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

Bin Zhou¹,² and Christoph Knote¹

¹Model-based Environmental Exposure Science, Faculty of Medicine, University of Augsburg, Universitätsstraße 2, 86159, Augsburg, Bavaria, Germany
²Potsdam Institute for Climate Impact Research (PIK), Member of the Leibniz Association, Telegrafenberg A31, 14473, Potsdam, Brandenburg, Germany

Correspondence: Bin Zhou (bin.zhou@med.uni-augsburg.de)

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 µm (PM².⁵), SO₂, NO₂, and O₃ - 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 O₃ 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 PM².⁵ exposure, rendering the health effect of increased O₃ from wildfires larger than the effect of increased PM².⁵. We attributed this largely to the spatially more widespread impact of wildfires on O₃. 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 (Naheher et al., 2007; Wegesser et al., 2009).
Wildfires emit large amounts of particulate matter (PM), nitrogen oxides (NO\textsubscript{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 (NH\textsubscript{3}), SO\textsubscript{2}, NO\textsubscript{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\textsubscript{x} and VOCs from wildfires lead to additional ozone (O\textsubscript{3}) 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\textsubscript{2.5}), SO\textsubscript{2}, NO\textsubscript{2}, and O\textsubscript{3} – 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.

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.

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 km$^2$ to be consistent with the configuration of the numerical model employed in this study (Fig. 1a).

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$_{2.5}$, O$_3$, NO$_2$, and SO$_2$ into the atmosphere (CAMS, 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. 1b-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$_{2.5}$, O$_3$, NO$_2$, and SO$_2$ 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., 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.

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 inlet 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. 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).
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 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 major air pollutants.

### 2.3 Model evaluation

We used the European Environmental Agency (EEA)'s AirBase air quality (including PM$_{2.5}$, O$_3$, NO$_2$, and SO$_2$) 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", "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 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 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

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:

$$C_{p_i,J,d} = \frac{\sum_{j} \left( \text{pop}_j \times c_{i,j,d} \right)}{\sum_{j} \text{pop}_j}$$

(1)

where $\text{pop}_j$ 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

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 (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 $BM_d$.

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 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.
Given a linear relationship and an increase of population exposure ($\Delta C$) of 10 $\mu g/m^3$, the RR is defined as

$$RR = e^{\beta \Delta C}.$$  \hspace{1cm} (2)

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

$$RR(\Delta X) = e^{\beta(\Delta X - X_0)}.$$  \hspace{1cm} (3)

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 $J$:

$$\Delta C_{P_{i,J}} = C_{P_{i,J,fire}} - C_{P_{i,J,no-fire}}.$$  \hspace{1cm} (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):

$$PAF = 1 - \frac{1}{RR}.$$  \hspace{1cm} (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:

$$E_{i,J} = \sum_{d=1}^{N} (BM_{J,d} \times PAF_{i,J}),$$  \hspace{1cm} (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.

### Table 1

<table>
<thead>
<tr>
<th></th>
<th>PM$_{2.5}$</th>
<th>O$_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Orellano et al. (2020)</td>
<td>1.0065 (1.0044–1.0086)</td>
<td>1.0043 (1.0034–1.0052)</td>
</tr>
<tr>
<td>WHO (2013)</td>
<td>1.0123 (1.0045–1.0201)</td>
<td>1.0029 (1.0014–1.0043)</td>
</tr>
<tr>
<td>Liu et al. (2019)*</td>
<td>1.0068 (1.0059–1.0077)</td>
<td>–</td>
</tr>
<tr>
<td>Vicedo-Cabrera et al. (2020)</td>
<td>–</td>
<td>1.0018 (1.0012–1.0024)</td>
</tr>
</tbody>
</table>

* 2-day moving average is used as the exposure metric instead of the daily mean used elsewhere.
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 (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 ($x \sim N(\mu, \sigma^2)$), the standard error (SE) of each data is derived as:

$$SE(x) = \frac{(CI_{\text{upper}} - CI_{\text{lower}})}{2 \cdot z_{1-0.05/2}},$$

(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 $\text{Var}(x) = SE(x)^2$.

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:

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

(8)

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:

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

(9)

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

The second approach is based on the Monte Carlo method. We respectively generated the normally distributed random 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 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$_3$ 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 ($\Delta$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 \( \text{O}_3 \), which is determined by the presence of sunlight, as \( \text{O}_3 \) is formed primarily by photo-chemical reactions.

Figures C3 and C4 show the temporal pattern of \( \text{NO}_2 \) and \( \text{SO}_2 \) concentrations respectively, while their spatial patterns are shown in Fig. D1 and D2. The hot spots of \( \text{NO}_2 \) and \( \text{SO}_2 \) 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 \( \text{O}_3 \), increases of \( \text{NO}_x \) and \( \text{SO}_2 \) 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 \( \text{PM}_{2.5} \), the diurnal pattern of \( \text{NO}_2 \) and \( \text{SO}_2 \) concentration is determined by the mixing layer height. Meanwhile, we found the expected increases in the concentration of \( \text{OH} \) 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 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 \( \text{PM}_{2.5} \) and \( \text{O}_3 \) 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 \( \text{NO}_2 \) and \( \text{SO}_2 \) within the simulation period and region. Therefore, we estimate the wildfire-associated health impacts based primarily on \( \text{PM}_{2.5} \) and \( \text{O}_3 \) exposures.

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 \( \text{PM}_{2.5} \) and \( \text{O}_3 \) 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.

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 \( \text{PM}_{2.5} \) and \( \text{O}_3 \). 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 \( \text{PM}_{2.5} \) and \( \text{O}_3 \) 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 Orellano et al. (2020), there are 87 (95% CI: 56-118) deaths attributable to the short-term exposure to wildfire-caused \( \text{PM}_{2.5} \). 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 O$_3$, the daily mean concentration is used, while the daily mean 8-h average (DMA8) for O$_3$. B&H stands for Bosnia and Herzegovina.

With regard to the excess deaths attributable to short-term O$_3$ 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 O$_3$ than to PM$_{2.5}$. As the relative risk of exposure to O$_3$ 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 O$_3$ due to a longer overall atmospheric lifetime of O$_3$ (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 O$_3$. 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 O$_3$ 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 O$_3$, 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 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 emissions from wildfires and subsequent secondary chemical and physical processes. To date, the health impact assessment of air 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 simplification 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 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 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 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 underestimation of particular matter and an overestimation of O$_3$ in urban areas (Im et al., 2015a). Due to its complex nature, the 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 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).

Subject to the RR value used, the estimated health burdens in association with short-term O$_3$ 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 O$_3$, 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 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 PM$_{10}$, another widely used proxy indicator for 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 chemistry 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 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).

<table>
<thead>
<tr>
<th>Name</th>
<th>ISO-2</th>
<th>ISO-3</th>
<th>Pop. (in Mio.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Albania</td>
<td>AL</td>
<td>ALB</td>
<td>2.84</td>
</tr>
<tr>
<td>Bosnia and Herzegovina (B&amp;H)</td>
<td>BA</td>
<td>BIH</td>
<td>3.28</td>
</tr>
<tr>
<td>Bulgaria</td>
<td>BG</td>
<td>BGR</td>
<td>6.93</td>
</tr>
<tr>
<td>Cyprus</td>
<td>CY</td>
<td>CYP</td>
<td>1.21</td>
</tr>
<tr>
<td>Egypt</td>
<td>EG</td>
<td>EGY</td>
<td>102.33</td>
</tr>
<tr>
<td>Greece</td>
<td>GR</td>
<td>GRC</td>
<td>10.72</td>
</tr>
<tr>
<td>Croatia</td>
<td>HR</td>
<td>HRV</td>
<td>4.05</td>
</tr>
<tr>
<td>Israel</td>
<td>IL</td>
<td>ISR</td>
<td>9.22</td>
</tr>
<tr>
<td>Italy</td>
<td>IT</td>
<td>ITA</td>
<td>59.55</td>
</tr>
<tr>
<td>Lebanon</td>
<td>LB</td>
<td>LBN</td>
<td>6.83</td>
</tr>
<tr>
<td>Montenegro</td>
<td>ME</td>
<td>MNE</td>
<td>0.62</td>
</tr>
<tr>
<td>North Macedonia (N. Macedonia)</td>
<td>MK</td>
<td>MKD</td>
<td>2.07</td>
</tr>
<tr>
<td>Malta</td>
<td>MT</td>
<td>MLT</td>
<td>0.53</td>
</tr>
<tr>
<td>Romania</td>
<td>RO</td>
<td>ROU</td>
<td>19.29</td>
</tr>
<tr>
<td>Serbia</td>
<td>RS</td>
<td>SRB</td>
<td>6.91</td>
</tr>
<tr>
<td>Slovenia</td>
<td>SI</td>
<td>SVN</td>
<td>2.10</td>
</tr>
<tr>
<td>Tunisia</td>
<td>TN</td>
<td>TUN</td>
<td>11.82</td>
</tr>
<tr>
<td>Turkey</td>
<td>TR</td>
<td>TUR</td>
<td>84.34</td>
</tr>
<tr>
<td>SUM</td>
<td></td>
<td></td>
<td>334.62</td>
</tr>
</tbody>
</table>
Table A2. Excess number of deaths for individual countries and the entire region estimated based on wildfires-caused PM$_{2.5}$ and O$_3$ 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.

<table>
<thead>
<tr>
<th>Country</th>
<th>Method I (PM$_{2.5}$)</th>
<th>Method II (PM$_{2.5}$)</th>
<th>Method I (O$_3$)</th>
<th>Method II (O$_3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Albania</td>
<td>2 [1,2]</td>
<td>2 [1,2]</td>
<td>5 [3,6]</td>
<td>5 [3,6]</td>
</tr>
<tr>
<td>B&amp;H</td>
<td>1 [1,2]</td>
<td>1 [1,2]</td>
<td>3 [2,4]</td>
<td>3 [2,4]</td>
</tr>
<tr>
<td>Croatia</td>
<td>1 [1,2]</td>
<td>1 [1,2]</td>
<td>4 [3,5]</td>
<td>4 [3,5]</td>
</tr>
<tr>
<td>Cyprus</td>
<td>0 [0,0]</td>
<td>0 [0,0]</td>
<td>1 [0,1]</td>
<td>1 [0,1]</td>
</tr>
<tr>
<td>Egypt</td>
<td>9 [6,13]</td>
<td>9 [6,13]</td>
<td>48 [33,63]</td>
<td>48 [34,64]</td>
</tr>
<tr>
<td>Israel</td>
<td>1 [0,1]</td>
<td>1 [0,1]</td>
<td>4 [3,4]</td>
<td>4 [3,4]</td>
</tr>
<tr>
<td>Lebanon</td>
<td>1 [0,1]</td>
<td>1 [0,1]</td>
<td>2 [2,3]</td>
<td>2 [2,3]</td>
</tr>
<tr>
<td>Malta</td>
<td>0 [0,0]</td>
<td>0 [0,0]</td>
<td>0 [0,0]</td>
<td>0 [0,0]</td>
</tr>
<tr>
<td>Montenegro</td>
<td>1 [0,1]</td>
<td>1 [0,1]</td>
<td>1 [1,2]</td>
<td>1 [1,2]</td>
</tr>
<tr>
<td>N. Macedonia</td>
<td>1 [1,2]</td>
<td>1 [1,2]</td>
<td>4 [3,6]</td>
<td>4 [3,6]</td>
</tr>
<tr>
<td>Romania</td>
<td>15 [10,21]</td>
<td>15 [10,21]</td>
<td>46 [34,57]</td>
<td>46 [34,58]</td>
</tr>
<tr>
<td>Slovenia</td>
<td>0 [0,0]</td>
<td>0 [0,0]</td>
<td>1 [1,1]</td>
<td>1 [1,1]</td>
</tr>
<tr>
<td>Tunisia</td>
<td>1 [1,2]</td>
<td>1 [1,2]</td>
<td>4 [3,6]</td>
<td>4 [3,6]</td>
</tr>
</tbody>
</table>
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. \( \sigma \) 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.

<table>
<thead>
<tr>
<th>Metric</th>
<th>Expression</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean Bias (MB)</td>
<td>( \frac{1}{N} \sum_{i=1}^{N} (M_i - O_i) )</td>
<td>((-\infty, +\infty))</td>
</tr>
<tr>
<td>Normalized Mean Bias (NMB)</td>
<td>( \sum_{i=1}^{N} (M_i - O_i) / \sum_{i=1}^{N} O_i )</td>
<td>((-\infty, +\infty))</td>
</tr>
<tr>
<td>Normalized standard deviation</td>
<td>( \sigma_M / \sigma_O )</td>
<td>([0, +\infty))</td>
</tr>
<tr>
<td>Root mean square error (RMSE)</td>
<td>( \sqrt{\frac{1}{N} \sum_{i=1}^{N} (M_i - O_i)^2} )</td>
<td>([0, +\infty))</td>
</tr>
<tr>
<td>Correlation coefficient (( \rho ))</td>
<td>( \sum_{i=1}^{N} (M_i - \bar{M})(O_i - \bar{O}) / (\sigma_M \sigma_O) )</td>
<td>([0, 1])</td>
</tr>
<tr>
<td>Centred RMSE</td>
<td>( \sqrt{\sigma_M^2 + \sigma_O^2 - 2\sigma_M \sigma_O \rho} )</td>
<td>([0, +\infty))</td>
</tr>
<tr>
<td>Index of agreement (IOA)</td>
<td>( 1 - \frac{\sum_{i=1}^{N} (M_i - O_i)^2}{\sum_{i=1}^{N} [(M_i - \bar{O}) + (O_i - \bar{O})]^2} )</td>
<td>([0, 1])</td>
</tr>
</tbody>
</table>
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 O$_3$. It has a mediocre performance for NO$_2$, while it scores lowest in estimating SO$_2$. 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 O$_3$. The bias for NO$_2$, albeit small, fluctuates considerably among stations, and the same fluctuations are observed for SO$_2$. The particularly problematic simulations for SO$_2$ may be ascribed to the model configuration that the point source emissions of SO$_2$ (e.g., from power plants and industrial sites) are assigned directly to the immediate surface, leading a considerable overestimation of the surface ambient concentration.

<table>
<thead>
<tr>
<th></th>
<th>MB [$\mu g/m^3$]</th>
<th>NMB</th>
<th>RMSE [$\mu g/m^3$]</th>
<th>IOA</th>
</tr>
</thead>
<tbody>
<tr>
<td>PM$_{2.5}$</td>
<td>day</td>
<td>-4.60 (2.97)</td>
<td>-0.39 (0.20)</td>
<td>6.45 (2.45)</td>
</tr>
<tr>
<td></td>
<td>hour</td>
<td>-6.54 (3.20)</td>
<td>-0.50 (0.19)</td>
<td>9.21 (3.36)</td>
</tr>
<tr>
<td>O$_3$</td>
<td>day</td>
<td>25.74 (19.18)</td>
<td>0.45 (0.39)</td>
<td>32.29 (12.99)</td>
</tr>
<tr>
<td></td>
<td>hour</td>
<td>25.79 (19.21)</td>
<td>0.46 (0.39)</td>
<td>38.06 (11.74)</td>
</tr>
<tr>
<td>NO$_2$</td>
<td>day</td>
<td>2.32 (7.41)</td>
<td>0.29 (0.61)</td>
<td>7.55 (5.12)</td>
</tr>
<tr>
<td></td>
<td>hour</td>
<td>-0.48 (9.96)</td>
<td>0.13 (0.50)</td>
<td>13.93 (10.43)</td>
</tr>
<tr>
<td>SO$_2$</td>
<td>day</td>
<td>8.70 (20.17)</td>
<td>0.88 (1.75)</td>
<td>13.40 (19.54)</td>
</tr>
<tr>
<td></td>
<td>hour</td>
<td>5.49 (19.77)</td>
<td>0.38 (1.47)</td>
<td>15.26 (25.44)</td>
</tr>
</tbody>
</table>
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 0.9 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$_{2.5}$. 

https://doi.org/10.5194/nhess-2023-111
Preprint. Discussion started: 18 July 2023
© Author(s) 2023. CC BY 4.0 License.
Figure B2. Evaluation of $O_3$ concentration simulated by WRF-Chem using AirBase data. The AirBase observations of $O_3$ 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 $O_3$ variation drops slightly with the mean IOA decreasing from 0.65 to 0.51.
Figure B3. Evaluation of NO$_2$ concentration simulated by WRF-Chem using AirBase data. The model demonstrated a mediocre performance in simulating both daily and hourly mean of NO$_2$. 

https://doi.org/10.5194/nhess-2023-111
Preprint. Discussion started: 18 July 2023
© Author(s) 2023. CC BY 4.0 License.
Figure B4. Evaluation of SO$_2$ concentration simulated by WRF-Chem using AirBase data. It remains a huge challenge to simulate the near-surface concentration SO$_2$. The model is prone to overestimate the SO$_2$ concentration, as the point source SO$_2$ emissions from inventories are assigned to the immediate surface. However, as SO$_2$ is not a major constituent in wildfire smokes, the resulting impacts on the conclusions of this study is considered minimal.
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 NO$_2$ concentration for each country and the entire region of interest.
Figure C4. Diurnal pattern of multi-month hourly mean SO$_2$ 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 NO\textsubscript{2} concentration simulated by WRF-Chem under the fires scenario and the difference relative to the no-fires scenario. a) NO\textsubscript{2} concentration simulated with fires scenario, b) NO\textsubscript{2} concentration with fires scenario relative to no-fires (\(\Delta\text{NO}_2 = \text{NO}_2\text{fires} - \text{NO}_2\text{no-fires}\)), c) NO\textsubscript{2} concentration attributable to wildfires in percentages (\(\Delta\text{NO}_2/\text{NO}_2\text{fires}\)). The elevation of wildfire-caused NO\textsubscript{2} is mainly observed in the Balkans, Greece, and South Italy.
Figure D2. Spatial pattern of multi-month mean SO$_2$ concentration simulated by WRF-Chem under the fires scenario and the difference relative to the no-fires scenario. a) SO$_2$ concentration simulated with fires scenario, b) SO$_2$ concentration with fires scenario relative to no-fires ($\Delta$SO$_2$ = SO$_2^{\text{fires}}$ - SO$_2^{\text{no-fires}}$), c) SO$_2$ concentration attributable to wildfires in percentages ($\Delta$SO$_2$/SO$_2^{\text{fires}}$). The elevation of wildfire-caused SO$_2$ 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 ($\Delta OH = OH_{\text{fires}} - OH_{\text{no-fires}}$), c) OH concentration attributable to wildfires in percentages ($\Delta OH/OH_{\text{fires}}$).
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

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.

Acknowledgements. The authors acknowledge the European Forest Fire Information System of the European Commission for providing the data on total number of wildfires and total area of burnt areas. We acknowledge use of the WRF-Chem preprocessor tools mozbc, bio_emiss, fire_emiss and anthro_emiss provided by the Atmospheric Chemistry Observations and Modeling Lab (ACOM) of NCAR. The authors gratefully acknowledge computing time at the Augsburg Linux Compute Cluster (ALCC). Additional thanks go to Dr. David Jean du Preez for proofreading this paper.
References


https://doi.org/10.5194/nhess-2023-111
Preprint. Discussion started: 18 July 2023
© Author(s) 2023. CC BY 4.0 License.


