



Modelling street level
PM₁₀ across Europe

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Modelling street level PM₁₀ concentrations across Europe: source apportionment and possible futures

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Abstract

Despite increasing emission controls, particulate matter (PM) has remained a critical issue for European air quality in recent years. The various sources of PM, both from primary particulate emissions as well as secondary formation from precursor gases, make this a complex problem to tackle. In order to allow for credible predictions of future concentrations under policy assumptions, a modelling approach is needed that considers all chemical processes and spatial dimensions involved, from long-range transport of pollution to local emissions in street canyons. Here we describe a modelling scheme which has been implemented in the GAINS integrated assessment model to assess compliance with PM₁₀ (PM with aerodynamic diameter < 10 µm) limit values at individual air quality monitoring stations reporting to the AirBase database. The modelling approach relies on a combination of bottom up modelling of emissions, simplified atmospheric chemistry and dispersion calculations, and a traffic increment calculation wherever applicable. At each monitoring station fulfilling a few data coverage criteria, measured concentrations in the base year 2009 are explained to the extent possible and then modelled for the past and future. More than 1850 monitoring stations are covered, including more than 300 traffic stations and 80 % of the stations which exceeded the EU air quality limit values in 2009. As a validation, we compare modelled trends in the period 2000–2008 to observations, which are well reproduced. The modelling scheme is applied here to quantify explicitly source contributions to ambient concentrations at several critical monitoring stations, displaying the differences in spatial origin and chemical composition of urban roadside PM₁₀ across Europe. Furthermore, we analyse the predicted evolution of PM₁₀ concentrations in the European Union until 2030 under different policy scenarios. Significant improvements in ambient PM₁₀ concentrations are expected assuming successful implementation of already agreed legislation; however, these will not be large enough to ensure attainment of PM₁₀ limit values in hot spot locations such as Southern Poland and major European cities. Re-

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maining issues are largely eliminated in a scenario applying the best available emission control technologies to the maximal technically feasible extent.

1 Introduction

Particulate matter (PM) has become a major concern for public health in recent years (WHO, 2003, 2013). Especially particles with an aerodynamic diameter below $2.5\ \mu\text{m}$ ($\text{PM}_{2.5}$) have been associated with increased mortality mainly due to cardiovascular diseases. The most important sources of primary PM emissions include domestic combustion in household heating, road traffic, and industrial combustion. In addition to the emissions of primary particulate matter, particles are also formed in ambient air by chemical and physical processes from precursor gases.

Current European legislation (EU, 2008) states legally binding limit values on ambient concentrations of PM below $10\ \mu\text{m}$ diameter (PM_{10}): daily average PM_{10} concentrations must not exceed $50\ \mu\text{g m}^{-3}$ for more than 35 calendar days per year, and the annual mean concentration must not exceed $40\ \mu\text{g m}^{-3}$. Furthermore, a target value on $\text{PM}_{2.5}$ (annual mean concentration $< 25\ \mu\text{g m}^{-3}$) will become effective as a limit value in 2015.

Despite tightening of emission control legislation, EU Member States have been facing severe difficulties to attain these limit values (EEA, 2012). Compliance problems have been widespread and continuous at many locations. As the EU is currently revising its air quality legislation and planning new national emission ceilings for 2030, the question arises how compliance will evolve under different policy scenarios.

Modelling capacities of atmospheric PM have improved strongly in recent years. An overview of the state of the art modelling approaches is given by Rouil and Bessagnet (2013).

The GAINS integrated assessment model (Amann et al., 2011) is employed in the revision of the EU Thematic Strategy on Air Pollution (TSAP) as a policy tool to test the impacts of different pollution control options and calculate least cost solutions for

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achieving given policy targets (Amann et al., 2013). GAINS calculates particulate matter as the sum of primary PM, secondary inorganic aerosols caused by anthropogenic emissions of NH_3 , SO_2 , and NO_x , and secondary organic aerosols as a result of anthropogenic releases of non-methane volatile organic compounds (VOC).

5 We have recently introduced a downscaling scheme in GAINS to model NO_2 concentrations at different kinds of monitoring stations in the EU, including roadside stations (Kiesewetter et al., 2014). Here a similar scheme is developed which is now in use to assess future attainment of PM_{10} limit values in GAINS. In line with the methodology applied for NO_2 , we model annual mean concentrations based on past monitoring
10 data. At each air quality monitoring station, measured concentrations in the base year 2009 are disaggregated into contributions from regional background, urban increment, and roadside increment if appropriate. Individual contributions are then subject to the changes in the responsible emissions to calculate concentrations for scenario years.

This paper presents an introduction to the methodology used, a validation of trends
15 against observations, and applications of the model in the context of the revision of the EU air quality legislation. We quantify for several stations with high ambient concentrations the source contributions, pointing out large differences in the composition, and present an estimate of the evolution of PM_{10} concentrations in Europe until 2030 under different policy assumptions.

20 The remainder of this article is organised as follows: the modelling scheme is detailed in Sect. 2. A validation of modelled trends against independent observations for the years 2000–2008 is presented in Sect. 3. Uncertainties and shortcomings of the methodology are discussed in Sect. 4. Section 5 presents results: source contributions to different stations are analysed, and the evolution of compliance with limit values
25 in the EU is assessed under different assumptions for the evolution of anthropogenic emissions. Summary and conclusions are given in Sect. 6.

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2 Methodology

European legislation states two different limit values for PM_{10} concentrations (EU, 2008): annual mean concentrations must not exceed a value of $40 \mu\text{g m}^{-3}$, and daily average concentrations must not exceed $50 \mu\text{g m}^{-3}$ for more than 35 days in a calendar year. Out of these two limit values, the limit on daily average concentrations has proven more challenging to attain: e.g., while in 2009 more than 640 monitoring stations did not attain the daily limit value, only about 240 stations reported annual mean concentrations $> 40 \mu\text{g m}^{-3}$ (numbers refer to stations in the EU with more than 80 % data coverage). All of the latter did not attain the daily limit either. Hence, an assessment of future compliance with PM_{10} standards must focus on the daily limit value.

All calculations in GAINS are done on an annual mean basis and hence cannot address daily exceedances directly. However, a compact linear relation exists between the annual mean and the 36th highest daily average which is decisive for attainment of the daily limit value (see Fig. 1, showing observations from the AirBase¹ database in 2009): a 36th highest daily average of $50 \mu\text{g m}^{-3}$ corresponds to an annual mean concentration of $29.6 \mu\text{g m}^{-3}$. In a similar approach, Stedman et al. (2007) used a quadratic relationship between the number of days with PM_{10} concentrations greater than $50 \mu\text{g m}^{-3}$ and the annual mean to derive an equivalent annual mean concentration of $31.5 \mu\text{g m}^{-3}$. Hence we assess compliance with respect to an equivalent annual mean limit value of $30 \mu\text{g m}^{-3}$.

The modelling approach is similar to the one laid out by Kieseewetter et al. (2014) for NO_x and NO_2 . A schematic overview of the modelling approach is shown in Fig. 2. The 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. To ensure quality of the data, we consider only stations with more than 80 % temporal coverage of the

¹AirBase, the European air quality database. <http://acm.eionet.europa.eu/databases/airbase/>

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daily mean data. For any roadside monitoring station that fulfils this requirement, we first identify contributions from the ambient background and local road traffic emissions, and then model each of these contributions individually. The background itself is modelled as the sum of regional background contributions (primary and secondary) from Europe-wide emissions, an urban increment related to primary PM emissions from low-level sources, natural dust, and – if appropriate – a residual regarded as contribution from unknown sources. As a pessimistic assumption, this residual may be left constant in scenario calculations, as done with NO₂ residuals (Kieseewetter et al., 2014); a more realistic treatment attempts an allocation of this residual to natural contributions, regional and local emissions, as detailed below. Differences are only relevant in limited parts of Europe where the bottom up calculated concentrations significantly underestimate observations in 2009.

The following sections provide a description of the methodology for modelling the different contributions to the background (Sect. 2.1), and the roadside increment (Sect. 2.2). The synthesis of the different steps is described in Sect. 2.3.

2.1 Modelling background concentrations

Bottom up calculation of background concentrations is done in two steps, a coarse resolution transfer calculation and a fine scale increment relying on local emissions. All steps described here are done for PM₁₀ and PM_{2.5} independently; however, as the focus of this article is on PM₁₀ we do not mention PM_{2.5} explicitly here. Regional background concentrations are calculated from linear transfer coefficients at a resolution of 0.5° (lon) × 0.25° (lat) or roughly 28 km × 28 km, based on sensitivity calculations with the EMEP Chemistry Transport Model (Simpson et al., 2012). The EMEP model contains secondary inorganic as well as organic aerosol formation and calculates PM₁₀ concentrations from the source pollutants primary PM (PPM₁₀), NH₃, NO_x, SO₂, and non-methane volatile organic compounds (VOC). In order to match the expected situation best, expected emissions for the year 2020 under current legislation were used as base case for the EMEP model calculations. In each of the sensitivity runs, country

total emissions of one pollutant p from one source region r were reduced by 15% to calculate linear transfer coefficients $\pi(i, p, r)$ from r to each grid cell i ,

$$\pi(i, p, r) = \frac{[\text{PM}_{10}]_{\text{base}}(i) - [\text{PM}_{10}]_{\text{red}}(i)}{0.15E_{\text{base}}(p, r)} \quad (1)$$

with $E(p, r)$ denoting country total emissions of pollutant p in region r and the subscripts _{base} and _{red} referring to the model run with full 2020 emissions and that with reduced emissions, respectively. Fifty-seven source regions are included, covering Europe and the surrounding sea regions, as described by Kieseewetter et al. (2014).

PM_{10} concentrations for each EMEP grid cell i are then calculated as the sums of contributions from all source regions r and pollutants p ,

$$\text{PM}_{10}(i) = \delta_{\text{PM}_{10}} + \sum_{r=1}^{57} \sum_{p \in \{P, A, N, S, V\}} \pi(i, p, r) \cdot E(p, r) \quad (2)$$

with P, A, N, S, V denoting the source pollutants primary PM_{10} (PPM₁₀, “P”), NH₃ (“A”), NO_x (“N”), SO₂ (“S”), VOC (“V”). $\delta_{\text{PM}_{10}}$ denotes the residual resulting from non-linearities in the system and boundary conditions; it is calculated as difference between the sum of linear contributions from base case emissions and the base case concentrations modelled with the full EMEP CTM. This model-intrinsic residual is slightly negative in the Po valley, and between 0.5 and 2 $\mu\text{g m}^{-3}$ in the rest of Europe.

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 credible as long as changes in the three precursor gases are similar.

The $0.5^\circ \times 0.25^\circ$ resolution of the linear transfer coefficients is not sufficient to calculate realistic urban background PM concentrations. Kieseewetter et al. (2014) used

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a full year simulation performed with the CHIMERE Chemistry Transport Model (Menut et al., 2013) with a grid resolution of 0.125° (lon) \times 0.0625° (lat) or roughly $7\text{ km} \times 7\text{ km}$ to calculate for NO_x a sub-grid increment to the urban background level. Here we use the same simulation to derive a concentration increment for PM_{10} . As the formation of secondary PM takes place on timescales of hours, the urban increment is calculated as a function of primary PM emissions. For the CHIMERE model runs used here, Cuvelier et al. (2013) showed that most of the concentration increment from the $28\text{ km} \times 28\text{ km}$ to the $7\text{ km} \times 7\text{ km}$ resolution is explained by emissions of primary PM. This approach is used here to calculate a regression coefficient ξ relating increments in the PM_{10} concentration to additional emissions of PPM_{10} , so that in a sub-grid cell m of the $28\text{ km} \times 28\text{ km}$ grid cell i the PM_{10} concentration is calculated as

$$[\text{PM}_{10}](m) = [\text{PM}_{10}](i(m)) + \xi(i(m)) \cdot \{e_L(m) - e_L(i(m))\}, \quad (3)$$

with $e_L(m)$ the low level emissions in m and $e_L(i(m))$ the same averaged over the corresponding EMEP grid cell i . The parameter ξ relates the pattern of concentration increments to the pattern of emissions. It depends largely on the meteorological characteristics of the area in question. Although calculated only for 2009, ξ introduces a parametrisation of the urban increment with low level emissions that can easily be transferred to different scenario years. Since this resolution-dependent concentration increment is relevant mostly in urban areas, we refer to it also as urban increment, although it is calculated for every EMEP grid cell regardless of its location and may also be negative in sub-urban grid cells. EMEP grid cells containing parts of the same urban area are combined in the regression analysis, enhancing the statistical significance of the calculation. Each major city is thus assigned a single characteristic value of ξ .

A map of ξ for the whole domain of the CHIMERE model is shown in Fig. 3. Large differences are visible between different regions owing to the different meteorological conditions that influence boundary layer mixing. Particularly, the effect of low wind speed and frequent inversion layers is visible in Alpine regions and the Po valley, whereas

the higher wind speeds lead to correspondingly lower ξ values close to the Atlantic or North Sea shorelines.

R^2 values for the regression used in ξ calculation are high especially in major urban areas with significant PM emissions. Major European cities like Paris, London, Berlin, Madrid show values around 0.9 or higher.

While the urban background in large urban areas is represented well by the 7 km \times 7 km concentrations, concentrations in smaller cities are underestimated as the CHIMERE grid cells are not small enough to capture inner city concentrations. Adopting the methodology described by Kieseewetter et al. (2014), we use population density on a $0.01^\circ \times 0.01^\circ$ grid to redistribute domestic and light duty vehicle emissions and apply Eq. (3) to inner urban emission densities for 376 European cities with more than 100 000 inhabitants.

2.2 Modelling the traffic increment

Roadside concentrations of PM are typically a few $\mu\text{g m}^{-3}$ higher than concentrations in ambient urban background air (around $5 \mu\text{g m}^{-3}$ on the European average, see Fig. 6, but with a large spread); the difference originates from traffic related emissions of particles in the street canyon itself. We define the PM_{10} roadside increment as

$$\Delta[\text{PM}_{10}] = [\text{PM}_{10}]_{\text{road}} - [\text{PM}_{10}]_{\text{B}} \quad (4)$$

with $[\text{PM}_{10}]_{\text{road}}$ and $[\text{PM}_{10}]_{\text{B}}$ the roadside and urban background concentrations of PM_{10} (equivalently for fractions of PM_{10} or other tracers).

On time scales relevant for the mixing of air within street canyons, secondary particle formation can be neglected. Traffic related PM originates not only from combustion processes, but contains also a significant fraction of non-exhaust emissions from brake and tyre wear, road surface abrasion, and resuspension of road dust (Thorpe and Harrison, 2008).

The coarse fraction of PM ($\text{PM}_{\text{coarse}} = \text{PM}_{2.5-10} = \text{PM}_{10} - \text{PM}_{2.5}$) has been found to consist almost entirely of non-exhaust particles (Harrison et al., 2012), and at the same

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time is more affected by resuspension as it may accumulate on the road surface. Between different regions, large differences exist in the size partitioning and thus exhaust or non-exhaust origin of the PM₁₀ roadside increment: in London, Harrison et al. (2001) determined a roughly even split of the roadside increment in PM_{2.5} and PM_{coarse}, while in Nordic countries the coarse fraction dominates, caused by the widespread use of studded tires and application of traction sanding in winter (Kupiainen et al., 2005; Gustafsson et al., 2009).

As both the sources and the dispersion behaviour of fine and coarse traffic related PM are different, fine and coarse fractions are treated individually in the traffic increment calculation. Only few monitoring sites in Europe enable a distinction of fine and coarse roadside increment from observations. Thus, in our model the components are estimated via a correlation with the NO_x roadside increment, of which measurements are widely available.

The approach followed here distinguishes and idealises the fine and coarse fractions of PM. We assume that primary PM_{2.5} is dispersed like NO_x, which is chemically inert at the timescales involved, while PM_{coarse} is subject to accumulation and resuspension. The activity that causes the concentration increments in NO_x and PM_{2.5} is the same (namely vehicular emission in the street canyon in question), hence we can write

$$\Delta[\text{PM}_{2.5}] = \Delta[\text{NO}_x] \cdot \frac{E_{\text{PM}_{2.5}}}{E_{\text{NO}_x}} \quad (5)$$

with $E_{\text{PM}_{2.5}}$ and E_{NO_x} the national total emissions of each pollutant from road traffic. Due to the lack of station specific data we assume that the fleet composition at any station is well represented by the national average for urban conditions. A similar concept has been used by Boulter et al. (2006) for estimating the resuspension contribution to the roadside PM increment. Figure 4 shows this relation for Marylebone Road traffic station in London, using AirBase daily observations for the year 2009. Some roadside stations also show good correlation between $\Delta[\text{NO}_x]$ and $\Delta[\text{PM}_{10}]$; however, we do not use this relation but focus on the fine fraction here. To avoid unrealistically large PM_{2.5} roadside

increments in case of observational errors, the fine fraction is limited to 90 % of the total PM_{10} increment in the base year.

The coarse fraction of the traffic increment is then estimated as the residual

$$\Delta[PM_{\text{coarse}}] = [PM_{10}]_{\text{obs}} - [PM_{10}]_B - \Delta[PM_{2.5}] \quad (6)$$

with $[PM_{10}]_B$ and $[PM_{10}]_{\text{obs}}$ the observed background and roadside concentrations, respectively.

Once the fine and coarse fractions of the roadside increment are estimated for the base year, each of them is scaled individually with the appropriate trend in urban $PM_{2.5}$ or PM_{coarse} road traffic emissions (exhaust + non-exhaust). The trend in PM_{coarse} traffic emissions is essentially proportional to the trend in traffic volume as these non-exhaust emissions are not controlled on a large scale so far. As the PM_{coarse} roadside increment contains a significant fraction of re-suspended dust, the assumption that concentrations scale proportional to emissions may be too pessimistic, as the additional contribution of a single vehicle to dust resuspension decreases with total traffic volume (Boulter, 2005).

Wherever possible, the same background stations are used for PM_{10} and NO_x in the roadside increment calculation. Provided that sufficient temporal overlap exists (> 75 % of all days in 2009), $\Delta[PM_{10}]$ and $\Delta[NO_x]$ are calculated as annual averages over all days when NO_x and PM_{10} roadside and background stations provide data. If station pairs are not available, NO_x and PM_{10} background are calculated independently; if for a station pair sufficient overlap period is not available, $\Delta[PM_{10}]$ and $\Delta[NO_x]$ are calculated without temporal synchronisation.

2.3 Combination of the different modelling steps

Model calculations are possible for every station in the AirBase database which fulfils a few data coverage criteria: for background stations, all stations with more than 80 % coverage of daily mean PM_{10} concentration data are included. For roadside stations, in addition NO_x data are required for the same station, and at least one suitable PM_{10} and

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one NO_x background station, ideally identical, are needed. All of these stations must fulfil the 80 % temporal coverage criterion. With these criteria, a total of around 1860 PM₁₀ stations are covered by the model, of which 316 are traffic stations and 492 did not attain the equivalent limit value as defined in Sect. 2 in 2009 (315 if contributions from natural dust and sea salt are subtracted).

Calculations involve two steps: first, the calculation is done for the base year 2009, determining the observed background PM₁₀, modelled background PM₁₀ as described in Sect. 2.1, and the residual from calculated to observed background. GAINS transfer coefficients pertain only to anthropogenic emissions. Suspension and dispersion of natural dust and sea salt are calculated in the EMEP CTM for the year 2009. These natural fields are subtracted from observations before determining the residual. For a traffic station, the fine and coarse fractions of the observed roadside increment are calculated as described in Sect. 2.2.

As a second step, calculations are done for any scenario year by replacing base year emissions with emissions for the scenario year in question. GAINS calculates emissions bottom up from projections of anthropogenic activity, estimated shares of emission control technologies and appropriate emission factors for each technology (Klimont et al., 2002). GAINS provides emissions typically in five year intervals extending from 2000 to 2030; for other years emissions are interpolated linearly between these points.

In case of a positive residual in base year background concentrations (model under-explaining observations), the residual may be related to natural dust, re-suspension of dust, missing emissions or a missing representation of boundary layer inversions in the EMEP or CHIMERE model simulations. While the unexplained residual is kept constant in the NO₂ scheme (Kieseewetter et al., 2014), this treatment seems too pessimistic for PM₁₀ in some European regions: particularly in Southern Poland, extreme measured concentrations are at some stations not matched by the model. However, both temporal profile as well as geographical distribution of the offsets suggest a clear relation to domestic combustion in winter, indicating that domestic emissions are underesti-

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mated in emission inventories, or boundary layer mixing is overestimated in the CTM simulations. Consequently, a simple “best estimate” disaggregation of the residual concentration is undertaken. First, the residual is disaggregated into a regional and a local unexplained component; the regional component is determined as the linear interpolation of unexplained residuals at nearby rural background monitoring stations, while the remainder is by definition caused by local emissions. Within the regional component, natural dust is increased up to a reasonable maximum (the PM₁₀ dust fields used in the CHIMERE simulation, which are considerably higher than the EMEP dust fields that are used regularly in GAINS), and the rest is assumed to be composed like the modelled 28 km × 28 km concentrations at this location. The local residual component, on the other hand, is assumed to be related to an underestimation of local emissions or their enhancement through inversion situations, and are attributed proportionally to the gridded primary PM emissions within a radius of 20 km. While this methodology can only provide a rough estimate and takes into account only “known unknowns”, it still seems more realistic than keeping the residual constant.

If the residual is negative (model over-explaining observed background), the ratio of observed to calculated background PM₁₀ in the base year is used to scale calculated concentrations in scenario years.

3 Validation

Validating a model which calculates PM concentrations for roughly 1860 air quality monitoring stations is challenging. Here we show a comparison of bottom up calculated background PM concentrations for various background stations in Europe, and a validation of trends at background and roadside monitoring stations. Since the model is constrained by observations in the base year, validating absolute modelled concentrations at roadside monitoring stations is not possible.

Figure 5 compares PM_{2.5} and PM₁₀ background concentrations from bottom up modelling to observed concentrations at background monitoring stations, for urban and

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rural background stations separately. This provides a validation of the background calculation methodology from linear transfer coefficients plus downscaling to the urban background level. Each dot in the figure represents the annual mean at one monitoring station. The offset to the 1 : 1 line is compensated in scenario calculations as described in Sect. 2.3. We here use a subset of the model performance indicators proposed by Thunis et al. (2012): absolute bias, normalised mean bias, and correlation coefficient. PM_{2.5} concentrations are generally well modelled with a residual of $-2.5 \mu\text{g m}^{-3}$ (normalised mean bias -15%) remaining on the European average, 94 % of stations between a factor of two margins from the observations. The mean bias decreases to $-0.9 \mu\text{g m}^{-3}$ (-5%) at urban background stations located in cities $> 100\,000$ inhabitants, where urban polygons were defined as described by Kieseewetter et al. (2014) (black dots in Fig. 5a). Urban background stations in smaller cities for which urban polygons are not defined (open circles in Fig. 5a) have a considerably higher offset of $-6.6 \mu\text{g m}^{-3}$ or a normalised mean bias of -36% . This points to the added value of the last downscaling step beyond the 7 km CHIMERE grid resolution wherever possible, and at the same time supports the re-allocation of local residuals to nearby primary PM emissions as described in Sect. 2.3. At rural background stations (Fig. 5b) the model has a mean bias of $-1.9 \mu\text{g m}^{-3}$ (-15% normalised mean bias).

The performance of the model is less encouraging for the coarse PM fraction. The spatial variability between stations is underestimated, leading to an average bias of $-6.5 \mu\text{g m}^{-3}$ or 26 % of observed PM₁₀ in the base year (for urban background stations, $-3.2 \mu\text{g m}^{-3}$ or -12% at stations within urban polygons, compared to $-10.8 \mu\text{g m}^{-3}$ or -37% at stations without urban polygons). Correlation coefficients between model and observations are 0.76 and 0.83 for urban background and rural background PM_{2.5}, respectively, and around 0.6 for PM₁₀. Aside from uncertainties in direct anthropogenic emissions of PM or its precursors, offsets partly arise from uncertainties in the natural emissions and effects of re-suspended dust.

For the full PM₁₀ model, since offsets in the base year are compensated, only trends can be validated. Modelled trends in the decade 2000–2009 are compared to observa-

tions in Fig. 6. Here, model predictions at different categories of monitoring stations are compared to the annually averaged observations (only stations with at least five years of data are included here).

Different observational methods are applied in different locations. Particularly the use of the Tapered Element Oscillating Microbalance (TEOM) causes difficulties in comparing results to the standard gravimetric method as some semi-volatile compounds are lost in the measurement process due to the necessary heating of the sample (e.g. Hauck et al., 2004). Scaling factors are usually applied to correct for these offsets to the reference method; however, there is no uniform methodology how these are calculated across the EU. TEOM measurement data from France exhibit a step increase when a new methodology (adjustments based on TEOM Filter Dynamics Measurement System measurements) was introduced in 2007 to include the semi-volatile components. To establish a consistent time series and foster comparison with other monitoring sites, raw data from French TEOM measurement sites before 2007 were scaled by average correction factors as reported by AIRPARIF (2011a): +20 % for roadside stations and +30 % for background stations.

Trends are well captured by the model: slight declines of around $-0.36 \mu\text{g m}^{-3} \text{yr}^{-1}$ (urban background), $-0.45 \mu\text{g m}^{-3} \text{yr}^{-1}$ (traffic), and $-0.48 \mu\text{g m}^{-3} \text{yr}^{-1}$ (rural background) are seen in the decade 2000–2009. The decline in observed roadside PM_{10} concentrations is stronger than modelled ($-0.71 \pm 0.20 \mu\text{g m}^{-3} \text{yr}^{-1}$), which is due to a stronger decline in the roadside increment in observations. This possibly points to successful local measures that have been implemented during this decade in order to reduce dust suspension from road traffic at hot spot sites (e.g. dust binding and street cleaning measures in Scandinavian countries, changes in winter road maintenance) and that are not represented in the Europe wide emission calculation scheme. The conclusion from Fig. 6 is that rural and urban background concentrations are on average modelled well, while the model may be slightly pessimistic for future roadside concentrations.

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4 Uncertainties and caveats

The simplifications needed in a Europe wide modelling of PM down to individual street canyons lead to considerable uncertainty. Kieseewetter et al. (2014) provided a thorough discussion of the uncertainties associated with the roadside NO₂ calculation scheme which follows a very similar approach. Hence, we here only provide a short discussion of the uncertainties specific to the PM scheme and refer the reader to the cited reference for a more general treatment.

Limitations induced by the linearised approach taken here have been mentioned in Sect. 2.1, and are discussed by Amann et al. (2011).

Considerable uncertainties stem from the emission inventory used for the base year. The emission inventory itself is described by Kieseewetter et al. (2014). Emissions from domestic combustion are uncertain in critical regions such as Southern Poland or Bulgaria, where this sector is believed to be of key importance. Test runs with the CHIMERE CTM revealed that domestic heating emissions in Southern Poland are considerably underestimated in official reportings and previous versions of GAINS. Consultations with national experts led to the conclusion that this discrepancy is likely caused by the more widespread use of low quality coal for household heating in coal mining and adjacent areas than previously assumed. As a preliminary solution, domestic combustion emissions from provinces with active coal mines were multiplied by a factor 8, while these in neighbouring provinces were adjusted by a factor of 4. These adjusted emissions lead to a distinctively better match of modelled with measured PM₁₀ concentrations in Poland.

While such a flat correction factor adjusts the average well, at some monitoring stations a significant unexplained share remains (particularly in small cities, while concentrations in large cities are a bit overestimated). As a worst case scenario this residual may be left constant, as it is not explained by the emission inventory (including adjustments). However, in this case several regions would have little chances of attaining air quality limit values, which seems unrealistic in case of targeted action such as as-

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sumed in the policy scenarios. Therefore, residuals were site-specifically attributed to their likely sources as described in Sect. 2.3; however, the air quality benefits achieved under control scenarios are in these regions subject to considerable uncertainty.

While unit emissions of particles and aerosol precursors from combustion processes are well quantified, non-exhaust emissions are more uncertain, and suspension of natural or road dust is not well quantified at all. Road dust resuspension is only considered in the roadside increment in our scheme, where it is included in the residual from calculated PM_{2.5} increment to the full PM₁₀ increment. However, this simple scheme does not take account of the many factors usually considered in detailed road dust resuspension models such as Nortrip (Denby et al., 2013). Detailed input data as required in these models are not readily available for hundreds of roadside monitoring stations in Europe. The estimation of fine and coarse roadside increment from the proportionality to the NO_x increment creates a strong dependency on the quality of observations, particularly on inter-comparability of PM and NO_x observations.

PM concentrations are subject to strong inter-annual variability (see Fig. 6) due to changeable meteorological conditions and dust episodes. Due to practical limitations in computing time, the urban increment calculation with 7 km × 7 km resolution could only be performed for one year, which was selected as the most recent year with AirBase observations and meteorological fields available at the starting time of this work. Judging from the historical trend shown in Fig. 6, 2009 does not seem to show unusually high or low concentrations in relation to other years on the European average; however, we do acknowledge that the reliance on one year introduces systematic station related uncertainty in modelled concentrations for the future.

Given the uncertainties and approximations, it is clear that this modelling scheme is not able to, nor is it supposed to, substitute detailed local scale modelling. A Europe wide integrated model must make compromises, and there is definitely space for refinements in the methodology in the future. Results for individual stations need to be used with care, results are best analysed as an ensemble. Still, as a more detailed look at individual stations shows, the model is able to give a reasonable representation of

different stations with different characteristics (Sect. 5.1). Hence, it offers the unique possibility of studying – with all uncertainties and caveats mentioned – the effects of Europe wide air quality policy choices on ambient concentrations at the whole variety of monitoring stations available in Europe.

5 Results and discussion

This section applies the modelling scheme introduced in this article to quantify source contributions to PM₁₀ concentrations for a set of critical stations (Sect. 5.1), and to provide an outlook on the evolution of Europe wide PM₁₀ concentrations and the possible attainment of limit values under future emissions (Sect. 5.2).

5.1 Source allocation of PM₁₀: examples of critical stations

Thanks to the structure of the model, the source composition of modelled PM₁₀ in terms of component and origin can be traced for every single station. This section attempts to give some examples for source attributions of PM₁₀ at urban monitoring stations in the base year.

Figure 7 shows the spatial allocation of origin for seven monitoring stations in the base year. The set is rather arbitrary but stations were selected as examples for critical stations with different characteristics. PM₁₀ concentrations are disaggregated into contributions from natural dust and sea salt, transboundary, national, urban, and street canyon increments, similar to the categories used e.g. by Lenschow et al. (2001); all of the anthropogenic contributions are further split into fine and coarse PM fractions. To arrive at the disaggregation shown here, regional background levels have been determined from the interpolation of nearby rural background stations, and unexplained residuals are allocated to missing emissions as described in Sect. 2.3. Before the re-allocation, residuals at these stations were between –20% and 20%.

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Stations selected here are located in Paris (FR04058, A1 Saint Denis), Krakow (PL0038 A²), Torino (IT0469A, Consolata), Stockholm (SE0003A, Hornsgatan), Essen (DENW134, Gladbecker Str.), London (GB0682A, Marylebone Road), and Vienna (AT9RINN, Rinnböckstraße). While all of these stations exceeded the 30 $\mu\text{g m}^{-3}$ equivalent limit value in 2009, source allocations show large differences in the reasons for the exceedances. Five of the six stations shown are traffic stations, with Krakow – the station with the second highest 2009 annual mean among them – being the only exception as an urban background station. With urban background concentrations at this level, the situation at curbside locations may be expected to be even worse. All spatial source categories shown have their part, although contributions of each fraction vary strongly between stations: while Torino is shielded by the alps and consequently transboundary transport contributes only little to ambient PM, Vienna or Essen are significantly influenced by transboundary transport of pollution due to their geographical locations. Conversely, a high regional background related to Italian emissions is found in Torino, whereas in Stockholm the influence of Swedish emissions outside the city itself is almost negligible. The regional background, composed of natural, transboundary and national contributions, is around 20 $\mu\text{g m}^{-3}$ in most of the cities included here; lower levels are found in London and Stockholm. Such regional background levels leave only little room for urban and roadside increments if a limit of 30 $\mu\text{g m}^{-3}$ is to be matched, pointing to the multi-scale nature of the problem. Extreme differences are seen in both the urban and roadside increments, relating to local emission densities in the domestic and transport sectors as well as to atmospheric mixing conditions in the boundary layer (for the urban increment) or the layout of the street canyon. Note the strong differences regarding the split of the roadside increment into fine and coarse PM fractions, as it is estimated from the observed NO_x increment. While the fine fraction, caused mostly by exhaust emissions, slightly dominates at most stations, a dominating coarse component is found in regions with intensive use of traction sanding in winter or even studded tires such as in Stockholm. Both extreme examples, London Maryle-

²AirBase station name: MpKrakowWIOSPrad6115

bone Road (large fine increment) and Hornsgatan (large coarse increment), offer PM_{2.5} observations which confirm the split of the roadside increment; in Torino and Vienna the PM_{coarse} fraction of the roadside increment seems rather high and may be a bit over-estimated.

A large roadside increment can be viewed as an opportunity – if the main cause of the problem is a local one, local action has a chance to alleviate the problem. If, on the other hand, only Europe wide policy measures are adopted, which address only the fine, combustion generated particulates, cities with strong resuspension of road dust will face severe difficulties in reducing ambient concentrations.

Figure 8 shows the chemical composition of PM at the same set of monitoring stations as before. Chemical constituents are split up into natural, primary anthropogenic PM (PPM), secondary inorganic aerosol (SIA), and secondary organic aerosol (SOA), for both fine and coarse fractions. The primary coarse component includes non-exhaust emissions and resuspended dust, which is not distinguished explicitly in the model.

Comparing the chemical composition to observations is challenging for two reasons. Firstly, measured composition data are usually only available on a short term basis, often for episodes of high pollution; however, during such episodes the contributions can deviate significantly from the annual mean. Secondly, measured source categories are not easily translated into composition as modelled in GAINS. Hence, while a complete validation of the chemical composition is beyond the scope of this article, the purpose of this section is to point out a few characteristics.

The fine fraction constitutes about two thirds (59–73 %) of total PM₁₀ at six out of the seven stations, with Stockholm being the only exception (only 27 %) for the reasons discussed above. As for the spatial origin of PM, large differences are also encountered in terms of chemical composition. Dust and sea salt contribute 1–5 µg m⁻³ to PM₁₀, mostly in the coarse fraction. The largest contribution to PM₁₀ comes from primary particles (49–85 %); however, in the fine fraction, secondary aerosol concentrations are slightly higher than primary ones in Vienna and Essen.

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Secondary inorganic aerosol concentrations are straightforward to be compared to observations. AIRPARIF (2011b) report SIA concentrations of $6.5 \mu\text{g m}^{-3}$ at Paris roadside locations, which is matched well by GAINS ($6.3 \mu\text{g m}^{-3}$). For Vienna, Bauer et al. (2006) give annual average SIA concentrations of $11 \mu\text{g m}^{-3}$, close to the values shown in Fig. 8 ($10.5 \mu\text{g m}^{-3}$); however, measurements were made in 2004. In Stockholm, SIA formation is considerably lower, with the $3.6 \mu\text{g m}^{-3}$ modelled in the range of observations reported by Querol et al. (2004) ($3\text{--}5 \mu\text{g m}^{-3}$).

Among the stations included here, the highest SIA contribution in absolute terms is modelled in Krakow ($12.7 \mu\text{g m}^{-3}$) due to high SO_2 emissions and subsequent sulphate formation in this region. Overall, SIA contributes 10 % (Stockholm) to 34 % (Vienna) to PM_{10} . 80–95 % of the SIA is in the fine fraction of PM, with only minor contributions in the coarse fraction (essentially NaNO_3). Secondary organic aerosol formation is modelled but not of significant importance ($0.3\text{--}2.1 \mu\text{g m}^{-3}$ or 1–6 % of $\text{PM}_{2.5}$), with the highest values found in Torino.

Due to the simplifications of the model construction, the source attribution presented here can only give a rough estimate. It is meant to show the differences between individual stations and regions rather than provide exact results for which urban scale modelling based on local emission inventories is needed.

5.2 An outlook on the attainment of air quality standards

The modelling scheme described in this article has been applied in the ongoing revision of the EU air quality legislation to derive estimates of compliance with limit values under various emission scenarios. Here we show results for two specific scenarios, assuming either a political stagnation at currently approved emission control legislation (“CLE” = current legislation scenario), or a very ambitious policy scenario applying the most efficient control technologies available (“MTFR” = maximum technically feasible reductions scenario).

Figure 9 shows the trends of PM and precursor gas emissions under the scenarios used. The CLE scenario was used as the baseline case for the revision of the EU

Thematic Strategy on Air Pollution (TSAP); it has been described in detail by Amann et al. (2013), with recent updates described by Amann et al. (2014).

Considerable decreases in $PM_{2.5}$ and SO_2 , NO_x and VOC emissions are expected under current legislation from ongoing implementation of exhaust cleaning technologies. No further reductions are expected for PM_{coarse} , and hardly any for NH_3 emissions.

Analysis conducted for the TSAP revision has highlighted the potential for emission reductions beyond the baseline case. The MTRF scenario assumes that (within certain limitations of feasibility) all pollution sources are equipped with the best available emission control technology. Emissions under the MTRF scenario for 2030 are shown as circles in Fig. 9. Considerable reductions beyond the baseline are possible for all pollutants, however, this may come at relatively high costs. Realistic strategies are usually based on a partial closure of the gap between baseline and full application of the best available technologies. The strength of the GAINS model is then to find cost-optimal solutions for given health or air quality targets. However hypothetical for practical implementation, the MTRF scenario provides a quantification of what is possible in terms of emission reductions *without changing the levels of anthropogenic activities*, i.e. no behavioural changes and no switches to other fuel classes or renewable energy generation other than assumed in the baseline case which relies on the latest PRIMES-2013 scenario for energy consumption.

Figure 10 shows distributions of modelled PM_{10} concentrations at all stations covered in the modelling scheme, for the base year as well as the scenario year 2030, comparing the modelled evolution under CLE and MTRF scenarios. Since EU legislation allows for natural contributions to be subtracted from measured concentrations, dust and seasalt fields as used in the EMEP model are subtracted here from total modelled concentrations. While about 320 (17 %) of the stations exceed the equivalent limit value of $30 \mu g m^{-3}$ in 2009 (dashed), increasing controls on emissions are expected to result in decreasing concentrations and consequently a higher fraction of attainment of the limit value across the EU already in the baseline case. However, after 2020 con-

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centration decreases are slow, and about 100 (6%) of the stations operative in 2009 are expected to remain above the equivalent limit value in 2030. A large amount of stations remains close to the equivalent limit value, so that definite statements about compliance are difficult.

5 Considering that the equivalent limit value is defined on a statistical base, with some stations exceeding the daily limit value even at annual mean concentrations below $30 \mu\text{g m}^{-3}$ (Fig. 1), and also taking into account inter-annual meteorological variability, only stations below $25 \mu\text{g m}^{-3}$ should be considered to be in safe compliance. This $5 \mu\text{g m}^{-3}$ margin corresponds to the mean range of inter-annual Europe wide PM_{10} variations as seen in Fig. 6. More than 12% of the stations are not expected to meet this $25 \mu\text{g m}^{-3}$ limit in 2030 under CLE assumptions.

10 Full propagation of the maximum technically feasible emission reduction technologies would improve the compliance situation drastically, eliminating close to all stations above $30 \mu\text{g m}^{-3}$ (0.7%), and bringing 97% of the stations below $25 \mu\text{g m}^{-3}$. Several stations remain at annual mean concentrations close to the limit value, so that attainment of the limit value is not certain, particularly in years with unfavourable meteorological conditions. Additional local efforts may be warranted to ensure compliance in these cases.

15 Critical areas are identified easily in Fig. 11 showing a map of air quality monitoring stations colour coded by their modelled PM_{10} concentrations under the CLE scenario in 2030. From the discussion above, only the “green” stations below $25 \mu\text{g m}^{-3}$ can be assumed to be in relatively safe compliance.

20 Difficulties are expected to remain in several European cities, Southern Poland and bordering areas in Czech and Slovak Republics, Northern Italy, and Bulgaria. Different causes are responsible for the remaining difficulties: large cities are mainly under pressure from increasing traffic, with the unregulated non-exhaust emissions (and dust resuspension) eventually becoming dominant, while typically relatively clean fuels are used for household heating. If traffic volumes within large cities increase further, and if no additional measures on non-exhaust emissions are taken, several cities may move

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out of the compliance zone again. As non-exhaust road traffic emissions are influenced to a significant degree by road maintenance applied in winter, when inversion layers are frequent and emissions from domestic heating are maximal, several stations may violate the limit on daily exceedances in spite of annual means staying below 30 $\mu\text{g m}^{-3}$. Here, additional local action (e.g. restrictions on studded tyre use in Scandinavian countries, enhanced street cleaning, traffic restrictions) may be required to ensure safe attainment of the limit values. Eastern European countries, on the other hand, suffer from the widespread use of solid fuels such as low-grade coal or inefficient wood burning. Efficient emission cleaning technologies can improve the situation dramatically, as shown in Fig. 10; however, a hypothetical switch to cleaner fuels would provide for even better results.

6 Conclusions

This paper presents an introduction to the station based modelling methodology that has been introduced in the GAINS integrated assessment model to calculate concentrations of PM₁₀ and estimate compliance with limit values. Results are calculated for a total of around 1860 monitoring stations reporting to AirBase. The modelling approach is based on explaining observed concentrations for the base year 2009 to the extent possible with a chain of simplified atmospheric chemistry and transport calculations with models of different scales. Concentrations for other years are then calculated by substituting emissions from the GAINS bottom up emission calculation scheme.

Due to the complexity of the system involving different spatial scales, simplifications are necessary. The modelling scheme is not intended to replace detailed small scale dispersion modelling. The focus here is to provide an estimate of the effects of Europe wide air quality policies on the attainment of limit values. Although results are calculated for each station individually, they are best evaluated on an ensemble base, as individual emission trends are not calculated for each station. On the contrary, GAINS quantifies for each station the effects of Europe-wide policy measures.

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Different locations face different challenges for attaining safe PM levels. Both the geographical origin as well as the chemical composition vary considerably. While parts of the PM problem – particularly secondary aerosol formation – are related to transboundary transport in many EU Member States, calling for synchronised EU wide action, cities also suffer from the local increment generated mainly by household heating and road traffic.

Historical trends in observed concentrations are well reproduced by the model, a prerequisite for trustworthy conclusions on the future evolution. For the future, under the assumption of successful implementation of current legislation, reductions in ambient PM₁₀ concentrations are expected and consequently a higher attainment of the PM₁₀ limit value. However, current legislation is not expected to lead to Europe wide attainment of the PM₁₀ limit value. Challenges are foreseen particularly in Eastern Europe, where widespread use of coal and inefficient wood burning in domestic heating hampers significant improvement, and in several major urban areas which suffer from increasing road traffic and stagnating household emissions. Considering that many of the remaining exceeding stations are located in densely populated areas, a significant proportion of the European population can be expected to remain exposed to PM concentrations violating EU air quality standards unless further political action is taken.

A range of technical emission control measures is readily available to decrease PM and precursor emissions beyond the baseline, as discussed by Amann et al. (2014). Exploiting the full range of emission controls available, concentrations could be decreased significantly further, and most cases of severe non-compliance persisting in 2030 could be eliminated. However, even in this scenario, safe attainment of the limit value is not achieved at all stations given uncertain meteorological conditions and possible single events. A solution could lie in the switch to cleaner fuels in domestic heating such as natural gas in Eastern European Member States.

Another challenge to safe attainment of limit values specific to urban areas is the possibly increasing burden of road dust re-suspension. Although the linear relation used in this approach is pessimistic, it seems logical that more traffic generates more

dust. A simple solution to this problem is yet to be found; targeted measures such as dust binding or enhanced road cleaning may be helpful to ensure that reductions in exhaust emissions are not compensated by increases in suspended dust.

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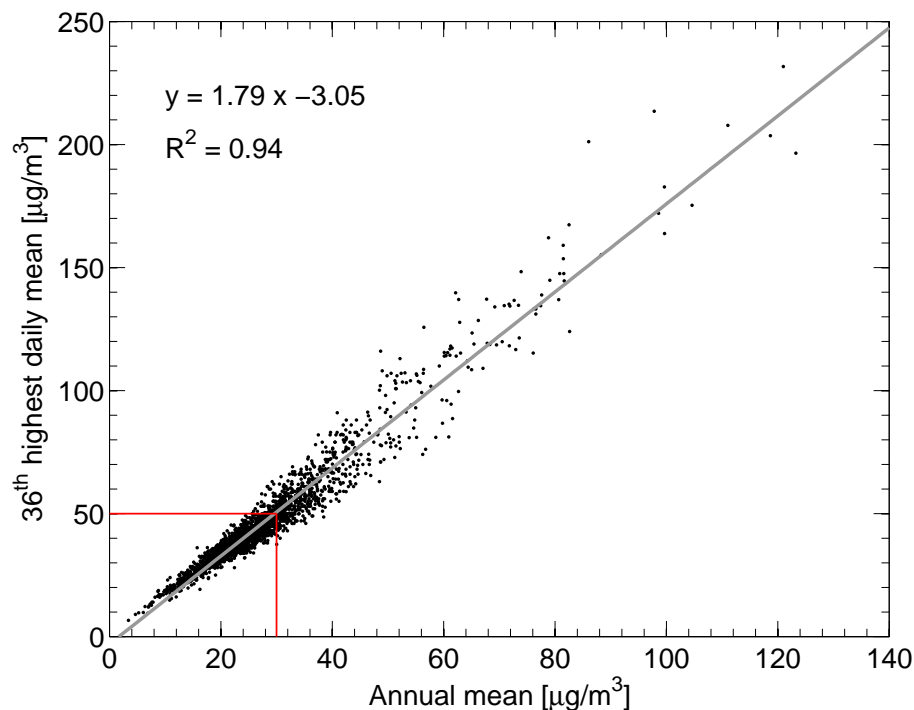


Figure 1. Relation between annual mean concentrations and the 36th highest daily average concentration in AirBase observations (data: all AirBase stations in 2009 with > 80% daily data coverage). The limit on daily exceedances of $50 \mu\text{g}/\text{m}^3$ is well represented by an annual mean limit of $30 \mu\text{g}/\text{m}^3$.

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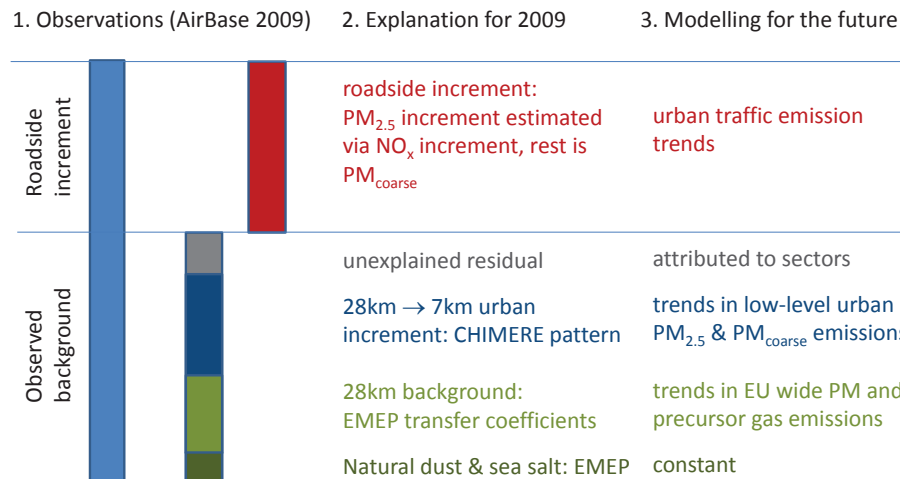


Figure 2. Schematic overview of the PM₁₀ modelling scheme for roadside stations.

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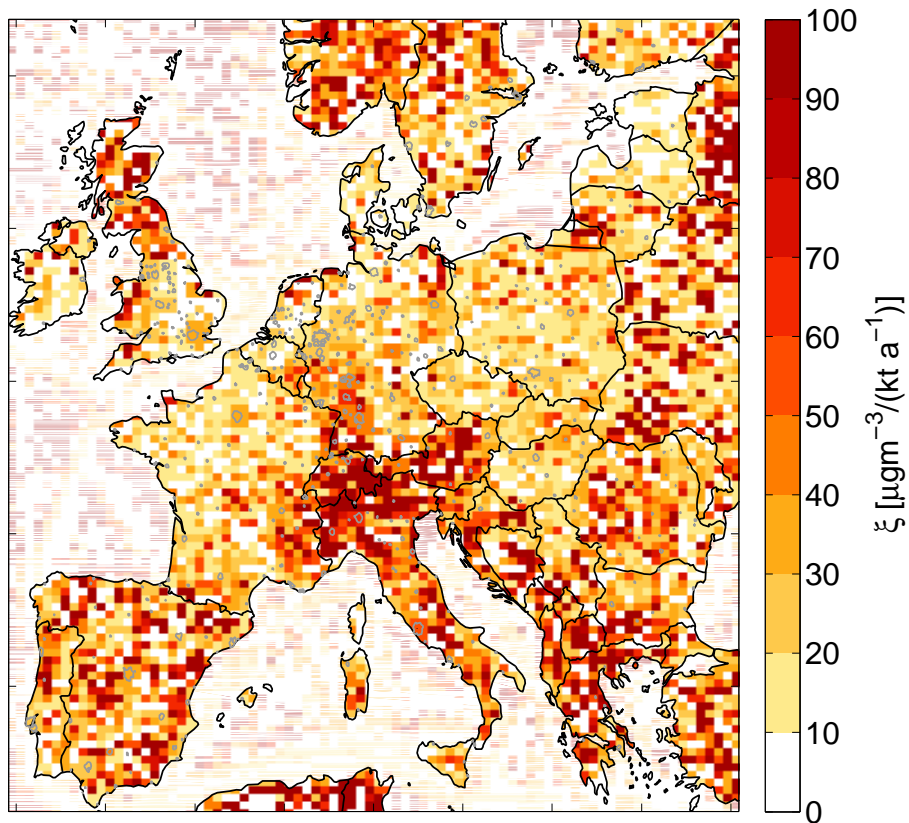


Figure 3. The regression coefficient ξ relating additional primary PM emissions within each EMEP grid cell to PM concentration increments.

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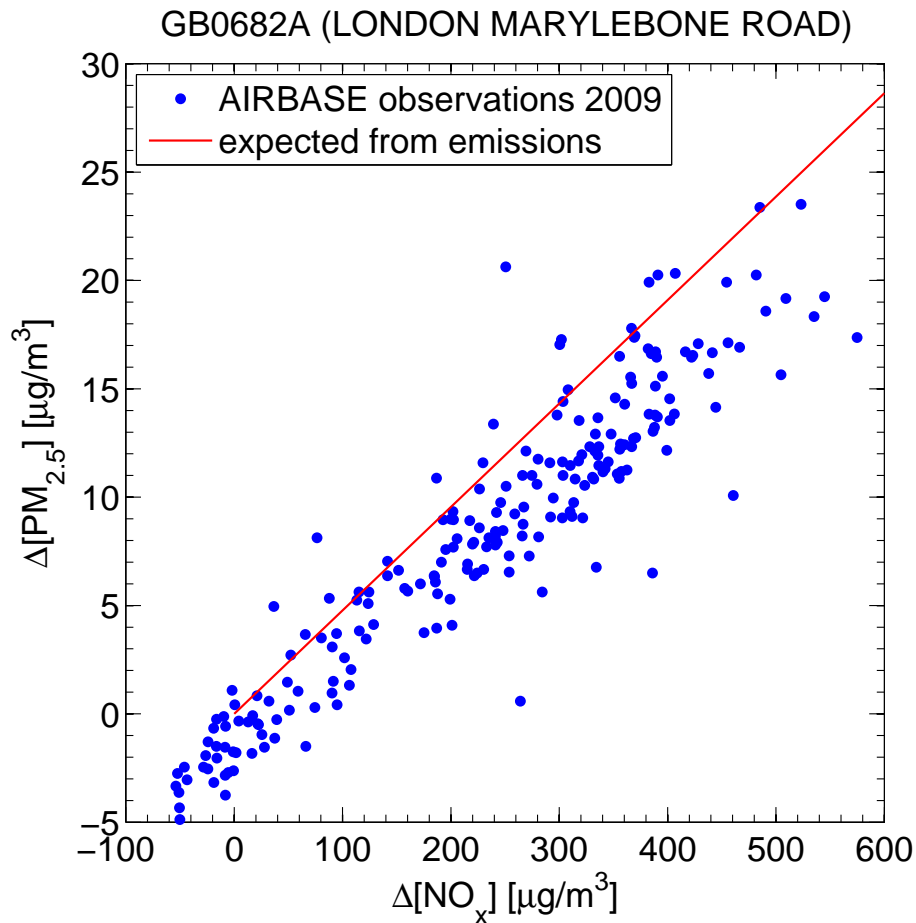


Figure 4. Roadside increments of NO_x and PM_{10} at Marylebone Road monitoring site, London: daily mean AirBase observations in 2009.

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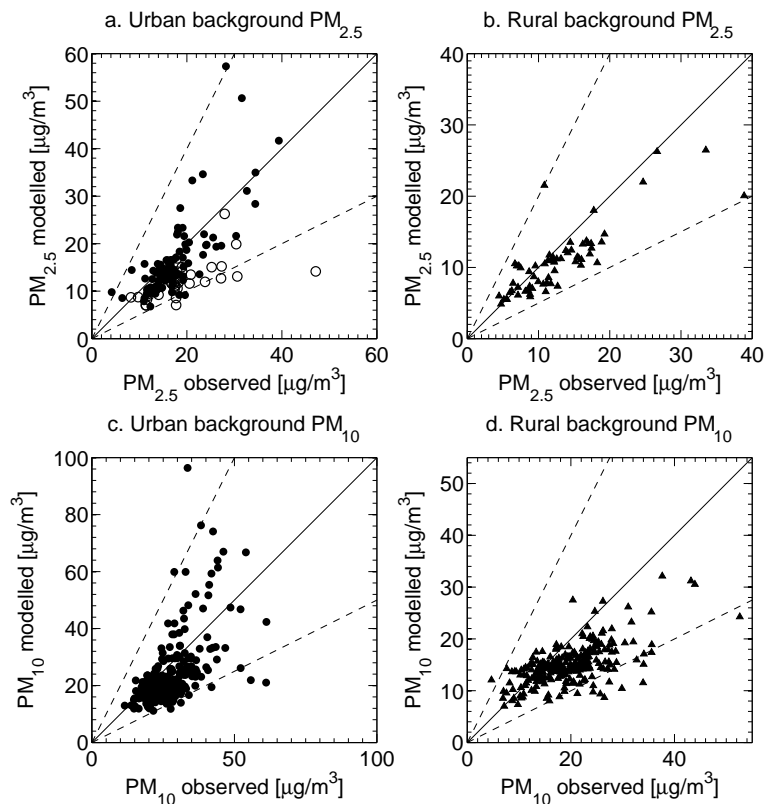


Figure 5. Bottom-up calculated vs observed PM_{2.5} and PM₁₀ concentrations at urban and rural background monitoring stations in 2009. Panel (a) distinguishes into stations located in cities > 100 000 inhabitants (dots) and those not (circles). For better viewing, only urban stations in cities > 100 000 inhabitants are shown for PM₁₀ (c).

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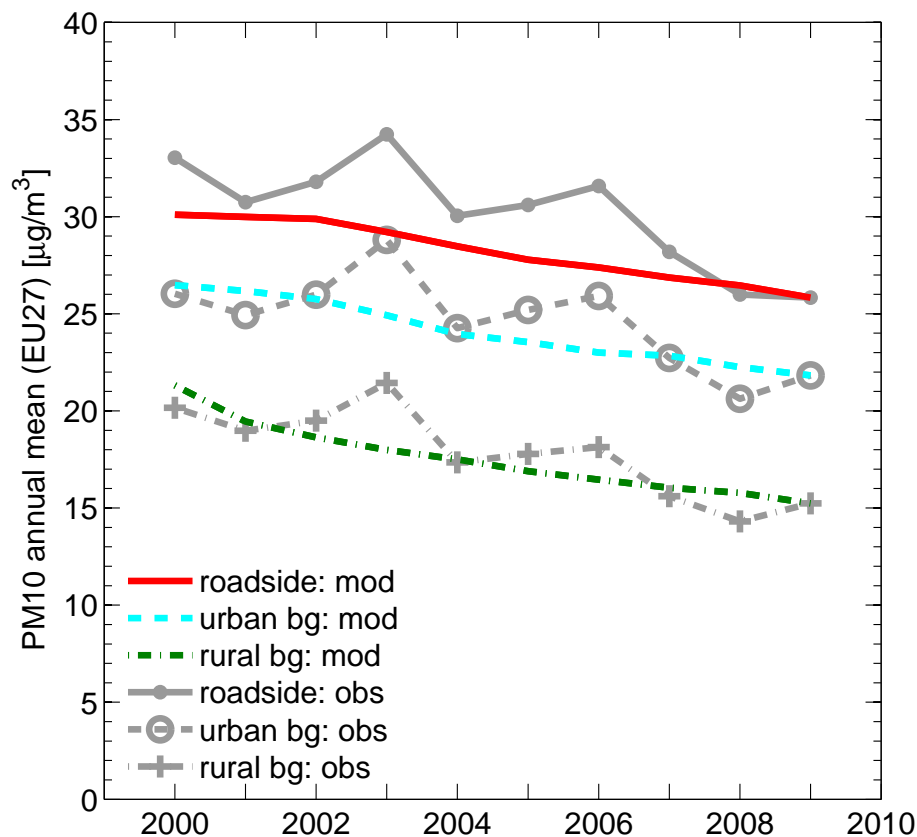


Figure 6. Time series of modelled and observed PM₁₀ averaged across different categories of monitoring stations in the EU.

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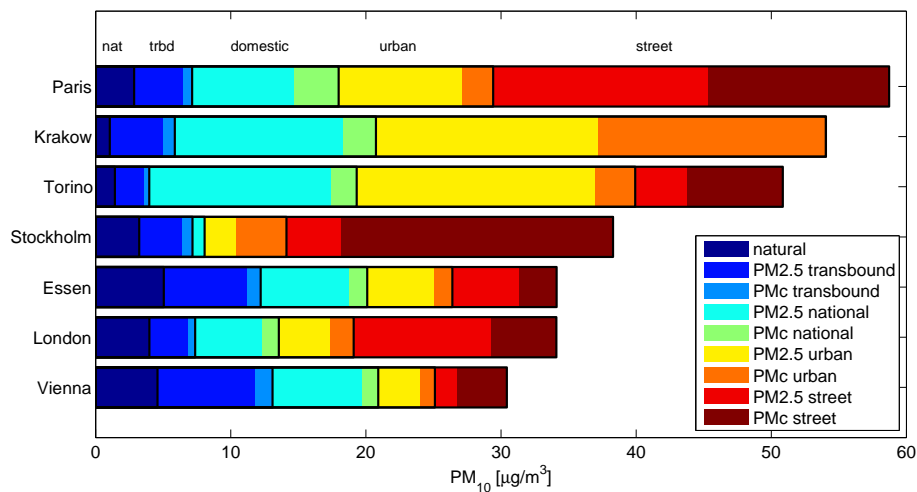


Figure 7. Modelled composition of PM₁₀ at seven monitoring stations with different characteristics in the year 2009: spatial source contributions. “nat”: natural, “trbd”: transboundary.

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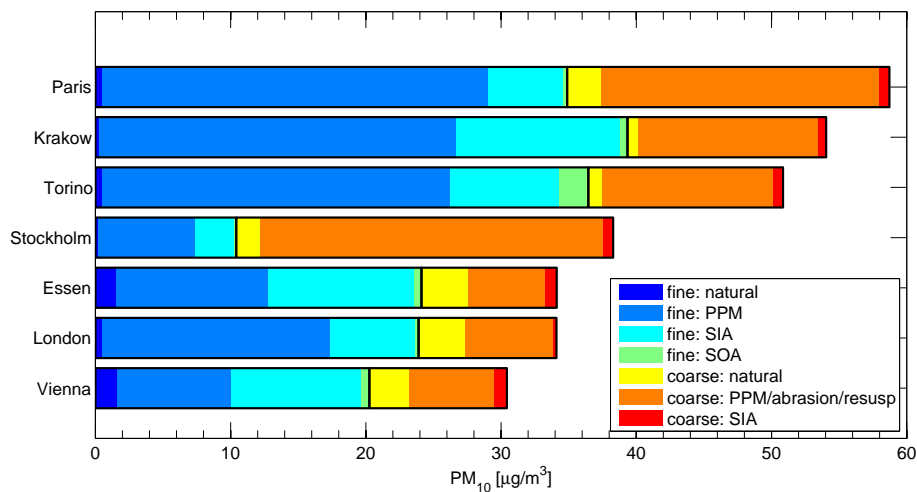


Figure 8. Modelled composition of PM₁₀ at seven monitoring stations with different characteristics in the year 2009: chemical composition.

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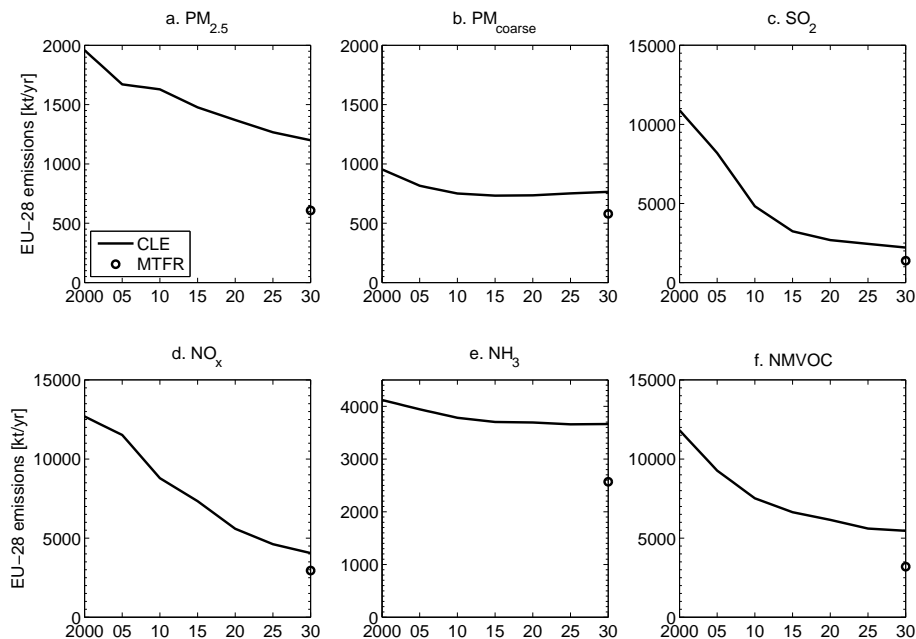


Figure 9. Bottom up calculated emissions of PM and its precursor gases in the EU-28 under current legislation (lines) and the maximum technically feasible reductions in 2030 (circles).



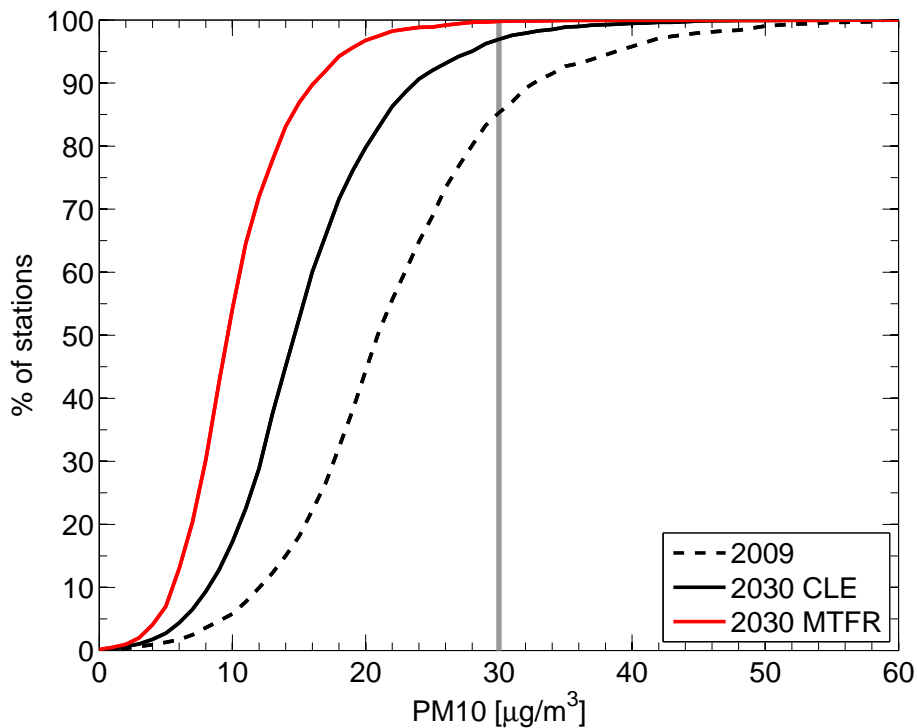


Figure 10. Cumulative distribution of PM₁₀ concentrations modelled at all stations covered in GAINS, for the base year 2009 and scenario year 2030, assuming either current legislation (CLE) or maximum technically feasible emission reductions (MTR). The equivalent annual mean limit value of 30 µg m⁻³ is indicated as grey line. Natural contributions are not included.

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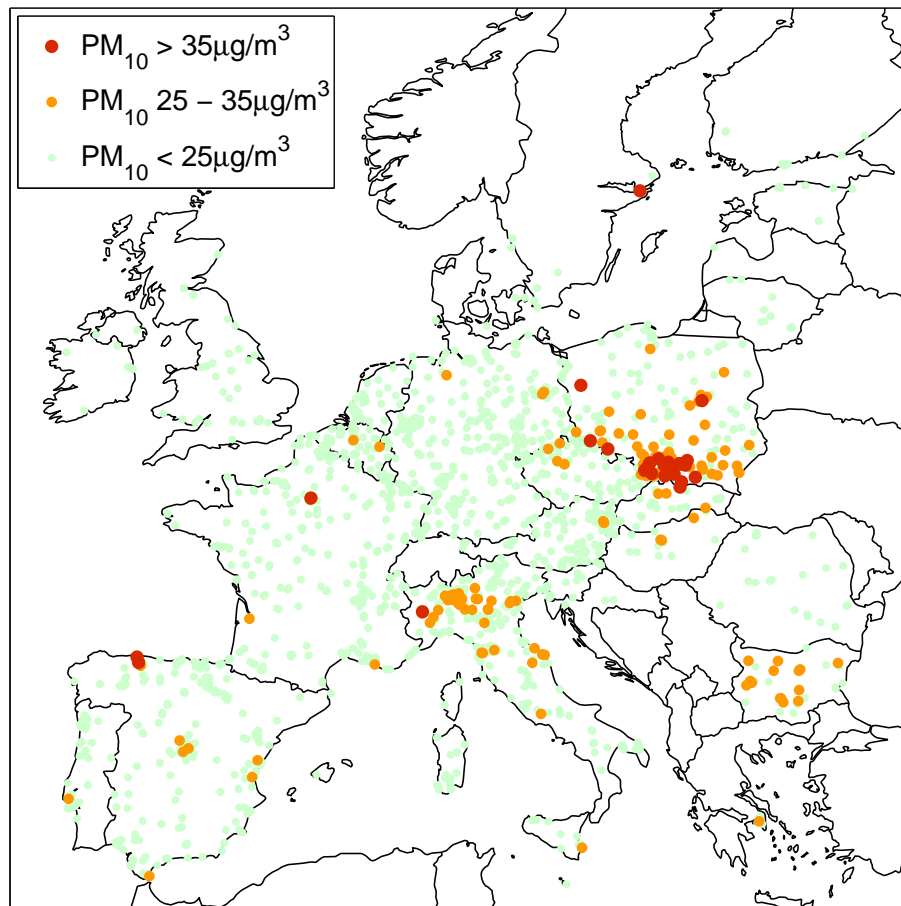


Figure 11. Modelled annual mean PM_{10} concentrations at AirBase stations for the year 2030 under the CLE scenario.

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