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Available Data on Naphthalene Exposures: Strengths and Limitations

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Purpose of this Document

Naphthalene has been the subject of a number of exposure and risk assessments over the last 26 years (EPA, 1980, 1982, 1984, 1988a,b, 2002; ATSDR, 2005, and Preuss et al., 2003). The purpose of this document is to summarize the findings of these studies and other recent publications and present a description of what is known and unknown about human exposure to naphthalene. The exposures investigated in this report are tailored to the toxicological issue that is the focus of the conference. That issue is, "What are the risks associated to chronic inhalation exposures to naphthalene?" As a result this report will focus on chronic inhalation exposure to U.S. populations and give less attention to dermal and oral exposures and acute inhalation exposures. The assessment will consider occupational exposures and exposure from environmentally mediated sources, indoor sources, and consumer products[1].

The resources available for the preparation of this report have precluded performing an in-depth analysis of the available data in each of the studies cited by ATSDR, 2005 and Preuss et al 2003 or the more recent publications. The approach used in this report is to use the readily available data to characterize the sources of exposure that affect the general U.S. population and then to evaluate the sources that affect specific sub-populations. This approach is used in the Voluntary Children's Chemical Evaluation Program (VCCEP) for the chemicals acetone, benzene, and xylenes (ACC, 2003, 2005; 2006). Within this framework the available data will be evaluated and where possible, conclusions will be generated on the range of chronic exposures to naphthalene that occur in the general populations and the sub-populations. The limitations of the data will be discussed and areas where additional research could be performed will be identified.

Naphthalene's Relevant Chemical Properties

Naphthalene is a white solid that readily sublimes at room temperature. Its chemical structure consists of two fused benzene rings. Naphthalene is the simplest member of the poly aromatic hydrocarbons (PAHs). A summary of the compound's properties are given in Table 1. As these properties indicate, the compound is a semivolatile. In most indoor and outdoor environments the compound is encountered as a vapor. In cigarette smoke the compound is found in the particulate phase. The compound has a saturated air concentration of 600,000 ug/m^3; however, at higher concentrations the compound tends to partition to surfaces.

Naphthalene is rapidly removed from air by reaction with hydroxyl radicals. The half life in air is 8 h (Bernhard and Simonich, 2000; Lu et al., 2005). Naphthalene has a short half life in surface waters and soils due to biodegradation and photolysis (ATSDR, 2005).

[1] This report does not review abuse of naphthalene such as sniffing or ingesting mothballs.

Table 1. Select Properties of Naphthalene	
Molecular weight	128.19 g/mole
Color	White
Physical state	Solid
Melting point	80.5 °C
Boiling point	218 °C
Density at 20 °C	1.145 g/mL
Odor:	Strong (tar or mothballs)
Odor threshold:	
Water	0.021 mg/L
Air	440 ug/m^3
Solubility:	
Water at 25 °C	31.7 mg/L
Organic Solvents	Soluble
Partition coefficients:	
Log Kow	3.29
Log Koc	2.97
Vapor Pressure	0.087 mmHg
	ATSDR, 2005

Naphthalene has been found to occur in ground water under anaerobic conditions where there is an on-going source such as the presence of petroleum products in the aquifer. Naphthalene does not bioaccumulate to any extent due to a relatively low Kow value and the ability of organisms to metabolize the compound. Naphthalene is found in certain foods at low levels and higher levels can occur in foods that are smoked or charbroiled. Because of these characteristics, inhalation exposure is the dominant route of exposure for naphthalene (EPA, 2002) and exposures occur at locations that are proximate to the sources of release in both time and space.

Sources of Naphthalene Exposures

Naphthalene is a product of incomplete combustion of biomass and petroleum products Preuss et al., 2003, Schauer 2001. The compound is also found in cigarette smoke and is generated during the grilling or charbroiling of meats (ATSDR, 2005). Naphthalene is also found in a number of commercial and consumer products. The following table presents the levels of naphthalene in petroleum products. As the table indicates, the naphthalene occurs in at levels of roughly 0.25% in many petroleum products and at higher levels in jet fuels. Naphthalene is also a component of asphalt (ATSDR, 2005). Use of asphalt on roads results in long term sources of release for naphthalene (Lu et al. 2005).

Table 2. Concentration of Naphthalene In Petroleum Products		
Petroleum Product	**Range in Weight %**	**Average Weight %**
Gasoline	0.15-0.36	0.25
Diesel Fuels (#2)	0.01-0.80	0.26
Kerosene	0.15-0.46	0.31
Jet Fuel-4	0.25-0.50	0.25
Jet Fuel-5	*	0.57
Jet Fuel-7	*	0.72
Jet Fuel-8	*	1.1
No. 2 Fuel Oil	0.009-0.40	0.22
No. 6 Fuel Oil	0.0021-0.015	0.0042
Lubricating and Motor Oils	0.00005-0.25	0.059
*Single data point		Potter and Simmons,1998

Table 3 presents a summary of information on naphthalene in consumer products. The products fall into two categories. The first group includes products where naphthalene is a component of a hydrocarbon mixture used as a fuel additive or as a carrier in pesticides. These products would not be anticipated to contribute significantly to indoor air levels.

Table 3. Levels of Naphthalene in Various Consumer Products			
Brand	**Category**	**Form**	**Percent**
STP Gas Treatment	Auto products	liquid	1-2
STP Octane Performance Booster	Auto products	liquid	1-2
STP Oxygenated Gas Treatment	Auto products	liquid	0-3
STP Super Concentrated Fuel Injector Cleaner	Auto products	liquid	0-2
STP Gas Treatment-08/03/2000	Auto products	liquid	0-2
STP Complete Fuel System Cleaner	Auto products	liquid	21-42
STP Fuel Injector/Carburetor Cleaner	Auto products	liquid	0-2
STP Fuel Injector/Carburetor Cleaner-08/07/1998	Auto products	liquid	0-2
STP Gas Treatment-09/30/1998	Auto products	liquid	0-2
STP Octane Performance Booster-11/13/1998	Auto products	liquid	0-2
STP Oxygenated Gas Treatment-12/24/1998	Auto products	liquid	1-2
STP Super Concentrated Intake Valve Cleaner	Auto products	liquid	0-2
STP Fuel Injector/Carburetor Cleaner-01/18/2000	Auto products	liquid	0-2
STP Fuel Injector and Carburetor Treatment 12 Fl. Oz.	Auto products	liquid	0-2
Spectracide Brush Killer Concentrate	Pesticides	liquid	0.7
Enoz Old Fashioned Mothballs	Pesticides	solid	99.9
Repel Pet and Stray Repellent	Pesticides	powder	16.0
Snake Away	Pesticides	granules	7.0
Bonide Mosquito Beater Granules	Pesticides	granules	4.50
Bonide Shotgun Rabbit & Dog Repellent	Pesticides	powder	4.50
Monsanto Lasso Herbicide (agricultural)	Pesticides	liquid	0.09-0.15
NLM Household Products Database and packaging information for specific products			

The second group is the pesticide products in which naphthalene is an active ingredient to control insects that damage clothing or to repel pests. In addition, the Internet is replete with guidance recommending the use of naphthalene mothballs to repel skunks and raccoons from attics and gardens[2].

Naphthalene is released into the air from a number of industries including coking, petroleum refining, aluminum production, wood treatment (creosote), industries that use asphalt (paving and roofing), and industries using petroleum products in particular jet fuels.

Measurement of Naphthalene in Air

ATSDR, 2005 and Preuss et al., 2003 have summarized the majority of air data available before 2003. In this report we have taken the data and organized the information into the following framework.

Ambient levels:

- Naturally occurring levels (pristine areas);
- Levels in suburban/urban areas

Indoor air levels:

- Homes
- Homes with naphthalene products.

Occupational Exposure Levels:

Low exposure industries

- Chemical industry
- Refineries
- Asphalt
- Aluminum production

High exposure industries

- Wood treatment (creosote)
- Coke production
- Jet fuel related

For each of these categories a range for the long term average levels is proposed based on available data. This range is intended to describe both the uncertainty and the interindividual variation in the long term average concentrations to which individuals in the relevant populations are exposed. The range is described using a log scale of concentration in units of ½ of log 10 ug/m^3 (1, 3, 10, 3, 100, …). The goal in this effort is capture the sense of the available data and to place the data in an order of magnitude framework to demonstrate each source of exposure's relative importance. As new data are developed these ranges may change.

[2]See http://www.ghorganics.com/page6.html
http://www.canr.uconn.edu/ces/ipm/homegrnd/htms/8racoon.htm
http://solutionsforyourlife.ufl.edu/hot_topics/environment/nuisance_wildlife.html

Levels of Naphthalene in Pristine Air

A number of studies have reported naphthalene levels in locations other than urban areas. Naphthalene is expected to occur at low levels in pristine air because of natural sources (biomass burning). Based on data from White and Hardy (1994) and Hoff and Chan (1987) a range of 0.0001 to 0.003 is proposed for a long term average concentration in pristine air.

Levels of Naphthalene in Suburban and Urban Air

Studies of Naphthalene in urban air have reported that levels have high spatial and temporal variability (Reisen, 2003 and Reisen and Arey, 2005). Elevated levels are directly related to traffic with the highest levels being reported in highway tunnels and areas with high vehicular traffic (Lu et al., 2005; ATSDR, 2005). Most of the data in the literature on levels in the U.S. predate current controls on vehicles and gasoline reformulation and may be overestimates of current exposures. Lu et al., reports modeling of naphthalene levels in the Los Angeles basin that range from 0.0 to 0.5 ug/m^3 in the summer and 0 - 1 ug/ m^3 in winter. These predictions were shown to be reasonably consistent with recent measurements. Because of the limited relevant monitoring data for areas outside of Southern California, it is difficult to provide an estimate of current range of annual averages of naphthalene in the suburban and urban areas. However, because Southern California is known to be representative of urban areas with air pollution problems, the levels reported by Lu et al. can be taken as a conservative estimate for the U.S. urban air. A range of 0.001 to 1 ug/m^3 is proposed for levels in suburban and urban air.

Levels of Naphthalene in Vehicles

Naphthalene levels in vehicles can be elevated by the vehicle’s or other vehicles' exhaust entering the cabin. Lu et al., 2005 cites Batterman et al., (2002) that naphthalene concentration of 1.2 ug/m3 in buses in Detroit, Michigan. ATSDR cites an average level from Lofgren et al., 1991 of 4.5 ug/m^3 (ATSDR, 2005). Lu et al. has suggested that based upon data on the elevation of benzene and other aromatics in vehicles, vehicle air levels for naphthalene be set at three times the ambient levels. Based on this recommendation, vehicle levels are estimated to range from 0.003 to 3 ug/m^3.

Levels of Naphthalene in Indoor Air

Very limited data are available on naphthalene in indoor air in the US. The data are at most 24-h average samples and may not be reflective of long term average exposures. However, the data clearly indicate that residential indoor air levels are higher than ambient levels. This finding is reasonable since a number of activities can result in releases of naphthalene to the indoor environment. These include:

- Smoking;
- Kerosene space heaters;
- Wood stoves;

- Emissions from vehicles and stored petroleum products in attached garages;
- Cooking; and
- Use of products containing naphthalene (mothballs).

The available data suggest that indoor air levels are likely to fall into the range of 0.1 to 10 ppm. Appendix A of this report presents a one-compartment model of a residence. This model demonstrates that modest releases from these sources of naphthalene are capable of causing levels in this range.

Levels of Naphthalene in Indoor Air from Use of Naphthalene Products

The list of products in Table 3 are all intended to be used in a garage or outside of the home with the exception of products designed to control insect damage (mothballs). Mothballs are designed to be used in enclosed spaces (boxes, bags, or closets) and in these closed spaces the vapor builds up and suppresses insects. However such containers are not vapor-proof and act as continuous sources of naphthalene in indoor air. Zhu et al., 2003 reported that levels of 0.3 to 13 ug/m^3 in bedrooms adjacent to closets containing mothballs. Such products can also cause exposures by elevating levels in the individuals' breathing zone when wearing clothing or using bedding that was treated with mothballs. The levels of exposures from these sources are not well documented.

There is also evidence of instances where mothballs are used as area fumigants to control a wide range of pests. Such off-label uses have the potential to cause higher exposures since large amounts are placed on open trays in attics or other portions of the home. ATSDR cites Linick (1983) as reporting a measurement of 100 ug/m^3 in one home where mothballs were misused. This level was measured after remediation and likely underestimates actual exposures. Based on the available data and the estimates provide in Appendix A, homes using mothballs are estimated to have long term levels that range from 1 to 100 ug/m^3 for typical use and 10 to 300 ug/m^3 for misuse scenarios.

Levels of Naphthalene in Other Indoor Environments

Data on naphthalene in day care centers were reported by Wilson et al., 1999. The mean level measure in 9 day care samples was 0.2 ug/m^3. No other data were identified for other indoor environments. Because of this limited data no estimates were made for other indoor environments.

Occupational Levels of Naphthalene

Elevated levels of naphthalene in the workplace have been shown to occur in a number of industries. In general the amount and quality of data in various industries is not large and varies in quality. In many instances the data are based on short term grab samples.

The level of exposures varies greatly from industry to industry. Preuss et al.,2005 reported that 3 hr grab samples in coking and coal tar industries ranged from <1 to 703 ug/m^3, with higher levels occurring in the coal tar and coking industries than in the product of refractory materials or graphite electrodes. Data on exposure to naphthalene in military personnel exposure to jet fuel was reported by Egeghty et al., 2003. Levels for task involving a high potential for exposure ranged from 12 to 3,900 ug/m^3. Preuss et al., 2003 presented a summary table for occupation exposure information in the U.S. and Europe.

Based on these data the daily exposures to naphthalene fall in to two ranges. For individuals in the following industries, daily exposures appear to fall in the range of 10 to 300 ug/m^3.

- Refining and petroleum industries;
- Asphalt industries paving and roofing;
- Industries that use pitch to manufacture refractory materials or graphite electrodes;

Levels in the following industries' daily exposures fall into a higher range of 100 to 3,000.

- Creosote production and use;
- Workers exposed to jet fuels;
- Coal tar and coke industries;
- Production of naphthalene from coal tar;
- Production of mothballs; and
- Chemical industries that use naphthalene as a raw material.

Summary of Existing Data and General Conclusions

Figure 1 presents a summary of the levels of naphthalene that the general U.S. population experiences outdoors, indoors, and in vehicles and the levels experienced by specific sub-populations. The first four categories in the chart affect the general population. The remaining categories present levels that affect only a small portion of the U.S. population. Figure 1 intentionally uses a symbol of a cloud to emphasize the uncertainty in the range of levels for the different sources and the tentative nature of this assessment.

While there are a number of significant limitations to the existing naphthalene data, a number of general observations can be made on the patterns of exposure to naphthalene in the U.S. population.

1. Background exposures to naphthalene for most individuals will be a function of both outdoor and indoor exposures with indoor sources making a greater contribution for at least some individuals.
2. Mothballs are a poorly characterized but significant source of exposure. This source has the greatest potential to result in exposures greater than 10 ug/m^3 for children and for adults outside of the workplace.
3. Occupation exposures will be the dominate source of exposure for workers in the affected industries.

Cigarettes and Naphthalene Exposure

Direct exposure to naphthalene occurs from the use of tobacco products. Estimates of the inhalation dose per cigarette are reported to range from 0.3 to 4 ug (Preuss et al., 2003). For a smoker consuming a pack a day this would correspond to a dose of 6-80 ug. A 24 hour exposure to air levels of 0.3 ug/m^3 to 4 ug/m^3 would produce a similar dose range in adults. This suggests the cigarette smokers receive doses that are similar in magnitude to the doses that they receive from both outdoor and residential exposures. While no studies have been made for naphthalene, Wallace (1996) concluded that the dominant source of exposure to benzene for smokers is direct exposure rather than environmental tobacco smoke. A similar finding would be expected for naphthalene.

Potential Areas for Exposure Research

Data on naphthalene are limited. Prior to the findings in 2000 of carcinogenic activity in rodents, naphthalene was regarded as a widespread contaminant in air that was of minimal concern. As a result naphthalene was rarely the focus of specific studies. Naphthalene was included in surveys as a compound that was detectable with broad spectrum analytical techniques, as one of the components of petroleum products, or as one of the PAHs. In many instances data collected on naphthalene were not thoroughly analyzed and were included only for the sake of completeness[3]. This has led to a relatively small amount of information on the compound in the published literature and few systematic attempts to evaluate the range and variation in chronic exposures in the U.S. populations.

Second, naphthalene levels are highly variable over time and space. Variation in sampling protocols (duration of the sampling, exact location in the home or in the city where the air sample is taken and the time of day the sample is taken) can influence the findings of a survey. The air monitoring data for naphthalene generally consists of grab samples of varying durations collected in different countries using different analytical methods.

Third, the ranges of values reported by individual studies are not likely to be representative of the ranges of long term average exposures of individuals in the sampled environments. The reason for this is that variation in long term average exposures are driven by factors such as variation in duties over a carrier in an industry, job tenure, population mobility, and long term patterns in consumer behavior that are not captured by short term samples. For these reasons the range and variation in the reported measurements should not be used as an indication of the range of chronic exposures to naphthalene in the general population or in any of the sub populations.

A final problem with the data on suburban and urban levels of naphthalene is that older studies are likely to overestimate current exposures. Air levels in urban and suburban

[3] See the lack of analysis of the naphthalene data in Chuang et al., (1999) and Wilson et al., (1999).

areas are believed to be driven in large part by vehicle emissions. Naphthalene as an aromatic contributes to photochemical smog and ozone formation. Programs to reduce emissions of aromatics such as the new vehicle standards and use of reformulated gasoline in many cities have resulted in a reduction in aromatics such as benzene and naphthalene levels in the last 10 years (ACC 2006; Lu et al., 2005). Evidence of the impact of such programs can be seen in Shauer et al., 2002 who reported that current catalytic converters decreased levels of naphthalene by a factor of 50 in automobile exhaust. Because of these changes, levels of naphthalene reported before 2000 may not be representative of current U.S. levels (Lu et al., 2005).

Because of these issues the following areas for exposure research are suggested:

1. Identify, collect, and organize existing data on naphthalene. These data should be used to confirm where data gaps exist for the compound and how the future studies, surveys, or modeling projects should be designed.
2. Perform surveys of levels in major urban areas to establish what are the current levels of exposure for the U.S. population.
3. Perform surveys of use of consumer products containing naphthalene and cooking and heating practices that release naphthalene.
4. Include naphthalene in future surveys of indoor air pollutants. Develop targeted survey for homes that use naphthalene containing products.
5. Consider the development of chronic residential exposure models based on the models used for residential pesticides.
6. Perform additional occupational monitoring surveys for naphthalene related industries.

The importance of the specific research projects will be dependent on the hazard finding for the compound. If chronic exposures to levels below 10 ug/m^3 are deemed to be of low concern then exposures from outdoor sources may be of low concern.

The available exposure data also suggest that the following considerations be made in assessing risks from naphthalene. First, when evaluating the risks associated with naphthalene it should be recognized that exposure to naphthalene from smoking may be different than other exposures, since naphthalene in cigarette smoke is reported to be bound to particles and not in the form of vapor. Second, biomonitoring studies need to recognize that dermal and oral sources of naphthalene may significantly contribute to body burdens in individuals with low inhalation exposures. Third, exposures to environmental sources of naphthalene are variable over a day and may have a strong seasonal variation. Low level dose-response models may need to consider this variation.

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Appendix A. Modeling Residential Levels of Naphthalene in Indoor Air

Approximate long term average indoor air levels can be estimated based on a few housing characteristics and the source strength (Jayjock et al., 2006). The relationship between the mass of a compound released to indoor air can be approximated by assuming that the home can be modeled as a single compartment. Using this assumption and for compounds where their removal rate is determined by the residential air exchange the indoor air concentration can be determined by the following equation:

Indoor Air Concentrations = Average Outdoor Air Concentration + S/(Q*V*24)

where S is the mass of the compound emitted into the residences air from a source in a day, Q is the hourly air exchange rate, and V is the volume house.

In this assessment the value of Q and V are taken from EPA's Exposure Factors Handbook estimates for the average U.S. residence.
$Q = 0.63\ h^{-1}$
$V = 369\ m^3$

Using this model the amount of naphthalene required to increase indoor air concentrations by various amounts can be calculated. The following table presents the amounts associated with different indoor air concentrations.

Naphthalene Source Strength and Average Indoor Air Levels			
Outdoor Air Concentration ug/m3	**Indoor Air Concentrations ug/m3**	**Source ug/d**	**Source g/yr**
0.2	0.3	600	0.2
0.2	0.5	1700	0.6
0.2	1	4400	1.6
0.2	3	15000	5.6
0.2	10	54000	20
0.2	30	160000	60
0.2	100	550000	200
0.2	200	550000	400

A single box of moth balls (Enoz Old Fashioned Mothballs) contains 396 g of naphthalene. Thus one box of mothballs is sufficient to raise the indoor air concentration to 200 ug/m^3 in a residence for the duration of a year. In smaller homes or homes with lower air exchange rates the amounts required would be less.

Figure 1. General Ranges of Naphthalene Air Concentrations in Different Populations