Science of the Total Environment 922 (2024) 171314

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Fires as a source of annual ambient $PM_{2.5}$ exposure and chronic health impacts in Europe

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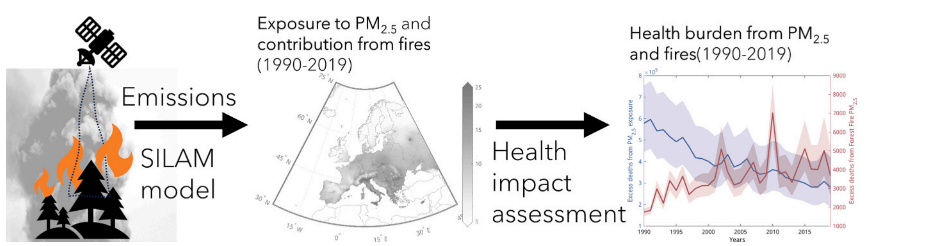
HIGHLIGHTS

- Population weighted $PM_{2.5}$ exposure and related health burden decreased over Europe between 1990 and 2019
- Excess deaths from fires increased by more than 100% during the same period.
- We found that the contribution of fires to excess deaths from $PM_{2.5}$ in Eastern Europe were at least three times higher as compared to Western and Central Europe
- Such increasing contribution from fires to ambient $PM_{2.5}$ exposure and related health burden are counteracting policies and efforts to achieve clean air across Europe

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Kai Zhang

Keywords:
Fires
Ambient PM2.5
Health burden
Fire mortality fractions

ABSTRACT

Chronic exposure to ambient $PM_{2.5}$ is the largest environmental health risk in Europe. We used a chemical transport model and recent exposure response functions to simulate ambient $PM_{2.5}$, contribution from fires and related health impacts over Europe from 1990 to 2019. Our estimation indicates that the excess death burden from exposure to ambient PM2.5 declined across Europe at a rate of 10,000 deaths per year, from 0.57 million (95 % confidence intervals: 0.44–0.75 million) in 1990 to 0.28 million (0.19–0.42 million) in the specified period. Among these excess deaths, approximately 99 % were among adults, while only around 1 % occurred among children. Our findings reveal a steady increase in fire mortality fractions (excess deaths from fires per 1000 deaths from ambient $PM_{2.5}$) from 2 in 1990 to 13 in 2019. Notably, countries in Eastern Europe exhibited significantly higher fire mortality fractions and experienced more pronounced increases compared to those in Western and Central Europe. We performed sensitivity analyses by considering fire $PM_{2.5}$ to be more toxic as compared to other sources, as indicated by recent studies. By considering fire $PM_{2.5}$ to be more toxic than other $PM_{2.5}$ sources results in an increased relative contribution of fires to excess deaths, reaching 2.5–13 % in 2019. Our results indicate the requirement of larger mitigation and adaptation efforts and more sustainable forest management policies to avert the rising health burden from fires.

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https://doi.org/10.1016/j.scitotenv.2024.171314
Received 26 October 2023; Received in revised form 29 January 2024; Accepted 25 February 2024
Available online 28 February 2024

1. Introduction

Long term exposure to ambient $PM_{2.5}$ (particulate matter <2.5 μm in diameter) is associated with multiple health outcomes, including morbidity and mortality from respiratory diseases, cardiovascular diseases, cerebrovascular diseases, acute lower respiratory infections, neoplasms of the lungs, diabetes, adverse birth outcomes and neonatal diseases (Brook et al., 2010; Cohen et al., 2017; Dockery and Pope III, 1994; Ito et al., 2011; Murray, 2015; Schraufnagel et al., 2019). Policies aiming to reduce the total $PM_{2.5}$ mass to avert the health impacts must target specific source sectors. Keeping pace with advancements in atmospheric chemistry models, development of emission inventories and satellite remote sensing, recent studies have informed about the sources of $PM_{2.5}$ at multiple scales, ranging from urban agglomerations to national scale (Chowdhury et al., 2022; Clappier et al., 2017; Khomenko et al., 2023; Lelieveld et al., 2015; Thunis et al., 2018; Upadhyay et al., 2018). Recent studies (Chowdhury et al., 2022; Lelieveld et al., 2015) have found that >75 % of the ambient $PM_{2.5}$ may be associated with anthropogenic sources but regional and seasonal variability is large. Although certain sources, such as the domestic use of solid fuels for cooking and household activities, play a substantial role, contributing to over 20 % of global PM2.5 exposure and related health effects (with even higher regional impacts), other sources like transportation, wild fires, industrial emissions, and power generation account for 5–10 % of ambient $PM_{2.5}$ exposure and health effects worldwide (Chowdhury et al., 2023, 2022).

Though much of the focus globally is towards mitigating anthropogenic sources of ambient $PM_{2.5}$ and other ambient air pollutants, controlling wildfires, which may cause episodes of extreme air pollution were until recently under-recognized as a potential source without credible mitigation options. With an increase in global population, more people are living in vulnerable areas close to forests. A recent study (Newton et al., 2020) found that about 1.6 billion of the Earth's rural population reside within 5 km of a forest which makes up 20 % of the global population. The World Bank also estimates about 1.6 billion people to be 'forest dependent', which increases human activity in and around forests (https://www.worldbank.org/en/news/feature/2016/03/18/why-forests-are-key-to-climate-water-health-and-livelihoods). In Europe, >95 % of the fires are human induced (Leone et al., 2009). Once a human induced fire starts, progressively warmer and drier conditions driven by climate change help to spread them, also such conditions make trees more susceptible to insects and pests, which weaken or kill trees, making the forests more susceptible.

The European Forest Fire Information System (https://effis.jrc.ec.europa.eu/) reported that Europe experienced the second most intense wildfire season in 2022, since 2000. Notably, much of Europe was hit by a record-breaking heatwave in 2022, which was accompanied by prolonged dry periods (Ballester et al., 2023). This made meteorological conditions conducive towards fire growth and smoke dispersion. Indeed, the emergence and intensity of wildfires arise from the intricate interplay of natural elements, including factors like the availability of fuel and occurrences of lightning, along with the human influences which include land management practices and ignition sources. Meteorological factors play a crucial role, particularly when they combine in ways that foster extreme fire weather conditions. The assessment of extreme fire weather often relies on fire weather indices, which integrate daily weather variables (including relative humidity, air temperature, wind speed, and precipitation) associated with fuel moisture and fire behavior (Flannigan and Harrington, 1988; Jolly et al., 2015). Over the last four decades, regions displaying increased temperature and wind speed, prolonged rain-free intervals, and decreased relative humidity have concurrently demonstrated an extended duration of the fire weather season, which is expected to increase under changing climate in the future (Im et al., 2022; Jain et al., 2022; Jolly et al., 2015; Lund et al., 2023; Miller et al., 2023; Richardson et al., 2022).

Wildfires emit numerous toxic mixtures of pollutants including black carbon, organic aerosols, carbon monoxide and oxides of sulfur and nitrogen into the air (Liu and Yang, 2023; Partanen and Sofiev, 2022; Singh, 2022). In close proximity to the fire, $PM_{2.5}$ levels can surpass WHO's 24-hour guideline by over 30 times (Bolaño-Diaz et al., 2022; Graham et al., 2021; Storey and Price, 2022). Additionally, pollutants emitted from these fires can stay in the air over long time facilitating their large-scale dispersion and impact of the population across continents (Gong and Wang, 2021; Hänninen et al., 2009; Holanda et al., 2023; Singh et al., 2022). Wildfire have been associated with multiple health outcomes on acute exposure (Chen et al., 2021, 2017; Cleland et al., 2021; Yu et al., 2023). A recent study found that each additional day of exposure to wildfire smoke was associated with 0.49 % (95 % CI 0.41–0.59 %) increase in risk of preterm birth (<37 weeks) in California (Heft-Neal et al., 2022). Multiple studies (Aguilera et al., 2021; Chen et al., 2021; Noah et al., 2023; Rice et al., 2021) have shown that wildfire smoke is positively associated with cardiovascular and respiratory morbidity and mortality including exacerbations of asthma and chronic obstructive pulmonary diseases (COPD). Borchers Arriagada et al. (2020) found that elevated exposure to $PM_{2.5}$ during Australian bushfires of 2019–2020 was associated with 417 (95 % CI 153–680) excess deaths and thousands of hospitalizations for cardiovascular and respiratory problems. Zhou et al. (2021) found that wildfire smoke amplified the effect of short-term exposure to $PM_{2.5}$ on COVID 19 in counties on the west coast of USA (United States of America). Consequently, long term exposure to wildfires was associated with premature mortality and increased morbidity including respiratory diseases, cancers, mental health and associated with health-related economic outcomes or health care costs (Grant and Runkle, 2022). Studies also find that wildfire smoke can be more toxic as compared to emissions from other sources like industries and power generation (Aguilera et al., 2021; Chen et al., 2021). Aguilera et al. (2021) found that wildfire smoke can result in 10 times higher risk of respiratory hospitalizations as compared to other sources of $PM_{2.5}$. Chowdhury and colleagues found that globally, >5 % of the deaths associated with chronic exposure to ambient $PM_{2.5}$ can be linked to forest fires, which increases further to 7.5 % upon considering anthropogenic organic aerosols and black carbon to be twice more toxic as compared to other components in $PM_{2.5}$.

Over the last couple of decades, the major sources of $PM_{2.5}$ emissions have changed due to factors including policy interventions, shifts in industrial and social practices and advancement of technologies (Crippa et al., 2020). In Europe, over the last few decades, multiple policy interventions were implemented aiming to reduce emissions from various major sources, including improving emission standards for vehicles, banning coal use for heating, introducing industrial emission controls, national air pollution control programs in multiple countries, among other measures. This has resulted in a decrease in anthropogenic emissions and exposure to $PM_{2.5}$ in most of Europe (Alikhani Faradonbeh et al., 2021; Gu et al., 2023; Jiang et al., 2020). Despite new insights on major sources of $PM_{2.5}$ by multiple new studies focusing on bottom-up techniques (Chowdhury et al., 2023; Lelieveld et al., 2015; McDuffie et al., 2021), only few studies have attempted to understand the long-term evolution of sources of $PM_{2.5}$ globally and in Europe (Gu et al., 2023) as such studies are often constrained by availability of computational resources. Specifically, there is little information on the evolution of wildfires, which has both human and climate dependance, as a source of $PM_{2.5}$ in Europe. In this study, we aim to explore the escalating significance of wildfires as a source of $PM_{2.5}$ exposure in Europe over a thirty-year period (1990–2019) by utilizing simulations from a global meso-scale dispersion model. Moreover, we assess the health impact caused by fire $PM_{2.5}$ and correlate changes in this burden over the three decades with shifts in baseline mortality rates epidemiology, demography, and $PM_{2.5}$ exposure. Additionally, we conduct sensitivity analyses based on recent evidence of heightened toxicity associated with emissions from the fires.

2. Methods

2.1. Model and emission inventory

We use the SILAM (System for Integrated modeLling of Atmospheric composition, http://silam.fmi.fi), an offline 3D chemical transport model (Kouznetsov and Sofiev, 2012; Sofiev, 2002; Sofiev et al., 2015, 2010) designed for atmospheric composition and air quality simulations at horizontal scales ranging from sub-kilometer scale and up to the whole globe. For the current study, SILAM was run at ~0.2° × 0.2° horizontal resolution with 19 vertical layers up to 106 hPa over continental Europe and surrounding islands. The model was run from 1979 to 2019, with the first year dismissed as a spin-up period for the model, of which data from 1990 to 2019 were used in this study. Boundary conditions were obtained from global SILAM simulations at horizontal resolution of ~2° × 2° and a temporal resolution of 3 h. The simulations were governed by meteorological variables obtained from the European reanalysis ERA5. The gas-phase chemistry, including secondary organic aerosols formation is based on CBM05 (Woody et al., 2015; Yarwood et al., 2010). The global SILAM simulations included stratospheric gas-phase reactions and heterogeneous reactions in polar stratospheric clouds (Damski et al., 2007; Kouznetsov et al., 2020; Sofiev et al., 2020). The aerosol microphysical processes (e.g. coagulation, condensation) are parameterized following the DMAT equilibrium-based model (Sofiev, 2000).

We used the monthly varying Community Emissions Data System (CEDS) anthropogenic emission inventory (Hoesly et al., 2018) at 0.5° × 0.5° resolution for the primary emitted species like SO_2 (sulfur dioxide), NOx (nitrogen oxides) (oxides of nitrogen), CO (carbon monoxide), BC (black carbon), OC (organic carbon), NH_3 (ammonia) and speciated NMVOCs (non-methane volatile organic compounds). For European scale runs the anthropogenic emissions were remapped to 0.1° × 0.1° grid using CAMS-GLOB (v2.1) emission for year 2015 as a proxy for the distribution inside a coarser grid of CEDS emissions.

Fire emissions posed a particular challenge for the study: homogeneous datasets based on active-fire or burnt-area observations and covering several decades do not exist, owing to changes of satellites, their capabilities, resolution, and sensitivity. Therefore, we used the dataset generated by the fire prediction model of Integrated System for wild-land fires, IS4FIRES (Soares et al., 2015; Sofiev et al., 2012, 2009). We distinguish 7 types of fuel associated with land-use: grass, agriculture waste, tropical forest, temperate forest, boreal forest, shrub, tundra. IS4FIRES data were previously used in a short-term evaluation of the fire-related mortality in Europe (Kollanus et al., 2017). The FFM (Fire Forecasting Model) of IS4FIRES is a time-agnostic machine-learning algorithm trained and evaluated against the MODIS Fire Radiative Power product, 2003–2022 (training period was 2003–2014, evaluation was performed over 2015–2020) (Sofiev et al., in prep.). An independent evaluation was performed against the GFEDv4 fire emission database, which is based on burnt-area observations, thus being independent from the active-fire observations.

DMS, sea-salt and mineral dust are generated interactively in the model driven mainly by meteorology. N2O emissions from soil (used only in global runs) is a static map and NOx emissions from lightning are based on climatology (from GEIA – Global Emissions Initiative inventory). In global runs biogenic emission are from MEGAN and CAMS-BIO inventories while the European scale simulations directly use the MEGAN model for biogenic isoprene and monoterpene emissions.

Fire originated PM emissions are tracked by having separate variables for particulate matter from fires. Fire source is taken explicitly in the simulations. The wild and vegetation fires (together termed as 'fires') were assumed to release a "fire-induced PM", which has a prescribed size distribution but no specific chemical composition, i.e., the fire-induced aerosol is a total PM originated from fires, including primary PM released by the fire and secondary PM produced from the fire-originated precursors. The transformations are assumed to occur fast enough, so that at the regional/continental scale they can be considered instant. Apart from eliminating the source apportionment step with its uncertainties, this approach allowed for a direct identification of emission factors by fitting the SILAM predictions to the satellite, Aeronet sun-photometer network, and in-situ total-PM measurements (see SI Text). SILAM has also been extensively evaluated in numerous international retrospective studies (Blechschmidt et al., 2020; Kukkonen et al., 2012; Marécal et al., 2015; Petersen et al., 2019) and real-time operational applications (http://atmosphere.copernicus.eu, https://dust.aemet.es, https://ews.tropmet.res.in, http://www.asdf-bj.net/gafis/index.html, https://hpfx.collab.science.gc.ca/~svfs000/na-aq-mm-fe/dist/, https://www.nrlmry.navy.mil/aerosol/).

2.2. Estimation of excess deaths

Combined with the generated data on exposure to $PM_{2.5}$, we use the recently formulated MR-BRT (meta regression Bayesian, regularized, trimmed) (meta-regression-Bayesian, regularized, trimmed) exposure response function (Murray et al., 2020; Pozzer et al., 2023), also used in the 2019 iteration of Global Burden of Disease, to estimate the age-categorized relative risks (RR) for exposure to ambient $PM_{2.5}$. MR-BRT includes cause-specific risk expressions for ischemic heart disease (IHD), stroke (ischemic and hemorrhagic), chronic obstructive pulmonary disease (COPD), lung cancer (LC) and Type II diabetes (T2DM) among adults (population with age > 25 years), acute lower respiratory tract infection (ALRI) among children (population under the age of 5). The splines in MR-BRT are fitted using information on RR and $PM_{2.5}$ exposure from epidemiological studies of ambient $PM_{2.5}$, household air pollution and secondhand smoking. The excess death burden was estimated at 5 × 5km resolution, linearly interpolating the $PM_{2.5}$ exposure simulated by SILAM, and was estimated by age and disease category, as in our earlier studies by

$$M_{c,a,d} = P_a \times BM_{a,d} \times \frac{RR_{c,a,d} - 1}{RR_{c,a,d}}$$

Excess deaths were estimated separately for adults and children at each 5 year intervals. *RR* (*c,a,d*) were derived using MR-BRT functions for all diseases by age, where *c,a,d* denotes concentration of $PM_{2.5}$, population age and disease respectively. Age specific RRs (Relative Risks) for IHD and stroke, are obtained using MR-BRT. For LC, T2-DM, and COPD uniform RR (*c,d*) were used across all age groups among adults. BM (*a,d*) is the baseline mortality rate per 100,000 population, obtained from the GBD (http://ghdx.healthdata.org/gbd-results-tool) for all countries in Europe and considered to remain uniform within a country at 5 × 5km resolution by age and disease. *P* (*a*) is the exposed population in a grid by age; the age distributions at 5-year intervals (adults > 25 years), and <5 years for children were obtained from the Global Burden of Disease (GBD) (http://ghdx.healthdata.org/record/ihme-data/gbd-2019-population-estimates-1950–2019) which are then merged with the latest population data at about 1 × 1 km (re-gridded to 5 × 5 km) horizontal resolution from Global Human Settlement dataset (https://ghsl.jrc.ec.europa.eu/datasets.php) to obtain the age-specific population (P(*a*)) at each 5 × 5 km grid. The Global Human Settlement dataset is available at every five-year interval from 1975 to the present, which we interpolate at yearly resolution.

Studies (Aguilera et al., 2021; Chen et al., 2021) have found that wildfires can be more toxic as compared to other $PM_{2.5}$ owing to its chemical composition and smaller sizes. A large share of $PM_{2.5}$ emissions from fires are black carbon, primary organic carbon and secondary organic carbon which may result in higher oxidative stress as compared to other components in $PM_{2.5}$. It should be noted that widely used exposure response functions like the Integrated Exposure-Response function (IER), Global Exposure Mortality Model and MR-BRT are formulated by assuming equal toxicity for all the components and sources of $PM_{2.5}$ (Burnett et al., 2018, 2014; Burnett and Cohen, 2020). To account for which we perform sensitivity studies by considering fire

$PM_{2.5}$ to be n ($n = 2,5,10$) times more toxic as compared to other sources of $PM_{2.5}$ using methods described in our previous study (Chowdhury et al., 2022). In brief, to assume fire $PM_{2.5}$ to be n times more toxic, the concentration of fire $PM_{2.5}$ was multiplied by n and the contribution from other sources were decreased accordingly, keeping the total $PM_{2.5}$ and associated excess deaths constant.

2.3. Changes in different factors

We also calculate the variation in excess deaths caused by $PM_{2.5}$ exposure for each year in comparison to 1990. We then attribute the changes observed in each country to changes in key factors, namely, baseline mortality rates, population size, population age structure and $PM_{2.5}$ exposure. To assess the relative importance of each individual factor, as previously detailed in our study (Chowdhury et al., 2020), we estimate the excess death burden for a specific year by introducing the factor of interest for that corresponding year while keeping all other factors constant at 1990 levels. Therefore, the difference between excess deaths for that year and 1990 represents the impact of that specific factor on the total excess deaths. The cases, 'BM', 'Population size', 'Population age' and 'Exposure' were developed to derive the impact of changing baseline mortality, population size, population age structure, and PM2.5 exposure respectively on excess death burden for a consequent year relative to 1990 levels. Important to consider is that the fractional contributions of the transitional factors are not additive and require cautious interpretation.

3. Results

3.1. Trends in exposure to ambient $PM_{2.5}$

To simulate the surface concentration of ambient $PM_{2.5}$, we utilized the SILAM chemical transport model (Sofiev et al., 2015), which was thoroughly validated against multiple measurement datasets and satellite retrievals. Please refer to the Supplementary Information, Figs. S1–S3 and SI Text for further details on the $PM_{2.5}$ evaluation. Our study primarily concentrates on understanding and estimating the chronic health impacts resulting from exposure to ambient $PM_{2.5}$ in Europe, with a specific focus on the contribution of fires. Therefore, we specifically examine annual averages of ambient $PM_{2.5}$ and the role of fires. The investigation of daily and seasonal variations of ambient $PM_{2.5}$ and fire contributions are beyond the scope of this study.

Our simulations show that the population weighted $PM_{2.5}$ exposure across Europe declined from 22.3 μg/m3 in 1990 to 10.8 μg/m^3 in 2019 (Fig. 1). It was simulated to decrease by 53 %, 52 % and 46 % in Western, Central and Eastern Europe, respectively in 2019 as compared to 1990 (Fig. 1.), supported by observational data. Fig. S4 illustrates the three regions in Europe. Significant decreases were also simulated in the populated countries of Germany (63 %), France (55 %), Spain (47 %) and Italy (43 %). Population weighted $PM_{2.5}$ for all the countries and the three regions of Europe can be found in SI Data1.

Our simulations showed that population weighted $PM_{2.5}$ decreased at rates of 0.37 μg/m^3, 0.34 μg/m^3, and 0.2 μg/m^3 per year over Western, Central, and Eastern Europe, respectively. The declining trend was more pronounced in the late 1990s. Over the period 1990–1999, $PM_{2.5}$ exposure was reduced by >35 % in all the European countries compared to 1990 levels. The population weighted PM2.5 by country over Europe for both 1990 and 2019 can be seen in Fig. S5. Such profound improvement in $PM_{2.5}$ levels were largely driven by multiple directives enforced in the last decade of the 20th century including the Large Combustion Plant Directive of 1988, introduction of EURO standards for light and heavy-duty vehicles in 1992, emission limits for engines used in non-road mobile machinery of 1997, VOC (volatile organic compounds) Stage 1 directive of 1994.

The improvement in air quality continued into the 21st century supported by the 2008 Ambient Air Quality Directive and more recently adopting the European Union (EU) Directive on National Emission reduction Commitments (NEC) of 2016. Across Europe, on average, fine particulate matter emissions decreased by ~40 % from 1990 to 2004 and ~25 % between 2005 and 2019, reflected on the trends of ambient $PM_{2.5}$ exposure. We estimate 34, 31, 63 and 17 % of population in Europe, Western, Central and Eastern Europe to be exposed to European $PM_{2.5}$ standard of 25 μg/m^3 respectively in 1990, which decreased to 3, 3, 0 and 3 % in 2019 respectively. The percentage of population in all

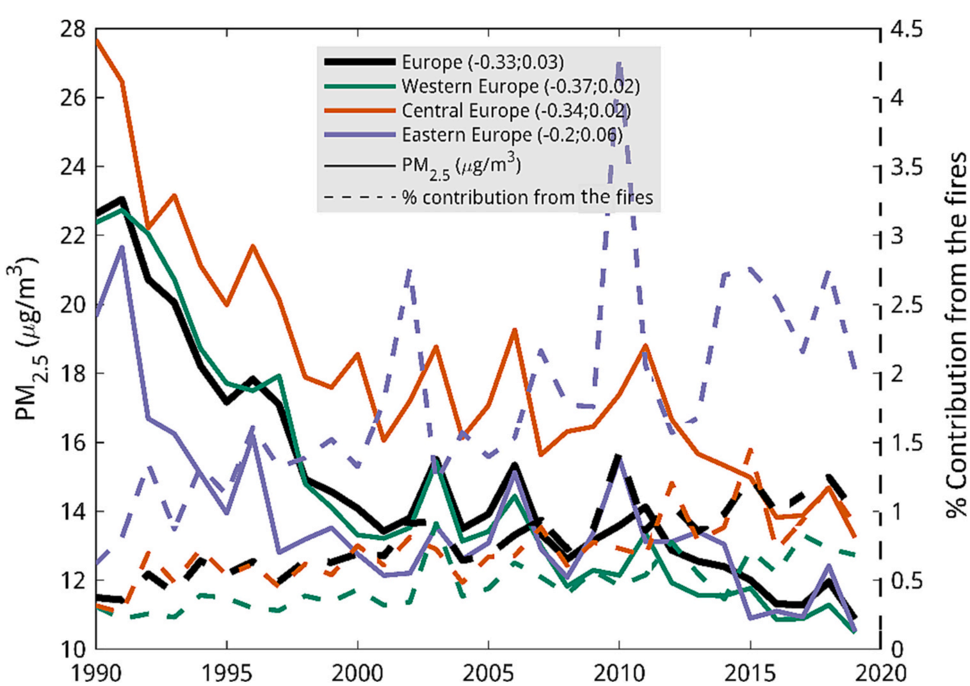

Fig. 1. Population weighted $PM_{2.5}$ exposure (solid lines) and percentage contribution of fires (dotted lines) over Europe (black), Western (green), Central (brown) and Eastern Europe (purple) from 1990 to 2019. The numbers in parenthesis indicate the rate of change of population weighted $PM_{2.5}$ and changing percentage contribution of fires in units/year.

European countries residing in locations exceeding the European standard, WHO (World Health Organization) guideline and Interim Targets for years 1990–2019 are listed in SI Data2.

3.2. Contribution from fires

We observe a rising trend in the contributions of fires over the 30-year period 1990–2019 (Fig. 1). Our estimation indicates that, in 1990, fires accounted for 0.3 % of the population weighted ambient $PM_{2.5}$. Its share increased to 1.2 % in 2018 and 1 % in 2019 at a rate of 0.03 % per year. The absolute exposure to fire $PM_{2.5}$ also increased from 0.08 μg/m^3 in 1990 to 0.15 μg/m^3 in 2018 and 0.1 μg/m^3 in 2019. This underscores the growing significance of fires as a source of ambient PM2.5 exposure in Europe, particularly when taking into account the decreasing emissions from sources such as industries, transportation, and power generation.

We find large heterogeneity in the contribution of fires to ambient $PM_{2.5}$ across Europe. In Russia and Ukraine, the contribution from fires were found to be ~2 % in 2019, an increase of >100 % as compared to the 1990 levels. Fig. 2 (a, b) depicts the contribution of fires to population weighted $PM_{2.5}$ by country in Europe for 1990 and 2019 respectively. The rate of change in the contribution of fires to ambient $PM_{2.5}$ between 1990 and 2019 reveals a positive trend across all European countries (Fig. 2c). The trend is especially pronounced in Eastern European and Mediterranean countries, including Spain, Portugal, and Greece. The absolute exposure to fire $PM_{2.5}$ also increased at least twofold in most Eastern European and Mediterranean countries. We find peak contributions of fires in 2010 over Europe, largely driven by prevalent fires in Eastern Europe. While the 2003 fires in Western Europe led to ~50 % increase in annual exposure to fire $PM_{2.5}$ over Western Europe as compared to the previous year. Please see SI Data3 for information on the contribution of fires by country and year.

3.3. Excess deaths from ambient $PM_{2.5}$ and contribution from fires

Using the MR-BRT exposure response functions, we estimate the excess death burden from exposure to ambient $PM_{2.5}$ to decrease over Europe from 0.57 million (95 % confidence intervals 0.44–0.75) million in 1990 to 0.28(0.19–0.42) million, of which ~99 % and <1 % occur among adults and children respectively (Figs. 3, S6). Fig. 3 depicts the trend in excess mortality burden from exposure to ambient $PM_{2.5}$ in Europe from 1990 to 2019. Russia (42 (29–64 CI: 95 %) thousand) is estimated to have the highest annual excess deaths in Europe followed by Ukraine (36 (23–57) thousand) and Italy (35 (26–47) thousand) respectively, in 2019. We find that excess deaths from ambient $PM_{2.5}$ exposure decreased in most European countries, most notably in Germany (from 84 (66–110) thousand in 1990 to 25 (13–39) thousand in 2019) and United Kingdom (from 35 (26–48) thousand in 1990 to 9 (6–13) thousand in 2019) at a rate of 1685 and 1085 deaths per year respectively. In Russia, Ukraine, and Italy the excess death burden decreased at a rate of 871, 1064 and 776 deaths/year respectively. However, in Central European countries of Albania, Macedonia and Montenegro, excess deaths from ambient $PM_{2.5}$ increased at a rate of 14, 5 and 3 deaths/year respectively. Table 1 lists the health burden among adults for the top five countries ranked by total excess deaths in 2015. Fig. S7 depicts the spatial distribution of excess deaths by country in Europe for 1990 and 2019 and data for all the European countries across the 30 years are listed in SI Data4. Overall, we find excess deaths to decrease more significantly in Western Europe (from 2.7 (2–3.5) million in 1990 to 1.1 (0.8–1.6) million in 2019) as compared to in Eastern

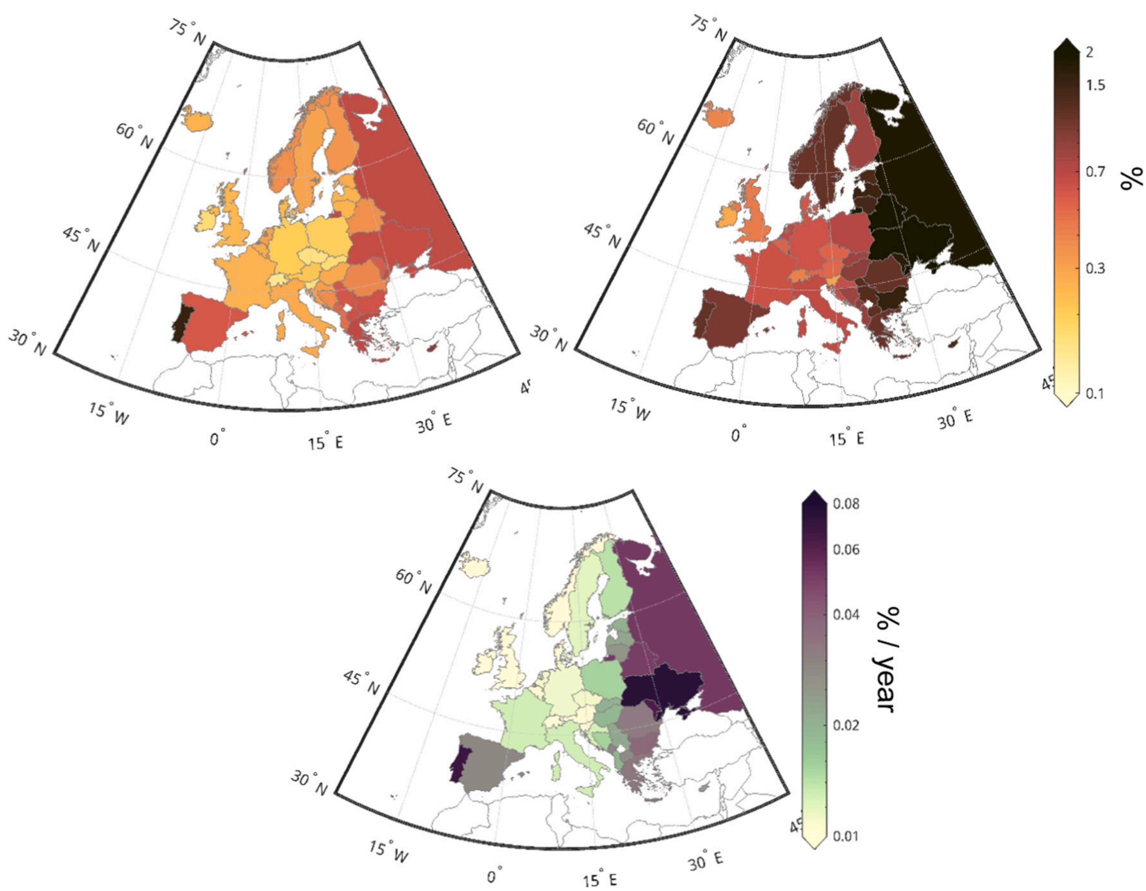

Fig. 2. (a) Spatial distribution (by country) of percentage contribution of forest fires to population weighted ambient $PM_{2.5}$ exposure in 1990 and (b) 2019. (c) Spatial distribution rate of change (%/year) of forest fire's contribution to ambient $PM_{2.5}$.

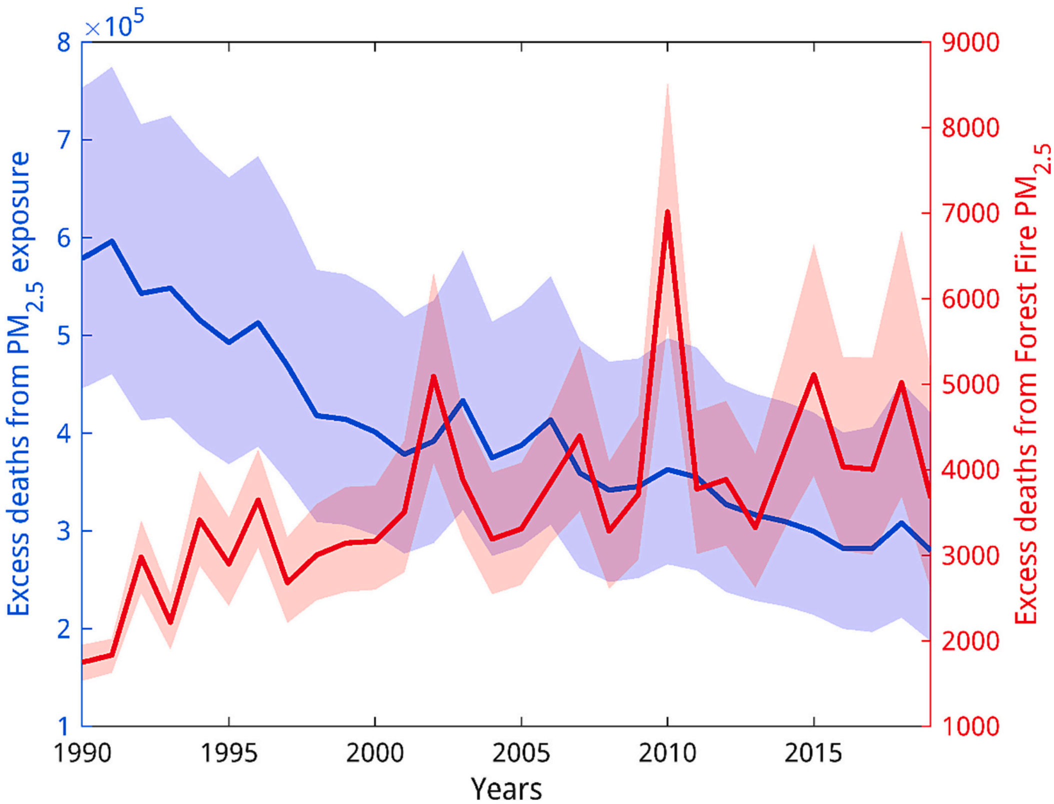

Fig. 3. Excess deaths from ambient $PM_{2.5}$ exposure (in blue lines) and from fire $PM_{2.5}$ (in red lines) in Europe from 1990 to 2019.

Europe (from 1.5 (1.1–2) million in 1990 to 0.8 (0.5–1.3) million in 2019) and Central Europe (from 1.1 (0.8–1.6) million in 1990 to 0.7 (0.5–1.1) million in 2019) respectively (Table 2).

In 1990, we estimated 90 (69 to 117) excess deaths per 100,000 population (excess death rates) in Europe due to exposure to ambient $PM_{2.5}$ which declined significantly to 42 (27 to 62) in 2019. The corresponding PM2.5-related excess death rates decreased significantly in the three European regions from 1990 to 2019 (Table 2), however they are significantly higher in Eastern and Central Europe as compared to those in Western Europe (Tables 1, 2). We estimate that IHD (48 %) and stroke (26 %) are the leading causes of death from exposure to ambient $PM_{2.5}$ in Europe in 2019, followed by lung cancer (13 %), COPD (8 %) and T2-DM (6 %) (Fig. S6). The distribution of excess deaths by disease varies considerably by European regions (SI Data5). Notably, our estimates for excess deaths in Europe are considerably lower (less than half) as compared to previous studies (Chowdhury et al., 2020; Lelieveld et al., 2019) that use the Global Exposure Mortality Model (GEMM), while being comparable to with the most recent estimate from GBD (Chowdhury et al., 2022; Murray, 2015). The reasons for such differences are described in our previous studies and elsewhere (Chowdhury et al., 2022; Pozzer et al., 2023).

While we estimate the excess deaths from exposure to ambient $PM_{2.5}$ to decrease significantly in Europe, by >50 % from 1990 to 2019, the excess deaths attributable to fires were estimated to increase by >100 %, from 1700 (1500–2000) in 1990 to 3700(2600–5100) in 2019. Spatial distribution of excess deaths per country from fire $PM_{2.5}$ in 1990 and 2019 over Europe are depicted in Fig. S8. We estimate that fires contribute to 0.7 %, 0.9 % and 2.3 % of the total excess deaths in Western, Central and Eastern Europe respectively in 2019, which are considerably higher compared to their contribution in 1990 (0.23, 0.21 and 0.5 % in Western, Central and Eastern Europe respectively).

We find that in 2019, 13 (9–18) out of 1000 excess deaths from ambient $PM_{2.5}$ exposure to be associated with fire $PM_{2.5}$ (termed as 'fire mortality fractions'), an increase from 2 (2.6–3.4) in 1990 (Figs. 4, 5, S9). The increase in fire mortality fractions from 1990 to 2019 were found to be considerably higher in Eastern Europe (5 (4.4–5.6) in 1990 to 23.9 (16.5–34.5) in 2019) as compared to Western and Central Europe (Table 2, Fig. S9). In Russia and Ukraine, the fire mortality fractions increased from 5.8 (5–6.6) and 4.8 (4.3–5.2) in 1990 to 21.9 (15.4–30.8) and 27 (18.5–39.2) in 2019 respectively (Table 1, Figs. 4, S9). In Spain and Portugal, the corresponding fire mortality fractions were found to increase from 5 (4–6) and 13 (11–15) in 2019 to 12.1 (8.5–17.1) and 14.7 (10.2–21.2) respectively (Fig. 4). We also found a considerable increase in fire mortality fractions in Scandinavian countries of Norway (5.1 (3.8–6.2) in 1990 to 22.1 (14.4–33.2)) and Sweden (3.3 (2.4–4.5) in 1990 to 18.8 (12.9–28.4) in 2019). Comparably, the fire mortality fractions were found to be relatively lower in highly populated Western European countries of Germany, France, and Great Britain (Fig. 4). The fire mortality fractions for all the countries from 1990 to 2019 are listed in SI Data6.

3.4. Sensitivity studies

The fires release large proportions of very fine organic aerosols, often at sub-micron scales, which are more toxic compared to emissions from other sources due to their chemical composition and size. A recent study found that wildfire $PM_{2.5}$ may be more toxic as compared to other sources of $PM_{2.5}$, resulting in ten times more respiratory hospitalizations (Aguilera et al., 2021). Based on this and other recent evidence, we assume fire $PM_{2.5}$ to be 10 times more toxic (10T) as compared to other $PM_{2.5}$. We also make alternative assumptions of fire $PM_{2.5}$ being 2 and 5 times more toxic (2T and 5T) as compared to other $PM_{2.5}$. Under the 10T assumption, we find that fires may contribute to ~13.6 % of total $PM_{2.5}$ related excess deaths in 2019 over Europe as compared to 1.3 % under equality toxicity (EqT) assumption (Fig. 6, Table 1). The corresponding contributions under 2T and 5T assumptions are 2.6 % and 6.7 % respectively. We find that under the 10T assumption, 25 % of the total excess deaths in Eastern Europe may be associated with fires in 2019, as

Table 1
Population weighted $PM_{2.5}$, related excess deaths and age standardized mortality rates (excess deaths from $PM_{2.5}$/1000 population), fire mortality fractions and fire mortality fractions by considering forest fire $PM_{2.5}$ to be 2, 5 and 10 times more toxic as compared to other sources of $PM_{2.5}$. The numbers are listed for 2019 and 1990 and the top 5 countries sorted by the excess deaths estimated for 2019.

Country	Population Weighted $PM_{2.5}$		Excess deaths from $PM_{2.5}$		Excess deaths from $PM_{2.5}$/100000 population		Fire mortality fractions (excess deaths from forest fires/1000 excess deaths from $PM_{2.5}$)							
							Considering all $PM_{2.5}$ to be equally toxic		Considering forest fires to be 2× more toxic		Considering forest fires to be 5× more toxic		Considering forest fires to be 10× more toxic	
	1990	2019	1990	2019	1990	2019	1990	2019	1990	2019	1990	2019	1990	2019
Russia	18.4	11.1	67 (52–86)	43 (29–64)	92 (72–119)	58 (39–86)	5.8 (5–6.6)	21.9 (15.4–30.8)	11.7 (10.2–13.2)	43.8 (30.9–62.1)	29.6 (25.7–33.5)	111.7 (77.9–159.7)	60.1 (51.9–68.6)	230.4 (158.2–335.7)
Ukraine	22.8	10.1	66 (51–86)	36 (23–57)	129 (99–168)	82 (52–130)	4.8 (4.3–5.2)	27 (18.5–39.2)	9.6 (8.5–10.5)	54.3 (37.1–79.1)	24.1 (21.4–26.4)	138.4 (93.9–202.8)	48.9 (43.3–53.7)	285.1 (191.2–423.7)
Italy	36.9	21	56 (45–69)	35 (26–47)	98 (80–121)	59 (44–79)	1.6 (1.4–1.6)	5.1 (4.3–5.9)	3.1 (2.9–3.3)	10.1 (8.6–11.8)	7.9 (7.3–8.3)	25.5 (21.6–29.8)	15.9 (14.7–16.8)	51.8 (43.7–60.6)
Germany	25.8	9.7	84 (65–110)	26 (17–39)	106 (82–138)	31 (21–46)	1.3 (1.2–1.5)	6.8 (8.1–13.2)	2.7 (2.4–2.9)	13.7 (9.8–19)	6.7 (6.1–7.3)	34.3 (24.5–47.9)	13.5 (12.1–14.7)	69.1 (49.2–96.7)
Poland	23.8	11.7	39 (31–49)	17 (11–26)	102 (81–129)	45 (29–68)	1.4 (1.3–1.5)	7.4 (5.3–10.3)	2.8 (2.6–2.9)	14.8 (10.6–20.6)	6.9 (6.4–7.3)	37.1 (26.5–51.8)	13.9 (12.8–14.7)	107.9 (83.4–138.1)

Table 2
Same as Table 1, but for the 3 major regions.

Country	Population Weighted $PM_{2.5}$		Excess deaths from $PM_{2.5}$		Excess deaths from $PM_{2.5}$/100000 population		Fire mortality fractions (excess deaths from forest fires/1000 excess deaths from $PM_{2.5}$)							
							Considering all $PM_{2.5}$ to be equally toxic		Considering all $PM_{2.5}$ to be equally toxic		Considering all $PM_{2.5}$ to be equally toxic		Considering all $PM_{2.5}$ to be equally toxic	
	1990	2019	1990	2019	1990	2019	1990	2019	1990	2019	1990	2019	1990	2019
Western Europe	22.4	10.5	2.7 (2–3.5)	1.1 (0.8–1.6)	75 (57–98)	28 (19–40)	2.4 (2–2.7)	7.3 (5.4–9.9)	4.7 (4–5.4)	14.6 (10.8–19.8)	11.9 (10.2–13.6)	36.8 (21.2–49.8)	24.1 (20.7–27.7)	74.3 (54.7–100.9)
Central Europe	27.7	13.2	1.4 (1.1–1.9)	0.7 (0.5–1.1)	122 (95–156)	64 (42–100)	2.1 (1.9–2.3)	9.4 (6.8–12.9)	4.3 (3.9–4.6)	18.8 (13.5–26)	10.8 (9.8–11.5)	47.6 (34–65.9)	21.7 (19.7–23.3)	96.7 (68.7–134.8)
Eastern Europe	19.7	10.5	1.5 (1.1–2)	0.8 (0.5–1.3)	104 (80–135)	65 (42–100)	5 (4.4–5.6)	23.9 (16.5–34.5)	10.1 (8.9–11.3)	48 (33.1–70)	25.5 (22.3–28.6)	122.3 (83.6–178.4)	51.7 (45.1–58.3)	251.9 (169.8–373)

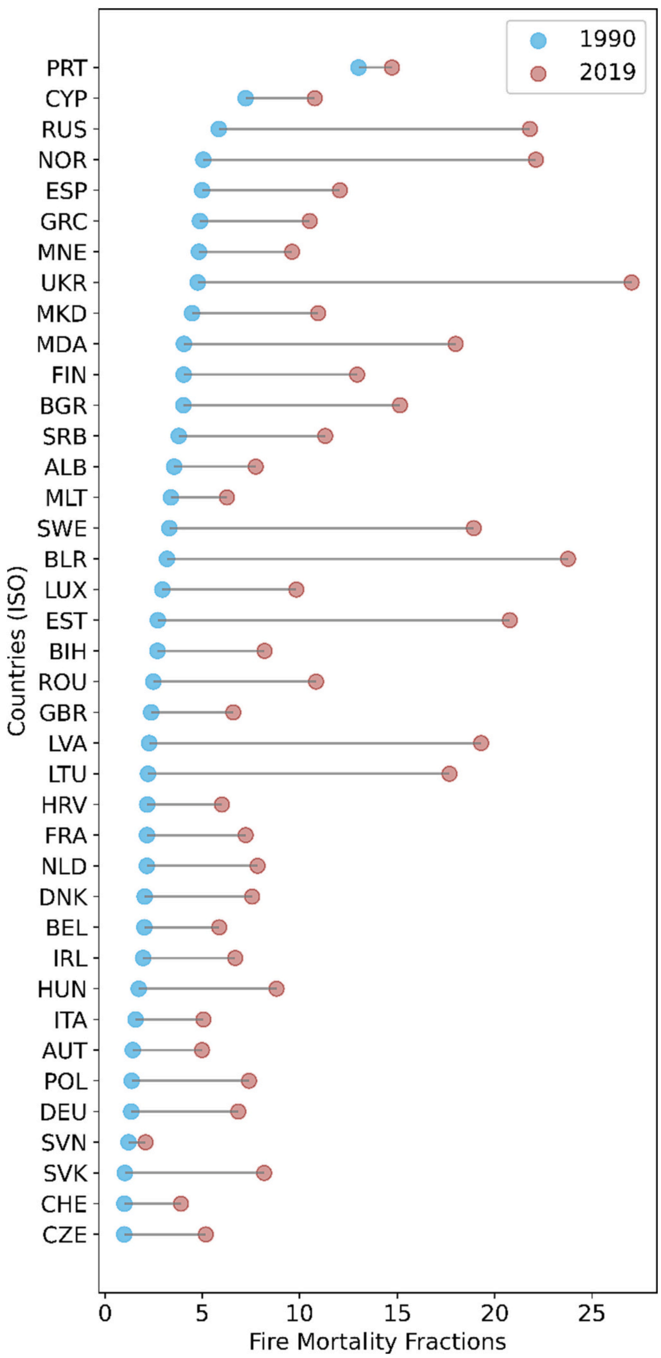

Fig. 4. Fire mortality fractions (excess deaths from fire $PM_{2.5}$/1000 excess deaths from ambient $PM_{2.5}$ exposure for 1990 (blue dots) and 2019 (brown dots) for all the studied European countries). The 95 % CIs for the central values represented here can be found in SI Data 6.

compared to 10 % and 7 % in Central and Western Europe respectively. In Ukraine, Belarus, Russia, and Norway the fire mortality fractions under the 10T assumption were estimated to be 285 (191–423), 245 (153–393), 230 (158–336) and 225 (151–332) respectively in 2019 (SI Data7). Nonetheless, we acknowledge the uncertainties related to our assumption of enhanced toxicity for fire $PM_{2.5}$, which highlights the importance of gaining a better understanding of the mechanisms and magnitude of the associated health risks from fires over chronic exposure.

In addition, we also estimated the relative contributions of the major transitional factors (i.e. BM, population size, population age and exposure) to excess death burden changes from 1990 to a consequent year, which is critical for understanding the effectiveness and progress of air quality mitigation policies. From an epidemiological perspective, as health care improves over time (which encompasses factors like nutritional status and medical care), the likelihood of mortality from a disease is anticipated to decline. Simultaneously, in Europe, there is a demographic shift towards older age groups, which increases the vulnerability to dying from non-communicable diseases associated with $PM_{2.5}$ and fire $PM_{2.5}$ exposure. We find that ageing populations and increases in population size were associated with 53 % and 57 % increase in excess deaths in Europe from 1990 to 2019 (Fig. 7). This was counteracted by improvements in baseline mortality rates and reductions in $PM_{2.5}$ exposure resulting in 41 % and 54 % decrease in excess death burden in the same period. The impact of an ageing population is more significant in Central Europe (resulting in 68 % increase in excess deaths in 2019 from 1990) as compared to in Eastern (42 %) and Western (52 %) Europe, whereas the impacts of improvement in baseline mortality rates were less significant in Eastern Europe (resulting in 17 % decrease in excess death burden in 2019 from 1990) compared to 53 % and 43 % decrease in Western and Central Europe (Fig. S10). Effective air pollution mitigation efforts across Europe resulted in significant decrease in excess deaths associated with $PM_{2.5}$ by 50 %, 48 % and 44 % in Western, Eastern and Central Europe respectively.

4. Uncertainties and limitations

In this study, the 95 % confidence intervals (CIs) are derived by combining the uncertainties in baseline mortality rates from the GBD data and those related to the MR-BRT splines used to calculate relative risk, as described in a previous study. While we acknowledge that the confidence intervals do not encompass the additional uncertainties related to the modelling of $PM_{2.5}$, and uncertainties in model inputs (e.g. anthropogenic emission inventories and model schemes) we observe that the comparison with measurements generally yields robust results (SI Text, Figs. S1, S2). We perform model simulations at $\sim$20 $\times$ 20 km spatial resolution, which though sufficient to capture the urban-rural gradient in $PM_{2.5}$, may average out the fine-scale urban and curb-side increments $PM_{2.5}$ at measurement sites. Nonetheless, a prior study found that the resolution of $PM_{2.5}$ calculations, ranging from 10 km to 100 km, has minimal impact on the $PM_{2.5}$ exposure estimations (Kushta et al., 2018). Instead, the uncertainties in excess mortality estimates are primarily influenced by the parameters of the exposure response functions. Although we recognize that there are limited chronic impact studies linking fire $PM_{2.5}$ exposure to health effects (Grant and Runkle, 2022), we employ the GBD exposure response function to evaluate the role of fires in relation to contributions from other sources.

We find that the rate of change in PM2.5 associated with wildfire to increase significantly over the time period (1990–2019) at 0.0008 μg/m3/year (significant at $p < 0.05$). Additionally, the rate of change in fire $PM_{2.5}$ and $PM_{2.5}$ are significant over large parts of Europe (at $p < 0.05$, please see Fig. S11a, b). However, when estimating excess deaths, it is crucial to consider fluctuations in baseline disease rates, which, despite decreasing across most European countries, are prone to statistical and methodological uncertainties stemming from variations in health registry data and documentation (Pozzer et al., 2023).

Furthermore, we acknowledge uncertainties related to the formulation of Exposure Response Functions (ERFs). These uncertainties are statistical and involve variations arising from (i) the number of cohort studies used and the assumed functional shape, and (ii) the exclusive reliance on cohort studies from specific regions, specifically the U.S., Europe, and China. Additionally, the ERFs inherently encompass uncertainties in the low concentration threshold, below which no adverse effects from pollution are anticipated, as these thresholds are derived mathematically once the ERFs are estimated (Pozzer et al., 2023). All these factors combined, results in a large spread in Fig. 3, although the rate of changes in population weighted fire $PM_{2.5}$ exposure and $PM_{2.5}$

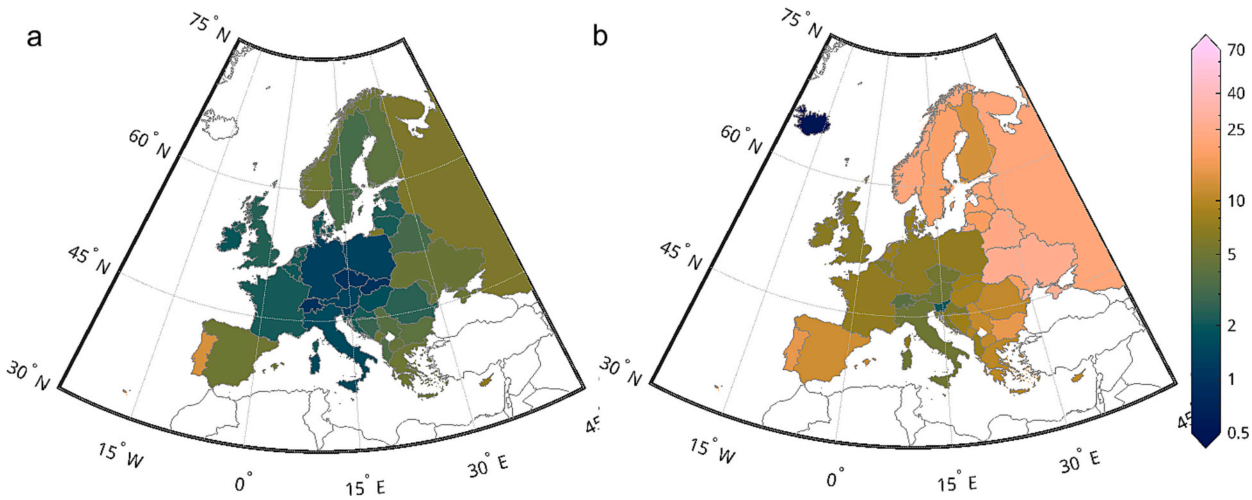

Fig. 5. Spatial distribution of fire mortality fractions in 1990 (a) and 2019 (b).

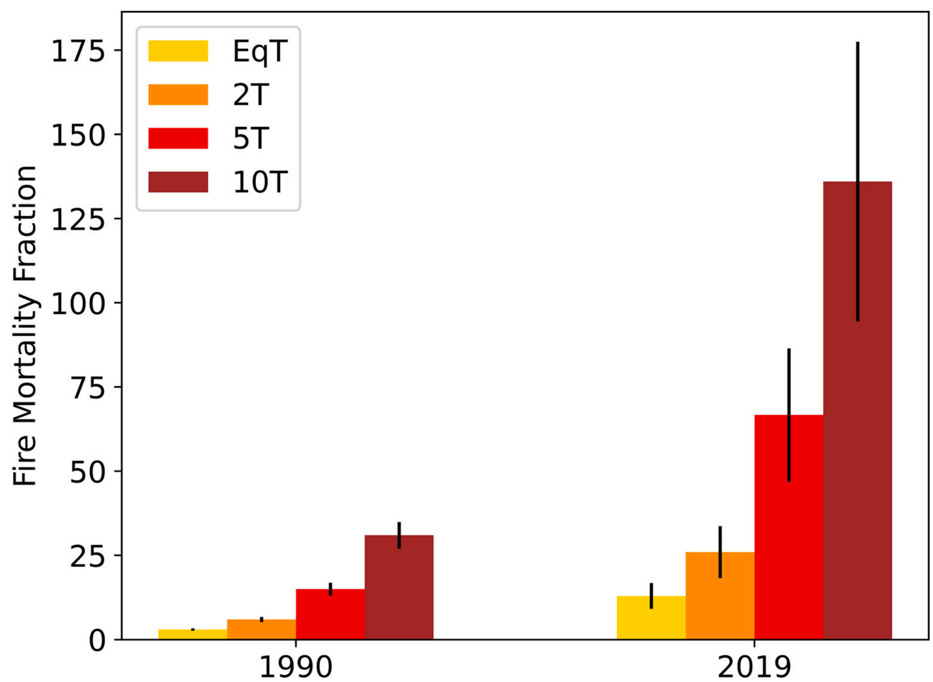

Fig. 6. Fire mortality fractions while considering forest fire $PM_{2.5}$ to be n ($n = 2, 5, 10$) times toxic as compared to other $PM_{2.5}$ sources. The equal toxicity (EqT) scenario is depicted in yellow bars.

exposure are statistically significant.

In the present study, we addressed the chronic exposure to emissions from fires and assessed their impacts on human health. While chronic exposure to is more important and is strongly associated with mortality. It is crucial, however, to recognize that acute exposure to emissions generated by wildfires holds immediate significance. These occurrences may lead to abrupt and substantial spikes in air pollutant concentrations within a brief timeframe. Such acute exposure scenarios have the potential to induce a rapid onset of respiratory diseases, exacerbate pre-existing respiratory and cardiovascular conditions, prompt emergency room visits, and contribute to elevated cardiovascular and respiratory morbidity. Notably, the current assessment did not perform health impact assessment for cardiovascular and respiratory morbidity resulting from acute exposure, warranting further exploration in future research.

While it should be noted that various emission inventories in general have similarly large uncertainties for organic aerosols, we acknowledge that the use of different emission inventories for anthropogenic activities will influence somewhat the calculated $PM_{2.5}$ exposures. However, arguably the primary uncertainty in the emission data originates from fire emission modelling. Due to the sporadic character of fires, nearly the only comprehensive way to monitor them is to use satellites, either recording the burnt area scars, or evaluating the power of active fires. Both have strong and weak points, but both share the ultimate dependency on the existence and capabilities of satellites. Any change of the satellite fleet can lead to a factor of times jumps in the fire inventory and, consequently, emission (e.g., adding the second MODIS instrument in 2002 tripled the registered fire radiative power – see http://is4fires.fmi.fi). It's important to mention that while we incorporate fires stemming from various land-use-related fuels such as grass, agricultural waste, tropical forest, temperate forest, boreal forest, shrub, and tundra, we do not distinguish emissions from distinct fire types in this study.

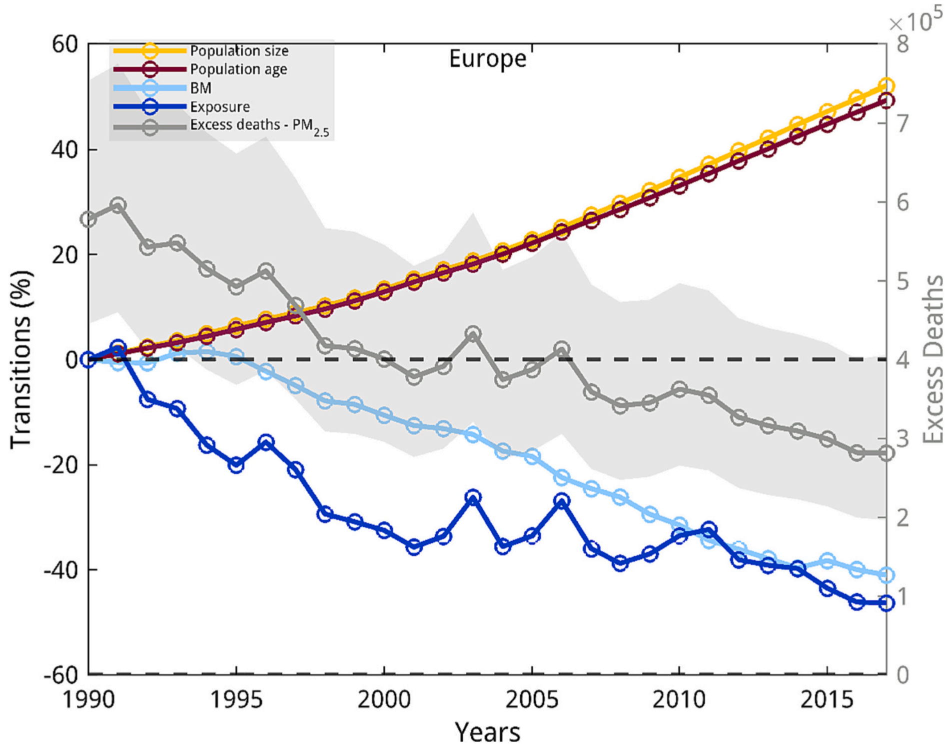

Fig. 7. Relative contribution of the four transition factors (i.e. Population size, Population age, baseline mortality rates (BM) and exposure) to $PM_{2.5}$ related excess deaths in Europe between 1990 and a consequent year.

Minimizing the uncertainty related to changes in the fire observations throughout the considered period, we used the machine-learning Fire Forecasting Model, which was trained and tested over the best-available homogeneous time series of fires provided by two MODIS instruments onboard Aqua and Terra satellites. They provided 20 years of well-calibrated time series, 2003–2022. This period was sufficient to establish the correlations between the weather conditions, fire danger indices, and the registered fires. However, the nature of the time-agnostic algorithm does not include sociology-driven trends: the same weather situation (including history of the season) would lead to the same prediction of fires and fire emission regardless of the specific year. The model therefore showed a slight upward trend in fire intensity following the growing weather-driven fire danger. However, as IS4FIRES data show, forest management and fire prevention measures in many European regions resulted in the actual trend to be closer to neutral or, sometimes, even declining (Daalen et al., 2022). This does not change the conclusions of the current study (the difference in predicted and observed fire intensity trends is quite small) but the absolute trend of the fire contribution suggested above should be considered rather as an upper-level estimate.

We conduct sensitivity analyses by assuming that emissions from fires are more toxic than those from other sources, given the substantial release of black carbon and organic aerosols associated with higher oxidative stress and increased toxicity, as indicated by recent toxicological assessments (Bates et al., 2019; Park et al., 2018). However, it is important to acknowledge that, studies find mixed effects for emissions from fires, especially on cardiovascular outcomes (Black et al., 2017) and the epidemiological studies informing our assessments focus on acute exposures and respiratory hospitalizations (Aguilera et al., 2021). We also note that while recent research addresses morbidity and mortality risks linked to long-term exposure to black carbon (Janssen et al., 2011; Yang et al., 2021), discrepancies exist in reported increased risks from other $PM_{2.5}$ components emitted from wildfires. Despite these uncertainties, we contend that our assumptions for the sensitivity study, pending further epidemiological analyses for chronic exposure, highlight potential influences on policy decisions for improved wildfire management.

5. Conclusions

Using a long-term (1990–2019) high resolution chemical transport model simulation we find that the population weighted ambient $PM_{2.5}$ exposure decreased by >50 % over Europe from 1990 to 2019, with substantial improvements in all the three regions (i.e. Western, Central and Eastern Europe). The reductions in air pollution primarily stem from multiple policies and directives which resulted in advancements in combustion processes in both industrial processes and residential heating, improved fuels for transportation, reduced coal use for power generation with minor improvements in contributions from agricultural sources- which are the leading cause of anthropogenic $PM_{2.5}$ in Europe (Annesi-Maesano, 2017; Kuklinska et al., 2015). However, despite such improvements, exposure to ambient $PM_{2.5}$ remains the largest environmental health risk in Europe. Applying the latest MR-BRT exposure response function, our estimation indicates that the burden of excess deaths from exposure to ambient $PM_{2.5}$ declined across Europe, decreasing from 0.57 million (0.44–0.75 million) in 1990 to 0.28 million (0.19–0.42 million) between 1990 and 2019. Our estimates of excess deaths for Western Europe and Central Europe are comparable to the most recent GBD estimates. However, the estimates are notably three times lower for Eastern Europe, which might be attributed to the inclusion of only the European part of Russia in our study area.

Our findings concur with previous studies (Chowdhury et al., 2022; McDuffie et al., 2021) that find fires to be a significantly smaller source of ambient $PM_{2.5}$ related excess deaths in Europe as compared to anthropogenic sources like transportation, industries, agriculture, and power generation. For the first time, our study yields results indicating a rising relative significance of fire contributions to the excess death burden between 1990 and 2019. We found 13 (9–18) out of 1000 excess deaths from ambient $PM_{2.5}$ exposure to be associated with fire $PM_{2.5}$ in 2019, a 6-fold increase from the 1990 levels. We found that the fire mortality fractions in Eastern Europe were at least three times higher as compared to Western and Central Europe. Ukraine, Belarus, Russia and Norway were found to have the largest increase in fire mortality fractions from 1990 to 2019. Noteworthily, climate change accompanied by rising temperatures, intense droughts, and prolonged dry seasons, is responsible for the growing frequency of wildfires in Europe and climate change is also anticipated to be the primary driver of wildfires in the coming decades (Im et al., 2022). While our study does not explore the impact of climate change on the meteorological factors of importance for modelling fire emissions and related excess deaths, we recognize that a substantial portion of the observed changes in fire $PM_{2.5}$ from 1990 to 2019 is likely linked to the changing climate.

As sensitivity analysis, we assign higher toxicity to fire $PM_{2.5}$ as compared to the other anthropogenic and natural sources of $PM_{2.5}$ based on toxicological and epidemiological studies of short-term health outcomes (Aguilera et al., 2021; Chen et al., 2021), pending extensive investigation of long-term impacts. Under the 10T assumption, fires would account for >13 % of excess deaths from ambient $PM_{2.5}$ exposure in 2019, and more than a quarter of excess deaths in Eastern Europe could be attributed to fires. Additionally, our findings indicate that the rapid increase in the ageing population across Europe is elevating the number of vulnerable individuals at risk of mortality from exposure to ambient $PM_{2.5}$ and fire $PM_{2.5}$.

Such increasing contribution from fires to ambient $PM_{2.5}$ exposure and related health burden are counteracting policies and efforts to achieve clean air across Europe (Annesi-Maesano, 2017). Multiple studies find that extreme fire weather has intensified globally as well as over Europe (Jain et al., 2022; Jolly et al., 2015; Lund et al., 2023). Acknowledging the growing threat of fires, especially intensified by climate change, the European Union (EU) has emphasized the importance of improved cooperation among member states to effectively manage and combat wildfires. The EU has proactively addressed the issue of wildfires by enacting various initiatives and strategies focused on prevention, preparedness, and response (European Commission 2021; Gamboa et al., 2023, https://effis.jrc.ec.europa.eu/reports-and-publications/annual-fire-reports). These efforts aim to minimize the adverse impacts of wildfires on human health and the ecosystem. The effectiveness of these fire management policies is evident in the reduced pace of fire $PM_{2.5}$ increase across all European regions from 2010 onwards (see Fig. 1), coinciding with the implementation of most emergency management services related to fire management. Nevertheless, in recent years, Europe has experienced significant occurrences of large wildfires and should emissions indeed prove to be more toxic than $PM_{2.5}$ emissions from other sources on chronic exposure, as indicated in our sensitivity analyses, larger coordinated efforts on more sustainable forest management are required to avert the large associated health burden.

CRediT authorship contribution statement

Sourangsu Chowdhury: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Visualization, Writing – original draft, Writing – review & editing. **Risto Hänninen:** Data curation, Formal analysis, Methodology, Software, Writing – review & editing. **Mikhail Sofiev:** Data curation, Formal analysis, Funding acquisition, Writing – review & editing. **Kristin Aunan:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

The authors acknowledge the Belmont Forum Climate, Environment and Health-I project, HEATCOST (Academy of Finland grant no. 334798 and Research Council Norway grant no. 310672), the European Union's Horizon 2020 research and innovation program under Grant Agreement 820655 (EXHAUSTION) and the European Union's Horizon 2020 cooperation and support action program under Grant Agreement 101003966 (ENBEL).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.171314.

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