## **ORIGINAL PAPER**



# **Changes in the concentration of volatile organic compounds and aldehydes in newly constructed houses over time**

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## **Abstract**

Nowadays, people spend most of their time indoors; thus, the indoor environment greatly afects human health because of exposure to chemicals indoors. Thus, in collaboration with Japanese house builders, a list of building materials having low volatile organic compound concentrations was compiled herein. The air-quality samples from seven newly built houses (seven bedrooms and seven living rooms) were collected and tested for 64 volatile organic compounds and two aldehydes. Air samples were obtained from the house with no furniture or household goods and were sampled 1 week after construction and repeated after a month. Furthermore, the test results with a 2009 survey of indoor air quality in newly constructed houses were compared. One week after construction, the xylene, styrene, toluene, formaldehyde, and ethylbenzene concentrations were less than half the standard guideline values set in Japan. The main substances detected in the samples were 2-butanone, acetone, ethanol, ethyl acetate, butyl acetate, and undecane. The statistical signifcance of the changes in substance concentrations over time was examined via Wilcoxon signed-rank test. One month after construction, the concentrations of all chemical substances had undergone a statistically signifcant reduction, except butyl acetate. The median of total volatile organic compounds in living rooms was 291  $\mu$ g/m<sup>3</sup>, and the maximum was 354  $\mu$ g/m<sup>3</sup>; both were under Japan's provisional total volatile organic compound target of 400  $\mu$ g/m<sup>3</sup>. These values were significantly lower than those recorded by other researchers. This study concludes that using experimentally identifed low volatile organic compound materials efectively improves the indoor air quality.

**Keywords** Environmental preventive medicine · Indoor air quality · Sick building syndrome · Total volatile organic compounds

# **Introduction**

With recent technological developments, houses are now constructed to be more energy efficient and new construction emphasizes airtightness and heat insulation. Improved airtightness and heat insulation has had the unexpected

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consequence that the volatile organic compounds (VOCs) that are emitted from interior materials and buildings probably circulate within the room, which deteriorates the indoor air quality (IAQ). Indoor concentrations of VOCs generally exceed the concentrations outdoors (Begerow et al. [1995](#page-8-0); Shields et al. [1996\)](#page-9-0). On average, people spend~90% of their time indoors in a day (Klepeis et al. [2001](#page-8-1)). Therefore, IAQ has a great effect on human health.

VOCs have been suspected as one of the major causes of the symptoms that are grouped together as "sick building syndrome" such as fatigue, asthmatic symptoms, eye and mucous-membrane irritation, and headache (Norbäck et al. [1995](#page-9-1); Wieslander et al. [1997](#page-9-2); Wolkoff et al. [1998](#page-9-3); Seife [1999;](#page-9-4) Norbäck et al. [2000;](#page-9-5) Wolkoff and Nielsen [2001](#page-9-6); Cox et al. [2002;](#page-8-2) Mølhave [2003](#page-8-3); Katsoyiannis et al. [2008](#page-8-4); Herberth et al. [2009;](#page-8-5) Heinrich [2011](#page-8-6); Mori and Todaka [2011](#page-8-7); Azuma et al. [2016\)](#page-8-8). Reducing the volatile compounds in



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indoor atmospheres is an important step in preventing these symptoms. Therefore, governing bodies including the World Health Organization have instituted guideline concentrations for several VOCs in indoor air. Many national building codes also state that new houses and building must have safe and healthy IAQ (Japanese Ministry of Health Lab and Welf [1997;](#page-8-9) Finland's Ministry of the Environment [2002;](#page-8-10) The Swedish National Board of Housing, Building and Planning and Boverket [2002](#page-9-7); Ministry of Transport Building and Housing of Denmark [2018\)](#page-8-11).

Several studies have focused on the IAQ of newly built buildings and its change over time (Seifert [1992](#page-9-8); Wolkoff [1999;](#page-9-9) Scholz and Santl [1999;](#page-9-10) Bornehag and Stridh [2000](#page-8-12); Rehwagen et al. [2003](#page-9-11); Raw et al. [2004](#page-9-12); Järnström et al. [2006\)](#page-8-13). These studies indicate that the concentrations of VOCs in the indoor air of newly built buildings are much higher than the guideline values specifed by each country, and Holøs et al.  $(2019)$  indicated that the off-gassing phase would last at least 2 years. However, Tuomainen et al. [\(2001\)](#page-9-13) showed that VOC concentrations in new houses constructed using low-emitting materials could be reduced by ventilating the houses for 1 week, and Noguchi et al. ([2016](#page-8-15)) also concluded that operating the ventilation system for 3 months would efectively reduce VOCs in newly constructed buildings. Herein, in cooperation with a residential building company, we investigated whether the IAQ of seven newly built houses, constructed using low-emission materials, could achieve good air quality; we also noted how much the VOC concentration decreases over time, specifcally after 1 week and 1 month of the construction. This study frst identifed the following fve compounds: styrene, formaldehyde, ethylbenzene, toluene, and xylene, which are mentioned in "Housing Quality Assurance Act" by the Japanese Ministry of Land, Infrastructure, Transport, and Tourism; this act was enacted to promote quality housing construction. Then, we checked the concentration values of the six most abundant substances and the TVOC to compare our results with that of another study.

As preliminary tests, the emission rates of these scheduled compounds from building materials (flooring, wallpaper, and substrate panel) were measured in a small test chamber, and low-emission materials were selected and used for the construction of the houses.

Indoor airborne chemicals are emitted not only from construction materials but also from furniture, household goods, and personal care products. Therefore, to evaluate the effect of construction materials on IAQ, measurements must be taken before furniture and household goods are brought into the rooms. Conversely, once construction is completed, houses are typically handed over to the owner in short order. Some studies have sampled the air in newly built houses before delivery, but few studies have taken measurements of the same house at two diferent times, without the infuence



of furniture and household goods. It is difficult to measure indoor air chemicals in newly built houses in the same situation at diferent times, and no studies which compare those results were found. Thus, seven newly built houses (seven bedrooms and seven living rooms) for 64 VOCs and two aldehydes were checked at diferent times and compared in this study. These measurements were performed together with records of temperature and humidity, and samples were collected within 1 week of the end of construction and again 1 month later. None of the homes had furniture and household goods inside during the 1-month period between samples. For the sake of this research, the move-in dates were delayed by 1 month after completion, in cooperation with the house owners.

The houses measured in this study were built in and around Tokyo by a single residential building frm. Seven houses were chosen from houses built by this frm between March 2016 and February 2017. The data were collected between March 2016 and March 2017 and were analyzed at the Center for Preventive Medical Sciences, Chiba University, and MC Evolve Technologies Corporation. The present study, referred to as IAQ-S 2015 below, is also compared with data from a study of newly built houses in Japan conducted by Onuki A et al. in [2009](#page-9-14) (IAQ-S 2009) to gain insight into the characteristics of IAQ in newly built houses at these times (Onuki et al. [2009\)](#page-9-14).

# **Materials and methods**

## **Characteristics of houses**

The owners of these houses were asked for permission to measure the levels of VOCs and aldehydes in indoor air shortly after construction, with completion judged as the last fnishing nailing. The targeted houses were all framed with light-gage steel. Wood floors (solid wood, painted finish, and sheet fnish) were installed in all rooms. All walls were either covered with wallpaper or applied with starch adhesives.

#### **Indoor air sampling and analysis**

Herein, measurements based on "Standard methods of air sampling and measurement" issued by the Ministry of Health, Labour and Welfare of Japan (MHLW [2000\)](#page-8-16) were performed. All windows and doors were left open for 30 min for ventilation before collecting the air samples; all open doors and windows were closed for>5 h. Air samples were then collected from the living rooms and bedrooms of the houses via active sampling for 30 min using the apparatus shown in Fig. [1](#page-2-0). The rooms' ventilation systems were engaged, and their air conditioning units were turned of <span id="page-2-0"></span>**Fig. 1** Equipment setup used for active sampling **a** living room and **b** bedroom



<span id="page-2-1"></span>**Table 1** List of analyzed aldehydes and VOCs



while collecting the samples. The environmental factors of temperature and humidity were recorded. These samples quantifed 64 VOCs and two aldehydes (Table [1](#page-2-1)). We conducted the sampling and analysis according to ISO 17025, and HPLC analysis and GC/MS analysis were performed according to ISO16000-3 and ISO16000-6, respectively.





## <span id="page-3-0"></span>**VOCs**

Tenax-TA® (porous polymer resin based on 2,6-diphenylene oxide), purchased from Sigma-Aldrich, was used for active sampling of VOCs. These tubes were pumped with the sampled air at 100-mL/min flow rate for 30-min collection period. The air samples were analyzed after collection using an Agilent 6890 gas chromatograph (Agilent Technologies Inc.) equipped with a TurboMatrix ATD unit (PerkinElmer, Inc.) and an Agilent MSD 5973 N quadrupole mass spectrometer. The Tenax-TA was heated at 260 °C for 10 min for thermal desorption. The transfer-line temperature and split ratio were 220 °C and 7:1, respectively. The gases were separated for 2 min using HP-VOC columns (60 m $\times$ 0.32 mm; inner diameter 1.8 µm) (Agilent Technologies Inc.) at 35 °C. Columns were frst heated to 95 °C at 15 °C/min. These were then heated to 105  $\degree$ C at 2.5  $\degree$ C/min. Columns were finally heated to 250 °C at 5 °C/min. As the carrier gas, helium (purity > 99.9999%) was used for chromatography; it was then flowed into the column at  $3.0$  mL min<sup>-1</sup>. Recovery rates of toluene, ethylbenzene, xylene, styrene, and tetradecane in Tenax-TA was 96~106 (%). The identifed VOCs were quantifed in scan mode (at mass-to-charge, or m/z, ratios of 35–550). Each compound's standard substance was used for determining the 64 VOCs' concentration levels. As the toluene equivalent of all substances, TVOC was calculated with carbon lengths from C6 (n-hexane) to C16 (hexadecane).

#### **Aldehydes**

2,4-Dinitrophenylhydrazine (DNPH) cartridges that were designed for ketones and aldehydes (GL Sciences Inc.) were used for aldehyde measurements. Measurements were conducted at a pump flow rate of 1.0 L/min for the collection period of 30 min. The gas tube was eluted after sampling using a syringe containing 10 mL of acetonitrile at a flow rate of 2–5 mL/min. The volume was set to 5.0 mL for high-performance liquid chromatography (HPLC); the analysis was conducted using HPLC Agilent 1260 Infnity LC from Agilent Technologies. Inertsil ODS-SP columns  $(250 \times 4.6 \text{ mm}^2)$ ; internal diameter 5.0 µm) from GL Sciences were used for separation, with a column oven temperature of 40  $\degree$ C and an injection volume of 10 µL. A mixed solution containing acetonitrile and water was the mobile phase;



1.0 mL/min was the column fow rate. Acetonitrile/water solution (52:48 mixing ratio) was fowed for 15 min followed by acetonitrile/water (mixing ratio 52:48) for 20 min, and then acetonitrile/water (mixing ratio 75:25) for 23 min. Concentration levels of two compounds were calculated using the standard substance of each compound. Recovery rates of formaldehyde and acetaldehyde in DNPH cartridge was  $101~105~$  (%).

#### **Emission rate test**

In a chamber of 20 L, the emission rate tests were conducted using the small-chamber method published in JIS A 1901 (Ministry of Land, Infrastructure, Transport and Tourism and JIS [2015](#page-8-17)), which is based on ISO 16000-9. The test conditions were adjusted to obtain the desired test temperature, relative humidity, ventilation frequency, and test loading rate of 28  $\degree$ C, 50%, 0.5 times/h, and 2.25 m<sup>2</sup>/m<sup>3</sup>, respectively. The emission rates for the two aldehydes and 64 VOCs were measured on the seventh day after confning the plates in the small chamber.

#### **Statistical analysis**

The Wilcoxon signed-rank test was performed on all data to determine whether the concentration of chemical substances had undergone a statistically signifcant change over the month after measurements conducted by us. All analyses were conducted using the IBM SPSS Statistics package, version 24. A statistically significant result of  $p$  values <0.05 was considered.

# **Results and discussion**

## **Indoor environment characteristics**

Table [2](#page-3-0) shows the median and maximum temperatures and relative humidity levels in the indoor environments.

Within 1 week after construction, the temperatures of the living rooms were in the range  $12.0-20.5$  °C with a mean  $\pm$  SD of 13.9 °C  $\pm$  3.8 °C, and the bedroom temperatures were in the range  $11.5-15.8$  °C with a mean  $\pm$  SD of 13.6  $\degree$ C  $\pm$  1.6  $\degree$ C. One month after construction,

the temperatures of the living rooms were in the range 12.2–30.2 °C with a mean  $\pm$  SD of 17.5 °C  $\pm$  8.5 °C, and the bedroom temperatures were in the range 11.9–27.6 °C with a mean  $\pm$  SD of 16.7 °C $\pm$ 6.9 °C.

The humidity range in the living rooms for the frst measurement was 48–68% with a mean  $\pm$  SD of 51.3%  $\pm$  7.9% and that in the bedrooms was  $47-78\%$  with a mean  $\pm$  SD of  $55.8\% \pm 12.6\%$ . One month after construction, the humidity range in the living rooms was  $42-70\%$  with a mean  $\pm$  SD of  $53.5\% \pm 12.0\%$  and that in the bedrooms was 44–76% with a mean  $\pm$  SD of 57.9%  $\pm$  11.1%.

Table [3](#page-4-0) shows the median and maximum values of the five substances listed in the Japanese "Housing Quality Assurance Act," as measured in the present study, listed along with the data from IAQ-S 2009 for comparison.

Table [4](#page-5-0) lists the median and maximum concentrations of the six most abundant substances and the TVOC, along with data from IAQ-S 2009. At 1 month after the construction, TVOC had a median value of 291  $\mu$ g/m<sup>3</sup> and a maximum of 354  $\mu$ g/m<sup>3</sup> in the living room, and a median value of 189 μg/m<sup>3</sup> and a maximum of 310 μg/m<sup>3</sup> in the bedroom; these results confrm that the houses met the TVOC target of 400  $\mu$ g/m<sup>3</sup> in Japan. These concentrations were also much lower than those recorded in IAQ-S 2009.

The statistical signifcance of the change in IAQ over the month between these samples was determined via the Wilcoxon signed-rank test. The median values of the top six substances decreased signifcantly, except for ethyl acetate.

In the living room and bedroom, airborne chemical concentrations were signifcantly lower 1 month after the construction than at 1 week after construction (Fig. [2](#page-6-0)).

All five targeted substances concentrations were below the guideline values both 1 week after construction and 1 month after construction. Furthermore, those concentrations were less than half the guideline values specifed by the national act.

The concentrations of standardized substances in the houses tested by us were found to be lower than those data from IAQ-S 2009.

All concentrations were lower than the guideline values by about 0.67 to 0.1 times. The diference in the maximum values was even larger, with the values lower by about 0.25 to 0.003 times. In the houses tested by us, the concentrations of all substances were sufficiently lower than the guideline values. The median values of the top six substances were almost equal. The maximum values in this study were about 0.43 to 0.03 times lower than the maximum values recorded in the study IAQ-S 2009. As for TVOC, the median value in this study was about 0.33 times lower and the maximum value about 0.05 times lower than the data from 2009. Since the measurements were completed within 6 months after the completion of construction in IAQ-S 2009, it was



**Table 3** Changes in the concentrations of fve regulated compounds

Table 3 Changes in the concentrations of five regulated compounds

dND:Not Detected

<span id="page-4-0"></span>ND:Not Detected

cThese fve substances are described in the Japanese law about the promotion of quality assurance in housing



cThe six most abundant substances

The six most abundant substances

 $\Delta T$ VOC as the toluene equivalent of substances with carbon lengths from C6 (n-hexane) to C16 (hexadecane)

 $\rm{}^{4}TVOC$  as the toluene equivalent of substances with carbon lengths from C6 (n-hexane) to C16 (hexadecane)

<span id="page-5-0"></span> $\mathcal{D}$  Springer

emission source. With this preliminarily data about the low-VOC building materials, builders will be able to build healthier houses. In Europe, a list of substances and associated emission lim-

its, EU-LCI values, have been developed and were issued in (European Collaborative Action [2013](#page-8-19)). In Japan, on the other hand, a labeling system for building materials is only prescribed for formaldehyde. Development of such improved labeling on building materials is expected also in Japan to protect the people's health.

IAQ than those tested in 2009. At the second sampling, the temperature was higher by 2.0–2.7  $\degree$ C at the median value than the first time. Among the reported environmental parameters, temperature infuences the VOC emissions from building materials (Bremer et al.  $1993$ ; Wolkoff  $1998$ ; Yang  $1999$ ); therefore, these diferences in the temperature likely had some efects on the concentrations of VOCs in the second samples; a slight increase in temperature led to a significant decrease in chemical concentrations in the second samples even with this effect. The results indicate that the concentrations were at healthy levels according to the guideline values by the national act even 1 week after construction ended, and that the risk of chemical exposure was greatly reduced after 1 month. From this result, it may be preferable to move to

emphasized that the IAQ-S 2015 houses had much better

new houses 1 month after the construction. The emission rates of interior and building materials chosen in this study are shown in Table [5.](#page-7-0) These materials were examined by the chamber test. All five substances mentioned in the national act were present in very low concentrations, and estimating those emission sources was difficult. Regarding the six most abundant substances, as the interior materials used in the foors, walls, and ceilings are considered to have almost the same contribution rate, the emission source was estimated from the results of the emission rate test. The contribution rates of the sash windows, substrate panels, and doors are low considering the area used, but large amounts of adhesive and sealing agents are utilized in those materials, and it is assumed that solvent chemicals such as 2-butanone and acetone are emitted from them. Acetone also originates from the flooring (solid wood), and ethanol primarily originates from the foorboards (painted fnish and solid wood), sash windows, and doors. Ethyl acetate is provided by the flooring (sheet finish and solid wood) and substrate panels. Flooring and substrate panels are also assumed to be the main sources of butyl acetate. The emission of undecane was not detected in the preliminary chamber tests conducted; however, a previous study found its volatilization from floor adhesives, foor waxes, and wood stains (Sarigiannis et al. [2011](#page-9-17)). More research is needed regarding the sources of this compound. All fve substances mentioned in the national act were at low concentrations. It was difficult to estimate the



<span id="page-6-0"></span>**Fig. 2 a** Changes in 2-butanone concentration over time, **b** changes in butyl acetate over time, **c** changes in ethyl acetate concentration over time, **d** changes in acetone concentration over time, **e** changes in etha-

nol concentration over time, **f** changes in undecane concentration over time, and **g** changes in TVOC concentration over time





<span id="page-7-0"></span><sup>a</sup>ND: Not detected<br>bBuilding materials selected this study bBuilding materials selected this study aND: Not detected

Furthermore, in the houses examined by us, the six most abundant substances have no guideline values in Japan's national standards. Although the health risks due to these compounds have not been clarifed, unregulated substances have often been used as substitutes for regulated solvents. In recent years, some reports have indicated their adverse health impacts on humans (Yu and Crump [1998;](#page-9-18) Ravindra et al. [2001;](#page-9-19) Kamijima et al. [2002;](#page-8-20) Tuomainen et al. [2004](#page-9-20); Gallego-Iniesta García et al. [2010](#page-8-21)). Thus, to prevent diseases and symptoms which are caused by indoor air pollutants, the concentrations of these unregulated substances should still be reduced and minimized.

# **Conclusion**

The reduction in the concentration levels of the five substances, which are regulated by national act, six most abundant substances, and TVOC in the newly built houses over 1 month, was confrmed. The results in this study suggest that the concentrations would possibly be low enough even 1 week after construction ended, and that the concentrations of chemical substances reduced even further after a month if the building materials are carefully chosen.

The research group is constantly updating a database of low-VOC building materials (Suzuki et al. [2018\)](#page-9-21). By choosing low-VOC building materials in advance, houses with low concentrations of volatile chemicals can be constructed.

As a next step of the research, measuring the indoor air chemicals with furniture will be planned. The airborne chemical substances in a room often are estimated to arise from furniture and other home goods. Additionally, investigating the intervention of the efficiency of ventilation systems will be planned.

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