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Review article

Volatile and semi-volatile organic compounds in landfill gas: Composition characteristics and health risks

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ABSTRACT

Gas emitted from landfills contains a large quantity of volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs), some of which are carcinogenic, teratogenic, and mutagenic, thereby posing a serious threat to the health of landfill workers and nearby residents. However, the global hazards of VOCs and SVOCs in landfill gas to human health remain unclear. To quantify the global risk distributions of these pollutants, we collected the composition and concentration data of VOCs and SVOCs from 72 landfills in 20 countries from the core database of Web of Science and assessed their human health risks as well as analyzed their influencing factors. Organic compounds in landfill gas were found to primarily result from the biodegradation of natural organic waste or the emissions and volatilization of chemical products, with the concentration range of 1×10^{-1}–1×10^{6} μg/m3. The respiratory system, in particular, lung was the major target organ of VOCs and SVOCs, with additional adverse health impacts ranging from headache and allergies to lung cancer. Aromatic and halogenated compounds were the primary sources of health risk, while ethyl acetate and acetone from the biodegradation of natural organic waste also exceeded the acceptable levels for human health. Overall, VOCs and SVOCs affected residents within 1,000 m of landfills. Air temperature, relative humidity, air pressure, wind direction, and wind speed were the major factors that influenced the health risks of VOCs and SVOCs. Currently, landfill risk assessments of VOCs and SVOCs are primarily based on respiratory inhalation, with health risks due to other exposure routes remaining poorly elucidated. In addition, potential health risks due to the transport and transformation of landfill gas emitted into the atmosphere should be further studied.

1. Introduction

The generation of municipal solid waste (MSW) has drastically increased with the rapid growth rates of economy and population. As in other countries, there was a significant increase in the production of China's MSW with 235 million tons in 2020, a 90.4% increase compared to 2010 (Ministry of housing., 2021). Globally, annual generation of MSW is expected to reach 3.4 billion tons by 2050 (Haza et al., 2018).

Landfill is one of the most important means of MSW disposal worldwide. Large amounts of gases are produced in landfills through complex physical, chemical, and biochemical processes and being emitted into the atmosphere through cover soils or landfill wells, thereby threatening the safety and health of landfill workers and nearby residents. Waste in landfills includes natural organic materials, such as vegetables, meat, and trees, and synthetic substances, such as chlorinated disinfectants and petrochemicals, which releases a large variety and quantity of volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs) (Slack et al., 2005).

Previous studies on landfills have focused on the following three aspects: (1) landfill gas, in particular, methane and greenhouse gas emission (Lucernoni et al., 2017); (2) landfill leachate composition, degradation and treatment characteristics, and groundwater pollution characteristics (Abiriga et al., 2020; Mehmet et al., 2020; Oturan et al., 2015; Yang et al., 2021); and (3) degradation of organics and humus

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formation (Liu et al., 2021; Yang and Antonietti, 2020). Previous studies on landfill gas have primarily focused on methane, and the human health risks and transformation characteristics of VOCs and SVOCs remaining overlooked. Many VOCs and SVOCs in landfill gas are mutagenic, teratogenic, and carcinogenic and can cause human health risks, such as irritating respiratory tract, damaged central nervous system, and even cancer (Allen et al., 1997; Butt et al., 2016). Human health risks posed by VOCs and SVOCs in landfill gas have been lately receiving attention but have remained spatially limited to individual landfills (Liu et al., 2016a; Piccardo et al., 2022; Wang et al., 2021). Only few studies have been conducted about the compositions and concentrations of VOCs and SVOCs (Wu et al., 2018; Yao et al., 2019). Therefore, this study aimed to (1) globally characterize the sources, compositions, and concentrations of VOCs and SVOCs produced by landfills; (2) assess their potential human health risks to workers and nearby residents and address the biological mechanisms underlying those risks; and (3) identify their primary drivers.(Yang and Antonietti, 2020)

2. Materials and methods

2.1. Data collection

Scientific studies published during 2002–2022 were searched with the subject words of “VOC, SVOC, VOCs, SVOCs, volatile organic compound, semi-volatile organic compound, volatile organic compounds, or semi-volatile organic compounds” and “landfill gas” in the core database of Web of Science. As shown in Table S1, 53 publications, including 72 landfills in 20 countries, were obtained on the VOC and SVOC data worldwide. For the lowest concentrations, the terms used in the original studies included “ND” (not detected) and “<LOQ” (below limit of quantification), and n.a. (not available) was used to indicate studies reporting only maximum concentrations.

2.2. Sampling and measurement techniques

The sampling techniques applied in 53 studies included sampling bags, vacuum canisters, glass-bombs, sorbent tubes, and flux boxes, with the sampling bags being the most common one. The measurement methods applied in 53 studies included gas-vent sampling, velocity measurement, surface flux chambers, production model of landfill gas, concentration ratios estimated relative to methane, and inverse dispersion modeling, with the surface flux chambers being the most common one. Most of these studies in our data chose helium as gas matrix during the measure process (Chiriac et al., 2007; Ding et al., 2012; Fang et al., 2012; He et al., 2015; Kim et al., 2012; Lim et al., 2018), and the rest used nitrogen as gas matrix (Duan et al., 2020; Liu and Zheng, 2020).

2.3. Risk assessment

Based on carcinogenicity, the International Agency for Research on Cancer (IARC) classifies VOCs and SVOCs into various groups, including Group 1 (carcinogenic to humans), Group 2A (probably carcinogenic to humans), and Group 2B (possibly carcinogenic to humans) (Yao et al., 2019). Based on the concentration data of VOCs and SVOCs extracted from 53 studies (Table S1), human health risk assessment was conducted to estimate their health risks to landfill workers and nearby residents. However, we excluded the VOC and SVOC data from organized emissions, such as soil pores and manifolds before burning in a flare. Exposure to pollutants in the atmosphere occurs via the following three pathways: dermal absorption, ingestion, and respiratory inhalation. As the exposed population at landfills are primarily workers and the pollutant carcinogenicity parameters are incomplete, human health risk assessment conducted in this study was limited to respiratory inhalation. Non-carcinogenic and carcinogenic effects of exposure to odorous volatile compounds by inhalation were evaluated via the methodology recommended by the United States Environmental Protection Agency (Usepa, 2009). The exposure concentration (EC, $\mu g{\cdot}m^{-3}$) can be estimated as follows:

$$EC = \frac{Ca \times ET \times EF \times ED}{AT \times 365 \times 24} \tag{1}$$

where Ca is airborne pollutant concentration ($\mu g{\cdot}m^{-3}$), ET is daily exposure time ($8\ h{\cdot}d^{-1}$), EF is exposure frequency ($260\ d{\cdot}yr^{-1}$);, ED is exposure duration (in 25 yr), and AT is the average time of effect expressed as 75 yr for carcinogenic effect and 25 yr for non-carcinogenic effect.

The carcinogenic risk (CR) of individual pollutants (ΣCR) can be estimated using Eq. (2)

$$CR = EC \times IUR \tag{2}$$

where IUR is inhalation unit risk $(\mu g{\cdot}m^{-3})^{-1}$. When IUR $= 2 \times 10^{-6}$ $(\mu g{\cdot}m^{-3})^{-1}$, two additional cancer cases (upper bound) per one million people are expected if exposed to 1 μg of chemical per m^3 of air every day during the average time of effect (Piccardo et al., 2022). When $CR > 1$, carcinogenic risk is considered to be possible.

The non-carcinogenic risk of individual pollutants (HI) can be estimated using Eq. (3)

$$HI = \frac{EC}{RfC \times 1000\mu g/m^3} \tag{3}$$

where RfC is the reference concentration ($mg{\cdot}m^{-3}$). When $HI > 1$, non-carcinogenic risk is considered to be possible.The toxicological parameters of VOCs and SVOCs are shown in Table S5.

2.4. Statistical analysis

Statistical analysis was conducted using GraphPad Prism 8.0. Figures and maps were produced using OriginPro 2018 and ArcGIS 10.8, respectively.

3. Results and discussion

3.1. Sources, compositions, and concentrations of VOCs and SVOCs in landfill gas

3.1.1. Sources of organic pollutants in landfill gas

Organic compounds in landfill gas were found to primarily result from the biodegradation of natural organic waste or the emissions and volatilization of chemical products (Duan et al., 2021a, 2021b). As presented in Table 1, hydrocarbon, nitrogen, oxygen, and sulfur compounds and terpenes originate through the biodegradation of organic waste, such as kitchen waste, vegetables, and fruits, and their compositions and concentrations depend on the biodegradation stage of waste. The aromatic and halogenated compounds originate from the direct volatilization of chemical products or the biological degradation of natural organic wastes (Duan et al., 2014). The aromatic compounds primarily originate from petrochemicals, such as coal tar, asphalt, and crude oil, among which benzene was the most common, which is derived from chemical products, such as gasoline, paints, and varnishes (Table S2). The halogenated compounds primarily originated from chemical products, such as disinfectants, pesticides, and degreasers (Dincer et al., 2006), among which the most common was tetrachloroethylene, which is derived from dry cleaners, dye solvents, and detergents (Table S3).

3.1.2. Compositions of VOCs and SVOCs in different countries

VOCs and SVOCs in 72 landfills of 20 countries were reported. The top five reported countries were China, South Korea, the United Kingdom, Spain, and the United States (Figure S1), all belonging to the high-income class, except China that belongs to the mid-to-high income

Table 1
Sources of organic compounds in landfills.

Compounds	Sources	References
Oxygenated compounds	Vegetables, fruits, kitchen waste, yard waste, medical waste, solvents, and plastic packaging	(Duan et al., 2021a; Zou et al., 2003; Butt et al., 2016)
Sulfocompounds	Kitchen waste, gypsum drywall, and wall board	(Duan et al., 2021b; James and Stack, 1997; Lucernoni et al., 2017; Yaghmaien et al., 2019; Butt et al., 2016)
Nitrogen compounds	Proteins, rubber, and kitchen waste	(Abiriga et al., 2020; Chiriac et al., 2007; James and Stack, 1997; Wu et al., 2017)
Hydrocarbons	Edible oils, paper, plastic packaging, fragrant detergents, solvents, refriger ants, waste cooking oil, and pesticides	(AlAhmad et al., 2012; Duan et al., 2021b; Lucernoni et al., 2017)
Terpenes	Yard waste, plant waste, household aromatic cleaners, air fresheners, and pharmaceuticals	(Duan et al., 2021b; James and Stack, 1997; Zou et al., 2003)
Aromatic compounds	Coal tar, asphalt, crude oil, plastic bags, paints, vehicle exhaust emissions, food containers, food with high fat content, paper, yard waste, solubilizers, and yard/green waste.	(Abiriga et al., 2020; AlAhmad et al., 2012; Baghanam et al., 2020; James and Stack, 1997; Liu et al., 2016b; Zou et al., 2003; Randazzo et al., 2022)
Halogenated compounds	Pesticides, insecticides, disinfectants, cleaners, plastics, dry cleaners, paint strippers, dye solvents, degreasers, soaps, paints, varnishes, and refrigerants	(AlAhmad et al., 2012; Dincer et al., 2006; Gonzalez et al., 2013; Piccardo et al., 2022; Butt et al., 2016)

class. Briefly, the level of the national economic growth was positively correlated with the study of VOCs and SVOCs in landfills.

These 72 landfills analyzed globally were located in 40 cities. As shown in Fig. 1, the top three cities with the most detected pollutants were Shanghai ($n = 132$), Hangzhou ($n = 116$), and Beijing ($n = 91$). However, no>40 pollutants were detected in landfill gas emissions from most of the remaining cities. Aromatic compounds were the most reported landfill gas in all countries (Figure S2). The aromatic compounds included toluene, ethylbenzene, and xylenes, as detected in landfills on several continents, including Europe, North America, and Asia. As the major components of benzene, toluene, ethylbenzene, and xylene (BTEX), these three compounds are widely present in chemicals expended by humans in everyday life, such as gasoline, dyes, and paints. The halogenated compounds, in particular, tetrachloroethylene and trichloroethylene, were also commonly reported. Among the top 50 reported VOCs and SVOCs in landfill gas, a small number of sulfur compounds was present, with methyl mercaptan ($n = 12$) and dimethyl sulfide ($n = 12$) being detected frequently primarily through the degradation of sulfur-containing food waste, such as methionine.

3.1.3. *Concentrations of VOCs and SVOCs*

A significant spatial difference was observed in the concentrations of VOCs and SVOCs in landfill gas worldwide. The concentrations of the same pollutants in the different countries varied by 6–8 orders of magnitude over a range of 1×10^{-1}–1×10^{6} μg/m^3. As shown in Fig. 2, the aromatic compounds were present at very high concentrations in narrow ranges in certain landfills. Compared with those of the aromatic compounds, the overall concentrations of the halogenated compounds were slightly low but differed widely between specific pollutants. Although a small number of sulfur compounds were reported in a few studies, their overall concentrations were high. For example, dimethyl sulfide, and methyl mercaptan generally occurred at higher concentrations than the other pollutants. In China, this was primarily because the high proportion of kitchen waste was characterized by high salt, oil, and water contents, which is more likely to produce high concentrations of sulfur and oxygen compounds (Liu et al., 2018). The overall concentrations of oxygen and hydrocarbon compounds were less variable than those of sulfur compounds and showed a decreasing trend with the increasing carbon atoms.

The concentrations of VOCs and SVOCs in landfill gas were affected by the sampling and measurement method. As show in Table S4, the chemical composition of the sampling bag method may undergo a significant loss or change during storage, or vacuum canisters may lose analytes owing to wall effect, matrix effect, and condensation and VOCs or SVOCs loss in sorbent tubes due to reaction between sulphur compounds and carbon-based sorbent tubes. In addition, human activities will also affect the concentration of VOCs and SVOCs. The dumping and compacting work of waste in the landfill will promote the volatilization of VOCs and SVOCs in the waste, resulting in an increase in the concentration in the air (Chiriac et al., 2007; Duan et al., 2014).

3.2. *Potential human health risk assessment*

Because only the respiratory inhalation is considered and the dermal absorption and ingestion is not considered, the results of human health

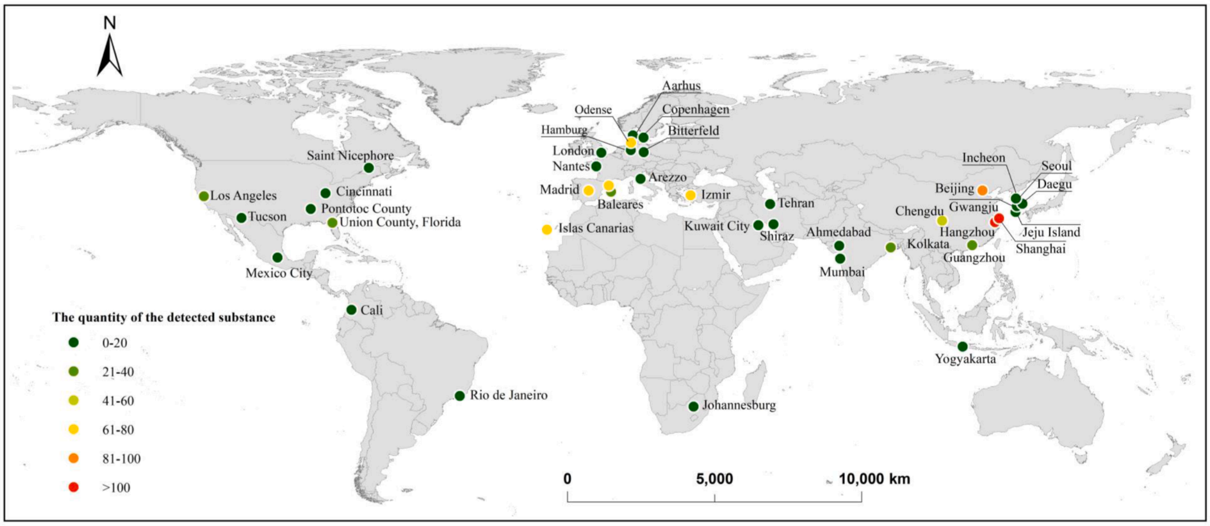

Fig. 1. Quantity and spatial distribution of VOCs and SVOCs detected in landfill gas in cities.

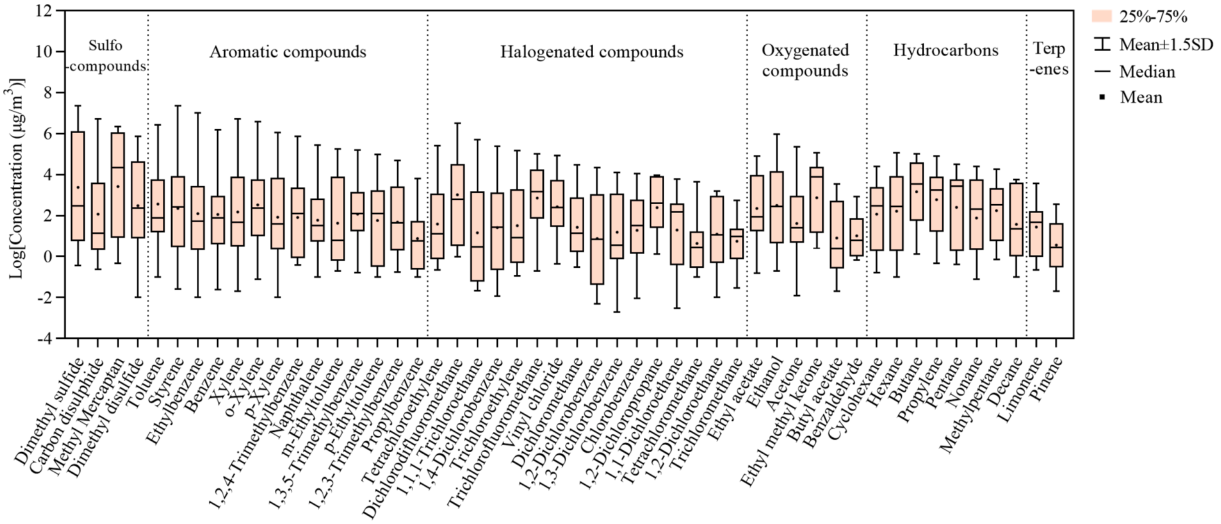

Fig. 2. Concentrations of the top 50 VOCs and SVOCs detected in landfill gas in cities.

risk assessment may be not completely accurate. As the concentration data of VOCs in the papers are not all sampled in the way of simulating human respiratory inhalation, the results of human health assessment in this paper can only be called potential human health risk assessment.

3.2.1. *Impacts of VOCs and SVOCs on human health*

(1) Cancer *risks posed by the inhalation of VOCs and SVOCs*

The inhalation of VOCs and SVOCs can cause cancer (Liu et al., 2014; Yang et al., 2017). As presented in Table 2, chloroform, tetrachromethane, hexanal, and 1,4-dioxane can cause breast caner. Benzene may cause leukemia, and 1,4-dichlorobenzene may cause head and neck cancer. The lung is the major target organ of polycyclic aromatic hydrocarbons (PAH) carcinogenicity, and the inhalation of PAHs can cause lung cancers; PAHs can also cause skin, bladder, and urinary system cancers as a function of different routes, concentrations, and exposure environments. For example, the increased risk of skin cancer follows high dermal exposure to PAHs, whereas the increased risk of bladder and other organ cancers is primarily found in workers in industries with high exposure to PAHs (Boffetta et al., 1997).

(2) Non-carcinogenic risks posed by the inhalation of VOCs and SVOCs

The impacts of VOCs and SVOCs on human health are primarily concentrated in the respiratory system as respiratory inhalation is the major route of human exposure to VOCs and SVOCs. As presented in Table 3, α-pinene, 2-methyl-1,2-butadiene, and aldehydes, such as hexanal and octanal, are associated with the deterioration of lung function (Cakmak et al., 2014), while chlorobenzene, 1,2-dichlorobenzene, 1,3-dichlorobenzene, and 1,4-dichlorobenzene can cause asthma. Asthma can lead to irreversible emphysema and permanent lung damage. Long-term lung damage increases the risks of asthma attack and heart disease (Kim et al., 2013).

Table 2
Carcinogenic health effects of VOCs and SVOCs.

Diseases	Compounds	References
Breast cancer	Chloroform, tetrachloromethane, hexanal, and 1,4-dioxane	(Calderon et al., 2022)
Leukemia	Benzene	(Zhou et al., 2015)
Head and neck cancer	1,4-dichlorobenzene	(Taware et al., 2018)
Lung cancer	PAHs	(Ben et al., 2004; Boffetta et al., 1997; Bosetti et al., 2005)
Skin cancer	PAHs	(Boffetta et al., 1997)
Bladder cancer	PAHs	(Boffetta et al., 1997; Bosetti et al., 2005)
Carcinoma of urinary system	PAHs	(Bosetti et al., 2005)

Table 3
Non-carcinogenic health effects of VOCs and SVOCs.

Diseases	Compounds	References
Lung function deterioration	Hexanal, octanal, nonanal, decanal, benzene, styrene, α-pinene, 2-methyl-1,2-butadiene, naphthalene, and 2-furancarboxaldehyde	(Cakmak et al., 2014; Martins et al., 2012)
Asthma	Benzene, toluene, ethylbenzene, p-xylene, m-xylene, styrene, chlorobenzene, 1,2-dichlorobenzene, 1,3-dichlorobenzene, and 1,4-dichlorobenzene	(Martins et al., 2012; Rumchev et al., 2004)
Allergies	Formaldehyde, hexane, octane, nonane, decane, toluene, ethylbenzene, p ± m-xylene, o-ethyltoluene, m-ethyltoluene, p-ethyltoluene, and chlorobenzene	(Kim et al., 2013; Lehmann et al., 2001; Martins et al., 2012)

In addition to adversely affecting the human respiratory system, VOCs and SVOCs can cause allergies, headache, eye irritation, nausea, and vomiting (Duan et al., 2014; Kim et al., 2013; Martins et al., 2012). As presented in Table 3, naphthalene and other aromatic compounds, such as benzene, toluene, and ethylbenzene, can cause allergic reactions in addition to asthma and lung function deterioration (Lehmann et al., 2001; Martins et al., 2012; Rumchev et al., 2004), while chlorobenzene and alkanes, such as hexane and octane, increase the likelihood of allergic reactions to milk and egg albumen (Lehmann et al., 2001). Atopic reactions increase the risk of allergies, which can cause shock and eventually death.

3.2.2. *Global distribution of potential human health risks*

The carcinogenic risks of various landfill pollutants at the maximum (CR_{max}) and mean (CR_{mean}) concentrations were estimated for the 40 cities worldwide. Globally, 30 out of 34 pollutants may cause cancer in humans. As shown in Fig. 3, Hangzhou ($n = 20$), Shanghai ($n = 19$), and Union County of Florida ($n = 12$) exhibited the largest number of carcinogenic VOCs and SVOCs, with the remaining cities exhibiting less than ten such compounds. The aromatic and halogenated compounds were the major sources of carcinogenic risks (Figs. 4 and S3). The aromatic compounds were commonly detected with high exceedance rates in landfills, presenting the highest carcinogenic risk. The halogenated compounds exceeded risk limits at a slightly lower exceedance rate than

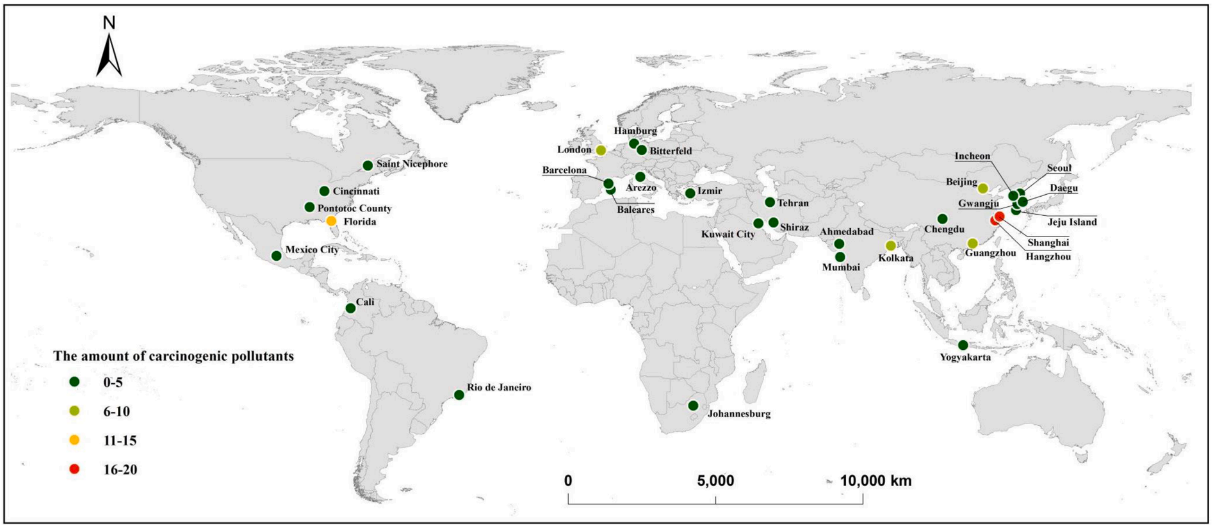

Fig. 3. Number and spatial distribution of VOCs and SVOCs with carcinogenic risk in landfill gas in cities.

that of the aromatic compounds; however, their frequent detections suggested a high carcinogenic risk. Formaldehyde, acetaldehyde, acrylonitrile, and aniline were also identified as carcinogenic (Fig. 4). Propylene oxide was detected in only one landfill despite its high carcinogenic risk ($CR_{max} = 1.5$). Therefore, more attention should be paid to the carcinogenic aromatic and halogenated compounds detected with high exceedance rates, such as ethylbenzene, benzene, carbon tetrachloride, and methylene chloride.

The non-carcinogenic risks of pollutants at the maximum (HI_{max}) and mean (HI_{mean}) concentrations detected were also assessed for the 40 cities worldwide. As shown in Fig. 5, Shanghai ($n = 29$), Hangzhou ($n = 27$), and United County of Florida ($n = 20$) released more non-carcinogenic pollutants than the other cities ($n < 20$). Both HI_{max} and HI_{mean} were greater than unity for 38 out of 52 pollutants worldwide, indicating that most organic compounds in landfill gas caused non-carcinogenic risks (Fig. 6). Pollutants with excess non-carcinogenic risks primarily included aromatic, halogenated, and oxygen compounds. Being emitted from landfills, VOCs and SVOCs showed a lower exceedance rate for non-carcinogenic risks than for carcinogenic risks (Figure S4). The aromatic compounds, such as benzene, naphthalene, and toluene, and the halogenated compounds, such as trichloroethylene, tetrachloroethylene, and chlorobenzene, not only presented high human health risk but also were frequently reported in landfills, thereby emphasizing the urgent need to focus on them. Being a potential hazard to human health, ethyl acetate, typically reported in landfills, also deserves special attention in mitigating the health risks.

Landfill is a significant source of health risks for on-site workers and nearby residents. In total, 13 pollutants, including benzene, 1,1,2-

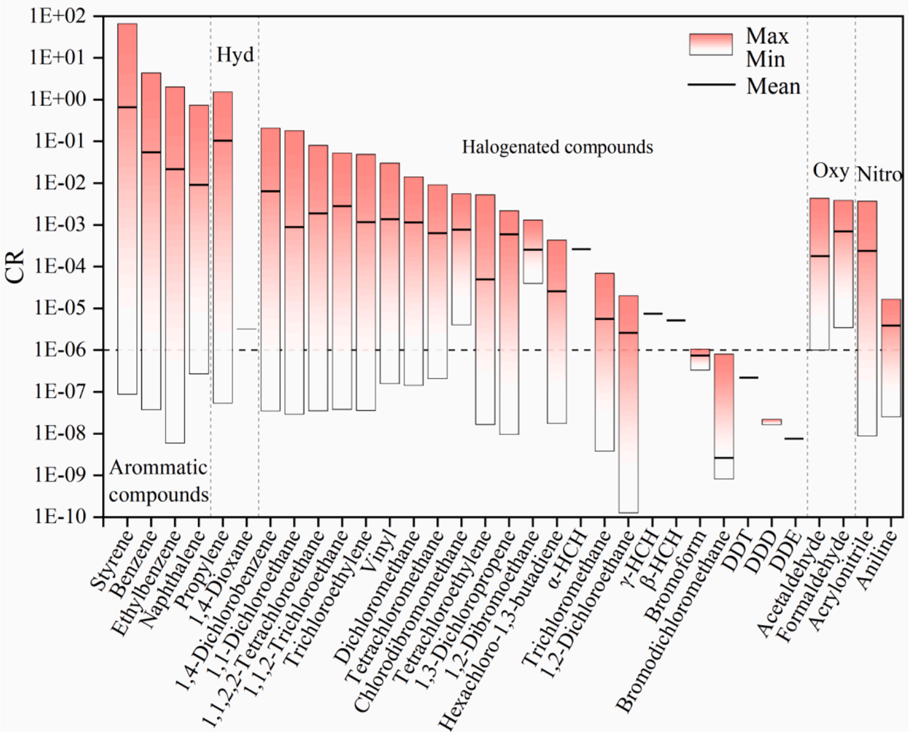

Fig. 4. Carcinogenic risk assessment of VOCs and SVOCs detected in landfill gas in cities (Hyd: hydrocarbons; Oxy: oxygenated compounds; and Nitro: nitrogen compounds).

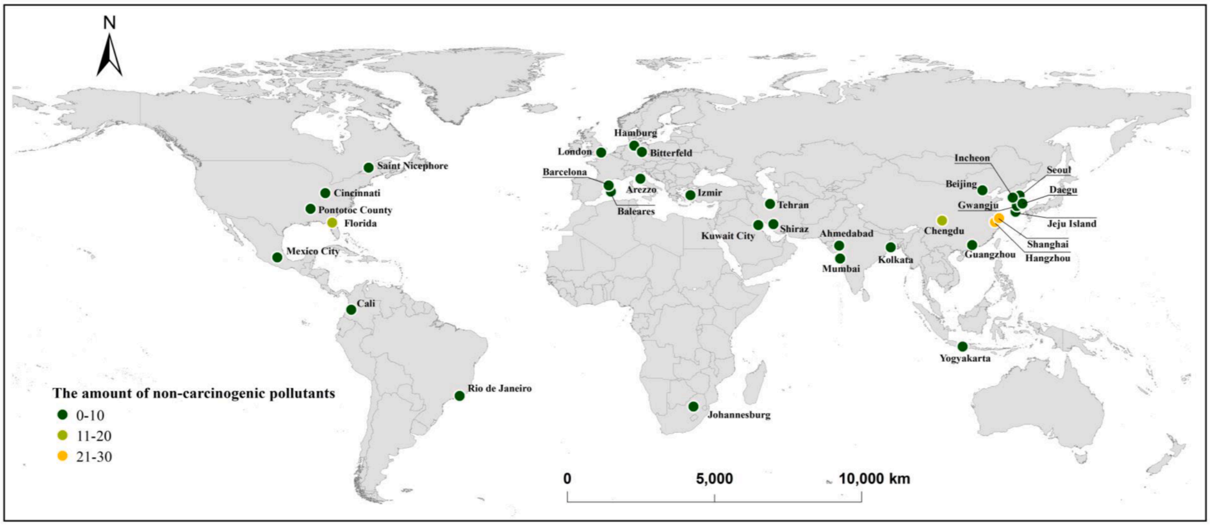

Fig. 5. Number and spatial distribution of VOCs and SVOCs with non-carcinogenic risk in landfill gas in cities.

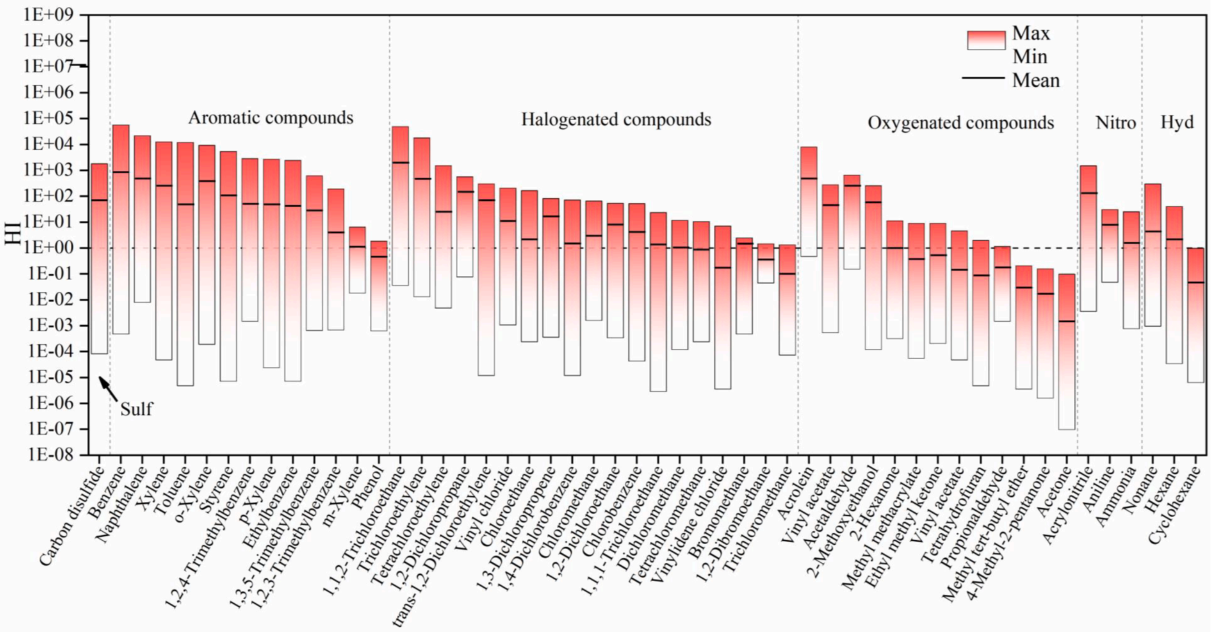

Fig. 6. Non-carcinogenic risk assessment of VOCs and SVOCs detected in landfill gas in cities (Sulf: sulfocompounds; Hyd: hydrocarbons; and Nitro: nitrogen compounds).

trichloroethane, acetaldehyde, and aniline, posed both carcinogenic and non-carcinogenic risks, among which ten comprised aromatic and halogenated compounds. Briefly, the aromatic and halogenated compounds from landfills were the dominant contributors to the health risks.

3.2.3. *Continental distribution of potential human health risks*

The halogenated and aromatic compounds were the major sources of landfill gas-related health risk in the continents of North America, Europe, and Asia. The halogenated compounds were the major source of cancer risk, followed by the aromatic compounds. However, both the halogenated and aromatic compounds were the major sources of non-carcinogenic risk (Figures S5–S10). For the remaining four continents, only limited data were obtained from South America and Africa (i.e., Brazil, Colombia, and South Africa), and health risks primarily arose from formaldehyde, acetaldehyde, and benzene (Figures S11–S12).

According to the descending order of comprehensive risks of human health, continents followed this order: Asia > Europe > North America (Figures S5–S10). The most diverse and highest quantity of pollutants with relatively high risks detected in landfill occurred in Asia than in other continents (Figure S10). For example, organic matter/carbon disulfide, nonane, and hexane did not pose a non-carcinogenic risk in North America and Europe but posed a human health risk in Asia. Organic matters such as aniline, acetone, methyl ethyl ketone, and methyl isobutyrate did not pose human health risks in North America but posed non-carcinogenic risks in Asia. Although oxygenated, sulfurous and nitrogen compounds were detected in landfills in Europe and North America but did not pose human health risks. However, most of these pollutants may pose human health risks in Asia owing to their high concentrations. The health risks from trichloroethylene were also lower in Europe than in Asia likely owing to the strict regulation of trichloroethylene issued by the European Chemicals Agency in 2010. Trichloroethylene has been banned for metal cleaners, which is a potential source of trichloroethylene detected at landfills (Allen et al., 1997; Chiriac et al., 2007). Landfills in South America and Africa posed low human health risks, with only one or two hazardous pollutants in each country, primarily owing to the small number of local studies. No risk assessment was performed for Oceania and Antarctica owing to the lack of data.

3.2.4. Country-level potential human health risks

(1) North American countries

In North America, the United States was subjected to the highest health risks from landfill gas, followed by Canada and Mexico. The carcinogenic risks of landfill gas in the United States and Canada were primarily associated with the aromatic and halogenated compounds (Figure S13). In the United States, various landfill pollutants posed carcinogenic risks, with 1,4-dichlorobenzene, 1,1-dichloroethane, and styrene as the top three pollutants. However, ethylbenzene was the most hazardous landfill gas in Canada. According to the limited number of studies, no VOCs and SVOCs that caused human health risks were observed in Mexican landfills.

Similar to the results of the carcinogenic risk assessment, the United States exhibited more non-carcinogenic pollutants and higher non-carcinogenic risks at landfills than the other North American countries. The halogenated and aromatic compounds were the major sources of non-carcinogenic risk in the United States and Canada likely owing to the extensive use of such products (Figure S14). For example, 1,1,2-trichloroethane and benzene posed health risks in the United States. Such hazardous pollutants from landfills, including xylenes, ethylbenzene, toluene, naphthalene, trichloroethylene, and tetrachloroethylene, were fewer in Canada than in the United States. In Mexico, only toluene from landfills had potential health risks.

(2) European countries

The overall health risk of VOCs and SVOCs in landfill gas was lower in the European countries than in the United States, with slight differences between the countries (Figure S15). The European countries were in the following descending order of CR_{max}: the United Kingdom > Germany > Italy > Spain. The overall health risk was relatively high in the United Kingdom owing to the six halogenated compounds with carcinogenic toxicity: trichloroethylene, tetrachloroethane, vinyl chloride, 1,1,2,-trichloroethane, tetrachloroethylene, and benzene. Similar to the United Kingdom, the halogenated compounds from landfills were the major source of carcinogenic risks in Germany. α-HCH and γ-HCH cause cancer risk but were rarely detected in landfill gas. Dichloromethane, vinyl chloride, trichloroethylene, and tetrachloroethylene required special attention as they not only have cancer risk but also have been frequently detected in landfill gas. In Italy and Spain, landfill gas exhibited low carcinogenic risk owing to the low CRmax value of pollutants and their small number detected.

The results of the non-carcinogenic and carcinogenic risk assessments were similar in the European countries. The UK and Germany were the two countries with the highest overall risk (Figure S16). The halogenated compounds, such as vinyl chloride, trichloroethylene, and 1,1,1-trichloroethane, were the major sources of non-carcinogenic risk. The non-carcinogenic risk of organic substances was lower in Italy than in the United Kingdom and Germany. No pollutants exhibited non-carcinogenic risk in Spain, as the pollutants (i.e., ethyl acetate, benzene, xylene, and vinyl chloride) detected in landfill gas remained at low concentrations. Overall, vinyl chloride and trichloroethylene showed both carcinogenic and non-carcinogenic risks that mostly exceeded the threshold in the European countries, which need to be reduced.

(3) Asian countries

The aromatic compounds from landfills were the major source of carcinogenic risks in Asia (Figure S17); however, the causes differed between the countries. As the oil-producing countries, the petrochemical industry is the mainstay of Kuwait and Iran, with a large quantity of the aromatic compounds being generated during production and processing. South Korea, China, India, and Turkey are the large consumers of petrochemicals, and the aromatic compounds primarily originate from the volatilization of used petrochemicals. The country ranking in terms of CRmax was of the following descending order: Kuwait > South Korea > China > Iran > India > Turkey. Kuwait, South Korea, and Iran were at a high risk from individual landfill pollutants but with fewer pollutants with carcinogenic risk. Overall, landfills in China were the ones that most threatened human health. A large variety of landfill pollutants in China caused carcinogenic risks, with predominantly halogenated compounds, such as trichloroethylene and methylene chloride, followed by aromatic compounds, such as styrene and benzene. Commonly used to make sanitary balls in China before 1993, naphthalene was also a potential source of carcinogenic risks. In addition, attention should be paid to formaldehyde, acetaldehyde, and acrylonitrile as these three pollutants not only pose high carcinogenic risks but also have been frequently detected in landfills. In India and Turkey, the carcinogenic risks of VOCs and SVOCs from landfills were rated low and largely associated with the oxygen, aromatic, and halogenated compounds. Most of the pollutants, such as benzene, styrene, and chloroform, showed the potential to cause carcinogenic risks in India but not in Turkey.

A large variety and quantity of VOCs and SVOCs with non-carcinogenic risks were found in Chinese landfills (Figures S18 and S19). The pollutants that exceeded the limits included the aromatic, halogenated, oxygen, and nitrogen compounds. In addition, carbon disulfide, frequently detected in Chinese landfills, exhibited non-carcinogenic risks (HI) well above the acceptable level. The situations in South Korea and Kuwait were similar, as non-carcinogenic risks from landfills in both countries were caused by the aromatic compounds, predominantly benzene and xylene. A difference was that, in South Korea, carbon disulfide with high health risks were detected in multiple landfills. Landfills in Turkey, Iran, and India posed low health risks. There were 26 non-carcinogenic pollutants found in Turkish landfills, of which acetaldehyde, acrolein, 1,2-dichloropropane, 1,1-dichloroethylene, and trichloroethylene may pose non-carcinogenic health risks. In Iran, six non-carcinogenic pollutants were detected, among which benzene, xylene, and acetaldehyde showed the potential to cause non-carcinogenic health risks. Among the 18 non-carcinogenic pollutants detected in India, acrolein and benzene were the potential sources of human health risks.

(4) Other countries

Only few studies and data on VOCs and SVOCs existed from landfill in South American, Oceanic, and African countries, such as Brazil, Colombia, and South Africa. Considering the limited data, carcinogenic risks were based on the average concentrations of pollutants. Health risks primarily arose from formaldehyde and acetaldehyde in Brazil and from benzene in Colombia and South Africa (Figure S11). Acetaldehyde from landfills showed the potential to cause health risks in Brazil. Although detected, the aromatic compounds, such as acetone, toluene, and ethylbenzene, presented low concentrations and low risks. In Colombia and South Africa, no pollutants were detected that could harm human health (Figure S12).

Globally, the top two countries with comprehensive human health risks were China and the United States. In China, the landfill pollutants were characterized by high-risk values, rich diversity, and frequent detections, which are related to the rapid development of Chinese economy. Landfill in European countries generally posed a weak threat to workers and nearby residents.

3.3. Factors influencing the health risks of VOCs and SVOCs

After being produced from the biodegradation of waste or from their direct volatilization, VOCs and SVOCs enter the atmosphere through cover soils, thereby adversely affecting the life and health of residents around the landfill sites. Air temperature, relative humidity, air pressure, wind direction, and wind speed are the major factors that influence the health risks of VOCs and SVOCs. High temperatures aggravate the atmospheric pollution of VOCs and SVOCs, as they cause VOCs and SVOCs to diffuse fast in the air, thereby exerting a spatially wider negative impact on the environmental and public health. As water vapor in the air can adsorb VOCs and SVOCs, high relative humidity is not conducive to the diffusion of these compounds, thereby worsening the air pollution over landfills. In contrast, high air pressure decreases the atmospheric concentrations of VOCs and SVOCs, thereby decreasing both carcinogenic and non-carcinogenic risks. Ding et al. (2012) found that the air pollution and health risks of VOCs and SVOCs at a landfill site in Hangzhou severely grew under the high temperature, high relative humidity, and low air pressure. Sea breeze that increases air temperature, relative humidity, and air pressure could also exacerbate the atmospheric pollution of VOCs and SVOCs over landfills in coastal areas. The wind direction and speed determine the diffusion direction and transport rate of VOCs and SVOCs, respectively. The faster the wind speed, the faster the migration rates of VOCs and SVOCs are and the stronger the dilution of the concentrations of VOCs and SVOCs, thereby decreasing health risks. VOCs and SVOCs not only transport in the atmosphere but also undergo chemical reactions under sunlight. Duan et al. (2014) detected more reduced sulfides in the air in winter owing to the decomposition and photochemical reactions of VOCs and SVOCs. Allen et al. (1997) suggested that the emissions of VOCs and SVOCs stimulate photochemical reactions in the air over landfills, thereby accumulating ozone, benzene, and vinyl chloride (Butt et al., 2016). Liu et al. (2016) revealed that BTEX can react with –OH or NO_x to produce secondary organic aerosols, ozone, and other oxides, depending on the intensity of solar radiation, relative concentrations of aromatic compounds, and meteorological conditions. Atmospheric stability play an important role in the pollutant dispersion. Tansel and Inanloo (2019) estimated that their impact range expanded by 194–1164% in stable atmosphere conditions (early morning or night) compared to that in other conditions.

Several studies have been conducted on the transport distance of VOCs and SVOCs from landfills. As shown in Figure S20, using the Gaussian model, Liu et al. (2018) quantified that reduced sulfide can travel as far as 1,000 m. Cai et al. (2015) estimated from 1,955 landfills that VOCs and SVOCs transport in the range of 400–1,000 m. Lu et al. (2013) determined that, among aromatic compounds released from a landfill site in Beijing, benzene can move approximately 1,500 m under normal conditions. China's Pollution Control Standards for Municipal Solid Waste Landfills (Draft for Comments) - Preparation Notes (People's Republic of China, 2022) (Pollution Control Standards., 2022) required a buffer distance of 500–1,500 m for the protection of residential areas from landfill gas. Currently, 5.57% of the landfills in China is located within 500 m of residential areas, with 37.88% within 1,000 m (Table S6), suggesting that landfills are already threatening the health of surrounding residents. The impact range of various VOCs and SVOCs emitted from landfills depends on multiple factors, such as landfill amount, waste composition, management level, geographical location, and meteorological conditions. In addition, complex mechanisms, such as superposition and synergy, play a significant role in the transport of VOCs and SVOCs; however, studies in this field of research are lacking.

4. Summary and outlook

Landfill gas is one of the major sources of VOCs and SVOCs in the environment. This study systematically addressed the composition characteristics, health risks, and influencing factors of VOCs and SVOCs and drawn the following main conclusions:

(1) Landfill gas is a non-negligible source of health risks for landfill workers and nearby residents. Landfill gas poses human health risks in almost all the countries considered, in particular, in China, followed by the developed European countries and the United States.

(2) The aromatic and halogenated compounds are the primary contributors to landfill gas hazards. As these substances primarily originate from the volatilization of chemical products, their hazards to human health can be reduced by improving the utilization rate of chemical products and reducing the total amount of these wastes entering landfills.

(3) The risks posed by compounds such as acetaldehyde, formaldehyde, and ethyl acetate also deserve special attention. Owing to their high concentrations and low thresholds, some VOCs and SVOCs caused human health risks beyond the acceptable levels. Typically, they were frequently detected with high exceedance rates.

(4) The health risks of VOCs and SVOCs from landfills are primarily influenced by air temperature, relative humidity, air pressure, wind direction, and wind speed. Owing to their increased transport distance, VOCs and SVOCs of different concentrations and hazards exerted a wider impact range and adversely affected residents within 1,000 m of landfills.

This paper systematically summarized the composition, source and concentration of VOCs and SVOCs in landfill gas, and has a more complete and clear understanding of them. Potential human health risk assessment was conducted at the global, continental and national levels to fully understand the threat of VOCs and SVOCs to human health in different regions. However, due to the lack of toxicity parameters in other pathways, it is impossible to calculate the human health risk assessment of VOCs and SVOCs through other pathways. In addition, the propagation mechanism of VOCs and SVOCs diffusing from landfills to the air lacks further research. In the future, studies can be conducted to address the following aspects:

(1)Despite the non-negligible human health risks posed by VOCs and SVOCs, most of the previous studies only assessed their respiratory risks, with health risks from other pathways of exposure remaining unclear. Moreover, the lack of certain carcinogenicity parameters for VOCs and SVOCs renders it impossible to estimate their health risks.

(2)The reactions and transport pathways of VOCs and SVOCs in the atmosphere, as well as the mechanisms and roles of interactions between VOCs and SVOCs, remain poorly understood.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

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