FORMALDEHYDE 283

## 5. POTENTIAL FOR HUMAN EXPOSURE

#### 5.1 OVERVIEW

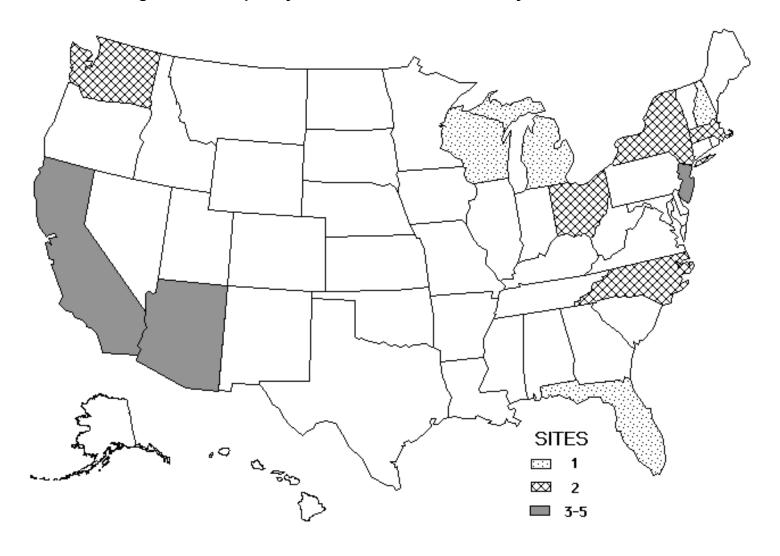
Formaldehyde is produced by both anthropogenic and natural sources. Combustion processes account directly or indirectly for most of the formaldehyde entering the environment. Direct combustion sources include power plants, incinerators, refineries, wood stoves, kerosene heaters, and cigarettes.

Formaldehyde is produced indirectly by photochemical oxidation of hydrocarbons or other formaldehyde precursors that are released from combustion processes (NRC 1981). During smog episodes, indirect production of formaldehyde may be greater than direct emissions (Fishbein 1992). Oxidation of methane is the dominant source of formaldehyde in regions remote from hydrocarbon emissions (Staffelbach et al. 1991). Other anthropogenic sources of formaldehyde in the environment include vent gas from formaldehyde production; exhaust from diesel and gasoline-powered motor vehicles; emissions from the use of formaldehyde as a fumigant, soil disinfectant, embalming fluid, and leather tanning agent; emissions from resins in particle board, and plywood; emissions from resin-treated fabrics and paper; waste water from the production and use of formaldehyde in the manufacture of various resins and as a chemical intermediate; and waste water from the use of formaldehyde-containing resins (EPA 1976a; Kleindienst et al. 1986; NRC 1981; Verschueren 1983). Natural sources of formaldehyde include forest fires, animal wastes, microbial products of biological systems, and plant volatiles.

Formaldehyde has been identified in at least 26 of the 1,428 current or former EPA National Priorities List (NPL) hazardous wastes sites (HazDat 1996). However, the number of sites evaluated for formaldehyde is not known. The frequency of these sites within the United States can be seen in Figure 5-1.

Although formaldehyde is found in remote areas, it probably is not transported there but is generated from longer-lived precursors that have been transported there (NRC 1981). Formaldehyde is soluble and will transfer into rain and surface water. Based upon the Henry's law constant for formaldehyde, volatilization from water is not expected to be significant. No experimental data were found concerning the adsorption of formaldehyde to soil, but because of the low octanol/water partition coefficient (log  $K_{ow}$ =0.35) (SRC 1995b), little adsorption to soil or sediment is expected to occur. No evidence of bioaccumulation has been found.

Figure 5-1. Frequency of NPL Sites with Formaldehyde Contamination



The input of formaldehyde into the environment is counterbalanced by its removal by several pathways. Formaldehyde is removed from the air by direct photolysis and oxidation by photochemically produced hydroxyl and nitrate radicals. Measured or estimated half-lives for formaldehyde in the atmosphere range from 1.6 to 19 hours, depending upon estimates of radiant energy, the presence and concentrations of other pollutants, and other factors (Atkinson and Pitts 1978; DOT 1980; EPA 1982; Lowe et al. 1980; Su et al. 1979). When released to water, formaldehyde will biodegrade to low levels in a few days (Kamata 1966). In water, formaldehyde is hydrated; it does not have a chromophore that is capable of absorbing sunlight and photochemically decomposing (Chameides and Davis 1983).

Levels of formaldehyde in the atmosphere and in indoor air are well documented. In a survey of ambient measurements of hazardous air pollutants, a median formaldehyde concentration of 2.5 ppb was found for a total of 1,358 samples collected at 58 different locations (Kelly et al. 1994). In several separate studies involving rural and urban areas in the United States, atmospheric concentrations ranged from 1 to 68 ppb (Grosjean 1982; Salas and Singh 1986; Schulam et al. 1985; Singh et al. 1982). Generally, indoor residential formaldehyde concentrations are significantly higher than outdoor concentrations. Formaldehyde concentrations measured in complaint homes (homes where people have complained of adverse symptoms), mobile homes, and homes containing large quantities of particle board or ureaformaldehyde foam insulation (UFFI) have been measured at 0.02 ppm to 0.8 ppm, with levels as high as 4 ppm, sufficient to cause irritating symptoms, observed in some instances (Gold et al. 1993). Since the time many of the above monitoring studies were performed, plywood and particle board manufacturing methods have been changed to reduce the formaldehyde emission levels in the finished product (EPA 1996). Similarly, home construction methods have changed and the use of UFFI has been greatly reduced since the mid-eighties (CPSC 1997). A recent pilot study on a newly constructed home reported localized formaldehyde concentrations of 0.076 ppm (Hare et al. 1996). Approximately 30 days after the installation of pressed wood products, the average indoor concentration attained a level of 0.035–0.45 ppm. Older conventional homes tend to have the lowest indoor concentrations of formaldehyde, with values typically less than 0.05 ppm (Gold et al. 1993), and mobile homes the highest due to their low rate of air exchange (Wolff 1991).

Although pressed wood products may be a source of formaldehyde in indoor air, there are numerous others. These include permanent press fabrics, fiberglass products, decorative laminates, paper goods, paints, wallpaper, and cosmetics (Kelly et al. 1996). Its presence in indoor air also results from combustion sources, such as stoves, heaters, or burning cigarettes (Matthews et al. 1985; NRC 1981).

Formaldehyde may also arise from the degradation of volatile organic chemicals commonly found in indoor air (Weschler and Shields 1996; Zhang et al. 1994b).

Formaldehyde is unstable in water; however, it has been detected in municipal and industrial aqueous effluents, rainwater, lake water, and some waterways (EPA 1976b; Hushon et al. 1980). Formaldehyde levels in rainwater collected in California are low, ranging from not detectable to 0.06 µg/mL (Grosjean and Wright 1983). Measured concentrations of formaldehyde range from 0.12 to 6.8 mg/L in fogwater (Igawa et al. 1989; Muir 1991); from 1.4 to 1.8 mg/L in cloudwater (Igawa et al. 1989); and from 0.25 to 0.56 mg/L in mist samples (Grosjean and Wright 1983). No data on formaldehyde levels in soil could be found in the literature.

Formaldehyde was found in three types of chewing tobacco (Chou and Que Hee 1994) and in cigarette smoke (Mansfield et al. 1977). Formaldehyde has also been found at levels ranging from 1 to 3,517 ppm in fabric samples (Schorr et al. 1974).

A major route of formaldehyde exposure for the general population is inhalation of indoor air; releases of formaldehyde from new or recently installed building materials and furnishings may account for most of the exposure. Environmental tobacco smoke may contribute 10–25% of the exposure. Since formaldehyde in food is not available in free form, it is not included in estimated exposures (Fishbein 1992). Consumers can be exposed to formaldehyde gas through its use in construction materials, wood products, textiles, home furnishings, paper, cosmetics, and pharmaceuticals. Dermal contact with formaldehyde containing materials, including some paper products, fabrics, and cosmetics, may also lead to consumer exposure. Commuters may be exposed to formaldehyde while riding in automobiles or subways, walking, and biking (Chan et al. 1991).

Occupational exposure to formaldehyde can occur during its production and during its use in the production of end products, in the garment industry, in the building materials industry, and in laboratories. Health care professionals may be exposed to formaldehyde vapors during preparation, administration, and/or cleanup of various medicines. Pathologists, histology technicians, morticians, and teachers and students who handle preserved specimens may also be exposed. The National Occupational Exposure Survey (NOES), conducted from 1981 to 1983 indicated that 1,329,332 workers in various professions were exposed to formaldehyde in the United States (NIOSH 1995b).

A subpopulation with potentially high exposures to formaldehyde are residents of mobile homes due to the frequent use of pressed wood products and their low rate of air exchange (Wolff 1991). Members of the general population who come in contact with a large amount of unwashed permanent press fabrics treated with formaldehyde-releasing resins may also be exposed to high levels.

#### 5.2 RELEASES TO THE ENVIRONMENT

According to the Toxics Release Inventory (TRI), in 1996, 21 million pounds (9.6 million kg) of formaldehyde were released to the environment from 674 domestic manufacturing and processing facilities (TRI96 1998). This number represents the sum of all releases of formaldehyde to air, water, soil, and underground injection wells. An additional 1.8 million pounds (0.8 million kg) were transferred to publicly owned treatment works (POTWs), and 1.3 million pounds (0.6 million kg) were transferred off-site (TRI96 1998). Table 5-1 lists amounts released from these facilities. The TRI data should be used with caution because only certain types of facilities are required to report (EPA 1995). This is not an exhaustive list.

### 5.2.1 Air

Formaldehyde is released to outdoor air from both natural and industrial sources. Combustion processes account directly or indirectly for most of the formaldehyde entering the atmosphere. One important source of formaldehyde is automotive exhaust from engines not equipped with catalytic converters (WHO 1989). Automobiles were found to emit about 610 million pounds (277 million kg) of formaldehyde each year (EPA 1976a). Emissions were reduced with the introduction of the catalytic converter in 1975 (Zweidinger et al. 1988); although they have been found to rise again with the introduction of oxygenated fuels (Kirchstetter et al. 1996). Gaffney and coworkers found that in urban areas the introduction of oxygenated fuels led to increased anthropogenic emissions of formaldehyde during the winter, the season these fuels are used (Gaffney et al. 1997). Formaldehyde in vehicle emissions in 1994 were found to increase by 13% within 2 months after the average oxygen content of fuels sold in the San Francisco Bay area increased from 0.3 to 2.0% by weight (Kirchstetter et al. 1996).

		Total of reported amounts released in pounds per year <sup>a</sup>						
State <sup>b</sup>	Number of Facilities	Air <sup>c</sup>	Water	Land	Underground Injection	Total Environment <sup>d</sup>	POTW <sup>e</sup> Transfer	Off-site Waste Transfer
AK	1	375	0	0	0	375	0	0
AL	31	477,346	1,312	361	0	479,019	2,854	23,887
AR	15	204,464	735	5	0	205,204	1,015	5,229
AZ	3	12,873	0	0	0	12,873	13,000	13,140
CA	25	417,661	250	769	0	418,680	68,609	3,658
CO	2	2,062	0	0	0	2,062	170	0
CT	6	27,833	4,581	0	0	32,414	29,641	16,967
DE	1	1,965	0	0	0	1,965	0	0
FL	6	95,126	1,522	37	0	96,685	0	1,000
GA	36	702,021	714	500	0	703,235	4,600	12,600
IA	4	30,736	400	0	0	31,136	580	1,605
IL	24	37,944	2,084	204	0	40,232	22,900	67,527
IN	15	76,532	0	0	0	76,532	47,761	5,171
KS	8	278,316	0	0	27,000	305,316	10,320	1,198
KY	8	65,275	21,902	0	0	87,177	1,510	27,180
LA	31	330,423	15,674	382	8,601,956	8,948,435	0	76,529
MA	10	60,059	10	1,500	0	61,569	809,791	17,273
MD	2	5,517	0	0	0	5,517	0	6,175
ME	3	121,198	0	0	0	121,198	5,229	58
MI	30	205,766	2,396	12,553	250	220,965	493,193	227,079
MN	15	536,241	0	0	0	536,241	17,250	3,945
MO	12	441,774	1,127	5	0	442,906	19,343	67,062
MS	16	477,785	1,102	2	0	478,889	0	4,671
MT	4	100,214	0	0	0	100,214	0	250
NC	39	1,048,939	6,689	1,201	0	1,056,829	15,905	38,161
NH	4	5,582	9,419	4,100	0	19,101	6,372	512
NJ	18	52,595	3,977	250	0	56,822	42,613	2,509
NM	1	4,570	0	0	0	4,570	0	0

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	_	Total of reported amounts released in pounds per year <sup>a</sup>						
State <sup>b</sup>	Number of Facilities	Air <sup>c</sup>	Water	Land	Underground Injection	Total Environment <sup>d</sup>	POTW <sup>e</sup> Transfer	Off-site Waste Transfer
NV	1	4,001	0	0	0	4,001	0	802
NY	23	280,861	8,409	40	0	289,310	8,103	56,005
ОН	49	1,031,374	87,225	87,335	13,000	1,218,934	126,617	173,090
OK	4	64,911	4	0	0	64,915	0	13,642
OR	25	1,070,119	3,600	20	0	1,073,739	24,928	15,676
PA	21	274,854	1,331	0	0	276,185	0	16,703
PR	4	2,766	0	0	0	2,766	20,827	530
RI	4	4,753	0	0	0	4,753	2,042	0
SC	39	752,317	57,542	3,800	0	813,659	16,718	217,365
SD	1	55,731	0	0	0	55,731	0	0
TN	8	295,461	1,170	397	0	297,028	8,329	5,493
TX	55	996,970	31,770	500	761,069	1,790,309	4,557	60,341
UT	4	18,313	0	0	0	18,313	23,568	285
VA	25	369,447	956	232	0	370,635	15,059	31,235
VT	1	333	0	0	0	333	0	0
WA	8	109,080	45,720	0	0	154,800	3,028	760
WI	22	138,117	7,422	213	0	145,752	17,204	53,649
WV	10	128,600	960	0	0	129,560	5,136	14,154
	Totals	11,419,200	320,003	114,406	9,403,275	21,256,884		

Source: TRI96 1998

Data in TRI are maximum amounts released by each facility
 Post office state abbreviations used
 The sum of fugitive and stack releases are included in releases to air by a given facility
 The sum of all releases of the chemical to air, land, water, and underground injection wells; and transfers off-site by a given facility

<sup>&</sup>lt;sup>e</sup> POTW = publicly-owned treatment works

Formaldehyde concentrations in jet engine exhaust have been found to range from 0.761 to 1.14 ppm (Miyamoto 1986). Formaldehyde is formed in large quantities in the troposphere by the oxidation of hydrocarbons (Calvert et al. 1972; WHO 1989) leading to elevated formaldehyde levels shortly after periods of high vehicular traffic (Grosjean et al. 1996).

Statistical analyses of formaldehyde data from four sites in New Jersey have been used to evaluate the effects of automobile traffic and photochemical formation on formaldehyde concentrations (Cleveland et al. 1977). Integrated formaldehyde concentrations during the hours 5:00 a.m. to 8:00 p.m. decreased from workdays to Saturdays to Sundays, corresponding to a decrease in motor vehicle traffic. On workdays, formaldehyde concentrations were higher on days with more photochemical activity.

Altshuller (1993) investigated the sources of aldehydes in the atmosphere during the night and early morning hours (between 9:00 p.m. and 9:00 a.m.). At night, the predominant sources of aldehydes should be the reaction of alkenes with O<sub>3</sub> and NO<sub>3</sub>; during the early daylight hours, OH radical reactions with alkenes and alkanes will also contribute to aldehyde production. Altshuller (1993) found that, although the emissions of formaldehyde from vehicular exhaust are substantial relative to emissions of alkenes from vehicular exhaust, the secondary atmospheric production of aldehydes from the alkenes emitted from all vehicular sources during the period 9:00 p.m. to 9:00 a.m. can exceed the primary emissions of formaldehyde. However, if there is a large shift in the future to vehicles fueled with methanol and/or natural gas, formaldehyde emissions from the exhaust could predominate over the secondary emissions of formaldehyde during the 9:00 p.m. to 9:00 a.m. period. It should be noted that the rate of secondary production of aldehydes during the period 9:00 p.m. to 9:00 a.m. is much less than during the late morning and afternoon hours (Altshuller 1993).

Grosjean et al. (1983) estimated the relative contributions of direct emissions and atmospheric photochemistry to levels of formaldehyde and other carbonyls in Los Angeles using measurements made simultaneously at a near-source site and at a site where the air quality was dominated by transport of polluted air masses from the downtown area (downwind smog receptor site). They found that the formaldehyde/carbon monoxide ratios were substantially higher at the downwind locations (average values, 3.2–11.7) than at the near-source site (average value, 1.8), indicating that photochemical production predominates over direct emissions in controlling formaldehyde levels in Los Angeles air. Using two models, their data were translated into formaldehyde photochemical production rates of 12–161 tons per day.

The amount of formaldehyde released to the atmosphere in 1996 by U.S. industrial facilities sorted by state is given in Table 5-1 (TRI96 1998). According to TRI96 (1998), an estimated total of 11.4 million pounds (5.2 million kg) of formaldehyde, amounting to approximately 54% of the total environmental release, was discharged to the air from 674 manufacturing and processing facilities in the United States in 1996. The TRI data should be used with caution since only certain types of facilities are required to report. This is not an exhaustive list.

Dempsey (1993) estimated the masses of selected organic emissions from hazardous waste incinerators (HWIs) on a nationwide scale using "reasonable worst-case" assumptions. Formaldehyde emissions formed during combustion of hazardous wastes were estimated to be 892 ng/L, which would result in a release of 7.8 tons of formaldehyde to the air per year. When compared to the 1990 TRI air release data from U.S. manufacturing operations, formaldehyde emissions from HWIs were found to be very small (0.12%).

There is a potential for release of formaldehyde to air from hazardous waste sites. Formaldehyde has been detected in air samples collected at 5 of the 26 hazardous waste sites where formaldehyde has been detected in some environmental medium (HazDat 1996).

Pressed wood products contribute to indoor formaldehyde levels. Combustion sources and phenol-formaldehyde resin bonded products generally are weak emitters to indoor air. Common indoor combustion sources include gas burners and ovens, kerosene heaters, and cigarettes (Matthews et al. 1985).

Formaldehyde also arises in the atmosphere from natural sources. In monitoring studies performed in New Mexico, 1993–94, Gaffney and coworkers looked at total (average) formaldehyde concentrations and formaldehyde:acetaldehyde ratios (Gaffney et al. 1997). These researchers reported that if formaldehyde and acetaldehyde were being formed solely from the atmospheric oxidation of naturally occurring alkenes, the ratio of the two chemicals would be expected to be about 10. In this study, the formaldehyde:acetaldehyde ratio was lowest during the winter months and highest in the summer months. Gaffney et al. (1997) concluded that atmospheric formaldehyde in urban areas resulted from both anthropogenic emissions and natural sources in the summer and primarily from anthropogenic sources during the winter.

In a study conducted at the Inhalation Toxicology Research Institute, formaldehyde release rate coefficients were measured for six types of consumer products (Pickrell et al. 1983). Release rates calculated per unit surface area (µg/m²/day) were used to rank the products in the following order: pressed wood products > >clothes . insulation products . paper products > fabric > carpet. Release rates from pressed wood products ranged from below the limit of detection for an exterior plywood to 36,000 µg/m<sup>2</sup>/day for some paneling. Other release rates were: for articles of new clothing not previously washed, 15–550 μg/m²/day; for insulation products, 52–620 μg/m²/day; for paper plates and cups, 75–1,000 μg/m²/day; for fabrics, from below the limit of detection to 350 μg/m²/day; and for carpets, from below the limit of detection to 65 µg/m<sup>2</sup>/day. In a follow-up to Pickrell's study (Pickrell et al. 1984), performed as a result of changes to product manufacturing processes, many of these release rates were reinvestigated. Formaldehyde release rates for a variety of bare urea-formaldehyde wood products (1/4 to 3/4") were reported to range from 8.6 to 1.578 µg/m<sup>2</sup>/hr, coated urea-formaldehyde wood products from 1 to 461 µg/m<sup>2</sup>/hr, permanent press fabrics from 42 to 214 µg/m<sup>2</sup>/hr, decorative laminates from 4 to  $50 \mu g/m^2/hr$ , fiberglass products from 16 to  $32 \mu g/m^2/hr$ , and bare phenol-formaldehyde wood products from 4 to 9 µg/m<sup>2</sup>/hr (Kelly et al. 1996). Paper grocery bags and towels had emission rates of 0.4 and  $<0.3 \mu g/m^2/hr$ , respectively. For wet products, the emission rates were: latex paint 591  $\mu g/m^2/hr$ ; more expensive latex paint 326 μg/m²/hr; fingernail hardener 215,500 μg/m²/hr; nail polish 20,700 μg/m²/hr; and commercially applied urea-formaldehyde floor finish 421 and 1,050,000 µg/m<sup>2</sup>/hr for base and topcoats, respectively (Kelly et al. 1996).

Formaldehyde may also arise in indoor air through the degradation of other organic compounds. Naturally occurring unsaturated hydrocarbons, such as limonene and pinene (which may also be released from consumer products), and anthropogenic compounds, such as 4-vinylcyclohexene (an emission from carpet padding), and other alkenes commonly found in indoor air have been found to produce formaldehyde via their initial reaction with ozone (Weschler and Shields 1996; Zhang et al. 1994b). Reiss and coworkers estimated that the amount of formaldehyde released by this process is 0.87 µg/sec in winter months and 2.43 µg/sec in the summer (Reiss et al. 1995).

In another study on indoor formaldehyde emissions, quasi steady-state emission rates of formaldehyde from new carpets were measured in a large-scale environmental chamber (Hodgson et al. 1993). The emission rates were 57.2 and 18.2  $\mu$ g/m²/hour at 24 and 168 hours, respectively, after the start of each experiment. Similar results were observed in a Swedish study where indoor formaldehyde levels were found to be higher in homes having wall to wall carpeting (Norback et al. 1995). Another recent

Swedish study on indoor emissions reported that oil-based skin care products known to contain formaldehyde precursors (donors) released formaldehyde to the air even if the material had been in storage for one year (Karlberg et al. 1998).

Formaldehyde release from pressed wood products is due to latent formaldehyde. During the pressing process, hot steam from moist wood particles transfers heat, formaldehyde, and other volatiles from the surface of the mat to the core of the board where unreacted urea-formaldehyde resin components accumulate. The resulting formaldehyde concentration in the core is approximately twice that of the surface. Release of formaldehyde is diffusion-controlled and gradually decreases over time (Meyer and Hermanns 1985). Formaldehyde can also be produced by hydrolytic cleavage of unreacted hydroxymethyl groups in the formaldehyde resins. Melamine formaldehyde resins generally are more stable, and the amounts of formaldehyde emitted from them are much lower (WHO 1989).

A formaldehyde emission rate of 0.48  $\mu$ g/kilojoule has been determined for normal operation of unvented gas ranges; however, this emission rate leads to relatively low indoor formaldehyde concentrations (Moschandreas et al. 1986). Formaldehyde emission rates ranging from 0.43 to 4.2  $\mu$ g/kilojoule were measured from eight well tuned, unvented gas-fired space heaters operated at full fuel input in an environmental chamber with low ventilation (Traynor et al. 1985). In another study, formaldehyde emissions factors from unvented kerosene heaters ranged from 0.18 to 0.47  $\mu$ g/kilojoule (Woodring et al. 1985).

Using an average cigarette consumption rate of approximately 10 cigarettes per indoor compartment per day with a measured formaldehyde emission rate of 0.97±0.06 mg/hour, Matthews et al. (1985) calculated an average formaldehyde emission rate of 0.4±0.03 mg/hour over 24 hours. Triebig and Zober (1984) report that the level of formaldehyde in side stream cigarette smoke is fifty times higher than main stream smoke, while the National Research Council (NRC 1986) put the value at five to eight times more formaldehyde in side stream smoke. Levels of formaldehyde in nonsmoking office buildings ranged from not detected to 0.22 ppm, while they ranged from not detected to 0.6 ppm where smoking was permitted (Sterling et al. 1987).

Formaldehyde can also be emitted into indoor air from fish during cooking. Amounts of formaldehyde that formed in a headspace when various kinds of fish flesh were heated at 200 EC ranged from 0.48  $\mu$ g/g of mackerel to 5.31  $\mu$ g/g of sardine (Yasuhara and Shibamoto 1995).

The general population may also participate in activities that release formaldehyde to outdoor air. The concentration of formaldehyde in mosquito-coil smoke was found to be 10.9–21.3 ppm/g (Chang and Lin 1998). Composting household waste was also found to emit formaldehyde to air (Eitzer et al. 1997).

#### 5.2.2 Water

Formaldehyde is released to water from the discharges of both treated and untreated industrial waste water from its production and from its use in the manufacture of formaldehyde-containing resins.

Formaldehyde can also be formed in seawater by photochemical processes (Mopper and Stahovec 1986).

Calculations of sea-air exchange have indicated that this process is probably a minor source of formaldehyde in the sea (Mopper and Stahovec 1986).

The amount of formaldehyde released to surface water and POTWs in 1996 by U.S. industrial facilities sorted by state is shown in Table 5-1 (TRI96 1998). According to TRI96 (1998), an estimated total of 320,003 pounds (145,153 kg) of formaldehyde, amounting to 2% of the total environmental release, was discharged to surface water in 1996. An additional 1.9 million pounds (0.8 million kg) of formaldehyde were discharged to POTWs (TRI96 1998). The TRI data should be used with caution since only certain facilities are required to report (EPA 1995). This is not an exhaustive list. As a result of secondary treatment processes in POTWs, only a fraction of the formaldehyde that enters POTWs is expected to be released subsequently to surface water; however, this percentage is not known for formaldehyde. Experiments conducted at three full-scale drinking water treatment plants and a pilot plant provided evidence that ozone treatment resulted in the production of measurable levels of formaldehyde in all of the plants studied (Glaze et al. 1989).

There is a potential for release of formaldehyde to water from hazardous waste sites. Formaldehyde has been detected in surface water samples collected at 5 of the 26 hazardous waste sites and in groundwater samples collected at 4 of the 26 hazardous waste sites where formaldehyde has been detected in some environmental medium (HazDat 1996).

# 5.2.3 Soil

Formaldehyde is released to soils through industrial discharges and through land disposal of formaldehyde-containing wastes. The amount of formaldehyde released to land in 1996 by U.S. industrial facilities sorted by state is shown in Table 5-1 (TRI96 1998). According to TRI96 (1998), an estimated total of 114,406 pounds (51,894 kg) of formaldehyde, amounting to 0.54% of the total environmental release, was discharged to land from U.S. manufacturing or processing facilities in 1996. An additional 9.4 million pounds (4.3 million kg), constituting about 44% of total environmental emissions, were released via underground injection (TRI96 1998). Also, some of the estimated 1.3 million pounds of formaldehyde wastes transferred off-site (see Table 5-1) may be ultimately disposed of in land. The TRI data should be used with caution since only certain facilities are required to report (EPA 1995). This is not an exhaustive list.

There is a potential for the release of formaldehyde to soil from hazardous waste sites. Formaldehyde has been detected in soil samples collected at 1 of the 26 hazardous waste sites and in sediment samples collected at 1 of the 26 hazardous waste sites where formaldehyde has been detected in some environmental medium (HazDat 1996).

#### 5.3 ENVIRONMENTAL FATE

In reviewing the fate of formaldehyde in the environment, it should be noted that the environmental factors that influence the bioavailability to humans of formaldehyde from contaminated air, water, or plant material have not been studied.

## 5.3.1 Transport and Partitioning

Formaldehyde is released to the atmosphere in large amounts and is formed in the atmosphere by the oxidation of hydrocarbons. However, the input is counterbalanced by several removal paths (Howard 1989). Because of its high solubility, there will be efficient transfer into rain and surface water, which may be important sinks (NRC 1981). One model has predicted dry deposition and wet removal half-lives of 19 and 50 hours, respectively (Lowe et al. 1980). Although formaldehyde is found in remote areas, it probably is not transported there, but is generated from longer-lived precursors that have been transported (NRC 1981).

No information concerning the fate of formaldehyde in groundwater could be found in the literature. Based upon the Henry's law constant for formaldehyde (3.27x10<sup>-7</sup> atm-m<sup>3</sup>/mol) (Dong and Dasgupta 1986), volatilization from water should not be significant (Lyman 1982). Also, little adsorption to sediment would be expected to occur.

No information could be found concerning the adsorption of formaldehyde to soil. However, its low octanol/water partition coefficient (log  $K_{ow}$  = 0.35) (SRC 1995b) suggests that adsorption to soil is low. Using the log  $K_{ow}$  of 0.35, a log  $K_{oc}$  of 1.57 can be calculated (Lyman 1982). This value of log  $K_{oc}$  suggests a very high mobility and leaching potential of formaldehyde in soil (Swann et al. 1983). At high concentrations, formaldehyde gas adsorbs somewhat to clay mineral, which is important to its use as a soil fumigant (De and Chandra 1979).

Plants, such as kidney beans and barley, can absorb gaseous formaldehyde through their leaves (EPA 1976a). Experiments performed on a variety of fish and shrimp showed no evidence of the bioaccumulation of formaldehyde (Hose and Lightner 1980; Sills and Allen 1979). Because formaldehyde is rapidly metabolized (Casanova et al. 1988), bioaccumulation is not expected to be important.

## 5.3.2 Transformation and Degradation

Formaldehyde undergoes a number of different transformation and degradation reactions in the environment as discussed in the following sections. The resulting environmental transformation products within different media are shown in Table 5-2.

#### 5.3.2.1 Air

Formaldehyde is removed from the atmosphere by direct photolysis and oxidation by photochemically produced hydroxyl radicals. Formaldehyde absorbs ultraviolet (UV) radiation at wavelengths of 360 nm and longer (DOT 1980); therefore, it is capable of photolyzing in sunlight. A half-life of 6 hours has been measured for photolysis in simulated sunlight (Su et al. 1979). There are two photolytic pathways, one producing H<sub>2</sub> and CO, and the other producing H and HCO radicals (Calvert et al. 1972; Lowe et al. 1980). When the rates of these reactions are combined with estimates of actinic irradiance, the predicted half-life of formaldehyde due to photolysis in the lower atmosphere is 1.6 hours at a solar zenith angle of

Table 5-2. Environmental Transformation Products of Formaldehyde by Medium

Reaction	Comments	Reference
Air		
$CH_2O + hv 6 HCO + H$	Photolysis, pathway 1	Calvert et al. 1972
$CH_2O + hv 6 H_2 + CO$	Photolysis, pathway 2	Calvert et al. 1972
$CH_2O + NO_3 6 HCO + HNO_3$	H-atom abstraction by NO <sub>3</sub> radical	Kao 1994
$CH_2O + HO 6 H_2O + HCO$	H-atom abstraction by HO radical	NRC 1981
Dilute Aqueous Solution		
$CH_2O + H_2O + CH_2(OH)_2$	Formation of <i>gem</i> -diol methylene glycol; $k_{298E}$ = $7.0 \times 10^3$ M atm <sup>-1</sup>	Kumar 1986
<b>Concentrated Solution</b>		
$CH_2O 6 H(CH_2O)_nOH + C_3H_6O_3$	Formation of paraformaldehyde and trioxane	EPA 1991c
Cloudwater		
$CH_2(OH)_2 + OH 6 CH(OH)_2 + H_2O (1)$	Formation of formic acid;	Chameides and Davis
$CH(OH)_2 + O_2 6 HO_2 + (HCOOH)_{aq}(2)$	$k_1 = 2x10^9 \text{ M}^{-1} \text{ s}^{-1}$ $k_2 = 4.5x10^9 \text{ M}^{-1} \text{ s}^{-1}$	1983

40 degrees (Calvert et al. 1972). Based on its rate of reaction with photochemically produced hydroxyl radicals, formaldehyde has a predicted half-life of approximately 19 hours in clean air and about half that time in polluted air (Atkinson and Pitts 1978; DOT 1980; EPA 1982). Lowe et al. (1980) report the lifetime of formaldehyde in the sunlit atmosphere, due to photolysis and reaction with hydroxyl radicals, is 4 hours. Singh et al. (1982) report an estimated daily loss rate of formaldehyde of 88.2% on the basis of hydroxyl radical activity and photolysis. The hydroxyl-radical-initiated oxidation of formaldehyde also occurs in cloud droplets to form formic acid, a component of acid rain (Chameides 1986; Chameides and Davis 1983). Calculations by Benner and Bizjak (1988) suggest that removal of formaldehyde by H<sub>2</sub>O<sub>2</sub> is probably negligible in atmospheric droplets. When formaldehyde is irradiated in a reactor, the half-life is 50 minutes in the absence of NO<sub>2</sub> and 35 minutes in the presence of NO<sub>2</sub> (Bufalini et al. 1979; EPA 1976a). The primary products formed are formic acid and CO (Su et al. 1979). The reaction of formaldehyde with nitrate radicals, insignificant in the daytime, may be an important removal mechanism at night (NRC 1981). Formaldehyde reacts with the NO<sub>3</sub> radical by H-atom extraction with a half-life of 12 days, assuming an average nighttime NO<sub>3</sub> radical concentration of 2x10<sup>9</sup> molecules per cm<sup>3</sup> (Atkinson et al. 1984).

#### 5.3.2.2 Water

When released to water, formaldehyde will biodegrade to low levels in a few days (Howard 1989). In nutrient-enriched seawater, there is a long lag period (. 40 hours) prior to measurable loss of formaldehyde by presumably biological processes (Mopper and Stahovec 1986). Formaldehyde in aqueous effluent is degraded by activated sludge and sewage in 48–72 hours (EPA 1976a; Hatfield 1957; Heukelekian and Rand 1955; Verschueren 1983). In a die-away test, using water from a stagnant lake, degradation was complete in 30 hours under aerobic conditions and 48 hours under anaerobic conditions (EPA 1976a). Bhattacharya and Parkin (1988) used anaerobic chemostats to study fate and kinetic effects of sludge and continuous additions of formaldehyde to acetate and propionate enrichment systems. The high reduction of formaldehyde with continuous addition is indicative of biodegradation, since the combination of volatilization, adsorption, and chemical transformation should account for less than 25% of the removal. Up to 80% of the formaldehyde was removed, with biodegradation accounting for 55–60%.

Chameides and Davis (1983) postulated that formaldehyde dissolved in cloudwater should not photolytically decompose because CH<sub>2</sub>(OH)<sub>2</sub> is not a chromophore, and that the more probable fate of

dissolved formaldehyde is oxidation by OH with the ultimate formation of formic acid. Experiments performed by Mopper and Stahovec (1986) suggest that formaldehyde is formed and consumed in seawater as a result of a number of interacting photochemical, biological, and physical processes. Diurnal fluctuations of formaldehyde ranging from 15 to 50 nM were measured in humic-rich waters off the west coast of Florida over a 3-day sampling period.

Concentrated solutions containing formaldehyde are unstable, both oxidizing slowly to form formic acid and polymerizing (Gerberich et al. 1980). In the presence of air and moisture, polymerization takes place readily in concentrated solutions at room temperature to form paraformaldehyde, a solid mixture of linear polyoxymethylene glycols containing 90–99% formaldehyde (EPA 1984).

#### 5.3.2.3 Sediment and Soil

The fate of formaldehyde in soil has not been determined (Howard 1989).

## 5.4 LEVELS MONITORED OR ESTIMATED IN THE ENVIRONMENT

Reliable evaluation for human exposure to formaldehyde depends in part on the reliability of supporting analytical data from environmental samples and biological specimens. In reviewing data on formaldehyde levels monitored in the environment, it should also be noted that the amount of chemical identified analytically is not necessarily equivalent to the amount that is bioavailable.

#### 5.4.1 Air

A recent survey of emission data from stationary and mobile sources was used as input for an atmospheric dispersion model to estimate outdoor toxic air contaminant concentrations for 1990 for each of the 60,803 census tracts in the contiguous United States (Woodruff et al. 1998). The average long term background concentration estimated for formaldehyde was 0.2 ppb (Woodruff et al. 1998).

In a survey of ambient measurements of hazardous air pollutants, a median formaldehyde concentration of 2.5 ppb was found for a total of 1,358 samples collected at 58 different locations (Kelly et al. 1994). Air samples collected daily in Schenectady, New York, during the months of June through August 1983 had formaldehyde concentrations ranging from 1 to 31 ppb. There was a significant daily variation that

appeared to correlate with traffic conditions (Schulam et al. 1985). In the same study, formaldehyde levels measured on the summit of Whiteface Mountain in Wilmington, New York, ranged from 0.8 to 2.6 ppb. Ambient formaldehyde concentrations at the California State University Los Angeles campus ranged from 2 to 40 ppb during the period from May to June 1980; concentrations in Claremont, California, ranged from 3 to 48 ppb from September 19 to October 8, 1980 (Grosjean 1982).

Formaldehyde was measured in urban ambient air in 8 cities for 2- to 3-day periods from June 1980 to April 1984. The average concentrations were 11.3 ppb in St. Louis, Missouri (June 5–7, 1980); 12.3 ppb in Denver, Colorado (June 23–24, 1980); 2.3 ppb in Denver, Colorado (April 1–2, 1984); 19 ppb in Riverside, California (July 8–10, 1980); 14.3 ppb in Staten Island, New York (April 3–4, 1981); 18.5 ppb in Pittsburgh, Pennsylvania (April 15–16, 1981); 11.3 ppb in Chicago, Illinois (April 27–28, 1981); 15.5 ppb in Downey, California (February 28–March 1, 1984); and 3.8 ppb in Houston, Texas (March 18–19, 1984). The maximum concentration in each city ranged from 5.5 ppb in Denver in April to 67.7 ppb in Downey (Salas and Singh 1986; Singh et al. 1982).

Formaldehyde was detected in 99% of 48 ambient air samples obtained in Ohio urban centers, June-July 1989, at a mean and maximum concentration of 3.0 and 15.5 ppb, respectively (Spicer et al. 1996). The diurnal changes in concentration were found to be consistent with initial direct emissions from vehicles followed by secondary photochemical production and, ultimately, atmospheric removal. These data indicate that formaldehyde concentrations in urban atmospheres are expected to be the highest during, or shortly after rush hour, or other periods of high vehicular traffic. Similar results were obtained when temporal formaldehyde concentrations were measured during a smog event in California, September 1993 (Grosjean et al. 1996). In central Los Angeles, higher formaldehyde concentrations were found to correlate with vehicular traffic while at a downwind location, the concentration was found to spike later in the day. The average atmospheric formaldehyde concentration for all sites was 5.3 ppbv (parts per billion by volume) in this study, while the background concentration from San Nicolas island (off the Southern California coast) was 0.8 ppmv (parts per million by volume). Average annual formaldehyde concentrations in California have been reported to vary from a minimum of 3.2 ppbv in the San Francisco Bay area to 4.9 ppbv in the South Coast Air Basin (Seiber 1996).

Several monitoring studies have been conducted in the United States to measure the concentration of formaldehyde in indoor environments (EPA 1987d; Gammage and Hawthorne 1985; Hawthorne et al. 1985, 1986b; Stock 1987). The results for a variety of housing types and ages have been compiled by Gold et al. (1993) and are presented in Table 5-3. Much of the data was collected in older homes, in

Table 5-3. Indoor Concentrations of Formaldehyde in U.S. Homes

		Concer	ntration (ppm)		
Building type	Number	Range	Mean	References	
With UFFI	>1,200	0.01-3.4	0.05-0.12	EPA 1987d; Gammage and Hawthorne 1985	
Without UFFI	131	0.01-0.17	0.025-0.07	Gammage and Hawthorne 1985	
Complaint Mobile homes	>500	0.00-4.2	0.1–0.9	Gammage and Hawthorne 1985	
Non-complaint Conventional, randomly selected	560	<0.005-0.48	0.027-0.091	EPA 1987d; Stock 1987	
Mobile homes, randomly selected	. 1,200	<0.01-2.9	0.091-0.62	EPA 1987d	
By age Mobile homes new	260		0.86	Gammage and Hawthorne 1985	
older, occupied  Conventional homes	_		0.25	Gammage and Hawthorne 1985	
0–5 years 5–15 years >15 Overall	18 11 11 40	<0.02-0.4	0.08 0.04 0.03 0.06	Hawthorne et al. 1985, 1986b Hawthorne et al. 1985, 1986b Hawthorne et al. 1985, 1986b Hawthorne et al. 1985, 1986b	

UFFI = Urea formaldehyde foam insulation

homes that had UFFI, or in homes in which occupants had filed complaints of formaldehyde irritant symptoms. Since the time many of these monitoring studies were performed, plywood and particle board manufacturing methods have been changed to reduce the formaldehyde emission levels in the finished product (EPA 1996). Similarly, home construction methods have changed, and the use of UFFI has been greatly reduced since the mid-eighties (CPSC 1997). A recent pilot study on a newly constructed and unoccupied house set up to measure formaldehyde emissions from construction materials had a maximum localized formaldehyde concentration of 0.076 ppm, which occurred shortly after a high loading of pressed wood materials, such as kitchen cabinets (Hare et al. 1996). The average indoor concentrations measured in this study were 0.035–0.45 ppm, which was attained approximately 30 days after either high or low loadings of formaldehyde releasing materials were installed.

Earlier studies evaluating randomly selected, non-complaint homes have appeared in the literature. Despite the wide variety of conditions under which these residential formaldehyde studies were conducted, results for given types of housing were consistent within certain broad ranges. In general, indoor residential formaldehyde concentrations were significantly higher than outdoor concentrations which ranged from 0.002 to 0.006 ppm in remote, unpopulated regions from 0.01 to 0.02 ppm (and sometimes 0.05 ppm) in highly populated areas and industrial urban air (Gold et al. 1993). The range of formaldehyde concentrations measured in complaint homes, mobile homes, and homes containing large quantities of particle board or UFFI were 0.02–0.8 ppm, with levels as high as 4 ppm, sufficient to cause irritating symptoms, observed in some instances. Formaldehyde levels in more recently built (<1 year old) conventional homes generally were within the range of 0.05–0.2 ppm, with few measurements exceeding 0.3 ppm. Older conventional homes had the lowest indoor concentrations of formaldehyde with values typically less than 0.05 ppm (Gold et al. 1993), consistent with the expected decrease in latent formaldehyde release from wood-based building materials as they age (EPA 1996; Zinn et al. 1990).

The Texas Department of Health measured formaldehyde in 443 mobile homes between April 1979 and May 1982 at the request of the occupants. Concentrations ranged from below detectable limits (<0.5 ppm) to 8 ppm (Norsted et al. 1985). Of the homes #1 year old, 27% had mean concentrations \$2 ppm, while 11.5% of older homes has concentrations \$2 ppm. The concentration of formaldehyde in mobile homes would be expected to be higher than that found in conventional homes due to their lower rate of air exchange (Wolff 1991).

Sexton et al. (1989) selected an age-stratified random sample of 470 mobile homes, from the more than 500,000 in California, for measurement of 1-week, average indoor formaldehyde concentrations during the periods July–August 1984 and February–March 1985. They observed relatively little variation in formaldehyde concentrations between summer and winter, with average 1-week formaldehyde values of 0.07–0.09 ppm. Of the homes, 31% exceeded the maximum concentration of 0.1 ppm formaldehyde recommended at the time of the study by AIHA, EPA, and the American Society of Heating, Refrigerating and Air Conditioning Engineers. The investigators noted that formaldehyde levels appeared to have been decreasing in mobile homes manufactured since about 1980, probably as a result of increased use of low-formaldehyde-emitting building materials.

The Indiana State Board of Health measured formaldehyde in four specific office and commercial establishments that had poor ventilation. The concentrations ranged from 0.01 to 1.01 ppm. In one case, the source of formaldehyde was urea-formaldehyde foam. In the others, it was particle board, plywood subflooring, and furniture (Konopinski 1983).

Shah and Singh (1988) collected indoor and outdoor data for volatile organic chemicals from both residential and commercial environments. The average daily outdoor concentration of formaldehyde for all outdoor site types (remote, rural, suburban, urban, and source-dominated) was 8.3 ppbv; the average daily indoor concentration of formaldehyde was 49.4 ppbv.

Zhang et al. (1994b) made simultaneous indoor and outdoor measurements of aldehydes at six residential houses in suburban New Jersey during the summer of 1992. Formaldehyde was the most abundant aldehyde, with a mean outdoor concentration of 12.53 ppb (. 60% of the total outdoor aldehyde concentration) and a mean indoor concentration of 54.56 ppb (. 87% of the total indoor aldehyde concentration).

Krzyzanowski et al. (1990) measured formaldehyde concentrations in 202 households in Pima County, Arizona, and found an average value of 26 ppb. Concentrations varied slightly with locations in the house, with the highest levels generally found in the kitchens. Only a few concentrations exceeded 90 ppb, with a maximum value of 140 ppb. The average indoor formaldehyde concentrations measured in homes in Pullman, Washington, ranged from approximately 5–72 ppb (Lamb et al. 1985).

Formaldehyde concentrations measured on two different days inside a tavern during normal smoking conditions were 85–72 ppb (Lofroth et al. 1989). In the same study, the average airborne formaldehyde yield of a cigarette was found to be 2 mg. Levels of formaldehyde in nonsmoking office buildings ranged from not detected to 0.22 ppm, while it ranged from not detected to 0.6 ppm where smoking was permitted (Sterling et al. 1987).

#### 5.4.2 Water

Formaldehyde is unstable in water; however, it has been detected in municipal and industrial aqueous effluents, including those resulting from chemical, oil, and coal processing (EPA 1976b; Hushon et al. 1980). Formaldehyde was not detected in the National Organics Reconnaissance Survey of Suspected Carcinogens in Drinking Water (EPA 1975).

Formaldehyde levels in rainwater collected in California were low, ranging from not detectable to  $0.06~\mu g/mL$  (Grosjean and Wright 1983). Concentrations of free formaldehyde measured in fogwater in Corvallis, Oregon, ranged from 0.4 to 3~mg/L with a volume-weighted mean of 1.8~mg/L (Muir 1991). Free formaldehyde concentrations in fogwater in Riverside, California, ranged from  $4.1~to~228~\mu M$  (0.12-6.8~mg/L), with approximately half of the samples measuring less than  $90~\mu M$  (3~mg/L) (Igawa et al. 1989). Stratus cloudwater at Henninger Flats, California, which typically is highly acidic and concentrated in inorganic pollutants, had concentrations of free formaldehyde ranging from  $45.9~to~61.5~\mu M$  (1.4-1.8~mg/L), comparable to the mid-range of Corvallis fogwater concentrations (Igawa et al. 1989). Formaldehyde concentrations ranging from  $11~to~142~\mu M$  were found in cloudwater samples collected in the Los Angeles Basin (Richards et al. 1983). Formaldehyde concentrations in mist samples in Long Beach and Marina del Ray, California, were  $0.25-0.56~\mu g/mL$ , respectively (Grosjean and Wright 1983).

#### 5.4.3 Sediment and Soil

No data on formaldehyde levels in sediment and soil could be found in the literature.

## 5.4.4 Other Environmental Media

Fresh shrimp from four local commercial markets in Atlanta, Georgia were found to have formaldehyde levels ranging from 0.39 to 1.44 mg/kg (Radford and Dalsis 1982). In the same study, shrimp kept live in the laboratory were found to contain 0.99 mg/kg immediately after sacrifice, and the level rose to 2.15 mg/kg after refrigeration for 6 days.

Chou and Que Hee (1994) measured the concentrations of carbonyl compounds in artificial saliva leachates of three chewing tobaccos. They found formaldehyde concentrations of 110, 670, and 530 ng/mL (0.11, 0.67, and 0.53 µg/mL, respectively) in three different commercial brands. Mansfield et al. (1977) used liquid chromatography to measure formaldehyde as a combustion product in tobacco smoke from six different brands of American filter tip cigarettes. The average amount of formaldehyde by brand ranged from 45.2 to 73.1 µg/cigarette and from 5.1 to 8.9 µg/puff. Triebig and Zober report that the level of formaldehyde in side stream cigarette smoke is fifty times higher than main stream smoke (Triebig and Zober 1984) while the National Research Council put the value at five to eight times more formaldehyde in side stream smoke (NRC 1986).

Formaldehyde has also been found at levels ranging from 1 to 3,517 ppm in 112 fabric samples, with 18 of the samples having a free formaldehyde concentration greater than 750 ppm (mg/kg) (Schorr et al. 1974).

### 5.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

The contribution of various atmospheric environments to the average exposure to formaldehyde is given below (Fishbein 1992):

# FORMALDEHYDE 5. POTENTIAL FOR HUMAN EXPOSURE

Source	mg/day
Air Outdoor air (10% of time)	0.02
Indoor air Home (65% of time) Conventional Prefabricated (chipboard)	0.5–2.0 1.0–10.0
Workplace (25% of time) Without occupational exposure With 1 mg/m³ occupational exposure	0.2 <b>–</b> 0.8 5
Environmental tobacco smoke	0.1–1
Smoking (20 cigarettes/day)	1

The major route of exposure is inhalation of indoor air, with releases of formaldehyde from chipboard and other building and furnishing materials composing the bulk of the exposure. Environmental tobacco smoke contributes 10–25% of the total indoor exposure. Since formaldehyde in food is not available in free form, the intake of formaldehyde from the ingestion of food (1–10 mg/day) has not been included (Fishbein 1992).

Consumers can be exposed to formaldehyde gas through its emission from construction materials, wood products, textiles, home furnishings, paper, cosmetics, and pharmaceuticals (Bartnik et al. 1985; Pickrell et al. 1983; WHO 1989). Members of the general population may also be exposed to formaldehyde by dermal contact with many of these products. Based on a study of dermal absorption of formaldehyde in rats from cosmetic products, Bartnik et al. (1985) made rough estimates of the formaldehyde that could be absorbed through the use of products such as hand cream or sun-tan lotion. In the case of hand cream, approximately 2 g is used per application, containing 2 mg formaldehyde. Assuming 5% absorption, 0.1 mg of formaldehyde would be absorbed. For a whole-body product such as sun-tan lotion, approximately 17 g of the product (containing 17 mg of formaldehyde) would be used, resulting in an absorption of 0.85 mg of formaldehyde.

A study was conducted in Boston to examine the commuter's exposure to six gasoline-related volatile emissions, including formaldehyde, in four different commuting modes (Chan et al. 1991). The mean formaldehyde concentrations (in μg/m³) measured while driving private cars, riding in subways, walking, and biking were 5.1 (n=40), 4.5 (n=38), 5.5 (n=31), and 6.3 (n=11), respectively. The maximum concentrations in all commuting modes were usually three to five times higher than the mean

concentrations. A similar study in the New York/New Jersey area found that the mean in-vehicle concentration of formaldehyde during commutes was  $0.3 \mu g/m^3$  (Lawryk et al. 1995; Lawryk and Weisel 1996).

Occupational exposure can occur during the production of end products in which formaldehyde is used, in the garment industry, during various preservation processes, and in laboratories. Health care professionals (pharmacists, physicians, veterinarians, dentists, nurses, etc.) may be exposed to vapors during the preparation, administration, and/or cleanup of various medicines. Pathologists and histology technicians, morticians, and teachers and students who handle preserved specimens may also be exposed (Fleisher 1987; Holness and Nethercott 1989; Korky et al. 1987; Perkins and Kimbrough 1985; Skisak 1983). The laser cutting of felt, woven fabrics, formica, plexiglass, and acrylic materials has been found to release formaldehyde (Kiefer and Moss 1997). Formaldehyde exposure for workers at a fiberglass insulation manufacturing plant were found to range from 49 to 516 µg/m³ (Milton et al. 1996a). Midrange photocopiers (30–135 copies per minute) have also been found to emit formaldehyde (Leovic et al. 1996).

The National Occupational Exposure Survey (NOES), conducted from 1981 to 1983, indicated that 1,329,332 workers employed in various professions were potentially exposed to formaldehyde in the United States (NIOSH 1995b). The NOES database does not contain information on the frequency, concentration, or duration of exposure; the survey provides only estimates of workers potentially exposed to the chemical in the workplace. OSHA has estimated that in the late 1980s over 2 million workers in over 112,000 firms were exposed to formaldehyde; about 45% of these workers are estimated to be in the garment industry. About 1.9 million were exposed to levels of formaldehyde between 0.1 ppm and 0.5 ppm (mainly in apparel, furniture, paper mills, and plastic molding); approximately 123,000 were exposed to levels of formaldehyde between 0.5 and 0.75 ppm (mainly in apparel, textile finishing, furniture, laboratories, and foundries); and about 84,000 were exposed to between 0.75 and 1 ppm (mainly in apparel, furniture, and foundries) (OSHA 1996).

OSHA has estimated that in the United States approximately 107,000 employees are exposed to formaldehyde concentrations greater than 1 ppm, and approximately 430,000 employees are exposed to concentrations ranging from 0.5 to 1 ppm (Fishbein 1992). An initial evaluation of formaldehyde exposures in a sewing plant using fabrics with a postcure resin found time-weighted average (TWA)

exposure levels #1.2 ppm, with a mean of 0.9 ppm (Luker and Van Houten 1990). A modification that decreased the amount of residual formaldehyde in the fabric reduced worker exposure by 80–85%. Area concentrations of formaldehyde in a plywood company ranged from 0.28 to 3.48 ppm; the average personal exposure was 1.13 ppm formaldehyde (Malaka and Kodama 1990). Airborne formaldehyde concentrations ranging from 0.187 to 0.783 ppm have been measured during particle-board-sanding operations (Stumpf et al. 1986). Formaldehyde concentrations of 0.5–7 ppm have been measured in leather tanning facilities (Stern et al. 1987). An average airborne formaldehyde level of 0.36±0.19 ppm was measured during 22 embalming procedures (Holness and Nethercott 1989). Fire fighters are exposed to formaldehyde concentrations as high as 8 ppm during knockdown (bringing the main body of fire under control) and #0.4 ppm during overhaul (searching for and extinguishing hidden fire) (Jankovic et al. 1991). Formaldehyde levels #0.3 ppm have been measured inside a fire fighter's self-contained breathing apparatus (Jankovic et al. 1991).

The current OSHA permissible exposure limit (PEL) for all workplaces covered by OSHA is 0.75 ppm for an 8-hour TWA, and the short-term exposure limit (STEL) is 2 ppm over a 15-minute period. The "action level" is 0.5 ppm measured over 8 hours (OSHA 1996). NIOSH has recommended a 0.1 ppm ceiling exposure concentration over a 15-minute period in the workplace (NIOSH 1992).

## 5.6 EXPOSURES OF CHILDREN

This section focuses on exposures from conception to maturity at 18 years in humans and briefly considers potential pre-conception exposure to germ cells. Differences from adults in susceptibility to hazardous substances are discussed in Section 2.6 Children's Susceptibility.

Children are not small adults. A child's exposure may differ from an adult's exposure in many ways. Children drink more fluids, eat more food, and breathe more air per kilogram of body weight, and have a larger skin surface in proportion to their body volume. A child's diet often differs from that of adults. The developing human's source of nutrition changes with age: from placental nourishment to breast milk or formula to the diet of older children who eat more of certain types of foods than adults. A child's behavior and lifestyle also influence exposure. Children crawl on the floor; they put things in their mouths; they may ingest inappropriate things such as dirt or paint chips; they spend more time outdoors. Children also are closer to the ground, and they do not have the judgement of adults in avoiding hazards (NRC 1993).

Children are exposed to formaldehyde mainly from its presence in air. Formaldehyde levels are typically higher indoors (Gold et al. 1993) where younger children may spend a significant amount of time. Exposure for children outdoors will be similar to adults. It is likely to be greatest during rush hour commutes (Spicer et al. 1996) or near known sources of formaldehyde release, such as factories using formaldehyde or during the use of products (Shah and Singh 1988) that emit formaldehyde. Children may also be dermally exposed to formaldehyde as a result of its release from permanent press fabrics or its presence in cosmetic products. Only a limited number of studies that address formaldehyde exposure to children or body burden measurements have been identified (Krzyzanowski et al. 1990; Wantke et al. 1996a).

In the home, formaldehyde sources include household chemicals, pressed wood products (especially when new) (EPA 1996), combustion sources (NRC 1986), and some new fabrics (Schorr et al. 1974) and garments. A number of common household products may release formaldehyde to indoor air, including antiseptics, medicines, dish-washing liquids, fabric softeners, shoe-care agents, carpet cleaners, glues, adhesives, and lacquers (Kelly et al. 1996). If children use or play with some of these products, or are present when they are used, additional exposure to formaldehyde may occur. Many cosmetic products contain formaldehyde and some, such as nail polish and nail hardeners, contain high levels (Kelly et al. 1996). If children place these products in their mouth or on their skin, or sniff them, they will be exposed to elevated levels of formaldehyde.

New carpets have been found to release formaldehyde (Hodgson et al. 1993). Older carpets, especially wall-to-wall carpets, have been found to be a sink for formaldehyde if there are other indoor sources (Norback et al. 1995). In addition, some carpet cleaners may contain formaldehyde. Infants may be exposed to formaldehyde while crawling on carpets or on newly cleaned carpets (IARC 1982; WHO 1989). Similarly, young children may be exposed while sitting or playing on indoor carpeting.

Formaldehyde is typically not found in water or soil, and children are not expected to be exposed by these routes. Because it is a gas, formaldehyde is not brought home on a parent's work clothes or tools. Occupants of newly constructed homes, including children, may be exposed to formaldehyde due to its release from pressed wood construction materials (see Section 5.7), a process that slowly decreases with time. As discussed above, formaldehyde is released to indoor air from many sources. Children that live in mobile homes may be exposed to higher levels of formaldehyde compared to those that live in conventional homes because mobile homes have lower air exchange rates. Children that live in

households that have a cigarette, cigar, or pipe smoker will also be exposed to higher levels of formaldehyde due to its presence in side stream smoke (Triebig and Zober 1984).

Formaldehyde is released from many pressed wood products used in the construction of furniture (Konopinski 1983). When placed in a new crib manufactured from these materials, infants may be exposed because of their proximity to the furniture's structural components. Also, small rooms that have new furniture manufactured from pressed wood products installed may have localized, elevated concentrations of formaldehyde because of their low total volume.

Formaldehyde is highly water soluble, very reactive, and is rapidly metabolized by tissues at portals of entry. Thus, parental exposure at typical indoor, outdoor, or occupational levels is not expected to result in exposure to parental germ cells or the developing fetus (See Section 2.6 for additional information).

Since formaldehyde is typically found in the air, and since its concentration in indoor air is typically higher than that found outdoors, formaldehyde exposure to children can be reduced by bringing fresh air into the home. This can be accomplished by opening windows or using ventilation fans. It has been established that coated or laminated pressed wood products release less formaldehyde than those that are uncoated (Kelly et al. 1996; Pickrell et al. 1983), so sealing these surfaces would be expected to reduce formaldehyde levels in the home. Since formaldehyde is also formed in indoor air by the degradation of other volatile organic chemicals, such as solvents (Weschler and Shields 1996), commonly found in the home, removing the source of these materials or using them with adequate ventilation will reduce indoor levels of formaldehyde.

Material that acts as a sink for formaldehyde, such as carpets (Norback et al. 1995), can also be removed from the home to lower levels. Securing cosmetic products and other materials that have higher formaldehyde concentrations away from children's reach and not allowing individuals to smoke in the house will also lower levels of exposure.

#### 5.7 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

Workers in industries where formaldehyde is used or released may receive potentially high exposures. Members of the general population who live in newly constructed homes or homes where pressed wood products have recently been installed may be exposed to high levels of formaldehyde by inhalation for short periods of time until the latent formaldehyde has been released. Exposures in mobile homes are expected to be higher than conventional homes due to their lower rate of air exchange (Wolff 1991). Members of the general population that handle large amounts of permanent press fabrics treated with formaldehyde-releasing resins may also receive potentially high exposures. The use of some cosmetics, such as nail hardeners, may result in high short-term exposure.

Smokers and persons who live in a home with a cigarette smoker also may be exposed to higher levels of formaldehyde. Environmental tobacco smoke, which is a combination of diluted sidestream smoke released from a cigarette's burning end and mainstream smoke exhaled by an active smoker, can contribute 10–25% (0.1–1 mg/day) of the total average indoor exposure to formaldehyde (Fishbein 1992).

#### 5.8 ADEQUACY OF THE DATABASE

Section 104(i)(5) of CERCLA, as amended, directs the Administrator of ATSDR (in consultation with the Administrator of EPA and agencies and programs of the Public Health Service) to assess whether adequate information on the health effects of formaldehyde is available. Where adequate information is not available, ATSDR, in conjunction with the NTP, is required to assure the initiation of a program of research designed to determine the health effects (and techniques for developing methods to determine such health effects) of formaldehyde.

The following categories of possible data needs have been identified by a joint team of scientists from ATSDR, NTP, and EPA. They are defined as substance-specific informational needs that if met would reduce the uncertainties of human health assessment. This definition should not be interpreted to mean that all data needs discussed in this section must be filled. In the future, the identified data needs will be evaluated and prioritized, and a substance-specific research agenda will be proposed.

#### 5.8.1 Identification of Data Needs

**Physical and Chemical Properties.** The physical and chemical properties of formaldehyde are well characterized and allow prediction of the transport and transformation of the compound in the environment.

**Production, Import/Export, Use, Release, and Disposal.** Knowledge of a chemical's production volume is important because it may indicate the magnitude of environmental contamination and human exposure. Data regarding the production, trend, use pattern, and disposal of formaldehyde are available.

According to the Emergency Planning and Community Right-to-Know Act of 1986, 42 U.S.C. Section 11023, industries are required to submit chemical release and off-site transfer information to the EPA. The Toxics Release Inventory (TRI), which contains this information for 1996, became available in May of 1998. According to the Toxics Release Inventory (TRI), in 1996, 21 million pounds (9.6 million kg) of formaldehyde were released to the environment. This database will be updated yearly and should provide a list of industrial production facilities and emissions.

**Environmental Fate.** The environmental fate of formaldehyde in air has been well studied (Bufalini et al. 1979; Calvert et al. 1972; Chameides and Davis 1983; DOT 1980; EPA 1976a; Kao 1994; Kelly et al. 1994; Lowe et al. 1980; Su et al. 1979). Formaldehyde is removed from the atmosphere by direct photolysis and oxidation by photochemically produced hydroxyl and nitrate radicals. Solutions of formaldehyde are unstable, both oxidizing slowly to form formic acid and polymerizing. When released to water, formaldehyde will biodegrade to low levels in a few days, with little adsorption to sediment (Howard 1989; Lyman 1982). No significant volatilization from water is expected to occur (Lyman 1982). No experimental or estimated values for the half-life of formaldehyde in ambient water were found in the literature. Aqueous solutions of formaldehyde that are released to soil may leach through the soil (Lyman 1982; SRC 1995b; Swann et al. 1983). Although formaldehyde is known to biodegrade under both aerobic and anaerobic conditions, the fate of formaldehyde in soil is still unknown (Howard 1989). There is a need for data on the fate and transport of formaldehyde in soil, including half-life values. The environmental fate of formaldehyde's predominant degradation product, formic acid, is well documented. In air, formic acid is expected to rapidly degrade (half-life approximately 1 month) through

its reaction with hydroxyl radicals or it is expected to undergo wet deposition to the earth's surface (HSDB 1999). In water or soil, formic acid is expected to rapidly biodegrade.

Bioavailability from Environmental Media. Inhalation is the major pathway for exposure to formaldehyde (Fishbein 1992). The environmental factors that may influence the bioavailability of formaldehyde from contaminated air, water, or plant material have not been studied. The role of sorption may not be significant in determining the bioavailability of formaldehyde from soil since formaldehyde is not expected to adsorb to soils and sediments (Lyman 1982). The factors affecting the bioavailability of formaldehyde from soil and other environmental media need further investigation. The bioavailability of formaldehyde from the ingestion of food is not known although it is not expected to be significant.

**Food Chain Bioaccumulation.** Experiments performed on a variety of fish and shrimp showed no evidence of the bioaccumulation of formaldehyde (Hose and Lightner 1980; Sills and Allen 1979). Because formaldehyde is rapidly metabolized (Casanova et al. 1988), bioaccumulation is not expected to be important. No further information is needed.

Exposure Levels in Environmental Media. The levels of formaldehyde in air are well documented (EPA 1987d; Gammage and Hawthorne 1985; Gold et al. 1993; Grosjean 1982; Hawthorne et al. 1985, 1986a; Kelly et al. 1994; Krzyzanowski et al. 1990; Lamb et al. 1985; Lofroth et al. 1989; Norsted et al. 1985; Salas and Singh 1986; Schulam et al. 1985; Sexton et al. 1989; Shah and Singh 1988; Singh et al. 1982; Stock 1987; Zhang et al. 1994a). Some data on the concentrations of formaldehyde in rainwater, fogwater, and mist have been reported (Grosjean and Wright 1983; Igawa et al. 1989; Muir 1991), but more recent data on the levels of formaldehyde in water would be useful in assessing exposures. No data on levels of formaldehyde in soil were found in the literature. Soil concentrations also are needed to assess exposures.

Reliable monitoring data for the levels of formaldehyde in contaminated media at hazardous waste sites are needed so that the information obtained on levels of formaldehyde in the environment can be used to assess the potential risk of adverse health effects in populations living in the vicinity of hazardous waste sites.

Exposure Levels in Humans. Data are available concerning exposure levels in occupational settings (Anonymous 1984a; Fishbein 1992; OSHA 1996). Results of the National Occupational Exposure Survey (NIOSH 1995b) are indicative of the number of workers potentially exposed to formaldehyde in various occupations; however, the database does not contain information on the frequency, concentration, or duration of exposure. Available data indicate that the general population may be exposed to formaldehyde through inhalation and dermal contact; however, because of the wide variety of exposure sources (conventional homes versus mobile homes, smoking versus nonsmoking environment, usage patterns for consumer products, etc.), it is difficult to estimate average daily intakes by these routes. Up-to-date comprehensive monitoring data would be useful in determining the typical amount of formaldehyde to which the general population is exposed. This information is necessary for assessing the need to conduct health studies on these populations and persons residing near hazardous waste sites.

**Exposures of Children.** Because formaldehyde is found predominately in the air, the main exposure pathway for children is the same as adults. Formaldehyde concentrations in indoor air have been reported, although many of the studies were obtained before manufacturing methods for formaldehyde-releasing building materials were changed to reduce emissions. Current, comprehensive studies that account for all formaldehyde release sources to indoor air would aid in determining the body burden to children. Studies that take into account the different lifestyles of children would be also helpful. For example, formaldehyde emissions to indoor air may be source dominated (e.g., emission from pressed wood furniture). For a given emission level, the resulting formaldehyde concentration would be expected to be greater in children's rooms compared to those of adults because the volume is smaller. Information to address child specific formaldehyde exposure issues is not available.

**Exposure Registries.** No exposure registries for formaldehyde were located. This substance is not currently one of the compounds for which a subregistry has been established in the National Exposure Registry. The substance will be considered in the future when chemical selection is made for subregistries to be established. The information that is amassed in the National Exposure Registry facilitates the epidemiological research needed to assess adverse health outcomes that may be related to exposure to this substance. Workers in industries where formaldehyde is used or produced should be considered for inclusion in an exposure registry. Given that formaldehyde is routinely found in indoor air samples, members of the general population should also be considered for inclusion in the exposure registry.

# 5.8.2 Ongoing Studies

As part of the Third National Health and Nutrition Evaluation Survey (NHANES III), the Environmental Health Laboratory Sciences Division of the National Center for Environmental Health, Centers for Disease Control and Prevention, will be analyzing human blood samples for formaldehyde and other volatile organic compounds. These data will give an indication of the frequency of occurrence and background levels of these compounds in the general population.

The Federal Research in Progress (FEDRIP 1996) database provides additional information obtainable from a few ongoing studies that may fill in some of the data needs identified in Section 5.8.1. These studies are summarized in Table 5-4.

Table 5-4. Ongoing Studies on the Potential for Human Exposure to Formaldehyde

Investigator	Affiliation	Research description	Sponsor
K. Knapp, D. Pahl, and F. Black	EPA-AREAL Research Triangle Park, NC	Characterization of emissions, including formaldehyde, from motor vehicles using both traditional and alternative fuels.	EPA
B.J. Collier	Louisiana State University Baton Rouge, LA	Development of textile materials for environmental compatibility and human health and safety.	USDA
D.V. Sandberg and R.D. Ottmar	Pacific Northwest Forest and Range Experiment Station Portland, OR	Assessment of firefighter exposures to potential health hazards, including formaldehyde.	USDA
T. Shibamoto	University of California Davis, CA	Isolation and identification of mutagens and carcinogens in foods.	USDA
J.S. Gaffney	Argonne National Laboratory Argonne, IL	Atmospheric chemistry of organic oxidants and aldehydes.	USDOE
P. Davidovits	Boston College Chestnut Hill, MA	Laboratory studies of heterogeneous gas- liquid interaction of atmospheric trace gases, including formaldehyde.	NSF
R.C. Bales and M. Conklin	University of Arizona Tucson, AZ	Determination of the atmosphere-to-snow transfer function for peroxide and formaldehyde, whose deposition is reversible.	NSF
R.C. Bales and M. Conklin	University of Arizona Tucson, AZ	Distribution of reactive chemical species, including formaldehyde, in ice and snow.	NSF
V.A. Mohnen	State University of New York Albany, NY	Examination of the involvement of natural hydrocarbons for the formation of peroxide and formaldehyde in air.	NSF
B.G. Heikes	University of Rhode Island Kingston, RI	Background atmospheric measurements of hydrogen peroxide, organic hydro-peroxides, and formaldehyde.	NSF
S.S. Que Hee	UCLA School of Public Health Los Angeles , CA	Carbonyl compounds air sampling method.	NIOSH
R.R. Fall	University of Colorado at Boulder Boulder, CO	Biogenic sources of oxygenated hydrocarbons in the troposphere.	NSF

EPA = Environmental Protection Agency; NSF = National Science Foundation; USDA = United States Department of Agriculture; USDOE = United States Department of Energy; NIOSH = National Institute for Occupational Safety and Health