



Fence Line Monitoring of Petroleum Refining Plants in Korea: UV-DOAS Analysis of SO₂, NO₂, and Benzene Emissions for Comprehensive Management

Cheonwoong Kang^{1,3} · Dongwoo Kim^{1,3} · Jeonghun Kim² · Sunghwan Cho² · Jungung Yu² · Daeil Kang² · Poongmo Park³ · Kijoon Jeon^{1,3}

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Abstract

Increasing public interest in air pollutants driven by climate change and high concentrations of particulate matter (PM) has highlighted the importance of managing pollutants, such as SO₂, NO₂, and benzene. These compounds are known precursors of secondary PM that affect the climate and human health, with benzene being particularly carcinogenic. Benzene is a volatile organic compound (VOC) that can persist in the atmosphere for approximately 9.4 days, potentially traveling long distances and contributing to the generation of secondary organic aerosols (SOAs) through photochemical reactions, leading to elevated fine dust concentrations. To comprehensively manage emissions, leaks, and point sources, a fence line monitoring study was conducted at petroleum refining industrial complexes in Daesan-eup, Chungcheongnam-do, and Yeosu-si, Jeollanam-do in Korea. Ultraviolet Differential Optical Absorption Spectroscopy (UV-DOAS) technology was used to confirm measurement reliability and monitor emissions. High SO₂ concentrations emitted in a specific direction were identified at the Yeosu-si, and further investigation identified the flare stack of a suspected plant as the emission source. Fence line monitoring was effective in identifying and addressing unknown high-concentration emission sources and devising measures to manage fugitive emissions and leaks.

Keywords Fence line monitoring · UV-DOAS · SO₂ · NO₂ · Benzene

Introduction

The Korean Ministry of Environment reports that 70% of Korea's PM_{2.5} is generated secondarily through photochemical reactions in the atmosphere. They are establishing multifaceted policies to manage the emissions of various

gases, such as nitrogen oxides (NO_x), sulfur oxides (SO_x), NH₃, and volatile organic compounds (VOCs), which are precursors that generate secondary particulate matter (PM). PM_{2.5} has a small aerodynamic diameter of under 2.5 μm; therefore, it can penetrate deep into the human lungs and cause adverse health effects, such as cardiovascular disease, respiratory disease, and cancer [1–4]. PM_{2.5} is directly emitted in various environments and conditions, such as biomass combustion, coal combustion, vehicle emissions, soil dust, and industrial emissions [5–7]. Pollutants emitted from pollution sources are produced secondarily in the atmosphere through photochemical reactions. They can be divided into primary and secondary aerosols [8]. Secondary aerosols include secondary organic aerosols (SOAs) and secondary inorganic aerosols (SIAs). SOAs are produced by photochemical reactions of VOCs [9, 10]. SIAs mainly consist of sulfate, nitrate, and ammonium, which are produced by the photochemical reactions of gaseous pollutants, such as NO_x and sulfur dioxide (SO₂) in the atmosphere [11, 12].

Cheonwoong Kang and Dongwoo Kim have contributed equally to this work.

✉ Kijoon Jeon
kjjeon@inha.ac.kr

¹ Department of Environmental Engineering, Inha University, 100 Inha-ro, Michuhol-gu, Incheon 22212, Republic of Korea

² Air Pollution Engineering Division, National Institute of Environmental Research, 42, Hwangeyeong-ro, Seo-gu, Incheon 22689, Republic of Korea

³ Particle Pollution Research and Management Center, 36, Gaetbeol-ro, Yeonsu-gu, Incheon 21999, Republic of Korea

Petroleum refining plants have various emission facilities, including processing, combustion, and fugitive emissions. The pollutants emitted from these facilities negatively impact human health and must be managed [13]. In the Korea, various pollutant emission facilities are measured by attaching automatic environmental measurement equipment (TMS, telemonitoring system) or are managed by periodic sampling and measurement. However, it is difficult to measure and manage pollutants in fugitive emission facilities; therefore, they are managed by applying a leak detection and repair (LDAR) program. The LDAR program regularly manages facilities that may leak or emit fugitive materials. The LDAR program is effective in reducing fugitive pollutants [14, 15], but requires a lot of manpower and is difficult to achieve effectiveness for irregular emissions. More advanced management is required than the LDAR program, one of which is fence line monitoring regulation. Fence line monitoring regulations have been introduced in the United States to accurately measure and manage benzene. Fence line monitoring is a regulation that comprehensively manages pollutants emitted from a plant by measuring at the plant boundary, rather than measuring and managing various emission sources individually [16]. Benzene is a volatile organic compound that is toxic by itself [17], with a maximum lifetime of 9.4 days, which is much longer than that of *m*-xylene. Benzene can travel long distances and has a sufficiently long lifetime for photochemical reactions.

In this study, SO₂, NO₂, and benzene (which are PM_{2.5} precursors and pollutants emitted from various emission sources) were monitored using an open path ultraviolet differential optical absorption spectroscopy (UV-DOAS) analyzer near a petroleum refining plant, and the results were analyzed using wind direction and wind speed data. We intend to explore the possibility of utilization in terms of air environment policies, such as tracking and managing the main emission sources without dividing the points and fugitive emission sources.

Materials and Methods

The measurements were conducted in two regions. Before installation, calibration was performed in the laboratory to confirm the reliability of the UV-DOAS. To determine the results of calibration, it was checked whether $R^2 \geq 0.98$ (which is the standard for linearity test results among the internal quality control regulations in the Korean Air Pollution Measurement Network operation guidelines) was satisfied. Afterward, the UV-DOAS was installed next to the Dokgot-ri Air Quality Monitoring Station located in Daesan-eup, Chungcheongnam-do, Korea to check whether UV-DOAS on-site measurements were possible. SO₂, NO₂, and benzene were measured for 19 days from March 13 to March

31, 2021. The results were compared with the Dokgot-ri measurement station data to confirm the reliability of the UV-DOAS measurement values and determine whether on-site measurements were possible. Finally, a UV-DOAS was installed on one side of the site boundary of a plant approximately 950 m in length located in the petroleum refining industrial complex in Yeosu-si, Jeollanam-do, Korea, and SO₂, NO₂, and benzene were measured from November 29, 2021, to January 19, 2022. To understand the behavioral characteristics of the pollutants, UV-DOAS measurements and meteorological observation data were analyzed.

UV-DOAS Analyzer

UV-DOAS analyzers can measure various chemical species in an optical path free of obstacles, allowing light to pass through. Currently, Korea's air pollution monitoring network measures pollutants in the air by sampling and analyzing samples at points. The UV-DOAS analyzer had a larger spatial measurement range and good measurement time resolution, enabling real-time measurement compared with the measurement methods of the air pollution monitoring network and the UV-DOAS measurement method. Therefore, it is advantageous to identify trends in the concentration of the target substance.

The UV-DOAS used in this study was an AR500 model commercially developed by a Swedish manufacturer, OPSIS. The equipment consists of a pair of emitters (EM150) and receivers (RE150) to form an optical path and is composed of an optical fiber cable that transmits to the analysis unit with little wavelength loss. The light source was equipped with a xenon lamp with a power of 150 W and it could stably irradiate in the 200–400 nm range, which is the UV wavelength necessary for measuring pollutants. The light emitted from the light source reaches the light receiver and finally the analyzer, which is used to measure pollutant concentration in the optical path. The analysis department performed spectroscopy using the grating method. The concentrations of the pollutants were measured using the absorbance wavelength. DOAS technology for analyzing pollutants follows the Lambert–Beer law and uses the principle of measuring the absorption luminosity of chemical species present as pollutants in the air that react with the wavelength of light.

Selecting Validation Measurement Data

The UV-DOAS could adjust the measurement time for each measurement item. It has the advantage of performing real-time measurements, because the measurement time interval is short. Additionally, it indicates a monitoring value called the light level, which is a numerical value that represents the luminance of the light received when transmitting light from the emitter to the receiver (0–100%). The light level

was used to determine whether the emitter and receiver were in the correct line. If the light level is low, sufficient light for analysis does not reach the analyzer, which can adversely affect the correct measurement values. Data where the light level was less than 20% of the 30-min average were classified as unreliable and excluded from the 24-h average value. Additionally, 48 30-min average data items were generated per day, and a day was completely excluded from the measurement if there were more than 24 pieces of data that were less than 20% of the 30-min average light level data. Consequently, data from 14 of 19 days measured in Daesan-eup, Chungcheongnam-do, and data from 52 of 60 days measured in Yeosu-si, Jeollanam-do were used.

UV-DOAS Calibration Method

Before installing the UV-DOAS in the field, calibration was performed using a standard gas to confirm the reliability of the measurement data. The equipment for calibrating the UV-DOAS analyzer consisted of a light source, optical receiver, optical gas cell, and calibration bench on which the optical gas cell could be placed, analysis unit, optical fiber cable, and standard gas. Light sources and optical receivers installed and measured in the field have large reflectors because they need to send light far away, whereas those used for calibration do not need to send light far away; therefore, the reflectors are small. However, the xenon lamp used as the light source and the material of the reflector was the same; therefore, there was no change in the wavelength required for analysis. The measurement times and distances were identical to those used in the actual field. The measurement time was set to 30 s for SO₂ and NO₂ and 60 s for benzene, and the measurement distance was set to 870 m.

The laboratory where the calibration was performed had a constant temperature of 21 °C. Humidity was not measured, because the standard gas was a dry gas diluted with N₂. All the measurement data were expressed to the first decimal place and a calibration procedure was performed after performing a reference calibration to check whether the analysis unit was operating normally.

The calibration parameters used were SO₂, NO₂, and benzene, and 99.999% N₂ gas was used as the zero point. The concentrations of SO₂ and NO₂ were 192 ppm and 188 ppm, respectively, as certified by Korea, but the concentration of benzene was 50 ppm, which was not officially certified, so it was replaced with the gas manufacturer's report. The concentration analyzed in the analysis section varied depending on the standard gas concentration and the length of the optical gas cell. The standard gas had one concentration for each item, but the optical gas cell had three lengths: 100, 250, and 350 mm. Linearity was confirmed by varying the analyzed concentrations.

Verifying UV-DOAS Field Applicability

In Korea, statistics on chemical emissions by region and company are publicly available. In Korea, oil refineries, chemical plants, and steel mills are located in Chungcheongnam-do and Jeollanam-do, where various types of air pollutants (such as SO₂, NO₂, and benzene) are emitted. As of 2021, approximately 51 tons of benzene will be emitted annually in Chungcheongnam-do and 31 tons per year in Jeollanam-do. The UV-DOAS was installed immediately next to the Dokgot-ri measuring station in Daesan-eup, Chungcheongnam-do, and the UV-DOAS measurement data were compared with those of the Dokgot-ri measuring station (Fig. 1a). The Dokgot-ri measurement

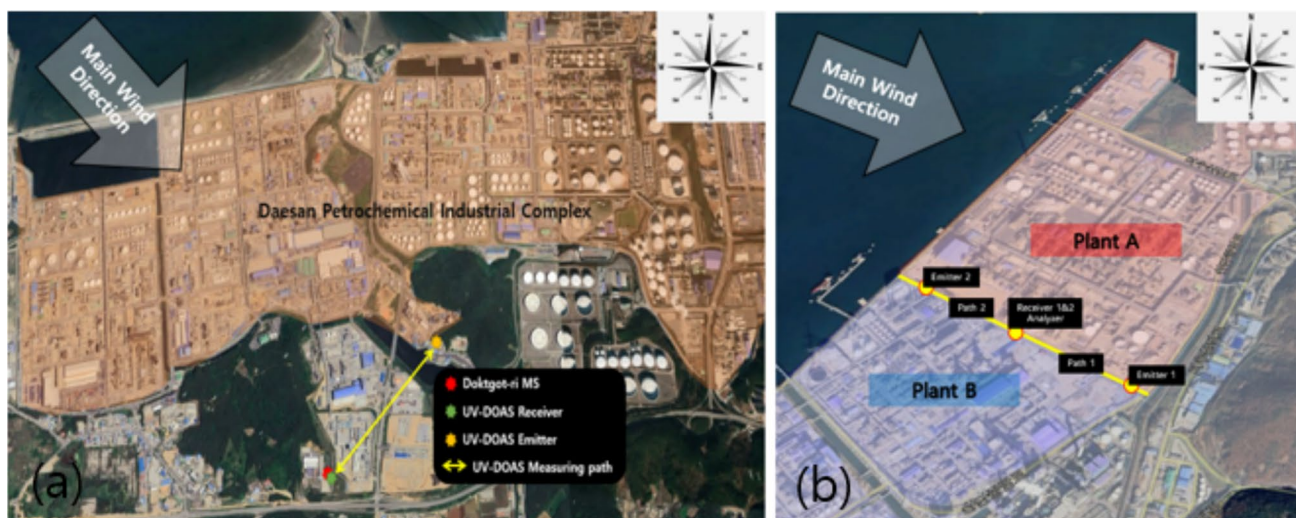


Fig. 1 Two measurement areas of UV-DOAS; **a** Daesan-eup test bed location and **b** Yeosu-si test bed location

station is operated by the Ministry of the Environment of Korea and the measured data are disclosed to the public. In this experiment, the Dokgot-ri measurement station measurement data were considered reliable. All Dokgot-ri measurement station data were converted to 24-h average data. Using the converted 24-h average data, we compared the differences and trends in the measured data and verified whether the UV-DOAS can measure SO₂, NO₂, and benzene in the atmosphere in the field.

Fence Line Monitoring of Petroleum Refining Plant

A UV-DOAS was installed at the site boundary between two different petroleum refining plants located in Yeosu-si, Jeollanam-do, and the SO₂, NO₂ and benzene concentrations were measured (Fig. 1b). The two petroleum refining plants are operated by different companies and have different processes; therefore, it was expected that the characteristics of the pollutants emitted into the air would differ. Plant A included crude-oil refining, aromatic production facilities, and flare stacks, whereas plant B included PVC production and NCC processes.

The measured concentration, wind direction, and wind speed data were used to analyze and create a pollution map. Wind direction and wind speed data were used from the Korea Meteorological Administration Disaster Prevention and Weather Observation (AWS, Automatic Weather Observation System) Yeosu Industrial Complex branch (34.84946, 127.71496). We confirmed the direction of pollutants that emitted high concentrations using pollutant concentration data based on wind direction and wind speed. This study measured plants A and B (which are expected to have different air pollutant emission characteristics) to determine which plant contributed the most. Subsequently, SkyDOAS was employed to track the main emission sources in detail. SkyDOAS is an analyzer developed by FluxSense in Sweden to measure atmospheric pollutant emissions. It measures pollutants between the sun and the measurement equipment and calculates emissions. The analyzer utilizes a telescope made of optical fiber and a Czerny-Turner spectrometer with a CCD camera to measure scattered light in the UV and visible ranges at the zenith angle. The system comprises a quartz telescope connected via an optical fiber to a 303 mm focal length spectrometer, which features a thermoelectrically cooled CCD camera with a resolution of 1024 × 255 pixels. In Korea, there is a research case that measured and calculated sulfur oxide emissions using vehicle equipped with SkyDOAS [18]. Using SkyDOAS, measurements were obtained while driving inside a plant that contributed to high-concentration emissions, and the main source was tracked.

Results and Discussion

Verification of UV-DOAS Analyzer Reliability

A calibration experiment was performed using standard gas to verify the reliability of the UV-DOAS analyzer, and the possibility of field application was confirmed by installing the UV-DOAS near the Dokgot-ri measuring station in Daesan-eup, Chungcheongnam-do, Korea. A linearity of $R^2 = 0.99$ was confirmed for SO₂, NO₂, and benzene using the UV-DOAS calibration experiment. This satisfies the linearity test result standard of $R^2 \geq 0.98$ among the internal quality control regulations in the Korea Air Pollution Monitoring Network operation guidelines. This result confirms the reliability of the UV-DOAS data.

After confirming the device performance of the UV-DOAS using a standard gas, the UV-DOAS was installed next to the Dokgot-ri measurement station and compared and analyzed with the Dokgot-ri station measurement data. The R^2 values of SO₂, NO₂, and benzene were 0.82, 0.84, and 0.71, respectively, confirming similar trends (Fig. 2). However, the Dokgot-ri measuring station measurement method is a point measurement method that collects samples at one point and analyzes only the collected samples, whereas the UV-DOAS measurement method forms an optical path similar to a line and measures pollutants moving across the optical path. Differences may have occurred, because this was a line-measurement method. However, the difference was small, and the trends were similar, confirming the reliability of the measurement data and the field applicability of the UV-DOAS.

Petroleum Refining Plant Fence Line Monitoring Results

After confirming the reliability of the UV-DOAS, it was installed and measured on one side of the site boundary of a petroleum refining plant in Yeosu, Jeollanam-do, Korea. Wind direction and speed data were used to analyze the measurement results and identify the main emission sources of specific pollutants. Disaster prevention weather observation data approximately 2 km from the measurement point were used for wind direction and wind speed data. During the monitoring period, the main wind direction was west northwest (WNW), the average wind speed was 4.39 m/s, and the temperature ranged from -4.5 to 10.9 °C in winter.

Generally, SO₂ and NO₂ are emitted from combustion processes. However, since many employees work in the petrochemical industry, it is expected that emissions will also be emitted from vehicles such as passenger cars

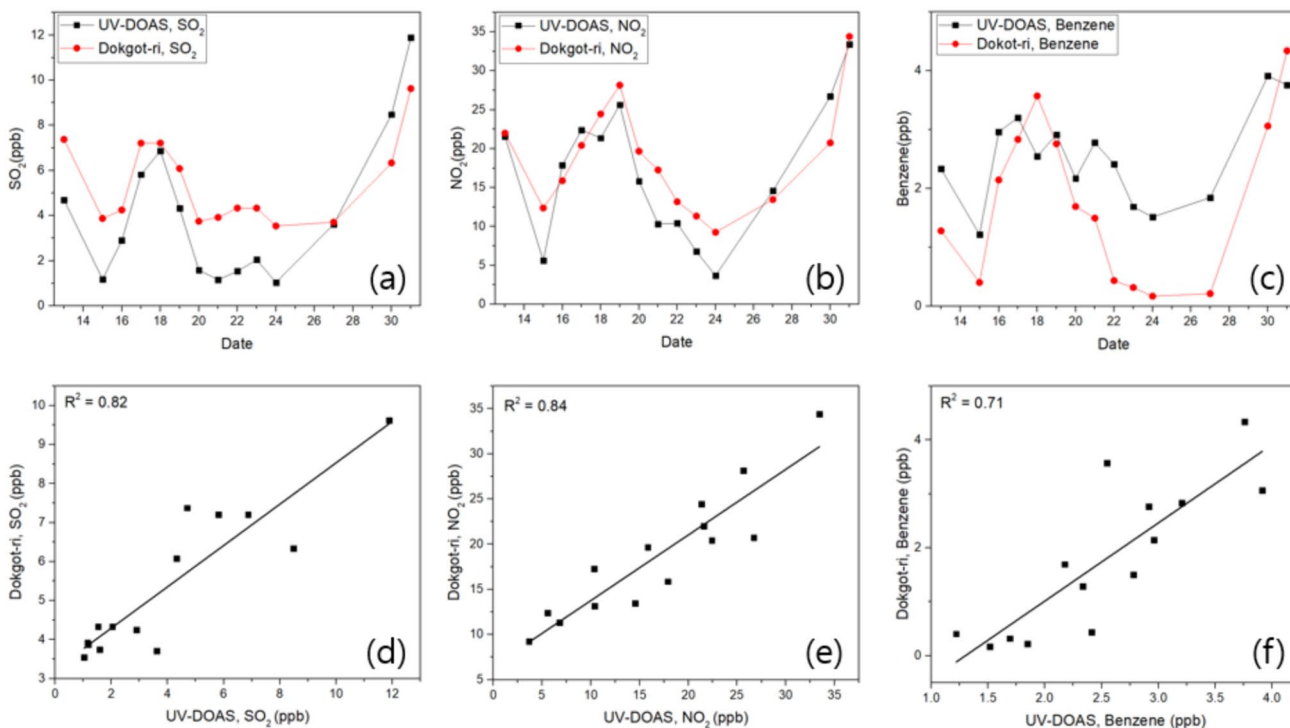
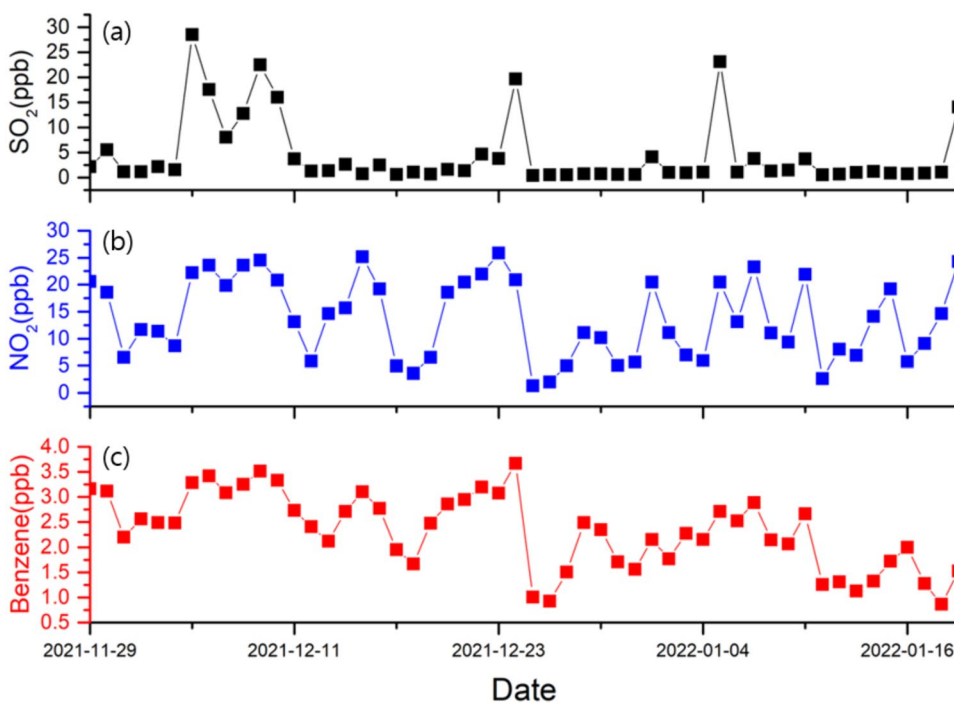


Fig. 2 UV-DOAS and Dokgot-ri measuring station daily average data of **a** SO₂, **b** NO₂, and **c** benzene and R² result, the correlation coefficient between UV-DOAS and Dokgot-ri measurement station data of **d** SO₂, **e** NO₂, and **f** benzene

driven by employees or industrial trucks. Benzene is emitted from chemical reactions and separation processes, but can also leak or be released from flanges and storage tanks.

In the case of SO₂ among the measurement results, high concentrations were observed intermittently; however, the trends of NO₂ and benzene were different from those

Fig. 3 Daily average results measured at Yeosu-si test bed for **a** SO₂, **b** NO₂, and **c** benzene



of SO_2 (Fig. 3). Additionally, NO_2 and benzene fluctuated less than SO_2 and were less affected by wind direction. This could have led to the possibility that NO_2 and benzene emission sources were scattered throughout the area. NO_2 and benzene were being emitted from Plant A, but they were not dominant, and it could be expected that they were being emitted from vehicles or from Plant B. Therefore, wind direction and wind speed data were used to identify the emission sources of SO_2 , NO_2 , and benzene and to confirm the pollutant behavior characteristics. High concentrations of pollutants were generally observed in the direction of plant A, but mean concentrations of pollutants were also observed in the direction of plant B in the case of NO_2 and benzene (Fig. 4). However, extreme emissions were emitted from plant A in the case of SO_2

(Fig. 5). The SO_2 concentration was higher than usual for 7 days: December 5, December 6, December 8, December 9, December 10, December 24, 2021, and January 5, 2022. On 5 out of 7 days, high concentrations were observed when the main wind was northeasterly; however, on 2 days (December 10, 2021, December 24, 2021), high concentrations were observed when the main wind was northwesterly (sea breeze). However, it was necessary to examine the SO_2 concentration in detail for the wind direction, because the main wind speed was considered. We confirmed that high concentrations were monitored when the wind was blowing east from plant A when we checked the SO_2 data on December 10 and 24, 2021 in detail (Fig. 6). Meanwhile, it was difficult to identify specific emission sources for NO_2 and benzene and it was difficult to confirm

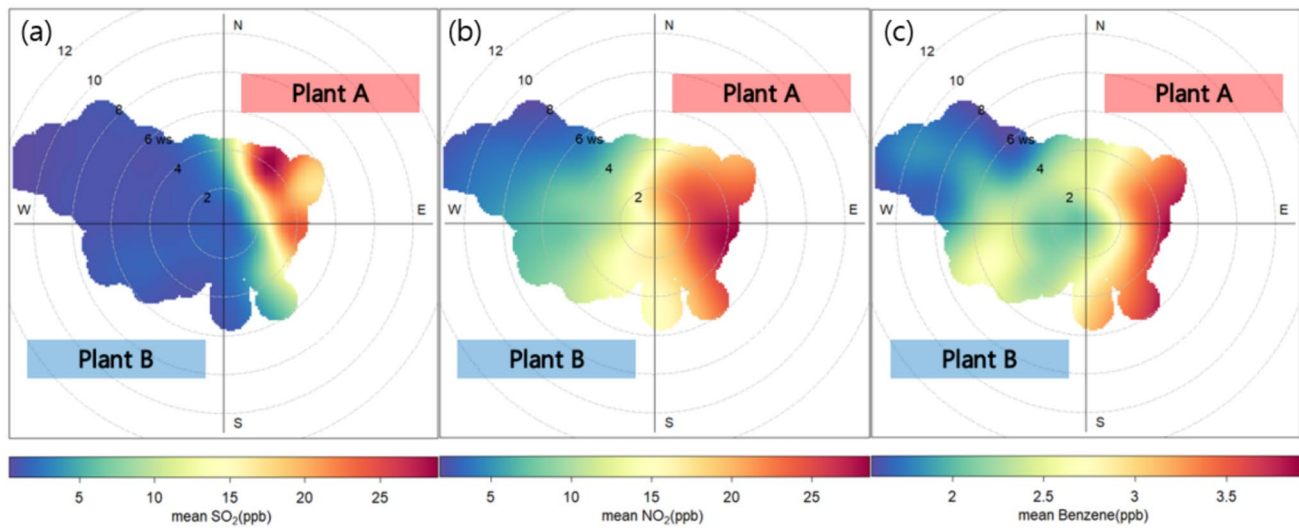


Fig. 4 Pollution map using Yeosu-si measurement results for **a** SO_2 , **b** NO_2 , and **c** benzene

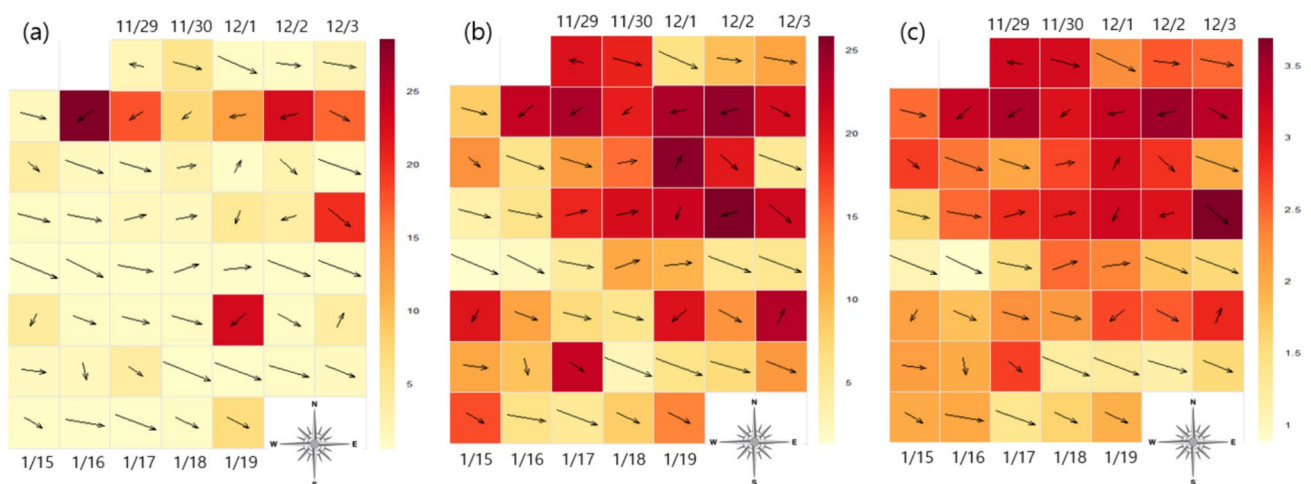


Fig. 5 Daily wind direction (arrow direction), wind speed (arrow length), and concentration (color) of **a** SO_2 , **b** NO_2 , and **c** benzene

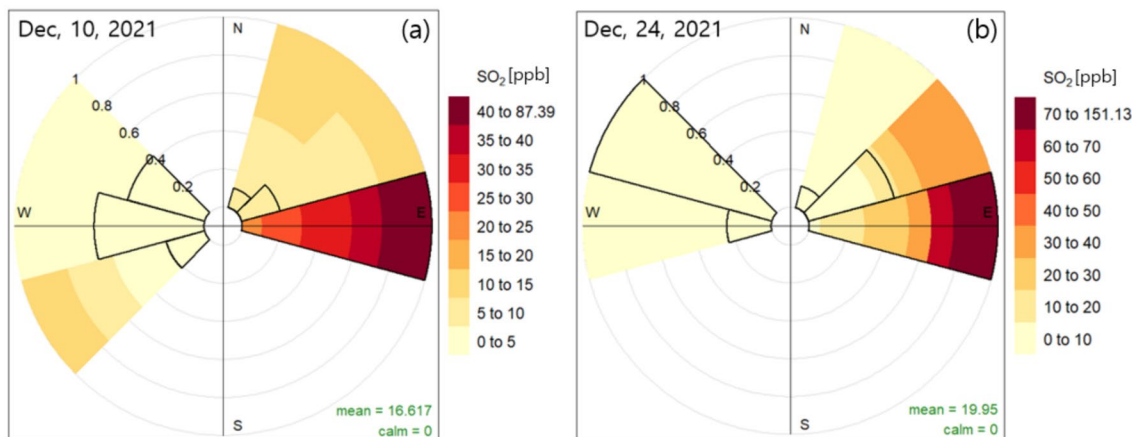


Fig. 6 SO₂ concentration results according to wind direction on a day when the main wind is northwest and SO₂ is high on **a** Dec 10 and **b** Dec 24 in 2021

the correlation with wind direction. There is a possibility that emission sources are scattered or that fugitive emissions occur.

As the measurement point is an industrial complex where petroleum refining plants are located, there were some differences in the measured concentrations between weekdays and weekends during the measurement period. The concentrations of all the measured compounds tended to decrease on weekends rather than on weekdays (Table S1). The weekend concentration of SO₂ was 3.37 ppb, which was 28% lower

than that of the weekday concentration (4.67 ppb). The NO₂ concentration decreased by 32% to 14.47 ppb on weekdays and 9.84 ppb on weekends. The benzene concentration decreased by 16% to 2.47 ppb on weekdays and 2.07 ppb on weekends. The wind rose on weekdays and weekends during the measurement period (Fig. 7). The wind direction and speed were similar on weekdays and weekends, which appears to be owing to the operation rate of the emission source and the reduction in traffic volume, rather than the effect of the wind direction and speed.

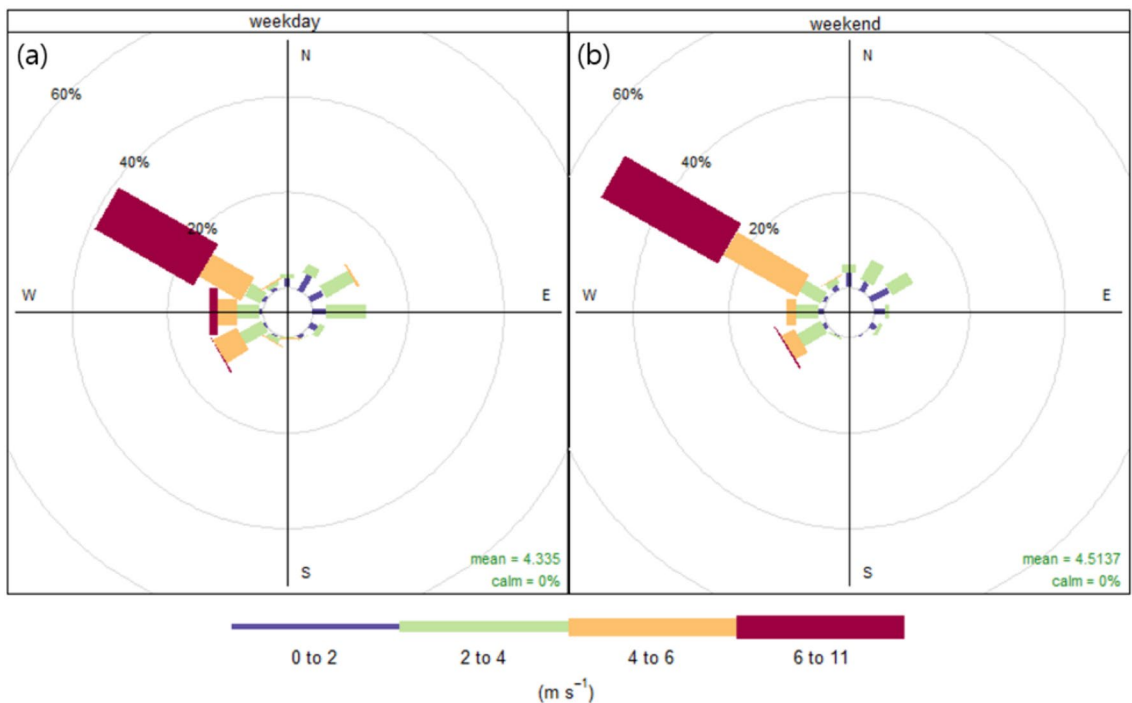


Fig. 7 Yeosu-si test bed wind rose based on a **a** weekday average and **b** weekend average

The UV-DOAS measurement results were compared with those from the Wolnae-dong Atmospheric Environment Monitoring Station to determine the emission characteristics of SO₂ and NO₂ (Fig. 8). SO₂ and NO₂ were measured at the Wolnae-dong Atmospheric Environment Monitoring Station, approximately 3.7 km away from the UV-DOAS. Despite being 3.7 km away from each other, there was little difference in the measured data for NO₂ and a similar trend was observed. However, the concentration of SO₂ did not exhibit a similar trend. We confirmed that SO₂ had an

emission source near the UV-DOAS by comparing the pollution map results with the Wolnae-dong Air Quality Monitoring Station measurement data. Therefore, efforts are required to manage it.

SO₂ Measurement Inside Plant A Using SkyDOAS

Plant A was expected to emit high concentrations of SO₂. A high peak of SO₂ was confirmed near the flare stack using SkyDOAS (Fig. 9). Gas containing sulfur is

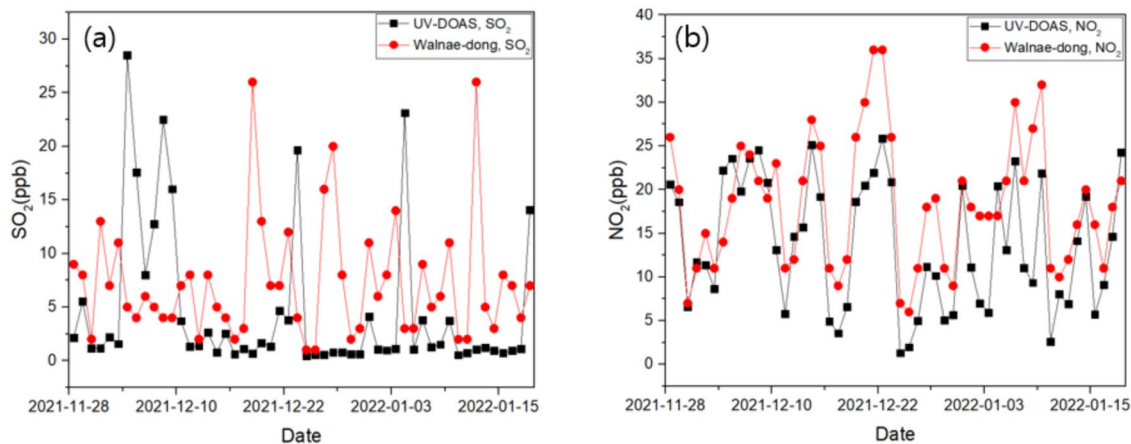


Fig. 8 UV-DOAS and Wolnae-dong measuring station daily average data of a SO₂ and b NO₂

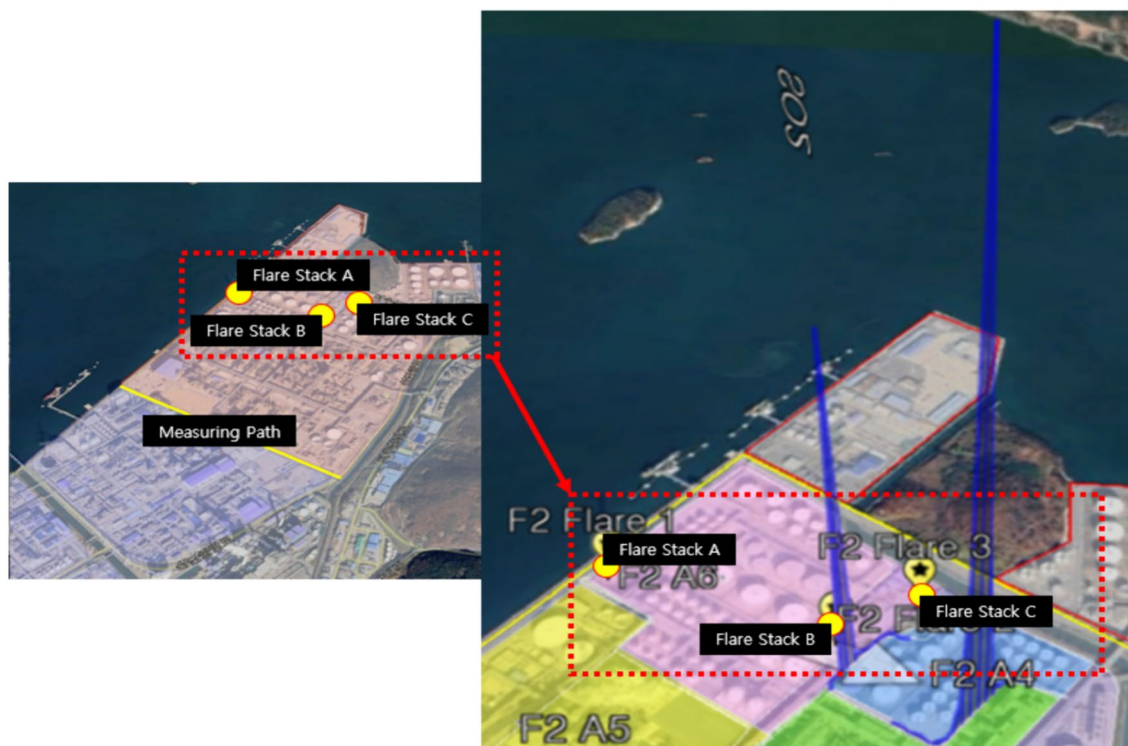


Fig. 9 SO₂ peak measured with SkyDOAS (blue) and flare stack location in plant A

emitted from the flare stack without being incinerated or processed. Currently, there are insufficient measures for managing the pollutants emitted from flare stacks. Therefore, a system that can simultaneously manage point and fugitive emission sources (such as fence line) is required.

Conclusions

This study obtained basic data on the fence line monitoring method, which is a new management method at the plant or company site level, rather than on the existing air pollutant emission facility management method in Korea. The UV-DOAS (the optical remote-sensing device used in this study) is useful for monitoring real-time data on air pollutants emitted from point or fugitive emission sources. It was more economical, because no manpower was required for sample collection and analysis. It can also be measured over a long period of time, and it is easy to check the behavioral characteristics of air pollutants emitted from emission sources and track major emission sources using measured concentration, wind direction, and wind speed data. Based on the monitoring results of UV-DOAS, it is possible to identify major emission sources that were previously unknown by utilizing measurement instruments such as SkyDOAS, which can measure the vicinity of the plant process. An integrated concentration profile was obtained by driving downwind from the source and intersecting the plume with the measurement path. This can help in planning specific means and methods for managing sources. This method is expected to be used in future air pollutant reduction evaluations.

Fence line monitoring is a method of measuring in the immediate vicinity of a plant, and the measured concentration varies depending on the distance between the emission source and the measurement point, making it difficult to manage by applying existing Korean air quality standards. SO₂ and NO₂ are maintained at low concentrations in the atmosphere through continuous emission control and the development of reduction facilities; however, it is more difficult to meet air quality standards for benzene. In this study, 2.8 ppb benzene was observed, which is higher than the Korean benzene air quality standard of approximately 1.5 ppb (the Korean benzene air quality standard is an annual average, but in this study, it was an average of 52 days). Therefore, long-term monitoring is necessary, but it is expected to be monitored at a higher concentration than the air quality standard. Therefore, separate standards are required for fence line monitoring of benzene. Standards that consider human risks and long-term measurement results must be established to introduce fence line monitoring regulations.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s11814-024-00220-3>.

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Data availability The dataset used in this study are available upon request from the corresponding author.

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