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Impact of international Maritime Organization 2020 sulfur content regulations on port air quality at international hub port

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ABSTRACT

In this study, various characteristics of port air quality that are affected by harbor activities, including ship emissions, were assessed at an international hub port (Busan port) and other major ports in South Korea from November 2017 to December 2020. We conducted an in-depth analysis of aerosol characteristics to investigate the impact of ship emissions, harbor activities, and emission control measures (reinforced limits of sulfur content in fuel oil, from 3.5% to 0.5% mass fraction) on the air quality around the Busan port, South Korea. Our results demonstrate that the impact of ship emissions on port air quality varied significantly according to chemical species. The largest impact on port air quality from ship emissions and air mass pathways (e.g., port wind sector) was confirmed by significantly higher concentrations of SO_2 at the port. Our results of regression discontinuity design, a quasi-experimental impact evaluation method, revealed that the implementation of reinforced regulations (i.e., International Maritime Organization 2020 policy) significantly reduced SO_2 and NH_4^+ concentrations at the Busan port, but did not affect the concentrations of $PM_{2.5}$ and NO_2.

1. Introduction

International trade has grown significantly over the past few decades, resulting in heavy ship traffic, increases in global exhaust emissions (carbon dioxide (CO_2), nitrogen oxide (NO_x), sulfur dioxide (SO_2), and particulate matter (PM)), and significant contributions to global anthropogenic emissions (Eyring et al., 2005; Endresen et al., 2007; UNCTAD, 2015). Additionally, local emissions from ships in coastal areas, including harbors, have contributed to increasing ozone (O_3) concentrations by up to 29 ppb in the south coast of California (Vutukuru and Dabdub, 2008) and by 15 ppb in Busan, a hub port city in South Korea, East Asia (Song et al., 2010). Port activities, including ship cruising in harbor areas, tend to deteriorate air quality; thereby affecting human health detrimentally. Therefore, the International Maritime Organization (IMO) has imposed regulations to limit the sulfur content in fuel oil used by ships operating outside designated sulfur emission control areas (SECAs). The sulfur content regulation, which allowed a maximum of 3.5% m/m (mass by mass) until December 31, 2019, was reduced to a maximum of 0.5% m/m from January 1, 2020 (IMO, 2020 policy) outside the SECAs. Beginning on September 1, 2020, major ports in South Korea were designated as SECA (0.1% sulfur content limit). However, characterizing the air quality around these ports is challenging owing to the diverse range of port emission sources and cruising speeds of the ships. The various emission sources include ocean going vessels (OGVs) at the coast, ship hoteling in the terminal, transit through shipping channels, cargo handling equipment (cranes and forklifts, among others), diesel trucks, and trains at the port.

The emission characteristics of air pollutants from OGVs and diesel on-terminal equipment, including trucks, were significantly different. OGVs emit large amounts of SO_2 because of the high sulfur content (the current limit of 0.5%) in low-grade heavy fuel oil (HFO), whereas significant quantities of black carbon (BC) and carbon monoxide (CO) are emitted from diesel using on-terminal freight-handling equipment, such as cranes and forklifts (Ault et al., 2009; Moldanová et al., 2009). Heavy diesel trucks that load and unload containers and freight in ports also lead to high concentrations of air pollutants (Kozawa et al., 2009), whereby significant public health concerns associated with BC and other air pollutants exist (Wu et al., 2009).

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Several studies have evaluated the effect of sulfur emission control policies, such as SECAs (0.1% sulfur content), mostly on the emissions of air pollutants (not ambient concentrations) in the major ports of the world (Kotchenruther, 2017; Wan et al., 2019; Zhang et al., 2019). However, studies on the effect of the IMO 2020 rule concerning sulfur content (from 3.5% to 0.5% outside the SECAs) on port air quality in non-SECA zones have not been performed. In this study, we quantitatively evaluated the impact of the IMO 2020 policy (starting from January 1, 2020) on air quality in the highly polluted port regions of South Korea, based on observational data analysis. Because SO_2 is the most signficant contributor to the strength of air pollutants in port areas (National Institute of Environmental Research (NIER), 2021), the IMO 2020 policy can affect port air quality (SO_2, SO_4^{2-}, and $PM_{2.5}$) as it related to the magnitude of sulfur content in fuel oil. To estimate the environmental policy effect on port air quality, we applied the regression discontinuity design (RDD) frame, a statistical tool, to a time-series dataset in which time is a running variable and treatment begins at a particular threshold in time (Chen and Whalley, 2012; Gallego et al., 2013; Zhang et al., 2020). To the best of our knowledge, this is the first study to examine the effect of the new IMO 2020 emission control policy on port air quality. Additionally, we investigated the effect of different air mass pathways (i.e., wind direction) on air quality in the study area affected by natural (marine) and anthropogenic (port facilities and ship emissions) sources before and after the implementation of the IMO 2020 policy. We also assessed $PM_{2.5}$ chemical characteristics via an intensive field campaign at the target area of this study: the Busan port.

2. Materials and methods

2.1. Data and methods

The air quality at six major ports (Busan (BS1 and BS2), Incheon (IC), Ulsan (US), Gwangyang (GY), and Samcheok (SC)) was investigated to assess the effect of port-related activities on the concentration of air pollutants. Note that the SC port was selected as the control, as it is known to be mostly unaffected by ship emissions, port activities, and transport from nearby urban areas. The BS port is the largest Korean port located in the Busan metropolitan city, which has an area of 770 km^2, a population of approximately 3.4 million, and 1.4 million registered vehicles as of 2020. The BS port is the fifth busiest container port in the world by cargo tonnage (known as the "Cargo Gateway of Asia") and is the largest transshipment port in East Asia (Song and Shon, 2014). The total harbor volumes (quantity of goods transported or freight tons and the sum of container and non-container volumes) at BS, GY, US, and IC ports in 2018 (and 2019) were 28.4% (28.5%), 18.6% (18.8%), 12.5% (12.3%), and 10.1% (9.6%) of the national harbor volume, respectively, whereas those at SC were less than 0.3% (0.4%) (2020 Port Management Information System, Port-MIS, https://new.portmis.go.kr/portmis/websquare/websquare.jsp?w2xPath = /portmis/w2/main/intro.xml). At the BS port, the cargo volume from container ships (95%) in 2019 was much higher than that (5%) of non-container ships (e.g., bulk carriers and general cargo ships), while the rest of the ports (GY, US, and IC) contained more dominant cargo volume from non-container ships (65–96%) (Table S1). Depending on the nature of the cargo handled by each port, the effects on the air quality of the ports may differ. In addition, the BS port is the pivotal international trade port in South Korea, accounting for 78% of the national import and export overseas container volume.

The geographical locations of the air quality monitoring (AQM) sites in the near-port areas used in this study are shown in Fig. 1. The BS port consists of two AQM sites (BS1 and BS2). For all other ports, the monitoring site closest to each port was selected as the port site. Initially, most air pollutants were routinely recorded at hourly intervals at each AQM site by the Korean Ministry of Environment. A detailed analysis of the particulate and gaseous air pollutants ($PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO) was conducted using hourly values throughout the study period (from November 1, 2017 to December 31, 2020). At the harbor AQM site of the BS2 port, the concentrations of air pollutants were routinely measured since November 2017. In addition, at the BS2 port, the observational datasets of eight water-soluble inorganic ions (Cl^-, SO_4^{2-},

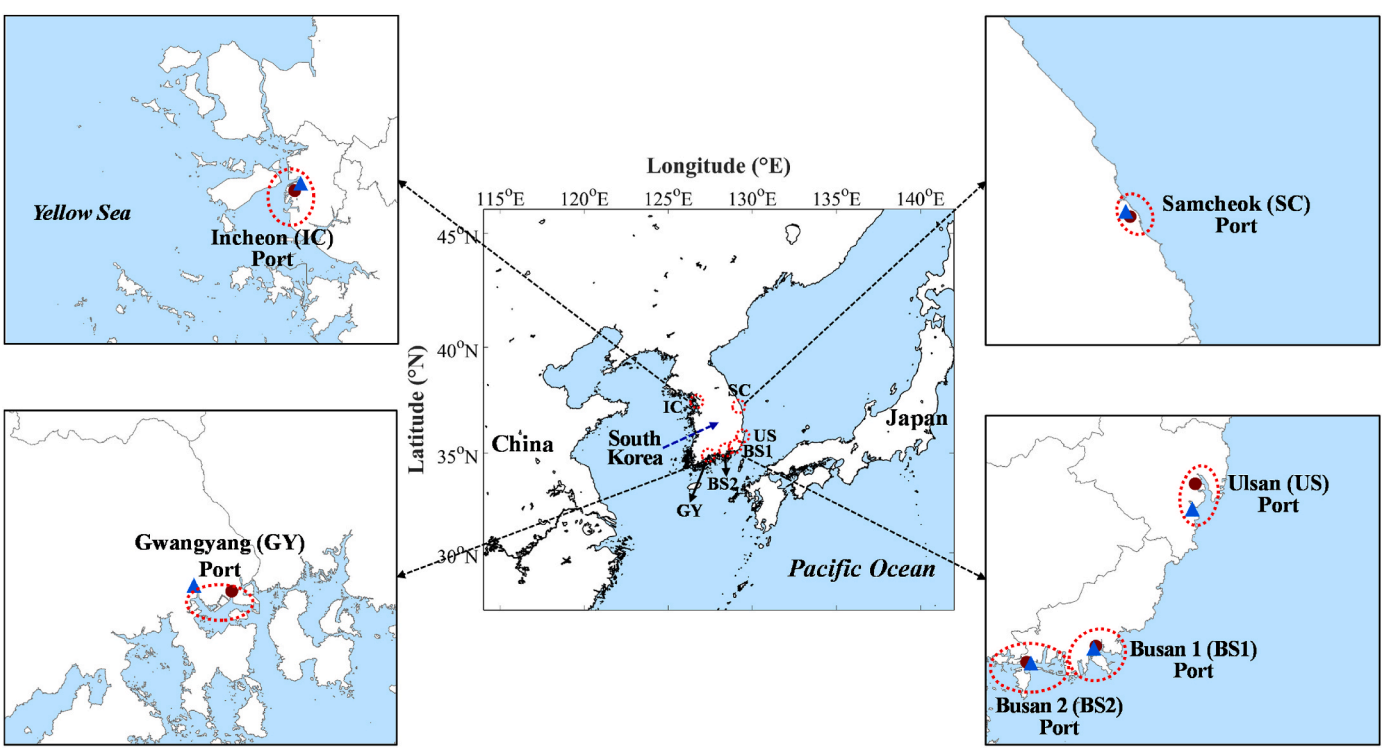


Fig. 1. Geographical locations of six port areas for analysis of port air quality. The AQM (solid circles) and meteorological monitoring sites (solid triangles) closest to each port were selected. In addition, $PM_{2.5}$ components were measured at the port site (BS2) in Busan.

NO_3^-, NH_4^+, Na^+, K^+, Ca^{2+}, and Mg^{2+}) and two carbonaceous species (elemental carbon (EC) and organic carbon (OC)) in $PM_{2.5}$ were routinely measured on an hourly basis since May 2018. The operation of this site is managed by the Busan Institute of Health & Environment (BIHE), which implements quality assurance and quality control for chemical analysis and provides detailed information on the chemical analysis of $PM_{2.5}$. Inorganic ion concentrations were measured by ion chromatography using a URG-9000D-INTEGRION Ambient Ion Monitor (AID) (Dionex, USA). The concentrations of OC and EC were measured using a thermal/optical transmittance carbon aerosol analyzer (Sunset Laboratory Inc.).

To characterize the aerosol properties of the port area, a short-term field campaign was conducted from February 12 to March 3, 2019, at a monitoring station at the AQM site (BS2, 5 m away). A detailed description of the aerosol property measurements is provided in the Supplementary Materials. Gaseous precursors, including nitric acid (HNO_3) and ammonia (NH_3), and the particle size distribution and chemical composition (NO_3^-, SO_4^{2-}, NH_4^+, and BC) of $PM_{2.5}$ were also measured during the field campaign. The chemical composition (ions and BC) and size distribution of PM were analyzed by particle into liquid sampler-ion chromatography, scanning mobility particle sizer (SMPS), and aethalometer. The SMPS (Model 5416, GRIMM Aerosol Technik) was used to measure the number concentration of aerosols based on the particle size by classifying and counting the particles using the electrical properties of particles (Joshi et al., 2017). In the case of BC concentration, values in the 880-nm range were used (Park et al., 2010). Furthermore, twenty-one metallic compounds and elements (Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Ba, Fe, Ni, Cu, Zn, As, Se, Br, and Pb) for $PM_{2.5}$ were measured using an energy dispersive X-ray fluorescence spectrometer (ED-XRF, ARL QUANTX EDXRF Spectrometer, Thermo. Inc., USA). A detailed description of the metallic element measurements was provided by Lee et al. (2020). In addition, detailed information on the chemical properties of aerosols (non-sea-salt sulfate (NSS–SO_4^{2-}) and the oxidation rates of gaseous precursors) is provided in the Supplementary Materials.

Meteorological data, including temperature, relative humidity (RH), wind speed, and wind direction, which were observed at a monitoring station adjacent to the port (Fig. 1), were used to study the meteorological effects on port air quality. To assess the downwind effect of port-related activities (and ship emissions) on air quality, we analyzed the characteristics of pollution wind-rose and differences in the mean concentrations of air pollutants between two different wind sectors, such as prevalent winds from ports (i.e., port wind sector (PS)) and non-port directions (i.e., non-port wind sector (NPS)). The wind directions corresponding to PS at the six ports were as follows: 101–236° (BS1), 191–304° (BS2), 124–236° (GY), 124–169° (US), 169–349° (IC), and 349−146° (SC). In addition, NPS corresponded to all wind directions, excluding PS.

2.2. Regression discontinuity design (RDD)

We applied the statistical RDD tool to evaluate the effect of the IMO 2020 policy on air quality in the harbor areas of Busan (BS1 and BS2 sites). For RDD analysis, hourly datasets of PS at each site were selected and then averaged on a daily basis (high-frequency dataset). As the IMO 2020 policy was officially implemented on January 1, 2020, this day was taken as the cutoff to estimate the effects of the treatment. In this study, we applied a non-parametric estimation method with a bandwidth of one year in RDD because the policy effect could not be fully reflected in the consideration of seasonal effects in a short time period. A detailed explanation of the application of RDD to time series is provided in Hausman and Rapson (2018). The average experimental effect on the cutoff value was estimated using the following equation:

$$Y_i = \beta_0 + \beta_1 Z_i + \beta_2 (X_i - X_c) + \beta_3 Z_i (X_i - X_c) + \lambda C_i + \varepsilon_i \quad (1)$$

where β_0 is the intercept; X_i and Y_i are the day and daily air quality data (SO_2, NO_2, and $PM_{2.5}$), respectively, in time series; β_1 is the estimate of the treatment effect (IMO, 2020 policy); β_2 and β_3 are predicted outcomes (Y_i) from assignment; X_c is the treatment cutoff; Z_i is a dummy variable equal to 0 (zero) for air quality data in 2019 and 1 for air quality data in 2020; C_i is the control variable; and ε_i is a random error term. The interaction between the assignment and treatment variables (the third term on the right-hand side of Eq. (1)) was modeled. In this method, a rectangular kernel was selected as the base option, which gives the same weight to samples in the bandwidth. To improve the reliability of the regression, we selected several control variables C_i, such as wind speed, precipitation, maximum temperature (T_{max}), and minimum temperature (T_{min}), which can influence the air quality (e.g., concentrations). Seasonality was controlled using seasonal dummy variables. The impact of the IMO 2020 policy on air quality was graphically checked using the locally estimated scatterplot smoothing (LOESS) method. LOESS visualizes a continuous function of variable interest (air quality) with respect to the running variable (time). In the LOESS smoothing, α (determining the width of the moving window) was set to 0.5 and a 2nd-order polynomial was used.

3. Results and discussion

3.1. Impact of ship-related activities on port air quality before and after IMO 2020 policy

The air quality of the six ports, in terms of annual mean air pollutant concentrations, before (2018–2019) and after the IMO 2020 policy implementation are summarized in Table 1. The characteristics of air pollutants at the ports (BS1, BS2, GY, US, IC, and SC) varied according to the port emission characteristics. The highest PM (PM_{10} and $PM_{2.5}$) concentrations during 2018–2019 were recorded at the IC port (46.0–46.1 and 22.7–26.1 μg m^{-3}, respectively), followed by the US port (41.7–45.7 and 21.9–28.1 μg m^{-3}, respectively). The ratio of $PM_{2.5}$ to PM_{10} ranged from 0.49 to 0.68 (mean of 0.57) in 2018–2019 and 0.41–0.59 (0.52) in 2020. The contributions of $PM_{2.5}$ emissions from ships to the total emissions in the Ulsan and Incheon cities in 2019 were 21% and 18%, respectively (Fig. S1) (NIER, 2021). Unlike PM, the highest SO_2 concentration was found at the BS2 port (11.1–12.6 ppb), followed by the US port (9.9–10.8 ppb), whereas relatively high NO_2 concentrations were found at IC (29.6–32.8 ppb) and BS ports (29.6–30.3 ppb).

As shown in Table 1, a significant decrease in SO_2 concentrations at the ports was observed from January 1, 2020, owing to the reduction in the sulfur fuel content limit (0.5% m/m) by IMO for ships operating outside designated SECAs. The BS port showed the largest reduction in mean SO_2 concentrations in 2020, which was 52% (BS1) and 56% (BS2) of the 2018–2019 values (i.e., before the implementation of the IMO, 2020 policy). At the other ports, the reductions in SO_2 concentrations (except for SC) were 42% (IC), 31% (GY), and 31% (US). These SO_2 reductions were confirmed by monthly changes in SO_2 concentrations at the six ports during the study period (Fig. 2). Insignificant harbor volume changes at most of the ports (except for BS port) in 2020 supported the findings that the new regulations were responsible for the considerable decrease in SO_2 concentration (Fig. S2 and Table S2). Unlike SO_2, the largest decrease in mean $PM_{2.5}$ concentration was observed at the GY port (40%), followed by the US port (27%), whereas the largest decrease in mean PM_{10} was observed at the IC port (23%), followed by the BS2 port (22%) (Table 1). Different patterns in $PM_{2.5}$ (and PM_{10}) are likely to be related to the dominance of secondary PM production by the gas-to-particle conversion compared to primary PM at the port (Genga et al., 2017; Jeong et al., 2017; Liu et al., 2021). Compared with concentrations over 2018–2019, the largest decrease in mean NO_2 was observed at the IC port (24%), followed by the GY port (17%).

A similar effect of the sulfur content control strategy in ship fuel oil (SECA) on air quality was observed in port areas (Kotchenruther, 2017;

Table 1
Statistical summary of annual mean concentrations of air pollutants and meteorological variables at the air quality monitoring (AQM) sites in the six ports during 2018–2020. The AQM and meteorological monitoring sites are located closest to each port.

	BS		GY	US	IC	SC
	BS1	BS2				
PM_{10} (μgm^{-3})	36.4/36.1/28.9[a]	39.0/36.8/29.4	32.0/33.1/28.2	45.7/41.7/34.3	46.0/46.1/35.3	31.8/31.8/24.9
$PM_{2.5}$ (μgm^{-3})	21.7/22.1/17.4	21.5/22.5/17.5	20.5/18.3/11.7	28.1/21.9/18.0	22.7/26.1/18.3	17.9/17.4/12.9
$PM_{2.5}/PM_{10}$ ratio	0.60/0.61/0.59	0.54/0.60/0.59	0.68/0.54/0.41	0.62/0.53/0.52	0.49/0.57/0.51	0.53/0.53/0.52
SO_2 (ppb)	9.9/8.6/4.4	12.6/11.1/5.2	8.3/5.0/4.3	10.8/9.9/7.1	8.5/8.0/4.8	2.4/2.5/2.5
NO_2 (ppb)	29.9/30.3/27.2	29.6/30.1/29.4	18.3/16.2/14.3	24.7/23.2/21.3	32.8/29.6/23.6	14.4/13.6/11.3
CO (ppb)	391/397/383	378/381/365	495/491/492	628/496/553	523/534/510	378/360/345
O_3 (ppb)	25.6/28.4/25.0	24.2/33.5/29.8	29.0/36.7/33.4	21.8/27.7/29.5	23.5/29.1/30.1	31.2/31.3/26.9
Temp (°C)	15.8/16.4/16.0	15.3/16.0/15.6	14.7/15.2/14.9	14.4/15.5/15.1	12.6/13.2/12.9	13.3/14.3/13.8
RH (%)	59.7/59.3/62.7	62.4/61.9/62.7	69.8/65.8/65.1	68.9/66.6/64.0	65.2/65.3/66.4	59.9/59.1/60.2
Wind speed ($m\ s^{-1}$)	1.0/1.0/1.0	1.6/1.5/1.5	1.4/1.4/1.4	3.4/3.3/3.4	3.0/2.9/3.1	1.8/1.7/1.8
Wind direction[b]	NE/NE/NE	W/W/W	WNW/WNW/WNW	NNE/NNE/NNE	NNW/NNW/NNW	WSW/WSW/WSW

[a] Values in 2018/2019/2020.
[b] Dominant wind direction.

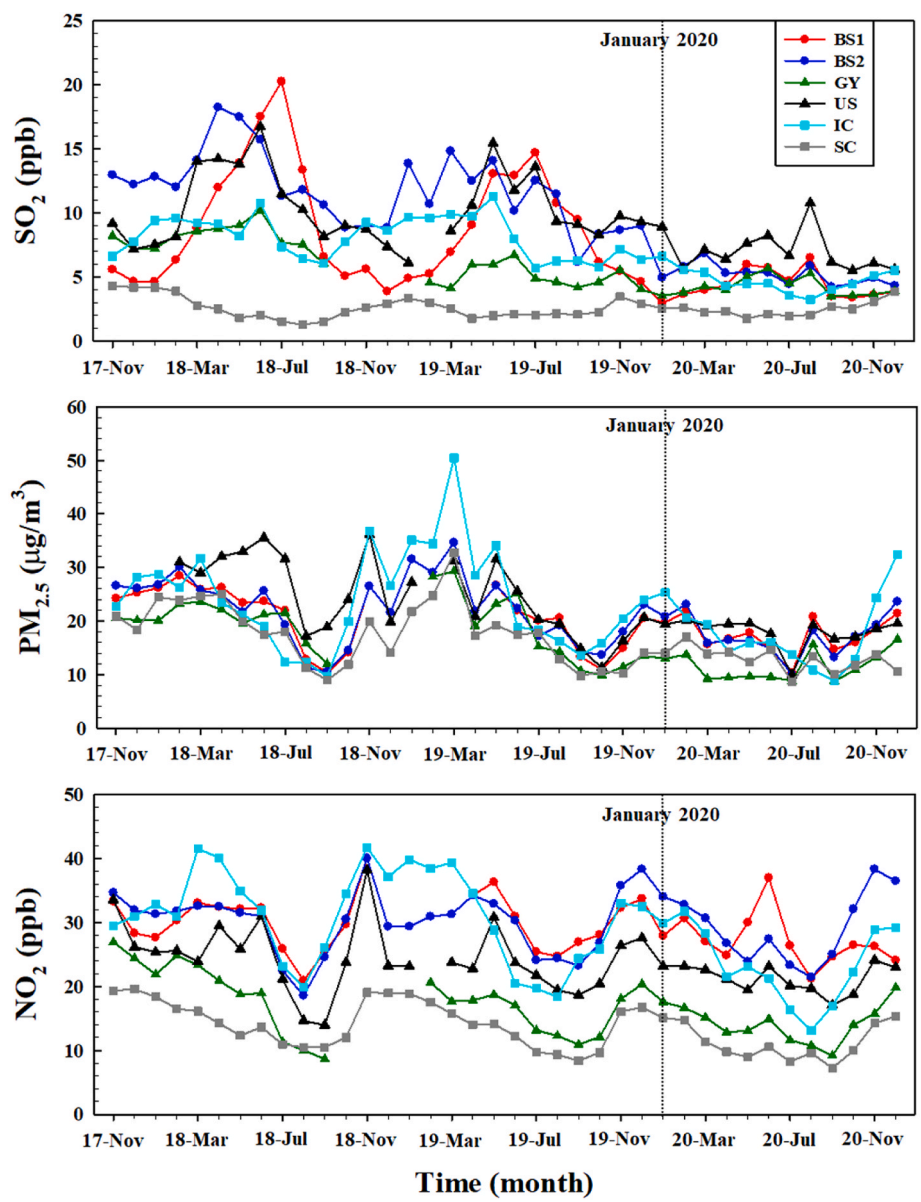


Fig. 2. Monthly variations in concentrations of SO_2, $PM_{2.5}$, and NO_2 in six port areas during study period (November 2017 to December 2020).

Wan et al., 2019; Zhang et al., 2019). In Chinese SECAs, a large reduction in SO_x emissions (e.g., <56%) was observed by changing to < 0.5% residual marine fuel oil during ship hoteling, whereas only insignificant changes in NO_x emissions were reported (Wan et al., 2019). According to an observational study in Shanghai, China, there was a significant decrease in SO_2 concentration (27–55%) at two port sites and urban sites, after the implementation of the domestic SECA policy (Zhang et al., 2019). SO_2 reduction at the Shanghai port was slightly lower than that at the BS port in the present study. Kotchenruther (2017) determined that the annual mean $PM_{2.5}$ concentrations in North America after implementation of SECA regulation decreased by 50% starting in 2010 (below 1% S) and by 74.1% starting in 2015 (below 0.1% S) compared with pre-SECA levels. The reduction in $PM_{2.5}$ concentration (36–43%) due to the IMO 2020 policy at the GY port was comparable to the SECA effect (for below 1% S) on $PM_{2.5}$ in North America. After September 2020, significant SO_2 concentration decreases were observed at most ports owing to the designation of SECA in the major ports of South Korea (Fig. 2).

SO_2 concentrations at the ports were moderately correlated with harbor volumes, with Pearson correlation coefficients (r) of 0.58 (BS1) and 0.41 (BS2). There was no significant monthly variation in harbor volume, except for the BS port (Fig. S2). There were also no significant monthly and daily variations in the total number of incoming and outgoing vessels (Fig. S3). In addition, the SO_2 concentration in the ports was significantly correlated with the relative ship emission strength (r = 0.79, Fig. S4), and this exhibited a higher correlation than those (r = 0.41 and 0.75, respectively) for $PM_{2.5}$ and NO_x. According to the NIER (2021), the contributions of SO_x (sum of SO_2 and SO_3) emissions from ships to total SO_x emissions in Busan and Ulsan cities in 2019 were 75% and 7%, respectively, and the contributions of NO_x (sum of NO and NO_2) emissions from ships to total NO_x emissions in Busan and Incheon cities were 40% and 13%, respectively (Fig. S1). Ships were the predominant source of SO_x emissions in Busan city, whereas industrial processes, electricity generation, and manufacturing were the major emission sources in other cities. In addition, ships were major NO_x emission sources in Busan city, whereas on-road mobile sources accounted for most of the NO_x emissions in other cities. Because the contribution of on-road mobile sources emitted from the port and urban areas in Busan city to $PM_{2.5}$ at the BS2 port was estimated to be significant (49.8%), the impact of urban on-road mobile sources on port $PM_{2.5}$ may be significant (personal communication, E. Jang, 2022). In contrast, the impact of urban on-road mobile sources on port SO_2 might be negligible owing to very low SO_x emissions in urban areas (Fig. S1). Because the emissions of air pollutants from other sources (e.g., factories and buildings) in several Korean cities were negligible (Fig. S1), the impact of these emissions on the port air quality is expected to be insignificant. The higher NO_2 concentration at the IC port can be attributed to traffic emissions and regional air mass transport from the Seoul metropolitan area, including Incheon city. Therefore, SO_2 concentrations in port areas were most likely to be affected by ships and port activities, whereas PM and NO_2 concentrations in these areas could be affected by nearby urban emission sources.

Ship emissions and port facilities at container terminals can significantly affect ambient fine PM levels and air quality, especially in areas near the port. Significant $PM_{2.5}$ emissions (348 ton y^{-1}, 17% of total ship emissions) in 2019 were observed from diesel on-terminal equipment at major freight handling facilities at the BS port, with significant $PM_{2.5}$ emissions (214 ton y^{-1}) from yard tractors (Busan, 2019) (https://busanpa.com/kor/Board.do?mCode=MN1449). The BS2 port accounted for approximately 56% of the total $PM_{2.5}$ emissions from the cargo handling equipment (CHE) of the total BS port. The $PM_{2.5}$ emissions from OGV and CHE at the BS port in 2015 were 555 ton y^{-1} and 347 ton y^{-1}, respectively, indicating a significant emission contribution by CHE at the port (https://busanpa.com/kor/Board.do?mCode=MN1449). Significant $PM_{2.5}$ emission by railway locomotives was also reported at the highly emission-controlled port terminal compared with emissions from port activities, including high-duty vehicles, CHE, and ship emissions (Mousavi et al., 2018).

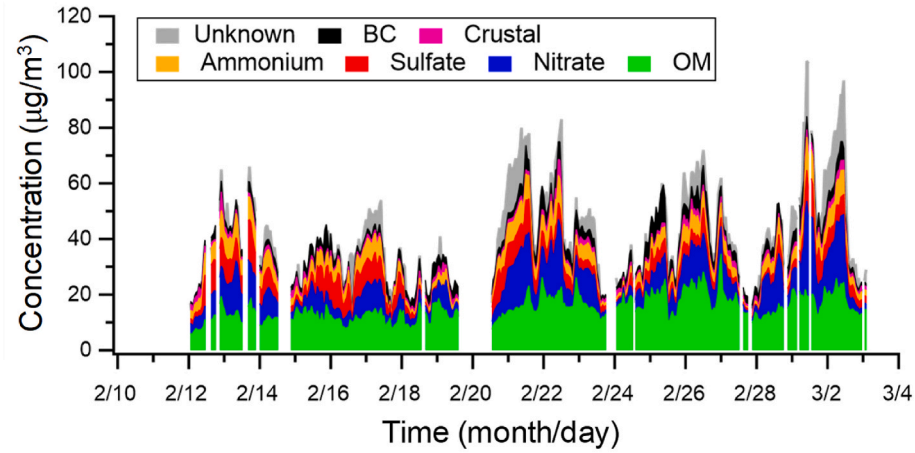
The formation of secondary inorganic aerosols (SIA: NO_3^-, SO_4^{2-}, and NH_4^+) at the BS port was assessed by measuring the sulfate oxidation ratio (SOR) and nitrate oxidation ratio (NOR) (Table S3). The values of SOR and NOR were higher than 0.25 and 0.1, respectively, suggesting the fast potential formation of SIA in the atmosphere (Colbeck and Harrison, 1984; Ohta and Okita, 1990); in contrast, the values at the port air were lower than 0.25 and 0.1, respectively, suggesting the slow formation of SIA in freshly emitted aerosols from port sources. During our study period, the SOR and NOR at the BS port were less than 0.15 and 0.05, respectively. In addition, the IMO 2020 policy implementation showed an increase in the SOR for all seasons and no clear patterns in the NOR during most of the seasons covered during the study period. Despite the reductions in SO_2 and NO_2 at the BS port in 2020, the higher SOR and NOR were likely to be attributed to strong gas-phase chemical conversion (or strong oxidizing capacity) and high RH (Table 1). The high RH favored partitioning into the particle phase and contributed to a heterogeneous pathway (Wang et al., 2005; Shon et al., 2013; Han et al., 2021; Li et al., 2021).

The mean concentrations of $PM_{2.5}$, organic matter (OM), NO_3^-, SO_4^{2-}, NH_4^+, and BC during the intensive field campaign at BS2 port were 39.8 ± 17.9, 15.2 ± 4.6, 8.7 ± 5.8, 4.9 ± 3.2, 4.6 ± 2.3, and 3.5 ± 2.2 μg m^{-3}, respectively (Figs. 3 and S5). The major components of $PM_{2.5}$ were OM (39%), NO_3^- (19.1%), SO_4^{2-} (11.6%), NH_4^+ (10.6%), BC (9.3%), and crustal (4.2%) (Fig. 3). Organics and SIA accounted for a major portion of $PM_{2.5}$ in the atmosphere of the BS2 port. The PMs were generally distributed in a relatively freshly emitted particle size range of 50–60 nm (Fig. S6). The mean concentrations of gas-phase precursors were 34.4 ± 14.8 ppb of NO_2, 8.9 ± 8.5 ppb of SO_2, 3.1 ± 2.3 ppb of NH_3, and 1.5 ± 0.9 ppb of HNO_3 (Fig. S5). In addition, the mean BC_{370nm}/BC_{880nm} ratio, a marker of BC sources, was 1.15 ± 0.14 (Fig. S7). Wu et al. (2007) compared the BC ratios from biofuels and vehicle fuels at 370 and 880 nm wavelengths and reported values of 1.74 for coal, 1.04 for diesel, and 1.14 for gasoline. Yu et al. (2014) compared the BC_{370nm}/BC_{880nm} ratios for biomass burning and non-biomass burning periods and reported the values to be 1.29 and 0.95, respectively. Therefore, the BC_{370nm}/BC_{880nm} ratio obtained in this study was close to the general range of diesel and gasoline, implying the significance of mobile sources and on-terminal equipment at major freight-handling facilities and heavy-duty diesel trucks at the BS port terminals.

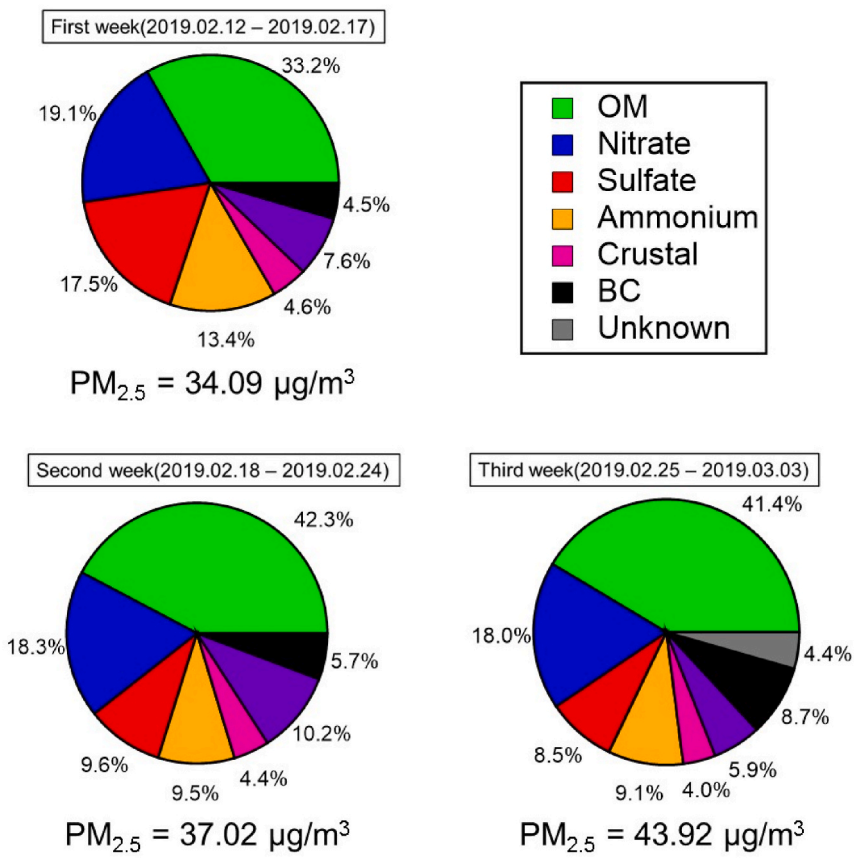
The molar ratio of NH_4NO_3 and $(NH_4)_2SO_4$ appeared mainly in the ammonia-rich range (Fig. S8), indicating a homogeneous reaction (Pathak et al., 2009). The concentration of excess NH_3 at the BS2 port during the field campaign was estimated to identify the atmospheric conditions in the SIA formation for $PM_{2.5}$. If the calculated value of excess NH_3 was less than 0 (zero), it indicates that nitrate production in $PM_{2.5}$ was in the NH_3-limited state; if the value was greater than 0, it indicated an HNO_3-limited (or NH_3-rich) state. The definition of excess NH_3 is provided in the Supplementary Materials. The mean concentration of excess NH_3 during this study period was 0.075 ± 0.128 μmol m^{-3}, indicating an NH_3-rich condition (Fig. S5). This showed that the environment had sufficient NH_3 to form $(NH_4)_2SO_4$ and NH_4NO_3 by neutralizing H_2SO_4 and HNO_3 in the study area. High concentrations of NH_3 and HNO_3 can provide favorable physicochemical conditions for NH_4NO_3 (Seinfeld and Pandis, 2016).

3.2. Impact of air mass pathways on port air quality before and after IMO 2020 policy

Air pollution signals, including ship emissions at the port, can be clearly determined in terms of air mass transport pathways (e.g., wind direction). The port air quality was analyzed in terms of two different wind sectors (PS vs. NPS) (Table 2). At the BS ports, SO_2 concentrations in PS before the IMO 2020 policy (2018–2019) were higher than those in



(a)



(b)

Fig. 3. (a) Temporal variations in concentrations of $PM_{2.5}$ components and (b) chemical composition of $PM_{2.5}$ at BS2 port during 2019 field campaign.

NPS by a factor of 2.6–3.6, which were higher (a factor of 3–4) than the urban mean (not shown). Similarly, both PM and NO_2 concentrations in PS at the BS ports were higher than those in NPS by a factor of 1.1–1.5 and 1.1–1.3, respectively. The PM and NO_2 concentrations in PS at the other ports (except for IC) were also higher than those in NPS, although the difference was smaller compared to SO_2 concentrations. The large increase in SO_2 levels in PS from ship emissions was confirmed by the simultaneous increase in the concentrations of vanadium (V) and Ni (Fig. S9), which are markers for aerosols from heavy oil combustion (e.g., ship engines) (Ault et al., 2009; Schembari et al., 2014). However, ship emissions at the SC port were not significant because the site was selected as a control site (minimum ship impact).

The concentrations of key pollutants (e.g., SO_2, NO_2, and $PM_{2.5}$) at all ports decreased significantly after the IMO 2020 policy, and the magnitude of the concentration decrease was higher in PS than in NPS. For instance, the mean SO_2 concentrations in PS at the BS port after the IMO 2020 policy reduced by 69% and 68% at the BS1 and BS2 ports, respectively, compared with those in 2018–2019. In other major ports, the differences in SO_2 concentrations in PS before and after the IMO 2020 policy ranged from 23% (GY) to 51% (IC), with a mean of 35%. For

Table 2
Statistical summary of the concentrations of air pollutants at the AQM sites in the six ports for port wind (PS) and non-port wind sectors (NPS) during 2018–2020.

	BS						GY			US			IC			SC		
	BS1			BS2														
	PS[a]	NPS[b]	PS/NPS[c]	PS	NPS	PS/NPS	PS	NPS	PS/NPS	PS	NPS	PS/NPS	PS	NPS	PS/NPS	PS	NPS	PS/NPS
PM_{10}	45.2/	33.8/	1.34/	46.1/	33.0/	1.40/	35.1/	30.1/	1.17/	55.7/	44.6/	1.25/	49.8/	44.9/	1.11/	29.4/	33.3/	0.88/
	39.3	35.2	1.12	41.1	33.4	1.23	36.4	31.5	1.15	46.4	41.0	1.13	50.6	41.3	1.22	30.3	32.7	0.93
(μgm^{-3})	/30.9[d]	/28.4	/1.09	/31.0	/27.9	/1.11	/31.2	/27.5	/1.13	/37.7	/33.8	/1.12	/36.4	/33.2	/1.10	/25.1	/24.7	/1.02
$PM_{2.5}$	26.5/	20.3/	1.31/	26.3/	17.5/	1.50/	22.0/	19.7/	1.11/	30.8/	27.8/	1.11/	23.4/	21.8/	1.07/	16.4/	18.8/	0.87/
	23.4	21.8	1.07	25.4	20.3	1.25	19.8	17.5	1.13	22.3	21.8	1.02	28.5	22.9	1.25	16.1	18.2	0.89
(μgm^{-3})	/17.2	/17.3	/1.00	/18.4	/16.5	/1.12	/12.5	/11.5	/1.09	/20.0	/17.7	/1.13	/18.0	/18.3	/0.98	/12.4	/13.2	/0.94
$PM_{2.5}$/	0.59/	0.60/	0.99/	0.57/	0.51/	1.12/	0.66/	0.69/	0.96/	0.59/	0.63/	0.95/	0.45/	0.45/	0.99/	0.52/	0.54/	0.95/
PM_{10}	0.61	0.61	0.99	0.62	0.59	1.06	0.54	0.54	1.01	0.48	0.54	0.90	0.57	0.54	1.06	0.51	0.53	0.95
Ratio	/0.56	/0.61	/0.92	/0.60	/0.60	/1.00	/0.40	/0.42	/0.97	/0.53	/0.52	/1.01	/0.50	/0.55	/0.90	/0.50	/0.54	/0.93
SO_2 (ppb)	20.8/	6.7/6.4	3.12/	20.7/	5.8/5.3	3.56/	10.2/	7.2/4.2	1.42/	12.6/	10.6/	1.20/	9.5/9.1	7.3/6.4	1.30/	2.1/2.4	2.6/2.5	0.79/
	16.5		2.58	18.3		3.43	6.6		1.57	10.7	9.8	1.09			1.41			0.96
	/5.7	/3.9	/1.46	/6.3	/4.1	/1.54	/6.2	/3.7	/1.68	/7.8	/7.0	/1.11	/4.6	/5.0	/0.92	/2.2	/2.7	/0.81
NO_2 (ppb)	35.8/	28.2/	1.27/	33.2/	26.7/	1.24/	20.2/	17.3/	1.17/	27.5/	24.4/	1.13/	32.2/	33.6/	0.96/	12.2/	15.8/	0.77/
	33.8	29.4	1.15	32.0	28.7	1.12	17.5	15.6	1.13	24.1	23.1	1.04	28.7	30.9	0.93	12.2	14.4	0.84
	/31.8	/25.8	/1.23	/31.0	/27.8	/1.12	/16.6	/13.8	/1.20	/25.9	/20.7	/1.25	/21.5	/26.8	/0.80	/10.2	/12.0	/0.85
CO (ppb)	366/	398/	0.92/	388/	370/	1.05/	527/	477/	1.10/	536/	638/	0.84/	477/	579/	0.83/	346/	397/	0.87/
	365	406	0.90	391	374	1.05	515	478	1.08	409	508	0.80	499	586	0.85	343	371	0.93
	/352	/393	/0.90	/356	/371	/0.96	/553	/479	/1.15	/506	/557	/0.91	/466	/577	/0.81	/328	/358	/0.92
O_3 (ppb)	28.3/	24.8/	1.14/	24.3/	24.2/	1.01/	32.9/	26.7/	1.23/	25.6/	21.4/	1.20/	27.4/	18.8/	1.45/	34.5/	29.2/	1.18/
	31.2	27.6	1.13	34.8	32.5	1.07	43.3	33.4	1.30	35.0	26.7	1.31	34.0	21.9	1.55	33.0	30.2	1.09
	/24.9	/24.8	/1.00	/29.2	/30.2	/0.97	/39.2	/31.8	/1.23	/35.6	/28.7	/1.24	/34.6	/22.9	/1.51	/28.3	/26.2	/1.08

[a] PS: winds from port direction, BS1 101–236°, BS2 191–304°, GY 124–236°, US 124–169°, IC 169–349°, SC 349−146°.
[b] NPS: all wind directions excluding PS.
[c] The ratio of PS/NPS dataset.
[d] Values in 2018/2019/2020.

PS at the BS port, PM (PM_{10} and $PM_{2.5}$) decreased by 21–35% and NO_2 by 3–11% compared with the concentrations before the IMO 2020 policy implementation. In general, sulfur-related pollutant species, such as SO_2 and $PM_{2.5}$, can seriously affect human health (e.g., disease and premature deaths) and economic costs in urban polluted areas (Pope and Dockery, 2006; Wu et al., 2020; Han et al., 2021). According to Wu et al. (2020), the total mean mortality and its economic cost owing to the exposure of excessive SO_2 concentrations in Beijing, China was estimated to be 884 cases per year and 384.89 million RMB Yuan, respectively, and the economic cost accounted for 0.0014% of the total annual GDP of Beijing. In addition, Tian et al. (2013) reported that the 2015 emission reduction scenario in China resulted in a decrease in SO_2 and NO_x emissions, which further reduced economic costs. Although the target area of this study is different from that in previous studies, the IMO 2020 policy effect was likely to affect port air quality, including decreases in SO_2 emissions and concentrations of sulfur-related pollutant species, as well as decreases in health impact and economic cost in the study area. If a global sulfur cap of 0.1% m/m is imposed outside the SECAs (open ocean), the magnitude of these decreases will increase.

To assess the impact of ship emissions and clean marine sources on port air quality, we further investigated the characteristics of $PM_{2.5}$ components, which were obtained from the BIHE measurement at BS2 port from May 2018 to December 2020, and categorized them into two wind sectors (PS vs. NPS) (Table S4). Unlike the criteria air pollutants, most $PM_{2.5}$ components (except for EC and SO_4^{2-}) showed slight differences in their concentrations between PS and NPS. From January to December 2019 (before the IMO, 2020 policy), the EC and SO_4^{2-} concentrations were significantly higher in PS than in NPS (48–51% increase), thereby suggesting a strong influence of port-related anthropogenic sources. The elevated concentrations of BC and PM in the downwind area of ports were observed during periods of increased port activity (Steffens et al., 2017). Additionally, the concentrations of NH_4^+ and OC in PS were slightly higher than those in NPS (24–27% increase). The differences in the concentrations of NO_3^-, Na^+, K^+, and Ca^{2+} between PS and NPS were insignificant (≤l0%). In contrast, Cl^- and Mg^{2+} concentrations in PS decreased by approximately 30% and 8%, respectively, compared with those in NPS, probably owing to more acidic aerosols in PS (Sarin et al., 2010).

Since the implementation of the IMO 2020 policy, the concentrations of most $PM_{2.5}$ components (except for Cl^- and Mg^{2+}) at the BS2 port decreased, ranging from 8% (EC) to 76% (Na^+), with slightly larger decreases in PS (13–79%) than in NPS (1.4–76%) (Table S4). The IMO 2020 policy controls the sulfur content in fuel oil (or sulfur emission control), and thus, the concentrations of pollutant species related to sulfur can decrease. Because air pollutants emitted from various sources in PS are transported to the air quality sites in the port areas, their concentrations in PS can be higher than those in NPS. Therefore, port-related anthropogenic sources can affect the concentrations of their emission markers (e.g., EC and SO_4^{2-}). In addition, the sulfur content of the fuel oil caused the difference in the concentrations before and after the IMO 2020 policy. For instance, the fractions (55% and 57%) of SO_4^{2-} and EC in PS in the total concentrations of PS and NPS in 2020 were slightly lower (60% and 60%, respectively) than those in 2019 (before the IMO, 2020 policy). On the other hand, the differences in the concentrations of $PM_{2.5}$ components, except for EC and Cl^-, between PS and NPS in 2020 were insignificant compared with those in 2019. Among the $PM_{2.5}$ components, the four ions of Na^+, Mg^{2+}, Ca^{2+}, and K^+, which are the major ions in seawater and sea-salt aerosols, showed no difference in the concentrations between PS and NPS. The contribution of the clean marine aerosol component to the observed aerosol mass was assessed by estimating the sea-salt SO_4^{2-} content. As shown in Fig. S10, the aerosol at the BS2 port was almost entirely affected by anthropogenic sources, which was supported by a small sea-salt sulfate fraction (~4%) in the observed sulfate aerosol. The maximum NSS-SO_4^{2-} fraction was observed in the W wind sector of PS.

Ship emissions account for 24% of the total EC emissions in Busan (NIER, 2019). EC emissions can also be attributed to tailpipe exhaust from heavy-duty diesel vehicles and on-terminal equipment at major freight handling facilities in the BS port. Therefore, both ship engines and on-terminal equipment are significant sources of EC emissions at ports. Emissions from container ships at berth showed a significant fraction of EC (36%), EC-OC mixtures (34%), and Na-rich particles (21%) (Xiao et al., 2018). High OC concentrations may be related to the production of secondary OC owing to the emission of semi-volatile organic compounds from on-road mobile sources in harbors (Shon et al., 2012). In addition, NH_4^+ levels in PS in our study suggested the significance of on-road mobile sources of on-terminal heavy-duty vehicles (not shown).

3.3. Statistical analysis of the effect of IMO 2020 policy on port air quality

RDD in time series examines the impact on ambient air quality by estimating the change in air pollutant concentrations at the time the policy occurred (cutoff) (Davis, 2008; Chen and Whalley, 2012; Gallego et al., 2013; Zhang et al., 2020). This method can be applied to ambient air quality data available at daily or hourly intervals over a long time period (e.g., years). It can be also applied to environmental policy in which many potential time-varying confounders are assumed to change smoothly across the date of the policy change (Hausman and Rapson, 2018). Therefore, statistical analysis using RDD is essential for demonstrating the robustness of our observational analysis results of the IMO 2020 policy effect. The RDD analysis on port air quality during the periods before and after implementation of the IMO 2020 policy was performed at the BS ports only (BS1 and BS2). In summary, the sharp decrease in SO_2 concentration at the cutoff (January 1, 2020) for the BS2 port was 57% (Fig. 4), which was very similar to the observational analysis result (56%) and that for the BS1 port was 38% (52% in observational analysis). In contrast to the observational analysis results (Section 3.1), the concentrations of NO_2 and $PM_{2.5}$ at the BS1 and BS2 ports did not decrease after the implementation of the IMO 2020 policy. As such, we can infer that the policy had a direct effect on SO_2 level due to fuel sulfur content drop but an insignificant indirect effect on NO_2 and $PM_{2.5}$ levels at the harbor. A potential reason for the increase in $PM_{2.5}$ concentration (49%) after the implementation of the IMO 2020 policy at the BS1 port might be related to enhanced ammonium nitrate (NH_4NO_3) formation owing to SO_2 reduction in an ammonia-rich environment, as was derived from the 2019 field campaign conducted in the study area (Fig. S5). Interestingly, the NH_4^+ concentration at the BS2 port decreased significantly (57%) after the implementation of the IMO 2020 policy. The SO_2 and NH_4^+ concentration reduction (both 57%) is highly likely to be related to the secondary formation of ammonium sulfate ($(NH_4)_2SO_4$) through gas (e.g., SO_2 and NH_3) to particle conversion.

In general, RDD robustness was checked by the McCrary test, which is a useful diagnostic tool for assessing the internal validity of an RDD (Jacob et al., 2012). However, when time is assigned as a running variable, discontinuities in both the conditional density of the running variable and the outcome variable (i.e., air quality data) cannot be checked because of the uniform density of the time variable. Thus, the robustness of our results was checked by the bandwidth choice between full and half-bandwidth, placebo test, and kernel choices (uniform (i.e., rectangular) vs. triangular). Falsification (or placebo) tests were performed to determine whether the casual impact of air quality change was caused by the assignment rule (IMO, 2020 policy) with cutoffs on December 1, 2019 and February 1, 2020. The LATEs (β_1) difference for different bandwidths were not insignificant (e.g., −7.55 vs. −7.20) (Table 3 and Fig. 4). The LATEs discontinuity at the cutoff for two different placebo cutoffs were not likely to be present, especially for cutoff on February 1, 2020 (not shown). The SO_2 LATEs were also not significantly different for rectangular and triangular options in the kernel function at the BS2 port (e.g., −7.55 and −7.11, respectively).

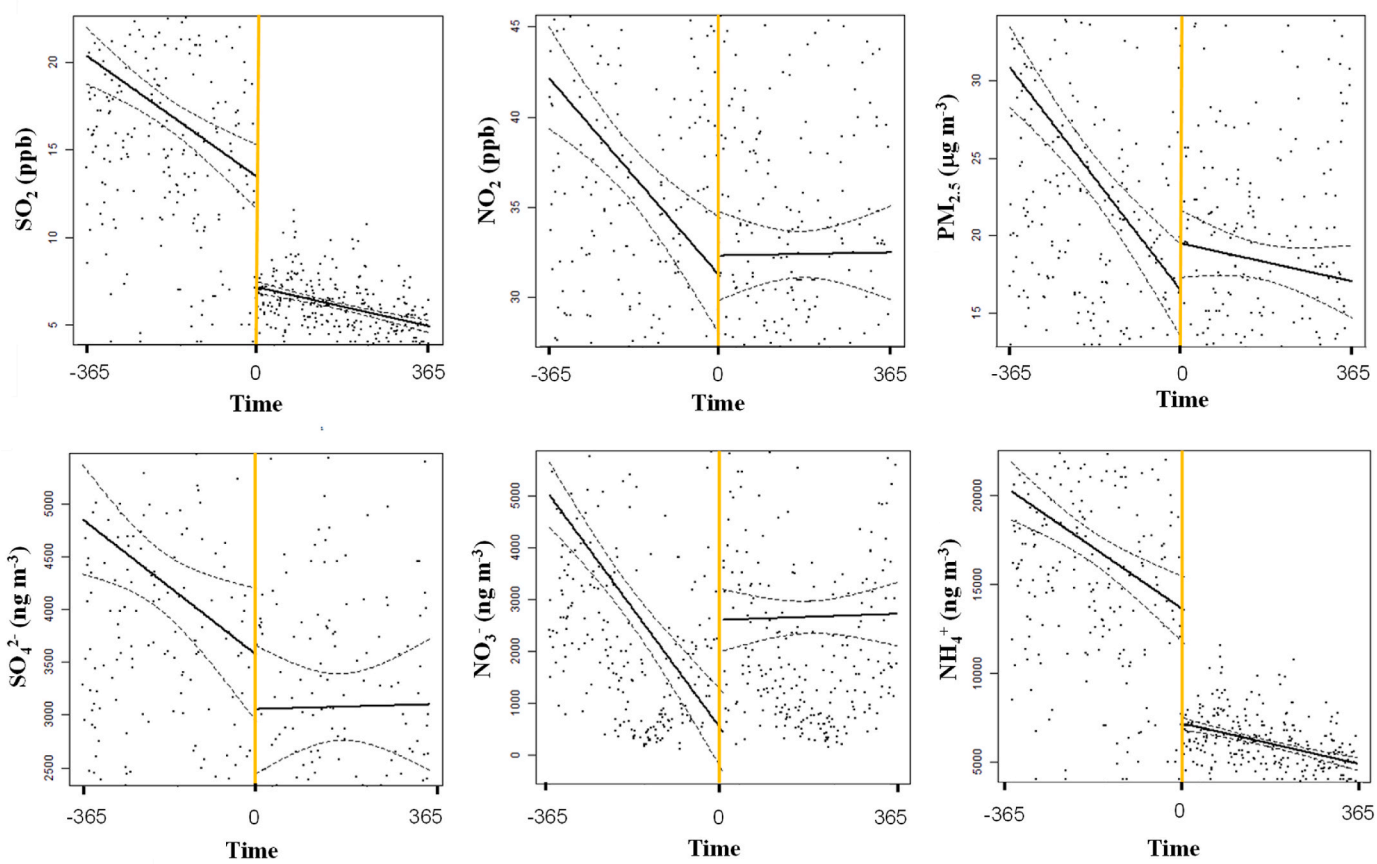


Fig. 4. RDD results for SO_2, NO_2, $PM_{2.5}$, and SIA (SO_4^{2-}, NO_3^-, and NH_4^+) in $PM_{2.5}$ at BS2 port.

Table 3
Statistical summary of average treatment effects (β_1) at the cutoff for the PS dataset at the BS2 port due to IMO 2020 policy.

	BS2						BS1		
	SO_2 (ppb)	NO_2 (ppb)	$PM_{2.5}$ (μg m^{-3})	SO_4^{2-}(μg m^{-3})	NO_3^-(μg m^{-3})	NH_4^+ (μg m^{-3})	SO_2 (ppb)	NO_2 (ppb)	$PM_{2.5}$ (μg m^{-3})
Bandwidth (365)[a]	−7.55***	−0.92	2.07	0.16	0.75	−7.67***	−2.30***	−4.04	7.14*
Half-bandwidth (182.5)	−7.20***	−0.65	2.83	−0.46	0.77	−7.20***	0.77	0.15	4.81

For BS2, The number of case = 618 for full bandwidth and 307 for half-bandwidth.
***$p < 0.001$; **$p < 0.01$; *$p < 0.05$.
[a] The number in parenthesis is bandwidth. Kernel function: rectangular.

The continuity tests of predetermined variables of meteorological conditions, including T_{max}, T_{min}, wind speed, RH, and precipitation, which are key factors for controlling air quality, were performed using LOESS (Fig. S11). There were no discontinuities (drop) of these variables before and after the implementation of the IMO policy, suggesting that the decrease in SO_2 concentration was not driven by meteorological conditions.

3.4. Confounding factor

Coronavirus disease (COVID-19) was first identified in Wuhan, China in December 2019, and the World Health Organization (WHO) declared it an international public health emergency on January 30, 2020, and a pandemic on March 11, 2020. Thus, the COVID-19 pandemic could be a confounding factor in the effect of the IMO2020 policy. The COVID-19 pandemic showed significant NO_2 concentration changes in China collected from the Tropospheric Monitoring Instrument (TROPOMI) onboard ESA's Sentinel-5 satellite (Dutheil et al., 2020). In central China, NO_2 emissions decreased by as much as 30% in February 2020 (Dutheil et al., 2020; NASA, 2020). However, the COVID-19 pandemic did not significantly impact port air quality in South Korea compared with the application of the IMO 2020 policy. Unlike central China, there was no significant difference in tropospheric NO_2 column concentration between February 2019 and February 2020 as well as in other months (March−June 2019 and 2020) (http://www.temis.nl/airpollution/no2.html, Fig. S12).

To estimate the effect of ship emission variations by the COVID-19 pandemic outbreak on port air quality in South Korea, the number of ships entering and departing ports before and after the COVID-19 outbreak was analyzed for the period from 2019 to 2020 (Fig. S13). There were no significant differences in the number of ships entering and departing ports during these periods. For instance, compared with 2019, the number of ships entering and departing the BS port daily in 2020 was reduced by 5.5% and 5.2%, respectively. In addition, the number of cold ironing facilities (or alternative maritime power (AMP)) at the BS, GY, and IC ports in 2020 was negligible, accounting for less than 0.05% of the total number of ships entering and departing ports (Port-MIS, https://new.portmis.go.kr/portmis/websquare/websquare.jsp?w2xPath=/portmis/w2/main/intro.xml). Therefore, the significant impact of ship emissions on port air quality can be attributed to

significant decreases in SO_2 concentrations following reductions in the sulfur fuel content limit and not the COVID-19 outbreak or the use of AMP.

4. Summary and conclusions

In this study, we investigated the impacts of ship traffic, harbor activities, and emission control measures (IMO, 2020) on port air quality based on observational and statistical analyses (RDD) using port AQM concentration data from South Korea from November 2017 to December 2020. The characteristics of the key pollutants at the ports varied according to the port emission characteristics. The concentration of SO_2, a strong port-related signal, was the highest at the BS port (international hub port), indicating that its main source was ships, whereas NO_2 and $PM_{2.5}$ concentrations were mainly affected by on-road mobile sources. Port air quality showed the properties of freshly emitted aerosols according to the oxidation ratio of sulfur and nitrogen oxides and BC ratios. The strong impact of anthropogenic sources related to harbor activities on port air quality was confirmed by air mass pathway analysis (PS vs. NPS) with SO_2 enhancement by a factor of 2.6–3.6 at the BS ports.

The observational analysis of the effect of IMO 2020 showed the largest reduction in SO_2 (a mean of 54%) and relatively low reductions in $PM_{2.5}$ and NO_2 (20% and 6%, respectively) at the two BS ports (BS1 and BS2). Differences in the reduction of key pollutants are likely related to the competition between the primary and secondary production of these pollutants. The strong IMO 2020 effect on the concentration of a key pollutant (SO_2) at the BS ports obtained from the observational analysis was confirmed by RDD, suggesting a strong direct impact of SO_2 levels from lower fuel sulfur content. In contrast to the observational analysis, the statistically insignificant effect of IMO 2020 on $PM_{2.5}$ and NO_2 suggested an insignificant indirect effect on NO_2 and $PM_{2.5}$ levels at the ports. Interestingly, a significant reduction in the NH_4^+ concentration at the BS2 port suggests the significance of secondary aerosol formation in port air quality. Therefore, more studies on the indirect effect of the IMO 2020 policy on secondary $PM_{2.5}$ formation using 3-D chemical transport models are required. Furthermore, an in-depth analysis of the impact of SECA designation on port air quality will be conducted in future studies.

CRediT authorship contribution statement

Sang-Keun Song: Writing – original draft, Conceptualization, Investigation, Formal analysis, Writing – review & editing, Validation. **Zang-Ho Shon:** Conceptualization, Methodology, Supervision, Writing – review & editing. **Soo-Hwan Moon:** Formal analysis, Software, Visualization. **Tae-Hyoung Lee:** Data collection and analysis, Validation. **Heon-Sook Kim:** Formal analysis, Software. **Se-Hwa Kang:** Data collection, Formal analysis. **Gee-Hyeong Park:** Data share, Validation. **Eun-Chul Yoo:** Data share, Validation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2022.131298.

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