Journal of Hazardous Materials 496 (2025) 139549

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Characteristics and health risk-oriented source of VOCs contamination in a typical non-ferrous metal smelting plant and its surrounding area for prioritizing the VOCs control strategies

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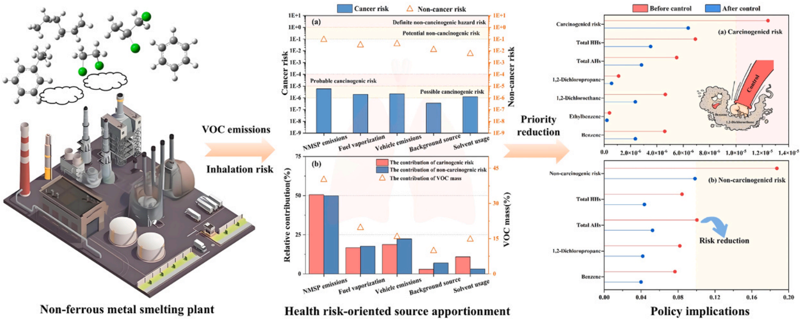
HIGHLIGHTS

- The contaminant characteristics of VOCs in a typical NMSP and its surrounding areas were systematically revealed.
- The concentration was significantly negatively correlated with the distance from the NMSP ($p < 0.01$).
- The carcinogenic risks in NMSP and RA exceed the acceptable risk level.
- NMSP emissions contributed 50.54 % of the carcinogenic risk and 49.84 % of the non-carcinogenic risk in RA.
- Precise control of AHs and HHs can reduce the carcinogenic risk by one category and eliminate non-carcinogenic concerns in RA.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
VOCs
Non-ferrous metal smelting
Spatial distribution
Health risk-oriented source apportionment
Mitigation policy

ABSTRACT

Non-ferrous metal smelting plant (NMSP) discharges quantities of volatile organic compounds (VOCs) into the surrounding residential areas (RA). A total of 92 VOC species were identified. VOC concentrations in NMSP ranged from 88.1 to 154.4 $\mu g/m^3$, with aromatic hydrocarbons (AHs) and aliphatic hydrocarbons (AlHs) accounting for more than 75.0 % of TVOCs. Spatial distribution revealed a significant negative correlation between VOCs concentration and distance from NMSP ($p < 0.01$), and contaminants predominantly transported southward over short and medium distances. Health risk assessment revealed that the inhalation carcinogenic risks in NMSP and RA were 3.0×10^{-5} and 1.2×10^{-5}, exceeding the acceptable risk level (1.0×10^{-6}). Benzene (AHs), 1,2-dichloroethane (halogenated hydrocarbons; HHs), and 1,2-dichloropropane (HHs) were the primary contributors. Health risk-oriented source apportionment further showed that NMSP emissions contributed 50.54 % of the carcinogenic risk and 49.84 % of the non-carcinogenic risk in RA. Further prioritizing the control efficiency of AHs and HHs emissions from NMSP to over 96.1 % and 97.1 % would reduce the carcinogenic risk level by one

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https://doi.org/10.1016/j.jhazmat.2025.139549
Received 18 June 2025; Received in revised form 5 August 2025; Accepted 11 August 2025

risk category and eliminate non-carcinogenic concerns in RA. Greater emphasis should be placed on high-risk compounds, rather than solely on high-concentration compounds. This study provides critical insight for prioritizing VOC control strategies to maximize environmental and health co-benefits.

1. Introduction

According to the World Health Organization (WHO), volatile organic compounds (VOCs) are defined as organic compounds with boiling points in the range of 50 – 260 ℃ [42]. VOCs primarily exist in the troposphere, which can react with nitrogen oxides by photochemical reaction, acting as key precursors for ozone and fine particulate matter formation [10,9]. Previous studies have identified several primary VOC emission sources, including vehicle emissions, fuel combustion, solvent usage, and industrial emissions [36]. Particularly, industrial emissions are an important contributor to VOC levels, with studies indicating that industrial sources constitute up to 51.3 % of total VOC emissions in economically developed regions of China [46]. Furthermore, it is particularly concerning that anthropogenic VOCs emitted from industrial areas can impact surrounding regions through atmospheric dispersion, significantly increasing exposure concentrations of VOCs, and thereby increasing the health risk for nearby residents [53]. Residents near these industrial areas are constantly exposed to VOCs.

VOCs enter the human body primarily through inhalation, leading to carcinogenic effects and health risks. These compounds can cause chronic damage to the respiratory, hematopoietic, and nervous systems, potentially resulting in diseases such as asthma [20,58]. Particular attention should be paid to high-risk VOCs due to their heightened toxicity and potential for long-term harm. However, it is important to note that not all VOCs have the same level of toxicity to humans. The toxic effects of VOCs are not only related to their concentration but also closely associated with factors such as composition, molecular structure, and functional groups of VOCs [37]. Zheng et al. demonstrated that benzene, oxygenated VOCs, and 1,2-dichloroethane are classified as carcinogens and potentially toxic substances [68]. Chronic exposure to benzene and 1,2-dichloroethane can lead to a reduction in hematopoietic stem cell counts and their colony-forming abilities, as well as cause gastrointestinal hemorrhage [39,47]. Conversely, isobutane, a commonly occurring VOC, exhibits no carcinogenic effects [19]. Therefore, accurately characterizing the VOCs concentration and toxicological profiles is crucial for effective health risk assessment and informed policy development for VOCs management and control.

China, as the largest global producer and consumer of non-ferrous metals [25], has continuously increased its production, with output reaching 79.2 million tons in 2024, representing a 4.3 % increase over the previous year (www.miit.gov.cn). Current research on industrial VOC emissions predominantly focuses on sectors such as petrochemical refining [14], chemical manufacturing [5], and coking [53]. In contrast, studies on atmospheric pollutants from the non-ferrous metal smelting industry have primarily focused on heavy metals and gaseous mercury [48], with limited attention paid to VOC emissions. Non-ferrous metals are primarily produced through pyrometallurgical and hydrometallurgical processes, including smelting, anode refining, and electrolytic refining [30]. In these processes, VOCs are inevitably produced. In these processes, VOCs are inevitably produced. For example, coal combustion, which is frequently used as fuel during the smelting process, releases substantial aromatic VOC emissions [6]. During electrolysis refining, organic solvents are commonly utilized as diluents and extractants for metal separation and ion purification, resulting in the generation of a substantial amount of VOCs [44]. VOC emissions from the metal smelting and rolling industry have reached approximately 101,000 tons per year in China [7]. The emission of numerous VOCs during the smelting process can spread to nearby residential areas, posing a health risk to residents surrounding the NMSP. Therefore, characterizing VOC emissions from NMSP and adjacent areas is crucial for accurately assessing associated health risks for both NMSP workers and nearby residents.

However, current regulatory policies often employ uniform standards for controlling fixed species VOC emissions, and primarily focus on high-concentration compounds. These policies generally lack source-specific management strategies as well as prioritized regulation of contaminants that pose high health risks [57]. For example, in the Yangtze River Delta region of China, it would be more effective to not only prioritize the control of benzene emissions according to the national standard, but also to focus on the control of ethylbenzene and 1,2-dichloropropane emissions from the paints and coatings industry due to their high toxicity [38]. Many residents living near industrialized areas are at risk of VOC exposure [31], which indicates that the current regulatory policies need to be further optimized. It is of particular concern that the synergistic effect of multiple VOC sources on resident health risks. Therefore, the development of a health risk-oriented VOC source apportionment method is crucial for quantitatively evaluating the contribution of different emission sources to the overall health risks faced by residents. This is of great significance for the formulation of effective and targeted VOC prevention and control strategies.

This study investigates a representative NMSP and its surrounding area, aim to: (1) characterize VOCs emissions from NMSP; (2) explore the spatial distribution and dispersion trajectory of VOCs emitted by NMSP; (3) identify potential sources of hazardous VOC emissions using a health risk-oriented source apportionment; (4) identify critical VOCs emission sources affecting the health of workers and residents; (5) provide recommendations to enhance VOCs prevention and control. The findings of this study provide critical insights for air quality management and environmental restoration efforts in industrialized regions and nearby residential areas.

2. Methodology

2.1. Sample collection area

We selected a representative NMSP to collect atmospheric samples in Hubei Province, China. The sampling area was centered around the NMSP, extending over an 8 × 7 km rectangular area. Sampling was carried out from April 2021, with predominantly cloudy and sunny weather conditions (Table S1). In order to ensure comprehensive and representative sampling, the sampling area was divided into three categories based on population density and land use type: inside the NMSP, surrounding residential areas (RA), and control areas (CA) located approximately 20 km away from NMSP. During the sampling process, we collected 40 atmospheric samples. Fig. S1 provides the specific locations of all the sampling sites in detail. All the NMSP samples were collected outside the workshop. Ambient air samples were gathered utilizing 2.7 L electropolished Summa canisters (Entech Instruments Inc., USA).

2.2. Sample analysis

For the analysis, 1 ppm standard gas from the photochemical assessment monitoring station (PAMS) and 1 ppm TO-15 (Linde Spectra Environment Gases, USA) were injected into pre-concentration gas chromatography-mass spectrometry (GC-MS) system (7890B GC-5977C MS, Agilent Technologies, USA), and the concentration calibration curve was drawn. Qualitative and quantitative analysis was performed using the NIST database and the retention time of each compound [53]. The method detection limit (MDL) of each compound was 0.02–0.31 μg/m^3.

Detailed analysis methods are provided in the SI.

2.3. Quality assurance and control

In order to ensure the cleanliness of the summa canisters before sample collection. We first used high-purity nitrogen (purity of 99.99 %) to thoroughly clean the canisters five times, and pumped the canisters to a vacuum state (internal pressure less than 10 Pa) after the cleaning. A strict leak detection procedure was performed on all canisters to prevent leakage. To minimize uncertainties during analysis, after every 10 atmospheric samples and an additional blank sample from a group, an internal standard was added to all samples before analysis. In addition, the instrument was calibrated before use to ensure optimal performance, including the required resolution, qualitative ability, and sensitivity. On-site blank canisters were also prepared, and levels of target compounds detected in all blank samples were below the minimum detection limit (MDL) of each compound.

2.4. Data analysis

For data analysis, we used IBM SPSS Statistics 26 software to perform correlation analysis and non-linear fitting analysis. Source analysis was conducted using the Positive Matrix Factorization (PMF) method to accurately identify and quantify the contribution of source species and profiles of VOCs in the research area. To evaluate the potential inhalation risks of VOCs to human health, we calculated both cancer risk and non-cancer risk of specific contaminants. Oracle Crystal Ball 11.1.3.0.0 was used for the Monte Carlo simulation. Under the condition of uncertain information, random phenomena were simulated using an appropriate probability distribution. A detailed description analysis process, including the model and methods used, is available in the SI.

3. Results and discussion

3.1. Characteristics of VOC emissions from NMSP

A total of 92 VOCs were detected in the collected samples (Table S4). Based on their chemical structures and element types, the detected VOCs were categorized into five groups: aliphatic hydrocarbons ($\sum_{31}$ AIHs), aromatic hydrocarbons ($\sum_{18}$ AHs), halogen hydrocarbons ($\sum_{34}$ HHs), oxygenated VOCs ($\sum_{8}$ OVOCs), and carbon disulfide (Ss). Fig. 1 and Fig. S2 illustrate the atmospheric concentrations of various VOCs in

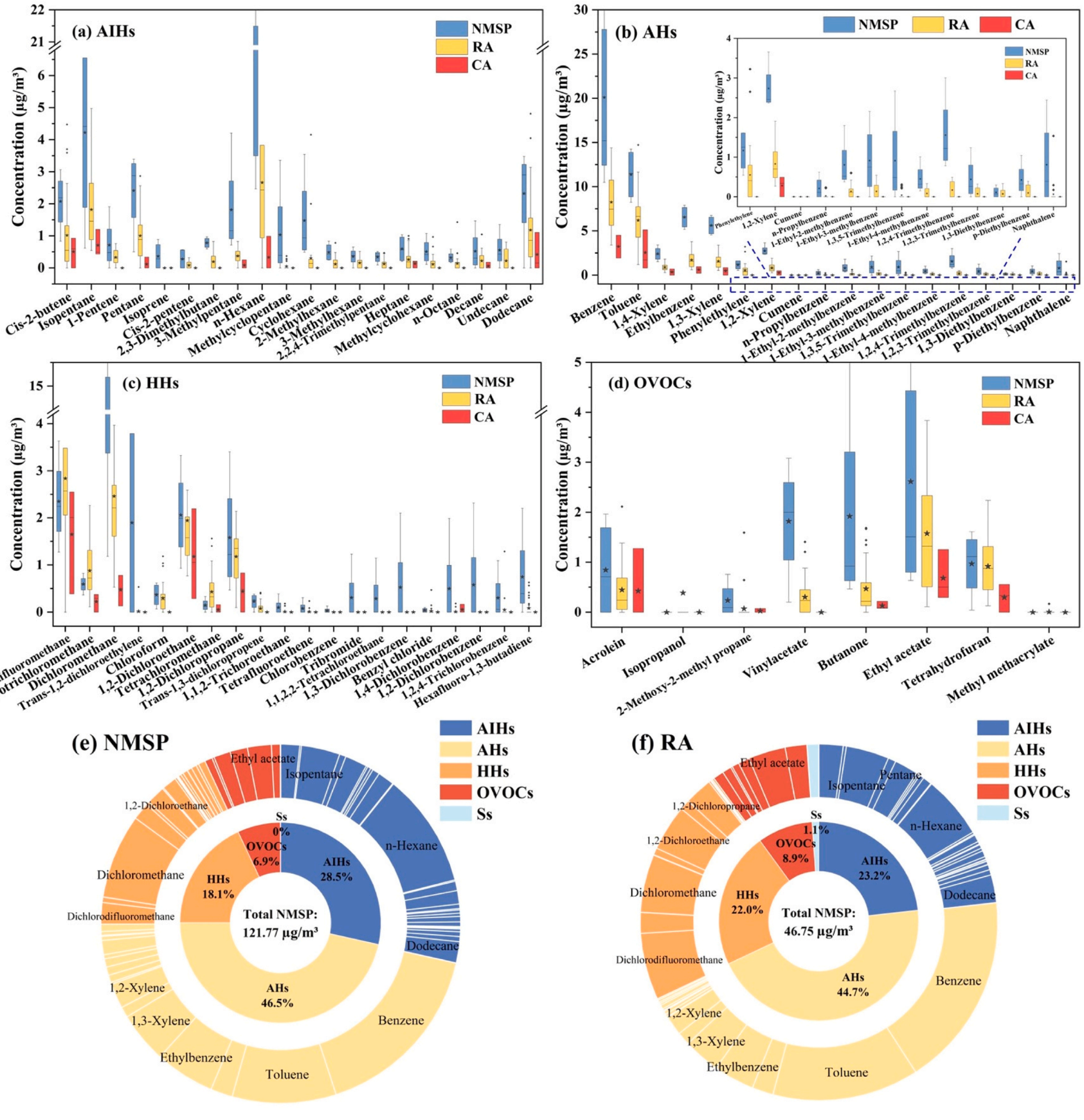

Fig. 1. The contamination levels of the main contaminants of AHs (a), AIHs (b), HHs (c), and OVOCs (d) in different functional zones, and the concentration proportions of the compounds in NMSP (e) and SA (f).

NMSP. The concentration of TVOCs in NMSP ranged from 88.1 to 154.4 μg/m^3 . The detection frequencies of AIHs, AHs, HHs, OVOCs, and Ss in the four workshops were 70 %, 79 %, 35 %, 63 %, and 25 %, respectively. AHs were identified as the predominant contaminants from smelting emissions, with their concentration reaching 56.6 ± 19.8 μg/m^3 , of which the average concentrations of benzene, toluene, and ethylbenzene reached 20.1, 11.4, and 6.6 μg/m^3 , respectively. To investigate atmospheric VOCs contamination levels around the NMSP, the compound concentration characteristics across different functional areas are presented in Fig. 1 and Fig. S3. The study found that the concentrations of TVOCs in NMSP, RA, and CA were 121.8 ± 30.5, 46.7 ± 16.0, and 15.5 ± 8.6 μg/m^3 , respectively, and AHs in NMSP were 2.7- and 7.5-fold that of RA and CA, respectively. The VOC concentrations in NMSP were significantly higher than those in RA and CA. During the smelting process, coke combustion is necessary to dissolve metal and remove impurities at high temperatures. However, substantial emissions of VOCs are generated through both incomplete combustion of fossil fuels Peng et al. [41] and the presence of organic waste materials in raw materials [45,63].

In addition, the emissions of AIHs and HHs from the NMSP need to be given special attention. The concentrations of AIHs and HHs reached 34.7 and 22.0 μg/m^3 , respectively, with n-hexane and dichloromethane exhibiting the highest concentrations within these groups, ranging from 2.5 to 33.9 μg/m^3 and 1.2–17.3 μg/m^3 . Comparing different functional areas, AIHs and HHs in NMSP were significantly higher than those in RA and CA ($p < 0.05$). Specifically, the AIH levels in NMSP were 3.2- and 14.7-fold higher than those in RA and CA, respectively, while HH concentrations were 2.1- and 5.4-fold higher, respectively. Benzene and dichloromethane were the compounds with the highest concentrations among the AIHs and HHs, respectively, and their concentration in NMSP were 4.7- and 3.8-fold that of in RA, and 37.9- and 19.6-fold that of in CA, respectively. These findings indicate substantial AIH and HH emissions during metal smelting processes. In the electrolysis workshop for non-ferrous metals, VOCs compounds such as dichloromethane, n-hexane, and toluene are utilized as electrolyte additives and extractants to improve metal purity. However, this process leads to the leakage or release of VOCs, thereby resulting in increased workers' exposure risks. Chang et al., [4].

In order to better reflect the contamination level of VOCs emitted by NMSP, the VOC emission characteristics from NMSP were compared with other industries (Table S5). The VOC concentrations emitted from NMSP rank at an upper-medium level among industrial emission sources. Compared with the coking plant [53] and the shipbuilding industry [27], the concentration of AHs emitted by NMSP is lower, but it is higher than that of the petrochemical refinery [14]. This elevated level of AHs is primarily attributed to incomplete coal combustion during metal smelting and refining processes [52], as well as to the presence of naturally occurring organic matter in the raw materials and organic flotation agents from mineral separation [45,63]. Moreover, the concentration of AIHs in NMSP is higher than that of the coking plant, automobile industry [27], shipbuilding industry, battery material plant [35], and electronic waste dismantling [32], which is primarily attributed to the use of organic solvents as cleaning agents and electrolyte additives, such as n-hexane. The dominant contaminants in various industries are closely linked to their respective technological processes.

3.2. *The exposure level of VOCs in the residential areas around NMSP*

To better assess the impact of the NMSP on surrounding residential areas, we compared the average concentrations of VOCs in RA and CA, both of which are residential areas (Table S6 and Fig. S4). TVOCs in RA were 3.0-fold higher than those in CA. AHs and AIHs were the dominant compounds emitted from NMSP, exhibiting 2.8- and 4.6-fold higher concentrations in RA than in CA, respectively. Notably, AIHs exhibited the highest incremental exposure among all compound groups. Benzene, toluene, n-hexane, and dichloromethane, which contributed significantly to the TVOC in NMSP, these compounds in RA were 2.6-, 2.4-, 8.1-, and 5.2-fold higher than in CA, respectively. Overall, the VOC concentration levels in RA were significantly higher than those in CA ($p < 0.05$). In RA, AHs accounted for the largest proportion, reaching 44.7 %, which was consistent with the VOC characteristics observed in industrial cities influenced by metal smelting [49]. VOCs emitted from smelting and refining processes, electrolytic processes, and solvent extraction used in non-ferrous metal smelting operations disperse into the surrounding environment via atmospheric transport, thereby posing a threat to nearby residents [53].

In order to further investigate the residential exposure levels near the NMSP, we compared the VOC concentrations in RA with those in residential areas around the world (Table S7). By comparison, it was found that the TVOC concentration of RA was comparable to those reported in a rural town in southeastern Nigeria [1] and a metropolitan city in Seoul, South Korea [26]. It was lower than the levels observed in traffic-dominated areas in Beijing, China [33] and Cheras, Malaysia [69], as well as in a coking contaminated area in Shanxi, China [53]. Conversely, TVOCs concentration in RA was higher than in the commercial-residential area in southern Burnaby, Canada [56] and the control area in La Plata, Argentina [28]. These comparisons indicate that the level of TVOC contamination in RA around NMSP is relatively high and requires increased attention.

3.3. *Spatial distribution characteristics of VOCs*

To visually observe the diffusion of VOCs emissions from NMSP to surrounding areas, the spatial distribution maps of various VOCs groups were drawn using Inverse Distance Weighting. As shown in Fig. 2 and Fig. S5, the spatial distribution of AIHs, AHs, HHs, OVOCs, and TVOCs were observed across the entire monitoring area. The NMSP area showed significant red coloration, demonstrating significantly higher concentrations of TVOCs, AIHs, and AHs compared to surrounding areas. Both the NMSP and RA were hotspots of TVOCs, AIHs, and AHs, suggesting that activities associated with NMSP may significantly contribute to contamination in RA. The spatial distribution diagrams of TVOCs, AIHs, and AHs clearly demonstrated that the residential area on the right side of NMSP was affected by metal smelting, and its concentration was higher than in other areas. This observation is consistent with the conclusion drawn by Deng et al., who observed similar spatial trends involving polycyclic aromatic hydrocarbons and their derivatives near NMSP [12]. VOCs were discharged into the atmosphere, and they would enter the surrounding environment through atmospheric diffusion, resulting in an increased exposure risk of VOCs near NMSP.

Analysis of the diffusion direction of VOCs (Fig. 2 and Fig. S5) through the spatial distribution map shows that four types of contaminants, TVOCs, AIHs, AHs, and OVOCs, mainly diffused eastward, seriously affecting the residential areas in the east. The diffusion pattern might be related to the local topography and perennial wind direction. There were mountains in the north and west of the study area. Due to the obstruction of the mountains, the atmospheric flow was not smooth, and the diffusion of contaminants to the northwest was blocked, thus resulting in the accumulation of contaminants around the east NMSP. Yuan et al. also found similar diffusion phenomena in the study of VOCs in Gaoxiong [59]. Additionally, during the monitoring period, the local winds were predominantly westerly (Fig.S6). The synergistic effects of topographical barriers and wind direction significantly contributed to the eastward dispersion of VOCs [43]. Notably, the concentration of HHs near the residential area in the southwest of NMSP is relatively high, which may be attributed to the presence of other outdoor emission sources, such as the use of crop pesticides [22,4].

To deeply understand the relationship between VOC concentration and diffusion distance, a curve fitting method was employed. The relationship between distance and concentration data was evaluated by thousands of iterations using various functions. The function $Con = a + b \times EXP(Dis/c)$ was selected. As depicted in Fig. 2 and Fig. S5, AIHs

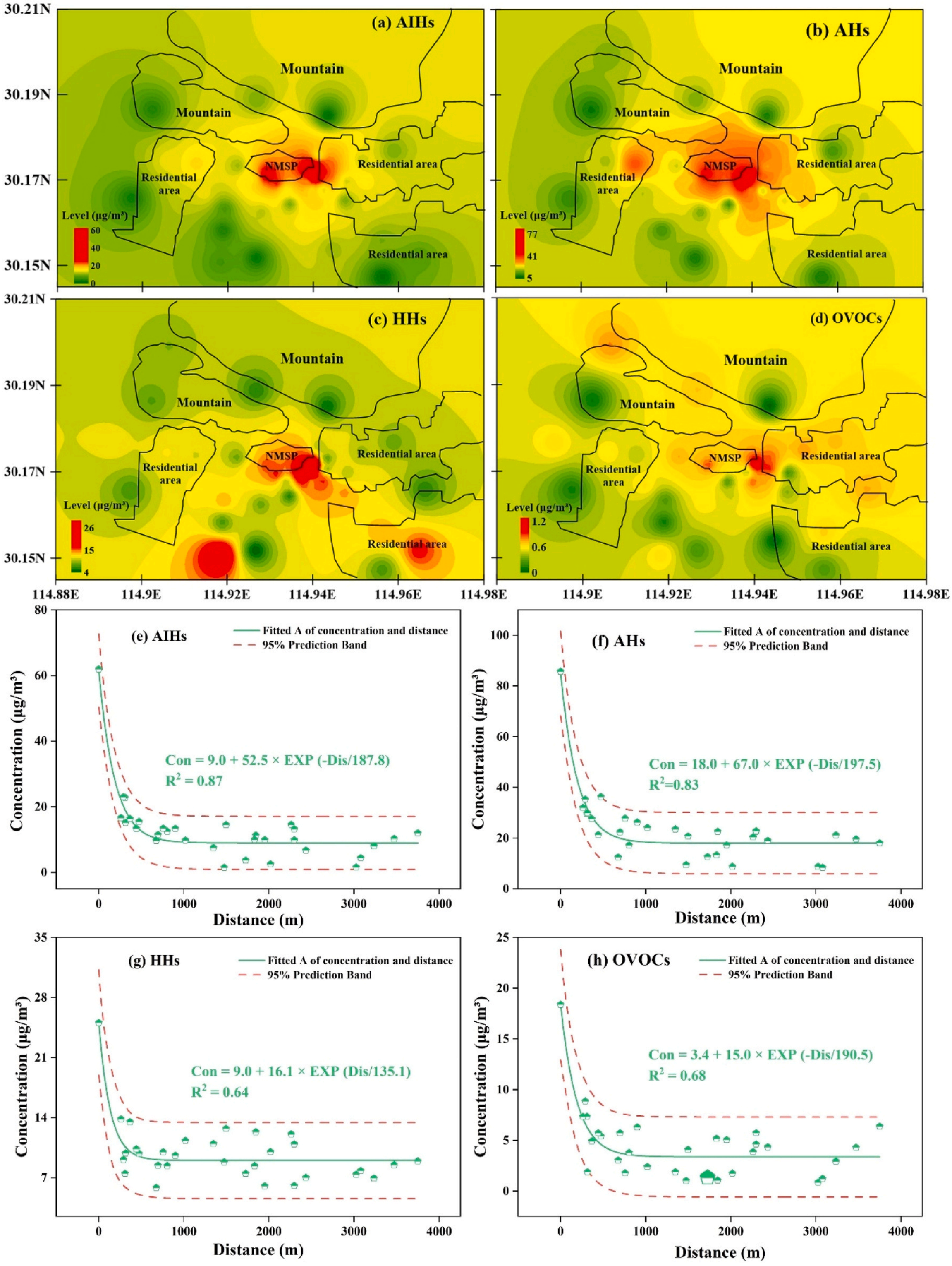

Fig. 2. Spatial distributions of (a) AIHs, (b) AHs, (c) HHs, and (d) OVOCs concentrations (μg/m^3); (e-h) spatial fitting analysis of AIHs, AHs, HHs, and OVOCs concentrations with non-fitting to distance from the NMSP.

($R^2 = 0.87, p < 0.01$), AHs ($R^2 = 0.83, p < 0.01$), and TVOCs ($R^2 = 0.77$, $p < 0.01$) exhibited strong fitting effects. Whereas HHs ($R^2 = 0.64$, $p < 0.01$) and OVOCs ($R^2 = 0.68$, $p < 0.01$) demonstrated a moderate fitting result. In these curves, the a value more accurately represents local VOC exposure levels, while the b value, which is modulated in the logarithmic function, better reflects the rate at which concentration decreases with increasing distance. Due to intensive emissions from NMSP, the a values for TVOCs and AHs were significantly higher than those for other groups. Furthermore, compared with the b value, the concentrations of these two major groups exhibited a more pronounced decrease with increasing distance. Overall, within a radius of 1000 m, the concentration of contaminants sharply decreased with distance, whereas beyond 1000 m, the VOC concentration became less affected by the change in distance [12]. The results showed that the VOCs emitted by the metal smelting process of NMSP diffused into the surrounding areas via atmospheric transport, and the surrounding residential areas were contaminated to varying degrees depending on their distances from NMSP.

Meteorological conditions significantly affect the VOCs diffusion dynamics. In order to explore the long-distance migration trajectories of VOCs emitted by NMSP, we used a forward trajectory model to calculate [18]. By integrating the trajectories throughout the entire observation period, it can be clearly shown in Fig. S7 that the diffusion processes were predominantly governed by five air masses. Influenced by westerly and southeasterly winds, VOCs emitted by NMSP were predominantly transported to the south. Medium- and short-distance transport accounted for 76.39 % of total movements, with nearly 80 % of the air mass being transported to the south. Specifically, the trajectory in Jiangxi province (green and purple pathways, 43.05 %), the trajectory in Hubei province (blue pathway, 18.06 %), and the trajectory in Hunan province (red pathway, 15.28 %) were relatively short, and the speed of movement was relatively slow. A smaller portion (5.56 %) exhibited long-distance transport toward the northwest (yellow pathway), and lower VOC concentrations implied a limited impact on the northwestern regions. Therefore, it was necessary to implement stringent emission control of VOCs from NMSP to improve air quality in the southern areas.

3.4. Source apportionment

3.4.1. Diagnostic ratio of specific compounds

Potential emission sources of contaminants can be identified through the ratio of specific compounds [23]. Fig. S8 and Fig. S9 show the correlation and scatter plots of VOC components. It is evident that benzene and toluene, as well as isobutane and n-butane, exhibited a significantly strong positive correlation ($p < 0.01$). The ratio of toluene to benzene (T/B) was often used to distinguish potential combustion sources [2]. The T/B ratio is less than 2, indicating combustion sources as the main VOCs emission contribution. Specifically, the T/B ratio below 1 suggests predominant contributions from coal and biomass combustion [34]. The T/B of all samples ranged from 0.17 to 1.9, with an average of 0.7 (Fig. S9), indicating that emissions in RA were significantly influenced by coal combustion activities in NMSP and biomass combustion in the residential area. Another diagnostic ratio, the ratio of isopentane/pentane (i/p), can be applied to distinguish the primary source of VOCs. The i/p ratio was in the range of 1.8–4.6 represents fuel evaporative emissions [29,34], while a ratio between 1.5 and 3.0 represents vehicle emissions [51]. The observed i/p ratios ranged from 0.61 to 4.84 (average = 1.8), predominantly between 1.5 and 4, suggesting that both fuel evaporative and vehicle emissions are important contributors. Therefore, the diagnostic ratio analysis collectively indicated coal combustion (from NMSP), fuel evaporation, and vehicle emissions as the primary VOCs sources in the study area.

3.4.2. PMF source apportionment

In this study, the EPA PMF5.0 version was used to analyze the sources apportionment of VOCs in RA, determining the contributions from each potential source. 41 representative VOCs with a detection frequency exceeding 40 % were input into the PMF model [60]. Five factors were determined according to the change rate of Q_{ture} / Q_{robust} (Fig. S10). As shown in Fig. 3a, the five identified factors were identified as background source, solvent usage, NMSP emissions, fuel vaporization, and vehicle emissions.

Factor 1 primarily included dichlorodifluoromethane (65.49 %) and fluorotrichloromethane (48.51 %). Fluorinated compounds usually have a prolonged atmospheric lifetime [16,53]. Therefore, the factor 1 was identified as the background source. Factor 2 was mainly composed of benzene (70.22 %), n-hexane (67.08 %), toluene (59.20 %), and dichloromethane (54.44 %). These compounds were the primary compounds detected in NMSP. Furthermore, Wang et al. also reported significant proportions of benzene and toluene in metal smelting emissions [49]. Therefore, Factor 2 was attributed to NMSP emissions. Factor 3 featured primarily C_4-C_5 alkanes and alkenes. Specifically, it included pentane (53.14 %), 2,3-dimethylbutane (47.76 %), and isopentane (52.77 %). Pentane is the most abundant VOCs species in gasoline evaporation emissions [67]. Additionally, Wu et al. identified C_4-C_5 alkanes and alkenes, including pentane, isopentane, and 2-methylpentane, as typical indicators of fuel vaporization [54]. Consequently, factor 3 was attributed to fuel vaporization. Factor 4 mainly contained long-chain alkanes, such as undecane (87.12 %), n-octane (60.38 %), and decane (68.35 %), which are recognized tracers of diesel combustion [2,43]. Considering heavy traffic and vehicular activities in the study area, factor 4 was determined to be associated with vehicle emissions. The characteristic species of factor 5 were primarily ethyl acetate (64.16 %), tetrachloromethane (61.16 %), and chloroform (51.75 %). Previous studies indicate that chloroform and tetrachloromethane, as two of the most common organic solvents in industry, are often used in industrial production [22]. Moreover, chlorinated hydrocarbons are widely applied as solvents in manufacturing processes, coatings, and inks [17]. Therefore, factor 5 was identified as solvent usage.

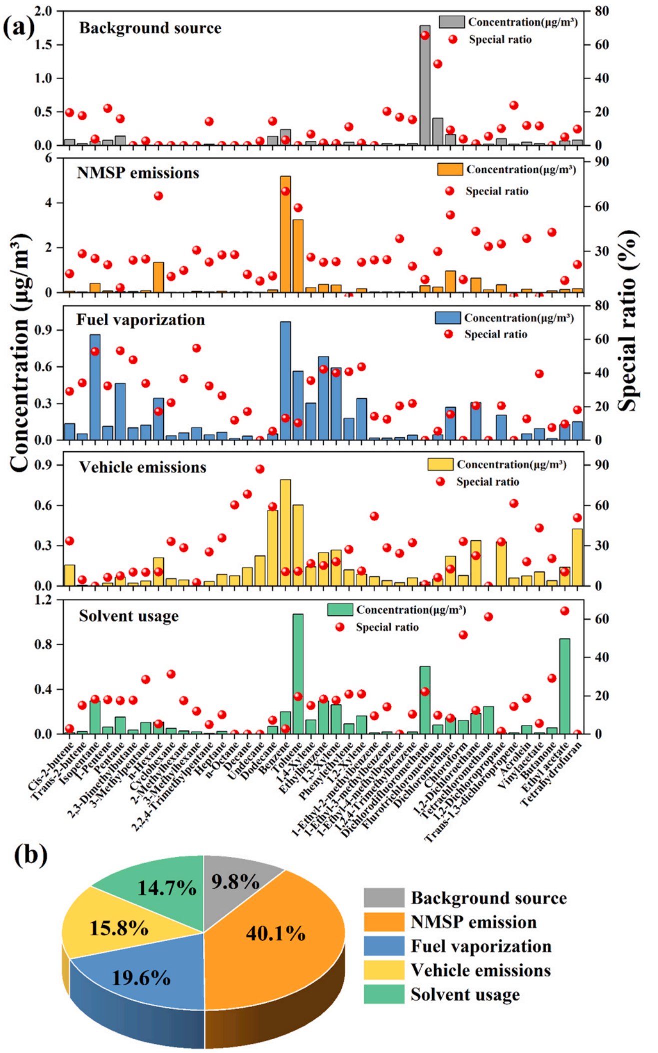

Fig. 3. The source analysis of VOCs (a) and the specific contribution of each source (b) obtained from the PMF model.

Fig. 3b displays the source contribution percentages in RA. In residential areas, NMSP emissions were the primary contributors, with a contribution rate of 40.1 %. NMSP has emerged as the dominant VOCs source due to the abundance of coal combustion and the use of organic solvents. The strong positive correlations observed between benzene, n-hexane, toluene, and dichloromethane in NMSP and RA (Fig. S8) provide evidence that these compounds share a common emission source [21], emphasizing the direct impact of NMSP emissions on the living environment of nearby residents. Fuel vaporization represented the second largest contributor to VOCs, accounting for 19.6 % of TVOCs contamination. Cooking activities by local residents and fuel evaporation from large cargo trucks likely contribute to the elevated proportion of this source within the residential area. These findings are consistent with both the PMF analysis and the diagnostic ratio method. In addition, vehicle emissions, solvent usage, and background source contributed 15.8 %, 14.7 %, and 9.8 % to TVOC contamination, respectively. Consequently, it is essential to control VOC emissions at NMSP and optimize fuel consumption to lower atmospheric VOC levels and enhance air quality in the studied areas.

3.5. *Inhalation health risk assessment*

In order to assess the health risks associated with VOCs exposure from NMSP and its surrounding environment, inhalation carcinogenic risks (ICR) were assessed for 16 selected compounds, while non-carcinogenic hazard quotients (HQ) were calculated for 21 selected compounds [24,64]. As shown in Fig. 4 and Table S10, the carcinogenic hazard ratio values ranked as follows: NMSP (3.0×10^{-5}) $>$ RA (1.2×10^{-5}) $>$ CA (5.2×10^{-6}). The carcinogenic risks in all three areas exceeded the threshold of 10^{-6}, indicating a possible carcinogenic risk level [65]. The carcinogenic risks at NMSP were 2.5- and 5.77-fold higher than those in RA and CA, respectively. Additionally, compared with residential areas, the carcinogenic risk in RA was 2.3-fold greater than that in CA, indicating that residents surrounding the NMSP were more significantly affected by NMSP emissions. Among 16 selected VOCs for carcinogenic hazard risk assessment, benzene, 1,2-dichloroethane, and naphthalene posed a potential carcinogenic risk (1×10^{-5} - 1×10^{-4}), contributing 74.2 % to the total carcinogenic risk in NMSP, these compounds significantly affect the health of workers within NMSP. In addition, ethylbenzene, 1,2-dichloropropane, 1,1,2,2-tetrachloroethane, and hexafluoro-1,3-butadiene were also categorized as posing a possible cancer risk, contributing 5.0 %, 4.8 %, 5.0 %, and 5.0 %, respectively. In RA, benzene, 1,2-dichloroethane, and 1,2-dichloropropane were identified as the dominant carcinogenic contaminants, contributing 83.1 % of total carcinogenic potential. This composition exhibits a substantial overlap with the primary carcinogenic compounds identified between NMSP and RA.

As shown in Fig. 4c, non-carcinogenic hazard ratios were highest at NMSP (0.52), followed by RA (0.19) and CA (0.06). The non-carcinogenic risk associated with exposure levels within NMSP was 2.74- and 8.67-fold higher than in RA and CA, respectively. The non-carcinogenic risk within the NMSP exceeded the safety threshold of 0.1, indicating significant health concerns [66], in which benzene and 1,2-dichloropropane contributed the most to this risk, accounting for 35.3 % and 20.8 %, respectively. Additionally, compared with RA and CA, the non-carcinogenic risk levels in RA exceeded the threshold of 0.1, indicating potential health risks for residents across the RA monitoring area, with benzene and 1,2-dichloropropane risks exceeding 2.7-fold in CA. CA did not exhibit non-carcinogenic risks. The carcinogenic risk in RA was higher than that reported in traffic-dominated areas [69] and metropolitan cities [26]. The emission of VOCs from NMSP had significantly increased the health risk level for the surrounding residents.

Fig. S12 illustrates the health risks among populations of different age groups. It shows that adults have the highest carcinogenic risk from

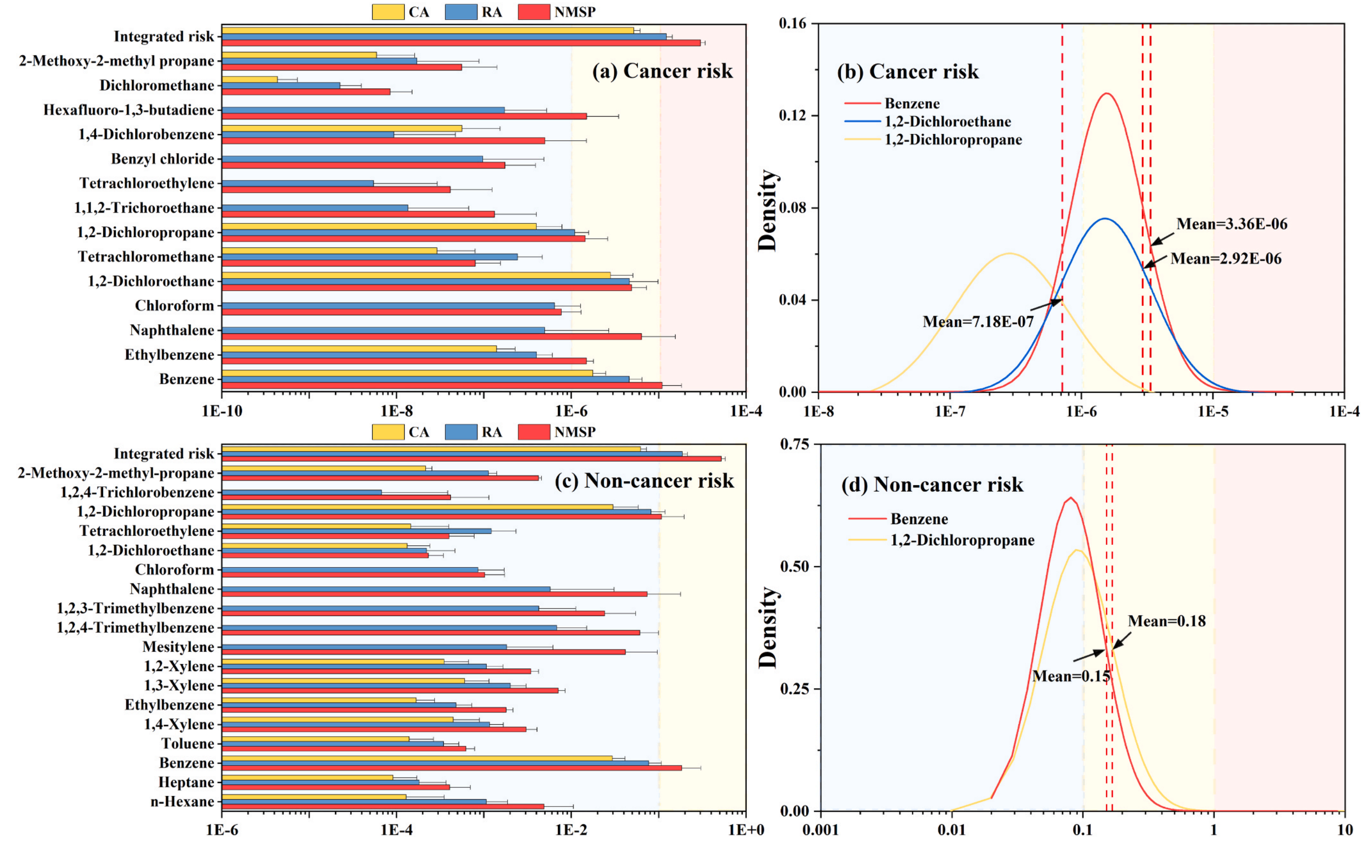

Fig. 4. (a) Carcinogenic risks and (c) non-carcinogenic risks in various functional areas. The probability distribution diagram simulated via Monte Carlo simulation, (c) represents the carcinogenic risk in RA, and (d) represents the non-carcinogenic risk in RA. In carcinogenic risk, the blue, yellow, and red areas represent negligible risk ($ICR < 10^{-6}$), possible risk ($10^{-6} < ICR < 10^{-5}$), and probable risk ($10^{-5} < ICR < 10^{-4}$), respectively. In non-carcinogenic risk, the blue, yellow, and red areas represent negligible risk ($HQ < 0.1$), probable risk ($0.1 < HQ < 1$), and definite hazard risk ($HQ > 1$), respectively.

VOC exposure in NMSP, with TVOCs posing a carcinogenic risk of up to 1.5×10^{-4}. This indicates a significant direct carcinogenic risk that substantially increases the health hazards for long-term employees in NMSP. Comparing different age groups, the carcinogenic risk follows the order: adults > children > adolescents. This difference primarily results from adults experiencing exposure times approximately 5.8 times longer than those of children and adolescents, significantly increasing the inhaled contaminant exposure for adults [24]. The RA region shows potential carcinogenic and non-carcinogenic risks across all age groups, highlighting concerns regarding the environmental quality around NMSP.

Considering parameter variability and uncertainty, we used a Monte Carlo simulation to estimate the probability distribution of carcinogenic and non-carcinogenic risks for compounds that critically impact residential areas (RA). Probability risk and sensitivity analyses for benzene, 1,2-dichloroethane, and 1,2-dichloropropane are displayed in Fig. 4 and Fig. S13. Benzene exhibited the highest average carcinogenic risk (3.08×10^{-6}), followed by 1,2-dichloroethane (2.94×10^{-6}). The average non-carcinogenic risk of benzene and 1,2-dichloropropane were 0.15 and 0.18, respectively, exceeding the safety level. The probability of long-term exposure to benzene, 1,2-dichloroethane, and 1,2-dichloropropane exceeding the safe limits was 90.96 %, 88.78 %, and 24.37 %, respectively. The probability of long-term exposure to benzene and 1,2-dichloropropane exceeding the non-carcinogenic safe limits was 55.41 % and 61.47 %. The results of the sensitivity analysis revealed that contaminant concentration was the most influential parameter on risk estimation, followed by IUR value. Dai et al. also obtained similar results while evaluating the carcinogenic risk of indoor inhalation [11].

Compared with other industrial zones, the health risks of VOC emissions from NMSP are at a moderate level. Although the carcinogenic risk is lower than that reported for coking plants [53] and e-waste dismantling sites [32], it is comparable to that of petrochemical refineries [14], and notably higher than that of the automobile industry [50] and battery material plants [35]. This comparison underscores the need for increased attention to VOC emissions from the NMSP.

Based on the health risk assessment described above, to prevent potential harm of VOCs emissions from NMSP, it is crucial to strictly control high-concentration VOCs like n-hexane, isopentane, benzene, and toluene. Additionally, particular attention must be paid to naphthalene and HHs, including 1,2-dichloroethane, 1,2-dichloropropane, 1,1,2,2-tetrachloroethane, and hexafluoro-1,3-butadiene at NMSP, which pose significant health risks even at low exposure levels due to their high carcinogenic/non-carcinogenic risk coefficients.

3.6. Health risk-oriented source apportionment

VOCs contaminant sources in RA region include background source, NMSP emissions, fuel evaporation, vehicle exhaust, and solvent usage, each source has a distinct impact on residents' health risks. Therefore, it is crucial to accurately quantify the contribution of each VOCs source to overall health risks. This understanding will facilitate the development of targeted VOC control strategies and mitigate potential health hazards. In this study, a health risk-oriented source apportionment method was employed to evaluate the contribution rates of various VOCs sources to residents' health risk [2,57]. As shown in Fig. 5a, the carcinogenic risk values of different VOCs sources ranged from 3.5×10^{-7} to 5.87×10^{-6}. Except for the background source at a safe level, all other sources reached potential carcinogenic risk thresholds. Notably, vehicle exhaust has emerged as another important health risk source following NMSP emissions. Non-carcinogenic risk values for various VOCs sources ranged from 0.006 to 0.09, all below the potential non-carcinogenic risk threshold of < 0.1. Compared with various emission sources (Fig. 5b), the sum of carcinogenic risk of NMSP emissions (50.54 %), vehicle emissions (18.75 %), and fuel vaporization (16.77 %) exceeded 80 % of the total carcinogenic risk. Specifically, VOCs emitted from NMSP account for over half of the health risk in nearby residential areas. Regarding non-carcinogenic risk, NMSP emission, fuel evaporation, and vehicle emission contributed 49.84 %, 17.68 %, and 23.71 %, respectively. Vehicle emissions were identified as another critical non-carcinogenic risk source after NMSP emissions. Therefore, to effectively control VOCs contamination in RA, it is imperative to strictly reduce VOC emissions from NMSP and enhance the regulatory oversight of other emission sources, such as vehicle exhaust.

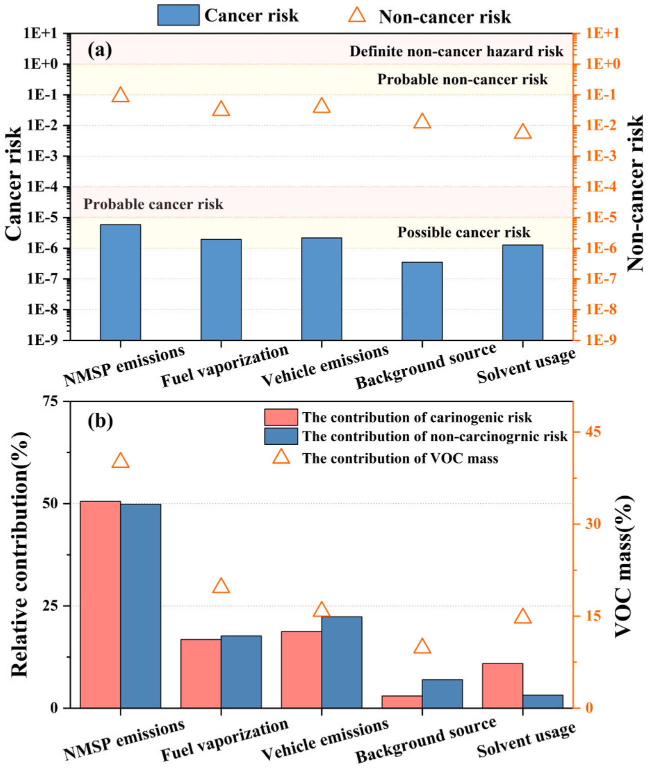

Fig. 5. Carcinogenic risk and non-carcinogenic risk of VOC sources (a) and relative contribution (b).

3.7. Policy implications

In recent years, the government has formulated a series of policies aimed at controlling air contamination. Background investigations revealed that the studied NMSP employs the two-stage treatment process involving wet scrubbing for dust removal and sodium-alkali desulfurization for flue gas treatment and sulfur recovery. However, for the non-ferrous metal smelting industry, the current treatments predominantly address particulate matter, sulfur dioxide, and heavy metals, while ignoring VOCs [48,8]. The health risk-oriented source apportionment results showed that NMSP emissions and vehicle emissions were the main sources of VOCs in residential areas surrounding NMSP. Therefore, VOC emissions should be mitigated directly at their origins. On the one hand, enterprises should enhance the control of waste gas generated in the NMSP production process, and on the other hand, local authorities should enforce traffic management strategies to restrict heavy trucks' operation during specific periods. The spatial distribution of carcinogenic risk and non-carcinogenic risk are shown in Fig. 4 and Fig. S14. The primary contributors to carcinogenic / non-carcinogenic risk are benzene (AHs), 1,2-dichloroethane (HHs), and 1,2-dichloropropane (HHs). Due to the discharge from NMSP smelting, the risk of carcinogenesis and non-carcinogenesis in surrounding areas had increased significantly, notably raising carcinogenic risks near residential areas to the southeast.

Given the high concentration and significant carcinogenic potential of benzene (AHs), 1,2-dichloroethane (HHs), and 1,2-dichloropropane

(HHs), their potential impact on the health of surrounding residents requires careful consideration. Therefore, prioritizing the removal efficiency of AHs and HHs would be an effective strategy for reducing the carcinogenic risk to surrounding residents. A variety of VOCs treatment technologies are currently available, including adsorption, absorption, membrane separation, condensation, thermal incineration, and advanced oxidation. However, the efficacy of these methods varies significantly depending on the physicochemical properties of the contaminants [62]. Integrated treatment systems thus represent the most effective approach for the synergistic control of diverse VOCs [13]. Considering the relatively low treatment costs and highly efficient removal efficiencies associated with AHs and HHs [15,40], the combined adsorption-condensation-catalytic oxidation process demonstrates promising efficiency, potentially achieving removal efficiencies of 96.1 % and 97.1 % for AHs and HHs in exhaust gas, respectively [3, 55,61]. Integrating this process with the existing tail gas treatment system in NMSP could substantially decrease VOCs emissions. According to the health risk-oriented source apportionment results, NMSP emissions were the biggest contributor to health risk. Implementing the aforementioned process for emission control, as illustrated in Fig. 6, the carcinogenic risk in RA would decrease by 48.7 %, while the non-carcinogenic risk would be reduced by 47.5 %. This optimization would lower the carcinogenic risk level for surrounding residents to a possible risk level (6.4×10^{-6}), and the non-carcinogenic risk is lowered to 0.098, below the threshold of concern. Notably, benzene, 1,2-dichloroethane, and 1,2-dichloropropane collectively contribute 83.2 % and 86.6 % to the reduction in carcinogenic and non-carcinogenic risk, respectively. Therefore, in order to effectively reduce the health risk in the studied residential area, it was necessary to give priority to controlling the AHs and HHs emissions from NMSP, specifically targeting benzene, 1,2-dichloroethane, and 1,2-dichloropropane. Although dichloromethane, n-hexane, and toluene in the study area were at high concentrations, their associated health risks were relatively low. Hence, policy development should emphasize the control of targeting contaminants that pose substantial health risks to workers and surrounding residents, rather than solely focusing on high-concentration compounds.

In the RA region, carcinogenic risks were not only associated with NMSP emissions but also contributed by vehicle emissions, fuel vaporization, and solvent usage. Table S11 lists the priority VOCs for control measures in RA. Specifically, benzene, 1,2-dichloroethane, and 1,2-dichloropropane need reductions of 78.3 %, 78.4 %, and 8.3 %, respectively, to reduce the health risks of each VOCs below the risk threshold. If achieving complete elimination of health risks from TVOCs would necessitate the implementation of even more stringent control strategies. NMSP emissions were the most influential emission source in RA. Therefore, to effectively mitigate carcinogenic risks in RA, it is essential to implement stringent control measures for organic waste gases emitted by NMSP, as well as exhaust gases from vehicles and VOCs discharged during solvent evaporation and solvent usage.

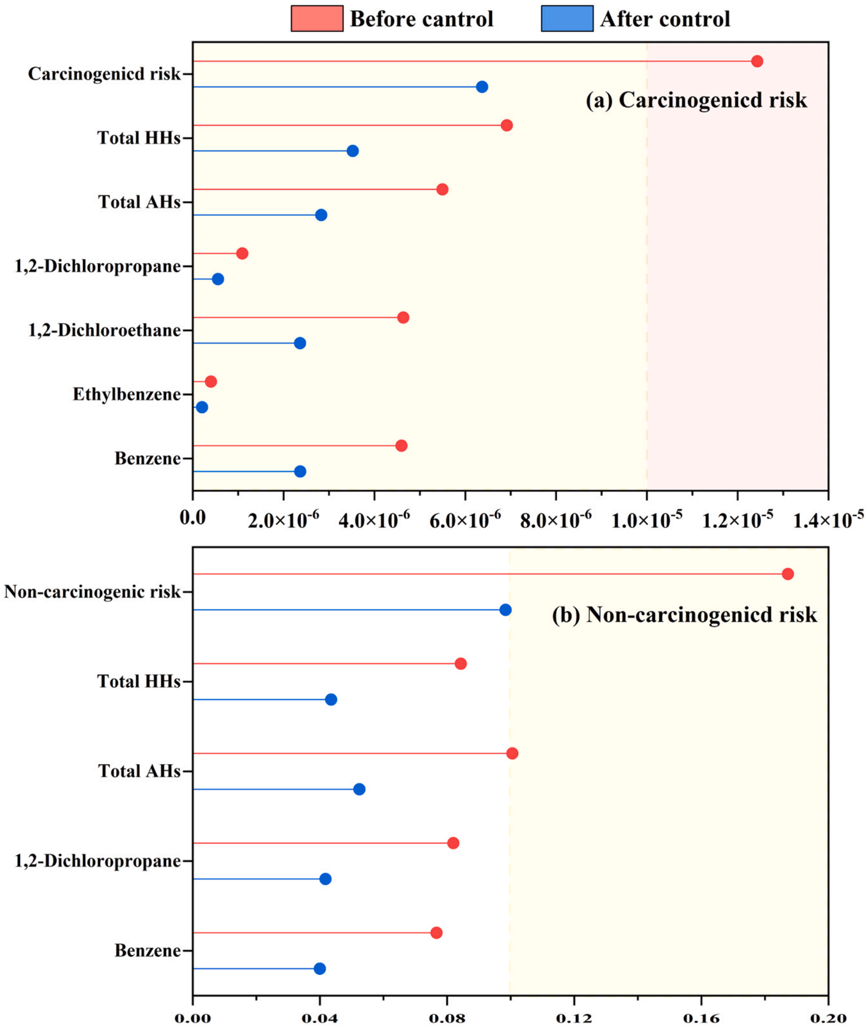

Fig. 6. Carcinogenic risk and non-carcinogenic risk of RA before and after NMSP emission source control.

4. Conclusion

Atmospheric samples were collected in NMSP and its surrounding areas to investigate VOCs contamination associated with smelting activities. The concentration of TVOCs in NMSP ranged from 88.13 to 154.38 μg/m^3, AHs and AIHs were identified as the characteristic contaminant group, with benzene, toluene, and n-hexane being the predominant individual compounds. A negative correlation was observed between the concentrations of contaminants and the distance from NMSP, combined with their spatial distribution, indicating that the emission of NMSP had a serious impact on nearby residential areas. PMF source apportionment identified six dominant sources: background source, solvent usage, NMSP emissions, fuel vaporization, vehicle emissions, and industrial emissions. Among these, NMSP emissions were the dominant source of VOCs, contributing 40.1 % to the TVOCs in RA. The results of health risk assessment showed that the NMSP and RA exhibited potential carcinogenic and non-carcinogenic risks, with benzene, 1,2-dichloroethane, and 1,2-dichloropropane identified as the major contributing compounds. Health risk-oriented source apportionment revealed that the carcinogenic risk value of all derived emission sources ranged from 3.50×10^{-7} to 5.87×10^{-6}, and all of them had a possible risk except the background source. The health risk distribution showed that NMSP had a serious impact on the surrounding population. If the emissions of AHs and HHs from NMSP are effectively controlled, the carcinogenic risk level in nearby residential areas can be reduced by one risk category, and the non-carcinogenic risk will become a negligible risk. Specifically, benzene, 1,2-dichloroethane, and 1,2-dichloropropane collectively contribute 83.2 % and 86.0 % to the reduction in carcinogenic and non-carcinogenic risk, respectively. A notable distinction exists between high-risk contaminants and high-concentration contaminants. Consequently, future regulatory policies targeting NMSP emissions should prioritize the reduction of benzene, 1,2-dichloroethane, and 1,2-dichloropropane due to their high health risk contributions.

Environmental implication

The characteristics of VOC contamination from NMSP emissions and their associated health impacts on surrounding residents were systematically investigated. High concentrations of VOCs were detected within NMSP, and the concentration was significantly negatively correlated with the distance from NMSP. Health risk-oriented source apportionment revealed that NMSP emissions accounted for 50.54 % of the carcinogenic risk and 49.84 % of the non-carcinogenic risk in RA. Targeted control of AHs and HHs can reduce carcinogenic risk by one category and eliminate non-carcinogenic concerns in RA. This study provides novel insights and policy guidance for the control of VOC emissions from industrial sources.

CRediT authorship contribution statement

Wanjun Wang: Formal analysis, Conceptualization. **Meicheng Wen:** Writing – review & editing, Supervision, Funding acquisition, Formal analysis, Conceptualization. **Jin Huang:** Methodology, Investigation, Data curation. **Jukun Xiong:** Formal analysis, Data curation. **Weiqiang Deng:** Writing – original draft, Resources, Methodology, Investigation, Formal analysis, Data curation. **Zhizhao Guo:** Writing – original draft, Investigation, Formal analysis, Data curation.

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.

Acknowledgements

This study was financially supported by Guangdong Provincial Natural Science Fund for Distinguished Young Scholars (2022B1515020051), National Natural Science Foundation of China (42377364, 42077332 and 41991312), and Science and Technology Program of Guangzhou (2025A04J5187).

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2025.139549.

Data availability

Data will be made available on request.

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