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Hazard Assessment of Chemical Air Contaminants Measured in Residences

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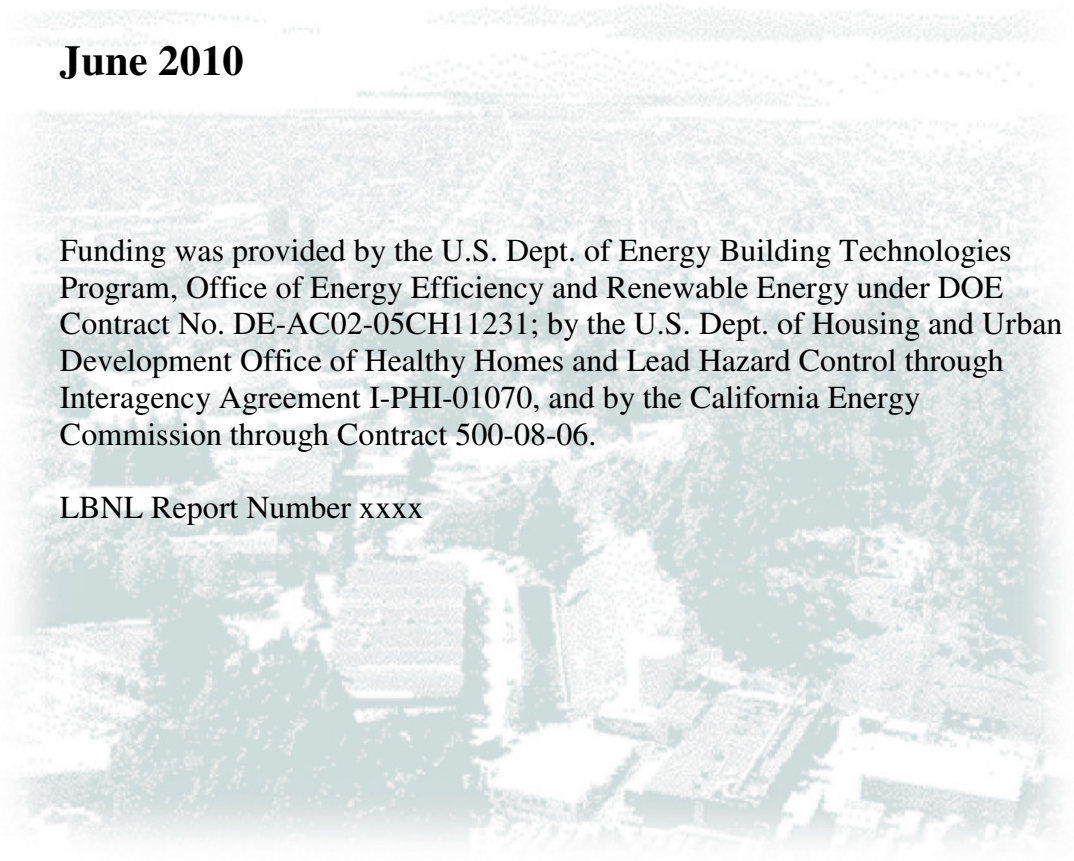
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Abstract

Identifying air pollutants that pose a potential hazard indoors can facilitate exposure mitigation. In this study, we compiled summary results from 88 published studies reporting measurements of chemical pollutants in residences. These data were used to calculate representative mid-range and upper bound concentrations relevant to chronic exposures for 321 pollutants and representative peak concentrations relevant to acute exposures for 5 activity-associated pollutants. Representative concentrations are compared to available chronic and acute health standards for 108 pollutants. Fifteen pollutants are identified as contaminants of concern for chronic health effects in a large fraction of homes. Nine pollutants are identified as potential chronic health hazards in a substantial minority of homes and an additional nine are identified as potential hazards in a very small percentage of homes. Nine pollutants are identified as priority hazards based on robustness of reported concentration data and fraction of residences that appear to be impacted: acetaldehyde; acrolein; benzene; 1,3-butadiene; 1,4-dichlorobenzene; formaldehyde; naphthalene; nitrogen dioxide; and PM_{2.5}. Activity-based emissions are shown to pose potential acute health hazards for PM_{2.5}, formaldehyde, CO, chloroform, and NO₂.

Introduction

The importance of the residential environment to cumulative air pollutant exposures has been demonstrated in numerous studies (Edwards et al. 2001; Weisel et al. 2005). As outdoor air pollutant concentrations decrease and residential air exchange rates are lowered with improved air tightness (Sherman et al. 2002), the contribution of indoor pollutant sources to overall exposure is expected to become increasingly more significant.

The management and mitigation of health risks and disease burden associated with indoor air pollutant exposures can be advanced using the environmental health approaches of hazard analysis and risk assessment. Hazard analysis is a binary identification of pollutants that may cause harm under some prevailing conditions. Risk assessment attempts to quantify the probability and/or extent of harm that would be caused under a given set of conditions. Identified contaminants of concern can be managed in many ways including reducing emission sources and designing ventilation systems to achieve dilution and removal so as to maintain concentrations below harmful levels. The American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) Standard 62.1 allows this latter approach as an alternative to prescriptive ventilation rates designed to achieve acceptable indoor air quality in many buildings (ASHRAE 2007). Sherman and Hodgson (2004) suggested that residential ventilation rates should be set to reduce formaldehyde concentrations below hazardous levels. Two recent studies examined pollutants posing chronic health hazards in residences. Levin and Hodgson (2003) and Dawson and McAlary (2009) identified volatile organic compounds (VOCs) that potentially pose an elevated cancer and non-cancer risk respectively by comparing concentrations to published health standards. As part of a broad examination of semi-volatile organic compounds (SVOCs) in indoor environments, Weschler and Nazaroff (2008) reviewed available data on residential concentrations (air and surface) of these chemicals. Mendell (2007) reviewed 21 epidemiological studies to identify pollutants and common household items that are potential indoor-risk drivers.

This paper presents the results of a hazard analysis designed to identify chronic and acute chemical contaminants of concern in U.S. residences. We undertook a literature review to identify and compile data on measured pollutant concentrations for volatile organic compounds (VOCs), semi-volatile organic compounds (SVOCs), metals, and criteria pollutants. From these data, we determined broadly representative mid-range and upper-bound concentrations relevant to assessing chronic pollutant exposures. We also compiled elevated short-term and peak concentrations resulting from episodic activities. These concentrations were compared to chronic and acute health guidelines or standards set by various agencies including the U.S. Environmental Protection Agency (USEPA), the World Health Organization (WHO), and the California Environmental Protection Agency (CalEPA). This analysis yielded a list of acute and chronic health hazards that may be used as a foundation for ongoing residential indoor air quality management efforts.

Approach

Literature Review to Identify Residential Measurements

The initial step of this work was a review of recent studies reporting measurements of pollutant concentrations in residences. The review was focused on the U.S. but also covered data from

other industrialized countries. The review focused first on studies that measured pollutant concentrations relevant to chronic exposures. Many studies reported results from integrated samples collected over periods of 24 hours or more in occupied homes. Some reported concentrations measured over shorter periods in homes that were unoccupied or measured during periods when no substantial pollutant-generating activities were occurring. A second set of studies was identified to obtain data on elevated short-term and peak concentrations resulting from pollutant generating activities. These data included time-resolved or short-term sampling at times and/or for rooms in which pollutant generating activities were occurring. The activities were in some cases scripted and in some cases occupant initiated.

We used the ISI web of knowledge database as our main search engine. We also reviewed proceedings from the 2009 Healthy Building Conference held in Syracuse, NY and the 2008 Indoor Air conference held in Lyngby, Denmark; we scanned titles and abstracts from the conferences for relevance. We conducted the search based on pollutant search terms, ignoring data from developing countries. This search yielded 86 articles that were relevant to acute and chronic exposure in residences.

Our review considered all chemical contaminants measured in residential air regardless of source. The contaminants considered thus include some emitted purely from indoor sources, some that enter predominantly from outdoors, and some having both indoor and outdoor sources. Table 1 lists the reference, study location, pollutant measurements, sample period, and pollutant classes measured in each of the studies with chronic-exposure relevant concentration data. Much of the data applicable to chronic hazard assessment were collected during large exposure studies. The studies were of occupied homes and generally designed to avoid extreme emission sources. Sixty-seven studies used sampling durations on the order of one or more days. Eight studies used shorter sample durations but took steps to reduce the impact of any recent pollutant-generating occupant activities. As an example, in a study comparing VOCs in homes using fuel oil versus control homes, the New York State Department of Health (2006) measured concentrations over a two-hour period. To compensate for the short measurement time, the study was limited to homes that did not regularly use and had not recently used VOC containing products.

Studies with data relevant to assessing short-term peak concentrations and acute exposures are listed in Table 2. These studies reported concentrations measured during scripted events or during occupant activities such as cooking or cleaning that happened to occur during sampling. The reported concentrations were either calculated from time-resolved measurement or from short duration integrated samples collected with the express intent of measuring air quality following specific events or activities. These sampling periods tended to be on the order of a few hours, however some studies reported peak concentrations from highly time resolved data.

Data Compilation

Of the articles collected in the initial screening, 75 reported data relevant to chronic exposure. Based on these 75 reports, we compiled a database of summary statistics for chronic-exposure relevant concentrations for SVOCs, VOCs, metals, and criteria pollutants. From this database, we calculated weighted summary statistics for each pollutant. When calculating summary

statistics, we weighted statistics from individual studies by the number of unique measurements in each study. Typically this was the number of homes in which measurements were made, though some studies included repeat measurements for some homes. This approach was used in a previous compilation effort (Dawson et al. 2009). Results include the total number of studies measuring the pollutant; the total number of unique measurements of a pollutant across all studies and weighted arithmetic mean, 25th, 50th, 75th, and 95th percentile values. A complete set of summary statistics is available for each of the criteria pollutants. Available data for VOCs varied from compound to compound. Each VOC listed has at least one study with mean or median values reported. Benzene was measured in more studies (15) than any other VOC. Fewer data were found for SVOCs. Naphthalene was reported in nine studies, but for some of the SVOCs only a Top of Range, TOR, value was reported. Since SVOC data are so limited, TOR values are included in the data summary.

We used the database and summary statistics to determine representative mid-range and upper-bound concentrations relevant to chronic exposures. When sufficient data were available to calculate a weighted median concentration, we used that value as the representative mid-range concentration. If not, we used the weighted mean value. The upper bound representative concentration for each pollutant is based on the highest concentration for which a summary statistic was available. For most compounds, this was the 95th percentile concentration. This statistic was not used when one of two situations applied: (1) when, owing to variations in reporting and in the values measured in different studies, the 95th percentile concentration was lower than one or more other summary statistics, or (2) none of the studies reporting data for a given compound included a 95th percentile value. Using the summary statistic with the highest concentration also leads to a more conservative selection of chemical contaminants of concern. We used the 95th percentile value as a representative upper bound for all the criteria pollutants, for 72 of the 83 VOCs with available 95th percentile values, and for both of the SVOCs with available 95th percentile values. For the remaining compounds we set the representative upper-bound value to the highest weighted statistic. For some of the SVOCs, only a TOR value is available, and in these cases we set the representative upper bound value to the TOR value and did not define a representative mid-range value.

Hazard Assessment

We completed the hazard assessment by comparing the compiled summary statistics for representative mid-range and upper-bound chronic-relevant concentrations to available chronic and the activity-associated short-term concentrations to acute health standards. Various governmental organizations publish standards or guidelines that specify either safe or hazardous pollutant concentrations for chronic and acute exposures. Such standards or guidelines are available for diverse sets of chemicals including criteria pollutants, hazardous air pollutants, and toxic air contaminants. Chronic health issues can take a lifetime to manifest and published health standards for chronic exposure are established to protect people exposed continuously for years to decades and up to a life-time. Health standards for acute exposures are typically specified for averaging times of 1 h to 1 day but can include levels above which even shorter exposure may be hazardous. Tables S8-S11 in the online supplemental list the health standards we used in the hazard assessment.

The U.S. Environmental Protection Agency (USEPA) sets National Ambient Air Quality Standards (NAAQS) for six criteria pollutants specified in the 1970 Clean Air Act: carbon monoxide (CO), nitrogen dioxide (NO₂), ozone, particulate matter (PM), lead, and sulfur dioxide. The standards are set to protect the most sensitive subset of the population. Several of these standards have been tightened since their inception in the 1970s. The USEPA has recently adopted a 1 hour NO₂ standard of 0.1 ppm (190 µg m⁻³) (USEPA 2010). For some criteria pollutants, the California Environmental Protection Agency (CalEPA) has set standards that are more stringent than USEPA standards. Many governmental bodies outside of the U.S. promulgate standards for the same pollutants. The World Health Organization (WHO) tends to publish the most health-protective standards (WHO 2005), but unlike USEPA standards, these are recommendations or goals rather than legally mandated targets.

Title III of the 1990 Clean Air Act Amendments established a new regulatory category for chemical air contaminants that are known or suspected to cause serious health effects; 189 chemicals were named to the initial list of hazardous air pollutants (HAPs, also called “air toxics”), of which 187 are still on the list. The USEPA is charged to maintain and update this list, which includes VOCs, SVOCs, metals, and polycyclic organic matter (POM). The CalEPA maintains a separate list of toxic air pollutants referred to as Toxic Air Contaminants (TACs). There is considerable overlap between the CalEPA TAC the USEPA HAP lists, but there are some key differences. For a subset of these pollutants the USEPA has listed chronic non-cancer reference concentrations (*RfCs*) and cancer unit risk estimates (*UREs*) through its Integrated Risk Information System (IRIS) and Health Effects Assessment Summary Tables (HEAST). Non-cancer *RfCs* report the exposure concentrations that are assumed to represent a safe level in that they are unlikely to cause health effects even for sensitive subgroups of the population. *UREs* estimate the incremental increase in cancer risk that accrues for each 1 µg m⁻³ increase in chronic exposure. The California Office of Environmental Health Hazard Assessment (OEHHA) publishes non-cancer Reference Exposure Levels (*RELs*) and its own cancer *UREs*. In addition to the California and USEPA values, the U.S. Occupational Safety and Health Administration (OSHA) sets reference concentrations for workplace exposures, and the Agency for Toxic Substances and Disease Registry (ATSDR) publishes *RfCs* for chronic exposure. Since OSHA regulations are intended to protect generally healthy adult workers, their allowable concentrations tend to be higher than those set for HAPs/TACs by the USEPA and CalEPA.

Whereas exposure concentration limits are specified for acute effects and for chronic non-cancer endpoints, concentration-based standards are not uniformly available for cancer. The European Union and the CalEPA have estimated no-effect concentration levels based on an acceptable level of risk. The USEPA has not defined a generally acceptable cancer risk level for HAPs. However, a case-specific determination was made in the 1989 Benzene National Emission Standard for Hazardous Air Pollutants (NESHAP). This rule set an upper limit of acceptability of 1 in 10⁴ lifetime cancer risk for highly exposed individuals and the goal of reducing lifetime risk to 1 in 10⁶ for the general public. In consideration of this range, we used available cancer *UREs* to calculate acceptable exposure concentration for cancer risk that correspond to a lifetime incremental risk of 1 in 10⁵ assuming 70 years of continuous exposure. The resulting cancer-based exposure concentration values are health protective and comparable to but not necessarily equivalent to the *RfCs*.

In addition to the chemical pollutants that have available health-based concentration standards, there are several contaminants of emerging concern with comparably limited toxicity data. These include the following pollutants: SVOCs that are HAPs/TACs with no available health-based standards; SVOCs that are not HAPs/TACs including pesticides and brominated fire retardants; short-lived products of indoor secondary organic aerosol (SOA) chemistry; and ultra fine particles (UFPs). Since the toxicological and epidemiological data are as yet insufficient to set standards for these compounds, they are treated only qualitatively in this paper.

As a final point, although the our analysis focuses on the method of comparing measured concentrations to health-based standards to establish hazard, we note that there are three indoor air hazards that are already well established—radon, Second-Hand Tobacco Smoke, and carbon monoxide (CO). The health effects of exposure to radon have recently been reviewed by Al-Zoughool and Kreski (2009). Several major reviews have compiled data on measured concentrations and health effects of Secondhand Tobacco Smoke (Surgeon General 2006). For carbon monoxide (CO) there is evidence of acute hazard from hospital emergency room visits and deaths, along with growing concern about possible chronic health effects at levels not previously identified as harmful (Ashley et al. 2005).

Results and Discussion

Summary Statistics

Summary statistics compiled for chronic exposure-relevant concentrations are provided in Tables S2-S5 in the online supplemental information. Table S1 lists concentrations for the criteria pollutants, Table S2 lists the VOC concentrations, Table S3 lists the SVOC concentrations and Table S4 lists concentrations for metals that are components of airborne particulate matter. Of the pollutants measured, 193 have more than one type of summary statistic.

Large variations are seen in indoor concentrations for many of the chemical contaminants measured to date. Based on what has been reported by others (Hodgson et al. 2003; Dawson et al. 2009), this was expected. Mean concentrations vary among chemicals by more almost nine orders of magnitude from ethanol ($9 \times 10^2 \mu\text{g m}^{-3}$) to BDE85 ($1 \times 10^{-6} \mu\text{g m}^{-3}$). Differences between the highest and lowest summary statistic values vary widely by compound: 66 varied by more than a factor of ten and ten varied by more than a factor of 100. The largest variations are seen for cesium (Cs) and 1,1-dichloroethene, which each varied by more than a factor of 2000, followed by 1,4-dichlorobenzene and chlorobenzene, which varied by a factor of 500.

Potential Health Hazards from Chronic Exposures

In this section we compare our representative indoor air concentrations to the relevant standards for chronic health hazards. In all of the figures in this section, the bars indicate the representative mid-range concentration with a line that extends to the representative upper bound concentration. All the graphs are arranged, in decreasing order, by the ratio of the mid-range concentration to the lowest available health standard.

Criteria Pollutants

Figure 1 presents representative indoor concentrations of criteria pollutant along with standards developed by the USEPA (NAAQS), CalEPA, and WHO. The figure shows that the

representative mid-range concentration for PM_{2.5} is above the WHO annual standard and very close to the NAAQS annual standard. The representative upper bound concentration for PM_{2.5} is above both the NAAQS and WHO 24-hour standards. Figure 1 shows that the representative upper bound for NO₂ is above the annual average values set by both WHO and EPA, and above the 1 hour WHO value. Thus, in some homes, NO₂ concentrations averaged over periods of days or longer can exceed both chronic and short-term acute health-based standards.

HAP/TACs

Figures 2 to 4 compare three subclasses of HAP/TACs concentrations to available chronic health standards. Of the compounds evaluated, 20 have representative mid-range or upper bound concentrations that exceed at least one standard. The majority of the 20 pollutants have indoor concentrations that exceed cancer standards only; only four of the pollutants have indoor concentrations that exceed a non-cancer standard. This is an important point since the chronic exposure concentration “standards” used to assess cancer hazards in this study are health protective and inferred from *UREs* (not directly set) as described in the methods section.

Figure 2 illustrates how representative concentrations for HAP/TAC VOCs compare to standards developed directly by USEPA, CalEPA, ADSTR, and OSHA as well as the cancer exposure standards calculated from *UREs*. Of the 63 pollutants with applicable health *RfCs*, *RELs*, and *UREs*, eighteen pollutants have representative indoor concentrations higher than at least one health standard. Of those eighteen, fourteen have concentrations that exceed the cancer value only, two exceed non-cancer endpoints only, and two are potential hazards for both cancer and non-cancer effects. Eleven of the pollutants have representative mid-range concentrations above standards, indicating a potential hazard in a large percentage of homes. Two pollutants identified as hazards, acrolein and formaldehyde, have representative mid-range concentrations above the CalEPA acute standards. Figure 2 also shows that for ten additional pollutants, representative concentrations are within an order of magnitude of at least one health-based standard.

Figure 3 compares the SVOC HAP/TAC concentrations to applicable standards. Of the 108 semi-volatile compounds for which residential indoor air measurements were reported, only 21 have relevant health standards. Of these 21, only naphthalene is identified as a hazard using this methodology. Naphthalene concentrations exceed calculated limits for cancer in many homes and non-cancer chronic standards are exceeded in high concentration homes. Data were sparse for SVOCs other than naphthalene; most other compounds were reported in only one or two studies each. Lindane and heptachlor had concentrations below, but within an order of magnitude of health standards.

Figure 4 compares particle-bound metals to relevant standards. Of the compounds for which measurements are available, only chromium and cadmium appear to pose a hazard. Nickel, chlorine, and arsenic were measured in some homes at concentrations within an order of magnitude of a health representative. There were only one or two studies available for each of these pollutants as well.

In total, we identified 16 chemical pollutants having representative concentrations that were below but within an order of magnitude of at least one health standard. For these pollutants we reviewed the studies that contributed data to their calculated representative concentration values.

The intent was to assess whether the data were sufficient to reach a robust determination that the compound is not present at hazardous levels, or if the potential for hazard is uncertain due to data scarcity.

Five of these chemical pollutants are identified as potential hazards in a small percentage of homes. Sufficient data exist to determine that bromomethane, trichloroethane, chlorine, and 1,2-dichloroethane are present at levels exceeding at least one health standard in U.S. homes with the highest concentrations. Bromomethane exceeded the most health-protective standard in fewer than 5% of the 439 homes measured in one U.S. study (see Table 3). Chlorine, trichloroethene, and 1,2-dichloroethane were measured at concentrations above health standards in a limited number of Texas, New York City, and Saschwtchewan homes respectively. The potential for propanal to be present at hazardous levels is uncertain. Representative concentrations for propanal are within an order of magnitude of some standard primarily owing to high concentrations measured outside of the U.S. Propanal is identified for now as a potential hazards in a small percentage of homes; additional data are needed to clarify this assessment.

An additional five pollutants were identified as possible hazards in a small percentage of homes because available data were insufficient to reach a robust determination that levels are reliably below health-based standards. Lindane, heptaclor, nickel, and arsenic benchmark values are all based on very limited data from one or two studies each. More data are needed to to reliably determine that these compounds are not substantial hazards in U.S. homes.

The remaining six pollutants with representative concentrations close to at least one health standard do not appear to be substantial residential indoor-air hazards. Large studies have measured toluene, m/p-xylene, and MTBE and several of these have 95th percentile concentrations substantially below the most health-protective standard. Representative concentrations for 1,2-dibromomethane and 1,1,2-trichloroethane are biased by a single study with a very high minimum detection level (MDL) and a large number of non-detects that set the concentration to a level of half the MDL (NYDOSHS 2006). There is no positive evidence that these compounds represent a hazard in U.S. homes.

Health-based standards have been established for only a subset of the chemicals known to be present in indoor air. Over 40% of the pollutants with available concentration data do not have available health standards. Tables S6 and S7 in the online supplemental material list the compounds without standards. Table S6 lists the assessed carcinogenicity for chemicals included in the IRIS database and indicates that six are thought to be possible or probable carcinogens. The remaining 102 pollutants are listed in Table S7.

Potential Health Hazards from Chronic Exposures in New Homes

As new homes are added to the existing housing stock, there is concern that increasing home tightness may lead to reduced ventilation and hence higher pollutant concentrations indoors and hope that newer building materials will have reduced emission rates. We looked at new homes separately to investigate variations in hazard profiles. Of the 74 studies reviewed, only 3 focused on new homes. Offermann et al.(2009) measured pollutant concentrations in 108 detached single-family homes in California built between 2002 and 2004; measurements were conducted during 2006-2008 when homes were 1.7 to 5.5 years old. Park and Ikeda (2006) measured

concentrations in 219 new homes (built in 2000) in Japan during the summers of 2000-2003. Hodgson et al. (2000) measured VOC concentrations in 11 new manufactured and site-built US homes within 10 months of construction. Summary statistics for new homes are included in Table S5 in the online supplemental. Similar to older homes, PM_{2.5} concentrations are of concern in new homes. NO₂ mid-range and upper bound representative concentrations measured in new homes are below the standards, yet NO₂ is assumed to be a potential hazard in new homes with any unvented natural gas appliance(s). The rationale is that emission rates are of similar magnitude whereas dilution from whole house ventilation is reduced relative to older homes.

A limited number of VOCs and SVOCs have been measured in new homes. We plot representative concentrations and available standards for these in Figure 5. For comparison, the representative concentrations for all homes are also plotted. The hazards identified were a subset of the hazards identified for all homes and most appear as hazards for cancer only.

Potential Acute Exposures from Episodic Indoor Sources

Several studies have looked at specific events or activities in the home that give rise to high transient pollutant concentrations. Table 2 shows the sample durations (or integration times) and measured values of PM_{2.5}, CO, NO₂, chloroform, and formaldehyde associated with some episodic events and activities in the home. For studies that did not report sample duration, the reported peak concentration is included in Table 2. Peak concentrations were reported for studies that used highly time resolved instrumentation and refer to the single highest value measured. For these measurements sampling times were not reported, but are likely on the order of one minute based on the instrumentation used. Figure 6 compares the highest concentration for each pollutant to acute standards from WHO, USEPA, and CalEPA.

The review identified nine studies reporting PM mass concentrations. Fortmann and Dariher (2001) measured PM_{2.5} during and after prescribed event in the kitchen of a house and showed that concentrations can be several orders of magnitude larger than acute health standards for several hours in homes. Singer et al. (2006) and Coleman et al. (2008) showed that use of terpene-containing products in the presence of ozone can cause particle generation events that lead to concentrations above acute standards for at least half a day.

The use of unvented gas cooking appliances and fireplaces can lead to CO and NO₂ concentrations above acute standards for several hours. Gordon et al. (2008) measured concentrations of CO and NO₂ in 30 homes with unvented natural gas fireplaces over periods of up to a few days in each home with occupants directed to not alter their appliance use patterns. During their period of measurement, NO₂ concentrations exceeded acute (1-h) standards in 80% of the homes and the 8-h CO standard was exceeded at least once in 20% of the homes. Dutton (2001) operated an unvented fireplace in a single home through a series of scripted events and found that CO concentrations could exceed EPA standards.

Limited information is available in the literature for other pollutants and activities. Cooking was shown to elevate levels of formaldehyde above acute standards for several hours. Kerger and Schmidt (2000) found that showering for 12 minutes elevated bathroom concentrations of chloroform above acute standards for half an hour. Although in this case, chloroform

concentrations may be less than the standard over the course of an entire hour, shower durations longer than 12 min could easily lead to concentrations above acute standards for an hour.

Additional Contaminants of Concern

Whereas the analysis in preceding sections depend on the availability of health-based standards, this section explores the potential hazard associated with indoor air contaminants for which no specific concentration-based standards or guidelines have been established. Of the 321 compounds with available indoor concentrations measurements, 40% did not have available health standards to aid in the identification of indoor hazards. In this section a similar method will be used to identify indoor chemical hazards based on mechanistic, epidemiological and toxicological evidence.

It has been shown that SVOC concentrations can be an order of magnitude larger indoors than outdoors and moderately to highly sorbing compounds can persist indoors for weeks to several years (Weschler et al. 2008). Bio-monitoring studies have shown that SVOCs appear in human blood and urine samples (Wilford et al. 2005; Canosa et al. 2007; Mannino et al. 2008) and there is significant epidemiological evidence that specific chemical classes may have harmful effects on the human body (Darnerud 2003; Legler et al. 2003; Miyazaki et al. 2004; Ghisari et al. 2009) including endocrine disruption that may affect the behavior of hormones in the human body. Increasing attention is being devoted to the potential hazards associated with indoor SVOCs. SVOCs indoors quickly absorb to available surfaces including human skin. Despite low concentrations indoors, indoor air is a medium for transporting SVOCs from surfaces to skin where they can potentially accumulate and be absorbed into the body (Weschler et al. 2008). Recently, the USEPA has designated phthalates and polybrominated diphenyl ethers (PBDEs) as chemicals of concern (USEPA 2010). Several phthalates and PBDEs were found to have measurable indoor concentrations in this study.

Ultra fine particles (UFP), <100 nm in diameter, make up more than 90% of the number count of PM_{2.5}, but only 10% of the mass (Buonanno et al. 2009). Health scientists have hypothesized that the small size and large surface area of UFP may lead to greater toxicity per unit mass than for the larger diameter particle fraction of PM_{2.5} (Delfino et al. 2005; Gwinn et al. 2006; Peters et al. 2006). It has been suggested that the small size of these particles may make them more dangerous and potentially lead to translocation of particles to the blood stream and other organs or acting on the autonomic nervous system (Knol et al. 2009). UFP emissions are from both primary sources such as natural gas combustion and food preparation, as well as secondary sources such as ozone reactions with terpenes in cleaning materials (Singer et al. 2006). Bhangar et al. (2010) showed that in houses where people are at home and awake, and presumably undertaking everyday indoor activities, particle number (PN) concentrations are consistently higher than outdoors, by as much as a factor of 3. The same study showed that PN concentrations when people are home and asleep or when homes are unoccupied are consistently lower than concentrations outdoors, again underlining the importance of indoor sources. Recent expert review determined that there is sufficient evidence supporting the harmfulness of UFP (Knol et al. 2009), however no standard has been set.

Summary of Identified Hazards

Table 3 summarizes the results of the hazards analysis. The table subdivides the chronic hazards into three groups: hazards in most homes, hazards in some homes (on the order of 5-50%) and hazards in very few homes (on the order of a few percent or less) based on what percentage of the available data has concentrations greater than available standards. These groupings are based on our representative mid-range and upper-bound concentrations that generally derive from weighted median and 95th percentile values of reported concentrations in homes. The table also indicates the type of hazard (cancer or non-cancer), and the level of certainty. The level of certainty reflects whether we believe that the available data is representative of the current state of US homes and was based on the number of available studies, whether reported concentrations were above a standard in U.S. homes or only in homes outside of the U.S, and, in a few cases, information about concentrations outdoors.

Of the 15 compounds identified in most homes, nine were identified as priority chronic hazards in U.S. residences: acetaldehyde, acrolein, benzene, 1,3-butadiene, 1,4-dichlorobenzene, formaldehyde, naphthalene, NO₂, and PM_{2.5}. These are nine of the ten pollutants identified as hazards with a high level of certainty in most homes. The tenth pollutant, carbon tetrachloride, was used extensively as a refrigerant in the past, but was banned as part of the Montreal Protocol and has been largely phased out. Due to a long atmospheric lifetime, carbon tetrachloride is still present in the atmosphere at hazardous concentrations.

The pollutants identified as acute hazards are, for the most part, a subset of the pollutants identified as chronic pollutants. Chloroform was additionally identified as posing a potential acute hazard.

Our results are similar to those identified by the reviews done by Dawson et al.(2009), Koistinen et al.(2008), and Loh et al.(2007) with some distinct differences. Loh et al.(2007) identified a similar subset of high priority VOC and SVOC chemical air pollutants using a combination of measurements and modeling. Our review identified a similar set of VOC and SVOC priority pollutants with the addition of acrolein, which was not included in their study. Dawson et al.(2009) identified benzene as having an elevated cancer risks in most homes by comparing concentrations of a subset of 10 VOCs to available standards. Koistinen et al.(2008) identified 5 priority pollutants in European homes, formaldehyde, CO, NO₂, benzene, and naphthalene. With the exception of CO, these pollutants were identified as priority pollutants in this study as well. The difference appears to be due to higher long term concentrations in European homes.

Summary and Conclusions

This analysis identified mid-range and upper bound chronic exposure relevant representative concentrations for over 300 chemical pollutants and acute exposure relevant concentrations for 5 indoor activity related chemical pollutants. The results are summarized in Table 3. Comparisons of pollutant concentrations to relevant health standards indicate 15 pollutants that are chronic hazards in most homes, 9 that are chronic hazards in some homes (on the order of 5-50%), and 6 that are chronic hazards in very few homes (fewer than 5%). Additionally, 6 pollutants were identified as potential acute health hazards indoors. Of those 31 chemical hazards, 9 were identified as priority chemical pollutants in U.S. homes:

- acetaldehyde,
- acrolein,
- benzene,
- 1,3-butadiene,
- 1,4-dichlorobenzene,
- formaldehyde,
- naphthalene,
- NO₂,
- PM_{2.5}.

Over 40% of the pollutants with available indoor concentration data did not have available health standards whether due to a lack of toxicity data or because the pollutant is non-hazardous. There are a vast number of pollutants in the indoor environment that cannot be said to be at safe or unsafe levels. Mechanistic and epidemiological evidence has suggested that certain pollutants, such as UFPs and select SVOCs including phthalates and brominated flame retardants, pose a potential hazard to human health. Further toxicological and epidemiological research is needed to quantify the risks posed by emerging pollutants of concern.

It is important to note that, despite the large number of articles included in the study, that data included here is not fully representative of indoor exposures across the country due to biases in sampling locations, the large number of pollutants that were not measured by any study, and because the effect of residential concentrations on personal exposure is not perfectly understood.

There were several areas where data for particular sub-populations and pollutant groups were lacking. Relatively few concentration measurements were available for new homes, only three studies. Analysis of the two studies that focused on new homes indicated a similar hazard profile as seen in older homes. More data is needed for SVOCs. These pollutants have recently come to the attention of health professionals and, with the exception of naphthalene, have not been sampled as extensively as VOCs in the indoor environment. There is also insufficient data for the majority of US states. The measurement studies that have focused on the US have disproportionately focused on California and the east coast. Of the 40 studies in the US, 34 focused on only 7 states. The remaining states, representing diverse climates, have been largely underrepresented. The data compiled here is also largely for single-family homes. Despite the inclusion of six studies that surveyed lower socioeconomic status, more data is needed for multi-unit residential structures. More work is needed to determine the relative contributions of location and sub-standard housing to risks. Finally, the data have mostly been taken in developed areas, this may lead to slight overestimation of concentrations indoors due to the effects of infiltration from the outdoors.

This work successfully reduced a list of 321 indoor chemical pollutants to a set of 9 priority chemical hazards. The identification of a succinct group of chemical hazards in indoor air will allow for the design of mitigation strategies in the indoor residential environment that target these priority pollutants.

Figures and Tables

Table 1 Publications with chronic exposure relevant concentrations. (F=formaldehyde, NA=naphthalene, A=acrolein, P=PM_{2.5}, N=NO₂, O=ozone, C=CO). For some of the studies only one VOC or SVOC was included for these studies the individual pollutant is indicated instead of the pollutant class.

	Study	Sample Period	Location	US homes	New homes	Criteria Pollutants	VOCs	Aldehyde	SVOCs	Metals	Number of samples
1	(Topp et al. 2004)	2 weeks	Hamburg/Erfurt, Germany			N	X				2524
2	(Park et al. 2006)	24 hrs	Japan		X		X	X			2151
3	(Geyh et al. 2000)	6 months	Upland, California	X		O					1980
4	(Wilson et al. 1986)	1 week	Southern California	X		N					1800
5	(Rehwagen et al. 2003)	4 weeks	Leipzig, Germany				X		X		1499
6	(Garcia-Algar et al. 2003)	7-15 days	UK, Spain			N					1438
7	(Williams et al. 2009)	5 days	Detroit, Michigan	X		P					973
8	(Lee et al. 1998)	48 hrs	Boston, Massachusetts	X		N					942
9	(Raw et al. 2004)	2 weeks	England, UK			N,C					812
10	(Levy 1998)	48 hrs	Varied	X		N					617
11	(Kirchner et al. 2009)	7 days	Nationwide, France			C,P					570
12	(Weisel et al. 2005)	48 hrs	LA, CA; Houston, TX; Elizabeth, NJ	X		P	X	X			121-554
13	(NYDOSH 2006)	2-12 hrs	New York	X			X		NA		4-546
14	(Saborit et al. 2009)	24 hrs	Wales/W. Midlands, England				X		X		91-500
15	(Cyrus et al. 2000)	1 week	Hamburg/Erfurt, Germany			N					404
16	(Marchand et al. 2008)	30-95 mins	Strasbourg, France				X	X			244-286
17	(Turpin et al. 2007)	48 hrs	LA, CA; Houston, TX; Elizabeth, NJ	X		P			X	X	157-275
18	(Croxford et al. 2006)	1 week	London, UK			C					270
19	(Jia et al. 2008)	3-4 days	Ann Arbor, Michigan	X			X		X		251-257
20	(Avol et al. 1998)	24 hrs	Los Angeles, California	X		O					241
21	(Jo et al. 2006)	2 hrs	Daegu, South Korea			C					240
22	(Heroux et al. 2009)	24 hrs	Saskatchewan, California				X	X	NA		217
23	(Long et al. 2000)	12 hrs	Boston, Massachusetts	X		P					211
24	(Offermann 2009)	24 hrs	California	X	X	P,C, N	X	X	X		31-211
25	(Jarvis et al. 2005)	14 days	United Kingdom			N					203
26	(Edwards et al. 2001)	48 hrs	Helsinki, Finland				X	X	X		201
27	(Gordon et al. 2008)	6-7 days	Arizona	X			X				170
28	(Jia et al. 2008)	3-4 days	Southeast Michigan	X			X	X	NA		159
29	(Sexton et al. 2004)	2 days	Minneapolis/St. Paul	X			X				132
30	(Rudel et al. 2003)	24 hrs	Cape Cod, Massachusetts	X					X		102-120
31	(Fromme et al. 2004)	7 hrs	Berlin, Germany						X		61-118
32	(Simons et al. 2007)	3 days	Baltimore, Maryland	X		O, P, N					95-109
33	(Weisel 2006)	24 hrs	New Jersey	X			X				7-100
34	(Zota et al. 2005)	2 weeks	Boston, Massachusetts	X		N					100
35	(Guo et al. 2009)	24 hrs	Hong Kong				X				100
36	(Gilbert et al. 2006)	7 days	Quebec City, Canada			N		X			96
37	(Lee et al. 2002)	6 days	Southern California	X		N					92
38	(Miller et al. 2009)	24 hrs	Commerce City, Colorado	X		C,P					92-97
39	(Phillips et al. 2005)	24 hrs	Cities, Oklahoma	X			X				90
40	(Kinney et al. 2002)	48 hrs	West central Harlem, New York	X		P	X	X		X	18-88
41	(Lee et al. 2002)	6 days	Southern California			N,O					88-102
42	(Sorensen et al. 2005)	48 hrs	Copenhagen, Denmark			P,N					73-85
43	(Dodson et al. 2008)	24 hrs	Boston, Massachusetts	X			X	X			83
44	(Janssen et al. 2005)	24 hrs	Amsterdam/Helsinki			P				X	82
45	(Garrett et al. 1999)	24 hrs	Latrobe Valley, Victoria, Australia			N					80
46	(Zhu et al. 2005)	24 hrs	Ottawa, Ontario, CA				X		X		75
47	(Raymer et al. 2009)	24 hrs	Sacramento, California	X			X	X			47-70
48	(Johnson et al. 2004)	3 days	Columbus, Ohio	X		C					67
49	(Sakai et al. 2004)	24 hrs	Nagoya, Japan			N	X	X			64
50	(Baxter et al. 2007)	3-4 days	Boston, Massachusetts	X		N,P				X	62
51	(Kornartit et al. 2010)	1 week	UK			N					60
52	(Gilbert et al. 2005)	24 hrs	Prince Edward Island, Canada					X			59
53	(Leaderer et al. 1999)	24 hrs	SW and central VA	X		P,SO2					58
54	(Piechocki-Minguy et al. 2006)	24 hrs	Lille, France			N					44
55	(Malkin-Weber et al. 2009)	7 days	Central North Carolina	X				X			36
56	(Zhang et al. 1994)	6 days	New Jersey	X			X	X			36

57	(Harrad et al. 2006)	28 days	Birmingham, UK						X		31
58	(Gustafson et al. 2007)	24 hrs	Hagfors, Sweden				X	X			21-23
59	(Na et al. 2004)	24 hrs	Riverside County, California	X		P				X	20
60	(Stranger et al. 2009)	24 hrs	Antwerp, Belgium			P				X	19
61	(Zipprich et al. 2002)	48 hrs	Richmond, VA	X		N					19
62	(Mukerjee et al. 1997)	3 weeks	Lower Rio Grande Valley, Texas	X		P	X				6-15
63	(Strandberg et al. 2006)	2 weeks	Hagfors, Sweden						X		15
64	(Batterman et al. 2009)	7 days	Southeastern Michigan	X					X		12
65	(Johnson-Restrepo et al. 2009)	8 hrs	Albany, New York	X					X		12
66	(Chao 2001)	48 hrs	Hong Kong			O					10
67	(Toms et al. 2009)	31 days	Brisbane, Queensland, Australia						X		10
68	(Seaman et al. 2009)	24 hrs	California	X			A				9
69	(Missia et al. 2008)	7 days	Europe				X	X			8
70	(Hodgson et al. 2000)	3 days	Southeastern United States	X	X		X				8
71	(Arhami et al. 2009)	5 days	San Gabriel Valley/Riverside, California	X		P, C, N					8
72	(Kamens et al. 1991)	24 hrs	Chapel Hill, North Carolina	X		P					8-9
73	(Koziel et al. 2001)	8 hrs	Waterloo, Ontario, Canada					X			4
74	(Kuntasal et al. 2005)	2 hrs	Ankara, Turkey				X		X		1

Table 2 Short-term concentrations during typical indoor residential activity.

Chronic Health Hazards

Hazards in most homes	Studies	Hazard*	Level of Certainty
acetaldehyde**	11,12,16,22,24,32,43,47,52,56,58,69	C/NC	high
acrolein**	11,12,22,52,68	NC	high
benzene**	1,5,11,12,13,14,19,22,24,26,28,33,39,40,43,46,52,58,62,69,74	C	high
butadiene, 1,3-**	14,19,22,33,40,46,58,62	C	high
carbon tetrachloride	12,13,29,49,62	C	high
dichlorobenzene, 1,4- **	2,12,13,19,24,28,29,33,35,40,43,49	C	high
formaldehyde**	2,11,12,16,24,27,35,36,40,43,47,49,52,55,56,58,69,71	C/NC	high
naphthalene**	5,13,14,19,22,26,28,46,74	C/NC	high
NO2**	1,4,8,9,10,15,24,25,32,34,36,37,41,50,51,54,61,70,72	NC	high
PM2.5**	7,11,12,17,23,24,32,38,40,42,44,50,53,59,60,62,70,72	NC	high
acrylonitrile	46	C	medium
chromium	17,40,59,62	C	medium
hexachlorobutadiene	13	C	low
benzyl chloride	13	C	low
vinyl chloride	42	C	low

Hazards in some homes

chloroform	12,13,19,24,28,29,33,40,43,46,49,62	C	high
environmental tobacco smoke	not applicable	C/NC	high
ethylbenzene	1,2,5,12-14,19,26,28,33,35,40,43,46,49,62,69,71	C	high
methylene chloride	12,13,22,29,33,40,43,46	C	high
radon	not applicable	C	high
tetrachloroethene	5,11,12,13,19,24,28,29,40,43,46,49,62	C	high
cadmium	1,13,59	C	medium
dichloropropane,1,2-	13,46	NC	medium
ethanol	13,22	C	medium
tetrachloroethane, 1,1,2,2-	13,46	C	medium

Hazards in very few homes

bromomethane	13	NC	high
chlorine	17,44,50,59,62	NC	high
CO	9,11,18,21,24,38,48,71	NC	high
dichloroethane, 1,2-	13,22,46	C	high
trichloroethene	2,5,11,12,13,19,29,40,43,46,49,62	C	high
propanal	2,12,47,56,69	C	low

Acute Health Hazards

acrolein	11,12,22	SI	high
formaldehyde	2,24,35,55,56,73,76	SI	high
CO	77,78	H	high
PM2.5	11,12,23,24,32,59,60,62,76,83-88	R/H	high
NO2	4,77-81	R	high
chloroform	75	RD	low

*NC=noncancer, C=cancer, SI=sensory irritation, H=cardiovascular, R=respiratory, RD=reproductive/developmental

**priority hazards

Table 3 Pollutants that potentially pose an adverse indoor health risk. Study numbers correspond to study lists in table1 and table 2.

Pollutant	Activity	Conc. (µg/m³)	Measurment Duration	Source (Study #)
<u>Chloroform</u>	12min shower	157	32min	Kerger et al. 2000 (75)
<u>Formaldehyde</u>	oven cleaning	417	5.5hrs	Fortmann et al. 2001 (76)
	gas)	129	3hrs	Fortmann et al. 2001 (76)
<u>NO₂</u>	unvented fireplace use	2422	1hr	Gordon et al. 2008 (77)
	oven cleaning	1435	5.5 hrs	Fortmann et al. 2001 (76)
	cooking french fries (gas)	772	2.5hrs	Fortmann et al. 2001 (76)
	unvented fireplace use	677	4 hrs	Dutton et al. 2001 (78)
	cooking	355	4 min	Park et al. 2008 (79)
	maximum in kitchen	243	3.7(2.6)hrs	Franklin et al. 2006 (80)
	maximum in kitchen	209	7.3hrs	Noy et al. 1990 (81)
<u>CO</u>	unvented fireplace use	114000	2hr	Dutton et al. 2001 (78)
	unvented fireplace use	20486	1hr	Gordon et al. 2008 (77)
<u>PM_{1.1}</u>	cleaning products	89	12hr	Singer et al. 2006 (82)
<u>PM_{2.5}</u>	oven cleaning	6381	5.2hrs	Fortmann et al. 2001 (76)
	cooking fish (gas stove)	3146	3hrs	Fortmann et al. 2001 (76)
	maximum in house	2842	peak conc.	Morawska et al. 2003 (83)
	constantly sooting candle	1400	1hr	Pagels et al. 2009 (84)
	cooking	745	peak conc.	He et al. 2004 (85)
	grilling bacon	389	peak conc.	Buonanno et al. 2009 (86)
	expirement)	215	steady state	Coleman et al. 2008 (87)
	candle vapour eucalypt oil	132	24hrs	He et al. 2004 (85)
	maximum in house	105	peak conc.	Stranger et al. 2007 (88)
	hair dryer	45	peak conc.	He et al. 2004 (85)
	washing machine	43	peak conc.	He et al. 2004 (85)
	sweeping	35	peak conc.	He et al. 2004 (85)
	kerosene lamp	32	1min	He et al. 2004 (85)
	oil lamp	30	1min	He et al. 2004 (85)

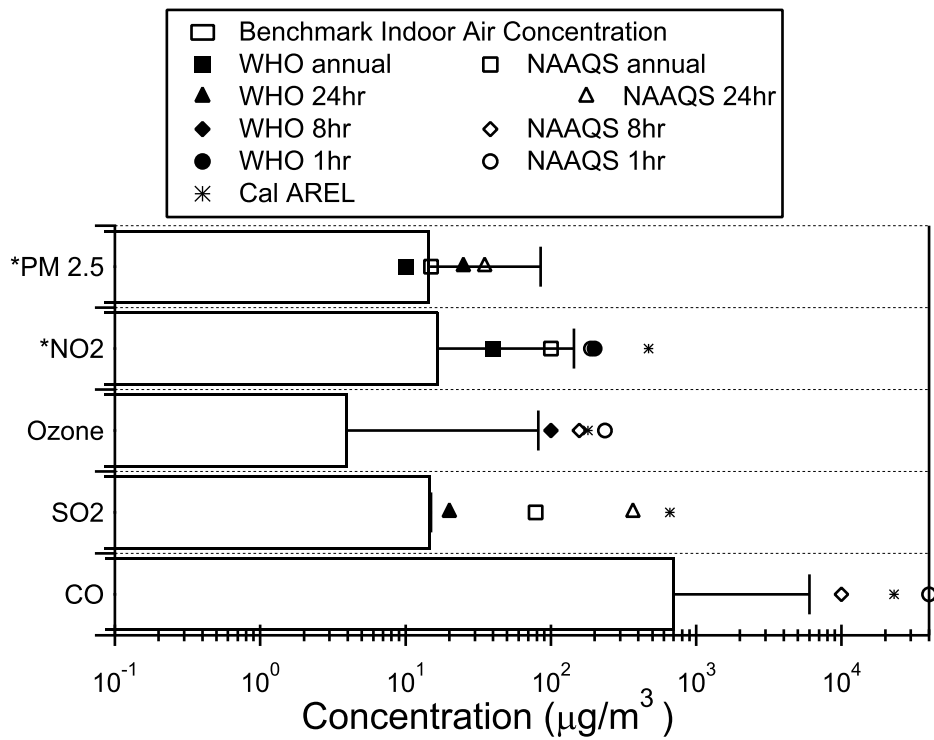


Figure 1 Representative indoor air concentrations compared to relevant national and international standards. Line extends to the upper bound indoor concentration. CAL AREL is the CalEPA acute reference exposure level (1hr).

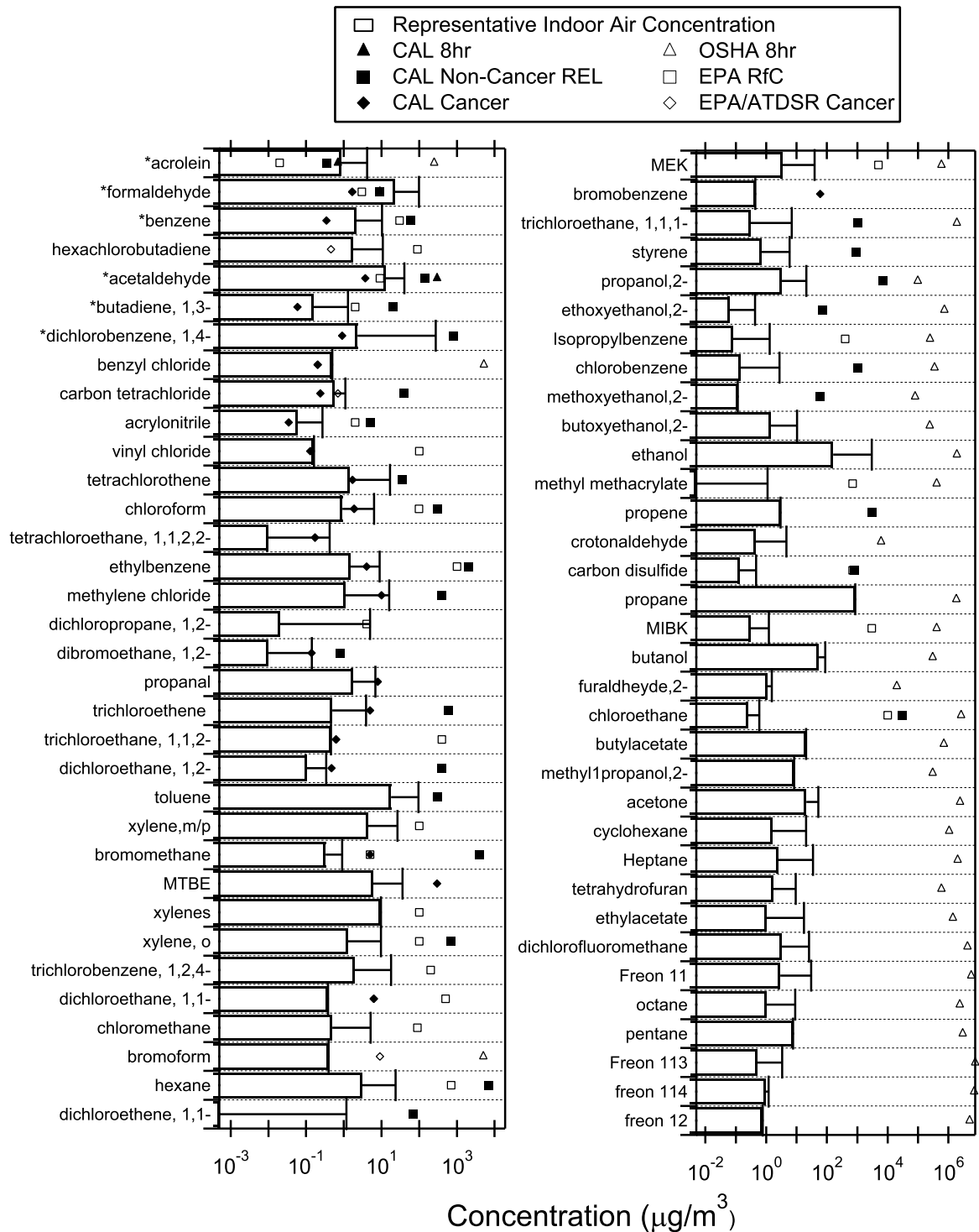


Figure 2 Our representative indoor air concentrations compared to volatile organic HAP/TAC health standards. The line extends to representative upper bound indoor concentration. Cancer, RfC, and REL standards are for chronic long term exposure (70 years). OSHA standards are for workday exposure for a significant portion of a lifetime. (CAL=CalEPA, EPA=USEPA). Priority pollutants are identified with an asterisk.

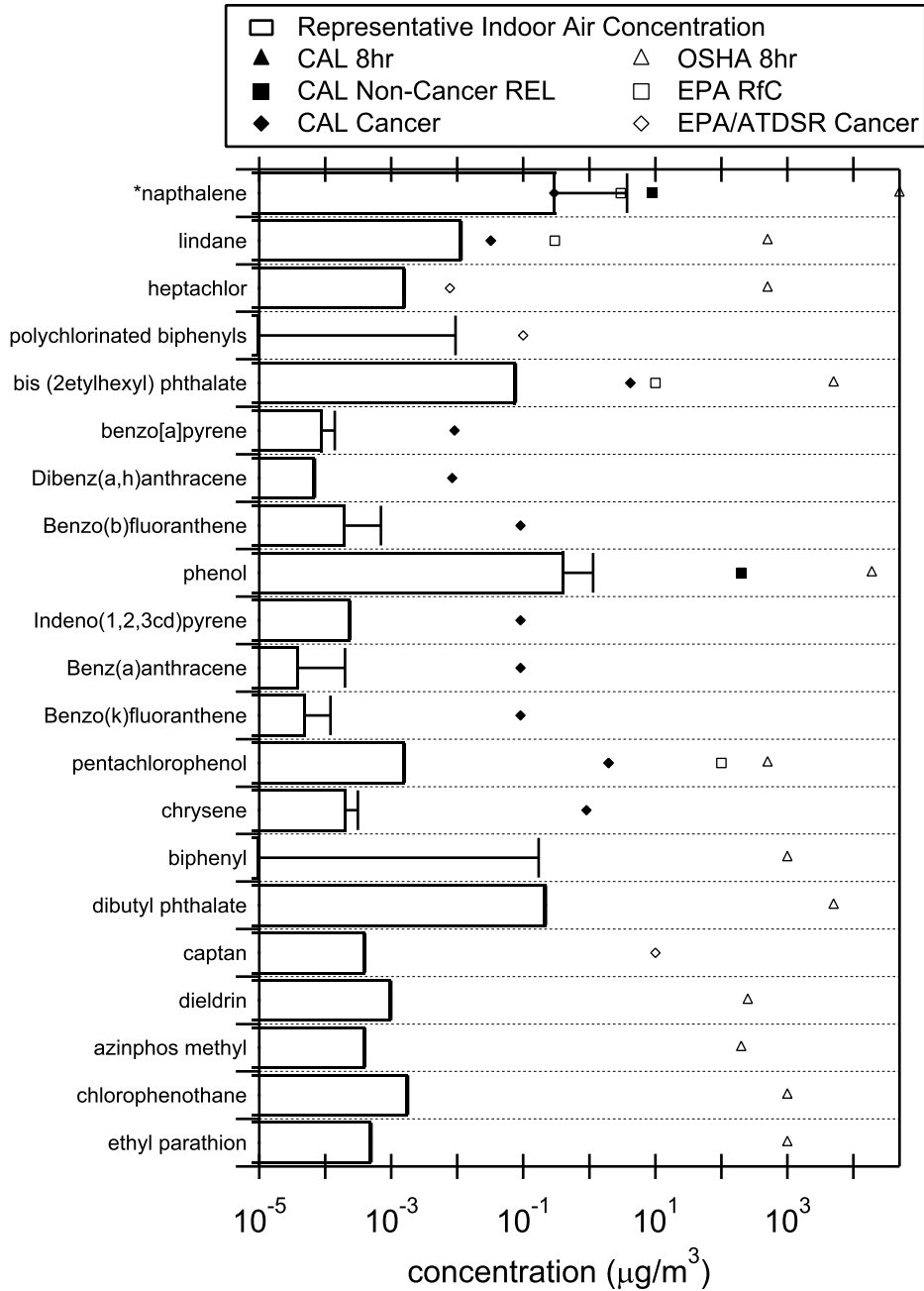


Figure 3 Representative indoor air concentrations compared to SVOC HAP/TAC health standards. The line extends to our upper bound representative indoor concentration. Cancer, RfC, and REL standards are for chronic long term exposure (70 years). OSHA standards are for workday exposure for a significant portion of a lifetime. (CAL=CalEPA, EPA=USEPA). (CAL=CalEPA, EPA=USEPA). Priority pollutants are identified with an asterisk.

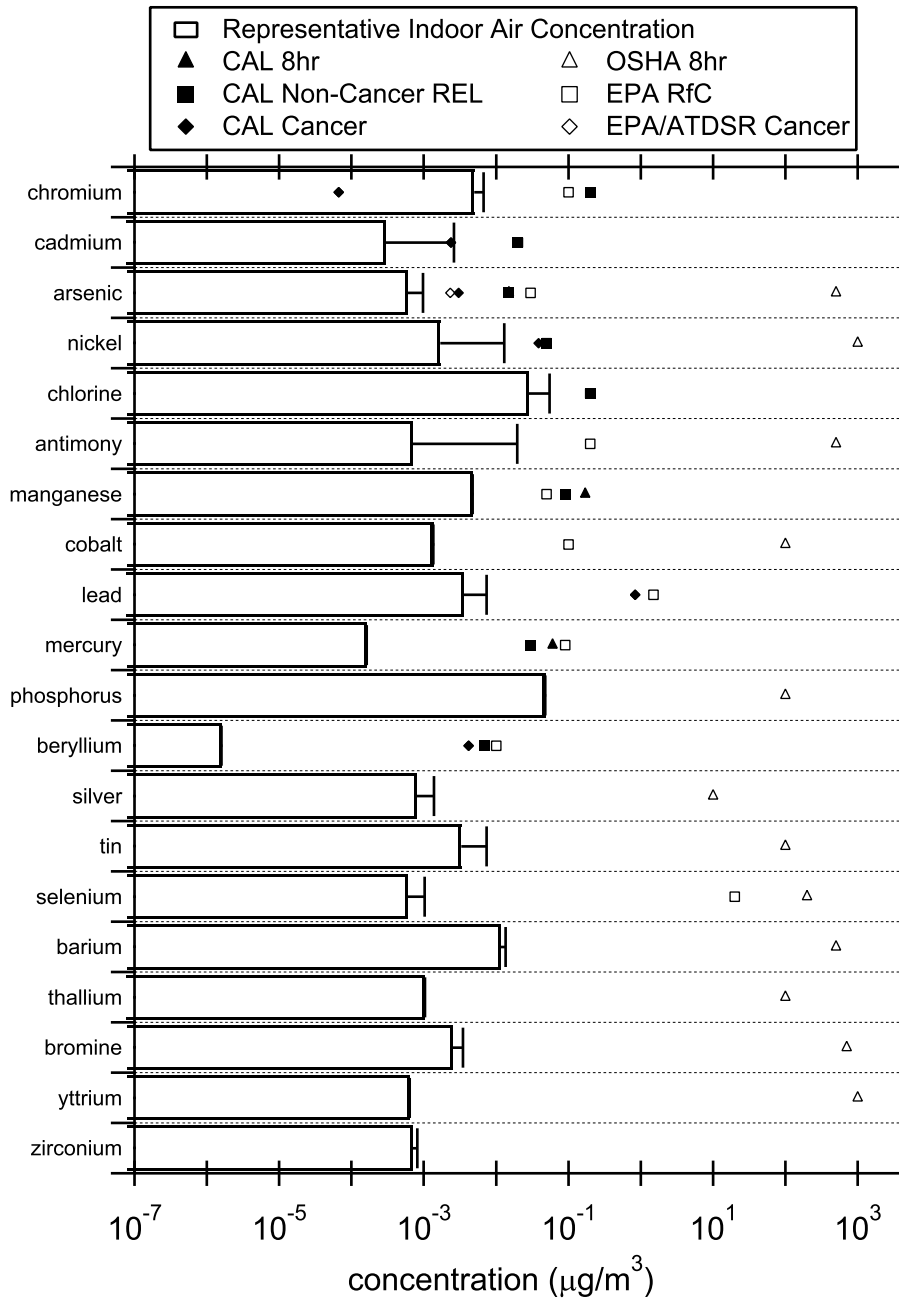


Figure 4 Representative indoor air concentrations compared to metal HAP/TAC health standards. The line extends to the upper bound representative indoor concentration. Cancer, RfC, and REL standards are for chronic long term exposure (70 years). OSHA standards are for workday exposure for a significant portion of a lifetime. (CAL=CalEPA, EPA=USEPA).

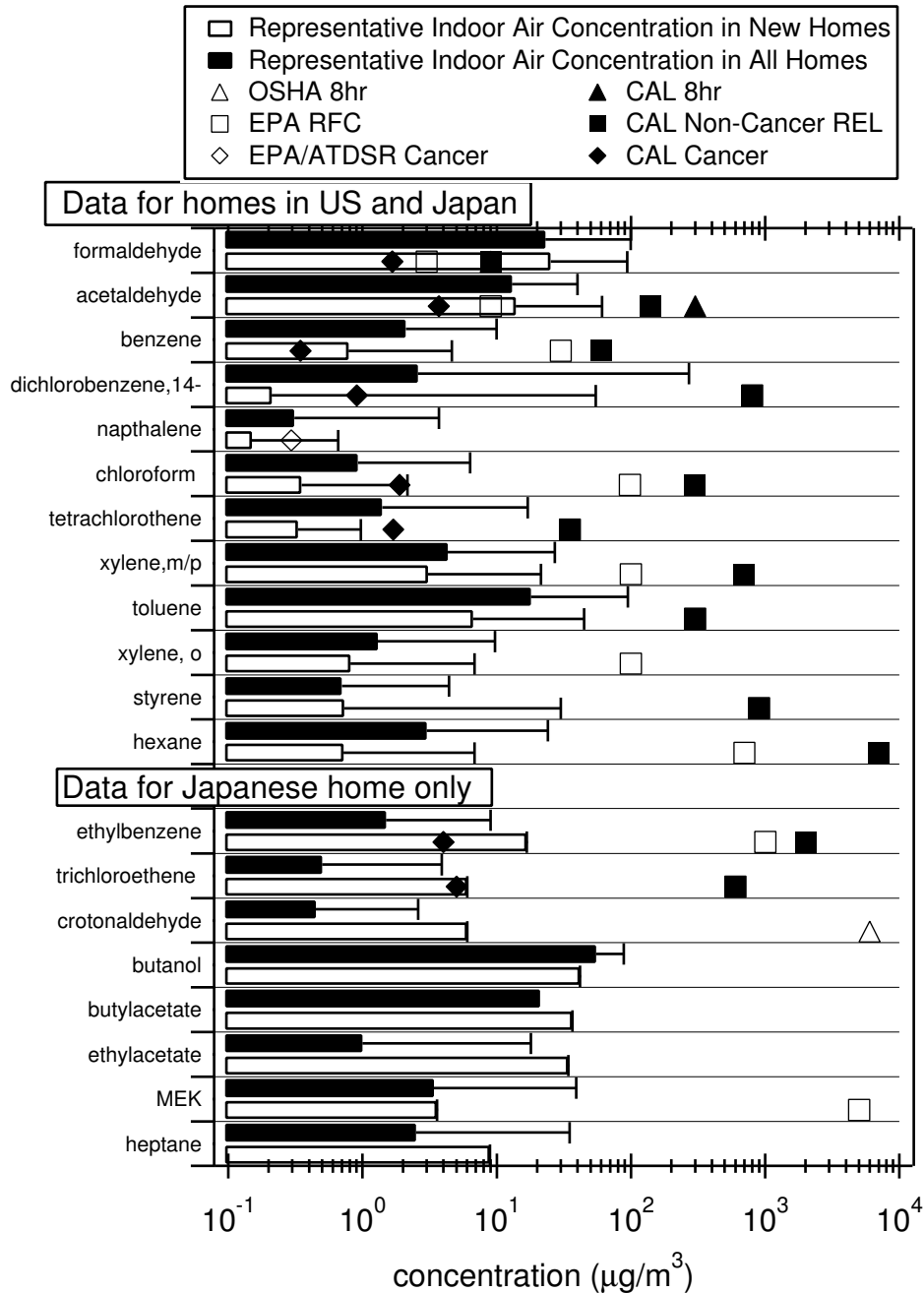


Figure 5 Representative indoor air concentrations in new homes compared to HAP/TAC standards. Error bar extends to the upper bound representative indoor concentration. Cancer, RfC, and REL standards are for chronic long term exposure (70 years). OSHA standards are for workday exposure for a significant portion of a lifetime. (CAL=CalEPA, EPA=USEPA). Representative indoor air concentrations in all homes are added for comparison.

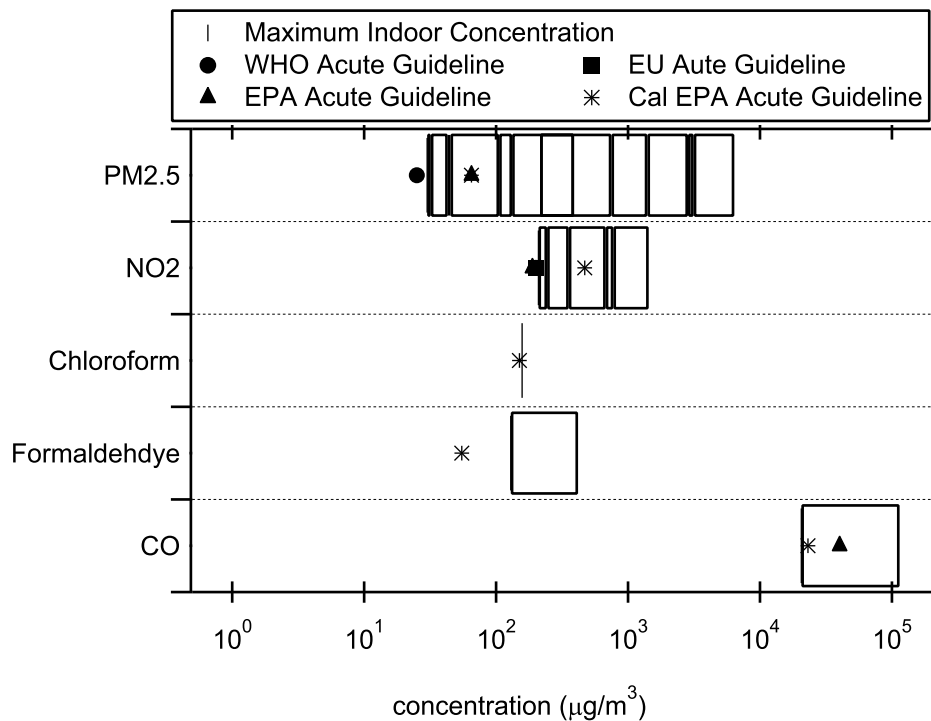


Figure 6 Range of acute concentration measurements compared to acute standards for WHO (1hr standard for NO₂, 24 hr for PM_{2.5}), EPA standards (1 hr for PM_{2.5}, NO₂, and CO), and CalEPA (1 hr for CO, chloroform, and formaldehyde). The bars represent the range of measured acute concentrations and each vertical line in the bar represents an individual measurement

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