

1 **Sectorial and regional uncertainty analysis of the contribution of anthropogenic emissions**  
2 **to regional and global PM<sub>2.5</sub> health impacts**

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4 Monica Crippa<sup>1</sup>, Greet Janssens-Maenhout<sup>1</sup>, Diego Guizzardi<sup>2</sup>, Rita Van Dingenen<sup>1</sup>, Frank  
5 Dentener<sup>1</sup>

6 <sup>1</sup>European Commission, Joint Research Centre (JRC), Via E. Fermi 2749, I-21027 Ispra (VA),  
7 Italy

8 <sup>2</sup>Didesk Informatica, Verbania (VB), Italy

9

10 **Abstract**

11 In this work we couple the HTAP\_v2.2 global air pollutant emission inventory with the global  
12 source receptor model TM5-FASST to evaluate the relative contributions of the major  
13 anthropogenic emission sources (power generation, industry, ground transport, residential,  
14 agriculture and international shipping) to air quality and human health in 2010. We focus on  
15 particulate matter (PM) concentrations because of the relative importance of PM<sub>2.5</sub> emissions in  
16 populated areas and the well-documented cumulative negative effects on human health. We  
17 estimate that in 2010, depending on the region, annual averaged anthropogenic PM<sub>2.5</sub>  
18 concentrations varied between ca 1 and 40 µg/m<sup>3</sup>, with the highest concentrations observed in  
19 China and India, and lower concentrations in Europe and North America. The relative  
20 contribution of anthropogenic emission sources to PM<sub>2.5</sub> concentrations varies between the  
21 regions. European PM pollution is mainly influenced by the agricultural and residential sectors,  
22 while the major contributing sectors to PM pollution in Asia and the emerging economies are the  
23 power generation, industrial and residential sectors. We also evaluate the emission sectors and  
24 emission regions in which pollution reduction measures would lead to the largest improvement  
25 on the overall air quality. We show that air quality improvements would require regional  
26 policies, in addition to local and urban scale measures, due to the transboundary features of PM  
27 pollution. We investigate emission inventory uncertainties and their propagation to PM<sub>2.5</sub>  
28 concentrations, in order to identify the most effective strategies to be implemented at sector and  
29 regional level to improve emission inventories knowledge and air quality. We show that the  
30 uncertainty of PM concentrations depends not only on the uncertainty of local emission  
31 inventories, but also on that of the surrounding regions. Finally, we propagate emission  
32 inventories uncertainty to PM concentrations and health impacts.

33

34 **1 Introduction**

35 Ambient particulate matter pollution ranks among the top five risk factors globally for loss of  
36 healthy life years and is the largest environmental risk factor (Lim et al., 2013;Anderson et al.,  
37 2012;Anenberg et al., 2012;Cohen et al., 2017). The world health organization (WHO, 2016)  
38 reported about 3 million premature deaths worldwide attributable to ambient air pollution in  
39 2012. Health impacts of air pollution can be attributed to different anthropogenic emission  
40 sectors (power generation, industry, residential, transport, agriculture, etc.) and sector-specific

1 policies could effectively reduce health impacts of air pollution. These policies are usually  
2 implemented under national legislation (Henneman et al., 2017; Morgan, 2012), while in Europe  
3 transboundary air pollution is also addressed by the regional protocol under the UNECE  
4 Convention on Long-Range Transport of Air Pollution (CLRTAP). At city/local level, several  
5 studies have been developed to assess the contribution of sector specific emissions to PM<sub>2.5</sub>  
6 concentrations with the aim of designing air quality plans at local and regional level (Karagulian  
7 et al., 2015; Thunis et al., 2016). Indeed, particulate matter can travel thousands of kilometers,  
8 crossing national borders, oceans and even continents (HTAP, part A, 2010). Local, regional and  
9 international coordination is therefore needed to define air pollution policies to improve globally  
10 air quality and possibly human health. The CLRTAP's Task Force on Hemispheric Transport of  
11 Air Pollution looks at the long-range transport of air pollutants in the Northern Hemisphere  
12 aiming to identify promising mitigation measures to reduce background pollution levels and its  
13 contribution to pollution in rural as well as urban regions. Although primary PM<sub>2.5</sub> (particulate  
14 matter with a diameter less than 2.5 µm) and intermediately lived (days-to-weeks) precursor  
15 gases can travel over long distances, the transboundary components of anthropogenic PM are  
16 mainly associated with secondary aerosols which are formed in the atmosphere through complex  
17 chemical reactions and gas-to-aerosol transformation, transport and removal processes, of  
18 gaseous precursors transported out of source regions (Maas and Grennfelt, 2016). However, the  
19 most extreme episodes of exposure often occur under extended periods of low wind speeds and  
20 atmospheric stability, favoring formation of secondary aerosols close to the source regions.  
21 Secondary aerosol from anthropogenic sources consists of both inorganic -mainly ammonium  
22 nitrate and ammonium sulfate and ammonium bisulfate and associated water, formed from  
23 emissions of sulphur dioxide (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>) and ammonia (NH<sub>3</sub>), and organic  
24 compounds involving thousands of compounds and often poorly known reactions (Hallquist et  
25 al., 2009). Exposure to and impact from aerosols on humans can be estimated by a variety of  
26 approaches, ranging from epidemiological studies to pure modelling approaches. The Burnett et  
27 al. (2014) risk-response methodology is often used in models to estimate premature  
28 deaths/mortality (PD) due to air pollution exposure, e.g. in Lelieveld et al. (2015) and Silva et al.  
29 (2016), who report a global mortality in 2010 due to air quality issues induced by anthropogenic  
30 emissions of 2.5 and 2.2 million people, respectively. A higher global mortality is found in a  
31 more recent work by Cohen et al. (2017) accounting for 3.9 million premature deaths/year due to  
32 different model assumptions. In Europe, Brant et al. (2013) estimate 680 thousand premature  
33 deaths, which is twice as high as the numbers reported for the CAFE (Clean Air for Europe)  
34 study (Watkiss et al., 2005). Recently, using the same emission database as in this study, Im et  
35 al. (2017) report a multi-model mean estimate of PD of 414.000 (range 230-570 thousand) for  
36 Europe and 160 thousand PDs for the USA. At the global scale, models, in some cases using  
37 satellite information (Brauer et al., 2015; Van Donkelaar et al., 2016), are the most practical  
38 source of information of exposure to air pollution. However, model calculations are subject to a  
39 range of uncertainties related with incomplete understanding of transport, chemical  
40 transformation, removal processes, and not the least, emission information.

41 This work is developed in the context of the TF HTAP Phase 2 (Galmarini et al., 2017a), where a  
42 number of models are deployed to assess long-range sensitivities to extra-regional emissions,  
43 using the same HTAP\_v2.2 anthropogenic emission inventory (Janssens-Maenhout et al., 2015).  
44 Differences in model results illustrate uncertainties in model formulations of transport, chemistry  
45 and removal processes, and are addressed in separate studies (Liang et al., 2018), but not of  
46 uncertainties in emission inventories. The objective of this study is to evaluate the relevance of

1 uncertainties in regional sectorial emission inventories (power generation, industry, ground  
2 transport, residential, agriculture and international shipping), and their propagation in modelled  
3 PM<sub>2.5</sub> concentrations and associated impacts on health. To this end we couple the HTAP\_v2.2  
4 global emission inventory for the year 2010 and the global source-receptor model TM5-FASST  
5 (TM5-FAst Scenario Screening Tool) to estimate global air quality in terms of PM<sub>2.5</sub>  
6 concentrations. The regional and global scale, the focus on annual PM<sub>2.5</sub> and associated health  
7 metrics, warrants the use of the TM5-FASST model. However, the most extreme episodes of  
8 pollution may occur at more local-to-regional scales justifying the need for local. For instance, a  
9 recent study performed over hundreds of cities in Europe (Thunis et al., 2017) shows that in  
10 order to comply with the standards prescribed by the Air Quality Directives and the health  
11 guidelines by WHO, local actions at the city scale are needed.

12 Specifically, we show that the impact of emission inventory uncertainty on mortality estimates is  
13 comparable with the range of uncertainty induced by air quality models and population exposure  
14 functions. We also investigate the uncertainties in PM<sub>2.5</sub> from within the region to extra-regional  
15 contributions. A second objective of this analysis is to evaluate the importance of emission  
16 uncertainties at sector and regional level on PM<sub>2.5</sub>, to better inform local, regional and  
17 hemispheric air quality policy makers on the potential impacts of sectors with larger  
18 uncertainties (e.g. residential and agriculture) or regions (e.g. developing and emerging  
19 countries).

20

## 21 **2 Methodology**

### 22 **2.1 TM5-FASST model and emission perturbations**

23 The TM5-FASST model is extensively documented in a companion publication in this special  
24 issue. Van Dingenen et al., (2018) provide an extensive evaluation of the model, model  
25 assumptions and performance with regard to linearity and additivity of concentration response to  
26 different size of emission perturbations and future emission scenarios. The validation of TM5-  
27 FASST against the full TM5 model runs is extensively discussed by van Dingenen et al. (2018),  
28 as well as the validity of the assumptions of linearity and additivity behind this reduced form-  
29 model. Below we summarize the most important features of relevance for this work, and refer for  
30 more detail to Van Dingenen et al., (2018).

31 In order to calculate PM<sub>2.5</sub> concentrations corresponding to the HTAP\_v2.2 emissions, we use  
32 the native 1°x1° resolution source-receptor gridmaps obtained for TM5-FASST\_v0 (Van  
33 Dingenen et al., 2018). The TM5-FASST source-receptor model is based on a set of emission  
34 perturbation experiments (-20 %) of SO<sub>2</sub>, NO<sub>x</sub>, CO, NH<sub>3</sub>, and VOC and CH<sub>4</sub> using the global  
35 1°x1° resolution TM5 model, the meteorological year 2001 (which was also used for the HTAP  
36 Phase 1 experiments) and the community emission dataset prepared for the IPCC AR5 report  
37 (RCP, Representative Concentration Pathway) emissions for the year 2000 (Lamarque et al.,  
38 2010). TM5-FASST uses aggregated regional emissions (i.e. one annual emission value per  
39 pollutant or precursor for each of the 56 regions + shipping), with an implicit underlying 1°x1°  
40 resolution emission spatial distribution from RCP year 2000 which was partly based EDGAR  
41 methodology and gridmaps. The concentration of PM<sub>2.5</sub> contributing from and to each of 56  
42 receptor regions is estimated as a linear function of the emissions of the source regions, including  
43 the aerosol components BC, primary organic matter (POM), SO<sub>4</sub>, NO<sub>3</sub>, and NH<sub>4</sub>. While

1 Secondary Organic Aerosol (SOA) from natural sources is included in the model calculations  
 2 using the parameterisation described in Dentener et al. (2006), no explicit treatment of  
 3 anthropogenic SOA is considered, since no reliable emission inventories of SOA precursor gases  
 4 was available, and formation processes were not included in the parent TM5 model. A recent  
 5 study by Farina et al. (2010) indicates a global source of 1.6 Tg, or ca 5.5 % of the overall SOA  
 6 formation due to anthropogenic SOA. The relative importance of anthropogenic SOA ranges  
 7 regionally widely, and is deemed higher in regions with less VOC emission controls. We  
 8 speculate that the inclusion of SOA would possibly lead to a somewhat larger role of the  
 9 transboundary pollution transport, mainly for those sectors emitting PM and VOCs (e.g.  
 10 residential, and to some extent transport and industry).

11  
 12 Under the assumption that the individual sector contributions add up linearly to total  $PM_{2.5}$  – this  
 13 assumption is further evaluated in Van Dingenen et al. (2018) - the comparison of  $PM_{2.5}$   
 14 concentrations calculated for the reference and scenario case yields an estimation of the  
 15 contribution of each sector to total  $PM_{2.5}$  concentrations.

16  
 17 Specifically, the reduced-form model TM5-FASST is computing the concentration resulting  
 18 from an arbitrary emission scenario  $E_i$  using a “perturbation approach”, i.e. the difference  
 19 between  $E_i$  and  $E_{i,ref}$  ( $dE$ ) is considered as a perturbation on  $E_{ref}$  and the resulting concentration is  
 20 evaluated as a perturbation  $dPM$  on the reference concentration:

$$21 \quad 22 \quad dPM(\mathbf{y}) = \sum_i SRC_i[x, y] \cdot [E_i(x) - E_{i,ref}(x)] \quad (\text{Eq. 1})$$

$$23 \quad SRC_i[x, y] = \frac{\Delta PM_{ref}(y)}{\Delta E_{i,ref}(x)} \quad (\text{Eq. 2})$$

24 where the summation is made over all primary emitted components and precursors ( $i$ ) for  
 25 secondary components, and  $SRC_i[x, y]$  is a set of Source-Receptor Coefficients describing the  
 26 linearized relationship between each precursor emission of specific components and  
 27 concentration for each pair of source ( $x$ ) and receptor ( $y$ ) region. Van Dingenen et al. (2018)  
 28 explain in detail how the ‘perturbation approach’ can be also applied also for evaluating the  
 29 attribution by sector as well as by source region. Thus to calculate total  $PM_{2.5}$  concentration in  
 30 each receptor region, the 56 source region individual contributions must be summed. Using this  
 31 approach, it is possible to evaluate the  $PM_{2.5}$  concentrations from “within-region” and “extra-  
 32 regional”  $PM_{2.5}$  emissions. The extra-regional contribution represents the RERER metric  
 33 (Response to Extra-Regional Emission Reduction) for a specific region used across the whole  
 34 HTAP experiment (Galmarini et al., 2017b), in particular focusing on the  $PM_{2.5}$  concentration  
 35 reduction due to the contribution of the emissions of each anthropogenic sector (Eq. 3):

$$36 \quad RERER = \frac{\sum R(\text{foreign regions})}{\sum R(\text{all regions})} \quad (\text{Eq. 3})$$

37

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

39

1 As depicted in Fig. S1, the 56 TM5-FASST regions cover the entire globe, but their areal extent  
2 differs in terms of size, population, emission magnitude and presence of neighbouring countries  
3 (e.g. Europe comprises 18 TM5-FASST regions). In order to make smaller regions (e.g.  
4 European countries) comparable with larger regions (like USA, China and India), in this work an  
5 aggregation procedure to 10 world regions (refer to Table S2) has been applied (China+, India+,  
6 SE Asia, North America, Europe, Oceania, Latin America, Africa, Russia and Middle East). In  
7 this work we focus on particulate matter due to its negative effects on human health (WHO,  
8 2013;Pope and Dockery, 2006), (Worldbank, 2016). The TM5-FASST model includes an  
9 assessment of the premature mortality due to ambient PM<sub>2.5</sub> concentrations on exposed  
10 population following the methodology developed by Burnett et al. (2014), as discussed in Sect.  
11 4. Health impacts due to indoor air pollution or ozone are not evaluated in this work.

12 The aim of this work is to address the uncertainty of sector specific emissions from this  
13 inventory in a quantitative way as well as the differences we observe from one region to the  
14 other, based on the uncertainty of activity data and emission factors. As discussed in the next  
15 section, the reason to use HTAP\_v2.2 and not e.g. the RCP2000 as the basis for our assessment  
16 of emission propagation is that the TF HTAP aims at bringing policy relevant information, and to  
17 this end, it has compiled a policy relevant emission inventory (HTAP\_v2.2) for the most recently  
18 available year. While the RCP2000 was at the basis of the FASST calculations, and presented the  
19 best community emissions effort at the time, the HTAP\_v2.2 inventory is now day much more  
20 accurate in particular given the focus on regional (and not so much gridded) emission analysis of  
21 our work.

## 22 **2.2 HTAP\_v2.2 emissions**

23 The global anthropogenic emission inventory HTAP\_v2.2 for the year 2010 is input to the global  
24 source-receptor model TM5-FASST to evaluate PM<sub>2.5</sub> concentrations for each world  
25 region/country with the corresponding health effects. The HTAP\_v2.2 inventory includes for  
26 most countries official and semi-official annual anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub>, CO  
27 (carbon monoxide), NMVOC (non-methane volatile organic compounds), PM<sub>10</sub> (particulate  
28 matter with a diameter less than 10 µm) PM<sub>2.5</sub>, BC (black carbon) and OC (organic carbon) by  
29 country and sector (Janssens-Maenhout et al., 2015). Here we focus on the 6 major  
30 anthropogenic emission sectors contributing to global PM<sub>2.5</sub> concentrations, namely the power  
31 generation (“power”), non-power industry, industrial processes and product use (“industry”),  
32 ground transportation (“transport”), residential combustion and waste disposal (“residential”),  
33 agriculture (“agriculture”) and international shipping (“ship”). International and domestic  
34 aviation emissions are not considered in this study due to the lower contribution to air pollution  
35 compared to other anthropogenic sectors. It should be noted that agricultural emissions do not  
36 include agricultural waste burning and forest and savannah fires. Details on the emissions  
37 included in each aggregated sector can be found in Janssens-Maenhout et al. (2015). In addition  
38 to the reference HTAP\_v2.2 emissions for the year 2010, a set of emission perturbation scenarios  
39 has been created by subtracting from the reference dataset the emissions of each sector.

## 40 **2.3 Emission inventory uncertainties**

41 In order to investigate how computed PM<sub>2.5</sub> concentrations are affected by the uncertainty of  
42 emission inventories, we perform a sensitivity analysis testing the upper and lower range of  
43 HTAP\_v2.2 emissions including their uncertainties. Aggregated emissions of a certain pollutant

1  $p$ , from a sector  $i$  and country  $c$  are calculated as the product of activity data (AD) and emission  
 2 factors (EF), therefore the corresponding uncertainty ( $\sigma_{i,c,p}$ ) is calculated as following:

$$3 \quad \sigma_{EMI\ i,c,p} = \sqrt{\sigma_{ADi,c}^2 + \sigma_{EF,i,p,c}^2} \quad (\text{Eq.4})$$

4 where  $\sigma_{AD}$  and  $\sigma_{EF}$  are the uncertainties (%) of the activity data and emission factors for a certain  
 5 sector, country and pollutant. Uncertainty values of the activity data by sector and country are  
 6 obtained from Table 2 of Janssens-Maenhout et al. (2017, in review) and Olivier et al. (2016).  
 7 Using this approach, the uncertainty in the global total anthropogenic CO<sub>2</sub> emissions is estimated  
 8 to range from -9% to +9% (95% confidence interval), which is the result from larger  
 9 uncertainties of about  $\pm 15\%$  for non-Annex I countries, whereas uncertainties of less than  $\pm 5\%$   
 10 are obtained for the 24OECD90 countries for the time series from 1990 (Olivier et al, 2016)  
 11 reported to UNFCCC. Uncertainty values for the emission factors of gaseous pollutants are  
 12 retrieved from the EMEP/EEA Guidebook (2013) and Bond et al. (2004) for particulate matter.  
 13 In this work we assume that reported countries emissions are based on independent estimations  
 14 of activity data and emission factors EFs, hence no cross-country correlation structure is  
 15 assumed. This is in contrast to bottom-up gridded emission inventories like EDGAR, where the  
 16 use of global activity datasets may lead to correlated errors between countries.

17 Therefore, we can calculate the overall uncertainty  $\sigma_{EMI\ p,c}$  for a certain pollutant ( $p$ ) due to all  
 18 sectors ( $i$ ) in a specific country ( $c$ ) with the following equation (EMEP/EEA, 2013).

$$19 \quad \sigma_{EMI\ p,c} = \sqrt{\sum_i \left( \sigma_{EMI\ i,c,p} * \frac{EMI_{i,c,p}}{EMI_{tot,c,p}} \right)^2} \quad (\text{Eq. 5})$$

21  
 22 where  $EMI_{i,c,p}$  (in kton) represents the emission of a certain pollutant in a certain country from a  
 23 specific sector ( $i$ ) and  $EMI_{tot,c,p}$  (in kton) the corresponding emissions from all sectors for that  
 24 country and pollutant.

25 Table S3, reports the overall uncertainty calculated for each pollutant and for each TM5-FASST  
 26 region. Using an additional constraint that EFs and activities cannot be negative, a lognormal  
 27 distribution of the calculated uncertainties is assumed (Bond et al., 2004). Therefore we can  
 28 calculate the upper and lower range of emission estimates multiplying and dividing the reference  
 29 emissions by  $(1+\sigma_{p,c})$ , respectively. We do not account for the uncertainties of the atmospheric  
 30 transport model and the uncertainties due to aggregation, which are larger over smaller TM5-  
 31 FASST regions. Based on the upper and lower emission range per region, new TM5-FASST  
 32 model runs have been performed per source region to retrieve the corresponding range of  
 33 concentrations in receptor regions (therefore the total number of computations is  $56*2$  for the  
 34 uncertainty analysis).

35

36

### 1 3 TM5-FASST modelling results

#### 2 3.1 Regional contributions to PM<sub>2.5</sub> concentrations

3 Figure 1 provides a global perspective on the fraction of within-region and extra-regional PM<sub>2.5</sub>  
4 concentrations for 10 aggregated world receptor regions using emissions of the year 2010, with  
5 the extra-regional fraction (RERER metric) broken down into source region contributions.  
6 Annual average population weighted anthropogenic PM<sub>2.5</sub> concentrations (refer to Van Dingenen  
7 et al., (2018) for the calculation of this metric) ranged from few  $\mu\text{g}/\text{m}^3$  (e.g. in Oceania or Latin  
8 America), around 7-8  $\mu\text{g}/\text{m}^3$  for North America and Europe, and up to 33-39  $\mu\text{g}/\text{m}^3$  in China+  
9 (including also Mongolia) and India+ (including also the rest of South Asia). Anthropogenic  
10 PM<sub>2.5</sub> pollution in China+ and India+ is mainly affected by large emission sources within the  
11 country (98 and 96%, respectively), although 4 % of the Indian anthropogenic PM<sub>2.5</sub> pollution is  
12 mainly transported from the Gulf region and Middle East. North America (98%) and Oceania  
13 (98%) are mainly influenced by within-regional pollution due to their geographical isolation  
14 from other regions. TM5-FASST computations attributed 11 % of the PM<sub>2.5</sub> in Europe to extra-  
15 regional sources; for the Middle East and Gulf region extra-regional contributions amount to  
16 18% (mainly from Europe and Russia), for Africa 25% (mainly from Europe and Middle East),  
17 and Russia 28% (mainly from Europe, Middle East and Gulf region and China). Shipping  
18 emissions are not considered in this figure due to their international origin, while inland  
19 waterways emissions are still included in the ground transport sector. Transboundary air  
20 pollution is known to be an important issue in the rest of Asia, in particular for pollution  
21 transported from China to Korea and Japan (Park et al., 2014) and we estimate that the  
22 contribution of transported PM is up to 40% in South Eastern Asia (mainly from China and  
23 India). Within-region and extra-regional PM<sub>2.5</sub> concentrations for all the TM5-FASST regions  
24 are reported in Table S2.

25 Focusing on Europe, Fig. 2 shows within-region (in black) vs. extra-regional absolute  
26 population-weighted PM<sub>2.5</sub> concentrations (in  $\mu\text{g}/\text{m}^3$ ) for 16 EU countries plus Norway and  
27 Switzerland, defined in TM5-FASST, as well as the source regions contributing to this pollution.  
28 Regional annual averages of population weighted PM<sub>2.5</sub> concentrations in Europe vary between  
29 2-4  $\mu\text{g}/\text{m}^3$  in Northern European countries (like Finland, Norway and Sweden) up to 10-12  
30  $\mu\text{g}/\text{m}^3$  for continental Europe. Although most of EU annual average PM<sub>2.5</sub> concentrations are  
31 below the World Health Organization Air Quality Guideline of 10  $\mu\text{g}/\text{m}^3$  PM<sub>2.5</sub> (as annual  
32 average), these values represent only regional averages while several exceedances in urban areas  
33 are often observed in Europe. As further discussed in Sect. 3.2, an additional contribution to  
34 PM<sub>2.5</sub> concentrations comes from the shipping sector, mainly influencing Mediterranean  
35 countries (like Italy, Spain and France) and countries facing the North Sea, Baltic Sea and  
36 Atlantic Ocean (e.g. Benelux, Sweden, Great Britain, etc.). Transboundary air pollution from  
37 external regions contributes by 27% to 75 % and on average by 51 % to PM<sub>2.5</sub> pollution in  
38 European countries. Countries surrounded by oceans, are mainly influenced by within-region  
39 pollution due to their geographical isolation from other source regions (e.g. Italy, Spain, Great  
40 Britain and Norway); therefore the fraction of extra-regional pollution ranges from 27% to 35%.  
41 The largest extra-regional contributions are calculated for Hungary (75%, mainly from Austria,  
42 Czech Republic, Rest of Central EU, Poland and Germany), Czech Republic (67%, mainly from  
43 Poland, Germany and Austria), Austria and Slovenia (66%, mainly from Czech Republic,  
44 Germany and Italy), Sweden+Denmark (65%, mainly from Germany, Norway and Poland),

1 Bulgaria (63%, mainly from Romania), and Greece (61%). The remaining EU countries are both  
2 affected by within-region and extra-regional pollution (the latter ranging from 40% to 59%),  
3 highlighting the importance of transboundary transport of PM<sub>2.5</sub> concentrations. For example  
4 Switzerland is influenced by the pollution coming from France, Italy and Germany; Rest of  
5 Central EU by Poland and Germany; Germany by France and Benelux; Poland by Czech  
6 Republic and Germany. Interestingly, Romania, Bulgaria, Greece and Hungary are also  
7 significantly affected by the pollution transported from Ukraine and Turkey, which is included in  
8 the “rest of the world” contribution of Fig. 2. Our results are consistent with the findings of the  
9 latest UNECE Scientific Assessment Report (Maas and Grennfelt, 2016), which highlights the  
10 importance of transboundary transport of organic and inorganic PM.

### 11 **3.2 Sectorial contributions to PM<sub>2.5</sub> concentrations**

12 Figure 3 shows the relative sectorial contributions to anthropogenic PM<sub>2.5</sub> concentrations for the  
13 56 TM5-FASST receptor regions, separating the fraction of extra-regional (RERER) (shaded  
14 colors) and within-region pollution, while Table 1 shows regional average values of sector-  
15 specific relative contributions. In most African regions (except Egypt) anthropogenic PM<sub>2.5</sub>  
16 concentrations are mainly produced by emissions in the residential sector. Agriculture is an  
17 important sector for Egypt, while Northern Africa is strongly influenced by shipping emissions  
18 in the Mediterranean (30%). PM<sub>2.5</sub> in emerging economies in Asia, Latin America and Middle  
19 East are dominated by PM<sub>2.5</sub> concentrations from the residential sector, power generation and  
20 industrial. Asian countries, China, India, Indonesia and Philippines are mainly influenced by  
21 within-region pollution with the largest contributions coming from power, industry and  
22 residential sectors. Japan is characterised by the contribution of local sources like transport and  
23 agriculture but it is also affected by transported pollution from China, especially from the  
24 industrial sector. Anthropogenic PM<sub>2.5</sub> in the remaining Asian countries is influenced by more  
25 than 50% by the pollution coming from China (e.g. Vietnam, Malaysia, Thailand, Mongolia,  
26 South Korea, Taiwan) or India (e.g. Rest of South Asia and South Eastern Asia) from the power,  
27 industry and residential activities. A different picture is seen for Europe where according to our  
28 calculations, annual PM concentrations stem mainly from the agricultural and residential sectors  
29 with a somewhat lower contribution from the transport sector. In Eastern European countries  
30 noticeable contributions are also found from the power and industrial sectors due to the relatively  
31 extensive use of polluting fuels like coal. PM<sub>2.5</sub> concentrations in USA and Canada are mostly  
32 from the power, industry and agricultural sectors. In Oceania industry and agriculture are the  
33 most important sectors. PM<sub>2.5</sub> from ship emissions mainly affect coastal areas of North Africa,  
34 SE Asia (e.g. in Japan, Taiwan, Malaysia, Indonesia and Philippines), Mediterranean countries  
35 (Spain 11%, Italy 5%, France 7% of their corresponding country totals), Northern EU regions  
36 (Great Britain 10%, Norway 6%, Sweden and Denmark 10% of their corresponding country  
37 totals) and Oceania (22% of the regional total). Over the international areas of sea and air no  
38 distinction between within-region and extra-regional concentrations is reported. Further details  
39 on within-region and extra-regional concentrations can be found in section S2 of the  
40 Supplementary Material.

### 41 **3.3 Gridded PM<sub>2.5</sub> concentrations**

42 Figure 4 shows the global 1°x1° gridmaps of anthropogenic PM<sub>2.5</sub> concentrations in 2010 for the  
43 reference case as well as the contribution from each of the major anthropogenic emission sectors.  
44 Anthropogenic PM<sub>2.5</sub> is ubiquitous globally and covers a range from a μg/m<sup>3</sup> or less over the

1 oceans and seas to more than  $50 \mu\text{g}/\text{m}^3$  over Asia. As shown in Fig. 3, the most polluted  
2 countries in Asia are China, India and Rest of South Asia (which includes Afghanistan,  
3 Bangladesh, Bhutan, Nepal and Pakistan) with annual average anthropogenic  $\text{PM}_{2.5}$   
4 concentrations ranging from  $29$  to  $40 \mu\text{g}/\text{m}^3$ ; Mongolia and North Korea, Vietnam, South Korea,  
5 Rest of South Eastern Asia (including Cambodia, Lao People's Democratic Republic and  
6 Myanmar), Thailand, Japan and Taiwan are rather polluted areas with  $\text{PM}_{2.5}$  concentration in the  
7 range of  $6$  to  $14 \mu\text{g}/\text{m}^3$ . The highest annual  $\text{PM}_{2.5}$  concentrations in Africa are computed in Egypt  
8 ( $11 \mu\text{g}/\text{m}^3$  as annual average), Republic of South Africa ( $6.1 \mu\text{g}/\text{m}^3$  as annual average) and  
9 Western Africa ( $4.0 \mu\text{g}/\text{m}^3$  as annual average). The highest pollution in Europe is observed in the  
10 Benelux region, Italy and in some of the Eastern countries (e.g. Romania, Bulgaria and Czech  
11 Republic), while in Latin America the most polluted areas are Chile ( $13.7 \mu\text{g}/\text{m}^3$  as annual  
12 average) and Mexico ( $4.2 \mu\text{g}/\text{m}^3$  as annual average). Middle East, the Gulf region, Turkey,  
13 Ukraine and former USSR are also characterised by  $\text{PM}_{2.5}$  concentrations ranging between  $7.5$   
14  $\mu\text{g}/\text{m}^3$  and  $9.2 \mu\text{g}/\text{m}^3$  as annual averages.

15 The TM5-FASST model developed by van Dingenen et al. (2018) has been validated against  
16 concentration estimates derived from the WHO database (WHO, 2011, 2014, 2016) and satellite-  
17 based measurements (van Donkelaar et al., 2010, 2014). General good agreement is found  
18 between the  $\text{PM}_{2.5}$  concentrations modeled by TM5-FASST and the measured ones reported in  
19 the WHO database for Europe (within 20% deviation), North America (within 5% deviation) and  
20 partly China due to the higher accuracy of the measurements. The comparison for Latin America  
21 and Africa is much less robust (40-60% deviation from the 1:1 line) and the scatter possibly  
22 highlights a non-optimal modeling of specific sources relevant for these regions by TM5-FASST  
23 (e.g. large scale biomass burning) by the TM5-FASST model. Similar results are also found  
24 comparing regional averages of urban stations from WHO against the FASST population  
25 weighted average of grid cells. The TM5-FASST modeled  $\text{PM}_{2.5}$  concentrations have been  
26 compared to satellite products which are based on aerosol optical depth measurements together  
27 with chemical transport model information to retrieve from the total column the information of  
28 PM concentrations in the lowest layer of the atmosphere (Boys et al., 2014; van Donkelaar et al.,  
29 2010, 2014). The regional comparison shows consistent results with the ground based  
30 measurements comparison (e.g. good agreement for EU and USA within 10% deviation, while  
31 lower agreement for developing and emerging countries).

32 In our work, modelled  $\text{PM}_{2.5}$  concentrations are in the range of the measurements and satellite-  
33 based estimates provided in several literature studies (Brauer et al., 2012; Brauer et al.,  
34 2015; Boys et al., 2014; Evans et al., 2013; Van Donkelaar et al., 2016), reporting for the whole  
35 Europe annual averaged  $\text{PM}_{2.5}$  concentrations in the range between  $11$  and  $17 \mu\text{g}/\text{m}^3$ , for Asia  
36 from  $16$  to  $58 \mu\text{g}/\text{m}^3$ , Latin America  $7$ - $12 \mu\text{g}/\text{m}^3$ , Africa and Middle East  $8$ - $26 \mu\text{g}/\text{m}^3$ , Oceania  $6$   
37  $\mu\text{g}/\text{m}^3$  and North America  $13 \mu\text{g}/\text{m}^3$  (note that measurements and satellite estimates would not  
38 separate anthropogenic and natural sources of PM, e.g. dust, large scale biomass burning, while  
39 the concentrations in this study consider anthropogenic emissions alone).

40 In order to understand the origin of global  $\text{PM}_{2.5}$  concentrations, we look at sector specific maps  
41 (Fig. 4). The power and industrial sectors are mainly contributing to PM concentrations in  
42 countries having emerging economies and fast development (e.g. Middle East, China and India),  
43 while the ground transport sector is a more important source of PM concentrations in  
44 industrialised countries (e.g. North America and Europe) and in developing Asian countries. The  
45 residential sector is an important source of PM all over the world, also affecting indoor air

1 quality (Ezzati, 2008;Lim et al., 2013;Chafe et al., 2014). Africa and Asia are strongly influenced  
2 by PM concentrations produced by this sector due to the incomplete combustion of rather dirty  
3 fuels and solid biomass deployed for domestic heating and cooking purposes. Interestingly, the  
4 agricultural sector is affecting pollution in Asia as well as in Europe (Backes et al., 2016;  
5 Erisman et al., 2004) and North America, confirming the findings of the UNECE Scientific  
6 Assessment Report and several other scientific publications (Maas and Grennfelt, 2016;Pozzer et  
7 al., 2017;Tsimpidi et al., 2007;Zhang et al., 2008). The residential and agriculture sectors are less  
8 spatially confined, and emissions more difficult to be effectively regulated than point source  
9 emissions of the industrial and power sectors (e.g. in Europe the Large Combustion Plant  
10 Directive, the National Emission Ceilings or the Industrial Emissions, the Euro norms for road  
11 transport, etc.). Finally, shipping is mainly contributing to the pollution in countries and regions  
12 with substantial coastal areas, and with ship tracks on the Mediterranean Sea, the Atlantic,  
13 Pacific and Indian Oceans, as depicted in Fig. 4.

### 14 **3.4 Uncertainty from emissions**

#### 15 **3.4.1 Propagation of emission uncertainties to anthropogenic PM<sub>2.5</sub> concentrations**

16 Table 2, as well as Fig. 5, report the annual average anthropogenic PM<sub>2.5</sub> concentrations (µg/m<sup>3</sup>)  
17 estimated by TM5-FASST with the uncertainty bars representing the upper and lower range of  
18 concentrations due to emission inventories uncertainty. The extra-regional contribution to  
19 uncertainty is also addressed as well as the contribution of the uncertainty of primary particulate  
20 matter emissions to the upper range of PM<sub>2.5</sub> concentrations (Table 2). We estimate that primary  
21 PM emissions represent the dominant source of uncertainties, contributing from 45% to 97% to  
22 the total uncertainty in anthropogenic PM<sub>2.5</sub> concentrations for each country/region.

23 Figure 5 depicts the results of the propagation of the lowest and highest range of emissions  
24 including their uncertainty to PM<sub>2.5</sub> concentrations in Asia (Fig 5a) and - in more detail- Europe  
25 (Fig 5b), highlighting the contribution of within-region and extra-regional PM<sub>2.5</sub> concentrations  
26 and uncertainties (error bars). Due to their large sizes, Indian and Chinese PM<sub>2.5</sub> concentrations  
27 and uncertainties are mainly affected by uncertainties from the residential, transport and  
28 agricultural sectors within these countries. Interestingly, in South Eastern and Eastern Asia  
29 uncertainties in PM<sub>2.5</sub> are strongly influenced by the Indian residential emissions. On the other  
30 hand, PM<sub>2.5</sub> in Thailand, Japan, Taiwan, South Korea, Mongolia and Vietnam are strongly  
31 affected by the uncertainty in the Chinese residential and industrial emissions. Consequently  
32 reducing the uncertainties in the Chinese and Indian emission inventories will help improving the  
33 understanding the long-range contribution of PM<sub>2.5</sub> pollution in most of Asian countries.

34 In Europe, the highest uncertainties in PM<sub>2.5</sub> concentrations are associated with the emissions  
35 from the residential, agriculture and transport sectors. In most of the Central and Eastern  
36 European countries modelled PM<sub>2.5</sub> is strongly affected by the uncertainty of transported extra-  
37 regional pollution, produced from the residential, agricultural and transport sectors. Conversely,  
38 uncertainties in Norway are dominated by national emissions, mainly from the residential and  
39 transport sectors, and in Italy from the residential and agriculture sectors. The remaining  
40 European countries are affected both by within-country and imported uncertainties. Fig. 5c  
41 represents the results of the propagation of the emissions range including their uncertainty to  
42 PM<sub>2.5</sub> concentrations for North America, Latin America, Oceania and Russia, while Fig 5d  
43 displays emission uncertainties for Africa, Middle East and the Gulf region. The uncertainty in

1 the USA agricultural and residential emissions affect more than 50% of modelled Canadian  
2 PM<sub>2.5</sub> concentrations and the uncertainty in Mexico and Argentina is influenced by similar  
3 magnitudes (30-50%) by neighbouring countries. The uncertainty in within-region emissions,  
4 especially from the residential sector, dominates the overall levels of PM<sub>2.5</sub> uncertainties in Latin  
5 America. However, in addition, Chili's own agriculture and power sectors contribute  
6 significantly to the overall uncertainty levels. PM<sub>2.5</sub> levels in most of the African regions are  
7 strongly affected by the uncertainty in their own residential emissions, while in Egypt they are  
8 mostly influenced by the agricultural sector uncertainties (refer to Fig. 5d). Interestingly,  
9 anthropogenic PM<sub>2.5</sub> in Northern Africa is influenced by uncertainties in Italian emissions  
10 uncertainty as well as those from shipping emissions. Conversely, the Middle East and Turkey  
11 regions are influenced by a range of extra-regional emission uncertainties (e.g. Middle East is  
12 affected by the uncertainty of Turkey, Egypt and the Gulf region, while Turkey by Bulgaria, Gulf  
13 region and rest of Central EU).

14

### 15 **3.4.2 Ranking the sector specific contribution to emission uncertainties**

16 Figure 6 shows the average sector relative contribution to total emission inventory related  
17 uncertainty for the main PM<sub>2.5</sub> concentration precursors and world regions. These contributions  
18 can be interpreted as a ranking of the most effective improvements to be taken regionally to  
19 better constrain their inventories and reduce the final formation of PM<sub>2.5</sub> concentrations. The  
20 complete overview of all TM5-FASST regions is provided in Fig. S2, where the share of each

21 term of the sum of Eq. 5  $\left(\sigma_{EMI_{i,c,p}} * \frac{EMI_{i,c,p}}{EMI_{tot,c,p}}\right)^2$ , representing the sector contribution to the  
22 uncertainty of each pollutant in each region, is reported. SO<sub>2</sub> uncertainties mainly derive from  
23 the power generation sector especially countries with a dominant coal use; however, substantial  
24 contributions are also computed for the industrial sector in South Africa, Asia, Norway, some  
25 Latin American countries, Canada and Russian countries. Interestingly, for SO<sub>2</sub> some  
26 contributions are also observed from the residential sector in Africa and from the transport sector  
27 in some Asian countries (e.g. Korea, Vietnam, Indonesia, South Eastern Asia, etc.). Smith et al.  
28 (2011) report a range of regional uncertainty for SO<sub>2</sub> emissions up to 30%, while our estimates  
29 are slightly higher (up to 50%). NO<sub>x</sub> emissions uncertainty mainly stems from the transport  
30 sector, although some contributions are also seen from power generation in Russia, countries  
31 strongly relying on gas (e.g. Russia), the Middle East and the residential sector in Africa.  
32 Depending on the region, CO uncertainty (not shown) is dominated by either the transport or  
33 residential (particularly in Africa and Asia) sectors and for some regions by a similar  
34 contribution of these two sectors. NMVOC emission uncertainties mainly derive from poorly  
35 characterized industrial, transport and residential activities due to the complex mixture and  
36 reactivity of such pollutants. As expected, NH<sub>3</sub> emission uncertainty is dominated by the  
37 agricultural sector which appears to be less relevant for all other pollutants. Among all air  
38 pollutants, primary PM<sub>2.5</sub> represents one of the most uncertain pollutant due to very different  
39 combustion conditions, different fuel qualities and lack of control measures (Klimont et al.,  
40 2017).

41 Primary particulate matter emissions should be mainly improved for the residential, transport and  
42 in particular industrial sectors. Black carbon emission inventories should be better characterised  
43 in Europe, Japan, Korea, Malaysia etc. for the transport sector, where the higher share of diesel

1 used as fuel for vehicles leads to higher BC emissions; in addition, BC emissions from the  
 2 residential sector require further effort to better define EFs for the different type of fuels used  
 3 under different combustion conditions. To constrain and improve particulate organic matter  
 4 emissions, efforts should be dedicated to improve residential emissions estimates. Therefore, in  
 5 the following section, we try to assess one of the major sources of uncertainty in the residential  
 6 emissions in Europe which is the use of solid biofuel.

### 7 **3.4.3 Assessing the uncertainty in household biofuel consumption with an independent** 8 **inventory in Europe**

9 The combustion of solid biomass (i.e. biofuel) for household heating and cooking purposes is  
 10 one of the major sources of particulate matter emissions in the world. Wood products and  
 11 residues are widely used in the residential sector, but national reporting often underestimates the  
 12 emissions from this sector, due to the fact that often informal economic wood sales are not  
 13 accurately reflected in the official statistics of wood consumption (AD) (Denier Van Der Gon et  
 14 al., 2015). An additional uncertainty is related to the lack of information in the inventory  
 15 regarding the emission factors (EF) variability, which depends on the combustion efficiency and  
 16 type of wood (Weimer et al., 2008;Chen et al., 2012). In our work we estimate the uncertainty  
 17 attributable to wood combustion in the residential sector ( $\sigma_{AD,RES,bio}$ ) by comparing it to the  
 18 recent TNO RWC (residential wood combustion) inventory of Denier van der Gon et al. (2015),  
 19 which includes a revised biomass fuel consumption with the corresponding EDGARv4.3.2  
 20 activity data (Janssens-Maenhout et al., 2017), as shown in Table S4. In the TNO RWC  
 21 inventory, wood use for each country has been updated comparing the officially reported per  
 22 capita wood consumption data (from GAINS and IEA) with the expected specific wood use for a  
 23 country including the wood availability information (Visschedijk et al., 2009;Denier Van Der  
 24 Gon et al., 2015). We can therefore assume that the TNO RWC inventory represents an  
 25 independent estimate of wood consumption in the residential sector, allowing a more precise  
 26 uncertainty estimation of the AD for this sector. Assuming that emissions are calculated as the  
 27 product of AD and EF, the corresponding uncertainty can be calculated with Eq. 4, where  $\sigma_{AD}$   
 28 ranges from 5 to 10% for European countries and Russia as reported for international statistics  
 29 (Olivier et al., 2016). We can therefore calculate the residential emission factors uncertainty of  
 30 each individual pollutant ( $\sigma_{EF,p}$ ) from Eq. 4. In addition, based on the comparison of the recent  
 31 estimates of wood consumption provided by TNO RWC AD, which should match better with  
 32 observations and the EDGARv4.3.2 ones, we can evaluate the mean normalized absolute error  
 33 (MNAE) considering all  $N$  countries, following Eq. 6 (Yu et al., 2006), which represents our  
 34 estimate of  $\sigma_{AD,RES,bio}$ .

35

$$36 \quad \text{MNAE} = \frac{1}{N} * \sum_j^N \frac{|TNO\ RWC_j - EDGARv4.3.2_j|}{TNO\ RWC_j} \quad (\text{Eq.6})$$

37

38 We estimate a value of  $\sigma_{AD,RES,bio}$  of 38.9% which is much larger compared to the 5-10%  
 39 uncertainty reported for the fuel consumption of the international statistics ( $\sigma_{AD}$ ). The issue of  
 40 biofuel uncertainty mainly affects rural areas where wood is often used instead of fossil fuel.

41 Then, using Eq. 4 and the calculated  $\sigma_{AD,RES,bio}$  and  $\sigma_{EF,p}$ , we can evaluate a new  $\sigma_{EMI,p,RES,bio}$

1 for the residential sector including the uncertainty of the AD due to the use of wood as fuel for  
2 this sector, as reported in Table S5. Comparing the results shown in Table S5 with the factor of  
3 two uncertainty values expected for PM emissions from the residential sector (Janssens-  
4 Maenhout et al., 2015), we derive that the uncertainty associated with the emission factors for  
5 biomass combustion in the residential sector is the dominant source of uncertainty compared to  
6 the uncertainty in wood burning activity data. Large increases in reported biomass usage for  
7 domestic use has been noted in IEA energy statistics for some European countries (IEA,  
8 2013,2014,2015,2016) and further increases are expected as countries are shifting their  
9 methodologies to estimate biofuel activity data away from fuel sales statistics to a modelling  
10 approach based on energy demand. In addition, several EU countries are increasing the use of  
11 biomass in order to accomplish the targets set in the context of the renewable energy directive  
12 (2009/28/EC) as reported in their national renewable energy action plans  
13 (<http://ec.europa.eu/energy/node/71>). When comparing the UNFCCC and the TNO RWC data, a  
14 higher value of  $\sigma_{AD,RES,bio}$  is obtained (59.5% instead of 38.9%), although its effect on the final  
15 residential emission uncertainty is less strong, as shown in Table S6. Table 3 shows the impact of  
16 biofuel combustion uncertainty in the residential sector on PM<sub>2.5</sub> concentrations. Upper-end  
17 uncertainties indicate that PM<sub>2.5</sub> concentrations could be between 2.6 and 3.7 times larger than  
18 those derived from the HTAP\_v2.2 inventory.

#### 19 **4 Health impact assessment**

20 Annual population weighted PM<sub>2.5</sub> concentrations represent the most robust and widely used  
21 metric to analyse the long-term impacts of particulate matter air pollution on human health (Pope  
22 and Dockery, 2006;Dockery, 2009). The mortality estimation in TM5-FASST is based on the  
23 integrated exposure-response functions defined by Burnett et al. (2014). The increased risk from  
24 exposure to air pollution is estimated using exposure-response functions for five relevant deaths  
25 causes, namely Ischemic heart disease (IHD), Cerebrovascular Disease (CD, stroke), Chronic  
26 Obstructive Pulmonary Disease (COPD), Lung Cancer (LC), Acute Lower Respiratory  
27 Infections (ALRI). The relative risk (RR) represents the proportional increase in the assessed  
28 health outcome due to a given increase in PM<sub>2.5</sub> concentrations (Burnett, 2014).

29 In this section, we investigate the impact of total and sector-specific anthropogenic population  
30 weighted PM<sub>2.5</sub> concentrations on health and we show comparisons with mortality estimates  
31 provided by WHO and scientific publications (Silva et al., 2016). Figure 7 represents the  
32 premature deaths (PD) distribution due to air pollution, using population weighted PM<sub>2.5</sub>  
33 concentrations and representative for anthropogenic emissions in the year 2010. The most  
34 affected areas are China and India, but also some countries of Western Africa and urban areas in  
35 Europe (in particular in the Benelux region and Eastern Europe). Our computations indicate that  
36 annual global outdoor premature mortality due to anthropogenic PM<sub>2.5</sub> amounts to 2.1 million  
37 premature deaths, with an uncertainty range related to emission uncertainty of 1-3.3 million  
38 deaths/year. In our work we only evaluate how the uncertainty of emission inventories influences  
39 the health impact estimates focusing on the interregional aspects (i.e. we do not evaluate effects  
40 of misallocation of sources within regions) and not all the other sources of uncertainties, such as  
41 the uncertainty of concentration-response estimates, of air quality models used to estimate  
42 particulate matter concentrations, etc. An overview of the propagation of the uncertainty  
43 associated with an ensemble of air quality models to health and crop impacts is provided by  
44 Solazzo et al. (2018, submitted). Solazzo et al. find in their analysis over the European countries

1 a mean number of PDs due to exposure to PM<sub>2.5</sub> and ozone of approximately 370 thousands  
2 (inter-quantile range between 260 and 415 thousand). Moreover, they estimate that a reduction in  
3 the uncertainty of the modelled ozone by 61% - 80% (depending on the aggregation metric used)  
4 and by 46% for PM<sub>2.5</sub>, produces a reduction in the uncertainty in premature mortality and crop  
5 loss of more than 60%. However, we show here that the often neglected emission inventories'  
6 uncertainty provides a range of premature deaths of ±1.1 million at the global scale, which is in  
7 the same order of magnitude of the uncertainty of air quality models and concentration-response  
8 functions (Cohen et al., 2017). In 2010, using our central estimate, 82% of the PDs occur in fast  
9 growing economies and developing countries, especially in China with 670 thousand and India  
10 with an almost equal amount of 610 thousand PD/year. Table 4 summarizes our estimates of  
11 premature mortality for aggregated world regions, with Europe accounting for 210 thousand  
12 PD/year and North America 100 thousand PD/year.

13 Our results are comparable with Lelieveld et al. (2015) and Silva et al. (2016) who, using the  
14 same Burnett et al. (2014) methodology, estimate a global premature mortality of 2.5 and 2.2  
15 million people, respectively, due to air quality in 2010 for the same anthropogenic sectors.  
16 However, a recent work published by Cohen et al. (2017) estimates a higher value of global  
17 mortality (3.9 million PD/year) mainly due to a lower minimum risk exposure level set in the  
18 exposure response function, the inclusion of the urban increment calculation and the contribution  
19 of natural sources. When comparing mortality estimates we need to take into account that  
20 several elements affect the results, like the resolution of the model, the urban increment subgrid  
21 adjustment (including information on urban and rural population, refer to van Dingenen et al.  
22 2018), the inclusion or not of natural components, the impact threshold value used, and RR  
23 functions. In this study we use the population weighted PM<sub>2.5</sub> concentration (excluding natural  
24 components) at 1x1 degree resolution as metric for estimating health effects due to air, with a  
25 threshold value of 5.8 µg/m<sup>3</sup>, no urban increment adjustment, and relative risk functions  
26 accordingly with Burnett et al. (2014). We also estimate that 7 % of the global non accidental  
27 mortalities from the Global Burden of Disease (<http://vizhub.healthdata.org/gbd-compare>;  
28 Forouzanfar et al. (2015)) are attributable to air pollution in 2010; 8.6% of total mortality in  
29 Europe is due to air pollution, ranging from less than 1% up to 17% depending on the country;  
30 similarly, Asian premature mortality due to air quality is equal to 8.7% of total Asian mortality,  
31 with 10.6% contribution in China and 8.5% in India. Lower values are found for African  
32 countries and Latin America where other causes of mortalities are still dominant compared to  
33 developed countries.

34 Table 5 shows the number of premature deaths for each receptor region, highlighting the  
35 premature mortality induced within the country itself and outside the receptor region. The PD  
36 induced by Chinese and Indian emissions are mainly found within these two countries; however,  
37 the annual PDs caused by China and India in external regions contribute for ca 700 thousand and  
38 ca 500 thousand PD/year, respectively, representing more than 50% of the global mortality.  
39 Clearly, reducing emissions and emission uncertainties in these two regions will have therefore  
40 the largest over-all benefit on global air quality improvement and understanding as well as on  
41 global human health. For most of the TM5-FASST regions, PDs due to anthropogenic emissions  
42 within the source region are higher than the extra-regional contributions. However, there are  
43 marked exceptions, such as the Gulf region, Hungary, Czech Republic, Mongolia, etc., where the  
44 extra-regional and within-region contributions to mortality are at least comparable. In fact  
45 Hungary and Czech Republic are strongly influenced by polluted regions in Poland (mainly);

1 likewise Mongolia is suffering from the vicinity of source in China. The Gulf region produces a  
2 lot of its own pollution, but is also influenced by transport from Africa and Eurasia as reported  
3 by Lelieveld et al. (2009).

4 Detailed information on the premature deaths for each TM5-FASST region and the contributing  
5 anthropogenic emission sectors is shown in Figs. 8a and 8b. Health effects induced by air quality  
6 in industrialized countries are mainly related with agriculture (32.4% of total mortality or 68  
7 thousand PD/year), residential combustion (17.8% or 37 thousand PD/year) and road transport  
8 (18.7% or 39 thousand PD/year) for Europe and with power generation (26.4% or 26 thousand  
9 PD/year), industry (19% or 19 thousand PD/year), residential (17% or 17 thousand PD/year) and  
10 agriculture (24.0% or 24 thousand PD/year) for North America. The health impacts observed in  
11 most Western EU countries is due both to within-regions and extra-regional pollution, while in  
12 several Eastern EU countries the impact of neighbouring countries is even larger compared to  
13 within-region pollution. The premature deaths induced by international shipping emissions  
14 represent 5.5% of total EU PD, in the range the results of Brandt et al. (2013a) (ca 50 thousand  
15 PDs). PM related mortality in developing countries and fast growing economies is mostly  
16 affected by industrial (up to 42% in China or 279 thousand PD/year) and residential activities  
17 (ranging from 27% in China and 76% in Western Africa), and also by power generation (up to  
18 24% in India or 113000 PD/year). Chinese emissions have a strong impact on China, Japan,  
19 Vietnam, Mongolia+Korea, Thailand while the Indian emissions impact the rest of South and  
20 South Eastern Asia. Reducing Chinese and Indian emissions will reduce the PM related mortality  
21 in almost all countries in Asia. Our results are in agreement with the study of Oh et al. (2015)  
22 where they highlight the role of transported pollution from China in affecting Korean and other  
23 South Eastern Asian countries PM<sub>2.5</sub> concentrations and health effects, as well as the need of  
24 international measures to improve air quality.

## 25 **Conclusions**

26 We coupled the global anthropogenic emission estimates provided by the HTAP\_v2.2 inventory  
27 for 2010 (merging national and regional inventories) to the global source receptor model TM5-  
28 FASST, to study PM<sub>2.5</sub> concentrations and the corresponding health impacts, including an  
29 evaluation of the impacts of uncertainties in national emission inventories. Annual and regionally  
30 averaged anthropogenic PM<sub>2.5</sub> concentrations, corresponding to the 2010 emissions, vary  
31 between ca 1 and 40 µg/m<sup>3</sup>, with the highest annual concentrations computed in China (40  
32 µg/m<sup>3</sup>) India (35 µg/m<sup>3</sup>), Europe and North America (each 8 µg/m<sup>3</sup>). Anthropogenic PM<sub>2.5</sub>  
33 concentrations are mainly due to emissions within the source region, but extra-regional  
34 transported air pollution can contribute by up to 40%, e.g. from China to SE Asia, from EU to  
35 Russia, etc.). Moreover, due to the transport of PM between European countries, EU wide  
36 directives can help improving the air quality across Europe.

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

1 Using the HTAP\_v2.2 emission inventory and TM5-FASST, we also evaluate how the  
2 uncertainty in sectors and regions propagates into PM<sub>2.5</sub>. The aim of our analysis is to provide  
3 insights on where the emission inventories of each country could be improved, because of their  
4 highest uncertainty and highest contribution to the formation of PM<sub>2.5</sub> concentrations. The  
5 uncertainty of PM concentrations depends in variable proportions to the uncertainties of the  
6 emissions within receptor regions, and surrounding regions. We show that reducing the  
7 uncertainties in the Chinese and Indian emission inventories (e.g. from industry and residential  
8 sectors) will be highly relevant for understanding the long-range sources of PM<sub>2.5</sub> pollution in  
9 most of Asian countries. Here we demonstrate how analysis of uncertainties in national/regional  
10 sectorial emission inventories can further inform coordinated transboundary and sector-specific  
11 policies to significantly improve global air quality. Among all anthropogenic emission sectors,  
12 the combustion of biomass for household purposes represents one of the major sources of  
13 uncertainties in emission inventories both in terms of wood consumption and emission factor  
14 estimates. Further effort is therefore required at national level to better characterize this source.

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

20

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1 **Tables and Figures**

2 **Table 1 - Sector specific contribution [%] to annual anthropogenic PM<sub>2.5</sub> concentrations for aggregated world**  
3 **regions. The largest contributing sectors (above a threshold of 15%) are shaded in blue.**

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

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1 **Table 2 - Annual average PM<sub>2.5</sub> concentrations (µg/m<sup>3</sup>) with upper and lower range in brackets due to**  
 2 **emission inventories uncertainty (1 standard deviation, σ). The upper and lower range of PM<sub>2.5</sub>**  
 3 **concentrations are calculated as the reference concentrations multiplied and divided by (1+σ) respectively.**  
 4 **The third column reflects the fractional uncertainty due to the contribution of primary PM<sub>2.5</sub> emissions.**

World region	TM5-FASST region	PM <sub>2.5</sub> concentration (µg/m <sup>3</sup> )	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)
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%

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<b>World region</b>	<b>TMS-FASST region</b>	<b>PM<sub>2.5</sub> concentration (<math>\mu\text{g}/\text{m}^3</math>)</b>	<b>Fraction of uncertainty due to primary PM emissions (%)</b>
<b>Africa</b>	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%
<b>Gulf/Middle East</b>	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%
<b>Latin America</b>	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%
<b>NA</b>	Canada	4.3 (2.4 - 8.3)	66%
	USA	7.8 (4.4 - 14.4)	71%
<b>Russia</b>	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%
<b>Oceania</b>	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%

**Table 3 - PM<sub>2.5</sub> concentrations due to the residential sector emissions in Europe, European part of Russia, Ukraine and Turkey and uncertainty range including the uncertainty in the biomass consumption for the same sector.**

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

**Table 4 – Absolute and population size normalized number of premature deaths/year due to anthropogenic PM<sub>2.5</sub> air pollution in world regions and corresponding uncertainty range.**

	<b>PD (thousand deaths/year)</b>	<b>Normalized PD (deaths/year/million people)</b>
<b>China+</b>	670 (350 - 100)	669
<b>India+</b>	610 (270 - 960)	609
<b>Europe</b>	260 (140 - 480)	405
<b>SE Asia</b>	150 (83 - 250)	50
<b>Russia</b>	110 (67 - 240)	449
<b>North America</b>	100 (55 - 170)	306
<b>Africa</b>	74 (34 - 160)	90
<b>Middle East</b>	56 (32 - 97)	237
<b>Latin America</b>	26 (14 - 53)	49
<b>Oceania</b>	0.055 (0.034 - 0.12)	2

**Table 5 – Number of premature deaths for each receptor region including the within-region and extra-regional attribution. For the RERER metric refer also to Table S2.**

<b>world regions</b>	<b>TM5-FASST region code</b>	<b>PDs in receptor region (deaths/year)</b>	<b>Within-region PDs (deaths/year)</b>	<b>Extra-regional PDs (deaths/year)</b>
<b>Africa</b>	<b>Eastern Africa</b>	16705	8218	8487
<b>Africa</b>	<b>Egypt</b>	17282	11380	5902
<b>Africa</b>	<b>Northern Africa</b>	5424	3427	1997
<b>Africa</b>	<b>Rep. of South Africa</b>	9065	8797	268
<b>Africa</b>	<b>Southern Africa</b>	345	322	23
<b>Africa</b>	<b>Western Africa</b>	25081	19785	5296
<b>Asia</b>	<b>China</b>	655870	643129	12741
<b>Asia</b>	<b>Indonesia</b>	17780	14803	2977
<b>Asia</b>	<b>India</b>	474660	412298	62362
<b>Asia</b>	<b>Japan</b>	25636	15181	10455
<b>Asia</b>	<b>South Korea</b>	25295	7510	17784
<b>Asia</b>	<b>Mongolia+North Korea</b>	12657	4076	8581
<b>Asia</b>	<b>Malaysia</b>	2014	1058	957
<b>Asia</b>	<b>Philippines</b>	121	94	27
<b>Asia</b>	<b>Rest of South Asia</b>	134280	67170	67110
<b>Asia</b>	<b>Rest of South Eastern Asia</b>	23316	3814	19502
<b>Asia</b>	<b>Thailand</b>	21231	10495	10736
<b>Asia</b>	<b>Taiwan</b>	3443	1028	2415
<b>Asia</b>	<b>Vietnam</b>	30750	20286	10464
<b>Europe</b>	<b>Austria+Slovenia</b>	6073	1806	4267
<b>Europe</b>	<b>Bulgaria</b>	4739	1709	3030
<b>Europe</b>	<b>Benelux</b>	9090	4201	4889
<b>Europe</b>	<b>Switzerland</b>	3200	1568	1632
<b>Europe</b>	<b>Czech Republic</b>	7936	2696	5240
<b>Europe</b>	<b>Germany</b>	36256	18595	17661
<b>Europe</b>	<b>Spain+Portugal</b>	11291	8487	2804
<b>Europe</b>	<b>Finland</b>	0	0	0
<b>Europe</b>	<b>France</b>	22046	13320	8727
<b>Europe</b>	<b>Great Britain+Ireland</b>	13949	9459	4490
<b>Europe</b>	<b>Greece+Cyprus</b>	3117	1133	1984
<b>Europe</b>	<b>Hungary</b>	14211	3820	10391
<b>Europe</b>	<b>Italy+Malta</b>	24417	16312	8105
<b>Europe</b>	<b>Norway</b>	674	516	158
<b>Europe</b>	<b>Poland+Baltic</b>	28686	15877	12809
<b>Europe</b>	<b>Rest of Central EU</b>	6764	3418	3346

Europe	Romania	14155	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	10478	8447	2031
Latin America	Rest of Central America	3413	2772	640
Latin America	Rest of South America	4489	4164	325
Middle East	Gulf region	15176	11225	3951
Middle East	Middle East	6784	2804	3980
Middle East	Turkey	34151	24191	9960
North America	Canada	3262	1491	1771
North America	USA	97877	90176	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	10757	6420	4337
Russia	Russia (Asia)	1348	601	746
Russia	Russia (EU)	25149	12704	12445
Russia	Ukraine	71724	44604	27120

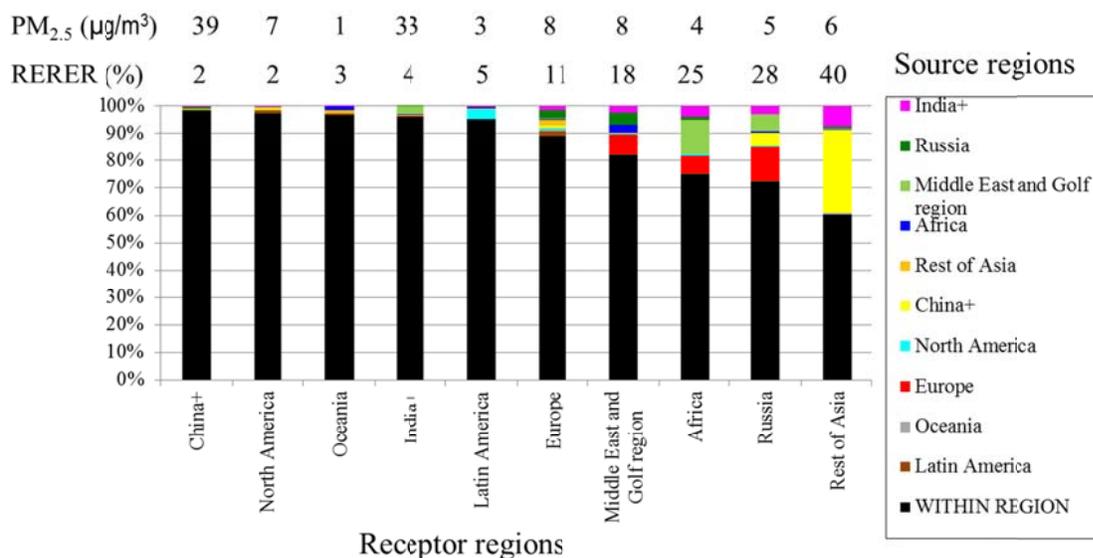


Figure 1 – Within-region vs. imported extra-regional anthropogenic population weighted PM<sub>2.5</sub> concentrations [%] for aggregated world regions. Annual average population weighted anthropogenic concentrations (in µg/m<sup>3</sup>) are reported on top of each bar together with the RERER metric (%). Ship emissions were not included.

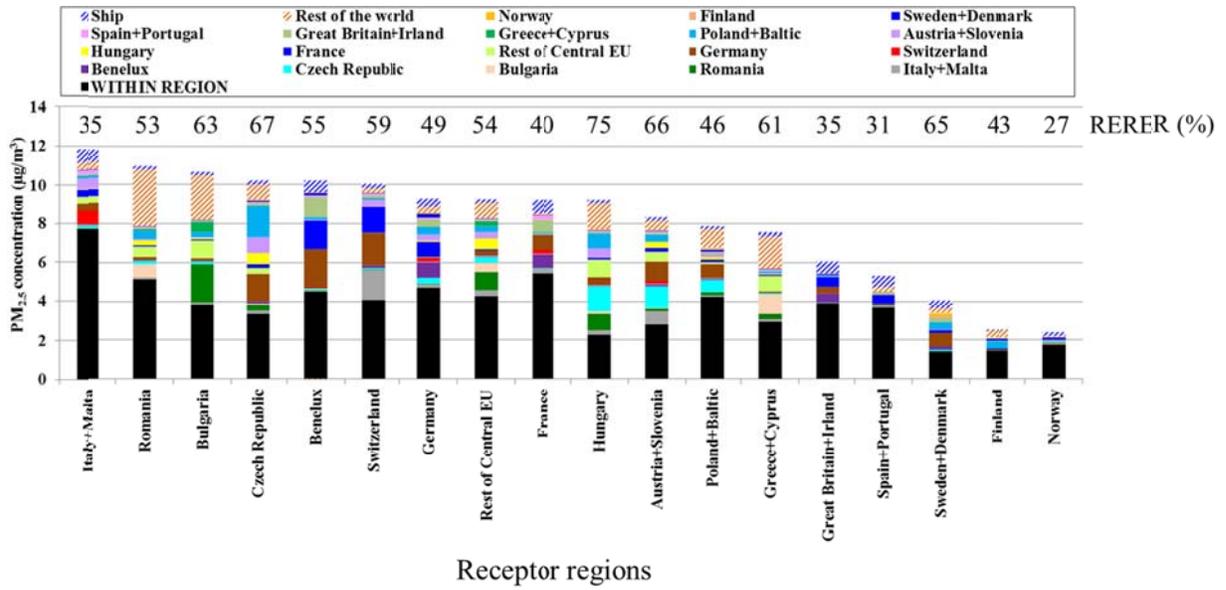


Figure 2 – Anthropogenic PM<sub>2.5</sub> concentrations in 18 countries and sub-regions in Europe separated in within-region and extra-regional contributions. The RERER metric (%) is reported on top of each bar.

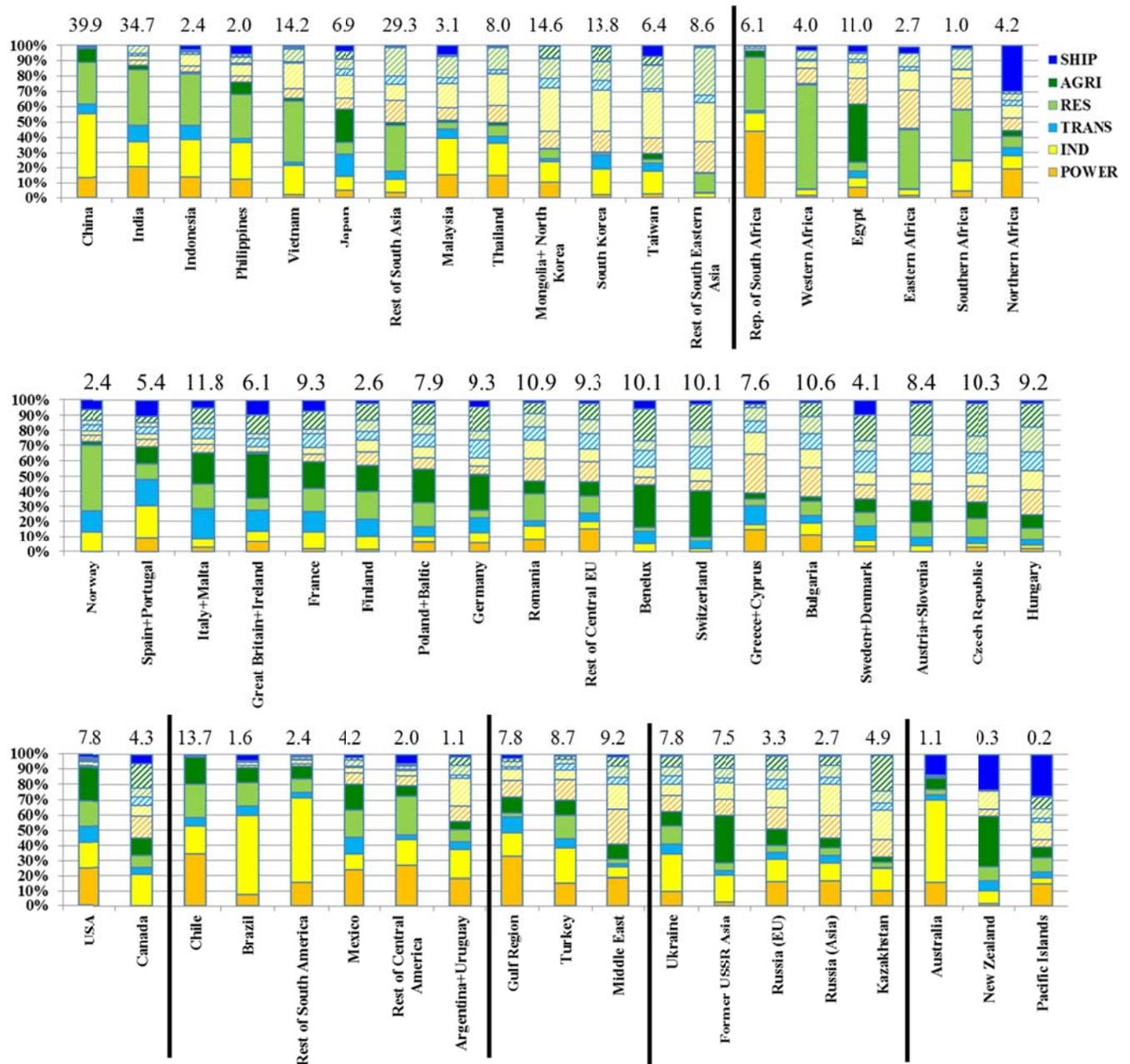


Figure 3 - Fraction of within-region and extra-regional (shaded areas) anthropogenic PM<sub>2.5</sub> concentrations separate by sector for receptor region within the macro-regions: Asia and Africa (upper panel), Europe (middle panel), North America, Latin America, Middle East, Russia and Oceania (lower panel). Annual averaged anthropogenic concentrations (in  $\mu\text{g}/\text{m}^3$ ) are reported on top of each bar. The RERER metric (%) for the 56 TM5-FASST regions is also reported in Table S2.

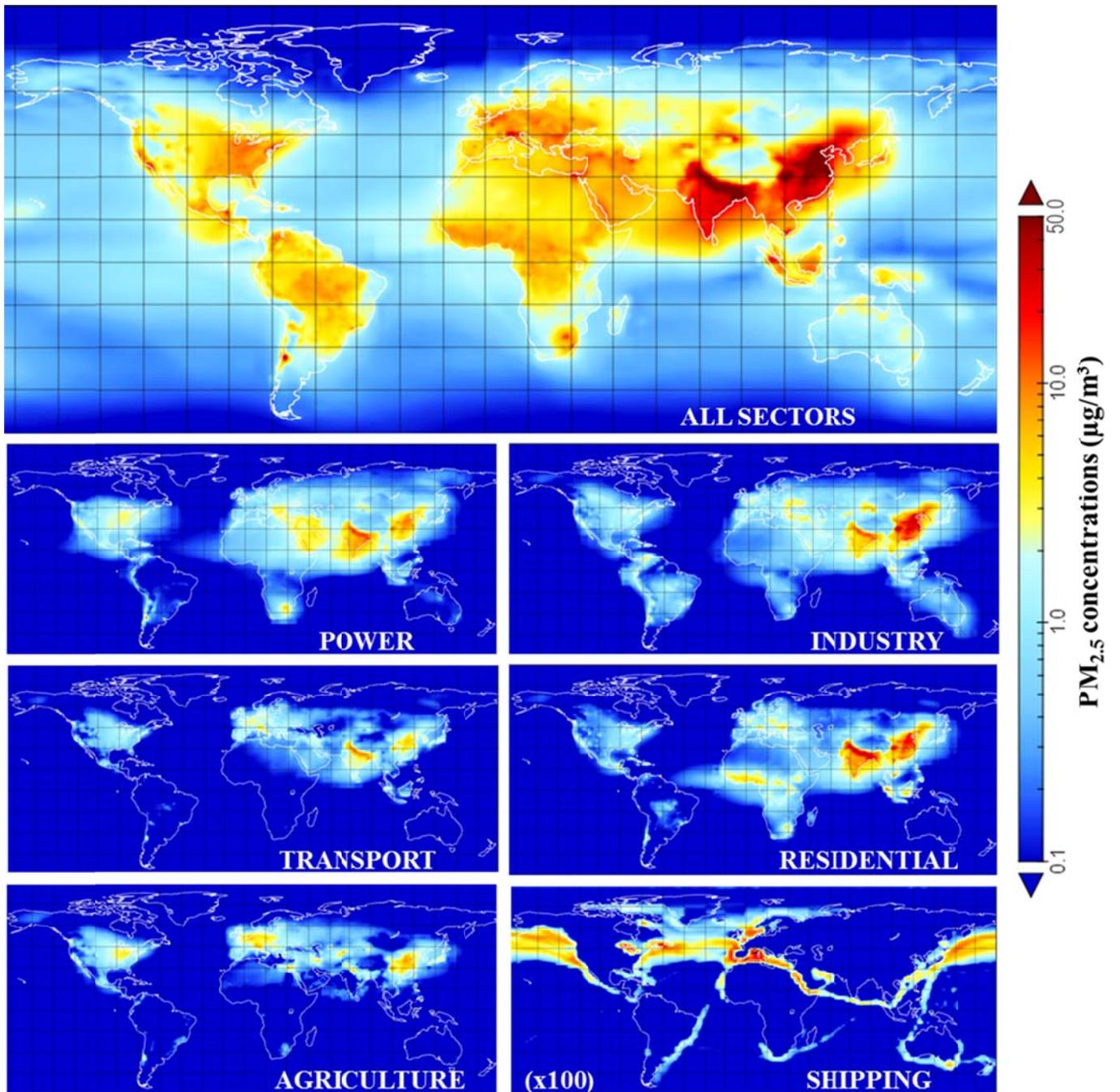


Figure 4 – Total anthropogenic PM<sub>2.5</sub> concentrations (µg/m<sup>3</sup>) and sectorial contributions using 2010 emissions.

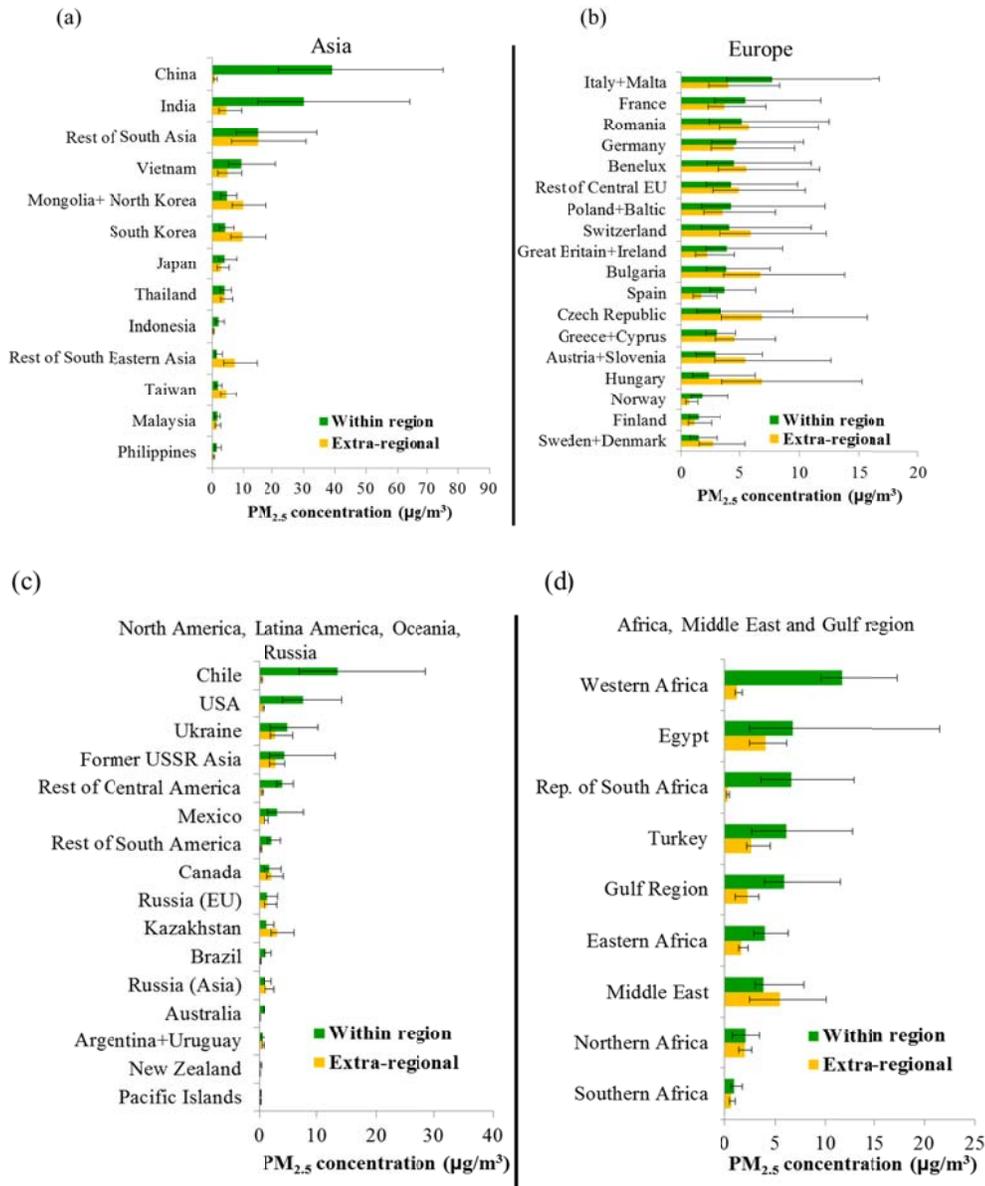


Figure 5 - Within-region and extra-regional anthropogenic PM<sub>2.5</sub> concentrations for Asia (panel *a*), Europe (panel *b*), North America, Latin America, Oceania and Russia (panel *c*) and Africa, Gulf region and Middle East (panel *d*). The error bars are calculated multiplying and dividing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

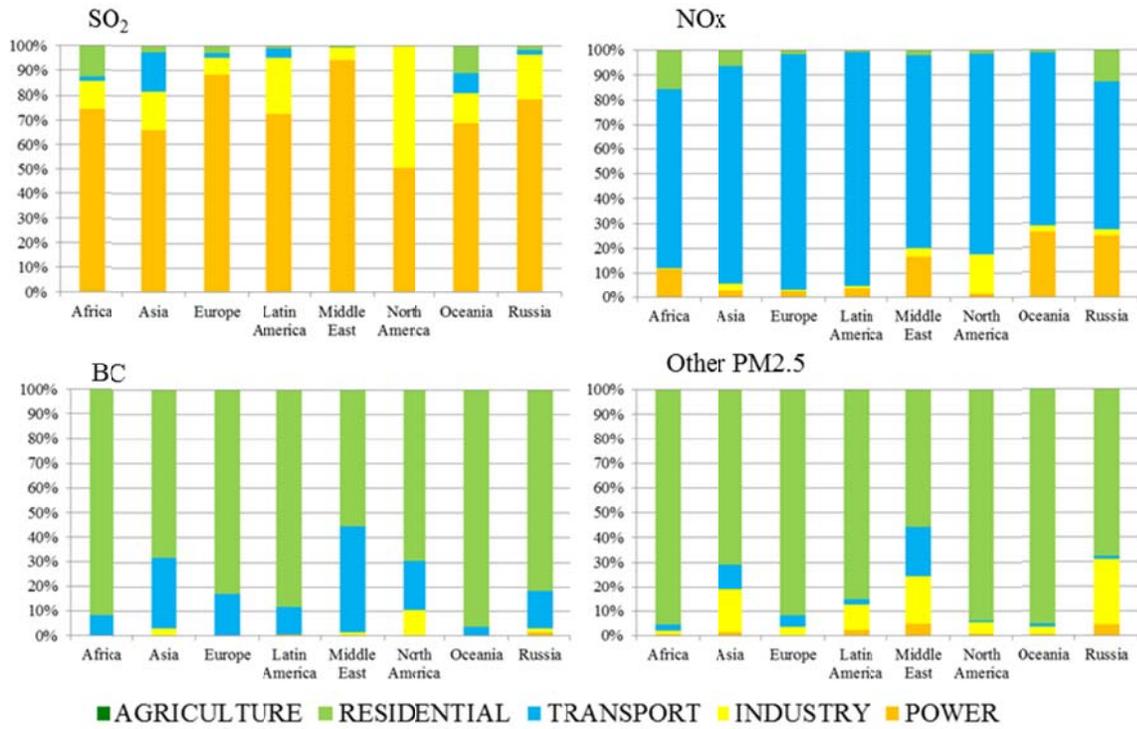
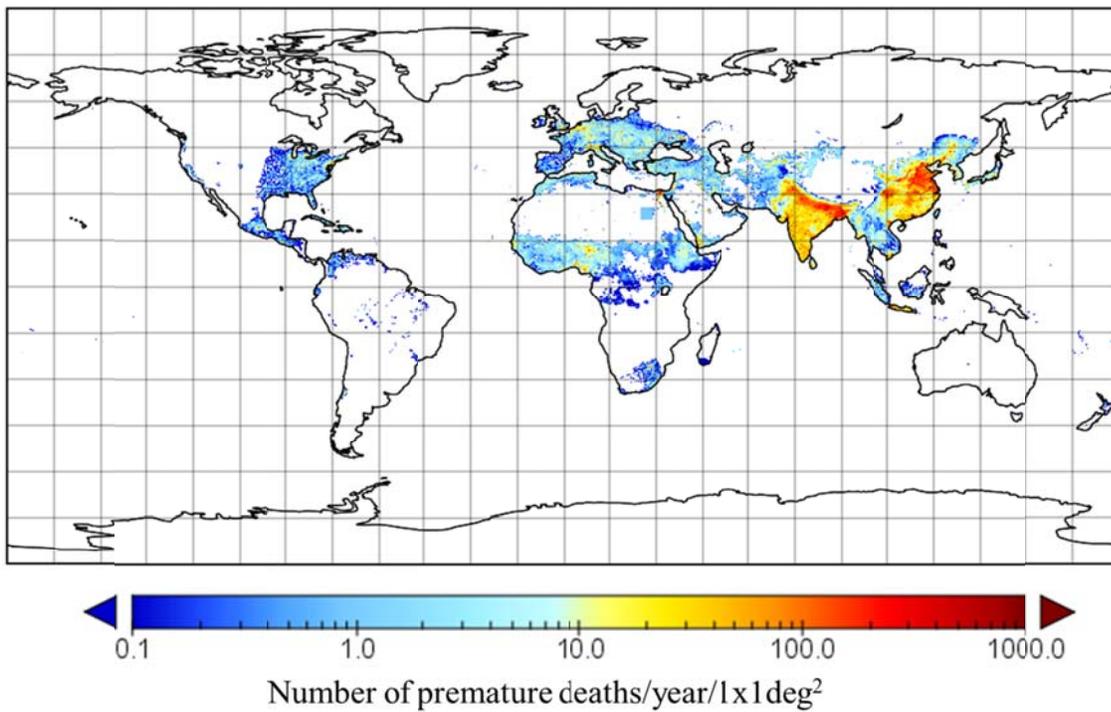
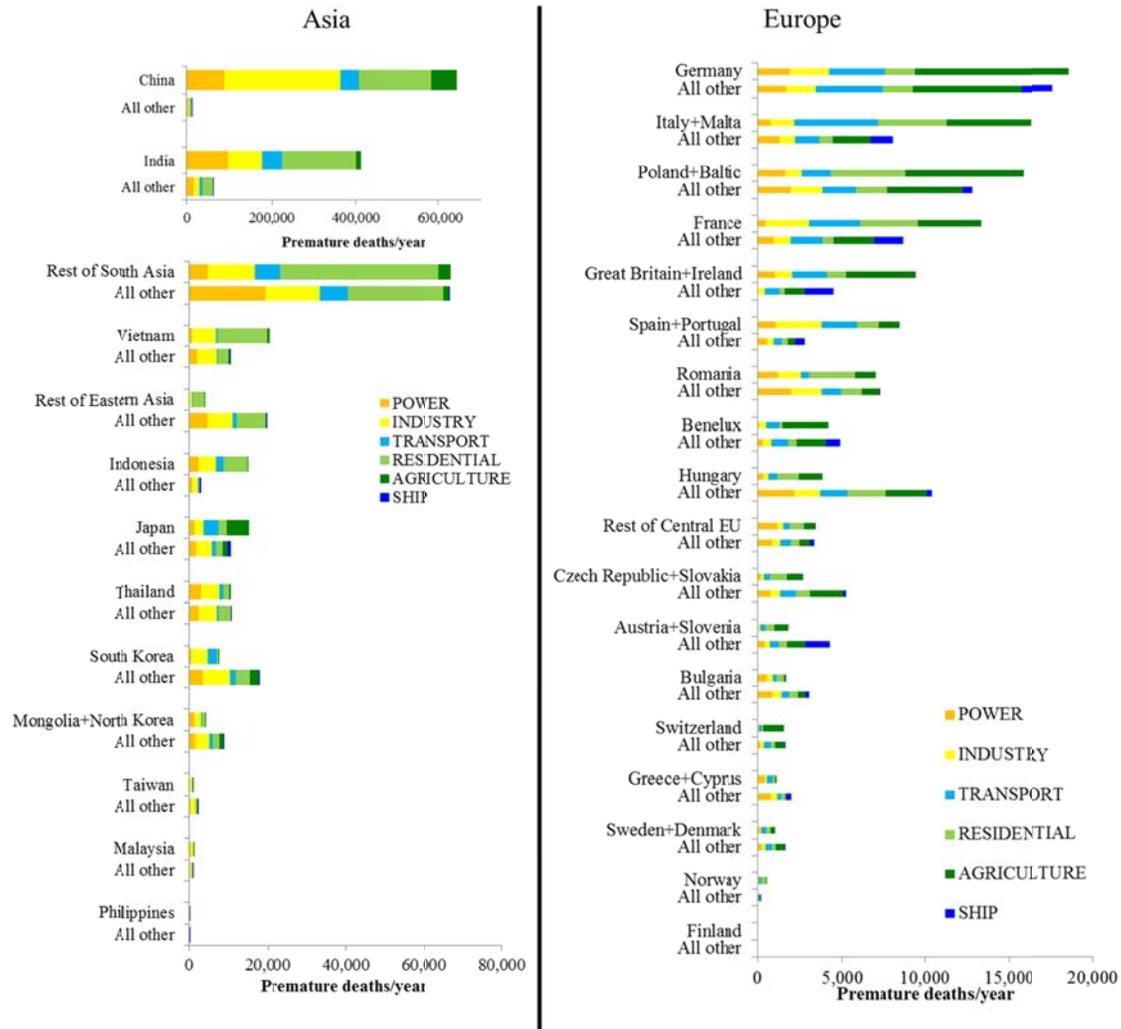


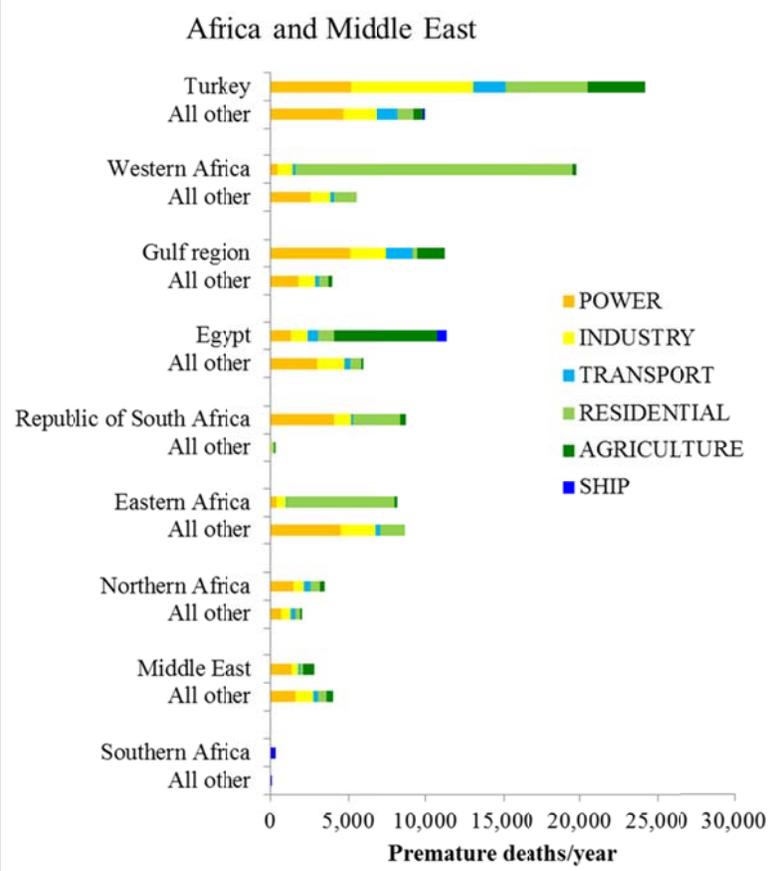
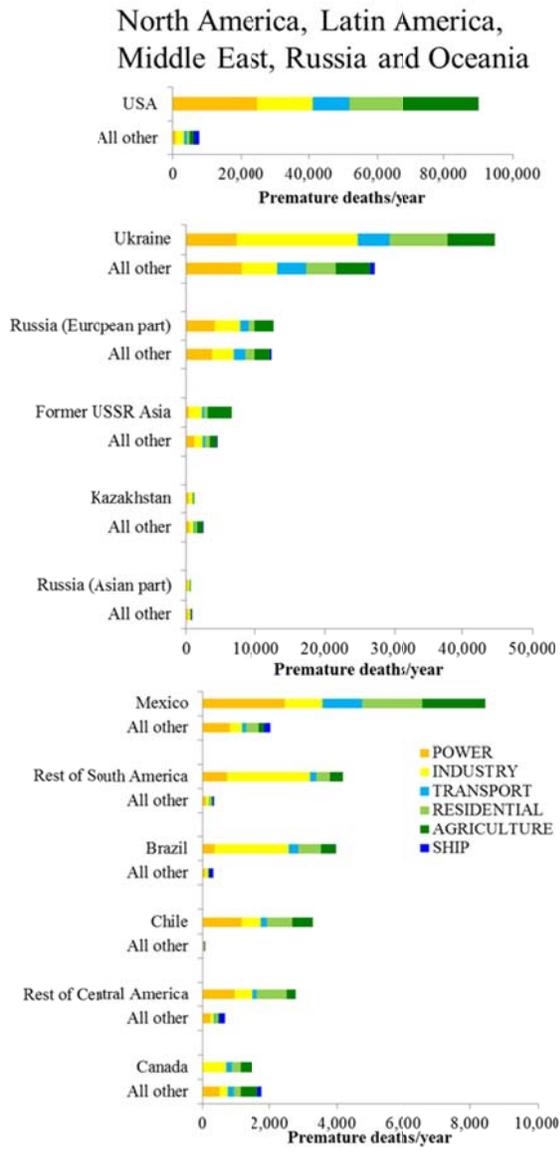
Figure 6 – Contribution of anthropogenic sectors to the emission uncertainty of various pollutants for different world regions.



**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<sub>2.5</sub> population weighted concentrations in the TM5-FASST receptor regions of Asia (left) and Europe (right).**



**Figure 8b – Anthropogenic emission sector contributions to premature mortality (deaths/year) due to PM<sub>2.5</sub> 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.**