

CHAPTER 3

22643

Human exposure to air pollutants

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Sources of human exposure

General points

3.1. Determination of the risk to human health from air pollutants requires a knowledge of five basic factors:

- (a) the source and emission rates of the pollutant;
- (b) its dispersion and transport in the atmosphere;
- (c) individual exposure;
- (d) individual dose;
- (e) the adverse effects resulting from these doses.

The distinction between exposure and dose is important: **exposure** refers to contact between an airborne pollutant and the human body; it is usually expressed in terms of the pollutant concentration, the duration of exposure and the frequency of exposure. **Dose**, in contrast, refers to the amount of pollutant which is absorbed by the body.

Factors influencing pollutant concentrations outdoors

3.2. Pollutant concentrations vary considerably in time and space as a result of a number of factors which are detailed below.

3.3: *The types of sources, and their location.* For example, sources with tall stacks, such as large power stations, will have much less influence on local concentrations for a given amount of emission than sources emitting close to groundlevel, such as motor vehicles or domestic fires.

3.4: *The rates of emission from these sources, and the number and location of the sources.* Emission rates may vary on a seasonal basis (e.g. sulphur dioxide emissions from electricity generation are generally higher in the colder winter months), on a diurnal basis (e.g. urban vehicle numbers are the highest during rush hours) or on a day-to-day basis (e.g. urban vehicle numbers are usually lower at weekends).

3.5: *Meteorological conditions.* These may influence pollutant concentrations in a number of ways. Atmospheric sta-

bility is often crucial in providing conditions for a smog episode, when a layer of cold air is trapped close to the ground and pollutant concentrations may build up to high levels. Conversely, concentrations may be low when windspeeds are high and no inversion exists. Rainfall may rapidly reduce the concentrations of soluble pollutants such as sulphur dioxide. For some secondary pollutants, such as ozone, concentrations are highest in hot, sunny conditions when the reactions leading their formation proceed most rapidly. Wind direction and circulation patterns may also be important both on a local scale, and on a long-range scale, with air masses which have passed over areas of continental Europe drifting over the U.K.

3.6: *Atmospheric chemical reactions and physical removal processes.* The fate of a pollutant once it is emitted depends largely on its reaction with other chemical constituents, its possible physical transformation (for example, sulphur dioxide may be oxidized and then dissolve in cloud droplets, ultimately to fall as acid rain), and its rate of deposition to vegetation and other ground surfaces [18]. The rate of chemical reaction or physical deposition is not constant, but may depend on climatic conditions, the concentrations of other pollutants, and the nature of the ground surface. Finally, it should be recognized that important chemical reactions can take place within the lung; for example, sulphuric acid aerosol may be neutralized by reaction with the ammonia which all humans exhale [19].

3.7. The relationships between the above factors and pollutant concentration can be described quantitatively, and computer models are commonly used to estimate the concentration of a pollutant at a given place and time. Such models vary in the accuracy of these estimates, with estimates generally being fairly reliable for chemically inert substances over fairly short distances, and less reliable for long-range transport of substances with a complex atmospheric chemistry.

Factors influencing pollutant concentrations indoors

3.8. It is important to note that in the U.K. most individuals spend the majority of their time indoors: one estimate for all developed countries is that, over the population as a whole,

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Table 3.1. Major outdoor and indoor sources of air pollutants

Pollutant	Outdoor sources	Indoor sources
Sulphur Dioxide	Domestic heating Power stations Industrial works Waste incinerators Diesel vehicles	Domestic coal burning Kerosene heaters
Nitrogen Oxides	Transport Power stations Industrial works Waste incinerators	Gas cookers and heaters } unvented Kerosene heaters Tobacco smoke
Particulates	Domestic heating Power stations Transport Industrial works Waste incineration	Tobacco smoke Wood and coal burning Gas and kerosene heaters
Ozone	Secondary pollutant from nitrogen oxides and VOCs	Photocopiers } (poorly maintained) Laser printers
Formaldehyde	Secondary pollutant	Insulation Furnishing Consumer products

89% of time is spent indoors [20]. Knowledge of exposure to air pollution indoors is therefore critical in assessing potential health effects. The concentration of an air pollutant in an indoor environment is influenced by a number of factors, of which the most important are:

- the number and location of indoor sources, and their rates of pollutant emission;
- the characteristics of source use. While some sources may operate continuously, others, such as gas cookers or photocopiers, may operate intermittently;
- the size and structure of the building;
- the rate of air infiltration from outside;
- the ventilation and mixing of air within the indoor environment;
- the removal rate of the pollutant, by physical deposition to surfaces or by chemical transformation;
- the outdoor concentration of the pollutant.

As for outdoor air pollution, these and other factors can lead to large temporal and spatial variations of indoor air pollutant concentrations.

3.9. In the case of some pollutants, especially secondary pollutants such as ozone and sulphate aerosol, indoor emissions are relatively unimportant compared to outdoor sources, and the important factor is the rate of penetration of outdoor pollution. For other pollutants, such as radon or environmental tobacco smoke (ETS), indoor sources are more important than those outdoors, and indoor concentrations are predominantly controlled by emissions from indoor sources and their dispersion and deposition within the building. Finally, for some pollutants such as carbon monoxide or nitrogen dioxide, there are both important indoor and outdoor sources. Table 3.1 provides a summary of the major indoor and outdoor sources for certain air pollutants.

3.10. With a quantitative knowledge of the factors listed above it is possible to develop simple mathematical models to predict indoor pollutant concentrations. The simplest of these envisage the indoor environment as a box and apply a mass balance formula [21]. Such models calculate the total mass of pollutant (from which concen-

trations can be calculated knowing the volume of the box) at any point in time as a function of four possible factors: a pollutant emission rate; an infiltration rate — a function of external concentration and a volume penetration rate; a deposition rate — a function of internal concentration and a deposition velocity; and a recirculation removal rate — a function of internal concentration, volume flow rate through e.g. an air conditioner, and the removal rate in the recirculation system.

Measurement techniques

General points

3.11. For most pollutants a range of measurement techniques is available. An important distinction is between **active** methods which require air to be drawn through a detecting device or chemical absorbent, and **passive** methods which rely on diffusion or deposition for the pollutant to reach the detection unit or absorbant surface. Active methods can also be usefully be divided into those that provide a **continuous**, real-time record of pollutant concentrations, and those which only provide a **time-averaged** concentration. These latter methods normally rely on chemically or physically absorbing the pollutant over a period of hours, followed by subsequent analysis in the laboratory.

3.12. The choice of measurement technique will depend on the objectives of the exercise, and the budget available, as well as the performance of the technique itself. In the context of effects on human health a number of different possible objectives may be identified. For example:

- (a) to demonstrate compliance with an air quality standard set to protect human health. In such cases, the measurement method is often defined in the relevant legislation, and the choice of measurement method is often constrained by the averaging time used to define the air quality standard; for example, when this is defined as a 1-hour average, methods providing daily or weekly averages will not be adequate;
- (b) to provide the information needed to issue smog alerts or public health warnings. In this case, it is essential that continuous real-time information is available;
- (c) to provide data on exposure for use in epidemiological studies of health effects. In this case, the emphasis will be on estimates of the exposure of the individual participants, and thus there may be a need to estimate personal exposure, either directly or indirectly;
- (d) to locate areas with high pollutant concentrations. For such spatial studies, there is a need for a large number of sites but less importance attached to having continuous

measurement; in such situations, the use of simple, cheap passive samplers which provide long-term mean concentrations is often most appropriate.

3.13. In terms of data quality, a number of criteria need to be considered when selecting an appropriate measurement technique.

- (a) *The detection limit of the technique*: this refers to the concentration below which it is not possible to measure the pollutant concentration with the required precision. When a technique relies on absorbing pollutants over a period of time, the detection limit may be defined by the total mass of absorbed pollutant; thus the minimum duration of exposure required to obtain a measurement of a given precision will depend on the concentration to be measured.
- (b) *The precision of the technique*: this refers to the degree of random scatter in the measurement, or the repeatability of the measurement. This would typically be addressed by operating replicate monitors in the same position, or by carrying out replicate measurements on the same sample in the laboratory.
- (c) *The accuracy of the technique*: this refers to the extent to which the measured value deviates from the 'true' value. This deviation, or bias, may be constant or it may be related to the concentration of the pollutant being determined. Bias can arise from a number of sources, but one of the most important is interference from other gases; interference can lead both to positive and to negative bias, depending on the pollutant involved.

Assessment of personal exposure

3.14. Defining human exposure to air pollutants is not straightforward. Most pollutant measurements are based on static devices which provide a record of the concentrations at one point in space. However, most humans are highly mobile and in the course of a day may spend time in many different locations — in the house, in the garden, in the office, in the car, on the street, etc. Thus the exposure of an individual over the course of the day will be a complex function of the temporal variation in pollutant concentration, the spatial variation in pollutant concentration and the activity pattern of the individual. This is illustrated in Figure 3.1, which shows the exposure to nitrogen oxide (NO₂) of a hypothetical commuter over the course of a day. In this particular example, the work exposure is not particularly significant, but in other cases occupational exposure may be the most important element of exposure. Analysis of exposure patterns such as that shown in Figure 3.1 will show large variations between individuals; for any specific pollutant, it may be possible to identify individuals, or groups, who because of their

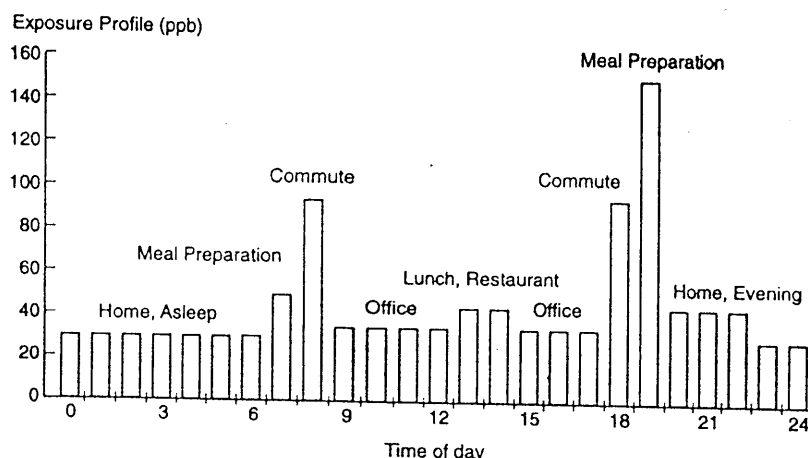


Figure 3.1. Exposure profile to nitrogen dioxide for a hypothetical commuter in an urban environment. (From Loth & Ashmore [37].)

life-style, occupation, etc. have a high exposure and thus a high risk of adverse effects.

3.15. Most pollutant monitors are placed at specific fixed locations — so-called fixed site monitors (FSMs). While this approach to monitoring has considerable value, it gives a very poor indication of the actual pollution exposure experienced by individuals because of their mobility. Since it is the actual exposure of an individual which will determine the likelihood of any adverse effect on the health of that individual, the ability to determine personal exposure is of crucial importance in assessing the health effects of air pollution.

3.16. There are several methods of obtaining an estimate of personal exposure to pollutants, which are summarized in Figure 3.2 [22]. In the **direct** approach, personal monitors are worn or carried by individuals. Most such personal monitors, such as those developed for NO_2 and for respirable particles, can only provide a time averaged concentration, although continuous personal monitors have been used for carbon monoxide (CO), and could be developed for other pollutants. Because of the need for portability, robustness and a light weight, personal monitors are usually less sensitive than the equivalent FSM, and this can restrict their use at ambient concentrations. The use of direct personal monitoring is expensive and time consuming, and it is thus less well-suited to

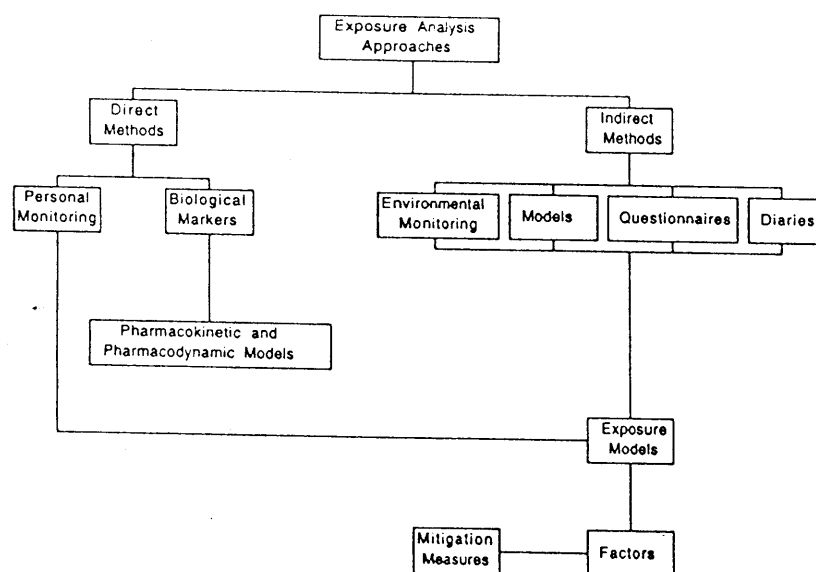


Figure 3.2. Summary of possible approaches for analysis of personal exposure to air pollutants. (From National Research Council [22].) Reprinted with permission from *Human Exposure Assessment for Airborne Pollutants* (1991) p. 43. © 1985 National Academy of Sciences, Washington, DC, USA.

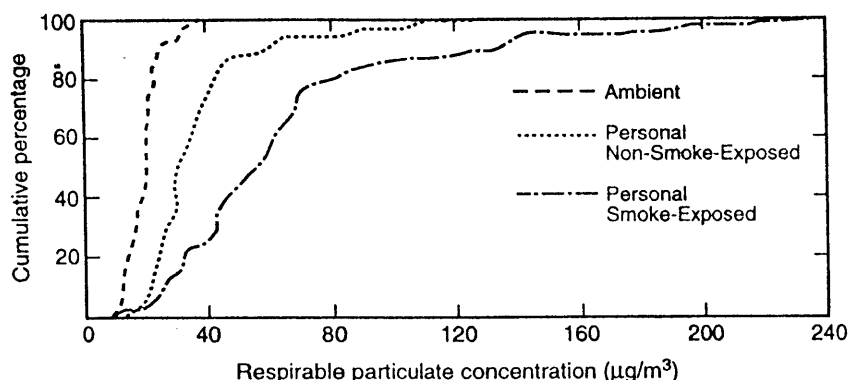


Figure 3.3. Cumulative frequency distributions of central site ambient concentrations, and person exposure in individuals exposed or not exposed to tobacco smoke, for RSP concentrations in two towns in Tennessee. (From Spengler *et al.* [25], reprinted with permission from Environmental Sciences & Technology (1985) Vol. 19, p. 705. © 1985 American Chemical Society, Washington, DC, USA).

estimation of the personal exposure of populations, or large groups. However, it is the only method of directly determining the exposure of individual subjects.

3.17. In the **indirect** approach, estimates of personal exposure are based on models which combine information on pollutant concentrations with a number of other pieces of information. Such models are based on the concept of the micro-environment, which refers to locations, such as kitchen, living room, office, street, restaurant, car, or garage, in which pollutant concentrations are broadly determined by the same factors. To obtain an indirect estimate of personal exposure, a model must combine information on: the time-activity patterns of individuals — when and for long they occupy each micro-environment; on the characteristics of these micro-environments — for example, what type of cookers are used in the kitchen or traffic densities on streets; and on pollutant concentrations in these micro-environments. Estimates of pollutant concentrations may be obtained from personal monitoring, from monitoring at a fixed location in each micro-environment, or from computer models of indoor or outdoor pollutant dispersion.

3.18. Exposure models combine these various types of information in one of two distinct ways. The first of these, the simulation approach, effectively simulates individual exposure from a set of time-activity patterns. For example, in the SHAPE model for CO exposure [23], micro-environment concentrations are expressed relative to those at a background FSM. It is then possible to use the actual FSM data for a particular day, in combination with time-activity records and micro-environmental concentrations, to estimate the time-resolved personal exposure of individuals over the course of the day. The second of these approaches, initially

devised by Duan [24], involves statistically combining records of time-activity patterns of a sample of individuals with a set of micro-environment concentrations to generate time-weighted personal exposures; this method is simpler, but does not provide the level of detail of the time-pattern of personal exposure. A key feature of both these approaches is that micro-environmental concentrations are defined not just as a mean, but as a frequency distribution, so that the range of personal exposures within a population can be estimated.

3.19. A number of studies have compared personal exposures, estimated directly or indirectly, with estimates made on the basis of concentrations recorded simultaneously at FSMs. Consistently large differences have been demonstrated between them. For example, Figure 3.3 shows the frequency distribution of personal exposures to respirable particles (RSP) in two towns in Tennessee, estimated by Spengler *et al.* [25]. The personal exposures are considerably higher than those recorded at a FSM in the town centre, even in those individuals not exposed to tobacco smoke. The personal exposures, estimated using a micro-environmental model developed by Letz *et al.* [26], have both a higher mean and a greater standard deviation than those at the FSM. Thus, it is clear that data from FSMs provides an unreliable estimate of the actual exposure of individuals for use in epidemiological studies or in assessment of policy options.

Major pollutants of concern

General points

3.20. The major pollutants of concern in relation to allergy are summarized in Table 3.1. The pollutants may conven-

Table 3.2. Sources of emissions as fraction of total U.K. in 1990 (from U.K. QUARG [30])

Source	Percentage of total emissions				
	Sulphur dioxide	Black smoke	Nitrogen oxides	Carbon monoxide	VOCs
Road transport	2	46	51	90	41
Electric supply industry	72	6	28	1	—
Other industry	19	14	9	4	52
Domestic	3	33	2	4	2
Other	7	1	9	—	4
Total (kT)	3774	453	2719	6659	2396

iently be divided into primary pollutants, such as sulphur dioxide (SO_2) and carbon monoxide (CO), which are emitted directly into the atmosphere, and secondary pollutants such as ozone (O_3) which are formed by subsequent chemical reactions in the atmosphere. The distinction is not always a clear one; in the case of nitrogen oxides, for example, most primary emissions are in the form of nitric oxide (NO), which is oxidized to nitrogen dioxide (NO_2) in the atmosphere. Although most NO_2 is thus secondary, there is a small primary emission of NO_2 from motor vehicles.

3.21. Table 3.2 gives a quantitative breakdown of the major outdoor sources of the primary pollutants for the U.K. as a whole. For any area, however, the quantitative breakdown of

sources may be very different; in general local, low level sources will become more significant. Thus Figure 3.4 illustrates the national variation in the proportion of ground level nitrogen oxides which is due to road transport; this shows a variation from 30% or lower in remote rural areas to over 70% in the centre of large cities [27].

3.22. While some of the pollutants listed in Table 3.1 are distinct chemical species, others are much more complex in their physical and chemical constitution. This applies particularly in the case of airborne particles. The variation in physical size and chemical character of particulates can partly be related to their origin, which can be either primary — emitted from sources such as diesel vehicles, coal fires or indus-

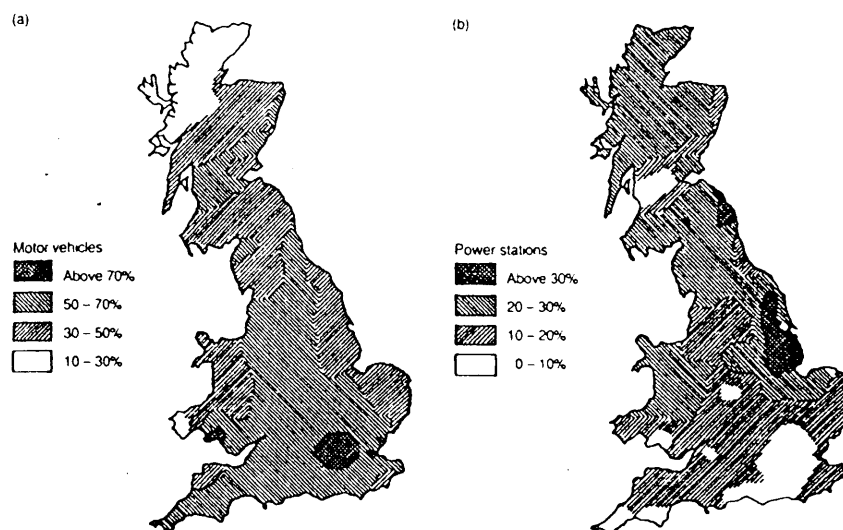


Figure 3.4. Modelled contributions to U.K. ground-level NO_2 concentrations from (a) motor vehicles and (b) power stations. (From U.K. PORG [27].)

trial works, or secondary — formed in the atmosphere from condensation or chemical reactions. The impacts of particles on human health may be due to the physical effects of the particles, or due to chemical components, such as heavy metals, organic compounds or acid species.

Sulphur dioxide

3.23. SO_2 can be emitted from combustion of any material containing sulphur, with emissions being related to its sulphur content. Hence emissions from fuels such as coal and kerosene are much higher than, for example, natural gas.

3.24. Historically, the major source contributing to high outdoor concentrations of SO_2 in the U.K. was domestic coal burning; under inversion conditions, especially during the winter, SO_2 emissions from these sources could lead to severe pollution episodes, such as the London smog of December 1952. The elimination of this source in most areas of the U.K., together with other measures such as reductions in the sulphur content of heating oil, has led to a dramatic fall in urban concentrations over the last 40 years. It is noticeable that those towns where domestic coal burning still continues (e.g. Belfast and some coal mining areas) now have the highest SO_2 concentrations in the U.K. The major source of SO_2 nationally is now large combustion plants, whose tall stacks normally disperse pollutant effectively. However, under certain meteorological conditions, the plumes from such plants can contribute to high ground-level concentrations. Indoor, the major source of SO_2 is also fuel combustion.

3.25. There are a number of possible techniques for the measurement of SO_2 concentrations, depending on the time resolution and specificity required. Historically, measurements at a large number of U.K. sites were obtained using a method in which air was bubbled through a dilute hydrogen peroxide solution in a glass Dreschel bottle for a period of 24 hours. SO_2 reacts with hydrogen peroxide in the bubbler to form sulphuric acid and subsequent titration provides a measure of total acidity [28]. However, because the method is based on a measurement of acidity, there may be interference from other acidic or alkaline compounds in the atmosphere; for example if the bubbler is situated close to a source of ammonia, SO_2 levels may be significantly underestimated. There are similar bubbler methods, such as the pararosaniline method [29] which provide 24-hour mean concentrations which are more specific to SO_2 ; however, the chemical analysis involved in these methods is more complex than that in the total acidity method. Continuous measurements of SO_2 concentrations are made using UV fluorescence monitors, but these are much more expensive.

Nitrogen oxides

3.26. Like SO_2 , the major source of nitrogen oxides is fuel combustion. However, unlike SO_2 , it is not the nitrogen content of the fuel which is important in determining the emission rate but the temperature of combustion, which affects the rate of reaction between atmospheric nitrogen and oxygen. Hence, fuels with a high combustion temperature such as natural gas, tend to have a higher emission rate than fuels such as kerosene which have a lower combustion temperature. Emissions from motor vehicles increase with speed, with emissions at 120 km/h being 2–3 times greater than those at 20 km/h [30]. The fitting of catalytic converters should in principle reduce emissions by about 90%, although the poorer catalyst performance in the first few minutes of a journey, and maintenance failures, will in practice reduce this figure. Data from on-road tests in urban areas suggest a reduction in emissions of about 75% with the catalytic converter [31]. It should also be noted that although nitrogen oxide emissions from diesel cars are lower than those from petrol engines, once three-way catalytic converters are fitted to petrol engines, emissions are about half of those from diesel cars.

3.27. There are a range of oxides of nitrogen either emitted into the atmosphere or formed there by secondary reactions. The two most important are nitric oxide (NO), which is the major primary emission, and NO_2 , which has small primary emissions, and is largely produced in the atmosphere by oxidation of NO by ozone, a reaction which can take less than one minute. However, when the supply of ozone is limited, as it often is close to major roads, the conversion rate is slow and much of the emitted nitrogen oxides may remain in the form of the relatively harmless NO.

3.28. There are also significant indoor sources of nitrogen dioxide, especially gas cookers and gas and paraffin heaters which are not properly ventilated or maintained. Measurements suggest that in homes without such devices, concentrations are below those outdoors by a factor of about 50%. However, in kitchens with unventilated gas cookers, mean NO_2 concentrations may be 2–4 times those outdoors with higher concentrations during those periods when the cooker is in use [32]. Studies have also shown large variations between individual houses in indoor NO_2 levels [33].

3.29. Continuous measurements of oxides of nitrogen may be made using chemiluminescent monitors, which are based on a reaction with ozone. These instruments respond not only to NO and NO_2 but also to some extent to other minor nitrogen oxides. These interferences are probably trivial in typical urban situations but may be more important in re-

mote locations. An important recent development is the use of passive diffusion tubes to measure NO_x concentrations; these tubes contain a chemical absorbant of NO_2 , triethanolamine, with the total amount of absorbed NO_2 then being determined colorimetrically. These tubes are cheap, and thus can be easily used to obtain detailed spatial information; they have also been used for micro-environmental and personal monitoring. However, they can only determine mean concentrations, typically over 1–2 weeks, and there is evidence that diffusion tubes tend to overestimate actual outdoor NO_2 concentrations.

Ozone

3.30. Ozone is formed as a result of a complex series of atmospheric reactions from a range of precursors. The most important of these are nitrogen oxides, volatile organic compounds (VOCs) and carbon monoxide. Some of the key reactions are photochemical — they require the presence of light — and they also proceed more rapidly at higher temperatures. Furthermore, emission rates for VOCs tend to be higher at higher temperatures. Ozone is a major component of 'photochemical smogs', or 'summer smogs', but these also contain high concentrations of aerosols, and gases such as hydrogen peroxide and peroxyacetyl nitrate. Outdoors, ozone concentrations tend to be high on hot, sunny days, and the frequency of ozone episodes of potential concern for human health is much higher in hot dry summers such as 1990 and 1989 or during the scorching summer of 1976 when daily peak O_3 concentrations in SE England were greater than 200 ppb for 2 weeks.

3.31. In addition to these sources, ozone can also result from downward fluxes from the stratospheric ozone layer, and from chemical reactions in the background troposphere. There is some evidence from various locations around the world that these background tropospheric concentrations have increased over the last 100 years from about 10 ppb to 20–30 ppb in the present day. During photochemical episodes in the U.K., concentrations may exceed 100 ppb, with the highest concentration ever recorded being 258 ppb in the summer of 1976.

3.32. There are few major indoor sources of ozone, although poorly maintained photocopiers or laser printers can affect indoor concentrations in small badly ventilated rooms. However, in general, indoor concentrations are lower than those outdoors, although there are few measurements in the U.K. to demonstrate this. Of course, hot sunny days with high O_3 concentrations are also the days on which people tend to spend the greatest amount of time outdoors, or have windows open when they are indoors.

3.33. Continuous measurements of ozone can be made using a number of techniques, but that now almost universally adopted is based on the absorption of UV radiation.

Particulates

3.34. The chemical and physical nature of particulates is strongly dependent on the source of the material, and heavily influences their impacts on health. The concentration and mix of particles in the atmosphere is highly dynamic, but essentially is determined by three factors: the emission of particles from primary sources; the creation of particles in the atmosphere by vapour condensation or chemical reaction between gases and vapours; and their removal by sedimentation, diffusion or impaction.

3.35. These particles vary greatly in size, and may range from 100 μm in diameter to less than 0.1 μm . This variation in size affects the physical processes by which particles are deposited, with the larger particles with greater mass tending to deposit more quickly. This is also of great importance in determining the extent to which particles penetrate into the lung. Particles more than 10 μm in diameter will almost all be deposited in the nasopharyngeal region, and it is only the fine particles, with relatively small diameters, which penetrate into the smaller bronchi and alveoli. Recent epidemiological evidence suggests that it is the particulate fraction below 2–3 μm which is most closely related to long-term adverse effects on health [15].

3.36. In practice, atmospheric particles are often described on the basis of the measurement technique used. Historically, the U.K. measurement networks have employed a method in which air is drawn through a filter over a period of 24 hours, and then its darkness is measured — hence the term Black Smoke. While this provides a cheap and easy method, its dependence on the blackness of the particulate make it less appropriate than in the days when coal smoke predominated. The other parameters of interest are the mass concentration, the particle size distribution, and the chemical composition. Size distribution is most commonly assessed using multi-stage impactors, which remove particles of progressively smaller diameter; the mass and chemical composition of these size fractions can then be determined. For routine monitoring, a single size-selective inlet is used to restrict the measurement to particles below a certain size, with 10 μm often being used as a cut-off (so-called PM_{10}). In other cases, a smaller cut-off of 2.5 μm is used to provide a measure sometimes referred to as respirable suspended particulate (RSP). A recent development is the design of instruments to give better time resolution, such as the Tapered Element Oscillating Microbalance (TEOM) now being used to provide hourly

mean PM_{10} concentrations from stations in the U.K. Enhanced Urban Network. An alternative to the use of gravimetric techniques is the use of instruments that measure the degree of light-scattering caused by the particulate; these provide real-time data, but measure only total particle number concentrations.

3.37. The atmosphere naturally contains a variety of particulate material, such as dust, salt spray, pollen, fungi, and *motus*. Forty years ago, the major anthropogenic source of particulate in U.K. urban areas was domestic coal burning. In most areas, this source is now removed, and the most important current sources include motor vehicles, especially those with diesel engines, industry and construction. Indoor sources of particulates are primarily combustion for heating and cooking, and tobacco smoke.

3.38. The relationship between indoor and outdoor concentrations depends very much on the particle size. For particles in the size range 2.5–10 μm , studies have shown that, in the absence of indoor sources, indoor concentrations are much lower than those outdoors, whereas for particles below 2.5 μm , differences between indoor and outdoor concentrations are small [34].

3.39. A particulate fraction of interest in relation to health impacts is acid aerosol, especially sulphate aerosol, for which there is evidence both from controlled human exposure studies and from epidemiological work of adverse effects (see Section D). Sulphate aerosols are secondary particulates, produced by the oxidation of SO_2 and its dissolution in water droplets; small acid droplets can also form by condensation of acid gases. Since the rate of oxidation of SO_2 is typically only a few per cent per hour [30], sulphate aerosol tends to be a regional rather than a local pollutant. Sulphuric acid aerosol, once formed, can be neutralized, primarily by ammonia to form ammonium sulphate aerosol. High sulphate aerosol concentrations can be associated with both winter smogs and summer smogs. Because of its relatively small diameter, sulphate aerosol concentrations tend to show relatively little difference between indoors and outdoors [35].

3.40. The most common method of measuring acid aerosol involves drawing air through a diffusion denuder tube and then collecting it on a Teflon filter pack. The denuder tube serves to remove ammonia and other bases which otherwise would neutralize acidity collected on the filter. The acidity is then extracted from the filter and determined.

Formaldehyde

3.41. Although some human exposure to formaldehyde can occur in the outdoor environment the most important sources

of exposure are indoors. Outdoors, concentrations may be elevated around specific chemical processes involving formaldehyde, and it is a minor component of photochemical smogs. Indoors, the major contributing sources are insulating materials especially urea-formaldehyde cavity wall insulation, chipboard and fabrics while smoking is also a significant source in some situations. Formaldehyde can also be present in high concentrations in some occupational settings, e.g. chipboard factories.

3.42. Average concentrations in homes typically lie between 50 and 100 $\mu g/m^3$, with occasional buildings having higher values; concentrations outdoors are typically one order of magnitude lower. In the case of houses with urea-formaldehyde insulation, concentrations may be higher in the first few weeks after installation.

Environmental tobacco smoke

3.43. Although cigarettes are smoked both indoors and out, it is only in indoor environments that concentrations will build up to significant levels. Environmental tobacco smoke (ETS) is not a single compound, but a complex mixture of chemicals in both the particle and vapour phases; one estimate is that it contains more than 3800 different chemicals. The most important chemicals in this mix are: carbon monoxide, nitrogen oxides, respirable particles, and a complex mix of organic compounds including nicotine, formaldehyde, benzene, nitrosamines, and polycyclic aromatic hydrocarbons.

3.44. For these reasons, it is impossible to measure ETS directly; instead the usual approach is to measure one or more of these key components which are of medical concern, such as CO or particulates, as a marker; however, since these have other sources, they are not specific to ETS. Biological markers of ETS exposure have also been used to assess personal exposure. Of these, the presence of nicotine or cotinine in blood, urine and saliva is specific for ETS exposure, and provides good semi-quantitative estimates of integrated exposure over a period of hours or days [22]. However, there is no reliable, specific and quantitative biological marker of long-term exposure to ETS.

3.45. As with any indoor pollutant, the concentrations experienced will depend on the number and intensity of sources and the ventilation characteristics. For these reasons, the highest concentrations are typically found in crowded clubs, pubs and bars. Each chemical component of ETS will have its own characteristic rate of removal from the atmosphere; generally, carbon monoxide and nitrogen oxides deposit relatively slowly, while nicotine and respirable particles are deposited relatively quickly [36]. For this reason, it is difficult to cite typical concentrations of ETS. However, there is little doubt

that concentrations of key components of ETS, such as respirable particulates, are higher in homes or offices where smoking occurs [25].

Pollutant mixture

3.46. The above text has considered each individual pollutant in isolation. However, because these pollutants often have similar sources, and because poor dispersion conditions will lead to the build-up of high concentrations of many pollutants, air pollution episodes are frequently characterized by mixtures of pollutants, e.g. smoke and sulphur dioxide, with high levels of sulphate aerosol, in the historic London smogs. Similarly, the combination of specific sources and poor ventilation conditions can lead to the build-up of high concentrations of several pollutants together indoors. When considering effects of air pollutants on allergic disease, it is also important to consider whether conditions leading to high pollutant concentrations also lead to high exposures to allergens.

Summary and future research directions

3.47. Any epidemiological study of the impact of air pollution on human health requires a precise and unbiased estimate of subjects' exposure to the air pollutants of concern. In practice, this is often difficult because of the cost and technical limitations of the measuring techniques which are available,

but more importantly because of the mobility and unique time activity patterns of individuals. The assessment of personal exposure is recognized to be a major limitation in epidemiological studies, and this is an important priority for future research in the U.K., both in terms of direct personal monitors and indirect modelling methods [37].

3.48. In terms of the changing prevalence of allergic diseases, it is also important to evaluate critically the real changes which have occurred in human exposure. While the sources of outdoor air pollution in U.K. cities have changed dramatically over the past 40 years, there have also been important changes in indoor sources of air pollution, and in people's activity patterns.

3.49. Finally, effective pollution control policy will require an understanding of how measures to alter emissions from different sources influence personal exposure, and hence health outcomes. Of critical importance here is a knowledge of the frequency distribution of individual personal exposures within a population, and this implies further development of suitable indirect techniques of exposure estimation. It is also important to evaluate carefully all potential sources and the potential benefits of controlling them; for example, because of the length of time people spend indoors, it may be that for some pollutants measures to reduce indoor exposure are potentially more effective than those which reduce outdoor exposure.