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Review article

Contributions to cities' ambient particulate matter (PM): A systematic review of local source contributions at global level

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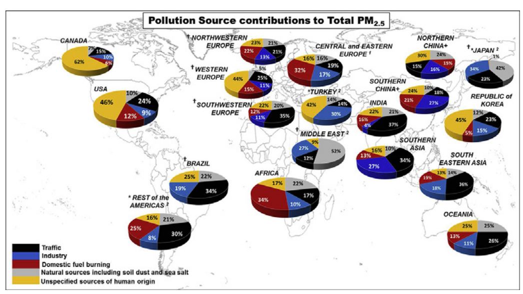
HIGHLIGHTS

- Typical shares of the ambient sources of $PM_{2.5}$ by country and by region were estimated.
- Traffic has been targeted as important contributor to ambient air pollution in cities.
- A database for Source Apportionment studies as of August 2014 has been compiled.

GRAPHICAL ABSTRACT



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ABSTRACT

For reducing health impacts from air pollution, it is important to know the sources contributing to human exposure. This study systematically reviewed and analysed available source apportionment studies on particulate matter (of diameter of 10 and 2.5 microns, PM_{10} and $PM_{2.5}$) performed in cities to estimate typical shares of the sources of pollution by country and by region. A database with city source apportionment records, estimated with the use of receptor models, was also developed and available at the website of the World Health Organization.

Systematic Scopus and Google searches were performed to retrieve city studies of source apportionment for particulate matter. Six source categories were defined. Country and regional averages of source apportionment were estimated based on city population weighting.

A total of 419 source apportionment records from studies conducted in cities of 51 countries were used to calculate regional averages of sources of ambient particulate matter. Based on the available information, globally 25% of urban ambient air pollution from $PM_{2.5}$ is contributed by traffic, 15% by industrial activities, 20% by domestic fuel burning, 22% from unspecified sources of human origin, and 18% from natural dust and salt. The available source apportionment records exhibit, however, important heterogeneities in assessed source categories and incompleteness in certain countries/regions.

Traffic is one important contributor to ambient PM in cities. To reduce air pollution in cities and the substantial disease burden it causes, solutions to sustainably reduce ambient PM from traffic, industrial activities and biomass burning should urgently be sought. However, further efforts are required to

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improve data availability and evaluation, and possibly to combine with other types of information in view of increasing usefulness for policy making.

1. Introduction

New evidence on exposure-risk information and improved global exposure estimates (Brauer et al., 2012; Burnett et al., 2014) suggest higher exposure to ambient particulate matter (PM) than previously estimated (WHO, 2014a). An important fraction of the exposure leading to those health impacts occurs in cities, due to the higher density of human activities and their emissions to the air. According to the region, however, high emissions can also occur from domestic fuel use in rural or peri-urban areas, particles associated with long range transport, or high occurrence of natural dusts. Due to growing population numbers, increasing urbanization, and economic growth, those health impacts may even get worse in much of the world if no adequate action is taken. Initial analyses are confirming this trend (WHO, 2014a, 2014b). PM is currently considered to be the best indicator for health effects of ambient air pollution (Burnett et al., 2014; WHO, 2014a). Many human activities contributing to ambient PM are further contributing to climate change, and are therefore associated with additional health impacts (Karagulian et al., 2014). In order to take actions to reduce exposure to air pollution, and hence the associated health impacts, 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, biomass burning/residential activities, re-suspended dust, sea salt and other unspecified pollution sources of human origin.

This study aims at a systematic review of local case studies of sources of ambient particulate matter (PM) around the world and synthesizing this information to estimate the main contributors of ambient PM in cities in different world regions.

2. Methods

2.1. Study design

We estimated the average contributions to urban ambient PM from key emission source categories for world regions by averaging available results of local source apportionment (SA) studies. For the purpose of this study, countries have been grouped into 19 country-regions according to the IAM (Integrated Assessment Modelling) (Parson and Fisher-Vanden, 1995) and United Nation geoscheme (UN, 2013) (Table A.1, Supplementary material). These regions are based on similar socio–economic parameters for countries belonging to the same region. Our analysis comprised three steps: (1) identifying data sources, (2) defining common source categories and extracting data, and (3) estimating regional averages of source contributions.

2.2. Data sources

Source apportionment (SA) studies are local studies determining the contributing sources to ambient PM measured at representative monitoring stations. Chemical compounds analyzed in the measured particulate matter define some "chemical fingerprints" which hint the attribution of the particulate matter to sectors as sources of ambient PM (Belis et al., 2014).

Source apportionment of ambient PM 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:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \tag{1}$$

where x_{ij} is the concentration of the jth species in the ith sample, g_{ik} is the contribution of kth source to ith sample, f_{kj} is the concentration of the jth species in the kth source, and e_{ij} is the residual for each sample/species.

RMs are commonly used to apportion PM concentration on the basis of its chemical composition: major ions, carbonaceous fractions, trace elements and organic markers. From the physio-chemical analysis of PM components, it is possible to estimate the pollution sources attributed to the total PM_{10} and $PM_{2.5}$ mass, along with associated uncertainties.

We systematically searched the Scopus database and the World Wide Web using Google search for peer-reviewed SA studies with combinations of the following key words: source apportionment, receptor models, particulate matter (PM), aerosols, PM_{10}, $PM_{2.5}$ (i.e. particulate matter smaller than 10 and 2.5 μm, respectively), ambient PM, air pollution, air quality. The research covered the period between 1990 and 2014 (year of publication). Data gathered from SA studies were reviewed according to criteria described in the following sections. Reviewed SA data were used to populate a database (WHO, 2015).

2.3. Source categories

Six categories of main sources of ambient particulate matter (PM) commonly found in SA studies which used receptor models and compatible for grouping, have been used for the purpose of this analysis: traffic, industry, domestic fuel burning, natural sources including soil dust (re-suspended) and sea salt, and unspecified sources of pollution of human origin. The categories were characterized by the profile of their chemical components (Table A.2, Supplementary material) to match the six identified categories using the component profile of the European Guide on Air Pollution Source Apportionment with Receptor Models (Belis et al., 2014). The selected categories of sources of pollution are described in the following sections.

Traffic is a source category that includes different kinds of emissions from various vehicle types. In addition to primary PM emissions from exhaust, and the emissions of organic and inorganic gaseous PM precursors from the combustion of fuels and lubricants, vehicles emit significant amounts of particles through the wear of brake linings, clutch, and tires (Amato et al., 2009; Belis et al., 2013). These are deposited onto the road and then re-suspended by vehicle traffic together with crustal/mineral dust particles and road wear material.

Industry is a heterogeneous category including mainly emissions from oil combustion, coal burning in power plants and emissions from different types of industries (petrochemical, metallurgic, ceramic, pharmaceutical, IT hardware, etc.) and from harbor-related activities. Industrial sources are sometimes mixed with unidentified combustion sources or traffic (Belis et al., 2013).

Domestic fuel burning includes wood, coal and gas fuel for cooking or heating. A typical case of household air pollution coal burning for domestic heating is found in Central Europe where wood and coal are used for domestic heating.

Natural sources including soil dust and sea salt. Dust is characterized by elements abundant in the earth's crust rocks and the soil. These components of PM are associated with the re-suspension from fields or bare soils by local winds. When reported separately, road dust was included in the traffic source category in this study (Belis et al., 2013). Even though re-suspension of natural soils and road-dust emitted and re-suspended by vehicular traffic can be distinguished using markers of brakes, tires, and road wear, separating these two contributions may be challenging. **Sea salt** particles in the air can be found close to the coast. In continental areas, it can be transported by wind from coastal areas, or come from road salt (Seinfeld and Pandis, 2006).

The "**Unspecified sources of human origin**" category mainly includes secondary particles formed from unspecified pollution sources of human origin. Primary particle emissions include mechanically generated particles and primary carbonaceous particles. Primary particles also include carbonaceous fly-ash particles produced from high temperature combustion of fossil fuels in coal power plants.

Secondary particles are formed in the atmosphere through reactions of primary gaseous pollutants (nitrogen dioxide NO_2, ammonia NH_3, sulfur dioxide SO_2, non-methane volatile organic compounds NMVOCs). Secondary particles can either be inorganic or organic aerosols (Finlayson-Pitts and Pitts, 2000).

Inorganic sulphate aerosols are formed from agricultural NH_3 emissions that combine with SO_2 from, shipping and industrial activities and power generation. Inorganic nitrate aerosols are formed through a combination of NH_3 with NOx emissions from traffic, power generation, industrial and residential sources. Secondary inorganic aerosols may be associated to "long range transport", since the residence time of sulphates and nitrates in the atmosphere is between 3 and 9 days (Seinfeld and Pandis, 2006). Secondary organic aerosols (SOAs) can be emitted directly (e.g., from combustion of biomass, wood, coal – primary organics), formed in the air through combinations with other substances (secondary) or formed from biogenic VOCs. Among SOAs there are NMVOCs from industrial processes, industrial and household consumption of solvents (e.g. in paints, degreasers and stain removers) via evaporation, combustion processes and, transport emissions. In addition, most NMVOC play an important role in ozone formation. Certain NMVOC are harmful to health or even carcinogenic (e.g. benzene).

Main economic sectors attributed to the "Unspecified sources of human origin" source category may also include industrial sources and long-range transport of mixed pollutant sources including the industry, traffic, agriculture and shipping.

As reported below, dust and sea salt have been combined together and, in the following, we will referrer to them as natural sources. In addition, with the term "SA records" we will specifically refer to the data and metadata reported by SA case studies carried out for PM_{10} or $PM_{2.5}$. On the other hand, with the term "SA studies" we will refer to the work and methodology carried out for the estimation of SA data.

2.4. Quality indicator for source apportionment records

We developed a quality rating for SA records reported in the database. The rating has been based on the number of the source categories of ambient PM identified by the records and on the time duration of the measurements (annual or seasonal). The rating has not considered the type of receptor model used to estimate the number of source categories. Receptor model outputs are strongly related to the protocol followed for the source apportionment analysis rather than to the model itself (Belis et al., 2014).

The rating criteria for SA records are shown in Table 1. Records reporting all five source categories and, with measurements carried out on an annual basis have been rated as "6", that means a "high (quality)" study. 15% of the records were rated as "6". On the other hand, records reporting five source categories but, with measurements carried out on seasonal basis, have been rated as "5" that means a "partial high" (quality) study. Using these criteria, other records have been quality rated as "average" ("4"), "partial average" ("3"), "basic" ("2") and "partial basic" ("1") (Table 1).

The majority of the records (39%) has been carried out on annual basis and reported four source categories. These records were rated as "average" (quality) records. A non-negligible number of records (17%) reported three out of five source categories. Instead only 2% of the records reported 2 sources categories. SA records reporting three and two source categories were therefore rated as "basic".

2.5. Computation of regional averages of source apportionment

Regional averages of SA records represent the city population-weighted averages of particulate matter attributed to economic sector in each country-region for records. Country-weighted averages have also been developed, and are based on the same approach. Source contributions to urban ambient $PM_{2.5}$ and PM_{10} were analysed and reported separately. In addition, for studies analyzing both components, the chemical speciation of $PM_{2.5}$ and PM_{10} attributed to source categories have been compared.

For each source category, the regional averages were computed as the city population-weighted averages after dividing the city population by the number of case studies available for the same city:

$$\boldsymbol{S_i} = \frac{\sum_{\boldsymbol{x}=1}^{\boldsymbol{n}} \frac{\boldsymbol{Cpop_x}}{\boldsymbol{N_c}} * \boldsymbol{s_{xi}}}{\sum \boldsymbol{Cpop}} \quad (2)$$

where S_i is the average SA value for source category i, $Cpop_x$ the city population for each study x, N_c the number of times a city C is represented, and s_{xi} the SA record for each SA study x and source category i. For regions with three records or less, the arithmetic (rather than weighted) average was taken, in order to consider them as random samples. Where several studies were available for the same city, those studies with two or more source categories missing were excluded if more complete studies were available. When a source category was missing for a study, averages have omitted that source category for that study. In the case the total of the weighted averages of the categories was not equal to 100%, the same multiplier was applied to all source categories in order to reach 100%.

Furthermore, we used the following approaches:

a) source categories "dust" and "sea salt" were combined into one source "Natural Sources (dust and sea salt)"

Table 1
Quality of collected SA records.

Quality indicator for SA records				
Number of source categories	Duration of the measurement	Number of SA records (% to the total number of records)	Rating	Quality rating description
5	Year	80 (15%)	6	High
	Season	14 (3%)	5	Partial high
4	Year	208 (39%)	4	Average
	Season	73 (14%)	3	Partial average
3	Year	89 (17%)	2	Basic
	Season	50 (9%)	1	Partial basic
2	Year	10 (2%)	2	Basic
	Season	5 (1%)	1	Partial basic

b) only records with annual averages were considered with the exception of records from countries where measurements were only performed in a particular seasons.

3. Results

3.1. Source apportionment records

3.1.1. The global source apportionment database

We collected and populated a database (DB) with 529 records from SA studies using receptor models. These records are from studies published between 1990 and 2014 and cover cities of 51 worldwide countries for a total of 560 million people (Table 2). 284 out of the 529 total records were carried out in countries other than Europe. Listing of the SA records that populate the DB is reported in the Supplementary material (Table A.3) and include: name of site location, country, country region, site typology, PM class, methodology (type of Receptor Model), literature references, number of sources of ambient PM, time period of the measurement, and rating of the SA record as defined in Table 1. The database is openly available at the website of the World Health Organization (WHO, 2015). SA studies have been ordered alphabetically by country and then by site location within each country. Most of the SA records have been carried out in urban areas (77%) followed by rural (14%), remote (5%) and, industrial (4%) areas. These four site categories also include sub-categories as reported in Table A.3 (Supplementary material).

The largest number of SA records was found for the region of Northern, Western and Southern Europe (239) followed by the USA (106) and the China regions (43). Between 14 and 23 SA records were found for the Rest of Americas, Brazil, India region, Central Europe and South Eastern Asia. Between three and eight SA records per region were found for each of the remaining regions.

Sources of ambient PM have been grouped following criteria reported in European Guide on Air Pollution Source Apportionment with Receptor Models (Belis et al., 2014) and in recent work published by Belis et al. (2013). As reported above, we considered five categories of ambient PM: “traffic”, “industry”, and “domestic fuel burning”, “unspecified sources of human origin” and, “natural sources (dust and sea salt)”.

“Unspecified sources of human origin” were identified in all the records (100%) followed by natural source (dust and sea salt) (89%), traffic (87%), industry (66%) and domestic fuel burning (45%) (Table 2). Therefore, not all records identified the same number of sources. Annual SA measurements were identified in 73% records, whereas the remaining records (27%) performed measurements during specific seasons of the year.

We identified 14 types of receptor models used for the estimation of sources of ambient PM. Positive Matrix Factorization (PMF) and Chemical Mass balance were the models mostly used in 45% and 19% of the SA records, respectively (Table A.3, Supplementary material).

3.1.2. Source apportionment records in urban areas

In this work, we considered 419 out of 529 records from SA studies carried out before the year 1990 in urban areas (96%) and in a limited number of industrial site locations (4%) with high population density. 218 out of 529 SA records were for countries other than Europe. Fig. 1 shows the geographical locations of SA records for $PM_{2.5}$ and PM_{10} for urban and industrial sites considered for the calculation of regional averages of source contributions performed in this work. Only SA measurements collected after the year 1990 were considered for the calculation of regional averages of source contribution according to the approaches discussed in Section 2.5.

3.2. Global and regional averages of source contributions

The present analysis of SA studies highlights the complexity of mapping source categories of individual studies to a common set of source categories. Many of the SA studies use very different source categories that were mainly determined by the available techniques for the chemical analyses. In addition, in most of SA studies, a large portion of PM remains unexplained. This often includes secondary particles (here called “unspecified sources of human origin”). However, the chemical fractions and source categories responsible for these unexplained fractions might differ quite substantially across studies.

3.2.1. $PM_{2.5}$ global and regional averages

With the above caveats, the 419 SA records used in this study

Table 2
Summary of the typology of the collected SA records.

Site typology (51 countries 560 mill. people)	Number and percentage of SA records published as of August 2014[a]	
Urban	397	77%
Rural[b]	74	14%
Remote[b]	25	5%
Industrial	22	4%
	Total = 529	
Ambient PM source categories identified in SA records		
Traffic	462	87%
Industry	347	66%
Domestic fuel burning	238	45%
Unspecified sources of human origin	524	100%
Natural sources (dust and sea salt)	468	89%
Duration of the measurements		
Annual average SA records	387	73%
Seasonal SA records	70 (winter)	13%
	20 (spring)	4%
	31 (summer)	6%
	20 (fall)	4%

[a] Listing of the Source Apportionment records can be found in Table A.3 of the Supplementary material.
[b] Rural and Remote sites have not been considered for the analysis carried out in this work.

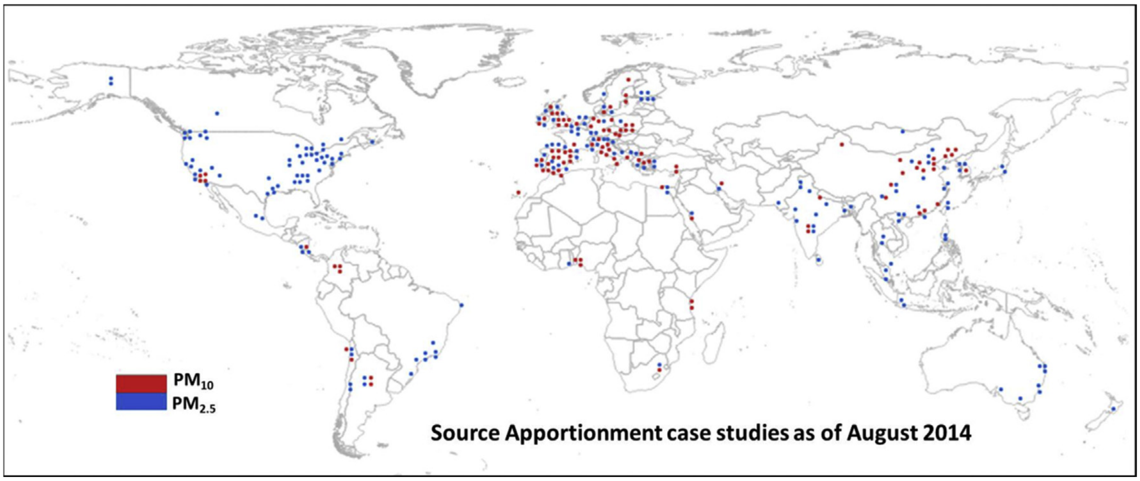

Fig. 1. Geo-location of Source Apportionment studies for $PM_{2.5}$ and PM_{10} considered for the regional averages of source contributions performed in this study. Rural and remote sites have not been displayed.

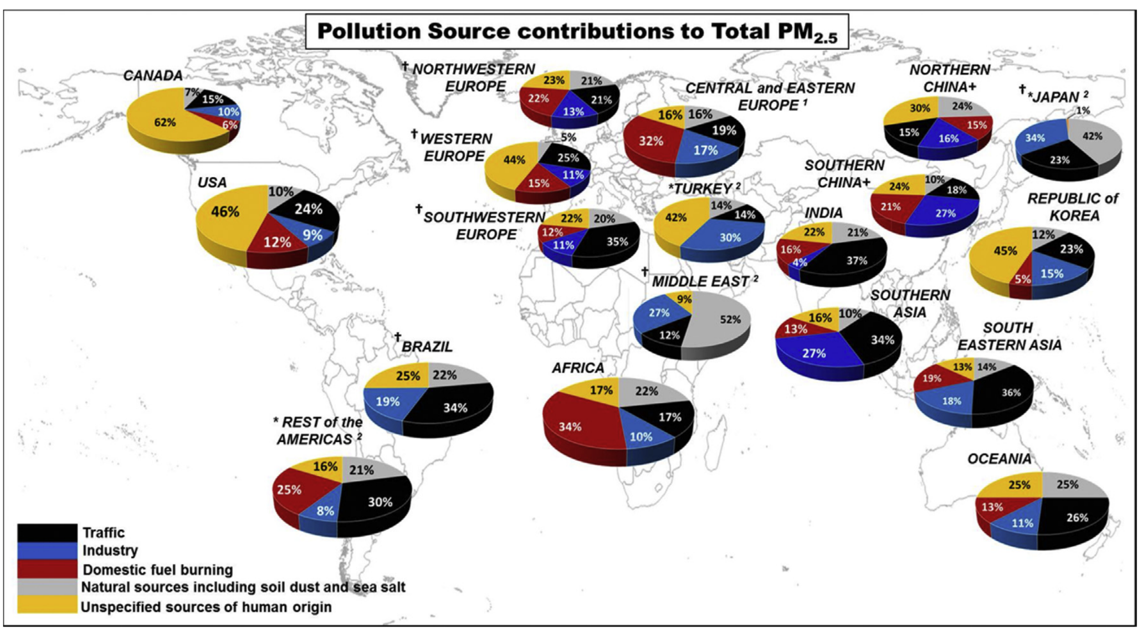

Fig. 2. Population-weighted averages for relative source contributions to total $PM_{2.5}$ in urban sites. *, †regions in which (*) unspecified sources of human origin and (†) domestic fuel burning sources have not been assessed. [1]Based only on one study including domestic fuel burning, and therefore only provides indicative results. [2]Based only on two studies, and therefore only provides indicative results.

suggest that, from global averages of source contributions, 25% of urban ambient particulate matter ($PM_{2.5}$) was contributed by traffic, 15% by industrial activities including power generation, 20% from domestic fuel burning, 22% from unspecified sources of human origin, and 18% from natural dust and sea salt (Table 3).

The contributions by source category varied substantially across regions. Traffic was the main contributor to urban ambient $PM_{2.5}$ in several regions, including India (37%), South Eastern Asia (36%), Southwestern Europe (35%), Southern Asia (34%), Brazil (33%), and the Rest of the Americas (30%) (Fig. 2 and Table 3). Most studies did not clearly separate local small combustion sources from other source categories, and contribution could have been erroneously attributed to traffic or industrial activities resulting in overestimation of these sectors. Most SA studies allocated traffic based on primary PM emissions from vehicles. These primary emissions mainly include diesel soot and non-exhaust emissions such as brakes and tires. On the other hand, secondary inorganic compounds, such as secondary inorganic nitrates formed from NO_x emissions, are usually not apportioned to traffic.

Industrial activities had the highest contributions from human activities in, Japan (34%), Middle East and Southern Asia (27%), Turkey (30%), Brazil (19%), Central Europe (17%), and South Eastern Asia (18%) (Fig. 2 and Table 3).

Domestic fuel burning emerged as the main contributor in Africa (34%), and in Central and Eastern Europe (32%) although based on very few records in this region (Fig. 2 and Table 3). Domestic fuel burning was also important in the Rest of the Americas (25%), Northwestern Europe (22%), the Southern China region (21%), South Eastern Asia (19%), and India (16%).

Unspecified pollution sources of human origin, mostly attributed to secondary particle formation, accounted for important contributions in Canada (62%), USA (46%), Republic of Korea (45%),

Table 3
Percent of source attribution to urban ambient $PM_{2.5}$ and PM_{10} by source category.

Region[a]	Traffic[b]		Industry[b]		Domestic fuel burning[b]		Unspecified source of human origin[b]		Natural sources (dust and sea salt)[b]	
	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}
Africa	17	34	10	6	34	21	17	14	22	25
Brazil	34	na	19	na	na	na	25	na	22	na
Canada	15	na	10	na	6	na	62	na	7	na
Central and Eastern Europe	19	8	17	18	32	45	17	26	16	3
Northern China+	15	25	16	21	15	19	30	18	24	17
Southern China+	18	17	27	22	21	7	24	27	10	27
India	37	34	4	14	16	11	22	12	21	29
Southern Asia	34	na	27	na	13	na	16	na	10	na
Japan	23	na	34	na	na	na	1	na	42	na
Republic of Korea	23	21	15	17	5	3	45	43	12	16
Middle East	12	15	27	23	na	4	9	14	52	44
Oceania	26	na	11	na	13	na	25	na	25	na
Rest of the Americas	30	38	8	26	25	na	16	12	21	24
South Eastern Asia	36	na	18	na	19	na	13	na	14	na
Turkey	14	16	30	29	na	na	42	39	14	16
USA	24	30	9	na	12	15	46	44	10	11
Northwestern Europe	21	12	13	12	22	24	24	19	21	33
Western Europe	25	28	11	22	15	7	44	22	5	21
Southwestern Europe	35	23	11	11	12	7	22	20	20	39
Low- and middle-income countries[c]	24	25	16	16	22	18	21	19	20	24
High-income countries[c]	26	30	12	21	17	15	29	25	17	22
World	25	25	15	18	20	15	22	20	18	22

na: not available.
[a] See Table A.1 (Supplementary material) for country grouping.
[b] Rural and remote sites have not been considered for the analysis carried out in this work.
[c] Classification according to the World Bank list of economies for the year 2012.

Western Europe (44%), and Turkey (42%) (Fig. 2 and Table 3).

Natural dust has been found as the main contributor of $PM_{2.5}$ in the Middle East (52%). Instead, relevant contribution have been found in Japan (42%), followed by Oceania (25%), Brazil and Africa (22%).

3.2.2. PM_{10} global and regional averages

Analysis of global and regional averages of source contributions to urban PM_{10} levels has been also carried out (Fig. 3 and Table 3). Globally, 25% of urban ambient particulate matter (PM_{10}) was contributed by traffic, 18% by industrial activities including power generation, 15% from domestic fuel burning, 20% from unspecified sources of human origin, and 22% from natural dust and sea salt and (Table 3).

At regional level, traffic showed higher contribution to PM_{10} rather than $PM_{2.5}$ in the Rest of the Americas (38%), Africa and India (34%), USA (30%, based on only one study), Western Europe (28%), and Northern China region (25%). This might explained by the fact

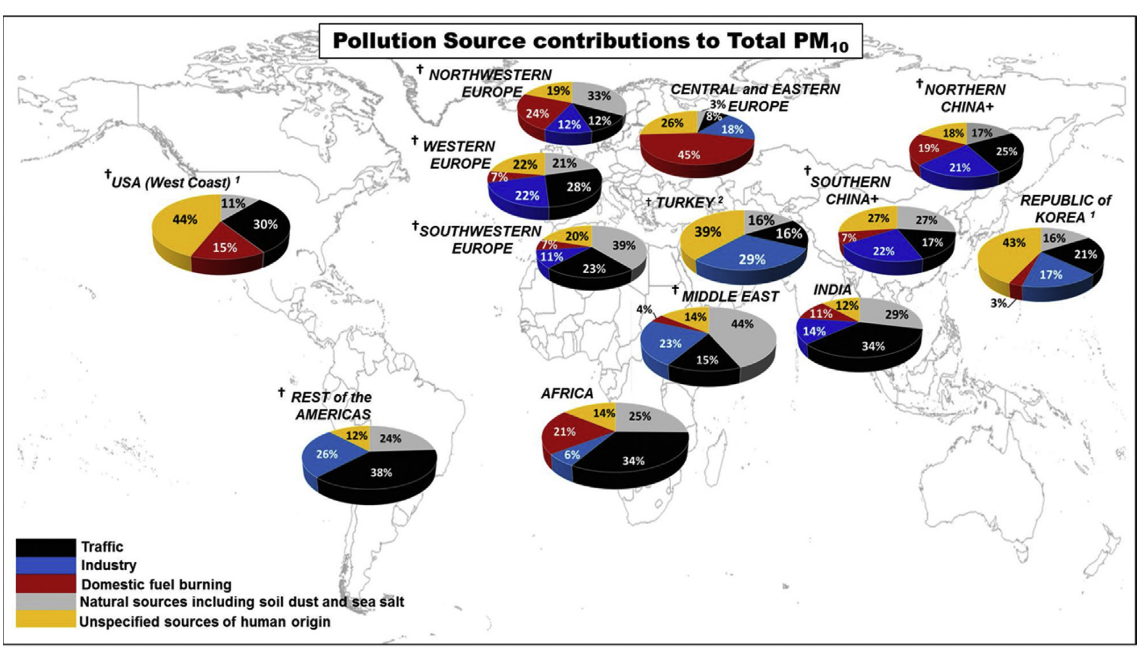

Fig. 3. Population-weighted averages for relative source contributions to total PM_{10} in urban sites. † regions in which domestic fuel burning have not been assessed. [1]Based only on one study, and therefore only provides indicative results. [2]Based only on one study including traffic, and therefore only provides indicative results.

that several SA studies have apportioned part of re-suspended dust to the traffic source rather than to the natural sources. This portion of dust comes from road wear material that is re-suspended by traffic vehicles and assigned to the PM_{10} fraction.

Industry has been found as important contributor of PM_{10} in Turkey (29%) as well as Rest of the Americas (26%), Western Europe (22%) and in the China regions (21–22%).

Domestic fuel burning emerged as the main contributor to PM_{10} in Central and Eastern Europe (45%) followed by Northwestern Europe (24%), Africa (21%), and the Northern China region (19%).

With the exception of Middle East and Central and Eastern Europe, natural sources (dust and sea salt) showed higher contribution to PM_{10} compared to $PM_{2.5}$.

In general, unspecified sources of human origin (secondary particles) showed lower contribution to PM_{10} rather than $PM_{2.5}$. This is in line with the evidence that secondary particles are classified as fine and therefore assigned to $PM_{2.5}$. Major contribution of this source to PM_{10} was found in the USA (44%, based on only one study), Republic of Korea (43%, based only one study), Turkey (39%), and in the Southern China region (27%).

Natural source emerged as the main contributor in Middle East (44%), Southwestern Europe (39%), Northwestern Europe (33%), the Southern China Region (27%), Africa (25%), and Rest of the Americas (24%).

We want to stress out that the above findings need to be seen in the perspective of the number and quality of the SA records used for this analysis (Table 4).

The absolute contributions to ambient particulate matter (PM) by region and source are listed in Fig. A.1 ($PM_{2.5}$) and Fig. A.2 (PM_{10}) of the Supplementary material. The Northern/Southern China, India, Southern Asia and Africa regions, showed the highest urban $PM_{2.5}$ concentrations in average. On the other hand, the Middle Eastern, Northern China, Indian and African regions showed the highest absolute urban PM_{10} concentrations.

A significant number of sites reported source contribution for both $PM_{2.5}$ and PM_{10} in the Western European region and the Rest of the Americas. In those regions, averages of differences between PM_{10} and $PM_{2.5}$ showed differences below 10% for each category (Figs. A.3 and A.4, Supplementary material). Dust and sea salt showed higher contribution in PM_{10} rather than in $PM_{2.5.}$ On the other hand, unspecified sources of human origin, industry and traffic showed higher contribution in $PM_{2.5}$ rather than in PM_{10}. This confirms that natural sources are mostly associated to the PM_{10} fraction whereas, secondary particle formation from anthropogenic activities are associated to the $PM_{2.5}$ fraction.

We reported the ratings of SA records averaged by region to give an estimate of the quality of SA studies carried out in worldwide regions (Table 4). Most of low and high income countries showed a rating between 3.5 and 5, that is and an indicator of "average" quality. Only few region regions such as, Canada, Republic of Korea and Oceania showed "high" rating. On the other hand, no region was found with the "basic" rating (Table 4). Finally, at worldwide level, the overall rating was found as "average" for both $PM_{2.5}$ and PM_{10}.

4. Discussion and conclusions

This work represents a first attempt to compile available source apportionment (SA) studies of particulate matter at the global scale. It is found that source categories distinguished in individual SA records vary greatly, and a mapping to a common set of source sectors remains ambiguous. Furthermore, although atmospheric modeling studies indicate a large contribution from solid fuel combustion in households, this category is often not distinguished in many SA studies, or lumped together with traffic emissions. Also, secondary aerosols are often not allocated to the source of their precursor emissions, which might mislead results. While identification of specific pollution sources could provide essential information to guide policy makers in reducing pollution at local level, the imperfections of current SA studies mandate cautious interpretation of the available results. Particularly, the low relative contribution from local small scale combustion of solid fuels, i.e., cook stoves and heating using biomass, wood and coal, suggested by the current SA studies contradicts the much larger role that is

Table 4
Average rating of $PM_{2.5}$ and PM_{10} records calculated at regional and global level.

Region[a]	Number of records ($PM_{2.5}$)[b]	Average rating of records ($PM_{2.5}$)[c]	Number of records (PM_{10})[b]	Average rating of records (PM_{10})[c]	Income classification
Africa	4	5.0	7	4.6	L
Brazil	8	3.1	0	na	L
Canada	6	5.5	0	na	H
Central and Eastern Europe	3	4.7	8	2.5	L
Northern China+	12	3.8	13	5.0	L
Southern China+	8	3.1	6	4.3	L
India	7	4.4	3	3.7	L
Southern Asia	6	3.2	0	na	L
Japan	2	3.5	0	na	H
Republic of Korea	3	5.7	1	6.0	H
Middle East	2	2.5	2	3.5	L
Oceania	8	5.5	0	na	L
Rest of the Americas	13	3.2	11	3.0	H
South Eastern Asia	12	4.3	0	na	L
Turkey	2	5.0	3	3.0	L
USA	64	4.7	1	3.0	H
Northwestern Europe	20	4.4	21	4.5	H
Western Europe	10	4.4	13	4.6	H
Southwestern Europe	56	3.2	84	3.5	H
Low- and middle-income countries[d]	72	4.0	42	4.0	
High-income countries[d]	152	3.9	131	3.7	
World	224	3.9	173	3.8	

na: not available.
[a] See Table A.1 (Supplementary material) for country grouping.
[b] Rural and Remote sites have not been considered for the analysis carried out in this work.
[c] Rating description as quality indicator: 6 = High; 5 = Partial High; 4 = Average; 3 = Partial Average; 2 = Basic; 1 = Partial Basic.
[d] Classification according to the World Bank list of economies for the year 2012 (The World Bank, 2013). H=High income; L = Low income.

found in other studies (Brauer et al., 2012; Junninen et al., 2009).

Although current estimates from SA studies need further validation, the regional averaging of 419 local SA records to ambient PM indicates which could be the most important contributors to urban disease from ambient air pollution. Contributions vary importantly across regions, with traffic, industrial activities, domestic fuel burning or natural dusts being the most prominent contributors. Limitations of the current records stem from important uncertainties in the SA studies, the total exposure to ambient $PM_{2.5}$ and the disease burden estimations. The main sources of uncertainties include (a) the limited number and representativity of available SA studies for the respective urban populations in each region; (b) the relatively large portions of unclearly attributed categories, namely "unspecified" and "dust"; and (c) the difficulties in attribution to source categories experienced in certain SA studies.

Regarding the uncertainty (a) listed above, while as many as 419 records from SA studies conducted mostly in urban areas could be identified and used in this study, a number of regions have no or very few data. These include for example Africa, parts of Eastern Europe (i.e. the Kazakhstan Region, Ukraine Region and Russia), and Northern Africa. Source contribution to ambient PM in low- and middle income countries is particularly poorly documented with 114 records, as compared to 305 records in high-income regions (corresponding to six per urban inhabitant in low- and middle income countries). Ambient air pollution is however highest in low- and middle income countries (WHO, 2014b). Also for PM_{10}, the information available in certain regions can be scarce.

In terms of unclearly attributed categories (uncertainty (b)), studies allocate 18% of $PM_{2.5}$ and 22% of PM_{10} to "dust", i.e. an unspecified category of mostly natural origin, instead 22% of $PM_{2.5}$ and 20% of PM_{10} was allocated to "unspecified sources of human origin". In certain regions, however, the total share of unclearly attributed particles can reach more than 50%, e.g. in North America (Canada) or the Middle East. This may come from mixtures of particles from industrial activities such as power plants and particles from secondary reactions of chemical compounds attributed to transport emissions that render attribution more difficult. In the Middle East, desert dusts can amount for a large share of fine particles.

Uncertainty (c), difficulties to attribute source categories in certain SA studies, because of the lack of information referring to specific chemical compounds that permit identification of unique pollution sources.

The quality or representativity of studies according to the criteria used for the rating of SA records in this study did not vary much according to the income level. However, the mean overall rating corresponded to "average" only, and could be improved in the future. SA studies are not always representative for all seasons of the year, and often lack important source categories.

Whereas, for most of regions (excluding Middle East and Central and Eastern Europe), natural sources have shown higher contribution in PM_{10} rather than in $PM_{2.5,}$ the "unspecified source of human origin", industry and traffic, have shown higher contribution in $PM_{2.5}$ rather than in PM_{10}. This indicates that the influence of natural sources is relatively high on PM_{10} than $PM_{2.5}$ concentrations and, combustion sources are more important in $PM_{2.5}$ than in PM_{10} formation. However, we should also consider ultrafine particles (particulate matter smaller than 100 nm, PM_1) produced in large quantities by vehicular combustion and their role for causing several adverse human health effects (Knibbs et al., 2011). Therefore, the analysis carried out in this work for $PM_{2.5}$ might have different outcomes if ultrafine particles formation would be attributed to the traffic source category.

More extensive networks of air quality monitoring stations in low-and middle-income countries would allow for more SA studies and provide a more accurate picture of pollution sources, provided they follow a quality-controlled approach that distinguishes all sectors. The results of this study however already provide an approximate indication of which human activities and natural sources are important contributors to disease due to cities' ambient PM. It provides key information for policy makers in the health and environment sectors, and those sectors which are determining the polluting sources of concern (energy, transport, industry).

This study provides regional and country averages of major sources of urban ambient PM, which are larger scale indications for the main categories contributing to particular matter. For planning local actions in view of reducing the adverse health effects from ambient PM, in locations with locally specific industries, additional local information is required.

This study however suggests traffic, industry, and biomass burning in low- and middle income countries as important contributors to fine particle in cities. Prioritizing healthy transport solutions, which may lead to additional reductions in noncommunicable disease rates, such as appropriate physical activity levels and reduced release of agents leading to climate change, may be a promising solution in view of disease prevention. Reductions of industrial emissions and access to clean energy in households will complete the picture in the progress towards a healthier and more sustainable environment.

Finally, this work summarized the current available source apportionment case studies performed around the world. In order to help policy makers to obtain a convincing and realistic attribution of sources of ambient PM, the average outcome of existing studies made in this work requires further data collection. Future work might be oriented toward the integration of information from emission inventories, dispersion modelling, time series and chemical monitoring in source apportionment studies.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.08.087.

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