

Impact of wildfires on particulate matter in the Euro-Mediterranean in 2007: sensitivity to ~~the parameterization~~ some parameterizations of emissions in air quality models

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Abstract.

This study examines the uncertainties on air quality modeling associated with the integration of wildfire emissions in chemistry-transport models (CTMs). To do so, aerosol concentrations during the summer 2007, which was marked by severe fire episodes in the Euro-Mediterranean region especially in Balkan (20–31 July 2007, 24–30 August 2007) and Greece (24–30 August 2007), are analysed. Through comparisons to observations from surface networks and satellite remote sensing, we evaluate the abilities of two CTMs, Polyphemus/Polair3D and CHIMERE, to simulate the impact of fires on the regional particulate matter (PM) concentrations and optical properties. During the two main fire events, fire emissions may contribute up to 90% of surface PM_{2.5} concentrations in the fire regions (Balkans and Greece), with a significant regional impact associated with long-range transport. Good general performances of the models and a clear improvement of PM_{2.5} and aerosol optical depth (AOD) are shown when fires are taken into account in the models with high correlation coefficients.

Two sources of uncertainties are specifically analysed in terms of surface PM_{2.5} concentrations and AOD using sensitivity simulations: secondary organic aerosol (SOA) formation from intermediate and semi-volatile organic compounds (I/S-VOCs) and emissions' injection heights. The analysis highlights that surface PM_{2.5} concentrations are highly sensitive to injection heights (with a sensitivity that can be as high as 50% compared to the sensitivity to I/S-VOCs emissions which is lower than 30%). However, AOD which is vertically integrated is less sensitive to the injection heights (mostly below 20%), but highly sensitive to I/S-VOCs emissions (with sensitivity that can be as high as 40%). The maximum statistical dispersion, which quantifies uncertainties related to fire emissions modeling, is up to 75% for PM_{2.5} in Balkan and Greece, and varies between 36 and 45% for AOD above fire regions.

The simulated number of daily exceedance of World Health Organization (WHO) recommendations for PM_{2.5} over the considered region reaches 30 days in regions affected by fires and ~ 10 days in fire plumes, which is slightly underestimated compared to available observations. The maximum statistical dispersion (σ) on this indicator is also large (with σ reaching 15 days), showing the need for better understanding of the transport and evolution of fire plumes in addition to fire emissions.

1 Introduction

The Mediterranean area is directly affected by large aerosol sources leading to an European maximum in aerosol loading (Putaud et al., 2010; Nabat et al., 2013; Rea et al., 2015). Observations show the influence of a complex mixture of different sources (Dall'Osto et al., 2010; Gerasopoulos et al., 2011; Boselli et al., 2012). The pollution transport pathways in the region are controlled by the very specific orography of this closed sea, but also by the influence of the large circulation patterns (Lelieveld et al., 2002; Lionello et al., 2006) due to its location between the subtropical high-pressure systems, the mid-latitude westerlies and low pressure systems. Mineral dust contributes significantly to pollution episodes in the Euro-Mediterranean area, further increasing aerosol loads associated with local anthropogenic sources (Querol et al., 2009; Gobbi et al., 2007; Kaskaoutis et al., 2008; Nabat et al., 2013; Rea et al., 2015). During summer, high concentrations of organic aerosols, mostly of biogenic origin, are observed in the western Mediterranean (El Haddad et al., 2013; Chrit et al., 2017). Although the precursors are biogenic volatile organic compounds, the formation of organic aerosols is partly explained by the influence of anthropogenic emissions (Kanakidou and Tsigaridis, 2000; Carlton et al., 2010; Sartelet et al., 2012).

Vegetation fires are another sizable sporadic source that needs to be accounted for, especially during summer when the hydrological and meteorological conditions favor their occurrence and spread. Depending on the severity of the fire season, their contribution to the atmospheric aerosol loading and thus to the impairment of the local and regional air quality can be significant (Barnaba et al., 2011; Rea et al., 2015). However, quantifying their contribution remains a challenge due to large uncertainty in emissions and transport.

Most fire episodes in Europe occur in southern countries (Portugal, Spain, France, Italy, Greece) with $\sim 500 \cdot 10^3$ ha burned every year (Barbosa et al., 2009; Turquety et al., 2014). On average, only $\sim 2\%$ of fires contribute to $\sim 80\%$ of the area burned due to clusters of fires that merge into "mega-fires" (San-Miguel-Ayanz et al., 2013). Although ignitions are mainly of anthropogenic origin (negligence, arson, agricultural practices) [according to San-Miguel-Ayanz et al. \(2013\) and the European Forest Fire Information System \(EFFIS\) \(JRC, 2008\) of the European Joint Research Center \(JRC\)](#), fire spread depends on meteorological conditions. It is favored by hot and dry conditions (heat waves and associated droughts), especially if they are preceded by wet winter and spring (fuel accumulation) (Pereira et al., 2005; Hernandez et al., 2015). Fires are also important in eastern Europe, Ukraine, western Russia and Turkey. They are usually smaller and associated with agricultural practices like waste burning and land clearing (Korontzi et al., 2006; Stohl et al., 2007; Turquety et al., 2014).

Turquety et al. (2014) estimate that, on average for the 2003-2010 time period and the Euro-Mediterranean region, total yearly fire emissions amount to $\sim 30\%$ of anthropogenic emissions for $\text{PM}_{2.5}$ (particulate matter with diameter $\leq 2.5 \mu\text{m}$). It is all the more critical as the fire episodes are concentrated during the summer and usually last less than 10 days (so that emissions are very concentrated in time resulting in dense plumes). However, the uncertainty associated with fire emissions is also very large, estimated to ~ 100 to 200% (e.g. Urbanski et al. 2011; Turquety et al. 2014). Uncertainties are also linked to the modeling of the temporal variability of emissions. Improving the diurnal cycle may for instance be critical for some fire events (Rea et al., 2016). In addition to emissions, the modeling of wildfires' impact on atmospheric chemistry using chemistry-transport models (CTMs) requires a good knowledge of the emissions' injection height. Indeed, the energy released by fires

can trigger or enhance convection (pyroconvection) and there by injecting emissions at high altitude. Several parameterizations have been developed in recent years (Freitas et al., 2007; Rio et al., 2010; Sofiev et al., 2012) and are increasingly implemented in CTMs. However, comparisons to observations of fire plume's height highlight the difficulty of correctly capturing the vertical shape of fire emissions (Sofiev et al., 2012; Val Martin et al., 2012; Rémy et al., 2016). This will then influence the simulated transport pathways and its regional impact. Injecting above the boundary layer will lower the local impact but result in larger-scale transport. The chemical evolution of fire plumes is still not well understood and not well represented. Primary organic aerosols (POA) are directly emitted by biomass burning into the atmosphere. However, secondary organic aerosols (SOA) are produced through gas-to-particle of oxidation products of volatile organic compounds (VOCs) (with saturation concentration C^* higher than $10^6 \mu\text{g.m}^{-3}$), intermediate organic compounds (I-VOCs) (with saturation concentration C^* in the range of 10^4 – $10^6 \mu\text{g.m}^{-3}$) and semi-volatile organic compounds (S-VOCs) (with saturation concentration C^* in the range 0.1 – $10^4 \mu\text{g.m}^{-3}$), Low volatility organic compounds (L-VOCs) (with saturation concentration C^* lower than $0.1 \mu\text{g.m}^{-3}$) (Lipsky and Robinson, 2006; Robinson et al., 2007; Grieshop et al., 2009; Huffman et al., 2009; Cappa and Jimenez, 2010; Fountoukis et al., 2014; Tsimpidi et al., 2010; Woody et al., 2016; Giancarlo et al., 2017). The formation of secondary organic aerosols (SOA) from the ageing of biomass burning organic precursors is likely to strongly affect aerosol loading and properties in biomass burning plumes. The major organic precursors are thought to be intermediate, semi and low volatility organic compounds (I/S/L-VOCs), (May et al., 2013a; Koo et al., 2014; Konovalov et al., 2015; Giancarlo et al., 2017). However, I/S/L-VOC emissions are not well characterised and their gas-phase emission is often missing from emission inventories (Robinson et al., 2007). Their emissions are often estimated from particulate matter emissions (Couvidat et al., 2012; Giancarlo et al., 2017). The chamber experiments of May et al. (2013a) characterised the volatility distribution of I/S/L-VOCs emissions into different volatility classes. Recent studies have proven that considering I/S-VOCs emitted from biomass burning, shows a major improvement of the agreement between the simulated and observed organic aerosol (OA) (Koo et al., 2014; Konovalov et al., 2015; Giancarlo et al., 2017). Konovalov et al. (2015) find that ignoring I/S-VOCs from biomass burning underestimates strongly the ratio of $\Delta PM_{10} / \Delta CO$ (by a factor of 2) in the city of Kuopio (Finland) and thus leads to an underestimation of the OA concentrations.

The objective of this study is to evaluate the capabilities of ~~current two~~ CTMs to simulate the impact of wildfires on the regional particulate matter budget (~~PM_{2.5}~~, 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 PM_{2.5}. The total loading of aerosols over the region is evaluated using comparisons of AOD to observations. After an evaluation of two CTMs (CHIMERE and Polyphemus/Polair3D) through comparisons to observations from surface networks and remote sensing, a sensitivity analysis to the injection heights and I/S-VOCs emissions is conducted in order to quantify the uncertainties associated to these two parameterizations, in terms of AOD and surface PM concentrations. This analysis is focused on the summer 2007 which was marked by extreme meteorological conditions (consecutive heat waves and drought) and severe fire episodes in the eastern Mediterranean and Europe. According to the European Forest Fire Information System (EFFIS), 2007 was well above the average of the previous last 3 decades in terms of burned area (574 361 against 495 471 ha burned respectively) (JRC, 2008). The burning season was particularly severe for Greece where the burned area reached ex-

treme values. This case study is particularly interesting since it was well captured by satellite sensors as the resulting smoke plumes, fanned by north-easterly winds, were transported over the sea crossing the south Ionian Sea and reaching the northern part of the African continent. Several studies have highlighted the important enhancements in atmospheric gases (Turquety et al., 2009; Coheur et al., 2009; Hodnebrog et al., 2012) and aerosols (Liu et al., 2009; Kaskaoutis et al., 2011) due to these
5 fire episodes. Modeling analyses have investigated secondary production in the fire plumes. While Hodnebrog et al. (2012) showed limited ozone impact on average during the summer, Poukpou et al. (2014) investigated more precisely the fire event in Peloponnese (Greece) at the end of August 2007. They found enhancements of CO and NO_x concentrations mainly over the burnt areas due to the biomass burning. Due to the non-linear dependence of O₃ on NO_x levels, the near surface O₃ values were reduced (-34%) over the Peloponnese, but increased (+52%) over the sea at 500 km downwind. Here, the sensitivity of
10 regional aerosol loadings, both primary and secondary, to modeling configurations are analyzed.

This paper is structured as follows. In the second section, a brief description of the chemistry transport models and the different tools and methodology used in this work, are given. Then, section 3 presents statistics for model-to-data comparison to assess the models performance during the summer 2007 and more specifically during the main fire events. Then, uncertainties
15 related to the integration of wildfires in CTMs (injection height, I/S-VOCs emissions) are discussed. Finally, section 4 describes the contribution of wildfires to air quality threshold exceedences as well as the associated uncertainties.

2 Simulation experiments

2.1 Chemistry-transport models

Two CTMs are used for this study. [CHIMERE simulations \(Menut et al., 2013\) allow us to perform inter-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.](#) The sensitivity analysis is undertaken using the Polyphemus modeling platform of air quality (Mallet et al., 2007) with the chemistry transport model Polair3D (Sartelet et al., 2012). ~~Results are compared to simulations by the CHIMERE-CTM (Menut et al., 2013).~~
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Table 2.1 summarizes the main characteristics of the simulations.
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Table 2.1. Main characteristics of the Polyphemus and CHIMERE simulations.

Meteorology	ECMWF
Boundary conditions	From nesting simulation
Chemical mechanism	- Polyphemus: CB05 - CHIMERE: Melchior2
Horizontal resolution	- Large domain: $0.5^{\circ} \times 0.5^{\circ}$ - Small domain: $0.25^{\circ} \times 0.25^{\circ}$
Vertical resolution	- Polyphemus: 14 levels (surface–12km) - CHIMERE: 19 levels (surface–200hPa)
Biogenic emissions	MEGAN
Anthropogenic emissions	EMEP
Fire emissions	APIFLAME
Dust emissions	Surface and soil databases (Menut et al., 2013) Briant et al. (2017)

For the simulations presented in this work, Polyphemus is used with the Carbon Bond 05 model (CB05) (Yarwood et al., 2005) for the gas-phase chemistry (modified following Kim et al. (2011) for SOA formation) and with the Size resolved Aerosol Model (SIREAM) (Debry et al., 2007) for aerosol dynamics (coagulation, condensation/evaporation). SIREAM uses a sectional approach and the dry particle diameter is discretized with 5 sections between 0.01 μm and 10 μm . The simulations are performed on 14 vertical levels extending from the ground to 12 km.

The version 2016 of the regional CTM CHIMERE is used for this work (Menut et al., 2013; Mailler et al., 2016). Simulations were conducted using the reduced chemical mechanism Modele Lagrangien de Chimie de l’Ozone a l’échelle Regionale 2 (MELCHIOR2), which includes 44 species and 120 reactions, and the aerosol module described in Bessagnet et al. (2004) (nucleation, coagulation, absorption). This module is also based on a sectional representation of the size distribution. For this simulations, 10 bins from 40 nm to 40 μm are used and simulations are performed on 19 vertical levels extending from the surface to 250 hPa (σ -pressure coordinates).

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. \(2017\).](#) Thermodynamics of inorganic aerosols are modeled using ISORROPIA (Nenes et al., 1999) with a bulk equilibrium approach. Bulk equilibrium is also used for SOA formation, and the partitioning between the gas and particle phases is done with The Secondary Organic Aerosol Processor (SOAP) (Couvidat and Sartelet, 2015) in Polyphemus. Photolysis rates are calculated using the FastJX model (version7.0b for CHIMERE) (Wild et al., 2000). Their online calculation in CHIMERE allows to represent the attenuation by clouds and aerosols, while the attenuation by clouds in Polyphemus is modeled by multiplying the clear-sky photolysis rates by a correction factor (Real and Sartelet, 2011).

Both models (Polyphemus and CHIMERE) are driven by meteorological conditions simulated by the European Center for Medium-Range Weather Forecasts (ECMWF, [ERA-Interim](#)) model. Initial and boundary conditions from MOZART4-GEOS5 6-hourly simulation outputs are used (Emmons et al., 2010). Simulations are undertaken using two nested domains ([Figure A1 in Appendix A](#)). One large domain ($0.5^{\circ} \times 0.5^{\circ}$ horizontal resolution) covering Europe and North Africa to provide realistic dust sources and a smaller domain at $0.25^{\circ} \times 0.25^{\circ}$ horizontal resolution over the Mediterranean area (presented in Figure 2.1).

Anthropogenic emissions are derived from the EMEP emissions inventory [for 2007](#) (European Monitoring and Evaluation Program, [www.emep.int](#)). The inventory species are disaggregated into real species using speciation coefficients (Passant, 2002). The aggregation into model species is done following Middleton et al. (1990). Primary particulate matter emissions are given in total mass by the EMEP emission inventory. They are speciated into dust, primary organic emissions (POA) and black carbon (BC), and distributed into 5 diameter bins (Sartelet et al., 2007).

Biogenic emissions [of isoprene and terpenes \(\$\alpha\$ -pinene, \$\beta\$ -pinene, limonene and humulene\)](#) are calculated using the Model of Emissions of Gases and Aerosols from Nature (~~MEGAN~~) [with the standard MEGAN LHIV database MEGAN-L for Polyphemus and MEGAN v2.04 for CHIMERE](#) (Guenther et al., 2006). Sea-salt emissions are parameterized following Monahan (1986). The mineral dust emissions are calculated using soil and surface databases (Menut et al., 2013) and with a spatial extension of potentially emitting area in Europe as described in Briant et al. (2017). The daily fire emissions are detailed in section 2.2.

In Polyphemus, I/S/L-VOC emissions are estimated by multiplying the primary organic emissions (POA) by a factor of 1.5, following the ~~measurements of different samples representative of the French vehicular fleet~~ (Zhu et al., 2016; Zhu et al., 2016) [chamber measurements](#) (Robinson et al., 2007; Robinson et al., 2016; Robinson et al., 2016). The factor of 1.5 is used for both anthropogenic and fire emissions to estimate the gas-phase I/S/L-VOCs that are not included in the inventories. I/S/L-VOCs emissions are assigned to 3 surrogates species: POAIP, POAmP and POAhP (for compounds of low, medium and high volatilities respectively), of saturation concentration C^* : $\log_{10}(C^*) = -0.04, 1.93, 3.5$ respectively. The volatility distribution at emissions of I/S/L-VOCs is detailed in Couvidat et al. (2012) (25%, 32%, and 43% of I/S/L-VOCs are assigned to POAIP, POAmP and POAhP respectively). It corresponds to the volatility distribution measured by May et al. (2013a) for biomass burning [aerosol emissions](#). Each primary ~~aerosol~~ [I/S/L-VOCs](#) undergoes one OH-oxidation reaction in the gas phase with a kinetic rate constant equal to 2.10^{-11} ~~molecule⁻¹.cm³.s⁻¹~~ [molecule⁻¹.cm³.s⁻¹](#), leading to the formation of secondary ~~surrogate in the same volatility bin~~ [surrogates](#): SOAIP, SOAmP and SOAhP. The ageing of the primary aerosols reduces the volatility of the secondary product by a factor of 100 and increases the molecular weight by 40% (Couvidat et al., 2012).

30 2.2 Fire emissions

Daily fire emissions are calculated using the APIFLAME fire emissions' model v1.0 described in Turquety et al. (2014). The carbon emission associated with a specific fire is calculated using the MODIS burned area product at 500 m resolution (MCD64 product) (Giglio et al., 2009), multiplied by the consumed fuel load specific to the vegetation burned. The CORINE LAND COVER (CLC) is used for vegetation attribution, and the biomass density is estimated based on simulations by the ORCHIDEE

vegetation dynamics and carbon cycle model (Maignan et al., 2009). Turquety et al. (2014) estimated an uncertainty of $\sim 100\%$ on daily carbon emissions using an ensemble approach. This is in agreement with estimates for other daily inventories (e.g. GFED (Van der Werf et al., 2010)).

Emissions for each species are derived from the carbon emissions using the emissions factors from Akagi et al. (2011). These emission factors are provided in terms of g species per Kg Dry Matter (DM) burned (g.kg^{-1}) for all relevant species observed in biomass burning plumes and for different standard vegetation types that match to Mediterranean landscapes (chaparral, temperate forest, crop residue, pasture maintenance and savanna). The contribution of these vegetation types to the burned area detection over the Mediterranean region during the time period studied is 37.2% for temperate forest, 32.7% for savanna, 9.6% for chaparral and 19.9% for crop residue. Emissions for inventory species are then converted into emissions for model species using model-specific aggregation matrices (Yarwood et al., 2005). For aerosols, the difference between emissions factors provided for the total $\text{PM}_{2.5}$ and for the main primary emissions (organic and black carbon, small amounts of inorganics) and for the total $\text{PM}_{2.5}$ is modelled as a specific, inert and unidentified species grouping other fine mode primary particulate matter PPM_{fine} . PPM_{fine} corresponds to all the unidentified fine particles emitted by wildfires which are incorporated to consider the differences between $\text{PM}_{2.5}$ emissions and the total of all $\text{PM}_{2.5}$ speciated emissions. The difference between PM_{10} and $\text{PM}_{2.5}$ emission factors is attributed to emissions of coarse mode PM ($\text{PPM}_{\text{coarse}}$).

Figure 2.1 shows a map of total primary organic carbon emissions (OC) from fires in the Euro-Mediterranean region during the summer 2007. Four main areas are affected by wildfires: Balkan (sub-region MedReg1), Greece (sub-region MedReg2), Southern Italy (sub-region MedReg3) and Algeria (sub-region MedReg4).

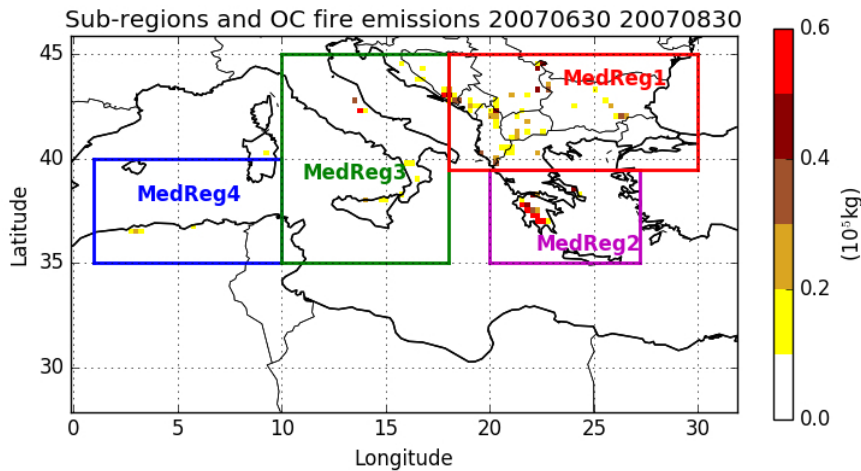


Figure 2.1. Map of the nested domain over the Mediterranean area with a spatial resolution of $0.25^\circ \times 0.25^\circ$. The total organic carbon emissions ($\text{kg. (grid cell)}^{-1}$) from fires during the summer of 2007 are presented. The sub-regions used in this study are also indicated in colored boxes: MedReg1 (Balkan + Eastern Europe), MedReg2 (Greece), MedReg3 (Italy) and MedReg4 (Algeria).

Total daily fire emissions for the four studied areas are plotted in Figure 2.2. In all regions, fire emissions are occasional but very intense. The largest fires in the simulated domain occur in Balkan (sub-region MedReg1) between 20 July and 31 July 2007, and in Greece (sub-region MedReg2) between 24 August and 30 August. In Algeria (sub-region MedReg4), fires mainly occur at the end of August and beginning of September (28 August – 1 September). In Southern Italy (sub-region MedReg3), fires are observed between 9 July and 31 July 2007.

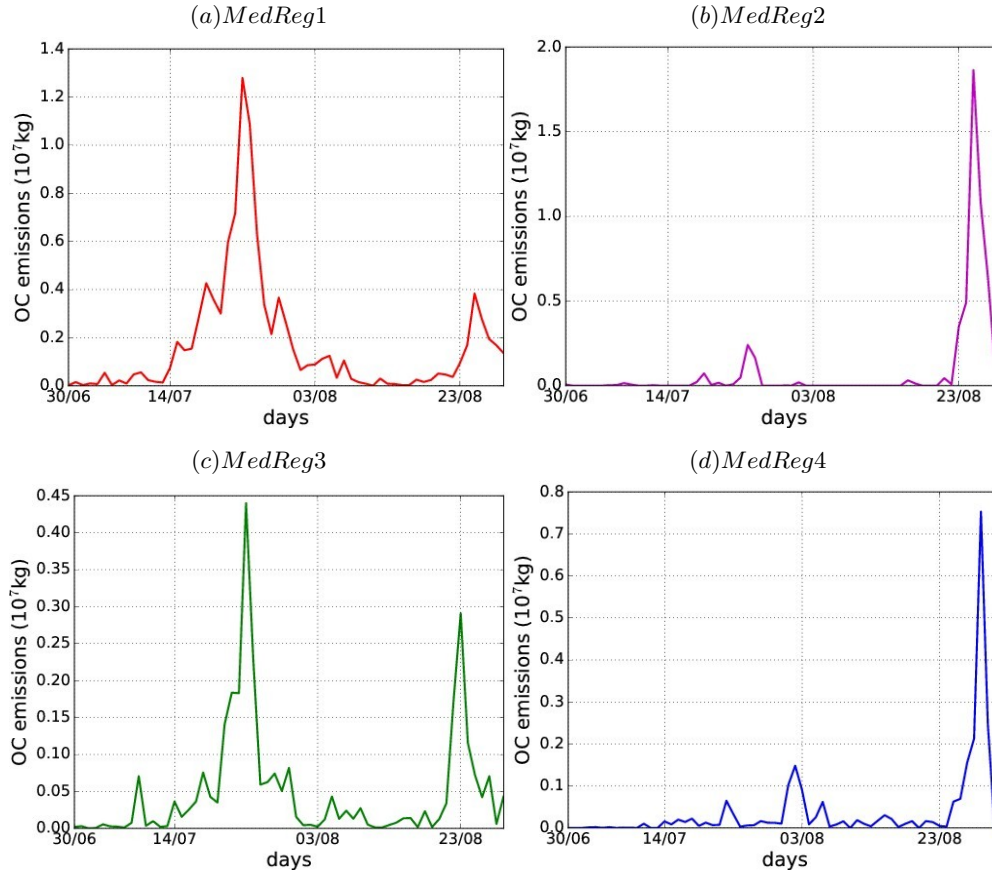


Figure 2.2. Daily total OC emissions calculated by APIFLAME during the summer of 2007 in the four sub-regions of Figure 2.1.

2.3 Model sensitivity experiments

Two different parameters, considered critical for modeling fire events, are tested using sensitivity simulations with the Polyphemus CTM: fire emissions of I/S-VOCs and emissions’ injection heights. Another simulation is also conducted to evaluate the impact of PPM_{fine} emissions. In the simulations discussed here, four different configurations of the model are used:

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1. Simulation *Poly-ref* : fire emissions are homogeneously mixed the planetary boundary layer (PBL), but no lower than 1 km. The percentage of fire emissions injection is divided homogeneously depending on layers’ thickness (I/S/L-VOCs included).
 2. Simulation *Poly-noI/S-VOCs* : fire emissions in the PBL but without I/S-VOCs. ~~POA emissions are taken into account and modelled~~ S-VOC emissions in the gas phase. In this sensitivity study, the semi-volatile properties of POA are not considered, and POA emissions are modeled as LVOCs.
 3. Simulation *Poly-NoPPM* : fire emissions in the PBL, with I/S/L-VOCs but without PPM_{fine} .
 4. Simulation *Poly-3km* : fire emissions injected up to 3km with: 20% under 1km, 80% between 1 and 3km. Note that, in this case, 78% of fire emissions are injected above the PBL. 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, Mims et al. (2010) estimated that 26% of the fire plumes exceed the boundary layer.

In addition, a simulation with the CHIMERE model (without I/S-VOCs and with fires in the PBL)(*CHIMERE-ref*) allows an inter-model comparison. Simulations without fires are also computed with both models (*Poly-Nofires* and *CHIMERE-Nofires*). The set up of the different simulations is summarized in Table 2.2.

Table 2.2. Summary of the configurations used in different simulations. (N/A: not applicable)

Simulation	Fire emissions	I/S-VOCs from fire	Fire emissions' injection height	PPM_{fine}
Simulation <i>Poly-ref</i>	Yes	Yes	Between 1km and PBL	Yes
Simulation <i>Poly-3km</i>	Yes	Yes	20% under 1km 80% between 1 and 3km	Yes
Simulation <i>Poly-NoPPM</i>	Yes	Yes	Between 1km and PB -PBL	No
Simulation <i>Poly-noI/S-VOCs</i>	Yes	No	Between 1km and PB -PBL	Yes
Simulation <i>CHIMERE-ref</i>	Yes	Yes No	Between 1km and PB -PBL	Yes
Simulation <i>CHIMERE-Nofires</i>	No	N/A	N/A	N/A
Simulation <i>Poly-Nofires</i>	No	N/A	N/A	N/A

3 Model evaluation

3.1 Observations

Daily observations from the European network AIRBASE 5 are used for PM concentrations. Due to the relatively coarse horizontal resolution, only background stations are included in the present paper. The investigated stations are indicated in Figure 3.1. PM₁₀ concentrations in the Mediterranean area are strongly affected by dust, which are difficult to simulate due to their sporadic nature and the fact that their main sources are located out of the model domain. ~~Since dust is not in the scope of this paper~~In order to evaluate more specifically the uncertainties associated with fine particles (largest contribution from fires), and to minimize the contribution from dust, the analysis focuses on PM_{2.5}.

Surface observations are complemented by remote sensing observations of aerosol optical properties. AOD level 2.0 data (at 550 nm) and Angstrom coefficient α (derived from AOD at 500 and 870 nm) from the AERONET (AErosol RObotic NETwork) ground-based network of sun photometers (Holben et al., 1998) are used. The uncertainty on AOD is estimated to less than 0.02 (Holben et al., 2001). AOD level 2.0 observations are missing in Lecce University (in Italy) during the first event, and in Blida (in Algeria) during the second one. Since AOD Level 1.5 observations at 500 nm (before filtering) are available for the latter, we have chosen to include these observations for comparison in the next sections. The considered stations are indicated in Figure 3.1.

AOD observations at 550 nm from the MODIS instrument on board the Terra (equator crossing time at 10:30, ascending node) and Aqua (equator crossing time at 13:30, ascending node) satellites are also used in order to get a more complete regional view. The MOD04 and MYD04 (for Terra and Aqua, respectively) from collection 5.2 data, available at $10 \times 10 \text{ km}^2$ are used (Remer et al., 2005). The expected uncertainty on AOD is $\Delta\tau = \pm 0.05 \pm 0.15\tau$ (τ is the optical thickness) over land (Chu et al., 1998; King et al., 1999) and $\Delta\tau = \pm 0.03 \pm 0.05\tau$ over ocean (King et al., 1999), with a good agreement with ground based measurements (Remer et al., 2005). Deep Blue AOD (Sayer et al., 2013) is used when available (over bright areas).

3.2 Comparison method

A set of statistical indicators are used for the comparison of model simulations to surface measurements: the root mean square error ($RMSE$), the correlation coefficient (R), the mean fractional bias (MFB) and the mean fractional error (MFE). These are defined as:

$$5 \quad RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (c_i - o_i)^2} \quad (1)$$

$$R = \frac{\sum_{i=1}^n (c_i - \bar{c})(o_i - \bar{o})}{\sqrt{\sum_{i=1}^n (c_i - \bar{c})^2} \sqrt{\sum_{i=1}^n (o_i - \bar{o})^2}} \quad (2)$$

$$MFB = \frac{1}{n} \sum_{i=1}^n \frac{c_i - o_i}{(c_i + o_i)/2} \quad (3)$$

10

$$MFE = \frac{1}{n} \sum_{i=1}^n \frac{|c_i - o_i|}{(c_i + o_i)/2} \quad (4)$$

with o_i the observed concentration at time and location i , c_i the modeled concentration at time and location i , and n the number of data.

Boylan and Russel (2006) proposed for PM that a model performance criterion (level of accuracy acceptable for standard
15 modeling applications) is met when $MFE \leq +75\%$ and MFB is within $\pm 60\%$, and a model performance goal (level of accuracy considered to be close to the best a model can be expected to achieve) is met when $MFE \leq 50\%$ and MFB is within $\pm 30\%$. In the following, the MFB and MFE are computed at each station and averaged.

3.3 Overview of the three months period

The statistical evaluation of the simulations during the summer 2007 (from 15 July to 30 August 2007) is presented in
20 Table 3.1 for $PM_{2.5}$ concentrations and Table 3.2 for AOD at 550 nm. Globally, the $PM_{2.5}$ concentrations and AOD are well reproduced by the models, although they are slightly underestimated compared to measurements. For AOD, the model performance and goal are always met. For $PM_{2.5}$ concentrations, the model performance is always met, and the model goal is met for the model errors MFEs. The model-to-measurements correlations range between 46% and 83% for all the simulations when fires are included. The model errors MFEs are similar for outputs from CHIMERE and Polyphemus. However,
25 CHIMERE outputs have lower biases (higher concentrations are AOD values), while Polyphemus outputs have higher correlations. The models-to-measurements comparisons tend to improve when fire are taken into account, with lower MFEs and higher correlations. The improvement is stronger for AOD than for $PM_{2.5}$ concentrations, because the stations used for AOD models-to-measurements comparisons are closer to regions affected by wildfires than the stations used for $PM_{2.5}$ models-to-measurements comparisons, as discussed below.

Figure 3.1 shows the mean surface concentrations of PM_{2.5} simulated by Polyphemus (*Poly-ref*) over the Euro-Mediterranean domain, from 15 July to 30 August 2007. The 8 AIRBASE stations used for the models-to-measurements comparison are also presented in Figure 3.1. The mean simulated PM_{2.5} concentrations can reach 60 ~~$\mu\text{g}\cdot\text{m}^{-3}$~~ $\mu\text{g}\cdot\text{m}^{-3}$ in regions affected by wildfires (Balkan, sub-region MedReg1 and Greece, sub-region MedReg2). Only two AIRBASE stations, GR0039A in Greece and IT0459A, in Italy are close to regions affected by wildfires.

Figure [B1 in Appendix B](#) shows that conclusions for CHIMERE are similar to those for Polyphemus.

[Figure 3.1](#) also shows the mean modeled AOD, which can be as high as 0.72 in average in Balkan and in Greece, and the 6 Aeronet stations used for the models-to-measurements comparisons. 5 out of the 6 stations are located in regions affected by wildfires.

This evaluation shows good general performance of the models and a clear improvement of PM_{2.5} and AOD when fires are included, allowing a more precise analysis of the model’s behavior during the strongest fire events.

Table 3.1. Statistics of models-to-measurements comparisons for the mean daily PM_{2.5} concentrations during the summer 2007 (AIRBASE station number = 8).

Simulations	Mean observed PM _{2.5}	Mean simulated PM _{2.5}	Correlation (%)	MFB (%)	MFE (%)
<i>Poly-ref</i>	13.2	9.3	82	-32	42
<i>Poly-noI/S-VOCs</i>	13.2	9.3	82	-32	42
<i>Poly-3km</i>	13.2	9.1	82	-33	43
<i>Poly-NoPPM</i>	13.2	9.1	82	-33	43
<i>Poly-Nofires</i>	13.2	8.4	78	-37	46
<i>CHIMERE-Nofires</i>	13.2	11.2	70	-15	39
<i>CHIMERE-ref</i>	13.2	11.3	67	-10	39

Table 3.2. Statistics of models-to-measurements comparisons for mean daily AOD at 550 nm during the summer 2007 (AERONET station number = 6).

Simulations	Mean observed AOD	Mean simulated AOD	Correlation (%)	MFB (%)	MFE (%)
<i>Poly-ref</i>	0.27	0.22	62	-14	34
<i>Poly-noI/S-VOCs</i>	0.27	0.21	64	-18	35
<i>Poly-3km</i>	0.27	0.22	64	-15	34
<i>Poly-NoPPM</i>	0.27	0.22	62	-14	34
<i>Poly-Nofires</i>	0.27	0.19	56	-24	39
<i>CHIMERE-Nofires</i>	0.27	0.23	36	-7	39
<i>CHIMERE-ref</i>	0.27	0.24	46	-6	36

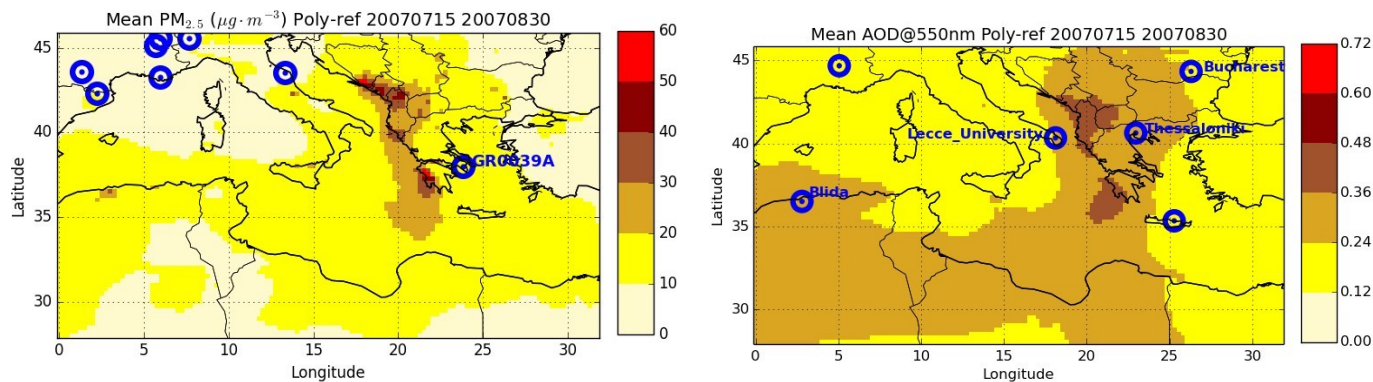


Figure 3.1. Daily mean [surface](#) PM_{2.5} and AOD at 550 nm from the *Poly-ref* simulation averaged over the summer of 2007 (the 8 AIRBASE and 6 AERONET stations, used in this work, are represented here in blue dots).

3.4 Fire events

To better understand the sensitivity of PM_{2.5} concentrations and AOD to I/S-VOCs emissions and the fire injection heights during the fire events, the composition of PM during fire peaks and the evaluation of fire contribution are examined. The locations of the fire peaks during the two main fire events are first detailed.

5 3.4.1 Locations of the fire peaks

The contribution of fires to PM_{2.5} concentrations simulated by Polyphemus during the two main fire events (20-31 July and 24-30 August) is presented in Figure 3.2 as the relative difference between the reference simulation (*Poly-ref*) and the simulation without fire emissions (*Poly-Nofires*). The largest contribution is simulated over the Balkan and Eastern Europe during the first period (sub-region MedReg1), and Greece (sub-region MedReg2) and Algeria (sub-region MedReg4) during the second

period. The impact reaches up to 90% locally on average during each fire event. The contribution of fires remains large ($>60\%$) over most of the eastern Mediterranean basin, and part of the western basin at the end of August due to long-range transport of fire plumes. [Figure C1 in Appendix C shows that the contribution of fire for CHIMERE are similar to that for Polyphemus.](#) Only the closest AIRBASE and AERONET stations to fire regions (where fire contribution is higher than 50%) are used in models-to-measurements comparisons in the next sections. During the first fire event, the most affected stations are: GR0039A in Greece (sub-region MedReg2) and Bucharest in Romania (sub-region MedReg1), while during the second fire event, stations in Greece (Thessaloniki in sub-region MedReg2), Italy (Lecce University in sub-region MedReg3) and in Algeria (Blida in sub-region MedReg4) are the most influenced. However, since the models show the same behavior during the second fire event over the stations GR0039A and Thessaloniki, we choose to focus only on GR0039A.

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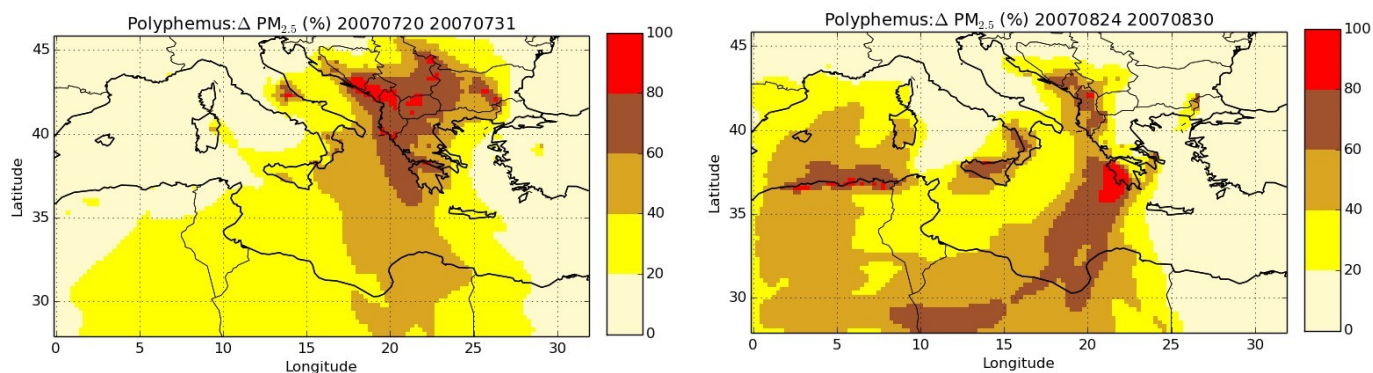


Figure 3.2. Left panel: relative difference of [surface](#) $PM_{2.5}$ concentrations between simulations *Poly-ref* and *Poly-NoFires* during the first fire event. Right panel: relative difference of [surface](#) $PM_{2.5}$ concentrations between simulations *Poly-ref* and *Poly-NoFires* during the second fire event.

3.4.2 Aerosol composition during fire peaks

~~Figure 3.3 shows the~~ [The](#) composition of surface $PM_{2.5}$ concentrations simulated during the first ~~peak (on 25 July 2007) at the AIRBASE station GR0039A~~ fire event over MedReg1 and during the second one over MedReg2 are shown in Figures 3.3 and 3.4 respectively. These two subregions are the areas most affected by fires (high fire emissions (Figure 2.1) especially during the first fire event for Medreg1 and during the second fire event for MedReg2). The upper left panel shows the composition of surface $PM_{2.5}$ concentrations for the simulation without fire *Poly-NoFires* (background surface $PM_{2.5}$ concentrations), while the upper right panel shows the composition of surface $PM_{2.5}$ concentrations due to fires (differences between the simulations *Poly-ref* and *Poly-NoFires*). ~~In the~~ [If wildfires are not taken into account \(simulation without fire\) and during the first fire event over MedReg1, organic and inorganic aerosols contribute equally \(42.6%, organic aerosols, inorganic aerosols and \$PPM_{fine}\$ \(dust\) contribute equally 40.5%\)](#) to the surface $PM_{2.5}$ concentrations (35, 31 and 34). The contribution of ~~black carbon is low (about 5 PPM_{fine} (dust), black carbon are lower (15%, 1%).~~ As noted by Chrit et al. (2017), most of sum-

mer organic aerosols are from biogenic sources in this region. ~~Figure 3.3 also shows~~ If wildfires are not taken into account during the second fire event over MedReg2, inorganics and PPM_{fine} are the predominant component in the composition of $PM_{2.5}$ concentrations (56.5% and 27.9%). Lower contributions are simulated for black carbon (1.2%) and organic aerosol (14.3%). Figures 3.3 and 3.4 also show the composition of surface $PM_{2.5}$ concentrations due to fires for the simulations *Poly-ref*, *Poly-NoI/S-VOCs* and *Poly-NoPPM* (differences between the simulations *Poly-ref* and *Poly-Nofires*), *Poly-NoI/S-VOCs* (differences between the simulations *Poly-NoI/S-VOCs* and *Poly-Nofires*), *Poly-NoPPM* (differences between the simulations *Poly-NoPPM* and *Poly-Nofires* respectively). Organic). During the first fire event over MedReg1, organic aerosol is predominant in the contribution of fires (between 52.47% and 87.85% of the contribution). Organic aerosol is mostly composed of POA and SOA from I/S/L-VOCs (62.46 to 84.46 to 80%). Note that POA and SOA from L-VOCs (low volatile organic compounds) are important even in the simulation when I/S-VOCs are not taken into account in fire emissions (49.46%), because POA are then assigned to L-VOCs. The contribution from inorganics (7.108 to 13%) and black carbon (2.3 to 6%) are low.

During the second fire event over MedReg2, similar $PM_{2.5}$ composition is found. Organic aerosol (mainly the POA and SOA from I/S/L-VOCs) is the most important component contributing to $PM_{2.5}$ from fires (between 46% and 81% of the contribution). The contribution from inorganics (9 to 12%) and black carbon (5 to 6%) are lower. The contribution of PPM_{fine} from fires (if it is included) is very significant, 27% for the simulations with I/S-VOCs and 36% otherwise. Because PPM_{fine} emissions are incorporated to consider the difference between $PM_{2.5}$ emission factors and the total of all PM included in specific species, the contribution of PPM_{fine} may be overestimated (double counting with I/S-VOCs for instance).

In our study, inorganics (mainly sulfate, sea salt and ammonium) contribute highly to $PM_{2.5}$ composition, if fire emissions are not considered. Similar results are found in Fountoukis et al. (2011) who showed the high contribution of sulfate, sea salt and ammonium to PM over Europe during May 2008. However, when fire emissions are taken into account, the contribution of inorganics becomes lower than the contribution of organics (8 to 9% of inorganics against 40 to 80% of organics). Focusing on the contribution from fires, sulfate, ammonium and nitrate are the predominant components in the composition of inorganics from fires: between 55.7% and 67.6% for sulfate, between 26.8 and 38.7% for ammonium and 5.6 to 13.6% for nitrate.

Similar $PM_{2.5}$ composition is found during the second fire peak, and in the concentrations simulated by CHIMERE (not shown here). Surface $PM_{2.5}$ concentrations from fire simulated by CHIMERE are composed in the first and second peak events mainly of organic aerosol, mostly composed by primary organic carbon (OCAR) (48.46%) which corresponds to I/S/L-VOCs in Polyphemus, and of PPM_{fine} (37.39%). The contributions from inorganics (8.29%), black carbon (4.45.2%) and SOA (2.7%) are low.

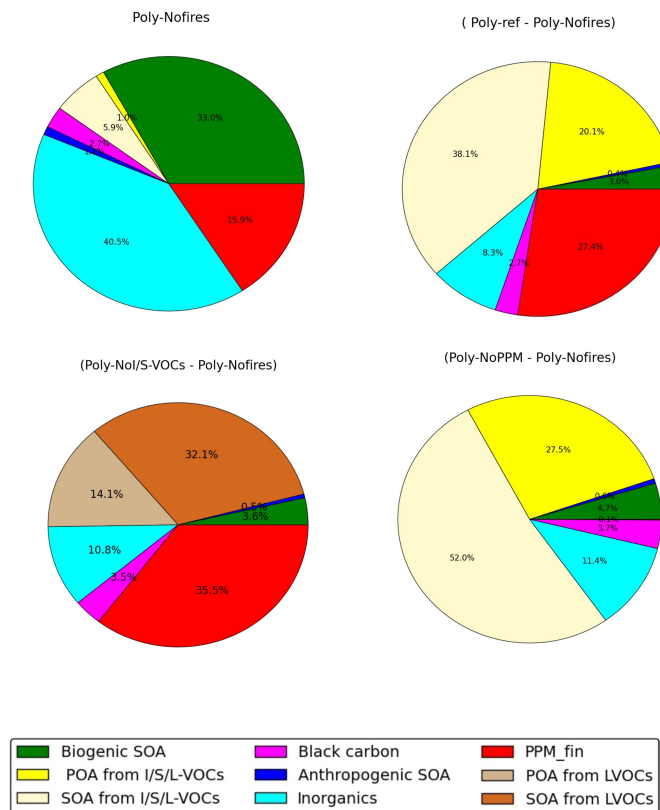


Figure 3.3. Composition of surface PM_{2.5} concentration at over MedReg1 during the GR0039A station on 25 July 2007 first fire event (upper left panel, simulation Poly-nofires Poly-Nofires). Composition of surface PM_{2.5} concentration due to fires (upper right panel: simulation Poly-ref; lower left panel: simulation Poly-NoI/S-VOCs; lower right panel: simulation Poly-NoPPM).

3.4.3 Evaluation of fire contribution

Figure 3.5 shows time series of daily observed and simulated aerosols at stations with significant impact from fires background suburban and background rural stations with available data during fire events and with a fire contribution higher than 10% (PM_{2.5} in Greece (sub-region MedReg2), AOD at 550 nm in Italy (sub-region MedReg3), Romania (sub-region MedReg1) and Algeria (sub-region MedReg4). A significant increase in AOD and PM_{2.5} concentration is observed during the major fire episodes on 20–28 July and 24–30 August, associated with large contributions from fires.

The daily average AOD, observed by MODIS and simulated for the sub-regions of Figure 2.1, are shown in Figure 3.6. Background AOD and daily variability are consistent with MODIS for both models in the sub-regions of Figure 2.1, with high correlation coefficients (~90% on average for Polyphemus, 80% for CHIMERE).

As shown in Figures 3.5 and 3.6, injecting emissions higher (simulation *Poly-3km*) significantly lowers surface PM concentrations (compared to the simulation *Poly-ref*), even if the maximum injection height remains conservative. Not taking into

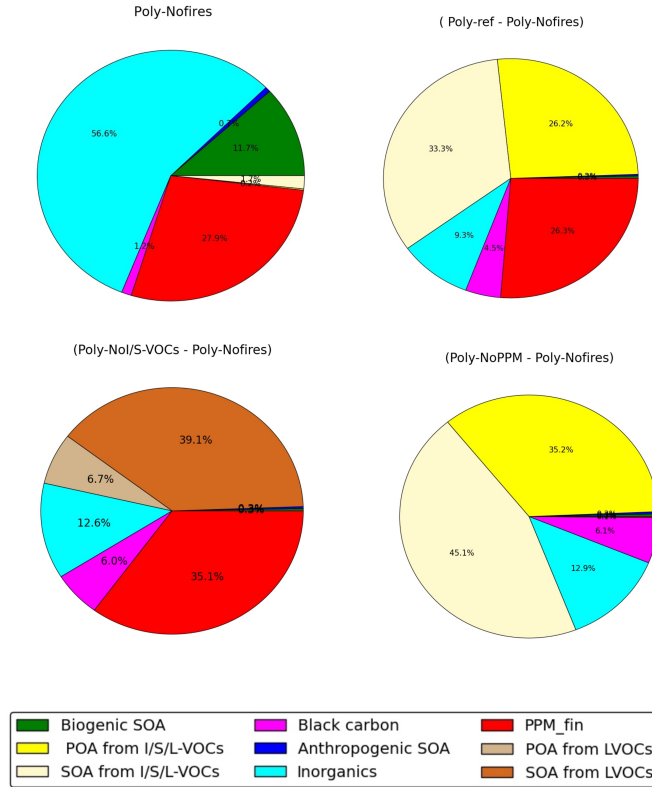


Figure 3.4. Composition of surface PM_{2.5} concentration over MedReg1 during the first fire event (upper left panel, simulation Poly-Nofires). Composition of surface PM_{2.5} concentration due to fires (upper right panel: simulation Poly-ref; lower left panel: simulation Poly-NoI/S-VOCs; lower right panel: simulation Poly-NoPPM).

account SOA from I/S-VOCs directly reduces emissions thus having strong influence on PM concentrations (-20% compared to *Poly-ref*). A reduction of the same order of magnitude is obtained when PPM_{fine} is not accounted for (*Poly-NoPPM*), suggesting that the incorporation of this species in APIFLAME could compensate for the missing I/S-VOCs emissions.

For the first fire event, Figure 3.7 shows maps of the daily mean AOD at 550 nm from MODIS, modeled by Polyphemus (Poly-Ref) and CHIMERE (CHIMERE-ref). The simulated AOD is generally in a good agreement with the observations in terms of localization. The AOD calculated from Poly-Ref and CHIMERE-ref simulations is close to observations in Balkan. However, it seems overestimated in the fresh plume and further downwind (reaching ~0.65 for Poly-Ref and 0.52 for CHIMERE-ref). Results of Poly-noI/S-VOCs are close to those of the CHIMERE model, which does not include I/S-VOCs emissions. Compared to MODIS AOD (Figure 3.6), the simulations including I/S-VOCs (*Poly-Ref*, *Poly-3km* and *Poly-NoPPM*) overestimate AOD during the fire events, while simulations without I/S-VOCs underestimate AOD. This is more pronounced in the two sub-regions MedReg1 and MedReg2, where the mean modeled AOD values are overestimated by about 30% for *Poly-Ref* and *Poly-3km* and

10% for *Poly-NoPPM*. Compared to AIRBASE ground measurements (Figure 3.5), the peak corresponding to the first event (25 July) is well modeled in the simulation *Poly-ref* compared to PM_{2.5} observations at GR0039A (Athens suburbs, Greece, sub-region MedReg2). The temporal variations of the mean simulated PM_{2.5} concentrations are consistent with observations, with high correlation coefficients (>88%). Background levels of PM_{2.5} are slightly underestimated compared to observations.

5 This can be explained by an underestimation of dust long-range transport or an underestimation of local emissions. The peak of PM_{2.5} concentration is slightly underestimated in all the other simulations. At [the GR0035A station, the temporal tendencies of the simulated PM_{2.5} concentrations are consistent with the observations. However, the first PM_{2.5} peak is underestimated since PM_{2.5} levels are strongly underestimated \(bias= -85%\). The observed background levels are significantly higher than those at GR0035A station. This suggests the influence of local emissions that are missing in the model or limitations due to the](#)

10 [coarse model resolutions which can not represent this station.](#)

[At](#) Bucharest, two high AOD peaks are modeled (Figure 3.5). Compared to AERONET ground observations, the first peak is well modeled on 23 July (0.46 against measurements 0.5) and underestimated on 30 July (0.51 against measurements 0.82 but simulated one day after the observations, probably due to uncertainty in the MODIS fire detection). The observed values of Ångström exponent are lower on 23 July ($\alpha \sim 0.57$), indicating a large fraction of coarse mode particles (probably dust trans-

15 port), than on 30 July ($\alpha \sim 1.6$, large fraction of fine mode particles from fires). These under-estimations or over-estimations of the model's AOD, depending on the data set used for the evaluation (AERONET vs MODIS), underline uncertainties in AOD retrievals from measurements, which have already been observed by numerous studies (Li et al., 2009; Wu et al., 2017; Boiyo et al., 2017).

During the second fire event, the contribution from fires becomes predominant on 24 August (beginning of the event according to MODIS fire observations). The best agreement with PM_{2.5} in Greece is obtained for the simulations *Poly-3km* and CHIMERE, although the latter overestimates surface concentrations on the following days. [At the GR0035A station, the second observed peak during the second fire event is more than twice higher than the observed peak at the GR0039A and is measured one day earlier. In the simulations, the enhancement due to fires is similar in shape and magnitude, clearly highlighting the difficulty to simulate the exact temporal variability of emissions and transport of fire plumes in CTMs. Moreover, the station](#)

25 [GR0035A is probably affected by dust episode during the second fire event.](#)

For AOD at Lecce University, all simulations show good agreement to observations. According to AERONET level 1.5 measurements in Blida (in Algeria, sub-region MedReg5), an AOD peak (0.55) is observed on 27 August. The AOD simulated by CHIMERE and Polypohemus are consistent with the measurements mainly for *Poly-ref*, *Poly-3km*, *Poly-NoPPM* and *CHIMERE-ref*. *Poly-noI/S-VOCs* shows the lowest AOD values at Lecce University (0.38) and Blida (0.33). This suggests

30 that taking into account I/S-VOCs emissions leads to higher and more realistic AOD at these stations.

In Blida, three peaks are simulated for 2 August, 6 August and 16 August. The observed values of the Ångström exponent are equal to $\alpha \sim 1.18$ and $\alpha \sim 1.14$ for the first and third peaks respectively, which indicates fine mode particles. Therefore, the first and third peaks are attributed mostly to fires in Algeria on 2 August and 16 August. However, emissions are probably underestimated, as all the models under-estimate the AOD fire-peaks. Since a lower value of the Ångström exponent is observed

35 on 6 August ($\alpha \sim 0.91$), this second peak is probably attributed to dust.

This analysis highlights the strong regional impact during intense events on both AOD and PM_{2.5} concentrations but also the difficulties in representing their amplitudes and variability. Considering the uncertainty on fire data and emissions (~100%) and on the observations (1-2% for AERONET observations (Eck et al., 1999) and ~34% for AIRBASE observations (Bovchaliuk, 2013)), the performance obtained is considered very reasonable. The spread in the different model configurations tested shows the additional uncertainty on the modeling of fire impact. Observations generally lie within the simulated variability but it is difficult to extract the best model configuration (it depends on the event and on the station).

For the first fire event, Figure 3.7 shows maps of the daily mean AOD at 550 nm from MODIS, modeled by Polyphemus (*Poly-Ref*) and CHIMERE (*CHIMERE-ref*). The simulated AOD is generally in a good agreement with the observations in terms of localization. The AOD calculated from *Poly-Ref* and *CHIMERE-ref* simulations is close to observations in Balkan. However, it seems overestimated in the fresh plume and further downwind (reaching ~ 0.65 for *Poly-Ref* and 0.52 for *CHIMERE-ref*). Results of *Poly-noI/S-VOCs* are close to those of the CHIMERE model, which does not include I/S-VOCs emissions. During the second fire event in Greece, the simulated AOD for *Poly-Ref* and for *CHIMERE-ref* is about 1 and 0.9 respectively as shown in Figure D1 in Appendix D. The observed AOD can reach 0.9. However, both models overestimate AOD values in the fire plume (reaching 0.98 for *Poly-ref* and 0.88 for *CHIMERE-ref* against 0.7-0.8 in Greece and 0.5 from MODIS. The fire plume is less pronounced in observations than in simulations.

Day-to-day comparisons for four selected days (24, 25, 27 and 29 August 2007) are shown in Figure E1 in Appendix E. The simulated AOD is consistent with the observations in terms of localization and general transport pathways. However, the simulated AOD is much higher in the Greek fires' plume compared with MODIS observations during the peak of emissions (25-29 August). This probably reflects too low temporal variability in the emissions. In the simulations, emissions are assumed constant during the day but comparisons suggest shorter temporal variability. This is also apparent in the time series of Figure 3.6, over MedReg2: the peak for the second fire event is twice longer in the simulations (double peak starting on the 25 August 2007) than in the observations. This peak over two days instead of one in the simulations suggests an overestimate of emissions during this event which is also observed with respect to surface observations in Greece. The first peak corresponds better to observations. This shows that uncertainties are not only related to total emissions but also to their temporal variability and the associated transport pathways.

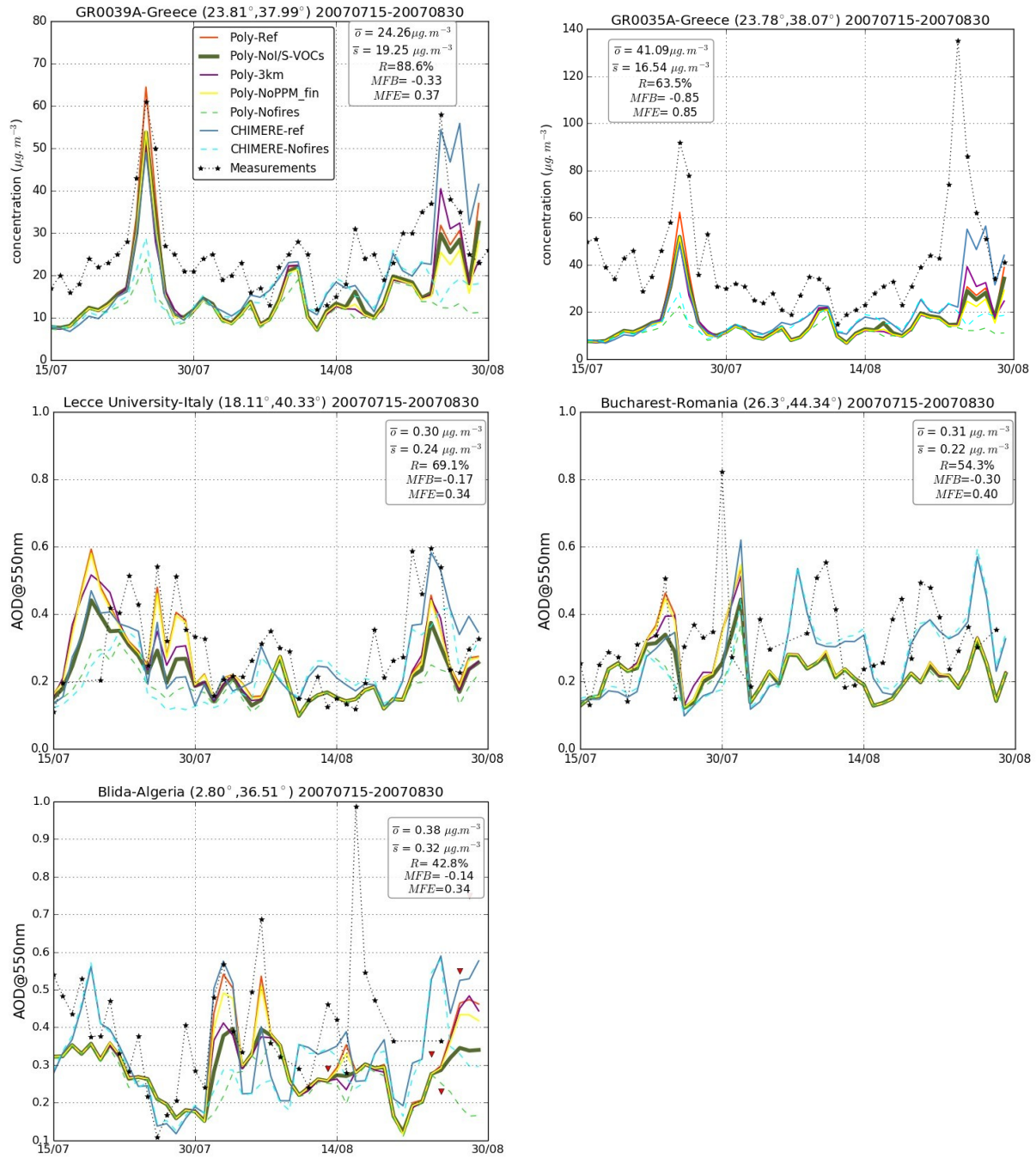


Figure 3.5. Time series from 15 July to 30 August of daily mean surface PM_{2.5} concentrations at the AIRBASE [station-stations](#) GR0039A and [GR0035A](#) daily mean AOD at 550 nm at three AERONET stations (Lecce University, Bucharest, Blida). The red triangles in Blida station correspond to AERONET measurements using AOD level 1.5 AOD data at 500 nm. Statistics for simulation *Poly-Ref* are shown at each station.

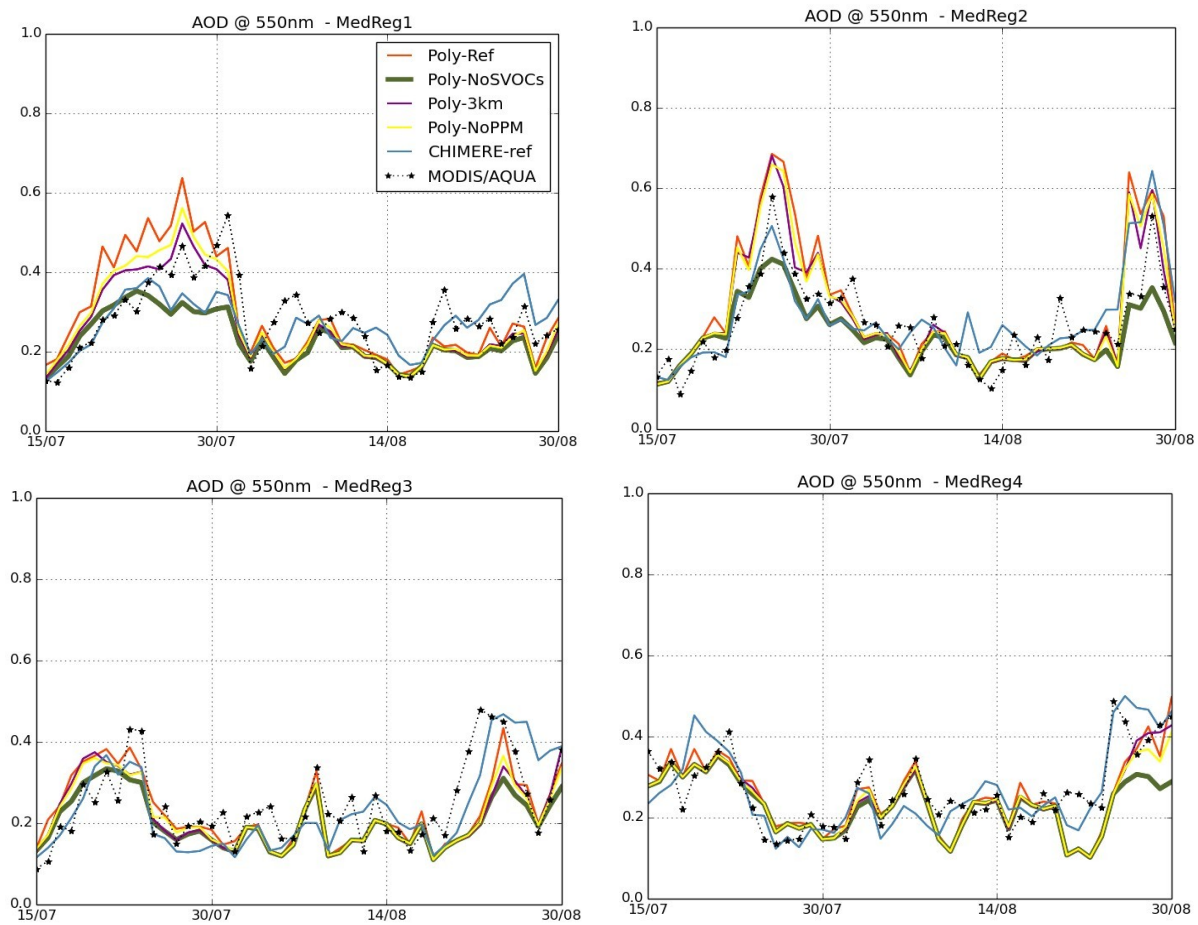


Figure 3.6. Daily mean AOD at 550 nm observed by MODIS and simulated by Polyphemus and CHIMERE from 15 July to 30 August 2007, in the sub-regions of Figure 2.1.

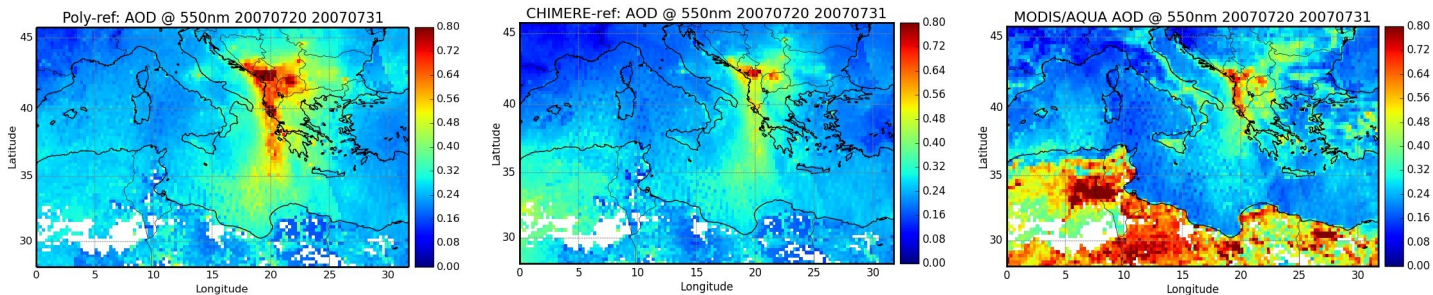


Figure 3.7. Mean total AOD (at 550 nm) from MODIS/AQUA, *Poly-Ref* and *CHIMERE-ref* during the first fire event (20-31 July 2007).

4 Uncertainty and sensitivity analysis

The sensitivity of the modeling of I/S-VOCs emissions and injection heights on simulated surface $PM_{2.5}$ concentrations and AOD is now evaluated regionally over the Mediterranean domain.

The sensitivity of the model results to the I/S-VOCs emissions and injection heights is compared to the inter-model sensitivity presented in Figure 4.1, which shows the relative differences between a sensitivity simulation (*CHIMERE-ref*, *Poly-noI/S-VOCs*, *Poly-3km*) and the reference simulation *Poly-ref*. To focus on fires impact, only the $PM_{2.5}$ concentrations and AOD exceeding $15 \mu g \cdot m^{-3}$ and 0.25 respectively, are taken into account when computing the relative differences between the simulations. It is worthy to note that the arbitrary choice made in this work (injecting between 1 and 3 km) may overestimate the impact of injection height on surface $PM_{2.5}$ concentrations and underestimate it on long range transport (injecting fire emissions at or below 3 km remains conservatively low).

The inter-model sensitivity is low (relative differences below 20%) for both surface $PM_{2.5}$ concentrations and AOD, except in Balkan (sub-region MedReg1), where it can reach 50% locally. The high inter-model differences are slightly more spread horizontally for surface $PM_{2.5}$ concentrations than for AOD. Furthermore, this region of high inter-model sensitivity corresponds to the region where the sensitivity to the injection height is the highest for $PM_{2.5}$ concentrations. It may therefore be linked to differences in the models ~~vertical discretisation (the~~ vertical discretisation. 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.

Neglecting I/S-VOCs emissions leads to a decrease in surface $PM_{2.5}$ concentrations. The impact of I/S-VOCs emissions on surface $PM_{2.5}$ concentrations over the fire regions is mostly under 20%, but reaches 30% locally. The impact of I/S-VOCs emissions is spread over larger areas than the inter-model difference.

I/S-VOCs emissions have a higher impact on AOD than on surface $PM_{2.5}$ concentrations, since adding I/S-VOCs increases the total integrated $PM_{2.5}$ concentrations: the increase can be as high as 40% in Balkan, 30% in Greece and the fire plume.

Injecting above the boundary layer results in larger scale transport for $PM_{2.5}$ concentrations that leads to the highest impact on surface $PM_{2.5}$ concentrations (40 to 50% near the fire regions and 30% in the fresh plume and further downwind). However, the impact of the injection height on AOD is lower, but still significant (mostly under 20%, but reaching 30% locally). Similar

- results are found in Turquety et al. (2007); Chen et al. (2009); Stein (2009); Daskalakis et al. (2015); Gonzi et al. (2015). In fact, previous studies highlighted the high sensitivity of long range transport of carbon monoxide (CO) to wildfires injection height (Turquety et al., 2007; Gonzi et al., 2015). It also leads to a reduction of concentrations at the surface Chen et al. (2009). The sensitivity analysis of Stein (2009) estimates a strong reduction in the surface PM_{2.5} concentrations ($> 10 \text{ } \cancel{\mu\text{g}\cdot\text{m}^{-3}} \text{ } \mu\text{g}\cdot\text{m}^{-3}$)
- 5 caused by fire emissions injection height over the United States. According to Daskalakis et al. (2015), assumptions on the injection heights of fire emissions can also result regionally in up to 30% differences in the calculated tropospheric lifetime of pollutants. This can lead to significant interactions between isoprene and fire emissions. Daskalakis et al. (2015) showed that these interactions affect the effectiveness of isoprene to produce secondary aerosols (up to 18%).
- 10 This analysis highlights that injecting above the boundary layer is more critical for surface PM_{2.5} concentrations than integrating I/S-VOCs emissions, since 78% of fire emissions are emitted above the boundary layer. However, for AOD and vertically integrated concentrations, integrating I/S-VOCs emissions is more critical than the injection heights.

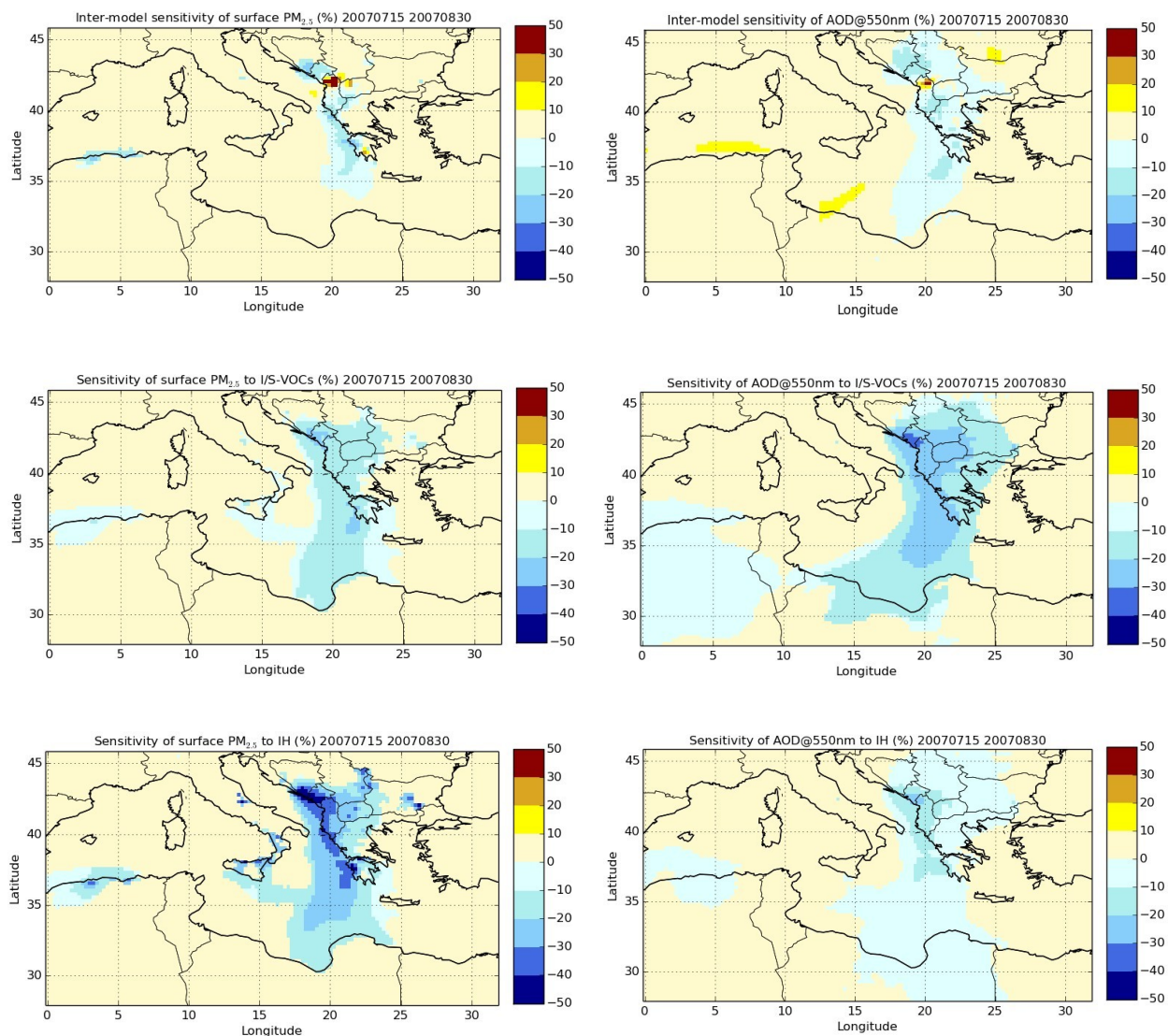


Figure 4.1. Sensitivity of surface $\text{PM}_{2.5}$ concentrations (left panels) and AOD at 550 nm (right panels) to the CTM used (*CHIMERE-ref* or *Poly-ref*, upper panels), the modelling of I/S-VOCs emissions (*Poly-noI/S-VOCs* or *Poly-ref*, middle panels) and the injection height (IH) (*Poly-3km* or *Poly-ref*, lower panels) during the summer 2007 (15 July to 30 August). The simulation *Poly-ref* is used in all panels to estimate the relative differences.

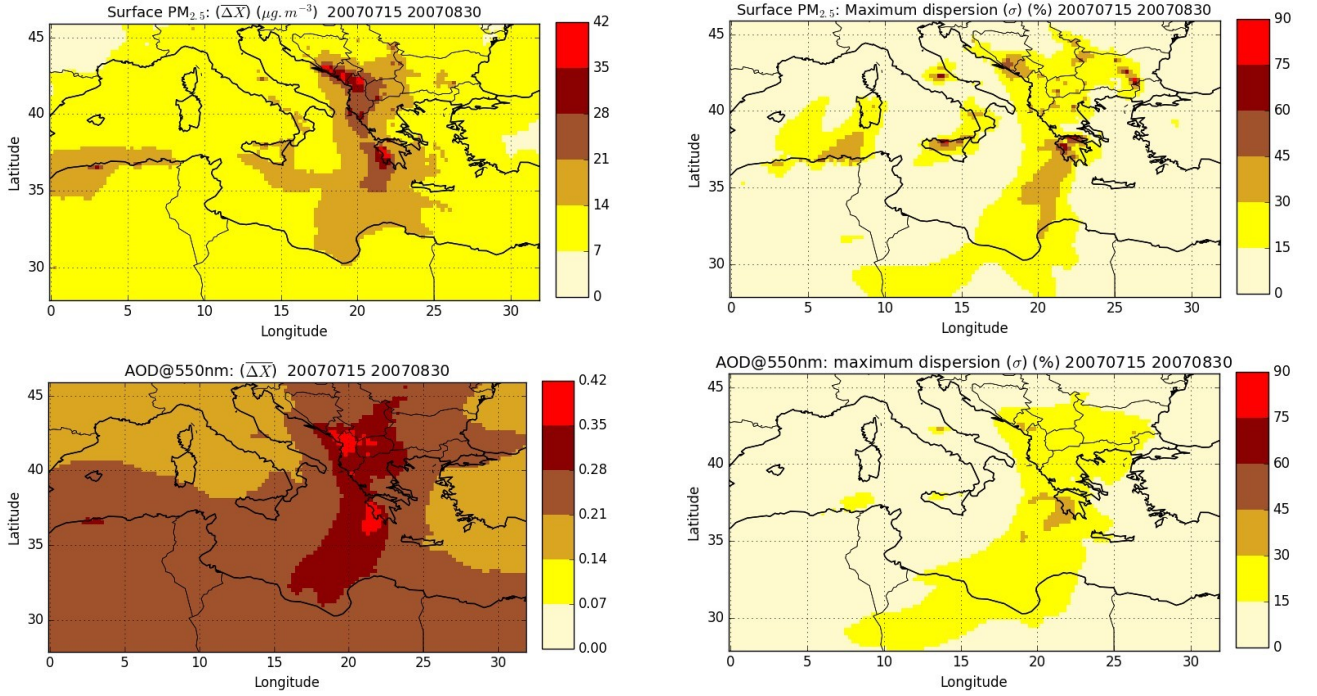


Figure 4.2. Mean surface $\text{PM}_{2.5}$ concentrations and AOD (estimated using four Polyphemus sensitivity simulations) (left panel) and their relative maximum [statistical](#) dispersion (σ) (right panel) during the summer 2007.

The ensemble of the sensitivity simulations provides an estimate of the uncertainty of the modeling of fire plumes. To quantify uncertainties related to fire emissions modeling, the maximum [statistical](#) dispersion (σ) is used as a statistical estimator. The maximum [statistical](#) dispersion (σ) for surface $\text{PM}_{2.5}$ and AOD calculated as:

$$\sigma = \frac{\sqrt{\frac{1}{N} \sum_{i=1}^N (X_i - \overline{\Delta X})^2}}{\overline{\Delta X}} \cdot 100 \quad (5)$$

5

$$\overline{\Delta X} = \frac{1}{N} \sum_{i=1}^N (X_i) \quad (6)$$

with X refers to either surface $\text{PM}_{2.5}$ concentrations or AOD, N is the total number of the simulations with fire emissions included (N is equal to 4).

10

Figure 4.2 shows the average surface $\text{PM}_{2.5}$ concentrations and AOD estimated using four Polyphemus simulations and the maximum [statistical](#) dispersion (σ) for surface $\text{PM}_{2.5}$ concentrations and AOD. The maximum [statistical](#) dispersion related to simulated surface $\text{PM}_{2.5}$ concentrations and AOD is higher near the fire regions. The mean surface $\text{PM}_{2.5}$ concentrations

5 estimated from the four Polyphemus simulations can reach $42 \mu\text{g}\cdot\text{m}^{-3}$ in Balkan and Greece with a statistical dispersion that can be as high as 75%. Lower mean surface $\text{PM}_{2.5}$ concentrations are calculated for Algeria, in the fresh fire plume and further downwind ($28 \mu\text{g}\cdot\text{m}^{-3}$ with a maximum statistical dispersion reaching 45%). The mean AOD estimated as the average of four Polyphemus simulations can reach 0.42 in Balkan and Greece, 0.36 in the fire plume with a statistical dispersion that can reach 45% and 36% respectively.

4.1 Air quality exceedances

Air quality impact is evaluated by analyzing the number of threshold exceedances during the summer of 2007. It corresponds to the number of times daily averaged $\text{PM}_{2.5}$ surface concentrations exceeds $25 \mu\text{g}\cdot\text{m}^{-3}$ (World Health Organisation recommendation, (Krzyzanowski and Cohen, 2008)).

The number of exceedances predicted by the model is first compared to exceedances observed by AIRBASE (~~only one station is available~~). ~~During the~~ stations. Table F1 in Appendix F presents the modeled and observed $\text{PM}_{2.5}$ exceedances at each AIRBASE stations during the whole summer 2007, ~~14~~ and during fire events. Generally, the models (Polyphemus and CHIMERE) underestimates $\text{PM}_{2.5}$ ~~exceedances were observed and only 7 are simulated at~~ air quality exceedances because the horizontal resolution used here does not allow the representation of local pollution (especially for the station GR0035A). Better performance is observed during fire events than the whole period. Near fire regions, at the stations GR0039A ~~by Poly-ref~~ and ~~9 by CHIMERE-ref~~, the number of exceedance is well-modeled, in spite of the slight underestimation during both fire events compared to the observed $\text{PM}_{2.5}$ exceedances. However, at the GR0035A station, the underestimation of $\text{PM}_{2.5}$ exceedances is sharp. This can be explained by the strong underestimation of the background $\text{PM}_{2.5}$ levels as shown in Figure 3.5. Far from fire regions, the $\text{PM}_{2.5}$ exceedances modeled by Polyphemus are in a good agreement with the observed ones especially during the two fire events. The simulated $\text{PM}_{2.5}$ exceedances by CHIMERE in the fire plume are overestimated at some stations. Figure 4.3 shows the ~~simulated number of days concerned by a additional days with~~ $\text{PM}_{2.5}$ ~~exceedance from fires~~ exceedances due to fires simulated by Polyphemus (difference between the *Poly-ref* and *Poly-Nofires*). Most are concentrated around fire sources, mainly in Balkan (30 days). Figure G1 in Appendix G shows that conclusions for CHIMERE are similar to those for Polyphemus. Fires cause up to 49.5% of the total simulated exceedances from 15 July to 30 August 2007.

During the fire periods, surface $\text{PM}_{2.5}$ concentrations simulated in *Poly-ref* in MedReg1 and MedReg2 sub-regions are composed mainly of ~~primary organic matter (7.3OM (54% with 9261.6% due to fire) for MedReg1 and secondary organic aerosol (19.152.3% with 78.960.1% due to fires for MedReg2), and inorganics (27% with 8% due to fires for MedReg1 and 15% with 9% due to fires for MedReg2).~~

Figure 4.3 shows that the maximum statistical dispersion on the simulation of $\text{PM}_{2.5}$ exceedances can reach 15 days in regions affected by fires, particularly 5 days in GR0039A, 12 days in the fire plume and 6 days further downwind, based on our ensemble of sensitivity simulations.

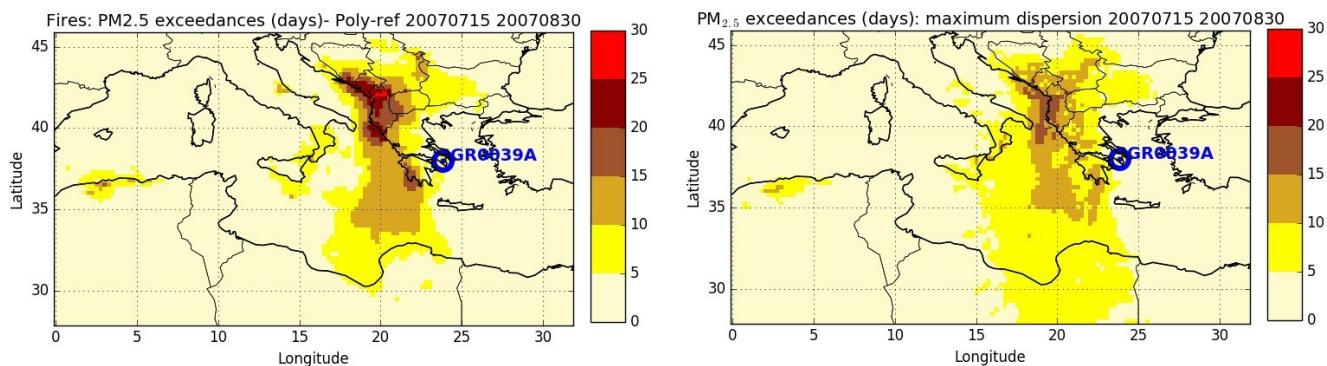


Figure 4.3. The additional days with PM_{2.5} exceedance (days) from exceedances due to fires simulated by Polyphemus (difference between the *Poly-ref* and *Poly-Nofires*) (left panel) and the maximum statistical dispersion (σ) related to fire emissions for additional days of PM_{2.5} exceedances days due to fires (right panel) during the summer 2007 (from 15 July to 30 August 2007).

5 Conclusions

A sensitivity analysis was undertaken to estimate the uncertainty of the parameterization of wildfire emissions into air quality models over the Euro- Mediterranean region during the summer 2007.

The ability of Polyphemus/Polair3D and CHIMERE to simulate regional surface PM_{2.5} concentrations and optical properties (particularly aerosol optical depth (AOD) was evaluated based on comparison to available measurements. The general evaluation compared to background AIRBASE (8 AIRBASE stations and 6 AERONET stations). Only two out of the 8 AIRBASE stations (GR0039A and AERONET surface observations shows good performances (with high correlation coefficients) for surface PM_{2.5} concentrations and AOD. 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 the most affected by wildfires (background suburban and rural stations with fire contribution 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. A clear improvement is noticed when fires are taken into account. The contribution of fires is large in Balkan and Eastern Europe during the first fire period (20–31 July) and Greece and Algeria during the second period (24–30 August).

Near fires, PM_{2.5} is mostly composed of organic aerosol (5247% to 8785%), with a strong contribution from I/S/L-VOCs (6246% to 8480%). The comparison between simulated AOD and satellite observations (MODIS) highlights consistent daily variability for both models in all sub-regions with high correlation coefficients ($\sim 90\%$ on average for Polyphemus, 80% for CHIMERE). Moreover, simulations including I/S-VOCs (*Poly-Ref*, *Poly-3km* and *Poly-NoPPM*) overestimate AOD during the fire events, by about 30% for *Poly-Ref* and *Poly-3km* and 10% for *Poly-NoPPM*, mainly in the two sub-regions MedReg1 and MedReg2. The overestimation is very low (10) in the simulation Poly-NoPPM, where I/S-VOCs are taken into account, but Since unidentified primary particles (PPM_{fine}) emitted from biomass burning are not considered in Poly-NoPPM, the simulated AOD values are closer to MODIS observations. This suggests that these primary particles which are not identified

in the emission inventory, PPM_{fire} could correspond to I/S-VOCs. During the first fire event, compared to AIRBASE measurements (at GR0039A in Greece, sub-region MedReg2), the temporal simulated surface $\text{PM}_{2.5}$ concentrations are well reproduced by the models. The simulation *Poly-Ref* reproduces well the $\text{PM}_{2.5}$ peaks with a high correlation coefficient ($>88\%$) and a low bias. The $\text{PM}_{2.5}$ peaks are slightly underestimated by all the other simulations. Compared to AERONET ground measurements, the measured AOD tends to be underestimated by the models at Bucharest (Romania, sub-region MedReg1). During the second fire event, surface $\text{PM}_{2.5}$ concentrations are well reproduced by the simulations *Poly-3km* and *CHIMERE-ref* compared to AIRBASE measurements, but they are underestimated by the other simulations. The modeled AOD are well simulated compared to AERONET observations at Lecce University and Blida by *CHIMERE-ref* and all the Polyphemus simulations except for *Poly-noI/S-VOCs*, which shows low AOD values at these stations. This suggests that taking into account I/S-VOCs improves the simulated AOD values and leads to more realistic ones at these stations. The analysis of the two fire events also shows strong regional impact. Taking into account the uncertainty on fire emissions ($>100\%$) and on observations, the performance of the models is considered reasonable. More surface observations in remote regions would be 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.

Two critical parameters, SOA formation from I/S-VOCs and injection heights, are considered as the two main sources of uncertainties in the calculation of wildfires' impact on surface $\text{PM}_{2.5}$ concentrations and AOD. 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 $\text{PM}_{2.5}$ concentrations are particularly sensitive to the 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.

~~The~~ The statistical dispersion of the ensemble of simulations based on different configurations of Polyphemus is used to evaluate the maximum uncertainty on surface $\text{PM}_{2.5}$ concentrations and AOD associated with these two parameters. During the summer 2007, the maximum statistical dispersion (σ) is as high as 75% for surface $\text{PM}_{2.5}$ in the Balkans and Greece and varies between 36 and 45% for AOD above fire regions. The number of daily exceedance of WHO recommendation of $25 \mu\text{g}\cdot\text{m}^{-3}$ for $\text{PM}_{2.5}$ 24-hour mean reaches 30 days for the fire region and 10 – 15 days for the fire plume over the simulated period of 46 days. The maximum statistical dispersion on this indicator is large (σ reaching 15 days close to fires), showing the need for better understanding of the transport and evolution of fire plumes in addition to fire emissions.

Over Europe, inorganics (mainly sulfate, sea salt and ammonium) contribute highly to $\text{PM}_{2.5}$ composition, when fire emissions are not considered (e.g., Fountoukis et al. 2011; Chrit et al. 2018). 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 due to wildfires is found to be low compared to the formation of organics. However, our simulation

takes into account emissions of inorganic precursors such as ammoniac (NH_3) with emission factors from Akagi et al. (2011). Several studies (R'honi et al. 2013; Van Damme et al. 2014; Whitburn et al. 2017), show that large emissions of NH_3 are released by biomass burning. Whitburn et al. (2017) studied the enhancement ratios NH_3/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 Whitburn et al. (2017), the emission ratios NH_3/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 NH_3 and concentrations of inorganics should be evaluated close to fire regions.

In this study, PPM_{fine} emissions correspond to emissions factors of $\text{PM}_{2.5}$ minus primary aerosols emissions of OM, BC and sulfate. This model species is used to fill a gap in current evaluation of emissions and is therefore very uncertain. This study shows that considering PPM_{fine} emissions and I/S/L-VOC emissions may be redundant and may correspond to the same “missing emissions mass”. Several models actually partly treat this missing part by considering I/S/L-VOCs emissions from particulate matter 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 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. (2012) use a factor of 3.4 for PM emissions based on the comparison between simulations and AOD observations. 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. 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 $\text{PM}_{2.5}$ concentrations.

It is worth to mention that uncertainties in wildfires modeling can be associated with other uncertainties such as meteorological inputs. Garcia-Menendez et al. (2014) found that simulated $\text{PM}_{2.5}$ concentrations at urban sites displayed large sensitivities to wind perturbations within the error range of meteorological inputs. Rea et al. (2016) added that special attention must be paid to the PBL height, which can have a considerable impact on the fire emissions injection heights. Therefore an assessment of uncertainties related to meteorological data should be investigated.

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project. We are grateful to the EEA for maintaining and providing the AirBase database (more particularly for the GR0039A station). We thank the principal investigators and their staff for establishing and maintaining the AERONET sites used in this study: Brent Holben for Blida, Maria Rita Perrone for Lecce University and Doina Nicolae for Bucharest Inoe. We finally wish to thank the National Aeronautics and Space Agency (NASA) for the availability of the MODIS data.

Appendix A

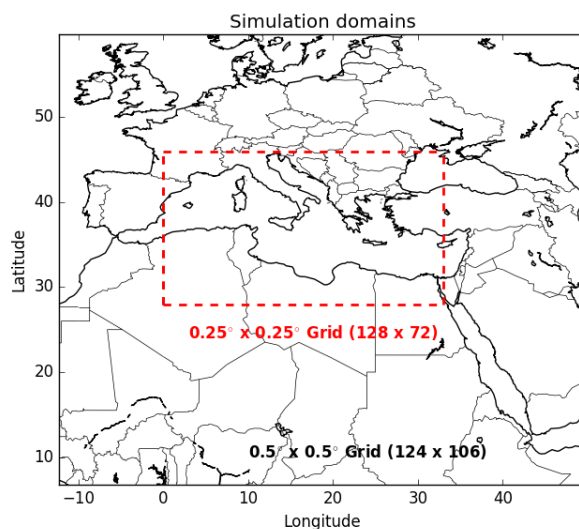


Figure A1. Simulation domains including one large domain (with a $0.5^\circ \times 0.5^\circ$ horizontal resolution) and a smaller domain (at a $0.25^\circ \times 0.25^\circ$ horizontal resolution) delimited by the dotted red box.

Appendix B

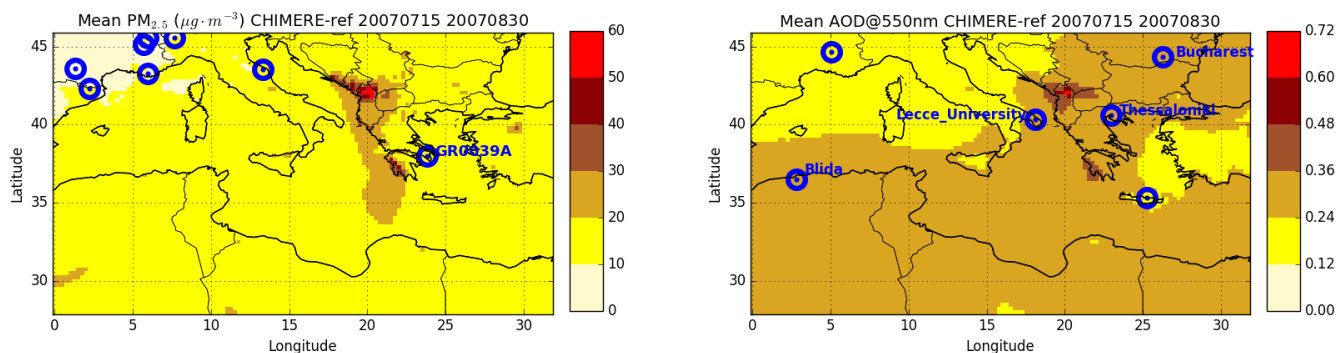


Figure B1. Daily mean surface $PM_{2.5}$ and AOD at 550 nm from the *CHIMERE-ref* simulation averaged over the summer of 2007 (the 8 AIRBASE and 6 AERONET stations, used in this work, are represented here in blue dots).

Appendix C

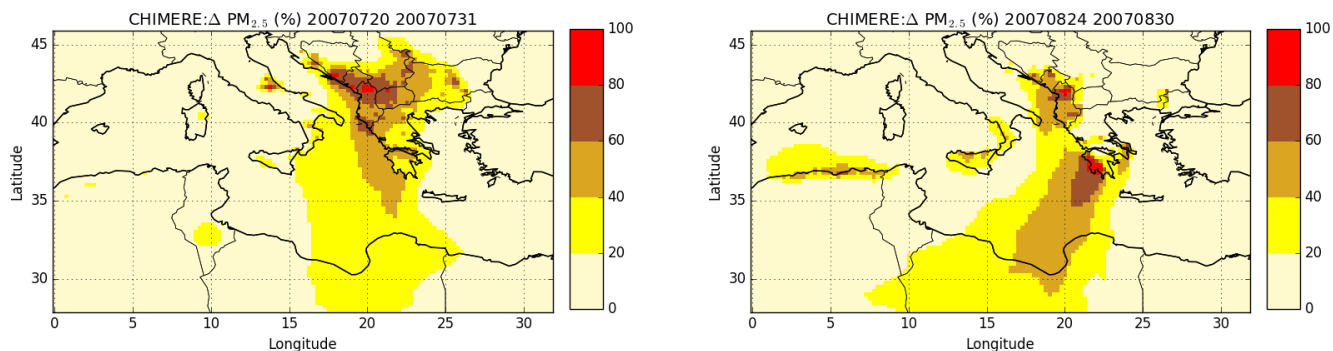


Figure C1. Left panel: relative difference of surface $PM_{2.5}$ concentrations between simulations *CHIMERE-ref* and *CHIMERE-Nofires* during the first fire event. Right panel: relative difference of surface $PM_{2.5}$ concentrations between simulations *CHIMERE-ref* and *CHIMERE-Nofires* during the second fire event.

Appendix D

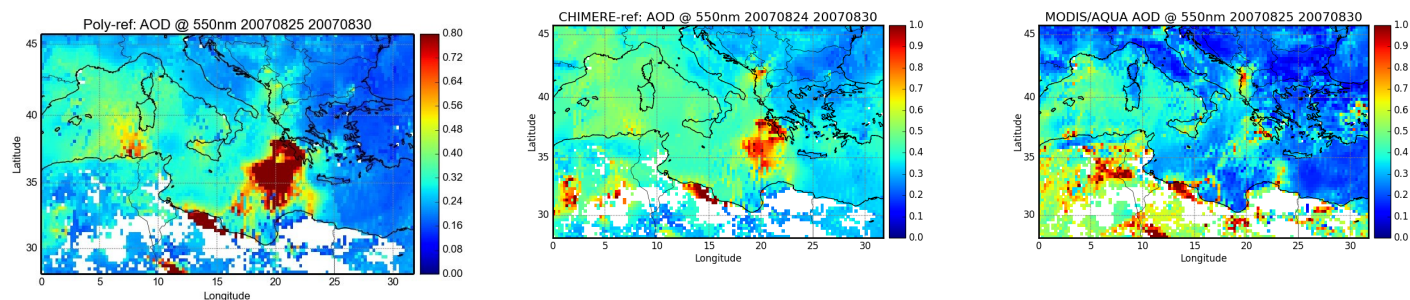


Figure D1. Mean total AOD (at 550 nm) from MODIS/AQUA, *Poly-Ref* and *CHIMERE-ref* during the second fire event (24-30 August 2007).

Appendix E

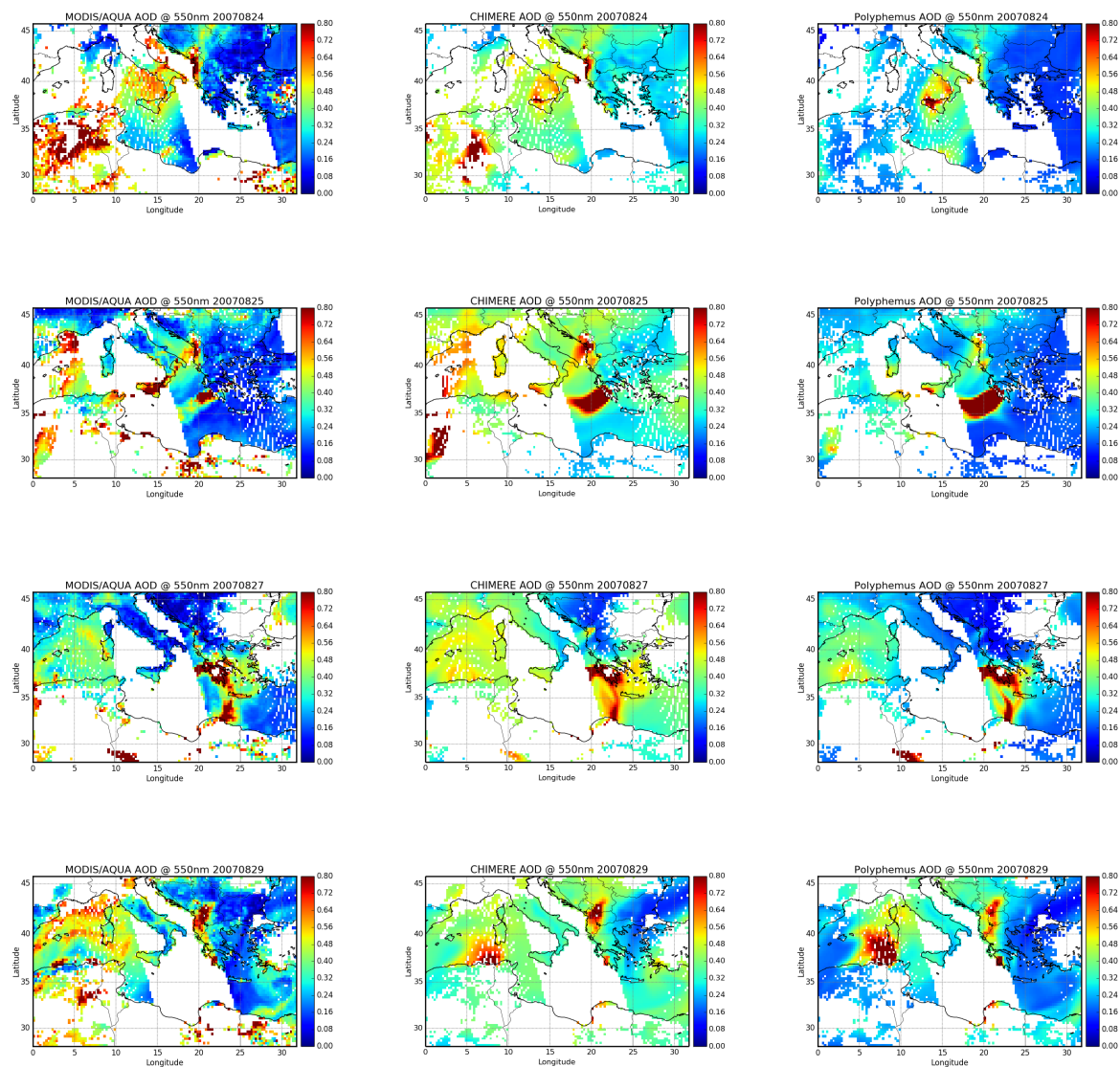


Figure E1. Total AOD (at 550nm) from MODIS/AQUA and the corresponding Polyphemus AOD (*Poly-Ref*) and CHIMERE AOD (*CHIMERE-ref*) for four selected days (24, 25, 27 and 29 August 2007).

Appendix F

Table F1. Modeled and observed PM_{2.5} exceedances at each AIRBASE stations. Values between brackets correspond to modeled PM_{2.5} exceedances for simulations without fire emissions (*Poly-Nofires* and *CHIMERE-Nofires*).

AIRBASE sta- tions	Observed PM _{2.5} exceedances			Modeled PM _{2.5} exceedances <i>Poly-Ref</i>			Modeled PM _{2.5} exceedances <i>CHIMERE-ref</i>		
	Whole pe- riod	First event	Second event	Whole pe- riod	First event	Second event	Whole pe- riod	First event	Second event
GR0039A	14	5	5	7(0)	3(0)	4(0)	9(2)	3(1)	5(0)
GR0035A	37	12	7	7(0)	3(0)	4(0)	9(2)	3(1)	5(0)
IT0459A	4	2	2	0(0)	0(0)	0(0)	0(2)	0(0)	0(0)
FR12021	0	0	0	0(0)	0(0)	0(0)	2(2)	0(0)	2(2)
ES0010R	0	0	0	1(1)	0(0)	1(1)	2(2)	0(0)	2(2)
FR15043	0	0	0	0(0)	0(0)	0(0)	4(4)	0(0)	4(4)
FR33101	0	0	0	0(0)	0(0)	0(0)	3(3)	0(0)	3(3)
FR03043	5	0	3	2(2)	0(0)	2(2)	4(4)	0(0)	4(4)
FR33102	0	0	0	0(0)	0(0)	0(0)	3(3)	0(0)	3(3)

Appendix G

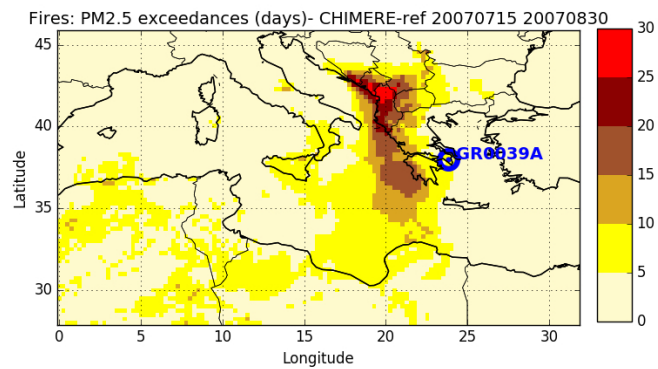


Figure G1. The additional days with PM_{2.5} exceedances due to fires simulated by CHIMERE (difference between the *CHIMERE-ref* and *CHIMERE-Nofires*) during the summer 2007 (from 15 July to 30 August 2007).

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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 #1

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

The main general comment for the manuscript is that several of the results presented can be considered as tentative since they are provided for and refer to a single site (GR0039A).

We agree that the lack of available surface data close to the fire region is a limitation to our model evaluations. That is why an evaluation is also done using satellite observations.

A rural background AIRBASE station is located close to the fire region and discussed in the specific comments (comment 5). The station GR0035A is located in the same area (Athens) in Greece as the station GR0039A, already included. For GR0035A, observations show significantly higher background PM_{2.5} levels with large variability due to local influences. We consider that this specific station is probably strongly influenced by local emissions and decided not to add additional figures that may be confusing (the model resolution cannot resolve local influences). However, following to reviewer's comment, we added it in the revised version since it also shows the high spatial variability of the transported fire plumes.

In addition to observations of surface concentrations, the remote sensing observations from MODIS and AERONET are considered. Even if comparison to remote sensing adds another level of uncertainty (calculation of optical properties, vertically integrated datasets), it improves the representativity of the model validation.

A sentence on the problem of data availability for the evaluation of the composition and impact of fire plumes has been added to the conclusions (page 24, lines 4-11 in the revised paper): “The ability of Polyphemus/Polair3D and CHIMERE to simulate regional surface PM_{2.5} 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 the most affected by wildfires (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.”

In addition, in the manuscript more results are presented for the first summer period wildfire event (20-31 July 2007) although the maximum of emissions is during the second summer period wildfire event (24-30 August 2007) as shown in Figure 2.

More Figures and results are added in the appendix of the revised paper for the second wildfire event (24-30 August 2007). Figures of the PM_{2.5} composition in MedReg2 (most affected during that time period) are added for Polyphemus simulations (Cf. Specific comment 6). The fire contribution to PM_{2.5} composition is almost the same for both events. Figures of the mean total AOD at 550nm from MODIS, for Poly-ref and CHIMERE-ref during the second fire event also shows similar conclusions as during the first fire event.

Specific comments

1. **Page 6, lines 20-22:** Instead of primary aerosol oxidation, are you probably referring to the oxidation of I/S/L-VOCs by OH in the gas phase?

The sentence (page 6, lines 20-22): “Each primary aerosol undergoes one OH-oxidation reaction in the gas phase with ...” is replaced by “Each primary I/S/L-VOC undergoes one OH oxidation reaction in the gas phase with ...”

2. **Page 8, lines 8-9:** Please use a reference and explain why POA emissions taken into account are modeled as LVOCs.

In page 8, POA emissions are modeled as LVOC in a sensitivity study. This aims at studying the impact of ignoring I/S-VOC emissions in the gas phase and ignoring the semi-volatile properties of POA. The sentence (page 8, lines 8-9) : “fire emissions in the PBL but without I/S-VOCs” is modified to “fire emissions in the PBL but without I/S-VOC emissions in the gas phase. In this sensitivity study, the semi-volatile properties of POA are not considered, and POA emissions are modeled as LVOCs”.

3. **Pages 11-12, Section 3.3:** The overall performance of CHIMERE model is shown to be slightly better than Polyphemus model considering also the statistics in Tables 3 and 4. Under this view, you have to present results of CHIMERE model in Figures 3, 4 and 11 additional to those of Polyphemus model for comparison.

It is not easy to determine which model is better in terms of statistics, as the performance varies with the statistics considered. For PM_{2.5} statistics, the overall performance of CHIMERE is slightly better for bias, but it is slightly worse for correlation. In terms of errors, CHIMERE is slightly better for PM_{2.5}, but Polyphemus is slightly better for AERONET AOD.

However, figures for CHIMERE (Figure A.1, Figure A.2, Figure A.3) are added in the appendix A, C and G respectively, in the revised paper. Conclusions for CHIMERE are similar to those for Polyphemus.

Figure A.1 is added to the appendix A

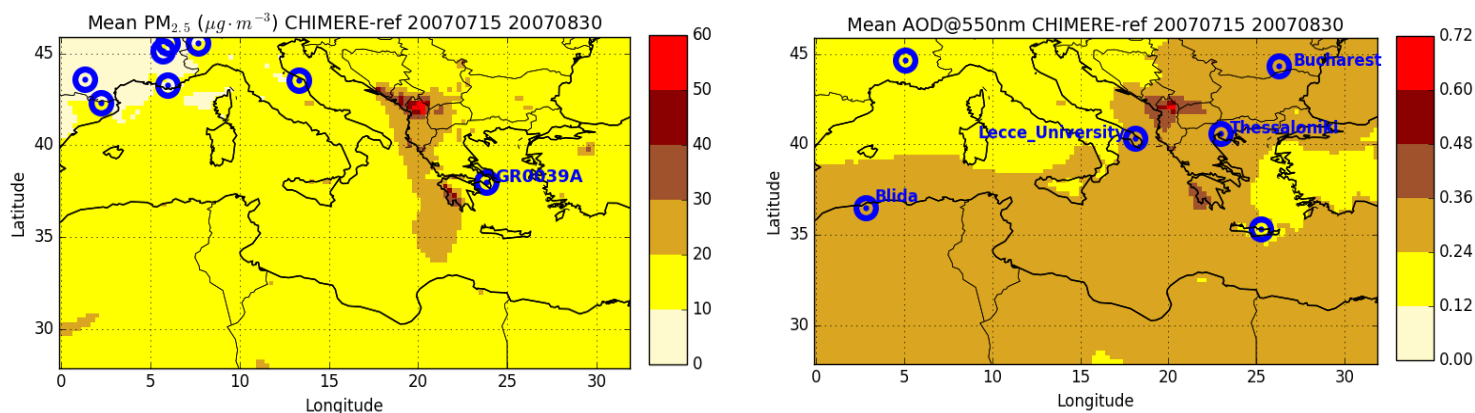


Figure A.1: “Daily mean PM2.5 and AOD at 550 nm from the simulation CHIMERE-ref averaged over the summer of 2007. The 8 AIRBASE and 6 AERONET stations, used in this work, are represented here in blue dots.

The sentence “Figure B1 in Appendix B shows that conclusions for CHIMERE are similar to those for Polyphemus.” is added to the revised paper (page 11, line 26).

Figure A.2 is added to the appendix C:

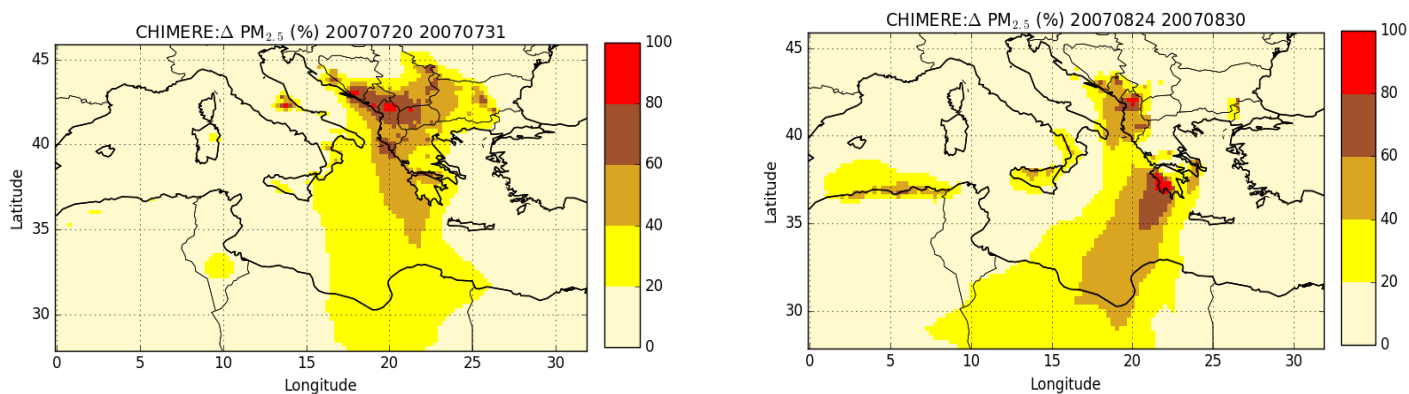


Figure A.2: “Contribution from fires to the total surface PM2.5 as the difference between reference simulations and simulations without fires for CHIMERE during the first (left panel) and second (right panel) fire events.”

The sentence “Figure C1 in Appendix C shows that the contribution of fires for CHIMERE are similar to that for Polyphemus.” is added to the revised paper (page 13, line 5).

Figure A.3 is added to the appendix G:

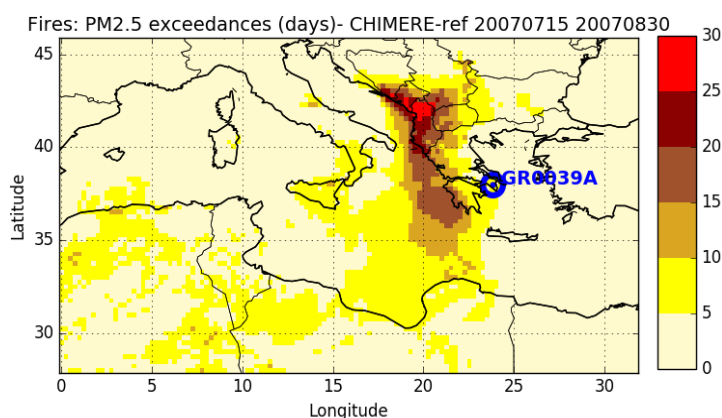


Figure A.3: PM2.5 exceedance (days) from fires simulated by CHIMERE (difference between the Poly-ref and Poly-Nofires) during the summer 2007 (from 15 July to 30 August 2007).”

The sentence “Figure G1 in Appendix G shows that conclusions for CHIMERE are similar to those for Polyphemus.” is added to the revised paper (page 25, lines 22-23).

4. Pages 12, Figure 3: Presenting similar maps using the data of MODIS is necessary to support better the comments in the manuscript about Figure 3.

Comparisons to MODIS are addressed in the response to the specific comment 8.

5. Page 13, Section 3.4.1: Figure 4 reveals several parts of the Mediterranean area being affected by the wildfire plumes for example Central and South Italy, Sicily, Bulgaria, almost the whole country of Greece etc. In the manuscript you are referring to only 2 stations being affected by the wildfire emissions during the first event and 3 stations during the second one. Aren’t there additional AIRBASE or AERONET stations that could be used to support your analysis? (For example GR0035A is also a background suburban station where PM2.5 concentrations are being measured; is there a reason for not using measured data from this station?). Is the performance of the model good also for the remaining areas that are not affected by the fires?

5.1) aren’t there additional AIRBASE or AERONET stations that could be used to support your analysis?

Measurement stations have been selected using the contribution from fires. Only stations with relative contribution higher than 10% are kept. In addition, for surface concentration observations, only background suburban and background rural stations with available data during the fire events are chosen in this study (Figure 3 left panel in the paper).

In the revised paper page 15 line 2, the sentences: “Figure 6 shows time series of daily observed and simulated aerosols at stations with significant impact from fires ...” are replaced by: “ Figure 3.5

shows times series of daily observed and simulated aerosols at background suburban and background rural stations with available data during fire events and with a fire contribution higher than 10% ...”

As already explained in the answer to the general comments, another station in Greece (GR0035A) meets this criteria, close to the station already included and discussed in detail.

In the revised paper, we added in Figure 2.5 the time series of daily mean surface PM_{2.5} at that station (GR0035A). Although the temporal tendencies of the simulated PM_{2.5} concentrations at GR0035A are consistent with the observations, background PM_{2.5} levels are strongly underestimated (Bias= -85%). The observed background levels are significantly higher than those at the GR0039A station. This suggests the influence of local emissions that are missing in the model or that the resolution of the simulations does not allow to represent. This was the reason why the GR0035A was not initially considered in the paper.

However, following the reviewer's suggestion to include more surface sites, we have chosen to include it in the revised version because it is a good illustration of the high spatial variability of the transported fire plume which can not be captured in regional CTM simulations.

At the GR0035A station, the second observed peak during the second fire event is more than twice higher than the observed peak at the GR0039A and is measured one day earlier. In the simulations, the enhancement due to fires is similar in shape and magnitude, clearly highlighting the difficulty to simulate the exact temporal variability of emissions and transport of fire plumes in CTMs. Moreover, the station GR0035A is probably affected by a dust episode during the second fire event. Similar conclusions are found for AOD at the station Thessaloniki (near the station GR0035A), which seems to be also affected by dust especially during the second fire event. The observed value of Angstrom exponent at Thessaloniki station is lower on the 25th August ($\alpha=0.95$) suggesting that a large fraction of coarse mode particles (probably dust transport). Here, we didn't include the Thessaloniki station due to the lack of available AOD measurements during the first fire event where the fire contribution is more pronounced.

The sentences are added to page 17 line 11 in the revised paper: “At the GR0035A station, the temporal tendencies of the simulated PM_{2.5} concentrations are consistent with the observations. However the first PM_{2.5} peak is underestimated since PM_{2.5} levels are strongly underestimated (bias= -85%). The observed background levels are significantly higher than those at GR0035A station. This suggests the influence of local emission that are missing in the model or limitations due to the coarse model resolutions which can not represent this station.”

and we added also these sentences to page 17 line 26 in the revised paper: “At the GR0035A station, the second observed peak during the second fire event is more than twice higher than the observed peak at the GR0039A and is measured one day earlier. In the simulations, the enhancement due to fires is similar in shape and magnitude, clearly highlighting the difficulty to simulate the exact temporal variability of emissions and transport of fire plumes in CTMs. Moreover, the station GR0035A is probably affected by a dust episode during the second fire event.”

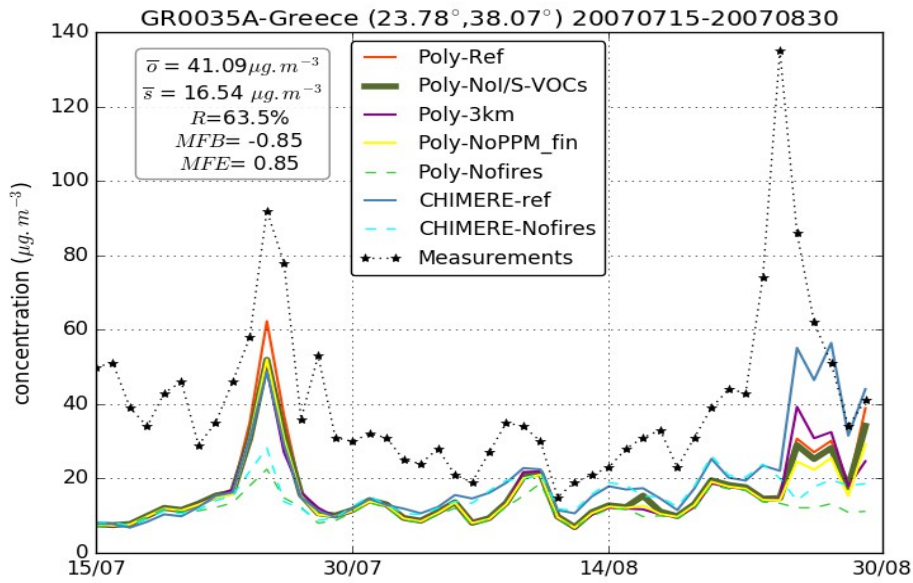


Figure 6: Times series from 15 July to 30 August of daily mean surface PM2.5 concentrations at the AIRBASE station GR0035A.

5.2) Performance of the models (CHIMERE and Polyphemus) for areas which are not affected by fires:

Table C.1: Statistics of models-to-measurements comparisons for the mean daily PM2.5 from the reference Polyphemus simulation (from 15 July to 30 August 2007).

Station	localization	Mean observed PM2.5	Mean simulated PM2.5	Correlation (%)	MFB (%)	MFE(%)
FR12021	Toulouse- France	10.48	6.44	85.5	-48	48
ES0010R	Spain	9.54	7.95	59.4	-22	36
FR15043	Grenoble- France	10.57	8.28	75.8	-26	31
FR33101	Chambery- France	6.84	8.56	87.3	22	28
FR03043	Marseille- France	14.93	10.16	92	-43	43
FR33102	Chambery Le haut- France	9.74	8.37	86	-17	23
IT0495A	Italy	17.39	8.19	83.4	-76	76

Table C.2: Statistics of models-to-measurements comparisons for the mean daily PM2.5 from the reference CHIMERE simulation (from 15 July to 30 August 2007).

Station	localization	Mean observed PM2.5	Mean simulated PM2.5	Correlation (%)	MFB (%)	MFE(%)
FR12021	Toulouse- France	10.48	8.80	76.6	-22	32
ES0010R	Spain	9.54	8.41	56.1	-13	30
FR15043	Grenoble- France	10.57	10.44	73.9	-3	32
FR33101	Chambery- France	6.84	10.47	67.6	40	44
FR03043	Marseille- France	14.93	10.71	85	-39	41
FR33102	Chambery Le haut- France	9.74	10.19	61.3	1	35
IT0495A	Italy	17.39	10.78	67	-49	51

Regarding the statistical evaluation of the models, the mean simulated PM2.5 at the different stations which are not affected by fires is in good agreement with the observations, except for IT0495A where PM2.5 are underestimated compared with surface PM2.5 measurements. The model performance is always met and the model goal is met only for the model errors MFEs. The models-to-measurements correlations range between 59.9 and 92% for all the stations.

Generally, the performance of the two models (Polyphemus and CHIMERE) is good for the areas which are not affected by fires, except at IT0495A where PM2.5 background levels in the simulations are underestimated most probably due to dust.

Since this paper focuses on the evaluation of models performance near the most affected stations by fires, we choose not to include the model performance at the stations which are not affected by fires in the revised paper.

6. Page 14-15, Section 3.4.2: The results presented in this section refer only to a single station and only to the fire event of the first period. Under this view, they are of very limited reference and cannot be used to characterize the aerosol composition in the Mediterranean areas affected by the fire plumes. Similar results with those presented in Figure 5 have to be presented for different stations and wider areas affected by the fire emissions and separately for both fire peak events in order possible differences to be identified.

Figure 5 page 15 is replaced by Figure 3.3 and Figure 3.4 in the revised paper.

Figure 3.3 and Figure 3.4 show the composition of surface PM2.5 concentrations for two wider areas which are the subregions: MedReg1 (for the first fire event) and MedReg2 (for the second fire event). These two subregions are the areas most affected by fires (high fire emissions (Figure 2)) especially during the first fire event for Medreg1 and during the second fire event for MedReg2.

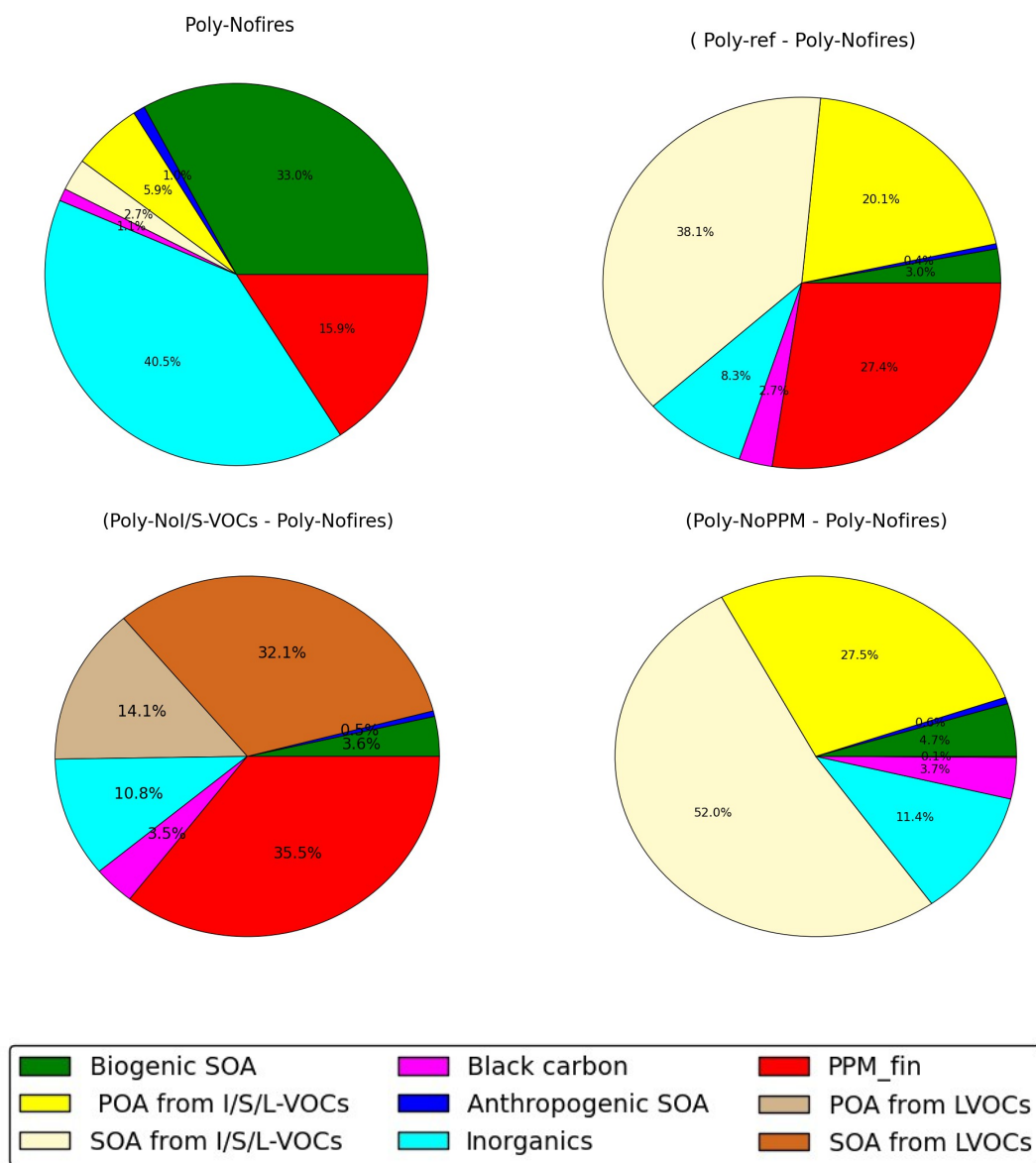


Figure 3.3: Composition of surface PM_{2.5} concentration over MedReg1 during the first fire event (upper left panel, simulation Poly-Nofires). Composition of surface PM_{2.5} concentration due to fires (upper right panel: simulation Poly-ref; lower left panel: simulation Poly-NoI/S-VOCs; lower right panel: simulation Poly-NoPPM).

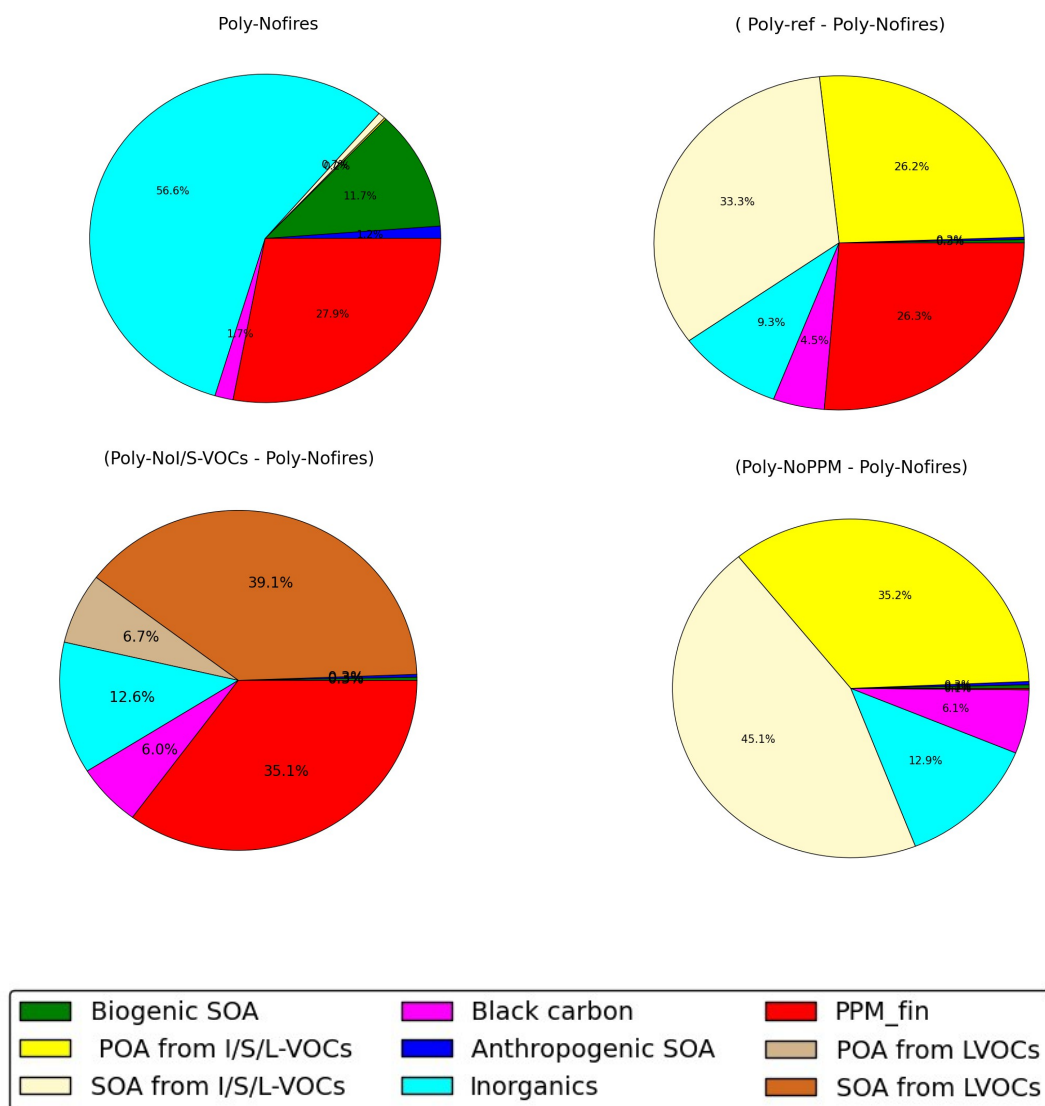


Figure 3.4: Composition of surface PM_{2.5} concentration over MedReg2 during the second fire event (upper left panel, simulation Poly-Nofires). Composition of surface PM_{2.5} concentration due to fires (upper right panel: simulation Poly-ref; lower left panel: simulation Poly-NoI/S/L-VOCs; lower right panel: simulation Poly-NoPPM).

Similar composition is found for surface PM_{2.5} concentrations over two wider areas: MedReg2 during the second fire event and over MedReg1 during the first fire event for both models (CHIMERE and Polyphemus).

During the first fire event over MedReg1, organic aerosols contribute significantly to surface PM_{2.5} concentrations. Their contribution is larger than those of other species such as inorganics (11%) PPMfine (dust) (15%) and black carbon (5%). When we consider fire emissions, we found that organic aerosols which are mostly composed of POA and SOA from I/S/L-VOCs, are predominant in the contribution of fires (between 47% and 85% of the contribution). Lower contribution are found for inorganics (8% to 13%) and black carbon (3% to 6%).

During the second fire event over MedReg2, PM_{2.5} composition in Poly-Nofire is similar to PM_{2.5} composition during the first fire event over MedReg1. The same conclusions are found as during the first fire event: the organic aerosols are the most important contributor in the composition of PM_{2.5} from fires (46% to 81%). Lower contributions are found for inorganics (9 to 12%) and black carbon (5 to 6%).

The sentences (page 14 line 2-23) are corrected as followed: “The composition of surface PM2.5 concentrations simulated during the first fire event over MedReg1 and during the second one over MedReg2 are shown in Figures 3.3 and 3.4 respectively. These two subregions are the areas most affected by fires (high fire emissions (Figure 2)) especially during the first fire event for Medreg1 and during the second fire event for MedReg2.

The upper left panel shows the composition of surface PM2.5 concentrations for the simulation without fire Poly-NoFires (background surface PM2.5 concentrations), while the upper right panel shows the composition of surface PM2.5 concentrations due to fires (differences between the simulations Poly-ref and Poly-Nofires). If wildfires are not taken into account (simulation without fire) and during the first fire event over MedReg1, organic and inorganic aerosols contribute equally (42.6%, 40.5%) to the surface PM2.5 concentrations. The contribution of PPMfine (dust), black carbon are lower (15% , 1%). As noted by Chrit et al. (2017), most of summer organic aerosols are from biogenic sources in this region.

If wildfires are not taken into account during the second fire event over MedReg2, inorganics and PPMfine are the predominant component in the composition of PM2.5 concentrations (56.5% and 27.9%). Lower contributions are simulated for black carbon (1.2%) and organic aerosol (14.3%).

Figures 3.3 and 3.4 also show the composition of surface PM2.5 concentrations due to fires for the simulations Poly-ref (differences between the simulations Poly-ref and Poly-Nofires), Poly-NoI/S-VOCs (differences between the simulations Poly-NoI/S-VOCs and Poly-Nofires) and Poly-NoPPM (differences between the simulations Poly-NoPPM and Poly-Nofires). During the first fire event over MedReg1, organic aerosol is predominant in the contribution of fires (between 47% and 85% of the contribution). Organic aerosol is mostly composed of POA and SOA from I/S/L-VOCs (46% to 80%). Note that POA and SOA from L-VOCs (low volatile organic compounds) are important even in the simulation when I/S-VOCs are not taken into account in fire emissions (46%), because POA are then assigned to L-VOCs. The contribution from inorganics (8% to 13%) and black carbon (3% to 6%) are low. During the second fire event over MedReg2, similar PM2.5 composition is found. Organic aerosol (mainly the POA and SOA from I/S/L-VOCs) is the most important component contributing to PM2.5 from fires (between 46% and 81% of the contribution). The contribution from inorganics (9% to 12%) and black carbon (5 to 6%) are lower.

Similar PM2.5 composition is found not only during the second fire event over MedReg2 but also in the concentrations simulated by CHIMERE (not shown here). Surface PM2.5 concentrations from fire simulated by CHIMERE are composed in the first and second fire events mainly of organic aerosol, mostly composed of primary organic carbon (OCAR) (46%) which corresponds to I/S/L-VOCs in Polyphemus, and of PPMfine (39%). The contributions from inorganics (9%), black carbon (5.2%) and SOA from VOCs (2.7%) are low.”

7. Page 16, lines 4-8: Comments for Figure 8 is better to be written at the end of the section 3.4.3 after the comments for Figures 6 and 7.

Comments for Figure 8 are moved at the end of the section 3.4.3 (page 18 line 12 in the revised paper).

8. Page 16, Figure 8: Please present and comment on similar maps based on the models and MODIS results for the second fire event in August.

Figure D.1 is added to Appendix D and E.1 are added to Appendix E in the revised paper.

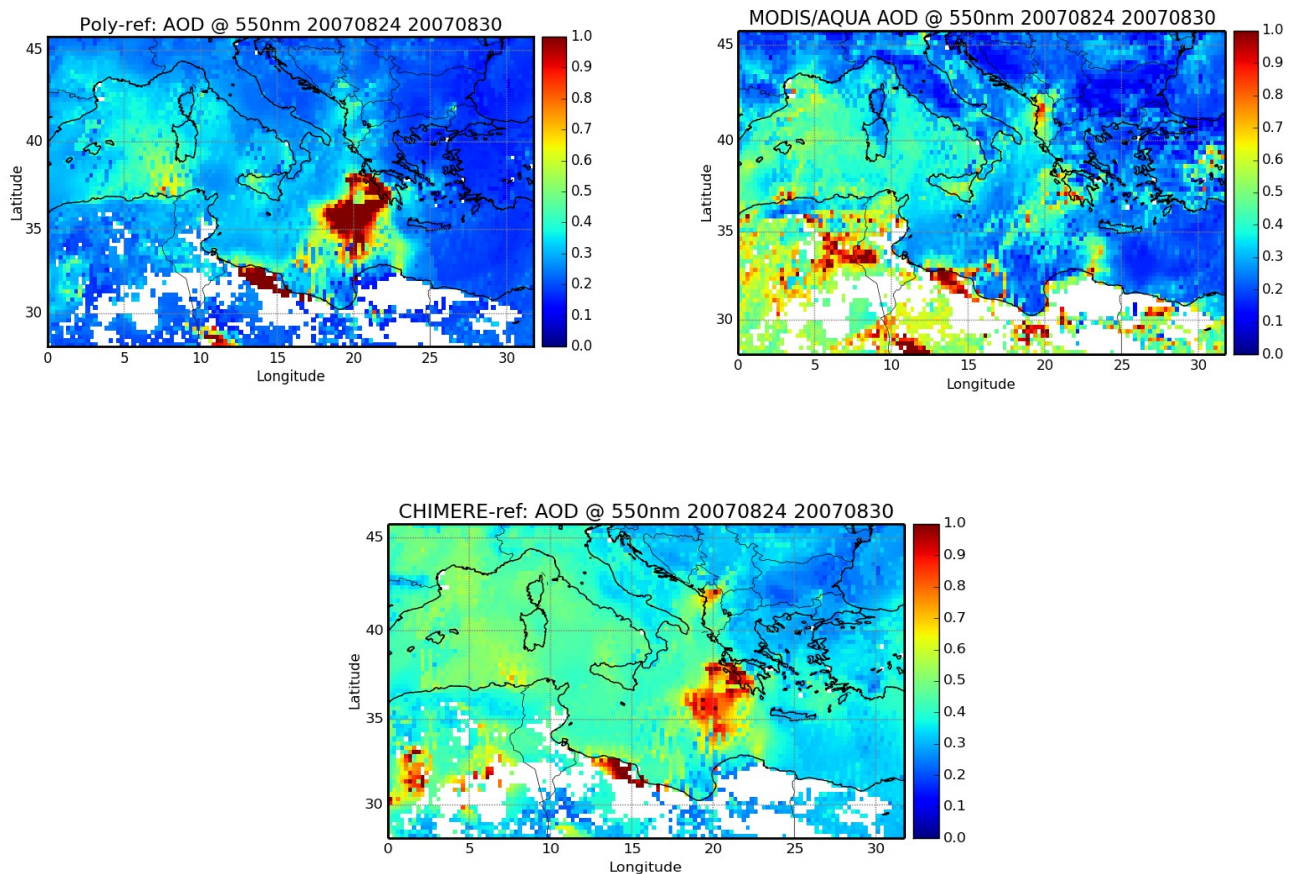


Figure D.1: “Mean total AOD (at 550 nm) from MODIS/AQUA, Poly-Ref and CHIMERE-ref during the second fire event (24-30 August 2007).”

These sentences are added in the revised paper (page 18 line 16) (after moving the comments for Figure 8 at the end of the section 3.43 page 18 line 12): “ During the second fire event in Greece, the simulated AOD for Poly-ref and for CHIMERE-ref is about 1 and 0.9 respectively as shown in Figure D.1 in Appendix D. The observed AOD can reach 0.9. However, both models overestimate AOD values in the fire plume (reaching ~0.98 for Poly-ref and 0.88 for CHIMERE-ref against 0.7-0.8 in Greece and 0.5 from MODIS. The fire plume is less pronounced in observations than in simulations.

Day-to-day comparisons for four selected days (24, 25, 27 and 29 August 2007) are shown in Figure E.1 in Appendix E. The simulated AOD is consistent with the observations in terms of localization and general transport pathways. However, the simulated AOD is much higher in the Greek fires' plume compared with MODIS observations during the peak of emissions (25-29 August). This probably reflects too low temporal variability in the emissions. In the simulations, emissions are assumed constant during the day but comparisons suggest shorter temporal variability. This is also apparent in the time series of Figure 3.6 in the paper, over region MedReg2: the peak for the second fire event is twice longer in the simulations (double peak starting on the 25 August 2007) than in the observations. This peak over two days instead of one in the simulations suggests an overestimate of emissions during this event which is also observed with respect to surface observations in Greece. The first peak corresponds better to observations. This shows that uncertainties are not only related to total emissions but also to their temporal variability and the associated transport pathways.”

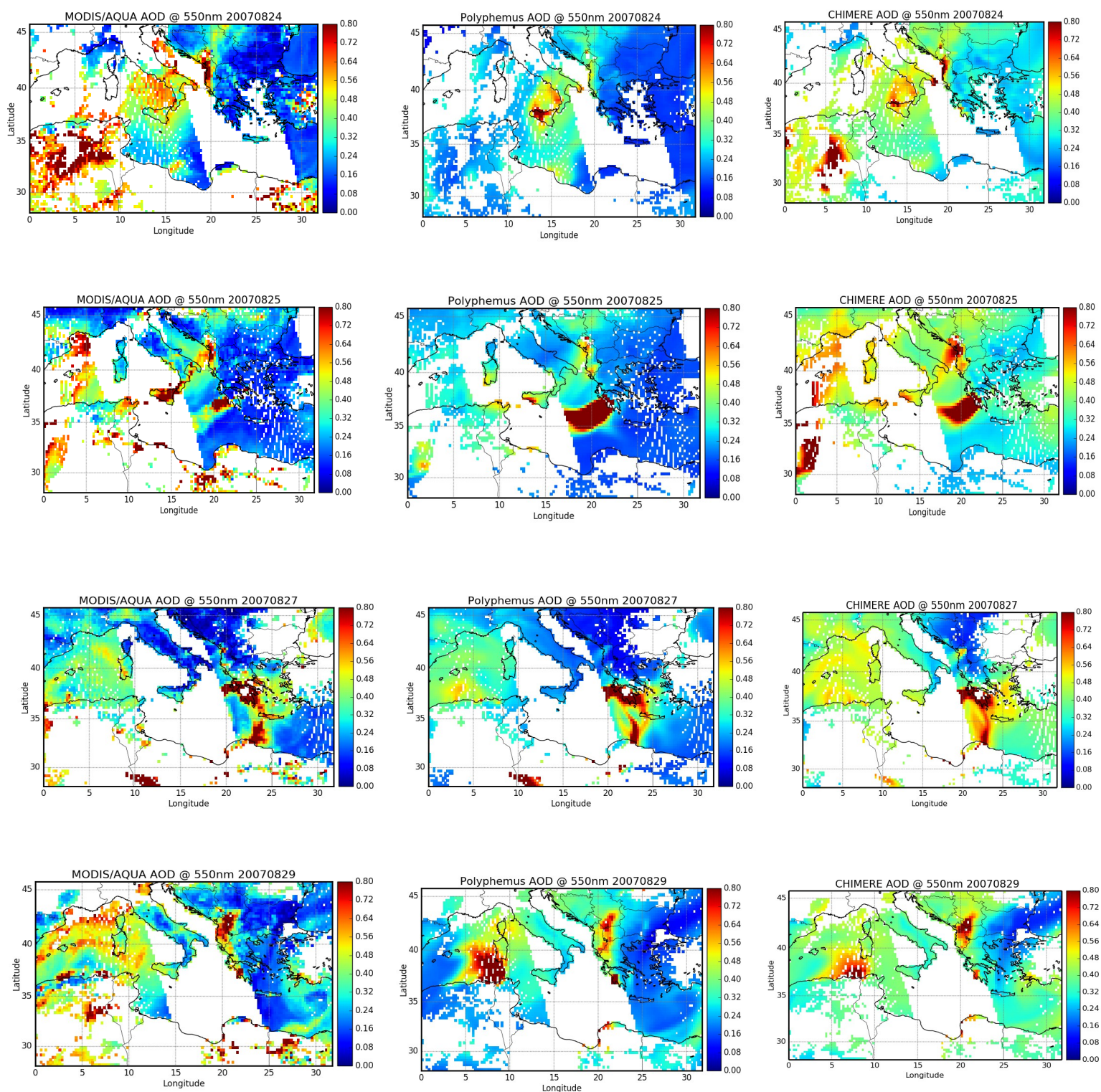


Figure E.1: “Total AOD (at 550 nm) from MODIS/AQUA and the corresponding Polyphemus AOD (Poly-Ref) and CHIMERE AOD (CHIMERE-ref) for four selected days (24, 25, 27 and 29 August 2007).”

9. Page 23, Section 4.1: In lines 10-12, it is commented that according to the Polyphemus model results the number of days, when the WHO air quality limit for the daily PM2.5 values is exceeded in GR0039A station, is 7 while the corresponding number based on measurements is 14. This corresponds to a -50% bias by the model. Is it acceptable? No data are provided for additional stations in the study area rendering the results presenting in this section very tentative. Further evidence is necessary to prove the acceptable performance of Poly-ref runs (and CHIMERE-ref runs, please see na previous comment) in the estimation of the PM2.5 air quality exceedences supporting better the necessity for the maps in the Figure 11 to be presented.

Table F.1: Modeled and observed PM2.5 exceedances at each AIRBASE stations. Values between brackets correspond to modeled PM2.5 exceedances for simulations without fire emissions (Poly-Nofires and CHIMERE-Nofires).

Airbase station	Observed PM2.5 exceedances			Modeled PM2.5 exceedances Poly-ref			Modeled PM2.5 exceedances CHIMERE-ref		
	Whole period	First fire event	Second fire event	Whole period	First fire event	Second fire event	Whole period	First fire event	Second fire event
GR0039A	14	5	5	7 (0)	3 (0)	4 (0)	9 (2)	3 (1)	5 (0)
GR0035A	37	12	7	7 (0)	3 (0)	4 (0)	9 (2)	3 (1)	5 (0)
IT0459A	4	2	2	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)
FR12021	0	0	0	0 (0)	0 (0)	0 (0)	2 (2)	0 (0)	2 (2)
ES0010R	0	0	0	1 (1)	0 (0)	1 (1)	2 (2)	0 (0)	2 (2)
FR15043	0	0	0	0 (0)	0 (0)	0 (0)	4 (4)	0 (0)	4 (4)
FR33101	0	0	0	0 (0)	0 (0)	0 (0)	3 (3)	0 (0)	3 (3)
FR03043	5	0	3	2 (2)	0 (0)	2 (2)	4 (4)	0 (0)	4 (4)
FR33102	0	0	0	0 (0)	0 (0)	0 (0)	3 (3)	0 (0)	3 (3)

Table F.1 is added to the revised paper in the appendix F.

The sentences in page 23, lines 10-12: “The number of exceedances predicted by the model is first compared to exceedances observed by AIRBASE (only one station is available). During the summer 2007, 14 PM2.5 exceedances were observed and only 7 are simulated at GR0039A by Poly-ref and 9 by CHIMERE-ref.” are replaced by “Table F1 in Appendix F presents the modeled and observed PM2.5 exceedances at each AIRBASE stations during the whole summer 2007 and during fire events. Generally, the models (Polyphemus and CHIMERE) underestimates PM2.5 air quality exceedances because the horizontal resolution used here does not allow the representation of local pollution (especially for the station GR0035A). Better performance is observed during fire events than the whole period. Near fire regions, at the stations GR0039A, the number of exceedance is well-modeled, in spite of the slight underestimation during both fire events compared to the observed PM2.5 exceedances. However, at the GR0035A station, the underestimation of PM2.5 exceedances is sharp. This can be explained by the strong underestimation of the background PM2.5 levels as shown in Figure 3.5. Far from fire regions, the PM2.5 exceedances modeled by Polyphemus are in a good agreement with the observed ones especially during the two fire events. The simulated PM2.5 exceedances by CHIMERE in the fire plume are overestimated at some stations .”

10. Page 23, Figure 11: Figure 11 is not clearly explained in the manuscript. What is presented in Figure 11 (left panel)? Is it the additional days with exceedences due to fires (i.e. (days with exceedences Poly-ref) minus (days with exceedences Poly-Nofires)))?

This sentence (page 23 lines 13-15): “Figure 11 shows the simulated number of days concerned by a PM2.5 exceedance from fires (difference of PM2.5 exceedance days between the Poly-ref and Poly-Nofires). Most are concentrated around fire sources, mainly in Balkan (30 days).” is replaced by “Figure 4.3 shows the additional days with PM2.5 exceedences due to fires simulated by Polyphemus (difference of PM2.5 exceedance days between the Poly-ref and Poly-Nofires). Most are concentrated around fire sources, mainly in Balkan (30 days).”

Figure 11 's caption “PM2.5 exceedance (days) from fires simulated by Polyphemus (difference between the Poly-ref and Poly-Nofires) (upper left panel) and simulated by CHIMERE (difference between the Poly-ref and Poly-Nofires) (upper right panel) and the maximum dispersion (σ) related to fire emissions for PM2.5 exceedences days (lower left panel) during the summer 2007 (from 15 July to 30 August 2007).” is replaced by “The additional days with PM2.5 exceedences due to fires simulated by Polyphemus (difference between the Poly-ref and Poly-Nofires) and the maximum dispersion (σ) related to fire emissions for additional days of PM2.5 exceedences due to fires (lower left panel) during the summer 2007 (from 15 July to 30 August 2007).

11. Page 23, lines 16-18: It is commented by the authors that PM2.5 concentrations are composed mainly of organic matter. However adding the percentages in the parentheses (i.e. 7.3% for POA and 19.1% for SOA), a percentage total contribution of 26.1% is estimated. Is this contribution to PM2.5 the main? Please clarify.

The sentence (page 23, lines 16-18): “During the fire periods, surface PM2.5 concentrations simulated in Poly-ref in MedReg1 and MedReg2 sub-regions are composed mainly of primary organic matter (7.3% with 92% due to fire) and secondary organic aerosol (19.1% with 78.9% due to fires).” is replaced by “During the fire periods, surface PM2.5 concentrations simulated in Poly-ref in MedReg1 and MedReg2 subregions are composed mainly of OM (54% with 61.6% due to fire for MedReg1 and 52.3% with 60.1% due to fires for MedReg2), and inorganics (27% with 8% due to fires for MedReg1 and 15% with 9% due to fires for MedReg2).”

P.S.: During each fire event, over each subregion, the percentage (P) presenting the contribution of each component (X) from fire, is calculated as follows : $P\% = [X]_{poly-ref} - [X]_{poly-Nofire} / [PM2.5]_{Poly-ref}$.

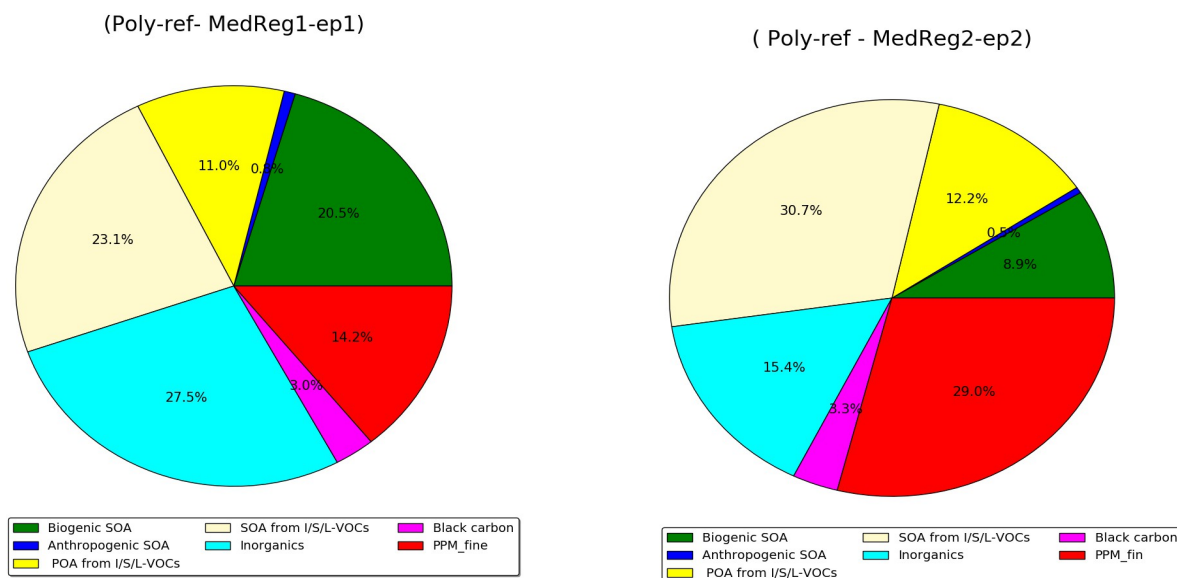


Figure: composition of surface concentrations over MedReg1 during the first fire event and MedReg2 during the second fire event for simulation Poly-Ref.

12. Page 24, lines 9-10: The percentages have been estimated for one site and for one fire event and have a very limited reference (spatial and temporal). Please see a previous comment of mine so as to update the conclusions.

We replaced the percentages given for one site and for one fire event by percentages for wider areas during the two fire events : MedReg1 during the first fire event and MedReg2 during the second fire event.

The sentence in page 24, lines 9-10: “Near fires, PM_{2.5} is mostly composed of organic aerosol (52% to 87%), with a strong contribution from I/S/L-VOCs (62% to 84%).” is replaced by “Near fires, PM_{2.5} is mostly composed of organic aerosol (47% to 85%), with a strong contribution from I/S/L-VOCs (46% to 80%).”

13. Page 24, lines 10-12: Which Figure or Table supports these high correlation values and where in the manuscript are these results commented? Is it Figure 7?

Figure 7 supports these high correlation values. These results are commented (page 15 lines 6-8).

14. Page 24, lines 14-16: This conclusion is not straightforward and requires further clarifications not only in conclusions but also in the manuscript. In addition, it is not clear how overestimations in simulated results may indicate missing emissions.

Indeed, this sentence is not clear. The sentence (page 24, lines 14-16): “The overestimation is very low (10%) in the simulation Poly-NoPPM, where I/S-VOCs are taken into account, but unidentified primary particles emitted from biomass burning are not.” is replaced in the revised paper by “Since unidentified primary particles (PPM_{fine}) emitted from biomass burning are not considered in Poly-NoPPM, the simulated AOD values are closer to MODIS observations. This suggests that PPM_{fine} could correspond to I/S-VOCs.”

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 #2

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:

A) This paper tries to evaluate model estimates of fire-related PM concentrations through evaluation with data from surface networks and satellite remote sensing. However, I think this work is flawed due to the decision to use only PM measurements. For some reason they do not make use of the rather extensive measurements which are available (Airbase, EMEP, or even AMS data) of sulfate, nitrate, ammonium, or other inorganic components. This is a serious problem I think. For example, Fountoukis et al (2011) suggested significant contributions of sulfate, nitrate and ammonium to PM1 mass, and backed up their analysis with comparison to measurements. Van Damme et al. (2014) showed enormous levels of NH3 from biomass burning, which presumably caused high PM2.5 also.

Figure 1 shows the PM2.5 composition for simulation “Poly-Nofires” over the Euro-Mediterranean region and during the summer 2007. Inorganics, mainly sulfate (23.5%), sea salt (23.6%) and ammonium (7.6%), contribute highly to PM2.5 composition. Similar results are found in Fountoukis et al. (2011) who showed also that sulfate, sea salt and ammonium contribute highly to the PM1 mass (30%, 14% and 13% respectively) over Europe during May 2008.

Several other studies evaluated the performance of Polyphemus to simulate sulfate, ammonium and nitrate concentrations (Sartelet et al., 2007; Zhang et al., 2013; Chrit et al., 2018). Chrit et al., (2018) evaluated the model Polyphemus over the western Mediterranean region during the ChArMEx campaign, by comparison to in-situ aerosol measurements performed during three consecutive summers (2012, 2013 and 2014). According to Chrit et al. (2018), sulfate and ammonium concentrations satisfy the performance and goal criteria, while sodium satisfies only the performance criteria. Chrit et al. (2018) found that chloride and nitrate were both underestimated compared to observations. They explained that this underestimation is probably caused by uncertainties in the measurements and difficulties in representing the gas-particle partitioning. Sartelet et al. (2007) showed that the concentrations of inorganics modeled with Polyphemus over Europe are consistent with observations (EMEP and AIRBASE). Model performance criteria are met for sulfate, nitrate and ammonium. Zhang et al. (2013) used Polyphemus to simulate air quality in July 2001 over western Europe. They evaluate the model's performance using surface measurements from EMEP and AIRBASE stations for inorganics. The simulated concentrations of inorganic PM are in a good agreement with EMEP observations, although sulfate and nitrate concentrations are underestimated.

The formation of inorganics because of wildfires is found to be low compared to the formation of organics. However, our simulation takes 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 NH_3/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 NH_3/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 inorganics, more observations are required to provide emissions values of NH_3 and concentrations of inorganics should be evaluated close to fire regions.

During the summer 2007, AMS data are not available. The PM data from AIRBASE and EMEP are scarce in our domain. We only have a few PM_{2.5} stations and only 3 stations (far from the studied fire regions: 2 in Spain and 1 in France) for inorganic concentrations. Evaluation with measurements is therefore not possible. However, we looked more closely at the contribution of inorganics to the simulated concentrations.

Poly-Nofires - whole summer 2007- MedReg

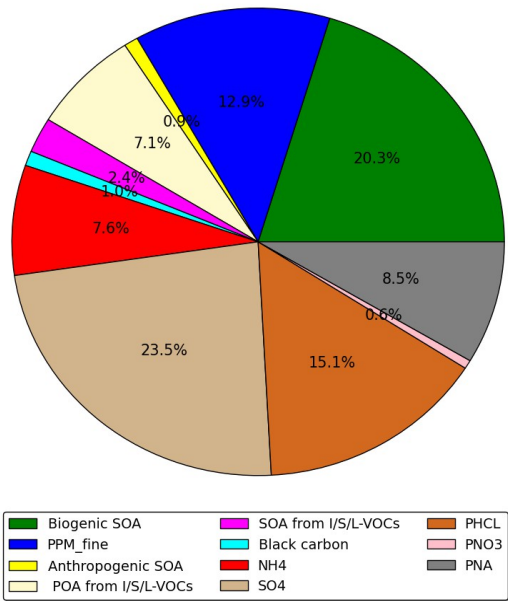


Figure 1: Composition of surface PM_{2.5} (simulation Poly-No-fires) over the Euro-Mediterranean region during summer 2007

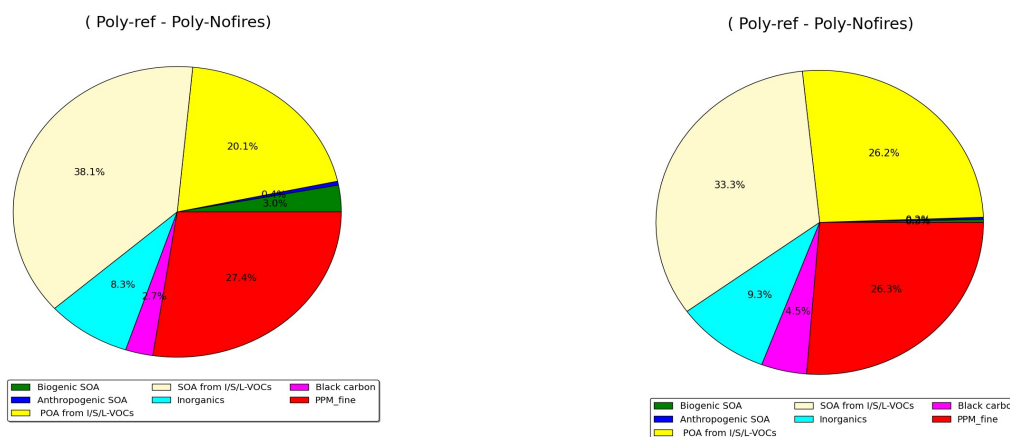


Figure 2: Composition of surface PM_{2.5} concentration due to fires (left panel: simulation Poly-ref over MedReg1 during the first fire event and right panel: simulation Poly-ref over MedReg2 during the second fire event).

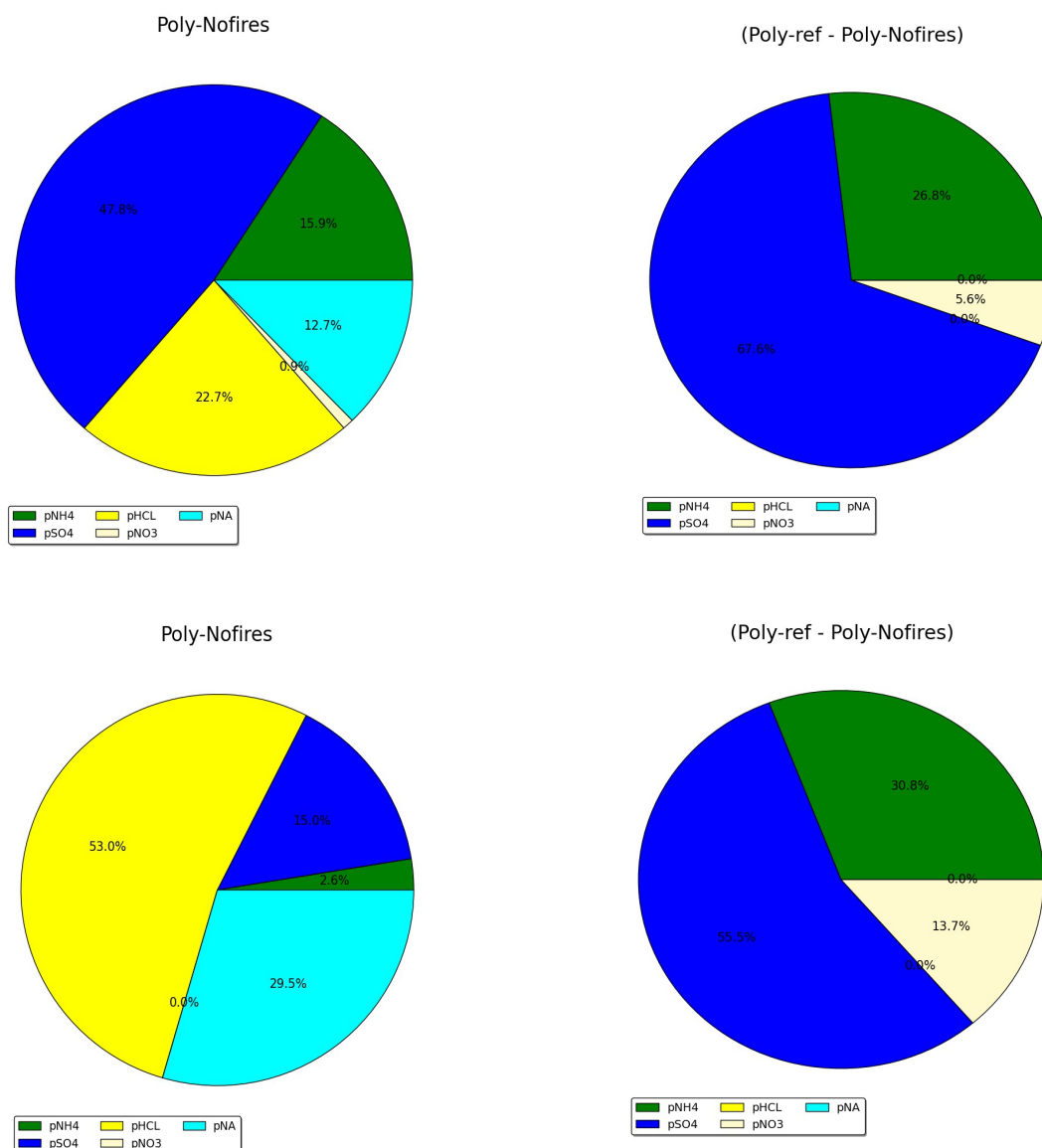


Figure 3: Composition of surface inorganic concentrations over Medreg1 during the first fire event (upper left panel, simulation Poly-nofires) and over MedReg2 during the second fire event (lower left panel, simulation Poly-nofires). Composition of surface inorganics due to fires over Medreg1 during the first fire event due to fires (upper right panel: simulation Poly-ref) and over MedReg 2 during the second fire event (lower right panel: simulation Poly-ref).

The contribution of inorganics is low compared to the contribution of organics. Figure 2 and Figure 3 show the composition of surface PM_{2.5} concentrations due to fires for simulation Poly-ref and the composition of surface inorganic concentrations respectively over MedReg1 for the first fire event and over MedReg2 for the second fire event.

The contribution of inorganics from fires varies between 8.3 and 9.3%. Focusing on the contribution from fires to inorganics, sulfate, ammonium and nitrate are the predominant components: between 55.7% and 67.6% for sulfate, between 26.8 and 38.7% for ammonium and 5.6 to 13.6% for nitrate.

These sentences are added to page 14 line 24 of the revised paper: “In our study, inorganics (mainly sulfate, sea salt and ammonium) contribute highly to PM_{2.5} composition, if fire emissions are not considered. Similar results are found in Fountoukis et al. (2011) who showed the high contribution of sulfate, sea salt and ammonium to PM over Europe during May 2008. However, when fire emissions are taken into account, the contribution of inorganics from fires is lower than the contribution of organics (8 to 9% of inorganics against 40 to 80% of organics). Focusing on the contribution from fires, sulfate, ammonium and nitrate are the predominant components of inorganics from fires: between 55.7% and 67.6% for sulfate, between 26.8 and 38.7% for ammonium and 5.6 to 13.6% for nitrate.”

B) Further, on p10 they claim that by focusing on PM_{2.5} they avoid contributions from dust, but this simply isn't the case. The low-diameter tail of coarse-mode dust (and sea-salt) contributes of course to PM_{2.5}, and measurements in southern Europe have shown that this can be a significant fraction even over long-periods (Putaud et al., 2010). (This same publication also demonstrates clearly the importance of non-organic compounds in contributing to PM_{2.5}.)

I see no reason to believe that changes in model performance attributed here to various assumptions on OA emissions could not be masked or mistaken for problems with inorganic pollutants. As well as attending to the points raised below, I would encourage the authors to do a thorough analysis of the model performance for all the major compounds, and across many sites. I am afraid I cannot recommend this paper for publication without major revision.

Indeed, the low-diameter tail of coarse-mode dust (and sea-salt) contributes of course to PM₁₀, but we choose not to consider PM₁₀ in this paper (and therefore focus on PM_{2.5}) in order to reduce the uncertainties related to dust emissions as much as possible.

Sentences page 10 line 5-7 “PM₁₀ concentrations in the 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. Since dust is not in the scope of this paper, the analysis focuses on PM_{2.5}.” are replaced by “ PM₁₀ concentrations in the Mediterranean area are strongly affected by dust, which are difficult to simulate due to their sporadic nature and the fact that their main sources are located out of the model domain. In order to evaluate more specifically the uncertainties associated with fine particles (largest contribution from fires), and to minimize the contribution from dust, the analysis focuses on PM_{2.5} .”

Other points:

0. The title is actually quite misleading. The paper only addresses some aspects of OM emissions, not emissions in general.

The title : “Impact of wildfires on particulate matter in the Euro-Mediterranean in 2007: sensitivity to the parameterization of emissions in air quality models.” is replaced by : “Impact of wildfires on particulate matter in the Euro-Mediterranean in 2007: sensitivity to some parameterizations of emissions in air quality models.”

1. The word dispersion, used in eg line 17 of the abstract, is usually associated with turbulent mixing in the air pollution context. Please use another term to make clear that here you mean statistical spread.

The word “dispersion” used in the paper is replaced by the “statistical dispersion” to avoid any kind of confusion.

2. p2, L20. Give reference for statements about anthropogenic origin.

The sentence page 2, line 20-21 : “Although ignitions are mainly of anthropogenic origin (negligence, arson, agricultural practices) , fire spread depends on meteorological conditions.” is replaced by: “Although ignitions are mainly of anthropogenic origin (negligence, arson, agricultural practices) according to San-Miguel-Aynanz et al. (2013) and the European Forest Fire Information System (EFFIS) of the European Joint Research Center (JRC), fire spread depends on meteorological conditions.”

3. p3, L8. Give reference for the volatility limits and definitions. Also, do you really mean from 1e4, or from 0.32e4 (see eg Table 4 of Murphy et al., 2014) ?

The volatility borders are very uncertain, but in this work we followed the volatility limits given in Robinson et al. (2007). The reference is added in the revised paper (page 3, line 12).

Reference:

- Robinson et al., 2007: Allen L. Robinson, Neil M. Donahue, Manish K. Shrivastava, Emily A. Weitkamp, Amy M. Sage, Andrew P. Grieshop, Timothy E. Lane, Jeffrey R. Pierce, Spyros N. Pandis: Rethinking organic Aerosols: Semi volatile Emissions and Photochemical aging, Science, 315, 1259-1262, DOI:10.1126/science.113360

4. Units (e.g. p3. L8) should not be in italics.

All the units in italics are corrected in the revised version of the paper.

5. p3, L23. Change 'current CTMs' to 'two CTMs'. Your study says little about the many other CTMs used in Europe.

The sentences page 3, line 23: “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 (mainly PM_{2.5}).” is replaced by “The objective of this study is to evaluate the capabilities of two CTMs to

simulate the impact of wildfires on the regional particulate matter budget .”

6. Table 1 is very vague and uses many acronyms that haven’t been explained yet. What is ECMWF, which version? What does ’from nesting simulations’ mean? Which version of MEGAN? Which version of EMEP emissions? Give more specific details and references here.

Table 1 summarizes the main characteristics of the Polyphemus and CHIMERE simulations. All the Acronyms used in this Table are explained in the text (from page 5 lines 1-21 to page 6 lines1-23). The clarifications below are added in the revised paper.

6-1 What is ECMWF, which version ?

The sentence page 5 line 20-21 “Both models (Polyphemus and CHIMERE) are driven by meteorological conditions simulated by the European Center for Medium-Range Weather Forecasts (ECMWF) model. ” is replaced by “Both models (Polyphemus and CHIMERE) are driven by meteorological conditions simulated by the European Center for Medium-Range Weather Forecasts (ECMWF, ERA-Interim) model.”

6-2 What does ’from nesting simulations’ mean?

Boundary conditions are from a simulation undertaken using a large domain ($0.5^\circ \times 0.5^\circ$, horizontal resolution) covering Europe and North Africa (see Figure G).

The Figure G is added in the appendix A in the revised paper.

The sentence page 6 line 3: “ Simulations are undertaken using two nested domains.” is replaced by “ Simulations are undertaken using two nested domains (Figure A1 in Appendix A).”

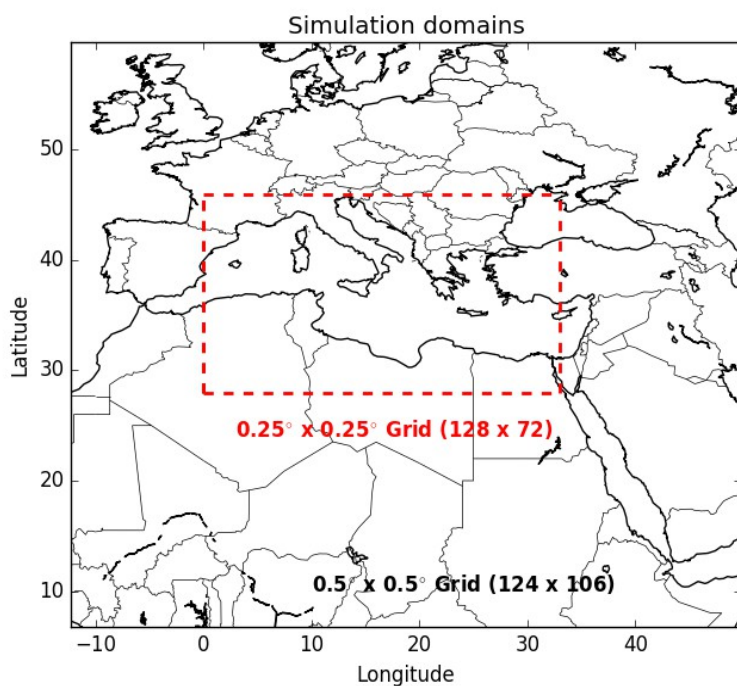


Figure G: Simulation domains including one large domain (with a $0.5^\circ \times 0.5^\circ$ horizontal resolution) and a smaller domain (at a $0.25^\circ \times 0.25^\circ$ horizontal resolution) delimited by the dotted red box.

6-3 Which version of MEGAN?

The sentence page 6 line 9-10 “Biogenic emissions are calculated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006).” is replaced by “ Biogenic emissions are calculated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) with the standard MEGAN LHIV database MEGAN-L for Polyphemus and MEGAN v2.04 or CHIMERE (Guenther et al., 2006).”

6-4 Which version of EMEP emissions?

The sentences page 6 line 4-5 “Anthropogenic emissions are derived from the EMEP emissions inventory (European Monitoring and Evaluation Program, www.emep.int).” is replaced by “Anthropogenic emissions are derived from the EMEP emissions inventory for 2007. (European Monitoring and Evaluation Program, www.emep.int)”

7. P6, L3. The nesting factor is here given as 2:1, but usually one uses an odd ratio to avoid interpolation errors (e.g.

<https://earthscience.stackexchange.com/questions/8067/why-is-wrf-most-often-configured-at-31-nesting-ratio>). **Have the authors checked if their results are robust?**

The recommendation to use an odd ratio applies to WRF simulations. However, this is not required by Polyphemus nesting. Indeed the interpolation is done properly in 2D in Polyphemus, and it is accurate because the vertical levels do not vary with time/space.

8. p6, L9. Biogenic emissions of what? I assume isoprene and terpenes, but which?

The sentence page 6 lines 9-10 “ Biogenic emissions are calculated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006).” is replaced by “Biogenic emissions of isoprene and terpenes (α -pinene, β -pinene, limonene and humulene) are calculated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006).”

9. p6, L17-24. The explanation of the VBS scheme used is unclear. Firstly, where do the numbers -0.04, 1.93 and 3.5 come from? They are not suggested by May et al. 2013, and not obvious to the reader. Secondly, L21 suggests that OH+POA leads to SOA compounds in the same VBS bin, but L22-23 suggests a reduction in volatility equivalent by a factor of 100. Which is it?

In this work, we didn't use a VBS scheme. We used the one-step oxidation scheme developed by Couvidat et al. (2012) where the emission distribution is based on the fitting of the curve of dilution of diesel exhaust from Robinson et al. (2007). This emission distribution is approximately similar to the one measured by May et al. (2013a) for biomass burning (see Table A). According to Couvidat et al. (2012), I/S/L-VOCs emissions are assigned to 3 surrogates species: POAIP, POAmP and POAhP (for compounds of low, medium and high volatilities respectively), of saturation concentration C^* : $\log_{10}(C^*) = -0.04, 1.93, 3.5$ respectively.

C^* ($\mu\text{g}/\text{m}^3$)	Couvidat et al. (2012) emissions distribution	May et al. (2013a) emissions distribution for biomass burning
$10^{-0.04}$	0.25	0.20 0.20
$10^{1.93}$	0.32	0.10 0.20
$10^{3.5}$	0.43	0.4

Table A: the emission distribution of I/S/L-VOCs given by Couvidat et al. (2012) and May et al. (2013a)

The sentences in page 6, lines 17-24: “I/S/L-VOCs emissions are assigned to 3 surrogates species: POAIP, POAmP and POAhP (for compounds of low, medium and high volatilities respectively), of saturation concentration C^* : $\log(C^*) = -0.04, 1.93, 3.5$ respectively. The volatility distribution at emissions of I/S/L-VOCs is detailed in Couvidat et al. (2012) (25%, 32%, and 43% of I/S/L-VOCs are assigned to POAIP, POAmP and POAhP respectively). It corresponds to the volatility distribution measured by May et al. (2013) for biomass burning aerosol emissions. Each primary aerosol undergoes one OH-oxidation reaction in the gas phase with a kinetic rate constant equal to $2.10^{-11} \text{ molecule}^{-1} .\text{cm}^3 .\text{s}^{-1}$, leading to the formation of secondary surrogate in the same volatility: SOAIP, SOAmP and SOAhP. The ageing of the primary aerosols reduces the volatility of the secondary product by a factor of 100 and increases the molecular weight by 40% (Couvidat et al., 2012). ” are replaced by “I/S/L-VOCs emissions are assigned to 3 surrogates species: POAIP, POAmP and POAhP (for compounds of low, medium and high volatilities respectively), of saturation concentration C^* : $\log(C^*) = -0.04, 1.93, 3.5$ respectively. The volatility distribution at emissions of I/S/L-VOCs is detailed in Couvidat et al. (2012) (25%, 32%, and 43% of I/S/L-VOCs are assigned to POAIP, POAmP and POAhP respectively). It corresponds to the volatility distribution measured by May et al. (2013a) for biomass burning aerosol emissions. Each primary aerosol undergoes one OH-oxidation reaction in the gas phase with a kinetic rate constant equal to $2.10^{-11} \text{ molecule}^{-1} .\text{cm}^3 .\text{s}^{-1}$, leading to the formation of secondary surrogates: SOAIP, SOAmP and SOAhP. The ageing of the primary aerosols reduces the volatility of the secondary product by a factor of 100 and increases the molecular weight by 40% (Couvidat et al., 2012).”

10. p6, L17. The use of 'log' is ambiguous. In most VBS papers it means log10, but in other papers (especially mathematical) and common programming languages (eg python) it means the natural logarithm, log_e (or ln). I suggest using log10 to be clear.

The sentence page 6, line 17 : “I/S/L-VOCs emissions are assigned to 3 surrogates species: POAIP, POAmP and POAhP (for compounds of low, medium and high volatilities respectively), of saturation concentration C^* : $\log(C^*) = -0.04, 1.93, 3.5$ respectively.” is replaced by “I/S/L-VOCs emissions are assigned to 3 surrogates species: POAIP, POAmP and POAhP (for compounds of low, medium and high volatilities respectively), of saturation concentration C^* : $\log_{10}(C^*) = -0.04, 1.93, 3.5$ respectively.”

11. p6, L33-34. Akagi provides emission factors for several ecosystems, but non are a close match to Mediterranean landscapes. And some of these emitted isoprene and terpenes, which also form SOA. Which emissions were in fact used?

The sentence in page 6 line 33 “Emissions for each species are derived from the carbon emissions using the emissions factors from Akagi et al. (2011).” is replaced by “Emissions for each species are derived from the carbon emissions using the emissions factors from Akagi et al. (2011). These emission factors are provided in terms of g species per Kg Dry Matter (DM) burned (g.kg⁻¹) for all relevant species observed in biomass burning plumes and for different standard vegetation types that match to Mediterranean landscapes (chaparral, temperate forest, crop residue, pasture maintenance and savanna). The contribution of these vegetation types to the burned area detection over the Mediterranean region during the time period studied is 37.2% for temperate forest, 32.7% for savanna, 9.6 % for chaparral and 19.9% for crop residue.”

The isoprene and terpenes (α -pinene, β -pinene, limonene and humulene) emissions from fires are considered in this work.

12. Also, L33 suggests that only carbon emissions were used, but many studies suggest that fires are a major and sometimes huge source of gases including NH₃ (eg van der Werf et al., 2017, Van Damme et al., 2014), and hence of inorganic PM. Were such emissions examined for their impact on PM? Were they checked?

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) were rather on the lower end of those reported in Akagi et al. (2011) (used here). Therefore, we believe we did not underestimate NH₃ emissions. However, the impact on inorganic concentrations is low, as discussed previously in comment 1.

13. p7, The notation PPM_{fine} is very confusing. This is usually used to represent emissions of any primary particulate matter including OM, BC and inorganics. Am I right in saying that your PPM_{fine} is PM_{2.5} - OC - BC - SIA? In which case I would expect most of be essentially OM-OC - is that what you mean? To further the confusion with this PPM_{fine} notation, you introduce PPM_{coarse} which is more traditional, just PM₁₀-PM_{2.5}. So, PPM_{fine} excludes OC, BC and SIA, and consists largely of O and H, whereas PPM_{coarse} consists of OM+BC+SIA. As I said, confusing.

PPM_{fine} corresponds to all the unidentified fine particles emitted by wildfires which are incorporated to consider the differences between PM_{2.5} emissions and the total of all PM included in specific species. Therefore, PPM_{fine} corresponds to PM_{2.5}-OM-BC-SIA.

This is why we believe that PPM_{fine} includes actually OM and I/S/L-VOCs.

For clarity, the following sentences are added in page 7 lines 11-14: “PPM_{fine} corresponds to all the unidentified fine particles emitted by wildfires which are incorporated to consider the differences between PM_{2.5} emissions and the total of all PM_{2.5} speciated emissions.”

14. p7, Figure 1. It is difficult to see the yellow fire labels on top of the yellow background. Change the colors.

In the revised paper, the Figure 1 page 7 is replaced by the Figure A below.

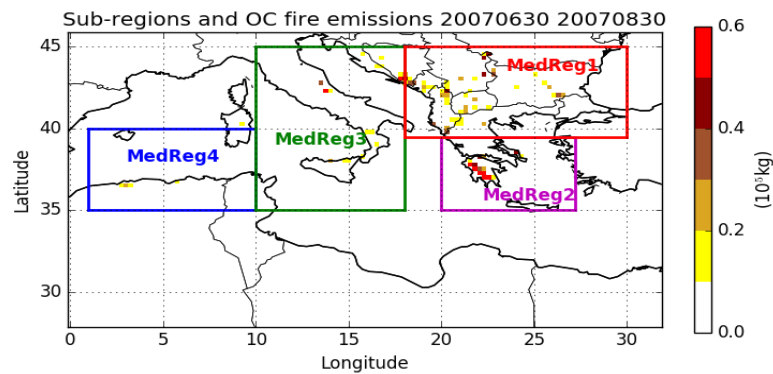


Figure A: Map of the nested domain over the Mediterranean area with a spatial resolution of $0.25^\circ \times 0.25^\circ$. The total organic carbon emissions(kg. (grid cell) $^{-1}$) from fires during the summer of 2007 are presented. The sub-regions used in this study are also indicated in colored boxes: MedReg1 (Balkan + Eastern Europe), MedReg2 (Greece), MedReg3 (Italy) and MedReg4 (Algeria).

15. p9. Table 2.

- The Poly-ref emissions are said to be introduced between 1km and the PBL. I assume you mean between ground level and min(PBL,1km)? If not, I have a serious problem with your base-case!

Indeed, we meant that fire emissions in the simulation Poly-ref by are injected in min (PBL, 1km).

- What is PB?

We mean by PBL : Planetary Boundary Layer.

“PB” in Table 2, page 9 is replaced by “PBL (Planetary Boundary Layer)”.

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 PM_{2.5} 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 PM_{2.5} 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 in page 27 line 26- page 28 line 4 in the revised paper)

“Over Europe, inorganics (mainly sulfate, sea salt and ammonium) contribute highly to

PM2.5 composition, when fire emissions are not considered (e.g., Fountoukis et al., 2011; Chrit et al., 2018). 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 due to wildfires is found to be low compared to the formation of organics. However, our simulation takes into account emissions of inorganic precursors such as ammoniac (NH₃), with emission factors from Akagi et al. (2011).

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)

These sentences: “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 the most affected by wildfires (background suburban and rural stations with fire contribution 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. ” are added in page 26 lines 4-11 in the revised paper.

The sentence :“More surface observations in remote regions would be 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.” is added in page 27 lines 9-11 in the revised paper.

(3-Details concerning the emissions of PPM_{fine} and I/S/L-VOCs are added in the conclusion, page 28 lines 5-13 in the revised paper)

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

This study shows that considering PPM_{fine} emissions and I/S/L-VOC emissions may be redundant and may correspond to the same “missing emissions mass”.

Several models actually partly treat this missing part by considering I/S/L-VOCs 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 the primary organic aerosols (POA) by a factor of 1.5 following 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- Discussion on uncertainties on the burned area is added in page 28 lines 14-15 in the revised paper)

“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- Discussion on uncertainties due to deposition is added in page 28 lines 16-21 in the revised paper)

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 PM_{2.5} concentrations.

#Specific comments :

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, lines 18 – 20 in the revised paper: “CHIMERE simulations (Menut et al., 2013) allow us to perform inter-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 PM_{2.5} 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 aerosol optical depth (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 the most affected by wildfires (background suburban and rural stations with fire contribution 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.”

These sentences are added to page 27 lines 9-11: “More surface observations in remote regions would be 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 (page 28 lines 16-21 in the revised paper).

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 strengthened 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 PPM_{fine} 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 PM_{2.5} and the uncertainties in its definition (expected overestimation). How is this missing part treated in the other models?

PPM_{fine} are all the unidentified fine particles emitted by wildfires which are incorporated to consider the differences between PM_{2.5} emissions and the total of all PM included in specific species. There are a lot of uncertainties related to the estimation of PPM_{fine} 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 Figure 1”.

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 PM_{2.5}”

The legend in Figure 3 in page 12 “Daily mean PM_{2.5} 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.”