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Indoor and Built Environment 2005 14: 165
DOI: 10.1177/1420326X05052564

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Exposure Assessment for Formaldehyde and Acetaldehyde in the Workplace

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Key Words

Aldehyde · Formaldehyde · Acetaldehyde · Risk assessment · Occupational health · Indoor environment

Abstract

Personal exposure and possible cancer risk to formaldehyde and acetaldehyde were appraised in 8 work places at a university in Brazil. Levels of formaldehyde measured ranged from 22.5 to 161.5 $\mu\text{g}\cdot\text{m}^{-3}$ and from 18.3 to 91.2 $\mu\text{g}\cdot\text{m}^{-3}$ for acetaldehyde. The personal exposure, expressed as the potential dose in indoor air, was calculated to range from 129.8 to 930.4 $\mu\text{g}\cdot\text{day}^{-1}$ (low exposure) and 183.9 to 1318.1 $\mu\text{g}\cdot\text{day}^{-1}$ (medium exposure) for formaldehyde and 105.5 to 525.3 $\mu\text{g}\cdot\text{day}^{-1}$ (low exposure) and 149.5 to 744.2 $\mu\text{g}\cdot\text{day}^{-1}$ (medium exposure) for acetaldehyde. The indoor/outdoor ratio showed the existence of indoor sources of the compounds which were mainly in practical classes and research laboratories. The highest formaldehyde and acetaldehyde levels were found where chemical reagents were manipulated. Relating the levels found to the permissible limit given by the US OSHA showed there was no particular risk

although some formaldehyde levels did exceed the lower exposure limit of the US agency NIOSH. Any cancer risk would be highest for female technicians and teaching researchers.

Introduction

The quality of air in work places [1], schools [2,3], leisure centres [4] and homes [5,6,7] has been monitored in several countries over the last 2 decades. Such buildings usually have a low air change rate and this results in a considerable increase in the concentration of any chemical compounds produced in those places. Moreover, we spend more than 90% of our time in indoor environments including houses, offices, leisure centres shopping malls, restaurants, libraries and others so increasing any exposure and consequently the risk to human health [8,9]. In consequence, the World Health Organisation (WHO) recognises disease produced by poor indoor air quality as a public and occupational health problem [10].

Recently, a working group, convened by the IARC Monographs Programme [11] have concluded that formaldehyde is carcinogenic to humans. Based on new information, the expert working group has determined

that there is now sufficient evidence that exposure to formaldehyde can cause naso-pharyngeal cancer in humans, a rare cancer in developed countries. Formaldehyde was previously classified by IARC as Group 2A (probably carcinogenic to humans) but has now been reclassified as Group 1 [12]. To date its permissible exposure limit (PEL) is unchanged and varies from 0.016 to 0.75 ppm averaged over an 8-h workshift, but may not exceed a 0.1 ppm average for longer than 15 min of the work period [13]. Presenting less hazard to health, acetaldehyde is classified by IARC as group 2B, a possible human carcinogen based on sufficient evidence in animals and inadequate evidence in humans [12] and the legal airborne PEL is 200 ppm averaged over 8-h workshift [13].

People exposed for some time to certain chemical compounds, mainly in work places (industries, laboratories, hospitals, etc.) have a high probability of acquiring degenerative diseases [14]. This must include employees, teachers and students (though these have a smaller exposure) exposed to a number of chemical compounds in schools, research centres and universities, during the academic year, when carrying out research or simply by working near to places handling such compounds (bystander exposure).

The risk to human health may be estimated by calculation of the increased probability of development of cancer and is usually estimated only for subjects that suffer chronic exposure for an appreciable part of their working life [15, 16].

The objective of the present study was a preliminary evaluation of the exposure to formaldehyde and acetaldehyde experienced by employees and students working in a Brazilian university and from these to estimate the risk to their health.

Materials and Methods

Sampling Sites

Samples were taken indoors at the university during term-time when students, other employees, teachers and teacher-researchers were present. The places sampled were offices, classrooms, research laboratories, laboratories used by practical classes, a library, a print room used for letterpress printing and outdoors. The samples were taken in the laboratories of chemistry, biochemistry, physiology, chemical engineering and food processing, places where practical classes used several chemical compounds. The main characteristics of each sampling site are shown in Table 1.

Table 1. Characteristics of the sampling sites

| Sampling sites (n = 2) | Activity | Number of people (average) | Ventilation type |
|------------------------|----------------------------|----------------------------|------------------|
| WP1 | office room | 1 | natural |
| WP2 | office room | 1 | air-conditioned |
| WP3 | classroom | 40* | natural |
| WP4 | research laboratory | 5* | natural |
| WP5 | class practical laboratory | 20* | natural |
| WP6 | class practical laboratory | 20* | air-conditioned |
| WP7 | central library | 10* | natural |
| WP8 | print room | 2 | natural |
| EP | outdoors | | |

*Estimated.

A database was used to obtain information on the chemical agents used at the places studied, other potential sources, residence time, personal activities, etc.

Sampling

The sampling methodology used followed the proposals of Kuwata et al. [17], using classical gas sampling coupled through a C₁₈ Sep-Pak column (Waters, Milford, MA) impregnated with 2,4-dinitrophenylhydrazine (2,4-DNPH) in acid. The downstream end of the cartridge was connected to a calibrated flow meter. During a sampling period, air passed through the cartridge at a flow rate of 0.8 to 1.2 L·min⁻¹ (total sample volume of 100 L). Each cartridge was sealed with Teflon[®] tape immediately after sampling, then wrapped in aluminium foil and refrigerated.

Analysis

The cartridges were eluted with 5 ml of HPLC-grade acetonitrile (ACN) and a 20 µl volume injected into an HPLC Shimadzu (model LC-10 AD). The analytical conditions were as follows: MetaChem Technologies C₁₈ column (25 × 0.46 cm, 5 µm); gradient mobile phase: 70–30% ACN/water solution for 7 min, 77–23% ACN/water for 2 min and then 70/30% ACN/water for 1 min; the mobile-phase flow rate was 1 mL·min⁻¹. The DNPH derivatives were detected at 350 and 365 nm with a Diode array (SPD-10AVP) UV/VIS detector. A calibration curve was constructed by direct injection of standard mixtures with known amounts of 2,4-DNPH-derivatives in ACN. Cartridge blanks were analysed to determine the background level of 2,4-DNPH-derivatives.

Exposure to Formaldehyde and Acetaldehyde

The exposure for an individual can be calculated as the average of the exposure, expressed as a potential dose (PD). The exposure (PD) for an individual (i) due to intake processes (inhalation or ingestion) can be calculated from the equation of the USEPA [18]:

$$PD_i = \sum_{j=1}^n C_j IR_i T_{ij}$$

where C is the concentration of the pollutant ($\mu\text{g}\cdot\text{m}^{-3}$), IR is the inhalation rate ($\text{m}^3\cdot\text{h}^{-1}$), T is the exposure time ($\text{h}\cdot\text{day}^{-1}$), and j the microenvironment.

Due to the difficulty of measuring precisely the correct inhalation rate for each individual, the PD was estimated for a time of 8 h (for exposure) and using the IR for low inhalation ($0.75\text{m}^3\cdot\text{h}^{-1}$) and medium inhalation ($1.02\text{m}^3\cdot\text{h}^{-1}$), as described in Exposure factors handbook – USEPA [19].

Risk Assessment to Formaldehyde and Acetaldehyde

The calculation of the risk assessment from exposure to formaldehyde and acetaldehyde was estimated using the chronic daily intake – CDI ($\text{mg}\cdot\text{kg}\cdot\text{day}^{-1}$) calculated from the following equation [18–21]:

$$CDI = (CA \cdot IR \cdot ED \cdot EF \cdot L) / (BW \cdot ATL \cdot NY)$$

where CA is the contaminant concentration ($\text{mg}\cdot\text{m}^{-3}$); IR the inhalation ratio ($\text{m}^3\cdot\text{h}^{-1}$); ED the exposure duration ($\text{h}\cdot\text{week}^{-1}$); EF the exposure frequency ($\text{week}\cdot\text{year}^{-1}$); L the length of exposure (years); BW the body weight (kg); ATL the average lifetime (the period over which exposure is averaged, 63 years) and NY the number of days per year (365 days).

To facilitate the estimate of the calculated risk the following were assumed: 1.02m^3 as the inspired air per hour, a body weight of 70kg for men and 60kg for women. In Brazil employees spend at work places, on average, 8h per day, 5 days per week totalling 251 days of work in the year. Thus, 2,008h of annual exposure and 80,320h taken as the length of time of exposure over a working lifetime (equivalent to 40 years of exposure).

Results

Level of Carbonyl Compounds and Ratio

The carbonyl compounds were found in amounts ranging from 22.5 to $161.5\mu\text{g}\cdot\text{m}^{-3}$ for formaldehyde and 18.3 to $91.2\mu\text{g}\cdot\text{m}^{-3}$ for acetaldehyde (Table 2). The highest levels were observed at the places where chemical agents were handled; research laboratories and practical classes (WP4, WP5 and WP6).

The formaldehyde/acetaldehyde ratio ranged from 1.1 to 1.8 following the patterns of other Brazilian studies [9,22]. However, the outdoor ratio (Table 2) is at variance with reports for most Brazilian studies but is similar to that found in other parts of the world.

The health-based limit suggested by Aquino Neto and Brickus [23] for formaldehyde was used to evaluate the levels in indoor air. Although not accepted as an official limit, the level is sensibly based and is used for real world situations in Brazil. Using this showed that levels at the WP6 site were 60% above the limit level suggested (Figure 1) while those at the WP4 site, although below, are however very close of the limit.

Indoor/Outdoor (I/O) Ratio

The I/O ratio of formaldehyde and acetaldehyde ranged from 1.2 to 8.9 and 1.2 to 5.9, respectively (Figure 2). The highest values were found at the WP4, WP5 and WP6 sites.

Potential Dose (PD)

The personal exposure, expressed as PD for the levels of formaldehyde and acetaldehyde measured in the indoor air, are given in Table 3. The highest personal exposure observed was at the WP4 and WP6 sites.

Table 2. Formaldehyde (HCHO) and acetaldehyde (CH_3CHO) levels

| Sampling sites | HCHO ($\mu\text{g}\cdot\text{m}^{-3}$) | CH_3CHO ($\mu\text{g}\cdot\text{m}^{-3}$) | HCHO/ CH_3CHO |
|----------------|--|---|-------------------------------|
| WP1 | 32.3 | 18.3 | 1.8 |
| WP2 | 41.0 | 26.2 | 1.6 |
| WP3 | 22.5 | 19.0 | 1.2 |
| WP4 | 96.5 | 79.4 | 1.2 |
| WP5 | 56.5 | 38.1 | 1.5 |
| WP6 | 161.5 | 91.2 | 1.8 |
| WP7 | 36.5 | 33.0 | 1.1 |
| WP8 | 42.2 | 35.6 | 1.2 |
| Outdoors | 18.2 | 15.4 | 1.2 |

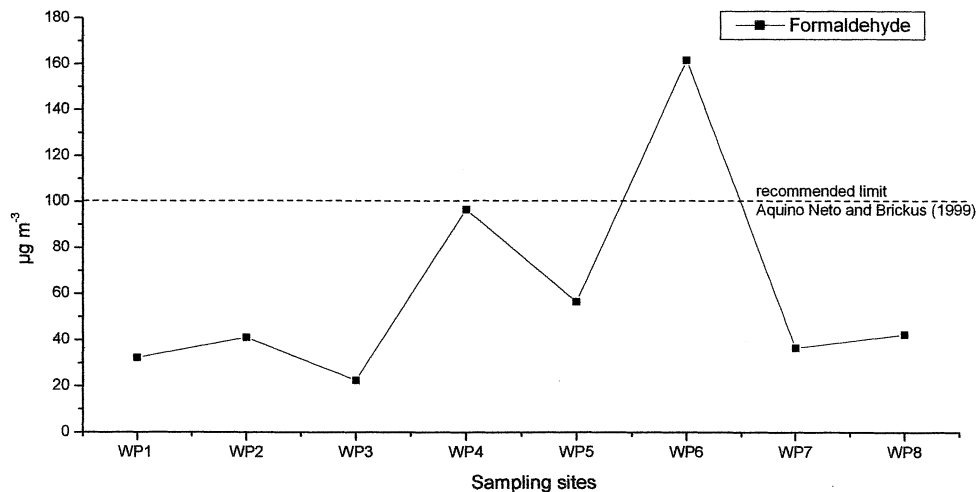


Fig. 1. Distribution of formaldehyde and acetaldehyde levels.

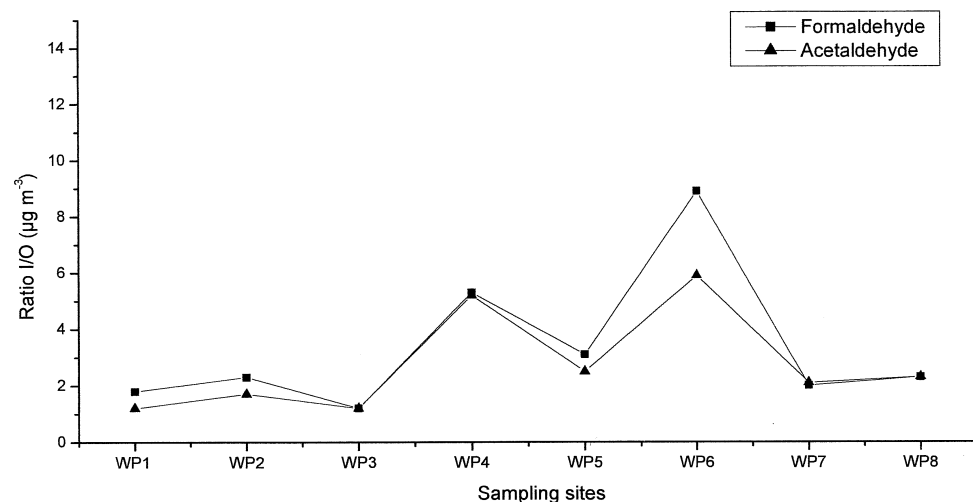


Fig. 2. Indoor/outdoor ratio of formaldehyde and acetaldehyde.

CDI

The values for the CDI, averaged over a lifetime, for each contaminant by each exposure pathway ranged from 8.22×10^{-4} to $6.13 \times 10^{-3} \text{ mg} \cdot \text{kg}^{-1} \cdot \text{day}^{-1}$ for formaldehyde and 9.48×10^{-4} to $3.46 \times 10^{-3} \text{ mg} \cdot \text{kg}^{-1} \cdot \text{day}^{-1}$ for acetaldehyde. In all the places the risk for women was larger due a smaller body size (Figure 3(a) and 3(b)).

Discussion

Formaldehyde and Acetaldehyde Levels

The formaldehyde and acetaldehyde levels found in the places studied, excluding the laboratories, were similar to the levels reported by other authors (see Table 4). However, it was shown that at the WP2 site, which

was air-conditioned, the levels were enriched by around 79% for formaldehyde and 70% for acetaldehyde, when compared to the WP1 site which had similar characteristics but no air-conditioning. This increased concentration was expected and is due to the liberation of carbonyl compounds from various construction and furnishing materials in the offices and human activity [7,24], compounded by the low renewal of air in air-conditioned rooms. In addition, cleaning products are potential sources of various chemical compounds in the air of offices and houses. According to Wolkoff et al. [6] and Nazaroff and Weschler [25] such products release high levels of VOCs, that remain in ambient air for a long time if the air-change rate is low.

The highest levels of formaldehyde and acetaldehyde were measured in WP6 followed by WP4 and WP5.

Table 3. Personal exposure to formaldehyde and acetaldehyde

| Sites | Formaldehyde ($\mu\text{g}\cdot\text{day}^{-1}$) | | Acetaldehyde ($\mu\text{g}\cdot\text{day}^{-1}$) | |
|-------|--|-----------------|--|-----------------|
| | Low exposure | Medium exposure | Low exposure | Medium exposure |
| WP1 | 186.2 | 263.8 | 105.5 | 149.5 |
| WP2 | 236.3 | 334.8 | 151.1 | 214.0 |
| WP3 | 129.8 | 183.9 | 109.6 | 155.3 |
| WP4 | 555.8 | 787.4 | 457.3 | 647.8 |
| WP5 | 325.6 | 461.3 | 219.3 | 310.7 |
| WP6 | 930.4 | 1318.1 | 525.3 | 744.2 |
| WP7 | 210.2 | 297.7 | 190.3 | 269.6 |
| WP8 | 243.3 | 344.7 | 204.8 | 290.2 |

These are all places where practical classes in physiology and organic chemistry are held periodically. The explanation for the extreme levels found at the WP6 site is because of the use of several direct and precursor sources of formaldehyde and acetaldehyde in those rooms.

Overall, the main factors responsible for high levels are the high use of chemical compounds and the low renewal of the indoor air (often because of air-conditioning), this last fact is considered by many authors [8,25,26].

Formaldehyde/Acetaldehyde Ratio

According to various Brazilian authors [22,27–28], the use of ethanol in the national fuel produces ambient levels in which the formaldehyde/acetaldehyde ratio, is less than 1. This is in contrast to the ratios observed in this study, which were larger than 1. Since our results are single measurements and may differ at other times, this behaviour should be verified more precisely. The existence of a refinery, approximately 50m from the area of study may have contributed to the levels of compounds measured.

Indoor/Outdoor Ratio

Analysis of the data show the existence of point sources at the WP4, WP5 and WP6 sites due to handling of a variety of chemical reagents. Elsewhere, the I/O ratio does not exceed 2.3 for formaldehyde and acetalde-

hyde. These values are below the mean levels for formaldehyde (2.6) and acetaldehyde (2.1) found in offices in Rio de Janeiro by Brickus et al. [29].

Personal Exposure and Risk Assessment

The PD calculation shows the level of exposure for the various groups of people to chemical compounds. As expected, people that work in places where chemical agents are handled are subject to larger exposures, increasing their risk of health effects.

At the WP1 and WP2 sites, the personal exposures were smaller when compared to those measured in studies in offices in Mexico [24]. However according to the authors of this study, in addition to common potential sources in offices for aldehydes, the existence of smokers increases considerably the concentrations of the studied compounds and consequently people's exposure.

There are several difficulties that limit the estimate of the risk assessment, mainly due to several variables that cannot be defined precisely when studying groups of people. Guo et al. [1] describes the factors that limit and hinder even a rough estimate of the risk assessment. However, by making some assumptions an estimate can be made and used in political argument concerned with people's occupational health, as well as to monitor those things that influence professional activity in workers.

The inhalation or breathing rate is an important variant and can increase the uncertainty in any estimate of risk. Linn et al. [30] carried out a study that estimated the inhalation rates for "high-risk" sub-population groups exposed to ozone (O_3) in their daily activities in the Los Angeles area. In this study several factors were recorded for each group: their daily activities; change in location (indoors to outdoors, or in a vehicle); self-estimated inhalation rates and time spent during each activity or at each location. Subjective inhalation rates were defined as slow (normal walking); medium (faster than normal walking); and fast (hard physical work or strenuous exercise) and the values calculated for healthy adults were $0.72\text{ m}^3\cdot\text{h}^{-1}$ (slow), $1.02\text{ m}^3\cdot\text{h}^{-1}$ (medium) and $3.06\text{ m}^3\cdot\text{h}^{-1}$ (fast) while the mean for asthmatic adults was

Table 4. Comparisons of formaldehyde and acetaldehyde levels with other sites

| Site | Formaldehyde ($\mu\text{g}\cdot\text{m}^{-3}$) | Acetaldehyde ($\mu\text{g}\cdot\text{m}^{-3}$) | Activity | Reference |
|----------------|--|--|------------|-----------|
| Mexico City | 97.0 | 47.0 | office | [24] |
| São Paulo | 29.0 | 17.0 | library | [30] |
| Hong Kong | 27.0 | – | classroom | [2] |
| Rio de Janeiro | 21.3 | 15.0 | print room | [9] |

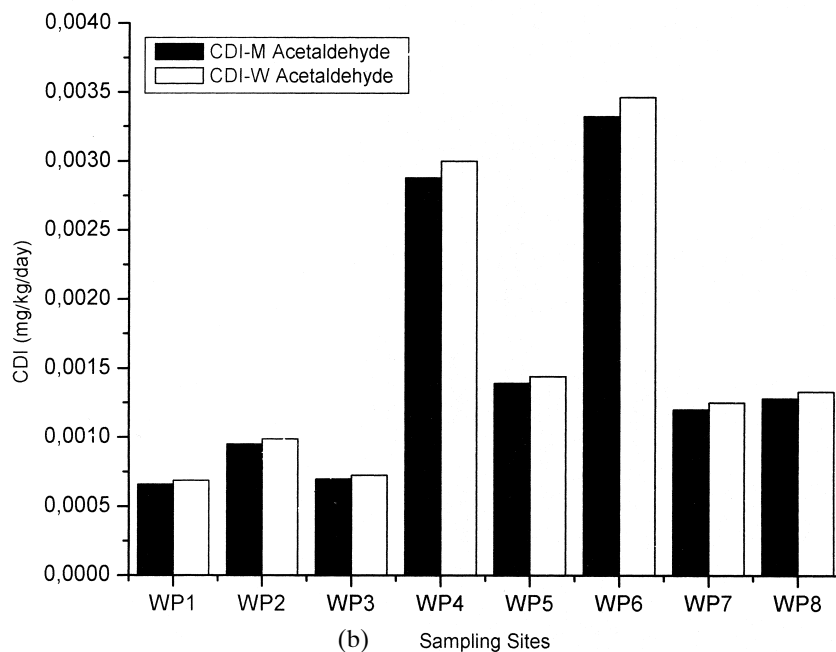
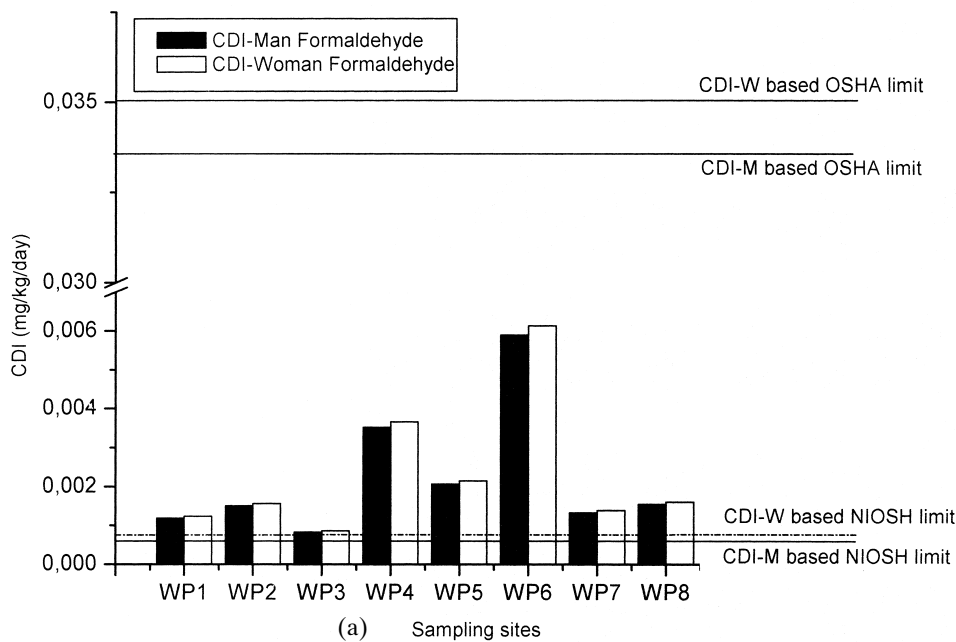


Fig. 3. Chronic daily intake of formaldehyde (a) and acetaldehyde (b).

$1.02\text{ m}^3\text{ h}^{-1}$. This same author reported that the diary data showed that most individuals spent most of their time (in a typical day) indoors at a low activity level.

An advantage of this type of study is that diary data can provide rough estimates of inhalation rates which are useful in exposure assessments for gaseous chemical compounds. Another advantage is that inhalation rates were divided into those for various sub-populations (i.e.,

healthy outdoor adult workers, healthy children, asthmatics and construction workers).

Several international, national and state authorities have established regulations or guidelines for the use and production of formaldehyde, because of an assumed cancer risk (now confirmed [11]). The difficulty is knowing what level of the compound to permit as an exposure limit where the probability that such exposure

will cause disease is effectively zero. As with most chemical exposures, the question does not have an easy answer. From the 1980s to the present time the various limits have decreased and are now less than 10% of the earlier limits [31]. With the recent reclassification by IARC [11], exposure limits for formaldehyde may be reduced lower still.

The US Occupational Safety and Health Administration (OSHA) have established a Permissible Exposure Limit (PEL) of 0.75 ppm for an 8h Time-Weighted Average (TWA) [13]. A more rigid criterion is the Recommended Exposure Limit (REL) for occupational exposure at 0.016 ppm in 8h (TWA) defined by the US Institute for Occupational Safety and Health (NIOSH) [13].

Using the OSHA limit (converted for mg·kg·day) none of the places monitored had levels that exceeded this standard (see Figure 3(a)). Levels of acetaldehyde were less than 1% of the limit values (13.12 mg·kg·day for men and 13.67 mg·kg·day for women). These places can therefore be classified as of low risk. On the other hand,

using the more stringent exposure limit of NIOSH, we have shown that at all the places studied the cancer risk varied from medium to high for formaldehyde (see Figure 3(a)). This would apply mainly to technical staff and teacher-researchers who work in the classrooms, practical classes and research laboratories for usually more than 8 h per day over 40 years of academic activity.

The teachers who only take classes (practical and theoretical) are moderately exposed over their academic life. The students are exposed to only a small risk due to the relatively short time they spend in practical classes. The data also showed that women are more exposed than men and consequently have a higher cancer risk.

Acknowledgements

The authors thank Mr F.V.M. Pereira for his technical assistance and Dr João Sarkis Yunes for the donation of C18 Sep-Pak cartridges.

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