

# Indoor Air Pollution

## *Sources, Levels, Chemistry, and Fates*

At the beginning of this book, we presented some discussion of health-based air quality standards. In the final chapter, which follows this one, the scientific bases of control measures for various pollutants are discussed. In between, the complex chemistry that occurs in both polluted and remote atmospheres, and that converts the primary pollutants into a host of secondary species, has been detailed. To provide further perspective on airborne gases and particles and human exposure levels, we briefly treat indoor air pollution in this chapter. As we shall see, for many species it is simply a question of emissions leading to elevated levels indoors. However, there is some chemistry that occurs in indoor atmospheres as well, and it is of interest to compare this to that occurring outdoors.

From the point of view of health impacts, it is the combination (not necessarily linear) of concentrations and duration of exposure to a pollutant or combination of pollutants that is important. In this regard, it is noteworthy that most individuals spend the majority of their time indoors, even in relatively moderate climates. For example, in California, people spend 87% of their time indoors on average, 7% in enclosed transit systems, and only 6% outdoors (Jenkins *et al.*, 1992). Similarly, Quackenboss *et al.* (1986) report that only 15% of the average day in Portage, Wisconsin, was spent outdoors in summer and less than 5% in winter. As a result, elevated concentrations of air pollutants indoors can have a significant impact on human health and can lead to enhanced chemical sensitivities (Hileman, 1991) as well as other health impacts such as cancer. Establishing the contribution of indoor air pollution to carcinogenicity and assessing the relative risks (e.g., Tancredi *et al.*, 1987) are complex and difficult. An interesting approach suggested recently is pet epidemiology, discussed in detail by Bukowski and Wartenberg (1997).

The term “indoors” is used in the literature to refer to a variety of environments, including homes, work-

places, and buildings used as offices or for recreational activities. In addition, a number of studies have been carried out to measure various compounds inside vehicles during commutes. As we shall see, and consistent with expectations, levels measured indoors are characteristic both of the particular sources present and, to a significant extent, of the outdoor concentrations of the species. We shall not, in general, distinguish in this chapter between the various types of indoor environments but rather focus on the sources of various compounds and their indoor chemistry.

Table 15.1 summarizes the major species of concern for indoor air pollution and some of their sources (Su, 1996). We focus in this chapter primarily on those species common to indoor and outdoor air environments, including oxides of nitrogen, volatile organic compounds (VOC), CO, ozone, the OH radical, SO<sub>2</sub>, and particles. In addition, a brief discussion of radon is included since this has been one of the major foci of concern in the past with respect to indoor air pollution.

### A. RADON

Radon (<sup>222</sup>Rn) is formed by the radioactive decay of uranium, <sup>238</sup>U (Fig. 15.1a). As a result, the highest concentrations tend to be associated with soils derived from rocks with a high uranium content (Nazaroff and Nero, 1988; Boyle, 1988; Nero, 1989; Mose and Mushrush, 1997). Because radon is a gas that diffuses out of the soil, it can enter homes through cracks in the foundation, around loose-fitting pipes and wall joints, and through floor drains (e.g., Nero, 1989). The concentrations found in a home depend on the type of soil (including the moisture content) on which it sits and the extent of Rn penetration into the house. They also depend on the house ventilation rate and the particular location in the house in which the measurement is

TABLE 15.1 Some Indoor Air Pollutants and Their Sources<sup>a</sup>

Species	Sources
Radon ( <sup>222</sup> Rn)	Soil and some masonry building materials
Oxides of nitrogen	Combustion
Volatile organic compounds (including HCHO)	Building materials, carpets, solvents, paints, household and personal care products, air fresheners, pesticides, mothballs, humans, treated water
CO	Combustion
O <sub>3</sub>	Outside air, photocopying machines, electrostatic air cleaners
SO <sub>2</sub>	Combustion
Particles	Combustion
Asbestos	Building materials, handheld hair dryers
Microorganisms	Air conditioners, cool-mist humidifiers

<sup>a</sup> Adapted from Su (1996) and Nero (1988).

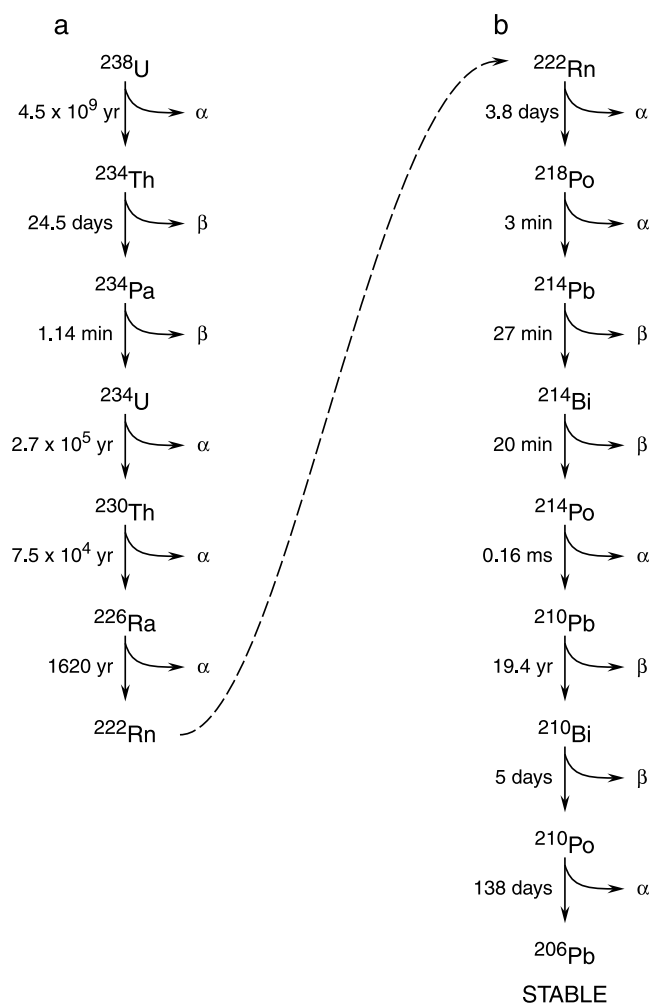


FIGURE 15.1 The major radioactive decay paths in (a) the uranium series leading to the formation of <sup>222</sup>Rn and (b) of <sup>222</sup>Rn to form <sup>206</sup>Pb. Half-lives are shown to the left of each process.

made, with the basement typically having the highest concentrations (e.g., Liu *et al.*, 1991). Interestingly, because homes are often warmer than their surroundings, a “chimney effect” occurs that draws gases, including radon, into the house from the surroundings (e.g., Osborne, 1987; Nero, 1989; Turk *et al.*, 1990; Hintenlang and Al-Ahmady, 1992).

Other sources include building materials such as concrete that are made from the earth’s crustal materials and hence can contain significant amounts of uranium and radium (Nazaroff and Nero, 1988). Radon dissolves in water, and hence degassing from household water can also be a source. For example, Osborne (1987) reported that the radon concentration in a bathroom increased by more than two orders of magnitude during a 15-min period that a shower was running.

The health concerns associated with <sup>222</sup>Rn are primarily associated with its radon daughters. As a noble gas, radon is unreactive in air and is both readily inhaled and exhaled. However, a significant portion of its daughters are positively charged ions that are expected to attract water vapor and become hydrated; the formation of clusters with other ions is also likely (e.g., Castleman, 1991). Uptake on existing aerosol particles also occurs readily, and such particles can then be deposited in the respiratory tract, providing a source of radioactive emissions directly to the lung. Effects such as lung cancer may then ensue.

The health effects associated with radon, as well as sources and mitigation measures, are discussed in detail in several National Research Council reports (1988, 1991), in the book edited by Nazaroff and Nero (1988), and in the International Commission on Radiological Protection Report (1994). Initial risk assessments were based on data from underground miners who were exposed to relatively high levels of radon and its progeny. However, there has been considerable controversy over the extrapolation to lower levels in homes [e.g., see summaries by Nazaroff and Teichman (1990) and Peto and Darby (1994)].

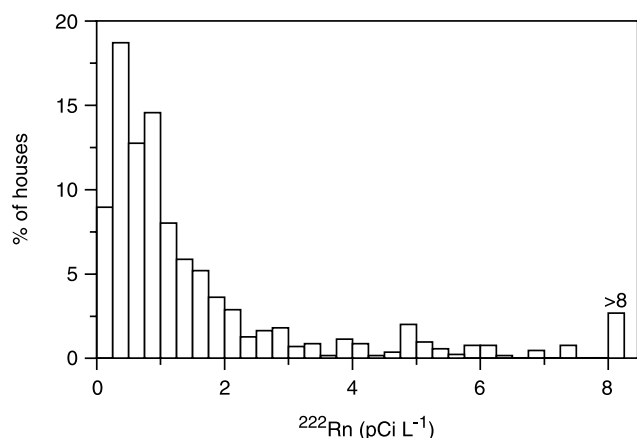


FIGURE 15.2 Distribution of  $^{222}\text{Rn}$  measured in homes in the United States (adapted from Nero *et al.*, 1986).

A number of studies have been carried out to determine the concentrations of radon in homes (e.g., see Nero *et al.*, 1986; Alter and Oswald, 1987; Turk *et al.*, 1990; Liu *et al.*, 1991; and Mose and Mushrush, 1997). Units used to express the concentration of  $^{222}\text{Rn}$  are picocuries per liter ( $\text{pCi L}^{-1}$ ), with 1 pCi being the amount of substance that gives 2.2 radioactive decays per minute, or becquerels per cubic meter ( $\text{Bq m}^{-3}$ ), where  $1 \text{ pCi L}^{-1} = 37 \text{ Bq m}^{-3}$  (e.g., see Nazaroff and Teichman, 1990).

Figure 15.2 shows the distribution of levels measured in homes in the United States (Nero *et al.*, 1986). As might be expected given the variables that affect  $^{222}\text{Rn}$  concentrations in homes, the concentrations vary widely, from  $<0.1$  to  $>8 \text{ pCi L}^{-1}$ . Indeed, Alter and Oswald (1987) report a few single measurements of up to  $4000 \text{ pCi L}^{-1}$ . For comparison, the U.S. EPA recommends levels below  $4 \text{ pCi L}^{-1}$  ( $150 \text{ Bq m}^{-3}$ ), which is exceeded by about 7% of U.S. homes (Nero *et al.*, 1986), and the International Commission on Radiological Protection (1993) recommends  $5.4\text{--}16 \text{ pCi L}^{-1}$  ( $200\text{--}600 \text{ Bq m}^{-3}$ ). Typical outdoor concentrations in continental areas are  $0.1\text{--}0.4 \text{ pCi L}^{-1}$  (Nazaroff and Nero, 1988).

## B. OXIDES OF NITROGEN

### 1. Levels of $\text{NO}_x$

A large number of studies of NO and  $\text{NO}_2$  have been carried out in many different indoor air environments. Because of air exchange, indoor levels are generally higher when outdoor levels increase (e.g., Hoek *et al.*, 1989; Rowe *et al.*, 1991; Hisham and Grosjean,

1991a; Spengler *et al.*, 1994; Weschler *et al.*, 1994; Baek *et al.*, 1997). However, enhanced indoor levels are found when combustion sources are present. These include gas stoves, kerosene heaters, water heaters, and cigarette smoke (e.g., Wade *et al.*, 1975; Marbury *et al.*, 1988; Ryan *et al.*, 1988; Petreas *et al.*, 1988; Hoek *et al.*, 1989; Pitts *et al.*, 1989; Spengler *et al.*, 1994; Levy *et al.*, 1998). While combustion generates primarily NO, the focus indoors has been on  $\text{NO}_2$  because of its health impacts (see Chapter 2.).

For example, Spengler *et al.* (1994) report that personal exposures to  $\text{NO}_2$  were 10 ppb higher in homes in the Los Angeles area having gas ranges with pilot lights compared to those having electric ranges and 5 ppb higher if the gas range did not have a pilot light. Levy *et al.* (1998) report  $\text{NO}_2$  concentrations indoors and outdoors, as well as personal exposures, in 18 different cities in 15 countries in the Northern Hemisphere over a two-day period in February and March when the use of combustion was expected to be higher. The ratio of indoor to outdoor concentrations ranged from  $0.3 \pm 0.2$  in Berlin, Germany, to  $2.8 \pm 2.8$  in Tokushima, Japan. Average  $\text{NO}_2$  concentrations over the two-day period indoors ranged from 5.5 ppb in Kuopio, Finland, to 63 ppb in Mexico City. Outdoors, the range of concentrations was from 12 ppb in Geneva, Switzerland, to 52 ppb in Seoul, Korea.

Again, the use of gas stoves was highly correlated with indoor  $\text{NO}_2$ , with an indoor/outdoor concentration ratio of 1.19 for homes with a gas range compared to 0.69 for those without a gas stove. The ratio was even higher for homes with a kerosene space heater, 2.3 compared to 0.85 without such a heater (Levy *et al.*, 1998). Both the indoor and outdoor concentrations of  $\text{NO}_2$  were higher in cities where at least 75% of the homes had gas stoves; for example, the mean outdoor  $\text{NO}_2$  concentration in such gas-intensive cities was  $38 \pm 20$  ppb, compared to  $14 \pm 6$  ppb in cities where fewer than 25% of the households had gas ranges.

High concentrations of  $\text{NO}_2$  have also been measured in indoor skating rinks where the use of ice resurfacing machines powered by propane, gasoline, or diesel fuel results in significant emissions (e.g., Brauer and Spengler, 1994; Brauer *et al.*, 1997; Pennanen *et al.*, 1997). Mean concentrations of  $\text{NO}_2$  of  $\sim 200$  ppb have been reported, with some rinks having concentrations up to 3 ppm! The indoor-to-outdoor ratios of the arithmetic mean concentrations varied from about 1 to 41, with an overall mean of 20. Figure 15.3 shows the effect of the particular fuel used on the indoor  $\text{NO}_2$  concentrations (Brauer *et al.*, 1997). Propane gave the highest values, followed by gasoline and diesel. The  $\text{NO}_2$  concentrations when the resurfacers were electric were similar to those outdoors.

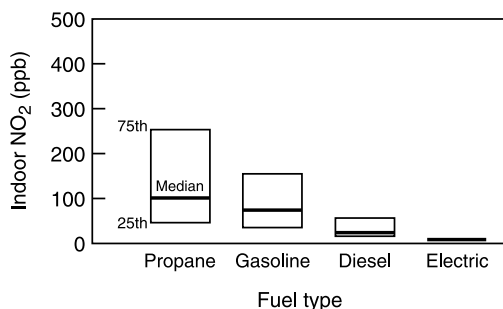


FIGURE 15.3 Indoor concentrations of  $\text{NO}_2$  measured in ice skating rinks using different types of power for the ice resurfacers. The median and the 25th and 75th percentiles are shown in each case (adapted from Brauer *et al.*, 1997).

In the absence of such sources of  $\text{NO}_x$ , indoor and outdoor concentrations are quite similar (e.g., Weschler *et al.*, 1994), since removal of  $\text{NO}$  and  $\text{NO}_2$  indoors, e.g., on surfaces, is relatively slow. However, as discussed shortly, although the surface reaction of  $\text{NO}_2$  is relatively slow, it is still of interest since it generates nitrous acid ( $\text{HONO}$ ). Different surfaces found inside homes have been found to have different removal rates for  $\text{NO}_2$ . Figure 15.4, for example, shows measured rates of removal of  $\text{NO}_2$  by a number of common household materials (Spicer *et al.*, 1989). Large variations in removal rate (and hence the formation of products such as  $\text{NO}$  and  $\text{HONO}$ ; see later) are evident, varying from negligible for plastic storm windows to quite large for wallboard.

In short, there is a variety of evidence that there are higher levels of  $\text{NO}_2$  indoors when combustion sources are present and that the concentrations generated indoors can be quite substantial in some circumstances. One word of caution is in order, however, particularly with regard to earlier measurements of  $\text{NO}_2$ . As discussed in the following section, significant concentrations of  $\text{HONO}$  are generated both by a heterogeneous reaction of  $\text{NO}_2$  on surfaces and by direct emissions from combustion sources. In some measurement methods used for  $\text{NO}_2$ ,  $\text{HONO}$  is also detected and hence reported as  $\text{NO}_2$ . This is particularly true for the  $\text{O}_3$  chemiluminescence method and for electrochemical sensors (e.g., Spicer *et al.*, 1994), so that  $\text{NO}_2$  reported using these techniques should be regarded as upper limits to its concentrations. This problem can be circumvented through the use of denuders to remove  $\text{HONO}$  prior to sampling into the instrument.

## 2. $\text{HONO}$ and $\text{HNO}_3$

As discussed in Chapter 7.C.1,  $\text{HONO}$  is formed by the reaction of  $\text{NO}_2$  with water on surfaces. The reac-

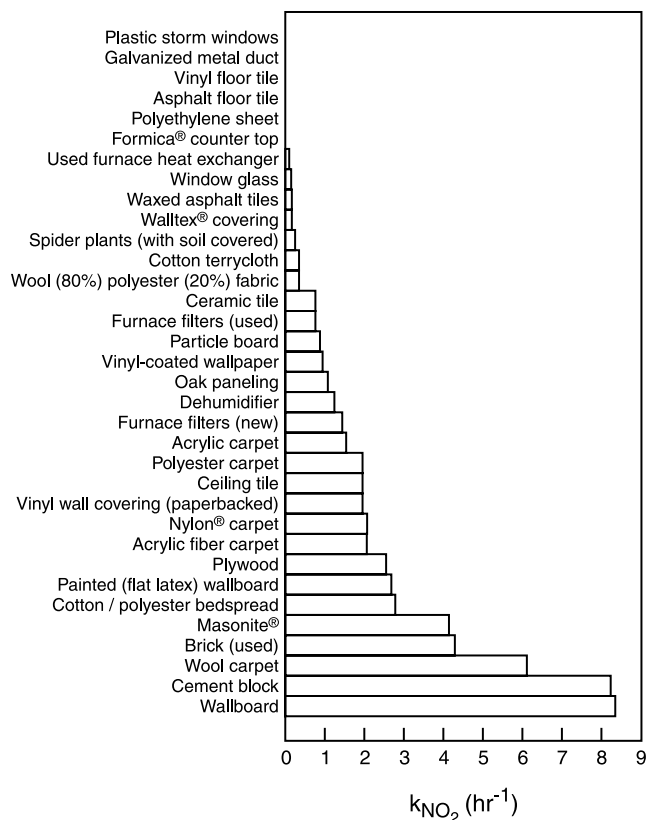


FIGURE 15.4 Rate constants for  $\text{NO}_2$  removal on  $3.3\text{-m}^2$  samples of different materials commonly found in homes (adapted from Spicer *et al.*, 1989).

tion is usually represented as



although the detailed mechanism is not known; gaseous  $\text{HNO}_3$  is not generated in equivalent amounts, which has been attributed to its remaining adsorbed on the surface. This overall reaction occurs on a variety of surfaces in the laboratory and hence might be expected to also occur on surfaces in other environments, such as homes. This, indeed, is the case.

Pitts *et al.* (1985) first used differential optical absorption spectrometry (DOAS) to establish unequivocally that  $\text{NO}_2$  injected into a mobile home forms  $\text{HONO}$ . Interestingly, the dependence of the rate of  $\text{HONO}$  generation on the  $\text{NO}_2$  concentration was similar to that measured in laboratory systems, consistent with production in, or on, a thin film of water adsorbed on surfaces. A number of studies have confirmed that the behavior is similar to that in laboratory systems; i.e., the rate of production of  $\text{HONO}$  increases with  $\text{NO}_2$  and with relative humidity. Indoor levels of  $\text{HONO}$  as high as 8 ppb as a 24-h average and 40 ppb as a 6-h

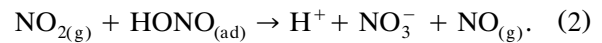
average have been reported in normal, in-use buildings and homes (Febo and Perrino, 1991; Spengler *et al.*, 1993; Weschler *et al.*, 1994).

The ratio of HONO to  $\text{NO}_2$  indoors can be quite large, up to  $\sim 0.15$  (e.g., Febo and Perrino, 1991; Brauer *et al.*, 1990, 1993; Spengler *et al.*, 1993). This can be compared to typical values of a few percent outdoors (see Chapter 11). High levels of HONO (up to  $\sim 30$  ppb) have also been measured in automobiles in use in polluted urban areas, and again, the ratio of HONO to  $\text{NO}_2$  was quite large,  $\sim 0.4$ , compared to 0.02–0.03 measured outdoors in the same study (Febo and Perrino, 1995).

Figure 15.5, for example, shows one set of measurements of  $\text{NO}_2$ , HONO, and NO after injection of  $\text{NO}_2$  into a home used for research purposes (Spicer *et al.*, 1993). As  $\text{NO}_2$  decays, HONO is formed. Small amounts of NO are also generated, as has been observed in laboratory studies (e.g., Spicer *et al.*, 1989). Delayed release of HONO was observed, suggesting that HONO was adsorbed in part on the surfaces and was subsequently released to the gas phase. For example, when the house was purged with outside air, the HONO levels dropped; however, on closing up the house again, the levels increased immediately to a few ppb. Degassing of HONO from the interior surfaces of automobiles has also been reported (Febo and Perrino, 1995). Interestingly, in measurements made in a commercial office building, indoor HONO concentrations were observed not to be enhanced under conditions of high indoor  $\text{O}_3$ , suggesting that dissolved HONO in a surface film of water is readily oxidized to nitrate by  $\text{O}_3$  (Weschler *et al.*, 1994).

The generation of NO in Fig. 15.5 was attributed by Spicer and co-workers to a reaction of gaseous  $\text{NO}_2$

with adsorbed HONO:



The same process was hypothesized to explain some time periods in a commercial office building when indoor NO actually exceeded outdoor NO (Weschler *et al.*, 1994).

As is the case in laboratory systems, equivalent amounts of  $\text{HNO}_3$  are not observed as might be expected from the stoichiometry of reaction (1), likely due to  $\text{HNO}_3$  remaining on the surface after formation and/or being taken up by surfaces. For example, Spicer *et al.* (1993) used atmospheric pressure ionization mass spectrometry (see Chapter 11.A.2), which could measure  $\text{HNO}_3$  with a sensitivity of 50 ppt, during the  $\text{NO}_2$  decay and HONO formation in the research home.  $\text{HNO}_3$  was only observed, and in small concentrations ( $< 1$  ppb), when an unvented space heater was operating, and it decayed rapidly in the absence of sources. Similarly, Salmon *et al.* (1990) measured  $\text{HNO}_3$  indoors and outdoors at five museums in the Los Angeles area and found that indoor concentrations were less than  $\sim 40\%$  of those outdoors, and typically about 10%. The accumulation of nitrate on indoor surfaces in a commercial building has been reported by Weschler and Shields (1996a) and attributed to the formation and uptake of  $\text{HNO}_3$  via reactions of  $\text{NO}_3$  (see later) and/or oxidation of nitrite (i.e., adsorbed HONO) in an aqueous surface film.

Subsequently, it was shown that HONO is also directly emitted by gas stoves (Pitts *et al.*, 1989). For example, Fig. 15.6 shows the concentrations of  $\text{NO}_2$  and HONO measured using DOAS when two top burners of a new, residential gas kitchen stove were turned on in a mobile home with both the central ventilation and air conditioning running.  $\text{NO}_2$  from the gas stove emissions reached almost 300 ppb, and HONO about

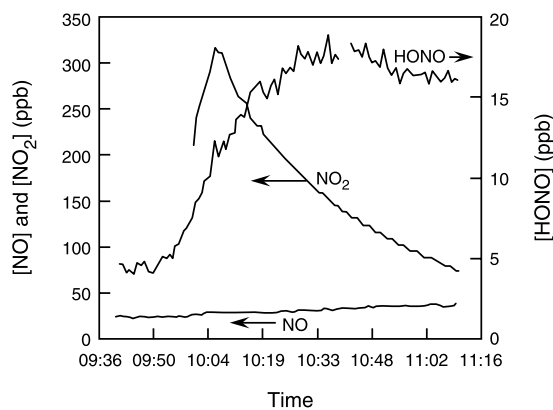


FIGURE 15.5 Concentrations of  $\text{NO}_2$ , HONO, and NO after injection of  $\text{NO}_2$  into a research house (adapted from Spicer *et al.*, 1993).

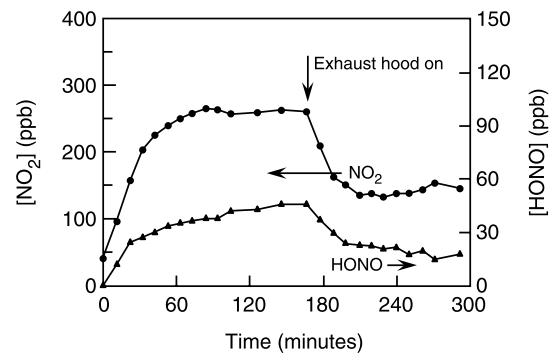


FIGURE 15.6  $\text{NO}_2$  and HONO measured in a mobile home with two burners of a kitchen stove on; the above-range exhaust was turned on at the time shown (adapted from Pitts *et al.*, 1989).

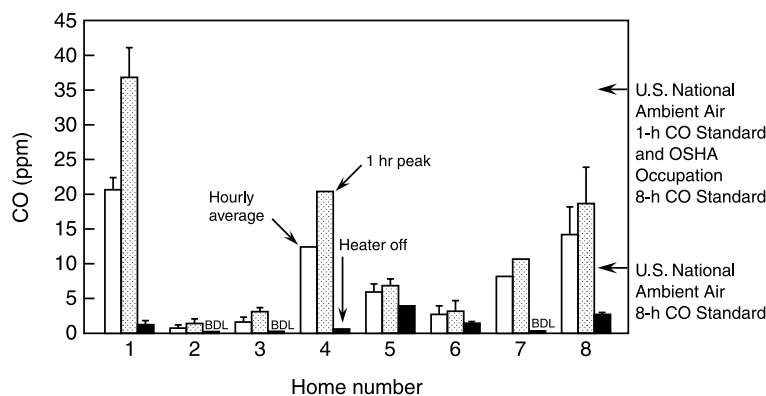


FIGURE 15.7 Measured concentrations of CO in eight homes using unvented kerosene heaters: dark shading, hourly average concentrations with heater off; no shading, hourly average concentrations with heater on; light shading, peak 1-h concentration (adapted from Mumford *et al.*, 1991).

45 ppb. When the exhaust hood (which was externally vented) above the range was turned on, the NO<sub>2</sub> and HONO decreased substantially. Similar production of HONO from kerosene and propane space heaters has been observed (e.g., see Pitts *et al.*, 1989; Brauer *et al.*, 1990; Febo and Perrino, 1991; and Vecera and Dasgupta, 1994). While the mechanism generating such substantial concentrations is not known, it may involve the recombination of OH with NO as the combustion gases cool.

In a house used for investigating indoor air pollution that had natural gas fueled appliances (a convective heater, a radiant heater, and a range with four burners), both the surface reaction of NO<sub>2</sub> and the direct combustion emissions contributed significantly to the measured indoor HONO. When an appliance was operational, the contribution of direct emissions was the more important source (Spicer *et al.*, 1993).

In short, the “dark reaction” of NO<sub>2</sub> with water on surfaces is ubiquitous and occurs not only in laboratory systems but also indoors. The combination of this heterogeneous reaction with combustion sources of HONO can produce significant concentrations of HONO indoors. As a result, there is a concern regarding the health impacts of nitrous acid, not only because it is an inhalable nitrite but also because it is likely the airborne acid present in the highest concentrations indoors.

### C. CO AND SO<sub>2</sub>

As for NO<sub>x</sub>, combustion sources such as gas stoves and kerosene heaters can be significant sources of indoor CO. Figure 15.7, for example, shows measured

CO concentrations in eight mobile homes with unvented kerosene heaters either off or on (Mumford *et al.*, 1991). Both the hourly average and peak 1-h concentrations are shown for the situation with the heater on, whereas only the 1-h average for the heater off is shown. Also shown are the United States 1- and 8-h standards and the Occupational Safety and Health Administration (OSHA) 8-h standard. In three of the homes, the average CO concentrations exceeded the 8-h standard and in one home, the 1-h ambient air and OSHA standard was exceeded.

The ratio of indoor to outdoor concentrations of CO in homes using gas stoves has been measured to be 1.2–3.8 (Wade *et al.*, 1975), with the highest ratios found close to the source. Similarly, higher CO levels indoors compared to outdoors have been reported for restaurants in Korea, with those using charcoal burners as well as gas giving much higher concentrations (Baek *et al.*, 1997). Figure 15.8, for example, shows the mean indoor-to-outdoor concentrations of CO and some other air pollutants measured in restaurants in Korea using either gas only or a combination of gas and

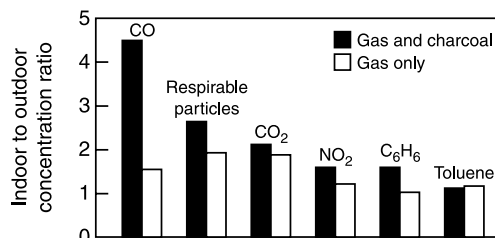


FIGURE 15.8 Median ratios of indoor-to-outdoor concentrations of CO and some other air pollutants measured in restaurants in Korea where either gas alone (□) or gas with charcoal (■) was used for cooking (adapted from Baek *et al.*, 1997).

charcoal. The use of charcoal increases this ratio to more than four; in one such restaurant, an indoor concentration of 90 ppm CO was measured.

In buildings where motor vehicle exhaust can be entrained from outdoors or attached parking garages, elevated indoor CO levels may also result (e.g., Hodgson *et al.*, 1991).

On the other hand, in homes and offices where there was no direct indoor source of CO, the indoor-to-outdoor ratio was about one, and sometimes less. For example, in Riyadh, Saudi Arabia, CO concentrations were measured indoors and outdoors; the indoor-to-outdoor ratio varied from 0 to 2, but was typically below one (Rowe *et al.*, 1989).

There have been a number of measurements of CO in the "indoor environment" of automobiles. Given that cars are major CO sources in urban areas, one might expect higher concentrations of CO during commutes and this is indeed the case. Typical CO concentrations of  $\sim 9$ – $56$  ppm have been measured inside automobiles during commutes in major urban areas (Flachsbart *et al.*, 1987; Koushki *et al.*, 1992; Ott *et al.*, 1994; Dor *et al.*, 1995; Fernandez-Bremauntz and Ashmore, 1995). This can be compared to peak outdoor levels of  $\sim 10$  ppm in highly polluted urban areas (see Chapter 11.A.4c). Thus, a significant enhancement of CO inside automobiles during commutes is common. For example, Chan *et al.* (1991b) report a ratio of the in-vehicle CO concentration to that outdoors of  $\sim 4.5$  in Raleigh, North Carolina.

As is the case for CO, SO<sub>2</sub> levels indoors and outdoors tend to be similar if there are no combustion sources indoors. For example, Hisham and Grosjean (1991b) report that the ratio of indoor-to-outdoor SO<sub>2</sub> concentrations averaged 0.89 for three museums in southern California, with a range from 0.36 to 1.92. On the other hand, quite high SO<sub>2</sub> concentrations can result when there are indoor combustion sources. For example, in China, where unvented stoves using coal are used extensively for cooking and heating, average SO<sub>2</sub> levels indoors are typically  $250 \mu\text{g m}^{-3}$  (96 ppb) during the summer and  $750 \mu\text{g m}^{-3}$  (287 ppb) during the winter, compared to average annual outdoor concentrations of  $72$ – $94 \mu\text{g m}^{-3}$  (28–36 ppb) (Florig, 1997; Ando *et al.*, 1996).

#### D. VOLATILE ORGANIC COMPOUNDS

Volatile organic compounds (VOC) are ubiquitous components not only of ambient air but also of indoor

air environments, including offices, commercial and retail buildings, and homes (Shah and Singh, 1988). There are three sources/categories for VOC: (1) entrainment of air from outside the building, (2) emissions from building materials, and (3) human activities inside buildings.

As might be expected given the nature of the sources, a very large variety of organic compounds have been identified and measured indoors (e.g., Brown *et al.*, 1994; Crump, 1995; Kostianen, 1995). These number in the hundreds of different compounds, with the particular species and their concentrations depending on the particular sources present as well as the air exchange rates. Table 15.2 summarizes some of the types of organics that have been measured in indoor air and typical sources (Tichenor and Mason, 1988; Crump, 1995). Because of the VOC sources present indoors, the indoor-to-outdoor concentration ratios are quite large for many compounds.

For example, Table 15.3 shows some typical ratios of indoor-to-outdoor concentrations for specific compounds found in each of the classes shown in Table 15.2, which are frequently present indoors (Brown *et al.*, 1994). These data are based on a review of the literature and include data from a number of different countries. The ratio is for all but one compound substantially greater than one. Also shown in Table 15.3 is a typical range of concentrations expressed as the overall weighted average of the geometric mean, where the weighting was done using the number of available measurements. Some of the compounds associated with the three sources—entrainment from outdoors, emissions from building materials, and anthropogenic activities—are now briefly reviewed.

*Entrainment of air from outdoor sources.* Entrainment of outdoor air through ventilation systems brings with it the species found in ambient air, which have been discussed throughout this book. Some of them, such as HNO<sub>3</sub> and to a lesser extent O<sub>3</sub>, can be removed on surfaces such as those in air conditioning systems, and hence the indoor concentrations tend to be lower than those outdoors. Others such as NO tend to have similar concentrations indoors and outdoors if there are no significant combustion sources indoors (e.g., Weschler *et al.*, 1994). In the case of hydrocarbons, the concentrations of compounds that do not have significant indoor sources tend to be about the same as the outdoor concentrations. For example, Lewis and Zweidinger (1992) measured VOC in 10 homes in winter and showed that the concentrations of ethene, benzene, 2-methylpentane, methylcyclopentane, 2,2,4-trimethylpentane, and 2,3-dimethylbutane indoors were within experimental error of those outdoors.

TABLE 15.2 Some VOCs Measured Indoors and Their Sources<sup>a</sup>

Class of Compounds	Species	Typical Sources
Hydrocarbons	Aliphatic hydrocarbons	Paints, adhesives, gasoline, combustion products, floor waxes
	Aromatic hydrocarbons (toluene, xylenes, ethyl benzene, trimethylbenzenes, styrene, benzene)	Insulation, textiles, disinfectants, plastics, paints, smoking
	Terpenes (limonene, $\alpha$ -pinene)	Scented deodorizers, polishes, fabrics, fabric softeners, cigarettes, food, beverages
	PAHs	Combustion products (smoking, wood burning, kerosene heaters)
Oxygenated organics	Acrylic acid esters, epichlorohydrin	Monomers may escape from polymers
	Alcohols	Aerosols, window cleaners, paints, paint thinning, cosmetics, adhesives
	Ketones	Lacquers, varnishes, polish removers, adhesives
	Ethers	Resins, paints, varnishes, lacquers, dyes, soaps, cosmetics
	Esters	Plastics, resins, plasticizers, lacquer solvents, flavors, perfumes
	Ethylene oxide	Sterilizers (hospitals)
Other organics	Toluene diisocyanate	Polyurethane foam aerosols
	Phthalic acid anhydride	Epoxy resins
	Sodium dodecyl sulfate	Carpet shampoo
Chlorinated organics	Benzyl chloride	Vinyl tiles
	Tetrachloroethylene	Dry-cleaned clothes
	Chloroform	Chlorinated water
	1,1,1-Trichloroethane	Dry-cleaned clothes, aerosol sprays, fabric protectors
	Carbon tetrachloride	Industrial-strength cleaners
	<i>p</i> -Dichlorobenzene	Moth crystals, room deodorants

<sup>a</sup> Adapted from Crump (1995) and Tichenor and Mason (1988).

There are, however, some specific outdoor sources that can lead to higher concentrations of certain VOCs indoors than in the general outdoor air environment. For example, gases generated in landfills or from petroleum contamination can migrate through the soil and groundwater to adjacent buildings and homes to give larger indoor concentrations, particularly in basements and crawl spaces, than otherwise expected (e.g., Moseley and Meyer, 1992; Hodgson *et al.*, 1992; Fischer *et al.*, 1996). In one such case, the total hydrocarbon concentration was measured to be 120 ppm in a crawl space beneath the floor of a school where petroleum contamination was present from adjacent sources, compared to <80 ppb outdoors (Moseley and Meyer, 1992). Although concentrations in various rooms

were lower, they were still elevated compared to outdoors, ranging from 0.13 to 3.4 ppm.

The use of pesticides *outside* buildings can also lead to enhanced concentrations of these compounds indoors. For example, Anderson and Hites (1988) measured the concentrations of chlorinated pesticides indoors and found elevated levels inside, e.g., a factor of 7 times higher for  $\gamma$ -chlordane compared to outdoor levels. One home that had the highest indoor concentrations had been treated with chlordane about a decade earlier, presumably by subsurface injection from which the pesticide migrated into the house through cracks in the basement walls. Enhanced levels of chlorpyrifos were observed indoors in homes where soil surrounding the home had been treated on a regular



TABLE 15.3 Ratio of Indoor-to-Outdoor Concentrations for Some VOCs and Typical Ranges of Concentrations<sup>a</sup>

Compound	Indoor / outdoor ratio (range of concentration ( $\mu\text{g m}^{-3}$ ))	Compound	Indoor / outdoor ratio (range of concentration ( $\mu\text{g m}^{-3}$ ))
<i>n</i> -Alkane		Alcohols	
<i>n</i> -Pentane	3	2-Propanol	>73
<i>n</i> -Hexane	9	<i>n</i> -Butanol	5 (<1)
<i>n</i> -Heptane	4 (1-5)	Aldehydes	
<i>n</i> -Octane	7 (1-5)	Acetaldehyde	5
<i>n</i> -Nonane	14 (1-5)	Butanal	2 (1-5)
<i>n</i> -Decane	19 (5-10)	Hexanal	>5
<i>n</i> -Undecane	20 (1-5)	Nonanal	5 (5-10)
<i>n</i> -Dodecane	20 (1-5)	Ketones	
<i>n</i> -Tridecane	>6	Acetone	12 (20-50)
<i>n</i> -Tetradecane	16 (1-5)	Methyl ethyl ketone	4 (1-5)
<i>n</i> -Pentadecane	>5 (1-5)	Esters	
Branched cycloalkanes		Ethyl acetate	15 (5-10)
2-Methylpentane	2	Aromatic hydrocarbons	
2-Methylhexane	2	Styrene	10 (1-5)
3-Methylhexane	3	Benzene	3 (5-10)
Cyclohexane	4 (1-5)	Toluene	6 (20-50)
Halogenated compounds		Ethylbenzene	6 (5-10)
Trichlorofluoromethane	10	<i>m</i> - and <i>p</i> -xylene	6 (10-20)
1,2-Dichloroethane	12 (<1)	<i>o</i> -Xylene	6 (5-10)
Dichloromethane	6 (10-20)	<i>n</i> -Propylbenzene	4
Chloroform	5 (1-5)	1,3,5-Trimethylbenzene	4 (1-5)
Carbon tetrachloride	2 (1-5)	1,2,4-Trimethylbenzene	15 (5-10)
1,1,1-Trichloroethane	9 (20-50)	(1-Methylethenyl)benzene	5
1,1-Dichloroethene	13 (1-5)	Naphthalene	4 (<1)
Trichloroethylene	6 (1-5)	Terpenes	
Tetrachloroethylene	5 (5-10)	Camphene	20 (10-20)
<i>p</i> -Dichlorobenzene	5 (5-10)	$\alpha$ -Pinene	23 (1-5)
<i>m</i> -Dichlorobenzene	0.4 (<1)	Limonene	80 (20-50)
		Total VOC	7

<sup>a</sup> Adapted from Brown *et al.* (1994) and references therein.

basis. Similarly, aldrin and dieldrin (used as termiticides in the United States from the 1950s to the 1970s) were shown to be present indoors in a home in which the surrounding soil and/or concrete blocks used for the basement walls had been treated during construction; as might be expected given the source, the highest levels were in the basement (Wallace *et al.*, 1996).

Another source of VOC is motor vehicle emissions, which can be drawn into buildings from outdoors or parking garages (e.g., Perry and Gee, 1994; Daisey *et al.*, 1994). For example, motor vehicles were major sources (responsible for  $\geq 75\%$ ) of 12 of 39 individual compounds measured in a dozen buildings by Daisey *et al.* (1994). Of the 12 compounds, 5 were alkanes and 7 were aromatics. Similarly, Baek *et al.* (1997) report that

vehicle emissions are important VOC sources indoors in Korea during the summer in homes and offices, as has been reported in the United States (e.g., Hodgson *et al.*, 1991; Daisey *et al.*, 1994).

**Building materials.** Emissions associated with building materials are major contributors to indoor levels of VOC. Table 15.4 summarizes some individual VOCs that have been associated with some building materials (Tichenor and Mason, 1988; Crump, 1995). The overall weighted average geometric mean concentrations of some individual organics measured indoors in buildings that were more than 3 months old are shown in Table 15.5 (Brown *et al.*, 1994). New buildings often have higher concentrations of certain compounds compared to older buildings. For example, enhanced levels of

TABLE 15.4 Some VOCs from Building Materials<sup>a</sup>

Source	Compounds
Carpet adhesive	Toluene
Floor adhesive (water based)	Nonane, decane, undecane, dimethyloctane, 2-methylnonane, dimethylbenzene
Particleboard	Formaldehyde, acetone, hexanal, propanol, butanone, benzaldehyde, benzene
Moth crystals	<i>p</i> -Dichlorobenzene
Floor wax	Nonane, decane, undecane, dimethyloctane, trimethylcyclohexane, ethylmethylbenzene
Wood stain	Nonane, decane, undecane, methyloctane, dimethylnonane, trimethylbenzene
Latex paint	2-Propanol, butanone, ethylbenzene, propylbenzene, 1,1'-oxybis[butane], butylpropionate, toluene, formic and acetic acids
Water-based acrylic wall paint	1,2-Propanediol, isomers of 2,4,4-trimethyl-1,3-pentanediol monoisobutyrate
Furniture polish	Trimethylpentane, dimethylhexane, trimethylhexane, trimethylheptane, ethylbenzene, limonene
Polyurethane floor finish	Nonane, decane, undecane, butanone, ethylbenzene, dimethylbenzene
Room freshener	Nonane, decane, undecane, ethylheptane, limonene, substituted aromatics (fragrances)
Particleboard	Formaldehyde, acetone, hexanol
Vinyl flooring	Alkyl aromatics, dodecane, 2,2,4-trimethyl-1,3-pentanediol diisobutylate, 2-ethyl-1-hexanol, phenol, cresol, ethyl hexyl acetate, ammonia
Floor varnish	Butyl acetate, <i>N</i> -methylpyrrolidone
Laminated cork floor tile	Phenol
Carpets	4-Phenylcyclohexene, styrene, 4-ethenylcyclohexene, 2-ethyl-1-hexanol, nonanol, heptanol
Silicone caulk	Methyl ethyl ketone, butyl propionate, 2-butoxyethanol, butanol, benzene, toluene
Paint	Dibutyl phthalate
Acrylic sealant	Hexane, dimethyloctanols
Creosote-impregnated timber	Naphthalene, methylnaphthalenes

<sup>a</sup> Adapted from Tichenor and Mason (1988), Crump (1995), Reiss *et al.* (1995b), and Wolkoff (1998).

*n*-dodecane, *n*-decane, and *n*-undecane, the xylenes, and 2-propanol have been measured in new buildings, and the total VOC concentration is generally larger (by factors of 4–23) compared to established buildings (Brown *et al.*, 1994).

Kostiainen (1995) identified more than 200 individual VOCs indoors in 26 houses. In addition, they compared the VOC concentrations in normal houses to those where complaints of odors or illness had been registered. A number of different VOCs were present at increased concentrations in the houses with complaints compared to the normal houses; these included a variety of aromatic hydrocarbons, methylcyclohexane, *n*-propylcyclohexane, terpenes, and chlorinated compounds such as 1,1,1-trichloroethane and tetrachlorethene.

Carpets are a major source of VOCs in homes. For example, Sollinger *et al.* (1993, 1994) have identified 99

different VOCs emitted from a group of 10 carpet samples, and Schaeffer *et al.* (1996) identified more than 100 different VOCs emitted from the carpet cushion alone. Emissions come not only from the carpet fibers but also from the backing materials and the adhesives used to bind the carpet to the backing. As a result, the individual compounds emitted by carpets can vary substantially, depending on the carpet construction. For example, Table 15.6 shows some of the compounds emitted by three different types of carpets: (1) with a styrene–butadiene rubber (SBR) latex adhesive to bind the primary and secondary backings; (2) with a polyvinyl chloride (PVC) secondary backing and made in the form of tiles; and (3) with a polyurethane foam secondary backing. With the SBR adhesive, the major VOCs emitted are 4-phenylcyclohexene (responsible for the “new carpet” smell) and styrene. With the PVC backing, the major compounds are vinyl ac-

TABLE 15.5 Overall Weighted Average Geometric Mean Concentrations of Some Individual VOCs Indoors<sup>a</sup>

Compound	Type of building	Number of measurements	Concentration ( $\mu\text{g m}^{-3}$ )
<b>Hydrocarbons</b>			
<i>n</i> -Hexane	Dwelling	656	5
	Office	26	12
<i>n</i> -Nonane	Dwelling	592	5
<i>n</i> -Decane	Dwelling	1085	5
Camphene	Mobile home	44	14
Limonene	Dwelling	584	21
Benzene	Dwelling	2171	8
Toluene	Dwelling	792	37
Ethylbenzene	Dwelling	1867	5
<i>o</i> -Xylene	Dwelling	1518	6
<i>m</i> - and <i>p</i> -xylene	Dwelling	1587	18
1,2,4-Trimethylbenzene	Dwelling	619	6
1,2,3-Trimethylbenzene	Office	152	9
<i>m</i> -Methylethylbenzene	Office	168	8
<b>Oxygenated organics</b>			
Acetone	Dwelling	86	32
Methanol	School	11	29
Ethanol	Dwelling	39	120
Acetic acid	School	5	12
Butyric acid	School	5	25
Methyl ethyl ketone	Dwelling	316	4–21
Diethyl ketone	School	12	6
Phenol	School	5	9
Nonanal	Dwelling	15	7
Ethyl acetate	Dwelling	302	8
	School	12	10
<b>Chlorinated organics</b>			
Dichloromethane	Dwelling	101	17
Chloroform	Office	20	10
1,2-Dichloroethene	Dwelling	35	11
1,1,1-Trichloroethane	Dwelling	1580	24
Tetrachloroethene	Dwelling	1919	7
<i>p</i> -Dichlorobenzene	Dwelling	1881	8

<sup>a</sup> Adapted from Brown *et al.* (1994).

etate (used as a copolymer in the production of PVC), 1,2-propanediol, and 2,2,4-trimethylpentane, whereas the polyurethane backing emitted primarily 2,6-di-*tert*-butyl-4-methylphenol (BHT), a compound often used as an antioxidant. Many of the compounds emitted are known to be used in the manufacturing processes (e.g.,  $\epsilon$ -caprolactam is used in Nylon-6 production) and/or are common solvents. For example, such species as 1,2-propanediol, 2,2,4-trimethylpentane, 1,1,1-trichloroethane, toluene, 1-butanol, and the dipropylene glycol methyl ethers may be used during the production of various carpet components.

Emissions of VOC from carpets tend to decrease with time and increase with temperature. For example,

Fig. 15.9 shows the concentrations in a test chamber of 4-phenylcyclohexene, styrene, and 4-ethenylcyclohexene, another compound associated with SBR adhesives (Weschler *et al.*, 1992b), from one carpet as a function of time (Hodgson *et al.*, 1993). While styrene and 4-ethenylcyclohexene decrease rapidly with time, the decay of 4-phenylcyclohexene is much slower. In a house used for field studies, the concentration of 4-phenylcyclohexene remained above 2 ppb almost two months after installation, well above the odor threshold of 0.5 ppb.

Figure 15.10 shows the temperature dependence of the concentrations of two VOCs emitted by carpets, styrene and benzothiazole, into a test chamber (Sol-

TABLE 15.6 Some Organics Emitted from Carpets<sup>a</sup>

Compound	SBR <sup>b</sup> latex backing adhesive	PVC <sup>b</sup> backing	Polyurethane backing
4-Phenylcyclohexene	D	—	—
Styrene	D	—	—
Vinyl acetate	—	D	—
1,2-Propanediol	—	D	—
2,2,4-Trimethylpentane	—	D	—
Formaldehyde	—	+	—
2-Ethyl-1-hexanol	—	+	—
2,6-Di- <i>tert</i> -butyl-4-methylphenol (BHT)	—	—	D
1-Butanol	—	—	+
Hexamethylcyclotrisiloxane	—	—	+
Dipropylene glycol methyl ethers	—	—	+
2-Methyl-1-propene	+	—	+
Acetaldehyde	—	+	—
Acetone	+	—	+
1- and 2-propanol	—	—	+
Trimethylsilanol	—	—	+
1,1,1-Trichloroethane	—	—	+
Acetic acid	—	+	—
Alkane HCs	—	+	—
2,2,5-Trimethylhexane	—	—	+
4-Ethenylcyclohexane	+	—	—
$\epsilon$ -Caprolactam	—	—	+

<sup>a</sup> Adapted from Hodgson *et al.* (1993); D = dominant emission, + = observed, — = not observed.

<sup>b</sup> SBR = styrene-butadiene rubber latex adhesive; PVC = polyvinyl chloride.

linger *et al.*, 1994). Both increase with temperature, but styrene at a much slower rate. Given the temperature control in many homes, the change in emissions over most temperature ranges encountered in dwellings may

not be large. However, in homes without temperature control or in automobiles, for example, the temperature dependence could be significant.

Emission rates from other building materials such as flooring, paints, varnishes, and sealants also tend to increase, not surprisingly, with temperature (e.g.,

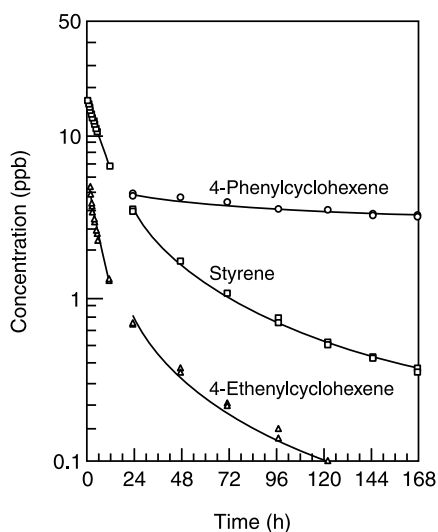


FIGURE 15.9 Concentrations of three VOCs emitted from a carpet sample made using SBR adhesives as a function of time in a test chamber (adapted from Hodgson *et al.*, 1993).

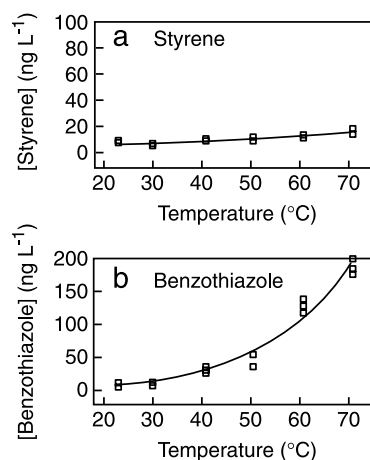


FIGURE 15.10 Temperature dependence of styrene and benzo-thiazole emitted from carpets into a test chamber (adapted from Sollinger *et al.*, 1994).

TABLE 15.7 Effect of Temperature, Relative Humidity, and Replacement of Air by N<sub>2</sub> on VOC Emissions from Some Common Building Materials<sup>a,b</sup>

Building product	Temperature (°C)		RH (%)	Air replaced by N <sub>2</sub>
	35	60		
Carpet				
2-Ethylhexanol	+	+	+	+
4-Phenylcyclohexene	+	+	NC	NC
PVC flooring				
2-Ethylhexanol	NC	+	NC	+
Phenol	NC	+	NC	+
Sealant				
Hexane	+	+	NC	NC
Dimethyloctanols	+	+	+	+
Varnish				
Butyl acetate	+	+	NC	NC
N-Methylpyrrolidone	+	+	NC	NC
Wall Paint				
1,2-Propanediol	NC	+	+	NC
Texanol <sup>c</sup>	NC	+	NC	NC

<sup>a</sup> Adapted from Wolkoff (1998).

<sup>b</sup> A “+” sign means an increase of 20% or more in concentration. NC means less than 20% change.

<sup>c</sup> Texanol = isomers of 2,4,4-trimethyl-1,3-pentanediol monoisobutyrate.

Wolkoff, 1998; Haghghat and de Bellis, 1998). Table 15.7, for example, shows the effects of temperature and relative humidity on the emissions of particular compounds associated with carpet, PVC flooring, sealants, varnish, and wall paint (Wolkoff, 1998). Interestingly, exposure of these samples to N<sub>2</sub> rather than air also increased the emissions in some cases. However, using increased temperatures to “bake-out” buildings and hence lower the concentrations of indoor VOCs does not appear to be particularly effective. For example, Bayer (1991) reports that the total VOC concentrations from particleboard are about the same after as before a 5-day bake-out at 88°C. Similarly, significant levels of HCHO have been observed in a mobile home even after 20 years of use in a hot ambient air environment (Pitts *et al.*, 1989; see later).

The dependence of VOC emissions from building materials on relative humidity is more complex, with some emissions increasing with relative humidity, but others not. For example, Sollinger *et al.* (1994) report that the VOC emissions from carpets did not change with relative humidity over the range from 0 to 45% RH. On the other hand, the emissions of formic and acetic acids from latex paints have been reported to

increase dramatically with relative humidity; for example, for one paint sample the emission rate for acetic acid almost tripled when the relative humidity was changed from 4–5% to 5–23% (Reiss *et al.*, 1995b).

A number of different aldehydes have been measured indoors (e.g., see Crump and Gardiner, 1989; Lewis and Zweidinger, 1992; Zhang *et al.*, 1994a; Daisey *et al.*, 1994; and Reiss *et al.*, 1995a), some of which are directly emitted and some of which are formed by chemical reactions indoors of VOCs such as styrene (see Section F). Of these, there is an enormous amount of evidence for direct emissions of HCHO from building materials. Interest in formaldehyde emissions and levels in homes and other buildings stems from its well-known health effects, which include possible human carcinogenicity and eye, skin, and respiratory tract irritation (Feinman, 1988). Formaldehyde is emitted from urea–formaldehyde foam insulation as well as from resins used in reconstituted wood products such as particleboard and plywood (Meyer and Reinhardt, 1986); urea–formaldehyde resins comprise about 6–8% of the weight of particleboard and 8–10% of medium-density fiberboard (Meyer and Hermanns, 1986). Other sources include permanent press fabrics (such as draperies and clothing), floor finishing materials, furniture, wallpaper, latex paint, varnishes, some cosmetics such as fingernail hardener and nail polish, and paper products (Kelly, 1996; Howard *et al.*, 1998).

Many measurements of HCHO have been made in indoor air environments. In conventional homes, average concentrations are typically about 10–50 ppb (e.g., Stock, 1987; Zhang *et al.*, 1994a; Reiss *et al.*, 1995a). Sexton *et al.* (1989) measured concentrations of HCHO in 470 mobile homes in California and found geometric mean concentrations of 60–90 ppb, although maximum values of over 300 ppb were recorded in some cases. In a similar study in Wisconsin, levels up to 2.8 ppm were measured (Hanrahan *et al.*, 1985). Higher levels are typically found in mobile homes because of the reconstituted wood products (e.g., particleboard and plywood) used in their construction. Interestingly, HCHO does not appear to be a significant product of natural gas combustion, as levels in dwellings with and without gas stoves turned on are not significantly different (e.g., Pitts *et al.*, 1989; Zhang *et al.*, 1994a).

Temperature is again an important determinant of HCHO levels. Figure 15.11, for example, shows the concentrations of HCHO as well as of formic acid and methanol measured using FTIR in a research mobile home as function of time as the temperature increased. At 70°F, the average HCHO concentration was 27 ppb but increased to 105 ppb at 100°F (Pitts *et al.*, 1989).

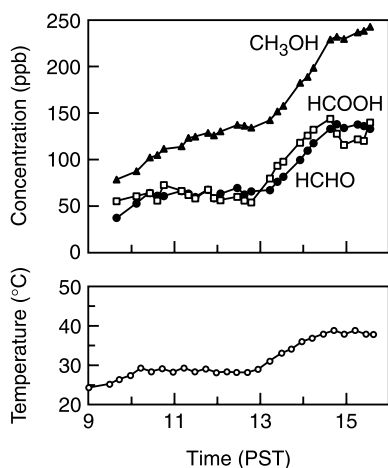


FIGURE 15.11 Concentrations of HCHO, HCOOH, and  $\text{CH}_3\text{OH}$  measured using long-path FTIR in a mobile home as a function of temperature. Although the gas stove burners were on, these were shown in separate experiments not to be the source of these organics (adapted from Pitts *et al.*, 1989.)

HCOOH has also been observed from outgassing in a mobile home trailer by chemical ionization mass spectrometry during measurements of  $\text{HNO}_3$  (Huey *et al.*, 1998) and at lower concentrations (mean of 10 ppb) in conventional homes (Reiss *et al.*, 1995a).

Figure 15.12 summarizes the ratio of indoor-to-outdoor concentrations of HCHO and higher aldehydes as well as formic and acetic acids measured in some conventional homes. Concentrations of all of these compounds, except possibly propionaldehyde, are significantly higher indoors, suggesting that not only

HCHO but higher aldehydes and ketones as well as acids have significant indoor sources (Lewis and Zweidinger, 1992; Zhang *et al.*, 1994a, 1994b; Zhang and Liou, 1994; Reiss *et al.*, 1995a). As discussed later, reactions of hydrocarbons with ozone indoors is a potential source, in addition to direct emissions.

It should be noted that while building materials are sources of a variety of VOCs, they can also adsorb organics as well (e.g., Van Loy *et al.*, 1997). As a result, building surfaces and contents may act as reservoirs of organics, slowly releasing compounds over a period of time.

*Anthropogenic activities.* There are many sources of VOCs associated with human activities in buildings. For example, mixtures of  $\text{C}_{10}$  and  $\text{C}_{11}$  isoparaffinic hydrocarbons, which are characteristic of liquid process copiers and plotters, have been identified in office buildings in which these instruments were in use (Hodgson *et al.*, 1991). Emissions of a number of hydrocarbons and aldehydes and ketones have been observed during operation of dry-process copiers; these include significant emissions of ethylbenzene, *o*-, *m*-, and *p*-xylenes, styrene, 2-ethyl-1-hexanol, acetone, *n*-nonanal, and benzaldehyde (Leovic *et al.*, 1996). Enhanced levels of acetaldehyde in an office building in Brazil were attributed to the oxidation of ethanol used as a cleaning agent (Brickus *et al.*, 1998), although levels outdoors were also enhanced due to the use of ethanol as a fuel (see Chapter 16.D.4). Pyrocatechol has been measured in an occupational environment where meteorological charts are mapped on paper impregnated with this compound (Ekinja *et al.*, 1995), and

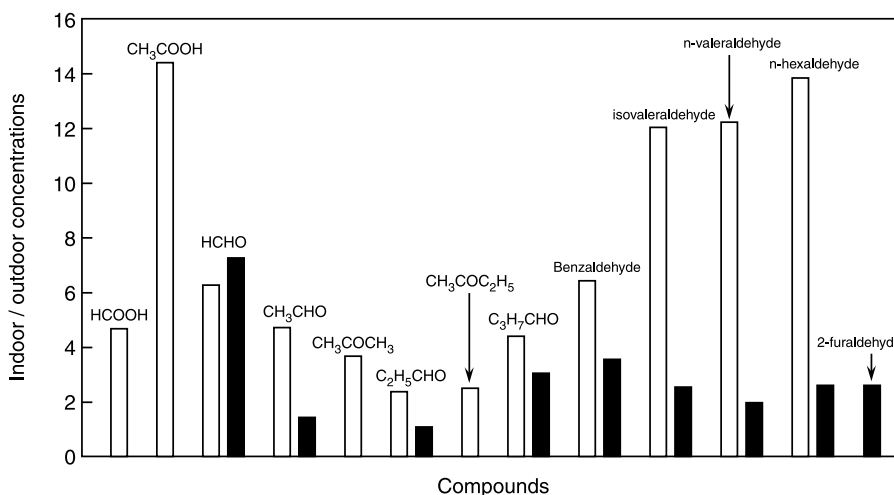


FIGURE 15.12 Ratio of indoor-to-outdoor concentrations of formic and acetic acids and some carbonyl compounds in some homes: unshaded, data from Reiss *et al.*, 1995a, for summer data; shaded, data from Zhang *et al.*, 1994a.

*p*-dichlorobenzene is observed when mothballs containing this compound are in use (e.g., Tichenor *et al.*, 1990; Chang and Krebs, 1992).

Elevated concentrations of the *n*-C<sub>13</sub> to *n*-C<sub>18</sub> alkanes and branched-chain and cyclic analogs were measured in a building having a history of air quality complaints; the source was found to be volatilization from hydraulic fluids used in the building elevators (Weschler *et al.*, 1990).

Enhanced levels of chlorinated compounds have been observed indoors due to human activity as well. For example, increased levels of perchloroethylene have been observed from unvented dry-cleaning units (e.g., Moschandreas and O'Dea, 1995) and volatilization of chlorinated organics such as chloroform from treated tap water can occur (e.g., McKone, 1987). Other sources include the use of household products. For example, chloroform emissions have been observed from washing machines when bleach containing hypochlorite was used (Shepherd *et al.*, 1996). It is interesting that emissions of organics associated with the use of washing machines are decreased when the machine is operated with clothes inside (Howard and Corsi, 1998).

Of course, activities such as smoking result in enhanced levels not only of nicotine (e.g., Thompson *et al.*, 1989) but also of a variety of other gases associated with cigarette smoke (e.g., California Environmental Protection Agency, 1997; Nelson *et al.*, 1998). For example, using 3-ethenylpyridine as a marker for cigarette smoke, Heavner *et al.* (1992) estimated that 0.2–39% of the benzene and 2–49% of the styrene measured in the homes of smokers were from cigarette smoke.

Humans emit a variety of VOCs such as pentane and isoprene (e.g., Gelmont *et al.*, 1981; Mendis *et al.*, 1994; Phillips *et al.*, 1994; Jones *et al.*, 1995; Foster *et al.*, 1996). In addition, emissions from personal care products have been observed. Decamethylcyclopentasiloxane (D5), a cyclic dimethylsiloxane with five Si–O units in the ring, and the smaller D4 analog, octamethylcyclotetrasiloxane, are used in such products as underarm deodorant and antiperspirants at concentrations up to 40–60% by weight (Shields and Weschler, 1992; Shields *et al.*, 1996). Increased concentrations of D5 have been measured in offices and are correlated to human activity, as expected if personal care products were the major source (Shields and Weschler, 1992). In some cases, increased concentrations attributable to emissions from silicone-based caulking materials were also observed (Shields *et al.*, 1996).

The use of pesticides indoors can lead to very large concentrations not only of the pesticide but of the additional VOCs used as a matrix for the pesticide, which represent most (>95%) of the mass of the material as purchased. For example, Bukowski and

Meyer (1995) predict that VOC concentrations immediately after the application of a fogger could reach levels of more than 300 mg m<sup>-3</sup>!

However, lower levels of pesticides themselves are common after use inside homes. For example, Lewis *et al.* (1988) reported the presence of 24 pesticides in homes, ranging in concentration from 0.002 to 15 μg m<sup>-3</sup> (the latter for chlorpyrifos), while outdoor levels measured simultaneously were much lower, from <0.001 to 0.4 μg m<sup>-3</sup>. The compounds present indoors at the highest concentrations were those used recently at the home. Similarly, Whitmore *et al.* (1994) reported 22 pesticides in homes in Jacksonville, Florida, and Springfield, Massachusetts, at indoor levels up to 0.5 μg m<sup>-3</sup> indoors, but only 0.04 μg m<sup>-3</sup> outdoors. The pesticides not only were present as gases but also adsorbed to dust particles in the home, particularly for the less volatile compounds. Indeed, higher concentrations of some pesticides have been found in dust than in air (e.g., Roinestad *et al.*, 1993).

There are a few data that suggest that pesticides can undergo reactions indoors. For example, Wallace *et al.* (1996) observed that the aldrin levels inside a home decreased with time, whereas those of dieldrin did not. Dieldrin had been applied with aldrin but is also an oxidation product of aldrin. One of the reasons for the lack of change in dieldrin may be that it was being formed as the aldrin decayed; however, this could not be differentiated from the effects of a lower vapor pressure of dieldrin, which could lead to lower overall removal rates. In the same study, pentachloroanisole was also measured inside the home and attributed to formation by degradation of pentachlorophenol, which is used as a wood preservative and termiticide.

Not surprisingly, some indoor organics are readily taken up on building surfaces, such as carpets and wallboard, and are subsequently released into the room. For example, Chang *et al.* (1998) showed that some of the alcohols found in indoor air environments are taken up by carpets and gypsum board and could be desorbed back into the gas phase later. However, revolatilization was observed to be slow, and Chang and co-workers estimated that it would take more than a year to remove the adsorbed organics. Similarly, Van Loy *et al.* (1998) showed that nicotine from environmental tobacco smoke can be readily adsorbed and then desorbed and that surfaces can hold significant amounts of nicotine. As a result of this reversible adsorption–desorption process, measurable levels of organics can be maintained indoors after the initial exposure by slow degassing from surfaces.

Not surprisingly, the concentrations of VOCs from automobile exhaust are higher in the “indoor environment” of automobiles during commutes. For example,

Duffy and Nelson (1997) report during commutes in Sydney, Australia, that the benzene concentrations inside vehicles were 10–25 times those in ambient air and that the concentrations of 1,3-butadiene were more than 55–115 times greater. The source appeared to be primarily from the exhausts of surrounding vehicles. Similar enhancements of benzene and other VOCs such as toluene, ethylbenzene, and the xylenes in automobiles and buses have been reported in many countries, including Korea (Jo and Choi, 1996), Taiwan (Chan *et al.*, 1993), and the United States (e.g., Chan *et al.*, 1991a,b; Lawryk and Weisel, 1996).

## E. OZONE

Because O<sub>3</sub> decomposes on surfaces, indoor levels are usually lower than those outdoors due to the decomposition that occurs as the air passes through air conditioning systems and impacts building surfaces (e.g., Reiss *et al.*, 1994). The measured ratio of indoor-to-outdoor concentrations of ozone vary from 0.1 to 1, but are typically around 0.3–0.5 (e.g., Druzik *et al.*, 1990; Hisham and Grosjean, 1991b; Liu *et al.*, 1993; Weschler *et al.*, 1989, 1994; Gold *et al.*, 1996; Jakobi and Fabian, 1997; Avol *et al.*, 1998; Drakou *et al.*, 1998; Romieu *et al.*, 1998). Buildings with low air exchange with outside air tend to have lower ratios, ~0.1–0.3 (Druzik *et al.*, 1990; Weschler *et al.*, 1994; Romieu *et al.*, 1998). For example, Gold *et al.* (1996) estimate that at outdoor ozone concentrations of 170 ppb in Mexico City, the indoor-to-outdoor ratio of O<sub>3</sub> at a school was  $0.71 \pm 0.03$  with the windows and doors open, which maximized the exchange with outside air,  $0.18 \pm 0.02$  with the windows and doors closed and the air cleaner off, and  $0.15 \pm 0.02$  with the windows and doors closed and the air cleaner on.

There are some additional sources of O<sub>3</sub> indoors. These include dry-process photocopying machines, laser printers, and electrostatic precipitators (e.g., Leovic *et al.*, 1996; Wolkoff, 1999). Indeed, it is not unusual to detect O<sub>3</sub> by its odor during operation of some copy machines and laser printers.

In the “indoor environment” in cars, ozone levels tend to be significantly less than in the surrounding area. For example, Chan *et al.* (1991b) report that in-vehicle O<sub>3</sub> concentrations during commutes in Raleigh, North Carolina, were only about 20% of those measured in the local area at a fixed station. There are several contributing factors to these low concentrations. One is that NO concentrations are higher near roadways, so that O<sub>3</sub> is titrated to NO<sub>2</sub> by its rapid reaction with NO. A second is that O<sub>3</sub> can decompose

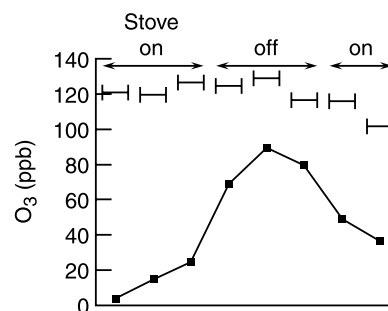


FIGURE 15.13 Measured concentrations of outdoor O<sub>3</sub> and indoor O<sub>3</sub> with and without a gas stove on: horizontal bars, outdoor O<sub>3</sub>; solid squares, indoor O<sub>3</sub> (adapted from Zhang and Liou, 1994).

on the surfaces of the automobile air conditioning system.

A similar titration effect has been observed inside homes where there are combustion sources of NO. Figure 15.13 shows measured outdoor and indoor levels of O<sub>3</sub> in one study with a gas stove turned on or off. Although the outdoor concentration remained relatively constant, the indoor levels were much lower when the gas stove was on compared to when it was off. Although NO<sub>2</sub> was not reported in this study, its levels presumably rose when the stove was on as the O<sub>3</sub> reacted with NO to form NO<sub>2</sub>.

## F. INDOOR VOC-NO<sub>x</sub>-O<sub>3</sub> CHEMISTRY

As discussed in detail throughout this book, there is rich and complex chemistry involving volatile organic compounds (VOCs), oxides of nitrogen, and ozone in ambient air. One might therefore anticipate similar chemistry in indoor air environments, and although there are far fewer studies, this does indeed appear to be the case. Weschler and Shields (1997b) and Wolkoff *et al.* (1997, 1999) review VOC-NO<sub>x</sub> chemistry that could potentially be important in indoor air environments and the implications for human exposures.

Although the chemistry occurring indoors is fundamentally the same as that occurring outdoors and discussed throughout this book, there are some important differences as well. For example, the time available for the chemistry to occur indoors is determined by the ventilation rate of the building. In addition, the light intensity and spectral distributions are quite different from those outdoors, decreasing the relative importance of photochemical reactions and increasing that of “dark reactions.” For example, Pitts *et al.* (1985) followed the loss of methyl nitrite (CH<sub>3</sub>ONO) in brightly lit rooms indoors in a mobile home used for the HONO

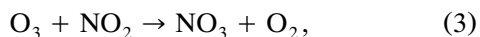


studies; no loss was observed over a 5-h period, whereas this compound photolyzes rapidly outdoors.

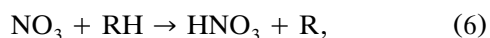
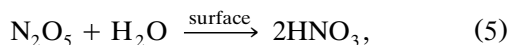
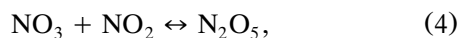
Finally, the concentrations of the reactants, both absolute and relative, are different from those in most outdoor environments. As we have already seen, the concentrations of VOCs tend to be larger due to the large number of sources indoors, whereas that of  $O_3$  tends to be smaller due to its removal on surfaces, e.g., of air conditioning systems. In the absence of combustion sources, concentrations of oxides of nitrogen tend to be similar to those outdoors.

A number of models that incorporate chemical reactions have been developed and applied to indoor air environments. For example, Özkaynak *et al.* (1982) developed a model that incorporated simple  $NO_x$  chemistry. A more comprehensive model that included VOC- $NO_x$  chemistry as well was developed by Nazaroff and Cass (1986) for indoor air environments (it included ventilation, emissions, and also removal by wall loss) and applied to museum environments. Measurements of NO,  $NO_2$ , and  $O_3$  both indoors and outdoors were also made for testing and refining the model predictions. As expected based on the discussion of ozone in Section E,  $O_3$  concentrations indoors, both measured and predicted, were lower than those outdoors due to removal at surfaces; however, removal by reaction with NO, particularly in the morning when NO concentrations were high, was also a significant contributor. The model tended to consistently underpredict NO indoors, which they attribute to its generation in the surface reaction of  $NO_2$  discussed earlier, which was not included in the model chemistry. Although other species were not measured, the results of the model suggest that  $NO_3$  and  $N_2O_5$  levels indoors are greater than those outdoors and that under some circumstances, the same may be true for  $HNO_3$  and  $H_2O_2$ .

Weschler and co-workers (Weschler *et al.*, 1992a, 1994) suggested that the formation of  $NO_3$  indoors,



followed by its well-known secondary chemistry to form  $HNO_3$ ,



may be an important indoor source of  $HNO_3$ . Indirect evidence for an indoor source is the measurement of the indoor-to-outdoor ratio (I/O) of  $HNO_3$  and  $SO_2$  in residences in several locations on the east coast of the United States during the summer months. The I/O

ratio for  $HNO_3$  was about the same as that for  $SO_2$  at one location and larger at a second site. This is not expected, since  $HNO_3$  is much more rapidly lost to surfaces than is  $SO_2$ . In a third location, the I/O ratio for  $HNO_3$  was smaller than that of  $SO_2$ , but not by as much as would be expected from its rapid wall loss. Such data are indicative of an indoor source of  $HNO_3$ , and Weschler *et al.* (1992a) suggest reactions (3)–(6).

Evidence for such indoor chemistry has also been obtained by measuring NO,  $NO_y$ , and  $O_3$  indoors and outdoors in an office building (Weschler *et al.*, 1994). For example, as  $O_3$  rises and NO decreases during the morning hours, the  $O_3$  reaction with NO indoors leads to faster decay of NO than otherwise expected and a slower rate of increase of  $O_3$ . On the other hand, when NO is rising and  $O_3$  falling in the early evening, this reaction speeds up the decay of  $O_3$  and slows the increase in NO.

Weschler and Shields (1997b) suggest that with the higher concentrations of VOCs indoors, their reactions with  $O_3$ ,  $NO_3$ , and OH may be important. There is some experimental evidence that this is indeed the case. For example, carpet exposed to  $O_3$  in a chamber generated HCHO, benzaldehyde, benzoic acid, and acetophenone, all expected products from the reaction of  $O_3$  with styrene (Zhang *et al.*, 1994b) emitted from the latex adhesive used to bind the backing to the carpet; styrene decreased simultaneously (Weschler *et al.*, 1992b). The formation of a series of  $C_5$ – $C_{10}$  aldehydes was observed, which appeared to be from the reaction of  $O_3$  with nonvolatile organics associated with the carpet fibers. Concentrations of HCHO increased by up to a factor of 3 and  $CH_3CHO$  by up to a factor of 20 in the presence of  $O_3$ . Interestingly, no additional effect was observed when  $NO_2$  was also present, suggesting that the nitrate radical was not a significant contributor to the formation of these aldehydes and ketones.

Salthammer *et al.* (1999) examined emissions from commonly available coatings used on furniture and identified numerous oxidation products. These were observed without the addition of oxidants such as ozone, indicating that oxidation in air (perhaps including photodecomposition for some compounds) under typical conditions is sufficient to generate such products. For example, emissions of 2-ethylhexanol were identified from di-2-ethylhexyl phthalate, used as a plasticizer in many coatings.

Reiss *et al.* (1995b) exposed latex paint to  $O_3$  and observed the production of HCHO as well as  $CH_3CHO$  and  $CH_3COCH_3$  for some paint samples. They proposed that these were formed by the reactions of  $O_3$  with some remaining double bonds that were not fully reacted during the process in which the  $CH_2=CHR$

was polymerized to form the latex paint. Similarly, Chang and Guo (1998) and Fortmann *et al.* (1998) report emissions of hexanal during the drying of an alkyd paint; since hexanal was not a component of the paint itself, they proposed that it was formed by the oxidation of unsaturated fatty acid esters in the alkyd resin.

A correlation between indoor ozone and the concentrations of carbonyl compounds and organic acids in homes has been reported in several studies and attributed at least in part to indoor  $O_3$  reactions (e.g., Reiss *et al.*, 1995a; Zhang and Lioy, 1994; Zhang *et al.*, 1994a–c). Not only HCHO and  $CH_3CHO$  but also larger aldehydes have been measured indoors, with indoor concentrations for all but possibly propionaldehyde being much larger than those outdoors (Fig. 15.12). The same is true for formic and acetic acids, which can be formed by ozone reactions from reaction of the Criegee biradicals with water vapor (see Chapter 6.E.2). Zhang *et al.* (1994c) report that indoor formic acid concentrations increased with the indoor concentration of  $O_3$  and with relative humidity, as expected if the reaction of the HCHOO Criegee biradical with water vapor was a significant indoor source.

However, as discussed by Reiss *et al.* (1995a), separating the contribution of ozone reactions from other factors such as temperature and relative humidity, which also affect direct emissions, is difficult. For example, while the production rate of oxygenated organics is correlated with the ozone removal rate, the latter is also correlated with temperature. As a result, both reaction and increased direct emission rates due to higher temperatures may be contributing to these enhanced indoor levels.

Ozone can also react with components found in air ducts. For example, Morrison *et al.* (1998) reported that the sealant and neoprene gaskets used in the ducts emitted VOCs into the airstream, but at relatively low levels compared to the typical concentrations found indoors. However, reaction with  $O_3$  led to increased emissions of aldehydes, particularly the  $C_5$ – $C_{10}$  aldehydes.

The mechanism of the reaction of  $O_3$  with alkenes was discussed in detail in Chapter 6.E.2. It was seen there that these reactions serve as an indoor source of OH through decomposition of the Criegee biradical. Weschler and Shields (1996b) proposed that the indoor reaction of  $O_3$  with alkenes could serve as a source of OH and calculated that at 20 ppb  $O_3$  and average indoor alkene concentrations, a steady-state OH concentration of about  $2 \times 10^5$  molecules  $cm^{-3}$  might be expected.

Subsequently, they measured OH concentrations in an office building using the rate of decay of 1,3,5-tri-

methylbenzene, which reacts with OH but not  $O_3$  (Weschler and Shields, 1997a). Although  $O_3$  and *d*-limonene were injected during the experiment, their concentrations were chosen to be similar to that measured under normal operating conditions. An average OH concentration of  $7 \times 10^5$  molecules  $cm^{-3}$  was obtained, which is about an order of magnitude smaller than daytime peak OH concentrations outdoors but also more than an order of magnitude larger than those outdoors at night (see Chapter 11.A.4i).

As discussed in Chapter 9.C.2, some of the larger alkenes such as terpenes form particles containing low-volatility organics on oxidation with ozone. Hence particle formation might be expected indoors in the presence of such compounds, and indeed this has been observed (Weschler and Shields, 1999).

In short, much of the chemistry that has been observed outdoors also occurs indoors. However, the relative importance of various reactions may be somewhat different due to the different absolute and relative concentrations of the reactants, the lower photolysis rates, the exchange of air with outdoor air, and the presence of relatively large surface areas, which can both remove various species and act as substrates for heterogeneous reactions.

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## G. PARTICLES

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With the epidemiological studies suggesting increased mortality associated with particles (see Chapter 2), there has been increasing interest in indoor particle concentrations compared to outdoor levels. A number of studies have examined this over the years and are summarized in a review by Wallace (1996).

In general, if there are no indoor sources of particles, the levels indoors tend to reflect those outdoors. For example, application of a mass balance model to measurements of indoor and outdoor particle concentrations in Riverside, California, indicated that 75% of  $PM_{2.5}$  and 65% of  $PM_{10}$  in a typical home were from outdoors (Wallace, 1996). Similar conclusions were reached by Koutrakis *et al.* (1991, 1992) for homes in two counties in New York. For example, they report that 60% of the mass of particles in homes is due to outdoor sources. However, the contribution to various individual elements in the particles varies from 22% for copper to 100% for cadmium.

There are some differences in indoor levels of particulate matter in areas with low outdoor compared to high outdoor levels. In the case of high outdoor levels, the indoor concentrations tend to be somewhat lower than those outdoors; for example, Colome *et al.* (1992)

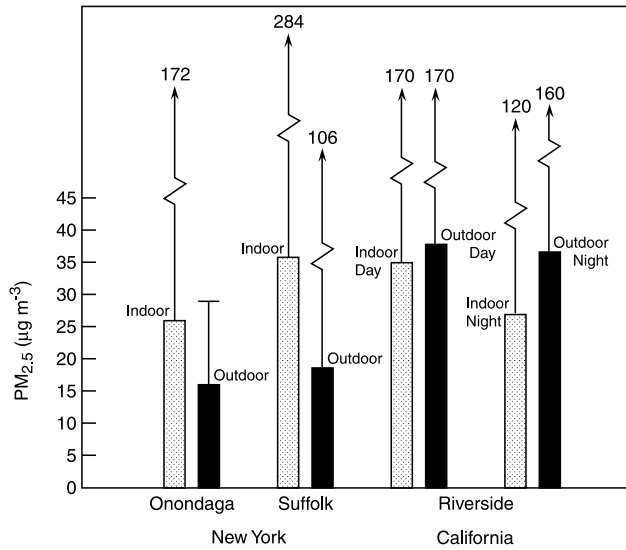


FIGURE 15.14 Geometric mean  $PM_{2.5}$  concentrations indoors and outdoors in Onondaga and Suffolk Counties in New York state and in Riverside, California. The Riverside data are weighted means. The error bars represent the maximum values for the New York sites and the 98th percentile for the Riverside studies (data from Wallace, 1996; based on Sheldon *et al.*, 1989, and Pellizzari *et al.*, 1993).

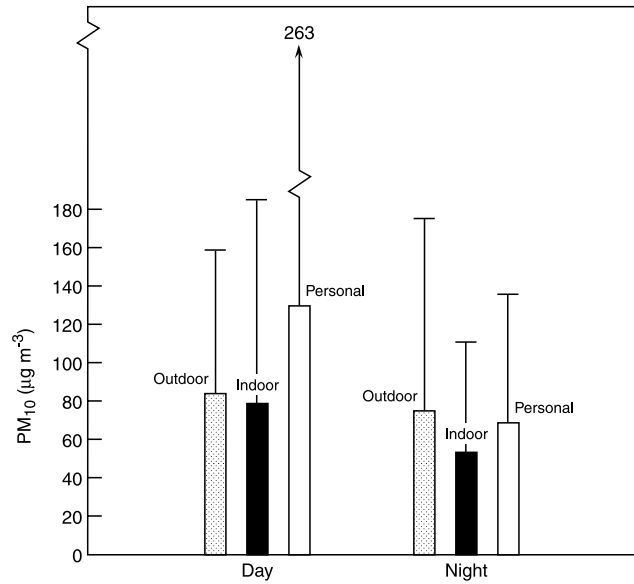


FIGURE 15.15 Geometric mean  $PM_{10}$  concentrations outdoors, indoors, and by personal exposure monitors during the day and at night in Riverside, California, weighted to provide estimates of concentrations for household-days or person-days. The error bars are 90th percentile (data from Clayton *et al.*, 1993).

report that the ratio of indoor-to-outdoor median concentrations of  $PM_{10}$  is 0.7 in residences in southern California. On the other hand, when outdoor levels are low, indoor levels tend to be higher. This is illustrated by the data in Fig. 15.14, which shows indoor and outdoor levels of  $PM_{2.5}$  measured in two counties in New York state (Sheldon *et al.*, 1989) and in Riverside, California (Pellizzari *et al.*, 1993), as summarized by Wallace (1996).

Nighttime mass concentrations indoors tend to be smaller than those during the day, probably because of the decreased activity. Interestingly, when individuals wear personal exposure monitors to measure their actual exposure to particles, the measured mass concentrations tend to be higher than those measured with fixed monitors located indoors. Both of these are illustrated by the data in Fig. 15.15, which shows the geometric mean concentrations of  $PM_{10}$  in Riverside, California, measured outdoors, indoors, and with personal exposure monitors (Clayton *et al.*, 1993). Because of the study design, the measurements were weighted to provide estimates of the concentrations in terms of household-days or person-days, respectively. Indoor levels at night are about 50–70% of those during the day. However, the personal exposure concentrations are 165% of the measured indoor concentrations during the day and 128% at night. This has been dubbed the “body cloud” or “personal cloud” (Rodes, 1991). The reasons for the higher concentrations are not

clear. While skin flakes and clothing fibers contribute to this, their levels do not appear to be sufficiently high to explain the observed levels (Özkaynak *et al.*, 1996). In addition, the elemental composition of the personal exposure particles in some studies has been observed to be very similar to that of indoor aerosol samples, indicating that different sources are not likely to be responsible (Özkaynak *et al.*, 1996; Wallace, 1996). It may be that the activity of the individual is responsible, at least in part. For example, carpets have been shown to be a reservoir of dust particles that can be suspended when walking, vacuuming, etc. (Thatcher and Layton, 1995; Leese *et al.*, 1997). Such activity leads to resuspension of the larger particles preferentially. For example, cleaning activities increased the airborne concentrations of 10- to 25- $\mu\text{m}$  particles by a factor of 30, of 5- to 10- $\mu\text{m}$  particles by a factor of 11, and of 0.5- to 1- $\mu\text{m}$  particles insignificantly (Thatcher and Layton, 1995). Enhanced levels of particulate sulfate and  $H^+$  in personal exposure monitors compared to indoor values (ratios of 1.25 and 3.1, respectively) have been reported (Suh *et al.*, 1992), although Özkaynak *et al.* (1996) report that sulfur in particles in the “personal cloud” was the only element not enhanced compared to indoor levels.

A major source of increased particles indoors is cigarette smoking. (e.g., Spengler *et al.*, 1981; Quackenboss *et al.*, 1989; Neas *et al.*, 1994). Figure 15.16 shows

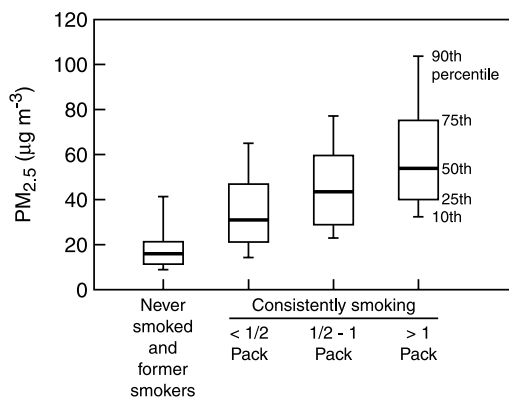


FIGURE 15.16 Annual average concentrations of  $PM_{2.5}$  indoors as a function of smoking in the home. The data are shown in percentiles as marked. (Adapted from Neas *et al.*, 1994.)

the annual average concentrations of  $PM_{2.5}$  measured indoors in homes with nonsmokers and for homes occupied by smokers as a function of the number of packs of cigarettes smoked per day (Neas *et al.*, 1994). There is a steady increase in the indoor levels of  $PM_{2.5}$  as a function of the number of cigarettes smoked, with the increase in  $PM_{2.5}$  estimated to be in the range of 25–45  $\mu\text{g m}^{-3}$  (Wallace, 1996).

In addition to the contribution to the *mass* concentrations of indoor particles, cigarette smoke is of concern because of the mutagens, carcinogens, and toxic air contaminants that are emitted (e.g., see Löfroth *et al.*, 1991; Chuang *et al.*, 1991; California Environmental Protection Agency, 1997; and Nelson *et al.*, 1998). Thus, a variety of both gaseous and particulate polycyclic aromatic hydrocarbons (PAH) and compounds (PAC) have been identified in buildings with cigarette smoke (e.g., Offermann *et al.*, 1991; Mitra and Ray, 1995). Indeed, in the homes of smokers, almost 90% of the total PAH was from tobacco smoke (Mitra and Ray, 1995). Higher levels of mutagenic particles have also been shown to be associated with indoor air containing cigarette smoke (e.g., Lewtas *et al.*, 1987; Löfroth *et al.*, 1988, 1991; Georghiou *et al.*, 1991).

Other significant sources identified in a number of studies are cooking, the use of kerosene heaters, wood burning, and humidifiers. For example, a study carried out under the auspices of the U.S. Environmental Protection Agency, the TEAM study (*Total Exposure Assessment Methodology*), indicated that an increase in  $PM_{10}$  of  $\sim 10\text{--}20 \mu\text{g m}^{-3}$  could be attributed to cooking (Wallace, 1996). This source will obviously depend on the amount of cooking, the types of cooking, and the ventilation. For example, Löfroth *et al.* (1991) measured emissions of particles ranging from 0.07 to 3.5 mg per gram of food cooked, depending on the

particular food. Baek *et al.* (1997) measured indoor and outdoor concentrations of particles in homes, offices, and restaurants in Korea and report ratios of 1.3, 1.3, and 2.4, respectively. The higher value in restaurants, even those using only gas and not charcoal, suggests a significant contribution from cooking.

Kerosene heaters can be significant sources of particles under some circumstances. For example, kerosene heaters were reported to contribute to indoor  $PM_{2.5}$  in homes in Suffolk County, New York, but not Onondaga County; wood stoves and fireplaces and gas stoves did not contribute in either case (Koutrakis *et al.*, 1992; Wallace, 1996). A similar conclusion was reached in a study of eight mobile homes in North Carolina (Mumford *et al.*, 1991).

However, it should be noted that even where kerosene heaters do not contribute significantly to particle *mass* concentrations, they may still be important in terms of health effects. This is because of the composition of the particles emitted, which include polycyclic aromatic compounds and other mutagenic species, as well as sulfate (Leaderer *et al.*, 1990; Traynor *et al.*, 1990). For example, Traynor *et al.* (1990) studied the emissions from unvented kerosene space heaters and identified a number of PAHs (naphthalene, phenanthrene, fluoranthene, anthracene, chrysene, and indeno[*c,d*]pyrene) and nitro-PAHs (1-nitronaphthalene, 9-nitroanthracene, 3-nitrofluoranthene, and 1-nitropyrene), in addition to a host of other gaseous species. Baek *et al.* (1997) also reported increased levels of a number of gases indoors in homes and offices in Korea due to the use of kerosene heaters.

In studies of indoor air in eight mobile homes, Mumford *et al.* (1991) identified the PAHs and nitro-PAHs measured in emissions from kerosene heaters by Traynor *et al.* (1990), as well as a number of compounds that may be animal carcinogens, such as cyclopenta[*c,d*]pyrene, benz[*a*]anthracene, benzofluoranthenes, benzo[*a*]pyrene, and benzo[*ghi*]perylene. While the mass concentrations of  $PM_{10}$  did not increase with the kerosene heater on in six of the eight homes studied, the particles in five of the homes had increased mutagenicity using TA98 with or without S9 added (see Chapter 10.C.4).

In short, not only the mass emissions but also the nature of the compounds emitted must be taken into account in assessing the health effects of indoor particles.

Humidifiers can be a significant source of airborne particles if tap water is used because as the water evaporates from the aerosol, the solids that were in water are left as particles. For example, Highsmith *et al.* (1988, 1992) showed that the airborne particle concentration increased linearly with the total dissolved

solids in the water used in the humidifier. When an ultrasonic humidifier was operated in a closed single room using tap water with total dissolved solids of 303 mg L<sup>-1</sup>, concentrations of fine and coarse particles of 6307 and 771 μg m<sup>-3</sup>, respectively, were generated. When distributed over the whole house, the corresponding levels were 593 and 65 μg m<sup>-3</sup>, respectively. The whole-house values fell to 41 and 13 μg m<sup>-3</sup> when bottled water with total dissolved solids of 24 mg L<sup>-1</sup> was used (Highsmith *et al.*, 1992).

On the other hand, the use of evaporative (“swamp”) coolers appears to decrease particulate matter indoors. For example, Quackenboss *et al.* (1989) report levels of PM<sub>2.5</sub> and PM<sub>10</sub> in homes having such coolers (for both smokers and nonsmokers) that are about half that of homes without them.

Where indoor heating and cooking involves the use of coal or biomass, indoor particle concentrations can be extremely large. For example, Florig (1997) and Ando *et al.* (1996) report that in China typical indoor total suspended particle (TSP) concentrations can be in the range from 250 to 900 μg m<sup>-3</sup> in homes using coal and 950–3500 μg m<sup>-3</sup> in those using biomass fuels. These levels can be compared to annual average outdoor concentrations of 250–410 μg m<sup>-3</sup>. The high concentrations associated with coal burning combined with the mutagenic nature of the emissions have been suggested to be responsible for enhanced lung cancer in China (Mumford *et al.*, 1987). Similarly, Davidson *et al.* (1986) measured TSP concentrations of 2900–42,000 μg m<sup>-3</sup> in homes in Nepal that used biomass fuels, compared to outdoor levels of 280 μg m<sup>-3</sup>. For particles with diameters less than 4 μm, the levels ranged from 870 to 14,000 μg m<sup>-3</sup>.

Similar conclusions regarding the relative indoor and outdoor concentrations have been reached in studies of office and commercial buildings. For example, Ligocki *et al.* (1993) measured indoor and outdoor concentrations of particles and their components at five museums in southern California. The indoor-to-outdoor ratios of particle mass varied over a wide range, depending to a large extent on the ventilation and filtration systems in use. Ratios varied from 0.16 to 0.96 for particles with diameters less than 2.1 μm and from 0.06 to 0.3 for coarse particles with diameters greater than this.

The chemical composition of particles collected in the museums was also compared to that outdoors using a mass balance model. The results indicated that there were significant indoor sources of fine particle organics and that this source(s) was a significant fraction of the total indoor fine particle organic concentration (Ligocki *et al.*, 1993). A similar conclusion was reached by Naik *et al.* (1991) in measurements made in a telephone

switching office. The levels of the *n*-C<sub>27</sub> through *n*-C<sub>33</sub> alkanes in the fine particle fraction were all elevated, suggesting indoor sources such as waxes, polishes, and lubricants. Enhanced levels of dibutyl phthalate and di(2-ethylhexyl) phthalate were also observed in fine particles and attributed to plasticizers used in floor polishes and vinyl products.

Turk *et al.* (1989) measured particle concentrations in 38 buildings that had both smoking and nonsmoking areas. The average mass concentration of respirable particles in smoking areas was 70 μg m<sup>-3</sup> compared to 19 μg m<sup>-3</sup> in nonsmoking areas, whereas the outdoor concentrations were essentially the same, 19 μg m<sup>-3</sup>.

In another study, Ott *et al.* (1996) measured particle concentrations inside and outside a tavern before and after a smoking ban was instated. Average respirable suspended particle mass concentrations were 57 μg m<sup>-3</sup> above the outdoor concentrations prior to the ban, compared to 6–13 μg m<sup>-3</sup> afterward. Cooking and resuspended dust also contributed to the indoor particle mass concentration, but at concentrations about 20–25% of that due to cigarette smoke.

In short, the indoor concentrations of particles depend on the outdoor levels, the ventilation system and exchange rates, and the presence of indoor sources such as cigarette smoke.

In many nonresidential buildings, deposition is of particular interest because of the potential for damage to materials in museums, offices, cultural objects, and industrial sites (e.g., Sinclair *et al.*, 1988, 1990a,b; Nazaroff *et al.*, 1990a; Salmon *et al.*, 1994, 1995). Deposition onto both horizontal and vertical surfaces is of concern, but these can show different behaviors as a function of particle size. For example, deposition of particles to vertical and horizontal surfaces was measured inside five museums. Horizontal deposition velocities increased from ~10<sup>-6</sup> to 10<sup>-3</sup> m s<sup>-1</sup> as particle size increased from about 0.1 to 30 μm as expected for gravitational settling (see Chapter 9.A.3). However, the dependence on particle size of the uptake onto vertical surfaces, which is influenced by thermal and air flow fields (Nazaroff *et al.*, 1990b), was variable, increasing with particle size in some cases, decreasing in others, and, in some, showing no dependence on particle size (Ligocki *et al.*, 1990).

A number of models have been developed for particles indoors (e.g., Nazaroff and Cass, 1989a; Sinclair *et al.*, 1990b; Nazaroff *et al.*, 1990a; Weschler *et al.*, 1996; Wallace *et al.*, 1996, and references therein). This is a complex problem, given the number of potential sources, different deposition velocities for particles of different sizes (e.g., see Chapter 9.A.3 and Nazaroff and Cass (1989b)), the different particle compositions, and the effects of outdoor concentrations and ventila-

tion rates. However, reasonably good agreement has been obtained in many cases between modeled and observed concentrations.

## H. PROBLEMS

1. As discussed in this chapter, a relatively new area in indoor air pollution is that of hydroxyl radical chemistry. However, the importance of indoor OH chemistry (as well as O<sub>3</sub> and NO<sub>3</sub> chemistry) is determined by the rates of the reactions compared to the rate of air exchange. An OH concentration of  $7 \times 10^5 \text{ cm}^{-3}$  has been reported in an indoor air environment by Weschler and Shields (1997a). Assess the importance of the OH reaction for the removal of limonene indoors compared to its removal by reaction with O<sub>3</sub>. The O<sub>3</sub>-limonene rate constant is  $2 \times 10^{-16}$  and the OH-limonene rate constant is  $1.7 \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ . Take the O<sub>3</sub> concentration to be 20 ppb. How do these compare with a typical air exchange rate of  $0.75 \text{ h}^{-1}$  used in these experiments?

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