



An estimate of excess mortality resulting from air pollution caused by wildfires in the eastern and central Mediterranean basin in 2021

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Abstract. Wildfires result in human fatalities not only due to the direct exposure to flames, but also indirectly through smoke inhalation. The Mediterranean basin with its hot and dry summers is a hotspot for such devastating events. The situation has further been aggravated in recent years by climate change as well as a growing and aging population in the region. To assess the health impacts due to short-term exposure to air pollution created by the 2021 summer wildfires in the eastern and central Mediterranean basin, we used a regional-scale chemistry transport model to simulate concentrations of major air pollutants such as fine particulate matter with an aerodynamic diameter less than $2.5 \mu\text{m}$ ($\text{PM}_{2.5}$), SO_2 , NO_2 , and O_3 - in a fire and a no-fire scenario. Elevated short-term exposure of the population to air pollutants are associated with excess all-cause mortality using relative risks (RRs) for individual pollutants from previously published meta-analyses. Our estimates indicate that the short-term exposure to wildfire-caused changes in O_3 accounted for 289 (95% CI:214-364) excess deaths in total over the entire region of investigation during the wildfire season between mid-July to early October 2021. This is followed by 87 (95% CI: 56 - 118) excess deaths due to elevated $\text{PM}_{2.5}$ exposure, rendering the health effect of increased O_3 from wildfires larger than the effect of increased $\text{PM}_{2.5}$. We attributed this largely to the spatially more widespread impact of wildfires on O_3 . Our study concludes with a discussion on uncertainties associated with the health impact assessment based on different air pollutants.

1 Introduction

Air pollution is a central issue in public health globally given its well-documented association with adverse health effects (Brunekreef and Holgate, 2002; Anenberg et al., 2010; Lelieveld et al., 2020). Exposure to air pollution, both long-term and short-term, is estimated to cause millions of premature deaths and lost years of healthy life each year (WHO, 2021). Air pollution caused by wildfires is becoming a subject of increasing concern due to the higher toxicity associated with its chemical composition (Naeher et al., 2007; Wegesser et al., 2009).



Wildfires emit large amounts of particulate matter (PM), nitrogen oxides (NO_x), as well as carbon monoxide (CO) and volatile organic compounds (VOCs) into the atmosphere (Schneider and Abbatt, 2022). Wildfires increase PM levels through direct emission of particles as well as the formation of secondary PM from the oxidation of the emitted ammonia (NH₃), SO₂, NO_x, and VOCs. These species are oxidized to less volatile sulfates, nitrates, and secondary organic aerosols, respectively, which then condense onto pre-existing particles or form new ones (Kroll et al., 2020). Some VOCs are in part emitted with semi- and low volatility and will condense onto particles upon dilution and cooling without the need of further photochemical reactions.

Increased NO_x and VOCs from wildfires lead to additional ozone (O₃) formation downwind through photo-chemical reactions (Jaffe and Wigder, 2012), as well as increases in the concentrations of the hydroxyl radical (OH). The reader is referred to Xu et al. (2021) for a summary of observational evidence from a large recent airborne field campaign. Both increases result in an increased atmospheric oxidation capacity, which in turn further favors the formation of secondary PM (Hobbs et al., 2003; Akagi et al., 2011).

Smoke from wildfires can travel large distances and affect the health of a much larger population than the effects of the actual fire (Bencherif et al., 2020). Most recently, prominent examples are wildfires in the Amazon in 2019 (Butt et al., 2021), Australia in 2019-2020 (Graham et al., 2021), and those in the United States in recent years (Burke et al., 2021; Kramer et al., 2019; Xie et al., 2020). It has been shown that frequency and magnitude of fire events are increasingly affected by human activities and have been exacerbated by climate change (Bowman et al., 2020; Jolly et al., 2015).

Globally, wildfire smoke is estimated to cause more than 330,000 premature deaths each year during 1997 - 2006 (Johnston et al., 2012). This is traditionally considered to be dominated by the effects of inhaling (fine) particulate matter (e.g., Fann et al., 2018). As climate change worsens, together with updated evidence of impacts of wildfires on human health (Chen et al., 2021; Haikerwal et al., 2015), wildfires are projected to result in increased human and material losses in the near future (Xu et al., 2020). Furthermore, wildfires can lead to a higher susceptibility to other (respiratory) diseases, with reports of an amplified risk of COVID-19 cases and deaths in wildfire seasons (Zhou et al., 2021; Schwarz et al., 2022) as a prime example. This may imply that previous studies may systematically underestimate the actual health impacts of wildfires, as they fail to adequately address the underlying relationship between wildfires and other environmental and health concerns.

Historically, the Mediterranean basin which is characterized by hot and dry summers has been negatively affected by wildfires. With increased warming and declining precipitation, the Mediterranean basin is expected to experience an increase in the frequency and scale of wildfires (Ruffault et al., 2020; Cos et al., 2021). Meanwhile, the rapid population growth in some countries (e.g., Egypt, Israel, and Tunisia) and an ageing population in others (e.g., southern European countries) renders the Mediterranean basin ever more vulnerable to the unfavorable consequences of climate change (Linares et al., 2020). All this demands a comprehensive assessment of population exposure to wildfire-caused air pollution.

In this context, the 2021 summer wildfires in the eastern and central Mediterranean basin serve as an indicator of future wildfire impacts. In our work we used this case to assess the health impacts due to short-term exposure to air pollution from wildfire smoke. An online-coupled atmospheric chemistry transport model was employed to simulate concentrations of major air pollutants – fine particulate matter with a diameter of 2.5 μm or less (PM_{2.5}), SO₂, NO₂, and O₃ – in a fire and a no-fire



scenario. Elevated short-term exposure to air pollutants are associated with excess all-age all-cause mortality using relative risks (RRs) for individual pollutants based on previously published meta-analyses. We estimated the excess mortality attributable to wildfires for the entire region and the countries included, with detailed discussions on the uncertainties associated with the estimates.

60 2 Data and Methods

2.1 Study area and period

The study area covers 18 countries and regions in the central and eastern Mediterranean basin with a total population of 334.62 million in 2020, of which over half resides along its coastal areas and hydrological basins (World Bank, 2020). For a full list of countries included in the study and their respective populations, please refer to Tab. A1 in Appendix A.

65 We used the UN WPP-Adjusted population count GPWv4 dataset of the Gridded Population of the World, Version 4 (GPWv4) data from the Socioeconomic Data and Application Center (SEDAC) for 2020 (CIESIN, 2018) to describe the spatial population distribution. From the original resolution of 2.5 arc-minute (approx. 5 km) data were aggregated to a grid of about 20 x 20 km² to be consistent with the configuration of the numerical model employed in this study (Fig. 1a).

70 The study period was 15 July - 02 October 2021, covering the major wildfire events in summer 2021. The severe dry conditions and heatwaves prevailing in this period have resulted in many intense and long-lasting wildfires across the region, emitting large amounts of air pollutants, e.g., PM, NO_x, and SO₂ into the atmosphere (CAM5, 2021). According to the European Forest Fire Information System (EFFIS) (EFFIS, 2021), there were more than 1800 wildfires with a burnt area of 10 hectares or larger occurring in the region and period of this study, which burnt 589,400 hectares area in total. This makes 2021 the second worst fire season in the European Union since 2000, surpassed only by 2017 (San-Miguel-Ayanz et al., 2022). As shown in Fig. 1
75 b-c, the worst hit countries include Turkey, Greece, Italy, Tunisia, and in the Balkans (e.g., Albania, Montenegro, and North Macedonia).

2.2 Model description

Concentrations of PM_{2.5}, O₃, NO₂, and SO₂ were simulated using the Weather Research and Forecasting model coupled to Chemistry (WRF-Chem) model (version 4.2.1), a fully online-coupled regional atmospheric chemistry model (Grell et al.,
80 2005). The Model for Ozone and Related chemical Tracers, version 4 (MOZART-4) gas-phase chemistry mechanism (Emmons et al., 2010) with considerable updates to the chemistry of VOCs (Knote et al., 2014) was used to predict trace gas concentrations. Aerosol characteristics were simulated with the 4 size-bin implementation of the MOSAIC aerosol module (Zaveri et al., 2008). This includes a simplified formulation of secondary organic aerosol formation (Hodzic and Jimenez, 2011), including that from wildfires.

85 Analyses interlaced with hourly forecasts from the Global Forecasting System (GFS) of the National Centers for Environmental Prediction (NCEP) made available through the NOAA Operational Model Archive and Distribution System (NOMADS,

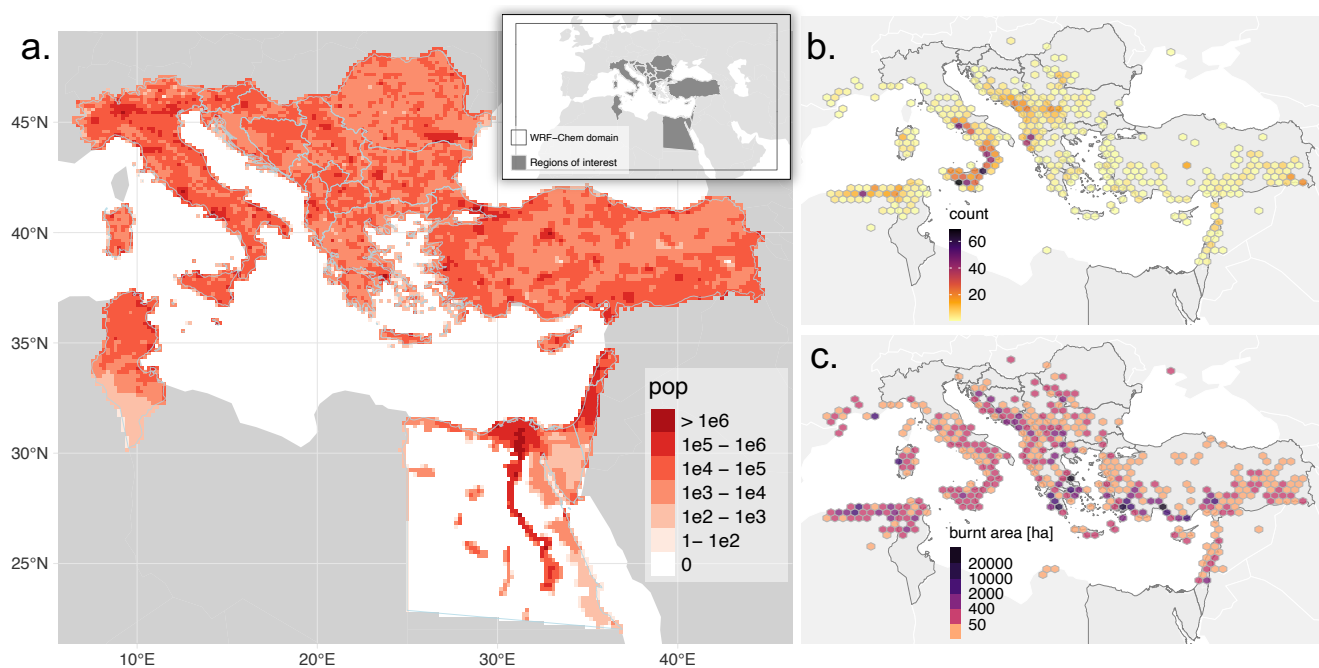


Figure 1. The study region overlaid with the population data and wildfire statistics. a) Population counts per country from the Gridded Population of the World, Version 4 (GPWv4) data for 2020 (CIESIN, 2018). The light gray rectangle in the inset demarcates the simulation domain. b-c) the total number of wildfires and total burnt areas, respectively, based on the European Forest Fire Information System (EFFIS) data between 15-July and 02-October, 2021. Countries included in the study are outlined in black.

(Rutledge et al., 2006)) were used as initial and boundary conditions for meteorological variables. Simulations from the Whole Atmosphere Community Climate Model (WACCM) model created by the Atmospheric Chemistry Observations & Modeling Laboratory (ACOM) of the National Center of Atmospheric Research (NCAR) served to provide initial and boundary conditions for trace gases and particles (<https://www.acom.ucar.edu/waccm/>, last accessed 02.03.2022).

The EDGAR v5.0 dataset was used for prescribing anthropogenic emissions (Crippa et al., 2021). The model simulations also considered biogenic emissions from plants (Guenther et al., 2006), desert dust (LeGrand et al., 2019) and sea spray (Gong, 2003). Emissions of trace gases and particles from wildfires were included by the Fire INventory from NCAR (FINN) model (Wiedinmyer et al., 2011) in version 1.5, based on daily observations by the MODIS and VIIRS satellite instruments.

95 The plume-rise scheme used is originally based on Freitas et al. (2007). Archer-Nicholls et al. (2015) have coupled it to the MOSAIC aerosol scheme also used in this work, and evaluated it for biomass burning episodes in Brazil. As part of Knote et al. (2014) and Knote et al. (2015) we made sure to extend this coupling to the improved MOZART gas-phase mechanism and MOSAIC scheme described therein. Secondary organic aerosol formation from biomass burning is considered using the observationally-constrained approach described in (Hodzic and Jimenez, 2011).



100 This model configuration has already been evaluated in different regions of the world and used to simulate the concentrations of various air pollutants (Graham et al., 2021; Butt et al., 2021).

The model domain in this study covers the central and eastern Mediterranean basin (24.54° W - 63.54° E, 16.95 - 56.57° N) at 20 km horizontal resolution, with 33 vertical levels (up to 10 hPa) (Fig. 1a). Model meteorology was re-initialized every 48 hours with meteorological analyses from GFS, and then allowed to be evolved freely within the boundary layer. Above
105 the boundary layer, grid nudging was performed to keep mesoscale features in line with GFS analyses. Aerosol and trace gas fields were carried over without re-initialization between runs. This setup had been developed previously to strike an optimal balance between realistically considering aerosol-radiation and aerosol-cloud interactions whilst also keeping in line with actual weather development (Im et al., 2015b).

Two simulations, a fire and a no-fire scenario, were performed to quantify the contribution of wildfires to concentrations of
110 major air pollutants.

2.3 Model evaluation

We used the European Environmental Agency (EEA)'s AirBase air quality (including PM_{2.5}, O₃, NO₂, and SO₂) time series data sets (E1a & E2a) to evaluate the WRF-Chem simulations. AirBase data reported by EEA's member states are provided either at hourly or daily intervals. Based on type and location, AirBase stations are classified as rural (including "rural-remote",
115 "rural-regional"), sub-urban (including "rural-nearcity"), and urban. All AirBase data are quality-checked and flagged with different levels of verification (European Environment Agency, 2021). We only used data with "verification code" equal to 1 (verified) or 2 (preliminary verified) in the metadata, and only from stations where more than 75% of observations were present during the entire period. Nevertheless, the verified data from each AirBase station could still contain outliers which we defined as observations with four standard deviations from its mean over the entire study period and were thus removed. For stations
120 where only daily means were recorded, hourly WRF-Chem values were averaged to daily means for comparison. The data were downloaded using `airbase 0.2.7` python library (<https://pypi.org/project/airbase/>).

For WRF-Chem simulations of each air pollutant, the conventional statistical metrics, such as mean bias (MB), normalized mean bias (NMB), root mean square error (RMSE), and index of agreement (IOA), were calculated (see Tab. B1). In addition, Taylor diagrams (Taylor, 2001) were plotted to compare the model's performance at different locations (rural, suburban, and
125 urban) and averaging frequencies (daily versus hourly mean).

2.4 Health impacts assessment (HIA)

Health impacts of short-term exposure to wildfire-caused air pollution were estimated using a well-established methodology (WHO, 2016). Details of the main steps are given below.



2.4.1 Determination of population exposure to air pollution

130 Population exposure was quantified for each country by calculating the population-weighted concentrations of air pollutants using GWPv4 population data and pollutant concentration data from the WRF-Chem simulations. For country J on day d , its population weighted exposure (C_p) to a pollutant i - say, i is $PM_{2.5}$, is then calculated as:

$$C_{p_{i,J,d}} = \frac{\sum_{j \in J} (pop_j \times c_{i,j,d})}{\sum_{j \in J} pop_j} \quad (1)$$

135 where pop_j is the population counts in any grid cell j pertaining to country J ; $c_{i,j,d}$ is the daily mean air pollutant concentration for $PM_{2.5}$, SO_2 , and NO_2 , whereas the daily maximum 8 hour average (DMA8) is used for O_3 . Population-weighted exposure was computed for both fire and non-fire scenarios, with their difference representing the additional health burden attributable to wildfires.

2.4.2 Estimate of exposure-associated health risk

140 To estimate the exposure-associated health risk, we used both baseline health statistics (here e.g., mortality) and an exposure-response-function (ERF) for individual air pollutants.

The baseline all-cause mortality data in both genders (deaths per annum) for 2019 (the latest year for which data are available) were downloaded from the Global Health Data Exchange (GHDx) (Global Burden of Disease Collaborative Network, 2020). As shown in Fig. A1, the annual mortality rates of countries range from 515 (Israel) to 1791 (Bulgaria) deaths per 100,000 population. To interpolate the annual deaths to the summer months (July-August-September) 2021, we used the multi-annual
145 (2010-2019) average of monthly deaths as proxy. Monthly baseline mortality data are collected by the European Statistical Office (Eurostat) for its member and associated states (Eurostat, 2021). For countries where monthly deaths are not available, e.g., Tunisia, Israel, and Egypt, the monthly mean deaths averaged over all other countries in this region were used as proxy (Fig. A2). Total monthly deaths were then equally distributed to each day of the month. We denoted the daily number of deaths (baseline mortality) as BM_d .

150 An ERF associates the proportional increase in exposure of an air pollutant with the potential adverse health outcomes, typically expressed as relative risk (RR). The RR is a ratio of incidences (e.g., deaths) exposed to air pollution relative to incidences with no exposure. The RR can be estimated either by 1) pre-defined formulas or ranges of values from studies or meta-analyses, or 2) by integrated ERF approaches (Burnett et al., 2014; Cohen et al., 2017). Here, we chose the former approach, adopting RR values for short-term exposure mainly from a recently published systematic review by Orellano et al. (2020). To assess the
155 uncertainty due to different RR estimates, we additionally calculated health impact estimates with RRs from different reviews published before (World Health Organization, 2013; Orellano et al., 2020; Vicedo-Cabrera et al., 2020; Liu et al., 2019). All of these RRs refer to risk of all-cause all-age mortality associated with short-term exposure to air pollutants, and thus no age groups are specified throughout this study. The RRs together with their respective 95% confidence intervals (CIs) are listed in Tab. 1.



	Relative risk (95% CI)	
	PM _{2.5}	O ₃
Orellano et al. (2020)	1.0065 (1.0044–1.0086)	1.0043 (1.0034–1.0052)
WHO (2013)	1.0123 (1.0045–1.0201)	1.0029 (1.0014–1.0043)
Liu et al. (2019)*	1.0068 (1.0059–1.0077)	–
Vicedo-Cabrera et al. (2020)	–	1.0018 (1.0012–1.0024)

* 2-day moving average is used as the exposure metric instead of the daily mean used elsewhere.

Table 1. Relative risks (RR_{Δ10}) with 95% confidence interval (CI) of all-cause mortality associated with short-term exposure to PM_{2.5} and O₃ for an increase of 10 μg/m³ concentration, obtained from selected meta-analysis or multi-city studies published before.

160 Given a linear relationship and an increase of population exposure (ΔC) of 10 μg/m³, the RR is defined as

$$RR = e^{\beta \Delta C}. \quad (2)$$

From Eq. 2 we can back out the coefficient $\beta = \ln RR / \Delta C$ and use it to estimate RR for arbitrary ΔX:

$$RR(\Delta X) = e^{\beta(\Delta X - X_0)}, \quad (3)$$

175 where X_0 is the theoretical minimum risk exposure level, below which no additional risk is assumed. In line with previous works (e.g., Graham et al., 2021; Macintyre et al., 2016), we assume X_0 to be zero. We then derived excess exposures to an air pollutant i emitted from wildfires as the difference in population exposure between the fire and no-fire simulations for a country J :

$$\Delta C p_{i,J} = C p_{i,J}^{fire} - C p_{i,J}^{no-fire} \quad (4)$$

To account for the health impacts on the population level, the population attributable fraction (PAF), defined as the fraction of adverse health outcomes in a population attributable to a specific exposure, is computed using the formula (Mansournia and Altman, 2018):

$$PAF = 1 - \frac{1}{RR} \quad (5)$$

The excess number of deaths attributable to wildfire-caused exposure to air pollutant i for a country J within the entire period of simulation ($E_{i,J}$) is computed as:

$$175 \quad E_{i,J} = \sum_{d=1}^N (BM_{J,d} \times PAF_{i,J}), \quad (6)$$

where $BM_{J,d}$ is the baseline mortality (in number of deaths) of the country J on day d obtained before, and N is the total number of simulation days.



2.4.3 Estimate of uncertainty due to error propagation in excess mortality

Numerous sources of uncertainty exist for such a health impact assessment which have been discussed in details elsewhere
180 (WHO, 2016). In this study, we accounted for the uncertainty arising from the multiplication of baseline mortality and relative risk, both of which are bounded with their respective 95% CIs. Assuming a normal distribution for both data sets ($x \sim \mathcal{N}(\mu, \sigma^2)$), the standard error (SE) of each data is derived as:

$$SE(x) = (CI^{\text{upper}} - CI^{\text{lower}})/(2 \cdot z_{1-0.05/2}), \quad (7)$$

where $z_{1-0.05/2}$ is the 0.975 quantile of the standard normal distribution ($z_{0.975} \approx 1.96$). Then, the variance $\text{Var}(x) = SE(x)^2$.
185 As PAF depends linearly on $1/RR$ (Eq. 5), its SE and variance are linear derivatives of those for $1/RR$.

To quantify the propagation of the uncertainties in the excess mortality estimates, two approaches were used. The first approach is based on the delta method (Ver Hoef, 2012). For the product of baseline mortality and PAF (as in Eq. 6), the joint standard error is calculated below:

$$SE(\hat{x}_1 \hat{x}_2) = \sqrt{\hat{x}_1^2 \text{Var}(\hat{x}_2) + \hat{x}_2^2 \text{Var}(\hat{x}_1) + 2\hat{x}_1 \hat{x}_2 \text{Cov}(\hat{x}_1, \hat{x}_2)}, \quad (8)$$

190 where \hat{x}_1 and \hat{x}_2 represent the expectations of baseline mortality and PAF, respectively. Since baseline mortality and PAF are mutually independent, their covariance $\text{Cov}(\hat{x}_1, \hat{x}_2)$ is zero. Equation 8 can be further shortened as:

$$SE(\hat{x}_1 \hat{x}_2) = \sqrt{\hat{x}_1^2 \text{Var}(\hat{x}_2) + \hat{x}_2^2 \text{Var}(\hat{x}_1)} \quad (9)$$

The 95% confidence intervals for the product $x_1 x_2$ are $\hat{x}_1 \hat{x}_2 \pm 1.96 SE(\hat{x}_1 \hat{x}_2)$.

The second approach is based on the Monte Carlo method. We respectively generated the normally distributed random
195 samples of BM and PAF (with a sample size $N = 1000$) based on the expectations and SEs derived from Eq. 7. The 2.5% and 97.5% percentiles of the sample composed of the element-wise product of BM and PAF designated the lower and upper bounds of the 95% CIs for their product. Repeating for 100 times, the respective mean values of lower and upper bounds quantified the uncertainty for the estimated health impacts.

We used the R software (version 4.1.1) (R Core Team, 2021) and the *sf* package (version 1.0-4) (Pebesma, 2018) to perform
200 the statistical and geo-spatial analyses.

3 Results

3.1 Model validation

Overall, WRF-Chem model performance is on par with recent multi-model inter-comparison studies of air quality models over Europe and North America (Im et al., 2015a, b) and produced accurate estimates for the concentrations of $PM_{2.5}$ and O_3
205 when compared to the AirBase dataset. Table B1 lists the model performance metrics for simulations of each air pollutant. The WRF-Chem model is prone to overestimate the concentration of air pollutants relative to the AirBase observations, except $PM_{2.5}$.



Figures B1-B4 in Appendix B show normalized Taylor diagrams for WRF-Chem simulations of each air pollutant for rural, sub-urban, and urban AirBase stations. The estimates of $PM_{2.5}$ from the WRF-Chem simulations were marginally better for daily mean values than for hourly values when compared to the observations, whereas the performance exhibited no significant difference between stations of different types. The correlation coefficients between the simulated and observed concentrations (on polar axes) range from 0.4 to 0.7 (Fig. B1). By comparison, aggregating hourly observations of O_3 to daily means does not help increase the model performance. On the contrary, the model performance of reproducing the O_3 variation dropped with the mean IOA decreasing from 0.65 to 0.51 (Tab. B2). The type of station does not seem to affect the model performance in estimating O_3 concentration.

The stringent criteria on the data's status of verification have significantly reduced the number of AirBase stations available for model evaluation. The most stations remained for evaluation are located in a limited number of countries (e.g., Italy, Greece, and Bulgaria) and mostly in urban or suburban areas. We note that a perfect simulation, especially of urban stations cannot be expected due to the very different representativeness of station measurements and the 20 km horizontal resolution of WRF-Chem. Furthermore, the uneven spatial spread of stations and the over-representation of certain station type in the model evaluation could further worsen the performance metrics.

3.2 Spatio-temporal patterns of air pollutant concentrations

Figure 2a shows the spatial pattern of the WRF-Chem simulated $PM_{2.5}$ concentration as average over the entire period based on the fire scenario. The mean $PM_{2.5}$ concentrations range from 1 to $150 \mu g/m^3$ across the region. High $PM_{2.5}$ loads are visible in regions that are already heavily polluted even in the absence of wildfires, e.g., in the Po Valley of Italy and the capital region of Serbia.

Figure 2b and c show the spatial pattern of fires-caused $PM_{2.5}$ concentration relative to that under no-fires scenario, and the percentage of $PM_{2.5}$ loads attributable to wildfires. The increased $PM_{2.5}$ concentrations caused by wildfires are mainly observed in the Balkans, Greece, and southern Italy, coinciding with the distribution of fire events within the simulation period (Fig. 1 b-c).

Diurnally, the $PM_{2.5}$ concentration is subject to the mixing layer height (Fig. C1). The daytime troughs reflect strong turbulent exchange and dilution within the mixed layer, whereas stable nighttime boundary layer enables $PM_{2.5}$ to accumulate (Manning et al., 2018).

Figure 3 shows the spatial pattern of WRF-Chem-simulated O_3 concentrations. In contrast to the $PM_{2.5}$ pattern, the O_3 concentration demonstrates an upward gradient from urban agglomerations to non-urban surroundings, indicating titration by large urban NO_x emissions (Sillman, 1999).

The overall effect of the wildfires on O_3 is more widespread due to the longer atmospheric formation and lifetime of O_3 , and is hence visible on a country level (Fig. C2). Interestingly, the border region between Bulgaria and Romania is observed to be one of regions most affected by wildfire-caused O_3 pollution, which may be accounted for by the location-specific topography and the fact that it is located downwind of a number of fires that occurred during the season. The region on the lower Danube

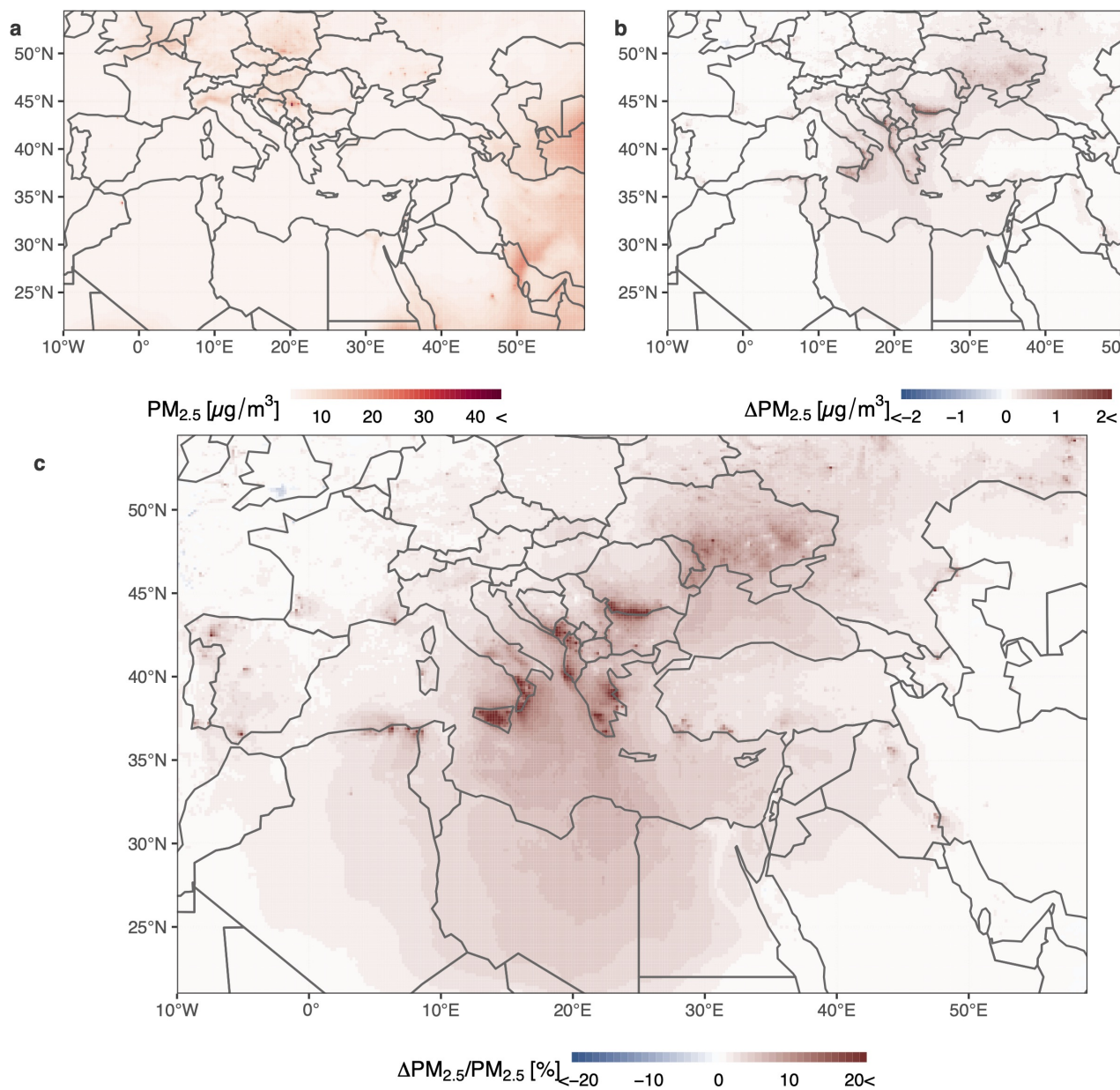


Figure 2. Spatial pattern of multi-month mean PM_{2.5} concentrations simulated by WRF-Chem under the fires scenario and the difference relative to the no-fires scenario. a) PM_{2.5} concentration simulated under fires scenario, b) PM_{2.5} concentration under fires scenario relative to no-fires ($\Delta\text{PM}_{2.5} = \text{PM}_{2.5}^{\text{fires}} - \text{PM}_{2.5}^{\text{no-fires}}$), c) PM_{2.5} concentration attributable to wildfires in percentages ($\Delta\text{PM}_{2.5}/\text{PM}_{2.5}^{\text{fires}}$). The increased PM_{2.5} concentrations caused by wildfires are mainly observed in the Balkans, Greece, and southern Italy, whereas the regions with high background industrial pollution (hot spots in the panel a) show a slight decrease in ambient PM_{2.5} concentration.



plain is further bounded by the Carpathian Mountains to the North and West and by the Balkan Mountains to the South which forms a semi-closed topographical feature, channeling and concentrating air pollutants emitted from wildfires.

Figure C2 shows the diurnal pattern of O_3 , which is determined by the presence of sunlight, as O_3 is formed primarily by photo-chemical reactions.

245 Figures C3 and C4 show the temporal pattern of NO_2 and SO_2 concentrations respectively, while their spatial patterns are shown in Fig. D1 and D2. The hot spots of NO_2 and SO_2 are principally found in cities, along artery roads and shipping routes, and in large point sources like power plants and oil and gas refineries (Fig. D2a). By comparison to O_3 , increases of NO_x and SO_2 are less visible on a country level (Fig. C3 and C4), as their effects are limited locally due to their relatively shorter atmospheric lifetime, and the dilution into comparatively high pre-existing background concentrations. Similar to $PM_{2.5}$, the
250 diurnal pattern of NO_2 and SO_2 concentration is determined by the mixing layer height. Meanwhile, we found the expected increases in the concentration of OH in the fire simulation (Fig. D3), which will lead to additional formation of secondary PM.

3.3 Population-weighted exposure to major air pollutants

Figure 4 depicts the multi-month mean population-weighted exposures to four air pollutants under fires (red dots), and no-fires (blue dots) scenarios. For the time series of population-weighted exposures to air pollutants in individual countries, please
255 refer to Figs. E1-E4. Countries in Balkans (e.g., Albania, Serbia, North Macedonia, Montenegro, and Bulgaria), Romania, and Greece are among the most affected regions by the wildfire-caused $PM_{2.5}$ and O_3 pollution, in line with the incidence of wildfires within the simulation period. By contrast, wildfires have not resulted in a discernible increase of population exposure to NO_2 and SO_2 within the simulation period and region. Therefore, we estimate the wildfire-associated health impacts based primarily on $PM_{2.5}$ and O_3 exposures.

260 3.4 Wildfire-caused excess mortality

Figure 5 a-b shows the excess number of deaths for each country estimated based separately on RRs of short-term exposure to $PM_{2.5}$ and O_3 obtained from the meta-analysis by Orellano et al. (2020). The 95% CIs are estimated using the Monte Carlo method described in Section 2.4.3. The exact number of deaths for each country and the entire region are available in Tab. A2.

265 Countries with a large population such as Italy and Egypt are estimated to have higher excess deaths due to short-term exposure to wildfire-caused $PM_{2.5}$ and O_3 . Although wildfires are not frequently observed in Egypt (Fig. 1 b), the deaths estimated are significant and can be attributed to the transport of air pollution caused by wildfires occurring elsewhere in the Mediterranean basin, clearly indicating the widespread impact of wildfires on the whole Mediterranean basin.

Figure 5c-d sum up the excess deaths attributable to short-term exposure to $PM_{2.5}$ and O_3 in the entire region of investigation during mid-July to early October, 2021, based on RRs suggested by different publications. Based on the RR values from
270 Orellano et al. (2020), there are 87 (95% CI: 56-118) deaths attributable to the short-term exposure to wildfire-caused $PM_{2.5}$. This estimate is close to the one based on Liu et al. (2019) – 90 (95% CI: 73-108). In comparison, 164 (95% CI: 57-270) excess deaths are estimated based on the RRs from World Health Organization (2013), albeit with a pronounced range of uncertainty.

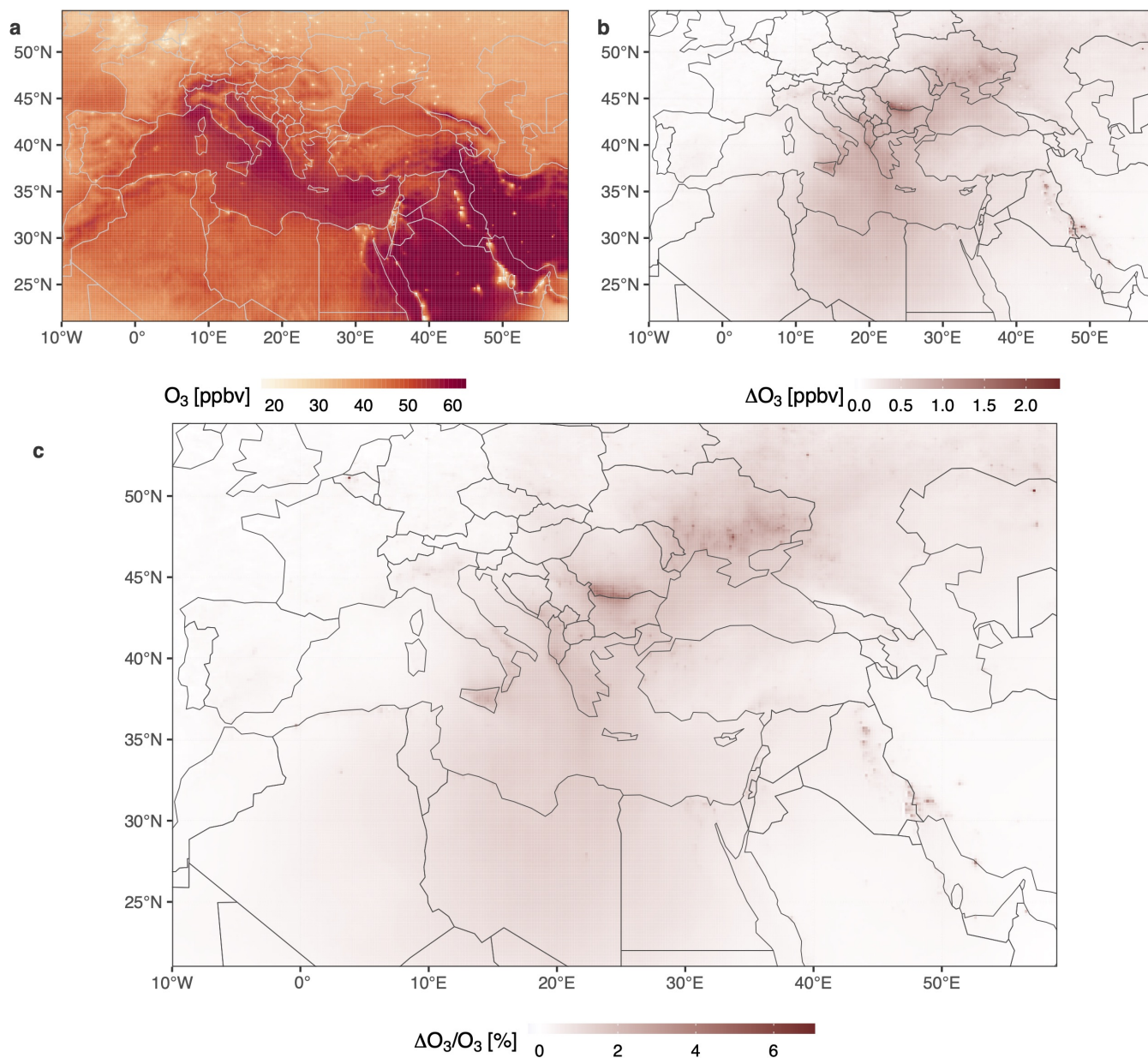


Figure 3. Spatial pattern of multi-month mean O_3 concentrations simulated by WRF-Chem under the fires scenario and the difference relative to the no-fires scenario. a) O_3 concentration simulated under fires scenario, b) O_3 concentration under fires scenario relative to no-fires ($\Delta O_3 = O_3^{\text{fires}} - O_3^{\text{no-fires}}$), c) O_3 concentration attributable to wildfires in percentages ($\Delta O_3 / O_3^{\text{fires}}$).

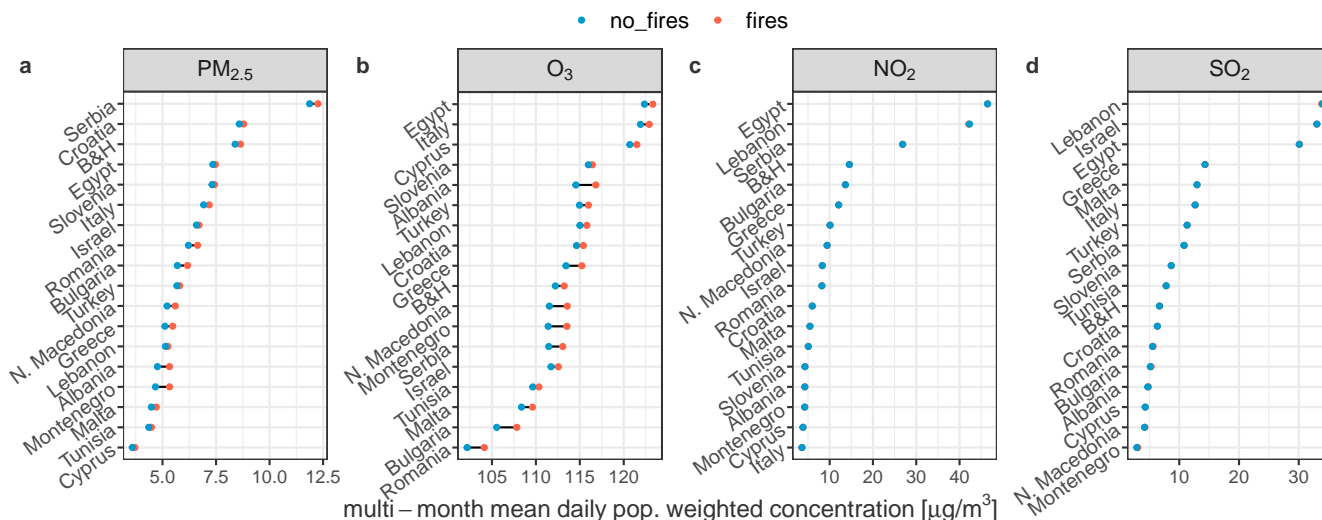


Figure 4. Multi-month daily mean population weighted concentration of air pollutants, fires scenario versus no-fires scenario. For pollutants other than O₃, the daily mean concentration is used, while the daily mean 8-h average (DMA8) for O₃. B&H stands for Bosnia and Herzegovina.

With regard to the excess deaths attributable to short-term O₃ exposure, 289 (95% CI: 214 - 364) excess deaths are estimated based on Orellano et al. (2020), remarkably exceeding the estimates based on World Health Organization (2013) (195, 95% CI: 93 - 297) and Vicedo-Cabrera et al. (2020) (121, 95% CI: 77 - 166).

4 Discussion

In general, we found larger health impacts due to wildfire-associated exposure to O₃ than to PM_{2.5}. As the relative risk of exposure to O₃ is actually lower than that of PM_{2.5} exposure (Tab. 1), the reason for this surprising finding is shown to be attributable to the more widespread impact of wildfires on O₃ due to a longer overall atmospheric lifetime of O₃ (about a month) in contrast to PM_{2.5} (days to weeks) (Task Force on Hemispheric Transport of Air Pollution (TF HTAP), 2010; Liang et al., 2018).

We refrained from deciding whether the excess deaths attributed to each pollutant can simply be added up to generate a synthesized estimate of health impacts as a result of the simultaneous exposure to multiple pollutants. The solution to this problem demands a thorough knowledge of correlations between health impacts of each pollutant (World Health Organization, 2013), and analysis of their confounding effects (Anderson et al., 2012; Bell et al., 2007). Although several statistical methods have been proposed to address the multi-collinearity issues in concurrent exposure (Stafoggia et al., 2017; Wei et al., 2020), they have not been widely adopted, including in studies that underlay the systematic reviews by Orellano et al. (2020); World Health Organization (2013). Therefore, results based on these reviews should be interpreted with caution, as confounders were

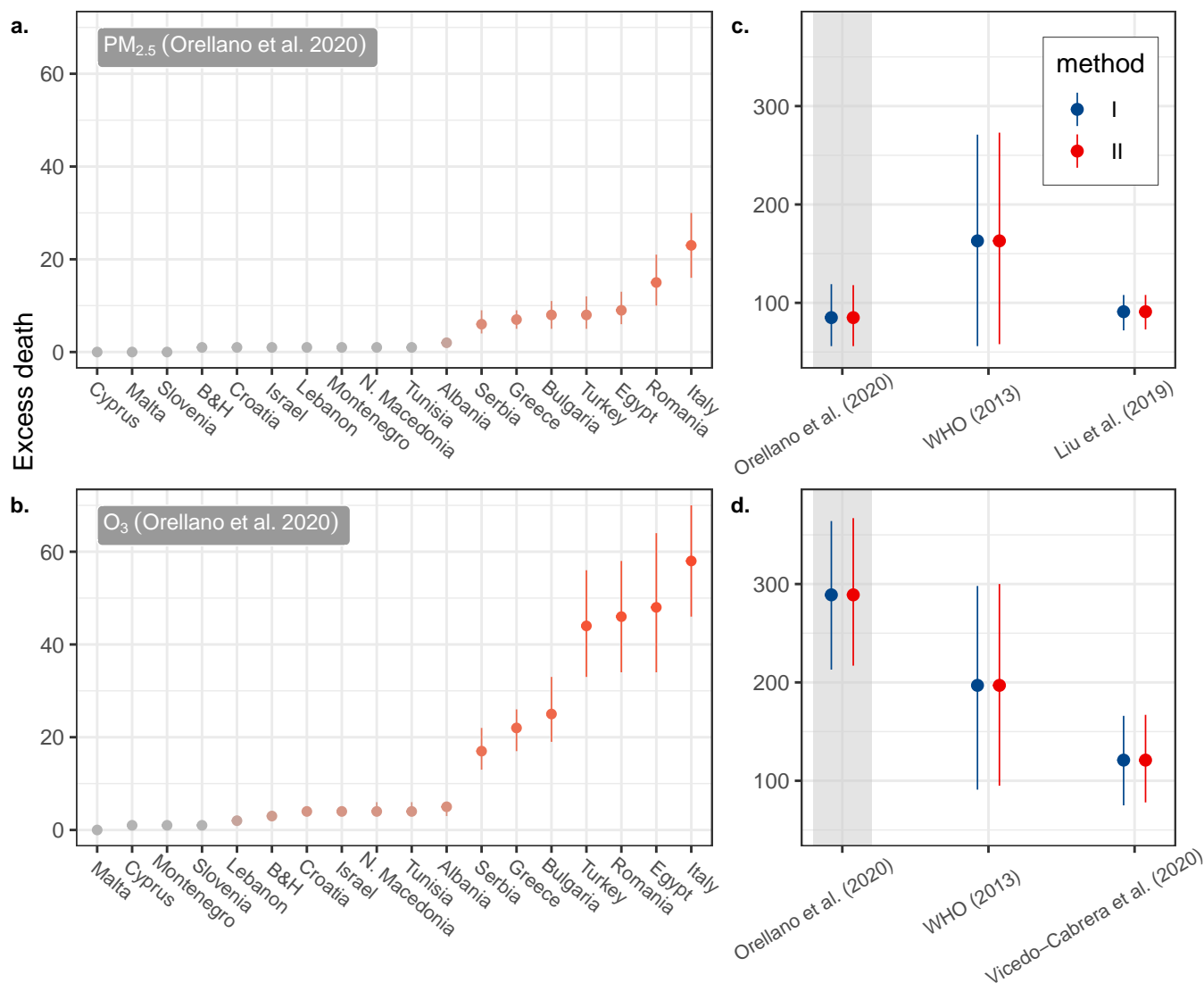


Figure 5. Excess deaths with uncertainties estimated based on relative risks of short-term exposure to PM_{2.5} and O₃. a-b) wildfire-caused excess deaths with 95% CIs for each country included in the study based on relative risk values for short-term PM_{2.5} and O₃ exposures from Orellano et al. (2020). c) Total excess deaths estimated using relative risk values from different meta-analyses (with the vertical gray band indicating the ones used in panels a and b), based on short-term exposure to PM_{2.5} and O₃, respectively. The 95% CIs were estimated using the Delta (I) and Monte Carlo (II) methods. In Liu et al. (2019), the 2-day moving average of daily mean PM_{2.5} concentrations was used to estimate the excess mortality.



not adequately adjusted for. As a result, it remains inconclusive whether to sum up the death estimates based on different air
290 pollutants. All the aforementioned aspects can substantially affect the outcomes of the health impact assessment, making it
difficult to narrow down the range of real health impacts attributable to wildfire-caused air pollution beyond what we have
shown here.

The WRF-Chem model enables a temporally and spatially resolved exposure estimate, while accounting for primary emis-
sions from wildfires and subsequent secondary chemical and physical processes. To date, the health impact assessment of air
295 pollution is limited to a subset of its proxies (e.g., $PM_{2.5}$, NO_2 , O_3), with their exposures being measured by mass. This sim-
plification is increasingly challenged by mounting evidence that the toxicity of air pollution depends to a large extent on the
chemical composition and atmospheric ageing rather than the mass itself. Especially, several studies point to the fact that PM
arising from wildfires is more toxic to lungs compared with PM from normal ambient air (Wegesser et al., 2009; Dong et al.,
2017; Xu et al., 2020). In the future, the ability of such modeling systems to estimate further harmful trace gases as well as
300 chemically-speciated PM may help overcome such limitations, leading to better association and causation in epidemiological
analysis.

We have accounted for uncertainties arose from the baseline mortality data and ERFs. However, we have to acknowledge
several sources of uncertainty that have not been taken into account within this study. The health impacts estimated within the
study is limited to the excess mortality due to short-term exposure to air pollution aggravated by wildfires. This does not take
305 into account direct loss of life and hospital admissions caused by the direct exposure to radiant heat/smoke/flames of wildfires,
nor the consequent impairment of life quality.

Though broadly validated, the WRF-Chem model results contain a certain degree of uncertainty in exposure estimation. The
uncertainty could originate from the model configuration (e.g., the horizontal and vertical grid setting of simulation domain,
and parameterization schemes adopted to account for the atmospheric physics and chemistry), model inputs (e.g., boundary
310 meteorology, and inventories on anthropogenic and fires-associated emissions), as well as the representation of particles and
their size distribution within the model (Im et al., 2018).

The horizontal resolution of 20 km set in the WRF-Chem simulation may be too coarse to represent urban areas and their
related impacts on the dispersion and transformation of air pollutants sourced from wildfires. This may lead to an underes-
timation of particular matter and an overestimation of O_3 in urban areas (Im et al., 2015a). Due to its complex nature, the
315 model-associated uncertainty has not been addressed in the study. However, this can be the focus of future research.

A further source of uncertainty lies in the ERFs adopted for the health impact assessment. Although ERFs derived from meta-
analysis are thought to deliver less biased and impartial evidence on the association between exposure and health outcomes,
they are prone to several other biases, e.g., publication bias and language bias, which may distort the evidence (Page et al.,
2021). Meanwhile, current ERFs are based primarily on exposures measured in urban or suburban settings, making the exposure
320 estimate for rural areas more prone to misclassification errors. Even though rural areas normally exhibit a lower level of PM and
are thought to be less polluted, the rural air can demonstrate similar levels of cellular oxidative potential as in cities, due largely
to more toxic chemicals emitted from agriculture activities (Wang et al., 2022). On the other hand, neither measurements by



monitoring networks nor model simulations, are able to reproduce the individual exposure which is subject to a variety of factors such as personal behavior, socioeconomic status, and preexisting health conditions (Evangelopoulos et al., 2020).

325 Subject to the RR value used, the estimated health burdens in association with short-term O₃ exposure differ markedly. The RR value suggested by Orellano et al. (2020) was attained through a systematic review based on the most recent studies, serving as an update of the previous report by World Health Organization (2013). In comparison, Vicedo-Cabrera et al. (2020) suggested a considerably lower RR for short-term exposure to O₃, resulting in a smaller estimate of excess deaths. As both studies differ in methodology and underlying database leading to the RRs, the reason for the discrepancy remains unexplored
330 and is beyond the scope of this study.

In all estimates, the choice of methods for quantifying the uncertainty of excess deaths exhibits a minor impact on the results. The Delta method, compared with the Monte Carlo method, is prone to slightly underestimate the lower and upper bounds of the 95% CI.

We excluded the health impact assessment based on short-term exposure to PM₁₀, another widely used proxy indicator for
335 air pollution, to avoid double counting of the PM-associated health effects. This is based on the fact that PM_{2.5} accounts for an overwhelming proportion of PM-associated health effects (Lu et al., 2015; Liu et al., 2019).

5 Conclusions

We have assessed the health impacts due to the short-term exposure to air pollution caused by wildfires over the eastern and central Mediterranean basin in the summer 2021. The exposures were estimated using a fully coupled atmospheric chem-
340 istry model under fire and no-fire scenarios, respectively, while the consequent health impacts were quantified based on well-established ERFs from selected systematic reviews. We estimated that the 2021 summer wildfires result in an excess number of deaths ranging from approximately 87 (95% CI: 56-118) to 289 (95% CI: 214 - 364), depending on the targeted pollutants which population are exposed to, and on their respective ERFs. Future work is needed to reduce the uncertainties resulted from estimates of both exposures and health effects, while simultaneously augmenting the computational performance of the
345 methodology used. To this end, it is worth further exploring the ensemble or hybrid approaches which combine both physically based atmospheric chemistry transport models and computationally efficient statistical models (Im et al., 2018; Conibear et al., 2021; Di et al., 2019; Shtein et al., 2019; Hough et al., 2021).



Appendix A: Population and mortality data

Table A1. Countries included in the study and their respective population in 2020, obtained from the World Bank's World Development Indicators database (World Bank, 2020).

	name	ISO-2	ISO-3	pop. (in Mio.)
1	Albania	AL	ALB	2.84
2	Bosnia and Herzegovina (B&H)	BA	BIH	3.28
3	Bulgaria	BG	BGR	6.93
4	Cyprus	CY	CYP	1.21
5	Egypt	EG	EGY	102.33
6	Greece	GR	GRC	10.72
7	Croatia	HR	HRV	4.05
8	Israel	IL	ISR	9.22
9	Italy	IT	ITA	59.55
10	Lebanon	LB	LBN	6.83
11	Montenegro	ME	MNE	0.62
12	North Macedonia (N. Macedonia)	MK	MKD	2.07
13	Malta	MT	MLT	0.53
14	Romania	RO	ROU	19.29
15	Serbia	RS	SRB	6.91
16	Slovenia	SI	SVN	2.10
17	Tunisia	TN	TUN	11.82
18	Turkey	TR	TUR	84.34
	SUM			334.62



Table A2. Excess number of deaths for individual countries and the entire region estimated based on wildfires-caused PM_{2.5} and O₃ loads within the simulation period, with 95% confidence intervals (CIs) in brackets. The methods I and II refer to the Delta and Monte Carlo methods for the uncertainty estimation, respectively, which are described in details in Section 2.4.3 in the main text. The Delta method slightly underestimate the excess deaths compared to the Monte Carlo method. However, the difference is not significant.

Country	PM _{2.5}		O ₃	
	Method I	Method II	Method I	Method II
Albania	2 [1,2]	2 [1,2]	5 [3,6]	5 [3,6]
B&H	1 [1,2]	1 [1,2]	3 [2,4]	3 [2,4]
Bulgaria	8 [5,11]	8 [5,11]	25 [18,32]	25 [19,33]
Croatia	1 [1,2]	1 [1,2]	4 [3,5]	4 [3,5]
Cyprus	0 [0,0]	0 [0,0]	1 [0,1]	1 [0,1]
Egypt	9 [6,13]	9 [6,13]	48 [33,63]	48 [34,64]
Greece	7 [5,9]	7 [5,9]	22 [17,26]	22 [17,26]
Israel	1 [0,1]	1 [0,1]	4 [3,4]	4 [3,4]
Italy	23 [16,31]	23 [16,30]	58 [46,70]	58 [46,70]
Lebanon	1 [0,1]	1 [0,1]	2 [2,3]	2 [2,3]
Malta	0 [0,0]	0 [0,0]	0 [0,0]	0 [0,0]
Montenegro	1 [0,1]	1 [0,1]	1 [1,2]	1 [1,2]
N. Macedonia	1 [1,2]	1 [1,2]	4 [3,6]	4 [3,6]
Romania	15 [10,21]	15 [10,21]	46 [34,57]	46 [34,58]
Serbia	6 [4,9]	6 [4,9]	17 [12,22]	17 [13,22]
Slovenia	0 [0,0]	0 [0,0]	1 [1,1]	1 [1,1]
Tunisia	1 [1,2]	1 [1,2]	4 [3,6]	4 [3,6]
Turkey	8 [5,12]	8 [5,12]	44 [32,56]	44 [33,56]
SUM	87 [56,118]	87 [57,118]	289 [214,364]	289 [218,366]

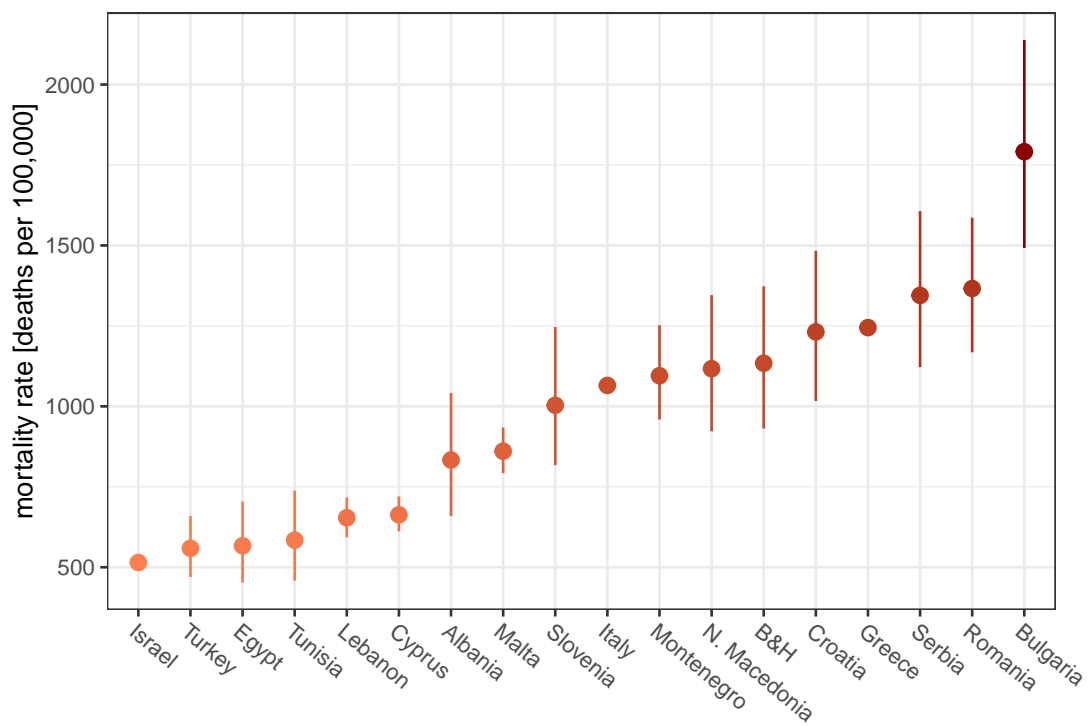


Figure A1. Baseline all-cause mortality in both genders 2019 for each country with 95% confidence intervals, downloaded from the Global Health Data Exchange Global Burden of Disease Collaborative Network (2020). The annual mortality rates of countries range from 515 (Israel) to 1791 (Bulgaria) deaths per 100,000 population. B&H stands for Bosnia and Herzegovina.

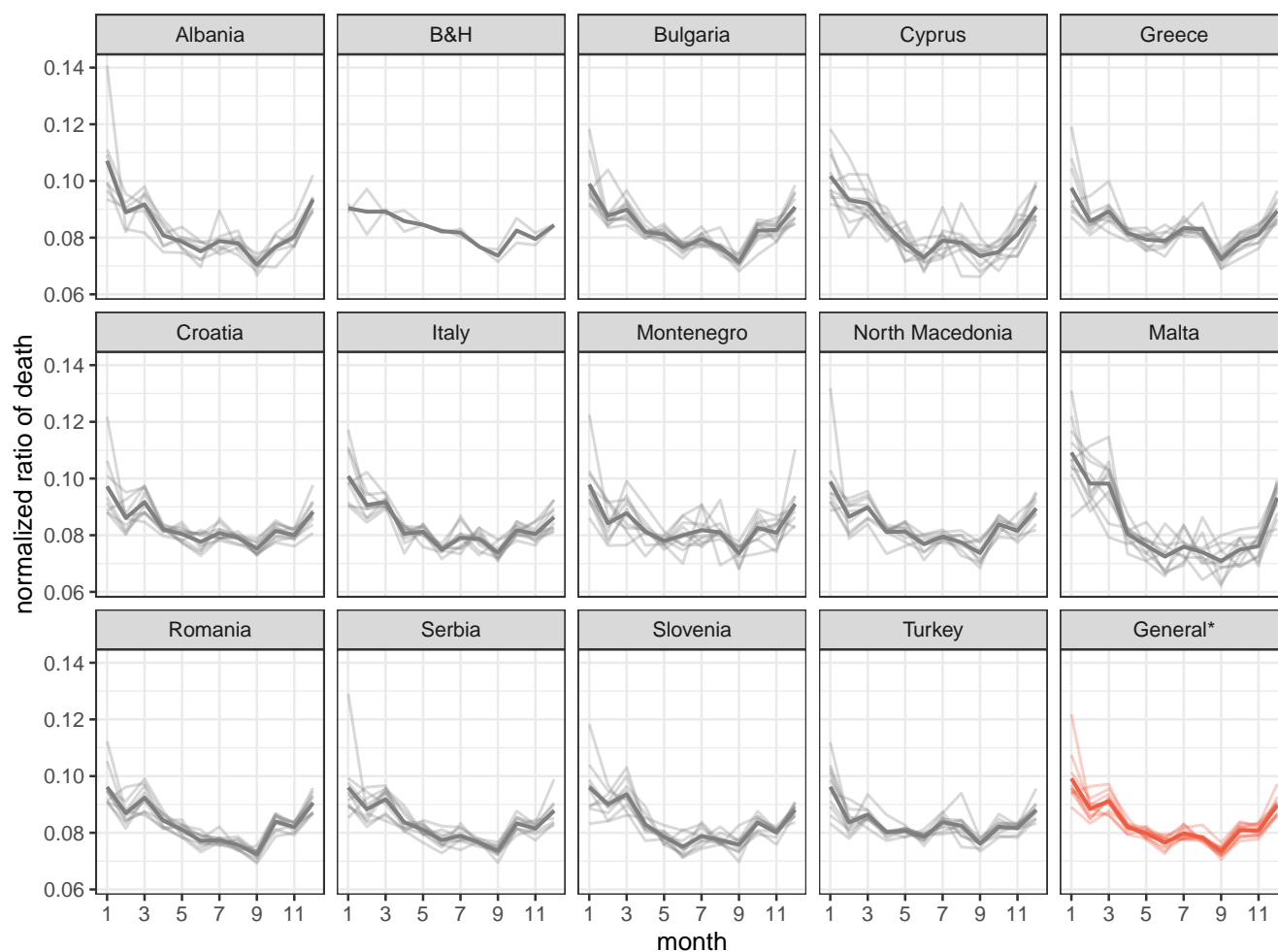


Figure A2. Multi-annual monthly mean normalized ratio of death averaged across 2010-2020 (dark gray, bold), underlaid with data for individual years (light gray) downloaded from Eurostat (2021). For countries where monthly death data are not available (e.g., Egypt, Israel, Lebanon), the monthly mortality is calculated based on the regional mean value averaged across all other countries (General*).



Appendix B: Model performance of WRF-Chem

Table B1. Performance metrics used in this study for model evaluation. N is the number of observations. M_i and O_i are modelled and observed values, respectively, while the overline indicates the arithmetic mean. σ represents the standard deviation. Normalized standard deviation and correlation coefficient (Pearson's) are presented in a Taylor diagram (Figs. B1-B4). The index of agreement (IOA) is a standardized measure of the degree of model prediction error with a range between 0 and 1 (Willmott, 1981). A value of 1 indicates a perfect match, while 0 the worst.

Metric	Expression	Range
Mean Bias (MB)	$\frac{1}{N} \sum_{i=1}^N (M_i - O_i)$	$(-\infty, +\infty)$
Normalized Mean Bias (NMB)	$\sum_{i=1}^N (M_i - O_i) / \sum_{i=1}^N O_i$	$(-\infty, +\infty)$
Normalized standard deviation	σ_M / σ_O	$[0, +\infty)$
Root mean square error (RMSE)	$\sqrt{\frac{1}{N} \sum_{i=1}^N (M_i - O_i)^2}$	$[0, +\infty)$
Correlation coefficient (ρ)	$\sum_{i=1}^N (M_i - \bar{M})(O_i - \bar{O}) / (\sigma_M \sigma_O)$	$[0, 1]$
centred RMSE	$\sqrt{\sigma_M^2 + \sigma_O^2 - 2\sigma_M \sigma_O \rho}$	$[0, +\infty)$
Index of agreement (IOA)	$1 - \frac{\sum_{i=1}^N (M_i - O_i)^2}{\sum_{i=1}^N (M_i - \bar{O} + O_i - \bar{O})^2}$	$[0, 1]$



Table B2. Performance metrics of the WRF-Chem for simulating daily and hourly mean concentrations of individual air pollutants from AirBase stations. Metrics shown are mean value of mean bias (MB), normalized mean bias (NMB), root mean square error (RMSE), and index of agreement (IOA), with their standard deviations in parentheses. Based on IOA, the model performs relatively well to reproduce the variations of PM_{2.5} and O₃. It has a mediocre performance for NO₂, while it scores lowest in estimating SO₂. The model shows a generally positive bias relative to the AirBase observations for most air pollutants considered here except PM_{2.5}. The model underestimates PM_{2.5} while overestimating O₃. The bias for NO₂, albeit small, fluctuates considerably among stations, and the same fluctuations are observed for SO₂. The particularly problematic simulations for SO₂ may be ascribed to the model configuration that the point source emissions of SO₂ (e.g., from power plants and industrial sites) are assigned directly to the immediate surface, leading a considerable overestimation of the surface ambient concentration.

		MB [$\mu\text{g}/\text{m}^3$]	NMB	RMSE [$\mu\text{g}/\text{m}^3$]	IOA
PM _{2.5}	day	-4.60 (2.97)	-0.39 (0.20)	6.45 (2.45)	0.57 (0.10)
	hour	-6.54 (3.20)	-0.50 (0.19)	9.21 (3.36)	0.50 (0.09)
O ₃	day	25.74 (19.18)	0.45 (0.39)	32.29 (12.99)	0.51 (0.16)
	hour	25.79 (19.21)	0.46 (0.39)	38.06 (11.74)	0.65 (0.13)
NO ₂	day	2.32 (7.41)	0.29 (0.61)	7.55 (5.12)	0.52 (0.15)
	hour	-0.48 (9.96)	0.13 (0.50)	13.93 (10.43)	0.47 (0.13)
SO ₂	day	8.70 (20.17)	0.88 (1.75)	13.40 (19.54)	0.40 (0.15)
	hour	5.49 (19.77)	0.38 (1.47)	15.26 (25.44)	0.36 (0.10)

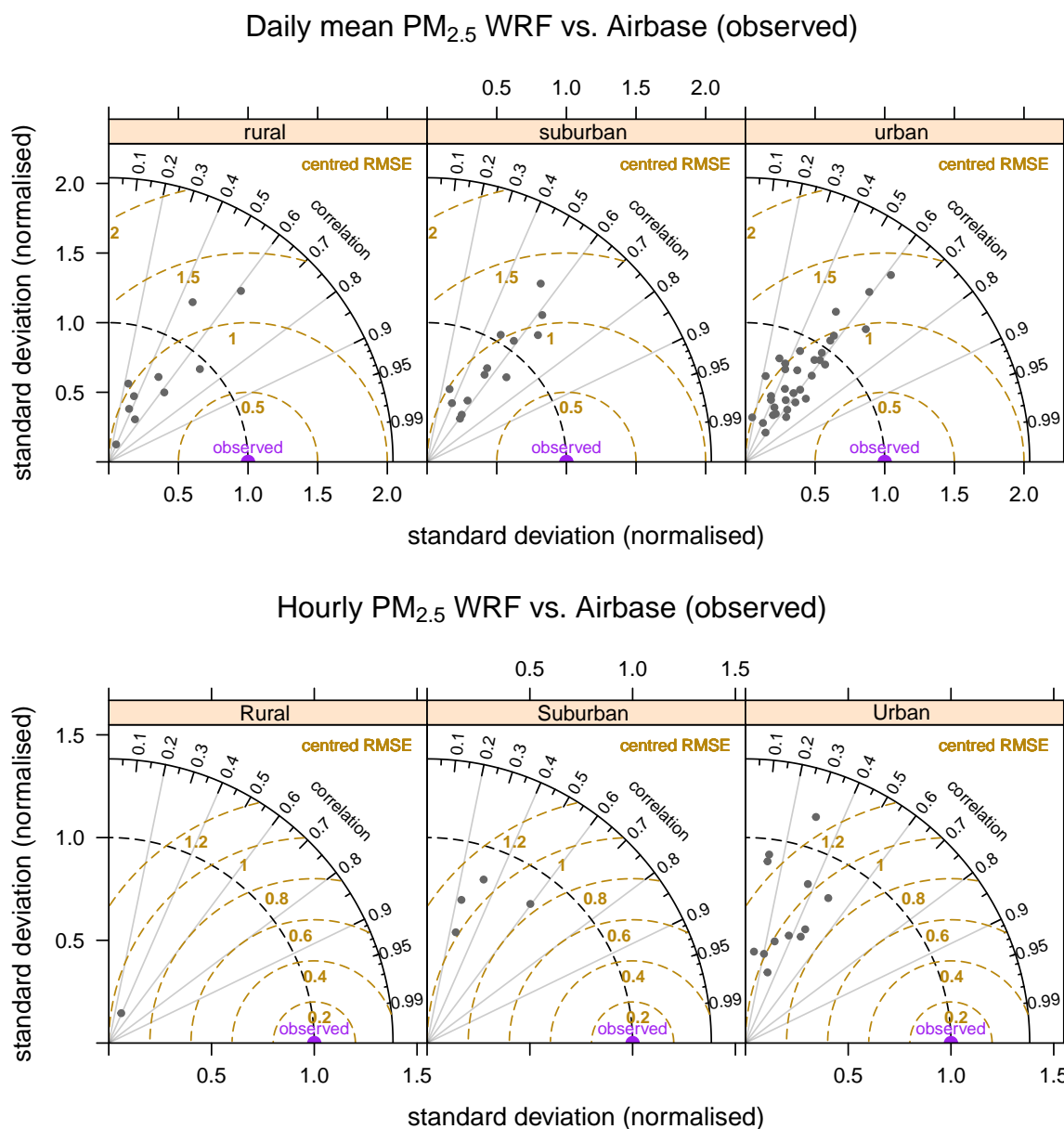


Figure B1. Evaluation of PM_{2.5} concentration simulated by WRF-Chem using AirBase E1a & E2a air quality data. We only used the verified AirBase data with "verification code" equal to 1 (verified) or 2 (preliminary verified) in the metadata, and data from stations where more than 75% of observations are present during the entire period. Each AirBase station is spatially matched to a WRF grid cell (20 km). The hourly WRF values are also averaged to daily means for comparison with AirBase data from stations where only daily means were recorded. The model achieved a correlation coefficient of larger than 0.5 and centred RMSE below 1.0 for most stations in simulating daily mean PM_{2.5}. It slightly outperformed the model for reproducing the hourly mean PM_{2.5}.

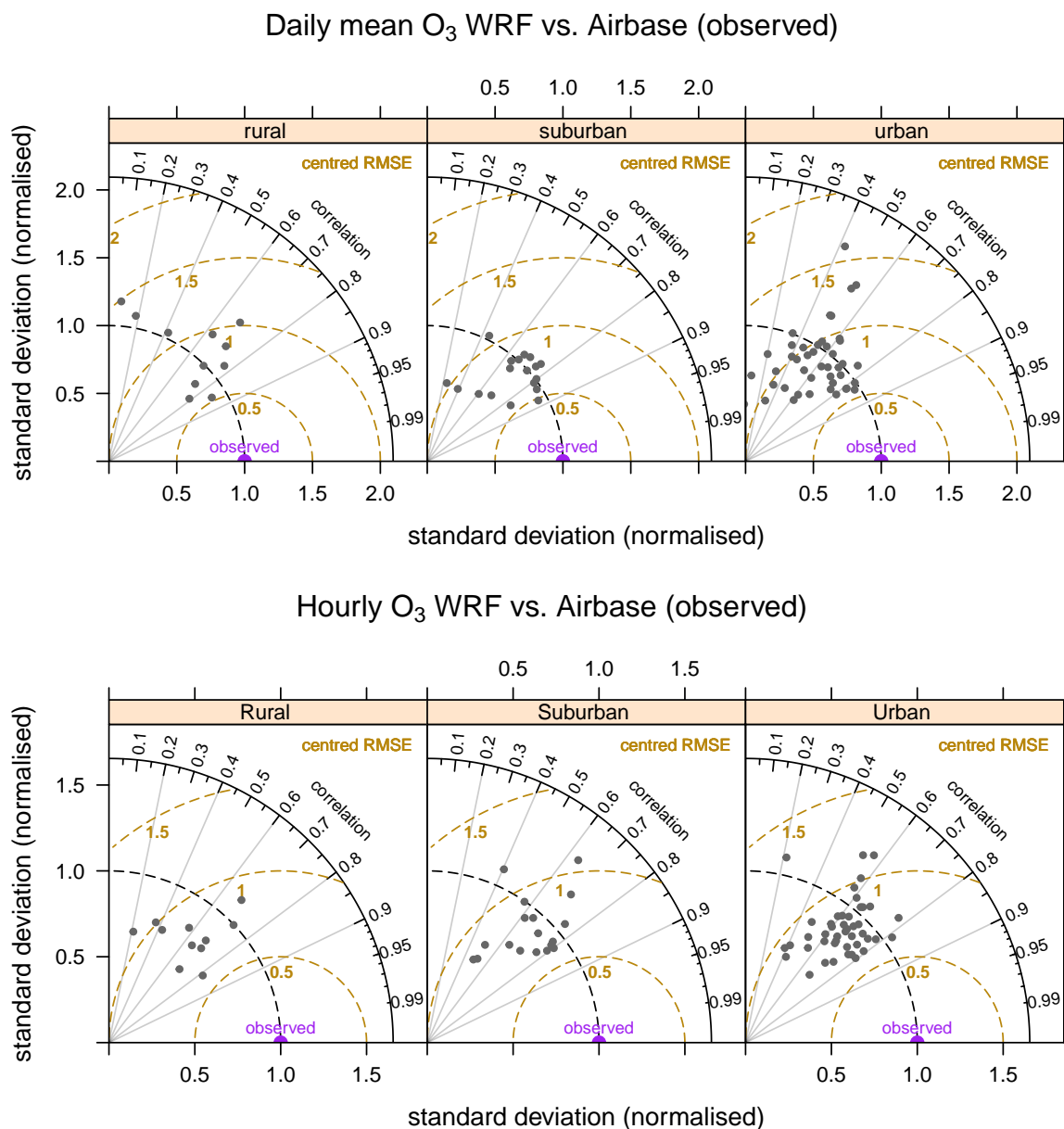


Figure B2. Evaluation of O₃ concentration simulated by WRF-Chem using AirBase data. The AirBase observations of O₃ are collected hourly. Aggregating hourly values to daily aggregates does not help increase the model performance. On the contrary, the model performance of reproducing the O₃ variation drops slightly with the mean IOA decreasing from 0.65 to 0.51.

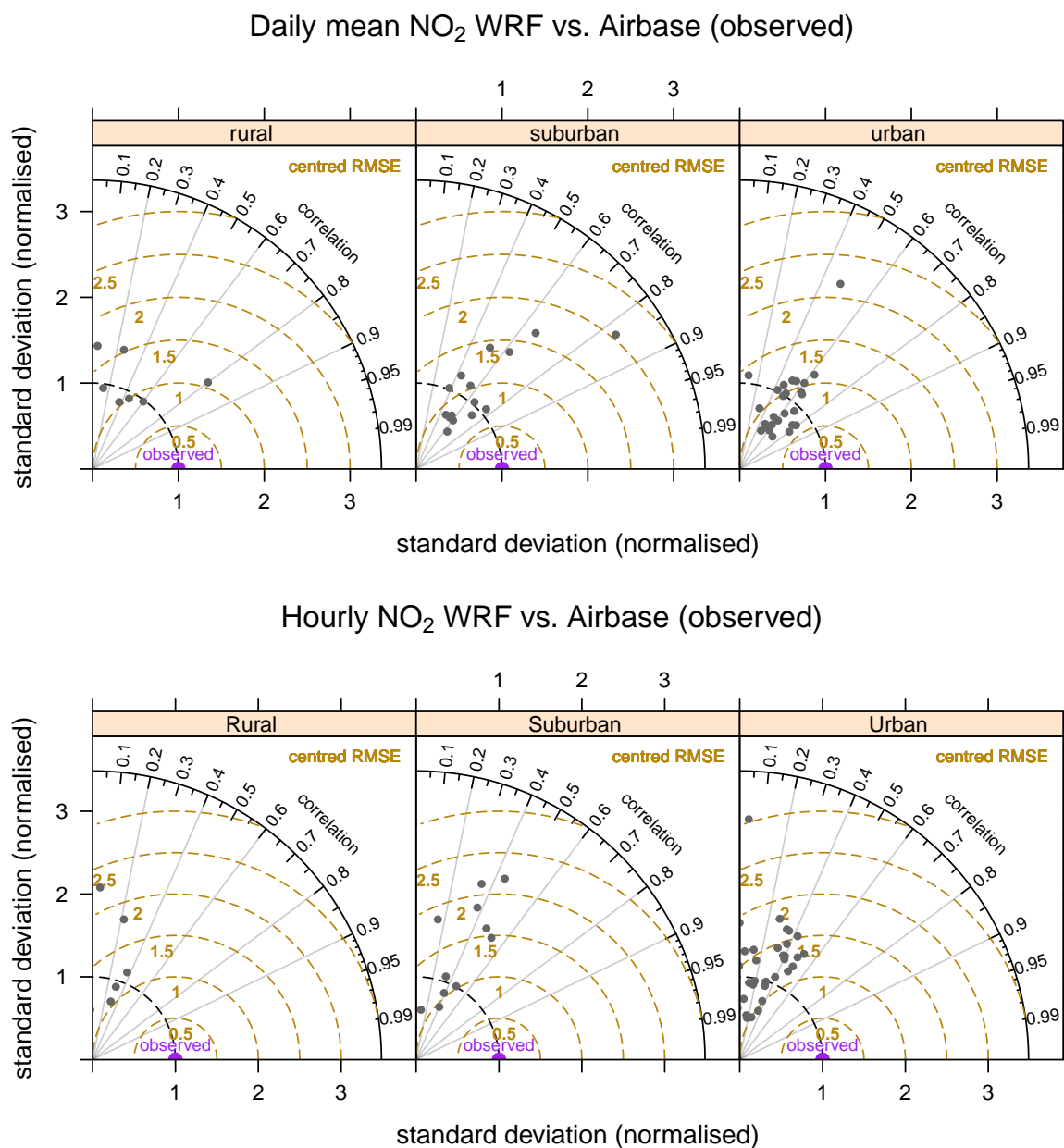


Figure B3. Evaluation of NO₂ concentration simulated by WRF-Chem using AirBase data. The model demonstrated a mediocre performance in simulating both daily and hourly mean of NO₂.

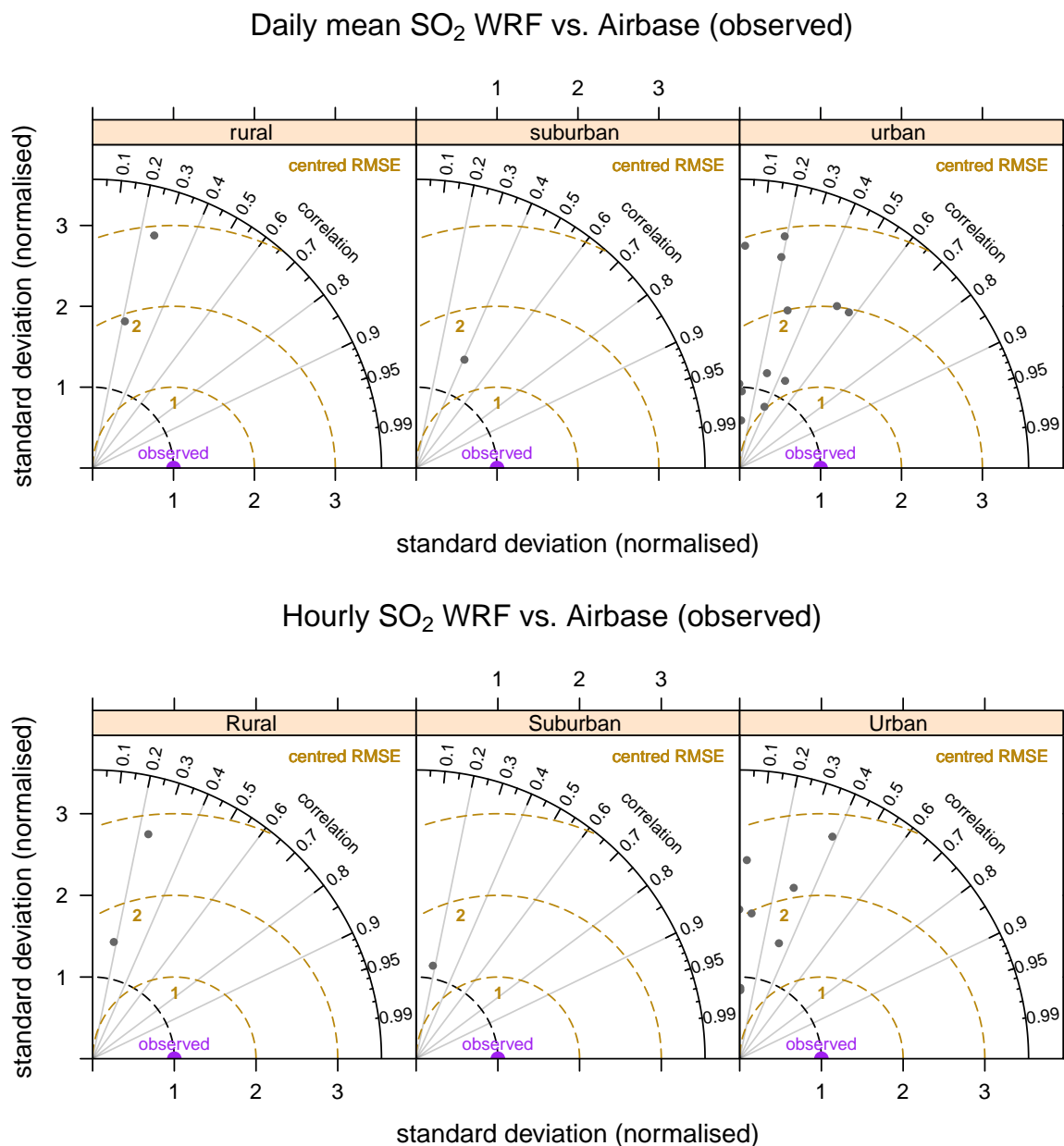


Figure B4. Evaluation of SO₂ concentration simulated by WRF-Chem using AirBase data. It remains a huge challenge to simulate the near-surface concentration SO₂. The model is prone to overestimate the SO₂ concentration, as the point source SO₂ emissions from inventories are assigned to the immediate surface. However, as SO₂ is not a major constituent in wildfire smokes, the resulting impacts on the conclusions of this study is considered minimal.



350 Appendix C: Diurnal pattern of multi-month hourly mean concentration of air pollutants

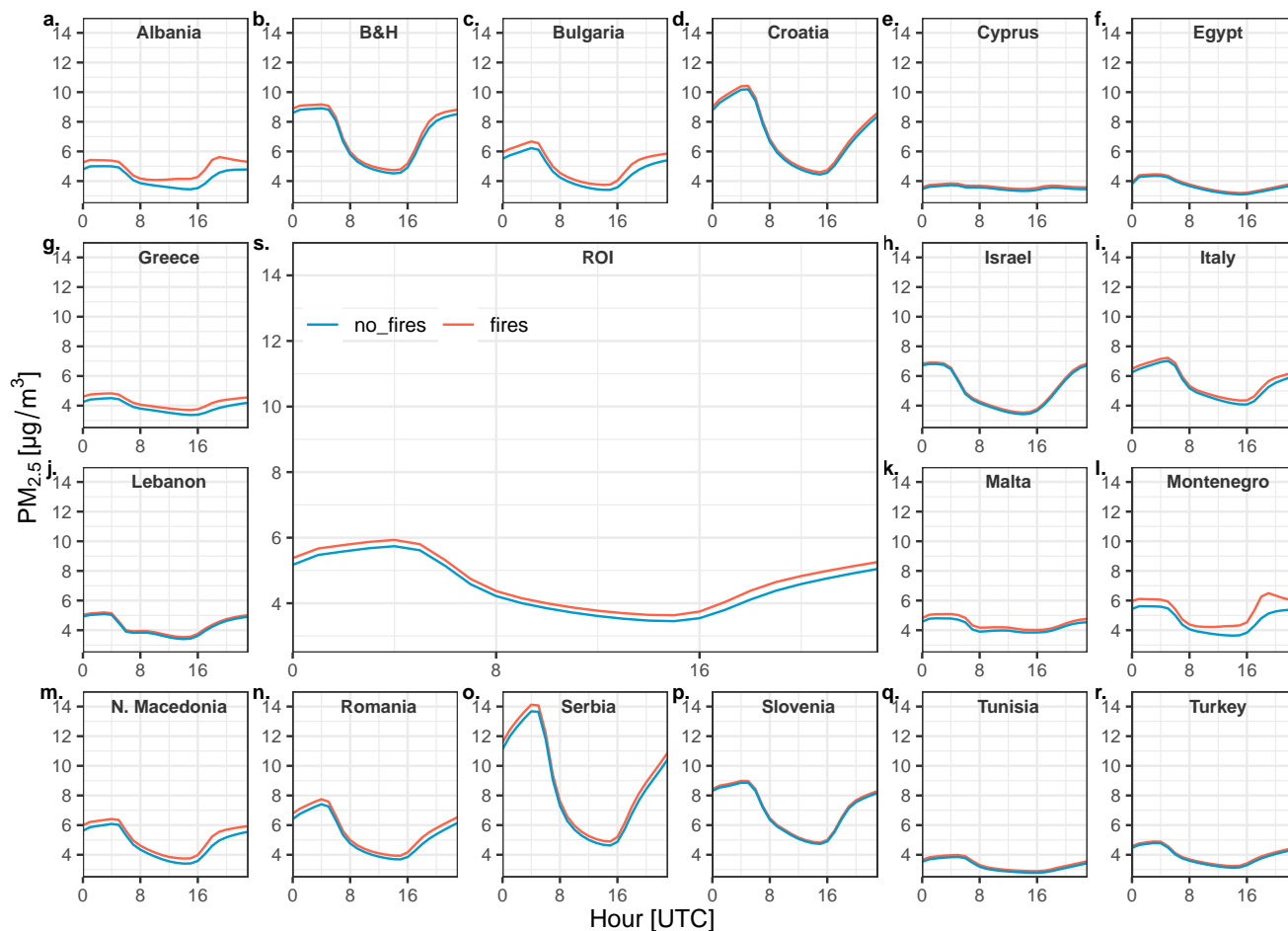


Figure C1. Diurnal pattern of multi-month hourly mean ambient $PM_{2.5}$ concentration for each country and the entire region of interest (ROI). In most countries, the $PM_{2.5}$ concentration follows a pattern subject to the mixing layer height. The daytime troughs reflect strong turbulent exchange and dilution within the mixed layer, whereas stable nighttime boundary layer enables $PM_{2.5}$ to accumulate Manning et al. (2018). The wildfire-caused elevation of $PM_{2.5}$ concentration in all countries can be ascribed to the emissions of gas-phase air pollutants such as NO_x , and VOCs from wildfires. These species are oxidized to form less volatile nitrates, and secondary organic aerosols (SOAs), respectively, and condense into the particle phase Kroll et al. (2020).

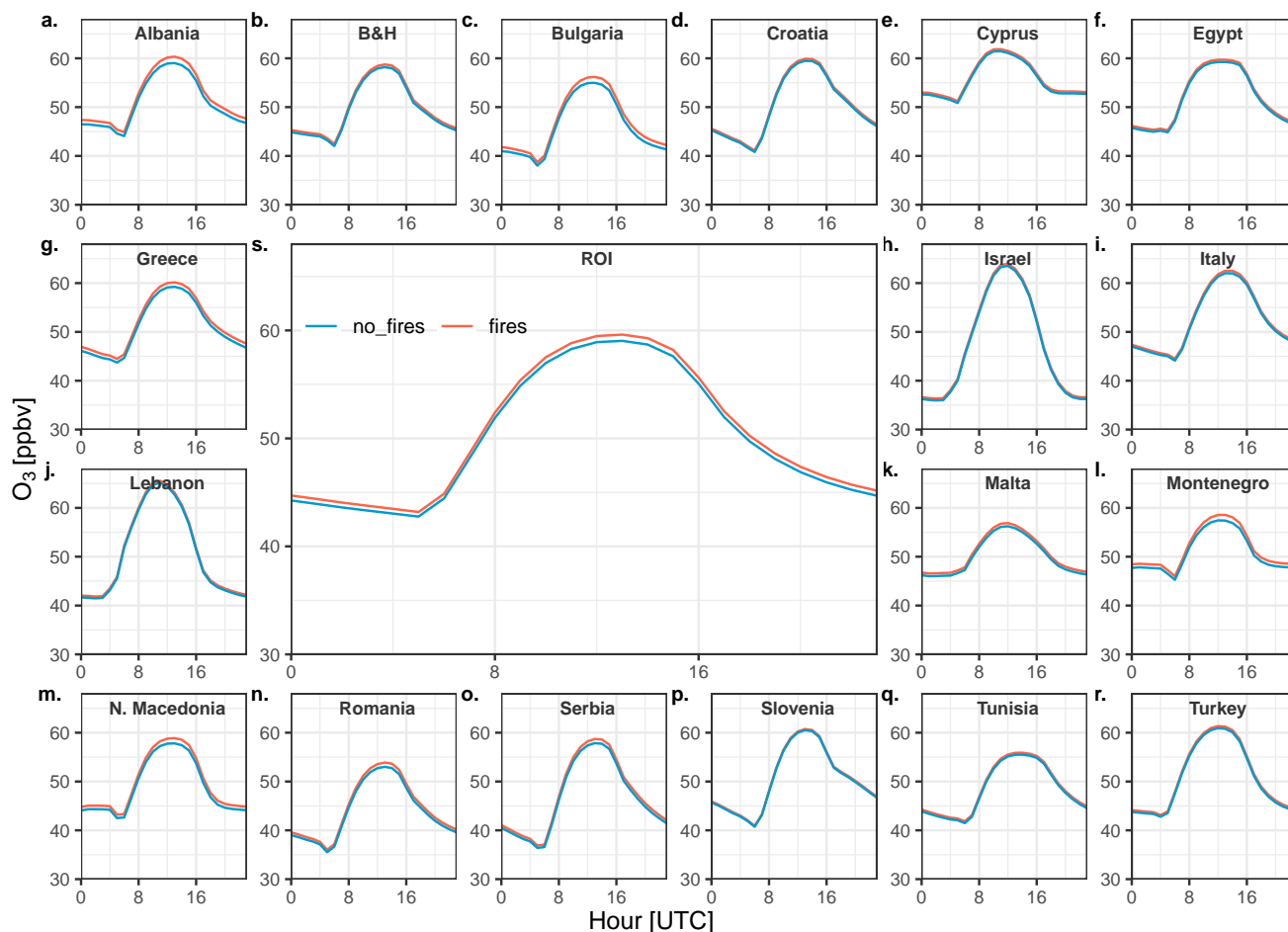


Figure C2. Diurnal pattern of multi-month hourly mean O_3 concentration for each country and the entire ROI. The biomass burning presents a significant source of O_3 precursors such as carbon monoxide (CO), volatile organic compounds (VOCs), and nitrogen oxide (NO_x), which form O_3 in the presence of solar radiation Holzinger et al. (1999). Therefore, the concentration of O_3 starts to increase after the sunrise and often reaches its maximum in the afternoon (between LT1200 and 1600). As the time of the day (horizontal axis) is in UTC, the local time (LT) of each country may be offset, subject to the time zone which the country is located in.

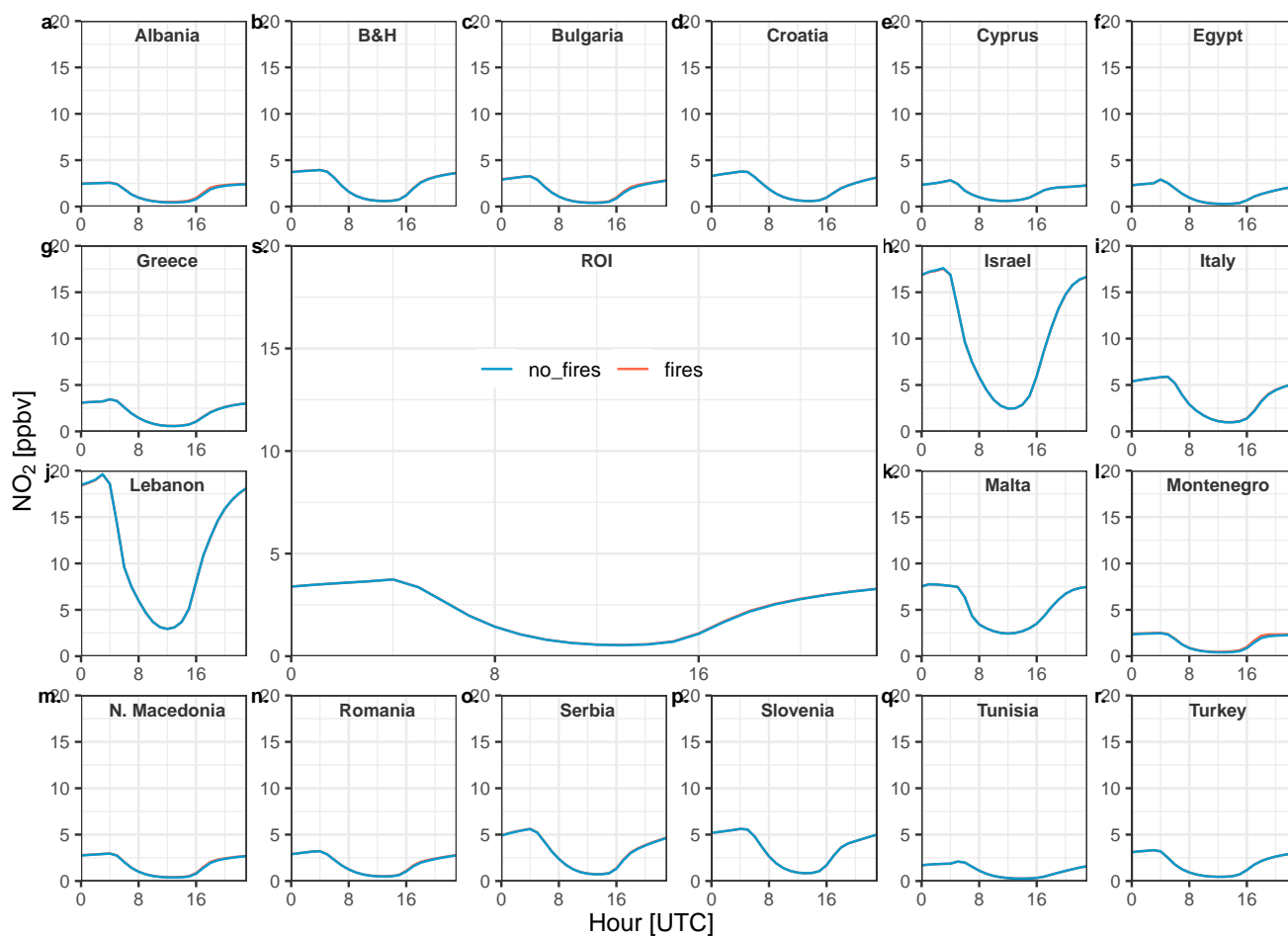


Figure C3. Diurnal pattern of multi-month hourly mean NO₂ concentration for each country and the entire region of interest.

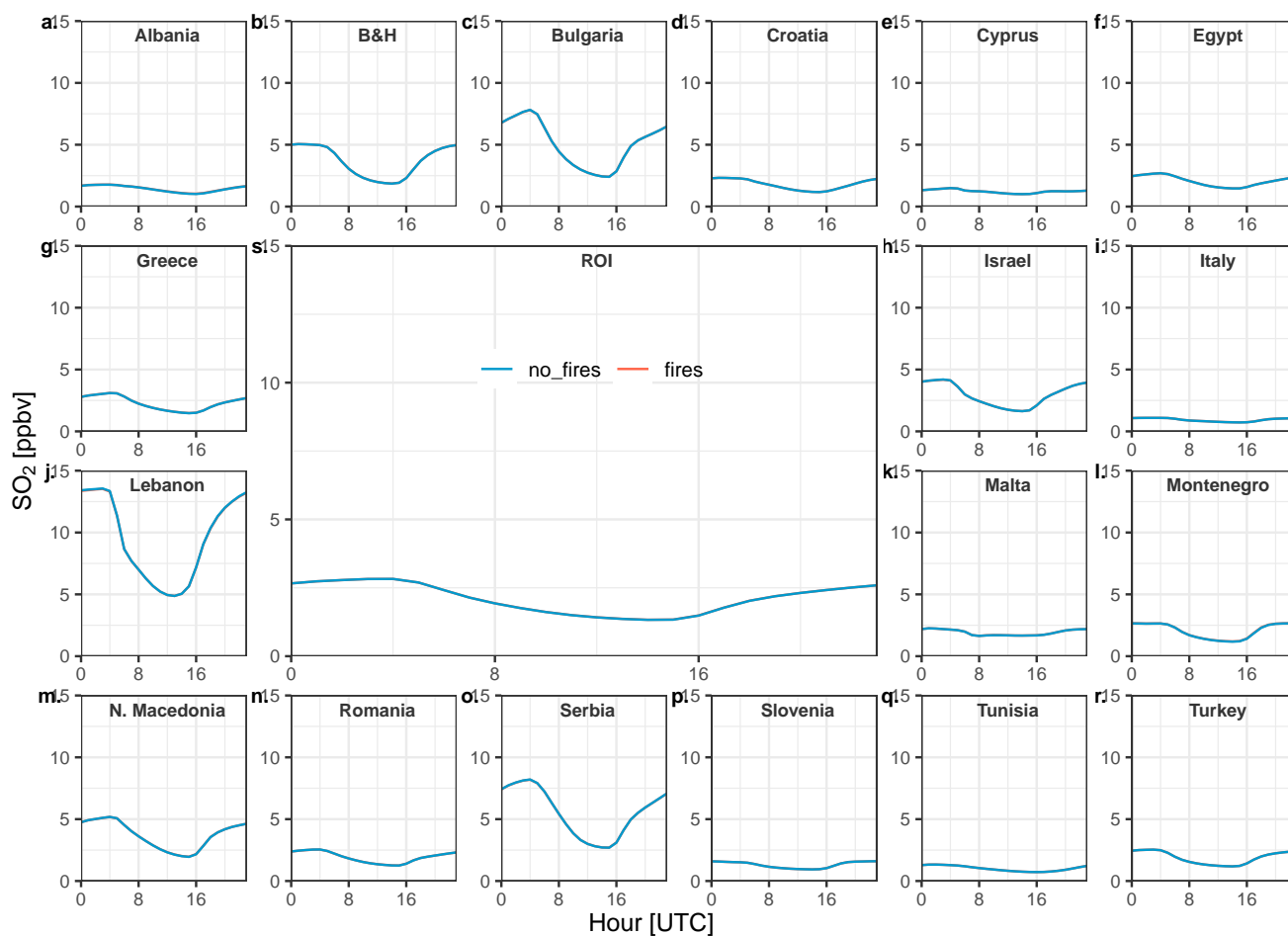


Figure C4. Diurnal pattern of multi-month hourly mean SO₂ concentration for each country and the entire region of interest.



Appendix D: Spatial pattern of WRF-simulated multi-month mean concentration of air pollutants

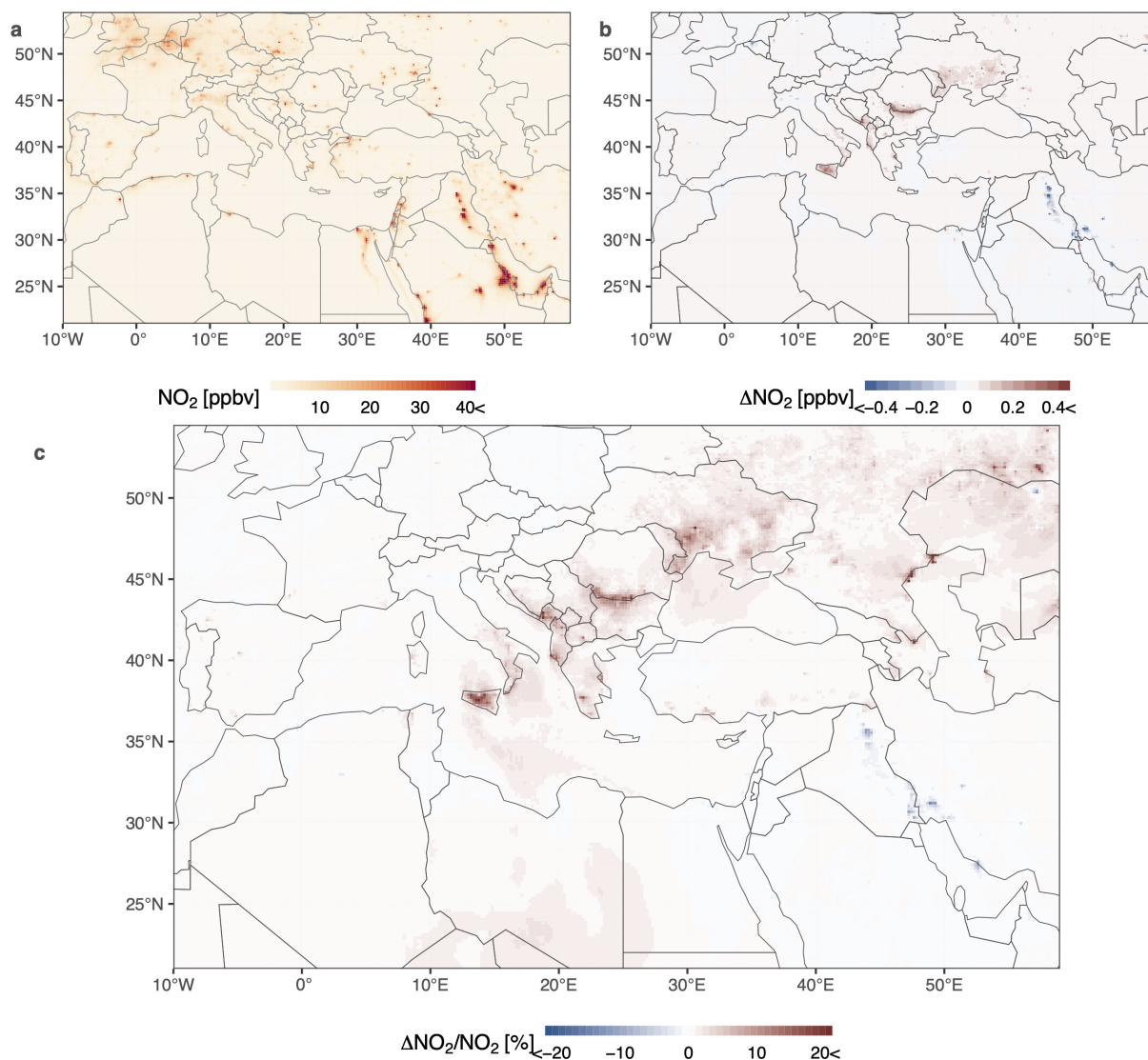


Figure D1. Spatial pattern of multi-month mean NO₂ concentration simulated by WRF-Chem under the fires scenario and the difference relative to the no-fires scenario. a) NO₂ concentration simulated with fires scenario, b) NO₂ concentration with fires scenario relative to no-fires ($\Delta\text{NO}_2 = \text{NO}_2^{\text{fires}} - \text{NO}_2^{\text{no-fires}}$), c) NO₂ concentration attributable to wildfires in percentages ($\Delta\text{NO}_2/\text{NO}_2^{\text{fires}}$). The elevation of wildfire-caused NO₂ is mainly observed in the Balkans, Greece, and South Italy.

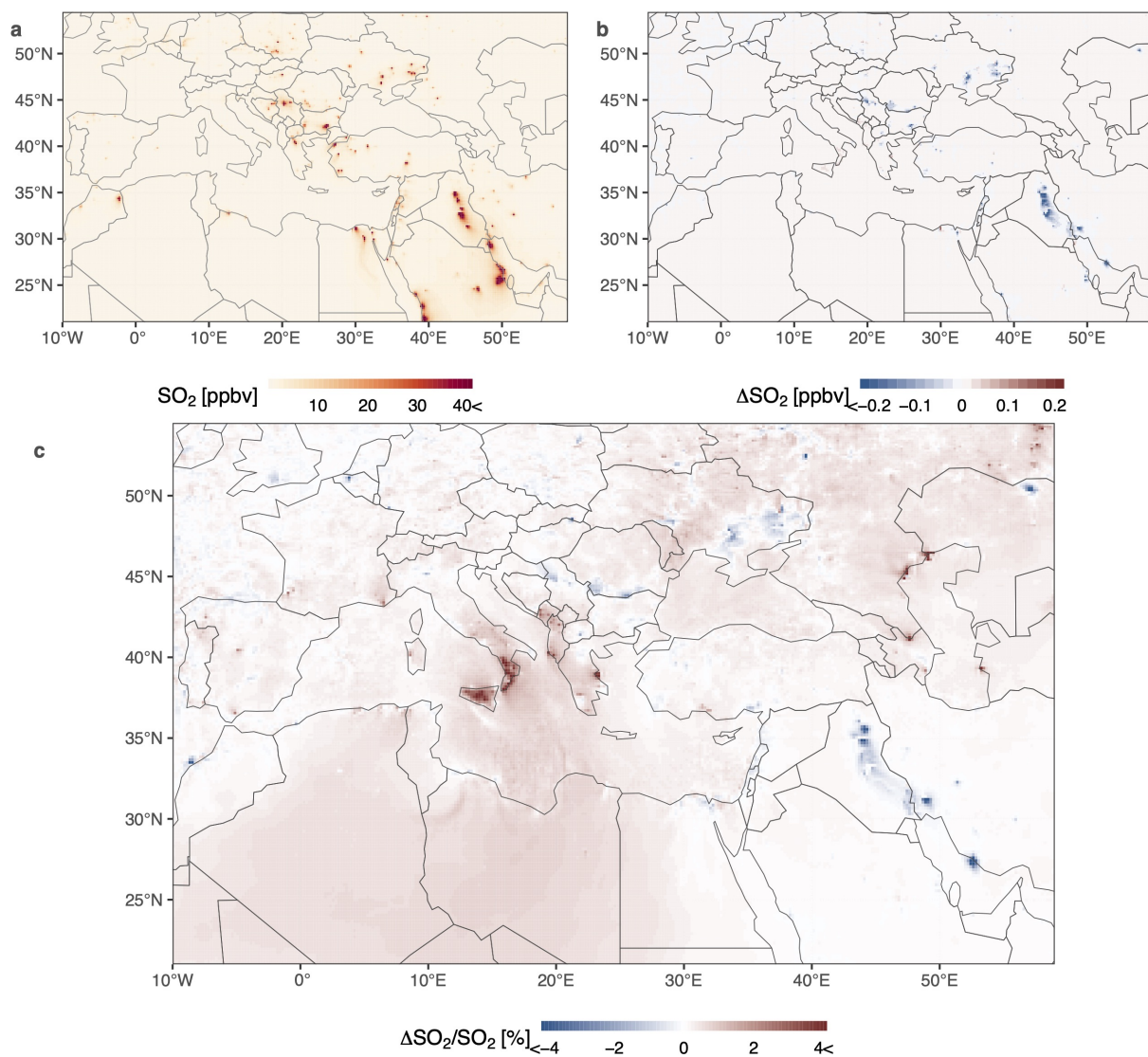


Figure D2. Spatial pattern of multi-month mean SO₂ concentration simulated by WRF-Chem under the fires scenario and the difference relative to the no-fires scenario. a) SO₂ concentration simulated with fires scenario, b) SO₂ concentration with fires scenario relative to no-fires ($\Delta\text{SO}_2 = \text{SO}_2^{\text{fires}} - \text{SO}_2^{\text{no-fires}}$), c) SO₂ concentration attributable to wildfires in percentages ($\Delta\text{SO}_2/\text{SO}_2^{\text{fires}}$). The elevation of wildfire-caused SO₂ is mainly observed in Albania, Greece and South Italy.

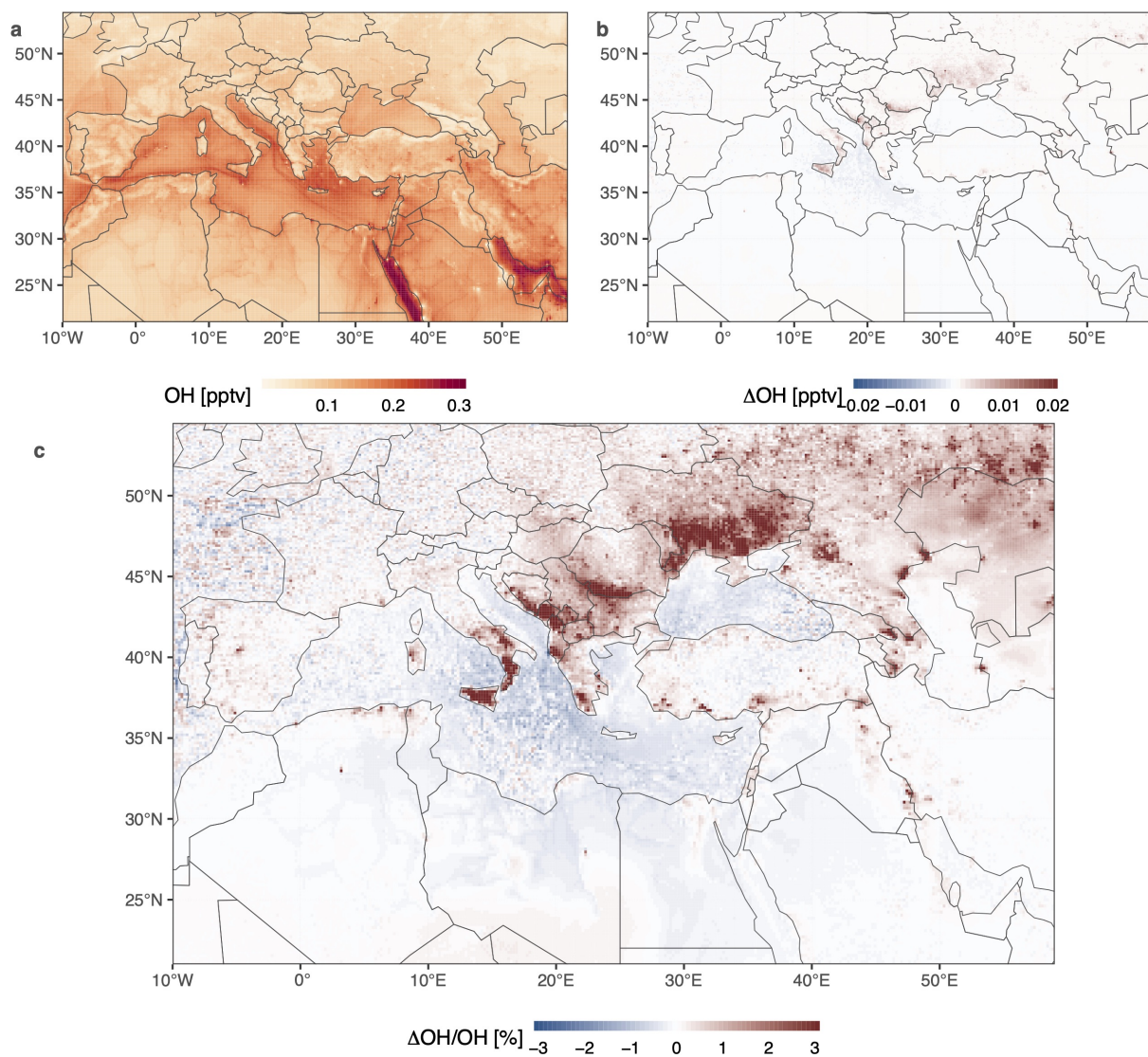


Figure D3. Spatial pattern of multi-month mean hydroxyl radical (OH) concentration simulated by WRF-Chem under the fires scenario and the difference relative to the no-fires scenario. a) OH concentration simulated with fires scenario, b) OH concentration with fires scenario relative to no-fires ($\Delta\text{OH} = \text{OH}^{\text{fires}} - \text{OH}^{\text{no-fires}}$), c) OH concentration attributable to wildfires in percentages ($\Delta\text{OH}/\text{OH}^{\text{fires}}$).



Appendix E: Time series of population weighted concentration difference

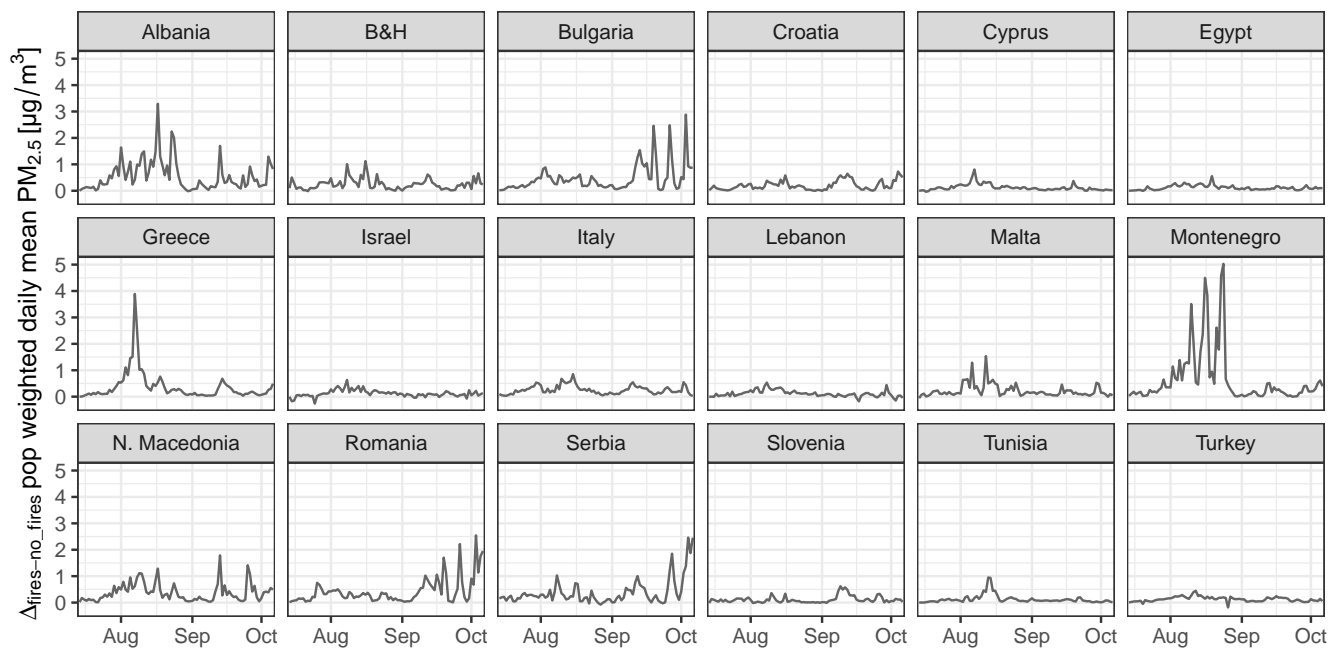


Figure E1. Time series of 24h-mean population weighted concentration difference of $PM_{2.5}$ (fires - no_fires) for each country.

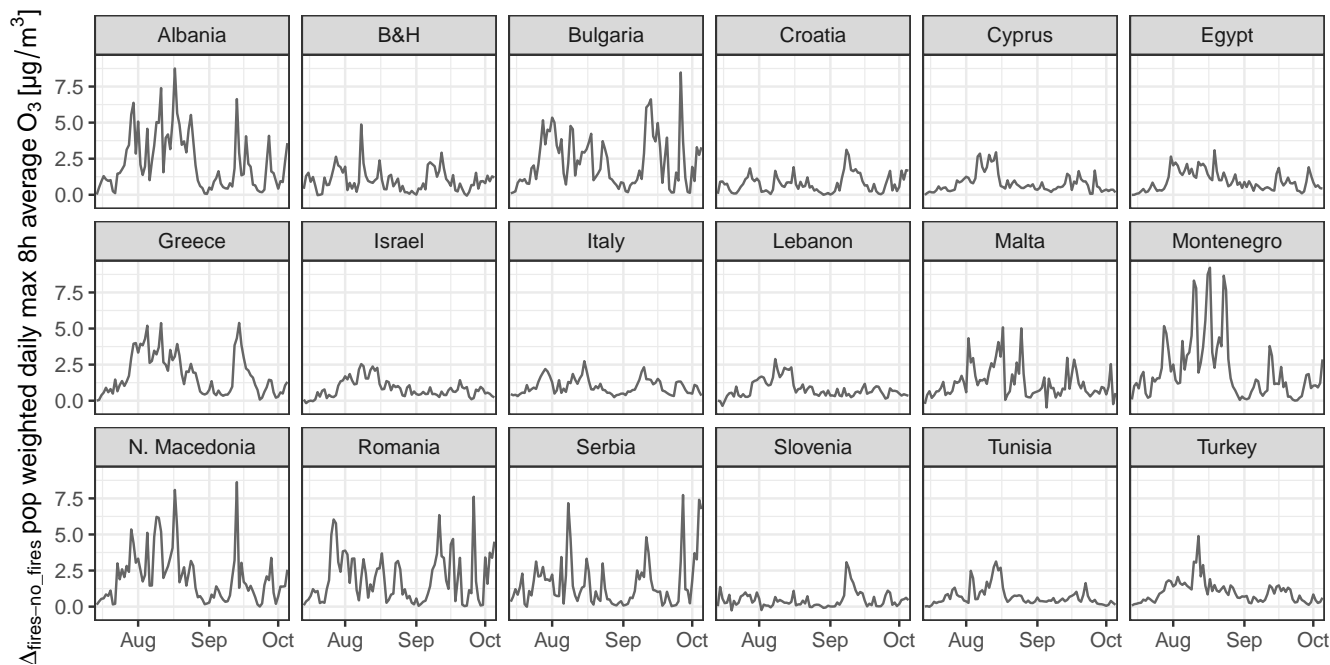


Figure E2. Time series of population weighted daily maximum 8-h average (DMA8) difference of O_3 (fires - no_fires) for each country.

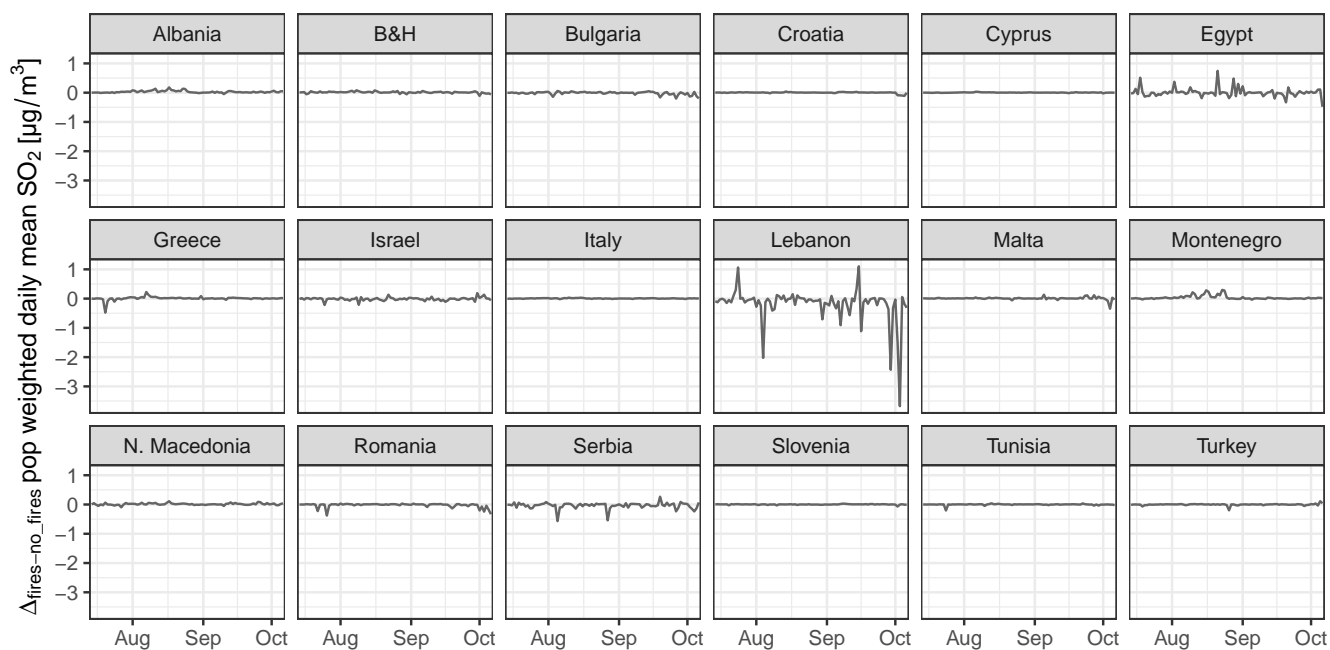


Figure E3. Time series of 24h-mean population weighted concentration difference of SO_2 (fires - no_fires) for each country.

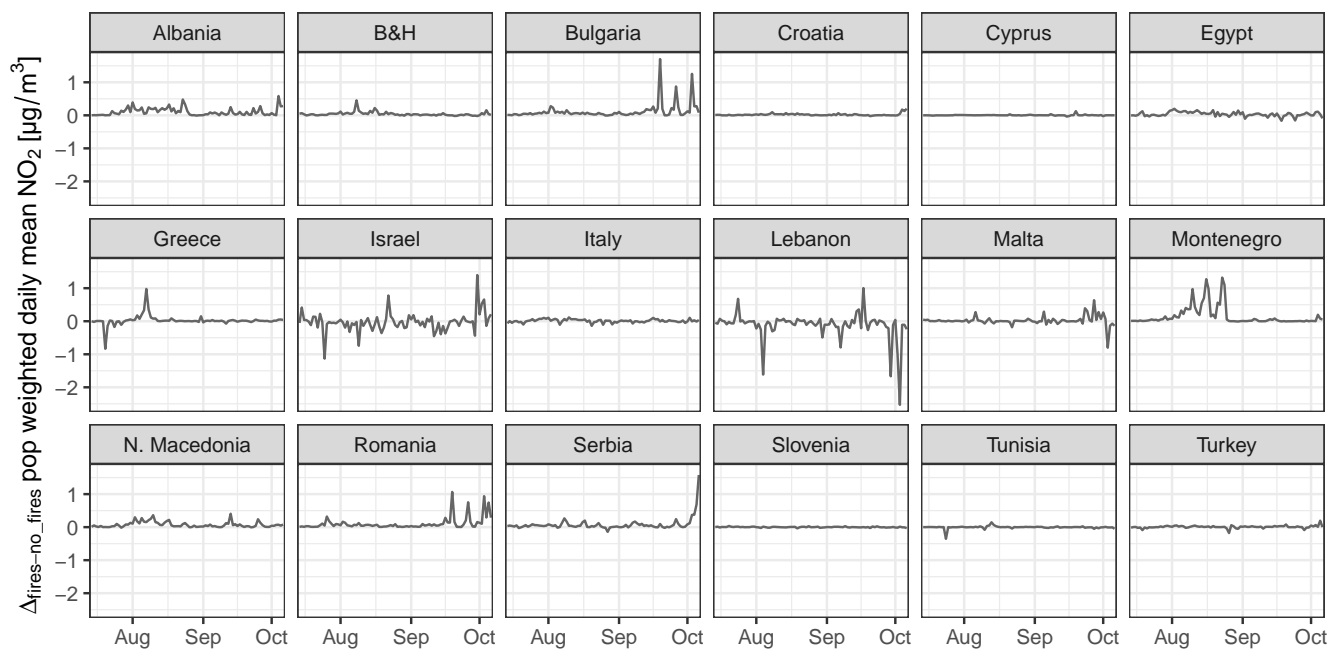


Figure E4. Time series of 24h-mean population weighted concentration difference of NO_2 (fires - no_fires) for each country.



Data availability. Original WRF-Chem simulation data are available upon request to the corresponding author.

Code and data availability. Data and code used for data analysis and visualization are available on Zenodo, <https://doi.org/10.5281/zenodo.8120613>

355 *Author contributions.* Both authors were involved in the study design and conceptualized the methodology. CK performed the WRF-Chem simulation. BZ curated, analysed and visualized the data. BZ wrote the original draft of the manuscript. CK reviewed and edited the manuscript. CK administrated the project and funding acquisition.

Competing interests. The authors declare no conflict of interest.

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References

- 365 Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D., and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, *Atmospheric Chemistry and Physics*, 11, 4039–4072, <https://doi.org/10.5194/acp-11-4039-2011>, 2011.
- Anderson, G. B., Krall, J. R., Peng, R. D., and Bell, M. L.: Is the relation between ozone and mortality confounded by chemical components of particulate matter? Analysis of 7 components in 57 US communities, *American Journal of Epidemiology*, 176, 726–732, <https://doi.org/10.1093/aje/kws188>, 2012.
- 370 Anenberg, S. C., Horowitz, L. W., Tong, D. Q., and West, J. J.: An estimate of the global burden of anthropogenic ozone and fine particulate matter on premature human mortality using atmospheric modeling, *Environmental Health Perspectives*, 118, 1189–1195, <https://doi.org/10.1289/ehp.0901220>, 2010.
- Archer-Nicholls, S., Lowe, D., Darbyshire, E., Morgan, W. T., Bela, M. M., Pereira, G., Trembath, J., Kaiser, J. W., Longo, K. M., Freitas, S. R., Coe, H., and McFiggans, G.: Characterising Brazilian biomass burning emissions using WRF-Chem with MOSAIC sectional aerosol, *Geoscientific Model Development*, 8, 549–577, <https://doi.org/10.5194/gmd-8-549-2015>, 2015.
- Bell, M. L., Kim, J. Y., and Dominici, F.: Potential confounding of particulate matter on the short-term association between ozone and mortality in multisite time-series studies, *Environmental Health Perspectives*, 115, 1591–1595, <https://doi.org/10.1289/ehp.10108>, 2007.
- Bencherif, H., Bègue, N., Pinheiro, D. K., du Preez, D. J., Cadet, J. M., Lopes, F. J. d. S., Shikwambana, L., Landulfo, E., Vescovini, T., Labuschagne, C., Silva, J. J., Anabor, V., Coheur, P. F., Mbatha, N., Hadji-Lazaro, J., Sivakumar, V., and Clerbaux, C.: Investigating the long-range transport of aerosol plumes following the amazon fires (August 2019): A multi-instrumental approach from ground-based and satellite observations, *Remote Sensing*, 12, 1–18, <https://doi.org/10.3390/rs12223846>, 2020.
- 380 Bowman, D. M. J. S., Kolden, C. A., Abatzoglou, J. T., Johnston, F. H., van der Werf, G. R., and Flannigan, M.: Vegetation fires in the Anthropocene, *Nature Reviews Earth & Environment*, 1, 500–515, <https://doi.org/10.1038/s43017-020-0085-3>, 2020.
- 385 Brunekreef, B. and Holgate, S. T.: Air pollution and health, *Lancet*, 360, 1233–1242, [https://doi.org/10.1016/S0140-6736\(02\)11274-8](https://doi.org/10.1016/S0140-6736(02)11274-8), 2002.
- Burke, M., Driscoll, A., Heft-Neal, S., Xue, J., Burney, J., and Wara, M.: The changing risk and burden of wildfire in the United States, *Proceedings of the National Academy of Sciences of the United States of America*, 118, 1–6, <https://doi.org/10.1073/PNAS.2011048118>, 2021.
- Burnett, R. T., Arden Pope, C., Ezzati, M., Olives, C., Lim, S. S., Mehta, S., Shin, H. H., Singh, G., Hubbell, B., Brauer, M., Ross Anderson, H., Smith, K. R., Balme, J. R., Bruce, N. G., Kan, H., Laden, F., Prüss-Ustün, A., Turner, M. C., Gapstur, S. M., Diver, W. R., and Cohen, A.: An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure, *Environmental Health Perspectives*, 122, 397–403, <https://doi.org/10.1289/ehp.1307049>, 2014.
- 390 Butt, E. W., Conibear, L., Knote, C., and Spracklen, D. V.: Large Air Quality and Public Health Impacts due to Amazonian Deforestation Fires in 2019, *GeoHealth*, 5, 1–16, <https://doi.org/10.1029/2021GH000429>, 2021.
- 395 CAMS: Wildfires wreaked havoc in 2021, CAMS tracked their impact, <https://atmosphere.copernicus.eu/wildfires-wreaked-havoc-2021-cams-tracked-their-impact>(Accessed:2022-02-01), 2021.
- Chen, G., Guo, Y., Yue, X., Tong, S., Gasparri, A., Bell, M. L., Armstrong, B., Schwartz, J., Jaakkola, J. J. K., Zanobetti, A., Lavigne, E., Nascimento Saldiva, P. H., Kan, H., Royé, D., Milojevic, A., Overcenco, A., Urban, A., Schneider, A., Entezari, A., Vicedo-Cabrera, A. M., Zeka, A., Tobias, A., Nunes, B., Alahmad, B., Forsberg, B., Pan, S.-C., Íñiguez, C., Ameling, C., De la Cruz Valencia, C., Åström, C., Houthuijs, D., Van Dung, D., Samoli, E., Mayvaneh, F., Sera, F., Carrasco-Escobar, G., Lei, Y., Orru, H., Kim, H., Holobaca, I.-H.,
- 400



- 405 Kyselý, J., Teixeira, J. P., Madureira, J., Katsouyanni, K., Hurtado-Díaz, M., Maasikmets, M., Ragettli, M. S., Hashizume, M., Stafoggia, M., Pascal, M., Scortichini, M., de Sousa Zanotti Stagliorio Coêlho, M., Valdés Ortega, N., Ryti, N. R. I., Scovronick, N., Matus, P., Goodman, P., Garland, R. M., Abrutzky, R., Garcia, S. O., Rao, S., Fratianni, S., Dang, T. N., Colistro, V., Huber, V., Lee, W., Seposo, X., Honda, Y., Guo, Y. L., Ye, T., Yu, W., Abramson, M. J., Samet, J. M., and Li, S.: Mortality risk attributable to wildfire-related PM_{2.5} pollution: a global time series study in 749 locations, *The Lancet Planetary Health*, 5, e579–e587, [https://doi.org/10.1016/S2542-5196\(21\)00200-X](https://doi.org/10.1016/S2542-5196(21)00200-X), 2021.
- CIESIN: Gridded Population of the World, Version 4 (GPWv4): Population Count Adjusted to Match 2015 Revision of UN WPP Country Totals, Revision 11, <https://doi.org/10.7927/H4PN93PB>, 2018.
- 410 Cohen, A. J., Brauer, M., Burnett, R., Anderson, H. R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R., Feigin, V., Freedman, G., Hubbell, B., Jobling, A., Kan, H., Knibbs, L., Liu, Y., Martin, R., Morawska, L., Pope, C. A., Shin, H., Straif, K., Shaddick, G., Thomas, M., van Dingenen, R., van Donkelaar, A., Vos, T., Murray, C. J., and Forouzanfar, M. H.: Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015, *The Lancet*, 389, 1907–1918, [https://doi.org/10.1016/S0140-6736\(17\)30505-6](https://doi.org/10.1016/S0140-6736(17)30505-6), 2017.
- 415 Conibear, L., Reddington, C. L., Silver, B. J., Chen, Y., Knote, C., Arnold, S. R., and Spracklen, D. V.: Statistical Emulation of Winter Ambient Fine Particulate Matter Concentrations From Emission Changes in China, *GeoHealth*, 5, 1–13, <https://doi.org/10.1029/2021GH000391>, 2021.
- Cos, J., Doblas-Reyes, F., Jury, M., Marcos, R., Bretonnière, P.-A., and Samsó, M.: The Mediterranean climate change hotspot in the CMIP5 and CMIP6 projections, *Earth System Dynamics Discussions*, pp. 1–26, <https://doi.org/10.5194/esd-2021-65>, 2021.
- 420 Crippa, M., Guizzardi, D., Muntean, M., and Schaaf, E.: EDGAR v5.0 Global Air Pollutant Emissions, <http://data.europa.eu/89h/377801af-b094-4943-8fdc-f79a7c0c2d19>, 2021.
- Di, Q., Amini, H., Shi, L., Kloog, I., Silvern, R., Kelly, J., Sabath, M. B., Choirat, C., Koutrakis, P., Lyapustin, A., Wang, Y., Mickley, L. J., and Schwartz, J.: An ensemble-based model of PM_{2.5} concentration across the contiguous United States with high spatiotemporal resolution, *Environment International*, 130, 104909, <https://doi.org/10.1016/j.envint.2019.104909>, 2019.
- 425 Dong, T. T., Hinwood, A. L., Callan, A. C., Zosky, G., and Stock, W. D.: In vitro assessment of the toxicity of bushfire emissions: A review, *Science of the Total Environment*, 603–604, 268–278, <https://doi.org/10.1016/j.scitotenv.2017.06.062>, 2017.
- EFFIS: Total burnt areas and total number of fires in 2021., <https://effis.jrc.ec.europa.eu/applications/data-and-services>(accessed: 2021-12-01), 2021.
- 430 Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geoscientific Model Development*, 3, 43–67, <https://doi.org/10.5194/gmd-3-43-2010>, 2010.
- European Environment Agency: AirBase air quality data (E1a & E2a), <https://discomap.eea.europa.eu/map/fme/AirQualityExport.htm>, 2021.
- Eurostat: Deaths (total) by month, 2010–2019, 2021.
- 435 Evangelopoulos, D., Katsouyanni, K., Keogh, R. H., Samoli, E., Schwartz, J., Barratt, B., Zhang, H., and Walton, H.: PM_{2.5} and NO₂ exposure errors using proxy measures, including derived personal exposure from outdoor sources: A systematic review and meta-analysis, *Environment International*, 137, 105500, <https://doi.org/10.1016/j.envint.2020.105500>, 2020.
- Fann, N., Alman, B., Broome, R. A., Morgan, G. G., Johnston, F. H., Pouliot, G., and Rappold, A. G.: The health impacts and economic value of wildland fire episodes in the US: 2008–2012, *Science of the total environment*, 610, 802–809, 2018.



- Freitas, S. R., Longo, K. M., Chatfield, R., Latham, D., Silva Dias, M. A. F., Andreae, M. O., Prins, E., Santos, J. C., Gielow, R., and
440 Carvalho Jr., J. A.: Including the sub-grid scale plume rise of vegetation fires in low resolution atmospheric transport models, *Atmospheric
Chemistry and Physics*, 7, 3385–3398, <https://doi.org/10.5194/acp-7-3385-2007>, 2007.
- Global Burden of Disease Collaborative Network: Global Burden of Disease Study 2019 (GBD 2019) Results., [http://ghdx.healthdata.org/
gbd-results-tool?params=gbd-api-2019-permalink/4e1860d6fad20ccf64c5e6f7e1f7a24b](http://ghdx.healthdata.org/gbd-results-tool?params=gbd-api-2019-permalink/4e1860d6fad20ccf64c5e6f7e1f7a24b), 2020.
- Gong, S.: A parameterization of sea-salt aerosol source function for sub-and super-micron particles, *Global biogeochemical cycles*, 17, 2003.
- 445 Graham, A. M., Pringle, K. J., Pope, R. J., Arnold, S. R., Conibear, L. A., Burns, H., Rigby, R., Borchers-Arriagada, N., Butt, E. W., Kiely,
L., Reddington, C., Spracklen, D. V., Woodhouse, M. T., Knote, C., and McQuaid, J. B.: Impact of the 2019/2020 Australian Megafires
on Air Quality and Health, *GeoHealth*, 5, 1–17, <https://doi.org/10.1029/2021GH000454>, 2021.
- Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled “online” chemistry
within the WRF model, *Atmospheric Environment*, 39, 6957–6975, 2005.
- 450 Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using
MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmospheric Chemistry and Physics*, 6, 3181–3210, 2006.
- Haikerwal, A., Akram, M., Monaco, A. D., Smith, K., Sim, M. R., Meyer, M., Tonkin, A. M., Abramson, M. J., and Dennekamp, M.:
Impact of fine particulate matter (PM_{2.5}) exposure during wildfires on cardiovascular health outcomes, *Journal of the American Heart
Association*, 4, 1–10, <https://doi.org/10.1161/JAHA.114.001653>, 2015.
- 455 Hobbs, P. V., Sinha, P., Yokelson, R. J., Christian, T. J., Blake, D. R., Gao, S., Kirchstetter, T. W., Novakov, T., and Pilewskie,
P.: Evolution of gases and particles from a savanna fire in South Africa, *Journal of Geophysical Research: Atmospheres*, 108,
<https://doi.org/10.1029/2002jd002352>, 2003.
- Hodzic, A. and Jimenez, J. L.: Modeling anthropogenically controlled secondary organic aerosols in a megacity: a simplified framework for
global and climate models, *Geoscientific Model Development*, 4, 901–917, <https://doi.org/10.5194/gmd-4-901-2011>, 2011.
- 460 Holzinger, R., Wameke, C., Hansel, A., Jordan, A., Lindinger, W., Scharffe, D. H., Schade, G., and Crutzen, P. J.: Biomass burning as a
source of formaldehyde, acetaldehyde, methanol, acetone, acetonitrile, and hydrogen cyanide, *Geophysical Research Letters*, 26, 1161–
1164, <https://doi.org/10.1029/1999GL900156>, 1999.
- Hough, I., Sarafian, R., Shtein, A., Zhou, B., Lepeule, J., and Kloog, I.: Gaussian Markov random fields improve ensemble predictions of
daily 1 km PM_{2.5} and PM₁₀ across France, *Atmospheric Environment*, 264, 118 693, <https://doi.org/10.1016/j.atmosenv.2021.118693>,
465 2021.
- Im, U., Bianconi, R., Solazzo, E., Kioutsioukis, I., Badia, A., Balzarini, A., Baró, R., Bellasio, R., Brunner, D., Chemel, C., Curci, G., Denier
van der Gon, H., Flemming, J., Forkel, R., Giordano, L., Jiménez-Guerrero, P., Hirtl, M., Hodzic, A., Honzak, L., Jorba, O., Knote, C.,
Makar, P. A., Manders-Groot, A., Neal, L., Pérez, J. L., Pirovano, G., Pouliot, G., San Jose, R., Savage, N., Schroder, W., Sokhi, R. S.,
Syrakov, D., Torian, A., Tuccella, P., Wang, K., Werhahn, J., Wolke, R., Zabkar, R., Zhang, Y., Zhang, J., Hogrefe, C., and Galmarini, S.:
470 Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2.
Part II: Particulate matter, *Atmospheric Environment*, 115, 421–441, <https://doi.org/10.1016/j.atmosenv.2014.08.072>, 2015a.
- Im, U., Bianconi, R., Solazzo, E., Kioutsioukis, I., Badia, A., Balzarini, A., Baró, R., Bellasio, R., Brunner, D., Chemel, C., Curci, G.,
Flemming, J., Forkel, R., Giordano, L., Jiménez-Guerrero, P., Hirtl, M., Hodzic, A., Honzak, L., Jorba, O., Knote, C., Kuenen, J. J.,
Makar, P. A., Manders-Groot, A., Neal, L., Pérez, J. L., Pirovano, G., Pouliot, G., San Jose, R., Savage, N., Schroder, W., Sokhi, R. S.,
475 Syrakov, D., Torian, A., Tuccella, P., Werhahn, J., Wolke, R., Yahya, K., Zabkar, R., Zhang, Y., Zhang, J., Hogrefe, C., and Galmarini, S.:



- Evaluation of operational on-line-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2. Part I: Ozone, *Atmospheric Environment*, 115, 404–420, <https://doi.org/10.1016/j.atmosenv.2014.09.042>, 2015b.
- Im, U., Brandt, J., Geels, C., Hansen, K. M., Christensen, J. H., Andersen, M. S., Solazzo, E., Kioutsioukis, I., Alyuz, U., Balzarini, A., Baro, R., Bellasio, R., Bianconi, R., Bieser, J., Colette, A., Curci, G., Farrow, A., Flemming, J., Fraser, A., Jimenez-Guerrero, P., Kitwiroon, N.,
480 Liang, C.-K., Nopmongcol, U., Pirovano, G., Pozzoli, L., Prank, M., Rose, R., Sokhi, R., Tuccella, P., Unal, A., Vivanco, M. G., West, J., Yarwood, G., Hogrefe, C., and Galmarini, S.: Assessment and economic valuation of air pollution impacts on human health over Europe and the United States as calculated by a multi-model ensemble in the framework of AQMEII3, *Atmospheric Chemistry and Physics*, 18, 5967–5989, <https://doi.org/10.5194/acp-18-5967-2018>, 2018.
- Jaffe, D. A. and Wigder, N. L.: Ozone production from wildfires: A critical review, *Atmospheric Environment*, 51, 1–10,
485 <https://doi.org/10.1016/j.atmosenv.2011.11.063>, 2012.
- Johnston, F. H., Henderson, S. B., Chen, Y., Randerson, J. T., Marlier, M., DeFries, R. S., Kinney, P., Bowman, D. M., and Brauer, M.: Estimated Global Mortality Attributable to Smoke from Landscape Fires, *Environmental Health Perspectives*, 120, 695–701, <https://doi.org/10.1289/ehp.1104422>, 2012.
- Jolly, W. M., Cochrane, M. A., Freeborn, P. H., Holden, Z. A., Brown, T. J., Williamson, G. J., and Bowman, D. M.: Climate-induced
490 variations in global wildfire danger from 1979 to 2013, *Nature Communications*, 6, 1–11, <https://doi.org/10.1038/ncomms8537>, 2015.
- Knote, C., Hodzic, A., Jimenez, J. L., Volkamer, R., Orlando, J. J., Baidar, S., Brioude, J., Fast, J., Gentner, D. R., Goldstein, A. H., Hayes, P. L., Knighton, W. B., Oetjen, H., Setyan, A., Stark, H., Thalman, R., Tyndall, G., Washenfelder, R., Waxman, E., and Zhang, Q.: Simulation of semi-explicit mechanisms of SOA formation from glyoxal in aerosol in a 3-D model, *Atmospheric Chemistry and Physics*, 14, 6213–6239, <https://doi.org/10.5194/acp-14-6213-2014>, 2014.
- 495 Knote, C., Hodzic, A., and Jimenez, J. L.: The effect of dry and wet deposition of condensable vapors on secondary organic aerosols concentrations over the continental US, *Atmospheric Chemistry and Physics*, 15, 1–18, <https://doi.org/10.5194/acp-15-1-2015>, 2015.
- Kramer, H. A., Mockrin, M. H., Alexandre, P. M., and Radeloff, V. C.: High wildfire damage in interface communities in California, *International Journal of Wildland Fire*, 28, 641–650, <https://doi.org/10.1071/WF18108>, 2019.
- Kroll, J. H., Heald, C. L., Cappa, C. D., Farmer, D. K., Fry, J. L., Murphy, J. G., and Steiner, A. L.: The complex chemical effects of
500 COVID-19 shutdowns on air quality, *Nature Chemistry*, 12, 777–779, <https://doi.org/10.1038/s41557-020-0535-z>, 2020.
- LeGrand, S. L., Polashenski, C., Letcher, T. W., Creighton, G. A., Peckham, S. E., and Cetola, J. D.: The AFWA dust emission scheme for the GOCART aerosol model in WRF-Chem v3. 8.1, *Geoscientific Model Development*, 12, 131–166, 2019.
- Lelieveld, J., Pozzer, A., Pöschl, U., Fnais, M., Haines, A., and Münzel, T.: Loss of life expectancy from air pollution compared to other risk factors: A worldwide perspective, *Cardiovascular Research*, 116, 1910–1917, <https://doi.org/10.1093/cvr/cvaa025>, 2020.
- 505 Liang, C. K., West, J. J., Silva, R. A., Bian, H., Chin, M., Davila, Y., Dentener, F. J., Emmons, L., Flemming, J., Folberth, G., Henze, D., Im, U., Jonson, J. E., Keating, T. J., Kucsera, T., Lenzen, A., Lin, M., Tronstad Lund, M., Pan, X., Park, R. J., Pierce, R. B., Sekiya, T., Sudo, K., and Takemura, T.: HTAP2 multi-model estimates of premature human mortality due to intercontinental transport of air pollution and emission sectors, *Atmospheric Chemistry and Physics*, 18, 10 497–10 520, <https://doi.org/10.5194/acp-18-10497-2018>, 2018.
- Linares, C., Díaz, J., Negev, M., Martínez, G. S., Debono, R., and Paz, S.: Impacts of climate change on the public health of the
510 Mediterranean Basin population - Current situation, projections, preparedness and adaptation, *Environmental Research*, 182, 109 107, <https://doi.org/10.1016/j.envres.2019.109107>, 2020.
- Liu, C., Chen, R., Sera, F., Vicedo-Cabrera, A. M., Guo, Y. Y.-L., Tong, S., Coelho, M. S., Saldiva, P. H., Lavigne, E., Matus, P., Valdes Ortega, N., Osorio Garcia, S., Pascal, M., Stafoggia, M., Scortichini, M., Hashizume, M., Honda, Y., Hurtado-Díaz, M., Cruz, J., Nunes,



- 515 B., Teixeira, J. P., Kim, H., Tobias, A., Íñiguez, C., Forsberg, B., Åström, C., Ragettli, M. S., Guo, Y. Y.-L., Chen, B.-Y., Bell, M. L., Wright, C. Y., Scovronick, N., Garland, R. M., Milojevic, A., Kyselý, J., Urban, A., Orru, H., Indermitte, E., Jaakkola, J. J., Rytí, N. R., Katsouyanni, K., Analitis, A., Zanobetti, A., Schwartz, J., Chen, J., Wu, T., Cohen, A., Gasparri, A., and Kan, H.: Ambient Particulate Air Pollution and Daily Mortality in 652 Cities, *New England Journal of Medicine*, 381, 705–715, <https://doi.org/10.1056/nejmoa1817364>, 2019.
- 520 Lu, F., Xu, D., Cheng, Y., Dong, S., Guo, C., Jiang, X., and Zheng, X.: Systematic review and meta-analysis of the adverse health effects of ambient PM_{2.5} and PM₁₀ pollution in the Chinese population, *Environmental Research*, 136, 196–204, <https://doi.org/10.1016/j.envres.2014.06.029>, 2015.
- Macintyre, H. L., Heaviside, C., Neal, L. S., Agnew, P., Thornes, J., and Vardoulakis, S.: Mortality and emergency hospitalizations associated with atmospheric particulate matter episodes across the UK in spring 2014, *Environment International*, 97, 108–116, <https://doi.org/10.1016/j.envint.2016.07.018>, 2016.
- 525 Manning, M. I., Martin, R. V., Hasenkopf, C., Flasher, J., and Li, C.: Diurnal Patterns in Global Fine Particulate Matter Concentration, *Environmental Science and Technology Letters*, 5, 687–691, <https://doi.org/10.1021/acs.estlett.8b00573>, 2018.
- Mansournia, M. A. and Altman, D. G.: Population attributable fraction, *BMJ (Online)*, 360, 2–3, <https://doi.org/10.1136/bmj.k757>, 2018.
- Naeher, L. P., Brauer, M., Lipsett, M., Zelikoff, J. T., Simpson, C. D., Koenig, J. Q., and Smith, K. R.: Woodsmoke health effects: A review, *Inhalation Toxicology*, 19, 67–106, <https://doi.org/10.1080/08958370600985875>, 2007.
- 530 Orellano, P., Reynoso, J., Quaranta, N., Bardach, A., and Ciapponi, A.: Short-term exposure to particulate matter (PM₁₀ and PM_{2.5}), nitrogen dioxide (NO₂), and ozone (O₃) and all-cause and cause-specific mortality: Systematic review and meta-analysis, *Environment International*, 142, 105 876, <https://doi.org/10.1016/j.envint.2020.105876>, 2020.
- Page, M. J., Sterne, J. A. C., Higgins, J. P. T., and Egger, M.: Investigating and dealing with publication bias and other reporting biases in meta-analyses of health research: A review, *Research Synthesis Methods*, 12, 248–259, <https://doi.org/10.1002/jrsm.1468>, 2021.
- 535 Pebesma, E.: Simple Features for R: Standardized Support for Spatial Vector Data, *The R Journal*, 10, 439–446, <https://doi.org/10.32614/RJ-2018-009>, 2018.
- R Core Team: R: A Language and Environment for Statistical Computing, R Foundation for Statistical Computing, Vienna, Austria, <https://www.R-project.org/>, 2021.
- Ruffault, J., Curt, T., Moron, V., Trigo, R. M., Mouillot, F., Koutsias, N., Pimont, F., Martin-StPaul, N., Barbero, R., Dupuy, J. L., Russo, A., and Belhadj-Khedher, C.: Increased likelihood of heat-induced large wildfires in the Mediterranean Basin, *Scientific Reports*, 10, 1–9, <https://doi.org/10.1038/s41598-020-70069-z>, 2020.
- 540 Rutledge, G. K., Alpert, J., and Ebisuzaki, W.: NOMADS: A climate and weather model archive at the National Oceanic and Atmospheric Administration, *Bulletin of the American Meteorological Society*, 87, 327–341, <https://doi.org/10.1175/BAMS-87-3-327>, 2006.
- San-Miguel-Ayanz, J., Durrant, T., Boca, R., Maianti, P., Liberta', P., Artes Vivancos, T., Jacome Felix Oom, D. P., Branco, A., De Rigo, D., Ferrari, D., Pfeiffer, H., Grecchi, R., Nuijten, D., Onida, M., and Loffler, P.: Advance Report on Forest Fires in Europe, Middle East and North Africa 2021, Tech. rep., <https://doi.org/10.2760/039729>, 2022.
- 545 Schneider, S. R. and Abbatt, J. P.: Wildfire atmospheric chemistry: climate and air quality impacts, *Trends in Chemistry*, 4, 255–257, <https://doi.org/https://doi.org/10.1016/j.trechm.2021.12.004>, special Issue: Third Anniversary, 2022.
- Schwarz, L., Dimitrova, A., Aguilera, R., Basu, R., Gershunov, A., and Benmarhnia, T.: Smoke and COVID-19 case fatality ratios during California wildfires, *Environmental Research Letters*, 17, 014 054, <https://doi.org/10.1088/1748-9326/ac4538>, 2022.
- 550



- Shtein, A., Kloog, I., Schwartz, J., Silibello, C., Michelozzi, P., Gariazzo, C., Viegi, G., Forastiere, F., Karnieli, A., Just, A. C., and Stafoggia, M.: Estimating daily PM_{2.5} and PM₁₀ over Italy using an ensemble model, *Environmental Science and Technology*, 54, acs.est.9b04279, <https://doi.org/10.1021/acs.est.9b04279>, 2019.
- Sillman, S.: The relation between ozone, NO_x and hydrocarbons in urban and polluted rural environments, *Atmospheric Environment*, 33, 1821–1845, [https://doi.org/10.1016/S1352-2310\(98\)00345-8](https://doi.org/10.1016/S1352-2310(98)00345-8), 1999.
- 555 Stafoggia, M., Breitner, S., Hampel, R., and Basagaña, X.: Statistical Approaches to Address Multi-Pollutant Mixtures and Multiple Exposures: the State of the Science, *Current environmental health reports*, 4, 481–490, <https://doi.org/10.1007/s40572-017-0162-z>, 2017.
- Task Force on Hemispheric Transport of Air Pollution (TF HTAP): Hemispheric Transport of Air Pollution 2010. Part A: Ozone and Particulate Matter. *Air Pollution Studies No. 17*, 17, 2010.
- 560 Taylor, K. E.: Summarizing multiple aspects of model performance in a single diagram, *Journal of Geophysical Research: Atmospheres*, 106, 7183–7192, <https://doi.org/10.1029/2000JD900719>, 2001.
- Ver Hoef, J. M.: Who invented the delta method?, *American Statistician*, 66, 124–127, <https://doi.org/10.1080/00031305.2012.687494>, 2012.
- Vicedo-Cabrera, A. M., Sera, F., Liu, C., Armstrong, B., Milojevic, A., Guo, Y. Y. L. L., Tong, S., Lavigne, E., Kyselý, J., Urban, A., Orru, H., Indermitte, E., Pascal, M., Huber, V., Schneider, A., Katsouyanni, K., Samoli, E., Stafoggia, M., Scortichini, M., Hashizume, M., Honda, Y., Ng, C. F. S., Hurtado-Diaz, M., Cruz, J., Silva, S., Madureira, J., Scovronick, N., Garland, R. M., Kim, H., Tobias, A., Íñiguez, C., Forsberg, B., Åström, C., Ragettli, M. S., Röösli, M., Guo, Y. Y. L. L., Chen, B. Y., Zanobetti, A., Schwartz, J., Bell, M. L., Kan, H., and Gasparrini, A.: Short term association between ozone and mortality: global two stage time series study in 406 locations in 20 countries, *The BMJ*, 368, 1–10, <https://doi.org/10.1136/bmj.m108>, 2020.
- 565 Wang, Y., Puthussery, J. V., Yu, H., Liu, Y., Salana, S., and Verma, V.: Sources of cellular oxidative potential of water-soluble fine ambient particulate matter in the Midwestern United States, *Journal of Hazardous Materials*, 425, 127777, <https://doi.org/10.1016/j.jhazmat.2021.127777>, 2022.
- Wegesser, T. C., Pinkerton, K. E., and Last, J. A.: California wildfires of 2008: Coarse and fine particulate matter toxicity, *Environmental Health Perspectives*, 117, 893–897, <https://doi.org/10.1289/ehp.0800166>, 2009.
- Wei, Y., Wang, Y., Wu, X., Di, Q., Shi, L., Koutrakis, P., Zanobetti, A., Dominici, F., and Schwartz, J. D.: Causal effects of air pollution on mortality rate in Massachusetts, *American Journal of Epidemiology*, 189, 1316–1323, <https://doi.org/10.1093/aje/kwaa098>, 2020.
- 575 WHO: Health Risk Assessment of air pollution, World Health Organization, pp. 1–40, 2016.
- WHO: WHO global air quality guidelines. Particulate matter (PM_{2.5} and PM₁₀), ozone, nitrogen dioxide, sulfur dioxide and carbon monoxide, 2021.
- Wiedinmyer, C., Akagi, S., Yokelson, R. J., Emmons, L., Al-Saadi, J., Orlando, J., and Soja, A.: The Fire INventory from NCAR (FINN): A high resolution global model to estimate the emissions from open burning, *Geoscientific Model Development*, 4, 625–641, 2011.
- 580 Willmott, C. J.: On the validation of models, *Physical Geography*, 2, 184–194, <https://doi.org/10.1080/02723646.1981.10642213>, 1981.
- World Bank: Total population, <https://data.worldbank.org/indicator/SP.POP.TOTL> (Accessed 11 Dec. 2021), 2020.
- World Health Organization: Health risks of air pollution in Europe – HRAPIE project, World Health Organization (WHO), p. 60, 2013.
- Xie, Y., Lin, M., and Horowitz, L. W.: Summer PM_{2.5} Pollution Extremes Caused by Wildfires Over the Western United States During 2017–2018, *Geophysical Research Letters*, 47, 1–11, <https://doi.org/10.1029/2020GL089429>, 2020.
- 585 Xu, L., Crounse, J. D., Vasquez, K. T., Allen, H., Wennberg, P. O., Bourgeois, I., Brown, S. S., Campuzano-Jost, P., Coggon, M. M., Crawford, J. H., DiGangi, J. P., Diskin, G. S., Fried, A., Gargulinski, E. M., Gilman, J. B., Gkatzelis, G. I., Guo, H., Hair, J. W., Hall, S. R., Halliday, H. A., Hanisco, T. F., Hannun, R. A., Holmes, C. D., Huey, L. G., Jimenez, J. L., Lamplugh, A., Lee, Y. R., Liao, J., Lindaas, J., Neuman,



- 590 J. A., Nowak, J. B., Peischl, J., Peterson, D. A., Piel, F., Richter, D., Rickly, P. S., Robinson, M. A., Rollins, A. W., Ryerson, T. B.,
Sekimoto, K., Selimovic, V., Shingler, T., Soja, A. J., St. Clair, J. M., Tanner, D. J., Ullmann, K., Veres, P. R., Walega, J., Warneke, C.,
Washenfelder, R. A., Weibring, P., Wisthaler, A., Wolfe, G. M., Womack, C. C., and Yokelson, R. J.: Ozone chemistry in western U.S.
wildfire plumes, *Science Advances*, 7, <https://doi.org/10.1126/sciadv.abl3648>, 2021.
- Xu, R., Yu, P., Abramson, M. J., Johnston, F. H., Samet, J. M., Bell, M. L., Haines, A., Ebi, K. L., Li, S., and Guo, Y.: Wildfires, Global
Climate Change, and Human Health, *New England Journal of Medicine*, 383, 2173–2181, <https://doi.org/10.1056/NEJMs2028985>, 2020.
- 595 Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for simulating aerosol interactions and chemistry (MOSAIC), *Journal of
Geophysical Research: Atmospheres*, 113, 2008.
- Zhou, X., Josey, K., Kamareddine, L., Caine, M. C., Liu, T., Mickley, L. J., Cooper, M., and Dominici, F.: Excess of COVID-19
cases and deaths due to fine particulate matter exposure during the 2020 wildfires in the United States, *Science Advances*, 7, 1–12,
<https://doi.org/10.1126/sciadv.abi8789>, 2021.