European Commission

JRC TECHNICAL REPORTS

Attribution of anthropogenic $PM_{2.5}$ to emission sources

A global analysis of source-receptor model results and measured source-apportionment data

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2017






Joint Research Centre

EUR 28510 EN

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JRC Science Hub
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JRC104676

EUR 28510 EN

PDF ISBN 978-92-79-66599-8 ISSN 1831-9424 doi:10.2760/344371

Ispra: European Commission, 2017

How to cite this report: Karagulian F., Van Dingenen R., Belis C.A., Janssens-Maenhout G., Crippa M., Guizzardi D., Dentener F., *Attribution of anthropogenic* $PM_{2.5}$ *to emission sources*, EUR 28510 EN, doi 10.2760/344371

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Abstract / executive summary

Outdoor air pollution – in particular fine particulate matter, $PM_{2.5}$ - is a major issue of concern in the public health domain. Efficient control strategies depend on the understanding of the major sources contributing to the atmospheric $PM_{2.5}$ burden.

This report performs for the first time a comparison between $PM_{2.5}$ source apportionment results from in-situ point measurements, obtained from a screened literature review, and from global air quality modelling using the in-house developed global source-receptor air quality model TM5-FASST.

Both experimental and numerical modelling techniques have been proven to provide valuable information about the emitting sectors contributing to ambient PM2.5. JRC has specific expertise in the source attribution of in-situ measured $PM_{2.5}$ composition at selected spots using well-established and validated source-apportionment techniques, as well as in the global and regional scale bottom-up modelling of $PM_{2.5}$ levels and compounds, using sector-specific emission inventories as input. Both approaches address the issue of source apportionment, but at a completely different spatial scale and resolution. The global TM5-FASST model takes as input the EDGAR-HTAP V2 global air pollution emission inventory, separated by major economic sector, and computes corresponding pollutant concentrations by sector. The experimental source apportionment data are obtained with receptor modelling techniques applied on the observed in-situ chemical composition of PM2.5.

The outcome of the global model shows that in regions with the highest $PM_{2.5}$ levels (China, India, Western Africa), residential burning is the major contributing sector whereas in western Europe it is the agricultural sector, followed by road transport.

A comparison between the global model and regionally aggregated in-situ source apportionment results shows a reasonable agreement in the $PM_{2.5}$ concentrations attributed to the industry sector. On the other hand, for the transport sector large discrepancies are found between the two methodologies. These discrepancies can be tentatively attributed to the different resolution at which the concentration levels are evaluated, and the limited spatial representativeness of some pollution hotspots for the in-situ measurements, but there may also be underlying uncertainties in the emission inventories used as input for the global model.

1 Introduction

It has become increasingly clear that environmental impacts of anthropogenic air pollutants are pressing problems for present and future generations. New estimates just released by the World Health Organization (WHO) confirm that air pollution is now the world's largest single environmental health risk (WHO, 2013). 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 premature deaths (WHO, 2014). The most recent Global Burden of Disease study (Forouzanfar et al., 2015) showed that air pollution from household solid fuel burning and outdoor particulate matter pose the highest environmental risk to human health worldwide, comparable to more widely appreciated risk factors such as tobacco smoking, alcohol consumption and obesity. They have been ranked as the seventh and eighth most important risk factors in global mortality respectively.

In Europe, outdoor air pollution from particulate matter and ozone is estimated to cause 230 000 – 400 000 premature deaths (IHME, 2016; Lelieveld et al., 2015) . Estimated loss of statistical life expectancy ranges between 1 and 2 months in clean background conditions (Sweden, Scotland) to 1.5 years and more in the Po valley, Poland, parts of Romania and Bulgaria (Kiesewetter et al., 2015). The current European Air Quality Directive specifies a target annual mean concentration value of 25µg/m³. This value largely exceeds levels considered safe by the World Health Organization who specify a guideline annual mean concentration of 10µg/m^3. Several European monitoring sites exceed the $PM_{2.5}$ limit value of 25µg/m³, and only a few stations currently meet the WHO guideline value. Urban concentrations of $PM_{2.5}$ are frequently far above the WHO guideline value.

Understanding the sources of these pollutants is crucial to identifying 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 with increased exposure to particulate matter (Burnett et al., 2014a; Schwartz et al., 2010). In particular, exposure to $PM_{2.5}$ has been associated with increased risk of premature death from ischemic heart disease, stroke, chronic obstructive pulmonary disease, acute lower respiratory infections and lung cancer (Burnett et al., 2014a) , while ozone has been associated with respiratory infections and disease(Jerrett et al., 2009).

The health impact of ambient pollution can almost entirely be attributed to fine airborne suspended particles ($PM_{2.5}$), composed of a mixture of organic and inorganic compounds, originating from a variety of anthropogenic sectors. 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). BC is a major component of emissions from fossil fuel combustion and biomass burning. OC is directly emitted from primary sources or produced by chemical reactions involving gaseous organic precursors (Pandis et al., 1992).

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 sulphur and nitrogen oxides, drive the formation of inorganic secondary $PM_{2.5}$ (Secondary Inorganic Aerosols, SIA) in the form of ammonium nitrate and sulphate, through successive photo-chemical reactions in air. Transformation of volatile and semi-volatile 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).

Primary $PM_{2.5}$ is closely associated with areas of high fossil-fuel combustion, with peak concentrations clearly linked to the emission areas. Secondary components of $PM_{2.5}$ are formed over longer time and spatial scales, providing a sufficient lifetime for the particulate end products to be distributed over a larger geographical area because of the photochemical and cloud-phase chemical processes involved, and thus having not only local but also regional, and even hemispheric impacts (Finlayson-Pitts and Pitts, 2000). In general, $PM_{2.5}$ at a given location is composed of local, regional and long-range contributions with relative magnitudes depending on the specific type of location, in particular its vicinity to emitting sources.

In this study, we use a global pollutant emission inventory and a source-receptor model for air pollutant formation and 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 (HTAPv2) has been used to calculate total emissions for each country and major economic sector, representative of the most important air pollution sources worldwide.

Present day and future emissions are ingested by JRC's source receptor model (TM5-FASST) which calculates atmospheric concentration attributed to each economic sector. To cross check the model output with independent data deriving from a different approach, 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 using receptor-oriented models. In this work, we compare gridded data (1°x1°) from the source receptor model with point source contribution estimates derived from measurements. Another factor contributing to the differences between the two approaches is that certain sources are better tracked by one methodology than the other e.g. dust is most likely to be tracked by receptor-oriented models and secondary inorganics are better seen by Chemistry-Transport Models (CTMs).

2 Methods

2.1 The global air quality model TM5-FASST

For this study we apply the global air quality source-receptor model TM5-FASST (FAst Scenario Screening Tool) to compute spatially resolved $PM_{2.5}$ concentrations attributed by sectors. Evaluating contributions from various sectors involves a large number of model runs (see below) for which we use TM5-FASST, an in-house developed reduced-form source receptor model (SRM) for global air quality and impacts. In this type of model, the relation between the emissions of a compound i from a source region x and the resulting pollutant j concentration (where $j = i$ in case of a primary component) at a receptor location y 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:

$$C_{ij}(x,y) = c_0(\mathrm{y}) + A_{ij}(x,y)E_i(x) \quad (1)$$

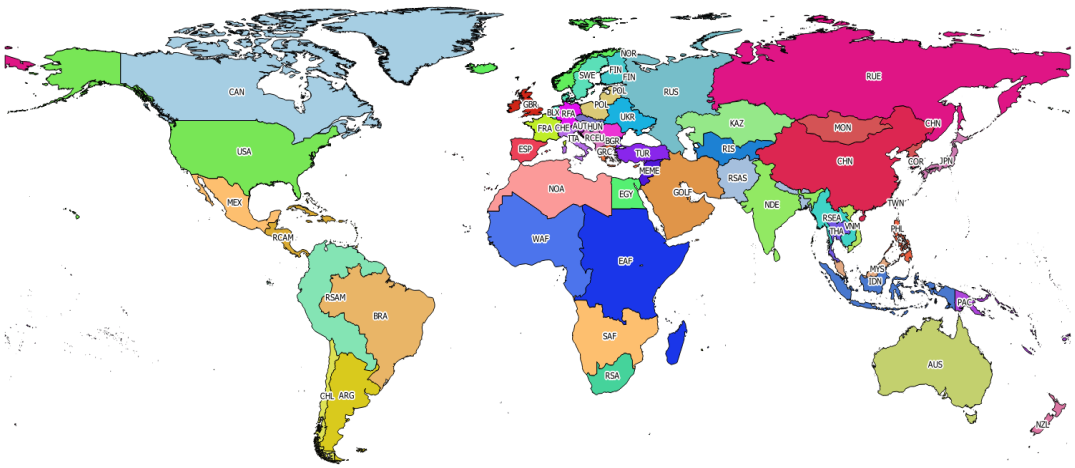
Where $C_{ij}(x,y)$ is the concentration of species j at the receptor region y formed from precursor i emitted at source region x, $E_i(x)$ is the emission rate (kg/yr) of precursor i at source region x, $A_{ij}(x,y)$ is the so-called source-receptor coefficient (SRC) between source location x and receptor region y for emitted precursor i leading to end product j, and $c_0(y)$ 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., 2006b) . E.g. SO_2 emissions, dominated by industrial and power generation, are typically emitted at a few hundred meters altitude whereas NO_x from ground transport is emitted in the first model layer (about 60 meters).

With n_x and n_y being the number of source and receptor regions respectively, the SRCs are stored as matrices with dimension $[n_x, n_y]$, and are available for each precursor and for each resulting component from that precursor. The SRCs have been derived from a set of dedicated runs with the full chemical transport model TM5-CTM (Krol et al., 2005) 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 (Figure 1). 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 aggregation to 23 larger regions is reported in Annex A1.

— A primary set of 360 x 180 receptor grid cells was defined at the TM5 model's native 1° x 1° resolution. A reference run with the IPCC AR5 representative concentration pathways (RCP)' global emission dataset for all relevant air pollutants for the year 2000 was performed, including sulphur dioxide (SO_2), nitrogen dioxide (NO_2), BC, OC (or primary organic matter POM), non-methane VOCs (NMVOC), and ammonia (NH_3). This run produces reference concentration maps of particulate 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.

Figure 1. Definition of the 56 source regions within TM5-FASST. EU27 is represented by 16 regions.



Source: JRC analysis

— 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 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 $\Delta C_{ij}(x,y)$:

$$\Delta C_{ij}(x,y) = C_j(y)|_{E_{i,x}^{Ref}} - C_j(y)|_{0.8 \cdot E_{i,x}^{Ref}} \quad (2)$$

with $E_{i,x}^{Ref}$ the emission strength of precursor *i* in source region *x*

— In addition 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*, resulting from arbitrary emissions $E_i(x)$ of *all* its precursors *i* at *all* source regions *x* is obtained by overlaying emission-scaled delta fields $\Delta C_{ij}^0(x,y)$ for all source regions (scaled on the 20% perturbation):

$$C_j(y) = C_j^0(y) + \sum_x \sum_i \Delta C_{ij}(x,y) \cdot \left[\frac{E_{i,x} - E_{i,x}^{Ref}}{0.2 E_{i,x}^{Ref}} \right] \quad (3)$$

The linear equations (3) with associated source-receptor matrices (SRC) constitute the kernel' of TM5-FASST, assuming that the emission-concentration sensitivities, determined from the 20% perturbation run, are valid for any emission perturbation on the reference run. The validity of this assumption was tested applying larger emission perturbations (-80%,+100%) on a limited set of representative regions. These tests indicate that TM5-FASST $PM_{2.5}$ values, based on the extrapolation of the 20%

perturbation, are reproducing the full CTM results within 1% for SO_4, BC and POM, and within 5% for NO_3 and NH_4 (Van Dingenen et al., manuscript in preparation).

The TM5-FASST model requires as input regionally aggregated emission data and returns 1°x1° resolution global pollutant concentration fields. Still it should be noted there is an implied fixed underlying 1° x 1° resolution gridding of the emissions (i.e. the spatial distribution of the emission fields used in the reference year 2000 run and perturbation calculations). Furthermore, 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 the resulting concentration fields that have been calculated with the full TM5 model from natural emissions (Dentener et al., 2006a) using meteorology for 2001. Dust is mainly responsible for country-averaged annual mean exposure levels above 50 µg/m³. Therefore, the performance of the model in the high $PM_{2.5}$ range end (above 120 µg/m³) is linked to the performance of the dust model.

The TM5-FASST tool produces as primary 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.

A specificl application of TM5-FASST further considers input emissions on a sector by sector basis. The contribution of each individual sector s_i to $PM_{2.5}$ concentration ($\mathrm{PM}_{2.5}(s_i)'$) 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:

$$\mathrm{PM}_{2.5}(s_i)' = PM_{2.5}(E_{tot}) - PM_{2.5}(E_{tot} - E_i) \tag{4}$$

The total $PM_{2.5}$ concentration includes the anthropogenic part, modelled from the full anthropogenic emission set, plus the natural components (mineral dust + sea salt), which are equally treated as a 'sector' contribution. Calculations of $PM_{2.5}$ 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 $PM_{2.5}$ components, like primary BC, POM, sea salt and dust are by definition behaving linearly. Also in this particular version of TM5, secondary organic aerosol as a product from NMVOC is not calculated.

Because of the linearized approach in TM5-FASST the sum over all sectors of the $\mathrm{PM}_{2.5}(s_i)$ does not exactly match $PM_{2.5}(E_{tot})$. We therefore 'calibrate' with a factor R_i the sectorial contributions as follows:

$$\mathrm{PM}_{2.5}(s_i) = R_i PM_{2.5}(E_{tot}) \tag{5}$$

$$R_i = \frac{\mathrm{PM}_{2.5}(s_i)'}{\sum_i \mathrm{PM}_{2.5}(s_i)'} \tag{6}$$

R_i values have been calculated per grid point at 1° x 1° resolution.

For this study, emissions segregated by sector are available, which allowed us to estimate the $PM_{2.5}$ concentrations from the Transport, Residential, Agriculture, Industry, Energy and Large Scale (LS) Biomass Burning sectors, as well as the contribution of natural mineral dust.

The native resolution of the TM5-FASST model corresponds to a nominal grid size of 1°x1° degree (nominally 100 km x 100 km) and hence calculated $PM_{2.5}$ concentrations are obtained as grid cell averages. Population exposure estimates have been improved by taking into account sub-grid pollutant gradients which tend to correlate with 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 products are gridded data at a 0.125°

x 0.125° resolution with adjusted (urban-incremented) $PM_{2.5}$ concentrations that nevertheless conserve native 1° x 1° $PM_{2.5}$ concentration.

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 $PM_{2.5}$ interpolated and urban-increment adjusted concentration for each sector at the highest available resolution for population grid maps (0.125° x 0.125°) (Leitão et al., 2014).

Sector specific $PM_{2.5}$ concentrations are population-weighted according to the following equation:

$$(Population\ Weighted\ PM_{2.5})_i = \frac{\sum_{i,j}[\mathrm{PM}_{2.5}(s_i)]\times N(population)_j}{\sum_j N(population)_j} \quad (7)$$

where $N(population)_j$ is the population number in gridcell j belonging to the country.

Population grid-maps for the year 2010 were obtained from the Gridded Population of the World (GPWv3) released by (Columbia University Center for International Earth Science Information Network (CIESIN), 2014).

2.2 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.

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 (Janssens-Maenhout et al., 2012, 2015). 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) grid-maps 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 has been harmonized and aggregated to provide total emissions (kg/Year) by country and activity 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: LS Biomass Burning includes agricultural (FAOSTAT) waste burning and forest fires from the Global Fire Emissions Database, version 3 (GFED3) (van der Werf et al., 2010).

Absolute and relative 2010 emissions of the main pollutant precursors for $PM_{2.5}$ concentrations are shown in Figure 2 and Figure 3. For the year 2010, the largest emission strength of pollutant is represented by CO with 1000 Tg/year, followed by NMVOCs with 150 Tg/year, NO_x with 135 Tg/year, SO_2 with 110 Tg/year, $PM_{2.5}$ with 85 Tg/year, NH_3 with 50 Tg/year, POM with 44 Tg/year and, BC with 8 Tg/year (Figure 2)

Relative contributions of the above pollutants to the sector contributions are shown in Figure 3. Main findings showed that worldwide:

— Transport sector contributes to 31% NO_x, 16% CO and 12% BC emissions.

— Energy sector contributes to 41% SO_2 and 21% NO_x emissions.

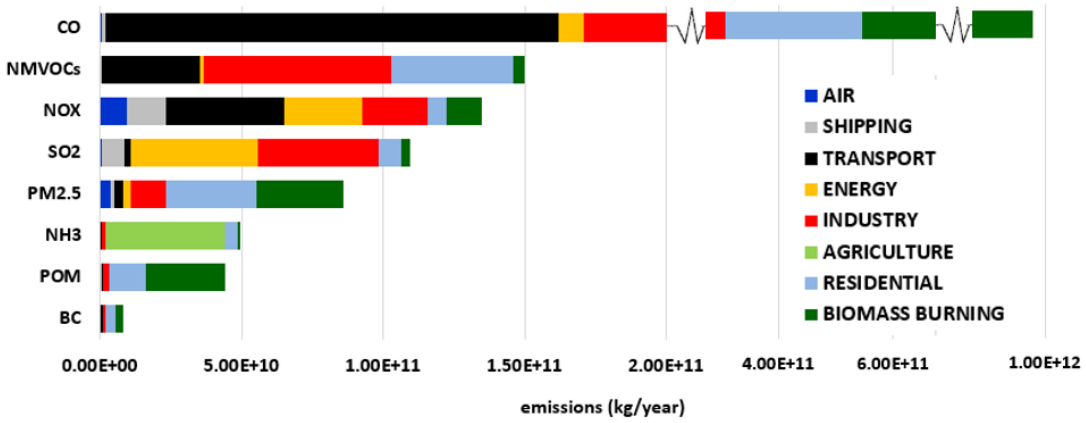
— Industry sector contributes to 44% NMVOCs, 39% SO_2, 17% NO_x, 14% $PM_{2.5}$ and CO, and 12% BC emissions.

— Agriculture sector mostly contributes to 85% NH_3 emissions.

— Residential activities are responsible of 42% BC, 37% PM2.5, 30% POM (OC), 28% (NMVOCs) and 24% CO emissions.

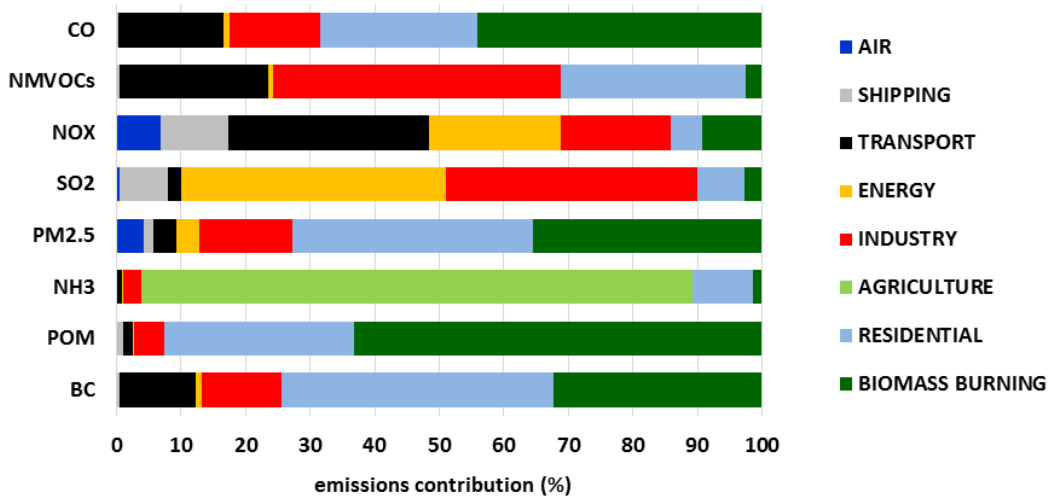
— LS Biomass Burning (labelled as "Biomass Burning" in the legend) contributes to 63% POM, 44% CO, 36% $PM_{2.5}$ and, 32% BC emissions.

Figure 2. Global pollutant emissions for all anthropogenic sectors (kg/Year) (year 2010).



Source: JRC analysis

Figure 3. Sector relative contribution to global emissions by pollutant for the year 2010.



Source: JRC analysis

2.3 Validation with $PM_{2.5}$ source apportionment data

We carried out a comparison of the TM5-FASST modelled concentrations with source apportionment data estimated from measured $PM_{2.5}$ mass concentrations.

Source apportionment (SA) allows the identification of ambient air "pollution sources" and the quantification of their contribution to pollution levels (Belis et al., 2014). This can be achieved 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-oriented models (RMs) are 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:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \quad (8)$$

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 typically used to apportion Particulate Matter (PM) concentration. To that end, a dataset with a rather large amount of data consisting of chemical constituents gathered from a number of observations (samples) is required. The most commonly used chemical species are: major ions (sulphate, nitrate and ammonium), carbonaceous fractions (OC and EC), trace elements and organic markers (e.g. levoglucosan).

RMs account for the measurement uncertainty and provide uncertainty estimation of the model output using uncertainty propagation or randomization methods (e.g. bootstrap).

RMs have been traditionally classified into those which explicitly use information about the emission fingerprints (chemical mass balance) and those which do not use any a priori information on source chemical profiles (factor analysis methods).

The most commonly used RMs are Chemical Mass Balance (CMB) (Watson et al., 1997), and Positive Matrix Factorization (PMF). The original PMF most used versions are PMF 2 and ME-2 (Paatero, 1997, 1999).The EPA free versions are: CMB 8.2, PMF 3 and PMF 5. Other less popular tools are based on eigenvalue methods such as PCA and UNMIX. The availability of source profiles repositories is essential for performing and validating the

RM output. At present two public databases are available: the US-EPA SPECIATE and the JRC SPECIEUROPE (Pernigotti et al., 2016).

In this study we compare the results of a quality-controlled set of urban PM2.5 source-apportionment studies with the modelled sector attribution obtained with TM5-FASST. The source-apportionment dataset consists of 419 records from studies conducted in cities of 51 countries (Karagulian et al., 2015). The records in the source apportionment dataset are not fully homogeneous because they were obtained in different time windows using a variety of RMs. In addition, the number of records varies between different countries/regions.

3 Results

3.1 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. Figure 4 shows global maps of $PM_{2.5}$ concentration attributed to each of the contributing sectors (Transport, Energy, Industry, Agriculture, Residential activities and Large Scale Biomass Burning), and their relative contribution to total $PM_{2.5}$. Relative contributions to total $PM_{2.5}$ are intended to highlight the importance of a specific contributing sector in certain geographical regions and where most of the population lives. The high contribution from agriculture relative to the residential sector and transport $PM_{2.5}$ in Northern Europe is noteworthy. Also the residential sector and transport are standing out as major contributing sectors in Europe. In Asia the residential sector, industry and energy production are major contributing sources, whereas sub-equatorial Africa and South-America are strongly impacted by large-scale forest fires. Note that also at high northern hemispheric latitudes forest fires (Canada, Siberia) are important sources of PM2.5, due to the lack of significant other (anthropogenic) sources. Total $PM_{2.5}$ remains however very low in these areas.

Further population-weighted averaging has been carried out for 23 larger world regions (Figure 5 and

Figure **6**). Raw data by country are reported in Table T5 in the Annex A2. Total anthropogenic $PM_{2.5}$ concentration shows the highest country-average concentration in the China (55 µg/m³) and India (51 µg/m³) region, followed by Western Africa (28 µg/m³) and the Korea region (24 µg/m³). The lowest total $PM_{2.5}$ concentration values was found in Oceania (3 µg/m³), followed by Central America and Mexico with values of about 9 µg/m³.

Making a regional break-down by sector, we find that the highest $PM_{2.5}$ concentrations attributable to transport are observed in India (5 µg/m³) and China (3.5 µg/m³) followed by Korea (3.2 µg/m³) and Western Europe (2.9 µg/m³). 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 (13%) and Middle East (12%). We must keep in mind that for large countries like China and India, country-average concentrations and fractions are masking large spatial heterogeneities.

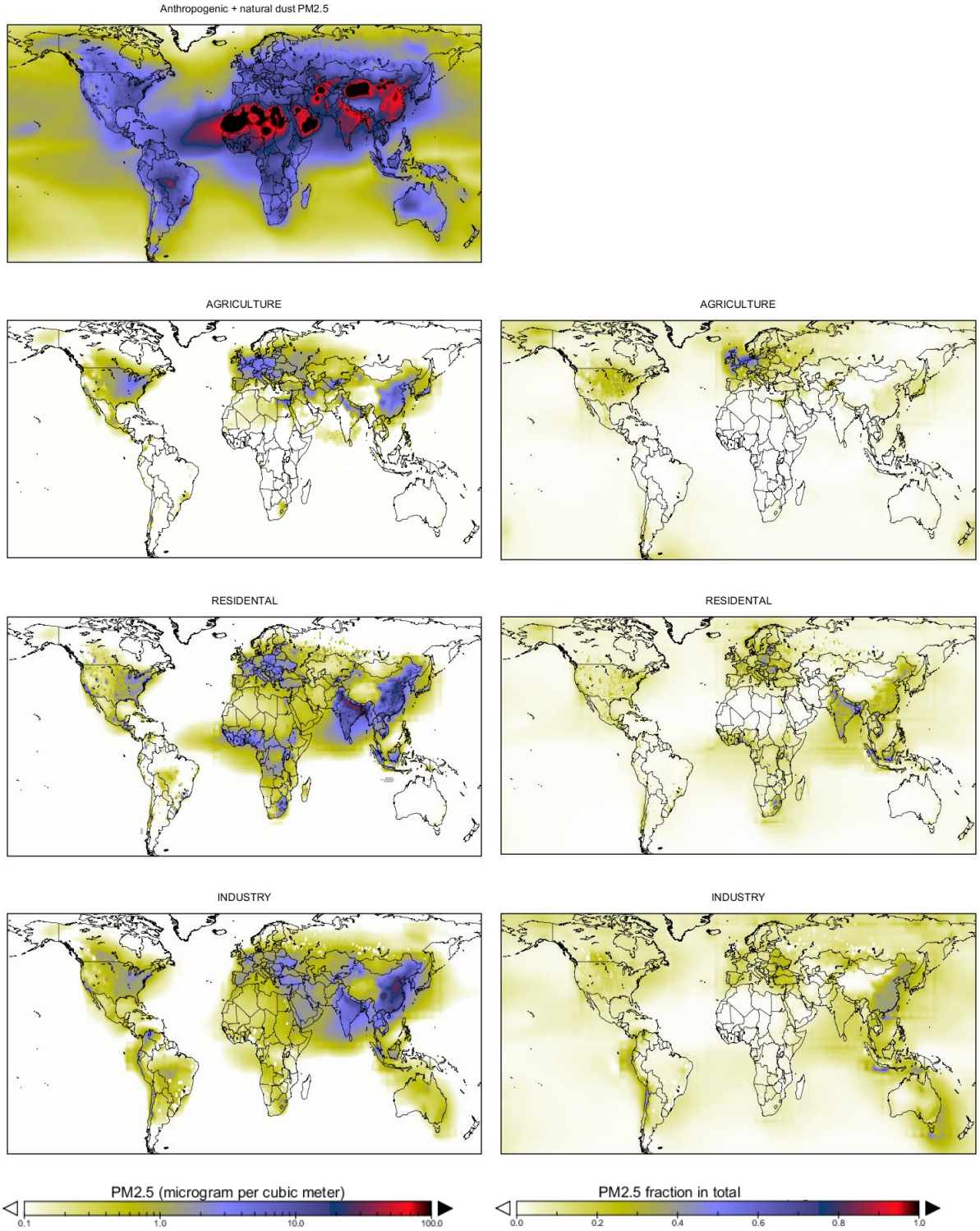
The $PM_{2.5}$ concentrations attributable to Industry are highest in China (21 µg/m³), India and Korea (9 µg/m³) followed by the Ukraine and South Eastern Asia (4 µg/m³). By comparison the relative contribution of Industry to Total $PM_{2.5}$ shows high values spots in China (38%), Korea (37%), Oceania (32%) followed by Ukraine, Kazakhstan and Turkey (25%).

The highest concentrations of $PM_{2.5}$ attributable to the Energy sector are observed in India (10.5 µg/m³) and China (8 µg/m³) followed by Middle East (5.4 µg/m³) and Turkey (4.5 µg/m³). The relative contribution to Total $PM_{2.5}$ from Energy is high in the Middle East (39%), Turkey (28%), Northern Africa (27%) and Mexico (26%)

Agriculture has highest $PM_{2.5}$ values in China (6.2 µg/m³) and in Western Europe (5.2 µg/m³) followed by Central Europe (4.8 µg/m³). The relative contribution to Total $PM_{2.5}$ from agriculture is highest in Kazakhstan (36%), Western Europe (33%), Northern Africa (32%) and Japan (28%).

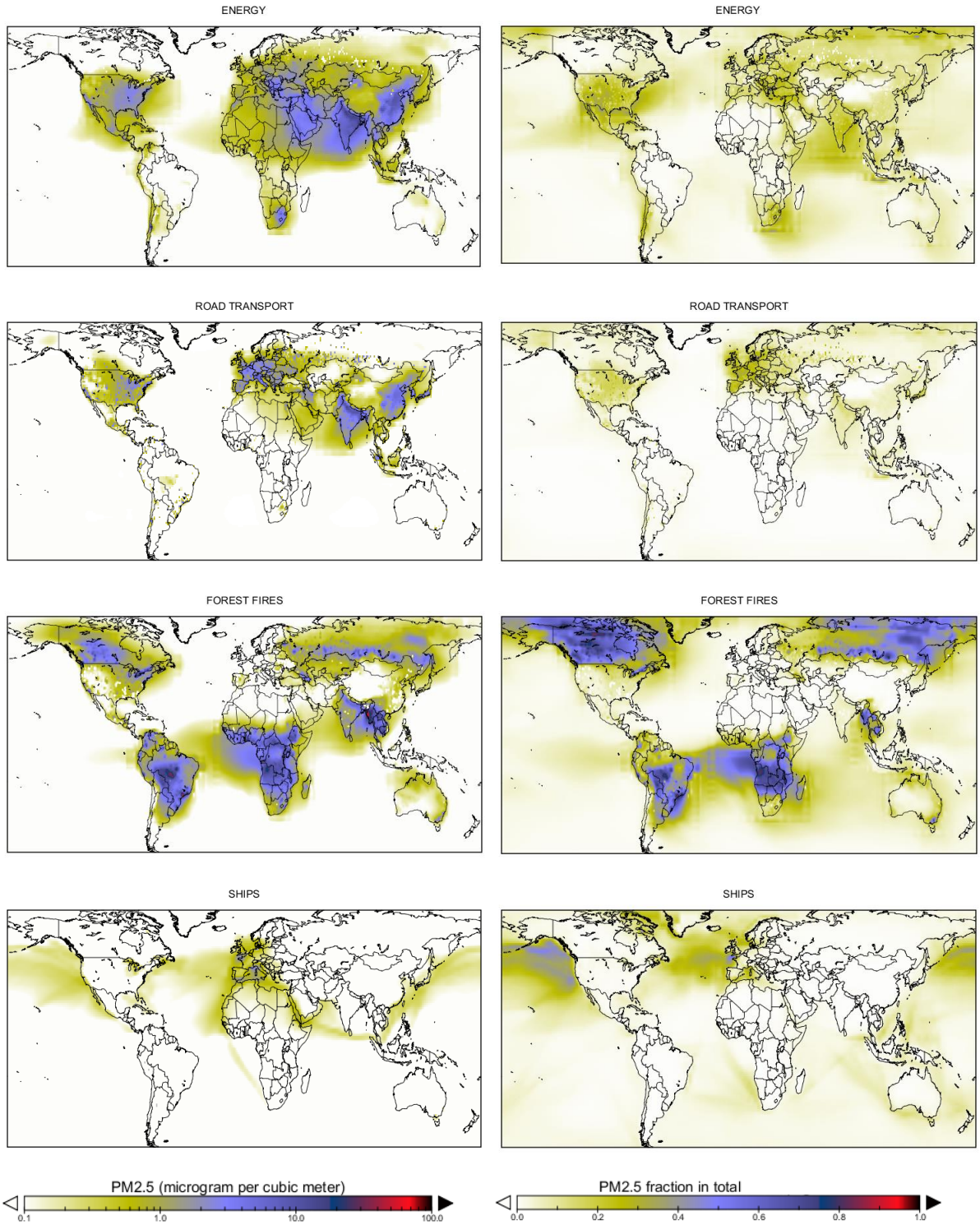
Residential activities, such as heating and cooking, show the highest $PM_{2.5}$ concentrations in India (21.3 µg/m³), and China (14.7 µg/m³) followed by Western Africa (11.4 µg/m³). By comparison the relative contribution to Total $PM_{2.5}$ from the residential sector is highest in India region (42%), followed by Western Africa (41%), Central America (38%) and Eastern Africa (35%).

Figure 4. Left page: Year 2010 total $PM_{2.5}$ (upper left panel, anthropogenic plus mineral dust) and contribution of individual sectors to $PM_{2.5}$ concentrations (µg/m³). Right page: relative contribution (fraction) of sector-derived $PM_{2.5}$ to total $PM_{2.5}$ (anthropogenic plus mineral dust). Grid maps have been linearly interpolated from native 1°x1° resolution to 0.125°x0.125°.



Source: JRC analysis

Figure 4 (continued)



Source: JRC analysis

Large scale Biomass Burning is strongly related to seasonal fires. High $PM_{2.5}$ concentrations are observed in Western Africa and Brazil (6 µg/m³), followed by South East Asia (4.8 µg/m³) and Southern Africa (4.6 µg/m³). 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%).

3.2 Comparison of modelled $PM_{2.5}$ concentrations with measured data

The above findings provide 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 (Brauer et al., 2012; Burnett et al., 2014b). 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 many regions (WHO, 2014).

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 have 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, residential activities, re-suspended dust, sea salt and other unspecified pollution sources of human origin. In the following we review outcomes of in-situ source-apportionment and evaluate the consistency between these results and the ones obtained from the global sector attribution study presented above.

$PM_{2.5}$ concentration by pollution sector obtained the TM5-FASST runs were compared with the $PM_{2.5}$ concentration data reported in the WHO dataset released in 2014 (WHO, 2014). 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.

However, when comparing models and measurements data we might consider that:

— 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 parameterised in the TM5-FASST with a global pseudo-source strength of about 19 Tg. Therefore, modelled $PM_{2.5}$ concentrations could be underestimated in some regions when compared with real ambient $PM_{2.5}$ measurements.

— WHO $PM_{2.5}$ concentrations are from monitoring stations located at different types of sampling sites including traffic, urban background and residential and are biased towards high-pollution locations.

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 7, Figure 8). Both measured (WHO) and modelled (TM5-FASST) $PM_{2.5}$ concentrations show values up to 20-25 µg/m³ in the USA and Western Europe. Instead, values up to 80 µg/m³ can be observed in the India and China regions from both measurement and modelled $PM_{2.5}$ concentrations.

Figure 5. Population weighted annual anthropogenic $PM_{2.5}$ average concentrations by economic sector (2010)

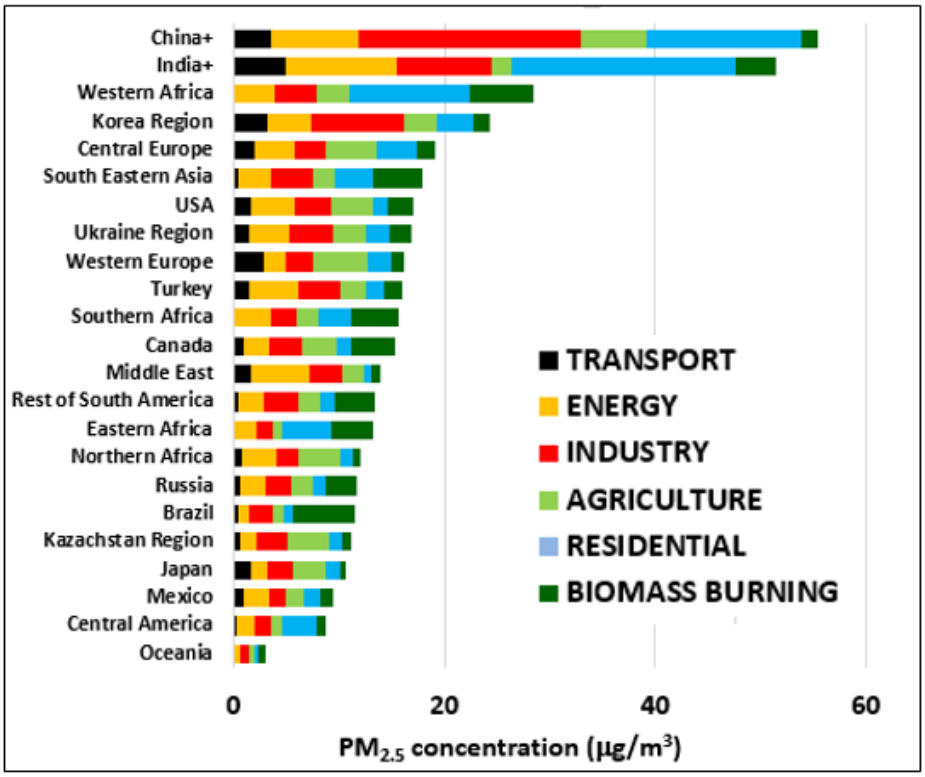
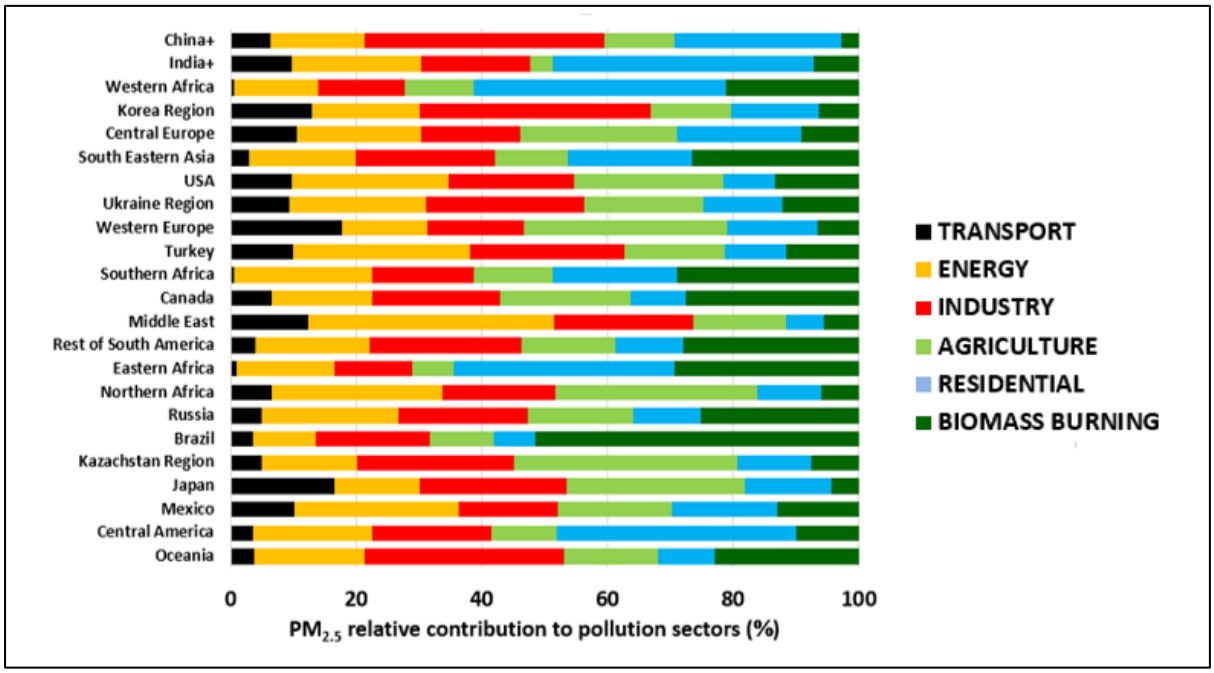
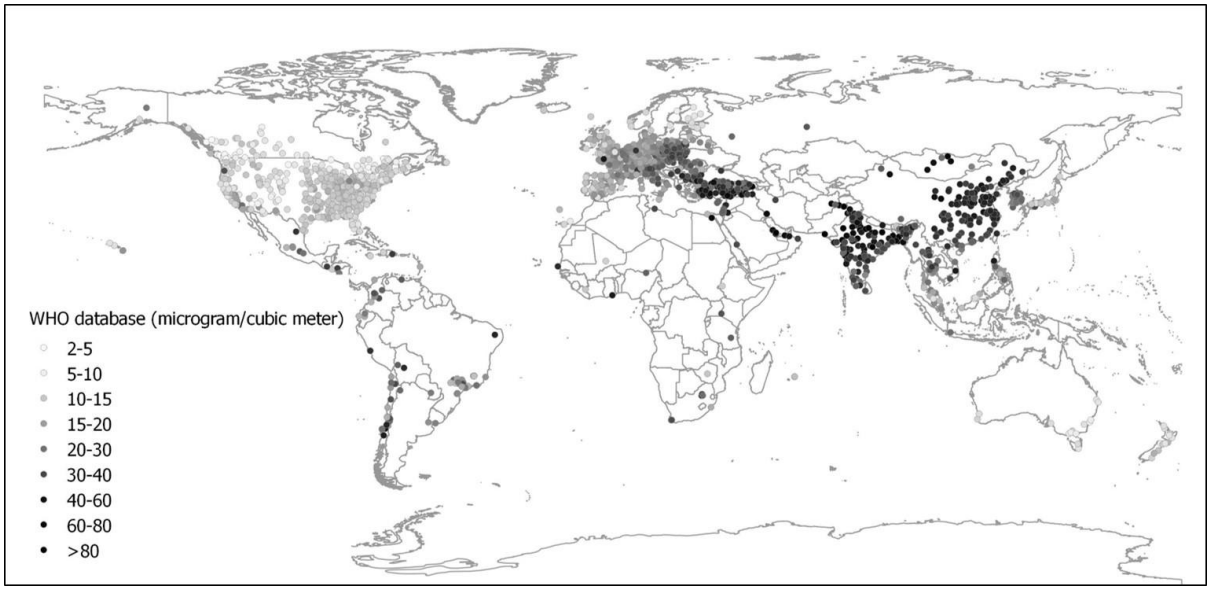


Figure 6. Relative contribution of economic sectors to annual total $PM_{2.5}$ (2010)



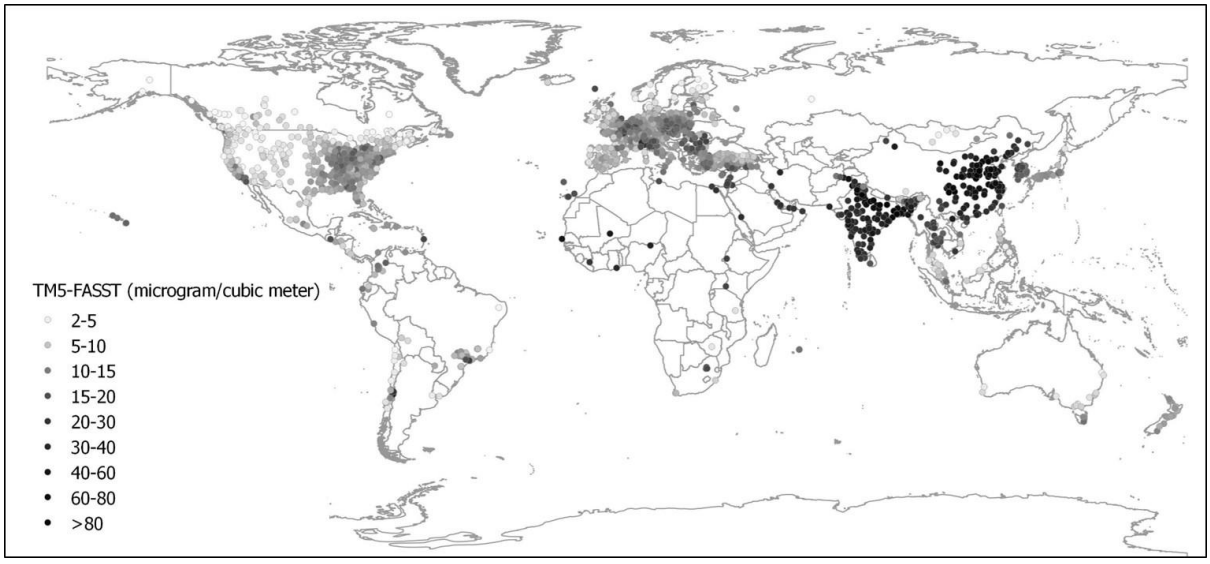
Source: JRC analysis

Figure 7. Total $PM_{2.5}$ concentrations from the WHO Ambient Air Pollution (AAP) dataset updated at the year 2014



Source: JRC analysis

Figure 8. Total $PM_{2.5}$ concentrations extracted from the TM5-FASST interpolated high-resolution grid-map of Total $PM_{2.5}$ concentration averaged over 0.125° x 0.125° grid-cell (year 2010)



Source: JRC analysis

Global average $PM_{2.5}$ concentrations are about 21.36 µg/m³ and 20.56 µg/m³ for WHO and TM5-FASST, respectively. Instead, the global maximum $PM_{2.5}$ concentration is 153 µg/m³ and 142 µg/m³ 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.

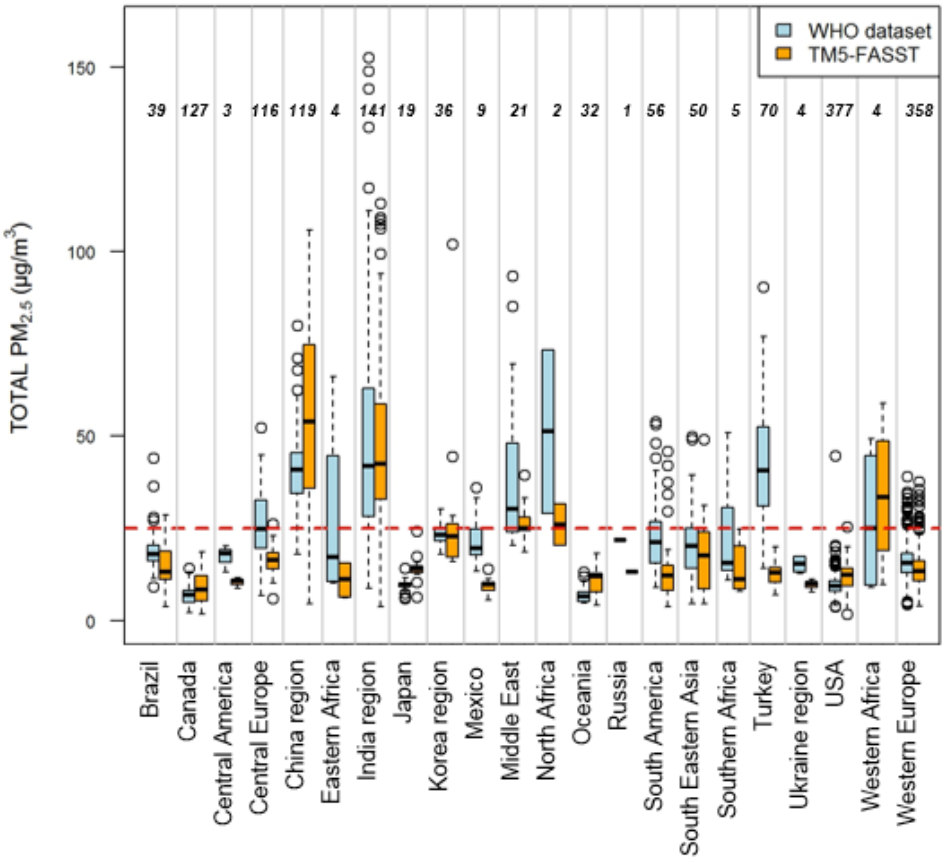
Quantitative comparison of the statistical distribution between the WHO and TM5-FASST Total $PM_{2.5}$ concentrations has been carried out for 22 larger regions. As shown in Figure

9, for regions such as Western Europe and USA, the median value of $PM_{2.5}$ concentrations from WHO and TM5-FASST is quite close. There are several values classified as outliers by the statistical tool used for this analysis (Cai, 2013). 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.

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 9).

Figure 9. Comparison between the WHO Ambient Air Pollution (AAP) dataset (2014) representative for 2008-2013 and TM5-FASST data for Total PM2.5. Boxplots represent the $PM_{2.5}$ concentrations 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 $PM_{2.5}$.



Source: JRC analysis

Regional population-weighted as well as area averaged Total $PM_{2.5}$ have been calculated for WHO and TM5-FASST, in the latter case extracting the grid cell values corresponding to the WHO database locations (Figure 10, Figure 11). Averages have been calculated using the population and the surface area of the cities reported in the WHO database. Globally, the resulting population-weighted average urban $PM_{2.5}$ concentration from TM5-

FASST is about 17% lower than the$PM_{2.5}$ concentrations from WHO data, whereas the area-weighted $PM_{2.5}$ concentration from TM5-FASST is about 24% lower than the average WHO $PM_{2.5}$ concentration from the WHO data.

The major issue arising comparing output of the TM5-FASST model and point measurement data, is the representativeness of the point measurements for the regional scale. As shown in Figure 10 and Figure 11, 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. WHO data from Western Europe and USA, composed of a large number of individual data, are deemed to better represent the "regional" 1°x1° resolution of TM5-FASST. On the other hand, for regions with smaller datasets such a China and India, the few point measurements obtained in urban environments cannot be representative of a larger region and therefore, they cannot be compared with the output of a coarse resolution model such as the TM5-FASST even with a sub-grid parameterisation.

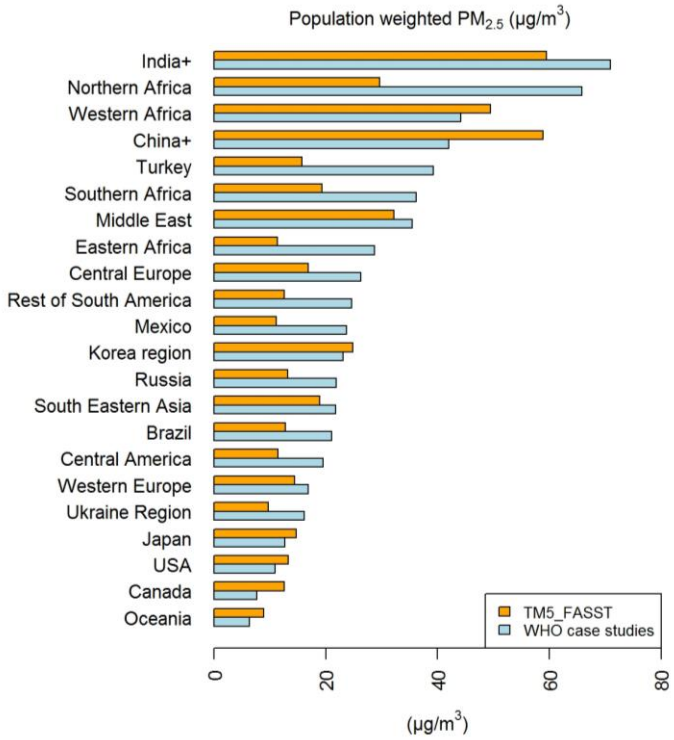
The non-representativeness of point data for the regional concentration is not the only possible explanation for the discrepancy between model and measurements: TM5-FASST is a simplified numerical representation of the complex meteorological and chemical processes taking place between emission and regional transport of pollutants, it is based on a single meteorological year, its output depends critically on the validity of input emissions. Possible contributions to the uncertainty are

- Emission inventories from (HTAP) that can have incomplete coverage in some regions
- Anthropogenic SOAs are not modelled in the TM5-FASST model but it is part of the measured $PM_{2.5}$ concentrations
- Meteorological data in the TM5-FASST model are referred to 2001 while measurements relate to various years
- Linearization of the TM5-FASST Source Receptor Model and the use of emission-concentration sensitivities based on year 2000 emissions in the source-receptor matrices.
- Uncertainties in the parent models' parameterization of atmospheric processes like transport, chemistry and removal.

Apart from 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. $PM_{2.5}$ Several sources of uncertainties may affect the correct estimation of measured total $PM_{2.5}$:

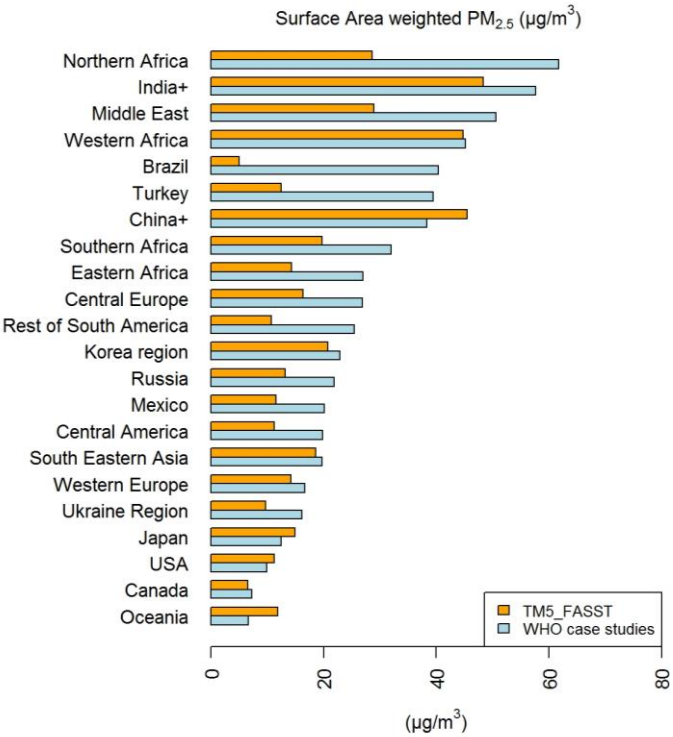
- Experimental errors (field and laboratory activity) and sampling method (usual uncertainty for sampling $PM_{2.5}$ is about 2.0 μg/m³ for both TEOM-FDMS and gravimetric method)
- Temporal coverage of the measurements (season) and yearly emission data used in the TM5-FASST model
- Quality Control/Quality Assurance of the measurement protocols
- Representativeness of measurement location for larger area.

Figure 10. 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 PM2.5. (year 2010).



Source: JRC analysis

Figure 11.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 PM2.5. (year 2010)



Source: JRC analysis

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

In the past decade, tools have been developed for improving capabilities of source resolution and source contribution quantification (Karagulian and Belis, 2012). "Pollution sources" identified and quantified by receptor models can be somehow compared to "pollution sectors" defined in the emission inventories.

Recent work (Belis et al., 2013) 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 peer-reviewed publications have been used to screen and classify worldwide data on PM_{10}, $PM_{2.5,}$ and, PM_1 up to 2014. These studies are based on urban, sub-urban, remote, rural, and industrial site locations

Here, SA data for $PM_{2.5}$ mass concentrations are compared with TM5-FASST $PM_{2.5}$ modelled concentrations.

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

- Sea Salt (including road salting)
- Secondary Inorganic Aerosols (SO4, NO3, NH4)
- Crustal/Re-Suspended Dust
- Traffic (only ground transportation including road dust)
- Industry-Oil Combustion (include point sources and power plants for energy production)
- Biomass Combustion (small-scale use of biofuel and biomass waste burning such as, wood and wood waste burning)
- Residential and small-scale non-industrial combustion
- Secondary Organic Aerosol (usually estimated from PM10)
- Unexplained/other sources

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

Sources from SA studies such as Traffic, Residential Heating (including biofuel) and, partially Industry-Oil Combustion can be compared with the emission sectors (used in TM5-FASST) of Transport, Residential, Industry and Energy, respectively. On the other hand, identification of the "agricultural sector" as a pollution source in SA studies is more problematic because of the secondary nature of the resulting $PM_{2.5}$ (involving longer time scales) and multiple sources contributing to the resulting component (mostly ammonium nitrate, with ammonium resulting from agriculture and nitrate from other sources, see Figure 3).

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

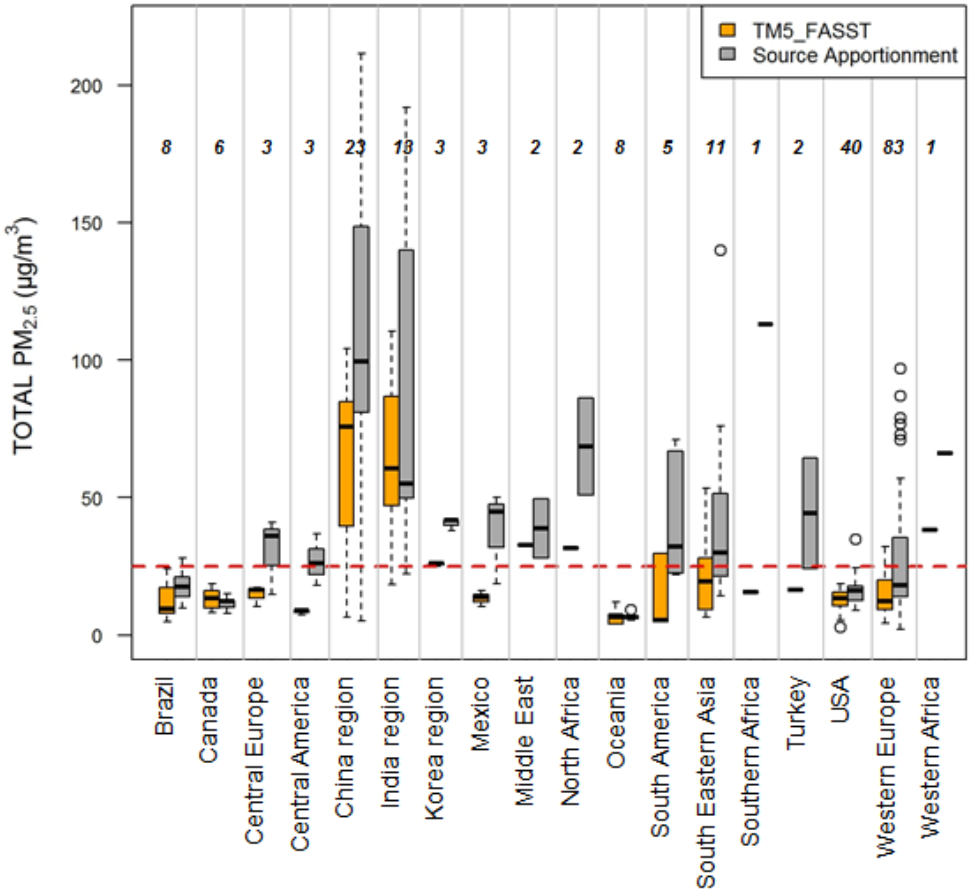
— 150 case studies for the Industry pollution factor/source

The studies mentioned above provide data both on Total PM2.5, as well as the source attribution. We will compare both metrics with our TM5-FASST model results for year 2010.

3.3.1 Total $PM_{2.5}$: TM5-FASST vs SA case studies

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

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



Source: JRC analysis

As we can see, TM5-FASST data values fall in the same range of SA data for India, USA, Western Europe, Canada and Oceania and with the exception of China and India, median values are also below 25 µg/m³, the EU limit value for Total $PM_{2.5}$.

As for the WHO data, population-weighted average and area average of Total $PM_{2.5}$ have been calculated for SA and TM5-FASST data. Averages have been calculated using the population and the surface area of the cities used in the SA database. Regional averages are shown in Figure 13 and Figure 14. Globally, population-weighted Total $PM_{2.5}$ concentrations from TM5-FASST are about 30% lower than $PM_{2.5}$ concentrations from SA data, whereas area-weighted Total $PM_{2.5}$ concentrations from TM5-FASST are about 33% lower than SA $PM_{2.5}$ measured concentrations.

As discussed previously, the major issue 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 analysed for this work cannot necessarily be considered a representative subset.

3.3.2 $PM_{2.5}$ from Transport: TM5-FASST vs SA case studies

212 case studies have been selected to compare Transport $PM_{2.5}$ concentrations from SA and TM5-FASST model estimations. Figure 15 shows a statistical analysis for SA and TM5-FASST Transport $PM_{2.5}$ concentrations.

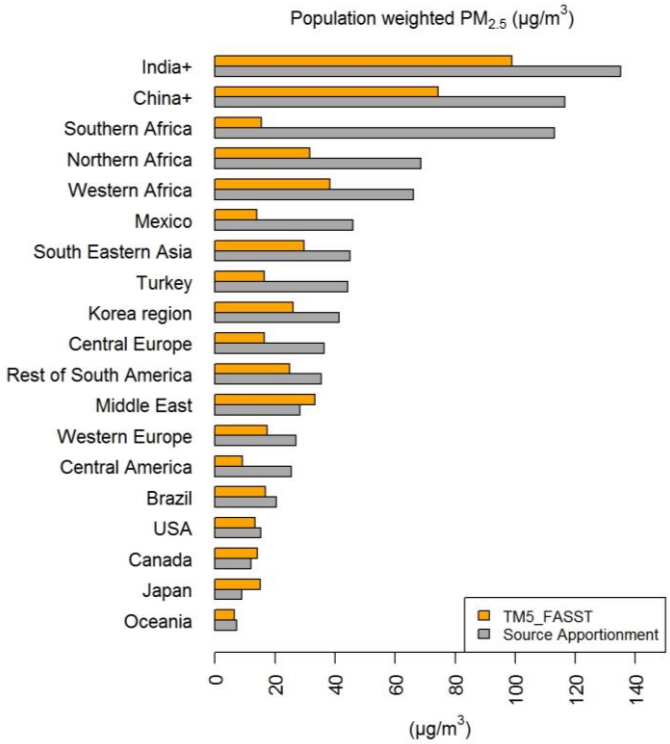
Population-weighted average and area average of Transport $PM_{2.5}$ have been calculated for the SA and TM5-FASST data. Regional averages are shown in Figure 16 and Figure 17. Regional averaged concentration values are given in Table T3 of Annex 2. Large discrepancies are observed between the SA and TM5-FASST modelled datasets: globally, population-weighted Transport $PM_{2.5}$ concentrations from TM5-FASST are about 70% lower than $PM_{2.5}$ concentrations from SA data whereas area-weighted weighted Transport $PM_{2.5}$ concentration from TM5-FASST is about 77% lower than SA $PM_{2.5}$ measured concentration.

Large discrepancies between SA and TM5-FASST Transport $PM_{2.5}$ concentrations might be tentatively attributed 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 from Transport emission are not modelled in the TM5-FASST model.
- Bias in transport sector emission data used as input to the TM5-FASST model.

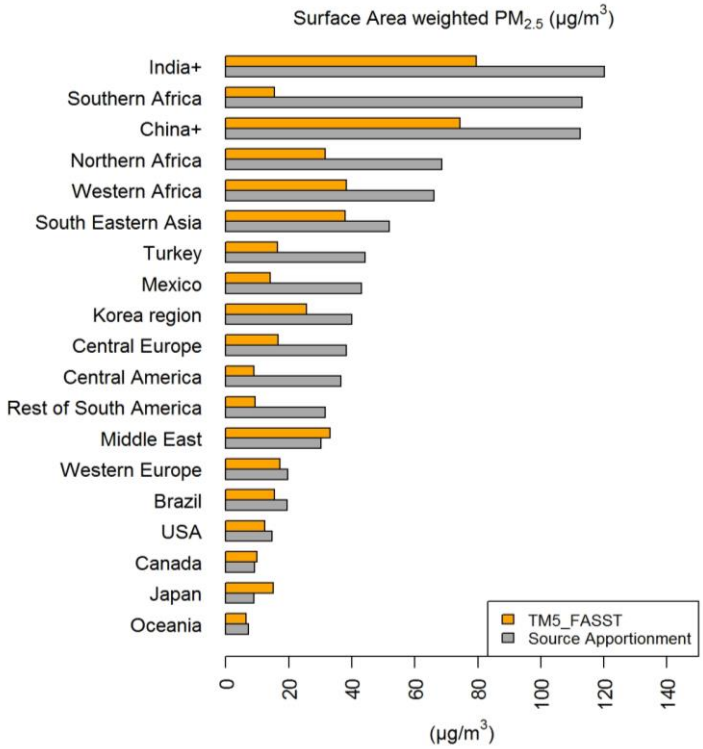
Further, in SA case studies, transport-induced re-suspended road dust is in principle mixed with the Transport source, whereas in TM5-FASST only long-range transport of dust from large desert regions is included as a separate source, but not a potential re-suspension of local road-dust. Recent SA works carried out in six major cities in India, estimate that 40-60% of PM_{10} can be attributed to traffic-related re-suspended dust. A similar situation is observed for $PM_{2.5}$ (CPCB, 2014).

Figure 13. 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).



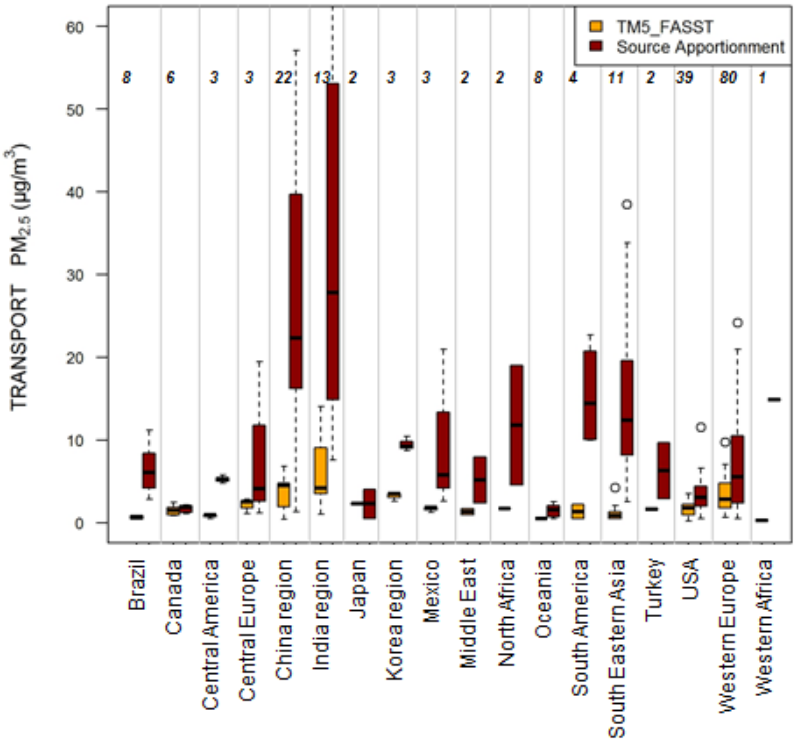
Source: JRC analysis

Figure 14.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).



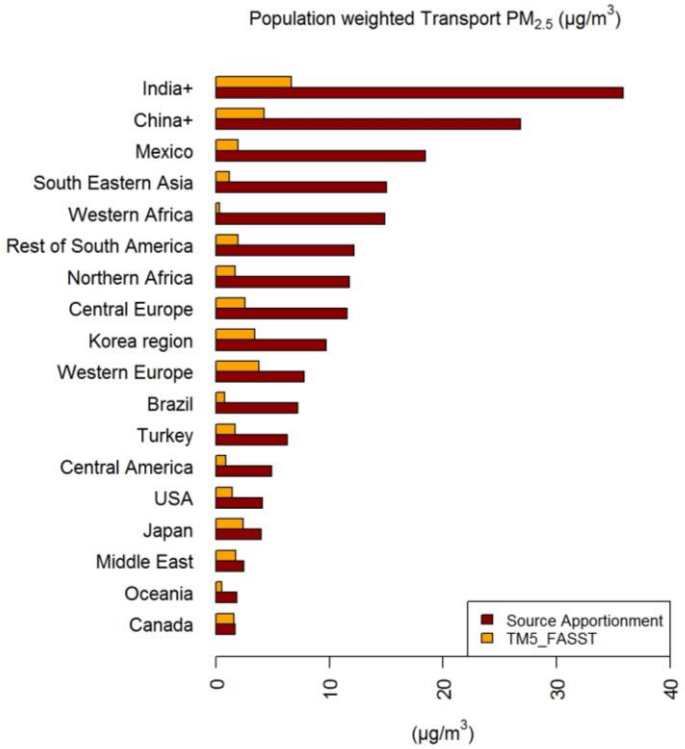
Source: JRC analysis

Figure 15. Comparison between TM5-FASST and Source Apportionment data for Transport $PM_{2.5}$ concentrations. Boxplots represent the $PM_{2.5}$ concentrations 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.



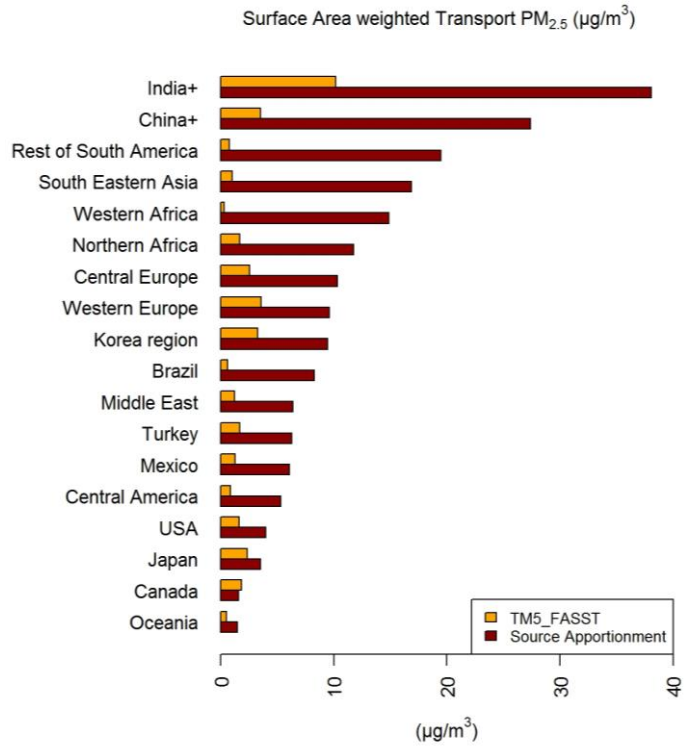
Source: JRC analysis

Figure 16. 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).



Source: JRC analysis

Figure 17. 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).



Source: JRC analysis

3.3.3 $PM_{2.5}$ from Industry: TM5-FASST vs SA case studies

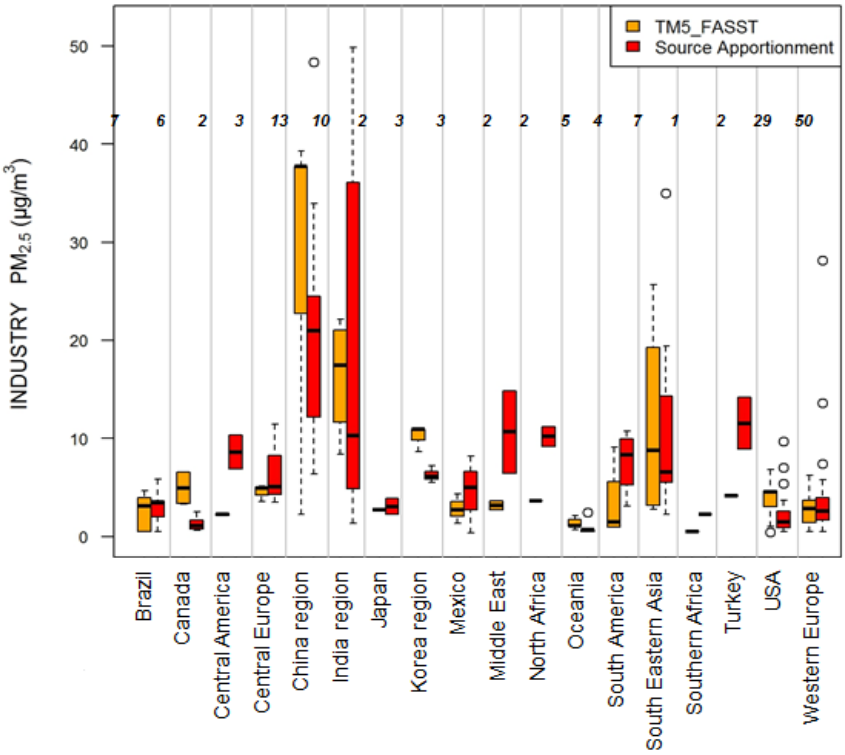
150 case studies have been selected to compare Industry $PM_{2.5}$ concentrations from SA and TM5-FASST model estimations. Figure 18 shows the comparison between Industry $PM_{2.5}$ from TM5-FASST model and SA studies. Low $PM_{2.5}$ concentrations are observed in Western Europe and USA. Instead, higher concentrations are found in the India and China. 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 19 and Figure 20.

There are 2 effects which could cause the SA estimates to exceed the TM5-FASST attribution of Industry to PM2.5:

1. the definition of the "Industry" source in SA studies is commonly grouped together with Power Plants because of similar combustion processes leading to observed pollution trends, which is somewhat different from the "pure" Industrial sector defined in the HTAP emission inventory.
2. In particular power plants represent strong point sources that are confined in local areas, whereas TM5-FASST model calculations based on an implicit underlying emission gridding at a resolution of 1°x1° (roughly 100kmx100km).

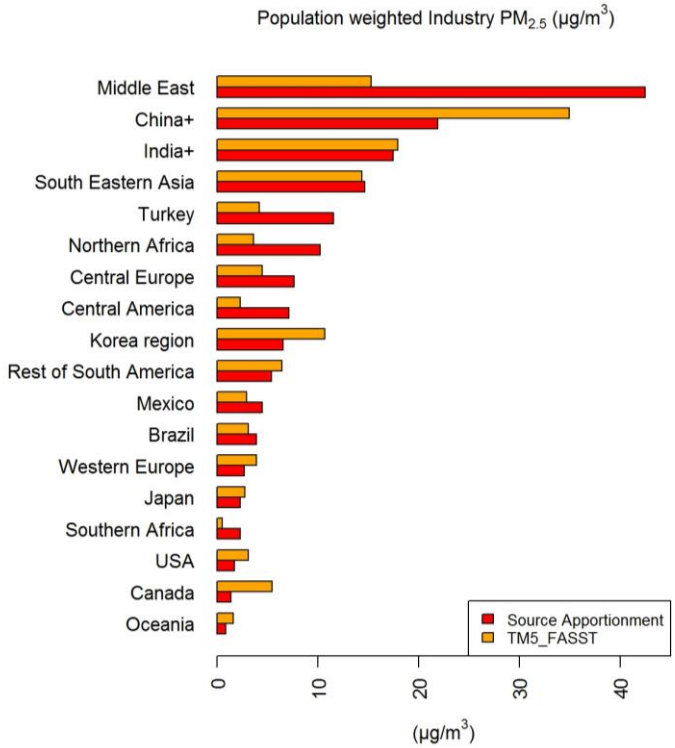
Nonetheless, for this source category, differences are lower than for the Transport/Traffic source: globally, population-weighted Industry $PM_{2.5}$ concentrations from TM5-FASST are about 13% higher than $PM_{2.5}$ concentrations from SA data whereas area-weighted weighted Industry $PM_{2.5}$ concentration from TM5-FASST was about 17% higher than SA $PM_{2.5}$ measured concentrations. Values are given in Table T4, Annex2. Largest discrepancy is found for a single location in Argentina and more systematically in the Eastern Mediterranean.

Figure 18. Comparison between TM5-FASST and Source Apportionment data for Industry $PM_{2.5}$ concentrations. Numbers indicate the amount of data available in each region.



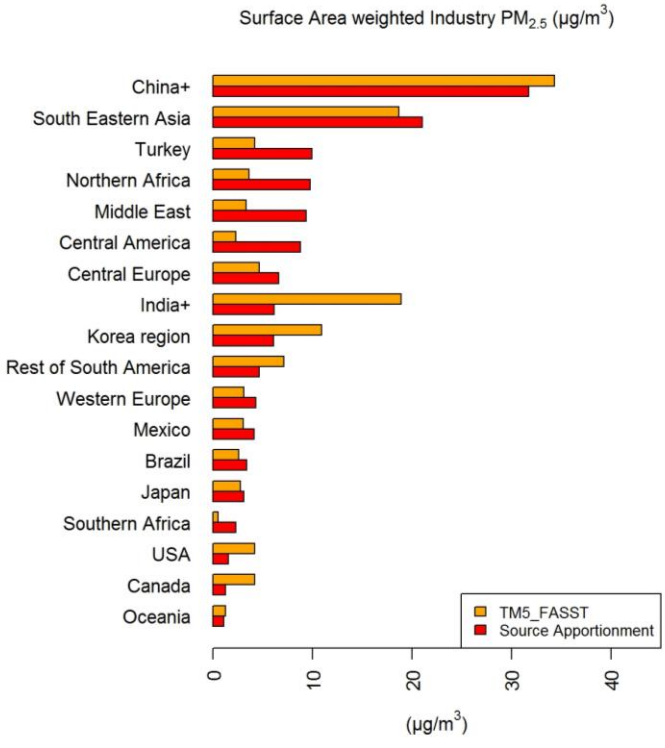
Source: JRC analysis

Figure 19. 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).



Source: JRC

Figure 20. 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).



Source: JRC analysis

4 Conclusion and outlook

This report compares the relative contributions to total $PM_{2.5}$ concentrations from different emission sectors as calculated by the global TM5-FASST model with in-situ source-apportionment data from urban locations over the globe. This is the first work comparing sector attributed modelled $PM_{2.5}$ concentrations with $PM_{2.5}$ point measurements from the WHO database (2014) and Source Apportionment case studies.

The global model results are derived from global emission inventories for the year 2010 (HTAP-v2). Results showed that emerging economies such as China and India have the highest concentration of $PM_{2.5}$ from the Industry and Residential sector. China and India are the countries with the highest $PM_{2.5}$ concentrations with 55 µg/m³ and 55 µg/m³, respectively. Industry is the largest economic sector in China contributing 38% of total $PM_{2.5}$ emissions whereas India has a larger contribution from Residential emissions (42%) followed by Western Africa (40%).

The Transport sector showed largest absolute values in India, China and Korea. However, the biggest relative contribution of Transport to $PM_{2.5}$ has been observed in Western Europe (18%) and Japan (17%). The Energy sector showed highest values for India and China but has its largest relative contribution (38%) to $PM_{2.5}$ in the Middle East.

Agriculture is an important source of $PM_{2.5}$ in the Kazakhstan region (36%) as well as in Northern Africa and Western Europe (32%). Residential activities are important sources of to $PM_{2.5}$ in India (42%), Western Africa (40%) and Central America (38%). Finally, large scale Biomass Burning is the major source of $PM_{2.5}$ in Brazil (51%), Southern Africa (29%) the Rest of South America (28%) and Canada (27%).

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 different spatial representativeness of the urban background dataset when compared with the coarser output of the chemical transport model that underpins TM5-FASST. This highlights the importance of having a comprehensive up-to-date dataset for source-attributed $PM_{2.5}$. More SA data sets are needed from emerging countries where most of the highest pollution levels are recorded and where there are high population densities. In addition, there is an urgent need to harmonize the pollutant source-attribution (referred to economic sectors) in $PM_{2.5}$ formation, between the SA datasets and emission data.

Although emission inventory data used in this study are representative for medium-sized urban agglomerates, the model used to derive PM formation from primary and secondary pollutants is still too coarse to be used to assess air sources of air pollution at local levels. On the other hand, at regional level, the model clearly showed its potential to evaluate the distribution of pollution sources between countries and consequently show substantial differences between different economies.

Source apportionment results represent the best means of identifying local pollution sources of PM using measurement data from monitoring stations around the world. Although globally there is not a homogeneous distribution of data, the potential of this technique will increase as the number of source apportionment studies increases. The inconsistency observed between modelled results and source apportionment data is clearly associated with the different spatial distribution of the pollution source that has been assessed against the modelled one. The two methods (air quality model and source apportionment) find a better agreement in regions with a denser spatial distribution of point measurements (USA and Europe).

Therefore, from this study it is not possible to make a general direct comparison between modelled data and source apportionment data at local level. On the other hand, at regional level, source apportionment studies can be extrapolated to larger areas and can give an estimation of regional pollution.

The spatial distribution of emissions inventories, modelled data and source apportionment data can be definitely enhanced with the integration of additional data such as satellite data and regional – local air quality models with resolutions around 1km x 1km. Statistical integrated methodologies such as the geographically regression modelling (van Donkelaar et al., 2016) might represent a step forward in filling gaps between measurements and modelled data

The present work has identified the need to:

— Improve the spatial resolution (using appropriate downscaling techniques) of the TM5-FASST model to better target emissions at city scales. This includes addressing the issue of re-suspended road dust.

— Focus on case studies dealing with megacities where there is a more accurate knowledge of local emissions and measurements.

— Use Source Apportionment (SA) data to validate the performance and output of the TM5-FASST model.

— Make efforts to understand the causes of the underestimation $PM_{2.5}$ due to Transport in the TM5-FASST model compared to SA case studies.

— Improve the source separation and identification between model and measurements.

— Call for a better cooperation between experimental field research and modellers to fill the gap between SA data and modelled data.

Also from the measurement side, some issues can be identified:

— 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, at best represent 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 needs to be applied to estimate $PM_{2.5}$ levels.

  The SA dataset has been built gathering all available case studies published by 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, several of the SA $PM_{2.5}$ measurements are more representative of urban agglomerates.

Comparison at regional level of Total $PM_{2.5}$ concentrations with the $PM_{2.5}$ dataset from WHO showed several discrepancies. These are mainly attributable to the inability of the TM5-FASST model to target point sources of $PM_{2.5}$. In addition, the WHO dataset cannot be representative of all the regions where the TM5-FASST model generates its $PM_{2.5}$ estimates. Obviously, model results obviously depend on the quality of the emission data used as input

The same issue has been observed with even 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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List of abbreviations and definitions

AAP	Ambient Air Pollution
BC	Black Carbon
EDGAR	Emission Database for Global Atmospheric Research
EMEP	The European Monitoring and Evaluation Programme
GFED3	Global Fire Emissions Database version 3
GIS	Geographic Information System
HTAP	Hemispheric Transport of Air Pollutants
IPCC AR5	Intergovernmental Panel on Climate Change - 5th Assessment Report
LS	Large-Scale
MICS	Model Intercomparison Study
NMVOC	Non-methane volatile organic compounds
OC	Organic Carbon
PAH	Polycyclic aromatic hydrocarbons components
PM	Particulate Matter
$PM_{2.5}$	Fine particulate matter with an aerodynamic diameter smaller than 2.5 µm
PM10	Fine particulate matter with an aerodynamic diameter smaller than 10 µm
POM	Particulate organic matter
REAS	Regional Emission Inventory for Asia
SA	Source Apportionment
SIA	Secondary Inorganic Aerosol
SOA	Secondary Organic Aerosol
SRC	Source-receptor coefficient
TEOM-FDMS	System for continuous monitoring of $PM_{2.5}$ mass based on Tapered Element Oscillating Microbalances with Filter Dynamics Measurement Systems
TM5-CTM	Chemical Transport Model version 5
TM5-FASST	Reduced-Form Fast Scenario Screening Tool based on the chemical transport model TM5-CTM
TNO	Applied Research in Natural Sciences (Toegepast Natuurwetenschappelijk Onderzoek)
VOC	Volatile Organic Compounds
WHO	World Health Organisation

List of Figures

ANNEX 1: FASST Regions definition

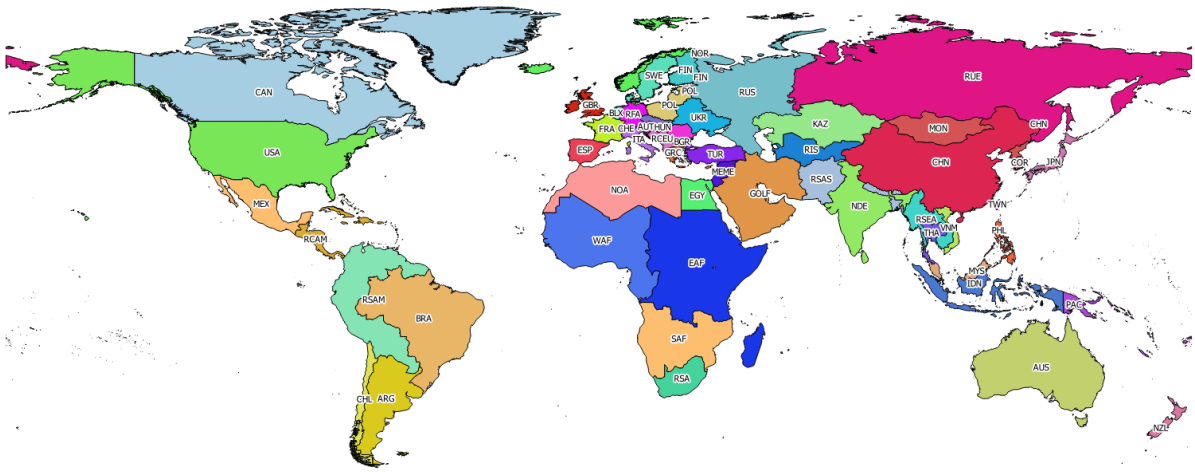

Table A1. List of the FASST codes/names and the correspondent ISO codes of the belonging countries

FASST code	**FASST region name**	**ISO codes of the included countries**
ARG	Argentina, Falklands and Uruguay	ARG FLK URY
AUS	Australia	AUS
AUT	Austria, Slovenia and Liechtenstein	AUT SVN LIE
BGR	Bulgaria	BGR
BLX	Belgium, Luxemburg and Netherlands	BEL LUX NLD
BRA	Brazil	BRA
CAN	Canada and Greenland	CAN GRL
CHE	Switzerland	CHE
CHL	Chile	CHL
CHN	China, Hong Kong and Macao	CHN HKG MAC
COR	South Korea	KOR
EAF	Eastern Africa	CAF TCD SDN ETH SOM KEN UGA COD RWA TZA MDG ERI DJI COM BDI BID MUS REU SYC SDS SOL
EGY	Egypt	EGY
ESP	Spain and Portugal	ESP PRT GIB
FIN	Finland	FIN
FRA	France and Andorra	FRA AND

GBR	Great Britain and Ireland	GBR IRL GGY IMN JEY
GOLF	Gulf states	BHR IRQ KWT OMN QAT SAU ARE YEM IRN
GRC	Greece and Cyprus	GRC CYP
HUN	Hungary	HUN
IDN	Indonesia and East Timor	IDN TLS
ITA	Italy, Malta, San Marino and Monaco	ITA VAT SMR MCO MLT
JPN	Japan	JPN
KAZ	Kazakhstan	KAZ
MEME	Israel, Jordan, Lebanon, Palestine Territories and Syria (Near East)	ISR JOR PSE LBN SYR PSX
MEX	Mexico	MEX
MON	Mongolia and North Korea	MNG PRK
MYS	Malaysia, Singapore and Brunei	MYS SGP BRN
NDE	India, Maldives and Sri Lanka	IND LKA MDV
NOA	Morocco, Tunisia, Libya and Algeria	MAR DZA ESH TUN LBY SAH
NOR	Norway, Iceland and Svalbard	NOR ISL SJM
NZL	New Zealand	NZL
PAC	Pacific Islands and Papua New Guinea	FJI NCL SLB VUT FSM GUM KIR MHL NRU MNP PLW NFK TKL ASM COK PYF NIU PCN TON TUV WLF WSM PNG
PHL	Philippines	PHL
POL	Poland and Baltic states	POL EST LVA LTU
RCAM	Central America and Caribbean	PAN NIC HND GTM SLV ANT KNA LCA VCT TTO TCA VIR BLZ AIA ATG ABW BHS BRB VGB CYM DMA CUB DOM GRD GLP HTI JAM MTQ MSR PRI CRI
RCEU	Serbia, Montenegro, The Former Yugoslavian Republic of Macedonia and Albania (Rest of Central Europe)	SCG MKD HRV BIH ALB SRB MNE
RCZ	Czech Republic and Slovakia	CZE SVK
RFA	Germany	DEU
RIS	Rest of former Soviet Union	KGZ TKM UZB TJK
ROM	Romania	ROU

RSA	Republic of South Africa, Swaziland and Lesotho	ZAF SWZ LSO
RSAM	Rest South America	BOL COL ECU GUF GUY PER SUR VEN PRY PRA
RSAS	Rest of South Asia	AFG BGD BTN NPL PAK
RSEA	Cambodia, Laos and Myanmar	KHM LAO MMR
RUE	Eastern part of Russia	RUE
RUS	Russia, Armenia, Georgia and Azerbaijani	RUS ARM GEO AZE
SAF	Southern Africa (ex RSA)	AGO NAM ZMB BWA ZWE MOZ MWI MYT
SWE	Sweden and Denmark	SWE DNK FRO
THA	Thailand	THA
TUR	Turkey	TUR
TWN	Taiwan	TWN
UKR	Ukraine, Belarus and Moldova	BLR MDA UKR
USA	United States	USA SPM BMU
VNM	Vietnam	VNM
WAF	West Africa	COG CNQ GAB GIN CMR NGA NER MLI BEN GHA BFA CIV SEN GMB GNB SLE LBR STP CPV SHN TGO GNQ MRT

Source: JRC analysis

ANNEX 2: FASST and SA results by region

Table T1. Data values for population and area weighted $PM_{2.5}$ concentration regional 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)

Country region	**$PM_{2.5}$ WHO (Area-weighted**	**$PM_{2.5}$ TM5-FASST (Area-weighted)**	**$PM_{2.5}$ WHO (Pop. weighted)**	**$PM_{2.5}$ TM5-FASST (Pop. 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

Source: JRC analysis

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

Country Region	**$PM_{2.5}$ SA (Area-weighted)**	**$PM_{2.5}$ TM5-FASST (Area-weighted)**	**$PM_{2.5}$ SA (Pop. weighted)**	**$PM_{2.5}$ TM5-FASST (Pop. 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

Source: JRC analysis

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).

Country Region	**Transport $PM_{2.5}$ SA (Area-weighted)**	**Transport $PM_{2.5}$ TM5-FASST (Area-weighted)**	**Transport $PM_{2.5}$ SA (Pop. weighted)**	**Transport $PM_{2.5}$ TM5-FASST (Pop. 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

Source: JRC analysis

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

Country Region	Industry $PM_{2.5}$ SA (Area-weighted)	Industry $PM_{2.5}$ TM5-FASST (Area-weighted)	Industry $PM_{2.5}$ SA (Pop. weighted)	Industry $PM_{2.5}$ TM5-FASST (Pop. 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

Source: JRC analysis

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KJ-NA-28510-EN-N

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Publications Office

doi:10.2760/344371
ISBN 978-92-79-66599-8