

## Residential Exposure and Risk Levels to Ambient Formaldehyde and Acetaldehyde According to Distance from Industrial Area in Metropolitan City

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### Abstract

The present study evaluated residential exposure to atmospheric formaldehyde and acetaldehyde according to distance from the a dyeing industry complex (DIC). This purpose was achieved by measuring concurrently the outdoor air concentrations in residences near the DIC and a certain distance away, plus the outdoor air concentrations at two industrial areas within the DIC boundary. Formaldehyde concentrations (median values of 24.3 and 22.5  $\mu\text{g}/\text{m}^3$  in IS1 and IS2, respectively) were higher than acetaldehyde concentrations (median values of 7.4 and 7.3  $\mu\text{g}/\text{m}^3$  in IS1 and IS2, respectively) at both sites. However, there was no significant difference in the industrial outdoor air concentrations of both formaldehyde and acetaldehyde between the two sites. In addition, the median formaldehyde concentration from the residential site near the DIC (RS1) was about 1.5 times higher than that from the residential site far away from the DIC(RS2), and the median acetaldehyde concentration from RS1 was about 1.3 times higher than that from RS2. It is noteworthy that the mean or median risk as well as these maximum risks are well above the USEPA's permissible risk level of  $10^{-6}$  from environmental exposure. This suggests that appropriate management for formaldehyde and acetaldehyde is necessary in order to decrease risk of the residents of study areas, regardless of the distance from the DIC.

**Key Words :** Dyeing industry, Statistical analysis, Industrial site, Permissible risk level, Environmental exposure

### 1. Introduction

Aldehydes are environmental carbonyl compounds, which are ubiquitously present in atmospheric air of urban areas, of interest because of their adverse health effects. Residential outdoor concentrations of several aldehydes in urban areas have been measured in several previous studies (Báez et al., 2003; Sax et al., 2004; Liu et al., 2006). Furthermore, formaldehyde and acetaldehyde are well known as two most

prevalent carbonyl compounds in urban atmospheric air (Liu et al., 2006). Formaldehyde has been considered as carcinogenic to humans on the basis of sufficient evidence from several epidemiological surveys (Hauptmann et al., 2004; Pinkerton et al., 2004; Coglianò et al., 2005), and has been classified in Group I by the International Agency for Research on Cancer (IARC, 2004). Acetaldehyde, another abundant carbonyl hydrocarbon in urban air, has been known as probable human carcinogen, and has been classified in Group B by USEPA (USEPA, 2003). These characteristics of airborne formaldehyde and acetaldehyde warrant the survey of exposure levels to these carbonyl compounds in various residential areas.

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Most previous studies on urban aldehyde levels were conducted around typical residences or roadway-adjacent residences (Báez et al., 2003; Sax et al., 2004; Liu et al., 2006). However, less information is available on aldehyde levels urban areas adjacent to dyeing industry-concentrated complex, although dyeing industry is one of urban industries associated with emission of aldehydes, since aldehydes are widely employed in dyeing and finishing processes (Wicks et al., 1994; Xiao et al., 2007; Zhang et al., 2008). The Daegu dyeing industrial complex (DIC) has been established in 1980 in the northwest area of Daegu, which is the third largest city in Korea with a population of nearly 2.5 million and population density of 2812/km<sup>2</sup>. The DIC currently includes over a hundred of dyeing industrial companies and borders on certain residences. According to the Daegu Regional Environmental Management Agency, the number of complaints from nearby residents regarding air pollution has dramatically increased since 1996, although the clear reason for this has not been determined. Residents who live near the complex have continuously complained about potential adverse health effects due to elevated residential exposure to air pollutants emitted from the DIC compared to residents living further away from such a source, although the exact effects of such exposure and exposure evaluations have been based on personal observations and judgment. Such consideration has been supported by the fact that proximity to air pollution sources has long been associated with high personal exposure to environmental pollutants (Gelencser et al., 1997; Jo and Moon, 1999; Fischer et al., 2000). Nevertheless, it is noteworthy that exposure evaluations based solely on proximity to air pollution sources are not robust and require confirmation (Huang and Batterman, 2000).

One important deficiency in relating the environmental exposure to adverse health effects is the lack of measured exposure data (Needham and Sexton,

2000). As such, the current study evaluated residential exposure to atmospheric formaldehyde and acetaldehyde according to distance from the DIC. This purpose was achieved by measuring concurrently the outdoor air concentrations in residences near the DIC and a certain distance away. The outdoor air concentrations at two industrial areas within the DIC boundary were also measured concurrently for comparison with the residential concentrations.

## 2. Methods

### 2.1. Study design

A series of surveys was designed to measure the concentrations of two prevalent aldehydes (formaldehyde and acetaldehyde) in residential outdoor air based on distance from the DIC and in the industrial ambient air within the DIC. No indoor samples were collected for the two aldehydes, as most of the residents in the surveyed homes objected to the noise from the aldehyde sampling pumps. Two residential sites (RS1 and RS2) were selected: RS1, located within 100 meters of the boundary of the DIC and RS2, between 500 and 800 meters away. Fifty households were surveyed in each area for a paired study. Even though RS1 was defined as a residential site within 100 meters of the boundary of the DIC, 44 out of 50 households in RS1 were actually located within 35 meters of the boundary of the DIC. The 100 m distance was the maximum distance from the boundary of the DIC to a house in RS1. All houses had a semi-western style with external structures similar to those of western houses. However, other characteristics, such as the construction materials and home furnishings, were different from traditional western homes. Each pair of households in the two sites was simultaneously surveyed on a standard workday (Monday through Friday) by collecting a daytime 12-hour residential outdoor air sample. These air samples were collected in the yard of each home.

Concurrently, industrial outdoor air concentrations were measured on the roofs of two four-storey buildings within the boundary of the DIC. The two industrial sites (IS1 and IS2) were located near the center of the DIC. At the end of sampling day, the residents in the surveyed homes were interviewed on odor complaints, the use of tobacco products by any family members, the type of fuel used for cooking and heating, the use of solvents, and the presence of any attached garage activities. Meanwhile, meteorological data measured during the sampling period were obtained from the Daegu meteorological office.

## 2.2. Sampling and analysis

The sampling was conducted for three fall months (October, November and December). The sampling and analytical procedure was a modification of the U.S. EPA Method TO-11A (1999). The formaldehyde and acetaldehyde were collected on Sep-Pak DNPH-silica cartridges (Waters, USA) using Handy Samplers (KIMOTO HS-7, Japan) at a nominal flow rate of 100 mL/min. The flow rates were measured using a digital flow meter (Field-Cal 650, Humonics Inc., USA) prior to and following the collection of each sample. The average of these two rates was then used as the sample flow rate in all volume calculations. No samples departed by more than 10% from the initial flow rate during this study. The residential outdoor air samples were collected 1.8 meters from the ground, whereas the industrial outdoor air samples were collected at a height of 1.8 meters from the roof of the industrial companies. To avoid any negative interference from O<sub>3</sub>, a PTFE tube (1-cm i.d. and 10 cm length) with KI coating was used as an O<sub>3</sub> scrubber for all samples (USEPA, 1999; Tejada, 1986). After sampling, the cartridges were capped and stored in a refrigerator prior to extraction.

The aldehydes were extracted with 4 mL of acetonitrile (ACN) (HPLC grade), and then analyzed by a high-performance liquid chromatograph (HPLC)

with UV detection (Shimadzu LC-10A, Japan) (Uchiyama et al., 2004). The chromatographic separations were carried out using a solvent isocratic elution at a flow rate of 1.5 mL/min in an analytical column (0.46 x 250 cm, Shimadzu CLC-ODS (M)), preceded by a C<sub>18</sub> insert guard column. An isocratic run was used for over 25 min with 65% water, 30% ACN, and 5% tetrahydrofuran.

The quality assurance (QA) program included field blank cartridges, spiked samples and duplicate measurements, and inter-laboratory comparisons. To check the quantitative response, known standards of formaldehyde and acetaldehyde prepared from purchased solutions of aldehyde DNPH derivatives (Supelco Inc., USA) were directly injected into a cartridge to transfer the target compounds to the HPLC. When the quantitative response differed by more than 10% from that predicted by the specified calibration equation, a new calibration equation was determined.

## 3. Results

### 3.1. QA results

The mean concentrations measured in the field blanks for formaldehyde and acetaldehyde were 0.18 µg/m<sup>3</sup> with a range of 0.07 to 0.32 µg/m<sup>3</sup> and 0.38 µg/m<sup>3</sup> with a range of 0.33 to 0.63 µg/m<sup>3</sup> (using an assumed sampling volume of 72 L), respectively. The detection limits, defined as 3 times the standard deviation of the field blanks, were 0.13 µg/m<sup>3</sup> for formaldehyde and 0.24 µg/m<sup>3</sup> for acetaldehyde. The laboratories of two institutes compared their analytical techniques by analyzing the same 4-mL ACN extracts of five samples. The analytical errors were within 10% for all five samples. The recovery efficiency was calculated to be 93% for formaldehyde and 95% for acetaldehyde. Twenty duplicate samples were collected for each sampling period to test the precision of the sampling and analytical techniques. The mean relative

standard deviations of the duplicate measurements were less than 10% for the two aldehydes.

### 3.2. Comparison of Two Industrial Sites for Ambient Aldehyde Levels

The statistical summary of the industrial outdoor air concentrations measured on the roofs of two four-storey buildings within the boundary of the DDIC is presented in Table 1. Formaldehyde concentrations (median values of 24.3 and 22.5  $\mu\text{g}/\text{m}^3$  in IS1 and IS2, respectively) were higher than acetaldehyde concentrations (median values of 7.4 and 7.3  $\mu\text{g}/\text{m}^3$  in IS1 and IS2, respectively) at both sites. There was no significant difference in the industrial outdoor air concentrations of both formaldehyde and acetaldehyde between the two sites (Table 2). For these two aldehydes, the industrial outdoor air concentrations were significantly different from the residential outdoor air concentrations.

### 3.3. Comparison of Residential and Industrial Sites for Ambient Aldehyde Levels

The summary of the indoor and outdoor air

concentrations of formaldehyde and acetaldehyde measured in the two residential areas (RS1 and RS2) according to distance from the DIC are also displayed in Table 1. RS1 was within 100 meters of the boundary of the DDIC, while RS2 was between 500 and 800 meters away. Similar to the industrial outdoor air concentrations, formaldehyde concentrations were significantly higher than acetaldehyde concentrations in the outdoor air from both residential areas (Table 2). In addition, the outdoor concentrations of both formaldehyde and acetaldehyde were significantly different between RS1 and RS2 ( $p < 0.05$ ). The median formaldehyde concentration from RS1 was about 1.5 times higher than that from RS2, and the median acetaldehyde concentration from RS1 was about 1.3 times higher than that from RS2. The real-time outdoor concentrations were almost always higher in RS1, although for a few sampling periods, the results were reversed.

### 3.4. Correlation between residential and industrial sites

Non-parametric correlation analysis of the outdoor

**Table 1.** Summary of aldehyde concentrations ( $\mu\text{g}/\text{m}^3$ ) measured in two residential sites (RS1 and RS2) and two industrial sites (IS1 and IS2)<sup>a</sup>

Site	Formaldehyde				Acetaldehyde			
	Mean	SD	Median	Range	Mean	SD	Median	Range
RS1	19.5	5.7	18.8	9.7-35.2	5.6	4.9	4.8	1.0-19.8
RS2	13.7	4.8	12.7	4.7-28.2	4.1	1.8	3.7	1.1-8.1
IS1	25.1	5.8	24.3	17.2-38.3	8.0	3.3	7.4	2.4-15.9
IS2	24.6	5.6	22.5	16.9-39.2	7.5	3.2	7.3	1.8-20.1

<sup>a</sup>RS1, residential site near dyeing industrial complex (DIC); RS2, far away from the DIC; IS1, industrial site 1; IS2, industrial site 2; and SD, standard deviation.

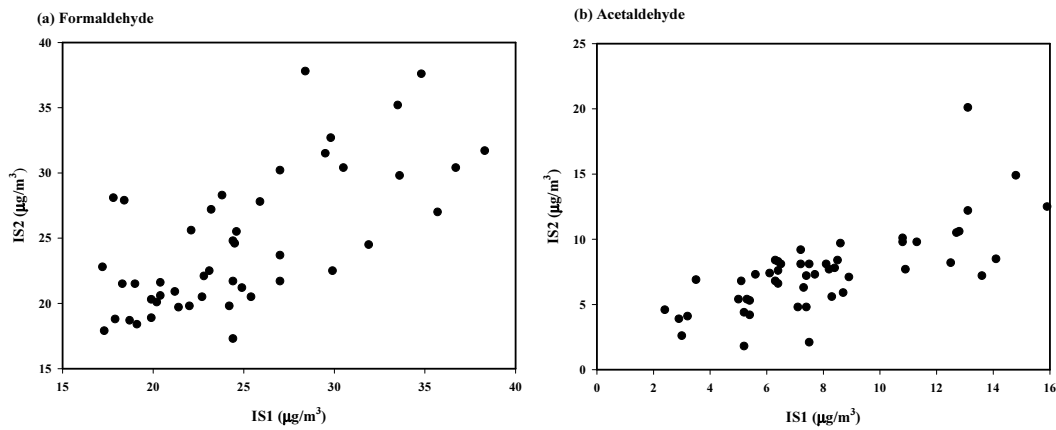
**Table 2.** Duncan's Multiple Range Group for formaldehyde and acetaldehyde concentration difference between sampling sites<sup>a</sup>

Aldehyde	IS1	IS2	RS1	RS2
Formaldehyde	A	A	B	C
Acetaldehyde	A	A	B	C

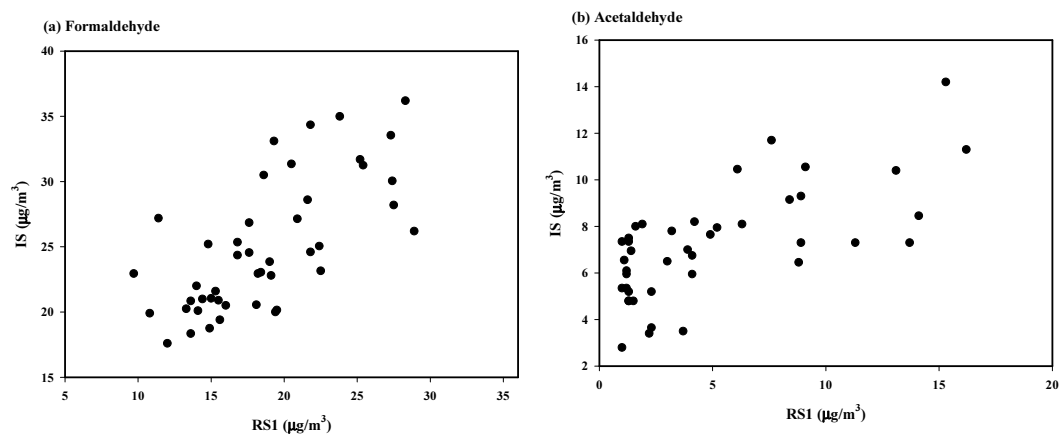
<sup>a</sup>IS1, industrial site 1; IS2, industrial site 2; IS, combined data of IS1 and IS2; RS1, residential site near dyeing industrial complex (DIC); and RS2, far away from the DIC. In Duncan's grouping, the sampling sites sharing different letter represent a significant difference from other site(s).

air levels between the industrial and residential areas was performed. Since similar correlation patterns were found for the two industrial sites (Fig. 1), instead of using separate concentration distributions, a combined concentration distribution of the two industrial sites was employed to calculate the correlations between the industrial and residential areas. The combined concentration distribution was obtained by averaging the paired concentrations measured in the two industrial sites on each sampling day. The correlation

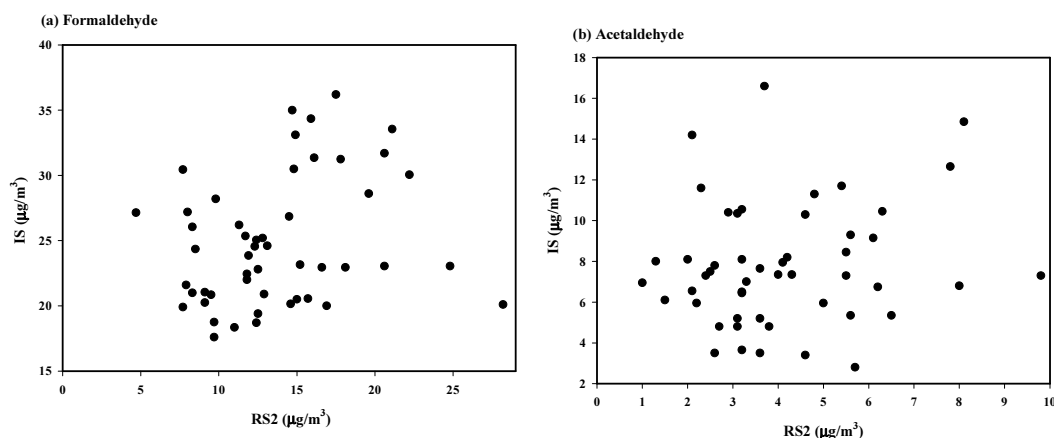
in the both formaldehyde and acetaldehyde levels between industrial areas and RS1 was statistically significant (Fig. 2), whereas the correlation between industrial areas and RS2 was not statistically significant for either formaldehyde or acetaldehyde levels (Fig. 3). Meanwhile, the correlation between RS1 and RS2 was not statistically significant for either formaldehyde or acetaldehyde levels (Fig. 4).



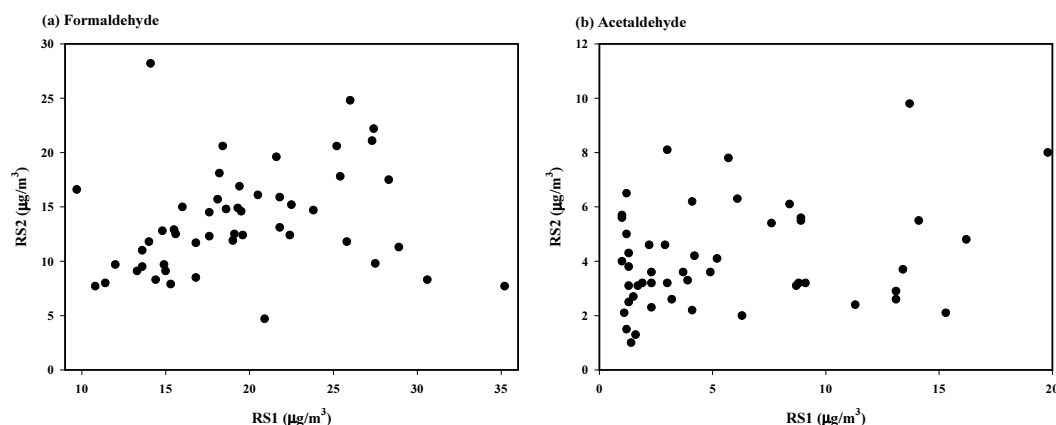
**Fig. 1.** Correlation of aldehyde concentrations ( $\mu\text{g}/\text{m}^3$ ) between two industrial sites (IS1 and IS2): (a) formaldehyde ( $R^2 = 0.47$ ) and (b) acetaldehyde ( $R^2 = 0.53$ ).



**Fig. 2.** Correlation of aldehyde concentrations ( $\mu\text{g}/\text{m}^3$ ) between residential site (RS1) near a dyeing industrial complex and industrial sites (IS): (a) formaldehyde ( $R^2 = 0.49$ ) and (b) acetaldehyde ( $R^2 = 0.46$ ). The industrial-site data represent average of those of two industrial sites (IS1 and IS2).



**Fig. 3.** Correlation of aldehyde concentrations ( $\mu\text{g}/\text{m}^3$ ) between residential site (RS2) far away from a dyeing industrial complex and industrial sites (IS): (a) formaldehyde ( $R^2 = 0.07$ ) and (b) acetaldehyde ( $R^2 = 0.03$ ). The industrial-site data represent average of those of two industrial sites (IS1 and IS2).



**Fig. 4.** Correlation of aldehyde concentrations ( $\mu\text{g}/\text{m}^3$ ) between residential site (RS1) near a dyeing industrial complex (DIC) and residential site (RS2) far away from the DIC: (a) formaldehyde ( $R^2 = 0.05$ ) and (b) acetaldehyde ( $R^2 = 0.07$ ).

### 3.5. Residents' interview

The residents in the surveyed homes were interviewed for any odor complaints, exposure information, and their daily activities related to the 12-hour period preceding the sampling periods. The nearby residents complained of a weak or strong odor on most of the sampling days, whereas the residents living further away from the DIC rarely complained for any odor. Most but not all of the houses used liquid propane gas or electricity for cooking and heating. As usual, none

of the participants reported any use of the target aldehydes in the home during the monitoring period. None of the households had an attached garage. Furthermore, no other potential sources of the target aldehydes were observed around the surveyed homes.

## 4. Discussion

The current study measured the outdoor air levels of two prevalent carbonyl compounds (formaldehyde

and acetaldehyde) within the DIC boundary and two residential areas, one adjacent to and the other a certain distance away from the DIC. The industrial outdoor air concentrations of both formaldehyde and acetaldehyde substantially higher than the residential outdoor air concentrations. Based on the knowledge that these compounds are employed in dyeing and finishing processes (Wicks et al., 1994; Xiao et al., 2007; Zhang et al., 2008), it is suggested that the DIC is the primary cause for the elevated industrial levels of the two aldehydes. However, the outdoor air concentrations of formaldehyde and acetaldehyde were significantly higher in RS1 than in RS2. A possible cause for this discrepancy is the amount of these compounds used as solvent in textile coating process. According to the DIC, formaldehyde is employed much more than acetaldehyde in the process, even though information on exact amounts are unavailable. Furthermore, the outdoor air concentrations in RS1 substantially exceeded other residential outdoor air concentrations in earlier studies (Liu et al., 2006; Marchand et al., 2006), whereas those in RS2 were compatible with. Liu et al. (2006) previously reported on median residential outdoor air concentrations of formaldehyde and acetaldehyde at 6.4 and 5.4  $\mu\text{g}/\text{m}^3$ , respectively. Marchand et al. (2006) reported on mean residential outdoor air concentrations of formaldehyde and acetaldehyde at 6.8 and 5.3  $\mu\text{g}/\text{m}^3$ , respectively. Although motor vehicles are closely associated with ambient levels of formaldehyde (Schmid et al., 2001; Kristensson et al., 2004), according to the city-traffic department, traffic loads in the two residential areas are not significantly different. As such, motor vehicles were eliminated as

the major cause for the difference in the present outdoor air aldehyde levels between the two residential areas. Rather, the higher aldehyde levels in RS1 would primarily be attributed to transportation of aldehydes from the DIC, since the other known major sources were not observed down-stream or around the DIC, or around the residential areas. This assertion is further supported by three parameters: correlation of outdoor air levels between industrial and residential sites; meteorological conditions during sampling period; and interview results on odor. The aldehyde levels measured at the industrial site and from RS1 were significantly correlated, although there was no significant correlation in the levels of the other target compounds. The frequencies of wind directions monitored during sampling period are shown in Table 3. Three most frequent wind directions (W, 16%; NW, 38%; and SW, 8%) represented above 60% of wind direction. Meanwhile, one wind direction (SE) exhibited the frequency of wind direction just 1%. The wind direction over the remaining time was distributed uniformly across the other compass points. Since the two residential sites are located in the east-southern area of the DIC, the wind directions between N and W correspond to downwind for the residential areas from the DIC. These wind directions favor transporting air pollutants emitted from the DIC to RS1, thereby causing elevated aldehyde levels in the residential site. Finally, the interview results implied that the residential aldehyde levels in RS1 were closely associated with residential odor nuisance due to air pollutants transported from the DIC, even though this information is subject to a certain error and bias due to the personal judgment involved when assessing the

**Table 3.** Average wind speed and direction frequency in the study area

Wind speed (m/s)	Wind direction frequency (%)							
	N	NW	W	SW	S	SE	E	NE
2.5	4	38	16	8	1	11	6	16

magnitude of the nuisance.

The exposure levels were not absolutely, but were relatively estimated for residents, who live in near and far away the DIC, assuming that atmospheric aldehyde levels also influence linearly the indoor aldehyde exposure levels. As such, exposure time was assumed to be 24 h for both outdoor and indoor exposure. The personal exposure levels to aldehydes for the residents were estimated from the equation of the USEPA (1992):

$$E_p = C \times BR \times t$$

where  $C$  is the concentration of aldehydes ( $\mu\text{g}/\text{m}^3$ ),  $BR$  is the inhalation rate ( $\text{m}^3/\text{h}$ ), and  $t$  is the exposure time (h/day). For exposure calculus, the breathing rate of air was estimated for an average person ( $BR = 0.63 \text{ m}^3/\text{h}$ ) according to EPA exposure factors (USEPA, 1990). A daily residence time of 24 h was considered as exposure time ( $t$ ). Mean, median and maximum aldehyde concentrations in the unit of  $\text{mg}/\text{m}^3$  converted from  $\mu\text{g}/\text{m}^3$  unit were utilized to calculate the mean, median and maximum risk, respectively. Inhalation unit risk estimates of  $1.3 \times 10^{-5} (\text{mg}/\text{m}^3)^{-1}$  (USEPA, 1991a) and  $2.2 \times 10^{-6} (\text{mg}/\text{m}^3)^{-1}$  (US EPA, 1991b) were used for the calculation of risks from formaldehyde and acetaldehyde exposure, respectively.

The aldehyde exposure estimates from inhalation of air are presented in Table 4. The maximum exposure estimates of formaldehyde and acetaldehyde were 532 and 299  $\mu\text{g}/\text{m}^3$ , respectively, for residents who live near the DIC, whereas they were 426 and 122  $\mu\text{g}/\text{m}^3$  for those living far away from the DIC. These

exposure levels for the residents who live near the DIC result in the maximum risk estimates of  $4.6 \times 10^{-4}$  and  $4.4 \times 10^{-5}$  from formaldehyde and acetaldehyde exposure, respectively. For those living far away from the DIC, the maximum risk estimates were  $4.6 \times 10^{-4}$  and  $4.4 \times 10^{-5}$  from formaldehyde and acetaldehyde exposure, respectively. It is noteworthy that the mean or median risk as well as these maximum risks are well above the USEPA's permissible risk level of  $10^{-6}$  from environmental exposure (USEPA, 1999). Accordingly, it is suggested that appropriate management for formaldehyde and acetaldehyde is necessary in order to decrease risk of the residents of study areas, regardless of the distance from the DIC.

## 5. Conclusions

It was confirmed that the atmospheric levels of both formaldehyde and acetaldehyde was higher in residential site near the DIC as compared with those for residential site far away from the DIC, resulting in elevated exposure and corresponding health risk. This result was primarily attributed to transportation of aldehydes from the DIC, since the other known major sources were not observed down-stream or around the DIC, or around the residential areas. The result was further supported by three parameters: correlation of outdoor air levels between industrial and residential sites; meteorological conditions during sampling period; and interview results on odor.

**Table 4.** Relative exposure ( $\mu\text{g}/\text{day}$ ) of residents living near (RS1) and far away (RS2) from dyeing industrial complex and corresponding risk estimation

Aldehyde	Residents	Exposure			Risk		
		Mean	Median	Max	Mean	Median	Max
Formaldehyde	RS1	295	284	532	$2.5 \times 10^{-4}$	$2.4 \times 10^{-4}$	$4.6 \times 10^{-4}$
	RS2	207	192	426	$1.8 \times 10^{-4}$	$1.7 \times 10^{-4}$	$3.7 \times 10^{-4}$
Acetaldehyde	RS1	85	57	299	$1.2 \times 10^{-5}$	$1.0 \times 10^{-4}$	$4.4 \times 10^{-5}$
	RS2	62	56	122	$0.9 \times 10^{-5}$	$0.8 \times 10^{-5}$	$1.8 \times 10^{-5}$



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