Macedonian Journal of Chemistry and Chemical Engineering, Vol. **43**, No. 2, pp. xx–xx (2024)

MJCCA9 –

ISSN 1857-5552
e-ISSN 1857-5625

Received: September 1, 2024
Accepted: November 27, 2024

DOI: 10.20450/mjcce.2024.2953
Original scientific paper

ASSESSMENT OF VOLATILE ORGANIC COMPOUNDS IN INDOOR ENVIRONMENTS ACROSS NORTH MACEDONIA AND KOSOVO USING PASSIVE SAMPLING*

Vllaznim Mula[1,2*], Jane Bogdanov[2], Jasmina Petreska Stanoeva[2], Lulzim Zeneli[1*], Avni Berisha[3], Zoran Zdravkovski[2*]

[1]*University "Fehmi Agani" 50000 Gjakova, Kosovo*
[2]*Institute of Chemistry, Faculty of Natural Sciences and Mathematics, Ss. Cyril and Methodius University in Skopje, 1000 Skopje, N. Macedonia*
[3]*Department of Chemistry, Faculty of Mathematics and Natural Sciences, University of Prishtina, 10000 Prishtina, Kosovo*

vllaznim.mula@uni-gjk.org; lulzim.zeneli@uni-gjk.org; zoran@ukim.edu.mk

Humans are exposed to various volatile organic compounds (VOCs) through ingestion, inhalation, or skin contact, potentially leading to adverse health consequences. In this study, the distribution of various VOCs in indoor air in North Macedonia and Kosovo was investigated for the first time. From March to December 2023, the monthly relative abundance of VOCs was monitored using Radiello® passive/diffusive samplers, followed by gas chromatography–mass spectrometry (GC-MS) analysis.

A total of 60 samples were collected from 17 sites, where over 70 individual VOCs belonging to several classes were identified. Benzene, toluene, ethylbenzene, and xylenes (BTEX) compounds were the most abundant in the chemical stockroom, contributing up to 77.84 % of the total VOC relative abundance. In the printing store/workplace, alkylbenzenes were predominant, comprising 69.72 %. Alcohols and ethers were primarily detected in the coffee shop, whereas organosiloxanes were dominant in the hair salon, accounting for 33.87 % and 54.46 % of the total VOCs, respectively. Terpenes were a significant component in the meat factory, representing 65.78 % of the total VOCs, while hydrocarbons were most prevalent in the oil & lubricants store, accounting for 58.23%. Halogenated compounds were primarily found in the superstore, constituting 20.42 % of total VOCs. The VOC composition results indicate that their presence in indoor environments may pose both acute and chronic health risks.

Keywords: volatile organic compounds; indoor air; air pollution; passive sampling; GC-MS

ПРОЦЕНА НА ИСПАРЛИВИТЕ ОРГАНСКИ СОЕДИНЕНИЈА ВО ЗАТВОРЕНИ ПРОСТРИИ ВО СЕВЕРНА МАКЕДОНИЈА И КОСОВО СО ПРИМЕНА НА ПАСИВНО ЗЕМАЊЕ ПРИМЕРОЦИ

Луѓето се изложени на голем број испарливи органски соединенија (VOCs) преку храната, вдишувањето или контактот со кожата, што потенцијално може да доведе до негативни здравствени последици. Во оваа студија, за прв пат е испитана дистрибуцијата на различни VOCs во воздухот во затворени простории во Северна Македонија и Косово. Релативната застапеност на различни класи испарливи органски соединенија беше следена во периодот од март до декември 2023 година. Примероците беа земани месечно, со примена на пасивни примеркувачи од марката Radiello®, а потоа анализирани со гасна хроматографија спрегната со масена спектрометрија (GC-MS).

* Dedicated on the occasion of the Golden Jubilee of the *Macedonian Journal of Chemistry and Chemical Engineering.*

Во текот на студијата беа собрани вкупно 60 примероци од 17 локации, при што беа идентификувани над 70 соединенија групирани во неколку класи. Соединенијата од типот на бензен, толуен, етилбензен и ксилени (BTEX) беа најзастапени во складиште со хемикалии, придонесувајќи со 77,84 % од вкупната содржина на VOCs. Во печатницата доминираа алкилбензените, со застапеност од 69,72 %. Алкохолите и етерите беа детектирани во примероците собрани во кафетерија, додека органосилоксаните беа доминантни во фризерски салон, со релативна застапеност од 33,87 % и 54,46 %, соодветно. Терпените беа најприсутни во фабрика за месо и сочинуваа 65,78 % од вкупната содржина на VOCs, додека јаглеводородите доминираа во складиште за нафта, со застапеност од 58,23 %. Халогенираните соединенија главно беа детектирани во хипермаркет, каде сочинуваа 20,42 % од вкупната содржина на VOCs.

Резултатите од анализата на VOC покажуваат дека нивното присуство во затворени простории е значајно, што може да претставува потенцијален здравствен ризик.

Клучни зборови: испарливи органски соединенија, воздух во затворени простории; загадување на воздух; пасивно примеркување; GC-MS

1. INTRODUCTION

Indoor air pollution presents a critical global challenge, especially in developing nations such as North Macedonia and Kosovo. With individuals spending approximately 90% of their time indoors, they are frequently exposed to elevated levels of pollutants, including volatile organic compounds (VOCs).[1–4]

Previous research has demonstrated that indoor air can contain VOC concentrations up to ten times higher than the air found outdoors.[5–9] The World Health Organization (WHO) estimates that exposure to air pollution is responsible for millions of deaths annually, ranking as the fourth most significant global health risk factor.[10]

Exposure to VOCs in indoor environments has been linked to various acute and chronic health effects. These effects include sick building syndrome (SBS), allergies, respiratory issues, asthma, bronchitis, cardiovascular disease, leukemia, cancer, and severe physical and mental health complications.[5,11–13]

Previous research, such as the study by Yingjie Li et al., titled "Profiles and monthly variations of selected volatile organic compounds in indoor air in Canadian homes: Results of Canadian national indoor air survey 2012–2013," highlighted the dominance of six VOCs: limonene, decamethylcyclopentasiloxane, toluene, hexanal, nonanal, and α-pinene. These compounds were detected in the highest concentrations, collectively accounting for 54 % of the total VOCs monitored across 3,524 Canadian residential homes.[14]

Similarly, a study by Bin Yuan et al., titled "Source profiles of volatile organic compounds associated with solvent use in Beijing, China," revealed that toluene and C_8 aromatics were the predominant species, constituting 76 % of the total VOCs emitted during paint applications. Additionally, the analysis of printing emissions identified heavier alkanes and aromatics, including *n*-nonane, *n*-decane, *n*-undecane, toluene, and *m*/*p*-xylene, as the dominant compounds.[15]

These studies highlight the variability in VOC composition across different indoor settings and geographical locations, reflecting the diverse sources contributing to indoor air pollution. Primary contributors, as identified in various studies, include construction materials, furnishings, paints, glues, tobacco smoke, cleaning products, carpets, textiles, personal care products, solvents, floorings, and cooking, among others.[11–13]

Despite these concerning statistics, there is a significant lack of scientific data on indoor air quality, particularly regarding VOCs in North Macedonia and Kosovo. To address this gap, a specialized study was conducted to investigate the distribution and characteristics of VOCs across various indoor environments.

Several techniques and methodologies are available for assessing VOC distribution in indoor environments. This study adopted a comprehensive approach based on internationally recognized standards, including ISO 16000-5:2007 and the American Society for Testing and Materials (ASTM) D6196-03.[16,17] Using Radiello® passive/diffusive samplers combined with gas chromatography-mass spectrometry (GC-MS) analysis, VOC distribution was measured across diverse indoor environments in North Macedonia and Kosovo.[18]

The findings of this study offer valuable insights into the indoor air quality in North Macedonia and Kosovo through the analysis of a wide range of VOCs. This research contributes significantly to global discussions on indoor air quality, highlighting the complexity of the VOC landscape.

The diversity of the VOCs identified underscores the importance of developing a comprehensive approach to formulating effective strategies for managing and improving indoor air quality in these regions. These insights are crucial for policymakers tasked with safeguarding indoor air quality in these regions, highlighting the global relevance of addressing this crucial issue.

2. EXPERIMENTAL SECTION

2.1. *Sampling sites*

The study was conducted across various indoor environments in North Macedonia and Kosovo. Sampling sites were carefully selected to represent a range of indoor environments, including educational, industrial, commercial, and residential settings. Consideration was given to factors such as the strategic placement of passive samplers, by avoiding direct proximity to emission sources, optimizing air circulation, by maintaining unobstructed airflow, and ensuring both security and accessibility. Prior approval was obtained from the inhabitants or owners of each site, in accordance with ethical guidelines and with respect for property rights.[19]

From March to December 2023, a total of 60 indoor air samples were collected from 17 diverse sites, including a coffee shop, textile factory, superstore, and other establishments (see Table 1). For each 28-day sampling interval, a new adsorbing cartridge was used to prevent cross-contamination and to ensure accurate VOC measurements.

T a b l e 1

Description of indoor air sampling locations

Location	**Type of environment**	**Description**
North Macedonia	Coffee shop	Urban area, smoking is permitted
Kosovo	Textile factory	Urban area, a sampler is situated within the fabric washing section.
North Macedonia	Superstore	Urban area, a sampler is situated within the washing and cleaning products section of the superstore.
Kosovo	Oil & lubricants store	Urban area, a sampler is located in the store's stockroom.
North Macedonia	Residential settings	Urban area, one sampler is placed in the living room of an apartment, and another is located in the living room of a house.
North Macedonia	Organic laboratory and chemical stockroom	Urban area, one sampler is situated in the organic laboratory, and another is located in the chemical stockroom of the institute.
Kosovo	Meat factory	Suburban area, the sampler is located within the sausage processing and drying section of the factory.
Kosovo	Hair salon	Urban area, a women-only hair salon. A sampler is located in the center of the salon.
Kosovo	Kebab shop	Urban area, using charcoal grill. A sampler is located in the center of the kebab shop.
Kosovo	School	Urban area, one sampler is situated in a classroom, and another is located in the main hall of the primary school.
Kosovo	Electrical store	Urban area, small store selling electrical supplies. A sampler is situated in the center of the store.
Kosovo	Printing	Urban area, one sampler is located in the store, and another is in the windowless basement of the workplace.
Kosovo	Perfume shop	Urban area, a sampler is located in the shop within the shopping center.

2.2. *Sampling methods and sample preparation*

The Radiello® passive sampler (RAD 130), developed by Fondazione Salvatore Maugeri in Padova, Italy, consists of a cylindrical adsorbing cartridge housed within a white diffusive body. This design allows VOCs to be sampled by diffusion across a microporous polyethylene membrane.[18]

This choice stems from the sampler's demonstrated efficacy in capturing a broad spectrum of indoor VOCs, as well as its user-friendly operation, minimal maintenance requirements, and effective use of activated charcoal. The charcoal boasts an extensive surface area and intricate structure, ideal for adsorbing compounds pertinent to our research. Furthermore, its pump-free and electricity-independent operation aligns perfectly with the needs of our study.[18,19]

Endorsed by the European Union, the Radiello® sampler follows approved methodologies, ensuring comparability with other passive sampling techniques and establishing its status as a reference sampler.[20]

Passive/diffusive samplers were strategically positioned indoors at the center of each monitoring site, at heights ranging from approximately 1.5 meters to 3 meters, to effectively capture variations in VOC concentrations and account for diverse sources.

After completing the sampling process, each adsorbent-loaded cartridge was securely sealed in a glass tube and transported to the laboratory. The samples were then stored at 4 °C for up to one week before undergoing extraction and GC-MS analysis. For desorption, 2 ml of dichloromethane was added to the glass tube containing the adsorbent, with 100 µl of anisole as an internal standard. The mixture was allowed to sit at ambient temperature for 30 minutes with occasional shaking to facilitate desorption. After extraction, the extract was transferred to a vial and analyzed by GC-MS.[19]

Based on previous research, carbon disulfide was replaced with dichloromethane for extraction, and anisole was selected as the internal standard for its ability to produce a stable and distinct peak in GC-MS analysis, without interference from the target analytes. Its consistent reference signal enabled the determination of the relative abundance of VOCs, which is particularly useful since passive sampling methods, like those used in our study, do not provide absolute concentrations. Consequently, our analysis focuses on the relative ratios of individual VOCs, reflecting their comparative abundance within the sample. This approach is essential given the limitations of passive sampling in determining absolute VOC concentrations.[19,21,22]

Using relative abundance with an internal standard offers several advantages for assessing VOC composition. One key advantage is its simplicity and cost-effectiveness. This approach does not require expensive equipment or standards, making it a practical and affordable option for large-scale studies or long-term monitoring of VOCs. Furthermore, it enables efficient and rapid analysis, particularly when assessing the relative distribution of VOCs over time, which is essential for understanding their presence and variability in indoor air.

Another advantage of passive diffusion is the ability to conduct continuous and long-term monitoring of VOCs. Since passive samplers can be deployed over extended periods without requiring frequent maintenance or calibration, they serve as an excellent tool for monitoring VOC distribution across various indoor environments. This is particularly useful for capturing fluctuations in VOC composition, which may vary over time or with changing environmental conditions. The use of an internal standard also enhances the precision of relative comparisons between compounds, providing a clear indication of the relative abundance of different VOCs within the sample. Moreover, this method is non-intrusive and can be easily implemented in hard-to-reach areas, further supporting its use for environmental monitoring.

However, there are some limitations to this approach. The primary disadvantage is that the method provides relative quantification rather than absolute concentration values. While this allows for comparison between different VOCs within a sample, it does not offer precise concentration measurements, which may be necessary for regulatory purposes or detailed risk assessments. Another limitation is that the accuracy of the method can be influenced by environmental factors such as temperature, humidity, and airflow, which may impact the absorption rate of VOCs into the passive sampler. Additionally, this method may not capture all VOCs, particularly those with very low volatility, which could limit the comprehensiveness of the data.

2.3. *GC-MS analysis*

The analyses of VOCs were conducted using an Agilent 6890N gas chromatograph coupled with a single quadrupole 5975B mass selective detector, employing split injection and the internal standard method. The system employed an HP-5MS capillary column (30 m × 250 µm, film thickness 0.25 µm) coated with a nonpolar stationary phase of (5%-phenyl)-methylpolysiloxane. Helium served as the carrier gas, with a constant flow rate of 1 ml/min. The temperature program for the gas chromatograph began at 35 °C (held for 5 minutes), then gradually increased to 90 °C at a rate of 5 °C/min, holding for 3 minutes. The temperature was further ramped up to 280 °C at 10 °C/min for 2 minutes, followed by a final ramp to 300 °C at 5 °C/min. The injector temperature was maintained at 250 °C, with an injected volume of 2 µl and a split ratio of 1:2. For mass spectrometry, the MS source temperature was set to 230 °C, the quadrupole temperature to 150 °C, and the mass-to-charge ratio (*m/z*) scan range was 35 – 500 amu. Each analysis had a total runtime of 40 minutes.[19]

The samples were analyzed using Agilent MassHunter v.10.0 software, with compound identification based on the NIST Library of Mass Spectra database (NIST MS Search 2.0), using probability values above 85 %. The relative abundance of each individual VOC in indoor air was calculated by dividing the surface area of each compound by the total surface area of all compounds. The resulting quotient was then multiplied by 100 % to express the relative abundance as a percentage.

3. RESULTS AND DISCUSSION

The findings presented here highlight the diverse range of VOCs identified in the monitored indoor environments. Our study systematically categorizes these compounds into specific categories, including BTEX, alkylbenzenes, alcohols and ethers, aldehydes and ketones, organosiloxanes, terpenes, hydrocarbons, esters, halogenated compounds, and miscellaneous compounds. This classification not only improves our understanding of the sources and composition of indoor air pollutants but also facilitates more focused discussions and analyses, ultimately contributing to a deeper understanding of indoor air quality issues.

Figures 1–3 present representative chromatograms of VOCs detected in various indoor environments in North Macedonia and Kosovo. These chromatograms illustrate common indoor air pollution profiles, highlighting the main compounds identified in these settings. The data provide a foundation for comparing VOC distribution across different indoor environments and offer insight into the factors contributing to indoor air pollution.

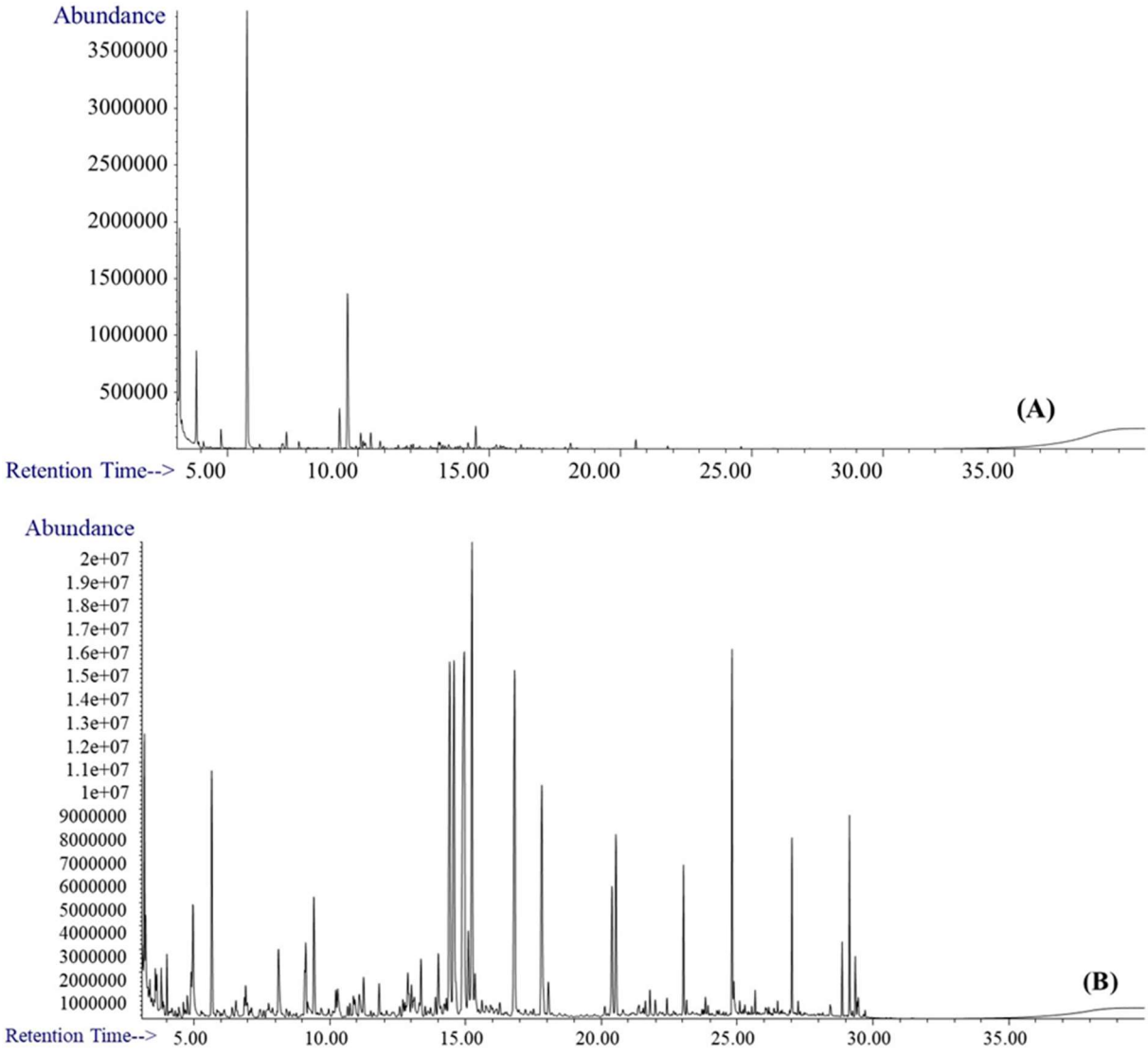

Fig. 1. Representative VOC chromatograms from various indoor environments in North Macedonia and Kosovo. **(A)** Chemical stockroom. **(B)** Coffee shop.

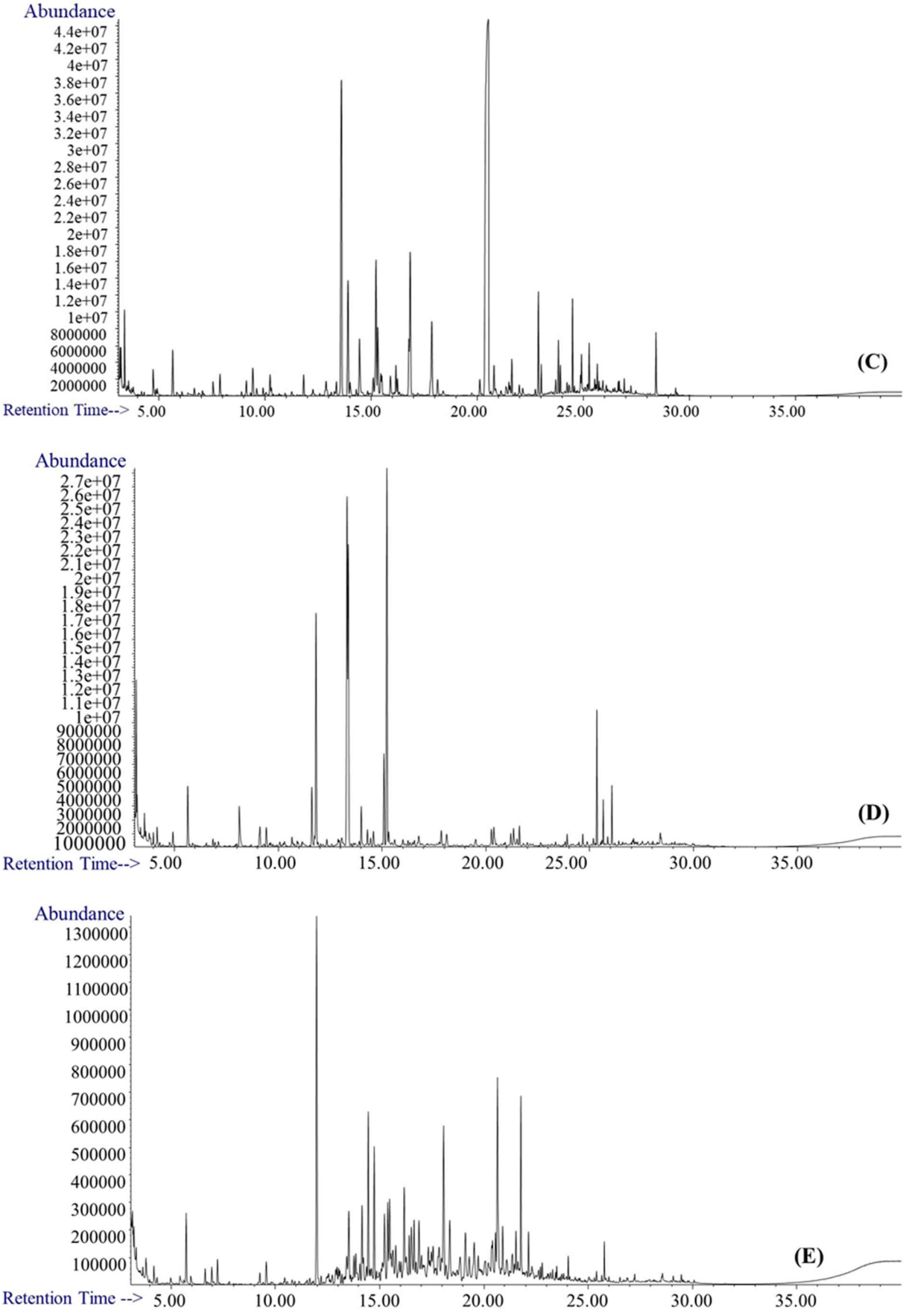

Fig. 2. Representative VOC chromatograms from various indoor environments in North Macedonia and Kosovo. **(C)** Hair salon. **(D)** Meat factory. **(E)** Oil & lubricants store.

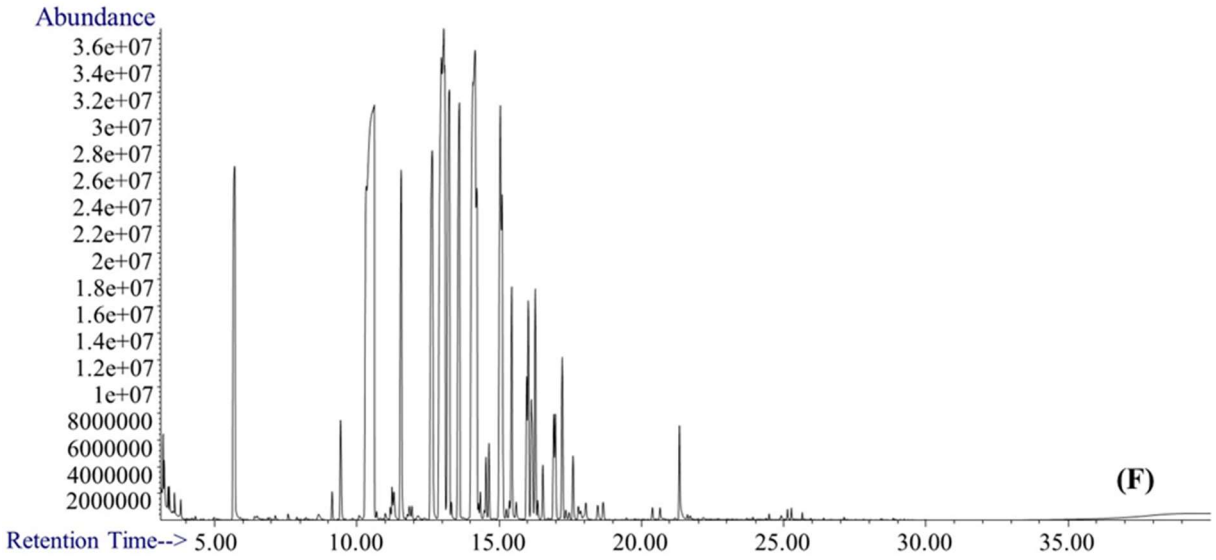

Fig. 3. Representative VOC chromatograms from various indoor environments in North Macedonia and Kosovo. **(F)** Printing store – workplace.

Figures 4 and 5 provides a comprehensive overview of the average relative abundance of the groups of VOCs detected across all indoor environments monitored. The analysis presented in these figures serves as the foundation for the detailed results and discussions that follow, offering deeper insights into these findings.

Previous research conducted in various countries has investigated the composition of VOCs in indoor environments across diverse settings. A particular focus has been placed on several categories of VOCs: **aromatic hydrocarbons** (e.g., BTEX, various trimethylbenzene isomers, and n-propylbenzene); **alkanes** (e.g., *n*-pentane, *n*-hexane, *n*-heptane, *n*-decane, *n*-dodecane, *n*-tetradecane, methylcyclopentane, and cyclohexane); **alcohols** (e.g., propan-1-ol, butan-1-ol, 2-methylpropan-1-ol, and 2-ethylhexan-1-ol); **terpenes** (e.g., α-pinene, β-pinene, *d*-limonene, and camphene); **aldehydes** (e.g., formaldehyde, acetaldehyde, butanal, and benzaldehyde); **ketones** (e.g., acetone and butan-2-one); **volatile methyl siloxanes** (e.g., hexamethyldisiloxane (L2), decamethyltetrasiloxane (L4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6)); **halogenated compounds** (e.g., chlorobenzene, 1,3-dichlorobenzene, 1,2-dichloroethane, and tetrachloromethane); **glycol/glycol ether** (e.g., 1-methoxypropan-2-ol, 2-butoxyethanol, and 2-phenoxyethanol); **heterocyclic compounds** (e.g., tetrahydrofuran and 2,5-dimethylfuran), among others.[13] Most of these pollutants were detected in the indoor air monitoring locations, as indicated by the results of the study presented below.

In this study, varying relative abundances of **BTEX** compounds (benzene, toluene, ethylbenzene, and xylenes) were observed in all indoor air-monitored locations. The chemical stockroom (77.84%), the organic laboratory (45.92%), the electrical store (38.91%), and the textile factory (32.52%) exhibited the highest concentrations of BTEX. Among the BTEX components, toluene and xylenes were found to have the highest percentages in the observed locations.

In the printing store, **alkylbenzenes** accounted for the highest relative abundance among the total VOCs, comprising 34.83 %. In contrast, this proportion increased significantly to 69.72% in the windowless basement workshop. Key compounds identified within this group included 1,3,5-trimethylbenzene, 1-ethyl-2-methylbenzene, 1-ethyl-4-methylbenzene, propylbenzene, isopropyl-benzene, and 2-ethyl-1,4-dimethylbenzene, among others.

The highest relative abundance of **alcohols and ethers** were found in the coffee shop (33.87 %), followed by the kebab shop (20.86 %), the perfume shop (15.67%), and the printing store (11.26 %). Commonly detected compounds in this group included 1-methoxypropan-2-ol, 1-(2-methoxy-1-methylethoxy)propan-2-ol, 3,7-dimethyloctan-3-ol, 1-(2-methoxypropoxy)propan-2-ol, 2,6-dimethyloct-7-en-2-ol, and 1-octen-3-ol, among others.

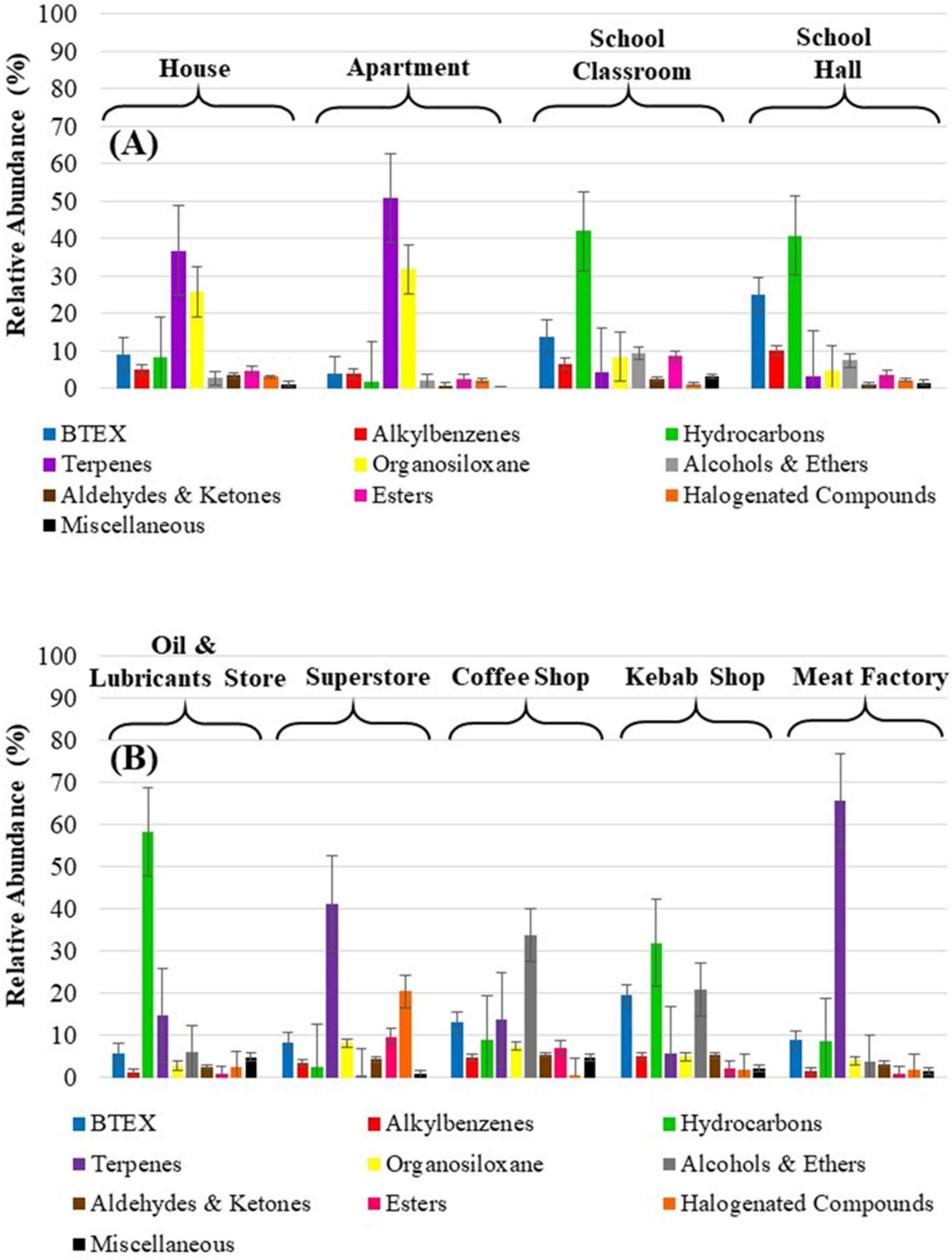

Fig. 4. The average relative abundance (%) of all VOC categories from sampling locations during March to December 2023:
(A) House, apartment, school classroom, and school hall;
(B) Oil & lubricants store, superstore, coffee shop, kebab shop, and meat factory.

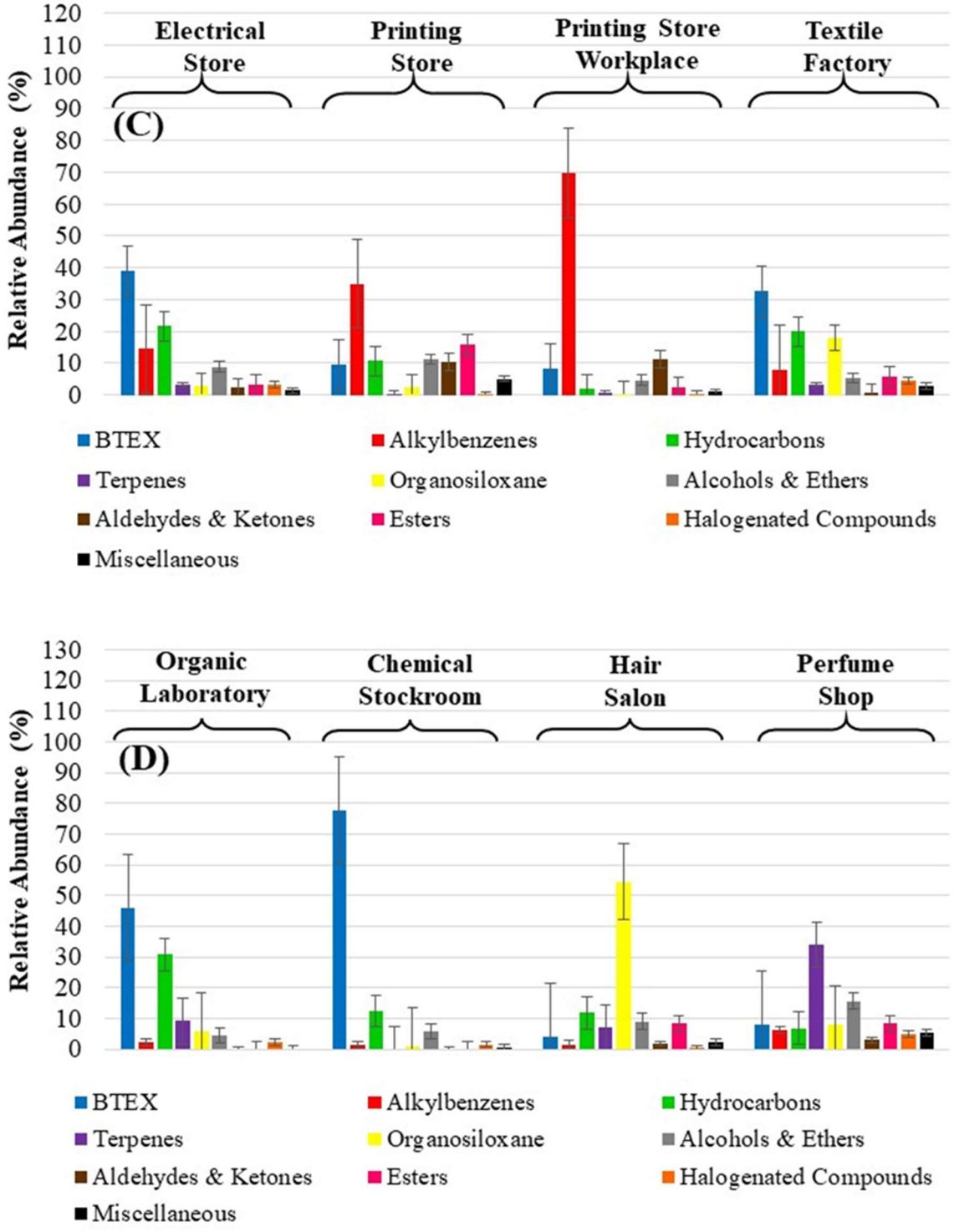

Fig. 5. The average relative abundance (%) of all VOC categories from sampling locations during March to December 2023:
(C) Electrical store, printing store, printing store-workplace, and textile factory;
(D) Organic laboratory, chemical stockroom, hair salon, and perfume shop

Aldehydes and ketones were detected in all monitored locations, with relative abundances ranging from 0.71 % to 11.22 %, except in the organic laboratory and chemical stockroom. Commonly detected compounds in this group included cyclohexanone, 1-(3-methylphenyl)ethenone, furan-2-carbaldehyde, 6-methylhept-5-en-2-one, hexanal, nonanal, and octanal, among others.

The hair salon displayed the highest relative abundance of **organosiloxane** compounds, reaching 54.46 %. The apartment setting followed closely with a relative abundance of 31.78%, while the

house showed 25.78 %, and the textile factory had 17.82 %. Commonly detected compounds in this group included decamethylcyclopentasiloxane (D5), octamethylcyclotetrasiloxane (D4), octamethyltrisiloxane, dodecamethylpentasiloxane, and methyltris(trimethylsiloxy)silane, among others.

Terpene compounds exhibited the highest relative abundance in the following settings: meat factory (65.78 %), apartment (50.88 %), hypermarket (41.33 %), house (36.84 %), and perfume shop (34.01 %). Notable compounds within this group commonly detected in the monitored indoor air included limonene, α-pinene, β-pinene, phellandrene, eucalyptol, linalool, and 3-carene, among others.

In the oil & lubricants store, **hydrocarbons** dominated, accounting for 58.23 % of the total relative abundance, a level higher than those observed in other areas. This category included a diverse range of hydrocarbon compounds, from those with low carbon numbers to those with chains exceeding 10 atoms, excluding aromatic hydrocarbons from this classification.

Esters were observed in all sampled locations, with relative abundances ranging from 0.88 % to 15.91 %, except in the organic laboratory and the chemical stockroom. Notable compounds in this group included 2-(2-butoxyethoxy)ethyl ethanoate, ethyl 5-oxotetrahydrofuran-2-carboxylate, 2-butoxyethyl ethanoate, methyl benzoate, butyl ethanoate, diethyl phthalate, butyl butanoate, among others.

The superstore exhibited the highest relative abundance of **halogenated compounds**, accounting for 20.42 % of the total identified VOCs. Notably, 1,3-dichlorobenzene alone contributed 18.73 % to the overall VOC composition in the superstore. Commonly detected halogenated compounds in the monitored indoor air included 1,3-dichlorobenzene, 1,4-dichlorobenzene, 2-chloro-2-methylbutane, 4-bromoheptane, 1-chlorooctane, 4-chloroheptane, among others.

In all monitored indoor air locations, the most frequently detected compounds within the **miscellaneous** group included 2,5-dimethylfuran, pyridine, 2-methylbutanenitrile, 2,6-dimethylpyrazine, and other unidentified VOCs.

A complete list of all VOCs detected in indoor air, along with their semi-quantitative data (peak area) and relative abundances for all samples, is provided in the **supplementary table**.

Based on the available data, as well as our monitoring results, it is evident that indoor air in the monitored locations contains VOCs from a diverse range of sources. These sources include, but are not limited to building materials, combustion processes, household products, personal care items, tobacco smoke, air fresheners, heating appliances, solvents, cleaning products, paints, office equipment, ink, toner, industrial processes, plastic materials, electronic devices, inadequate ventilation, and external factors such as vehicular emissions.[11–13,23–26]

The compounds identified in our monitoring underscore the significant health risks of indoor air pollution, including acute and chronic effects such as sick building syndrome. The severity of these effects may vary depending on factors such as concentration, exposure duration, and individual sensitivity.[3,27–29]

Acute effects associated with VOC exposure include irritation of sensory organs, respiratory tract irritation, headaches, dizziness, visual disorders, skin irritation, breathing difficulties, nausea, mucous membrane irritation, confusion, fatigue, and general malaise, among others. Chronic exposure poses an even greater risk, correlating with an increased likelihood of cardiovascular and pulmonary diseases, asthma, heart disease, liver damage, cancer, adverse birth outcomes, congenital malformations, and neurobehavioral issues, among other health concerns. In extreme cases, chronic exposure can result in premature death.[5,6,12,25,28,30–32]

Notably, BTEX compounds elicit particular concerns due to their classification as carcinogens or potential carcinogens by reputable regulatory bodies. Benzene is classified as carcinogenic to humans and animals by the U.S. Environmental Protection Agency (USEPA), and the International Agency for Research on Cancer (IARC) categorizes benzene as Group 1 (carcinogenic to humans). Ethylbenzene is classified as Group 2B (possibly carcinogenic to humans), while toluene and xylenes are categorized as Group 3 (not classifiable as to their carcinogenicity to humans) by IARC. The carcinogenic properties of benzene, in particular, highlight the urgent need for stringent regulatory measures to minimize exposure to this hazardous substance.[19,33,34]

Furthermore, our inquiry highlights the detrimental effects associated with certain VOCs, particularly decamethylcyclopentasiloxane (D5) and octamethylcyclotetrasiloxane (D4). These compounds have been shown to negatively impact various physiological systems, including the endocrine and reproductive systems, with potential implications for immune function, as well as the liver, lungs, and kidneys. Notably, while D4 meets the criteria for persistence, bioaccumulation, and toxicity, authorities classify D5 as highly persistent and bioaccumulative, with uncertainty surrounding its toxicity.[35,36]

Tobacco smoke, a significant contributor to indoor VOC concentrations, has unequivocally

been classified as a Group A carcinogen by the USEPA since 1994, highlighting the serious health risks it poses. According to the WHO, tobacco smoke is responsible for approximately 90% of lung cancer deaths.[27,37,38]

Additionally, VOCs emitted by air fresheners are considered potentially toxic or hazardous under federal regulations in the United States and standards set by the EPA, with some classified as carcinogenic. These emissions are one of the primary sources of VOCs in monitored indoor air locations.[24,39]

These findings emphasize the urgent need for comprehensive measures to address indoor air pollution, including improved ventilation, removal of VOC sources, and increased public awareness of the associated health risks.

4. CONCLUSION

This study addresses a significant gap in indoor air quality monitoring in North Macedonia and Kosovo, where volatile organic compounds (VOCs) had not been previously monitored. Comprehensive monitoring was conducted at seventeen sites spanning educational, industrial, commercial, and residential areas, employing Radiello® diffusive/passive sampler devices.

The findings reveal a diverse range of sources contributing to the VOC profile in indoor air, with compounds detected that are known to cause adverse health effects. These effects encompass a wide spectrum of acute and chronic health issues, including sick building syndrome (SBS).

The study highlights the critical need for continuous year-round monitoring of VOC concentrations in various indoor environments. Furthermore, it underscores the necessity of implementing a national program to promote and enforce indoor air quality standards pertaining to VOCs.

REFERENCES

(1) Huang, L.; Qian, H.; Deng, S.; Guo, J.; Li, Y.; Zhao, W.; Yue, Y., Urban Residential Indoor Volatile Organic Compounds in Summer, Beijing: Profile, Concentration and Source Characterization. *Atmos Environ* **2018**, *188*, 1–11. https://doi.org/10.1016/j.atmosenv.2018.06.009

(2) Arı, A.; Ertürk Arı, P.; Yenisoy-Karakaş, S.; Gaga, E. O., Source Characterization and Risk Assessment of Occupational Exposure to Volatile Organic Compounds (VOCs) in a Barbecue Restaurant. *Build Environ* **2020**, *174*, 106791. https://doi.org/10.1016/j.buildenv.2020.106791

(3) Sakai, N.; Yamamoto, S.; Matsui, Y.; Khan, M. F.; Latif, M. T.; Ali Mohd, M.; Yoneda, M., Characterization and Source Profiling of Volatile Organic Compounds in Indoor Air of Private Residences in Selangor State, Malaysia. *Sci Total Environ* **2017**, *586*, 1279–1286. https://doi.org/10.1016/j.scitotenv.2017.02.139

(4) Srivastava, P., Volatile Organic Compounds in Indoor Environments in Mumbai, India. *Sci Total Environ* **2000**, *255* (1–3), 161–168. https://doi.org/10.1016/S0048-9697(00)00465-4

(5) Irga, Peter. J.; Mullen, G.; Fleck, R.; Matheson, S.; Wilkinson, Sara. J.; Torpy, Fraser. R., Volatile Organic Compounds Emitted by Humans Indoors – A Review on the Measurement, Test Conditions, and Analysis Techniques. *Build Environ* **2024**, *255*, 111442. https://doi.org/10.1016/j.buildenv.2024.111442

(6) Otgonbyamba, O.-E.; Ganbat, G.; Khuyag, S.-O.; Altangerel, E.; Ganbold, B.; Bayanjargal, A.; Bat-Erdene, A.; Chuluunbaatar, B.; Badrakh, B.; Batbaatar, S., Health Risk Assessment of Volatile Organic Compounds for Children in Indoor Air, Ulaanbaatar, Mongolia. *Aerosol Air Qual Res* **2023**, *23* (8), 230028. https://doi.org/10.4209/aaqr.230028

(7) Goodman, N. B.; Steinemann, A.; Wheeler, A. J.; Paevere, P. J.; Cheng, M.; Brown, S. K., Volatile Organic Compounds within Indoor Environments in Australia. *Build Environ* **2017**, *122*, 116–125. https://doi.org/10.1016/j.buildenv.2017.05.033

(8) Wetzel, T. A.; Doucette, W. J., Plant Leaves as Indoor Air Passive Samplers for Volatile Organic Compounds (VOCs). *Chemosphere* **2015**, *122*, 32–37. https://doi.org/10.1016/j.chemosphere.2014.10.065

(9) Montaluisa-Mantilla, M. S.; García-Encina, P.; Lebrero, R.; Muñoz, R., Botanical Filters for the Abatement of Indoor Air Pollutants. *Chemosphere* **2023**, *345*, 140483. https://doi.org/10.1016/j.chemosphere.2023.140483

(10) World Health Organization, *WHO Global Air Quality guidelines: Particulate Matter (PM2.5 and PM10), Ozone, Nitrogen Dioxide, Sulfur Dioxide and Carbon Monoxide.* World Health Organization 2021. https://iris.who.int/handle/10665/345329 (accessed 2024-04-14).

(11) Paciência, I.; Madureira, J.; Rufo, J.; Moreira, A.; De Olivera Fernandes, E., A Systematic Review of Evidence and Implications of Spatial and Seasonal Variations of Volatile Organic Compounds (VOC) in Indoor Human Environments. *J. Toxicol. Environ. Health, Part B* **2016**, *19* (2), 47–64. https://doi.org/10.1080/10937404.2015.1134371

(12) Liu, N.; Bu, Z.; Liu, W.; Kan, H.; Zhao, Z.; Deng, F.; Huang, C.; Zhao, B.; Zeng, X.; Sun, Y.; Qian, H.; Mo, J.; Sun, C.; Guo, J.; Zheng, X.; Weschler, L. B.; Zhang, Y., Indoor Exposure Levels and Risk Assessment of Volatile Organic Compounds in Residences, Schools, and Offices in China from 2000 to 2021: A Systematic Review. *Indoor Air* **2022**, *32* (9). https://doi.org/10.1111/ina.13091

(13) Vera, T.; Villanueva, F.; Wimmerová, L.; Tolis, E. I. An Overview of Methodologies for the Determination of Volatile Organic Compounds in Indoor Air. *Appl Spectrosc Rev* **2022**, *57* (8), 625–674. https://doi.org/10.1080/05704928.2022.2085735

(14) Li, Y.; Cakmak, S.; Zhu, J. Profiles and Monthly Variations of Selected Volatile Organic Compounds in Indoor Air in Canadian Homes: Results of Canadian National

Indoor Air Survey 2012–2013. *Environ Int* **2019**, *126*, 134–144. https://doi.org/10.1016/j.envint.2019.02.035

(15) Yuan, B.; Shao, M.; Lu, S.; Wang, B. Source Profiles of Volatile Organic Compounds Associated with Solvent Use in Beijing, China. *Atmos Environ* **2010**, *44* (15), 1919–1926. https://doi.org/10.1016/j.atmosenv.2010.02.014

(16) ASTM D6196-03. *Standard Practice for Selection of Sorbents, Sampling, and Thermal Desorption Analysis Procedures for Volatile Organic Compounds in Air*. https://www.astm.org/d6196-03.html (Accessed 2024-04-14)

(17) ISO 16000-5:2007. *Indoor Air — Part 5: Sampling Strategy for Volatile Organic Compounds (VOCs)*. https://www.iso.org/standard/37388.html (Accessed 2024-04-14)

(18) Radiello®. *Radiello® Manual*. https://d3pcsg2wjq9izr.cloudfront.net/files/2285/download/724345/radiello_manual_final.pdf (Accessed 2024-04-14)

(19) Mula, V.; Bogdanov, J.; Stanoeva, J. P.; Zeneli, L.; Zdravkovski, Z., Monitoring Volatile Organic Compounds in Air Using Passive Sampling: Regional Cross-Border Study between N. Macedonia and Kosovo. *Aerosol Air Qual Res* **2024**, *24* (1), 230170. https://doi.org/10.4209/aaqr.230170

(20) Król, S.; Zabiegała, B.; Namieśnik, J., Monitoring VOCs in Atmospheric Air II, Sample Collection and Preparation. *TrAC, Trends Anal. Chem.* **2010**, *29* (9), 1101–1112. https://doi.org/10.1016/j.trac.2010.05.010

(21) Joos, P. E.; Godoi, A. F. L.; De Jong, R.; De Zeeuw, J.; Van Grieken, R., Trace Analysis of Benzene, Toluene, Ethylbenzene and Xylene Isomers in Environmental Samples by Low-Pressure Gas Chromatography–Ion Trap Mass Spectrometry. *J Chromatogr A* **2003**, *985* (1–2), 191–196. https://doi.org/10.1016/S0021-9673(02)01843-5

(22) Sofronievska, I.; Petreska Stanoeva, J.; Bogdanov, J.; Stefova, M. Assay of Volatile Organic Compounds in Urban Air Using Passive Sampling and Gas Chromatography Coupled to Mass Spectrometry. *Maced. J. Ecol. Environ.* **2022**, *24* (2), 103–113. https://doi.org/10.59194/MJEE22242103s

(23) Baya, M. P.; Bakeas, E. B.; Siskos, P. A. Volatile Organic Compounds in the Air of 25 Greek Homes. *Indoor Built Environ.* **2004**, *13* (1), 53–61. https://doi.org/10.1177/1420326X04036007

(24) Lin, K.-H.; Tsai, J.-H.; Cheng, C.-C.; Chiang, H.-L., Emission of Volatile Organic Compounds from Consumer Products. *Aerosol Air Qual Res* **2022**, *22* (9), 220250. https://doi.org/10.4209/aaqr.220250

(25) Xu, J.; Szyszkowicz, M.; Jovic, B.; Cakmak, S.; Austin, C. C.; Zhu, J., Estimation of Indoor and Outdoor Ratios of Selected Volatile Organic Compounds in Canada. *Atmos Environ* **2016**, *141*, 523–531. https://doi.org/10.1016/j.atmosenv.2016.07.031

(26) Pośniak, M.; Makhniashvili, I.; Koziel, E., Volatile Organic Compounds in the Indoor Air of Warsaw Office Buildings. *Indoor Built Environ.* **2005**, *14* (3–4), 269–275. https://doi.org/10.1177/1420326X05054071

(27) Parra, M. A.; Elustondo, D.; Bermejo, R.; Santamaría, J. M., Quantification of Indoor and Outdoor Volatile Organic Compounds (VOCs) in Pubs and Cafés in Pamplona, Spain. *Atmos Environ* **2008**, *42* (27), 6647–6654. https://doi.org/10.1016/j.atmosenv.2008.04.026

(28) Nunes, C. R. de O.; Sánchez, B.; Gatts, C. E. N.; De Almeida, C. M. S.; Canela, M. C., Evaluation of Volatile Organic Compounds Coupled to Seasonality Effects in Indoor Air from a Commercial Office in Madrid (Spain) Applying Chemometric Techniques. *Sci. Total Environ.* **2019**, *650*, 868–877. https://doi.org/10.1016/j.scitotenv.2018.09.095

(29) Chi Sing Chan; Shun Cheng Lee; Chan, W.; Kin Fai Ho; Linwei Tian; Sen Chao Lai; Yok Sheung Li; Yu Huang., Characterisation of Volatile Organic Compounds at Hotels in Southern China. *Indoor Built Environ.* **2011**, *20* (4), 420–429. https://doi.org/10.1177/1420326X11409458

(30) Zhang, Z.-F.; Zhang, X.; Zhang, X.; Liu, L.-Y.; Li, Y.-F.; Sun, W., Indoor Occurrence and Health Risk of Formaldehyde, Toluene, Xylene and Total Volatile Organic Compounds Derived from an Extensive Monitoring Campaign in Harbin, a Megacity of China. *Chemosphere* **2020**, *250*, 126324. https://doi.org/10.1016/j.chemosphere.2020.126324

(31) Grenga, P. N.; Gallagher, M. J.; McGahan, M. E.; Raymond, D. M.; Priefer, R. Assessment of Airborne Total Volatile Organic Compounds of Niagara Falls Residences as Compared to Resident Lifestyle. *Indoor Built Environ.* **2011**, *20* (2), 226–231. https://doi.org/10.1177/1420326X10389277

(32) Reese, K.; Tebehaevu, O.; Balanay, J. A. G., Volatile Organic Compound Emission in a University Printing Press Facility in Eastern North Carolina. *Indoor Built Environ.* **2021**, *30* (3), 426–434. https://doi.org/10.1177/1420326X19896840

(33) World Health Organization. *Agents Classified by the IARC Monographs,* Volumes 1–135. https://monographs.iarc.who.int/agents-classified-by-the-iarc/ (Accessed 2024-04-14)

(34) U.S. Environmental Protection Agency. *Initial List of Hazardous Air Pollutants with Modifications*. https://www.epa.gov/haps/initial-list-hazardous-air-pollutants-modifications (accessed 2024-04-14).

(35) Gentry, R.; Franzen, A.; Van Landingham, C.; Greene, T.; Plotzke, K., A Global Human Health Risk Assessment for Octamethylcyclotetrasiloxane (D 4). *Toxicol Lett* **2017**, *279*, 23–41. https://doi.org/10.1016/j.toxlet.2017.05.019

(36) Molinier, B.; Arata, C.; Katz, E. F.; Lunderberg, D. M.; Liu, Y.; Misztal, P. K.; Nazaroff, W. W.; Goldstein, A. H., Volatile Methyl Siloxanes and Other Organosilicon Compounds in Residential Air. *Environ Sci Technol* **2022**, *56* (22), 15427–15436. https://doi.org/10.1021/acs.est.2c05438

(37) Sheu, R.; Stönner, C.; Ditto, J. C.; Klüpfel, T.; Williams, J.; Gentner, D. R., Human Transport of Thirdhand Tobacco Smoke: A Prominent Source of Hazardous Air Pollutants into Indoor Nonsmoking Environments. *Sci Adv* **2020**, *6* (10). https://doi.org/10.1126/sciadv.aay4109

(38) Xie, J.; Wang, X.; Sheng, G.; Bi, X.; Fu, J., Determination of Tobacco Smoking Influence on Volatile Organic Compounds Constituent by Indoor Tobacco Smoking Simulation Experiment. *Atmos Environ* **2003**, *37* (24), 3365–3374. https://doi.org/10.1016/S1352-2310(03)00354-6

(39) Steinemann, A. Ten Questions Concerning Air Fresheners and Indoor Built Environments. *Build Environ* **2017**, *111*, 279–284. https://doi.org/10.1016/j.buildenv.2016.11.009