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Contribution and uncertainty of sectorial and regional emissions to regional and global PM$_{2.5}$ health impacts

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Abstract. In this work we couple the HTAP_v2.2 global air pollutant emission inventory with the global source receptor model TM5-FASST to evaluate the relative contributions 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 well-documented cumulative negative effects on human health. We estimate that in 2010, depending on the region, annual averaged anthropogenic PM$_{2.5}$ concentrations varied between ca. 1 and 40 µg m^{-3}, with the highest concentrations observed in China and India, and lower concentrations in Europe and North America. The relative contribution of anthropogenic emission sources 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 air quality improvements would require regional policies, in addition to local- and urban-scale measures, due to the transboundary features of PM pollution. 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 modelling. 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. Countries with high emission uncertainties are often impacted by the uncertainty of pollution coming from surrounding regions, highlighting the need for effective efforts in improving emissions not only within a region but also from extra-regional sources. Finally, we propagate emission inventory uncertainty to PM concentrations and health impacts. We estimate 2.1 million premature deaths per year with an uncertainty of more than 1 million premature deaths per year due to the uncertainty associated only with the emissions.

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., 2013; Anderson et al., 2012; Anenberg et al., 2009; Cohen et al., 2017). 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 reduce health impacts of air pollution. These policies are usually implemented under national legislation (Henneman et al., 2017; Morgan, 2012), while in Europe transboundary air pollution is also addressed by the regional protocol under the UNECE Convention on Long-Range Transboundary of Air Pollution (CLRTAP). At city and local level, several studies have been developed to assess the contribution of sector-specific emissions to PM$_{2.5}$ (particulate mat-

ter with a diameter less than 2.5 µm) concentrations with the aim of designing air quality plans at local and regional level (Karagulian et al., 2015; Thunis et al., 2016). Indeed, particulate matter can travel thousands of kilometres, crossing national borders, oceans and even continents (HTAP, part A, Dentener et al., 2010). Local, regional and international coordination is therefore needed to define air pollution policies to improve global air quality and possibly 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, to identify promising mitigation measures to reduce background pollution levels and their contribution to pollution in rural as well as urban regions. Although primary $PM_{2.5}$ and intermediately lived (days-to-weeks) precursor gases 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, transport and removal processes of gaseous precursors transported out of source regions (Maas and Grennfelt, 2016). However, the most extreme episodes of exposure often occur under extended periods of low wind speeds and atmospheric stability, favouring the formation of secondary aerosols close to the source regions. 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 sulfur dioxide (SO_2), nitrogen oxides (NO_x) and ammonia (NH_3) – and organic compounds involving thousands of compounds and often poorly known reactions (Hallquist et al., 2009). Exposure to and impact from aerosols on humans can be estimated by a variety of approaches, ranging from epidemiological studies to pure modelling approaches. The Burnett et al. (2014) risk–response methodology is often used in models to estimate premature deaths (PDs) due to air pollution exposure, e.g. in Lelieveld et al. (2015) and Silva et al. (2016), who report a global mortality in 2010 due to air quality issues induced by anthropogenic emissions of 2.5 and 2.2 million people, respectively. A higher global mortality is found in a more recent work by Cohen et al. (2017), who account for 3.9 million premature deaths per year due to different model assumptions. In Europe, Brant et al. (2013) estimate 680 000 premature deaths, which is twice as high as the numbers reported for the CAFE (Clean Air for Europe) study (Watkiss et al., 2005). Recently, using the same emission database as in this study, Im et al. (2018) report a multi-model mean estimate of PD of 414 000 (range 230–570 000) for Europe and 160 000 PDs for the USA. 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 to incomplete understanding of transport, chemical transformation, removal processes and, not least, emission information.

This work is developed in the context of the TF HTAP Phase 2 (Galmarini et al., 2017), where a number of models are deployed to assess long-range sensitivities to extra-regional emissions, using the same HTAP_v2.2 anthropogenic 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 (Liang et al., 2018), but not of uncertainties in emission inventories. The objectives and novelties of this study are the evaluation of (i) the relative contribution of anthropogenic emission sources to $PM_{2.5}$ concentrations at global scale, (ii) the emission sectors and emission regions in which pollution reduction measures would lead to the largest improvement on the overall air quality, and (iii) the relevance of uncertainties in regional sectorial emission inventories (power generation, industry, ground transport, residential, agriculture and international shipping), and their propagation in modelled $PM_{2.5}$ concentrations and associated impacts on health. This work applies the global source–receptor model TM5-FASST (TM5-FAst Scenario Screening Tool), which is extensively described and evaluated in this special issue (Van Dingenen et al., 2018), and couples it to the HTAP_v2.2 global emission inventory for the year 2010 to estimate global air quality and associated health impacts in terms of $PM_{2.5}$ concentrations. The regional and global scale and the focus on annual $PM_{2.5}$ and associated health metrics warrant the use of the TM5-FASST model. However, the most extreme episodes of pollution may occur at more local-to-regional scales, justifying the need for local measures. For instance, a recent study performed over hundreds of cities in Europe (Thunis et al., 2017) shows that in order to comply with the standards prescribed by the Air Quality Directives and the health guidelines by WHO, local actions at the city scale are needed.

Specifically, we show that the impact of emission inventory uncertainty on mortality estimates is comparable with the range of uncertainty induced by air quality models and population exposure functions. We also investigate the uncertainties in $PM_{2.5}$ from within the region to extra-regional contributions. Based on our analysis of the importance of emission uncertainties at sector and regional level on $PM_{2.5}$, we aim at informing local, regional and hemispheric air quality policy makers on the potential impacts of sectors with larger uncertainties (e.g. residential and agriculture) or regions (e.g. developing and emerging countries).

2 Methodology

2.1 TM5-FASST model and emission perturbations

This work is an application of the TM5-FASST model, which is extensively documented in a companion publication in this special issue. Van Dingenen et al. (2018) provide an extensive evaluation of the model, model assumptions and perfor-

mance with regard to the linearity and additivity of concentration response to different size of emission perturbations and future emission scenarios. Below we summarise the most important features of relevance for this work and refer for more detail to Van Dingenen et al. (2018).

In order to calculate $PM_{2.5}$ concentrations corresponding to the HTAP_v2.2 emissions, we use the native $1° \times 1°$ resolution source–receptor grid maps obtained for TM5-FASST_v0 (Van Dingenen et al., 2018). The TM5-FASST source–receptor model is based on a set of emission perturbation experiments (-20%) of SO_2, NO_x, CO, NH_3, and volatile organic compounds (VOCs) and CH_4 using the global $1° \times 1°$ resolution TM5 model, the meteorological year 2001 (which was also used for the HTAP Phase 1 experiments) and the community emission dataset prepared for the IPCC AR5 report (RCP, Representative Concentration Pathway) for the year 2000 (Lamarque et al., 2010). TM5-FASST uses aggregated regional emissions (i.e. one annual emission value per pollutant or precursor for each of the 56 regions + shipping), with an implicit underlying $1° \times 1°$ resolution emission spatial distribution from RCP year 2000 which was partly based EDGAR methodology and grid maps. 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 secondary organic aerosol (SOA) from natural sources is included in the model calculations using the parameterisation described in Dentener et al. (2006), no explicit treatment of anthropogenic SOA is considered, since no reliable emission inventories of SOA precursor gases was available, and formation processes were not included in the parent TM5 model. A recent study by Farina et al. (2010) indicates a global source of 1.6 Tg, or ca. 5.5 % of the overall SOA formation due to anthropogenic SOA. The relative importance of anthropogenic SOA varies widely by region, and is deemed higher in regions with fewer VOC emission controls. Inclusion of SOA would possibly lead to a somewhat larger role of the transboundary pollution transport (Farina et al., 2010; Peng et al., 2016; Shiraiwa et al., 2017), mainly for regions and sectors with large PM and VOC emissions (e.g. residential, and to some extent transport and industry).

Under the assumption that the individual sector contributions add up linearly to total $PM_{2.5}$ – this assumption is evaluated in Van Dingenen et al. (2018) and holds in most regions within 15 % error – 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.

Specifically, the reduced-form model TM5-FASST is computing the concentration resulting from an arbitrary precursor emission strength E_i using a first-order perturbation approach, i.e. for each PM component j, the change in concentration dPM_j resulting from a change in emission strength $E_i(x)$ of precursor i in source region x, relative to a reference emission $E_{i,\text{ref}}(x)$, is approximated by the first linear term of a Taylor expansion of PM as a function of emissions:

$$dPM_j(y) \cong A_{ij}[x,y][E_i(x) - E_{i,\text{ref}}(x)], \qquad (1)$$

where

$$A_{ij}[x,y] = \frac{\Delta C_j(y)}{\Delta E_i(x)} \text{ with } \Delta E_i(x) = 0.2 E_{i,\text{ref}}(x). \qquad (2)$$

$A_{ij}[x,y]$ is a set of independently computed source–receptor matrices, expressing the linearised emission–concentration response between each relevant precursor (i) emission and PM component j concentration, for each pair of source (x) and receptor (y) regions (Van Dingenen et al., 2018).

In Sect. S1.2 we explain in detail how Eq. (1) can also be applied for evaluating the attribution by sector as well as by source region, based on the work by Van Dingenen et al. (2018). Thus to calculate total $PM_{2.5}$ concentration in each receptor region, the 56 source regions' individual contributions must be summed. Using this approach, it is possible to evaluate the $PM_{2.5}$ concentrations from "within-region" and "extra-regional" $PM_{2.5}$ emissions. The extra-regional contribution represents the RERER metric (Response to Extra-Regional Emission Reduction) for a specific region used across the whole HTAP experiment (Galmarini et al., 2017), in particular focusing on the $PM_{2.5}$ concentration reduction due to the contribution of the emissions of each anthropogenic sector (Eq. 3):

$$\text{RERER} = \frac{\sum R(\text{foreign regions})}{\sum R(\text{all regions})}, \qquad (3)$$

where R represents the concentration response to each sector emission decrease.

As depicted in Fig. S1 in the Supplement, the 56 TM5-FASST regions cover the entire globe, 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 the evaluation of external impacts on smaller regions (e.g. European countries) comparable to those of larger regions (like the USA, China and India), in this work an aggregation procedure to 10 world regions (refer to Table S2 in the Supplement) has been applied (China+, India+, SE Asia, North America, Europe, Oceania, Latin America, Africa, Russia and the 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 an exposed population following the methodology developed by Burnett et al. (2014), as discussed in Sect. 4. Health impacts due to indoor air pollution or ozone are not evaluated in this work.

In the following, we will address the uncertainty of sector-specific emissions from this inventory in a quantitative way as well as the differences we observe from one region to the other, based on the uncertainty of activity data and emission factors. As discussed in the next section, the reason to use HTAP_v2.2, and not for example the RCP2000 as the basis for our assessment of emission propagation is that the TF HTAP aims at bringing policy-relevant information, and to this end, it has compiled a policy-relevant emission inventory (HTAP_v2.2) for the most recently available year. While the RCP2000 was at the basis of the FASST calculations, and presented the best community emissions effort at the time, the HTAP_v2.2 inventory is now much more accurate, in particular given the focus on regional emission analysis of our work.

2.2 HTAP_v2.2 emissions

The global anthropogenic emission inventory HTAP_v2.2 for the year 2010 (Janssens-Maenhout et al., 2015) 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, NO_x, 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). Here we focus on the six 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"). International and domestic aviation emissions are not considered in this study due to the lower contribution to air pollution compared to other anthropogenic sectors. It should be noted that agricultural emissions do not include agricultural waste burning and forest and savannah fires. 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 emission perturbation scenarios has been created by subtracting from the reference dataset the emissions of each sector.

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 (EFs); therefore the corresponding uncertainty ($\sigma_{i,c,p}$) is calculated as following:

$$\sigma_{\text{EMI}i,c,p} = \sqrt{\sigma_{\text{AD}i,c}^2 + \sigma_{\text{EF}i,p,c}^2},$$ (4)

where σ_{AD} and σ_{EF} are the uncertainties (%) of the activity data and emission factors for a certain sector (i), country (c) and pollutant (p). Uncertainty values of the activity data by sector and country are obtained from Table 2 of Janssens-Maenhout et al. (2017) and Olivier et al. (2016). Using this approach, the uncertainty in the global total anthropogenic CO_2 emissions is estimated to range from −9 % to +9 % (95 % confidence interval), with larger uncertainties of about ±15 % for non-Annex I countries, and uncertainties of less than ±5 % are obtained for the 1990 OECD countries[1] for the time series from 1990 (Olivier et al., 2016) reported to UNFCCC. 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. In this work we assume that reported countries' emissions are based on independent estimations of activity data and EFs, and hence no cross-country correlation structure is assumed. This is in contrast to bottom-up gridded emission inventories like EDGAR, where the use of global activity datasets may lead to correlated errors between countries.

Therefore, we can calculate the overall uncertainty $\sigma_{\text{EMI}p,c}$ with the following equation (EMEP/EEA, 2013).

$$\sigma_{\text{EMI}p,c} = \sqrt{\sum_i \left(\sigma_{\text{EMI}i,c,p} \cdot \frac{\text{EMI}_{i,c,p}}{\text{EMI}_{\text{tot},c,p}} \right)^2},$$ (5)

where $\text{EMI}_{i,c,p}$ (in kt) represents the emission of a certain pollutant (p) in a certain country (c) from a specific sector (i) and $\text{EMI}_{\text{tot},c,p}$ (in kt) 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 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. Based on the upper and lower emission range per region, new TM5-FASST model runs have been performed per source region to retrieve the corresponding range of concentrations in receptor regions (therefore the total number of computations is $56 \cdot 2$ for the uncertainty analysis).

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[1] OECD countries in 1990: Australia, Austria, Belgium, Canada, Denmark, Finland, France, Germany, Greece, Iceland, Ireland, Italy, Japan, Luxembourg, Netherlands, New Zealand, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, United Kingdom, United States.

3 TM5-FASST modelling results

In this section, we first provide "central" estimates of regional (Sect. 3.1), sectorial (Sect. 3.2) and gridded (Sect. 3.3) contributions, whereas the corresponding uncertainty estimates are discussed from Sect. 3.4 onward.

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 world receptor regions using emissions of the year 2010, with the extra-regional fraction (using the RERER metric) broken down into source region contributions. Annual average population-weighted anthropogenic PM$_{2.5}$ concentrations (refer to Van Dingenen et al., 2018, for the calculation of this metric) range from a few µg m^{-3} (e.g. in Oceania or Latin America), around 7–8 µg m^{-3} for North America and Europe, 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; RERER 2 %–4 %), although 4 % of the Indian anthropogenic PM$_{2.5}$ pollution is mainly transported from the Gulf region and the Middle East, as was also observed by Venkataraman et al. (2018). North America (98 %) and Oceania (98 %) are mainly influenced by within-regional pollution due to their geographical isolation from other regions. TM5-FASST computations attribute 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 the Middle East), and Russia 28 % (mainly from Europe, the Middle East and the Gulf region, and China). Shipping emissions are not considered in this figure due to their international origin, while inland waterway emissions are still included in the ground transport sector. Transboundary air pollution is known to be an important issue in the rest of Asia, in particular for pollution transported from China to North and South 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. Regional annual averages of population-weighted PM$_{2.5}$ concentrations in Europe vary between 2 and 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 the computed annual average PM$_{2.5}$ concentrations for Europe 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 in urban areas are often observed in Europe. As further 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). Transboundary air pollution from external regions contributes by 27 % to 75 % and on average by 51 % to PM$_{2.5}$ pollution in European countries. 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, the Czech Republic, the rest of central EU, Poland and Germany), the Czech Republic (67 %, mainly from Poland, Germany and Austria), Austria and Slovenia (66 %, mainly from the Czech Republic, Germany and Italy), Sweden + Denmark (65 %, mainly from Germany, Norway and Poland), Bulgaria (63 %, mainly from Romania), and Greece (61 %).

The remaining EU countries are affected by both 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; the rest of the central EU by Poland and Germany; Germany by France and Benelux; and Poland by the Czech Republic and Germany. Interestingly, Romania, Bulgaria, Greece and Hungary are also significantly affected by the pollution transported from 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), which highlights the importance of transboundary transport of organic and inorganic PM. As discussed in Sect. 3.4, insights into the uncertainty of within-region and extra-regional contributions to PM$_{2.5}$ concentrations are provided in Fig. 5 for each TM5-FASST region.

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 (RERER) (shaded colours) 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 Mediter-
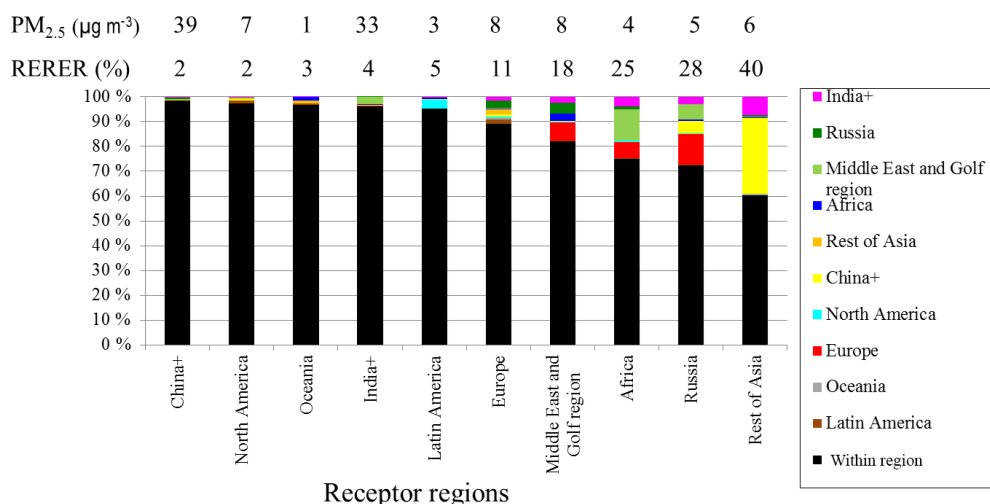
Figure 1. Within-region vs. imported extra-regional anthropogenic population-weighted PM$_{2.5}$ concentrations (%) for aggregated world regions based on "central" estimates. Annual average population-weighted anthropogenic concentrations (in µg m^{-3}) are reported on top of each bar together with the RERER metric (%). Shipping emissions are not included.
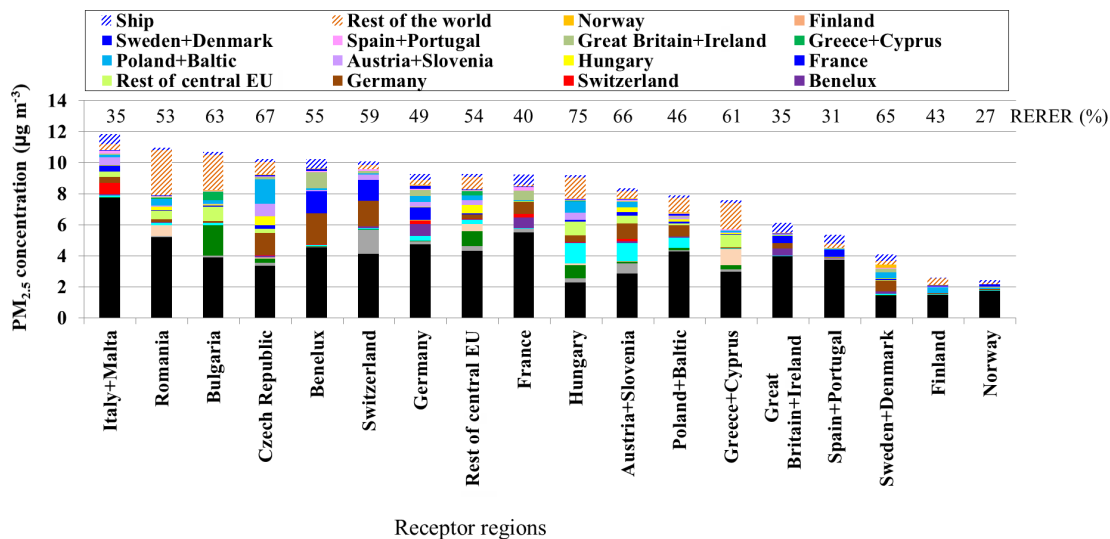
Figure 2. Anthropogenic PM$_{2.5}$ concentrations in 18 countries and sub-regions in Europe separated in within-region and extra-regional contributions. The RERER metric (%) is reported on top of each bar.

ranean (30 %). PM$_{2.5}$ concentrations in emerging economies in Asia, Latin America and the Middle East are dominated by the residential sector, power generation and industry. Asian countries such as China, India, Indonesia and the Philippines are mainly influenced by within-region pollution, with the largest contributions coming from power, industry and residential sectors. PM$_{2.5}$ pollution in Japan is characterised 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. the rest of south Asia and south-eastern Asia) from the power, industry and residential activities. A different picture is seen for Europe where, according to our calculations, annual PM concentrations stem mainly from the agricultural and residential sectors, with a somewhat lower contribution from the transport sector. In eastern European countries noticeable contributions are also found from the power and industrial sectors due to the relatively extensive use of polluting fuels like coal. PM$_{2.5}$ concentrations in USA and Canada are mostly from the power, industry and agricultural sectors. In Oceania industry and agriculture are the most important sectors. PM$_{2.5}$ from ship emissions mainly affects coastal areas of northern Africa, SE Asia (e.g. in Japan,
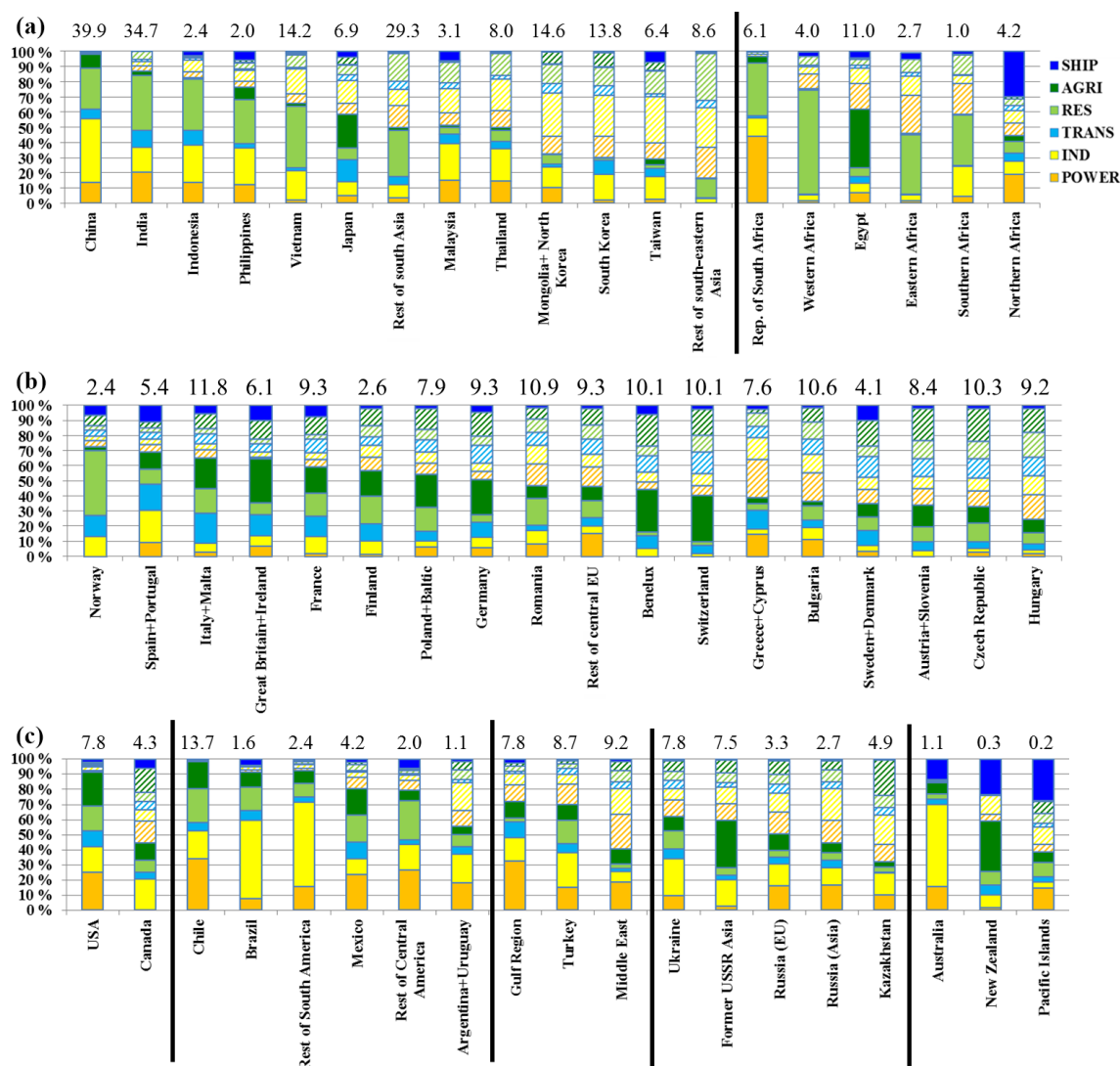
Figure 3. Fraction of within-region and extra-regional (shaded areas) anthropogenic $PM_{2.5}$ concentrations separated by sector for receptor region within the macro-regions: Asia and Africa (a), Europe (b), North America, Latin America, the Middle East, Russia, and Oceania (c). Annual averaged $PM_{2.5}$ anthropogenic concentrations (in $\mu g\,m^{-3}$) are reported on top of each bar. The RERER metric (%) for the 56 TM5-FASST regions is also reported in Table S2.

Taiwan, Malaysia, Indonesia and the Philippines), Mediterranean countries (Spain 11 %, Italy 5 %, France 7 % of their corresponding country totals), northern EU regions (Great Britain 10 %, Norway 6 %, Sweden and Denmark 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 Sect. S2 of the Supplement.

3.3 Gridded $PM_{2.5}$ concentrations

Figure 4 shows the global $1° × 1°$ grid maps of anthropogenic $PM_{2.5}$ concentrations in 2010 for the reference case as well as the computed contributions from each of the major anthropogenic emission sectors. Anthropogenic $PM_{2.5}$ is ubiquitous globally and covers a range from a few $\mu g\,m^{-3}$ or less over the oceans and seas to more than $50\,\mu g\,m^{-3}$ over Asia. As shown in Fig. 3, the most polluted countries in Asia are China, India and the rest of south Asia (which includes Afghanistan, Bangladesh, Bhutan, Nepal and Pakistan), with annual average anthropogenic $PM_{2.5}$ concentrations ranging from 29 to $40\,\mu g\,m^{-3}$; Mongolia and North Korea, Vietnam, South Korea, the rest of south-eastern Asia (including

Table 1. Sector-specific contribution (%) to annual anthropogenic PM$_{2.5}$ concentrations for aggregated world regions based on the "central" estimates which do not consider uncertainty. The largest contributing sectors (above a threshold of 15 %) are shaded in bold font.

	Power	Industry	Transport	Residential	Agriculture	Shipping
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
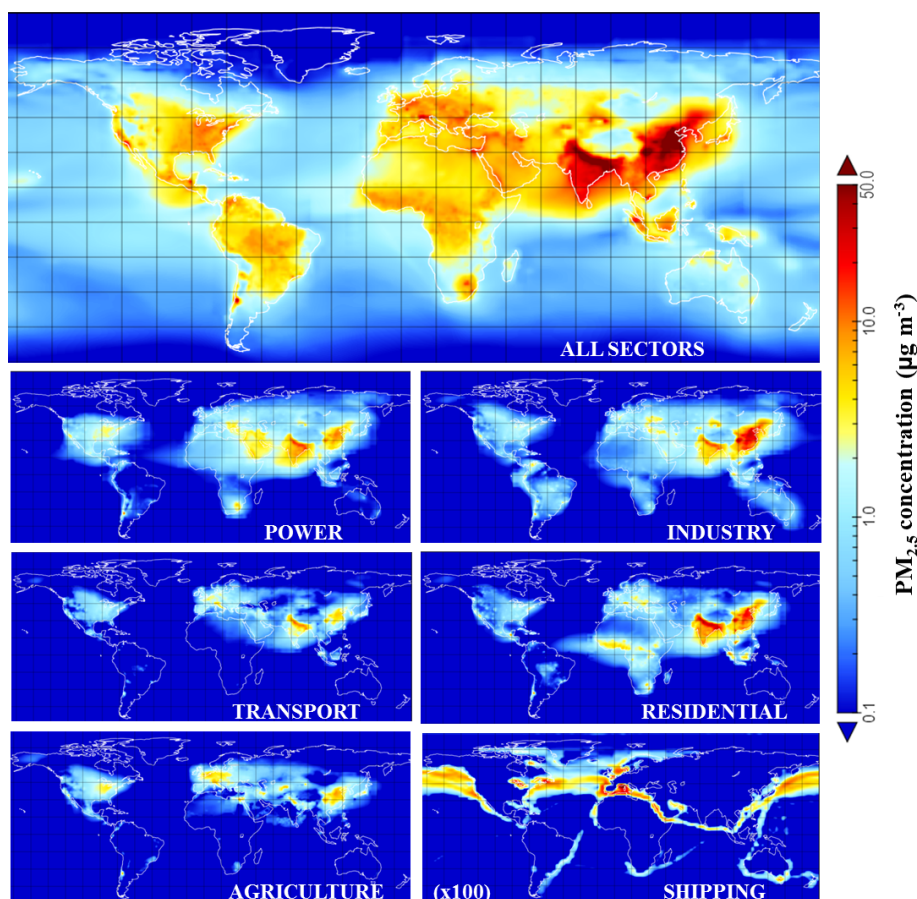
Figure 4. Total anthropogenic PM$_{2.5}$ concentrations (µg m^{-3}) and sectorial contributions using 2010 emissions.

Cambodia, Lao People's Democratic Republic and Myanmar), Thailand, Japan, and Taiwan are rather polluted areas, with PM$_{2.5}$ concentration in the range of 6 to 14 µg m^{-3}. The highest annual PM$_{2.5}$ concentrations in Africa are computed in Egypt (11 µg m^{-3} as annual average), the 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 the 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). The Middle East, the Gulf region, Turkey, Ukraine and the 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. Table 2 reports annual average PM$_{2.5}$ concentrations and the corresponding uncer-

tainty range for each TM5-FASST region as discussed in Sect. 3.4.

The TM5-FASST model (Van Dingenen et al., 2018) is also validated against concentration estimates derived from the WHO database and satellite-based measurements (van Donkelaar et al., 2010, 2014). The TM5-FASST modelled $PM_{2.5}$ concentrations are compared to satellite products which are based on aerosol optical depth measurements together with chemical transport model information to retrieve from the total column the information of PM concentrations in the lowest layer of the atmosphere (Boys et al., 2014; van Donkelaar et al., 2010, 2014). The regional comparison of annul mean population-weighted concentrations shows consistent results with the satellite-based retrievals (e.g. rather good agreement for the globe as a whole, EU and USA within less than 15 % deviation, with lower agreement for developing and emerging countries). Section S4 in the Supplement of the paper by van Dingenen et al. (2018) also reports the comparison between the $PM_{2.5}$ concentrations modelled by TM5-FASST and the measured ones reported in the WHO database, showing rather good agreement for Europe, North America and partly for China due to the higher accuracy of the measurements. The comparison for Latin America and Africa is much less robust and the scatter possibly highlights non-optimal modelling of specific sources relevant for these regions by the TM5-FASST model (e.g. large-scale biomass burning).

In our work, 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, 2015; Boys et al., 2014; Evans et al., 2013; Van Donkelaar et al., 2016), reporting annual averaged $PM_{2.5}$ concentrations for all of Europe in the range between 11 and 17 $\mu g\,m^{-3}$, for Asia from 16 to 58 $\mu g\,m^{-3}$, Latin America 7–12 $\mu g\,m^{-3}$, Africa and the Middle East 8–26 $\mu g\,m^{-3}$, Oceania 6 $\mu g\,m^{-3}$, and North America 13 $\mu g\,m^{-3}$ (note that measurements and satellite estimates would not separate anthropogenic and natural sources of PM, e.g. dust, large-scale biomass burning, while the concentrations in this study consider 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 with emerging economies and fast development (e.g. the 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 an important source of PM all over the world, also affecting indoor air quality (Ezzati, 2008; Lim et al., 2013; Chafe et al., 2014). PM concentrations in Africa and Asia are strongly influenced by this sector due to the incomplete combustion of rather dirty fuels and solid biomass deployed for domestic heating and cooking purposes. Interestingly, the agricultural sector is strongly affecting pollution in

Asia as well as in Europe (Backes et al., 2016; Erisman et al., 2004) and North America, confirming the findings of the UNECE Scientific Assessment Report and several other scientific publications (Maas and Grennfelt, 2016; Pozzer et al., 2017; Tsimpidi et al., 2007; Zhang et al., 2008). The residential and agriculture sectors are less spatially confined, and it is more difficult to regulate emissions effectively than point source emissions of the industrial and power sectors (e.g. in Europe the Large Combustion Plant Directive, the National Emission Ceilings Directive, the Industrial Emissions Directive, the European emission standards for road transport). Finally, shipping is mainly contributing to the pollution in countries and regions with substantial coastal areas, and with ship tracks on the Mediterranean Sea and the Atlantic, Pacific and Indian Oceans, as depicted in Fig. 4.

3.4 Uncertainty from emissions

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

Table 2, as well as Fig. 5, report the annual average anthropogenic $PM_{2.5}$ concentrations ($\mu g\,m^{-3}$) estimated by TM5-FASST with the uncertainty bars representing the upper and lower range of concentrations due to emission inventory 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 (Table 2). Primary PM emissions represent the dominant source of uncertainties, contributing from 45 % to 97 % to the total uncertainty in anthropogenic $PM_{2.5}$ concentrations for each country/region.

Figure 5 depicts the results of the propagation of the lowest and highest range of emissions including their uncertainty to $PM_{2.5}$ concentrations in Asia (Fig. 5a) and – in more detail – Europe (Fig. 5b), highlighting the contribution of within-region and extra-regional $PM_{2.5}$ concentrations and the corresponding 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. Consequently reducing the uncertainties in the Chinese and Indian emission inventories will help in improving the understanding of the long-range contribution of $PM_{2.5}$ pollution in most 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 pollu-

Table 2. Annual average PM$_{2.5}$ concentrations (μg m^{-3}) with upper and lower ranges in brackets due to emission inventory uncertainty (1 standard deviation, σ). The upper and lower range of PM$_{2.5}$ concentrations are calculated as the reference concentrations multiplied and divided by $(1 + \sigma)$ respectively. The third column reflects the fractional uncertainty due to the contribution of primary PM$_{2.5}$ emissions.

World region	TM5-FASST region	PM$_{2.5}$ concentration (μg 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 %
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 %
North America	Canada	4.3 (2.4–8.3)	66 %
	USA	7.8 (4.4–14.4)	71 %

Table 2. Continued.

| World region	TM5-FASST region	PM$_{2.5}$ concentration (µg m^{-3})	Fraction of uncertainty due to primary PM emissions
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 %

tion, produced from the residential, agricultural and transport sectors. Conversely, uncertainties in Norway are dominated by 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-country and imported uncertainties. Figure 5c 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 Fig. 5d displays emission uncertainties for Africa, the 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 %) from neighbouring countries. The uncertainty in within-region emissions, especially from the residential sector, dominates the overall levels of PM$_{2.5}$ uncertainties in Latin America. However, in addition, Chile's own 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. 5d). Interestingly, anthropogenic PM$_{2.5}$ in northern Africa is influenced by uncertainties in Italian emissions as well as those from shipping emissions. Conversely, the Middle East and Turkey regions are influenced by a range of extra-regional emission uncertainties (e.g. the Middle East is affected by the uncertainty of Turkey, Egypt and the Gulf region, while Turkey is affected by Bulgaria, the Gulf region and the rest of the 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 PM$_{2.5}$ concentration precursors and world regions. These contributions can be interpreted as a 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 contributions is provided in Fig. S2, where the share of each term of the sum of Eq. (5) $\left(\sigma_{\text{EMIi,c,p}} \cdot \frac{\text{EMI}_{i,c,p}}{\text{EMI}_{\text{tot},c,p}}\right)^2$ represents the sector contribution to the uncertainty of each pollutant in each region. SO$_2$ uncertainties mainly derive from the power generation sector, especially in countries with dominant coal use; however, substantial contributions are also computed for the industrial sector in South Africa, Asia, Norway, some Latin American countries, Canada and Russia. Interestingly, for SO$_2$ some contributions are also observed from the residential sector in Africa and from the transport sector in some Asian countries (e.g. North and South Korea, Vietnam, Indonesia, southeastern Asia). Smith et al. (2011) report a range of regional uncertainty for SO$_2$ emissions up to 30 %, while our estimates are slightly higher (up to 50 %). NO$_x$ emission uncertainty mainly stems from the transport sector, although some contributions are also seen from power generation in Russia, countries strongly relying on gas (e.g. Russia), the Middle East and the residential sector in Africa. Depending on the region, CO uncertainty (not shown) is dominated by either the transport or residential (particularly in Africa and Asia) sectors and for some regions by a similar contribution of these two sectors. NMVOC emission uncertainties mainly derive from poorly characterised industrial, transport and residential activities 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. Among all air pollutants, primary PM$_{2.5}$ represents one of the most uncertain pollutant due to very different combustion conditions, different fuel qualities and lack of control measures (Klimont et al., 2017).

Primary particulate matter emissions should be mainly improved for the residential, transport and in particular industrial sectors. Black carbon emission inventories should be better characterised in Europe, Japan, North and South 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 define EFs for the differ-
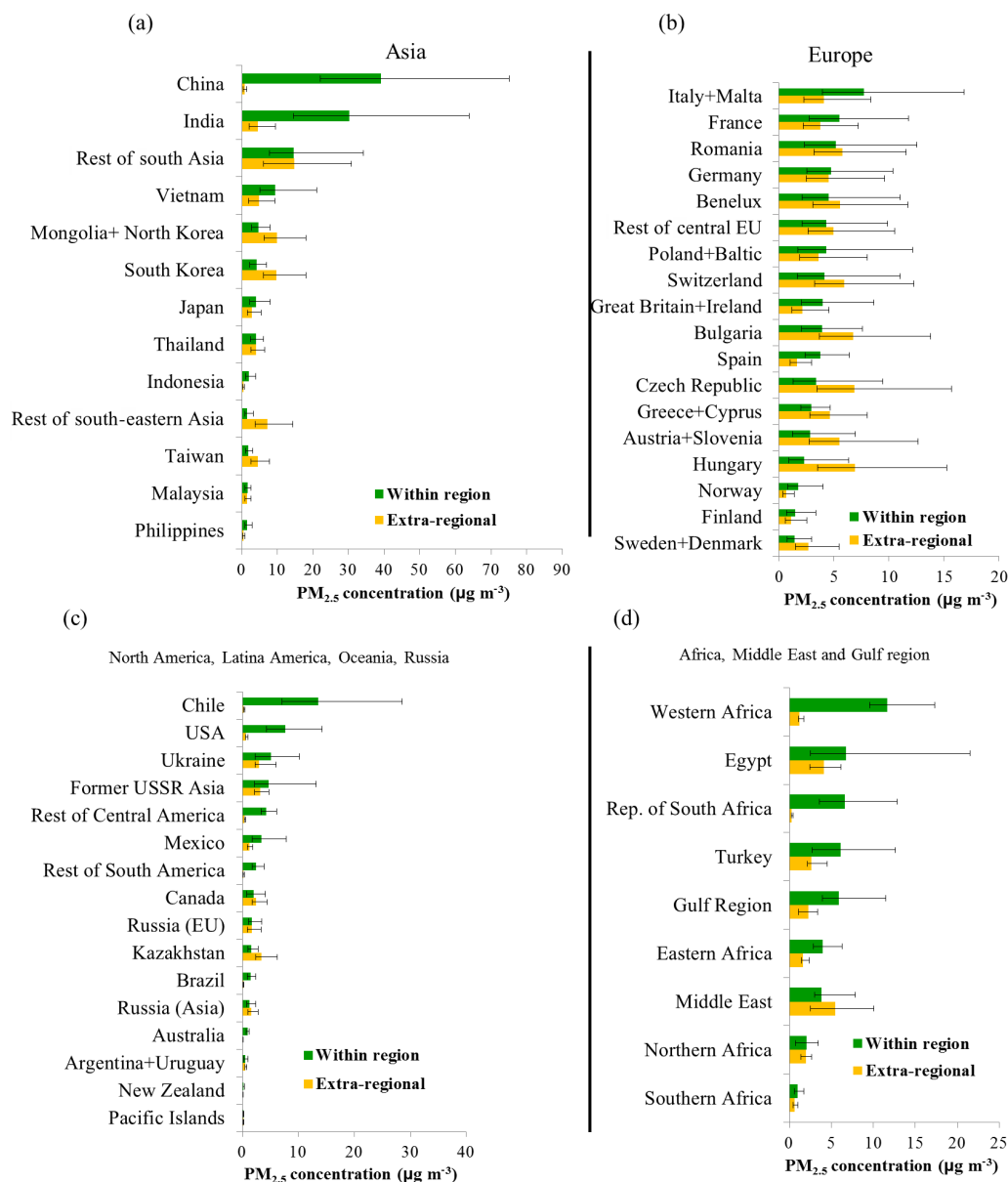
Figure 5. Within-region and extra-regional anthropogenic PM$_{2.5}$ concentrations and emission-related uncertainties for Asia **(a)**; Europe **(b)**; North America, Latin America, Oceania and Russia **(c)**; and Africa, the Gulf region and the Middle East **(d)**. The error bars are calculated by multiplying and dividing the reference emissions by $(1 + \sigma)$ as discussed in Sect. 2.3.

ent type of fuels used under different combustion conditions. To constrain and improve particulate organic matter emissions, efforts should be made to improve residential emissions estimates. 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 widely used in the residential sector, but national reporting often underestimates the emissions from this sector, 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).

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

An additional uncertainty is related to the lack of information in the inventory regarding the 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 residential sector (σ_{AD,RES_bio}) by comparing it to the recent TNO RWC (Netherlands Organization for Applied Scientific Research, 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), 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, Greenhouse Gas and Air Pollution Interactions and Synergies, and IEA, International Environmental Agency) 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. (4), 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 emis-

sion factors uncertainty of each individual pollutant (σ_{EFp}) from Eq. (4). 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 data, we can evaluate the mean normalised absolute error (MNAE) considering all N countries, following Eq. (6) (Yu et al., 2006), which represents our estimate of σ_{AD,RES_bio}.

$$\text{MNAE} = \frac{1}{N} \cdot \sum_j^N \frac{\left|\text{TNO RWC}_j - \text{EDGARv4.3.2}_j\right|}{\text{TNO RWC}_j} \quad (6)$$

We estimate a value of σ_{AD,RES_bio} of 38.9 %, which is much larger compared to the 5 %–10 % uncertainty reported 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. (4) and the calculated σ_{AD,RES_bio} and σ_{EFp}, we can evaluate a new σ_{EMIp,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-2 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 uncertainty in wood burning activity data. Large increases in reported biomass usage for domestic use

Table 3. PM$_{2.5}$ concentrations due to the residential sector emissions in Europe, European part of Russia, Ukraine and Turkey and the 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

has been noted in IEA energy statistics for some European countries (IEA, 2016) and further increases are expected as countries are shifting their methodologies to estimate biofuel activity data away from fuel sales statistics to a modelling approach based on energy demand. In addition, several EU countries are increasing the use of biomass in order 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, last access: April 2019). 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 on PM$_{2.5}$ concentrations in the residential sector. 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$_{2.5}$ concentrations represent the most robust and widely used metric to analyse the long-term impacts of particulate matter air pollution on human mortality (Pope and Dockery, 2006; Dockery, 2009). As described in Sects. 2.5 and S5 of the paper by Van Dingenen et al. (2018), 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 causes of death, namely ischaemic heart disease (IHD), cerebrovascular disease (CD, stroke), chronic obstructive pulmonary disease (COPD), lung cancer (LC) and acute lower respiratory infections (ALRIs). The relative risk (RR) represents the proportional increase in the assessed health outcome due to a given increase in PM$_{2.5}$ concentrations (Burnett et al., 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 recent scientific publications (Silva et al., 2016). Figure 7 represents the 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 per year. In our work we only evaluate how the uncertainty of emission inventories influences the health impact estimates focusing on the interregional aspects (i.e. we do not evaluate effects of misallocation of sources within regions) and not all the other sources of uncertainties, such as the uncertainty of concentration-response estimates, of air quality models used to estimate particulate matter concentrations, etc. An overview of the propagation of the uncertainty associated with an ensemble of air quality models to health and crop impacts is provided by Solazzo et al. (2018). Solazzo et al. (2018) find in their analysis over the European countries a mean number of PDs due to exposure to PM$_{2.5}$ and ozone of approximately 370 000 (inter-quantile range between 260 000 and 415 000). Moreover, they estimate that a reduction in the uncertainty of the modelled ozone concentration by 61 %–80 % (depending on the aggregation metric used) and by 46 % for PM$_{2.5}$ produces a reduction in the uncertainty in premature mortality and crop loss of more than 60 %. However, we show here that the often neglected emission inventories' uncertainty provides a range of premature deaths of ±1.1 million at the global scale, which is of the same order of magnitude of the uncertainty of air quality models and concentration-response functions (Cohen et al., 2017). In 2010, using our central estimate, 82 % of the PDs occur in fast-growing economies and developing countries, especially in China with 670 000 and India with an almost equal amount of 610 000 PDs per year. Table 4 summarises our estimates of premature mortality for aggregated world re-

Table 4. Absolute and population size normalised number of premature deaths per year due to anthropogenic $PM_{2.5}$ air pollution in world regions and corresponding uncertainty range.

	PD (deaths per year)
China+	6.7×10^5 (3.5×10^5–1.0×10^6)
India+	6.1×10^5 (2.7×10^5–9.6×10^5)
Europe	2.6×10^5 (1.4×10^5–4.8×10^5)
SE Asia	1.5×10^5 (8.3×10^4–2.5×10^5)
Russia	1.1×10^5 (6.7×10^4–2.4×10^5)
North America	1.0×10^5 (5.5×10^4–1.7×10^5)
Africa	7.4×10^4 (3.4×10^4–1.6×10^5)
Middle East	5.6×10^4 (3.2×10^4–9.7×10^4)
Latin America	2.6×10^4 (1.4×10^4–5.3×10^4)
Oceania	5.5×10^1 (3.4×10^1–1.2×10^2)

gions, with Europe accounting for 210 000 PDs per year and North America 100 000 PDs per year.

Our results are comparable with Lelieveld et al. (2015) and Silva et al. (2016) who, using the same Burnett et al. (2014) methodology, estimate 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 higher value of global mortality (3.9 million PDs per year) mainly due to a lower minimum risk exposure level set in the exposure response function, the inclusion of the urban increment calculation and the contribution of natural sources. 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 (including information on urban and rural population; refer to Van Dingenen et al., 2018), the inclusion or not of natural components, the impact threshold value used, and RR functions. In this study, we use the population-weighted $PM_{2.5}$ concentration (excluding natural components) at $1 \times 1°$ resolution as the metric for estimating health effects due to air pollution, with a threshold value of $5.8\,\mu g\,m^{-3}$, no urban increment adjustment and relative risk functions accordingly with Burnett et al. (2014). We also estimate that 7 % of the global non-accidental mortalities from the Global Burden of Disease (IHME, 2015, http://vizhub.healthdata.org/gbd-compare, last access: April 2019; Forouzanfar et al., 2015) are attributable to air pollution in 2010; 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.

Table 5 shows the number of premature deaths for each receptor region, highlighting the premature mortality induced within the country itself and outside the receptor 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 contribute an additional 700 000 and ca. 500 000 PDs per year, respectively, representing more than 50 % of the global mortality. Clearly, reducing emissions and emission uncertainties in these two regions will therefore have the largest overall benefit on global air quality improvement as well as on global human health. As explained in Sect. 3.1, PDs attributed to internal and external emissions are directly linked (proportional) to the internal and external $PM_{2.5}$ contributions. For most of the TM5-FASST regions, PDs due to anthropogenic emissions within the source region are higher than the extra-regional contributions. However, there are marked exceptions, such as Hungary, the Czech Republic, Mongolia, etc., where the extra-regional and within-region contributions to mortality are at least comparable. For instance, Hungary and the Czech Republic are strongly influenced by polluted regions in Poland (mainly); likewise, Mongolia is affected by the vicinity of sources in China. The Gulf region produces a lot of its own pollution but is also influenced by transport from Africa and Eurasia as reported by Lelieveld et al. (2009).

Detailed information on the premature deaths for each TM5-FASST region and the contributing anthropogenic emission sectors is shown in Fig. 8a and b. Health effects induced by air quality in industrialised countries are mainly related to agriculture (32.4 % of total mortality or 68 000 PDs per year), residential combustion (17.8 % or 37 000 PDs per year) and road transport (18.7 % or 39 000 PDs per year) for Europe and with power generation (26.4 % or 26 000 PDs per year), industry (19 % or 19 000 PDs per year), residential (17 % or 17 000 PDs per year) and agriculture (24.0 % or 24 000 PDs per year) for North America. The health impacts observed in most western EU countries is due to both within-region and extra-regional pollution, while in several eastern EU countries the impact of neighbouring countries is even larger compared to within-region pollution. The premature deaths induced by international shipping emissions represent 5.5 % of total EU PD, which is in the range of the results of Brandt et al. (2013) (ca. 50 000 PDs). PM-related mortality in developing countries and fast-growing economies is mostly affected by industrial (up to 42 % in China or 279 000 PDs per 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 113 000 PDs per year). Chinese emissions have a strong impact on China, Japan, Vietnam, Mongolia + North Korea, and 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 the $PM_{2.5}$ concentrations and health stan-

Table 5. Number of premature deaths for each receptor region including the within-region and extra-regional attribution based on $PM_{2.5}$ "central" estimates, which do not consider uncertainty. For the RERER metric refer also to Table S2.

World regions	TM5-FASST region name	PDs in receptor region (deaths per year)	Within-region PDs (deaths per year)	Extra-regional PDs (deaths per year)
Africa	Eastern Africa	16 705	8218	8487
Africa	Egypt	17 282	11 380	5902
Africa	Northern Africa	5424	3427	1997
Africa	Rep. of South Africa	9065	8797	268
Africa	Southern Africa	345	322	23
Africa	Western Africa	25 081	19 785	5296
Asia	China	655 870	643 129	12 741
Asia	Indonesia	17 780	14 803	2977
Asia	India	474 660	412 298	62 362
Asia	Japan	25 636	15 181	10 455
Asia	South Korea	25 295	7510	17 784
Asia	Mongolia + North Korea	12 657	4076	8581
Asia	Malaysia	2014	1058	957
Asia	Philippines	121	94	27
Asia	Rest of south Asia	134 280	67 170	67 110
Asia	Rest of south-eastern Asia	23 316	3814	19 502
Asia	Thailand	21 231	10 495	10 736
Asia	Taiwan	3443	1028	2415
Asia	Vietnam	30 750	20 286	10 464
Europe	Austria + Slovenia	6073	1806	4267
Europe	Bulgaria	4739	1709	3030
Europe	Benelux	9090	4201	4889
Europe	Switzerland	3200	1568	1632
Europe	Czech Republic	7936	2696	5240
Europe	Germany	36 256	18 595	17 661
Europe	Spain + Portugal	11 291	8487	2804
Europe	Finland	0	0	0
Europe	France	22 046	13 320	8727
Europe	Great Britain + Ireland	13 949	9459	4490
Europe	Greece + Cyprus	3117	1133	1984
Europe	Hungary	14 211	3820	10391
Europe	Italy + Malta	24 417	16 312	8105
Europe	Norway	674	516	158
Europe	Poland + Baltic	28 686	15 877	12 809
Europe	Rest of central EU	6764	3418	3346
Europe	Romania	14 155	6979	7176
Europe	Sweden + Denmark	2650	1021	1629
Latin America	Argentina + Uruguay	133	75	58
Latin America	Brazil	4261	3968	293
Latin America	Chile	3332	3283	49
Latin America	Mexico	10 478	8447	2031
Latin America	Rest of Central America	3413	2772	640
Latin America	Rest of South America	4489	4164	325
Middle East	Gulf region	15 176	11 225	3951
Middle East	Middle East	6784	2804	3980
Middle East	Turkey	34 151	24 191	9960
North America	Canada	3262	1491	1771
North America	USA	97 877	90 176	7701
Oceania	Australia	28	25	3
Oceania	New Zealand	24	15	9
Oceania	Pacific Islands	3	1	2
Russia	Kazakhstan	3389	1100	2290
Russia	Former USSR Asia	10 757	6420	4337
Russia	Russia (Asia)	1348	601	746
Russia	Russia (EU)	25 149	12 704	12 445
Russia	Ukraine	71 724	44 604	27 120
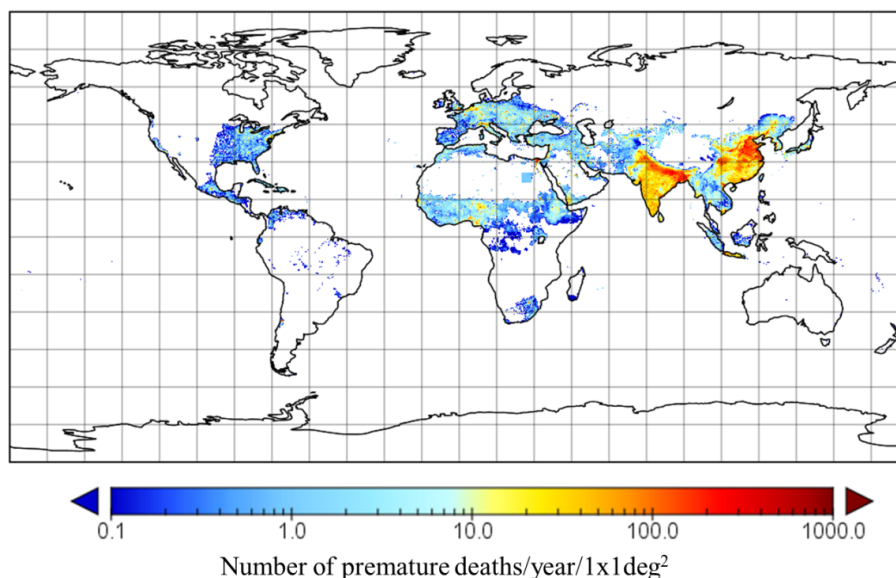
Figure 7. Global distribution of premature deaths in 2010 caused by anthropogenic particulate matter pollution estimated using the methodology described in Burnett et al. (2014). A threshold value of $5.8\,\mu g\,m^{-3}$ is assumed and no urban increment adjustments are considered. The relative risk functions of Burnett et al. (2014) are used for the premature death dose-response estimates.

dards of North and South Korea and other south-eastern Asian countries, as well as the need for international measures to improve air quality.

5 Conclusions

In this work we couple the global anthropogenic emission estimates provided by the HTAP_v2.2 inventory for 2010 (merging national and regional inventories) with 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 $40\,\mu g\,m^{-3}$, with the highest annual concentrations computed in China ($40\,\mu g\,m^{-3}$, range: $22.4–76.6\,\mu g\,m^{-3}$), India ($35\,\mu g\,m^{-3}$, range: $16.6–73.4\,\mu g\,m^{-3}$), North America ($8\,\mu g\,m^{-3}$, range: $4.4–14.4\,\mu g\,m^{-3}$) and Europe (on average ca. $8\,\mu g\,m^{-3}$, range: $5–18\,\mu g\,m^{-3}$). 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). Moreover, due to the transport of PM between European countries, EU-wide directives can help improve 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 the most $PM_{2.5}$ 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 the HTAP_v2.2 emission inventory and TM5-FASST, we also evaluate how the uncertainty in sectors and regions propagates into $PM_{2.5}$ concentrations. The aim of our analysis is to provide insights into where improvement of country emission inventories would give the largest benefits, 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 more accurate quantification of the contribution of the long-range sources to $PM_{2.5}$ pollution in most Asian countries. Here we demonstrate how analysis of uncertainties in national and 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 characterise this source.

Finally, we analyse the air quality effects on health. Global health effects due to $PM_{2.5}$ concentrations calculated with
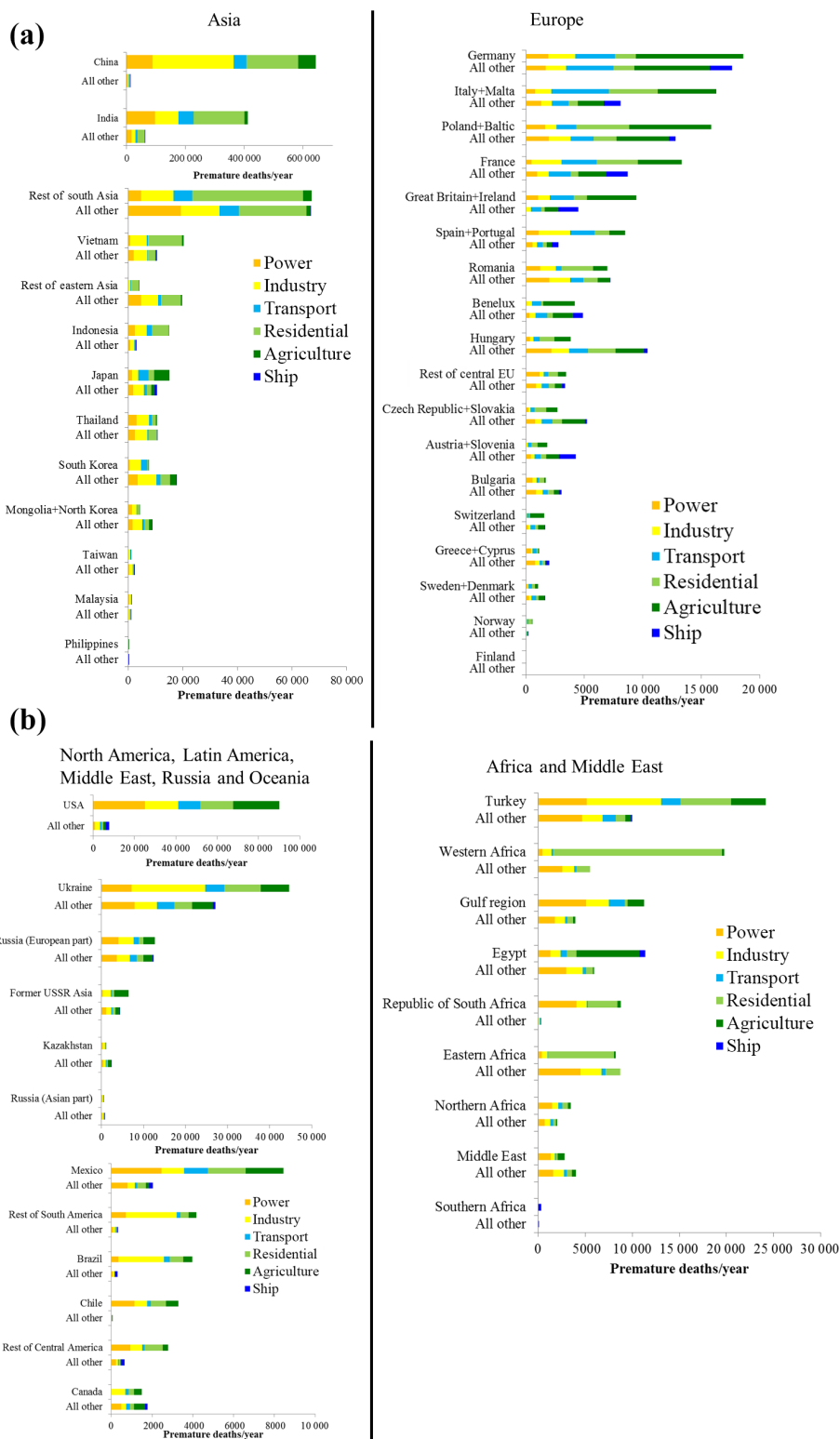
Figure 8. Anthropogenic emission sector contributions to premature mortality (deaths per year) due to PM$_{2.5}$ population-weighted concentrations in the TM5-FASST receptor regions of **(a)** Asia (left) and Europe (right) and **(b)** North America, Latin America, Russia, the Middle East and Oceania (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. Sector and region contributions pertain to the "central" emission estimates.

TM5-FASST and anthropogenic emissions in 2010 are estimated to be ca. 2.1 million premature deaths per year, but the uncertainty associated with emission ranges between 1 and 3.4 million deaths per year, with the largest fraction of PD (82 %) in developing countries.

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