Issues in Environmental Science and Technology

Airborne Particulate Matter

Sources, Atmospheric Processes and Health
Volume 42

Edited by R. E. Hester, R. M. Harrison
and Xavier Querol

**ROYAL SOCIETY
OF CHEMISTRY**

Airborne Particulate Matter
Sources, Atmospheric Processes and Health

ISSUES IN ENVIRONMENTAL SCIENCE AND TECHNOLOGY

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42
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Preface

Airborne particulate matter has been a highly topical subject for the past 20 years or so, although it has been a problem pollutant for very much longer. Historical records of problems with coal smoke pollution go back many centuries, but the first good quantitative records date back to the early 20th century when it was measured as ''black smoke''. An episode of severe pollution by black smoke and sulfur dioxide in December 1952 is believed to have caused around 4000 premature deaths in London alone, and comparable events occurred in other parts of the world. The recognised toxicity of black smoke and sulfur dioxide led to the development of mitigation policies, which greatly improved the quality of the atmosphere to the point that it was felt that there were no significant remaining effects on public health arising from these pollutants in most developed countries. However, the application of more advanced epidemiological methods in the 1990s demonstrated that significant public health impacts of exposure to airborne particles, now measured by the mass metrics $PM_{2.5}$ and PM_{10}, existed in North America and Europe and, by implication, in other parts of the world. Whilst most aspects of local air quality continued to improve in developed countries, this was a time when less developed countries were growing their economies rapidly, with an accompanying substantial increase in the concentration of airborne particles. As a result, the World Health Organisation's Global Burden of Disease Project ranks exposure to outdoor particles as the ninth largest preventable cause of disease in the global population.

This volume of the *Issues* series addresses airborne particulate matter from a global perspective. The first chapter, by Marc Guevara of the Barcelona Supercomputing Centre, gives quantitative information on the sources of particle emissions in a European context. Not only are particles emitted directly into the atmosphere, but they also form within the atmosphere from the oxidation of gases and undergo chemical transformations in the atmosphere. The latter secondary fraction typically makes the dominant contribution to $PM_{2.5}$ concentrations. In the second chapter, Neil Donahue of Carnegie Mellon University and co-authors pose the question: What are

the sources of particles? Their focus is on the secondary particles that are formed within the atmosphere and how the sources affect human exposure to airborne particles, both by mass and by number.

One of the key advances in the past 20 years has been the development of receptor modelling methods that take air quality data and use it to provide estimates of the quantitative contribution of different sources to measured concentrations. In the third chapter, John Watson and Judy Chow of the Desert Research Institute, Nevada, outline the principles of these methods, and in the fourth chapter, Philip Hopke of Clarkson University, New York State, provides further insights into methods and gives some case studies from North America. The following two chapters take data from two major European experimental studies to provide case studies of the application of receptor modelling methods to source apportionment. In doing so, they give powerful insights into the sources of particles in European cities. The AIRUSE LIFE+ team led by Xavier Querol of the Consejo Superior de Investigaciones Cientificas (CSIC) describes a study of cities in southern Europe. This is complemented by a study from northern Europe (the Joaquin Project) led by Edward Roekens of the Flanders Environment Agency, which has applied similar methods to five cities within northern Europe.

The subsequent two chapters provide insights into the apportionment of $PM_{2.5}$ (fine particles) and particulate matter of various size fractions in two of the world's hotspots. In Chapter 7, Mei Zheng and colleagues from Peking University consider the source apportionment of $PM_{2.5}$ within China, and in Chapter 8, Mukesh Khare and Isha Khanna of the Indian Institute of Technology, Delhi, review case studies of source apportionment from the Indian sub-continent.

No such volume would be complete without also considering the adverse health effects of airborne particles. Consequently, in Chapter 9, Frank Kelly of Kings College, London, reports on the health effects of airborne particles and how these relate to their composition, size and source.

We are delighted to have engaged such a distinguished group of authors, including several world leaders in their respective fields, to provide a truly authoritative and up-to-date volume describing many of the key aspects of airborne particulate matter. We believe that this will prove to be of widespread interest and be of great value to a range of communities, including policymakers, physical and life scientists, and students taking advanced courses in a range of environmental and health-related fields.

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Ronald E. Hester
Roy M. Harrison
Xavier Querol

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Roy M. Harrison is Queen Elizabeth II Birmingham Centenary Professor of Environmental Health in the University of Birmingham. He was previously Lecturer in Environmental Sciences at the University of Lancaster and Reader and Director of the Institute of Aerosol Science at the University of Essex. His more than 400 publications are mainly in the field of environmental chemistry, although his current work includes studies of human health impacts of atmospheric pollutants as well as research into the chemistry of pollution phenomena. He is a past Chairman of the Environment Group of the Royal Society of Chemistry for whom he edited 'Pollution: Causes, Effects and Control' (RSC, 1983;

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Emissions of Primary Particulate Matter

M. GUEVARA

ABSTRACT

Particulate matter (PM) accounts for a complex group of air pollutants with properties and impacts that vary according to its composition and size. The emission rates, size and composition of primary PM emissions are challenging to determine since they depend not only on the sector considered, but also on the fuel properties, technology and other characteristics of the emission process. At the European level, fine carbonaceous particles are generally the dominant components of primary PM emissions, the most important sources of organic and black carbon being residential biomass combustion and diesel vehicle engines, respectively. On the other hand, soil particles generated by wind erosion processes, traffic resuspension, mining and construction operations, and agricultural land management activities are large contributors to the coarse fraction of primary PM emissions. European PM emissions are decreasing as a result of implemented EU legislation mainly focused on road transport and large point sources. Nevertheless, emissions released by residential solid fuel appliances have been increasing due to a lack of regulations, a tendency that is expected to change with the eco-design directive. The decrease of traffic PM exhaust emissions has also increased the importance of traffic non-exhaust emissions, a major source of metals in urban areas.

1   Introduction

Particulate matter (PM) is a generic term used to describe a mixture of solid particles and liquid droplets (aerosols) that vary in size and composition, depending on the location and time[1] (Table 1).

Table 1 Sources of origin and main components of coarse $PM_{10-2.5}$, fine $PM_{2.5}$ and ultrafine $PM_{0.1}$ primary particles.[a]

PM fraction	Sources of origin	Main components	Contribution
Coarse particles ($PM_{10-2.5}$)	Agricultural activities	Agricultural soil, OC	$+++$
	Traffic resuspension	Road dust	$+++$
	Windblown dust/ construction and mining activities/industrial resuspension	Si, Al, Ti, Fe	$+++$
	Tyre and brake wear	Cu, Zn	$++$
	Combustion in energy and manufacturing industries (coal, coke, heavy oil)	EC	$++$
	Wind-land fires and volcanoes	Volcanoes' ashes, burned OC	$+$
	Biological sources	Plant debris and fungal spores	$+$
	Ocean spray	Na, Cl, Mg	$+$
Fine ($PM_{2.5}$) and	Diesel-fuelled vehicle engines	BC	$+++$
Ultrafine particles ($PM_{0.1}$)	Biomass combustion	OC, PAHs	$+++$
	Maritime traffic	BC, OC, SO_4^{-2}	$++$
	Combustion in energy and manufacturing industries	Pb, Cd, As, Cr, V, Ni, Se, SO_4^{-2}	$++$
	Processes in non-metallic industries	Si, Al, Fe	$+$
	Metal processing activities	Pb, Cd, Cr, Zn	$+$

[a] $+++$ High contribution; $++$ Medium contribution; $+$ Low contribution.

PM is made up of a large number of components, including elemental or black carbon (BC) and organic carbon (OC) compounds, sulfate (SO_4^{-2}), nitrate (NO_3^-), trace metals, crustal material (*i.e.* soil particles) and sea salt.[2] PM also comes in a wide range of sizes and includes PM with diameter less than or equal to 10 μm (PM_{10}), PM with diameter less than or equal to 2.5 μm ($PM_{2.5}$), also denoted as fine particles, PM with diameter less than or equal to 0.1 μm ($PM_{0.1}$), also denoted as ultrafine particles (UFP), and PM with diameter less than or equal to 0.05 μm ($PM_{0.05}$), also denoted as nanoparticles.[3]

In terms of source of origin, PM can be directly emitted from anthropogenic (man-made) or natural sources (*i.e.* primary PM), or formed in the atmosphere from a series of gaseous combustion by-products such as volatile organic compounds (VOCs), ammonia (NH_3), oxides of sulfur (SO_x) and oxides of nitrogen (NO_x) (*i.e.* secondary PM). Primary PM originates predominantly from combustion (*e.g.* vehicle engines) and high-temperature processes (*e.g.* smelting and welding industrial operations),[4,5] as well as from mechanical disruption processes and man- or wind-induced events causing suspension of particles (*e.g.* traffic resuspension of street dust).[6,7]

On the other hand, secondary PM is formed by gas-to-particle conversion in the atmosphere and/or condensation of gaseous compounds on pre-existing aerosol particles, mainly involving NO_x, SO_x, NH_3 and VOCs, which may react with O_3, $^\bullet OH$ and other reactive molecules forming secondary inorganic aerosols (SIA) and secondary organic aerosols (SOA).[3]

Unlike other pollutants, such as SO_2 or NH_3, PM describes a complex group of air pollutants with properties and impacts that vary according to their composition and size. For instance, BC is linked to a range of climate impacts (*e.g.* increased temperatures) owing to its capability of directly absorbing light, reducing the albedo of snow and ice and interacting with clouds.[8] On the other hand, several European cohort studies have reported that short- and long-term exposure to $PM_{2.5}$ is associated with a number of health risks, such as lung cancer.[9] The results of these studies have formed the basis for the International Research Agency on Cancer (IARC) to classify PM as carcinogenic to human beings (Group 1).[10]

The main objective of the present chapter is to describe and analyse the main factors that characterize European primary PM emissions, including: main sources of origin, size distribution and chemical composition (speciation), current emission inventories, trends and regulations, and mitigation measures. Despite having a significant contribution to ambient particle concentrations,[11] secondary PM is not considered in the present chapter. The complexity of the atmospheric aerosol processes and other factors (*e.g.* precursor gases) influencing its formation suggest the need for treating it separately in a more extensive study.

Section 2 of this chapter lists and describes the main anthropogenic and natural emission sources that contribute to total PM emissions in Europe. In Sections 3 and 4 a thorough analysis of the size distribution and speciation of PM emissions is conducted, respectively. Section 5 describes the main European PM emission inventories currently used, while Section 6 performs an analysis of PM trends in Europe. Finally, Section 7 focuses on current regulations and mitigation measures that affect PM emissions.

2 Source Categories

Primary PM is derived from a wide range of sources (both natural and anthropogenic), the contribution of each one varying with the location, season and time of day[12] (Figure 1).

This section introduces and describes the sources that currently present the most significant contributions to European PM emissions.

2.1 Residential Combustion

Recently, interest has grown in biomass combustion as an environmentally friendly way of heating homes whilst at the same time reducing climate change impact and contributing to energy security. In this sense, the use of wood and other biomass in residential small combustion installations has

(a)

(b)
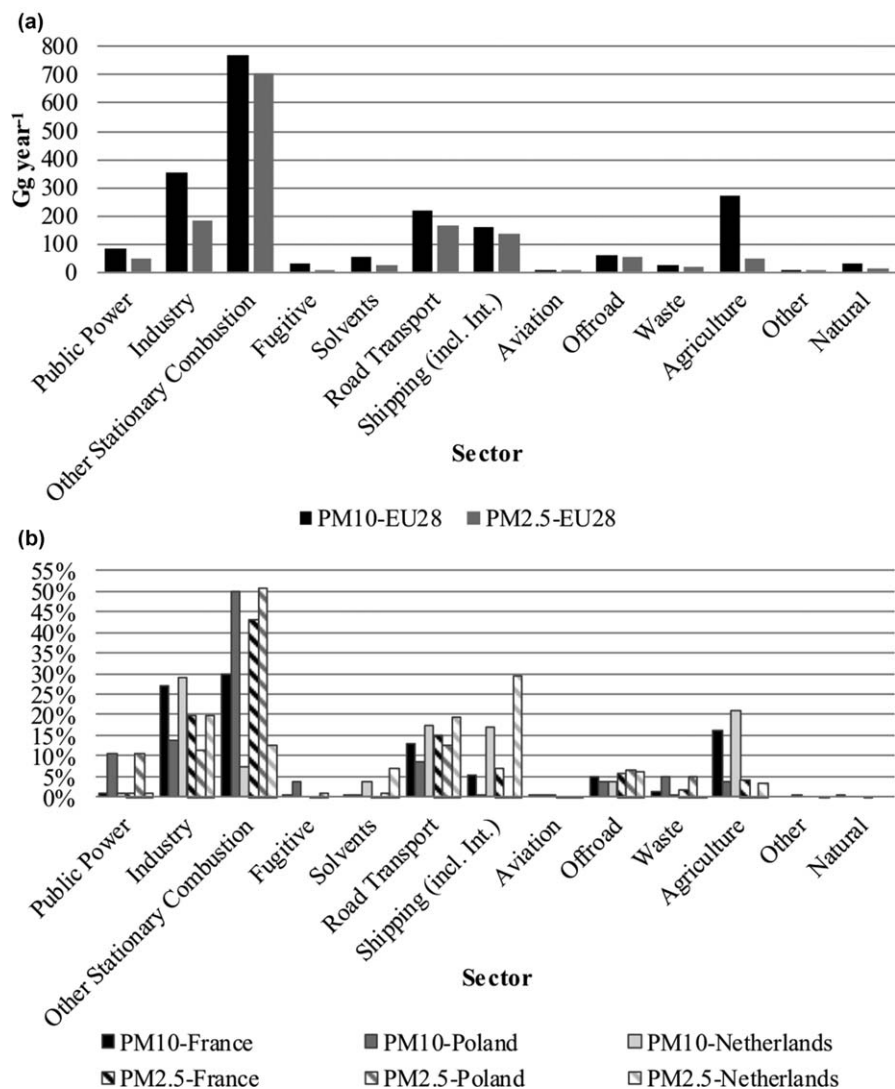
Figure 1 PM$_{10}$ and PM$_{2.5}$ annual emissions (Mg year^{-1}) per pollutant sector in the EU-28 region (a) and contribution (%) of each pollutant sector to total PM$_{10}$ and PM$_{2.5}$ emissions in France, Poland and The Netherlands in the year 2013 (b).[14]

been enhanced by several greenhouse gas strategies and targets for renewable energy. For instance, in 2014 the United Kingdom introduced the Domestic Renewable Heat Incentive (RHI), a financial support programme for renewable heat that offers payments to households for the installation of biomass heating systems to provide central heating and hot water. Moreover, the increase during the economic crisis of other fuel prices typically used in

the residential sector (*e.g.* fuel oil) also triggered the use of biomass, which is less expensive, especially in those countries more affected by the recession, such as Greece.[13]

Despite being labelled as a renewable fuel that can contribute to mitigating climate change, the combustion of biomass in small heating combustion installations is currently a major source of primary PM emissions, especially in wintertime. In 2013, emissions released from small residential combustion appliances were reported as the largest source of PM_{10} (38%) and $PM_{2.5}$ (52%) in the EU-28 region.[14] The high contribution of residential wood combustion (RWC) is mainly owing to the fact that: (i) burning conditions are often inefficient (*i.e.* low combustion temperatures, which leads to incomplete combustion) and (ii) household appliances used for the combustion of biomass usually lack emission controls or regulations.

In each European country, the contribution of RWC towards the total PM_{10} and $PM_{2.5}$ emissions varies depending on its energy balance (*i.e.* relative amount of biomass consumed at the residential level with respect to other fuels) and the type of appliances used and fuels burned. The amount of appliances (*e.g.* fireplace, woodstove, pellet stove, pellet boilers) and biofuels (*e.g.* cork oak, pine, olive pit) used for RWC is very large and their use varies from one country to another. A survey conducted in Portugal reported that the main appliances used for RWC in the country are fireplaces (43%) and woodstoves or traditional ovens (24%).[15] On the other hand, in Finland the biggest portion of wood is burnt in masonry heaters and ovens (38%), log boilers (24%) and sauna stoves (15%), with fireplaces representing only 5% of the total combustion installations used.[16] Masonry heaters and ovens have qualities that allow better burning conditions, higher efficiency and subsequently lower PM emissions than conventional fireplaces. While in 2013 the amount of biomass used in the residential sector was 62% higher in Finland than in Portugal,[17] the amount of Finish $PM_{2.5}$ emissions reported for the same year and sector was just 28% larger than in Portugal.[14]

Several studies have shown that the amount of PM emitted varies widely with category of burning appliance and biomass type.[18–20] One of the most recent studies focusing on this topic was developed under the framework of the AIRUSE LIFE project. Emissions from different biofuels and appliances (those most prevalent in southern Europe) were analysed to obtain a detailed characterisation of emission profiles resulting from RWC. Results from this and previous studies shown that open fireplaces are the appliances that present the highest particulate emission factors (EFs; amount of pollutant emitted per activity unit) owing to low temperatures, which contribute to inefficient combustion. Compared to modern eco-labelled woodstove, $PM_{2.5}$ EF from traditional fireplaces can be up to 10–50 times higher.[18,19] Variations in the PM emissions can also be found within the same type of appliance owing to the operation conditions (*e.g.* air-staging settings and the thermal load).[21] On the other hand, the highest EF are observed for biomass fuels other than pellets (*e.g.* olive pit, shell of pine, nuts, almond shell), the variations being related to the different ash contents of the fuels.[22]

Apart from fuel parameters and operation conditions, the measurement protocol applied is another important factor that influences the variation of EF for the same appliance type. A detailed survey and review of the various RWC EF in use in Europe concluded that the most important type of measurement techniques are filter measurements, which measure only solid particles, and dilution tunnel measurements, which measure solid particles and condensates of semi-volatile organics.[23] The EF compiled by the study presented a high variation as a function of the technique used. For instance, EF for conventional wood stoves obtained with filter measurement ranged from 64 to 87 mg MJ^{-1}, while measurements in dilution tunnels showed results in the range from 340 to 544 mg MJ^{-1}. The choice to use a filter measurement- or dilution tunnel measurement-based EF can have a great impact when estimating PM RWC emissions and analysing the contribution of this source to primary organic aerosols (POA).[24]

2.2   Road Transport

Road transport is one of the main sources of PM in urban areas. In 2013, road transport alone was responsible for 12% and 13% of total primary PM_{10} and $PM_{2.5}$ emissions in the EU-28 region, respectively.[14] Nevertheless, at the city level these contributions can go up to 40–50%, thus constituting the main urban emission source.[25–27]

PM emissions from traffic are categorised according to the mode of formation.[28] The combustion of fuels, mainly gasoline and diesel, in internal combustion engines (exhaust emissions) is generally assumed as the principal mechanism by which PM is formed. On the other hand, road transport also involves the interactions between vehicles and the road surface and the use of brakes, which can result in the release of PM emissions. This category of emissions is known as non-exhaust emissions and includes: (i) tyre wear, (ii) brake wear, (iii) road surface wear and (iv) resuspension. The first three sources involve mechanical abrasion, grinding, crushing and corrosion processes, while the last one refers to the resuspension of the dust collected on the road surface owing to vehicle-generated turbulence.

The quantification of exhaust traffic emissions mainly depends on the engine type, engine age (*i.e.* Euro categories set up by European legislation), after-treatment technology, fuel properties (*e.g.* fuel sulfur content), level of maintenance of the vehicle, environmental conditions and driving conditions.[5] Exhaust emission rates from vehicles can be estimated from controlled conditions in laboratories (*i.e.* engine and chassis dynamometer studies) or real-world conditions (*i.e.* tunnel, remote sensing, on-road and on-board measurements).[29] The use of both approaches indicates that in general PM emission rates from diesel vehicles are significantly higher compared to those from gasoline ones, and that Heavy Duty Diesel Vehicles (HDDVs) are the highest emitters among the different diesel vehicle categories.[30–32] However, increasingly restrictive European diesel emission standards (Section 7) have resulted in a clear reduction of diesel PM emission levels by about

80–90%. In some cases vehicles equipped with diesel particle filters (DPF) (part of Euro 4 and all from Euro 5 and on) even show lower PM levels than gasoline vehicles.[33] The effect of speed on PM exhaust emissions is also increasingly reduced with the introduction of new Euro standards. Generally speaking, low-speed operations lead to higher emission rates. Nevertheless, and as shown by the two reference vehicle emission models in Europe (COPERT; COmputer Programme to calculate Emissions from Road Transport and HBEFA; Handbook of Emission Factors), the shape of emission rates *vs.* speed curves is flatter for the new emission standards than the old ones.[30–32] On the other hand, PM emission rates significantly increase during acceleration as well as with aggressive driving or heavy load conditions.[33]

Non-exhaust emissions are more difficult to quantify than exhaust emissions owing to the strong influence of not only the type of vehicle and traffic conditions, but also the material properties (*e.g.* tyre type, road pavement, grain size) and meteorological factors (*e.g.* temperature, road wetness).[6,34] Non-exhaust particles derived from resuspension processes seem dominant in terms of mass, although this can vary from one country to another owing to the effect of humidity, the use of studded tyres and the contribution from road sanding.[35] Resuspension PM_{10} emission rates estimated by roadside measurements on inner-city urban roads across Europe present a wide variation: UK (14–23 mg VKT^{-1}), Germany (57–109 mg VKT^{-1}), Denmark (46–108 mg VKT^{-1}), Finland (121 mg VKT^{-1}), Sweden (198 mg VKT^{-1}).[36–38] Several campaigns have pointed out a strong correlation between HDDVs and resuspension, the emission rate for this class of vehicle being up to 20 times higher than that for passenger cars.[39] Resuspension emissions in motorways tend to be lower than those in other types of roads (especially urban streets) since higher average vehicle speeds and traffic intensity lead to a lower on-road dust reservoir.[36] The large variation in the resuspension emission rates make them applicable only to the site of study or areas with similar characteristics. During recent years, different numerical approaches have been developed with the intention of reducing the dependency of non-exhaust emission quantification on local measurements.[40] One of the most recent models, the NORTRIP model, is capable of estimating non-exhaust traffic PM emissions based on the impact of surface wetness, the buildup of dust on the road surface, the surface moisture and the effects of applying traction maintenance measures (*e.g.* salting and sanding).[41]

Several studies across Europe have pointed out that the contribution of non-exhaust emissions to PM_{10} can be comparable or even higher than that of exhaust emissions, especially in Scandinavian and Mediterranean countries, owing to studded tyres and road sanding in the former and drier climates in the latter.[42] The contribution of non-exhaust emissions to total PM_{10} in urban areas is expected to grow during the coming years up to approximately 80–90% by 2020.[43] This increase is the result of a combination of several actions that are currently in place to reduce PM emissions from motor exhausts (both at legislative and technological levels) and a lack of abatement measures for non-exhaust emissions.

2.3   Energy and Manufacturing Industries

Emissions from energy (power plants and refineries) and manufacturing industries represent the second-largest source of primary PM_{10} (28%) and $PM_{2.5}$ (21%) in the EU-28 region.[14] However, and with the exception of certain cities close to industrial environments,[44] the contributions of these activities to primary PM in urban areas is less pronounced than that of road transport (around 10%).[25,45]

There are three main mechanisms by which industrial PM is formed. The first involves fuel combustion processes (*e.g.* coal, oil, coke) in conventional boilers, furnaces, gas turbines, reciprocating engines or other combustion devices. PM emissions can also arise from non-combustion processes, such as mechanical treatments of raw materials (non-metallic industries) or casting operations (iron and steel industries). Emissions derived from both combustion and non-combustion processes are usually channelled through ducts (*i.e.* stacks), which makes them more controllable. Finally, industrial PM emissions can also occur during the handling, transport and storage of dusty raw materials (*e.g.* clinker, cement). These emissions, referred to as ''diffuse'', are more complicated to quantify and control than the channeled ones, owing to the difficulties in determining their flux and location of occurrence inside the industrial areas.

PM emissions from combustion processes are mainly characterized by the type of fuel and technology used. Once released into the atmosphere, meteorological parameters (*e.g.* temperature, pressure) also play a key role in their vertical distribution and subsequent transportation.[46] Fuels with significant ash content (*i.e.* coal, oil and coke) have the highest potential to emit primary channeled PM. In the past, the Best Available Techniques (BAT) in large coal-fired power plants have been translated into abatement technologies, such as electrostatic precipitators (EP) or fabric filters (FF), which have allowed a great reduction of PM emissions. As a result, the current emission rate from a fluidised bed boiler ($\geq$300 MW) working with brown coal can be up to 4 times lower than the emissions derived from a gas oil reciprocating engine.[47] Combustion processes related to public electricity and heat production facilities alone presented a contribution of 4% to total PM_{10} and $PM_{2.5}$ primary emissions (in the EU-28 region in 2013.[14] These contributions largely vary from one country to another owing to the different energy generation systems. In Poland, where the production of electricity and heat mainly comes from coal-fired power plants, contribution from the public power sector goes up to 11% (PM_{10} and $PM_{2.5}$), while in France, where the main source of energy is nuclear, public power only accounts for 1% of total PM_{10} and $PM_{2.5}$ (Figure 1).[17]

Non-combustion channeled emissions are mainly associated with non-metallic mineral and iron and steel industries.[47] In the first case, PM emissions largely originate from pre- and after-treatments (*e.g.* milling processes in the cement industry), while in the iron and steel sector emissions are generated in sintering and pelletizing plants as well as in blast

furnaces, used for the production of pig iron, and basic oxygen, open hearth and electric arc furnaces, used for the production of steel. Most of the time these emissions are conducted through stacks and subsequently controlled by efficient filters. Nevertheless, specific industrial processes, such as laser sintering of ceramic tiles, can entail non-controlled particle emissions, which can impact worker exposure.[4]

Cement, steel, ceramic and mining industries, in which bulk materials are usually stored, transported and handled in open air, are the facilities that present more potential for diffuse PM emissions.[48] These types of emissions are not only influenced by the characteristics of the industrial processes but also by meteorological factors (*e.g.* wind speed, precipitation) and material characteristics (*e.g.* raw material moisture content and particle size). The estimation of EF for this source has shown a large variation (from 7 to 400 g PM_{10} t^{-1} product) depending on the type of operation (*e.g.* transport of material on unpaved road) and control measure applied (*e.g.* enclosure and use of bag filters during handling operations).[49]

2.4 Maritime Traffic

Maritime traffic is a key component of the European economy. Compared to other modes of transport (*e.g.* trucks, trains) ship traffic is more fuel-efficient (*i.e.* fuel used per tonne-kilometre). The use of ships increased by more than 20% during the 1995–2012 period (with an average growth rate of 1% per year), and in 2012 the shipping sector was the second most used mode of freight transport in the EU-28 with 1401 billion tonne-kilometres (tkm), right after road transport (1692 billon tkm).[50] According to a recent report by the International Maritime Organization (IMO), it is expected that this form of transport will continue increasing in the future owing to globalization and the increase of global-scale trade.[51]

At the same time, maritime transport is considered an important contributor to primary PM in coastal areas[52] and subsequently to European coastal air quality degradation,[53] especially in the North Sea and the Mediterranean basin. Ship manoeuvring and hoteling operations (ships at berth), which occur in port areas usually located near cities, have been reported to contribute largely to primary PM emissions. In the Greek ports of Piraeus, Santorini, Mykonos, Corfu and Katakolo a total of 94.3 t year^{-1} PM emissions from cruise ships was estimated for the year 2013,[54] 85% of which was related to hoteling operations and 11.5% to the manoeuvring phase. On the other hand, ship hoteling in the port of Rotterdam (the Netherlands) was estimated to generate 248 t year^{-1} of PM_{10} in 2010.[55] At the European level, primary shipping emissions have been reported to influence atmospheric aerosol concentrations in coastal areas within about 1–7% of PM_{10} and 1–20% of $PM_{2.5}$.[53]

PM emissions from maritime traffic are mainly owing to combustion processes that take place in the ship engines. There are three main factors

that control the total amount of emissions released by ships: engine load factor, engine type and fuel type.[52,56,57]

The powering of ships is delivered by their main engines (ME) and auxiliary engines (AE), which present different load factors (from 0 to 100%) depending on the operative profile of the ship. During the cruising and manoeuvring operations, the ME usually presents the highest load factor (50–75% during cruising and 10–30% during manoeuvring), while during the hoteling phase the AE is the main source of emissions (*i.e.* to cover the electricity requirements of the ship) and the MEs are switched off or running at low load (*e.g.* to provide power for pumps to load and unload liquid cargo). The dependence of PM emission factors on engine load may vary from ship to ship. Nevertheless, a recent review reported that at loads lower than 25%, emission rates can be significantly increased (up to 6.5 times).[58]

In terms of type of engine, ships can be equipped with marine diesel engines (slow-speed diesel engines, SSD; medium-speed diesel engines, MSD and high-speed diesel engines, HSD), steam turbines or gas turbines. SSD generate a greater fraction of hydrocarbons (HCs) than MSD and HSD, which may result in an increase in total PM emissions because of the formation of HC aerosols.[59]

The fuels used in maritime transport include marine heavy fuel oil (HFO), marine diesel oil (MDO), marine gasoline oil (MGO) and, more recently, liquefied natural gas (LNG). HFO is a residual product of the oil refinery process and its fuel sulfur content (FSC) can be up to 3.5%, while in the case of MDO/MGO the FSC is around 0.03%. The FSC has a crucial influence on PM emissions as primary sulfate is linearly dependent on it.[51] A review of published data from on-board studies on PM emissions from ships indicated ranges of emission rates between 0.18 and 0.48 g kWh^{-1} for MDO and 0.56 to 2.12 g kWh^{-1} for HFO.[57] Nevertheless, the same review study also indicated that the levels of fine and UF particle emissions are not necessarily reduced by this fuel shift.

PM emissions within port areas are not only produced owing to maritime traffic but also during loading/unloading operations of solid cargoes from ships (*e.g.* clinker, tapioca, phosphate). These operations generate dust that is firstly deposited in the dockside and later resuspended by the effects of port-related traffic or wind. The problematic issue relating to this emission source is similar to that found for diffuse industrial emissions (Section 2.3). In the framework of the LIFE project HADA (Automatic Tool for Environmental Diagnostic), average PM$_{10}$ EF up to 140 ± 30 g min^{-1} were estimated for several operations and types of cargoes in Spanish harbours, which states the relevance of this source to port dust emissions.[60]

2.5 Agricultural Activities

During the last few years, agricultural activities, including fertilizer application, manure management and animal housing, have attracted scientific attention since they are the main European sources of NH$_3$ (their

contribution is around 90%) and subsequently important contributors to secondary PM.[61] Nevertheless, agriculture also presents a notable contribution to primary PM_{10}, with a contribution of up to 14% in the EU-28 emission inventory in 2013.[14] The main activities that contribute to the formation of this pollutant include storage, handling and transport of agricultural products, manure management, agricultural waste burning, land preparation and harvesting.

Just as in the case of traffic resuspension (Section 2.2), emissions from land preparation and harvesting are not regulated by the Convention on Long-range Transboundary Air Pollution (CLRTAP) and are not included in the official emission inventories reported by the Member States (MS). Consequently, studies about the contribution of land management activities are currently scarce and a significant knowledge gap exists. Despite the small amount of available dedicated research, some studies have acknowledged that agricultural land operations (*e.g.* ploughing and harrowing) together with harvesting may create dust plumes, and although much of this dust is rather coarse-sized, significant amounts are carried in suspension over long distances, contributing to the background atmospheric dust load.[62] The contribution of these activities have been estimated to be around 5% of total primary PM_{10} in the EU-27 emission inventory,[63] but it can be more significant in countries and regions characterised by large agricultural regions ,such as The Netherlands.[64] Nevertheless, studies in which emission potentials were estimated for different land management activities present a wide range of values, showing EF variations of a factor of up to 50.[65]

2.6   Natural Sources

Natural sources, which involve no direct or indirect human activity, can present high contributions to total PM emissions. The sources included under this category are: (i) windblown (desert and local) dust, (ii) sea salt aerosols, (iii) volcanoes, (iv) primary biological aerosol particles and (v) wildland fires.[74,77,84]

Windblown dust defines the fugitive dust generated and transported by wind action. This occurs mainly in arid and semi-arid regions, although the process can also occur in surfaces covered by vegetation or man-made covers (*e.g.* roads, buildings). The major sources of dust are located in North Africa, the Saharan sources being considered as the most active ones in the world.[66] Recent estimates of the amount of dust exported annually from North Africa (usually referred to as desert dust or African dust) suggest that 400–2200 Tg year^{-1} is a plausible emission range.[67] A large fraction of the African dust is regularly transported from its source northwards across the Mediterranean to southern Europe,[68] and sometimes as far north as the United Kingdom.[69] Desert dust emissions have a significant impact on the background particle levels in the Mediterranean basin as they are responsible for a significant percentage (up to 70%) of PM_{10} daily level exceedances of the EC standard at background monitoring stations, especially in Spain and during the

summertime.[70] On the other hand, in Europe there are also potentially erodible surfaces (local dust reservoirs) that can generate fugitive dust emissions. According to a study developed under the NatAir European project, the yearly amount of PM_{10} emitted by wind from the European territory is approximately in the range of 0.66–0.88 Tg year^{-1},[7] of which emissions from agricultural areas constitute an estimated 52%. Spain, together with France and Italy, are the European countries where the most important local dust reservoirs are located.[71] In the case of Spain, loamy soils in central Aragon (NE Spain) are often eroded by strong Cierzo winds, reporting observed dust events with vertical flux ranging from 0.4 to 70 µg m^{-2} s^{-1}.[72]

Sea salt aerosols under 10 µm in diameter are the dominant aerosols in marine surface air and can make a significant contribution to land-based PM levels, especially when surface wind speeds are high.[73] At the European level, the annual contribution that sea-salt emission makes to PM_{10} was estimated as 20 Tg year^{-1} for the year 2009,[74] the highest production of sea-salt found on the Atlantic Ocean during winter time, while in the Mediterranean Sea the highest emissions were estimated over the Aegean Sea during summer. A significant part of the variability in the emission estimation comes from the uncertainty associated with the parameterization of the sea-salt emission process, which mainly depends on surface wind speed as well as sea surface temperature, wave height and water salinity, among other parameters.[75]

Primary particles emitted by volcanic eruptions are formed through magma fragmentation and erosion of the vent walls. Volcanic aerosol emissions generally exhibit coarse size distribution and are mainly characterised by their plume height, mass eruption rate and their vertical distribution of mass (with the fine ashes concentred at the top).[76,77] Volcanic ash emission rates vary according to the eruptive style and the intensity and duration of the eruption. During the Eyjafjallajökull eruption, which took place in Iceland between April and May of 2010 and caused significant economic and social disruption in Europe, a total fine ash (diameter 2.8–28 µm) emission of 8 ± 4 Tg was found.[77] European volcanic activity is mainly limited to Iceland and the Mediterranean areas of Greece and Italy.[76,78] Nevertheless, volcanic particles can undergo long-range transport in the atmosphere since they have the potential to produce transient peaks in PM levels not only near the volcano area but also within distances of thousands of km.[79] Besides direct emissions, resuspension and dispersal of freshly deposited volcanic fine ash by wind also have a large impact on PM_{10} levels. Looking again at the example of the 2010 Eyjafjallajökull eruption, PM_{10} concentrations of up to 2000 µg m^{-3} were registered in areas that were never hit directly by the eruptive plume owing to resuspended ash.[80]

Primary biological aerosol particles consist of material that derives from biological processes.[74,81] These types of aerosols are transferred into the atmosphere without any change in their chemical composition and they mainly include pollen, plant debris, fungal spores, bacteria and viruses. At the European level, the contribution of plant debris and fungal spores to

PM_{10} emissions has been estimated at 0.12 Tg year^{-1}.[74] However, there is currently a rather unsophisticated approach applied for the estimation of these emissions. The EF are not directly obtained but derived from a few sets of measured plant debris atmospheric concentrations that are compared to atmospheric concentrations of other compounds, for which the emission fluxes are known.[81] Moreover, emission rates are considered independent of the surface type or vegetation (excluding barren land and water area) and are temporally scaled (3 month periods) using observed seasonal cycles of plant debris and spore mass.[74] Hence, there is a need to better understand the release mechanisms associated with these primary biological aerosols (*e.g.* meteorological patterns that may influence the emission fluxes) and subsequently refine their emission estimates.

Wind-land fires, also referred as wildfires, are caused by burning forests, shrublands, grasslands and other vegetation (excluding agricultural waste burning). For the region of Europe, the global Fire INventory model (FINN) reported a total of 0.39 Tg year^{-1} and 0.22 Tg year^{-1} PM_{10} and $PM_{2.5}$ annual average emissions (2005–2009),[82] while the Global Fire Assimilation System (GFAS) estimated an average of 0.74 Tg year^{-1} and 0.46 Tg year^{-1} for annual PM_{10} and $PM_{2.5}$ emissions (2003–2011).[83] Wildfire emissions are especially relevant in forested Mediterranean countries, such as Spain, Portugal, France, Greece and Italy, where summers are drier and hotter than other European countries. These five southern MS present a combined average of 400 000 hectares of forestland burn every year and are estimated to be responsible for 0.17 Tg year^{-1} $PM_{2.5}$ average annual emissions (2003–2011).[84] Emissions from open vegetation fires basically depend on the land area burnt, the type of vegetation (*i.e.* fuel material), the amount of organic matter available, the properties and condition of the fuel material (*e.g.* dry, wet, decayed), and the combustion stage (*i.e.* flaming, smouldering).[82–84] Several laboratory studies and field campaigns indicate a wide variation in the emission factors associated with specific fuel types, most of them confirming that PM_{10} mass is dominated by $PM_{2.5}$ mass concentration.[85] As in the case of volcano emissions, the injection height of wildfire emissions is a critical parameter in the transport of the particles released to the atmosphere. Several factors, such as the energy released from the fire, fuel type and local meteorological conditions, determine the plume height, which can reach altitudes of up to 6.1–8.7 km above the surface.[86]

3   Particle Size Distribution

As previously stated, PM comes in a wide range of sizes according to its aerodynamic diameter, including: coarse particles ($PM_{2.5-10}$; diameter between 10 μm and 2.5 μm), fine particles ($PM_{2.5}$; diameter less than or equal to 2.5 μm), ultrafine particles (UFP) ($PM_{0.1}$; diameter less than or equal to 0.1 μm) and nanoparticles ($PM_{0.05}$; diameter less than or equal to 0.05 μm). The size of PM is directly linked to its potential for causing health problems since smaller particles penetrate further down the respiratory tract and even

transfer to extrapulmonary organs, including the central nervous system.[87] While most severe adverse health effects have been typically associated with $PM_{2.5}$, other epidemiological studies suggest that PM_1 may have a greater potential for adverse health impacts.[88] The relative amounts of particles present in each size are expressed by mass concentration in the case of $PM_{2.5-10}$ and $PM_{2.5}$ and by number concentration (PNC) in the case of aerosols with diameters between 0.1 and 0.05 µm owing to their negligible mass.

Coarse particles are usually associated with mechanical disruption processes (*e.g.* crushing, grinding, and abrasion of surfaces) and the suspension of dust. Traffic non-exhaust emissions (wear processes and resuspension) are assumed to be dominated by the $PM_{2.5-10}$ fraction,[38] although in some cases particles in the fine particle range have also been found (approximately 15%).[89] Similarly, emissions derived from agricultural activities are mainly associated with the coarse size[62] as well as the diffuse emissions related to handling, transport and storage of dusty raw materials.[60] Regarding sea salt aerosols, approximately 95% of their total mass is in the coarse mode,[90] although in Atlantic zones its contribution to $PM_{2.5}$ can be up to 11%.[11] $PM_{2.5-10}$ tends to have a local impact (1 to 10s of km) and to settle on the ground through dry deposition processes (*e.g.* gravitational sedimentation) in a matter of hours. This is not the case for coarse particles related to wind-blown desert dust, which can be transported over thousands of km (Section 2.6).

Primary $PM_{2.5}$, UFP and nanoparticles are mainly formed from combustion and high-temperature processes, and industrial operations. Road transport, in particular diesel engines, is the major source of primary $PM_{0.1}$ and $PM_{0.05}$ emissions in urban environments,[91,92] with reported contributions of up to 97% of the total PNC.[93] Many of the PM produced by RWC as well as maritime traffic is also below 1 µm.[22,56] On the other hand, primary UFP and nanoparticle emissions from industrial processes such as tile sintering and laser ablation operations are also receiving increasing attention.[4] As opposed to coarse particles, PM in the accumulation mode (diameter between 0.1 and 2.5 µm) tend to have longer lifetimes (days to weeks) as they settle slowly and have low diffusivities, their travel distance being up to thousands of km.[3] On the other hand, UFP usually present lifetimes that go from minutes to hours owing to their tendency towards growth into the accumulation mode.

According to European official reported emissions in the year 2013, 32% of total primary PM_{10} emissions are considered to be in the $PM_{2.5-10}$ fraction and 68% in the $PM_{2.5}$ fraction.[14] In the coarse fraction, agricultural activities are the ones that present the largest contribution (36%), together with mining and construction activities (10%) and non-exhaust traffic emissions (9%). On the other hand, the fine fraction is mainly dominated by residential combustion (58%), energy and manufacturing industries (21%), and road transport (13%). Regarding UFP emissions, they can be indirectly obtained from primary particle number (PN) emission inventories (expressed as

numbers of particles instead of mass) since PN emissions are dominated by UFP emissions and the difference between them is relatively small.[92] Recent primary PN emission inventories for Europe indicate a significant contribution from traffic as well as shipping emissions, especially in coastal urban areas such as Oslo, with total shares of 75% and 15%, respectively.[91] France, Spain, Germany, Italy, UK and Poland are reported as the major PN emitters in the EU-28 region, the sum of their traffic emissions representing approximately 72% of the total PN road transport emissions in EU-28.[93]

4   Speciation

Primary PM includes as principal components organic carbon (OC), black carbon (BC), trace metals, crustal material (*i.e.* soil particles), sea-salt and, to a lesser extent, sulfates (SO_4^{-2}). The chemical makeup of PM varies across Europe, depending on the emission source categories that characterize the region of study.

Carbonaceous particles (BC and OC) are generally the dominant components of primary PM emissions. The carbon fraction of PM is identified as having significant impacts on health, climate change, atmospheric photochemistry and aerosol–cloud interactions.[94] Primary BC and OC are mainly formed by incomplete combustion processes and are predominantly present in the fine and UF particle fractions.[67,92,95,96] BC is sometimes also defined using other terms, such as elemental carbon (EC), soot or graphitic carbon. While all the terms are used to denote light-absorbing carbon in atmospheric aerosol particles, each one of them identifies the specific instrument or measurement technique used to measure the quantity of the component. For instance, BC and EC are often used to indicate optical and thermal measurement methods, respectively. In this chapter, the term BC is generically used.

According to the EDGAR-HTAP_v2 global emissions inventory, the total amounts of primary anthropogenic (land-based, excluding ship emissions) BC and OC emissions released in Europe during 2010 were 0.38 Tg year^{-1} and 0.64 Tg year^{-1}, respectively, since the transport and residential sector is responsible for around 90% of the total emissions.[97] On the other hand, European official BC anthropogenic emissions report a total of 0.14 Tg year^{-1} released in the EU-28 region during 2013, the largest contributions being those of the residential (34%), traffic (32%), and national and international shipping (6%) sectors.[14] The contribution of emission sources to particulate carbonaceous emissions may vary according to PM size. The size-resolved emission inventory of carbonaceous particles addressed in the EUCAARI project indicates that the emission of OC in the fine fraction is dominated by the residential combustion of wood and coal, while the largest sources of EC in the UFP fraction are diesel transport and residential combustion.[95] On the other hand, and according to the TNO_MACC_II emission inventory, the most important sources of coarse OC and coarse BC in terms of total mass are not the transport and residential sectors but rather the

agriculture (agricultural waste burning) and the power plants and industry sectors, respectively.[96]

Emission ratios of BC and OC to PM are critical to determine since they vary according to a large number of parameters, including fuel type, technology, combustion process efficiency, emission control and size of particles. Diesel engines are estimated as the largest contributors to primary BC emissions,[26,95] while gasoline engines are known to release a higher fraction of OC.[98] Nevertheless, some studies have pointed out that gasoline vehicle UFP emissions are dominated by the BC fraction.[99] In cases where advanced after-treatments are used (*e.g.* DPFs), a significant reduction of the BC fraction is also observed.[100] In the case of biomass combustion, OC generally dominates the PM emissions in small traditional appliances (*e.g.* fireplaces), while more efficient combustion installations show larger EC relative fractions owing to higher combustion temperatures and flaming combustion.[18] Fuel properties also influence the OC and BC contents in the particles emitted, with higher hydrocarbon (HC) emission rates contributing to higher OC contents.[20] The OC fraction released from biomass burning provides an important contribution to benzo[*a*]pyrene (B*a*P),[101] a polycyclic aromatic hydrocarbon (PAH) reported by the IARC as a probable carcinogen in humans.[102] In the case of maritime traffic, the OC fraction is typically larger than the BC one.[56] PM ship emissions of OC increase with the fuel sulfur content, whereas BC appears to have a significant dependence on the engine load and engine settings but not on the FSC.[56–58] SSD engines are found to generate a greater amount of OC fraction since they typically have a larger fraction of HCs coming through the engine.[58]

Crustal material includes soil particles generated by wind erosion processes (including desert dust contributions), traffic resuspension, handling, transport and storage of materials, and agricultural land management activities, among others. The main components that can be associated with crustal material include aluminium (Al), silicium (Si), calcium (Ca) and iron (Fe), which are usually associated with the coarse fraction ($PM_{2.5-10}$).[103] In Europe, soil particle emissions typically represent 5 to 20% of the ambient PM_{10} mass;[104] the contribution is higher in south-western and south-eastern Europe owing to the warmer and drier climate and the higher influence of African dust intrusions.

Tyre and brake wear emissions (as well as resuspension) are a major source of metals in urban areas.[28,103] In terms of heavy metals, brake wear is the most important source of emissions for copper (Cu), while for tyre wear the most important emission is zinc (Zn).[35] Tyre and brake wear sources represented 77% and 33% of total Cu and Zn emissions in the EU-28 region in 2013, respectively.[14] These two emission sources together also significantly contribute to total lead (Pb) emissions (10%), which in the past was dominated by gasoline exhaust emissions until the phasing out of leaded fuels in Europe.[105] Tyre and brake emissions also include other trace metals, such as arsenic (As), nickel (Ni), antimony (Sb), iron (Fe) and barium (Ba),

the composition presenting a large variability across Europe owing to the dependence on the manufacturer and brand.[28,42] Outside urban areas, the metal concentrations of PM may partly originate from different sources such as energy and manufacturing combustion or industrial processes. Heavy metals are most abundant in high-temperature metal processing activities,[2,103] and hence the production of iron and steel is a key contributor to total European emissions of Pb, cadmium (Cd), mercury (Hg), chromium (Cr) and Zn.[14]

Primary sulfate aerosols usually present a residual fraction of total PM emissions (between 2 and 4%).[27,96] Nevertheless, sulfur emitted in the form of particles is important in combustion processes of high-sulfur fuels, which mainly occur in energy and manufacturing industries and in shipping. The fraction of primary sulfate to total PM is mainly influenced by the FSC of the fuel consumed; during coal combustion it has been reported to range from 10 to 45%,[106] while in the case of marine residual oil combustion (*i.e.* HFO) it can account for up to 80% of the weight of the emitted particles.[51,56] Sulfate is becoming less and less significant as a primary PM component owing to the general tendency to substitute coal for natural gas in the public power sector and HFO for MDO and LNG in the maritime sector (Section 7.4).

4.1 PM Speciation Source Profiles

PM speciation source profiles indicate the chemical species that comprise the PM emissions released from a specific source. These speciation profiles, commonly expressed as the mass ratio of each species to the total PM, are used to characterize the different components that are associated with individual pollutant sources. Currently there are different repositories of PM speciation source profiles freely accessible, with the objective of being used for different purposes, such as creating speciated PM emission inventories for photochemical air quality modelling[27] or providing input to the Chemical Mass Balance (CMB) receptor models (RM).[101] One of the best-known repositories is the United States EPA SPECIATE database, which has been publicly available since 1988, and it currently contains around 3000 entries.[107] Source profiles from this American repository are usually used in European emission and air quality modelling exercises owing to the scarcity of official and well-established European databases. With the objective of filling this gap, a new database of PM speciation source profiles in Europe has been recently developed (SPECIEUROPE).[2]

The SPECIEUROPE is a repository developed in the framework of the Forum for Air quality Modelling in Europe (FAIRMODE) that contains the chemical composition of PM emission sources reported in European scientific papers and official reports. Currently, SPECIEUROPE consists of 209 PM speciation profiles, combining measured, composite, calculated (from stoichiometric composition) and derived (results of source apportionment studies) profiles.

5   European PM Emission Inventories

Emission inventories are datasets used to estimate the amount of air pollutants being emitted to the atmosphere, caused by an anthropogenic or natural activity, at a certain geographical location for a given period of time. Emission inventories are generally recognized as key inputs to atmospheric modelling, especially when they are used to design effective control measures to mitigate the adverse impact of air pollution.[45] Statistical methods of source apportionment to indirectly assess pollutant sources from measurements have also proved the requirement of emission inventories as input data.[103] Therefore, during recent years a significant amount of emission datasets have been developed either for scientific or regulatory purposes.[108]

At the European level, the most used inventories to determine PM emissions and its impacts on air quality are: the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP),[109] the Emission Database for Global Atmospheric Research (EDGARv4.2),[110] the EDGAR-HTAP_v2,[97] the EMEP emission inventory,[111] the TNO-MACC-II emission inventory[96] and the Greenhouse gas and Air pollution INteractions and Synergies (GAINS) model.[112] Each of them presents different emission estimation methodologies, spatial resolutions, temporal coverages and applications (Table 2).

All of these European inventories focus their attention on the PM_{10} and $PM_{2.5}$ fractions (carbonaceous components included in some cases) giving no particular attention to UFP. This is a consequence of the fact that current European legislation on primary PM emissions is based on particle mass and not on particle number. However, the increasing evidence of the adverse health impacts related to UFP has also increased the attention on PN emission inventories. Numerous research studies and European projects, such as PARTICULATES or TRANSPHORM, have consolidated emission factor databases for constructing PN emission inventories in Europe. As a consequence, during recent years some of the aforementioned emission inventories have been revised in order to include PN emission estimations.[91,93,95] Nevertheless, the estimation of PN emissions is associated with a higher uncertainty than that linked with PM_{10} and $PM_{2.5}$ emission estimations. For instance, while the uncertainty of PM emissions from traffic sources has been reported to be between 10 and 20%,[113] the overall uncertainty of vehicular PN emissions can be up to 144–169% when aftertreatment device effects are included.[91] This increase of the uncertainty is mainly related to the set-ups of the measurements that define PN vehicle emission factors, including: (i) the consideration or not of volatile PN and (ii) the definition of the lower size cut-off used in the measurement. Considering that traffic is the most intensively studied source category for PN emissions, similar or higher uncertainty values can be assumed for other pollutant sectors.

Despite being well established and showing important improvements, European inventories are still not able to characterize primary PM emissions

Table 2 Summary of European emission inventories currently used in the scientific community for scientific and regulatory purposes.

Name	Source	Emission sources	Pollutants	Temporal resolution/ coverage	Spatial resolution/ coverage	Use	Approach used
ACCMIP	109	Anthropogenic biomass burning	SO_2, NO_x, CO, NMVOC, CH_4, NH_3, BC and OC	Decadal, 1850–2000	$0.5° \times 0.5°$, Global	Scientific	Combination of other inventories (RETRO, GAINS, EMEP)
EDGARv4.2	110	Anthropogenic biomass burning	SO_2, NO_x, CO, NMVOC, CH_4, NH_3, PM_{10}	Annual, 1970–2008	$0.1° \times 0.1°$, Global	Regulatory scientific	Combination of national AF with specific EF, disaggregated using different spatial proxies
EDGAR-HTAP_v2	97	Anthropogenic biomass burning	SO_2, NO_x, CO, NMVOC, CH_4, NH_3, PM_{10}, $PM_{2.5}$, BC and OC	Annual, 2008–2010	$0.1° \times 0.1°$, Global	Regulatory scientific	Compilation of different regional gridded inventories with EDGAR v4.2 spatial proxies
EMEP	111	Anthropogenic	SO_2, NO_x, CO, NMVOC, CH_4, NH_3, $PM_{2.5-10}$, $PM_{2.5}$	Annual, 1980–2013	$0.1° \times 0.1°$, European	Regulatory scientific	National emission inventories reported by parties and assigned to the EMEP grid
TNO-MACC-II	96	Anthropogenic	SO_2, NO_x, CO, NMVOC, CH_4, NH_3, PM_{10} and $PM_{2.5}$ (broken down into EC, OC, SO_4^{-2}, Na and other minerals)	Annual, 2003–2009	$1/8° \times 1/16°$, European	Scientific	Downscaling of National emission inventories through the use of specific spatial proxies
GAINS	112	Anthropogenic	SO_2, NO_x, CO, NMVOC, CH_4, NH_3, PM_{10}, $PM_{2.5}$, $PM_{0.1}$	Annual, 1990–2030	50 km $\times$ 50 km, European	Regulatory	Combination of national AF with specific EF and grid maps

to a satisfying level of detail. Reported emissions often present data gaps, missing sources and high uncertainties for the applied emission factors,[114] which entail high discrepancies between the different emission datasets.[115] This fact is especially relevant for fugitive emissions related to industrial and agricultural activities, where a problem of data gaps exists. On the other hand, the non-inclusion of key sources, such as traffic resuspension, in the national emission inventories reported under the CLRTAP, which are later used in well-established emission inventories such as EMEP or TNO_MACC-II, also entails large uncertainties.

Comparisons between European emission inventories with local emission inventories developed at the regional or urban scale have also pointed out significant discrepancies, especially in terms of allocation and total amount of PM residential biomass emissions.[116,117] These differences mainly come from the fact that emission inventories at European or national levels usually tend to rely to a larger degree on top-down approaches, while emission inventories developed for local and urban applications rely to a larger degree on bottom-up approaches. Both methods require information concerning activity factors (*e.g.* total amount of fuel consumed) and emission factors per activity (*e.g.* amount of pollutant emitted per activity unit). Nevertheless, emissions compiled through a bottom-up approach are based on specific information for each sector, such as housing units or number of vehicles per road link for domestic heating and traffic emissions, respectively. Alternately, top-down approaches are based on the disaggregation of variables defined at the regional or national level (*e.g.* fuel sold or consumed) in smaller areas based on auxiliary spatial surrogates that represent the activity (*e.g.* population density for wood burning emissions), thus achieving a higher spatial detail. Bottom-up approaches allow high spatial and temporal detail, although they also require a greater amount of data and thus more resources.

6   Long-term Trends in Europe

According to the European Union emission inventory report under the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP),[14] total emissions of primary PM_{10} have reduced by 19% across the EU-28 region between 2000 and 2013, driven by an 18% reduction in emissions of $PM_{2.5}$. On the other hand, BC emissions have seen a reduction of 35% over the same period.

The difference between the BC trend and that of PM_{10} and $PM_{2.5}$ is owing to significantly decreasing emissions in BC from road and off-road transport since 2000 (a decrease of 50% and 60%, respectively). The majority of the reduction in PM_{10} and $PM_{2.5}$ emissions has taken place in the anthropogenic sectors of public power (20% and 13%), industry (35% and 27%) and road transport (25% and 34%) owing to: (i) a fuel-switching from coal to natural gas for electricity generation, (ii) an introduction of after-treatment technologies in new vehicles, such as DPF (driven by the legislative Euro

standards) and (iii) an implementation of BATs in the industrial sector, in-
cluding improvements in the performance of pollution abatement equip-
ment. Moreover, a marked decrease has been recorded since 2008 in the
hardest-hit countries by the economic crisis (*i.e.* Italy, Portugal and Spain).
The influence of the economic recession on PM has also been reported by
different European interpretation trend studies.[118]

During 2013 the contribution of residential combustion to total PM
emissions significantly increased in comparison to 2000 (by 13% in PM_{10}
and 17% in both $PM_{2.5}$ and BC) and it is the only sector in which emissions
have risen between 2000 and 2013 (by 11% for PM_{10}, 13% for $PM_{2.5}$ and 12%
for BC) (Figure 2). This evolution can partly be explained by the increase of
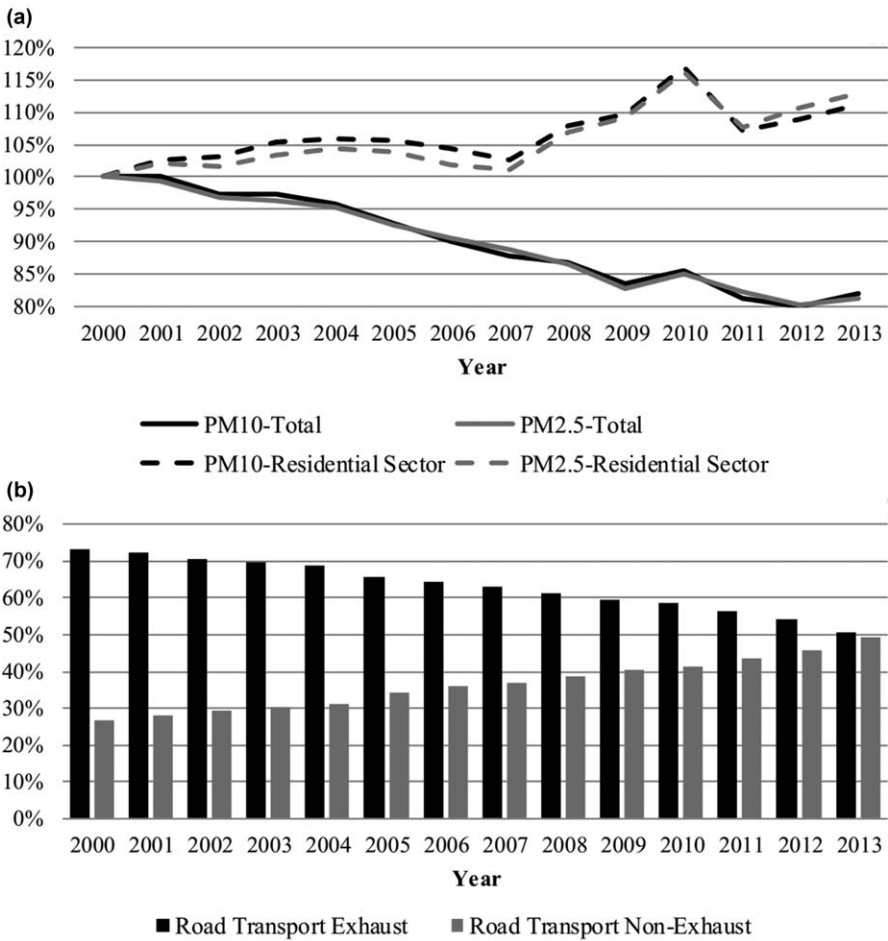
Figure 2   Trend of PM_{10} and $PM_{2.5}$ emissions (%) from total pollutant sources and
residential combustion sources (a) and trend of the contribution (%) of
exhaust and non-exhaust emissions in total road transport PM_{10} emission
in the EU-28 region (b).[14]

biomass burning at the residential level, especially in Eastern European countries (see section below). Moreover, during this period, European efforts have been especially focused on exhaust diesel PM emissions control strategies, which has caused an important decrease in the exhaust road transport's contribution to total BC (from 43% to 33%). The decrease of traffic PM exhaust emissions has increased the importance of non-exhaust emissions in the coarse fraction; the relative contribution from non-exhaust emissions in road transport has increased from 27% to 49% for PM_{10} from 2000 to 2013 (Figure 2). For these results it is important to note that traffic resuspension emissions are not included in the official MS inventories and subsequently road transport non-exhaust contributions may be underestimated.

Looking at the variations between countries, the largest reductions of PM_{10} for 2000–2013 have been reported by Cyprus (66%), France (35%) and Hungary (35%). In the case of $PM_{2.5}$, Cyprus and France are also among those countries that have shown the greatest reduction (73% and 42%, respectively) together with The Netherlands (50%). BC emission reductions are led by The Netherlands (61%), the United Kingdom (53%) and France (44%). The large reduction of BC observed in the case of The Netherlands is partly explained by the increase of the market share of hybrid-electric vehicles during this period (9.7%), the largest of the whole EU-28 region.[119] However, despite all the reductions observed, France was the MS with the largest contribution to total PM_{10}, $PM_{2.5}$ and BC emissions in 2013 (14.4, 14.2 and 26.7%, respectively), which was also the case back in 2000. These results can be explained by the patterns observed in the road transport and residential combustion sectors: (i) diesel dominated the French passenger car market with a 66% of the total share[119]—the EU-28 share was 53% in 2013—and (ii) biomass was, after natural gas, the fuel most used in French households (28%) and represented 19% of the total biomass consumed in the EU-28 region.[17]

In contrast to the aforementioned countries, PM emissions have increased in some countries since 2000; the greatest increases have been reported by far in Romania for all PM emissions (PM_{10}, 21%; $PM_{2.5}$, 31% and BC, 43%). The explanation for this lies in the fact that the use of biofuels and waste in the residential combustion sector increased by 27% during this period.[17] With this increase, Romania rose from the 7th to 4th position in the list of top contributors to $PM_{2.5}$ and BC emissions in EU-28 (just after Italy). Similarly, Bulgaria has also increased its levels of PM emissions during the same period owing to a rise in residential biomass consumption (53%). Although the emissions have dropped by 11%, Poland is another Eastern European country in which residential emissions have a significant impact (50% of total $PM_{2.5}$). In this case, the main fuel consumed is coal, which represented 68% of the total coal consumed at the residential level in the EU-28 region in 2013.[17]

Emissions of primary PM_{10}, $PM_{2.5}$ and BC are expected to decrease across the EU-28 region in the coming years as vehicle technologies are further improved and stationary fuel combustion emissions are controlled through abatement techniques or the use of low-sulfur fuels (natural gas). However, it