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.

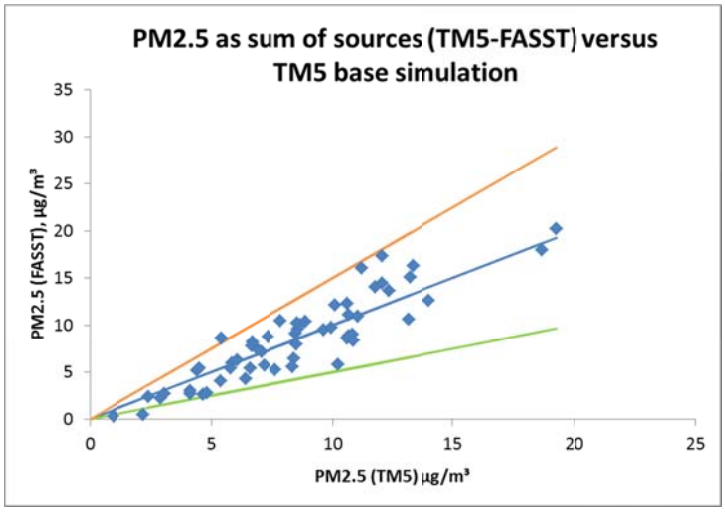
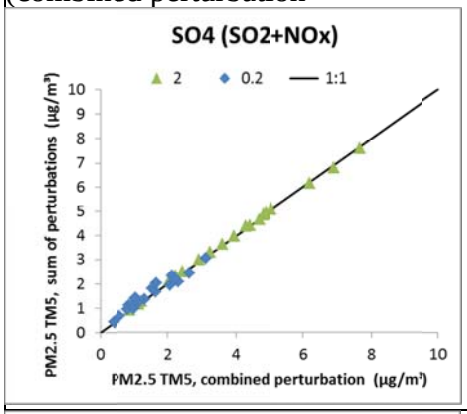
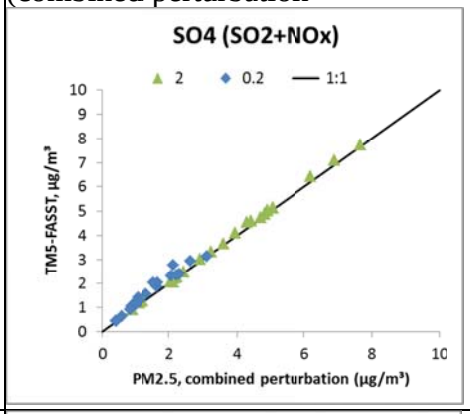
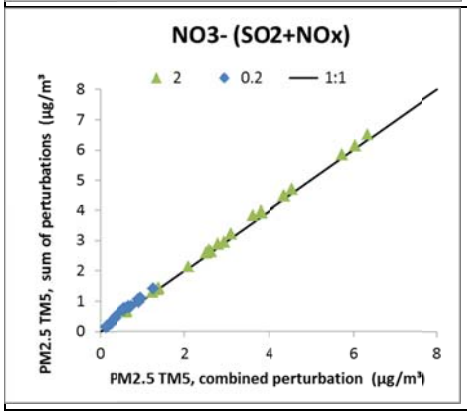
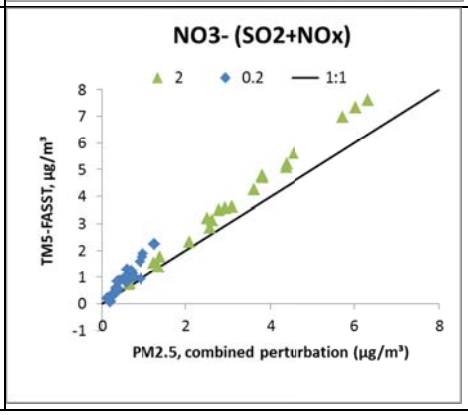


Figure A: Scatter plot of regionally averaged PM2.5 concentration (including all anthropogenic components) obtained as the sum of individual source region contribution by linear scaling of

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
	
	

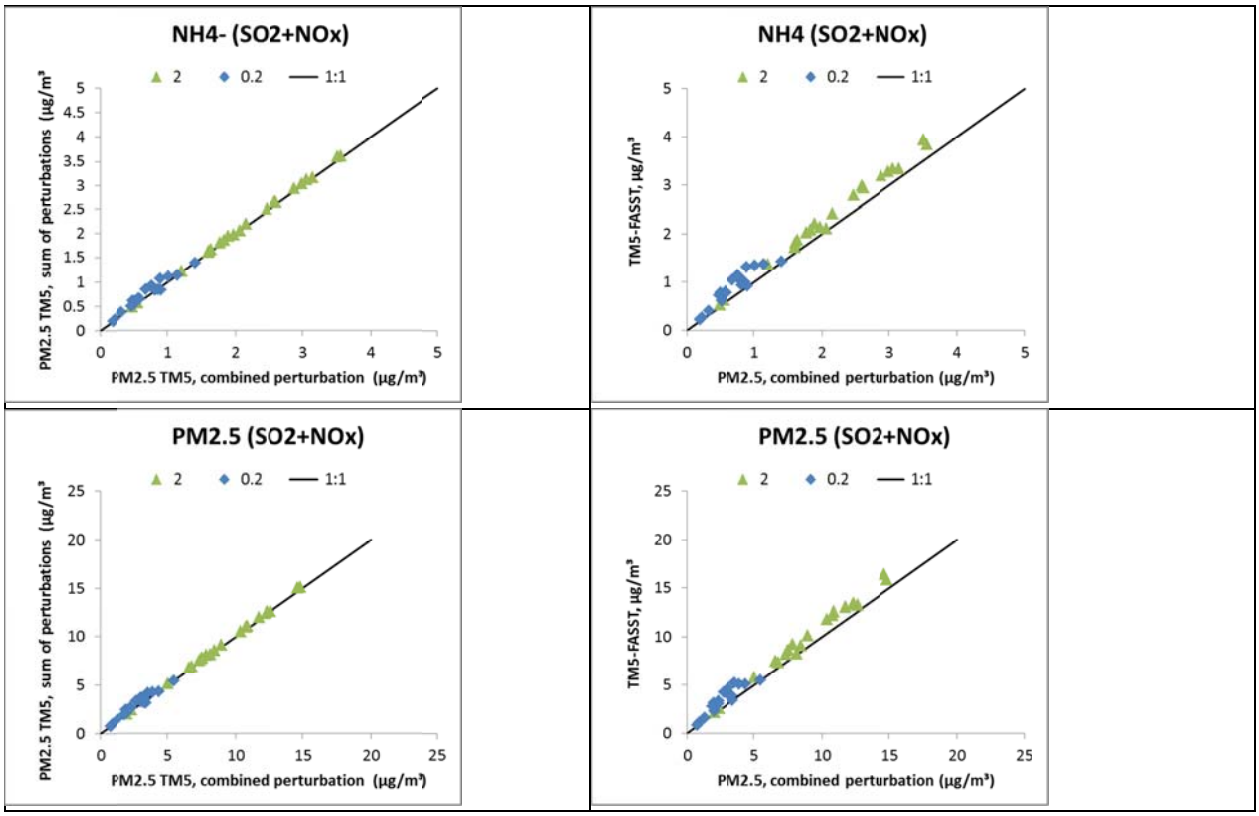


Figure 1. Additivity and linearity test of perturbations using TM5 outcome for regional population-weighted mean secondary inorganic PM2.5 concentrations for 3 perturbation magnitudes (green: +100%, red: -20%, blue: -80% relative to base simulation emissions). X-axis: simultaneous perturbation of SO2 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.

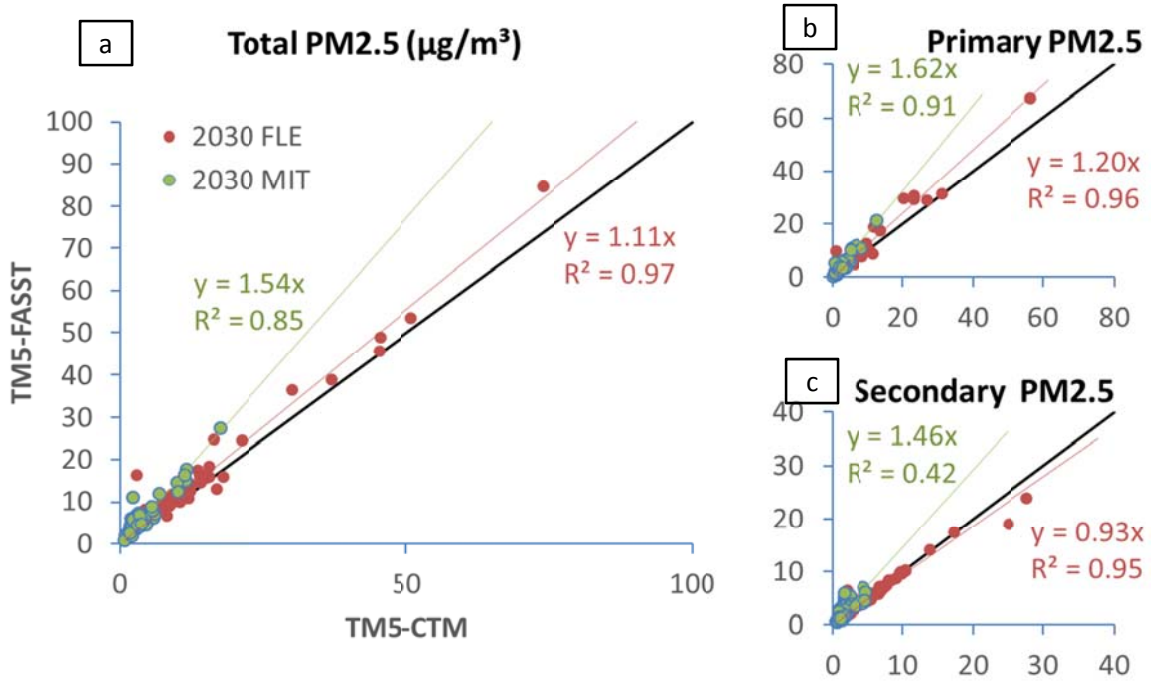


Figure 2. (a) PM2.5 concentration obtained with TM5-FASST versus TM5-CTM for high (FLE, red dots) and low (MIT, green dots) emission scenarios (see text). Each point represents the population-weighted mean over a TM5-FASST receptor region. Black line: 1:1 relation. breakdown for (b) primary (BC+POM+other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

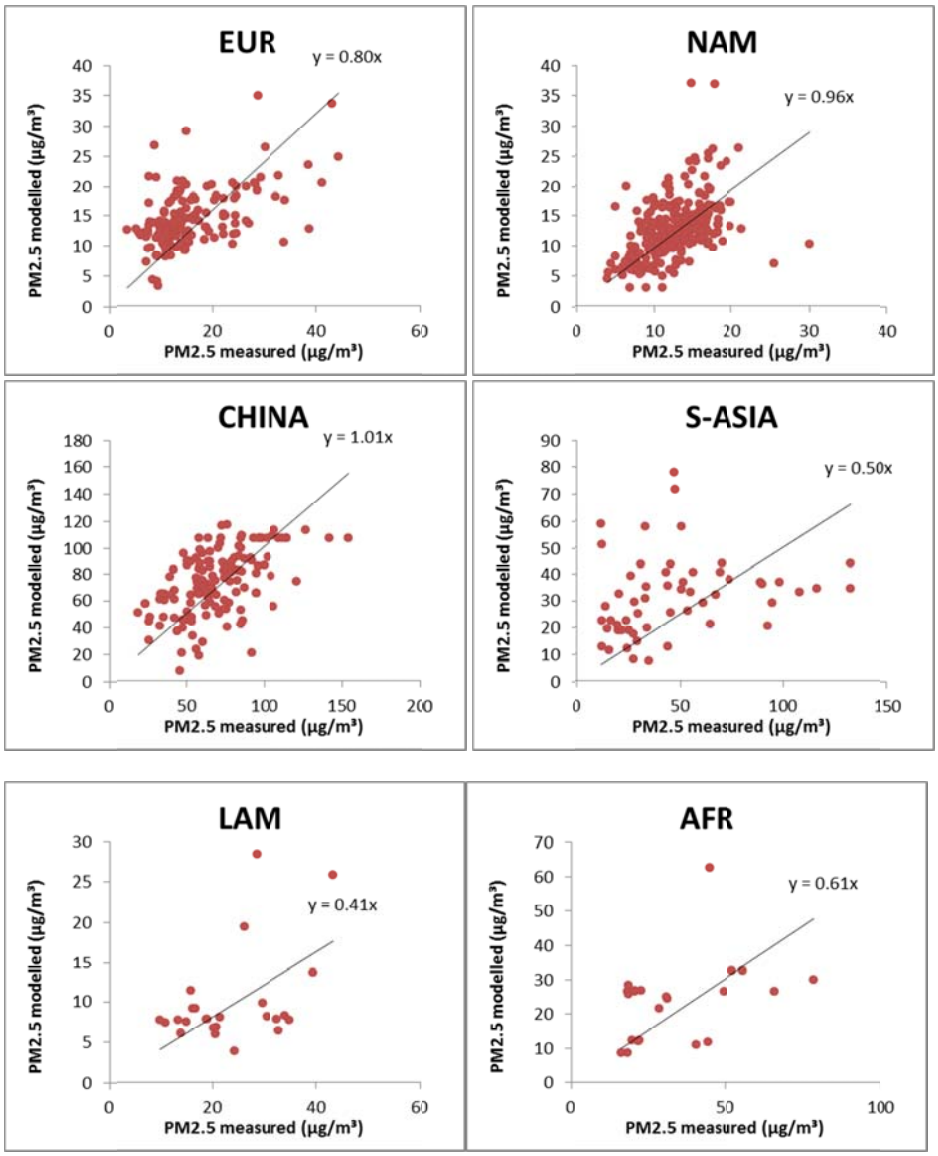


Figure 3 - TM5-FASST grid-cell mean (with urban increment parameterisation) versus individual monitoring stations (WHO consolidated database, including both measured and estimated PM2.5).

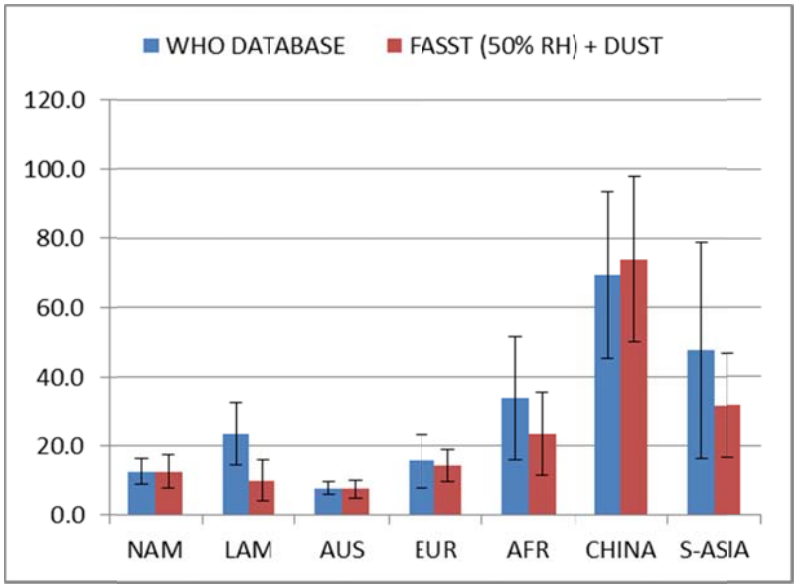
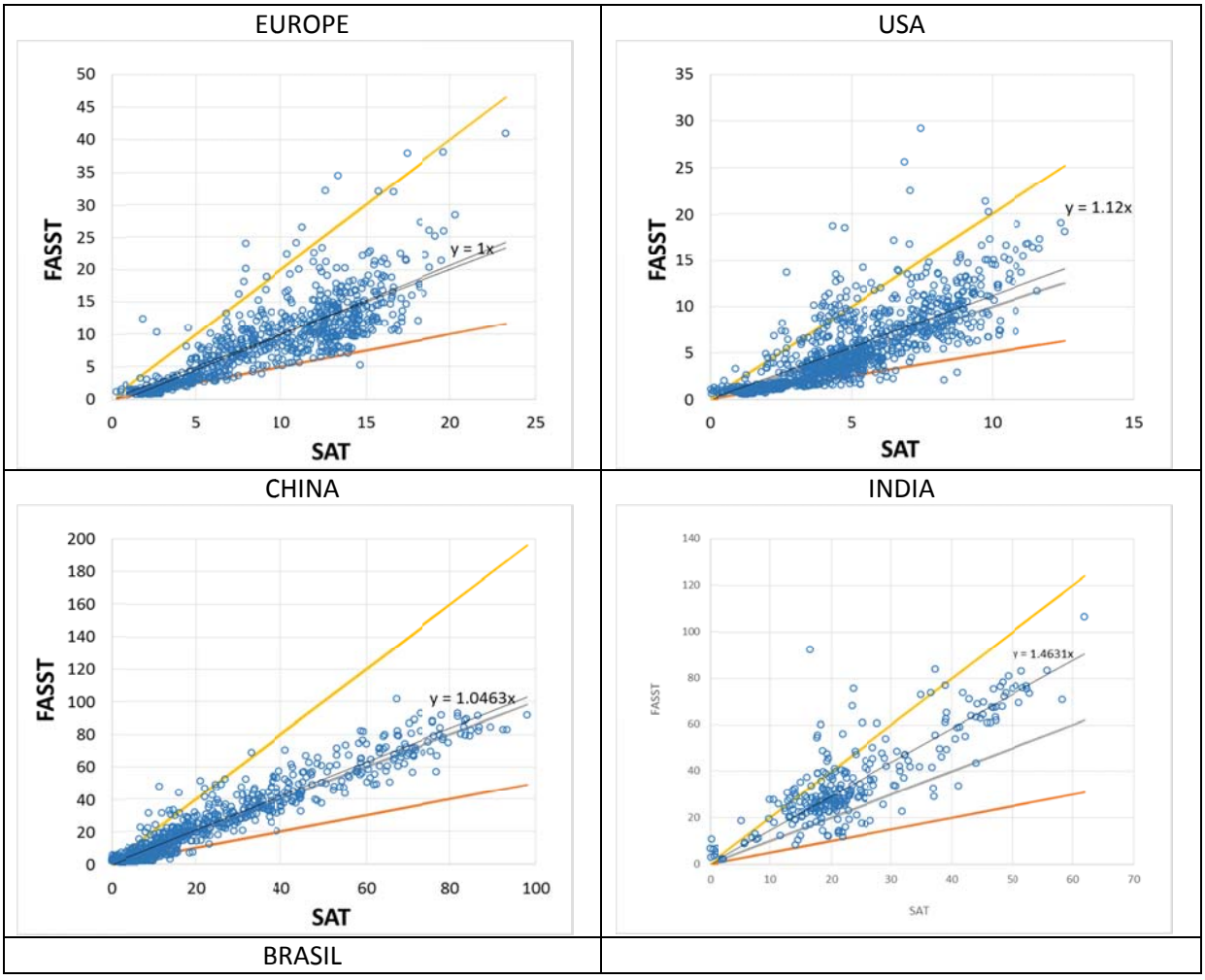


Figure 4 - WHO regional average of urban stations (+/- 1 stdev) and FASST population weighted average of grid cells.



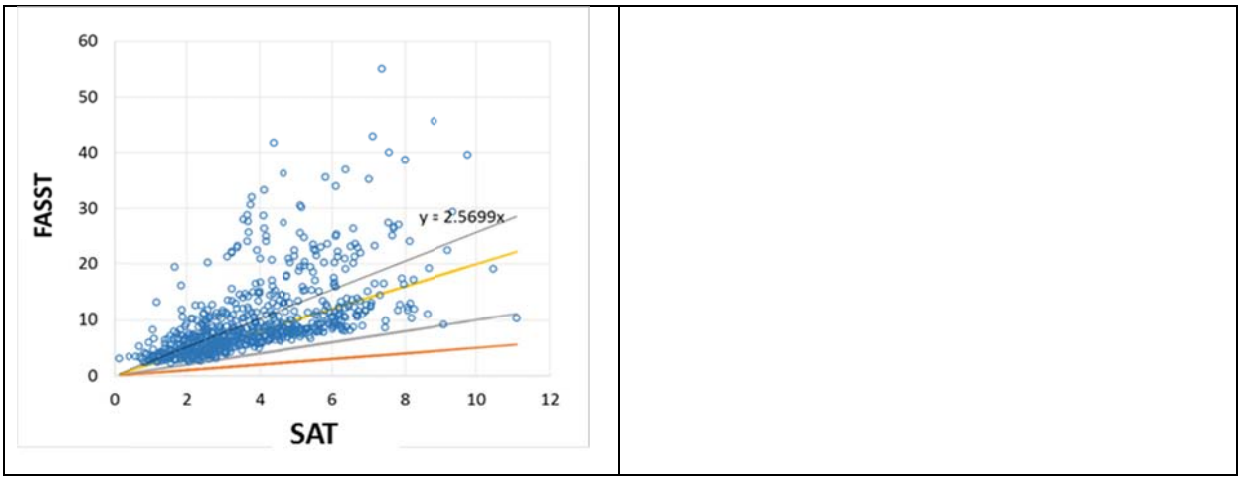


Figure 5 - Comparison between TM5-FASST and satellite products for world regions (Boys et al., 2014; van Donkelaar et al., 2010, 2014).

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).

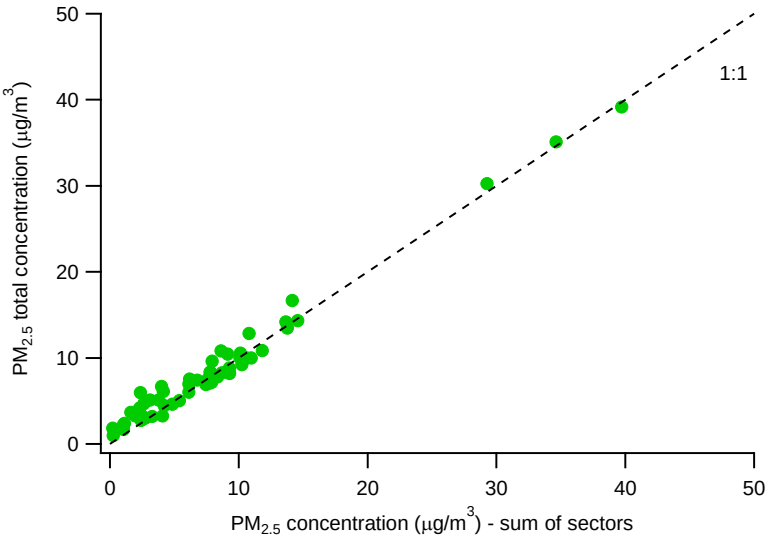


Figure 1 – Comparison between the total modeled PM2.5 concentration and the sum of the sectors.

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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