



Modelling PM_{2.5} impact indicators in Europe: Health effects and legal compliance



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ABSTRACT

Long-term exposure to fine particulate matter (PM_{2.5}) has been shown to have significant negative impacts on human health. It is estimated that current levels of air pollution shorten the statistical life expectancy of European citizens by several months. The GAINS integrated assessment model calculates shortening of life expectancy from population exposure to PM_{2.5} using epidemiologically-derived health impact functions. In addition, GAINS estimates PM_{2.5} concentrations at 1875 air quality monitoring stations located in diverse environments ranging from remote background locations to busy street canyons. In this article, different approaches to dealing with the PM_{2.5} pollution problem are compared. We assess for the present and future the attainment of EU and WHO air quality standards for PM_{2.5} and estimate the loss of life expectancy under different policy scenarios developed for the ongoing revision of the EU Air Quality Legislation.

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1. Introduction

During recent years, several long-term cohort studies have demonstrated a strong association between human exposure to fine particulate matter below 2.5 μm aerodynamic diameter (PM_{2.5}) and premature death (Dockery et al., 1993; Pope et al., 2002; Henschel and Chan, 2013; Beelen et al., 2014). Long-term exposure to elevated levels of PM_{2.5} leads to increased risk for a variety of diseases, such as cardiovascular disease, ischemic heart disease, and respiratory diseases (Lim et al., 2012). Effects of short-term exposure to ambient PM_{2.5} have also been demonstrated; however, these are not the topic of this study.

Consequently, PM_{2.5} has received increased attention in air quality legislation in recent years, prompting the establishment of guideline and legally binding limit values in many countries as well as the European Union (EU). The World Health Organisation (WHO) states a guideline value of 10 $\mu\text{g}/\text{m}^3$ annual mean concentration that should not be exceeded in order to ensure healthy conditions. The United States Environmental Protection Agency has recently revised its standard to be 12 $\mu\text{g}/\text{m}^3$, while the EU set forth a target value of 25 $\mu\text{g}/\text{m}^3$ that is to become a limit value in 2015 (EU, 2008). Compliance with this limit has to be established at a fixed set of

individual air quality monitoring stations, largely congruent with the stations reported to AirBase, the European Air Quality Database.

The existence of a threshold value of PM_{2.5} exposure that can be considered safe for human health is still under debate, partly because few observations are available at clean environments. For the cohorts analysed in recent assessments, no levelling off of effects could be observed (Crouse et al., 2012; WHO, 2013). For most of the populated places in Europe and worldwide however, this question is of less relevance as their PM concentrations are considerably above such levels. Despite successful efforts to reduce ambient PM_{2.5} concentrations in Europe, concentration levels are well above the WHO guideline value (EEA, 2013). Hot spot areas are found in the Po valley in Northern Italy, parts of Poland, and many of the large cities in both old and new member states of the EU, where the WHO guideline is exceeded by factors of up to five.

PM is both a local and a regional issue, as contributions stem both from long-range transboundary transport of pollution, urban increments in cities, and local traffic increments within street canyons. Important sources of PM emissions include domestic heating, particularly coal and wood burning, road transport with contributions from both exhaust (mostly diesel) as well as non-exhaust emissions from brake and tyre wear, and industrial combustion. However, as PM consists not only of primary particles but also secondary particles formed in the atmosphere from precursor gases, a range of other pollutants and anthropogenic activities from which they are emitted, such as agriculture, contribute strongly to

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the problem. Secondary inorganic aerosol consists of sulphate formed from sulphur dioxide (SO₂) emissions, nitrate formed from reactive nitrogen oxides (NO_x) emissions, and ammonium formed from ammonia (NH₃) emissions. Emissions of non-methane volatile organic compounds (VOCs) are responsible for the formation of secondary organic aerosol (SOA). For an integrated approach to tackle the PM problem, all sources need to be taken into account to design a control strategy.

Several research groups have established integrated assessment modelling tools that have been used to quantify the effects of different policy options on anthropogenic emissions and their impacts on environmental parameters, on a national (Oxley and ApSimon, 2007; Oxley et al., 2013), regional (Amann et al., 2011) or global scale (Van Dingenen et al., 2014). The GAINS integrated assessment model (Amann et al., 2011) provides for the whole of Europe a consistent framework that combines bottom-up emission calculations from projections of anthropogenic activity in all economic sectors and data on control technologies and their emission factors for various pollutants with an impact calculation scheme, providing as output variables (among others) projections on premature mortality from exposure to ambient air pollution, as well as projections of ambient PM_{2.5} concentrations at 1875 air quality monitoring stations reporting to the AirBase European Air Quality Database. Hence, GAINS provides estimates of two quantities relevant to policy: The actual target of air quality legislation is the reduction of health and environmental damage,¹ which cannot be measured directly, while limit values pertain to measured concentrations at individual stations which are supposed to be representative of their surroundings.

Presently, the EU is undertaking a revision of the existing air quality legislation, with the aim to establish new national emission reduction commitments for the year 2030. The GAINS model has been extensively employed in this policy process to provide an analysis of the expected evolution of emissions and their reduction potentials, and to quantify the projected impacts of different policy options.

This paper explains the methodology used in GAINS and provides for the first time projections of station-based PM_{2.5} concentrations as well as projections of calculated health benefits for different scenarios in the year 2030. We analyse a baseline scenario assuming only implementation of currently agreed legislation, a mitigation scenario investigating the potential for reductions if all available control technologies are employed to their maximum feasible extent, and a policy scenario corresponding to the EU Commission proposal for a new Emission Ceilings Directive for 2030 as published December 2013 (European Commission, 2013). We investigate the two different indicators and quantify their relationship, which may become relevant if the currently rather high limit value is to be revised in the future.

This paper is structured as follows. Section 2 reiterates the GAINS methodology for calculating premature mortality, as well as PM_{2.5} concentrations for individual stations. Section 3 provides a validation of the modelled station-by-station concentrations. Section 4 presents results in terms of station concentrations and premature mortality for 2009 and projections for the year 2030 following the different scenarios. A simple relation between the two indicators is introduced and discussed. Conclusions are presented in Section 5.

2. Methodology

In the following sections, the calculation of ambient concentrations in GAINS is explained. We start by explaining the calculation of ambient PM_{2.5} concentrations on the 7 × 7 km² grid used in GAINS (Sect. 2.1). These gridded concentrations are used directly in the health impact calculation, as detailed in Sect. 2.2. Going beyond

the grid level, Sect. 2.3 explains how the calculated gridded concentrations are used together with past monitoring data to derive concentration estimates for individual monitoring stations reporting to the Airbase network of observations.

2.1. Calculating exposure to ambient PM_{2.5}

Anthropogenic exposure to PM_{2.5} is usually analysed at urban background concentration levels. Ambient concentrations vary considerably within cities (Cyrus et al., 2012), and in particular local traffic emissions lead to considerably higher concentrations within urban street canyons. Roadside increments thus need to be considered when assessing compliance at individual stations (as described in Sect. 2.3), however, they are not taken into account when looking at long-term average population exposure.

In the GAINS integrated assessment model, PM_{2.5} concentrations at the urban background level are calculated as the sum of regional background contributions (primary and secondary) from Europe-wide emissions, and an urban increment related to primary PM emissions from local low-level sources. Calculation of the regional background relies on linearised source-receptor simulations performed with the EMEP Chemistry Transport Model (EMEP CTM, Simpson et al., 2012) using five different meteorological years with a resolution of 0.5° × 0.25° (roughly 28 × 28 km²). The urban increment is derived from a full year simulation of the CHIMERE CTM (Menut et al., 2013) run at a resolution of 0.125° × 0.0625° or roughly 7 × 7 km². Both EMEP and CHIMERE models used the same emission gridding by country and SNAP sector on the 7 × 7 km² grid, as described by Kieseewetter et al. (2014, 2015).

Ambient concentrations at the 28 × 28 km² grid level are related to emissions of primary PM and PM precursors (NH₃, NO_x, SO₂, and VOC) in 53 European source regions (EU-28, neighbouring countries, and 10 sea regions) as

$$[PM_{2.5}]_{28}(\mathbf{i}) = \delta(\mathbf{i}) + \sum_{r=1}^{53} \sum_p \pi(r, p, \mathbf{i}) E(r, p) \quad (1)$$

with $E(r, p)$ the annual total emissions of pollutant p in region r in a given year, π the PM_{2.5} transfer coefficient for pollutant p from region r to receptor grid cell \mathbf{i} , and $\delta(\mathbf{i})$ a constant from the source-receptor calculations accounting for boundary conditions and non-linearities in the atmospheric chemistry and dispersion.

The linear approach does not take into account the cross-dependencies between different precursors for secondary inorganic aerosol formation; in particular, it does not explicitly calculate an equilibrium state between ammonium sulphate and ammonium nitrate formation but assumes that the modelled effects of reducing one pollutant by 15% can be extrapolated linearly. It is clear that this approach has its limitations, in particular if emission changes are unbalanced between different precursors. Modelled concentrations are realistic as long as changes in precursor gases are similar.

The 0.5° × 0.25° resolution of the atmospheric dispersion calculation is not sufficient to reproduce measured urban background PM concentrations. Starting from the modelled fields at 0.5° × 0.25°, a downscaling scheme is applied which increases the resolution by a factor of 16, essentially based on a redistribution of primary PM concentrations according to the primary PM emission densities from low-level sources (transport and domestic sectors), using a full year simulation of the CHIMERE CTM with the same emission gridding as used in the transfer coefficient calculations. A detailed description is given by Kieseewetter et al. (2015).

In a further step, urban polygon shapes have been developed for most European cities with more than 100000 inhabitants, as described by Kieseewetter et al. (2014). These polygons are used to distinguish between urban and rural parts of the 0.125° × 0.0625° grid cells, leading to a further enhancement of inner urban PM_{2.5} concentrations and thus overall population exposure.

2.2. Health impact calculations

Several cohort studies have investigated the relation between PM_{2.5} exposure and mortality and derived values for Relative Risk RR_{PM}. Here we use results from the HRAPIE (Henschel and Chan, 2013) and REVIHAAP (WHO, 2013) meta-analyses, stating that the risk of natural death from all causes is increased by 6.2% per 10 µg/m³ PM_{2.5} (95% confidence interval 4.1–8.4%; Hoek et al., 2013). This is in close agreement with earlier results from the American Cancer Society cohort study (Pope et al., 2002, 2009a,b) which gave an estimate of 6% per 10 µg/m³ PM_{2.5} (95% CI 2–11%), while the more recent ESCAPE analysis (Beelen et al., 2014) reported significantly higher relative risks (13% per 10 µg/m³ PM_{2.5}, 95% CI 1–25%).

The recent Global Burden of Disease assessment also quantified non-linear relative risk functions for death from individual diseases such as ischemic heart disease (Burnett et al., 2014; Lim et al., 2012). The exact shapes of the dose response curves have been the subject of scientific discussion, particularly relating to departures from linearity and possible lower threshold doses. So far no clear evidence of a safe level of exposure or a threshold below which no adverse health effects occur could be identified (WHO, 2013). Burnett et al. (2014) use in their model a counterfactual low level of PM_{2.5} between 5.8 and 8.8 µg/m³, below which no health effects are assumed; however, they maintain that this level is mainly used as a technical quantity in their model representing the lowest exposure reached

¹ Note, e.g., the formulation of the EU Thematic Strategy on Air Pollution, which states the objective to attain “levels of air quality that do not give rise to significant negative impacts on, and risks to human health and the environment”.

anywhere, and may be adapted if further evidence for health effects below this point is found.

There has been a scientific debate about possible differences in toxicity of different components of PM. Despite targeted research efforts, however, no clear evidence has so far been found for different impacts of different components of PM (WHO, 2013), and thus we assume a proportionality of premature mortality to total PM_{2.5} mass concentration. Furthermore we do not distinguish individual diseases here and rather focus on all-cause mortality, as an analysis of WHO statistics on causes of death revealed important inconsistencies in reporting standards across countries, which would introduce an artificial and systematic bias in health impact assessments between countries.

Mechler et al. (2002) have introduced the methodology used in GAINS. Loss of life expectancy is calculated from exposure to ambient PM_{2.5} using a Cox proportional hazards model (Cox, 1972). A key assumption in all calculations presented here is that the cohort exposure is kept constant for the whole lifetime. We specify that the age-specific risk of dying for adults is related to the level of PM_{2.5} as follows:

$$RR_{PM} = \exp(\beta [PM]) \cong 1 + \beta [PM] \quad (2)$$

using a first-order Taylor expansion, which is justified because β is small. We take into account the fact that exposure to air pollution affects only natural mortality, while accidents and suicide (about 5% of deaths) are unchanged. For the whole population the coefficient β in Eq. (2) thus becomes 0.00588 per 1 $\mu\text{g}/\text{m}^3$ PM_{2.5}. Cohort- and country-specific mortality data extracted from life table statistics (United Nations, 2011) are used to calculate for each cohort the baseline survival function over time. The survival function $l_c(t)$ indicates the percentage of a cohort c alive after time t elapsed since starting time w_0 . $l_c(t)$ is an exponential function of the sum of the mortality rates $\mu_{a,b}$, which are derived for each country from life tables with a as age and b as calendar time. Accordingly, for a cohort aged c in the starting year 2010, $l_c(t)$ is

$$l_c(t) = \exp\left(-\sum_{z=c}^t \mu_{z,z-c+w_0}\right) \quad (3)$$

The baseline survival function $l_c(t)$ is modified by the exposure to PM_{2.5} to

$$\bar{l}_c(t) = \exp\left(-\left(1 + \beta [PM]\right) \sum_{z=c}^t \mu_{z,z-c+w_0}\right). \quad (4)$$

Consistent with the methodology introduced by Pope et al. (2002), increased risk applies only to people older than $w_0 = 30$ years, and younger cohorts are only included when they reach the age of 30. For a given exposure to PM_{2.5} (PM), life expectancy e_c is calculated as the integral over the remaining life time:

$$e_c = \int_c^{w_1} \bar{l}_c(t) dt = \int_c^{w_1} \exp\left(-\left(1 + \beta [PM]\right) \sum_{z=c}^t \mu_{z,z-c+w_0}\right) dt \quad (5)$$

where w_1 is the maximum age considered (100 years) and RR_{PM} the relative risk for a given concentration of PM_{2.5}. With some simplifying assumptions and approximations (Vaupel and Yashin, 1986), the change in life expectancy per person (Δe_c) in a cohort c due to PM_{2.5} can be expressed as:

$$\Delta e_c \cong \beta [PM] \int_c^{w_1} l_c(t) \log l_c(t) dt \quad (6)$$

where l_c is the baseline survival function.

For all cohorts in a country r the change in life expectancy Δe_r is then calculated as the averages of the change in life expectancy for the cohorts living in the 7×7 km² grid cells i of the country (region) r :

$$\Delta e_r = \sum_{c=w_0}^{w_1} \frac{Pop_{c,r}}{Pop_i} \Delta e_{c,i} = \beta \sum_{i \in r} [PM]_i \frac{1}{Pop_r} \sum_{c=w_0}^{w_1} Pop_{c,r} \int_c^{w_1} l_{c,r}(t) \log l_{c,r}(t) dt \quad (7)$$

where

- $\Delta e_{c,r}$ Change in life expectancy for cohort c in country r
- $Pop_{c,r}$ Population in cohort c in country r
- Pop_i Total population in grid cell i in country r
- Pop_r Total population in country r .

PM concentration levels correspond to the bottom up calculated urban background levels on the 7×7 km² grid, as described in Sect. 2.1. As a simplification, national average population numbers and life tables supplied by the United Nations (UN, 2011) are applied to all grid cells in a given country, assuming that the age structure is the same across the whole country. Population density data corresponding to the year 2000 have been obtained from the Joint Research Centre of the European Commission (JRC-IES) on a $0.01^\circ \times 0.01^\circ$ grid (Gallego, 2010) and re-gridded to the 7×7 km² grid used here. For grid cells containing parts of cities

with >100000 inhabitants for which urban polygons have been defined (see Kiesewetter et al., 2014), the total grid population is split into urban and rural population, and exposure is calculated separately for both parts of the grid cell. The spatial distribution of population is left constant in time.

GAINS calculations of air pollution related mortality include only bottom-up modelled concentrations of anthropogenic PM. It is presently not clear whether particles from natural sources such as sea salt and wind-blown dust affect human health. Furthermore, the focus of GAINS is not so much on the absolute quantification of effects but rather on comparison of emission control scenarios. Since no mitigation strategies exist for natural particles, any potential effects of these would cancel out between two different air pollution control scenarios. The explicit exclusion of natural background aerosol shifts the origin of the assumed dose–response curve, thereby implying a threshold level as high as the natural background aerosol.

The health impact is calculated for the total population of the year 2010; cohorts are followed until the year 2110. As the cohort studies that determined the relative risk used here included only participants over 30 years of age, also GAINS calculates health effects starting at this age. People who are currently below 30 years are subject to the increased risk only after crossing this threshold.

2.3. Station-based concentration calculation

GAINS calculates concentrations of ambient PM_{2.5} for 1875 individual monitoring stations reporting to AirBase,² the European air quality database. The methodology is described in detail in Kiesewetter et al. (2015) and will only be repeated briefly here.

Similar to the approach laid out by Kiesewetter et al. (2014) for NO_x and NO₂, the station-based modelling scheme combines past monitoring data with bottom-up emission modelling and a simplified atmospheric chemistry and dispersion calculation. The starting point of all calculations is monitoring data reported to AirBase in 2009. For each monitoring station with sufficient data coverage, contributions from the ambient background and local road traffic emissions are identified, and then each of these contributions is modelled individually. The bottom-up modelling scheme laid out in Section 2.1 is used to explain as far as possible the observed background concentrations. Residuals between bottom-up modelled and measured background concentrations are attributed to their likely sources of origin and then scaled with the respective trends in the projections for the future, so that the station-by-station calculation scheme is constrained to observed values in 2009. The attribution process is detailed in the Technical Appendix.

The availability and spatial coverage of PM_{2.5} monitoring stations in the EU is much less than in the case of PM₁₀ or NO₂. For the station calculations, GAINS relies on monitoring data for 2009, the same meteorological year for which the CHIMERE atmospheric model calculations were performed. The network of stations is still being built up, and while some regions already had a dense coverage, others – particularly some of the most affected regions in the new Member States – did not have a sufficient number of monitoring stations operational in 2009. In order not to leave out these areas, GAINS extends the PM_{2.5} calculations to all PM₁₀ stations covered by the model, effectively estimating a measured PM_{2.5} concentration from the modelled concentration and the unexplained residual in PM₁₀. This residual is allocated to the fine and coarse fractions in the same ratio as the country average for those stations where both PM_{2.5} and PM₁₀ are available.

Roadside stations assume an additional increment on top of urban background levels, related to road traffic emissions within the immediate surroundings of the monitoring station. Instead of bottom-up estimates of site-specific emission densities and dispersion, the roadside increment observed in the base year is scaled by the national trend in primary PM_{2.5} road traffic emissions for urban driving conditions (exhaust and non-exhaust). We take into account that the fleet composition and average driving speeds differ from the national average in the typical urban environment but do not distinguish trends at individual stations. This simplification assumes that the national average fleet composition and driving regime that is used for calculation of national urban road transport emissions would be representative of any given roadside station.

Due to the mentioned lack of PM_{2.5} station data, identification of the base year roadside increment is not straightforward, as there are only few station pairs of a roadside and a background station within a meaningful distance. For locations where roadside PM_{2.5} observations are available but the corresponding background station supplies only PM₁₀, PM_{2.5} concentrations at the background site are estimated as described, leading to a well-defined PM_{2.5} roadside increment. To estimate roadside PM_{2.5} concentrations where either the roadside or both roadside and background stations supply only PM₁₀, we use a simple scaling model to estimate the roadside increment of PM_{2.5} based on the following assumptions:

² AirBase, the European air quality database; online at <http://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-8>.

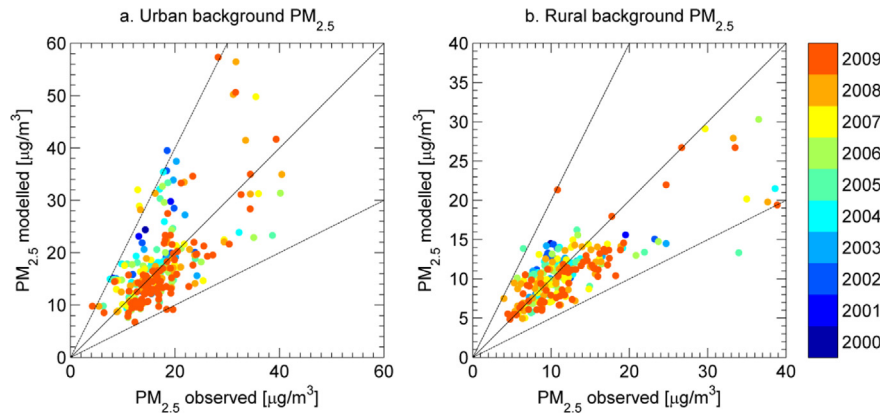


Fig. 1. Bottom-up modelled versus observed concentrations of $PM_{2.5}$ at urban (a) and rural (b) background stations. Modelled fields of natural $PM_{2.5}$ have been added to the anthropogenic concentrations. Modelled concentrations correspond to the levels inside urban polygons in panel a, and the 7×7 km concentrations in panel b.

- 1) The roadside increment consists entirely of primary particles from road traffic (exhaust and non-exhaust); secondary aerosol formation can be ignored at the time scales involved in the exchange with background air masses.
- 2) $PM_{2.5}$ disperses like a chemically inert gas. If roadside $PM_{2.5}$ observations are missing, the base year roadside increment can thus be derived from the proportionality to the observed increment in nitrogen oxides (NO_x) at the same station, measurements of which are widely available. The $PM_{2.5}$ roadside increment is then estimated as

$$\Delta[PM_{2.5}] = \frac{E_{PM_{2.5}}}{E_{NO_x}} \Delta[NO_x] \quad (8)$$

where $\Delta[\cdot]$ denotes the concentration increment from urban background to roadside levels for a pollutant, and E_x represent the national total emissions of pollutant x from road traffic in urban areas.

Assumption 1 is frequently taken, as the mixing time between roadside and urban background air masses is in the order of seconds to minutes, while secondary aerosol formation takes place on the order of hours (Seinfeld and Pandis, 1997). Assumption 2 is needed because roadside observations of $PM_{2.5}$ are still scarce, even though the monitoring network has increased significantly in recent years. Restricting the analysis to stations operational in 2009 would result in an uneven distribution across Europe and exclude some of the worst affected areas e.g. in the new EU Member States. Using this scaling ratio allows us to extend the analysis to all roadside stations which monitor NO_x and PM_{10} , and have NO_x and PM_{10} background observations available.

The temporal evolution of the roadside increment is assumed to be proportional to the national trend in road traffic emissions of $PM_{2.5}$. This implies that the fleet mix and driving conditions at any station are well represented by the national average, that temporal changes in traffic intensities follow the national trends and do not fluctuate significantly from year to year, and that the atmospheric mixing conditions in a given scenario year do not deviate significantly from those in the year 2009.

3. Validation

A wide range of criteria for model validation and performance evaluation is available (Bennett et al., 2013; Thunis et al., 2012). As noted by Bennett et al. (2013), performance evaluation needs to be tailored to the purpose of the model. In our case, there are different aspects of the model which can be validated, mostly pertaining to the atmospheric part of the calculations.

Firstly, simplifications are taken regarding the linear approximations involved in both the atmospheric dispersion and the health impact calculations, which need to be validated against the full non-linear formulation in each case. The linearised methodology for atmospheric chemistry-dispersion relations used in GAINS has been shown to be in close agreement with corresponding simulations of the full EMEP atmospheric model (Amann et al., 2011). Mechler et al. (2002) have compared the linearised approach to the life expectancy calculations to a full integration and

found close agreement. These linearization aspects are therefore left out of the validation shown here.

Secondly, we need to ensure that modelled ambient concentrations of $PM_{2.5}$ match real world observations sufficiently well to make the modelling credible. In this regard, it is important to note the differences between the station-by-station calculation and the gridded background calculation. The 7×7 km fields of anthropogenic $PM_{2.5}$ which are derived directly from bottom-up emission calculations, linearised chemistry-dispersion calculations at 28×28 km, and the downscaling step to 7×7 km/urban polygons, are used for the health impact calculation in GAINS. The station-by-station calculation, which is relevant for compliance with the limit values, is based on the 7×7 km/urban concentration fields but is constrained by the observed values at the station in the year 2009.

We focus here on the evaluation of model performance for background stations. With a perfect model and perfect observations, the observed concentration at a background monitoring site (be it urban or rural) would be expected to match exactly the 7×7 km/urban bottom-up modelled concentrations plus natural PM.

Two indicators of model performance are analysed: the scatter between observations and modelled concentrations, expressed by the Pearson correlation coefficient R , and the mean bias, calculated as arithmetic mean of observations minus model. Fig. 1 shows scatter plots for annual average $PM_{2.5}$ concentrations at urban background (a) and rural background (b) locations for the years 2000–2009 (colour coded). Station comparisons show some scatter ($R = 0.76$ for urban background stations, $R = 0.83$ for rural background stations).

Comparing absolute concentrations, overall agreement is good, and there is hardly any mean bias present (model underestimating observations by less than $1 \mu\text{g}/\text{m}^3$ on a Europe wide average) between observed and modelled values for these station categories, as may be seen in Fig. 1, which compiles annual average observations from all years and all $PM_{2.5}$ stations covered by the model, colour-coded by the year of observation. Only stations located inside a defined urban polygon are included in Fig. 1a. Concentrations at urban background stations located in smaller cities are somewhat underestimated by the 7×7 km fields, so that the overall set of modelled stations shows a mean negative bias of $2.5 \mu\text{g}/\text{m}^3$ to observations.

The biases for individual stations show an apparent lack of reliable local information on emissions in some areas. Notably, inner urban concentrations are overestimated by the model in parts of Poland and Northern Italy, while smaller settlements are often underestimated, pointing to possible issues with the spatial distribution of household emissions between cities and the

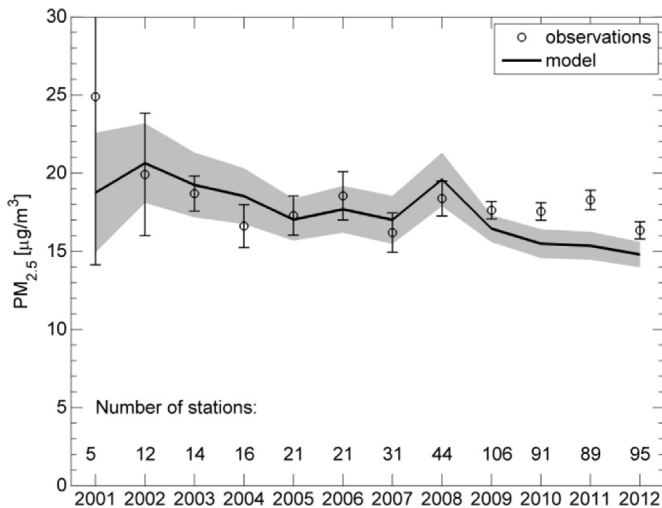


Fig. 2. Europe wide annual average $PM_{2.5}$ concentrations at urban background stations in cities with more than 100000 inhabitants (same set as Fig. 1a), comparing observations to the bottom-up modelled concentrations for multiple years. All such urban background $PM_{2.5}$ stations operational in each year and covered by GAINS are included; numbers of stations are given for each year. Standard deviations of the mean are shown as error bars for the observations and as shaded area for the modelled concentrations. Note that concentrations from 2010 onwards are based on modelled emission numbers which have not been validated.

countryside. For health impact calculations, which are based entirely on the bottom up modelled concentration fields shown in Fig. 1, local biases for individual sites are of limited effect as long as country wide impacts are analysed. For the purpose of modelling compliance with individual stations, a calibration to monitoring data to incorporate local conditions is necessary to derive the best possible concentration estimate. In this calibration scheme, offsets between modelled and observed background values are corrected by either a best guess of the sectoral split of the unexplained part, or a scaling of the modelled concentrations if they exceed observations. The estimation process is detailed in the [Technical Appendix](#).

A second criterion to ask from our model is whether the time evolution of modelled concentrations is able to capture observed trends in the past, particularly as meteorological conditions vary from year to year in reality while they are fixed in the model. A practical challenge in this regard is that measurements of $PM_{2.5}$ at the beginning of the 21st century are rather sparse and the number of stations has increased rapidly over the years. Therefore, deriving a robust Europe wide trend is not possible as the amount of stations with a sufficient time span available would be too small. Nonetheless, we may analyse whether the model shows systematic deviations over time or not. Fig. 2 shows bottom-up modelled $PM_{2.5}$ concentrations for the mean of all background stations in major urban areas with more than 100000 inhabitants for the years 2001–2012. The figure includes all stations operational in any given year and covered by GAINS. Thus we do not limit the ensemble of stations considered here to those covering a certain number of years, which would effectively remove many stations from the comparison in later years. Fig. 2 is not suited to derive a Europe wide trend. Lines are shown as guides to the eye and solely for the purpose of validating the model against observations for multiple years. The spreads (standard deviations of the mean) of observed and modelled concentrations are shown as shaded area for modelled values and as error bars for observations. Note that we limit the set of stations to those operational in 2009. Agreement between model and observations is excellent for the years 2001–2009. Year to year variations in concentrations are related to

the changing set of stations as well as inter-annual meteorological variability, of which the latter is only reflected in observations. In the years after 2009 model predictions seem too low; however, concentrations after 2010 are derived from emissions which are interpolated linearly between 2010 and estimated emissions for 2015, and effects of the financial crisis are not yet accounted for. An update to validated emission inventories for 2015 is needed before performance after 2010 can be assessed.

4. Results

For estimating current and future health impacts, we employ the emission scenarios used by the European Commission for the proposal for a new directive on national emission ceilings in December 2013 (EU Commission, 2013). They have been described by Amann et al. (2014a).

The assumed evolution of macroeconomic drivers (population, GDP growth, energy use, agricultural activities) is taken from external sources, in this case from projections of the PRIMES model (E3MLab, 2013). Emission scenarios are then created in GAINS by assuming application of different emission control measures in the various countries and economic sectors.

Three emission scenarios are analysed here. The starting point is a scenario assuming successful implementation of currently agreed emission control legislation (“current legislation”, CLE) but no further measures beyond it. Starting from this baseline, we discuss consequences of additional emission control measures in the EU (outside the EU, emissions are assumed to remain at baseline levels). Full implementation of best available technology leads to a “maximum technically feasible reduction” (MTFR) scenario, which represents the lowest level of emissions achievable with all currently available technologies represented in the GAINS model, but excluding fuel switches or behavioural changes. Increasingly tight emission controls, however, come at increasing marginal costs for implementing them. Realistic scenarios thus aim for a partial exploitation of the full potential. GAINS includes an optimization module (Wagner et al., 2013) which identifies the most cost-effective strategy to reach a given policy target within the feasible range of emissions and their impacts. The third scenario analysed here is a cost optimal solution corresponding to the proposal made by the EU Commission for further emission reductions (“Clean Air Policy Package”, CLAPP). In the design of the CLAPP, the policy target was set to a 67% ambition level on reducing premature mortality from exposure to ambient $PM_{2.5}$ in 2030 (expressed in number of life years lost in the EU), with 0% corresponding to current legislation and 100% corresponding to the MTFR case.

As the mitigation scenarios used here do not consider any non-technical measures, care needs to be taken in interpreting them. Some of the measures contained in the MTFR scenario have very high marginal costs; in practice, emission reductions may be achieved at lower costs if behavioural changes or fuel switches are allowed. CLAPP and MTFR scenarios are based on the same anthropogenic activity pathways (particularly the same fuel use) as the CLE baseline (PRIMES-2013), which was generated before the current EU climate targets were formulated. Hence these scenarios do not yet take account of the proposed targets (40% CO_2 reduction by 2030, 27% penetration of renewable energy). A study conducted for the European Parliament (Amann et al., 2014b) highlighted that synergies in climate and air pollution control policies would lead to higher overall potentials for emission reductions and a correspondingly lower MTFR scenario.

Using the CLE, CLAPP, and MTFR emission scenarios, we quantify in the following sections the ambient $PM_{2.5}$ concentrations and resulting loss of life expectancy both in the recent past and for 2030, the target year for the revision of the EU air quality legislation.

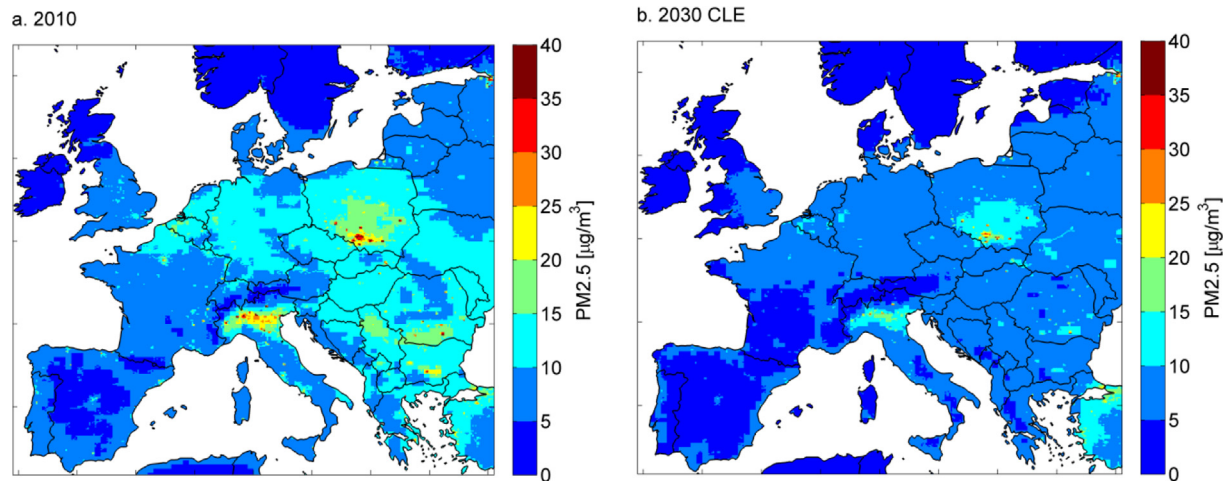


Fig. 3. Ambient $PM_{2.5}$ concentrations from anthropogenic emissions as modelled in GAINS at the 7×7 km grid level (urban background), for two different scenario years, assuming current legislation (CLE).

4.1. Future ambient $PM_{2.5}$ concentrations

Fig. 3 shows bottom-up modelled anthropogenic $PM_{2.5}$ concentrations at the 7×7 km grid level, for the year 2010 (a) and the year 2030 under CLE assumptions (b). Individual cities can easily be identified as concentration hot spots above the regional background, particularly in Northern Italy, Poland, Romania and Bulgaria. The localized nature of these hot spots clearly indicates the necessity of the downscaling step to the 7×7 km/urban background level. Due to the limited resolution of the image, a further distinction between concentrations inside and outside urban polygons is not made for this figure. The validation against observations demonstrates the need to take into account this step wherever possible, as $PM_{2.5}$ levels in several medium-sized or smaller European cities are underestimated in the 7×7 km fields, particularly if cities are divided between neighbouring grids.

While regional background $PM_{2.5}$ concentrations range between 5 and $15 \mu\text{g}/\text{m}^3$ in most parts of Europe, at hot spot areas modelled anthropogenic concentrations exceed $25 \mu\text{g}/\text{m}^3$ (Fig. 3a: the Po valley in northern Italy, south-west Poland, and parts of Romania and Bulgaria). Modelled concentrations are more reliable for EU countries due to the higher availability of information on sectoral emission data there (gridded and national totals). It is interesting to note that the broad spatial pattern of $PM_{2.5}$ concentrations in 2010 estimated using the methodology described here, and the identified hot-spot areas, are rather similar to the 2010 $PM_{2.5}$ concentration map generated by Horálek et al. (2013). The method used by these authors starts from the 2010 $PM_{2.5}$ measurements from Airbase, supplemented by “pseudo $PM_{2.5}$ data” based on measured PM_{10} , and mapped by means of a kriging approach using additional data such as EMEP model output, altitude, wind speed, surface solar radiation and population density.

Implementation of existing legislation is expected to decrease regional background PM and at urban hotspots (Fig. 3b). However, care is needed when interpreting the concentration fields for 2030, as elevated concentrations are still present in several urban areas (Po valley, southern Poland) but are not easily visible due to the limited resolution of the image.

4.2. Scenario analysis: loss of life expectancy

We calculate total loss of life expectancy for the reference year 2010 (in comparison to a hypothetical perfectly clean world) and then compare the relative benefits of different emission control

scenarios in 2030 to this starting point. By analysing differences, some of the uncertainties mentioned above (existence of a threshold value, impacts of natural aerosols) are eliminated and the relative benefits of different emission control scenarios are subject to less uncertainty than the absolute loss of life expectancy for a given scenario and year. To facilitate comparison of different scenarios, the same population structure is used in all cases, so that the population of 2010 is exposed to modelled concentrations and then followed to the age of 100 using the same baseline mortality rates which are modified by ambient PM following Eq. (7), assuming constant ambient PM for the whole lifetime.

As described in Sect. 2.2., the statistical loss of life expectancy is calculated from the 7×7 km modelled fields of anthropogenic $PM_{2.5}$, with a further distinction of urban and rural parts of the grids for cities >100000 inhabitants. The GAINS estimate for the absolute loss of life expectancy attributable to ambient $PM_{2.5}$ levels as modelled for 2010 is shown in Fig. 4a. Estimated values range between 1 and 2 months in clean background conditions (Sweden, Scotland) to 1.5 years and more in the Po valley, Poland, parts of Romania and Bulgaria. On an EU average, around 60% of the calculated exposure and related loss of life expectancy is from secondary aerosols and 40% from primary PM.

As discussed in Section 2, the absolute loss of life expectancy is subject to uncertainty due to questions of lower threshold and effects of natural aerosols. The focus of our health impact calculation is thus on the comparison of different scenarios or scenario years. While the absolute loss of life expectancy would vary if a lower threshold were introduced (on the EU average by about 0.6 months per $\mu\text{g}/\text{m}^3$), results for differences between scenarios are independent of threshold assumptions as long as no significant proportion of the population lives below threshold levels. For ambient concentrations resulting from the emission scenarios used here this is not the case for threshold levels up to $7.5 \mu\text{g}/\text{m}^3$ as used in the Global Burden of Disease study (Lim et al., 2012) provided that in the case of a threshold also natural sources should be included.

Fig. 4b shows the population average change in life expectancy that is obtained from switching constant 2010 conditions to constant levels of $PM_{2.5}$ as in the CLE case for 2030. Significant gains in life expectancy are calculated mainly in the new member states and the Po valley, with values ranging between less than two months gain in most of Western Europe and around six months in hot spots.

For the EU-28, the CLE scenario projects an increase of 2.3 months of life expectancy, with the values for individual countries

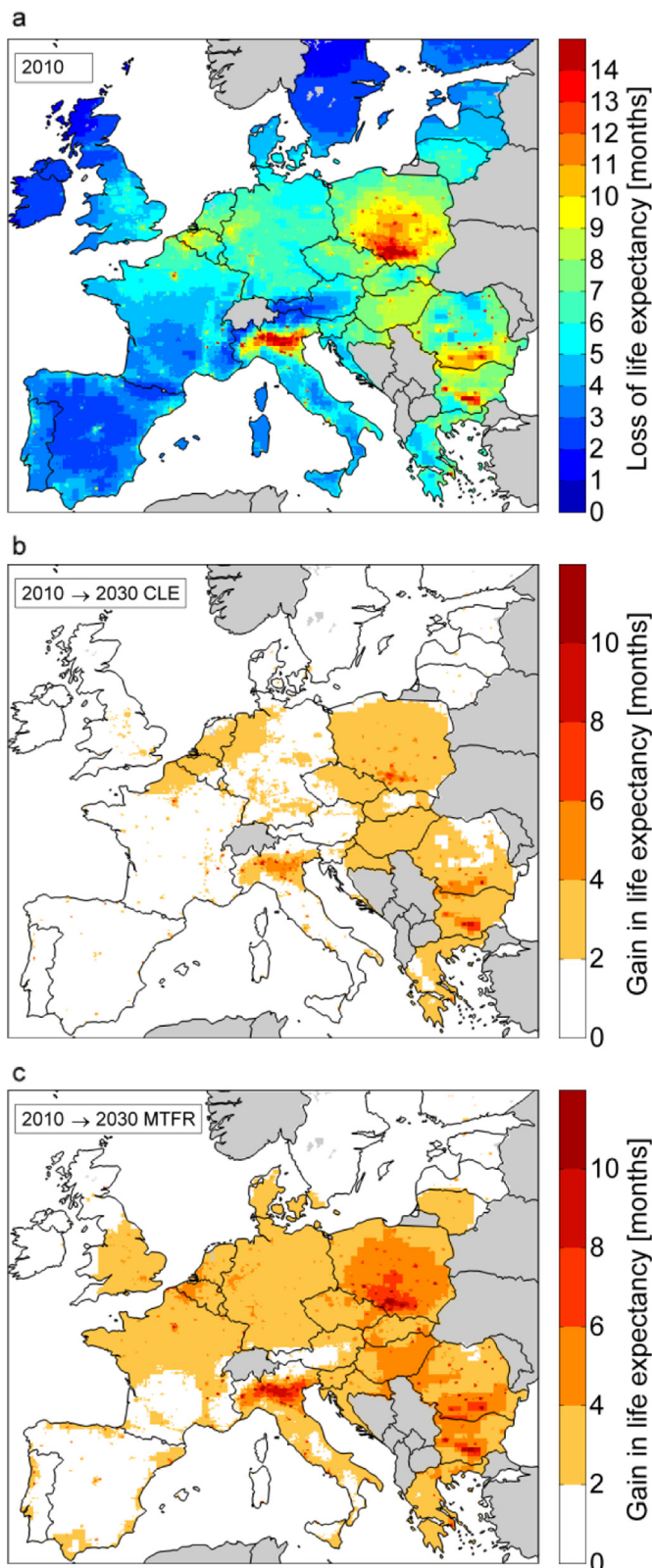


Fig. 4. Shortening of life expectancy assuming constant lifelong exposure to anthropogenic $PM_{2.5}$ levels as in 2010 (a), and relative to this the gains in life expectancy attained for 2030 levels under current legislation (b), and for the maximum technically feasible emission reduction scenario (c).

ranging between 0.7 (Ireland and Sweden) and 4.5 months (Bulgaria). In absolute terms, however, in Belgium, Poland, the Czech Republic, Hungary and Romania people would still lose more than six months on average even in 2030.

Maximal implementation of the currently available emission controls as foreseen in the MTFR scenario (Fig. 4c) would lead to significantly higher gains in life expectancy, particularly in the most polluted regions where up to a year could be gained. As in the CLE case, improvements would be highest in the new Member States, with country average gains in life expectancy amounting to 5.5 months in Romania, 5.9 months in Bulgaria, and 6.4 months in Poland. Thereby, all countries except Cyprus (which is under strong transboundary influence from non-EU countries) would recover between 40 and 60% of the life expectancy lost in 2010; also in relative terms, the more polluted regions would benefit the most.

4.3. Scenario analysis: compliance with EU limit value and WHO guideline

Following the declines in emissions of PM and its precursors, the station-specific model projects decreases in $PM_{2.5}$ concentrations at all individual stations, although of different magnitude depending on the different sectoral-spatial contributions to the local concentration.

Fig. 5 shows modelled concentrations of $PM_{2.5}$ at all PM_{10} stations for the year 2030, as expected under the CLE scenario. Fig. 5 displays all modelled PM stations which report either PM_{10} or $PM_{2.5}$ in 2009. If only genuine $PM_{2.5}$ stations are considered, densities would become very low in some regions in the New Member States. Understandably, the picture is similar to the pure bottom-up modelled 7×7 km fields in Fig. 3a. Many urban areas show concentrations around $20 \mu\text{g}/\text{m}^3$, clearly above the WHO guideline value, while the current EU limit value is attained at most stations with exceptions in Poland and the Slovak Republic. There are large differences in the densities of monitoring station distribution, pointing to an important issue with the assessment of compliance based on single stations. To be representative for the average pollution levels citizens are exposed to, a certain density as well as suitable positioning of monitoring stations is required.

Cumulative distributions of calculated $PM_{2.5}$ concentrations for all stations are shown in Fig. 6. Consistent with European legislation, natural dust and sea salt (amounts as calculated from the EMEP CTM) are not included in this graph as they may be subtracted from measured concentration when determining compliance with a given limit value. While the large majority of stations (>90%) is already in compliance with the $PM_{2.5}$ limit value of $25 \mu\text{g}/\text{m}^3$ in 2009, only 12% of stations, mainly located in remote background locations, have met the WHO guideline value of $10 \mu\text{g}/\text{m}^3$ in this year. In the year 2030, this fraction is projected to increase significantly, with around 41% of stations attaining the WHO guideline and close to all (>98%) stations complying with the EU limit value. Thus, even without further political action, the current limit value is assumed to be widely attained, but at the same time it does not stimulate further emission controls that would be needed to attain the WHO guideline.

Implementation of additional emission control measures would improve this situation considerably. Under the MTFR scenario, concentrations would decrease below $10 \mu\text{g}/\text{m}^3$ at around 74% of stations, while under the CLAPP policy scenario only 63% of the stations would reach this value. The US EPA standard of $12 \mu\text{g}/\text{m}^3$, for comparison, could be attained at 89% of stations provided a full implementation of emission controls (MTFR), and at 83% of the stations under the CLAPP scenario.

It should be emphasized that, due to the steep increase of the cumulative distribution curves for the year 2030 around the WHO

guideline value, percentage numbers stated here are subject to large uncertainty and should only be taken as indicative values. The exact levels of natural PM which may be subtracted from measured concentrations when determining compliance with the limit value introduce significant uncertainty. In the numbers stated and the lines shown in Fig. 6 we take a conservative approach and subtract only sea salt and natural dust fields calculated by the EMEP CTM for the meteorological situation of 2009. The CHIMERE CTM, which has been used in the downscaling scheme, estimates significantly higher amounts of natural dust for the same conditions. Subtracting the CHIMERE natural dust fields from projected station concentrations in 2030 would increase attainment of the WHO guideline by about 13% for the CLAPP scenario in 2030, while inclusion of all natural aerosols would decrease attainment rate by 7%. Natural PM concentrations as modelled by the EMEP CTM are shown as shaded area for the year 2009; the same range applies for other scenario years as the natural components are assumed to remain constant over time.

While this issue points to uncertainties in the absolute estimates of compliance with given limit values, differences are systematic for individual stations and thus the relative changes between scenarios and scenario years are more robust, whichever absolute amount of natural $PM_{2.5}$ is subtracted.

4.4. Relationship between compliance and health impacts

In the previous sections, station by station concentrations and mortality from exposure to ambient background concentrations have been analysed separately. Ultimately, however, the target of air quality limit values is the restriction of health impacts for citizens. Hence, the question arises how these quantities are related, i.e. how the station concentrations translate into the population exposure index of the air quality directive, i.e., population-weighted averages, whether the stations are representative of exposure in the area they are supposed to represent, and how a tighter limit value might be set in order to reach a given level of anthropogenic exposure.

Compliance in the EU is assessed for air quality management zones, which are regarded as the primary territorial units for assessment and management of air quality under the air quality directives.³ The definition of the zones is the responsibility of national authorities, and different Member States follow different approaches in the designation of zones. In some countries, zones follow provincial boundaries and separate only major cities, while other countries have defined detailed zones. We employ the zones as defined by Member States in 2010 and do not attempt to evaluate the comparability of zones here, but rather use the polygons as the best information available for a reasonable grouping of stations and link between single sites and gridded exposure fields.

Compliance with a given limit value is determined by the highest measured concentration at any site inside the zone.⁴ Anthropogenic exposure is usually associated with background concentrations, while individual stations e.g. close to major roads may report considerably elevated concentrations. The question arises how the different quantities are related: the mean of measured ambient concentrations, the maximum of measured ambient concentrations, and actual population exposure.

³ The definition of air quality management zones used here was obtained from <http://www.eea.europa.eu/data-and-maps/data/zones-in-relation-to-eu-air-quality-thresholds-4>.

⁴ In the strict legal sense, compliance is determined from a large subset of stations, i.e. not every single AirBase station is considered. We do not take this fine distinction into account here.

Fig. 7 shows a comparison of the population-weighted mean modelled anthropogenic background $PM_{2.5}$ concentrations to the average measured concentrations in each air quality management zone in 2009. To make axes comparable, modelled fields of natural $PM_{2.5}$ are subtracted from observations. The size of the symbols corresponds to the area of the zone. Open circles represent zones with only one monitoring station, with increasingly dark shading indicating a higher number of stations. A large variation in zone sizes and station density is obvious (note the logarithmic scale for zone area). Zones with higher ambient concentrations tend to supply a better coverage of observations; nonetheless, there are cases where only 1–2 stations are available for zones of 10,000 km^2 and more. A close linear relationship is observed between the mean of the stations and the population-weighted mean concentration ($R^2 = 0.62$) in particular for zones with a high number of stations in which the point measurements are able to represent the average well. The linear fit is shown in the Figure (constrained to the origin); the population-weighted average concentration is related to the mean of monitoring stations by a factor of 0.76. Outliers are mainly observed for zones in which only a single station is available, which is not necessarily representative of the average. If all such zones are removed from the sample, R^2 for the constrained linear fit increases to 0.67. The region of Katowicze in Southern Poland stands out as a zone with many stations for which the modelled exposure ($37 \mu g/m^3$) is considerably higher than the mean of individual stations ($29 \mu g/m^3$), which is related to the over-estimation of urban background stations in Polish cities mentioned in Sect. 3.

Fig. 7 demonstrates a compact relation between averaged point measurements and the mean population exposure in a given zone, thus providing both support for the usefulness of air quality indicators associated with station means (such as the average exposure indicator mentioned in the EU Air Quality Directive) as well as the modelling capacities of GAINS.

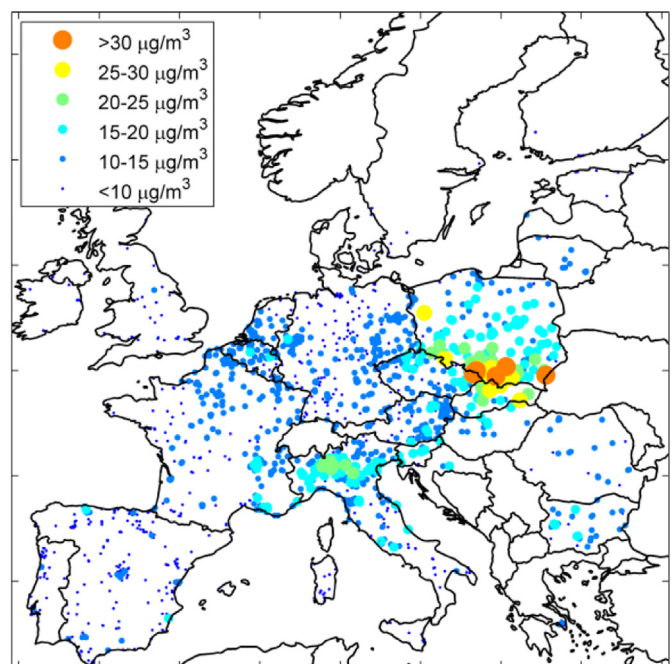


Fig. 5. Modelled $PM_{2.5}$ concentrations at all PM monitoring stations covered by GAINS, for the year 2030 under the assumption of current legislation. Natural dust and sea salt fields as modelled in the EMEP CTM are subtracted from calculated station concentrations.

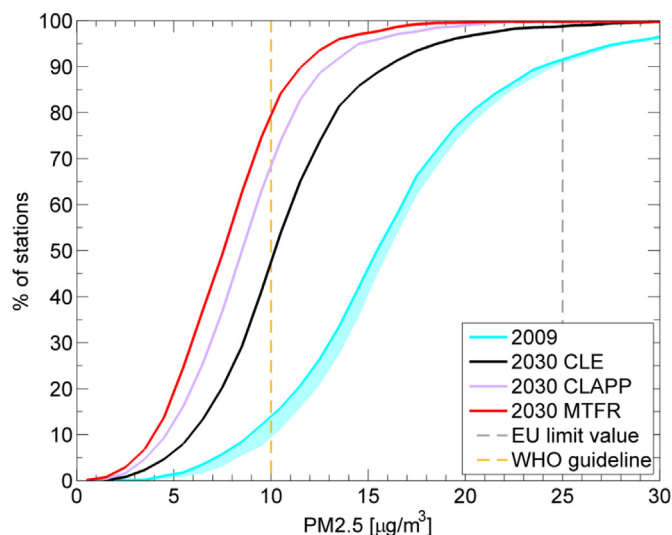


Fig. 6. Cumulative distributions of modelled $PM_{2.5}$ concentrations at all monitoring stations covered by GAINS, for the year 2009 and the year 2030 under current legislation (CLE), the EU commission proposal for revised emission ceilings (Clean Air Policy Package, CLAPP), and the maximum technically feasible reductions (MTFR). Lines correspond to station concentrations with EMEP dust and sea salt fields subtracted; for 2009, the shift of the curve by the subtraction of natural PM is shown as shaded area.

Compliance with the actual annual mean limit value of currently $25 \mu\text{g}/\text{m}^3$ is decided from the highest measured concentration in a zone, thus the question arises if a similar relation can be established for this indicator. Fig. 8 shows a similar comparison as Fig. 7, however using the highest measured concentration in each zone on the x axis. Again, modelled natural $PM_{2.5}$ fields have been subtracted from observations to make the axes comparable. As a guide to the eye, the black fit line from Fig. 7 has been replicated. A shift of the cloud is clearly visible, corresponding to the strong variations of observed concentrations within zones. The figure distinguishes

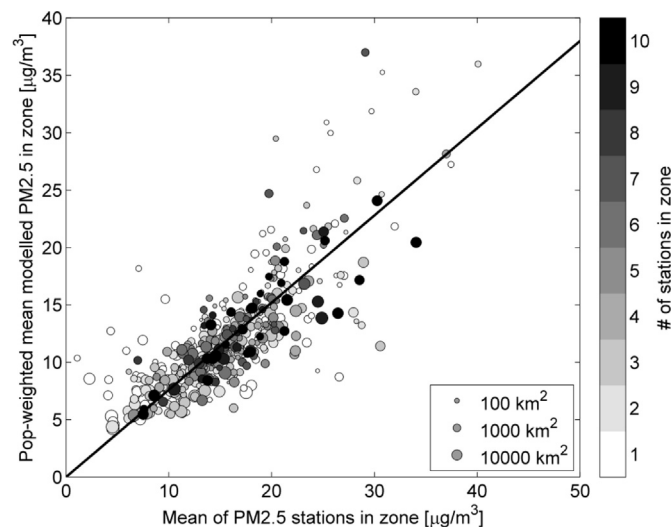


Fig. 7. Relation between the average of concentrations measured at monitoring station in a given air quality management zone, and the population weighted average modelled $PM_{2.5}$ concentration in the zone, for the base year 2009. The size of the symbol represents the area covered by the zone (logarithmic scale, samples shown in lower right corner), and the colour indicates the number of stations in each zone. Natural PM has been subtracted from observations to make concentrations comparable. Black line: Linear fit to mean of stations, constrained to origin.

cases in which the highest concentration is observed at roadside locations (triangles) from those where the highest station is a background site (circles). Understandably, the highest concentrations in many locations are observed at roadside stations. However, there is also a number of zones in which variability among stations representing urban background conditions is high, and in fact the highest concentrations across all stations analysed here are observed at background locations in Southern Poland (black dots with x values $\geq 40 \mu\text{g}/\text{m}^3$). A linear relation is less compact than to the mean of stations in a zone. Nevertheless 47%–56% of the variance in exposure is explained by a linear relation, depending on whether we constrain the regression to the origin. The corresponding regression line, shown in red in Fig. 8, is consequently shifted from the black line representing station means. In this case, we obtain a conversion factor of 0.65 between the highest station in a given zone and the average population exposure.

From the presented analysis, it is obvious that the two indicators – population average exposure and compliance at peak stations – are inherently different; a combined approach will thus be needed relying on both Europe wide measures to decrease average exposure and local measures to address local hotspots.

5. Discussion and conclusions

This paper has discussed the $PM_{2.5}$ related air quality indicators calculated in the GAINS integrated assessment model. Premature mortality is calculated from anthropogenic $PM_{2.5}$ concentrations on a $7 \times 7 \text{ km}$ grid, and compliance with limit values is calculated from $PM_{2.5}$ concentrations at monitoring stations, based on a combination of bottom-up modelled fields and past observations.

As an application of the modelling scheme, we explored potential impacts of the emission control scenarios in 2030 that are used as a basis for the revision of the EU air quality legislation. Ultimately, the driver for limit values on ambient concentrations of $PM_{2.5}$ has been concern for human health. The EU aims in its 7th Environment Action Program at attaining “levels of air quality that do not give rise to significant negative impacts on, and risks to human health and environment”. While emission ceilings are set for each Member State individually, the legal indicator for air quality is compliance with a fixed limit value on annual mean concentrations at individual monitoring stations.

As shown in Section 4, current EU emission control legislation will lead to a significant decrease of ambient $PM_{2.5}$ concentrations in comparison with past/present levels. By the year 2030, more than 98% of all stations can be assumed to meet the current EU limit value on $PM_{2.5}$ of $25 \mu\text{g}/\text{m}^3$ annual mean concentration without any further political action. However, at the same time the life expectancy of European citizens will remain shortened by several months, and acceptable levels of ambient $PM_{2.5}$ as indicated by the WHO guideline cannot be reached without further measures.

From the above discussion, it is obvious that the current limit value on $PM_{2.5}$ is hardly suited to motivate further emission controls in Europe's most polluted areas. The question arises how a limit value might be set which would achieve this but would still be attainable with reasonable efforts. From the analysis presented in Section 4.3, attainment of the WHO guideline seems infeasible at around 25% of the analysed monitoring stations. However, a large majority of stations (80–90%) is projected to have anthropogenic $PM_{2.5}$ concentrations below $12 \mu\text{g}/\text{m}^3$ in 2030 under the MTRF as well as CLAPP scenarios, hence attaining the US EPA standard with Europe-wide emission control measures. For the remaining stations, local measures such as traffic regulations, and fuel switches in domestic heating would be needed on top of the purely technical measures analysed here. Using the linear relation derived in Sect. 4.4., this concentration level would translate into an average

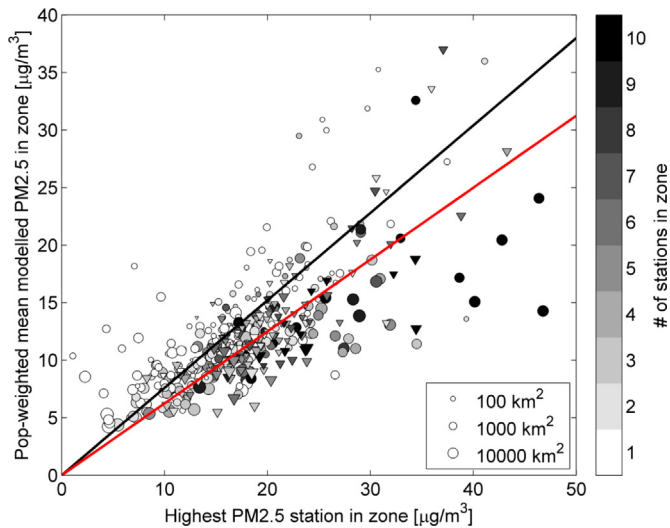


Fig. 8. Relation between the highest monitoring station in a given air quality management zone, and the population weighted average modelled $PM_{2.5}$ concentration in the zone, for the base year 2009. Circles indicate zones with a background station as the highest site, while triangles indicate a roadside station highest. The size of the symbol represents the area covered by the zone (logarithmic scale, samples shown in lower right corner, as Fig. 7), and the colour indicates the number of stations in each zone. Natural PM has been subtracted from observations. Black line: Linear fit to mean of stations, repeated from Fig. 7. Red line: Linear fit to highest station in each zone as shown in this figure (both regression lines constrained to origin). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

population exposure of $7.9\text{--}10\ \mu\text{g}/\text{m}^3$ depending on the linear fit used. Applying EU average life table factors, this corresponds to $4.9\text{--}6.3$ months of life expectancy lost; hence a limit value in the order of the US standard would limit loss of life expectancy to roughly half a year everywhere.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envsoft.2015.02.022>.

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