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Outdoor trends and indoor investigations of volatile organic compounds in two high schools of southern Italy

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Abstract
Volatile organic compounds (VOCs) are a class of ubiquitous substances that are present in outdoor and indoor air. They are emitted by a wide range of sources and can penetrate and accumulate specifically in indoor environments. Concern is growing among the scientific community regarding the potential health impacts of exposure to a high concentration of VOCs in indoor spaces. Due to their still-developing respiratory and immune systems, children may be the most fragile subjects in this regard, and therefore, the study of indoor air quality in schools is of the utmost importance. In this work, the concentrations of total volatile organic compounds (TVOCs) and of 20 specific compounds belonging to this class were determined in a school in Squinzano, a town in the province of Lecce (Apulia region, southern Italy). Sampling was carried out in indoor (classrooms and bathrooms) and outdoor (terrace) areas using passive diffusive samplers for VOCs and photoionization detectors for TVOCs. Average concentrations of both TVOC (303 ± 47 μg m^{-3}) and individual VOCs (< 0.5 μg m^{-3}) were below levels of concern; however, specific indoor sources such as cleaning activities and student occupancy were responsible for peaks in TVOC concentrations above harmless levels for children and school staff. The data were then compared to the ones obtained in a similar study conducted in a school in Galatina, another town of the Apulia region, highlighting the impact of the surrounding outdoor environment on the indoor profile of VOCs.

Keywords VOCs · Schools · Indoor environments · Air quality monitoring

Introduction

Volatile organic compounds (VOCs) are defined by the US Environmental Protection Agency (EPA) as "any carbon compound, excluding carbon monoxide, carbon dioxide, carbonic acid, metal carbides or carbonates, and ammonium carbonate, that participates in atmospheric photochemical reactions" (Environmental Protection Agency 2023). These substances are emitted by a number of sources, including fossil fuel use, biomass burning, incomplete combustion (traffic, industrial emissions, petroleum refining, and heating), fuel production (distribution and use), and solvent use (Zhang et al. 2021; Bergomi et al. 2022). In addition, VOCs can also be emitted by a wide range of indoor sources, such as products used in construction, decoration, and restoration of homes and buildings,and paints, waxes, cleaning products, solvents, coating materials, varnishes, and glues (Lucattini et al. 2018). Due to the wide variety of sources that can emit these species, the nature of volatile compounds is strongly site-specific, and concentrations are also influenced by other factors, such as atmospheric reactivity and meteorological conditions (De Lima et al. 2020; Zhang et al. 2021; Sarigiannis et al. 2011).

Due to the large number of emissive sources, volatile organic compounds are a class of ubiquitous compounds in ambient air that contribute significantly to air pollution.

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Indeed, it has been shown in numerous studies that a high concentration of VOCs is associated with acute episodes of air pollution (Zhang et al. 2021; Song et al. 2019). In addition to being emitted directly from numerous sources, VOCs are also key precursors for the formation of tropospheric ozone and secondary organic aerosol (SOA) (Zhang et al. 2021): the latter is an important component of fine particulate matter and a major contributor to haze pollution, whereas the former is the main cause of photochemical smog formation (Ramirez et al. 2012). Despite their ubiquitous presence in outdoor environments, VOCs are also one of the main indoor pollutants. This is because VOCs can penetrate from outdoors and be emitted by numerous indoor sources (Al-Awadi 2018). This leads to the accumulation of these substances, and therefore, concentrations are often greater indoors compared to outdoors (De Lima et al. 2020); in fact, indoor/outdoor (I/O) ratios are usually greater than 1 (Portela et al. 2021; Mishra et al. 2015). Because people spend between 80 and 95% of their time indoors, total human exposure to air pollutants is dominated by indoor air pollution (Carrer et al. 2000), and VOCs play a key role.

Due to the very large number of substances that are part of volatile organic compounds, there is great variability in terms of chemical composition and therefore also in terms of adverse effects on human health. Some compounds are harmless, whereas others can cause irritation and sensitization reactions of the eyes and the respiratory tract, show neurotoxic effects (e.g., toluene and xylenes), or even be known or suspected human carcinogens. One of the most harmful compounds, benzene, is a definite human carcinogen, classified by the International Agency for Research on Cancer (IARC) in group 1 (World Health Organization International Agency for Research on Cancer 2004). Also, many halogenated VOCs, including tetrachloroethylene, trichloroethylene, and 1–2-dibromoethane, are considered possible carcinogens to humans and are therefore classified as group 2 by the IARC (Ramirez et al. 2012). Indoor and outdoor environments are often characterized by the simultaneous presence of a wide variety of VOCs; therefore, recent studies are investigating a possible link between total volatile organic compounds (TVOCs) and certain health issues (Rumchev et al. 2007). Indeed, high concentrations of total indoor VOCs appear to pose a risk for cancer development, including lung cancer, blood cancer (leukemia and non-Hodgkin's lymphoma), liver cancer, kidney cancer, and biliary tract cancer (Ramirez et al. 2012). It has also been suggested that there may be an association between TVOCs and a complex set of nonspecific symptoms called sick building syndrome (SBS), mucous membrane irritation, and decreased tear film stability (Carrer et al. 2000).

Based on this scientific evidence, several institutions are starting to set limits on the concentration of VOCs in both indoor and outdoor spaces. The current air quality legislation in the European Union (European Council and Parliament Directive 2016/2284) aims to limit emissions of non-methane volatile organic compounds (NMVOCs) between 2020 and 2029 by specific amounts for each member nation (European Union 2016). However, to date, the only legal limit in the EU applies to the ambient air concentration of benzene (5 μg m^{-3}) (European Union 2008), and there are still no internationally accepted legal limits regarding the concentrations of any volatile organic compounds in indoor environments (Goodman et al. 2017). In this regard, international guidelines on indoor air quality (IAQ) started to include TVOC concentrations as a parameter for the evaluation of IAQ. The German Federal Environmental Agency, the World Health Organization (WHO), the RESET Standard, and the LEED Green Building Rating System have different IAQ grading systems based on TVOC levels, which agree on 500 μg m^{-3} as an upper limit to guarantee a harmless environment (Sensirion 2019).

Given the large number of hours that students and school staff spend indoors, IAQ in schools is an issue of growing concern. The most fragile individuals are certainly the children, whose organs are most vulnerable to airborne pollutants because of their higher respiratory rate and their still-developing respiratory and immune systems (World Health Organization 2006). In fact, prolonged exposure to indoor environments with poor air quality can lead to serious side effects, such as asthma, allergic disorders, and other respiratory diseases, and have a direct negative impact on the cognitive skills of the pupils and consequently on their academic performance (Portela et al. 2021; De Gennaro et al. 2013). Among the various classes of pollutants, several studies suggest that, in school environments, volatile organic compounds are directly responsible for several of the aforementioned negative health symptoms (Daisey et al. 2003; Madureira et al. 2015).

The studies that have been conducted to date on the presence of VOCs in school environments highlight the site-specific nature of these substances, both in terms of type and concentration. Liu et al. 2022a, b reported high mean concentrations of benzene, toluene, and xylenes in Chinese schools (40.4 μg m^{-3}, 46.0 μg m^{-3}, and 111 μg m^{-3}, respectively), with a large variability between different provinces. Lucialli et al. (2020) found levels of BTEX ranging between 0.10 and 29 μg m^{-3} in Italian schools located in highly polluted areas. Conversely, the most abundant VOCs in schools of central–southern Spain were aldehydes, namely formaldehyde (11–66 μg m^{-3}), and hexanal (9–27 μg m^{-3}) emitted from specific indoor sources (Villanueva et al. 2018), whereas terpenes were the majority of the VOCs (6–35 μg m^{-3}) in a series of Italian schools (De Gennaro et al. 2013). Finally, benzene, toluene, and formaldehyde were the main VOCs in Turkish schools (29 μg m^{-3}, 87 μg m^{-3},

and 106 μg m^{-3}, respectively), followed by naphthalene and xylenes (Sofuoglu et al. 2011).

Due to the site-specific nature of these pollutants, it is important to carry out as many case studies worldwide as possible to try and tackle this internationally relevant issue. In this framework, the aim of the present work is to investigate the presence of VOCs in a school located in a suburban area of the municipality of Squinzano (province of Lecce) to be compared with measurements obtained in another school, also located in the province of Lecce, but close to an industrial area and near several other schools. The concentration of TVOCs in different school environments (bathrooms, classrooms, terrace) was monitored continuously using photoionization detectors. In parallel, passive diffusive devices were used to sample volatile organic compounds of interest, which included BTEX (benzene, toluene, ethylbenzene, and xylenes), halogenated VOCs, and other alkylated derivates of benzene. This dual approach, rarely employed in similar studies, enabled us to determine the total amount of VOCs in indoor spaces, while determining the contribution of several compounds of greatest health concern. Analysis of individual VOCs allowed us to determine the nature and impact of both indoor and outdoor sources on air quality in school environments.

Materials and methods

Study area

The study was conducted in a high school in the small town of Squinzano (40° 26′ 5.82″ N, 18° 2′ 32.09″ E, 48 a.s.l.), located in the northernmost limits of the province of Lecce, in the Apulia region of southern Italy (Fig. 1). This town is in one of the rural parts of the region and has around 14,000 inhabitants. It is an important agricultural center with many olive trees and vineyards. The school includes about 250 students per year, with an average of 20 students per class. The results from this campaign were compared to the one obtained in a similar study conducted in a school in another Apulian town, Galatina (Ielpo et al. 2021). The choice of schools and the sampling period were driven by internal school management, with which our research institute (CNR ISAC Institute of Atmospheric Sciences and

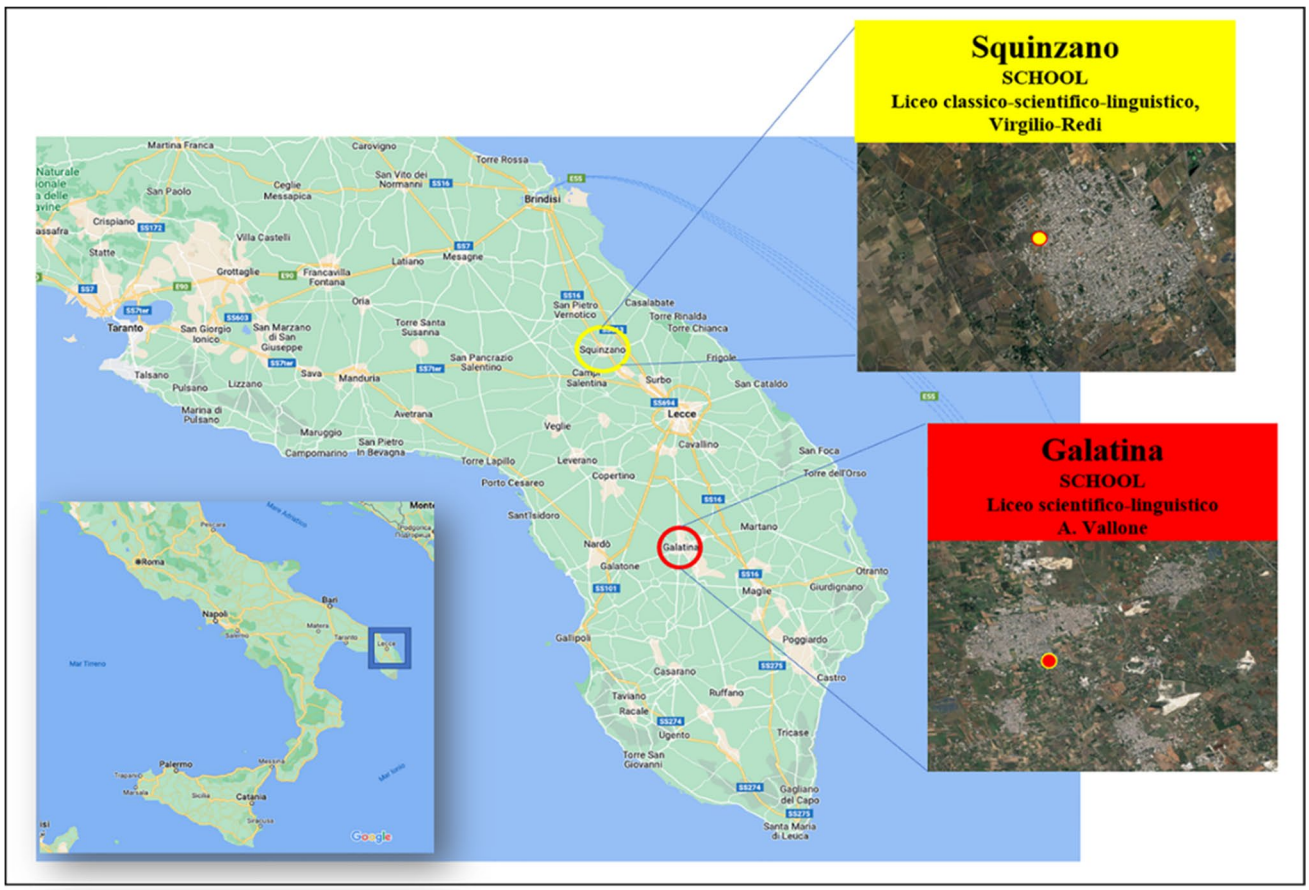

Fig. 1 Location of the two schools considered in this study: Liceo classico-scientifico-linguistico Virgilio-Redi (Squinzano) and Liceo scientifico-linguistico A. Vallone (Galatina)

Climate, National Research Council) had ongoing previous collaborations.

Volatile organic compounds—passive sampling

Volatile organic compounds were sampled using passive diffusive samplers (Radiello®, Fondazione Salvatore Maugeri-IRCCS, Padova, Italy). These systems consist of a white diffusive body (RAD 120), a support plate (RAD 121), a vertical adapter (RAD 122), and an adsorbent cartridge (RAD 130). The cartridges were constructed of stainless steel (100 mesh, 5.8 mm diameter), packed with activated carbon (30–50 mesh; 530 ± 30 mg), and placed inside a polyethylene container (60 × 16 mm) with a diffusion membrane of 25 µm porosity and 1.7 mm thickness that allowed controlled diffusion of pollutant gases to the adsorbent tube (diffusive path length of 18 mm). After sampling, the collected samples were stored at 4 °C until analysis.

Diffusive samplers were placed in four different locations within the school: the women's bathroom (B1), the men's bathroom (B2), a classroom (4B), and outdoors (O). Both male and female bathrooms were chosen in order to investigate possible different indoor sources, such as the use of cosmetic products and different gender inclinations to cigarette smoking. The monitoring campaign was conducted from 13 May 2019 to 3 June 2019 continuously, for a total of 22 days. Details regarding sampling locations and times are listed in Table 1.

Table 1 Sampling periods and locations

Sample	Location	Start date	Start time	End date	End time
985 QU	B2	13 May 2019	12:40	17 May 2019	11:15
984 QU	B1	13 May 2019	12:35	17 May 2019	11:10
982 QU	O	13 May 2019	12:02	17 May 2019	11:20
983 QU	4B	13 May 2019	12:17	17 May 2019	11:10
987 QU	B2	17 May 2019	11:15	20 May 2019	10:57
986 QU	B1	17 May 2019	11:10	20 May 2019	10:50
989 QU	O	17 May 2019	11:20	20 May 2019	10:32
988 QU	4B	17 May 2019	11:18	20 May 2019	10:20
993 QU	B2	20 May 2019	10:58	24 May 2019	12:25
992 QU	B1	20 May 2019	10:54	24 May 2019	12:30
991 QU	O	20 May 2019	10:35	24 May 2019	12:17
990 QU	4B	20 May 2019	10:25	24 May 2019	12:10
996 QU	B2	24 May 2019	12:25	27 May 2019	09:15
995 QU	B1	24 May 2019	12:35	27 May 2019	09:05
994 QU	O	24 May 2019	12:17	27 May 2019	09:32
Q 832 I	4B	24 May 2019	12:15	27 May 2019	09:25
998 QU	B2	27 May 2019	09:20	31 May 2019	12:20
997 QU	B1	27 May 2019	09:10	31 May 2019	12:25
999 QU	4B	27 May 2019	09:30	31 May 2019	12:15
Q 836 I	B2	31 May 2019	12:20	3 June 2019	13:25
Q 849 I	B1	31 May 2019	12:25	3 June 2019	13:20
Q 834 I	O	31 May 2019	12:10	3 June 2019	13:42
Q 835 I	4B	31 May 2019	12:15	3 June 2019	13:35
Q 792 I	B2	3 June 2019	13:30	7 June 2019	10:13
Q 793 I	B1	3 June 2019	13:25	7 June 2019	10:09
Q 795 I	O	3 June 2019	13:45	7 June 2019	10:20
Q 794 I	4B	3 June 2019	13:38	7 June 2019	10:03

Gas chromatography coupled to mass spectrometry (GC-MS) analysis

A total of 20 different VOCs (methyl tertiary-butyl ether; 1,2-dichloroethane; chloroform; carbon tetrachloride; trichloroethylene; bromodichloromethane; tetrachloroethylene; dibromochloroethylene; bromoform; trichloroethylene; benzene; toluene; ethylbenzene; m-,p-xylene; o-xylene; styrene; isopropylbenzene; 1,3,5-trimethylbenzene; 1,2,4-trimethylbenzene; and 1,2,3-trimethylbenzene) were extracted from the cartridges and analyzed using gas chromatography coupled to mass spectrometry (GC–MS). Considering that the nature of the VOCs emitted in school environments is strongly site-specific, the choice of which species to analyze was made according to the results of previous studies on indoor air quality conducted in several sites, including kindergartens, by the laboratories of the Dipartimento di Igiene e Prevenzione Sanitaria (DIPS) of ATS Città Metropolitana di Milano, in which the current analyses were also carried out. Such compounds include BTEX, other alkylated derivates of benzene, and halogenated VOCs, which are among the ones of greatest concern to the general public.

The procedures employed were carried out according to the specifications of the UNI EN TS/13649 technical standard. Specifically, the compounds were extracted using 2 mL of carbon disulfide (CS_2), stirring the solution occasionally over 30 min, either manually or with the help of a

mechanical stirrer (Vortex, VWR Collections). The extraction solvent was prepared to contain the internal standards (1,2-dichloroethane-d4; toluene-d8; and 4-bromofluorobenzene) at a concentration of 5 g mL^{-1}.

The analysis was carried out using GC–MS (Agilent Technologies Inc. 8890 GC System, Santa Clara, CA, USA) with a DB-624 column (60 × 0.25 mm internal diameter and 1.4 m film thickness) and using helium as the carrier gas. A total of 1 µL of solution was injected using an autosampler (GERSTEL robotic MPS multipurpose sampler) in splitless mode. The oven temperature was 48 °C for the initial 4 min and increased by 5 °C/min to 110 °C (holding 7 min), then increased by 6 °C/min to 165 °C (holding 0 min), and finally increased by 20 °C/min to 220 °C (holding 4 min). Overall, the duration of the chromatographic run was 40 min. Electron ionization (EI), conducted at 70 eV, was used as the ionization technique. The identification and quantification of the 20 VOCs tested were carried out by SIM and SCAN monitoring modes.

The limit of detection (LOD) and limit of quantification (LOQ) were 0.012 µg and 0.04 µg, respectively. These values are referred to the mass contained within the 2 mL extracted. This could correspond to different concentration values expressed in µg m^{-3} for every analyte in each one of the samplers.

Following the chemical analysis, the concentrations in air of the different VOCs were calculated using Eq. (1):

$$C = (M/(Q_k \times t)) \times 10^6, \quad (1)$$

where C is the concentration (µg m^{-3}), M is the mass (µg), Q_k is the sampling rate (mL min^{-1}), and t is the sampling time (min). In turn, Q_k was calculated using Eq. (2).

$$Q_k = Q_{298} \times (K/298)^{1.5} \quad (2)$$

where Q_{298} is the sampling rate at 298 K indicated by the manufacturer (mL min^{-1}) and K is the average absolute temperature during the sampling period (Table 2).

Table 2 Average absolute temperatures during sampling

Sample	Average absolute temperature / K
985 QU	314.09
984 QU	314.09
982 QU	314.10
983 QU	314.10
987 QU	316.65
986 QU	316.65
989 QU	316.65
988 QU	316.59
993 QU	317.86
992 QU	317.86
991 QU	317.86
990 QU	317.86
996 QU	318.68
995 QU	318.61
994 QU	318.67
Q 832 I	318.68
998 QU	319.19
997 QU	319.19
999 QU	319.19
Q 836 I	318.18
Q 849 I	318.18
Q 834 I	318.25
Q 835 I	318.25
Q 792 I	320.94
Q 793 I	320.96
Q 795 I	320.94
Q 794 I	320.94

Total volatile organic compound real-time monitoring

Photoionization detectors (PIDs) (Corvus, Ion Science Ltd., Fowlmere, UK) were used to monitor TVOC concentrations (expressed as ppm), along with temperature (°C) and relative humidity (%). The PID high-sensitivity technology consists of a Krypton UV lamp operating at 10.6 eV ionization potential, two collecting electrodes, and an electrical signal converter. The sampling concentration range for TVOC was between 0.01 and 50 ppm, with an accuracy of 134 ± 5 ppb. The limit of detection (LOD) was equal to 5 ppb. The measured TVOC values were automatically corrected for relative humidity thanks to the PID's featured built-in humidity compensation, which takes advantage of integrated humidity and temperature sensors. Prior to the experimental campaign, the PIDs were calibrated using isobutylene as the calibration gas (thus, TVOC concentrations are expressed and discussed in ppmv isobutylene equivalent). During the monitoring campaign, TVOC concentrations were recorded with a 1 s time resolution, and the concentrations were averaged every 15 min.

The monitoring sites were chosen in order to obtain maximum representativity of the data, as described below. Simultaneous indoor and outdoor monitoring of TVOC concentrations was carried out by placing one PID inside the classroom (4B) and another on the outdoor terrace of the school. Inside the classroom, the detector was placed about 1 m above the floor and at least 2 m away from windows, doors, or active heating systems. The choice of the monitoring location was carefully designed to make the measurements as representative as possible of the entire room volume and of the air breathed by the students. Moreover, a distance of at least 2 m was ensured between the windows

and the detector to avoid any air turbulence near the monitoring system, which could cause potential fluctuations in the data. Outdoor measurements were taken to establish external reference concentrations. The PID was placed on the lateral terrace of the school, on the first floor. The detector was located 1.5 m above the floor and 8 m away from the only wall present on the terrace. The other sides were completely exposed to outdoor air.

Results

Data from the Squinzano experimental campaign were compared with measurements taken at a school in the municipality of Galatina (Ielpo et al. 2021). The comparison was made in order to highlight the differences between the VOC profiles of the two schools, considering that, unlike Squinzano, Galatina is located in an industrial area with higher traffic rates at the school entrance and exit.

Average indoor VOC concentrations

Table 3 summarizes the number of samples showing VOC concentrations below the limits of detection (< LOD), between the limits of detection and the limits of quantification (< LOQ), and above the limits of quantification (> LOQ). VOC data measured at the Squinzano school were compared with the data measured in Galatina. The results highlight a limited presence of the analyzed VOCs in all the observed environments. Most of the samples were associated with VOC concentrations below the limits of detection (LOD) or the limits of quantification (LOQ).

With a few differences between the two schools, the most abundant species in both environments were the BTEX (benzene, toluene, ethylbenzene, and xylenes) compounds, along with other alkylated benzene derivates, such as 1,2,4-trimethylbenzene. The presence of halogenated VOCs was scarce in both sites, except for chloroform and carbon tetrachloride in Squinzano, which were detected above the LOQ in around half of the samples. Nevertheless, aside from two exceptions (11.83 μg m^{-3} and 14.35 μg m^{-3} sampled on 08/02/2019 in Galatina in one of the classrooms and outdoors, respectively), the observed concentrations of individual species were always below 5 μg m^{-3}, which is the annual limit for benzene in ambient air set by the European Union in the Air Quality Directive 2008/EC/50 and currently the only legal frame of reference for VOC concentrations (European Union 2008).

The average indoor concentration values of the main VOCs (BTEX and alkylbenzenes) observed in the two sites were compared (Fig. 2).

The concentration of alkylbenzenes was calculated by summing the levels of isopropylbenzene; 1,2,3-trimethylbenzene; 1,2,4-trimethylbenzene; and

Table 3 Number of samples showing VOC concentrations below the limits of detection (<LOD), below the limits of quantification (<LOQ), and above the limits of quantification (>LOQ) determined in the schools of Squinzano and Galatina (Ielpo et al. 2021)

VOC	Number of samples < LOD		Number of samples < LOQ		Number of samples > LOQ	
	Squinzano	Galatina	Squinzano	Galatina	Squinzano	Galatina
Benzene	25	0	1	0	1	35
Toluene	7	0	4	0	16	35
Ethylbenzene	1	12	4	5	22	18
p-,m-Xylene	1	11	1	5	25	19
o-Xylene	2	19	20	5	5	11
Styrene	26	34	1	1	0	0
Isopropylbenzene	19	22	8	13	0	0
1,2,3-Trimethylbenzene	13	26	14	2	0	7
1,2,4-Trimethylbenzene	0	17	2	0	25	18
1,3,5-Trimethylbenzene	1	22	25	1	1	12
Methyl tertiary-butyl ether	26	35	1	0	0	0
Trichloroethylene	27	34	0	0	0	1
1,2-Dichloroethane	27	29	0	0	0	6
Chloroform	8	17	8	18	11	0
Carbon tetrachloride	11	17	2	14	14	4
Trichloroethylene	27	35	0	0	0	0
Bromodichloromethane	27	34	0	1	0	0
Tetrachloroethylene	6	25	17	1	4	9
Dibromochloroethylene	27	–	0	–	0	–
Bromoform	27	–	0	–	0	–

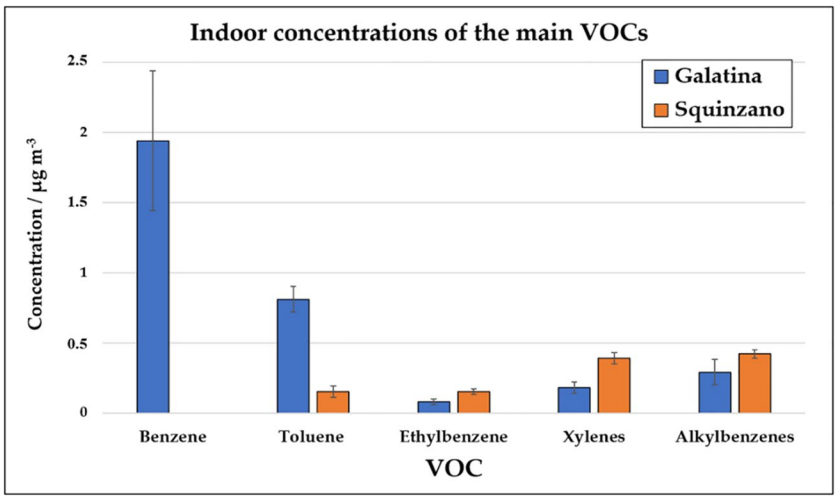

Fig. 2 Average indoor concentration values of the main VOCs observed in the schools of Squinzano and Galatina (Ielpo et al. 2021)

1,3,5-trimethylbenzene. In samples where the analyte was detected between the LOD and the LOQ, 0.5LOQ was used as the value for its concentration. As shown in Figure 2, the main difference between the two sites is represented by the levels of benzene; in Squinzano, this VOC was detected above the LOD in only two out of the twenty-seven samples analyzed (for this reason, no average value was calculated), whereas in Galatina, this was the most abundant VOC (1.94 µg m^{-3}). A similar trend was also observed for toluene: once again, the average indoor concentrations were lower in Squinzano with respect to Galatina. With regard to the other species, no significant differences were observed between the two sites, and in both cases, the average concentrations were always below 0.5 µg m^{-3}. In terms of alkylbenzenes, in both schools, the main compound was 1,2,4-trimethylbenzene, which accounted for more than 50% of the total amount.

VOCs in the classrooms

In order to evaluate the spatial distribution of the VOCs within the different school environments, average concentrations were calculated for specific areas, such as classrooms. Figure 3 shows the average concentrations registered in the classrooms of the two sites (benzene was not included in the histogram in order to better highlight the differences between the other major VOCs, as its concentrations were an order of magnitude greater than all the other species).

A similar trend to the one observed for the average indoor values was reflected also in the classrooms. Once again, benzene was detected only sporadically above the LOD in the classrooms of Squinzano, whereas it was the main VOC in the classrooms of Galatina (2.37 µg m^{-3}). Instead, only traces of all other non-halogenated species were observed in both sites, with concentrations rarely exceeding 0.5 µg m^{-3}. Finally, the halogenated VOCs were almost never detected above the LOQ.

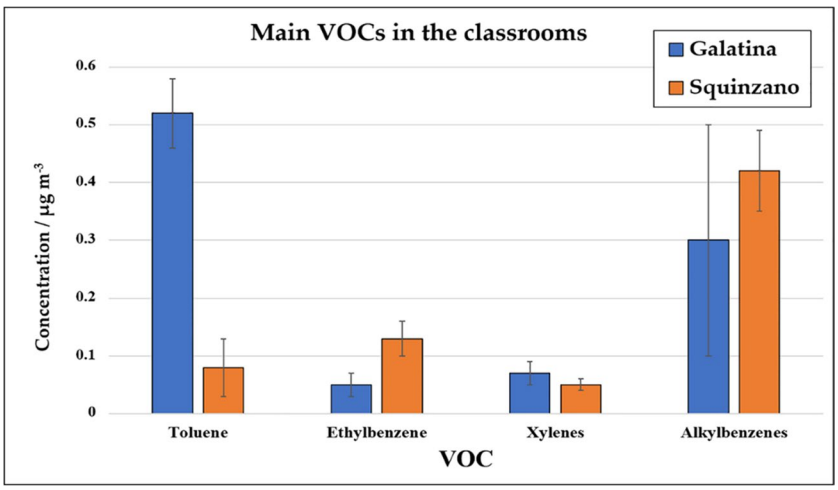

Fig. 3 Average concentration values of the main VOCs observed in the two classrooms of the schools of Squinzano and Galatina (Ielpo et al. 2021)

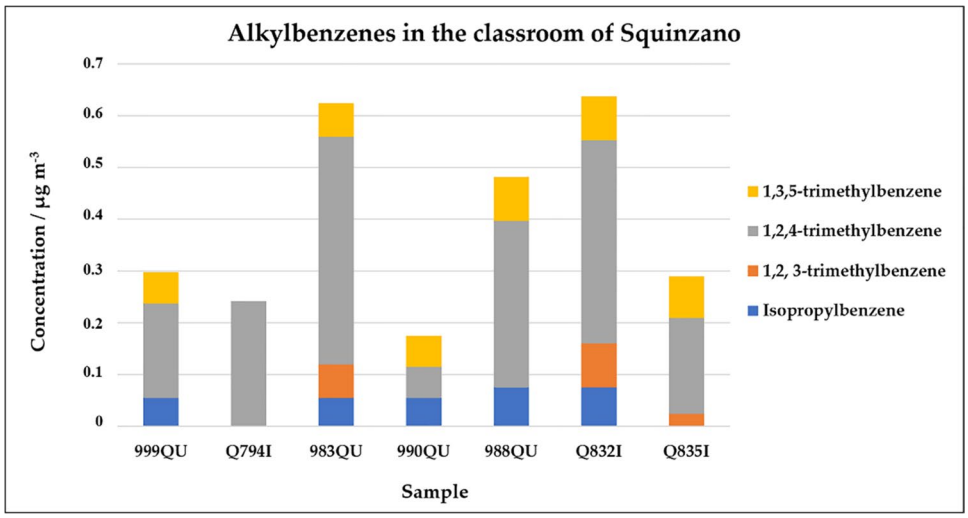

Fig. 4 Concentrations of alkylbenzenes in the classroom of Squinzano

In Squinzano, alkylbenzenes were the most abundant class of VOCs. Specifically, the most abundant compound within this class was 1,2,4-trimethylbenzene (Fig. 4).

Among the analyzed samples, the concentration of this compound was always greater than the sum of all other alkylbenzenes (except for sample 990QU). Overall, the concentration of these substances was comparable to the levels observed in Galatina, in which 1,2,4-trimehtylbenzene was also the most abundant compound of the class.

VOCs in the male (B2) and female (B1) bathrooms

Other environments that were investigated were the male and female bathrooms. Figure 5 shows the average indoor concentrations of the main VOCs registered in the male and female bathrooms.

With regard to the non-halogenated VOCs, a similar trend observed for the classrooms was registered. Benzene and toluene were the least abundant VOCs in Squinzano, present in much lower concentrations with respect to Galatina. Among the other species, similar concentrations were recorded in the two different locations and within the two environments studied (classrooms and bathrooms). The only exception was a greater concentration of xylenes in the bathrooms with respect to the classrooms, observed in both sites. Once again, the main component of alkylbenzenes was 1,2,4-trimehtylbenzene.

However, it is interesting to point out that detectable levels of chloroform and carbon tetrachloride were found in both the male and female bathrooms of Squinzano. Indeed, in most of the analyzed samples, these two compounds were detected at concentrations above the LOQ. This was not the case for Galatina, in which both VOCs were never found in concentrations above the LOQ, in line with the results of the other halogenated compounds.

Indoor/outdoor (I/O) ratios

I/O ratios are commonly employed to evaluate the sources of indoor pollutants. In this study, the indoor and outdoor average concentrations of the main VOCs were compared in both investigated sites (Fig. 6).

In Squinzano, the values are similar for all the species (no I/O ratio was calculated for benzene in Squinzano, because the concentrations of this compound were below the LOD in almost all samples), whereas in Galatina, the I/O ratios varied greatly between different compounds (from 0.60 for benzene to 3.60 for alkylbenzenes). Moreover, the I/O ratios for all species in Squinzano were around 1, suggesting no significant differences between the outdoor and indoor concentrations. In Galatina, it was possible to observe a greater indoor presence of alkylbenzenes, ethylbenzene, and toluene, whereas benzene was found in higher concentrations outdoors.

With regard to the halogenated VOCs, the only result worth mentioning is the average outdoor concentration of carbon tetrachloride in Squinzano (0.30 ± 0.04 µg m^{-3}), which was the only halogenated species detected above the LOQ in the outdoor environment. This value is in line with the concentrations observed in the male bathrooms and higher than the ones reported in the female bathrooms (and the classrooms, in which it was not detected above the LOQ). This suggests the presence of outdoor sources for this pollutant. The same conclusion cannot be drawn for chloroform, which was the only other halogenated VOC detected above the LOQ in the bathrooms of Squinzano. Indeed, this compound was not detected outdoors, indicating the presence of a specific indoor source.

Total volatile organic compound concentration

As already mentioned, the monitoring of TVOCs is of great interest in indoor environments because of the

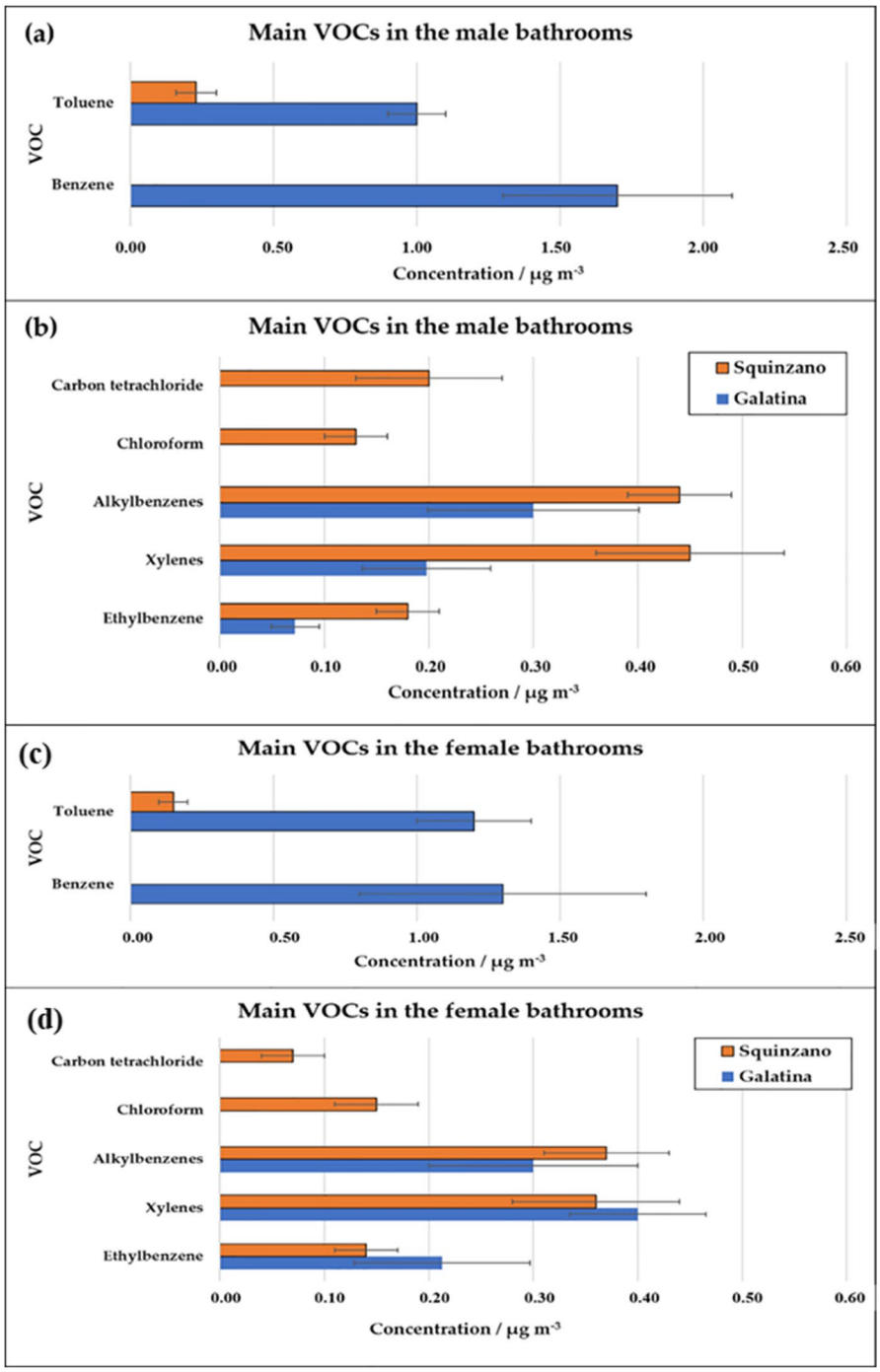

Fig. 5 Average concentration values in the schools of Squinzano and Galatina (Ielpo et al. 2021) of toluene and benzene in the male (**a**) and female (**c**) bathrooms and of carbon tetrachloride, chloroform, alkylbenzenes, xylenes, and ethylbenzene in the male (**b**) and female (**d**) bathrooms

adverse effects they have on human health. Keeping their concentrations monitored is therefore a priority, and recently, air purification systems have been developed that are able to significantly reduce VOC concentrations in indoor environments (Fermo et al. 2021). The concentration of TVOCs, and therefore the efficiency of the air purification systems, is monitored with photoionization detector devices, such as the one used in this study. Figure 7 shows the real-time TVOC concentrations in Squinzano for the period from 20 May 2019 to 2 June 2019, both inside and outside the school. As evident, indoor concentrations are systematically lower than outdoors, except in the cases of indoor peaks. The average TVOC value for the indoor site was 0.13 ± 0.02 ppm (303 ± 47 µg m^{-3}), while it was 0.18 ± 0.03 ppm (420 ± 70 µg m^{-3}) for the outdoor site, in contrast to the results obtained for the TVOC monitoring in Galatina, where the indoor concentrations were systematically higher than the outdoor ones (Ielpo et al.

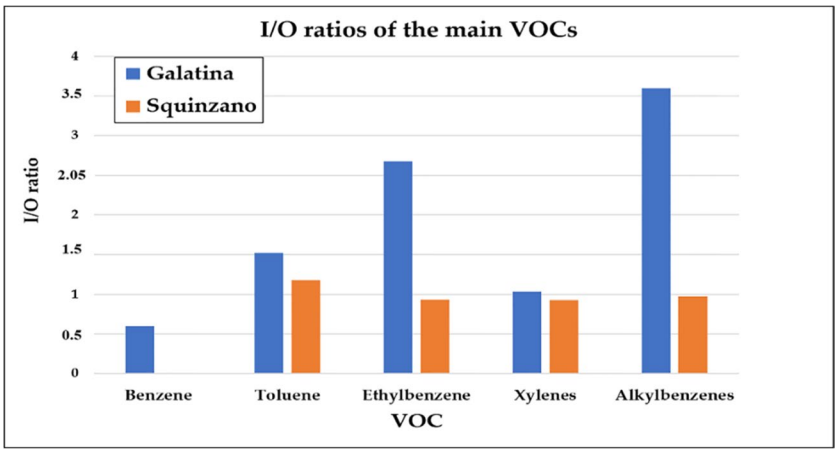

Fig. 6 I/O ratios of the main VOCs in Squinzano and Galatina (Ielpo et al. 2021)

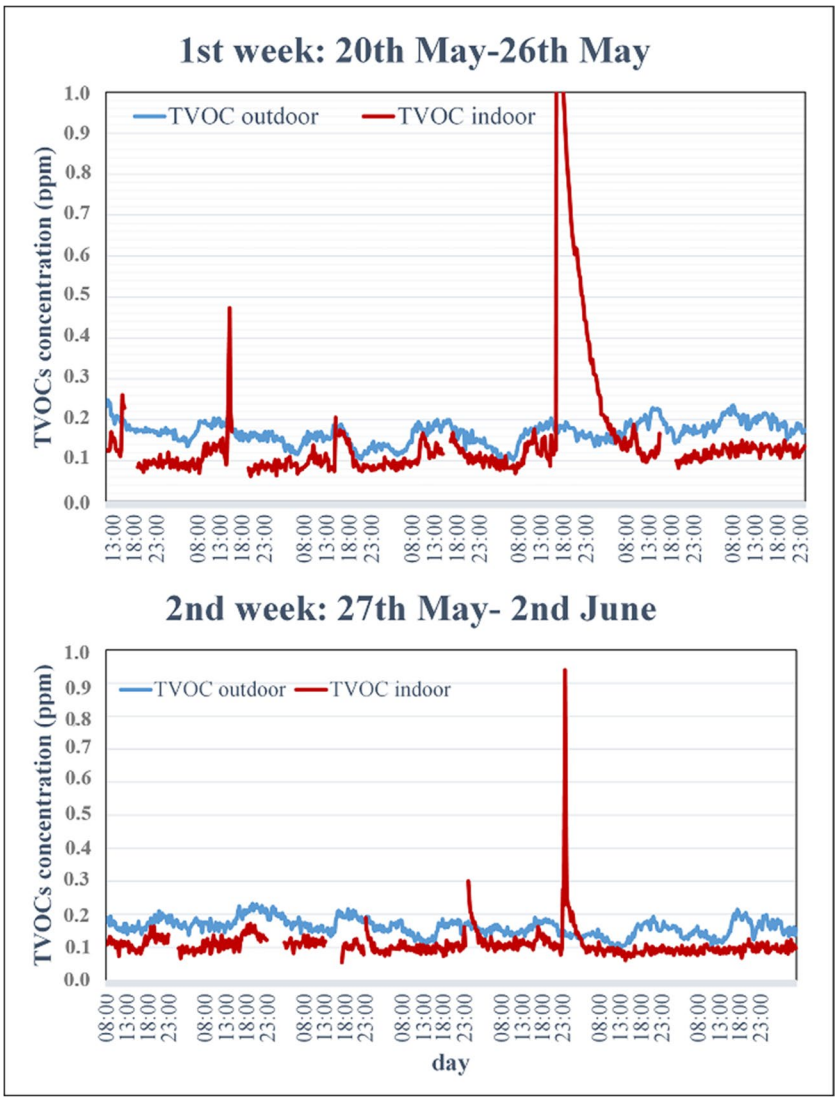

Fig. 7 Indoor and outdoor TVOC concentration values over the period from 20 May 2019 (Monday) to 2 June 2019 (Monday) in Squinzano

2021). Average indoor TVOC concentrations were below the limit (500 μg m^{-3}) indicated in several IAQ guidelines (Sensirion 2019) defining harmless conditions.

A more thorough analysis of the daily TVOC profiles shows, as expected, that the average value of indoor TVOC concentrations during school hours is higher than those

registered during Sundays, suggesting a non-negligible impact on student occupancy. In addition, the daily profiles show the presence of two peaks in TVOC concentrations during the mornings and afternoons, which are not directly related to penetration from outside. The morning peaks, recorded during daytime school hours, are probably due to student occupancy, while the afternoon TVOC peaks suggest a link between TVOC concentrations and cleaning activities. Despite average TVOC concentrations were below levels considered harmful, the concentrations observed during the peaks (often > 2 ppm, hence > 4667 μg m^{-3}) are related to poor and not acceptable IAQ according to several guidelines (Sensirion 2019).

A correlation between the peaks in the concentrations of TVOC and the corresponding passive sampler was not observed. This indicates that the compounds responsible for the rise in the concentration were not part of the 20 species quantified in this study. For this reason, additional substances were searched in the chromatograms obtained using SCAN mode. Only peaks clearly distinguishable from the background were considered, with a 70% or greater match between the proposed mass spectrum and the MS library. This qualitative analysis revealed the presence of other species which can, at least in part, account for the differences between the concentrations observed with the PID and with the passive samplers (Table 4).

Except for sample 990 QU, all the samples showed the presence of D-limonene. This monoterpene is emitted by several plants and is commonly found in nature (Król et al. 2014). However, its presence is not limited to outdoor environments, since there are also numerous indoor sources which can emit D-limonene, such as air fresheners, waxes, and cleaning agents (Vartiainen et al. 2016). Indeed, this terpene was found ubiquitously in both indoor and outdoor environments in Squinzano. Considering the location and the activities taking place in the school, the most probable indoor source is the use of cleaning products, whereas plant emissions probably dominate outdoor emissions. Similar considerations can be made for α-pinene, which is one of the most abundant biogenic terpenes emitted by plants, but is also a common indoor air pollutant due to its presence in building materials and household products (Waidyanatha et al. 2022). Unlike D-limonene, this substance was found only sporadically in all indoor environments (male bathroom, female bathroom, classroom) and was not detected in outdoor areas, indicating an indoor source with a limited impact.

Other substances which were found sporadically in the samples were tetramethylbenzene, methylcyclohexane, and 2-butoxyethanol. The first two are possible by-products of combustion processes (Ballesteros et al. 2015) which, considering the geographic location and characteristics of the site, probably derive from vehicle exhaust emissions. Instead, 2-butoxyethanol is an indoor air pollutant deriving from the use of cleaning products, specifically glass-cleaning and degreasing products (Paciência et al. 2019). Indeed, this glycol ether was found in combination with toluene, pinene, limonene, and xylenes in the analysis of a typical glass cleaner (Rella et al. 2012). All these species were detected in the school's indoor spaces, further supporting the idea of a common source deriving from afternoon cleaning activities. Based on these considerations, it is highly probable that the two large peaks in concentration are also due to the effect of afternoon cleaning activities. Both peaks occur on a Friday evening and stretch throughout the night. The high concentrations observed could be due to a combination of factors, such as more intense and thorough cleaning prior to the weekend, coupled with the complete closure of the school which facilitates indoor accumulation of the volatile organic compounds.

Table 4 Additional substances detected in the chromatograms using SCAN analysis

Sample	Location	Substances detected
985 QU	B2	D-Limonene, α-pinene, tetramethylbenzene
984 QU	B1	D-Limonene
982 QU	O	D-Limonene
983 QU	4B	D-Limonene, tetramethylbenzene, methylcyclohexane
987 QU	B2	D-Limonene
986 QU	B1	D-Limonene, 2-butoxyethanol, tetramethylbenzene
989 QU	O	D-Limonene
988 QU	4B	D-Limonene, α-pinene, methylcyclohexane
993 QU	B2	D-Limonene
992 QU	B1	D-Limonene
991 QU	O	D-Limonene, tetramethylbenzene
990 QU	4B	–
996 QU	B2	D-Limonene
995 QU	B1	D-Limonene, α-pinene, tetramethylbenzene
994 QU	O	D-Limonene
Q 832 I	4B	D-Limonene
998 QU	B2	D-Limonene, tetramethylbenzene
997 QU	B1	D-Limonene
999 QU	4B	D-Limonene, 2-butoxyethanol
Q 836 I	B2	D-Limonene
Q 849 I	B1	D-Limonene
Q 834 I	O	D-Limonene
Q 835 I	4B	D-Limonene
Q 792 I	B2	D-Limonene
Q 793 I	B1	D-Limonene
Q 795 I	O	D-Limonene
Q 794 I	4B	D-Limonene, α-pinene, tetramethylbenzene

Discussion

Several studies in the last years have highlighted possible links between exposure to high concentrations of VOCs and different types of illnesses, ranging from nose and eye irritation to different forms of cancer (Rumchev et al. 2007). The exposure to VOCs can be particularly harmful for children, whose respiratory and immune systems are still under development (Rivas et al. 2014). In addition, children spend most of their time inside school buildings, and because VOCs tend to accumulate preferentially in indoor environments, the exposure to this class of compounds is a topic of great interest and concern. Moreover, due to the lack of limits and regulations on the indoor concentrations of VOCs, both as a whole (TVOCs) and on individual compounds (e.g., benzene and toluene), many of these sites remain essentially unmonitored.

The concentrations observed in this study (both TVOC and single compounds) were below the values observed in numerous other similar studies (Al-Awadi 2018; Sofuoglu et al. 2011; Bertoni et al. 2002). Unfortunately, comparisons cannot be made directly with all the analyzed species since literature data are present only for BTEX and few other gases (e.g., formaldehyde), whereas information on the concentrations of other VOCs is lacking. In fact, most of the values reported in the literature regard the compounds of the greatest health concern, such as benzene, or those typically found in higher concentrations, such as toluene and xylenes. Few data can be found regarding the concentrations of other volatile organic compounds in schools, such as the alkylated derivatives of benzene that were detected in the present study. However, the concentrations of all individual VOCs and average TVOCs were significantly below the values considered harmful to human health (Rumchev et al. 2007; Goodman et al. 2017). This being true, as highlighted in the WHO Guidelines for indoor air quality, no safe level of exposure can be recommended for dangerous compounds such as benzene, and concentrations should be reduced as much as possible (WHO 2010). Indeed, benzene concentrations below the legal limit of 5 μg m^{-3} are still associated with excess lifetime risks of 1/100,000 (1.7 μg m^{-3}) and 1/1,000,000 (0.17 μg m^{-3}). This compound was not detected in Squinzano; however, average concentrations in Galatina were between 1 and 3 μg m^{-3} in several indoor environments (Ielpo et al. 2021).

Moreover, indoor school areas represent unique environments with a potential need for different VOC concentration limits. In a similar study, Madureira et al. (2015) found that, despite concentrations of individual VOC and median TVOC being much lower than the recommended value proposed by different internationally accepted guidelines, TVOC, toluene, and *o*-xylene exposure levels were associated with episodes of wheeze in children. This shows that children may suffer from adverse health effects even if the concentrations remain below the recommended values and also highlights the importance of carrying out continuous monitoring in parallel to passive sampling, since concentrations may vary significantly and average values may not be the best parameters to assess air quality conditions and their implications on human health. In fact, continuous monitoring of TVOCs in Squinzano allowed us to recognize several peaks in concentrations during weekdays, related to specific indoor activities and student occupancy. The concentrations reached in these peaks were often above the guidelines suggested for a harmless environment. Indeed, most literature studies on this topic focus either on the measurement of TVOCs (Mundackal and Ngole-Jeme 2022; Bayani et al. 2021; Liu et al. 2022a), or on the quantification of individual VOCs (De Gennaro et al. 2013; Lucialli et al. 2020; Villanueva et al. 2018), whereas case studies in which both are carried out simultaneously are rarer (Hu et al. 2020; Al-Awadi 2018). As occurred in this study, the fact that the peaks in TVOC concentration were not fully accounted for by the levels of the individual VOCs is an indication that the latter approach is the most complete and should be carried out in order to have a clear picture of indoor air quality, even more so considering that both TVOC levels and the concentration of harmful individual VOCs have been used as parameters to assess indoor air quality.

The simultaneous presence of all or most BTEX in the samples suggests the existence of sources common to them. The most common ones are vehicular traffic, biomass burning, industrial emissions, cigarette smoke, and solvent use (Zhang et al. 2021; Sarigiannis et al. 2011); however, the specific nature of the sources and the concentration of the VOCs can vary greatly from site to site. In this case, it is most probable that the geographical location of the two schools influenced the overall emission profile. Indeed, many studies show a correlation between the degree of urbanization and the level of air contamination by VOCs (Gilli et al. 1994). The school in Galatina is located in an industrial area and therefore more affected by industrial and traffic emissions, which are known emitters of both benzene and toluene. Another source of emission in Galatina was tobacco smoke, which contributed particularly to the enhanced levels of benzene in the classrooms and outdoors. The same effect was not recorded in Squinzano, and the suburban location of this site, away from the major sources of pollutant emission, may explain the absence of high concentrations of VOCs, such as benzene and toluene. Compared with typical values of greater urban areas (Carrer et al. 2000), the concentrations observed in both sites were lower.

With regard to the halogenated VOCs, another similar study (Al-Awadi 2018) found concentrations at least an order of magnitude higher than in the present work, mainly due to indoor sources, such as the use of perfumes, hand sanitizers, solvents, reagents in laboratories, air fresheners, and other common applications. In the same study, it was also evident that the variability in the concentrations of these VOCs is very high and is closely related to the presence of specific sources. The detected levels of chloroform and carbon tetrachloride in the bathrooms of Squinzano suggest the use of specific solvents and/or cleaning products. However, the presence of carbon tetrachloride in similar concentrations in the outdoor environment indicates an outdoor source, at least for this halogenated VOC, despite the fact that Squinzano is not close to any of the typical emitters of carbon tetrachloride, such as the petrochemical industry (Dumanoglu et al. 2014). Considering the very low values of the detection and quantification limits associated with the method and in light of the previous considerations on the overall VOC concentrations, it is possible to conclude that the use of such products does not represent a particular concern for the environments studied.

Regarding the I/O ratios, the difference between the two sites could be due to the different sampling periods. In Squinzano, sampling was carried out during spring (13 May 2019–3 June 2019), whereas in Galatina, it was performed during winter (1 February 2019–12 March 2019). We outlined that the choice of the sampling periods was indicated by internal school management. Due to the lower temperatures and worse weather conditions in wintertime, air exchange rates are higher in spring than in winter, allowing for greater circulation between the indoor and outdoor environments. Indeed, the I/O ratios recorded in Squinzano for all the major VOCs indicate similar indoor and outdoor concentrations, suggesting a good air exchange and no visible impact of indoor sources on air quality. Conversely, except for benzene (I/O = 0.6), in Galatina all the major VOCs were associated with higher indoor concentrations due to penetration from outdoors and subsequent accumulation, presence of specific indoor sources, or both.

Despite not being able to perform sampling during the same months in both schools, it was possible to highlight the significant impact of the external environments on indoor type and concentration of volatile organic compounds. Emblematic is the case of benzene, which was emitted by several outdoor sources in Galatina, and high concentrations were found also indoors, whereas the absence of outdoor sources in Squinzano was reflected in indoor concentrations below the limit of detection. At the same time, the different trends in TVOC concentrations in the classrooms highlighted the impact of indoor sources and activities. A lower number of pupils attending the Squinzano school could be one of the reasons behind lower I/O ratios with respect to Galatina. Moreover, the difference in the type of VOCs detected indoors (chlorinated species in Squinzano) is also due to site-specific indoor sources.

Building on the results of this study, the necessity to carry out more studies on the air quality inside schools remains. First, because the number of published literature works in this field is still scarce and in order to obtain a clear and representative picture of the air quality (in terms of VOCs) in Italian and European schools, more studies need to be conducted. In fact, as already highlighted, the nature and concentration of volatile organic compounds are highly dependent on the site under investigation and can vary greatly between different areas. This may be particularly the case for Italian schools because the geographical landscape varies significantly between the northern and southern parts of the country, along with the number and types of emission sources. However, the results of this study provide additional data in terms of the type and concentration of VOCs in school environments and may also represent a point of reference for future similar studies, both at the national and international scale.

Moreover, another further development of this work may include the study of a larger number of VOCs, such as oxygenated species. In fact, despite the concentrations of TVOC being comparable between the two sites, the concentrations of the specific species often varied, benzene being the most evident case. Moreover, investigating the presence of a wider range of substances may be useful to determine the nature of the species responsible for the peaks in indoor TVOC concentrations observed. Given the large differences in reactivity, volatility, and chemical structure, in order to perform a complete speciation of airborne VOCs, it is necessary to use more than one type of sampling cartridge in order to selectively trap all of the species. In this regard, thermal desorption is preferred to chemical desorption in the qualitative analysis due to advantages in terms of sensitivity, reduced loss of signals, and reduced interferences. Finally, given that the toxicity of volatile organic compounds varies greatly among the different species, being able to determine the concentration of a larger number of substances is essential in order to fully assess the air quality within the schools.

Conclusions

A comparison of the indoor and outdoor concentrations of VOCs in two different schools of the Apulia region (southern Italy) was performed in this study. Real-time monitoring of TVOC concentrations was carried out, along with the sampling and analysis of 20 volatile organic compounds. Overall, the results show good air quality, particularly at the Squinzano school, in terms of VOCs, with average concentrations below levels considered harmful to human health. For the school

in Galatina, there remains a criticality for benzene, which, although below legal limits, has values of attention considering the young age of the students. Moreover, peaks in the concentration of TVOC above levels considered harmless were observed in both cases, strictly related to specific indoor activities. Differences between the two monitored environments were due to both indoor activities and sources and the characteristics of the surrounding outdoor environment. Specific activities, such as cigarette smoking in the classrooms of Galatina or solvent use in the bathrooms of Squinzano, were instrumental in shaping the indoor profile of VOCs in both schools. At the same time, the differences in the surrounding environment, with Squinzano located in a more rural and less industrialized area, were equally significant in affecting the concentrations of volatile organic compounds, especially BTEX.

The importance of this work stems from the fact that, to the knowledge of the authors, the topic of VOCs in school environments has not been extensively studied in scientific literature. Moreover, almost all these sites are unmonitored due to the lack of specific regulations limiting the indoor concentrations of volatile organic compounds. However, before introducing internationally accepted legislation, it is important to assess the air quality conditions of the widest possible pool of schools. In this regard, the data collected in this study represents a step forward; particularly, the quantitative analysis of single compounds may also represent an important point of reference for future studies. In fact, despite other studies being conducted in this field, the literature data highlighted a large variability between countries and between schools located in areas with a similar background, reinforcing the concept of the site-specific nature of these pollutants. Therefore, it is important to be able to conduct the highest number of studies in order to obtain a clear picture of the overall air quality conditions.

Hopefully, this work will pave the way toward a more complete and in-depth analysis of the air quality within schools, aimed at safeguarding children's health. Further developments of this study will include conducting another monitoring campaign with the use of a newly purchased high-sensitivity VOC monitoring station capable of measuring carbon dioxide, in order to confirm the impact of student occupancy on the levels of TVOCs (morning peaks). Moreover, the speciation of a larger number of VOCs will be carried out, focusing on other classes, such as oxygenated species, which have been detected in other studies and that may explain the afternoon peaks in concentrations observed with the PIDs.

Author contribution The project was conceptualized by PI, CM, and PF. The methodology was developed by PI, CM, and AG, who also performed on-site sampling. Formal analysis was carried out by SG, whereas data curation and elaboration were performed by AB, under the supervision of PF and VC. The original draft was written by AB and then revised and edited by all the other authors.

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Data availability This manuscript has no associated data.

Declarations

Competing Interests The authors declare no competing interests.

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