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Characteristics of the Chemical Components of PM_{2.5} in the Dangjin Region, South Korea, and Evaluation of Emission Source Contributions During High-Concentration Events

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Abstract

Fine particulate matter (PM_{2.5}; aerodynamic diameter $\leq 2.5 \mu\text{m}$) remains a challenging policy for industrialized coastal regions throughout East Asia. In this study, we present a multi-year chemical characterization of PM_{2.5} and identify key factors contributing to extreme pollution events in Dangjin, a heavy-industry hub on Korea's west coast. Between August 2020 and March 2024, 24-h gravimetric filters (up to $n = 245$; 127–280 valid analyses depending on constituent) were collected twice weekly in winter–spring and weekly in summer–autumn. Meteorological data and 48-h backward HYSPLIT trajectories guided source interpretation. The mean PM_{2.5} concentration was $26.22 \pm 15.29 \mu\text{g}/\text{m}^3$ ($4.74\text{--}95.31 \mu\text{g}/\text{m}^3$). The mass was highest in winter ($30.83 \mu\text{g}/\text{m}^3$). Secondary inorganic ions constituted 60.3% of the aerosol, with nitrate comprising 29.7%. A nitrate-to-sulfate ratio of 1.94 indicated a stronger influence from mobile NO_x emissions compared to that from coal combustion. The trajectory analysis showed north-easterly transport from Eastern China, followed by local stagnation, which promoted rapid ammonium-nitrate formation. Regional transport contributes to severe PM_{2.5} episodes, with their magnitude increased by local NO_x and NH₃ emissions. Our findings suggest that effective mitigation strategies in coastal industrial corridors require coordinated control of long-range transport and domestic measures focused on vehicles and ammonia-rich industries.

Keywords: PM_{2.5} concentration; heavy metals; carbonaceous species; water-soluble ions



Received: 29 August 2025

Revised: 5 October 2025

Accepted: 6 October 2025

Published: 13 October 2025

Citation: Kim, Y.-h.; Park, S.-Y.; Jang, H.; Moon, J.-E.; Lee, C.-M.

Characteristics of the Chemical Components of PM_{2.5} in the Dangjin Region, South Korea, and Evaluation of Emission Source Contributions During High-Concentration Events. *Toxics* **2025**, *13*, 869. <https://doi.org/10.3390/toxics13100869>

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1. Introduction

PM_{2.5} has been regulated in Korea under national air quality standards since 2015. PM_{2.5} with an aerodynamic diameter of $<2.5 \mu\text{m}$, which is smaller than that of PM₁₀, can penetrate deep into the lungs and exacerbate respiratory diseases such as asthma, as well as increase the risk of premature death owing to cardiovascular complications [1,2]. The World Health Organization has categorized PM_{2.5} as a Group 1 carcinogen, posing a serious threat to public health [3]. Additionally, PM_{2.5} has a relatively high number of particles per unit volume and a large surface area, facilitating the adsorption of various chemical substances, such as heavy metals, ionic species, and carbonaceous components. Toxic elements such as Cd and As, which have adverse health effects, can be introduced into the human body via PM_{2.5} and may result in various negative health outcomes [4].

The primary chemical constituents of $PM_{2.5}$ comprise water-soluble ions (SO_4^{2-} , NO_3^- , and NH_4^+), carbonaceous species (organic and elemental carbon), and metals (Fe, Pb, and Cr) [5]. The composition and concentration of these constituents vary based on the characteristics of proximal emission sources [6].

In South Korea, high $PM_{2.5}$ concentrations frequently result from domestic sources and long-range transported aerosols outside the country [7,8]. Typically, foreign contributions are estimated to account for 30–50% of the annual average concentration and up to 60–80% during high-concentration episodes. A joint study by the National Institute of Environmental Research of Korea, China, and Japan estimated that domestic sources contribute approximately 51.2% to annual $PM_{2.5}$ levels in Korea. In contrast, China and Japan contribute 32.1% and 1.5%, respectively [9].

Dangjin, a heavy industry hub on Korea's west coast, has several primary sources, such as coal-fired power plants, industrial complexes, and steel facilities near residential and commercial areas. Consequently, local emissions considerably influence particulate matter levels, and high-concentration events have also been observed owing to long-range transport from China. Additionally, according to the 2022 Air Quality Annual Report published by the National Institute of Environmental Research, Dangjin, which is situated in the Chungnam region, exhibits the highest average PM_{10} and $PM_{2.5}$ concentrations in the province. Consequently, numerous studies have been conducted on particulate matter's spatial and temporal distributions and source identification to develop effective control strategies [10–12].

We investigated the $PM_{2.5}$ concentrations and their chemical components in Dangjin over 3 years and 7 months, from August 2020 to March 2024. We characterized seasonal variations and compared our findings with previous studies to identify the distinctive features of $PM_{2.5}$ concentrations in Dangjin. Additionally, through multifaceted analyses during high-concentration events, we explored the influencing factors to obtain foundational data and inform future $PM_{2.5}$ management strategies and policy developments in the region.

2. Materials and Methods

2.1. Monitoring Site

Dangjin is an industrial region with a national industrial complex and several general industrial complexes, 13 comprising the Seongmun National Industrial Complex, which covers 12.0 km². However, part of the Asan National Industrial Complex is officially located in Asan, which borders Dangjin. In particular, the general industrial complexes are also extensive; however, the national industrial complexes alone cover approximately 16,877 m² [13]. To investigate the distribution characteristics of $PM_{2.5}$ and its chemical components in Dangjin's ambient air, we selected a monitoring site in a residential area close to major industrial facilities.

The air monitoring site was set at coordinates (36°56'30.4'' N, 126°47'04.1'' E) to represent the air quality of both industrial and residential areas in central Dangjin (Figure 1). To ensure an accurate representation of the local air quality without notable nearby interference, samples were collected from a building rooftop presumed to be minimally affected by immediate surrounding sources.

As of July 2024, 496 companies were operating within Dangjin's industrial complexes. Of these, 200 companies were in the Seongmun Industrial Complex (one of the national industrial complexes), and 142 companies were in the Godae-Bugok District of the Asan National Industrial Complex, which partially extends into Dangjin. The operational rates for these complexes were 46% and 95.7%, respectively. Machinery, petrochemicals, and steel accounted for approximately 76% of the industries within the Seongmun Industrial

Complex; other operations included valve manufacturing, metal storage, chemical dye production, and surface treatment. In the Asan Industrial Complex, steel, semiconductors, power generation, and automobile parts manufacturing accounted for approximately 50% of all industries [14].

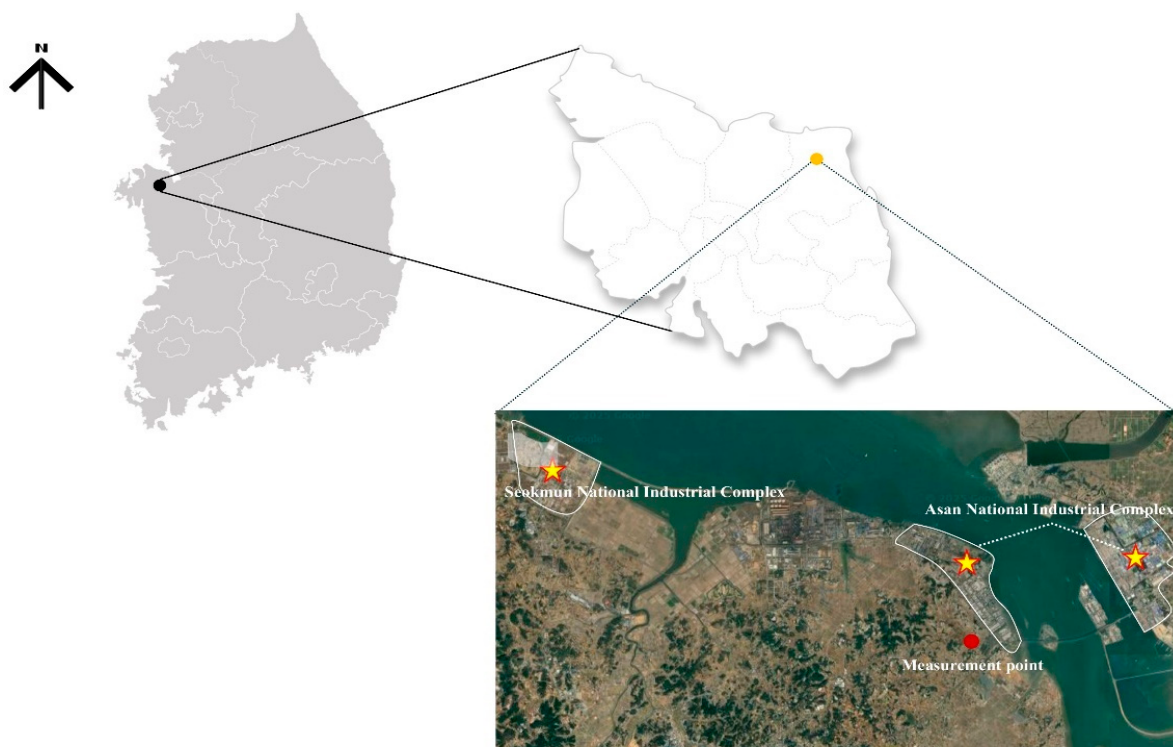


Figure 1. Air monitoring stations within the Dangjin industrial complex. Background map source: Google Maps <https://www.google.com/maps> (accessed on 10 April 2025).

2.2. Sample Collection

This study was conducted over 3 years and 7 months, from August 2020 to March 2024, by selecting measurement points and collecting samples in a national industrial park in Dangjin. Measurements were conducted twice a week (Monday and Wednesday) during winter, which typically has high concentrations of fine particulate matter [15], and during spring, which is frequently affected by yellow dust [16]. During summer and autumn, measurements were conducted once a week (Monday). The dataset and subsequent analyses excluded samples affected by extreme weather events such as typhoons, heavy rainfall, or instrument failure, and the overall sampling strategy followed the protocol described in our previous study [17]. PM_{2.5} was collected using a gravimetric method with a low-volume air sampler (PMS-204, APM Co., Bucheon, Republic of Korea), which conforms to the U.S. EPA Federal Reference Method for PM_{2.5} measurement (Online Resource 1).

Sampling ran continuously for 24 h from 09:00 at a constant flow of 16.7 L/min on each measurement day. All equipment setup, sample acquisition, and transport activities were implemented strictly with the “Guidelines for Installation and Operation of Air Pollution Measurement Networks” provided by the Ministry of Environment and the National Institute of Environmental Research [18]. Two independent sampling devices were employed to facilitate analysis of ionic constituents, trace metals, and carbon fractions. Each device was outfitted with Teflon filters (PTFE, 2.0 µm pore size, Ø 27 mm) and quartz fiber filters (Pure Quartz fiber, Ø 47 mm), following NIER specifications.

The Teflon filters were placed in sterile Petri dishes, sealed with paraffin film, and refrigerated until analysis, and the quartz filters were used after the heating treatment. To

prevent the loss of PM_{2.5} components through volatilization and reaction, the collected dust was transported in an ice box filled with refrigerant to maintain a low temperature, and a buffered transport device was used to prevent loss of PM_{2.5} particles owing to shaking or impact during transport, and to prevent static electricity.

2.3. Gravimetric Analysis

Each filter was conditioned and weighed for 24 h under controlled temperature and humidity before and after sample collection. Laboratory blanks (LAB) and field blanks (FB) were used to correct the measured PM_{2.5} mass concentrations. For each batch of filters, a new LAB was kept in the laboratory during the sampling period and reweighed post-collection to verify measurement accuracy. Similarly, a fresh FB was transported to the field site without exposure to airflow, then returned and reweighed to assess handling stability. The PM_{2.5} mass concentration was subsequently calculated using the following equation:

$$\text{PM}_{2.5}(\mu\text{g}/\text{m}^3) = \frac{(W_f - W_i) - (W_{\text{FBf}} - W_{\text{FBi}})}{V_a} \quad (1)$$

where W_i and W_f denote the initial and final weights (μg) of the LAB filters; W_{FBi} and W_{FBf} indicate the weights (μg) of the FB filters measured before and after PM_{2.5} sampling; and V_a represents the total air volume (m^3) drawn through the sampler.

2.4. Ionic Composition Analysis

The ionic constituents of PM_{2.5} were determined, targeting five cationic (Na^+ , NH_4^+ , K^+ , Mg^{2+} , and Ca^{2+}) and three anionic species (SO_4^{2-} , NO_3^- , and Cl^-). To enhance analyte extraction efficiency, the filter was positioned such that its collection surface faced the bottom of a beaker, and 200 μL of ethanol (reagent grade or higher) was added to promote precipitation. Subsequently, 20 mL of ultrapure water (resistivity $\geq 18 \text{ M}\Omega\cdot\text{cm}$) was introduced, and the solution was stirred at 120 rpm for 120 min using a magnetic stirrer.

Following the extraction, the liquid was passed through a 110 mm diameter membrane or a syringe filter with a 0.1- μm pore size. Afterward, ionic species were quantified using ion chromatography (IC) under conditions presented in Online Resource 2.

Calibration standards were prepared for each target ion, and calibration curves were constructed accordingly. The coefficient of determination (R^2) for all analytes was >0.999 , indicating high analytical linearity. The atmospheric concentrations of ionic species in PM_{2.5} were then computed using the following equation:

$$C = ((X_1 - X_2) \times S) \div F \quad (2)$$

where C denotes the atmospheric concentration of ionic species ($\mu\text{g}/\text{m}^3$); X_1 and X_2 are the concentrations ($\mu\text{g}/\text{mL}$) in the sample and blank filters, respectively; S indicates the extracted solution volume (mL); and F refers to the total sampled air volume (m^3).

2.5. Carbon Component Analysis

The flame ionization detector was used to analyze the carbon component of PM_{2.5} (Online Resource 3). The collected filters were baked at 550 $^\circ\text{C}$ for >4 h to remove impurities such as organic components and cut into 1.5 cm^2 sample pieces for analysis. To prevent sample contamination owing to the use of sample autoinjectors, a maximum of 10 samples were analyzed consecutively. Additionally, the temperature was maintained at a low level, and the analysis was conducted on days with high humidity. The concentration of carbon in PM_{2.5} was calculated using the following equation:

$$C = ((X_1 - X_2) \times S) \div F \quad (3)$$

where C denotes the atmospheric concentration of carbon species ($\mu\text{g}/\text{m}^3$); X_1 and X_2 are the surface concentrations ($\mu\text{g}/\text{cm}^2$) from the sampled and blank filters, respectively; S indicates the analyzed filter area (cm^2); and F refers to the total volume of air sampled (m^3).

2.6. Trace Elemental Composition Analysis

The trace elemental composition of $\text{PM}_{2.5}$ was analyzed using energy-dispersive X-ray fluorescence (ED-XRF). A total of nineteen elements were quantified, including Al, Ti, V, Mn, Fe, Ni, Co, Zn, As, Sr, Mo, Cd, Ba, Pb, P, S, Cr, Si, and Cu. The procedures were carried out in accordance with the national guidelines for air pollution monitoring networks established by the Ministry of Environment and the National Institute of Environmental Research. Technical details of the analytical equipment are provided in Online Resource 4.

ED-XRF, which has a three-dimensional optical system structure, is a quantitative and qualitative analysis instrument that measures the wavelength and intensity of unique fluorescent X-rays emitted by each element and can measure harmful substances in the atmosphere in extremely small amounts. Standard solutions were prepared for the analysis of trace elemental composition. Additionally, calibration curves were generated for each standard substance, and the coefficient of determination (R^2) was >0.999 . The concentration of trace elemental components in $\text{PM}_{2.5}$ was calculated using the following equation: the mass of components in 1 m^3 of air at 0°C and 760 mmHg .

$$C = \frac{(C_s - C_{bk}) \times A_u}{V_s} \quad (4)$$

where C represents the concentrations ($\mu\text{g}/\text{m}^3$) of the trace elements in the atmosphere, C_s is the mass (ng/cm^2) of heavy metals collected on the filter, C_{bk} is the mass (ng/cm^2) of heavy metals on the blank filter, A_u is the total area (cm^2) of the filter used for sampling, and v_s is the total volume (m^3) of the air sampled.

2.7. Quality Control (QA/QC)

To calculate the accurate value of the measured item, precision control was performed once yearly, and the method detection limit (MDL), relative standard deviation, and the coefficient of determination of the black curve (R^2)'s straightness were calculated for precision control. The MDL is the standard deviation of the measurement multiplied by 3.14, and the relative standard deviation (unit: %) is the standard deviation divided by the mean value of n consecutive measurements and multiplied by 100. Finally, a calibration curve was generated using two or more standards with concentrations within the quantification range, and the coefficient of determination of the straightness of the curve was calculated. If the R^2 is <0.99 , the curve is rewritten. The MDL values ranged from 0.001 (Ag) to $1.547 \mu\text{g}/\text{m}^3$ (Sb), and the R^2 of the calibration curves for each component was >0.99 .

2.8. Statistical Analysis

This study calculated descriptive statistics such as mean, standard deviation, and minimum and maximum values to determine the spatial and temporal distribution of $\text{PM}_{2.5}$ concentrations and chemical constituents in $\text{PM}_{2.5}$. Subsequently, Pearson correlation analysis was used to evaluate the correlation between major ionic components (NO_3^- , SO_4^{2-} , and NH_4^+), carbon components (organic carbon [OC] and elemental carbon [EC]), and trace elements, and one-way analysis of variance was used to test for seasonal and annual differences in concentrations. The significance level for all statistical analyses was set at 0.05 . In addition, we identified the causes of high concentrations by comparing

the increase in major elements during high-concentration events ($\text{PM}_{2.5} > 75 \mu\text{g}/\text{m}^3$) to normal times.

2.9. Weather Information Processing and the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) Model

Meteorological data from Dangjin City were used to conduct atmospheric analysis, based on records obtained from the Automatic Weather System operated by the Korea Meteorological Administration. To determine the origin of increased $\text{PM}_{2.5}$ concentrations, backward trajectory simulations were performed using the HYSPLIT model. Developed as part of the Real-time Environmental Applications and Display System by the National Oceanic and Atmospheric Administration, HYSPLIT estimates atmospheric transport pathways based on outputs from numerical weather prediction models.

In this study, meteorological input fields were obtained from the Global Data Assimilation System, operated by the National Centers for Environmental Prediction, with a temporal resolution of 1 h and a spatial resolution of 1° . Backward trajectory simulations were conducted using the HYSPLIT model with meteorological input from the Global Data Assimilation System (GDAS1, $1^\circ \times 1^\circ$, 1 h resolution). Trajectories were computed for a 72 h backward period, starting at arrival heights of 500, 1000, 2000, and 3000 m above ground level, with additional cases at 5000 m to capture higher-altitude transport. The arrival point was the Dangjin monitoring site (36.99° N , 126.62° E). The model top was set at 10,000 m, with vertical motion fields obtained from the meteorological input data. Trajectory outputs were recorded at 1 h intervals.

High $\text{PM}_{2.5}$ episodes were identified based on daily mean concentrations, with exceedances defined as days when concentrations exceeded $75 \mu\text{g}/\text{m}^3$, the threshold for issuing $\text{PM}_{2.5}$ advisories in Korea. These selected episodes were further assessed using wind rose plots and HYSPLIT simulations to differentiate between transboundary inflow and local emissions, in conjunction with evaluating changes in chemical composition and particle size distribution.

3. Results

3.1. Meteorological Data

Based on data from the Dangjin Automatic Weather System between August 2020 and March 2024, the average temperature was 12.9° C , with an average high temperature of 35.1° C and an average low temperature of -16.2° C . Cumulative precipitation totaled 1364 mm over the study period. The prevailing wind directions were southerly (39.76%) and northerly (28.65%), indicating that winds predominantly blew from the south or north.

Among the meteorological conditions facilitating air pollution episodes, calm conditions, which are defined as wind speeds of $\leq 0.5 \text{ m/s}$, accounted for 18.34% of the study period. However, this percentage indicates that wind-free periods were relatively limited, and stagnant air masses associated with high $\text{PM}_{2.5}$ days can prolong the residence time of transported particulates [19]. Further, calm conditions may facilitate increases in locally formed secondary particles, potentially increasing $\text{PM}_{2.5}$ particle concentrations [20].

3.2. $\text{PM}_{2.5}$ Seasonal Distribution and Chemical Components

Throughout the study, $\text{PM}_{2.5}$ levels exhibited significant variation, ranging from 4.74 to $95.31 \mu\text{g}/\text{m}^3$, with a mean value of $26.22 \pm 15.29 \mu\text{g}/\text{m}^3$. The daily variation of $\text{PM}_{2.5}$ concentrations is illustrated in Figure 2, showing distinct fluctuations corresponding to seasonal and meteorological influences. Seasonally, the average concentration in spring was $30.58 \pm 14.10 \mu\text{g}/\text{m}^3$, which decreased to $15.68 \mu\text{g}/\text{m}^3$ in summer, the lowest among the four seasons, before increasing to $22.11 \pm 13.56 \mu\text{g}/\text{m}^3$ in the fall. Winter had the

highest value at $30.83 \pm 16.31 \mu\text{g}/\text{m}^3$. These findings are consistent with those of previous studies [15,16], indicating relatively high winter concentrations owing to heating emissions, meteorological factors, and springtime increases, which are influenced by Asian dust events. Therefore, the seasonal $\text{PM}_{2.5}$ trends observed in Dangjin are consistent with the typical distribution patterns reported in other regions of Korea.

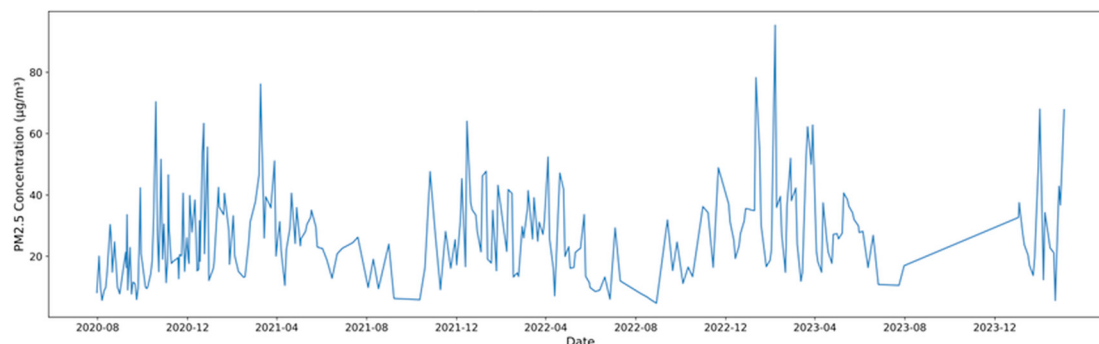


Figure 2. Daily variation of $\text{PM}_{2.5}$ mass concentration.

The mean NO_3^- concentration in $\text{PM}_{2.5}$ during the monitoring period was $5.69 \pm 7.02 \mu\text{g}/\text{m}^3$, accounting for the largest proportion at 29.7% of the total $\text{PM}_{2.5}$ mass; OC followed this at 28.3% and SO_4^{2-} and NH_4^+ each at 16.1%. The annual and seasonal average chemical compositions of $\text{PM}_{2.5}$ are presented in Figure 3. These three secondary inorganic ions collectively comprised more than half (61.9%) of the total $\text{PM}_{2.5}$. When the $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio is >1 , mobile emission sources such as vehicular exhaust are more influential, whereas ratios <1 indicate a considerable effect from stationary sources such as coal combustion [21]. Therefore, with a $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio of 1.84 in the study area, mobile emissions—particularly those from automobiles—appear to contribute more to $\text{PM}_{2.5}$ than from stationary sources. The relationship between the $\text{NO}_3^-/\text{SO}_4^{2-}$ and $\text{NH}_4^+/\text{SO}_4^{2-}$ ratios is illustrated in Figure 4. Additionally, the $\text{NH}_4^+/\text{SO}_4^{2-}$ ratio was consistently >1.5 across all seasons, indicating that under ammonia-rich atmospheric conditions in Dangjin, interactions with acidic substances may enhance $\text{PM}_{2.5}$ formation [22,23].

Carbonaceous compounds accounted for approximately 28.9% of the total $\text{PM}_{2.5}$ mass. This fraction introduces OC into the atmosphere through direct emissions and secondary formation driven by photochemical reactions involving volatile organic compounds (VOCs) [24]. In contrast, EC primarily results from combustion-related processes, including burning fossil fuels and biomass, and is a marker of primary emissions [25]. The interpretation of OC/EC ratios provides insights into emission sources: values <1 typically indicate coal combustion, whereas ratios >1 indicate substantial influence from secondary processes, including biomass burning [26–28]. An OC/EC ratio ≥ 2.0 is indicative of secondary organic carbon (SOC) formation via atmospheric photochemical transformations [29,30].

This study's OC/EC ratio was 8.72, indicating that secondary formation was more influential than primary sources; this may be attributed to the local characteristics of the sampling site. The site, which is situated near a major roadway, is considerably affected by vehicular emissions and is surrounded by numerous industrial facilities that emit combustion-related pollutants. Additionally, pollutants generated in adjacent regions can be carried by wind, contributing to increased contamination. These local factors likely promote the accumulation of primary pollutants and facilitate active photochemical reactions, thereby increasing the production of secondary OC.

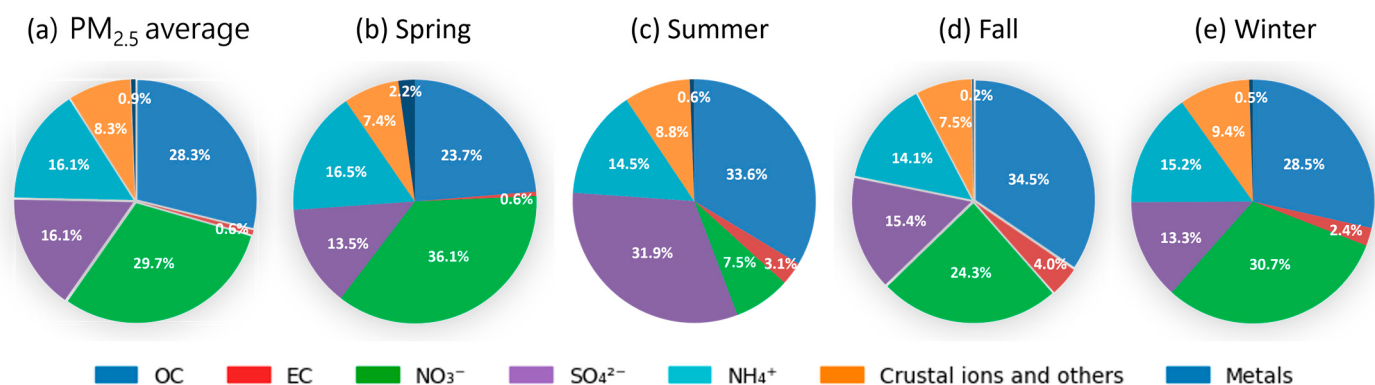


Figure 3. (a) Annual and seasonal (b–e) average chemical compositions of $PM_{2.5}$.

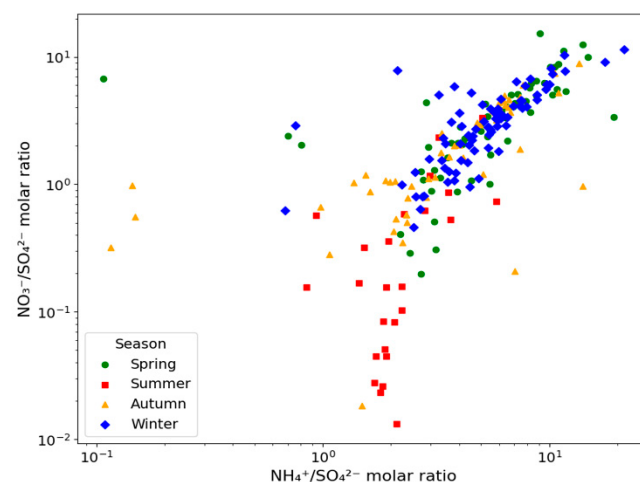


Figure 4. Nitrate-to-sulfate molar ratio as a function of ammonium-to-sulfate molar ratio in $PM_{2.5}$.

Although trace elements account for a relatively minor fraction of the total mass, numerous toxic metals within $PM_{2.5}$ can adversely affect human health even at low concentrations [31]. Therefore, continuous monitoring is crucial. This study measured the mass concentrations of 19 trace elements—S, Al, Si, Fe, Zn, Mn, Pb, Ti, Ba, P, As, Cu, Cd, Cr, Mo, Ni, V, Sr, and Co. Their combined contribution was under 10% of the total $PM_{2.5}$, with an annual average concentration of $1.4 \mu\text{g}/\text{m}^3$ (Table 1).

The correlation analysis of primary $PM_{2.5}$ components revealed strong inter-component correlations in all seasons except for summer (Table 2). In summer, the correlation coefficient between NO_3^- and SO_4^{2-} was -0.15 , indicating a negative relationship. This finding is consistent with a previous study [32], which revealed that when temperatures exceed 30°C , atmospheric NO_3^- exists primarily as gaseous HNO_3 . Owing to the increased solar radiation and temperatures in summer, NO_3^- remains in its gaseous form rather than transitioning to the particulate phase; consequently, the resulting inverse correlations among components differ from those in other seasons.

In contrast, in winter, NH_4^+ and NO_3^- exhibited the strongest correlation (0.94). As previously mentioned, NO_3^- concentrations were higher in winter than in other seasons, and secondary NO_3^- primarily exists as ammonium nitrate (NH_4NO_3).

Figure 5 shows the seasonal concentration distributions of $PM_{2.5}$ and all its chemical constituents. An analysis of each substance by season indicated that $PM_{2.5}$ concentrations were generally the highest in winter and lowest in summer. This outcome can be attributed to coal combustion for heating and stagnant atmospheric conditions in winter and, conversely, to high mixing heights and frequent precipitation events in summer, consistent with previous findings [33]. The seasonal order of $PM_{2.5}$ concentra-

tions was winter > spring > fall > summer, with winter exhibiting marginally higher levels than spring.

Table 1. Concentration of substances in the Dangjin area.

Substance (Unit)		N	Mean	S.D.	Max	<i>p</i> -Value ^a
PM _{2.5} (µg/m ³)		245	26.22	15.29	95.31	0.047
Ion (µg/m ³)	Cl [−]	245	0.66	0.68	4.91	0.000
	NO ₃ [−]	245	5.69	7.02	44.29	0.058
	SO ₄ ^{2−}	245	2.90	2.12	14.18	0.002
	Na ⁺	245	0.37	0.36	3.23	0.000
	NH ₄ ⁺	245	2.82	2.93	23.08	0.023
	K ⁺	209	0.27	0.27	3.00	0.000
	Mg ²⁺	127	0.11	0.08	1.00	0.078
	Ca ²⁺	245	0.13	0.13	0.80	0.000
Carbon (µg/m ³)	EC	280	0.64	0.29	2.09	0.060
	OC	280	5.58	2.95	16.62	0.448
Elements (ng/m ³)	Al	51	324.74	627.24	3680	0.027
	Ti	245	8.31	12.23	140.61	0.000
	V	245	1.10	1.22	6.48	0.000
	Mn	245	17.12	15.48	78.68	0.000
	Fe	245	193.80	186.70	1549.08	0.000
	Ni	245	1.54	1.91	20.09	0.000
	Co	245	1.01	1.04	8.22	0.000
	Cu	245	7.27	7.68	42.44	0.000
	Zn	245	62.24	51.21	234.06	0.000
	As	225	3.28	4.95	37.00	0.000
	Sr	245	0.65	1.02	6.33	0.000
	Mo	140	1.35	1.98	12.00	0.000
	Cd	245	1.82	3.34	22.09	0.000
	Ba	245	5.36	7.06	39.82	0.000
	Pb	245	23.31	49.52	621.90	0.003
	P	245	5.69	8.32	54.80	0.000
	S	245	928.19	962.45	4909.62	0.000
	Cr	245	2.43	2.72	22.85	0.000
	Si	245	302.29	563.26	6458.81	0.000

^a ANOVA by sampling years.

Table 2. Pearson correlation analysis of chemical components in PM_{2.5}.

Spring	NO ₃ [−]	SO ₄ ^{2−}	NH ₄ ⁺	OC	Summer	NO ₃ [−]	SO ₄ ^{2−}	NH ₄ ⁺	OC
SO ₄ ^{2−}	0.51				SO ₄ ^{2−}	−0.15			
NH ₄ ⁺	0.85	0.67			NH ₄ ⁺	0.15	0.92		
OC	0.55	0.34	0.62		OC	0.79	−0.07	0.19	
EC	0.47	0.26	0.46	0.78	EC	0.52	0.09	0.27	0.50
Fall	NO ₃ [−]	SO ₄ ^{2−}	NH ₄ ⁺	OC	Winter	NO ₃ [−]	SO ₄ ^{2−}	NH ₄ ⁺	OC
SO ₄ ^{2−}	0.64				SO ₄ ^{2−}	0.74			
NH ₄ ⁺	0.92	0.73			NH ₄ ⁺	0.94	0.76		
OC	0.62	0.31	0.54		OC	0.69	0.48	0.62	
EC	0.41	0.26	0.41	0.87	EC	0.32	0.12	0.26	0.72

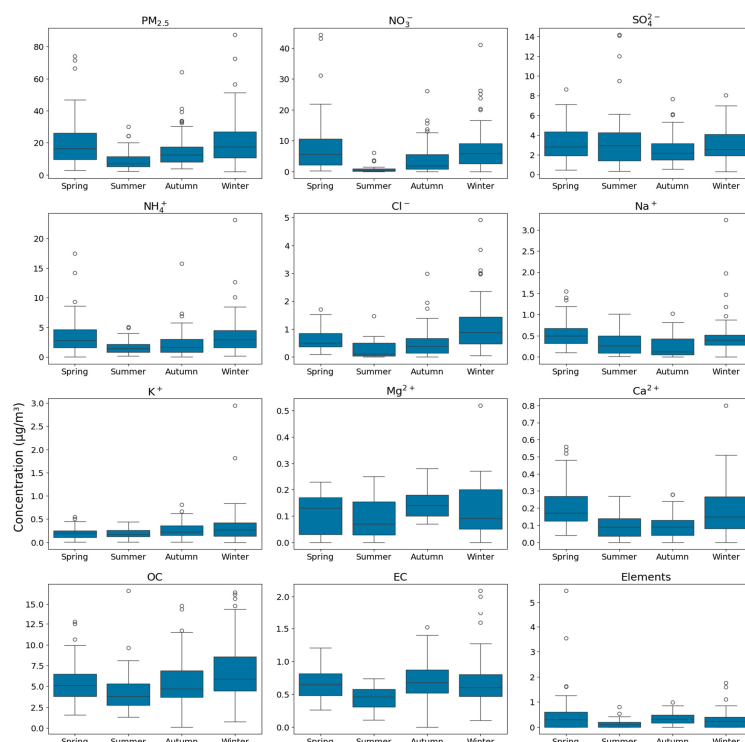


Figure 5. Seasonal variation of PM_{2.5} components.

Circles represent outliers beyond 1.5 times the interquartile range (IQR). Increased NO_3^- and NH_4^+ concentrations were observed in both spring and winter, possibly owing to low-temperature phase changes, long-range transport, and stagnant conditions [34]. In contrast, SO_4^{2-} was highest during summer, likely influenced by high temperatures, high humidity, intense solar radiation, and a deep mixing layer, all promoting SO_4^{2-} formation [35].

Cl^- averaged $1.07 \pm 0.87 \mu\text{g}/\text{m}^3$ in winter, exceeding levels in other seasons. Although coarse-mode Cl^- is typically associated with marine sources, in fine-mode particles, increased Cl^- levels typically result from industrial processes, waste incineration, and coal combustion [36]. Consequently, coal combustion for heating has likely played a notable role in winter. K^+ and Mg^{2+} concentrations exhibited similar seasonal patterns, remaining relatively low from spring to summer and slightly increasing in the fall. This pattern may be attributed to the lower total PM_{2.5} mass in the fall, which proportionally increases the K^+ and Mg^{2+} concentrations. These ions commonly originate from marine sources, soil, and biomass burning [37].

Ca^{2+} and Na^+ concentrations are influenced by soil erosion, resuspension, and marine sources, and are typically found in high quantities in coarse particles [38]. However, Ca^{2+} primarily originates from the soil, whereas Na^+ predominantly results from marine sources. In spring, strong winds and stagnant air possibly contributed to both ions becoming essential components of PM_{2.5}. Spring had the highest proportion of heavy metals, likely due to Asian dust events transporting soil from deserts in China and Mongolia via prevailing westerly winds [39].

3.3. Characteristics of High-Concentration PM_{2.5} Components

This study defines high-concentration events as days during which the PM_{2.5} advisory threshold ($75 \mu\text{g}/\text{m}^3$) is exceeded consecutively for at least 24 h. The temporal variations in daily average PM_{2.5} are shown in Figure 6, highlighting the three identified high-concentration events. Three such high-concentration events were identified and analyzed. To investigate changes in major components during each event, the mean values

for the three high-concentration days were compared with those for the seasonal averages, excluding the event days. Among these events, PM_{2.5} concentrations were the highest in Event 3 (95.31 µg/m³), followed by Events 2 (78.23 µg/m³) and 1 (76.14 µg/m³). Notably, concentrations of ammonium (NH₄⁺), nitrate (NO₃[−]), sulfate (SO₄^{2−}), and OC significantly increased during these high PM_{2.5} episodes, indicating that these components were key contributors.

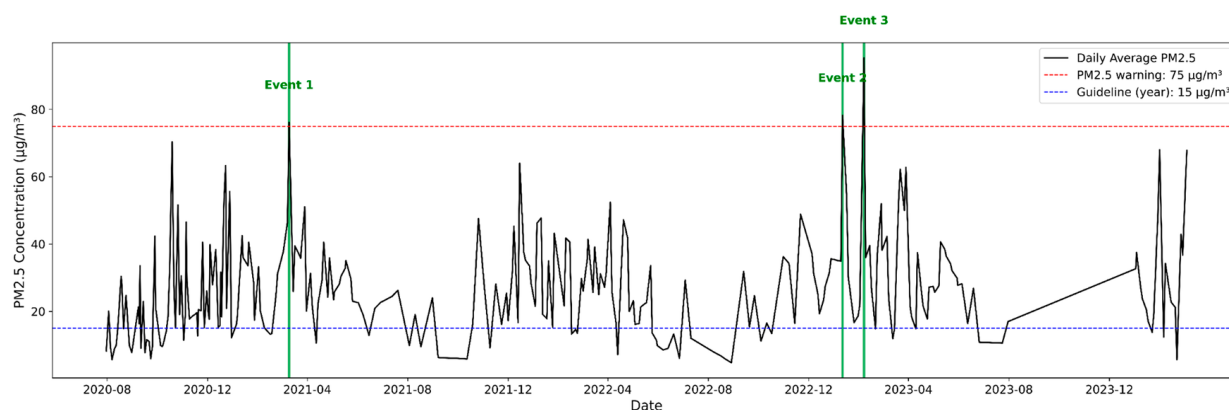


Figure 6. Temporal variations in daily average PM_{2.5}.

The percentage changes of major components during each high-concentration event are shown in Figure 7. Ammonium (NH₄⁺) exhibited the largest increase relative to its seasonal average during these high-concentration periods: approximately 4.6-, 3.1-, and 6.4-fold increases in Events 1, 2, and 3, respectively. Ammonium typically combines with nitrate (NO₃[−]) and sulfate (SO₄^{2−}) to form ammonium nitrate (NH₄NO₃) and ammonium sulfate ((NH₄)₂SO₄), respectively [40]. Consequently, it plays a major role as a secondary component in high-concentration PM_{2.5}. In Event 3, which occurred in winter, the substantially high ammonium concentration further accentuated its linkage to nitrate.

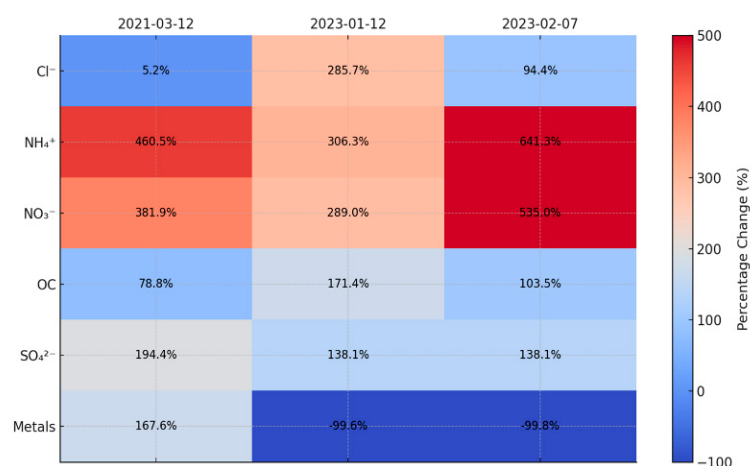


Figure 7. Change Rates of Major Components in High-Concentration PM_{2.5}.

Nitrate (NO₃[−]) concentrations increased by approximately 3.8-fold in Event 1, 2.9-fold in Event 2, and 5.3-fold in Event 3 relative to seasonal averages. Nitrate is a secondary pollutant formed when nitrogen oxides (NO_x) undergo oxidation reactions in the atmosphere, originating mainly from mobile sources, particularly vehicle exhaust [41]. The exceptionally high increase during Event 3 possibly indicates a combination of heating (during winter) and vehicular emissions. Moreover, the low mixing-layer height and stagnant conditions in winter likely facilitated nitrate accumulation, thereby amplifying ambient concentrations.

Sulfate (SO_4^{2-}) increased by approximately 2-fold in Event 1 and 1.4-fold in Events 2 and 3 compared to seasonal averages, primarily associated with stationary sources, such as coal combustion [42]. Sulfate is formed secondarily through oxidation reactions of sulfur dioxide (SO_2) in the atmosphere. Emissions from these stationary sources can increase ambient sulfate concentrations.

OC levels in high-concentration events increased by approximately 0.8-, 1.7-, and 1.0-fold in Events 1, 2, and 3, respectively, compared to seasonal averages. OC is partly formed via the photochemical oxidation of VOCs in the atmosphere and is vital in high $\text{PM}_{2.5}$ episodes [43]. This study's OC/EC ratio was 8.72, indicating that SOC contributes substantially to the total OC, exceeding primary emissions. Low mixing-layer heights and stagnant conditions likely promoted VOC oxidation reactions, further enhancing SOC formation. Additionally, interactions with other components, such as nitrate (NO_3^-) and sulfate (SO_4^{2-}), may have contributed to the formation of complex aerosols during these episodes.

4. Causes of High-Concentration Occurrences

Wind direction and wind speed data are important meteorological factors influencing variations in fine particulate matter concentrations. In particular, the effect on surrounding areas can vary considerably depending on the daily changes in prevailing wind direction [44,45]. This study used wind roses based on wind direction and wind speed data from Dangjin City to examine the influence of surrounding emission sources on fine particle concentrations. Additionally, the prevailing wind direction in each case and the physical and chemical changes of $\text{PM}_{2.5}$ were investigated. Figure 8 shows the wind roses, backward trajectory analysis, and physical and chemical changes for three high-concentration $\text{PM}_{2.5}$ cases measured in Dangjin during the study period.

Case 1 occurred over two consecutive days, from 10 to 12 March 2021, during the spring season. The $\text{PM}_{2.5}$ concentration was $76.14 \mu\text{g}/\text{m}^3$, with a prevailing northerly wind. The average wind speed was 1.19 m/s, and the calm condition ratio was 35.42%, indicating the highest wind speed and lowest calm condition ratio among the high-concentration cases. The backward trajectory analysis indicated that air parcels likely originated from the northern continental regions of East Asia, gradually ascended to higher altitudes, and subsequently entered Korea. All air masses in the upper and lower layers were transported from northwestern continental areas, contributing to the observed influence. The chemical composition was as follows: NO_3^- (42.4%), NH_4^+ (23.8%), OC (14.5%), SO_4^{2-} (11.8%), metals (4.1%), and EC (1.3%). During this springtime high-concentration case, SO_4^{2-} was relatively high owing to an increase in SO_2 , a precursor to SO_4^{2-} . The increase in SO_4^{2-} during spring high-concentration periods may be due to the high levels of SO_2 (a gaseous precursor) and more active gas-phase reactions under lower relative humidity than those of low-concentration periods in spring. Therefore, this springtime high-concentration event is likely associated with long-range transported air masses from the northern continental regions, as suggested by the backward trajectory analysis.

Case 2 occurred in winter, from 11 to 12 January 2023. The $\text{PM}_{2.5}$ concentration was $78.23 \mu\text{g}/\text{m}^3$, with a prevailing northerly wind and some observed northwesterly winds. The average wind speed was 0.68 m/s, and the calm condition ratio was 58.8%, the highest among the cases. The significant increase in the concentrations of NO_3^- and NH_4^+ in the atmosphere was primarily due to their long residence time, a characteristic pattern particularly associated with winter heating fuel usage. The backward trajectory analysis showed that air masses during this period mainly originated from northern and northeastern continental regions and moved toward Korea, where local stagnation further contributed to the high-concentration event. The chemical composition was as follows:

NO_3^- (34.6%), OC (22.3%), NH_4^+ (17.4%), SO_4^{2-} (9.6%), and EC (1.5%). Compared to the normal periods, NO_3^- and NH_4^+ concentrations increased by approximately 1.2 and 1.18, respectively, whereas Cl^- concentrations increased by a factor of 5.5, which was a notable increase. During this period, heating activities involving coal combustion and chlorides emitted from metal processing and manufacturing processes in the Asan National Industrial Complex were analyzed as the primary causes for increased Cl^- levels. Therefore, the combined influence of long-range transported air masses and domestic industrial activities likely contributed to this high-concentration event.

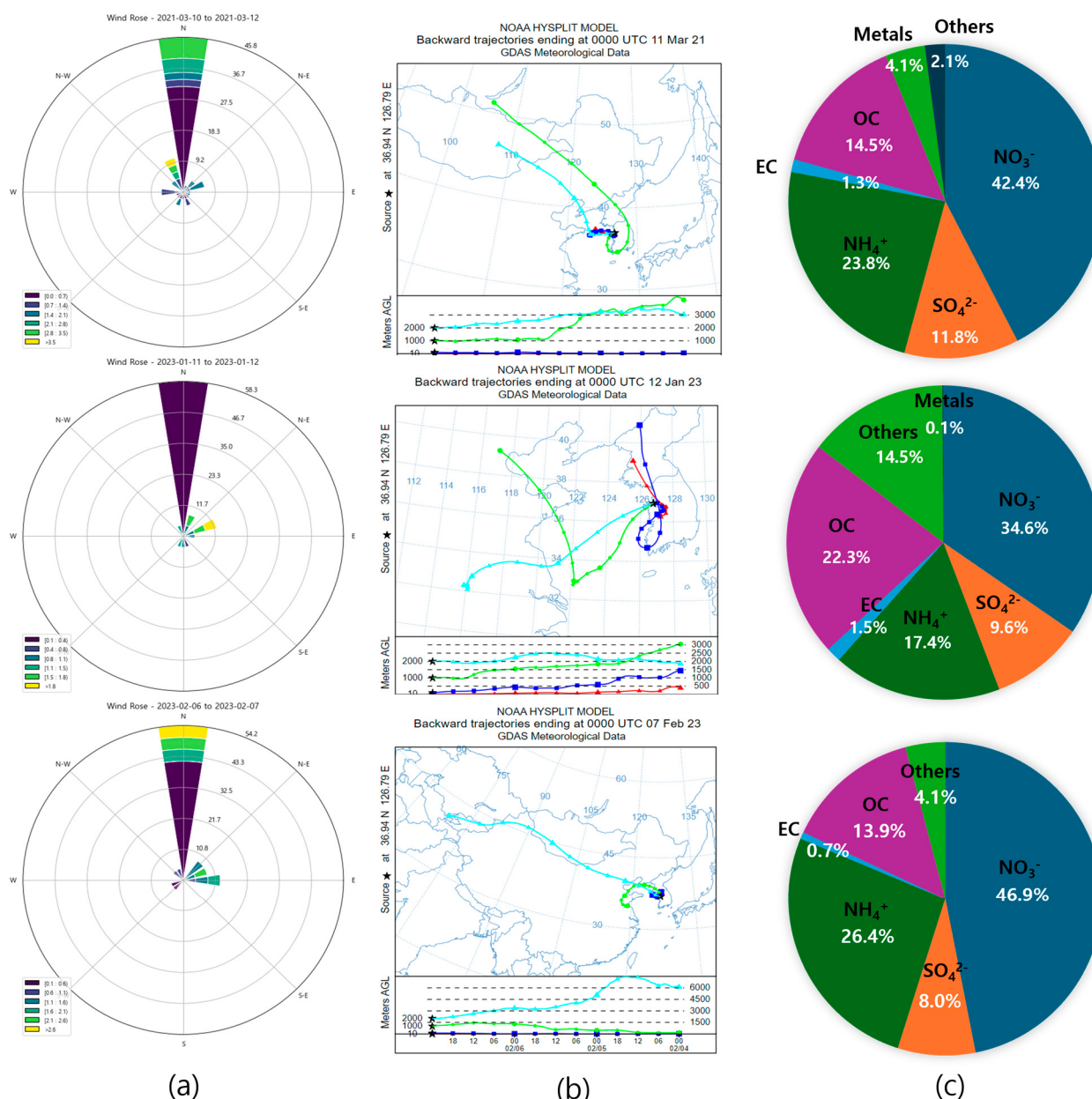


Figure 8. (a) Windrose; (b) Back-trajectory; (c) Chemical composition ratio. In (b), colored lines denote air mass trajectories at various arrival heights, and the black star marks the Dangjin monitoring site.

Case 3 also occurred during winter, from 6 to 7 February 2023, with a $\text{PM}_{2.5}$ concentration of $95.31 \mu\text{g}/\text{m}^3$. The prevailing wind was northerly, and some easterly winds were observed. The average wind speed was 0.92 m/s , and the calm condition ratio was 50.0%, a level that can facilitate local airflow stagnation. The backward trajectory analysis showed that air masses mainly originated from eastern continental regions of East Asia and moved

along the west coast before reaching Korea, contributing to the high-concentration event. The chemical composition analysis indicated the following concentrations: NO_3^- (46.9%), NH_4^+ (26.4%), OC (13.9%), SO_4^{2-} (8.0%), and EC (0.8%). The notable increase in NO_3^- can be attributed to low temperatures and high humidity during winter, which promote the partitioning of nitrates into the aerosol phase and enhance heterogeneous reactions under high humidity conditions. The long-range transport of continental air masses and local stagnation were important factors in increasing nitrate levels. These combined factors resulted in the formation of the high-concentration $\text{PM}_{2.5}$ event in Case 3.

The backward trajectory analyses in Cases 1–3 were based on the HYSPLIT model, which provides simulated transport pathways. Actual transport may be more complex, and observational validation would strengthen these results. As backward trajectories primarily indicate the pathways of air masses rather than direct source regions, their interpretation should be made with caution, particularly in cases where air masses may have passed through regions with lower pollutant concentrations that could dilute the observed levels. Therefore, the results of this study are interpreted as supportive evidence, and more precise source apportionment methods, such as the Weighted Potential Source Contribution Function (WPSCF), are suggested for future applications.

5. Conclusions

This study analyzed $\text{PM}_{2.5}$ concentrations and chemical composition in the Dangjin region from August 2020 to March 2024. The average $\text{PM}_{2.5}$ concentration was $26.22 \pm 15.29 \mu\text{g}/\text{m}^3$, with the lowest seasonal mean in summer ($15.68 \mu\text{g}/\text{m}^3$) and the highest in winter ($30.83 \pm 16.31 \mu\text{g}/\text{m}^3$).

Among chemical components, NO_3^- was the dominant contributor ($5.69 \pm 7.02 \mu\text{g}/\text{m}^3$, 29.7% of total $\text{PM}_{2.5}$), followed by OC (28.3%), SO_4^{2-} (15.3%), and NH_4^+ (15.3%), with secondary inorganic ions together accounting for 60.3% of the mass. The $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio (1.94) suggested a stronger influence from mobile sources, while the OC/EC ratio (8.72) indicated a substantial contribution from secondary organic carbon.

During high-concentration events ($>75 \mu\text{g}/\text{m}^3$), levels of NO_3^- , NH_4^+ , SO_4^{2-} , and OC increased significantly. NH_4^+ showed the greatest enhancement, up to 6.4-fold in Case 3 (winter), while NO_3^- increased up to 5.3-fold. These events coincided with stagnant meteorological conditions, reduced mixing-layer heights, and enhanced wintertime fuel combustion.

The backward trajectory analysis revealed that high-concentration events were driven by both long-range transported air masses and domestic emissions. In Case 1 (spring), air masses from the northern continental regions were associated with elevated $\text{PM}_{2.5}$ levels. Case 2 (winter) reflected the combined effects of domestic heating and industrial activities under stagnant conditions, together with regional-scale inflow from the northeast. In Case 3 (winter), air masses from eastern continental regions and local stagnation contributed to the most severe pollution episode.

Unlike previous studies that focused on short-term observations or limited chemical species, this work provides multi-year data with detailed chemical composition, offering new insights into the combined effects of domestic emissions and transboundary transport.

This study enhances understanding of the seasonal characteristics of $\text{PM}_{2.5}$ and the interrelationships among its primary components, elucidating the roles of mobile and stationary sources during high-concentration episodes. In addition, by examining the effect of seasonal meteorological conditions on $\text{PM}_{2.5}$ levels, this study underscores the need for airflow analysis and careful consideration of emission sources when formulating air pollution management strategies and policies. As major chemical components of fine particulate matter (ions, organic carbon, and trace elements) can present potential

health risks, systematic measures to reduce exposure during high-concentration periods are essential. In particular, the observed concentrations in this study frequently exceeded the World Health Organization (WHO) air quality guidelines, indicating potential risks for adverse health outcomes. A preliminary risk consideration highlights that sustained exposure to elevated levels of nitrates, sulfates, and carbonaceous species may increase respiratory and cardiovascular disease burden in the affected population. Enhancing the management of stationary emission sources (industrial complexes) and implementing complementary policies to reduce emissions from mobile sources (vehicle exhaust) are expected to effectively mitigate high-concentration events. Future research should include more precise source apportionment and long-range transport modeling to better elucidate the contributions of pollutants at the regional level.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/toxics13100869/s1>. Table S1: Specifications of the measurement device used in this study; Table S2: Ion chromatography analysis equipment and conditions; Table S3: Flame ionization detector analysis equipment and conditions; Table S4: Energy dispersive X-ray fluorescence analyzer and conditions.

Author Contributions: C.-M.L. and Y.-h.K.; methodology, C.-M.L., Y.-h.K. and H.J.; investigation, Y.-h.K., S.-Y.P., H.J. and J.-E.M.; data curation, C.-M.L., Y.-h.K. and J.-E.M.; data collection and analysis, C.-M.L., Y.-h.K. and S.-Y.P.; writing—original draft preparation, Y.-h.K.; writing—review and editing, C.-M.L., S.-Y.P. and Y.-h.K.; visualization, Y.-h.K.; supervision, C.-M.L.; project administration, C.-M.L. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Most of the data generated or analyzed during this study are included in this published article and Supplementary Materials. Additional datasets are available from the corresponding author upon reasonable request.

Acknowledgments: The authors acknowledge the contributions of all individuals who assisted with field investigations and data processing.

Conflicts of Interest: The authors declare no conflicts of interest.

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