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On-site passive flux sampler measurement of emission rates of carbonyls and VOCs from multiple indoor sources

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ABSTRACT

In indoor environments with high levels of air pollution, it is desirable to remove major sources of emissions to improve air quality. In order to identify the emission sources that contribute most to the concentrations of indoor air pollutants, we used passive flux samplers (PFSs) to measure emission rates of carbonyl compounds and volatile organic compounds (VOCs) from many of the building materials and furnishings present in a room in a reinforced concrete building in Tokyo, Japan. The emission flux of formaldehyde from a desk was high (125 $\mu g/m^2/h$), whereas fluxes from a door and flooring were low (21.5 and 16.5 $\mu g/m^2/h$, respectively). The emission fluxes of toluene from the ceiling and the carpet were high (80.0 and 72.3 $\mu g/m^2/h$, respectively), whereas that from the flooring was low (9.09 $\mu g/m^2/h$). The indoor and outdoor concentrations of formaldehyde were 61.5 and 8.64 $\mu g/m^3$, respectively, and those of toluene were 43.2 and 17.5 $\mu g/m^3$, respectively. The air exchange rate of the room as measured by the perfluorocarbon tracer (PFT) method was 1.84/h. Taking into consideration the area of the emission sources, the carpet, ceiling, and walls were identified as the principal emission sources, contributing 24%, 20%, and 22% of the formaldehyde, respectively, and 22%, 27%, and 14% of the toluene, respectively, assuming that the emission rate from every major emission sources could be measured. In contrast, the door, the flooring, and the desk contributed little to the indoor levels of formaldehyde (1.0%, 0.54%, and 4.1%, respectively) and toluene (2.2%, 0.31%, and 0.85%, respectively).

1. Introduction

Since the 1990s in Japan, the construction of buildings that are tightly sealed and have low ventilation rates, and the use of some new types of building materials, have often resulted in indoor air quality problems. Many inhabitants were reported to be suffering from sick building syndrome (SBS) and from multiple chemical sensitivity (MCS) [1,2]. The indoor air guidelines for formaldehyde, acetaldehyde, toluene, ethylbenzene, xylene, and *p*-dichlorobenzene set by the Japanese Ministry of Health, Labour and Welfare are 100, 48, 260, 3800, 870 and 240 $\mu g/m^3$, respectively, based on hazard assessments [3–6]. In Japan, plywood and particle board have recently been classified into four classifications, depending on formaldehyde emission rates as measured using an emission chamber or by desiccator methods [7,8], and the surface areas of each type of material that can be used in house construction are limited by the Building Standards Law [9]. Thus, the indoor concentrations of the chemicals which set the guideline value have been reduced in newly constructed buildings. Unfortunately, formaldehyde can continue to be emitted from resin adhesives even in buildings that are 10 or more years old due to hydrolysis [10], and the indoor concentrations of formaldehyde are therefore still high in many older dwellings. In addition, emission sources of the other chemicals, such as furniture and appliance, still remain in many dwellings. It is therefore necessary to remove major emission sources even in older dwellings in order to improve indoor air quality.

Although it would be desirable to remove the primary emission sources, it is difficult to determine which sources significantly affect the indoor air quality because there are several possible sources in indoor environments, such as flooring, doors, closets, desks, beds, etc. Emission chambers [11–15], the desiccators [16], and field and laboratory emission cells (FLEC) [17–19] have been used to measure the emission rates of chemical compounds from building materials.

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The emission rates from source materials, however, cannot be measured in an actual room by using the chamber or desiccator methods. Thus, the emission sources cannot be identified in an actual room by using these methods. Since the emission rates from building materials can be accurately measured in both the field and the laboratory by using FLECs, FLEC was often used to measure the emission rates from several materials in chamber or houses [20], to obtain the model parameters (diffusion and partition coefficients) [21,22], and to estimate the surface reactions on the materials [23–25]. This instrument is, however, unsuitable for multipoint field sampling because of size, weight, and cost considerations. To identify the emission sources that may be primarily responsible for the indoor pollution, the passive flux samplers (PFS) has been proposed to measure the emission rates from several sources in occupied dwellings [26,27]. The emission rates measured by PFSs were reported to be proportional to those measured by the chamber method [27].

Many researchers proposed the indoor emission source model for several type of sources, such as coating materials, building materials, evaporating solvent pools, pesticide and so on [28–30]. These models can estimate the emission rates from the materials under several conditions in the chamber. Emission rates from indoor sources in occupied dwellings are, however, difficult to be estimated by using these models because the detail characteristics of source materials are often unknown. There are still few data about the relations between the emission rates from indoor sources and indoor concentrations in occupied dwellings.

In this study, emissions of carbonyl compounds and volatile organic compounds (VOCs) from indoor materials were measured in an occupied room to determine the emission sources that contributed most to indoor concentrations of chemicals in the air. In order to evaluate the PFS measurements, the indoor concentrations measured in the room were compared with estimates calculated by non-equilibrium mass balance equations using the emission rates measured by PFSs and the air exchange rate measured by PFT.

2. Methods

Emission rates and indoor concentrations of carbonyl compounds and VOCs, together with the air exchange rate, were measured in an occupied room in a three-story reinforced concrete building in May 27, 2003.

2.1. Measurements in room

The characteristics of the room in which measurements were taken in this study are given in Table 1, and the room layout is shown in Fig. 1. After leaving all the windows and doors open for 30 min, all the windows and doors were closed, and the exhaust fan in the bathroom was left on for about 16 h until the sampling ended. During the sampling, four people were present in the room (Fig. 1).

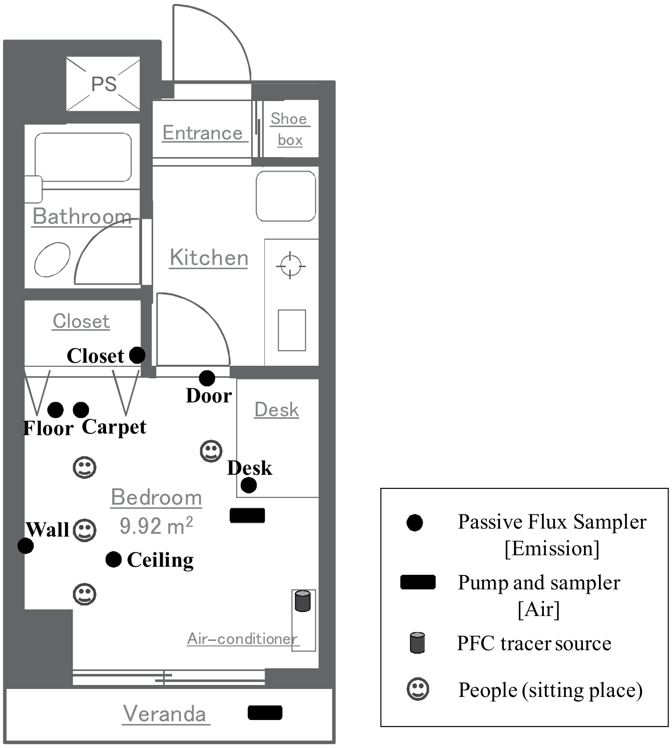

Fig. 1. Layout of room in which measurements were taken, and sampling points in the room.

Table 1
Characteristics of room and building in which measurements were taken.

Building age	Nine years
Location	Kita-ku, Tokyo, Japan
Residential structure	Three-story reinforced concrete building One-room apartment house
Room characteristics	Second floor, south-facing
Room layout	One bedroom, one kitchen, and one unit bathroom
Bedroom volume	22.6 m^3

2.2. Emission flux

PFSs were used to measure the emission flux from each source in the room. 2,4-dinitrophenylhydrazine (DNPH) sheets (OLBO827; Supelco, Co., USA) and Carbotrap B (20/40 mesh, 360 mg, Supelco, Co., USA) were used as the adsorbents for carbonyl compounds and for VOCs, respectively (Fig. 2). The depth of petri dishes (as an approximation of thicknesses of boundary layers at surfaces of materials in the room) was set at 10.0 mm in this study because the thickness of the gas-phase boundary layer was reported as 10–30 mm in four ventilated rooms when the speed of indoor air movement was 50–120 mm/s at 20 mm above the surface [31] and the speed of indoor air movement was high in the present study due to the electrical fan. The PFSs were placed on each of the materials to be sampled. The carpet, ceiling, closet, door, flooring, desk, and wall were selected as likely emission sources in view of surface area. Carbonyl compounds and VOCs, after being emitted from a material, diffuse to the inside of the PFSs and are adsorbed onto the adsorbents (Fig. 3). After 255 min of sampling, the DNPH sheet and Carbotrap B were removed from the petri dishes and were individually stored in aluminized bags at 4 °C until analysis. Adsorbed DNPH-carbonyl derivatives were extracted from the adsorption filter in 5 mL of acetonitrile (HPLC grade, Wako Pure Chemicals Co., Ltd., Japan) by ultrasonication with a W-113 MK-2 (Honda Electronics Co., Japan) for 30 min at 24 kHz. The extracted DNPH-carbonyl derivatives were analyzed using a high performance liquid chromatograph (HPLC; Agilent1100, Agilent technology, USA) with a photodiode array detector at a wavelength of 365 nm. The mobile phase was a 65% acetonitrile and 35% water mixture with a flow rate of 1 mL/min. Twenty microliters of the analyte was injected onto a XDB-C18 packed column (ZORBAX Eclipse; length 250 mm, inner diameter 4.6 mm; particle size 5 μm), which was maintained at 35 °C. Adsorbed VOCs were thermally desorbed with an ATD400 (PerkinElmer Inc., USA) and were analyzed using a gas chromatograph and mass spectrometer (GC–MS; Agilent6890-Agilent5973, Agilent technology, USA). Parameters for the auto thermal desorption system were set as follows: desorption time: 10 min, desorption temperature: 300 °C, desorption flow: 10 mL/min, second trap temperature: 5 °C, second desorption temperature: 320 °C, and outlet split ratio: 1:15. A HP5-MS capillary column