이학석사 학위논문

Analysis of a Visibility Trend and its Contributing Factors in Korea for 2012-2018

2012-2018년 한반도 시정 변화와 기여요인 분석

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Abstract

Visibility is determined by light extinctions due to gases and particles in the atmosphere as well as meteorological conditions. In particular, fine particular matters (PM) are one of important factors for affecting visibility, which is thus known to be a representative indicator of sensible air pollution. Despite of continued decreases of PM concentrations in South Korea, the public, however, perceive that PM air pollution in South Korea has worsened over the past. To understand and explain this disparity, we here use long-term hourly visibility observations at six sites in South Korea for 2012-2018 and analyze contributing factors to their variations including PM concentrations, and its chemical compositions along with meteorological conditions. We find that annual mean visibility in Seoul and Daejeon has improved by 0.7 km and 0.4 km, respectively, for the past 7 years, while Gwangju, Ulsan, Jeju, and Baengnyeong have shown its degradations by 0.7 km, 2.9 km, 0.6 km, 1.4 km for the same period. For high PM seasons, the frequency of hourly poor visibility (< 6.7 km) in Seoul, however, has increased by 11% in winter and Daejeon has also shown an increase of poor visibility frequency by 13% in spring. The frequencies of hourly poor visibility in Gwangju, Ulsan, and Baengnyeong have increased by 9%, 15%, 13%, respectively, regardless of seasons. Our analysis reveals that PM composition changes from sulfate to nitrate aerosols are a major factor for increasing hourly poor visibility frequencies in Seoul, Daejeon, and Baengnyeong, whereas meteorological conditions including relative humidity and windspeed changes are important factors for visibility degradations in Gwangju, Ulsan, and Jeju. We find that nitrate aerosols account for about 53% of visibility degradations in all regions. Increases of nitrate aerosol concentrations are driven by NO_x emission changes and the reduction of sulfate aerosol concentrations, which makes additional NH_3 available for ammonium nitrate production in the atmosphere.

Keywords : Visibility, Light extinction coefficient, Sensible air quality, $PM_{2.5}$, Nitrate

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1.INTRODUCTION

Visibility degradation is a decrease in visible distance due to light scattering and absorption by particles or gases in the atmosphere (Jung et al., 1996). Therefore, a visibility change is significantly related to factors, including solar radiation, clouds, precipitation, and atmospheric chemical compositions such as gases and particles (Jung et al., 1992; Choi et al., 1993; Lee et al., 1995; Jung et al., 2000). It is known that the most of the public sense visibility degradations as worsening of air quality, in particular, for particulate matter (PM) air quality changes in the atmosphere (Kim et al., 2002; Han et al., 2004; Lee et al., 1996).

In South Korea, high PM concentrations are one of serious concerns for the public (SEF, 2017). In 2013, World Health Organization (WHO) denounced PM as a first-class carcinogen (IARC, 2013) and consequently, PM forecasting and warning systems were implemented in early 2014 in South Korea. In addition, since March 2018, more frequent warnings due to the strengthening of Korea's environmental standards for PM air quality criteria have also contributed to raising public awareness of PM issues. Particulate matters in the atmosphere, depending on their diameters (D), are classified as PM_{10} (D <= 10 μm), and $PM_{2.5}$ (D <= 2.5μm) (Nel et al., 2005). Despite of a growing public concern for PM air pollution, PM_{10} and $PM_{2.5}$ concentrations in South Korea have continuously decreased on an annual mean basis over the past (Yeo et al., 2019).

Effects of chemical and optical properties of PM on visibility degradation have been actively investigated using observations at Interagency Monitoring of Protected Visual Environments (IMPROVE) in the United States, North American Research Strategy for Tropospheric Ozone (NARSTO) in Canada, the United States, and Mexico, and European Experiment on Transport and Transformation of Environmentally Relevant Trace Constituents (EURO-TRACΠ) in Europe (Kim et al., 2003). China has improved visibility through research on the correlation between visibility and PM (Whang et al., 2006) and analysis of various weather factors (Zhang et al., 2010). In South Korea, there are many studies, investigating the characteristics of visibility changes and their long-term trends (Lee et al., 2006), spatial and temporal visibility changes (Lee et al., 2006), and factors for visibility degradations (Jung et al., 1996).

However, previous studies in South Korea relied on visibility observations conducted with a traditional method, which measures a maximum distance with the naked eye (Kim et al., 1998) so that it is naturally prone to large uncertainty. Especially, the higher the visibility, the greater the uncertainty of the visibility value is because the naked eye is not that sensitive to visibility changes as it increases. In order to overcome this limitation, the light intensity observations from the transmitter have been used to calculate visibility up to 20 km (Jung et al., 1996). These two methods, however, show some discrepancies of visibility observations depending on the environmental conditions (Lee et al., 2015).

In this study, we conduct an analysis of a long-term visibility trend for 2012-2018 and examine contributing factors for its change in South Korea, where visibility had been measured using the traditional eye observations for 2012-2016 and its measurement was switched to using the light intensity observations after 2017 and onward (Lee et al., 2015). Prior to the analysis, we use observations of PM and meteorological variables to reconstruct long-term visibility data, which are consistent with visibility observations with the two method and use this reconstructed dataset for our trend analysis.

Despite the fact the annual average concentration of PM in South Korea tends to decrease continuously (Figure 1), the anxiety of people's air pollution is growing. Based on our reconstructed visibility dataset, as a representative sensible index that can visually identify air pollution, we conduct the analysis of its changes in the annual average and in the frequency of poor visibility for six regions in Korea from 2012 to 2018. We also investigate the relationship between the visibility degradation and PM air quality and try to understand how much of a visibility change can be attributed to PM air quality degradation in South Korea.

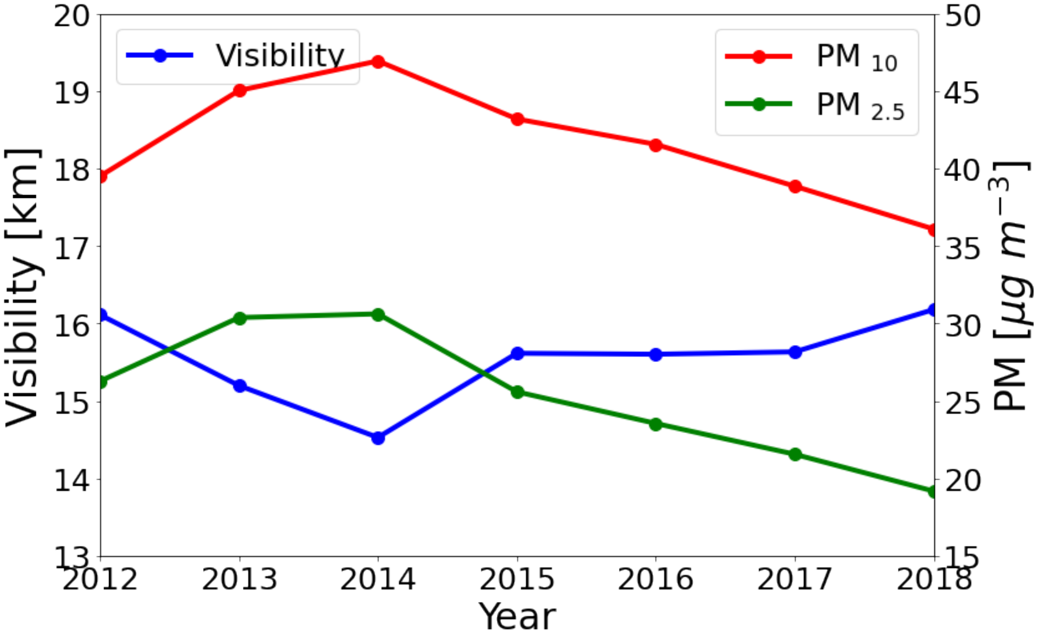

Figure 1. Annual mean concentrations of PM_{10} (red closed circles), $PM_{2.5}$ (green closed circles) averaged at all Airkorea sites for 2012-2018. Annual mean visibility is indicated with blue closed circles.

2. Data and Methods

2.1. Observations

In our analysis, we used meteorological observations at ground sites of the Korea Meteorological Administration (KMA) from 2012 to 2018 in major cities in South Korea, including visibility, yellow dust, precipitation, relative humidity, and wind speeds. Hourly PM_{10} and $PM_{2.5}$ concentrations and $PM_{2.5}$ chemical components (SO_4^{2-}, NO_3^-, K^+, Mg^{2+}, Ca^{2+}, organic carbon (OC), elemental carbon (EC)) measured at six sites of the National Institute of Environmental Research (NIER) were also used (Figure 2) . From 2012 to 2016, NOx and SO_2 emissions data from the National PM Information Center were used for the analysis (Table 1).

We divided South Korea into the metropolitan area (Seoul), central area (Daejeon), Honam area (Gwangju), Yeongnam area (Ulsan), Jeju, and Baengnyeong for the analysis of regional changes. Visibility observations with the naked eye were conducted every hour during the day and every three hours at night (18:00 to 03:00). From January 1, 2017 and onward, the observation method has changed from the eye observation to the instrument observation.

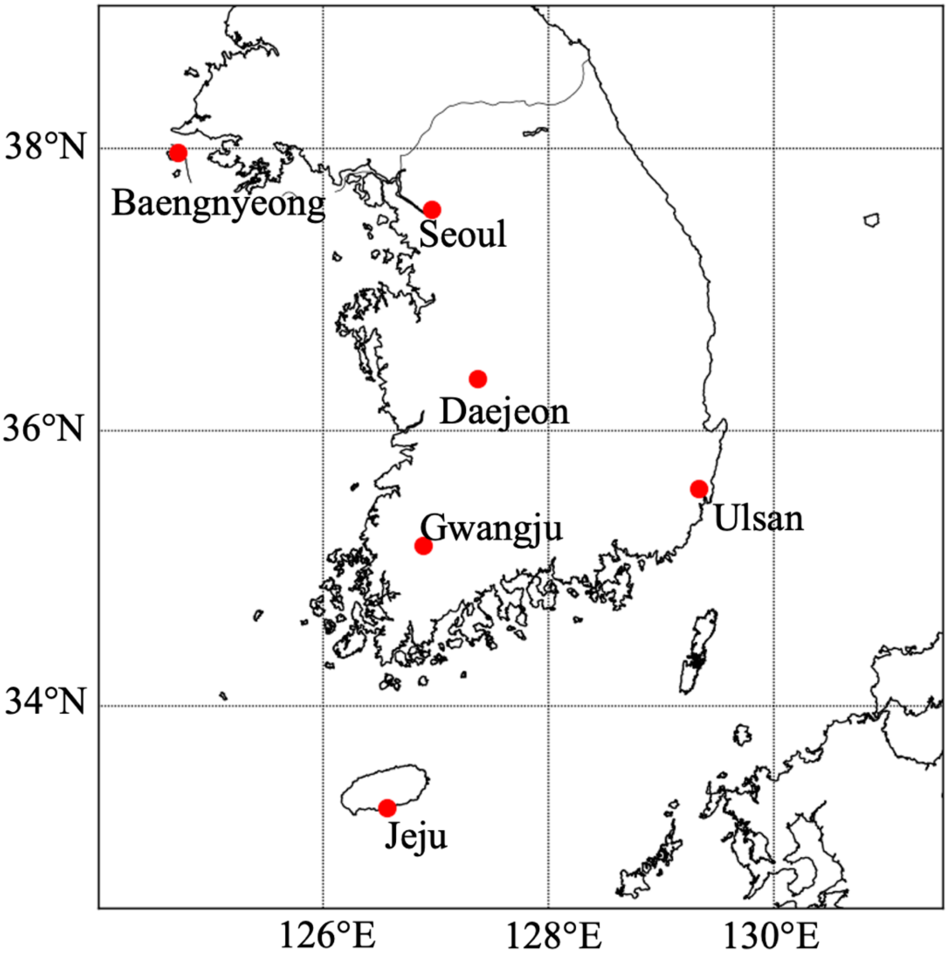

Figure 2. Locations of six ground sites : Seoul (37.57°N, 126.96°E), Daejeon (36.37°N, 127.37°E), Gwangju (35.17°N, 126.89°E), Ulsan (35.58°N, 129.33°E), Jeju (33.24°N, 126.56°E), and Baengnyeong (37.97°N, 124.71°E).

Table 1. Data descriptions.

	Regions	Valuable	Period
National Institute of Environmental Research	Seoul (108), Daejeon (133), Gwangju (156), Ulsan (152), Jeju (189), Baengnyeong (102)	PM_{10}, $PM_{2.5}$, SO_4^{2-}, NO_3^-, K^+, Mg^{2+}, Ca^{2+}, OC, EC	2012 - 2018
Korea Meteorological Administration		Visibility, Temperature, precipitation, windspeed, relative humidity, yellow dust	2012 - 2018
National PM Information Center		NOx and SO_2 emissions	2012 - 2016

2.2. Data Reconstruction

Visibility observations may differ depending on the measurement methods (Lee et al., 2015). In particular, the weather conditions (clear, rain, fog, etc.) significantly affect the eye observation, which shows a discrepancy from that of the instrument observation (Lee et al., 2015). Therefore, it is important to calibrate visibility observations by removing spurious signals.

Precipitation is a typical meteorological phenomenon that causes poor visibility. Annual mean visibility in the presence of precipitation decreases in Seoul, Daejeon, Gwangju, Ulsan, Jeju, and Baengnyeong by 4.97 km, 4.35 km, 3.87 km, 4.74 km, 5.73 km, and 7.65 km, respectively. Another cause of deterioration of visibility is yellow dust. The mean visibility when yellow dust occurs was lower than the mean visibility without yellow dust by 0.57 km, 0.53 km, 0.51 km, 0.54 km, 0.76 km, and 0.72 km in Seoul, Daejeon, Gwangju, Ulsan, Jeju, and Baengnyeong, respectively. According to the Meteorological guidelines of the KMA, fog is defined as when horizontal visibility is less than 1 km and relative humidity is close to 100%. In order to investigate the effect of PM and its composition on visibility, the hourly visibility data with precipitation and fog during the analysis period were removed, the daily visibility data with yellow dust was also removed based on the date of yellow dust observation by the KMA.

A systematic gap between visibility datasets by two different methods exists and has been a significant limitation for the long-term visibility analysis because visibility observations have been conducted by naked eye until 2016 and later switched to the

Table 2. Light extinction coefficient equations of IMPROVE and NIER.

Institute	Composite equation for light extinction coefficient
IMPROVE (1994)	3xf(RH){[NHSO]+[NHNO]}+4[OMC]+10[EC]+1[FS]+0.6[CM]+b_abs,NO_2 +b_Ray
IMPROVE (2007)	2.2 × fS(RH) × [Small Ammonium Sulfate] + 4.8 × fL(RH) × [Large Ammonium Sulfate] + 2.4 × fS (RH) × [Small Ammonium Nitrate] + 5.1 × fL(RH) × [Large Ammonium Nitrate] + 2.8 × [Small Organic Mass] + 6.1 × [Large Organic Mass] + 10 × [Elemental Carbon] + 1 × [Fine Soil] + 1.7 × fSS(RH) × [Sea Salt] + 0.6 × [Coarse Mass] + Rayleigh Scattering (Site Specific) + 0.33 × [NO_2 (ppb)]
NIER (2006)	0.91x(3[NHSO]x(1-RH/100)$^{-0.7}$)+1.34x(3[NHNO]x(1-RH/100)$^{-0.7}$) +1.06x(4[OMC]x(1-RH/100)$^{-0.4}$) +0.98x(10[EC]) +1x(2[FS])+(2[FS]) +1x(0.6x[CM])+153.53

NHSO = 1.375[SO_4^{2-}], NHNO = 1.29[NO_3^-], OMC = 1.41[OC], FS = 1.89[Al] +2.14[Si] +1.40[Ca] +1.43[Fe], CM =[PM_{10}]-[$PM_{2.5}$]

instrument observation. In order to make a consistent visibility dataset, we employ a statistical equation to reflect light extinctions by gases and aerosols, which can be used to compute reconstructed visibility. Table 2 shows light extinction equations used for visibility calculations developed by IMPROVE (Sisler et al., 1993) and NIER (NIER, 2013). They are based on observed PM component concentrations and ambient relative humidity (RH). We evaluated all these equations by comparing their results against the instrument visibility data for 2017-2018.

We find that the correlation coefficients between the reconstructed visibility and the instrument visibility in South Korea are 0.82 using the NIER equation, 0.26 using the 1994 IMPORVE equation, and 0.21 using the 2007 IMPROVE equation for 2017-2018. To convert light extinctions into visibility, we used the following *koschemeider formula (Equation 1)*.

$$Visibility = \frac{3.912}{light\ extinction\ coefficient}, \tag{1}$$

We apply the NIER equation, which shows highest correlation coefficient (0.82) with the instrument observation data. However, it has a theoretical maximum value of visibility, 25.4 km, which is significantly different from the maximum visibility observations by the naked eye. In particular, good visibility (> 20 km) accounts for about 30% of the total visibility. To solve this problem, low visibility values (< 17 km) were used for our analysis. Additionally, the reconstructed visibility data were corrected by fitting the mean distribution of the daily mean visibility using the NIER equation to the observed mean visibility. The correlation coefficient between the reconstructed visibility and the instrument observation increases to 0.86, which is higher than using the NIER equation (0.82).

2.3. Poor visibility frequency

Definition of poor visibility is quite arbitrary. For example, Landscherg (1974) in the United States used visibility (< 10 km) as poor visibility, while Corfield (1968) in the UK and Nomoto (1983) in Japan used much smaller value (< 5 km), and Yong-seung Jung (1992) and Seung-beom Lee (1994) defined poor visibility (< 1 km). In this study, we use poor visibility (< 6.7 km), which is the lowest 20% of daily mean visibility in South Korea. Using this criterion, annual, seasonal, and diurnal (rush hour) frequencies of poor visibility were calculated. The poor visibility ratio was obtained by dividing the poor visibility frequency by the total number of observations in each region.

3. Results and discussion

3.1. Annual visibility trend

The annual average concentration of PM_{10} and $PM_{2.5}$ from 2012 to 2018 in the South Korea has decreasing trend overall (Figure 1). Unlike the PM concentrations that decrease in all regions, the annual average visibility is divided into two categories, regions showing an increasing trend, and regions showing a decreasing trend. (Figure 3). In Seoul and Daejeon, annual mean visibility increased by 0.7 km and 0.4 km, respectively, and decreased by 0.3 km in Gwangju, 3 km in Ulsan, 0.3 km in Jeju, and 1.3 km in Baengnyeong for 2012-2018. Comparing the slope of the linear regression equation during the analysis period using the annual average concentration for each region (Table 3), it can be seen that the visibility of Gwangju, Ulsan, Jeju, and Baengnyeong are decreasing despite the steady decrease in PM concentration.

As the increase or decrease in the visibility cannot be explained simply by PM concentration, the analysis of change in concentrations of PM constituents (sulfate, nitrate, OC, and EC) and meteorological conditions were conducted . In Figure 4, sulfate decreases up to 23% in all regions from 2013 to 2017, and rapidly increases in Gwangju, Ulsan, and Jeju in 2018. Nitrate tends to increase by 26%, OC decreases by 7%, and EC decreases by 32% during the analysis period in all regions. Relative humidity increases in all regions until 2015, and then tends to increase in Baengnyeong, decrease in Seoul and Daejeon.

Table 3. Slope of linear regression lines of annual mean visibility by region.

Location	Visibility ($km\ year^{-1}$)	$PM_{2.5}$ ($\mu g\ m^{-3}\ year^{-1}$)
Seoul	0.2032	-2.274
Daejeon	0.0484	-1.532
Gwangju	-0.0751	-3.372
Ulsan	-0.5772	-3.773
Jeju	-0.0197	-3.194
Baengnyeong	-0.2122	-0.533

In Gwangju, Ulsan, and Jeju, the relative humidity has increased until 2016 and decreased after 2016. The annual mean wind speed steadily decreases from 2.8 m s^{-1} to 1.5 m s^{-1}during the analysis period and decreases in all regions, and in the case of Baengnyeong, the wind speed is twice higher than compared to other regions.

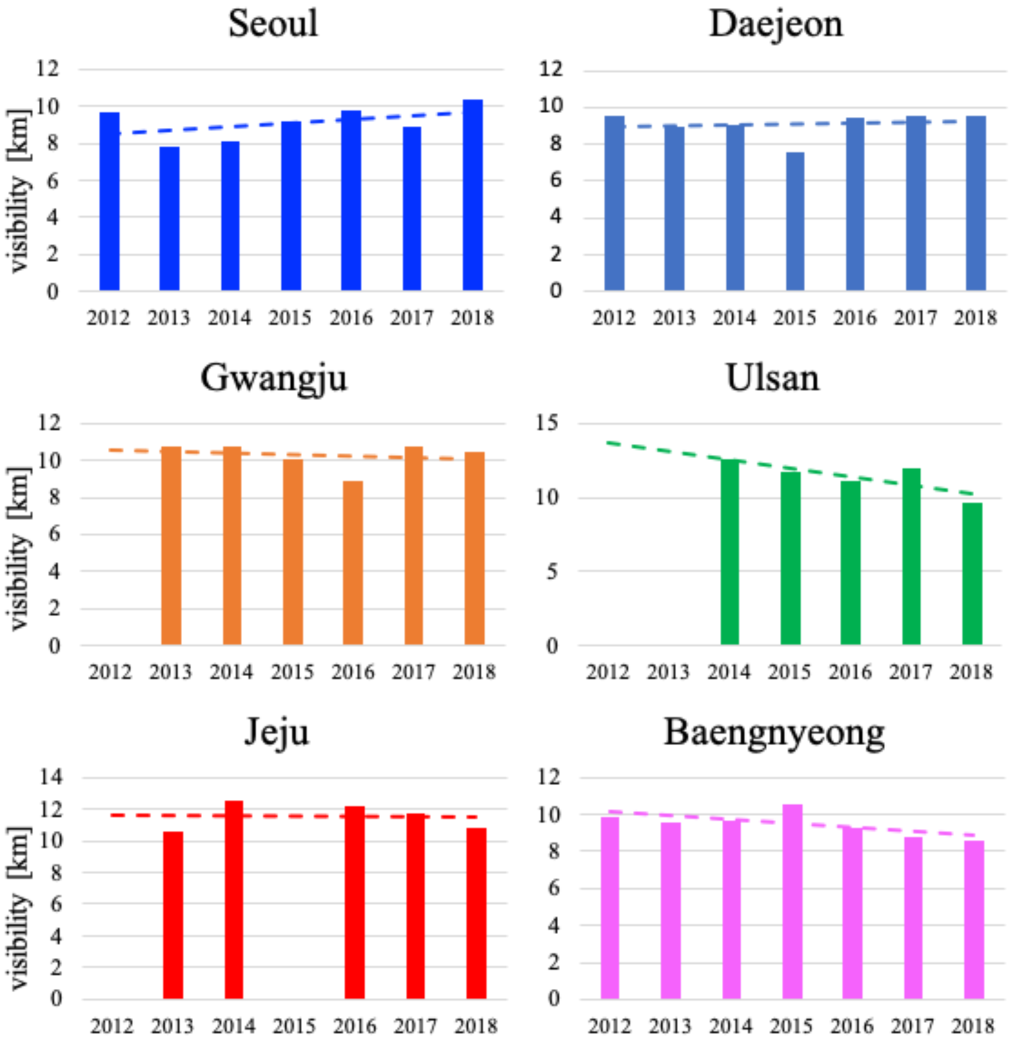

Figure 3. Annual mean trend of reconstructed visibility less than 17 km by corrected visibility equation. Dotted line indicate annual mean trend of the each area. Seoul and Daejeon has increasing trend while Gwangju, Ulsan, Jeju and Baengnyeong has decreasing trend.

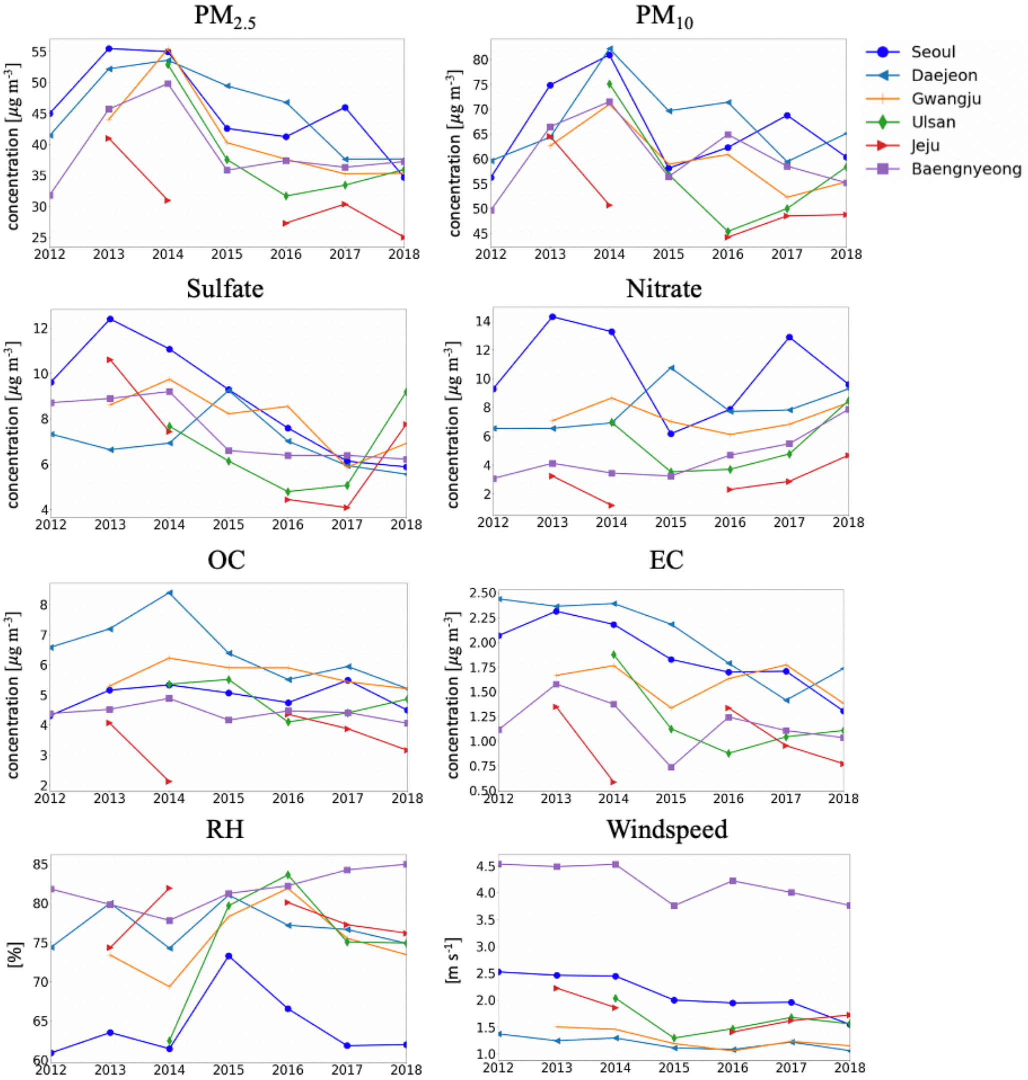

Figure 4. Annual variation of PM and PM components (sulfate, nitrate, OC and EC), relative humidity (RH), and windspeed.

3.2. Poor visibility analysis

3.2.1. Annual trend of poor visibility ratio

Despite the increase in the average visibility, if the poor visibility ratio increases, there is a possibility that the public consider air quality is not improved. We analyze the trend of poor visibility ratio for six sites and it is shown in Figure 5 (a). The poor visibility ratio by year in black, and the trend of poor visibility ratio decreased by 2% in Seoul and 0.2% in Daejeon, and 5% in Jeju, and increased by 9%, 15% and 12% in Gwangju, Ulsan, and Baengnyeong, respectively. This indicates that the visibility of Seoul, Daejeon and Jeju is improving, and that of the rest of the sites are worsening. However, in Jeju, poor visibility ratio decreased until 2014 by 4%, but the ratio increased from 2016 to 2018 by 11%. Through this, it can be confirmed that from 2012 to 2018 in Seoul and Daejeon, the public feel that the air quality is getting better while Gwangju, Ulsan, and Baengnyeong's air quality is getting worse. In Jeju, since 2016, it is deduced that the air quality in Jeju has been deteriorating.

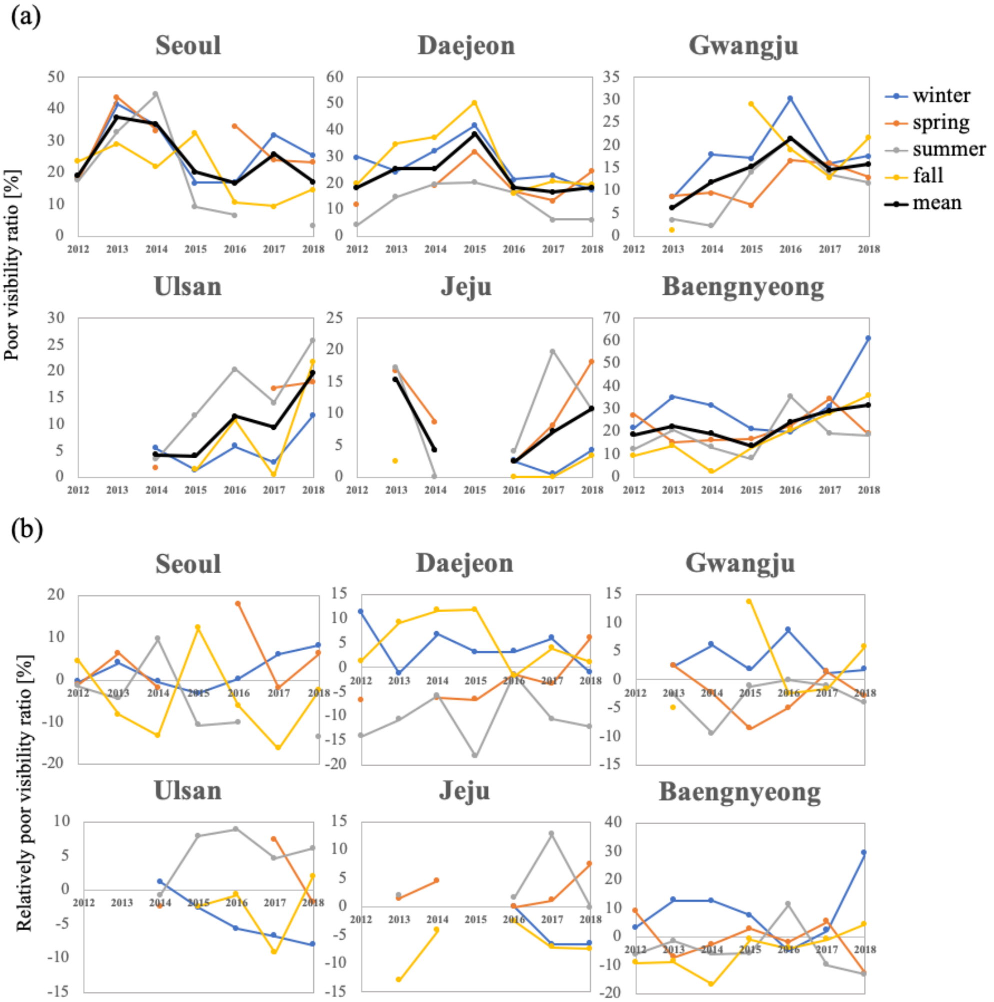

Figure 5. Annual and seasonal poor visibility ratio changes (a) Annual poor visibility ratio in black, winter in blue, spring in orange, summer in gray, and fall in yellow. (b) Seasonal poor visibility ratio – mean poor visibility ratio.

3.2.2. Seasonal trend of poor visibility ratio

Figure 5 (a) shows the ratio of poor visibility for each season to find out the characteristics of seasonal trends, winter in blue, spring in orange, summer in gray, and fall in yellow. The change in the ratio of poor visibility in each season is not significantly different from the average value. It has decreasing trend in Seoul and Daejeon, and increasing trend in Gwangju, Ulsan, and Baengnyeong. In Jeju, it has decreasing trend overall, however, increases since 2016. In order to investigate if the poor visibility ratio anomaly particularly high in a specific season, the annual mean poor visibility ratio was subtracted from the poor visibility ratio for each season. That is seasonal relative poor visibility ratio of each region and indicates how much the seasonal poor visibility ratio changed from the average in Figure 5 (b).

Since 2015, in Gwangju, ratio of relative poor visibility in spring has increased by 9%, and the poor visibility ratio in summer increases by 10% from 2014 to 2017. Relative poor visibility ratio of Baengnyeong increased rapidly since 2016 in winter, and the ratio reached 29%. In Seoul, the relative poor visibility ratio has been increasing since 2015 in winter, and the ratio increased by 11% in 2018 compared to 2015. In the case of Daejeon, the relative poor visibility ratio increases rapidly by 13% in spring. In Jeju, the relative ratio of spring has increased by 7% from 2016. Seasonal analysis showed that the ratio of relative poor visibility increased rapidly in winter and spring, which could explain the deterioration of air quality except Ulsan. It is confirmed the ratio has a certain tendency to increase in winter and spring, which high PM period, for each region. If the ratio of relative poor visibility in a particular season continued to increase compared to the previous year, the public feels that the air quality has deteriorated compared to the previous year.

3.2.3 Trend of poor visibility ratio during rush hour

The changes in visibility of rush hours was analyzed to investigate sensible air pollution changes when people were active outside. Figure 6 shows the poor visibility ratio at each rush hour period (morning rush hour : 7-9 am, afternoon rush hour : 5-7 pm) by region. The morning rush hour showed a higher ratio of poor visibility than the afternoon rush hour in all regions.

In Seoul and Daejeon, where the mean poor visibility ratio is decreasing, even during rush hour, the ratio has decreasing trend. In Seoul, the poor visibility ratio decreases by 5% in the morning and decreases by 2% in the afternoon, and Daejeon decreases by 2% in the morning and 12% in the afternoon rush hour. In Ulsan and Baengnyeong, the ratio of poor visibility increases on both morning and afternoon rush hour. the poor visibility ratio of Ulsan increased by 12% in morning and 6% in afternoon rush hour. In Baengnyeong, 18% in morning and 10% in afternoon rush hour.

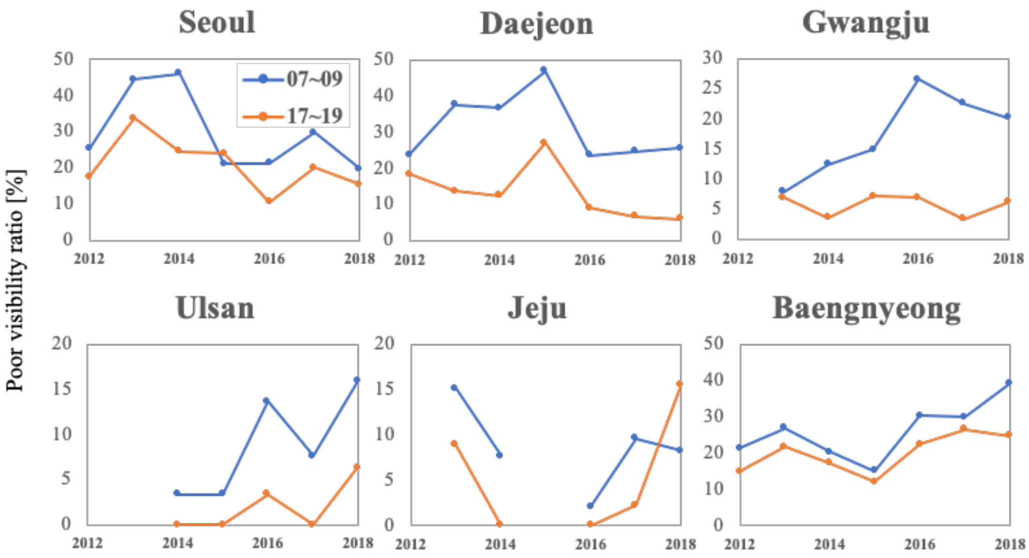

Figure 6. Poor visibility ratio at morning rush hour (07-09) in blue, at afternoon rush hour (17-19) in orange, and diurnal mean in gray. Morning rush hour ratio is larger than afternoon rush hour ratio.

Gwangju and Jeju has opposite trend on morning and afternoon rush hour. In Gwangju, the poor visibility ratio on morning rush hour increases by 20% while afternoon rush hour decreases by 6%. In Jeju, the poor visibility ratio on morning rush hour decreases by 8% while afternoon rush hour increases by 15%. The increase of poor visibility ratio in Gwangju has a greater impact on the morning rush hour, and the decrease in the poor visibility ratio in Jeju also has a great influence on morning rush hour than afternoon rush hour. The changes of poor visibility ratio in each region can be deduce from the analysis of morning rush hour which has a greater effect on the poor visibility ratio than afternoon rush hour.

3.3. Conditions affecting poor visibility

3.3.1 PM composition variation

In order to investigate the major factor that causes poor visibility, the concentration changes of sulfate, nitrate, OC, and EC are shown in Figure 7. The ratio of poor visibility in Seoul can be explained by the changes in nitrate and sulfate concentrations from 2012 to 2015, the change in nitrate follows the trend of poor visibility. Nitrate continues to increase since 2015, but as OC and EC decrease, and therefore contributes more to the poor visibility ratio decline in 2016. The ratio of poor visibility in Daejeon increased by 15% until 2015, where the value shows maximum, and both sulfate and nitrate concentrations also showed peaks in 2015. While sulfate and nitrate decrease until 2017, and the concentration of nitrate increases in 2018, this coincides with the decrease and increase of the ratio of poor visibility in Daejeon. The poor visibility ratio in Baengnyeong is increasing from 17% to 29%. Although the concentration of sulfate decreased by 5 $\mu g\ m^{-3}$, the concentration of nitrate increased by 10 $\mu g\ m^{-3}$, so an increase in the rate of poor visibility can be explained by nitrate than sulfate.

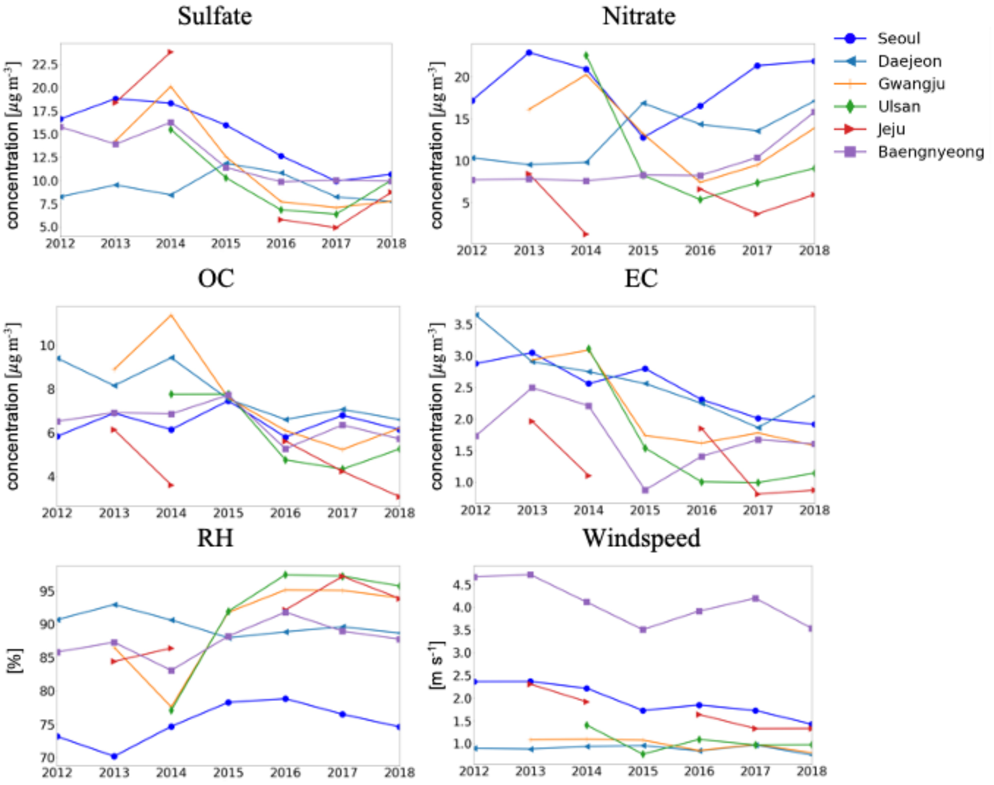

Figure 7. Annual variation of PM components, relative humidity, and windspeed at poor visibility. Regional annual variation of each component when reconstructed visibility lower than 6.47km. Sulfate, OC, EC, and windspeed has decreasing trend. Trend of nitrate and RH are depends on regions**.**

In Gwangju, The poor visibility ratio starts at 5% in 2013 and continues to increase, then reaches the highest in 2016 (35%) and becomes 20% in 2017. However, the concentrations of nitrate, sulfate, OC, and EC are peaked in 2014. Since it has a tendency to decrease after that, the analysis of the concentration of PM components cannot explain the increase in the trend of poor visibility in Gwangju. As in Gwangju, the ratio of poor visibility in Ulsan has steadily increased from 3% to 18% since 2014, however, since the concentrations of sulfate, nitrate, OC, and EC are all decreased. Therefore, the change in the concentration of PM components cannot explain the increase of the poor visibility ratio. In Jeju, the ratio decreases by 8% during 2013 to 2014. This is explained by the rapid decrease of concentrations of nitrate, OC, and EC. However, from 2016 to 2017, the increasing trend in the ratio of poor visibility cannot be explained with the decreasing trend in the concentration of PM components.

3.3.2. Meteorological conditions variation

Trends of Gwangju, Ulsan, and Jeju, where poor visibility ratio changes were poorly linked to variations in the concentration of PM components, can be explained by variations in meteorological conditions. In 2014, when the concentration of sulfate and nitrate was the highest in Gwangju, the relative humidity was the lowest at 77%. In 2016, while the concentrations of nitrate and sulfate were low, the relative humidity was 95% or higher. The increase of relative humidity increases hygroscopicity of sulfate and nitrate even if the concentrations of sulfate and nitrate are small, it causes poor visibility with a large growth factor. In addition, wind speed was 0.85 m s^{-1} which can cause poor visibility. In

2017, the ratio of poor visibility decreased by 10% because the relative humidity decreased slightly and the wind speed increased, but after that, the poor visibility ratio increased again in 2018 due to the increase in nitrate and sulfate concentrations and the decrease in wind speed.

In Ulsan, increase of poor visibility ratio also can be explained by the relatively high relative humidity since 2014. Although the concentrations of the PM components all decreased until 2016, the relative humidity which peaked in 2016 could explain the increase in the poor visibility ratio. In 2018, the relative humidity decreases, however the concentration of all the components that make up the PM is increasing, which explains the increase in the poor visibility ratio. The relative humidity of Jeju increased from 92% to 97% in 2016-2017, and wind speed decreased from 1.7 m s^{-1} to 1.3 m s^{-1}. These weather conditions seem to increase the poor visibility ratio regardless of decrease in the concentration of PM components.

In Gwangju, Ulsan, and Jeju, even if the concentration of sulfate and nitrate was low, the ratio of poor visibility was high if the relative humidity is high. It can be seen that the visibility of these sites is more affected by relative humidity and wind speed than by the effect of PM. Therefore, it is necessary to conduct the analysis on meteorological factors as well as the concentration of PM components depending on the region.

3.4 Variations of nitrate and sulfate

3.4.1 Relative light extinction coefficient budget changes

Figure 8 shows the relative light extinction coefficient budget for 2012 and 2018, which represents the relative influence of each PM component on poor visibility. The mean relative light extinction coefficient budget of nitrate is the most dominant, that is, it has the greatest impact on poor visibility, followed by sulfate, OC, EC, CM, and FS. The mean of relative nitrate extinction budget is about 51.5% and sulfate is about 32.5%. Compared to 2012 (Gwangju and Jeju : 2013, and Ulsan : 2014), the average of regional relative nitrate budget increased more predominantly by 53% in 2018 and the sulfate ratio decreased by 24% except in Jeju and Ulsan. As such, the effect of nitrate on the reduction of visibility is large, and it is gradually increasing, suggesting that nitrate will be a more important factor in analyzing visibility forward. However, since the budget of sulfate is increasing in Jeju and Ulsan compared to 2012 (Jeju : 2013), it is necessary to analyze the causes affecting the concentration trends of nitrate and sulfate. The main causes affecting the concentrations of the two are precursor emissions changes and the thermodynamic and stoichiometric nature of sulfate, nitrate formation.

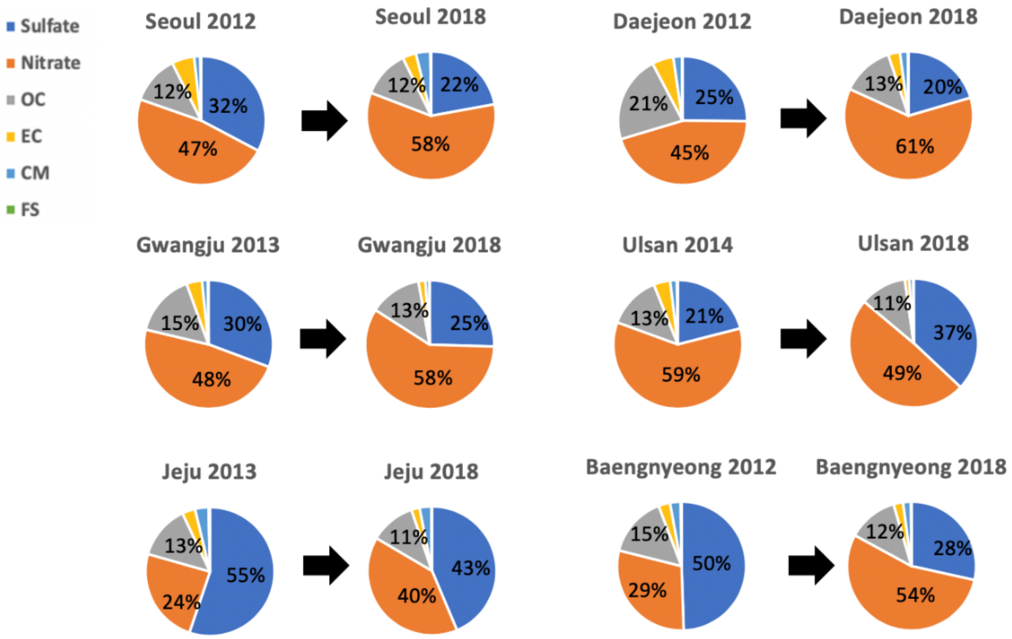

Figure 8. Relative extinction coefficient budget for each species at poor visibility (2012, 2018) Regional PM component budget changed in 2012 and 2018 respectively (Gwangju, Jeju Island in 2013, Ulsan in 2014).

3.4.2 Concentration change according to NOx and SO_2 emissions

Sulfate concentrations are decreasing in almost all regions, nitrate concentrations are decreasing in Gwangju and Jeju, Ulsan, and increasing in Seoul, Daejeon and Baengnyeong. In order to find out the causes of the different fluctuations by region, the changes in the emissions of NOx and SO_2 from 2012 to 2016 were analyzed. Table 4 indicates the annual emission rate change of NOx and SO_2 and changes in the annual concentration of nitrate and sulfate from 2012 to 2016.

Table 4. Annual (mol) concentration changes of nitrate, sulfate, NOx and SO_2.

Annual changes (% yr^{-1})		Seoul	Daejeon	Gwangju	Ulsan	Jeju	Baengnyeong
Concentration	Nitrate	4.5	7.7	0.4	-14.8	8.8	18.1
	Sulfate	-5.8	-1.04	-9.11	-8.81	-5.2	-6.5
Emission	NOx	4.5	5.3	0.8	-3.6	15.9	-
	SO_2	-2.5	-8.8	-15.2	-3.9	-4.6	-
Mol concentration	Nitrate	1.2	1.4	0.1	-5.4	1.2	2.2
	sulfate	-0.9	-0.3	-1.1	-1.5	-1.0	-1.0

The rate of increase in nitrate in Seoul is consistent with the rate of increase in NOx emissions at 4.5%, which indicates that most of the changes in emissions affected the concentration of nitrate. Changes in nitrate concentrations in Gwangju and Jeju are less than changes in NOx emissions. Decreases of NOx emission in Gwangju affect about 50% of nitrate concentration changes, and in Jeju NOx emission affect about 55% to nitrate concentration changes. The change in nitrate is much larger than the change in NOx emissions in Daejeon and Ulsan, which implies nitrate concentration can be inferred that not only the concentrations were changed by changes in NOx emissions, but also other factors, such as external influx, may have been affected.

Analyzing the relationship between sulfate concentration change and SO_2 emission change, Daejeon and Gwangju have less concentration change than emission change. SO_2 emission reduction of Daejeon and Gwangju affects about 30% and 50% in sulfate concentration change, respectively. Sulfate concentration changes in Seoul, Ulsan and Jeju are greater than the emission change, and it can be inferred that the concentration has changed due to the change of the emission, as well as the influence of other factors.

3.4.3 Increase of nitrate due to sulfate reduction

Nitrate and sulfate are exist in the state of ammonium nitrate and ammonium sulfate in the atmosphere. Ammonium nitrate are composed with ammonia and nitrate with a stoichiometric ratio of 1: 1, and in ammonium sulfate, ammonia and sulfate are combined with a ratio of 2: 1. Theoretically, a reduction in sulfate can make ammonium nitrate twice as much as the reduced amount. Table 4 shows the the mol concentration changes of sulfate and nitrate by region in order to confirm if the increasing concentration of nitrate is due to the decreasing concentration of sulfate. In Gwangju, Ulsan, and Jeju, both nitrate and sulfate are decreasing, so it cannot be confirmed that the reduction of sulfate is achieved by an increase in nitrate.

Except Ulsan, mol concentration changes of sulfate decreases and nitrate increases. Double the reduction in sulfate affects nitrate increase by 62% in Seoul, 4.5% in Gwangju, and 58% in Jeju. Reduction of sulfate of Daejeon and Baengnyeong can explain 36% and 94% of the increase in increase of nitrate mol concentration, respectively. The change in the molar concentration of sulfate in Baengnyeong is almost identical to the change in molar concentration of nitrate. This indicated that the increase in nitrate was mainly due to sulfate reduction. Especially in Daejeon, increase in nitrate concentration was much greater than the rate of change in NOx emissions and it can be deduced that the concentration of nitrate was changed by not only sulfate reduction and NOx emission change but also other mechanism such as external inflow.

In conclusion, in order to improve the air quality, efforts to reduce the concentrations of nitrate and sulfate, especially among PM components is necessary. However, if we only care about reducing the concentrations of these two, it can lead to worsening air quality because a decrease in sulfate concentration doubles the concentration of nitrate, which significantly affects poor visibility. Therefore, understanding of chemical reactions of each component must be carried out, and appropriate reduction policies suitable for regional characteristics such as relative humidity and wind speed must be implemented to effectively improve air quality.

4. Summary and Conclusions

Even though the concentration of PM is decreasing, people tend to feel that the air quality is getting worse. Long-term analysis and poor visibility analysis of visibility were conducted in six regions to investigate the cause. We conducted analysis of the change in annual, seasonal, and rush hour visibility. It was found that the annual average visibility is increasing in Seoul and Daejeon, and the ratio of poor visibility is decreasing. In Gwangju, Ulsan, Jeju, and Baengnyeong, the annual average visibility is not only lowered, but the ratio of poor visibility is increasing, indicating that the visibility is worsening. In the seasonal analysis, unlike the increase in annual average visibility in Seoul and Daejeon, the ratio of poor visibility in winter and spring is rapidly increasing, and the increase in poor visibility ratio causes a decrease in air quality.

Poor visibility changes in Seoul, Daejeon, and Baengnyeong can be explained by variations in the concentration of PM and PM composition, especially sulfate and nitrate. However, in Gwangju, Ulsan, and Jeju, with the relative humidity and wind speed we can explain poor visibility ratio changes better than the only changes in PM concentration. Our analysis describes four factors influencing the increase and decrease of visibility: nitrate, sulfate, RH, and WS, and how these factors can explain observes changes in visibility and poor visibility in each region.

The relative light extinction budget of nitrate for poor visibility is about 50%, and it has a greater influence on poor visibility than sulfate, which is about 30%. In Seoul, Daejeon, Gwangju, Jeju, and Baengnyeong, the nitrate budget increased significantly in 2018 and the sulfate budget decreased in 2018 compared to 2012, and the nitrate effect is likely to increase even further. Factors affecting the increase in nitrate include increased NOx emissions and reduced ambient sulfate concentrations. Theoretically, nitrate is produced twice as much as the reduction of sulfate, which was confirmed in Baengnyeong. Daejeon, an area where nitrate has increased more than twice the declining sulfate amount, may be affected by an increase in emissions or the possibility of foreign inflows. Since the increase in nitrate is much greater than the increase in NOx emissions, there is a possibility of foreign influx such as Chinese transport.

In order to make better air quality in the future, it is necessary to pay attention to the reduction and relative changes of PM composition, and it is especially important to reduce the concentration of nitrate. In addition, because meteorological conditions may be more important than PM concentration depending on regional characteristics, effective reduction policies should be implemented in consideration of meteorological characteristics of each region.

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국문 초록

2012-2018 년 한반도 시정 변화와 기여요인 분석

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시정은 대기의 혼탁도를 나타내는 척도로써 공기 중의 다양한 가스상 물질과 입자상 물질들의 빛 산란과 기상 조건에 의해 결정된다. 특히 체감 대기오염의 대표적인 지표로 알려진 미세먼지는 시정에 영향을 끼치는 중요한 물질 중 하나이다. 그러나 한국의 미세먼지 농도가 지속해서 감소하는 추세임에도 불구하고, 국민들은 과거보다 한국의 대기오염이 더 악화되었다고 인식하고 있다. 이러한 인식의 차이를 이해하고 설명하기 위해 2012년부터 2018년 동안 한국의 6개 지역에서 관측된 시정 자료를 사용하여, 미세먼지를 구성하는 성분 농도(질산염, 황산염, 유기 탄소, 원소 탄소)와 기상 조건(상대습도, 풍속 등)이 시정과 저시정의 빈도율 변화에 얼마나 기여하는지를 분석하였다. 서울과 대전의 연평균 시정은 지난 7년 동안 각각 0.7 km, 0.4 km씩 개선되었으며, 광주, 울산, 제주, 백령도는 각각 0.7 km, 2.9 km, 0.6 km, 1.4 km 씩 감소하였다. 광주와 울산, 백령도는 계절과 관계없이

저시정 빈도율이 각각 9%, 15%, 13%씩 증가한 반면, 서울은 겨울에, 대전은 봄에 한정하여11%, 13%씩 증가하였다. 서울과 대전, 백령도의 저시정 빈도율의 증가는 미세먼지를 구성하는 주요 성분이 황산염에서 질산염으로 변화하는 것으로 설명할 수 있는데, 이에 반해 광주, 울산, 제주도의 시정 저하에는 상대습도와 풍속의 변화가 더 크게 기여하고 있음을 알아내었다. 또한, 모든 지역에서 미세먼지 성분들 중 가장 크게 시정 저하에 기여하는 물질은 질산염으로 53% 기여하고 있다. 질산염의 농도 변화는 질소산화물의 배출량 변화와 황산염 농도의 감소로 인한 대기 중의 암모니아 생성으로 설명할 수 있다.

주요어 : 시정, 빛 흡광계수, 체감 대기질, 초미세먼지, 질산염

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