The authors are grateful to the Referee for these additional comments that helped in improving the manuscript. The paper by Van Dingenen et al. (submitted, 2018) about the TM5-FASST methodology is now accepted for final publication, therefore we hope that with the additional information provided below as well as the changes done in the manuscript will help in solving all the concerns.

The authors coupled the HTAP_v2.2 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. As I noted in my previous review, I find that what the paper is trying to do is important. However, I still find that the objective of the paper is unclear throughout the paper and I am not sure if this paper should be stand-alone or should be combined together with the Van Dingenen et al. (2018) paper that is currently under review for ACP. Most importantly, I do not understand the rationale behind quantifying health impacts from sectorial emissions, given that the uncertainty is so high.

First, the biggest problem I have with this paper is that there are significant underestimations of PM2.5 concentrations in many countries and to me, the linearity estimation for PM2.5 is not satisfactory. I am not convinced that there is new science in the paper and as one of the reviewers was suggesting, maybe this paper, combined with the Van Dingenen et al. (2018) paper should probably be moved to GMD to discuss potential of the new tool for assessing air quality and health impacts.

The paper by van Dingenen et al. (2018) is accepted for final publication in ACP. In our manuscript we clarify the role of our paper being an application of the TM5-FASST methodology.

"This work is an application of the TM5-FASST model, which is extensively documented in a companion publication in this special issue. Van Dingenen et al., (2018) provide an extensive evaluation of the model, model assumptions and performance with regard to linearity and additivity of concentration response to different size of emission perturbations and future emission scenarios."

Moreover, in the supplementary material we provide additional information on the assumptions of linearity as reported below:

## S1.2 – Sector and source region attribution using the TM5-FASST source-receptor relationships

### S1.2.1 - Attribution by sector

The TM5-FASST methodology uses a local perturbation approach in the vicinity of a reference simulation, where the total concentration of component (or metric) $j$ in receptor region $y$, resulting from emissions of all $n_i$ precursors $i$ in all $n_x$ source regions $x$, is obtained as a perturbation on the base-simulation concentration (Van Dingenen et al., 2018). Hence, the $PM_{2.5}$ concentration in region $y$ for an emission scenario different from the reference scenario is obtained as:

$$\mathrm{PM}(y) = PM_{ref}(y) + \Delta\mathrm{PM}(y) \quad (1)$$

The perturbation term $\Delta\mathrm{PM}(y)$ is obtained from the linear scaling of the difference between scenario and reference emission (i.e. the emission perturbation):

$$\Delta\mathrm{PM}(y) = \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} A_{ij}[x_k, y] \cdot [E_i(x_k) - E_{i,ref}(x_k)] \quad (2)$$

where the summation runs over $n_i$ precursor species, $n_j$ $PM_{2.5}$ components and $n_x$ source regions, and $A_{ij}[x_k, y]$ is the source-receptor coefficient, expressing the emission-concentration response sensitivity in the vicinity of the reference conditions, evaluated from a 20% emission perturbation (see Van Dingenen et al., 2018):

$$A_{ij}[x, y] = \frac{\Delta PM_{ref}^{j}(y)}{\Delta E_{i,ref}(x)} \quad (3)$$

with $\Delta E_{i,ref}(x)$=0.2$E_{i,ref}(x)$ and $\Delta PM_{ref}^{j}(y)$ the corresponding $PM_{2.5}$ component *j* response.

Eq. (2) can also be applied to attribute individual sector contributions to the pollutant concentration by setting the “emission perturbation” equal to the emission contribution of a single sector. The $PM_{2.5}$ contribution from the single sector S equals

$$\Delta\mathrm{PM'}_S(y) = \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} A_{ij}[x_k, y] \cdot [E_{S,i}(x_k)] \quad (4)$$

Having obtained the marginal $PM_{2.5}$ contributions from the individual sectors, the total $PM_{2.5}$ can be re-composed as the sum from all $n_S$ sectors S:

$$\mathrm{PM'}(y) = \sum_{s=1}^{n_S} \Delta\mathrm{PM'}_s(y) \quad (5)$$

However, due to non-linearities in emission-concentration responses, the sum of all individual sector contributions may not exactly match the total $PM_{2.5}$ obtained from Eqs. (1) and (2) where we write $E_i(x_k)$ as the sum of the emissions by sector:

$$\mathrm{PM}(y) = PM_{base}(y) + \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} A_{ij}[x_k, y] \cdot \left[\sum_{s=1}^{n_S} E_{s,i}(x_k) - E_{i,ref}(x_k)\right] \quad (6)$$

$\mathrm{PM'}(y)$ from Eq. 5 and $\mathrm{PM}(y)$ from Eq. 6 are equivalent if

$$PM_{ref}(y) = \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} A_{ij}[x_k, y] \cdot E_{i,\mathrm{ref}}(x_k) \quad (7)$$

Using Eq. 3 this is equivalent to the condition that

$$PM_{ref}(y) = \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} A_{ij}[x_k, y] \frac{\Delta PM(y)}{0.2E_{i,\mathrm{ref}}(x_k)} E_{i,ref}(x_k) \tag{8}$$

or

$$PM_{ref}(y) = \sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} 5.\Delta PM(y) \tag{9}$$

In other words, total $PM_{2.5}$ will be correctly reproduced as the sum of the individual sector contributions if and only if the $PM_{2.5}$ base concentration can be approached by 5 times the 20% perturbation response, implying a perfectly linear emission-concentration response for all precursors. Figure A1.1 shows the correspondence between regionally aggregated $\sum_{j=1}^{n_j} \sum_{k=1}^{n_x} \sum_{i=1}^{n_i} 5.\Delta PM$and $PM_{ref}$. The agreement is satisfactory although not perfect. In order to restore the closure between the total $PM_{2.5}$ and the sum of the sectors, we therefore rescale the sector contributions such that their sum corresponds to the total $PM_{2.5}$ obtained from the local perturbation calculation, i.e. we use the relative contribution by sector resulting from Eq. 5 and apply them onto the total $PM_{2.5}$ obtained from Eq. 6.

$$\Delta\mathrm{PM}_S(y) = \frac{\Delta\mathrm{PM}'_S(y)}{\sum_{S=1}^{n_S} \Delta\mathrm{PM}'_S(y)} \mathrm{PM}(y) \tag{10}$$

**S1.2.2 Attribution by source region**

The marginal contribution of an individual source regions ($x$) to the total $PM_{2.5}$ concentration in a given receptor region ($y$) is obtained (via Eq. 2) from

$$\Delta\mathrm{PM}'_x(y) = \sum_{j=1}^{n_j} \sum_{i=1}^{n_i} A_{ij}[x, y] \cdot E_i(x) \tag{11}$$

Similar as for the sector break-down, the emission perturbation has been replaced by an extrapolation of the SR coefficient over the total emission magnitude in a given source region, and non-linearities may lead to non-closure between the sum of all $\Delta\mathrm{PM}'_x(y)$ and total $PM_{2.5}$ obtained from the local perturbation as in Eqs. (1) and (2). In order to restore the closure we apply the same scaling procedure as in Eq. 10:

$$\Delta\mathrm{PM}_x(y) = \frac{\Delta\mathrm{PM}'_x(y)}{\sum_{k=1}^{n_k} \Delta\mathrm{PM}'_{x_k}(y)} \mathrm{PM}(y) \tag{12}$$

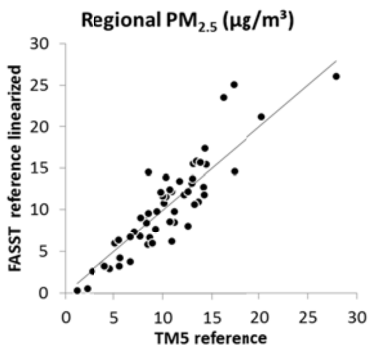


**Figure S1.2 - Scatter plot of regionally aggregated $PM_{2.5}$ concentrations. Y-axis: FASST linearized extrapolation of a 20% emission perturbation towards 100%, versus the full TM5 computation, for the FASST reference emission scenario (RCP year 2000, se Van Dingenen et al., 2018). The Figure evaluates the validity of Eq. 9.**

Second, the objective needs to be better defined. As for the two objectives of the study, there are two sentences in the manuscript: P. 2, l. 46 "The objective of this study is to evaluate the relevance of uncertainties in regional sectorial emission inventories, and their propagation in modelled PM2.5 concentrations and associated impacts on health."

We thank the Reviewer for the comment regarding the objectives of the paper and we clarified the objectives of the paper as described below.

We kept the sentence in the introduction (reported below) since it represents the key objective of the uncertainty analysis.

"The objective of this study is to evaluate the relevance of uncertainties in regional sectorial emission inventories, and their propagation in modelled PM2.5 concentrations and associated impacts on health."

P. 3, l. 15 "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 or regions." The two are very similar and I am not sure if the second objective is necessary.

We rephrased as following:

Based on our analysis on the importance of emission uncertainties at sector and regional level on $PM_{2.5}$, we aim at informing local, regional and hemispheric air quality policy makers on the potential impacts of sectors with larger uncertainties (e.g. residential and agriculture) or regions (e.g. developing and emerging countries).

On p. 4, l. 12, the aim of this work is explained "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." Furthermore, later in the text on p. 13 l. 38, the authors state, "[i]n our work we only evaluate how the uncertainty of emission inventories influences the health impact estimates focusing on the interregional aspects and not all the other sources of uncertainties." The authors should be consistent in what the objective and the aim of this work is throughout the paper.

We rephrased as following:

In the following, we will address the uncertainty of sector specific emissions from this inventory in a quantitative way as well as the differences we observe from one region to the other, based on the uncertainty of activity data and emission factors.

Third, the paper should have all the methodologies related to the objective in the paper. For example, if the objective of this paper is indeed on quantifying health impacts, I think the premature mortality calculation methodology should move from the Van Dingenen et al. (2018) to this paper and the crop damage should be taken out from this paper.

We agree with the Reviewer about the need of knowing the details of the methodology applied to estimate the health effects in TM5-FASST, as well as other methodological assumptions. In our manuscript we have summarized the key features the reader need to know in order to understand the results discussed in out manuscript. However, we cannot report all the details about the TM5-FASST methodologies which are extensively described both in the main text and in the supplementary material of the work by Van Dingenen et al. (2018). To help the reader in linking our manuscript with the paper by Van Dingenen et al. (2018), we added the information about the sections of the paper by Van Dingenen et al. (2018) where to find these methodological information. For example we now report that:

As described in Sect 2.5 and S5 of the paper by Van Dingenen et al. (2018), the mortality estimation in TM5-FASST is based on the integrated exposure-response functions defined by Burnett et al. (2014).

Fourth, the writing could be improved, as it is often difficult to follow, as described in minor comments below.

Minor comments:

1. P. 1, l. 29 Not sure what the authors mean by "improve emission inventories knowledge and air quality"

We corrected as following: "improve emission inventories knowledge and air quality modeling".

2. P. 2, l. 9 Not sure what the authors mean by “improve globally air quality and possibly human health”
   The sentence now reads:
   Local, regional and international coordination is therefore needed to define air pollution policies to improve globally air quality and possibly human health.

3. P. 2, l. 13-20 I am unsure what the authors mean in the two sentences.
   This paragraph aims at giving the context of air quality issues which are not only happing locally but also at regional and global scale. Then we focus on a short description of particulate matter composition and formation, being the compound we look at in this publication.

   “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 PM2.5 (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). 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 (SO2), nitrogen oxides (NOx) and ammonia (NH3), and organic compounds involving thousands of compounds and often poorly known reactions (Hallquist et al., 2009).”

4. P. 2, l. 35 414.000 à 414,000 or 414 thousand
   Changed to 414 thousand
5. P. 4, l. 28 “can be also applied also” à delete the second “also”

Change done as requested

6. P. 5 l. 19 “now day much more” à “now much more?”
   Change done as requested

7. P. 6 l. 10 Not sure what the authors mean by “24OECD90 countries”

A footnote has been inserted to identify the OECD countries in 1990:
OECD countries in 1990: Australia, Austria, Belgium, Canada, Denmark, Finland, France, Germany, Greece, Iceland, Ireland, Italy, Japan, Luxembourg, Netherlands, New Zealand, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, United Kingdom, United States.

8. P. 14 l. 25 How did the authors come up with a threshold value of 5.8 µg/m3?

The threshold value of 5.8 µg/m3 comes from literature (Anenberg et al., 2010) and it is fully described in the work by Van Dingenen et al. (2018).

References

Anenberg, S. C., Horowitz, L. W., Tong, D. Q. and West, J. J.: An estimate of the global burden of anthropogenic ozone and fine particulate matter on premature human mortality using atmospheric modeling, Environ. Health Perspect., 118(9), 1189–1195, doi:10.1289/ehp.0901220, 2010.

Van Dingenen, R., Dentener, F., Crippa, M., Leitao, J., Marmer, E., Rao, S., Solazzo, E., and Valentini, L.: TM5-FASST: a global atmospheric source-receptor model for rapid impact analysis of emission changes on air quality and short-lived climate pollutants, Atmos. Chem. Phys. Discuss., 2018, 1-55, 10.5194/acp-2018-112, 2018.