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International Agency for Research on Cancer

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1. EXPOSURE DATA

1.1 Definition of outdoor air pollution

Air pollution is the presence in the air of one or more substances at a concentration or for a duration above their natural levels, with the potential to produce an adverse effect (derived from Seinfeld & Pandis, 2006). This definition implicitly acknowledges that some substances that are considered to be air pollutants are present naturally. Although some air pollutants are solely anthropogenic, or nearly so (e.g. chlorofluorocarbons and, for most purposes, some products of fossil fuel combustion), many, including ozone, particulate matter (PM), sulfur dioxide (SO_2), carbon monoxide (CO), and polycyclic aromatic hydrocarbons (PAHs), may also result from natural processes (Table 1.1). Anthropogenic activities have led to increases in many air pollutants to levels that have adverse impacts on human and environmental health. What is key in terms of assessing the potential health effects of air pollution are the levels of the thousands of substances present, recognizing that their composition varies from location to location, so the term "air pollution" can refer to very different exposure mixtures.

1.1.1 Characteristics of exposure to outdoor air pollution

(a) Overview

Exposure to outdoor air pollutants occurs virtually continuously, across microenvironments, including indoors. The composition of the mixture and the absolute levels of the air pollutants change, and many different air pollutants are present. The understanding of these exposures is complicated by the fact that the composition is seldom, if ever, well characterized in any environment, much less in all of the locations an individual may traverse. Although most of the more-abundant pollutants are known and can be measured, many trace species have not been identified, much less quantified routinely. Most trace pollutants typically are not measured; instead, they are characterized as being part of a class of pollutants that are more readily measured together (e.g. many organic compounds are aggregated in measurements), or they may be linked to other indicators.

In light of the complexities discussed in this *Monograph*, there is no standardized way to characterize exposure to outdoor air pollution. This text focuses on outdoor air pollutants, classes of pollutants, pollutant mixtures (characterized by source and/or components), and pollutant indicators that are of specific interest as potentially leading to human cancer due to direct contact with humans through their presence in outdoor air. This includes exposures to outdoor

Table 1.1 Major air pollutants and pollutant classes of interest, their physical state, and their sources

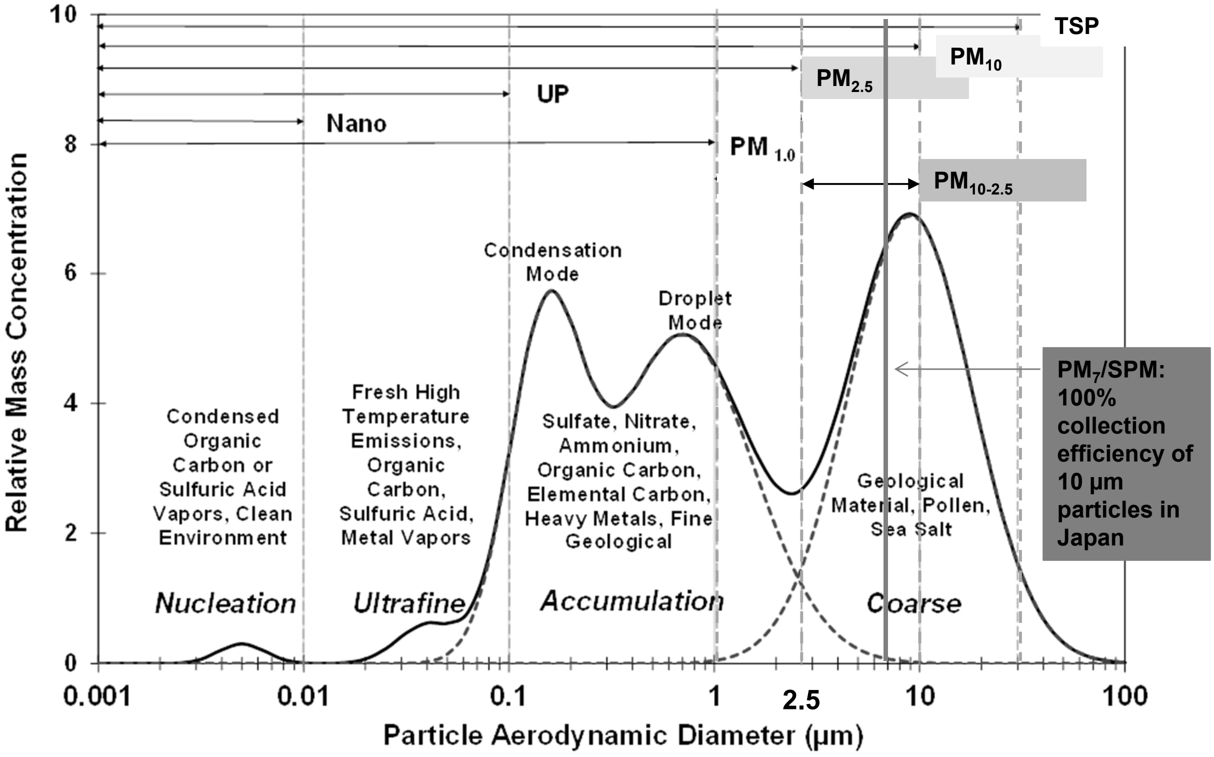
Pollutant/pollutant class	Examples	Physical state	Major sources
Photochemical oxidants	Ozone	Gas	Generated from NO_x, VOCs, and CO, as well as natural processes (e.g. stratosphere)
Sulfur dioxide (SO_2)	SO_2	Gas	Fossil fuel combustion, natural emissions
Carbon monoxide (CO)	CO	Gas	Fossil fuel combustion, particularly spark-ignition engines; oxidation of biogenic VOC emissions
Nitrogen oxides (NO_x)	NO_2	Gas	Combustion processes
Hazardous air pollutants (HAPs)	Benzene, 1,3-butadiene, formaldehyde, acids	Gas	Incomplete combustion, chemical processing, solvent use
Mercury (Hg)	Hg^0, methyl mercury	Gas and particulate	Coal combustion, ore refining, natural
Lead (Pb)	Pb	Particulate	Leaded fuel combustion, lead processing
PM, including $PM_{2.5}$, PM_{10}, inhalable PM, TSP	Inorganic ions (e.g. sulfate); metal oxides; carbonaceous material, including organic carbon (OC) and elemental carbon (EC)	Particulate (condensed phase)	Dust storms, fossil fuel combustion, biomass fuel combustion, biogenic emissions, fertilizer use, gas-to-particle conversion
Organic carbon (OC)	Hopanes, steranes, polycyclic aromatic hydrocarbons, levoglucosan (hundreds of species present, not all identified or quantified)	Particulate	Fossil and biomass fuel combustion, vegetative detritus, oxidation of gaseous organic compounds

CO, carbon monoxide; EC, elemental carbon; Hg^0, elemental mercury; NO_2, nitrogen dioxide; NO_x, nitrogen oxides; OC, organic carbon; PM, particulate matter; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; SO_2, sulfur dioxide; TSP, total suspended particles; VOCs, volatile organic compounds.
Prepared by the Working Group.

pollutants and their reaction products outdoors and indoors. Exposures to pollutants generated indoors and contained indoors (e.g. from cooking and heating) and to smoking are not considered, but pollutants generated indoors that migrate outdoors are considered. An increased focus is on those specific species, classes of pollutants, indicators, and mixtures that are most relevant to cancer in humans.

There are multiple ways to parse exposure to outdoor air pollution. First, the phase can be considered. Air pollutants are typically classified as being gaseous or PM, which contains suspensions of very small particles (with diameters of a few micrometres or less, down to nanometre scales) that are liquid and/or solid matter. Complicating this classification is the fact that some pollutants move between phases (e.g. semivolatile organic compounds [SVOCs]). A second consideration is whether the pollutant is emitted directly, and thus is primary (e.g. dust), or is formed in the atmosphere, and thus is secondary (e.g. ozone). Some pollutants are both primary and secondary (e.g. formaldehyde). Primary and secondary pollutants are described in more detail in Section 1.2. A third approach would be to consider where the exposure takes place (e.g. outdoors, in a car, or indoors) and whether the pollutant mixture has been significantly altered by that microenvironment. A fourth consideration is the source of the pollutant mixture (e.g. fuel combustion, chemical manufacture) or whether the mixture is dominated by secondary compounds.

Fig. 1.1 Major features of atmospheric particle mass distribution



PM$_{10}$, particulate matter with particles of aerodynamic diameter < 10 µm; PM$_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; SPM, suspended particulate matter; TSP, total suspended particles; UP, ultrafine particles.
Adapted from Watson (2002). Visibility: science and regulation. *J Air Waste Manag Assoc*, 52(6):628–713, by permission of the Air & Waste Management Association. (http://www.awma.org).

(b) Air pollutants

Air pollutants and pollutant classes of interest, their physical state, and their major sources are summarized in Table 1.1.

Gaseous compounds of interest include, but are not limited to: ozone; nitrogen oxides (NO_x), which comprise nitrogen oxide (NO) and nitrogen dioxide (NO_2) (in the atmosphere, NO is oxidized to NO_2, often rapidly in the presence of ozone); SO_2; and a myriad of volatile organic gases (often referred to as organic gases or volatile organic compounds [VOCs]). VOCs include aldehydes (e.g. formaldehyde), ketones, aromatics (e.g. benzene), alkanes, and other classes. PM is even more complex (and less well understood). PM is characterized by both its size and its chemical composition (see Fig. 1.1). Particle sizes range across about 5 orders of magnitude, from the nanometre scale (e.g. as clusters of molecules) up to grains of dust on the order of tens of micrometres. Although the distribution of particle sizes can be measured, most measurements capture the mass in specific size ranges; for example, PM$_{2.5}$ is PM with particles of aerodynamic diameter less than 2.5 µm. Other common size classes are PM$_{10}$ (PM < 10 µm), total suspended particles (TSP), and ultrafine PM (PM < 0.1 µm). Coarse PM is taken as the fraction with diameters from 2.5 µm to 10 µm. These size classes are relevant to PM dynamics in the atmosphere and uptake in the human body, and reflect size ranges used in health studies.

Broad classes of chemical compounds can be considered in PM: inorganic ionic compounds (e.g. sulfate), metal oxides (e.g. silicon oxide), and carbonaceous PM, which can in turn be classified as organic carbon (OC) and elemental carbon (EC).

EC is not truly pure carbon and is defined by the measurement approach; in some cases, the term black carbon (BC) is used, again being defined by the measurement approach.

OC comprises hundreds of compounds, and although the common inorganic compounds are reasonably well known and measured, only a fraction of the organic compounds in PM have been specifically identified (Schauer et al., 1996). Significant headway has recently been made to better understand the general structure of the unidentified compounds (Gentner et al., 2012; Liu et al., 2012).

In addition, some inorganic compounds (e.g. ammonium and nitrate) and a myriad of organic compounds (e.g. SVOCs and intermediate-volatility organic compounds) can move between the condensed (particle) phase and the gas phase.

Atmospheric chemistry and transport, and infiltration indoors, alter the exposure mixtures. Ozone, a major component of photochemical smog, is formed in the atmosphere due to reactions of NO_x with VOCs and CO. Sulfate PM is derived primarily from the oxidation of SO_2 (a gas), followed by gas-to-particle conversion. Although formaldehyde is emitted from some building materials and chemical facilities, more is formed in the atmosphere from the oxidation of other organic compounds, and is also destroyed by further reactions. A potentially important, but less well understood, set of chemistry involves larger organic molecules. Oxidation of gaseous organic molecules can lower their vapour pressure, leading to condensation, and they thus become part of the OC mixture in PM. There can be multiple generations of reactions, and some reactions can also take place in or on the particles themselves.

(c) Role of sources in the composition of outdoor air pollutants

Here, an overview is provided of the information described in detail for each pollutant in Section 1.2, focusing on sources.

Fossil fuel combustion leads to elevated levels of NO_x, VOCs, and carbonaceous PM. Reactions involving these compounds can further contribute to elevated levels of other compounds, such as ozone. Furthermore, any impurities in the fuel, for example sulfur and metals such as mercury, are also emitted, sometimes as a gas (e.g. SO_2) or as PM (e.g. fly ash, including metals such as selenium, vanadium, and nickel) or as both (e.g. mercury). The combustion process is complex, leading to a mixture of organic gases and PM that is just as complex, comprising species that were originally present in the fuel (e.g. benzene) and products of incomplete combustion. Some products of incomplete combustion include partially oxidized organic components (e.g. aldehydes), pyrolysis products (e.g. 1,3-butadiene and carbonaceous aerosol), and more oxidized products such as CO and carbon dioxide (CO_2) and PAHs. Some of these organic species of concern, including dioxins, quinones, and PAHs, are known or suspected carcinogens (see Table 1.2). The relative abundance of the constituents depends on the fuel type and combustion conditions. Historically, spark-ignition engines typically produced relatively less PM and NO_x than diesel, but more CO and VOCs. The carbonaceous PM from combustion is particularly complex (for detailed information, see HEI, 2013 and IARC, 2013a). Not all automobile emissions are from the exhaust pipe; brake and tyre wear can lead to copper and asbestos emissions, as well as resuspended dust. Biomass fuel combustion leads to emissions that can be similar to those from fossil fuel combustion, at least at the macroscopic level. Typically biomass combustion in open burning and in stoves takes place at lower temperatures, leading to lower NO_x emissions but higher CO emissions. Organic gases are emitted, as is carbonaceous PM, along with impurities in the fuel (e.g. potassium). The major differences are at the molecular level. Chemical plants and other industries have been responsible for elevated levels of additional pollutants, including heavy metals and organic air toxics, and the compounds involved are process-specific. The air toxics may be due to leakage in the plant or from incomplete combustion of flaring used to control emissions.

Natural processes also play a role. Wildland fires, natural and human-related, lead to emissions of CO, NO, and organic gases (including air toxics) and PM. Lightning forms NO_x and ozone. Sea spray generates PM. Volcanoes emit sulfur oxides (SO_x), mercury, and other metals. Wind raises dust. Plants emit organic gases, some of which are highly reactive. Microbial activity leads to emissions of NO_x and ammonia, as well as bioaerosols. Biogenically emitted VOCs and NO_x react to form ozone and organic PM. In many cases, natural and anthropogenic sources, such as natural VOCs and emissions of NO_x and SO_x from fossil fuel combustion, interact to produce higher pollutant levels than would be present with either type alone. Together, these sources and the related atmospheric processing lead to air pollutant mixtures of tremendous complexity and variation, although at any one time most of the pollutants are present, just at varying levels.

Carbonaceous PM is a major component of outdoor PM in general, and it will take on a larger role as the levels of other components of PM are reduced. Characterizing the components of primary and secondary carbonaceous PM is difficult, and even specialized studies have typically identified and quantified a relatively small fraction of the potentially thousands of components (Schauer et al., 1996). Therefore, carbonaceous PM is often classified more simply as EC and OC. To the extent that they have been characterized, carbonaceous PM emissions typically resemble the components in the fuel they are derived from (and, in the case of internal combustion engines, the lubricating oil), along with pyrolysis products. OC from internal combustion engines includes PAHs, hopanes, steranes, and partially oxidized products of the underlying fuels and lubricant (Zheng et al., 2002; Gentner et al., 2012; Isaacman et al., 2012; Liu et al., 2012; Zhao et al., 2013). OC from biomass combustion contains large amounts of levoglucosan, a pyrolysis product of cellulose combustion. Atmospheric processing increases the complexity of the OC mixture. Secondary organic compounds are formed in part from the oxidation of gaseous organic compounds, which then have a lowered vapour pressure, leading to condensation. Organic compounds that were originally emitted as PM can volatilize, react, and then recondense. As discussed below, recent advances in analytical methods are leading to a more detailed understanding of both emitted and outdoor OC at the molecular level (Gentner et al., 2012; Isaacman et al., 2012).

(d) Role of spatial scales of pollutants

Concentrations of outdoor air pollutants vary across microenvironments, depending on source characteristics. A typical urban area is affected by the surrounding regional background pollutants, which have evolved from a variety of processes (e.g. chemistry, dispersion, and deposition, along with emissions from natural processes). Pollutant concentrations in a city increase due to the variety of urban emissions characteristic of populated areas, leading to elevated levels of primary and processed pollutants on spatial scales similar to the size of the city (1 km to tens of kilometres). On top of the urban mixture, locally elevated levels of freshly emitted pollutants occur over smaller scales (0 m to hundreds of metres). Specific locations that experience high levels of air pollution include sites near and on roadways (including in vehicles), in fire plumes, and near chemical facilities. Karner et al. (2010) assessed near-roadway pollutant gradients and found that CO concentrations dropped by 90% to near background levels in about 170 m, and that concentrations of most other roadway-emitted species dropped to near background levels by 570 m. There are also locally large exposure gradients around factories and industrial complexes, although such gradients can be quite complex, depending on source characteristics. In contrast, plumes from power plants and fires, although they are diluted, can still be identified for tens to thousands of kilometres (Ryerson et al., 1998; Forster et al., 2001). Secondary pollutants (e.g. ozone, sulfate, and part of the OC) are found regionally with relatively smaller gradients. Dust has impacts at multiple scales; locally generated dust can have large impacts locally, and dust storms can lead to intercontinental transport.

For pollutants generated outdoors, concentrations may be lower when the pollutants are transported indoors. However, since people tend to spend most of their time indoors, most of the exposure to those pollutants occurs indoors. Also, indoor environments can lead to unique exposures as pollutants generated outdoors come into a very different environment, allowing new chemical pathways to occur. Studies have quantified exposures to specific chemicals or classes of chemicals across environments.

1.1.2 Pollutant concentrations

A better perspective on the potential impacts of air pollutants and air pollution is gained by considering typical levels of the major pollutants; more detailed descriptions of concentrations across regions are given in Section 1.4.

For primary pollutants, urban concentrations can be many hundreds of times background levels; ozone levels do not vary as dramatically. Approximate concentration ranges are given because pollutant concentrations vary by orders of magnitude between urban areas and within an urban area, depending on the location or day (or time of day). Although many measurements target a specific agent, the concentrations can be an indicator of a mixture and the presence of other compounds.

Ozone is produced naturally, and pre-industrial levels are estimated to have been approximately 30 μg/m³. Background levels are now about twice that, due to worldwide anthropogenic emissions of NO_x and VOCs, along with natural emissions. Because ozone is produced photochemically, levels are typically highest during sunny periods with reduced atmospheric dispersion. In urban areas with photochemical pollution, ozone levels have reached much higher levels (likely > 1000 μg/m³ in Los Angeles in the 1970s), but current levels in urban areas are much lower (e.g. summertime peaks of < ~400 μg/m³). Ozone reacts with NO, so in areas where NO_x emissions are high and/or photochemical activity is low, ozone levels are depressed and can be well below background levels. When elevated levels of ozone are found in urban areas, levels of other photochemical oxidants are likely to be elevated as well, including aldehydes, organic acids, organonitrates, inorganic acids, hydrogen peroxide, and photochemically produced PM (typically in the fine fraction) (Finlayson-Pitts & Pitts, 2000a, 2000b, and references therein).

PM levels vary dramatically and depend on which size fraction is being considered (Seinfeld & Pandis, 2006, and references therein). PM_{10} levels are higher than $PM_{2.5}$ levels because that size range (PM_{10}) includes the particles between 2.5 μm and 10 μm in diameter, in addition to particles with diameters smaller than 2.5 μm. The ratio between the two is location- and time-dependent. For 21 regions analysed worldwide in 2005, Brauer et al. (2012) estimated the ratio of annual average levels ($PM_{2.5}/PM_{10}$) to range from 0.13 to 0.94. Areas with high levels of dust or sea salt will have a considerably higher fraction of coarse PM, but if the PM is derived from combustion emissions or atmospheric reactions, the $PM_{2.5}$ can be a large proportion of the PM_{10}. In pristine areas or after rainstorms, $PM_{2.5}$ levels can be below 1 µg/m³. In more typical urban atmospheres, annual average levels of $PM_{2.5}$ are on the order of 4 µg/m³ to tens of micrograms per cubic metre (Brauer et al., 2012; Cooper et al., 2012). Background levels of SO_2 are very low, less than 1 µg/m³, except near natural sources (Finlayson-Pitts & Pitts, 2000b). In urban areas or near point sources, however, SO_2 levels can exceed tens of micrograms per cubic metre.

OC is present in urban atmospheres due to direct emissions and photochemical production. Levels are typically on the order of 1–10 µg/m³ but can be higher during stagnation events. Levels of the individual organic compounds that comprise OC are much lower, reflecting the hundreds of substances likely to be present. EC levels are typically less than OC levels, by about a factor of 2 or more, depending on the source distributions and photochemical activity. OC and EC levels are often highly correlated, because they share some of the same sources, and can also be correlated with levels of CO and NO_x (e.g. HEI, 2013).

Background CO levels are on the order of 50 µg/m³, varying both spatially and temporally (Seinfeld & Pandis, 2006). In urban areas with high levels of spark-ignition engine emissions, levels are about 10 times background levels and can be as high as 1000 or more times background levels during adverse meteorological conditions near sources. Concentrations tend to peak in cooler periods, when dispersion is reduced and cold-start emissions from vehicles are increased. CO levels in urban areas in developed countries have dropped dramatically due to automotive emission controls.

Average lead concentrations have decreased dramatically in countries where regulations have reduced the lead content in on-road motor vehicle gasoline. In the USA, lead concentrations in cities dropped from more than 5 µg/m³ in the early 1980s to about 0.1 µg/m³ after the use of leaded fuel was discontinued (EPA, 2010). Elevated lead concentrations are still found where leaded fuel is used and around lead processing facilities.

NO_x are emitted naturally by combustion and from microbial activity, leading to levels of 0.02–10 µg/m³. Urban concentrations can reach more than 1000 µg/m³, although they are typically more on the order of 10–100 µg/m³ (Seinfeld & Pandis, 2006).

A large number of other potentially hazardous air pollutants (HAPs; sometimes referred to as air toxics) are found in the atmosphere. Different organizations maintain different lists of air toxics. In many cases these pollutants are generally associated with specific source types and are found at elevated levels in limited geographical areas around point sources; exceptions include pollutants such as 1,3-butadiene from engines, PAHs from fossil and biomass fuel combustion, and mercury, which is long-lived and is deposited and re-emitted (UNEP, 2008). Special studies have produced information on potential levels of exposures. Levels of many of the compounds can be found in Seinfeld & Pandis (2006) and references therein, and are discussed below.

1.1.3 Pollutants classified by IARC

Outdoor air contains many substances that have been evaluated by IARC in the past (Table 1.2). The concentrations of these agents in the atmosphere are often very low, much lower than the levels that are found in environments where past epidemiological studies linking the substance to cancer may have occurred. Recently, IARC classified diesel engine exhaust in Group 1 and gasoline engine exhaust in Group 2B (IARC, 2013a). These are both significant components of urban air pollution mixtures, and include PM and other compounds.

Table 1.2 Agents in outdoor air that are established or probable IARC carcinogens[a]

Agent	CAS no.	Evaluation	Volume (reference)
Metals and fibres			
Arsenic and inorganic arsenic compounds	7440-38-2	1	100C (IARC, 2012a)
Asbestos		1	100C (IARC, 2012a)
Beryllium and beryllium compounds	7440-41-7	1	100C (IARC, 2012a)
Cadmium and cadmium compounds	7440-43-9	1	100C (IARC, 2012a)
Chromium (VI)	18540-29-9	1	100C (IARC, 2012a)
Lead compounds, inorganic/organic		2A/3	87 (IARC, 2006)
Nickel, metallic/compounds		2B/1	100C (IARC, 2012a)
Silica dust		1	100C (IARC, 2012a)
Organic chemicals			
1,3-Butadiene	106-99-0	1	100F (IARC, 2012b)
Benzene	71-43-2	1	100F (IARC, 2012b)
Ethylene oxide	75-21-8	1	100F (IARC, 2012b)
Formaldehyde	50-00-0	1	100F (IARC, 2012b)
Halogenated chemicals			
Ethylene dibromide	106-93-4	2A	71 (IARC, 1999)
2,3,7,8-Tetrachlorodibenzo-*para*-dioxin	1746-01-6	1	100F (IARC, 2012b)
Tetrachloroethylene	127-18-4	2A	106 (IARC, 2014a)
Trichloroethylene	79-01-6	1	106 (IARC, 2014a)
1,2,3-Trichloropropane	96-18-4	2A	63 (IARC, 1995)
Vinyl bromide	593-60-2	2A	97 (IARC, 2008)
Vinyl chloride	75-01-4	1	100F (IARC, 2012b)
Vinyl fluoride	75-02-5	2A	97 (IARC, 2008)
Polycyclic aromatic hydrocarbons			
Benzo[*a*]pyrene	50-32-8	1	100F (IARC, 2012b)
Cyclopenta[*cd*]pyrene	27208-37-3	2A	92 (IARC, 2010a)
Dibenz[*a*,*h*]anthracene	53-70-3	2A	92 (IARC, 2010a)
6-Nitrochrysene	7496-02-8	2A	105 (IARC, 2013a)
-Nitropyrene	5522-43-0	2A	105 (IARC, 2013a)
2-Nitrotoluene	88-72-2	2A	101 (IARC, 2013b)
Mixtures			
Biomass fuel (primarily wood), indoor emissions from household combustion of		2A	95 (IARC, 2010b)
Coal, indoor emissions from household combustion of		1	100E (IARC, 2012c)
Coal tar pitch	65996-93-2	1	100F (IARC, 2012b)
Coke production		1	100F (IARC, 2012b)
Creosotes	8001-58-9	2A	92 (IARC, 2010a)
Diesel engine exhaust		1	105 (IARC, 2013a)
Frying, emissions from high-temperature		2A	95 (IARC, 2010b)
Mineral oils, untreated or mildly treated		1	100F (IARC, 2012b)
Polychlorinated biphenyls	1336-36-3	1	107 (IARC, 2014b)
Polybrominated biphenyls	59536-65-1	2A	107 (IARC, 2014b)
Tobacco smoke, second-hand		1	100E (IARC, 2012c)
Wood dust		1	100C (IARC, 2012a)

[a] Established or probably carcinogens include Group 1 and Group 2A. The Working Group noted that many agents in Group 2B are also detected in outdoor air, such as gasoline engine exhaust, several individual polycyclic aromatic hydrocarbons, and acetaldehyde.
Prepared by the Working Group.

1.2 Sources of air pollutants

1.2.1 Introduction

Although there are hundreds of sources of outdoor air pollution, the source categories that are the largest contributors to most air pollutants in many locations are: vehicle emissions; stationary power generation; other industrial and agricultural emissions; residential heating and cooking; re-emission from terrestrial and aquatic surfaces; the manufacturing, distribution, and use of chemicals; and natural processes (Unger et al., 2010). Given the large differences in the number and density of these sources as well as in their design, fuel source, and effectiveness of emission control technology, the relative contribution of these sources to air pollution concentrations and exposures varies considerably across locations.

Daily, weekly, and seasonal changes in source activity, as well as meteorological factors, can also lead to very large changes in the temporal trends in atmospheric pollutant concentrations and the relative contributions from different sources.

Sources of air pollutants can be divided into several types. These can be helpful in understanding the spatial and temporal distribution of source emissions, which has a large impact on exposures to emissions from different sources. Sources are commonly classified into three broad groups: primary, secondary, and re-emission sources. A primary source results from the direct emissions from an air pollution source. In contrast, a secondary source results from the formation of a pollutant in the atmosphere from the chemical reaction of precursors emitted from air pollution sources. Finally, a re-emission source results from primary or secondary pollutants depositing on the Earth's terrestrial or aquatic surfaces, followed by re-emission to the atmosphere.

Not all pollutants fall exclusively into one group, but in many locations, the classification of a pollutant into these categories can provide insight into exposure gradients. Secondary and re-emission sources tend to have smaller temporal and spatial concentration gradients than primary sources, due to the physical processes controlling their emissions. Primary sources can be further subdivided into point sources, mobile sources, and area sources. Point sources' emissions are from emissions stacks and tend to lead to very large spatial and temporal gradients in concentration. Mobile sources are associated with transportation and tend to have large spatial gradients close to roadways but tend to be more homogeneous away from roadways in urban areas. Area sources are sources with relatively dispersed emissions over large areas and lead to relatively constant source contributions over space but can have very large temporal changes in emissions. In addition, fugitive sources, including VOCs and dust, result from the leakage of gases from storage and handling facilities and the resuspension of dust, respectively. The nature of these source categories leads to source contributions and exposures that can be parameterized with physical and statistical models to represent pollutant concentrations, given knowledge of emission factors.

Estimates of the source contribution to pollutant concentrations in the atmosphere and to exposures can be obtained with transport models, receptor models, or hybrid models that integrate aspects of transport models and receptor models. Transport models use emissions inventories along with mathematical representations of wind speed and direction to estimate pollutant concentrations over time and space. Receptor models use measurements of pollutants at a given location or from personal exposure measurements to elucidate the sources of the pollutants (EPA, 2014; European Commission, 2014). Reasonable confidence in source apportionment models usually requires agreement between transport and receptor models, but this is not always achieved if the applied models are not adequately developed.

In locations or scenarios where transport and receptor models have not been developed, the use of emissions inventories and source-specific intake fractions can provide reasonable estimates of exposures and the sources of the exposures.

Table 1.3 provides a global anthropogenic emissions inventory of key global pollutants by sector in 2000. On a global average, the power and industry sectors were the two major anthropogenic sources of SO_2 emission. These two sectors together with biomass burning and on-road transportation also contributed greatly to NO_x emission. Biomass burning, household biofuel, on-road transportation, and industry were the most important sources of carbonaceous emissions, including CO, BC, OC, and VOCs (Unger et al., 2010). It is important to note that the relative source contribution and absolute source contribution to these pollutants vary considerably across different regions of the world, across urban areas, and across seasons.

1.2.2 Photochemical oxidants

Photochemical oxidants are secondary pollutants that are formed during photochemical reactions in the atmosphere. These oxidants have short lifetimes but are continuously formed and destroyed through chemical reactions, leading to pseudo-steady-state concentrations that are important for chemical processing and can be inhaled. These oxidants include ozone, hydrogen peroxide, acids, peroxyacetyl nitrate, and reactive radicals. The reactive radicals, which include hydroxyl radical, oxygen radical, hydrogen radical, and several other radicals, have very short lifetimes and are not commonly measured (Finlayson-Pitts & Pitts, 2000a). A large number of VOCs, SVOCs, and non-volatile organic compounds are also produced in photochemical smog, and some are oxidants (see Section 1.2.10). Ozone is often used as an indicator for these oxidant compounds.

Photochemical oxidants are formed in the presence of sunlight from the chemical reactions of VOCs and NO_x. A more detailed discussion of the sources of NO_x and VOCs is presented in Sections 1.2.6 and 1.2.10, respectively.

Given the nonlinear response of ozone production from the reaction of VOCs and NO_x, the relative source contributions to ozone cannot be directly scaled from the relative source contributions to VOCs and NO_x. Chemical transport models are needed to apportion the incremental ozone to sources (Cohan et al., 2005).

1.2.3 Particulate matter

The size of atmospheric particles can be related to their sources, due to the physical processes that form atmospheric particles and the atmospheric processes that control the fate and evolution of particle size distributions in the atmosphere.

Coarse PM (particles with aerodynamic diameters between 2.5 µm and 10 µm) is generated largely by physical processes, including resuspension of soil and road dust, sea spray, agricultural tilling, vehicular abrasion (i.e. tyre and brake wear), and fugitive dust emission from industrial sources.

Accumulation mode particles (particles with diameters between 0.2 µm and 2.5 µm) comprise predominantly the condensation of secondary inorganic and organic compounds and coagulated nuclei mode particles (particles with diameters < 0.2 µm). These particles comprise predominantly secondary sulfate and bisulfate ion, secondary nitrate ion, secondary ammonium ion, and carbonaceous PM from primary and secondary sources, but also include some crustal materials due to the fact that accumulation mode particles include supermicrometre particles.

Nuclei mode particles originate predominantly from combustion sources and atmospheric nucleation. They have relatively short

Table 1.3 Global anthropogenic emissions inventory of air pollutants by sector in 2000

Sector	NO_x[b]	CO[a]	NMVOCs[a]	SO_2[a]	BC[c]	OC[c]	CH_4[a]	NH_3[b]	N_2O[a]	CO_2[a]
Industry	6.0	51	33.6	63.2	769	2559	2.7	0.2	0.7	8414
Power	7.8	12	33.3	57.7	22	18	93.9	0.1	0.1	9127
Household fossil fuel	0.9	27	1.2	8.1	453	486	1.7	2.2	0.02	3390
Household biofuel	2.2	237	27.3	3.1	1471	7823	13.8	0	0.2	495
On-road transportation	8.7	186	33.8	3.7	1235	1630	0.9	0	0.1	4276
Off-road (land) transportation	1.8	13	4.6	2.0	588	292	0.008	0	0.003	390
Shipping	2.9	0.1	0.02	7.3	97	136	0.028	0	0.003	428
Aviation	0.7	0	0	0.2	11	0	0.006	0	0.020	654
Agricultural waste burning	0.2	16	2.0	0.2	371	2266	0.8	1.4	0.020	0
Waste/landfill	0.04	4	2.7	0.05	0	0	58.2	2.7	0.3	0
Biomass burning	10.2	507	31.3	2.7	3500	37 200	21.2	1.8	0.9	2740
Animals	0	0	0	0	0	0	88.5	21.1	3.2	0
Agriculture	0	0	0	0	0	0	39.4	12.6	6.6	0

BC, black carbon; CH_4, methane; CO, carbon monoxide; CO_2, carbon dioxide; N_2O, nitrous oxide; NH_3, ammonia; NMVOCs, non-methane volatile organic compounds; NO_x, nitrogen oxides; OC, organic carbon; SO_2, sulfur dioxide.

[a] Expressed in teragram (Tg) full molecular mass/year

[b] Expressed in teragram (Tg) nitrogen/year

[c] Expressed in gigagram (Gg) full molecular mass/year

Adapted from Unger et al. (2010). Attribution of climate forcing to economic sectors. *Proc Natl Acad Sci U S A*, 107(8):3382–7. doi: 10.1073/pnas.0906548107 PMID:20133724, with permission from PNAS.

atmospheric lifetimes before they either grow to become accumulation particles or coagulate to form accumulation particles. Nuclei mode particles tend to be enriched in carbonaceous aerosols and metals from the combustion of heavy oil and fuel as well as emissions from the high-temperature processing of metals.

Coarse PM comprises predominantly inorganic crustal materials, abrasion particles from mobile sources and industrial sources, and sea spray.

It should be noted that $PM_{2.5}$ includes nuclei mode particles and accumulation mode particles and PM_{10} includes nuclei mode particles, accumulation mode particles, and coarse particles (Watson, 2002).

Source apportionment efforts for PM have typically been directed at source apportionment of particle mass; however, there are some studies that have been used to apportion the sources of components of PM (Querol et al., 2007; Heo et al., 2013).

Zhang et al. (2007) analysed the bulk composition of fine PM at more than 30 sites in the Northern Hemisphere, including urban, rural, and remote locations. They found that organic compounds accounted for 18–70% of the PM mass, sulfate ion accounted for 10–67%, nitrate ion accounted for a few percent to 28%, and ammonium ion accounted for 7–19% of the PM mass. EC and crustal materials are also important contributors to fine PM in the context of human exposure and health. Crustal material typically contributes 5–20% to $PM_{2.5}$ in most locations in Europe and the USA (Chow & Watson 2002; Belis et al., 2013), and EC usually contributes about 5–10% of the fine PM mass. Although $PM_{2.5}$ levels in China are much higher than those in cities in North America and Europe, the relative composition in megacities in China is similar (Chan & Yao, 2008; Cao et al., 2012). In addition, sea spray and road salt (used in cold climates to melt snow and ice on roadways) can account for up to 5–10% of fine PM mass (Chow & Watson 2002; Belis et al., 2013).

Sulfate ion in fine and ultrafine PM is predominantly from the oxidation of SO_2, which is largely from the combustion without emission controls of sulfur-containing fossil fuels. More information on the sources of SO_2 is provided in Section 1.2.4.

The contribution of nitrate ion and ammonium ion to fine PM is influenced by the fact that the two major forms of nitrate ion – nitric acid and ammonium nitrate – are semivolatile compounds, which can exist in both the gas phase and the particle phase. Atmospheric chemistry, temperature, and humidity control the rate of NO_x conversion to nitric acid. Further details are given in Section 1.2.6

The sources of carbonaceous fine PM have been a large area of research over the past decade, and the tools to understand the contribution of primary sources of carbonaceous PM and the split between primary and secondary organic aerosols are quite advanced and show reasonably good agreement (Docherty et al., 2008; Snyder et al., 2009a; Zhang et al., 2009a; Heo et al., 2013). In contrast, it is still difficult to quantify the specific sources of secondary organic aerosols at this time. The primary sources of fine particle organic aerosols are dominated by combustion sources, including gasoline-powered engines, diesel-powered engines, coal and residual oil combustion, biomass burning, and food cooking operations (Schauer et al., 1996; Bond et al., 2004). As previously noted, the distribution of sources and their fuels, operations, and degree of emission controls can have a very large impact on their relative contributions to primary organic aerosols, which can be dominated by mobile sources in cities such as Los Angeles (USA), Tel Aviv (Israel), Amman (Jordan), and Mexico City (Mexico) (Stone et al., 2008; von Schneidemesser et al., 2010; Heo et al., 2013), by biomass burning in locations such as Kathmandu (Nepal) and rural North Carolina (USA) (Sheesley et al., 2007; Stone et al., 2010), or by multiple combustion sources in locations such as Beijing (China) (Zheng et al., 2005).

EC emissions are mainly in the submicrometre range, and the contribution of EC to atmospheric PM is largely in the $PM_{2.5}$ fraction. EC is mainly from pyrolysis during combustion from sources including coal combustion, fuel oil combustion, diesel engines, poorly operating gasoline engines, and biomass burning. As PM controls are being placed on most stationary power generation sources, as well as diesel engines, in Europe, the USA, and Canada, the concentrations of EC in these locations continue to decrease. In regions of the world where diesel engine emissions are not being controlled and there are large primary emissions from residual fuel and solid fuel combustion, these sources dominate contributions to EC.

Source contributions to PM_{10} can be represented as the sum of source contributions to fine PM plus source contributions to coarse PM. In the Los Angeles Basin (USA), coarse PM was found to have average contributions of about 50% from crustal material, 20% from secondary inorganic ions, 20% from OC, and 10% from sea spray (Cheung et al., 2011). Similar results were observed in the United Kingdom (Yin & Harrison, 2008). In locations affected by dust storms, the dust contributions to coarse PM and fine PM can be significantly larger in terms of concentrations and relative contribution.

Emissions inventory data can provide an assessment of sources of primary emissions of PM on a global or local scale (Bond et al., 2004; Corbett et al., 2007).

1.2.4 Sulfur dioxide

Natural sources of SO_2 include the atmospheric oxidation of sulfur compounds emitted from microbial activity in the ocean and from the anaerobic degradation of organic material in terrestrial environments. In some locations, such as Mexico City and parts of Japan, SO_2 emissions from volcanoes also affect urban areas and SO_2 exposures (de Foy et al., 2009; Kitayama et al., 2010).

However, in most locations in the world that are influenced by anthropogenic emissions, SO_2 emissions from natural sources are usually much lower than anthropogenic emissions. SO_2 in urban and industrialized areas is largely from the combustion without emission controls of sulfur-containing fuels and from uncontrolled metal processing facilities that roast sulfide ores to make metal oxides. Emissions inventories can provide a good understanding of the sources of SO_2, given the ability to accurately estimate sulfur contents of fuels (Bhanarkar et al., 2005; Smith et al., 2011; Ozkurt et al., 2013). Many countries have adopted regulations and technologies to reduce sulfur levels in gasoline and diesel fuels; however, there are still a large number of countries around the world that do not have good controls for SO_2 emissions and have not reduced sulfur levels in mobile-source fuels. Historically, there have been petroleum refining and coal liquefaction facilities that have removed sulfur during fuel processing and emitted it as SO_2 directly to the atmosphere. It is unclear whether such facilities are still operating, but they may be important sources in some local areas where adequate emission controls do not exist.

In addition, in some regions where coal is burned for residential heating and cooking, very high exposure to SO_2 can occur.

1.2.5 Carbon monoxide

The formation of CO is largely due to poor mixing of combustion air and combustion fuel, resulting in incomplete combustion. The dominant sources of outdoor concentrations of CO in urban areas are on-road transportation (gasoline- or diesel-powered engines) (IARC, 2013a), off-road engines, and biomass burning activity.

The use of catalytic convertors to convert emissions of CO to CO_2 for on-road gasoline-powered engines decreases CO emissions.

In rural areas and locations where biomass fuels are commonly used for residential cooking and heating, outdoor concentrations of CO are typically dominated by these biomass burning activities. Likewise, forest fires and controlled burns of vegetation can also be very large sources of CO.

Several global assessments of CO can be used to understand the regional distribution of CO sources using emissions inventory and chemical transport models (Holloway et al., 2000). On an urban scale, inverse models can be used to understand the local contributions of sources to CO (Bergamaschi et al., 2000).

1.2.6 Nitrogen oxides

Globally, the sources of NO_x are dominated by fossil fuel combustion, microbial activity in soils, and biomass burning, with smaller contributions from lightning and stratospheric oxidation of nitrous oxide (N_2O).

In urban areas, fossil fuel combustion is often the dominant source and includes stationary power generation, diesel-powered engines, and gasoline-powered engines. There has been some concern that diesel aftertreatment technologies aimed at reducing PM emissions will shift the distribution of NO_x emissions towards NO_2, which will lead to higher NO_2 exposures near roadways (Grice et al., 2009).

In rural areas where residential combustion of solid fuels is common, the residential combustion of solid fuels and microbial activity in soils are typically the dominant sources of NO_x.

Ammonia is a primary pollutant and on national scales is emitted largely as a result of agricultural practices, including direct emissions from livestock waste, emissions from spreading of manure, and emissions from the use of synthetic fertilizers (Battye et al., 2003). In urban areas, ammonia emissions are dominated by mobile-source emissions (Battye et al., 2003), which result from three-way catalytic converters over-reducing NO_x to ammonia (Fraser & Cass, 1998).

As part of the photochemical cycle, NO reacts with ozone to form NO_2, and NO_2 undergoes photolysis in the presence of sunlight to form NO. This photochemical cycle is a key component of ozone formation and the production of photochemical oxidants.

Chemical transport models that use emissions inventories have been very successful at modelling both near-roadway (Karner et al., 2010) and continental-scale NO_x concentrations (Stedman et al., 1997; Martin et al., 2003). Such models are an effective means of quantifying the sources of NO_x on different time scales for current and future scenarios.

1.2.7 Lead and other toxic metals

Non-volatile metals are components of atmospheric PM and can greatly influence its biological activity. Industrial sources can be very large sources of metals that can be found in atmospheric PM even though the metals are not major contributors to particle mass (Schauer et al., 2006; Snyder et al., 2009b). In the absence of industrial sources, roadway emissions and stationary power generation are typically the largest source of many toxic metals in the urban atmosphere. The braking systems of motor vehicles and underground public transportation emit metals that are potentially of concern for human exposure, including iron, copper, chromium, strontium, manganese, and antimony (Schauer et al., 2006; Kam et al., 2013). Stationary power generation that does not have suitable particle controls can have substantial impacts on metal concentrations and exposures. In locations where residual oils are used for heating and emission controls do not exist, very high concentration of nickel and vanadium can be found in atmospheric PM (Peltier et al., 2009). Likewise, coal fly ash can contain relatively high levels of arsenic, copper, chromium, zinc, antimony, selenium, and cadmium (Ratafia-Brown, 1994), and if the fly ash is not controlled with aftertreatment technologies, then emissions will contribute to an increased presence of toxic metals in the PM downwind of the facility. In developing countries, the uncontrolled emissions from brick kilns, waste incineration, and cement plants are important sources of metals to communities close to these facilities (Christian et al., 2010; Tian et al., 2012). There are very few comprehensive studies of the emissions inventory of fine particulate metals; Reff et al. (2009) provided an assessment of the spatially resolved emissions inventory for 10 metals classified as air toxics by the United States Environmental Protection Agency (US EPA) from 84 source categories.

1.2.8 Volatile metals, including mercury

Atmospheric mercury concentrations are largely dominated by gaseous elemental mercury (GEM), reactive gaseous mercury (RGM), and particulate mercury (Hg-P). RGM and Hg-P are formed in the atmosphere from the oxidation of GEM. Global anthropogenic emissions of mercury have been assessed by Pacyna et al. (2010). Globally, in 2005, burning of fossil fuel (mostly coal) was the largest single source of GEM emissions, accounting for about 45% of the anthropogenic emissions; artisanal/small-scale gold mining was responsible for about 18%, and industrial gold production accounted for 5–6%. Other mining and metal production activities and cement production were each responsible for about 10% of global anthropogenic releases to the atmosphere. The proportion of emissions from waste incineration and product-use sources is more difficult to estimate (Pacyna et al., 2010).

GEM is a global pollutant that has an atmospheric lifetime in the range of months to years. In most urban and rural outdoor locations, GEM levels are typically in the range of 2–10 ng/m^3, and the concentrations of RGM and Hg-P are typically in the range of tens to hundreds of pictograms per cubic metre. Local sources of GEM, including anthropogenic sources and re-emissions from terrestrial and aquatic surfaces, can increase local concentrations to 5–10 ng/m^3, or hundreds of nanograms per cubic metre near large mercury sources (Manolopoulos et al., 2007).

In addition to mercury, other volatile metals have been measured in the atmosphere, including alkyl-lead compounds (Wang et al., 1997), arsines and methyl arsines (Mestrot et al., 2009), and selenium compounds (Zhang et al., 2002).

1.2.9 Polycyclic aromatic hydrocarbons

Poor combustion conditions can lead to high emissions of PAHs and are often associated with liquid and solid fuel combustion. Benzo[*a*]pyrene (B[*a*]P) is a specific PAH formed mainly from the burning of organic material, such as wood, and from car exhaust fumes, especially from diesel vehicles. B[*a*]P pollution is predominantly a problem in countries where domestic coal and wood burning is common (EEA, 2013).

In 2007, it was estimated that the global total atmospheric emission of 16 PAHs came from residential/commercial biomass burning (60.5%), open-field biomass burning (agricultural waste burning, deforestation, and wildfire) (13.6%), and petroleum consumption by on-road motor vehicles (12.8%) (Shen et al., 2013).

1.2.10 Other organic compounds, including VOCs, SVOCs, and particulate organic matter

Thousands of organic compounds can be found in the atmosphere. They are components of fossil fuel, partially combusted components of fossil fuel, and pyrolysis products of fossil fuel; industrial chemical, food cooking, and biomass burning emissions; biogenic compounds emitted from plants; and organic compounds formed in the atmosphere (EEA, 2013; Oderbolz et al., 2013). These compounds include VOCs, non-volatile organic compounds that are present in atmospheric PM, and SVOCs that are present in both the gas phase and the particle phase. Many known or suspected carcinogens (Table 1.2) come from combustion sources; they include benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, and naphthalene (EPA, 2006). Industrial facilities and consumer products are also important sources of aromatic VOCs, oxygenated VOCs, and halogenated VOCs. These chemicals include benzene, toluene, xylenes, ethylbenzene, methyl ethyl ketone, acetophenone, and trichloroethylene. In addition, some VOCs of potential concern are also formed in the atmosphere from photochemical reactions; these include formaldehyde, acetaldehyde, and nitrobenzene. There is also a group of persistent organic pollutants (POPs), which include many SVOCs such as polychlorinated biphenyls, polybrominated biphenyls, furans, and dioxins, and several pesticides and insecticides that can be directly emitted from air pollution sources or re-emitted from previous contamination through volatilization or resuspension of soil material (EPA, 2006; EEA, 2013).

The three major sources of VOCs in Asia are stationary combustion, solvent and paint use, and transportation; the proportion of each of these sources varies between 25% and 50%, depending on the region (Kurokawa et al., 2013). In Europe, solvent and product use was reported to contribute to about half of the total VOC emissions; the contributions of three other major sources of VOCs – commercial, institutional, and household energy use; road transportation; and energy production – were 10–20% each (EEA, 2013). In the USA, the relative source contribution reported in 2008 by the US EPA was 50% for transportation and 20% each for solvent use and industrial processes (EPA, 2013d).

In recent years, significant progress in the development of emissions inventories has been made, including the current and future emissions of dioxins (Quass et al., 2004). To assess the sources of organic compounds that both are formed in the atmosphere and react in the atmosphere, such as formaldehyde, chemical transport models are needed (Zheng et al., 2005). Several integrated assessments of emissions inventories of toxic organic compounds have been conducted and are used to provide an integrated risk from these sources by source and receptor (George et al., 2011; Luo & Hendryx, 2011).

1.2.11 Mineral dust and fibres

Resuspended dust from roadways, agricultural lands, industrial sources, construction sites, and deserts is a major source of PM in many regions of the world. Roadway dust also contains metals associated with motor vehicles (Schauer et al., 2006). Agricultural soils often contain metals that accumulate from fertilizer and animal waste, and the content of dusts from industrial sources and construction sites will depend on the specific process activities occurring at those facilities.

Although fibres, such as asbestos, are not commonly measured in the outdoor atmosphere, they can be part of the atmospheric pollution mixture. The use of asbestos has been restricted or banned in many countries. However, outdoor air pollution with asbestos may still arise in some areas from releases from asbestos-containing building materials, asbestos brakes used on vehicles, and asbestos mining activity (IARC, 2012a).

1.2.12 Bioaerosols

Bioaerosols are part of the atmospheric PM. The term “bioaerosol” refers to airborne biological particles, such as bacterial cells, fungal spores, viruses, and pollens, and their products, such as endotoxins (Stetzenbach et al., 2004). A wide range of these biological materials have been measured in the outdoor atmosphere, including moulds, spores, endotoxins, viruses, bacteria, proteins, and DNA (Yeo & Kim, 2002). Knowledge about the dynamics and sources of airborne microbial populations is still scanty. Bioaerosols are believed to be ubiquitous, and studies demonstrate the long-range transport of microorganisms and biological particles in the atmosphere (Gandolfi et al., 2013). Bioaerosols may derive from many sources, for example plants, suspension of soils containing biological materials, cooking, and burning of biological materials.

1.3 Outdoor air pollution measurement methods

Measurements of gaseous and particle air pollutants have been used for exposure assessment and epidemiological studies. Methods include passive sampling (e.g. diffusion-based methods on an absorbent) and active sampling with pumps. Most methods for regulated gas pollutants (e.g. CO, NO_2, ozone, and SO_2) use in situ continuous monitors for hourly averaged concentrations. Airborne particles are sampled mostly using integrated sampling systems over a 24-hour period with defined inlets, sampler surfaces, filter substrates/holders, pumps, and flow controllers. Filter substrates are used to measure mass concentration by gravimetry. These substrates can be further analysed for their major components to explain the measured mass. Continuous monitoring for mass by β attenuation monitoring, an inertial balance, or particle light scattering (as a surrogate for PM mass) have been used at many central monitoring sites since the early 1980s. Continuous PM component measurements for precursor gases, elements, ions, and carbon along with particle number, size, and single particle measurement have been available since the late 1990s.

1.3.1 Overview

Table 1.4 (available online at: http://monographs.iarc.fr/ENG/Monographs/vol109/Table1.4-online.pdf) summarizes different measurement methods by pollutant, including relevant references that provide greater detail on measurement principles, practicality, measurement standards, detection limits, accuracy, precision, interferences, and intercomparability for different environments and costs. Table 1.4 (available online) also identifies opportunities for quantifying a wide range of pollutants beyond those that are currently regulated by ambient air quality standards (AAQS) (Chow & Watson, 2011; Billionnet et al., 2012; Pachon et al., 2012). The major categories in Table 1.4 (available online) are individual gases, multiple gases, PM for mass and chemical components, particle number and size, and mercury.

In Table 1.4, (available online) references associated with topic headings are more general reviews for that category, and more detailed treatments are associated with each method. The cited references are intended to inform about past measurement methods – for example, the British Smoke measurement of filter darkening dates from the 1920s (Hill, 1936; Brimblecombe, 1987) and has been used in retrospective exposure studies – as well as newly emerging technologies. A few critical reviews and textbooks provide broad descriptions of gas and particle measurements (Chow, 1995; Landsberger & Creatchman, 1999; Finlayson-Pitts & Pitts, 2000a; McMurry, 2000; Cohen & McCammon, 2001; Wilson et al., 2002; Clemitshaw, 2004; Fehsenfeld et al., 2004; Heard, 2006; Chow et al., 2008; Wexler & Johnston, 2008; Kulkarni et al., 2011; Chow & Watson, 2012), and detailed procedures for certain methods have been published by ASTM (2013), the US EPA (2013a, 2013b), ISO (2013), and the European Union (EU) (CEN, 2013).

1.3.2 Types of air quality monitors

Pollutants for which air quality standards have been established in many countries (called "criteria pollutants" under the statute defined by the US EPA, i.e. CO, NO_2, SO_2, ozone, $PM_{2.5}$ mass, PM_{10} mass, TSP, and lead) are monitored in populated areas to determine compliance with the AAQS, and these data can often be downloaded for specific study areas and time periods (e.g. EPA, 2013c). Gases are recorded continuously as hourly averages, and PM mass concentrations may be hourly or 24-hour averages.

Sampling sites are selected to represent neighbourhood (~1–5 km) to regional (100–1000 km) scales (EPA, 1997, 1998; Chow et al., 2002), although some are also located in near-road environments (~30–50 m) dominated by vehicle exhaust or in industrial fenceline environments. Monitoring methods and procedures are specified for compliance purposes, and the data are used to determine exceedances of the AAQS. Compliance data can be used as a first estimate of human exposure, but these can be supplemented with additional measurements in microscale (1–10 m) environments to obtain more representative exposure estimates.

Passive sampling is the most cost-effective approach for estimating spatial gradients around compliance monitors, surveying additional pollutants, identifying hotspots, and estimating individual exposure. However, passive samplers need to be co-located with calibrated gas and particle sampling systems at the central monitoring site to establish equivalence and comparability. Substrate passive samplers are simple, inexpensive, and unobtrusive and require no power (Kot-Wasik et al., 2007; Zabiegała et al., 2010).

Passive samplers can detect long-term averages to very low levels, depending on the environment. Diffusion tube and badge-type passive samplers are in most common use, and the references in Table 1.4 (available online) identify impregnants suitable for several gaseous pollutants and specific VOCs. A passive PM sampler coupled with microscopic analysis is also listed.

In active sampling, an air mover (e.g. pump or blower) draws air through a substrate or absorbing solution, pumps it into a container, or directs it into an in situ sensor. Accurate and precise flow rates and sample durations must be quantified (Watson et al., 2013).

The largest variety of compounds is obtained from laboratory analysis of the substrates or container contents, but this is at the expense of averaging times and labour to change samples. Some of this is compensated for with in situ continuous instruments, which collect and analyse the sample in the field, but this comes at an even higher cost.

The trend is towards microsensors that use battery-powered miniature pumps and can be placed in microenvironments or carried by an exposure subject (Marć et al., 2012; Moon et al., 2012; Steinle et al., 2013). Miniature samplers are in common use for PM and certain gases, and the detection limits of optical light scattering/absorption systems and electrochemical gas sensors have been improving.

Particle scattering by nephelometer with a $PM_{2.5}$ inlet and a smart heater to remove moisture under high relative humidity (e.g. > 65%) is often used as a surrogate for $PM_{2.5}$ mass (Chow et al., 2006; Watson et al., 2008).

Remote sensing measures the scattering and absorption of infrared, visible, and ultraviolet radiation at different wavelengths along a sight path. Path lengths may range from a few metres, used for in-plume monitoring, to thousands of kilometres for geostationary satellites (Hidy et al., 2009; Hoff & Christopher, 2009). Satellite remote sensing estimates for PM, NO_2, SO_2, and some other pollutants often correspond to urban and industrial areas, but spatial resolution is limited to about 10 km.

1.4 Environmental occurrence and human exposure

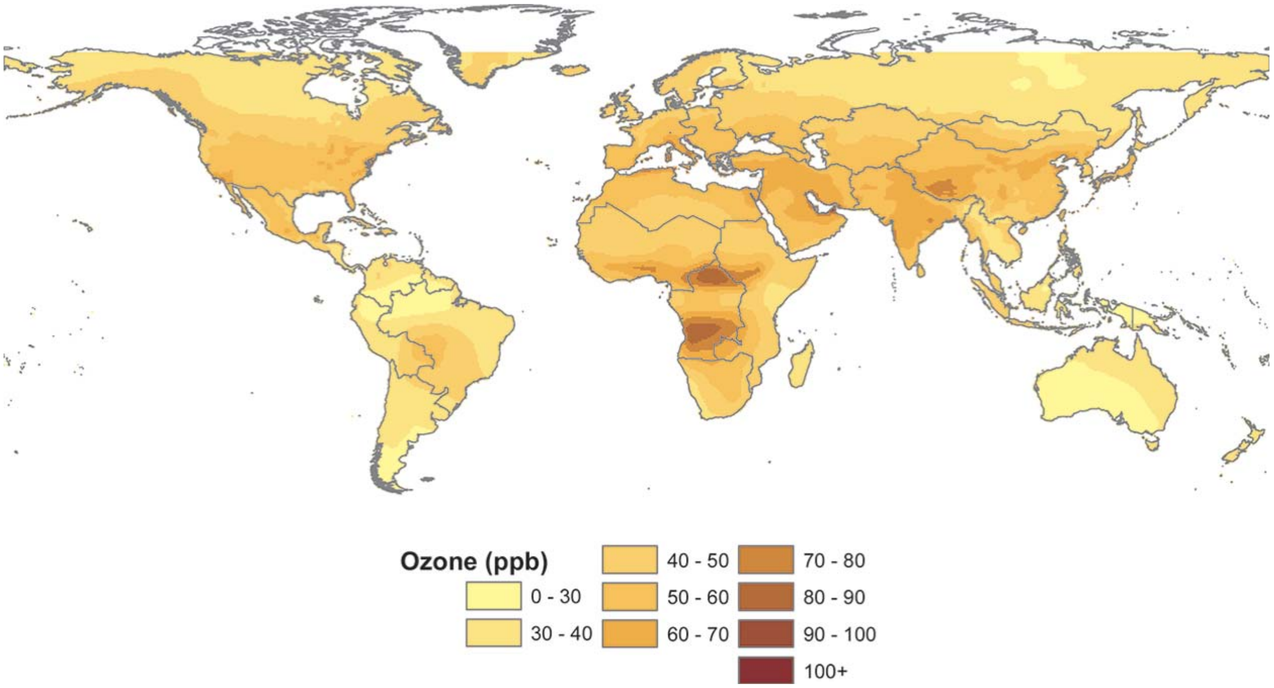
This section describes concentrations of air pollutants measured throughout the world. Because measurement approaches and methodologies differ by location, direct comparisons between countries of levels measured by ground-based monitoring should be made with caution. Furthermore, there are major differences in the overall availability of routine measurement data between countries. Although they are not available for all constituents of interest, satellite-based approaches provide estimates in a consistent manner for the entire globe and are therefore useful to identify spatial patterns (Lee et al., 2009; Brauer et al., 2012; Lamsal et al., 2013).

For ozone, a global chemical transport model simulation of seasonal maximum concentrations is available. The estimated levels of ozone are highest in North America, Latin America, Europe, and South and East Asia, as well as parts of Africa. For these regions, seasonal (3-month) hourly maximum ozone concentrations in 2005 were estimated to be greater than 40 ppb [80 μg/m³], with concentrations in some areas in parts of Asia and Africa greater than 80 ppb [160 μg/m³] (Fig. 1.2). As expected, given that ozone is a secondary pollutant, the spatial variability of the ozone concentration is less pronounced than that of $PM_{2.5}$, and levels are not as systematically higher in the rapidly developing countries of Asia (Brauer et al., 2012).

For $PM_{2.5}$, the concentration in 2005 was estimated to be high (> 50 μg/m³) in South and East Asia. Similarly high concentration estimates due to airborne mineral dust, rather than combustion emissions, were reported in North Africa, central Asia, and Saudi Arabia (Brauer et al., 2012; Fig. 1.3).

Global variation in NO_2 generally follows the spatial variation in combustion sources such as motor vehicle exhaust. Broad regional patterns of higher NO_2 concentrations correspond to

Fig. 1.2 Estimated seasonal (3-month) hourly maximum ozone concentrations (ppb) from a global chemical transport model (TM5) for 2005



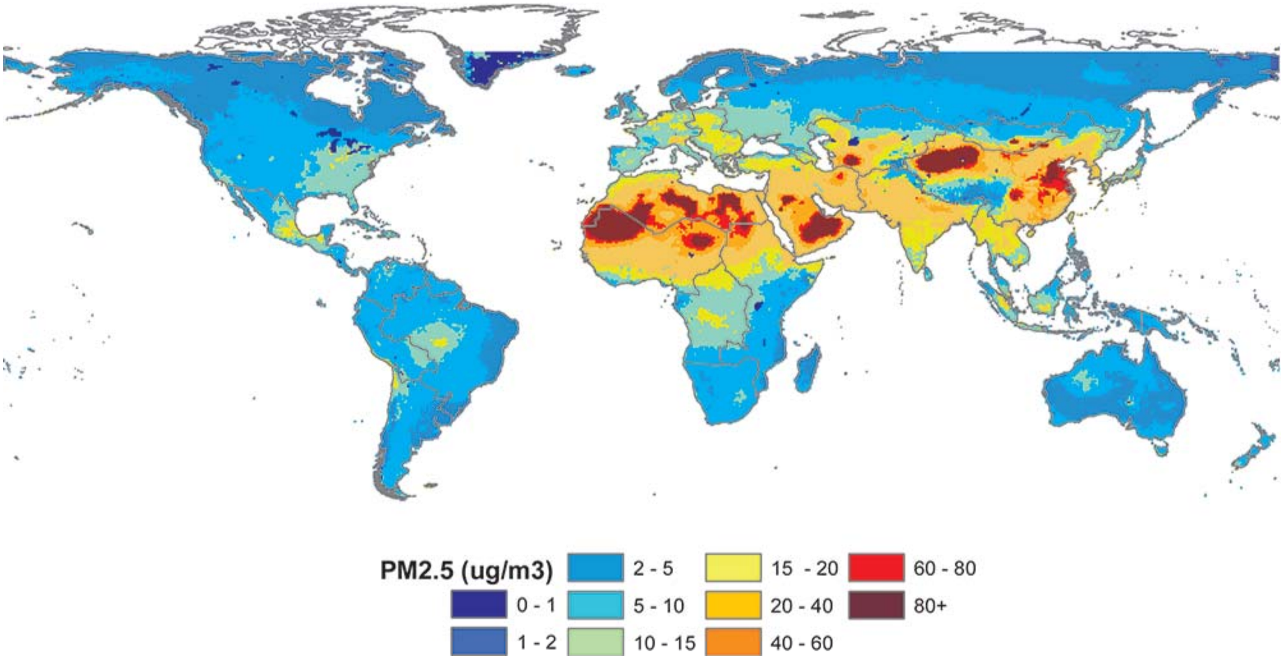
Ozone concentrations in µg/m³ = 2 × ozone concentrations in ppb.
Reprinted from Brauer et al. (2012). Exposure assessment for estimation of the global burden of disease attributable to outdoor air pollution. *Environ Sci Technol*, 46:652–660, with permission from the American Chemical Society. Copyright 2012, American Chemical Society.

population density, although absolute levels vary considerably according to economic development and air quality management programmes. In urban areas, lower concentrations are observed in cities in India (0.2–12 ppb [0.38–22.9 µg/m³]), substantially higher concentrations in cities in China (0.3–8 ppb [0.57–15.3 µg/m³]), and levels varying across this range for cities in the USA and Europe, reflecting differences in per capita fuel consumption. Globally, NO_2 concentrations increase in proportion to population raised to an exponent that varies by region (Lamsal et al., 2013).

Elevated SO_2 levels are observed over urban and industrial areas, especially in eastern China. Specific plumes related to volcanic activity are also evident in the satellite-based estimates. For example, the SO_2 plume from the Nyamuragira eruption in the Democratic Republic of the Congo can extend to South Asia (Lee et al., 2009).

High levels of formaldehyde are found in tropical regions in Africa and South America, where biogenic and biomass burning sources are important. High levels are also found in South-East Asia, resulting from biomass burning and anthropogenic sources. Seasonal variations in formaldehyde levels reflect increased biogenic and biomass burning emissions during summer in deciduous forests (mid-latitudes) and during the dry season in tropical forests (Amazon and Africa) (De Smedt et al., 2012).

Fig. 1.3 Estimated 2005 annual average $PM_{2.5}$ concentrations (μg/m³)



The $PM_{2.5}$ estimates are generated from the grid cell average of satellite-based estimates and chemical transport model (TM5) simulations that are calibrated with a prediction model incorporating surface measurements.
$PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm.
Reprinted from Brauer et al. (2012). Exposure assessment for estimation of the global burden of disease attributable to outdoor air pollution. *Environ Sci Technol*, 46:652–660, with permission from the American Chemical Society. Copyright 2012, American Chemical Society.

1.4.1 Outdoor pollutant concentrations

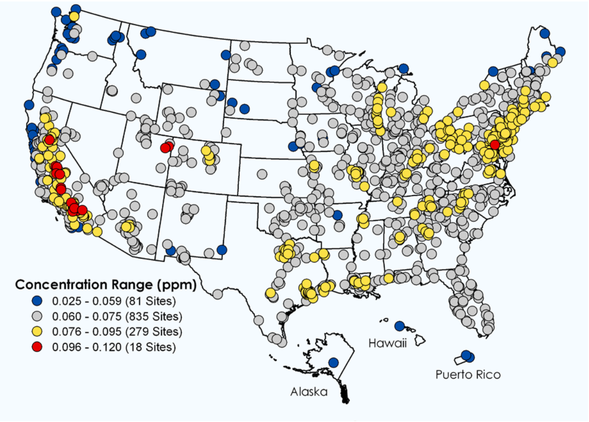
(a) North America (USA, Canada, and Mexico)

Measurements of air pollutant concentrations in North America at the country level are summarized in detail in this section. Differences in network composition, sampling and analysis methods, and available summary information hamper direct comparisons between countries. However, several global databases are available for a limited number of pollutants, which allow direct comparisons. The Global Burden of Disease Study 2010 provided estimates of $PM_{2.5}$ and ozone globally at about 10 × 10 km resolution, combining estimates from a chemical transport model, an approach using satellite retrievals of aerosol optical depth and a chemical transport model, and available measurements (Brauer et al., 2012). For 2005, the population-weighted annual mean $PM_{2.5}$ concentration in North America was estimated as 13 µg/m³, and the population-weighted seasonal 3-month hourly maximum ozone concentration was 57 ppb. Fig 1.2 and Fig. 1.3 present the estimated concentrations.

(i) USA

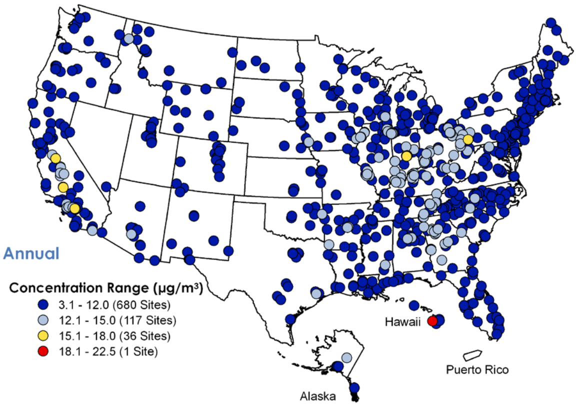
The US EPA collates a comprehensive database of outdoor air pollutant measurements conducted at about 3000 locations by state and local air quality monitoring agencies following Federal Reference Methods for the criteria air pollutants (ozone, NO_x/NO_2, SO_2, $PM_{2.5}$/PM_{10}, CO, and lead). This network (EPA, 2011a) was initiated in 1978, although monitoring approaches and specific pollutants that have been included have changed over time. At a subset of about 250 of these sites, several air toxics such as VOCs, metals in PM_{10}, and mercury are monitored (EPA, 2012a). This network is complemented by about

Fig. 1.4 Annual fourth-highest daily maximum 8-hour ozone concentrations in 2010 in the USA (applicable NAAQS is 0.075 ppm)



NAAQS, National Ambient Air Quality Standards.
Reprinted from EPA (2012c). Air quality monitoring information. Available from: http://www.epa.gov/airtrends/factbook.html.

Fig. 1.5 Annual average (98th percentile of 24-hour concentrations) $PM_{2.5}$ concentrations in 2010 in the USA

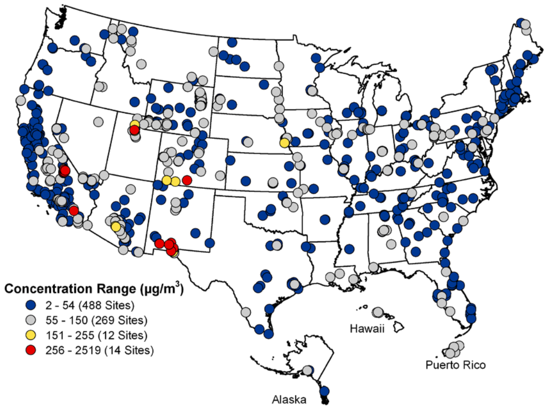


$PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm.
Reprinted from EPA (2012c). Air quality monitoring information. Available from: http://www.epa.gov/airtrends/factbook.html.

180 chemical speciation network monitoring sites (EPA, 2013e), where specific components of $PM_{2.5}$ are measured. Several smaller routine monitoring networks are also operated with specific objectives, for example the IMPROVE network to assess the impacts of air pollution on visibility in protected environments (IMPROVE, 2012). In addition, the National Air Toxics Trends Station (NATTS) Network (EPA, 2012b) provides long-term monitoring data for air toxics at 27 sites (20 urban, 7 rural) across the country.

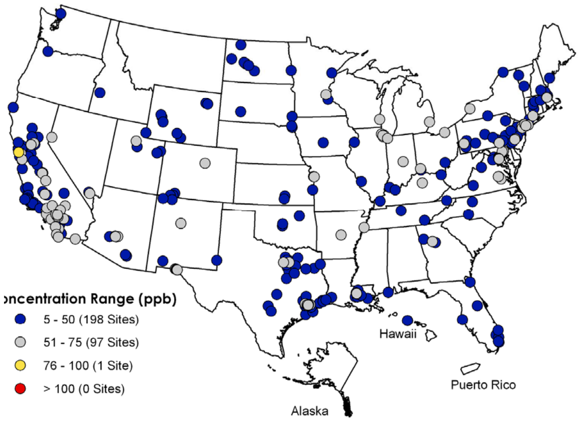
Regular status and trends reports provide information on concentrations of criteria and toxic air pollutants (EPA, 2012c). Summary information for 2010 is presented in Fig. 1.4, Fig. 1.5, Fig. 1.6, Fig. 1.7, and Fig. 1.8 and shows substantial variability in outdoor pollutant concentrations across the USA. The highest concentrations of ozone were observed in California as well as the Ohio River valley, the New England states, Texas, and several south-eastern states. Ozone levels are more heterogeneous over space, with most sites reporting levels below the National Ambient Air Quality Standards (NAAQS) of 75 ppb (annual fourth-highest daily maximum 8-hour concentration) (Fig. 1.4). Concentrations (annual average) of $PM_{2.5}$ were highest in California, Indiana, Pennsylvania, and Hawaii. Current levels of $PM_{2.5}$ (annual average) are below 15 µg/m³ at all but 6 (of > 700) reporting monitoring sites (Fig. 1.5). During winter periods, high concentrations of $PM_{2.5}$ were measured in regions where wood burning is prevalent, such as the Pacific Northwest and Alaska (EPA, 2012c). High PM_{10} concentrations were observed in California as well as Utah, Colorado, and New Mexico, especially in arid regions or industrial areas with multiple coarse particle sources (Fig. 1.6). NO_2 concentrations generally correspond to population density, with the highest concentrations observed in California, the Midwest, and the East Coast (Fig. 1.7). Annual average NO_2 levels have a range of 1–28 ppb, with a mean across 142 Metropolitan Statistical Areas of 10 ppb, well below the NAAQS of 53 ppb. To further describe spatial patterns at high resolution (30 m), Novotny et al. (2011) used a combination of satellite-based estimates and land-use

Fig. 1.6 Annual average (2nd highest maximum of 24-hour concentrations) PM_{10} concentrations in 2010 in the USA



PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm.
Reprinted from EPA (2012c). Air quality monitoring information. Available from: http://www.epa.gov/airtrends/factbook.html.

Fig. 1.7 NO_2 (98th percentile of 1-hour daily maximum) concentrations in 2010 in the USA



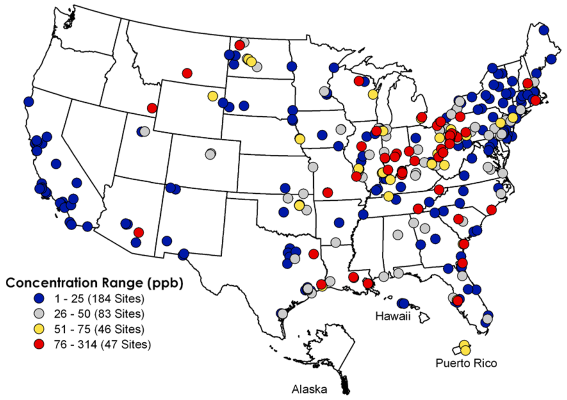
NO_2, nitrogen dioxide.
Reprinted from EPA (2012c). Air quality monitoring information. Available from: http://www.epa.gov/airtrends/factbook.html.

characteristics to model NO_2 across the USA. Using this approach, a population-weighted mean annual average concentration of 10.7 ppb was estimated. SO_2 concentrations are highest in the Upper Midwest and portions of the Northeast, where coal-fired power generation and industrial sources are common (Fig. 1.8). The 1-hour maximum SO_2 levels ranged from 0.1 ppb to 10.5 ppb, with a mean across 178 Metropolitan Statistical Areas of 2.4 ppb, well below the annual mean NAAQS of 30 ppb. Lead concentrations are much higher near stationary sources such as metals processing, battery manufacturing, and mining (~8 times the concentrations at sites not located near stationary sources) (Fig. 1.9). Levels of airborne lead (maximum 3-month average) are mostly below 0.07 µg/m³ (about half the level of the current NAAQS of 0.15 µg/m³), but levels as high as 1.4 µg/m³ have been measured at a subset of sites (EPA, 2012c).

For all of the criteria pollutants, concentrations have decreased over the past 10 years, after even larger decreases in earlier periods. $PM_{2.5}$ and PM_{10} concentrations show steady reductions that coincide with emissions reduction programmes (EPA, 2012c). Nationally, between 2001 and 2010, 24-hour $PM_{2.5}$ and PM_{10} concentrations declined by 28% and 29%, respectively.

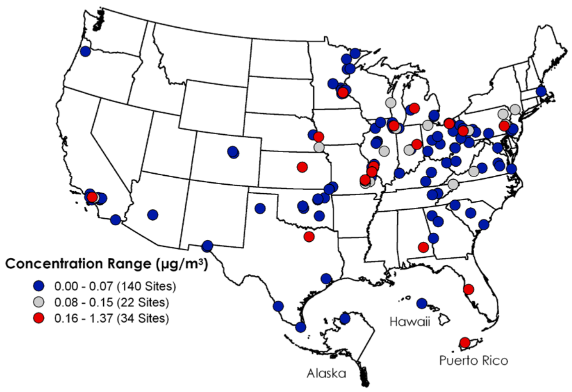
The US EPA operates several networks (the Urban Air Toxics Monitoring Program [UATMP], the National Air Toxics Trends Station [NATTS] Network, and the Community-Scale Air Toxics Ambient Monitoring [CSATAM] Program) that collect information on outdoor concentrations of HAPs. The 2010 report includes data from samples collected at 52 monitoring sites that collected 24-hour air samples, typically on a 1-in-6 day or 1-in-12 day schedule. Of these, 24 sites sampled for 61 VOCs, 30 sites sampled for 14 carbonyl compounds, 26 sites sampled for 22 PAHs, 14 sites sampled for 11 metals, and 23 sites sampled for hexavalent chromium (EPA, 2012d). The report provides detailed summary (and individual site) statistics on all of the measured pollutants, and a risk-based screening approach is applied to identify pollutants of highest priority based

Fig. 1.8 SO_2 (99th percentile of daily 1-hour maximum) concentrations in 2010 in the USA



SO_2, sulfur dioxide.
Reprinted from EPA (2012c). Air quality monitoring information. Available from: http://www.epa.gov/airtrends/factbook.html.

Fig. 1.9 Lead (maximum 3-month average) concentrations in 2010 in the USA



Reprinted from EPA (2012c). Air quality monitoring information. Available from: http://www.epa.gov/airtrends/factbook.html.

on the proportion of measurements exceeding risk-based screening levels. These "pollutants of interest" and 2010 summary concentrations are presented in Table 1.5. Information on trends is provided for individual sites, most of which indicate small decreases over the past about 8 years of monitoring, but data are not systematically analysed for temporal trends at the national level (EPA, 2012d).

The US EPA also produces the National-Scale Air Toxics Assessment (NATA) as a screening risk assessment tool that is used to identify pollutants and locations of specific concern in relation to potential cancer risk from air pollution and to assess trends. The most recent NATA, for 2005, was published in 2011 (EPA, 2012e) and includes information on 177 HAPs identified in the Clean Air Act as well as diesel PM.

In addition to government reporting, several research projects have reported outdoor concentrations of HAPs at the national scale. The Health Effects Institute (HEI) summarized outdoor concentrations of seven priority mobile-source air toxics (acetaldehyde, acrolein, benzene, 1,3-butadiene, formaldehyde, naphthalene, and polycyclic organic matter) because it was determined that mobile sources were a sizeable source of human exposure and existing data suggested potential for adverse health effects at outdoor concentrations. The report provides summaries of outdoor concentrations for each of these pollutants, except naphthalene (for which outdoor concentrations are reported as being < 1 µg/m³) (HEI, 2007).

(ii) Canada

In Canada, the National Air Pollution Surveillance (NAPS) network was initiated in 1969 and currently includes about 300 sites in more than 200 communities located in every province and territory of the country. Monitoring is focused on SO_2, NO_2, ozone, CO, PM_{10}, and $PM_{2.5}$, and a suite of 50 elements (including metals such as arsenic, lead, and mercury), 14 inorganic and organic anions, and 11 inorganic cations are measured in PM samples. Additional measurements of trace contaminants, including VOCs, PAHs, polychlorinated biphenyls (PCBs), and dioxins, are made at a subset of about 40 locations. Results are summarized in a series

Table 1.5 Summary concentrations of air toxics “pollutants of interest” in the USA for 2010

Compound	Mean	SD	Median	25th percentile	75th percentile
PAHs[a]					
Acenaphthene	3.98	7.69	2.04	0.983	4.30
Benzo[*a*]pyrene	0.131	1.32	0.02	0	0.1
Fluorene	4.82	8.09	2.91	1.75	5.39
Naphthalene	95.3	117	66.4	36.6	117
Metals[b]					
Arsenic (PM_{10})	0.558	0.535	0.415	0.240	0.700
Beryllium (PM_{10})	0.003	0.005	0.002	0.0003	0.004
Cadmium (PM_{10})	0.164	0.238	0.084	0.050	0.176
Lead (PM_{10})	3.67	4.95	2.32	1.45	3.74
Manganese (PM_{10})	6.82	11.6	4.03	2.15	7.97
Nickel (PM_{10})	1.06	0.915	0.845	0.594	1.22
Hexavalent chromium	0.037	0.129	0.018	0	0.032
Carbonyls[c]					
Acetaldehyde	1.06	0.718	0.893	0.597	1.29
Formaldehyde	2.01	1.80	1.66	1.09	2.47
VOCs[d]					
Acrylonitrile	0.017	0.084	0	0	0
Benzene	0.311	0.223	0.245	0.173	0.373
1,3-Butadiene	0.038	0.044	0.026	0.012	0.048
Carbon tetrachloride	0.567	1.38	0.037	0.013	0.272
Chloroform	0.038	0.119	0.020	0.014	0.031
p-Dichlorobenzene	0.019	0.105	0.007	0	0.019
1,2-Dichloroethane	0.003	0.008	0	0	0
Ethylbenzene	0.082	0.216	0.053	0.03	0.1
Tetrachloroethylene	0.025	0.029	0.016	0.008	0.03
Trichloroethylene	0.011	0.073	0	0	0
Vinyl chloride	0.0004	0.002	0	0	0

PAHs, polycyclic aromatic hydrocarbons; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; SD, standard deviation; VOCs, volatile organic compounds.

[a] PAHs: unit is ng/m^3.

[b] Metals: unit is ng/m^3.

[c] Carbonyls: unit is ppbv.

[d] VOCs: unit is ppbv.

Data extracted from the 2010 National Monitoring Programs Annual Report (EPA, 2012d) of the US EPA monitoring networks, which collect information on outdoor concentrations of hazardous air pollutants (Urban Air Toxics Monitoring Program [UATMP], National Air Toxics Trends Station [NATTS] Network, and Community-Scale Air Toxics Ambient Monitoring [CSATAM] Program).

of annual and summary reports (Environment Canada, 2010). Concentrations of major air pollutants have declined dramatically over the past about 40 years of measurement, as seen in Fig. 1.10, Fig. 1.11, Fig. 1.12, and Fig. 1.13.

In addition, the Canadian Air and Precipitation Monitoring Network operates 29 locations where $PM_{2.5}$ speciation is measured. Hystad et al. (2011) used a combination of satellite-based estimates and land-use characteristics to model the concentrations of $PM_{2.5}$ and NO_2 across Canada. National models for benzene, ethylbenzene, and 1,3-butadiene were also developed based on land use and source proximity characteristics (Hystad et al., 2011).

Setton et al. (2013) used data from the NAPS network (for 2006) and measurements reported in the literature or government reports since 2000 along with deterministic concentration gradients based on proximity to major roads and industrial sources to estimate exposure to several IARC Group 1 carcinogens in outdoor air in Canada. Table 1.6 presents estimated exposures to selected IARC Group 1, Group 2A, and Group 2B carcinogens in outdoor air in Canada for 2010, based on data from the CAREX database (CAREX Canada, 2013).

(iii) Mexico

The National Information System for Air Quality (SINAICA) operates about 50 sites in Mexico where ozone, NO_x, CO, SO_2, PM_{10}, TSP, and VOCs are measured (SINAICA, 2011). An additional network of about 60 sites is operated in Mexico City for the same general suite of pollutants (Secretaría del Medio Ambiente, 2013). Data from the air quality monitoring networks are centralized by the National Institute of Ecology, with detailed reports provided for specific airsheds. Parrish et al. (2011) provided summaries of trends of annual average concentrations in Mexico City over a 20-year period in which air quality has improved substantially (Fig. 1.14).

Annual average particulate PAH concentrations (in PM_{10}) collected at a site in Mexico City over a period of several years are summarized in Table 1.7. For several of the PAHs, concentrations increased during this 4-year period, even as PM_{10} concentrations decreased (Amador-Muňoz *et al.*, 2013).

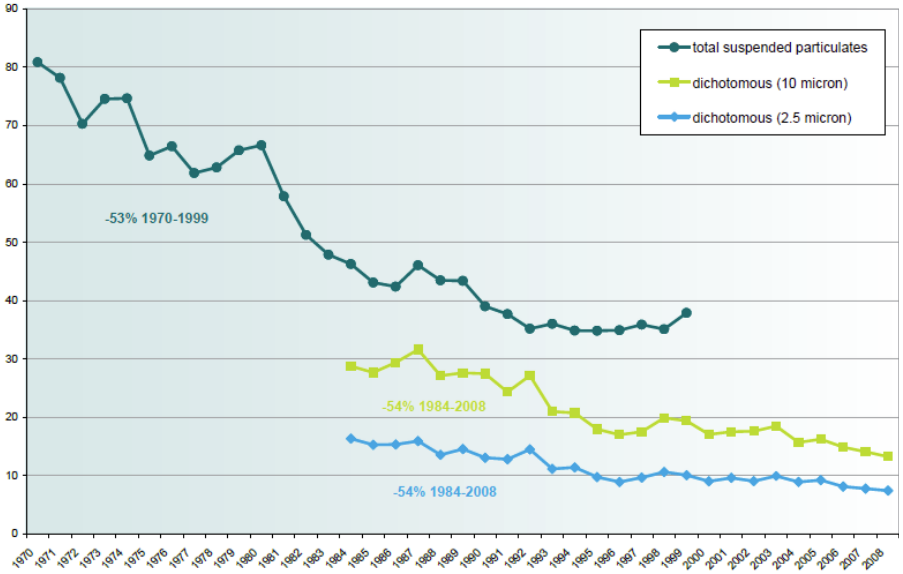
Mexico, Canada, and the USA operate a collaborative network for measurement of dioxins and furans, including nine stations in Mexico (five rural, two semi-urban, and two urban sites) (Cardenas et al., 2011). The mean concentrations for the background (rural) and semi-urban sites were 1.59 fg/m^3 and 18.6 fg/m^3, respectively, which are of the same order of magnitude as those reported by the outdoor monitoring networks in the USA and Canada. However, the mean concentration for the urban sites was 282 fg/m^3, which is significantly higher than concentrations measured at similar sites in the USA and Canada.

(b) Europe

In this section, information on outdoor concentration levels, spatial variation, and time trends in major outdoor air pollutants in Europe is summarized. Data are available from routine monitoring networks and several large research projects. This text focuses on concentration data from 38 countries that are members or cooperating members of the European Environment Agency (EEA), so that the spatial pattern across Europe is broadly represented.

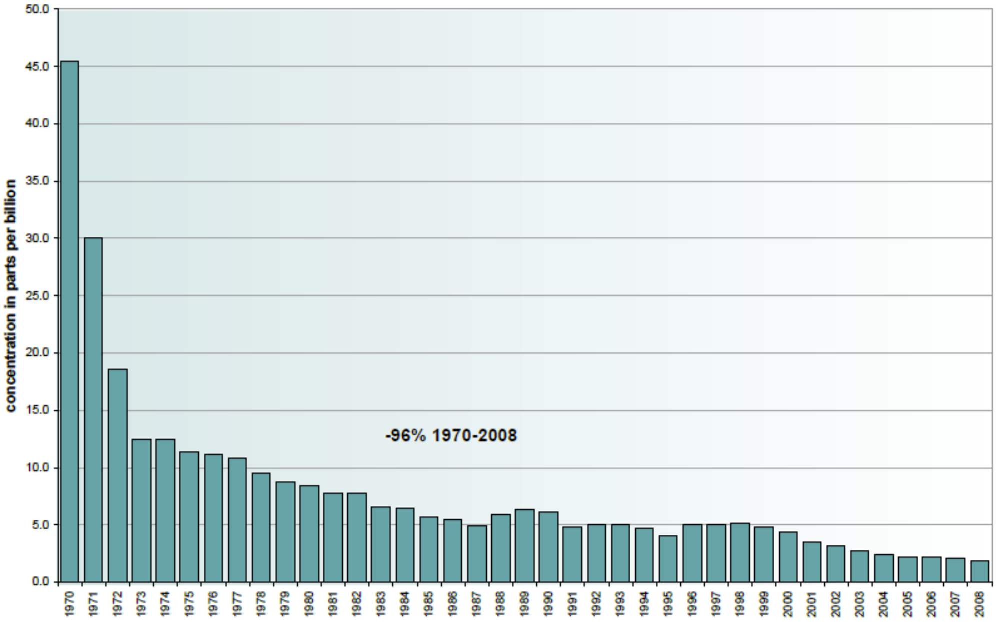
Routine monitoring networks are national in Europe; there is no comprehensive European network. EU Member States have to report their data to the EU, resulting in the European air quality database AirBase, maintained by the EEA, in which concentrations and metadata (site description, monitoring methods) are available (EEA, 2014). European reference methods have been defined for regulated pollutants. The EEA regularly reports assessment of air quality across Europe (e.g. EEA, 2012).

Fig. 1.10 Trends (1970–2008) in annual mean particle (TSP, PM_{10}, $PM_{2.5}$) concentrations measured at National Air Pollution Surveillance (NAPS) sites in Canada



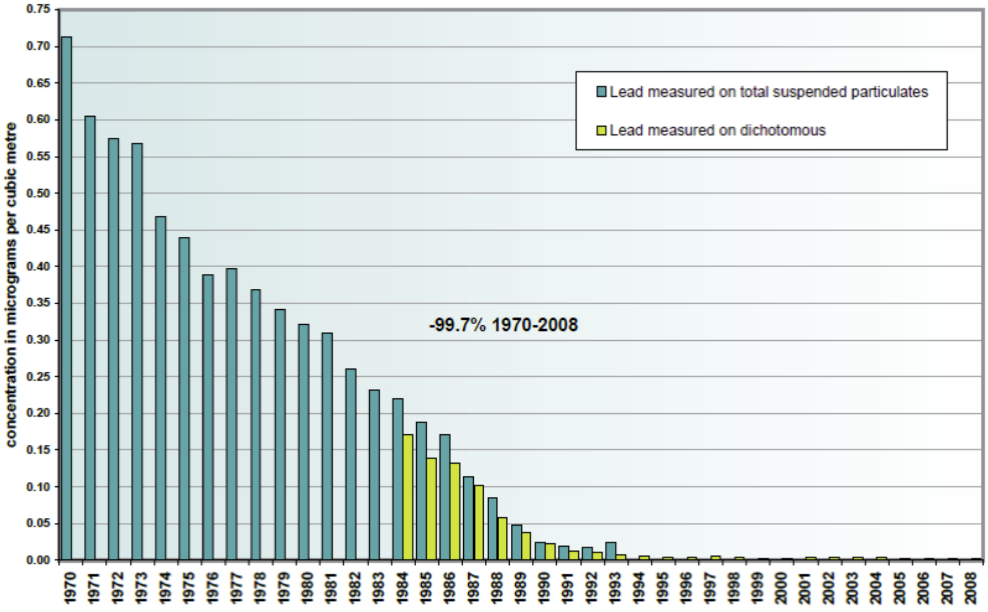
PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; SO_2, sulfur dioxide; TSP, total suspended particles.
Reprinted from Environment Canada (2010). National air pollution surveillance. Available from: http://www.ec.gc.ca/rnspa-naps/default.asp?lang=En&n=77FECF05-1#reports.

Fig. 1.11 Trends (1970–2008) in annual mean SO_2 concentrations at National Air Pollution Surveillance (NAPS) sites in Canada



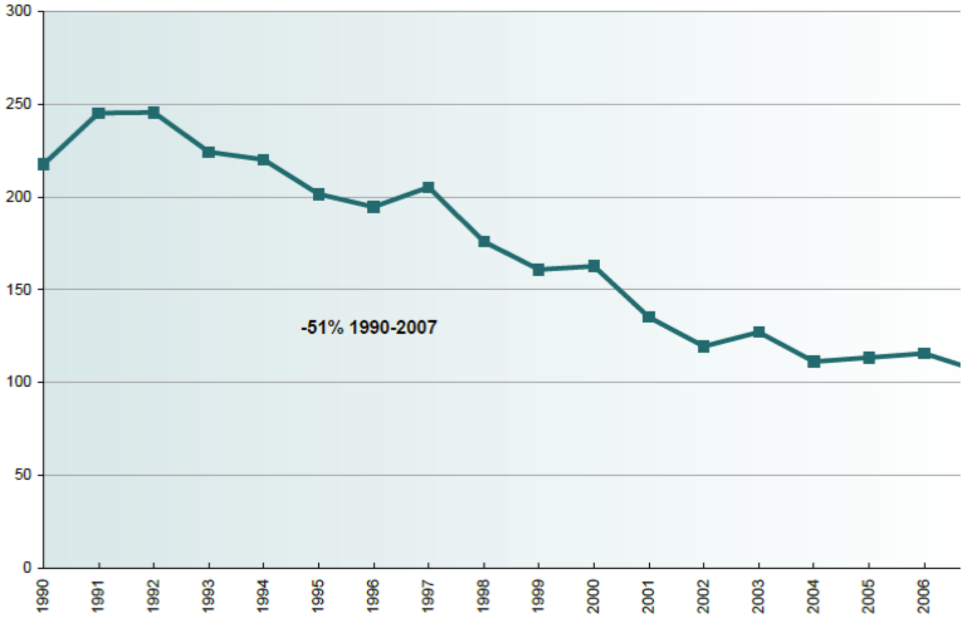
SO_2, sulfur dioxide.
Reprinted from Environment Canada (2010). National air pollution surveillance. Available from: http://www.ec.gc.ca/rnspa-naps/default.asp?lang=En&n=77FECF05-1#reports.

Fig. 1.12 Trends (1970–2008) in annual mean particulate lead concentrations at National Air Pollution Surveillance (NAPS) sites in Canada



Reprinted from Environment Canada (2010). National air pollution surveillance. Available from: http://www.ec.gc.ca/rnspa-naps/default.asp?lang=En&n=77FECF05-1#reports.

Fig. 1.13 Trends (1990–2007) in annual mean total volatile organic compounds (VOCs) at National Air Pollution Surveillance (NAPS) sites in Canada



Reprinted from Environment Canada (2010). National air pollution surveillance. Available from: http://www.ec.gc.ca/rnspa-naps/default.asp?lang=En&n=77FECF05-1#reports.

Table 1.6 Estimated exposures (in 2010) to selected IARC Group 1, Group 2A, and Group 2B carcinogens in outdoor air in Canada

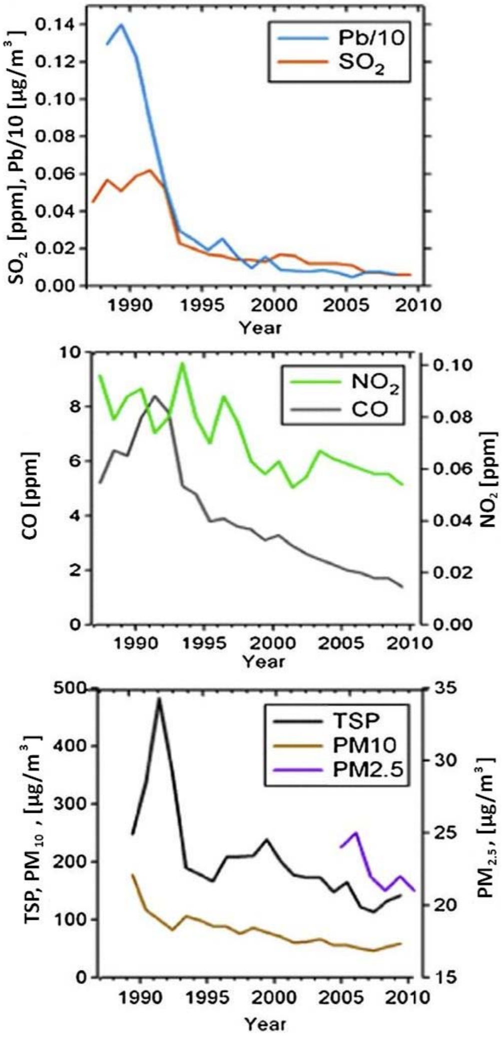
Compound	Mean concentration (μg/m³)
Group 1	
1,3-Butadiene	0.073
Benzene	0.84
Formaldehyde	1.4
Benzo[*a*]pyrene	1.1×10^{-4}
2,3,7,8-Tetrachlorodibenzo-*para*-dioxin	10^{-9}
Polychlorinated biphenyls (PCBs)	2.5×10^{-9}
Diesel engine exhaust	0.8
Arsenic	4.3×10^{-4}
Cadmium	1.1×10^{-4}
Hexavalent chromium	2×10^{-5}
Nickel compounds	5×10^{-4}
Group 2A	
Dichloromethane	0.68
Tetrachloroethylene	0.2
Lead compounds	0.0012
Group 2B	
Acetaldehyde	0.81
Chloroform	0.15
Ethylbenzene	0.55
Benzo[*b*]fluoranthene	4×10^{-4}
Benzo[*k*]fluoranthene	1.1×10^{-4}
Benz[*a*]anthracene	1.8×10^{-4}
Chrysene	2×10^{-4}
Indeno[1,2,3-*cd*]pyrene	1×10^{-4}

Prepared by the Working Group with data from CAREX Canada (2013).

The European Monitoring and Evaluation Programme (EMEP) is a European network of regional background stations that was designed in the 1970s in response to the observation of transboundary air pollution (Tørseth et al., 2012). The network includes measurements of SO_2, NO_2, sulfate/nitrate in aerosols, and more recently PM, ozone, and POPs.

Maps prepared by the EEA of the annual average concentrations across Europe of PM_{10}, $PM_{2.5}$, NO_2, SO_2, and ozone are presented in Fig. 1.15, Fig. 1.16, Fig. 1.19, Fig. 1.21, Fig. 1.22; other maps, for heavy metals in PM, CO, benzene, and PAH concentrations, can be found in the EEA report (EEA, 2012). These components were selected based on availability of at least reasonably comparable data across Europe.

Fig. 1.14 Trends in outdoor concentrations of lead, SO_2, NO_2, CO, and particles (TSP, PM_{10}, $PM_{2.5}$) in Mexico City



Plots show the average of the fifth-highest annual maximum from all stations with valid data for a given year.
CO, carbon monoxide; NO_2, nitrogen dioxide; Pb, lead; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 μm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 μm; SO_2, sulfur dioxide; TSP, total suspended particles.
Reprinted from Parrish et al. (2011). Air quality progress in North American megacities: a review. *Atmos Environ*, 45(39):7015–25, with permission from Elsevier.

Table 1.7 Annual medians of mass polycyclic aromatic hydrocarbons (PAHs) concentrations in PM_{10} (10th–99th percentile) (pg/m³) from the sampling days of 1999–2002 at a site in south-west Mexico City

PAH	1999 (*n* = 58)	2000 (*n* = 69)	2001 (*n* = 88)	2002 (*n* = 73)
Phenanthrene	116 (39–250)	122 (63–270)	141 (87–240)	135 (78–239)
Anthracene	21 (10–38)	17 (7–44)	25 (16–40)	21 (13–35)
Fluoranthene	230 (93–514)	204 (95–529)	260 (145–511)	253 (126–460)
Pyrene	334 (120–747)	290 (135–704)	387 (225–718)	322 (163–593)
Retene	134 (26–538)	5 (4–164)[d]	83 (51–258)	97 (49–261)
Benzo[*a*]anthracene	143 (46–361)	155 (61–403)	165 (87–334)	175 (82–380)
Chrysene[a]	212 (66–518)	200 (94–492)	187 (112–435)	234 (111–485)
Benzo[*b*]fluoranthene	588 (194–1179)	417[e] (147–963)	368[e] (199–814)	505 (314–1030)
Benzo[*k*]fluoranthene[b]	454 (159–896)	474 (240–1025)	382 (236–857)	440 (178–930)
Benzo[*e*]pyrene	506 (176–1052)	474 (235–950)	461 (290–924)	601 (356–1009)
Benzo[*a*]pyrene	240 (63–649)	313 (129–787)	274 (154–725)	357 (187–730)
Perylene	33 (0–92)[f]	53[e] (21–108)[f]	48[e] (25–108)[f]	74 (19–142)[g]
Indeno[1,2,3-*cd*]pyrene	734 (230–1587)	700 (306–1353)	623 (381–1388)	896 (506–1544)
Dibenzo[*a*,*h*]anthracene	45 (14–91)	43 (16–89)	43 (18–97)	46 (27–95)
Benzo[*ghi*]perylene	1342 (427–2793)	1289 (631–2644)	1342 (746–2891)	1856 (1058–2994)
Coronene	892 (267–1850)	755 (340–1624)	855 (49–2024)	1077 (564–2257)
Light PAH[c]	926 (293–2145)	686 (302–1595)	877 (531–1702)	836 (468–1636)
Heavy PAH[c]	5301 (1578–11 174)	4953 (2393–10 186)	4636 (2829–10 148)	6206 (3588–11 399)

PM_{10}, particulate matter with particles of aerodynamic diameter < 10 μm.

[a] Probably chrysene co-eluting with triphenylene

[b] Probably benzo[*k*]fluoranthene co-eluting with benzo[*j*]fluoranthene

[c] The values were calculated taking into account the corresponding PAH sum in each sampling day by year.

[d] Some values of retene were lower than the quantification limit.

[e] Contiguous medians were not statistically different.

[f] All daily values of perylene were lower than the quantification limit.

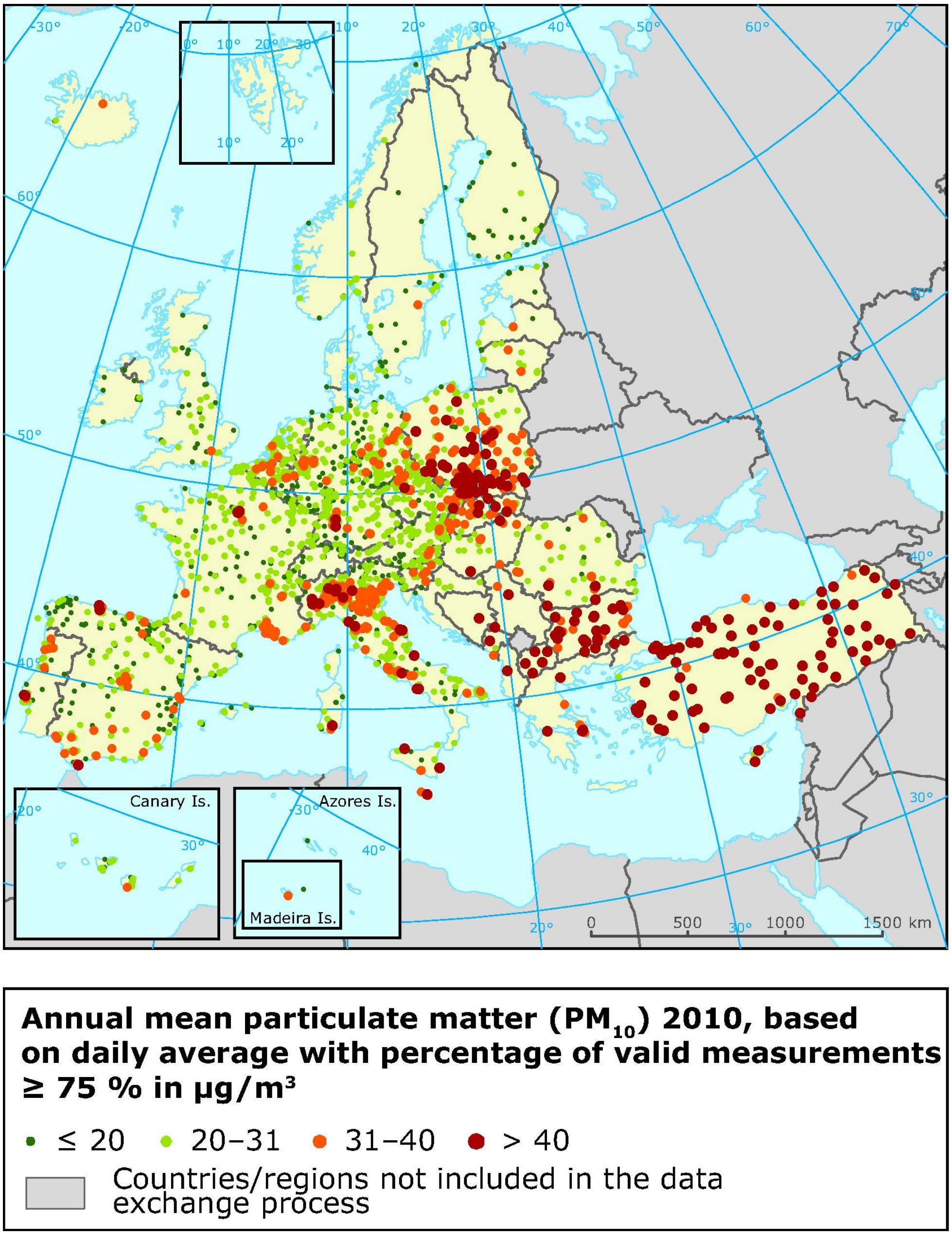
[g] Some daily values of perylene were lower than the quantification limit.

Adapted from Amador-Muñoz et al. (2013). Opposing seasonal trends for polycyclic aromatic hydrocarbons and PM_{10}: health risk and sources in southwest Mexico City. *Atmos Res*, 122:199–212. doi:10.1016/j.atmosres.2012.10.003, © with permission from Elsevier.

(i) PM_{10} and $PM_{2.5}$

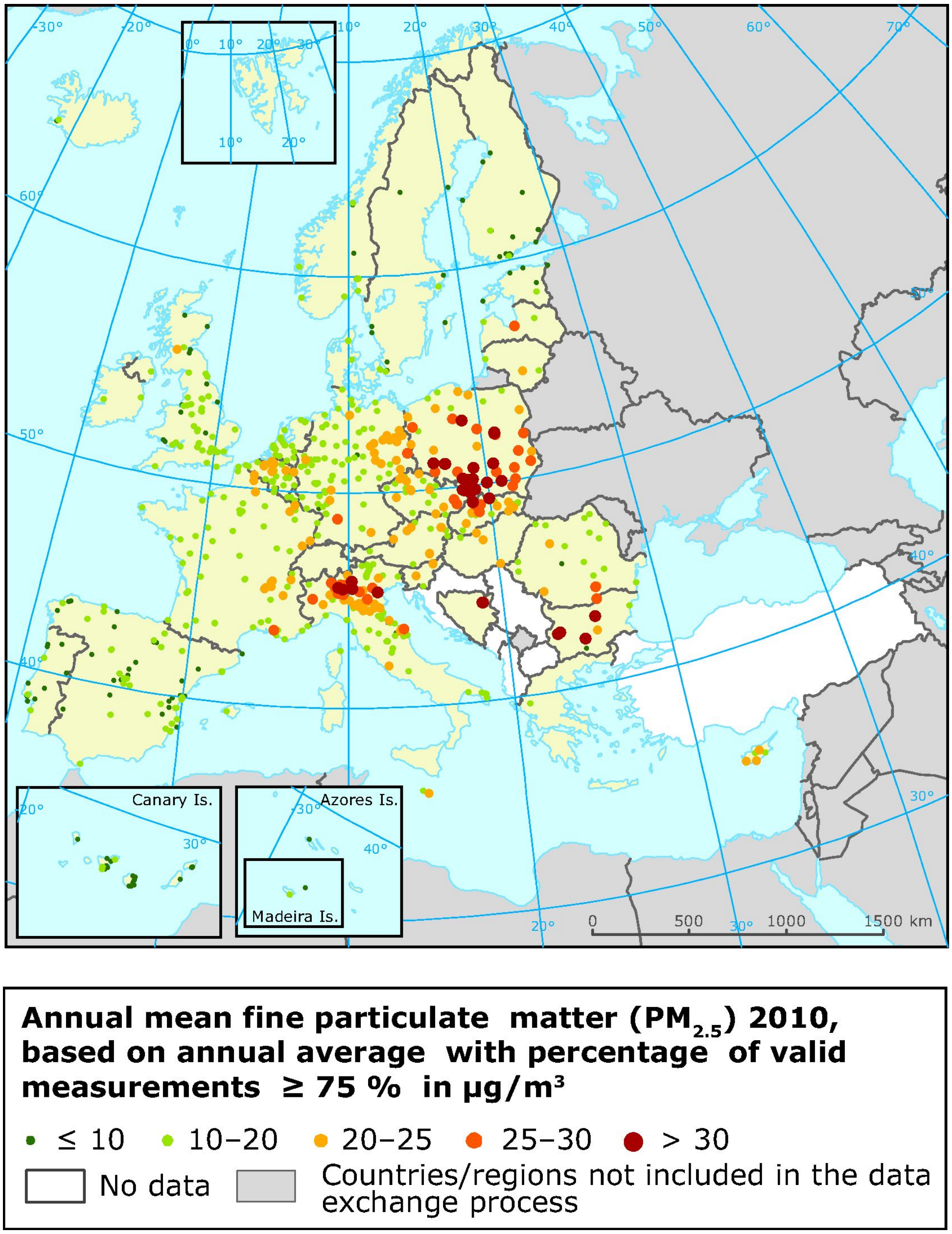
The PM_{10} and $PM_{2.5}$ maps (Fig. 1.15 and Fig. 1.16) show that concentrations are lower in northern Europe than in southern and eastern Europe. The PM_{10} map is based on a substantially larger number of sites than the $PM_{2.5}$ map, because $PM_{2.5}$ monitoring has not been fully developed within Europe because of later adoption of the air quality guideline for $PM_{2.5}$. Several research projects have broadly confirmed the general patterns across Europe (Hazenkamp-von Arx et al., 2004; Putaud et al., 2004, 2010; Van Dingenen et al., 2004; Eeftens et al., 2012). In the ESCAPE study (Eeftens et al., 2012), based on standardized gravimetric measurements using the Harvard impactor in 20 study areas, average $PM_{2.5}$ concentrations below 10 μg/m³ were found in northern Europe (Fig. 1.17). In southern European cities, for example Athens (Greece) and Turin (Italy), annual average $PM_{2.5}$ concentrations above 20 μg/m³ were measured. Relatively high concentrations were also found in the two central European cities Györ (Hungary) and Kaunas (Lithuania). Fig 1.17 further illustrates significant intra-urban spatial variation, particularly for coarse particles (calculated as PM_{10} – $PM_{2.5}$) and $PM_{2.5}$ absorbance. A regression analysis of $PM_{2.5}$ on PM_{10} concentrations for

Fig. 1.15 Annual mean concentrations of PM_{10} in 2010 in Europe



PM_{10}, particulate matter with particles of aerodynamic diameter < 10 μm.
The red dots indicate stations reporting exceedances of the 2005 annual limit value (40 μg/m³), as set out in the Air Quality Directive (EU, 2008).
The orange dots indicate stations reporting exceedances of a statistically derived level (31 μg/m³) corresponding to the 24-hour limit value, as set out in the Air Quality Directive (EU, 2008).
The light green dots indicate stations reporting exceedances of the WHO air quality guideline for PM_{10} of < 20 μg/m³ but not in exceedance of the limit values as set out in the Air Quality Directive (EU, 2008).
The dark green dots indicate stations reporting concentrations below the WHO air quality guideline for PM_{10} and implicitly below the limit values as set out in the Air Quality Directive (EU, 2008).
Reprinted from EEA (2012). Air quality in Europe – 2012 report. European Environment Agency Report 4/2012.

Fig. 1.16 Annual mean concentrations of $PM_{2.5}$ in 2010 in Europe



$PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm.

The red dots indicate stations reporting exceedances of the 2010 annual target value (25 µg/m³) plus at least 5 µg/m³.

The dark orange dots indicate stations reporting exceedances of the 2010 annual target value (25 µg/m³), as set out in the Air Quality Directive (EU, 2008).

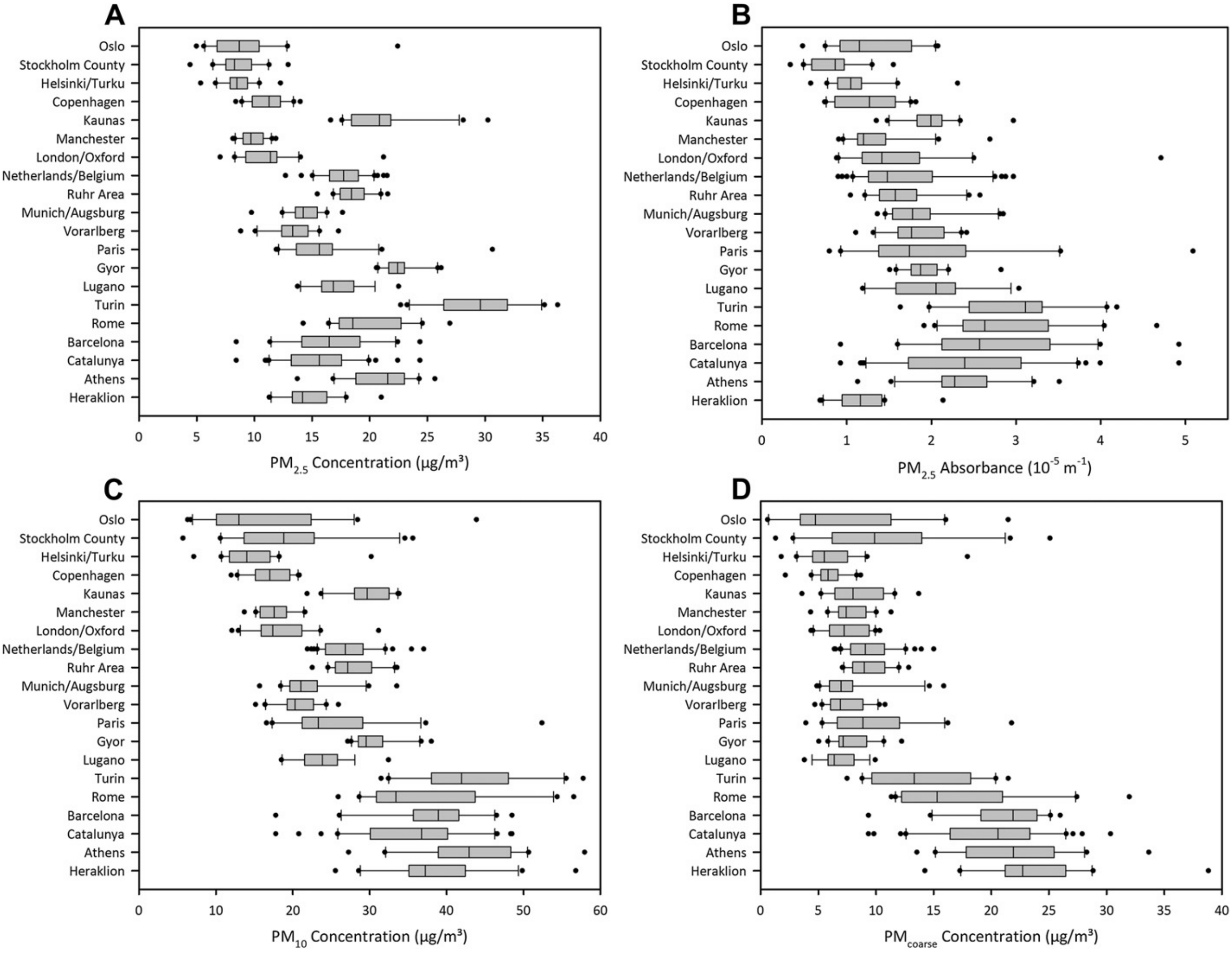
The light orange dots indicate stations reporting exceedances of the 2020 indicative annual limit value (20 µg/m³), as set out in the Air Quality Directive (EU, 2008).

The light green dots indicate stations reporting exceedances of the WHO air quality guideline for $PM_{2.5}$ of < 10 µg/m³ but not in exceedance of the target or limit values for $PM_{2.5}$ as set out in the Air Quality Directive (EU, 2008).

The dark green dots indicate stations reporting concentrations below the WHO air quality guideline for $PM_{2.5}$ and implicitly below the target and limit values for $PM_{2.5}$ as set out in the Air Quality Directive (EU, 2008).

Reprinted from EEA (2012). Air quality in Europe – 2012 report. European Environment Agency Report 4/2012.

Fig. 1.17 Spatial variation of 2009–2010 annual average particulate matter (PM) concentrations across Europe



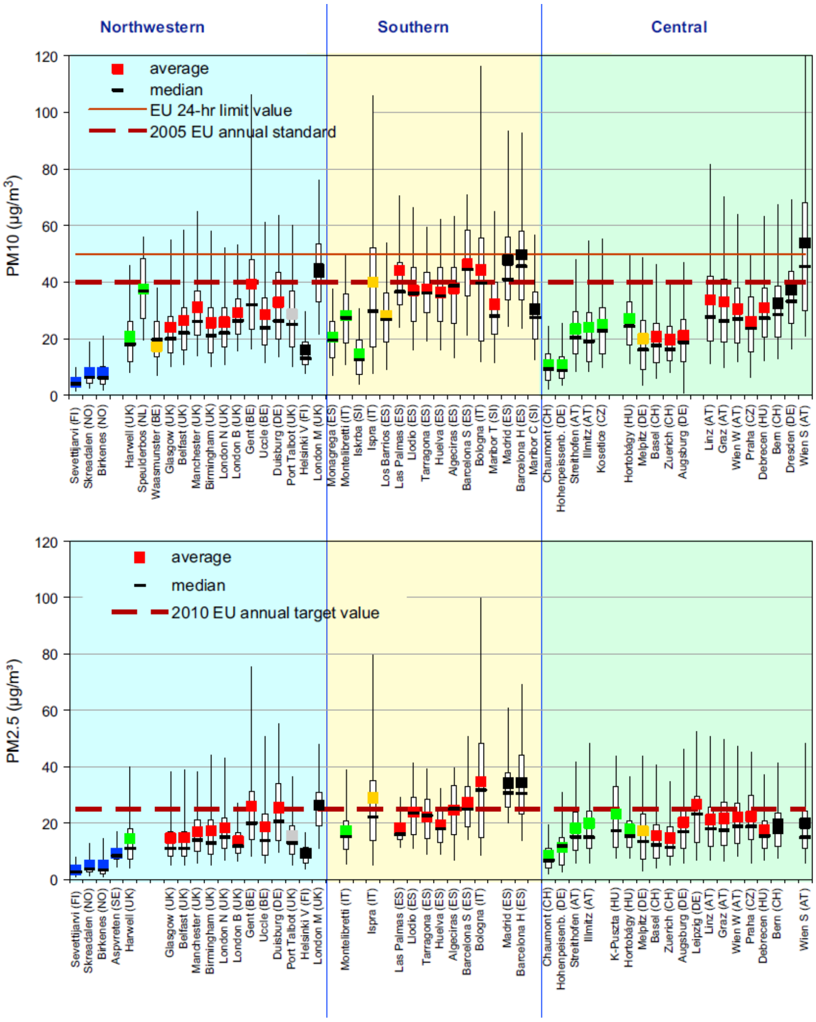
PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; PM_{coarse} calculated as $PM_{10} - PM_{2.5}$.

Median, 25th percentile, and 75th percentile are shown in the boxes; whiskers indicate the 10th and 90th percentiles, and individual outliers are shown as points.

Reprinted from Eeftens et al. (2012). Spatial variation of $PM_{2.5}$, PM_{10}, $PM_{2.5}$ absorbance, and PM_{coarse} concentrations between and within 20 European study areas and the relationship with NO_2 – results of the ESCAPE project. *Atmos Environ*, 62:303–317; with permission from Elsevier.

60 sites across Europe found site-specific slopes varying between 0.44 and 0.90 (Putaud et al., 2010). Fig 1.18 illustrates the spatial variation of $PM_{2.5}$ and PM_{10} concentrations across European cities. A large range of PM_{10} concentrations (5–54 µg/m³ annual average) is observed across the network. Urban background PM_{10} annual mean and median values are significantly larger in southern Europe (median, 36 µg/m³) than in north-western Europe (median, 24 µg/m³) and central Europe (median, 26 µg/m³). The range of $PM_{2.5}$ concentrations observed across the network (3–35 µg/m³ annual average) is similar to that of PM_{10}. In north-western and southern Europe, an increasing gradient in $PM_{2.5}$ is generally observed from rural to urban sites. In central Europe, $PM_{2.5}$ can be as large at rural sites as at urban sites (Putaud et al., 2010). The chemical composition of PM differs widely across Europe, with generally more carbonaceous matter in

Fig. 1.18 Spatial variation of 1996–2007 annual average particulate matter (PM) concentrations across Europe



PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm.

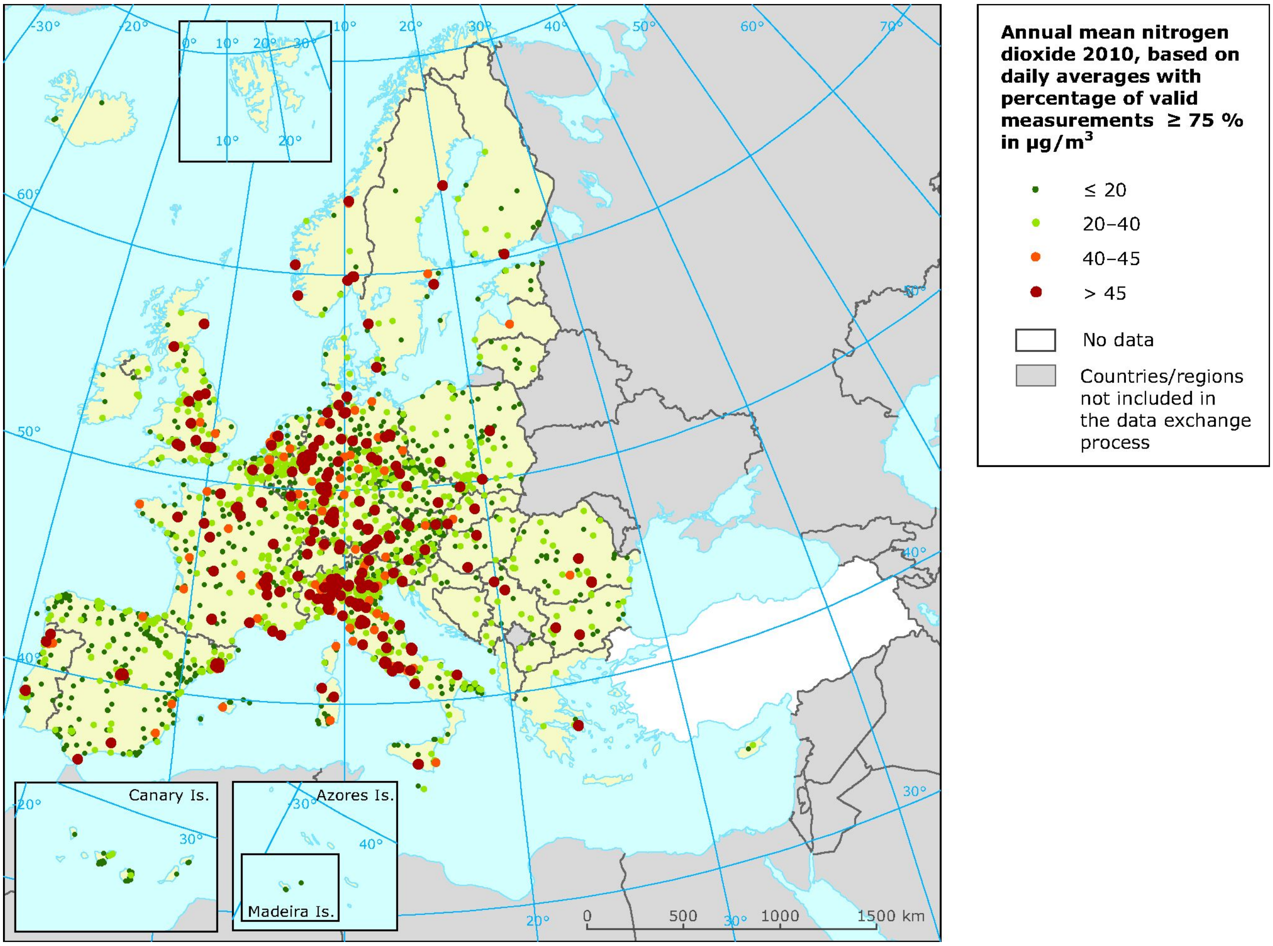
Median, 25th percentile, and 75th percentile are shown in the boxes; whiskers indicate the 10th and 90th percentiles, and individual outliers are shown as points.

Reprinted from Putaud et al. (2010). A European aerosol phenomenology – 3: physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe. *Atmos Environ*, 44(10):1308–1320; with permission from Elsevier.

central Europe, more nitrate in north-western Europe, and more mineral dust in southern Europe (Putaud et al., 2010; Tørseth et al., 2012). Table 1.8 presents the average contributions of major components to PM concentrations. The elemental composition of eight elements representing major sources across Europe has recently been published based on the ESCAPE study (de Hoogh et al., 2013). Significant variability of concentrations both within and between study areas across Europe was found.

There is a lack of comprehensive monitoring for the heavy metals lead, arsenic, cadmium, and nickel across Europe. In general, low concentration levels are measured (often below the lower assessment threshold), with the exception of sites located next to specific industries (EEA, 2012).

Fig. 1.19 Annual mean concentration of NO_2 in 2010 in Europe



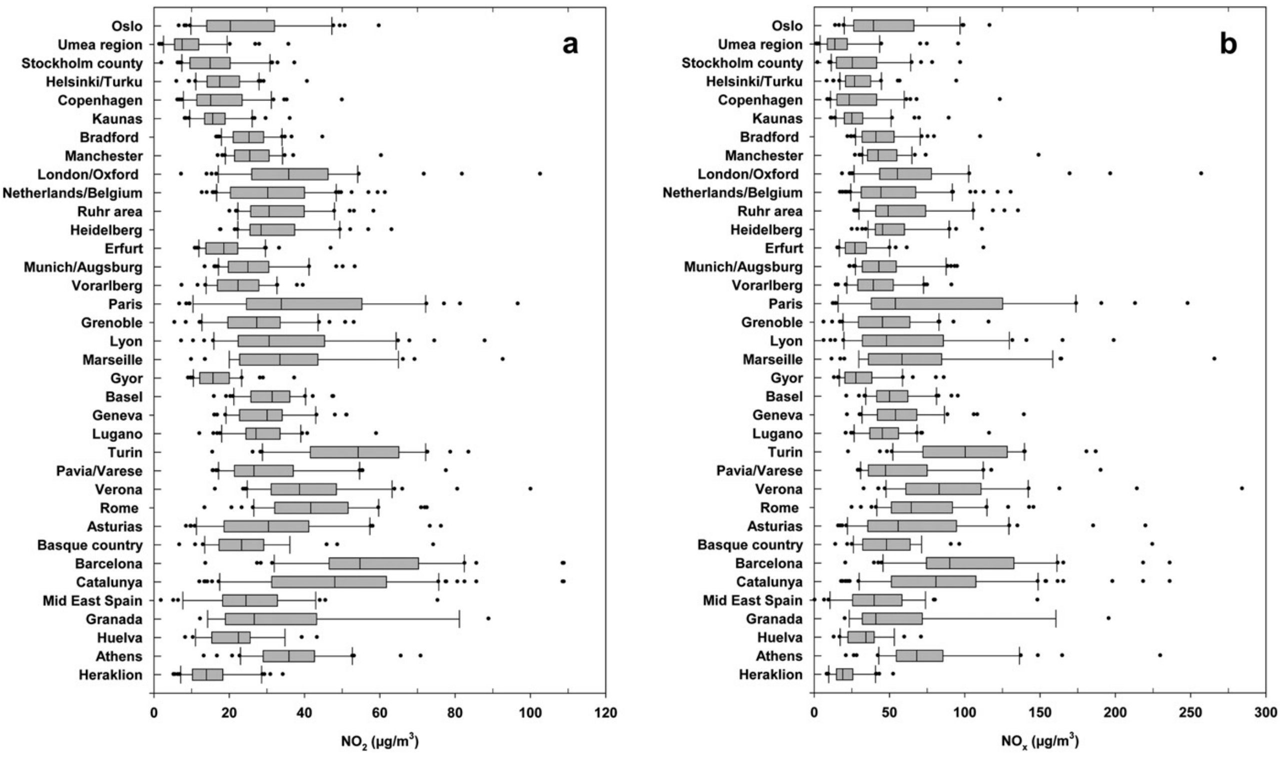
NO_2, nitrogen dioxide.
Orange and red dots correspond to exceedances of the annual limit value (40 µg/m³).
Red dots correspond to exceedances of the annual limit value plus 5 µg/m³.
Reprinted from EEA (2012). Air quality in Europe – 2012 report. European Environment Agency Report 4/2012.

There are no routine measurements of ultrafine particles available across Europe, as in other parts of the world. In individual cities, including Amsterdam (the Netherlands), Athens (Greece), Birmingham (United Kingdom), and Helsinki (Finland), total particle number counts are available. Research projects have included snapshots of spatial patterns across Europe (e.g. Puustinen et al., 2007). Urban background levels were about 10 000–20 000 particles/cm³ in four large cities, with substantially higher concentrations measured near major roads (Puustinen et al., 2007).

(ii) NO_2

The most striking feature of the NO_2 map is the higher concentrations in major cities (Fig. 1.19). European research studies have also shown a general north-to-south increasing gradient in NO_2 concentrations (Hazenkamp-von Arx et al., 2004; Cyrys et al., 2012). Fig 1.20 further illustrates significant intra-urban spatial variation, which exceeded between-area variability. In

Fig. 1.20 Spatial variation of 2008–2011 annual average nitrogen dioxide (NO_2) and nitrogen oxides (NO_x) concentrations across Europe



a Distribution of annual average concentration of NO_2 for each study area separately. **b** Distribution of annual average concentration of NO_x for each study area separately.

Median, 25th percentile, and 75th percentile are shown in the boxes; whiskers indicate the 10th and 90th percentiles, and individual outliers are shown as points.

In each study area, 40–80 sites were measured. Sites ordered from north to south.

Reprinted from Cyrys et al. (2012). Variation of NO_2 and NO_x concentrations between and within 36 European study areas: results from the ESCAPE study. *Atmos Environ*, 62:374–90; with permission from Elsevier.

virtually all study areas, there was at least one site in which the current EU annual average standard of 40 μg/m³ was exceeded.

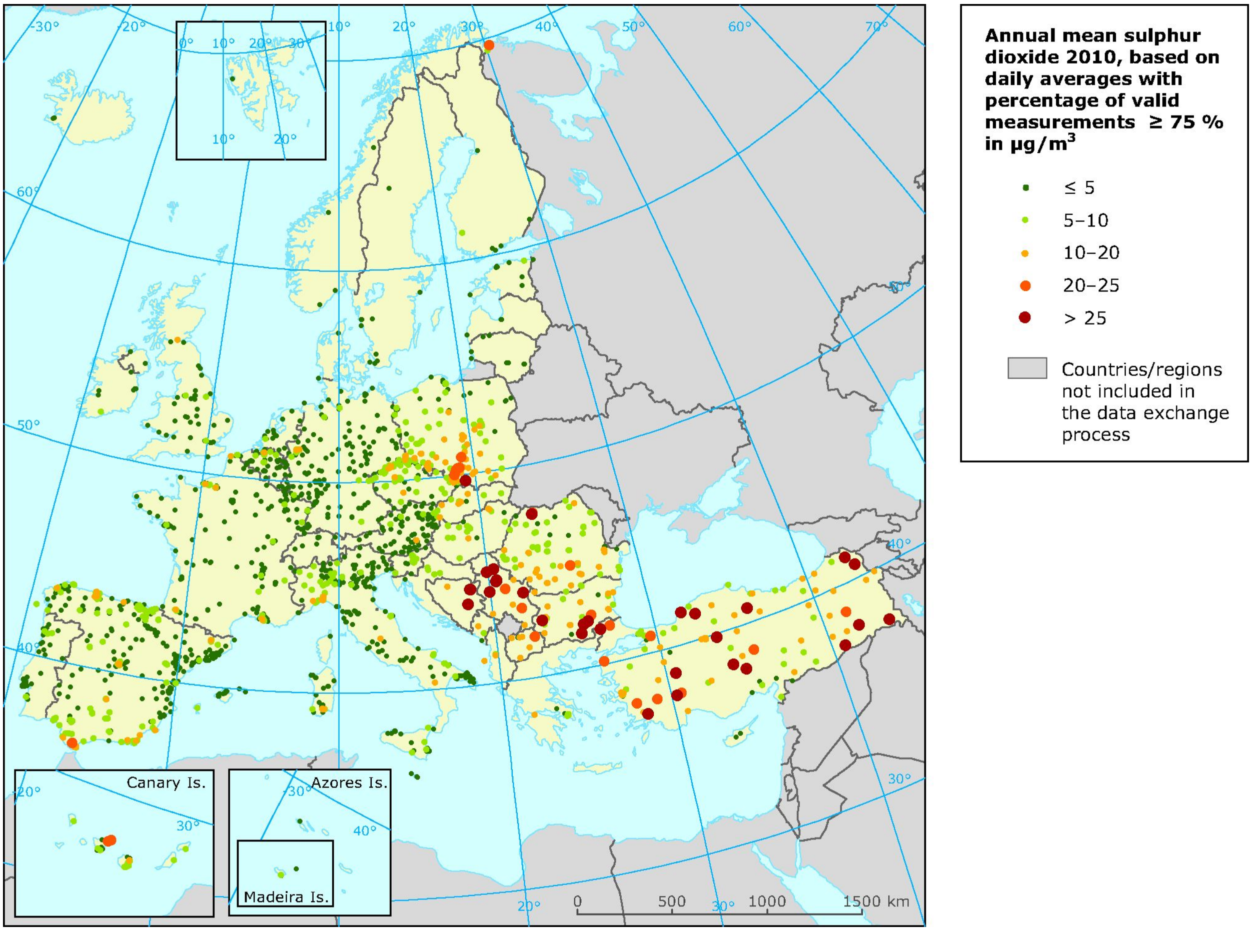
(iii) SO_2

Current average SO_2 concentrations in Europe are low, typically well below 10 μg/m³ in large parts of Europe (Fig. 1.21). The highest concentrations occur in eastern Europe, related to industrial activities and the remaining coal burning (EEA, 2012). Currently, emissions are predominantly from power generation (Tørseth et al., 2012). International shipping emissions have become a significant source because shipping emissions have been much less affected by policies than industrial emissions have (Tørseth et al., 2012).

(iv) CO

CO concentrations are typically low, due to the significant reduction in traffic emissions by catalytic converters. Still, the highest concentrations occur in urban areas, especially at traffic sites and occasionally at industrial locations (EEA, 2012). There is not a clear pattern across Europe.

Fig. 1.21 Annual mean SO_2 concentrations in Europe in 2010



The dark orange and red dots correspond to exceedances of the limit value (20 µg/m³) for the protection of vegetation.
Reprinted from EEA (2012). Air quality in Europe – 2012 report. European Environment Agency Report 4/2012.

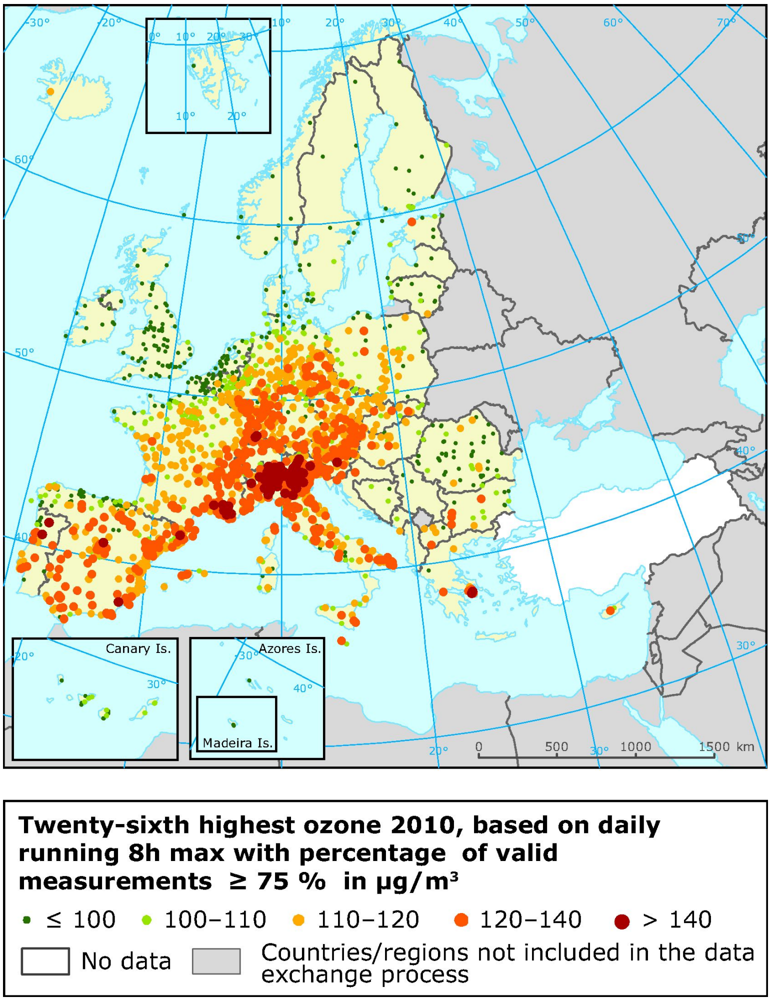
(v) Ozone

Fig. 1.22 shows the map of maximum 8-hour average ozone concentrations. The 26th highest value is shown because of the formulation of the EU standard (120 µg/m³ as an 8-hour maximum not to be exceeded on > 25 days). The highest concentrations occur in southern Europe and in Austria and Switzerland, related especially to higher temperatures and altitude (EEA, 2012). Ozone concentrations are generally higher at rural stations than at urban background stations. Concentrations at traffic sites are even lower, related to scavenging of ozone by NO (EEA, 2012).

(vi) Benzene

Current average benzene concentrations in Europe are low, typically below 5 µg/m³ in large parts of Europe. The highest concentrations occur at traffic sites and at industrial locations (EEA, 2012).

Fig. 1.22 Twenty-sixth highest daily maximum 8-hour average ozone concentration recorded in 2010 in Europe



The map shows the proximity of recorded ozone concentrations to the target value. At sites marked with dark orange and red dots, the 26th highest daily ozone concentrations exceeded the 120 μg/m³ threshold and the number of allowed exceedances by the target value.
Reprinted from EEA (2012). Air quality in Europe – 2012 report. European Environment Agency Report 4/2012.

(vii) Benzo[a]pyrene

Current average B[*a*]P concentrations in Europe are low, typically below 1 ng/m³ in large parts of Europe. There is no clear north-to-south gradient. The highest concentrations occur in areas with domestic coal or wood burning and industrial areas, particularly in eastern Europe (EEA, 2012).

(viii) Pollution trends

Fig. 1.23, Fig. 1.24, Fig. 1.25, and Fig. 1.26 show the trends in annual average concentrations of PM and major gaseous components based on the European AirBase database (EEA, 2012).

Annual average concentrations of PM_{10} and $PM_{2.5}$ have not decreased much since 2000 despite assumed decreases in emissions of precursors (Fig. 1.23). Between 1990 and 2004, a clear decrease (~44%) in total PM emissions

Table 1.8 Major constituent contributions to PM_{10}, $PM_{2.5}$, and PM_{coarse} in Europe

Region	Constituent	PM_{10}			$PM_{2.5}$			PM_{coarse}		
		Rural	Urban	Kerbside	Rural	Urban	Kerbside	Rural	Urban	Kerbside
North-western Europe	Mineral dust	4%	12%			5%	1%		26%	
	Sea salt	12%	10%	7%		4%	1%		15%	
	SO_4	13%	14%	8%		21%	18%		6%	
	NO_3	16%	14%	12%		16%			20%	
	OM	15%	18%	16%		25%			14%	
	EC	4%	5%	9%		7%			1%	
	TC	14%	18%	20%		25%			12%	
Southern Europe	Mineral dust	15%	21%	28%		11%	14%		42%	69%
	Sea salt	3%	12%	5%		6%	2%		22%	11%
	SO_4	16%	12%	12%		15%	15%		4%	5%
	NO_3	14%	9%	8%		7%	7%		11%	9%
	OM		26%			23%			13%	
	EC		6%			8%			2%	
	TC	13%	21%	28%		30%	38%		11%	
Central Europe	Mineral dust	9%	12%	15%	3%	5%	6%	22%	25%	29%
	Sea salt	2%	2%	2%	1%	1%	1%	2%	3%	5%
	SO_4	19%	15%	9%	17%	19%	12%	5%	4%	4%
	NO_3	13%	12%	8%	6%	13%	10%	10%	7%	6%
	OM	23%	21%	21%	15%	22%	26%	5%	15%	13%
	EC	6%	10%	17%	5%	14%	21%	3%	3%	10%
	TC	32%	32%	38%	19%	31%	35%	6%	14%	19%

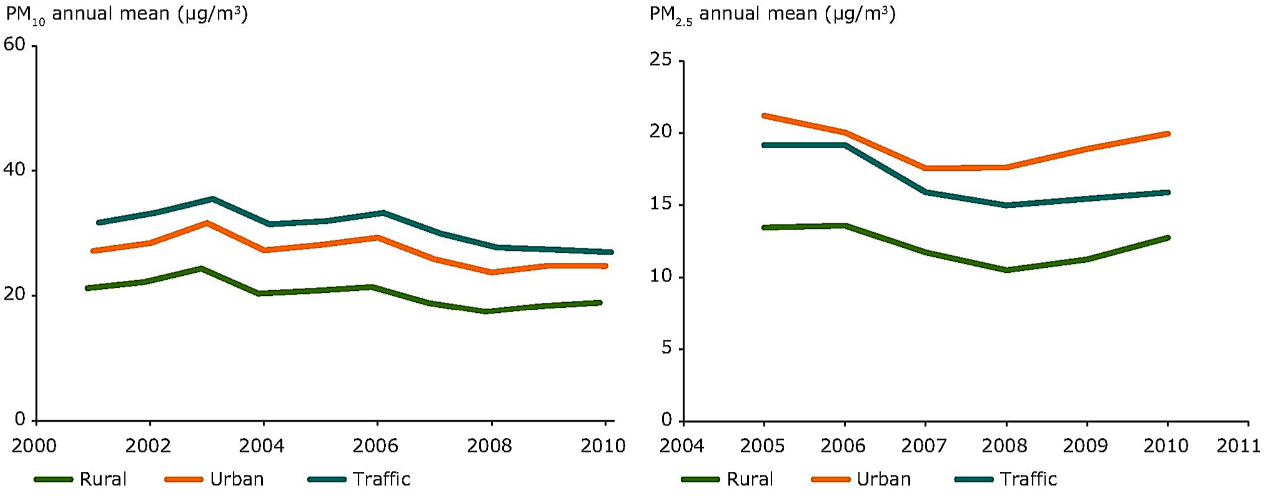
EC, elemental carbon; OM, organic matter; PM, particulate matter; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; PM_{coarse}, particulate matter with particles of aerodynamic diameter between 2.5 µm and 10 µm; TC, total carbon.
Adapted from Putaud et al. (2010). A European aerosol phenomenology – 3: physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe. *Atmos Environ*, 44(10):1308–20; with permission from Elsevier.

occurred (EEA, 2007; Harrison et al., 2008). At the EMEP regional background sites, concentrations of PM_{10} and $PM_{2.5}$ decreased by 18% and 27%, respectively, between 2000 and 2009 (Tørseth et al., 2012). Longer-term trends are difficult to quantify from monitoring networks because PM_{10} and especially $PM_{2.5}$ were often not measured until the 1990s. High annual average concentrations of PM_{10} and $PM_{2.5}$ were measured in research projects in central and eastern Europe in the 1990s (Hoek et al., 1997; Houthuijs et al., 2001). A series of studies in eastern Germany reported a significant decline in particle mass concentration, accompanied by an increase in concentrations of ultrafine particles (Kreyling et al., 2003).

Longer trends are available for sulfate, although sampling artefacts complicate the assessment (Tørseth et al., 2012). Consistent with the large reduction in SO_2 emissions, sulfate concentrations decreased by 70% between 1980 and 2009, mostly between 1990 and 2009 (56% reduction). Nitrate concentrations decreased by much less than sulfates (8% between 1990 and 2009), reflecting the smaller reduction in precursor emissions and a shift in the equilibrium with ammonia and nitric acid towards particulate nitrate (Tørseth et al., 2012).

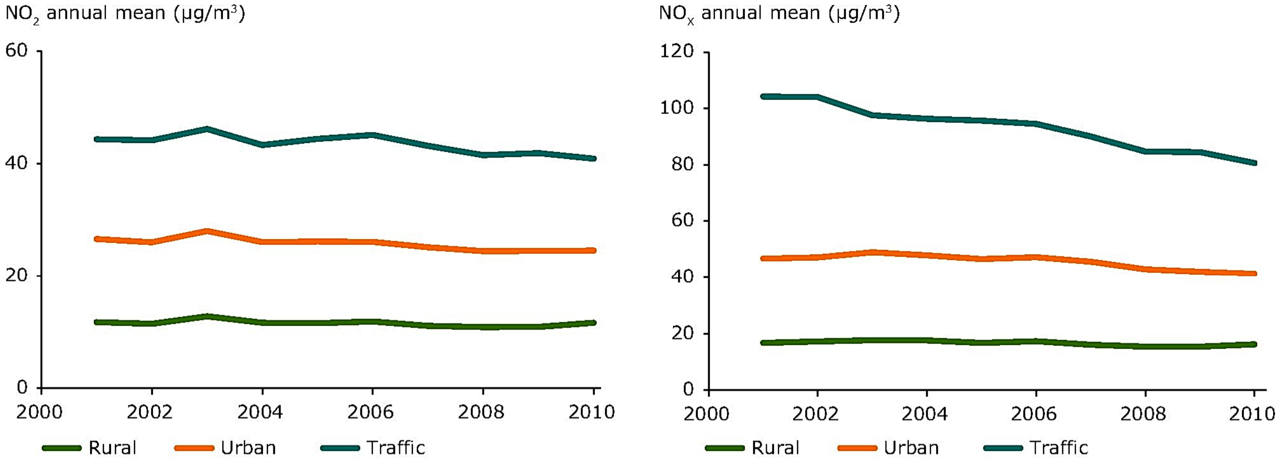
Annual average concentrations of NO_2 have remained fairly stable since 2000, whereas NO_x concentrations did decrease substantially at traffic sites (Fig. 1.24). At the EMEP regional

Fig. 1.23 Trends in annual average concentrations of PM_{10} (2001–2010) and $PM_{2.5}$ (2005–2010) by station type across Europe



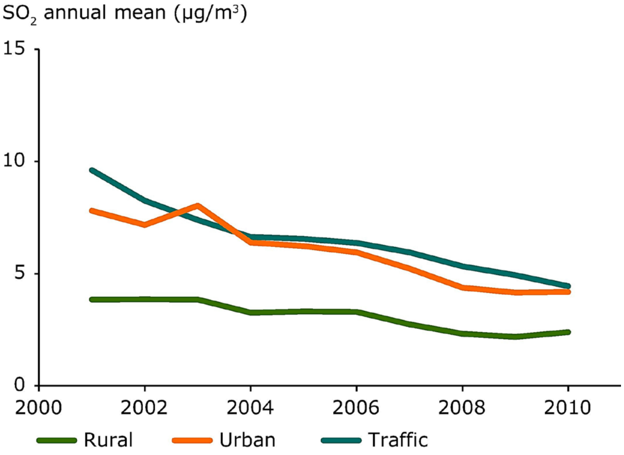
PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm.
All stations in European Union Member States with at least 75% data coverage for at least 8 years (PM_{10}) or 6 years ($PM_{2.5}$) were included in the analysis. Concentrations by station type are given in µg/m³.
Reprinted from EEA (2012). Air quality in Europe – 2012 report. European Environment Agency Report 4/2012.

Fig. 1.24 Trends in NO_2 and NO_x annual mean concentrations (2001–2010) by station type across Europe



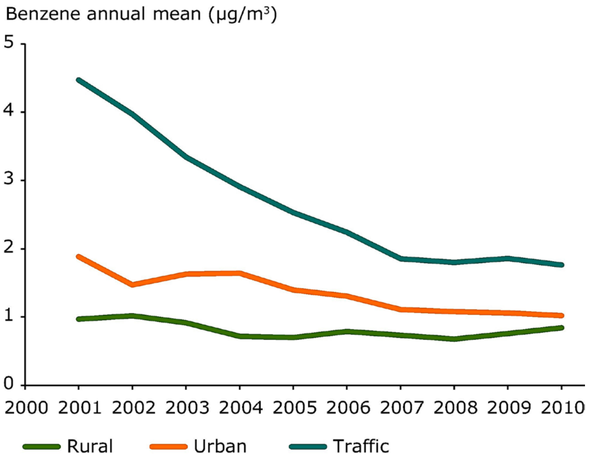
NO_2, nitrogen dioxide; NO_x, nitrogen oxides.
Reprinted from EEA (2012). Air quality in Europe – 2012 report. European Environment Agency Report 4/2012.

Fig. 1.25 Trend in annual average SO_2 concentrations (2001–2010) by station type across Europe



SO_2, sulfur dioxide.
All stations in European Union Member States with at least 75% data coverage for at least 8 years were included in the analysis.
Reprinted from EEA (2012). Air quality in Europe – 2012 report. European Environment Agency Report 4/2012.

Fig. 1.26 Trend in annual average mean benzene concentrations (2001–2010) by station type across Europe



All stations in European Union Member States with at least 75% data coverage for at least 8 years were included in the analysis.
Reprinted from EEA (2012). Air quality in Europe – 2012 report. European Environment Agency Report 4/2012.

background sites, NO_2 concentrations decreased by 23% between 1990 and 2009 (Tørseth et al., 2012). The decrease in NO_x concentrations is explained by lower emissions from motorized traffic, since NO_x emissions in Europe had increased, especially until about 1990, because of emissions from road transportation (Vestreng et al., 2009). The reduced fuel consumption and early technological changes in western Europe between 1980 and 1990 were not sufficiently effective to reduce emissions (Vestreng et al., 2009). After 1990, emissions decreased because of new technologies in western Europe and the economic recession in eastern Europe, while increasing car ownership in eastern Europe resulted in increased road traffic emissions from that region (Vestreng et al., 2009).

The limited decrease in NO_2 concentrations is due to an increase in primary NO_2 in road traffic emissions. Primary NO_2 emissions have gained importance compared with the ozone/NO_x equilibrium (Keuken et al., 2009; Mavroidis & Chaloulakou, 2011). The increase in primary NO_2 emissions has been attributed to increased use of diesel-powered vehicles, which emit a higher fraction of NO_2 compared with gasoline-powered vehicles (Grice et al., 2009; Anttila et al., 2011; Carslaw et al., 2011). In addition, the aftertreatment devices (such as oxidation catalysts) implemented for reducing PM emissions by diesel vehicles contribute to the increasing fraction of primary NO_2 in NO_x (Mavroidis & Chaloulakou, 2011; Williams & Carslaw, 2011). For diesel-fuelled vehicles equipped with catalytic diesel particulate filters, primary NO_2 fractions of about 40–50% are reported (Carslaw et al., 2007). A consequence of this trend is that the value of NO_2 as a marker of the mixture of traffic-related pollutants may have changed.

Concentrations of SO_2 have continued to decrease significantly in Europe at traffic sites, urban background sites, and regional background sites (Fig. 1.25). On average, concentrations were halved between 2000 and 2010

(EEA, 2012). Compared with the early 1990s, concentrations have decreased several-fold, due to significant reductions in the use of coal for power generation and other sources such as domestic heating, lower sulfur content in fuel, and substantial technological developments such as desulfurization at power plants (Tørseth et al., 2012). At the EMEP regional background sites, SO_2 concentrations decreased by 92% between 1980 and 2009, mostly between 1990 and 2009 (75% reduction) (Tørseth et al., 2012). Modest reductions in emissions between 1980 and 1989 occurred largely in western Europe, whereas large reductions between 1990 and 1999 occurred mainly in central and eastern Europe (Vestreng et al., 2007). Important factors were the drop in industrial activity in eastern Europe after the political changes in 1989 and a switch from solid fuel to oil and natural gas containing lower amounts of sulfur (Vestreng et al., 2007).

Concentrations of benzene have decreased substantially in the past decade, especially at traffic sites (Fig. 1.26). The main explanation for this trend is the lower benzene content of gasoline.

There are insufficient data from networks to specify a Europe-wide trend for B[*a*]P concentrations (EEA, 2012), although there are studies from selected locations. A study in Munich showed a decrease in concentrations by an order of magnitude between 1981 and 2001, with most of the change occurring before 1993 (Schauer et al., 2003). Large decreases in PAH concentrations have also been reported for the United Kingdom (Brown et al., 2013). Comparison of data from different sites in London showed a decrease in B[*a*]P concentrations from 10–100 ng/m^3 in the 1950s to less than 0.1 ng/m^3 currently. Median B[*a*]P concentrations of all sites in the current PAH network have decreased from about 1.4 ng/m^3 to 0.2 ng/m^3 (Brown et al., 2013). The decline in the past two decades was attributed to dramatically reduced emissions from industrial metal processing and a ban on burning agricultural stubble (Brown et al., 2013).

Overall, air quality has generally improved in Europe, and the mixture has clearly changed in composition.

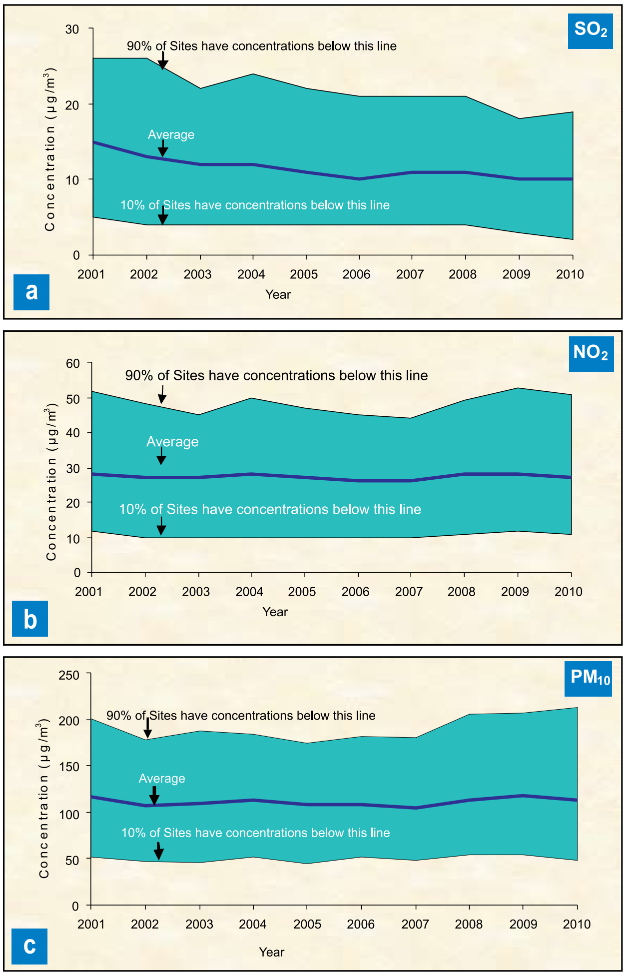
(c) Asia

(i) India

Outdoor air quality information in India is collected primarily by the National Air Quality Monitoring Programme (NAMP). Administered by the Central Pollution Control Board (CPCB), Ministry of Environment and Forests, Government of India, the NAMP network was initiated in 1984 with seven stations in the cities of Agra and Anpara (situated close to the National Capital Region). This network has steadily grown to include nearly 503 outdoor air quality monitoring stations across 209 cities in 26 states and 5 union territories in 2011. Criteria air pollutants listed under the earlier 1994 NAAQS and monitored under the NAMP include $PM_{10,}$ SO_2, and NO_2. Integrated 8-hour and 24-hour measurements are performed twice a week, resulting in about 104 observations from each station annually. In addition, CO, NH_3, lead, and ozone are monitored at selected locations. The NAAQS were recently revised (CPCB, 2009b). $PM_{2.5}$ and air toxics such B[*a*]P, arsenic, and nickel are now included in the revised NAAQS and are slowly being added to the routine monitoring performed under the NAMP. The CPCB collates the data received by the entire network in the central Environmental Data Bank. After completion of quality assurance/quality control, these data are made available in the public domain. This section summarizes pollutant-specific information available from the CPCB, with additional details from relevant published studies where available.

Analyses of CPCB data from 402 stations on criteria air pollutants for the 10-year period 2000–2010 indicate a decline in the national

Fig. 1.27 National mean concentrations derived from data across National Air Quality Monitoring Programme (NAMP) stations together with the 10th and 90th percentile for SO_2 (a), NO_2 (b), and PM_{10} (c) in India



NO_2, nitrogen dioxide; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; SO_2, sulfur dioxide.
Reprinted from CPCB (2012).

annual average SO_2 concentration; NO_2 levels remained largely unchanged, and PM_{10} levels showed a modest increase (Fig. 1.27). Although monitoring locations do not cover all cities across India, they do provide coverage across all states, indicating the extent of exposures that urban populations are likely to experience (CPCB, 2012).

The CPCB classifies the air quality at NAMP locations into four broad categories – low (acceptable), moderate, high, and critical levels of pollution – based on the exceedance factor (the ratio of annual mean concentration of a pollutant to that of the respective standard), as shown in Table 1.9.

By these criteria, the levels of SO_2 at most locations have not only declined but are mostly low across the locations monitored, whereas NO_2 and PM_{10} levels have remained moderately to critically high across many locations over the

Table 1.9 India Central Pollution Control Board (CPCB) criteria for classification of pollution levels

Pollution level	Annual mean concentration range (µg/m³)		
	SO_2	NO_2	PM_{10}
Low (L)	0–25	0–20	0–30
Moderate (M)	26–50	21–40	31–60
High (H)	51–75	41–60	61–90
Critical (C)	> 75	> 60	> 90

NO_2, nitrogen dioxide; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; SO_2, sulfur dioxide.
Adapted from CPCB (2012).

years (Fig. 1.28). Maximum levels in the most polluted states/cities often exceed the NAAQS by 2–5-fold, as may be seen in Table 1.10.

Limited information is currently available on chemical speciation of PM fractions or the differential distribution of PM and gaseous pollutants in relation to land use. In a recent national source apportionment study performed across six cities (CPCB, 2011), levels of PM_{10} and $PM_{2.5}$ in the outdoor air were consistently in excess of the NAAQS across background, kerbside, industrial, commercial, and residential sites, and winter and post-monsoon season levels were much higher than summer levels (CPCB, 2011). NO_2 levels were of concern at several locations, whereas SO_2, ozone, and CO levels were generally within the prescribed standards. Results from analyses of PM components indicate that EC and OC accounted for 20–45% of PM_{10} and 25–75% of $PM_{2.5}$ in cities. SO_4^{2-} and NO_3^- accounted for 10–30% of PM_{10} in cities. Vehicle exhaust, secondary particulates, construction activities, oil burning (e.g. diesel or heavy oil), biomass burning, coal combustion, kerosene combustion, and industrial emissions have been identified to be the dominant sources for criteria air pollutants in these cities (CPCB, 2011).

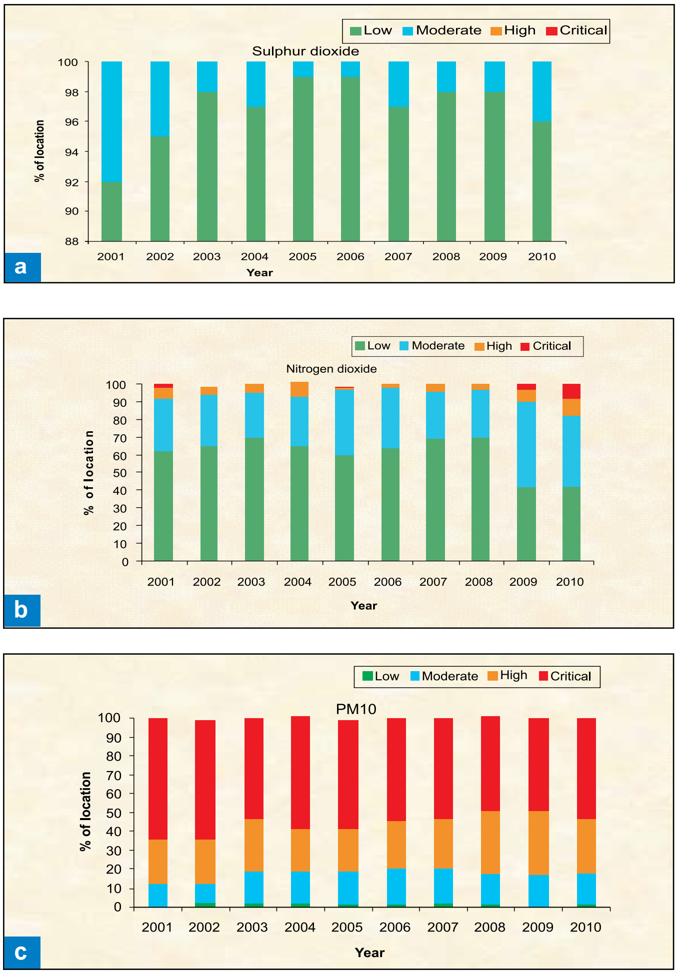
In addition, several industrial hotspots have been identified by the CPCB using the new risk assessment criteria of the Comprehensive Environmental Pollution Index (CEPI) (CPCB, 2009a). The CEPI weights the toxicity of the agents, the volume of emissions, the scale of the population exposed, and the exposure pathways involved. Of special relevance to carcinogenicity is the fact that unlike criteria air pollutant data provided by the NAMP, the CEPI includes weighted contributions from a range of compounds including probable carcinogens (US EPA Class 2 and 3 or substances with some systemic toxicity, such as VOCs, PAHs, and PCBs) as well as known carcinogens or chemicals with significant systemic or organ system toxicity (such as vinyl chloride, benzene, lead, radionuclides, hexavalent chromium, cadmium, and organophosphates) (CPCB, 2009a).

Data from the NAMP network of the CPCB provide the most comprehensive description of the status of air quality across Indian cities as far as criteria air pollutants are concerned. Air toxics are seldom monitored routinely, and hence information on air toxics is mostly contained in individual studies conducted by academic and/or research organizations.

(ii) China

As a result of the unprecedented rapid development in industrialization and urbanization in the past decades, many Chinese cities have air pollution levels well above health-based standards (HEI, 2010b; Gao et al., 2011), and air pollution associated with health impacts has become a growing concern (Zhang et al., 2010a). In this section, information on outdoor concentration

Fig. 1.28 Trends in pollution levels for 2000–2010 in India in relation to Central Pollution Control Board (CPCB) criteria given in Table 1.9



Reprinted from CPCB (2012).

levels, spatial variation, and time trends in major outdoor air pollutants is summarized. Data are extracted primarily from publications on air pollution and epidemiological research conducted in China, as well as from government routine monitoring networks.

In the last century, air pollution from coal combustion was the dominant type of air pollution in most cities in China, and the air pollution was severe. Various pollution control measures and devices have been gradually put in place for the industrial and residential sectors.

Coal will remain the major energy source in China for the near future. However, in recent years, outdoor air pollution in most Chinese cities has become a mixture of emissions from coal combustion, vehicles, and biomass burning, as well as from sandstorms in the north-western region (HEI, 2010b). The annual average levels of PM_{10}, SO_2, and NO_2 in 31 provincial capital cities in China are summarized in Fig. 1.29. The concentrations of PM_{10}, SO_2, and NO_2 in most large urban areas in China have generally stabilized or are decreasing (albeit with some

Table 1.10 Profile of the 10 most polluted Indian cities in 2010[a]

State	City	Minimum (µg/m³)	Maximum (µg/m³)	Annual average (µg/m³)	Standard deviation (µg/m³)	Air quality[b]
PM_{10} concentrations						
Madhya Pradesh	Gwalior	598	114	308*	107	C
Jharkhand	West Singhbhum	59	926	302*	229	C
Uttar Pradesh	Ghaziabad	162	510	290*	89	C
Chhattisgarh	Raipur	207	370	289*	39	C
Delhi	Delhi	46	748	261*	130	C
Haryana	Yamuna Nagar	64	523	261*	116	C
Jharkhand	Jharia	131	370	237*	40	C
Punjab	Khanna	152	283	231*	23	C
Punjab	Gobindgarh	125	534	224*	66	C
Punjab	Amritsar	181	258	219*	20	C
NO_2 concentrations						
West Bengal	Howrah	37	147	75*	25	C
West Bengal	Barrackpore	39	140	74*	24	C
Maharashtra	Badlapur	9	175	73*	37	C
Maharashtra	Ulhasnagar	8	162	68*	33	C
West Bengal	Durgapur	42	91	66*	11	C
West Bengal	Asansol	46	88	66*	10	C
West Bengal	Sankrail	28	120	65*	22	C
West Bengal	Raniganj	45	85	63*	10	C
West Bengal	Kolkata	23	142	62*	27	C
West Bengal	South Suburban	25	113	56*	23	C
SO_2 concentrations						
Jharkhand	Jamshedpur	27	42	35.4	1	M
Jharkhand	Saraikela Kharsawan	28	41	35	3	M
Maharashtra	Badlapur	5	86	32.3	15	M
Goa	Mormugao	7	253	31.8	35	M
Maharashtra	Ulhasnagar	5	109	31.2	17	M
Uttar Pradesh	Ghaziabad	21	37	30.3	3	M
Uttar Pradesh	Khurja	21	40	29.2	4	M
Maharashtra	Pune	10	96	28.7	15	M
Maharashtra	Chandrapur	12	35	21.3	4	L
Jharkhand	West Singhbhum	15	36	21	3	L

NAAQS, National Ambient Air Quality Standards; NO_2, nitrogen dioxide; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; SO_2, sulfur dioxide.

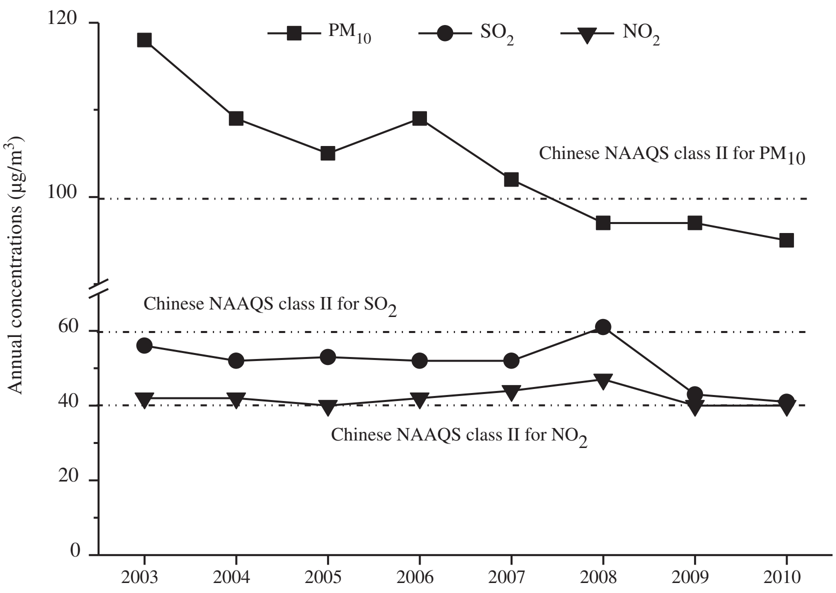
[a] Asterisks indicate cities where annual mean concentration exceeded the NAAQS of 60 µg/m³ (PM_{10}) or 40 µg/m³ (NO_2) or 50 µg/m³ (SO_2) for residential, industrial, and other areas.

[b] Classification based on criteria in Table 1.9: L, low; M, moderate; H, high; C, critical.

Compiled from CPCB (2012).

notable exceptions). However, along with the reductions in concentrations of PM_{10}, SO_2, and NO_2 in China, the pollution episodes of $PM_{2.5}$ and ozone in some city cluster areas suggest the degradation of regional air quality. As total air pollution sources and overall emissions increase in China, yet become more dispersed, regional and transboundary air quality issues are likely to become increasingly important. The mean concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2, and

Fig. 1.29 Annual average levels of PM_{10}, SO_2, and NO_2 in 31 provincial capital cities in China, 2003–2010



NO_2, nitrogen dioxide; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; SO_2, sulfur dioxide.
The dotted-dashed line indicates the annual level of the Chinese National Ambient Air Quality Standards (NAAQS) class II.
Reprinted from Shang et al. (2013). Systematic review of Chinese studies of short-term exposure to air pollution and daily mortality. *Environ Int*, 54:100–11. doi:10.1016/j.envint.2013.01.010 PMID:23434817, © with permission from Elsevier.

ozone reported in time-series studies conducted in China from 1990 to 2012 are presented in Table 1.11.

PM_{10}

As a result of energy restructuring, the annual average levels of PM_{10} in 31 provincial capital cities in China decreased by about 25% from 2003 to 2010 (Fig. 1.29); however, the levels are still high compared with elsewhere in the world. In 2010, the annual concentrations of PM_{10} were 121 µg/m³ in Beijing, 79 µg/m³ in Shanghai, 69 µg/m³ in Guangzhou, and 126 µg/m³ in Xi'an (National Bureau of Statistics of China, 2011). The spatial variations of PM_{10} in major Chinese cities suggest more serious particulate pollution in northern regions in China, due to the longer heating season in winter as well as the local topography and the impact of sandstorms. The nationwide distribution of air pollution levels is likely to be related to the spatial distribution of emission sources across the country (National Bureau of Statistics of China, 2012).

SO_2

Trends in air quality in 31 provincial capital cities in China from 2003 to 2010 suggest a significant decrease of about 30% in annual average SO_2 concentrations in urban areas, with the exception of an average increase in SO_2 concentration during 2008 (Fig. 1.29). The reductions in SO_2 have resulted from the use of low-sulfur fuels and the relocation of major coal-fired power plants and industrial facilities from urban areas to outside cities. In more recent years, annual levels of SO_2 were below 60 µg/m³ in most cities, and PM_{10} concentration levels continued to decrease (National Bureau of Statistics of China, 2012).

Table 1.11 Mean concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2, and O_3 reported in time-series studies conducted in China (1990–2012)

City, year(s)	Pollutant[a]					Reference
	PM_{10}	$PM_{2.5}$	SO_2	NO_2	O_3	
Beijing, 2003	141 (79)	—	60 (56)	—	—	Pan et al. (2008)
Beijing, 2004–2008	146 (92)	—	49 (49)	64 (26)	—	Zhang et al. (2010b)
Beijing, 2007–2008	172 (93)	82 (52)	—	—	—	Chen et al. (2011a)[b]
Shanghai, 2000–2001	91 (52)	—	43 (20)	33 (14)	—	Kan & Chen, (2003)
Shanghai, 2001–2004	102 (2)	—	45 (1)	67 (1)	63 (1)	Kan et al. (2008)
Shanghai, 2001–2004	102 (65)	—	45 (24)	67 (25)	63 (37)	Zhang et al. (2006b)
Shanghai, 2002–2003	112 (76)	69 (48)	38 (21)	59 (23)	—	Dai et al. (2004)[b]
Shanghai, 2004–2005	108 (2)	56 (1)	58 (1)	62 (1)	77 (3)	Huang et al. (2009)[b]
Shanghai, 2004–2005	108(2)	57 (1)	—	—	65 (3)	Kan et al. (2007)[b]
Shanghai, 2004–2008	105 (54)	55 (30)	—	—	—	Chen et al. (2011a)[b]
Shanghai, 2006–2008	86 (53)	—	53 (30)	56 (21)	—	Chen et al. (2011b)[b]
Guangzhou, 2004–2008	81 (45)	—	54 (36)	67 (30)	—	Huang et al. (2012b)
Guangzhou, 2006–2009	60 (24)	—	43 (21)	48 (26)	—	Yu et al. (2012)
Guangzhou, 2007–2008	—	70 (35)	50 (32)	66 (31)	—	Yang et al. (2012a)[b]
Tianjin, 2005–2007	105 (57)	—	68 (54)	47 (18)	—	Zhang et al. (2010c)
Hong Kong Special Administrative Region, 1995–1998	52 (25)	—	17 (12)	56 (20)	34 (23)	Wong et al. (2002)
Hong Kong Special Administrative Region, 1996–2002	52 (25)	—	18 (12)	59 (20)	37 (23)	Wong et al. (2008a)
Wuhan, 2000–2004	142 (64)	—	44 (25)	52 (19)	78 (41)	Qian et al. (2007)
Wuhan, 2001–2004	142	—	39	52	86	Wong et al. (2008b)
Pearl River Delta, 2006–2008	78	—	62	53	80	Tao et al. (2011)[b]
Xi'an, 2004–2008	131 (55)	—	48 (29)	39 (15)	—	Hou et al. (2011)
Xi'an, 2004–2008	—	177 (104)	—	—	—	Huang et al. (2012a)[b]
Anshan, 2004–2006	111 (60)	—	59 (74)	26 (16)	—	Chen et al. (2010)
Chongqing, 1995	—	147	213	—	—	Venners et al. (2003)[b]
Suzhou, 2006–2008	—	—	—	—	58 (40)	Yang et al. (2012b)
Hangzhou, 2002–2004	113	—	46	53	—	Ren et al. (2007)
Shenyang, 2006–2008	141 (66)	94 (52)	—	—	—	Chen et al. (2011a)[b]
Taiyuan, 2004–2008	132 (65)	—	77 (8)	23 (9)	—	Chen et al. (2011b)

NO_2, nitrogen dioxide; O_3, ozone; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; SO_2, sulfur dioxide.

[a] Mean concentrations are given in µg/m³; when available, standard deviations are given in parentheses.

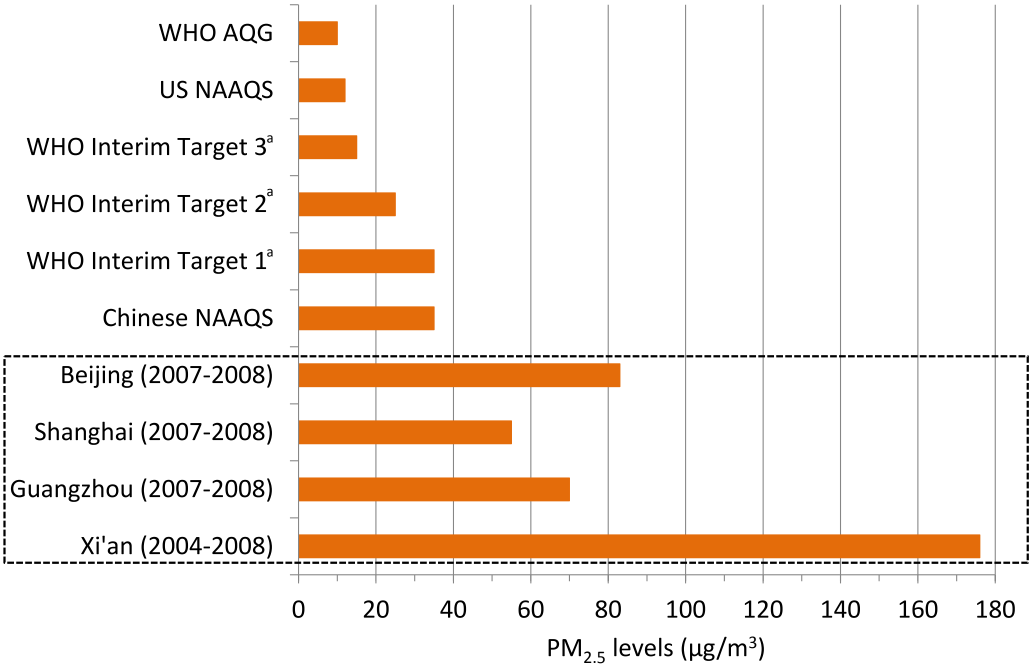
[b] Air pollution data collected not by state routine monitoring reporting system but by environmental science investigators.

Prepared by the Working Group.

NO_2

Due to tightened motor vehicle emission standards in place from the early 2000s, annual average NO_2 levels remained stable at 40 µg/m³, with some variations (Fig. 1.29). However, with the increasing numbers of motor vehicles in most Chinese cities, NO_2 levels in the more developed cities tend to be higher, at more than 45 µg/m³ (National Bureau of Statistics of China, 2012; Table 1.11).

Fig. 1.30 Comparisons of reported annual $PM_{2.5}$ levels (µg/m³) in Beijing, Shanghai, Guangzhou, and Xi'an with the Chinese national standards and international air quality standards



AQG, air quality guidelines; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter <2.5 µm; NAAQS, National Ambient Air Quality Standards.

[a] In addition to guideline values, WHO has proposed interim targets for each air pollutant in 2005. "These interim targets are proposed as incremental steps in a progressive reduction of air pollution, and are intended for use in areas where pollution is high. … Progress towards the guideline values should, however, be the ultimate objective of air quality management and health risk reduction in all areas" (WHO, 2006).

Prepared by the Working Group based on data from China Statistical Yearbook 2008–2009.

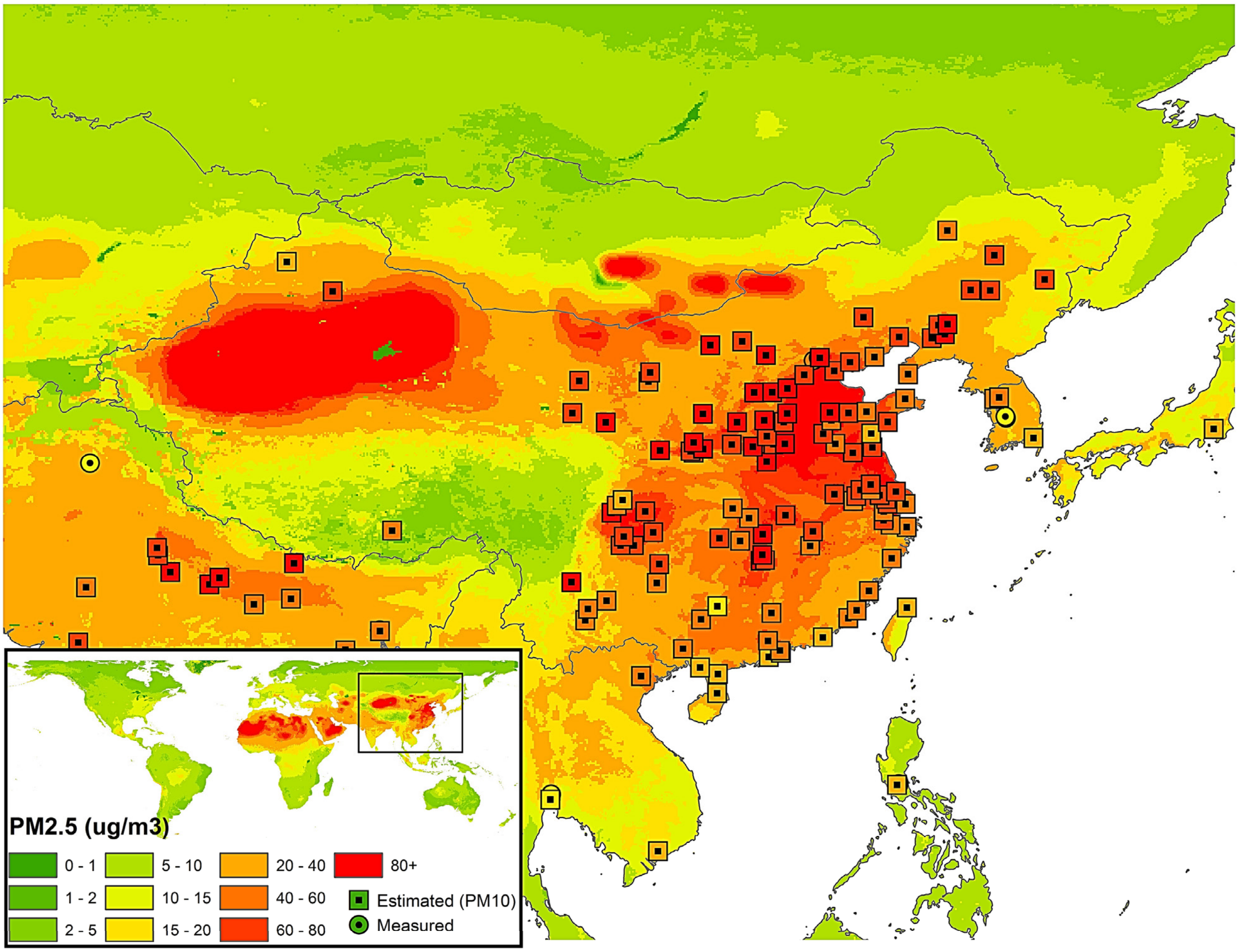
$PM_{2.5}$ and ozone

At present, very limited data are available on the annual levels of $PM_{2.5}$ and ozone, which were newly included in the revised Chinese AAQS released in March 2012 (MEPPRC, 2012).

To assess exposure, time-series studies have been conducted (Shang et al., 2013). The reported average concentrations of $PM_{2.5}$ and 8-hour ozone in these studies were in the ranges of 55–177 µg/m³ and 34–86 µg/m³, respectively (Table 1.11). In these studies, the reported $PM_{2.5}$ levels in Beijing, Shanghai, Guangzhou, and Xi'an were all well above the Chinese national standards and international air quality standards (Fig. 1.30). Brauer et al. (2012) estimated that the population-weighted annual average levels of $PM_{2.5}$ in East Asia had increased from 43 µg/m³ to 55 µg/m³ between 1990 and 2005, whereas they reported the highest measurement of annual average $PM_{2.5}$ concentration (in 2005) of 58 µg/m³ in Beijing and the highest derived $PM_{2.5}$ concentration (calculated from PM_{10} measurements) of 121 µg/m³ in Datong, a coal-mining centre in Shanxi Province (Brauer et al., 2012). In northern China, estimated $PM_{2.5}$ levels in 2010 were above 80 µg/m³ (Fig. 1.31).

Geological materials, organic materials, EC, and secondary aerosols (such as SO_4^{2-}, NO_3^-, and NH_4^+) are the primary components of $PM_{2.5}$ in China; however, due to source variations, the concentrations of primary $PM_{2.5}$ components vary significantly across locations and seasons (Niu et al., 2006; Cao et al., 2012). On average, SO_4^{2-}, NO_3^-, NH_4^+, organic materials, and EC

Fig. 1.31 Estimated levels of $PM_{2.5}$ (µg/m³) in 2010 in China



$PM_{2.5}$, particulate matter with particles of aerodynamic diameter <2.5 µm.
Compiled by the Working Group with data from Brauer et al. (2012).

account for more than 70% of the $PM_{2.5}$ mass in summer, whereas the percentage is even higher in winter (Cao et al., 2012).

Lead levels are still high in Chinese cities, reaching an average of 1.68 µg/m³ in Xi'an during winter. High correlations of lead with arsenic and SO_4^{2-} concentrations indicate that much of the lead derives from coal combustion rather than from leaded fuels, which were phased out by 2000 in China. Although limited fugitive dust markers were available, scaling of iron by its ratios in source profiles showed that in most of the cities, 20% of $PM_{2.5}$ derives from fugitive dust (Cao et al., 2012).

Photochemical smog, in the presence of solar radiation, is commonplace in city cluster areas of China with greatly increased numbers of vehicles (e.g. the Beijing–Tianjin–Hebei area and the Pearl River Delta region). Studies in these areas reported high concentrations of PM induced by photochemical smog. For example, in Shenzhen in 2004, the 24-hour average $PM_{2.5}$ and PM_{10} concentrations in summer were 35 µg/m³ and 57 µg/m³, respectively, and in winter were 99 µg/m³ and 137 µg/m³, respectively (Niu et al., 2006). In

Guangzhou, the summer 24-hour average $PM_{2.5}$ concentration was 97.5 µg/m³ (Wang et al., 2006).

Polycyclic aromatic hydrocarbons

Daily and hourly average or snapshot concentrations of outdoor PAHs in urban and industrial areas in China are high compared with elsewhere in the world (usually 10–20 ng/m³). Mean concentrations of 16 outdoor PAHs of up to 1400 µg/m³ were observed in Taiyuan, a coal-polluted city in central China, in December 2006 (Fu et al., 2010). Concentrations of PAHs in the gas phase were also reported at high levels, in particular in megacities and large cities (e.g. Beijing, Shanghai, and Hangzhou) (Liu et al., 2001, 2007; Wang et al., 2002; Zhang et al., 2009b; Zhu et al., 2009; Wei et al., 2011).

Volatile organic compounds

High daily and hourly average or snapshot concentrations of outdoor benzene, toluene, and xylene have been reported in Chinese megacities (e.g. Beijing, Shanghai, and Guangzhou) compared with the levels observed in the USA (Zou et al., 2003; Zhang et al., 2006a; Wei et al., 2007; Lu et al., 2008; Wang et al., 2010; Zhou et al., 2011).

(iii) Japan

As one of the most developed countries in Asia, Japan experienced serious pollution from industrial and automobile emissions in the 1950s and 1960s, and the main energy source shifted from coal to oil, making SO_2 a major air pollutant (Committee on Japan's Experience in the Battle Against Air Pollution, 1997). Air pollution levels declined after the introduction of pollution control measures in the 1970s. As an example, the nationwide annual average concentrations of SO_2 decreased to 0.015 ppm [42.3 µg/m³] in the 1970s and further to 0.006 ppm [16.9 µg/m³] in 1990 (Ministry of the Environment of Japan, 2011).

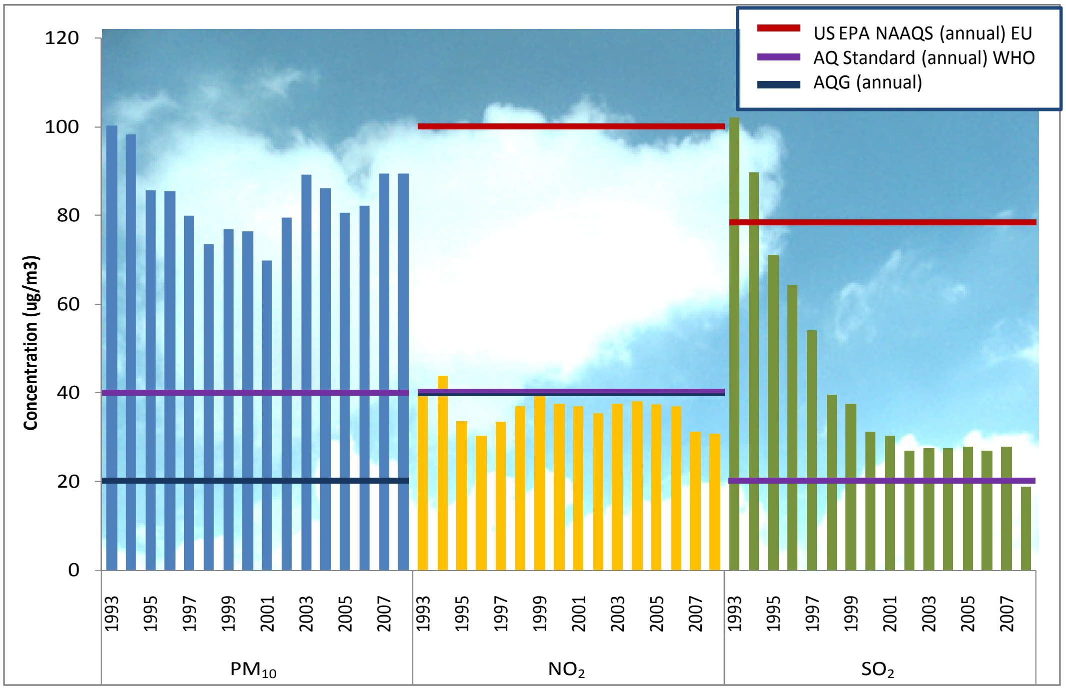
In contrast to the rapid decline in the concentrations of SO_2, pollution from mobile sources increased during the 1970s. The annual concentrations of NO_2 in 1970 were 0.035 ppm [70.9 µg/m³] at general sites and 0.042 ppm [85.1 µg/m³] at roadside sites; those of suspended PM (PM < 7 µm in diameter [SPM]) in 1975 were 50 µg/m³ at general sites and 84 µg/m³ at roadside sites (Ministry of the Environment of Japan, 2011). After the tightened mobile-source emission control regulations and measures were put in place, the concentrations of NO_2 and SPM declined gradually.

In 2011, the annual concentrations of major air pollutants in Japan were as follows: SO_2, 0.002 ppm [5.64 µg/m³] at general sites and 0.003 ppm [8.46 µg/m³] at roadside sites; NO_2, 0.011 ppm [22.3 µg/m³] at general sites and 0.021 ppm [42.5 µg/m³] at roadside sites; SPM, 20 µg/m³ at general sites and 22 µg/m³ at roadside sites; $PM_{2.5}$, 15.4 µg/m³ at general sites and 16.1 µg/m³ at roadside sites; and CO, 0.3 ppm [370 µg/m³] at general sites and 0.5 ppm [617 µg/m³] at roadside sites (Ministry of the Environment of Japan, 2011). In recent years, in addition to making the necessary efforts towards reducing the concentrations of these pollutants, Japan has also faced problems such as relatively high and stable concentrations of ozone in metropolitan areas (e.g. annual concentration of 0.028 ppm [59.2 µg/m³] in Tokyo in 2011) (Bureau of Environment of Tokyo, 2013).

(iv) Other Asian countries

Since the 1990s, most Asian countries have established national routine air quality monitoring networks for the criteria pollutants PM_{10}, SO_2, and NO_2, whereas the air quality data on $PM_{2.5}$ and ozone have been very limited. In the cities with routine air quality monitoring systems, some improvements in air quality have been achieved in the past decades; however, the levels of PM_{10} and SO_2 still exceed the World Health Organization (WHO) air quality guidelines (AQG) (Fig. 1.32; Clean Air Asia, 2010). PM_{10} has been a major pollutant in Asian cities, with

Fig. 1.32 Average of annual average outdoor air quality in selected Asian cities (1993–2008)



NAAQS, National Ambient Air Quality Standards; NO_2, nitrogen dioxide; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; SO_2, sulfur dioxide.
Air quality data are compiled by CAI-Asia Center from official sources (publications, personal communications) for 243 Asian cities, as of April 2010.
The US EPA NAAQS does not have a standard for PM_{10}, whereas the EU and WHO apply the same standards for NO_2 and SO_2.
Reprinted from Clean Air Asia (2010).

annual average PM_{10} concentrations well above the WHO AQG. Since 1995, most Asian cities have reported reduced NO_2 levels, with annual average concentrations below the WHO AQG. For SO_2, the annual average levels have decreased remarkably from the 1990s to the 2000s in most Asian cities, due to energy restructuring in the area.

PM_{10}

As of 2008, PM_{10} was still a major pollutant in Asia; annual average PM_{10} concentrations ranged from 11 µg/m³ to 375 µg/m³ in the 230 Asian cities with the highest levels observed in East and South-East Asia (Fig. 1.33; Clean Air Asia, 2010).

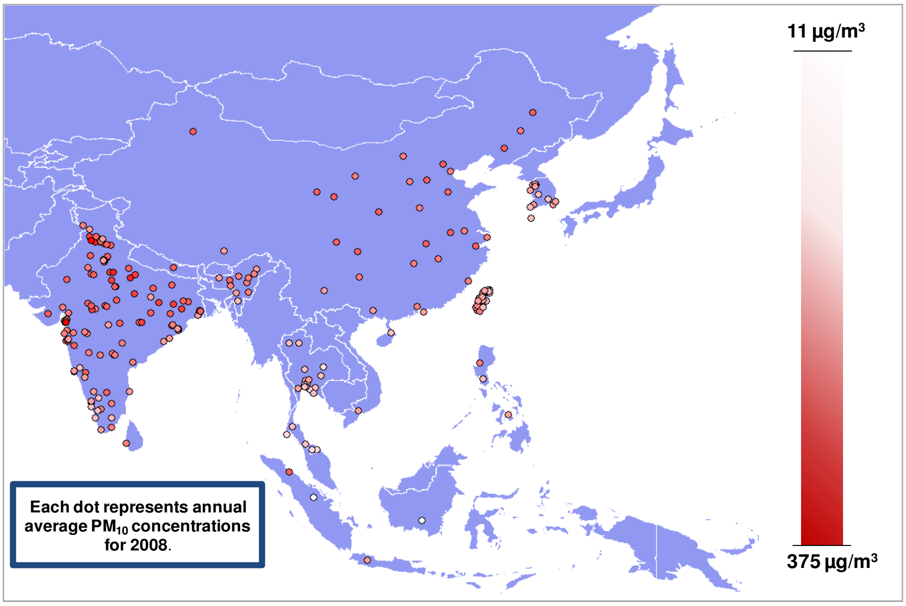
SO_2

In 2008, the monitoring data for 213 Asian cities showed that SO_2 levels were still high in some cities in East Asia, particularly those near industries. Annual average SO_2 concentrations ranged from 1.3 µg/m³ to 105 µg/m³. The mean of annual average SO_2 concentrations for 213 Asian cities was 18.7 µg/m³ in 2008. See Fig. 1.34 (Clean Air Asia, 2010).

NO_2

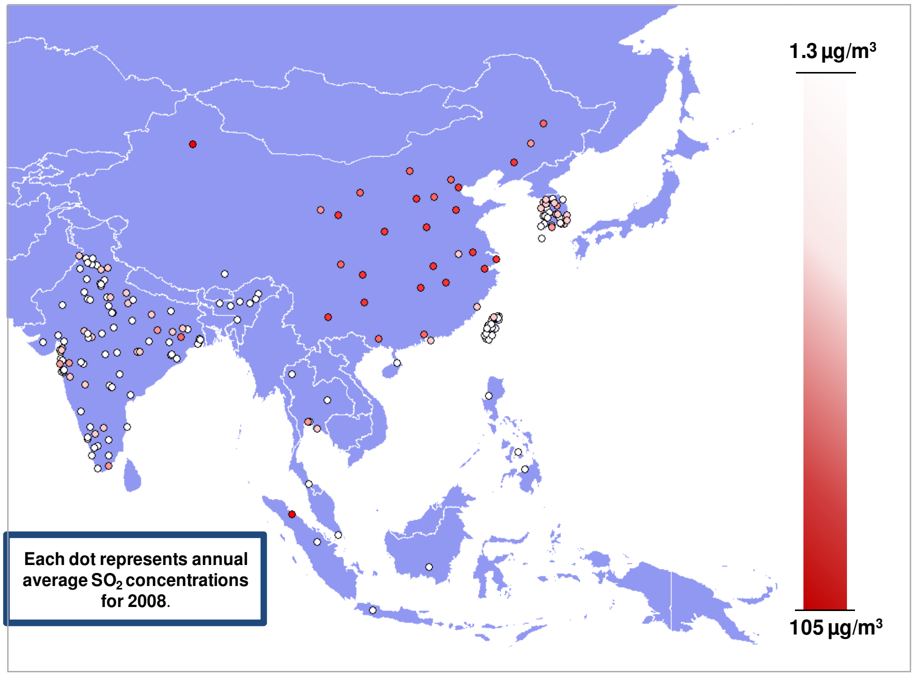
In 2008, annual average NO_2 concentrations ranged from 1.9 µg/m³ to 77 µg/m³ in 234 Asian cities; the mean of annual average NO_2 concentrations was 30.7 µg/m³. About 73% of the 234 cities had annual average NO_2 concentrations below the WHO AQG of 40 µg/m³. See Fig. 1.35 (Clean Air Asia, 2010).

Fig. 1.33 Annual PM_{10} concentrations (µg/m³) in 230 Asian cities



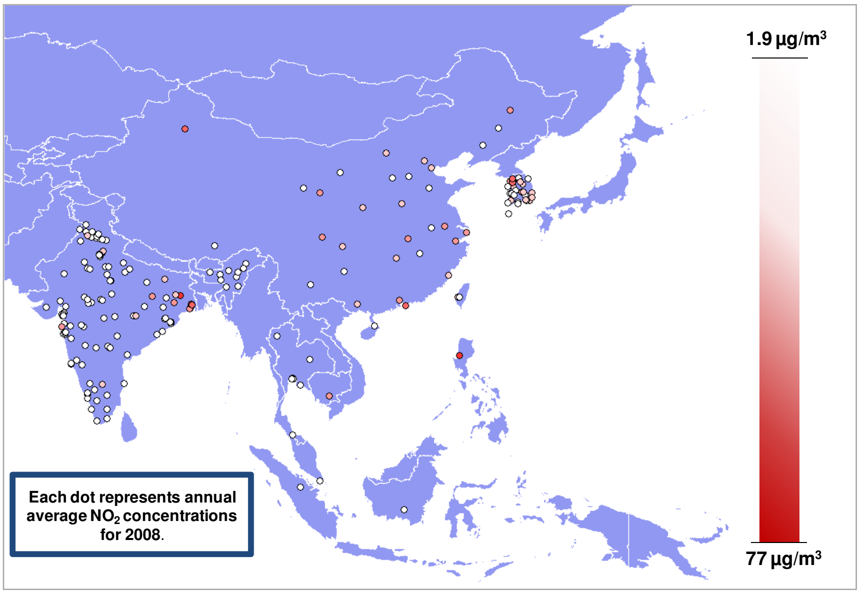
PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm.
Reprinted from Clean Air Asia (2010).

Fig. 1.34 Annual SO_2 concentrations (µg/m³) in 213 Asian cities



SO_2, sulfur dioxide.
Reprinted from Clean Air Asia (2010).

Fig. 1.35 Annual NO_2 concentrations (µg/m³) in 234 Asian cities



NO_2, nitrogen dioxide.
Reprinted from Clean Air Asia (2010).

$PM_{2.5}$

$PM_{2.5}$ levels have increased in medium to large Asian cities. Only a few Asian countries have set $PM_{2.5}$ air quality standards, and of those countries, none have standards equivalent to the WHO AQG, but generally the standards are close to the WHO interim target. The population-weighted annual average concentrations of $PM_{2.5}$ were estimated to range between 16 µg/m³ and 55 µg/m³, with the highest levels observed in East Asia, followed by South Asia, in 2005 (Brauer et al., 2012).

A multicity study examined the seasonal variations of $PM_{2.5}$ mass concentrations and species in mixed urban areas (2001–2004) in six Asian cities: Bandung (Indonesia), Bangkok (Thailand), Beijing (China), Chennai (India), Manila (Philippines), and Hanoi (Viet Nam) (Table 1.12). These cities differed in geographical location, topography, energy use, industry, mix of vehicles, and density. The climate of the region is dominated by monsoons, with two distinct seasons, dry and wet, although each dry and wet season may cover different months of the year in different countries. In these cities, the major components of $PM_{2.5}$ and PM_{10} were found to be organic matter (calculated in this study as 1.7 times the OC content); crustal material, including aluminium, calcium, silicon, titanium, iron, potassium, and their oxides; the secondary aerosols NO_3^- and SO_4^{2-}; and EC/BC. The "trace metals" group included all the remaining elements except crustal elements, sodium, and sulfur. OC was not analysed in most sites, except for the Bangkok Metropolitan Region and Beijing; hence, comparison of OC levels was not possible. In all these cities, the levels of PM_{10} and $PM_{2.5}$ were found to be high, especially during the dry season, frequently exceeding the US EPA standard for PM_{10} and $PM_{2.5}$, especially at the traffic sites (Kim Oanh et al., 2006).

PAHs in particles are also important in Asia. Shen et al. (2013) estimated that Asian countries contributed 53.5% of the global total PAH emissions, with the highest emissions from China (106 Gg) and India (67 Gg) in 2007.

Table 1.12 City-wise average mass and major components of $PM_{2.5}$ (μg/m³) during the dry and wet seasons in six cities in Asia (2001–2004)

Cities, country	Number of samples	Mass[a]	Crustal	Organic matter	Soot	Sea salt	NH_4^+	NO_3^-	SO_4^{2-}	Trace elements	Percentage of mass explained
Dry season $PM_{2.5}$											
Bangkok[b], Thailand	181	50	1.1	21.4	8.2	1.7	1.6	1.2	5.6	0.4	80
Beijing, China	142	168	9.9	64.3	18.7	1.6	12.5	14.2	20.8	1.5	40
Chennai, India	83	46									
Bandung, Indonesia	106	53	2.8	–	9.8	0.7	3.4	5.5	8.2	0.8	59
Manila, Philippines	407	44	1.4	–	21.6	0.9	–	–	(1.5)[c]	0.7	56
Hanoi[c], Viet Nam	75	124	7.5	–	–	1.1	–	–	(6.0)[c]	7.1	18
Wet season $PM_{2.5}$											
Bangkok[b], Thailand	106	18	0.9	–	5.3	1.5	0.5	0.4	2.4	0.3	71
Beijing, China	115	104	4.5	19.2	5.3	0.5	10.4	12.0	17.9	1.0	57
Chennai, India	10	42									
Bandung, Indonesia	38	38	3.1	–	7.5	0.8	3.9	3.5	6.3	1.5	71
Manila, Philippines	376	43	1.4	–	22.7	0.9	–	–	(0.8)	1.6	63
Hanoi[c], Viet Nam	21	33	4.0	–	4.3	–	–	–	–	3.8	47

[a] Average of all sites in the city.

[b] Bangkok metropolitan area.

[c] Hanoi metropolitan region.

Adapted from Kim Oanh et al. (2006).

(d) Other regions

(i) Africa

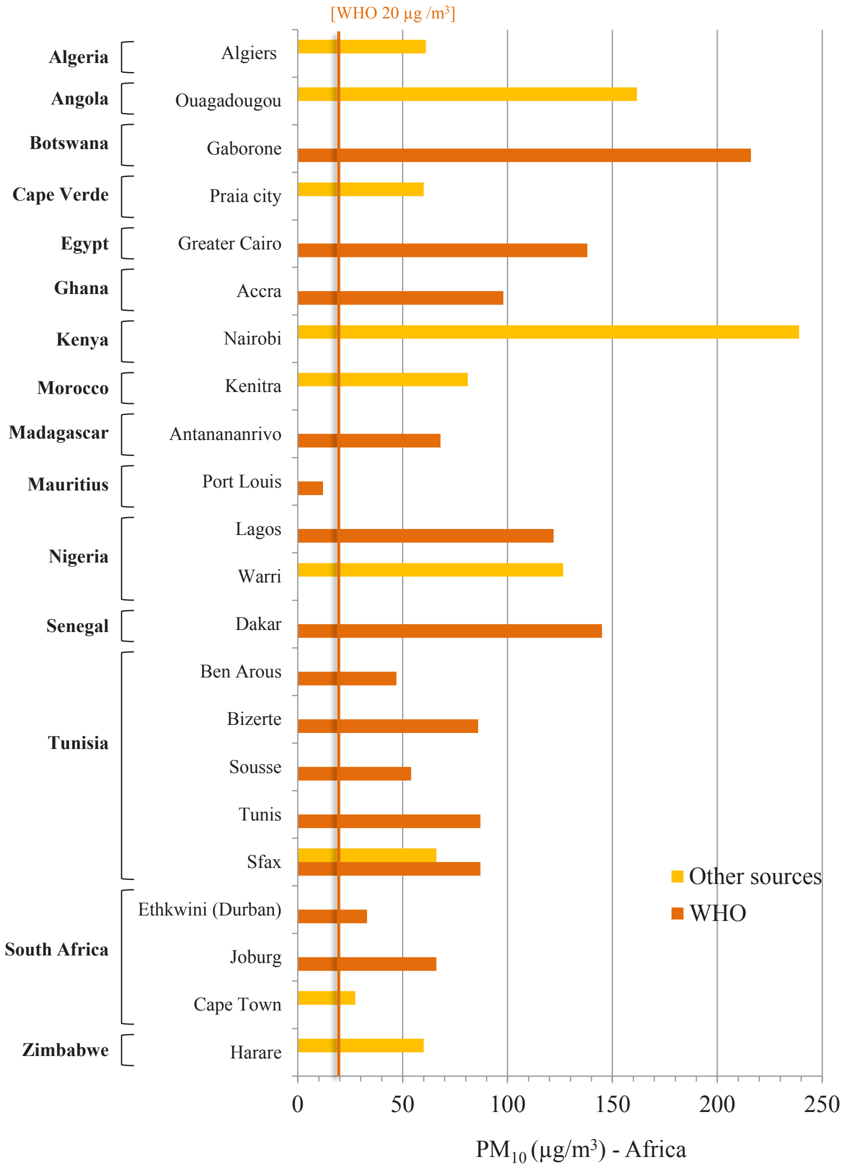
Measurements of air pollution in Africa are limited, and environmental agencies do not exist in all countries. World agencies provide some aggregate information for the continent, which can be complemented by local research, as air quality data in Africa are scarce.

Fig. 1.36 and Fig. 1.37 present the mean concentrations for PM measured with at least 2 months of monitoring coverage in selected African cities. The limited data show that the concentrations range from 7 μg/m³ to more than 100 μg/m³ for $PM_{2.5}$ and from 12 μg/m³ to more than 230 μg/m³ for PM_{10} in the African cities studied. Among the reported air pollution measurement campaigns, Dionisio *et al.* reported the geometric mean concentrations of $PM_{2.5}$ and PM_{10} along the mobile monitoring path [street-level monitoring] of 21 μg/m³ and 49 μg/m³, respectively, in the neighbourhood with the highest socioeconomic status and 39 μg/m³ and 96 μg/m³, respectively, in the neighbourhood with the lowest socioeconomic status and the highest population density in Accra, Ghana. The factors that had the largest effects on local PM pollution were nearby wood and charcoal stoves, congested and heavy traffic, loose-surface dirt roads, and trash burning (Dionisio et al., 2010a).

(ii) South America

Continuous measurements of air pollution are available in more than half of the South American countries. However, the spatial distribution of air monitoring stations in South America is not balanced. For instance, in Brazil monitoring stations are located mostly in large metropolitan regions and do not cover the remaining areas of Brazil; only 8 out of 27 Brazilian states (including the Federal District) have set up air monitoring networks. Fig 1.38 and Fig. 1.39 summarize the most recent data on PM concentrations in South American countries; concentrations ranged from

Fig. 1.36 PM_{10} concentrations (µg/m³) in selected African cities

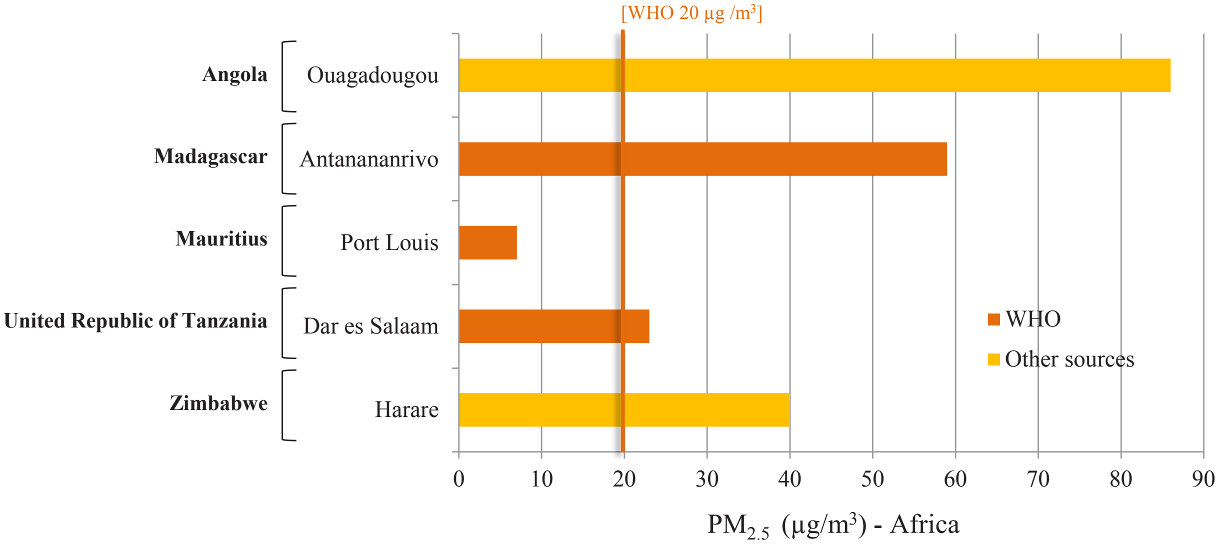


PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm.
Compiled by the Working Group with data from Lindén et al. (2012), WHO (2011), Tchuente et al. (2013), Almeida-Silva et al. (2013), Petkova et al. (2013), Laïd et al. (2006), WHO (2011), Abu-Allaban et al. (2007), Dionisio et al. (2010a, b), Arku et al. (2008), Mkoma et al. (2009, 2010), Wichmann & Voyi (2012), and Kuvarega & Taru (2008).

22 µg/m³ to 70 µg/m³ for PM_{10} and from 7 µg/m³ to 35 µg/m³ for $PM_{2.5}$.

Besides high PM concentrations measured in South American cities, high concentrations of formaldehyde were reported in some countries, such as Brazil. In downtown Rio de Janeiro, mean formaldehyde concentrations rose 4-fold from 1998 to 2002, to 96 µg/m³ (with peak 2-hour concentrations as high as 138 µg/m³) (Corrêa & Arbilla, 2005). A further 10-fold increase in formaldehyde concentrations was reported in Rio de Janeiro from 2001 to 2004, as a consequence

Fig. 1.37 $PM_{2.5}$ concentrations (µg/m³) in selected African cities



$PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm.
Compiled by the Working Group with data from Boman et al. (2009), Tchuente et al. (2013), Abu-Allaban et al. (2007), Dionisio et al. (2010a, b), Arku et al. (2008), Kinney et al. (2011), van Vliet & Kinney (2007), WHO (2011), Petkova et al. (2013), Mkoma et al. (2010), Worobiec et al. (2011), and Kuvarega & Taru (2008).

of the introduction of compressed natural gas vehicles in 2000 (Martins et al., 2007).

(iii) The Middle East

WHO (2011) depicts air monitoring information for 39% of the Middle East countries. Air pollution monitoring coverage in the Middle East is similar to that in South American countries – 67% of both regions have some type of air quality data available; however, the air pollution monitoring sites are not evenly distributed across Middle East countries. Fig 1.40 and Fig. 1.41 depict PM concentrations across the Middle East; concentrations mostly ranged from 25 µg/m³ to 100 µg/m³ for PM_{10} and from 50 µg/m³ to 300 µg/m³ for $PM_{2.5}$.

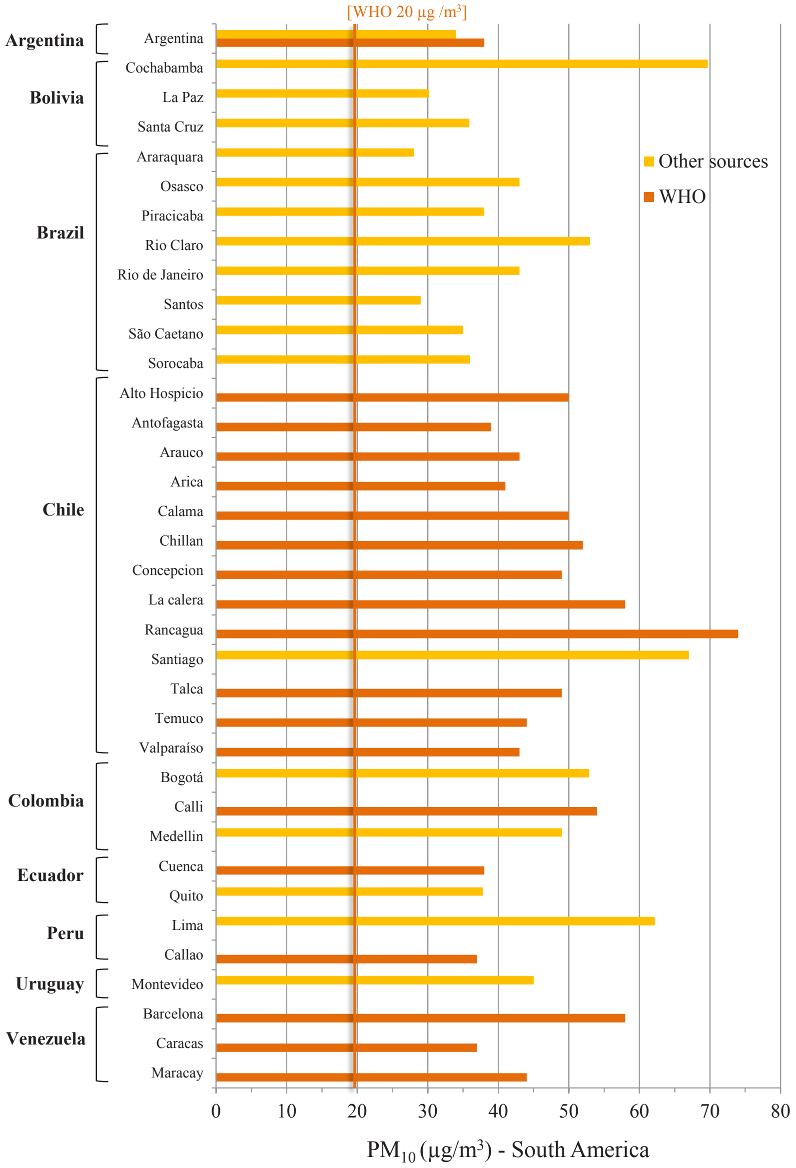
(iv) Australia

Between 1999 and 2008, there were significant decreases in the levels of air pollution in Australia. Levels of CO, NO_2, SO_2, and lead in urban areas declined to levels significantly below the national air quality standards. However, levels of PM and ozone did not decrease significantly over the time period. Between 1999 and 2008, the median 1-hour and 4-hour ozone levels varied between 0.02 ppm and 0.04 ppm in most Australian cities; the higher levels (~0.04 ppm) were observed in some areas including South East Queensland and Toowoomba. For PM, the median annual levels of PM_{10} remained at 15–20 µg/m³ and the $PM_{2.5}$ levels were 5–10 µg/m³ in most Australian cities in 2008 (Australian Government, 2010).

1.4.2 Exposure assessment in epidemiological studies

Epidemiological studies of relationships between air pollution exposure and cancer require long periods of observation and large populations. Therefore, it is virtually impossible with currently available approaches to assess exposure via personal monitoring (which is here distinguished from biomarkers of exposure, which are discussed in Section 1.4.3). Accordingly, epidemiological studies use outdoor air pollution concentrations as the primary basis for exposure

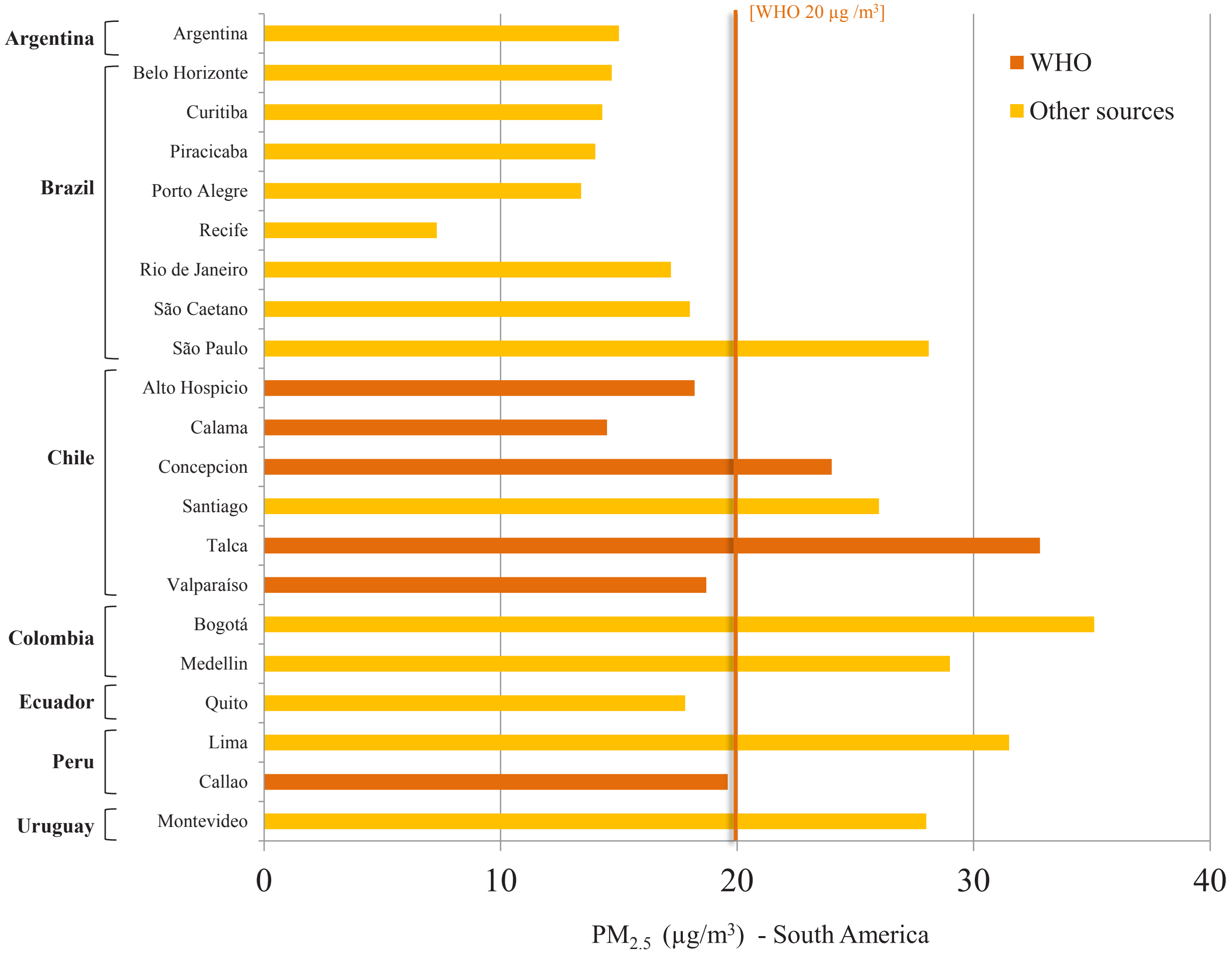
Fig. 1.38 PM_{10} concentrations (μg/m³) in selected South American cities



PM_{10}, particulate matter with particles of aerodynamic diameter < 10 μm.
Compiled by the Working Group with data from WHO (2011), Arkouli et al. (2010), Clean Air Institute (2012), CETESB (2013), FEAM (2011), IEMA (2007), de Miranda et al. (2012); INEA (2009), and RFF (2005).

estimation. Given that air quality monitoring is typically limited to measurements of a relatively small number of indicator pollutants collected at a limited number of discrete locations, epidemiological studies and risk assessments have typically used several approaches to estimate exposures of study subjects. Of particular importance for assessment of cancer is the ability to assess exposures over long time periods. An ideal assessment of long-term exposure requires both residential histories for the study population of interest and estimates of outdoor air pollution concentrations for periods of 20–30 years (life course). Prospective cohort studies following

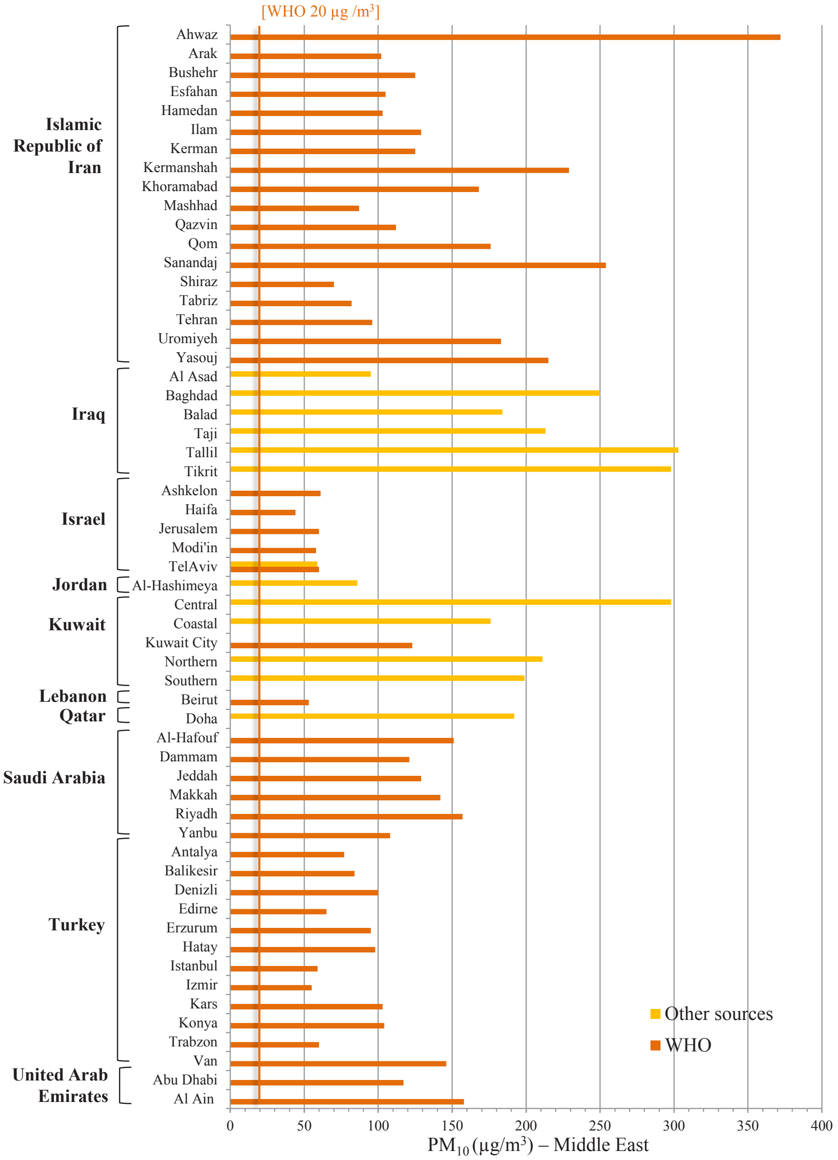
Fig. 1.39 $PM_{2.5}$ concentrations (µg/m³) in selected South America cities



$PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm.
Compiled by the Working Group with data from de Miranda et al. (2012), CETESB (2013), INEA (2009), WHO (2011), and Clean Air Institute (2012).

populations over long time periods with a focus on air pollution are rare; therefore, most studies require a retrospective exposure assessment approach. The ability to assign exposures retrospectively is often limited by the availability of historical exposure information or by the lack of residential histories. Several studies have evaluated the extent to which spatial patterns in measurements of NO_2 remain stable over time by repeating spatial measurement campaigns separated by periods of 7–18 years (Eeftens et al., 2011; Cesaroni et al., 2012; Gulliver et al., 2013; Wang et al., 2013). These studies suggest that although concentrations may change dramatically over time, the spatial patterns in concentrations remain quite similar. This suggests that studies of spatial contrasts in pollution based on information collected to represent one time period may be applied to other time periods using temporal trends, derived for example from a limited number of monitoring sites within the study area (Hystad et al., 2012). However, caution is needed in making extrapolations over longer periods of time, for more dynamic study areas, or for sites where major air pollution interventions took place.

Fig. 1.40 PM_{10} concentrations (µg/m³) in selected Middle East cities

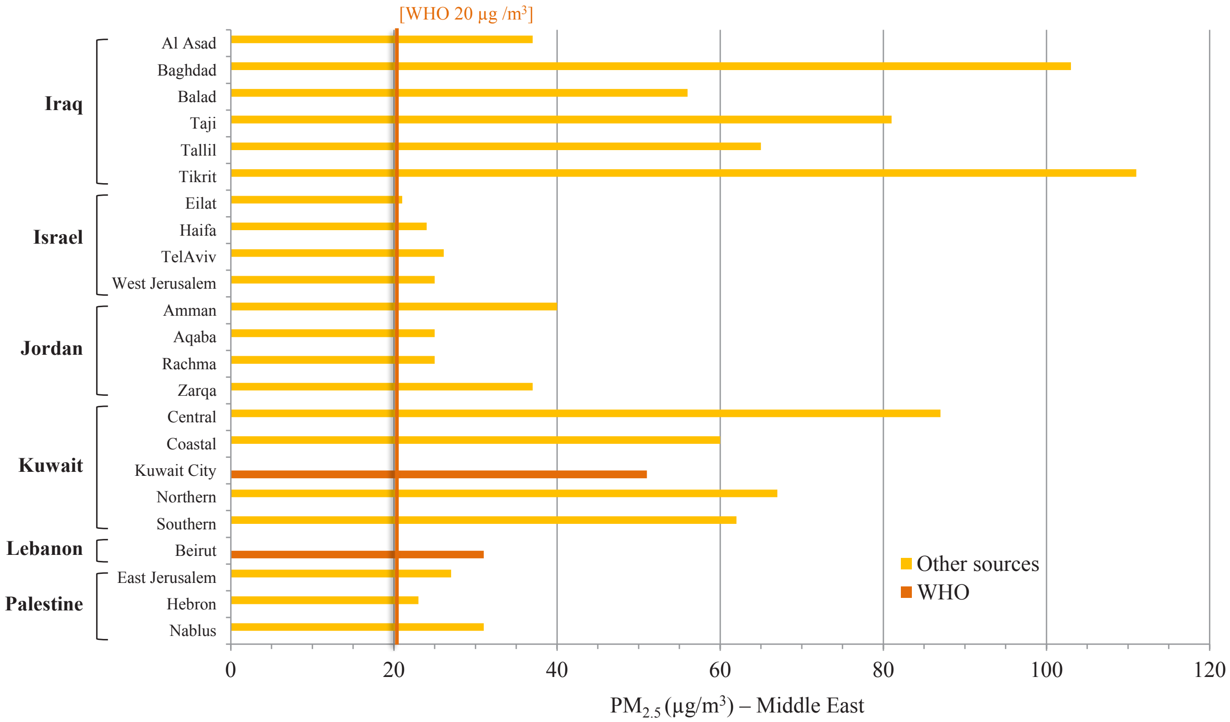


PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm.
Compiled by the Working Group with data from Vahlsing & Smith (2012), Khamdan et al. (2009), Naddafi et al. (2012), Brajer et al. (2012), WHO (2011), Mansourian et al. (2010), Engelbrecht et al. (2009), Agay-Shay et al. (2013), Israel Ministry of Environmental Protection (2010), Alnawaiseh et al. (2012), Saffarini & Odat (2008), Abu-Allaban et al. (2006), Al-Salem (2013), Alolayan et al. (2013), Saliba et al. (2010), Massoud et al. (2011), Qatar General Secretariat for Development Planning (2011), Al-Jeelani (2013), Rushdi et al. (2013), Khodeir et al. (2012), Munir et al. (2013), Meslmani (2004), Kara et al. (2013), Bayraktar et al. (2010), Kuzu et al. (2013), and Al Jallad et al. (2013).

(a) Outdoor air quality monitoring

The most traditional approach to estimate exposure is based on assignment of measured outdoor air pollutant concentrations to the study populations. Only rarely are these measurements specifically designed for the purposes of exposure assessment. One prominent exception is the Harvard Six Cities Study, in which air quality measurements in each study community were initiated at subject enrolment and continued in some form for much of the prospective follow-up

Fig. 1.41 $PM_{2.5}$ concentrations (µg/m³) in selected Middle East cities



$PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm.
Compiled by the Working Group with data from Khamdan et al. (2009), Brajer et al. (2012), Engelbrecht et al. (2009), von Schneidemesser et al. (2010), Sarnat et al. (2010), Agay-Shay et al. (2013), Alolayan et al. (2013), WHO (2011), Saliba et al. (2010), Massoud et al. (2011), Al-Jeelani (2013), Rushdi et al. (2013), Khodeir et al. (2012), Aburas et al. (2011), and Bayraktar et al. (2010).

period (Lepeule et al., 2012). In this case, the exposure assignment was based on a centrally located monitor in each community, and no adjustments were made for participants who changed addresses within each community as all subjects within a specific community were assigned the same exposure. A similar approach was applied in a Japanese cohort study where exposure was assigned based on address at study entry, and the analysis was restricted to those subjects who had resided in the study area for at least 10 years before enrolment and remained in the area during a 10-year follow-up period (Katanoda et al., 2011). In that study, the primary exposure metric of interest, $PM_{2.5}$, was estimated based on measured SPM levels using a subanalysis in which $PM_{2.5}$:SPM ratios were measured. Although this ratio was developed only for a specific time period and differed somewhat between study locations, a single ratio was applied to all areas. In the American Cancer Society's Cancer Prevention Study II (CPS-II) cohort (Turner et al., 2011), a single community-based monitor or the average of multiple monitors within each study community was used for exposure assignment. In that study, residential history was not considered because exposure assignment was based on the residential location at study entry. An identical method of assignment was used by Cao et al. (2011) in their assessment of air pollution and lung cancer in China. [Although these examples of exposure based on centrally located air quality monitors do not include within-city variation in concentrations, this approach to estimating exposure may be valid if the within-city variability in concentrations is less than the between-city variability, as might be the case for $PM_{2.5}$ but is less likely to be so for NO_2. Where individual exposures are imputed from central monitors, there will be resulting measurement error, which will have an impact on the bias and variance of subsequent effect size estimates. The importance of these errors will be greater if the inter-monitor variance is small relative to total inter-individual variance.]

Other approaches using community-based air quality monitors allow some level of individual-level exposure assignment based on within-area variability in pollutant concentrations, by assigning exposures based on the nearest monitor to the residential address of each study participant (Heinrich et al., 2013) or using geostatistical averaging such as inverse-distance weighting of measurements from available monitors within a defined study area (Lipsett et al., 2011). [All of these approaches do provide highly accurate descriptions of temporal variation at fine resolution and allow assessment of exposures during specific time windows.]

(b) Proximity measures

Although the above-mentioned approaches provide quantitative information on exposures to specific pollutants, they are limited in their ability to evaluate impacts of specific sources and are limited to areas with available outdoor pollution monitoring. In particular, many studies exclude subjects who reside beyond a specific distance from an available air monitoring site. Furthermore, there is increasing interest in evaluating differences within populations that may reside in the same community. One of the simplest approaches to estimating individual exposures is to measure proximity to specific pollutant sources, such as major roads (Heinrich et al., 2013) or industrial point sources (López-Cima et al., 2011). These examples estimate exposure by the distance (which may be described by linear or nonlinear functions) between a subject and a source. Source intensity measures, such as traffic counts over time or within a defined area, have also been used (Beelen et al., 2008; Raaschou-Nielsen et al., 2011). If subject residential histories are available, then such proximity estimates can be limited to specific time periods of interest or weighted over the full period of follow-up. As described in Section 1.4.1a, deterministic concentrations gradients based on proximity to major roads and industrial sources have been used to estimate exposure to several carcinogenic air pollutants in outdoor air in Canada (CAREX Canada, 2013; Setton et al., 2013). For each of these compounds, maps of estimated outdoor annual average concentrations allow exposure assignment at the individual level.

[Although proximity measures are simple to implement, often reflect gradients in measured concentrations, and allow studies of within-area exposure variation related to specific sources or source sectors, the relationship between proximity and levels of pollution will differ between studies conducted in different locations or at different times. This limits comparability of studies and does not allow quantification of adverse impacts in relation to pollutant concentrations. Furthermore, while the proximity measure is assumed to be a surrogate of exposure to air pollution, it may also reflect variation in other exposures (e.g. noise, in the case of traffic proximity) and in other potential determinants of health (e.g. socioeconomic status). Finally, proximity estimates generally have an overly simplistic representation of the physical processes related to pollutant fate and transport.]

(c) Atmospheric transport models

Given the understanding of a relatively high degree of variability in exposure within urban areas, often associated with motor vehicle traffic, several epidemiological studies have used dispersion models to estimate concentrations of specific air pollutants over space and time. In this approach, estimates of emissions and meteorological data are used (typically in a Gaussian dispersion model framework) to estimate the dispersion of pollutants within an airshed. Simple models do not consider any chemical transformation and are therefore most appropriate for non-reactive pollutants (e.g. CO); more sophisticated chemical transport models also incorporate a large number of chemical reactions and are designed to simulate atmospheric fate and transport, for example the production of secondary pollutants (Cesaroni et al., 2013). These models are designed for purposes other than health effects research, so their use in epidemiological studies has been opportunistic. In most cases, this approach has focused on estimating individual exposures to traffic-related pollutants within a single study area (Nyberg et al., 2000; Bellander et al., 2001; Gram et al., 2003; Nafstad et al., 2004; Naess et al., 2007; Raaschou-Nielsen et al., 2010, 2011), although there are examples of applications at the national level (Carey et al., 2013).

The Danish cohort studies (Raaschou-Nielsen et al., 2010, 2011) focus on traffic influences on NO_x and NO_2 combined with urban and regional background concentrations and have the notable advantages of both individual estimates of exposure and detailed residential histories, so that exposure estimates are a time-weighted average of outdoor concentrations at all addresses for each participant during the 34-year study period. The models include time-varying inputs on traffic levels and emissions and adjustments for street-canyon effects with time-varying information on building geometry. This approach also allows the estimation of exposure for different time windows, although estimates for the time of enrolment were strongly correlated ($r = 0.86$) with estimated exposures over the full period of follow-up (Raaschou-Nielsen et al., 2011).

The studies conducted in Oslo, Norway (Gram et al., 2003; Nafstad et al., 2003) incorporate emissions information for both traffic and point sources (industrial and space heating) to estimate individual-level exposures to SO_2 and NO_x for each year over a 25-year period. Deterministic gradients were used for subjects living in proximity to specific streets with the highest levels of traffic, and persons who moved to outside of Oslo were assigned a regional value for each year. Subjects moving from outside of Oslo were also assigned regional exposure values based on available outdoor monitoring network data. Subsequent analyses in Oslo have included estimates for $PM_{2.5}$ and PM_{10} (Naess et al., 2007) and incorporated emissions information from a larger set of source categories (traffic, road dust, wood burning) but were restricted to more recent and shorter time periods.

A very similar approach was used in a case–control analysis of lung cancer in Stockholm County, Sweden, in which individual exposures to SO_2, NO_2, and NO_x were estimated for each year over a 40-year period (Nyberg et al., 2000; Bellander et al., 2001). As in the Danish studies, the approaches applied in Oslo and Stockholm County allow individual exposure estimates covering different time windows.

The detailed data needed for dispersion modelling are seldom available at the national level. However, in a study in the United Kingdom, Carey et al. (2013) used emissions-based dispersion models to assign annual average concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2, and ozone for 1-km grid squares to the nearest postal code at the time of death. The model included emissions by source sector (e.g. power generation, domestic combustion, and road traffic), with pollutant concentrations estimated by summing pollutant-specific components, such as point and local area sources.

[Although dispersion models have a strong physical basis, even they are typically simplified representations of atmospheric transport that do not incorporate the complex physical and chemical transformations that occur after emission. Given their reliance on emissions, such models also are limited by the quality of emissions data as well as the lack of microscale meteorological measurements. Furthermore, dispersion models require specialized expertise to run, and there has been relatively little evaluation of dispersion models with measurements or integration of available measurements into the modelling effort. All of the above-mentioned examples are also limited to individual urban areas, given the data requirements of dispersion models, and therefore this approach is typically only applied to studies of within-city variation, which are usually focused on a single source sector, such as traffic. Although Carey et al. (2013) applied dispersion modelling at a national scale, their approach did not account for residential history or temporal changes in exposure and has a larger spatial resolution (1 km) than those of the Danish and Oslo models (~5 m).]

Although it has not been applied to epidemiological studies and is used as a screening-level assessment approach, the NATA (described in Section 1.4.1a) provides concentration estimates for several HAPs throughout the USA using a combination of dispersion, chemical transport, and exposure models (EPA, 2011b).

(d) Geospatial/land-use regression models

Land-use regression models or other geospatial statistical models have increasingly been used to assess chronic exposures to air pollution. In a simple form, estimates of source density and proximity can be used to estimate source-specific exposures. For example, Raaschou-Nielsen *et al.* used as supplementary exposure measures the presence of a street with a traffic density of more than 10 000 vehicles per day within 50 m of a residence and the total number of kilometres driven by vehicles within 200 m of the residence each day in a cohort analysis of cancer incidence for residents of two cities in Denmark (Raaschou-Nielsen et al., 2011). Chang *et al.* used the density of petrol stations as an indicator of a subject's potential exposure to benzene and other pollutants associated with evaporative losses of petrol or to air emissions from motor vehicles in a study of lung cancer in Taiwan, China (Chang et al., 2009). Although no evaluation of the exposure metric was conducted in this study, inverse distance to the nearest petrol station was associated with outdoor concentrations of benzene and xylene compounds in the RIOPA study in the USA (Kwon et al., 2006).

Land-use regression models are more sophisticated geospatial models in which pollutant measurements are combined with geographical predictors in a spatial regression model (Hoek et al., 2008a). This model is then used to predict concentrations of the air pollutant at unmeasured locations. These models have been especially useful in the assessment of exposure to variability in traffic-related air pollutant concentrations within urban areas. Note that the measurements used to develop models may be limited to available measurements from outdoor monitoring networks (Yorifuji et al., 2010, 2013), which are unlikely to fully capture the variability in outdoor concentrations or predictor variables, or from measurement campaigns of shorter duration (Cesaroni et al., 2013). Although land-use regression models often explain a high proportion (60–80%) of the variability in spatial measurements of air pollutant concentrations in a study area, if spatial correlation in model residuals exists, universal kriging may also be used for estimating exposures (Mercer et al., 2011). Universal kriging is a more generalized form of spatial modelling in which information from nearby (spatially correlated) measurements influences predictions through an estimated correlation structure.

In some cases these models may also incorporate temporal variation derived from outdoor monitoring network data, but most typically they provide estimates of spatial variability only, and it is assumed that this variability is stable over time – an assumption that has generally been supported by several measurement studies for periods of up to 18 years (Eeftens et al., 2011; Cesaroni et al., 2012; Gulliver et al., 2013; Wang et al., 2013). Models that do not rely on targeted measurement campaigns may also allow annual estimates to be made (Yorifuji et al., 2010, 2013).

Land-use regression estimates have also been combined with external monitoring data and proximity estimates in hybrid models. For example, Beelen et al. (2008) estimated exposure to outdoor air pollution at the home address at study entry as a function of regional, urban, and local components. The regional background concentrations were estimated using inverse-distance-weighted interpolation of measured concentrations at regional background outdoor monitoring sites. The urban component was estimated using land-use regression models developed using only regional and urban background monitoring site data, and the sum of the regional and urban contributions was defined as the background concentration. Background concentrations were estimated for NO_2, black smoke, and SO_2. Estimates were made for 5-year intervals during a 20-year study period. The local traffic contribution was based on several measures of traffic intensity and proximity. In addition, quantitative estimates for the local component were estimated with regression models incorporating field monitoring measurements and traffic variables. The local component was added to background concentrations for an overall exposure estimate for each pollutant. [Land-use regression models are relatively easy to implement and, given their use of pollutant measurements, are capable of providing reliable estimates of exposure to a large number of specific pollutants as well as source indicators (Jerrett et al., 2005). Confidence in model use depends on adequate geographical and pollutant monitoring data, especially the inclusion of targeted monitoring that characterizes variability both in air pollutant concentrations and in geographical predictors within the study area. Reliability can be quite high, especially with increasing numbers of observation locations.]

(e) Remote sensing

A more recent development for application to epidemiological studies has been the use of remote-sensing-based estimates of air pollution. For example, van Donkelaar et al. (2010) developed a global model of long-term average $PM_{2.5}$ concentration at a spatial resolution of about 10 × 10 km. This approach combines aerosol optical depth (AOD) (a measure of the scattered light from all aerosol within the total column between the Earth’s surface and the satellite) with information from a chemical transport model on the vertical stratification of aerosol as well as its composition to estimate time- and location-specific factors to relate AOD to surface $PM_{2.5}$. Estimates derived from this approach were combined with surface monitoring data and estimates from a different chemical transport model to estimate exposures for the Global Burden of Disease Study 2010 (Brauer et al., 2012; Lim et al., 2012). Useful satellite retrievals have been available since about 2000 and have been combined with available surface monitoring data to provide backcasted spatially resolved estimates for earlier periods (Crouse et al., 2012; Hystad et al., 2013), as described in more detail below. Satellite-based estimates are available globally for a small group of pollutants, including $PM_{2.5}$, NO_2, ozone, and formaldehyde (Brauer et al., 2012; De Smedt et al., 2012; Lamsal et al., 2013).

[Remote-sensing-based estimates have the advantage of providing estimates of concentrations essentially anywhere in the world by a consistent approach, although they are best suited to between-location contrasts, given the currently available resolution on the order of 10 × 10 km.]

(f) Remote sensing and land-use regression hybrid models

Remote-sensing-based estimates have also been combined with land use and other geographical predictors in hybrid land-use regression-type models. Canadian researchers developed national estimates of long-term average concentrations of $PM_{2.5}$ and NO_2 in which satellite-based estimates were combined with deterministic gradients related to traffic and industrial point sources (Hystad et al., 2011). Although these models were only spatial and did not include a temporal component, in a subsequent effort (Hystad et al., 2012), which was applied to a cohort analysis of lung cancer with detailed residential histories (Hystad et al., 2013), spatial satellite-based estimates for $PM_{2.5}$ and NO_2 and chemical transport model estimates for ozone were adjusted retrospectively with annual air pollution monitoring data, using either spatiotemporal interpolation or linear regression to produce annual estimates for a 21-year period. In addition, proximity to major roads, incorporating a temporal weighting factor based on mobile-source emission trends, was used to estimate exposure to vehicle emissions, and industrial point source location proximity was used to estimate exposures to industrial emissions. In the USA, Novotny et al. (2011) developed a national spatiotemporal land-use regression model with 30 m spatial resolution and 1 hour temporal resolution based on a single year of available regulatory monitoring network data, satellite-based estimates, and geographical predictors (population density, land use based on satellite data, and distance to major and minor roads). To date, this model has not been applied in epidemiological analyses.

More recently, a novel spatiotemporal approach combining AOD and daily calibration to available monitoring network measurements with land-use data (Kloog et al., 2011) was applied to investigate the effect of long-term exposures to $PM_{2.5}$ on population mortality (Kloog et al., 2013).

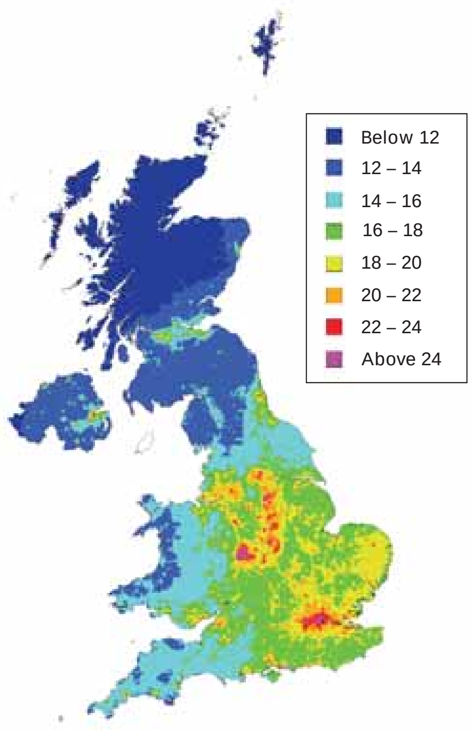
(g) Bioindicators (lichens/pine needles)

Although there are only limited examples of applications to epidemiological analyses, several approaches using environmental biomonitors such as lichens and pine needles (Augusto et al., 2010) as indicators of air pollution levels have been developed. For example, lichen biodiversity in north-eastern Italy was geographically correlated with both measurements of SO_2 and NO_3 and male lung cancer mortality, after correcting for spatial autocorrelation (Cislaghi & Nimis, 1997). In risk assessment, measures of PAHs and heavy metals in lichens have been used to estimate exposures (Augusto et al., 2012; Käffer et al., 2012) and cancer risk. Augusto *et al*. used measurements of multiple PAH species in lichens to develop a spatial model related to industrial point-source emissions of PAHs (Augusto et al., 2009). These approaches may prove to be useful in estimating historical exposures as the biomonitors can integrate deposited pollutant species over relatively long time periods.

1.4.3 Personal exposure and biomarkers

In recent decades a large number of studies have been published on personal exposure to major air pollutants (Wallace, 2000; Monn, 2001). Research conducted since the early 1980s has indicated that personal exposure may deviate significantly from concentrations measured at fixed sites in the outdoor environment. Subsequent research has identified factors that are responsible for differences between outdoor and personal exposure. In this section, the factors affecting personal exposure are summarized, followed by a discussion of validity studies in which indicators of exposure have been compared with actual measurements of personal exposure. There is also a brief discussion of the distinction between pollutants of outdoor origin and pollutants from indoor sources (Wilson et al., 2000; Ebelt et al., 2005; Wilson & Brauer, 2006).

Fig. 1.42 Estimated United Kingdom annual average background PM_{10} concentrations (μg/m³) during 2002



PM_{10}, particulate matter with particles of aerodynamic diameter < 10 μm.
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Personal monitoring studies have been conducted for most of the major air pollutants, including PM, NO_2, VOCs, and ozone (Monn, 2001). Studies measuring PM have often used integrated samplers sampling $PM_{2.5}$ or PM_{10}, but real-time instruments based on light scattering have been used as well. Recently, studies have also measured personal exposure to ultrafine particles (Wallace & Ott, 2011; Buonanno et al., 2014), focusing especially on commuters' exposures (Knibbs et al., 2011). Fewer studies have measured particle composition. Components that have been measured include EC or proxies of EC, aerosol acidity, PAHs, and elemental composition.

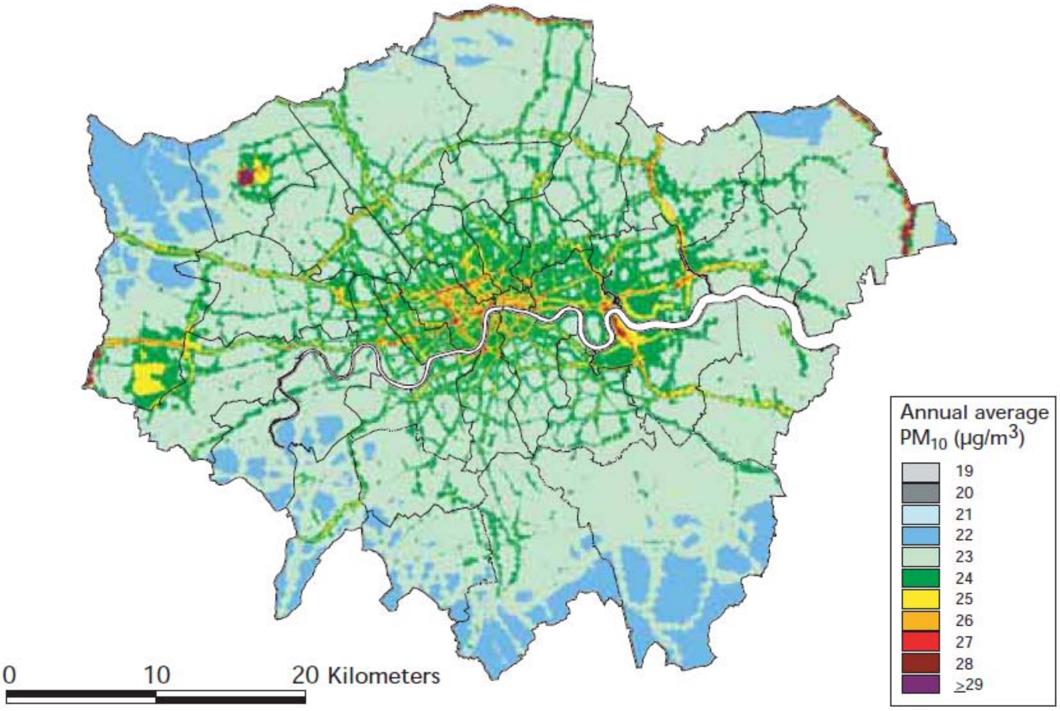
(a) Factors affecting personal exposure

For cancer, long-term average personal exposure is the biologically relevant exposure. Therefore, it is important to assess both the intensity of exposure and the duration. Exposure assessment in epidemiological studies of cancer and air pollution is often based on the residential address. Hence, residential history should be considered. A large number of studies have identified factors that affect the intensity of personal exposure to major air pollutants. These factors can be grouped into four broad groups: (i) concentration in outdoor air, at the residence and in the community; (ii) time–activity patterns, including residential history; (iii) infiltration of pollutants indoors; and (iv) indoor sources of pollutants. These factors are discussed further in the sections below, with a focus on air pollution including particles of outdoor origin.

(i) Concentration in outdoor air

People may be exposed to outdoor air pollutants directly while spending time outdoors. However, a significant fraction of the exposure to outdoor air pollutants occurs while spending time indoors, as people generally spend a large fraction of their time indoors and pollutants penetrate into the indoor environment. Because people spend a significant fraction of their time in or near their own home, exposure in epidemiological studies is often characterized based on the residential address. Residential address information is generally available from ongoing epidemiological studies designed for purposes other than studying air pollution effects. Most often the outdoor concentration at the address is characterized. A large number of studies have evaluated spatial variation of outdoor air pollution (Monn, 2001; HEI, 2010b). Spatial variation can be present at various scales, ranging from global to local (HEI, 2010b). Examples of the various scales of variation are illustrated in Fig. 1.42 and Fig. 1.43, and in Fig. 1.3 in Section 1.4.1a.

Fig. 1.43 Annual average PM_{10} concentrations (µg/m³) in London calculated for 2004



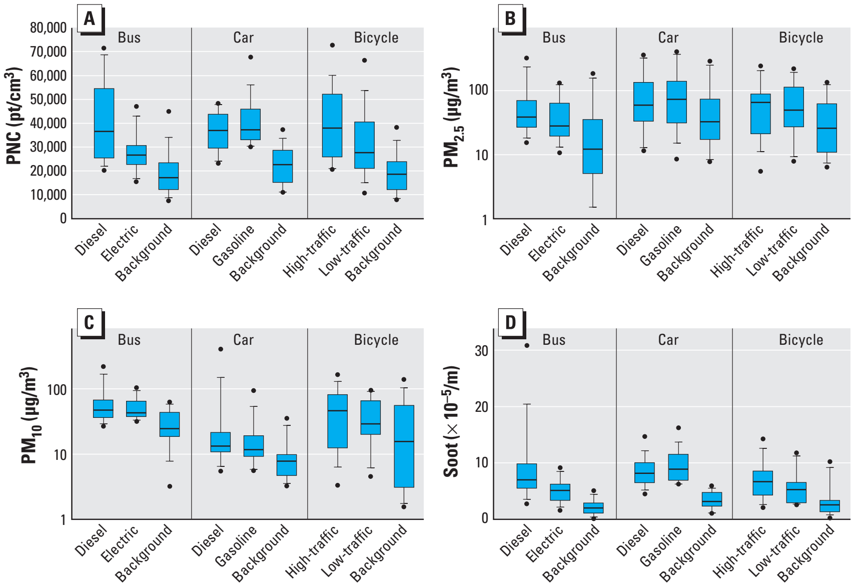
PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm.
Reprinted from Air Quality Expert Group (2005). © Crown copyright 2005.

Contrasts across countries within a continent are discussed further in Section 1.4.1.

As Fig. 1.43 illustrates, within urban areas, significant spatial variation is present related to proximity to major roads. Large gradients with distance to major roads have been identified for traffic-related pollutants, including NO_2, CO, benzene and other VOCs, EC, and ultrafine particles (HEI, 2010b). Gradients are relatively small for $PM_{2.5}$ and PM_{10} compared with, for example, EC (HEI, 2010b; Janssen et al., 2011). A summary of studies measuring both $PM_{2.5}$ or PM_{10} and BC reported an average ratio of 2 for BC and 1.2 for PM concentrations at street sites compared with urban background levels (Janssen et al., 2011). Spatial gradients vary significantly by pollutant and are nonlinear near major roads, with steep decreases in the first 50–100 m and smaller decreases up to about 300–500 m (HEI, 2010b). In compact urban areas, gradients from major roads are much smaller.

A growing number of studies have documented significant exposures to a range of traffic-related air pollutants, including fine and ultrafine particles, EC, and VOCs, while in transit, including walking, cycling, car and

Fig. 1.44 Concentrations in modes of transportation and at the urban background location on corresponding sampling days



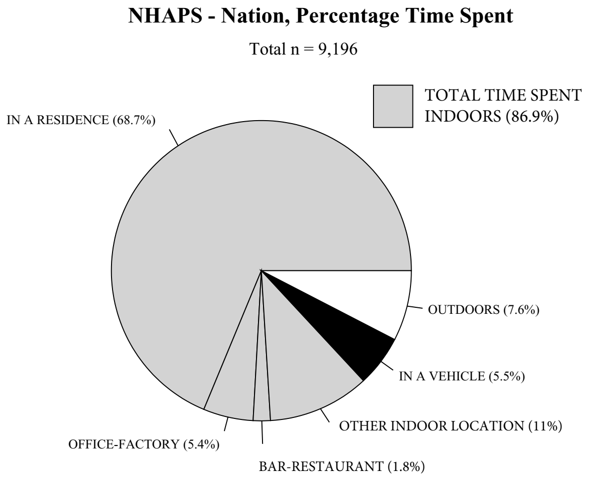
PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; PNC, particle number concentration.
Reproduced from *Environmental Health Perspectives* (Zuurbier et al., 2010).

bus driving, and underground (Fig. 1.44; Kaur et al., 2007; de Hartog et al., 2010; Zuurbier et al., 2010; de Nazelle et al., 2011). Commuters' exposures further differ significantly with route, and despite the relatively short time typically spent in traffic, significant contributions to average personal exposure may occur (Marshall et al., 2006; Kaur et al., 2007; Van Roosbroeck et al., 2008; de Nazelle et al., 2013; Dons et al., 2012, 2013). Van Roosbroeck et al. (2008) found that time spent in traffic was a significant predictor of 48-hour personal exposure to soot and $PM_{2.5}$ in elderly adults in the Netherlands. A study in 62 volunteers in Belgium reported that 6% of time was spent in traffic, but the contribution to the measured 24-hour average personal exposure to BC was 21%, and to calculated inhaled doses was 30% (Dons et al., 2012). Home-based activities, including sleep, accounted for 65% of time, 52% of exposure, and 36% of inhaled dose (Dons et al., 2012). For volunteers in Barcelona, Spain, in-transit exposures accounted for 6% of time, 11% of NO_2 exposure, and 24% of inhaled dose (de Nazelle et al., 2013). Setton and co-workers documented that ignoring residential mobility in epidemiological studies using individual-level air pollution may (modestly) bias exposure response functions towards the null (Setton et al., 2011).

(ii) Time–activity patterns

A range of surveys in developed countries have shown that most people spend a large fraction of their time indoors. An example is shown from the large National Human Activity Pattern Survey (NHAPS) in the USA (Fig. 1.45; Klepeis et al., 2001). On average, subjects spent 87% of their time indoors, of which a large fraction was spent in their own residence. Time spent outdoors accounted for about 2 hours of the day. These broad patterns have been found in other surveys as well (Jenkins et al., 1992; Leech et al., 2002).

Fig. 1.45 Time spent in various microenvironments by subjects in the USA



Reprinted from Klepeis et al. (2001) by permission from Macmillan Publishers Ltd: *Journal of Exposure Science and Environmental Epidemiology*, Klepeis NE, Nelson WC, Ott WR, Robinson JP, Tsang AM, Switzer P et al. The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants, Volume 11, Issue 3, pages 231–252, copyright (2001).

However, individual time–activity patterns differ substantially, related to factors such as age, employment, and socioeconomic status. A recent survey showed that German children spent on average 15.5 hours per day in their own home (65% of time), 4.75 hours in other indoor locations (for a total of 84% of time spent indoors), and 3.75 hours outdoors (16% of time) (Conrad et al., 2013). The German survey did not distinguish between "outdoors" and "in traffic," which may be partly responsible for the share of time spent outdoors.

Time–activity patterns vary significantly over the day, as does the air pollution concentration, supporting the use of more dynamic exposure estimates (Beckx et al., 2009). Time–activity patterns may thus differ across population groups (related to age, sex, employment status, socioeconomic position, and other factors), contributing to contrasts in exposure between population groups beyond contrasts in outdoor concentrations. A study in Delhi, India, showed a high proportion of time spent indoors, with significant variability across population groups (Saksena et al., 2007).

The contribution to time-weighted average exposure is a function of the time spent in a microenvironment and the concentration in that microenvironment. Thus, for pollutants that infiltrate poorly indoors, the relatively short time spent outdoors, including in transit, may nevertheless amount to a significant fraction of total exposure.

Because of the large fraction of time spent in the home, residential history is an important determinant of long-term average exposure to air pollution. In epidemiological studies, air pollution exposure is often assigned based on the most recent address or the address at recruitment into the (cohort) study. Because a significant number of subjects may change address before inclusion in the study or during follow-up, misclassification of exposure may occur. This is particularly problematic because limited information is available about the critical window of exposure. In a study in California of children with leukaemia, residential mobility differed with age and socioeconomic status, and accounting for residential mobility significantly affected the assigned neighbourhood socioeconomic status and urban/rural status (Urayama et al., 2009). A case–control study in Canada reported that in the 20-year exposure period, 40% of the population lived at the same address (Hystad et al., 2012). The correlation between air pollution exposure estimates with and without residential history was 0.70, 0.76, and 0.72 for $PM_{2.5}$, NO_2, and ozone, respectively. About 50% of individuals were classified into a different $PM_{2.5}$, NO_2, and ozone exposure quintile when using study-entry postal codes and spatial pollution surfaces, compared with exposures derived from residential histories and spatiotemporal air pollution models (Hystad et al., 2012). Recall bias was reported for self-reported residential history, with lung cancer cases reporting more residential addresses than controls (Hystad et al., 2012). In a Danish cohort study, exposure was characterized as the average concentration of all addresses 20–25 years before enrolment and during follow-up weighted with the time lived at an address (Raaschou-Nielsen et al., 2011). People moved on average 2.4 times before enrolment and 0.3 times during follow-up. Exposure estimates from different periods were highly correlated (Section 1.4.2).

(iii) Infiltration of pollutants indoors

Because people generally spend a large fraction of their time indoors and outdoor air pollution infiltrates indoors, this section examines relationships between outdoor and indoor pollutant levels.

Mass-balance models have been used extensively to describe the concentration in indoor air as a function of outdoor air and indoor sources. The indoor concentration of an air pollutant can be expressed simply as $C_{ai} = F_{inf}\, C_a$, where C_{ai} = is the indoor pollutant concentration originating from outdoors, F_{inf} is defined as the infiltration factor, and C_a is the ambient (outdoor) concentration. The infiltration factor describes the fraction of outdoor pollution that penetrates indoors and remains suspended. Penetration efficiency depends on several factors, including the air velocity, the dimensions of the opening, and the particle size, with ultrafine and especially coarse particles penetrating less efficiently (Liu & Nazaroff, 2001).

Hänninen et al. (2011) evaluated the original data of European studies of indoor–outdoor relationships for $PM_{2.5}$. The overall average infiltration factor was 0.55, illustrating significant infiltration of outdoor fine particles. A review including European and North American studies reported infiltration factors of 0.3–0.82 for $PM_{2.5}$ (Chen & Zhao, 2011). Since people in Europe and North America spend a large fraction of their time indoors, human exposure to fine particles of outdoor origin occurs mostly indoors. Infiltration factors were consistently higher in the summer than in the winter (Hänninen et al., 2011). A study in seven cities in the USA included in the MESA Air study also reported high infiltration factors in the warm season (Allen et al., 2012). The implication is that for the same outdoor concentration, the actual human exposure is higher in the summer than in the winter. Higher infiltration factors in the summer are explained by higher air exchange rates in the summer than in the winter.

In the four European cities included in the RUPIOH study, infiltration factors for ultrafine particles assessed by total particle number counts were somewhat lower than those for $PM_{2.5}$ (Table 1.13; Hoek et al., 2008b) but higher than those for coarse particles. A large study in Windsor, Ontario, Canada, that measured total particle number counts reported infiltration factors of 0.16–0.26, with a large variability for individual homes (Kearney et al., 2011). The lower infiltration of ultrafine particles is consistent with lower penetration and higher decay rates due to diffusion losses compared with accumulation mode particles. Studies in the USA that measured particle size distributions have also found lower infiltration factors, on the order of 0.5 for particles in the ultrafine range and up to 0.7 for $PM_{2.5}$ (Abt et al., 2000; Long et al., 2001; Sarnat et al., 2006). A study conducted in a Helsinki, Finland, office found that indoor particle number concentrations tracked outdoor concentrations well but were only 10% of the outdoor concentrations (Koponen et al., 2001). A study in two empty hospital rooms in Erfurt, Germany, reported a high correlation between indoor and outdoor concentrations of $PM_{2.5}$, black smoke, and particle number concentration and an indoor–outdoor ratio of 0.42 for total number concentration, compared with 0.79 for $PM_{2.5}$ (Cyrys et al., 2004). There is thus a large range in reported infiltration factors, related to differences in air exchange rates and building characteristics, and likely also to differences in measurement methods across studies.

Table 1.13 Infiltration factors estimated as regression slope for the relationships between indoor and outdoor 24-hour average concentrations of different particle metrics from the RUPIOH study

Pollutant	Helsinki, Finland	Athens, Greece	Amsterdam, Netherlands	Birmingham, United Kingdom
$PM_{2.5}$	0.48	0.42	0.39	0.34
$PM_{10} - PM_{2.5}$	0.14	0.16	0.11	0.13
PNC	0.42	0.42	0.19	0.22
Soot	0.63	0.84	0.78	0.71
Sulfate	0.59	0.61	0.78	0.61

PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; PNC, particle number concentration.
Data from Hoek et al. (2008b).

The composition of particles infiltrated indoors also differs from the outdoor composition. Infiltration factors for EC exceeded those for $PM_{2.5}$ significantly (Fig. 1.46). EC is concentrated in submicrometre particles, is non-volatile, and has few indoor sources (Noullett et al., 2010). Although smoking affects EC levels, the impact is less than on $PM_{2.5}$ concentrations (Götschi et al., 2002). A detailed analysis of the RIOPA study showed that 92% of the indoor EC concentration was due to outdoor EC, whereas the corresponding contribution for $PM_{2.5}$ was 53% (Meng et al., 2009).

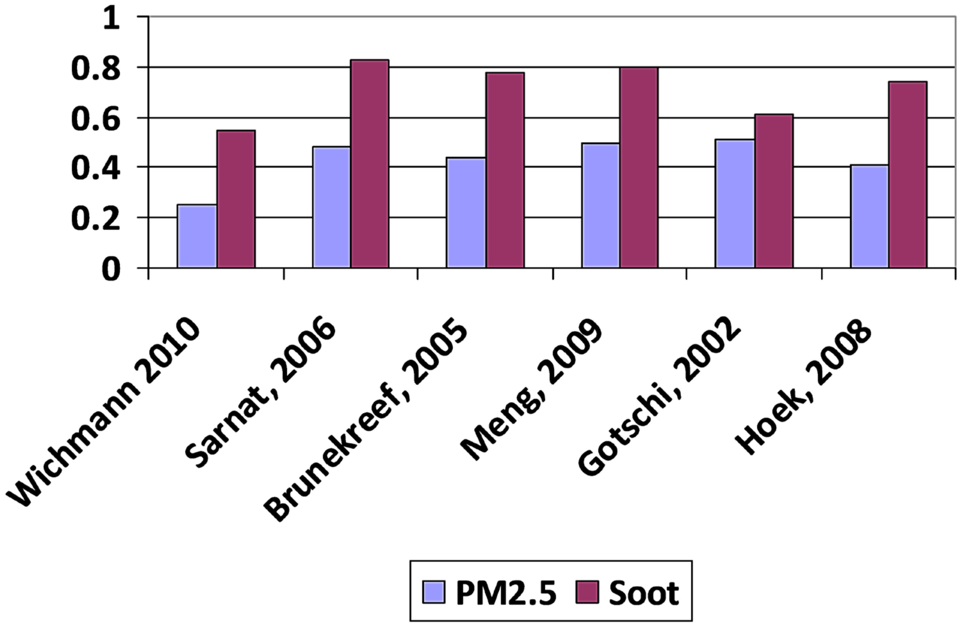
Sulfates have few indoor sources and high infiltration factors (Noullett et al., 2010). Indoor concentrations of SO_2 in the absence of indoor sources (e.g. unvented kerosene heaters) are typically low, related to large losses to indoor surfaces (Koutrakis et al., 2005).

Nitrates typically show low infiltration factors, ranging from 0.05 to 0.2 in studies in Europe and the USA (Sarnat et al., 2006; Hoek et al., 2008b). Indoor concentrations of NO_2 in the absence of indoor sources (e.g. gas cooking, unvented heaters) are substantially lower than outdoor concentrations (Monn, 2001). In a recent review of studies of personal and outdoor NO_2 exposure, the overall average personal–outdoor regression slope was between 0.14 and 0.40, depending on the study type (Meng et al., 2012a). A study in Spain reported indoor–outdoor slopes of 0.20 and 0.45 for two cities, after adjusting for the large influence of gas cookers and gas heaters (Valero et al., 2009). Personal exposure may be affected by more factors, but studies have shown that the indoor concentration is the dominant factor, with large heterogeneity observed between studies (Monn, 2001; Meng et al., 2012a).

Indoor ozone concentrations are typically low because ozone is a highly reactive component with a high decay rate and no indoor sources in residences (Monn, 2001). Indoor–outdoor ratios of between 0.2 and 0.8 were reported in previous studies, depending on air exchange rates (Monn, 2001). A recent analysis of the DEARS study in Detroit, USA, reported a personal–outdoor regression slope of 0.03 in summer and 0.002 in winter (Meng et al., 2012b), even lower than that for NO_2 and much lower than that for $PM_{2.5}$.

In large-scale epidemiological studies, indoor measurements of infiltration factors are not feasible. Hystad and co-workers developed a model for $PM_{2.5}$ infiltration based on measurements in 84 North American homes and publicly available predictor variables, including meteorology and housing stock characteristics (Hystad et al., 2009). A model including season, temperature, low building value, and heating with forced air predicted 54% of the variability in measured infiltration factors (Hystad et al., 2009). Low building value increased infiltration factors, increasing exposure contrasts across different

Fig. 1.46 Infiltration factors for $PM_{2.5}$ and soot (EC, BC) measured in the same study



BC, black carbon; EC, elemental carbon; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm.
Compiled by the Working Group with data from Wichmann et al. (2010), Sarnat et al. (2006), Brunekreef et al. (2005), Meng et al. (2009), Götschi et al. (2002), and Hoek et al. (2008b).

socioeconomic groups. Other modelling studies in North America reported similar results, with a substantial fraction of the variability of infiltration factors explained by factors including window opening, air exchange rate, and presence or use of central air conditioning and forced air heating, with indications that predictors differ by season (Clark et al., 2010; Allen et al., 2012).

(iv) Indoor sources of pollutants

Numerous indoor sources, including tobacco smoking, cooking, heating, appliances, consumer products, building construction, and activities such as vacuum cleaning, have been identified to affect indoor concentrations and personal exposure for a wide range of air pollutants (Weschler, 2009).

Indoor sources affect different pollutants to a different degree. As noted above, sulfate and EC are affected more by outdoor air pollution than by indoor sources. Sulfate has therefore been used to evaluate the personal or indoor exposure to particles of outdoor origin (Sarnat et al., 2002).

(b) Pollutants from both indoor and outdoor sources

Several authors have stressed the importance of distinguishing between personal exposure from all sources and exposure indoors to pollutants from indoor and outdoor sources (Wilson et al., 2000; Ebelt et al., 2005; Wilson & Brauer, 2006). The discussion was initiated in the framework of temporal studies of $PM_{2.5}$ showing often modest correlations between total personal $PM_{2.5}$ and outdoor $PM_{2.5}$ concentrations. Scientific reasons to separate the two sources include that particle composition differs significantly depending on the source and that different particle composition might influence health effects. Furthermore, if the interest is in evaluating the health effects of outdoor pollution, then exposure to the same pollutant from indoor sources should be treated as a potential confounder. The implication is that to assess agreement between often used exposure metrics and personal exposure, personal exposure to pollutants of outdoor origin should be evaluated. It may also be important to distinguish pollution exposures originating from outdoor versus indoor sources for policy purposes.

(c) Validation studies

In this context, validation studies are studies that compare exposure metrics used in epidemiological studies (e.g. modelled outdoor concentration) with personal exposure monitoring, which is usually considered as a more valid method of individual exposure assessment. A critical issue is that the correct comparison must be made between exposure metrics and personal exposure, depending on the epidemiological study design and the health outcome of interest. For time-series studies of acute events, the interest is in the longitudinal (within-subject) variation in exposure levels, whereas for cohort studies assessing long-term exposures, the interest is in the between-subject variation of long-term averages.

Very few studies have assessed the validity of long-term outdoor exposure estimates as used in epidemiological studies for estimating long-term average personal exposure, in contrast to the large literature on the temporal correlation of outdoor and personal exposure over shorter time intervals (Avery et al., 2010). It is challenging to collect sufficient personal exposure data to represent a long-term average exposure in a large group of subjects. Consequently, most of the personal monitoring studies discussed previously rely on a single or a few 24-hour measurements. First, studies evaluating fine-spatial-scale outdoor exposure metrics are discussed. Next, studies assessing differences in personal exposure between cities are discussed.

A study in Amsterdam reported significantly higher outdoor concentrations of $PM_{2.5}$, soot, PAHs, and benzene measured near high-traffic homes compared with low-traffic homes (Fischer et al., 2000). These contrasts were also found for indoor concentrations; for example, for soot, concentration ratios of 1.8 for high- versus low-traffic homes were found for both indoor and outdoor measurements (Fischer et al., 2000). Another study in Amsterdam reported ratios of soot concentrations for high- versus low-traffic homes of 1.19 to 1.26 for 24-hour measurements indoors and of 1.29 for personal exposure (Wichmann et al., 2005). A study in Utrecht comparing air pollution exposures of elderly adults living near major roads versus minor roads found larger differences for soot than for $PM_{2.5}$ and NO_2 using both personal and environmental measurements (Van Roosbroeck et al., 2008).

A study among volunteers in Helsinki, Barcelona, and Utrecht found a significant correlation between long-term average residential outdoor soot concentrations estimated by city-specific land-use regression models and measured average personal exposure (Montagne et al., 2013). Within the individual cities, no consistent association was found between land-use regression-modelled NO_2 and $PM_{2.5}$ concentrations and personal exposures, but modelled and measured exposures to all pollutants were highly correlated when all data from all three cities were combined. The finding of strong correlations between modelled and measured exposures in the combined data from the three cities may be relevant for studies exploiting exposure contrasts across cities.

Two Dutch studies in children reported significant correlation between NO_2 exposure measured at school and personal exposure, which remained after accounting for indoor sources including gas cooking (Rijnders et al., 2001; van Roosbroeck et al., 2007). In contrast, a Canadian study where the 72-hour personal NO_2 exposure of elderly adults was measured in three seasons found no relationship between the modelled long-term average outdoor concentration and the personal exposure measurements (Sahsuvaroglu et al., 2009).

Two studies reported consistently higher population average personal exposures in European cities with higher outdoor concentrations (Monn et al., 1998; Georgoulis et al., 2002). Personal NO_2 exposure was highly correlated with outdoor concentration in a study in eight Swiss cities and towns with large contrasts in outdoor NO_2 concentration (Monn et al., 1998). The correlation between community average outdoor concentration and personal exposure was $R^2 = 0.965$ (Fig. 1.47; Monn, 2001).

(d) Social inequalities in air pollution exposure

There is a large literature that has evaluated contrasts in air pollution exposures in association with socioeconomic status (O'Neill et al., 2003). In general, higher outdoor air pollution concentrations have been observed for subjects with lower socioeconomic status, related to residential location (O'Neill et al., 2003). However, the contrast in air pollution exposures across socioeconomic groups differs significantly between study areas and spatial scales, with several studies showing higher concentrations for individuals with higher socioeconomic status (Deguen & Zmirou-Navier, 2010). A study in Rome, Italy, reported that subjects living close to major roads had a higher socioeconomic position than subjects living further away from major roads (Cesaroni et al., 2010).

Most studies of air pollution and socioeconomic status have evaluated outdoor pollutant concentrations with little attention to time–activity patterns and indoor exposures. Higher indoor concentrations were reported in low-income subjects, related to outdoor concentrations, indoor sources, and housing characteristics (Adamkiewicz et al., 2011). A study in Vancouver, Canada, reported higher wood smoke exposures and intake fractions in low-income neighbourhoods (Ries et al., 2009).

(e) Biomarkers of exposures

Biomarkers of exposure to outdoor air pollution have not been commonly used as the main method of exposure assessment in large-scale epidemiological studies of outdoor air pollution and cancer. However, associations between biomarkers of exposure and biomarkers of effect have been evaluated in smaller studies with tens to hundreds of subjects (see Section 4). In this context, biomarkers can contribute to elucidating the pathway from exposure to cancer. Biomarkers could additionally be useful in retrospective exposure assessment, if appropriate biological material has been stored; however, a limitation of many biomarkers for this purpose is their relatively short half-life (Scheepers, 2008).

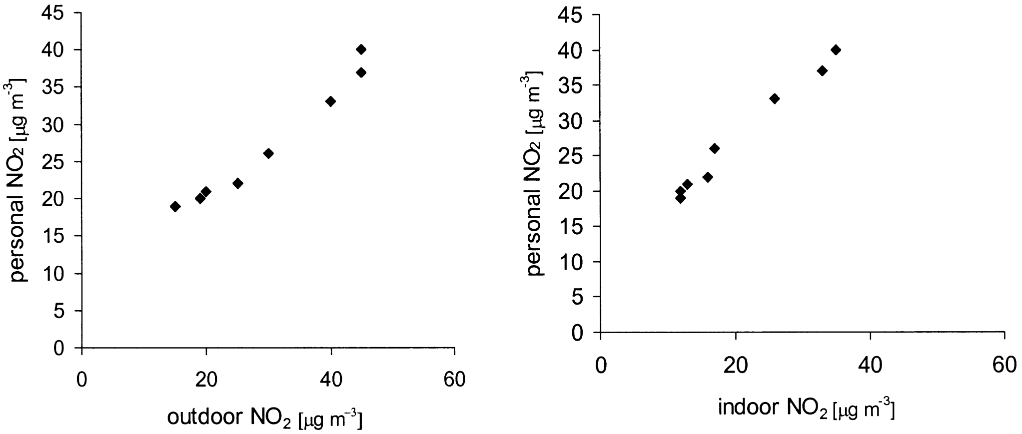
Associations of biomarkers with exposure to air pollution have been described in several recent reviews (Barbato et al., 2010; Møller & Loft, 2010; Demetriou et al., 2012; DeMarini, 2013; Rylance et al., 2013). Demetriou et al. (2012) specifically considered the utility of potential biomarkers of exposure to air pollution in a systematic review. The evidence of an association with external exposure was considered to be strong for 1-hydroxypyrene (1-OHP), DNA adducts, and oxidized nucleobases, particularly 8-oxo-7,8-dihydro-2′-deoxyguanosine (8-oxodG). Studies in a wide variety of populations, including children, mail carriers, traffic police, and professional drivers, have repeatedly found increases in 1-OHP and in the frequency of DNA adducts in more exposed subjects (Demetriou et al., 2012).

It should be noted that the same markers can often be interpreted as indicators of early biological effects, as well as of exposure (DeMarini, 2013). Studies using these biomarkers and other markers of effect are reviewed in detail in Section 4.

1.4.4 Occupational exposure of outdoor workers

See Table 1.14.

Fig. 1.47 Scatterplot for outdoor–personal ($R^2 = 0.965$) and indoor–personal ($R^2 = 0.983$) NO_2 ratios of aggregated data (annual mean estimates) in eight Swiss cities



N (indoor) = 1501, *N* (outdoor) = 1544, *N* (personal data) = 1494; NO_2, nitrogen dioxide.
Reprinted from Monn (2001). *Atmospheric Environment*, Volume 35, Monn C, Exposure assessment of air pollutants: a review on spatial heterogeneity and indoor/outdoor/personal exposure to suspended particulate matter, nitrogen dioxide and ozone, Pages 1–32, Copyright (2001), with permission from Elsevier.

Workers who spend significant amounts of time outdoors may be occupationally exposed to outdoor air pollution. Although workers such as farmers, miners, and construction workers can face exposure to polluted air, emissions related to their work processes are the primary concern (e.g. diesel exposure in miners). Outdoor air pollution becomes an occupational exposure for workers who spend most or all of their working hours in polluted outdoor environments. Exposures for professional drivers, urban traffic police, mail carriers, toll booth operators, municipal workers, street vendors, service workers, and other outdoor occupations are often influenced by traffic-related emissions. Exposures from microenvironments influenced by polluted air can also be important for specialized groups of workers such as subway/underground metro workers and wildfire firefighters; the contribution of outdoor air pollution to occupational exposure in these instances can be substantial. However, few studies are designed to capture this contribution. Relying on fixed outdoor air quality monitors without exposure monitoring or reconstruction often fails to capture the range of exposures for such workers. This section describes the range of exposures to outdoor air pollution in occupational situations experienced by workers in selected jobs, as listed above. See Table 1.14.

(a) Traffic police

Urban traffic police are constantly exposed to traffic-related emissions while controlling traffic, and they may also be regarded as a model for worst-case exposures for air toxics. Many studies of traffic police have relied on outdoor air quality monitoring for criteria pollutants to highlight the potential for high occupational exposures directly attributable to the outdoor environment.

A review of traffic-related exposures (Han & Naeher, 2006) cited additional studies that reported high levels of outdoor exposures for VOCs including benzene, xylene, and toluene in the Republic of Korea (Jo & Song, 2001), India (Mukherjee et al., 2003), and Italy (Bono et al., 2003).

Traffic police on duty at the roadside had significantly higher environmental exposures to PAHs compared with police on office

Table 1.14 Exposure of outdoor workers to air pollutants

Occupation	Exposure measure	Outdoor air concentration (range, if provided)	Location	Comments	Reference
Traffic police	Personal exposure to respirable particulate matter (PM$_5$)	113–878 µg/m^3; average, 322 µg/m^3	Greater Mumbai, India	Similar levels have been reported in Nepal (Majumder et al., 2012)	Kulkarni & Patil (1999)
	Corresponding outdoor air PM$_{10}$ concentration at the nearest air quality monitor	170–320 µg/m^3; average, 143 µg/m^3			
	Breath CO concentrations in non-smoking police officers after work shift	0.46–2.95 ppm	Ankara, Turkey		Atimtay et al. (2000)
	Breath CO concentrations in non-smoking police officers before work shift	0.7–3.37 ppm			
	Corresponding outdoor air concentrations	6.26–23.89 ppm			
	TWA exposure to benzene in traffic police	Geometric mean, 6.8 µg/m^3	Rome, Italy		Crebelli et al. (2001)
	TWA exposure to benzene in indoor workers	Geometric mean, 3.5 µg/m^3			
	PAH exposure for traffic police on active duty at the roadside	74.25 ng/m^3	Thailand	Traffic police on active duty at the roadside had significantly higher environmental exposures to PAHs compared with police on office duty	Ruchirawat et al. (2002)
	PAH exposure for police on office duty	3.11 ng/m^3			
	1-OHP in traffic police on active duty at the roadside	0.181 ± 0.078 µmol/mol creatinine	Thailand		Ruchirawat et al. (2002)
	1-OHP in police on office duty	0.173 ± 0.151 µmol/mol creatinine			
Automobile drivers	Benzene	55.6 (± 9.3) µg/m^3	Manila, Philippines	Jeepney drivers	Balanay & Lungu (2009)
	Toluene	196.6 (± 75.0) µg/m^3			
	Ethylbenzene	17.9 (± 9.0) µg/m^3			
	m,*p*-xylene	72.5 (± 21.1) µg/m^3			
	o-xylene	88.5 (± 26.5) µg/m^3			
	Benzene in urban air	11.8 (± 2.2) µg/m^3			
	Toluene in urban air	83.7 (± 40.5) µg/m^3			
	o-xylene in urban air	38.0 (± 12.1) µg/m^3			
	Toluene in rural air	14.0 (± 6.0) µg/m^3			
	o-xylene in rural air	24.7 (± 11.9) µg/m^3			

Table 1.14 (continued)

Occupation	Exposure measure	Outdoor air concentration (range, if provided)	Location	Comments	Reference
Street vendors/small business operators	Total PAHs on main roads	7.10–83.04 ng/m^3	Bangkok, Thailand	Different occupations in 5 traffic-congested areas of Bangkok	Ruchirawat et al. (2005)
	Benzene levels on main roads	16.35–49.25 ppb			
	Total PAHs at nearby temples (control sites)	1.67–3.04 ng/m^3			
	Benzene levels at nearby temples (control sites)	10.16–16.25 ppb			
	Total PAHs in street vendors	16.07 ± 1.64 ng/m^3			
	Benzene in street vendors	21.97 ± 1.50 ppb			
	Total PAHs in monks and nuns from nearby temples	5.34 ± 0.65 ng/m^3			
	Benzene in monks and nuns from nearby temples	13.69 ± 0.77 ppb			
	Afternoon urinary 1-OHP levels (creatinine) in clothes vendors	0.12 µmol/mol creatinine			
	Afternoon urinary 1-OHP levels (creatinine) in grilled-meat vendors	0.15 µmol/mol creatinine			
	Afternoon urinary 1-OHP levels (creatinine) in controls	0.04 µmol/mol creatinine			
	Afternoon urinary *t*,*t*-MA levels in both groups of street vendors	0.12 mg/g creatinine			
	Afternoon urinary *t*,*t*-MA levels in controls	0.08 mg/g creatinine			
	Benzene in street vendors	83.7 ± 45.0 µg/m^3	Mexico City		Meneses et al. (1999)
	Benzene in office workers	45.2 ± 13.3 µg/m^3			

CO, carbon monoxide; 1-OHP, 1-hydroxypyrene; PAHs, polycyclic aromatic hydrocarbons; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; PM_5, particulate matter with particles of aerodynamic diameter < 5 µm; *t*,*t*-MA, *trans*,*trans*-muconic acid; TWA, time-weighted average.

duty (74.25 ng/m³ vs 3.11 ng/m³) in Thailand (Ruchirawat et al., 2002). Similar observations were reported from another study in Thailand (Arayasiri et al., 2010), which measured benzene and 1,3-butadiene exposures.

(b) Professional drivers

Research from around the world indicates that concentrations of particles and other air toxics in transportation microenvironments on and near roadways and inside vehicles often exceed nearby outdoor levels.

Such exposures are of concern for professional vehicle drivers, especially in developing-country settings, given the rapid increases in high-emitting vehicle fleets, vehicle use, and long exposure durations in and near traffic. For example, a 1997 study in Delhi, India, reported that concentrations of $PM_{5.0}$ and CO inside vehicles exceeded the high urban background concentrations by 1.5–10 times depending on vehicle type (Saksena et al., 2007). Although relatively few studies have been able to characterize outdoor air pollution exposures for automobile drivers, the ratio of reported in-vehicle concentrations to outdoor concentrations indicates the potential for extreme exposures (Apte et al., 2011; Fig. 1.48).

High in-vehicle concentrations would lead to high time-integrated exposures, as reported in the Delhi study (Apte et al., 2011). For example, a typical time-integrated exposure during an average daily commute (1.9 hours/day for auto-rickshaw users; Saksena et al., 2007) is nearly 2-fold higher than entire-day PM exposures for urban California residents (Fruin et al., 2008), the average in-home exposure contributions for residents of seven San Francisco Bay Area single-family homes (Bhangar et al., 2011), and the average for occupants of Beijing high-rise apartments (Mullen et al., 2011). During a typical daily work shift (10–16 hours; Harding & Hussein, 2010), auto-rickshaw drivers may receive very high PM exposures, up to an order of magnitude higher than those experienced during the average daily commute.

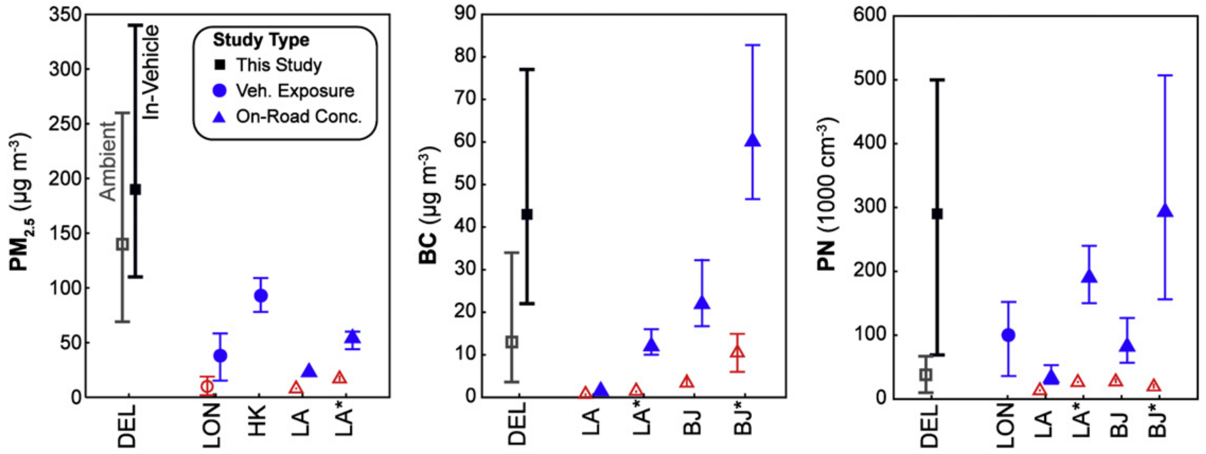
In a study that assessed the occupational exposure of jeepney drivers to selected VOCs in Manila, Philippines (Balanay & Lungu, 2009), personal sampling was conducted on 15 jeepney drivers. Area sampling was conducted to determine the background concentration of VOCs in Manila compared with that in a rural area. Both personal and area samples were collected for 5 working days. Samples were obtained using diffusive samplers and were analysed for VOCs including benzene, toluene, ethylbenzene, *m*,*p*-xylene, and *o*-xylene. The personal samples of drivers (collected for work-shift durations of 12–16 hours) had significantly higher concentrations for all selected VOCs than the urban area samples. Among the area samples, the urban concentrations of benzene and toluene were significantly higher than the rural concentrations. The personal exposures for all the target VOCs were not significantly different among the jeepney drivers.

A recent report (HEI, 2010a) that addressed contributions from mobile-source exposures to air toxics to exposures found that in-vehicle concentrations substantially exceeded outdoor concentrations for 1,3-butadiene, benzene, acrolein, formaldehyde, polycyclic organic matter, and diesel exhaust. This indicates substantial potential for high occupational exposures for many workers who spend long hours in vehicles.

(c) Street vendors/small business operators

Small-scale businesses, commonly street vending, operate primarily outdoors in many developing countries, especially in tropical countries, where weather poses fewer restrictions on spending time outdoors. Furthermore, in the absence of resources for air conditioning or other means of insulation from dust and heat, the work environment in many such small businesses is affected significantly by the prevailing outdoor air quality conditions. Traffic and industrial

Fig. 1.48 Comparison of in-vehicle concentrations in Delhi with those reported in other cities



BC, black carbon mass concentration; BJ, Beijing, China; DEL, Delhi, India; HK, Hong Kong Special Administrative Region; LA, Los Angeles, USA; LON, London, United Kingdom; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; PN, ultrafine particle number concentration.

Plots indicate the mean and range of concentrations.

Reprinted from Apte et al. (2011). *Atmospheric Environment*, Volume 45, Apte JS, Kirchstetter TW, Reich AH, Deshpande SJ, Kaushik G, Chel A *et al.*, Concentrations of fine, ultrafine, and black carbon particles in auto-rickshaws in New Delhi, India, Pages 4470–4480, Copyright (2011), with permission from Elsevier.

emissions thus become a source of occupational exposure. In a study conducted across various susceptible groups of the population with different occupations in five traffic-congested areas of Bangkok (Ruchirawat et al., 2005), the levels of total PAHs on the main roads at various sites were much higher than the outdoor levels in nearby temples (control sites).

In Mexico City, a significant proportion of the labour force works in informal markets, where many vendors spend long hours outdoors. Many workers in the service and transportation sectors experience similar conditions. In Mexico City, about 200 000 people work as taxi and bus drivers and more than 100 000 work as street vendors (SETRAVI, 2007); they have direct exposures to mobile-source emissions on high-traffic-density streets (Ortiz et al., 2002). Compared with indoor workers, these outdoor workers have higher exposures to PM, above the Mexican standard of 65 µg/m³, and 2 or more times higher exposures to ozone, benzene, toluene, methyl *tert*-butyl ether, and 11-pentane (Tovalin-Ahumada & Whitehead, 2007). A survey among outdoor workers found a relationship between their exposure to selected VOCs, ozone, and $PM_{2.5}$ and the presence of severe DNA damage (Tovalin et al., 2006).

1.5 Guidelines and regulations

In many countries around the world, air quality standards are in place for ozone, SO_2, NO_2, CO, PM, and lead (Pegues et al., 2012; Vahlsing & Smith, 2012). Table 1.15 provides a summary of the air quality standards for some example countries. Within countries that have air quality regulations, there is not a consistent approach to regulating important air pollutants. In the USA, these pollutants are referred to as criteria pollutants and NAAQS are established for these pollutants at the national level. The EU has parallel limits for these pollutants but also has air quality limits for benzene, arsenic, cadmium, nickel, and PAHs. National standards in Japan are not set for lead but are set for

Table 1.15 Air quality standards in selected countries (in µg/m^3)[a]

Country	SO_2	NO_2	O_3	$PM_{2.5}$	Lead	PAHs[b]	Benzene	Arsenic
Australia[c]								
1-hour	200 ppb [564]	120 ppb [243]	100 ppb [211]					
4-hour			80 ppb [169]					
1-day	80 ppb [226]			25				
Annual	20 ppb [56.4]	30 ppb [60.8]		8	0.50			
China (Class 2 areas)[d]								
1-hour	500	120	160					
24-hour	150	80		75				
Annual	60	40		35	1.0			
European Union[e]								
1-hour	350	200						
8-hour			120					
24-hour	125							
Annual		40		25	0.5	1 ng/m^3	5	6 ng/m^3
India (residential areas)[f]								
1-hour			180					
8-hour			100					
24-hour	80	80		60	1			
Annual	50	40		40	0.5		5	6 ng/m^3
Japan[g]								
1-hour	100 ppb [282]		60 ppb [127]					
8-hour								
24-hour	40 ppb [113]	40–60 ppb [81–122]		35				
Annual				15			3	
USA[h]								
1-hour	75 ppb [212]	100 ppb [203]						
8-hour			75 ppb [159]					
24-hour				35				
Annual		53 ppb [107]		12	0.15			
WHO[i]								
10-minute	500							
1-hour		200						
8-hour			100					
24-hour	20			25				
Annual		40		10				

NO_2, nitrogen dioxide; O_3, ozone; PAHs, polycyclic aromatic hydrocarbons; PM_{10}, particulate matter with particles of aerodynamic diameter < 10 µm; $PM_{2.5}$, particulate matter with particles of aerodynamic diameter < 2.5 µm; SO_2, sulfur dioxide; TSP, total suspended particles.

[a] Air quality standards are in µg/m^3 unless otherwise specified; conversion from ppb into µg/m^3 is given in square brackets.

Table 1.15 (continued)

[b] Expressed as concentration of benzo[*a*]pyrene.

[c] http://www.environment.gov.au/topics/environment-protection/air-quality/air-quality-standards#air

[d] http://www.mep.gov.cn/image20010518/5298.pdf; standards also exist for TSP, PM_{10}, benzo[*a*]pyrene, carbon monoxide, and fluorine.

[e] http://ec.europa.eu/environment/air/quality/standards.htm; standards also exist for carbon monoxide, PM_{10}, cadmium, and nickel.

[f] http://cpcb.nic.in/National_Ambient_Air_Quality_Standards.php; standards also exist for PM_{10}, carbon monoxide, ammonia, benzo[*a*]pyrene, and nickel.

[g] http://www.env.go.jp/en/air/aq/aq.html; standards also exist for TSP, carbon monoxide, trichloroethylene, tetrachloroethylene, dichloromethane, and dioxin.

[h] http://www.epa.gov/air/criteria.html; standards also exist for carbon monoxide.

[i] http://www.who.int/mediacentre/factsheets/fs313/en/ and WHO (2006).

benzene, trichloroethylene, tetrachloroethylene, dichloromethane, and dioxins. Similar lists of air pollutants are regulated in China and India with direct air quality standards. In some locations where air quality standards have not been developed, the WHO guidelines are used as a reference for air quality management. In many locations around the world, compliance with air quality standards and the WHO guidelines is not achieved.

Given the importance of specific industrial sectors on air pollution, sector-based regulations for emission controls have been developed (Lioy & Georgopoulos, 2011). Important examples are mandated controls on mobile sources, including gasoline-powered motor vehicles and diesel-powered vehicles (see the Annex of IARC, 2013a), stationary power generation, and Portland cement manufacturing. In the case of diesel engines, there are standards for new vehicles in all regions of the world that limit emissions of PM, NO_x, VOCs, and in some countries standards for gas-phase air toxic compounds such as benzene, formaldehyde, acetaldehyde, and butadiene (Diaz-Robles et al., 2013). Likewise, sector-based controls on coal-fired power plants are used to limit emissions of SO_2, NO_x, and mercury. These sector-based control requirements are established at both the national and the regional level, depending on the importance of specific sectors and the importance of the pollutants to local air quality problems. Some sector-based controls are also directed at consumer products and consumables used in industry. Examples include reformulated gasoline, reformulated paints, and replacement of environmental persistent chemicals in consumer goods.

Sector-based controls can take three forms: (1) risk-based (based on risk, not on every source); (2) technology-based (based on the "best" technology for all sources, regardless of risk; e.g. Maximum Achievable Control Technology standards, New Source Performance Standards, or Reasonably Available Control Technology standards); or (3) market-based (cap and trade, emissions taxes, and/or fees) (Farrell & Lave, 2004; Sovacool, 2011). A good example of a regulation or control strategy is that related to HAPs established by the USA: the National Emissions Standards for Hazardous Air Pollutants (NESHAPs) (EPA, 2013f) identify specific pollutants and emissions limits relevant to a wide range of industries.

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