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Long-term health impact assessment of total $PM_{2.5}$ in Europe during the 1990–2015 period

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HIGHLIGHTS

- $PM_{2.5}$ concentrations over Europe were used to perform a health impact assessment during the 1990–2015 period.
- Population weighted $PM_{2.5}$ concentrations were predicted to have declined by 0.8 $\mu g\ m^{-3}\ y^{-1}$ on average.
- The number of premature deaths due to exposure to $PM_{2.5}$ was predicted to have declined during the 1990–2015 period.
- The variability in the predicted number of premature deaths was higher in the 1990s compared to the 2000s.

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ABSTRACT

Several datasets of $PM_{2.5}$ concentrations over Europe during the 1990–2015 period, were used to calculate health impacts from chronic exposure to total particle matter below 2.5 µm (i.e. $PM_{2.5}$). The datasets used in the analysis include the European Topic Centre on Air Pollution and Climate Change Mitigation (ETC/ACM), the Copernicus Atmospheric Monitoring Service (CAMS), the Global Burden of Disease (GBD), the World Health Organization (WHO) as well as the EURODELTA-Trends (EDT) multi-model reanalysis developed specifically for Europe.

The exposure to ambient $PM_{2.5}$ concentrations was calculated as population weighted annual average $PM_{2.5}$ concentrations by country. The calculated exposure to $PM_{2.5}$ was later used as input in the health impact assessment (HIA) Alpha-RiskPoll (ARP) tool to retrieve the total number of premature deaths.

Our results indicate a substantial reduction in the number of premature deaths from $PM_{2.5}$ exposure in Europe over the 1990–2010 period, between nearly 30 and 50%. Putting all the data-sets together, even if they do not cover the whole period, a decrease of even around 60% is observed between 1990 and 2015. For the countries included in this study, the estimated number of premature deaths from $PM_{2.5}$ in 1990 was found to be around 960 000 (median of all the available datasets), whereas in 2015 it was found to be around 445 000. However, the variability in the estimated premature deaths from the different $PM_{2.5}$ datasets was found to be large during the

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early 90s (around a factor of 2). For the latest years of the investigated period (2005 onwards), where a relatively flat trend in the $PM_{2.5}$ exposure was observed, the differences between the different datasets were smaller.

Even though our results indicate a reduction in the number of premature deaths from chronic exposure to $PM_{2.5,}$ the numbers remain considerable in 2015, underlining the need to continue improving air quality in the future.

1. Introduction

Particulate matter (PM) induces negative effects on human health (Cohen et al., 2017; Crippa et al., 2019; Im et al., 2018; Pope and Dockery, 2006; WHO, 2013a). Particles with an aerodynamic diameter below 2.5 μm (i.e. $PM_{2.5}$), mainly arise from various anthropogenic sources (i.e. combustion processes) as well as from the conversion of high volatile biogenic organic compounds (BVOCs) in the particle phase after oxidation with various atmospheric oxidants (e.g. OH, O_3 and NO_3). Especially in this size range and in the sub-micron range (i.e. PM_1), the capacity of particles to penetrate deeper in the various sections of the lung and in the alveoli regions, is highly enhanced, which poses serious threats for human health. Numerous epidemiological studies have indicated a significant dependence between exposure to particle matter concentrations and number of deaths related to respiratory diseases (Atkinson et al., 2015). In Europe, short-term epidemiological studies, e.g. the APHEA-2 (Air pollution and Health: a European Approach) provided first information on daily mortality increase related to PM_{10}. The study included about 43 million individuals in 29 cities in Europe which were followed for more than 5 years in the beginning of the 90s. Results indicated an increase of about 0.6% in the all-cause daily mortality for a 10 μg m^{-3} increase in PM_{10} concentrations. The numbers of hospital admissions were also reported using information from 38 million people in 8 European cities, followed over 3–9 years during the first half of the 1990s. Asthma and chronic obstructive pulmonary disease (COPD) were found to increase by 1% for each 10 μg m^{-3} increase in PM_{10} concentrations, whereas hospital admissions for cardiovascular diseases (CVD) were found to increase by 0.5% for each 10 μg m^{-3} in PM_{10} concentrations, among people older than 65 years (Atkinson et al., 2001).

One of the main complications in investigating the health impact of fine particles is represented by their complex nature (Seinfeld and Pandis, 2012). The extent of the cumulative health effect of $PM_{2.5}$ (Xing et al., 2016) depend on the type of sources, the chemical composition as well as the physical properties of PM. Previous studies showed that vehicle traffic can significantly increase exposure and health risks, depending on the type of road and other factors such as meteorology and vehicle mix (Zhang and Batterman, 2013). Indeed, several epidemiological studies take the distance from major roads as a proxy for PM exposure (Volk et al., 2011). More recent epidemiological studies indicated that wildfire smoke is significantly associated with risk of respiratory morbidity (Liu et al., 2015). In a more recent study of Liu et al. (2017), an increase of 7.2% in the risk of respiratory hospital admissions was found during days with intense contribution of wildfire to $PM_{2.5}$ compared to days with no wildfire smoke activities. Moreover, recent laboratory experiments performed using state-of-the-art smog chamber facilities, indicated that cells deaths may occur when they are exposed to particles from wood combustion, along with DNA damage (Krapf et al., 2017).

Various studies have used Chemical Transport Model (CTM) output for conducting health impact assessments. Brandt et al. (2013) developed an integrated model system, EVA (Economic Valuation of Air pollution) to investigate the health-related economic cost for specific emission sources. Their study indicates that the major contributors to health-related cost associated with air pollution arise from power production, agriculture, road traffic, and nonindustrial domestic combustion, including wood combustions (Brandt et al., 2013). More recently, Solazzo et al. (2018) used simulated $PM_{2.5}$ and ozone concentration fields from 12 chemical transport models in the framework of the third phase of the Air Quality Model Evaluation International Initiative (AQMEII (Galmarini et al., 2017)) to calculate the impact of air pollution on premature deaths and crop yields over Europe for the year 2010. Their analysis showed a mean number of premature deaths due to exposure to $PM_{2.5}$ and ozone of approximately 370 000 (Solazzo et al., 2018). Im et al. (2018) investigated the health effect of total $PM_{2.5}$ for the year 2010 in the framework of the third phase of the AQMEII3 model inter-comparison exercise. The Economic Valuation of Air Pollution system was used to calculate the health impact of different pollutants (e.g. O_3 and $PM_{2.5}$) with exposure-response coefficients from various sources. The ensemble of all the participating models indicated around 390 000 ± 100 000 deaths for Europe (30°W-60°E, 25–70°N) in the year 2010 due to exposure to $PM_{2.5}$. Moreover, their study suggested that a total number of 47 000 premature deaths could be avoided with a 20% reduction of anthropogenic emissions in Europe. However, model results differed by up to a factor of 3 for the European results (12 models participated in the exercise). Cohen et al. (2017) investigated the global burden of disease (GBD) attributable to air pollution during the 1990–2015 periods. The analysis was conducted at different scales, i.e. global, regional and country levels. Resolved aerosol optical depth (AOD) fields retrieved from satellites were combined with chemical transport models, surface PM observations and geographical information to estimate the population exposure to $PM_{2.5}$ at a 0.1 × 0.1° resolution in 5 years steps. Different integrated dose-response functions for various causes of deaths were implemented based on risk estimates from different studies. Their results indicated that in 2015, about 4.2 million deaths were caused by long-term exposure to $PM_{2.5}$ which correspond to about 7.6% of the total global mortality risk due to this factor. Moreover, a decrease in the global rates of mortality between 1990 and 2015 was estimated because of the improved air quality conditions, but an increase in the absolute number of attributable deaths was predicted mainly because of the increased air pollution especially in China and India. In addition, burning of solid fuel was found to be the major cause of mortality in low-income and middle-income countries. Similarly, Lelieveld et al. (2015), used a global atmospheric chemistry model to investigate the contribution of various emissions sources to premature mortality for the year 2010. The study indicated that outdoor pollution (mainly $PM_{2.5}$), led to about 3.3 million premature deaths in the world, depending on the assumptions about particle toxicity, therefore making a substantial contribution to the health impact (Lelieveld et al., 2015).

Anthropogenic emissions have largely declined in Europe over the recent decades thanks to the switch to low-sulfur fuels (e.g. natural gas), the introduction of desulphurization as well as the new EURO standard for passenger cars. Tørseth et al. (2012) investigated trends of PM_{10} and $PM_{2.5}$ at 24 and 13 EMEP sites, respectively. Their study indicated average reductions of 18% and 27% of PM_{10} and $PM_{2.5}$ concentrations over the 2000–2009 period.

While previous studies have investigated long term health impact assessment of total $PM_{2.5}$ at global scale (Brauer et al., 2016; Cohen et al., 2017), in this study we provide novel information on the total number of premature deaths due to exposure to $PM_{2.5}$ with a specific focus on the European domain. We used available $PM_{2.5}$ estimates for Europe from the European Topic Centre on Air Pollution and Climate Change Mitigation (ETC/ACM) as well as the regional component of the Copernicus Atmospheric Monitoring Service (CAMS) which mainly covered specific years in the 2000s (Marécal et al., 2015). In addition, the novel multi-model EURODELTA-Trends (EDT) exercise, which built upon the establishment of the Convention on Long Range Transboundary Air Pollution (CLRTAP) in 1979, provided 21 years of continuous $PM_{2.5}$ concentrations over Europe from the year 1990, and with "real" year-to-year meteorological input data (Colette et al., 2017a). The EDT dataset was used to calculate the exposure to $PM_{2.5}$ as well as the number of premature deaths associated with it. Its estimates were compared with $PM_{2.5}$ exposure and premature deaths retrieved from the other datasets (e.g. including GBD).

The paper is organized as follows: In Section 2 information regarding the methods and data used for the study are reported. In Section 2.1 an overview of the different datasets of $PM_{2.5}$ concentrations available for Europe during the 1990–2015 period is presented. Section 2.2 describes the population data and natural deaths data used in the health impact assessment tool. Section 2.3 describes the methodology used for the health impact assessment. Results and discussions are presented in section 3 and section 4, respectively.

2. Methods

2.1. Overview of the different $PM_{2.5}$ datasets

Four different datasets of $PM_{2.5}$ concentration fields available for Europe were used in this study to quantify the health impacts of total $PM_{2.5}$:

- The European Topic Centre on Air Pollution and Climate Change Mitigation (ETC/ACM) provided $PM_{2.5}$ concentrations using the regression – interpolation – merging mapping methodology (Horálek et al., 2016), i.e. a kriging method, to interpolate the regression residuals of in-situ surface PM measurements from the Air Quality e-reporting database with the concentrations calculated by the EMEP MSC-W model (at 50 km resolution) and other supplementary data. Due to the limited availability of $PM_{2.5}$ measurements in Europe, PM_{10} observations at urban, sub-urban and rural areas were also used to derive $PM_{2.5}$ concentrations using a multi-parameter linear model (Denby et al., 2011). ETC/ACM $PM_{2.5}$ concentrations estimates were made available for the years 2005, 2007, 2008 and 2010 through to 2015. The years 2006 and 2009 were not included in this analysis because ETC/ACM data were not available for those years. In addition, from the year 2013 onwards, a different population data source, i.e. Geostat 2011, was used for the mapping, instead of the JRC population data. Therefore, the ETC/ACM data source was adjusted using the Geostat population estimates to make the data consistent for all the available years.
- The EURODELTA-Trends (EDT) project provided modeled air quality trends for Europe for the 1990–2010 period (Colette et al., 2017a). Five state-of-the-art chemical transport models, i.e. CHIMERE, EMEP MSC-W, LOTOS-EUROS, MATCH and MINNI, performed simulations of air pollution in Europe for the 21-years period using common anthropogenic emission inventories from IIASA's GAINS model (Klimont et al., 2017). Model simulations were performed at $0.25^{\circ} \times 0.4^{\circ}$ latitude and longitude resolution, which corresponds to about 25 km in the European domain. For this study, the outputs of the different models were corrected using data fusion techniques, i.e. kriging, by means of daily $PM_{2.5}$ and PM_{10} measurements available at urban, suburban and rural sites from the Air Quality e-reporting database. Kriging consist of a linear weighted combination of the measured $PM_{2.5}$ and PM_{10} values that deploys spatial autocorrelation among the data to retrieve the weights and uses the ensemble of EDT models as the external drift in the kriging. Under suitable assumptions on the priors, such techniques provides the optimal linear unbiased prediction of the intermediate values (Cressie, 1990). Only three models, CHIMERE (Mailler et al., 2017; Menut et al., 2013), EMEP MSC-W (Simpson et al., 2012) and LOTOS-EUROS (Manders et al., 2017; Sauter et al., 2012; Schaap et al., 2008) were used to perform the data fusion since output from the remaining models were not available at the time the kriging was performed. In addition, since no PM observations are available prior to 2000 and $PM_{2.5}$ measurements started even later, the fusion of EDT models output with observations was done for the 2000–2010 and 2008–2010 periods, for PM_{10} and $PM_{2.5}$, respectively. The requirement for the data fusion procedure was that the original modeled relative trend is conserved, though the absolute values are adjusted. Therefore, to perform data fusion for the entire periods, the ensemble median of raw modeled $PM_{2.5}$ fields (available for 1990–2010) was scaled using the ratio between the ensemble median of raw modeled $PM_{2.5}$ and the kriging of the model ensemble with $PM_{2.5}$ measurements during the 2008–2010 period. The retrieved ratio was then applied to every model participating in the EDT ensemble. Those corrections are spatially dependent and applied on the model grid as they are built with respect to the kriged analysis of observations.
- The regional component Copernicus Atmospheric Monitoring Service (CAMS) provided yearly $PM_{2.5}$ concentration fields calculated with several CTMs: CHIMERE (CHIa), EMEP MSC-W (EMPa), EURAD (RIUa), LOTOS-EUROS (KNMa), MATCH (SMHa), MOCAGE (MFMa) and SILAM (FMIa) including either data assimilation or data fusion techniques. Available PM measurements from the Air quality e-reporting database are used to improve the model outputs, with different techniques for the participating models. These are either Ensemble Kalman Filter, 3DVar, or optimal interpolation relying on kriging with an external drift (Marécal et al., 2015). Only two-third of the available PM measurements were used in the data assimilation or data fusion process to improve model performance (as the remaining data are used for evaluation purposes), making the CAMS products quite different from the other datasets presented here. The list of the models available in the CAMS products included in the regional CAMS-VRA (Validated analysis, i.e. using validated in-situ measurement) for the year 2014 are: CHIMERE, EMEP MSC-W, EURAD, LOTOS-EUROS, MATCH and SILAM. For the year 2015 the CHIMERE, EMEP MSC-W, EURAD, LOTOS-EUROS, MATCH, MOCAGE and SILAM data were used.
- The Global Burden of Disease (GBD) study provides global concentration fields of $PM_{2.5}$ at $0.1^{\circ} \times 0.1^{\circ}$ of longitude and latitude resolution (Brauer et al., 2016; Shaddick et al., 2018). The GBD13 dataset adopts similar procedures as used in the GBD10 dataset (van Donkelaar et al., 2010) to retrieve the $PM_{2.5}$ concentrations including new satellite-based estimates of $PM_{2.5}$ for the 1998–2012 period. The near surface $PM_{2.5}$ concentrations are retrieved by applying the relationship of $PM_{2.5}$ to AOD simulated by the GEOS-Chem model. The instruments used for the $PM_{2.5}$ estimates include the MODIS, MISR and SeaWiFS satellites as well as vertical profile information from the CALIOP satellite instrument. These sets of estimates were used to estimate the $PM_{2.5}$ concentrations for 2000, 2005, 2010 and 2011. For the year 1995 and 1990, the ratio between the GEOS-Chem model simulations for the year 2005 and the year of interest was used (with anthropogenic emission from the EDGAR dataset). In addition, simulations from the TM5-FASST model (Fast Scenario Screening Tool) were also included for the year 1990, 2000 and 2010 using emission estimates from the ECLIPSE v4a inventory. $PM_{2.5}$ measurements were collected for the 2010–2013 period from the same sources as in the GBD10 dataset and estimated from the $PM_{2.5}$ to PM_{10} ratio when not available (and excluding industrial and roadside sites when possible). A regression calibration approach, using the mean of satellite-based data (interpolated with the GEOS-Chem model), TM5-FASST simulations and available measurements, was used to correct the final global estimate of $PM_{2.5}$ concentrations at $0.1^{\circ} \times 0.1^{\circ}$ grid-cell resolution using a single global calibration function. Finally, The GDB15 dataset further expanded the methods and datasets used in GBD13 with additional air quality measurements and novel estimation

algorithms (Shaddick et al., 2018). In particular, the TM5 simulations were not included in the re-analysis, and the calibration functions can vary spatially using a Bayesian Hierarchical Model (Shaddick et al., 2018). The estimated global population weighted mean concentrations of $PM_{2.5}$ were made available at 11 km × 11 km resolution.

2.2. Population data

In order to retrieve the population weighted exposure to $PM_{2.5}$ concentrations, supplemented Geostat 2011 population density data provided by the ETC/ACM were used (Horálek et al., 2016). For specific countries for which population density was not available in the Geostat 2011 dataset, the data was complemented with other sources of population density: the JRC (Joint Research Centre) population data at a resolution of 100 m × 100 m (Gallego, 2010) and the population density data from the ORNL LandScan Global Population dataset. The latter provided population density estimates at 30 × 30 arcsecs resolution and it is based on population data for the year 2008 provided by the Geographic Studies Branch, US Bureau of Census. The ORNL data was re-projected from its original WGS 1984 30 × 30 arcsecs grids into the Geostat 2011 reference projection, i.e. ETRS89-LAEA5210 at 1 × 1 km resolution by EEA.

2.3. Health impact assessment: methodology

The quantification of health impacts was carried out with INERIS' version of the health impact assessment (HIA) tool Alpha-RiskPoll (ARP; developed by EMRC, and described in Schucht et al. (2015). ARP is regularly used in European Policy analyses such as the CAFE (Clean Air For Europe) program. The methods for benefits assessment used in ARP were first developed under the EC funded ExternE project (External cost of Energy) suite during the 1990s. These methods are extensively documented in Holland et al. (2005a,b,c, 2011) and Hurley et al. (2005). They have been applied since the end of the 1990s to cost-benefit assessments of EC and UNECE policies and are regularly and thoroughly reviewed (Krupnick et al., 2005; WHO, 2013a, 2013b).

The current version of the model relies on the methodology update as recommended by the WHO/Europe HRAPIE (Health Risks of Air Pollution in Europe) project (WHO, 2013a, 2013b). Its implementation in ARP is described in Holland (2014a,b). HRAPIE recommendations applied in ARP are the Concentration-Response Functions (CRFs) linking levels of pollutant exposure to a set of specific health endpoints (amongst which mortality and different morbidity impacts). The health endpoint "mortality due to chronic exposure to $PM_{2.5}$" is calculated for the age-group above 30 years based on the recommended Relative Risk (RR) of 1.062 for a 10 μg m^{-3} increase of $PM_{2.5}$ (95% confidence interval is 1.040–1.083) (ETC/ACM, 2017). Mortality effects are calculated for all-cause (natural) mortality, as linear functions and in

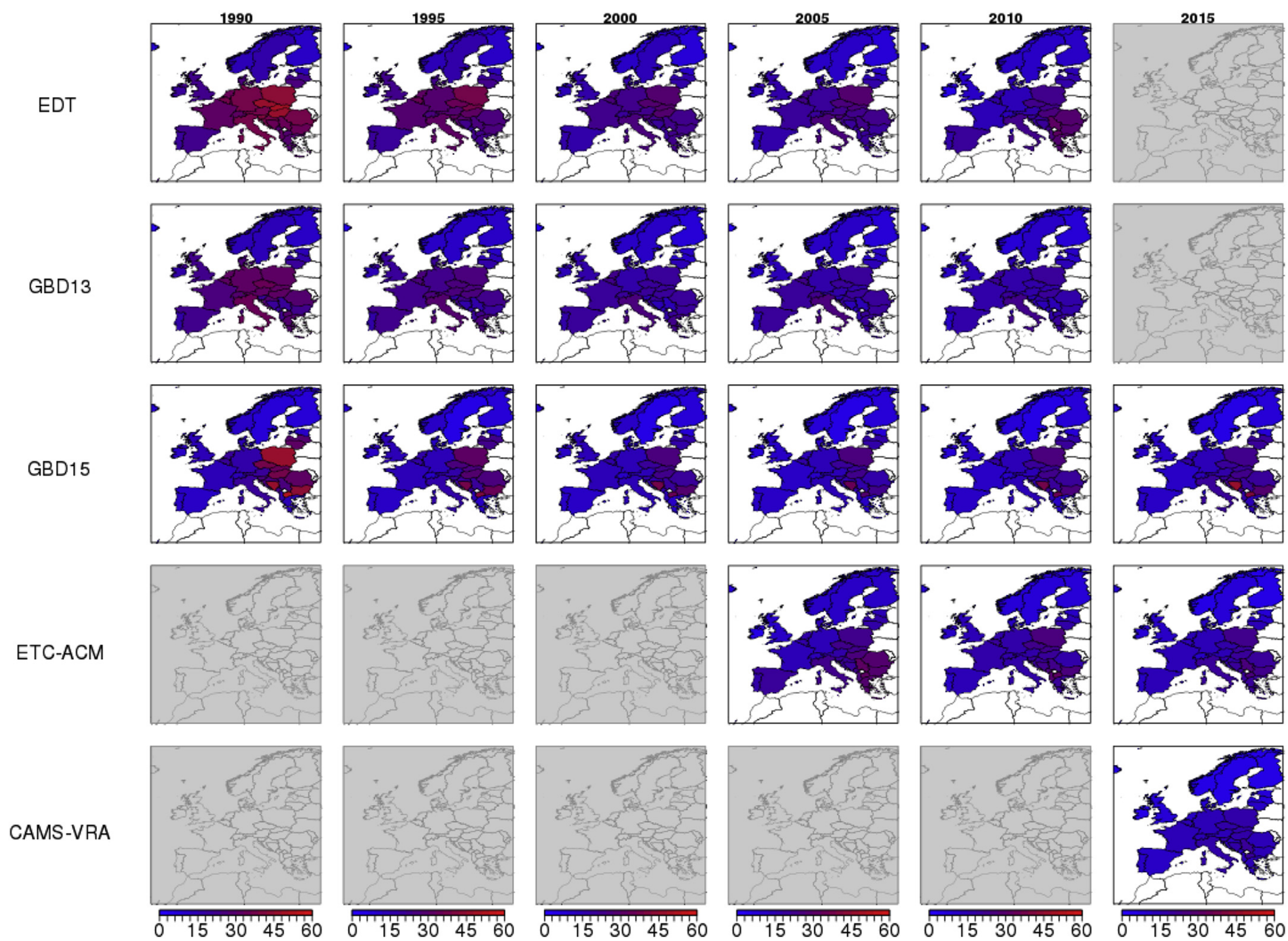

Fig. 1. Annual national average population weighted $PM_{2.5}$ concentration (μg m^{-3}), i.e. exposure to $PM_{2.5}$, as predicted by EDT (model ensemble), GBD13, GBD15, ETC-ACM and CAMS-VRA (model ensemble) datasets in 1990, 1995, 2000, 2005, 2010 and 2015. Grey maps indicate years for which the corresponding dataset is not available. Units are in μg m^{-3}.

response to a one-year pulse change without lag and without any threshold for $PM_{2.5}$ concentrations. Studies on long-term exposure give no evidence of a specific threshold, and epidemiological studies for the pollutants such as $PM_{2.5}$ have not provided direct evidence of the existence of such thresholds at the population level; but whether such threshold does apply for at least the pollutant considered in this study, is currently under debate (WHO, 2013a, 2013b). Also, various European studies of short-term exposure that have comprehensively investigated concentration–response functions have not revealed significant differences compared to the linear functions, and for ambient levels of $PM_{2.5}$ observed in Europe (WHO, 2013a, 2013b). Following current scientific advice, all particulate matter is treated as equally harmful, irrespective of source and chemical composition since a precise quantification of the health effects of individual PM components is not possible according to current knowledge (COMEAP, 2015; Miller et al., 2011; WHO, 2007, 2013b). In the present study, only one health endpoint is quantified: the mortality from chronic (long-term) exposure to $PM_{2.5}$, expressed in premature deaths per year. The present health outcomes are coherent with those described in de Leeuw and Horálek, 2016 and in the EEA's Air Quality in Europe reports (EEA, 2018). Indeed, these studies equally apply the HRAPIE methodology.

Total and age class specific population data relies on the UN's World Population Prospects, 2017 Revision. Information on mortality (all-age natural deaths, and 30 + years natural deaths) were extracted (and calculated) from the WHO Mortality Database (ICD-9 and ICD-10 classification, March and October 2017 updates, respectively). Age class information is not spatialized within countries: for a given country, the age distribution is assumed to be the same in all grid cells. For countries where population or mortality data were not available, gap filling was performed by using data from the latest available year. Furthermore, Andorra, Monaco and San Marino, are altogether excluded from the present study because they are not part of the AlphaRiskPoll country list. For Kosovo, the same exposure, baseline mortality and age distribution data as for Serbia are used. However, the total number of deaths attributed to $PM_{2.5}$ is split according to the relative population of Serbia and Kosovo under UNSCR 1244/99. For Lichtenstein, exposure estimates are available (for all data sets except for GBD13 and GBD15) but mortality and population data are limited. For this country we hence used the baseline mortality and age distribution of Austria, and mortality incidence data was scaled according to the population of Lichtenstein. The final list of countries taken from the single $PM_{2.5}$ concentration datasets as well as those used for the calculation of the total number of premature deaths is reported in Table S1.

3. Results and discussion

3.1. Exposure to $PM_{2.5}$

Fig. 1 shows the national annual average population weighted $PM_{2.5}$ concentrations, i.e. exposure to $PM_{2.5}$, as predicted by the different datasets for six years for which multiple datasets were available, i.e. 1990, 1995, 2000, 2005, 2010 and 2015. High variability in the exposure to $PM_{2.5}$ is visible especially during the early years of the period analyzed, i.e. the 90s, and especially between the EURODELTA-Trends (EDT) and the GBD15 datasets. In 1990, the models that participated in the EURODELTA-Trends exercise indicated an annual average population weighted $PM_{2.5}$ concentration between 29 and 37 $\mu g\,m^{-3}$, depending on model in the cluster (see section 2.3 for details on the available countries included in this analysis) whereas estimates were lower for the GBD13 and GBD15 datasets, i.e. around 27 and 20 $\mu g\,m^{-3}$, respectively (Table S2). The differences in predicted exposure for year of 1990, were in general higher in the central and eastern part of Europe especially for the EDT and GDB15 datasets (Fig. 1 and Table S2).

Fig. 2 shows the comparison in the predicted $PM_{2.5}$ exposure from all the datasets for the years 1990, 1995, 2000, 2005, 2010 and 2015. In all years, most points (i.e. population weighted means in specific

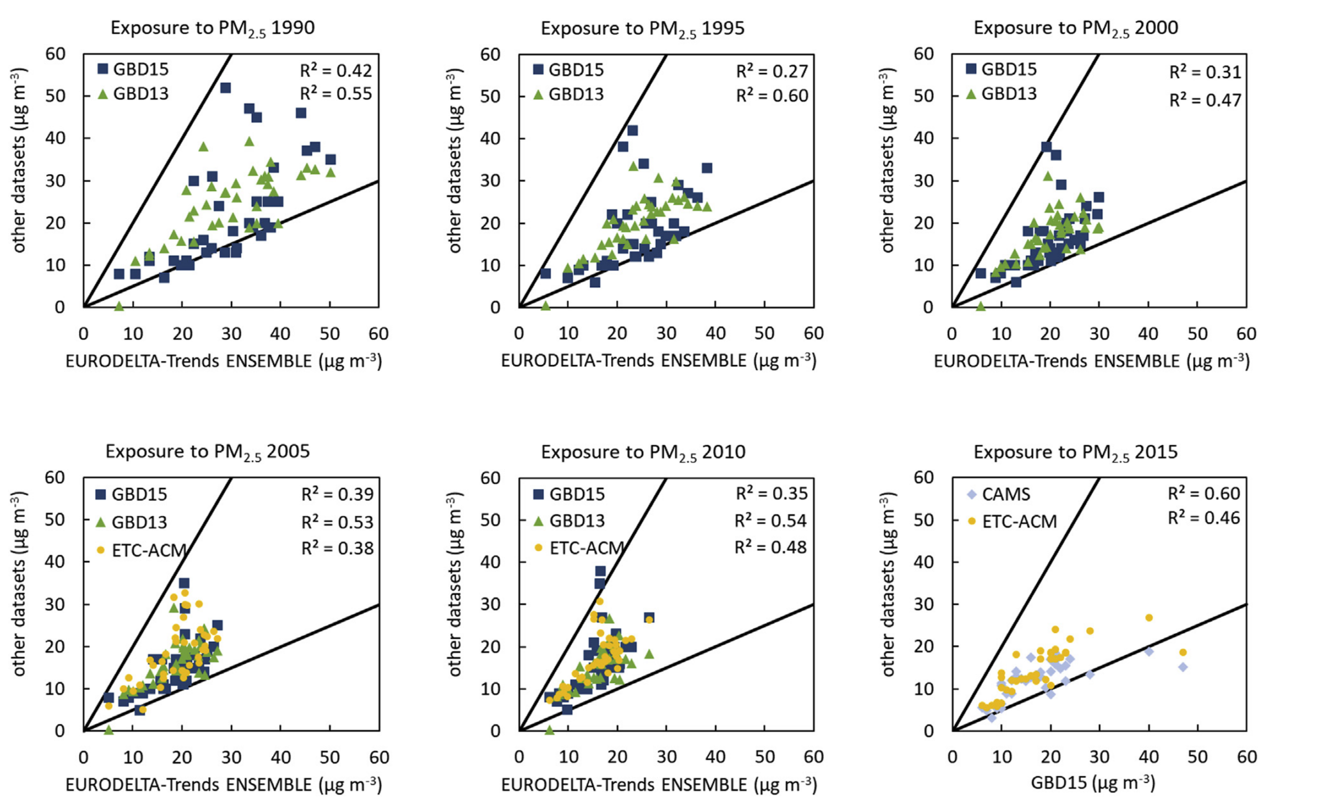

Fig. 2. Comparisons of the exposure to $PM_{2.5}$ ($\mu g\,m^{-3}$) between the different datasets (i.e. GBD13, GBD15, EDT, ETC-ACM and CAMS) for the 1990, 1995, 2000, 2005, 2010 and 2015 years. The black lines indicate the 1:2 and 2:1 line.

countries) were reproduced between the 1:2 line with larger variation in predictions in Macedonia and Bosnia-Herzegovina (i.e. points around 30–40 µg m^{-3} in the year 2010). In general, the EDT dataset correlated spatially (country by country) better with the GBD13 than the GBD15 datasets, with coefficients of determination (R^2) between 0.47 and 0.6 for GBD13 and between 0.27 and 0.42 for the GBD15 comparisons. In 2015, comparisons between the GBD15, CAMS and ETC-ACM revealed that most of the points were also reproduced within a factor of 2. Larger discrepancies were again found for Bosnia-Herzegovina (i.e. points around 40–50 µg m^{-3} in the year 2015).

Fig. 3 shows the variation (i.e. standard deviation) for all the countries where $PM_{2.5}$ exposure data were available for the years 1990, 1995, 2000, 2005, 2010 and 2015. Differences in the calculated exposure to $PM_{2.5}$ were between 5 and 10 µg m^{-3} in 1990 for most of the countries, whereas they were reduced in the more recent years, i.e. mostly well below 5 µg m^{-3} in 2010 and 2015 and with most of the datasets predicting similar values of exposure to $PM_{2.5}$ for the total of all the countries (Table S3 and Table S4). The latter was mainly attributed to the reduced uncertainty in the emission inventories used by the different chemical transport models and to the availability of $PM_{2.5}$

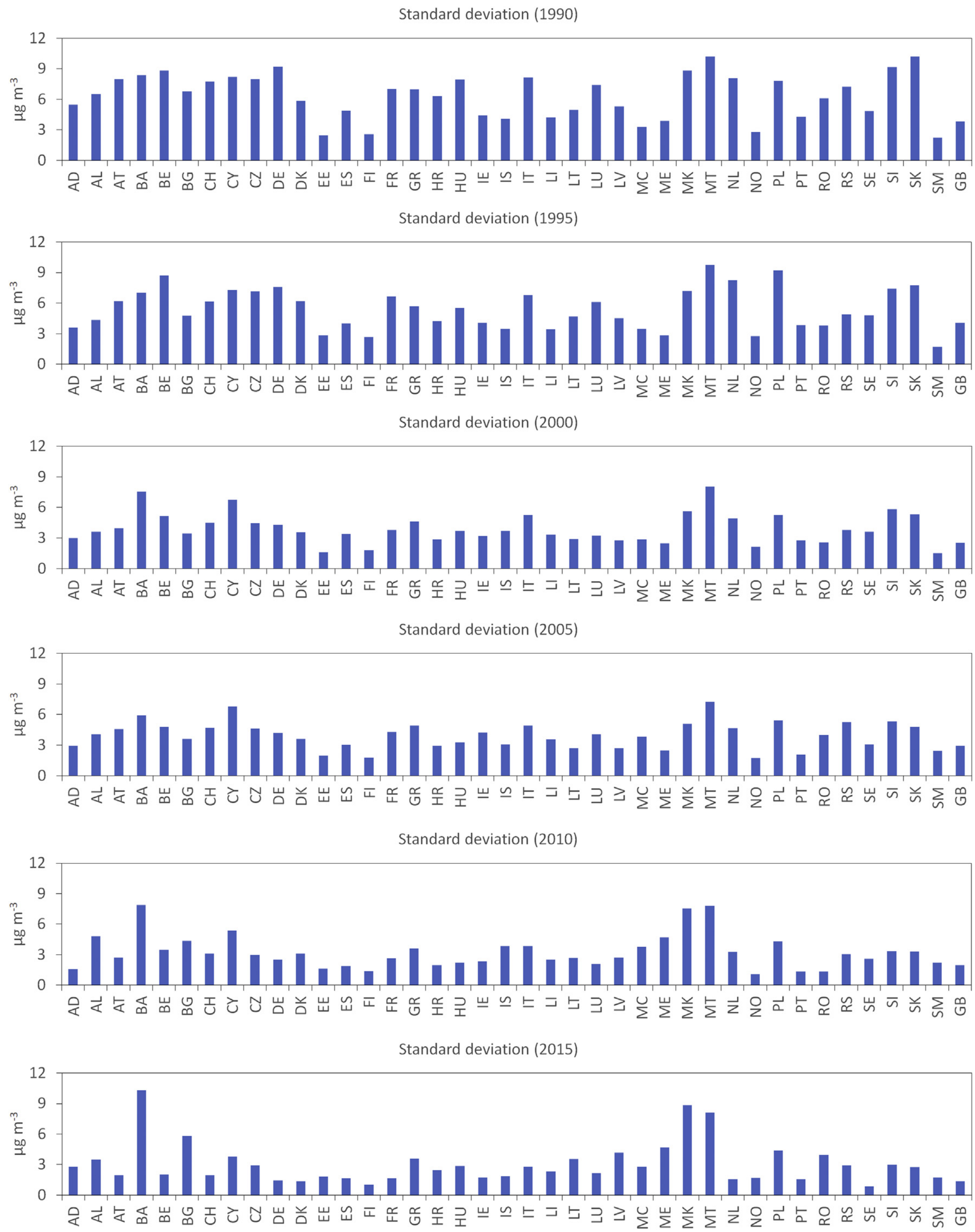

Fig. 3. The variability (standard deviation) in population weighted $PM_{2.5}$ concentrations (µg m^{-3}), i.e. exposure to $PM_{2.5}$, in 1990, 1995, 2000, 2005, 2010 and 2015 (from the top to the bottom) among the different sources of model data for all the available countries (indicated with their ISO codes).

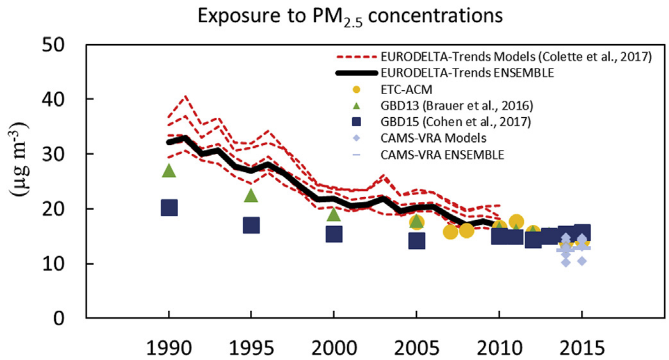

Fig. 4. Population weighted $PM_{2.5}$ concentrations (μg m^{-3}), i.e. exposure to $PM_{2.5}$, as modeled by all the available datasets for the 1990–2015 period (total of all the available countries).

observations in more recent years to perform the bias corrections.

Fig. 4 reports the exposure to $PM_{2.5}$ calculated over the 1990–2015 period, using all the datasets available in each year and for all the considered countries. A downward trend is clearly seen in all data sets, but to a varying extent. The analysis is complicated by the fact that different data sets cover different periods of time (see section 2.1). We first analyzed the trend in exposure to $PM_{2.5}$ between 1990 and 2010, because of the availability of three data sets over the whole period. Indeed, the GBD13 and GBD15 datasets overlap with five years of the EDT analysis (i.e. 1990, 1995, 2000, 2005 and 2010). Exposure to $PM_{2.5}$ was estimated to have declined by about 50%, 40% and 28% in the EDT ensemble median, GBD13 and GBD15 datasets, respectively, which corresponds to a difference of about a factor of nearly 2 for all the countries included in the analysis (Table 1). Thus, the EDT derived trend is larger than the GDB one, and especially than GBD15. Possible reasons for this will be discussed in the next section. In addition, we aimed at estimating a trend covering the whole period 1990–2015. For this, we derived a linear trend of the median of all the available datasets (Table 1), each EDT model counting individually. Such a procedure is justified, if errors in individual data sets are decorrelated between different years. Even if this is a strong hypothesis, it is probably partly true. For instance, emissions are certainly much more uncertain in the 90's than in more recent years. For GDB data sets, much less data were available in the 90's and the method is different for these years. Our alternative method yields a $PM_{2.5}$ exposure decline of 60% for the 1990–2015 period. It also can be noted that mainly from the year 2005 onwards, almost no trends in exposure to $PM_{2.5}$ are observed in most of the datasets (Fig. 4).

Fig. 5 shows the spatial distribution of the linear trends in exposure to $PM_{2.5}$ as predicted between 1990 and 2010, using the GBD13, GBD15 and EDT datasets, and between 1990 and 2015 using the median of all

Table 1
Linear relative and absolute trends in population weighted $PM_{2.5}$ concentrations, i.e. exposure to $PM_{2.5}$, as modeled by the EDT, GBD13 and GBD15 datasets for the 1990–2010 period and as modeled by the median of all the datasets (referred to as ALL) for the 1990–2015 period.

Countries	EDT (%)	GBD13 (%)	GBD15 (%)	ALL (%)	EDT (μg m^{-3} y^{-1})	GBD13 (μg m^{-3} y^{-1})	GBD15 (μg m^{-3} y^{-1})	ALL* (μg m^{-3} y^{-1})
Albania	−44.7	−26.2	−21.7	−47.3	−0.6	−0.3	−0.2	−0.5
Andorra	−38.1	−64.2	0.0	−60.3	−0.4	−0.8	0.0	−0.6
Austria	−52.8	−47.2	−22.0	−65.3	−1.0	−0.7	−0.2	−1.0
Belgium	−54.1	−36.1	−20.6	−68.7	−1.1	−0.5	−0.2	−1.1
Bos. & Her.	−46.5	−31.5	−19.4	−44.0	−0.7	−0.3	−0.4	−0.5
Bulgaria	−50.5	−25.6	−45.9	−38.1	−0.8	−0.3	−0.9	−0.4
Croatia	−50.9	−41.2	−27.8	−59.0	−0.9	−0.6	−0.3	−0.8
Cyprus	−30.6	−29.0	−5.1	−24.2	−0.4	−0.5	0.0	−0.2
Czech Repu.	−58.6	−50.6	−50.6	−65.1	−1.2	−0.8	−0.8	−1.1
Denmark	−49.6	−46.2	−25.0	−65.7	−0.7	−0.5	−0.2	−0.7
Estonia	−46.3	−15.8	−33.3	−51.3	−0.3	−0.1	−0.2	−0.3
Finland	−38.3	−24.2	−10.5	−47.2	−0.2	−0.1	0.0	−0.2
France	−48.7	−39.0	−14.5	−61.5	−0.8	−0.5	−0.1	−0.8
Germany	−59.7	−49.8	−19.8	−69.1	−1.0	−0.7	−0.2	−1.0
Greece	−37.5	−29.5	−9.5	−42.6	−0.5	−0.4	−0.1	−0.5
Hungary	−60.7	−44.1	−42.8	−65.3	−1.3	−0.7	−0.7	−1.1
Iceland	−25.1	−17.0	0.0	−26.8	−0.1	0.0	0.0	−0.1
Ireland	−47.0	−44.3	−18.5	−61.5	−0.4	−0.4	−0.1	−0.5
Italy	−47.5	−35.0	−24.2	−57.4	−0.9	−0.6	−0.2	−0.9
Latvia	−47.4	−28.0	−43.3	−55.9	−0.5	−0.2	−0.6	−0.5
Liechtenstein	−50.4	–	–	−64.5	−0.7	–	–	−0.7
Lithuania	−48.5	−37.3	−42.6	−55.1	−0.6	−0.3	−0.6	−0.6
Luxembourg	−55.1	−58.4	−20.6	−67.0	−0.9	−1.1	−0.2	−0.9
Macedonia	−38.0	−35.4	−39.0	−12.1	−0.5	−0.5	−0.9	−0.1
Malta	−38.7	−29.6	0.0	−49.0	−0.5	−0.4	0.0	−0.6
Monaco	−45.2	–	–	−60.6	−0.8	–	–	−0.9
Montenegro	−37.9	−35.4	−16.5	−35.4	−0.5	−0.3	−0.2	−0.3
Netherlands	−55.5	−45.2	−21.7	−68.9	−1.0	−0.7	−0.2	−1.0
Norway	−40.7	−32.7	−18.5	−53.2	−0.3	−0.2	−0.1	−0.3
Poland	−49.1	−43.3	−45.3	−56.3	−1.1	−0.6	−0.9	−1.0
Portugal	−45.6	−27.1	−7.8	−53.1	−0.5	−0.2	0.0	−0.4
Romania	−53.7	−38.9	−45.3	−55.0	−0.9	−0.5	−0.7	−0.7
San Marino	−46.8	–	–	−58.4	−0.8	–	–	−0.8
Serbia	−42.4	−36.6	−22.6	−43.7	−0.7	−0.3	−0.3	−0.5
Slovakia	−58.9	−52.1	−45.3	−66.7	−1.3	−0.8	−0.7	−1.2
Slovenia	−49.8	−39.3	−36.2	−61.9	−0.9	−0.4	−0.4	−0.9
Spain	−34.2	−27.2	0.0	−45.4	−0.4	−0.3	0.0	−0.4
Sweden	−43.6	−31.6	−29.4	−62.3	−0.4	−0.2	−0.1	−0.5
Switzerland	−50.5	−31.5	−15.9	−62.3	−0.8	−0.4	−0.1	−0.8
Unit. King.	−47.9	−46.6	−21.6	−60.8	−0.6	−0.5	−0.2	−0.6
All countries	−50.4	−40.4	−28.2	−59.9	−0.8	−0.5	−0.3	−0.8

(*) Calculated using the median of the EDT, GBD13, GBD15, ETC-ACM and CAMS datasets.

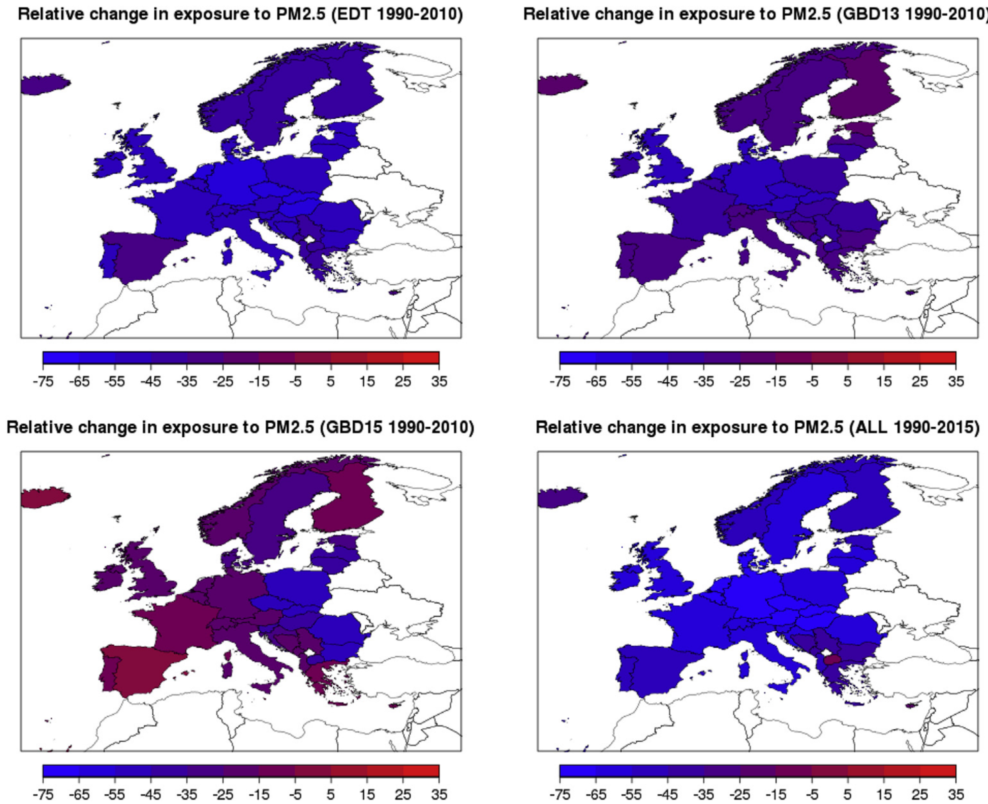

Fig. 5. Relative trends in country averaged population weighted $PM_{2.5}$ concentrations, i.e. exposure to $PM_{2.5}$, for the EDT, GBD13 and GBD15 datasets (1990–2010 period) and for the median of the EDT, GBD13, GBD15, ETC-ACM and CAMS-VRA dataset (referred to as ALL for the 1990–2015 period).

available datasets. In general, the EDT datasets predicted larger reductions in exposure to $PM_{2.5}$ during the 1990–2010 period compared to the GBD13 and especially to the GBD15 datasets. For the latter, the estimated relative reductions were closer to those estimated by the EDT dataset in some of the eastern European countries (Fig. 5 and Table 1), whereas discrepancies were particularly remarkable (up to a factor of 3–5) in Switzerland, Cyprus, Germany, Finland, France, Greece and Portugal. The median of all the available datasets for the 1990–2015 period indicated that exposure to $PM_{2.5}$ has declined by about 12–69% with larger reductions predicted in e.g. Germany and Netherlands and Belgium, and lower reductions predicted in eastern European countries such as Bulgaria, Montenegro and Macedonia (Fig. 5 and Table 1).

3.2. Discussion on the exposure to $PM_{2.5}$

The observed differences in the predicted exposure to $PM_{2.5}$ could be mainly related to the different methodologies and the underlying information used to retrieve the exposure in the studied years. The GBD and EDT datasets use a different methodology to estimate the $PM_{2.5}$ concentrations, especially in the early 1990s. In the GBD datasets simulated AOD values from the GEOS-Chem model are used to estimate surface level of $PM_{2.5}$ concentrations in combination with satellite-based measurements. Different anthropogenic emissions were used in the GBD and EDT datasets. The GEOS-Chem model simulations were based on the EDGAR emissions inventories whereas the EDT dataset is based on the ECLIPSE emission inventory. Recent studies focusing on Asia indicated that substantial differences could be found among the two emission inventories (Saikawa et al., 2017) and the need to further reconcile the differences among them. In addition, for the years 1990 and 1995, the concentrations were estimated by scaling the simulated $PM_{2.5}$ concentrations for 2005 using constant meteorological data. This is not the case for the EDT dataset where "real" year-to-year meteorology was used for each of the 21 years covered in the exercise. Finally, the approach used to correct and calibrate the gridded concentrations over the whole domain might also play an important role for estimating the global (for GBD) and regional (for EDT) $PM_{2.5}$ concentrations. The EDT gridded dataset was corrected using the raw model output data and the kriged analysis using available $PM_{2.5}$ measurements (mainly during the late 2000s) and applied to the whole 21-year simulation period. The measurements were retrieved from the AQ e-reporting database available for Europe. This approach differs from the one used in GBD13 where global $PM_{2.5}$ measurements (mainly between 2010 and 2013) were used to retrieve a single global calibration function that was later applied to correct the gridded $PM_{2.5}$ concentration fields, regardless of the specific geographic region (e.g. Europe, Asia). Even though several different calibration functions were tested for various regions (i.e. using $PM_{2.5}$ measurement belonging only to each specific geographical area), a single global calibration function was chosen due to the poor fit in some of the regions (Brauer et al., 2016). Nevertheless, differences in the $PM_{2.5}$ composition, and their trends, might significantly differ depending of the specific geographical area. This might help explaining the differences with the EDT dataset which was corrected specifically with European measurements. Other discrepancies between the two GBD datasets, i.e. GBD13 and GBD15, are likely due to the updated correction algorithm deployed in the updated version of the GBD dataset, i.e. GBD15. For the latter, global calibration functions could vary spatially in the domain, but TM5 model simulations were not included in the calibration process, contrary to GBD13.

Long-term measurements of $PM_{2.5}$ concentrations dating back to 1990 are very scarce in Europe. Moreover, the trends of the individual $PM_{2.5}$ components, i.e. both inorganic and organic phase, is yet not well understood due to the non-linear dependence of aerosol constituents on its gas-phase precursors. Tørseth et al. (2012) presented a trend analysis study for particulate sulfate, total nitrate (the sum of nitric acid and particulate nitrate) and total ammonium (the sum of ammonia and particulate ammonium) for the 1990–2009 period in Europe. Their

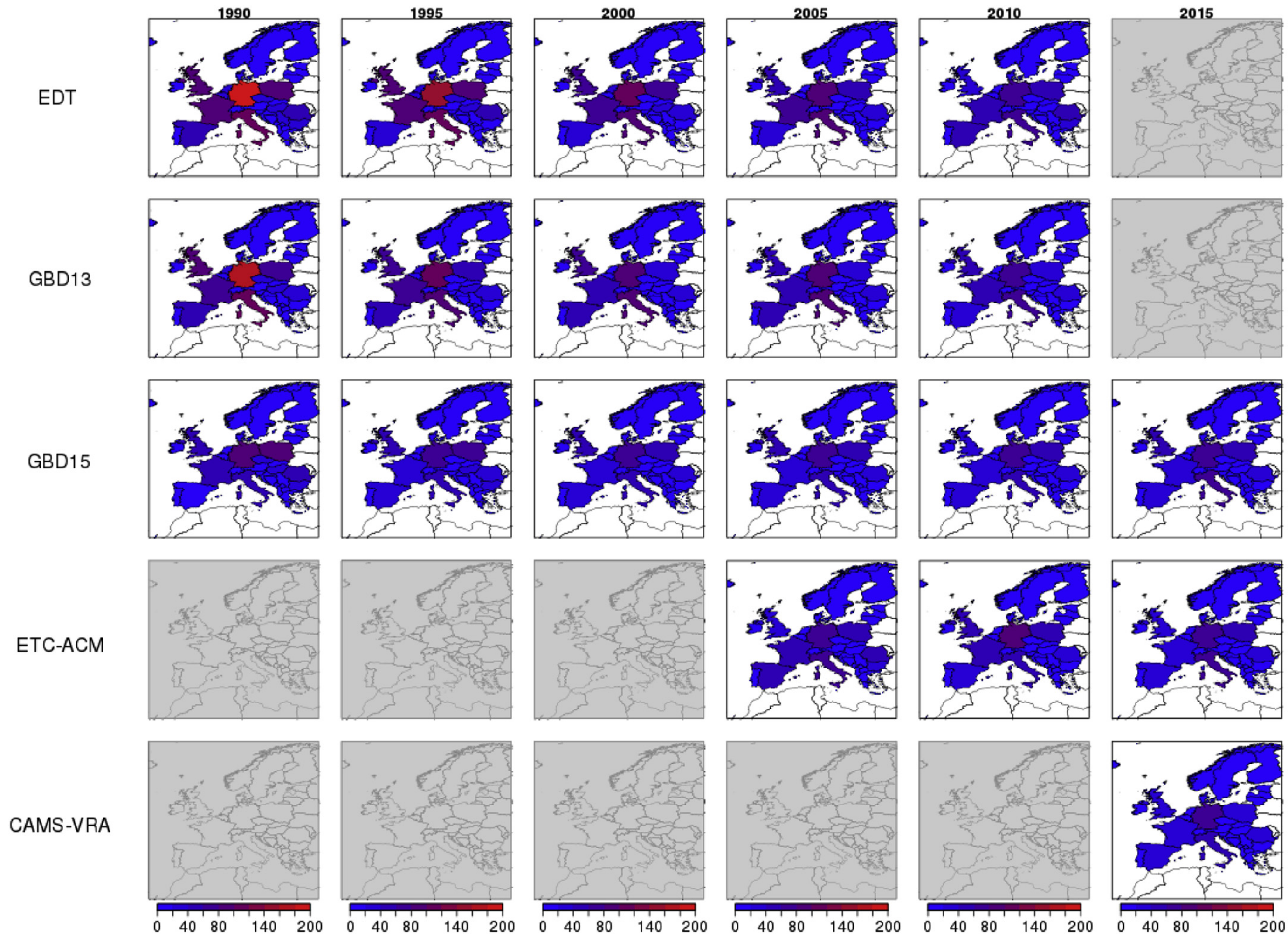

Fig. 6. Annual national average premature deaths due to exposure to $PM_{2.5}$, as predicted by EDT (model ensemble), GBD13, GBD15, ETC-ACM and CAMS-VRA datasets (model ensemble) in 1990, 1995, 2000, 2005, 2010 and 2015. Grey maps indicate years for which the corresponding dataset is not available. Units are estimated national total premature deaths (in thousands).

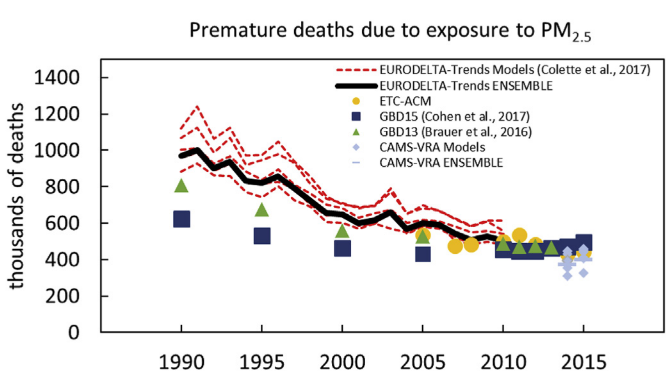

Fig. 7. Trends in premature deaths (in thousands of deaths) due to exposure to $PM_{2.5}$ as predicted by all the available datasets for the 1990–2015 period (total of all the available countries).

analysis indicated a relative reduction of 56% in particulate sulfate concentrations, 8% in total nitrate and 24% in total ammonium concentrations. More recently (Colette et al., 2016), reported a decrease of about 65% in particulate sulfate concentrations between the 1990–2012 period. Even though those trends were calculated at few specific sites, and only partly cover the $PM_{2.5}$ species, such reductions are at least not contradicting the one predicted by the EURODELTA-Trends exercise (i.e. around 50% reduction in exposure to $PM_{2.5}$ for the total of all the available countries). Ciarelli et al. (2019) showed that for the 1990–2010 period, the models which participated in the exercise could reproduce very well the observed trends in particulate sulfate, which were predicted to have declined to a greater extent respect to total nitrate and total ammonium concentrations, as also indicated by the observations. PM_{10} measurement are too scare in the 1990s in Europe to allow for an evaluation, but looking at the 2000–2010 period, the EURODELTA-Trends ensemble was found to capture well the overall decline with a 1.94%/yr average decline, whereas the observed decline at Air Quality e-reporting was 1.87%/yr (Colette et al., 2017b).

In the next section (section 3.3), the mortality trends (in terms of number of premature deaths) are presented based on the exposure analysis discussed in this section.

3.3. $PM_{2.5}$ health impact assessment trends

Fig. 6 shows the predicted number of premature deaths for all the datasets used in the study and for six specific years, i.e. 1990, 1995, 2000, 2005, 2010 and 2015. In line with the predicted exposure to $PM_{2.5}$, larger discrepancies between the datasets can be observed in the early 1990s than in more recent years (i.e. 2010 and 2015), consistent with the results for exposure to $PM_{2.5}$ (see section 2.1 and previous discussions in section 3.2). For the first analyzed year, i.e. 1990, health impact calculations based on the GBD15 dataset indicated about

Table 2
Premature deaths due to exposure to $PM_{2.5}$ as median of all the datasets available in 1990, 2010 and 2015.

Countries	Median (1990)	Median (2010)	Median (2015)
Albania	2227	1060	1183
Austria	17 308	8191	6489
Belgium	20 895	11 417	7489
Bos. & Her.	5462	2979	4006
Bulgaria	21 713	12 054	11 016
Croatia	10 744	5694	5174
Cyprus	716	543	580
Czech Repu.	32 615	13 029	10 789
Denmark	8438	4131	2935
Estonia	969	815	668
Finland	3057	2236	1600
France	88 872	47 671	35 755
Germany	177 708	80 350	66 283
Greece	15 531	11 279	10 490
Hungary	37 468	15 064	12 906
Iceland	75	78	52
Ireland	3273	1529	945
Italy	120 622	71 517	65 811
Kosovo*	3958	2524	2583
Latvia	3341	2481	1604
Liechtenstein	48	29	23
Lithuania	4078	3811	2848
Luxembourg	794	331	271
Macedonia	2297	2104	2903
Malta	416	315	360
Montenegro	815	552	492
Netherlands	24 488	13 961	9992
Norway	3412	2050	1359
Poland	94 165	56 966	44 903
Portugal	11 486	5851	6789
Romania	52 426	27 377	27 688
Serbia*	15 673	9994	10 226
Slovakia	14 792	6823	5424
Slovenia	4017	2071	2161
Spain	40 940	28 064	31 060
Sweden	8326	4787	3091
Switzerland	10 785	5963	4952
Unit. King.	86 872	41 386	32 779
All countries	960 419	498 168	444 535

(*) The same exposure and baseline mortality are used for Serbia and Kosovo, but mortality attributed to $PM_{2.5}$ is split by using the relative population of Serbia and Kosovo and with same population data for the years 1990 and 2015.

622 000 premature deaths from chronic exposure to $PM_{2.5}$ (representing the lowest estimate among all the datasets), whereas the model estimates in the EURODELTA-Trends exercise lie between about 881 000 and 1 118 000 premature deaths (Fig. 7). For the same year, the median of all the data sets indicated a total number of premature deaths of 960 000 (total of all considered countries, Table 2). The three countries with the highest total number of premature deaths according to the calculations were Germany, Italy and Poland with 178 000, 121 000 and 94 000 premature deaths due to exposure to $PM_{2.5}$, respectively (Table 2). For the last available year of this study, i.e. 2015, the total burden of premature deaths from chronic exposure to $PM_{2.5}$ was found to be largely reduced compared to 1990, with a total number of deaths for all the investigated countries of about 445 000 (median of all the datasets available in 2015, Table 2). This is in line with the study of Im et al. (2018) where around 390 000 ± 100 000 premature deaths for Europe (30°W-60°E, 25–70°N) for the year 2010 due to exposure to $PM_{2.5}$ were predicted. Also, the gradient of premature deaths among the different countries is comparable with the study performed by (Solazzo et al., 2018) even though their estimates are slightly lower, likely because of a minimum threshold concentration of 5.8 $\mu g\, m^{-3}$ used to perform the HIA calculations.

Fig. 7 reports the linear trends in premature deaths which, by construction, are similar to those obtained for exposure to $PM_{2.5}$: for the 1990–2010 period, the relative decline in the number of premature deaths was found to be around 52%, 41% and 30% for the EDT, GBD13 and GBD15 datasets, respectively, corresponding to an absolute declining rate of about 25 000 deaths y^{-1}, 16 000 deaths y^{-1} and 9000 deaths y^{-1} (Table 3). Taking again the median of all the datasets included in the analysis for the entire period (1990–2015) and for the total of all the countries, a decline of about 60% is found, corresponding to an absolute reduction rate of about 23 000 deaths y^{-1} (Fig. 7 and Table 3). Finally, mainly from the year 2005 onwards, almost no trends in the number of premature deaths due to exposure to $PM_{2.5}$ were indicated by most of the datasets (Fig. 7).

Recent health impact studies were performed with the GBD15 datasets over the US domain. Zhang et al. (2018), compared the trends in total number of deaths predicted using exposure calculated with the WRF-CMAQ model and the one available in the GBD15 datasets for the 1990–2010 period. Even though the methodology to retrieve the number of premature deaths attributable to exposure to $PM_{2.5}$ largely differs from the analysis presented here, their results also indicated a relatively flat trend in the total number of deaths predicted with the GBD15 dataset compared to the one predicted by the WRF-CMAQ model over the 1990–2010 period (Zhang et al., 2018).

Fig. 8 shows the spatial distribution of the linear trends in premature deaths as retrieved for the 1990–2010 period, using the GBD13, GBD15 and EDT datasets, and between 1990 and 2015 using the median of all available datasets. In general, The EDT datasets indicated larger reductions in the number of premature deaths during the 1990–2010 period compared to the GBD13 and especially to the GBD15 datasets. For the latter, the estimated relative reductions were closer in some of the Eastern European countries (Fig. 8 and Table 3), whereas discrepancies were higher in Switzerland, Cyprus, Finland, France, Greece and Portugal. The median of all the available datasets for the 1990–2015 period indicated that the number of premature deaths have declined by about 10–79% with larger reductions (above 70%) predicted in Lichtenstein, Denmark, the Czech Republic, Germany, Hungary and Luxemburg, and lower relative reductions (below 30%) predicted in Greece, Bosnia-Herzegovina, Montenegro, Cyrus and Iceland (Fig. 8 and Table 3). The increase in the total number of premature deaths over Macedonia during 1990–2015 is mainly due to the relatively low reduction in the exposure to $PM_{2.5}$ predicted between 2010 and 2015 by all the available datasets combined with an increase in the baseline mortality (not shown). For the GBD15 datasets the predicted increases in premature deaths for some of the available countries, i.e. Greece, Iceland, Malta and Spain, are mainly due to the absence of trends, or very weak trends, in the exposure to $PM_{2.5}$ (Table 1) and thus the predicted premature deaths were driven by the variation in the baseline mortality. We would like to notice that for some specific countries such as Bosnia and Herzegovina or Serbia and Montenegro, important gaps where found in terms of the mortality data (e.g. for Bosnia and Herzegovina data were available only for 1990, 1991, 2011 and 2014, and mortality for intermediate years was estimated using the latest available years) and the interpretation of the results for these countries must be taken with care.

3.4. Comparisons with the baseline mortality

In this section the comparison of premature deaths due to exposure to $PM_{2.5}$ with the baseline mortality is discussed. The baseline mortality, i.e. natural deaths for the age-group above 30 years, is used to estimate the total number of premature deaths due to $PM_{2.5}$ exposure based on the dose response function described in section 2.3. Fig. S1 shows the total number of natural deaths for age-group above 30 years as well as the contribution of premature deaths due to exposure to $PM_{2.5}$ to the base line mortality for all the countries included in the analysis (apart from Kosovo and Lichtenstein) and for the EDT dataset (1990–2010 period). The total number of natural deaths for age-group above 30 years varies between about 4 670 000 and 4 890 000 and the contribution of premature deaths due to exposure to $PM_{2.5}$ was found to

Table 3
Linear relative and absolute trends in premature deaths to the exposure to $PM_{2.5}$, as predicted by the EDT, GBD13 and GBD15 datasets for the 1990–2010 period and as predicted by the median of all the datasets (referred to as ALL) for the 1990–2015 period.

Countries	EDT (%)	GBD13 (%)	GBD15 (%)	ALL (%)	EDT (deaths y^{-1})	GBD13 (deaths y^{-1})	GBD15 (deaths y^{-1})	ALL (deaths y^{-1})
Albania	−28.4	−22.1	−19.0	−43.9	−28	−17	−12	−35
Austria	−58.7	−52.1	−28.6	−68.7	−500	−352	−122	−476
Belgium	−56.2	−37.7	−22.5	−69.8	−656	−334	−130	−676
Bos. & Her.	−45.5	−28.2	−15.5	−28.9	−107	−42	−54	−51
Bulgaria	−47.8	−21.5	−42.4	−37.3	−482	−156	−554	−279
Croatia	−47.7	−38.7	−24.7	−56.1	−231	−152	−81	−222
Cyprus	−30.1	−28.9	−5.0	−18.3	−11	−16	−1	−5
Czech Repu.	−66.5	−60.5	−60.6	−71.7	−971	−645	−714	−801
Denmark	−56.2	−53.8	−35.0	−72.4	−260	−217	−79	−286
Estonia	−40.5	−11.0	−27.7	−50.9	−26	−6	−13	−28
Finland	−40.5	−25.7	−12.0	−45.9	−63	−37	−12	−60
France	−49.3	−38.7	−14.1	−61.1	−2302	−1437	−284	−2305
Germany	−64.0	−54.7	−26.3	−71.5	−5827	−3985	−1122	−5066
Greece	−26.2	−16.5	6.8	−29.8	−202	−122	24	−182
Hungary	−66.1	−49.1	−48.0	−71.3	−1133	−602	−673	−943
Iceland	−15.4	−3.9	15.4	−10.1	−1	0	1	0
Ireland	−56.0	−54.0	−32.6	−68.7	−99	−85	−33	−99
Italy	−43.6	−29.7	−18.0	−52.6	−2549	−1559	−516	−2613
Kosovo	−37.2	−30.8	−15.9	−39.1	−68	−34	−21	−57
Latvia	−32.8	−7.9	−26.0	−48.4	−55	−8	−50	−70
Liechtenstein	−47.7	–	–	−78.7	−1	–	–	−2
Lithuania	−29.2	−9.5	−15.2	−39.5	−69	−15	−36	−78
Luxembourg	−60.4	−63.0	−28.0	−70.5	−22	−26	−6	−21
Macedonia	−9.6	−7.1	−10.6	32.6	−11	−8	−21	28
Malta	−27.1	−17.6	16.4	−36.3	−6	−4	2	−7
Montenegro	−29.0	−27.1	−5.3	−24.9	−11	−8	−2	−7
Netherlands	−53.7	−42.2	−18.3	−67.2	−770	−495	−130	−794
Norway	−48.1	−41.8	−29.3	−60.4	−92	−70	−42	−97
Poland	−51.8	−44.5	−46.5	−56.8	−2425	−1359	−2014	−2077
Portugal	−42.6	−22.8	−2.6	−51.1	−253	−110	−8	−245
Romania	−51.4	−32.6	−39.2	−53.3	−1249	−586	−819	−1015
Serbia	−37.2	−30.8	−15.9	−39.1	−270	−134	−84	−225
Slovakia	−56.7	−50.0	−43.0	−65.9	−380	−218	−203	−349
Slovenia	−50.4	−37.3	−34.1	−61.0	−100	−36	−41	−97
Spain	−22.1	−14.5	17.5	−32.6	−456	−313	167	−561
Sweden	−47.5	−36.3	−34.2	−65.8	−224	−134	−64	−273
Switzerland	−51.6	−32.0	−16.3	−62.0	−276	−155	−36	−269
Unit. King.	−56.9	−54.9	−33.5	−69.4	−2680	−2358	−961	−2719
All countries	−52.0	−41.1	−29.8	−60.2	−24 863	−15 833	−8747	−23 238

be around 20 and 10% over the 1990–2010 period (Fig. S1).

4. Conclusions

In this study, trends in the total number of premature deaths from chronic exposure to $PM_{2.5}$ concentrations were calculated based on different datasets of $PM_{2.5}$ in Europe during the 1990–2015 period. The new available EURODELTA-Trends dataset provided novel $PM_{2.5}$ concentrations estimates over Europe for the 1990–2010 period, corrected with data fusion technique based exclusively on European measurements. In this paper, we have presented a detailed comparison with other available datasets - GBD13 and GBD15 - to investigate the differences in the number of premature deaths as estimated using the different datasets. Even though the three datasets, i.e. EDT, GBD13 and GBD15, indicated substantial reductions in both exposure to $PM_{2.5}$ and related number of premature deaths for the overall countries' totals, large differences were observed for the absolute and relative reductions, especially at single country levels. The EDT datasets indicated about 52% reduction in the number of premature deaths for the countries' total whereas the GBD13 and GBD15 showed lower relative reductions rates, around 41% and 30%, respectively. Especially in the year 1990, the differences in the predicted number of premature deaths were found to be larger, i.e. around a factor of 2, compared to the final years of the investigated periods, and in line with previous studies (Im et al., 2018). Such discrepancies were mainly attributed to the different methodologies used in the datasets to estimate the $PM_{2.5}$ concentrations, and in particular to: (i) the different chemical model, i.e. GEOS-Chem, as well as anthropogenic emission inventory, i.e. EDGAR, used to correct satellite-based $PM_{2.5}$ estimates in the GBD datasets for the years 1990 and 1995, whereas the EDT dataset relies on the ECLIPSE anthropogenic emission datasets, (ii) the constant meteorological fields, i.e. based on the year 2005, used for correcting the GBD satellite-based estimates for the year 1990, whereas EDT used "real" year-to-year meteorology, (iii) the single correction function based on global $PM_{2.5}$ measurements that was used to correct the gridded $PM_{2.5}$ concentrations in the GBD13 datasets, whereas the EDT dataset was corrected exclusively with European measurements and (iv) the exclusion of chemical transport model simulation, i.e. TM5, from the GBD15 dataset.

Based on the linear trend of the median of all the available datasets, our results indicate that over the whole period the total number of premature deaths has declined by about 60%. This corresponds to an absolute reduction rate of about 23 000 deaths y^{-1} for all the countries included in the analysis. The total burden of premature deaths due to exposure to $PM_{2.5}$, and its relative reductions over the investigated period, were particularly elevated for countries such as Germany, Poland, Italy, France and the United Kingdom, i.e. ranging from 87 000 to 178 000 premature deaths in the year 1990 as median of all the datasets available, and which were found to be largely reduced at the end of the full investigated period, i.e. by between 53 and 71%.

In conclusion, the estimated reduction in the number of deaths due to exposure to $PM_{2.5}$ concentrations suggest that European legislation towards air quality mitigation strategies was effective in lowering the total burden of premature deaths related to $PM_{2.5}$. However, after the

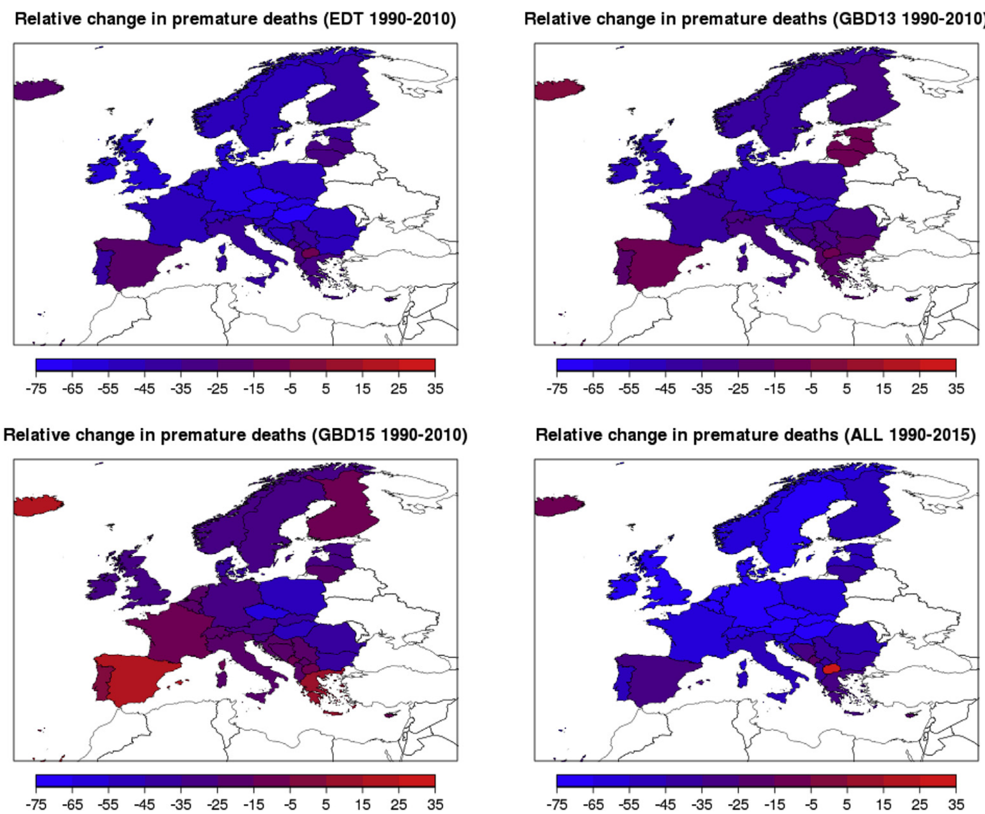

Fig. 8. Relative trends (%) in premature deaths due to exposure to $PM_{2.5}$, for the EDT, GBD13 and GBD15 datasets (1990–2010 period) and for the median of the EDT, GBD13, GBD15, ETC-ACM and CAMS-VRA dataset (referred to as ALL for the 1990–2015 period).

year 2005 onwards, the decrease slowed down considerably and almost no trends in the number of premature deaths due to exposure to $PM_{2.5}$ were indicated by most of the datasets. Health impacts from $PM_{2.5}$ in Europe remain considerable, with about 445 000 premature deaths in 2015 as indicated by the median of all the $PM_{2.5}$ datasets used in this study. Therefore, further efforts to reduce $PM_{2.5}$ concentrations are needed to lower the total burden of premature deaths. As more long-term measurements, and CTMs simulations, of the individual $PM_{2.5}$ components are becoming increasingly available, future health impact studies will need to focus on the specific health effects of the individual $PM_{2.5}$ components, since not all the different chemical species, i.e. organic and inorganic constituents, might have the same toxicity, as assumed in the present study. Another recognized focus is on the size distribution within the $PM_{2.5}$ fraction, since systematic measurements of ultrafine particles mass and the total number of particles are still scarce.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.aeaoa.2019.100032.

References

Atkinson, R.W., Anderson, H.R., Sunyer, J., Ayres, J., Baccini, M., Vonk, J.M., Boumghar, A., Forastiere, F., Forsberg, B., Touloumi, G., Schwartz, J., Katsouyanni, K., 2001. Acute effects of particulate air pollution on respiratory admissions: results from APHEA 2 project. Air Pollution and Health: a European Approach. Am. J. Respir. Crit.

Care Med. 164, 1860–1866. https://doi.org/10.1164/ajrccm.164.10.2010138.
Atkinson, R.W., Mills, I.C., Walton, H.A., Anderson, H.R., 2015. Fine particle components and health—a systematic review and meta-analysis of epidemiological time series studies of daily mortality and hospital admissions. J. Expo. Sci. Environ. Epidemiol. 25, 208–214. https://doi.org/10.1038/jes.2014.63.
Brandt, J., Silver, J.D., Christensen, J.H., Andersen, M.S., Bønløkke, J.H., Sigsgaard, T., Geels, C., Gross, A., Hansen, A.B., Hansen, K.M., Hedegaard, G.B., Kaas, E., Frohn, L.M., 2013. Contribution from the ten major emission sectors in Europe and Denmark to the health-cost externalities of air pollution using the EVA model system – an integrated modelling approach. Atmos. Chem. Phys. 13, 7725–7746. https://doi.org/10.5194/acp-13-7725-2013.
Brauer, M., Freedman, G., Frostad, J., van Donkelaar, A., Martin, R.V., Dentener, F., Dingenen, R. van, Estep, K., Amini, H., Apte, J.S., Balakrishnan, K., Barregard, L., Broday, D., Feigin, V., Ghosh, S., Hopke, P.K., Knibbs, L.D., Kokubo, Y., Liu, Y., Ma, S., Morawska, L., Sangrador, J.L.T., Shaddick, G., Anderson, H.R., Vos, T., Forouzanfar, M.H., Burnett, R.T., Cohen, A., 2016. Ambient air pollution exposure estimation for the global burden of disease 2013. Environ. Sci. Technol. 50, 79–88. https://doi.org/10.1021/acs.est.5b03709.
Ciarelli, G., Theobald, M.R., Vivanco, M.G., Beekmann, M., Aas, W., Andersson, C., Bergström, R., Manders-Groot, A., Couvidat, F., Mircea, M., Tsyro, S., Fagerli, H., Mar, K., Raffort, V., Roustan, Y., Pay, M.-T., Schaap, M., Kranenburg, R., Adani, M., Briganti, G., Cappelletti, A., D 'Isidoro, M., Cuvelier, C., Cholakian, A., Bessagnet, B., Wind, P., Colette, A., 2019. Trends of inorganic and organic aerosols and precursor gases in Europe: insights from the EURODELTA multi-model experiment over the 1990 – 2010 period. Geosci. Model Dev. Discuss. 1–49. https://doi.org/10.5194/gmd-2019-70.
Cohen, A.J., Brauer, M., Burnett, R., Anderson, H.R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R., Feigin, V., Freedman, G., Hubbell, B., Jobling, A., Kan, H., Knibbs, L., Liu, Y., Martin, R., Morawska, L., Pope, C.A., Shin, H., Straif, K., Shaddick, G., Thomas, M., Dingenen, R. van, Donkelaar, A. van, Vos, T., Murray, C.J.L., Forouzanfar, M.H., 2017. Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015. Lancet 389, 1907–1918. https://doi.org/10.1016/S0140-6736(17)30505-6.
Colette, A., Aas, W., Banin, L., Braban, F.C., Ferm, M., Ortiz, G.A., Llyin, I., Mar, K., Pandolfi, M., Putaud, J.-P., Shatalov, V., Solberg, S., Spindler, G., Tarasova, O., Vana, M., Adani, M., Almodovar, P., Berton, E., Bessagnet, B., Nizzetto, B.P., Boruvkova, J., Breivik, K., Briganti, G., Cappelletti, A., Cuvelier, C., Derwent, R., D'Isidoro, M., Fagerli, H., Funk, C., Vivanco, M.G., Haeuber, R., Hueglin, C., Jenkins, S., Kerr, J., De Leeuw, F., Lynch, J., Manders, A.M.M., Mircea, M., Pay, M.T., Pritula, D., Querol, X., Raffort, V., Reiss, I., Roustan, Y., Sauvage, S., Scavo, K., Simpson, D., Smith, I.R., Tang, S.Y., Theobald, M.R., Tørseth, K., Tsyro, S., van Pul, A., Vidic, S., Wallasch, M., Wind, P., 2016. Air Pollution Trends in the EMEP Region between 1990 and 2012.
Colette, A., Andersson, C., Manders, A., Mar, K., Mircea, M., Pay, M.P., Raffort, V., Tsyro, S., Cuvelier, C., Adani, M., Bessagnet, B., Bergström, R., Briganti, G., Butler, T., Cappelletti, A., Couvidat, F., D&apos, Isidoro, M., Doumbia, T., Fagerli, H., Granier, C., Heyes, C., Klimont, Z., Ojha, N., Otero, N., Schaap, M., Sindelarova, K., Stegehuis, A.I., Roustan, Y., Vautard, R., van Meijgaard, E., Vivanco, M.G., Wind, P., 2017a. EURODELTA-Trends, a multi-model experiment of air quality hindcast in Europe over 1990–2010. Geosci. Model Dev 10, 3255–3276. https://doi.org/10.5194/gmd-10-3255-2017.
Colette, A., Solberg, S., Beauchamp, M., Bessagnet, B., Malherbe, L., Guerreiro, C., 2017b. Long Term Air Quality Trends in Europe: Contribution of Meteorological Variability, Natural Factors and Emissions.
COMEAP, 2015. Statement of the Evidence for Differential Health Effects of Particulate Matter According to Source or Components. Public Health England.
Cressie, N., 1990. The origins of kriging. Math. Geol. 22, 239–252. https://doi.org/10.1007/BF00889887.
Crippa, M., Janssens-Maenhout, G., Guizzardi, D., Van Dingenen, R., Dentener, F., 2019. Contribution and uncertainty of sectorial and regional emissions to regional and global $PM_{2.5}$ health impacts. Atmos. Chem. Phys. 19, 5165–5186. https://doi.org/10.5194/acp-19-5165-2019.
de Leeuw, F., Horálek, J., 2016. Quantifying the Health Impacts of Ambient Air Pollution: Methodology and Input Data.
Denby, B., Horálek, J., de Smet, P., de Leeuw, F., 2011. Mapping Annual Mean PM2.5 Concentrations in Europe: Application of Pseudo PM2.5 Station Data.
EEA, 2018. Air Quality in Europe - 2018 Report.
ETC/ACM, 2017. Long-term Air Quality Trends in Europe. Fine Particulate Matter (PM2.5) Health Impacts.
Gallego, F.J., 2010. A population density grid of the European Union. Popul. Environ. 31, 460–473. https://doi.org/10.1007/s11111-010-0108-y.
Galmarini, S., Koffi, B., Solazzo, E., Keating, T., Hogrefe, C., Schulz, M., Benedictow, A., Griesfeller, J.J., Janssens-Maenhout, G., Carmichael, G., Fu, J., Dentener, F., 2017. Technical note: coordination and harmonization of the multi-scale, multi-model activities HTAP2, AQMEII3, and MICS-Asia3: simulations, emission inventories, boundary conditions, and model output formats. Atmos. Chem. Phys. 17, 1543–1555. https://doi.org/10.5194/acp-17-1543-2017.
Holland, M., 2014a. Implementation of the HRAPIE Recommendations for European Air Pollution CBA Work. Health Impact Assessment and Cost Benefit Analysis.
Holland, M., 2014b. Cost-benefit Analysis of Final Policy Scenarios for the EU Clean Air Package, Version 2, Corresponding to IIASA TSAP Report 11, Version 1.
Holland, M., Hunt, A., Hurley, F., Navrud, S., Watkiss, P., 2005a. Methodology for the CostBenefit analysis for CAFE: Volume 1: Overview of Methodology', Service Contract for Carrying out Cost-Benefit Analysis of Air Quality Related Issues, in particular in the Clean Air for Europe (CAFE) Programme.
Holland, M., Hurley, F., Hunt, A., Watkiss, P., 2005b. Methodology for the Cost-Benefit analysis for CAFE: Volume 3: Uncertainty in the CAFE CBA: Methods and First Analysis', Service Contract for Carrying out Cost-Benefit Analysis of Air Quality Related Issues, in particular in the Clean Air for Europe (CAFE) Programme.
Holland, M., Watkiss, P., Pye, S., 2005c. Cost-Benefit Analysis of Policy Option Scenarios for the Clean Air for Europe Programme' Service Contract for Carrying out Cost-Benefit Analysis of Air Quality Related Issues, in particular in the Clean Air for Europe (CAFE) Programme.
Holland, M., Wagner, A., Hurley, F., Miller, B., Hunt, A., 2011. Cost Benefit Analysis for the Revision of the National Emission Ceilings Directive: Policy Options for Revisions to the Gothenburg Protocol to the UNECE Convention on Long-Range Transboundary Air Pollution.
Horálek, J., de Smet, P., de Leeuw, F., Kurfürst, P., Benešová, N., 2016. European Air Quality Maps of PM and Ozone for 2014.
Hurley, F., Hunt, A., Cowie, H., Holland, M., Miller, B., Pye, S., Watkiss, P., 2005. Methodology for the cost-benefit analysis for CAFE: volume 2: health impact assessment', Service contract for carrying out cost-benefit analysis of air quality related issues. In: Particular in the Clean Air for Europe (CAFE) Programme.
Im, U., Brandt, J., Geels, C., Hansen, K.M., Christensen, J.H., Andersen, M.S., Solazzo, E., Kioutsioukis, I., Alyuz, U., Balzarini, A., Baro, R., Bellasio, R., Bianconi, R., Bieser, J., Colette, A., Curci, G., Farrow, A., Flemming, J., Fraser, A., Jimenez-Guerrero, P., Kitwiroon, N., Liang, C.-K., Nopmongcol, U., Pirovano, G., Pozzoli, L., Prank, M., Rose, R., Sokhi, R., Tuccella, P., Unal, A., Vivanco, M.G., West, J., Yarwood, G., Hogrefe, C., Galmarini, S., 2018. Assessment and economic valuation of air pollution impacts on human health over Europe and the United States as calculated by a multi-model ensemble in the framework of AQMEII3. Atmos. Chem. Phys. 18, 5967–5989. https://doi.org/10.5194/acp-18-5967-2018.
Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J., Schöpp, W., 2017. Global anthropogenic emissions of particulate matter including black carbon. Atmos. Chem. Phys. 17, 8681–8723. https://doi.org/10.5194/acp-17-8681-2017.
Krapf, M., Künzi, L., Allenbach, S., Bruns, E.A., Gavarini, I., El-Haddad, I., Slowik, J.G., Prévôt, A.S.H., Drinovec, L., Močnik, G., Dümbgen, L., Salathe, M., Baumlin, N., Sioutas, C., Baltensperger, U., Dommen, J., Geiser, M., 2017. Wood combustion particles induce adverse effects to normal and diseased airway epithelia. Environ. Sci. Process. Impacts 19, 538–548. https://doi.org/10.1039/C6EM00586A.
Krupnick, A., Ostro, B., Bull, K., 2005. Peer Review of the Methodology of Cost-Benefit Analysis of the Clean Air for Europe Programme.
Lelieveld, J., Evans, J.S., Fnais, M., Giannadaki, D., Pozzer, A., 2015. The contribution of outdoor air pollution sources to premature mortality on a global scale. Nature 525, 367–371. https://doi.org/10.1038/nature15371.
Liu, J.C., Pereira, G., Uhl, S.A., Bravo, M.A., Bell, M.L., 2015. A systematic review of the physical health impacts from non-occupational exposure to wildfire smoke. Environ. Res. 0, 120–132. https://doi.org/10.1016/j.envres.2014.10.015.
Liu, J.C., Wilson, A., Mickley, L.J., Dominici, F., Ebisu, K., Wang, Y., Sulprizio, M.P., Peng, R.D., Yue, X., Son, J.-Y., Anderson, G.B., Bell, M.L., 2017. Wildfire-specific fine particulate matter and risk of hospital admissions in urban and rural counties. Epidemiology 28, 77–85. https://doi.org/10.1097/EDE.0000000000000556.
Mailler, S., Menut, L., Khvorostyanov, D., Valari, M., Couvidat, F., Siour, G., Turquety, S., Briant, R., Tuccella, P., Bessagnet, B., Colette, A., Létinois, L., Markakis, K., Meleux, F., 2017. CHIMERE-2017: from urban to hemispheric chemistry-transport modeling. Geosci. Model Dev. (GMD) 10, 2397–2423. https://doi.org/10.5194/gmd-10-2397-2017.
Manders, A.M.M., Builtjes, P.J.H., Curier, L., Denier van der Gon, H.A.C., Hendriks, C., Jonkers, S., Kranenburg, R., Kuenen, J.J.P., Segers, A.J., Timmermans, R.M.A., Visschedijk, A.J.H., Wichink Kruit, R.J., van Pul, W.A.J., Sauter, F.J., van der Swaluw, E., Swart, D.P.J., Douros, J., Eskes, H., van Meijgaard, E., van Ulft, B., van Velthoven, P., Banzhaf, S., Mues, A.C., Stern, R., Fu, G., Lu, S., Heemink, A., van Velzen, N., Schaap, M., 2017. Curriculum vitae of the LOTOS–EUROS (v2.0) chemistry transport model. Geosci. Model Dev. (GMD) 10, 4145–4173. https://doi.org/10.5194/gmd-10-4145-2017.
Marécal, V., Peuch, V.-H., Andersson, C., Andersson, S., Arteta, J., Beekmann, M., Benedictow, A., Bergström, R., Bessagnet, B., Cansado, A., Chéroux, F., Colette, A., Coman, A., Curier, R.L., Denier van der Gon, H.A.C., Drouin, A., Elbern, H., Emili, E., Engelen, R.J., Eskes, H.J., Foret, G., Friese, E., Gauss, M., Giannaros, C., Guth, J., Joly, M., Jaumouillé, E., Josse, B., Kadygrov, N., Kaiser, J.W., Krajsek, K., Kuenen, J., Kumar, U., Liora, N., Lopez, E., Malherbe, L., Martinez, I., Melas, D., Meleux, F., Menut, L., Moinat, P., Morales, T., Parmentier, J., Piacentini, A., Plu, M., Poupkou, A., Queguiner, S., Robertson, L., Rouïl, L., Schaap, M., Segers, A., Sofiev, M., Tarasson, L., Thomas, M., Timmermans, R., Valdebenito, Á., van Velthoven, P., van Versendaal, R., Vira, J., Ung, A., 2015. A regional air quality forecasting system over Europe: the MACC-II daily ensemble production. Geosci. Model Dev. (GMD) 8, 2777–2813. https://doi.org/10.5194/gmd-8-2777-2015.
Menut, L., Bessagnet, B., Khvorostyanov, D., Beekmann, M., Blond, N., Colette, A., Coll, I., Curci, G., Foret, G., Hodzic, A., Mailler, S., Meleux, F., Monge, J.-L., Pison, I., Siour, G., Turquety, S., Valari, M., Vautard, R., Vivanco, M.G., 2013. CHIMERE 2013: a model for regional atmospheric composition modelling. Geosci. Model Dev. (GMD) 6, 981–1028. https://doi.org/10.5194/gmd-6-981-2013.
Miller, B., Hurley, F., Shafrir, A., 2011. Health Impact Assessment for the National Emissions Ceiling Directive (NECD) – Methodological Issues.
Pope, C.A., Dockery, D.W., 2006. Health effects of fine particulate air pollution: lines that connect. J. Air Waste Manag. Assoc. 56, 709–742. https://doi.org/10.1080/10473289.2006.10464485.
Saikawa, E., Kim, H., Zhong, M., Avramov, A., Zhao, Y., Janssens-Maenhout, G., Kurokawa, J., Klimont, Z., Wagner, F., Naik, V., Horowitz, L.W., Zhang, Q., 2017. Comparison of emissions inventories of anthropogenic air pollutants and greenhouse gases in China. Atmos. Chem. Phys. 17, 6393–6421. https://doi.org/10.5194/acp-17-

6393-2017.
Sauter, F., van der Swaluw, E., Manders, A.M.M., Wichink Kruit, R.J., Segers, A., Eskes, H., 2012. LOTOS-EUROS v1.8 Reference Guide. TNO, Utrecht, The Netherlands.
Schaap, M., Timmermans, R.M.A., Roemer, M., Boersen, G.A.C., Builtjes, P.J.H., Sauter, F.J., Velders, G.J.M., Beck, J.P., 2008. The LOTOS EUROS model: description, validation and latest developments. Int. J. Environ. Pollut. 32, 270. https://doi.org/10.1504/IJEP.2008.017106.
Schucht, S., Colette, A., Rao, S., Holland, M., Schöpp, W., Kolp, P., Klimont, Z., Bessagnet, B., Szopa, S., Vautard, R., Brignon, J.-M., Rouïl, L., 2015. Moving towards ambitious climate policies: monetised health benefits from improved air quality could offset mitigation costs in Europe. Environ. Sci. Policy 50, 252–269. https://doi.org/10.1016/j.envsci.2015.03.001.
Seinfeld, J.H., Pandis, S.N., 2012. Atmospheric Chemistry and Physics: from Air Pollution to Climate Change. Wiley.
Shaddick, G., Thomas, M.L., Green, A., Brauer, M., van Donkelaar, A., Burnett, R., Chang, H.H., Cohen, A., Dingenen, R.V., Dora, C., Gumy, S., Liu, Y., Martin, R., Waller, L.A., West, J., Zidek, J.V., Prüss-Ustün, A., 2018. Data integration model for air quality: a hierarchical approach to the global estimation of exposures to ambient air pollution. J. R. Stat. Soc. Ser. C Appl. Stat. 67, 231–253. https://doi.org/10.1111/rssc.12227.
Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L.D., Fagerli, H., Flechard, C.R., Hayman, G.D., Gauss, M., Jonson, J.E., Jenkin, M.E., Nyíri, A., Richter, C., Semeena, V.S., Tsyro, S., Tuovinen, J.-P., Valdebenito, Á., Wind, P., 2012. The EMEP MSC-W chemical transport model – technical description. Atmos. Chem. Phys. 12, 7825–7865. https://doi.org/10.5194/acp-12-7825-2012.
Solazzo, E., Riccio, A., Van Dingenen, R., Valentini, L., Galmarini, S., 2018. Evaluation and uncertainty estimation of the impact of air quality modelling on crop yields and premature deaths using a multi-model ensemble. Sci. Total Environ. 633, 1437–1452. https://doi.org/10.1016/j.scitotenv.2018.03.317.
Tørseth, K., Aas, W., Breivik, K., Fjæraa, A.M., Fiebig, M., Hjellbrekke, A.G., Lund Myhre, C., Solberg, S., Yttri, K.E., 2012. Introduction to the european monitoring and evaluation programme (EMEP) and observed atmospheric composition change during 1972–2009. Atmos. Chem. Phys. 12, 5447–5481. https://doi.org/10.5194/acp-12-5447-2012.
van Donkelaar, A., Martin, R.V., Brauer, M., Kahn, R., Levy, R., Verduzco, C., Villeneuve, P.J., 2010. Global estimates of ambient fine particulate matter concentrations from satellite-based aerosol optical depth: development and application. Environ. Health Perspect. 118, 847–855. https://doi.org/10.1289/ehp.0901623.
Volk, H.E., Hertz-Picciotto, I., Delwiche, L., Lurmann, F., McConnell, R., 2011. Residential proximity to freeways and autism in the CHARGE study. Environ. Health Perspect. 119, 873–877. https://doi.org/10.1289/ehp.1002835.
WHO, 2013a. Review of Evidence on Health Aspects of Air Pollution –REVIHAAP – First Results.
WHO, 2013b. Health Risks of Air Pollution in Europe – HRAPIE – Summary of Recommendations for Question D5 on "Identification of Concentration-Response Functions" for Cost-Effectiveness Analysis.
WHO, 2007. Health Relevance of Particulate Matter from Various Sources.
Xing, Y.-F., Xu, Y.-H., Shi, M.-H., Lian, Y.-X., 2016. The impact of PM2.5 on the human respiratory system. J. Thorac. Dis. 8, E69–E74. https://doi.org/10.3978/j.issn.2072-1439.2016.01.19.
Zhang, K., Batterman, S., 2013. Air pollution and health risks due to vehicle traffic. Sci. Total Environ. 0, 307–316. https://doi.org/10.1016/j.scitotenv.2013.01.074.
Zhang, Y., West, J.J., Mathur, R., Xing, J., Hogrefe, C., Roselle, S.J., Bash, J.O., Pleim, J.E., Gan, C.-M., Wong, D.C., 2018. Long-term trends in the $PM_{2.5}$- and O_3-related mortality burdens in the United States under emission reductions from 1990 to 2010. Atmos. Chem. Phys. Discuss. 1–25. https://doi.org/10.5194/acp-2018-498.