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Distribution of Indoor Concentrations and Emission Sources of Formaldehyde in Japanese Residences

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1. Introduction

Since the 1990s, tight sealing of buildings to save energy and new types of building materials have caused air pollution problems inside many houses in Japan. Many people are suffering from sick building syndrome (SBS), sick house syndrome (SHS), and multiple chemical sensitivity (MCS) in such houses. Formaldehyde has been reported to be one of the chemical substances responsible for causing SBS, SHS, and MCS symptoms, such as eye irritation, respiratory tract irritation, dizziness, fatigue, and neurotoxicity (Kim et al., 2000; Paustenbach et al., 1997; Shinohara et al., 2004). In addition, formaldehyde was reported to be a human carcinogen (IARC 2006).

Formaldehyde has been commonly used in a raw material for synthetic resins such as urea resin, melamine resin, phenolic resin, and synthetic rubber. These resins were used as adhesives in plywood, particle board, and wallpapers in building materials and furniture. The resins react with water to form formaldehyde due to hydrolysis. Formaldehyde has also been used as a bleaching agent and fungicide in wallpaper and curtains. Residual and formed formaldehyde can be emitted from building materials and furniture to the indoor environment of buildings.

Indoor concentrations of formaldehyde are higher in summer than in winter. In Japan, concentration levels were reported to be 39.9 ± 33 μg m^{-3} (Amagai et al., 2000), 34.7 ± 23 μg m^{-3} (Tokyo Metropolitan Government Bureau of Public Health, 2002), and 78.9 ± 22 μg m^{-3} (Shinohara et al., 1999) in summer, while concentrations in winter were 21.7 ± 14 μg m^{-3} (Tokyo Metropolitan Government Bureau of Public Health, 2002), 58.6 ± 20 μg m^{-3} (Shinohara et al., 1999), and 17.6 ± 1.8 μg m^{-3} (Sakai et al., 2004). The formaldehyde concentrations are higher in new houses than those in older houses. The geometric means (GM) of indoor formaldehyde concentrations in new houses were 64.9 μg m^{-3} (Tokyo Metropolitan Institute of Public Health, 2002) and 84.2 μg m^{-3} (Tateno et al., 1999), while those in old houses were 37.7 μg m^{-3} (Tokyo Metropolitan Institute of Public Health, 2002) and 63.6 μg m^{-3} (Tateno et al., 1999). Most of these data, however, were the results of 30-min average or 24-hour average levels.

Following the increase of SHS, an indoor guideline for 30-min average levels of formaldehyde was published by the Japanese Ministry of Health, Labor and Welfare (MHLW) in 1997 in which the level was set at 100 μg m^{-3} (0.08 ppm) based on the level that has been shown to cause nose and throat irritation in humans (MHLW, 2000). This guideline value is the same as that for chronic exposure published by the World Health Organization (WHO, 1999). In addition, plywood and particleboard in Japan is classified into four categories (F one-star to F four-star) depending on their formaldehyde emission rates, which are measured by using an emission chamber or desiccator methods. In the Japanese Building Codes revised in 2003, the areas in which these materials can be used in house construction are limited by their classification and by the ventilation capacity of the house (MLIT, 2003). The guidelines and law have been successful in reducing the mean indoor concentration of formaldehyde in new houses from 89 μg m^{-3} in 2000 to 32 μg m^{-3} in 2004 (MLIT, 2005).

Since the law applies only to new houses, older houses and furniture are not affected. Hence, when indoor formaldehyde levels are high, the major emission sources have to be identified and removed from older houses. Although it is desirable to remove the major emission sources of formaldehyde, it is difficult to determine which sources significantly affect indoor air quality because there are several possible sources, such as flooring, doors, closets, desks, beds, etc. An emission chamber method (IHCP, 1999; ASTM, 1996; 1997; JISC, 2003; Tichenor, 1989) and a desiccator method (JISC, 2001) have been used to measure the emission rates of chemical compounds from building materials. However, it is impossible or impractical to use these methods on-site in actual rooms because the emission sources to be measured must be placed in a desiccator or a chamber. Recently, field and laboratory emission cells (FLEC) (Risholm-Sundman et al., 1999; Uhde et al., 1998; Wolkoff et al., 1991), a passive flux sampler (PFS) (Fujii et al., 2003; Shinohara et al., 2001; Shinohara et al., 2007; Blondel & Plaisance, 2011), a passive emission colorimetric sensor (PECS) (Shinohara et al., 2008), and an advanced diffusive sampling emission cell (ADSEC) (Akutsu et al., 2000; Aoki et al., 2000) have been proposed and applied to measure the emission rates in the field.

In this study, the 24-h average levels of indoor formaldehyde were measured for 7 days over five seasons in 26 houses to obtain the chronic exposure levels of formaldehyde. In addition, the emission sources identification of formaldehyde in several Japanese houses are reported.

2. Method

2.1 Distribution of indoor formaldehyde concentrations

2.1.1 Survey and analysis

Twenty-four-hour average indoor and outdoor formaldehyde concentrations were measured for 7 days in three rooms of 26 Japanese residences over five seasons: the summer and autumn of 2005, and the winter, spring, and summer of 2006. To include a diverse section of residences, volunteers were solicited on the Internet bulletin boards of some corporations and selected according to family composition, residence architecture, and residence age. Thus, each residence had a different family composition, architecture, and age. The living room, bedroom, and one other room such as a child's room or the dining room/kitchen were selected in each residence for this study.

Formaldehyde was passively sampled for 24 hours with a DNPH (2,4-Dinitrophenylhydrazine)-coated silica gel cartridge (DSD-DNPH; Sigma-Aldrich Japan K.K., Japan) at the center of each room at a height of 1.8 m. Carbonyl compounds, extracted with 5 mL of acetonitrile (Wako Pure Chemical Industries, Ltd., Japan) from the sampling cartridge, were analyzed by HPLC (LC-10A, Shimadzu Co.).

The temperature and humidity were measured in each room and outdoors using a thermohygrometer (HL3631, AS ONE Co.). As part of the survey, the residents were asked in a questionnaire to record the durations for which windows were kept open and ventilation fans were run in each room every day. The air exchange rates were also measured using the perfluorocarbon tracer (PFT) method. The results of the air exchange rates were published previously (Shinohara et al., 2011).

2.1.2 Sampling rates, detection limit, recovery rates, and precision

To obtain the sampling rates of formaldehyde for the DSD-DNPH, the cartridges were left in a chamber, in which the concentrations of formaldehyde were controlled with diffusion tubes, for 24 h (N = 3). The lower limit of detection (LOD) and lower limit of quantitation (LOQ) were defined as 3σ and 10σ of the absolute amount of the analyte (N = 5). To evaluate the recovery efficiencies of absorbed formaldehyde from the DSD-DNPH cartridge, 1.0 μg of formaldehyde was spiked to the adsorbent of the DSD-DNPH in a closed vessel (N = 3) and was allowed to stand for 1 h. Then, the compounds were extracted and analyzed as described above. The obtained sampling rate, LOD and LOQ for 24-h sampling, and the recovery efficiency were 46.4 mL min^{-1}, 0.09 μg m^{-3} and 0.27 μg m^{-3}, and 98.3% ± 0.28%.

2.1.3 Statistics

Statistical analysis was conducted using SPSS 15.0 (SPSS Japan Inc., Japan). Two-way repeated analysis of variance (ANOVA) with a Scheffe test was employed for the statistical comparison of the logarithm of formaldehyde concentrations for different seasons and different residences.

The contribution of factor i (inter-residence variability, seasonal variability, and interaction of seasonal and inter-residence variabilities) to the air exchange rates and the contribution of the daily variability can be expressed with the adjusted variance, V_i and V_{daily}.

$$V_i = \left(SS_i - f_i * SS_{daily}\right) / f_i$$

$$V_{daily} = \left(SST - \sum V_i\right) / f_{daily}$$

where SS_T, SS_i, SS_{daily}, f_i, and f_{daily} are the total sum of squares, sum of squares due to factor i, sum of squares due to the daily variation and other errors, the degree of freedom associated with factor i, and the daily variability, respectively.

In addition, multiple linear regression analysis was conducted to determine the contribution of the architecture of the residence, climate, and human behavior, such as the air exchange rates, building age, construction materials, temperature, and humidity. Because the distribution of the indoor formaldehyde concentration had a log-normal distribution, the dependent variable was the log-transformed data on the daily formaldehyde concentrations in each season and the independent variables were the architecture of the residences (wooden or reinforced) (which was expressed by a dummy variable [1, 0]), air exchange rates, temperature, relative humidity, and other factors associated with the building's age (which was expressed by a dummy variable [1, 0, 0] for age < 1 year, [0, 1, 0] for 1-10 years, and [0, 0, 1] for >10 years). A step-down procedure was conducted under $p < 0.05$ of populated F and $p > 0.10$ of eliminated F.

2.2. Indoor emission sources of formaldehyde

The emission rates of formaldehyde were measured in 8 Japanese houses in 2007 as well as the indoor/outdoor concentrations and the air exchange rates. The 30-min average emission rates of formaldehyde from all emission sources were measured with PECS (Fig. 1; Yanagisawa sensor, Nippon Living Co. Ltd., Japan), developed by Shinohara et al. (2008), in 8 Japanese houses. The indoor formaldehyde was sampled for 30 min using a DNPH cartridge (Supelco Lp DNPH S10, Sigma-Aldrich Co., USA) at 200 mL/min with a pump (Pocket pump, SKC Inc., USA), extracted with acetonitrile (Wako Pure Chemical Industries, Ltd., Japan) and analyzed by HPLC (Agilent1100, Agilent Technologies Co., USA). The air exchange rates were measured by the CO_2 concentration-decay method.
The indoor concentrations of formaldehyde were estimated by the following equation, and the estimated value was compared with the measured one.

$$C_{in\_est} = \frac{\sum E_a \times A_a}{N \times V} + C_{out}$$

where C_{in_est} [μg m^{-3}] denotes the estimated indoor formaldehyde concentration; E_a [μg m^{-2} h^{-1}], the emission rate from source a; A_a [m^2], the surface area of source a; N [h^{-1}], the air exchange rate; V [m^3], the room volume; and C_{out} [μg m^{-3}], the outdoor concentration of formaldehyde.

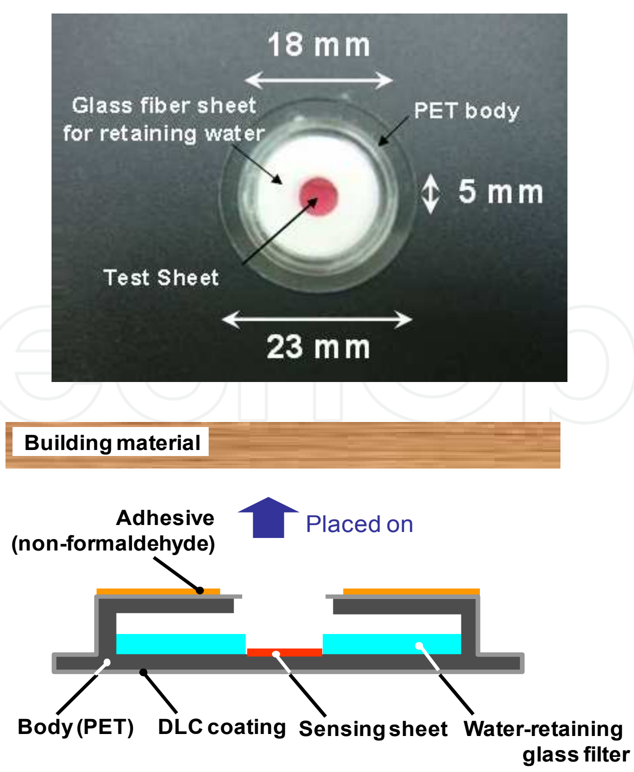

Fig. 1. Picture of PECS (Left) and structure of PECS (Right).

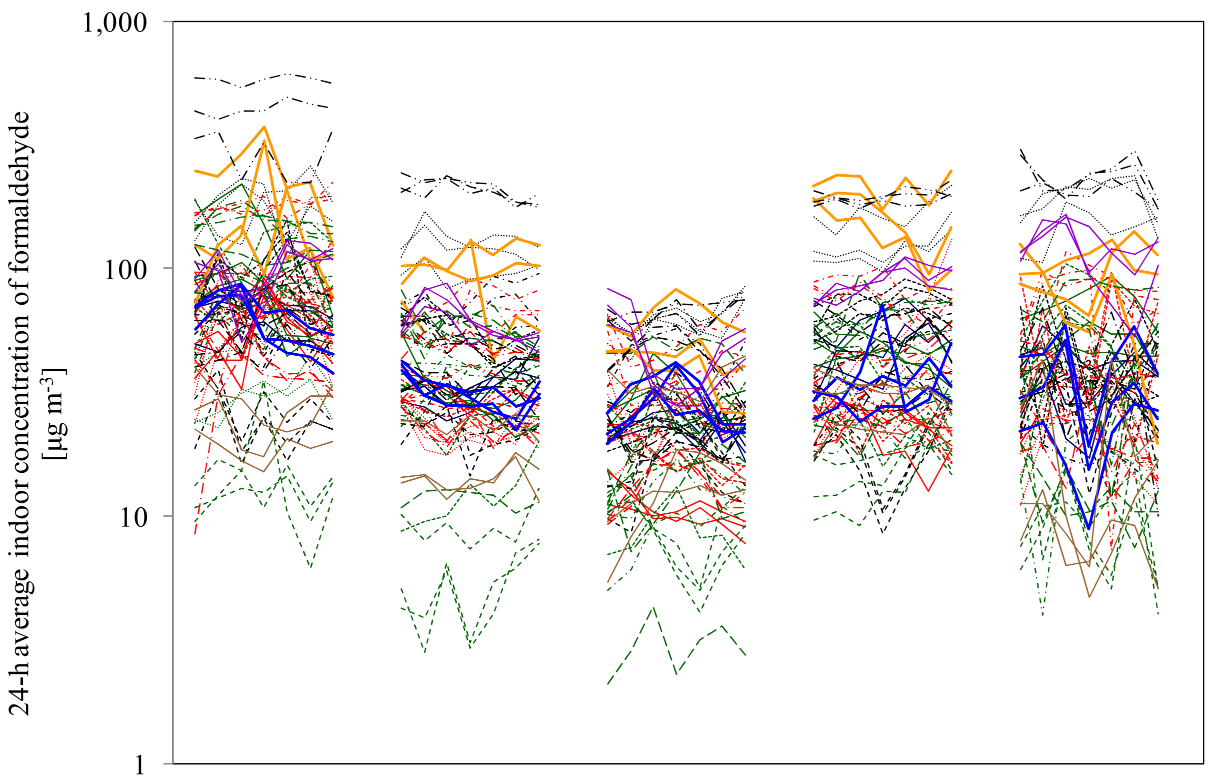

Fig. 2. 24-h average indoor formaldehyde concentrations during the survey. The same line shows data from three rooms of the same residence.

3. Results and discussion

3.1 Distribution of indoor formaldehyde concentrations

3.1.1 Average levels

The geometric mean (GM) of the weekly average indoor formaldehyde levels was 72.8, 38.4, 23.4, 45.4, and 41.1 μg m^{-3} in each of the 5 seasons mentioned in sec. 1.1.1, respectively, and those of the outdoor levels were 6.79, 4.11, 2.49, 4.69, and 3.88 μg m^{-3} (Fig. 2, Table 1). The GM of the yearly average indoor and outdoor formaldehyde concentrations (summer 2005 to spring 2006 and autumn 2005 to summer 2006) was 46.9 and 36.8, and 4.70 and 4.12 μg m^{-3}, respectively.

The percentage of rooms in which the weekly average concentrations were higher than the 30-min average Japanese indoor guideline were 24%, 8.1%, 0%, 12%, and 14% in each season, respectively. The percentage of rooms in which the 24-h average levels were higher than the 30-min average Japanese indoor guideline more than once a week were 43%, 12%, 0%, 19%, and 19% in each season, respectively. Although we did not measure the 30-min average levels of indoor formaldehyde, the 30-min average levels might be higher than the guideline levels in more rooms considering the diurnal variation.

A two-way repeated measures ANOVA revealed that the indoor formaldehyde concentrations differed significantly with respect to seasons, residences, and interaction of seasons and residences ($p < 0.01$). The Scheffe test revealed that the indoor formaldehyde concentrations in both summers and spring were significantly higher than those in winter ($p < 0.05$).

The indoor formaldehyde concentrations in the less than one-year-old residences (yearly average (summer 2005 to spring 2006 and autumn 2005 to summer 2006): 40.7 and 33.6 μg m^{-3}) were lower than those in one-year-old to ten-year-old residences (yearly average: 72.9 and 56.9 μg m^{-3}). The reason for this has been suggested that the revised Building Code in which formaldehyde emission rates and air exchange rates were set in 2003. Those in the more than ten-years-old residences (yearly average: 48.1 and 43.5 μg m^{-3}) were also lower than those in one-year-old to ten-year-old residences. This could be because of the natural decrease of formaldehyde emissions over time.

		Weekly average concentrations					Yearly average concentrations	
		Summer 2005	Autumn 2005	Winter 2006	Spring 2006	Summer 2006	Summer 2005 - Spring 2006	Autumn 2005 - Summer 2006
Indoor	Mean [μg m^{-3}]	94.4	49.4	27.8	58.2	56.7	57.3	46.2
	SD [μg m^{-3}]	88.6	42.3	17.5	48.6	56.0	44.0	37.1
	GM [μg m^{-3}]	72.8	38.4	23.4	45.4	41.1	46.9	36.8
	GSD [-]	2.0	2.0	1.9	2.0	2.2	1.9	1.9
Outdoor	Mean [μg m^{-3}]	7.46	4.68	2.82	4.80	5.05	4.92	4.29
	SD [μg m^{-3}]	3.36	2.38	1.53	1.05	3.70	1.51	1.20
	GM [μg m^{-3}]	6.79	4.11	2.49	4.69	3.88	4.70	4.12
	GSD [-]	1.6	1.7	1.6	1.2	2.3	1.4	1.4

Table 1. Weekly average and yearly average indoor formaldehyde concentrations. (Mean: Arithmetic mean, SD: Standard deviation, GM: Geometric Mean, GSD: Geometric standard deviation)

3.1.2 Variability of the indoor concentrations

The percentage contribution of inter-residence variability, seasonal variability, interaction of seasonal and inter-residence variabilities, and the daily variability to the total variability of the 24-h average air exchange rates in the present survey were 35%, 63%, 1.2%, and 0.12%, respectively. The daily variability was much lower than the other variabilities, inter-residence variability and seasonal variability. Therefore, it is considered that the yearly average levels of formaldehyde could be obtained by a 24-h measurement once every season.

3.1.3 Multiple linear regression analysis

The results of the multiple linear regression analysis (standardized regression coefficients β, and the probability p) are presented in Table 2. The indoor concentration of formaldehyde was strongly associated with temperature. This is because the activity of the hydrolysis reaction increases with temperature and water content, and the vapor pressure increases with temperature. However, the relative humidity was statistically eliminated in this analysis. This might be due to the colinearity. Therefore, the strong association between the temperature and the indoor formaldehyde concentrations can be considered to include the association between the humidity and indoor formaldehyde concentrations. The indoor concentration of formaldehyde was strongly and negatively associated with the air exchange rates. The air exchange can decrease the indoor formaldehyde levels because the outdoor levels were much lower than the indoor levels. In addition, the indoor concentration was negatively associated with wooden buildings and building age (< 1 or > 10 years old). The reasons for this were described above — the Building Code revised in 2003 and the natural decrease of emissions.

	Standardized regression coefficients β	Probability p
Ventilation rate	-0.385	<0.01
Temperature	0.523	<0.01
Wood house	-0.151	<0.01
Building age <1	-0.063	<0.05
Building age >10	-0.130	<0.01

Table 2. Results of multiple linear regression analysis.

3.2 Indoor emission sources of formaldehyde

Fabric furniture and covers, such as curtains, sheets and carpets, were the major source of formaldehyde in many houses. Although formaldehyde is used for permanent-press treatment or bleaching of clothes, the emission/use for clothes is not regulated. On the other hand, the emissions from interior building materials, such as flooring, walls, and ceilings were low. The low emission from the building materials could be because the newly built houses were regulated by the building code.

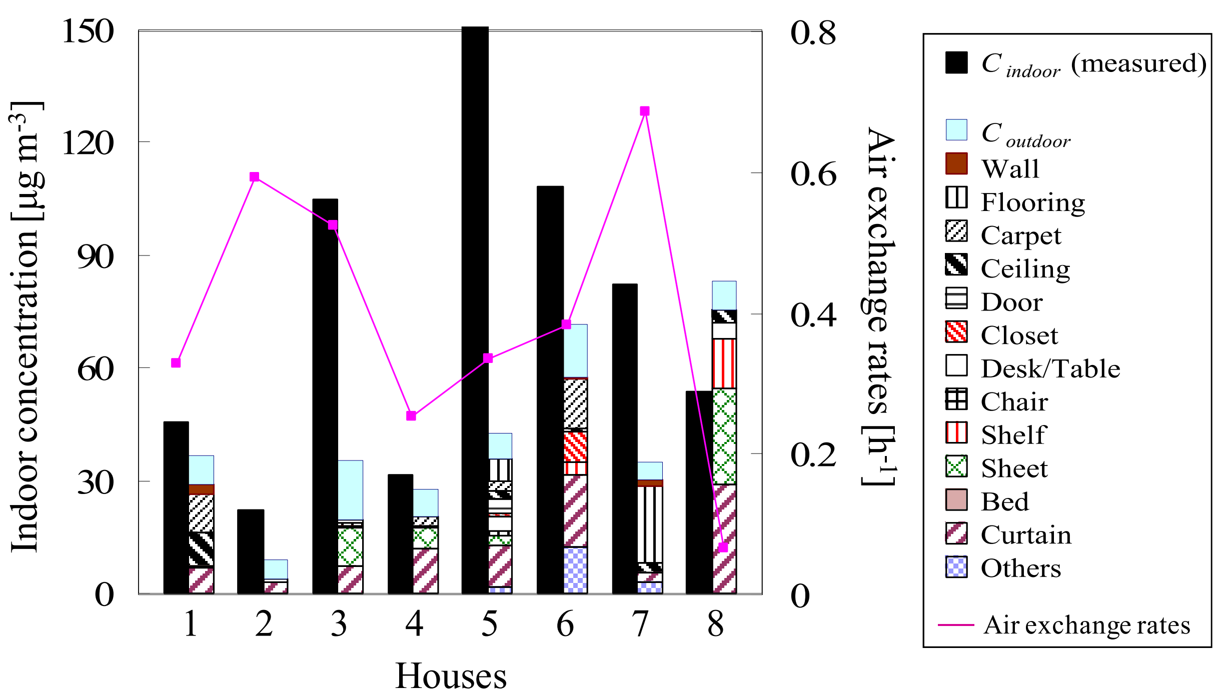

Fig. 3. Measured and calculated indoor concentrations of formaldehyde and the air exchange rates in 8 houses. House 1 and 2 are wooden houses, House 3, 4 and 5 are light-gauge steel houses, while Houses 6, 7 and 8 are reinforced concrete buildings.

In some houses (House 2, 3, 5, and 7), the measured indoor concentrations were significantly different from the calculated concentrations using the emission rates and the air exchange rates. This suggested that there were other emission sources of formaldehyde. Therefore, additional measurements of formaldehyde concentrations were carried out in closets and formaldehyde emission rates were measured around joint gaps on the ceiling and electric outlets on the wall. In the results, the in-closet concentrations were high in House 3 and 7 (> 150 μg m^{-3}). In house 2 and 5, the emission rates were high at the joint gaps. These results suggest that the spaces inside the closet, inside the wall, or under the roof were highly polluted because there are no strict regulations for these materials and spaces, and from there the polluted air could be introduced into the indoor living environment.

4. Conclusions

Formaldehyde has been often used in indoor environments in Japan. The weekly average indoor formaldehyde levels (GM) were 72.8, 38.4, 23.4, 45.4, and 41.1 μg m^{-3} in the summer 2005, autumn 2005, winter 2006, spring 2006, and summer 2006, respectively, and those of the outdoor levels were 6.79, 4.11, 2.49, 4.69, and 3.88 μg m^{-3}, respectively. The indoor formaldehyde concentrations in both summers and in spring were significantly higher than those in winter. The indoor concentration of formaldehyde was strongly associated with temperature, and negatively associated with the air exchange rates, building structure (wooden), and building age (< 1 and > 10 years old). High emission rates of formaldehyde were observed from the cloth furniture and fixtures, such as curtains, sheets and carpets, while low emission rates were observed from building materials. The effect of the intake of formaldehyde from inside closets, inside walls, and under roofs on the indoor formaldehyde levels cannot be ignored.

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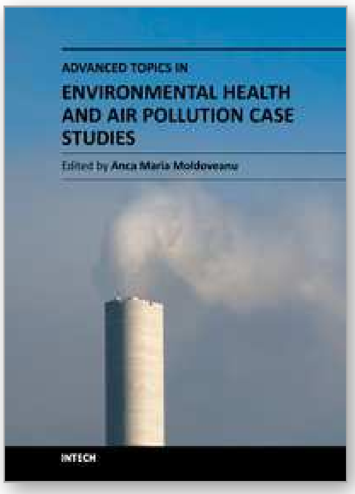

Advanced Topics in Environmental Health and Air Pollution Case Studies

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The book describes the effects of air pollutants, from the indoor and outdoor spaces, on the human physiology. Air pollutants can influence inflammation biomarkers, can influence the pathogenesis of chronic cough, can influence reactive oxygen species (ROS) and can induce autonomic nervous system interactions that modulate cardiac oxidative stress and cardiac electrophysiological changes, can participate in the onset and exacerbation of upper respiratory and cardio-vascular diseases, can lead to the exacerbation of asthma and allergic diseases. The book also presents how the urban environment can influence and modify the impact of various pollutants on human health.