Chapter 3
Anthropogenic Sources of Nanoparticles

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Abstract This chapter deals with anthropogenic sources of nanoparticles in atmosphere. The stationary and diffuse sources are described first: mining and extraction of fossil fuels, ferrous and non-ferrous ores and other mineral resources. Brief references are then made to secondary stationary sources of particulate matter which originate in thermal energy and power generation, ferrous and non-ferrous metallurgy, chemical industry and construction materials (cement, lime) production technologies. A separate section is dedicated to particulate matter resulted from municipal, toxic and hazardous waste incineration and its effect on the environment and human health. The authors also speak about emissions from point sources of nanoparticles, such as the residential heating sector that makes a major contribution to the total amount of nanoparticles produced by anthropogenic activities. Mobile nanoparticle sources include engines that are most frequently used in the present: plane engines, rocket engines, engines for road and water transport. Particulate matter emissions can be reduced by using alternative fuels or other forms of propulsion energy.

3.1 Introduction

Most human activities that take place in open air or produce emissions in the environment generate, directly or indirectly, fine powders lifted by air currents.

As it has already been shown [1], the major fraction of particulate matter (PM) floating in the atmosphere comes from space and it does not have a definite origin that astrophysicists and astrogeologists can explain [2]. Nevertheless, nanoparticles of anthropogenic origin are definitely among the most harmful particulates, not only to human and animal health, but also to the environment.

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And if the impact of natural particulates has determined the specific adaptation of living organisms along the time, most anthropogenic nanoparticles were generated in the nineteenth and twentieth centuries, when large amounts of countless harmful and change-resistant particulates were released in the atmosphere. Among these are cancer-causing, mutagenic and teratogenic chlorinated pesticides [3] that were largely used in agriculture until recently, heavy metal oxide PM of heavy metals, PM from the synthesis or chemical purification of various chemical compounds, fuel combustion or deliberate burning of vegetation for deforestation purposes in underdeveloped countries.

None of the above-mentioned categories has been the subject of long-term studies about their impact on health and the environment. The effects of nanoparticles on the living kingdom have been analysed only recently.

One cannot ignore the unprecedented development of scientific research into nanoapplications through the synthesis, characterisation and industrial production of such materials [4–9] which in turn generate various waste types in a similar state of matter.

Anthropogenic sources of nanoparticles are classified as primary, represented by ore exploitation, and secondary—PM resulted from industrial activities and energy production and transport activities.

Secondary sources can be stationary or mobile. The former are thermal power, chemical, milling, metallurgic or other industrial plants that are continuous and diffuse and, unlike the latter, produce and release large amounts of PM. The latter are mostly terrestrial, marine and air vehicle engines and rockets launched to the extra-atmospheric space. These are considered discontinuous point sources.

As far as global anthropogenic emissions are concerned, the amounts of nanoparticles released by stationary sources are equal or less than those released by engines, since the latter total the daily equivalents which account for 40 % of global pollution [10].

Table 3.1 is a synthetic presentation of several Australian anthropogenic sources of nanoparticles [11].

3.2   Primary Nanoparticle Sources

Depending on the materials extracted from quarries or by underground mining, PM concentration released during technological processes can have characteristics similar to those of natural nanoparticle emissions, as it results from the exploitation of mineral reserves accumulated in time, through orogenesis [12].

Consequently, the origin of nanoparticles is the direct exploitation of rocks by surface mining, evacuation through mine shafts or waste heaps produced either directly, by the separation of the useful fraction, or indirectly, through decantation, sedimentation or flotation, following the concentration of the useful fraction in ores. In either case, waste heaps comprise millions of PM which are more or less stable when rising in the atmosphere.

Table 3.1 A synthesis of sources of nanoparticles resulted from anthropogenic activities in several representative Australian areas [11]

Source Group	Source Type	Particulate matter <10 μm		
		Annual emissions (tonnes/year)	Proportion of annual anthropogenic emissions (%)	Cumulative (%)
GMR				
Domestic-Commercial	Solid fuel combustion	5,986	19.6	19.6
Off-Road Mobile	Industrial off-road vehicles and equipment	5,191	17.0	36.6
Industrial	Coal mining	4,154	13.6	50.3
Industrial	Generation of electrical power from coal	1,708	5,6	55.9
Industrial	Primary iron and steel production	1,444	4.7	60.6
On-Road Mobile	Exhaust emissions light duty—diesel	1,073	3.5	64.1
On-Road Mobile	Exhaust emissions heavy duty commercial—diesel	1,002	3.3	67.4
On-Road Mobile	Exhaust emissions passengers cars petrol	972	3.2	70.6
Industrial	Crushing, grinding or separating works	918	3.0	73.6
Industrial	Other land-based extraction	799	2.6	76.2
All	Other	7,253	23.8	100
Sydney				
Domestic-Commercial	Solid fuel combustion	4,503	34.3	34.3
Off-Road Mobile	Industrial off-road vehicles	1,152	8.8	43.1
On-Road Mobile	Exhaust emissions light duty—diesel	840	6.4	49.5
Industrial	Crushing, grinding or separating works	807	6.2	55.6
On-Road Mobile	Exhaust emissions passengers cars petrol	797	6.1	61.7
On-Road Mobile	Exhaust emissions heavy commercial duty—diesel	681	5.2	66.9
Industrial	Ceramics production (excluding gas)	606	4.6	71.5
Industrial	Other land-based extraction	418	3.2	74.7
Commercial	Poultry farming (meat)	237	1.8	76.5

Table 3.1 (continued)

Source Group	Source Type	Particulate matter <10 μm		
		Annual emissions (tonnes/year)	Proportion of annual anthropogenic emissions (%)	Cumulative (%)
Industrial	Petroleum refining	237	1.8	78.3
All	Other	2,848	21.7	100
Newcastle				
Domestic-Commercial	Solid fuel combustion	381	22.6	22.6
Industrial	Production of ammonium nitrate	207	12.3	34.8
Off-Road Mobile	Industrial off-road vehicles and equipment	125	7.4	42.2
Industrial	Bitumen pre-mix or hotmix production	114	6.8	49.0
Industrial	Primary aluminium production	111	6.6	55.6
Off-Road Mobile	Commercial ships	80	4.7	60.3
Industrial	Production of phosphate fertiliser	78	4.6	64.9
Industrial	Coal mining	71	4.2	69.1
On-Road Mobile	Exhaust emissions light duty—diesel	60	3.6	72.6
On-Road Mobile	Exhaust emissions heavy duty commercial—diesel	52	3.1	75.7
All	Other	409	24.3	100
Wollongong				
Industrial	Primary iron and steel production	1,442	65.8	65.8
Domestic-Commercial	Solid fuel combustion	248	11.3	77.1
Off-Road Mobile	Industrial off-road vehicles and equipment	187	8.5	85.6
On-Road Mobile	Exhaust emissions heavy duty commercial—diesel	40	1.9	87.5
On-Road Mobile	Exhaust emissions light duty—diesel	36	1.7	89.1
Industrial	Coke production	33	1.5	90.6
On-Road Mobile	Exhaust emissions passenger cars petrol	32	1.4	92.1
Industrial	Coal loading	19	0.9	93.0

Table 3.1 (continued)

Source Group	Source Type	Particulate matter <10 μm		
		Annual emissions (tonnes/year)	Proportion of annual anthropogenic emissions (%)	Cumulative (%)
Off-Road Mobile	Commercial ships	14	0.7	93.6
Industrial	Coal mining	14	0.6	94.2
All	Other	126	5.8	100.0
Non-Urban				
Industrial	Coal mining	4,056	30.1	30.1
Off-Road Mobile	Industrial off-road vehicles and equipment	3,727	27.6	57.7
Industrial	Generation of electrical power from coal	1,708	12.7	70.3
Domestic-Commercial	Solid fuel combustion	855	6.3	76.7
Industrial	Other land-based extraction	368	2.7	79.4
Industrial	Hard-rock gravel quarrying	360	2.7	82.1
Commercial	Poultry farming (meat)	272	2.0	84.1
Off-Road Mobile	Commercial off-road vehicles and equipment	261	1.9	86.0
On-Road Mobile	Exhaust emissions heavy duty commercial—diesel	228	1.7	87.7
Industrial	Primary aluminium production	202	1.5	89.2
All	Other	1,456	10.8	100.0

On the long run, countries rich in mineral deposits (see Fig. 3.1) accumulate the largest number of waste heaps and consequently release the largest amounts of nanoparticles in the atmosphere. The most vulnerable are the underdeveloped countries where ore is extracted with obsolete technology and old tools [13].

3.2.1 Fossil Fuel Exploitation

Regardless of their nature, fossil fuels can be exploited on the surface, in quarries or through underground extraction. High carbon concentrations (anthracite, pit coal) in fossil deposits require exploitation at considerable depth [14, 15].

Fig. 3.1 Countries with the most important mineral resources in Europe and Central Eurasia [13]

Coal extraction from quarries, a method that is almost generalised at the moment, is presented intuitively in Fig. 3.2.

When the soil has been stripped and the deposit uncovered (1), coal is excavated and transported either with large vehicles or via conveyor belts. Steps (2) and (3) represent different stages of quarry exhaustion. This technique, though very productive, is also very polluting, especially because of slowly sedimenting PM.

Generally, studies about toxic agents and PM released during surface mining [16] deal with the PM issue superficially; the concentration of micropollutants under the form of nanoparticles is not specified directly, but with global indicators of the TSP type (total suspended particulates), which are irrelevant for the subject of this book [17–19].

Recent studies [19, 20] give more specific details about toxic agent determinations in coal plants during daytime, using more precise indicators such as PM_{10} or $PM_{2.5}$.

Figure 3.3 indicates that in the air samples collected within the perimeter of the two quarries, the highest concentrations are those of the slowly sedimenting particles represented by PM_{10}, or $PM_{2.5}$ which seem to follow the same specific law of concentration variation, while coarser particles sediment immediately, so their concentration is less relevant.

As far as the $PM_{2.5}$ variation is concerned, daily concentrations recorded in the studied period in several places around the coal exploitations vary between 4.5 and 10.2 $\mu g/Nm^3$, while the average value of the whole period was about 7 $\mu g/Nm^3$.

The toxic gaseous agents found in the deposits (methane, nitrogen oxides, hydrogen sulphide, etc.) must be mentioned among the sources of nanoparticles reformed in the atmosphere. When released, these agents cause degradation through oxidation-reduction, hydrolysis, and neutralisation; in other words, they generate nanoparticles with a different chemical nature than that of the initial pollutants [18–20] (Table 3.2).

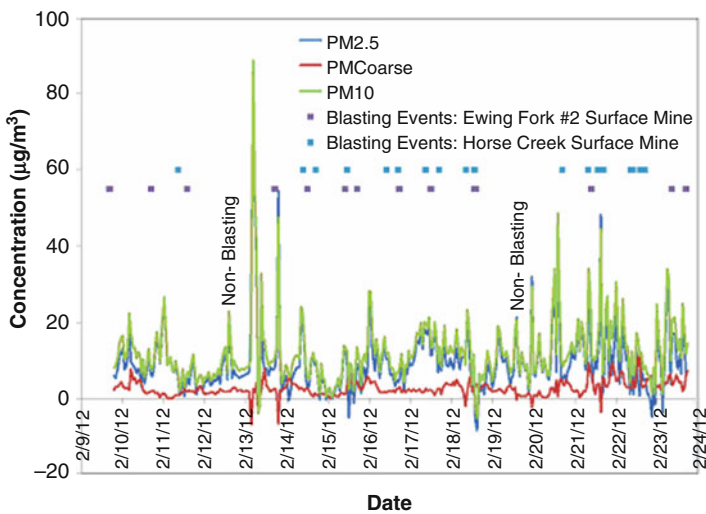**Fig. 3.2** Open-pit coal mining at Greenhills schematic [16]

Fig. 3.3 PM$_{10}$ and PM$_{2.5}$ variation near coal quarries in the Appalachians (West Virginia) [20]

Air pollutants are usually the same when coal is extracted through underground mining: PM and gas, including also methane (CH$_4$), sulphur dioxide (SO$_2$), nitrogen oxides (NO$_x$), hydrogen sulphide and carbon monoxide (CO) [21].

The mixture of these constituents is called *firedamp* and its presence in mine galleries can cause explosions and death by intoxication [22].

Accurate nanoparticle measurements expressed as PM$_{2.5}$ were taken under a thorough study conducted for several years in several mines of the Appalachian region. The research results are given in Fig. 3.4 [11].

The above results show that the most important fraction of fine particles expressed as PM$_{2.5}$ is not evacuated through airshafts; it originates in open-air

Table 3.2 PM$_{2.5}$ variation measured in several places around coal quarries in the Appalachians (West Virginia) [20]

End data	PM2.5 concentration (µg/m^3)			
	Residence (integrated)	School (integrated)	Beckley (integrated)	TEOM (continuous)
2/10/12	NS	NS	13.1	8.3
2/11/12	NS	NS	5.5	8.9
2/12/12 (Non-blasting)	5.7	NS	3.9	6.1
2/13/12	NS	5.5	4.7	14.2
2/14/12	7.7	6.9	8.7	9.4
2/15/12	5.2	5.6	4.5	3.7
2/16/12	8.9	8.9	5.8	8.9
2/17/12	10.2	9.4	8.4	12.0
2/18/12	8.9	8.8	10.6	8.1
2/19/12 (Non-blasting)	NS	6.7	7.4	8.9
2/20/12	8.0	7.4	7.4	12.1
2/21/12	8.4	6.3	7.3	13.1
2/22/12	6.9	6.1	5.5	9.3
2/23/12	9.2	NS	4.9	10.0
Average	7.3	7.1	7.0	9.5
2010 annual average for beckley	10.3			
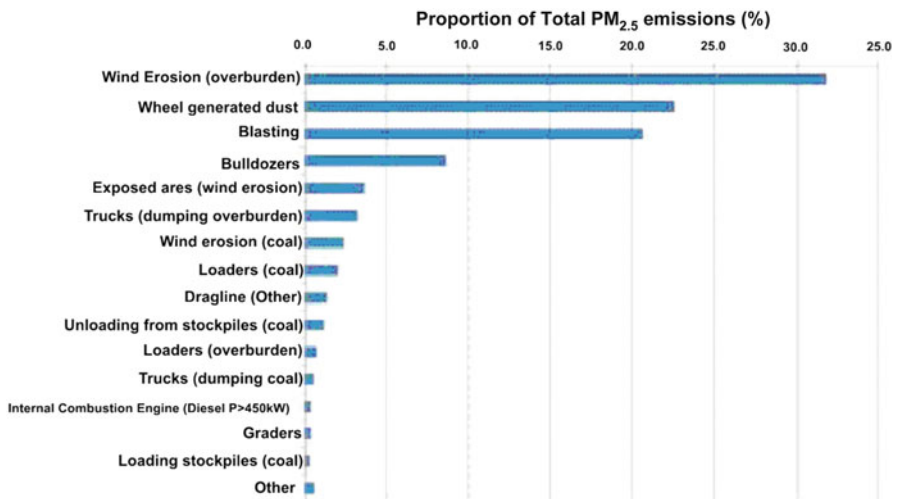
Fig. 3.4 Results of PM$_{2.5}$ measurements in coal mine perimeters [11]

Table 3.3 Mining and separation technologies employed at present [23]

Resource	Mining techniques	Extraction techniques
Iron	Surface mining (open pit mining)	Smelting and chemical reduction
Gold	Underground mining (shaft mining) and surface mining	Gold cyanidation is used. The ore is chemically treated to extract the gold
Copper	Surface mining (open pit mining)	Leaching is used to extract the copper using an acid
Platinum	Underground mining (shaft mining)	Chemical methods and as a byproduct of copper mining
Zinc	Underground mining (shaft mining)	Smelting and leaching
Chromium (Chrome)	Surface mining (open pit mining) and underground mining (shaft mining)	Smelting, redox reactions
Manganese	Surface mining (open pit mining) and underground mining (shaft mining)	Smelting and chemical processes
Diamonds	Surface mining (alluvial deposits) and underground mining (pipe mining)	Diamonds are extracted from rocks and in almost pure form
Phosphate	Surface mining (open pit mining)	Treatment with acid
Asbestos	Surface mining (open pit mining)	Extracted in fairly pure form

activities related to mining, performed especially in the coal storage area, where almost one-third of the whole measured quantity is recorded.

3.2.2 Ferrous and Non-ferrous Ore Extraction

Table 3.3 provides a synthetic presentation of the current mining techniques employed in the extraction of metals and minerals [23].

Iron ore is extracted both by surface and underground mining. The most important surface mining facilities are in South America (Bolivia and Brazil in particular), Western Australia, China, India, Ukraine and Canada. In the past years, these countries have replaced other countries with traditional iron ore mining on the market, such as France, Sweden or Germany. The most important iron ore deposit is at El Mutùn, in Bolivia, with an estimated amount of 40 billion tons of iron ore, of which over 50 % is made of magnetite (Fe_3O_4) and hematite (Fe_2O_3) [24]. Pyrite (FeS_2) is another mineral with significant industrial applications in iron and sulphuric acid production.

After extraction and crushing (and roasting, in case of pyrite) and before the metallurgic process starts, ore concentration occurs through sinterisation, producing a porous conglomerate which is essential for furnace processing, because it allows air circulation [25].

PM determination in an Indian open-pit mining facility [26] has provided the following $PM_{2.5}$ values (Fig. 3.5).

Logically, the highest concentrations of PM (about 20 µg/Nm^3) were measured in point A1, corresponding to the office building of the mining facility, and point

Fig. 3.5 PM$_{2.5}$ monthly averages in Baliparbat-Daitari, India
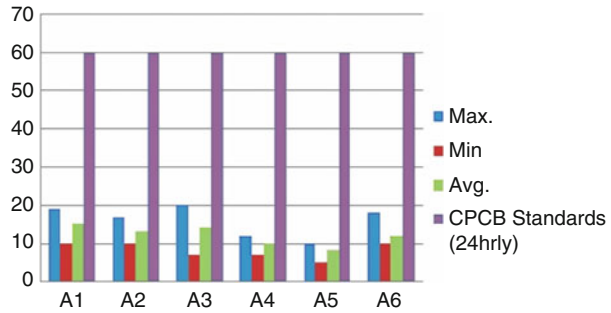
A3, near the OMC (the company that took the measurements) office building. However, similar concentrations were measured outside the mining facility, in various places of local social activity, which indicates that nanoparticles do no show sedimentation tendencies.

The amounts of metallic copper found in nature cannot cover the market demand. Therefore, it is exploited by mining and successive industrial processing. The most important copper ores are copper matte $CuO(OH)$, cuprite Cu_2O, tenorite CuO, algodonite Cu_6As, paramelaconite $(Cu^+Cu^{2+})_2O_3$ and chalcocite or copper pyrite CuS.

A long-term study on PM emissions in copper mining facilities provides relevant results in the field (Table 3.4) [27]. For these emissions, it is easy to notice a general decreasing tendency in the concentrations of all indicators characterising PM, which may be the consequence of both technological improvement along the past two decades and the state-of-the-art monitoring equipment, besides the specific legal limitations. PM$_{2.5}$ variations along the years—an increase of the initial value followed by a constant decrease towards the end of the studied period—may be the result of the intensive/extensive copper ore extraction activity depending on the metal demand on the market.

Studies similar to those conducted for copper extraction in India were carried out for a chromite mine belonging to the same company. The PM$_{10}$ values were between 65–85 μg/Nm^3 within the quarry perimeter and 45–76 μg/Nm^3 in Bangur, a locality near the quarry [28]. Although within the admitted range, most of these values are closer to the upper limit. The most important chromite ores, used almost exclusively for chromium production, are the chromite $(FeCr_2O_4)$ and the crocoite $PbCrO_4$ [29].

A comparison of the values recorded for chromite ore excavation (Table 3.5) with those recorded within the iron ore quarry (Table 3.6) will show that the former are higher without exception. This means that either the excavation tools crush the rocks into finer particles or the chromite mining facility is at a much lower altitude than the iron mining facility, and since there are no air currents to spread the PM, they accumulate in the extraction caldera. Such tendencies are also present in countries that use advanced technologies in surface mining [30].

Table 3.4 Average $PM_{2.5}$ values measured in a long-term study conducted in an open-pit mining facility in a copper ore deposit in Rosemont (Virginia, USA) [27]

Pollutant	Period	Year 1	Year 5	Year 10	Year 15	Year 20
PM/TSP	lb/h	1,273.17	1,544.97	1,376.67	1,361.96	1,009.35
	tpd	9.66	12.93	10.91	10.73	6.50
	tpy	2,923.53	3,370.34	2,859.10	2,787.85	1,658.96
PM_{10}	lb/h	443.43	513.41	470.14	464.83	371.71
	tpd	2.79	3.63	3.11	3.05	1.93
	tpy	852.11	961.72	830.18	801.94	495.68
$PM_{2.5}$	lb/h	54.88	61.98	57.66	56.84	46.64
	tpd	0.45	0.54	0.49	0.48	0.36
	tpy	141.60	152.52	139.39	135.64	101.67
CO	lb/h	3,516.38	3,516.38	3,516.38	3,516.38	3,516.38
	tpd	2.13	2.13	2.13	2.13	2.13
	tpy	644.83	615.22	611.73	447.98	184.94

Table 3.5 PM_{10} values measured in an open-pit mining facility for chromite extraction in Bangur (Uttar Pradesh, India) [28]

Mine Office (A1)		Results in μg/m³		
Sample No.	Date	RPM	SO$_2$	NO$_x$
1	03.04.11	85	<4.0	11.2
2	07.04.11	58	4.0	14.3
3	11.04.11	57	<4.0	19.5
4	15.04.11	65	<4.0	22.3
5	18.04.11	65	4.0	20.8
6	21.04.11	75	7.6	22.6
7	25.04.11	73	4.3	27.8
8	30.04.11	78	5.1	21.0
Bangur village (A2)				
1	03.04.11	59	6.1	16.9
2	07.04.11	57	6.7	13.8
3	11.04.11	45	6.0	10.2
4	15.04.11	51	<4.0	14.3
5	18.04.11	61	<4.0	14.1
6	21.04.11	73	5.0	18.4
7	25.04.11	53	<4.0	10.4
8	30.04.11	76	7.1	12.2
National ambient air quality standards	Annual	60	50	40
	24 h	100	80	80

Asbestos is an iron and sodium hydroxy silicate with the following formula: $Na_2Fe^{2+}{}_3Fe^{3+}{}_2Si_8O_{22}(OH)_2$. Its fibre size is compared with other sub-micrometre particles in Fig. 3.7 [32].

Table 3.6 Size of asbestos fibres revealed by measurements taken near an asbestos quarry in Quebec [32]

	Outdoor air MDDEP (Bisson and Couture, 2007) (f/ml)	Indoor air AVAQ (Marier et al. 2007) (PCMe fibres/ml)
Counting criteria	L >5 μm, D <3 μm, and ratio L/D >3:1	L >5 μm, D ≥0.25 μm and <3 μm and ratio L/D >3:1
Average	0.0029	0.0020
Minimum	0.00038	0.000553
Maximum	0.028	0.010
Upper limit of 95 % Cl	0.0035	0.0031

Concentrations vary from 6–22 μg/Nm^3 in indoor air to 28–96 μg/Nm^3 [33]

Once the use of asbestos in large-scale applications was limited because of its proven cancer-causing characteristic, asbestos mining decreased considerably, but did not stop. Consequently, in order to limit the danger of asbestosis and lung cancer, numerous studies have been carried out to establish strict and coherent rules for asbestos exploitation [31–34].

As shown in Fig. 3.6, asbestos fibres can vary in size from 80–90 nm to 10 μm. As a result, they are considered potentially dangerous for any kind of living cells [33].

Table 3.6 shows values of asbestos fibres measured inside and outside buildings situated near an asbestos quarry in Quebec (Canada).

### 3.2.3 Exploitation of Natural Construction Materials

Stone quarries and gravel pits are major sources of nanoparticle pollution, especially if stone crushers and/or sorting machines are used for sorting sand or crushed material by grain size.

Most technological processes of rock removal (or extraction by water bed dredging), transport (conveyor belts or trucks), grain sorting and storage take place in open air, therefore wind carries away stone dust and fine sand particles.

In a stone quarry located near Harare (Zimbabwe), in a usual working month, the level of PM expressed as respirable and inhalable PM varied with the distance from the crusher and the stone sorting machine, as shown in Fig. 3.7 [35].

The PM amounts in the two categories represent maximum and minimum values corresponding to discontinuous operations of unloading stone from the crusher. The obvious general tendency is a concentration decrease with the distance from the quarry machines. The smaller the particles are, the smaller the decreasing tendency is. PM_4 and $PM_{2.5}$ emissions in a silica sand quarry in Arizona were measured before and during plant operation with vibrating sieves [36]. The values exceeded the limits established in US EPA, but this is a general issue of any plants operating with particulate matter.

Fig. 3.6 Size of asbestos fibres compared with other materials
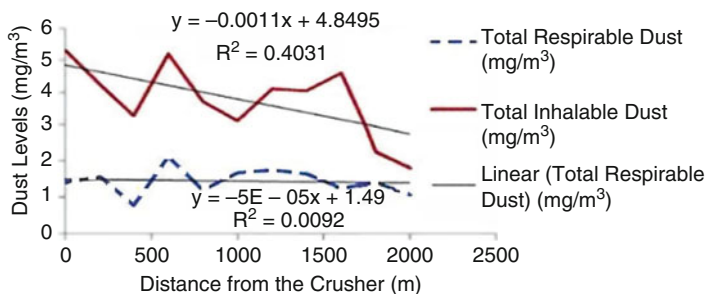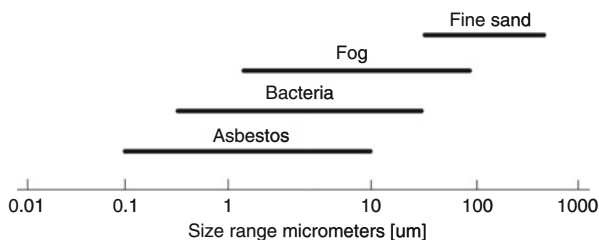
Fig. 3.7 Level of respirable and inhalable PM in the stone quarry at Pomona (Zimbabwe) in January 2011 [35]

3.3 Secondary Sources of Nanoparticles

Secondary sources of anthropogenic nanoparticles are among the most harmful sources, although their contribution is smaller by comparison with both natural PM and PM released during mining activities. If during the evolution of the biosphere its constituents adapted to the impact of natural PM, even to dust containing pathogens or toxic metals, through the parallel evolution of metabolism, PM resulting from industrial activities is more recent, therefore much more harmful to the living beings, both as a consequence of its diverse origins and its toxicity, since man has synthesised millions of substances so far.

Figure 3.8 [37] shows that the European regions with the highest concentration of atmospheric nanoparticles are grouped in the most developed Western European states: Leeds-Manchester industrial zone (Great Britain), Belgium, The Netherlands and Luxemburg (almost entirely); the Paris region and the lower course of the Seine (France), Ruhr Basin (Germany), the Turin-Milan-Venice-Florence area (Italy), the Bohemian Plateau (the Czech Republic) and the coal region of Silesia (Poland). In Eastern Europe, high concentrations of nanoparticles are found in the Donetsk Basin (Ukraine), where large iron and coal deposits are explored [38, 39].

Road transport is the major source of nanoparticle emissions released in the atmosphere (Fig. 3.9) [37].

Quite unexpectedly, the second major contribution to atmospheric pollution with PM comes from point residential sources, not from the energetic sector. Relevant
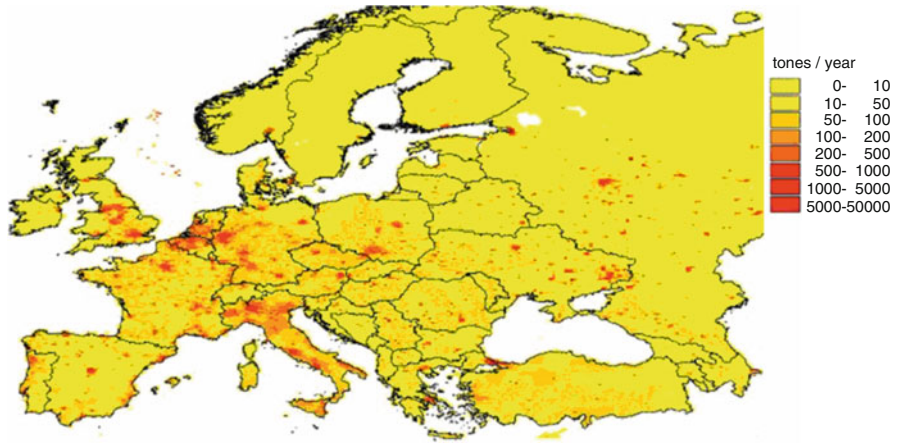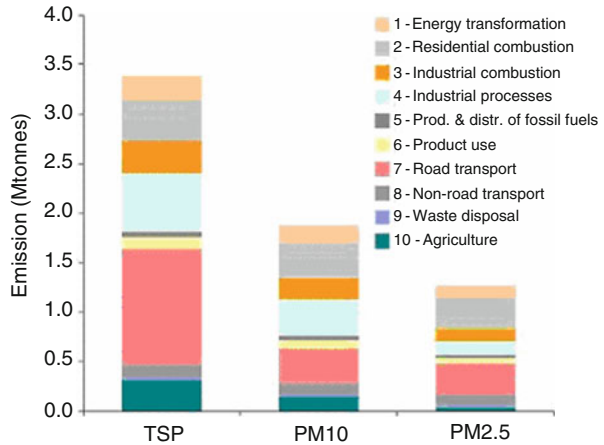
Fig. 3.8 Pan-European PM$_{2.5}$ emissions [37]

Fig. 3.9 Particle emissions in 15 European Union states, Iceland, Norway and Switzerland, in 2005 [37]


and almost equal contributions come from the industrial combustion sector (thermal and catalytic reformation of hydrocarbons, chemical conversion of coal, syngas production, etc.), other chemical processes, industrial processing of resources and the municipal and industrial waste incineration sector.

3.3.1 Stationary Sources of Nanoparticles

As mentioned at the beginning of this chapter, stationary sources of nanoparticles are industrial thermal power plants, chemical plants producing dust-containing products like fertilisers or solid pesticides, cement plants and municipal or special

Fig. 3.10 Size distribution of PM$_{2.5}$ flying ash collected from the stack of a thermal power plant in California (USA) [47]
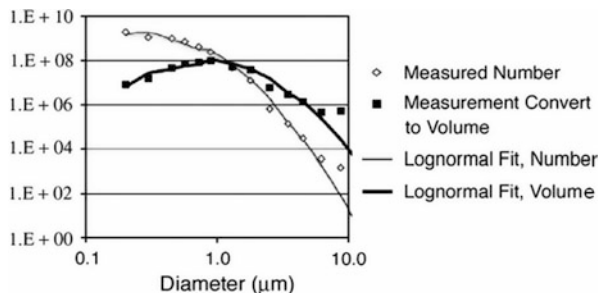
waste incineration plants. This category includes the point sources of the residential heating sector [40–44].

3.3.1.1 Thermal Energy and Power Generation Systems

Thermal power plants release PM in different ways: flying ash escaping from the electric filters mounted on the stack, bottom ash extraction, fine dust scattering and final ash storage, if ash is not dried and sprayed permanently [45, 46].

Studies on the PM$_{2.5}$ fraction of flying ash in a fluidised bed coal-fired thermal power plant have revealed the particle distribution shown in Fig. 3.10 [47].

The data presented so far indicate that about 50 % of the ash collected from the electrostatic filter bags is 100–1000 nm in size.

For the bottom ash resulted from fluidised bed combustion of crushed coal in several thermal power plants in Poland, the size distribution of nano- and microparticle-containing fractions is given in Fig. 3.11 [46].

The comparison of the size distribution of flying ash and bottom ash in modern thermal power plants in Poland and the USA indicates that PM$_{1-2.5}$ in the bottom ash are less than a quarter of the whole amount and half of the nanoparticle concentration in the flying ash.

The two types of ash differ very slightly in their chemical composition. The difference lies in the origin of the coal [46, 47]. Their typical chemical composition is given in Table 3.7 [47].

The composition was determined by high-resolution methods:

– Ion chromatography
– AAS spectrophotometry
– Total organic carbon
– X-ray spectroscopy

The current trend of the studies dealing with the recovery of unburnt carbon from coal ash, as well as the encouragement of renewable fuels in the energetic production require the use of vegetal pellet or dust (straw, wood) in combination with bottom ash [48].
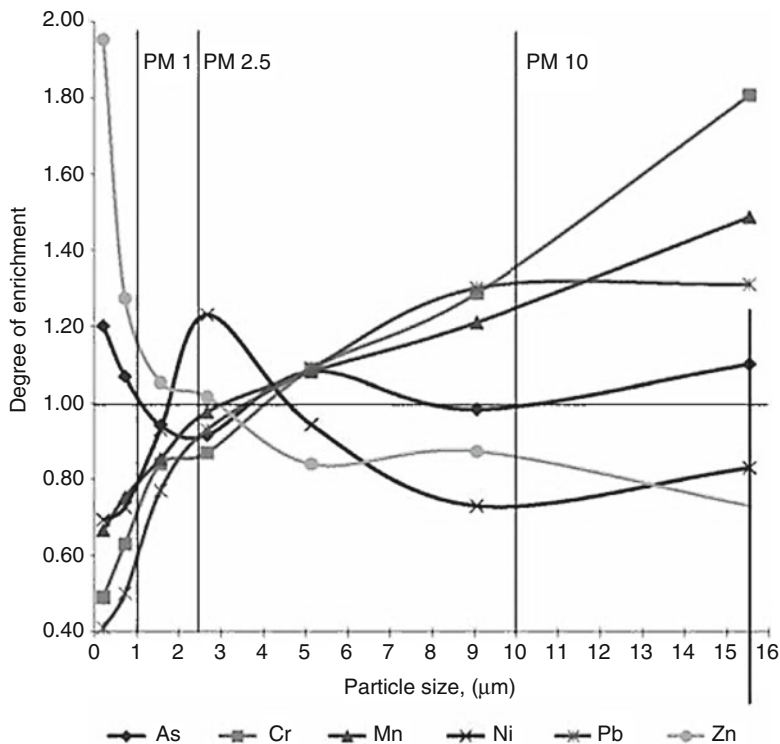
Fig. 3.11 Size distribution of bottom ash in a fluidised bed coal-fired thermal power plant

Under the circumstances, aerosol formation during combustion was studied in a fluidised bed 800 MW$_t$ h boiler, during combustion of finely divided wood, with and without coal or flying ash as alkaline capture additive.

Both the physico-chemical characteristics and the PM morphology of the aerosol samples were analysed. Sub-micrometre sizes were determined with the electron microscope (SEM/EDS and Met/EDS). Sub-micrometre particles (PM$_1$) with a concentration of 44–47 mg/Nm^3 were identified.

The Gaussian distribution of nanoparticles highlighted a characteristic maximum peak at about 200 nm. This peak consisted mainly of ultrafine aggregates (<100 nm) of particles rich in K, Cl and S formed through the homogeneous and heterogeneous vaporisation of solutions containing chloride, potassium, sodium and sulphate ions from the gaseous state.

In an experimental variant based on ash tree wood flour with the addition of alkaline flying ash, for a 1:2 or 1:4 mass ratio, the Gaussian distribution of sub-micrometre particles indicated sizes larger than those in the experiments without coal flying ash. The PM$_1$ concentration diminished from the above-mentioned value to 11–19 mg/Nm^3.

In this case, ultrafine particles (<60 nm) had a more regular, almost spherical shape, and the composition changed from the Na-K-Cl-S quaternary system to the

Table 3.7 Chemical composition of the same thermal power plant ash [47]

| Soluble ions by atomic absorption or ion chromatography	
Chloride	0.003
Nitrate	ND[a]
Phosphate	ND
Sulphate	3.97
Ammonium	0.03
Sodium ion	0.37
Potassium ion	0.06
Calcium ion	3.67
Carbon fractions by thermal/optical reflectance	
Organic carbon 1	0.05
Organic carbon 2	0.29
Organic carbon 3	0.34
Organic carbon 4	0.46
Pyrolysed carbon	0.09
Elemental carbon 1	0.50
Elemental carbon 2	1.40
Elemental carbon 3	1.28
Carbonate carbon	0.35
Elements by X-ray fluorescence	
Sodium	1.00
Magnesium	0.14
Aluminium	4.92
Silicon	12.43
Phosphorus	0.22
Sulphur	1.26
Potassium	1.23
Calcium	5.64
Titanium	0.65
Vanadium	0.06
Chromium	0.02
Manganese	0.03
Iron	4.16
Cobalt	ND
Nickel	0.004
Copper	0.02
Zinc	0.04
Selenium	0.009

[a]Not detected
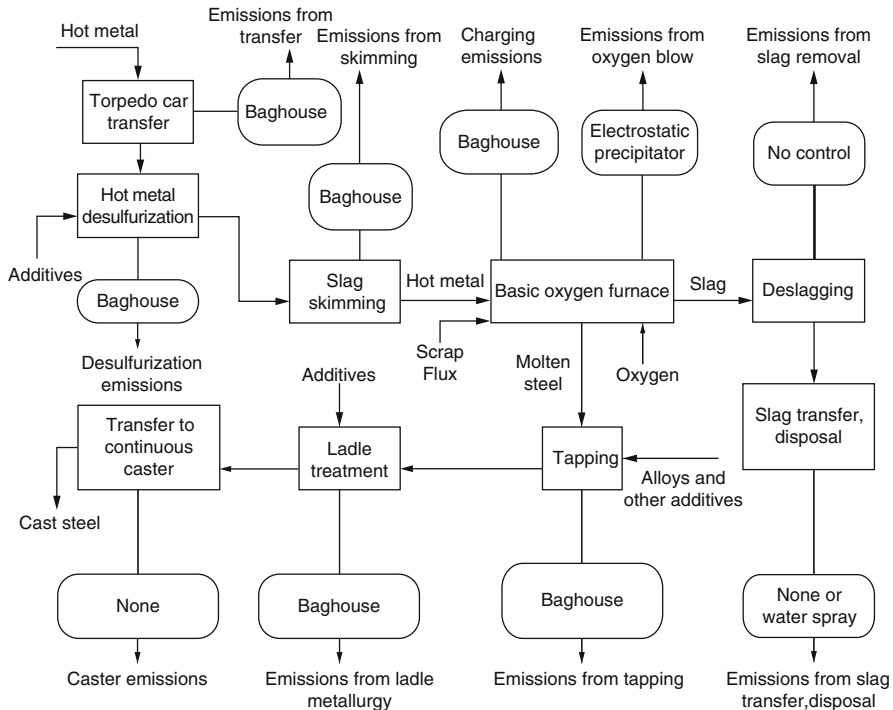
Fig. 3.12 Sources of nanoparticles in a steel plant in Michigan (USA) [51]

Ca-P-Si tertiary system, which demonstrated that the ions resulted mostly from the coal flying ash. This proved the optimisation/minimisation of the solid aerosol content of the coal flying ash added to the alternative dust fuel in fluidised bed coal-fired thermal power plants [48–50].

3.3.1.2 Ferrous and Non-ferrous Metallurgy

A recent thorough study on several steel plants with coke oven batteries in the Great Lakes area reveals both direct sources of $PM_{2.5}$ (Fig. 3.12) and sources that generate nanoparticles through *de novo* synthesis between PAH and the heavy metals released in the atmosphere (Fig. 3.13) [51].

The total amounts of $PM_{2.5}$ released from the steel plants are given in Table 3.8 [51].

The above-mentioned experimental data indicate that these amounts must not be ignored, especially because they include heavy metals under the form of both organic and organometallic compounds that are very dangerous for health and the environment.

As far as the orientative distribution of iron-containing nanoparticle concentration in large steel plant areas is concerned, the concentrations measured in various
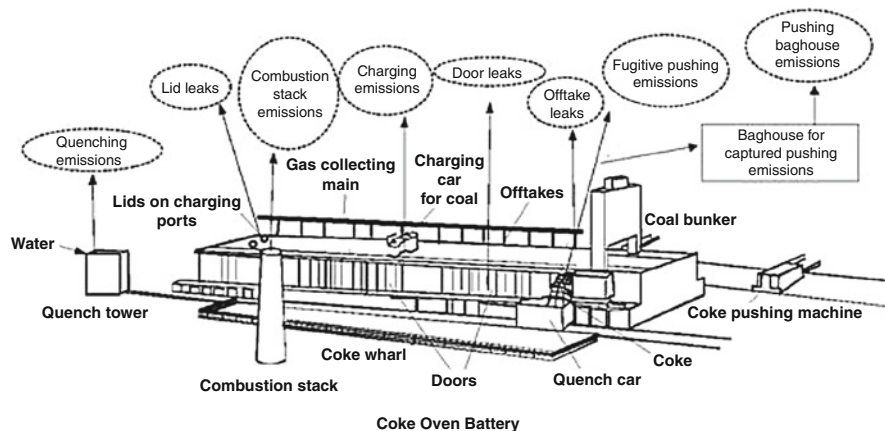
Fig. 3.13 Sources of nanoparticles in a coke oven battery in Michigan (USA) [51]

Table 3.8 Annual emissions of nanoparticles in a steel plant with a coke oven battery in Michigan (USA) [51]

Pollutant	Emissions (tpy)	Percent
SO_x	4,567	35
NO_x	5,616	43
PM-CON	1,876	14
$PM_{2.5}$-FIL	1,130	8
Total	13,189	100
HAP	Total emissions (tpy)	HAP in PM2.5 (tpy)
Manganese	13	7.2
Lead	1.9	0.7
Nickel	0.04	0.01
Chromium	0.2	0.1
Mercury	0.4	0.4
Total	15.5	8.4

adjacent points varied from 250 to 3,000 ng/Nm^3, depending on the environmental factors and the Saharan dust advection (Fig. 3.14) [52].

The days when the natural nanoparticle concentration was higher than that of the investigated emissions were days when Saharan dust was present in the atmosphere.

3.3.1.3 Chemical Industry

The sources of nanoparticles released by chemical product plants are so diverse that listing them here would go beyond the scope of the present monograph. This is because besides raw materials, half-finished and finished products under the form of PM with direct impact on air quality, any chemical plant releases, intentionally or unintentionally, gases that interact with other pollutants and generate PM.
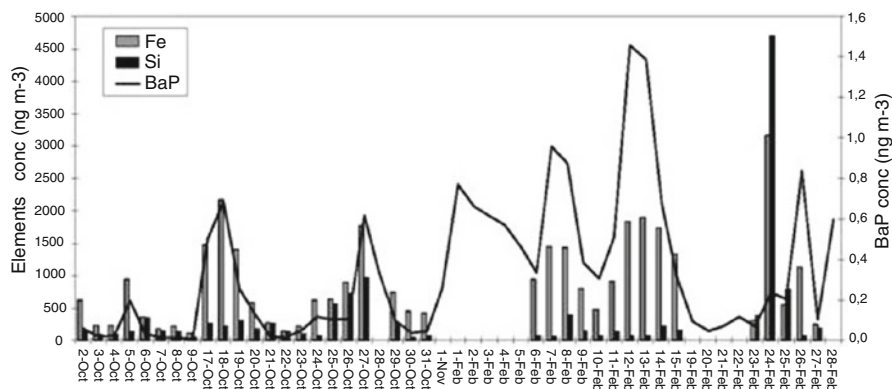
Fig. 3.14 Variation of iron and silicon concentration in PM in the Apulia (Italy) steel plant [52]

Consequently, fertilisers, pigments and pesticide production will be dealt with in short, as these are the most representative sub-branches that produce chemical dust particles.

A case of global investigation of pollutants generated on an industrial platform in South-Western Spain reveals the interaction between many metallic ions released from steel or other metallurgic plants with various constituents released in the atmosphere during phosphate rock processing under the form of NPK fertilisers or by road transport [54].

In a large NP (ammonium phosphate) fertiliser plant in India, air quality monitoring revealed $PM_{2.5}$ particles between 13 and 38 μg/Nm^3, values that are within the allowed local range (Table 3.9) [53].

Nanoparticles generated in pesticide production and large-scale uses in agriculture are harmful to man's health and the biosphere. Consequently, studies were conducted on their travel distance in air under the influence of environmental factors, their effect as nanoparticles or VOC in the atmosphere expressed as half-life [41].

The dyes and pigments industry releases fine inorganic or organic dust in the air [55–57]. Inorganic pigments are based on heavy metal salts whose ions have characteristic colours. Dyes that contain such pigments are mostly mixtures of pigments and siccative oils or other organic substances that can form polymerisable films. The most common ions found in inorganic pigments are chrome, nickel, iron, cobalt, lead, zinc and arsenic ions; as a result, the released nanometric dust is extremely dangerous [55].

A federal monograph of American industrial sources of nanoparticles highlights the annual PM concentrations, including inorganic pigments, and recommends measures to limit them these concentrations (see for example Table 3.10) [56].

As a rule, organic pigments are functional derivatives with chromofore groups that ensure the coloured perception of these substances by the human eye. Most of them (anilins, azoic derivatives, microdispersed pigments) are toxic and cause cancer. It has been proved that those pigments that are to be eliminated from current surface protection processes belong to the polychlorinated biphenyls class whose use was strictly limited long ago [57].

Table 3.9 Data about air quality monitoring at the fertiliser plant in Mangalore (Jaipur, India) [53]

Details		PM10 µg/ m^3	PM2.5 µg/ m^3	NH$_3$ µg/ m3	SO$_2$ µg/ m^3	NO$_x$ µg/ m^3	CO mg/ m^3	Hydrocarbon µg/ m^3	F$^-$ µg/ m^3
Jan. 2013	Max.	55	38	58	9	9	0.26	ND	ND
	Min.	22	15	<5	<5	<5	<0.1		
Febr 2013	Max.	58	36	57	9	9	0.23	ND	ND
	Min.	26	16	<5	<5	<5	<0.1		
March 2013	Max.	56	36	57	9	9	0.22	ND	ND
	Min.	25	13	<5	<5	<5	<0.1		

ND not detectable

3.3.1.4 Cement Industry

In the past 20 years, industrial systems of cement production have made great progress, so that they can set higher limits to environmental pollution with both gaseous constituents and PM. These very expensive technological changes were imposed by the conclusions founded on measurements taken in many cement factories around the world and mathematical models based on which the long-term effects of pollution under the influence of atmospheric factors were evaluated [58, 59].

The long-term measurements taken in a large cement factory in Harbin (China) during 1990 and 2010 indicate that, in the case of very fine dust, the PM$_{2.5}$ value dropped to half in less than 15 years (Fig. 3.15) [58].

Unfortunately, this obvious worldwide tendency of reducing atmospheric PM emissions does not characterise other pollutants released during cement production (CO, CO$_2$, NO$_x$, SO$_2$), as their value rose with production increase [59].

3.3.1.5 Waste Incineration

Waste incineration is one of the largely employed methods of waste removal both for reducing waste amounts and toxicity and energy recovering as a final alternative of thorough use. Depending on their destination, industrial incineration plants eliminate either municipal waste or flammable industrial, sanitary and toxic waste that must be destroyed with strictly controlled methods [60].

Municipal Waste Incineration

PM emissions in modern municipal waste incineration plants are limited under the provisions of EC Directive on waste incineration [61].

Table 3.10 Annual PM$_{2.5}$ concentrations in the St. Louis (USA) industrial area [56]

Facility Name	SCC	SCC L2	SCC L3	SCC L4	PM2.5-PRI emissions (tpy)	Control classification
American Commercial Terminals	30501011	Mineral products	Coal Mining, cleaning, and material handling (see 305319)	Coal transfer	1,052	Uncontrolled
Amerenue-Meramee Plant	101000226	Electric generation	Bituminius/sub-bituminous coal	Pulverised coal: dry bottom tangential (sub-bituminous coal)	754	Controlled
Pace Construction Co-Chesterfield	30500260	Mineral products	Asphalt concrete	Drum mix plant: rotary drum dryer/mixer, #2 oil-fired, counterflow	494	Uncontrolled
Dial Corp–Dial Corp	30113210	Chemical manufacturing	Organic acid manufacturing	Acetic acid via acetaldehyde	426	Uncontrolled
Dynegy Midwest Generation Inc	10100202	Electric generation	Bituminius/subbituminous coal	Pulverised coal: dry bottom (Bituminous coal)	350	Controlled
Dynegy Midwest Generation Inc	10100203	Electric generation	Bituminius/subbituminous coal	Cyclone furnace (Bituminous coal)	348	Controlled
Dynegy Midwest Generation Inc	10100203	Electric generation	Bituminius/Subbituminous coal	Pulverised coal: dry bottom	341	Controlled
Elementis Specialties Inc	30103553	Chemical manufacturing	Inorganic pigments	Pigment dryer	184	Uncontrolled
Elementis Specialties Inc	30103553	Chemical manufacturing	Inorganic pigments	Pigment dryer	184	Uncontrolled
Masterchem Industries Inc Imperial	30101401	Chemical manufacturing	Paint manufacturer	General mixing and handling	117	Uncontrolled
US Silica Company Pacific	30502511	Mineral products	Construction sand and gravel	Screening	113	Uncontrolled
Anheusr Busch Inc-St Louis	10200202	Industrial	Bituminius/Subbituminous coal	Pulverised coal: dry bottom	102	Controlled
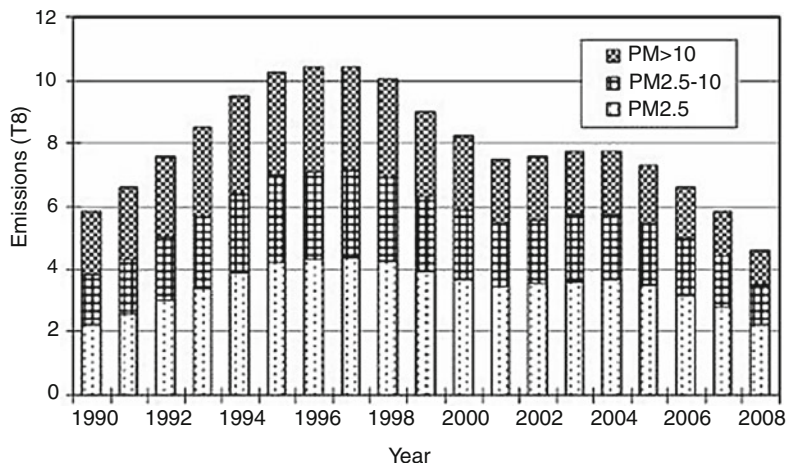
Fig. 3.15 Variation of PM values for dust emissions measured at the cement factory in Harbin (China) [58]

All municipal solid waste (MSW) incinerators are equipped with bag filters that retain particulate matter. In Great Britain, such incinerators contribute to atmospheric pollution with 0.042 % of the total $PM_{2.5}$ emissions [62].

Some studies show that, as far as municipal waste incineration is concerned, ultrafine particulate matter $PM_{0.1-1}$ can have significant effects on health and the environment. These studies hold that health risks are associated rather with the number of ultrafine particles than with their mass [63–65].

Measurements of nanoparticles released at the stack of a municipal waste incinerator with energy recovery located in Piacenza (Italy) [63] indicated that no particles larger than 2.5 μm in diameter were present in the emissions, but about 65 % of the measured $PM_{2.5}$ emissions were sub-micrometre particles (Fig. 3.16). Particles larger or smaller than 100 nm had a relatively equal distribution [63, 64].

The identification of the primary composition of stack emissions released by the same incinerator led to the conclusion that the improved particle retention systems were effective in removing 99.99 % of emissions, with similar efficiency in reducing particles of smaller diameter [65].

It has been found that the boiling points of the elements present in the incinerated waste are very important: higher boiling point elements like chromium were identified in larger particles, while lower boiling point elements like arsenic and cadmium were more present in ultrafine particles, making them more dangerous [65].

Nevertheless, the conclusions that several authors from North America and Japan and European researchers have drawn about the level of global pollution with ultrafine particles are unanimous: in the general context of anthropogenic
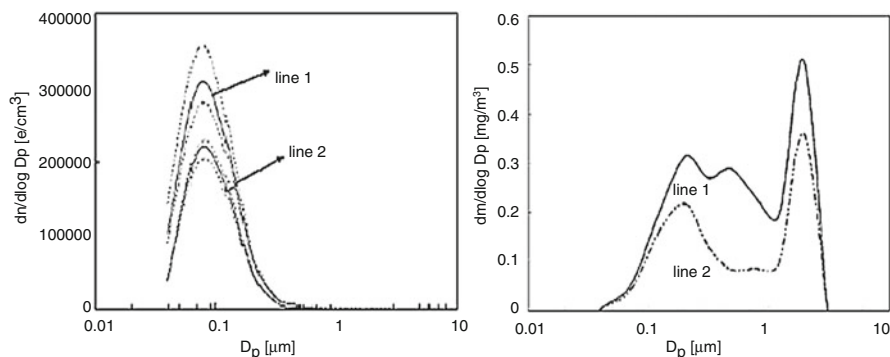
Fig. 3.16 Size distribution of stack nanoparticles in the municipal waste incineration plant in Piacenza (Italy)

emissions, the nanoparticle contribution resulted from MSW incineration plants is small [62].

Hazardous Waste Incineration

Hazardous waste is eliminated under specific regulations that are meant to guarantee the reduction to a minimum of direct emissions, through VOC or other forms of volatile or particulate waste release and stack emissions [66, 67].

Current norms regarding hazardous waste incineration require strict limits both for $PM_{2.5}$ emissions and indirect nanoparticle emissions under the form of heavy metals which are more or less volatile at high temperatures (Table 3.11) [62, 67].

Although incineration of any type of waste has a small contribution to anthropogenic nanoparticle emissions [62–65, 67], there are studies that attempt at finding a direct correspondence between thermal waste elimination and the statistical parameters of population health (Fig. 3.17) [68, 69].

According to these sources, waste incineration plants produce huge amounts of fine and ultrafine particles, because the current standard value of PM emissions (10 mg/m^3) specifies a filter retention rate of only 5–30 % for $PM_{2.5}$ and 0 (zero) for PM_1. In fact, most stack emissions are ultrafine particles of the most dangerous type. Fabric filters with Teflon membrane are not effective against particles under 200–300 nm which are very harmful to health [69]. In this case, what matters is the particle number and size, and not their weight [65, 68].

The distribution of particle size by weight will cause great errors, especially in hazardous waste incinerators with equipment for reducing nitrogen oxides, which may actually double the $PM_{2.5}$ emissions [69, 70].

Table 3.11 Emission limits recommended for incineration of hazardous waste and sewage sludge [67]

	Code	Name			
NFR source category	5.C.1.b.i, 5.C.1.b.ii, 5.C.1.b.iv	Industrial waste incineration including hazardous waste and sewage sludge			
Fuel	NA				
Not applicable	HCH				
Not estimated	Nh3, Cr, Cu, Zn, Se, Benzo(a)pyrene, Benxo(b)fluoranthene, Benzo(k) fluoranthene, Indeno(1,2,3-cd)pyrene, PCBs				
Pollutant	Value	Unit	95 % confidence interval		Reference
			Lower	Upper	
NO_x	0.87	kg/Mg waste	0.087	8.7	European Commission (2006)
CO	0.07	kg/Mg waste	0.007	0.7	European Commission (2006)
NMVOC	7.4	kg/Mg waste	0.74	74	Pasant (1993)
SO_2	0.047	kg/Mg waste	0.0047	0.47	European Commission (2006)
TSP	0.01	kg/Mg waste	0.001	2.3	European Commission (2006)
PM_{10}	0.007	kg/Mg waste	0.0007	0.15	US EPA (1996) applied on TSP
$PM_{2.5}$	0.004	kg/Mg waste	0.0004	0.1	US EPA (1996) applied on TSP
BC	3.5	% of $PM_{2.5}$	1.8	7	Olmez et al. (1998)
Pb	1.3	g/Mg waste	0.48	1.9	Theloke et al. (2008)
Cd	0.1	g/Mg waste	0.048	0.15	Theloke et al. (2008)
Hg	0.056	g/Mg waste	0.04	0.08	European Commission (2006)
As	0.016	g/Mg waste	0.01	0.019	Theloke et al. (2008)
Ni	0.14	g/Mg waste	0.048	0.19	Theloke et al. (2008)
PCDD/F	350	µg I-TEQ/ Mg waste	0.5	35,000	UNEP (2005)
Total 4 PAHs	0.02	g/Mg waste	0.007	0.06	Wild (1995)
HCB	0.002	g/Mg waste	0.0002	0.02	Berdowski et al. (1997)

Current modelling methods do not take into consideration the secondary particles reformed from atmospheric gas emissions, far from the incinerator stack, although a number of studies have shown that in 95 % of cases ultrafine heavy metal particles combine with polynuclear aromatic hydrocarbons (PAH) to produce PM_3 or smaller conglomerates. PAHs are toxic, mutagenic, teratogenic and cancer causing by nature and this kind of combinations increases lung cancer risk by almost eight times [69–72].
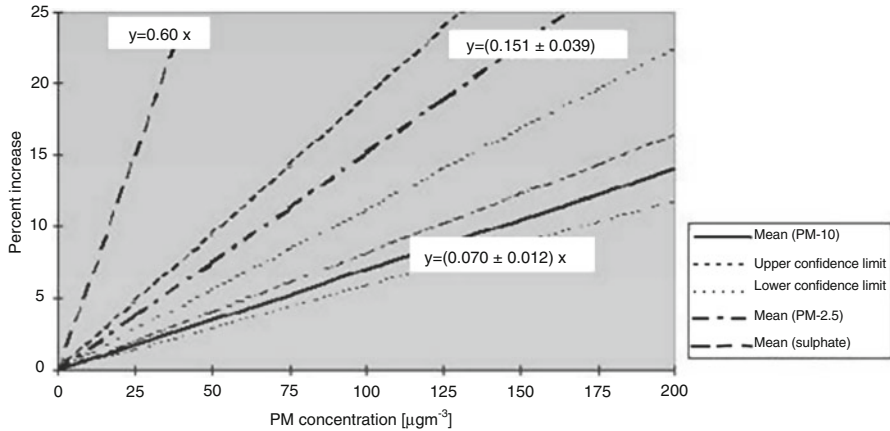
Fig. 3.17 Dependence of mortality on $PM_{2.5}$ and PM_{10} concentrations in Great Britain population [68]

3.3.1.6 Residential Heating

Irrespective of fuel nature, residential heaters have a major contribution to the anthropogenic accumulation of nanoparticles in the atmosphere (Fig. 3.18) [37].

Detailed studies on nanoparticles released by conventional residential and social heating sources (schools, offices, performance halls, etc.) in Fairbanks, Alaska indicate that of the total annual emission amount, the highest nanoparticle concentration comes from wood-fired heaters [73–75]. In order to limit these emissions in areas on the planet where residential heating is a long-term or constant necessity, programmes for the gradual reduction of wood combustion in favour of other low-emission fuels were suggested [75].

Given that fossil fuels obtained from exhaustible sources (coal, oil, natural gas) need to be replaced with renewable fuels (wood, straw, biodiesel, biobenzene), the policies of reducing nanoparticle emissions should be harmonised with the policies concerning the use of such fuel types.

3.3.2 Mobile Sources of Nanoparticles

Mobile sources of nanoparticles are engines. Regardless of fuel nature or mechanical drive, the modern classification of engines identifies two classes:

– External combustion engines;
– Internal combustion engines.
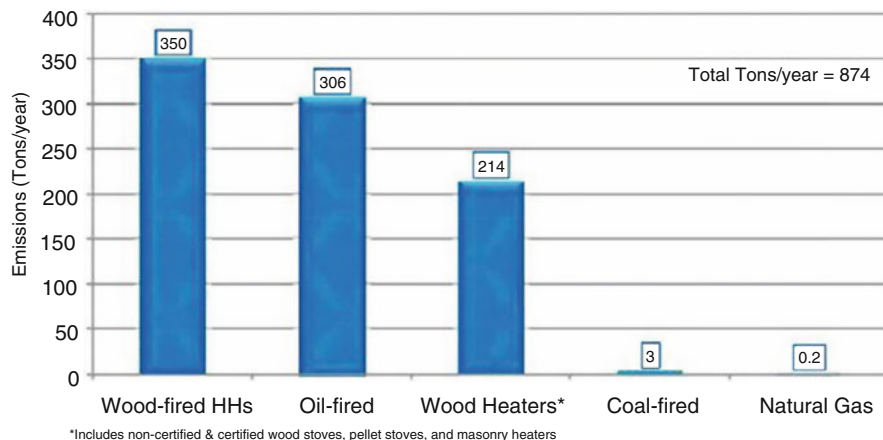
Fig. 3.18 Estimated PM$_{2.5}$ emissions caused by residential heating with various fuels in Fairbanks (Alaska) [75]

Functionally, external combustion engines produce vehicle propulsion through the adiabatic energy of gases that are burnt and evacuated at high pressure and speed—sometimes passing through a turbine. For this reason, many researchers do not include them in the class of proper engines. Additionally, rocket engines use other oxidisers, much stronger than oxygen, instead of air. This is because, on the one hand, rocket engines must produce as much energy as possible in a very short time, and on the other hand, air supply is no longer possible when leaving the atmosphere.

Most internal combustion engines are engines with spark (Otto) or compression (Diesel) ignition. If spark-ignition engines are almost exclusively petrol engines and are used for cars or smaller applications (boat engines, low-voltage generating sets, etc.), depending on their size, diesel engines use diesel oil, light or heavy fuel oil and even coal dust or carbon black (marine engines).

Historically speaking, one should also mention steam engines, either Watt-Carnot or Stirling, which were the basis for industrial and water and road transport development for almost 200 years. However, applications for historical machinery are anticipated for the future [76].

3.3.2.1 External Combustion Engines (Jet and Rocket Engines)

Almost without exception, medium or long-range wide body jet airliners use jet engines or turbojet engines. The distribution of nanoparticles released by jet engines in the northern hemisphere is given in Fig. 3.19. The analytical
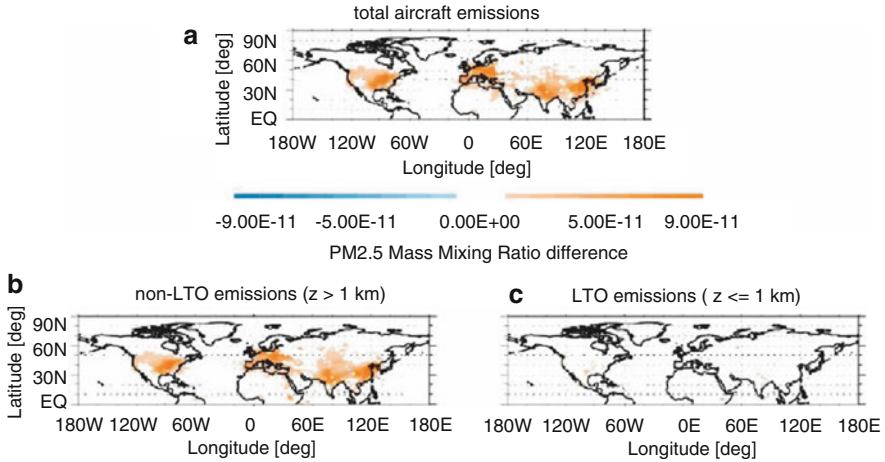
Fig. 3.19 PM$_{2.5}$ emissions from jet engines of planes travelling in the northern hemisphere [77]

determinations shown in these figures indicate that the most densely populated areas on the planet, with the largest number of airports and air routes, are also the most polluted with PM$_{2.5}$ resulting either directly, from kerosene combustion, or indirectly, from the interaction between exhaust gas constituents and the components in Earth's higher atmospheric layers, under the influence of solar radiation [77, 78].

A variation in emission concentration is also visible. Concentration varies with seasons and the altitude where the measurements were taken. It is known that the most frequent air routes for medium and long distances are in the troposphere, at 10,000–14,000 m (Fig. 3.20) [77].

Irrespective of their engine fuel, oxidiser or structure, rocket engines used in the present release, besides CO$_2$, significant amounts of carbon black in the stratosphere. The large carbon black volume is caused by the incomplete fuel combustion, given the low oxygen concentration in the upper atmospheric layers [79–81].

Recent models indicate that, given the current rate of 1,000 rocket launches per year, the effect of the warming potential caused by finely divided carbon particles—carbon black—is about 100,000 higher than the effect of CO$_2$ emissions. Although carbon nanoparticles do not always react with stratospheric ozone, through the solar radiation protection effect, they contribute to ozone depletion [79, 81]. Since carbon black absorbs radiation in the visible spectre and remains in the upper atmosphere for 5–10 years, its effects can lead, in the same period of time, to a temperature increase of 1 K at the two poles, causing changes in the pressure
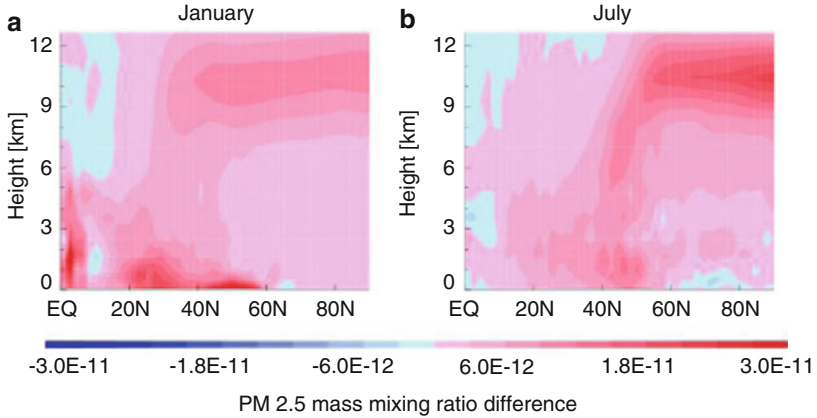
Fig. 3.20 Seasonal and altitude distribution of nanoparticles from aircraft in the northern hemisphere [77]
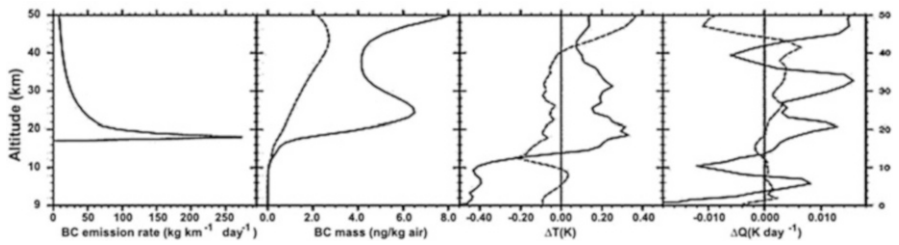

Fig. 3.21 Carbon black emissions in the atmosphere and temperature variations caused by rocket launches in space [79]

gradients [80]. Figure 3.21 shows both the variations in carbon emissions with altitude and the variation of the thermal gradient in relation to the same parameter [79].

3.3.2.2 Internal Combustion Engines

Despite the variety of internal combustion engines, most land and sea vehicles are equipped with petrol-fuelled stark-ignition (Otto) engines or diesel oil-fuelled compression engines. Large commercial ships are equipped with variants of diesel engines fuelled with heavy fuel oil or even coal dust.
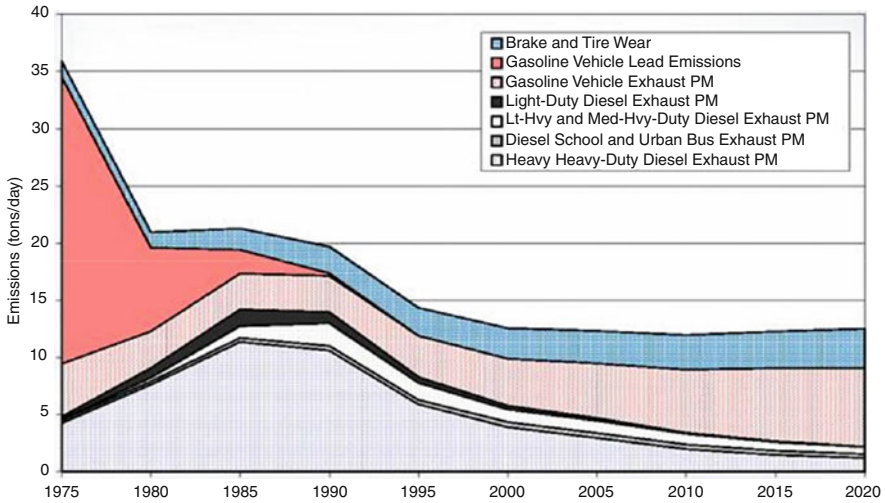
Fig. 3.22 Variation of PM$_{2.5}$ engine-released emissions along the time and in the near future, by fuel type [84]

Regardless of the fuel, these engines release nitrogen oxides, carbon monoxide (CO), particulate matter (PM), volatile organic compounds (VOC) and smaller amounts of other pollutants like sulphur dioxide (SO$_2$) and ammonia (NH$_3$).

Most non-electric trains and big trucks use diesel engines, unlike cars, which have petrol engines. Diesel engines release the same pollutants as petrol engines, but they produce much higher quantities of PM$_{2.5}$ and smaller amounts of VOC and CO (see for example the data presented in Fig. 3.22). Particulate matter released by diesel engines are considered very harmful, as the particles are extremely small and can be inhaled very easily [82–84].

Long distance, high capacity ships, as well as large-scale fish farming activities, naval military actions or large cruise liners are major producers of PM emissions. Accordingly, international agreements limit marine emissions to road traffic emission levels [84].

Nuclear-powered ships, used almost exclusively for military purposes, are vessels that do not release nanoparticles in the atmosphere.

A past and future perspective of PM$_{2.5}$ emissions released by both diesel and stark-ignition engines are presented in Fig. 3.23 [84]. The statistical data show that in the middle of the tenth decade of the past century, lead emissions disappeared completely, because the method of controlling autoignition with tetraethyl lead-based antiknock agents was abandoned.

The obvious decrease in nanoparticle emissions is the consequence of engine manufacturers' efforts to improve technical performances and the attempts to
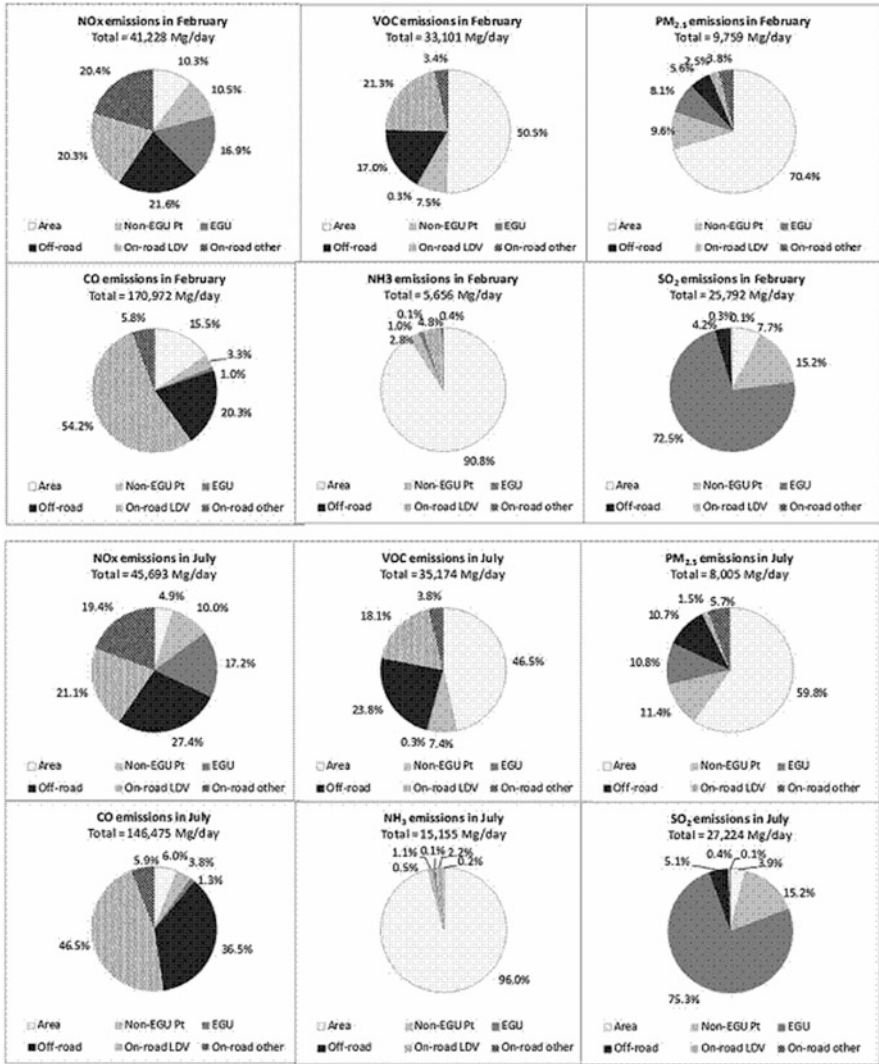
Fig. 3.23 Estimated polluting emissions; road traffic pollutants in central USA in February and July 2008 [83]

control global pollution through fuel cells, electrical engines or non-polluting fuel (water, hydrogen) engines, given the worrying perspective of fossil fuel depletion.

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