Interactive comment on "Impact of wildfires on particulate matter in the Euro-Mediterranean in 2007: sensitivity to the parameterization of emissions in air quality models" by Marwa Majdi et al.

Anonymous Referee #3

The authors wish to thank the anonymous referee for the very helpful comments and corrections. All corrections have been included in this new version. A response to the general and specific comments is provided below (in blue).

#General comments :

This paper is about the modeling of wildfires in air quality models and the associated uncertainties on the aerosols concentrations and optical properties. The article is based on several numerical simulations of the 2007 fires in the Mediterranean region with the Polyphemus and CHIMERE models. Simulations explore the sensitivity of these models to chemical related factors (mainly VOC emissions) and dynamics (injection height) . The scientific approach is sound. The work presented is substantial (several simulations have been done and analyzed) but the conclusions and discussions deserve more work before publication. Several conclusions written in the manuscript are expected and already known. The risk is that the impact of the publication to a broader scientific community remains limited unless the authors put the conclusions into a wider perspective. In particular, this would require to add a section dedicated to a scientific discussion including more references to previous works on the subject if possible.

A scientific discussion including previous work highlighting the impact of fire on the injection height is already mentioned in section 4 page 19 line 24 to page 20 line 7.
To highlight the importance and meaning of the sensitivity studies, the sentences line 29-32 page 24 : "The sensitivity to I/S-VOCs emissions and injection height is important as it shows the maximum impact of I/S-VOCs emissions and injection height on PM2.5 concentrations and AOD. The sensitivity is high (40 to 50%), but it may not explain order of magnitude differences that sometimes occur between models and observations." are replaced by: "Sensitivities to these key parameters are computed using simulations performed with different configurations of Polyphemus. These configurations are chosen to maximize the sensitivities. AOD is particularly sensitive to I/S-VOCs emissions (up to 40% sensitivity, while surface PM2.5 concentrations are particularly sensitive to injection heights (up to 50% sensitivity). These sensitivities are most of the time higher than inter-model sensitivities, which are mostly linked to the model vertical discretization close to fire emissions."

To broaden the conclusion and address the limits of our study following comments from another reviewer, paragraphs on other existing sources of uncertainties are added in the conclusion:

(1-The influence of NH3 emissions and the formation of inorganics is added)
Over Europe, inorganics (mainly sulfate, sea salt and ammonium) contribute highly to PM2.5 composition, when fire emissions are not considered (Fountoukis et al., 2011). However, during fire events the contribution of inorganics is lower than the contribution of

organics (8 to 9% from inorganics against 40 to 80% from organics). Focusing on this inorganic contribution from fires, sulfate, ammonium and nitrate are the predominant inorganic components. The formation of inorganics because of wildfires is found to be low compared to the formation of organics. However, our simulation does not take into account emissions of inorganic precursors such as ammoniac (NH3).

Several studies (R'Honi et al.,2013; Van Damme et al.,2014; Whitburn et al., 2017), show that large emissions of NH3 are released by biomass burning. Whitburn et al. (2017) studied the enhancement ratios NH3/CO for biomass burning emissions in the tropics using observations from the IASI satellite based instrument. They found a significant variability due to fire contribution. According to the Whitburn et al. (2017), the emission ratios NH3/CO in the tropics derived from IASI observations (as in Van Damme et al., 2014) are rather on the lower end of those reported in Akagi et al. (2011) that are used here.

If fire emissions are important for the regional budget of organics, more observations are required to provide emissions values of NH3 and concentrations of inorganics should be evaluated close to fire regions.

(2-Discussions on the lack of detailed observational data are also added)

Moreover, the ability of Polyphemus/Polair3D and CHIMERE to simulate regional surface PM2.5 concentrations and optical properties (particularly AOD) was evaluated based on comparison to available measurements (8 AIRBASE stations and 6 AERONET stations). Only two out of the 8 AIRBASE stations (GR0039A and GR0035A in Greece) and 3 out of 6 AERONET stations (Lecce University in Italy, Blida in Algeria and Bucharest in Romania) are used because they are they are the most affected by wildfires (background suburban and rural stations where fire contribution is higher than 10%). The lack of surface observations strongly limits this evaluation but it is partly complemented by comparisons to MODIS satellite-based observations of AOD.

Comparisons to surface and remote sensing observations show that the models can simulate enhancements of a good order of magnitude and +/- 1 day uncertainty in the timing. However, more surface observations in remote regions is necessary for a precise evaluation of the simulated long-range transport from fire emissions, the aerosol speciation within the plumes and the resulting impact on air quality.

(3-Details were added in the conclusion concerning the emissions of PPMfine and I/S/L-VOCs.)

In this study, PPMfine emissions correspond to emissions factors of PM2.5 minus primary aerosols emissions of OM and BC. This model species is used to fill a gap in current evaluation of emissions and is therefore very uncertain.

This study shows that considering PPMfine emissions and I/S/L-VOC emissions may be redundant and may correspond t the same “missing emissions mass”.

Several models actually partly treat this missing part by considering I/S/L-VOCs emissions from particulate matter emissions from biomass burning emissions (Koo et al., 2014; Konovalov et al., 2015) . For example, several studies estimated the I/S/L-VOCs emissions by multiplying he primary organic aerosols (POA) by a factor of 1.5 following the chamber measurements (Robinson et al., 2007; Zhu et al., 2016; Kim et al., 2016). Some studies/models do not consider specific species/surrogates to treat these missing emissions, but simply use a ratio to reduce uncertainties related to the estimation of PM emissions. For example, Kaiser et al. (2011) use a factor of 3.4 for PM emissions based on the comparison between simulations and AOD observations.

(4-Uncertainties on the burned area)

Uncertainty on the burned area and the associated temporal evolution (used as input to the calculation of emissions) is also high. Giglio et al. (2010) found that uncertainties on MODIS observation can reach about 5 days mainly due to cloud cover.

(5- Uncertainties due to deposition)
Deposition can be considered as a source of uncertainty on PM and AOD over the Euro-Mediterranean region during summer 2007. Roustan et al. (2010) pointed out the importance of dry and wet deposition over Europe while studying the sensitivity of Polyphemus to input data over the Europe with a focus on aerosols. They found that PM is sensitive to options influencing deposition such as wet diameter and aerosol density. They found that during both summer and winter, the uncertainties on wet diameter and aerosol density can reach 19% and 9% respectively. Uncertainties on these parameters can lead to uncertainties on the dry/wet deposition fluxes and thus on AOD and PM2.5 concentrations.

#<u>Specific comments</u> :

1) The added value of the CHIMERE model to the analysis must be better explained. The main conclusion I hold is the impact of the vertical resolution. The vertical resolution is certainly a factor of uncertainty for the representation of the boundary layer in general (with or without fire) and would deserve a full sensitivity analysis. At a minimum, this limitation of both models should be discussed.

A) The added value of the CHIMERE model to the analysis must be better explained:

These sentences are added in page 4, line 15 – 18 : "CHIMERE is added to perform a model-to-model comparison and to evaluate the capability of other current CTMs to simulate the impact of wildfires on the regional particulate matter budget and to quantify the uncertainties on air quality modeling related to the integration of fire emissions in CTMs."

B) The main conclusion I hold is the impact of the vertical resolution. The vertical resolution is certainly a factor of uncertainty for the representation of the boundary layer in general (with or without fire) and would deserve a full sensitivity analysis. At a minimum, this limitation of both models should be discussed:

The sentence where we pointed out the impact of the vertical resolution in the inter-model sensitivity (section 4 page 19 lines 14-16) is modified to stress the limitation of both models: "Furthermore, this region of high inter-model sensitivity corresponds to the region where the sensitivity to the injection height is the highest for PM2.5 concentrations. It may therefore be linked to differences in the models' vertical discretization. The models use different vertical coordinates and different numbers of vertical levels. The vertical resolution of the models is rather low as Polyphemus uses 14 vertical levels and CHIMERE uses 19 vertical levels."

2) The introduction refers to the study of PM2.5 and PM10. The latest are little discussed.

In this work, we focus only on PM2.5 since PM10 concentrations in the Euro-Mediterranean area are strongly affected by dust, which are difficult to simulate due to their sporadic nature and their main sources are located out of the model domain.
The sentence in the introduction page 3 lines 23-24 "The objective of this study is to evaluate the capabilities of current CTMs to simulate the impact of wildfires on the regional particulate matter budget (PM2.5 and PM10)." is replaced by : " The objective of this study is to evaluate the capabilities of current CTMs to simulate the impact of wildfires on the regional particulate matter

budget. In the Mediterranean region, surface PM10 is dominated by the contribution from dust (Rea et al., 2015). Since the focus of this study is on biomass burning, the discussion is centered on the simulation of surface PM2.5. The total loading of aerosols over the region is evaluated using comparisons of AOD to observations."

3) The fact that only one ground station was available to validate the model near fires moderates confidence in the findings. The paucity of the in-situ data (which is not the fault of the authors) should be pointed out in the general conclusion.

Sentences in the conclusion page 24-lines 4-7: "The ability of Polyphemus/Polair3D and CHIMERE to simulate regional PM2.5 concentrations and AOD was evaluated based on comparison to available measurements. The general evaluation compared to background AIRBASE and AERONET surface observations shows good performances (with high correlation coefficients) for surface PM2.5 concentrations and AOD." are replaced by:
"The ability of Polyphemus/Polair3D and CHIMERE to simulate regional surface PM2.5 concentrations and optical properties (particularly AOD) was evaluated based on comparison to available measurements (8 AIRBASE stations and 6 AERONET stations). Only two out of the 8 AIRBASE stations (GR0039A and GR0035A in Greece) and 3 out of 6 AERONET stations (Lecce University in Italy, Blida in Algeria and Bucharest in Romania) are used because they are they are the most affected by wildfires (background suburban and rural stations where fire contribution is higher than 10%). The lack of surface observations strongly limits this evaluation but it is partly complemented by comparisons to MODIS satellite-based observations of AOD.
Comparisons to surface and remote sensing observations show that the models can simulate enhancements of a good order of magnitude and +/- 1 day uncertainty in the timing. However, more surface observations in remote regions is necessary for a precise evaluation of the simulated long-range transport from fire emissions, the aerosol speciation within the plumes and the resulting impact on air quality."

4) Both models include wet and dry deposition but little information is provided on the approaches. Are dry and wet depositions sources of uncertainties for PM2.5 and AOD? Wet deposition might not be predominant during the studied period.

4.a) Both models include wet and dry deposition but little information is provided on the approaches.
The sentence in page 5 line 14: "Both models include wet and dry deposition" is replaced by : "Both models include wet and dry deposition. Deposition in Polyphemus is described in Sartelet et al. (2007) and in CHIMERE in Menut et al. (2013) and Mailler et al. (2017) ".

For particles, dry deposition is parameterized with a resistance approach following Zhang et al. (2001). In Polyphemus, the in-cloud and below-cloud scavenging is parameterized following Roselle and Binkowski (1999).

4.b) Are dry and wet depositions sources of uncertainties for PM2.5 and AOD? Wet deposition might not be predominant during the studied period.

In this study, we didn't investigate the uncertainties due to dry/wet deposition over the Euro-Mediterranean region. However, Roustan et al. (2010) pointed out the importance of dry and wet deposition over Europe and studied the sensitivity of air quality model (Polyphemus) to input data over the Europe with a focus on aerosols. They found that PM is sensitive to options influencing deposition such as wet diameter and aerosol density. They found during both summer and winter, the uncertainties on wet diameter and aerosol density can reach 19% and 9% respectively. Uncertainties on these parameters can lead to uncertainties on the dry/wet deposition fluxes and thus on AOD and PM2.5 concentrations. A discussion on these uncertainties is added in the conclusion.

5) The conclusion on the three months period evaluation says that the simulations are improved when fire emissions are included: is this surprising? It seems to me that the conclusion was expected unless there are previous studies that have concluded otherwise?

Indeed, it is not surprising that adding fire emissions, the simulations are improved. But, the main goal of this section is to evaluate statistically the performance of two CTMs to simulate regional PM2.5 concentrations and optical properties when fire emissions are considered, based on comparisons to available measurements. The main conclusion is that models are in good agreement with observations only when considering fire emissions. This shows the importance of considering these emissions during fire events but also gives confidence in the emissions included. Also, it compares the uncertainties linked to modeling the injection height, I/S/L-VOC emissions, and inter-model uncertainty to determine whether it is relevant to keep on improving models of injection height and I/S/L-VOC emissions. Conclusions to this point were strengthen in the current version.

6) Surface PM2.5 are particularly sensitive to the injection heights" is also an expected conclusion. Can the study help to decide between a PBL mixing of fire emissions vs. injection height above PBL?

In Table 3 and 4, the model shows almost similar statistics and performance using both configurations,in simulations Poly-ref (injection between 1 km and PBL) and Poly-3km (injection between 1 and 3 km). Therefore this study can not really help to decide between a PBL mixing of fire emissions .vs. injection above PBL. Injection heights should actually vary as a function of meteorological conditions and fire intensity using a pyroconvection scheme. Here we tested two extreme situations. We have already specified that in section 2.3 line 5: "This choice of sensitivity study may be viewed as conservative since, for example, injection heights are limited to 3 km. But it is also extreme since maximum injection at 3 km is imposed to all fires, resulting in injection above the boundary layer. This could be realistic, since based on the Multi-angle Imaging SpectroRadiometer (MISR) observations, however Mims et al. (2010) estimated that 26% of the fire plumes exceed the boundary layer."

7) The introduction of the PPMfine fraction remains a little obscure for me and would deserve a little more explanation. It is important to understand this variable in light of its significant contribution to the composition of PM2.5 and the uncertainties in its definition (expected overestimation). How is this missing part treated in the other models?

PPMfine are all the unidentified fine particles emitted by wildfires which are incorporated to consider the differences between PM2.5 emissions and the total of all PM included in specific species. There are a lot of uncertainties related to the estimation of PPMfine emissions.
Several models treat this missing part by considering I/S/L-VOCs emissions from particulate matter emissions from fire emissions (Koo et al., 2014; Konovalov et al., 2015). For example, several studies estimated the I/S/L-VOCs emissions by multiplying the primary organic aerosols (POA) by a factor of 1.5 following the chamber measurements (Robinson et al., 2007; Zhu et al., 2016; Kim et al., 2016).

Moreover, other studies/models do not consider specific species/ surrogates to treat these missing emissions, they use a ratio to reduce uncertainties related to the estimation of PM emissions. For example, Kaiser et al. (2011) use a factor of 3.4 for PM emissions based on the comparison between simulations and AOD observations.

#Technical corrections:

1) Figure 2: I assume that these are emissions calculated by APIFLAMME ?

The title of Figure 2 in page 8: "Daily total OC emissions during the summer of 2007 in the four sub-regions of Figure1"Is replaced by: " Daily total OC emissions calculated by APIFLAME during the summer of 2007 in the four sub-regions of Figure1".

2) Section 2.1: how are I/S/L VOCs represented in CHIMERE?

In CHIMERE, we do not consider I/S/L-VOCs.

3) Table 2: I am confused between the Table marks and the comment above concerning the configuration of the CHIMERE model. The comment refers to simulation without I/S-VOCs and with fires but the table for the CHIMERE-ref shows a "Yes" for I/S-VOCs

In Table 2 page 9: "yes" for I/S-VOCs from fire for simulation " CHIMERE-ref" is corrected and replaced by "No".

4) Change PB to PBL.

In Table 2 page 9 : "Between 1 km and PB" for Fire emissions' injection height is replaced by " Between 1 km and PBL".

5) Figure 3 & 4: Legend: add "surface PM2.5"

The legend in Figure 3 in page 12 "Daily mean PM2.5 and AOD at 550 nm from the Poly-ref simulation averaged over the summer of 2007 (the 8 AIRBASE and 6 AERONET stations, used in this work, are represented here in blue dots) " is replaced by "Daily mean surface PM2.5 and AOD at 550 nm from the Poly-ref simulation averaged over the summer of 2007 (the 8 AIRBASE and 6 AERONET stations, used in this work, are represented here in blue dots)".
The legend in Figure 4 in page 13 "Left panel: relative difference of PM 2.5 concentrations between simulations Poly-ref and Poly-Nofires during the first fire event. Right panel: relative difference of PM2.5 concentrations between simulations Poly-ref and Poly-Nofires during the second fire event." is replace by: "Left panel: relative difference of surface PM 2.5 concentrations between simulations Poly-ref and Poly-Nofires during the first fire event. Right panel: relative difference of surface PM2.5 concentrations between simulations Poly-ref and Poly-Nofires during the second fire event."