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**SUMMARY**
Indoor air quality surveillance in public premises, especially those hosting vulnerable populations such as children, was introduced in the second French national environment and health action plan and then regulated by the first “Grenelle Environnement” law, on August 3rd, 2009. A national pilot monitoring survey of indoor air quality in 310 French schools and day-care centres was performed in two phases from 2009 to 2011. The results of the first phase show that air quality is overall acceptable in 90% of the establishments with respect to the management values suggested by the French committee for public health. Nonetheless, a few cases required additional diagnoses or corrective measures. Furthermore, air stuffiness (based on $CO_2$ measurements) was found to be very high in 16% of the classrooms. The Mayors and School Principals were informed and provided with means to identify the main sources of pollution and to implement remediation actions.

**KEYWORDS**
Air pollution, air stuffiness, formaldehyde, benzene.

## 1 INTRODUCTION

Indoor air quality surveillance in public premises, especially those hosting vulnerable populations such as children, was introduced in the second French national environment and health action plan (NEHAP) and then regulated by the first “Grenelle Environnement” law, on August 3rd, 2009. A national pilot monitoring survey of indoor air quality in 310 French schools and day-care centres was performed in two phases from 2009 to 2011 (160 in 2009-2010 and 150 in 2010-2011).

The goal of this pilot monitoring survey was to validate monitoring protocols and management procedures to be implemented (*i.e.*, sampling strategy, pollutant source identification, remediation, etc.). The survey focused on two priority pollutants: formaldehyde, irritant to the nose and the respiratory tract and probable carcinogen, emitted by some building materials, furniture, glues, cleaning products; benzene, carcinogen, originating from combustion processes (exhaust gas in particular). Carbon dioxide was also monitored in order to determine an air stuffiness index as an indirect mean to assess pollutants accumulation in a closed space.

In addition, a simple audit of each building was carried out, including a description of each investigated room, heating and ventilation systems and cleaning habits. This audit could provide first clues of explanation when high concentrations were observed (failing ventilation system, specific sources...).

## 2 MATERIAL AND METHODS

### Chemical indicators: formaldehyde and benzene

The implemented protocols for formaldehyde and benzene monitoring were based on those developed through the work of the Central Laboratory for Air Quality Monitoring (LCSQA, 2008a).

Formaldehyde and benzene were measured using passive diffusion radial tubes. To be as representative of a long-term exposure of the children as possible, measurements were performed from Monday morning to Friday afternoon, *i.e* during 4.5 days (scholar holidays were not taken into account). As formaldehyde concentrations can vary greatly from season to season, this pollutant and benzene were measured at two different seasons (so called "winter (W)" and "summer (S)"). For the needs of the study, "summer" and "winter" periods were defined as follows: « summer » extends from mid-september to mid-october and from april to may ; « winter » extends from november to february. The analysis of formaldehyde was performed by chemical desorption, followed by high performance liquid chromatography (HPLC) coupled with UV detection. The analysis of benzene was carried out by thermal desorption, followed by gas chromatography (GC) coupled with flame ionization detection (FID) and mass spectrometry (MS). Details of the analytical methods are given in the LCSQA protocols (LCSQA, 2008a).

Temperature was also recorded all over the sampling periods and taken into account to correct the uptake rate for concentration calculation.

The uncertainty linked to the temporal sampling strategy for formaldehyde was also studied during the first phase of this pilot monitoring survey. For 12 establishments over 160, formaldehyde measurements were performed for 16 weeks (4.5 days) consecutively, a part during the summer period, another part during the winter period and the rest during the transition period between the 2 seasons.

### Indoor air stuffiness

A good indicator of the air stuffiness is the measurement of carbon dioxide concentrations ($CO_2$) from metabolic production. $CO_2$ concentrations were thus monitored continuously with a time step of 10 minutes for two consecutive weeks using an instrument developed by CSTB and based on a non-dispersive infrared sensor (Lum'Air®) (Ribéron *et al*., 2011). An air stuffiness index called ICONE (*Indice de CONfinement d'air dans les Ecoles*) was also developed in 2008 by CSTB (Ribéron et al., 2011), that is based on the frequency and intensity of $CO_2$ levels around defined threshold values of 1000 and 1700 ppm, and taking exclusively into account children occupancy periods. A classroom is considered occupied when at least half of the usual number of children is present. The air stuffiness level of the room is then expressed by a score from 0 to 5. A score of 0 corresponds to non-stuffy air ($CO_2$ level always less than 1000 ppm) and is the most favorable situation. A score of 5 corresponds to air with extreme stuffiness ($CO_2$ level always above 1700 ppm during children occupancy) and is the worst situation. Middle scores correspond to a gradient of variable exceeding situations.

The final result for a given classroom is the average of two weekly scores rounded to the nearest integer and corresponding to one of the six categories of air stuffiness (0 to 5). This calculation method allows minimizing the influence of  under-occupancy or special events that can take place during the week. The air stuffiness index reflects the quality of the air change during occupancy but does not provide any information during unoccupied periods. The score for one building is represented by the highest value registered among instrumented rooms.

**Operational measurements progress and building audit**

As indicated previously, measurements were performed in the presence of children during one to two consecutive weeks (1 or 2 x 4.5 days), depending on the targeted compound, and at two different seasons, except for $CO_2$ that was measured only during the winter period (when heating systems were switched on). All measurement systems were placed on Monday morning, before 8:30am to avoid disturbing courses. Passive diffusion tubes were generally hanged from the ceiling. The measurement was stopped when children left the classroom, on Friday after 4:45pm. One to height rooms per establishment were investigated, depending on its size (number of buildings, number of floors and rooms per building ...). The studied rooms were the same for both seasons. For benzene specifically, an outdoor measurement was also performed in order to evaluate the outdoor contribution. Replicates and field blanks were also performed in each establishment for quality assurance purposes.

The building audit consisted in a set of questions that included a general description of the facility, housekeeping/cleaning premises, products used for this purpose, aeration habits and ventilation system, activities, etc. This questionnaire was filled by a building expert. Audits were performed during the heating season in order to check heating systems (from November 2009 to February 2010). These diagnoses did not include any measurement.

## 3 RESULTS

### Chemical indicators

The measured formaldehyde and benzene concentrations were compared with reference values suggested by the French committee for public health (HCSP 2009 and HCSP 2010). These values take into account health protection values determined by the French agency for food, environmental and occupational health safety, and technical considerations such as the existence of chemical substitutes or alternative techniques. The reference value for long-term exposure to formaldehyde is 30 $\mu g/m^3$ with remediation actions needed for any observed level above 100 $\mu g/m^3$ (HCSP, 2009). The reference value for long-term exposure to benzene is 5 $\mu g/m^3$ with remediation actions needed for any observed level above 10 $\mu g/m^3$ (HCSP, 2010).

Annual average concentrations of formaldehyde and benzene for each establishment (mean of the annual concentrations registered in the different rooms) are detailed in Table 1. 89% of the investigated establishments had an average concentration of formaldehyde below 30 $\mu g/m^3$. For benzene, 43 % of the establishments had an average concentration lower than 2 $\mu g/m^3$. These levels are satisfactory and did not imply specific actions. 75 % of the establishments with a benzene concentration ranging from 2 to 5 $\mu g/m^3$ reported a mean level lower than 3 $\mu g/m^3$. In 75 % of the cases for which benzene concentrations were higher than 2 $\mu g/m^3$, indoor and outdoor levels were statistically equivalent.

Table 1. Annual mean levels of formaldehyde (FA) and benzene (BE) (n=160 establishments)

| Average concentration of FA (µg/m$^3$) | Proportion of establishments (%) | Average concentration of BE (µg/m$^3$) | Proportion of establishments (%) |
|---|---|---|---|
| 0 - 30 | 89.4 | 0 - 2 | 44.7 |
| 30 - 50 | 8.8 | 2 - 5 | 52.8 |
| 50 - 100 | 1.8 | 5 - 10 | 2.5 |
| > 100 | 0.0 | > 10 | 0.0 |

The uncertainty associated with the temporal sampling strategy for formaldehyde annual mean calculation is reported in Table 2. As indicated before, for 12 establishments, formaldehyde measurements were performed for 16 working weeks consecutively, a part during the “summer” period (S), another part during the “winter” period (W) and the rest during the transition period between the 2 seasons (T). For some establishments, several rooms were investigated. Each 16 week measurement in a room was considered as a measurement series, so the studied sample corresponds to 21 measurement series. Each series can be divided into 3 stratums (S, W, T). It was supposed that the 16 week measurement was representative of a whole year with its seasonal variations and that the defined 3 stratums represented a complete partition of the year.

Table 2. Theoretical uncertainty in annual mean calculation due to the temporal sampling strategy based on 21 measurement series.

| Temporal sampling strategies | Average uncertainty (%) | Standard deviation (%) |
|---|---|---|
| The year is divided into 3 stratums (S, W, T)<br>1 measurement per period, *i.e.* 3 measurements/year | 21 | 7 |
| The year is divided into 2 stratums (S, W)<br>1 measurement per period, i.e. 2 stratums/year | 26 | 8 |
| The year is equal to 1 stratum,<br>3 measurements/year | 25 | 6 |
| The year is equal to 1 stratum,<br>2 measurements/year | 32 | 8 |
| The year is equal to 1 stratum, ,<br>1 measurement/year | 46 | 12 |

The theoretical uncertainty due to the temporal sampling strategy was calculated according to the sample survey theory and depended on the concentration variance within each temporal stratum and on the number of measurements drawn in each stratum (Tillé, 2001 ; LCSQA, 2008b). We can see that dividing the year in different stratums is efficient for reducing the uncertainty. Indeed, for the same number of measurements during a year, the uncertainty is lower when stratums are considered. The second line (table 2) corresponds to the sampling strategy followed in the survey and appears to be the best compromise between cost (number of measurements) and precision (calculated uncertainty).

**Indoor air stuffiness**

Results for the air stuffiness index are compiled in Table 3. 23% of the investigated establishments had at least one room with a very high air stuffiness index, such as 4 or 5 (2% of the day-care centres, 18 % of the kindergartens and 47 % of the elementary schools). Quite all 10 oversea sites at Reunion island did not show any sign of stuffy air.

Table 3. Distribution of the air stuffiness index (n=160 schools and day-care centres)

| Icone | Distribution of the maximum air stuffiness index determined in each establishment (%) | | | |
|---|---|---|---|---|
| | All<br>n = 160 | Day-care centres<br>n = 46 | Nursing schools<br>n = 61 | Primary schools<br>n = 53 |
| 0 | 9.3 | 15.2 | 11.5 | 1.9 |
| 1 | 14.4 | 23.9 | 16.4 | 3.8 |
| 2 | 18.8 | 23.9 | 21.3 | 11.3 |
| 3 | 33.8 | 34.8 | 31.2 | 35.8 |
| 4 | 22.5 | 2.2 | 18.0 | 45.3 |
| 5 | 0.6 | 0 | 0 | 1.9 |
| INV | 0.6 | 0 | 1.6 | 0 |

*[*]INV: Invalid data or insufficient occupancy that does not allow the calculation of the index.*

## 4 DISCUSSION

Formaldehyde and benzene concentrations as well as air stuffiness indexes were compared. To do so, the annual average concentrations for each establishment were categorized into three classes, according to the reference values. For formaldehyde, the three classes were 0-30 µg/m$^3$, 30-50 µg/m$^3$ and > 50 µg/m$^3$. For benzene, the three classes were 0-2 µg/m$^3$, 2-5 µg/m$^3$ and > 5 µg/m$^3$. For the air stuffiness index, values of 4 and 5 were grouped together, as the latter occurred only once.

Then, we calculated the distribution of air stuffiness indexes for each class of pollutant concentration and we studied the evolution of this distribution according to the classes of benzene or formaldehyde concentrations (Figure 1).

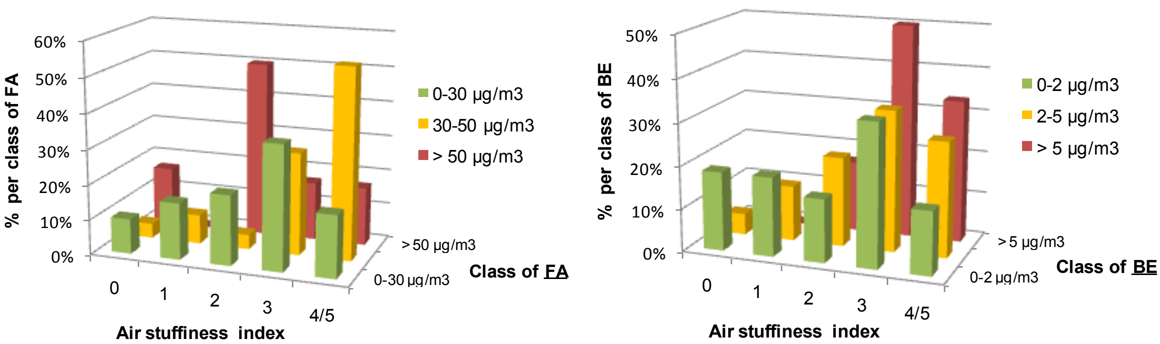


Figure 1. Distribution of air stuffiness indexes according to the different classes of formaldehyde (FA) (left) concentrations, or benzene (BE) (right) concentrations

The distribution of air stuffiness indexes did not seem to depend on the formaldehyde concentrations. For benzene, the proportion of establishments with levels higher than 5 µg/m$^3$ tended to be greater if the air stuffiness index was equal to or greater than 3. However, this tendency did not reflect a significant association between those 2 variables. Thus, a situation of high or very high air stuffiness was not sufficient to explain high pollutant concentration levels that also implied the presence of specific emission sources. Moreover, it is important to note that the comparison of air stuffiness indexes with benzene and formaldehyde levels must be made with caution since formaldehyde and benzene data included both occupied and unoccupied periods, especially nights (due to the limitations of the sampling method), whereas the air stuffiness index only took into account occupied periods. The occupied periods represent about 20-25 % of the 4.5 days sampling time. Another possible explanation is that ventilation conditions in classrooms may be different during occupied and unoccupied periods.

## 5 CONCLUSIONS

First phase results of this pilot monitoring survey showed that indoor air quality, based on the evaluation of two chemical indicators and an air stuffiness index, was satisfactory in 26 % of the investigated establishments (air stuffiness index below 3, annual concentration below 30 µg/m$^3$ for formaldehyde and below 2 µg/m$^3$ for benzene). However, 31% of the establishments registered a poor situation for at least one of the three parameters (air stuffiness index greater than 3 or annual concentration higher than 50 µg/m$^3$ for formaldehyde or higher than 5 µg/m$^3$ for benzene). These establishments were strongly encouraged to quickly conduct further investigations. Establishments with an air stuffiness index greater than 3 in at least one room were advised to improve their aeration during children occupancy and to check the ventilation system, where available.

Several actions planned in the second French NHAP should gradually improve air quality in schools and day-care centres in France, *i.e.* improving ventilation practices and reducing source emissions. In particular, as from January 1st, 2012, new building materials and decoration products are labelled according to their volatile organic compounds emissions. At last, another step towards a better indoor air quality in France is the statutory decree of December 2nd, 2011 that defines the mandatory monitoring of indoor air quality in some public premises hosting sensitive people. The guidelines of this mandatory monitoring are based on the study described in this paper.

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