Attribution of anthropogenic $PM_{2.5}$ to economic sectors: a global analysis

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1. Introduction and scope

It has become increasingly clear that environmental impacts of anthropogenic air pollutants are among the most pressing problems for present and future generations. New estimates just released by the World Health Organization confirm (WHO) that air pollution is now the world's largest single environmental health risk [1]. In March 2014, WHO reported that in 2012 around 7 million premature deaths could be attributed to in- and outdoor air pollution, that is one in eight of total global deaths [2].

Over the last few years, increasing attention is devoted to the evaluation of the health impacts of local air pollution [3–7] .

Understanding the sources of these pollutants is crucial to identify cost-effective emission reduction measures. Particulate matter, especially those particles with a diameter up to 2.5 microns ($PM_{2.5}$), is of particular concern because it penetrates deep into the respiratory system. Epidemiological studies consistently show increased mortality risk from exposure to $PM_{2.5}$. [8,9]

$PM_{2.5}$ is usually a complex mixture of various chemical components, emitted by different multi-pollutant sources. Primary $PM_{2.5}$ results from direct emissions of carbonaceous particles (elemental carbon, organic carbon) from combustion processes, but also from re-suspension of road dust, tire and brake wear, and agricultural sources. Among carbonaceous components, particular interest is dedicated to Black Carbon (BC) and Organic Carbon (OC). Black carbon (BC) particles are major components of emissions from fossil fuel combustion and biomass burning. Arguably, BC is one of the most important components causing global warming (IPCC; 2013) OC is directly emitted from primary sources or produced from chemical reactions involving gaseous organic precursors.[10,11]

BC, together with OC, is also an important component of diesel exhaust emissions and, has been associated with various adverse health effects due to its polycyclic aromatic hydrocarbons components (PAHs).

Gaseous pollutants, like sulfur and nitrogen oxides, drive the formation of inorganic secondary $PM_{2.5}$ (Secondary Inorganic Aerosols, SIA) in the form of nitrate and sulfate salts, through a successive photo-chemical reactions in air.

Whereas the formation of SIA in urban and regional pollution episodes is clearly associated with areas of high fossil-fuel combustion and anthropogenic emissions, secondary species have a sufficient lifetime to be distributed over a larger geographical area, thus having not only local and regional impacts, but global ones as well [12].

Transformation of organic species is also responsible for the formation of secondary organic aerosol (SOA). These compounds represent a major fraction of atmospheric particulates. Organic aerosols can form in a variety of reactions, with different environments around the world presenting variable conditions (temperature, humidity, and sunlight), precursors emissions (biogenic and anthropogenic volatile organic compounds, VOCs) and oxidants levels (ozone and radicals).

In this study, we use a global emission inventory and a source-receptor model for air pollutant transport to quantify the respective contribution of the different anthropogenic emission sources (from major human activity sectors) to $PM_{2.5}$. This information reveals the potential to assess the effectiveness of air quality directives addressing different human activities and even more importantly guidance for targeted and efficient air quality policies.

Specifically, a global compilation for 2010 of national and regional emission inventories (HTAP_v2) has been used to calculate total emissions for each country, and major economic sectors representative for the most important air pollution sources worldwide.

Present day and future emissions are ingested by source receptor model (TM5-FASST) calculating concentration attributed by economic sector. To verify the accuracy of the computation, a global Source Apportionment (SA) database has been compiled with $PM_{2.5}$ attributed to main pollution sources. SA data have covered 24 years of field measurements (1990-2014) dealing with particle sampling and post-analysis. In this work, we attempted to compare gridded data (1°x1°) from a source receptor model with point measurements.

To our knowledge, this is the first work that compares sector attributed modelled $PM_{2.5}$ concentrations with worldwide $PM_{2.5}$ source apportionment data at global scale.

2. The global air quality model TM5-FASST

For this study we apply the global chemical transport model TM5-FASST (FAst Scenario Screening Tool).[13,14] to compute spatially resolved $PM_{2.5}$ concentrations attributed by sectors. TM5-FASST is a reduced-form SRM (source receptor model): the relation between the emissions of a compound i from a source x in region S and the resulting pollutant j concentration (where $j = i$ in case of a primary component) at a receptor y_R in regions R is expressed by a simple functional relation which mimics the underlying meteorological and chemical processes. In the current version of TM5-FASST, the function is a simple linear relation:

$$\sum_R C_{i\rightarrow j,y_R,x} = C_0 + \sum_R A_{i\rightarrow j,x,y_R} E_{i,x} \quad (1)$$

Where $C_{i\rightarrow j,y_R,x}$ is the concentration of species *j* at the receptor region y_R formed from precursor *i* emitted at source *x*, $E_{i,x}$ is the emission rate (kg/yr) of precursor *i* at source *x*, $A_{i\rightarrow j,x,y_R}$ is the so-called source-receptor coefficient (SRC) between source location *x* and receptor region y_R for emitted precursor *i* leading to end product *j,* and C_0 is a constant. We note that this equation implicitly includes the vertical dimension, through the emission height assigned to specific emission sources [Dentener et al., 2006]. E.g SO2 emissions, dominated by industrial and power generation, are typically emitted at a few hundred meters altitude. NOx emissions from ground transport is emitted in the first model layer (ca. 60 meters).

The source-receptor coefficients are stored as matrices with dimension [x,y]and are available for each precursor and for each resulting component from that precursor. The SRCs have been derived from a set of runs with the full chemical transport model TM5-CTM (Krol et al., 2005; Van Dingenen 2015) by performing a set of emission perturbation runs for a defined set of source regions and precursor components compared to a base run. TM5-CTM explicitly solves the mass balance equations of the species using detailed meteorological fields and sophisticated physical and chemical process schemes. TM5-CTM covers the global domain with a resolution of 1°x1°. In particular, the applied procedure to calculate the SRCs was the following:

- 56 source regions were defined (see Figure 1) covering the global continents. Source receptor coefficients were also calculated between global international shipping and aviation as sources, and the global grid as receptor. Definition of The 56 source regions and their aggregration to 23 larger regions is reported in Appendix 1.
- 360 x 180 receptor regions were defined at a global 1° x 1° resolution. A reference run with a reference global emission dataset for all relevant air pollutants for the year 2000 was performed, including sulfate dioxide (SO_2), nitrogen dioxide (NO_x,), BC, OC (or primary organic matter POM), non-methane VOCs (NMVOC), and ammonia (NH_3). This run produces concentrations ofparticulate matter ($PM_{2.5}$) components (SO_4, NO_3, NH_4, BC, particulate organic matter – POM), trace gases (SO_2, NO, NO_2, NH_3, O_3, CO), and deposition fluxes of BC, N and S species at a global 1°x1° resolution.
- A series of perturbation runs were performed, where sequentially in each of the defined 56 source regions, pollutant emissions were reduced over the entire source region by 20% relative to the reference run. The resulting concentrations were then calculated as done for the reference run. The difference between the concentration field for a specific compound from each perturbation run and the reference run is a global 360x180 concentration field (1°x1° resolution), the so-called delta-field. Additional to the 56 continental source regions, separate perturbation runs were performed for aggregated international shipping emissions (occurring over the oceans) and for aviation, so that in total 58 source 'regions' are available.

Hence, the total concentration of component *j* in receptor region y_R, resulting from arbitrary emissions $E_i(x)$ of *all* its precursors *i* at *all* source regions *x* from sector k (characterized with either stack height or surface height) is obtained by overlaying emission-scaled delta fields $\Delta C_{i\rightarrow j,x,y_R}$ for all source regions (scaled on the 20% perturbation):

$$C_{i\to j}(\boldsymbol{y_R}) = \sum_{\boldsymbol{R}} C_{j,base}(\boldsymbol{y_R}) + \sum_{\boldsymbol{R}} \sum_{\boldsymbol{x}} \sum_{\boldsymbol{i}} \Delta C_{i\to j,x,y_R} \cdot \left[\frac{E_i(x)-E_{i,base}(x)}{0.2E_{i,base}(x)}\right] \quad (2)$$

Linear equations (2) with associated source-receptor matrices (SRC) are the 'kernel' of TM5-FASST.
Although input emission data are provided as regions or country aggregates, TM5-FASST implies an underlying gridding of the emissions (i.e. the emission fields used in the reference year 2000 run). Further, source-receptor relations are based on a fixed meteorological field (year 2001 meteorology). Natural $PM_{2.5}$ components (mineral dust, sea-salt) are not modelled with TM5-FASST. Instead we use fixed, pre-calculated sea-salt fields from the full TM5 model and fixed dust fields which have been calculated with the dust emission model by Ginoux from the year 2000 and with meteorology for 2001 [15]. Dust is mainly responsible for country-averaged exposure levels above 50 $\mu g/m^3$. Therefore, the performance of the model in the high $PM_{2.5}$ range end (above 120 $\mu g/m^3$) is linked to the performance of the dust model.

The TM5-FASST tool produces as output global gridded concentrations at 1° x 1° resolution of all $PM_{2.5}$ chemical components, which can be aggregated to country-population-weighted-mean $PM_{2.5}$ concentrations. Residual water is also included in the calculation of the total $PM_{2.5}$, assuming a value of 50% for relative humidity (RH), which is the humidity prescribed for the $PM_{2.5}$ mass measurement protocol in Europe.

When the emissions breakdown by sector is provided, the contribution of each individual sector to $PM_{2.5}$ concentration ($[\mathrm{PM_{2.5}(sector)_i}]'$) is evaluated by making the difference between a run with total (all sector) emissions, and a run with the sector of interest excluded from the total emissions:

$$[PM_{2.5}(sector)_i]' = [Total\ PM_{2.5}] - [Total\ PM_{2.5} - PM_{2.5}(sector)_i] \quad (3)$$

The total $PM_{2.5}$ concentration includes the anthropogenic part, modelled from the full anthropogenic emission set, plus the natural components (mineral dust and sea salt), which are equally treated as a 'sector' contribution. Calculations of PM2.5 in the parent TM5 model, would include non-linear interaction with oxidants; e.g. the oxidation of SO_2, as well as non-linear formation of NH_4NO_3. Other PM2.5 components, like primary BC, POM, sea salt and dust are by construction behaving fully linear. Also in this particular version of TM5, secondary organic aerosol

Because of the linearized approach in TM5-FASST by which $PM_{2.5}$ concentrations are calculated from a linear perturbation of a reference emission-concentration set (Equation 2), the sum over all sectors of the $[PM_{2.5}(sector)_i]$ does not exactly match $[Total\ PM_{2.5}]$. Indeed, emission changes exceed in most case the 20% emission perturbation that was applied for the calculation of the linearized emission-concentration relations. Therefore, the $PM_{2.5}$ concentrations are in most cases slightly underestimated (how much, is this for grid-points or countries) both for increasing and decreasing emissions, as consequence of the linearization of non-linear chemical processes.
We 'calibrate' with a factor R_i the sectorial contributions as follows:

$$[PM_{2.5}(sector)_i] = R_i[Total\ PM_{2.5}] \quad (4)$$

$$R_i = \frac{[PM_{2.5}(sector)_i]'}{\sum_i [PM_{2.5}(sector)_i]'} \quad (5)$$

R values have been calculated per gridpoint. Globally, R ranges from 0 to 0.84 with an average value of 0.17.

With individual sector emissions available, we estimated the $PM_{2.5}$ concentrations of sectors Transport, Residential, Agriculture, Industry, Energy and Biomass Burning, as well as the contribution of natural sources (dust + seas salt).

The native resolution of the TM5-FASST model corresponds to a nominal grid size of 1x1 degree (ca. 100 km x 100 km) and hence calculated $PM_{2.5}$ concentration are obtained as grid cell averages. Population exposure estimates have been improved by taking into account sub-grid pollutant gradients which are mostly driven by population density gradients. We applied a parameterization to reconstruct $PM_{2.5}$ sub-grid gradients within the 1° x 1° grid cell to a resolution of 0.125° x 0.125° (64 sub-grid cells), as described by Rao et al., 2012. The final product are gridded data at a 0.125° x 0.125° resolution with adjusted (urban-incremented) $PM_{2.5}$ concentration, which conserves however the regional native 1° x 1° $PM_{2.5}$ concentration. This methodology is described in the appendix (A1).

For health impact evaluation, a relevant metric is the population-weighted mean $PM_{2.5}$ by country. The population-weighted $PM_{2.5}$ average has been estimated for each Country and pollution sector using the population statistics for the year 2010 together with the $PM_{2.5}$ concentration for each sector at the resolution of 0.125° x 0.125°.
Sector specific $PM_{2.5}$concentrations are population-weighted according to the following equation:

$$(Population\ Weighted\ PM_{2.5})_i = \frac{\sum_{i,j}[PM_{2.5}(sector)_i] \times N(population)_j}{\sum_j N(population)_j} \quad (6)$$

where $N(population)_j$ is the population number of country or region *j* for sector *i*.

Population statistics for the year 2010 has been calculated using the Gridded Population of the World (GPWv3) released by the Centre for International Earth Science Network (CIESIN)[16] depicts the distribution of human population across the globe and is intended to demonstrate the spatial relationship of human populations and the environment across the globe. The gridded data set is largely constructed from national or subnational administrative units.

3. Air Pollution emissions

3.1. The EDGAR-HTAP V2 emission inventory

The focus of this study is the attribution of $PM_{2.5}$ concentrations to the contributing emitting sectors at global scale, with a consistent methodology across all regions, and based on a global up-to-date emission inventory that contains the required sectorial detail. The sector-separated emissions for this study are obtained from the

Hemispheric Transport of Air Pollution (HTAP V2) harmonized emissions database for the year 2010 [17]. The HTAP V2 dataset consists of 0.1° x 0.1° emission grid-maps of CO, SO_2, NO_x, NMVOC, NH_3, PM_{10}, $PM_{2.5}$, BC and POM for the years 2008 and 2010 [18,19]. This dataset uses nationally reported emissions combined with regional scientific inventories in the format of sector-specific grid-maps. HTAPv2 grid-maps have been harmonized and aggregated to provide total emission for each world country and activity. The grid-maps are complemented with the Emission Database for Global Atmospheric Research (EDGARv4.3) gridmaps for those regions where no other well-accepted regional data source is available. The global grid-map results from the cooperation of US-EPA, EPA-Canada, the Model Inter-comparison Study Asia (MICS-Asia group), EMEP/TNO Europe, the Regional Emission inventory for Asia (REAS) and the JRC EDGAR group. The primary objective is to serve the scientific community for hemispheric transport of air pollution.

The HTAP V2 dataset provides Total Emissions (kg/Year) by country and activity sector for the year 2010. The main pollutant sectors of interest are:

- **Air** (international and domestic aviation)
- **Shipping** (international shipping)
- **Energy** (power plant industry)
- **Industry** (manufacturing, mining, metal cement, solvent industry)
- **Transport** (ground transport including road, rail, pipeline, inland waterways). All types of fuels are included (including biofuels with short cycle C). Re-suspended dust from road transport is not included in $PM_{2.5}$ emissions. .
- **Residential** (heating/cooling of buildings and equipment/lighting of buildings and waste treatment)
- **Agriculture** (agriculture but not agricultural waste burning). NH_3 is the main chemical compound for this sector.
- **Additional to HTAP_v2: Biomass Burning** includes agricultural (FAOSTAT) waste burning and biomass burning from the Global Fire Emissions Database, version 3 (GFED3) [20].

Absolute and relative 2010 emissions of the main pollutant precursors for $PM_{2.5}$ concentrations are shown in Figure 2 and 2a. For the year 2010, the largest concentration of pollutant is represented by CO with 1000 Tg/year, followed by NMVOCs with 150 Tg/year, NOx with 135 Tg/year, SO_2 with 110 Tg/year, $PM_{2.5}$ with 85 Tg/year, NH3 with 50 Tg/year, POM with 44 Tg/year and, BC with 8 Tg/year (Figure 2).
Since the original TM5 calculations explicitly considered different heights of emissions [Dentener et al., 2006], the resulting source-receptor receptor calculations are mostly representative for the dominating sources of specific components in the original emission inventory; e.g. SO2 industry/power; BC transport in most regions, but biomass burning in other regions.
Relative contributions of the above pollutants to the sector contributions are shown in Figure 2a. Main findings showed that:

- Transport sector contributes to 31% NOx, 16% CO and 12% BC emissions. Worldwide?
- Energy sector contributes to 41% SO_2 and 21% NOx emissions.

- Industry sector contributes to 44% NMVOCs, 39% SO2, 17% NOx, 14% PM2.5 and CO, and 12% BC emissions.
- Agriculture sector mostly contributes to 85% NH3 emissions.
- Residential activities are responsible of 42% BC, 37% PM2.5, 30% POM (OC), 28% (NMVOCs) and 24% CO emissions.
- Biomass Burning contributes to 63% POM, 44% CO, 36% PM2.5 and, 32% BC emissions.

3.2. Validation with $PM_{2.5}$ source apportionment data

We carried out a comparison of the TM5-FASST modelled source attribution making use of Source Apportionment data estimated from measured $PM_{2.5}$ mass concentrations.

Source Apportionment (SA) is the identification of ambient air "pollution sources" and the quantification of their contribution to pollution levels [21]. This task can be reached using different methods: emission inventories, source-oriented models and receptor-oriented models (RMs). These (latter?) models have the advantage of providing information derived from real-world measurements. However, their applicability to very reactive species is limited. Receptor models (RM) are mostly used for source contribution estimation at local and regional level all over the world. A RM apportions the measured mass of an atmospheric pollutant at a given site to its emission sources by solving the mass balance equation:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \qquad (7)$$

where x_{ij} is the concentration of the j^{th} species in the i^{th} sample, g_{ik} is the contribution of k^{th} source to i^{th} sample, f_{kj} is the concentration of the j^{th} species in the k^{th} source, and e_{ij} is the residual for each sample/species.

RMs are commonly used to apportion Particulate Matter (PM) concentration on the basis of its chemical composition: major ions, carbonaceous fractions, trace elements and organic markers. From the physic-chemical analysis of $PM_{2.5}$ components, it is possible to estimate the pollution sources attributed to the total $PM_{2.5}$ mass, along with associated uncertainties.

4. Results

4.1. Sector specific contribution to total $PM_{2.5}$ concentrations

We report the main findings obtained from the TM5-FASST model using the EDGAR-HTAPv2 emission inventory. Population-weighted averages of the $PM_{2.5}$ concentration fields has been carried out for all countries and are visualized in Figure 3 to 9a. Further population-weighted averaging has been carried out for 23 larger regions that are representative for the entire world (Figure 10 and 10a). Raw data by Country are reported in Table T5 in the appendix.

Figures 3 to 9a show global maps of population-weighted $PM_{2.5}$ concentration attributed to each of the contributing sectors (Transport, Energy, Industry, Agriculture, Residential activities and Biomass Burning), and

their relative contribution to Total $PM_{2.5}$. Relative contributions to Total $PM_{2.5}$ are intended to highlights the importance of a specific contribution sector over certain geographical regions and where most of the population live. These values have been calculated from 0.125° x 0.125° gridded $PM_{2.5}$ output concentrations from TM5-FASST runs.

As shown in Figure 3, globally total anthropogenic and natural $PM_{2.5}$ concentration showed the highest country average concentration in the China (55 $\mu g/m^3$) and India (51 $\mu g/m^3$) region followed by Western Africa (28 $\mu g/m^3$) and the Korea region (24 $\mu g/m^3$). The lowest total $PM_{2.5}$ concentration values was found in Oceania (3 $\mu g/m^3$), followed by Central America and Mexico with values of about 9 $\mu g/m^3$.
In the following we report the main average contribution of the modelled PM2.5 concentrations according to pollution sector:

1) The highest $PM_{2.5}$ concentrations associated with the transport sector are observed in the India (5 $\mu g/m^3$) and China (3.5 $\mu g/m^3$) regions followed by Korea region (3.2 $\mu g/m^3$) and Western Europe (2.9 $\mu g/m^3$) (Figure 4 and 10). Instead, the largest relative contribution of this sector to Total $PM_{2.5}$ is found in Western Europe (18%) and Japan (17%) followed by Korea region (13%) and Middle East (12%) (Figure 4a and 10a). We must keep in mind that for large countries like China and India, 'average' concentrations are somewhat meaningless.
2) Industry $PM_{2.5}$ concentration shows the highest values in the China region (21 $\mu g/m^3$), India and Korea region (9 $\mu g/m^3$) followed by the Ukraine region and South Eastern Asia (4 $\mu g/m^3$) (Figure 5 and 10). Instead the Industry relative contribution to Total $PM_{2.5}$ shows high values spots in China (38%), Korea region (37%) , Oceania (32%) followed by Ukraine, Kazakhstan region and Turkey (25%) (Figure 5a and 10a).
3) The highest concentrations of $PM_{2.5}$ for the Energy sector are observed in the India (10.5 $\mu g/m^3$) and China (8 $\mu g/m^3$) regions followed by Middle East ((5.4 $\mu g/m^3$) and Turkey (4.5 $\mu g/m^3$) (Figure 6 and 10). Energy relative contribution to Total $PM_{2.5}$ shows high values in Middle East (39%), Turkey (28%), Northern Africa (27%) and Mexico (26%) (Figure 6a and 10a).
4) Agriculture has highest $PM_{2.5}$ values in the China region (6.2 $\mu g/m^3$) and in Western Europe (5.2 $\mu g/m^3$) followed by Central Europe (4.8 $\mu g/m^3$) (Figure 7 and 10). High Agriculture relative contribution to Total $PM_{2.5}$ is observed in the Kazakhstan region (36%), Western Europe (33%) Northern Africa (32%) and Japan (28%) (Figure 7a and 10a).
5) Residential activities, such as heating and cooking, show the highest $PM_{2.5}$ concentrations in the India (21.3 $\mu g/m^3$), and China (14.7 $\mu g/m^3$) region followed by Western Africa (11.4 $\mu g/m^3$) (Figure 8 and 10). Instead, the Residential relative contribution to Total $PM_{2.5}$ shows in the India region (42%), Western Africa (41%), Central America (38%) and Eastern Africa (35%) (Figure 8a and 10a).
6) Biomass Burning is strongly related to seasonal fires. High $PM_{2.5}$ concentrations are observed in Western Africa and Brazil (6 $\mu g/m^3$), followed by South East Asia (4.8 $\mu g/m^3$) and Southern Africa

(4.6 μg/m^3) (Figure 9 and 10). The largest relative contributions to Total $PM_{2.5}$ is observed in Brazil (51%) followed by Eastern and Southern Africa (29%), rest of South America (28%), Canada and South Eastern Asia (27%) and, Russia (25%) (Figure 9a and 10a).

The above findings are represented and important information for exposure studies to ambient air pollution. Recent studies showed that exposure to ambient air pollution is higher than previously thought, given the new evidence on exposure-risk information and also better global exposure estimates.[7,22,23]

Much of the exposure leading to those health impacts is believed to occur in cities, due to the higher concentration of human activities and their emissions to the air. Because of growing population numbers, increasing urbanization, and economic growth, those health impacts may even get worse in much of the world. Initial analyses are confirming this trend.[22,24] Many human activities contributing to air pollution are further contributing to climate change, and are therefore associated with yet additional health impacts.[25]

In order to take action to reduce health impacts, and hence exposure to air pollutants, it is essential to know the sources and activities contributing to local levels of pollution. For this reason, an increasing number of local studies on the contribution of sources to air pollution levels has been developed, most often at city level. Such studies generally consider various pollutants, and large groups of human and natural sources of pollutants, such as transport, industrial activities, biomass burning/residential activities, re-suspended dust, sea salt and other unspecified pollution sources of human origin.

5. Validation of results against measurements and Source Apportionment

5.1. Comparison of modeled Total $PM_{2.5}$ concentrations with the WHO database

$PM_{2.5}$ concentration by pollution sector obtained the TM5-FASST runs were compared with the $PM_{2.5}$ concentration data reported in the WHO dataset released on 2014 [26]. The 2014 version of the WHO Ambient Air Pollution dataset consists of urban air quality data with annual means of $PM_{2.5}$ for about 1600 cities from 91 countries for the years 2008-2013, and is therefore comparable to the emission dataset representative for 2010. The primary sources of data include publicly available national/subnational reports and web sites, regional networks such as the Asian Clean Air Initiative and the European Airbase, and selected publications.[1]
However, comparing models and measurements data we might consider that:

a) WHO $PM_{2.5}$ concentrations come from ambient measurements and therefore include secondary organic aerosols (SOA) from anthropogenic and biogenic origin. Only biogenic SOA has been highly parametrized modelled in the TM5-FASST with a global pseudo-source strength of about 19 Tg. Therefore, modelled $PM_{2.5}$ concentration could be underestimated in some regions when compared with real ambient $PM_{2.5}$ measurements.

b) WHO PM2.5 concentrations are from monitoring stations located at different type of sampling sites such as, traffic, urban background and residential.

Gridded Total $PM_{2.5}$ concentration values from TM5-FASST have been visualized with a GIS tool at the closest geographical coordinates of the WHO cities (Figure 11 and 11a).

Both measured (WHO) and modeled (TM5-FASST) $PM_{2.5}$ concentrations show values up to 20-25 $\mu g/m^3$ in the USA and Western Europe. Instead, values up to 80 $\mu g/m^3$ can be observed in the India and China regions from both measurement and modeled $PM_{2.5}$ concentrations.

World's average $PM_{2.5}$ concentration is about 21.36 $\mu g/m^3$ and 20.56 $\mu g/m^3$ for WHO and TM5-FASST, respectively. Instead, World's maximum $PM_{2.5}$ concentration is 152.60 $\mu g/m^3$ and 142.0 $\mu g/m^3$ for WHO and TM5-FASST, respectively. These latter values are found in India and China, respectively.

However, it should be pointed out that, in this comparison, TM5-FASST values correspond to an average of a 0.125° x 0.125 area, whereas the WHO data are point measurements. Therefore, point-to point correspondence cannot be expected.

Quantitative comparison of the statistical distribution between the WHO and TM5-FASST Total $PM_{2.5}$ concentrations has been carried out for 23 larger regions. As shown in Figure 11a, for regions such as Western Europe and USA, the median value of $PM_{2.5}$ concentration from WHO and TM5-FASST is quite close. There are several values classified as outliers by the statistical tool used for this analysis. Outliers have been as values beyond 1.5 times the interquartile range of the whole values for a specific region. Therefore, the presence of outliers does not always allow good correlation when comparing both set of data from WHO and TM5-FASST. For regions such as South East Asia, Korea, India, Canada, Brazil, Southern and Western Africa, the median value of $PM_{2.5}$ concentration shows good agreement between WHO and TM5-FASST data. However, for these regions, the range of values in WHO is larger than the one found from TM5-FASST calculations.

Instead, regions such as China, Eastern Africa, Middle East, Northern Africa, South America and Turkey show discrepancies between WHO and TM5-FASST data. In China, for instance, the TM5-FASST model overestimates the Total $PM_{2.5}$ concentration compared to the WHO data (Figure 11a).

Population-weighted average and area average of Total $PM_{2.5}$ has been calculated for WHO and TM5-FASST data. Averages have been carried out using the population and the surface are of the cities reported in the WHO database. Averages have been calculated by region (Figure 11b and Figure 11c) and by country (Figure A1a, and Figure A1b in the Appendix). We found several discrepancies between the WHO and TM5-FASST modelled datasets. Globally, population-weighted $PM_{2.5}$ concentration from TM5-FASST is about 17% lower than $PM_{2.5}$ concentrations from WHO data (Table T1, Appendix) .Whereas, area-weighted $PM_{2.5}$ concentration from TM5-FASST is about 24% lower than WHO $PM_{2.5}$ concentrations from WHO data (Table T1, Appendix).

The major issue arising comparing the coarse output of the TM5-FASST model and point measurement data, is the representativeness of the point measurements for wider regions in-between the point measurements.
As shown in Figure 11a, 11b and 11c, population and area weighed $PM_{2.5}$ averages calculated with city and surface area data resulted in several discrepancies when comparing WHO and TM5-FASST data. Only WHO data from Western Europe and USA, with large datasets, showed to be more representative of coarse surface areas when compared with TM5-FASS data. On the other hand, fore region with smaller datasets such a China and India, point measurements cannot be representative of a larger observational dataset and therefore, they cannot be compared with the output of a coarse resolution model such as the TM5-FASST even with a sub-grid parametrization.

The discrepancy between the TM5-FASST model and the WHO database might be partially explained by the fact that in the TM5-FASST only biogenic SOA has been modelled and not the anthropogenic SOA. However, the modelling of SOA is quite uncertain due to the complex and unknown chemical reactions happening in the atmosphere. SOA formation is highly variable depending on seasons, location and meteorological conditions. Usually, overall SOA concentrations might be reach up to few $\mu g/m^3$.

Apart from the emission input uncertainties, the TM5 model uncertainties and the linearization by the reduced form emission-concentration function in FASST, there may be issues with the suitability of measured data.
At this purpose, we have to make some considerations for the experimental estimation of measured Total $PM_{2.5}$ because of several sources of uncertainties that might affect the correct estimation of measured total $PM_{2.5}$:

(a) Experimental errors (field and laboratory activity)
(b) Sampling method (usual uncertainty for sampling $PM_{2.5}$ is about 2.0 $\mu g/m^3$)
(c) Temporal coverage of the measurements (season) and yearly emission data used in the TM5-FASST model
(d) Quality Control/Quality Assurance
(e) Representativeness of measurement location for larger area.

On the other hand, there are several uncertainties related to the TM5-FASST model for the estimation of the $PM_{2.5}$ concentrations. These uncertainties can be summarized as follow:

a) Emission inventories from (HTAP) that can have incomplete coverage in some regions
b) Anthropogenic SOAs are not modelled in the TM5-FASST model but it is part of the measured $PM_{2.5}$ concentrations
c) Meteorological data in the TM5-FASST model are referred to 2001 while HTAP modelled emissions for the base year 2010
d) Linearization of the Source Receptor Model instead of using the full TM5 model) resulted in the underestimation of the $PM_{2.5}$ concentrations.
   Uncertainties in the parent models' parametrization: emissions, transport, chemistry and removal.

5.2. Comparison/Validation with Source Apportionment $PM_{2.5}$ data

In the past decade, the number of Source Apportionment works and peer reviewed publications in this field exponentially increased and tools have been developed for improving capabilities of source resolution and source contribution quantification [33]. "Pollution sources" identified and quantified by receptor models can be somehow compared to "pollution sectors" defined in the emission inventories.

Recent work[34]performed a thorough review of published Source Apportionment (SA) studies resulting in a harmonized and quality assured dataset for Europe. Additional work has been performed for other worldwide regions. About 400 hundred peer-reviewed publications have been used to screen and classify worldwide data on PM_{10}, $PM_{2.5,}$ and, PM_1 up to year 2014. These studies are based on urban, sub-urban, remote, rural, and industrial site locations

In this work, SA data for $PM_{2.5}$ mass concentrations are compared with TM5-FASST $PM_{2.5}$ modeled concentrations.

The most common pollution sources estimated in the reviewed SA case studies are:

1) Sea Salt (including road salting)
2) Secondary Inorganic Aerosols (SO_4, NO_3, NH_4)
3) Crustal/Re-Suspended Dust
4) Traffic (only ground transportation including road dust)
5) Industry-Oil Combustion (include point sources and power plants for energy production)
6) Biomass Combustion (small-scale biomass and biomass waste burning such as, wood and wood waste burning)
7) Residential and small-scale non-industrial combustion
8) Secondary Organic Aerosol (usually estimated from PM_1)
9) Unexplained/other sources

The whole set of data gathered from the analysis of SA does not always give estimates of the above sources. In addition, for a correct attribution of the pollution sources, all chemical elements of $PM_{2.5}$ should be analyzed and compared to reference sources.

Only sources from SA studies such as Traffic, Residential Heating, Biomass Burning and, partially Industry-Oil Combustion can be compared with the emission sectors (used in TM5-FAST) of Transport, Residential, Biomass Burning, Industry and Energy, respectively.

For this comparison, we have considered SA case studies including:

- Total $PM_{2.5}$ mass closure of about 100% (about 100 case studies)
- Urban site locations (including residential area)

From the above SA case studies, we have selected those which could be associated to the same "pollution sector" as defined for the TM5-FASST according to emission inventory. :

- 213 case studies for the Traffic and Re-Suspended Dust pollution factor/source
- 150 case studies for the Industry pollution factor/source

These studies provide data both on Total $PM_{2.5}$, as well as the source attribution. We will compare both metrics with our TM5-FASST model results for year 2010.

5.2.1. Total $PM_{2.5}$: TM5-FASST *vs* SA case studies

220 case studies have been selected to compare Total $PM_{2.5}$ concentration from SA and TM5-FASST model estimations. Figure 12a shows a breakdown of the statistical analysis by region. Most of the SA studies have been performed Europe (83) and in the USA (40) and Europe. However, a non-negligible number of SA studies have been also performed in the China (23) and India (13) region, South Eastern Asia (11), Brazil and Oceania (8), and rest of South America (5).

As we can see, TM5-FASST data values fall in the same range of SA data for India, USA, Western Europe, Canada and Oceania whose median values are also below 25 $\mu g/m^3$, that is the EU limit value for Total $PM_{2.5}$. As for the WHO data, population-weighted average and area average of Total $PM_{2.5}$ has been calculated for SA and TM5-FASST data. Averages have been carried out using the population and the surface are of the cities used in the SA database. Regional averages are shown in Figure 12b and Figure 12c. Country averages are shown in Figure A2a and Figure A2b of the Appendix. Several discrepancies have been observed between the SA and TM5-FASST modelled datasets. Globally, population-weighted Total $PM_{2.5}$ concentration from TM5-FASST is about 30% lower than $PM_{2.5}$ concentrations from SA data (Table T1, appendix). Whereas, area-weighted Total $PM_{2.5}$ concentration from TM5-FASST is about 33% lower than SA $PM_{2.5}$ measured concentrations (Table T2, Appendix).

As said above, the major issue arising comparing the coarse output of the TM5-FASST model and point measurement data from Source Apportionment studies, is the representativeness of the point measurements for wider regions in-between the point measurements. In addition, the limited number of Source Apportionment studies analyzed for this work cannot necessarily need to be considered a representative subset of a larger observational dataset.

5.2.2. $PM_{2.5}$ from Transport: TM5-FASST vs SA case studies

212 case studies have been selected to compare Transport $PM_{2.5}$ concentration from SA and TM5-FASST model estimations. Figure 13a shows a statistical analysis for SA and TM5-FASST Transport $PM_{2.5}$ concentrations. Population-weighted average and area average of Transport $PM_{2.5}$ has been calculated for SA and TM5-FASST data. Regional averages are shown in Figure 12b and Figure 12c. Country averages are shown in Figure A2a and Figure A2b of the Appendix. Very large discrepancies have been observed between the SA and TM5-FASST modelled datasets: globally, population-weighted Transport $PM_{2.5}$ concentration from TM5-FASST was about 70% lower than $PM_{2.5}$ concentrations from SA data (Table T3, Appendix) Whereas, area-weighted weighted Transport $PM_{2.5}$ concentration from TM5-FASST was about 77% lower than SA $PM_{2.5}$ measured concentrations (Table T3, Appendix)

Large discrepancies between SA and TM5-FASST Transport PM2.5 concentrations might be tentatively attribute to:

- Low representativeness of the SA dataset as subset of a bigger dataset for large regions
- Difficulty to compare point measurement concentrations from SA studies with the output of a coarse model such as the TM5-FASST which produces regional $PM_{2.5}$ concentrations
- Anthropogenic SOAs and therefore SOA form Transport emission are not modelled in the TM5-FASST model.

In several SA case studies, the source of re-suspended Dust is often mixed with the Transport source. This leads to an over-estimation of the transport $PM_{2.5}$ concentration, in particular in those area where dust storms are recurrent such as in parts of China and India.[35–37] In TM5-FASST only dust from large desert regions are included, not a potential re-suspension or road-dust.

Recent SA works carried out in six major cities in India, estimate that 40-60% of PM_{10} is attributed to re-suspended Dust. Similar situation is observed for $PM_{2.5}$. [38] However, these estimates represent hot spots concentrations in the Indian region.
On the other end, the prediction of Dust by atmospheric chemistry transport models is very uncertain, due to complexities in describing source regions and source functions.
Re-Suspended Dust emissions are strongly related to meteorological conditions and episodic phenomena and are anyhow (in our study) only referring to long-range desert dust transport.

5.2.3. $PM_{2.5}$ from Industry: TM5-FASST vs SA case studies

150 case studies have been selected to compare Industry $PM_{2.5}$ concentration from SA and TM5-FASST model estimations.
As done for Transport $PM_{2.5}$, population-weighted average and area average of Industry $PM_{2.5}$ has been calculated for SA and TM5-FASST data. Regional averages are shown in Figure 13b and Figure 13c. Country averages are shown in Figure A3a and Figure A3b of the Appendix. Large discrepancies have been observed between the SA and TM5-FASST modelled datasets: globally, population-weighted Transport $PM_{2.5}$ concentration from TM5-FASST was about 13% higher than $PM_{2.5}$ concentrations from SA data (Table T4, Appendix) Whereas, area-weighted weighted Transport $PM_{2.5}$ concentration from TM5-FASST was about 17% higher than SA $PM_{2.5}$ measured concentrations (Table T4, Appendix)

Figure 17 shows the comparison between Industry $PM_{2.5}$ from TM5-FASST model and SA studies. Low $PM_{2.5}$ concentrations are observed in Western Europe and USA. Instead, higher concentrations are found in the India and China.

Figure 18 shows that agreement between model and measurements is in general better than for Transport. Largest discrepancy is found for a single location in Argentina and more systematically in the Eastern Mediterranean.
Emissions from industries represent point sources that are confined in local areas. Instead, outputs from the TM5-FASST model are representative for country and regions rather than local areas.. In addition, several SA studies have also the tendency to include the Energy sector in the Industry sector, with the consequence of erroneous estimation of Industry $PM_{2.5}$ concentration.

Brauer er al. work comparison

TM5-FASST results from the present study have been compared with results from similar study carried out by Brauer et al. (2012). In that work, global TM5 modelled $PM_{2.5}$ concentrations and satellite $PM_{2.5}$ estimation were combined with satellite-based $PM_{2.5}$ estimates. TM5 runs were performed with emissions for 2005, while using meteorology for 2001 as a representative period. $PM_{2.5}$ concentration estimations were derived from satellite observations of Aerosol Optical Depth (AOD), a measure.
We made a comparison between the TM5-satellite combined $PM_{2.5}$ concentration estimation and the TM5-FASST output from this work. As shown in Figure 14, TM5-FASST results were grouped according to the region grouping used in the Brauer et al. work (this is the region grouping following the WHO scheme).
Globally, satellite-based $PM_{2.5}$ estimates from Brauer et al. were 40% lower than the TM5-FASST modelled $PM_{2.5}$ concentrations estimated in this work (Figure 14, Table T4, Appendix).
On a first attempt, we might attribute discrepancies might be attributed to different emission inputs. TM5 model in Brauer et al. used emission from base year 2005 whereas, TM5-FASST model in this work used emission from base year 2010.

6. Conclusions and outlook

At the date, $PM_{2.5}$ is considered the most harmful pollutant having negative effects on human health. Therefore, there is the necessity to develop methodologies aiming to estimate the $PM_{2.5}$ concentrations attributed to different economic sectors.
The present study estimates the relative emission sector contributions to Total $PM_{2.5}$ concentrations using global emission inventory data as input of the TM5-FASST model. This model estimates high-resolution population-weighted $PM_{2.5}$ averages for each emitting sector, becoming the base for health impact evaluation.

This work highlighted the importance of estimating $PM_{2.5}$ mass concentration from economic sectors such as Traffic, Energy, Industry, Agriculture, Residential activities and Biomass Burning. The presented results are derived from global emission inventories for the year 2010 (HTAP-v2) and their processing with a global reduced-form chemical transport model TM5-FASST. Output $PM_{2.5}$ concentrations have been distributed according population density for the year 2010 and, population-weighted averages have been calculated. Results

showed that emerging economies such as the China and India region, having the highest concentration of $PM_{2.5}$ from the Industry and Residential sector. China and India are the countries with the highest $PM_{2.5}$ concentration with 55 $\mu g/m^3$ and 55 $\mu g/m^3$, respectively. Industry is the largest economic sector in China contribution to 38% China of total $PM_{2.5}$ emissions. Whereas India, showed the larger contribution of Residential emission (42%) followed by Western Africa (40%). Transport sector showed highest contribution to $PM_{2.5}$.

The Transport sector showed largest absolute values in the India, China and Korea region. However, the biggest contribution of Transport to $PM_{2.5}$ has been observed in Western Europe (18%) and Japan (17%). The Energy sector showed highest values over India and China but has its largest contribution (38%) to $PM_{2.5}$ in Middle East.
Agriculture showed important contribution to $PM_{2.5}$ in the Kazakhstan region (36%) as well as in Northern Africa and Western Europe (32%). Residential activities highly contributes to $PM_{2.5}$ formation in India (42%), Western Africa (40%) and Central America (38%). Finally, Biomass Burning is the major highest contributor for $PM_{2.5}$ formation in Brazil (51%), Southern Africa (29%) and Rest of South America (28%) and Canada (27%).

The above findings have been illustrated with absolute and relative population-weighted average $PM_{2.5}$ concentration map for each economic sector contributing to global pollution.

This is the first work comparing sector attributed modelled $PM_{2.5}$ concentrations with $PM_{2.5}$ point measurements form the WHO database (2014) and Source Apportionment case studies. Several discrepancies were found when comparing modelled results with population- and area-weighted point measurement of $PM_{2.5}$ concentrations averages. The biggest issue might be identified with the lack of representativeness of the point measurement dataset when compared with the coarse output of a chemical transport model such as the TM5-FASST. This highlighting the importance to have a more comprehensive up-to-date dataset for source-attributed $PM_{2.5}$. More data should be provided and analyzed from emerging countries where most of the highest pollution level are recorders and where there is high population density. In addition, there is the urgent need to harmonize the pollutant source-attribution (referred to economic sectors) to $PM_{2.5}$ formation.

The present work outlined the necessity to take actions for:

1) Improving the spatial resolution of the TM5-FASST model to better target the emissions at city scales.
2) Focusing on case studies dealing with megacities where there is a more accurate knowledge from local emissions and measurements.
3) Using Source Apportionment (SA) data to improve the performance and output of the TM5-FASST model.
4) Making efforts to understand the causes of the underestimation of Transport concentration from the TM5-FASST model compared to SA case studies.
5) Improving the source separation and identification between model and measurements. For instance, biomass burning is generally assigned to small domestic fires and residential heating in most of source

apportionment studies. Whereas, modelled data from emission inventories use large scales fires and residential emissions as separate inputs.

6) Calling for a better cooperation between experimental field research and modelers to fill the gap between SA data and modeled data.

I think the conclusions should more targeted to the scope of the study: to provide a first global analysis of source contributions to PM levels. In the end you need to convince readers that there is useful information from this study, not only what is not possible. Where are findings consistent, where can't we say much (because of large uncertainties), where are results definitely inconsistent, and if so do we understand that from source uncertainties.

The WHO dataset is entirely based on ground measurements that, when performed in locations representative for human exposure such as in residential or commercial areas, are best representing exposure in those specific locations. However, they are only available in a limited number of locations and cities worldwide. Furthermore, in many developing countries, measurements are often limited to PM10, and a conversion factor needed to be applied to estimate $PM_{2.5}$ levels.

The SA dataset has been build gathering all available case studies published until August 2014. About 70% of these studies have been performed in urban and residential areas. In addition, in bigger cities it was possible to gather several case studies performed in different locations of the city. Therefore, most of the SA $PM_{2.5}$ measurements are representative of urban agglomerates.

The TM5-FASST model gives estimates of $PM_{2.5}$ at global level with urban increment. The coarse output of the model has been subsequently sub-gridded in smaller cell. However, the original information always comes from the coarse output of the model. The TM5-FASST model output can be therefore considers as regional, being able to give a coarse estimate of the $PM_{2.5}$ concentration over entire regions.

The TM5-FASST model do not parametrize re-suspension of road dust that, based on recent studies, may represent up tp 40%-60% of $PM_{2.5}$ in developing countries and presumably in all worldwide city with Traffic from vehicles. Re-suspended dust is not modelled in the TM5-FASST and this is an obvious source of uncertainty that affect the correct $PM_{2.5}$ concentration estimation in agglomerated urban areas with high Traffic vehicles.

At the date, there are not studies about the factorization of road dust from re-suspended dust. Several SA studies were able to estimate the fraction of road-dust from re-suspended dust, based on its chemical composition. From the SA gathered for this work it was possible to determine an empirical relationship between Re-Suspended Dust and Transport $PM_{2.5}$ concentrations (Figure A15, Appendix). In that analysis, high concentration values come from cities in India and China where the Dust contribution is not negligible. This parametrization is an attempt to estimate the fraction of re-suspended dust (road dust) attributed to Transport. However, this parameterization is strongly dependent from local Traffic conditions and cannot be applied at a

global level. Therefore, more work should be done in order to understand how Re-Suspended Dust could be included in the Transport/Traffic sector.

Comparison at regional level of Total $PM_{2.5}$ concentrations with the $PM_{2.5}$ dataset from WHO showed several discrepancies. These are mainly attribute to the impossibility of the TM5-FASST model to target point sources of $PM_{2.5}$. In addition, the WHO dataset cannot be representative for all the regions where the TM5-FASST model generate its $PM_{2.5}$ estimates.

The same issue has been observed even with bigger discrepancies when the TM5-FASST outputs, for specific economic sectors, have been compared Source Apportionment results from worldwide case studies.

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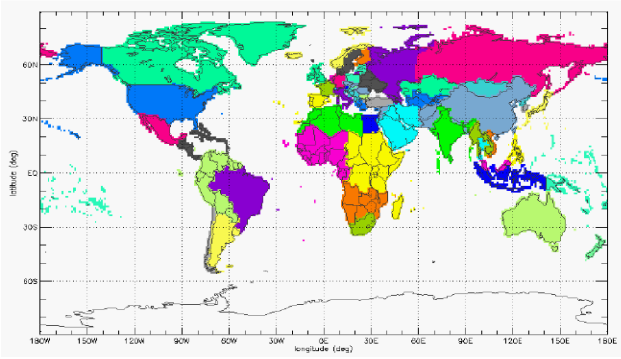

Figure 1. Definition of the 56 source regions within TM5-FASST

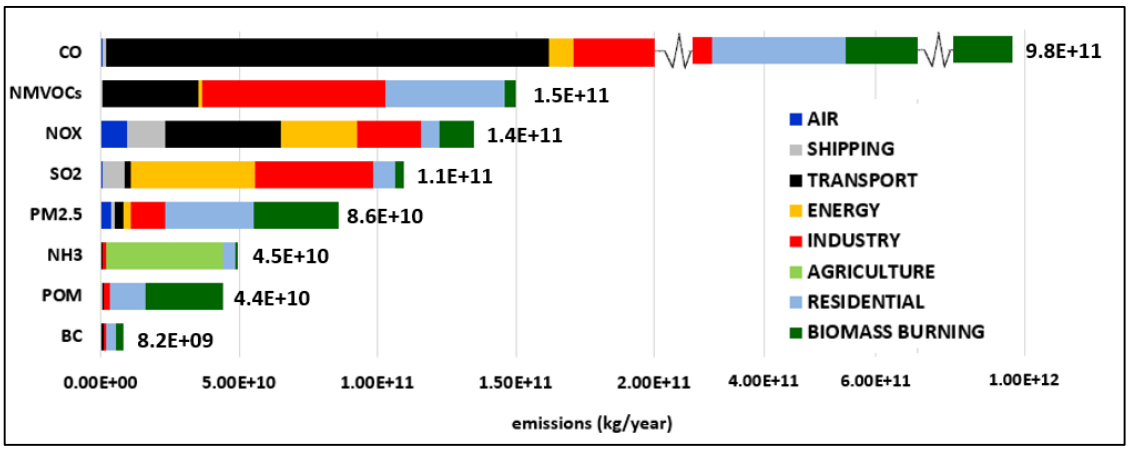

Figure 2. Total emissions for pollution sectors (kg/Year) calculated from country totals emissions (year 2010). C

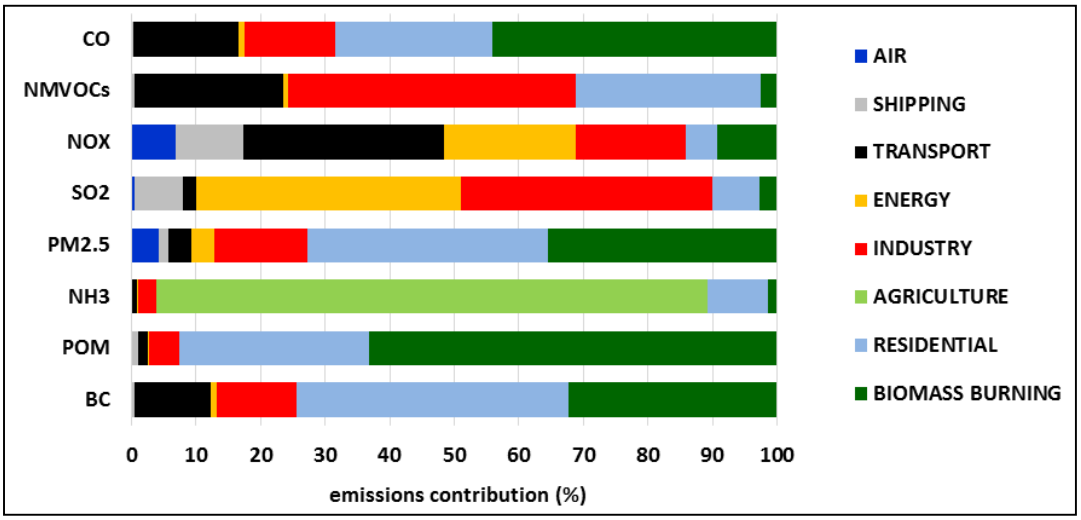

Figure 2a. Contribution [%] of chemical precursors to total emissions calculated from country totals emissions (year 2010).

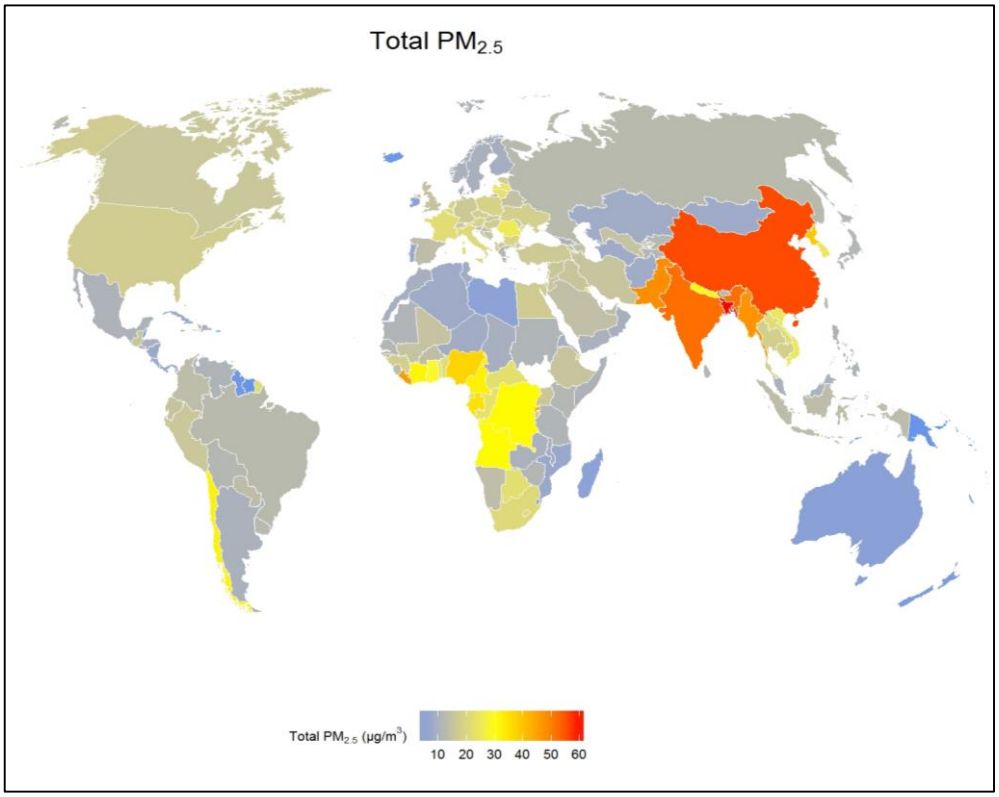

Figure 3. Population-weighted annual $PM_{2.5}$ concentration by country (2010). Color scheme is somewhat difficult. Choose more distinct colors? Not sure why numbers are presented as aggregated by country?

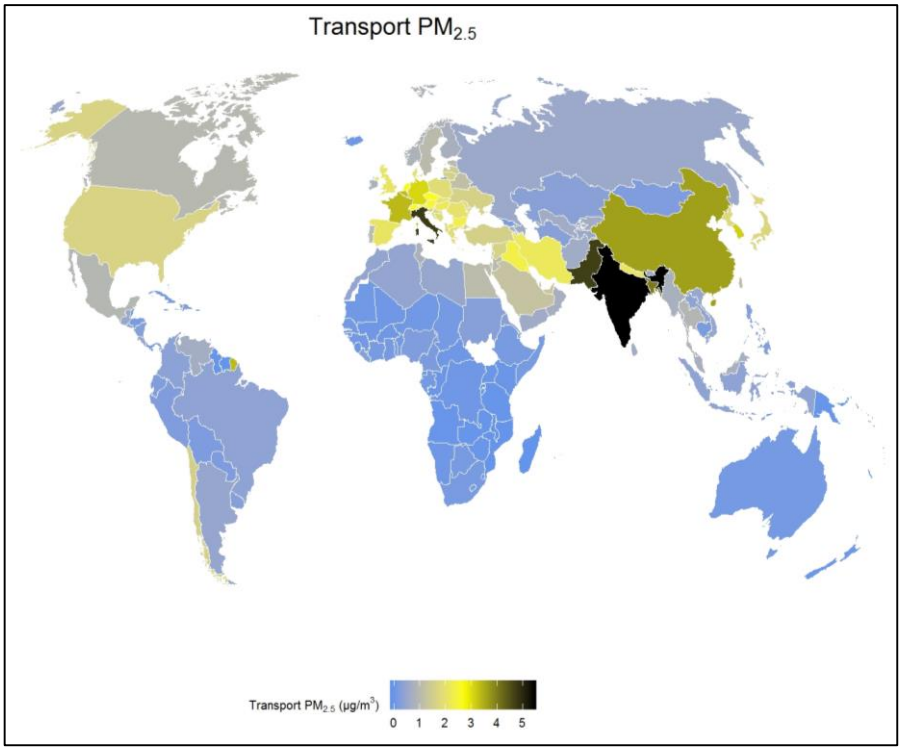

Figure 4. Population-weighted annual Transport $PM_{2.5}$ concentration by country (2010). Difficult color scheme.

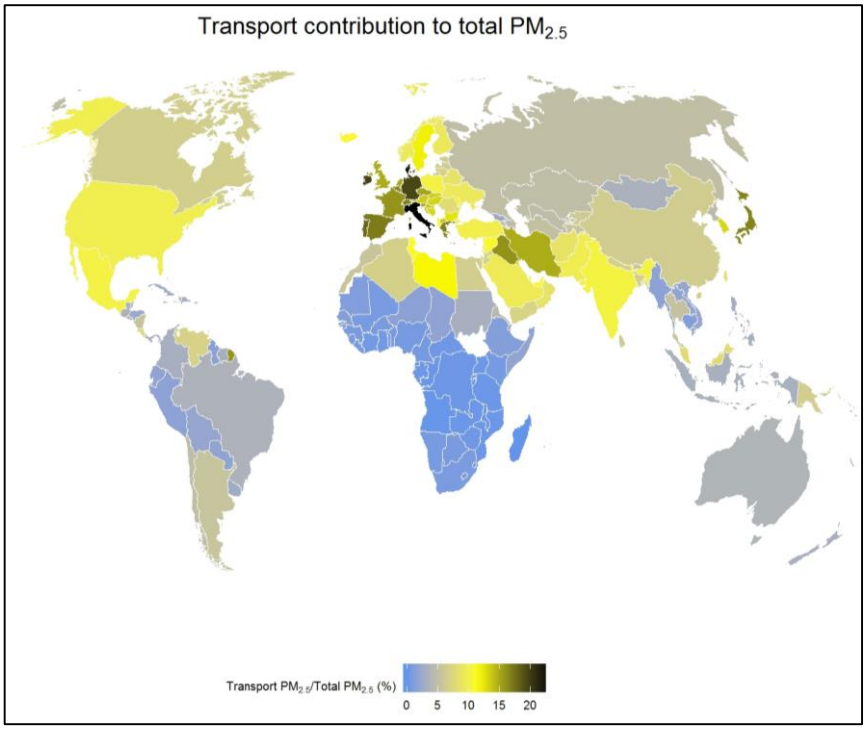

Figure 4a. Contribution of population-weighted Transport $PM_{2.5}$ to total $PM_{2.5}$ (2010).

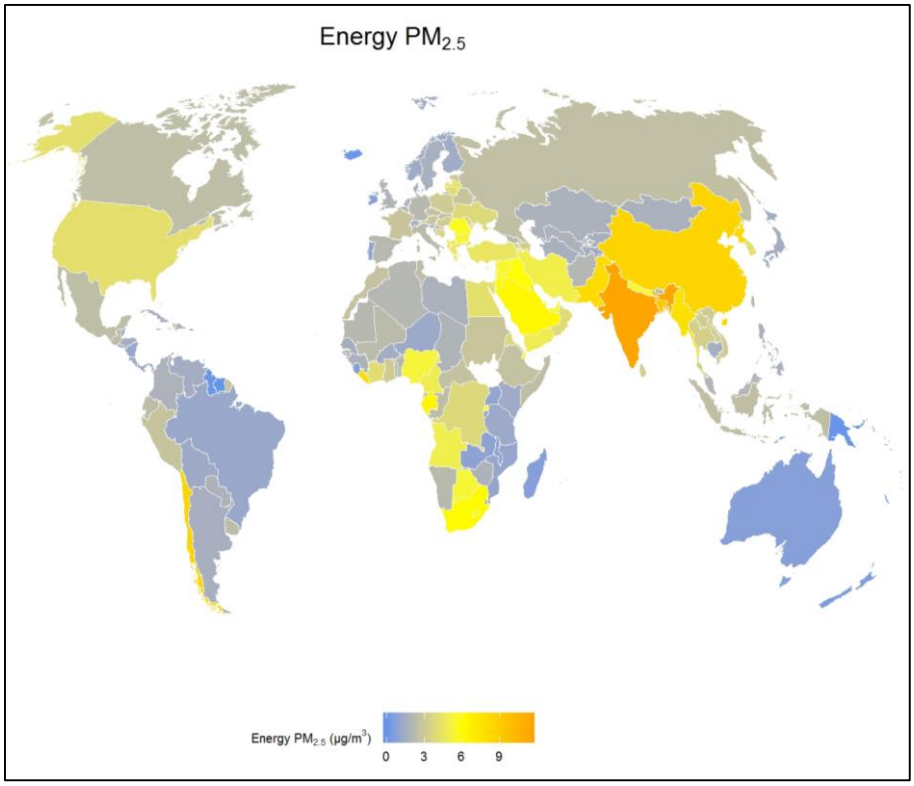

Figure 5. Population-weighted annual Energy $PM_{2.5}$ concentration by country (2010).

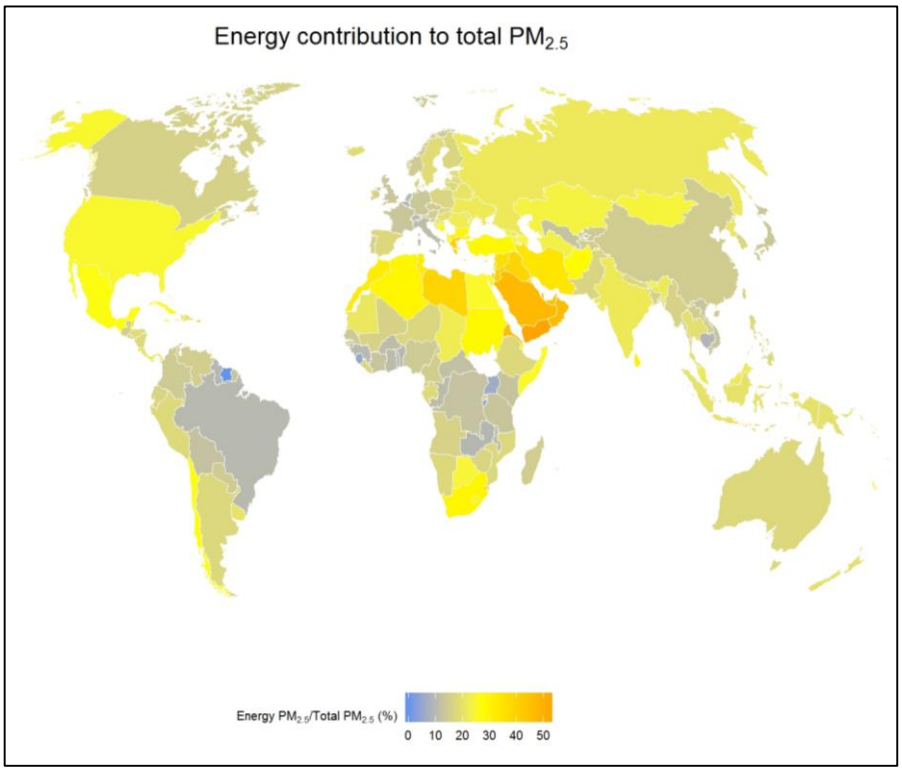

Figure 5a. Contribution of population-weighted Energy $PM_{2.5}$ to total $PM_{2.5}$ (2010).

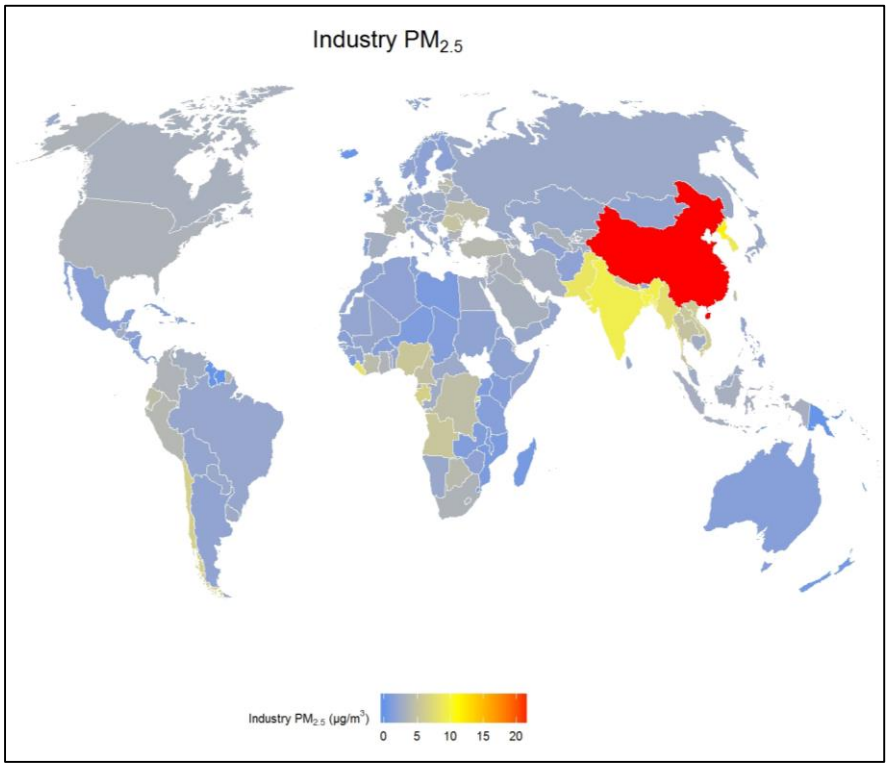

Figure 6. Population-weighted annual Industry $PM_{2.5}$ concentration by country (2010).

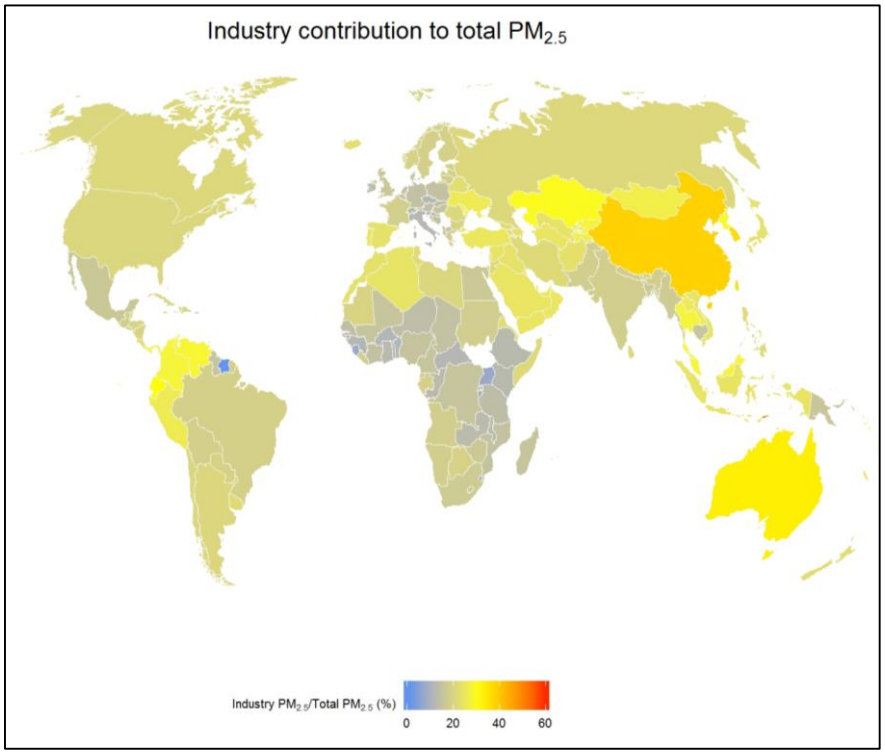

Figure 6a. Contribution of population-weighted Industry $PM_{2.5}$ to total $PM_{2.5}$ (2010).

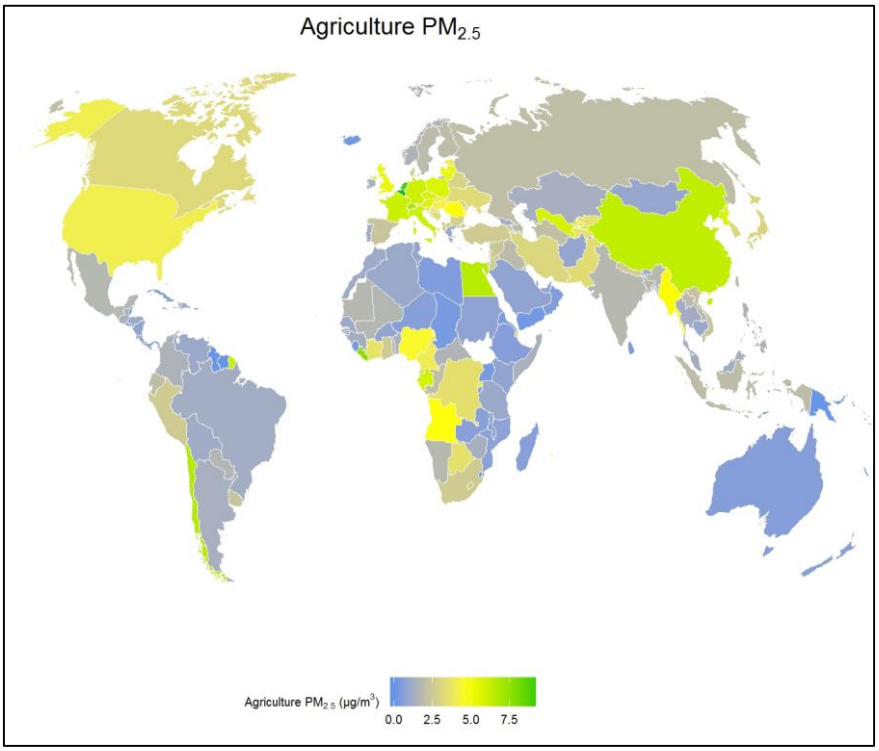

Figure 7. Population-weighted annual Agriculture $PM_{2.5}$ concentration by country (2010).

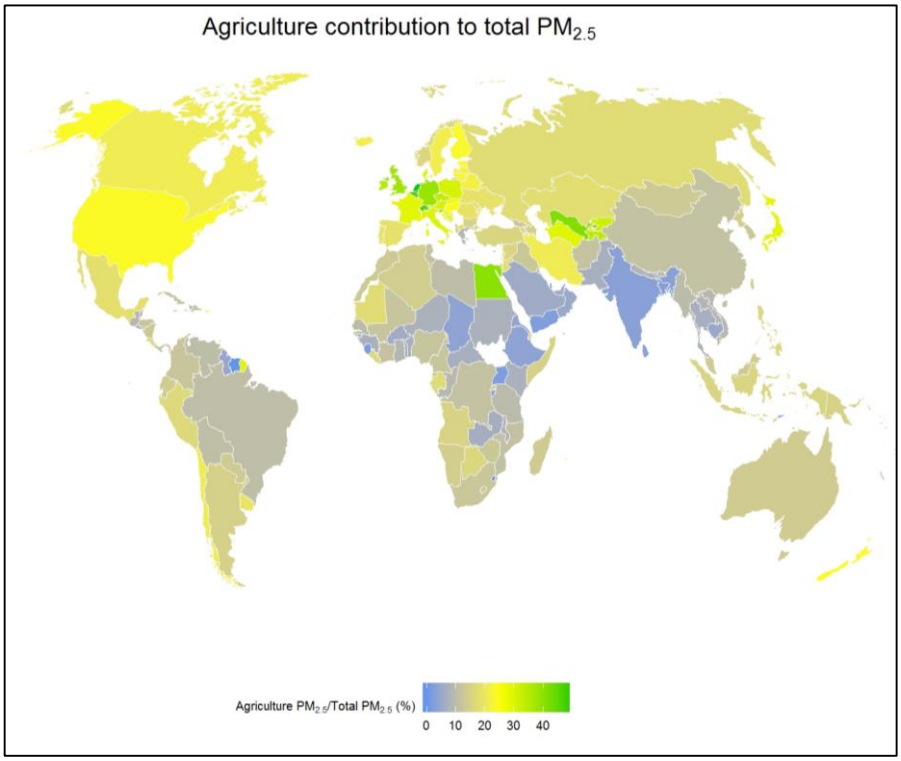

Figure 7a. Contribution of population-weighted Agriculture $PM_{2.5}$ to total $PM_{2.5}$ (2010).

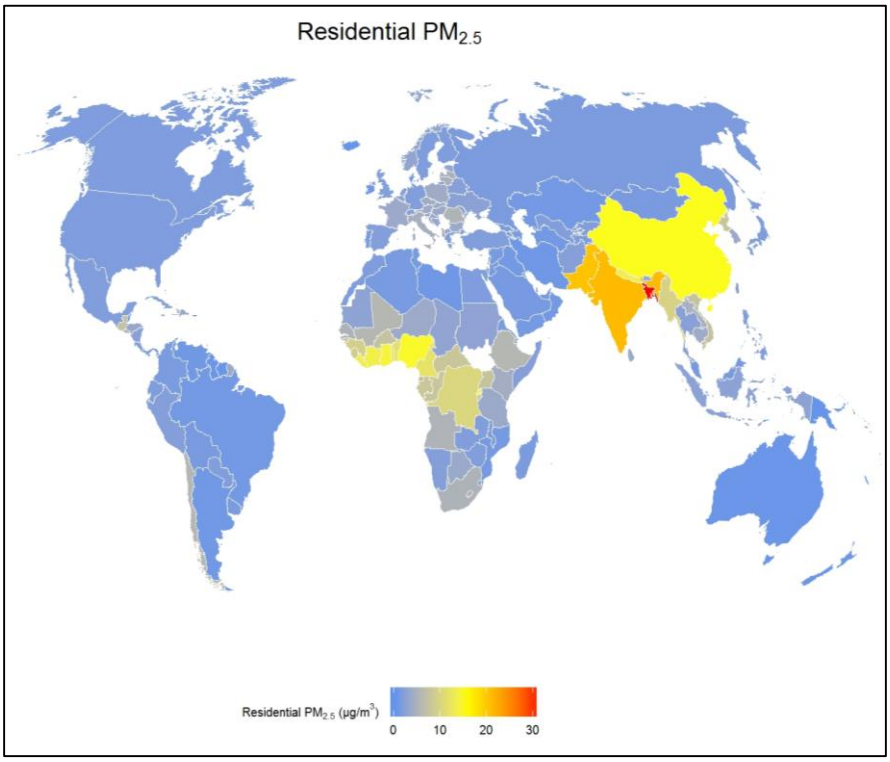

Figure 8. Population-weighted annual Residential $PM_{2.5}$ concentration by country (2010).

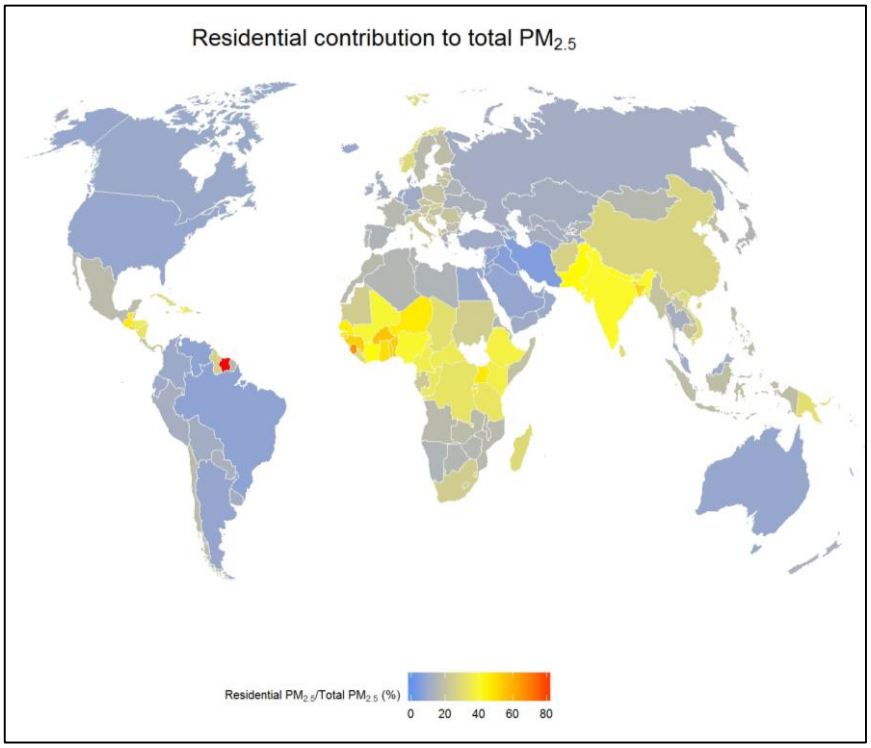

Figure 8a. Contribution of population-weighted Residential $PM_{2.5}$ to total $PM_{2.5}$ (2010).

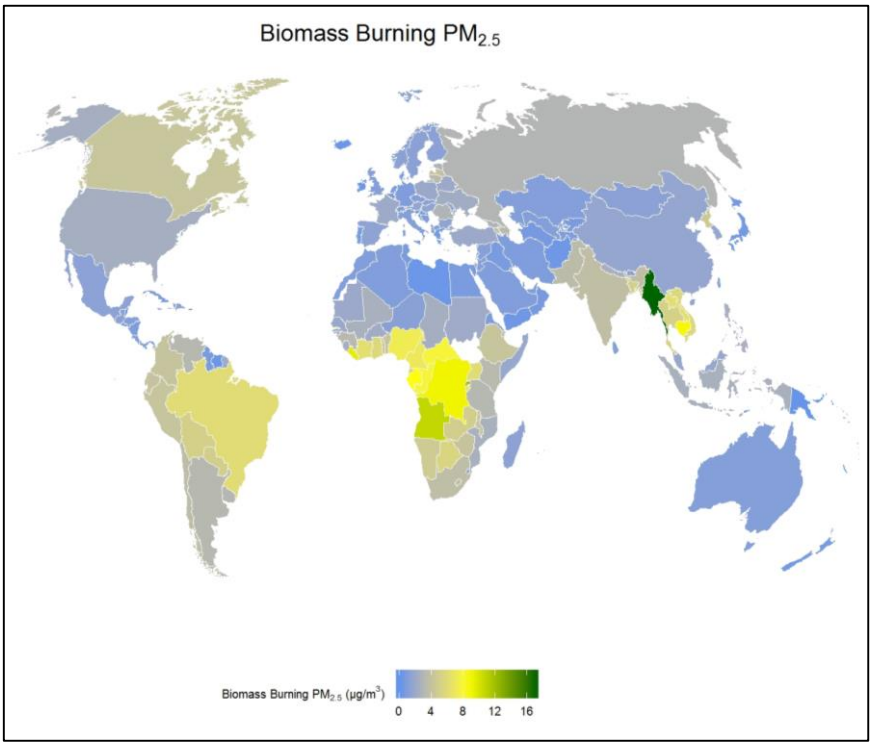

Figure 9. Population-weighted annual Biomass Burning $PM_{2.5}$ concentration by country (2010).

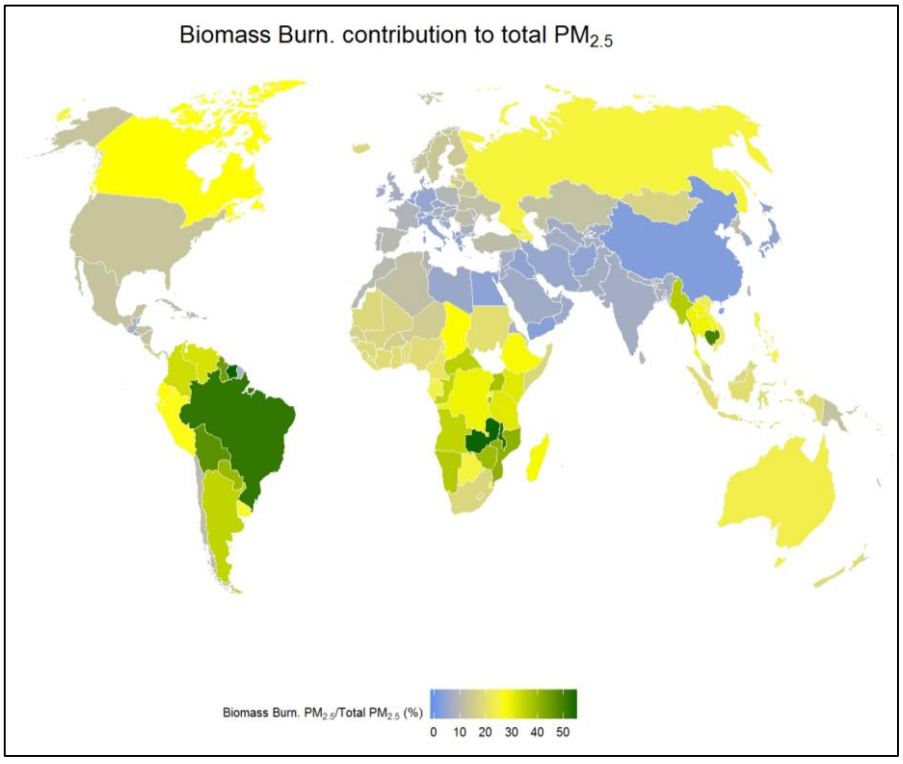

Figure 9a. Contribution of population-weighted Biomass Burning $PM_{2.5}$ to total $PM_{2.5}$ (2010).

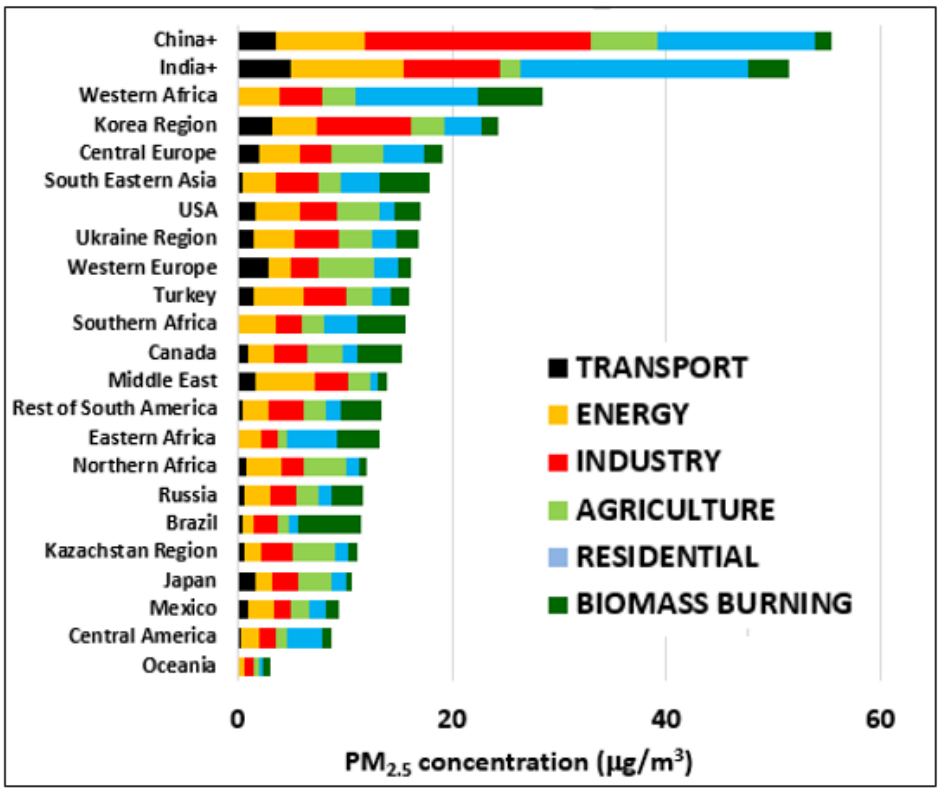

Figure 10. Population-weighted annual $PM_{2.5}$ fraction average calculated for economic sectors (2010).

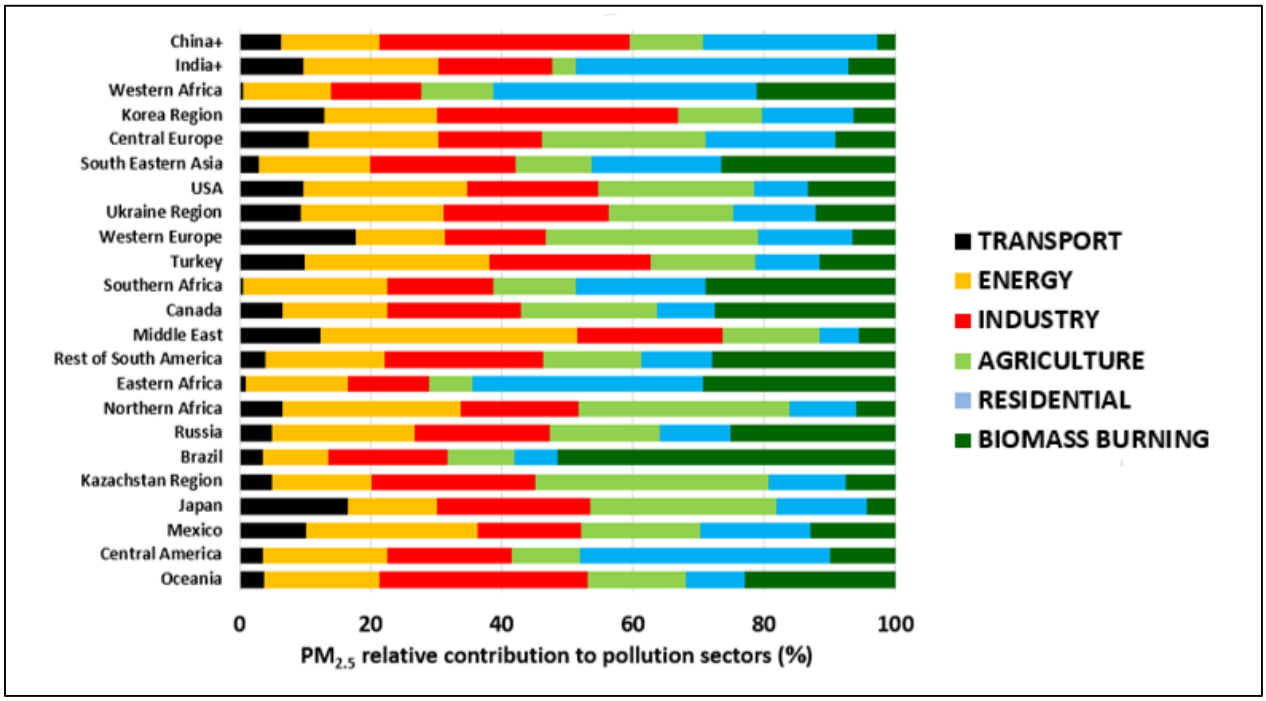

Figure 10a. Contribution of economic sectors to annual population-weighted Total $PM_{2.5}$ averages (2010).

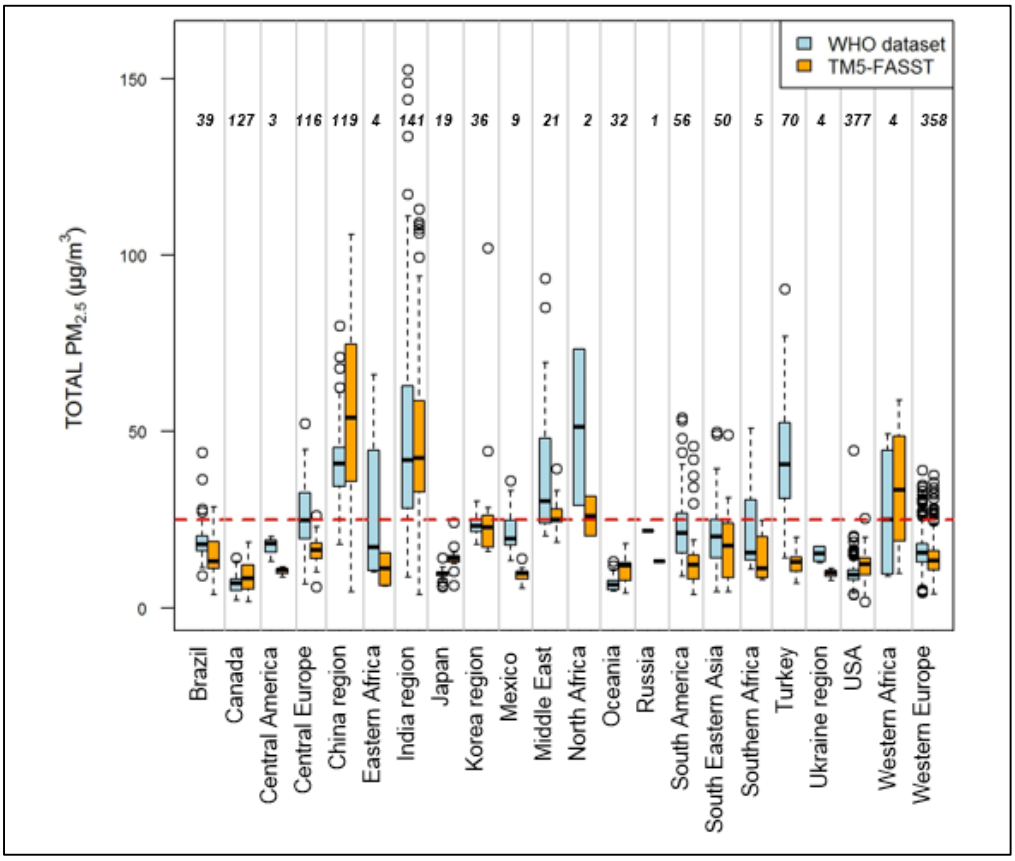

Figure 11a. Comparison between the WHO Ambient Air Pollution (AAP) dataset (2014) representative for 2008-2013 and TM5-FASST data for Total $PM_{2.5}$. Boxplots represent the $PM_{2.5}$ concentration median value (middle line), the upper and lower quartile (box), the minimum and maximum of all the data (whiskers) and the outliers (circles). Numbers indicate the amount of data available in each region. Red dashed line is the 25µg/m^3 European limit value for Total $PM_{2.5}$.

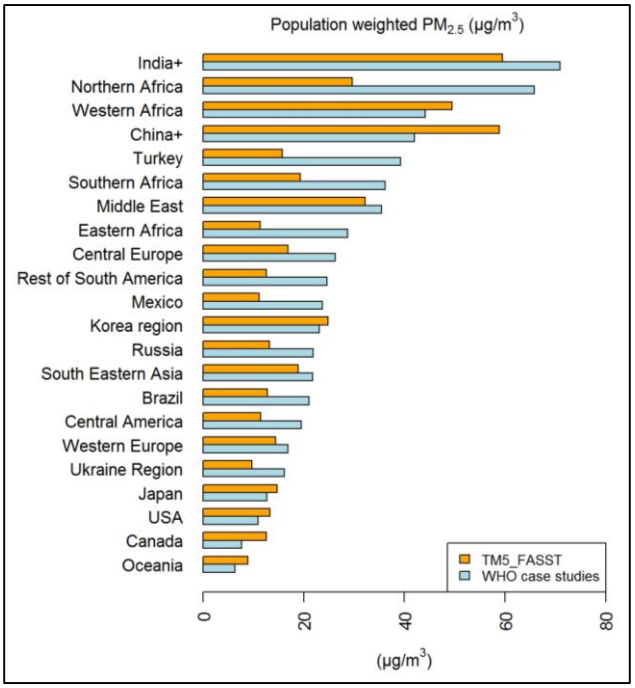

Figure 11b. Comparison between the population-weighted $PM_{2.5}$ concentration regional averages from the WHO Ambient Air Pollution (AAP) dataset (2014, representative for 2008-2013), and the TM5-FASST data for Total $PM_{2.5.}$ (year 2010).

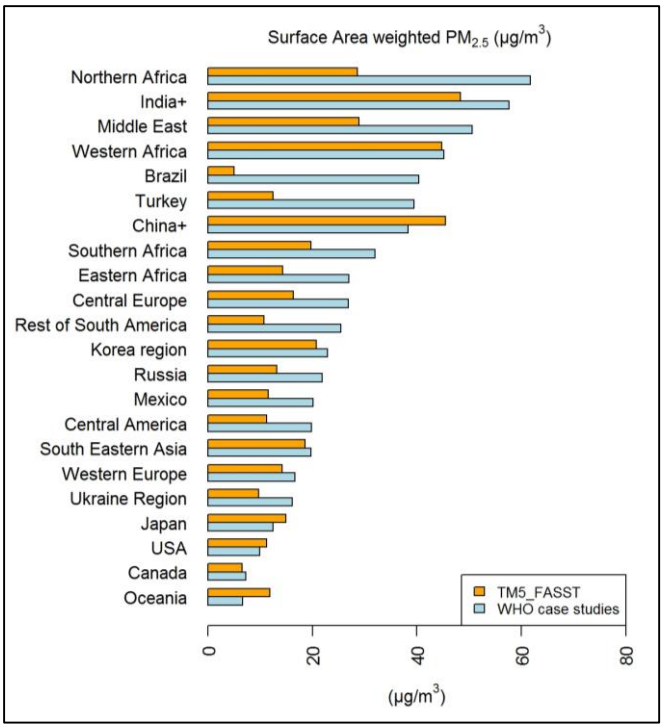

Figure 11c. Comparison between the area-weighted $PM_{2.5}$ concentration regional averages from the WHO Ambient Air Pollution (AAP) dataset (2014, representative for 2008-2013), and the TM5-FASST data for Total $PM_{2.5.}$ (year 2010).

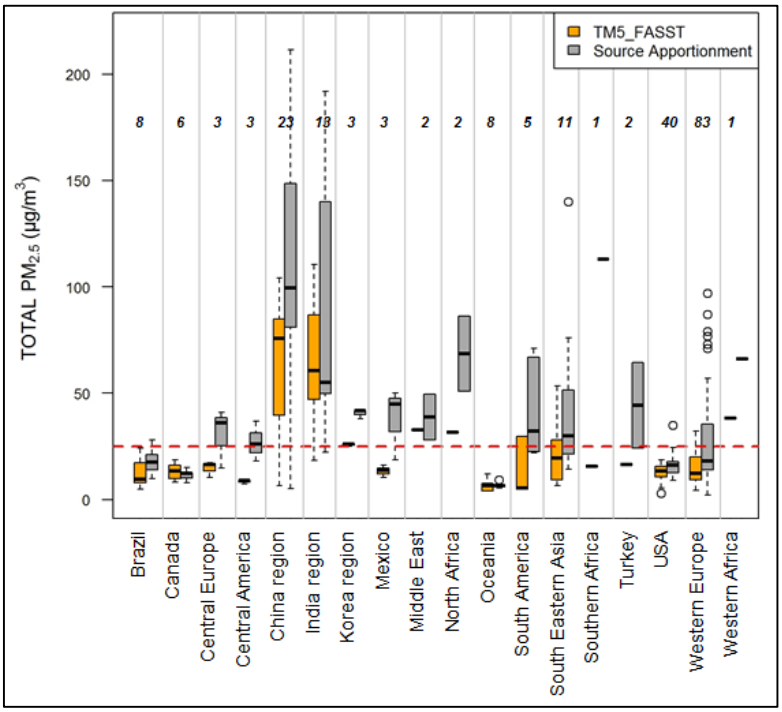

Figure 12a. Comparison between TM5-FASST and Source Apportionment data for Total $PM_{2.5}$ concentrations (up to year 2014).

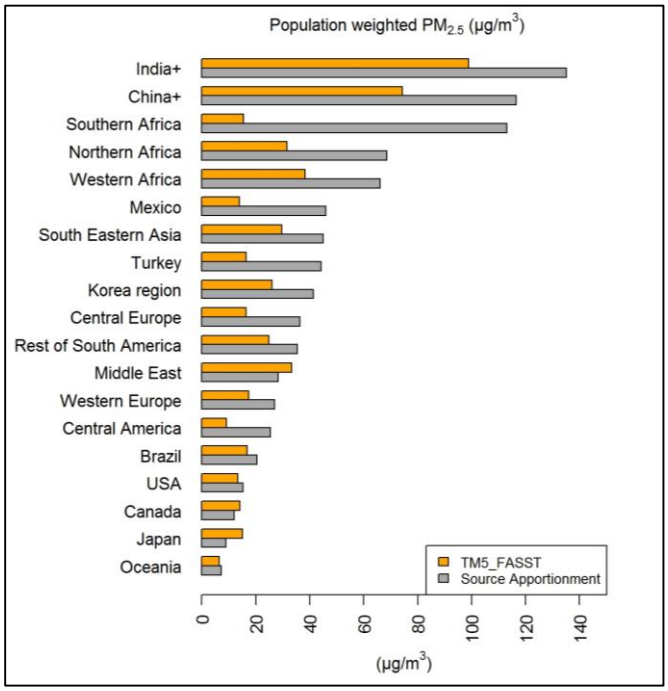

Figure 12b. Comparison between the population-weighted Total $PM_{2.5}$ concentration regional averages from Source Apportionment case studies (up to 2014) and the TM5-FASST modelled data for Total $PM_{2.5}$ (year 2010).

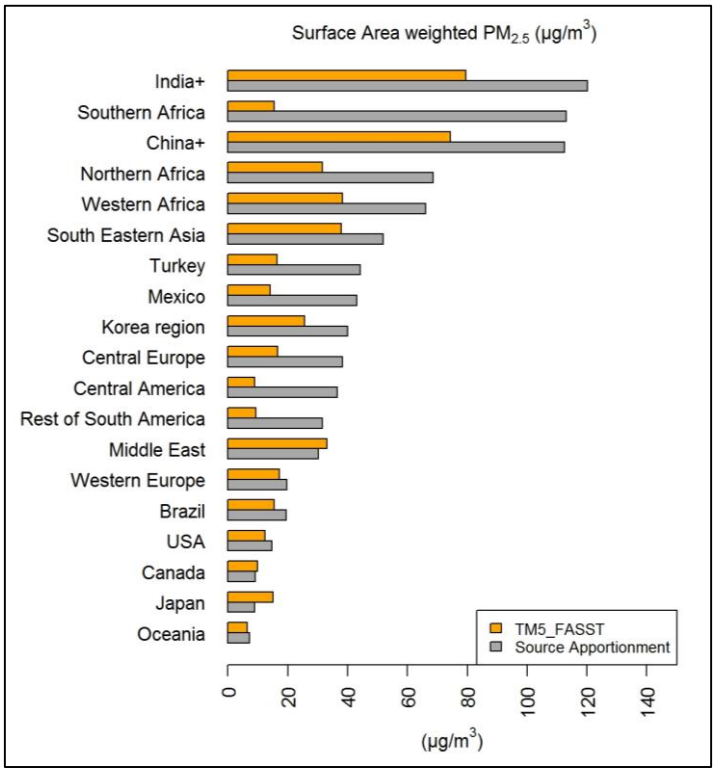

Figure 12c. Comparison between the area-weighted Total $PM_{2.5}$ concentration regional averages from Source Apportionment case studies (up to 2014) and the TM5-FASST modelled data for Total $PM_{2.5}$ (year 2010).

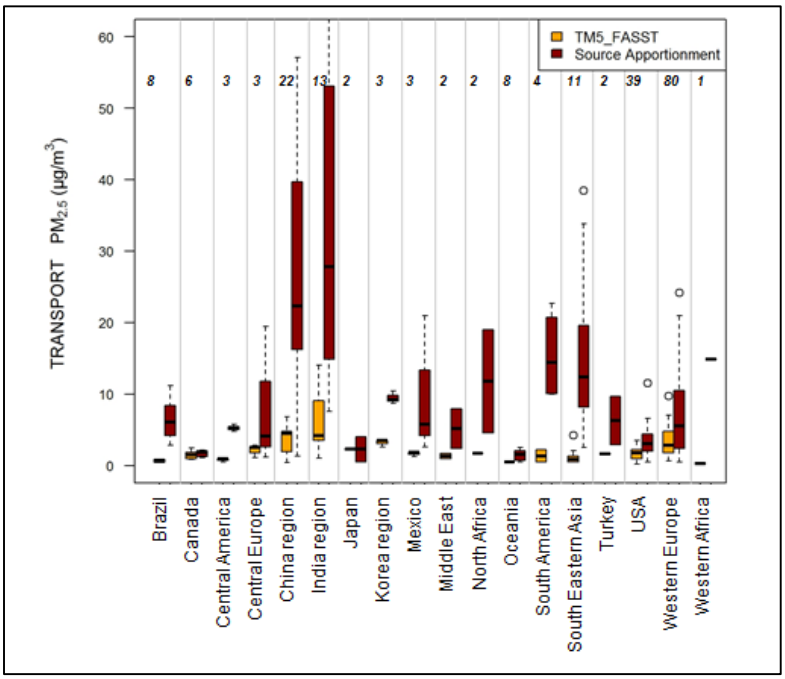

Figure 13a. Comparison between TM5-FASST and Source Apportionment data for Transport $PM_{2.5}$ concentrations.

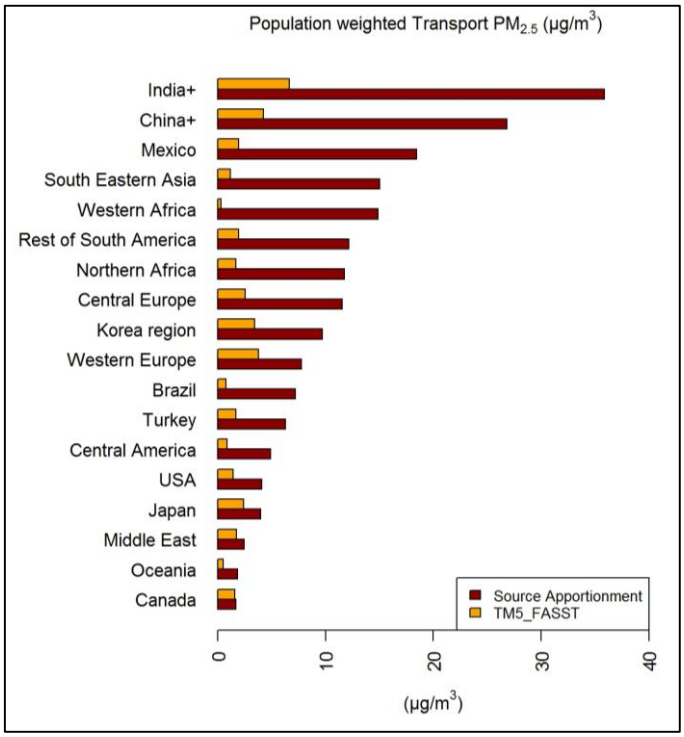

Figure 13b. Comparison between the population-weighted Transport $PM_{2.5}$ concentration regional averages from Source Apportionment case studies (up to 2014) and the TM5-FASST modelled data for Total $PM_{2.5}$ (year 2010).

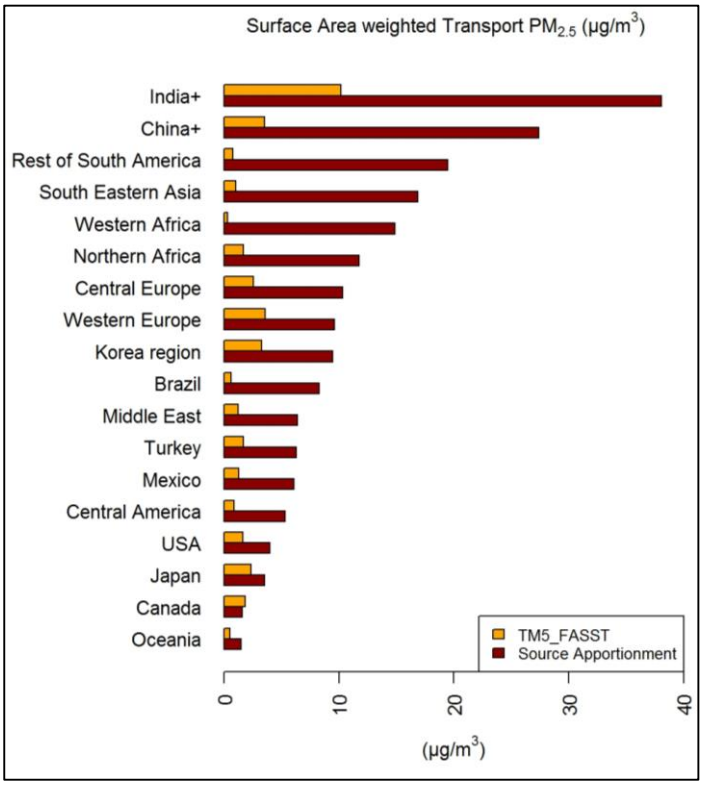

Figure 13c. Comparison between the area-weighted Transport $PM_{2.5}$ concentration regional averages from Source Apportionment case studies (up to 2014) and the TM5-FASST modelled data for Total $PM_{2.5}$ (year 2010).

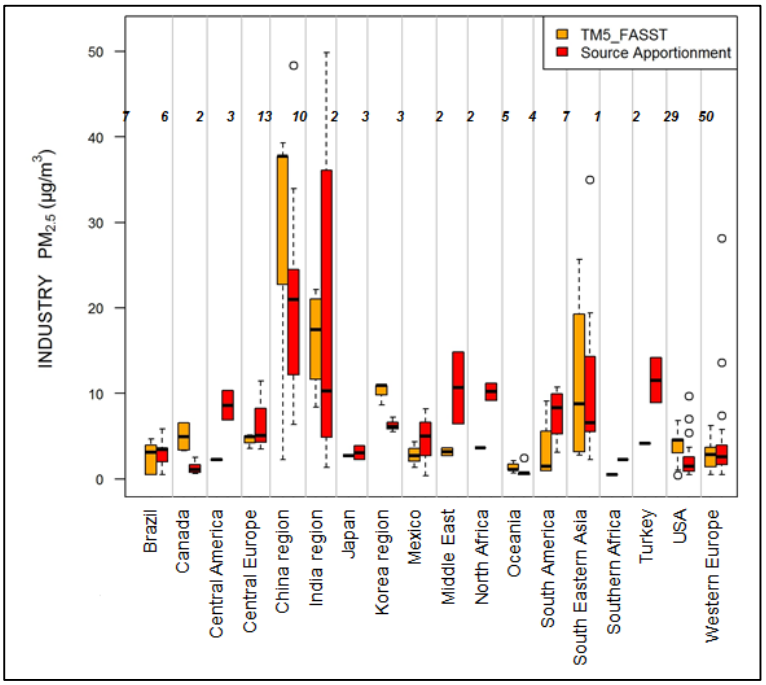

Figure 14a. Comparison between TM5-FASST and Source Apportionment data for Industry $PM_{2.5}$ concentrations. Numbers indicate the amount of data available in each region.

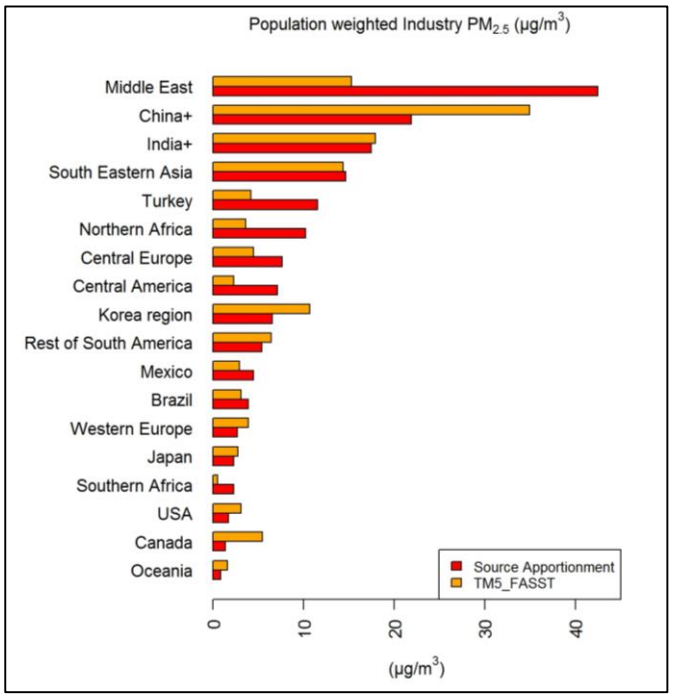

Figure 14b. Comparison between the population-weighted Industry $PM_{2.5}$ concentration regional averages from Source Apportionment case studies (up to 2014) and the TM5-FASST modelled data for Total $PM_{2.5}$ (year 2010).

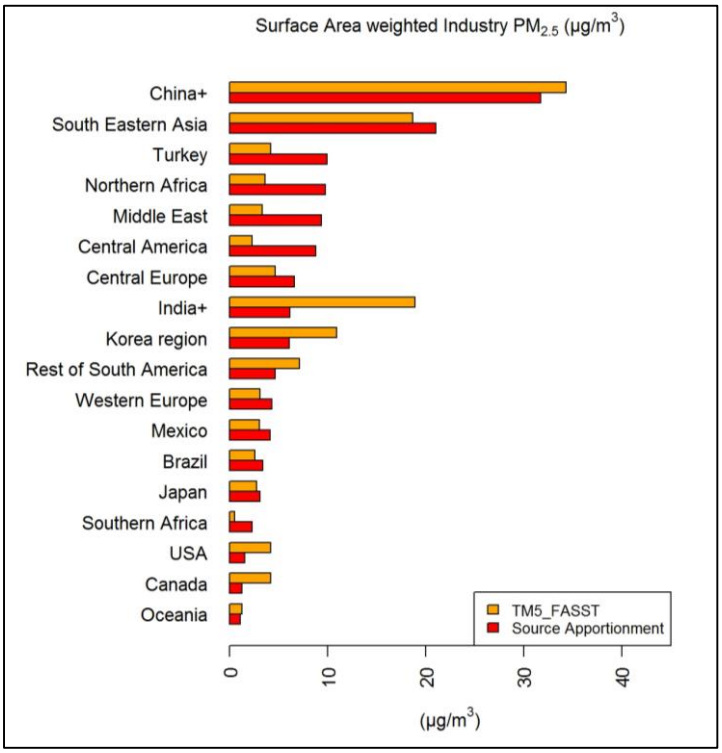

Figure 13c. Comparison between the area-weighted Industry $PM_{2.5}$ concentration regional averages from Source Apportionment case studies (up to 2014) and the TM5-FASST modelled data for Total $PM_{2.5}$ (year 2010).

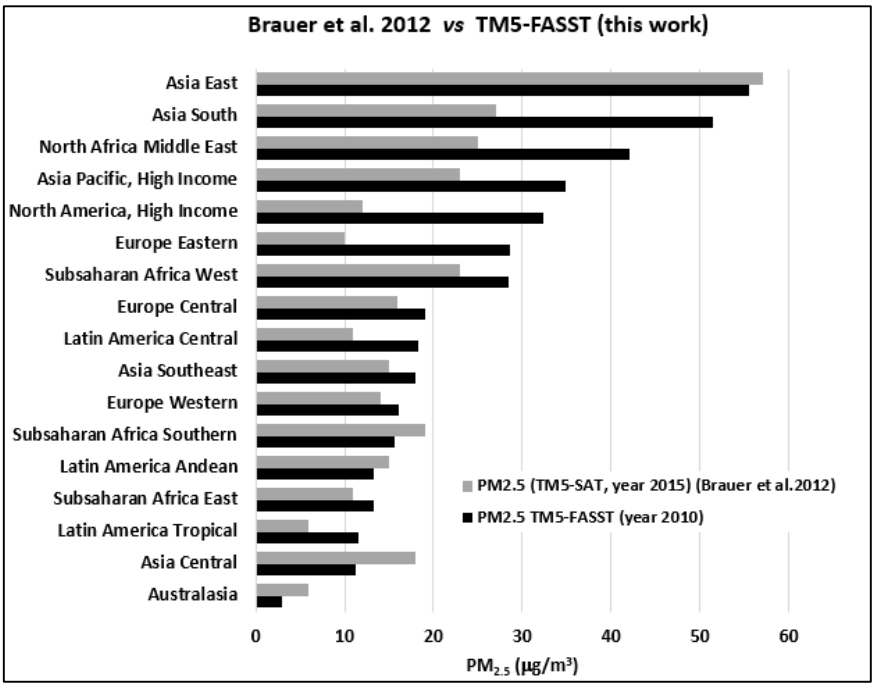

Figure 14. Comparison between the population-weighted Total $PM_{2.5}$ concentration averages from the TM5 model-satellite combination from Brauer et al. 2012 (data for year 2005) and the TM5-FASST modelled data for Total $PM_{2.5}$ (data for year 2010).

A1. Appendix. $PM_{2.5}$ sub-gridding parameterizations

For health impact studies, the TM5-FASST model includes a parameterization to redistribute the grid-cell area-averaged $PM_{2.5}$ concentration (~ 100 km x 100 km) to a finer scale resolution (0.125° x 0.125°, ~ 12.5 km x 12.5 km) in order to approach better the population exposure in urban areas (Leitão et al., 2013). This is accomplished by accounting for sub-grid gradients in population distribution and urban population fraction, and making assumptions on the locally enhanced emissions of transport and residential sectors. We chose the UN population dataset with a resolution of 0.125° x 0.125° to adjust the averaged (1° x 1°) $PM_{2.5}$ concentration population level (UN, 2011). Usually, highest pollutant concentrations are measured where population density is high. The sub-grid parameterization maintains the grid-cell average as obtained from the native 1° x 1° TM5-FASST resolution, but redistributes over the 64 sub-grids of each 1° x 1° native grid-cell, the concentration levels of primary $PM_{2.5}$ (BC, POM and other $PM_{2.5}$) from transport and residential sector making use of the known urban population fraction distribution within the 1° x 1° grid-cell. Secondary PM compounds, and primary $PM_{2.5}$ from other sectors are assumed to be homogenously distributed over the 1x1 grid cell. More details are available in Leitão et al., 2013.

Further work is on progress to implement a sub-gridding parameterization to reconstruct $PM_{2.5}$ sub-grid gradients within the 1° x 1° grid cell to a resolution of 0.1° x 0.1°. The basic assumption is that most of people lives in areas where $PM_{2.5}$ is produced, that is where anthropogenic activities occur.
A step forward has been done with the implementation of the TM5-FASST output with high-resolution satellite global estimate of $PM_{2.5}$. This has been possible by using the work carried out by van Donkelaar at al.(2010). In that work, global, satellite derived, annual average $PM_{2.5}$ surface concentrations (retrievals) were generated for the year 2001-2006 from daily Moderate Resolution Image SpectroRadiometer (MODIS) and a Multi-angle Imaging SpectroRadiometer (MISR) satellite instrument Aerosol Optical Dept (AOD). The geographic coverage of the van Donkelaar et al. (2010) dataset is nearly global with a horizontal resolution of 0.1° x 0.1° (~ 10 km x 10 km) at mid-latitudes.

While the TM5-FASST produces $PM_{2.5}$ grid-maps from (mostly) ground-based emissions, the satellite retrievals estimate $PM_{2.5}$ concentrations averaged for a column approximately up to 0.5-1 km above the earth surface, with GEOS-CHEM based vertical aerosol profiles scaling to the surface concentrations. Therefore, the sub-gridding procedure using satellite data might reflects some discontinuities between location where the emission occurs and the location where $PM_{2.5}$ satellite concentrations are retrieved. This could bring erroneous interpretations in the estimation of $PM_{2.5}$ concentration at urban location. Further work is then necessary in order to assure correct representation of urban conditions.

Preliminary simulations have been carried out to produce high resolution grid-maps at the resolution of 0.1° x 0.1° from down-gridding with satellite data. Population-weighted $PM_{2.5}$ averages calculated for all activity sectors at a resolution of 0.1° x 0.1° show to be consistent with the one calculated with the down-gridding accounting for gradients in population distribution.

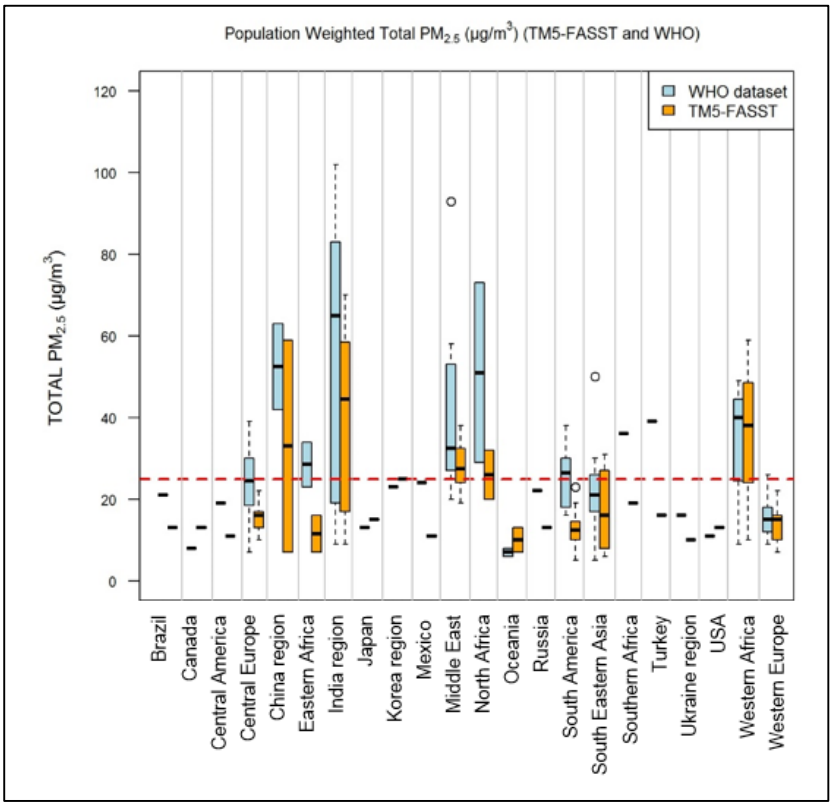

Figure A1a. Comparison between the WHO Ambient Air Pollution (AAP) dataset (2014) representative for 2008-2013 and TM5-FASST data for population-weighted averaged Total $PM_{2.5}$ country concentrations. Boxplots represent the median value (middle line), the upper and lower quartile (box), the minimum and maximum of all the data (whiskers) and the outliers (circles). Numbers indicate the amount of data available in each region. Red dashed line is the 25µg/m^3 European limit value for Total $PM_{2.5}$.

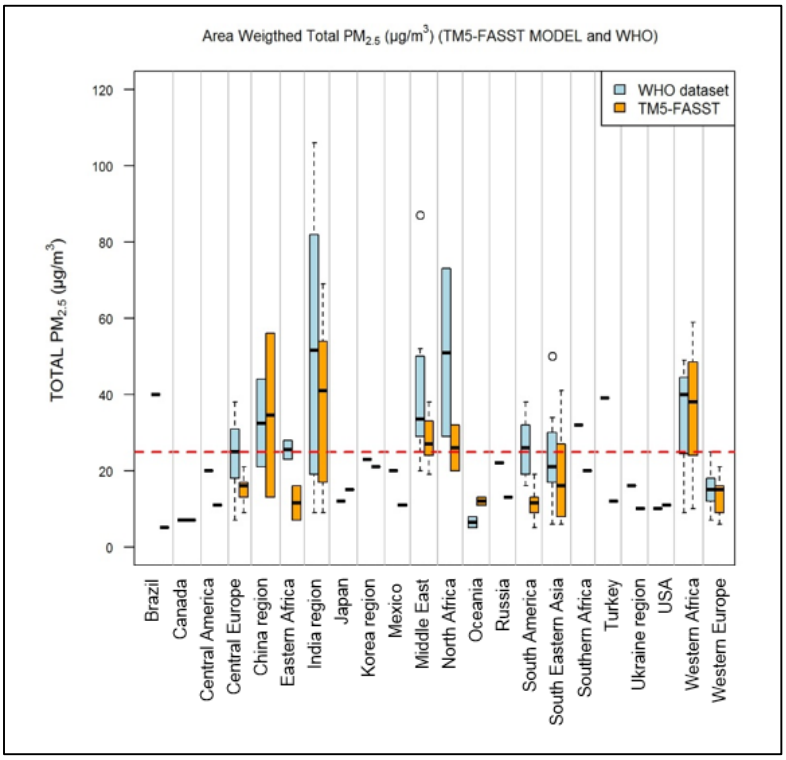

Figure A1b. Comparison between the WHO Ambient Air Pollution (AAP) dataset (2014) representative for 2008-2013 and TM5-FASST data for area-weighted averaged Total $PM_{2.5}$ country concentrations.

Country region	**$PM_{2.5}$ WHO (Pop.weighted)**	**$PM_{2.5}$ TM5-FASST (Area-weighted)**	**$PM_{2.5}$ WHO (Pop.weighted)**	**$PM_{2.5}$ TM5-FASST (Area-weighted)**
Oceania	6.63	11.79	6.26	8.86
Canada	7.27	6.51	7.66	12.56
USA	9.92	11.16	10.95	13.29
Japan	12.48	14.92	12.64	14.65
Ukraine Region	16.11	9.72	16.10	9.71
Western Europe	16.63	14.18	16.79	14.39
Central America	19.77	11.19	19.45	11.37
Brazil	40.31	5.01	21.06	12.72
South Eastern Asia	19.70	18.53	21.76	18.88
Russia	21.81	13.13	21.81	13.13
Korea region	22.87	20.74	23.05	24.77
Mexico	20.13	11.47	23.64	11.13
Rest of South America	25.38	10.70	24.63	12.53
Central Europe	26.87	16.37	26.27	16.82
Eastern Africa	27.01	14.29	28.67	11.32
Middle East	50.54	28.95	35.48	32.15
Southern Africa	31.97	19.66	36.19	19.33

Turkey	39.46	12.39	39.19	15.70
China+	38.33	45.49	41.99	58.92
Western Africa	45.20	44.74	44.11	49.43
Northern Africa	61.78	28.63	65.87	29.66
India+	57.63	48.35	70.92	59.47

Table T1. Data values for population and area weighted $PM_{2.5}$ concentration regonal averages calculated for the WHO Ambient Air Pollution (AAP) dataset (2014) representative for 2008-2013 and TM5-FASST modelled $PM_{2.5}$ concentrations (year 2010).

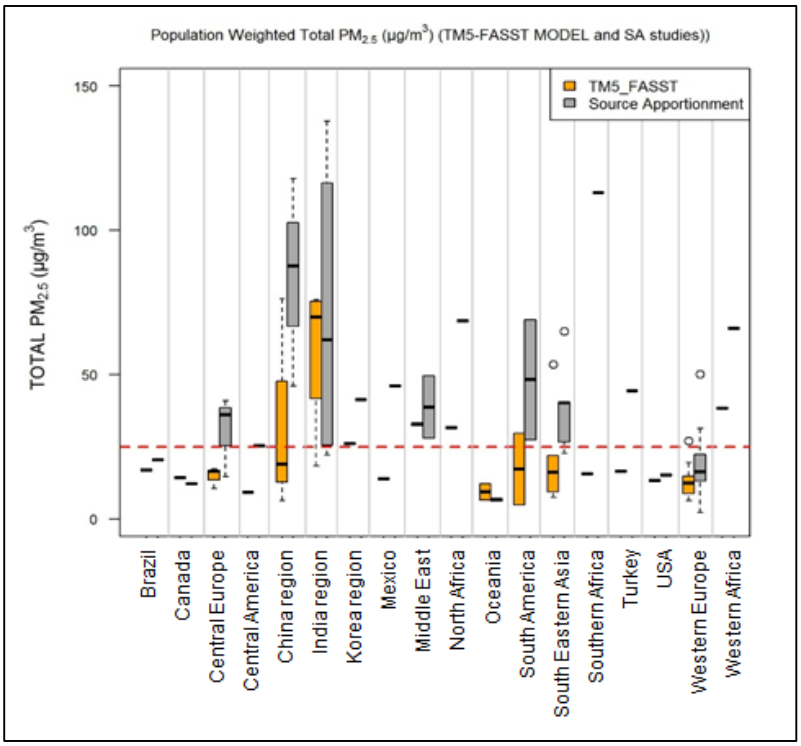

Figure A2a. Comparison between TM5-FASST and Source Apportionment data for population-weighted Total $PM_{2.5}$ country averages concentrations.

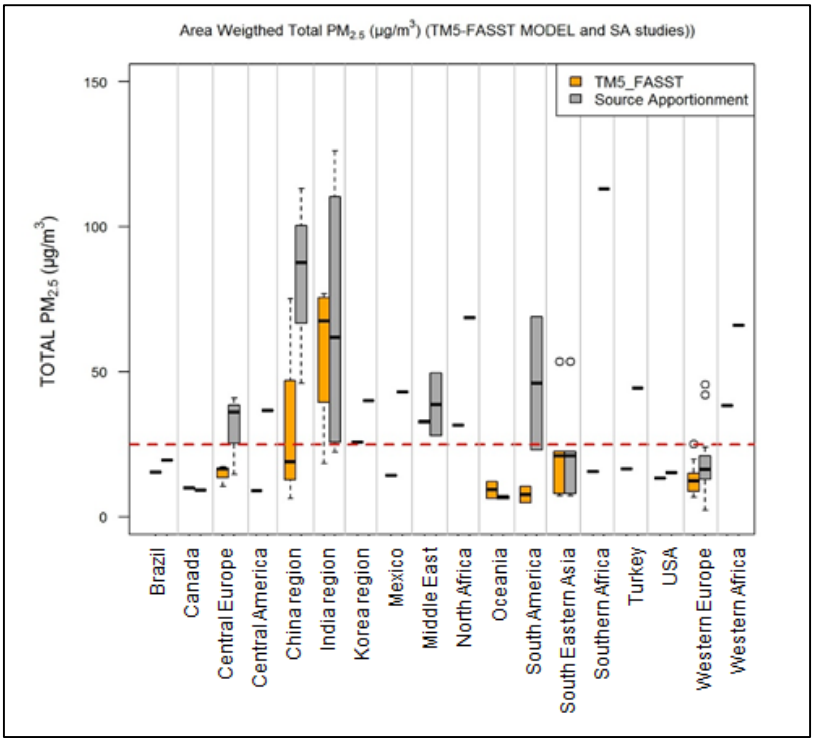

Figure A2b. Comparison between TM5-FASST and Source Apportionment data for area-weighted Total $PM_{2.5}$ country averages concentrations.

Country Region	**$PM_{2.5}$ SA (Pop.weighted)**	**$PM_{2.5}$ TM5-FASST (Area-weighted)**	**$PM_{2.5}$ SA (Pop.weighted)**	**$PM_{2.5}$ TM5-FASST (Area-weighted)**
Oceania	7.18	6.49	7.21	6.39
Japan	9.00	15.00	9.00	15.09
Canada	12.08	14.18	9.22	9.82
USA	15.26	13.38	14.71	12.43
Brazil	20.36	16.84	19.47	15.44
Central America	25.36	9.13	36.62	9.02
Western Europe	26.96	17.38	19.72	17.20
Middle East	28.25	33.26	30.28	33.17
Rest of South America	35.45	24.83	31.59	9.39
Central Europe	36.36	16.43	38.25	16.52
Korea region	41.37	26.09	40.05	25.70
Turkey	44.25	16.42	44.25	16.42
South Eastern Asia	44.91	29.61	51.97	37.88
Mexico	45.96	13.94	43.10	14.16
Western Africa	66.00	38.34	66.00	38.34
Northern Africa	68.65	31.58	68.65	31.58
Southern Africa	113.00	15.53	113.00	15.53

China+	116.55	74.25	112.41	74.27
India+	135.21	98.82	120.07	79.43

Table T2. Data values for population and area weighted $PM_{2.5}$ concentration regonal averages calculated for Source Apportionment Total $PM_{2.5}$ concentrations (up to year 2014) and TM5-FASST modelled $PM_{2.5}$ concentrations (year 2010).

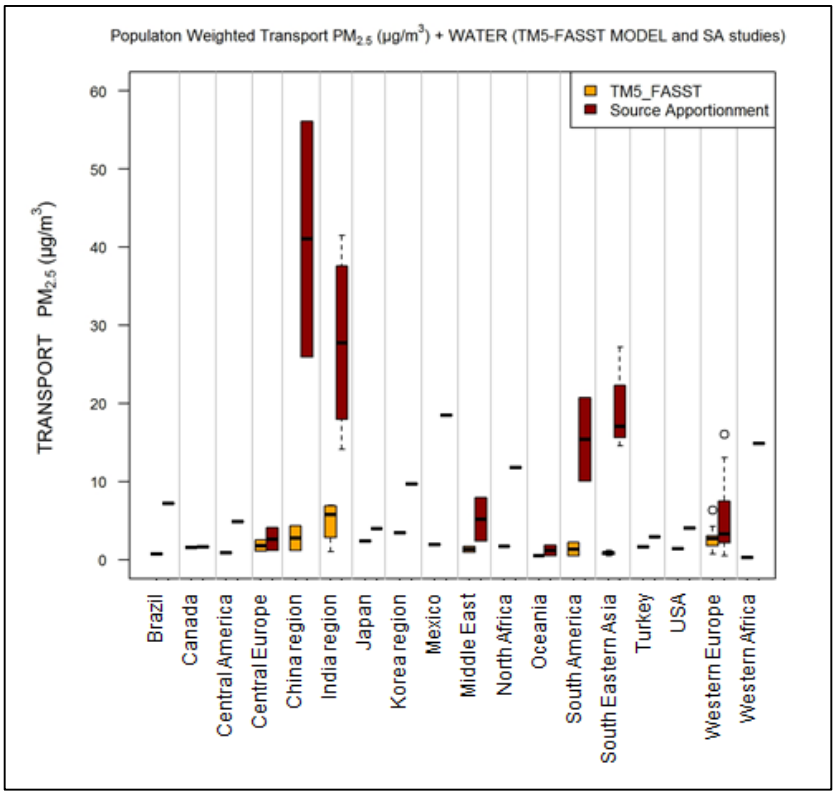

Figure A3a. Comparison between TM5-FASST and Source Apportionment data for population-weighted Transport $PM_{2.5}$ country averages concentrations.

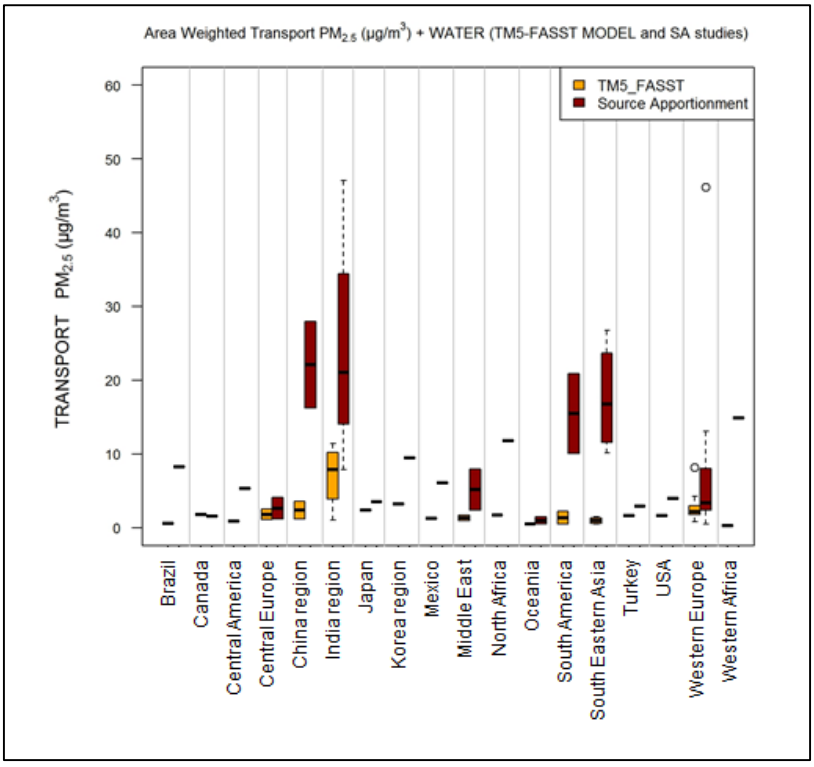

Figure A3b. Comparison between TM5-FASST and Source Apportionment data for area-weighted Transport $PM_{2.5}$ country averages concentrations.

Country Region	Transport $PM_{2.5}$ SA (Pop.weighted)	Transport $PM_{2.5}$ TM5-FASST (Area-weighted)	Transport $PM_{2.5}$ SA (Pop.weighted)	Transport $PM_{2.5}$ TM5-FASST (Area-weighted)
Canada	1.66	1.57	1.56	1.80
Oceania	1.84	0.50	1.45	0.50
Middle East	2.42	1.72	6.36	1.19
Japan	3.96	2.37	3.50	2.35
USA	4.07	1.42	3.99	1.64
Central America	4.90	0.86	5.31	0.86
Turkey	6.28	1.67	6.28	1.67
Brazil	7.20	0.74	8.26	0.61
Western Europe	7.74	3.77	9.57	3.54
Korea region	9.70	3.42	9.46	3.23
Central Europe	11.52	2.56	10.31	2.54
Northern Africa	11.76	1.69	11.76	1.69
Rest of South America	12.14	1.90	19.48	0.73
Western Africa	14.85	0.31	14.85	0.31
South Eastern Asia	15.00	1.14	16.85	0.98
Mexico	18.45	1.92	6.05	1.28
China+	26.85	4.23	27.39	3.51

India+	35.90	6.62	38.09	10.18

Table T3. Data values for population and area weighted $PM_{2.5}$ concentration regional averages calculated for Source Apportionment Transport $PM_{2.5}$ concentrations (up to year 2014) and TM5-FASST modelled $PM_{2.5}$ concentrations (year 2010).

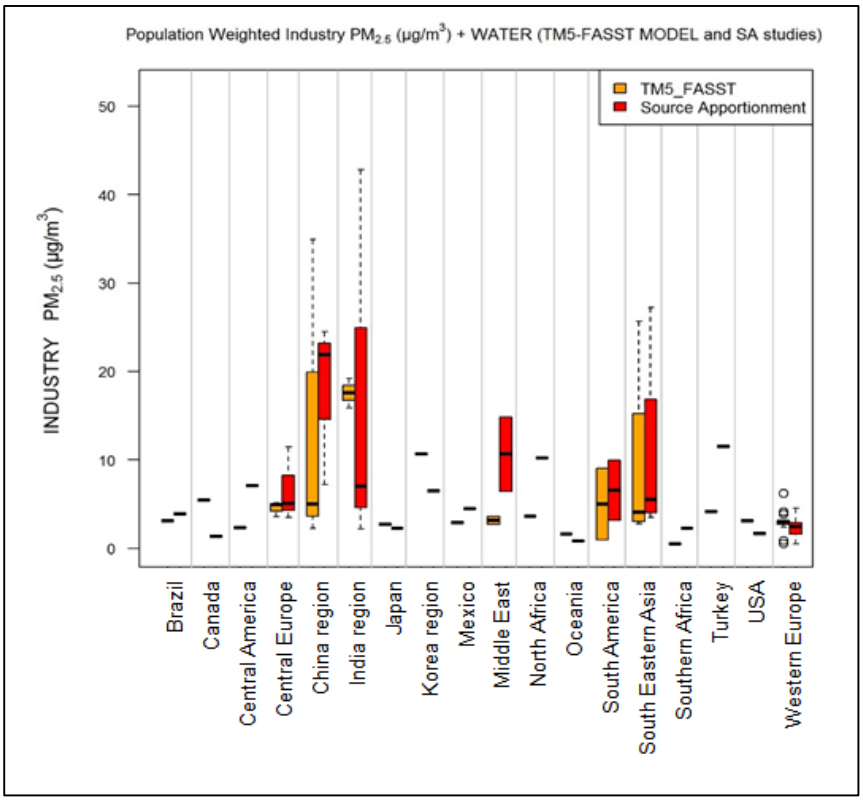

Figure A4a. Comparison between TM5-FASST and Source Apportionment data for population-weighted Industry $PM_{2.5}$ country averages concentrations.

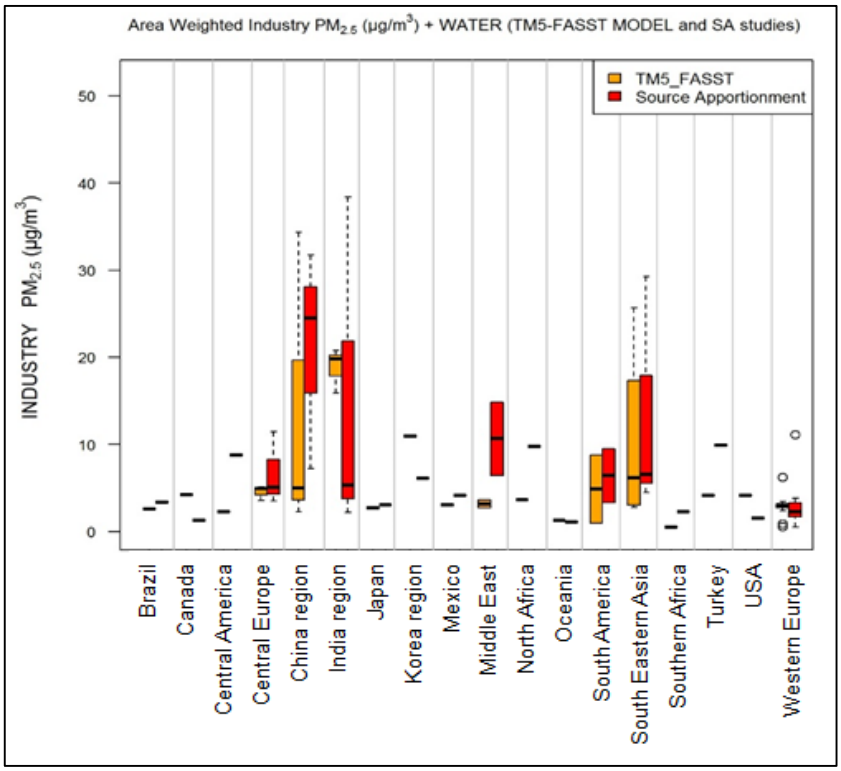

Figure A4b. Comparison between TM5-FASST and Source Apportionment data for area-weighted Industry $PM_{2.5}$ country averages concentrations.

Country Region	**Industry $PM_{2.5}$ SA (Pop.weighted)**	**Industry $PM_{2.5}$ TM5-FASST (Area-weighted)**	**Industry $PM_{2.5}$ SA (Pop.weighted)**	**Industry $PM_{2.5}$ TM5-FASST (Area-weighted)**
Oceania	0.86	1.59	1.06	1.26
Canada	1.36	5.47	1.27	4.19
USA	1.70	3.10	1.52	4.15
Southern Africa	2.26	0.50	2.26	0.50
Japan	2.29	2.74	3.06	2.74
Western Europe	2.71	3.89	4.30	3.11
Brazil	3.88	3.08	3.35	2.56
Mexico	4.47	2.92	4.14	3.02
Rest of South America	5.36	6.44	4.62	7.14
Korea region	6.52	10.70	6.10	10.92
Central America	7.11	2.30	8.77	2.26
Central Europe	7.62	4.45	6.60	4.66
Northern Africa	10.20	3.62	9.77	3.62
Turkey	11.54	4.18	9.91	4.18
South Eastern Asia	14.67	14.37	21.01	18.68
India+	17.49	17.91	6.14	18.92
China+	21.90	34.94	31.75	34.34

Middle East	42.48	15.26	9.38	3.33

Table T4. Data values for population and area weighted $PM_{2.5}$ concentration regonal averages calculated for Source Apportionment Industry $PM_{2.5}$ concentrations (up to year 2014) and TM5-FASST modelled $PM_{2.5}$ concentrations (year 2010).

Region	$PM_{2.5}$ TM5 (year 2000) (Brauer et al.2012)	$PM_{2.5}$ AVG (TM5-SAT) (Brauer et al.2012)	$PM_{2.5}$ TM5-FASST (year 2010)
Australasia	8	6	3
Asia Central	20	18	11
Latin America Tropical	7	6	12
Subsaharan Africa East	12	11	13
Latin America Andean	13	15	13
Subsaharan Africa Southern	22	19	16
Europe Western	14	14	16
Asia Southeast	14	15	18
Latin America Central	12	11	18
Europe Central	16	16	19
Subsaharan Africa West	24	23	28
Europe Eastern	11	10	29
North America, High Income	13	12	32
Asia Pacific, High Income	27	23	35
North Africa Middle East	26	25	42
Asia South	23	27	51
Asia East	56	57	55

Caribbean			add to Central America
Oceania			add to Asia south east
Latin America Southern			added to Latin America Andean
Sub-Saharan Africa Southern			include Sub-Saharan Africa Central

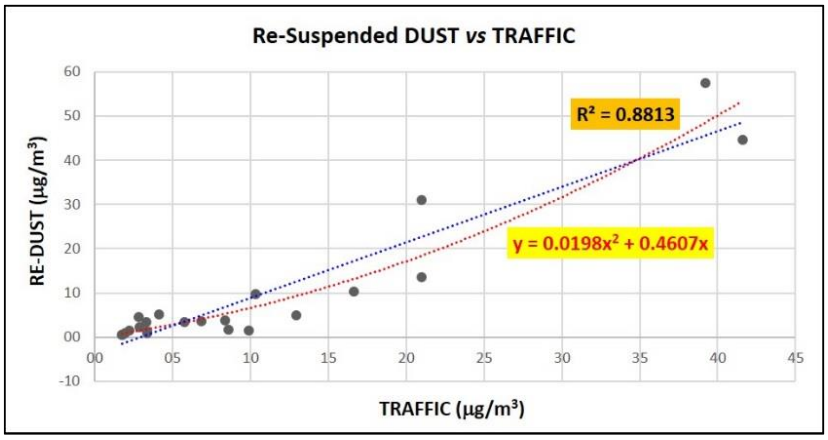

Figure A5. Relationship between Re-Suspended Dust and Traffic $PM_{2.5}$ concentrations found from Source Apportionment studies

Table T5. Population –weighted sectorial and total anthropogenic $PM_{2.5}$ calculated by TM5-FASST for world countries.

Population – weighted $PM_{2.5}$ fraction average ($\mu g/m^3$)	TRAN-SPORT	RESIDEN-TIAL	INDUSTRY	ENERGY	BIOMASS BURNING	AGRICUL-TURE	DUST	SEA SALT	Total Anthro-pogenic $PM_{2.5}$
Afghanistan	0.65	1.91	1.70	2.03	0.31	0.87	8.05	0.20	7.47
Albania	1.82	4.20	2.46	4.59	1.18	2.46	1.58	0.76	16.71
Algeria	0.48	1.05	1.67	2.07	0.86	1.00	4.67	2.79	7.13
Angola	0.04	5.03	5.34	4.93	10.67	4.69	0.07	1.19	30.70
Argentina	0.51	0.71	1.83	1.59	3.18	1.35	0.02	1.07	9.17
Armenia	0.75	1.56	3.11	3.54	3.64	2.32	3.53	0.47	14.92
Australia	0.15	0.32	1.33	0.70	0.95	0.55	0.09	2.81	4.00
Austria	2.49	3.57	2.49	2.77	1.50	5.74	0.36	0.58	18.56
Azerbaijan	0.59	1.23	3.04	3.24	3.28	1.85	6.49	1.77	13.23
Bahamas	0.15	0.67	1.03	1.39	0.83	0.85	1.20	7.57	4.92
Bahrain	1.99	2.20	4.91	8.04	2.35	1.85	17.03	3.58	21.34
Bangladesh	4.02	30.90	9.75	9.28	4.93	1.75	1.83	0.41	60.63
Belarus	1.18	1.85	2.98	2.78	1.84	3.00	0.39	0.54	13.63
Belgium	3.34	2.00	2.97	1.88	0.61	9.47	0.15	1.25	20.27
Belize	0.07	0.43	0.36	0.62	0.13	0.15	1.04	2.11	1.76
Benin	0.15	11.17	2.25	2.08	4.02	1.43	11.33	0.54	21.10
Bhutan	0.62	3.53	2.16	2.56	1.13	1.10	0.81	0.05	11.10
Bolivia	0.23	1.13	1.93	1.31	5.03	1.13	0.02	0.34	10.76
Bosnia-Herzegovina	1.86	2.99	1.93	3.60	0.83	3.04	0.87	0.66	14.25
Botswana	0.18	3.25	4.08	5.36	5.46	3.52	0.42	0.80	21.85
Brazil	0.41	0.77	2.09	1.17	5.97	1.20	0.05	1.18	11.61
Brunei Darussalam	0.69	0.90	2.57	2.18	1.98	1.88	0.03	1.59	10.20
Bulgaria	2.33	3.25	3.49	5.37	1.41	2.81	1.05	0.68	18.66
Burkina Faso	0.11	7.25	1.40	1.47	2.12	0.80	16.35	0.33	13.15
Burundi	0.03	7.82	1.08	0.84	6.42	0.52	0.44	0.64	16.71
Cambodia	0.22	3.57	2.70	1.62	8.82	0.94	0.18	1.05	17.87

Cameroon	0.22	11.65	4.77	4.88	6.99	3.96	12.11	0.28	32.47
Canada	1.00	1.35	3.12	2.44	4.21	3.19	0.06	0.56	15.31
Cape Verde	0.09	1.85	1.74	1.86	1.68	1.63	41.06	12.05	8.85
Central African Republic	0.10	7.67	2.38	2.61	7.80	1.53	4.79	0.22	22.09
Chad	0.15	2.54	1.16	1.85	2.31	0.33	28.59	0.40	8.34
Chile	1.59	5.41	6.29	8.48	3.59	6.66	0.02	1.45	32.02
China	3.62	15.09	21.61	8.42	1.53	6.36	7.87	0.45	56.63
Colombia	0.39	1.17	3.48	1.76	4.09	1.50	0.45	0.98	12.39
Comoros	0.01	0.86	0.80	0.83	1.02	0.74	0.13	6.01	4.26
Congo	0.11	9.95	4.59	3.80	9.04	3.48	0.32	0.32	30.97
Democratic Republic Congo	0.05	7.63	2.72	2.42	7.83	2.06	0.41	0.31	22.71
Costa Rica	0.49	1.27	1.38	1.31	0.97	1.09	0.75	2.42	6.51
Ivory Coast	0.18	13.38	4.34	4.04	6.18	3.62	5.99	0.49	31.74
Croatia	1.76	2.72	1.67	2.61	0.69	3.45	0.56	0.70	12.90
Cuba	0.17	1.71	1.19	1.61	0.69	0.71	1.53	3.95	6.08
Cyprus	1.01	0.83	3.05	4.36	0.87	0.65	2.38	1.42	10.77
Czech Republic	2.66	3.60	1.98	2.58	0.94	5.85	0.29	0.68	17.61
Denmark	1.93	0.92	1.25	1.42	0.60	2.64	0.06	1.12	8.76
Djibouti	0.40	3.21	3.96	5.21	3.14	2.98	11.71	1.27	18.90
Dominican Republic	0.27	2.96	1.72	1.81	1.08	1.07	3.41	3.23	8.91
East Timor	0.03	0.07	0.41	0.14	0.16	0.01	0.09	3.71	0.82
Ecuador	0.28	1.40	4.41	2.41	3.96	2.00	0.04	1.43	14.46
Egypt	1.07	1.33	2.66	4.18	0.65	6.63	7.72	1.48	16.52
El Salvador	0.31	4.66	1.62	1.41	0.43	0.62	0.78	1.49	9.05
Equatorial Guinea	0.09	6.68	3.51	3.48	5.59	3.12	2.17	0.49	22.47
Eritrea	0.36	1.22	1.78	3.77	0.69	0.36	12.14	1.35	8.18
Estonia	1.03	2.72	3.01	3.03	2.70	3.95	0.23	1.48	16.44
Ethiopia	0.18	5.51	1.74	2.60	4.16	0.55	6.14	0.80	14.74
Fiji	0.01	0.02	0.05	0.05	0.03	0.02	0.01	6.92	0.18
Finland	0.76	1.42	1.51	1.39	1.18	1.94	0.15	0.97	8.20
France	3.36	3.36	3.79	2.80	1.87	6.12	0.31	1.38	21.30
French Guiana	0.05	0.77	0.66	0.64	0.80	0.60	6.74	2.86	3.52
French Polynesia	0.00	0.01	0.01	0.01	0.01	0.01	0.01	7.23	0.05
Gabon	0.15	7.80	6.36	6.24	8.50	5.96	0.83	0.54	35.01

Gambia	0.25	10.63	6.39	6.44	6.93	6.00	32.46	1.08	36.64
Georgia	0.22	0.96	2.01	2.10	2.79	1.18	2.56	0.63	9.26
Germany	3.07	1.57	2.16	2.13	0.81	6.07	0.24	1.19	15.81
Ghana	0.16	14.06	3.45	3.10	5.61	2.57	7.98	0.72	28.95
Greece	1.88	1.44	1.93	4.54	0.49	0.98	1.64	1.46	11.26
Guadeloupe	0.12	0.63	0.62	0.68	0.54	0.57	6.57	9.11	3.16
Guam	0.01	0.05	0.07	0.06	0.04	0.01	0.19	8.39	0.24
Guatemala	0.52	7.05	2.67	2.36	1.19	1.38	0.67	1.17	15.17
Guinea	0.14	9.01	1.86	1.74	3.23	1.26	12.62	0.73	17.24
Guinea-Bissau	0.14	7.55	1.78	1.75	2.70	1.29	23.97	1.30	15.21
Guyana	0.02	0.32	0.19	0.12	0.57	0.06	4.80	2.82	1.28
Haiti	0.40	4.56	2.16	2.31	1.00	0.96	2.69	2.97	11.39
Honduras	0.20	2.92	1.59	1.52	1.00	1.01	1.08	2.20	8.24
Hong Kong	0.66	8.57	12.37	1.79	-2.11	-1.11	1.62	0.99	20.17
Hungary	2.19	3.22	2.45	3.22	1.39	4.18	0.37	0.52	16.65
Iceland	0.10	0.08	0.21	0.17	0.17	0.20	0.01	6.17	0.93
India	5.40	21.12	9.25	11.50	3.69	1.75	3.98	0.74	52.71
Indonesia	0.41	2.21	2.97	2.48	2.45	1.96	0.04	2.56	12.48
Iran	2.18	0.63	3.06	4.98	0.79	3.06	21.25	0.78	14.70
Iraq	2.36	0.74	3.40	5.75	0.66	1.90	7.92	0.51	14.81
Ireland	0.81	0.43	0.54	0.65	0.25	1.43	0.04	4.47	4.11
Israel	1.10	1.22	3.49	6.79	1.26	2.82	5.51	1.64	16.68
Italy	4.73	4.31	2.29	2.28	0.96	6.09	0.95	1.47	20.66
Jamaica	0.33	1.54	1.47	1.83	1.04	1.15	2.48	4.44	7.36
Japan	1.76	1.48	2.47	1.45	0.46	3.01	1.21	1.70	10.63
Jordan	1.05	1.02	2.98	6.00	1.02	2.60	7.30	1.30	14.67
Kazakhstan	0.36	0.82	2.18	1.74	0.92	1.30	7.82	0.20	7.32
Kenya	0.07	3.94	1.24	1.34	3.33	0.79	0.95	1.37	10.71
Dem. People's Rep. of Korea	1.77	7.56	11.60	8.20	4.45	5.74	2.69	0.48	39.32
Korea	3.15	3.41	8.92	4.16	1.54	3.11	2.63	1.02	24.29
Kuwait	2.72	1.17	4.89	7.81	2.21	2.20	19.74	1.67	21.00
Kyrgyz Republic	0.68	1.53	3.11	1.56	0.62	3.55	9.47	0.15	11.05
Lao People's Democratic Republc	0.41	4.41	5.42	3.20	6.19	1.84	0.44	0.66	21.47

Latvia	1.46	4.53	4.27	4.41	3.93	5.26	0.24	1.17	23.86
Lebanon	1.22	0.97	2.94	5.71	0.95	2.72	4.51	1.31	14.51
Lesotho	0.09	1.93	0.71	3.04	0.83	0.31	0.31	1.19	6.91
Liberia	0.34	13.26	8.12	7.93	9.42	7.67	4.03	0.59	46.74
Libyan Arab Jamahiriya	0.52	0.61	0.88	1.72	0.25	0.45	11.13	3.41	4.43
Lithuania	1.60	4.46	3.94	4.12	3.37	5.23	0.28	0.99	22.72
Luxembourg	3.85	2.16	2.99	2.39	1.09	7.31	0.25	1.62	19.79
Macedonia	2.13	2.97	2.17	4.87	0.80	2.73	1.36	0.55	15.67
Madagascar	0.00	1.26	0.65	0.66	1.25	0.59	0.02	3.64	4.41
Malawi	0.02	1.22	0.92	0.88	4.10	0.65	0.04	1.32	7.79
Malaysia	0.75	0.97	2.60	1.99	1.68	1.24	0.05	1.67	9.23
Mali	0.11	5.52	2.10	2.27	2.45	1.68	27.10	0.40	14.13
Martinique	0.08	0.67	0.63	0.69	0.56	0.58	7.51	9.92	3.21
Mauritania	0.12	2.36	1.93	2.14	1.84	1.76	118.97	1.01	10.15
Mauritius	0.00	0.03	0.04	0.05	0.08	0.03	0.01	11.86	0.23
Mexico	0.96	1.58	1.48	2.47	1.23	1.72	0.43	0.82	9.44
Federated State of Micron	0.00	0.02	0.02	0.03	0.02	0.00	0.05	6.28	0.09
Republic of Moldova	1.89	2.28	4.36	3.43	1.27	2.75	0.83	0.60	15.98
Mongolia	0.24	1.10	1.88	1.70	1.20	0.93	5.70	0.06	7.05
Morocco (includes Western	0.47	1.38	1.92	2.66	0.87	1.10	5.28	1.49	8.40
Mozambique	0.02	0.99	0.86	1.06	2.48	0.68	0.04	1.32	6.09
Myanmar	0.79	9.37	7.84	7.67	17.12	4.79	1.02	0.70	47.58
Namibia	0.10	1.74	2.20	2.20	4.58	1.79	0.30	1.18	12.61
Nepal	2.12	13.16	5.32	5.40	2.39	2.58	1.72	0.14	30.97
Netherlands	2.73	1.55	2.49	1.76	0.60	9.03	0.12	1.16	18.16
New Caledonia	0.01	0.02	0.11	0.08	0.03	0.03	0.04	8.55	0.28
New Zealand	0.08	0.31	0.54	0.49	0.47	0.60	0.00	8.53	2.49
Nicaragua	0.34	2.03	1.20	1.12	0.80	0.80	0.88	1.71	6.29
Niger	0.12	3.49	0.96	1.28	1.14	0.56	26.18	0.43	7.55
Nigeria	0.25	14.72	5.39	5.27	7.14	4.48	15.52	0.43	37.25
Norway	0.85	2.59	1.72	1.34	1.20	1.49	0.03	1.23	9.19
Oman	0.74	0.81	2.08	3.97	0.52	0.45	15.57	2.50	8.57
Pakistan	4.68	20.45	8.45	8.07	3.45	3.36	6.78	0.66	48.46
Panama	0.18	1.14	1.57	1.24	0.88	0.67	0.77	1.88	5.68

Papua New Guinea	0.03	0.14	0.07	0.09	0.06	0.07	0.01	1.71	0.46
Paraguay	0.23	1.54	2.31	1.77	4.85	1.64	0.01	0.33	12.34
Peru	0.26	1.74	3.91	2.73	4.14	2.55	0.02	1.02	15.33
Philippines	0.29	1.62	1.99	1.67	2.32	1.54	0.17	1.46	9.43
Poland	1.87	3.62	2.60	3.09	1.74	5.65	0.23	0.94	18.57
Portugal	1.05	0.87	1.42	0.92	0.49	1.11	0.81	1.48	5.86
Puerto Rico	0.01	0.03	0.04	0.16	0.01	0.08	4.29	3.47	0.33
Qatar	1.17	0.78	2.91	5.80	0.46	0.10	17.28	3.03	11.22
Reunion	0.00	0.18	0.22	0.24	0.27	0.22	0.01	10.79	1.13
Romania	2.02	4.95	5.07	5.58	2.84	4.76	0.80	0.57	25.22
Russia	0.58	1.25	2.38	2.53	2.95	2.02	1.32	0.40	11.71
Rwanda	0.06	13.38	6.18	5.95	12.05	5.55	0.66	0.70	43.17
Saint Lucia	0.07	0.57	0.51	0.57	0.45	0.46	7.62	10.06	2.63
Western Samoa	0.00	0.00	0.01	0.01	0.01	0.00	0.01	6.25	0.03
Sao Tome and Principe	0.02	1.59	0.42	0.22	2.76	0.00	0.95	1.24	5.01
Saudi Arabia	1.25	1.00	3.16	6.17	0.86	0.77	38.99	1.06	13.21
Senegal	0.14	4.85	1.42	1.50	1.76	1.08	43.65	1.26	10.75
Sierra Leone	0.09	8.44	0.84	0.64	2.32	0.26	7.19	0.72	12.59
Singapore	1.39	4.46	7.69	7.07	6.64	5.50	0.02	1.98	32.75
Slovakia	2.07	3.48	2.05	2.63	1.05	4.18	0.32	0.55	15.46
Slovenia	1.62	2.25	1.24	1.56	0.37	3.80	0.46	0.64	10.84
Solomon Islands	0.00	0.01	0.02	0.01	0.01	0.01	0.01	3.25	0.06
Somalia	0.18	1.82	1.95	2.43	1.70	1.43	2.54	1.29	9.51
South Africa	0.18	4.82	3.37	5.89	3.66	2.57	0.38	1.70	20.49
Spain	2.10	1.68	2.84	2.17	1.29	2.32	1.21	1.86	12.40
Sri Lanka	0.67	3.27	2.27	3.24	1.04	0.39	1.62	2.28	10.88
Sudan	0.32	2.40	1.73	2.80	1.98	0.75	12.28	0.58	9.98
Suriname	0.01	0.26	-0.01	-0.04	0.17	-0.08	5.13	1.19	0.31
Swaziland	0.04	1.68	0.55	2.10	1.56	0.00	0.12	1.15	5.93
Sweden	1.06	1.44	1.59	1.63	1.21	1.89	0.05	1.25	8.82
Switzerland	2.64	1.78	1.83	1.55	0.74	7.11	0.45	0.83	15.65
Syrian Arab Republic	1.63	1.02	3.52	5.43	1.06	2.35	3.82	0.78	15.01
Taiwan	1.10	1.94	4.65	2.21	1.23	1.90	1.65	3.22	13.03
Tajikistan	0.58	1.30	2.77	1.50	0.77	3.59	5.31	0.16	10.51

United Rep. of Tanzania	0.03	3.30	1.29	1.29	3.10	1.00	0.45	1.24	10.01
Thailand	0.93	2.05	4.96	3.36	5.15	1.21	0.33	0.76	17.66
Togo	0.15	12.41	2.51	2.27	4.47	1.65	10.65	0.57	23.46
Tonga	0.00	0.01	0.03	0.01	0.02	0.01	0.01	10.61	0.08
Trinidad and Tobago	0.02	0.27	0.11	0.14	0.08	0.03	6.19	6.79	0.65
Tunisia	0.81	1.07	1.57	2.36	0.70	0.85	3.79	3.71	7.36
Turkey	1.59	1.59	3.96	4.53	1.86	2.55	2.07	0.82	16.08
Turkmenistan	0.36	0.80	1.70	1.66	0.50	2.07	48.71	0.49	7.09
Uganda	0.06	7.31	1.04	1.00	5.92	0.35	0.98	0.97	15.68
Ukraine	1.62	2.19	4.47	3.85	2.14	3.25	0.89	0.59	17.52
United Arab Emirates	1.34	0.91	3.14	5.47	0.86	0.72	16.00	1.30	12.44
United Kingdom	2.07	1.64	2.18	1.83	0.99	5.26	0.07	1.25	13.97
United States of America	1.66	1.37	3.38	4.24	2.28	4.06	0.20	0.76	16.99
Uruguay	0.32	1.23	2.45	2.34	3.00	2.24	0.01	1.19	11.58
Uzbekistan	0.66	1.63	3.27	1.76	0.95	5.79	12.11	0.23	14.06
Vanuatu	0.01	0.02	0.06	0.05	0.04	0.04	0.02	7.62	0.22
Venezuela	0.68	0.65	2.81	1.47	3.14	1.02	2.73	1.52	9.77
Viet Nam	0.52	7.10	5.78	3.11	5.66	2.23	0.71	1.26	24.40
Yemen	0.64	0.89	2.19	4.80	0.31	0.23	36.82	1.46	9.06
Zambia	0.03	1.53	1.09	0.85	4.91	0.60	0.04	0.53	9.01
Zimbabwe	0.05	1.58	1.67	1.76	4.04	1.32	0.04	1.05	10.42