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Diffusion Characteristics of Chemicals Causing Sick-Building Syndrome

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Polluting concentrations of various chemical substances as a function of height from a floor are examined using a tin oxide odor sensor. The sensor has a high sensitivity for ammonia and alcoholic gases. The chemicals examined are formaldehyde, toluene, xylene and ethyl alcohol. All these chemicals cause sick-building syndrome. Sensor-output characteristics differ with each height. Exposure volume is derived by integrating the output characteristics. Integration is carried out on a time axis. The lower the height, the larger the exposure volume becomes. The volume as a function of height is expressed by a linear function, $y = ax + b$. In this expression, y is the volume and x the height. Four expressions of four chemicals examined are introduced. The coefficients of a and b in the expressions are also indicated as a linear function, $b = 0.077a + 0.978$. As for the results, the closer a person is to the floor, the higher the danger. It is thought that the rate indicates a harmful grade. Namely, the bedridden elderly, babies and newborns are harmed, with high probability, by chemicals such as volatile organic compounds.

1. Introduction

Various problems of natural conservation have appeared due to changes for the worse in the environment of the earth. These problems are caused by rapid economic growth and the desire for a comfortable life for human beings; therefore it is necessary to amend our lifestyle and enhance our environmental consciousness. Human beings must have a sense of crisis, especially with regard to the problems associated with carbon dioxide. The volume of various chemical pollutants generated must be kept within the limits of the cleansing ability of the earth. Buildings have been become very airtight and insulated to save heat energy. In Japan, the use of energy-saving architecture has grown remarkably since the 1970's.

Several kinds of indoor environmental problems have appeared as buildings become airtight.[1] One is the sick building syndrome.[2] This syndrome causes occupants to experience asthma, allergies, languid feelings and to decline in numbers. Over 50% of the population has some allergies. The problem of this disease was taken up early in the 1900's in Europe and America. In Japan, it was highlighted in an interpellation of the National Diet in 1996. Major chemicals causing the syndrome are formaldehyde, toluene, xylene and plasticizers. The former three chemicals are also addressed seriously on TV.

It is indispensable to understand the direction of spread and the exposure volume of the chemicals for a resident in an indoor environment. The exposure volume is defined by integrating the sensor output characteristics at a position in an experimental room on a time axis. It is estimated that the spread mainly occurs by gas diffusion and is also influenced by other factors.[3,4] In this study, diffusion experiments for the three chemicals and ethyl alcohol were carried out in a real domicile and the propagation of the chemicals was approximately understood. Results show that the exposure volume for each chemical is larger as the position of detection is lower, namely closer to the floor. The relationships between the volume and the height can be described as a linear function.

2. Experimental

Experiments were carried out in a real domicile which was built for a welfare investigation by the Ministry of International Trade and Industry of Japan. Four kinds of metal oxide odor sensors were installed at twelve points in the domicile.[5] Materials and highly sensitive chemicals are shown in Table 1. CH-E2 and CH-E3 have high sensitivity to volatile organic compounds (VOCs). CH-N has high sensitivity to ammonia, and AET-S to hydrogen sulfide gas. A photograph of these four sensors is shown in Fig. 1.[6]

The material in these four sensor-elements is a metal oxide semiconductor. Only AET-S is made of zinc oxide and the other sensor-elements are made of tin oxide. In this experiment, sensor arrays are installed at several points in a residential space and gas propagation can be understood when there is an air pollutant which has a detectable concentration.[7] The experiments, however, are carried out to understand the concentration changes of the chemicals as a function of height from the floor using a stand. Three sensor arrays are installed on the stand at the heights of 72, 132, and 195 cm from the floor.

Table 1
Odor sensors used fabricated by New COSMOS Inc. of Japan.

Type	Material	Highly sensitve gases and odors
CH-E2	SnO_2	VOC, CH_3CHO, CH_4, alcohol
CH-E3	SnO_2	high-sensitivity type of CH-E2
CH-N	SnO_2	NH_3, C_2H_5OH, CH_3OH
AET-S	ZnO	H_2S, CH_3SH, CH_3SCH_3, CH_3SSCH_3

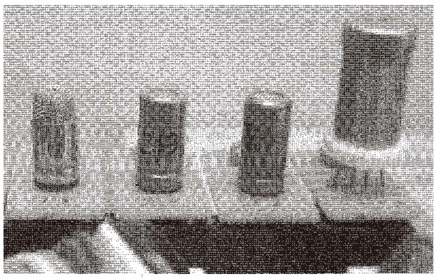

Fig. 1. Photograph of the odor sensors employed.

Four types of odor sensors are assembled in the unit. The positions and distances of the stand and polluting sources are indicated in Fig. 2. The height of the ceiling is 245 cm. The following chemicals are chosen as pollutants: formaldehyde, toluene, xylene and ethyl alcohol. Ethyl alcohol is also harmful to a person at high concentrations and long-term exposures.

Metal oxide semiconductor odor sensors are adopted to detect the concentration of an air pollutant. A gas detecting tube is generally used to detect a VOC.(8) CH-N output characteristics are used to derive the exposure volume. Other sensor outputs can be also used. It is necessary to use four kinds of sensors to identify the gas present.

3. Results and Discussion

It is very important to examine the distribution and concentration of an air pollutant in an indoor environment. The results are also useful in designing the building plan of a house. The diffusion distribution and the concentration of an existing pollutant are influenced by various environmental factors, namely temperature, humidity, air movement, time lapse and the design of the house. It is difficult to analyze all these matters. The experiments were performed under conditions in which all windows and doors were closed. VOCs usually arise from a board and curtain. Four kinds of pollutants were placed

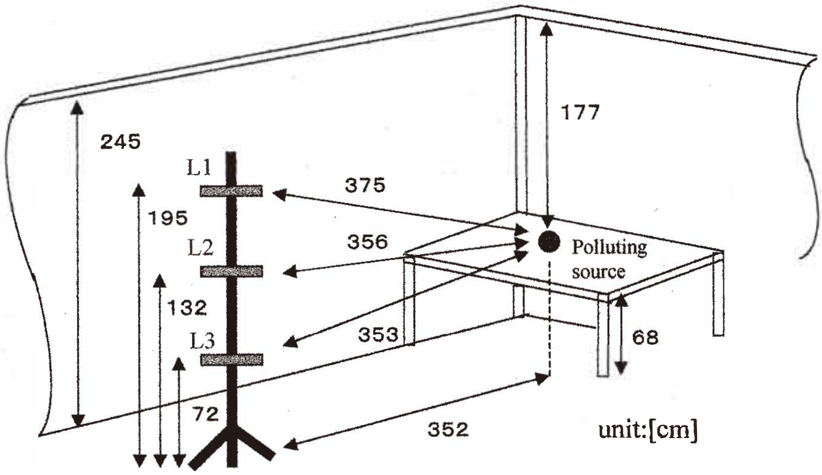

Fig. 2. Position of the sensor stand and polluting source.

on the table, as shown in Fig. 2. A formaldehyde solution (purity 37% + methanol 8%) 4 ml in volume was used as a pollutant. The volume of the other pollutants was 2 ml to roughly adjust the concentration to that of formaldehyde. CH-N outputs on the stand as a function of time were recorded every 2 s by a computer. Each pollutant was introduced 10 min after the beginning of the experiment and the pollutant was removed after 20 min and one of the windows slightly opened. The L3 on the stand (72cm) and the polluting source (68 cm from the floor) are almost same height above the floor.

The results for formaldehyde are shown in Fig. 3. There are three characteristics at the height positions shown in the figure, namely at L1, L2 and L3. The output level is higher with decreasing height of the sensor from the floor. Accordingly, the concentration becomes higher at lower levels. The sensor receptors responded 6 min after the pollutant was introduced. We can understand that the outputs increase slightly a minute after inserting the pollutant if the sensing responses are magnified. Exposure volume can be derived by integrating the sensor output from the time of introduction of a pollutant to the time of removal on the time axis. The interval is 20 min. Each derived volume is indicated in the figure. The volume in the parentheses indicates a value normalized by the value at L3. In this figure, note that the characteristics at L3 temporarily take a higher level than L2 and L1.

The exposure volume is larger with decreasing height, and the risk of dangerous exposure increases. Occupants are exposed largely by the pollutants at lower levels.

The results for toluene (purity 99.5%) are shown in Fig. 4. The exposure volume becomes higher as the position is lower. This phenomenon is the same as in the case of formaldehyde. Sensor outputs due to toluene and xylene fluctuate largely at L3. These phenomena are observed for all pollutants examined. The results for xylene (purity 99.5%)

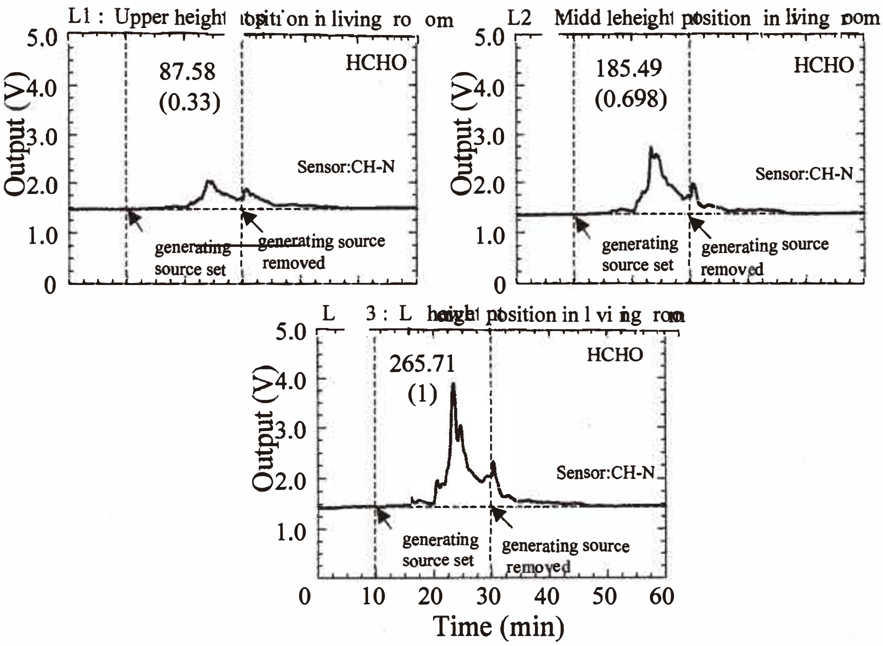

Fig. 3. CH-N sensor outputs for formaldehyde at the positions L1, L2 and L3 on the sensor stand.

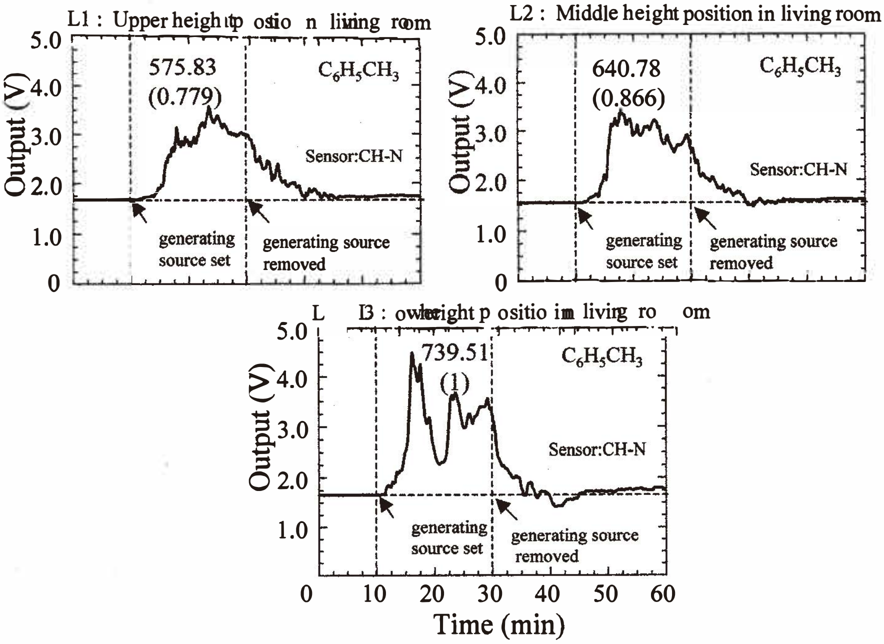

Fig. 4. CH-N sensor outputs for toluene at the positions L1, L2 and L3 on the sensor stand.

are shown in Fig. 5 and the results for ethyl alcohol are indicated in Fig. 6. The exposure volume for ethyl alcohol is very high because the sensor has high sensitivity to the gas. Each exposure volume of the pollutants examined using the sensor's output characteristics cannot be compared to the others. The volume decreases at higher positions for all examined pollutants. It is necessary to pay attention to bedridden elderly, babies and infants because they spend many hours at a lower position in a room. This must also be taken into account while an occupant is asleep. The important countermeasures are ventilation and foliage plants.(9) Each normalized exposure volume for the four chemicals examined, which is also called the exposure rate, is plotted in Fig. 7. The horizontal axis indicates height. The height of the ceiling from the floor is 245 cm. The characteristics are indicated by a nearly linear function. The exposure rate clearly decreases as the height of measurement increases. Although the gradient of the function for formaldehyde is large, it does not mean that the chemical is more dangerous than other chemicals. The slope depends on the sensor sensitivity to the chemicals. Approximate equations, which are expressed by a linear function ($y = ax + b$), are also indicated in the figure. The exposure rate at a certain height can be estimated using these functions. Coefficients a and b of the functions and physical chemistry constants for the chemicals examined are summarized in Table 2. Dispersions in two-dimensional space for coefficients a and b are shown in Fig. 8. In the figure, the horizontal axis is ($-a*10^3$) and the vertical axis is b. These two

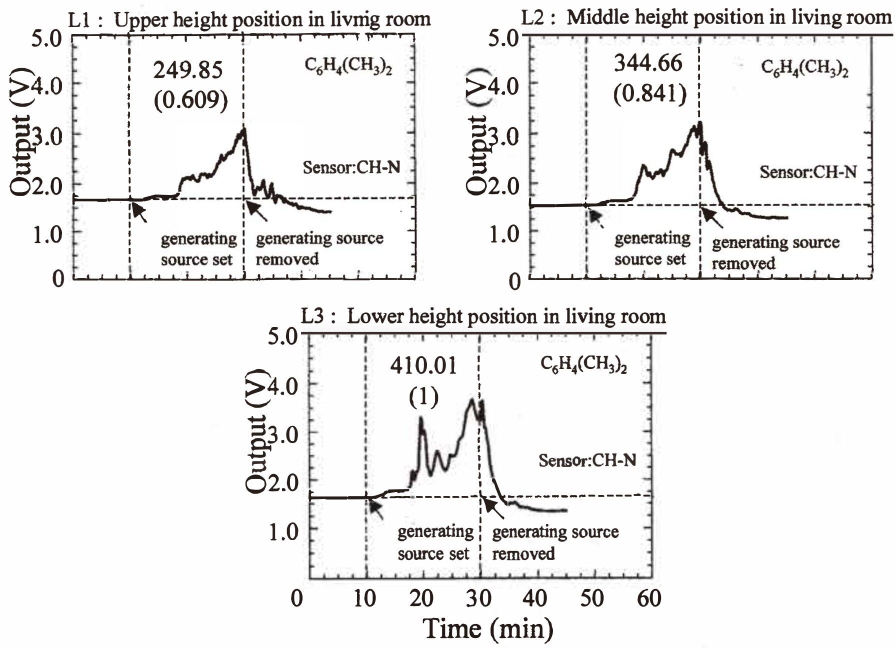

Fig. 5. CH-N sensor outputs for xylene at the positions L1, L2 and L3 on the sensor stand.

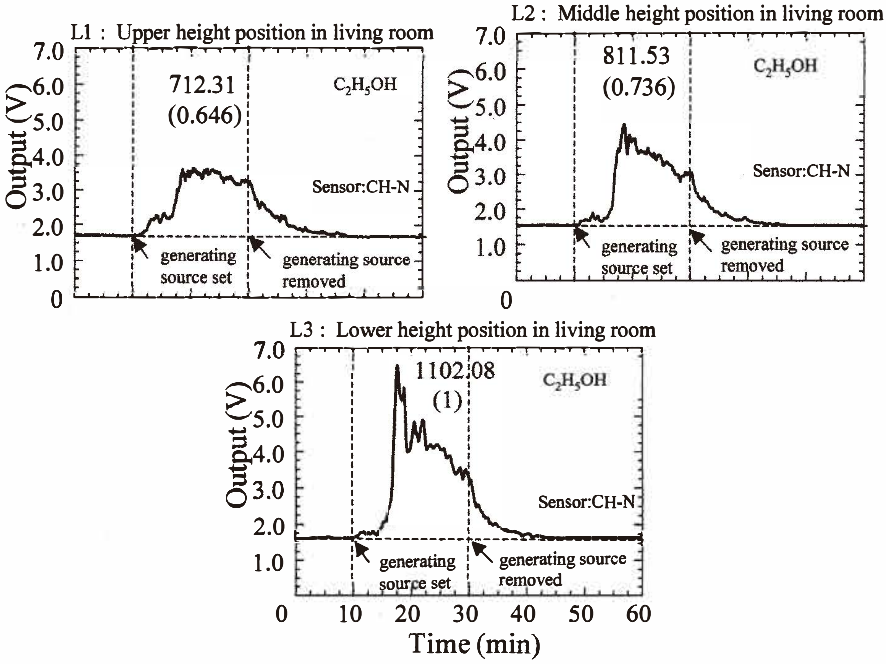

Fig. 6. CH-N sensor outputs for ethyl alcohol at the positions L1, L2 and L3 on the sensor stand.

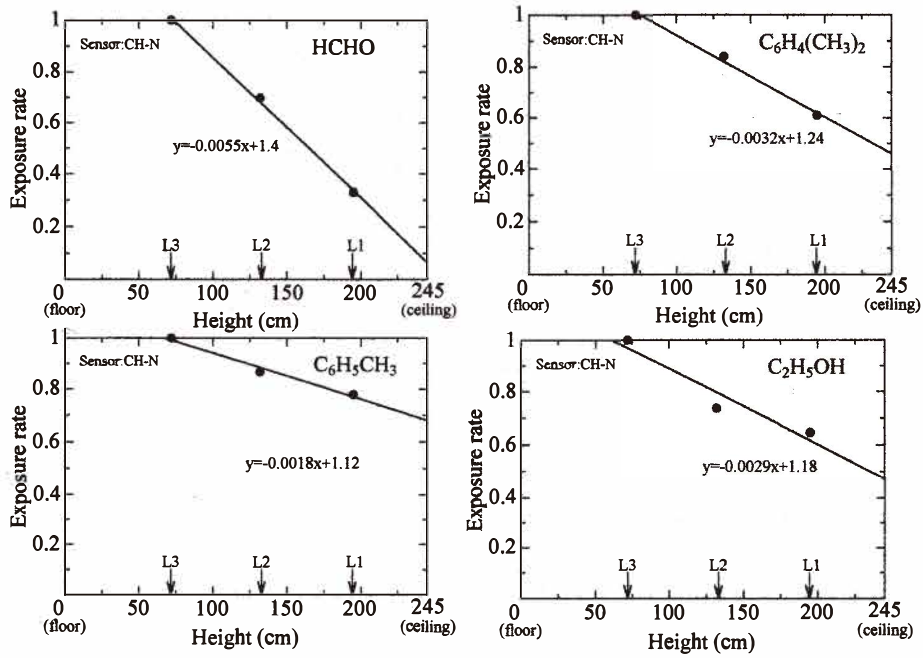

Fig. 7. Approximate exposure volume function of formaldehyde, toluene, xylene and ethyl alcohol as a function of height.

Table 2
Coefficients a and b, and physical chemistry constants for the chemicals examined.

chemical	molecular weight	boiling point (°C)	specific gravity	vapor density	a	b
HCHO	30.03	−19.2	0.815	1.03	−0.0055	1.40
$C_6H_4(CH_3)_2$	106.17	140.0	0.870	3.66	−0.0032	1.24
$C_6H_5CH_3$	92.13	110.8	0.866	3.14	−0.0018	1.12
C_2H_5OH	46.07	79.0	0.789	1.60	−0.0029	1.18

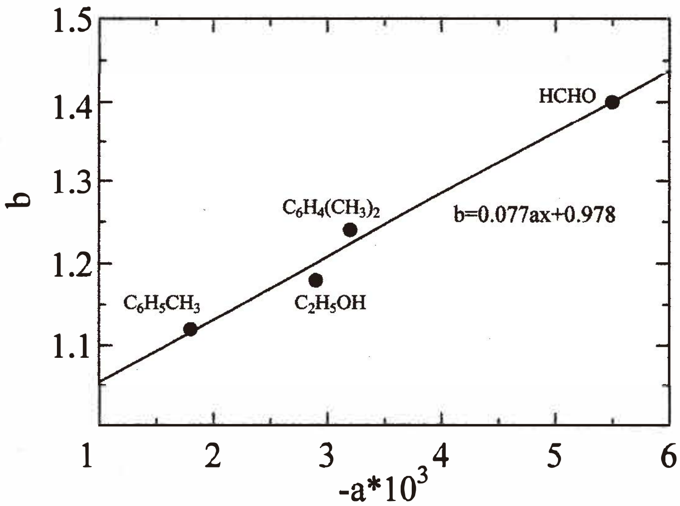

Fig. 8. Relationship of the coefficients *a* and *b*, expressed by a linear function.

coefficients can also be described by a linear function, namely *b* becomes larger as $-a*10^3$ increases. Coefficient *b* is the intercept in Fig. 7. Relationships derived for *a* and *b* can be expressed by the following equation.

$$b = 0.077\,a + 0.978$$

Removal characteristics are also examined using sensor outputs. These are described by the removal time tdec, and the concept is shown in Fig. 9. The term tdec expresses the time when the sensor output value (vremd), upon removal of a pollutant, decreases to 20%, and the time is measured from the moment at which a pollutant is removed and a window is slightly opened. Characteristics of tdec for the pollutants examined as a function of height are shown in Fig. 10(a). The tdec for the pollutants, except for xylene, decreases as

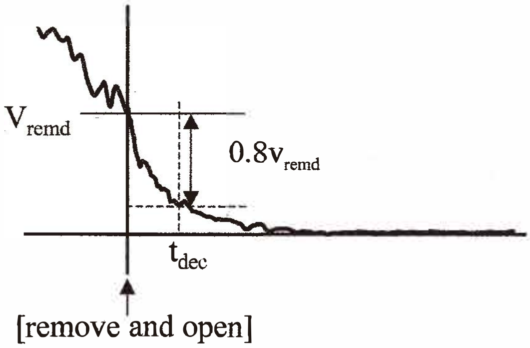

Fig. 9. Concept of removal period t_{dec}.

(a)

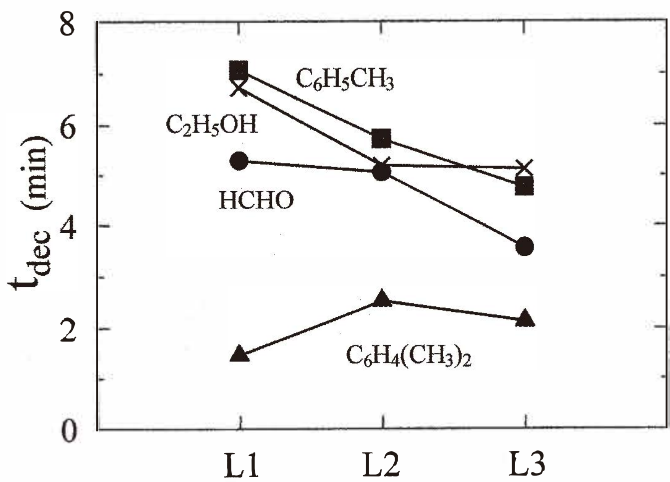

Fig. 10(a). Characteristics of t_{dec}. Parameters are pollutants examined in (a).

the height increases. This is also plotted for each pollutant in Fig.10(b), in which the parameter is height, namely L1, L2 and L3. Toluene and ethyl alcohol have a tendency to remain for a long time in a room. Xylene can easily flow out.

4. Conclusions

The relative concentration distribution of important air pollutants at three different heights is derived using tin oxide odor sensors. It is difficult to estimate the absolute distribution because there are many parameters which influence the gas diffusion equation. In this study, the exposure volumes of four main air pollutants are derived in a real

(b)

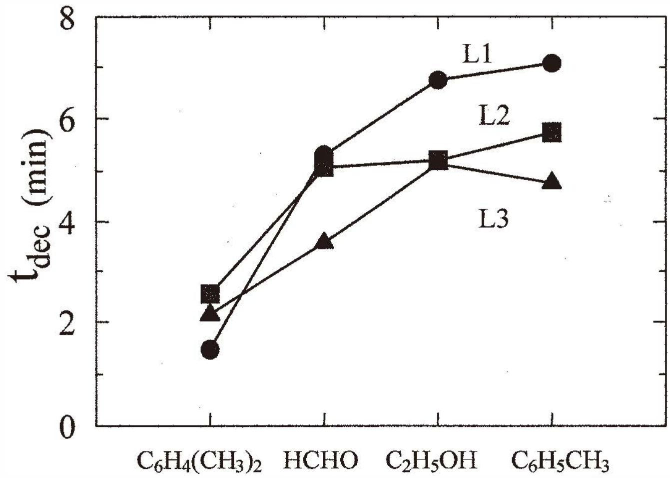

Fig. 10(b). (continued) Characteristics of t_{dec}. Parameters are height in (b).

domicile. The volume increases with decreasing height and is expressed by a linear function. Sick-building syndrome is mainly caused by the following chemicals: formaldehyde, toluene and xylene. It can be estimated from these results that considerable damage is done to bedridden elderly, babies and infants. It is also necessary to consider how to cope with the risk while occupants are asleep. The risk is expressed by the relative grade, which is a normalized value.

A pollutant is put on a table in this study. Pollutants actually arise from surfaces such as boards and curtains. It is necessary that experiments be carried out using these materials and that human surroundings be improved.

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References

1 T. Godish: Air Quality (Lewis Publishers, New York, 1997).
2 T. Salthammer: Organic Indoor Air Pollutants (Wiley-VCH, New York, 1999).
3 G. M. Barrow: Physical Chemistry (McGraw-Hill, 1988).
4 W. J. Moore: Physical Chemistry (Prentice-Hall, Inc., New Jersey, 1972).
5 T. Oyabu, H. Nanto, H. Kasahara, T. Onodera and N. Nakata: Sensors and Materials **11** (1999) 457.

6 A. Sawada, T. Oyabu, H. Nanto, T. Onodera and T. Katsube: Identification of Meal-Degree of a Solitude Aged using Odor Sensors, The Fourth Asian Fuzzy System Symposium, No. 0W2D-2 (May 31-June 3, 2000, Tsukuba Science City, Japan) 68.
7 T. Oyabu, T. Misawa, H. Kimura and H. Nanto: Materials Science & Engineering **C12** (2000) 89.
8 H. Levin: ASTM Standards on Indoor Air Quality, ASTM (West Conshohocken, PA, 1999).
9 T. Oyabu, T. Onodera, H. Nanto and N. Nakata: Purification Effect of Interior Plant for Indoor-Air Polluting Chemicals and Environmental Preservation, 4th International Conference on Engineering Design and Automation (EDA2000), No. 524 (Orlando, Florida, July 30-Aug. 2, 2000).