPEA) is used. A summary of the model skills is given in Table 3 - Table 5, while the mathematical formulation of the applied statistical parameters is given in Appendix A.

The mean value of PM₁₀ observed during the intense smog period was 45.2 $\mu\text{g m}^{-3}$, which is nicely captured by case 2 (50.8 $\mu\text{g m}^{-3}$), but overestimated by the baseline (58.7 $\mu\text{g m}^{-3}$), although the total PM₁₀ mass emitted by residential combustion is unchanged within a typical week. Nevertheless, the emitted mass amounts during weekdays (weekends) are reduced (increased) due to the revised weekly cycles (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. Although the mean bias between measurements and the two model runs is similar, the correlation of hourly data with observations is greatly improved in case 2 with an r^2 of 0.66 compared to only 0.24 in case 1 (Table 3). This is again related to the revisited temporal cycles of emissions, which represent the use of wood for space heating purposes in Athens much more realistically. In particular, the observed nighttime PM₁₀ peaks were on average 103.6 $\mu\text{g m}^{-3}$, more than two times higher than the mean daytime maximum (47.8 $\mu\text{g m}^{-3}$). Case 1 failed to represent these values (e.g. morning PPEA equals +173 %), due to the pre-crisis space heating (mainly office) during the working hours (cf. black line in Fig. 1a and Table 4). In contrast, case 2 is greatly improved (80% and 23%, respectively for morning and nighttime PPEA), showing realistic diurnal profiles during the winter period (under crisis), when the mean hourly concentrations of modeled and measured PM₁₀ ranged from 20 (morning) to 100 $\mu\text{g m}^{-3}$ (night; Figure 5a). Overall, the revised run shows improvements in the calculated PPEA values for improved more than the 70% of the day and nighttime PM₁₀ peaks during the intense smog period.

As evident in Table 3, the most important effect of the incorporation of emission revisions into the model runs is the improved representation of the mean submicron mass fractions (PM₁) of organic (OA) and black (or elemental) carbon (BC). In particular, r^2 equals 0.73 and 0.53 for the two components, respectively, both statistically significant at the 99% c.l. The chemical composition of oil combustion (central heating) greatly differs, thus the baseline run (chemical profiles suggested by the TNO) fails to represent winters during crisis years. Similarly to PM₁₀, the hourly peaks of both submicron species cannot be reproduced by the baseline run (Table 4). In case 2, the mean nighttime peak value of PM₁OA is predicted 69.1 $\mu\text{g m}^{-3}$, close to the measured value of 85.1 $\mu\text{g m}^{-3}$, while the outputs from case 1 are unrealistically low (19.6 $\mu\text{g m}^{-3}$). The

PPEA values for both carbonaceous species are significantly lower for case 2, ~~which leads to the improvement of i.e. for~~ the half PM₁OA and of all PM₁BC the daytime peaks during the intense smog period.

The mean diurnal cycles of both carbonaceous species (Figure 5b and c) are ~~satisfactorily represented~~ reproduced by case 2 ($r_{OA}=0.91$, $r_{BC}=0.8$), while case 1 is strongly biased, especially during daytime for PM₁BC (about 5-fold overestimation) and nighttime for PM₁OA (about 3-fold underestimation). The mean hourly evening peak of PM₁OA in the observations was 74.4 $\mu\text{g m}^{-3}$ at 23:00 UTC, ~~and nicely~~ captured by the revised run ($62.7 \mu\text{g m}^{-3}$ at 22:00 UTC, mean hourly bias equals -~~6.562.7~~ $\mu\text{g m}^{-3}$ at 22:00 UTC). The respective findings for PM₁BC are 9.3 $\mu\text{g m}^{-3}$ at 21:00 UTC in the observations and 11.3 $\mu\text{g m}^{-3}$ at 22:00 UTC in the simulation (mean hourly bias equals 3.5 $\mu\text{g m}^{-3}$). The largest diurnal amplitude during a typical smog day was observed for organics for which the mean minimum (daytime) concentration was around 10 times lower than the mean maximum (nighttime), while the day-night difference for PM₁BC was only by a factor of 7. This is related to the role of wood burning in each mass fraction, which is discussed below.

The fact that the model biases in the RWB fraction of the BC (case 2 – case 3), OC/BC, BC/TC, TC/PM₁₀, OA/PM₁₀ and BC/PM₁₀ are only small, indicates that the aerosol composition was accurately represented in case 2 during the intense smog period. The mean values from predictions and observations at the station Thissio are presented in ~~Table 5~~ Table 3. All model findings are in very good agreement with the observations in the city center. In particular, the mean fraction of RWB particles in the total mass of the predicted PM₁BC was 42 (45) %. This is a daily mean, i.e. the hourly fraction ranged from 25% (daytime) to 70% (nighttime) both in the measurements and the predictions (Figure 5c). RWB smog was estimated to comprise up to 50% of PM₁₀ concentrations during the intense smog period in Athens and to account for almost 80% of PM₁ organics (nighttime peak up to 90%; Figure 5b). It is interesting to note that in the case of organics, wood combustion continues to outweigh all other sources during the whole day (daytime value of more than 60%). Thus, the aerosol chemical composition during the economic crisis is completely altered with respect to the chemical profile of wintertime aerosols beforehand.

The OC/BC ratio calculated from the baseline run (1.1) is rather unrealistic, because it reflects typical urban environments, i.e. intense traffic, low biomass burning and limited regional contribution of aged aerosol. In contrast, both measurements (2.9) and revised predictions (2.8) are highlighting the influence by biomass burning and high secondary formation rates of organics (Szidat et al., 2009; Pio et al., 2011; Gianini et al., 2013; Airuse, 2014). The carbonaceous aerosol dominates PM₁₀ (62 %) in wood burning conditions, more than 70% of which corresponds to organics. The latter makes up half of the total PM₁₀ mass. Again, measurements and revised predictions are in agreement. The comparison with available measurements during increased wood burning in the Alpine area (Szidat et al., 2009) reveals similarities for the ratios EC/TC and EC/PM₁₀, while OC/BC and TC/PM₁₀ showed somewhat higher values during those experiments (cf. ~~Table 3~~ Table 5).

Overall, the exploitation of specialized, systematic measurements of BC_{wb} has been crucial for updating the conventional TNO-MACC_II emission database and a necessary step to accurately reproduce the aerosol pollution in Athens during the financial crisis. The revised temporal cycles and chemical profiles of the emissions from residential combustion significantly improved aerosol predictions, especially during the peak hours (daytime for PM₁₀ and PM₁BC and nighttime for PM₁OA).

Outliers (unrealistically high model values) occurred mainly during weekends with high nighttime temperatures outside the intense smog period (e.g. January, 12 and 18, cf. Figure 4) and are further discussed in Sect. 3.1.3.

3.1.2 Representative spatial aerosol fields

Figure 6 depicts the spatial distribution of the daily mean surface aerosol concentrations (PM_{10} , PM_{1OA} and PM_{1BC}) and corresponding fractions (TC/ PM_{10} , RWB and OC/EC), over the greater Athens area during the selected smog (left column) and mild (right column) event.

As seen, PM_{10} levels during these two events differ by more than a factor two (contours in Figure 6a and b), with concentrations during the smog event reaching $140 \mu g m^{-3}$ within the urban core, exceeding the [EU-National](#) alarm value for emission measures ($100 \mu g m^{-3}$; [Joint Ministerial Decision by FEK 3272B/23-12-2013](#)), and being above the standard daily EU limit ($50 \mu g m^{-3}$) over the entire Athens basin. Concentrations during the mild day reach up to $60 \mu g m^{-3}$, again exceeding the daily EU limit in the city center, which demonstrates the impact of RWB on air pollution throughout the whole winter period. This is related not only to the triggered accumulation of pollution due to meteorology and topography of Athens, but also to the secondary organic aerosol formation due to RWB. Indeed, the largest fraction of PM_{10} during the smog event is composed by carbonaceous matter (TC up to 80%, isolines in Figure 6a), 80% of which being organic. This is even more clear in Fig. 5c, where organics are elevated over the whole basin during the smog event (up to $75 \mu g m^{-3}$) and the RWB fraction constitutes between 60% (city outskirts) and 80% (city center) of PM_{1OA} . During the mild period, in contrast, the air pollution is mainly composed by the rest of the aerosol species (sulfates, ammonium, nitrates, rest; isolines in Figure 6b). PM_{1BC} reach very high levels under the RWB influence over the whole basin (daily values from 10 to $18 \mu g m^{-3}$; Figure 6 e), the 60% of which corresponds to BC_{wb} (not shown). On the contrary, PM_{1BC} is found below $8 \mu g m^{-3}$ during the mild day (Figure 6f). The surface gradient of the OC/BC ratio is very smooth, i.e. the effect of RWB on organic production and formation, is regional and independent of the local peaks. This pattern seems to be stable in time, i.e. the high OC/BC value (over 2.5) during the smog episode is similar to the mean value during the extended smog period (Table [Table 5](#)). Values around unity are found over the urban core, under typical heating and traffic-induced conditions (isolines in Figure 6f).

Overall, the aerosol levels at Thissio (all three RWB-affected components) can be considered representative of an extended urban area around the city center. 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 provided by Gratsea et al. (2016), who used CO measurements from several sites (NAPM regulatory monitoring network of Athens), to characterize Thissio as an urban background site, not intensively affected by local traffic and representative of the average background pollution conditions in Athens.

In order to further support the representativeness of the site in a more quantitative way, we calculated the mean, minimum and maximum values of a greater urban area ($118 km^2$, 15 cells; SW corner: 37.94° , 23.67°) and compared them to the model results at the grid point of Thissio. Indeed, the temporal mean (19 December – 22 January 2014) point values of PM_{1BC} , PM_{1OA} and PM_{10} (8.4 , 39.0 , $69.2 \mu g m^{-3}$) are very close to the spatio-temporal averages over the extended urban domain

(7.4, 37.8 and 64.9 $\mu\text{g m}^{-3}$, respectively; difference in the order of 3 - 7%). Furthermore, the hourly point values neither exceed the area peaks nor fall below the minimum values, while they correspond to 66-68% of the maximum hourly concentrations found in the selected area. Lastly, the linear correlation among the site and the mean domain values for all aerosol components is high (r^2 above 0.73, $N=817$) and close to the 1:1 line (slopes from 1.03 to 1.11 and intercepts below 0.8) for all species.

3.1.3 Heating demand and model bias

As already pointed out in Sect. 3.1.1, aerosol concentrations were occasionally overestimated during mild winter weekends (concurrent green spikes and orange lines in Figure 4a). In order to examine whether this is valid for the whole mild period (5-22 January, 2014), the respective mean PM_{10} levels ($T_{\min} > 8-9^\circ\text{C}$) were compared with the daily cycle predicted from case 3 (Figure 7). This run is indeed realistic, i.e. the actual wood burning during mild winter conditions in Greece is overestimated by the TNO-MACC_II database and by the subsequent atmospheric simulations (green line from 5 January and onwards; Figure 4a). Thus, we attempted to estimate a relation between mass concentrations and model bias with temperature conditions. This was performed by introducing the heating energy demand (HD) term, namely the energy needed to heat a home located in Athens. It is defined as the degrees below the base temperature, which is here chosen as 15.5 $^\circ\text{C}$ (Carbon Trust, 2006).

Indeed, the observed hourly concentration levels of submicron aerosol in the city center of Athens during wintertime tend to increase with decreasing temperature (black dots in Figure 8) and PM_{10} nighttime spikes are above 100 $\mu\text{g m}^{-3}$ only when air temperature falls below 8-9 $^\circ\text{C}$. The explanation is two-fold: during cold weather conditions, firstly, the need for heating increases and thus all means of heating including fireplaces and woodstoves are expected to maximize, resulting in high RWB emissions. Secondly, the same conditions coincide with reduced vertical mixing over Athens with mixing layer heights varying between 200 and 400 m, which support the accumulation of air pollutants near the surface.

In order to identify the link between the model discrepancies and the actual space heating demand in Athens, the trend of the nighttime PM_{10} model bias and the daily maximum heating demand (at the T_{\min} of each day) was examined for the whole period excluding the rainy events (green dots in Figure 8). There is a significant correlation between the two ($r^2 = 0.49$), which shows that when the demand for heating decreases (i.e. at milder temperatures) the model tends to overestimate the aerosol concentrations. In particular, all model bias from +25 $\mu\text{g m}^{-3}$ and above occur only when the HD is low ($<6.5^\circ\text{C}$), i.e. when nighttime air temperatures are above 9 $^\circ\text{C}$. Inversely, during the cold days (T_{\min} below 8 $^\circ\text{C}$) ~~that~~ the HD is increased ($>7.5^\circ\text{C}$), and consequently no model overestimation occurs.

This analysis shows the limitations of using average temporal profiles for the calculation of emissions from residential heating to feed model simulations. when the study period is extended and not focused on specific wintertime smog episodes. Evidently, the TNO-MACC_II residential wood combustion aerosol emission rates (using the updated temporal cycles proposed by the current study) are 'ideal' under typical winter conditions that lead to moderate or -increased residential heating demand and during stay-at-home days (e.g. weekdays, holidays). It should be noted that this study has focused on the

crisis period, i.e. a switch occurs in the domestic heating fuel from heating oil to wood, and that for a pre-crisis period or significant change in dominant heating fuels, the TNO-MACC_II emissions from residential heating should be further adjusted.

3.2 Impact of RWB smog on radiation and feedbacks on atmospheric composition

The effect of aerosols on total surface radiation during the simulated winter period was approximated by comparing case 2 with case 4. This is presented as a monthly mean in Figure 9a. As expected, the effect of total aerosol over the extended area of Athens is negative, i.e. the scattering of radiation by particles (organics, sulfates etc.) outweighs absorption by black carbon and the reflection of short-wave radiation (-1.9 Wm^{-2}) outweighs the enhanced trapping of long-wave radiation ($+0.5 \text{ Wm}^{-2}$). Nevertheless, the mean monthly effect does not exceed -1.4 Wm^{-2} at the urban core and by comparing the cases 2, 3 and 4 it is found that only 30% of this effect corresponds to particles from RWB. This means that the mean direct radiative effect (DRE) due to RWB at the surface does not exceed the value of -0.4 Wm^{-2} .

This small negative effect of particles on radiation is explained by the fact that most of the RWB emissions occur during nighttime when only the long-range terrestrial radiation is reflected (positive DRE values up to $+0.4 \text{ Wm}^{-2}$). The role of timing on the aerosol radiative impacts has already been analyzed for dust storm over the eastern Mediterranean (Remy et al., 2015), which stresses the important positive feedback between aerosol and meteorology during nighttime. Besides, the residual plume during the next day is weakened, due to removal and dispersion processes. Indicatively, the nighttime PM_{10} mean during the RWB smog episode (4-5 January, 2014) reached $145 \mu\text{g m}^{-3}$, while the daytime mean was $98 \mu\text{g m}^{-3}$, 50% of which corresponding to RWB particles. By further removing subtracting the absorbing BC aerosols ($14 \mu\text{g m}^{-3}$) from the RWB mean PM_{10} concentration value, it is found that during the peak smog event of the studied period only $35 \mu\text{g m}^{-3}$ corresponded to aerosols scattering in the short-wave, causing a DRE of -0.8 Wm^{-2} at the urban core (Figure 9b).

The related feedbacks on atmospheric composition areis found very small, in consistency with the low DRE value. In specific, the feedback on the mean monthly concentration values (case 2 – case 4), for all aerosol species, is found positive and in the range of $0.4 - 0.6 \mu\text{g m}^{-3}$, which indicatively represent only the 1% and 3% of the total PM_{10}OA or PM_{10}BC mean monthly concentration found at the urban core. This simulated slight increases in aerosol species concentrations due to the aerosol-radiation interaction,s areis associated with an aboutalmost 10 m lower mean height of the PBL height byin case 4 than bycompared to case 2, in accordance to the respective findings in Forkel et al. (2012).

Finally, we performed a run using the optical properties of aerosol from Vogel et al. (2009), which was compared to cases 2 and 3. The differences in the DRE were found to be negligible (not shown). This is explained by the fact that most of the differences that were imposed by the local conditions to the aerosol properties (cf. Sect. 2.4), correspond to the pure soot mode. Nevertheless, the atmospheric particles over an urban area correspond mainly to aged soot (coated with soluble material), a mode whose aerosol properties by Vogel et al. differ from the ones of this study on the long-wave part of the spectrum (cf. Figure 3). However, the wavelength range above $2 \mu\text{m}$ is unimportantly affected by particles, as explained above. Thus, the short-wave scattering in both runs did not change significantly, leading to similar findings for DRE.

4 Conclusions

This study examines the impacts of increasing use of wood burning for domestic heating on air pollution through model-based analysis of a case study in Athens, Greece, as an immediate consequence of the ongoing economic crisis. A cold period ($T_{\min} < 10\text{ }^{\circ}\text{C}$) with intense RWB smog conditions (nighttime PM_{10} hourly peaks from 75 to $173\text{ }\mu\text{g m}^{-3}$) was selected for the characterization of the aerosol levels and chemistry over the inner city center. Two representative events were distinguished for an analysis of the spatial representation of the smog plume during a cold ($T_{\min} = 8.2\text{ }^{\circ}\text{C}$) and a mild ($T_{\min} = 10.6\text{ }^{\circ}\text{C}$) winter day, with high and low nighttime peaks at the Thissio station in the city center (PM_{10} at 173 and $59\text{ }\mu\text{g m}^{-3}$, respectively).

The daily mean PM_{10} concentration at Thissio during the RWB smog period derived from the measurements and the model (in parenthesis) was $45.2\text{ (}50.8\text{)}\text{ }\mu\text{g m}^{-3}$. The respective values for the PM_1OA and BC were $28.4\text{ (}34.4\text{)}$ and $4.6\text{ (}7.1\text{)}\text{ }\mu\text{g m}^{-3}$. PM_{10} levels during the smog (cold) event exceeded the EU daily limit for the whole Athens basin (model outputs). This finding is primarily related to RWB particles, which comprised 50% of the total PM_{10} levels. The impact of RWB on air pollution persisted throughout winter, i.e. exceedances occurred also during the mild days, but confined to the urban core.

According to a spatio-temporal analysis of the current model results, the concentration values at Thissio are representative of an extended urban area, i.e. of the average pollution conditions in Athens. The day-to-night differences in the concentrations of different aerosol components were found to be very large (5-, 10- and 7-fold for PM_{10} , PM_1OA and PM_1BC , respectively) because of the intense RWB activity during nighttime and the changes in PBL. In particular, the observed (simulated) peaks in PM_{10} , PM_1OA and PM_1BC between 21:00 and 23:00 UTC were on average $90.5\text{ (}100.2\text{)}\text{ }\mu\text{g m}^{-3}$, $74.4\text{ (}62.7\text{)}\text{ }\mu\text{g m}^{-3}$ and $9.3\text{ (}11.3\text{)}\text{ }\mu\text{g m}^{-3}$, respectively. These values correspond to almost 70% of the maximum hourly concentrations revealed by the model for the extended urban area, further supporting that measurements at Thissio represent urban background conditions over the Athens basin.

The carbonaceous component (OA+BC) of PM_{10} in Athens during the smog period reached 62 (61) %, half of which corresponding to organic matter. This means that RWB completely alters the chemical profile of PM_{10} over Athens during wintertime, as the RWB smog fraction of PM_1OA (according to the simulation) reached from 60% during daytime to 90% during nighttime and BC_{wb} accounted for 25% during daytime to 70% during nighttime of BC_{tot} (according to both the measurements and simulation). The mean OC/BC ratio at Thissio from measurements (model outputs) was 2.9 (2.8), characteristic of the RWB influence and secondary formation of organics. Again, this value is representative for the whole Athens basin during wintertime, according to the spatio-temporal analysis of the model outputs.

Interestingly, the earth radiative budget was on average not altered significantly despite these high aerosol loads due to a compensation of long-wave and short-wave effects and the fact that RWB emissions are concentrated on nighttime hours when only long-wave effects are present. Overall, the mean direct radiative effect of wintertime RWB smog was estimated as low as -0.4 Wm^{-2} , with a subsequent feedback on the concentration of each of the simulated aerosol species less than $+0.6\text{ }\mu\text{g m}^{-3}$.

For the model to properly capture the wintertime aerosol values observed in Athens, a revision of the residential combustion emission sector was necessary. This was facilitated via a unique, long-term dataset of the wood burning indicator (BC_{wb}) obtained at Thissio station. In particular, the TNO-MACC_II RWB emissions for particulate matter were revised with respect to their temporal and chemical profiles. The updated model configuration was found to greatly improve the prediction of smog episodes during wintertime. 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. 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. Alternatively, an interactive 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.

5 Appendix A

The mean absolute normalized gross error (MANGE) is calculated by A1 and the correlation coefficient (r) by the A2:

$$MANGE = \frac{1}{N} \sum_{i=1}^N \frac{(E_i - O_i)}{O_i} 100\% \quad (A1),$$

$$r = \left[\frac{\sum_{i=1}^N (E_i - \bar{E})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^N (E_i - \bar{E})^2 \sum_{i=1}^N (O_i - \bar{O})^2}} \right] \quad (A2),$$

where E is the estimated (modeled) and O is the observed value of each parameter, paired in space and time for each i of N data pairs. \bar{E} and \bar{O} are the mean values of estimations and observations, respectively.

The paired peak estimation accuracy (PPEA) is calculated as below:

$$PPEA = \frac{1}{N} \sum_{i=1}^N \frac{(EM_i - OM_i)}{OM_i} 100\% \quad (A3),$$

where EM is the estimated (modeled) and OM is the observed peak one-hour value of each parameter, paired in space and time for each i of N data pairs.

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Table 1: Main characteristics of the COSMO-ART configuration and the current model setup. *Italic fonts indicate emissions not considered in the current model runs.*

Formulation / Mechanisms	COSMO-ART
Chemical mechanisms	Gaseous chemistry: RADMKA (Stockwell et al., 1990; Vogel et al., 2009) Inorganic aerosol chemistry: ISORROPIA II (Fountoukis and Nenes, 2007) Organic aerosol chemistry: VBS (Athanasopoulou et al., 2013) Wet scavenging and liquid-phase chemistry (Knote and Brunner, 2013)
Dynamic aerosol processes	Modal Aerosol Dynamics Model for Europe: MADESoot (Riemer et al., 2003) (11 overlapping log-normally distributed modes)
Interaction between aerosols and meteorology	Aerosol effects on radiation and temperature: GRAALS (Ritter and Geleyn, 1992) Aerosol activation (CCN) and ice nucleation (IN) parameterizations (Bangert et al., 2011, 2012)
Initial and boundary conditions	Meteorology: COSMO model application over Europe (7×7 km), driven by the German Weather Service GME global model (Majewski et al., 2002) Gaseous species (CO, HNO ₃ , NH ₃ , NO _x , NO ₃ , O ₃ , NMVOC): global outputs (2.5°×1.9°) from the MOZART model (Emmons et al., 2010) Aerosol species: 1 µg m ⁻³ for sulfates, 0.1 µg m ⁻³ for nitrates, 0.37 µg m ⁻³ for ammonium and 1 µg m ⁻³ for secondary organic aerosol (SOA) in the surface layer* (Athanasopoulou et al., 2013)
Input data	Anthropogenic, agricultural emissions (prescribed): TNO-MACC_II (Denier van der Gon et al., 2010; Kuenen et al., 2011, 2014), <i>Fire emissions (prescribed): GFED v.3 (van der Werf et al., 2010),</i> Biogenic activity (isoprene, a-pinene, a-limonene), <i>Desert dust (3 aerosol modes)</i> , Sea-salt (3 aerosol modes) and DMS production, <i>pollen and volcanic ash uplift and transport: online calculation (Vogel et al., 1995, 2006; Lundgren et al., 2013; Nightingale et al., 2000; Zink et al., 2013; Vogel et al., 2014).</i>
Vertical grid	40 levels (from surface to ca. 23 km; first layer is ca. 20 m thick)
Horizontal domain	Greater area of Greece 18 to 30 E°, 33 to 42 N° (0.025° × 0.025°)

Table 2: Description of modeling runs performed by the current COSMO- ART application from 17 December, 2013 to 22 January, 2014.

	Case	Description	Objective
1	Baseline	Original RWB emission data	Evaluation of the modified emission data from RWB in Greece
2	Revised*	Revised wintertime simulation	Aerosols over Athens during an intense RWB smog period
3	RWB-free*	Emissions from all residential combustion fuels, but wood	Contribution of RWB to the total aerosol load and chemistry and impact of RWB smog on radiation
4	Feedback free <u>No interaction*</u>	The interaction between aerosols and meteorology is switched off	Impact of total aerosol on radiation

* modifications in residential wood combustion emissions and in aerosol optical properties (cf. Sect. 2.3, 2.4)

Table 3: Averaged daily mean values and prediction skill metrics of the aerosol concentrations (and RWB mass fractions) against ground measurements (at Thessio) during the RWB smog period (19 December, 2013 – 05 January, 2014) in Athens. Numbers in bold represent the calculated statistics, given by equations A1 and A2. Mean values and prediction skill metrics of the aerosol concentrations (and selected mass fractions) against ground measurements (at Thessio) during the RWB smog period (19 December, 2013 – 05 January, 2014) in Athens. Numbers in bold represent the calculated statistics. Italic letters indicate maximum values and statistics.

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<u>Component</u>	<u>Parameter</u>	<u>Observations</u>	<u>Case 1 (baseline)</u>	<u>Mean hourly bias</u>	<u>r²</u>	<u>MANGE (%)</u>	<u>Case 2 (revised)</u>	<u>Mean hourly bias</u>	<u>r²</u>	<u>MANGE (%)</u>
<u>PM₁₀ μg m⁻³</u> (332 samples)	<u>Averaged daily mean (smog period)</u>	<u>45.2</u>	<u>58.7</u>	<u>15.0</u>	<u>0.24</u>	<u>95</u>	<u>50.8</u>	<u>19.3</u>	<u>0.66</u>	<u>83</u>
	<u>std. deviation</u>	<u>33.1</u>	<u>28.8</u>							
	<u>RWB fraction (%)*</u>						<u>51</u>			
<u>PM₁OA μg m⁻³</u> (333 samples)	<u>Averaged daily mean (smog period)</u>	<u>28.4</u>	<u>18.8</u>	<u>-8.2</u>	<u>0.39</u>	<u>65</u>	<u>34.4</u>	<u>6.6</u>	<u>0.73</u>	<u>118</u>
	<u>std. deviation</u>	<u>30.9</u>	<u>8.6</u>				<u>22.4</u>			
	<u>RWB fraction (%)*</u>						<u>78</u>			
<u>PM₁BC μg m⁻³</u> (212 samples)	<u>Averaged daily mean (smog period)</u>	<u>4.6</u>	<u>9.9</u>	<u>5.2</u>	<u>0.47</u>	<u>236</u>	<u>7.1</u>	<u>2.5</u>	<u>0.53</u>	<u>121</u>
	<u>std. deviation</u>	<u>3.8</u>	<u>6.7</u>				<u>4.7</u>			
	<u>RWB fraction (%)*</u>	<u>45</u>					<u>42</u>			

*RWB fraction predictions are the outcome of case 2 – case 3 differences. Measurements for the same fraction are only provided for BC.

<u>Component</u>	<u>Parameter</u>	<u>Observations</u>	<u>Case 1 (baseline)</u>	<u>Mean hourly bias</u>	<u>r²</u>	<u>MANGE (%)</u>	<u>Case 2 (revised)</u>	<u>Mean hourly bias</u>	<u>r²</u>	<u>MANGE (%)</u>
<u>PM₁₀</u> μg m ⁻³ (332 samples)	<u>daily mean (smog period)</u>	<u>45.2</u>	<u>58.7</u>	<u>15.0</u>	<u>0.24</u>	<u>95</u>	<u>50.8</u>	<u>19.3</u>	<u>0.66</u>	<u>83</u>
	<u>std. deviation</u>	<u>33.1</u>	<u>28.8</u>				<u>32.7</u>			<u>PPEA (%)</u>
	<u>mean daytime maximum</u>	<u>47.8</u>	<u>115.5</u>				<u>78.1</u>			<u>80</u>
	<u>mean nighttime maximum</u>	<u>103.6</u>	<u>80.9</u>				<u>110.1</u>			<u>23</u>
	<u>%</u>	<u>RWB fraction</u>						<u>51</u>		
<u>PM₁OA</u> μg m ⁻³ (333 samples)	<u>daily mean (smog period)</u>	<u>28.4</u>	<u>18.8</u>	<u>-8.2</u>	<u>0.39</u>	<u>65</u>	<u>34.4</u>	<u>6.6</u>	<u>0.73</u>	<u>118</u>
	<u>std. deviation</u>	<u>30.9</u>	<u>8.6</u>				<u>22.4</u>			<u>PPEA (%)</u>
	<u>mean daytime maximum</u>	<u>23.5</u>	<u>23.9</u>				<u>39.3</u>			<u>103</u>
	<u>mean nighttime maximum</u>	<u>85.1</u>	<u>19.6</u>				<u>69.1</u>			<u>9</u>
	<u>%</u>	<u>RWB fraction</u>						<u>78</u>		

PM₁BC	$\mu\text{g m}^{-3}$ (212 samples)	daily-mean (smog-period)	4.6	9.9	5.2	0.47	236	7.1	2.5	0.53	121
		std. deviation	3.8	6.7			PPEA (%)	4.7			PPEA (%)
		mean daytime maximum	14	27			590	15			258
		mean nighttime maximum	5.2	12.6			63	11.3			38
		% RWB fraction	45					42			
OC/BC		daily-mean (smog-period)	2.9	1.1				2.8			
BC/TC		daily-mean (smog-period)	28	47				27			
TC/PM₁₀		daily-mean (smog-period)	62	48				61			
OA/PM₁₀		daily-mean (smog-period)	50	32				51			
BC/PM₁₀		daily-mean (smog-period)	11	17				11			

[±] Organic aerosol (OA) predictions are divided by 1.6 (Turpin and Lim, 2001), to extract the carbon mass (OC) used for the calculation of the OC/BC ratio.

5 **Table 4: Mean daytime/night-time maximum values and prediction skill metrics of the aerosol concentrations against ground measurements (at Thissio) during the RWB smog period (19 December, 2013 – 05 January, 2014) in Athens. Numbers in bold represent the calculated statistics, given by equation A3.**

<u>Component</u>	<u>Time frame</u>	<u>Observations</u>	<u>Case 1 (baseline)</u>	<u>PPEA (%)</u>	<u>Case 2 (revised)</u>	<u>PPEA (%)</u>
PM ₁₀ $\mu\text{g m}^{-3}$ (332 samples)	<u>daytime</u>	47.8	115.5	173	78.1	80
	<u>night-time</u>	103.6	80.9	-8	110.1	23
PM ₁ OA $\mu\text{g m}^{-3}$ (333 samples)	<u>daytime</u>	23.5	23.9	119	39.3	103
	<u>night-time</u>	85.1	19.6	-58	69.1	9
PM ₁ BC $\mu\text{g m}^{-3}$ (212 samples)	<u>daytime</u>	14	27	590	15	258
	<u>night-time</u>	5.2	12.6	63	11.3	38

10 **Table 5: Averaged daily means of selected aerosol mass fractions against ground measurements at Thissio (Athens) during the RWB smog period (19 December, 2013 – 05 January, 2014) and at Swiss Alpine areas (winter 2005; Szidat et al., 2009). Organic aerosol (OA) predictions are divided by 1.6 (Turpin and Lim, 2001), to extract the carbon mass (OC) used for the calculation of the OC/BC ratio.**

<u>Component</u>	<u>Observations Athens / Alpine area *</u>	<u>Case 1 (baseline)</u>	<u>Case 2 (revised)</u>
OC/BC	2.9 / 3.7	1.1	2.8
BC/TC	0.28 / 0.23	0.47	0.27
TC/PM₁₀	0.62 / 0.89	0.48	0.61

<u>QA/PM₁₀</u>	<u>0.50 / -</u>	<u>0.32</u>	<u>0.51</u>
<u>BC/PM₁₀</u>	<u>0.11 / 0.10</u>	<u>0.17</u>	<u>0.11</u>

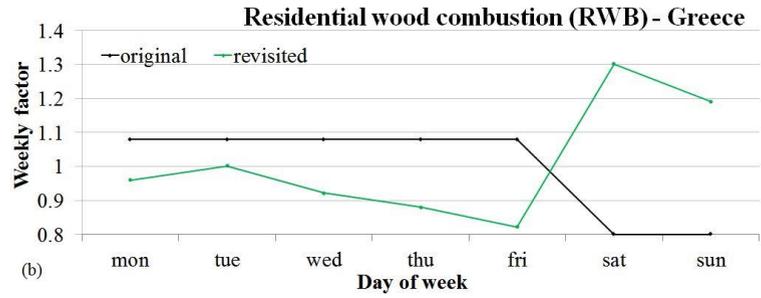
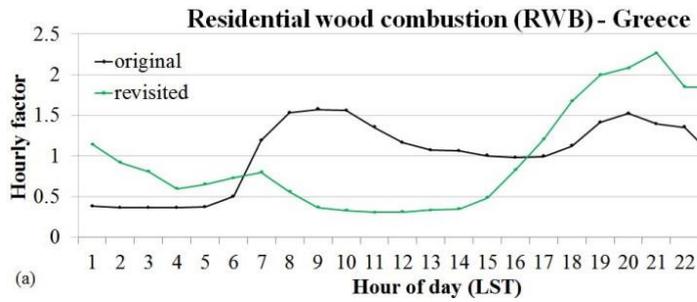


Figure 1: Original (black line) and revisited (green line): (a) diurnal and (b) weekly profiles, applied to the emissions from residential wood combustion (RWB) in Greece (TNO-MACC_II database; Denier van der Gon et al., 2011). The revisited temporal profiles correspond to the mean values of the long-term measurements (2013-2015) of the wood burning fraction of black carbon (BC_{wb}) in Athens, normalized with respect to the average value of each cycle.

5

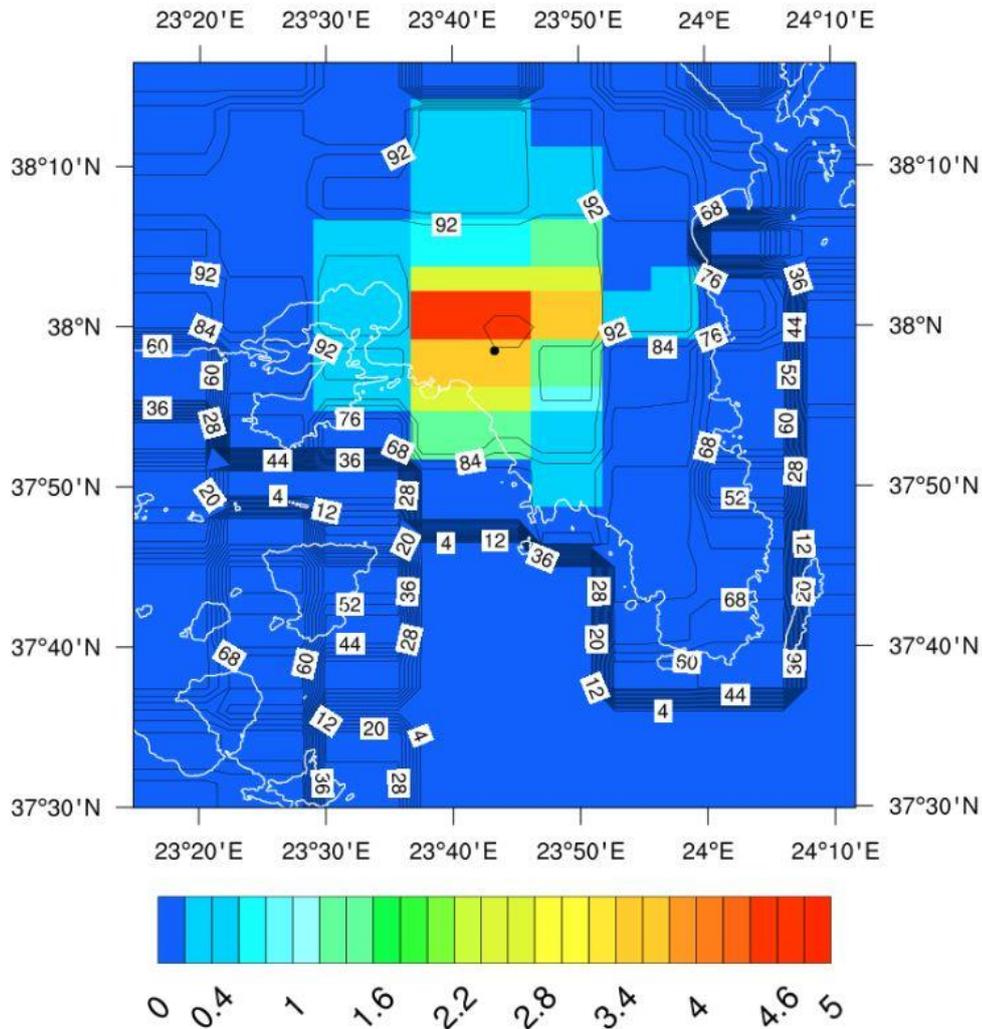


Figure 2: RWB aerosol emission rates ($\text{kg hr}^{-1} \text{ km}^{-2}$) (color contours) and their fraction of the total aerosol emissions (isolines in %) for a night hour of a weekday (Tuesday, 21.00 UTC), as retrieved from the revised emission (TNO-MACC_II) data. It is noted that the spatial resolution of the emissions ($0.125^\circ \times 0.0625^\circ$) is coarser than that of the model configuration ($0.025^\circ \times 0.025^\circ$). The city center of Athens is shown with the black mark.

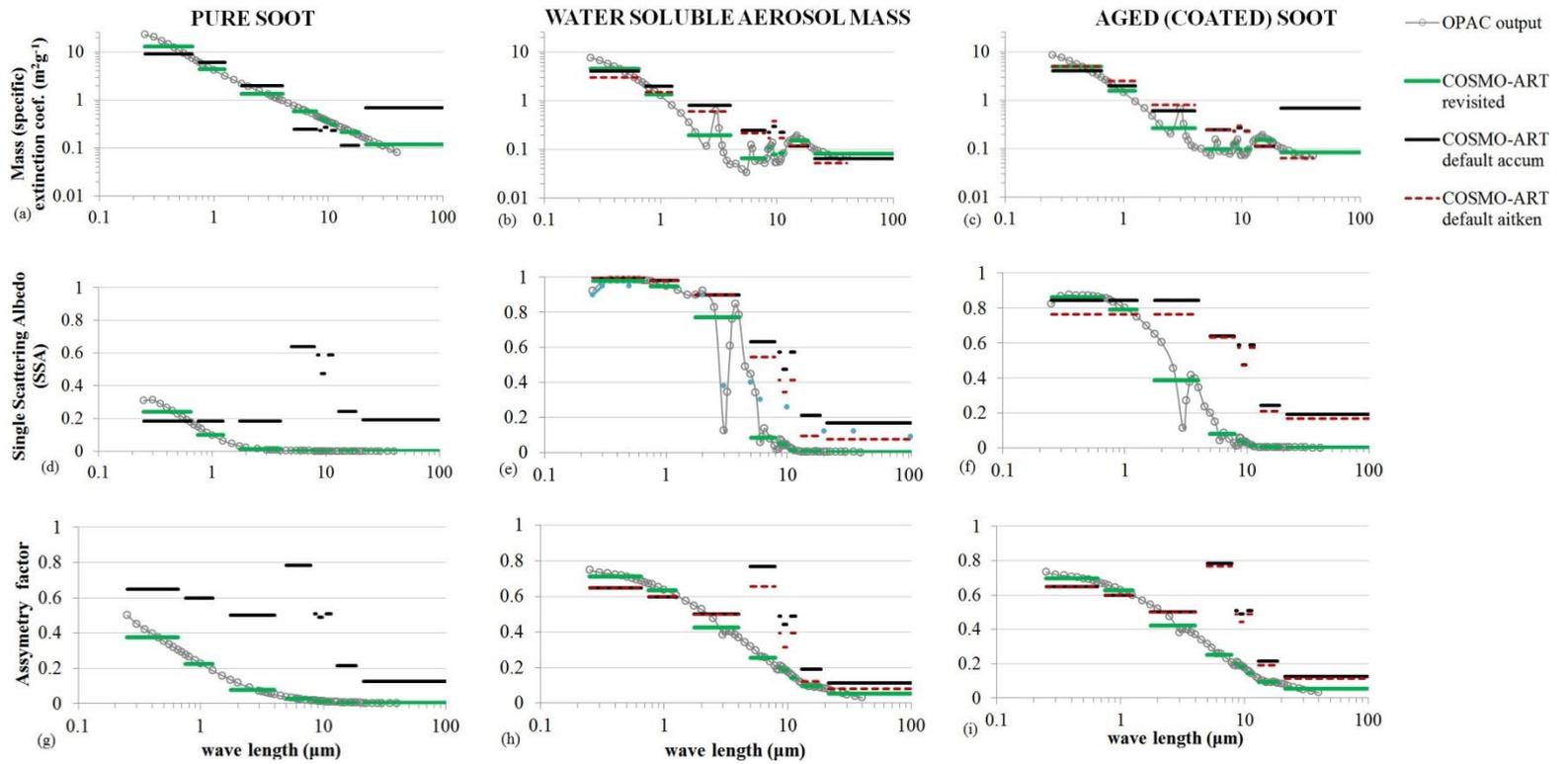
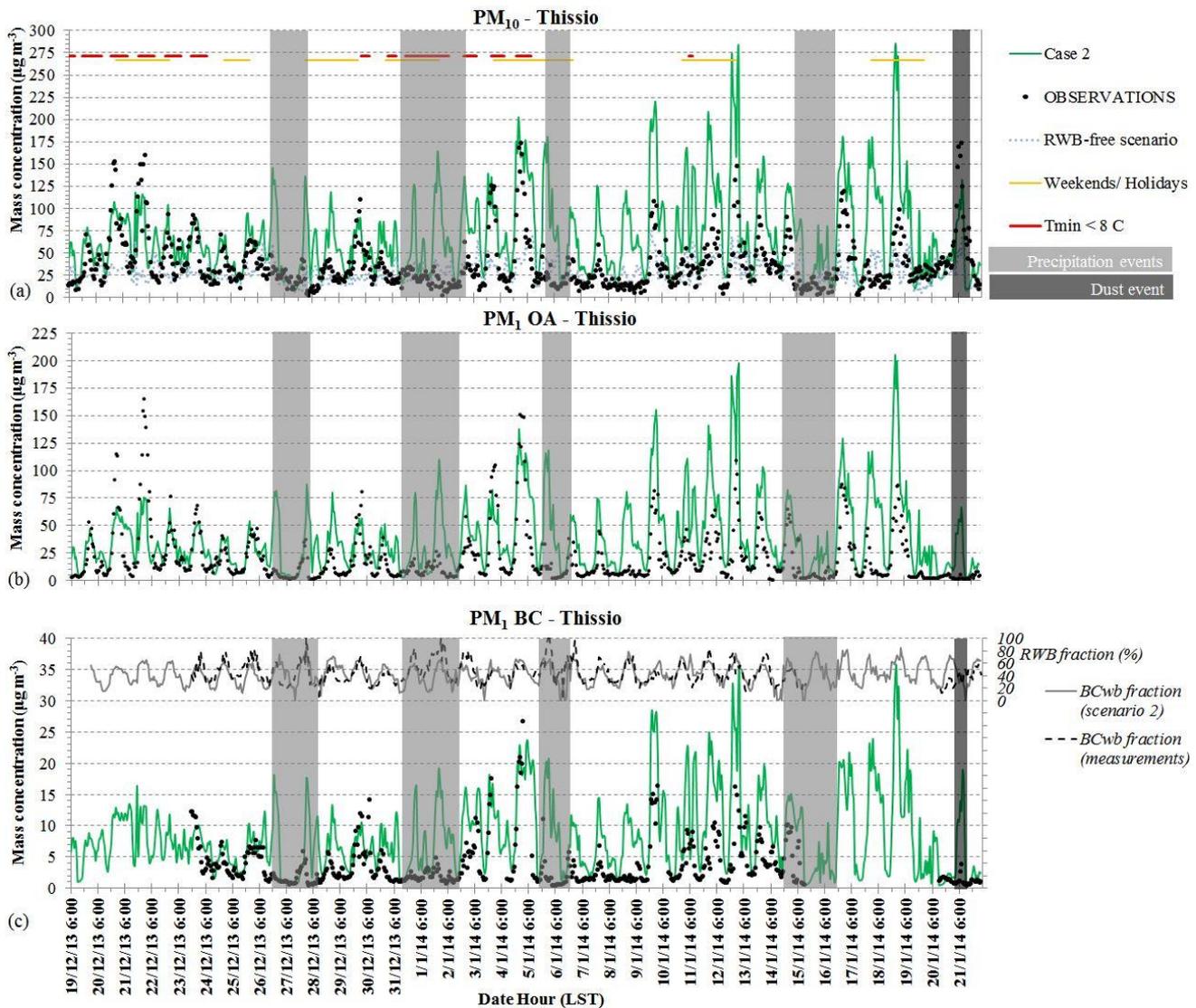
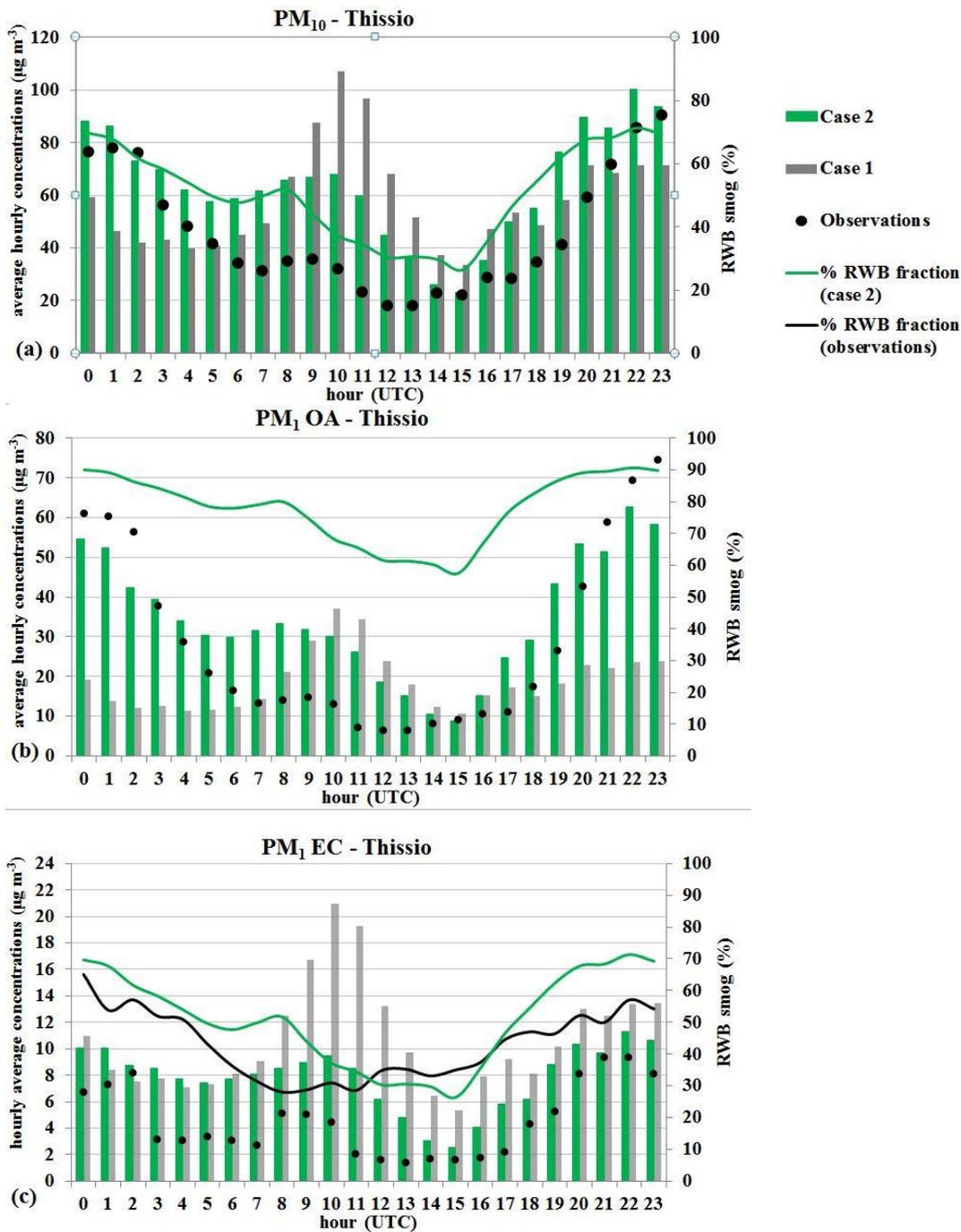


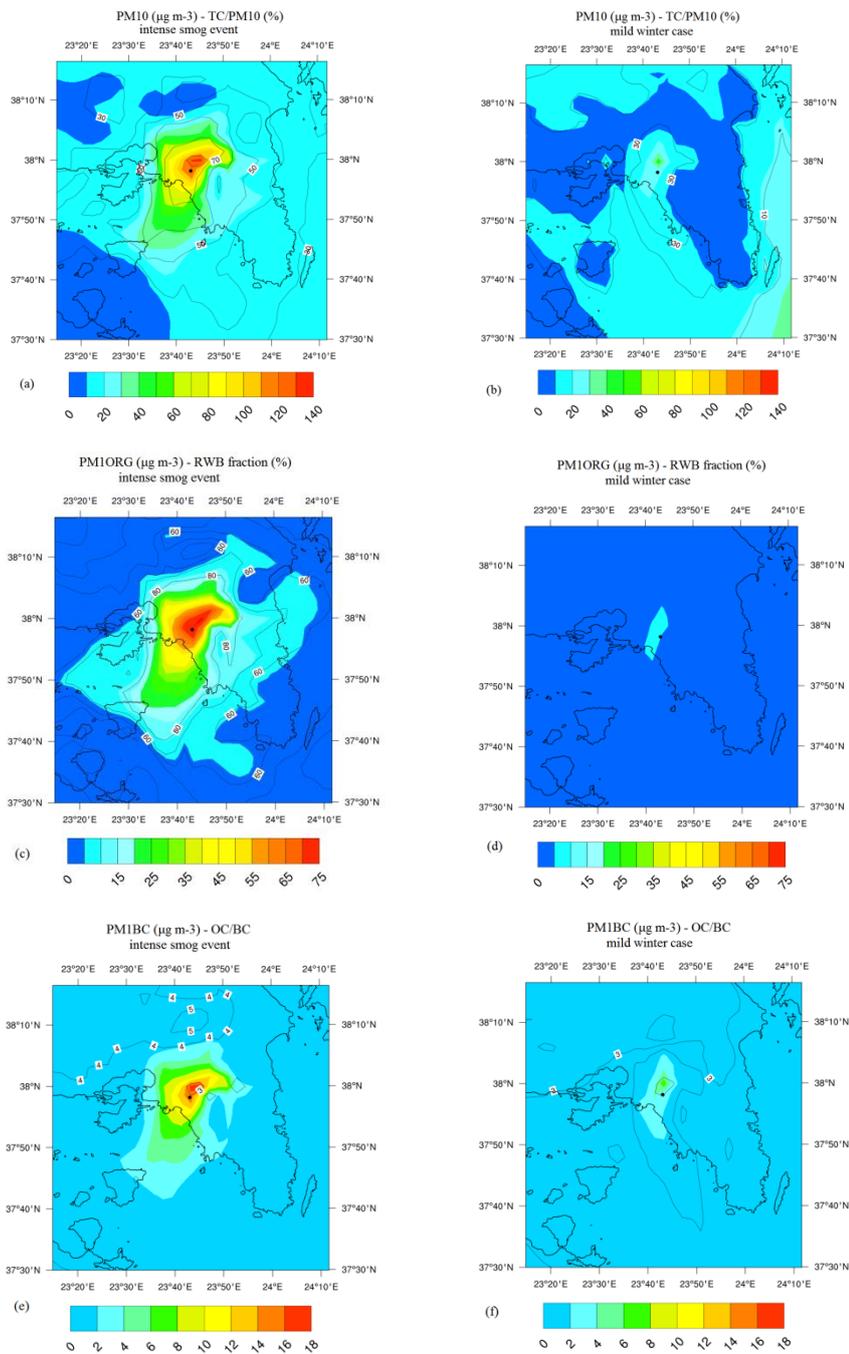
Figure 3: Specific extinction coefficient (a – c), single scattering albedo (d - f) and asymmetry factor (g - i) per wavelength, for different mixtures of submicron aerosol (pure soot, water soluble mass and coated soot): from the COSMO-ART configuration described in Vogel et al. (2009; black and red lines for the accumulation and nucleation mode, respectively). Soot is represented by a single mode), from the OPAC algorithm (grey lines), and the OPAC values adjusted to the COSMO-ART bands (green lines).



5 **Figure 4:** Time-series of the hourly concentrations ($\mu\text{g m}^{-3}$; left axis) of the: (a) total PM_{10} mass, (b) submicron organic aerosol ($\text{PM}_{1,\text{OA}}$) and (c) submicron black carbon ($\text{PM}_{1,\text{BC}}$), from the COSMO-ART application (case 2; green line) and the measurements (black dots). The RWB fraction (%) of black carbon (BC_{wb}), as predicted (case 2) and observed is shown by the grey and the black line, respectively (right axis). The cold nights (intense RWB smog episodes) and the weekends/holidays are shown with the red and the yellow line, respectively. The shadowed areas indicate: the rainy events (in light grey) and the dust event (in dark grey).



5 Figure 5: The mean daily cycle of mass concentrations ($\mu\text{g m}^{-3}$) from measurements (black dots) and from model outputs (case 1 in grey columns and case 2 in green columns) and of RWB fractions (%) from measurements (black line; Only BC_{wb} fraction is measured) and from model outputs (case 2 in green line) for: (a) PM_{10} , (b) $\text{PM}_{1\text{OA}}$, and (c) $\text{PM}_{1\text{EC}}$. All values refer to the site of Thissio (Athens), and they correspond to the intense smog wintertime period.



5 **Figure 6: Mean daily surface concentration ($\mu\text{g m}^{-3}$) fields of: (a) and (b) PM_{10} . Isolines represent the carbonaceous fraction (OA + BC) of PM_{10} (%), (c) and (d) $\text{PM}_{1\text{ORG}}$. Isolines represent the RWB fraction of $\text{PM}_{1\text{ORG}}$ (%), (e) and (f) $\text{PM}_{1\text{BC}}$. Isolines represent the OC/BC ratio. OC is approximated as OA divided by 1.6, a ratio suggested by Turpin and Lim (2001) for urban areas. All maps cover the extended area of Athens (Thessio is marked with the black dot) and derive from model outputs that correspond to the intense smog event (case 1; left column) and to the mild winter case (case 3; right column).**

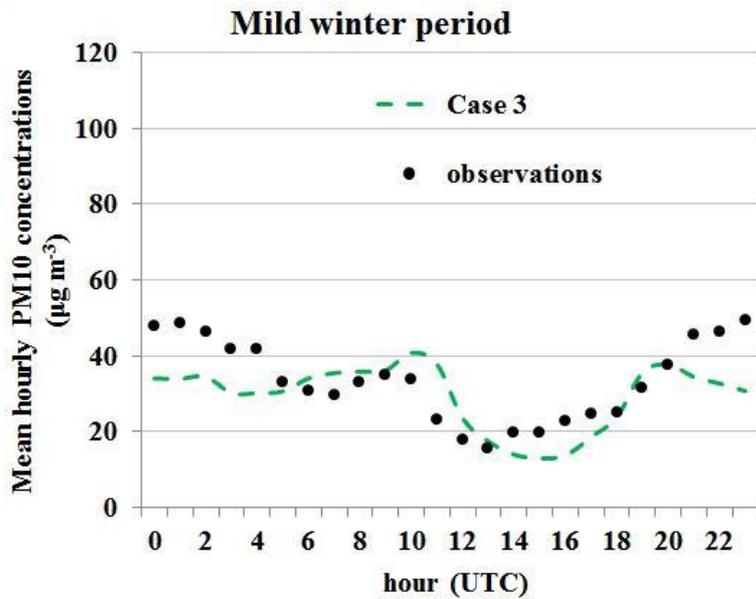


Figure 7: The linear correlation between air temperature ($^{\circ}\text{C}$) and submicron organic aerosol concentration peaks ($\text{PM}_{1\text{OA}}$ in $\mu\text{g m}^{-3}$, in black) and between the heating demand (HD, in $^{\circ}\text{C}$) and the model bias (for $\text{PM}_{1\text{OA}}$ in $\mu\text{g m}^{-3}$, in green), (b) the mean daily cycle of PM_{10} 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).

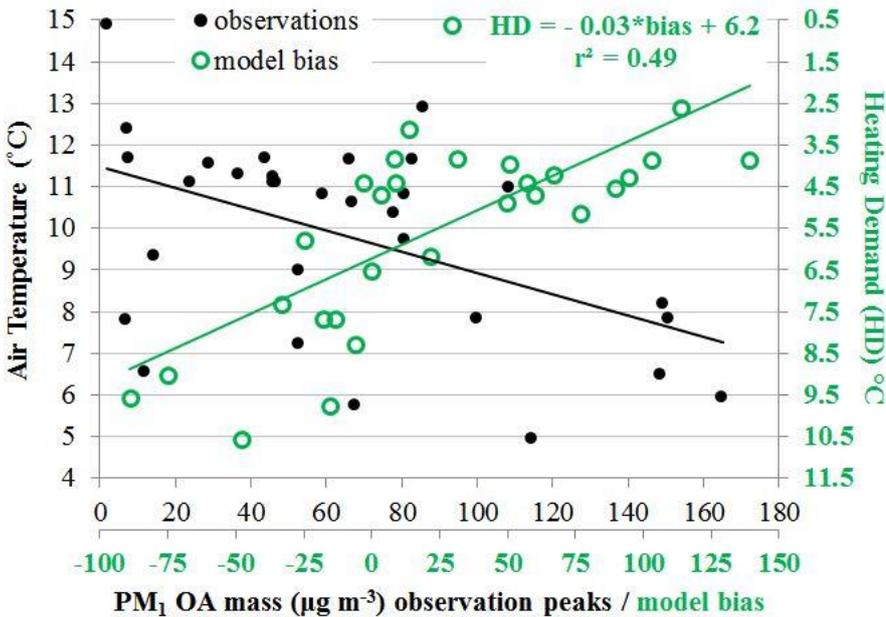


Figure 8: The linear correlation between air temperature ($^{\circ}\text{C}$) and submicron organic aerosol concentration peaks ($\text{PM}_{1\text{OA}}$ in $\mu\text{g m}^{-3}$, in black) and between the heating demand (HD, in $^{\circ}\text{C}$) and the model bias (for $\text{PM}_{1\text{OA}}$ in $\mu\text{g m}^{-3}$, in green). All data refer to the Thissio site (Athens).

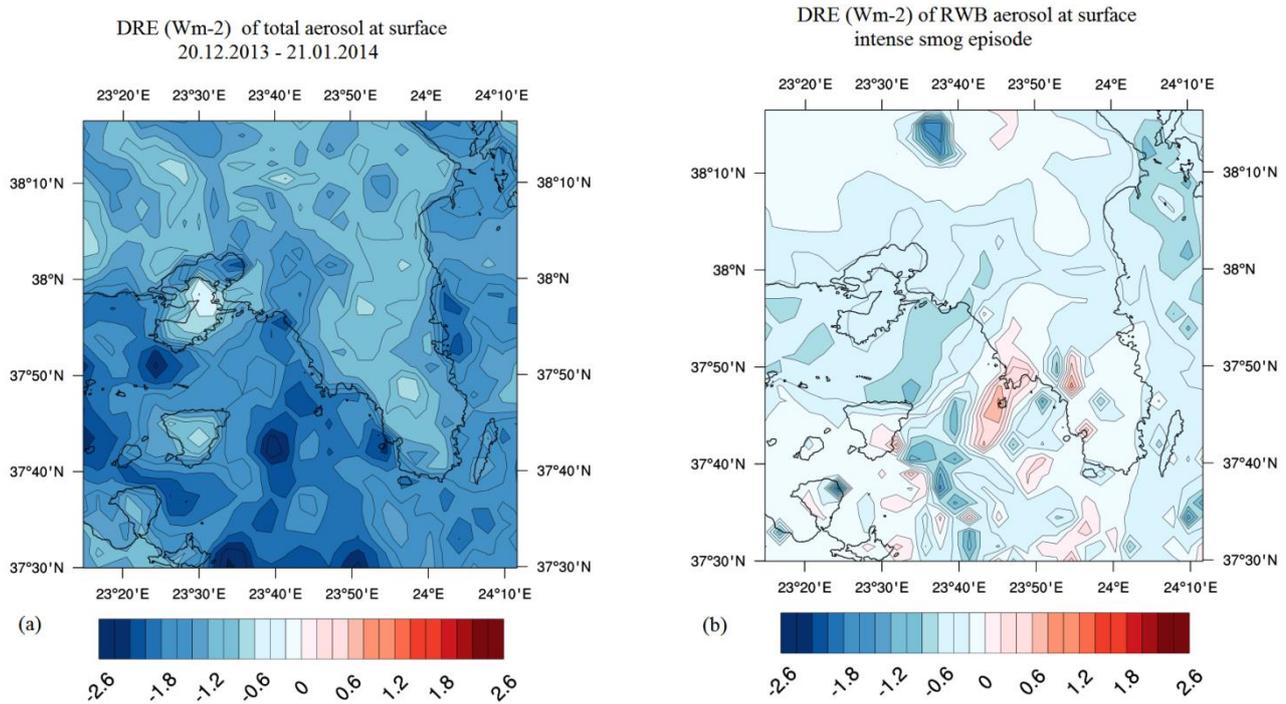


Figure 9: The spatial distribution of the direct radiative effect (Wm^{-2}) of the aerosol load at the ground level, over the extended area of Athens, during: (a) a winter month (20 December, 2013 – 21 January, 2014), taking into account the total aerosol load (mean monthly difference between case 2 and case 4) and (b) the RWB smog episode (4 – 5 January, 2014), isolating the RWB aerosol load (mean daily difference between case 2 and case 3).