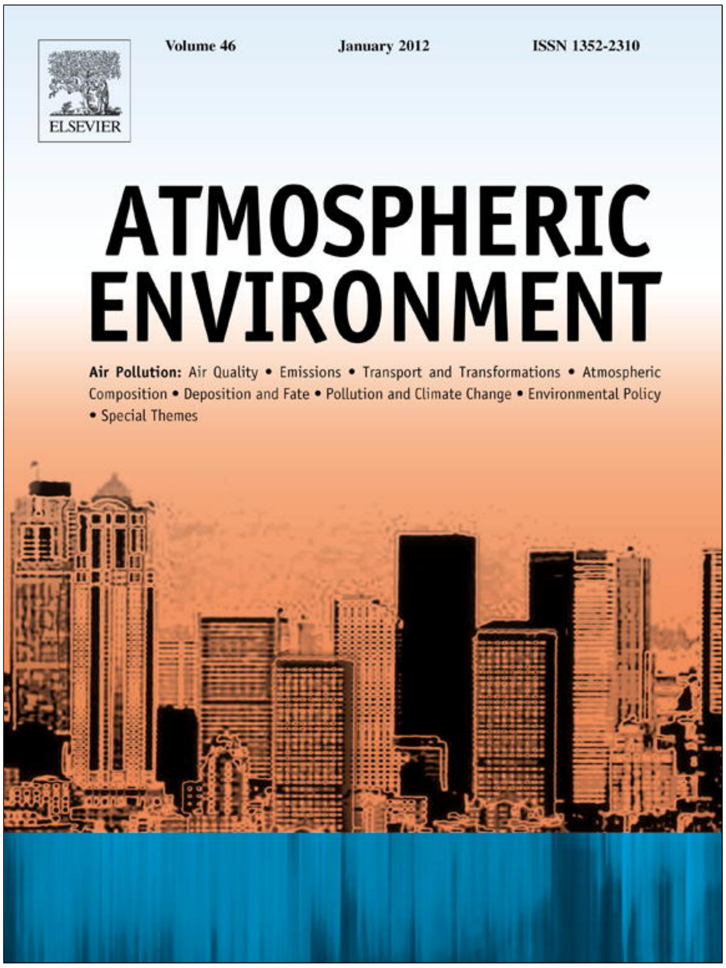
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Indoor aldehydes concentration and emission rate of formaldehyde in libraries and private reading rooms

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HIGHLIGHTS

► Indoor aldehydes levels in libraries are higher than outdoor levels.
► Indoor formaldehyde in private reading room is significantly high.
► Desk is the most significant source of formaldehyde in private reading rooms.
► Desk and building materials should be controlled for formaldehyde.

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ABSTRACT

Aldehydes are of particularly interest due to their potential adverse impact on human health. Formaldehyde is one of the most abundant indoor pollutants. To improve indoor air quality, identifying and removing the major emission sources of formaldehyde would be desirable. The purposes of this study were to determine aldehyde concentrations in libraries and reading rooms and to identify emission sources of formaldehyde in private reading rooms. Indoor aldehyde concentrations were quantified at 66 facilities, including public libraries, children's libraries, public reading rooms, and private reading rooms, in the Seoul metropolitan area. Emission fluxes of formaldehyde from the surfaces of desks, chairs, floors, walls, and ceilings in 19 private reading rooms were measured using a passive emission colorimetric sensor. Indoor aldehyde (formaldehyde, acetaldehyde, propioaldehyde, benzaldehyde, and hexaldehyde) levels were significantly higher than outdoor levels. Indoor formaldehyde geometric mean concentrations in private reading rooms (119.3 $\mu g\ m^{-3}$) were significantly higher than in public libraries (29.2 $\mu g\ m^{-3}$), children's libraries (29.3 $\mu g\ m^{-3}$), and public reading rooms (40.8 $\mu g\ m^{-3}$). Indoor formaldehyde levels were associated with relative humidity. In private reading rooms, the emission rates from desks (255.5 $\pm$ 214.8 $\mu g\ h^{-1}$) and walls (231.7 $\pm$ 192.3 $\mu g\ h^{-1}$) were significantly higher than that from chairs (79.6 $\pm$ 88.5 $\mu g\ h^{-1}$). Desks (31%) and walls (29%) were the major emission sources of formaldehyde in 14 facilities in which measurements exceeded the indoor standard of 100 $\mu g\ m^{-3}$. The age of interior materials was a significant factor for indoor formaldehyde emission flux. Controlling the emission rates of desks and walls is recommended to improve formaldehyde concentrations in private reading rooms.

1. Introduction

Aldehydes are of particularly interest due to their potential adverse impact on human health and their high level in indoor environment. The indoor level of aldehydes are often 1–7 higher than outdoor level (Baez et al., 2003; Zhang et al., 1994). Formaldehyde, the most abundant aldehydes in air, were emitted from

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carpets (Hodgson et al., 1993) and latex paints (Chang et al., 2002), along with various building materials, such as wood products made with urea-formaldehyde (UF) or phenol-formaldehyde resins (Kelly et al., 1999). Cigarette (Li et al., 2002) is also one of the sources of formaldehyde.

Formaldehyde was classified as a group 1 carcinogen for humans by the International Agency for Research on Cancer (IARC, 2006). In addition, exposure to formaldehyde can increase risks of asthma and allergies for children (Garrett et al., 1999; Rumchev et al., 2002). Long-term exposure to formaldehyde has been reported in occupational environments. Observed health effects of formaldehyde include upper respiratory tract irritation, eye irritation, and inflammatory and hyperplastic changes in the nasal mucosa (Edling et al., 1988). Sensory (eye and nasal) irritations in human were reported at 1 ppm of formaldehyde (Arts et al., 2006). Toxicities of other aldehydes are not well reported.

Several studies have monitored indoor levels of aldehyde in residential environments. The Building Research Establishment (BRE) in the United Kingdom carried out an indoor air survey from 1997 to 1999 (Raw et al., 2004). The geometric mean (GM), median, and maximum values from 3-day sampling of formaldehyde in bedrooms ($n = 833$) were 22.2, 24.0, and 171 μg m^{-3}, respectively. Aldehyde levels in 59 homes in Quebec City, Canada, were measured between January and April, 2002 (Gilbert et al., 2005). The indoor formaldehyde and acetaldehyde were ranged from 5.5 to 87.5 μg m^{-3} (GM = 33.2 μg m^{-3}) and from 4.4 to 79.1 (GM = 20.2 μg m^{-3}), respectively.

High levels of aldehyde in libraries were also reported. When aldehyde were measured in various public places (railway station, airport, shopping center, libraries, underground partaking garage, etc.) in Strasbourg area, France, the highest indoor level of formaldehyde were observed in public libraries (Marchand et al., 2006). The indoor formaldehyde levels in library 1 and 2 were 55.9 ± 4.9 μg m^{-3} and 33.7 ± 2.2 μg m^{-3}, respectively. Library 1 had many lines of racks filled with books and old journals, whereas library 2 was more lately built and had more empty places.

Although indoor aldehyde levels and their emission sources were investigated in previous studies, it is difficult to identify which sources are significant in real indoor environment. Methods of an emission chamber (ASTM, 1996; IHCP, 1999) and desiccators (JISC, 2001) can be used to estimate emission rate of formaldehyde. However, it is impossible to apply these methods in actual indoor environments, since emission sources should be placed in a chamber or desiccators. Applying recently developed passive emission colorimetric sensor (PECS) with a portable reflectance photometry device can be used to determine emission rate of formaldehyde in real indoor environments (Shinohara et al., 2008). The PECS allows multipoint field measurement with sampling time of 30 min. The aims of this study were to determine indoor aldehyde concentration in libraries and reading rooms and identify significant emission sources of formaldehyde in private reading rooms.

2. Methods

The Seoul metropolitan area has 188 public libraries and 42 children's libraries. The research team sent letters to 158 libraries requesting that they participate in an investigation of indoor air quality. The 158 libraries were subject to Korean Act of Indoor Air Qualities for Multiple-Use Facilities because of their gross area more than 3000 m^3. Ultimately, 22 libraries, including 11 public and 11 children's libraries, were selected as study sites. Although the study contained more public libraries than children's libraries, the same number of public and children's libraries were selected to compare aldehyde levels. In addition, 23 public reading rooms and 21 private reading rooms were recruited by telephone. Reading rooms are places where students or people can read books and study at their own desk. Field technicians asked for the information on the building age and remodeling. Libraries and reading rooms were chosen without any prior knowledge of the building age, remodeling status, or the period of operation to avoid selection bias.

Indoor and outdoor aldehyde levels were measured from September to December 2010. Aldehyde was sampled with a 2,4-dinitrophenylhydrazine (DNPH) cartridge (Supelco, Bellefonte, PA, USA) and an MP-Σ100H (Shibata, Tokyo, Japan) pump. Sampling was conducted for 30 min with a flow rate of 500 ml min^{-1} for indoor samples. Two sampling locations were selected and two different cartridges were used in each indoor environment. Therefore, four sample cartridges in total were obtained at each indoor environment. The average of the four sample cartridges was used as the indoor aldehyde concentration. Outdoor sampling was conducted for 60 min at a flow rate of 500 ml min^{-1} with one cartridge. An ozone scrubber (Waters, Milford, MA, USA) was connected to the inlet of the cartridges to remove the effects of ozone. In addition, the cartridge was wrapped in aluminum foil to block direct sunlight. All samplers were placed away from direct emission sources, 1 m away from the building wall and 1–1.5 m above the ground or floor.

Aldehydes were extracted from the cartridges using 5 ml acetonitrile. The extracts were analyzed by high-performance liquid chromatography (HPLC) (Series 200; PerkinElmer, Waltham, MA, USA) equipped with a UV–VIS detector (Flexar; PerkinElmer). A derivative solution of 20 μl was injected into a Brownlee choice C_{18} column (150 mm × 4.6 mm, 5 μm) and detected at a wavelength of 360 nm. The hydrazones were separated by a mobile phase of acetonitrile–water (70:30), which was pumped isocratically at a flow rate of 1.5 ml min^{-1}. Commercial hydrazone standard mixing solution (TO11/IP-6A Aldehyde/ketone-DNPH mix; Supelco) was used as a standard solution. For the calibration curves, a standard solution was diluted to create concentrations ranging from 0.15 to 3 μg ml^{-1}. The calibration curves used were linear over the concentration ranges. Of the aldehydes, formaldehyde, acetaldehyde, propioaldehyde, benzaldehyde, and hexanaldehyde were used as target compounds since other aldehydes were mostly below detection limits.

An additional survey of formaldehyde emission flux was conducted. The survey focused on private reading rooms because significantly higher levels of formaldehyde were found in those areas compared to other facilities (public libraries, children's libraries, and public reading rooms). This investigation occurred from February to March 2011. The twenty one private reading rooms were asked to permit additional measurement of formaldehyde emission flux. The two reading rooms with indoor formaldehyde concentrations of 180.4 μg m^{-3} and 83.4 μg m^{-3} refused to participate, thus measurements were conducted in 19 private reading rooms. The emission flux measurements were made in the same indoor environments where the previous aldehyde sampling was completed. Five materials (desks, chairs, floors, walls, and ceilings) were selected as likely emission sources of formaldehyde. Desks mainly consisted of coated medium-density fiberboard (MDF) and bare plywood panels. Chairs were mainly covered with artificial leather. Floors were mainly covered with polyvinyl chloride (PVC) laminated tiles. Walls were mainly covered with PVC coated paper. Ceilings were covered with PVC coated paper, gypsum board, and paint. PECS were used to measure the emission flux of formaldehyde from emission sources. The PECS is about coin sized and based on enzymatic reaction. Design and principle of the PECS had been described in detail elsewhere (Shinohara et al., 2008). A tape was used to measure the dimensions of all materials. While measuring indoor concentrations of aldehydes and emission fluxes from the five materials, temperature and relative humidity (RH) were simultaneously measured using a digital thermometer (303C, Hansung, Seoul, Korea).

The sampling procedure was as follows. First, since the enzymatic reaction requires water, a drop of distilled water was placed in each PECS. Second, the PECS was placed on the target material (i.e., a desk, chair, floor, wall, or ceiling in a reading room) with its face open for 30 min. Finally, each PECS was detached from the materials and analyzed using a portable reflectance photometry device (FB2111, Nippon living Corp., Okayama, Japan) in the field. The average coefficient of variance (CV) of 3 points of PECS for desk ($n = 16$), chair ($n = 14$), floor ($n = 14$), wall ($n = 16$), and ceiling ($n = 14$) were 5.7%, 7.6%, 4.9%, 7.4%, and 4.3%, respectively in the preliminary test.

The emission rate ($\mu g\ h^{-1}$) was estimated by multiplying the surface area of the material (m^2) by the emission flux ($\mu g\ m^{-2}\ h^{-1}$). The total emission rate was calculated by summing the emission rates for the investigated items. The total emission rate was calculated according to Eq. (1).

$$TE_i = \sum_{m=1}^{n} ER_{im} = \sum_{m=1}^{n} E_{im} \times A_{im} \quad (1)$$

where TE_i = Total emission rate ($\mu g\ h^{-1}$) of private reading room i; ER_{im} = Emission rate of material m in private reading room i; E_{im} = Emission flux of the material m ($\mu g\ m^{-2}\ h^{-1}$) in private reading room i; A_{im} = Surface area of the material m (m^2) in private reading room i.

The source contribution of each item was calculated based on the emission rate of the material. Each source contribution was calculated according to Eq. (2).

$$SC_{im} = \frac{ER_{im}}{TE_i} \times 100(\%) \quad (2)$$

where SC_{im} = the source contribution of material m in private reading room i (%).

Normality of the data distribution was verified by a Shapiro–Wilk test. Indoor aldehyde levels were log-transformed for statistical analyses due to their lognormal distribution. Generalized linear model (GLM) and analysis of variance (ANOVA) with Tukey's post-hot test were performed to compare indoor aldehyde levels among facility types and emission rates among the different materials in private reading rooms, respectively. A multiple linear regression model was employed to assess factors that can affect indoor level of aldehydes. The type of facilities (public library, children library, public reading room and private reading room), building age, remodeling, temperature and RH were considered as variables. Since six of 66 facilities did not have information on building age and remodeling, 60 facilities were included in the regression model. The building age of facilities were ranged from 1986 to 2010 years (median = 2004). Twenty seven of the 60 facilities were remodeled. Indoor temperature and RH in facilities were ranged from 13.5 to 29.4 °C (median = 23.8 °C) and from 21 to 71% (median = 41%), respectively. The regression models used stepwise method. A P-value of lower than 0.15 was considered as the significant factor for aldehyde concentrations. A Student's t test was used to compare formaldehyde emission rates of materials. A Pearson's correlation test was used to compare emission rates in private reading room with other variables. Simple linear regression was used to assess the relationship between total emission rates and indoor formaldehyde concentrations. SAS 9.2 (SAS Institute Inc., Cary, NC, USA) was used for all of the statistical analyses. All analysis except multiple linear regression analysis regarded P-value of 0.05 as significant. SigmaPlot 8.0 (Systat Software, Inc., Chicago, IL, USA) and Microsoft Office Excel 2007 (Microsoft Corp., Redmond, WA, USA) were used to draw graphs.

3. Results

Aldehydes were detected in all facilities. GM of indoor formaldehyde, acetaldehyde, propioaldehyde, benzaldehyde, and hexaldehyde concentrations were 51.4 $\mu g\ m^{-3}$ (geometric standard deviation, GSD = 2.6), 10 $\mu g\ m^{-3}$ (GSD = 2.3) $\mu g\ m^{-3}$, 3.9 $\mu g\ m^{-3}$ (GSD = 1.6), 2.9 $\mu g\ m^{-3}$ (GSD = 1.6), and 5.0 $\mu g\ m^{-3}$ (GSD = 2.2), respectively. Outdoor levels were not detected in 3 facilities. GM of outdoor formaldehyde, acetaldehyde, propioaldehyde, benzaldehyde, and hexaldehyde were 1.8 $\mu g\ m^{-3}$ (GSD = 1.9), 2.2 $\mu g\ m^{-3}$ (GSD = 1.7), 1.5 $\mu g\ m^{-3}$ (GSD = 1.4), 2.0 $\mu g\ m^{-3}$ (GSD = 1.2), and 2.8 $\mu g\ m^{-3}$ (GSD = 1.6), respectively. I/O ratio of formaldehyde, acetaldehyde, propioaldehyde, benzaldehyde, and hexaldehyde concentrations were significantly higher than 1. Especially, I/O of formaldehyde were 28.3.

Indoor formaldehyde, acetaldehyde, propioaldehyde, benzaldehyde, and hexaldehyde concentrations by types of libraries and reading rooms are presented in Table 1. Indoor levels of formaldehyde and hexaldehyde were significantly higher in private reading room than other facilities ($p < 0.05$). Indoor levels of benzaldehyde were significantly higher in public library and private reading room than other facilities ($p < 0.05$). However, indoor levels of acetaldehyde and propionaldehyde were not significantly different among facilities.

The final multiple stepwise regression models of aldehydes for facility type, building age, remodeling, temperature, and RH are presented in Table 2. Although formaldehyde had higher R^2 (0.41), other aldehydes had lower R^2. Formaldehyde levels were significantly associated with facility type and RH. Formaldehyde levels were higher in private reading room and with RH higher than 41%. Acetaldehyde levels were significantly associated with facility type and temperature. Acetaldehyde levels were higher in private reading room and with temperature higher than 24 °C. Benzaldehyde levels were higher in children library. Hexaldehyde levels were higher in public reading room and private reading room. Propioaldehyde levels did not have any variables at the 0.15 significance level.

Since indoor formaldehyde levels were significantly higher in private reading room, emission rate of formaldehyde in private reading room were further investigated. The emission rate of formaldehyde from an object was calculated by multiplying the emission flux by the area of each surface (Table 3). Emission fluxes were significantly higher from desks (15.3 ± 9.6 $\mu g\ m^{-2}\ h^{-1}$) than those from floors (6.6 ± 6.8 $\mu g\ m^{-2}\ h^{-1}$), walls (7.7 ± 10.4 $\mu g\ m^{-2}\ h^{-1}$), and ceilings (6.4 ± 5.1 $\mu g\ m^{-2}\ h^{-1}$) in 19 private reading rooms ($p < 0.05$).

Table 1
Aldehyde levels in public library, children library, public reading room and private reading room.

Aldehyde ($\mu g\ m^{-3}$)	PUL[a] ($n = 11$)	CHL[b] ($n = 11$)	PUR[c] ($n = 23$)	PRR[d] ($n = 21$)	P-value*
	GM (GSD)	GM (GSD)	GM (GSD)	GM (GSD)	
Formaldehyde	29.2 (2.2)	29.3 (3.1)	40.8 (2.1)	119.3 (1.7)*	<0.05
Acetaldehyde	8.9 (2.2)	7.2 (2.1)	9.1 (2.2)	14.2 (2.5)	0.12
Propionaldehyde	4.4 (1.7)	3.3 (1.5)	3.7 (1.4)	4.4 (1.6)	0.25
Benzaldehyde	3.9 (1.6)*	2.4 (1.5)	2.3 (1.5)	3.5 (1.4)*	<0.05
Hexaldehyde	4.9 (2.3)	4.9 (2.6)	3.1 (1.7)	8.4 (1.9)*	<0.05

* $P < 0.05$, statistically significant for GLM analysis.
[a] Public library.
[b] Children's library.
[c] Public reading room.
[d] Private reading room.

Table 2
Multiple stepwise regression model of ln-transformed aldehyde concentration and facility characteristics ($n = 60$).

Substance	Variable	Coefficient	Standard error	Significance
Formaldehyde ($R^2 = 0.41$)	Type of facility (reference: Public library)			
	Private reading room	1.42	0.22	<0.001
	RH ≥ 41%	0.52	0.21	0.02
Acetaldehyde ($R^2 = 0.15$)	Type of facility (reference: Public library)			
	Children library	−0.48	0.31	0.13
	Private reading room	0.51	0.24	0.04
	Temperature ≥ 24 °C	0.55	0.23	0.02
Benzaldehyde ($R^2 = 0.25$)	Type of facility (reference: Public library)			
	Children library	−0.38	0.15	0.01
	Public reading room	−0.26	0.16	0.10
	Remodeling (Yes vs. No)	−0.24	0.15	0.12
Hexaldehyde ($R^2 = 0.28$)	Type of facility (reference: Public library)			
	Public reading room	−0.42	0.20	0.04
	Private reading room	0.55	0.21	0.01

No significant difference in emission flux was seen between desks and chairs (9.4 ± 6.1 μg m^{-2} h^{-1}). The walls had the largest surface areas (35.8 ± 12.0 m^2) among the five types of materials ($p < 0.05$). The surface areas of desks (16.9 ± 11.7 m^2), floors (19.1 ± 10.3 m^2), and ceilings (19.1 ± 10.3 m^2) were all similar. Chairs had the smallest surface areas (7.7 ± 5.0 m^2).

In the 14 private reading rooms that exceeded the maintaining concentration (100 μg m^{-3}) of indoor formaldehyde standard established by the Korean Act of Indoor Air Qualities for Multiple-Use Facilities, desks had the highest emission fluxes, with a mean of 15.6 ± 9.1 μg m^{-2} h^{-1}. Emission fluxes from chairs, floors, walls, and ceilings were 11.2 ± 6.1 μg m^{-2} h^{-1}, 8.7 ± 6.7 μg m^{-2} h^{-1}, 9.5 ± 11.5 μg m^{-2} h^{-1}, and 7.5 ± 5.3 μg m^{-2} h^{-1}, respectively. However, emission fluxes from these 5 materials were not significantly different ($p = 0.09$). In the five private reading rooms that complied with the standard, emission fluxes from desks (14.4 ± 12.1 μg m^{-2} h^{-1}) were significantly higher than those from walls (2.4 ± 2.9 μg m^{-2} h^{-1}) and floors (0.7 μg m^{-2} h^{-1}) ($p < 0.05$). Emission fluxes from chairs (4.6 ± 3.3 μg m^{-2} h^{-1}) and ceilings (3.2 ± 3.0 μg m^{-2} h^{-1}) were not significantly different. In the facilities that exceeded the standard, emission fluxes from chairs, floors, and walls were significantly higher than in the facilities that complied with the standard ($p < 0.05$).

Emission flux was associated with facility age and certain materials. Negative correlations were observed between emission fluxes from floors, walls, and ceilings and the corresponding age of the material ($p < 0.05$). However, no significant correlations were found for desks ($p = 0.09$) and chairs ($p = 0.12$). The emission flux of PVC wallpaper (6.6 ± 4.2 μg m^{-2} h^{-1}) was significantly higher compared to painted surfaces (1.7 ± 2.2 μg m^{-2} h^{-1}) ($p < 0.05$). Fourteen private reading rooms used PVC wallpaper and four private reading rooms had painted wall surfaces. One private reading room used coated MDF and plywood panels as wall covering materials and the emission flux in the room was 46.8 μg m^{-2} h^{-1}.

Table 3
The average emission flux, surface area, and emission rate of 5 materials in 19 private reading rooms.

Material	Emission flux (μg m^{-2} h^{-1})	Surface area (m^2)	Emission rate (μg h^{-1})
Desk	15.3 ± 9.6	16.9 ± 11.7	255.5 ± 214.8
Chair	9.4 ± 6.1	7.7 ± 5.0	79.6 ± 88.5
Floor	6.6 ± 6.8	19.1 ± 10.3	125.8 ± 142.1
Wall	7.7 ± 10.4	35.8 ± 12.0	231.7 ± 192.3
Ceiling	6.4 ± 5.1	19.1 ± 10.3	116.2 ± 105.8

The emission rates of desks (255.5 ± 214.8 μg h^{-1}) and walls (231.7 ± 192.3 μg h^{-1}) were significantly higher than those of chairs (79.6 ± 88.5 μg h^{-1}) ($p < 0.05$). The average emission rates of floors and ceilings were 125.8 ± 142.1 μg h^{-1} and 116.2 ± 105.8 μg h^{-1}, respectively. The emission rates of desks (288.7 ± 217.6 μg h^{-1}) and walls (286.9 ± 191.3 μg h^{-1}) were significantly higher than those of chairs (96.2 ± 96.6 μg h^{-1}) in the 14 private reading rooms that exceeded the standard ($p < 0.05$). Emission rates from floors and ceilings in the 14 noncompliant facilities were 166.3 ± 145.6 μg h^{-1} and 142.3 ± 110.6 μg h^{-1}, respectively. In the five private reading rooms that complied with the standard, the emission rates of desks, chairs, floors, and ceilings were 162.5 ± 197.6 μg h^{-1}, 33.1 ± 36.0 μg h^{-1}, 12.3 ± 10.5 μg h^{-1}, 77.1 ± 83.5 μg h^{-1}, and 43.1 ± 40.0 μg h^{-1}, respectively. Fig. 1 shows the differences in emission rates between reading rooms classified by compliance. The emission rates of floors, walls, and ceilings were significantly higher in the 14 reading rooms that exceeded the standard ($p < 0.05$). Even though the result was not significant, the emission rates of desks ($p = 0.27$) and chairs ($p = 0.05$) were higher in the 14 private reading rooms that exceeded the standard. The total emission rate was calculated by summing the emission rates of the five materials. The total emission rates and formaldehyde concentrations in the 19 private reading rooms were correlated ($R^2 = 0.35$; Fig. 2).

For the emission rates in private reading rooms, the source contribution was calculated using Eq. (2). The highest source contribution in the 19 private reading rooms was observed with desks (34%). The source contributions of walls, ceilings, floors, and chairs were 27%, 15%, 14%, and 10%, respectively. In the 14 private reading rooms that exceeded the standard, desks had the highest source contribution (31%) among the five surfaces examined. The source contributions of walls, ceilings, floors, and chairs were 29%, 14%, 16%, and 10%, respectively. The source contributions of desks, walls, ceilings, floors, and chairs were 43%, 22%, 19%, 6%, and 10%, respectively, in the five private reading rooms that complied with the standard.

4. Discussion

Aldehydes levels in libraries and reading rooms were comparable to previous studies. Aldehyde levels at 20 university libraries in France ranged from 8.6 to 94.5 μg m^{-3} for formaldehyde, 3.7–25.9 μg m^{-3} for acetaldehyde, 0.7–16.3 μg m^{-3} for propioaldehyde, 0.2–5.3 μg m^{-3} for benzaldehyde, and 2.1–58.8 μg m^{-3} for hexaldehyde (Allou et al., 2008). Formaldehyde concentrations in

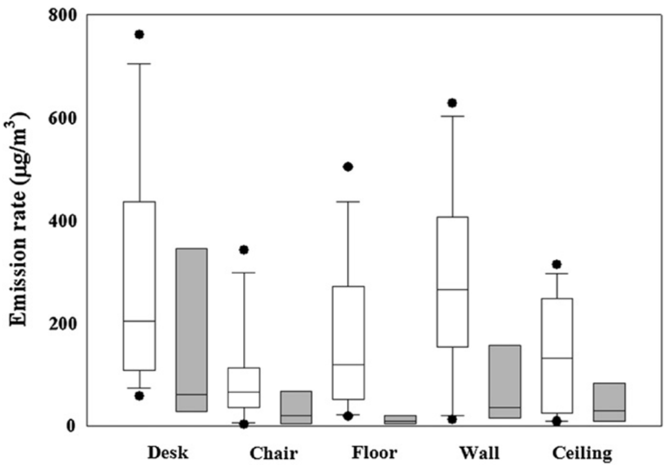

Fig. 1. Comparison of emission rates in private reading rooms that exceeded (white, $n = 14$) and complied with (gray, $n = 5$) the indoor standard.

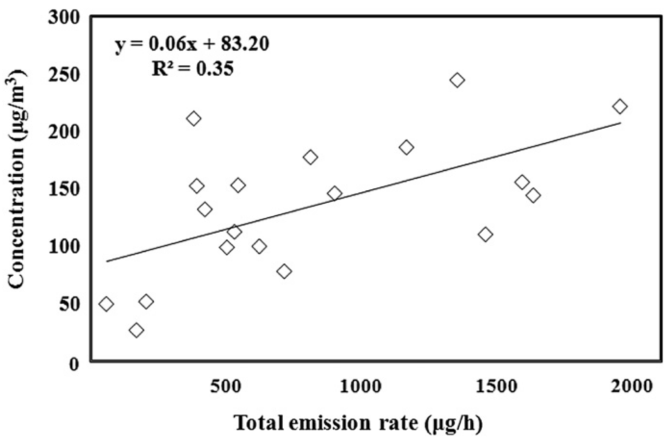

Fig. 2. Correlation between formaldehyde concentration and total emission rate in 19 private reading rooms.

16 libraries of the university of Modena in Italy ranged from 1.7 to 67.8 μg m^{-3} with an average value of 32.7 ± 23.9 μg m^{-3} (Fantuzzi et al., 1996).

Indoor aldehydes levels were significantly higher than outdoor level. The I/O ratios of aldehydes ranged from approximately 1–28. The formaldehyde had the highest I/O ratios. This indicated that there were strong emission sources of aldehydes in libraries and reading rooms. High levels of aldehydes were reported in Mexico City and Xalapa, Mexico (Baez et al., 2003). Indoor levels (houses, museums, and offices) of formaldehyde, acetaldehyde, and propioaldehyde were commonly 1–5 times higher than outdoor level. The mean and maximum I/O ratio of formaldehyde in residential house were 7.2 and 30.1, respectively (Zhang et al., 1994). Since people spend more than 80% of their time indoors (Yang et al., 2011), exposure in indoor areas contributes significantly more to total personal exposure than outdoor exposure. Young people can be exposed to high aldehydes levels in libraries since they tend to spend longer periods of time in these locations.

In addition to facility type, indoor aldehyde levels were associated with various factors including RH for formaldehyde, temperature for acetaldehyde. Indoor benzaldehyde and hexaldehyde levels were associated only with facility type, while propioaldehyde levels were not associated with any variable. Age of wall or floor coverings, smoking, carbon dioxide level and temperature had been reported their associations with indoor aldehyde levels (formaldehyde, acetaldehyde, pentanaldehyde, and hexaldehyde) (Clarisse et al., 2003). Other study indicated that formaldehyde levels depend on temperature and RH (Haghighat and Bellis, 1998).

Indoor levels of formaldehyde in private reading rooms were significantly higher than in other types of facilities. In private reading rooms, 15 of 21 facilities (71.4%) exceeded the indoor formaldehyde standard of 100 μg m^{-3}. This result was similar to what was found in a previous study conducted in Seoul and Gyeonggi Province from February to April 2008 (Park et al., 2008). In that study, indoor formaldehyde levels were measured in 15 private reading rooms. The arithmetic mean concentration was 136.7 ± 62.0 μg m^{-3} and 13 of 15 (86.7%) private reading rooms exceeded the standard. These high indoor formaldehyde levels may be due to the small spaces and low air-exchange rates (ACH) in private reading rooms. Although this study did not measure ACH in libraries and reading rooms, private reading rooms tended to have an airtight building structure with closed windows for the purpose of noise control. In public reading room in Seoul, Korea, ACH was about 0.5–0.6 h^{-1} (Hong et al., 2012).

Among the five items investigated in private reading rooms, desks had the highest formaldehyde emission flux. Desks in private reading rooms consisted of coated MDF and bare plywood panels. Even though the surface area of coated MDF was larger than that of bare plywood panels, the emission flux from coated MDF was negligible. Because they block formaldehyde release, surfaces of coated MDF showed no formaldehyde emissions in our preliminary tests. However, a strong formaldehyde emission flux was observed from the surface of bare plywood panels. Laminated coated materials have lower emissions of formaldehyde than bare plywood (Kelly et al., 1999). Bare plywood was found to be the main source of formaldehyde emissions (Kelly et al., 1999; Martinez and Belanche, 2000). Minimizing the use of bare plywood panels might achieve a reduction in emission rates from desks.

The age of interior materials was closely correlated with formaldehyde emission flux. Negative correlations were observed between material age and emission fluxes from floors, walls, and ceilings. When materials were older, emission flux decreased. A previous study determined that formaldehyde emission levels decreased linearly over time even though variation existed among the materials tested (Zinn et al., 1990). However, we found no significant correlations between emission fluxes from desks and chairs and the age of the item. Although *P*-values for desks and chairs were 0.09 and 0.12, respectively, a trend was observed for emission flux to decrease with the age of the material. Unlike flooring, walls, and ceilings, desks and chairs were produced by many manufacturers and their emission fluxes varied widely. Therefore, our sample size may have been insufficient for statistical analyses of correlations between age and emission fluxes from desks and chairs.

The emission flux of PVC wallpaper was significantly higher than that of painted wall surfaces. From this result, one can assume that the emission flux of PVC wallpaper, as a wall covering material, was a major source of formaldehyde emissions. Fourteen of 19 private reading rooms (74%) used PVC wallpaper as a wall covering material. PVC wallpaper and wall covering adhesives were known to be formaldehyde emission sources (Funaki and Tanabe, 2002). Therefore, the recommendation was made that private reading rooms should use formaldehyde-free wall materials and adhesives to reduce indoor formaldehyde levels.

When total emission rates were estimated by summing across the five materials, indoor formaldehyde levels and total emission rates were correlated. Although paper products may be one of source for formaldehyde (Marchand et al., 2006), their contribution to indoor level may be negligible. This is because the paper products are commonly carried by users of private reading rooms and have small surface area. Assuming similar ventilation rates among the private reading rooms, this correlation indicated that the five materials may be major emission sources. This result was similar to the findings of a previous study (Shinohara et al., 2009). The floors, walls, and ceilings and wood materials such as desks and closets were selected as emission sources of formaldehyde. The estimated concentration from the selected items was closely correlated with the actual indoor concentration of formaldehyde.

This study had a few limitations. First, indoor formaldehyde levels and emission fluxes were measured in different seasons. Formaldehyde levels were measured in the fall and emission fluxes were measured in spring. Indoor formaldehyde levels could have been affected by temperature and humidity in addition to the number of emission sources and the ventilation rate. Formaldehyde levels were higher when temperature and RH were higher (Haghighat and Bellis, 1998). However, the effects of temperature and RH differences between sampling periods may have been minimal. Temperature and RH differences between the two monitoring periods were 2.6 °C and 12.9%, respectively. Small temperature (±2 °C) and humidity changes (~10%) had negligible effects on formaldehyde emissions (Myers, 1985). Second, other formaldehyde sources in private reading rooms, such as books and textiles, were not measured because of their small surface areas and the impracticality of measuring formaldehyde emission flux with the PECS. While the exclusion of certain materials may have led to an underestimate of the total emission rate, their impact may have been negligible. Third, a single PECS was used to measure the emission flux from a single item. Single point measurements can be affected by variation in the emission rate. However, average CV of 3 points for desk, chair, floor, wall, and ceiling were low in the preliminary test. Fourth, the PECS was placed over cracks in laminate flooring. Since emissions from laminate surfaces were expected to be lower, the emission flux of the flooring could have been overestimated.

A total of 66 libraries and reading rooms were investigated indoor and outdoor levels of formaldehyde, acetaldehyde, propioaldehyde, benzaldehyde, and hexaldehyde. Most aldehydes had higher indoor levels than outdoor levels. Indoor aldehydes levels were associated with several factors (facility type, temperature, RH). Among several types of libraries and reading room, private reading room has significantly high formaldehyde levels. Based on measurements of formaldehyde emission rates, desks were identified as the most significant sources of formaldehyde in private reading rooms. Desks should be replaced to control or use lower emission of formaldehyde material. In addition, indoor building materials posed potential problems and contributed to high formaldehyde concentrations in private reading rooms. Emission rates from floors, walls, and ceiling in private reading rooms that exceeded the formaldehyde standard were significantly higher than in rooms that complied with the standard. Therefore, we recommend that emission rates from desk and building materials should be controlled in private reading rooms.

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