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Sectorial and regional uncertainty analysis of the contribution of anthropogenic emissions to regional and global $PM_{2.5}$ health impacts

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Abstract

In this work we couple the HTAPv2.2 global air pollutant emission inventory with the global source receptor model TM5-FASST to evaluate the relative contribution of the major anthropogenic emission sources (power generation, industry, ground transport, residential, agriculture and international shipping) to air quality and human health in 2010. We focus on particulate matter (PM) concentrations because of the relative importance of $PM_{2.5}$ emissions in populated areas and the proven cumulative negative effects on human health. We estimate that in 2010 regional annual averaged anthropogenic $PM_{2.5}$ concentrations varied between ca 1 and 40 $\mu g/m^3$ depending on the region, with the highest concentrations observed in China and India, and lower concentrations in Europe and North America. The relative contribution of anthropogenic emission source sectors to $PM_{2.5}$ concentrations varies between the regions. European PM pollution is mainly influenced by the agricultural and residential sectors, while the major contributing sectors to PM pollution in Asia and the emerging economies are the power generation, industrial and residential sectors. We also evaluate the emission sectors and emission regions in which pollution reduction measures would lead to the largest improvement on the overall air quality. We show that in order to improve air quality, regional policies should be implemented (e.g. in Europe) due to the transboundary features of PM pollution. In addition, we investigate emission inventory uncertainties and their propagation to $PM_{2.5}$ concentrations, in order to identify the most effective strategies to be implemented at sector and regional level to improve emission inventories knowledge and air quality. We show that the uncertainty of PM concentrations depends not only on the uncertainty of local emission inventories but also on that of the surrounding regions. Finally, we propagate emission inventories uncertainty to PM concentrations and health impacts.

1 Introduction

Ambient particulate matter pollution ranks among the top five risk factors globally for loss of healthy life years and is the largest environmental risk factor (Lim et al., 2012;Anderson et al., 2012;Anenberg et al., 2012). The world health organization (WHO, 2016) reported about 3 million premature deaths worldwide attributable to ambient air pollution in 2012. Health impacts of air pollution can be attributed to different anthropogenic emission sectors (power generation, industry, residential, transport, agriculture, etc.) and sector-specific policies could effectively

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reduce health impacts of air pollution. These policies are usually implemented under national legislation, while in Europe transboundary air pollution is also addressed by the regional protocol under the UNECE Convention on Long-Range Transport of Air Pollution (CLRTAP). Indeed, particulate matter can travel thousands of kilometers, crossing national borders, oceans and even continents (HTAP, part A, 2010). Therefore local, regional and international coordination is needed to define air pollution policies to improve globally air quality and human health. The CLRTAP's Task Force on Hemispheric Transport of Air Pollution looks at the long-range transport of air pollutants in the Northern Hemisphere aiming to identify promising mitigation measures to reduce background pollution levels and its contribution to pollution in rural as well as urban regions. Although primary $PM_{2.5}$ (particulate matter with a diameter less than 2.5 μm) can travel over long distances, the transboundary components of anthropogenic PM are mainly associated with secondary aerosols which are formed in the atmosphere through complex chemical reactions and gas-to-aerosol transformation of gaseous precursors transported over source regions (Maas and Grennfelt, 2016). Secondary aerosol from anthropogenic sources consists of both inorganic (mainly ammonium nitrate and ammonium sulfate and ammonium bisulfate and associated water, formed from emissions of sulphur dioxide (SO_2), nitrogen oxides (NOx) and ammonia (NH_3)) and organic compounds involving thousands of compounds and often poorly known reactions (Hallquist et al., 2009). Exposure of human to aerosol can be estimated by a variety of approaches, ranging from epidemiological studies to pure modeling approaches. At the global scale, models, in some cases using satellite information (Brauer et al., 2015;Van Donkelaar et al., 2016), are the most practical source of information of exposure to air pollution. However, model calculations are subject to a range of uncertainties related with incomplete understanding of transport, chemical transformation, removal processes, and not to the least emission information.

This work is developed in the context of the TF HTAP Phase 2 (Galmarini et al., 2017), where a set of models is deployed to assess long-range sensitivities to extra-regional emissions, using the prescribed HTAP_v2 emission inventory (Janssens-Maenhout et al., 2015). Differences in model results illustrate uncertainties in model formulations of transport, chemistry and removal processes, and are addressed in separate studies (West et al., 2017, in prep.), but not of uncertainties in emission inventories. The objective of this study is to evaluate the relevance of uncertainties in regional sectorial emission inventories (power generation, industry, ground transport, residential, agriculture and international shipping), and its propagation in modeled $PM_{2.5}$ concentrations and associated impacts on health, comparing the derived uncertainties in $PM_{2.5}$ from within the region to extra-regional uncertainties. A second objective of this analysis is to evaluate the importance of emission uncertainties at sector and regional level on $PM_{2.5}$, to better inform local, regional and hemispheric air quality policy makers on the potential impacts of less known emission sectors or regions. In this work we couple the HTAP_v2.2 global emission inventory for the year 2010 and the global source-receptor model TM5-FASST (TM5-FAst Scenario Screening Tool) to estimate global air quality in terms of $PM_{2.5}$ concentrations.

2 Methodology

2.1 HTAP_v2.2 emissions

The global anthropogenic emission inventory HTAP_v2.2 for the year 2010 is input to the global source-receptor model TM5-FASST to evaluate $PM_{2.5}$ concentrations for each world region/country with the corresponding health effects. The HTAP_v2.2 inventory includes for most countries official and semi-official annual anthropogenic emissions of SO_2, NOx, CO (carbon monoxide), NMVOC (non-methane volatile organic compounds), PM_{10} (particulate matter with a diameter less than 10 μm) $PM_{2.5}$, BC (black carbon) and OC (organic carbon) by country and sector (Janssens-Maenhout et al., 2015), downloadable at http://edgar.jrc.ec.europa.eu/htap_v2/index.php. Here we focus on the 6 major anthropogenic emission sectors contributing to global $PM_{2.5}$ concentrations, namely the power generation ("power"), non-power industry, industrial processes and product use ("industry"), ground transportation ("transport"), residential combustion and waste disposal ("residential"), agriculture ("agriculture") and international shipping ("ship"). It should be noted that agricultural emissions do not include agricultural waste burning. Details on the emissions included in each aggregated sector can be found in Janssens-Maenhout et al. (2015). In addition to the reference HTAP_v2.2 emissions for the year 2010, a set of scenarios has been created by subtracting from the reference dataset the emissions of each sector. Under the assumption that the individual sector contributions add up linearly to total $PM_{2.5}$, the comparison of $PM_{2.5}$ concentrations calculated for the reference and scenario case yields an estimation of the contribution of each sector to total $PM_{2.5}$ concentrations (Van Dingenen et al., 2017, in preparation).

2.2 TM5-FASST model

In order to calculate $PM_{2.5}$ concentrations from the HTAP_v2.2 emissions, the gridded TM5-FASST version 1.4b (Van Dingenen et al., 2017, in preparation). The TM5-FASST source-receptor model is based on a set of emission perturbation experiments (-20 %) of SO_2, NOx, CO, NH_3, and VOC and CH_4 using the global 1°x1° resolution TM5 model, the meteorological year 2001 (chosen for HTAP Phase 1) and the representative concentration pathway (RCP) emissions for the year 2000 (Lamarque et al., 2010). The concentration of $PM_{2.5}$ contributing from and to each of 56 receptor regions is estimated as a linear function of the emissions of the source regions, including the aerosol components BC, primary organic matter (POM), SO_4, NO_3, and NH_4. While SOA of natural sources is included using the parameterisation described in Dentener et al. (2006), no explicit treatment of anthropogenic SOA is considered. Specifically, the change in $PM_{2.5}$ concentrations, compared to a reference concentration in the receptor region y (dPM), induced by changes in precursor emissions in the source region x relativey to the reference case (dE), is estimated as following:

$$dPM(\boldsymbol{y}) = \sum_i SRC_i[x,y] \cdot \left[E_i(x) - E_{i,\mathrm{ref}}(x)\right] \quad \text{(Eq. 1)}$$

$$SRC_i[x,y] = \frac{\Delta PM_{ref}(y)}{\Delta E_{i,ref}(x)} \quad \text{(Eq. 2)}$$

where the summation is made over all primary emitted components and precursors (i) for secondary components, and $SRC_i[x,y]$ is a set of Source-Receptor Coefficients describing the linearized relationship between each precursor emission of specific components and concentration for each pair of source and receptor region. Therefore to calculate total $PM_{2.5}$ concentration in each receptor region, the sum of the 56 source region contributions must be taken into account. Using this approach, it is possible to evaluate the $PM_{2.5}$ concentrations from "within-region" and "extra-regional" $PM_{2.5}$ emissions. Further details about the TM5-FASST methodology and assumptions can be found in Van Dingenen et al. (2017, in preparation) and Leitão et al. (2013). As depicted in Fig. S1, the 56 TM5-FASST regions cover the entire globe,

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but their areal extent differs in terms of size, population, emission magnitude and presence of neighbouring countries (e.g. Europe comprises 18 TM5-FASST regions). In order to make smaller regions (e.g. European countries) comparable with larger regions (like USA, China and India), in this work an aggregation procedure to 10 world regions (refer to Table S2) has been applied (China+, India+, SE Asia, North America, Europe, Oceania, Latin America, Africa, Russia and Middle East). In this work we focus on particulate matter due to its negative effects on human health (WHO, 2013;Pope and Dockery, 2006), (Worldbank, 2016). The TM5-FASST model includes an assessment of the premature mortality due to ambient $PM_{2.5}$ concentrations on exposed population following the methodology developed by Burnett et al. (2014), as discussed in Sect. 4. Health impacts of indoor air pollution are not evaluated.

2.3 Emission inventory uncertainties

In order to investigate how computed $PM_{2.5}$ concentrations are affected by the uncertainty of emission inventories, we perform a sensitivity analysis testing the upper and lower range of HTAP_v2.2 emissions including their uncertainties. Aggregated emissions of a certain pollutant *p*, from a sector *i* and country *c* are calculated as the product of activity data (AD) and emission factors (EF), therefore the corresponding uncertainty ($\sigma_{i,c,p}$) is calculated as following:

$$\sigma_{EMI\ i,c,p} = \sqrt{\sigma^2_{ADi,c} + \sigma^2_{EF,i,p,c}} \qquad \text{(Eq.3)}$$

where σ_{AD} and σ_{EF} are the uncertainties (%) of the activity data and emission factors for a certain sector, country and pollutant. Uncertainty values of the activity data by sector and country are obtained from Janssens-Maenhout et al. and references therein (2017, submitted.), while uncertainty values for the emission factors of gaseous pollutants are retrieved from the EMEP/EEA Guidebook (2013) and Bond et al. (2004) for particulate matter. Differently from gridded emission inventories which often make use of similar proxies and spatial correlation structures, while errors in emissions may be correlated (e.g. the same systematic error in an estimate of EF introduced in the inventory for a number of countries), we assume here that reported countries emissions are based on independent evaluation of activity data and estimated EFs, and hence no cross-country correlation structure is assumed. Therefore, we can calculate the overall uncertainty $\sigma_{EMI\ p,c}$ for a certain pollutant (*p*) due to all sectors (*i*) in a specific country (*c*) with the following equation (EMEP/EEA, 2013).

$$\sigma_{EMI\ p,c} = \sqrt{\sum_i \left(\sigma_{EMI\ i,c,p} * \frac{EMI_{i,c,p}}{EMI_{tot,c,p}}\right)^2} \qquad \text{(Eq. 4)}$$

where $EMI_{i,c,p}$ (in kton) represents the emission of a certain pollutant in a certain country from a specific sector (i) and $EMI_{tot,c,p}$ (in kton) the corresponding emissions from all sectors for that country and pollutant.

Table S3, reports the overall uncertainty calculated for each pollutant and for each TM5-FASST region. Using an additional constraint that EFs and activities cannot be negative, a lognormal

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distribution of the calculated uncertainties is assumed (Bond et al., 2004); therefore we can calculate the upper and lower range of emission estimates multiplying and dividing the reference emissions by $(1+\sigma_{p,c})$, respectively. We do not account for the uncertainties of the atmospheric transport model and the uncertainties due to aggregation which are larger over smaller TM5-FASST regions.

3 TM5-FASST modelling results

3.1 Regional contributions to $PM_{2.5}$ concentrations

Figure 1 provides a global perspective on the fraction of within-region and extra-regional $PM_{2.5}$ concentrations for 10 aggregated receptor regions using emissions of the year 2010, with the extra-regional fraction broken down into source region contributions. Annual average population weighted anthropogenic $PM_{2.5}$ concentrations ranged from few µg/m^3 (e.g. in Oceania or Latin America), around 7-8 µg/m^3 for North America and Europe, and up to 33-39 µg/m^3 in China+ (including also Mongolia) and India+ (including also the rest of South Asia). Anthropogenic $PM_{2.5}$ pollution in China+ and India+ is mainly affected by large emission sources within the country (98 and 96%, respectively), although 4 % of the Indian anthropogenic $PM_{2.5}$ pollution is transported from the Gulf region. North America (98%) and Oceania (98%) are mainly influenced by within-regional pollution due to their geographical isolation from other regions. TM5-FASST computations attributed 11 % of the $PM_{2.5}$ in Europe to extra-regional sources; for the Middle East and Gulf region extra-regional contributions amount to 18% (mainly from Europe and Russia), for Africa 25% (mainly from Europe and Middle East), and Russia 28% (mainly from Europe, Middle East and Gulf region and China). Shipping emissions are not considered in this analysis due to their international origin. Transboundary air pollution is known to be an important issue in the rest of Asia, in particular for pollution transported from China to Korea and Japan (Park et al., 2014) and we estimate that the contribution of transported PM is up to 40% in South Eastern Asia (mainly from China and India). Within-region and extra-regional $PM_{2.5}$ concentrations for all the TM5-FASST regions are reported in Table S2.

Focusing on Europe, Fig. 2 shows within-region (in black) vs. extra-regional absolute population-weighted $PM_{2.5}$ concentrations (in µg/m^3) for 16 EU countries plus Norway and Switzerland, defined in TM5-FASST, as well as the source regions contributing to this pollution. Annual averages of $PM_{2.5}$ concentrations in Europe vary between 2-4 µg/m^3 in Northern European countries (like Finland, Norway and Sweden) up to 10-12 µg/m^3 for continental Europe. Although most of EU annual average PM2.5 concentrations are below the World Health Organization Air Quality Guideline of 10 µg/m^3 $PM_{2.5}$ (as annual average), these values represent only regional averages while several exceedances especially in urban areas are often observed in Europe. As discussed in Sect. 3.2, an additional contribution to $PM_{2.5}$ concentrations comes from the shipping sector, mainly influencing Mediterranean countries (like Italy, Spain and France) and countries facing the North Sea, Baltic Sea and Atlantic Ocean (e.g. Benelux, Sweden, Great Britain, etc.). From a European perspective, $PM_{2.5}$ represents a transboundary issue since extra-regional contributions range between 27% and 75% (on average 51%). Countries surrounded by oceans, are mainly influenced by within-region pollution due to their geographical isolation from other source regions (e.g. Italy, Spain, Great Britain and Norway); therefore the fraction of extra-regional pollution ranges from 27% to 35%. The largest extra-regional contributions are calculated for Hungary (75%, mainly from Austria, Czech Republic, Rest of Central EU, Poland and Germany), Czech Republic (67%, mainly from Poland, Germany and Austria), Austria and Slovenia (66%, mainly from Czech Republic, Germany and Italy), Sweden+Denmark (65%, mainly from Germany, Norway and Poland), Bulgaria (63%, mainly from Romania), Greece (61%). The remaining EU countries are both affected by within-region and extra-regional pollution (the latter ranging from 40% to 59%), highlighting the importance of transboundary transport of $PM_{2.5}$ concentrations. For example Switzerland is influenced by the pollution coming from France, Italy and Germany; Rest of Central EU by Poland and Germany; Germany by France and Benelux; Poland by Czech Republic and Germany. Interestingly, Romania, Bulgaria, Greece and Hungary are also significantly affected by the pollution transported by Ukraine and Turkey, which is included in the "rest of the world" contribution of Fig. 2. Our results are consistent with the findings of the latest UNECE Scientific Assessment Report (Maas and Grennfelt, 2016) where the importance of transboundary transport to PM, which in Europe mainly consists of secondary organic (not fully treated by TM5-FASST) and inorganic particles (e.g. ammonium nitrate and sulfate) formed from gaseous precursors, is highlighted. Therefore, in order to reduce regional mean PM concentrations, across-regional approach taking into account atmospheric transport and chemical transformations of pollutants over a wide scale could be considered.

3.2 Sectorial contributions to $PM_{2.5}$ concentrations

Figure 3 shows the relative sectorial contributions to anthropogenic $PM_{2.5}$ concentrations for the 56 TM5-FASST receptor regions, separating the fraction of extra-regional (shaded) and within-region pollution, while Table 1 shows regional average values of sector-specific relative contributions. In most African regions (except Egypt) anthropogenic $PM_{2.5}$ concentrations are mainly produced by emissions in the residential sector. Agriculture is an important sector for Egypt, while Northern Africa is strongly influenced by shipping emissions in the Mediterranean (30%). $PM_{2.5}$ in emerging economies in Asia, Latin America and Middle East are dominated by $PM_{2.5}$ concentrations from the residential sector, power generation and industrial. Asian countries, China, India, Indonesia and Philippines are mainly influenced by within-region pollution with the largest contributions coming from power, industry and residential sectors. Japan is characterized by the contribution of local sources like transport and agriculture but it is also affected by transported pollution from China, especially from the industrial sector. Anthropogenic $PM_{2.5}$ in the remaining Asian countries is influenced by more than 50% by the pollution coming from China (e.g. Vietnam, Malaysia, Thailand, Mongolia, South Korea, Taiwan) or India (e.g. Rest of South Asia and South Eastern Asia) from the power, industry and residential activities. A different picture is observed for Europe where according to our calculations, PM concentrations stem mainly from the agricultural and residential sectors with somewhat less contribution from the transport sector. In Eastern European countries relevant contributions are also found from the power and industrial sectors in Eastern European countries, related to the relatively extensive use of polluting fuels like coal. $PM_{2.5}$ concentrations in USA and Canada are mostly affected by the power, industry and agricultural sectors. In Oceania industry and agriculture are the most important sectors. $PM_{2.5}$ concentrations formed from ship emissions mainly affect coastal areas of North Africa, SE Asia (e.g. in Japan, Taiwan, Malaysia, Indonesia and Philippines), Mediterranean countries (Spain by 11%, Italy by 5%, France by 7% of their corresponding country totals), Northern EU regions (Great Britain by 10%, Norway by 6%, Sweden and Denmark by 10% of their corresponding country totals) and Oceania (22% of the regional total). Over the international areas of sea and air no distinction between within-region and extra-regional concentrations is reported. Further details on within-region and extra-regional concentrations can be found in section S2 of the Supplementary Material.

3.3 Gridded $PM_{2.5}$ concentrations

Figure 4 shows the global 1°x1° gridmaps of anthropogenic $PM_{2.5}$ concentrations in 2010 for the reference case as well as the contribution from each of the major anthropogenic emission sectors. Global $PM_{2.5}$ concentrations are ubiquitous and range from few µg/m^3 over the oceans and seas to more than 50 µg/m^3 over Asia. As shown also in Fig. 3, the most polluted countries in Asia are China, India and Rest of South Asia (which includes Afghanistan, Bangladesh, Bhutan, Nepal and Pakistan) with annual average anthropogenic $PM_{2.5}$ concentrations ranging from 29 to 40 µg/m^3; rather polluted areas are also found in Mongolia and North Korea, Vietnam, South Korea, Rest of South Eastern Asia (including Cambodia, Lao People's Democratic Republic and Myanmar), Thailand, Japan and Taiwan with $PM_{2.5}$ concentration in the range of 6 to 14 µg/m^3. The highest $PM_{2.5}$ concentrations in Africa are observed in Egypt (11 µg/m^3 as annual average), Republic of South Africa (6.1 µg/m^3 as annual average) and Western Africa (4.0 µg/m^3 as annual average). The highest pollution in Europe is observed in the Benelux region, Italy and in some of the Eastern countries (e.g. Romania, Bulgaria and Czech Republic), while in Latin America the most polluted areas are Chile (13.7 µg/m^3 as annual average) and Mexico (4.2 µg/m^3 as annual average). Middle East, the Gulf region, Turkey, Ukraine and former USSR are also characterised by $PM_{2.5}$ concentrations ranging between 7.5 µg/m^3 and 9.2 µg/m^3 as annual averages.

Modelled $PM_{2.5}$ concentrations are in the range of the measurements and satellite-based estimates provided in several literature studies (Brauer et al., 2012;Brauer et al., 2015;Boys et al., 2014;Evans et al., 2013;Van Donkelaar et al., 2016), reporting for the whole Europe annual averaged $PM_{2.5}$ concentrations in the range between 11 and 17 µg/m^3, for Asia from 16 to 58 µg/m^3, Latin America 7-12 µg/m^3, Africa and Middle East 8-26 µg/m^3, Oceania 6 µg/m^3 and North America 13 µg/m^3 (note that measurements and satellite estimates would not separate anthropogenic and natural sources of PM, e.g. dust, biomass burning, while the concentrations in this study pertain to anthropogenic emissions alone).

In order to understand the origin of global $PM_{2.5}$ concentrations, we look at sector specific maps (Fig. 4). The power and industrial sectors are mainly contributing to PM concentrations in countries having emerging economies and fast development (e.g. Middle East, China and India), while the ground transport sector is a more important source of PM concentrations in industrialised countries (e.g. North America and Europe) and in developing Asian countries. The residential sector is one of the most significant sources of PM all over the world, potentially also affecting indoor air quality. Africa and Asia are strongly influenced by PM concentrations produced by this sector due to the incomplete combustion of rather dirty fuels and solid biomass deployed for domestic purposes (both heating and cooking). Interestingly, the agricultural sector is affecting pollution in Asia as well as in Europe and North America, confirming the findings of the UNECE Scientific Assessment Report (Maas and Grennfelt, 2016). The residential and agriculture sectors are less spatially confined, and more difficult to effectively regulate with emission reductions than point source emissions of the industrial sectors (e.g. in Europe the Large Combustion Plant Directive, the National Emission Ceilings or the Industrial Emissions, the Euro norms for road transport, etc.). Finally, shipping is mainly contributing to the pollution in countries and regions with substantial coastal areas, and with ship tracks on the Mediterranean Sea, the Atlantic, Pacific and Indian Oceans, as depicted in Fig. 4.

3.4 Uncertainty from emissions

3.4.1 Propagation of emission uncertainties to $PM_{2.5}$ concentrations

Table 2, as well as Fig. 5, report the annual average $PM_{2.5}$ concentrations (µg/m^3) estimated by TM5-FASST with the uncertainty bars representing the upper and lower range of concentrations due to emission inventories uncertainty. The extra-regional contribution to uncertainty is also addressed as well as the contribution of the uncertainty of primary particulate matter emissions to the upper range of $PM_{2.5}$ concentrations (refer to Table 2). We observe that primary PM represents the dominant source of uncertainties in terms of emissions, contributing from 45% to 97% to the total uncertainty of $PM_{2.5}$ concentrations for each country/region.

Figure 5 depicts the results of the propagation of lowest and highest range of emissions including their uncertainty to $PM_{2.5}$ concentrations for Asia (panel *a*) and - in more detail- Europe (panel *b*), highlighting the contribution of within-region and extra-regional $PM_{2.5}$ concentrations and uncertainties (error bars). Due to their large sizes, Indian and Chinese $PM_{2.5}$ concentrations and uncertainties are mainly affected by uncertainties from the residential, transport and agricultural sectors within these countries. Interestingly, in South Eastern and Eastern Asia uncertainties in $PM_{2.5}$ are strongly influenced by the Indian residential emissions. On the other hand, $PM_{2.5}$ in Thailand, Japan, Taiwan, South Korea, Mongolia and Vietnam are strongly affected by the uncertainty in the Chinese residential and industrial emissions. Therefore our study finds that reducing the uncertainties in the Chinese and Indian emission inventories will be highly relevant for understanding the long-range contribution of $PM_{2.5}$ pollution in most of Asian countries.

In Europe the highest uncertainties in $PM_{2.5}$ concentrations are associated with the emissions from the residential, agriculture and transport sectors. In most of the Central and Eastern European countries modelled $PM_{2.5}$ is strongly affected by the uncertainty of transported extra-regional pollution, especially from the residential, agricultural and transport sectors. Conversely, uncertainties in Norway are dominated by the national emissions, mainly from the residential and transport sectors, and in Italy from the residential and agriculture sectors. The remaining European countries are affected both by within-region and imported uncertainties. Panel *c* of Fig. 5 represents the results of the propagation of the emissions range including their uncertainty to $PM_{2.5}$ concentrations for North America, Latin America, Oceania and Russia, while panel *d* for Africa, Middle East and the Gulf region. The uncertainty in the USA agricultural and residential emissions affect more than 50% of modelled Canadian $PM_{2.5}$ concentrations and the uncertainty in Mexico and Argentina is influenced by similar magnitudes (30-50%) by neighbouring countries. The uncertainty of within-region emissions, especially from the residential sector, dominates the overall levels of $PM_{2.5}$ uncertainties in Latin America. In addition, in Chile also the within-region agriculture and power sectors contribute significantly to the overall uncertainty levels. $PM_{2.5}$ levels in most of the African regions are strongly affected by the uncertainty in their own residential emissions, while in Egypt they are mostly influenced by the agricultural sector uncertainties (refer to Fig. 5, panel *d*). Interestingly, anthropogenic $PM_{2.5}$ in Northern Africa is influenced by Italian emissions uncertainty as well as by emissions from shipping. Conversely, the Middle East and Turkey regions are influenced by a range of extra-regional emission uncertainties (e.g. Middle East is affected by the uncertainty of Turkey, Egypt and the Gulf region, while Turkey by Bulgaria, Gulf region and rest of Central EU).

3.4.2 Ranking the sector specific contribution to emission uncertainties

Figure 6 shows the average sector relative contribution to total emission inventory related uncertainty for the main PM2.5 concentration precursors and world regions, representing the ranking of the most effective improvements to be taken regionally to better constrain their inventories and reduce the final formation of $PM_{2.5}$ concentrations. The complete overview of all TM5-FASST regions is provided in Fig. S2, where the share of each term of the sum of Eq. 4, representing the sector contribution to the uncertainty of each pollutant in each region, is reported. SO_2 uncertainties mainly derive from the power generation sector for most of the world countries especially those that are coal dominated; however, relevant contributions are also observed from the industrial sector in South Africa, Asia, Norway, some Latin American countries, Canada and Russian countries. Interestingly some contributions are also observed from the residential sector in Africa and from the transport sector in some Asian countries (e.g. Korea, Vietnam, Indonesia, South Eastern Asia, etc.). NOx emissions uncertainty mainly derives from the transport sector, although some contributions are also seen from the power generation in Russian countries with gas and Middle East and residential sector in Africa. Depending on the region, CO uncertainty is dominated by the transport or residential (particularly in Africa and Asia) sector and for some regions by a similar contribution of these two sectors. NMVOC emission uncertainties mainly derive from the industrial, transport and residential activities which are still not well characterized in terms of NMVOC emissions due to the complex mixture and reactivity of such pollutants. As expected, NH_3 emission uncertainty is dominated by the agricultural sector which appears to be less relevant for all other pollutants. Primary particulate matter emissions should be mainly improved for the residential and transport sectors and partly for the industrial one. Black carbon emission inventories should be better characterised in Europe, Japan, Korea, Malaysia etc. for the transport sector, where the higher share of diesel used as fuel for vehicles leads to higher BC emissions; in addition, BC emissions from the residential sector require further effort to better characterise them in terms of EFs for the different type of fuels used under different combustion conditions. To constrain and improve organic matter emissions, efforts should be dedicated to the residential emissions characterisation. Therefore, in the following section, we try to assess one of the major sources of uncertainty in the residential emissions in Europe which is the use of solid biofuel.

3.4.3 Assessing the uncertainty in household biofuel consumption with an independent inventory in Europe

The combustion of solid biomass (i.e. biofuel) for household heating and cooking purposes is one of the major sources of particulate matter emissions in the world. Wood products and residues are largely deployed in residential activities, but national reporting often underestimates the emissions from this sector in Europe, due to the fact that often informal economic wood sales are not accurately reflected in the official statistics of wood consumption (AD) (Denier Van Der Gon et al., 2015). An additional uncertainty is related to the lack of information in the inventory regarding the emission factors (EF) variability, which depends on the combustion efficiency and type of wood (Weimer et al., 2008;Chen et al., 2012). In our work we estimate the uncertainty attributable to wood combustion in the HTAP_v2.2 residential sector (σ_{AD,RES_bio}) by comparing it to the recent TNO RWC (residential wood combustion) inventory of Denier van der Gon et al. (2015), which includes a revised biomass fuel consumption, with the corresponding EDGARv4.3.2 activity data (Janssens-Maenhout et al., 2017, in prep.), as shown in Table S4. In the TNO RWC inventory, wood use for each country has been updated comparing the officially reported per capita wood consumption data (from GAINS and IEA) with the expected specific wood use for a country including the wood availability information (Visschedijk et al., 2009;Denier Van Der Gon et al., 2015). We can therefore assume that the TNO RWC inventory represents an independent estimate of wood consumption in the residential sector, allowing a more precise uncertainty estimation of the AD for this sector. Assuming that emissions are calculated as the product of AD and EF, the corresponding uncertainty can be calculated with Eq. 3, where σ_{AD} ranges from 5 to 10% for European countries and Russia as reported for international statistics (Olivier et al., 2016). We can therefore calculate the residential emission factors uncertainty of each individual pollutant ($\sigma_{EF,p}$) from Eq. 3. In addition, based on the comparison of the recent estimates of wood consumption provided by TNO RWC AD, which should match better with observations and the EDGARv4.3.2 ones, we can evaluate the mean normalized absolute error (MNAE) considering all *N* countries, following Eq. 5 (Yu et al., 2006), which represents our estimate of σ_{AD,RES_bio}.

$$\mathrm{MNAE} = \frac{1}{N} * \sum_j^N \frac{|TNO\ RWC_j - EDGARv4.3.2_j|}{TNO\ RWC_j} \qquad \text{(Eq.5)}$$

We estimate a value of σ_{AD,RES_bio} of 38.9% which is much larger compared to the 5-10% uncertainty assumed for the fuel consumption of the international statistics (σ_{AD}). The issue of biofuel uncertainty mainly affects rural areas where wood is often used instead of fossil fuel. Then, using Eq. 3 and the calculated σ_{AD,RES_bio} and $\sigma_{EF,p}$, we can evaluate a new σ_{EMI,p,RES_bio} for the residential sector including the uncertainty of the AD due to the use of wood as fuel for this sector, as reported in Table S5. Comparing the results shown in Table S5 with the factor of two uncertainty values expected for PM emissions from the residential sector (Janssens-Maenhout et al., 2015), we derive that the uncertainty associated with the emission factors for biomass combustion in the residential sector is the dominant source of uncertainty compared to the AD (wood consumption) uncertainty. Large increases in reported biomass usage for domestic use has been noted in IEA energy statistics for some European countries (IEA, 2013,2014,2015,2016) and further increases are expected as countries are shifting their methodologies to estimate biofuel activity data away from fuel sales statistics to a modeling approach based on energy demand. In addition, several EU countries have planned to increase the use of biomass to accomplish the targets set in the context of the renewable energy directive (2009/28/EC) as reported in their national renewable energy action plans (http://ec.europa.eu/energy/node/71). When comparing the UNFCCC and the TNO RWC data, a higher value of σ_{AD,RES_bio} is obtained (59.5% instead of 38.9%), although its effect on the final residential emission uncertainty is less strong, as shown in Table S6. Table 3 shows the impact of biofuel combustion uncertainty in the residential sector on $PM_{2.5}$ concentrations. Upper-end uncertainties indicate that $PM_{2.5}$ concentrations could be between 2.6 and 3.7 times larger than those derived from the HTAP_v2.2 inventory.

4 Health impact assessment

Annual population weighted $PM_{2.5}$ concentration represents the most robust and widely used metric to analyse the long-term impacts of particulate matter air pollution on human health, as demonstrated by several epidemiological studies (Pope and Dockery, 2006;Dockery, 2009). The mortality estimation in TM5-FASST is based on the integrated exposure-response functions defined by Burnett et al. (2014). The increased risk from exposure to air pollution is estimated using exposure-response functions for five relevant deaths causes, namely Ischemic heart disease (IHD), Cerebrovascular Disease (CD, stroke), Chronic Obstructive Pulmonary Disease (COPD), Lung Cancer (LC), Acute Lower Respiratory Infections (ALRI). The relative risk (RR) represents the proportional increase in the assessed health outcome due to a given increase in $PM_{2.5}$ concentrations (Burnett, 2014).

In this section, we investigate the impact of total and sector-specific anthropogenic population weighted $PM_{2.5}$ concentrations on health and we show comparisons with mortality estimates provided by WHO and scientific publications (Silva et al., 2016). Figure 7 represents the premature deaths (PD) distribution due to air pollution, using population weighted $PM_{2.5}$ concentrations and representative for anthropogenic emissions in the year 2010. The most affected areas are China and India, but also some countries of Western Africa and urban areas in Europe (in particular in the Benelux region and Eastern Europe). Our computations indicate that annual global outdoor premature mortality due to anthropogenic $PM_{2.5}$ amounts to 2.1 million premature deaths, with an uncertainty range related to emission uncertainty of 1-3.3 million deaths/year. In 2010, 82% of the PD occurs in fast growing economies and developing countries, especially in China with 670000 and India with an almost equal amount of 610000 PD/year. Table 4 summarizes our estimates of premature mortality for aggregated world regions, with Europe accounting for 210000 PD/year and North America 100000 PD/year.

Our results are comparable with Lelieveld et al. (2015) and Silva et al. (2016) who estimate using the Burnett et al. (2014) methodology a global premature mortality of 2.5 and 2.2 million people, respectively, due to air quality in 2010 for the same anthropogenic sectors. However, a recent work published by Cohen et al. (2017) estimates a global mortality of 3.9 million PD/year. When comparing mortality estimates we need to take into account that several elements affect the results, like the resolution of the model, the urban increment subgrid adjustment, the inclusion or not of natural components, the impact threshold value used, and RR functions. We also estimate that 7 % of the global non accidental mortalities are advanced by air pollution; 8.6% of total mortality in Europe is due to air pollution, ranging from less than 1% up to 17% depending on the country; similarly, Asian premature mortality due to air quality is equal to 8.7% of total Asian mortality, with 10.6% contribution in China and 8.5% in India. Lower values are found for African countries and Latin America where other causes of mortalities are still dominant compared to developed countries.

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Table 5 shows the number of premature deaths from a source region perspective, highlighting the premature mortality induced by each source region within the country itself and outside the emitting region. The PD induced by Chinese and Indian emissions are mainly found within these two countries; however, the annual PDs caused by China and India in external regions equal 54000 and 76000 PD/year, respectively, representing a high contribution of ca. 10 % to the global mortality. Clearly, reducing emissions and emission uncertainties in these two regions will have therefore the largest over-all benefit on global air quality improvement and understanding as well as on global human health. For most of the TM5-FASST regions, PD due to anthropogenic emissions within the source region are higher than the extra-regional contributions. However, there are marked exceptions, such the Gulf region, Hungary, Czech Republic, Mongolia, etc., where the extra-regional and within-region contributions to mortality are at least comparable.

Detailed information on the premature deaths for each TM5-FASST region and the contributing anthropogenic emission sectors is shown in Figs. 8a and 8b. Health effects induced by air quality in industrialized countries are mainly related with agriculture (32.4% of total mortality or 68000 PD/year), residential combustion (17.8% or 37000 PD/year) and road transport (18.7% or 39000 PD/year) for Europe and with power generation (26.4% or 26000 PD/year), industry (19% or 19000 PD/year), residential (17% or 17000 PD/year) and agriculture (24.0% or 24000 PD/year) for North America. The health impacts observed in most Western EU countries is due both to within-regions and extra-regional pollution, while in several Eastern EU countries the impact of neighbouring countries is even larger compared to within-region pollution. PM related mortality in developing countries and fast growing economies is mostly affected by industrial (up to 42% in China or 279000 PD/year) and residential activities (ranging from 27% in China and 76% in Western Africa), and also by power generation (up to 24% in India or 113000 PD/year). Chinese emissions have a strong impact on China, Japan, Vietnam, Mongolia+Korea, Thailand while the Indian emissions impact the rest of South and South Eastern Asia. Reducing Chinese and Indian emissions will reduce the PM related mortality in almost all countries in Asia. Our results are in agreement with the study of Oh et al. (2015) where they highlight the role of transported pollution from China in affecting Korean and other South Eastern Asian countries $PM_{2.5}$ concentrations and health effects, as well as the need of international measures to improve air quality.

Conclusions

We coupled the global anthropogenic emission estimates provided by the HTAP_v2.2 inventory for 2010 (merging national and regional inventories) to the global source receptor model TM5-FASST, to study $PM_{2.5}$ concentrations and the corresponding health impacts, including an evaluation of the impacts of uncertainties in national emission inventories. Annual and regionally averaged anthropogenic $PM_{2.5}$ concentrations, corresponding to the 2010 emissions, vary between ca 1 and 40 µg/m^3, with the highest annual concentrations computed in China (40 µg/m^3) India (35 µg/m^3), Europe and North America (each 8 µg/ m^3). Anthropogenic $PM_{2.5}$ concentrations are mainly due to emissions within the source region, but extra-regional transported air pollution can contribute by up to 40%, e.g. from China to SE Asia, from EU to Russia, etc.). Moreover, due to the transport of PM between European countries, EU wide directives can help improving the air quality across Europe.

For our analysis we aggregate our results derived from 56 TM5-FASST source regions, into 10 global regions to facilitate the comparison of results in regions of more equal size. The relative contribution of anthropogenic sectors to $PM_{2.5}$ concentrations varies in different regions. In Europe in 2010, the agriculture and residential combustion sectors contribute strongest to $PM_{2.5}$ concentrations and these sectors are also associated with relatively large emission uncertainties. $PM_{2.5}$ concentrations in China and other emerging economies are predominantly associated with the power generation, industry and residential activities.

Using HTAP_v2.2 and TM5-FASST, we also evaluate how the uncertainty in sectors and regions propagates to $PM_{2.5}$. The aim of our analysis is to provide insights on where the emission inventories of each country could be improved, because of their highest uncertainty and highest contribution to the formation of $PM_{2.5}$ concentrations. The uncertainty of PM concentrations depends in variable proportions to the uncertainties of the emissions within receptor regions, and surrounding regions. We show that reducing the uncertainties in the Chinese and Indian emission inventories (e.g. from industry and residential sectors) will be highly relevant for understanding the long-range sources of $PM_{2.5}$ pollution in most of Asian countries. Here we demonstrate how analysis of uncertainties in national/regional sectorial emission inventories can further inform coordinated transboundary and sector-specific policies to significantly improve global air quality. Among all anthropogenic emission sectors, the combustion of biomass for household purposes represents one of the major sources of uncertainties in emission inventories both in terms of wood consumption and emission factor estimates. Further effort is therefore required at national level to better characterize this source.

Finally, we analyse the air quality effects on health. Global health effects due to $PM_{2.5}$ concentrations calculated with TM5-FASST and anthropogenic emissions in 2010 are estimated to be ca 2.1 million premature deaths/year, but the uncertainty associated with emission ranges between 1-3.4 million deaths/year, of which the largest fraction (82%) occurs in developing countries.

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Tables and Figures

Table 1 - Sector specific contribution [%] to annual anthropogenic $PM_{2.5}$ concentrations for aggregated world region. The largest contributing sectors (above a threshold of 15%) are shaded in blue.

	POWER	INDUSTRY	TRANSPORT	RESIDENTIAL	AGRICULTURE	SHIP
Africa	26.7	16.1	3.6	37.9	8.2	7.4
China+	18.3	42	7.5	23.1	8.8	0.3
India+	20.8	19.4	11.4	45.2	3	0.2
SE Asia	17.1	35.9	9	27.2	7.4	3.4
Europe	15.1	14.3	18.7	19.7	27.7	4.4
Latin America	25.6	33.7	6.6	18.9	12.6	2.6
Middle East	37.9	25.2	9.7	11.7	13.7	1.8
Russia	23.5	30.9	8.6	13	23.1	0.8
North America	20.4	23.5	10.8	15.5	25.6	4.2
Oceania	13.9	30.7	5.1	9.8	18.6	21.8

Table 2 - Annual average $PM_{2.5}$ concentrations ($\mu g/m^3$) with upper and lower range in brackets due to emission inventories uncertainty (σ). The upper and lower range of $PM_{2.5}$ concentrations are calculated as the reference concentrations multiplied and divided by ($1+\sigma$) respectively. Uncertainty due to the contribution of primary $PM_{2.5}$ emissions.

World region	TM5-FASST region	$PM_{2.5}$ concentration ($\mu g/m^3$)	Fraction of uncertainty due to primary PM emissions (%)
Asia	South Korea	13.8 (8.3 - 24.9)	71%
	Japan	6.9 (3.8 - 13.3)	84%
	Mongolia+ North Korea	14.6 (9.0 - 25.9)	75%
	China	39.9 (22.4 - 76.6)	78%
	Taiwan	6.4 (3.7 - 10.9)	77%
	Rest of South Asia	29.3 (13.9 - 64.9)	87%
	India	34.7 (16.6 - 73.4)	86%
	Indonesia	2.4 (1.3 - 4.6)	86%
	Thailand	8.0 (5.1 - 12.6)	88%
	Malaysia	3.1 (1.8 - 5.2)	85%
	Philippines	2.0 (1.1 - 3.8)	80%
	Vietnam	14.2 (7.0 - 30.4)	92%
	Rest of South Eastern Asia	8.6 (4.6 - 17.6)	89%
Europe	Austria+Slovenia	8.4 (4.0 - 19.6)	59%
	Switzerland	10.1 (4.9 - 23.3)	52%
	Benelux	10.1 (5.2 - 22.7)	59%
	Spain+Portugal	5.4 (3.4 -9.4)	77%
	Finland	2.6 (1.3 - 5.8)	66%
	France	9.3 (5.0 - 19.0)	69%
	Great Britain+Ireland	6.1 (3.2 - 13.0)	66%
	Greece+Cyprus	7.6 (4.8 - 12.7)	74%
	Italy+Malta	11.8 (6.2 - 25.2)	64%
	Germany	9.3 (5.0 - 20.0)	54%
	Sweden+Denmark	4.1 (2.2 - 8.4)	65%
	Norway	2.4 (1.2 - 5.4)	89%
	Bulgaria	10.6 (5.4 - 21.6)	66%
	Hungary	9.2 (4.4 - 21.6)	60%
	Poland+Baltic	7.9 (3.6 - 20.2)	54%
	Rest of Central EU	9.3 (4.7 – 20.4)	63%
	Czech Republic	10.3 (4.8 - 25.1)	58%
	Romania	10.9 (5.5 - 24.1)	67%

1

World region	TM5-FASST region	$PM_{2.5}$ concentration ($\mu g/m^3$)	Fraction of uncertainty due to primary PM emissions (%)
Africa	Northern Africa	4.2 (2.3 - 4.3)	80%
	Egypt	11.0 (5.0 - 27.8)	46%
	Western Africa	4.0 (1.7 - 10.2)	96%
	Eastern Africa	2.7 (1.4 - 5.7)	89%
	Southern Africa	1.0 (0.5 - 2.2)	90%
	Rep. of South Africa	6.1 (3.1 - 12.5)	84%
Gulf/ Middle East	Middle East	9.2 (5.4 - 17.8)	58%
	Turkey	8.7 (4.9 - 17.1)	67%
	Gulf region	7.8 (4.7 - 14.5)	57%
Latin America	Brazil	1.6 (1.1 - 2.6)	85%
	Mexico	4.2 (2.1 - 9.2)	62%
	Rest of Central America	2.0 (1.0 - 4.0)	78%
	Chile	13.7 (7.3 - 29)	70%
	Argentina+Uruguay	1.1 (0.7 - 1.9)	77%
	Rest of South America	2.4 (1.6 - 3.9)	69%
NA	Canada	4.3 (2.4 - 8.3)	66%
	USA	7.8 (4.4 - 14.4)	71%
Russia	Kazakhstan	4.9 (3.2 - 8.9)	62%
	Former USSR Asia	7.5 (4.0 - 17.6)	49%
	Russia (EU)	3.3 (1.9 - 6.7)	57%
	Russia (Asia)	2.7 (1.7 - 5.1)	64%
	Ukraine	7.8 (4.2 - 15.9)	65%
Oceania	Australia	1.1 (0.8 - 1.4)	84%
	New Zealand	0.3 (0.1 - 0.5)	60%
	Pacific Islands	0.2 (0.1 - 0.4)	75%

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Table 3 - $PM_{2.5}$ concentrations due to the residential sector emissions in Europe, Russia (EU), Ukraine and Turkey and uncertainty range including the uncertainty in the biomass consumption for the same sector.

	$PM_{2.5}$ (μg/m^3) - RESIDENTIAL	$PM_{2.5}$ (μg/m^3)- RESIDENTIAL including biomass uncertainty
Romania	3.1	11.4
Czech Republic	2.9	10.7
Italy+Malta	3.6	10.6
Rest of Central EU	2.5	9.2
Hungary	2.5	9.1
Bulgaria	2.3	8.6
Poland+Baltic	2.2	8.3
Austria+Slovenia	2.2	7.1
Ukraine	1.7	6.1
France	2.1	6.0
Turkey	1.7	5.9
Norway	1.3	4.1
Switzerland	1.4	3.9
Greece+Cyprus	1.2	3.8
Germany	1.1	3.0
Spain+Portugal	1.0	2.7
Benelux	0.9	2.5
Sweden+Denmark	0.8	2.4
Finland	0.7	2.1
Great Britain+Ireland	0.7	1.8
Russia (EU)	0.4	1.3

Table 4 – Number of premature deaths/year due to anthropogenic $PM_{2.5}$ air pollution in world regions and corresponding uncertainty range.

	PD (deaths/year)
China+	6.7E+05 (3.5E+05 - 1.0E+06)
India+	6.1E+05 (2.7E+05 - 9.6E+05)
Europe	2.6E+05 (1.4E+05 - 4.8E+05)
SE Asia	1.5E+05 (8.3E+04 - 2.5E+05)
Russia	1.1E+05 (6.7E+04 - 2.4E+05)
North America	1.0E+05 (5.5E+04 - 1.7E+05)
Africa	7.4E+04 (3.4E+04 - 1.6E+05)
Middle East	5.6E+04 (3.2E+04 - 9.7E+04)
Latin America	2.6E+04 (1.4E+04 - 5.3E+04)
Oceania	5.5E+01 (3.4E+01 - 1.2E+02)

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Table 5 – Number of premature deaths caused by major source regions and their contribution to global mortality (within-region and extra-regional attribution).

world regions	TM5-FASST FASST codes	PD induced by source region (deaths/year)	Within-region PD (deaths/year)	Extra-regional PD (deaths/year)
Africa	Eastern Africa	9451	8218	1233
Africa	Egypt	11137	10783	354
Africa	Northern Africa	3904	3427	477
Africa	Rep. of South Africa	8813	8797	15
Africa	Southern Africa	248	32	216
Africa	Western Africa	19785	19785	0
Asia	China	696823	643129	53694
Asia	Indonesia	15352	14803	549
Asia	India	488319	412298	76021
Asia	Japan	15181	15181	0
Asia	South Korea	8789	7510	1279
Asia	Mongolia+ North Korea	8786	4076	4710
Asia	Malaysia	2225	1058	1167
Asia	Philippines	94	94	0
Asia	Rest of South Asia	113040	67170	45870
Asia	Rest of South Eastern Asia	4064	3814	250
Asia	Thailand	10898	10495	403
Asia	Taiwan	1028	1028	0
Asia	Vietnam	24401	20286	4115
Europe	Austria+Slovenia	3668	1674	1994
Europe	Bulgaria	5986	2269	3717
Europe	Benelux	12991	6057	6933
Europe	Switzerland	3036	1404	1632
Europe	Czech Republic	8957	3540	5417
Europe	Germany	33343	23001	10342
Europe	Spain+Portugal	10454	9541	914
Europe	Finland	0	0	0
Europe	France	23901	15148	8753
Europe	Great Britain+Ireland	12588	11157	1431
Europe	Greece+Cyprus	2112	1520	592
Europe	Hungary	4629	3889	740
Europe	Italy+Malta	18541	17373	1168
Europe	Norway	26	26	0
Europe	Poland+Baltic	23825	16811	7014
Europe	Rest of Central EU	9570	6239	3331
Europe	Romania	15374	8360	7014
Europe	Sweden+Denmark	90	88	2
Latin America	Argentina+Uruguay	114	75	39

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Latin America	**Brazil**	4089	3968	120
Latin America	**Chile**	3391	3283	108
Latin America	**Mexico**	9410	8447	964
Latin America	**Rest of Central America**	3569	2772	797
Latin America	**Rest of South America**	4205	4164	41
Middle East	**Golf region**	34270	11225	23046
Middle East	**Middle East**	3993	2804	1189
Middle East	**Turkey**	32442	24191	8252
North America	**Canada**	5279	1491	3788
North America	**USA**	92885	90176	2709
Oceania	**Australia**	1010	25	985
Oceania	**New Zealand**	15	15	0
Oceania	**Pacific Islands**	1	1	0
Russia	**Kazakhstan**	2000	1100	900
Russia	**Former USSR Asia**	7419	6420	999
Russia	**Russia (Asia)**	3607	601	3006
Russia	**Russia (EU)**	19419	12704	6714
Russia	**Ukraine**	57352	44604	12748

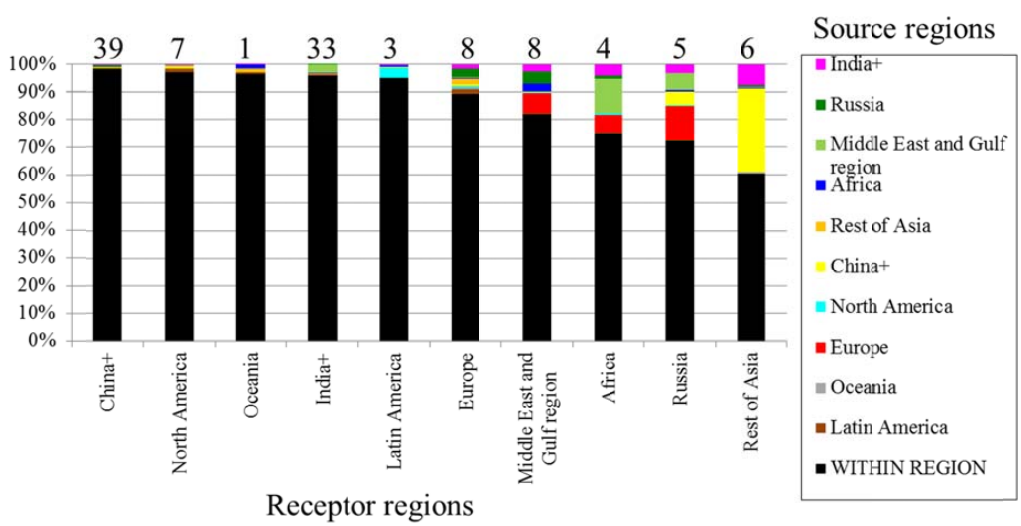

Figure 1 – Within-region vs. imported extra-regional anthropogenic $PM_{2.5}$ concentrations [%] for aggregated world regions. Annual average population weighted anthropogenic concentrations (in $\mu g/m^3$) are reported on top of each bar. Ship emissions were not included.

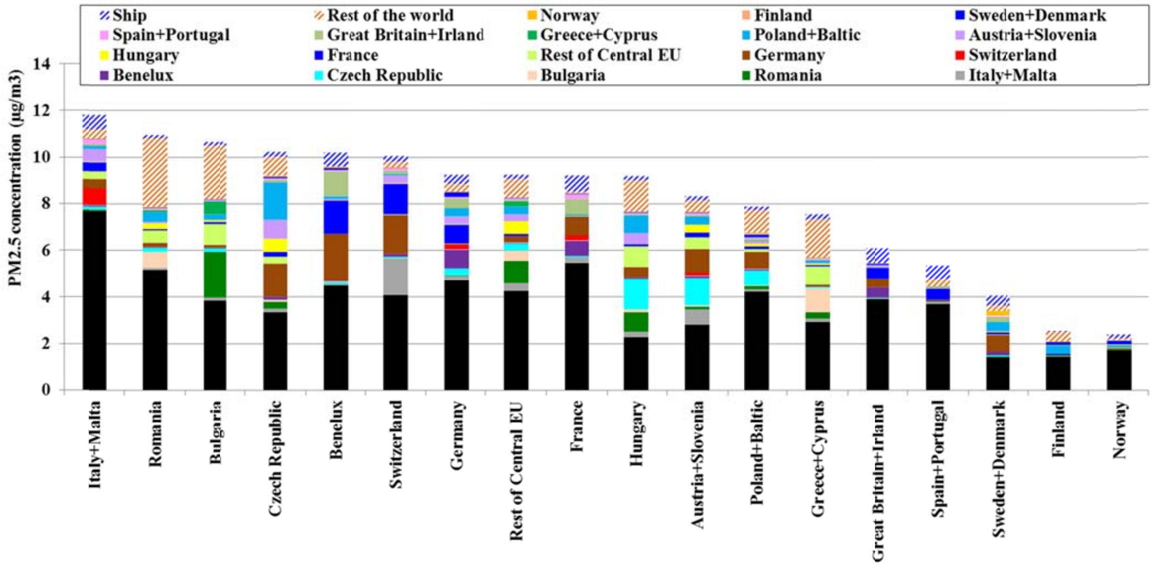

Figure 2 – Anthropogenic $PM_{2.5}$ concentrations in 18 countries and sub-regions in Europe separated in within-region and extra-regional contributions.

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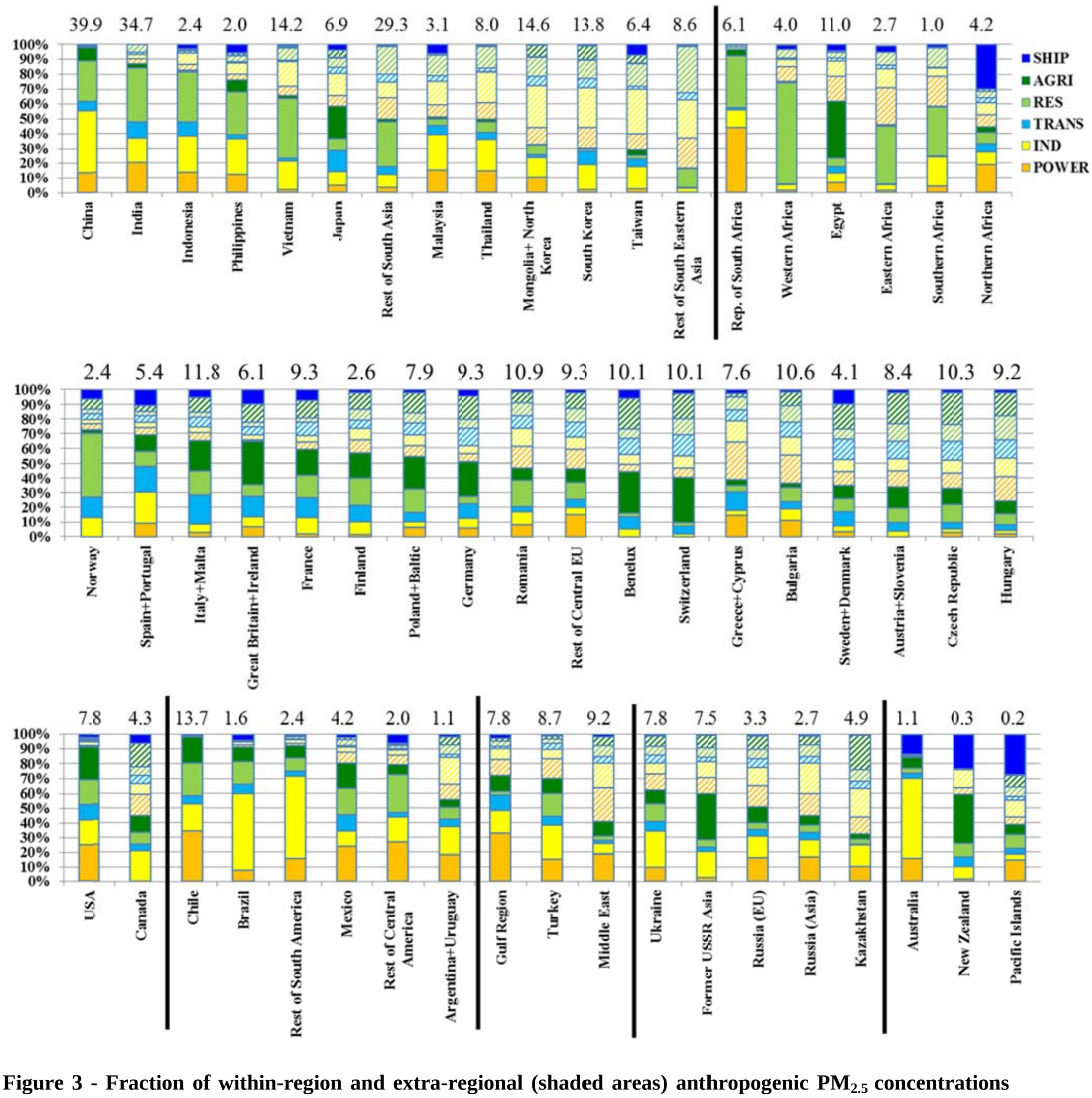

Figure 3 - Fraction of within-region and extra-regional (shaded areas) anthropogenic $PM_{2.5}$ concentrations separate by sector for receptor region within the macro-regions: Asia and Africa (upper panel), Europe (middle panel), North America, Latin America, Middle East, Russia and Oceania (lower panel). Annual averaged anthropogenic concentrations (in µg/m^3) are reported on top of each bar.

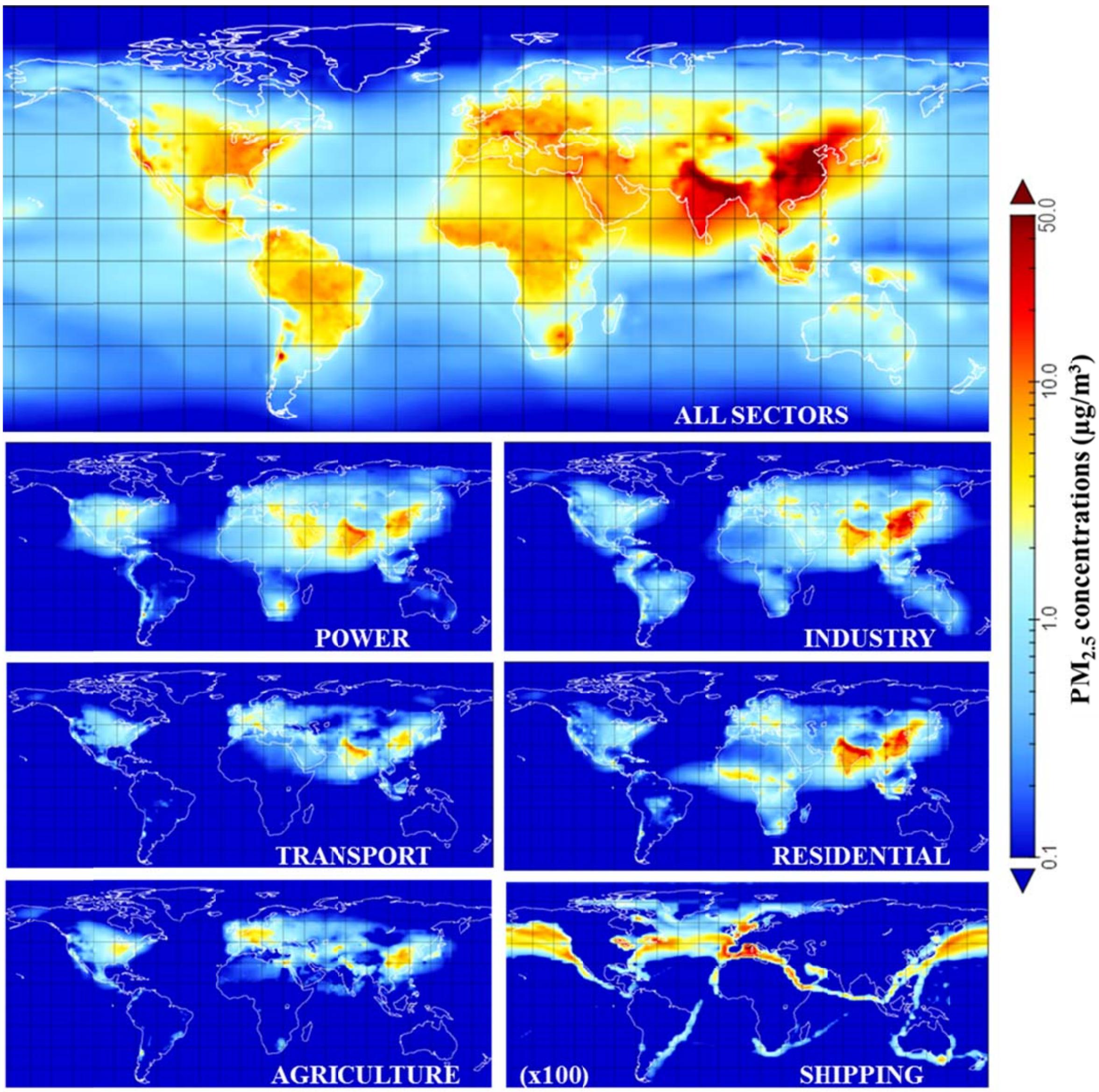

Figure 4 – Total anthropogenic $PM_{2.5}$ concentrations (μg/m^3) and sectorial contributions using 2010 emissions.

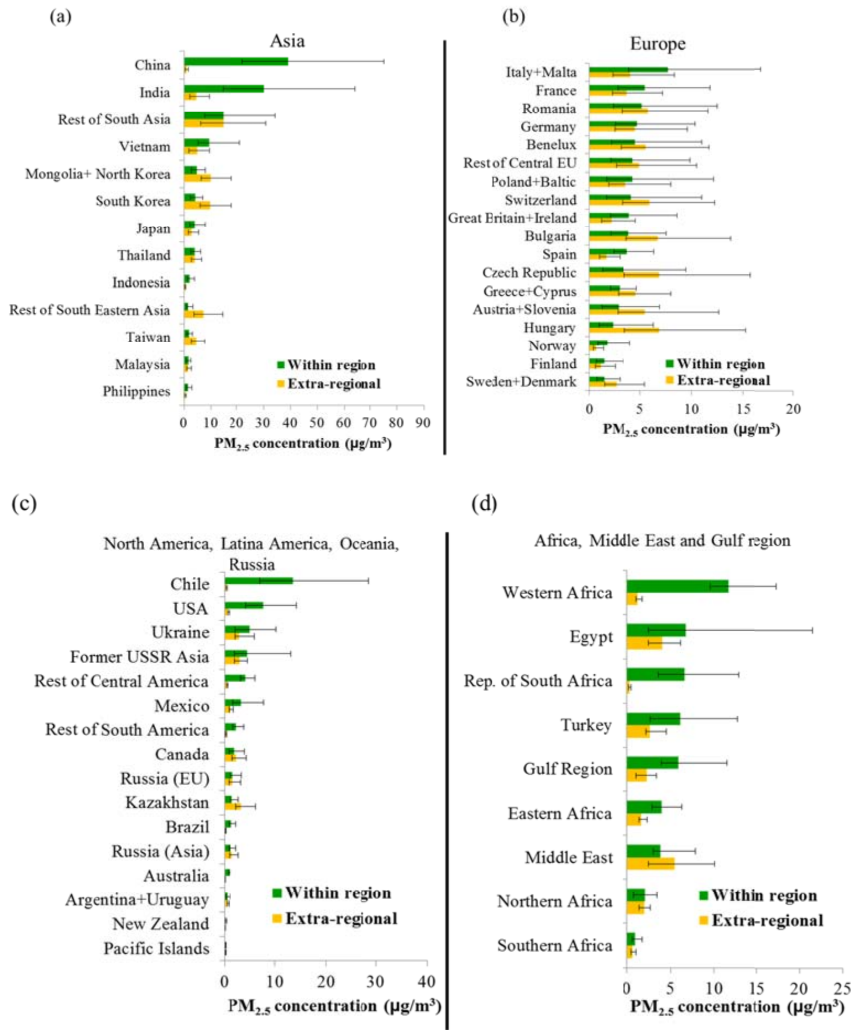

Figure 5 - Within-region and extra-regional anthropogenic $PM_{2.5}$ concentrations for Asia (panel *a*), Europe (panel *b*), North America, Latin America, Oceania and Russia (panel *c*) and Africa, Gulf region and Middle East (panel *d*). The error bars are calculated multiplying and dividing the reference emissions by (1+σ) as discussed in Sect. 2.3.

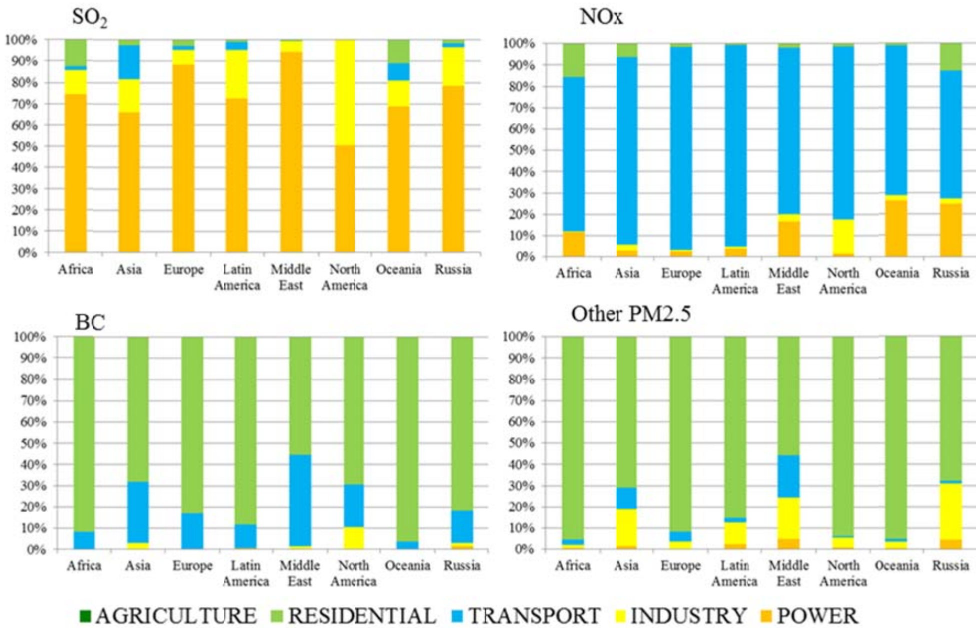

Figure 6 – Contribution of anthropogenic sectors to the emission uncertainty of various pollutants S for different world regions.

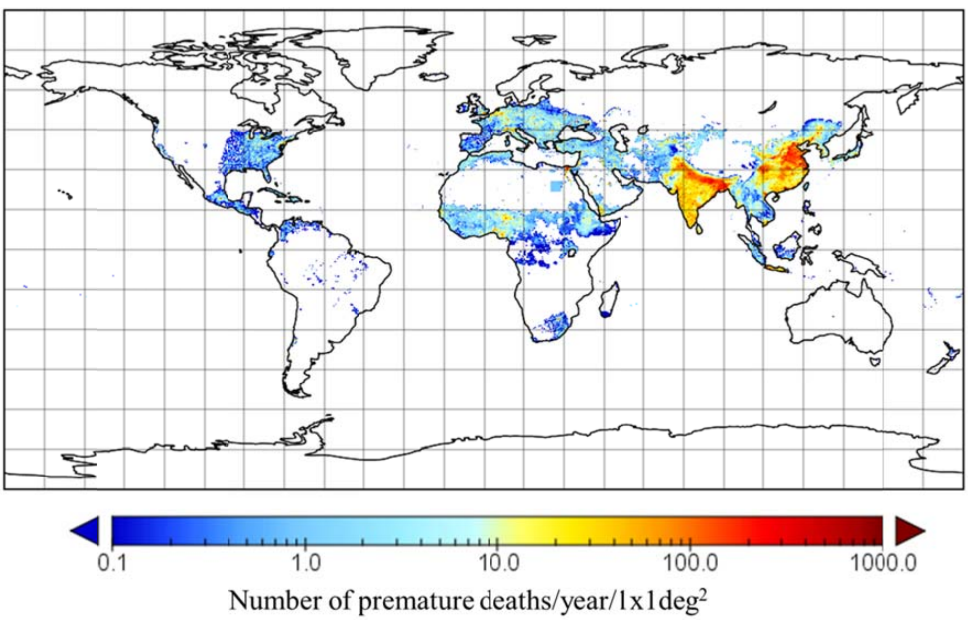

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Figure 7 – Global distribution of premature deaths in 2010 caused by anthropogenic particulate matter pollution estimated using the methodology described in Burnett et al. (2014).

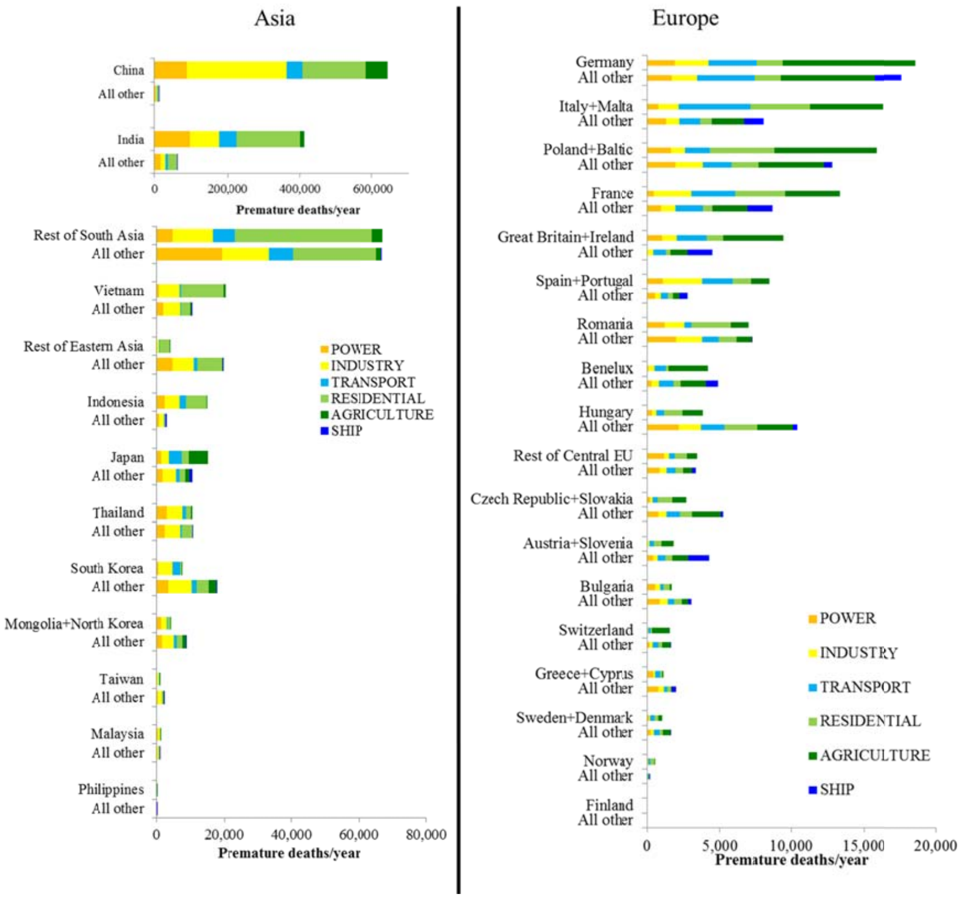

Figure 8a – Anthropogenic emission sector contributions to premature mortality (deaths/year) due to $PM_{2.5}$ population weighted concentrations in the TM5-FASST receptor regions of Asia and Europe.

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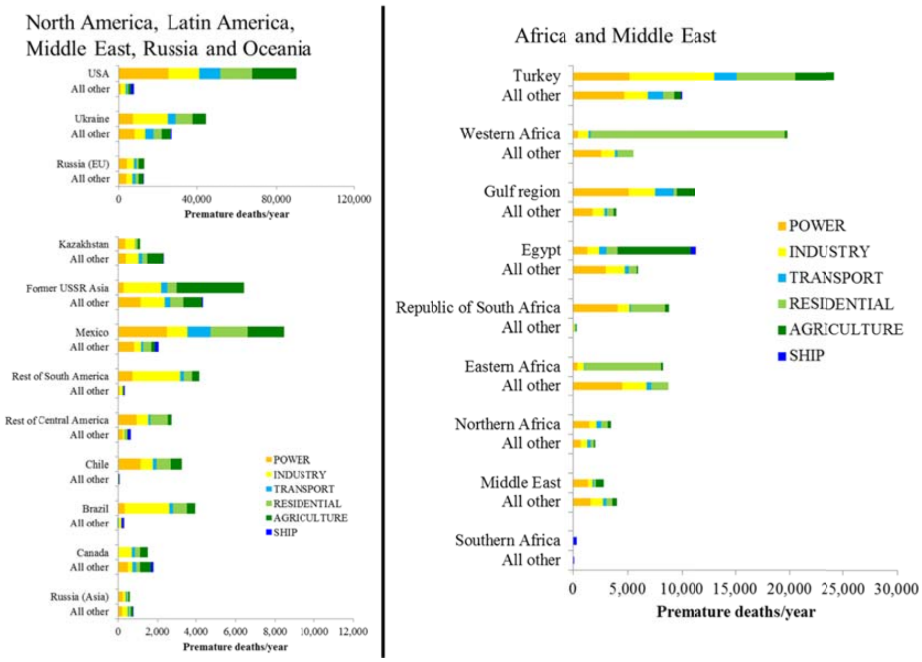

Figure 8b – Anthropogenic emission sector contributions to premature mortality (deaths/year) due to $PM_{2.5}$ population weighted concentrations in the TM5-FASST receptor regions of North America, Latin America, Russia, Middle East and Oceania (left hand side) and Africa (right hand side). Note that mortality estimates for Argentina+Uruguay, Australia, New Zealand and Pacific Islands are not reported being several orders of magnitude lower than other countries estimates.