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Diurnal variation and potential sources of indoor formaldehyde at elementary school, high school and university in the Centre Val de Loire region of France

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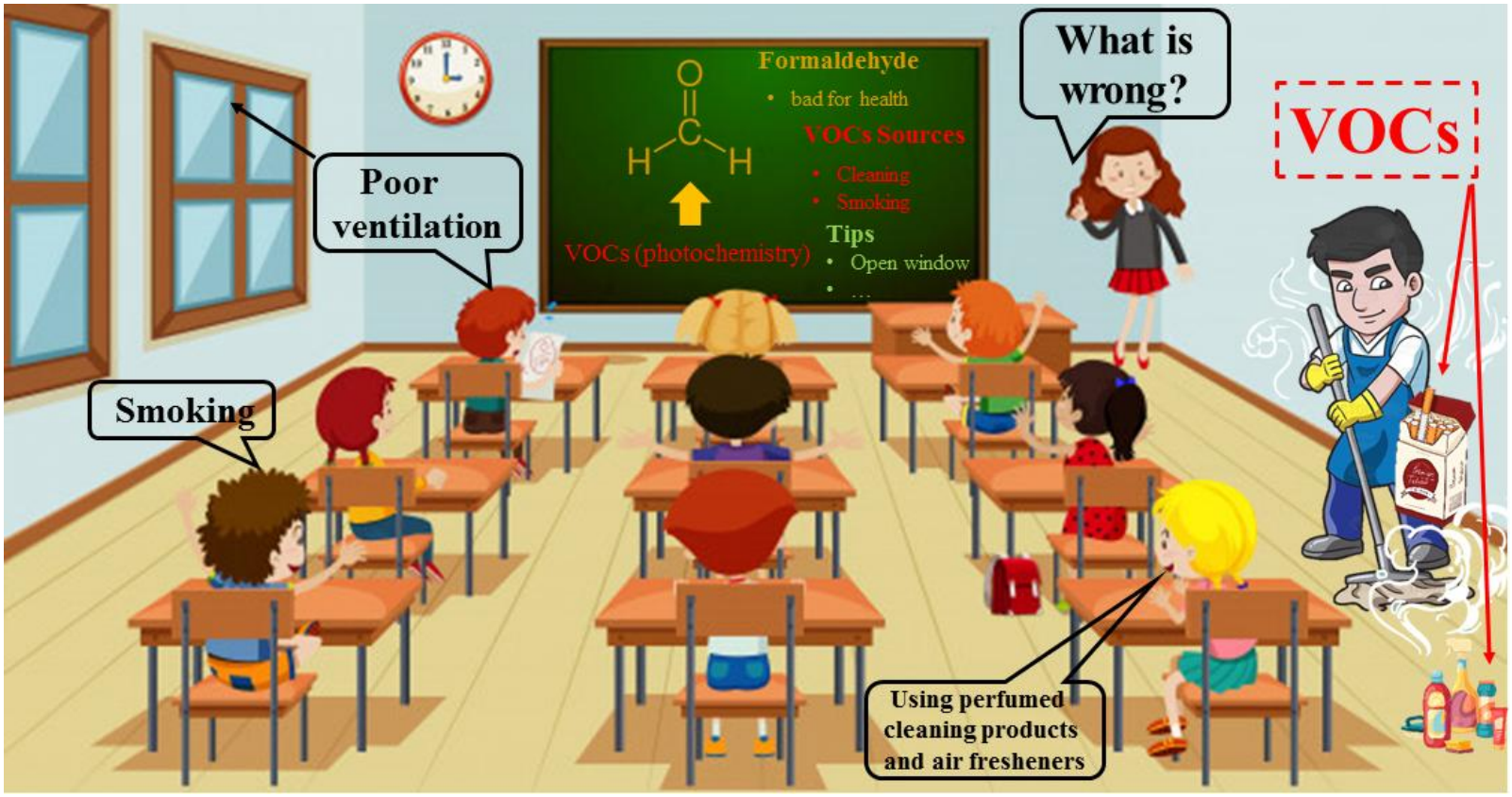

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ABSTRACT

Formaldehyde (HCHO) is one of the abundant indoor pollutants and has been classified as a human carcinogen by the International Agency for Research on Cancer (IARC). Indoor HCHO at schools is particularly important due to the high occupancy density and the health effects on children. In this study, high time resolved measurement of formaldehyde concentration was conducted in the classrooms at elementary school, high school and university under normal students' activities in three different locations in the Region Centre Val de Loire-France. Indoor average formaldehyde concentrations at those three educational institutions were observed to be in the range 10.96 - 17.95 μg/m^3, not exceeding the World Health Organization (WHO) guideline value of 100 μg/m^3. As expected, ventilation was found playing an important role in the control of indoor formaldehyde concentration. After opening windows for 30 minutes, formaldehyde level decreased by ~ 25% and 38% in the classroom at the elementary school and the high school, respectively. In addition to the primary sources, the objective of this study was also to determine potential secondary sources of indoor formaldehyde in these schools by measuring the other volatile organic compounds (VOCs) present in the classrooms by a Proton Transfer Reaction Time-of-Flight Mass Spectrometry (PTR-ToF-MS). The measurements suggest that the release of residue from tobacco smokers is one of the major sources of indoor HCHO at the high school, which increases HCHO by ~ 55% averagely

within 1 hour. Moreover, the control experiments conducted in the university suggests that VOCs such as that released from cleaning products like terpenes, can contribute to the increase of indoor formaldehyde levels through chemical reactions with ozone. This study confirms simple recommendations to reduce the indoors HCHO concentration in schools: use ventilation systems, limit the emissions like cigarette smoke or cleaning products. It also points out that the secondary sources of formaldehyde must be also considered in the classroom.

1. Introduction

Indoor air quality is an important occupational health and safety issue because people spend more than 80% of their time in indoor environments, where some pollutants can be at much higher concentration levels than outdoor (*Hodgson et al.*, 2002; *Jones*, 1999; *Lu et al.*, 2018). Indoor air quality at schools has attracted a lot of attention because of the poor air quality reported in classrooms and the health problems associated to it (*Annesi-Maesano et al.*, 2013; *Annesi-Maesano et al.*, 2012; *Baloch et al.*, 2020). Formaldehyde (HCHO) is one of the major and abundant indoor pollutants. It can be released directly from building and furnishing materials, cleaning agents, disinfectants, arts and crafts materials, personal care products and so on (*Chang et al.*, 2002; *Harčárová et al.*, 2020; *Salthammer*, 2019; *Salthammer et al.*, 2010). HCHO can also be formed by ozonolysis of alkenes present in indoor air (*Weschler*, 2001),

most often monoterpenes found in many consumer products, such as household products, air freshener and perfumed candles but also released from plants, wooden materials and paints. Short-term exposure to formaldehyde causes irritation of the airways and the eyes, while chronic exposure is associated with an increased risk of asthma (*Yu et al.*, 2020), and nasopharyngeal and sinonasal cancer (*HealthCanada*, 2005). Formaldehyde has been classified as a human carcinogen by the International Agency for Research on Cancer (*IARC*, 2006). In 2010, the World Health Organization (WHO) established an indoor air quality guideline for 30 min average exposure to formaldehyde of 100 µg/m^3 (*WHO*, 2011). This guideline has been re-evaluated for cancer risk assessment by *Nielsen et al.* (2017) who found that it has not been challenged by new studies. In France, the French Agency for Environmental and Occupational Health Safety (AFSSET) has suggested a healthy value of lower than 10 µg/m^3 and 50 µg/m^3 for long-term exposure and short-term exposure (2h), respectively (*AFSSET*, 2007). However, in 2011, French government established two target formaldehyde values for a long-term exposure: 30 µg/m^3 to be reached on January 1, 2015 and 10 µg/m^3 to be achieved on January 1, 2023 (Ministêre de L'Écoologie, du Développement Durable, des Transports et du Logement, 2011). In 2014, the Minister of the Environment postponed these obligations scheduled for January 2015, and in 2015 replaced it by a new decree (n° 2015-1000 of August 17,

2015, Ministère de la Transition écologique, 15 December 2020) with new deadlines: 2018 for nursery, elementary and nursery schools, January 1, 2020 for leisure centers and educational establishments of secondary school and January 1, 2023 for other establishments.

The regulatory framework for monitoring indoor air quality in these establishments includes:

- an assessment of the ventilation means that can be carried out by the establishments;
- implementation, as desired of a pollutant measurement campaign by an accredited body; or a self-assessment of air quality using the practical guide, allowing the establishment of an action plan (Ministère de la Transition écologique, 2019).

In recent decades, indoor HCHO concentration has been investigated intensively all over the world (*Brdarić et al.*, 2019; *Lee et al.*, 2018; *Norbäck et al.*, 2017; *Qin et al.*, 2020; *Yang et al.*, 2015). Studies demonstrate that the variation of HCHO concentration in indoor air varies depending on the age and type of buildings, ventilation, indoor air temperature, relative humidity (RH) etc. (*Clarisse et al.*, 2003; *Minami et al.*, 2002; *Parthasarathy et al.*, 2011; *Zhao et al.*, 2004). The concentration

of formaldehyde in indoor air is usually inversely correlated to the building age and air exchange rate (*Qin et al.*, 2020; *Zhao et al.*, 2004), but positively correlated to temperature and RH (*Parthasarathy et al.*, 2011). For example, *Zhao et al.* (2004) reported that the concentration of indoor formaldehyde showed a declining trend along with increase of building age and showed an increasing trend with temperature increase. *Minami et al.* (2002) reported that in a new private house, formaldehyde concentrations ranged between 0.07 and 0.23 ppm (corresponding to 93.8 and 308 μg/m^3) during the first months, while natural ventilation by opening windows was effective in decreasing the formaldehyde concentration. *Clarisse et al.* (2003) observed the formaldehyde levels in 61 dwellings in Paris were 21.7 μg/m^3, 24.3 μg/m^3, and 24.5 μg/m^3 in the kitchen, living room and bedroom, respectively.

Indoor air quality at schools is particularly important due to the high occupancy density and the health effects on children, which are more vulnerable to respiratory diseases such as allergic and asthma (*Daisey et al.*, 2003; *Zhao et al.*, 2008). Several recent studies were performed to survey indoor air pollution in schools and nurseries (*Baloch et al.*, 2020; *Becerra et al.*, 2020; *Brdaric et al.*, 2020; *Canha et al.*, 2016; *Michelot et al.*, 2013) and others have been focused on the incidence of respiratory problems (*Branco et al.*, 2020; *Huang et al.*, 2020; *Mohd Isa et al.*, 2020) and other symptoms like rhinitis, ocular, nasal and dermal problems, headache and fatigue

(*Norbäck et al.*, 2017). Other studies were concentrated on thermal conditions (*De Giuli et al.*, 2012; *Traumann et al.*, 2012), particulate matter (*Almeida et al.*, 2011; *Broich et al.*, 2012; *Devi et al.*, 2009; *Fromme et al.*, 2007; *Habil and Taneja*, 2011; *Lee and Chang*, 1999; 2000; *Wallace et al.*, 2011; *Yang et al.*, 2015), bioaerosol (*Kabir et al.*, 2012; *Pegas et al.*, 2011) and some specific groups of pollutants such as carbon dioxide (CO_2), carbon monoxide (CO), carbonyls, BTEX (benzene, toluene, ethylbenzene, xylenes) (*Brdarić et al.*, 2019; *Pegas et al.*, 2012; *St-Jean et al.*, 2012; *Stranger et al.*, 2008). In France, only one study dedicated to determine the potential sources of formaldehyde in educational institutions was reported in 2014 by *Poulhet et al.* (2014). The study was carried out during two campaigns in 2011 and 2012 in 8 schools of different cities of France where 29 primary sources of formaldehyde were identified. However, it was mainly emitted by ceiling and building materials and ranged from 1.2 to 252 $\mu g/m^2/h$. Some more recent studies have been devoted to characterize and measure the volatile organic compounds (VOCs) concentrations, including formaldehyde in schools. *Verriele et al.* (2016) studied 10 built energy-efficient French schools and levels of formaldehyde were around 15.9 $\mu g/m^3$. In 2016, *Cahna and coworkers* published a study on the relationship between ventilation and indoor air quality (IAQ) on 51 classrooms at 17 schools in France performed in 2010. Mean value of formaldehyde was 25±15 $\mu g/m^3$ (*Canha et al.*, 2016). A published

study in 2013 presented the monitoring of 310 French schools and day-care centers performed during campaigns in 2009 and 2010 (*Michelot et al.*, 2013). Concentration levels of formaldehyde were higher than 30 μg/m^3 for 10.6% of the investigated schools. However, most of these studies report the concentration levels of formaldehyde as an hourly or even daily/weekly average, which may hide the higher concentration which occurred in a short time period (few minutes). Moreover, only limited studies focus on the potential primary sources (*Poulhet et al.*, 2014). To our knowledge, so far no studies have been reported on the secondary sources of formaldehyde in schools in France.

The purpose of this work was to: (i) investigate the indoor levels and diurnal variation of formaldehyde at elementary school, high school and university in the Centre Val de Loire region of France by using a high time resolution HCHO monitor, and (ii) identify the potential primary and secondary sources of indoor formaldehyde at those educational institutions.

2. Material and methods

2.1 Site descriptions and sampling periods

Three representative classrooms from an elementary school, a high school and university of the Centre Val de Loire region of France were chosen for this study. The characteristics of investigated locations are summarized in Table 1. The elementary

school is situated in Bourges city, in the southeast of the central region of France, in an urban area close to the city center and near to streets with busy traffic. We have selected a 67-m^2 classroom that can receive 32 elementary-schoolers. It benefits from both northern and southern sun exposure and depends only on natural ventilation through two doors and seven windows. The school building is 140 years old. The high school is located in an urban area on the south side of Orleans city, near to streets with busy traffic. We have selected a 62-m^2 classroom that can receive 37 junior high school students. It benefits from western sun exposure and depends on a small controlled mechanical ventilation (CMV) and natural ventilation through two doors, five windows and two small natural ventilation grills. The school building is 24 years old. The third location was the Polytech Engineering school at the Orleans University campus which is located in a suburban area about ten kilometers south of Orleans city center and built in 1995. In this site, we selected a 81-m^2 classroom equipped for 48 students. It receives both southern and eastern sun exposure and depends only on natural ventilation through two doors and nine windows.

The measurements were conducted at the elementary school during the period from June 1 to June 18, 2012 (noted as “Elementary School-1”) and from February 1 to February 18, 2013 (noted as “Elementary School-2”). Measurements were carried out at the high school from March 9 to March 26, 2012 (noted as “High School-1”) and

from November 9 to November 26, 2012 (noted as “High School-2”). Measurements at the University campus were performed from January 17 to January 27, 2014 (noted as “University”). All measurements were performed in occupied classrooms. In addition, measurements on the unoccupied University’s classroom were also performed from June 26 to July 22, 2013 to identify the potential secondary sources of indoor formaldehyde.

2.2 Sampling and analyses

The instruments for indoor sampling were placed in a room adjoining the classrooms. All instruments were connected to an air-sampling manifold, equipped with a 15-m fluorinated ethylene propylene (FEP) Teflon tube, which reached the target classroom through the suspended ceiling. For each campaign, the sampling tube was placed in the middle of the classrooms and distance between the ceiling and air-entry point was set at 50 cm. A pump was used to sample the classroom air at a flow rate of 60 L/min. Consequently, all instruments received essentially identical air samples.

Formaldehyde was monitored by using an *on-line* HCHO analyzer (Aerolaser, AL 4021) with a temporal resolution of 30 seconds. The Aerolaser formaldehyde monitor is an extremely sensitive chemical analyzer, based on the transfer of gaseous HCHO from the atmosphere to a H_2SO_4 stripping solution. Quantification of the obtained HCHO liquid solution is based on the Hantzsch reaction with a detection by

fluorescence at λ=510 nm. A liquid calibration was made every week with a blank and two different dilutions of a HCHO standard solution (previously back titrated at 0.01 mol/L by pH metry method with HCl and Na_2CO_3), to obtain a curve with a coefficient of determination R^2 higher than 0.999. A liquid blank measurement was also done every morning during 15 minutes with a "zero" solution (from stripping solution of the system), to correct the baseline deviation. In these conditions, the limit of detection is estimated at 100 ppt and the uncertainty, determined by comparison of calibration factors, was less than 10% in this study. Meanwhile, the HCHO concentrations measured by an offline DNPH (2,4-Dinitrophenylhydrazine)-coated silica gel cartridge (Supelco) were used for HCHO inter-comparison in this study. The detailed description of the offline DNPH cartridge method is provided in the Supplementary materials (S1).

A Proton Transfer Reactor Time-of-Flight Mass Spectrometry (PTR-ToF-MS, IONICON) was used for monitoring VOCs with a temporal resolution of 10 minutes. Quantification by PTR-ToF-MS was reduced to some VOCs due to the limited number of gases available to be used as standards for a calibration. However, PTR-ToF-MS was also used to monitor qualitatively the variation trend of non-quantified VOCs in the classrooms air. At the meantime, VOCs were also sampled by a Perkin Elmer air sampler through a stainless-steel tube, using a pump at a flow rate of around

200 ml/min. Tubes were sampled every 2 hours on weekdays and every 3 hours at the weekend. Air Toxic tube analyses were performed using a thermal desorption technique with a Perkin Elmer gas chromatography separation and mass spectrometry (MS) detection. Blank samples were also analyzed under the same conditions. The detection limits were 0.01 ppb. Indoor temperature, relative humidity and CO_2 were examined by a QTrak monitor, the only instrument inside the classrooms.

Questionnaire: Two questionnaires were administered. The first one was provided to the school principals to collect general information about the building, furniture and maintenance activities, while the second one was provided to the teachers to obtain information about the specific daily activities and routine in class. The information was used to identify potential sources of pollutants and determine the effect of the natural ventilation supported by doors and windows.

2.3 Validation of methodology to monitor HCHO in the classrooms

Fig. 1(a) shows the comparison of HCHO concentration measured by both HCHO monitor and DNPH-Cartridges from January 20 to January 27, 2014 at the University classroom. As expected, they exhibit the same variation trend and match very well, implying the good quality of HCHO data obtained in this study. In the following discussion, all HCHO analysis results are based on the HCHO monitor data with high time resolution.

In addition, as the inlet point of the sampling tube was set at 50 cm from the ceiling for all measurements, we have studied the influence of the sampling height on the concentration of HCHO. Fig 1(b) shows the HCHO concentrations measured at the University classroom when variating the sampling height during the day. The figure shows that HCHO concentration follows a linear increase throughout the time. No significant change in the behavior of the HCHO concentration was evidenced when varying the sampling height.

3 Results and discussion

3.1 Indoor HCHO concentrations at three schools

Fig. 2 presents the variation of HCHO and CO_2 at the elementary school, high school and university, while Table 2 summarizes the corresponding average, minimum and maximum values together with indoor temperature (T) and relative humidity (RH). The average HCHO concentrations for Elementary School-1, Elementary School-2, High School-1, High School-2 and University measurements are 17.95, 11.73, 11.48, 10.96 and 13.33 μg/m^3, respectively. These values do not exceed the WHO guideline value of 100 μg/m^3 (*WHO*, 2011) and are comparable with other observations in schools in other places around the world (*Brdarić et al.*, 2019; *Norbäck et al.*, 2017) and in France (*Canha et al.*, 2016; *Poulhet et al.*, 2014; *Verriele et al.*, 2016). While average HCHO concentrations found in this study were lower than the French

regulation threshold of 30 μg/m^3 for long-term exposure applicable from January 2015, pretentions of a target value of 10 μm/m^3 are still not reached. It is noteworthy that the formaldehyde level in Elementary School-1 (17.95 μg/m^3) is higher than those observed in the other four measurements. This is probably due to: (1) the limited ventilation in the classroom at the elementary school which was based only on natural ventilation through doors and windows, and (2) relative higher average indoor temperature during this measurement (22 °C, Table 2). Formaldehyde is easy to be released from wood based products in the classroom under higher temperature. Moreover, photochemical production of HCHO is more active in hot seasons, even in an indoor environment (*Pang and Mu*, 2006).

As shown in Fig. 2, HCHO exhibits daily variations during working days, while it looks stable during the weekend. By comparison with the timetable of the class, as well as the information collected by the questionnaire, we found the formaldehyde level decreased when one or more windows were opened and increased when they were closed. Table 3 summarized the average, minimum and maximum decrease/increase percent of HCHO concentration after the windows were opened/closed for 30 minutes at elementary and high school classes. In the Elementary school-1 campaign, HCHO concentration decreased by around 25.5% when windows were opened, while it was observed to increase of around 34.1% when

windows were closed. Similarly, in the High School-1 measurements, HCHO concentration decreased by 37.8% when opening the windows and increased by 37.7% when closing them. Because Elementary School-2, High School-2 and University campaigns were conducted in winter where windows remain close, effect of window opening on the formaldehyde concentration was not carried out. In addition, HCHO shows the same variation trend as CO_2. As can be seen in Fig. S1, HCHO shows a positive correlation with CO_2 during the daytime (8:00-18:00) of working days, while it exhibits a weak correlation with CO_2 during the night (18:00-8:00) and weekend. This result implies that indoor HCHO concentration at schools was influenced by the activities of students.

3.2 Diurnal variation of indoor formaldehyde in three schools

Fig. 3 shows the statistical diurnal variation of indoor HCHO concentration at elementary school, high school and university. In the elementary school, indoor HCHO concentrations were stable during the weekend for both Elementary School-1 and Elementary School-2 measurements, while it showed a distinguished variation during working days. For the Elementary School-1 observation, HCHO concentration during the daytime (8:00-18:00) of working days was lower than that in the night (18:00-8:00). This was mainly due to windows and doors of the classroom at elementary school remained usually open during the daytime in summer (June).

Natural ventilation was effective for decreasing the formaldehyde concentration in indoor air (formaldehyde levels dropped by 25.5% after the windows are opened by 30 minutes, see Table 3). Furthermore, HCHO exhibited a clear daily pattern, with three steep falls around 8:00, 12:00 and 18:00 within a day (red arrows in Fig. 3(a1)). At around 8:00 am, students arrive in the morning and doors and windows are opened allowing indoor formaldehyde accumulated during the night to decreases. HCHO concentration increased with student class activities and showed several spikes during the period from 9:00 to 11:00. The second fall was exhibited around 12:00 when students left the classroom for lunch. After students returned to the classroom, HCHO concentration spikes were observed again from 13:00 to 17:00. The last fall occurred at around 18:00 when students finished their class and left the rooms. After that, the doors and windows were closed, and HCHO accumulated during the night time. In the Elementary School-2 observation, HCHO concentration during the daytime (8:00-18:00) of working days were higher than that in the night (18:00-8:00), and exhibited a clear daily pattern with a peak around 13:00. This difference compared to the Elementary School-1 campaign was attributed to the poorer ventilation during winter where windows and doors of the classroom were always closed even in the daytime. Thus, HCHO accumulated in the indoor air.

In the high school, the HCHO concentration was stable during the weekend but showed a particular variation during the working days. During the work days of the first campaign (High School-1), windows and doors were opened during the class. Therefore, HCHO showed a lower level during the daytime (8:00-18:00). It was notable that HCHO exhibited a peak around 8:15 am when students arrive to the classroom and few peaks after the breaks. This phenomenon is in contradiction with the observations made at the elementary school (Elementary School-1), where HCHO levels decreased when students arrive. It is suspected that tobacco smoke, desorbed from clothes or hair of students could be the origin of these morning HCHO peaks (see section 3.3). During the High School-2 measurement, windows and doors were mainly closed during the class and although a CMV ensured ventilation, HCHO concentration during the daytime (8:00-18:00) on working days was higher than that in the night (18:00-8:00). HCHO levels exhibited a clear daily pattern, with two peaks around 10:00 and 16:00 within a day. Interestingly, unlike that observed in High School-1 measurement, there was no typical HCHO peak, which proposed related with the tobacco smoke, around 8:15 am when students arrive. That's mainly because the windows and doors were closed during the High School-2 measurement, HCHO was continuously accumulated since the students arrive and the typical HCHO peak contributed from the tobacco smoke is immerged in the main peak at 10:00. The

decrease of HCHO at noon corresponded to the lunch time for students, when the classroom was empty.

In the university, HCHO concentration showed a relative weaker variation during the working days than that observed in elementary and high schools. By comparison with the timetable and occupancy of the class and the physical information of the classroom for these three schools, it is suggested that the weaker variation of HCHO at university was probably due to the larger volume and lower occupancy of the classroom. In addition, no typical HCHO peak (which may be related to tobacco smoke) was observed as well when the students arrive in the school. The same reason as that explained in High School-2 measurement, i.e., the closed windows and doors makes HCHO continuously accumulated since the students arrive and then the typical HCHO peak cannot be clearly identified. During the weekend, HCHO concentration is stable due to the lack of human activities.

3.3 Potential sources of indoor HCHO at schools

It is well known that formaldehyde can be released directly from building and furnishing materials, cleaning agents, disinfectants, arts and crafts materials, personal care products and so on. As shown in Fig. 2, during the working days in the high school, some sharp formaldehyde and CO_2 peaks were observed appearing when the students arrived in the morning or after some breaks. The formaldehyde level was

observed to rapidly increase by 4.5 to 11 μg/m^3 (55.3% on average) within one hour. This result implies that these formaldehyde peaks are strongly related to the student activities. It is well known that people can increase the indoor pollution levels by wearing dry cleaned clothes and perfumes. Moreover, smokers can increase indoor pollution, even if smoking inside the buildings is forbidden, because compounds can be retained in clothing and hair and then released into the classroom.

To determine if the tobacco smoke is responsible for the source of these formaldehyde peaks appearing upon students' arrival in the high school, the behavior of some tobacco compounds such as nicotine/anabasine ($C_{10}H_{14}N_2$), propyne (C_3H_4), myosmine ($C_9H_{10}N_2$), and nornicotine ($C_9H_{12}N_2$) were measured by a PTR-ToF-MS in the classroom (*Breiev et al.*, 2016; *Graus et al.*, 2010). As shown in Fig. 4, those tobacco emitted compounds were detected and showed the same trend as formaldehyde on the arrival of students in the high school-1 measurement. In addition, it is clear that there is no formaldehyde peak correlated with the children's arrival at elementary school-1 measurements, although it has the similar measurement condition as that in the high school-1 measurement, i.e., doors and windows opened. This is because elementary students don't smoke. This further supports the suggestion that the sharp formaldehyde peaks at the high school originating from the tobacco smoke from clothes or hair. Comparison between the measurements in high school and

elementary school shows that the residue from smokers' clothes or hair may further increase indoor HCHO by 55.3% in addition to human activities within 1 hour.

In addition to formaldehyde released directly from the surfaces inside the room, it can also be formed by oxidation of VOCs in ambient air (*Li et al.*, 2021; *Wolfe et al.*, 2016). Generally, using cleaning products in the classroom is one of the most common and important sources of indoor VOCs at schools (*Ramalho et al.*, 2015). For example, terpenes in the cleaning products can form formaldehyde through ozonolysis process in the indoor environment. To better understand the potential secondary sources of indoor formaldehyde at schools, a series of experiments, such as using the selected terpene-contained cleaning products or adding terpenes in the classroom, were planed and performed in an empty classroom at university (Fig. 5). For comparison, a reference experiment, i.e., without using cleaning products and keep the windows and doors closed, was carried out during the period from June 26 to July 3, 2014, The average HCHO concentration was 34.7 μg/m^3, a much higher value than that observed at the same classroom in January 2014 (13.33 μg/m^3, Table 2), when doors and windows were regularly opened by students and teachers. This result demonstrates that formaldehyde accumulates more easily in an enclosed environment. On July 4, some selected terpene-contained commercially purchased cleaning products (the chemical composition was analyzed by Gas Chromatography Mass

Spectroscopy (GC/MS) and presented in Fig. S2) were used to clean floors, tables, charts and windows in the classroom, and then the classroom was kept enclosed until July 10. Interestingly, no significant change of HCHO level was observed after using cleaning products, the average value over this period is 48.9 μg/m^3. This value is comparable with the average HCHO concentration (41.4 μg/m^3, from July 1 to July 3) before using cleaning products. As shown in Fig. 5, the ozone (O_3) concentration was very low (average values less than 4 μg/m^3) during both observation periods. It is evident from this study that the lack of indoor O_3 is restricting formaldehyde formation after using cleaning products.

To avoid the limitation of O_3 and better represent the condition of a real classroom in use, windows were opened for 20 minutes to bring ozone into the classroom before each of the following tests. On July 11 and July 15, two control experiments were conducted under normal conditions (i.e., without using any cleaning products and adding any VOCs, denoted as "control experiment"). On July 16 and July 18, the selected terpene-contained cleaning products were used inside the classroom. On July 12 and July 19, to figure out the contribution of the terpenes to increasing indoor formaldehyde levels through chemical reactions in schools, several terpenes which identified in the selected terpene-contained cleaning products (limonene, α-pinene, myrcene, sabinene, caryophyllene, decene, isoprene, linalool) were pre-mixed in the

lab and then introduced into the classroom by putting terpene-impregnated paper towels inside the classroom. For all these experiments (Fig. 5), a sharp decrease of HCHO with a distinct increase of O_3 was observed when opening windows. However after closing windows, the HCHO concentration increased with the consumption of O_3. To better understand the contribution of VOCs to the indoor formaldehyde, the comparisons of HCHO concentrations in presence and absence of cleaning products and terpenes in the classroom at the university are displayed in Fig. 6. It is obvious that after using cleaning products or adding terpenes inside the classroom, HCHO level increases faster than during the control experiment. These results imply that the VOCs from cleaning products such as terpenes can contribute to increase the indoor formaldehyde levels through chemical reactions, such as through ozonolysis process (*Weschler*, 2001) or photo-oxidation process (e.g., reaction with the OH radical formed by the photolysis of HONO in the classroom) (*Gómez Alvarez et al.*, 2013; *Zhou et al.*, 2021), at schools.

4 Conclusions

In the Framework of the FORMUL'AIR (FORmaldéhyde et siMUlations dans l'AIR: détermination des sources secondaires de formaldéhyde) project, measurements of indoor formaldehyde have been performed at elementary school, high school and university in Region Centre Val de Loire of France. The indoor average formaldehyde

concentrations at the three schools observed range from 10.96 to 17.95 μg/m^3, which is lower than the WHO guideline value of 100 μg/m^3 (*WHO*, 2011) and the current French regulation limit of 30 μg/m^3 for long term exposure. However, the threshold of 10 μg/m^3 considered for the French government, on January 2023, is not reached. Ventilation was found playing an important role in the control of indoor formaldehyde concentration. After opening windows for 30 minutes, indoor formaldehyde level decreased by 25.5% and 37.8% in the classroom at the elementary school and the high school, respectively. In addition, HCHO showed a positive correlation with CO_2 during the daytime (8:00-18:00) of weekdays, while it exhibited a weak correlation with CO_2 during the night (18:00-8:00) and at the weekend. This result implies that indoor HCHO concentration at schools was influenced by the activities of students. The PTR-ToF-MS measurements suggested that the releasing of the residue from tobacco smokers' clothes or hair is one of the primary sources of indoor HCHO at the high school, which increased HCHO concentration by an average of 55.3% within 1 hour. Moreover, the control experiments conducted at the university suggested that some VOCs released from cleaning products such as terpenes can contribute to increasing the indoor formaldehyde levels through chemical reactions.

Based on the results in this study, we believe that the following simple suggestions would effectively reduce the indoor HCHO concentration in Schools:

(1) Open the windows and doors regularly during the daytime on weekdays, and opening the windows and doors as long as possible before the students arrive in the morning is strongly recommended. Opening windows for 30 minutes can reduce indoor HCHO concentration by about 25-40%.

(2) Forbid the students from smoking. Indeed, HCHO from the clothes or hair of the tobacco smoker can be an important source of indoor HCHO, leading to an increase of ~ 55% within 1 hour.

(3) Use less emissive cleaning products and well ventilate afterward.

(4) Installation of adequate CMVs with high flow to guaranty the exchange of the indoor air in a short time would be a best investment to do for a best and healthy indoor air quality.

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Table 1. Characteristics of the studied schools.

Characteristics	Elementary School	High School	Polytech University
Location	Bourges	Orleans	Orleans
Classification	Urban (near to the city center)	Urban (near streets)	Sub-Urban (8 km south of the city center)
Champagne date	June 2012 and February 2013	March and November 2012	July 2013 and January 2014
Tested microenvironment	1 classroom	1 classroom	1 classroom
Room size	67 m^2 (8.95×7.46×3.03 m)	62 m^2 (8.23×7.52×2.70 m)	81 m^2 (12.50×6.50×2.90 m)
Windows	7	5	9
Doors	2	2	2
Floor	Tiled floor	Linoleum	Tiled floor
Ceiling	False ceiling	False ceiling	False ceiling
Board (Black or White)	4	1	2
Tables	17	18	42
Chairs	32	37	48
Built	1881	1997	1995
Ventilation	Natural	CMV*, Natural	Natural
Heating	Gas central system	Gas central system	Gas central system

CMV*: Controlled mechanical ventilation.

Table 2. Average, minimum and maximum values of HCHO, CO_2, Temperature and RH at elementary school, high school and university.

School	Period	HCHO (μg/m^3)			CO_2 (ppm)			Temperature (°C)			RH (%)		
		Ave	**Min**	**Max**	**Ave**	**Min**	**Max**	**Ave**	**Min**	**Max**	**Ave**	**Min**	**Max**
Elementary school	June 2012	17.95	4.98	32.85	532	367	2375	21.7	18.7	26.0	54.2	42.6	68.9
	February 2013	11.73	2.57	21.35	598	396	1954	16.4	8.8	22.0	42.2	26.3	62.0
High school	March 2012	11.48	2.81	22.67	522	349	2232	23.4	21.1	26.0	35.5	24.6	53.0
	November 2012	10.96	3.00	21.75	732	363	3363	21.1	17.6	24.9	43.9	32.1	59.5
University	January 2014	13.33	7.71	23.73	1016	230	4980	20.2	17.9	25.0	40.1	29.6	56.9

Table 3. Average, minimum and maximum decrease/increase percent of HCHO concentration after the windows are opened/closed in 30 minutes at elementary school and high school.

Campaign	Decrease percent (%)			Increase percent (%)		
	Ave	**Min**	**Max**	**Ave**	**Min**	**Max**
Elementary School-1	25.5	15.5	39.0	34.1	12.9	59.8
Elementary School-2	-	-	-	-	-	-
High School-1	37.8	20.3	67.4	37.7	19.6	73.3
High School-2	-	-	-	-	-	-

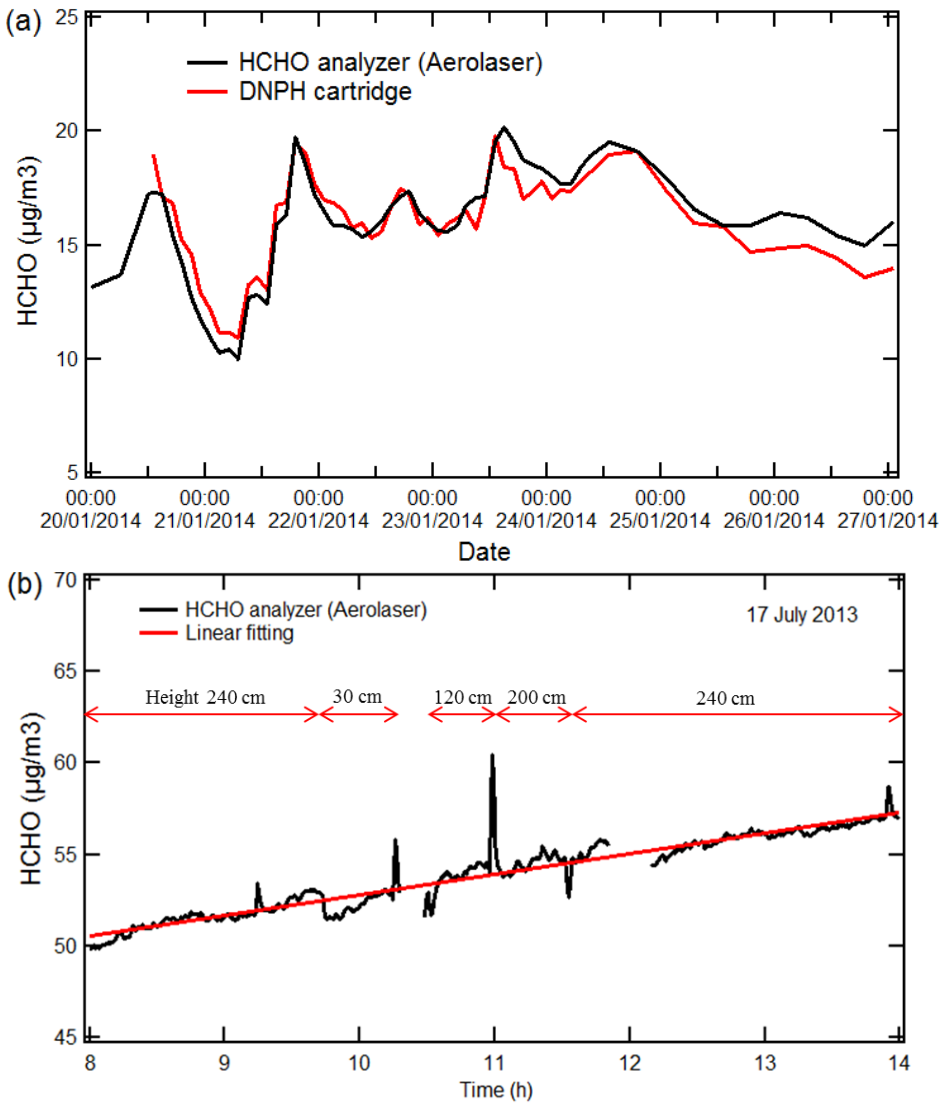

Fig. 1. Validation of HCHO data: (**a**) comparison of HCHO concentration measured by HCHO monitor and by DNPH-Cartridges during the period from January 20 to January 27, 2014 at Polytech University (**b**) Variation of HCHO concentration with sample height at Polytech University.

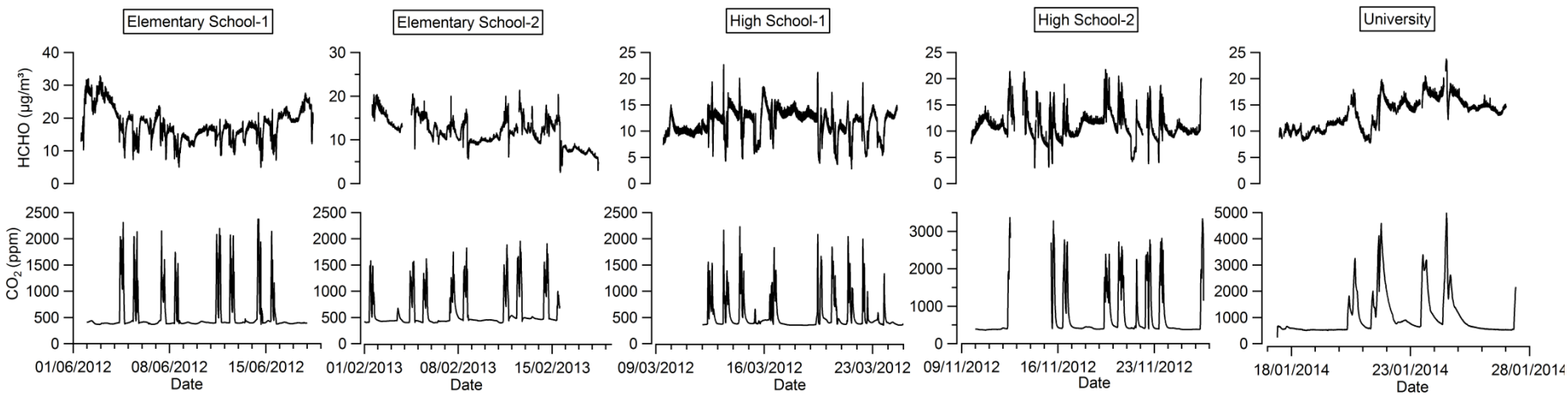

Fig. 2. Variation of HCHO and CO_2 concentration at elementary school, high school and university.

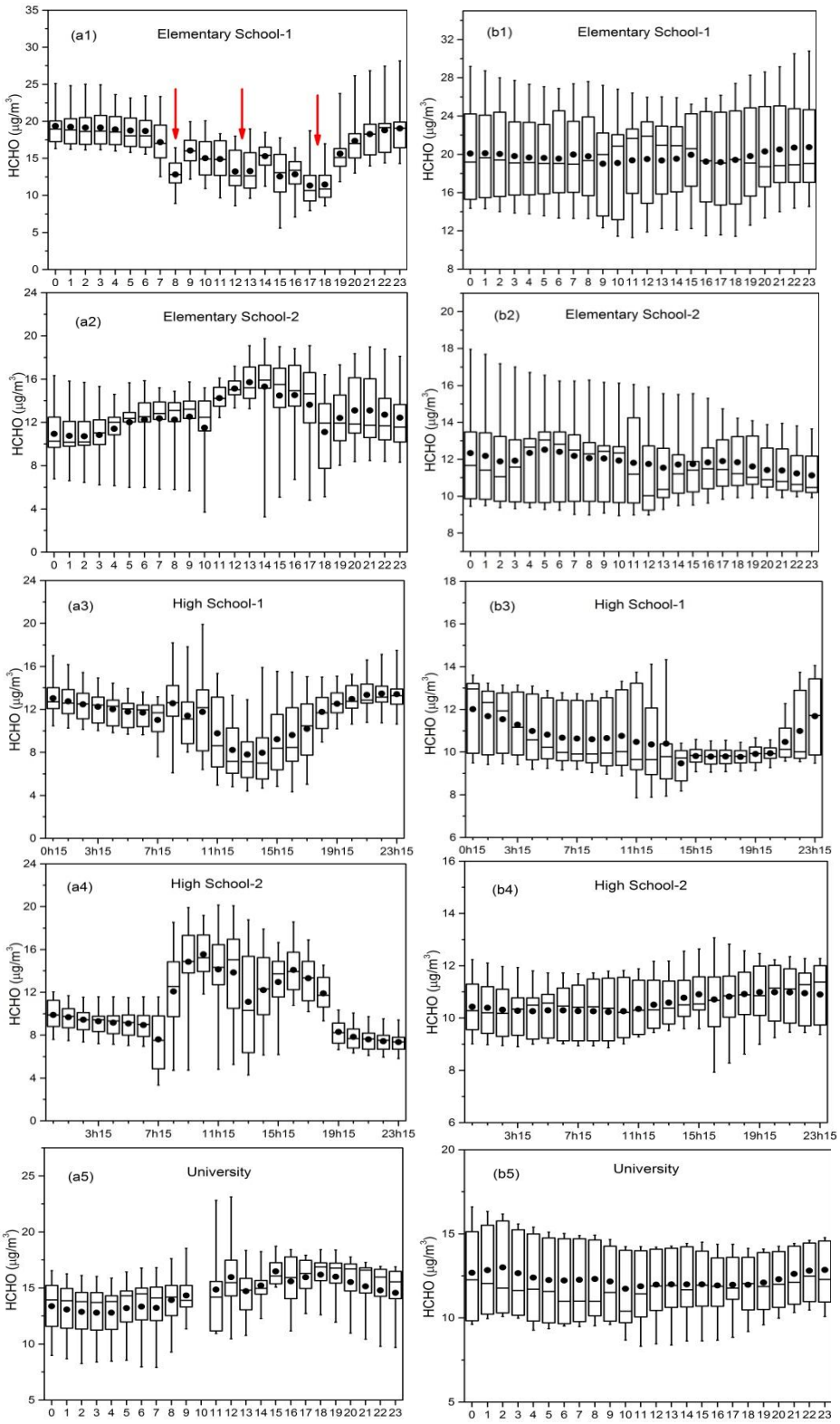

Fig. 3. Statistic diurnal variation of indoor HCHO during: (**a**) work days, and (**b**) weekend at elementary school, high school and university. The mean is represented by a dot, the dividing segment in the box is the median, the top and bottom box limits represent the 75th and 25th percentile, the error bars are related to the 95th and 5th percentiles.

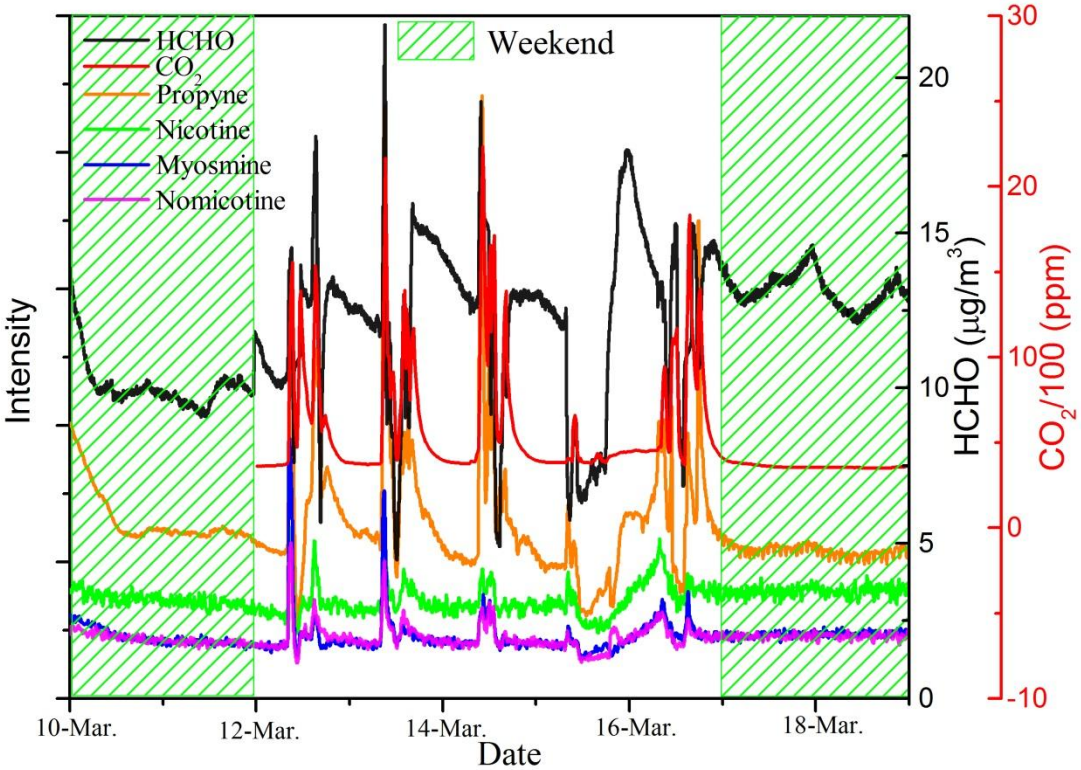

Fig. 4 Comparison of the formaldehyde variation and some tobacco compounds during the first week of campaign at High School-1.

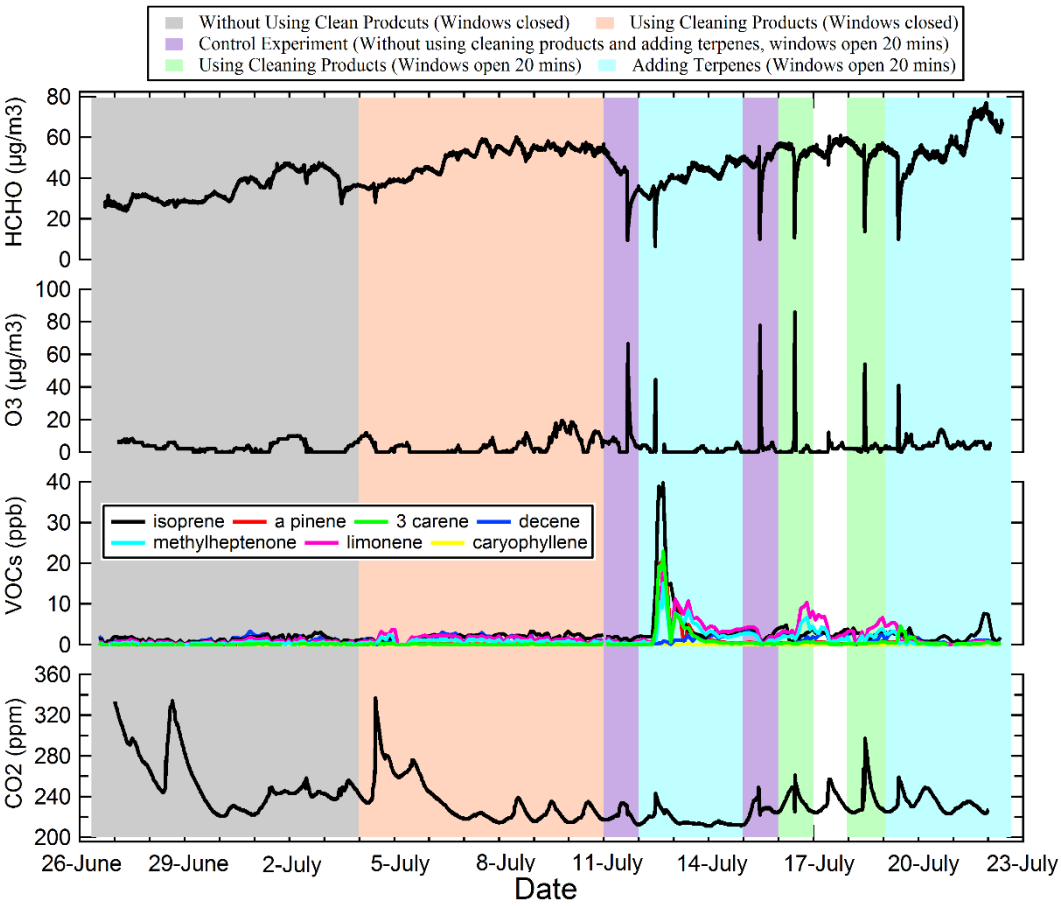

Fig. 5 Variation of indoor formaldehyde, VOCs and CO_2 at the university during the period from June 26 to July 22, 2013. The part marked in white corresponds to measurements during a day without specific experiment.

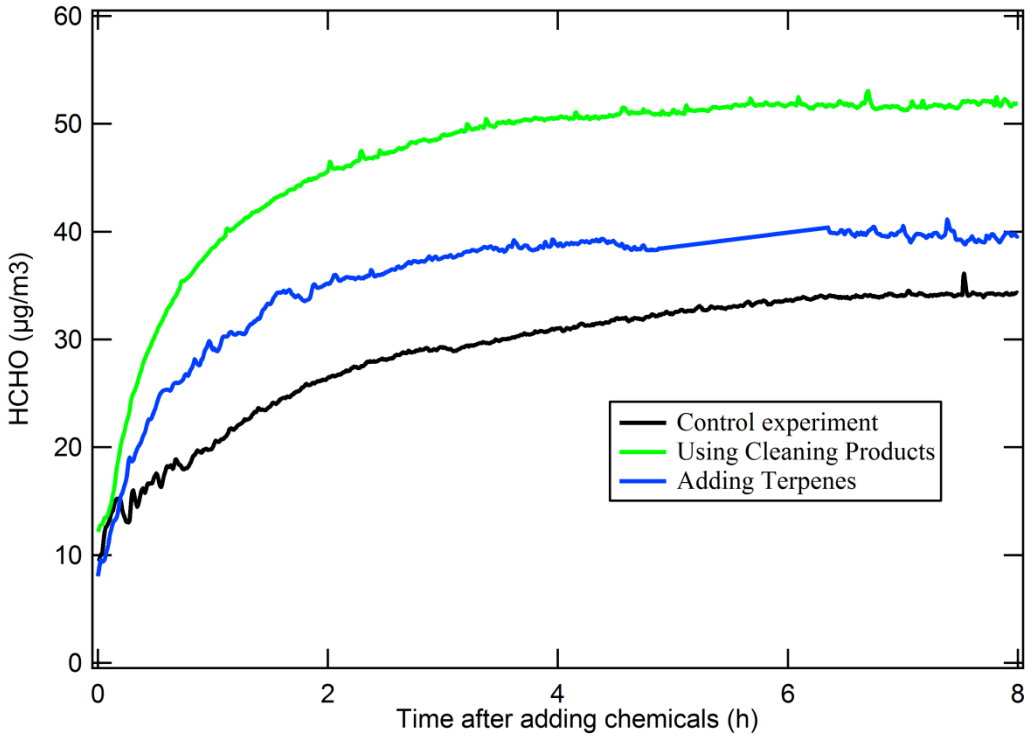

Fig. 6 Variation of HCHO concentration with and without using clean products and adding terpenes in the classroom at the university.