Impact of wildfires on particulate matter in the Euro-Mediterranean in 2007: sensitivity to the parameterization 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 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 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 $\sim$ 10 days in fire plumes, which is slightly underestimated compared to available observations. The maximum 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 $\sim 500\ 10^3$ 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 (negligeance, arson, agricultural practices), 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 $PM_{2.5}$ (particulate matter with diameter $\leq 2.5\ \mu m$). 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-

5 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 $10^6 \mu g.m^{-3}$), intermediate organic compounds (I-VOCs) (with saturation concentration C^* in the range of 10^4-$10^6 \mu g.m^{-3}$) and semi-volatile organic compounds (S-VOCs) (with saturation concentration C^* in the range 0.1–$10^4 \mu g.m^{-3}$),

10 Low volatility organic compounds (L-VOCs) (with saturation concentration C^* lower than $0.1 \ \mu g.m^{-3}$) (Lipsky and Robinson, 2006; Grieshop et al., 2009; Huffman et al., 2009; Cappa and Jimenez, 2010; Fountoukis et al., 2014; Tsimpidi et al., 2010; Woody et al., 2016; Ciarelli 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., 2013;

15 Koo et al., 2014; Konovalov et al., 2015; Ciarelli 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; Ciarelli et al., 2017). The chamber experiments of May et al. (2013) 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

20 and observed organic aerosol (OA) (Koo et al., 2014; Konovalov et al., 2015; Ciarelli 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 CTMs to simulate the impact of wildfires on the regional particulate matter budget ($PM_{2.5}$, PM_{10}). After an evaluation of two CTMs (CHIMERE and Polyphemus/Polair3D) through

25 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

30 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 extreme 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.,

35 2009; Kaskaoutis et al., 2011) due to these 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 O_3 on NO_x levels, the near surface O_3 values were reduced (-34%) over the Poleponnese, but increased (+52%) over the sea at

5 500 km downwind. Here, the sensitivity of 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

10 to assess the models performance during the summer 2007 and more specifically during the main fire events. Then, uncertainties 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

15 Two CTMs are used for this study. 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). Table 1 summarizes the main characteristics of the simulations.

Table 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°×0.5° - Small domain: 0.25°×0.25°
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

5   Aerosol Model (SIREAM) (Debry et al.) 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'echelle Regionale 2

10 (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. Thermodynamics of inorganic aerosols are modeled using ISORROPIA (Nenes

15 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).

20 Both models (Polyphemus and CHIMERE) are driven by meteorological conditions simulated by the European Center for Medium-Range Weather Forecasts (ECMWF) 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. One large domain (0.5°×0.5 horizontal resolution) covering Europe and North Africa to provide realistic dust sources and a smaller domain at 0.25°×0.25° horizontal resolution over the Mediterranean area (presented in Figure 1).

Anthropogenic emissions are derived from the EMEP emissions inventory (European Monitoring and Evaluation Program,
5 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 are calculated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther
10 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; Kim et al., 2016).
15 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: POAlP, 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 POAlP, POAmP and POAhP respectively). It corresponds to the volatility distribution measured by May et al.
20 (2013) for biomass burning. Each primary aerosol undergoes one OH-oxidation reaction in the gas phase with a kinetic rate constant equal to 2.10^{-11} $molecule^{-1}.cm^3.s^{-1}$, leading to the formation of secondary surrogate in the same volatility bin: SOAlP, 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).

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
30 vegetation dynamics and carbon cycle model (Maignan et al., 2009). Turquety et al. (2014) estimated an uncertainty of ~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). 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 main primary emissions (organic and black carbon, small amounts of inorganics) and for the total $PM_{2.5}$ is modelled as a specific, inert and unidentified species grouping other fine mode primary particulate matter PPM_{fine}. The difference between PM_{10} and $PM_{2.5}$ emission factors is attributed to emissions of coarse mode PM (PPM_{coarse}).

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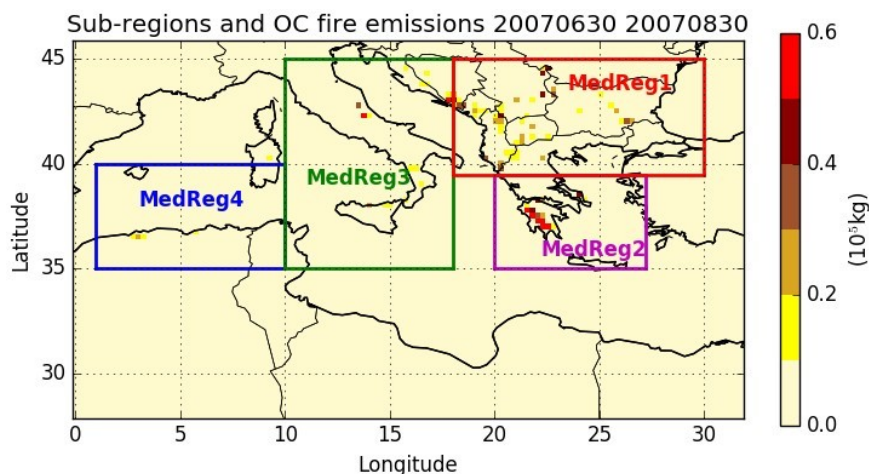
Figure 1. Map of the nested domain over the Mediterranean area with a spatial resolution of 0.25°×0.25°. The total organic carbon emissions (kg. (grid cell)$^{-1}$) from fires during the summer of 2007 are presented. The sub-regions used in this study are also indicated in colored boxes: MedReg1 (Balkan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

Total daily fire emissions for the four studied areas are plotted in Figure 2. In all regions, fire emissions are occasional but

10   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.
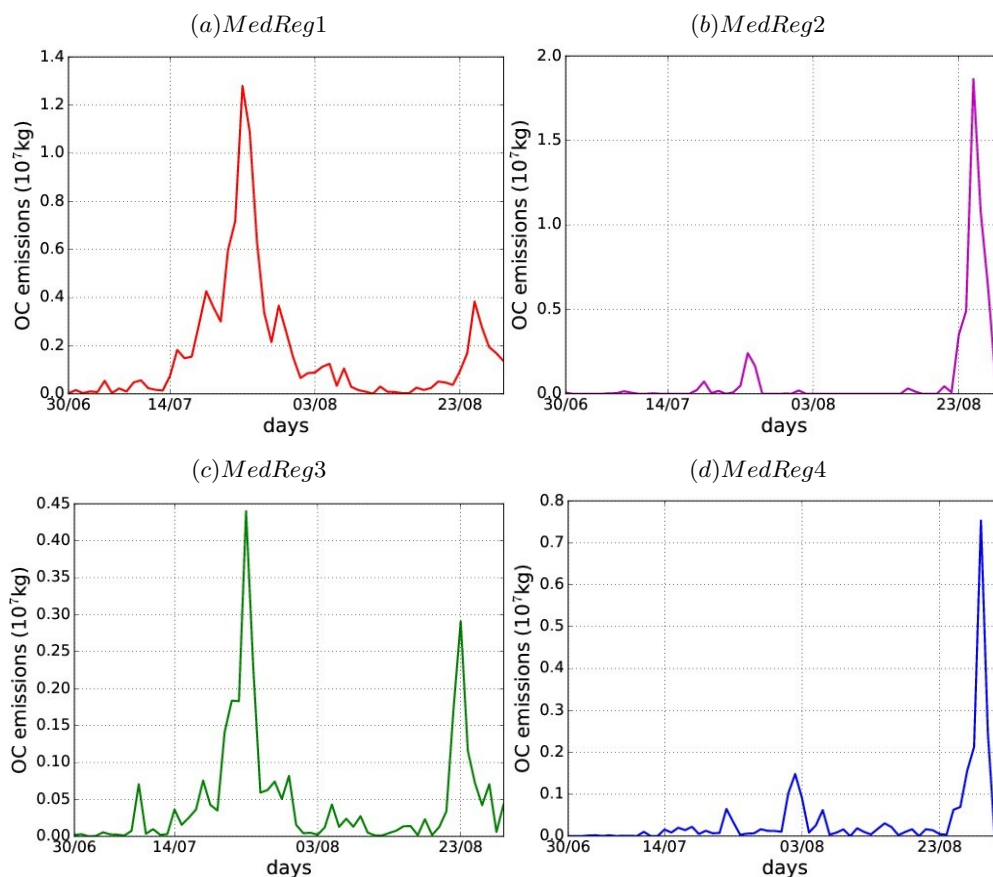
Figure 2. Daily total OC emissions during the summer of 2007 in the four sub-regions of Figure 1.

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:

5   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 as LVOCs.

10   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.

Table 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 *Poly-ref*	Yes	Yes	Between 1km and PBL	Yes
Simulation *Poly-3km*	Yes	Yes	20% under 1km 80% between 1 and 3km	Yes
Simulation *Poly-NoPPM*	Yes	Yes	Between 1km and PB	No
Simulation *Poly-noI/S-VOCs*	Yes	No	Between 1km and PB	Yes
Simulation *CHIMERE-ref*	Yes	Yes	Between 1km and PB	Yes
Simulation *CHIMERE-Nofires*	No	N/A	N/A	N/A
Simulation *Poly-Nofires*	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. PM_{10} 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 $PM_{2.5}$.

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.

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 10×10 km^2 are used (Remer et al., 2005). The expected uncertainty on AOD is $\Delta\tau= \pm0.05\pm0.15\tau$ (τ is the optical thickness) over land (Chu and Holben, 1998; King et al., 1999) and $\Delta\tau= \pm0.03 \pm0.05\tau$ 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:

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

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

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

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

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 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 Table 3 for PM$_{2.5}$ concentrations and Table 4 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, CHIMERE outputs have lower biais (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 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. The mean simulated PM$_{2.5}$ concentrations can reach $60 \, \mu g.m^{-3}$ 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 3 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. 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 (%)
Poly-ref	13.2	9.3	82	-32	42
Poly-noI/S-VOCs	13.2	9.3	82	-32	42
Poly-3km	13.2	9.1	82	-33	43
Poly-NoPPM	13.2	9.1	82	-33	43
Poly-Nofires	13.2	8.4	78	-37	46
CHIMERE-Nofires	13.2	11.2	70	-15	39
CHIMERE-ref	13.2	11.3	67	-10	39

Table 4. 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 (%)
Poly-ref	0.27	0.22	62	-14	34
Poly-noI/S-VOCs	0.27	0.21	64	-18	35
Poly-3km	0.27	0.22	64	-15	34
Poly-NoPPM	0.27	0.22	62	-14	34
Poly-Nofires	0.27	0.19	56	-24	39
CHIMERE-Nofires	0.27	0.23	36	-7	39
CHIMERE-ref	0.27	0.24	46	-6	36
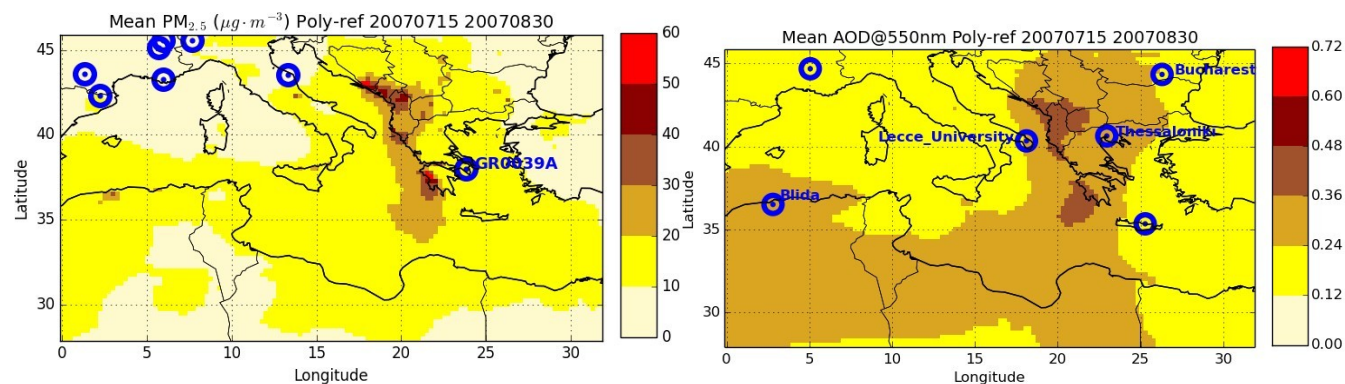
Figure 3. Daily mean PM$_{2.5}$ and AOD at 550 nm from the *Poly-ref* simulation averaged over the summer of 2007 (the 8 AIRBASE and 6 AERONET stations, used in this work, are represented here in blue dots).

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.

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 4 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.

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.
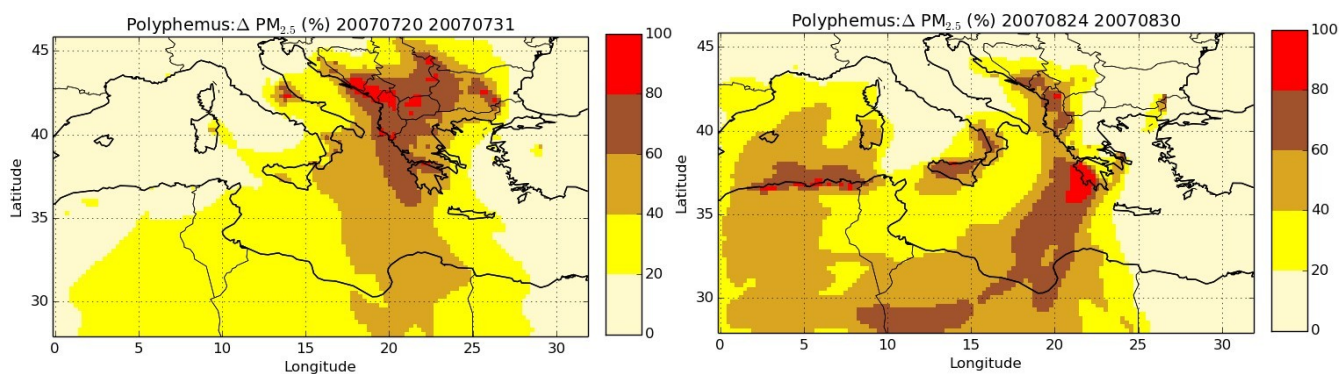
Figure 4. Left panel: relative difference of $PM_{2.5}$ concentrations between simulations *Poly-ref* and *Poly-Nofires* during the first fire event. Right panel: relative difference of $PM_{2.5}$ concentrations between simulations *Poly-ref* and *Poly-Nofires* during the second fire event.

3.4.2 Aerosol composition during fire peaks

Figure 5 shows the composition of surface $PM_{2.5}$ concentrations simulated during the first peak (on 25 July 2007) at the AIRBASE station GR0039A. 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 simulation without fire, organic aerosols, inorganic aerosols and PPM_{fine} (dust) contribute equally to the surface $PM_{2.5}$ concentrations (35%, 31% and 34%). The contribution of black carbon is low (about 5%). As noted by Chrit et al. (2017), most of summer organic aerosols are from biogenic sources in this region. Figure 5 also shows 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*, between the simulations *Poly-NoI/S-VOCs* and *Poly-Nofires* and between the simulations *Poly-NoPPM* and *Poly-Nofires* respectively). Organic aerosol is predominant in the contribution of fires (between 52% and 87% of the contribution). Organic aerosol is mostly composed of POA and SOA from I/S/L-VOCs (62% to 84%). 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%), because POA are then assigned to L-VOCs. The contribution from inorganics (7% to 10%) and black carbon (2% to 3%) are low.

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).

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 mainly of organic aerosols, mostly composed by primary organic carbon (OCAR) (48%), and of PPM_{fine} (37%). The contributions from inorganics (8.2%), black carbon (4.4%) and SOA (2.7%) are low.
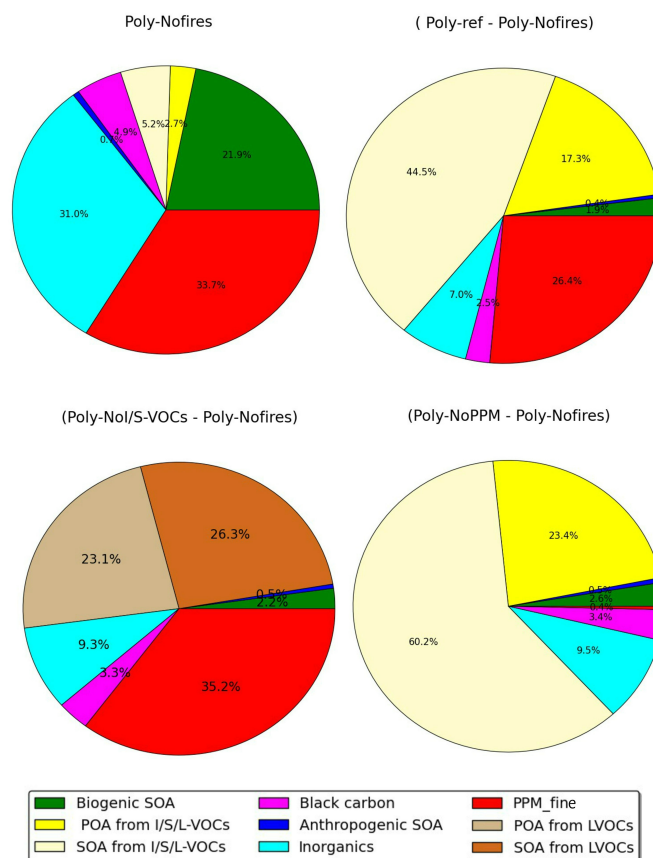
Figure 5. Composition of surface PM$_{2.5}$ concentration at the GR0039A station on 25 July 2007 (upper left panel, simulation *Poly-nofires*). Composition of surface PM$_{2.5}$ concentration due to fires (upper right panel: simulation *Poly-ref*; lower left panel: simulation *Poly-NoI/S-VOCs*; lower right panel: simulation *Poly-NoPPM*).

3.4.3 Evaluation of fire contribution

Figure 6 shows time series of daily observed and simulated aerosols at stations with significant impact from fires (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
5 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 1, are shown in Figure 7. Background AOD and daily variability are consistent with MODIS for both models in the sub-regions of Figure 1, with high correlation coefficients (∼90% on average for Polyphemus, 80% for CHIMERE).

As shown in Figures 6 and 7, injecting emissions higher (simulation *Poly-3km*) significantly lowers surface PM concen-
10 trations (compared to the simulation *Poly-ref*), even if the maximum injection height remains conservative. Not taking into

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 8 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 iclude I/S-VOCs emissions. Compared to MODIS AOD (Figure 7), 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 6), 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. 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 Bucharest, two high AOD peaks are modeled (Figure 6). 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 transport), 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. 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 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 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 ($\sim$100%) and on the observations(1-2% for AERONET observations (Eck et al., 1999) and $\sim$34% for AIRBASE observations (Bovchaliuk, 2013), the performance obtained is considered very reasonable. The spread in the different model configurations tested shows the additional uncertainty on the modeling of fire impact. Observations generally lie within the simulated variability but it is difficult to extract the best model configuration (it depends on the event and on the station).
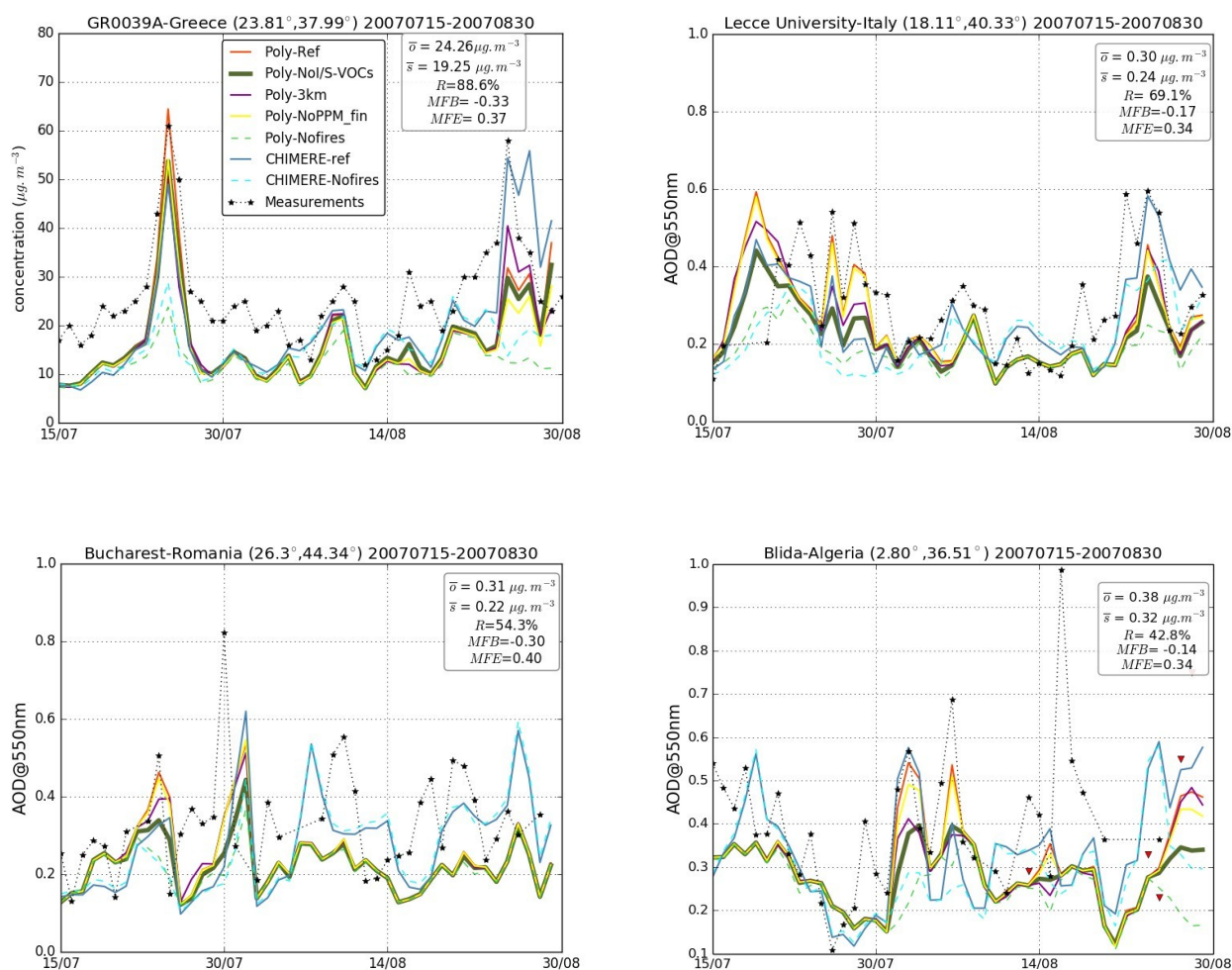
Figure 6. Time series from 15 July to 30 August of daily mean surface PM$_{2.5}$ concentrations at the AIRBASE station GR0039A and daily mean AOD at 550 nm at three AERONET stations (Lecce University, Bucharest, Blida). The red triangles in Blida station correspond to AERONET measurements using AOD level 1.5 AOD data at 500 nm. Statistics for simulation *Poly-Ref* are shown at each station.
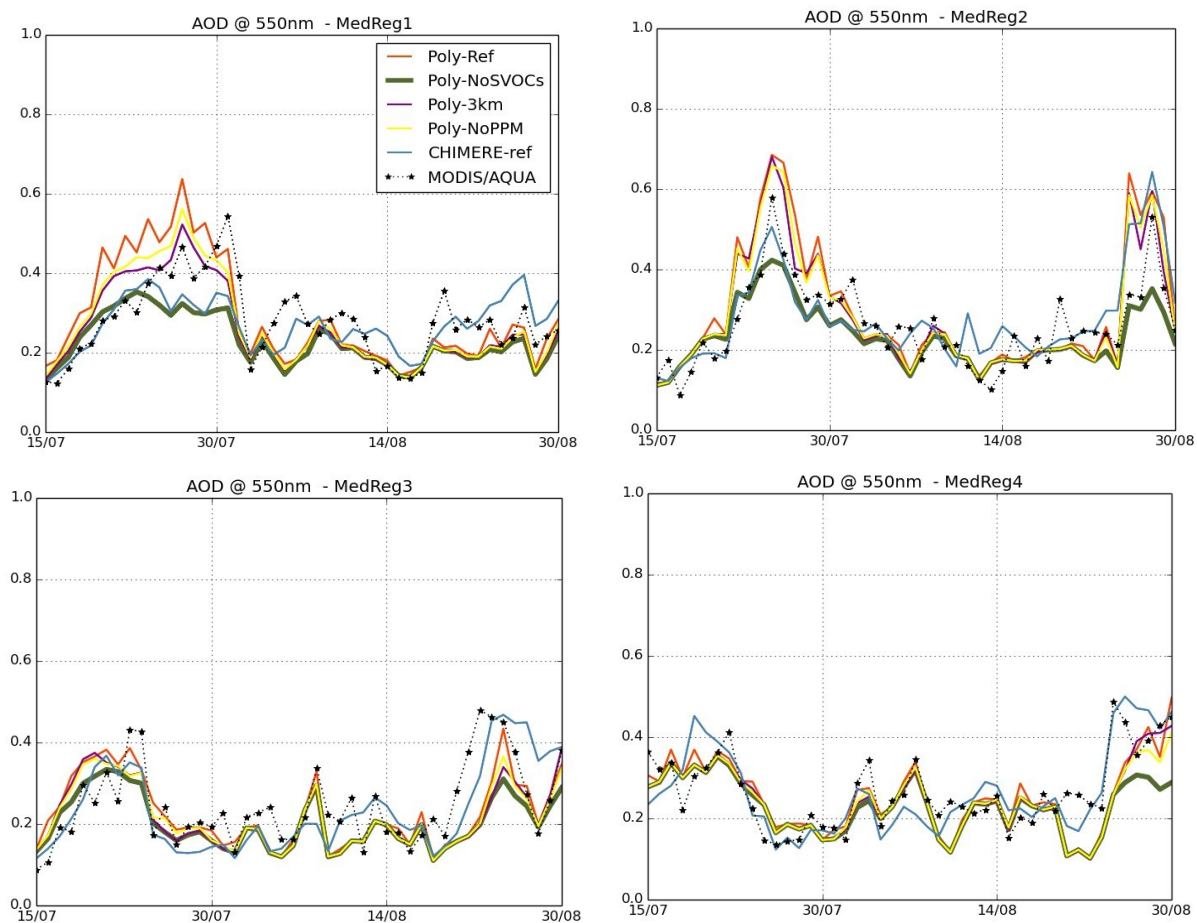
Figure 7. Daily mean AOD at 550 nm observed by MODIS and simulated by Polyphemus and CHIMERE from 15 July to 30 August 2007, in the sub-regions of Figure 1.
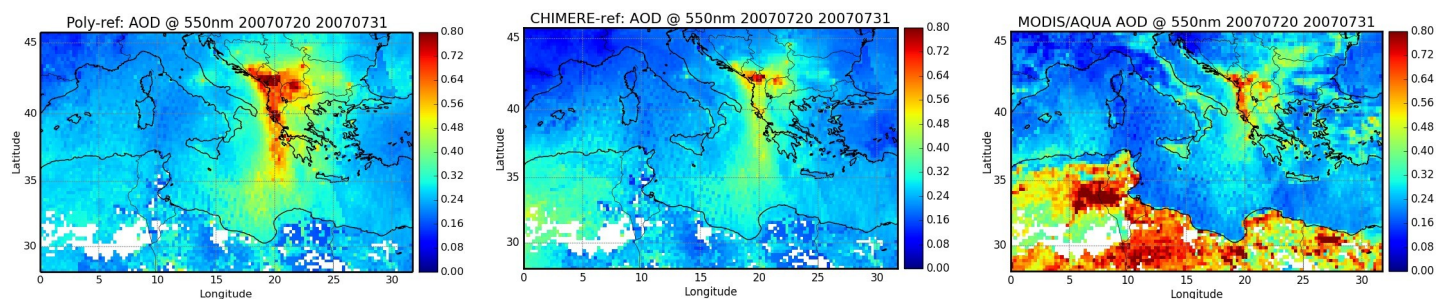
Figure 8. Mean total AOD (at 550 nm) from MODIS/AQUA, *Poly-Ref* and *CHIMERE-ref* during the first fire event (20-31 July 2007).

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 9, 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 $\mu g.m^{-3}$ 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 models use different vertical coordinates and different numbers of 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 sensivitity 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 ($> 10 \ \mu g.m^{-3}$) caused by fire emissions injection height over the United States. According to Daskalakis et al. (2015), assumptions on the injection

5   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%).

This analysis highlights that injecting above the boundary layer is more critical for surface $PM_{2.5}$ concentrations than

10   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.
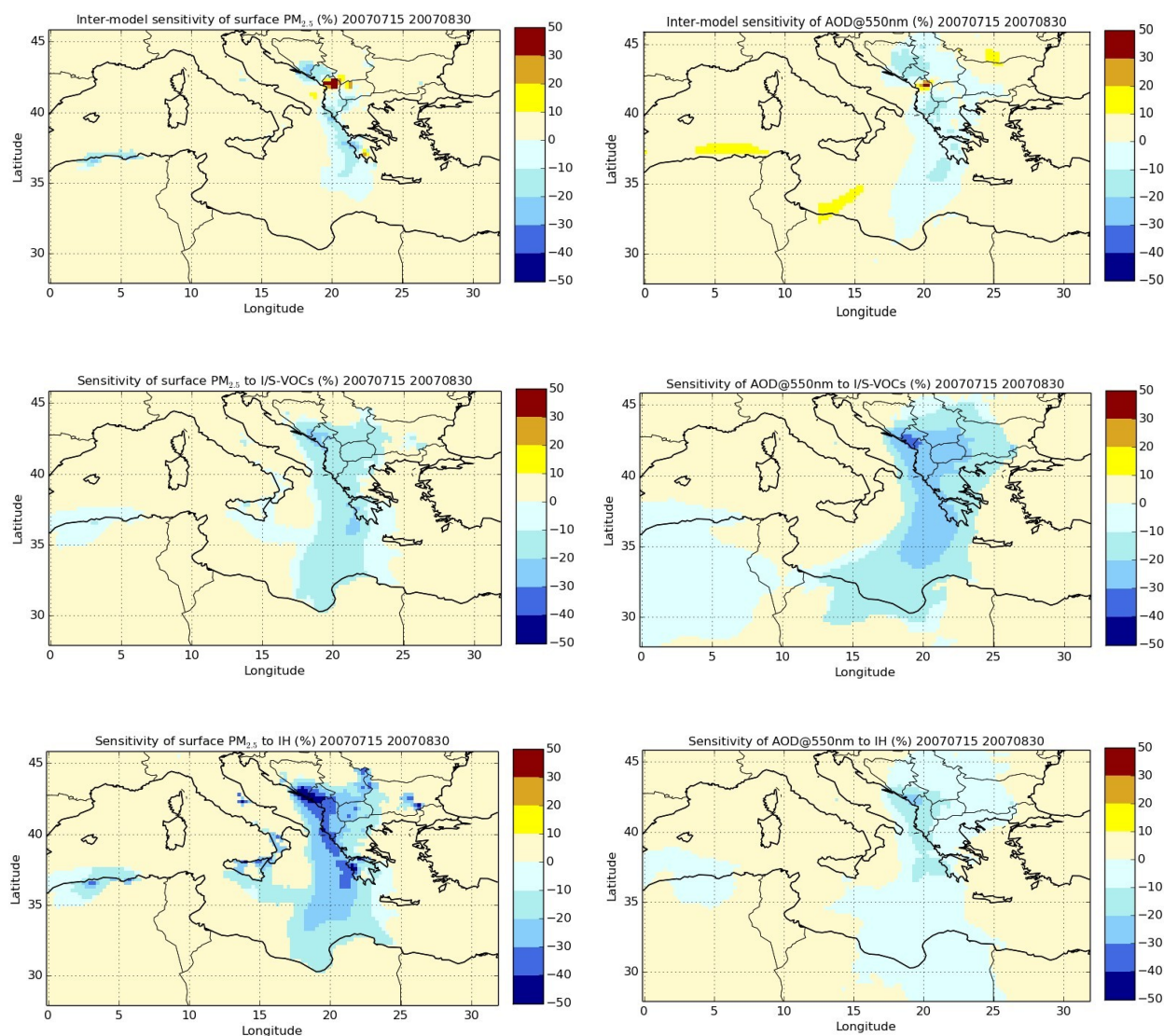
Figure 9. Sensitivity of surface PM$_{2.5}$ concentrations (left panels) and AOD at 550 nm (right panels) to the CTM used (*CHIMERE-ref* or *Poly-ref*, upper panels), the modelling of I/S-VOCs emissions (*Poly-noI/S-VOCs* or *Poly-ref*, middle panels) and the injection height (IH) (*Poly-3km* or *Poly-ref*, 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.
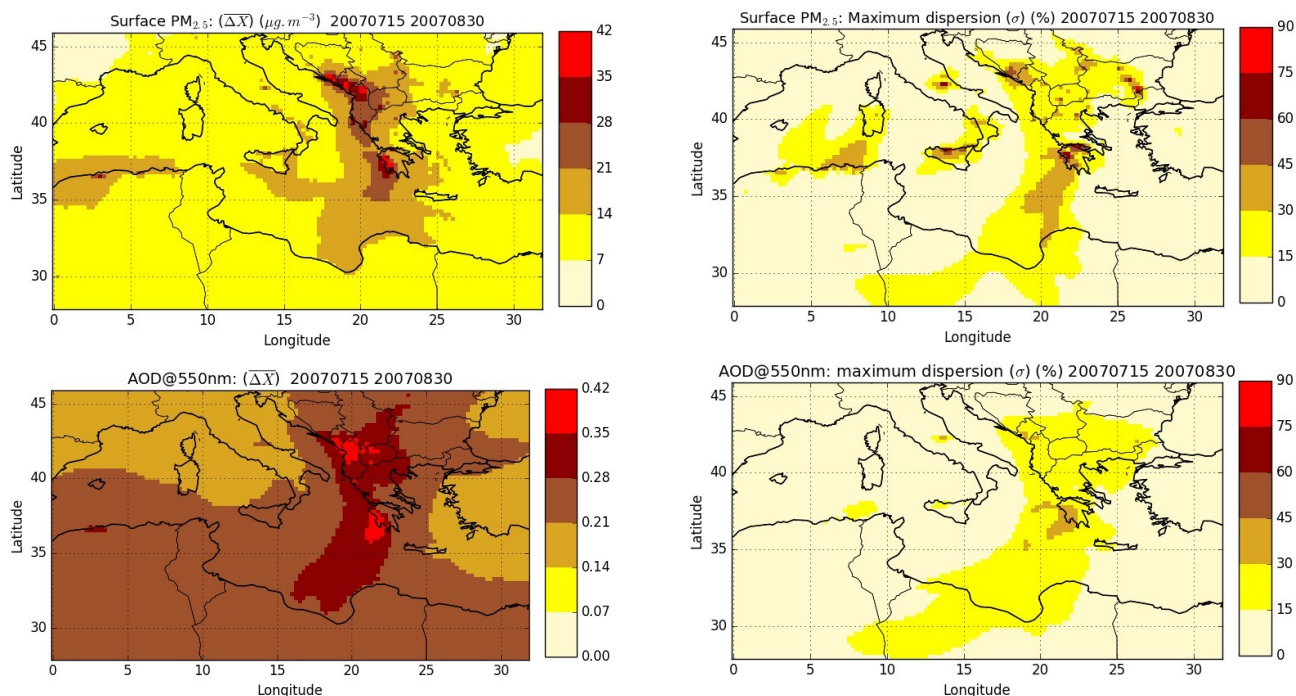
Figure 10. Mean surface $PM_{2.5}$ concentrations and AOD (estimated using four Polyphemus sensitivity simulations) (left panel) and their relative maximum dispersion (σ) (right panel) during the summer 2007.

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 dispersion (σ) is used as a statistical estimator. The maximum dispersion (σ) for surface $PM_{2.5}$ and AOD calculated as:

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

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

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

Figure 10 shows the average surface $PM_{2.5}$ concentrations and AOD estimated using four Polyphemus simulations and the maximum dispersion (σ) for surface $PM_{2.5}$ concentrations and AOD. The maximum dispersion related to simulated surface $PM_{2.5}$ concentrations and AOD is higher near the fire regions. The mean surface $PM_{2.5}$ concentrations estimated from the four

Polyphemus simulations can reach 42 $\mu g.m^{-3}$ in Balkan and Greece with a dispersion that can be as high as 75%. Lower mean surface PM$_{2.5}$ concentrations are calculated for Algeria, in the fresh fire plume and further downwind (28 $\mu g.m^{-3}$ with a maximum 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 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 PM$_{2.5}$ surface concentrations exceeds 25 $\mu g.m^{-3}$ (World Health Organisation recommendation, (Krzyzanowski and Cohen, 2008)).

10 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 PM$_{2.5}$ exceedances were observed and only 7 are simulated at GR0039A by *Poly-ref* and 9 by *CHIMERE-ref*.

Figure 11 shows the simulated number of days concerned by a PM$_{2.5}$ exceedance from fires (difference between the *Poly-ref* and *Poly-Nofires*). Most are concentrated around fire sources, mainly in Balkan (30 days). Fires cause up to 49.5% of the total 15 simulated exceedances from 15 July to 30 August 2007.
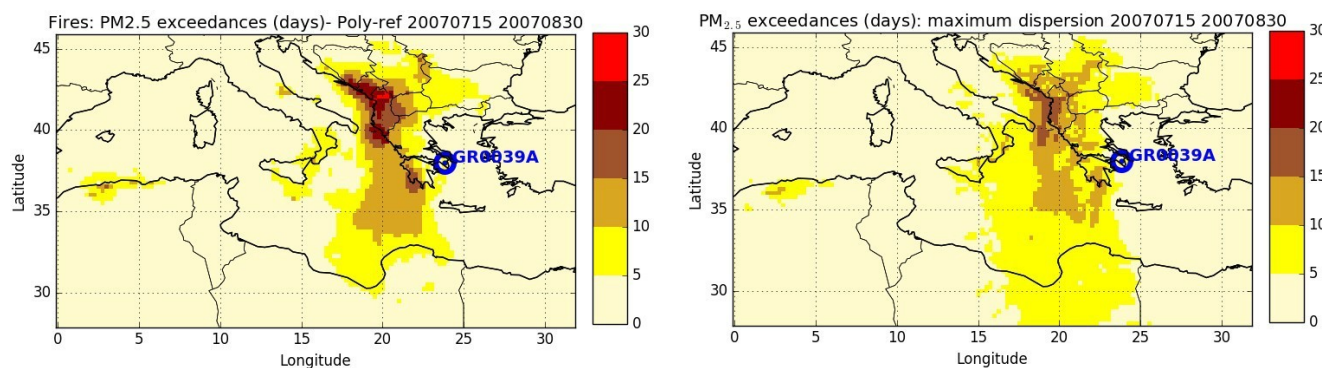
Figure 11. PM$_{2.5}$ exceedance (days) from fires (difference between the *Poly-ref* and *Poly-Nofires*) (left panel) and the maximum dispersion (σ) related to fire emissions for PM$_{2.5}$ exceedances days (right panel) during the summer 2007 (from 15 July to 30 August 2007).

During the fire periods, surface PM$_{2.5}$ 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).

Figure 11 shows that the maximum dispersion on the simulation of PM$_{2.5}$ exceedances can reach 15 days in regions affected 20 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.

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 $PM_{2.5}$ concentrations and optical properties (particularly 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 $PM_{2.5}$ concentrations and AOD. A clear improvement is noticed when fires are taken into account. The contribution of fires is large in Balkan and Easten 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 (52% to 87%), with a strong contribution from I/S/L-VOCs (62% to 84%). The comparison between simulated AOD and satellite observations (MODIS) highlights consistent daily variability for both models in all sub-regions with high correlation coefficients (∼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 unidentified primary particles emitted from biomass burning are not. This suggests that these primary particles which are not identified in the emission inventory, 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 $PM_{2.5}$ concentrations are well reproduced by the models. The simulation *Poly-Ref* reproduces well the $PM_{2.5}$ peaks with a high correlation coefficient (>88%) and a low bias. The $PM_{2.5}$ 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 $PM_{2.5}$ 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.

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 $PM_{2.5}$ concentrations and AOD. Sensitivities to these key parameters are computed using simulations performed with different configurations of Polyphemus. AOD is particularly sensitive to I/S-VOCs emissions (up to 40% sensitivity), while surface $PM_{2.5}$ concentrations are particularly sensitive to the injection heights (up to 50% sensitivity).

The dispersion of the ensemble of simulations based on different configurations of Polyphemus is used to evaluate the maximum uncertainty on surface $PM_{2.5}$ concentrations and AOD associated with these two parameters. During the summer 2007, the maximum dispersion (σ) is as high as 75% for surface $PM_{2.5}$ 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 $\mu g.m^{-3}$ for $PM_{2.5}$ 24-hour mean

5   reaches 30 days for the fire region and $10 - 15$ days for the fire plume over the simulated period of 46 days. The maximum 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 addiction to fire emissions.

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 $PM_{2.5}$ concentrations at urban sites displayed large sensitivities to

10   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.

Acknowledgements. CEREA and LMD are members of the Institut Pierre-Simon Laplace (IPSL). This research was partially funded by a

15   PhD grant from École des Ponts ParisTech, and by the french ministry of ecology, by CNRS-INSU (GMES-MDD program, NATORGA project). This study also received financial support from MISTRALS by ADEME, CEA, INSU, and Meteo-France, as part of the ChArMEx 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

20   Space Agency (NASA) for the availability of the MODIS data.

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