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A study on distribution characteristics of volatile organic compounds in Paju industrial complex area, using proton transfer reaction-time of flight mass spectrometry

So-Young Kim[1*]

Abstract

Paju City is located in the northwest of Gyeonggi-do, and its chemical emissions in 2020 were 1,287,917 kg, the 4th highest in Gyeonggi-do. In particular, the Munsan High-Tech Industrial Complex in Paju has LCD manufacturing plants and partner companies distributed in groups, and the volatile organic compounds used by these companies are causing many problems, such as causing bad odors, to the local community. In this sense, real-time analyzing equipment (proton-transfer-reaction time-of-flight mass spectrometry) was mounted on a vehicle for this study to look into the air quality around VOCs-using companies inside the High-tech Industrial Complex in Munsan, Paju from October 19 to October 21, 2020.

According to measurement results, toluene was detected the most at 25.7 ppb, followed by carbon tetrachloride (17.6 ppb), ethylbenzene (17.2 ppb), and xylene (8.5 ppb), which demonstrates that there is a need to control these substances to resolve the issue with VOCs in the region. In particular, benzene designated as the air quality standard was detected at 1.0 ppb in some sites, which is below the threshold (1.5 ppb). However, it was detected at 2.1 to 4.4 ppb, exceeding the threshold in most sites. Thus, continuous monitoring is expected to keep VOCs under control in Paju Industrial Complex down the road, using real-time measuring equipment.

Keywords Volatile organic compounds (VOCs), PTR-ToF-MS (proton transfer reaction-time of flight-mass spectrometer, Paju, LCD (liquid crystal display)

1 Introduction

Paju City is located in the northwest of Gyeonggi-do. There are 14 industrial complexes, including a liquid crystal display(LCD) complex, a publishing complex, and Munsan High-Tech, located in the area. According to the Pollutant Release Transfer Register (PRTR), the amount of chemicals discharged from Paju in 2020 was approximately 1,287,917 kg, the second highest level in Gyeonggi-do after Hwaseong, Pyeongtaek, and Ansan.

In particular, Munsan High-Tech Industrial Complex is a complex built between 2004 and 2010 in the Dangdong District and Seonyu District of Munsan, Paju, Gyeonggi-do, and is home to LCD manufacturing plants scattered sporadically in the metropolitan area and partner companies that manufacture related equipment, parts, and materials. Because they are distributed intensively, the emissions of volatile organic compounds such as toluene used in these businesses are causing many problems in the local community.

In particular, the government has banned the use of six substances in this area, including toluene, benzene,

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m-xylene, o-xylene, dichloromethane, and trichloroethylene, but there are many reports from residents and complaints about bad odors that some businesses are using these substances. On December 19, 2019, a chemical accident occurred at a company where toluene leaked and splashed into the eyes of workers, making it necessary to conduct a general investigation into VOCs, including banned substances such as toluene, in this area.

Therefore, in order to solve the VOC problem in this region, it seems necessary to identify which types of VOCs mainly have high concentrations and intensively manage the companies that mainly emit them.

VOCs are emitted from various industrial processes such as crude oil refining, chemical processing, use of organic solvents, automobile exhaust, painting, and synthetic resin manufacturing (Kim et al., 2021; Park et al., 2005), and exposure to humans can cause leukemia and central nervous system disease. It is reported to cause nervous system disorders, asthma, emphysema, breathing disorders, lung cancer, etc. (Son et al., 2005; Kim et al., 2021). In addition, it has been shown to affect a number of air pollutants, such as acting as a precursor for secondary organic aerosol (SOA), which reacts with ultraviolet rays in the atmosphere and produces photochemical smog, and causes global warming and bad odors. The importance of management is being highlighted (Liu et al., 2008).

However, although some VOCs surveys in the Paju area were conducted on publishing complexes, etc. (Lee et al., 2022), there is no survey on other areas, such as Munsan High-Tech Industrial Complex, where LCD-related companies are concentrated. In addition, since the national hazardous air pollutant measurement network and photochemical pollutant measurement network that can periodically monitor VOCs have not been established in Paju, an investigation into the overall VOC concentration distribution in the region is necessary to solve the VOC problem in the Paju area.

Accordingly, in this paper, a proton transfer reaction-time flight-mass spectrometer (PTR-ToF-MS), which can measure individual VOCs in seconds, was mounted on a vehicle, which has recently raised numerous odor complaints. The air quality in the area surrounding VOCs-using companies in the Munsan high-tech industrial complex in Paju was investigated. Based on these measurement results, an on-site survey was conducted on companies using VOCs near the high concentration measurement point, and numerous violations of environmental laws were discovered. Accordingly, this paper is expected to be used as basic data for preparing measures for the use of VOCs not only in the Paju industrial complex but also in future national industrial complexes.

2 Measurement and methodology

2.1 Selection of measuring sites

Among businesses licensed to handle VOCs in accordance with the Chemical Substances Act Control, five businesses handling toluene were selected as stationary sites to measure VOCs in the surrounding area of those VOC-handling businesses inside the High-tech Industrial Complex. These businesses mainly use toluene among VOCs. On the other hand, mobile measurements were conducted along the road circulating around the outskirts of the Seonyu District while passing through the stationary sites. As seen in Fig. 1, PTR-ToF MS was installed inside the vehicle and data was measured as the

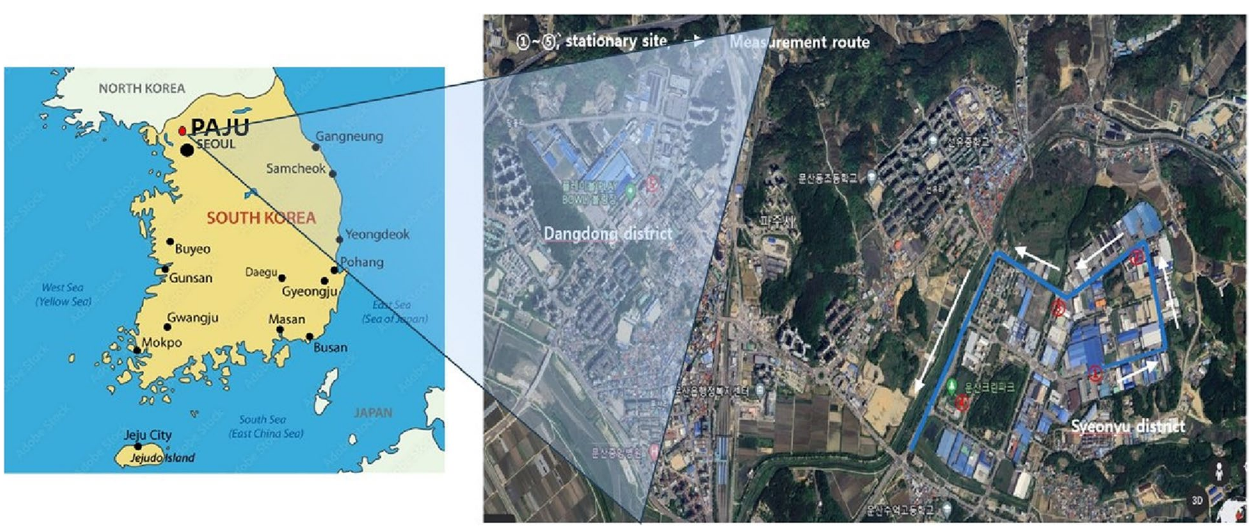

Fig. 1 View of measurement route and stationary site

vehicle moved around. It also stopped for about 10 min at the boundary of the company entrance for additional measurement. First, the vehicle started by measuring VOCs at the boundary of the first company (①) located on the eastern part of Seonyu General Industrial Complex, and moved northward, stopping for 10 min on the road in front of the second company (②) for measurement. Then, it moved to the southwest and stayed there for 10 min on the road in front of the third company (③) to measure VOCs and went to the south to measure VOCs by stopping for 10 min in front of Munsan Green Park (④) Road. Meanwhile, in the Dongdong District, only stationary measurements were taken on the road in front of the fourth company (⑤), with mobile measurements. The measurements were conducted on October 19, 2020, at Site ⑤ (Dangdong District, Stationary) and on Oct 21 at Sites ① to ④ (Seonyu District, Stationary + Mobile) and their measuring sites are as shown in Fig. 1.

2.2 Selection of measuring equipment and target substances

The PTR-ToF-MS(KORE Tec., USA) was used to measure VOCs in the High-tech Industrial Complex in Munsan, Paju. It is a device that can analyze VOCs in the atmosphere on a real-time basis without any pre-treatment. It analyzes VOCs through proton transfer reaction and is widely used in various research such as analysis of exhaust gases, aerial observation, and origin analysis of SOA as it can analyze individual concentrations of substances in the atmospheres up to seconds without any process of sampling and pre-treatment, compared to the existing GC-MS method(Blake et al., 2009; De Gouw & Warneke, 2006; Karl et al., 2007; Kim et al., 2013; Kim et al., 2021; Moser et al., 2005). In this study, 15 VOCs including toluene were selected and information regarding each substance was listed in the following Table 1. As a result of creating a calibration curve between 10 and 600 ppb for 15 types of VOC standard substances (supelco to-14) to be measured, R^2 was greater than 0.99. In this paper, 7 types are shown in pictures (Fig. 2).

Table 1 VOCs target compounds of PTR-ToF-MS

No.	Substance	ABB.	Mass	CAS NO	Formula
1	Benzene	BZ	78	71-43-2	C6H6
2	Toluene	TOL	92	108-88-3	C7H8
3	Bromomethane	BM	94	74-83-9	CH3Br
4	Styrene	STR	104	100-42-5	C8H8
5	Ethylbenzene	EBZ	106	100-41-4	C8H10
6	p-Xylene	XYL	106	106-42-3	C8H10
7	Chlorobenzene	CBZ	112	108-90-7	C6H5Cl
8	1,2,4-Trimethylbenzene	1,2,4-TMB	120	95-63-6	C9H12
9	Dichlorodifluoromethane	CCl2F2	120	75-71-8	CCl2F2
10	Dichlorobenzene	DBZ	147	95-50-1	C6H4Cl2
11	Carbontetrachloride	CBR4	153	56-23-5	CCl4
12	Tetrachloroethene	PCE	165	127-18-4	C2Cl4
13	1,2,4-Trichlorobenzene	1,2,4-TCB	181	120-82-1	C6H3Cl3
14	1,2-Dibromoethane	1,2-DM	187	106-93-4	C2H4Br2
15	hexachloro-1,3-butadiene	HEX	260	87-68-3	C4Cl6

3 Results and discussion

3.1 Weather conditions

During the measurement period, weather conditions such as wind directions, temperatures, and wind speeds were analyzed based on data from the Automatic Weather System (AWS) located in Paju. Figure 3 shows wind field diagrams using the AWS data measured in Guemsan, Paju on October 19 and October 21 (KMA, 2020). Winds mainly blew from the southwest and northeast on Oct 19 and from the southeast and northeast on October 21. For wind speeds, the daily average was similarly low with 0.9 m/s on the 19th and 0.8 m/s on the 21st and the frequency of winds blowing at 0.5 m/s or less stood at 45% on the 19th and 33% on the 21st, causing slower spread and dilution of pollutants on both days. Furthermore, although winds blew in different directions on the 2 days, their wind speeds and temperatures were quite similar. It is safe to say that there was no big difference in concentration between the 2 days due to the weather conditions.

3.2 VOCs composition ratio and concentration distribution characteristics between measurement points

The composition of VOCs measured at stationary sites in Dangdong and Seonyu districts of the High-tech Industrial Complex in Munsan, Paju indicates that toluene accounted for the largest share (19 to 45%) in all sites (Fig. 4). Lee et al. (2022) also measured 14 VOCs including toluene(TOL) at Paju Publishing Complex and TOL took up about 25% with slight differences depending on sites, showing similar results to this study. However, the ratio of TOL was a mere 3.2% in the results measured by Kim et al. (2021) at Daesan Industrial Complex in Chungnam, which is somewhat different from the result of this study. In general, VOCs tend to have various distribution patterns depending on types of emission sources, industrial structure, and process characteristics (Kim et al., 2021). Daesan hosts a cluster of petroleum chemical complexes and power plants, presenting a different business ratio compared to Paju.

In Seonyu District, after TOL, ethylbenzene(EBZ) was the second highest substance (17 to 27%), followed by carbon tetrachloride (CBR4) which showed a fairly high level of concentration (15 to 16%) at all sites excluding point 4. On top of that, benzene (BZ) that

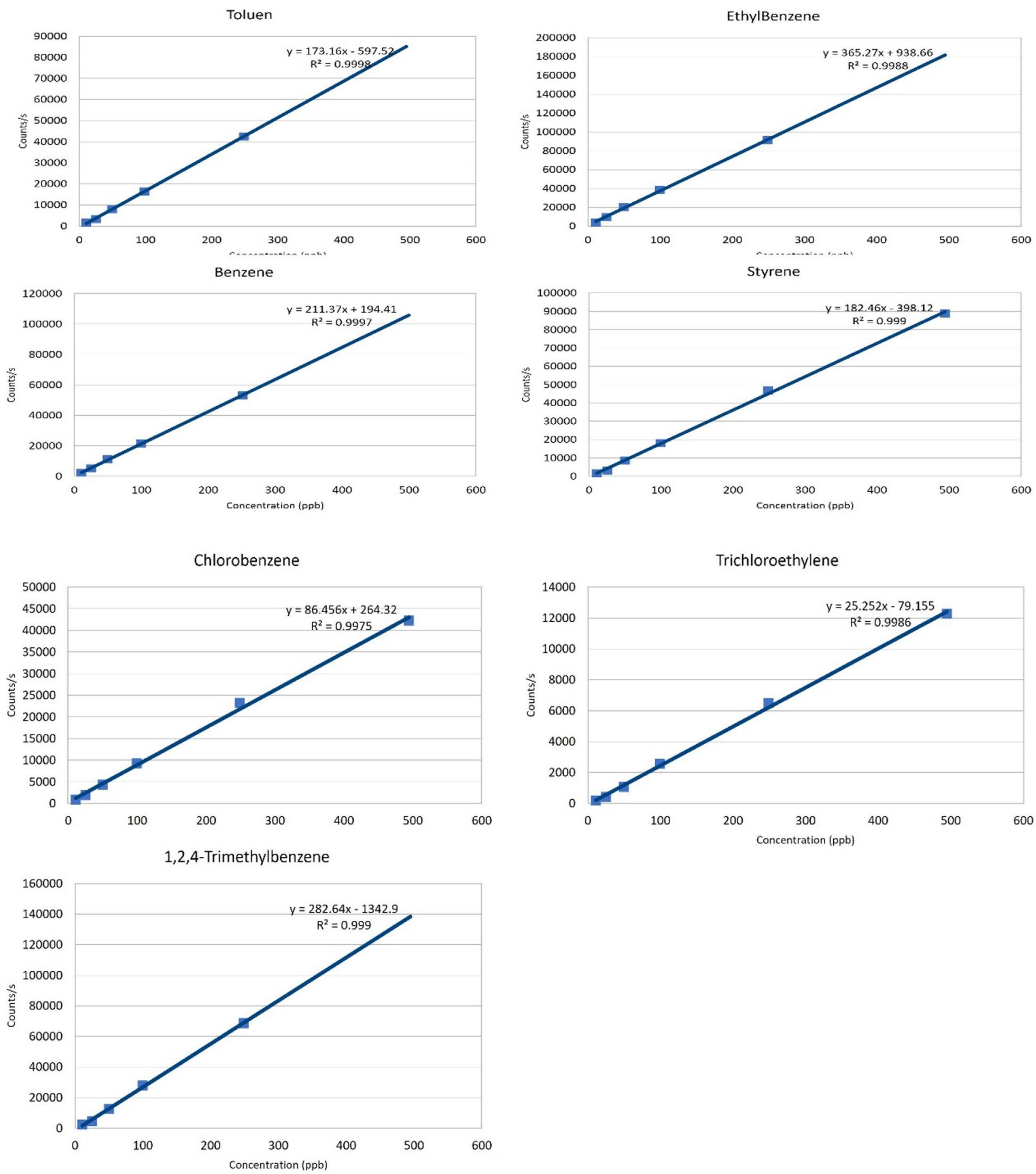

Fig. 2 Calibration curve of the substance to be measured

is the only VOC designated as the air quality standard in South Korea took up a maximum of 4% in all sites. Besides, others such as chlorobenzene (CBZ), 1,2,4-trimethylbenzene (1,2,4-TMB) and styrene (STR) were also detected to some extent. Rather than jumping to a conclusion simply with one or two measurements for this study, it is necessary to engage in discussions over the selection of target substances to be managed by conducting more follow-up studies down the road to solve the issue with VOCs including TOL, EBZ, CBR4,

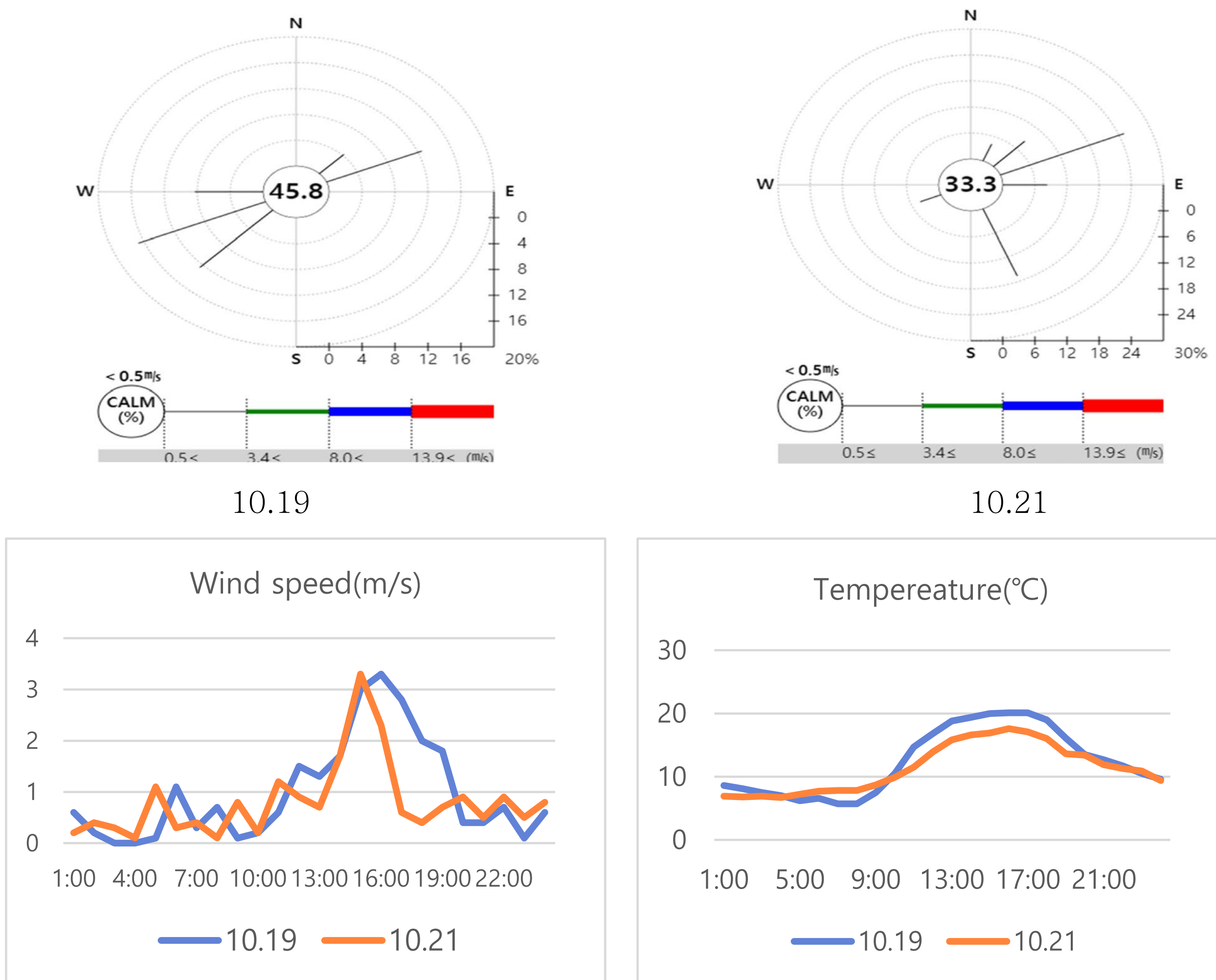

Fig. 3 Weather conditions during the measurement period

xylene (XYL), BZ which accounted for a high ratio in Seonyu District.

In point 5, VOCs were measured on October 19. The result indicated that toluene was the highest component with 19% which is lower than the levels measured at points 1 to 4. It was followed by XYL (17%) and EBZ(12%), validating that TOL, EBZ, and XYL were the main emissions.

In order to determine the concentration distribution of VOCs in the Munsan High-Tech Industrial Complex in Paju, the results of measurements taken while moving from point 1 to point 4 by vehicle are shown in Fig. 5 for each time period. Among the substances to be measured, TOL, EBZ, XYL, and CBR4 were measured at levels between 10 and 50 ppb, although there were differences depending on the substance, which was higher than other substances.

In particular, the average concentration of TOL was 25.7 ppb, the highest among the measured substances, and varied by more than 20 times from a minimum of 3.0 ppb to a maximum of 44.2 ppb depending on the location. Next, CBR4 appeared high at 17.6 ppb, followed by EBZ at 17.2 ppb and XYL at 8.5 ppb.

BZ showed a concentration of 1 to 3 ppb, although there were differences depending on the location, and most of them exceeded the air environment standard of 1.5 ppb. In particular, at point 5, the level was 0.97 ppb, which is lower than the air quality standard of 1.5 ppb.

However, points 1 to 4 were higher than 1.5 ppb and the maximum was 5.89 ppb, showing that BZ, etc. may exceed the standard in industrial complexes depending on the industry and process using VOCs.

In fact, according to the results measured by Kim et al. (2021) at the Daesan Industrial Complex, BZ was found to be about twice higher than the standard.

Figure 6 compares VOC concentrations between measurement points. Although there are differences between points, the median VOCs were at a

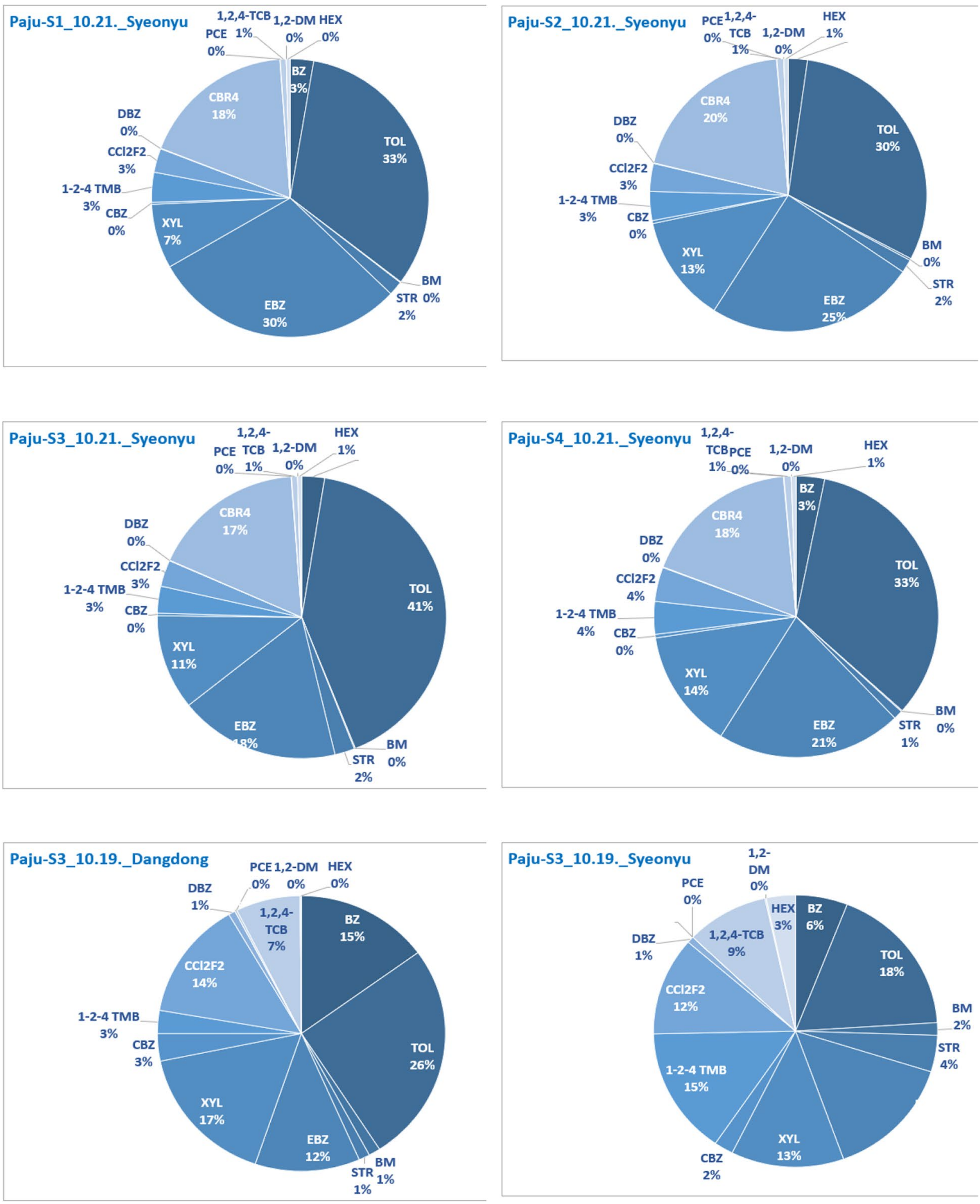

Fig. 4 Distribution ratio of each VOCs during the measurement period

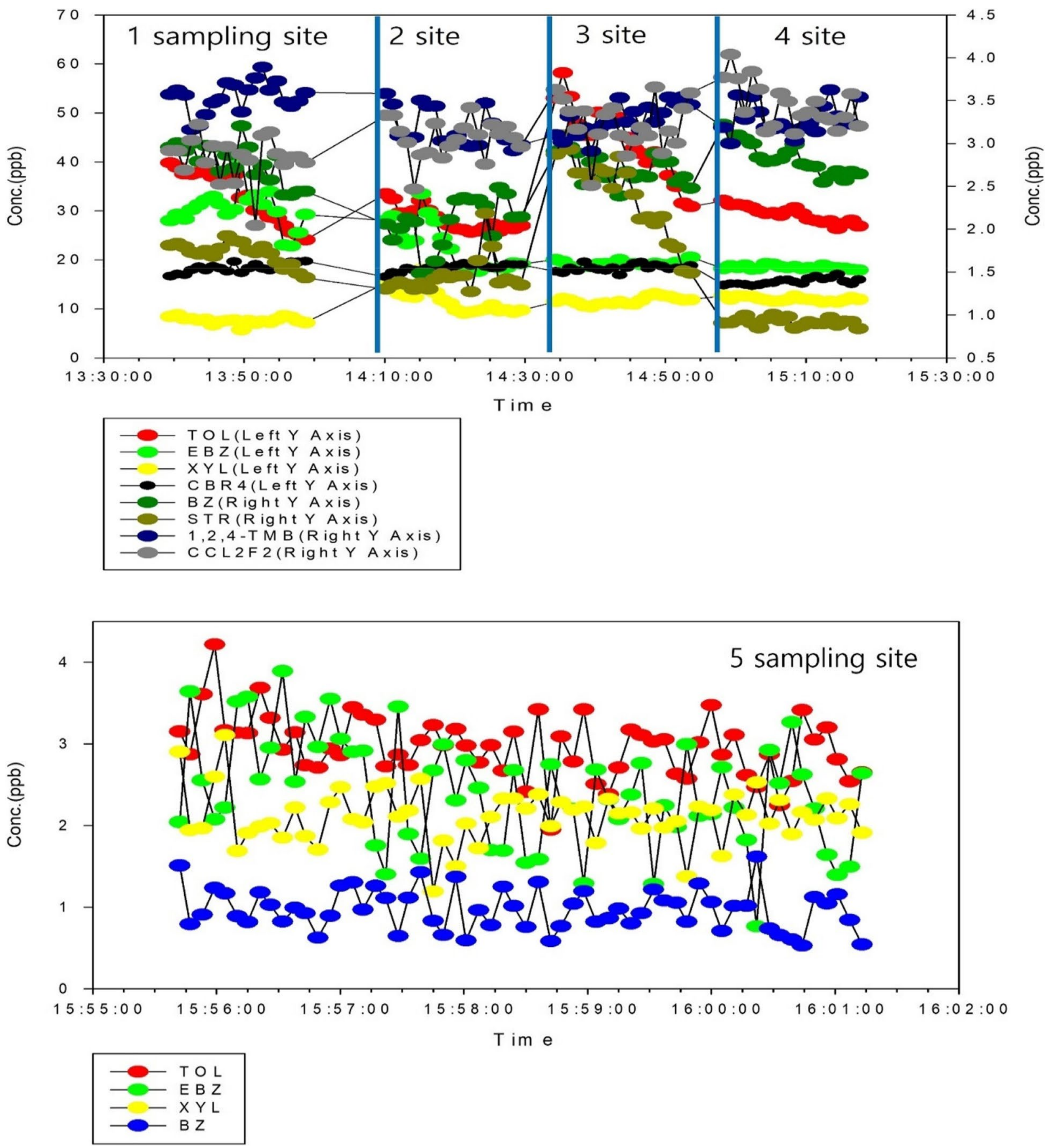

Fig. 5 Change in VOCs concentration at each measurement point over time

similar level overall, but the concentration distribution at points 1 to 4 was wider than that at point 5. The reason why outliers appeared at most points is because TOL was measured at a higher level than other substances. In particular, point 3 had the highest TOL compared to other points, resulting in the highest outlier.

In addition, there are more companies using VOCs around points 1 to 4 than around point 5, so it appears that the substances used by these companies have an impact on the surrounding air quality. Based on these results, as a result of conducting an on-site survey on companies using VOCs around points 1 to 5, violations of environmental laws, such as illegal use of chemicals

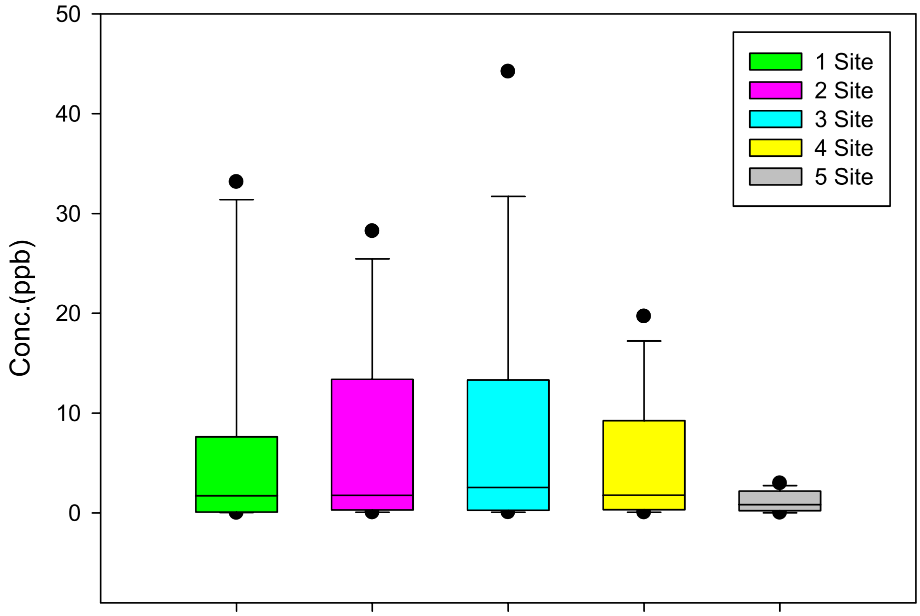

Fig. 6 Comparison of VOCs concentration distribution between measurement points

and non-compliance with air pollution prevention facilities, were confirmed at companies around points 1 to 4, and action was taken.

In general, when residential complexes are built around industrial complexes that use a variety of hazardous chemicals, complaints from residents due to the substances used by these companies continue to be raised. In order to solve this problem, local governments and governments periodically conduct crackdowns on companies handling hazardous chemicals in industrial complexes, but there are limits to fully investigating all target companies with limited manpower.

To solve this problem, equipment that can measure VOCs in real time is mounted on a vehicle, measured while moving, and then intensive on-site inspection is performed focusing on high-concentration areas. In fact, after measuring with a vehicle equipped with PTR-ToF-MS from 2020 to 2021 at the Sihwa and Banwol National Industrial Complexes, and focusing on the areas where high concentrations were measured, environmental law-related matters were intensively inspected between related organizations, and the target was generally document-oriented. The violation rate was found to be 20–50% higher than when the workplace was selected and inspected (Fig. 7).

However, such real-time equipment has the disadvantage of being expensive and requiring a lot of expertise to interpret the data. Therefore, in order to solve the VOC problem in the Paju high-tech industrial complex, install and operate a hazardous pollutant measurement network that can periodically measure VOCs or conduct monitoring using real-time measurement equipment.

3.3 Comparison of VOC concentration distribution with other regions and correlation analysis between substances

It was found that VOCs have various distribution patterns depending on types of emission sources, industrial structure, and process characteristics. Environmentally advanced nations such as the USA, the United Kingdom, and Japan have established and operated their environmental standards regarding VOCs since 1990. South Korea has also established its standards for the annual average of BZ (Byeon et al., 2009; Kim et al., 2021). Table 2 compares the results of research conducted in other areas with the concentrations of BZ, TOL, and XYL of this study. The measurements conducted at Gunsan Landfill in 2006 showed the following results: BZ (2.5 ppb), TOL (34.4 ppb), and m,p-XYL (10.2 ppb), which are quite similar to levels in this study. In terms of TOL, moreover, it was also comparable to the levels measured on the roadside (Daegu) and Pyeongdong Industrial Complex (Gwangju). However, it was even 30 times higher, compared to areas densely packed with petroleum chemical complexes in Yeosu and Daesan. For BZ, the concentration measured in this study was twice as high in some sites and it was also measured to a lower level compared to the measurement results at Daesan Industrial Complex. Thus, it can be described that VOCs have different distribution

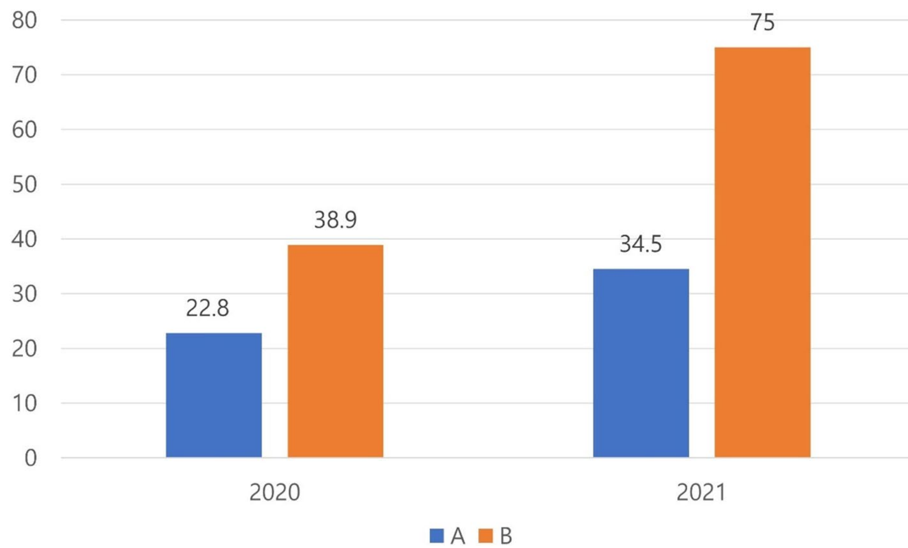

Fig. 7 Comparison of environmental law violation rates according to measurement-based target business site selection and document-based target business site selection

Table 2 Measured VOCs concentration in this study and other cities. (Unit:ppb)

Author	Year	Location	Site	Pollutants			
				Benzene	Toluene	m,p-Xylene	Sum
Jeon et al.	2003	Yeosu	Industrial	1.16	1.85	0.33	3.34
Jeon et al.	2005	Yeosu	Industrial	1.32	4.55	0.27	6.14
Baeket al.	2005	Pohang Gumi	Industrial Industrial	0.83 0.62	3.50 7.81	4.5 1.29	8.83 9.72
Jung et al.	2006	Nanjido Gyungsan	Landfill Landfill	31.8 2.5	259 34.4	10.4 10.2	301.2 47.1
Im et al.	2006	Ansan	Industrial	16.4	423	43.6	483
Park et al.	2006	Daegu Gyeonggsan	Roadside Sub-urban	1.18 0.89	20.4 4.31	1.87 0.87	23.5 6.07
Ryoo et al.	2010	Jeonju Gumam	Roadside Industrial	3.1 3.0	6.3 11.8	3.1 3.8	12.5 18.6
Baek et al.	2020	Daesan	Industrial Industrial	1.13 0.07	1.20 2.34	0.94 0.51	3.27 2.92
Kim et al.	2019	Daesan	Industrial	8.3	4.5	1.8	14.6
Kim et al.	2020	Haman Pyeongdong	Industrial Industrial	1.67 0.42	15.5 23.3	4.2 6.37	21.4 30.1
Kim et al.	2020	Daesan	Industrial	3.10	1.50	1.88	6.48
This study	2020	Paju	Industrial	2.6	25.7	8.5	36.7

characteristics according to the types of emission sources and industrial structure in the corresponding region.

Figures 8 and 9 show a scatter plot to identify the correlation between the materials to be measured. First, as a result of identifying the correlation between all target substances as shown in Fig. 7, although there are differences depending on the substance, TOL, EBZ, XYL, BZ, styrene, etc. showed a relatively positive relationship, but 1,2,4-TMB, CCl_2F_2, CBR_4, etc. showed a relatively low correlation between substances.

In Fig. 10, the correlation was analyzed again only for BZ, TOL, EBZ, and XYL, which had relatively high correlations between substances. As a result, it was found that as BZ increased, TOL also increased, and BZ and EBZ were also found to be similar.

At measurement points 1 to 4, many similar industries, such as TV flat panel display manufacturers, are

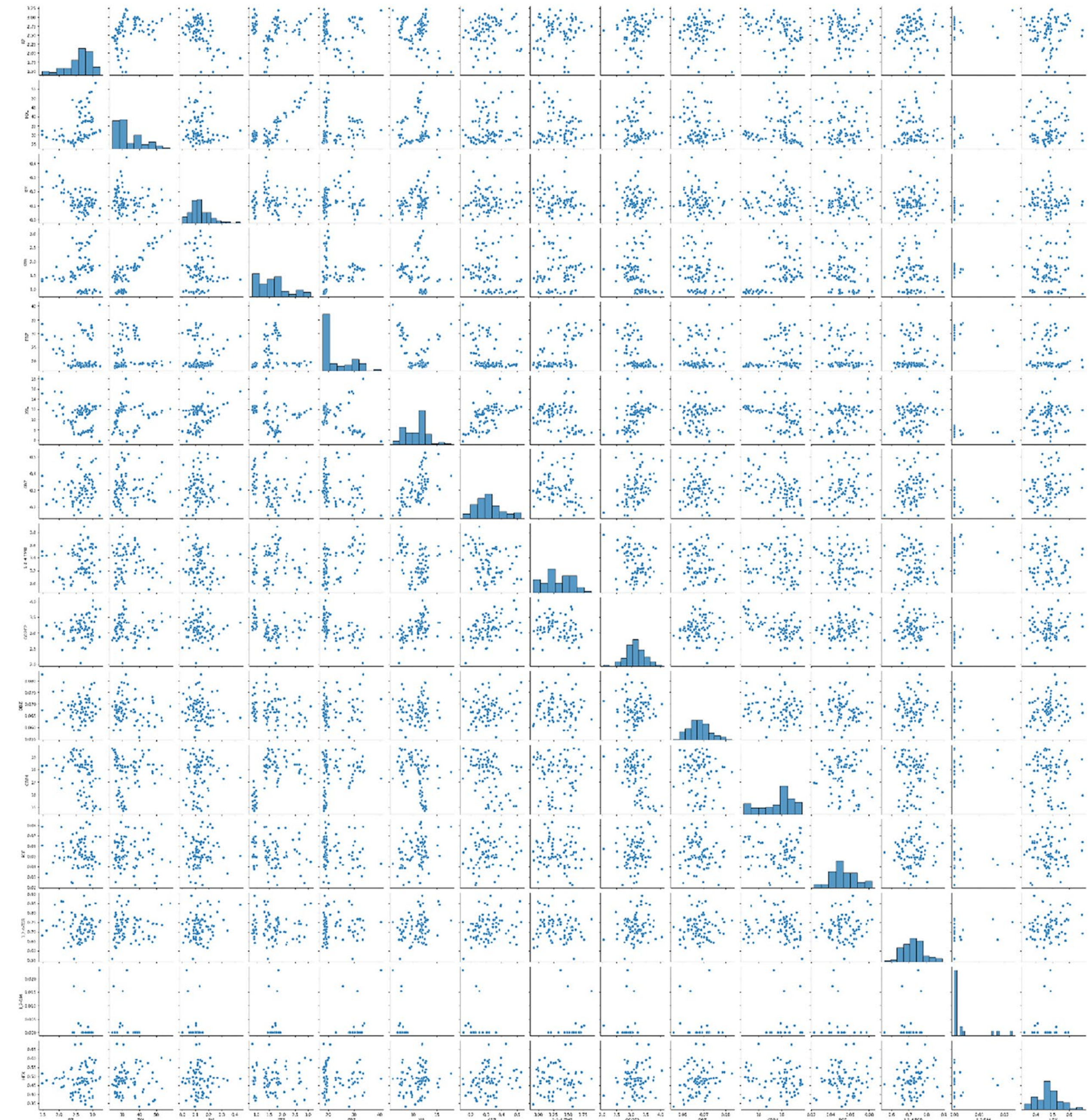

Fig. 8 Scatter plot between substances to be measured

distributed, so the emission characteristics appear to be similar to BZ and TOL used by these companies are emitted.

3.4 Spatial distribution of VOCs

Figure 9 shows the distribution of BTEX concentrations out of the results of local pollution levels measured by the mobile laboratory vehicle with real-time measuring equipment that ran near businesses handling VOCs. The concentrations were higher in proximity to business handling VOCs in the eastern and northern parts of sampling sites(1~4 points). When it moved westward, it became lower compared to the levels in the eastern and northern parts. In general, sampling sites(1~4 points) had high concentrations in their eastern and northern areas, indicating that companies

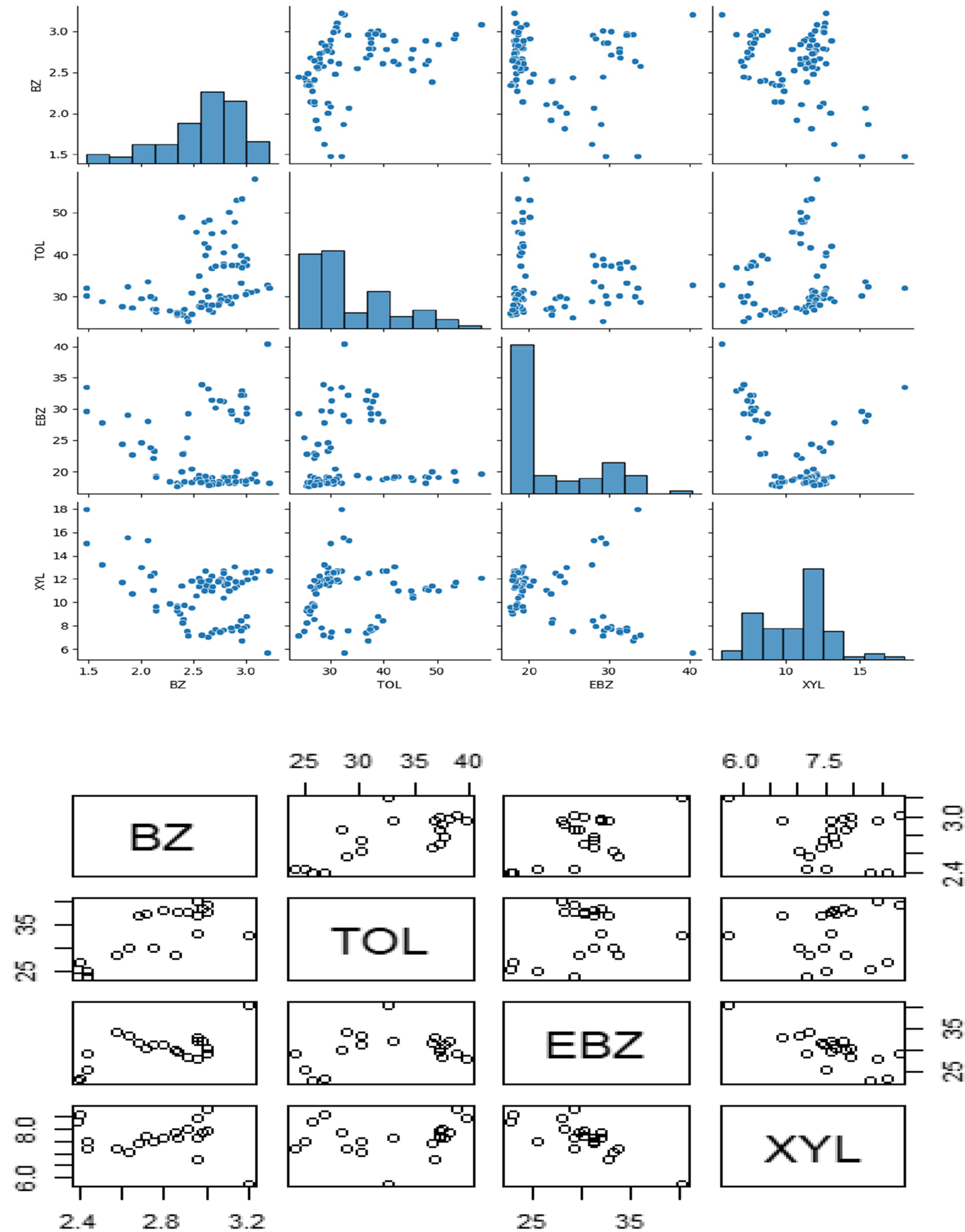

Fig. 9 Scatter plot of benzene, toluene, ethylbenzene, and xylene

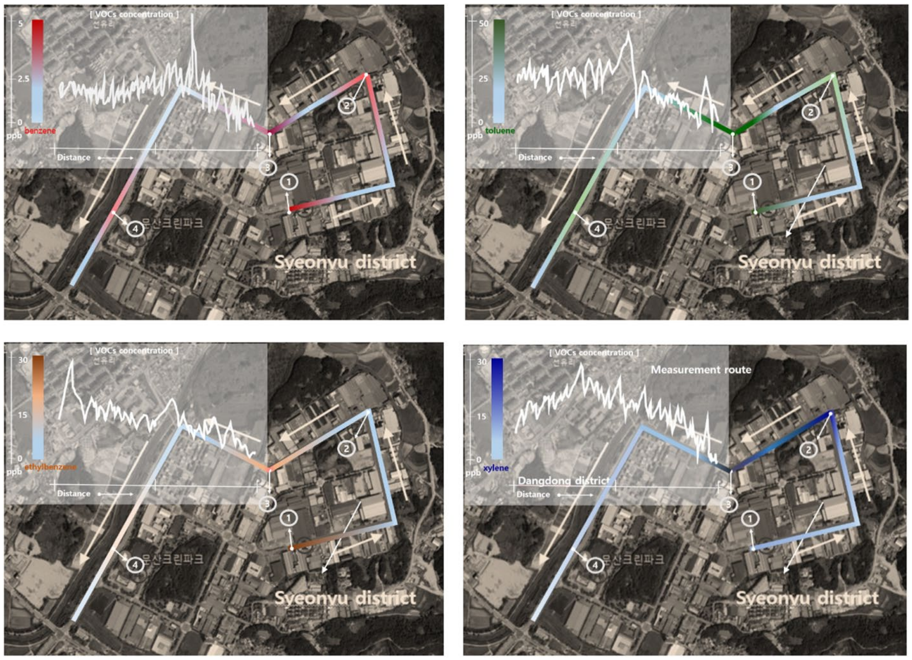

Fig. 10 Mapping results of VOCs measured using mobile laboratory

in this region could influence the local concentration of VOCs.

Based on these results, a general survey was conducted on companies using VOCs in the Munsan High-Tech Industrial Complex in Paju in 2020, and as a result, violations of environmental laws were discovered in relation to the VOCs handling process and air quality prevention facilities.

In general, when residential complexes are built around industrial complexes that use a variety of hazardous chemicals, complaints from residents due to the substances used by these companies continue to be raised. In order to solve this problem, local governments and governments periodically conduct crackdowns on companies handling hazardous chemicals in industrial complexes, but there are limits to fully investigating all target companies with limited manpower.

To solve this problem, equipment that can measure VOCs in real time is mounted on a vehicle and then moved while measuring within the industrial complex to determine which substances among the VOCs are distributed in the area and from which processes these substances are mainly emitted. The goal is to actively utilize the method of intensive on-site inspection of suspicious companies, focusing on high-concentration areas. In this way, it is expected that the problem caused by VOCs in the region can be solved by strengthening the guidance and crackdown on problem companies and reducing VOC emissions.

4 Conclusion

In order to determine the distribution of VOCs concentration in the Paju Munsan High-Tech Industrial Complex, a vehicle equipped with PTR-ToF-MS was used to measure air quality around VOCs handling businesses in the Paju Munsan High-Tech Industrial Complex, and the following conclusions were drawn.

Among the VOCs to be measured, toluene accounted for the highest proportion of 19~45%, followed by ethylbenzene (17~27%), xylene (17%), and carbon tetrachloride (15~16%). Among VOCs, benzene, which is the only one set as a domestic air environment standard, was found to have a composition ratio of up to 4% at all points.

The average concentration of toluene was the highest at 25.7 ppb and varied by more than 20 times depending on the location, from a minimum of 3.0 ppb to a maximum of 44.2 ppb. Benzene varies depending on the location, but if the concentration was 1 to 3 ppb, it mostly exceeded the air environment standard of 1.5 ppb.

Compared to studies in other regions, the results measured at the Gunsan landfill were similar to the toluene, benzene, and m,p-xylene levels in this study. In particular, toluene was similar to the results measured at the roadside (Daegu) and the Pyeongdong Industrial Complex (Gwangju) but was 30 times higher than the results measured at the Yeosu and Daesan petrochemical complexes.

Correlations for each VOC had slight differences depending on sites, however, at Sites 1, 2, and 3, substances like toluene, benzene, styrene, ethylbenzene, and xylene had a correlation coefficient of no less than 0.5. That said, at Site 4, there was no substance with significant correlation except for toluene, xylene, and benzene, showing different emission characteristics between Sites 1 to 3 and Site 4.

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Availability of data and materials
The datasets generated during and analyzed during the current study are available from the corresponding author on reasonable request

Declarations

Competing interests
The authors declare that they have no competing interests.

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