Science of the Total Environment 417–418 (2012) 61–67

Contents lists available at SciVerse ScienceDirect

Science of the Total Environment

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

A longitudinal study of aldehydes and volatile organic compounds associated with subjective symptoms related to sick building syndrome in new dwellings in Japan

Tomoko Takigawa [a], Yasuaki Saijo [b,*], Kanehisa Morimoto [c], Kunio Nakayama [c], Eiji Shibata [d], Masatoshi Tanaka [e], Takesumi Yoshimura [f], Hisao Chikara [f], Reiko Kishi [g,**]

[a] Department of Public Health, Okayama University Graduate School of Medicine, Dentistry and Pharmaceutical Sciences, 2-5-1 Shikata-cho, Kita-ku, Okayama 700-8558, Japan
[b] Department of Health Science, Asahikawa Medical University, Midorigaoka-higashi 2-1-1-1, Asahikawa 078-8510, Japan
[c] Department of Social and Environmental Medicine, Osaka University Graduate School of Medicine, 2–2 Yamadaoka, Suita 565-0871, Japan
[d] Department of Health and Psychosocial Medicine, Aichi Medical University School of Medicine, Yazakokarimata 1-1, Nagakute, Aichi Prefecture 480-1195, Japan
[e] Fukushima College, 1-1 Chigoike Miyashiro, Fukushima 960-8505, Japan
[f] Fukuoka Institute of Health and Environmental Sciences, 39 Mukouzano, Dazaifu 818-0135, Japan
[g] Hokkaido University Center for Environmental and Health Sciences, Kita 15, Nishi 7, Kita-ku, Sapporo 060-8638, Japan

ARTICLE INFO

Article history:
Received 23 June 2011
Received in revised form 22 December 2011
Accepted 22 December 2011
Available online 20 January 2012

Keywords:
Sick building syndrome
Aldehydes
Volatile organic compounds
Subjective symptoms
Questionnaire

ABSTRACT

To determine whether indoor chemicals act as possible environmental risk factors responsible for sick building syndrome (SBS)-related symptoms in new houses (<6 years old) in Japan, we studied 871 people living in 260 single-family houses in 2004 and 2005. We measured the indoor concentrations of aldehydes and volatile organic compounds and longitudinal changes in the living rooms over two consecutive years. Participants answered standardized questionnaires on SBS symptoms and lifestyle habits. Approximately 14% and 12% of subjects were identified as having SBS in the first and second year, respectively. According to analysis adjusted for sex, age, smoking, and allergic diseases, increases in aldehydes and aliphatic hydrocarbons contributed to the occurrence of SBS. Elevated levels of indoor aldehydes and aliphatic hydrocarbons increased the possible risk of SBS in residents living in new houses, indicating that source controls against indoor chemicals are needed to counter SBS.

1. Introduction

Domestic indoor air quality, especially in new dwellings, should be well maintained for the health and safety of occupants; otherwise, conditions such as sick building syndrome (SBS) may occur. SBS is characterized by various nonspecific subjective symptoms, including irritation of the eyes, nose, and throat; headache; and general fatigue, in rooms with deteriorated indoor air quality (Burge et al., 1987; Finnegan et al., 1984; Lyles et al., 1991; Mendell and Smith, 1990). This syndrome, observed since the 1970s, was acknowledged as a problem in Japan in the 1990s along with increases in indoor chemicals from building materials as the result of the insulation in modern dwellings (Godish, 1994; Saijo et al., 2004; Torii, 2002; World Health Organization, 1983). The problem is referred to as "sick house syndrome" in Japan because it usually occurs in residential dwellings (Ando, 2002; Seki et al., 2007).

Chemical substances have been considered the main factors associated with SBS symptoms in Japan, because most of the people affected live in newly built or renovated houses. The Japanese Ministry of Health, Labour and Welfare promulgated guideline values for indoor concentrations of 14 aldehydes and other volatile organic compounds (VOCs), and an advisable value for total VOCs in 2002 (Ministry of Health, 2002). Clinically, detailed criteria for sick house syndrome have been suggested in Japan with an emphasis on exposure to indoor chemicals (Miyajima et al., 2009). Epidemiologically, we have reported a positive relationship between indoor chemical concentrations and SBS symptoms in localized areas (Takeda et al., 2009; Takigawa et al., 2004).

Although there have been many studies on SBS, most of them are predominantly cross-sectional and target office and building workers (Bakke et al., 2008; Bobic et al., 2009; Brinke et al., 1998; Fisk et al., 2009; Mendell et al., 2008; Mendell and Mirer, 2009; Rios et al., 2009). We explored the association between questionnaire-based responses and measured environmental factors with SBS symptoms in dwellings, but these studies were also cross-sectional (Kishi et al., 2009; Takigawa et al., 2010). A longitudinal study in primary schools measured indoor chemical concentrations only during the last year the follow-up period (Norback et al., 1990). Although some studies have investigated the relationship between SBS symptoms and

* Corresponding author. Tel.: +81 166 68 2401; fax: +81 166 68 2409.
** Correspondence to: Hokkaido University Center for Environmental and Health Sciences North-15, West-7, Kita-ku, Sapporo 060-8638, Japan. Tel.: +81 11 706 5907; fax: +81 11 706 7805.
E-mail addresses: y-saijo@asahikawa-med.ac.jp (Y. Saijo), rkishi@med.hokudai.ac.jp (R. Kishi).

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doi:10.1016/j.scitotenv.2011.12.060

indoor chemical exposures in houses over time, symptoms were measured using questionnaires but chemicals were not measured in these dwelling (Sahlberg et al., 2009; Sahlberg et al., 2010). One study was performed in Japan in which ethnographic methods were used to identify psychosocial factors (Imai et al., 2008).

Our longitudinal study is of particular value because it involved both wide-ranging measurements of environmental chemicals and concurrent questionnaire surveys over a two-year period in many dwellings. The aim of this study was to clarify whether changes in indoor aldehydes and VOCs during two consecutive years affect SBS symptoms.

2. Methods

2.1. Study population and selection of houses

This study was conducted in six Japanese cities from September to December in 2004 and 2005. The cities involved in this study were located in the temperate zone, and the autumn season was chosen as the study period because it is that time of the year when air conditioner usage in Japan is low. In 2003, a preliminary questionnaire survey on indoor environments and SBS was sent to residents of 6080 newly built dwellings to be answered by a representative person from each dwelling. Dwellings $\leq$6 years old by 2003 were randomly chosen from building plan approval applications, which contain official data available for inspection. The number was reduced to 5589 after excluding questionnaires that were returned due to incorrect addresses and those sent to dwellings >7 years old. Responses were received from residents of 2298 dwellings in 2003 (response rate = 41.1%); of which, 1622 residents of 444 dwellings agreed to participate in the 2004 survey. Of the latter, 935 residents of 270 dwellings agreed to participate in the 2005 survey. We have previously reported cross-sectional results of the survey (Kishi et al., 2009; Takigawa et al., 2010). For this study, responses from 871 residents of 260 dwellings participating in both surveys from 2004 and 2005 and were deemed complete and subsequently included in the analysis.

This study was approved by the ethical board for epidemiological studies at Hokkaido University Graduate School of Medicine, the principal investigator's affiliation, and other applicable institutions. All study subjects provided written informed consent.

2.2. Environmental monitoring of chemical substances

Environmental monitoring was performed in each living room once a year from September to November in 2004 and 2005. Air samples were collected onto DSD-DNPH diffusive samplers for aldehydes and VOC-SD diffusive samplers for VOCs (both from Sigma-Aldrich, Tokyo, Japan). Samplers were placed approximately 1.5 m above the floor for 24 h. To eliminate contamination, blank field samples were simultaneously obtained, and the blank results were subtracted from the crude chemical concentrations. Temperature and relative humidity were also measured. Concentrations of 13 aldehydes (including one ketone, acetone) and 29 VOCs were quantified as previously described (Kataoka et al., 2012; Takigawa et al., 2004). In brief, the derivatives in the DSD-DNPH samplers were eluted with acetonitrile before analysis by a high-performance liquid chromatograph equipped with a UV detector. The VOC-SD samplers were desorbed with carbon disulfide before analysis with a gas chromatograph/mass spectrometer. These analysis methods were standardized to compare results obtained during the two survey years. The chemicals assayed included the major components of indoor chemicals detected in Japanese residences (Tanaka-Kagawa et al., 2005). Aldehydes and VOC concentrations lower than the limits of quantification (1.0 $\mu g/m^3$ each) were considered as half the limit of quantification (0.5 $\mu g/m^3$ each).

2.3. Questionnaire study

During both years, participants answered self-administered questionnaires based on the "questionnaire for national investigation for SBS and its potential risk factors in Japan" (Wang et al., 2007). The questions on subjective symptoms of SBS were derived from the Japanese version of the MM040EA, a validated questionnaire designed for epidemiological assessment of SBS symptoms (Andersson, 1998; Mizoue et al., 2001). The questionnaire included symptoms in 12 categories as follows: eye problems (itchy and irritated eyes), nasal problems (running or blocked nose and sneezing), dry throat, cough, dry/flushed facial skin, scaling/itching scalp or ears, dry/itching/red hands, fatigue, heavy-headedness, headache, nausea/dizziness, and concentration problems. The recall period was three months. For each symptom, the responses were "always," "sometimes," and "never." "Always" symptoms were defined as occurring $\geq$3 times per week, and "sometimes" symptoms as occurring once or twice per week. The respondents were also asked whether they attributed the symptom to their home environment. Any "always" or "sometimes" symptoms related to the home environment were designated as positive SBS symptoms. Those who complained about at least one positive symptom were classified as suffering from SBS.

Subjects were divided into four categories according to their SBS status over the two year study period as follows: "newly diseased," "ongoing," "recovered," and "symptom-free." Subjects without an SBS symptom in 2004 but with it in 2005 were categorized as "newly diseased," whereas subjects with a symptom in 2004 but without it in 2005 were categorized as "recovered." Subjects with SBS symptoms in both 2004 and 2005 were categorized as having "ongoing" SBS, whereas subjects without SBS symptoms during both years were categorized as "symptom-free."

The detailed questionnaire also included information on demographics (habitation area, duration of living in newly-built houses, age, and sex); personal characteristics (tobacco smoking, time spent at home, alcohol drinking, and allergic diseases); and indoor environment of the participating dwellings (condensation, fungi, pets, passive smoking, and use of moth repellent and air freshener). If the participants were too young or too old to read or write, another family member answered the questionnaire on their behalf.

2.4. Statistical analysis

Chemical indoor pollutant concentration in the living rooms was matched to each participant. To avoid multiple comparison problems, the concentration of chemical substances was categorized into families as aldehydes, aliphatic hydrocarbons, aromatic hydrocarbons, terpenes, halogens, esters, ketones, and alcohol. Changes in chemical concentrations were determined by subtracting the concentration in 2004 from that in 2005. The Wilcoxon test were used where appropriate. The change in concentration of each chemical family was assessed as a quintile. Odds ratios (ORs) and 95% confidence intervals (CIs) were calculated in an adjusted model. A multiple logistic regression analysis model was used to determine possible risk factors for newly diseased and ongoing SBS symptoms to adjust for confounding factors. We calculated adjusted ORs and 95% CIs for changes in SBS or each symptom over two years. We analyzed the newly diseased SBS risk among the nonSBS at baseline group, and the ongoing SBS risk factors among the SBS at baseline group. The quintile change in concentration of each chemical family was entered separately into the model adjusting for sex, age, tobacco smoking, allergic diseases, and each indoor chemical concentration from 2004. The test for linear trends was evaluated by modeling the quintile values as continuous variables in a logistic regression model, again adjusting for age and sex, tobacco smoking,