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Assessment of ventilation and indoor air pollutants in nursery and elementary schools in France

Abstract The aim of this study was to characterize the relationship between Indoor Air Quality (IAQ) and ventilation in French classrooms. Various parameters were measured over one school week, including volatile organic compounds, aldehydes, particulate matter ($PM_{2.5}$ mass concentration and number concentration), carbon dioxide (CO_2), air temperature, and relative humidity in 51 classrooms at 17 schools. The ventilation was characterized by several indicators, such as the air exchange rate, ventilation rate (VR), and air stuffiness index (ICONE), that are linked to indoor CO_2 concentration. The influences of the season (heating or non-heating), type of school (nursery or elementary), and ventilation on the IAQ were studied. Based on the minimum value of 4.2 l/s per person required by the French legislation for mechanically ventilated classrooms, 91% of the classrooms had insufficient ventilation. The VR was significantly higher in mechanically ventilated classrooms compared with naturally ventilated rooms. The correlations between IAQ and ventilation vary according to the location of the primary source of each pollutant (outdoor *vs*. indoor), and for an indoor source, whether it is associated with occupant activity or continuous emission.

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Key words: Air stuffiness index; CO_2; Volatile organic compounds; Aldehydes; Particulate matter.

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Practical Implications
Different ventilation indicators can be calculated based on CO_2 concentration, including air exchange rate (AER), ventilation rate (VR), and air stuffiness index. Each of them provides different information on ventilation conditions in classrooms. Ventilation must be improved in schools. Intraschool variability of air pollutant concentrations depends on the location of the pollutant primary source. Intraschool variability of ventilation conditions may not be negligible.

Introduction

Children constitute a population that is susceptible to exposure to air pollutants, not only because their respiratory and immune systems are not fully developed but also because they breathe higher air volumes in relation to their body weights (WHO, 2005). In this context, the Indoor Air Quality (IAQ) of school environments has become a growing concern within the scientific community; there is evidence that connects poor IAQ to negative impacts on students' health, performance, and attendance (Daisey et al., 2003; Mendell and Heath, 2005).

Classroom IAQ comprises a wide range of parameters, and although international guidelines have not been defined for classroom IAQ, an international effort has begun to characterize these microenvironments (Annesi-Maesano et al., 2013; Chatzidiakou et al., 2012). Several studies have monitored indoor air concentrations of gaseous compounds (Stranger et al., 2007), particles (Branis et al., 2005; Fromme et al., 2007, 2008; Tran et al., 2012, 2014), semi-volatile organic compounds (Lim et al., 2014; Wu et al., 2010), and allergens (Salo et al., 2009). Other studies have investigated outdoor air contributions (Almeida et al., 2011; Blondeau et al., 2005; Madureira et al., 2012), the role of different ventilation strategies (Geelen et al., 2008; Guo et al., 2008; Rosbach et al., 2013; Santamouris et al., 2008) and the relationships with student health (Daisey et al., 2003; Mendell et al., 2013; Simoni et al., 2011) and performance (Bakó-Biró et al., 2012; Haverinen-Shaughnessy et al., 2011; Shendell et al., 2004; Twardella et al., 2012; Wargocki and Wyon, 2007). Few studies have reported correlations between ventilation and IAQ in schools. Godwin and Batterman (2007) showed negative correlations between AER and indoor concentrations of toluene, m,p-xylenes, alpha-pinene, and limonene in a set of 64 schools in Michigan in the United States [aldehydes and particulate matter (PM) were not measured]. More recently, Chatzidiakou et al. (2015) studied the relationship between CO_2, ventilation rates (VRs), and selected pollutants in 18 classrooms from six schools in London. In France, some studies have been conducted in schools but only in limited numbers (Annesi-Maesano et al., 2012; Blondeau et al., 2005; Poupard et al., 2005; Tran et al., 2014) or only for a limited number of indoor air pollutants (Michelot et al., 2013).

Because of the lack of knowledge in this field in France, the French Indoor Air Quality Observatory (OQAI) was commissioned to assess children's exposure to various indoor air pollutants in nursery and elementary schools. A study was conducted in 51 classrooms in 17 French nursery and elementary schools. The specific objectives of the study were to (i) characterize IAQ of the studied classrooms using a multipollutant approach, (ii) characterize ventilation through different indicators, and (iii) study the relationships between indoor air concentrations and ventilation conditions, including the types of ventilation systems.

Materials and methods

Study site and school descriptions

The study area was the town of Clermont-Ferrand and the surrounding area, which has a population of 139 860 inhabitants in 43 km^2, and is located in the region of Auvergne in the center of France, which is 350 km south of Paris. A total of 17 schools were chosen on a voluntary basis to take part in this study and included 7 nursery and 10 elementary schools. Three classrooms were studied per school. The locations of the studied schools are shown in Figure 1. All schools were in urban areas except for school 7, which was located in a rural area.

Schools 1–10 were evaluated in the heating season (from January 11, 2010 to April 2, 2010, where the outdoor mean values of temperature and relative humidity were 6.4 ± 5.7°C and 59 ± 11%, respectively), and schools 11–17 were evaluated in the non-heating season (from April 26, 2010 to June 25, 2010, where the outdoor mean values of temperature and relative humidity were 17.0 ± 3.8°C and 60 ± 10%, respectively). Each school was studied during one full school week, from Monday to Friday. Details regarding the indoor values of temperature and relative humidity in the classrooms are presented in the Supporting Information (Section S1.2.).

The studied classrooms (a total of 51) had volumes ranging between 90 and 310 m^3. The majority of the classrooms (63%, n = 32) were located on the ground floor, 35% (n = 18) were located on the first floor, and only one classroom was located on the second floor. The mean number of children per classroom was 24 ± 4, and their ages ranged between 3 and 10 years old.

Regarding the type of ventilation, 73% of the classrooms (n = 37) had natural ventilation (windows and door openings), and 27% (n = 14) were equipped with a mechanical ventilation system (two classrooms had balanced systems, and 12 classrooms had exhaust-only systems).

The majority of classrooms (80%) were located in buildings of a traditional construction type (solid stone walls, solid bricks, or concrete blocks), 18% were located in buildings built using a concrete frame with stud walls, and only one classroom was located in a prefabricated building. All classrooms had their inner walls painted. Detailed information regarding each studied classroom is available in Table S1.

Sample collection and analytical methods

The sample collection and measurements were conducted in each studied classroom over one school week, from Monday morning (8:00 am) until Friday afternoon (5:00 pm).

Indoor concentrations of carbon dioxide (CO_2), relative humidity (RH), and temperature were measured continuously every 10 min with a Q-Trak Plus IAQ monitor 8552 (TSI Incorporated, Shoreview, MN, USA). The CO_2 sensor uses non-dispersive infrared technology with a measuring range of 0–5000 ppm [accuracy: ±(3% of reading + 50 ppm)]. The CO_2 sensors were calibrated prior to and after each sampling week at two concentrations (0 and 1500 ppm) with standard gas (Lind Gaz, Montereau, France). Relative humidity was measured with a capacitive sensor (5–95% RH measuring range and ±3% accuracy). The temperature was measured with a thermistor (0–50°C operating range and ±0.6°C accuracy). RH represents the ratio of the water vapor pressure to the saturation vapor pressure. The

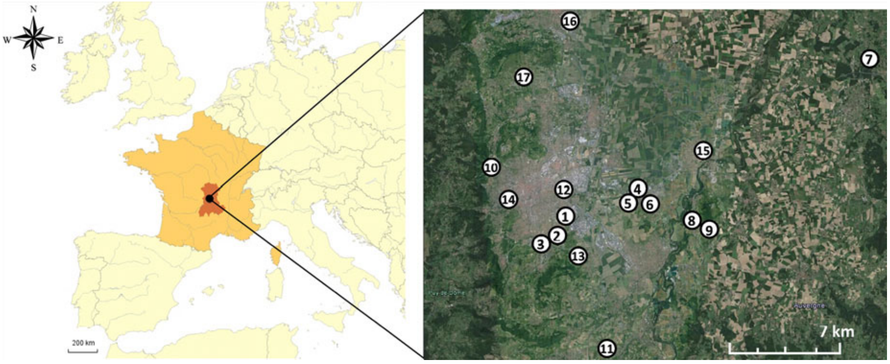

Fig. 1 Location of the Auvergne region (dark orange) in central France (left) and the 17 studied schools in the Clermont-Ferrand area (right)

latter depends directly on temperature; therefore, RH is dependent on both temperature and water content of the air. To consider this, the specific humidity (SH) was used rather than RH (calculation details provided in the Supporting Information).

The outdoor RH and temperature were recorded every 10 min with a Hygrolog data logger (Rotronic, Bassersdorf, Switzerland). The operating ranges for the RH and temperature sensors were 0–100% RH (±1.5% accuracy) and −10 to 50°C (±0.4°C accuracy), respectively.

Particulate matter ($PM_{2.5}$) concentrations were measured in all of the studied classrooms with gravimetric MicroVol samplers (Europa Environmental, Twyning, England) that were programmed to perform the sampling between 8:00 am and 5:00 pm each weekday, except for Wednesdays, which were days off for most of the French schools. The sampling flow rate was 1.8 ± 0.1 l/min, which was checked before and after sampling with a volumetric piston flow meter (Bios Dry Cal, DC Lyte, CO, USA). Particles were collected on a pre-weighted 37-mm PTFE filters with a 2-μm pore size (Pall Corporation, Port Washington, WI, USA). The total mass of particles was determined with a microbalance (1-μg resolution, Mettler MT-5; Sartorius, Dourdan, France) according to a standardized method (European Committee for Standardization, 2014; EN 12341). The quantification limit for the $PM_{2.5}$ weight was 22 μg.

Continuous measurements of particles with diameters between 0.3 and 20 μm were performed from Monday to Friday in 25 classrooms using an optical particle counter (Dust monitor 1.108; Grimm Aerosol Technik, Ainrin, Germany). The flow rate was 1.2 l/min. Measurements were recorded every 10 min. The device has a measuring range of 1–2000 particles/cm^3 and an accuracy of 2%. The device has 15 measuring channels with different size resolutions, namely 0.30–0.40, 0.40–0.50, 0.50–0.65, 0.65–0.80, 0.80–1.0, 1.0–1.6, 1.6–2.0, 2.0–3.0, 3.0–4.0, 4.0–5.0, 5.0–7.5, 7.5–10.0, 10.0–15.0, 15.0–20.0, and >20.0 μm. The final results are expressed in particles with sizes within the ranges 0.3–1.0 and 1.0–20.0 μm by summing the results of the measuring channels within the size ranges.

The sampling of aldehydes and VOCs was performed with passive samplers (Radiello, Fondazione Salvator Maugeri, Padova, Italy) that were exposed from Monday morning (8:00 am) to Friday afternoon (5:00 pm) in the classrooms and outdoors. Aldehydes were sampled by diffusion on cartridges impregnated with 2,4-dinitrophenylhydrazine (2,4-DNPH)-coated Florisil. DNPH reacts with carbonyls to form corresponding stable 2,4-DNPhydrazone derivatives. DNPhydrazones were extracted in acetonitrile, and four aldehydes (formaldehyde, acetaldehyde, butyraldehyde, and hexaldehyde) were analyzed by high-performance liquid chromatography and ultraviolet detection at a wavelength of 360 nm (Waters Corporation, Guyancourt, France). Volatile organic compounds were sampled by adsorption on graphitized charcoal tubes (Carbograph 4, Radiello, Fondazione Salvator Maugeri). The VOC samples were thermally desorbed (ATD 400; Perkin Elmer, Villepinte, France), and nine VOCs (benzene, ethylbenzene, toluene, m,p-xylenes, o-xylene, styrene, tetrachloroethylene, trichloroethylene, and 1,4-dichlorobenzene) were quantified by gas chromatography coupled with a flame ionization detector and a mass spectrometer (Saturn 200; Agilent, Les Ulis, France). Passive tube sampling rates were determined by Fondazione Salvator Maugeri (Italy) and were adjusted based on the average temperature during the sampling week using the equations provided by the manufacturer. For benzene, the sampling rate was adjusted according to the equation provided by Pennequin-Cardinal et al. (2005). The limits of detection (LOD) and limits of quantification (LOQ) for each compound are presented in Table 1. A field blank for $PM_{2.5}$, aldehyde, and VOC sampling was performed for each school to check for contamination during transport and manipulation.

Questionnaires

Two questionnaires were designed and administered to gather specific information about each studied school.

Table 1 Limits of detection (LOD) and limits of quantification (LOQ) of aldehydes and volatile organic compounds (VOCs)

Compounds	LOD (μg/m^3)	LOQ (μg/m^3)
Aldehydes		
Formaldehyde	0.6	1.1
Acetaldehyde	0.3	0.4
Butyraldehyde	0.05	0.2
Hexaldehyde	0.1	0.2
VOCs		
Benzene	0.4	1.1
Ethylbenzene	0.4	1.3
Toluene	0.3	0.9
m,p-xylenes	0.5	1.5
o-xylene	0.2	0.6
Styrene	0.1	0.3
Tetrachloroethylene	0.4	1.2
Trichloroethylene	0.4	1
1,4-dichlorobenzene	0.4	1.2

The technical characteristics of the school building were provided by the director of each school. The teacher described the type of activities and the number of classroom occupants with a frequency of 30 min for each day of the sampling week.

Air exchange rates and VRs

Air exchange rates (AERs) (air change per hour, /h) and VRs (air liters per second per person, l/s per person) were calculated for all classrooms using the build-up method, which relies on a computerized tool developed by Hänninen (2013) based on CO_2 concentrations. The AERs and VRs were calculated considering only the occupied periods. The number of students and teachers present during each analyzed build-up event was assessed through the questionnaires. When information was missing from the questionnaire, a theoretical number of students and teachers in the classroom was used.

Night-time AERs (nAERs) were calculated from monitored CO_2 concentrations using the decay method described in Ramalho et al. (2013). This method, which was developed in SAS software version 9.1.3 (Brie-Comte-Robert, France), automatically detects and calculates the AER for each decrease sequence observed after 7:00 pm, assuming no metabolic production of CO_2, a constant outdoor concentration of 400 ppm, and a homogeneous distribution of the indoor concentration. The method automatically performs a linear regression on the log-transformed concentration difference between the indoor and outdoor concentrations for each decrease event. The slope of the regression corresponds with the value of the AER during the event. Linear regressions of less than 1 h and with a determination coefficient of less than 0.9 were discarded. The value determined for each classroom represents the time-weighted average of all valid nAERs calculated over the school week.

The weekly averaged AER (wAER) was calculated by the weighted average of the AER and nAER based on a cumulative occupied period of 32 h and on an unoccupied/night period of 72 h, respectively, for a total of 104 h in the school week.

Air stuffiness index

Ribéron et al. (2011) developed an air stuffiness index, called ICONE (*Indice de CONfinement d'air dans les Ecoles*), which is a communication tool for occupants. The ICONE index is used to evaluate air stuffiness during occupied periods. The index considers the frequency and intensity of CO_2 concentrations compared with the defined threshold values of 1000 and 1700 ppm. These values were chosen to frame the threshold value of 1300 ppm required in France by the Departmental Health Regulations (RSDT, 1978). CO_2 concentrations measured during children's normal classroom attendance over a complete school week are used to calculate the ICONE index. Normal classroom attendance is defined when at least half of the usual number of children are present. Children occupancy periods associated with normal attendance ranged from 4 to 24.5 h during the week, with a mean value of 14.6 ± 5.3 h. Occupancy periods of less than 5 h are discarded. Subsequently, CO_2 values are classified according to their levels: n_0—values < 1000 ppm, n_1—values between 1000 and 1700 ppm and n_2—values > 1700 ppm. The ICONE air stuffiness index is then calculated by applying Equation 1, where f_1 is the proportion of CO_2 values between 1000 and 1700 ppm ($f_1 = n_1/(n_0 + n_1 + n_2)$) and f_2 is the proportion of CO_2 values above 1700 ppm ($f_2 = n_2/(n_0 + n_1 + n_2)$).

$$\mathrm{ICONE} = \left(\frac{2.5}{\log_{10}(2)}\right)\log_{10}(1 + f_1 + 3f_2) \quad (1)$$

The final results for characterizing a given classroom are rounded to the nearest integer. The air stuffiness level of the room is then expressed by a score, which ranges from 0 to 5, where 0 corresponds to non-stuffy air (the CO_2 concentration is always below 1000 ppm, which represents the most favorable conditions), and 5 corresponds to extremely stuffy air (the CO_2 concentration is always above 1700 ppm, which represents the worst conditions). ICONE scores between 0 and 5 correspond to an air stuffiness gradient: 0—none, 1—low, 2—average, 3—high, 4—very high, and 5—extreme air stuffiness (Ramalho et al., 2013). As such, the ICONE score represents a method of assessing occupants' exposure to air stuffiness. The higher the score, the higher the risk of exposure to elevated concentrations of other pollutants when specific pollutant sources are present or activated. In 2012, the ICONE air stuffiness index was integrated into the framework for the mandatory monitoring of IAQ in public buildings in France (MEDDTL, 2012).

Statistical treatment

An analysis of variance of the results was performed using nonparametric statistics at a significance level of 0.05. The Mann–Whitney *U*-test was used for binary independent groups, and the Kruskal–Wallis test was used for multiple independent groups. Correlations between variables were expressed by Spearman correlation coefficients because the continuous data distribution was neither normal nor log-normal. For analysis purposes, values below the LOD were replaced by LOD/2, and values below the LOQ were replaced by (LOD + LOQ)/2. All statistical analyses were conducted using the XLSTAT 2014 software (Addinsoft, Paris, France).

Results and discussion

Table 2 presents the mean values of all the studied parameters in the studied classrooms according to the sampling season, the type of classroom, and the type of ventilation, along with their statistical differences. Descriptions of the indoor climate parameters are available in the Supporting Information.

Ventilation indicators

The CO_2 concentration data were analyzed by different means to provide six different ventilation indicators, including (i) the indoor CO_2 concentration distribution, (ii) AERs, (iii) nAERs, (iv) wAERs, (v) VRs, and (vi) ICONE, the air stuffiness index.

CO_2 concentrations. Table 3 shows the overall results of the CO_2 concentration measurements for the studied classrooms during the period of occupancy. The mean CO_2 concentration in the 50 studied classrooms was 1290 ppm during the occupied period, with a median of 1250 ppm and a maximum of 2220 ppm.

A number of standards and guidelines for CO_2 concentration have been established for school environments. The European Standard EN15251 (European Committee for Standardization, 2007) and REHVA Guidebook 13 (d'Ambrosio Alfano et al., 2010) proposed performance-based standards that limit CO_2 concentration to 1500 ppm during a full school day. The American Society of Heating, Refrigeration, and Air Conditioning Engineers (ASHRAE) recommendations (US) specify an upper CO_2 limit of 700 ppm above the outdoor concentration for occupied classrooms, which generally corresponds to 1000–1100 ppm based on an average outdoor concentration between 300 and 400 ppm (NOAA, 2014). The French Departmental Health Regulations (RSDT, 1978) mandate that concentrations do not exceed 1300 ppm at any time in rooms in which smoking is prohibited.

Overall, the studied classrooms presented CO_2 concentration above 1000, 1300, and 1500 ppm during 65%, 46%, and 35% of the occupied period, respectively.

Table 2 Mean values ($\pm$standard deviation) of the indoor air quality parameters assessed in the classrooms according to the sampling season, type of school and type of ventilation. Bold values indicate significant *P*-values (Mann-Whitney *U* test)

		Season			Type of school			Type of ventilation		
Pollutant/parameter	All	Heating	Non-heating	*P*-value	Nursery	Elementary	*P*-value	Mechanical	Natural	*P*-value
Mean CO_2 (ppm)	1300 $\pm$ 400	1200 $\pm$ 400	1400 $\pm$ 400	0.168	1200 $\pm$ 400	1300 $\pm$ 400	0.140	1000 $\pm$ 200	1400 $\pm$ 400	**0.016**
AER (per hour)	1.4 $\pm$ 0.6	1.5 $\pm$ 0.7	1.2 $\pm$ 0.4	0.065	1.4 $\pm$ 0.6	1.3 $\pm$ 0.6	0.397	1.8 $\pm$ 0.5	1.2 $\pm$ 0.6	**0.001**
nAER (per hour)	0.2 $\pm$ 0.1	0.2 $\pm$ 0.1	0.1 $\pm$ 0.05	**0.015**	0.1 $\pm$ 0.1	0.2 $\pm$ 0.1	**0.032**	0.2 $\pm$ 0.1	0.2 $\pm$ 0.1	0.340
wAER (per hour)	0.5 $\pm$ 0.2	0.6 $\pm$ 0.2	0.4 $\pm$ 0.1	0.066	0.5 $\pm$ 0.1	0.5 $\pm$ 0.3	0.586	0.7 $\pm$ 0.1	0.5 $\pm$ 0.2	**0.024**
VR (l/s per person)	2.9 $\pm$ 1.6	3.3 $\pm$ 1.8	2.4 $\pm$ 1.0	0.119	3.2 $\pm$ 1.8	2.7 $\pm$ 1.6	0.213	4.2 $\pm$ 1.7	2.4 $\pm$ 1.4	**<0.001**
ICONE > 3 (%)	29	20	41	0.136	25	31	0.688	0	36	**0.032**
Temperature (°C)	23.0 $\pm$ 1.5	22.5 $\pm$ 1.5	23.6 $\pm$ 1.3	**0.012**	22.5 $\pm$ 1.3	23.3 $\pm$ 1.6	0.141	22.3 $\pm$ 1.1	23.3 $\pm$ 1.6	**0.032**
Spec. humidity (g/kg)	6.5 $\pm$ 1.9	5.3 $\pm$ 1.0	8.3 $\pm$ 1.4	**<0.001**	6.4 $\pm$ 2.2	6.6 $\pm$ 1.7	0.870	5.8 $\pm$ 1.8	6.8 $\pm$ 1.9	0.220
Formaldehyde ($\mu g/m^3$)	25 $\pm$ 15	19 $\pm$ 9	31 $\pm$ 17	**0.003**	28 $\pm$ 17	23 $\pm$ 13	0.346	18 $\pm$ 7	28 $\pm$ 16	**0.042**
Acetaldehyde ($\mu g/m^3$)	6.3 $\pm$ 2.1	5.5 $\pm$ 1.9	7.1 $\pm$ 2.1	**0.010**	6.5 $\pm$ 2.4	6.1 $\pm$ 2.0	0.634	4.8 $\pm$ 1.6	6.9 $\pm$ 2.1	**0.002**
Butyraldehyde ($\mu g/m^3$)	14 $\pm$ 9	10 $\pm$ 4	18 $\pm$ 11	**<0.001**	17 $\pm$ 12	12 $\pm$ 5	**0.049**	12 $\pm$ 3	15 $\pm$ 11	0.642
Hexaldehyde ($\mu g/m^3$)	12 $\pm$ 7	7.0 $\pm$ 4.7	17 $\pm$ 7	**<0.001**	13 $\pm$ 7	11 $\pm$ 8	0.287	7.6 $\pm$ 4.5	13 $\pm$ 8	**0.020**
Benzene ($\mu g/m^3$)	2.1 $\pm$ 2.2	2.8 $\pm$ 2.6	1.0 $\pm$ 0.3	**<0.001**	1.3 $\pm$ 0.4	2.2 $\pm$ 2.2	0.537	1.5 $\pm$ 0.3	2.2 $\pm$ 2.4	0.392
Toluene ($\mu g/m^3$)	5.2 $\pm$ 5.1	6.1 $\pm$ 6.4	3.8 $\pm$ 1.7	0.781	3.4 $\pm$ 1.5	5.0 $\pm$ 5.3	0.287	2.4 $\pm$ 0.7	5.8 $\pm$ 5.5	**0.008**
Ethylbenzene ($\mu g/m^3$)	2.2 $\pm$ 1.3	2.4 $\pm$ 1.5	1.9 $\pm$ 0.9	0.272	4.8 $\pm$ 0.7	2.1 $\pm$ 1.3	0.906	1.5 $\pm$ 0.2	2.4 $\pm$ 1.4	0.130
m,p-xylenes ($\mu g/m^3$)	4.4 $\pm$ 3.5	4.6 $\pm$ 3.5	4.1 $\pm$ 3.7	0.968	3.7 $\pm$ 3.0	3.9 $\pm$ 2.9	0.118	1.9 $\pm$ 0.2	5.0 $\pm$ 3.7	**0.001**
o-xylene ($\mu g/m^3$)	1.6 $\pm$ 2.1	1.5 $\pm$ 1.7	1.8 $\pm$ 2.5	0.412	1.2 $\pm$ 1.9	1.2 $\pm$ 1.4	0.180	0.3 $\pm$ 0.2	1.9 $\pm$ 2.2	**0.001**
Styrene ($\mu g/m^3$)	1.5 $\pm$ 0.7	1.6 $\pm$ 0.9	1.2 $\pm$ 0.2	0.250	1.2 $\pm$ 0.2	1.5 $\pm$ 0.7	0.420	1.2 $\pm$ 0.2	1.5 $\pm$ 0.8	0.439
$PM_{2.5}$ ($\mu g/m^3$)	22 $\pm$ 8	23 $\pm$ 9	21 $\pm$ 7	0.395	21 $\pm$ 9	23 $\pm$ 7	0.230	19 $\pm$ 6	24 $\pm$ 8	0.178
$PN_{[0.3-1]}$ (particles/cm^3)	53 $\pm$ 32	61 $\pm$ 38	44 $\pm$ 21	0.218	63 $\pm$ 37	46 $\pm$ 27	0.218	61 $\pm$ 36	51 $\pm$ 32	0.476
$PN_{(1-20)}$ (particles/cm^3)	19 $\pm$ 31	19 $\pm$ 26	20 $\pm$ 38	0.565	24 $\pm$ 38	15 $\pm$ 25	0.603	33 $\pm$ 45	16 $\pm$ 27	0.325

AER, air exchange rate; nAER, night-time AER; wAER, weekly averaged AER; VR, ventilation rate; PM, particulate matter; PN, Particle number concentration.

Table 3 Distribution of the CO_2 concentrations (ppm) during the occupied period over the sampling week (n = 50 classrooms)

CO_2 concentration (ppm)	Mean	Min	P5	P25	Median	P75	P95	Max
Weekly mean	1290	530	730	970	1250	1670	1900	2220
Minimum	440	360	380	410	430	460	570	600
Maximum	2440	580	1080	1880	2320	3180	3890	4310

Air exchange rates. Figure 2 presents the AERs for the studied classrooms during 459 analyzed CO_2 build-up events. A mean of 9 build-up events was analyzed per classroom, and each event had a mean duration of 43 min.

The mean AER during the occupied period varied between 0.3 and 3.1/h among all studied classrooms, with a mean value of 1.4 ± 0.6/h. The night AER varied between 0.03 and 0.54/h among all studied classrooms, with a mean value of 0.16 ± 0.13/h. The wAER varied between 0.20 and 1.0/h among all studied classrooms, with a mean value of 0.52 ± 0.22/h.

Ventilation rates. Figure 3 presents the VRs for the studied classrooms, which were determined from the AER calculations described above. The VRs varied between 0.6 and 8.2 l/s per person among all studied classrooms, with a mean value of 2.9 ± 1.6 l/s per person. These VRs are within ranges of values previously reported in the literature (Canha et al., 2013; Sundell et al., 2011).

Global standards and guidelines related to minimum classroom VRs are inconsistent. For instance, in the US, ASHRAE recommends a minimum VR of 7 l/s per person (ASHRAE, 2007) for teaching facilities. In Europe, the European Standard EN15251 (2007) and REHVA Guidebook 13 (2010) specify a minimum VR of 3 l/s per person in all occupied teaching and learning spaces. However, European countries have specific VR guidelines that vary substantially, from 4.2 to 12 l/s per person (Brelih and Seppänen, 2011). For instance, in France, the minimum recommended VR ranges between 4.2 and 5.0 l/s per person for classrooms equipped with mechanical ventilation (Ramalho et al., 2005; RSDT, 1978), depending on the type of activity being performed and on the numbers and ages of the children.

From the 51 studied classrooms, 37% (n = 19) presented mean VRs above 3 l/s per person (EN15251/REHVA guideline), and 9% (n = 9) presented mean VRs above 4.2 l/s per person, which is the minimum requirement by the French guidelines.

Among the 14 classrooms that were equipped with a mechanical ventilation system, 79% (n = 11) showed mean VRs above 3 l/s per person, and 43% (n = 6) presented mean VRs above 4.2 l/s per person. Among the 37 naturally ventilated classrooms, only 22% (n = 8) presented mean VRs higher than 3 l/s per person, and only 11% (n = 4) presented mean rates higher than 4.2 l/s per person.

In nursery classrooms, 43% (9 of 21) had mean VRs higher than 3 l/s per person, and 19% (4 of 21) had mean rates higher than 4.2 l/s per person. For elementary classrooms, 33% (10 of 30) had mean VRs higher than 3 l/s per person, and 20% (6 of 30) had mean rates higher than 4.2 l/s per person.

Air stuffiness index. Figure 4 shows the ICONE scores for the 42 classrooms for children occupancy periods greater than 5 h.

No extreme air stuffiness (ICONE score = 5) was found in any of the classrooms. Twelve classrooms (of 42; 28.5%) had very high air stuffiness (ICONE score = 4), 12 (28.5%) had high air stuffiness (ICONE score = 3), 10 (24%) had average air stuffiness

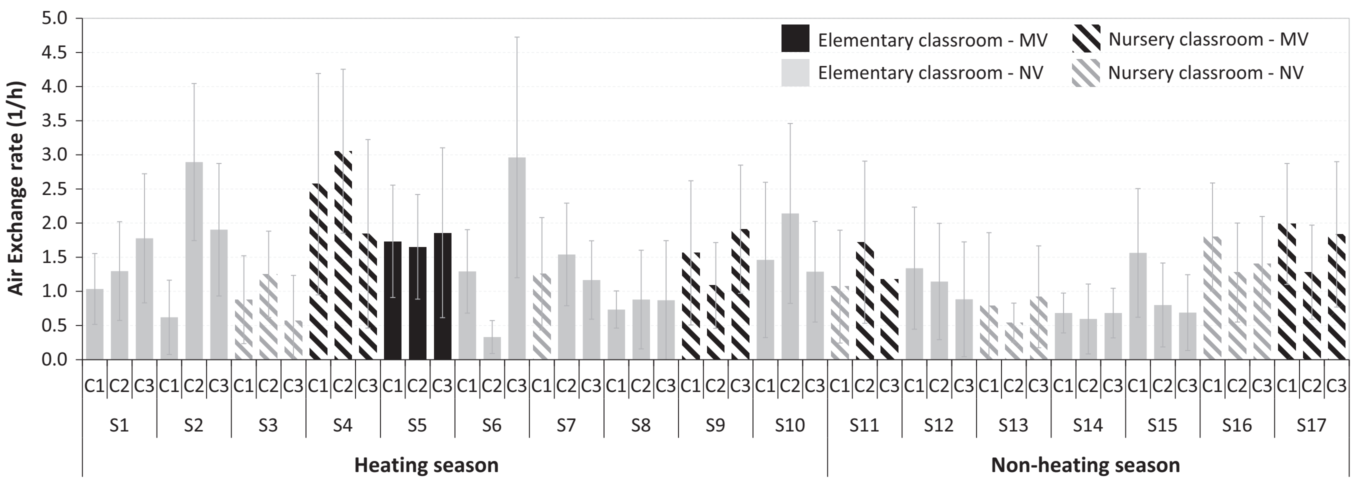

Fig. 2 Air exchange rates (per hour) of the studied classrooms (C) at the 17 schools (S) during the occupied period. MV, mechanically ventilated; NV, naturally ventilated

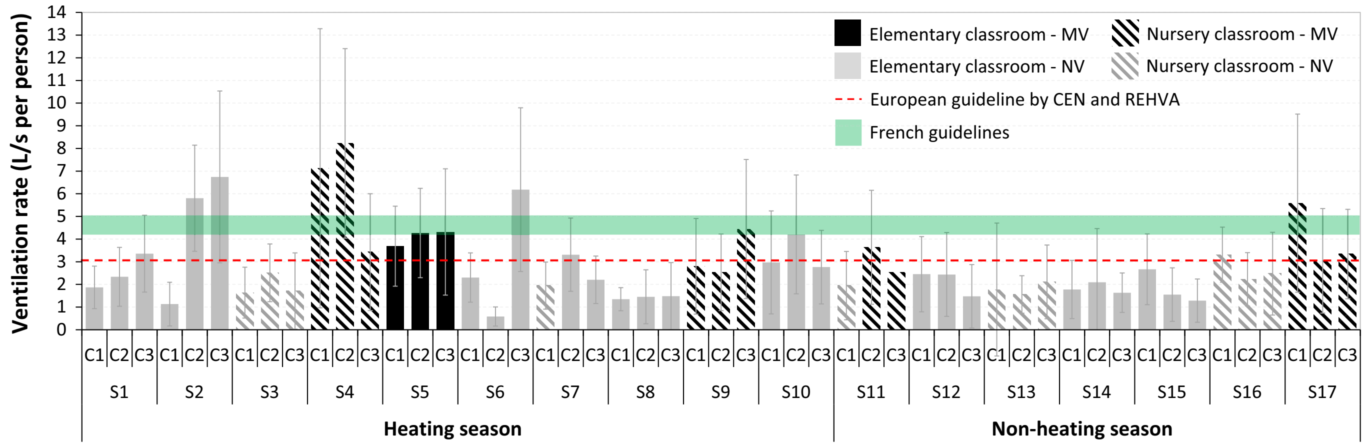

Fig. 3 Ventilation rates (l/s per person) of the studied classrooms (C) at the 17 schools (S) during the occupied period. MV, mechanically ventilated; NV, naturally ventilated

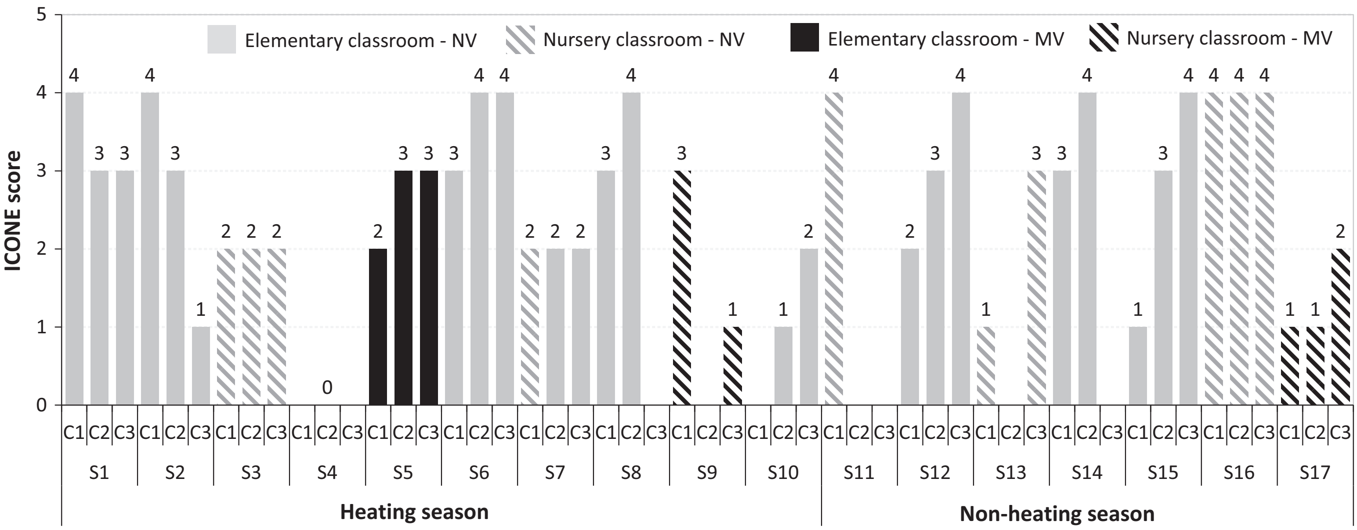

Fig. 4 ICONE air stuffiness index scores in the studied classrooms (C) at the 17 schools (S) during the occupied period. MV, mechanically ventilated; NV, naturally ventilated

(ICONE score = 2), and only 8 (19%) had no or little air stuffiness (ICONE score = 0 or 1).

Figure 5 shows the ICONE profiles in this study compared with the profiles that were calculated in 310 schools and day care centers distributed throughout France (Ramalho et al., 2013). The ICONE profiles obtained in the present study are similar to those described previously for both types of schools. Elementary schools have higher percentages of high air stuffiness (ICONE score ≥ 3) (present study: 69%; previous study: 67%) compared with that of nursery schools (present study: 38%; previous study: 29%).

Influence of various parameters on ventilation indicators. As summarized in Table 2, apart from nAER, all the studied ventilation indicators presented significant differences between classrooms with different types of ventilation, with mechanical ventilation showing higher VRs and AERs along with lower mean CO_2 concentrations during occupancy and ICONE scores, compared with natural ventilation. Mechanically ventilated classrooms presented VRs that were almost two times higher than naturally ventilated classrooms, as previously observed in other international studies (Chatzidiakou et al., 2012; Santamouris et al., 2008). No difference in nAER was found between naturally and mechanically ventilated classrooms. This can be explained by the fact that mechanical ventilation system are generally turned off during the night. As such, nAER represents mostly outdoor air infiltration. The nAER was significantly higher in elementary schools compared to nursery schools, which could indicate that nursery schools have tighter envelopes than elementary schools. This difference may be associated with the construction date of the building, whereby the nursery schools were constructed (median date: 1977) more

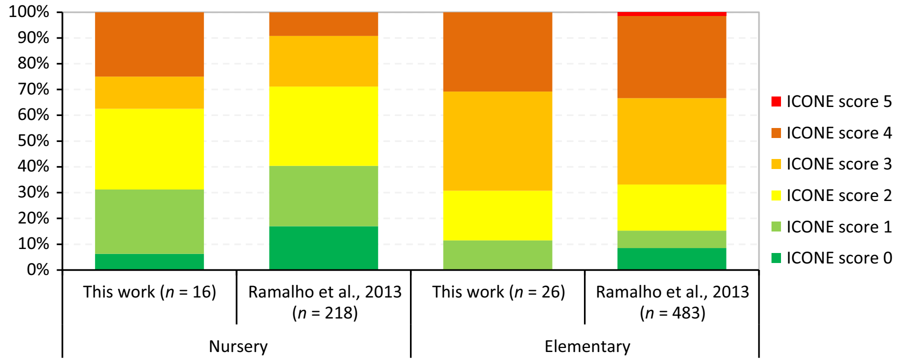

Fig. 5 ICONE profiles by type of school

recently on average than elementary schools (median date: 1967). The nAER was also found to be higher during the heating season compared to the non-heating season, which may be explained by greater stack effect during winter.

Gaseous pollutants

Table 4 summarizes the concentrations of VOCs and aldehydes measured in the classrooms and outdoors during a full school week, which includes both occupied and non-occupied periods.

Volatile organic compounds. The mean concentrations were always higher indoors compared to outdoors for all of the VOCs. For chlorinated hydrocarbons, both indoor and outdoor concentrations were at least 60% of the time, below the LOD. Because trichloroethylene, tetrachloroethylene, and 1,4-dichlorobenzene were measured at concentrations that were less than 30% above the limit of quantification, they were excluded from the statistical analysis. Among the aromatic hydrocarbons, toluene, ethylbenzene, m,p-xylenes, and styrene were detected and quantified in all classrooms. Benzene was detected in all samples but quantified in only 68% of them. No significant differences in VOC concentrations were found between the types of classroom (all P-values > 0.050 with the Mann–Whitney U-test), as shown in Table 2. Only benzene presented significantly higher concentrations in the heating season compared with the non-heating season ($P < 0.001$). Concentrations of toluene, m,p-xylenes, and o-xylene were significantly lower in mechanically ventilated classrooms compared with naturally ventilated classrooms (P-values < 0.050). The I/O ratios found for the BTEX compounds (benzene, toluene, ethylbenzene, and xylenes) were slightly above one, which is similar to those described by Stranger et al. (2008) in Flemish schools.

Aldehydes. Formaldehyde, acetaldehyde, and butyraldehyde were detected and measured in all 51 classrooms, but hexaldehyde was only detected in 90% of the classrooms. All of the aldehydes showed higher concentrations indoors compared to outdoors. Significantly higher concentrations were measured during the non-heating season for all of the aldehydes ($P < 0.050$; Table 2). This finding can be attributed to higher indoor temperatures and specific humidities, which enhance the emission of pollutants from materials and furniture (Sarigiannis et al., 2011). Only butyraldehyde showed significant differences in the mean concentration between classroom types, with higher values in the nursery compared to elementary schools. Compared to naturally ventilated classrooms, mechanically ventilated classrooms had significantly lower mean concentrations of formaldehyde, acetaldehyde, and hexaldehyde (P-values < 0.050). The indoor/outdoor ratio was always >8 for all aldehydes, which indicated that all of these pollutants likely had common indoor sources and/or shared a common outdoor sink, for example, photolysis and photochemistry.

Particulate matter

$PM_{2.5}$ mass concentrations. Among the 51 studied classrooms, $PM_{2.5}$ concentrations could not be assessed in six classrooms because of instrument malfunction (four classrooms) or because the sampling flow rate was outside of the acceptable range (two classrooms). Figure 6 shows the $PM_{2.5}$ concentrations in 45 classrooms. The mean indoor $PM_{2.5}$ concentration was 22 ± 8 $\mu g/m^3$, with values ranging from 10 to 47 $\mu g/m^3$. The mean $PM_{2.5}$ concentration of 23 ± 9 $\mu g/m^3$ and 21 ± 7 $\mu g/m^3$ was measured during the heating and non-heating seasons, respectively. No significant difference was observed according to the sampling season, type of classroom, or type of ventilation ($P > 0.050$).

All classrooms had indoor concentrations above 10 $\mu g/m^3$, which is the limit established by the WHO for $PM_{2.5}$ in ambient air over long exposure periods (annual mean) (WHO, 2006). The WHO also established a limit of 25 $\mu g/m^3$ per 24 h, which was exceeded in 31% of the classrooms in this study.

The indoor $PM_{2.5}$ concentrations assessed in this study are similar to concentrations previously observed

Table 4 Concentrations of major volatile organic compounds and aldehydes in classrooms and outdoors ($\mu g/m^3$; $n = 51$)

Pollutants ($\mu g/m^3$)	≤LOD %	LOD–LOQ %	≥LOQ %	Mean ± s.d.	Min	P5	P25	P50	P75	P95	Max	IAQ guideline	Mean I/O ratio ± s.d.
Benzene													
Classrooms	0	32	68	2.1 ± 2.2	<LOQ	<LOQ	<LOQ	1.4	1.6	7.7	8.5	1.7 $\mu g/m^3$ [a,b]	1.3 ± 0.3
Outdoors	0	53	47	1.7 ± 1.9	<LOQ	<LOQ	<LOQ	<LOQ	1.5	6.7	6.7	ALARA [c]	
Toluene													
Classrooms	0	0	100	5.2 ± 5.1	1.7	1.9	2.1	3.2	5.2	17.1	24.4	15 mg/m^3 [c,d]	2.1 ± 1.1
Outdoors	0	27	73	2.9 ± 3.9	<LOQ	<LOQ	<LOQ	1.7	2.3	15.4	15.4	300 $\mu g/m^3$ [c,e]	
Ethylbenzene													
Classrooms	0	0	100	2.2 ± 1.3	1.2	1.2	1.3	1.7	2.6	5.0	6.0	–	2.0 ± 2.2
Outdoors	0	80	20	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	3.2	3.2		
m,p-xylenes													
Classrooms	0	0	100	4.4 ± 3.5	1.6	1.7	1.9	2.8	7.1	11.0	14.9	20 mg/m^3 [c,d]	1.8 ± 1.5
Outdoors	0	27	73	2.5	<LOQ	<LOQ	<LOQ	1.7	2.3	7.8	7.8	200 $\mu g/m^3$ [c,e]	
o-xylene													
Classrooms	20	27	54	1.6 ± 2.1	<LOD	<LOQ	<LOQ	0.7	1.8	5.7	8.9	20 mg/m^3 [c,d]	1.8 ± 1.0
Outdoors	20	67	13	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	<LOQ	3.2	3.2	200 $\mu g/m^3$ [c,e]	
Styrene													
Classrooms	0	0	100	1.5 ± 0.7	0.9	1.0	1.1	1.3	1.4	3.3	4.0	2 mg/m^3 [c,d]	2.1 ± 2.1
Outdoors	0	67	33	0.4	<LOQ	<LOQ	<LOQ	<LOQ	0.8	1.1	1.1	250 $\mu g/m^3$ [c,e]	
Tetrachloroethylene													
Classrooms	74	12	14	1.1 ± 2.3	<LOD	<LOQ	<LOD	<LOD	<LOQ	5.4	11.5	250 $\mu g/m^3$ [a,e]	n/a
Outdoors	53	47	0	<LOQ	<LOD	<LOQ	<LOD	<LOD	<LOQ	<LOQ	<LOQ		
Trichloroethylene													
Classrooms	73	12	15	2.3 ± 6.4	<LOD	<LOD	<LOD	<LOD	<LOQ	20.8	28.2	23 $\mu g/m^3$ [a,b]	1.5 ± 0.4
Outdoors	60	33	7	1.3	<LOD	<LOD	<LOD	<LOD	<LOQ	14.9	14.9		
1,4-dichlorobenzene													
Classrooms	39	32	29	1.8 ± 3.9	<LOD	<LOD	<LOD	<LOQ	3	9.4	9.9	–	n/a
Outdoors	93	7	0	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOQ		
Formaldehyde													
Classrooms	0	0	100	25.1 ± 14.8	6.8	10.4	15.7	19.2	30.7	62.4	66.2	100 $\mu g/m^3$ [a,d]	20 ± 4
Outdoors	6	6	88	1.8	<LOD	<LOD	1.6	1.9	2.1	2.6	2.6	30 $\mu g/m^3$ [c,d]	
Acetaldehyde													
Classrooms	0	0	100	6.3 ± 2.1	2.7	4.3	4.3	6.1	8.1	10.2	10.7	200 $\mu g/m^3$ [c,e]	8.2 ± 1.9
Outdoors	6	0	94	1.3	<LOD	<LOQ	1.1	1.3	1.7	1.9	1.9		
Butyraldehyde													
Classrooms	0	0	100	14.1 ± 9.1	2.9	4.9	10.3	12.3	14.9	31.3	53.0	–	59 ± 1
Outdoors	18	0	82	3.6	<LOD	<LOD	2.3	4.2	5.3	6.1	6.1		
Hexaldehyde													
Classrooms	10	0	90	11.7 ± 7.4	<LOQ	<LOQ	6.3	10.9	15.2	27.0	29.1	–	54 ± 1
Outdoors	29	18	53	0.4	<LOD	<LOD	<LOQ	0.3	0.5	1.4	1.4		

LOD, limit of detection; LOQ, limit of quantification; s.d., standard deviation; P, percentiles; ALARA, as low as reasonably achievable.
[a]World Health Organization (2010).
[b]Lifetime exposure with acceptable risk at 10^{-5}.
[c]INDEX project (Koistinen et al., 2008; Kotzias et al., 2005).
[d]Short-term exposure.
[e]Long-term exposure.

in 24 schools in the Netherlands (mean of 23 ± 6 $\mu g/m^3$, Janssen et al., 2001), 108 schools in France (mean of 20 $\mu g/m^3$, Annesi-Maesano et al., 2012), and 64 schools in Germany (mean of 22 $\mu g/m^3$, Fromme et al., 2007). A small number of studies reported lower mean values compared to those in this study, including studies in Sweden (mean of 8 $\mu g/m^3$, Molnár et al., 2007), the US (mean of 17 ± 14 $\mu g/m^3$, John et al., 2007), and Portugal (mean of 10 $\mu g/m^3$, Almeida et al., 2011). Other studies in the literature have reported higher values compared to those in this work (Annesi-Maesano et al., 2013). For example, a mean concentration of 44 $\mu g/m^3$ was measured in schools in Beijing, China (Liu et al., 2004). In several studies, $PM_{2.5}$ concentrations were ten times greater than the present values, including a study in India in the winter (Habil and Taneja, 2011).

Particle number concentrations. Figure 7 shows the median particle number concentration measured in 25 classrooms based on occupancy. Concentrations were higher during occupied periods for both types of particles. Nevertheless, the difference between the occupied and non-occupied periods was greater for particles with diameters of 1–20 μm than for those with diameters of 0.3–1 μm. Overall, a mean of 53 ± 32 particles/cm^3

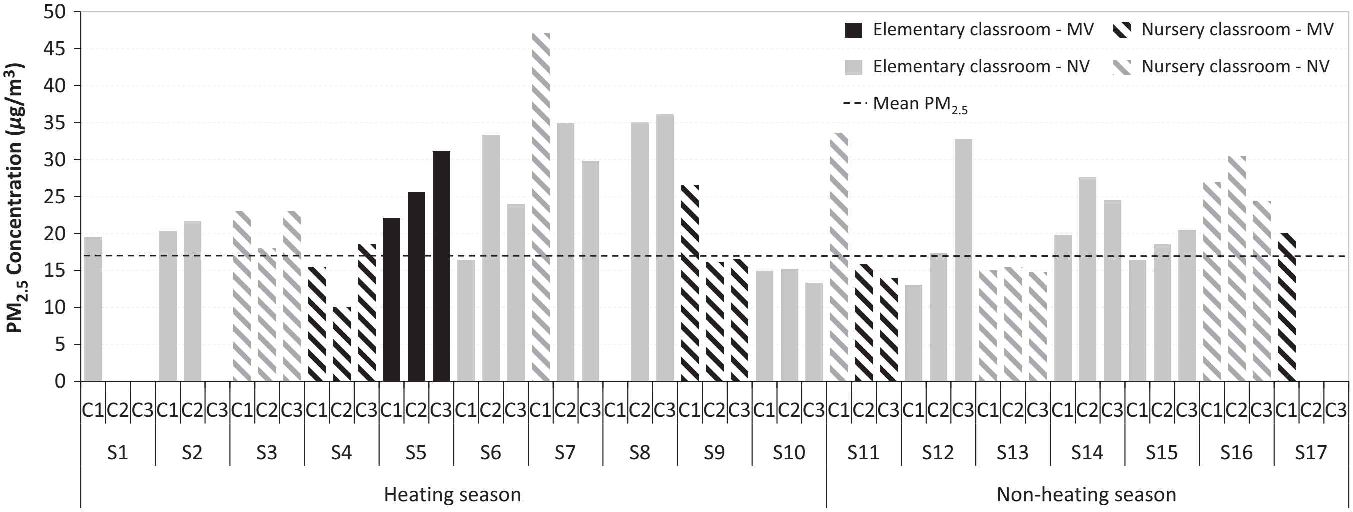

Fig. 6 Particulate matter ($PM_{2.5}$) concentrations ($\mu g/m^3$) in the studied classrooms (C) at the 17 schools (S). MV, mechanically ventilated; NV, naturally ventilated

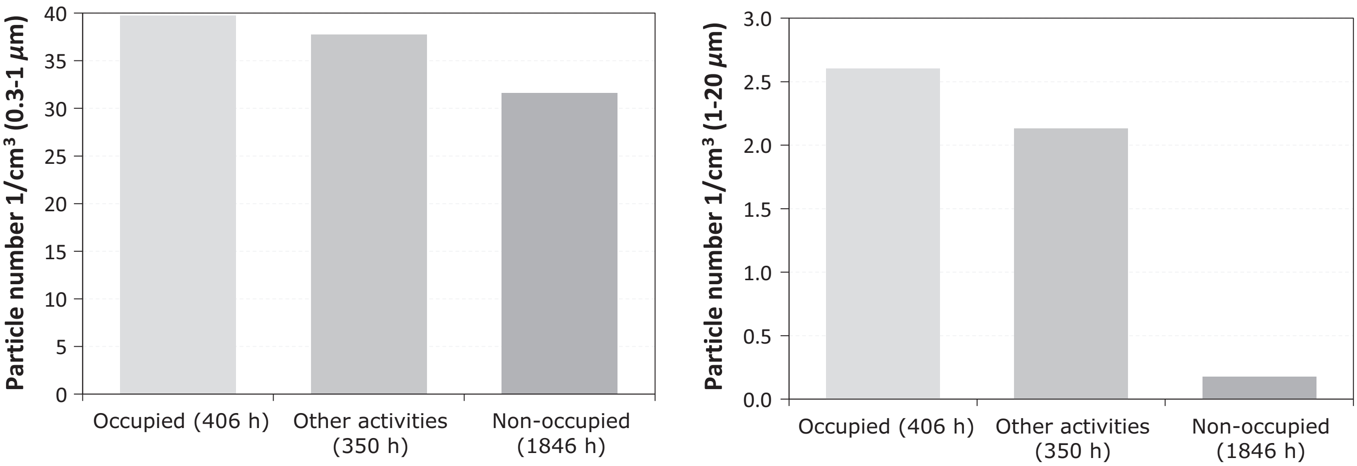

Fig. 7 Median concentrations (number of particles per cm^3) during the different sampling periods for particles with aerodynamic diameters of 0.3–1 μm (left) and 1–20 μm (right) during the different time periods

(range: 19–144 particles/cm^3) was measured for particles with a diameter between 0.3 and 1 μm, whereas for particles with a diameter between 1 and 20 μm, a lower mean value, namely 19 ± 31 particles/cm^3 (range: 1–105 particles/cm^3) was observed.

These results are similar to those observed by Blondeau et al. (2005) in eight French classrooms. Large particles presented higher concentrations during occupied periods compared with non-occupied periods, and this difference between periods was higher than for fine particles, the concentrations of which did not exhibit any important variation between types of sampling periods. This pattern is associated with the indoor generation of large particles by the occupants themselves, which can occur during school activities that create blackboard dust or resuspension of settled particles (Blondeau et al., 2005; Canha et al., 2014).

The comparison of fine and large particle concentrations based on the sampling season, type of classroom, and type of ventilation is shown in Table 2. For both types of particles, the mean concentrations were not significantly different ($P > 0.050$) in any of the studied cases.

Relationships between ventilation indicators and indoor air parameters

Table 5 presents the Spearman correlations between the ventilation indicators and the studied indoor air parameters. The Spearman correlations during the heating and non-heating seasons are presented in Tables S4 and S5, respectively.

Relationships between indoor air parameters. Strong correlation coefficients above 0.5 with P-values below 0.001 were found between all aldehydes and both the

Table 5 Spearman correlations between ventilation indicators and indoor air pollutants (significant correlations in bold)

Variables	CO_2	AER	nAER	wAER	VR	ICONE	Indoor T	Indoor SH	1	2	3	4	5	6	7	8	9	10	11	12	13
CO_2	1	**−0.34***	−0.20	−0.29	**−0.46****	**0.99*****	0.01	0.30	**0.47*****	**0.37***	0.17	0.17	0.11	**0.33***	0.13	**0.33***	**0.41***	0.12	**0.53*****	0.26	**0.47***
AER		1	0.26	**0.94*****	**0.94*****	**−0.42****	−0.13	−0.24	−0.24	**−0.38****	−0.16	−0.28	0.13	0.08	0.29	0.08	0.01	0.27	−0.19	0.01	0.13
nAER			1	**0.50****	0.27	−0.20	−0.23	**−0.54****	**−0.74*****	**−0.62*****	**−0.48****	**−0.56*****	0.18	−0.32	−0.12	−0.10	−0.20	−0.19	0.23	0.16	−0.07
wAER				1	**0.91*****	**−0.37***	−0.12	−0.29	**−0.39***	**−0.45****	−0.31	**−0.35***	0.11	0.12	0.24	0.21	0.13	0.12	−0.06	0.10	−0.08
VR					1	**−0.52*****	−0.20	−0.27	**−0.28***	**−0.40****	−0.19	**−0.31***	0.07	0.04	0.27	0.03	−0.04	0.30	**−0.30***	−0.11	0.02
ICONE						1	0.07	**0.33***	**0.47****	**0.45****	0.22	0.16	0.03	0.31	0.09	0.32	**0.41***	0.07	**0.58*****	0.18	**0.43***
Indoor T							1	**0.50*****	**0.43****	**0.46****	**0.37***	**0.40****	**−0.44****	−0.17	−0.31	−0.03	0.09	−0.17	−0.10	**−0.57****	−0.27
Indoor SH								1	**0.75*****	**0.63*****	**0.55*****	**0.75*****	**−0.61*****	0.23	−0.07	0.19	**0.42***	0.08	−0.03	−0.37	−0.09
1									1	**0.74*****	**0.61*****	**0.70*****	**−0.32***	**0.37***	0.12	0.24	**0.40***	0.22	−0.06	−0.18	0.11
2										1	**0.46*****	**0.61*****	−0.20	**0.33***	0.12	**0.34***	**0.39***	0.11	0.13	−0.07	−0.07
3											1	**0.52*****	**−0.64*****	−0.08	−0.18	−0.08	0.07	−0.16	−0.03	−0.13	0.08
4												1	**−0.48****	**0.31***	0.02	0.22	**0.31***	0.18	−0.09	−0.18	−0.03
5													1	**0.33***	0.30	0.22	0.05	0.29	0.26	**0.51***	0.18
6														1	**0.71*****	**0.80*****	**0.75*****	**0.58*****	0.16	0.15	−0.07
7															1	**0.85*****	**0.75*****	**0.73*****	0.17	0.35	−0.06
8																1	**0.91*****	**0.61*****	**0.35***	0.12	−0.04
9																	1	**0.64*****	**0.36***	0.08	−0.07
10																		1	0.09	0.06	0.01
11																			1	**0.56****	0.39
12																				1	0.07
13																					1

T, Temperature; SH, specific humidity; VR, Ventilation rate; nAER, night time AER; wAER, average AER during the school week.

Trichloroethylene, tetrachloroethylene and 1,4-dichlorobenzene are not presented because less than 30% of values are above LOQ.

1—Formaldehyde; 2—Acetaldehyde; 3—Butyraldehyde; 4—Hexaldehyde; 5—Benzene; 6—Toluene; 7—Ethylbenzene; 8—m,p-xylenes; 9—o-xylene; 10—Styrene; 11—$PM_{2.5}$; 12—$PN_{0.3-1}$; 13—PN_{1-20}.

**P*-value ≤ 0.050; ***P*-value ≤ 0.010; ***P-value ≤ 0.001.

SH and average indoor temperature. The correlations with SH were still significant during the heating and non-heating seasons but not with the indoor temperature. The association between aldehydes (specifically, formaldehyde) and humidity has already been reported in literature (Kim et al., 2013). Formaldehyde is widely used as an adhesive to manufacture building materials, furniture, and other wood-based household products (Yamashita et al., 2012). Increased humidity might enhance emissions of aldehydes from building materials and furniture (Pegas et al., 2011).

Benzene was negatively correlated with aldehydes, particularly butyraldehyde, but positively correlated with toluene and fine particles with a diameter of less than 1 μm. Toluene, ethylbenzene, xylenes, and styrene presented strong significant correlations between each other ($R_s > 0.5$, P-values < 0.001), which may indicate a common source.

Particulate matter ($PM_{2.5}$) was correlated with xylenes and fine particles with a diameter of less than 1 μm but not with larger particles.

Relationships between ventilation indicators. All of the ventilation indicators exhibited significant correlations with each other, with high correlations found between AER, wAER, and VR ($R_s > 0.90$, $P < 0.001$) and between the mean CO_2 concentration and ICONE score ($R_s = 0.99$, $P < 0.001$). nAER was only correlated with wAER. Although all of the indicators are based on indoor CO_2 monitoring, each one provides specific information regarding ventilation conditions in the classroom.

Air exchange rate is related to the efficiency of air renewal for a given volume of space and represents the best assessment of the ventilation constant, which can be interpreted both as an indoor sink and an outdoor source. However, because AER was only calculated during occupancy periods, it does not represent the ventilation constant during night-time, which may represent 75% of the school week. Weekly averaged AER takes both unoccupied and occupied periods into consideration and should be used for comparison with weekly averaged data. Night-time AER is more related to outdoor air infiltration through the building envelope compared with the other indicators (mechanical ventilation system turned off during night).

Ventilation rate represents the amount of airflow rate available for each occupant. VR is typically used for the design of the mechanical ventilation system and is useful for checking compliance with standards and guidelines.

The mean CO_2 concentration during occupancy depends on both metabolic production and ventilation conditions. This indicator can represent the level of accumulation that an indoor pollutant might reach when its sources are activated during occupancy periods, including particle resuspension. The ICONE index is a score based on the frequency of particular CO_2 thresholds being exceeded. The same average CO_2 concentration might lead to different scores for two measurements with completely different variances. The ICONE index represents a score related to the exposure of occupants to air stuffiness.

During the non-heating season, neither the CO_2 average nor the ICONE index were correlated with the other ventilation indicators, in contrast to what was observed during the heating season. This result indicates that these two parameters may be poor ventilation indicators during the non-heating season and may only represent the density of occupation.

The amount of time that the windows were open during the school week was considered as an additional ventilation indicator. However, because of information being poorly reported by the teachers, the duration could not be calculated, and hence, it was not used in this study. Another ventilation indicator that could have been used is the measurement of AER using a tracer gas technique (e.g. SF_6 injection). This method is accurate; however, the measurement is performed under specific conditions that may not represent the changes in ventilation conditions that might occur during a school week.

Relationships between ventilation indicators and indoor air pollutants. Weekly averaged data (i.e. VOC and aldehyde concentrations) were compared with wAER, which was calculated over the same timescale. Only three pollutants (formaldehyde, acetaldehyde, and hexaldehyde) were significantly and negatively correlated with wAER. The observed correlations were also significant during the heating season but not during the non-heating season. This result might indicate that for the heating season, the primary sources of these pollutants are located in the classrooms and an increase in the average AER lowers their respective indoor concentration. During the non-heating season, other factors, such as humidity, might compete with the dilution effect provided by an increase in the air exchange rate and could explain the lack of correlation. VOCs were not related with wAER for any season. Therefore, it can be assumed that the primary contribution to their indoor concentration comes from sources that are unaffected by changes in ventilation conditions, most likely outdoor sources.

Particulate matter and PN concentrations were compared with the indicators that represent ventilation conditions during occupancy periods, that is, the mean CO_2 concentration, ICONE index, AER, and VR. Fine particles below 1 μm in diameter were never correlated with any ventilation indicator for any season. $PM_{2.5}$ and PN_{1-20} were primarily positively correlated with CO_2 and the ICONE index but also to a lesser degree, with VR. The correlations remained significant in both seasons for $PM_{2.5}$ but not for PN_{1-20}. These correlations suggest that in schools, the CO_2 concentration or ICONE index could serve as a proxy for the presence of occupants, and hence, for their activities that could release particles in indoor air. As such, these two indicators cannot be completely related to ventilation whenever they are compared with human activity-related pollutants.

It should be noted that nAER was strongly correlated with all of the four aldehydes (most notably, with formaldehyde, $R_s < -0.74$, $P < 0.001$) and SH despite different time scales. Because nAER is also dependent on the season, the observed correlations are related to seasonal effects. During the heating season, nAER was significantly correlated with all aldehydes except butyraldehyde, whereas during the non-heating season, only formaldehyde remained correlated with nAER. These specific correlations may indicate that weekly averaged aldehyde concentrations are mostly driven by night-time periods.

It is important to consider the time scale of both the measurement and the ventilation indicators when determining correlations. The CO_2 concentration and ICONE index are indicators of both ventilation and occupancy conditions. Therefore, they should be better correlated with pollutants that vary in concentration according to specific human activities. The other ventilation indicators will correlate negatively with pollutants that have major indoor sources and not with pollutants that primarily are from the outdoors.

Intraschool variability

Measurements between classrooms from the same school were assumed to be independent in our analysis. However, this assumption may not be completely true when, for example, the primary sources of pollutants are outdoors or when all classrooms of a given school share the same ventilation system. On the other hand, indoor sources may be different between classrooms, and each teacher may exhibit a different behavior regarding opening windows. In these cases, the measurements between classrooms of the same school may not be correlated. Furthermore, the variability between classrooms may change depending on the season and the airing habits. To assess the degree of dependency between classrooms, intraschool and interschool variability [expressed as relative standard deviation (RSD)] were calculated for each parameter in the 17 schools (Table S6). The mean intraschool RSD was lower than the interschool variability for most of the measured parameters, except for the CO_2, ICONE index, and wAER. However, the intraschool variability varies greatly between schools. Nevertheless, intraschool RSD was always lower than interschool RSD for benzene, toluene, ethylbenzene, xylenes, styrene, $PN_{0.3-1}$, butyraldehyde, and SH. This result suggests that for these parameters, the major sources of variation are not related to a particular classroom, and rather, these parameters affect the entire school. For these parameters, a strong dependency exists between classrooms of a given school. In these cases, school-averaged data can be used for statistical analyses; however, for all other parameters, it will lead to a significant loss in information.

Conclusions

This study measured the IAQ using a multipollutant approach and ventilation in 51 classrooms. Different ventilation indicators, such as the mean CO_2 concentration (ppm), AER, nAER or wAER (per hour), VR (l/s per person), and the ICONE air stuffiness index, were used to assess classroom ventilation and to study their associations with measured IAQ parameters. The use of one or several indicators depends on the sampling period considered and the assigned objectives (e.g. compliance with guidelines, communication to occupants).

This study has improved the knowledge on correlations between ventilation conditions and IAQ. These correlations vary according to the location of the main source of each pollutant, outdoor vs. indoor, and when indoor, whether the source is associated with occupant activity or continuous emission.

These findings need to be confirmed with further studies that consider a larger set of classrooms. With this in mind, in 2013, the OQAI began a nationwide survey of 300 randomly selected French schools, including 600 classrooms. The sampling and analytical protocols that were implemented in the 17 schools of this study were shown to be feasible and relevant, and they are currently being deployed in the national survey.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Table S1. (A) Detailed information of the 30 classrooms monitored during the heating season (from 2010-01-11 to 2010-04-02); **(B)** Detailed information of the 21 classrooms monitored during the non-heating season (from 2010-04-26 to 2010-06-25); **(C)** Building characteristics, number and age of pupils, season of measurement according to the type of school.
Table S2. Temperature (°C) weekly means in the classrooms according to sampling periods and seasons ($n = 44$).
Table S3. Relative Humidity (%) weekly means in classrooms' indoors according to sampling periods and seasons ($n = 44$).
Table S4. Spearman correlations in the classrooms studied during the non-heating period (significant correlations in bold, P-value < 0.050).
Table S5. Spearman correlations in the classrooms studied during the heating period (significant correlations in bold, P-value < 0.050).
Table S6. Intraschool and interschool variability expressed as relative standard deviation (RSD).
Data S1. Indoor Climate Parameters.

References

Almeida, S.M., Canha, N., Silva, A., Freitas, M.C., Pegas, P., Alves, C., Evtyugina, M. and Pio, C.A. (2011) Children exposure to atmospheric particles in indoor of Lisbon primary schools, *Atmos. Environ.*, **45**, 7594–7599.

d'Ambrosio Alfano, F. R., Bellia, L., Boerstra, A., van Dijken, F., Ianniello, E., Lopardo, G., Minichiello, F., Romagnoni, P. and Gameiro da Silva, M. C. (2010) *Environment and Energy Efficiency in Schools (Part 1)*, REHVA guidebook 13, Brussels, Belgium, REHVA.

Annesi-Maesano, I., Hulin, M., Lavaud, F., Raherison, C., Kopferschmitt Blay, F., Charpin, D.A. and Denis, C. (2012) Poor air quality in classrooms related to asthma and rhinitis in primary schoolchildren of the French 6 Cities Study, *Thorax*, **67**, 682–688.

Annesi-Maesano, I., Baiz, N., Banerjee, S., Rudnai, P., Rive, S. and on behalf of the SINPHONIE Group. (2013) Indoor air quality and sources in schools and related health effects, *J. Toxicol. Environ. Health B Crit. Rev.*, **16**, 491–550.

ASHRAE (2007) *Ventilation for Acceptable Indoor Air Quality*, Atlanta, GA, American Society of Heating, Refrigerating and Air Conditioning Engineers (ASHRAE standard 62.1-2007).

Bakó-Biró, Zs., Clements-Croome, D.J., Kochhar, N., Awbi, H.B. and Williams, M.J. (2012) Ventilation rates in schools and pupils' performance, *Build. Environ.*, **48**, 215–223.

Blondeau, P., Iordache, V., Poupard, O., Genin, D. and Allard, F. (2005) Relationship between outdoor and indoor air quality in eight French schools, *Indoor Air*, **15**, 2–12.

Branis, M., Rezacova, P. and Domasova, M. (2005) The effect of outdoor air and indoor human activity on mass concentration of PM_{10}, $PM_{2.5}$, and PM_1 in a classroom, *Environ. Res.*, **99**, 143–149.

Brelih, N. and Seppänen, O. (2011) Ventilation rates and IAQ in European standards and national regulations. In: Proceedings of the 32nd AIVC Conference and 1st TightVent Conference in Brussels, 12 – 13 October 2011. Available online: www.aivc.org.

Canha, N., Almeida, S.M., Freitas, M.C., Täubel, M. and Hänninen, O. (2013) Winter ventilation rates at primary schools: comparison between Portugal and Finland, *J. Toxicol. Environ. Health A*, **76**, 400–408.

Canha, N., Almeida, S.M., Freitas, M.C., Trancoso, M., Sousa, A., Mouro, F. and Wolterbeek, H.Th (2014) Particulate matter analysis in indoor environments of urban and rural primary schools using passive sampling methodology, *Atmos. Environ.*, **83**, 21–34.

Chatzidiakou, L., Mumovic, D. and Summerfield, A.J. (2012) What do we know about indoor air quality in school classrooms? A critical review of the literature, *Intell. Build. Int.*, **4**, 228–259.

Chatzidiakou, L., Mumovic, D. and Summerfield, A. (2015) Is CO_2 a good proxy for indoor air quality in classrooms? Part 1: the interrelationships between thermal conditions, CO_2 levels, ventilation rates and selected indoor pollutants, *Build. Serv. Eng. Res. Technol.*, **36**, 129–161.

Daisey, J.M., Angell, W.J. and Apte, M.G. (2003) Indoor air quality, ventilation and health symptoms in schools: an analysis of existing information, *Indoor Air*, **13**, 53–64.

European Committee for Standardization. (2007) EN 15251: 2007. Indoor environmental input parameters for design and assessment of energy performance of buildings addressing indoor air quality, thermal environment, lighting and acoustics. CEN. Available at www.cen.eu.

European Committee for Standardization. (2014) EN 12341: 2014. Ambient air — Standard gravimetric measurement method for the determination of the PM10 or PM2,5 mass concentration of suspended particulate matter. CEN. Available at www.cen.eu.

Fromme, H., Twardella, D., Dietrich, S., Heitmann, D., Schierl, R., Liebl, B. and Ruden, H. (2007) Particulate matter in the indoor air of classrooms—exploratory results from Munich and surrounding area, *Atmos. Environ.*, **41**, 854–866.

Fromme, H., Diemer, J., Dietrich, S., Cyrys, J., Heinrich, J., Lang, W., Kiranoglu, M. and Twardella, D. (2008) Chemical and morphological properties of particulate matter (PM_{10}, $PM_{2.5}$) in school classrooms and outdoor air, *Atmos. Environ.*, **42**, 6597–6605.

Geelen, L.M.J., Huijbregts, M.A.J., Ragas, A.M.J., Bretveld, R.W., Jans, H.W.A., Doorn, W.J.V., Evertz, S.J.C.J. and Zijden, A.V.D. (2008) Comparing the effectiveness of interventions to improve ventilation behavior in primary schools, *Indoor Air*, **18**, 416–424.

Godwin, C. and Batterman, S. (2007) Indoor air quality in Michigan schools, *Indoor Air*, **17**, 109–121.

Guo, H., Morawska, L., He, C. and Gilbert, D. (2008) Impact of ventilation scenario on air exchange rates and on indoor particle number concentrations in an air-conditioned classroom, *Atmos. Environ.*, **42**, 757–768.

Habil, M. and Taneja, A. (2011) Children's exposure to indoor particulate matter in naturally ventilated schools in India, *Indoor Built Environ.*, **20**, 430–448.

Hänninen, O. (2013) Novel second degree solution to single zone mass-balance equation improves the use of build-up data in estimating ventilation rates in classrooms, *J. Chem. Health Saf.*, **20**, 14–19.

Haverinen-Shaughnessy, U., Moschandreas, D. and Shaughnessy, R.J. (2011) Association between substandard classroom ventilation rates and students' academic achievement, *Indoor Air*, **21**, 121–131.

Janssen, N.A.H., Vliet, P.H.N., Aarts, F., Harssema, H. and Brunekreef, B. (2001) Assessment of exposure to traffic related air pollution of children attending schools near motorways, *Atmos. Environ.*, **35**, 3875–3884.

John, K., Karnae, S., Crist, K., Kim, M. and Kulkarni, A. (2007) Analysis of trace elements and ions in ambient fine particulate matter at three elementary schools in Ohio, *J. Air Waste Manag. Assoc.*, **57**, 394–406.

Kim, J., Kim, S., Lee, K., Yoon, D., Lee, J. and Ju, D. (2013) Indoor aldehydes concentration and emission rate of formaldehyde in libraries and private reading rooms, *Atmos. Environ.*, **71**, 1–6.

Koistinen, K., Kotzias, D., Kephalopoulos, S., Schlitt, C., Carrer, P., Jantunen, M., Kirchner, S., McLaughlin, J., Mølhave, L., Fernandes, E.O. and Seifert, B. (2008) The INDEX project: executive summary of a European Union project on indoor air pollutants, *Allergy*, **63**, 810–819.

Kotzias, D., Koistinen, K., Kephalopoulos, S., Schlitt, C., Carrer, P., Maroni, M., Jantunen, M., Cochet, C., Kirchner, S., Lindvall, T., McLaughlin, J., Mølhave, Lars, Fernandes, E.O. and Seifert, B. (2005) *Final Report—The INDEX project: Critical Appraisal of the Setting and Implementation of Indoor Exposure Limits in the EU*, Luxembourg, European Commission, Directorate-General Joint Research Centre.

Lim, Y.W., Kim, H.H., Lee, C.S., Shin, D.C., Chang, Y.S. and Yang, J.Y. (2014) Exposure assessment and health risk of poly-brominated diphenyl ether (PBDE) flame retardants in the indoor environment of elementary school students in Korea, *Sci. Total Environ.*, **470–471**, 1376–1389.

Liu, Y., Chen, R., Shen, X. and Mao, X. (2004) Wintertime indoor air levels of PM10, PM2.5 and PM1 at public places and their contributions to TSP, *Environ. Int.*, **30**, 189–197.

Madureira, J., Paciência, I. and Fernandes, E.O. (2012) Levels and indoor–outdoor relationships of size-specific particulate matter in naturally ventilated Portuguese schools, *J. Toxicol. Environ. Health A*, **75**, 1423–1436.

MEDDTL. (2012), Décret no 2012-14 du 05/01/2012 Relatif à l'évaluation des moyens d'aération et à la mesure des polluants effectuées au titre de la surveillance de la qualité de l'air intérieur de certains établissements recevant du public, Journal Officiel de la République Française du 06/01/2012, Ministère de l'Ecologie, du Développement Durable, des Transports et du Logement.

Mendell, M.J. and Heath, G.A. (2005) Do indoor pollutants and thermal conditions in schools influence student performance? A critical review of the literature, *Indoor Air*, **15**, 27–52.

Mendell, M.J., Eliseeva, E.A., Davies, M.M., Spears, M., Lobscheid, A., Fisk, W.J. and Apte, M.G. (2013) Association of classroom ventilation with reduced illness absence: a prospective study in California elementary schools, *Indoor Air*, **23**, 515–528.

Michelot, N., Marchand, C., Ramalho, O., Delmas, V. and Carrega, M. (2013) Monitoring indoor air quality in French schools and day-care centers, *HVAC&R Res.*, **19**, 1083–1089.

Molnár, P., Bellander, T., Sällsten, G. and Boman, J. (2007) Indoor and outdoor concentrations of $PM_{2.5}$ trace elements at homes, preschools and schools in Stockholm, Sweden, *J. Environ. Monit.*, **9**, 348–357.

NOAA. (2014) National Oceanic and Atmospheric Administration (NOAA) - Earth System Research Laboratory (ESRL) - Global Monitoring Division (GMD) - Mauna Loa Observatory (MLO), Hawai, USA. Available online: http://www.esrl.noaa.gov.

Pegas, P.N., Alves, C.A., Evtyugina, M.G., Nunes, T., Cerqueira, M., Franchi, M., Pio, C.A., Almeida, S.N., Cabo Verde, S. and Freitas, M.C. (2011) Seasonal evaluation of outdoor/indoor air quality in primary schools in Lisbon, *J. Environ. Monit.*, **13**, 657–667.

Pennequin-Cardinal, A., Plaisance, H., Locoge, N., Ramalho, O., Kirchner, S. and Galloo, J.C. (2005) Performances of the Radiello diffusive sampler for BTEX measurements: influence of exposure conditions and determination of modelled sampling rates, *Atmos. Environ.*, **39**, 2535–2544.

Poupard, O., Blondeau, P., Iordache, V. and Allard, F. (2005) Statistical analysis of parameters influencing the relationship between outdoor and indoor air quality in schools, *Atmos. Environ.*, **39**, 2071–2080.

Ramalho, O., Kirchner, S., Ségala, C. and Ribéron, J. (2005) Impact de la ventilation sur la santé respiratoire des écoliers, *Pollut. Atmos.*, **185**, 63–68.

Ramalho, O., Mandin, C., Ribéron, J. and Wyart, G. (2013) Air stuffiness and air exchange rate in French schools and day-care centres, *Int. J. Vent.*, **12**, 175–180.

Ribéron, J., Derbez, M., Lethrosne, M. and Kirchner., S. (2011) Impact of airing behaviour on air stuffiness in schools and daycare centres: Development of a specific tool for ventilation management, 12th International conference on indoor air quality and climate, Indoor Air'2011, Austin (USA), June 5-10, short communication.

Rosbach, J.T.M., Vonk, M., Duijm, F., Ginkel, J.T., Gehring, U. and Brunekreef, B. (2013) A ventilation intervention study in classrooms to improve indoor air quality: the FRESH study, *Environ. Health*, **12**, 110–120.

RSDT. (1978) Règlement Sanitaire Départemental Type, Circulaire du 9 août 1978 relative à la révision du règlement sanitaire départemental type. 13 septembre 1978.

Salo, P.M., Sever, M.L. and Zeldin, D.C. (2009) Indoor allergens in school and day care environments, *J. Allergy Clin. Immunol.*, **124**, 185–192.

Santamouris, M., Synnefa, A., Asssimakopoulos, M., Livada, I., Pavlou, K., Papaglastra, M., Gaitani, N., Kolokotsa, D. and Assimakopoulos, V. (2008) Experimental investigation of the air flow and indoor carbon dioxide concentration in classrooms with intermittent natural ventilation, *Energ. Buildings*, **40**, 1833–1843.

Sarigiannis, D.A., Karakitsios, S.P., Gotti, A., Ioannis, I.L. and Katsoyiannis, A. (2011) Exposure to major volatile organic compounds and carbonyls in European indoor environments and associated health risk, *Environ. Int.*, **37**, 743–765.

Shendell, D., Prill, G., Fisk, R., Apte, W.J., Blake, M.G. and Faulkner, D. (2004) Associations between classroom CO_2 concentrations and student attendance in Washington and Idaho, *Indoor Air*, **14**, 333–341.

Simoni, M., Cai, G.H., Norback, D., Annesi-Maesano, I., Lavaud, F., Sigsgaard, T., Wieslander, G., Nystad, W., Canciani, M., Viegi, G. and Sestini, P. (2011) Total viable molds and fungal-DNA in classrooms and association with respiratory health and pulmonary function of European schoolchildren, *Pediatr. Allergy Immunol.*, **22**, 43–52.

Stranger, M., Potgieter-Vermaak, S.S. and Grieken, R.V. (2007) Comparative overview of indoor air quality in Antwerp, Belgium, *Environ. Int.*, **33**, 789–797.

Stranger, M., Potgieter-Vermaak, S.S. and Grieken, R.V. (2008) Characterization of indoor air quality in primary schools in Antwerp, Belgium, *Indoor Air*, **18**, 454–463.

Sundell, J., Levin, H., Nazaroff, W.W., Cain, W.S., Fisk, W.J., Grimsrud, D.T., Gyntelberg, F., Li, Y., Persily, A.K., Pickering, A.C., Samet, J.M., Spengler, J.D., Taylor, S.T. and Weschler, C.J. (2011) Ventilation rates and health: multidisciplinary review of the scientific literature, *Indoor Air*, **21**, 191–204.

Tran, D.T., Alleman, L.Y., Coddeville, P. and Galloo, J.C. (2012) Elemental characterization and source identification of size resolved atmospheric particles in French classrooms, *Atmos. Environ.*, **54**, 250–259.

Tran, D.T., Alleman, L.Y., Coddeville, P. and Galloo, J.C. (2014) Indoor–outdoor behavior and sources of size-resolved airborne particles in French classrooms, *Build. Environ.*, **81**, 183–191.

Twardella, D., Matzen, W., Lahrz, T., Burghardt, R., Spegel, H., Hendrowarsito, L., Frenzel, A.C. and Fromme, H. (2012) Effect of classroom air quality on students' concentration: results of a cluster-randomized cross-over experimental study, *Indoor Air*, **22**, 378–387.

Wargocki, P. and Wyon, D.P. (2007) The effects of moderately raised classroom temperatures and classroom ventilation

rate on the performance of schoolwork by children, *HVAC&R Res.*, **13**, 193–220.
World Health Organization. (2005) *Effects of Air Pollution On Children's Health and Development–A Review of the Evidence*, WHO Regional Office for Europe, Denmark, WHO Press.
World Health Organization. (2006) *Air Quality Guidelines for Particulate Matter, Ozone, Nitrogen Dioxide and Sulfur Dioxide—Global Update 2005—Summary of Risk Assessment*, Geneva, Switzerland, WHO Press.
World Health Organization. (2010) *WHO Guidelines for Indoor Air Quality: Selected Pollutants*, WHO Regional Office for Europe, Denmark, WHO Press.
Wu, Q., Baek, S.Y., Fang, M. and Chang, Y.S. (2010) Distribution and fate of polybrominated diphenyl ethers in indoor environments of elementary schools, *Indoor Air*, **20**, 263–270.
Yamashita, S., Kume, K., Horiike, T., Honma, N., Masahiro, F. and Amagai, T. (2012) Emission sources and their contribution to indoor air pollution by carbonyl compounds in a school and a residential building in Shizuoka, Japan, *Indoor Built Environ.*, **21**, 392–402.