Environmental Research 145 (2016) 39–49

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Assessment of inhalation exposure to indoor air pollutants: Screening for health risks of multiple pollutants in Japanese dwellings

Kenichi Azuma [a,b,*], Iwao Uchiyama [b], Shigehisa Uchiyama [c], Naoki Kunugita [c]

[a] Department of Environmental Medicine and Behavioral Science, Kinki University Faculty of Medicine, 377-2 Ohnohigashi, Osakasayama, Osaka 589-8511, Japan

[b] Sick-house Medical Science Laboratory, Division of Basic Research, Louis Pasteur Center for Medical Research, 103-5, Tanakamonzen-cho, Sakyo-ku, Kyoto 606-8225, Japan

[c] Department of Environmental Health, National Institute of Public Health, 2-3-6 Minami, Wako-shi, Saitama 351-0197, Japan

ARTICLE INFO

Article history:
Received 22 August 2015
Received in revised form
11 November 2015
Accepted 12 November 2015
Available online 6 December 2015

Keywords:
Airborne chemicals
Indoor air quality
Nationwide survey
Risk assessment
Screening

ABSTRACT

Over the past few decades, multiple low level indoor pollutants have been found in domestic dwellings. The types and concentrations of these indoor pollutants have not been consistent over time and have changed with alterations in lifestyle, the development of novel products used in housing, and the development of new measurement technologies. To clarify the highest risk pollutants for which health risks should be reduced, we conducted a health risk assessment of 49 indoor air pollutants measured in 602 houses during winter and summer from 2012 to 2014. Inhalation reference concentrations were determined, and the margins of exposure were estimated for each indoor pollutant from measured indoor air concentrations. Health risks due to ammonia and acidic gases, including formic acid, acetic acid, and hydrogen chloride, were also assessed. Overall, during both winter and summer, the highest risk pollutants were acrolein, nitrogen dioxide, benzene, formic acid, and hydrogen chloride. The health risks of propanal, acetaldehyde, and 1,4-dichlorobenzene were also high. Principal component analysis (PCA) suggested an independent principal component for 1,4-dichlorobenzene. The primary source of exposure to 1,4-dichlorobenzene in Japan is an indoor household insect repellent. The improvement of individual lifestyle and housing may be appropriate targets for reducing the risk associated with this compound. The provision of further information on the risk to consumers and promotion of changes in consumer consciousness are needed. PCA suggested that the health risks of indoor air pollutants are amalgamated into similar chemical families, such as aldehydes, aliphatic hydrocarbons, aromatic hydrocarbons, or acetic esters. Our results suggest that health-based guidelines or source control measures, based on these chemical families and similar health endpoints, are appropriate for reducing total health risk due to multiple low level indoor pollutants.

1. Introduction

People in modern societies spend more than 90% of their time indoors (Leech et al., 2002), and most of that time is spent in at home (Brasche and Bischoff, 2005; Matz et al., 2014). Thus, the quality of the indoor environment in households has a significant impact on public health. Indoor environmental hazards include biological and chemical pollutants, as well as physical factors such as thermal conditions, lighting, and noise. These hazards might cause or exacerbate a variety of adverse effects, including disease conditions, such as asthma, allergic disorders, cardiovascular diseases, dermatitis, mucosal irritation, central nervous system effects, and certain cancers (Wu et al., 2007).

There are several important barriers to developing policies to improve indoor home environments. It is difficult to develop regulations concerning indoor home environments because these regulations could affect the privacy of individuals. In addition, the regulations are not under the responsibility of one single department or ministry, and no special law comprehensively addresses this subject in many countries. Individuals in a housing environment may be exposed to a greater variety of chemicals, albeit at lower concentrations, compared with industrial workers in occupational environments. Numerous sources of chemical emissions are found in the housing environment depending on the lifestyle of the occupants. These sources include building materials, furniture, smoking, combustion appliances, insect repellents, and insecticides (Azuma et al., 2007, 2008). Hence, in contrast to the

* Corresponding author.
E-mail addresses: kenazuma@med.kindai.ac.jp (K. Azuma), iwao-u@cyber.ocn.ne.jp (I. Uchiyama), uchiyama@niph.go.jp (S. Uchiyama), kunugita@niph.go.jp (N. Kunugita).

http://dx.doi.org/10.1016/j.envres.2015.11.015
0013-9351/

situation for outdoor air and the air in work places, setting air quality standards does not seem to be the appropriate way to reduce indoor air pollution. Rather, guideline values for indoor air pollutants have been recommended (Harrison, 2002; Levin, 1998; Seifert, 1992; Seifert et al., 1999; WHO Europe, 2010), and many countries have developed such guideline values.

After conducting extensive housing exposure assessments in Japan from 1997 to 1999, the Ministry of Health, Labor and Welfare of Japan also developed indoor air quality guidelines for 13 chemicals [formaldehyde, toluene, xylene, 1,4-dichlorobenzene, ethylbenzene, styrene, chlorpyrifos, di-*n*-butyl phthalate, tetradecane, di-(2-ethylhexyl) phthalate, diazinone, acetaldehyde, and fenobucarb] from 1997 to 2002 (Azuma et al., 2008). Since then, the indoor air concentrations of formaldehyde, toluene, xylene, and ethylbenzene have notably decreased (Osawa and Hayashi, 2009). However, neither the types nor the concentrations of chemicals found indoors are consistent. Changes occur day-to-day, month-to-month, year-to-year, and decade-to-decade with alterations in lifestyles, the development of novel products used in housings, and the development of new measurement technologies (Weschler, 2009). Thus, a comprehensive investigation and an action for reducing health risks due to indoor air pollutants should be continued to protect public health.

To determine the highest risk indoor air pollutants for which health risks should be reduced, we proposed a risk-screening scheme for air pollutants (Azuma et al., 2007). The health risk levels of 93 chemicals were characterized from nationwide survey data of indoor air concentrations measured between 1995 and 2005, and their available hazard data (Azuma et al., 2007). As results, six chemicals, formaldehyde, acrolein, 1,4-dichlorobenzene, benzene, tetrachloroethylene, and benzo[a]pyrene were classified in the highest risk category (action level). After this study, we developed some passive samplers that were suitable for nationwide surveys of indoor air pollutants and conducted a nationwide survey in 602 houses during winter and summer from 2012 to 2014 (Uchiyama et al., 2015). In this survey, the measured data of indoor air concentrations of aldehydes and volatile organic compounds were obtained. In addition, for the first time, data for ozone, ammonia, and acid gases were obtained on a nationwide scale. These chemicals were commonly observed in this survey; however, health risks on a nationwide scale have not been evaluated thus far.

Based on these insights and results, we evaluated the health risks of the recent nationwide indoor exposure data in Japan and characterized health risk levels based on currently available hazard data. In addition, we attempted to analyze the dominant components for reducing risks.

2. Methods

2.1. Data collection for indoor exposure concentrations

Data from a nationwide survey in 602 houses, collected from 2012 to 2014 in Japan during winter and summer (Uchiyama et al., 2015), were used. Indoor air pollution was evaluated in 602 randomly selected houses in different regions of Japan. The number of houses in each region was allocated proportionally to the population size (Uchiyama et al., 2015).

Four types of diffusional passive samplers (Uchiyama et al., 2015) were used in the study: DSD-BPE/DNPH packed with 2,4-dinitrophenyl hydrazine and *trans*-1,2-bis (2-pyridyl) ethylene coated silica for measuring ozone and carbonyls; VOC-SD packed with Carboxen® 564 particles for measuring volatile organic compounds; DSD-TEA packed with triethanolamine impregnated silica for measuring acid gases; and DSD-NH_3 packed with phosphoric acid impregnated silica for measuring basic gases. Acid gases present in the air react with triethanolamine in the absorbent to form corresponding separated anions, such as formate from formic acid, acetate from acetic acid, chloride from hydrogen chloride, nitrite from nitrogen dioxide, and sulfate from sulfur dioxide, and were analyzed by ion chromatography.

Diffusional passive samplers and their attachments were sent to the participants. The samplers were placed near the center of the living room. The sampling time was 24 h in all houses. An instruction manual on the sampling procedure, including practical photographs, was distributed with the sampler. A data sheet to record the sampling time and the room where the sampling took place was also distributed. We analyzed the data based on the information from the recorded data sheet. Air sampling was performed within one week and samplers were sealed in an aluminum bag and immediately returned by mail. On receipt, samplers were stored in a refrigerator (4 °C) and analyzed within two weeks. These sampling procedures have been previously established for quality assurance and control (Yamada et al., 2013).

2.2. Hazard assessment

Inhalation reference concentrations (RfCs) or unit risks (URs) were determined for each pollutant based on the estimating scheme for non-cancer or cancer effects that was used in our previous study (Azuma et al., 2007). Firstly, the latest World Health Organization (WHO) air quality guidelines were primarily selected as RfC or UR. Secondly, we reviewed the latest documents or reports published by international and national agencies to obtain reliable observed effect levels in humans or animals for hazard assessment. The estimated no-observed adverse effect levels (NOAELs) for humans were derived for non-cancer effects (threshold approach) and URs of chemicals with the application of the non-threshold approach in hazard assessment of the agencies were used for cancer effects (Azuma et al., 2007).

Based on the systemic, developmental, reproductive, neurological, immunological, or lymphoreticular endpoints, the lowest sub-chronic or chronic NOAEL or the lowest-observed adverse effect level (LOAEL) was determined from these documents or reports for non-cancer effects. Studies on adverse health effects associated with long-term exposures of humans or experimental animals generally involve discontinuous exposures, such as 6 h per day for 5 days, per week for representing the occupational environment. Therefore, discontinuous exposures were adjusted to continuous exposures of 24 h per day for 7 days per week for the general living environment.

In the hazard assessment, when observed effect levels of inhalation exposure were not identified, NOAEL or LOAEL via inhalation exposure were determined from the observed effect levels of oral exposure studies. This is based on the assumption that chemicals that cause adverse health effects after exposure by ingestion cause the health effects at the same target site after intake into the body by inhalation and vice versa (OEHHA, 2005). To make this conversion, a reference human body weight of 50 kg and a reference human respiration rate/day of 15 m^3 (Japanese people) were used.

For deriving the estimated human NOAEL, uncertainty factors (UFs) were applied to the observed NOAEL or LOAEL. Uncertainty factor 1 (UF1) of 10 was applied to the LOAEL when a NOAEL was not available. Uncertainty factor 2 (UF2) was applied to extrapolation across durations: (I) exposures less than 8% of the expected lifetime were given a UF2 of 10, (II) exposures from 8% to 12% of the expected lifetime were given a UF2 of 3, and (III) exposures greater than 12% of the expected lifetime were given a UF2 of 1 (OEHHA, 2000). Uncertainty factor 3 (UF3) of 10 was applied to extrapolation from animal studies to a human situation.