Response to Anonymous Referee #2

We thank reviewer #2 for valuable comments which helped improve the manuscript. All the reviewer's points have been carefully considered, and our manuscript has been modified accordingly.

Below follow answers to the general and specific comments.

General comments:

This is an interesting model study about surface ozone levels as well as emissions and concentrations of air pollutants from wild fires in the Mediterranean region. The paper is well presented and most of the conclusions seem sound. The following general and specific comments should be considered before publication in ACP.

The paper includes substantial discussion on the evaluation and results of model simulations of fire emissions. This would merit a reflection in the title of the paper.

Answer:
We agree and have changed the title to "Impact of forest fires, biogenic emissions and high temperatures on the elevated Eastern Mediterranean ozone levels during the hot summer of 2007".

The authors suggest that the CO/NOx ratios in the fire emission data need to be revised. However, this is mainly based on the results from the WRF-Chem model which also seems to put most of the emissions close to the surface which means that the concentration contribution compared with the satellite data is minor due to the averaging kernels. Could it not be that underestimation of vertical mixing could explain the discrepancey between observed and simulated CO columns? The EMEP model is not used in this evaluation with the argument that CO concentrations in the upper troposphere are not realistic. What would be the result if a composite of EMEP results for the lower 2-3 km and WRF-Chem for the rest of the atmosphere were compared to the satellite columns? Would the same conclusions hold?

Answer:
A comparison of the plots where averaging kernels have been applied (Fig. 6) with the plots where averaging kernels have not been applied (Fig. A1 in the Supplementary Material) shows that the CO columns in the forest fire plumes are only slightly different, except the WRF-Chem simulation with GFED emissions which shows slightly higher values in the raw model data (w/o AK applied). Further, Fig. A2 in the Supplementary Material illustrates the effect of applying averaging kernels to the model data. If there were underestimation of vertical mixing in WRF-Chem, the raw model data plot (Fig. A1) should have given less discrepancy between observed and simulated CO columns in the forest fire plumes (as is the case for the background CO levels).

As suggested, a comparison of CO between the WRF-Chem and EMEP models is shown below (Fig. S1) as a vertical section for the lower 5 km. The figure shows that elevated CO levels due to the Peloponnese forest fire plume can be seen even above ~3 km height in both models. Also, there is no indication of less vertical mixing of CO fire emissions in WRF-Chem despite the fact that most of the emissions are implemented in the lowermost layer of this model. Based on this analysis, we think that our conclusions hold and that the CO/NOx ratios in the fire emission data need to be re-assessed.

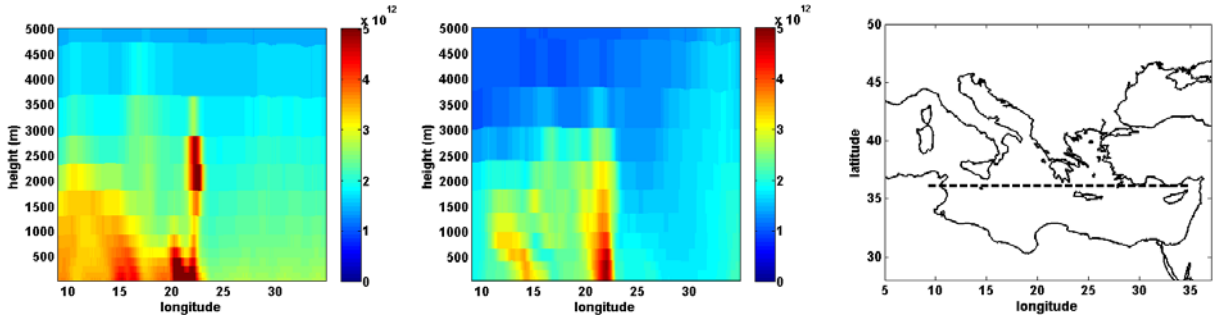

Figure S1. Vertical cross-section of CO concentrations (molec cm-3) modelled with WRF-Chem (left) and EMEP (middle) on 26 August 2007 (both models using GFEDv2 emissions). The location of the vertical section is shown by the dashed line in the right hand plot.

The difference in simulation surface ozone between the models merits some more discussion. The authors rule out differences in emissions. Could the higher surface ozone concentrations in WRF-Chem be due to less vertical mixing? Can inspection of vertical profiles of ozone or intercomparison of boundary layer heights or similar give some clues? This should be discussed.

Answer:
We agree to this point and have added a discussion in Section 3.3 in addition to a comparison of vertical cross-sections of ozone from the two models (Supplementary Material, Fig. A7).

Dry deposition is a very important loss process for ozone. It is unclear from the paper what is meant by the temperature effect on dry deposition. Do any of the models directly account for soil water availability in their calculation of stomata conductance? What is the difference between this effect and the temperature effect? This should be explained better.

Answer:
An explanation of the temperature dependence of dry deposition in the Wesely scheme of WRF-Chem (which is the model that was used for the dry deposition experiments) is now given in Section 2.1. A more precise formulation of what we mean with the temperature effect on dry deposition has also been added to the abstract and the introduction.

Specific comments:

p7618 l9 Suggest the wording "climate change impact research" instead
p7623 l10 Which meteorological variables were nudged?
p7623 l12 Was Oslo CTM2 driven by the same meteorological data as WRF-Chem? Please help the reader although this information is available in the given reference.
p7624 l13 Consider mentioning the chemical boundary conditions in the text for the EMEP model although it is given in table 1. Inconsistent to only mention boundary conditions for one of the models in the text.
p7637 l13 "very similar" is a too strong statement here since the results from WRFChem are clearly higher.
p7640 l13 Did Im and Kanakidou use the same isoprene emissions in their study? Please help the reader.
p7644 l16 I suggest writing "model results using WRF-Chem." Since the EMEP model was not fully compared to the CO measurements.

Answer: We agree with all the specific comments listed above and have changed the manuscript accordingly.

p7619 l25 What is meant by temperature dependency of dry deposition? Please specify. This is also needed in the abstract.
Answer: An explanation is now given in Section 2.1, and more precisely formulated in the abstract and introduction (see also answer to general comment).

p7667 Fig 7 Why are results shown only for part of the domain for the EMEP model? According to fig 1 the EMEP domain covers the whole area displayed in the figure.
Answer: The comparison with satellite observations required hourly model output in 3 dimensions. To avoid extremely large output files, only results from part of the EMEP domain was stored. This limited domain covers the region of most interest and should be sufficient for the discussion of the satellite comparison.

Response to Anonymous Referee #4

We thank reviewer #4 for valuable comments which helped improve the manuscript. All the reviewer's points have been carefully considered, and our manuscript has been modified accordingly.

Below follow answers to the general and specific comments.

General comments:

This is a very interesting paper focusing on a hot summer in the Eastern Mediterranean and investigating the impact of regional emissions from various sources including those from forest fires in Greece and other locations surrounding the Eastern Mediterranean basin on ozone levels using two different mesoscale models.

It represents a significant amount of modeling and analysis work. The conclusion drawn on the need for re-assessment of the CO/NOx ratios from forest fire emissions is a major contribution of this work. However, several clarifications are needed to make this paper appropriate for publication in Atmospheric Chemistry and Physics.

My major concern is about the potential double-counting of emissions from vegetation: To my understanding MEGAN (WRF/CHEM) and EMEP biogenic emissions are calculated based on land-use/vegetation maps that do not account (or do they?) for the forested areas burnt in 2007. Then the biomass burning emissions are added at the top of the biogenic VOC emissions? The authors need to clarify this aspect.

Answer:
The model's land-cover maps have no knowledge of the effects of the fires, and so in principle some land-area may be emitting BVOC at the same time as the area is supposedly burning. However, we do not feel that this is a large problem compared to the uncertainties in forest-fire and BVOC emissions in general. A short discussion has been added to the description of biogenic emissions in Section 2.3.

Clarifications are also needed on the consistency between the two applied parameterizations in the biogenic emissions (MEGAN) and the dry deposition scheme. If increasing temperature causes the plants stomata to close and thus reduces deposition, would it be that it also affects BVOC emissions? Are these two effects consistently taken into account or are they treated totally decoupled?

Answer:
These effects are taken into account in the models. A short discussion has been added at the end of Section 2.4.

Another point of concern is the accuracy of the plume chemistry as simulated by the mesoscale models. Due to high non linear behavior of O3 (and OH) to NOx levels, doubt that models with resolution of a few tens of km are able to capture the low oxidant levels in the center of a fresh plume. How such model limitation can affect the presented results and conclusions?

Answer:
A short discussion of plume chemistry (including references) has been added to the manuscript (Section 3.3.1). We agree that a model resolution of 25 km x 25 km is too coarse to resolve the small-scale chemistry occurring within the plume, but based on results from previous literature (e.g. Tie et al., 2010; Hodnebrog et al., 2011) we do not think that a finer grid resolution would yield substantial changes to our results. The issues surrounding grid-size are complex; as pointed out by Pleim and Ching (1993), decreasing grid size does not necessarily give more realistic ozone production rates. Furthermore, as forest fire plumes are area sources, the inaccuracies that arise due to coarse resolution are likely to be less than for point sources such as power plants.

Specific comments:

Page 7619, lines 10-15: this part fits better before the discussion of the CO levels.
Page 7619, line 25: add a reference
Page 7620, line 11: indicate that Poupkou et al. 2009 concerns summertime.
Page 7622, section 2: the coordinates of the 4 corners of the two model domains and the Eastern Mediterranean area can be provided here or in the Figure 1 caption.
Page 7622, line 21: the full model name (WRF-CHEM) should be given where it has been first mentioned.
Page 7623, line 11: the configuration of the Oslo CTM2 model should also be briefly described in the supplementary material as in Table 1 for the mesoscale models.
Page 7624, line 28: provide a reference for lumping to RADM2 species.
Page 7625: lines 1-4: How the NMVOC are speciated in the EMEP model?
Page 7627, line 12: : : : are ...
Page 7628: line 14: rephrase
Page 7628, line 23: are given
Page 7629, end of 1st paragraph: explain better the impact of emissions from fires on ozone production.
Page 7629, lines 16-17: ozone SURFACE measurements... in the EXTENDED region of interest... (Most of these stations are located upwind the fire events areas. This has to be mentioned).
Page 7629, section 3.2. Reference to Table 4 (in which geographic coordinates of the stations are missing) is needed here.
Page 7630, line 24: EMEP has also a relatively 'low' top at 100 hPa as given in Table 1.
Page 7630, line 25: WAS to be made...
Page 7631, last line: contributes
Page 7632: line 2: dilution in the model grid
Page 7634, line 19: reactivity of NOx to NOy: please rephrase.
Page 7635: lines 9-11: To my understanding Eastern Mediterranean stations are those in countries surrounding the East Mediterranean Basin, i.e. located roughly south of 45N. Thus some of the stations used for the model evaluation are in East Europe and not South East Europe. Please rephrase.
Page 7636, line 10: The strength of the emissions is also very different between the two models.

Page 7636, lines 14-20: besides the uncertainties in the emissions, overestimations at coastal sites such as Finokalia can be due to underestimated dry deposition due to the grid resolution that provides more water fraction in the particular grids cell, leading to accumulated O3 levels.
Page 7637, line 22: indicate which sensitivity study.
Page 7638, line 3: two orders of magnitude
Page 7639, line 21: GFED emissions are spread EQUALLY (?) over the 16 grid cells?
Page 7639, line 26: impact on ozone
Page 7641, line 9: These are areas that are experiencing very high temperatures in the Eastern Mediterranean.
Page 7642, line 16: This agrees with...
Page 7642, line 18-19: ' export of ozone': a fraction of ozone is exported but significant amounts should be formed by exported precursors.
Page 7643, line 24- 7644, line 1. When boundary conditions are kept the same and regional emissions are changing, the importance of long range transport of pollutants (e.g. O3/NOx/PAN/HNO3/CO) for the regional chemistry is also changing. Some rephrasing is needed to make this clear.
Page 7644, lines 4-7: As shown in Table 2 the largest source of VOC in the region during summer is vegetation. Therefore, ship emissions downwind of biogenic VOC emissions can also produce ozone.
Page 7645, line 5: the largest STUDIED contributor.
Page 7645, line 14: over land.
Page 7645, line 18: in the core of the plume : do not forget that model resolution is 25 km.
Page 7645, line 25: The point here is that all chemistry is faster both the O3 production and the O3 destruction reactions.
Table 2: Please add the surface of the studied region in the caption.
Table 5: For EMEP the 'percentile' titles have to be shifted to the right
Figure 6- caption: The data are... The model data have...
Figure 13 –caption: due to

Answer: We agree with all the specific comments listed above and have changed the manuscript accordingly.

Page 7620, lines 23-25 there is some repetition with page 7619 lines 20-25.
Answer: We prefer to keep the original sentence to emphasize that the climate of the Mediterranean region favours ozone formation.

Page 7621, line 19-22: Can you be more specific on the share between increasing BVOC emissions and decreasing dry deposition in the ozone increase in these earlier works? and compare your results to these earlier studies in the discussion session.
Answer: A more quantitative comparison with these earlier studies proves difficult due to the very different domains that have been used. Vieno et al. (2010) studied the hot 2003 summer in UK, while Solberg et al. (2008) included all of Europe in their domain. Direct comparison between these papers and our calculations over the Eastern Mediterranean in 2007 is therefore not sensible.

Page 7622, line 19: Table 1 should be updated to provide the first model layer heights and if possible, PBL layer heights (or how many layers are within the PBL roughly). The model calculation of temperatures, biogenic emissions and deposition can vary largely based on the first layer heights.
Answer: The full height of the lowermost level of each model has now been added to Table 1. The PBL layer heights vary largely in both time and space and therefore it does not make sense to present this variable as one single number.

Page 7622, section 2.1: since WRF-CHEM can simulate chemistry feedbacks on meteorology and considering that the periods that are studied in this paper concern forest fire, the aerosol scheme should also be mentioned (if used).
Answer: Due to computational constraints, no aerosol scheme has been used in WRF-Chem.

Page 7626: lines 17-20: Information on NMVOC speciated fire emissions as derived from this study would be useful. It can be provided in the supplement.
The dataset with the NMVOC speciated fire emissions have not been derived from this study. The source of these data has now been properly referenced (A. Heil, pers. comm.).

Page 7627, line 8: impact of total cloud fraction on PAR: Is this taken into account in both MEGAN and EMEP BVOC emissions calculations? If not, how much of the difference in the computed emissions can be attributed to that?
Answer: This is taken into account in both WRF/MEGAN and EMEP BVOC emissions calculation. The manuscript has been modified to make this point clear.

Page 7628, section 3.1. comparisons between modeled and observed surface temperatures in some representative stations can be presented (in supplement) in order to show how good the model was able to simulate the transport.
Answer: We consider this to be beyond the scope of this study.

Page 7631, lines 2-4: As written it is not clear if the difference in the retrieved CO columns is real or a measuring artifact. This sentence needs rephrasing for clarity.
Answer: A more detailed explanation of how the retrievals are affected by different thermal contrasts is now given.

Page 7631, line 8: fires in Algeria are not seen in Figure 2
Answer: The reason is that the fire emissions in Figure 2 are shown only for the region of the WRF-Chem domain, while the Algerian fires fall outside this region. Domain borders have been added to Figure 2 to indicate which regions are shown in each plot.

Page 7633, line 17: Could this discrepancy be due to the strength of the anthropogenic sources in Ukraine? Does this difference persist when the fire emissions impact is separated from the other sources?
Answer: Thank you for pointing this out. The results with the fire emissions impact separated from the other sources show that the high NO2 concentrations over Ukraine persist. Inspection of results with the anthropogenic emissions separated confirms that the discrepancy is due to the anthropogenic sources. The manuscript has been updated with this new information.

Page 7634, figure 7: A scatter plot comparison between model and satellite observations would be more informative.
Answer: We agree that some of the discrepancies between models and observations may be better explained with a scatter plot, but we concentrate here on the location and influence of the fire plumes and therefore find a contour plot of the horizontal distribution more useful for the evaluation.

Page 7639, line 16: Table 2 shows that NMVOC emissions in FINN are lower than in GFED, whereas those of CO and NOx are the other way around. Is there a reason for that?
Answer: The NMVOC emissions in FINN are slightly higher than in GFED, while the CO and NOx emissions are lower. Differences between FINN and GFED can vary regionally due to the various assumptions involved in the calculation of emissions (e.g. emission factors, fuel inputs, burned area). A short discussion of the differences between FINN and GFED has now been included in Section 2.3.

Page 7639, lines 23-24: Rephrase to discuss NO titration of O3, HNO3 formation, NMVOC oxidation.
Answer: Please see answer to general comment regarding plume chemistry.

Page 7639 last paragraph: This is interesting – Is there a difference in pollutant lifetimes between the two models driven by chemistry that is making this difference?
Answer: Previous studies show that the two chemical mechanisms predict similar O3 concentrations, but with higher O3 levels predicted by EMEP in NOx rich environments (Gross and Stockwell, 2003; Jimenez et al., 2003). Hence, differences related to plume rise or meteorology in the two models could be more probable explanations. The manuscript text has been updated with this information.

Page 7640, line 9: It is difficult to compare the BVOC emissions impact on O3 calculated by the two models, since the BVOC emissions are very different. A kind of normalization of impact on O3 to the BVOC emissions might be useful.
Answer: We agree that a normalization of O3 impact due to biogenic emissions would be interesting. However, the spatial distribution of both BVOC emissions and ozone production also varies, in different and complex ways, and there is no easy way to normalise this. It would require considerable work and new model simulations to investigate this, beyond the scope of this study.

Finally there are many references to "not shown" material in the results that I consider are important and would be useful to be presented in a supplementary material.
Answer: All "not shown" figures are now presented in a supplementary material.

References

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