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Tracing the Source of Particulate Matter in the Atmosphere over Seoul using Isotope Analysis

Ingyu LEE[1]

[1. First Author] Assistant Professor, Department of Civil & Energy System Engineering, Kyonggi University, Korea.
Email: ilee@kyonggi.ac.kr

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Abstract

Purpose: Particulate matter (PM) has been reported to affect human health and disease, as well as a wide range of industries. Several studies have shown its impact on the distribution industry affecting consumption patterns and sales. Various techniques have been used to identify the sources of these PMs. Stable isotopes ratio (SIR) has been applied in various fields including chemistry, ecology, environment, and food. Especially in environmental science fields, they have been used to track sources of water or air pollution. This study was carried out to examine the origin/source of PM in the atmosphere over Seoul Metropolitan. **Research design, data and methodology:** PM samples were collected from five different locations in Seoul, and stable isotope ratios of carbon, nitrogen, sulfur, and nitrate in PM10 were analyzed using an elemental analyzer-isotope ratio mass spectrometer. **Results:** These isotope values were compared with the data reported in the literature. The isotope ratio calculated in this study for each target element was within the range of reported values. **Conclusions:** The results revealed that δ13C and δ15N values obtained in this study were very similar to those of PMs collected in Paris. Based on the results, we hypothesized that a major emission source of PMs in Seoul might be combustion of liquidized natural gas. In comparison to other studies conducted during periods of high PM concentrations, the observed results are consistent with the hypothesis that pollution is primarily domestic stagnation in transportation and heating activities.

Keywords : Stable isotope ratio, Particulate matter, Emission source, Seoul.

JEL Classification Code : Q40, Q42, Q53, Q54, Q56

1. Introduction

In Korea, research on the health effects of particulate matter (PM) has been conducted since the 1990s. This research has included studies on the impact of fine and ultrafine PM on human health, which have been undertaken domestically and internationally (Bae, 2014; Kappos et al., 2014). PM has been reported to affect human health and disease, as well as a wide range of industries. Several studies have shown its impact on the distribution industry in terms of consumption patterns and sales (Bae & Baek, 2020; Han & Moon, 2022; Kim et al., 2022). Particulate matter in ambient air has been identified as impacting distribution and service industries. In particular, the distribution industry is susceptible to changes in the external environment, with consumers' purchasing behavior significantly influenced by such an atmosphere condition (Han & Moon, 2020). Consequently, this study aims to ascertain the extent to

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which domestic and foreign sources contribute to the pollution of particulate matter, which affects various industries.

Therefore, we focused on the origin of PMs using a technique that is comparing stable isotope ratio (SIR). Analysis of SIRs of elements has been applied in a variety of science fields including chemistry, ecology, environment, and food (Glibert et al. 2019; Ishikawa 2018; Santana-Mayor et al. 2019). Nowadays, it is used to investigate sources of water or air pollution (Guo et al. 2019; Schudel et al. 2018; Sriram et al. 2018). Especially, stable isotopic analysis of atmospheric particulate matter (PMs) is considered as a powerful tool for identifying emission sources for air pollution (Lim et al. 2019; Ouyang et al. 2019; Pan et al. 2020).

Widory et al. (2004) initiated research on SIRs to track potential sources of pollution in the atmosphere. According to the study, it appears to be difficult for us to identify pollution sources with common physicochemical properties of the detected pollutants. As such, they applied carbon and lead isotopes for the identification of aerosol sources in the atmosphere over Paris. Morera-Gómez et al. (2018) analyzed SIRs of carbon and nitrogen in particulate matter (PMs) emitted from various sources. The SIR results were compared with those obtained from the potential emission sources for tracking the major contributors of the local air pollution. PM_{10} (PMs with an aerodynamic diameter of less than 10 μm) is one of the major air pollutants in urban areas. It contains numerous inorganic and organic compounds, many of which can adversely affect human health and the environment (Fuzzi et al. 2015; Hegde et al. 2016). Widory (2007) conducted research in the downtown of Paris, France, where pollution sources were investigated by analyzing nitrogen SIRs in PM_{10} samples and by comparing them with those obtained from the corresponding library established for various emission sources such as vehicle exhaust, fuel combustion, natural gas, etc. The study revealed that most important nitrogen emission sources in Paris would be combustion of natural gas. However, due to the complex mechanism of PM_{10} formation, behavior of the particles in the atmosphere, and the seasonal variation in fuel consumption, it is difficult to clearly identify pollution sources by using only carbon and nitrogen SIRs (Morera-Gómez et al. 2018). In addition, SIRs of carbon and nitrogen could be changed by a variety of phenomena such as raining events, temperature changes, and pollution sources (Felix & Elliott 2014).

Therefore, stable isotopes of various elements as they are or as a part of a molecule are being utilized to characterize emission sources. NO_3^- and SO_4^{2-} are mainly created during the conversion process from gaseous N and S compounds to aerosol particles by secondary atmospheric chemical reactions (Yang et al. 2015). Agricultural activities are known as the major NH_4^+ source. A Significant amount of NH_4^+ is also emitted in urban area. It is emitted by vehicles equipped with a catalytic converter and eventually forms PMs as combined with other sulfur or nitrogen ions (Felix et al. 2014). To better understand the emission source and atmospheric behavior of PMs, analysis of a variety of isotope composition may be required for water-soluble inorganic ions in PMs (Felix et al. 2013; Pan et al. 2016; Sharma et al. 2015). Thus, various research on tracking pollution sources makes use of isotope ratios of NH_4^+, NO_3^-, SO_4^{2-} ions (Felix et al. 2014; Park et al. 2018; Yang et al. 2015). For example, Felix & Elliott (2014) characterized each air pollution source by comparing SIRs of NO_X-$\delta^{15}N/\delta^{18}O$ originated from organic, vehicle, soil and livestock samples. Stable isotope ratios of NO_X-$\delta^{15}N/\delta^{18}O$ of the samples collected from the northeastern region of the United States were compared with the SIR values of pollution sources listed in the library made in the States. The study concluded that vehicles were the main source of air pollution in the region. By analyzing time change of NO_3^--$\delta^{15}N$, Beyn et al. (2014) could trace down the shift of the pollution sources in Geesthacht, Germany over time; they monitored nitrogen deposits for four seasons. Mukai et al. (2001) measured sulfur and lead SIRs of the atmospheric aerosol of urban areas in China and Japan. They analyzed the regional sources characteristics by comparing sulfur SIRs with different fuel sources. Coal combustion was identified as the most significant source of air pollution in the industrial areas of the country by analyzing sulfur SIR. A summary of a variety of research was compared with the type of isotope ratio and tracing emission sources in Table 1.

Table 1: Isotope analysis with tracing emission sources in literature

City, Country	Isotope ratio	Emission Sources	Reference
Paris, France	C, N, Pb	Road, Industrial Park, Fuel, Natural gas	Widory et al., 2004; Widory, 2007
Northeastern, USA	N, O	Fertilized soil, Vehicle, Livestock manure	Felix & Elliott 2014
Geesthacht, Germany	NO_x	Coal powerplant, Road, Vehicle	Beyn et al., 2014
Cities in China and Japan	S, Pb	A various type of coal	Mukai et al., 2001

In Korea, only few studies have been carried out to characterize atmospheric particulate aerosols or PMs through the analysis of SIRs of elements (Hong et al. 2019; Park et al. 2018). Identifying the composition and sources of PMs is important in establishing a management program or policy for controlling air pollution. Therefore, the objective of present study is to characterize of PM_{10} by measuring SIRs of carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$), sulfur ($\delta^{34}S$), and nitrate (NO_3^--$\delta^{15}N/^{18}O$) in particles collected in the air over Seoul, Korea. Characterization of isotopic composition has been performed to track the potential sources of the collected PM_{10} by comparing the SIR values from this study with the ones published by other researchers. In addition, develop a response strategy and basic information for the distribution industry by analyzing the industrial impacts related to PMs.

2. Research Method

2.1. PM10 sampling location

Samples were collected from five different locations in Seoul. There were two sampling events during the winter on 8th Jan and 11th Mar. There are air quality monitoring stations operated by the Seoul Research Institute of Public Health and Environment for Seoul metropolitan government in the five selected locations shown in Fig. 1. The sampling site in Guro (1) is close to a semi-industrial area while in the north, there is a digital complex and southern circular roads. Mapo (2) site is a shopping district, while Gwangjin (3) site is a residential area. Songpa (4) is in a park. Yangjae (5) site is close to a residence area and to a highway. The characteristics of sample locations were presented in Table 2 for description on sampling sites.

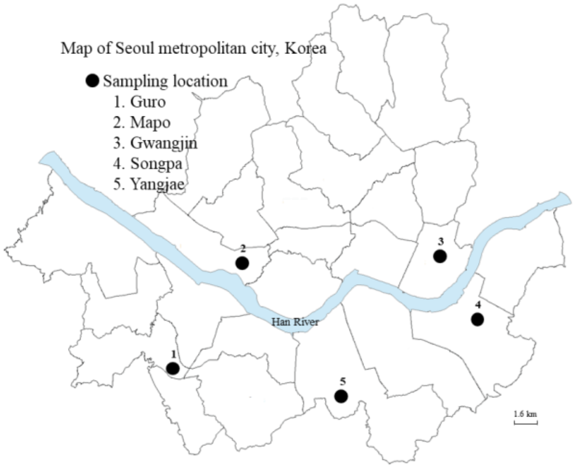

Figure 1: Sampling locations (Map of Seoul, Korea)

Guro (1) and Mapo (2) were designated as urban areas due to their high population density and geographical characteristics for applying urban location. Gwangjin (3), Songpa (4), and Yangjae (5), which are located in the suburbs of Seoul, were designated as peripheral areas. Guro site is 40 meters above sea level at Guro High School. This area is characterized by a combination of industrial and semi-industrial activities, with the Digital Complex situated to the north of the measurement site and the Southern Belt Highway located to the south. Mapo measurement site is situated at an elevation of 17 meters above sea level. This measurement site is located at the Mapo Art Centre, represented by low-rise buildings, including shops and residential properties. Gwangjin site, located at the former Arisu Water Treatment Plant, is 35 meters above sea level. This area is known for its high-rise apartments in the southern region, surrounded by apartment and office complexes. Songpa site, situated in the Olympic Park at 16 meters above sea level, is a predominantly residential area. Yangjae site is in the Olympic Park, 33 meters above sea level, and the Seoul Institute of Health and Environment. It is predominantly residential, nearby expressway and forest.

Table 1: Characteristics of sampling sites

ID	Name	Characteristic	Elevation (m)	PM_{10} Concentration ($\mu g\ m^{-3}$).
1	Guro	Heavy traffic, Commercial	40	67, 39
2	Mapo	Commercial, Residential	17	70, 41
3	Gwangjin	Residential	35	83, 44
4	Songpa	Residential, Suburban	16	69, 39
5	Yangjae	Heavy traffic, Residential	33	78, 43

2.2. PM_{10} Collection and Analytical Methods

PM_{10} in the air was collected using a high-volume air sampler (HVAS, Sibata, Kyoto, Japan) with a quartz fiber filter (Toyo Roshi Kaisha, Tokyo, Japan) at a flow rate of 1 $m^3\ min^{-1}$ for 24 h; the amount of PM_{10} on the filter was quantified using the gravimetric method. Ionic compounds in the collected particles were extracted through the method proposed by Kouvarakis et al. (2001). The samples were prepared for the size of Φ 50 mm using a punch with the collected filter, followed by crushing the filter for extraction of nitrate ionic compound and isotope analysis. These samples added 40 mL of deionized water in a conical tube and particulate nitrate extracted in a solution for 30 minutes using an ultrasonic digester. It was passed through a 0.20 µm membrane filter (Whatman®, Maidstone, UK) after shaking for 5 minutes. Then, nitrate (NO_3^-) concentration was measured using an ion chromatograph (DX-100, Dionex, California, USA).

2.3. Isotopic Analysis

PM_{10} samples were sealed in tin boats (4 mm×4 mm ×11 mm) and combusted in an elemental analyzer (EA) with 1150 ˚C oxidation and 850 ˚C reduction tubes. The combustion gas was composed of high-purity oxygen (>99.995%) and ultra-high-purity helium gas (>99.9999%). The carrier gas was helium (>99.9999%) with a 180 mL/min flow rate. The samples were gasified with nitrogen, carbon dioxide, and sulfur dioxide and simultaneously measured in an isotope ratio mass spectrometer (IRMS). SIRs are measured using a mass spectrometer (MS). This is an instrument that ionized gas based on the mass-to-charge ratio (m/z) of the target substance and passes it through a magnetic field to measure the SIR (Dawson & Brooks, 2001). SIR was analyzed using an EA-IRMS (Isoprime, Elementar-GV Instrument, Manchester, UK). it analyzed $^{13}C/^{12}C$, $^{15}N/^{14}N$, and $^{34}S/^{32}S$ values of PMs in the collected samples. Reproducibility of standard materials was obtained within about ±0.2 ‰ for both of carbon, nitrogen, and sulfur isotope ratio.

NO_3^--N^{15}/O^{18} was also analyzed using a denitrifying method; *Pseudomonas chlororaphis* (*P. chlororaphis*) was utilized to reduce NO_3^- to N_2O (Sigman et al. 2001). The solid type of P. chlororaphis was cultivated in tryptic soy broth (TSB) at 28 ℃ and for 3 d. After centrifugation of the cultures, only the cells were separated and cultured at 28 ℃ and for 7 d in a 100 mL TSB medium. The concentrated cells were placed into a tube. Then, the tube was purged with helium gas. The tube was sealed with a stopper and added with a sample containing NO_3^- extracted from the PM_{10}. NO_3^- in the sample were converted N_2O gas in the tube for 2 h. The N_2O gas generated was condensed with a trace gas pre-concentrator connected with a CO_2 cryo-trap. NO_3^--N^{15}/O^{18} in the gas was measured by IRMS (Isoprime, Elementar-GV Instrument, Manchester, UK). The stable isotope ratios of oxygen and nitrogen in the sample were adjusted with Vienna standard mean ocean water (VSMOW) for $^{18}O/^{16}O$, and with atmospheric nitrogen for $^{15}N/^{14}N$, respectively. The calculation of stable isotope consumption is always compared to a standard and the relative difference is expressed as a del value (δ) as follows.

$$\delta(‰) = \left(R_{sample}/R_{standard} - 1\right) \times 1000$$

A δ value of 0 means that the isotope ratio originated the sample (R_{sample}) is equal to that calculated by the standard ($R_{standard}$). A δ value of + (positive) means that the sample is more enriched in heavy isotopes than the standard, and a value of - (negative) means that the sample is deficient in heavy isotopes and lighter than the standard (McKinney et al., 1950).

3. Results and Discussion

3.1. SIRs of C, N, S in PM10 collected in Seoul

The results revealed that the stable isotope of carbon ($\delta^{13}C$) in PM_{10} collected from the air over Seoul were -25.5 to -24.2‰ (Fig. 2). The ratio of the C isotopes was similar to the PMs measured in the cities of northern China and central Mexico City (Cao et al. 2011; Górka et al. 2014; Grassi et al. 2006; López-Veneroni 2009). Conventionally, processes combusting petrol and natural gas have been considered as major PM emission sources (Widory et al. 2004). However, the stable isotope of carbon in PMs of the air over Seoul was similar to that of natural gas which is used as fuel for automobiles and home-heating. Compared to the isotope ratio values measured in an agricultural area of Tanzania, the area measured in this is quite different. This is probably because biomass-burning is commonly practiced in African countries (Mkoma et al. 2014).

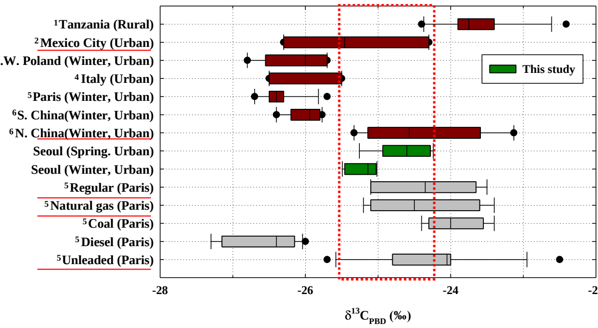

Figure 2: Composition of carbon isotope ratio in PM_{10}. Data was compared with ([1]Mkoma et al., 2014; [2]López-Veneroni, 2009; [3]Górka et al., 2012; [4]Grassi et al., 2006; [5]Widory et al., 2004; [6]Cao et al., 2011).

The isotopes of $\delta^{15}N$ in PM_{10} collected in this study ranged from -12.1 to 7.1‰ as shown in Fig. 3, The same range of isotope ratios of δ15N were measured in Paris, France and the southwestern region of Poland. Like the stable isotope ratio of carbon, the nitrogen isotope ratio was close to the one of natural gas (Górka et al. 2014; Kundu et al. 2010; Mkoma et al. 2014; Widory 2007). In fact, natural gas is used as fuel for public transportation and heating both in Korea and France. Notably, the nitrogen isotope ratio measured in Tanzania is significantly different from those of Seoul and Paris, possibly because air nitrogen source of the Tanzanian rural area would be different from those of Seoul and Paris.

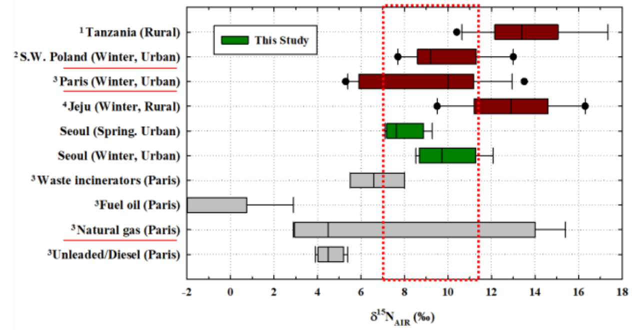

Figure 3: Composition of nitrogen isotope ratio in PM10. Data was compared with ([1]Mkoma et al., 2014; [2]Górka et al., 2012; [3]Widory, 2007; [4]Kundu et al., 2010).

In the case of $\delta^{34}S$, the range of sulfur isotope ratios was 5 to 9‰ (Fig. 4). $\delta^{34}S$ measured in the northern area of China ranged from 4 to 6‰ and that of Mainz, Germany was 2 to 14‰ (Mukai et al. 2001; Sinha et al. 2008). The stable isotope ratios of $\delta^{34}S$ measured in this study were quite different from those of gas emission from combustion as a fuel of coal in China (Cao et al., 2011). Germany and Korea are less affected by emissions from the combustion of a coal since these countries mainly use natural gas for heating.

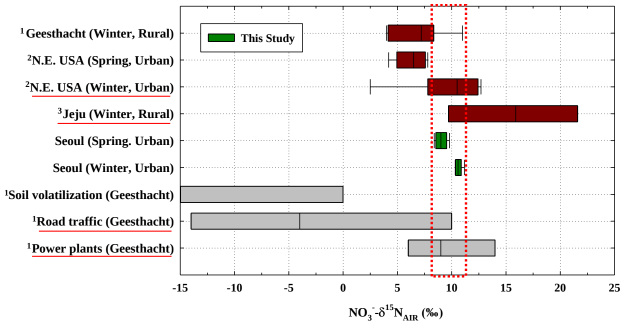

Figure 4: Composition of sulfur isotope ratio in PM10. Data was compared with (1Mukai et al., 2001; 2Sinha et al., 2008).

In this study, N and O stable isotopes of atmosphere-derived nitrate (NO_3^--$\delta^{15}N/\delta^{18}O$) were analyzed to identify potential sources of nitrogen oxides (NO_x) emission to the air pollution (Redling et al. 2013; Walters et al. 2015). In Seoul, NO_3^--$\delta^{15}N$ was 8-12 ‰ (Fig. 5) where it corresponded to NO_3^--$\delta^{15}N$ from the emission source of power plants and vehicles (Kundu et al., 2010; Beyn et al., 2014). It was distributed in similar ranges of isotope composition analyzed in urban areas of New York and Ohio in eastern United States (Elliott et al. 2009). Similar to the result of carbon and nitrogen stable isotope composition contributing to the emission source in atmosphere aerosol, it is assumed that nitrate isotopic composition from the PMs originated mainly from vehicles.

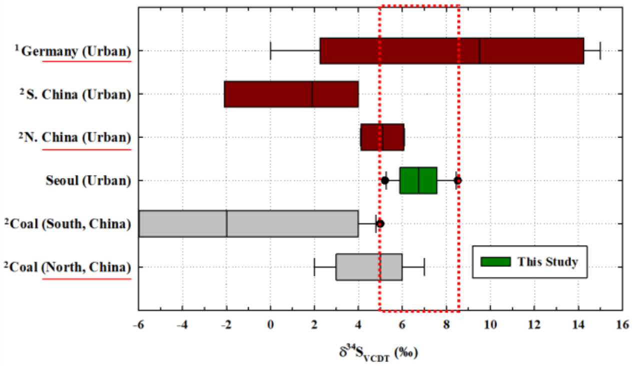

Figure 5: Composition of nitrogen isotope ratio of NO_3^- in PM_{10}. Data was compared from ([1]Beyn et al., 2014; [2]Elliott et al., 2009; [3]Kundu et al., 2010).

3.2. SIRs of NO_3^- in PM_{10} collected in Seoul

NO_3^--$\delta^{18}O$ was 75-90 ‰ which is similar to that of the city in the northeast area in the United States in winter (Fig. 6a). The isotope ratio measured in this study was characterized by dry depositions. There was a significant difference in the isotope ratio of NO_3^--$\delta^{15}N/\delta^{18}O$ depending on dry or wet deposition method (Felix and Elliott, 2014). There was a positive correlation between NO_3^--$\delta^{15}N$ and nitrate concentration of the PMs (Fig. 6b). It was also revealed that the concentration of NO_3^- was correlated to that of PM_{10} ($r = 0.90$, $p < 0.01$, $n = 10$ in this study, $r = 0.754$, $n = 23$; Górka et al., 2012). This indicated that the portion of emission sources with heavy NO_3^--$\delta^{15}N$ isotope ratio was higher compared to PM_{10} concentration in the ambient atmosphere. Therefore, it was assumed that the majority of PM_{10} originated from NO_X. However, this needs to be verified by establishing a nitrogen/nitrate isotope ratio library and comparing with isotopic composition of pollution sources in the future. Based on the hypotheses of this research, it is assessed that, in Seoul, vehicles are the major source of PM_{10} as well as NO_X.

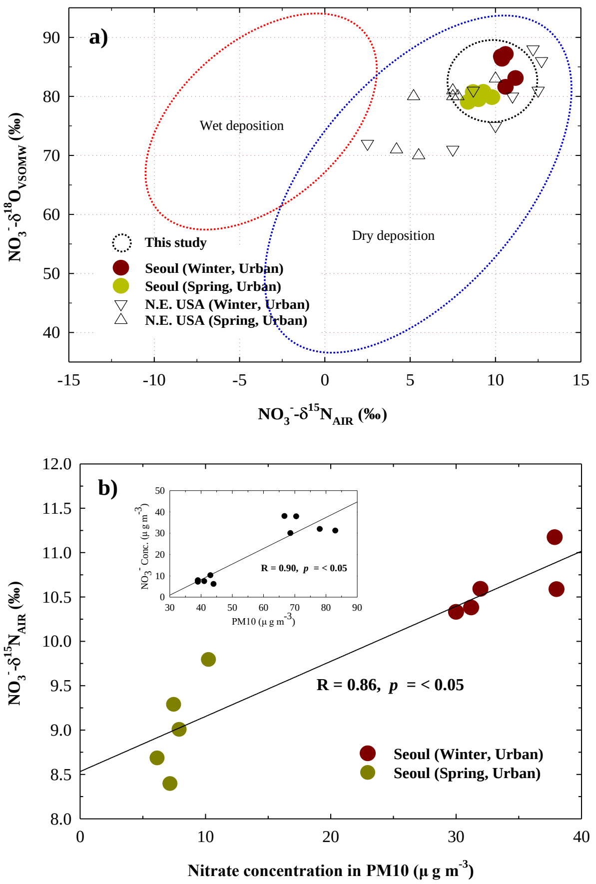

Figure 6: a) Composition of NO_3^--$\delta^{15}N$/$\delta^{18}O$ and b) comparison of NO_3^- and NO_3^--$\delta^{15}N$ in PM_{10}. Data was compared from Felix and Elliott (2014).

This study represents the inaugural attempt to measure stable isotope ratios in PM_{10} in Seoul, Korea, and to interpret the analysis data of stable isotope ratios by comparing them with pollution sources from similar values reported in the literature. In the future, it would be beneficial to investigate the isotope ratio inventories of various emission sources (e.g., automobiles, power plants, environmental infrastructures, soils, and concentrated animal feeding operations (CAFOs), etc.) in order to confirm the stable isotope ratios of PMs from the background area. By monitoring the PMs of the background area and the changes in the stable isotope ratio, it may be possible to track down sources of pollution more effectively.

3.3. The emission sources of PMs and its limitations

An air transport and dispersion model provided by National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (http://www.ready.noaa.gov) was utilized to find out the air flow patterns affecting the PM_{10} concentration in Seoul. The air-mass backward trajectories are classified using hybrid single particle lagrangian integrated trajectory (HYSPLIT), which has been used to predict movement path of a pollutant generated at a specific point over the time (Stein et al. 2015). The results of atmospheric transport pathway on the sampling day composed of the trajectories originated in the inner Mongolia region and the west sea of Korea nearby China (Fig. 7).

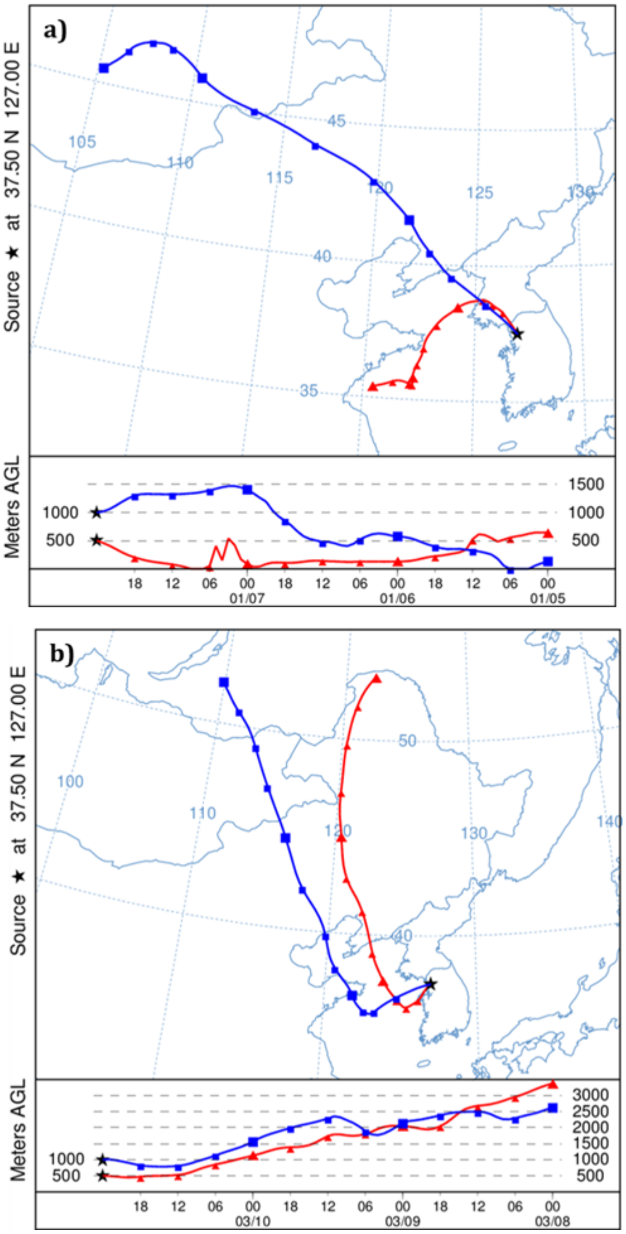

Figure 7: The atmospheric transport pathway of 72-h means backward air trajectories predicted by HYSPLIT for a) January and b) March at Seoul in Korea.

High concentrations of PM were observed in February (Shin et al., 2014). These indicate that PM_{10} concentrations in Seoul during the episode period were dominated by long-range transport from the China region as well as domestic sources (Kim et al., 2016). This study conducted sampling events during January and March, when high PM concentrations were observed. The sources of PM_{10} were similar during the measurement period of this study, and therefore the results of the isotope ratio comparison were also attributed to traffic and heating during the period of high PM_{10} concentration.

A backward trajectory of the atmosphere indicates that PM sources may influence long-range transportation from Mongolia or China. However, isotopic analyses confirm that the PM sources originated from domestic dominants. Nevertheless, further research is required to overcome the following limitations and reach more precise conclusions. The emission source tracking is limited by comparison with the isotope ratio results performed by overseas researchers. We propose the establishment of a library of an isotope ratio inventory of various emission sources. Additionally, we aim to monitor the isotope ratio of airborne particulate matter and its precursors over a seasonal or long-term period and identify the baseline concentration of stable isotopes in domestic background areas. These steps will significantly enhance our ability to trace emission sources with greater precision.

3.4. Effects of the PMs concentrations on distribution industry

The characteristics of PM emissions can be divided into 1) changes in PM concentration according to seasonal factors, 2) high concentration events, and 3) source differences. PM emissions affect various industries, such as distribution, health care, and services, resulting in changes in consumption patterns (Bae & Baek, 2020; Han & Moon, 2022; Kim et al., 2022). Therefore, a consumption strategy, when considering characteristics of PM emissions, holds the potential to positively influence the industries affected. The results of these studies can be used to evaluate consumption patterns and sales impacts in the distribution sector. In addition, it is used for product development and sales planning to respond to PM emissions.

4. Conclusion

In this study, the isotopic composition of carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$), and sulfur ($\delta^{34}S$) among the PM_{10} was analyzed to identify the emission source of PMs in Seoul. In addition, NO_3^--$\delta^{15}N/\delta^{18}O$ were analyzed using the denitrification method by microorganisms. The results of isotope ratios compared that $\delta^{13}C$ and $\delta^{15}N$ values obtained in this study were very similar to those of PMs collected in Paris. Based on the results, we hypothesized that a major emission source of PMs in Seoul might be combustion of liquidized natural gas. In comparison to other studies conducted during periods of high PM concentrations, the observed results are consistent with the hypothesis that pollution is primarily domestic stagnation in transportation and heating activities. The results revealed that most of the PM_{10} in Seoul originated from vehicles.

In the future, it is important to establish the inventories of the isotope ratios and monitor the isotope ratio of PM_{10} by season or for a long term. Analysis of the isotope ratio, together with additional and various stable isotope ratio libraries, can be efficiently used for tracking down the sources of pollution and the composition of PM_{10}. Additionally, it is expected that the results of this study can be used for the analysis of the sales patterns in the distribution industry. Identifying the sources of PM emission has revealed a significant domestic factor in the occurrence of high concentrations events. This finding, which holds promise for effective PM management, can be used to promote policies to reduce PM by establishing strict emission regulations and measures to supply eco-friendly fuels and electric vehicles. Therefore, regarding PM management, tracking the PM source can be essential in preparing policy implications.

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