French permanent survey on indoor air quality - Part 1.: Measurement protocols and quality control

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French Permanent Survey on Indoor Air Quality – Part. 1: Measurement Protocols and Quality Control

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Summary: *This paper focuses on a synthesis of the measurement and analytical protocols from the 2003-2005 campaign of the French permanent survey on indoor air quality and presents the associated quality control system including data traceability, quality code and interlaboratory tests. Preliminary exploitation of measurement and errors analysis are presented.*

Keywords: *pollutant measurements, field study, quality control*
Category: *Indoor air exposure*

1 Introduction

The French permanent survey on indoor air quality [1] conducts field measurement campaigns in different indoor environments, e.g. dwellings, offices, schools... Each campaign has a specific objective and as such needs specific development in survey strategy, questionnaires and measurement protocols. The first national campaign focused on dwellings was conducted from October 2003 to December 2005. The objectives of this campaign are first to provide target pollutant levels inside dwellings, second to identify sources and determinants of indoor air pollution and finally to assess occupants' exposure. A representative sample of 567 dwellings from the French national housing stock –approximately 24 million main homes– was randomly selected [2] and investigated. A pilot study based on 90 dwellings was first conducted to test measurement protocols [3] and the methodological approach used to assess population exposure [4].

Collected data quality is crucial in order to correctly assess population exposure and provide full data interpretation. Quality control was first initiated at the design of sampling protocols and choice of techniques and instrument. It was pursued with formation of technicians, who put designed protocols into practice and ensured correct transfer of monitored data to the database [5]. Associated laboratories conducted also daily quality control on samples storage, analysis and transfer of final results to the database. Finally, the database manager ensures quality control by tracking existing incoherencies and errors throughout the database. To summarize, quality control occurs before, during and after each measurement.

This paper focuses on a synthesis of the measurement and analytical protocols from the 2003-2005 campaign and presents the associated quality control system including data traceability, quality code and interlaboratory tests.

2 Methods

Thirty key pollutants were selected according to their health priority rank [6]. These pollutants are carbon monoxide (indoor and exhaled concentration), 20 target volatile organic compounds (benzene, ethylbenzene, m/p/o-xylenes, 1,2,4-trimethylbenzene, styrene, n-decane, n-undecane, trichlorethylene, tetrachloroethylene, 1,4-dichlorobenzene, 1-methoxy-2-propanol and acetate, 2-butoxyethanol and acetate, formaldehyde, acrolein, acetaldehyde, hexaldehyde), allergens (from dogs *Can f 1*, from cats *Fel d 1*, from dust mites *Der f 1* and *Der p 1*), radon and gamma radiation, particulate matter (PM_{10} and $PM_{2.5}$). Comfort parameters (relative humidity, temperature, carbon dioxide, exhaust airflow rate) are also measured. Different questionnaires provide characteristic data from investigated sites (building environment, furniture and equipments, rooms' description...), occupant's description, time activities diaries and allergic and respiratory symptoms. Questionnaires are fully detailed in an associated article [7].

A quality control system was set up for field sampling strategy and laboratory analysis. Technicians followed these protocols throughout the campaign and provided a quality code for every measurement. Quality codes are also provided by the laboratories and by the database administrator whenever errors or incoherencies are detected. There is one valid quality code for numerous non-conformity codes. This valid code means that sampling, transport/transfer and analysis are correct. However, some quality codes mean benign protocol deviation and data can therefore be considered useful for future analysis.

3 Measurement protocols

The measurement protocols of the 30 key indoor pollutants are presented hereafter, along with comfort parameters measurements.

Allergens

Cat and dog allergens (respectively *Fel d 1* and *Can f 1*) were measured in the living room by collecting suspended particulate matter on 37 mm diameter glass microfibre filters (Millipore). The measurement was realized during one hour at 20 litres per minute on three different filters (triplicates). Analyses were conducted at Hôpitaux Universitaires de Strasbourg (HUS) following the immuno-enzymatic ELISA method, a non amplified sandwich method using specific monoclonal antibodies. Limits of detection were 0.18 ng m^{-3} (*Fel d 1*) and 1.02 ng m^{-3} (*Can f 1*). Extended uncertainty for the sampling was determined to be ± 111% (*Fel d 1*) and ± 75% (*Can f 1*). Analytical extended uncertainty was much lower, about ± 26% (*Fel d 1*) and ± 21% (*Can f 1*).

Dust mite allergens (*Der f 1* and *Der p 1*) were measured in the collected dust by a vacuum cleaner from the bedroom mattress belonging to the reference occupant. Vacuum cleaner bags were then sent to Hôpitaux Universitaires de Strasbourg (HUS) for analysis, following the immuno-enzymatic ELISA method [8]. Limits of detection were 15.8 ng g^{-1} (*Der f 1*) and 26.4 ng g^{-1} (*Der p 1*). Analytical extended uncertainty was about ± 29% (*Der f 1*) and ± 25% (*Der p 1*).

Carbon dioxide (CO_2)

Carbon dioxide was measured to provide information on confinement and air renewal [9]. Carbon dioxide (along with temperature and relative humidity) was monitored during 7 days (10 min averages) by non dispersive infra-red probe (Q-track, TSI Inc.) in the bedroom. The instrument was verified and calibrated before investigation. Extended uncertainty was calculated to be ± 67 ppm at 1500 ppm target concentration.

Instruments were also yearly calibrated on temperature and relative humidity. These parameters were monitored in the living room as well using Hygrolog sensors (Rotronic).

Carbon monoxide (CO)

Carbon monoxide was monitored during 7 days (5 min averages) by electrochemical sensor (Draeger Pac III) in the living room, outdoors and in each room holding combustion equipment (gas heater, portable heater, etc.). All instruments were verified and calibrated before every investigation. Instrument resolution was 1 ppm although values between -3 and 3 ppm (noise fluctuation) were assumed to be not significantly different from 0. CO profiles presenting negative values lower than -3 were discarded as this indicated a drift in the electrochemical sensor response. Extended uncertainty was found to be ± 4.9 ppm for a target concentration of 50 ppm. The choice of instrument was oriented towards a security system able to warn occupants in case of high concentration of CO that may represent a danger.

Carbon monoxide was also measured in exhaled air of voluntary occupants at least 6 years old. The measurement was performed by means of a FIM CO-Tester Tx. This is an additional measurement added at the request of the Institut de Veille Sanitaire (InVS) to provide population exposure data to environmental tobacco smoke and other sources of carbon monoxide.

Exhaust air flow rate

Exhaust air flow rates were measured in every humid room (kitchen, bathroom, WC) where exhaust openings are present. An array of hot wires (SwemaFlow 233) provides an instantaneous measurement of exhaust air flow rate recorded on a PDA by the technician.

Particulate matter ($PM_{2.5}$ and PM_{10})

Mass concentrations of suspended particulate matter with an aerodynamic diameter below 2.5 µm ($PM_{2.5}$) and below 10 µm (PM_{10}) were measured in the living room. The chosen instrument was a model 2100 Mini-Partisol air sampler (Rüpprecht & Patashnick Co., Inc., distributed by Ecomesure), coupled to a ChemPass model 3400 sampling system integrating both $PM_{2.5}$ and PM_{10} PEMS impactor systems operating at 1.8 L min^{-1}. Technicians used flowrate calibrator DryCal DC-Lite (Bios International) in the field to check the correct flowrate in both PEMS impactors. The instrument was programmed to sample air during defined occupation hours of the investigation week, i.e. in the evening from 5 pm to 8 am the next day (Monday to Friday) and every time in the week-end. Pre-weighted 37 mm diameter PTFE membranes (2 µm porosity, Gelman Sciences) were used to collect particulate matter and then returned to the Laboratoire d'Hygiène de la Ville de Paris (LHVP), conducting the gravimetric measurement using a 1 µm sensitive electronic balance. Blank filters were left in the field to provide effective detection limit of the method.

Radon ^{222}Rn and gamma radiation

At the request of the Institut de Radioprotection et de Sûreté Nucléaire (IRSN), Radon and gamma radiation measurements were added to the survey. Passive measurement of Radon volumic activity is performed by accumulating alpha radiation from

^{222}Rn and its descendants (^{218}Po, ^{214}Po) on a 12 µm cellulose nitrate film (Kodalpha dosimeter) during 2 months. Both bedroom and living room are instrumented. Dosimeters are then sent to Dosirad, the laboratory in charge of the analysis.

External gamma radiation dose rate of cosmic and telluric origin is measured through a gamma radiameter of the Geiger-Müller type (Saphymo 6150 AD6), selecting energies between 60 keV and 1.2 MeV. The measurement stated in µSv h^{-1} is performed in the living room during 3 to 4 hours.

Volatile organic compounds and aldehydes

Volatile organic compounds (VOC) and aldehydes were collected by radial diffusive sampling [10] onto carbograph 4 adsorbents and 2,4-DNPH coated Florisil respectively. Both bedroom and outdoors are instrumented in each investigated dwelling. After 7 days exposure, adsorbents are sealed and sent to the laboratories in charge of analysis. Two different laboratories (CSTB and Fondazione Salvatore Maugeri (FSM)) perform the identification and quantification of VOC target compounds. Only one undertakes aldehyde cartridges analysis. Adsorbed VOCs were extracted through thermodesorption and analyzed by gas phase chromatography equipped with flame ionization detector and/or mass spectrometry [11]. Aldehyde-hydrazones formed in the cartridge were eluted by acetonitrile solvent and analyzed by liquid chromatography associated with a UV detector [12]. Detection limits were provided by both laboratories and are presented in Table 1.

Table 1. Analytical detection limits of VOCs and aldehydes expressed for a 7 days exposure.

	Detection limit (µg.m^{-3})	
	CSTB	FSM
Benzene	0.4	0.1
2-Butoxyethanol	0.4	0.2
2-Butoxyethyl acetate	0.3	0.6
n-Decane	0.06	0.2
1,4-Dichlorobenzene	0.03	0.2
Ethylbenzene	0.3	0.1
1-Methoxy-2-propanol	0.5	0.2
1-Methoxy-2-propyl acetate	0.7	0.2
Styrene	0.1	0.1
Tetrachloroethylene	0.4	0.02
Toluene	0.4	0.1
Trichloroethylene	0.4	0.3
1,2,4-Trimethylbenzene	0.02	0.1
n-Undecane	0.5	0.4
(m+p)-Xylenes	0.5	0.1
o-Xylene	0.2	0.1
Acetaldehyde	0.3	--
Acrolein	0.1	--
Formaldehyde	0.6	--
Hexaldehyde	0.1	--

Extended uncertainties were determined for some VOCs at 7 days exposure in indoor environment [13] and range from ± 20% (5 µg m^{-3} benzene or 19 µg m^{-3} toluene) to ± 27% (8 µg m^{-3} m/p-xylene). Another determination of measurement uncertainty of benzene leads to a value of ± 28% for a concentration of 2 µg m^{-3} closer to usual indoor values.

4 Quality codes

A quality code was associated with every sampling and analysis of a target pollutant. This code allows the validation or reject of collected data to integrate in subsequent statistical analyses. The technician fills all quality codes relevant to sampling, data transfer and questionnaires. The laboratory fills the part relevant to analyses.

The following quality codes were used throughout the survey campaign (Table 2 and 3). They can also be regrouped in different error types:

- protocol error: 1160, 1200, 2110, 2120, 2130, 2140, 2150, 2160, 2170, 2220, 3140, 3500;
- sampling error: 1000, 1330, 3600;
- laboratory error: 3000, 3130, 3210, 3220, 3400, 3710, 3720, 3730, 3740;
- instrument error: 1120, 1131, 1133, 1134, 1135, 3110, 3120;
- transport error: 2210, 4000, 4100;
- other sources of error: 1110, 1136, 1140, 1150, 3300.

Table 2. Quality codes for sampling.

Quality code	description
60000	valid sampling
1000	sampling not valid
1110	sampling not executable
1120	sampling instrument defect
1131	battery problem
1133	sampling instrument breakdown
1134	data retrieval problem
1135	empty data file
1136	insufficient supply
1140	sampling not performed
1150	occupant refusal
1160	other protocol condition not respected
1200	instrument calibration not valid
1330	manipulation error during sampling
2110	exposure time too low
2120	exposure time too high
2130	sampled volume too low
2140	sampled volume too high
2150	measurement not required (additional)
2160	flowrate too low
2170	flowrate too high
2210	invalid reception control
2220	storage temperature not respected

Table 3. Quality codes for analysis.

Quality code	description
0	valid analysis
3000	analysis not valid
3110	analytical instrument breakdown
3120	analytical instrument defect
3130	calibration error
3140	other protocol condition not respected
3210	manipulation error during analysis
3220	other laboratory problem
3300	sample not analysed
3400	sample not exploitable
3500	storage time too long
3600	missing information about sampling
3710	extrapolated data
3720	insufficient resolution
3730	extrapolated and insufficient resolution
3740	co-eluted target compound
4000	sample not received by the laboratory
4100	Donnée non reçue à l'équipe centrale

5 VOC inter-laboratory results

Interlaboratory comparison has been conducted for the analytical determination of target VOC concentration. Fifteen cartridges containing carbograph 4 adsorbents were spiked with a mix of 13 target VOCs at a given concentration level by an independent laboratory (Ecole des Mines de Douai). Five cartridges were sent to each laboratory for analysis along with blanks. Three different concentration levels were tested (around 5, 20 and 60 µg m^{-3}). Results are presented in Fig. 1.

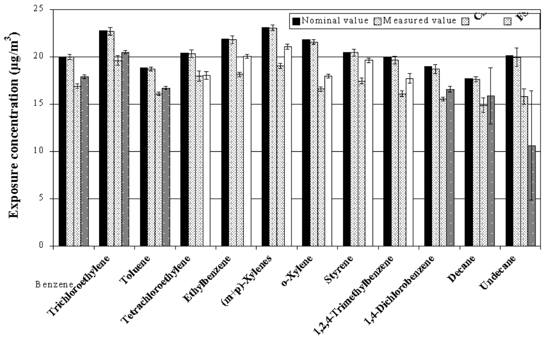

Fig. 1. VOC interlaboratory comparison results at intermediate concentration level. Error bars represent standard deviation.

Unexpected delay in the transmission of spiked tubes to the laboratory hinders the evaluation of the performance of a given laboratory as regards nominal value. However, laboratory comparison remains possible. Apart from alkanes, analytical differences between laboratories remain low and acceptable even if they are significant for some VOCs. The same conclusion is obtained at low or high concentration level.

The influence of storage conditions (temperature CQ = 2220 and duration CQ = 3500) on concentration of sampled VOCs and aldehydes was assessed (Fig. 2 and Fig. 3 respectively). A longer storage time (2 months instead of less than 1 month) has a significant impact on some VOC concentration: ethylbenzene (-15%), xylenes (-18%), 2-butoxyethanol (+75%), 1,2,4-trimethylbenzene (-16%), 1,4-dichlorobenzene (-26%), n-decane (-10%), n-undecane (-13%). No significant impact of storage temperature was observed.

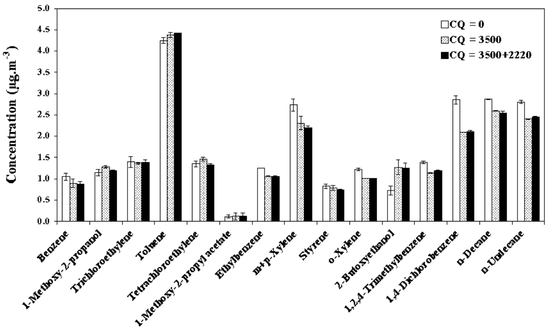

Fig. 2. Incidence of storage time (2 months vs < 1 month) and storage temperature (< 4°C vs ambient conditions) on concentration of sampled VOCs.

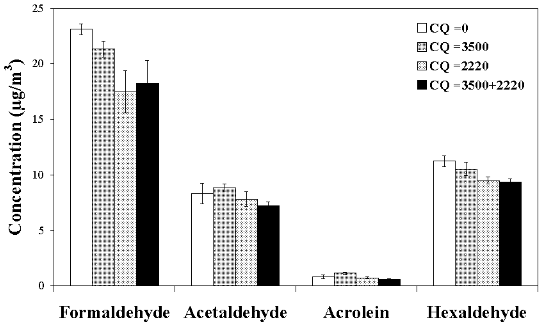

Fig. 3. Incidence of storage time (1 month vs < 15 days) and storage temperature (< 4°C vs ambient conditions) on concentration of sampled aldehydes.

Formaldehyde (-24%) and hexaldehyde (-16%) are the most sensitive to a storage temperature conditions and at a lesser extent to an increase in storage duration resulting in a respective loss of 8% and 6%. Data associated with 2220 quality code are then discarded from the final matrix.

Replicates were also used in the investigated dwellings to account for sampling and transport influence. Six samples per sampling point (indoor and outdoor) and per laboratory were installed in 12 different dwellings. Two examples (m+p)-xylenes and 1-methoxy-2-propanol are presented in Fig. 4. Results are acceptable for almost all compounds. Relative differences are more important in the low concentration range (< 2µg m^{-3}), but error in the absolute value remains acceptable. However, a serious deviation is observed for undecane at concentration over 20 µg m^{-3}.

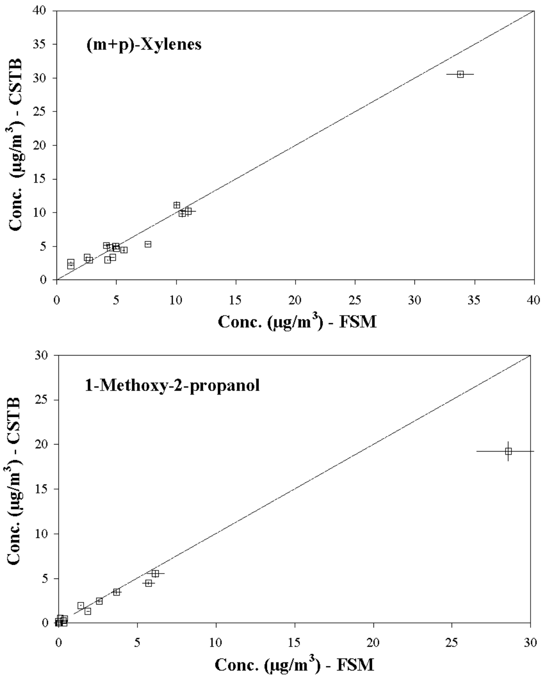

Fig. 4. Inter-comparison of VOC field replicates. Error bars represent laboratory standard deviation.

6 Distribution of quality codes

Analysis of quality codes distribution allows the identification of error sources associated with each pollutant measurement. This analysis was performed on all 567 investigated dwellings, but some analytical results are still not available at the writing of this paper. Nevertheless, this represents an enormous amount of data from single point measurements to data files covering a week measurement every 5 or 10 minutes.

The majority of quality codes associated with protocol deviation and with laboratory errors have been retained, as it was found that some quality codes do not represent sufficient information to decide whether or not a collected data can be exploited. Moreover, the use of quality codes was sometimes misinterpreted by technicians. As such, little confidence can be put in the raw completion of quality codes. Automated logical cross-validation rules are used in order to definitely keep or reject a given data. Nonetheless, the amount of valid data expressed by the quality codes is presented in table 4 for some key pollutants. The values herein must be taken as an upper margin. Current cross-validation rules will slightly lower the values. However, this will particularly affect particulate matter measurement as both $PM_{2.5}$ and PM_{10} are collected with a single mini-Partisol instrument and quality codes are not precise enough to decide the validity of data. Therefore, the following rules have been applied:

- PEMS initial flow rate tolerance: 1.8 ± 0.2 L/min. All values outside this range are rejected.
- Minimum sampling time of 5 days.
- Maximum difference between initial and final flow rate of 0.2 L/min.

The major identified source of error for both sampling and analysis is presented in table 5 for each pollutant. The inability to perform the sampling appears as the major error for aldehydes, carbon monoxide and VOC. This underlines the difficulty encountered by some technicians to realize the outdoor measurement. This is particularly true in block of flats without balconies and easy set up of samples outside the flat is impeded. The 1160 quality code (other protocol condition not respected) is mainly associated with cat and dog allergen measurement. This represents the technician inability to realize the sample in the absence of any pet which positively interacts with the measurement. Instrument fault also account for both carbon dioxide and particulate matter sampling errors, and in VOC analysis. Lack of fresh temperature is the main error source for aldehyde analysis. Damaged filters represent the main inability to perform the weighing of collected particles. Finally, the analysis of dust mite allergens needs a minimum dust mass that is sometimes not achieved.

Table 4. Estimation (upper margin) of valid rate for the sampling and analyses of pollutants based on quality codes.

	%valid sampling	%valid analyses
Aldehydes	99.4%	92.6%
Carbon dioxide	93.4%	--
Carbon monoxide (environmental)	94.0%	--
Cat and dog allergens	96.0%	97.3%
VOC	99.8%	93.7%
Dust mite allergens	96.7%	77.1%
Particulate matter	84.4%	97.1%

Table 5. Major identified source of error (occurrence).

	Main error code	
	sampling	analysis
Aldehydes	1110 (0.3%)	2220 (4.9%)
Carbon dioxide	1133 (1.9%)	--
Carbon monoxide (environmental)	1110 (1.3%)	--
Cat and dog allergens	1160 (2.2%)	1160 (0.7%)
VOC	1110 (0.1%)	3120 (2.4%)
Dust mite allergens	1150 (2.1%)	3400 (19.8%)
Particulate matter	1120 (3.8%)	2210 (1.6%)

Conclusion

The first campaign of the French national survey has investigated 567 dwellings from autumn 2003 to winter 2005. An enormous amount of data was collected and, associated to it, quality control data. Before beginning to analyze key data, quality control information must be exploited to define the final data set. Inter-laboratory comparison and designed experiments were undertaken to provide the highest confidence level in the final data set. The use of quality codes self-completed by technicians represents a good estimate of data validity but remains insufficient to be used as a decision tool. This role is assumed by logical cross-validation rules that are still being applied to the database. The sampling and analysis valid rate is satisfactory. Thus, a correct representativeness of the sample is preserved. And representativeness is essential in order to provide data interpretation and conclusions at a population level.

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