

DECLINE OF VOC CONCENTRATIONS WITH AGING OF HOUSES IN JAPAN

Motoya Hayashi[†], and Haruki Osawa²

¹ *Department of Living and Cultural Science, Miyagigakuin Women's University, 9-1-1, Sakuragaoka, Sendai, Japan*

² *Department of Environment & Fire Research Building Research Institute, 1, Tachihara, Tukuba, Japan*

ABSTRACT

The purpose of this investigation is to know the long term characteristics of VOC concentrations in houses built before the building code in 2003 and to know the need of the countermeasures in the houses were already built for example the improvement of the living habit, ventilation and building materials. The concentrations of VOCs were measured in these houses from 2000 to 2005. The results showed that the concentration of formaldehyde decreased in the first year. After that the decline of the concentration became invisible and the concentration changed only with the temperature. The characteristic of decline was thought to be caused by the two mechanisms of emission. One is an emission of concealed formaldehyde in the process of material production and another is an emission with the generation of formaldehyde from adhesives of urea resin and moisture. The concentration of toluene decreased rapidly in the first year. The concentrations of xylene, ethyl-benzene and styrene showed a similar change. But the concentrations of acetaldehyde which were measured from the summer of 2002 did not decrease and its concentrations in some houses were higher than the guideline even in the winter of 2005.

KEYWORDS

Indoor air quality, Passive sampling, Questionnaire survey, Statistics analysis

INTRODUCTION

In Japan, many countermeasures against indoor air pollution in houses have been taken by Government, building companies, building material manufacturers and other related groups from 1990s and a new building code with countermeasures against indoor air pollution has been forced by the Japanese government since 2003. According to this building code the concentration of formaldehyde is expected to be lower than the guideline: 0.08ppm, which was established by the Ministry of Health, Labour and Welfare of Japanese Government in 1993. This building code requires our consideration about the emission rate from building materials. The regulated materials are not only building materials which are used for interior finish but also materials which are concealed in walls, ceilings and floors. And the installation of ventilation equipments is required in all residential spaces. The required ventilation time is 0.5 times per hour. Before the building code, the concentrations of formaldehyde were higher than the guideline at the period of completion of houses in many cases. And some investigations showed that the concentrations of formaldehyde do not decrease with aging of houses soon. Volatile organic compounds are volatilizing from building materials and from furniture and articles which are carried into houses by residents. And the indoor concentrations do not decrease with the aging of houses. Therefore the influences of indoor pollution on the resident's health may continue long and these influences may become a cause of a multiple chemical sensitivity. Under these contexts the decline of concentrations with aging of houses was investigated.

[†] Corresponding Author: Tel: + 81 22 277 5793, Fax: + 81 22 277 5793
E-mail address: motoya-h@mgu.ac.jp

METHODS

The concentrations of formaldehyde, toluene, xylene, ethyl-benzene, styrene and acetaldehyde have been measured in winter and in summer during five years in about two hundred and fifty houses in which conditions of formaldehyde concentration was higher than the guideline: 0.08ppm in 2000. The first investigations in 2000 were carried out from summer to winter continuously. The number of investigated houses had been decreased gradually and became eighty four in the winter of 2005 as shown in figure 1. The concentrations were measured using passive samplers made by Advanced Chemical Sensor Inc. The samplers were sent to the residents from a laboratory. The residents placed samplers for 24 hours in the rooms and sent them back to the laboratory. The concentrations were specified using gas chromatographs in the laboratory. The characteristics of buildings and residents were checked using questionnaires. The indoor temperatures and the humidity were measured by residents and were checked on the questionnaires. The number of investigated houses changed. But the results of statistical analysis using the eighty four houses in which the concentrations were obtained at all the periods was quite similar to the results using all data. For example the difference between the average formaldehyde concentration of these eighty four houses and that of all houses is lower than 4%. Therefore the results using all data are reported in this paper.

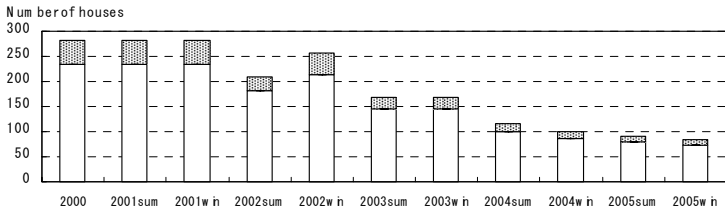


Figure 1 Number of investigated houses

RESULTS

Figure 2 shows Classification of investigated houses. They consist of apartment houses and detached houses. Most apartment houses were built with reinforced concrete structure. And the numbers of story were from one to three in the cases of 45% of the apartment houses. And those were from three to ten in the cases of the other apartment houses. The structure types of detached houses were various but most detached houses were built with wooden post and beam structure which is a common in Japan. One of the other structures was 2inch X4inch wooden stud structure which have been imported and built generally since 1980's. The others were prefabricated structures. Most of the prefabricated houses were produced in large factories by major housing companies. The percentage of classification of houses was similar to the percentage of all houses built recently in Japan.

Figure 3 shows the weather conditions and the humidity when the concentrations were measured in houses. In summer, the percentage of "cloudy" was a little low than that in winter. The temperatures were shown in figure 12. Naturally the temperatures were higher in summer and lower in winter. The humidity changed with the temperature. So it is hot and humid in summer and it is cold in winter. This annual change is typical in most areas of Japan. Figure 4 shows about opening windows. The percentage of opening windows does not contain the percentage in the case that the windows are opened for less than one hour. Naturally the percentage of opening windows is higher in summer than in winter.

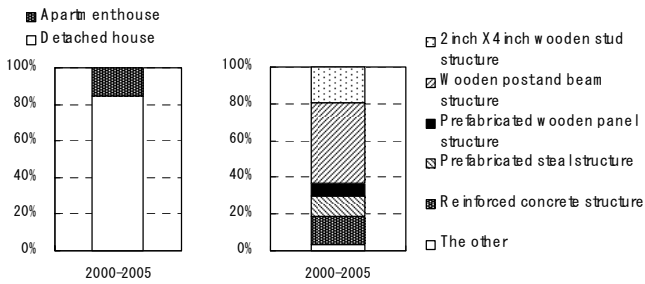


Figure 2 Classification of houses

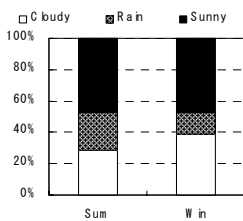


Figure 3 Weather and humidity

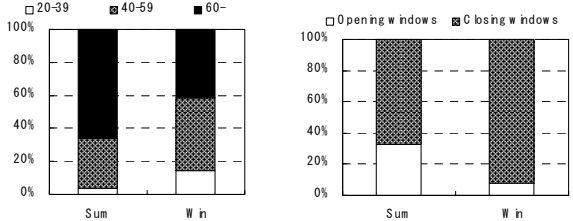


Figure 4 About opening windows

Figure 5 shows the changes of formaldehyde concentrations. In this figure the standard deviations of concentrations on each measurement period are showed using lines. The average of concentration of formaldehyde was higher than the guideline: 0.08ppm in 2000. It is because that the houses were chosen from the houses investigated in 2000 on the condition that the concentration exceeds the guideline. The concentration of formaldehyde decreased from 2000 to the winter of 2001 but the concentration increased in the summer of 2002. After 2003, the concentration increased in summer and decreased in winter. This annual change was repeated during the four years.

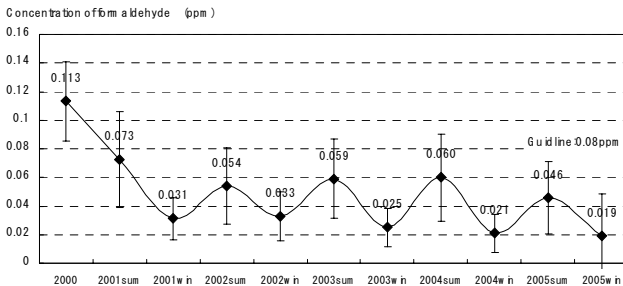


Figure 5 Decline of formaldehyde concentration

Figure 6 shows the concentrations of toluene. The concentration was higher than the guideline: 0.07ppm in 2000. The concentrations decrease and became lower than the guideline in the summer of 2001. The concentrations did not change with temperatures. Figure 7 shows the concentrations of

xylylene. The concentration was very lower than the guideline: 0.20ppm in 2000. The concentrations decrease during the first year and had been low during these five years. In 2003 the concentrations increased a little but the reasons were not clear. Figure 8 shows the concentrations of ethyl-benzene. The concentration was also very lower than the guideline: 0.88ppm even in 2000. The concentrations decrease during the first year and had been low after that. In the winter of 2003 the concentrations increased a little but the reasons were not clear. These temporary rises are thought to be caused by the pollution sources like furniture or insecticide which are carried in houses by residents.

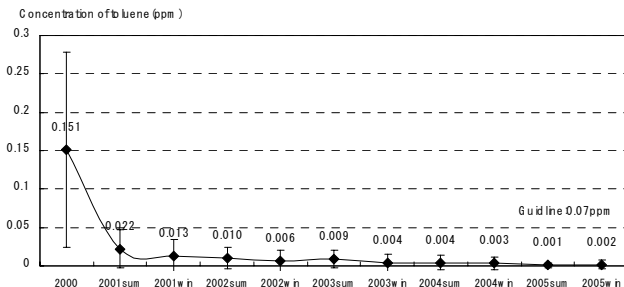


Figure 6 Decline of toluene concentration

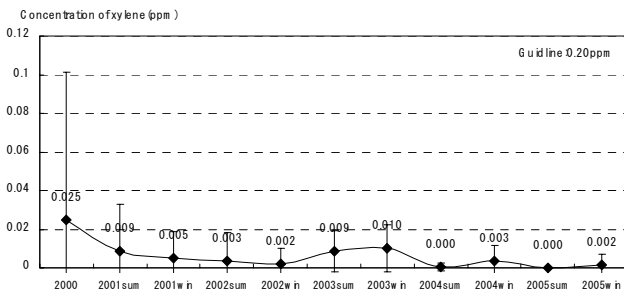


Figure 7 Decline of xylylene concentration

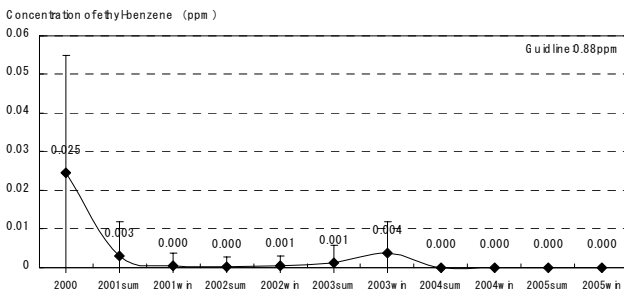


Figure 8 Decline of ethyl-benzene concentration

Figure 9 shows the concentrations of styrene. The average concentration was lower than the guideline: 0.05ppm in 2000. But in some houses the concentrations were higher than the guideline. The

concentrations decrease during the first year and had been lower during these five years. In 2003 the concentrations increased a little.

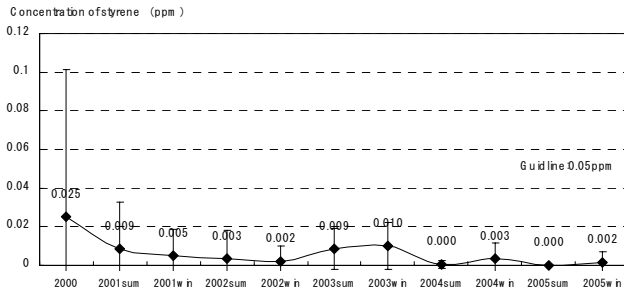


Figure 9 Decline of styrene concentration

Figure 10 shows the concentrations of acetaldehyde. The concentrations were measured after the summer of 2002. The concentration was not enough lower than the guideline: 0.03ppm in the summer of 2003. And the concentrations increased gradually during the four years. The reason of this increasing is not clear. One of the emission sources is smoking but the percentage of "smoking" was almost 11% in all measurement periods. The other source is thought to be building materials. Anyway it became cleared that the concentrations of acetaldehyde exceeded the guideline in some houses.

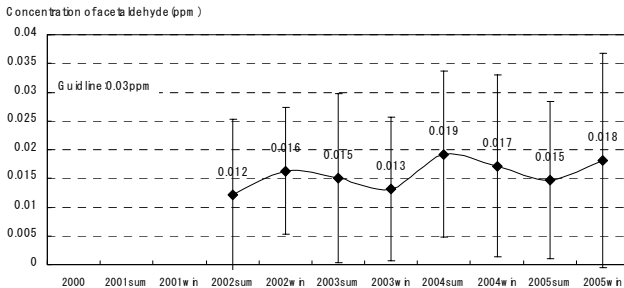


Figure 10 Decline of acetaldehyde concentration

Figure 11 shows the percentage of houses where the residents felt the physical change in houses. The residents answered the following question: Have you felt a change of physical condition which is thought to be caused by the chemical compound. If the answer was "yes", the following matters were checked: who felt the change, the physical conditions, when the change was felt. And the following physical conditions: nausea, headache, eczema, the pain of eyes, nose and throat were appealed on the questionnaires. The percentage of residents who felt the change of physical condition was higher in summer and lower in winter. This change was similar to that of the formaldehyde concentration shown in Figure 12. But the percentage decreased gradually and the fluctuation of the percentage also decreased. These results show that it is necessary to investigate the relationship between the indoor air quality and the feeling about the change of physical condition which are checked on the questionnaires. This investigation is expected to be made with the medical researchers.

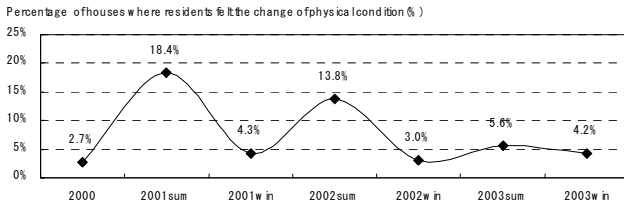


Figure 11 Percentages about change of physical condition with chemical compound

Figure 12 shows the change of temperature and concentrations. The concentrations of formaldehyde changed with the temperature. Figure 13 shows the relationship between temperatures and concentrations. In the case of formaldehyde the concentrations are distributed on a curve: "Calculation ($C_{25}=0.046$, $a=1.11$)" in Figure 13. This curve is calculated using the following equation.

$$C = C_{25} a^{T-25} \dots\dots\dots \text{Equation 1}$$

Where C: concentration, $C_{25} = E_{25} / (Q + \beta)$, E_{25} : emission rate when temperature is 25deg.C, Q: ventilation rate, β : a ratio of sink, a: coefficient of influence of temperature ($a = 1.11$: a value which was measured in small chambers).

The formaldehyde concentration in 2000 was not on the curve but the other concentrations fitted this curve generally. This shows that the ability of formaldehyde emission hardly decreased after 2001.

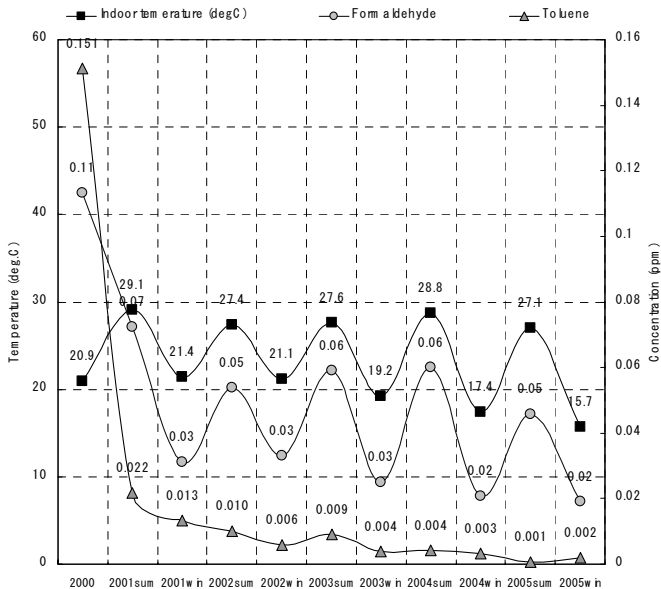


Figure 12 Change of temperature and concentrations

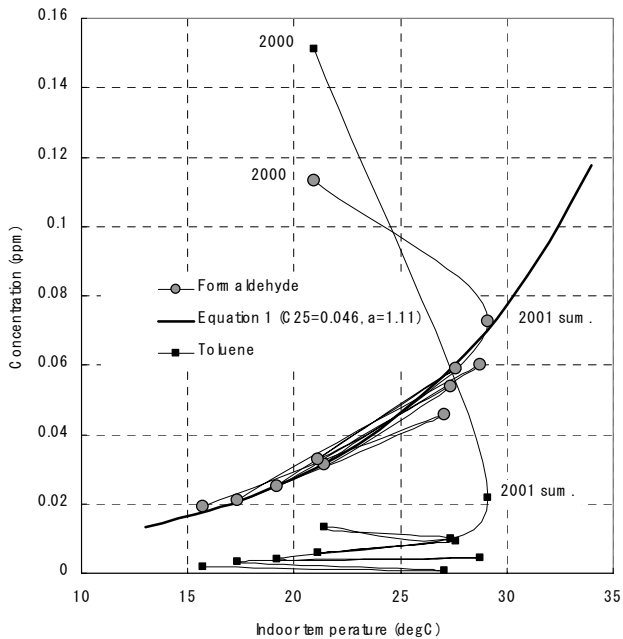


Figure 13 Relationship between temperatures and concentrations

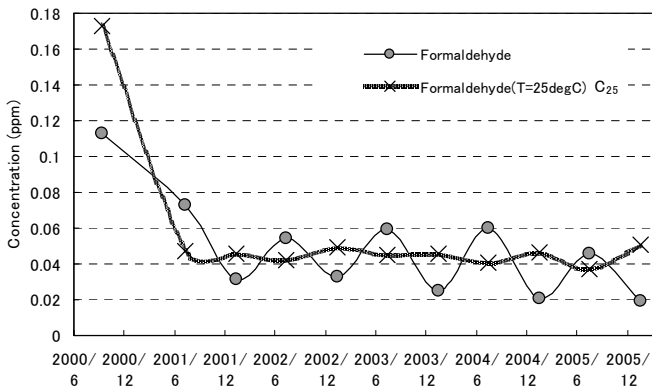


Figure 14 Change of the normalized formaldehyde concentration at a temperature :25deg.C

Figure 14 shows the measured concentrations of formaldehyde and the normalized concentrations: C₂₅ which is calculated using equation 1. The normalized concentration is thought to be an indicator of emission ability in a house. The normalized concentration decreased fast from 2000 to the summer of 2001 and was steady after that. This characteristic of concentration's decline may be caused by a

mechanism of emission from building materials. In Japanese houses main emission sources of formaldehyde were wooden building materials produced in factories like plywood and particle board. In the case of houses built in 2000, most of these materials were made using adhesives of urea resin which is made with formaldehyde. A part of formaldehyde was concealed in the materials when they were produced in factories. Soon after production these concealed formaldehyde may start to volatilize formaldehyde. On the other hand formaldehyde is generated from inside urea resin and moisture after production. The generation rate is influenced by the temperature and the humidity. And the generated formaldehyde diffuses to the surface and volatilizes into the indoor air. The calculated decline curve of normalized concentration shows that the emission of concealed formaldehyde finished in almost one year and the emission with the generation of formaldehyde continues for long time at least five years. The emission will continue until all the sources of formaldehyde are exhausted.

CONCLUSIONS

The investigation on the decline of indoor chemical pollution was carried out in Japanese houses. The concentrations of toluene, xylene, ethyl-benzene, styrene decrease fast and the concentrations were lower than the guidelines in most houses at least after one year. But the concentrations of formaldehyde and acetaldehyde were above the safety zone. In the case of formaldehyde the concentration decreased during the first one year but the concentration did not decrease at all during the next four years. The concentration became higher in summer every year. Therefore the long-term countermeasure against chemical pollution in summer is thought to be necessary. If residents have suffered some impairment by indoor chemical pollutions, renovations of building especially considering indoor air quality will be necessary not only in new houses but also in old houses.

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REFERENCES

1. Motoya Hyashi, Masamichi Enai and Yoshiko Hirokawa, "Annual characteristics of ventilation and indoor air quality in detached houses using a simulation method with Japanese daily schedule model", The International Journal of Building Science and its Applications 'BUILDING AND ENVIRONMENT' Vol.36, No.6 July 2001,pp.721-731
2. Hiroshi Yoshino, Kentaro Amano, Mari Matsumoto, Koji Netsu, Koichi Ikeda, Atsuo Nozaki, Kazuhiko Kakuta, Sachiko Hojo and Satoshi Ishikawa, "Long-Termed Field Survey of Indoor Air Quality and Health Hazards in Sick House", Journal of Asian Architecture and Building Engineering, Vol. 3 (2004) No. 2 pp.297-303