Service Contract on
Monitoring and Assessment
of Sectoral Implementation Actions
070307/2011/599257/SER/C3

Urban PM2.5 levels under the EU Clean Air Policy Package

TSAP Report #12

Version 1.0

Gregor Kiesewetter and Markus Amann
International Institute for Applied Systems Analysis IIASA

October 2014

The authors

This report has been produced by Gregor Kiesewetter and Markus Amann, working at the International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria.

Acknowledgements

This report was produced under the contract 'Services related to the assessment of specific emission reduction scenarios at EU and Member State level, notably reflecting national positions, the interaction with climate policy, and possible flexible implementation mechanisms', Specific Contract No. 070307/2013/666175/FRA/ENV.C.3 implementing Framework contract No ENV.C.3/FRA/2013/0013-IIASA of DG-Environment of the European Commission.

Disclaimer

The views and opinions expressed in this paper do not necessarily represent the positions of IIASA or its collaborating and supporting organizations.

The orientation and content of this report cannot be taken as indicating the position of the European Commission or its services.

Executive Summary

Fine particulate matter with a diameter below 2.5µm (PM2.5) poses a significant threat to human health. Current PM2.5 levels significantly exceed safe levels as defined by the WHO in many European cities, leading to a shortening of life expectancy of several months.

Ambient PM2.5 is composed of a variety of components, originating from local emissions as well as transboundary transport of pollution from natural as well as anthropogenic activities of many economic sectors. While primary PM is mainly emitted from traffic and household combustion, secondary PM is predominantly formed from precursor emissions from agriculture, industrial and traffic sources.

This report quantifies source contributions to urban PM2.5 levels in the EU Member States, using the station-based modelling approach that has recently been added to the GAINS modelling suite (Kiesewetter et al. 2013). For every urban roadside air quality monitoring station covered by the model, the approach disaggregates modelled PM2.5 into chemical, sectoral and spatial categories, encompassing sources of natural dust, transboundary transport of pollution, national emissions, urban emissions, and local traffic emissions.

This report compares - for Member States that have reported sufficient monitoring data to AIRBASE - the current levels of PM2.5 with the situation expected in 2030 under the Clean Air Policy Package that has been proposed by the Commission in December 2013. In many cases, the current transboundary contribution to PM2.5 already exceeds the WHO guideline values, so that uncoordinated national actions of individual Member States will not be sufficient to attain the WHO guideline value. The measures suggested by the Clean Air Policy Package would lead to a decrease of urban roadside PM2.5 concentrations by roughly a factor of two, and even more in some Member States. Most Member States would reach levels close to or even below the WHO standards of 10µg/m^3, and well below the current EU target value (which will be transformed into a limit value in 2015) of 25µg/m^3.

List of acronyms

CLE	Current legislation
EU	European Union
GAINS	Greenhouse gas - Air pollution Interactions and Synergies model
IIASA	International Institute for Applied Systems Analysis
kt	kilotons = 10^3 tons
µg/m^3	micrograms per cubic meter
NH_3	Ammonia
NH_4^+	Ammonium
NMVOC	Non-methane volatile organic compounds
NO_3^-	Nitrate
NO_x	Nitrogen oxides
PM10	Fine particles with an aerodynamic diameter of less than 10 µm
PM2.5	Fine particles with an aerodynamic diameter of less than 2.5 µm
PPM	Primary Particulate Matter
PRIMES	Energy Systems Model of the National Technical University of Athens
SNAP	Selected Nomenclature for Air Pollutants; Sector aggregation used in the CORINAIR emission inventory system
SIA	Secondary Inorganic Aerosol
SO_2	Sulphur dioxide
SO_4^{2-}	Sulphate
SOA	Secondary Organic Aerosol
TSAP	Thematic Strategy on Air Pollution
VOC	Volatile organic compounds

Table of contents

1 Introduction 4

1.1 Objective of this report 5

1.2 Structure of the report 5

1.3 Methodology 5

2 Source contributions to PM2.5 7

2.1 Spatial origin of PM2.5 at background stations 7

2.2 Sectoral-spatial source attribution of PM2.5 at urban traffic stations 8

3 Discussion and conclusions 20

More information on the Internet

More information about the GAINS methodology and interactive access to input data and results is available at the Internet at http://gains.iiasa.ac.at/TSAP. All detailed data of the scenarios presented in this report can be retrieved from the GAINS-online model (http://gains.iiasa.ac.at/gains/EUN/index.login?logout=1). The situation for 2009 is modelled using interpolated emissions from the scenario "PRIMES 2013 REF CLE" (in scenario group "TSAP_Dec_2013"). The 2030 Commission Proposal is listed in GAINS online as "B7 2030-Commission Proposal" and described in detail in Amann et al. 2014.

The graphs presented in this paper can also downloaded as a Powerpoint presentation at http://www.iiasa.ac.at/web/home/research/researchPrograms/MitigationofAirPollutionandGreenhousegases/TSAP-reports.en.html.

1 Introduction

Exposure to fine particulate matter has been shown to have significant negative impacts to human health (Pope III et al. 2002; 2011; WHO 2013). The Air Quality Directive (2008/50/EC) of the European Union (EC 2008) specifies limit values for ambient PM10, i.e., particulate matter with an aerodynamic diameter <10µm. Recent epidemiological evidence suggests health impacts to be strongly associated with PM2.5 i.e., particles with an aerodynamic diameter <2.5µm (e.g. WHO 2013). For PM2.5, the current Air Quality Directive specifies a target value of 25µg/m^3 annual mean concentration, which will be transformed into a legally binding limit value as of January 2015. This value is, however, by far higher than the levels considered safe by the World Health Organization (WHO), who specifies a guideline value of 10µg/m^3 annual mean concentration (World Health Organization 2006). For comparison, in a recent review of US air quality legislation, the annual PM2.5 standard in the United States has been tightened to 12µg/m^3 (US-EPA 2013).

While compliance with the current EU PM2.5 target value may be higher than with the PM10 air quality limit values, several monitoring sites are still exceeding the forthcoming PM2.5 limit value, and only few stations currently comply with the WHO guideline value. Particularly inner urban concentrations of PM2.5 are far above the WHO guideline value.

Health impacts from current levels of PM2.5 are considerable. GAINS estimates that in 2005 European average statistical life expectancy was shortened by 8.5 months due to the exposure to fine particulate matter (Amann et al. 2014). In the light of the objectives of the EU's Environment Action Programme, i.e., to achieve 'levels of air quality that do not give rise to significant negative impacts on, and risks to human health and environment', further action to reduce ambient PM2.5 will be necessary.

The origin of the PM2.5 problem is multi-faceted, with several spatial scales, economic sectors, and pollutants involved. Ambient PM2.5 does not only consist of primary particles but also contains natural components such as dust and sea salt, and a significant fraction of secondary particles formed in the atmosphere from precursor gases through well-known chemical reactions. The main compounds involved in the formation of secondary inorganic PM (or aerosol, SIA) are ammonium (NH_4^+), nitrate (NO_3^-) and sulphate (SO_4^{2-}), which are chemical products of the precursor gases ammonia (NH_3), nitrogen oxides (NO_x), and sulphur dioxide (SO_2). While, due to its low molecular weight, ammonium itself contributes only little to total PM mass, it is crucial for the formation of both main secondary inorganic PM species, ammonium sulphate ($(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3). Consequently, NH_3 emission reductions would have a significant effect on European PM2.5 levels (Beauchamp et al. 2013). Further contributions to ambient PM are made by secondary organic PM (or aerosol, SOA), which is formed from anthropogenic and natural emissions of non-methane volatile organic compounds (NMVOCs).

As noted above, different spatial scales are involved, from Europe-wide transboundary transport of pollution to local emissions within an urban street canyon. The importance of long range transport of PM in Europe has repeatedly been emphasised (Makra et al. 2011; Tsyro 2008; ApSimon et al. 2001; Malcolm et al. 2000). In particular the secondary aerosol formation from gaseous precursors introduces a strong international transboundary facet (Malcolm et al. 2000).

Figure 1.1 shows the concept of aggregation of contributions from different spatial scales to the local ambient levels inside a city. In addition to a natural background and transboundary transport of pollution, also emissions from distant sources located in the same country contribute to the regional background. Inside the city, a sharp increment is observed, caused mainly by low level emissions of primary PM, typically road transport and domestic heating. On top of the urban background, local traffic emissions lead to further concentration increases within individual street canyons. Due to this build-up of concentrations, urban roadside monitoring stations typically report the highest ambient concentrations in a given country.

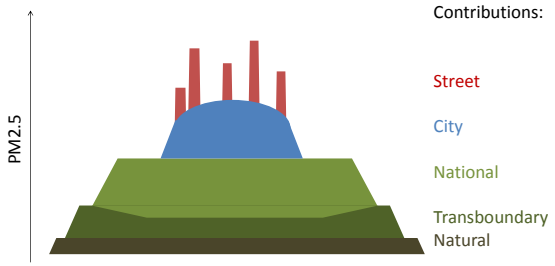

Figure 1.1 Build-up of PM2.5 contributions from different geographical origins.

Sources of PM and PM precursor gases are found throughout all economic sectors. Primary PM (PPM) is mainly emitted from combustion processes such as household heating, industrial combustion, or diesel engines. Unit emissions vary strongly across technologies and fuels used in each sector; e.g., PM emissions from gasoline cars are much lower than those from diesel engines, and natural gas heating produces much lower PM emissions than coal stoves. Of the precursor substances for secondary aerosol, NH_3 is almost exclusively emitted from agricultural processes (both animal husbandry as well as fertilizing of fields), while SO_2 is predominantly generated in the energy producing industry, and NO_x emissions are spread between the transportation, industry, and household combustion sectors.

Recently the European Commission has proposed a Clean Air Policy Package with the aim to further reduce the impacts of harmful emissions from industry, traffic, energy plants and agriculture on human health and the environment (EC 2013). The package includes a new Clean Air Programme for Europe with measures to ensure that existing targets are met in the short term, and new air quality objectives for the period up to 2030. The package also proposes a revised Directive on National Emission Ceilings with stricter national emission ceilings for the six main pollutants, as well as a new Directive to reduce pollution from medium-sized combustion installations.

The proposal of the European Commission has been informed by quantitative modelling of baseline emissions and associated impacts, of the scope for further emission reduction options, and of cost-effective emission reduction strategies with the GAINS Integrated Assessment Modelling suite by the International Institute for Applied Systems Analysis (IIASA). An assessment of the effects of the proposal on air quality in Europe has been provided in the TSAP Report #11 (Amann et al. 2014).

The GAINS model includes all economic sectors, chemical components, and spatial scales necessary for a comprehensive treatment of PM2.5 (Amann et al. 2011). Recently a downscaling methodology has been introduced which allows for the assessment of PM at individual air quality monitoring stations (Kiesewetter et al. 2013). The TSAP Report #11 included a quantification of health impacts due to PM2.5, and estimates on the attainment of PM10 limit values. PM2.5 concentrations have so far not been discussed explicitly. However, owing to the structure of the GAINS model, contributions to PM2.5 at each station are modelled individually and hence are traceable.

1.1 Objective of this report

This report presents an assessment of source contributions to PM2.5 levels at European monitoring stations. The present situation (based on the year 2009) is compared with the envisaged result of the Commission proposal for 2030 as discussed in TSAP Report #11 (Amann et al. 2014). We here focus on the attribution of PM2.5 concentrations at urban roadside stations to the source sectors and spatial contributions.

1.2 Structure of the report

The remainder of Section 1 provides a brief summary of the methodology. Section 2 presents the source allocation of PM2.5 concentrations in the European background and at urban traffic monitoring stations. Results are discussed and conclusions are drawn in Section 3. A more detailed technical explanation of refinements to the methodology is given in the Technical Appendix.

1.3 Methodology

This report employs the GAINS model system developed under the EC4MACS (European Consortium for Modelling of Air pollution and Climate Strategies) project, which was funded under the EU LIFE programme (www.ec4macs.eu). In particular, we use the station based methodology described by Kiesewetter et al. (2013; 2014a; 2014b), and apply it to PM2.5 levels at urban traffic stations.

A schematic overview is shown in Figure 1.2. The analysis combines bottom up emission modelling in GAINS, simplified atmospheric chemistry and dispersion calculations, and observations reported to the AirBase database of the European Environment Agency (EEA).

Measured urban background concentrations are explained to the extent possible with a linearised atmospheric dispersion scheme based on source-receptor calculations with the EMEP model (Simpson et al. 2012) at a resolution of 0.5°×0.25° (approx. 28×28km^2), and a more detailed, nested simulation with the CHIMERE CTM model (Menut et al. 2013) at a resolution of approx. 7×7km^2.

In order to provide for a good coverage of stations throughout the EU, the analysis is done not only for stations reporting PM2.5 in 2009, but includes all stations reporting PM10 and fulfilling the data coverage criteria to be covered by GAINS. The PM2.5 urban background concentration, either observed directly or estimated from observed PM10, is split into a regional component and an urban increment, based on the ratios of the 28×28km^2 concentrations modelled with the EMEP model and the 28→7km scale increments produced with the CHIMERE CTM model. Thereby, unexplained residuals between bottom-up modelled and measured urban background concentrations have been allocated proportional to the explained fractions, i.e., to their likely spatial and sectoral sources.

At roadside stations, the additional PM2.5 increment is derived from comparisons with related background observations or, if unavailable, calculated from scaling the observed NO_x roadside increment with the appropriate ratio in emissions.

Altogether, 1875 PM monitoring stations of all types are covered in GAINS. This analysis is based on 297 urban traffic stations that provide sufficient monitoring detail to be included in GAINS (i.e. PM2.5, PM10, NO_x, etc), located in 21 Member States. For the other Member States, insufficient data were available in 2009 to allow for the modelling of roadside stations. The full list of data requirement criteria is available in TSAP Report #9 (Kiesewetter et al. 2013).

A detailed explanation of the post-processing analysis is provided in the Technical Appendix.

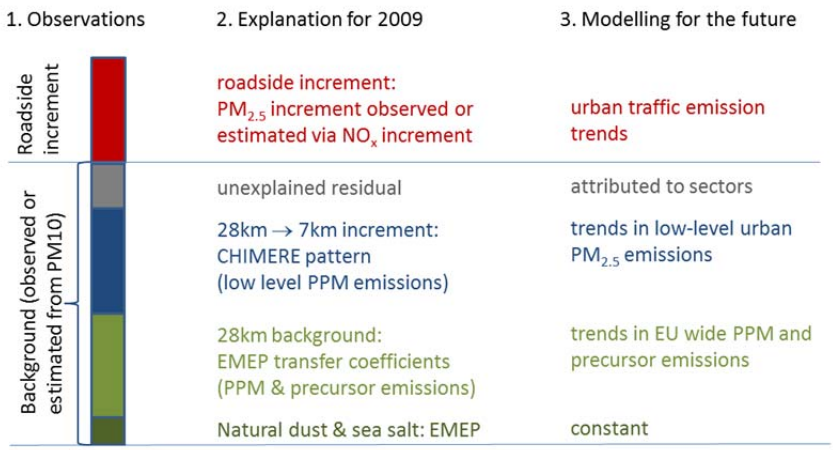

Figure 1.2. Overview of the PM2.5 station modelling scheme used in GAINS.

2 Source contributions to PM2.5

As described above, the source allocation calculations distinguish a large number of source sectors and countries of origin. However, for easier interpretation, the presentation in this report employs a more aggregated display:

Spatial origin:
Graphs distinguish contributions from transboundary transport of pollution, national emission sources (outside a city), emissions within the city, and traffic sources within the particular street canyon.

Sectoral contributions:

PPM is explicitly shown from industry (including energy industry, industrial combustion, industrial processes, solvent use, extraction and distribution of fuels, waste management) and traffic (road and non-road) sources.

Secondary aerosol is split into contributions involving industrial (SO_2 and NO_x) emissions and those involving traffic emissions (NO_x). Both of these components combine with ammonia from the only source agriculture, hence these contributions are attributed to "industrial + agriculture" and "traffic + agriculture" emissions respectively, to indicate the different sectors involved

Contributions from the domestic sector (mainly household heating) are shown as totals, including primary and secondary particles.

2.1 Spatial origin of PM2.5 at background stations

As an overview of the geographical origins of PM2.5 at rural and urban background stations, Figure 2.1 shows the modelled relative spatial source contributions to ambient PM2.5 at all stations classified as background stations in AirBase. The figures distinguish the four spatial source categories considered in this report: Natural (A), transboundary (B), national (C), and urban (D). Category C "national" contains contributions from emissions in the same country where the station is located, but excludes any local urban increment, which is shown in category D. Natural, international and national contributions together make up the regional background, i.e., the observed PM2.5 at rural background stations. The difference to PM observed at urban background stations is assumed to originate from urban emission sources, and is displayed in the category "urban".

Note that the percentages are relative to total PM2.5 in 2009, i.e., to observed PM2.5 wherever available, or estimated from observed PM10 otherwise (see Technical Appendix for details).

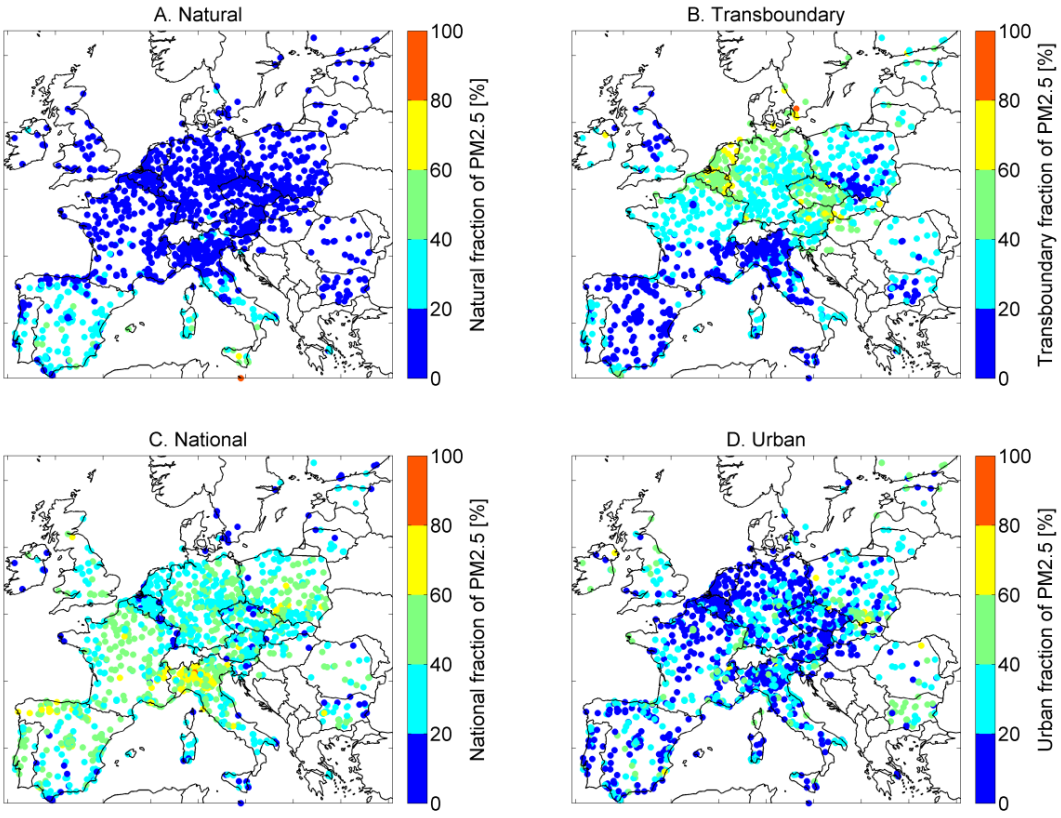

Figure 2.1. Spatial origins of PM2.5 at background monitoring stations covered by GAINS, shown as relative fractions of total modelled PM2.5 at each station in 2009.

2.2 Sectoral-spatial source attribution of PM2.5 at urban traffic stations

This section provides for each EU Member State with PM monitoring data from urban roadside stations reported in the AIRBASE database the allocation of observed PM2.5 concentrations into source contributions according to their spatial origin and chemical-sectoral composition as described in Section 1.3.

Graphs present averages for all modelled stations that are classified as urban traffic sites in AirBase and covered by the modelling scheme (see Kiesewetter et al., 2013 for details). The focus on urban traffic stations allows for the full spatial range of source contributions to be quantified, from long-range transboundary transport to local traffic emissions in the particular street canyon where a monitoring station is located.

However, some Member States provide monitoring data for only a few urban traffic stations, while many more urban background stations are reported. To increase the representativeness of the analysis in cases where only few traffic stations are reported, the average roadside increment calculated for the available roadside stations is applied also to the other stations for which no roadside increment could be derived from the observations. E.g., Poland reported for 2009 only three roadside monitoring stations with sufficient detail for the GAINS modelling, while observations for 142 urban background stations are provided.

The following pages contain figures showing source contributions, in alphabetical order of Member States. The spatial categories used in the plots are the same as in Figure 2.1, with the additional category of local traffic contributions from within the street canyon. For comparison, the WHO guideline value of 10µg/m^3 is shown.

Austria (29 stations)

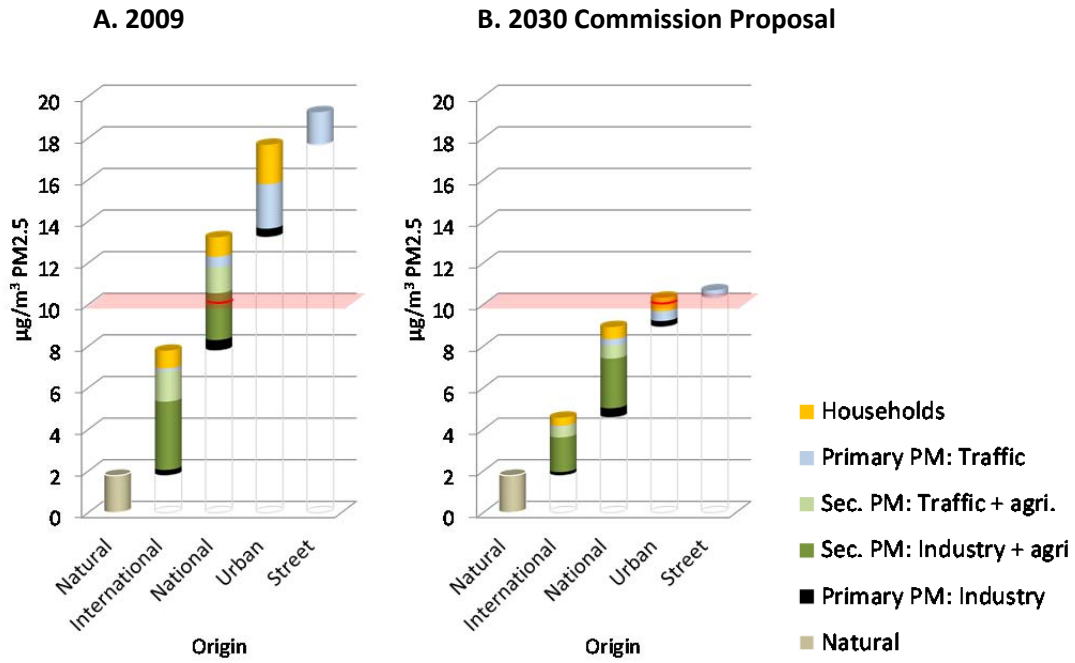

Figure 2.2. Source contributions to ambient PM2.5 at urban traffic stations in Austria (, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Belgium (4 stations)

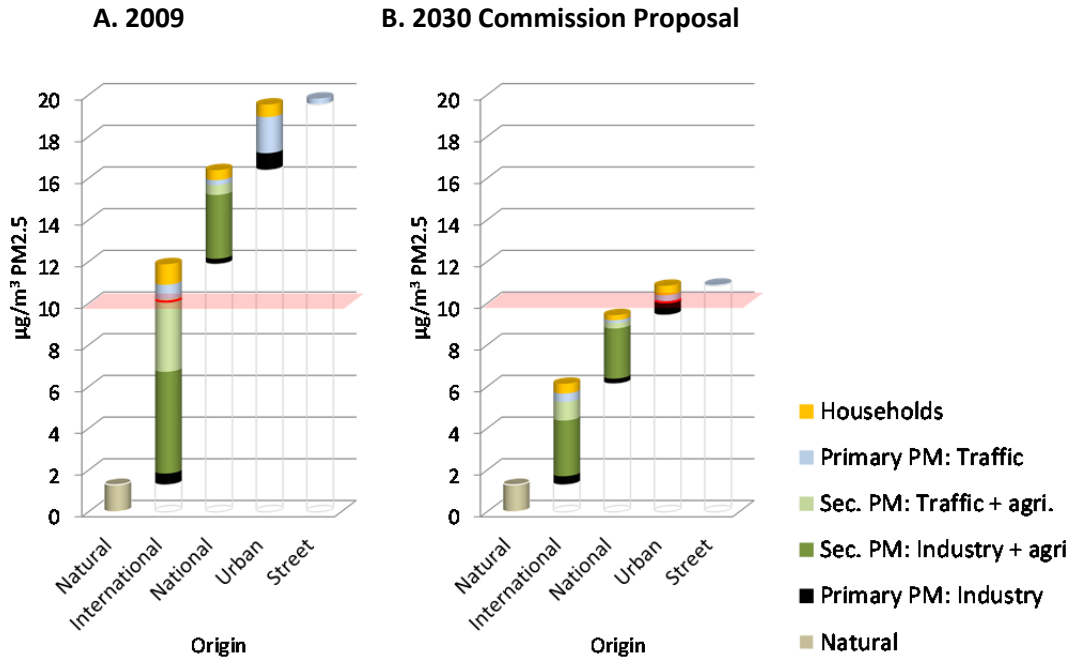

Figure 2.3. Source contributions to ambient PM2.5 at urban traffic stations in Belgium, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Bulgaria (14 stations

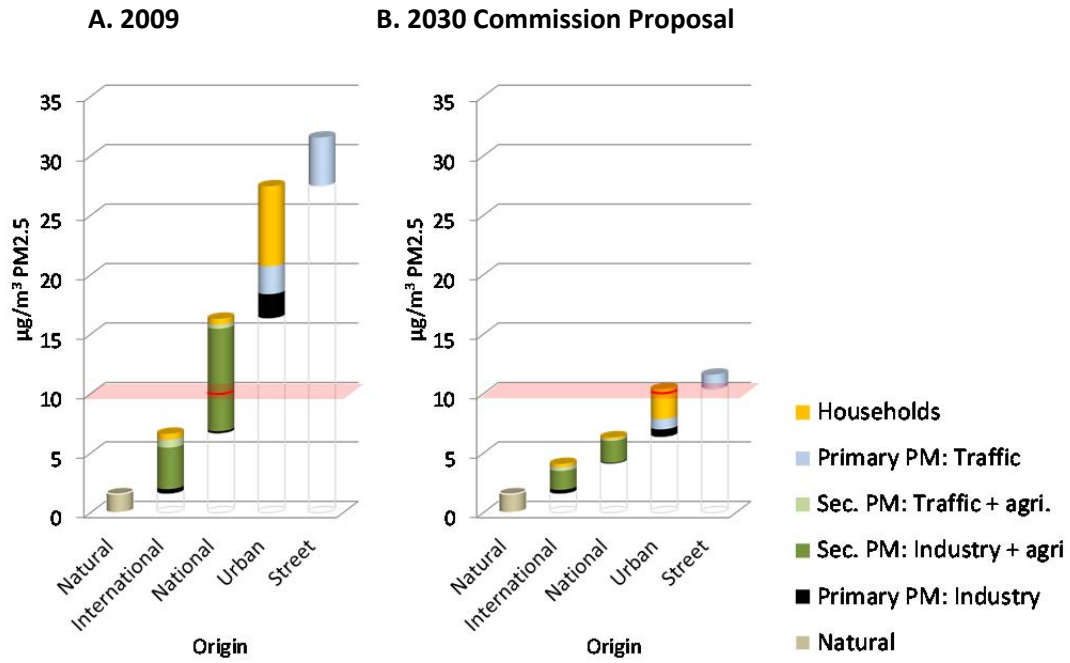

Figure 2.4. Source contributions to ambient PM2.5 at urban traffic stations in Bulgaria, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Czech Republic (33 stations)

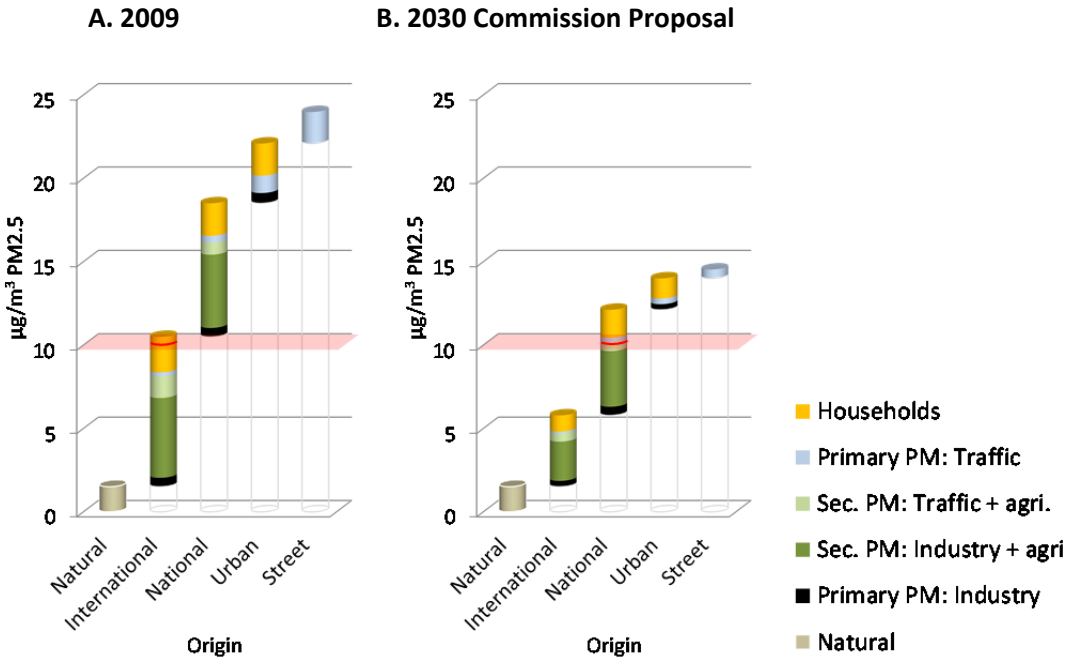

Figure 2.5. Source contributions to ambient PM2.5 at urban traffic stations in the Czech Republic, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Estonia (3 stations)

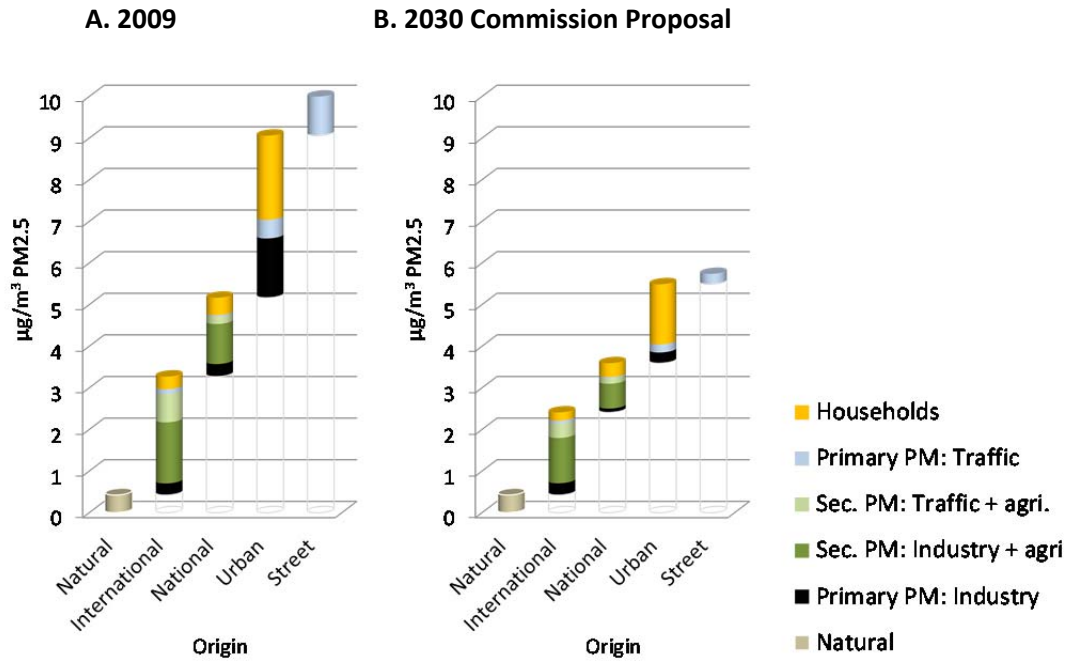

Figure 2.6. Source contributions to ambient PM2.5 at urban traffic stations in Estonia, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Finland (2 stations)

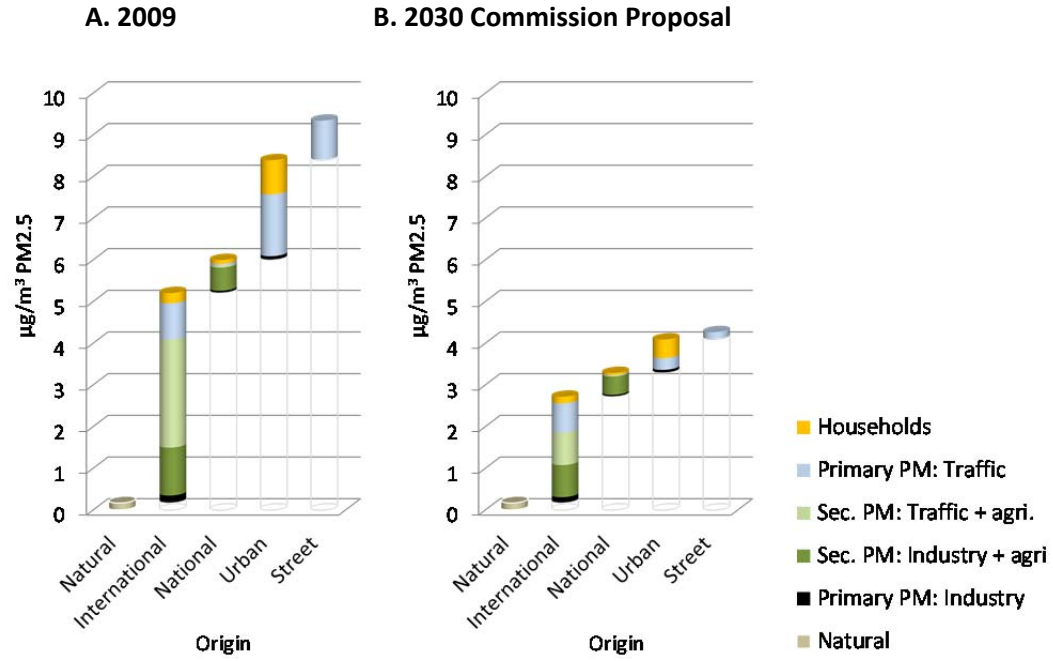

Figure 2.7. Source contributions to ambient PM2.5 at urban traffic stations in Finland, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

France (29 stations)

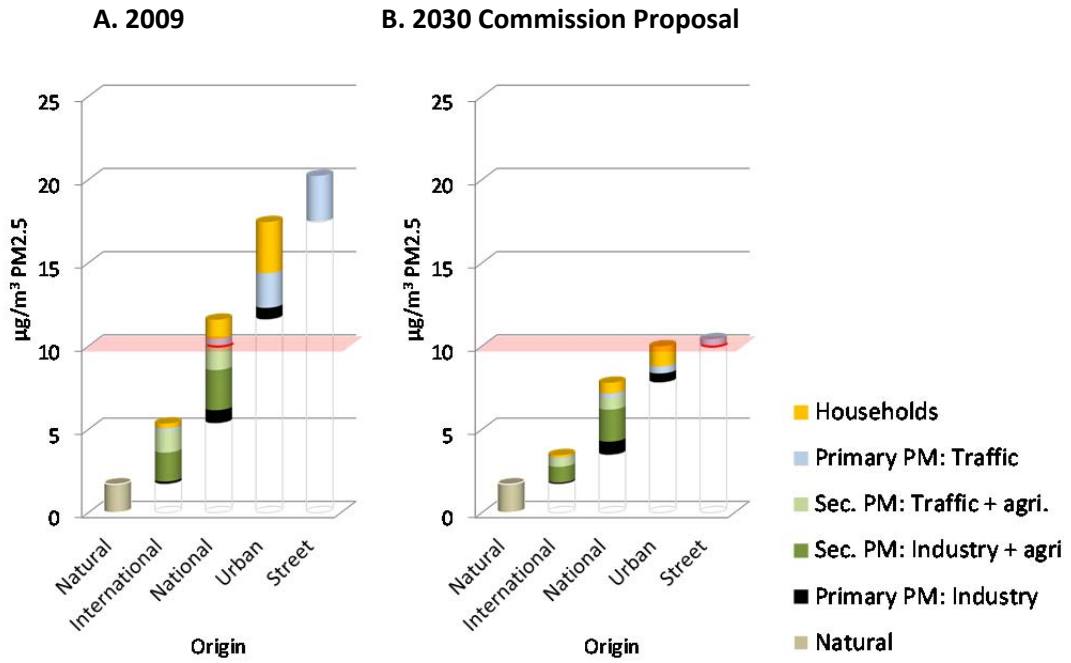

Figure 2.8. Source contributions to ambient PM2.5 at urban traffic stations in France, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Germany (79 stations)

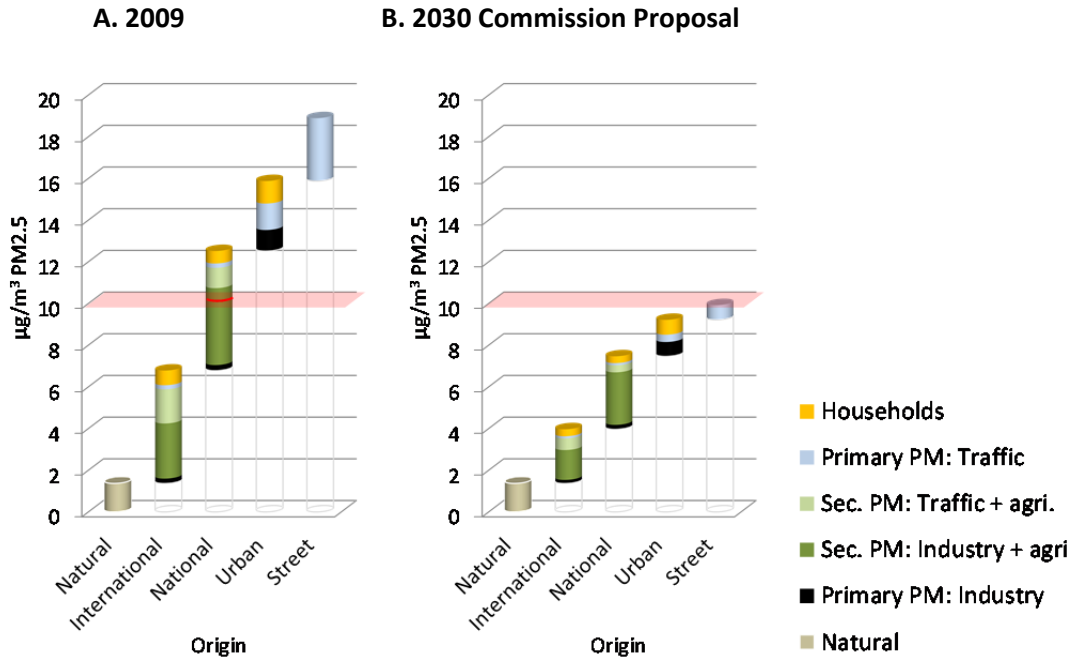

Figure 2.9. Source contributions to ambient PM2.5 at urban traffic stations in Germany, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Greece (3 stations)

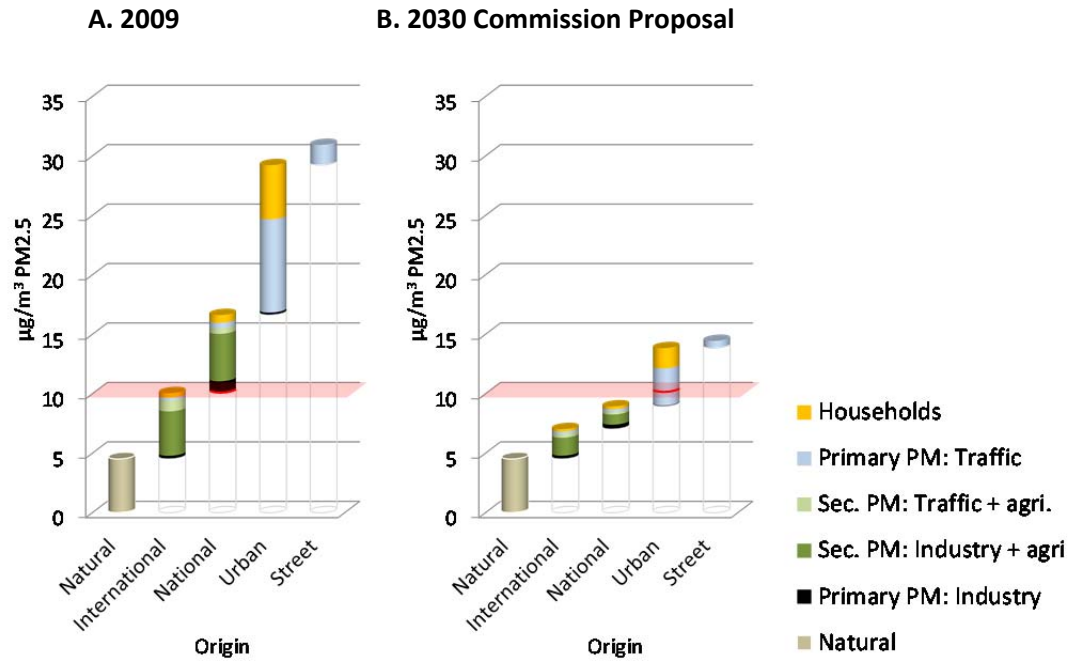

Figure 2.10. Source contributions to ambient PM2.5 at urban traffic stations in Greece, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Hungary (6 stations)

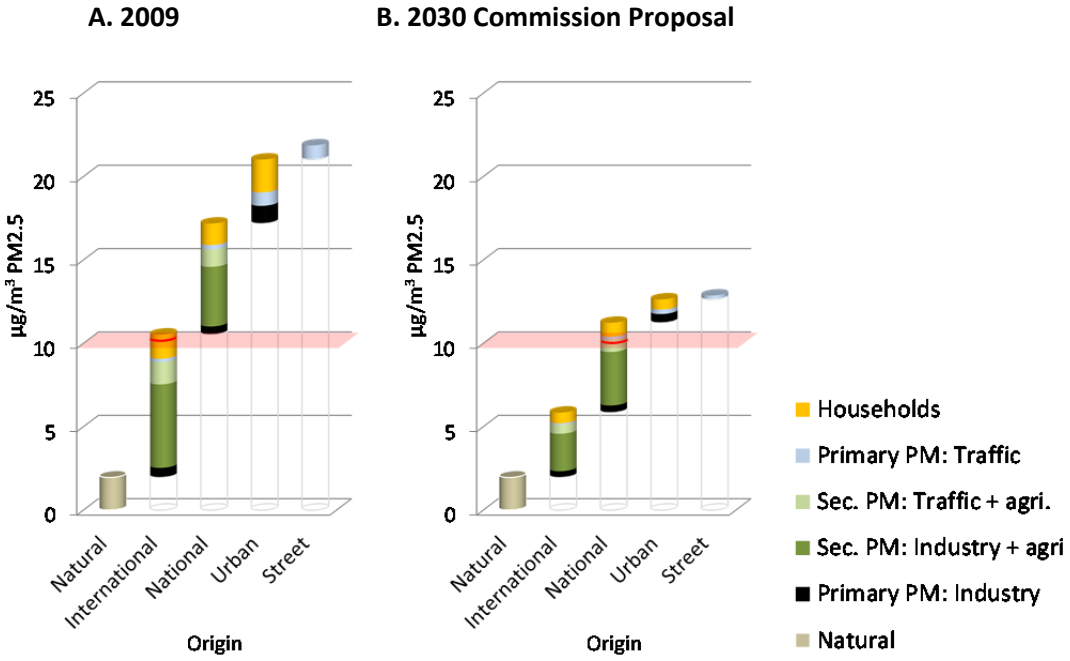

Figure 2.11. Source contributions to ambient PM2.5 at urban traffic stations in Hungary, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Ireland (2 stations)

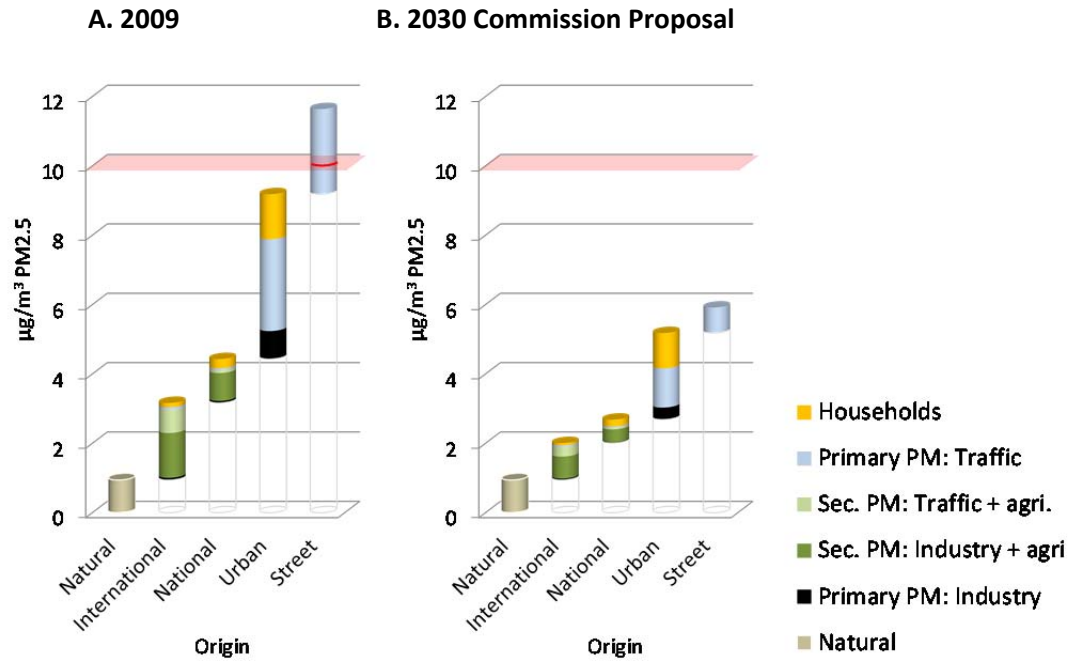

Figure 2.12. Source contributions to ambient PM2.5 at urban traffic stations in Ireland, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Italy (70 stations)

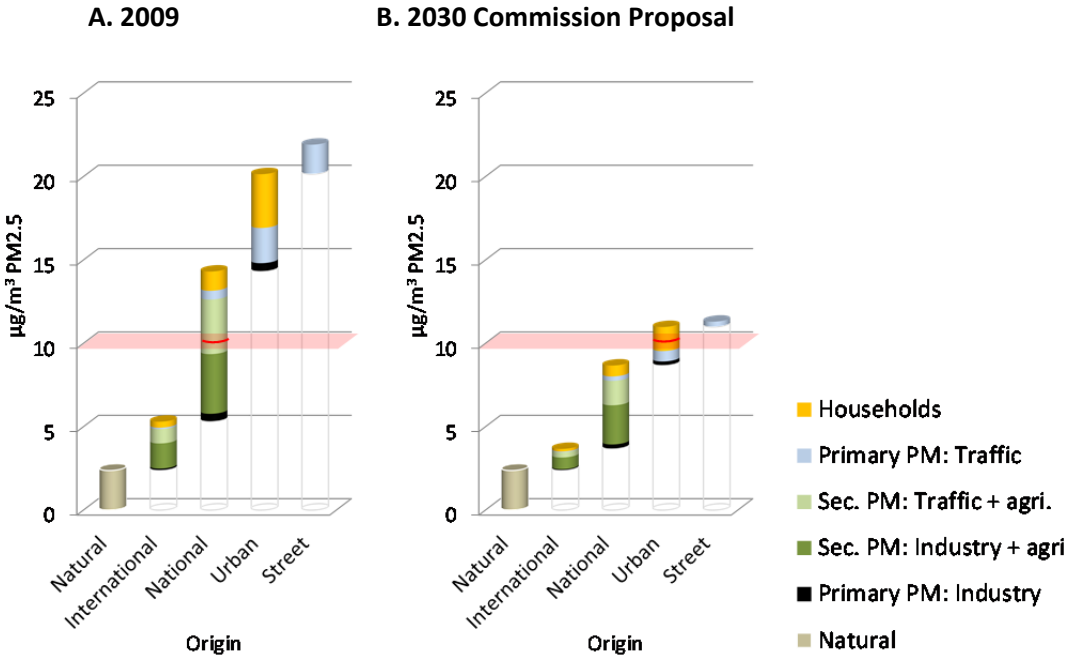

Figure 2.13. Source contributions to ambient PM2.5 at urban traffic stations in Italy, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Lithuania (5 stations)

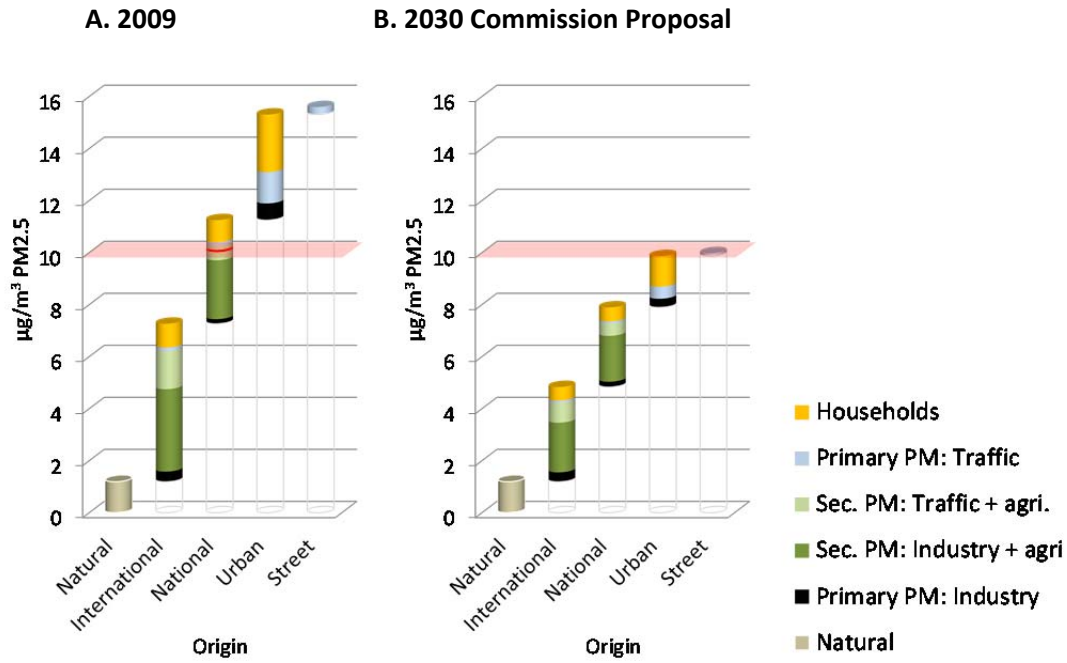

Figure 2.14. Source contributions to ambient PM2.5 at urban stations in Lithuania, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS. As Lithuania has not provided sufficient details for traffic stations in 2009, the street component could not be properly estimated.

Netherlands (5 stations)

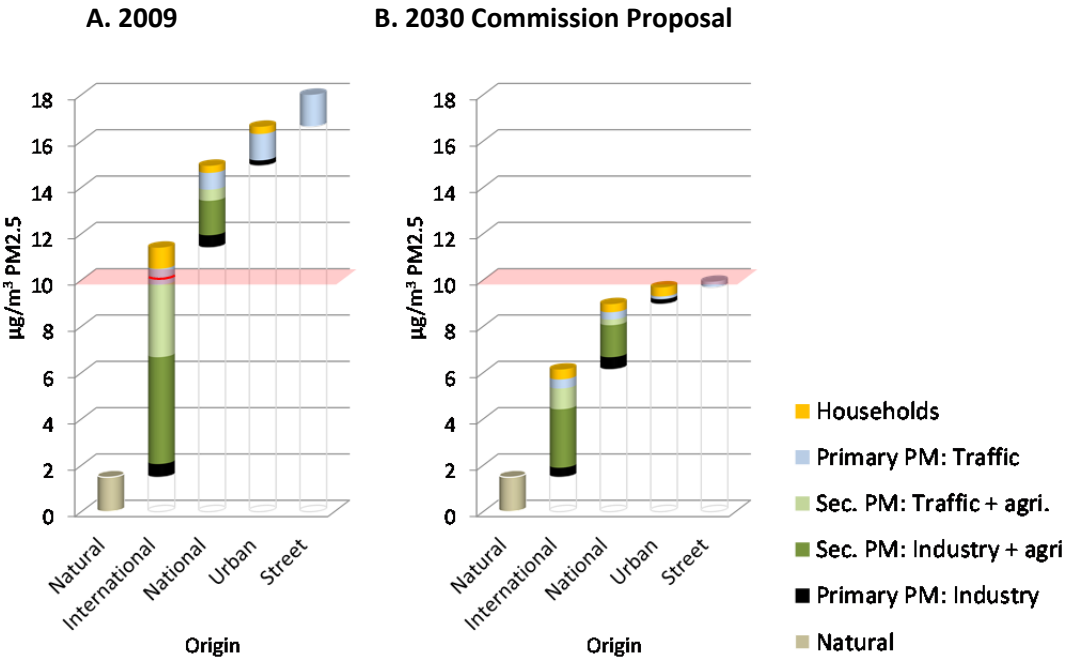

Figure 2.15. Source contributions to ambient PM2.5 at urban traffic stations in the Netherlands, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Poland (142 stations)

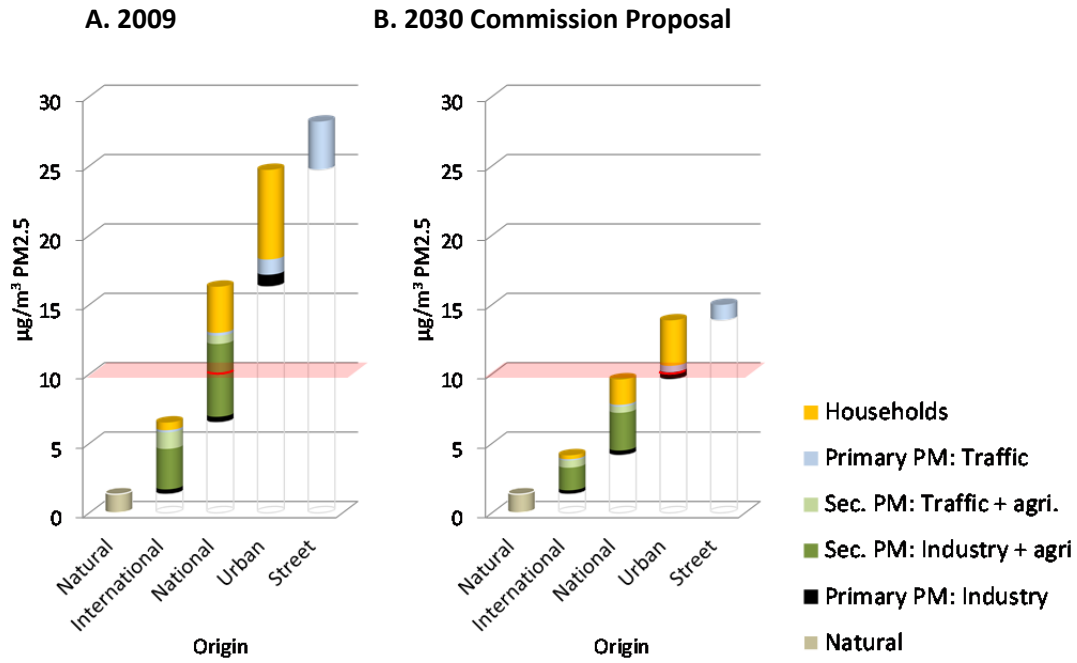

Figure 2.16. Source contributions to ambient PM2.5 at urban traffic stations in Poland, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Portugal (15 stations)

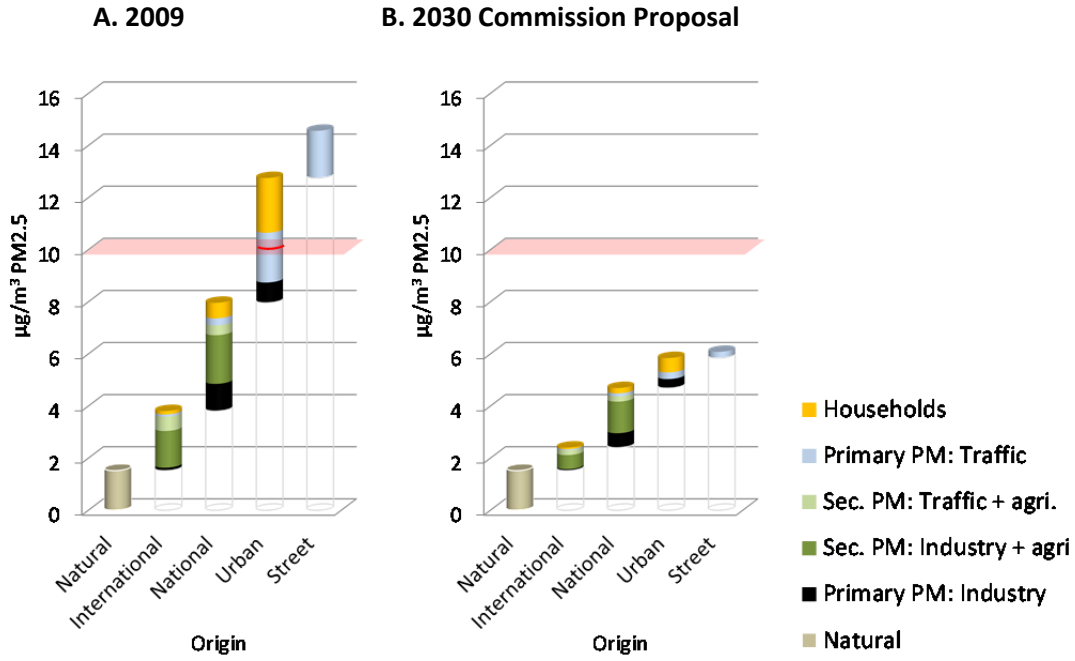

Figure 2.17. Source contributions to ambient PM2.5 at urban traffic stations in Portugal, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Romania (2 stations)

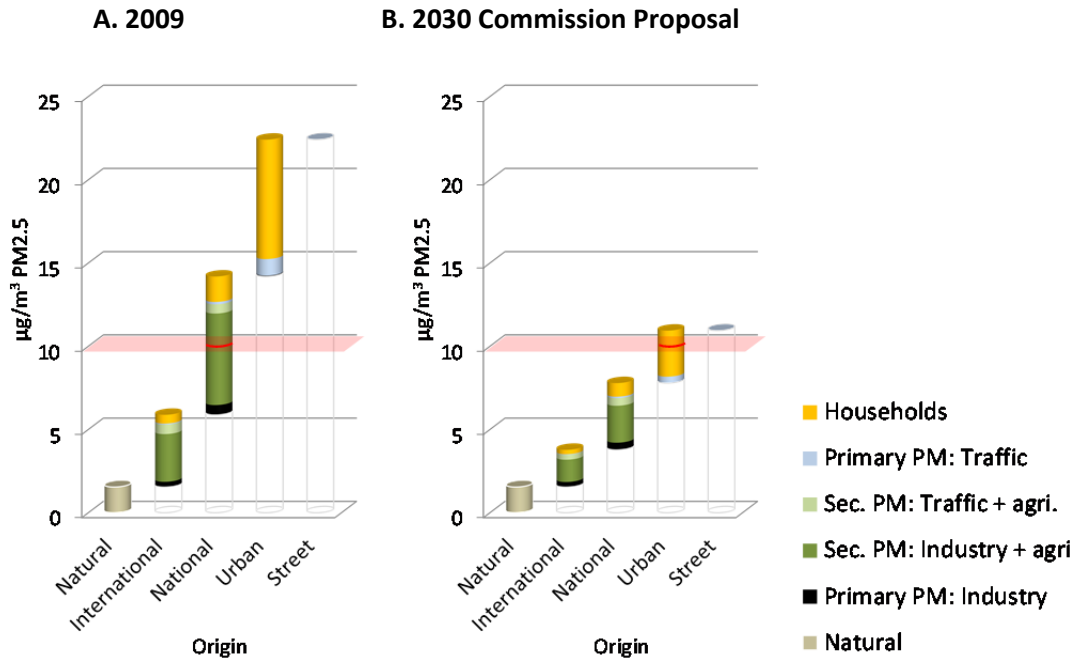

Figure 2.18. Source contributions to ambient PM2.5 at urban stations in Romania, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS. As Romania has not provided sufficient details for traffic stations in 2009, street components could not be estimated.

Slovakia (17 stations)

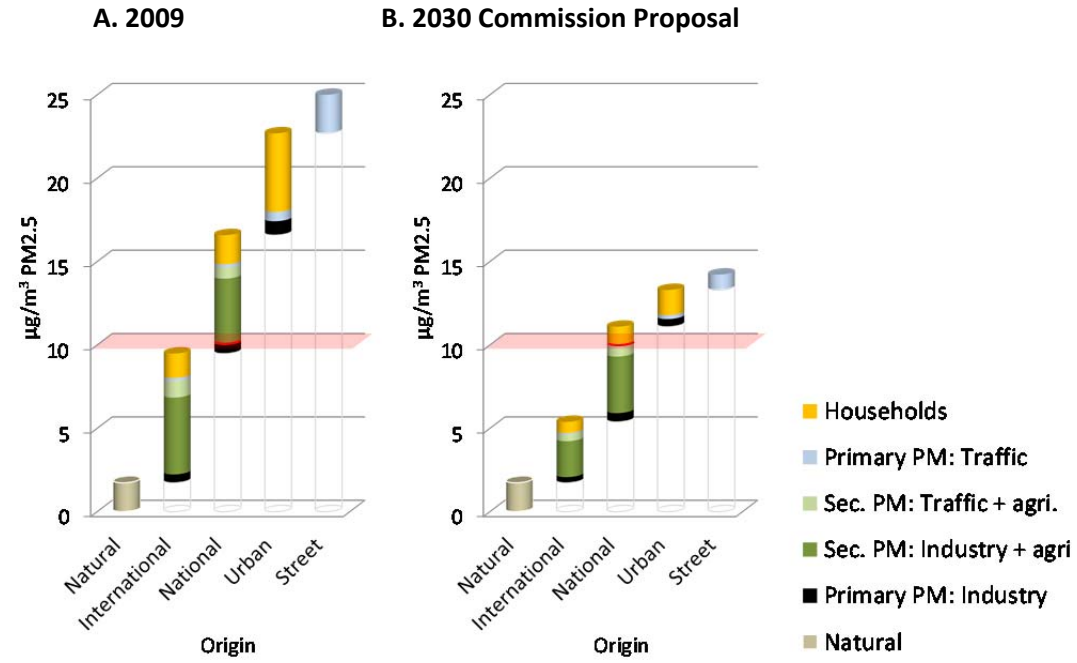

Figure 2.19. Source contributions to ambient PM2.5 at urban traffic stations in Slovakia, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Spain (30 stations)

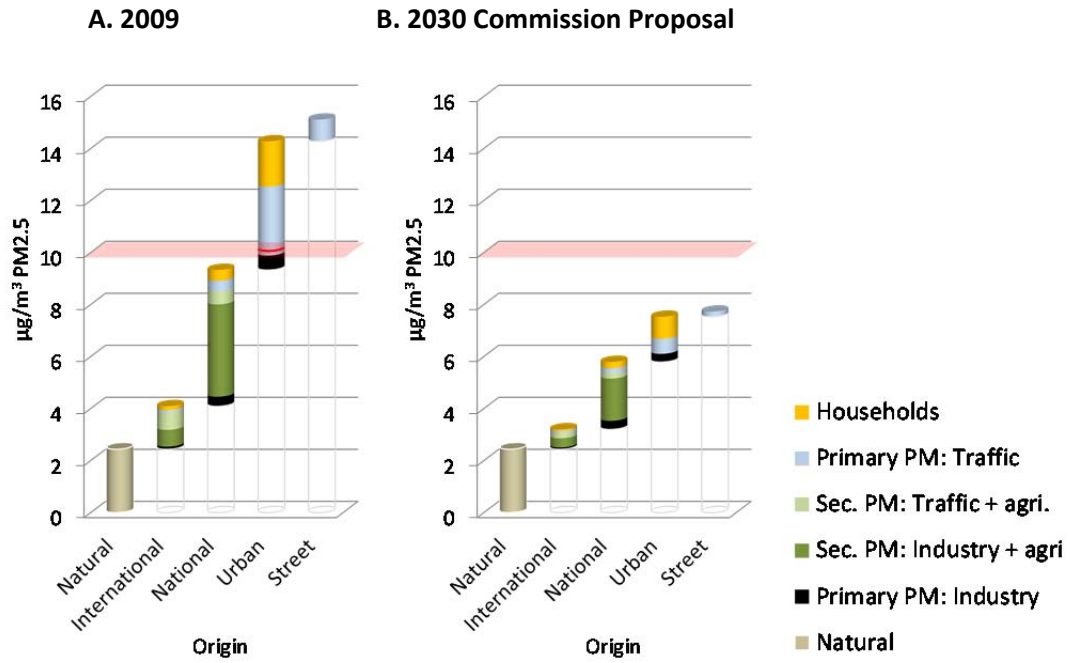

Figure 2.20. Source contributions to ambient PM2.5 at urban traffic stations in Spain, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Sweden (5 stations)

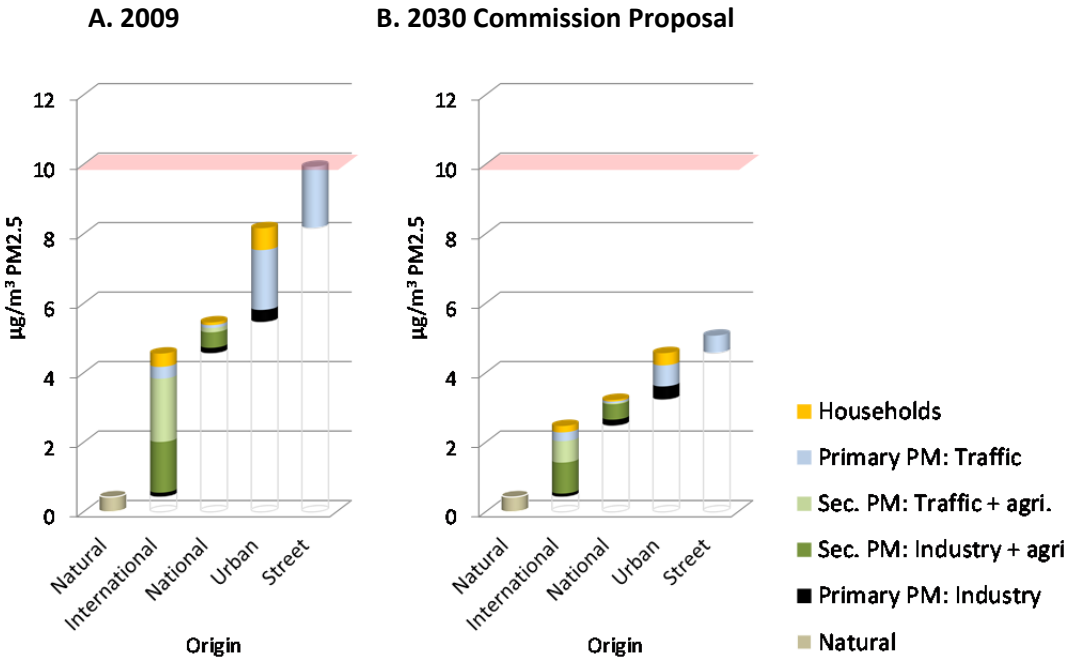

Figure 2.21. Source contributions to ambient PM2.5 at urban traffic stations in Sweden, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

United Kingdom (24 stations)

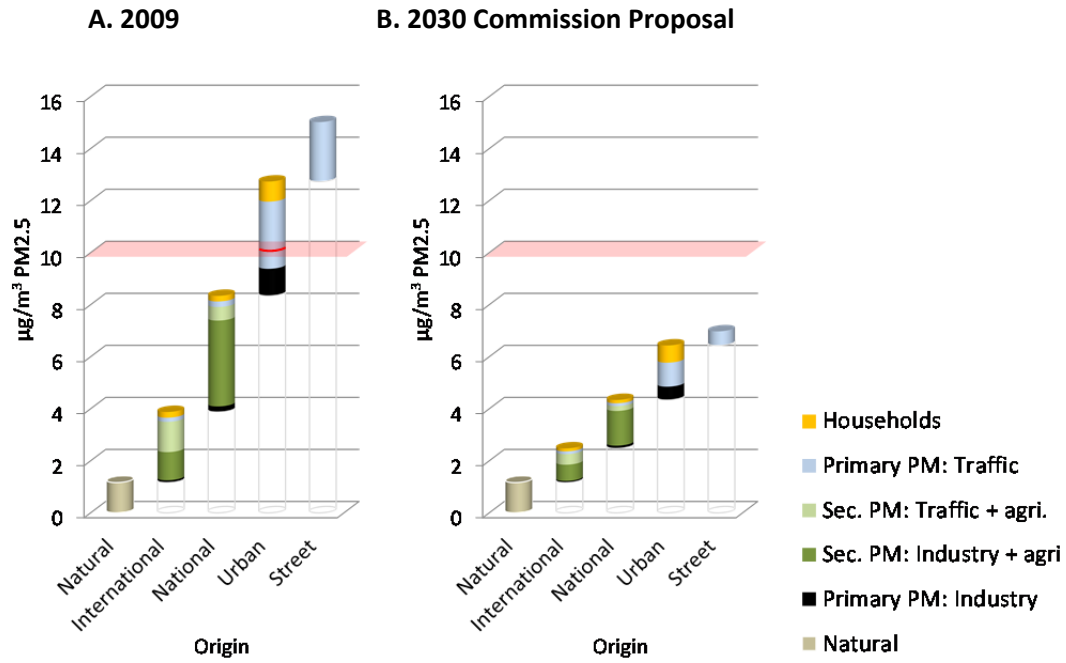

Figure 2.22. Source contributions to ambient PM2.5 at urban traffic stations in the UK, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

3 Discussion and conclusions

Average urban traffic PM2.5 concentrations exceeded the WHO standards in the vast majority of Member States in 2009. Peak values within each Member State are far higher than this average; overall, this presents a strong need for emission reductions for both legal compliance as well as public health considerations.

The source allocation presented in Chapter 2 shows that while source contributions vary strongly between individual countries, all spatial domains considered play their roles. In particular, it becomes clear that PM pollution cannot be considered a purely local problem. For several Member States, such as Belgium, Czech Republic, Netherlands, Hungary, Austria, transboundary transport of PM and precursor gases is a major contributor to urban PM2.5 levels, and it will be very difficult for these countries to decrease their urban PM2.5 to safe levels without coordinated international action. On the other hand, several regions show dramatic local increments, pointing to the possible effectiveness of local measures to reduce ambient PM2.5.

Transboundary transport is dominated by secondary pollution, while primary PM plays a role mostly for local sources. Hence, reductions in both primary PM and secondary precursor emissions will be needed to bring down PM2.5 to safe levels.

Emissions from household heating and road transport are the dominant sources of primary PM. In many Member States, especially those with very high PM concentrations (e.g., Poland, Czech Republic, Slovakia, Romania, and Bulgaria), domestic heating is the largest single source sector.

In the coming years, progressing introduction of particle filters for diesel vehicles is expected to reduce primary PM from road transport by almost two thirds. Remaining PM emissions from this sector will be mainly from non-exhaust sources (road abrasion, brake and tyre wear). Household heating will remain an important source of PM emissions, particularly in areas where coal or inefficient biomass burning is used. The anticipated decline in solid fuel use for heating together with the introduction of newer stoves would reduce emissions from this sector by ~25% without further policy interventions. More stringent product standards could cut emissions by another 66%. Accelerated substitution of inefficient burning of solid fuels in households by cleaner fuels such as natural gas or efficient biomass combustion could achieve additional emission reductions that are not considered in the Commission Proposal.

Overall, the Commission Proposal would lead to a decline of primary PM emissions by 51% by 2030. As a consequence, secondary aerosols are expected to become the dominant contributors to the remaining PM2.5 concentrations.

Many different source sectors are involved in the formation of secondary aerosols, and the various chemical processes make it difficult to uniquely trace them back to a single source. However, the formation of ammonium sulphate ($(NH_4)_2SO_4$, NH_4HSO_4) and ammonium nitrate (NH_4NO_3) is critically steered by the availability of NH_3. (Megaritis et al. 2013; Beauchamp et al. 2013). Ammonia (NH_3) emissions emerge predominantly from agricultural sources, and form, together with SO_2 and NO_x emissions mainly from power generation and industry, secondary inorganic particles.

Thereby, the future trends in secondary inorganic aerosols will depend critically on measures for agricultural NH_3 emissions. The Clean Air Policy Package of the European Commission proposes for 2030 a cut of NH_3 emissions by 27% relative to 2005. NO_x should be reduced by 69%, and SO_2 emissions by 81%.

Together with the proposed measures for primary particle emissions, this should reduce ambient PM2.5 levels by 50% or more in most Member States, as shown in Section 2.2. On average, urban roadside PM2.5 levels would then attain the WHO guideline value in seven Member States, although concentrations at some peak location could still be higher. At such places, however, there will be a realistic chance to control the remaining exceedance by local measures.

REFERENCES

Amann M, Bertok I, Borken-Kleefeld J, et al. (2011) Cost-effective control of air quality and greenhouse gases in Europe: modeling and policy applications. EMS 26:1489–1501. doi: 10.1016/j.envsoft.2011.07.012

Amann M, Borken-Kleefeld J, Cofala J, et al. (2014) The Final Policy Scenarios of the EU Clean Air Policy Package. TSAP Report #11. International Institute for Applied Systems Analysis, Laxenburg, Austria.

ApSimon HM, Gonzalez del Campo MT, Adams HS (2001) Modelling long-range transport of primary particulate material over Europe. Atmospheric Environment 35:343–352. doi: 10.1016/S1352-2310(00)00143-6

Beauchamp M, Bessagnet B, Guerreiro C, et al. (2013) Sensitivity analysis of ammonia emission reductions on exceedances of PM air quality standards. European Topic Centre on Air and Climate Change, Bilthoven, Netherlands

EC (2008) Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe.

EC (2013) Proposal for a Directive of the European Parliament and of the Council on the reduction of national emissions of certain atmospheric pollutants and amending Directive 2003/35/EC. European Commission (EC), Brussels, Belgium

Kiesewetter G, Borken-Kleefeld J, Heyes C, et al. (2013) Modelling compliance with NO2 and PM10 air quality limit values in the GAINS model. TSAP Report #9. International Institute for Applied Systems Analysis, Laxenburg, Austria

Kiesewetter G, Borken-Kleefeld J, Schoepp W, et al. (2014a) Modelling street level PM10 concentrations across Europe: source apportionment and possible futures. Atmos Chem Phys Discuss 14:18315–18354. doi: 10.5194/acpd-14-18315-2014

Kiesewetter G, Borken-Kleefeld J, Schöpp W, et al. (2014b) Modelling NO2 concentrations at the street level in the GAINS integrated assessment model: projections under current legislation. Atmos Chem Phys 14:813–829. doi: 10.5194/acp-14-813-2014

Makra L, Matyasovszky I, Guba Z, et al. (2011) Monitoring the long-range transport effects on urban PM10 levels using 3D clusters of backward trajectories. Atmospheric Environment 45:2630–2641. doi: 10.1016/j.atmosenv.2011.02.068

Malcolm AL, Derwent RG, Maryon RH (2000) Modelling the long-range transport of secondary PM10 to the UK. Atmospheric Environment 34:881–894. doi: 10.1016/S1352-2310(99)00352-0

Megaritis AG, Fountoukis C, Charalampidis PE, et al. (2013) Response of fine particulate matter concentrations to changes of emissions and temperature in Europe. Atmos Chem Phys 13:3423–3443. doi: 10.5194/acp-13-3423-2013

Menut L, Bessagnet B, Khvorostiyanov D, et al. (2013) CHIMERE 2013: a model for regional atmospheric composition modelling. Geosci Model Dev 6:981–1028.

Pope CA, Burnett RT, Turner MC, et al. (2011) Lung Cancer and Cardiovascular Disease Mortality Associated with Ambient Air Pollution and Cigarette Smoke: Shape of the Exposure–Response Relationships. Environ Health Perspect 119:1616–1621. doi: 10.1289/ehp.1103639

Pope III CA, Burnett RT, Thun MJ, et al. (2002) Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. JAMA: the journal of the American Medical Association 287:1132–1141.

Simpson D, Benedictow A, Berge H, et al. (2012) The EMEP MSC-W chemical transport model–technical description. Atmos Chem Phys 12:7825–7865.

Tsyro SG (2008) Regional model for formation, dynamics and Long-range transport of

atmospheric aerosol: Study of atmospheric aerosol properties in Europe. Russ Meteorol Hydrol 33:300–309. doi: 10.3103/S106837390805004X

US-EPA (2013) National Ambient Air Quality Standards for Particulate Matter; Final Rule. Federal Register 78, 10:3086–3287.

WHO (2013) Review of evidence on health aspects of air pollution – REVIHAAP Project. Copenhagen, Denmark

World Health Organization (2006) Air quality guidelines global update 2005: particulate matter, ozone, nitrogen dioxide, and sulfur dioxide. World Health Organization, Copenhagen, Denmark

A. Technical Appendix: Methodology

Since the number of stations reporting PM2.5 in 2009 is relatively low compared to PM10, especially in critical areas in the new Member States, all stations covered by the PM10 scheme are included in this analysis, regardless whether they report PM2.5 in 2009 or not. In case roadside PM2.5 observations are missing, the roadside increment is estimated from the NO_x traffic increment, scaled with the ratio of road traffic PM2.5 emissions over road traffic NO_x emissions. If PM2.5 urban background measurements are missing, the urban background PM2.5 is estimated as modelled PM2.5 + country average scaling factor × PM10 residual. The PM10 residual is known as PM10 urban background observations are required for any roadside station to be covered by the PM10 model. The scaling factor, representing the fraction of PM2.5 in PM10 residuals, is calculated from country average PM2.5 in PM10 residuals averaged at all stations where both measurements are available.

Figure A.1 gives a schematic overview of the re-allocation that is done in a post-processing step. For this analysis the term "regional background" is introduced, which is supposed to contain contributions from all sources except the city where the station is located. While the 28×28km^2 modelled background is a good first indicator for the regional background, it is not suitable in all places, as it does contain contributions from local emissions.

The basic assumptions behind the re-allocation are:

- EMEP natural dust fields as used in GAINS may be too low in some places. E.g. CHIMERE dust fields are usually higher. Thus, parts of the residual may be attributed to natural dust – however, only within certain limits.
- The residual may contain a regional as well as a local component. Only the regional component may be partly attributed to higher natural dust, while the local component is definitely related to either under-estimated local emissions or over-estimated mixing in the boundary layer.
- Secondary aerosol formation requires time and is therefore not considered a local phenomenon. As a simplification, all secondary aerosol must be contained in the regional background, while the urban increment contains only primary PM.
- consequently, a re-attribution of the 28×28km^2 modelled background concentrations into regional and local components is only needed for PPM.

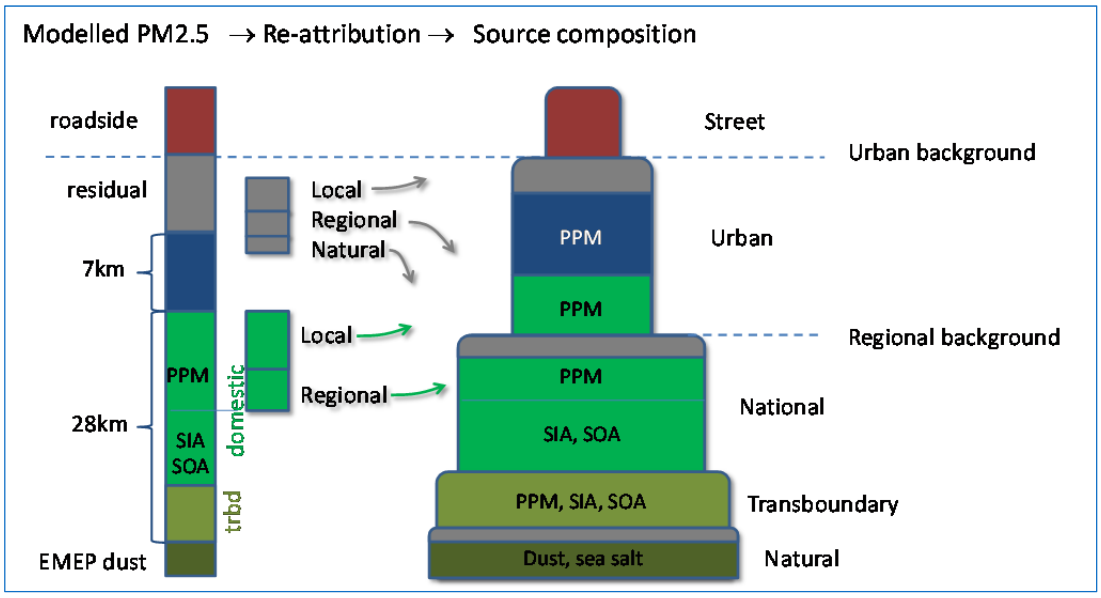

Figure A.3.1. Schema of the post-processing analysis. Modelled concentrations (left) are taken as basis and merged with the information on regional background PM2.5 levels. The residual is split up into regional and local contributions, as well as the PPM part of 28×28km^2 modelled concentrations, to arrive at the spatial-source allocation as presented in Chapter 2 (right).

The following base year information is used in the re-allocation:

- Urban background observations as used in the model
- rural background observations, interpolated to the site
- unexplained residuals at rural background sites, interpolated to the site
- Natural dust fields as used in the CHIMERE model runs, which are higher than the EMEP fields

The re-allocation distinguishes between cases where the urban background is underestimated by bottom up modelling (which is the case in the majority of sites), and those where the urban background is over-estimated.

The process consists of the following steps (here for stations in which the urban background is underestimated):

1) The unexplained residual is split into a regional part (i.e. interpolated residual from nearby rural background sites) and a remaining local part.
2) Out of the regional residual, as much as possible is allocated to natural dust (up to the dust fields used in CHIMERE), the rest is considered anthropogenic.
3) Anthropogenic regional residuals are attributed to source sectors and species (PPM/SIA/SOA) in the same shares as the domestic 28×28km modelled PM2.5 at the site
4) A regional background level $[PM2.5]_{RB}$ is defined as the minimum of:
    a. interpolated observed rural background PM2.5
    b. the modelled 28×28km^2 PM2.5 + EMEP natural fields + regional residual

    However, it must contain at least

    c. 28×28km^2 modelled transboundary PM2.5 + EMEP natural fields + regional residual + 28×28km^2 modelled domestic SIA

    so that

    $[PM2.5]_{RB}$ = max(min(a,b),c) .
5) The 28×28km^2 national PPM concentrations are split up into regional and local components according to the constraints imposed by regional background and regional residual defined in steps 1 and 3.
6) Local residuals are sector-wise attributed to PPM emissions within a range of 30km from the site, taken from the gridded CHIMERE 7×7km^2 emission inventory.

At stations where the urban background is overestimated, steps 1-3 and 6 are omitted. Regional background levels are defined as in step 4. A scaling factor is calculated as

$$f = \frac{[PM_{2.5}]_{B_{obs}} - [PM_{2.5}]_{nat}}{[PM_{2.5}]_{B_{28km}} + [PM_{2.5}]_{B_{7km}}}$$

with $[PM_{2.5}]_{B_{obs}}$ the observations, $[PM_{2.5}]_{B_{28km}}$ and $[PM_{2.5}]_{B_{7km}}$ the 28km modelled concentrations and the 7km urban increment, respectively, and $[PM_{2.5}]_{nat}$ the natural PM fields as calculated in the EMEP model for 2009. This scaling factor is then applied equally to all sectoral and chemical components in $[PM_{2.5}]_{B_{28km}}$ and $[PM_{2.5}]_{B_{7km}}$, so that their sum plus the unchanged natural dust and sea salt fields match observed PM2.5 levels.