The authors are grateful to Referee#1 for the helpful comments that helped improve the manuscript. Due to the strict link between this publication and the work recently submitted by van Dingenen et al. (submitted, 2018) about the TM5-FASST methodology, we offered the possibility to the Editor and the Reviewer to access the work of van Dingenen et al. (submitted, 2018) although not yet published in ACPD. Thanks to the Reviewer's comments, we also realized that some methodological aspects of the TM5-FASST tool could have been further developed also in the publication of van Dingenen et al. (submitted, 2018). Therefore, discussions on the comparison between PM2.5 modeled concentrations vs. the measured ones, as well as further details about the extension of the "perturbation approach" to the attribution of sectors and sources will be included in the review phase of the paper by van Dingenen et al. (submitted, 2018). We feel that we have been able to address all concerns, as outlined below.

Anonymous Referee #1

The manuscript by Crippa et al. investigates the regional and sectoral contributions to PM2.5 and associated health impacts throughout the world. This is accomplished through application of the TM5-FASST response tool. This topic is useful and their results are new, and also appropriate for the scope of this journal. They also provide a much needed estimation of how uncertainty in the emissions estimates propagate into uncertainties in PM2.5 estimates, which is a source of error not often well quantified in health impact studies. That being said, the manuscript good use more attention to previous works, especially in the introduction. These and some additional comments are highlighted below, which include requests for more information about the fidelity of the modeling estimates used here, and the impact of a few assumptions in its application that are made but not evaluated either through their own work presented here or references to literature (i.e. assuming PM2.5 responds linearly to emissions changes, or that anthropogenic SOA is negligible). Addressing these concerns constitutes major revisions, after which point this manuscript will be suitable for publication in ACP.

Major:

1.35: I wonder if the authors considered including some more recent estimates e.g. from the Global Burden of Disease project on estimated numbers of premature mortalities from ambient PM2.5 exposure, such as Cohen et al., The Lancet, 2017.

Ok, I see that relevant works be e.g. Lelieveld (2015), Silva (2016) or Cohen (2017) are finally discussed on page 11. Such works however should be discussed as part of the introduction and background information, in order to more clearly articulate the role of the present work.
In general the introduction was lacking in some detail with regards to previous works that have considered sector-specific health impacts, the role of model uncertainty vs emissions uncertainties or uncertainties in concentration-response parameterizations on estimates of PM2.5 health impacts.

As suggested by the Reviewer, the following sentences have been added to the introduction:

"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/mortality (PD) 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) accounting for 3.9 million premature deaths/year due to different model assumptions. In Europe, Brant et al. (2013) estimate 680 thousand 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. (2017) report a multi-model mean estimate of PD of 414.000 (range 230-570 thousand) for Europe and 160 thousand 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 with incomplete understanding of transport, chemical transformation, removal processes, and not the least, emission information."

2.1: Suggest adding references to any number of studies that have estimated the human health impacts of sector-specific policies for PM2.5 reduction.

As suggested by the reviewer we added in the manuscript the following some references related with studies on human health impacts of sector-specific PM2.5 contributions:

"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 Transport of Air Pollution (CLRTAP). At city/local level, several studies have been developed to assess the contribution of sector specific emissions to PM2.5 concentrations with the aim of designing air quality plans at local and regional level (Karagulian et al., 2015; Thunis et al., 2016)."

Equation 1: This equation is an approximation, not an equals sign. This should be clearly indicated, and the error associated with ignoring second-order terms should be discussed, either using evidence from the own authors work or from reference to many previous studies in the literature that have explored the nonlinear response of PM2.5 to emissions perturbations.

Equation 1 represents how PM concentrations can be estimated using the 20% perturbation which is the basis of the TM5-FASST methodology. So the equal sign is correct, although this equation represents an approximation due to errors both of the chemistry and transport modeling and to the emissions. We refer the Reviewer to the paper by van Dingenen et al (submitted, 2018) for details about the errors due the chemistry and transport, while in this work we address mainly the errors due to emissions. Below additional details about the TM5-FASST methodology:

The reduced-form model TM5-FASST is computing the concentration resulting from an arbitrary emission scenario E_s using a perturbation approach, i.e. the difference between E_s and E_{ref} (dE_s) is considered as a perturbation on E_{ref} and the resulting concentration is evaluated as a perturbation dPM on the reference concentration, hence:

$PM(E_s) = PM(E_{ref} + dE_s) = PM_{ref} + dPM = PM_{ref} + SRC \cdot dE_s$ (a)

Where $dE_s = E_s - E_{ref}$ and E_{ref} is the RCP reference scenario from which the *SRC* have been computed.

The contribution of a single sector *j* is calculated as the difference between the concentration including all sectors, and the concentration from the emissions excluding the single sector *j*

$PM(E_{s,j}) = PM(E_s) - PM(E_s - E_{s,j}) = SRC \cdot [dE_s - d(E_s - E_{s,j})] = SRC \cdot E_{s,j}$

If the linearity holds, the sum of $PM(E_{s,j})$ over all sectors j should be equal to $PM(E_s)$, or:

$$\sum_j PM(E_{s,j}) = PM_{ref} + SRC \cdot (E_s - E_{ref})$$

The TM5-FASST runs were performed for different scenarios, comparing the reference HTAP_v2.2 emissions with a scenario where emissions from one single sector were subtracted from the total emissions. Then comparing the reference case and each scenario (REF-sector$_i$), the contribution of each sector to PM2.5 concentrations is estimated. This approach is based on the assumption that the individual sector contributions add up linearly to total PM2.5, as mentioned in the paper. The paper by Van Dingenen et al. describing the whole TM5-FASST methodology has just been submitted to ACP (van Dingenen et al., submitted, 2018) Equation 1 represents the basis of the TM5-FASST method, since it describes how a variation in the emissions (delta emissions) determines a delta in PM2.5 based on the source receptor relationships.

The following discussion on how to apply the "perturbation approach" on the sector and source attribution will be also included in the paper by van Dingenen et al. (submitted, 2018):

Equation (2) expresses the 'perturbation' approach applied in the linearized TM5-FASST model, i.e. an arbitrary emission scenario is evaluated as a deviation from the base emission scenario, and the resulting pollutant concentration is obtained as the sum of the base concentration and a delta term, the latter proportional to the emission deviation from the base case (Figure 1).

A particular application of TM5-FASST is the attribution of the (anthropogenic) pollutant concentration to individual source regions or sectors. Due to the fixed contribution of the base concentration which does not contain information on the originating sources, Eq. (2) is not immediately suitable for such an analysis. Instead, we calculate for each individual source the contributing part by first evaluating all sources together ('total' simulation'), and subsequently subtracting the individual source emissions (E_s) from the total, evaluating the resulting pollutant concentration (C_{minus_s}), and making the difference with the 'total simulation' to obtain the single source contribution (C_s).

$C_{j,tot}(y) = C_{j,base}(y) + \sum_{n_x} \sum_{n_i} A_{ij}[x,y] \cdot [E_{i,tot}(x) - E_{i,base}(x)]$ (2)

$$C_{j,minus\_s}(y) = C_{j,base}(y) + \sum_{n_x}\sum_{n_i} A_{ij}[x,y] \cdot \left[E_{i,tot}(x) - E_{i,s}(x) - E_{i,base}(x)\right] \quad (4)$$

$$C^*_{j,s}(y) = C_{j,tot}(y) - C_{j,minus\_s}(y) = \sum_{n_x}\sum_{n_i} A_{ij}[x,y] \cdot E_{i,s}(x) \quad (5)$$

We can now reconstruct $C^*_{j,tot}$ as the sum of the individual source contributions:

$$C^*_{j,tot}(y) = \sum_{n_s} C^*_{j,s}(y) \quad (6)$$

$C^*_{j,tot}(y)$ is equivalent to $C_{j,tot}(y)$ in Eq. 2 only if

$C_{j,base}(y) = \sum_{n_x}\sum_{n_i} A_{ij}[x,y] \cdot E_{i,base}(x)$, in other words if the emission-concentration relation is perfectly linear and passes through the origin.

In reality there is some degree of non-linearity in most emission-pollutant relation as illustrated in Figs. 3 and 6. Figure A shows for each of the FASST regions the total $PM_{2.5}$ concentration obtained by Eq. 6 versus the TM5 base simulation result, illustrating the non-linearity error resulting from the application of Eq. 6. For 43 out of 56 regions, the deviation from the base simulation is less than 30%, only 3 regions (former Soviet Union, New Zealand and Pacific) deviate more than 50% from the TM5 model result. Consistency with the 'perturbation approach' is restored by simply rescaling the individual source contributions:

$$C_{j,s}(y) = \frac{C_{j,tot}(y)}{C^*_{j,tot}(y)} C^*_{s,j}(y) \quad (7)$$

This approach is valid for evaluating the attribution by sector as well as by source region.

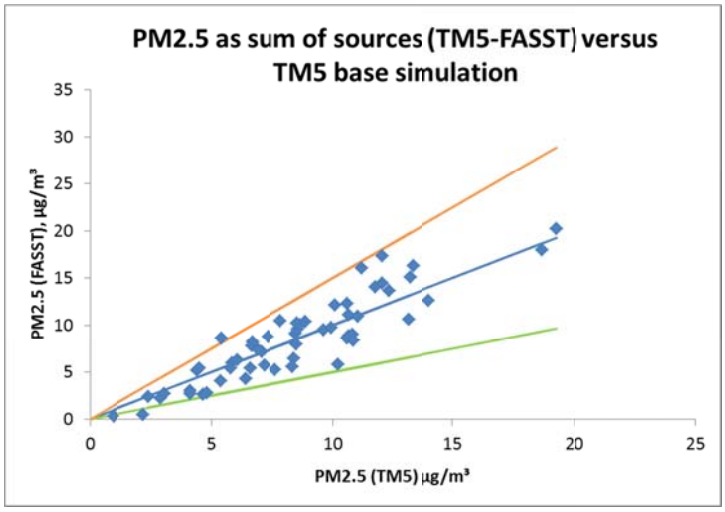
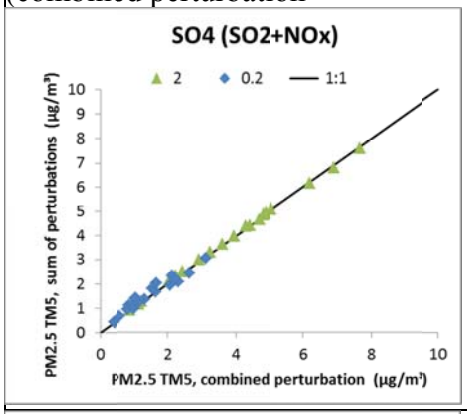
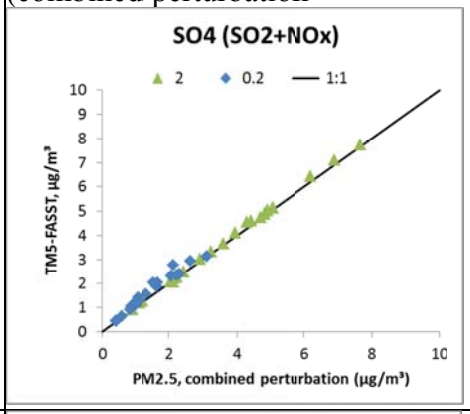
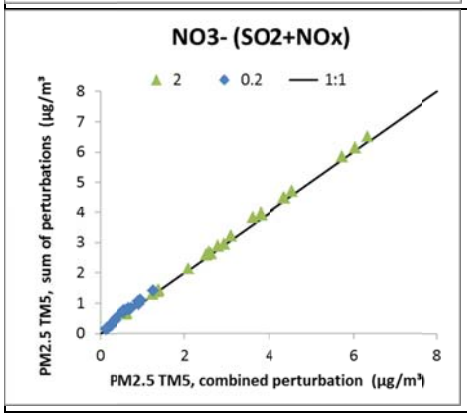
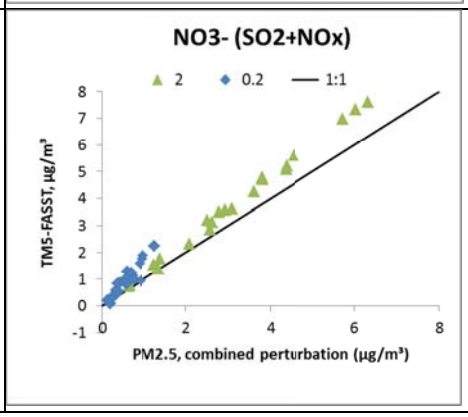


Figure A: Scatter plot of regionally averaged PM2.5 concentration (including all anthropogenic components) obtained as the sum of individual source region contribution by linear scaling of

their respective emissions with TM5-FASST source-receptor coefficients (Eq. 6), versus the regional average obtained by the full TM5 model.

Section 2.2: Some essential details of the TM5-FASST model are missing. What is the accuracy of the baseline PM2.5 (total, and speciated) concentrations estimated by TM5 fast compared to in situ measurements in different parts of the world? In locations where such data is not available, how do the model estimates compare to those from other models, or from remote-sensing derived products? How much error is expected owing to the coarse model resolution when estimating population-weighted concentrations, given the relatively high-resolution variability in population densities?

In the work by van Dingenen et al. (submitted on the 31st of January 2018 to the ACP HTAP special issue) details about the comparison between the linearized TM5-FASST model and the full TM5 runs are provided in Section 3.1 "Validation against the full TM5 model: additivity and linearity". They also report the linearity and additivity issues for PM2.5 and its speciation in Figure 3 of their paper, as shown below.

TM5 (sum of perturbations) vs TM5 (combined perturbation	FASST sum of extrapolated perturbations) vs TM5 (combined perturbation
	
	

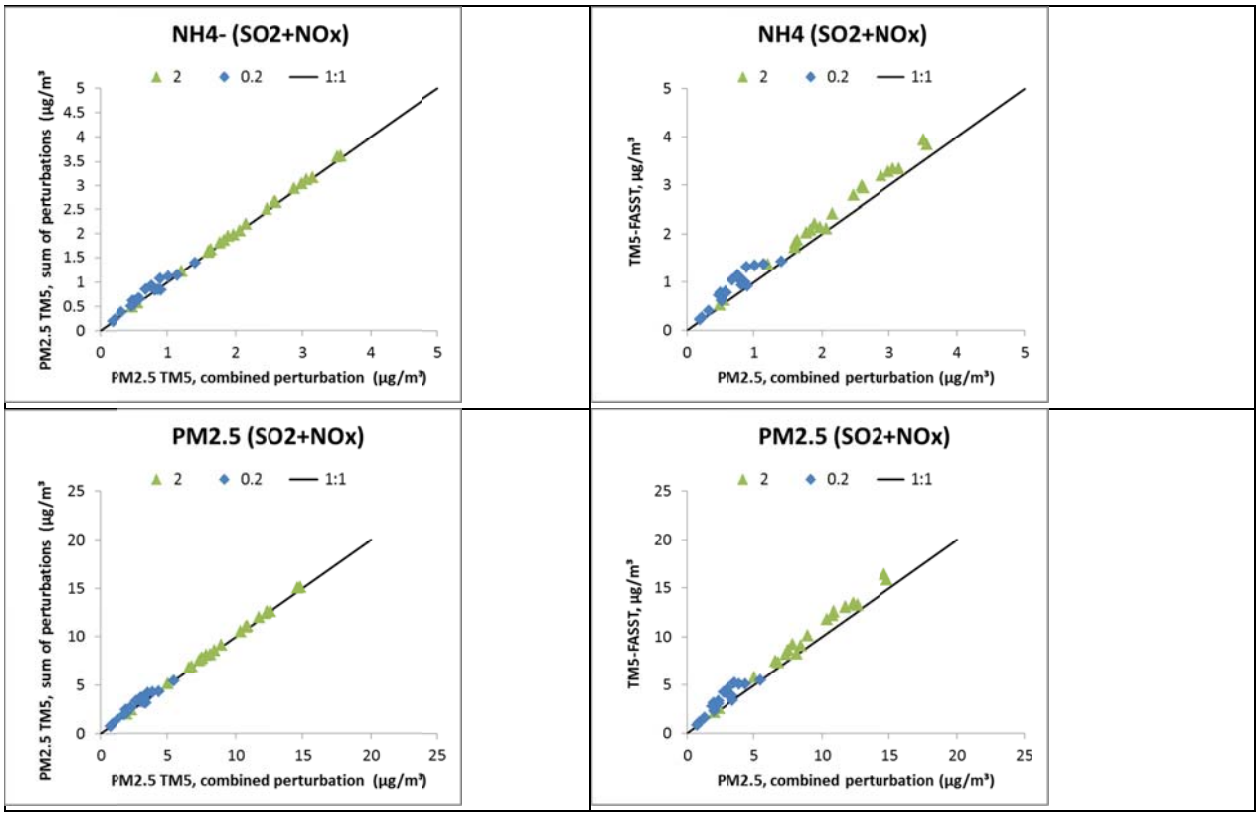


Figure 1. Additivity and linearity test of perturbations using TM5 outcome for regional population-weighted mean secondary inorganic PM2.5 concentrations for 3 perturbation magnitudes (green: +100%, red: -20%, blue: -80% relative to base simulation emissions). X-axis: simultaneous perturbation of SO2 and NOx emissions Left column Y-axis: sum of TM5 concentration response to two individual SO2 and NOx perturbations. Right column Y-axis: sum of linearly extrapolated individual 20% perturbations (FASST approach). Each point corresponds to the population-weighted mean concentrations over a receptor region (same regions as in Fig. 2).

Ok - while I do see that there is a single paragraph addressing this in very broad strokes, on page 7 (21-28), this evaluation is incommensurate with the scales of the paper. Given the regional, sectoral and species specificity of the source attribution results, the authors need to examine model fidelity on the same scales.

In addition, van Dingenen et al. (submitted, 2018) report in Fig.7 of their publication the comparison of modeled PM2.5 concentrations between the full TM5 runs and the TM5-FASST ones, as shown below.

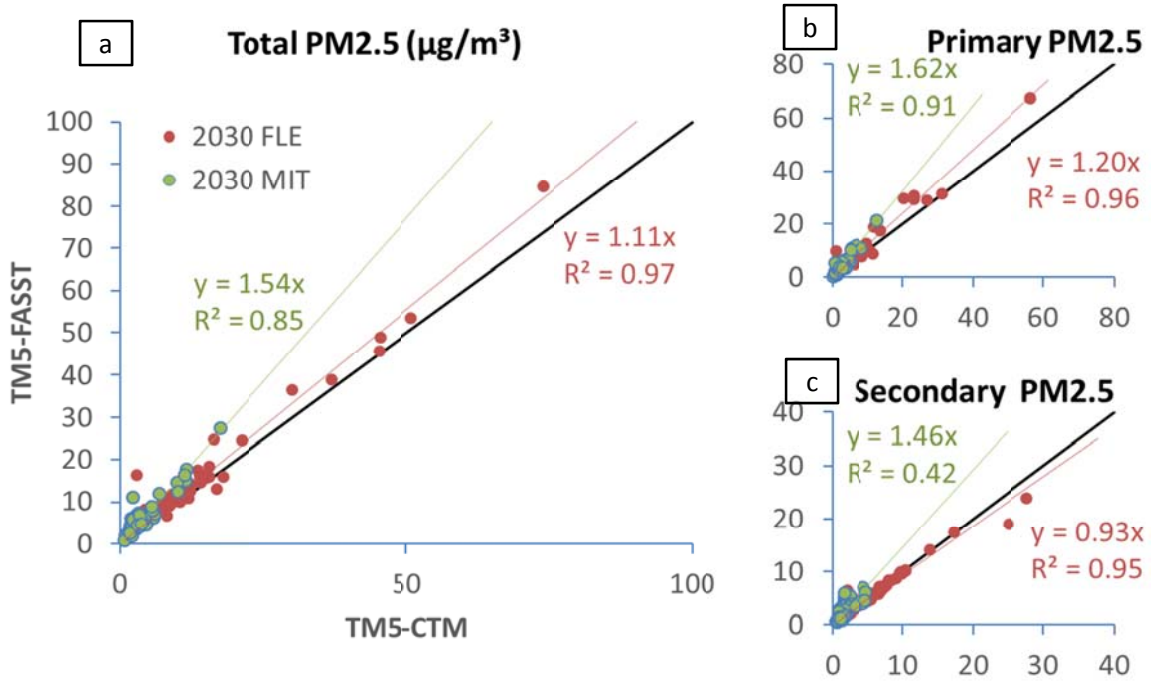


Figure 2. (a) PM2.5 concentration obtained with TM5-FASST versus TM5-CTM for high (FLE, red dots) and low (MIT, green dots) emission scenarios (see text). Each point represents the population-weighted mean over a TM5-FASST receptor region. Black line: 1:1 relation. breakdown for (b) primary (BC+POM+other primary PM2.5) and (c) secondary (SO4+NO3+NH4) PM components (same axis definitions as left plot).

In addition, van Dingenen et al. validated their modeled PM2.5 concentrations against in situ measurements and satellite derived data, as discussed in the following.

Figure 3 shows the comparison between the PM2.5 concentrations modeled by TM5-FASST and the measured ones reported in the WHO database for different world regions (i.g. EUR=Europe, NAM=North America, China, S-ASIA=Southern Asia, LAM=Latin America, AFR=Africa). This includes measurement points as well as PM2.5 concentration estimates based on a fraction of PM10 measurements (e.g. almost all points for the comparison in China are based on this second method). Quite good agreement is observed for Europe, North America and partly China where measurements have been performed over longer time compared to developing countries and they are based on quite consolidated methods. The comparison for Latin America and Africa is much less robust and the scatter possibly highlights a non-optimal modeling of large scale biomass burning for the TM5-FASST model. Figure 4 reports the comparison of WHO regional average of urban stations against the FASST population weighted average of grid cells. Similarly to the findings of Figure 1, the comparison for industrialized countries is very good, while for other developing regions the agreement is less satisfactory both due to less accurate measurements (e.g. reported by WHO) and lower quality modeling of specific sources by TM5-FASST (e.g. large scale biomass burning).

TM5-FASST modeled PM2.5 concentrations have been also validated against satellite products (see Figure 6) 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).

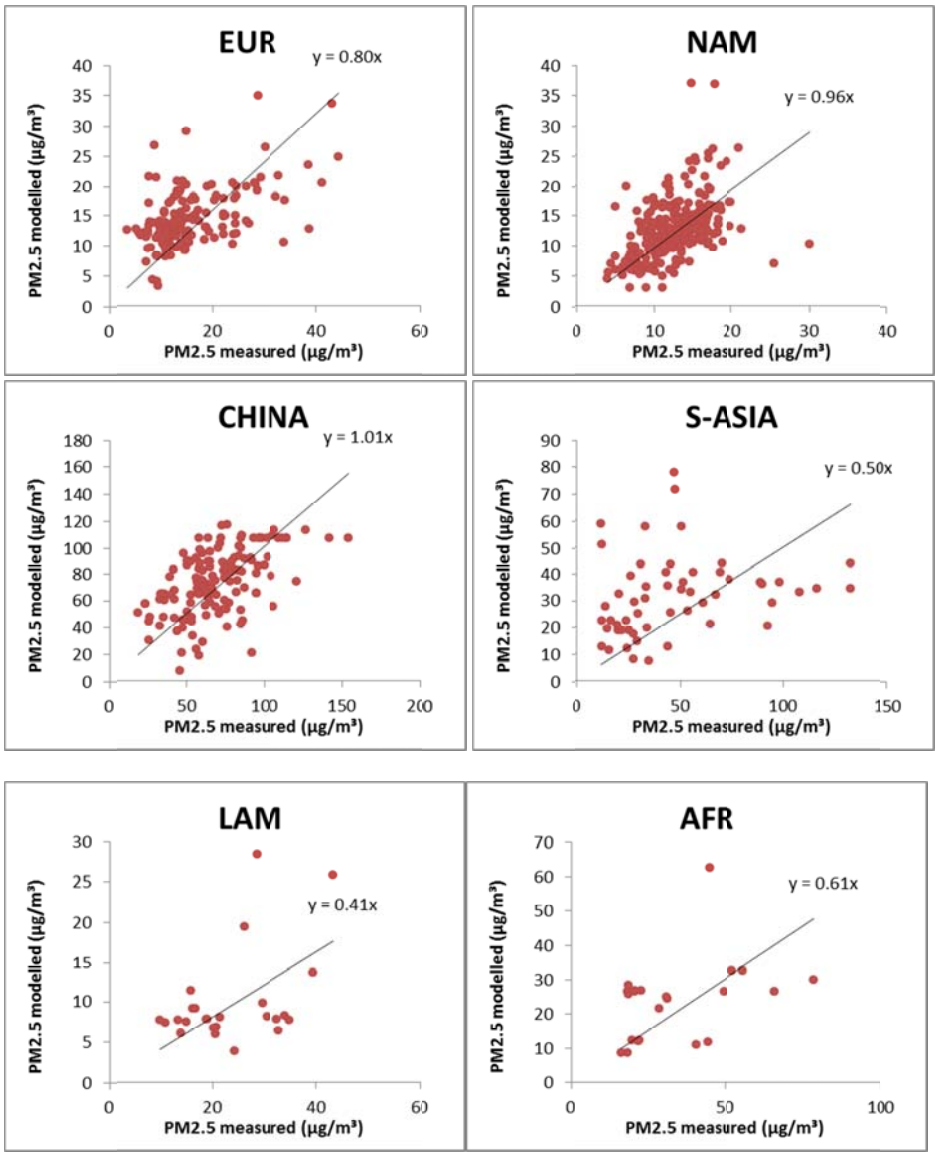


Figure 3 - TM5-FASST grid-cell mean (with urban increment parameterisation) versus individual monitoring stations (WHO consolidated database, including both measured and estimated PM2.5).

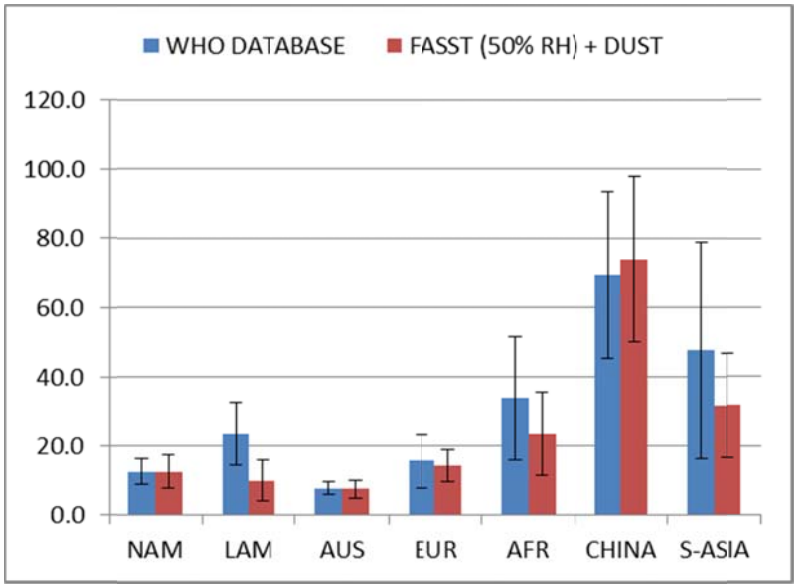


Figure 4 - WHO regional average of urban stations (+/- 1 stdev) and FASST population weighted average of grid cells.

Figure 5 shows the population-weighted country-averaged PM2.5 concentrations from the WHO database on outdoor air pollution (years 2011, 2014 and 2016) and from TM5-FASST for the year 2010 (using the ECLIPSE/HTAP emission inventory). WHO data are compiled from mostly urban or urban background monitoring stations, and include PM2.5 estimates based on measured PM10 when PM2.5 measurements are not available. FASST PM2.5 represents dry PM2.5 while WHO monitoring data include residual water from equilibration between 35 – 50%RH. TM5-FASST includes a generic fixed dust and sea-salt field, as well as an urban increment correction on primary anthropogenic PM2.5 where appropriate. TM5-FASST averages include all countries grid-cells, WHO includes only values form monitoring station locations and is therefore biased towards higher values compared to TM5-FASST.

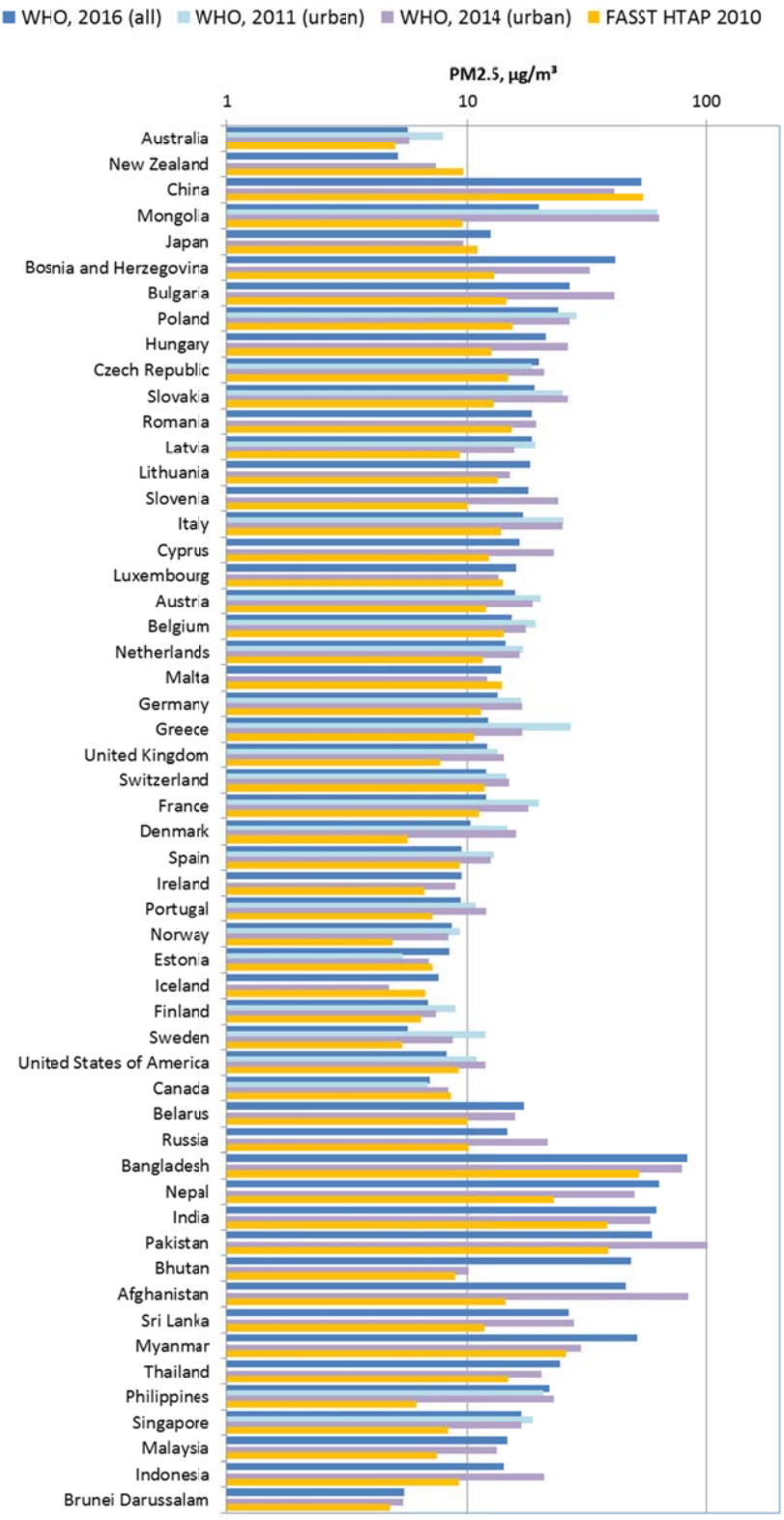

WHO, 2016 (all)
WHO, 2011 (urban)
WHO, 2014 (urban)
FASST HTAP 2010
PM2.5, µg/m³
1
10
100
Australia
New Zealand
China
Mongolia
Japan
Bosnia and Herzegovina
Bulgaria
Poland
Hungary
Czech Republic
Slovakia
Romania
Latvia
Lithuania
Slovenia
Italy
Cyprus
Luxembourg
Austria
Belgium
Netherlands
Malta
Germany
Greece
United Kingdom
Switzerland
France
Denmark
Spain
Ireland
Portugal
Norway
Estonia
Iceland
Finland
Sweden
United States of America
Canada
Belarus
Russia
Bangladesh
Nepal
India
Pakistan
Bhutan
Afghanistan
Sri Lanka
Myanmar
Thailand
Philippines
Singapore
Malaysia
Indonesia
Brunei Darussalam

Figure 5- Population-weighted country-averaged PM2.5 concentrations from the WHO database on outdoor air pollution (years 2011, 2014 and 2016) and from TM5-FASST for the year 2010 (using the ECLIPSE/HTAP emission inventory).

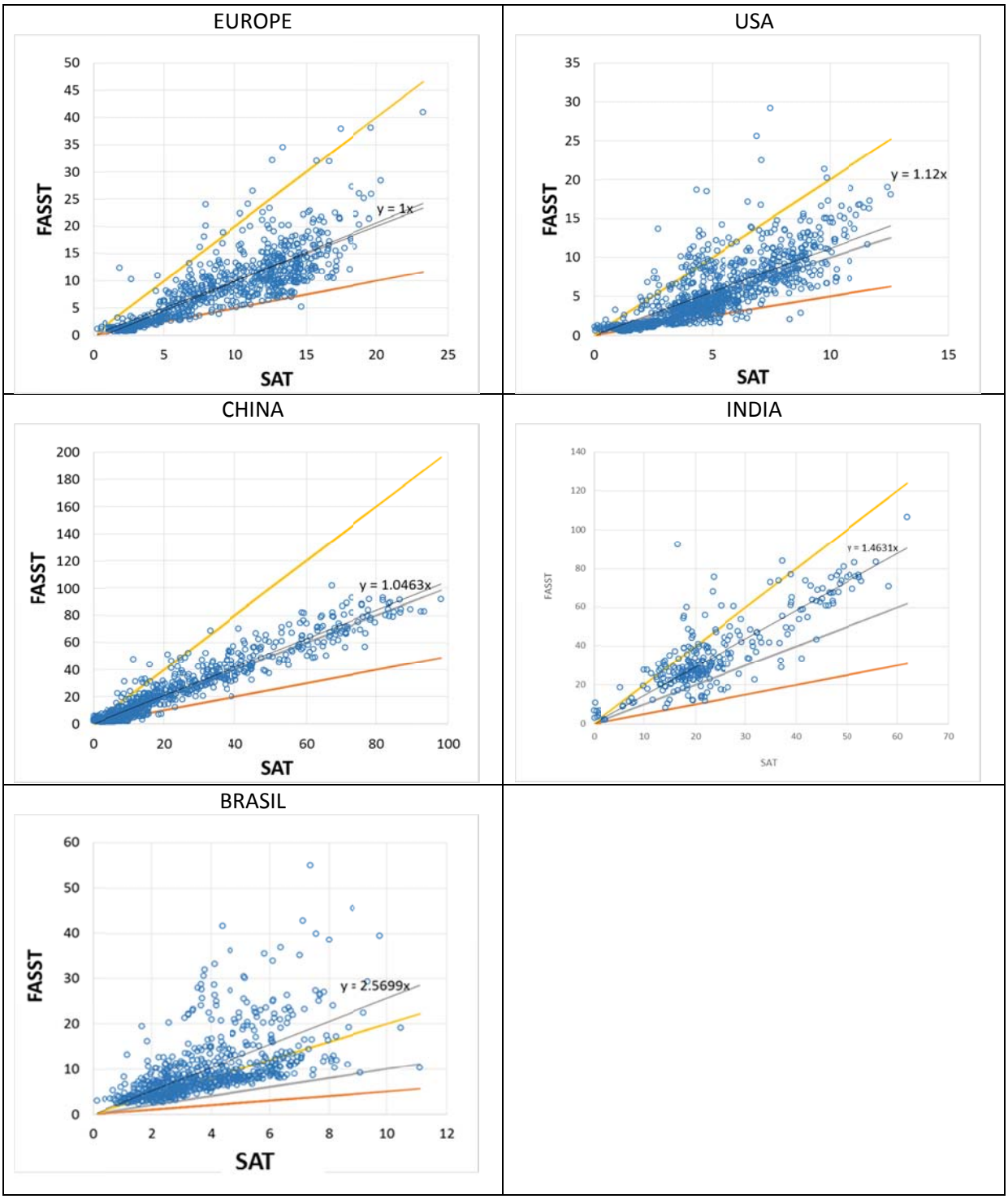


Figure 6 - Comparison between TM5-FASST and satellite products for world regions (Boys et al., 2014; van Donkelaar et al., 2010, 2014).

While for the full details and discussion with refer to Van Dingenen et al. (2018), we summarize their results in our manuscript as following:

In section 2.1 we added the following sentence:

"The TM5-FASST model 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 performance with regard to linearity and additivity of concentration response to different size emission perturbations and future emission scenarios. The validation of TM5-FASST against the full TM5 model runs is extensively discussed by van Dingenen et al. (2018), as well as the validity of the assumptions of linearity and additivity behind this reduced form-model. Below we summarize the most important features of relevance for this work, and refer for more detail to Van Dingenen et al., (2018)."

We added in the manuscript the following discussion in section 3.3:

"The TM5-FASST model developed by van Dingenen et al. (2018) has been validated against concentration estimates derived from the WHO database and satellite-based measurements (van Donkelaar et al., 2010, 2014). General good agreement is found between the PM2.5 concentrations modeled by TM5-FASST and the measured ones reported in the WHO database for Europe (within 20% deviation), North America (within 5% deviation) and partly China due to the higher accuracy of the measurements. The comparison for Latin America and Africa is much less robust (40-60% deviation from the 1:1 line) and the scatter possibly highlights a non-optimal modeling of specific sources relevant for these regions by TM5-FASST (e.g. large scale biomass burning) by the TM5-FASST model. Similar results are also found comparing regional averages of urban stations from WHO against the FASST population weighted average of grid cells. The TM5-FASST modeled PM2.5 concentrations have been 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 shows consistent results with the ground based measurements comparison (e.g. good agreement for EU and USA within 10% deviation, while lower agreement for developing and emerging countries)."

11.31: Cohen et al. (2017) also report a range for the total estimated global premature deaths from ambient PM2.5 - which should be repeated here. This is interesting to consider, as the source of the uncertainty in the Cohen paper is from uncertainty in the concentration-response relationships (IERs), not from uncertainties in the exposure estimates that may be owing to uncertainties (in part) from emissions. However, the range of values cited here (+/- 1.1 million) indicates that this uncertainty associated with emissions estimates is a factor, which hasn't been much considered previously. This is an import results of the present work which I believe could be highlighted more (i.e. by comparing the magnitude of the emissions-driven uncertainties to the magnitude of other types of uncertainties considered in different studies). Quantitative summary of this (similar to the final sentence of the manuscript) would be nice to see in the abstract as well.

The following sentences have been added:
"In our work we only evaluate how the uncertainty of emission inventories influences the health impact estimates focusing on the interregional aspects (we do not evaluate effects of misallocation of sources within regions) and not all the other sources of uncertainties often included in literature studies, 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, submitted). Solazzo et al. find in their analysis over the European countries a mean number of PDs due to exposure to PM2.5 and ozone of approximately 370 thousands (inter-quantile range between 260 and 415 thousand). Moreover, they estimate that a reduction in the uncertainty of the modelled ozone by 61% - 80% (depending on the aggregation metric used) and by 46% for PM2.5, produces a reduction in the uncertainty in premature mortality and crop loss of more than 60%. However, we show that the often neglected emission inventories' uncertainty provides a range of premature deaths of ±1.1 million at the global scale, which is in the same order of magnitude of the uncertainty of air quality models and concentration-response functions (Cohen et al., 2017)."

Minor:

2.10-2.14: What fraction of secondary PM2.5 long-range transport is owing to transport of the gas-phase precursors vs the transport of the secondarily formed PM2.5 itself?

To answer this question, which was not explicitly studied in this publication, but included in the model calculations, one has to consider 4 aspects: chemical lifetime of the precursor gases, atmospheric transport, transport distance, and removal processes of both precursors and aerosols. Lifetimes of precursor gases range from hours (NH3), hours-to-days (NOx) and several days (SO2). A back-of-the-envelope calculation assuming a lifetime 0.1 hour and a wind speed of 1 m/s, would indicate a transport distance of ca 8 km, and clearly most of the precursor would be oxidized before leaving the ca. 100x100 km TM5 gridbox. On the other hand a lifetime of 7 days and a wind speed of 10 m/s would imply that this precursor could travel thousands of km before 2/3 of it would be oxidized.

We propose to include the following phrase:

"Although primary $PM_{2.5}$ (particulate matter with a diameter less than 2.5 μm) 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)."

2.27: Clarify here that this inventory, and the prescribed emissions for these experiments, pertain only to anthropogenic emissions.
Done

3.14: Can the authors comment on the validity of this assumption, as backed up by their own investigations or those in previous studies in the literature?

We assume that individual sector contributions add up linearly to total PM2.5. The figure below shows the very good agreement between total PM2.5 concentrations and the sum of sector-specific concentrations for each receptor region. Additional details can be found in van Dingenen et al. (submitted, 2018).

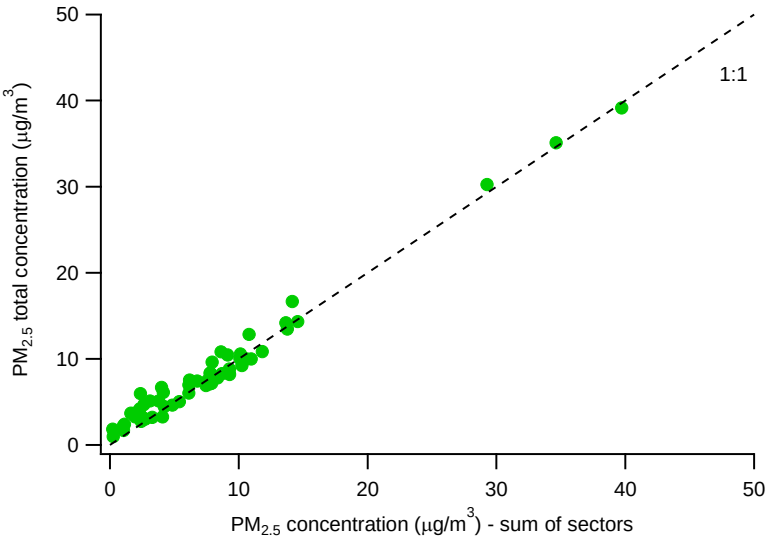


Figure 1 – Comparison between the total modeled PM2.5 concentration and the sum of the sectors.

3.23: The source-receptor modeling was based around a single year that didn't alight with the year of the emissions considered. To what extent does this misalignment potentially impact results? Or to what extent is the meteorology in this particular year representative of a climatological average? I guess I'm just wondering if the authors have checked if 2001 was for any reason particularly extreme with regards to temperature, precipitation, transport, or sources of natural PM2.5 such as biomass burning?

Anthropogenic emissions in general do not greatly vary from year to year and a large co-variation with specific meteorological conditions is considered not very important. Indeed such co-variation can be an important issue for natural emission. Biomass burning, sea salt and mineral dust are dependent among other factors on meteorological conditions. For the natural emissions of dust, sea salt and biomass burning we included the recommended gridded datasets made for AEROCOM phase 1 for the year 2000- indeed not aligning with the meteorological year 2001 used in the TM5 CTM in this study. There are three considerations of relevance for this paper. If the goal is to have the most accurate estimate of natural emissions, the use of a community endorsed dataset is probably a safe one, since model generated emissions would carry their own uncertainties. While, especially for mineral dust and biomass burning, there are

large inter-annual variations, these variations- at least at larger scales- are probably smaller than the emission uncertainties themselves. And finally, the use of 'constant' emission, allows factoring out their uncertainties, since the scope of the work is considering mostly anthropogenic emissions.

3.27: To what extent does not including anthropogenic SOA influence conclusions about the role of different sectors?

Unfortunately we do not have estimates of the contribution of anthropogenic SOA, as the gas phase chemical degradation scheme didn't include emissions of the relevant precursor gases. The importance of anthropogenic SOA ranges regionally widely, as demonstrated by a recent study by Farina et al. (2010) indicating a global source of 1.6 Tg, or ca. 5.5 % of the overall SOA formation. The relatively importance, however, may dependent regionally, and is deemed higher in regions with less VOC emission controls. Overall, we feel that the uncertainty stemming from our knowledge in SOA formation is higher than the omission of anthropogenic SOA. We would also like to mention that the development of the volatility-based SOA formation approach, means that the boundaries between 'primary' and 'secondary' SOA are disappearing, making it difficult to attribute organic aerosol to either primary, secondary (or natural-anthropogenic), as they strongly interact. Nevertheless, we speculate that the inclusion of SOA would possibly lead to a higher role of the transboundary pollution mainly for those sectors emitting PM and VOCs (e.g. residential, and to some extent transport and industry).

Therefore we added the following sentences to the manuscript:

"The importance of anthropogenic SOA ranges regionally widely, as demonstrated by a recent study by Farina et al. (2010) indicating a global source of 1.6 Tg, or ca. 5.5 % of the overall SOA formation. The relatively importance, however, may dependent regionally, and is deemed higher in regions with less VOC emission controls. We speculate that the inclusion of SOA would possibly lead to a higher role of the transboundary pollution mainly for those sectors emitting PM and VOCs (e.g. residential, and to some extent transport and industry)."

4.4: It seems that rather than aggregation the authors could consider some metrics that are normalized with regards to the country size or population.

In this work we decided to aggregate the 56 FASST regions into 10 world regions based on the geographical location and as much as possible the degree of development and emissions (of course African countries do not have all the same degree of development etc., but for us it made more sense to group them together instead of putting some African countries with Russian or Latin America countries because of similar size or population). Moreover, the population information is taken into account when calculating the population weighted PM concentrations for the aggregated regions. Population data are presented in Table S2. However, in order to make mortality results more comparable among countries we included the normalized PD metric in Table 4.

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

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

4.20: Here and elsewhere the Janssens-Maenhout (2017, submitted) paper is cited, although it's hard to evaluate what information is contained therein.

We clarified line 20 at page 4 as following:

"Uncertainty values of the activity data by sector and country are obtained from Table 2 of Janssens-Maenhout et al. (2017, submitted) and Olivier et al. (2016). Using this approach, the uncertainty in the global total anthropogenic CO2 emissions is estimated to range from -9% to +9% (95% confidence interval), which is the result from larger uncertainties of about +/-15% for non-Annex I countries, whereas uncertainties of less than +/-5% are obtained for the 24OECD90 countries for the time series from 1990 (Olivier et al, 2016) reported to UNFCCC."

About Figure 1: It's not clear – are the % contributions to the average PM2.5 in each region, or to the population-weighted average PM2.5 in each region?

Percentages represent the contributions to the population-weighted average PM2.5 in each region. Figure caption has been modified accordingly.

7.34: I think the impacts of the residential sector on indoor air quality are well known and have been documented in many previous studies that could be cited.

The following papers are now cited in the text:

The residential sector is one of the most significant sources of PM all over the world, potentially also affecting indoor air quality (e.g. Ezzati, 2008; Lim et al., 2013; Chafe et al., 2014).

7.39: Similarly, the role of the agricultural sector or NH3 in particularly has been noted in several previous and recent studies. The authors continue to cite only Maas and Grennfelt, 2016, despite the broader literature available for comparison.

The following papers are now cited in the text: Pozzer et al. (2017), Tsimpidi et al. (2007), Zhang et al. (2008), Backes et al. (2016) and Erisman et al. (2004).

"Interestingly, the agricultural sector is 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)."

8.9-11: Can the authors explain why primary emissions play such a large role in the uncertainty analysis, compared to their contribution to absolute PM2.5 concentration?

Primary PM emissions are mainly emitted from the residential, transport and to a smaller extent industrial sectors and they are characterized by the largest values of uncertainty. With the exception of the countries where the contribution of the power generation sector is relevant (which mainly leads to the formation of secondary inorganic components of PM), the other countries are dominated by the remaining sources highly emitting primary PM which are therefore strongly contributing to the final PM2.5 concentration.

9.20: Given that this work doesn't include anthropogenic SOA, what is the role of NMVOCs in PM2.5 formation? I guess I was just surprised to see these mentioned here.

In section 3.4.2 we rank the sector specific contribution to emission uncertainties for each of the pollutant provided by the HTAP_v2.2 inventory. As the Reviewer pointed out, TM5-FASST does not model SOA formation from anthropogenic VOCs. However, in order to provide a complete overview on the sector contribution to emission inventories' uncertainty we reported this information also for anthropogenic NMVOCs. This analysis wants to assess the emission inventories uncertainty and it is independent from the model or source-receptor model we use to estimate PM concentrations.

11.34: What is the "urban increment subgrid adjustment"?

As extensively discussed in van Dingenen et al. (submitted, 2018), to better represent the actual mean population exposure within a grid cell some adjustments are included in the TM5-FASST tool. A first adjustment is performed based on the assumption that the spatial distribution of primary emitted PM2.5 correlates with population density; then information on urban and rural population grids is included and further assumptions are also applied (e.g. primary PM2.5 from the residential and the surface transport sectors are contributing to the local (urban) increment, while other aerosol precursor components and other sectors are assumed to be homogenously distributed over the grid cell). Secondary PM2.5 is formed over longer time scales and therefore more homogeneously distributed at the regional scale.

The following sentence has been therefore added into the manuscript:

"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. (submitted, 2018)), the inclusion or not of natural components, the impact threshold value used, and RR functions."

11.33 - 35: I strongly agree that these factors are critical towards making these comparisons, as are sources of information such as population densities and baseline mortality rates. For those precise reasons, the authors should provide details on these aspects as used in their study, as have been provided in the cited works, in order to make such comparisons possible and meaningful.

The manuscript has been rephrased as following:

"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. (submitted, 2018)), the inclusion or not of natural components, the impact threshold value used, and RR functions. In this study we used pollution the population weighted PM2.5 concentration (excluding natural components) at 1x1 degree resolution as metric for estimating health effects due to air, with a threshold value of 5.8 μg/m3, no urban increment adjustment, and relative risk functions accordingly with Burnett et al. (2014).

12.10-12: What it is about these regions that given them such relatively large extra-regional contributions to PM2.5 health impacts?

As shown in Fig.3, Hungary, Czech Republic, Mongolia and the Gulf region are characterized by a very high fraction of transported pollution and therefore the corresponding extra-regional contribution to the health impacts is high.

The manuscript has been rephrased as following:

"However, there are marked exceptions, such as the Gulf region, Hungary, Czech Republic, Mongolia, etc., where the extra-regional and within-region contributions to mortality are at least comparable. In fact Hungary and Czech Republic are strongly influenced by polluted regions in Poland (mainly); likewise Mongolia is suffering from the vicinity of source 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)."

Editorial:

2.23: "not to the least" change to "not the least"
Done

2.35: "at sector" change to "at the sector"
Done

2.36: "on the potential" change to "of the potential"
Done

3.19: Some of this sentence seems to be missing.
The sentence has been corrected as following:
"In order to calculate $PM_{2.5}$ concentrations from the HTAP_v2.2 emissions, we deployed the gridded TM5-FASST version 1.4b (Van Dingenen et al., 2017, in preparation)."

4.16: "as following" change to "as follows"
Done

6.16: "across" change to "an across"
Done

8.23: "Europe the" change to "Europe, the"

Done

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The authors are grateful to Referee#2 for the helpful comments that helped improve the manuscript. Due to the strict link between this publication and the work recently submitted by van Dingenen et al. (submitted, 2018) about the TM5-FASST methodology, we offered the possibility to the Editor and the Reviewer to access the work of van Dingenen et al. (submitted, 2018) although not yet published in ACPD. We feel that we have been able to address all concerns, as outlined below. Thanks to the Reviewer's comments, we also realized that some methodological aspects of the TM5-FASST tool could have been further developed also in the publication of van Dingenen et al. (submitted, 2018). Therefore, discussions on the comparison between PM2.5 modeled concentrations vs. the measured ones, as well as further details about the extension of the "perturbation approach" to the attribution of sectors and sources will be included in the review phase of the paper by van Dingenen et al. (submitted, 2018). We feel that we have been able to address all concerns, as outlined below.

Anonymous Referee #2

The authors coupled the HTAPv2.2 global air pollutant emission inventory with the global source receptor model TM5-FASST to evaluate the relative contribution of the major anthropogenic emission sources to air quality and health in 2010. They focused on PM2.5 due to its negative impacts on human health. The objective of this paper is to evaluate the emissions uncertainties at sector and regional levels, and their propagation in modeled PM2.5 concentrations and associated impacts on health. Although the authors state that they have two objectives, I do not quite understand the difference between the two. I find that what the paper is trying to do is important but there are some major problems that need to be addressed before this can be published in ACP.

First, if the objective is to understand the health impacts of PM2.5, I believe that the authors need to make sure that their model simulations match with the observations. I do not find the existing comparison in the paper (p. 7, l. 21-28) very convincing. The authors could have at least compared with the recent WHO database of annual PM2.5 concentrations at various cities (http://www.who.int/phe/health_topics/outdoorair/databases/cities/en/). For the US, there is much better database that could be used (https://www.epa.gov/outdoor-air-quality-data/interactive-mapair- quality-monitors). The authors seem to allude that it is ok to not include the natural emissions but I disagree and think that the natural emissions need to be included in the model.

We acknowledge the suggestion of the Reviewer about the comparison of the PM2.5 concentrations estimated by TM5-FASST with other databases in addition to what already provided in our manuscript.
The EPA air quality statistics for USA for the year 2010 (https://www.epa.gov/outdoor-air-quality-data/air-quality-statistics-report) report an annual concentration of PM2.5 of 12.0 ug/m3 which is higher compared to our estimate (7.8 ug/m3) because measured PM2.5 concentrations include all sources of PM (e.g. large scale biomass burning and SOA from anthropogenic sources which are not accounted in our study).
However, the TM5-FASST model developed by van Dingenen et al. (submitted) has been validated against concentration estimates derived from the WHO database and satetellite-based

measurements (excluding dust and sea salt). We report below some details about these comparisons which will be included in the submitted manuscript by van Dingenen et al.
Figure 1 shows the comparison between the PM2.5 concentrations modeled by TM5-FASST and the measured ones reported in the WHO database for different world regions (i.g. EUR=Europe, NAM=North America, China, S-ASIA=Southern Asia, LAM=Latin America, AFR=Africa). This includes measurement points as well as PM2.5 concentration estimates based on a fraction of PM10 measurements (e.g. almost all points for the comparison in China are based on this second method). Quite good agreement is observed for Europe, North America and partly China where measurements have been performed over longer time compared to developing countries and they are based on quite consolidated methods. The comparison for Latin America and Africa is much less robust and the scatter possibly highlights a non-optimal modeling of large scale biomass burning for the TM5-FASST model. Figure 2 reports the comparison of WHO regional average of urban stations against the FASST population weighted average of grid cells. Similarly to the findings of Figure 1, the comparison for industrialized countries is very good, while for other developing regions the agreement is less satisfactory both due to less accurate measurements (e.g. reported by WHO) and lower quality modeling of specific sources by TM5-FASST (e.g. large scale biomass burning). Figure 3 shows the comparison between the population-weighted country-averaged PM2.5 concentrations from the WHO database on outdoor air pollution (years 2011, 2014 and 2016) and from TM5-FASST for the year 2010 (using the ECLIPSE/HTAP emission inventory).

TM5-FASST modeled PM2.5 concentrations have been also validated against satellite products (see Figure 4) 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).

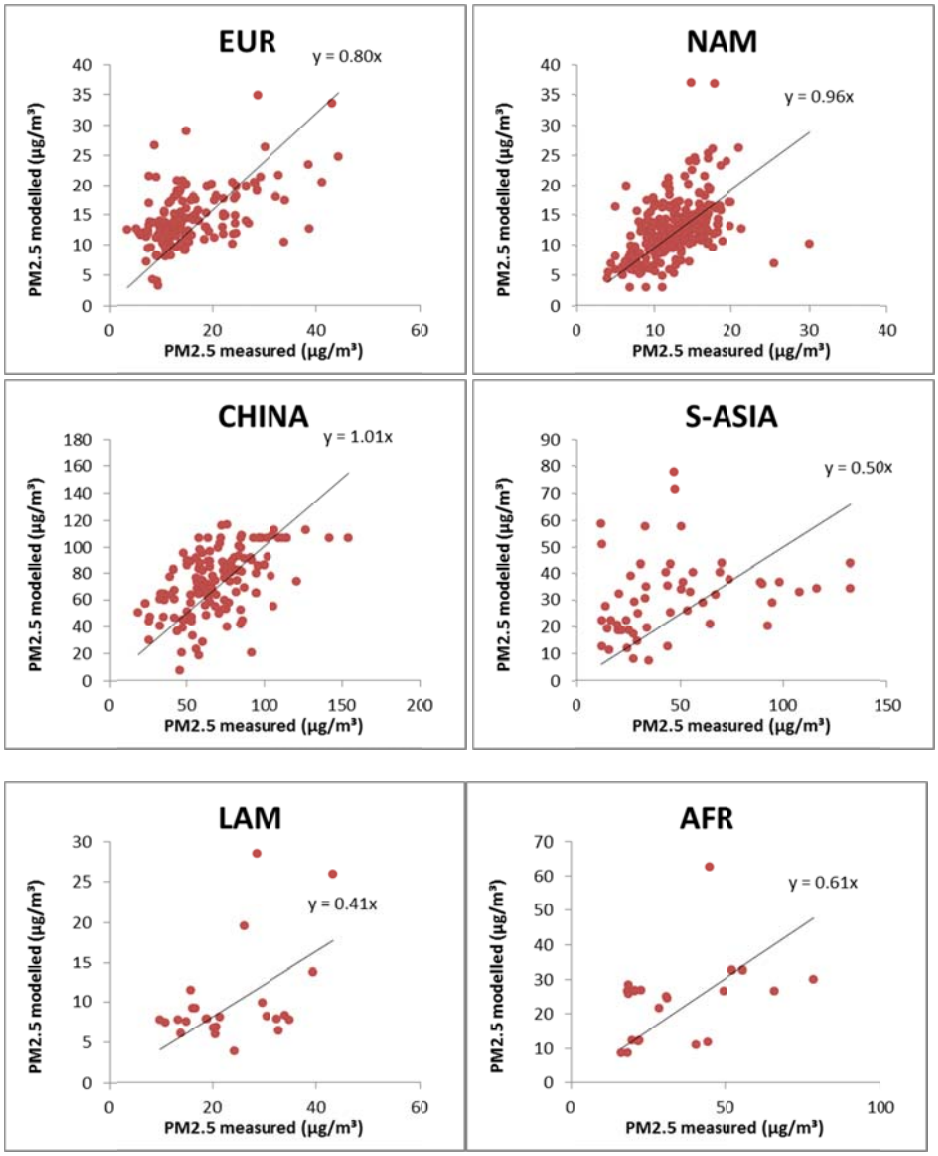

Figure 1 - TM5-FASST grid-cell mean (with urban increment parameterisation) versus individual monitoring stations (WHO consolidated database, including both measured and estimated PM2.5).

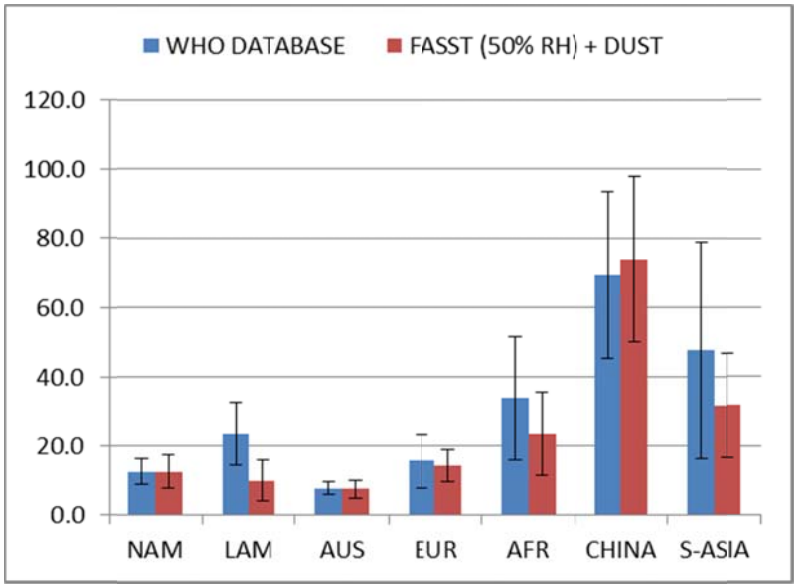

Figure 2 - WHO regional average of urban stations (+/- 1 stdev) and FASST population weighted average of grid cells.

Figure 3 shows the population-weighted country-averaged PM2.5 concentrations from the WHO database on outdoor air pollution (years 2011, 2014 and 2016) and from TM5-FASST for the year 2010 (using the ECLIPSE/HTAP emission inventory). WHO data are compiled from mostly urban or urban background monitoring stations, and include PM2.5 estimates based on measured PM10 when PM2.5 measurements are not available. FASST PM2.5 represents dry PM2.5 while WHO monitoring data include residual water from equilibration between 35 – 50%RH. TM5-FASST includes a generic fixed dust and sea-salt field, as well as an urban increment correction on primary anthropogenic PM2.5 where appropriate. TM5-FASST averages include all countries grid-cells, WHO includes only values form monitoring station locations and is therefore biased towards higher values compared to TM5-FASST.

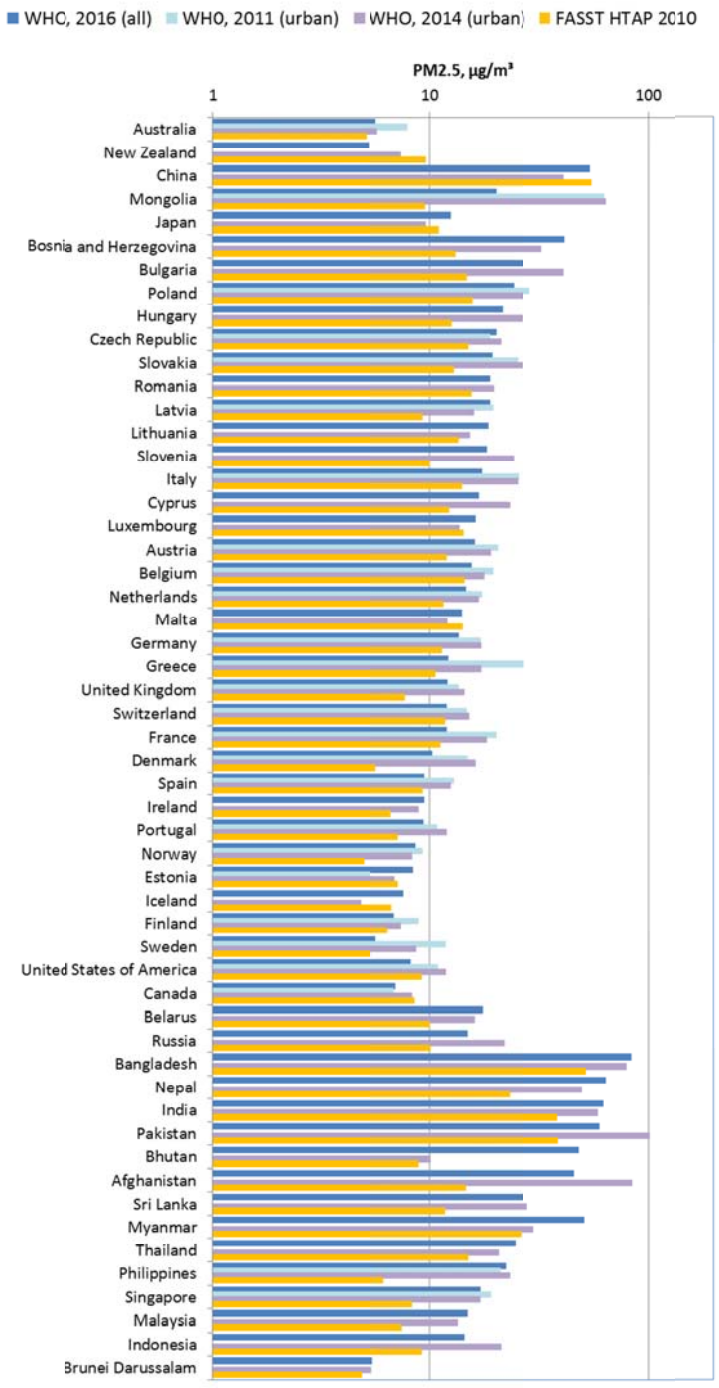

Figure 3 - Population-weighted country-averaged PM2.5 concentrations from the WHO database on outdoor air pollution (years 2011, 2014 and 2016) and from TM5-FASST for the year 2010 (using the ECLIPSE/HTAP emission inventory).

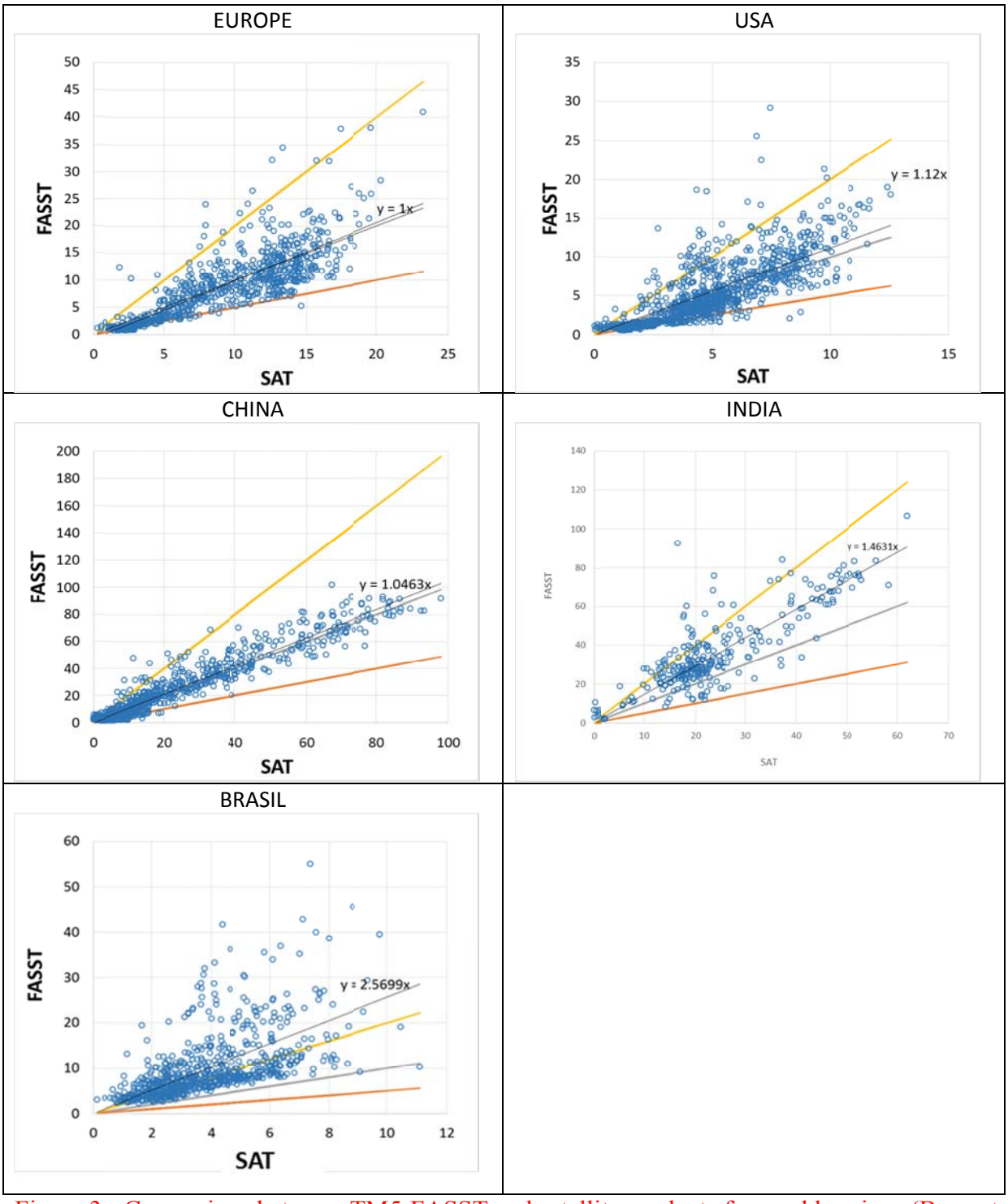

Figure 3 - Comparison between TM5-FASST and satellite products for world regions (Boys et al., 2014; van Donkelaar et al., 2010, 2014).

Therefore we added in the manuscript the following discussion in section 3.3:

"The TM5-FASST model developed by van Dingenen et al. (2018) has been validated against concentration estimates derived from the WHO database and satellite-based measurements (van Donkelaar et al., 2010, 2014). General good agreement is found between the PM2.5 concentrations modeled by TM5-FASST and the measured ones reported in the WHO database for Europe (within 20% deviation), North America (within 5% deviation) and partly China due to the higher accuracy of the measurements. The comparison for Latin America and Africa is much less robust (40-60% deviation from the 1:1 line) and the scatter possibly highlights a non-optimal modeling of specific sources relevant for these regions by TM5-FASST (e.g. large scale biomass burning) by the TM5-FASST model. Similar results are also found comparing regional averages of urban stations from WHO against the FASST population weighted average of grid cells. The TM5-FASST modeled PM2.5 concentrations have been 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 shows consistent results with the ground based measurements comparison (e.g. good agreement for EU and USA within 10% deviation, while lower agreement for developing and emerging countries)."

Second, I find that the emissions uncertainty estimate seems a little simplistic to only assess within the HTAP inventory, considering the existing differences among various inventories. Also, if the RCP emissions for the year 2000 are used as a baseline, to me it makes more sense to use RCP 2010 in their analysis, rather than switching to HTAP v2.2. Or if the HTAP is to be used, the uncertainty analysis should include the differences in estimates between RCP 2010 and HTAP v2.2. Also, it might be a good idea to compare with some other estimates in existing studies that have estimated emissions uncertainties for certain countries.

We would like to stress that the aim of this work is not to compare different emission inventories since this has already been done in other publications (specifically regarding the HTAP_v2 inventory, e.g. Janssens-Maenhout et al., 2015, Crippa et al., 2016), but we aim at addressing 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. There are several reasons to use HTAP_v2.2 and not e.g. the RCP2000 as the basis for our assessment of emission propagation. 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, we feel that it is now superseded by the more accurate HTAP_v2.2. Given our focus on regional (and not so much gridded) results, we feel that this choice is justified.

Therefore we added the following explanation in Section 2.1 of the manuscript:

"The aim of this work is to 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 e.g. 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 day much more accurate in particular given the focus on regional (and not so much gridded) emission analysis of our work."

Differently from CO2 for which emission uncertainties are much better know, literature studies dealing with uncertainty of emission inventories of all air pollutants show a lack of information on the corresponding uncertainties (while intercomparisons among different inventories are often shown). In addition, literature studies often make use of region- and sector-specific emission inventories and they do not provide a global view on all pollutants, sectors and regions (Hoesly et al., 2017).

However, we took into account the Reviewer's comment including some references with literature studies on emission inventory uncertainties.

Page 9, line 16: Smith et al. (2011) report a range of regional uncertainty for SO2 up to 30% while our estimates are slightly higher (up to 50%).

Page 9, line 24: "Among all air pollutants, represent one of the most uncertain pollutant due to very different combustion conditions, different fuel qualities and lack of control measures (Klimont et al., 2017)."

Third, I also find it problematic that important details and assumptions of TM5-FASST methodology are described in the paper that is still under preparation. I am assuming that the ΔPMref and ΔEref in Eq. 1 refer to the difference between the TM5-FASST simulation results for PM2.5 (and also PM10 as well?) using the RCP baseline and the perturbation (-20%) and the emissions themselves, respectively. However, I find it troublesome that these stay constant when the emissions change for all regions and sectors. We know that PM2.5 formation is a non-linear process and I do not believe it would work in a linear form for every region for every sector. If it does, maybe that is because simulation uses too coarse of a resolution and the result does not seem realistic.

The paper by van Dingenen et al. (submitted) has now been submitted to ACP. It contains a detailed description on the methodology and documents the validity of the linearity assumption for PM2.5 (the simulations were done only for PM2.5 and not PM10). Unfortunately anthropogenic SOA is not explicitly modeled in TM5 but treated as a pseudo-emission. In the manuscript we clarified the concept of dE and dPM as following:

"The reduced-form model TM5-FASST is computing the concentration resulting from an arbitrary emission scenario E_i using a perturbation approach, i.e. the difference between E_i and $E_{i,ref}$ *(dE)* is considered as a perturbation on E_{ref} and the resulting concentration is evaluated as a perturbation *dPM* on the reference concentration."

Also, it seems problematic that no explicit treatment of anthropogenic SOA is considered.

Unfortunately we do not have estimates of the contribution of anthropogenic SOA, as the gas phase chemical degradation scheme didn't include emissions of the relevant precursor gases. The importance of anthropogenic SOA ranges regionally widely, as demonstrated by a recent study by Farina et al. (2010) indicating a global source of 1.6 Tg, or ca. 5.5 % of the overall SOA formation. The relatively importance, however, may dependent regionally, and is deemed higher in regions with less VOC emission controls. Overall, we feel that the uncertainty stemming from our knowledge in SOA formation is higher than the omission of anthropogenic SOA. We would also like to mention that the development of the volatility-based SOA formation approach, means that the boundaries between 'primary' and 'secondary' SOA are disappearing, making it difficult to attribute organic aerosol to either primary, secondary (or natural-anthropogenic), as they strongly interact. Nevertheless, we speculate that the inclusion of SOA would possibly lead to a higher role of the transboundary pollution mainly for those sectors emitting PM and VOCs (e.g. residential, and to some extent transport and industry).

Therefore we added the following sentences to the manuscript:

"The importance of anthropogenic SOA ranges regionally widely, as demonstrated by a recent study by Farina et al. (2010) indicating a global source of 1.6 Tg, or ca. 5.5 % of the overall SOA formation. The relatively importance, however, may dependent regionally, and is deemed higher in regions with less VOC emission controls. We speculate that the inclusion of SOA would possibly lead to a higher role of the transboundary pollution mainly for those sectors emitting PM and VOCs (e.g. residential, and to some extent transport and industry)."

Is it correct that TM5-FASST simulations were run for each sector separately and also for all the sectors combined? That is how it looks like from Figure 4. If so, can the authors confirm that the sum of concentrations from each of the sectors run separately are similar to the values when the simulation was done including all the sector emissions together? It would be a nice test to check the linearity in the model. If the simulations were done in this way, then what was the reason equation 1 had to be used? The authors could have easily calculated the impact of each sector using these simulations instead?

In general, the reduced-form model TM5-FASST is computing the concentration resulting from an arbitrary emission scenario E_s using a perturbation approach, i.e. the difference between E_s and E_{ref} (dE_s) is considered as a perturbation on E_{ref} and the resulting concentration is evaluated as a perturbation dPM on the reference concentration, hence:

$$PM(E_s) = PM(E_{ref} + dE_s) = PM_{ref} + dPM = PM_{ref} + SRC \cdot dE_s \quad \text{(a)}$$

Where $dE_s = E_s - E_{ref}$ and E_{ref} is the RCP reference scenario from which the SRC have been computed.

The contribution of a single sector j is calculated as the difference between the concentration including all sectors, and the concentration from the emissions excluding the single sector j

$PM\ (E_{s,j}) = PM(E_s) - PM(E_s - E_{s,j}) = SRC \cdot [dE_s - d(E_s - E_{s,j})] = SRC \cdot E_{s,j}$

If the linearity holds, the sum of $PM(E_{s,j})$ over all sectors j should be equal to $PM(E_s)$, or:

$$\sum_j PM(E_{s,j}) = PM_{ref} + SRC \cdot (E_s - E_{ref})$$

In Figure 5 we compare both sides of the equation to demonstrate that indeed the linearity assumption holds sufficiently well.

A caveat of TM5-FASST is that no sector-specific SRC have been computed (except for international shipping which was evaluated separately), and consequently our single sector analysis implicitly assumes that the spatial distribution of pollutant emissions at the resolution considered here (1°x1°) is similar for all sectors within each source region. Taking into account that

(1) the spatial distribution of primary anthropogenic emissions is commonly generated using population density as the major proxy (except for large scale biomass burning) – e.g. domestic burning, transport, industry
(2) in many cases, the emission of secondary pollutant precursors is dominated by a single sector (e.g. NH_3 mainly from agriculture, NO_x mainly from transport, SO_2 mainly form energy production)

we deem that the spatial distribution of the individual sectors can be estimated sufficiently accurately for the present analysis, as shown in Figure 4 which has been obtained from the 'total' SRC, applied on single-sector emissions. A similar approach has been recently implemented by Liang et al. (2018) based on the HTAP2 source receptors.

The TM5-FASST runs were performed for different scenarios, comparing the reference HTAP_v2.2 emissions with a scenario where emissions from one single sector were subtracted from the total emissions. Then comparing the reference case and each scenario (REF-$sector_i$), the contribution of each sector to PM2.5 concentrations is estimated. This approach is based on the assumption that the individual sector contributions add up linearly to total PM2.5, as mentioned in the paper. The paper by Van Dingenen et al. describing the whole TM5-FASST methodology has just been submitted to ACP (van Dingenen et al., submitted) Equation 1 represents the basis of the TM5-FASST method, since it describes how a variation in the emissions (delta emissions) determines a delta in PM2.5 based on the source receptor relationships.

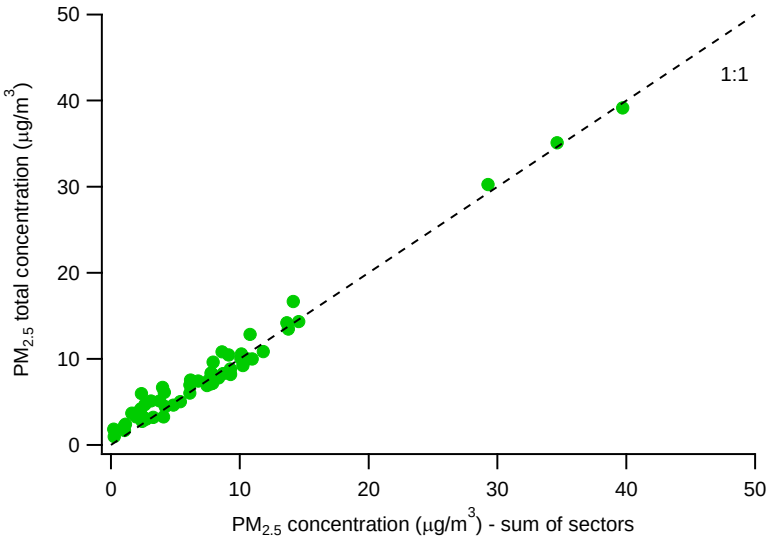

Figure 5 – Comparison between the total modeled PM2.5 concentration and the sum of the sectors.

The following discussion on how to apply the "perturbation approach" on the sector and source attribution will be also included in the paper by van Dingenen et al. (submitted, 2018):

Equation (2) expresses the 'perturbation' approach applied in the linearized TM5-FASST model, i.e. an arbitrary emission scenario is evaluated as a deviation from the base emission scenario, and the resulting pollutant concentration is obtained as the sum of the base concentration and a delta term, the latter proportional to the emission deviation from the base case (Figure 1).

A particular application of TM5-FASST is the attribution of the (anthropogenic) pollutant concentration to individual source regions or sectors. Due to the fixed contribution of the base concentration which does not contain information on the originating sources, Eq. (2) is not immediately suitable for such an analysis. Instead, we calculate for each individual source the contributing part by first evaluating all sources together ('total' simulation'), and subsequently subtracting the individual source emissions (E_s) from the total, evaluating the resulting pollutant concentration (C_{minus_s}), and making the difference with the 'total simulation' to obtain the single source contribution (C_s).

$$C_{j,tot}(y) = C_{j,base}(y) + \sum_{n_x}\sum_{n_i} A_{ij}[x,y] \cdot \left[E_{i,tot}(x) - E_{i,base}(x)\right] \quad (2)$$

$$C_{j,minus\_s}(y) = C_{j,base}(y) + \sum_{n_x}\sum_{n_i} A_{ij}[x,y] \cdot \left[E_{i,tot}(x) - E_{i,s}(x) - E_{i,base}(x)\right] \quad (4)$$

$$C^{*}_{j,s}(y) = C_{j,tot}(y) - C_{j,minus\_s}(y) = \sum_{n_x}\sum_{n_i} A_{ij}[x,y] \cdot E_{i,s}(x) \quad (5)$$

We can now reconstruct $C^*_{j,tot}$ as the sum of the individual source contributions:

$$C^*_{j,tot}(y) = \sum_{n_s} C^*_{j,s}(y) \quad (6)$$

$C^*_{j,tot}(y)$ is equivalent to $C_{j,tot}(y)$ in Eq. 2 only if

$C_{j,base}(y) = \sum_{n_x} \sum_{n_i} A_{ij}[x,y] \cdot E_{i,base}(x)$, in other words if the emission-concentration relation is perfectly linear and passes through the origin.

In reality there is some degree of non-linearity in most emission-pollutant relation as illustrated in Figs. 3 and 6. Figure A shows for each of the FASST regions the total $PM_{2.5}$ concentration obtained by Eq. 6 versus the TM5 base simulation result, illustrating the non-linearity error resulting from the application of Eq. 6. For 43 out of 56 regions, the deviation from the base simulation is less than 30%, only 3 regions (former Soviet Union, New Zealand and Pacific) deviate more than 50% from the TM5 model result. Consistency with the 'perturbation approach' is restored by simply rescaling the individual source contributions:

$$C_{j,s}(y) = \frac{C_{j,tot}(y)}{C^*_{j,tot}(y)} \, C^*_{s,j}(y) \quad (7)$$

This approach is valid for evaluating the attribution by sector as well as by source region.

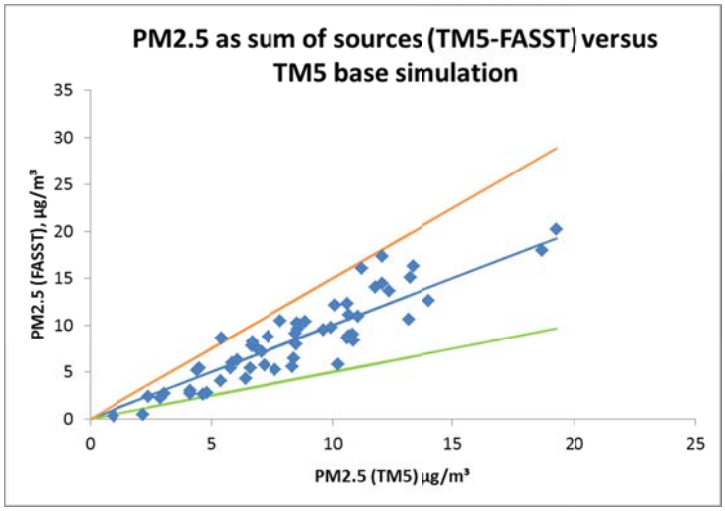

Figure A: Scatter plot of regionally averaged PM2.5 concentration (including all anthropogenic components) obtained as the sum of individual source region contribution by linear scaling of their respective emissions with TM5-FASST source-receptor coefficients (Eq. 6), versus the regional average obtained by the full TM5 model.

I have a hard time understanding the sentence on p. 9 l. 5-8. How do the authors determine the relative contribution to total emission inventory uncertainty? Are the authors using the

uncertainty for a specific sector over the total uncertainty for a specific pollutant as the "average sector relative contribution to total emission inventory"? If so, this does not necessarily take the magnitude of emissions into account and so maybe just looking at this value and deciding which sector to focus on might be a little too simplistic?

As discussed in Section 2.3, "uncertainties have been estimated for each emission sector for every country/region and pollutant. Then an overall uncertainty has been estimated using equation 5 (shown below) from the EMEP/EEA, 2013 Guidebook and which accounts for the weighted contribution of each sector to the overall uncertainty. Then the contribution of each sector to the overall uncertainty is given by the weight of each term of the equation compared to the others, so it does not correspond to the "average sector relative contribution to total emission inventory".

We rephrased as following:

"The complete overview of allTM5-FASST regions is provided in Fig. S2, where the share of each 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 uncertainty of each pollutant in each region, is reported."

Are the upper and the lower boundaries of PM2.5 concentrations (Table 2 and Figure 5) calculated based on the linear relationship between emissions per region? In other words, are they simply calculated from emissions, rather than running the simulation again in a chemical transport model?

To calculate the upper and lower boundaries of PM2.5 concentrations we used the TM5-FASST model and so they are based on the linear relationship between emissions per region. However, new emission datasets including the upper and lower range of uncertainty have been given as input for new TM5-FASST runs which gave us the upper and lower range of PM2.5 concentrations.

We added a sentence at the end of paragraph 2.3 to clarify our approach:

"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*2 for the uncertainty analysis)."

Minor comments:

1. I would like to see a figure that shows the 10 aggregated receptor regions, as it is unclear, for example, what China+ region includes. Does it just include Mongolia? Or also Korea and Japan?

   Table S2 of the Supplementary material already includes this information for all aggregated regions. China+ includes China and Mongolia+North Korea. We do not aim at having another Figure in the supplementary material about the regions aggregation, in order to avoid

repeating information already provided in a Table and to avoid misunderstandings with the map about the 56 TM5-FASST regions used for the model runs.

2. Why are some European countries lumped together in Figure 2 (Austria and Slovenia, for example), whereas others are not?

   The following explanation has been added in the Supplementary Material (S1) to explain the TM5-FASST regions aggregation.

"The 56 TM5-FASST regions were chosen to obtain an optimal match with integrated assessment models such as IMAGE (Eickhout et al., 2004; van Vuuren et al., 2007), MESSAGE (Riahi et al., 2007), GAINS (Höglund-Isaksson and Mechler, 2005) as well as the POLES model (Russ et al., 2007; Van Aardenne et al., 2007). The grouping of small countries was motivated by (a) finding a compromise between spatial resolution and computational effort required to obtain the set of source-receptor matrices for TM5-FASST and (b) avoiding inaccurate mapping of small individual countries that are represented by only a few 1°x1° grid cells.
Most European countries are defined as individual source regions, except for the smallest countries, which have been aggregated."

3. Why are there more countries in Figure 3 than in Figure 1?

   Figure 1 represents the global view using the 10 aggregated world regions, while figure 3 shows a disaggregated view making use of the original 56 TM5-FASST regions. The reason behind the aggregation to 10 regions is explained at page 4 of the manuscript: "In order to make smaller regions (e.g. European countries) comparable with larger regions (like USA, China and India), in this work an aggregation procedure to 10 world regions (refer to Table S2) has been applied (China+, India+, SE Asia, North America, Europe, Oceania, Latin America, Africa, Russia and Middle East)."

p. 2. l. 30-34: The sentence is too long and difficult to understand. Please rephrase the sentence.

The sentence has been rephrased as following:

"The objective of this study is to evaluate the relevance of uncertainties in regional sectorial emission inventories (power generation, industry, ground transport, residential, agriculture and international shipping), and its propagation in modeled $PM_{2.5}$ concentrations and associated impacts on health. We also investigate the uncertainties in $PM_{2.5}$ from within the region to extra-regional contributions."

p. 2 l. 36-37: The authors state that a second objective of the analysis is to "inform local, regional hemispheric air quality policy makers on the potential impacts of less known emission sectors or regions" but they are focusing on the "6 major anthropogenic emission sectors (l. 6-7, p. 3)." What do they mean by "less known emission sectors" then?

Less known emission sectors (and less regulated ones in terms of emissions) are the residential and agricultural sectors, so the sentence has been rephrased as following:

"A second objective of this analysis is to evaluate the importance of emission uncertainties at sector and regional level on PM2.5, to better inform local, regional and hemispheric air quality policy makers on the potential impacts of sectors with larger uncertainties less known emission sectors (e.g. residential and agriculture) or regions (e.g. developing and emerging countries).

p. 3. l. 19-20. This sentence is not finished.

The sentence has been corrected as following:

"In order to calculate PM2.5 concentrations from the HTAP_v2.2 emissions, we use the native 1°x1° resolution source-receptor gridmaps obtained for TM5-FASST_v0 (Van Dingenen et al., 2018, submitted)".

p. 3. L. 22. Why was such a coarse resolution used, when HTAPv2.2 is much finer?

At the time of creating the TM5-FASST Source receptor relationships (ca. 2007-2010), 1x1 degree global resolution was still of unprecedented high resolution (given hundreds of simulations) and more common was resolutions around 2 to 3 degrees (T42). Only since recently more global models are running on 1x1 degree or somewhat finer, but it is still difficult to make 100s of SR calculations. The 0.1x0.1 HTAP_v2 resolution is employed only in full by regional model studies that used global model results as boundary conditions.

The following sentence has been added for clarity in the manuscript:

"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°x1° resolution emission spatial distribution from RCP year 2000 which was partly based EDGAR methodology and gridmaps."

p. 3 l. 30 relativey ->relatively
correction done

p. 7 l. 37-39 Perhaps a reference to Bauer et al. (2016) would be appropriate here.

Some changes have been made in that section, adding also more references:

"In order to understand the origin of global PM2.5 concentrations, we look at sector specific maps (Fig. 4). The power and industrial sectors are mainly contributing to PM concentrations in countries having emerging economies and fast development (e.g. Middle East, China and India), while the ground transport sector is a more important source of PM concentrations in industrialised countries (e.g. North America and Europe) and in developing Asian countries. The residential sector is one of the most significant sources of PM all over the world, potentially also affecting indoor air quality (Ezzati, 2008; Lim et al., 2013; Chafe et al., 2014)."

p. 11 l. 36. It is unclear to me where this value (7% for the global non accidental mortalities) is coming from. Can you clarify or cite the source?

We cited the source of our estimates as following:

"We also estimate that 7 % of the global non accidental mortalities from the Global Burden of Disease (http://vizhub.healthdata.org/gbd-compare; Forouzanfar et al. (2015)) are attributable to air pollution in 2010;"

p. 12 l. 10 such the Gulf -> such as the Gulf

correction done

p. 19 Table 2. How do you quantify the uncertainty for a certain pollutant for a region?

The methodology behind the uncertainty estimates for a certain pollutant and region is described in Sect. 2.3 of the manuscript and with the equations 3 and 4.
Table S3 of the Supplementary material also provides region- and pollutant- specific emission uncertainties.

Reference:

Bauer, S.E., K. Tsigaridis, and R.L. Miller, 2016: Significant atmospheric aerosol pollution caused by world food cultivation. Geophys. Res. Lett., 43, no. 10, 5394-5400, doi:10.1002/2016GL068354.

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

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Abstract

In this work we couple the ~~HTAPv2~~HTAP_v2.2 global air pollutant emission inventory with the global source receptor model TM5-FASST to evaluate the relative ~~contribution~~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 ~~proven~~well-documented cumulative negative effects on human health. We estimate that in 2010 ~~regional~~, depending on the region, annual averaged anthropogenic $PM_{2.5}$ concentrations varied between ca 1 and 40 $\mu g/m^3$ ~~depending on the region~~, with the highest concentrations observed in China and India, and lower concentrations in Europe and North America. The relative contribution of anthropogenic emission ~~source sectors~~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 ~~in order to improve~~ air quality~~,~~ improvements would require regional policies ~~should be implemented (e.g. in Europe)~~, in addition to local and urban scale measures, due to the transboundary features of PM pollution. ~~In addition, we~~We investigate emission inventory uncertainties and their propagation to $PM_{2.5}$ concentrations, in order to identify the most effective strategies to be implemented at sector and regional level to improve emission inventories knowledge and air quality. We show that the uncertainty of PM concentrations depends not only on the uncertainty of local emission inventories, but also on that of the surrounding regions. Finally, we propagate emission inventories uncertainty to PM concentrations and health impacts.

1 Introduction

Ambient particulate matter pollution ranks among the top five risk factors globally for loss of healthy life years and is the largest environmental risk factor ~~(Lim et al., 2012~~(Lim et al., 2013;Anderson et al., 2012;Anenberg et al., 2012~~);~~Cohen et al., 2017). The world health organization ~~(WHO, 2016)~~(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 Transport of Air Pollution (CLRTAP). At city/local level, several studies have been developed to assess the contribution of sector specific emissions to PM2.5 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 kilometers, crossing national borders, oceans and even continents (HTAP, part A, 2010). ~~Therefore local~~Local, regional and international coordination is therefore needed to define air pollution policies to improve globally 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 aiming to identify promising mitigation measures to reduce background pollution levels and its contribution to pollution in rural as well as urban regions. Although primary $PM_{2.5}$ (particulate matter with a diameter less than 2.5 ~~μm)~~μm) 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~~ of gaseous precursors transported over source regions (Maas and Grennfelt, 2016). Secondary aerosol from anthropogenic sources consists of both inorganic (~~, 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, favoring 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 sulphur dioxide (SO_2), nitrogen oxides (NOx) and ammonia (NH_3~~))~~), and organic compounds involving thousands of compounds and often poorly known reactions ~~(Hallquist et al., 2009)~~(Hallquist et al., 2009~~). Exposure of human to aerosol can be estimated by a variety of approaches, ranging from epidemiological studies to pure modeling approaches.~~. 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/mortality (PD) 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) accounting for 3.9 million premature deaths/year due to different model assumptions. In Europe, Brant et al. (2013) estimate 680 thousand 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. (2017) report a multi-model mean estimate of PD of 414.000 (range 230-570 thousand) for Europe and 160 thousand PDs for the USA. At the global scale, models, in some cases using satellite information (Brauer et al., 2015~~;Van Donkelaar et al., 2016);~~Van Donkelaar et al., 2016), are the most practical source of information of exposure to air pollution. However, model calculations are subject to a range of uncertainties related with incomplete understanding of transport, chemical transformation, removal processes, and not ~~to~~ the least, emission information.

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This work is developed in the context of the TF HTAP Phase 2 ~~(Galmarini et al., 2017), where a set of models is deployed to assess long range sensitivities to extra-regional emissions, using the prescribed HTAP_v2~~(Galmarini et al., 2017a), 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)~~(Janssens-Maenhout et al., 2015). Differences in model results illustrate uncertainties in model formulations of transport, chemistry and removal processes, and are addressed in separate studies (~~West~~Liang et al., ~~2017, in prep.),~~2018), but not of uncertainties in emission inventories. The objective of this study is to evaluate the relevance of uncertainties in regional sectorial emission inventories (power generation, industry, ground transport, residential, agriculture and international shipping), and ~~its~~their propagation in ~~modeled~~modelled $PM_{2.5}$ concentrations and associated impacts on health~~, comparing~~. To this end we couple the ~~derived~~HTAP_v2.2 global emission inventory for the year 2010 and the global source-receptor model TM5-FASST (TM5-FAst Scenario Screening Tool) to estimate global air quality in terms of $PM_{2.5}$ concentrations. The regional and global scale, the focus on annual $PM_{2.5}$ and associated health metrics, warrants 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. 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 ~~uncertainties~~contributions. A second objective of this analysis is to evaluate the importance of emission uncertainties at sector and regional level on $PM_{2.5}$, to better inform local, regional and hemispheric air quality policy makers on the potential impacts of ~~less known emission sectors or regions. In this work we couple the HTAP_v2.2 global emission inventory for the year 2010 and the global source receptor model TM5-FASST (TM5-FAst Scenario Screening Tool) to estimate global air quality in terms of $PM_{2.5}$ concentrations.~~sectors with larger uncertainties (e.g. residential and agriculture) or regions (e.g. developing and emerging countries).

2 Methodology

~~2.1~~2.1 TM5-FASST model and emission perturbations

The TM5-FASST model 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 performance with regard to linearity and additivity of concentration response to different size of emission perturbations and future emission scenarios. The validation of TM5-FASST against the full TM5 model runs is extensively discussed by van Dingenen et al. (2018), as well as the validity of the assumptions of linearity and additivity behind this reduced form-model. Below we summarize 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°x1° resolution source-receptor gridmaps obtained for TM5-FASST v0 (Van Dingenen et al., 2018). ~~**HTAP_v2.2 emissions**~~

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

~~2.2 TM5-FASST model~~

~~In order to calculate $PM_{2.5}$ concentrations from the HTAP_v2.2 emissions, the gridded TM5-FASST version 1.4b (Van Dingenen et al., 2017, in preparation).~~ The TM5-FASST source-receptor model is based on a set of emission perturbation experiments (-20 %) of SO_2, NOx, CO, NH_3, and VOC and CH_4 using the global 1°x1° resolution TM5 model, the meteorological year 2001 (~~chosen~~which was also used for the HTAP Phase 1 experiments) and the ~~representative concentration pathway~~community emission dataset prepared for the IPCC AR5 report (RCP, Representative Concentration Pathway) emissions for the year 2000 ~~(Lamarque et al., 2010)~~(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°x1° resolution emission spatial distribution from RCP year 2000 which was partly based EDGAR methodology and gridmaps. 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 ~~of~~) from natural sources is included in the model calculations using the parameterisation described in Dentener et al. ~~(2006)~~(2006), no explicit treatment of anthropogenic SOA is considered~~. Specifically, the change~~, 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 ranges regionally widely, and is deemed higher in regions with less VOC emission controls. We speculate that the inclusion of SOA would

possibly lead to a somewhat larger role of the transboundary pollution transport, mainly for those sectors emitting PM and VOCs (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 further evaluated in Van Dingenen et al. (2018) - the comparison of $PM_{2.5}$ concentrations~~, compared to a reference~~ 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 ~~in the receptor region *y* (~~resulting from an arbitrary emission scenario E_i using a "perturbation approach", i.e. the difference between E_i and $E_{i,ref}$ *(dE)* is considered as a perturbation on E_{ref} and the resulting concentration is evaluated as a perturbation *dPM*~~), induced by changes in precursor emissions in the source region *x* relativey to~~ on the reference ~~case (dE), is estimated as following:~~concentration:

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

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

where the summation is made over all primary emitted components and precursors (*i*) for secondary components, and $SRC_i[x,y]$ is a set of Source-Receptor Coefficients describing the linearized relationship between each precursor emission of specific components and concentration for each pair of source (x) and receptor (y) region. ~~Therefore~~Van Dingenen et al. (2018) explain in detail how the 'perturbation approach' can be also applied also for evaluating the attribution by sector as well as by source region. Thus to calculate total $PM_{2.5}$ concentration in each receptor region, the ~~sum of the~~ 56 source region individual contributions must be ~~taken into account~~summed. Using this approach, it is possible to evaluate the $PM_{2.5}$ concentrations from "within-region" and "extra-regional" $PM_{2.5}$ emissions. ~~Further details about the TM5-FASST methodology and assumptions can be found in Van Dingenen et al. (2017, in preparation) and Leitão et al. (2013).~~ 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., 2017b), in particular focusing on the $PM_{2.5}$ concentration reduction due to the contribution of the emissions of each anthropogenic sector (Eq. 3):

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

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

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As depicted in Fig. S1, 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 smaller regions (e.g. European countries) comparable with larger regions (like USA, China and India), in this work an aggregation procedure to 10 world regions (refer to Table S2) has been applied (China+, India+, SE Asia, North America, Europe, Oceania, Latin America, Africa, Russia and Middle East). In this work we focus on particulate matter due to its negative effects on human health ~~(WHO, 2013;Pope and Dockery, 2006)~~(WHO, 2013;Pope and Dockery, 2006), (Worldbank, 2016). The TM5-FASST model includes an assessment of the premature mortality due to ambient $PM_{2.5}$ concentrations on exposed population following the methodology developed by Burnett et al. (2014), as discussed in Sect. 4. Health impacts ~~of~~due to indoor air pollution or ozone are not evaluated in this work.

The aim of this work is to 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 e.g. 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 day much more accurate in particular given the focus on regional (and not so much gridded) emission analysis of our work.

2.2 HTAP_v2.2 emissions

The global anthropogenic emission inventory HTAP_v2.2 for the year 2010 is input to the global source-receptor model TM5-FASST to evaluate $PM_{2.5}$ concentrations for each world region/country with the corresponding health effects. The HTAP_v2.2 inventory includes for most countries official and semi-official annual anthropogenic emissions of SO_2, NOx, CO (carbon monoxide), NMVOC (non-methane volatile organic compounds), PM_{10} (particulate matter with a diameter less than 10 μm) $PM_{2.5}$, BC (black carbon) and OC (organic carbon) by country and sector (Janssens-Maenhout et al., 2015). Here we focus on the 6 major anthropogenic emission sectors contributing to global $PM_{2.5}$ concentrations, namely the power generation ("power"), non-power industry, industrial processes and product use ("industry"), ground transportation ("transport"), residential combustion and waste disposal ("residential"), agriculture ("agriculture") and international shipping ("ship"). 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

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HTAP_v2.2 emissions including their uncertainties. Aggregated emissions of a certain pollutant *p*, from a sector *i* and country *c* are calculated as the product of activity data (AD) and emission factors (EF), therefore the corresponding uncertainty ($\sigma_{i,c,p}$) is calculated as following:

$$\sigma_{EMI\ i,c,p} = \sqrt{\sigma^2_{ADi,c} + \sigma^2_{EF,i,p,c}}$$

(Eq.~~3~~4)

where σ_{AD} and σ_{EF} are the uncertainties (%) of the activity data and emission factors for a certain sector, country and pollutant. Uncertainty values of the activity data by sector and country are obtained from ~~Janssens-Maenhout et al. and references therein (2017, submitted.), while uncertainty~~Table 2 of Janssens-Maenhout et al. (2017, in review) 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), which is the result from larger uncertainties of about ±15% for non-Annex I countries, whereas uncertainties of less than ±5% are obtained for the 24OECD90 countries 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)~~(2013) and Bond et al. (2004) for particulate matter. ~~Differently from gridded emission inventories which often make use of similar proxies and spatial correlation structures, while errors in emissions may be correlated (e.g. the same systematic error in an estimate of EF introduced in the inventory for a number of countries),~~In this work we assume ~~here~~ that reported countries emissions are based on independent ~~evaluation~~estimations of activity data and ~~estimated~~emission factors 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 $\sigma_{EMI\ p,c}$ for a certain pollutant (*p*) due to all sectors (*i*) in a specific country (*c*) with the following equation (EMEP/EEA, 2013).

$$\sigma_{EMI\ p,c} = \sqrt{\sum_i \left(\sigma_{EMI\ i,c,p} * \frac{EMI_{i,c,p}}{EMI_{tot,c,p}}\right)^2} \qquad \text{(Eq. 45)}$$

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

Table S3, reports the overall uncertainty calculated for each pollutant and for each TM5-FASST region. Using an additional constraint that EFs and activities cannot be negative, a lognormal distribution of the calculated uncertainties is assumed (Bond et al., 2004)~~; therefore~~. 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

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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*2 for the uncertainty analysis).

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3 TM5-FASST modelling results

3.1 Regional contributions to $PM_{2.5}$ concentrations

Figure 1 provides a global perspective on the fraction of within-region and extra-regional $PM_{2.5}$ concentrations for 10 aggregated world receptor regions using emissions of the year 2010, with the extra-regional fraction (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) ranged from few $\mu g/m^3$ (e.g. in Oceania or Latin America), around 7-8 $\mu g/m^3$ for North America and Europe, and up to 33-39 $\mu g/m^3$ in China+ (including also Mongolia) and India+ (including also the rest of South Asia). Anthropogenic $PM_{2.5}$ pollution in China+ and India+ is mainly affected by large emission sources within the country (98 and 96%, respectively), although 4 % of the Indian anthropogenic $PM_{2.5}$ pollution is mainly transported from the Gulf region and Middle East. North America (98%) and Oceania (98%) are mainly influenced by within-regional pollution due to their geographical isolation from other regions. TM5-FASST computations attributed 11 % of the $PM_{2.5}$ in Europe to extra-regional sources; for the Middle East and Gulf region extra-regional contributions amount to 18% (mainly from Europe and Russia), for Africa 25% (mainly from Europe and Middle East), and Russia 28% (mainly from Europe, Middle East and Gulf region and China). Shipping emissions are not considered in this ~~analysis~~figure due to their international origin, while inland waterways 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 Korea and Japan ~~(Park et al., 2014)~~(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.

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Focusing on Europe, Fig. 2 shows within-region (in black) vs. extra-regional absolute population-weighted $PM_{2.5}$ concentrations (in $\mu g/m^3$) for 16 EU countries plus Norway and Switzerland, defined in TM5-FASST, as well as the source regions contributing to this pollution. ~~Annual~~Regional annual averages of population weighted $PM_{2.5}$ concentrations in Europe vary between 2-4 $\mu g/m^3$ in Northern European countries (like Finland, Norway and Sweden) up to 10-12 $\mu g/m^3$ for continental Europe. Although most of EU annual average $PM_{2.5}$ concentrations are below the World Health Organization Air Quality Guideline of 10 $\mu g/m^3$ $PM_{2.5}$ (as annual average), these values represent only regional averages while several exceedances ~~especially~~ in urban areas are often observed in Europe. As 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, etc.). ~~From a European~~

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~~perspective, $PM_{2.5}$ represents a transboundary issue since extra-regional contributions range between~~Transboundary air pollution from external regions contributes by 27% to 75 % and ~~75% (~~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, Czech Republic, Rest of Central EU, Poland and Germany), Czech Republic (67%, mainly from Poland, Germany and Austria), Austria and Slovenia (66%, mainly from Czech Republic, Germany and Italy), Sweden+Denmark (65%, mainly from Germany, Norway and Poland), Bulgaria (63%, mainly from Romania), and Greece (61%). The remaining EU countries are both affected by within-region and extra-regional pollution (the latter ranging from 40% to 59%), highlighting the importance of transboundary transport of $PM_{2.5}$ concentrations. For example Switzerland is influenced by the pollution coming from France, Italy and Germany; Rest of Central EU by Poland and Germany; Germany by France and Benelux; Poland by Czech Republic and Germany. Interestingly, Romania, Bulgaria, Greece and Hungary are also significantly affected by the pollution transported ~~by~~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)~~(Maas and Grennfelt, 2016) ~~where the importance of transboundary transport to PM, which in Europe mainly consists of secondary organic (not fully treated by TM5-FASST) and inorganic particles (e.g. ammonium nitrate and sulfate) formed from gaseous precursors, is highlighted. Therefore, in order to reduce regional mean PM concentrations, across-regional approach taking into account atmospheric transport and chemical transformations of pollutants over a wide scale could be considered.~~, which highlights the importance of transboundary transport of organic and inorganic PM.

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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 colors) and within-region pollution, while Table 1 shows regional average values of sector-specific relative contributions. In most African regions (except Egypt) anthropogenic $PM_{2.5}$ concentrations are mainly produced by emissions in the residential sector. Agriculture is an important sector for Egypt, while Northern Africa is strongly influenced by shipping emissions in the Mediterranean (30%). $PM_{2.5}$ in emerging economies in Asia, Latin America and Middle East are dominated by $PM_{2.5}$ concentrations from the residential sector, power generation and industrial. Asian countries, China, India, Indonesia and Philippines are mainly influenced by within-region pollution with the largest contributions coming from power, industry and residential sectors. Japan is ~~characterized~~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. Rest of South Asia and South Eastern Asia) from the power, industry and residential activities. A different picture is ~~observed~~seen for Europe where according to our calculations, annual PM concentrations stem mainly from the agricultural and residential sectors with a somewhat ~~less~~lower contribution from the transport sector. In Eastern European countries ~~relevant~~noticeable contributions are also found from the power and

industrial sectors ~~in Eastern European countries, related~~due to the relatively extensive use of polluting fuels like coal. $PM_{2.5}$ concentrations in USA and Canada are mostly ~~affected by~~from the power, industry and agricultural sectors. In Oceania industry and agriculture are the most important sectors. $PM_{2.5}$ ~~concentrations formed~~ from ship emissions mainly affect coastal areas of North Africa, SE Asia (e.g. in Japan, Taiwan, Malaysia, Indonesia and Philippines), Mediterranean countries (Spain ~~by~~ 11%, Italy ~~by~~ 5%, France ~~by~~ 7% of their corresponding country totals), Northern EU regions (Great Britain ~~by~~ 10%, Norway ~~by~~ 6%, Sweden and Denmark ~~by~~ 10% of their corresponding country totals) and Oceania (22% of the regional total). Over the international areas of sea and air no distinction between within-region and extra-regional concentrations is reported. Further details on within-region and extra-regional concentrations can be found in section S2 of the Supplementary Material.

3.3 Gridded $PM_{2.5}$ concentrations

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

~~Modelled~~The TM5-FASST model developed by van Dingenen et al. (2018) has been validated against concentration estimates derived from the WHO database (WHO, 2011, 2014, 2016) and satellite-based measurements (van Donkelaar et al., 2010, 2014). General good agreement is found between the $PM_{2.5}$ concentrations modeled by TM5-FASST and the measured ones reported in the WHO database for Europe (within 20% deviation), North America (within 5% deviation) and partly China due to the higher accuracy of the measurements. The comparison for Latin America and Africa is much less robust (40-60% deviation from the 1:1 line) and the scatter possibly highlights a non-optimal modeling of specific sources relevant for these regions by TM5-FASST (e.g. large scale biomass burning) by the TM5-FASST model. Similar results are also found comparing regional averages of urban stations from WHO against the FASST population weighted average of grid cells. The TM5-FASST modeled $PM_{2.5}$ concentrations have been 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

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Donkelaar et al., 2010, 2014). The regional comparison shows consistent results with the ground based measurements comparison (e.g. good agreement for EU and USA within 10% deviation, while lower agreement for developing and emerging countries).
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;Brauer et al., 2015;Boys et al., 2014;~~Evans et al., 2013;Van Donkelaar et al., 2016)~~Evans et al., 2013;Van Donkelaar et al., 2016), reporting for the whole Europe annual averaged $PM_{2.5}$ concentrations in the range between 11 and 17 μg/m^3, for Asia from 16 to 58 μg/m^3, Latin America 7-12 μg/m^3, Africa and Middle East 8-26 μg/m^3, Oceania 6 μg/m^3 and North America 13 μg/m^3 (note that measurements and satellite estimates would not separate anthropogenic and natural sources of PM, e.g. dust, large scale biomass burning, while the concentrations in this study ~~pertain to~~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 having emerging economies and fast development (e.g. Middle East, China and India), while the ground transport sector is a more important source of PM concentrations in industrialised countries (e.g. North America and Europe) and in developing Asian countries. The residential sector is ~~one of the most significant sources~~an important source of PM all over the world, ~~potentially~~ also affecting indoor air quality~~.~~ (Ezzati, 2008;Lim et al., 2013;Chafe et al., 2014). Africa and Asia are strongly influenced by PM concentrations produced by this sector due to the incomplete combustion of rather dirty fuels and solid biomass deployed for domestic ~~purposes (both~~ heating and cooking~~).~~ purposes. Interestingly, the agricultural sector is 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 ~~(Maas and Grennfelt, 2016).~~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 emissions more difficult to be effectively ~~regulate with emission reductions~~regulated than point source emissions of the industrial and power sectors (e.g. in Europe the Large Combustion Plant Directive, the National Emission Ceilings or the Industrial Emissions, the Euro norms for road transport, etc.). Finally, shipping is mainly contributing to the pollution in countries and regions with substantial coastal areas, and with ship tracks on the Mediterranean Sea, the Atlantic, Pacific and Indian Oceans, as depicted in Fig. 4.

3.4 Uncertainty from emissions

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

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

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

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

3.4.2 Ranking the sector specific contribution to emission uncertainties

Figure 6 shows the average sector relative contribution to total emission inventory related uncertainty for the main $PM_{2.5}$ concentration precursors and world regions~~, representing the~~. 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 is provided in Fig. S2, where

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the share of each term of the sum of Eq. ~~4,~~5 $\left(\sigma_{EMI\ i,c,p} * \frac{EMI_{i,c,p}}{EMI_{tot,c,p}}\right)^2$, representing the sector contribution to the uncertainty of each pollutant in each region, is reported. SO_2 uncertainties mainly derive from the power generation sector ~~for most of the world countries~~ especially ~~those that are~~countries with a dominant coal ~~dominated~~use; however, ~~relevant~~substantial contributions are also ~~observed from~~computed for the industrial sector in South Africa, Asia, Norway, some Latin American countries, Canada and Russian countries. 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. Korea, Vietnam, Indonesia, South Eastern Asia, etc.). 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%). NOx emissions uncertainty mainly ~~derives~~stems from the transport sector, although some contributions are also seen from ~~the~~ power generation in ~~Russian~~Russia, countries ~~with~~ strongly relying on gas ~~and~~ (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) ~~sector~~sectors and for some regions by a similar contribution of these two sectors. NMVOC emission uncertainties mainly derive from ~~the~~ poorly characterized industrial, transport and residential activities ~~which are still not well characterized in terms of NMVOC emissions~~ due to the complex mixture and reactivity of such pollutants. As expected, NH_3 emission uncertainty is dominated by the agricultural sector which appears to be less relevant for all other pollutants. 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 ~~and~~, transport ~~sectors~~ and ~~partly for the~~in particular industrial ~~one~~sectors. Black carbon emission inventories should be better characterised in Europe, Japan, Korea, Malaysia etc. for the transport sector, where the higher share of diesel used as fuel for vehicles leads to higher BC emissions; in addition, BC emissions from the residential sector require further effort to better ~~characterise them in terms of~~define EFs for the different type of fuels used under different combustion conditions. To constrain and improve particulate organic matter emissions, efforts should be dedicated to ~~the~~improve residential emissions ~~characterisation~~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 ~~largely deployed~~widely used in the residential ~~activities~~sector, but national reporting often underestimates the emissions from this sector ~~in Europe~~, due to the fact that often informal economic wood sales are not accurately reflected in the official statistics of wood consumption (AD) ~~(Denier Van Der Gon et al., 2015)~~(Denier Van Der Gon et al., 2015). An additional uncertainty is related to the lack of information in the inventory regarding the emission factors (EF) variability, which depends on the combustion efficiency and type of wood ~~(Weimer et al., 2008;Chen et al., 2012)~~(Weimer et al., 2008;Chen et al., 2012). In our work we estimate the uncertainty attributable to wood combustion in the ~~HTAP_v2.2~~ residential sector (σ_{AD,RES_bio}) by comparing it to the recent TNO RWC (residential wood combustion) inventory of Denier van der

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Gon et al. (2015), which includes a revised biomass fuel consumption~~,~~ with the corresponding EDGARv4.3.2 activity data (Janssens-Maenhout et al., 2017~~, in prep.),~~), as shown in Table S4. In the TNO RWC inventory, wood use for each country has been updated comparing the officially reported per capita wood consumption data (from GAINS and IEA) with the expected specific wood use for a country including the wood availability information ~~(Visschedijk et al., 2009;Denier Van Der Gon et al., 2015)~~(Visschedijk et al., 2009;Denier Van Der Gon et al., 2015). We can therefore assume that the TNO RWC inventory represents an independent estimate of wood consumption in the residential sector, allowing a more precise uncertainty estimation of the AD for this sector. Assuming that emissions are calculated as the product of AD and EF, the corresponding uncertainty can be calculated with Eq. ~~3~~4, where σ_{AD} ranges from 5 to 10% for European countries and Russia as reported for international statistics ~~(Olivier et al., 2016)~~(Olivier et al., 2016). We can therefore calculate the residential emission factors uncertainty of each individual pollutant ($\sigma_{EF,p}$) from Eq. ~~3~~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 ones, we can evaluate the mean normalized absolute error (MNAE) considering all N countries, following Eq. ~~5~~6 ~~(Yu et al., 2006)~~(Yu et al., 2006), which represents our estimate of $\sigma_{AD,RES\ bio}$:

~~$$MNAE = \frac{1}{N} * \sum_{j}^{N} \frac{|TNO\ RWC_j - EDGARv4.3.2_j|}{TNO\ RWC_j}$$~~ (Eq.~~5~~6)

We estimate a value of $\sigma_{AD,RES\ bio}$ of 38.9% which is much larger compared to the 5-10% uncertainty ~~assumed~~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. ~~3~~4 and the calculated σ_{AD,RES_bio} and $\sigma_{EF,p}$, we can evaluate a new σ_{EMI,p,RES_bio} for the residential sector including the uncertainty of the AD due to the use of wood as fuel for this sector, as reported in Table S5. Comparing the results shown in Table S5 with the factor of two uncertainty values expected for PM emissions from the residential sector ~~(Janssens-Maenhout et al., 2015)~~(Janssens-Maenhout et al., 2015), we derive that the uncertainty associated with the emission factors for biomass combustion in the residential sector is the dominant source of uncertainty compared to the ~~AD (wood consumption)~~ uncertainty in wood burning activity data. Large increases in reported biomass usage for domestic use has been noted in IEA energy statistics for some European countries ~~(IEA, 2013,2014,2015,2016)~~(IEA, 2013,2014,2015,2016) and further increases are expected as countries are shifting their methodologies to estimate biofuel activity data away from fuel sales statistics to a ~~modeling~~modelling approach based on energy demand. In addition, several EU countries ~~have planned to increase~~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). When comparing the UNFCCC and the TNO RWC data, a higher value of $\sigma_{AD,RES\ bio}$ is obtained (59.5% instead of 38.9%), although its effect on the final residential emission uncertainty is less strong, as shown in Table S6. Table 3 shows the impact of biofuel combustion uncertainty in the residential sector on $PM_{2.5}$

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concentrations. Upper-end uncertainties indicate that $PM_{2.5}$ concentrations could be between 2.6 and 3.7 times larger than those derived from the HTAP_v2.2 inventory.

4 Health impact assessment

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

In this section, we investigate the impact of total and sector-specific anthropogenic population weighted $PM_{2.5}$ concentrations on health and we show comparisons with mortality estimates provided by WHO and scientific publications ~~(Silva et al., 2016)~~(Silva et al., 2016). Figure 7 represents the premature deaths (PD) distribution due to air pollution, using population weighted $PM_{2.5}$ concentrations and representative for anthropogenic emissions in the year 2010. The most affected areas are China and India, but also some countries of Western Africa and urban areas in Europe (in particular in the Benelux region and Eastern Europe). Our computations indicate that annual global outdoor premature mortality due to anthropogenic $PM_{2.5}$ amounts to 2.1 million premature deaths, with an uncertainty range related to emission uncertainty of 1-3.3 million deaths/year. ~~In 2010, 82% of the PD occurs in fast growing economies and developing countries, especially in China with 670000 and India with an almost equal amount of 610000 PD/year.~~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, submitted). Solazzo et al. 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 thousands (inter-quantile range between 260 and 415 thousand). Moreover, they estimate that a reduction in the uncertainty of the modelled ozone 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 in 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 thousand and India

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with an almost equal amount of 610 thousand PD/year. Table 4 summarizes our estimates of premature mortality for aggregated world regions, with Europe accounting for ~~210000~~210 thousand PD/year and North America ~~100000~~100 thousand PD/year.

Our results are comparable with Lelieveld et al. ~~(2015)~~(2015) and Silva et al. ~~(2016)~~(2016) who ~~estimate~~ 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)~~(2017) estimates a higher value of global mortality ~~of (~~3.9 million PD/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 1x1 degree resolution as metric for estimating health effects due to air, 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 ~~are advanced by~~from the Global Burden of Disease (http://vizhub.healthdata.org/gbd-compare; 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 ~~from a source~~for each receptor region ~~perspective~~, highlighting the premature mortality induced ~~by each source region~~ within the country itself and outside the ~~emitting~~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 ~~equal 54000~~contribute for ca 700 thousand and ~~76000~~ca 500 thousand PD/year, respectively, representing ~~a high contribution~~more than 50% of ~~ca. 10 % to~~ the global mortality. Clearly, reducing emissions and emission uncertainties in these two regions will have therefore the largest over-all benefit on global air quality improvement and understanding as well as on global human health. For most of the TM5-FASST regions, ~~PD~~PDs due to anthropogenic emissions within the source region are higher than the extra-regional contributions. However, there are marked exceptions, such as the Gulf region, Hungary, Czech Republic, Mongolia, etc., where the extra-regional and within-region contributions to mortality are at least comparable. In fact Hungary and Czech Republic are strongly influenced by polluted regions in Poland (mainly); likewise Mongolia is suffering from the vicinity of source 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 Figs. 8a and 8b. Health effects induced by air quality in industrialized countries are mainly related with agriculture (32.4% of total mortality or ~~6800068~~ thousand PD/year), residential combustion (17.8% or ~~3700037~~ thousand PD/year) and

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road transport (18.7% or ~~3900039~~ thousand PD/year) for Europe and with power generation (26.4% or ~~2600026~~ thousand PD/year), industry (19% or ~~1900019~~ thousand PD/year), residential (17% or ~~1700017~~ thousand PD/year) and agriculture (24.0% or ~~2400024~~ thousand PD/year) for North America. The health impacts observed in most Western EU countries is due both to within-regions and extra-regional pollution, while in several Eastern EU countries the impact of neighbouring countries is even larger compared to within-region pollution. The premature deaths induced by international shipping emissions represent 5.5% of total EU PD, in the range the results of Brandt et al. (2013a) (ca 50 thousand PDs). PM related mortality in developing countries and fast growing economies is mostly affected by industrial (up to 42% in China or ~~279000~~279 thousand PD/year) and residential activities (ranging from 27% in China and 76% in Western Africa), and also by power generation (up to 24% in India or 113000 PD/year). Chinese emissions have a strong impact on China, Japan, Vietnam, Mongolia+Korea, Thailand while the Indian emissions impact the rest of South and South Eastern Asia. Reducing Chinese and Indian emissions will reduce the PM related mortality in almost all countries in Asia. Our results are in agreement with the study of Oh et al. ~~(2015)~~(2015) where they highlight the role of transported pollution from China in affecting Korean and other South Eastern Asian countries $PM_{2.5}$ concentrations and health effects, as well as the need of international measures to improve air quality.

Conclusions

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

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

Using the HTAP_v2.2 emission inventory and TM5-FASST, we also evaluate how the uncertainty in sectors and regions propagates ~~to~~into $PM_{2.5}$. The aim of our analysis is to provide insights on where the emission inventories of each country could be improved, because of their highest uncertainty and highest contribution to the formation of $PM_{2.5}$ concentrations. The uncertainty of PM concentrations depends in variable proportions to the uncertainties of the emissions within receptor regions, and surrounding regions. We show that reducing the uncertainties in the Chinese and Indian emission inventories (e.g. from industry and residential

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sectors) will be highly relevant for understanding the long-range sources of $PM_{2.5}$ pollution in most of Asian countries. Here we demonstrate how analysis of uncertainties in national/regional sectorial emission inventories can further inform coordinated transboundary and sector-specific policies to significantly improve global air quality. Among all anthropogenic emission sectors, the combustion of biomass for household purposes represents one of the major sources of uncertainties in emission inventories both in terms of wood consumption and emission factor estimates. Further effort is therefore required at national level to better characterize this source.

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

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Tables and Figures

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

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

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Table 2 - Annual average $PM_{2.5}$ concentrations ($\mu g/m^3$) with upper and lower range in brackets due to emission inventories uncertainty ~~(σ)~~ (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+σ) respectively. ~~Uncertainty~~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 ($\mu g/m^3$)	Fraction of uncertainty due to primary PM emissions (%)
Asia	South Korea	13.8 (8.3 - 24.9)	71%
	Japan	6.9 (3.8 - 13.3)	84%
	Mongolia+ North Korea	14.6 (9.0 - 25.9)	75%
	China	39.9 (22.4 - 76.6)	78%
	Taiwan	6.4 (3.7 - 10.9)	77%
	Rest of South Asia	29.3 (13.9 - 64.9)	87%
	India	34.7 (16.6 - 73.4)	86%
	Indonesia	2.4 (1.3 - 4.6)	86%
	Thailand	8.0 (5.1 - 12.6)	88%
	Malaysia	3.1 (1.8 - 5.2)	85%
	Philippines	2.0 (1.1 - 3.8)	80%
	Vietnam	14.2 (7.0 - 30.4)	92%
	Rest of South Eastern Asia	8.6 (4.6 - 17.6)	89%
Europe	Austria+Slovenia	8.4 (4.0 - 19.6)	59%
	Switzerland	10.1 (4.9 - 23.3)	52%
	Benelux	10.1 (5.2 - 22.7)	59%
	Spain+Portugal	5.4 (3.4 -9.4)	77%
	Finland	2.6 (1.3 - 5.8)	66%

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	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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2

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

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	Russia (Asia)	2.7 (1.7 - 5.1)	64%
	Ukraine	7.8 (4.2 - 15.9)	65%
Oceania	Australia	1.1 (0.8 - 1.4)	84%
	New Zealand	0.3 (0.1 - 0.5)	60%
	Pacific Islands	0.2 (0.1 - 0.4)	75%

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

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

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

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

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

Table 5 – Number of premature deaths ~~caused by major source regions and their contribution to global mortality (~~for each receptor region including the within-region and extra-regional attribution~~). For the RERER metric refer also to Table S2.~~

~~world regions~~	~~TM5-FASST FASST codes~~	~~PD induced by source region (deaths/year)~~	~~Within-region PD (deaths/year)~~	~~Extra-regional PD (deaths/year)~~
~~Africa~~	~~Eastern Africa~~	~~9451~~	~~8218~~	~~1233~~
~~Africa~~	~~Egypt~~	~~11137~~	~~10783~~	~~354~~
~~Africa~~	~~Northern Africa~~	~~3904~~	~~3427~~	~~477~~
~~Africa~~	~~Rep. of South Africa~~	~~8813~~	~~8797~~	~~15~~
~~Africa~~	~~Southern Africa~~	~~248~~	~~32~~	~~216~~
~~Africa~~	~~Western Africa~~	~~19785~~	~~19785~~	~~0~~
~~Asia~~	~~China~~	~~696823~~	~~643129~~	~~53694~~
~~Asia~~	~~Indonesia~~	~~15352~~	~~14803~~	~~549~~
~~Asia~~	~~India~~	~~488319~~	~~412298~~	~~76021~~
~~Asia~~	~~Japan~~	~~15181~~	~~15181~~	~~0~~
~~Asia~~	~~South Korea~~	~~8789~~	~~7510~~	~~1279~~
~~Asia~~	~~Mongolia+ North Korea~~	~~8786~~	~~4076~~	~~4710~~
~~Asia~~	~~Malaysia~~	~~2225~~	~~1058~~	~~1167~~
~~Asia~~	~~Philippines~~	~~94~~	~~94~~	~~0~~
~~Asia~~	~~Rest of South Asia~~	~~113040~~	~~67170~~	~~45870~~
~~Asia~~	~~Rest of South Eastern Asia~~	~~4064~~	~~3814~~	~~250~~
~~Asia~~	~~Thailand~~	~~10898~~	~~10495~~	~~403~~
~~Asia~~	~~Taiwan~~	~~1028~~	~~1028~~	~~0~~
~~Asia~~	~~Vietnam~~	~~24401~~	~~20286~~	~~4115~~
~~Europe~~	~~Austria+Slovenia~~	~~3668~~	~~1674~~	~~1994~~
~~Europe~~	~~Bulgaria~~	~~5986~~	~~2269~~	~~3717~~
~~Europe~~	~~Benelux~~	~~12991~~	~~6057~~	~~6933~~

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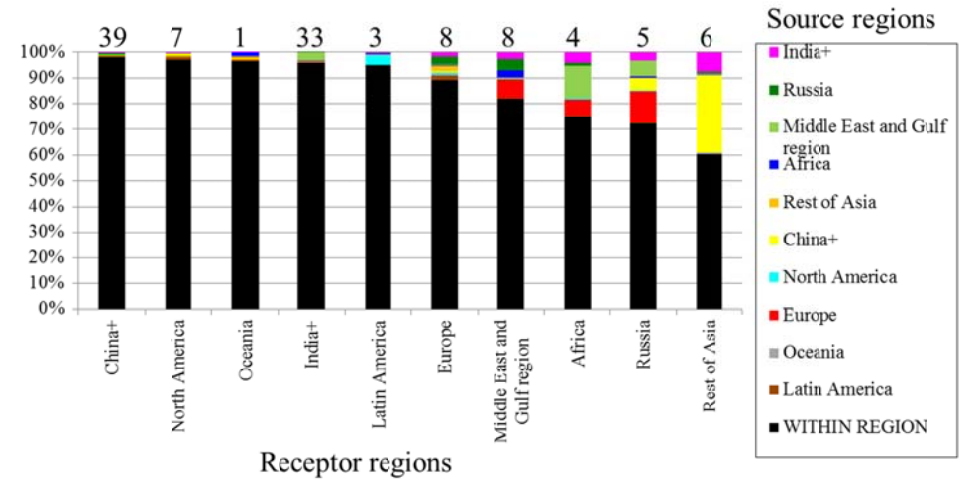
Europe	Switzerland	3036	1404	1632
Europe	Czech Republic	8957	3540	5417
Europe	Germany	33343	23001	10342
Europe	Spain+Portugal	10454	9541	914
Europe	Finland	0	0	0
Europe	France	23901	15148	8753
Europe	Great Britain+Ireland	12588	11157	1431
Europe	Greece+Cyprus	2112	1520	592
Europe	Hungary	4629	3889	740
Europe	Italy+Malta	18541	17373	1168
Europe	Norway	26	26	0
Europe	Poland+Baltic	23825	16811	7014
Europe	Rest of Central EU	9570	6239	3331
Europe	Romania	15374	8360	7014
Europe	Sweden+Denmark	90	88	2
Latin America	Argentina+Uruguay	114	75	39
Latin America	Brazil	4089	3968	120
Latin America	Chile	3391	3283	108
Latin America	Mexico	9410	8447	964
Latin America	Rest of Central America	3569	2772	797
Latin America	Rest of South America	4205	4164	41
Middle East	Golf region	34270	11225	23046
Middle East	Middle East	3993	2804	1189
Middle East	Turkey	32442	24191	8252
North America	Canada	5279	1491	3788
North America	USA	92885	90176	2709
Oceania	Australia	1010	25	985
Oceania	New Zealand	15	15	0
Oceania	Pacific Islands	1	1	0
Russia	Kazakhstan	2000	1100	900
Russia	Former USSR Asia	7419	6420	999
Russia	Russia (Asia)	3607	601	3006
Russia	Russia (EU)	19419	12704	6714
Russia	Ukraine	57352	44604	12748



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

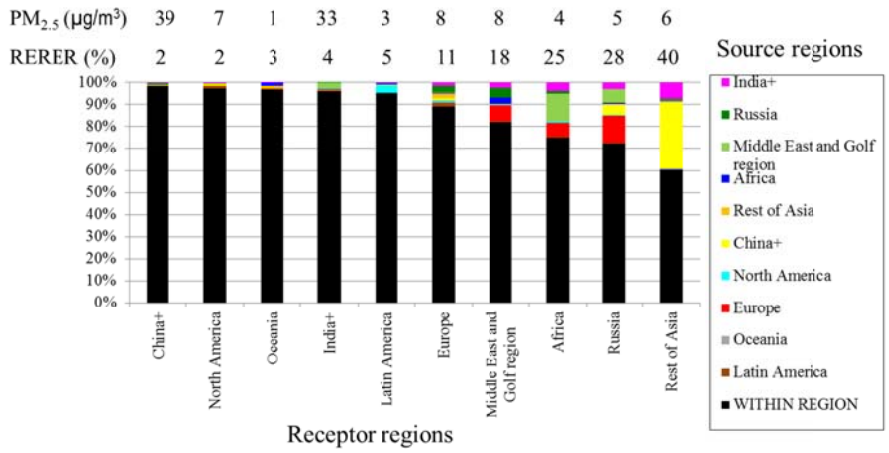


Figure 1 – Within-region vs. imported extra-regional anthropogenic population weighted $PM_{2.5}$ concentrations [%] for aggregated world regions. Annual average population weighted anthropogenic concentrations (in µg/m^3) are reported on top of each bar, together with the RERER metric (%). Ship emissions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

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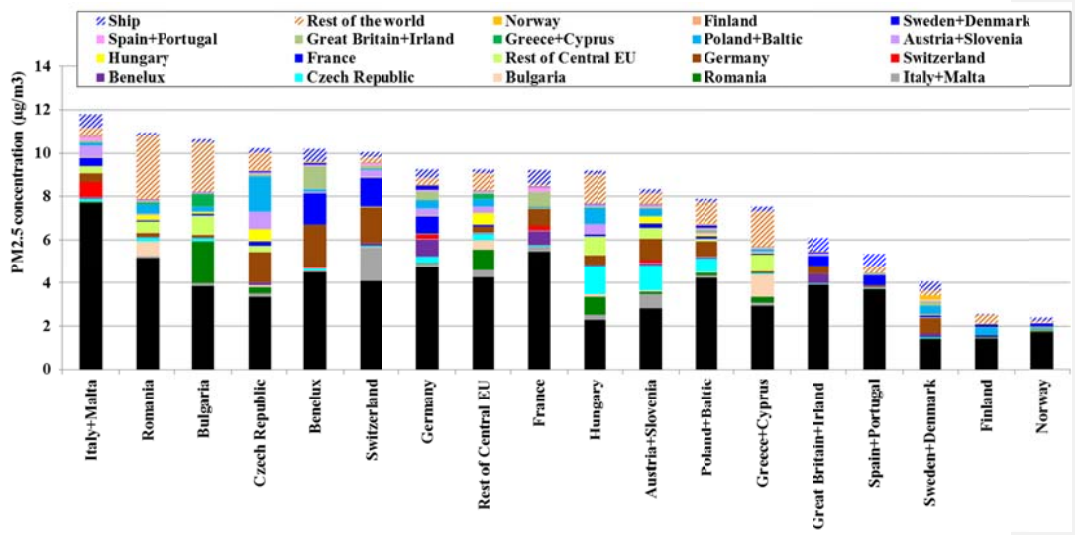

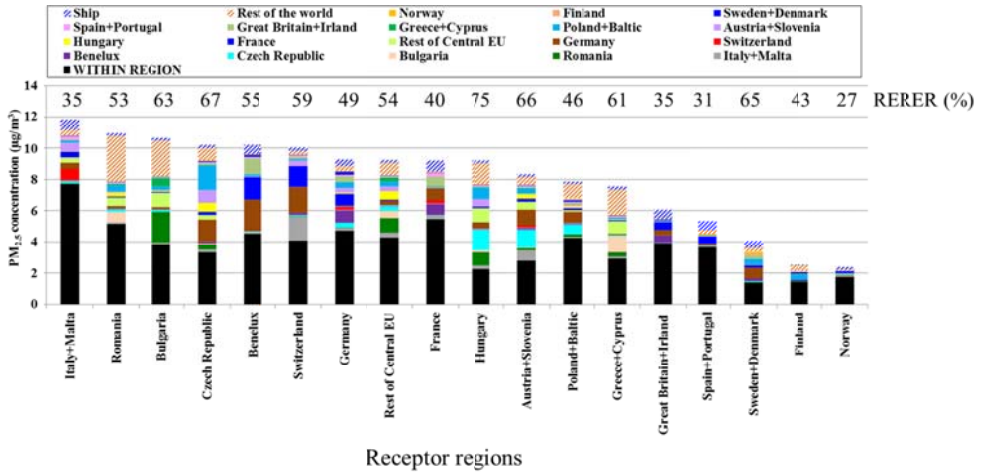

Figure 2 – Anthropogenic $PM_{2.5}$ concentrations in 18 countries and sub-regions in Europe separated in within-region and extra-regional contributions. The RERER metric (%) is reported on top of each bar.

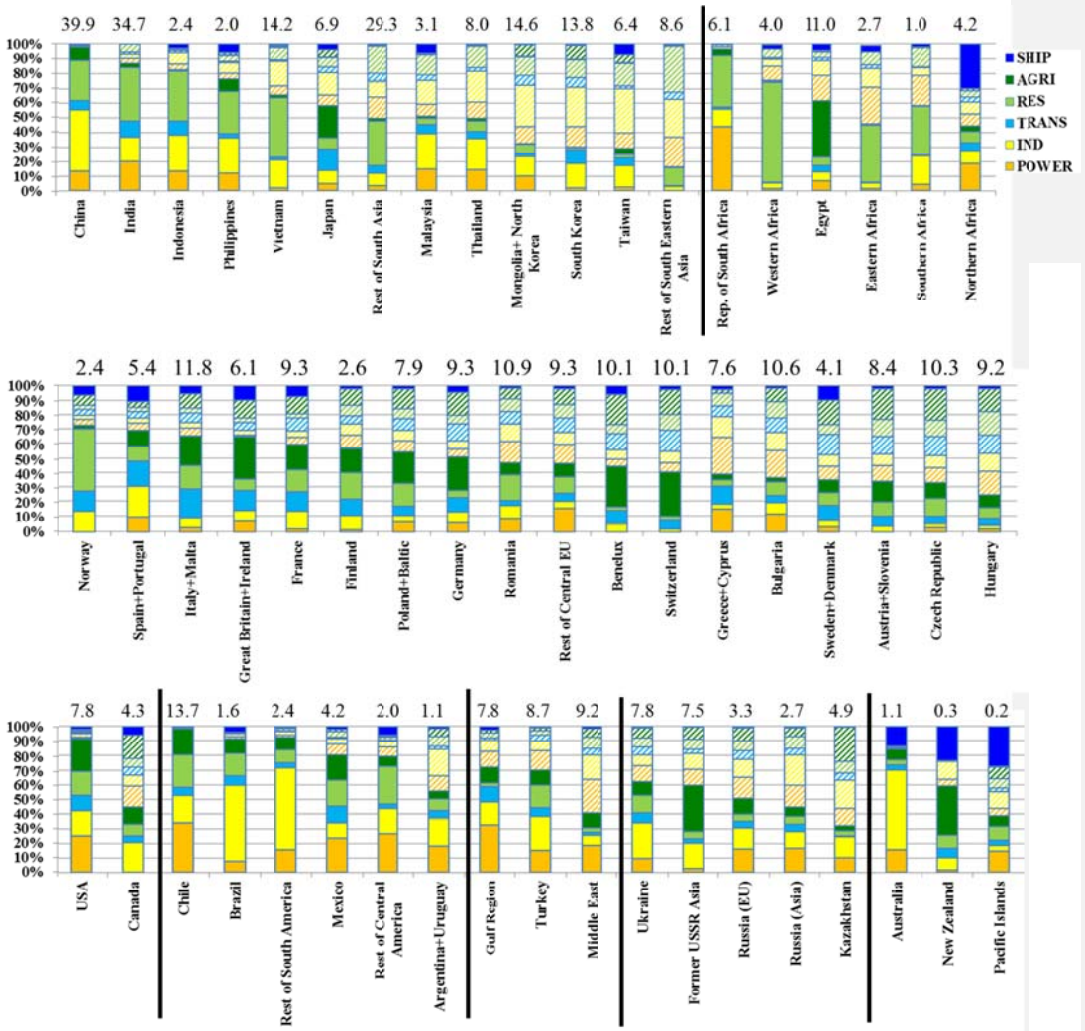

Figure 3 - Fraction of within-region and extra-regional (shaded areas) anthropogenic $PM_{2.5}$ concentrations separate by sector for receptor region within the macro-regions: Asia and Africa (upper panel), Europe (middle panel), North America, Latin America, Middle East, Russia and Oceania (lower panel).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

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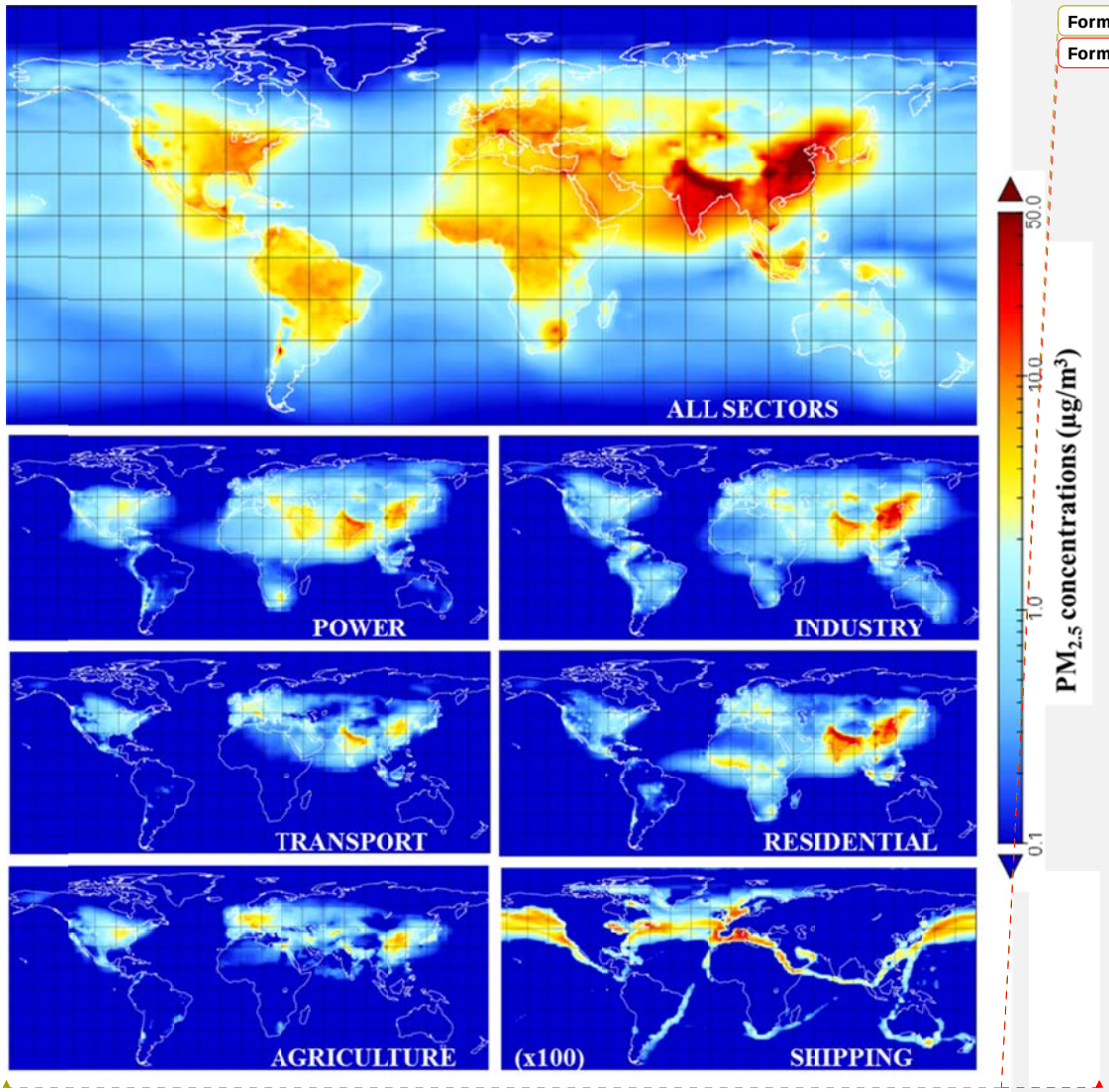

Figure 4 – Total anthropogenic $PM_{2.5}$ concentrations (µg/m^3) and sectorial contributions using 2010 emissions.

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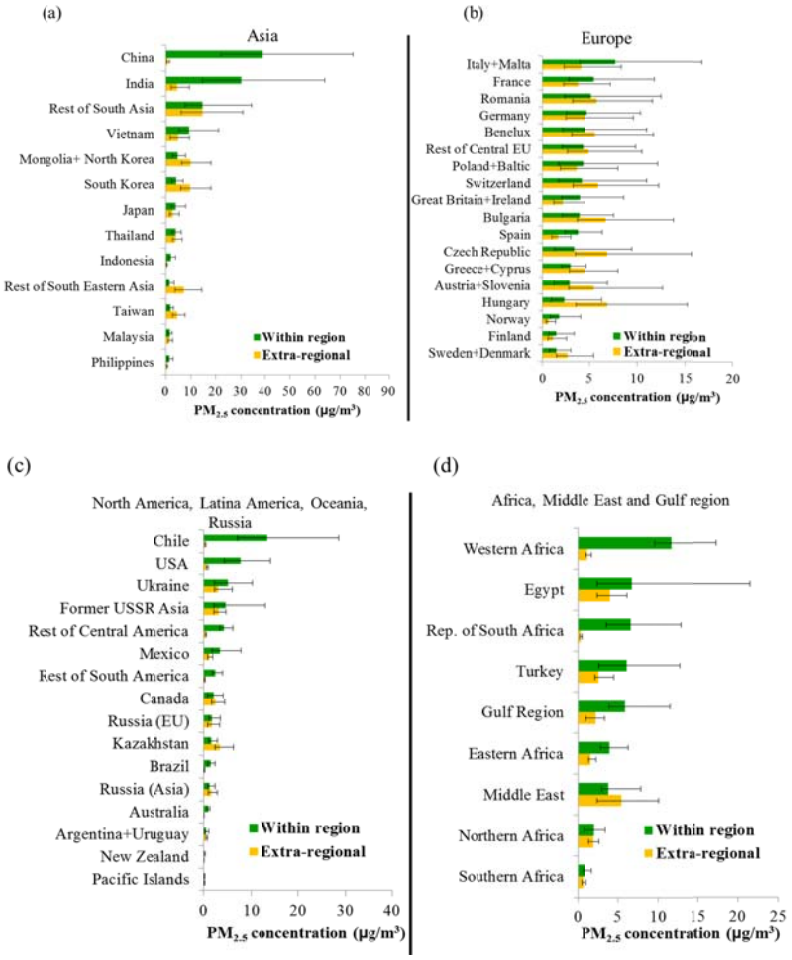

Figure 5 - Within-region and extra-regional anthropogenic $PM_{2.5}$ concentrations for Asia (panel *a*), Europe (panel *b*), North America, Latin America, Oceania and Russia (panel *c*) and Africa, Gulf region and Middle East (panel *d*). The error bars are calculated multiplying and dividing the reference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

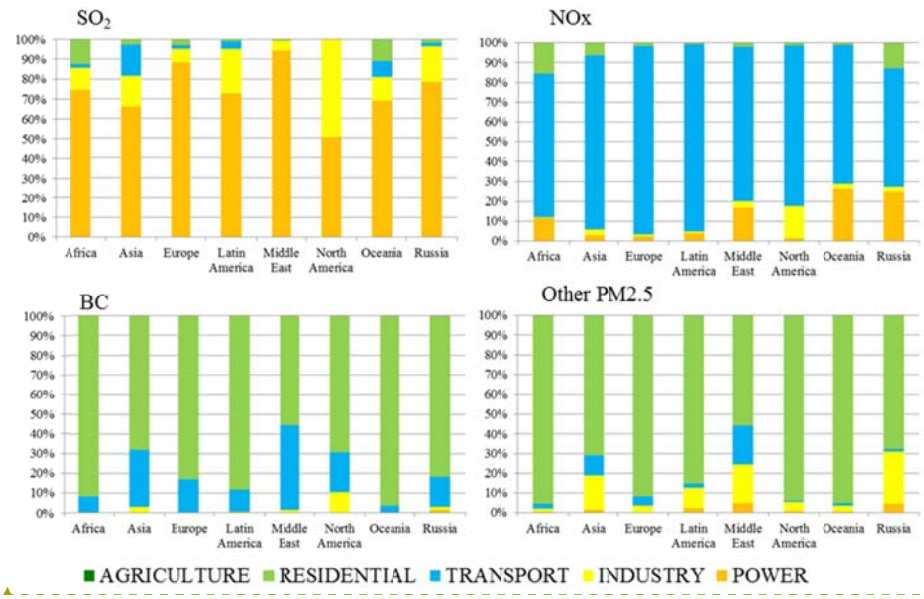

Figure 6 – Contribution of anthropogenic sectors to the emission uncertainty of various pollutants ~~S~~ for different world regions.

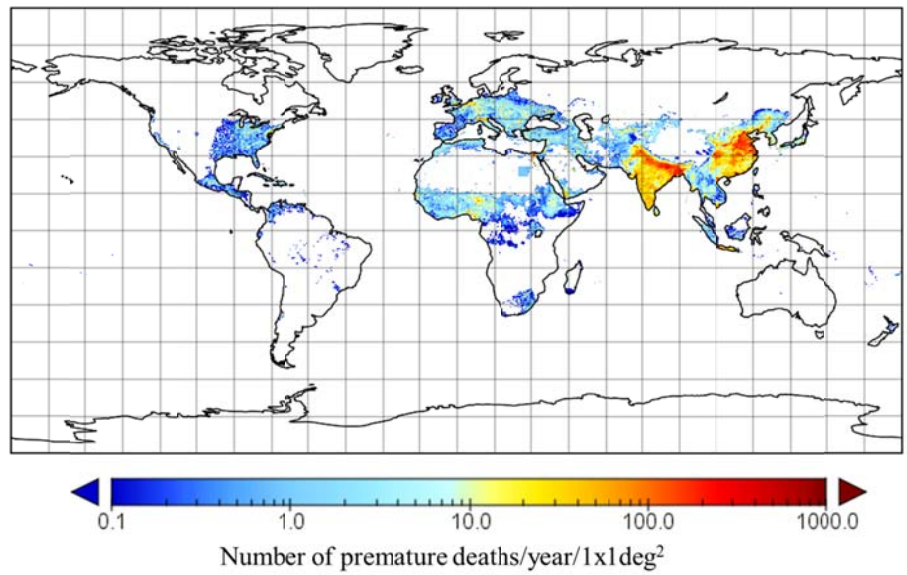

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Figure 7 – Global distribution of premature deaths in 2010 caused by anthropogenic particulate matter pollution estimated using the methodology described in Burnett et al. (2014).

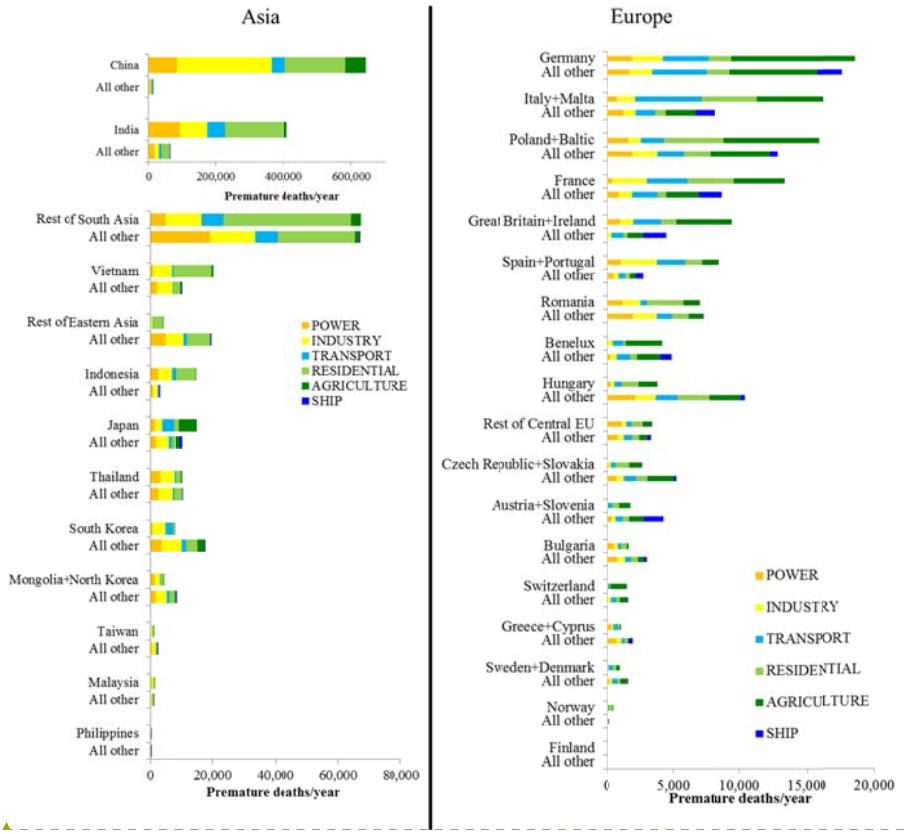

Figure 8a – Anthropogenic emission sector contributions to premature mortality (deaths/year) due to $PM_{2.5}$ population weighted concentrations in the TM5-FASST receptor regions of Asia (left) and Europe. (right).

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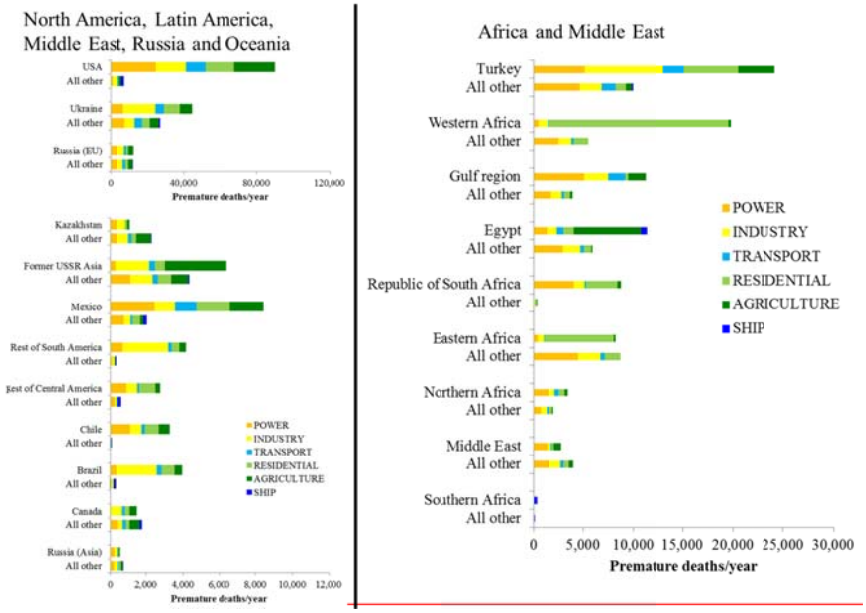
North America, Latin America, Middle East, Russia and Oceania
USA
All other
Ukraine
All other
Russia (EU)
All other
0
40,000
80,000
120,000
Premature deaths/year
Kazakhstan
All other
Former USSR Asia
All other
Mexico
All other
Rest of South America
All other
Rest of Central America
All other
Chile
All other
Brazil
All other
Canada
All other
Russia (Asia)
All other
POWER
INDUSTRY
TRANSPORT
RESIDENTIAL
AGRICULTURE
SHIP
0
2,000
4,000
6,000
8,000
10,000
12,000
Premature deaths/year
Africa and Middle East
Turkey
All other
Western Africa
All other
Gulf region
All other
Egypt
All other
Republic of South Africa
All other
Eastern Africa
All other
Northern Africa
All other
Middle East
All other
Southern Africa
All other
POWER
INDUSTRY
TRANSPORT
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SHIP
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5,000
10,000
15,000
20,000
25,000
30,000
Premature deaths/year

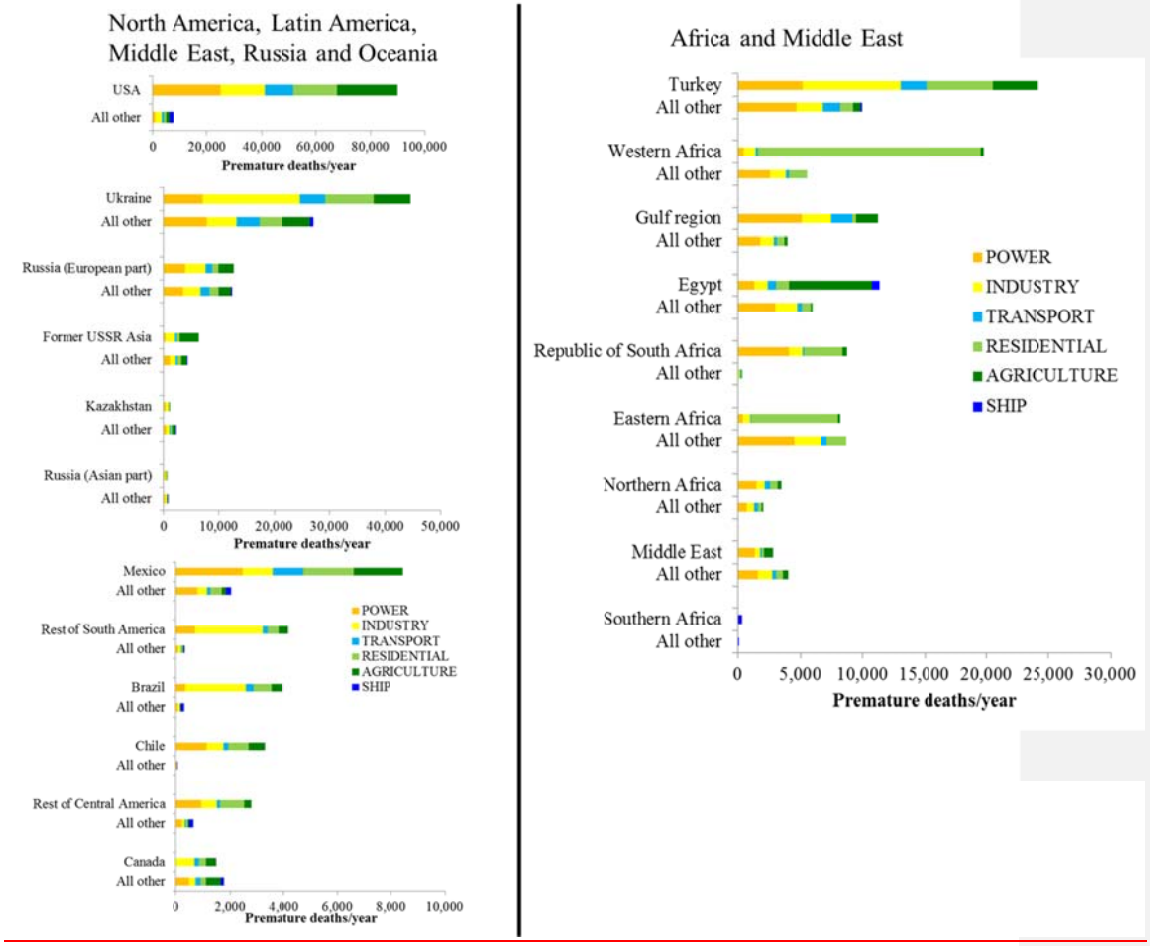

Figure 8b – Anthropogenic emission sector contributions to premature mortality (deaths/year) due to $PM_{2.5}$ population weighted concentrations in the TM5-FASST receptor regions of North America, Latin America, Russia, Middle East and Oceania (left hand side) and Africa (right hand side). Note that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

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