



## A simple method for screening emission sources of carbonyl compounds in indoor air

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### ABSTRACT

Volatile organic compounds (VOCs) emitted from building and furnishing materials are frequently observed in high concentrations in indoor air. Nondestructive analytical methods that determine the main parameters influencing concentration of the chemical substances are necessary to screen for sources of VOC emissions. Toward this goal, we have developed a new flux sampler, referred to herein as an emission cell for simultaneous multi-sampling (ECSMS), that is used for screening indoor emission sources of VOCs and for determining the emission rates of these sources. Because the ECSMS is based on passive sampling, it can be easily used on-site at a low cost. Among VOCs, low-molecular-weight carbonyl compounds including formaldehyde are frequently detected at high concentrations in indoor environments. In this study, we determined the reliability of the ECSMS for the collection of formaldehyde and other carbonyl compounds emitted from wood-based composites of medium density fiberboards and particleboards. We then used emission rates determined by the ECSMS to predict airborne concentrations of formaldehyde emitted from a bookshelf in a large chamber, and these data were compared to formaldehyde concentrations that were acquired simultaneously by means of an active sampling method. The values obtained from the two methods were quite similar, suggesting that ECSMS measurement is an effective method for screening primary sources influencing indoor concentrations of formaldehyde.

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### 1. Introduction

In recent years, buildings have become more airtight to decrease energy consumption. As a result, indoor concentrations of various chemical substances have increased. The presence of these substances in indoor air can cause serious health consequences, and the relationship between indoor air pollution and multi-organ hypersensitivity has been discussed previously [1–5]. Furthermore, some nonspecific symptoms including upper airway irritation, headache, muscle pain, joint pain, nausea, and eye irritation may be related to the presence of such chemicals in indoor air. Among such chem-

icals, volatile organic compounds (VOCs) such as formaldehyde are frequently detected in indoor environments [6–10].

For indoor formaldehyde concentration, the World Health Organization [11] and several countries, including Japan, have set a maximum guideline at  $100 \mu\text{g m}^{-3}$  (0.08 ppm). Building and furnishing materials can be major sources of formaldehyde and other VOCs [12], and in many cases formaldehyde is used as an adhesive for producing wood-based materials [13]. To reduce concentrations of indoor gaseous formaldehyde, new materials are being developed to replace conventional ones containing formaldehyde [14], and techniques for the removal of formaldehyde and other VOCs from indoor environments are also being researched [15,16]. However, there are still documented cases of indoor environments in which the above-mentioned maximum guideline is exceeded [17,18], even though manufacturers have made efforts to reduce the amounts of the chemicals emitted from building and furnishing materials.

Chamber test methods [19–21] and desiccator test methods [22–25] have been used to determine amounts of chemicals emitted from wood-based materials. However, these methods are destructive, requiring cutting of the test materials, and the tests

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must be conducted in laboratory setting. Therefore, the use of such methods to survey on-site emissions from building materials or from large furniture in existing buildings is impractical. Moreover, variations between results from the two kinds of methods mentioned above can arise from differences in testing conditions [26].

For on-site, nondestructive determination of emission rates of VOCs, some devices have been developed, including the field and laboratory emission cell [27,28] and a passive flux sampler [29]. These devices were developed solely for determining emission rates of VOCs. However, so many sources of formaldehyde and other VOCs exist in indoor environments that it is difficult to determine the main parameters influencing indoor air pollution [30] by means of an emission cell or a passive flux sampler. For adequate determination of VOCs, it is necessary to survey an indoor environment simultaneously at many positions including the ceiling, walls, floor, and furniture, and to determine individual emission rates from each of these sites. Therefore, an accurate survey requires many devices as well as simple, straightforward device operation and handling. For these reasons it is necessary to develop a cheaper and simpler method for the determination of VOCs in indoor environments.

Passive sampling techniques have been used for environmental monitoring, and various passive samplers have been developed to determine concentrations of many kinds of chemicals [31]. For example, passive samplers are used to collect nitrogen oxides [32]; sulfur oxides [32,33]; ammonia [34]; ozone [35]; chlorinated hydrocarbons [36]; aromatic hydrocarbons [37]; benzene, toluene, and xylene (BTX) [38]; and carbonyl compounds [39]. Passive samplers are inexpensive, low-maintenance, small, lightweight, and noiseless, and they do not need a power supply. For these reasons, they are particularly well suited for indoor sampling of abundant carbonyl compounds.

Recently, we have developed a screening device that features many of the aforementioned merits of passive samplers. This device has been developed not only for determining emission rates of VOCs from building and furnishing materials but also for screening the VOCs' indoor primary sources. We refer to the developed device as an emission cell for simultaneous multi-sampling (ECSMS), and the device is composed of commercially available materials. Thus, the ECSMS can be used to conduct indoor air surveys at a low cost. The primary emission sources in a polluted room can be determined by simultaneous use of multiple ECSMSs, and tens of ECSMSs can be used simultaneously in a single room because the device is small, lightweight, cheap, and easy to handle.

The objectives of the present study were (1) to determine the reliability of the ECSMS for collection of formaldehyde and other low-molecular-weight carbonyl compounds emitted from wood-based composites of medium-density fiberboards (MDFs) and particleboards (PBs); (2) to estimate the reliability of ECSMS for determining emission rates of formaldehyde from a bookshelf constructed of MDFs and PBs; and (3) to compare the indoor formaldehyde concentrations predicted from the measured emission rates with concentrations measured directly by an active sampling method.

## 2. Materials and methods

### 2.1. ECSMS sampling device

Schematic diagrams of the ECSMS device are shown in Fig. 1. The device is composed of the following parts: (1) a commercially available stainless steel container (area of opening, 94.6 cm<sup>2</sup>; depth, 3.8 cm; volume of inner space, 300 cm<sup>3</sup>) with a polyethylene seal (2) a passive sampler (Passive Gas Tubes 2,4-dinitrophenylhydrazine

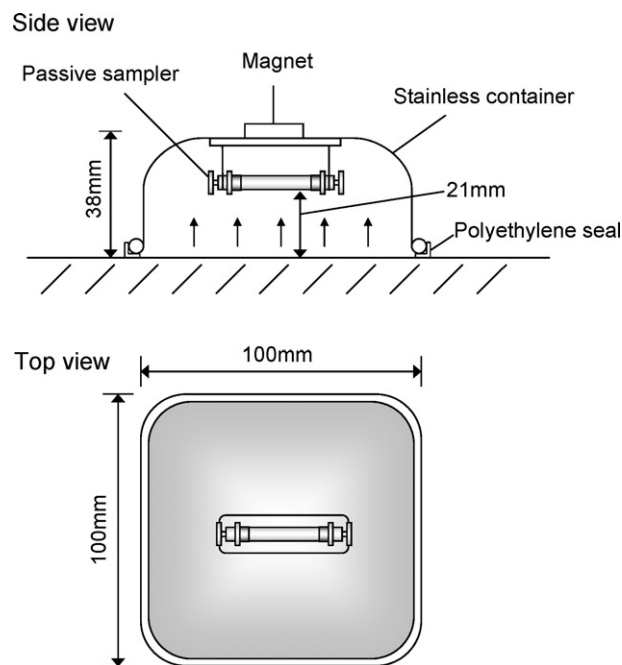


Fig. 1. Schematic diagrams of emission cell for simultaneous multi-sampling (ECSMS). Inside volume is 300 cm<sup>3</sup>, area of opening face is 94.6 cm<sup>2</sup>.

(DNPH) Silicagel [for aldehydes and ketones], Sibata Science Co., Tokyo, Japan); (3) a metal fitting to support the passive sampler; and (4) a magnet to affix the passive sampler, supported by the metal fitting, to the inside face of the stainless steel container. A 500-g weight was placed on top of the device during use.

### 2.2. Materials

The amount of formaldehyde emitted from a 9-mm-thick MDF (150 mm × 150 mm) containing formaldehyde as adhesive was determined by the desiccator method [24]. The formaldehyde emission level for the MDF was 0.5 mg L<sup>-1</sup> (F☆☆ grade, JIS A 5965 [40]). This MDF was used to examine the sampling rates of carbonyl compounds with the ECSMS.

A bookshelf constructed of MDFs and PBs was used to study the reproducibility of formaldehyde emission measurements made with the ECSMS. The dimensions of the bookshelf are shown in Fig. 2. The numbers in the figure indicate positions at which ECSMS measurements were taken. The parts of the positions 16, 17 and 18 were used MDFs, and the other parts were PBs.

Chamber tests were also conducted to estimate the reliability of the ECSMS for determining the emission rate and indoor concentration of formaldehyde originating from the bookshelf (Fig. 3). The inside faces of the chamber were covered with stainless steel 304, and the inside volume was 4.25 m<sup>3</sup>. Temperature and relative humidity were not controlled in these tests but were measured every 10 min with an RTR-53 thermorecorder (T&D Co., Nagano, Japan). A CO<sub>2</sub> monitor (Testo 435 Portable Multi-Function Instrument; Testo AG, Lenzkirch, Germany) was used to determine the air exchange rate in the chamber. Two active samplers (Precep-C DNPH cartridge [Short], Wako Pure Chemical Industries Co. Ltd., Osaka, Japan) connected in series with a miniature pump (MP-Σ30, Sibata Science Co.) were used to determine carbonyl concentrations in the chamber. The sampling rate was 1.0 L min<sup>-1</sup>, and the sampling period was 30 min. The used cartridges were sealed in aluminum bags individually and kept refrigerated at about -45 °C until analysis.

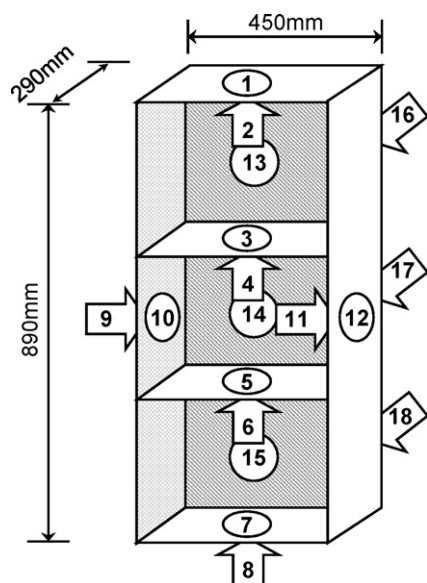


Fig. 2. Schematic diagram of the bookshelf and measurement position number.

### 2.3. Extraction and analysis of carbonyl compounds

The sampled materials were transferred from a sampler (active or passive) to a 10-mL glass test tube and were eluted with 2 mL of 30% acetonitrile aqueous solution. The eluted sample was then stirred for a minute and filtered through a Teflon filter (DISMIC-13HP; pore size 0.45  $\mu\text{m}$ , Toyo Roshi Kaisya, Ltd., Tokyo, Japan). The target carbonyl compounds were analyzed with a high-performance liquid chromatography (HPLC) system consisting of a PU-2089 Plus pump, an AS-2059 Plus autosampler, and a UV-2077 Plus ultraviolet-visible (UV-vis) detector (JASCO Co., Tokyo, Japan) set at 360 nm. A Wakosil-DNPH-II column (4.6 mm i.d.  $\times$  250 mm; Wako) was used to separate the carbonyl

compounds. Acetonitrile, Wakosil DNPH Eluent A, and Wakosil DNPH Eluent B were HPLC grade (Wako), and the water was double-distilled and filtered with a Milli-Q Gradient A10 system (Millipore Co., MA, USA). The column temperature was 35  $^{\circ}\text{C}$ , and the flow rate of the mobile phase was 1.0 mL  $\text{min}^{-1}$ . The HPLC conditions used for analysis of carbonyl compounds are shown in Table 1. The calibration standards contained DNPH derivatives of 16 carbonyl compounds (formaldehyde, acetaldehyde, propionaldehyde, acrolein, acetone, *i*-butyraldehyde, *n*-butyraldehyde, crotonaldehyde, *i*-valeraldehyde, *n*-valeraldehyde, benzaldehyde, *n*-hexanal, *o*-tolualdehyde, *m*-tolualdehyde, *p*-tolualdehyde, and 2,5-dimethylbenzaldehyde; each at a concentration of 10  $\mu\text{g mL}^{-1}$  in acetonitrile solution), and a formaldehyde standard solution (1 mg  $\text{mL}^{-1}$  in methanol solution) also was used. All the chemicals used for calibration standards were obtained from Wako and were of the highest purity available. The calibration range for the carbonyl compounds was 1–100 ng per injected solution.

### 2.4. Quality assurance/quality control

Table 2 shows detection limits and coefficients of variance for the HPLC analytical method. The detection limits were calculated as values of 3 times the standard deviations, and the detection limits ranged from 0.022 ng (acetaldehyde) to 0.076 ng (*p*-tolualdehyde).

### 2.5. Evaluation of the ECSMS

To confirm that the sampling rates of carbonyl compounds measured with the ECSMS were constant, we measured the collected amount of carbonyl compounds for the sampling period. As preparation for this experiment, passive samplers were individually affixed to the inside face of each ECSMS just before use, and the ECSMS was placed on a glass plate until the experiment began. Then the ECSMSs that housed a passive sampler were placed on the surface of the 9-mm-thick MDF mentioned in Section 2.2, and the amounts of carbonyl compounds collected by these ECSMSs were examined at 30, 60, 90, 120, 150, and 180 min. Temperature and relative humidity again were measured every 10 min with an RTR-53 thermorecorder (T&D Co.). The monitored temperature and relative humidity remained nearly constant during the experiment, with a temperature of  $28 \pm 0.5$   $^{\circ}\text{C}$  and relative humidity of  $50 \pm 5\%$ .

To determine the reproducibility of collection of carbonyl compounds by the ECSMS, formaldehyde was examined as a representative carbonyl compound. As preparation for this experiment, five passive samplers were individually affixed to the inside face of each of five ECSMSs just before use, and the ECSMSs were placed on a glass plate. Then a 5- $\mu\text{L}$  formaldehyde standard solution was individually spiked onto each of the five points on the glass plate where the ECSMSs were located. The standard solutions were applied to the plate by means of 10- $\mu\text{L}$  syringes (Hamilton Co., Bonaduz, Switzerland). The samples were allowed to incubate for 2 h, at which point the amounts of formaldehyde collected by the five ECSMSs were compared. In this experiment, temperature and relative humidity were not controlled.

To determine the reproducibility of emissions of carbonyl compounds from the bookshelf, we investigated variations in the amount of carbonyls collected at different positions on the same board of the bookshelf. Five ECSMSs were placed in different positions on the same board simultaneously. Because the bookshelf was made of MDFs and PBs, formaldehyde collection was examined for both types of boards. Measurement positions 5 (PB) and 17 (MDF) (see Fig. 2) were examined for 6 h each. Tests with the two sets of five ECSMSs (one set for MDF and one for PB) were run individually. Temperature and relative humidity were not controlled.

When a chemical substance is emitted from a material into indoor air, two diffusion processes occur: (1) diffusion inside the

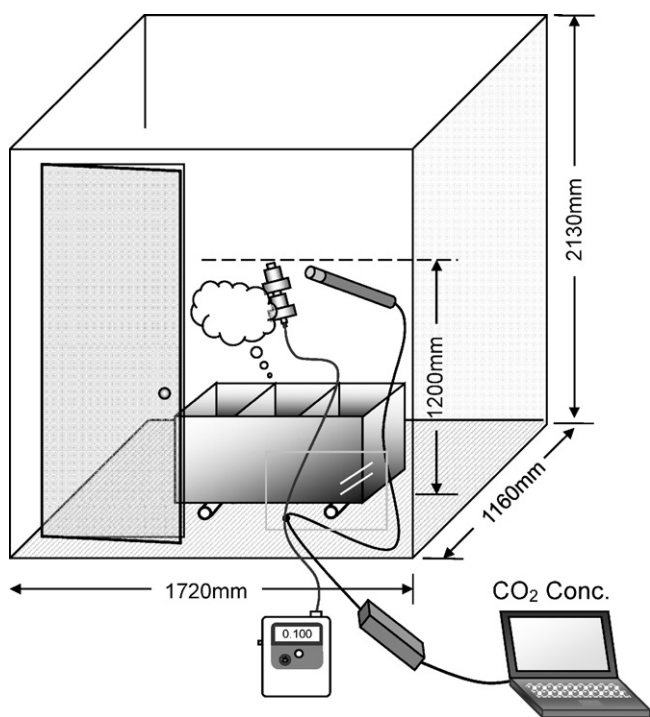


Fig. 3. Schematic diagram of the chamber system. Capacity of the chamber is 4.25  $\text{m}^3$ , quality of the inside is stainless steel 304.

**Table 1**  
HPLC conditions for analysis of carbonyl compounds.

Time (min)	Elution solvents <sup>a</sup> (%)				Elution conditions
	A	B	C	D	
0.0–0.1	70	0.0	30	0.0	70% A and 30% C hold from 0.0 to 0.1 min
0.1–20.0	70–0.0	0.0–70	30	0.0	70% A down to 0.0% and 0.0% B up to 70% over 0.1 to 20.0 min, 30% C hold from 0.1 to 20.0 min
20.0–30.0	0.0	70–100	30–0.0	0.0	70% B up to 100% and 30% C down to 0.0% over 20.0 to 30.0 min
30.0–40.0	0.0	0.0	70	30	70% C and 30% D hold from 30.0 to 40.0 min

<sup>a</sup> A is acetic acid/sodium acetate buffer (30%, pH 7.5) and methanol (70%) buffer solution, B is methanol (98%) buffer solution, C is water and D is acetonitrile.

material and (2) diffusion into a gas-phase boundary layer between the material surface and the indoor air. If process (1) is the rate-determining step, then the emission rate from the material surface is constant regardless of the thickness of the boundary layer. In such a case, we would be able to predict the indoor concentration of a chemical from the emission rate observed at the test material surface. However, if process (2) is the rate-determining step, then a concentration gradient of the chemical substance is present in the boundary layer. In such a case, the chemical concentration in the ECSMS would depend on the diffusion distance between the material surface and the passive sampler and the flux rate would be inversely proportional to the thickness of the boundary layer (Fick's Law):

$$J = -D \frac{dC}{dz} = -D \frac{C_{\infty} - C_0}{L}, \quad (1)$$

where  $J$  ( $\mu\text{g m}^{-2} \text{s}^{-1}$ ) is the chemical flux,  $D$  ( $\text{m}^2 \text{s}^{-1}$ ) is the diffusion coefficient,  $C$  ( $\mu\text{g m}^{-3}$ ) is the concentration of a chemical substance in the gas-phase boundary layer,  $z$  (m) is the diffusion distance,  $C_{\infty}$  ( $\mu\text{g m}^{-3}$ ) is the concentration of the chemical substance outside the boundary layer,  $C_0$  ( $\mu\text{g m}^{-3}$ ) is the concentration of the chemical substance on a material surface, and  $L$  (m) is the thickness of the boundary layer.

In this case, if the thickness of the boundary layer in the ECSMS is shorter than the distance between the material surface and the passive sampler, the emission rate observed with the ECSMS would be constant. In other words, we would be able to predict the indoor concentration of a chemical from the emission rate detected with the ECSMS.

To test this hypothesis, we collected carbonyl compounds at four different distances (6, 11, 16, and 21 mm) between the bookshelf surface and a passive sampler. To conduct this experiment, four ECSMSs were tested simultaneously to collect carbonyl compounds at four respective sites on the bookshelf (measurement positions 3, 10, 14, and 17 in Fig. 2). The magnets mentioned in Section 2.1 were also used as spacer materials to create the four distances. The

thickness of them was 5 mm per unit. The ECSMSs sampled the air for 6 h, and then the amounts of carbonyls collected at each diffusion distance were compared. In this experiment, temperature and relative humidity were not controlled.

## 2.6. Emission tests in a chamber

A chamber environment was used to test the reliability of the ECSMS for determining the emission rate and indoor concentration of formaldehyde from the bookshelf. To ventilate the inside of the chamber, the chamber door was opened slightly and was fixed at that position to ensure a steady air exchange rate.  $\text{CO}_2$  was used as a tracer gas to measure the air exchange rate, and a fan circulated the air in the chamber. A background measurement of  $\text{CO}_2$  was acquired, and then the chamber was filled with  $\text{CO}_2$  from a  $\text{CO}_2$  cylinder to achieve a concentration of 5000–6000 ppm. Air exchange was allowed to occur, during which the decreasing  $\text{CO}_2$  concentration was monitored. The following equation was used to determine the air exchange rate in the chamber:

$$N = \frac{2.303}{t} \times \log_{10} \left( \frac{C_1 - C_0}{C_t - C_0} \right), \quad (2)$$

where  $N$  ( $\text{h}^{-1}$ ) is the air exchange rate,  $t$  (h) is the elapsed time from the beginning of a measurement,  $C_1$  (ppm) is the  $\text{CO}_2$  concentration at the beginning of a measurement,  $C_t$  (ppm) is the  $\text{CO}_2$  concentration at time  $t$ , and  $C_0$  (ppm) is the background concentration of  $\text{CO}_2$ .

For chamber tests in which the bookshelf was used, background concentrations of carbonyl compounds were determined by the active samplers. Two active samplers were connected in series with the miniature pump (MP-Σ30, Sibata Science Co.), and the sampler cartridges were examined for the breakthrough of formaldehyde simultaneously with measurement of the concentrations. The samplers were placed 1200 mm above the floor in the center of the chamber. The sampling rate was  $1.0 \text{ L min}^{-1}$ , and the sampling period was 30 min. The bookshelf was then placed in the cham-

**Table 2**  
Detection limits and coefficients of variance for the analytical method<sup>a</sup> ( $n = 7$ ).

Carbonyl compound	Detection limit (ng)	Coefficient of variance for concentration (%)	Coefficient of variance for retention time (%)
Formaldehyde	0.023	1.08	0.11
Acetaldehyde	0.022	0.88	0.07
Acrolein	0.026	0.89	0.03
Acetone	0.035	0.97	0.03
Propionaldehyde	0.031	0.81	0.03
Crotonaldehyde	0.047	1.75	0.02
<i>n</i> -i-Butyraldehyde	0.043	0.42	0.03
Benzaldehyde	0.046	0.82	0.05
<i>i</i> -Valeraldehyde	0.058	1.59	0.03
<i>n</i> -Valeraldehyde	0.046	2.16	0.05
<i>o</i> -Tolualdehyde	0.044	2.31	0.06
<i>m</i> -Tolualdehyde	0.056	2.00	0.05
<i>p</i> -Tolualdehyde	0.076	2.33	0.04
<i>n</i> -Hexanal	0.037	2.53	0.02
2,5-Dimethylbenzaldehyde	0.052	2.83	0.03

<sup>a</sup> A variance index when it analyzed seven times of standard solution of the same concentration in HPLC.

ber and incubated for 40 h to achieve a steady state for emissions of carbonyl compounds from the bookshelf. Concentrations of carbonyls then were determined by the active samplers, by means of the method described above for the background concentration measurement. These tests were conducted on two occasions, in August 2007 and in October 2007. In addition, the active sampling process was repeated 64 h after the bookshelf was placed in the chamber in October, to confirm that the concentrations in the chamber remained constant. In these experiments, temperature and relative humidity were not controlled but were measured every 10 min with an RTR-53 thermorecorder (T&D Co.). The formaldehyde concentration in the chamber was defined as the difference between the value determined by active sampling with the bookshelf present and the background concentration value (i.e., the reported measurements are background-subtracted).

After the bookshelf had been incubated in the chamber for 40 h and the active sampling measurements were acquired, ECSMSs were installed on the bookshelf boards. The carbonyl fluxes emitted were collected simultaneously by the ECSMSs. The bookshelf consisted of 9 boards, and carbonyl emissions were measured on each of its 18 surfaces as shown in Fig. 2. The bookshelf was placed in the chamber with its backside atop two resin rods to enable measurements of fluxes from the rear face (see Fig. 3). The ECSMSs were left in place for 6 h, and temperature and relative humidity were measured every 10 min with an RTR-53 thermorecorder (T&D Co.).

The carbonyl emission rates were calculated by means of the following equation:

$$F = \frac{P}{at}, \quad (3)$$

where  $F$  ( $\mu\text{g m}^{-2} \text{h}^{-1}$ ) is the emission rate of a chemical substance,  $P$  ( $\mu\text{g}$ ) is the amount of the chemical substance collected,  $a$  ( $\text{m}^2$ ) is the area of the opening of the ECSMS, and  $t$  (h) is the collection period. In this experiment,  $a = 0.00946 \text{ m}^2$  and  $t = 6 \text{ h}$ .

### 3. Results and discussion

#### 3.1. Evaluation of the ECSMS

In the determination of ECSMS sampling rates for carbonyl compounds emitted from the 9-mm-thick MDF, formaldehyde, acetaldehyde, acetone, and *n*-hexanal were collected. The amounts of all four carbonyl compounds collected were proportional to the sampling period ( $R^2 = 0.998$  for formaldehyde, 0.995 for acetaldehyde, 0.993 for acetone, and 0.985 for *n*-hexanal; see Fig. 4). These results show that the ECSMS can simultaneously collect multiple low-molecular-weight carbonyl compounds. It was thus confirmed that the sampling rates of carbonyl compounds collected with ECSMS were constant, even though detection was delayed for a few minutes at the tests' initiation.

In the reproducibility tests for collection of formaldehyde with the ECSMS, the collected amount was  $3.91 \pm 0.84 \mu\text{g}$  (mean  $\pm$  SD,  $n = 5$ ), and the coefficient of variance was 2.1%.

In the reproducibility tests for emissions of carbonyl compounds from the bookshelf, formaldehyde was the major carbonyl collected, and the emission levels of the other carbonyls were remarkably low. Therefore, formaldehyde was chosen as a representative carbonyl compound for further reproducibility tests. The coefficient of variance for formaldehyde collected at bookshelf surface position 5 (PB) was 6.4%, and that for position 17 (MDF) was 9.5%. These values indicate that the ECSMS can be used to reproducibly determine the amount of formaldehyde emitted from MDFs and PBs.

The amounts of formaldehyde emitted from the bookshelf boards at varying diffusion distances (6, 11, 16, and 21 mm) between the board surface and the passive sampler in the ECSMS

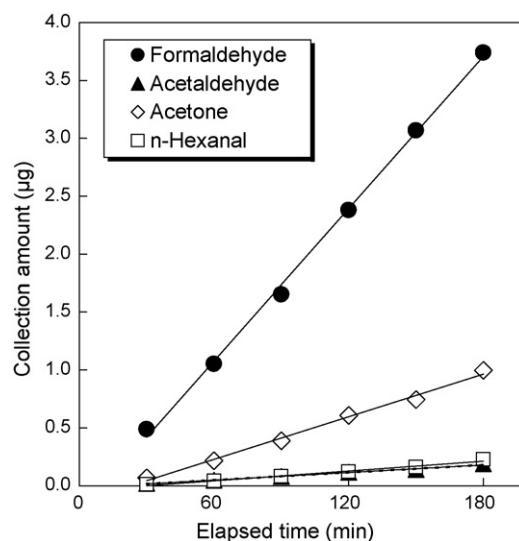


Fig. 4. Relationship between elapsed time and amounts of carbonyl compounds collected.

were determined at four positions on the bookshelf. For position 3, the amounts collected at 6, 11, 16, and 21 mm were 2.52, 2.24, 2.18, and 2.60  $\mu\text{g}$ , respectively; those collected at position 10 were 3.33, 3.44, 3.23, and 3.41  $\mu\text{g}$ , respectively; those collected at position 14 were 0.74, 0.74, 0.79, and 0.76  $\mu\text{g}$ , respectively; and those collected at position 17 were 2.61, 2.71, 2.61, and 2.92  $\mu\text{g}$ , respectively. The coefficients of variance for these formaldehyde amounts collected from each position were 8.7% for position 3, 2.7% for position 10, 3.2% for position 14, and 5.4% for position 17. The differences between these values may be explained by the difference in measurement positions on the bookshelf. Nevertheless, a specific trend in formaldehyde collection was not observed, thus suggesting that the thickness of boundary layer for formaldehyde in the ECSMS was less than the distance between the bookshelf surface and the ECSMS.

#### 3.2. Emission tests in a chamber

The air exchange rate in the chamber before the emission test was  $0.341 \text{ (h}^{-1}\text{)}$ , and that after the test was  $0.325 \text{ (h}^{-1}\text{)}$ . These values indicate that the air exchange rate remained nearly constant during the period of the chamber test. On the basis of these observed values, we used an air exchange rate of  $0.33 \text{ (h}^{-1}\text{)}$  when calculating formaldehyde concentrations in the chamber from ECSMS measurement data.

The formaldehyde background concentrations in the chamber were  $33.0 \mu\text{g m}^{-3}$  in August 2007 and  $22.0 \mu\text{g m}^{-3}$  in October 2007. The results and experimental conditions for the chamber tests are shown in Table 3. The formaldehyde concentrations observed in the October experiment at 40 and 64 h after test initiation were 108 and  $110 \mu\text{g m}^{-3}$ , respectively. These results suggest that the formaldehyde concentration in the chamber remained constant during this time period.

Table 4 shows the formaldehyde emission rates and the emission amounts from each position on the bookshelf, as well as the surface areas of each measurement position. The emission rates for the bookshelf boards determined in the August 2007 experiment were nearly 3 times those observed in October 2007. This difference in observed emission rates probably occurred because of natural variations in the temperature and relative humidity, which were not controlled during the experiments [41–43].

Finally, we predicted indoor air formaldehyde concentrations from the emission rates determined with the ECSMS as follows.

**Table 3**  
Experimental conditions during the chamber tests.

Date	Temperature (°C)			Relative humidity (%)			Formaldehyde concentration in the chamber ( $\mu\text{g m}^{-3}$ )	
	Mean	Max.	Min.	Mean	Max.	Min.	After 40 h <sup>a</sup>	After 64 h <sup>a</sup>
9–12 August 2007	27.5	27.6	27.3	74	74	70	297	–
11–14 October 2007	24.9	26.7	24.3	51	55	47	108	110

<sup>a</sup> The times express elapsed times after the installation of the shelf into the chamber.

**Table 4**  
Areas of the shelf board surfaces ( $\text{m}^2$ ), emission rates of formaldehyde ( $\mu\text{g m}^{-2} \text{h}^{-1}$ ) and emission amounts of formaldehyde ( $\mu\text{g h}^{-1}$ ) (ECSMS test results in August and in October).

Measurement location number of the shelf	Area of the board surface	Emission rate		Emission amount	
		August	October	August	October
1	0.122	180	78.7	22.0	9.58
2	0.116	89.8	26.6	10.4	3.07
3	0.116	140	51.3	16.1	5.93
4	0.116	187	72.1	21.6	8.33
5	0.116	141	58.5	16.2	6.76
6	0.116	187	69.6	21.6	8.04
7	0.116	129	50.3	14.9	5.81
8	0.122	112	44.3	13.7	5.40
9	0.258	169	67.6	43.6	17.4
10	0.237	152	71.1	36.0	16.8
11	0.237	153	76.7	36.1	18.2
12	0.258	191	57.0	49.2	14.7
13	0.116	75.1	24.4	8.68	2.82
14	0.116	79.6	23.9	9.20	2.76
15	0.116	71.9	20.8	8.30	2.40
16	0.116	213	73.1	24.7	8.45
17	0.116	163	58.3	18.8	6.73
18	0.116	162	56.1	18.7	6.48

If the air exchange rate in a room is constant, the amount of a chemical emitted per hour from a source can be expressed by the following equation:

$$E = FA, \quad (4)$$

where  $E$  ( $\mu\text{g h}^{-1}$ ) is the amount of chemical emitted from a material surface and  $A$  ( $\text{m}^2$ ) is the surface area.

The volume of air exchange in the room can be calculated by the following equation:

$$Q = NV, \quad (5)$$

where  $Q$  ( $\text{m}^3 \text{h}^{-1}$ ) is the air exchange volume,  $N$  ( $\text{h}^{-1}$ ) is the air exchange rate, and  $V$  ( $\text{m}^3$ ) is the volume of the room. Therefore, the chemical concentration in the room caused by emission from a source can be expressed as follows:

$$C = \frac{E}{Q}, \quad (6)$$

where  $C$  ( $\mu\text{g m}^{-3}$ ) is the chemical concentration in the room.

Assuming that the emission rates of the chemical substance are constant, the chemical concentration in the room can be calculated from the sum of the chemical emission amounts from the materials as follows.

$$C_C = \frac{\sum_{n=1}^x E_n}{Q} = \frac{\sum_{n=1}^x F_n A_n}{Q}, \quad (7)$$

where  $C_C$  ( $\mu\text{g m}^{-3}$ ) is the chemical concentration in a room,  $E_n$  ( $\mu\text{g h}^{-1}$ ) is the chemical emission amount from a material surface  $n$ ,  $F_n$  ( $\mu\text{g m}^{-2} \text{h}^{-1}$ ) is the chemical emission rate at  $n$ , and  $A_n$  ( $\text{m}^2$ ) is the surface area of  $n$ . For the bookshelf examined in the chamber tests,  $n$  is equal to the sum of positions 1 through 18.

Therefore, the formaldehyde concentrations (278 and  $106 \mu\text{g m}^{-3}$  for August 2007 and October 2007, respectively) predicted from the results of the chamber tests (Table 4) were in

good agreement with the actual measured concentrations ( $297$  and  $108 \mu\text{g m}^{-3}$  for August 2007 and October 2007, respectively; Table 3).

#### 4. Conclusions

We have developed a technique using an ECSMS to screen emission sources of chemical substances influencing indoor air pollution. In this study, the applicability of the ECSMS for determining indoor air concentrations of carbonyl compounds (mainly formaldehyde) was examined. The results show that this method was highly reliable for determining the emission rate of carbonyl compounds from building and furnishing materials. To compare indoor formaldehyde concentrations predicted from the emission rates with those of an active sampling method, chamber tests were conducted in August 2007 and October 2007. The concentrations predicted by the developed ECSMS method agreed with the results obtained by active sampling for both chamber tests. These results confirmed that this low-cost and simple ECSMS method can be useful for screening for indoor emission sources of formaldehyde.

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