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Technical Information

Analysis of the National Air Pollutant Emissions Inventory (CAPSS 2018) Data and Assessment of Emissions Based on Air Quality Modeling in the Republic of Korea

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ABSTRACT According to the 2018 National Air Pollutant Emissions Inventory (NEI), air pollutant emissions in the Republic of Korea comprised 808,801 tons of CO, 1,153,265 tons of NO_X, 300,979 tons of SO_X, 617,481 tons of TSP, 232,993 tons of PM_{10}, 98,388 tons of $PM_{2.5}$, 15,562 tons of black carbon (BC), 1,035,636 tons of VOCs, and 315,975 tons of NH_3. As for national emission contributions to primary $PM_{2.5}$ and PM precursors (NO_X, SO_X, VOCs, and NH_3), major source categories were the road sector for NO_X, the industry sector for SO_X and $PM_{2.5}$, and the everyday activities and others sector for VOCs and NH_3. In the case of emissions by region, the largest amount of NO_X was emitted from the Seoul Metropolitan Areas (SMA; Seoul, Incheon, and Gyeonggi-do, hereafter SMA) and the largest amounts of SO_X, $PM_{2.5}$, VOCs, and NH_3 were from the Yeongnam region. A 3D chemical transport modeling system was used to examine the uncertainty of the national air pollutant emissions based on the National Emission and Air Quality Assessment System (NEAS). Air quality was simulated using CAPSS 2018, and the simulation data were compared with observed concentrations to examine the uncertainties of the current emissions. These data show that emissions from five si (cities) (Pohang, Yeosu, Gwangyang, Dangjin, and Ulsan) need to be improved. Most of all, it is necessary to examine the emissions from places of business that use anthracite, which is the major $PM_{2.5}$ emission source, as fuel in these areas.

KEY WORDS NEI, CAPSS, CMAQ, NEAS, $PM_{2.5}$

1. INTRODUCTION

The government of the Republic of Korea announced the Comprehensive Measures on Fine Dust Management that aims to reduce particulate matter (PM) emissions by 30% (MOE, 2017), and implemented the strengthened plan on fine dust management for emergency and on a regular basis, which includes emergency reduction measures conducted during the periods recording high PM concentrations (MOE, 2018). However, the annual mean atmospheric concentration of $PM_{2.5}$ in 2018 was 23 μg/m^3, which exceeded the criterion of Korea (15 μg/m^3)

(MOE, 2019). Evidently, despite these aggressive reduction efforts of the government, atmospheric $PM_{2.5}$ concentrations were not significantly reduced and high PM concentrations were not mitigated while public awareness is low. Therefore, the Comprehensive Plans for Fine Dust Management and plan on air Environment Management by Region (SMA, Central area, Southern area, and Southeast area) were established and implemented (Kim *et al.*, 2022; MOE, 2020; MOE, 2019).

Since the characteristics of pollutant emission differ by area, it is necessary to identify the major emission sources and analyze their emission contributions to effectively improve PM emissions (Bae *et al.*, 2021). Air pollutant emissions have different characteristics in different regions depending on the topography and industrial structure. For example, SMA has the largest amounts of car-related pollutants in Korea as it has 50% of the country's total population and cars; whereas, Gangwon-do's annual air pollutant emissions are relatively low because of small population and underdevelopment of industrial complexes, which is the result of its mountainous topography (NAIR, 2021). In recent years, research has been conducted on the PM concentrations and emission status considering such regional characteristics (Gong *et al.*, 2021; Hwang *et al.*, 2021), and mutual impacts among neighboring areas, caused by PM emissions, have been analyzed (Kim *et al.*, 2021a, b, c; You *et al.*, 2020).

NAIR assesses and publishes the emissions of 9 air pollutants (CO, NO_X, SO_X, TSP, PM_{10}, $PM_{2.5}$, black carbon [BC], VOCs, and NH_3) for 17 dos (provinces) and metropolitan cities and 250 si (city), gun (county), gu (district) every year (note: Emissions from the sea were managed separately) (Choi *et al.*, 2021). Based on this, the central and local governments need to establish customized PM reduction measures to protect the health and property of local residents and minimize the economic loss of industries.

In this study, the 17 dos (provinces) and metropolitan cities were classified into 5 regions (SMA, Gangwon region, Chungcheong region, Honam region, and Yeongnam region) and changes in air pollutant emissions by region were analyzed using the 2018 national air pollutant emission estimation results. In addition, the uncertainty of domestic emissions was examined by region and pollutant through a comparison between the simulated concentrations using 3D chemical transport model with ground level observed concentrations.

2. NATIONAL AIR POLLUTANT EMISSION ESTIMATION METHOD AND IMPROVEMENTS

As for national air pollutant emissions, the measurement-based emission data of a tele-monitoring system (TMS) were utilized, as it was in previous studies (Choi *et al.*, 2021; Choi *et al.*, 2020; Yeo *et al.*, 2019), or the emissions of 9 pollutants (e.g., $PM_{2.5}$, NO_X, and SO_X) were estimated in 13 first-level categories, 56 second-level categories, and 240 third-level categories by applying

Table 1. Improvements in the emission estimation method.

Category	Improvement
Activity data	<Road transport> – A change in the method of counting the number of registered cars aged 10–15 years (integrated model year counting → individual model year counting) • (Before change) 10 to < 15 years • (After change) < 15 years, < 14 years, < 13 years, < 12 years, and < 11 years <Non-road transport> – (Construction equipment) An increase in the maximum car age subjected to the deterioration rate from 20–30 years – (Marine ships) An improvement in fuel consumption for passenger ships and fishing boats as well as subdivided criteria for the application of the sulfur content by oil type → Detailed classification of the fuels used for each ship <Agriculture> – An improvement in the method of counting livestock population • (Before change) Collect the latest information on the number of livestock population in as of fourth quarter • (After change) Collect the latest information on the number of livestock population as of the first, second, third, and fourth quarters

approximately 30,000 emission factors using approximately 300 statistical data from approximately 150 related organizations (e.g., pollutant-emitting places of business, and those related to transportation and meteorology) as activity data (NAIR, 2022). There were improvements in emission estimation method compared to 2017; The way to collect the activity data from road transport, non-road transport, and agriculture were improved. The details are as follows (NAIR, 2021) (Table 1).

3. 2018 NATIONAL AIR POLLUTANT EMISSION ESTIMATION RESULTS

3. 1 National Air Pollutant Emissions

In the 2018 NEI, the national emissions of air pollutants comprised 808,801 tons of CO; 1,153,265 tons of NO_X; 300,979 tons of SO_X; 617,481 tons of TSP; 232,993 tons of PM_{10}; 98,388 tons of $PM_{2.5}$; 15,562 tons of BC; 1,035,636 tons of VOCs; and 315,975 tons of NH_3 (Table 2).

The emission contributions of different emission

Table 2. 2018 air pollutant emissions and contributions by first-level category of emission sources. (Unit: metric tons/year)

Source category	CO	NO_X	SO_X	TSP	PM_{10}	$PM_{2.5}$	BC	VOCs	NH_3
Total	808,801 100.0%	1,153,265 100.0%	300,979 100.0%	617,481 100.0%	232,993 100.0%	98,388 100.0%	15,562 100.0%	1,035,636 100.0%	315,975 100.0%
Energy production	69,972 8.7%	104,420 9.1%	65,868 21.9%	4,305 0.7%	3,975 1.7%	3,308 3.4%	405 2.6%	9,161 0.9%	1,626 0.5%
Non industry	58,172 7.2%	87,599 7.6%	16,566 5.5%	1,439 0.2%	1,269 0.5%	890 0.9%	172 1.1%	2,936 0.3%	1,414 0.4%
Manufacturing industry	20,060 2.5%	168,967 14.7%	78,867 26.2%	117,150 19.0%	68,315 29.3%	35,099 35.7%	753 4.8%	3,579 0.3%	737 0.2%
Industrial processes	27,866 3.4%	57,020 4.9%	107,353 35.7%	11,975 1.9%	6,758 2.9%	5,189 5.3%	15 0.1%	188,247 18.2%	45,981 14.6%
Energy transport and storage								30,770 3.0%	
Solvent use								547,353 52.9%	
Road transport	213,568 26.4%	406,227 35.2%	217 0.1%	8,858 1.4%	8,858 3.8%	8,149 8.3%	4,935 31.7%	43,658 4.2%	3,322 1.1%
Non-road transport	195,020 24.1%	307,942 26.7%	29,831 9.9%	17,236 2.8%	17,232 7.4%	15,981 16.2%	7,014 45.1%	67,867 6.6%	126 0.04%
Waste	1,954 0.2%	12,492 1.1%	2,202 0.7%	338 0.1%	245 0.1%	209 0.2%	3 0.02%	57,735 5.6%	22 0.01%
Agriculture									249,777 79.0%
Other	7,556 0.9%	184 0.02%		560 0.1%	356 0.2%	320 0.3%	19 0.1%	737 0.1%	12,957 4.1%
Fugitive dust				427,916 69.3%	112,472 48.3%	18,025 18.3%	121 0.8%		
Biomass burning	214,632 26.5%	8,413 0.7%	76 0.03%	27,703 4.5%	13,514 5.8%	11,217 11.4%	2,125 13.7%	83,592 8.1%	14 0.00%

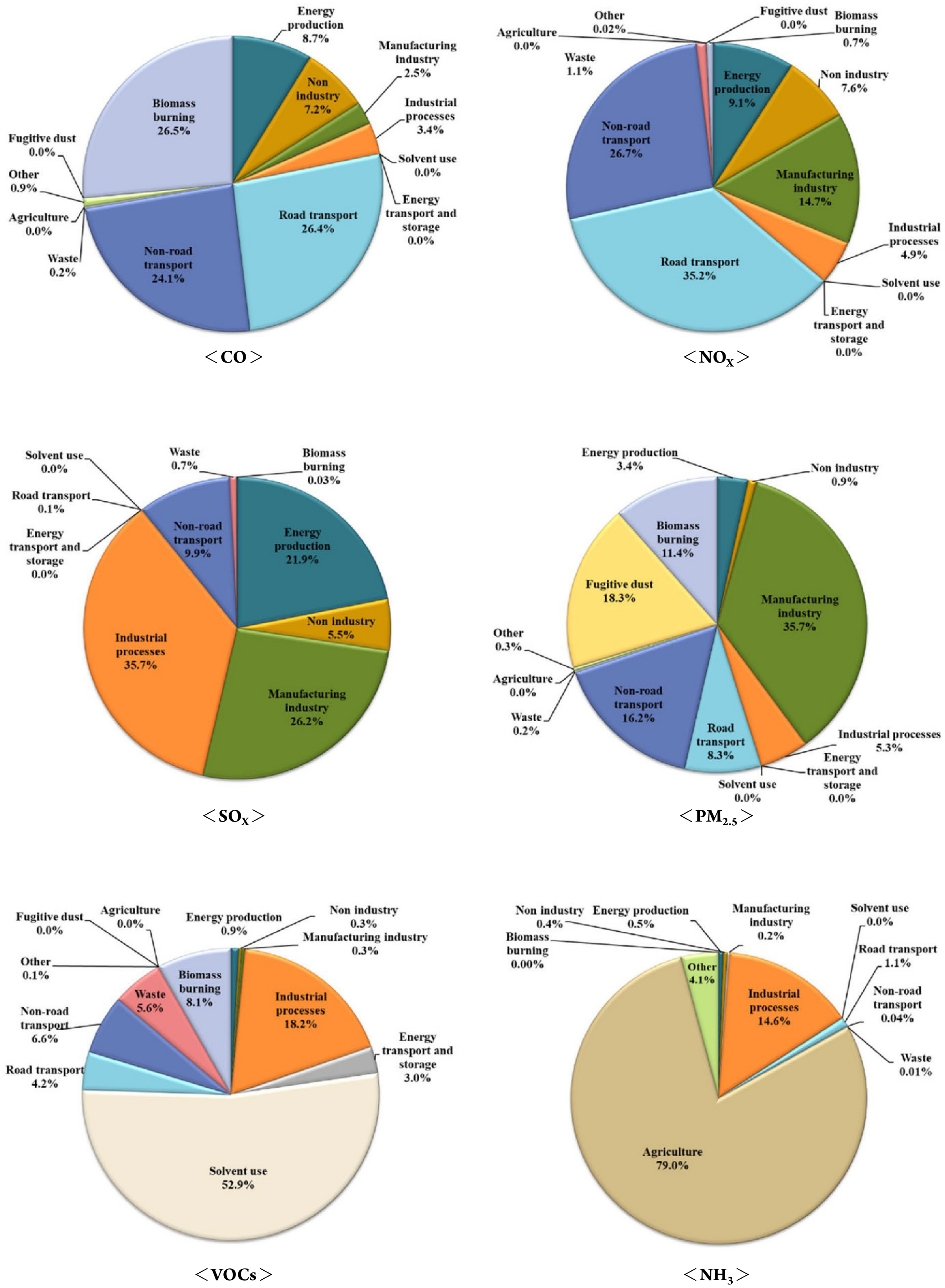


Fig. 1. 2018 emission contributions of different emission source categories by pollutant.

Table 3. Emission source classification.

Classification	Source category
Energy (oil refinery not included)	Energy production (public power generation, private power generation, and district heating)
Industry (oil refinery included)	Manufacturing industry Industrial processes Waste Oil refinery
Road	Road transport (passenger cars, vans, buses, freight cars, special cars, RVs, and two-wheeled vehicles)
Non-road	Non-road transport (railroads, ships, agricultural machinery, and construction machinery)
Everyday activities and others	Non-industry Energy transport and storage Solvent use Agriculture Others Fugitive dust Biomass burning

source categories by pollutant were as follows: biomass burning (26.5%), road transport (26.4%), and non-road transport (24.1%) for CO; road transport (35.2%), non-road transport (26.7%), manufacturing industry (14.7%) for NO_X; industrial process (35.7%), manufacturing industry (26.2%), energy production (21.9%) for SO_X; manufacturing industry (35.7%), fugitive dust (18.3%), non-road transport (16.2%) for $PM_{2.5}$; solvent use (52.9%), industrial process (18.2%) for VOCs; agriculture (79.0%), industrial process (14.6%) for NH_3 (Fig. 1).

For primary $PM_{2.5}$ and PM precursors (NO_X, SO_X, VOCs, and NH_3), the 13 first-level source categories were classified into five sectors (energy, industry, road, non-road, and everyday activities and others), as presented in Table 3. The national air pollutant emissions in 2018 were compared with those in 2017, and major causes of changes in emissions were analyzed.

NO_X emissions decreased by 3.1% compared to the previous year due to the replacement old cars with new cars in the road sector and the reinforcement of the emission control for power plants in the energy sector. SO_X emissions decreased by 4.6% compared to the previous year due to the reduction in fuel consumption (including B-C oil) of power plants and strengthened emission control. $PM_{2.5}$ emissions increased by 7.3% due to the increase in the number of ships and construction machinery registrations in the non-road sector. VOCs emissions decreased by 1.1% compared to the previous year due to the decline in paint supply in the everyday activities and others sector. NH_3 emissions increased due to the increase in fertilizer consumption and the number of livestock population in the everyday activities and others sector (Fig. 2).

3.2 Comparison of Air Pollutant Emissions by Region

To examine air pollutant emission characteristics and changes in emissions by region in Korea, the 17 dos (provinces) and metropolitan cities were grouped into the following five regions: SMA (Seoul, Incheon, and Gyeonggi-do), Gangwon region (Gangwon-do), Chungcheong region (Daejeon, Sejong, Chungcheongbuk-do, and Chungcheongnam-do), Honam region (Gwangju, Jeollabuk-do, and Jeollanam-do), and Yeongnam region (Busan, Daegu, Ulsan, Gyeongsangbuk-do, and Gyeongsangnam-do) (Table 4).

To examine pollutant emission characteristics by region, the current status of factors related to major pollutant emission, such as population, economy, large-scale places of business, cars, and construction machinery, was analyzed. For the analysis of the current status of the economy by region, Gross Regional Domestic Product (GRDP) data published by the Korean Statistical Information Service (KOSIS) were utilized. GRDP is the sum

(Unit: metric tons/year)

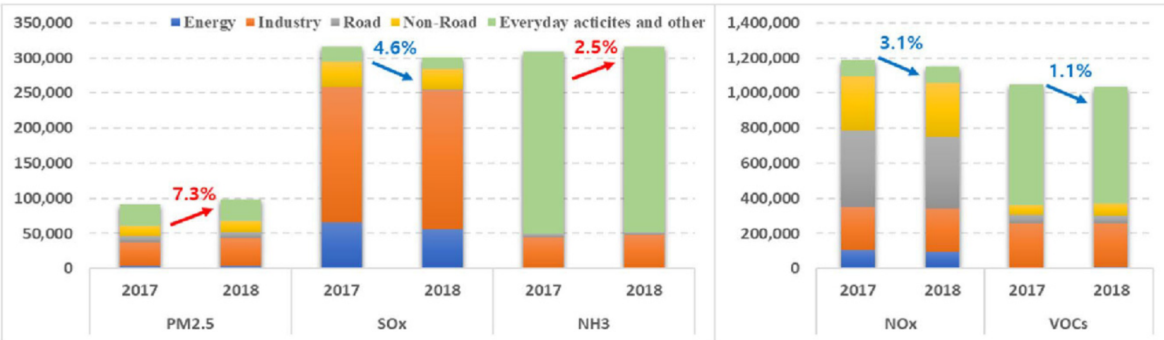
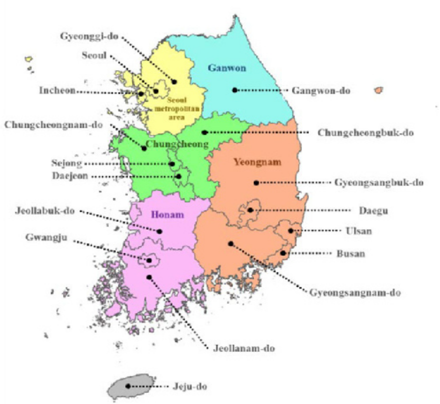
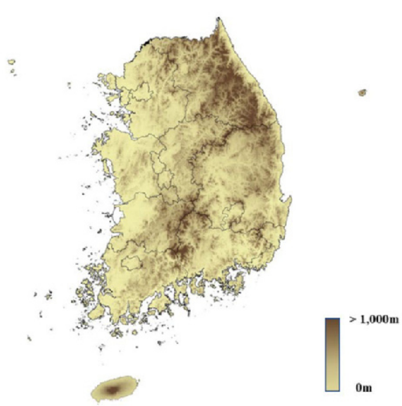


Fig. 2. 2018 Air pollutant emissions by sector.

Table 4. Classification of administrative districts by region.

Region	Administrative divisions	Region	Administrative divisions
SMA	Seoul	Honam region	Gwangju
	Incheon		Jeollabuk- do Jeollanam-do
	Gyeonggi-do	Yeongnam region	Busan
Gangwon region	Gangwon-do		Daegu
Chungcheong region	Daejeon		Ulsan
	Sejong		Gyeongsanbuk-do Gyeongsangnam-do
	Chungcheongbuk-do Chungcheongnam-do	Others	Jeju Island





of the market prices of all final goods and services produced in a fixed economic zone for a certain period of time. It is used to establish local financial and economic policies because it comprehensively represents the status of the local economy. As of 2018, SMA showed the highest values and proportions for both population (49.8%) and GRDP (52.2%), followed by the Yeongnam, Chungcheong, Honam, and Gangwon regions, and Jeju Island (Table 5).

According to the analysis results, the Yeongnam region had the largest number of large-scale places of business (annual pollutant emissions > 20 tons; 37.5%), followed by SMA (24.4%) and the Chungcheong region (19.1%). For the analysis of construction machinery, SMA had the largest number of registered vehicles (44.5%) and excavators (33.5%), followed by the Yeongnam region (26.8 and 27.3%, respectively) (Table 6).

Table 7 and Fig. 3 show the emissions by administrative division and region in 2018. SMA exhibited the largest emissions of CO (238,525 tons; 29.5%), NO_X (322,296 tons; 27.9%), and BC (5,215 tons; 33.5%). The Yeongnam region recorded the largest emissions of SO_X (113,601 tons; 37.7%), TSP (189.829 tons; 30.7%), PM_{10} (72,160 tons; 31.0%), $PM_{2.5}$ (32,945 tons; 33.5%), VOCs (344,649 tons; 33.3%), and NH_3 (81,881 tons; 25.9%).

The analysis on the major cause of changes in emissions and the comparison of regional and sectoral emissions based on emissions by region and pollutant is presented in the next section.

Table 5. GRDP by region in 2018.

	Population (thousands)		GRDP (trillion)	
Nationwide	51,826	100.0%	1,903	100.0%
SMA	25,797	49.8%	992	52.2%
Gangwon region	1,543	3.0%	47	2.5%
Chungcheong region	5,530	10.7%	238	12.5%
Honam region	5,179	10.0%	166	8.7%
Yeongnam region	13,110	25.3%	440	23.1%
Jeju Island	667	1.3%	20	1.1%

Source: KOSIS (Korean Statistical Information Service)

3.2.1 Analysis of Changes in Emissions for SMA

Almost half of the national population of Korea is concentrated in SMA, which consists of Seoul Metropolitan City (the capital), Incheon, and Gyeonggi-do, as it is the center of politics, economy, society, and culture. To improve the air pollution of SMA caused by high population density, traffic congestion, and industrialization, a separate law (Special Act On The Improvement Of Air Quality In Seoul Metropolitan Area, 2003) was enacted. Based on this, the Air Quality Management Plan in Seoul Metropolitan Area (2005) has been established and implemented. The plan includes strengthening of vehicle emission standards, supply of eco-friendly vehicles and expansion of infrastructure, details regarding total air pollutant emissions limitations for places of business, mandatory installation of VRU at gas stations, reinforcing the management of fugitive dust from vacant lands and places of business.

The population and economy indicators showed that SMA had the largest population (approximately 49.8%) and recorded the highest GRDP (approximately 52.2%) in 2018. The electric, electronic, and precision instrument manufacturing sector constituted the highest proportion of GRDP.

Air pollutant emissions from SMA in 2018 were estimated to be 17,162 tons of $PM_{2.5}$, 22,120 tons of SO_X, 322,296 tons of NO_X, 318,393 tons of VOCs, and 58,023 tons of NH_3. In addition, the contributions of each pollutant to the national emissions were as follows: $PM_{2.5}$ (17.4%), SO_X (7.3%), NO_X (27.9%), VOCs (30.7%), NH_3 (18.4%). $PM_{2.5}$ and VOCs emissions increased by 4.4% and 1.1% compared to the previous year, while SO_X,

Table 6. Current Status of places of business and the number of registered cars and construction machinery by region in 2018

Region	Places of business[1)]		Cars[2)]		Construction machinery[3)]	
	Number of registrations	Proportion (%)	Number of registrations	Proportion (%)	Number of registrations	Proportion (%)
SMA	1,000	24.4	10,319,869	44.5	168,093	33.5
Gangwon region	123	3.0	766,374	3.3	26,442	5.3
Chungcheong region	783	19.1	2,726,164	11.7	79,053	15.8
Honam region	644	15.7%	2,612,334	11.3%	82,348	16.4%
Yeongnam region	1,539	37.5%	6,224,236	26.8%	136,966	27.3%
Jeju Island	15	0.4%	553,578	2.4%	8,744	1.7%
Total	4,104	100.0%	23,202,555	100.0%	501,646	100.0%

*Sources: 1) Stack Emission Management System (SEMS), National Air Emission Inventory and Research Center, Ministry of Environment (Places of business represent large-scale places of business with annual NO_X, SO_X, and TSP emissions > 20 tons)
2) Number of registered cars: KOSIS (Korean Statistical Information Service)
3) Number of registered construction machinery: Ministry of Land, Infrastructure and Transport

Table 7. Air pollutant emissions by administrative divisions in 2018. (Unit: metric tons/year)

Dos (provinces) and metropolitan cities		CO	NO_X	SO_X	TSP	PM_{10}	$PM_{2.5}$	BC	VOCs	NH_3
	Total	808,801 100.0%	1,153,265 100.0%	300,979 100.0%	617,481 100.0%	232,993 100.0%	98,388 100.0%	15,562 100.0%	1,035,636 100.0%	315,975 100.0%
SMA	Seoul	59,091 7.3%	88,319 7.7%	1,095 0.4%	31,069 5.0%	15,130 6.5%	3,973 4.0%	1,498 9.6%	72,393 7.0%	3,469 1.1%
	Incheon	42,473 5.3%	54,996 4.8%	12,165 4.0%	22,496 3.6%	7,601 3.3%	2,701 2.7%	607 3.9%	55,061 5.3%	7,166 2.3%
	Gyeonggi-do	136,960 16.9%	178,981 15.5%	8,859 2.9%	84,050 13.6%	31,342 13.5%	10,488 10.7%	3,110 20.0%	190,940 18.4%	47,387 15.0%
	Sub total	238,525 29.5%	322,296 27.9%	22,120 7.3%	137,615 22.3%	54,074 23.2%	17,162 17.4%	5,215 33.5%	318,393 30.7%	58,023 18.4%
Gangwon region	Gangwon-do	50,996 6.3%	79,834 6.9%	13,802 4.6%	36,165 5.9%	9,772 4.2%	4,109 4.2%	749 4.8%	30,263 2.9%	14,848 4.7%
Chungcheong region	Daejeon	10,660 1.3%	16,051 1.4%	492 0.2%	5,271 0.9%	1,908 0.8%	653 0.7%	219 1.4%	16,758 1.6%	764 0.2%
	Sejong	4,956 0.6%	5,260 0.5%	80 0.0%	2,500 0.4%	1,024 0.4%	345 0.4%	123 0.8%	5,962 0.6%	2,760 0.9%
	Chungcheongbuk-do	42,067 5.2%	60,899 5.3%	7,223 2.4%	32,096 5.2%	9,462 4.1%	3,591 3.7%	853 5.5%	43,144 4.2%	16,981 5.4%
	Chungcheongnam-do	65,226 8.1%	107,613 9.3%	69,989 23.3%	81,841 13.3%	37,203 16.0%	18,129 18.4%	1,318 8.5%	78,132 7.5%	53,163 16.8%
	Sub total	122,909 15.2%	189,823 16.5%	77,784 25.8%	121,708 19.7%	49,598 21.3%	22,719 23.1%	2,513 16.1%	143,997 13.9%	73,667 23.3%
Honam region	Gwangju	7,956 1.0%	12,270 1.1%	173 0.1%	5,225 0.8%	1,710 0.7%	546 0.6%	153 1.0%	15,722 1.5%	968 0.3%
	Jeollabuk-do	46,257 5.7%	38,562 3.3%	3,761 1.2%	42,097 6.8%	10,629 4.6%	3,563 3.6%	773 5.0%	69,846 6.7%	35,197 11.1%
	Jeollanam-do	64,643 8.0%	105,269 9.1%	58,621 19.5%	71,464 11.6%	28,206 12.1%	13,156 13.4%	1,130 7.3%	88,958 8.6%	43,727 13.8%
	Sub total	118,856 14.7%	156,101 13.5%	62,555 20.8%	118,787 19.2%	40,545 17.4%	17,265 17.5%	2,056 13.2%	174,525 16.9%	79,892 25.3%
Yeongnam region	Busan	26,662 3.3%	49,951 4.3%	7,897 2.6%	17,031 2.8%	6,886 3.0%	2,644 2.7%	525 3.4%	42,340 4.1%	1,620 0.5%
	Daegu	17,213 2.1%	26,370 2.3%	2,595 0.9%	10,708 1.7%	3,911 1.7%	1,294 1.3%	338 2.2%	31,875 3.1%	1,668 0.5%
	Ulsan	31,400 3.9%	48,719 4.2%	42,794 14.2%	8,932 1.4%	4,080 1.8%	2,274 2.3%	298 1.9%	91,961 8.9%	15,129 4.8%
	Gyeongsangbuk-do	96,585 11.9%	104,098 9.0%	37,718 12.5%	107,358 17.4%	45,300 19.4%	22,007 22.4%	2,055 13.2%	89,304 8.6%	36,544 11.6%
	Gyeongsangnam-do	49,199 6.1%	73,050 6.3%	22,596 7.5%	45,799 7.4%	11,984 5.1%	4,726 4.8%	1,031 6.6%	89,168 8.6%	26,920 8.5%
	Sub total	221,058 27.3%	302,187 26.2%	113,601 37.7%	189,829 30.7%	72,160 31.0%	32,945 33.5%	4,248 27.3%	344,649 33.3%	81,881 25.9%
Jeju-do		11,130 1.4%	17,285 1.5%	1,836 0.6%	10,028 1.6%	3,495 1.5%	1,065 1.1%	223 1.4%	9,000 0.9%	7,655 2.4%
Sea*		45,327 5.6%	85,739 7.4%	9,282 3.1%	3,349 0.5%	3,349 1.4%	3,123 3.2%	557 3.6%	14,809 1.4%	8 0.0%

*Sea: Air pollutant emissions from maritime transport such as ships and fishing boats

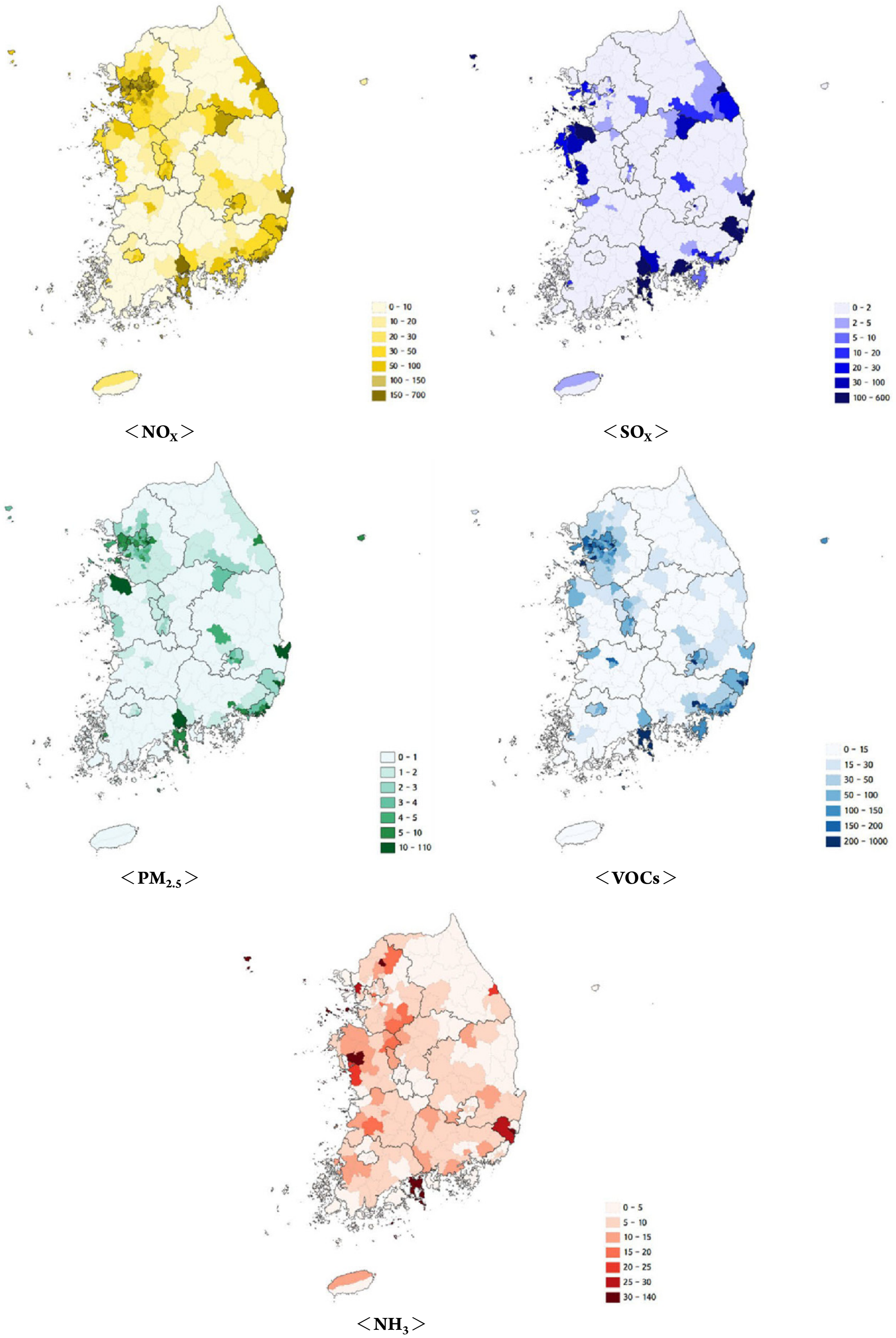


Fig. 3. 2018 Air pollutant emissions by administrative division (Unit: metric/km^2).

(Unit: metric tons/year)

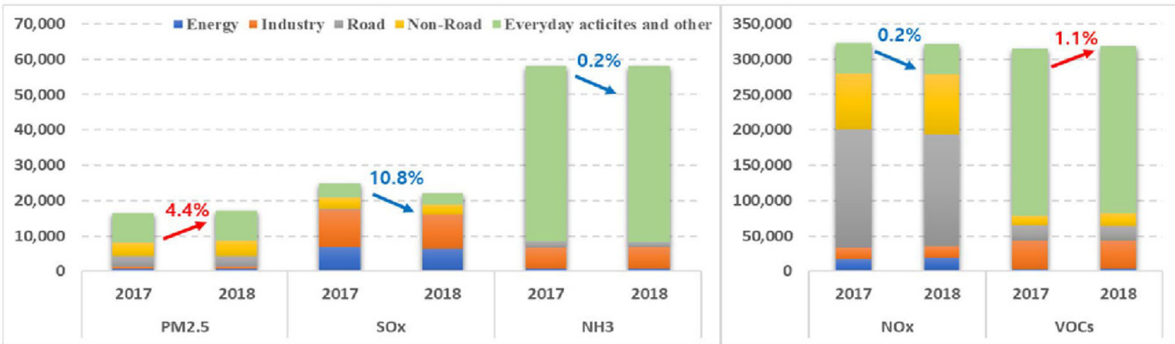


Fig. 4. Air pollutant emissions from SMA in 2018.

NO_X, and NH_3 emissions decreased by 10.8%, 0.2%, and 0.2%, respectively. Meanwhile, the contributions of NO_X and SO_X to the emissions from the road and industry sector respectively were the largest compared to other pollutants. In addition, $PM_{2.5}$, VOCs, and NH_3 contributed the largest to the emissions from the everyday activities and others sector (Fig. 4).

SMA's emissions from the road transport recorded the largest compared to other regions as it recorded the largest number of vehicles registered (44.5%), and VKT (40.5%) ($PM_{2.5}$ 36.1%, SO_X 40.2%, NO_X 38.9%, VOCs 46.9%, and NH_3 40.9%). SO_X, NO_X, VOCs, and NH_3 emissions decreased compared to those of the previous year. This is due to the replacement of old vehicles with new ones, which offset the effects of the increase in the number of vehicle registrations (3.1%, 307,000 units) and VKT (2.7%, 3,428 million km).

SMA's emissions from the non-transport sector were also the largest compared to those of other regions. (NO_X: 27.5%, $PM_{2.5}$: 28.0%, VOCs: 27.5%, and NH_3: 27.6%). The region's $PM_{2.5}$ and NO_X emissions from the construction machinery increased by 12.3% (366 tons) and 11.3% (6,867 tons) compared to those in the previous year. This was because construction machinery registrations (including excavators) increased by 9.9% (20,671 units) and the swaths of construction sites increased by 7.0% (5,001 m^2). VOCs emissions increased by 23.7% (3,568 tons) compared to that of the previous year. This was mainly because of the decrease in the number of registered leisure boat using gasoline in Incheon (by 26.4%, 880 units).

SO_X emissions decreased by 10.8% (2,686 tons) compared to those of the previous year. In particular, SO_X emissions from the industry sector decreased by a large margin (10.3%, 1,077 tons). This was due to the decreased consumption of industrial bituminous coal (66,000 tons, 19.5%) in Gyeonggi-do.

3.2.2 Analysis of Changes in Emissions for the Gangwon Region

Most of the Gangwon region, which is located in the northeastern part of Korea, is mountainous. Under the influence of such geographical conditions, industrial complexes are underdeveloped, which led to relatively low proportion of manufacturing-based industries.

The population and economy indicators showed that this region accounted for approximately 3.0% of the national population as of 2018. The GRDP of the region was approximately 2.5% of the national GRDP. More specifically, the public administration, defense and social security-related administration sector showed the highest proportion in GRDP. The manufacturing sector represented approximately 0.9% of the national GRDP.

Air pollutant emissions from the Gangwon region in 2018 were estimated to be 4,109 tons of $PM_{2.5}$, 13,802 tons of SO_X, 79,834 tons of NO_X, 30,263 tons of VOCs, and 14,848 tons of NH_3. In addition, the contributions of each pollutant to the national emissions were as follows: $PM_{2.5}$ (4.2%), SO_X (4.6%), NO_X (6.9%), VOCs (2.9%), NH_3 (4.7%). $PM_{2.5}$, SO_X, and NO_X emissions decreased by 0.1%, 2.3%, and 7.2%, respectively, compared to those of the previous year, whereas VOCs and NH_3 increased by 6.7% and 7.6%, respectively. Meanwhile, in the Gangwon region, the contributions of $PM_{2.5}$, VOCs, and NH_3 to the emissions from everyday activities and others sector respectively were the largest compared to other pollutants. In addition, NO_X and SO_X contributed the largest to the emissions from the indus-

(Unit: metric tons/year)

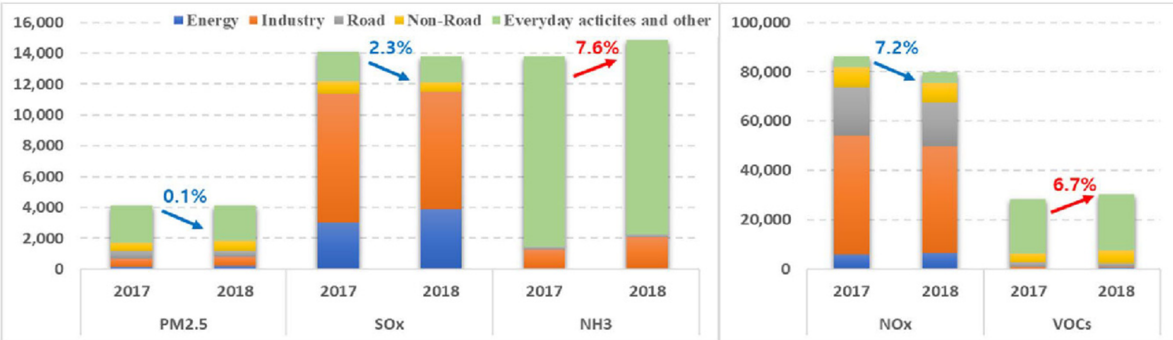


Fig. 5. Air pollutant emissions from the Gangwon region in 2018.

try sector (Fig. 5).

Emissions from the energy sector increased compared to those of the previous year (NO_X: 10.9%, SO_X: 29.6%, $PM_{2.5}$: 66.8%, VOCs: 50.2%, and NH_3: 58.0%). This was because the consumption of coal (including bituminous coal) and LNG increased by 43.7% (3,228,000 tons) and 62.2% (269 million m^3), respectively, due to the operation of new thermal power plants (coal and LNG).

VOCs emissions from the non-road transport sector increased by 38.0% (1,449 tons) compared to those of the previous year. This was mainly because of the increase in the number of registered leisure boat (by 47.2%, 1,570 units).

On the other hand, NO_X and SO_X emissions from the industry sector decreased by 10.4% (4,992 tons) and 9.1% (764 tons), respectively, compared to those of the previous year. This was due to the reduction in the fuel (bituminous coal) consumption of cement production facilities. NH_3 emissions increased by 67.1% compared to those in the previous year. This was mainly because emissions from $DeNO_X$ facilities in the industry sector increased by 67.7% (826 tons).

3.2.3 Analysis of Changes in Emissions for the Chungcheong region

The Chungcheong region, located in the center of Korea, consists of Daejeon Metropolitan City, Sejong Special Self-governing City, Chungcheongnam-do, and Chungcheongbuk-do. In the western part of the region, thermal power plants (coal and LNG), petrochemical complexes, iron and steel mills, and large manufacturing industries are located near trading ports. Meanwhile, in the eastern part of the region, high-value-added manufacturing industries (e.g., medicine and electronics) and food manufacturing industries are located.

The population and economy indicators showed that the region represented approximately 10.7% of the national population as of 2018. The GRDP of the region was approximately 12.5% of the national GRDP. The electric, electronic, and precision instrument manufacturing sector showed the highest proportion in GRDP, followed by the coal and petrochemical product manufacturing sector.

Air pollutant emissions from the Chungcheong region in 2018 were estimated to be 22,719 tons of $PM_{2.5}$, 77,784 tons of SO_X, 189,823 tons of NO_X, 143,997 tons of VOCs, and 73,667 tons of NH_3. In addition, the contributions of each pollutant to the national emissions were as follows: $PM_{2.5}$ (23.1%), SO_X (25.8%), NO_X (16.5%), VOCs (13.9%), NH_3 (23.3%). $PM_{2.5}$ and NH_3 emissions increased by 9.5% and 1.5%, respectively, compared to the previous year, whereas SO_X, NO_X, and VOCs emissions decreased by 1.8%, 5.9%, and 3.8%, respectively. Meanwhile, in the Chungcheong region, the contributions of $PM_{2.5}$, NO_X and SO_X to the emissions from the industry sector respectively were the largest compared to other pollutants. In addition, VOCs and NH_3 contributed the largest to the emissions from the everyday activities and others sector (Fig. 6).

In the case of the Chungcheong region, pollutant emissions from the energy sector were large compared to other regions ($PM_{2.5}$: 37.4%, SO_X: 36.9%, NO_X: 25.6%). NO_X and SO_X emissions from the energy sector decreased by 24.1% (7,838 tons) and 16.3% (4,081 tons), respectively, compared to those of the previous year. This was because of the reinforcement of the power plant emission management, which offset the effects of increased consumption

(Unit: metric tons/year)

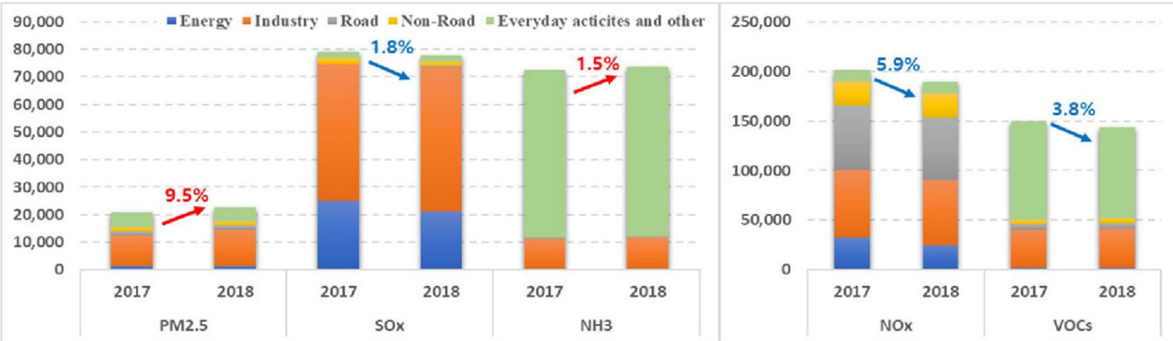


Fig. 6. Air pollutant emissions from the Chungcheong region in 2018.

of coal (including bituminous coal) in the coal-fired power plants of the region (1.0%, 444,000 tons) compared to the previous year.

$PM_{2.5}$ and SO_X emissions from the industry sector increased by 20.1% (2,242 tons) and 6.6% (3,295 tons) compared to those of the previous year. This was because of the increase in anthracite consumption in the primary metal industry (23.3%).

VOCs emissions decreased by 3.8% (5,737 tons) compared to those of the previous year. More specifically, VOCs emissions from the everyday activities and others sector decreased by 7.3% (7,246 tons) compared to those of the previous year. This was due to the emissions reductions (6,501 tons, 22.5%) caused by the decrease (21.8%) in the consumption of paint used for architecture and buildings in the region.

Meanwhile, NH_3 emissions represented 23.3% of the national emissions, and increased by 1.5% (1,084 tons) compared to those of the previous year. This was mainly because the emissions from the agriculture-manure management sector increased by 1.3% (720 tons), which was caused by a 3.3% (1,606,000 units) increase in the number of livestock population, including cows, pigs, and chickens.

3.2.4 Analysis of Changes in Emissions for the Honam Region

The Honam region, which consists of Gwangju Metropolitan City, Jeollabuk-do, and Jeollanam-do, is located in the southwestern part of Korea. It is Korea's representative breadbasket with wide plains, such as Honam and Naju plains. Thermal power plants (coal and LNG) and the nation's largest petrochemical complex are located in Yeosu, a southern part of the region, in addition to nearby iron and steel mills in Gwangyang.

The population and economy indicators showed that the region accounted for approximately 10.0% of the national population as of 2018. The GRDP of the region is approximately 8.7% of the national GRDP. More specifically, the coal and petrochemical product manufacturing sector showed the highest proportion of GRDP.

Air pollutant emissions from the Honam region in 2018 were estimated to be 17,265 tons of $PM_{2.5}$, 62,5554 tons of SO_X, 156,101 tons of NO_X, 174,525 tons of VOCs, and 79,892 tons of NH_3. In addition, the contributions of each pollutant to the national emissions were as follows: $PM_{2.5}$ (17.5%), SO_X (20.8%), NO_X (13.5%), VOCs (16.9%), NH_3 (25.3%). $PM_{2.5}$ and NH_3 emissions increased by 11.7% and 6.3%, respectively, compared to those of the previous year, whereas SO_X, NO_X, and VOCs decreased by 0.1%, 0.8%, and 2.5%, respectively. Meanwhile, the contributions of $PM_{2.5}$, and SO_X to the emissions from the industry sector, the contributions of NO_X to the emissions from the road sector, the contributions of VOCs and NH_3 emissions from the everyday activities and others sector were the largest in the region (Fig. 7).

In the case of the industry sector, $PM_{2.5}$, SO_X, and NO_X emissions increased by 30.9% (2,010 tons), 5.8% (2,671 tons), and 6.4% (2,666 tons), respectively, compared to those of the previous year. This was mainly because the increased consumption of coal, including anthracite, in the manufacturing sector (13.3%, 318,000 tons) in Jeollanam-do.

NO_X emissions from the road transport and non-road transport sectors decreased by 5.1% (2,611 tons) and 5.9% (2,078 tons), respectively, compared to those of the previous year. For road transport, emissions from the sector decreased because of the decrease in the number

(Unit: metric tons/year)

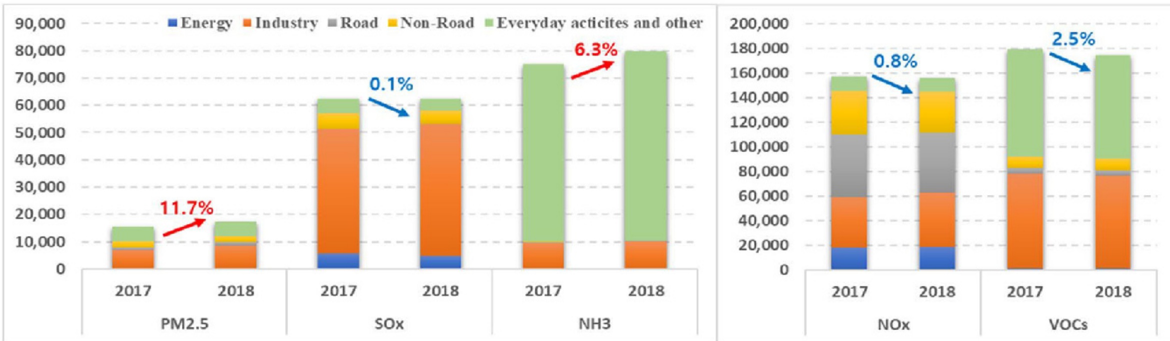


Fig. 7. Air pollutant emissions from the Honam region in 2018.

of old cars registrations and the replacement of old cars with new ones, which offset the impacts of the increase in the number of car registrations (3.0%, 77,000 units) in the region. In the case of the non-road transport, NO_X emissions decreased mainly because of the decrease in emissions from the non-road-construction machinery sector (13.6%, 1,674 tons) caused by the reduction in the number of registered construction machinery (14.6%, 6,162 units) in the region.

VOCs emissions decreased by 2.5% (4,561 tons) compared to those in the previous year. More specifically, this was because emissions by paint that is used for shipbuilding decreased by 16.9% (2,528 tons). For paint consumption, it decreased by 17.1% (4,606 kL) compared to those in the previous year.

NH_3 emissions increased by 6.3% (4,722 tons) compared to those in the previous year. The Honam region exhibited the largest NH_3 emissions in the country from the everyday activities and others sector. This was due to the 21.0% (1,335 tons) increase in emissions caused by a 20.8% (44,000 tons) increase in fertilizer consumption in farmlands, and the 5.0% (2,873 tons) increase in NH_3 emissions from the manure sector caused by a 10.2% (5,902,000 units) increase in the number of livestock population.

3.2.5 Analysis of Changes in Emissions for the Yeongnam Region

The Yeongnam region, which consists of Busan Metropolitan City, Daegu Metropolitan City, Ulsan Metropolitan City, Gyeongsangbuk-do, and Gyeongsangnam-do, is located in the southeastern part of Korea. Iron and steel manufacturing, shipbuilding, automobile manufacturing, and petrochemical industries as well as the nation's largest trading port (Busan Port) are located in the region.

The population and economy indicators showed that the region represented approximately 25.3% of the national population as of 2018. The GRDP of the region is approximately 23.1% of the national GRDP. More specifically, the machinery transport equipment, and other product manufacturing sector showed the highest proportion of GRDP, followed by electric, electronic, and precision instrument manufacturing and non-metallic mineral and metal product manufacturing sector.

Air pollutant emissions from the Yeongnam region in 2018 were estimated to be 32,945 tons of $PM_{2.5}$, 113,601 tons of SO_X, 302,187 tons of NO_X, 344,649 tons of VOCs, and 81,881 tons of NH_3. In addition, the contributions of each pollutant to the national emissions were as follows: $PM_{2.5}$ (33.5%), SO_X (37.7%), NO_X (26.2%), VOCs (33.3%), NH_3 (25.9%). $PM_{2.5}$ and NH_3 emissions increased by 7.0 and 1.2%, respectively, compared to those in the previous year, whereas SO_X, NO_X, and VOCs emissions decreased by 6.5%, 3.8%, and 1.8%, respectively. Meanwhile, the contributions of $PM_{2.5}$ and SO_X to the emissions from the industry sector were the largest in the region. In addition, VOCs and NH_3 contributed the largest to the emissions from the everyday activities and others sector (Fig. 8).

In the case of the Yeongnam region, air pollutant emissions from the industry sector were found to be the largest in Korea. In this region, emissions from the industry sector were 17,459 tons of $PM_{2.5}$, 79,097 tons of SO_X, 76,780 tons of NO_X, 95,344 tons of VOCs, and 17,153 tons of NH_3. Each pollutant represented 43.1% ($PM_{2.5}$), 40.1% (SO_X), 31.2% (NO_X), 38.2% (VOCs), and 36.6% (NH_3) of national emissions from the industry sector,

(Unit: metric tons/year)

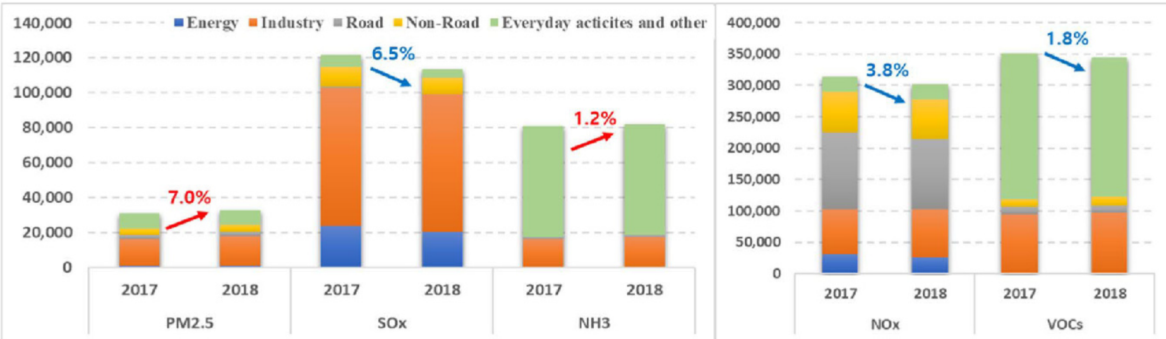


Fig. 8. Air pollutant emissions from the Yeongnam region in 2018.

respectively. $PM_{2.5}$ and NO_X emissions from the sector increased by 15.8% (2,380 tons) and 7.7% (5,459 tons), respectively, compared to those in the previous year. This was mainly because the consumption of coal, including anthracite, in the manufacturing sector increased by 15.1% (504 tons) compared to that in the previous year.

$PM_{2.5}$ emissions increased by 7.0% (2,156 tons) compared to those of the previous year. This was because of the increase in the consumption of anthracite in the industry sector. Meanwhile, NH_3 emissions also increased by 1.2% (982 tons) compared to the previous year. This was mainly because emissions from $DeNO_X$ facilities in the industry sector increased by 55.0% (1,282 tons).

NO_X emissions decreased by 3.8% (12,100 tons) compared to those in the previous year. These emissions decreased by 18.7% (5,854 tons) and 7.9% (9,581 tons) in the energy production and road transport sectors, respectively. For the energy production sector, this was mainly because of the reduction (4.1%) in the bituminous coal consumption by public power generation facilities and the reduction (19.6%, 5,292 tons) in emissions caused by the reinforcement of environmental facilities for power generation facilities. In the case of the road sector, the main cause of such reduction was the decrease in emissions caused by the reduction in the number of old vehicles, which offset the impacts of the increase in vehicles registrations and VKT increased by 2.0% (124,000 units) and 0.8% (750 million km), respectively, compared to those of the previous year.

SO_X emissions decreased by 6.5% (7,866 tons) compared to those of the previous year. This was mainly due to the emissions reductions in the energy sector (15.5%, 3,697 tons), the non-road sector (17.1%, 1,966 tons), and the everyday activities and others sector (29.7%, 2,000 tons). More specifically, for the energy sector, the main cause of such reduction was the decrease in emissions from public power generation facilities as it was for NO_X. In the case of the non-road sector, the emissions reductions caused by the decrease in the number of cargo ships entering the ports (6.9%, 6,640 units) and the decrease in the sulfur content in fuel (B-C oil). For the everyday activities and others sector, such reductions were due to the emissions reductions (49.3%, 1,424 tons) caused by the reduction in the consumption of fuel oil for cooling and heating (9.4%, 174,000 kL) in commercial and public facilities.

VOCs emissions from the everyday activities and others sector decreased by 1.8% (6,417 tons) compared to those of the previous year. This was mainly due to the decrease in emissions (6.7%, 8,609 tons) caused by the reduction in the consumption of paint at coating facilities (6.5%, 17,719 kL). VOCs emissions from the non-road sector, on the other hand, increased by 23.6% (2,662 tons). This was due to the emissions increase (47.0%, 2,844 tons) caused by an increase in the number of registered leisure boat (47.2%, 785 units).

4. ASSESSMENT OF UNCERTAINTY IN EMISSIONS USING AIR QUALITY MODELING

4.1 Methodology

The latest activity data and the best available emission factors were applied to the emissions data estimated above. Nevertheless, there are uncertainties in some emission sources. Old emission factors, activity data with low reliability, and missing emission sources are mentioned as the causes of such uncertainties (Kim *et al.*, 2020a; Lee *et al.*, 2019; Kim and Jang, 2014). Therefore, it is necessary to examine the uncertainty of the estimated emission data. Since air pollutants have different emission characteristics depending on the emission sources, it is difficult to verify emissions in a consistent way and present the results in a quantitative manner. To overcome such difficulties, a method of indirectly examining the accuracy of emissions has been used. This methodology is about comparing the concentrations data from monitoring stations with the results of air quality modeling, a process of converting air pollutants emissions into atmospheric concentrations using 3D chemical transport model (Bae *et al.*, 2020a, b; Kim *et al.*, 2020a).

As such, this study uses a method of utilizing 3D chemical transport model to examine the uncertainty of national air pollutant emissions. This study was conducted based on the National Emission and Air quality assessment System (NEAS). NEAS consists of the Weather Research and Forecasting (WRF) model, the Sparse Matrix Operator Kernel Emissions (SMOKE) model, and the Community Multiscale Air Quality (CMAQ) model. The detailed physico-chemical options used in the WRF and CMAQ models are presented in Supplementary Materials (Table S1). CAPSS 2018 was used for domestic emissions and the KORUSv5 data was used for overseas emissions. The domains and horizontal resolutions were for the simulation were as follows: Northeast Asia (27 km), the Korean Peninsula (9 km), and South Korea (3 km). And 2018 was selected as the target year for simulation (Supplementary Materials Fig. S1). NO_2 and SO_2 were selected as the target pollutants for which the uncertainty of emissions was to be examined by considering the following three aspects: 1) The two pollutants themselves are harmful to people's health. It is important to identify the emissions of their uncertainty as they are major precursors transformed into PM through secondary formation in the atmosphere; 2) It is easy to intuitively interpret the overestimation/underestimation as emissions and concentrations of NO_2 and SO_2 have a relatively linear relationship, nature of primary pollutants; 3) Since are NO_2 and SO_2 less affected by long-range transport, it is easy to assess the emissions of each region. However, this study suggests the results of comparison between simulated and observed concentrations of $PM_{2.5}$ as well because of the importance $PM_{2.5}$ has.

An error between the simulated and observed concentrations may occur due to various factors. Representative factors are the uncertainties of meteorological input data, emissions input data, and various physical and chemical equations included in atmospheric chemical transport models. This study assumes that the systematic bias found at a similar level in most regions drives from the errors between meteorological input data and atmospheric chemical transport models. This is to examine the model's errors occurring from the perspectives of emissions. And the study presents and analyzes the regions with large errors between observed and simulated concentrations while comparing their annual mean concentrations to examine the uncertainty of emissions from the perspectives of total emissions.

4.2 Comparison between Simulated and Observed Concentrations

Based on the locations of the urban air pollution monitoring network, errors between the simulated and observed annual mean concentrations across Korea were found to be 0.6 ppb (3%) for NO_2, 0.1 ppb (4%) for SO_2, and 5.6 μg/m^3 (24%) for $PM_{2.5}$. The simulated concentrations of gaseous pollutants were similar to the observed concentrations relative to $PM_{2.5}$. And $PM_{2.5}$ concentrations were underestimated compared to the observed concentrations. In addition, this study compared the simulated and observed concentrations at a provincial and metropolitan city level. The bias of the simulated NO_2 concentrations was found to range from −7.5 ppb (−35%, Chungcheongbuk-do) to 5.6 ppb (41%, Jeollanam-do). And high reproducibility was observed in Daejeon, Daegu, and Jeju with the error < 1 ppb (< 5%) (Fig. 9[a]). The bias of the simulated SO_2 concentrations ranged from −2.9 ppb (−66%, Seoul) to 7.4 ppb (144%, Jeollanam-do). Overestimation occurred in 5 out of 17 dos (provinces) and metropolitan cities. And it was particularly notable in Jeollanam-do, Gyeongsangbuk-do, and Ulsan (Fig. 9[b]). The bias of the simulated $PM_{2.5}$ concentrations ranged from −9.5 μg/m^3 (−35%, Jeollabuk-do) to 6.4 μg/m^3 (26%, Gyeongsangbuk-do), and underestimation occurred in 15 out of the 17 dos (provinces) and metropolitan cities (Fig. 9[c]).

For dos (provinces) and metropolitan cities that exhibited an underestimation, a tendency towards underestimation was generally observed in most of the municipalities as well. On the other hand, for dos (provinces) and metropolitan cities that showed an overestimation, high

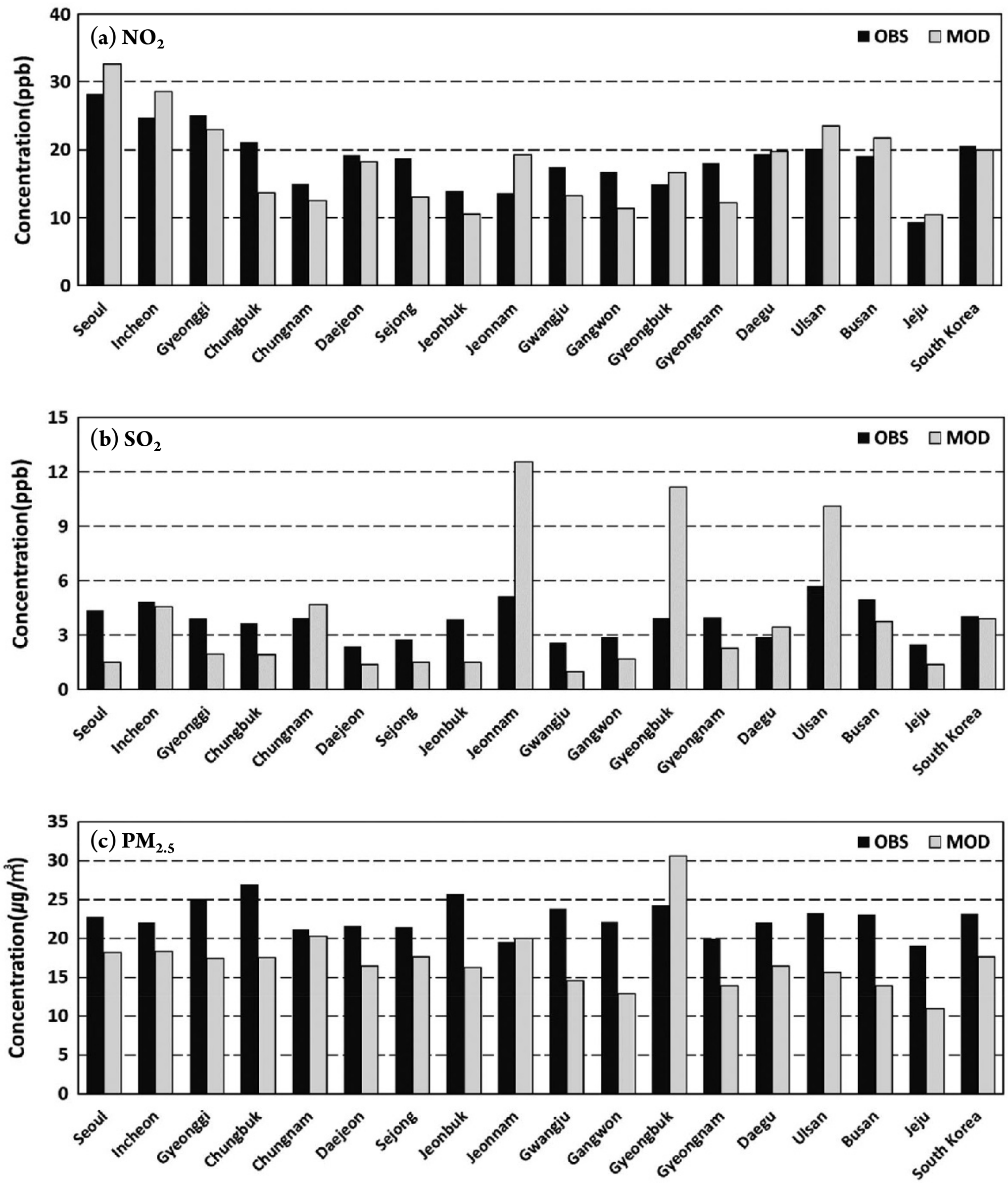


Fig. 9. Observed and simulated annual mean air pollutant concentrations. (a) NO_2, (b) SO_2, and (c) particulate matter with an aerodynamic diameter ≤ 2.5 μm ($PM_{2.5}$) concentrations by region.

simulated concentrations intensively occurred in some of the municipalities. Such municipalities include Gyeongsangbuk-do (Pohang), Jeollanam-do (Yeosu), Jeollanam-do (Gwangyang), and Chungcheongnam-do (Dangjin), and the simulated NO_2, SO_2, and $PM_{2.5}$ concentrations in those municipalities were 2–3 times higher than the observed concentrations of the same pollutants. However, for Ulsan, overestimation of SO_2 concentrations occurred at most of the air quality monitoring stations (11 stations, 73%), which was an exceptional case (Fig. 10).

This study assumes that the uncertainty of emissions would to be high for regions where the errors between the simulated and observed concentrations were large. The $PM_{2.5}$ concentrations in the atmosphere, however, are known to be affected in a complex manner by direct emissions from emission sources, secondary formation in the atmosphere by the chemical reactions of precursors, and long-range transport (Kim *et al.*, 2021d; Kim *et al.*, 2017a; Kim *et al.*, 2017b). The possibility that

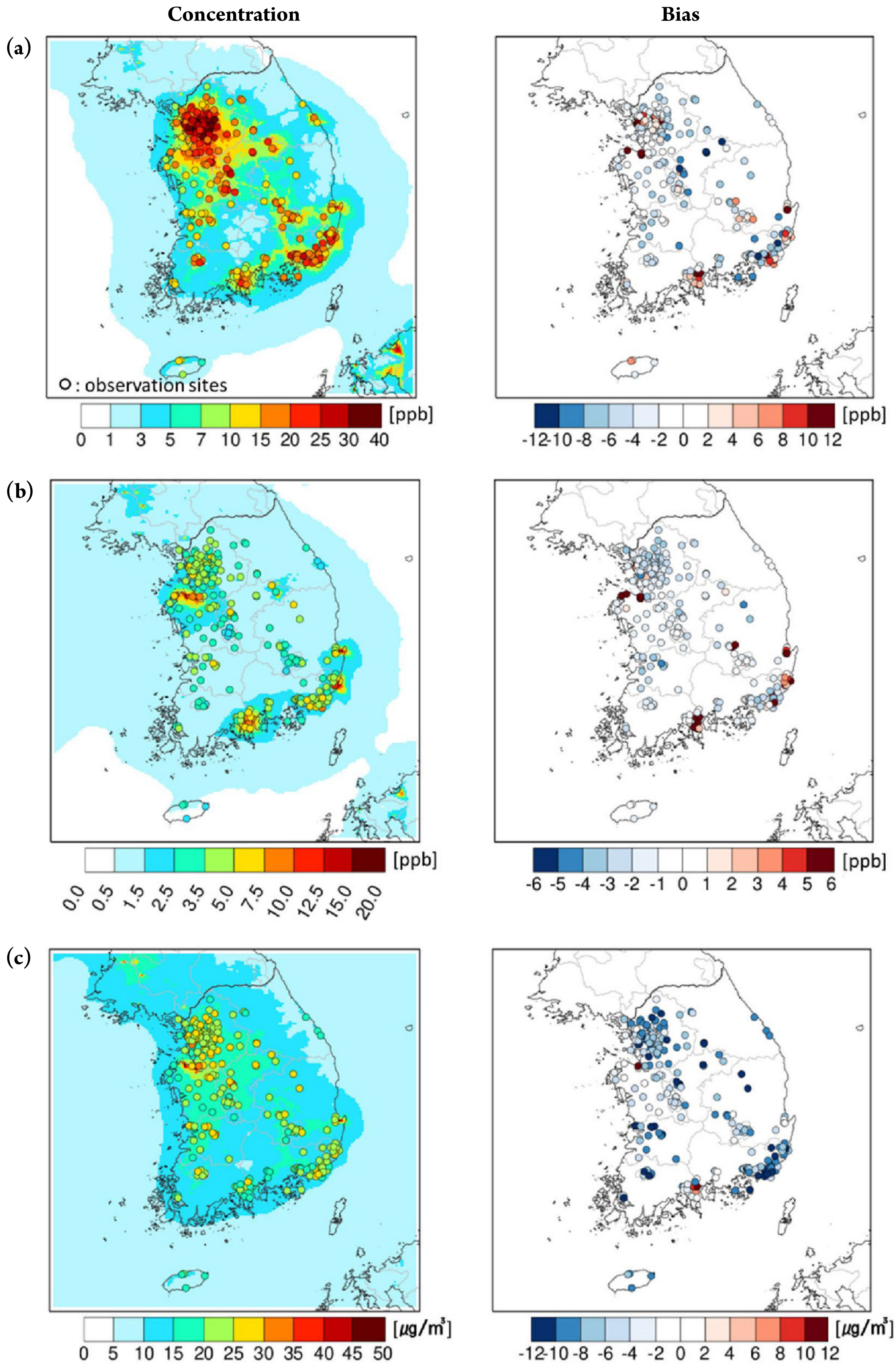


Fig. 10. Spatial distributions of observed (circle) and simulated (tile) annual mean air pollutant concentrations. (a) NO_2, (b) SO_2, and (c) $PM_{2.5}$ concentrations in 2018 and the bias between them by measurement point.

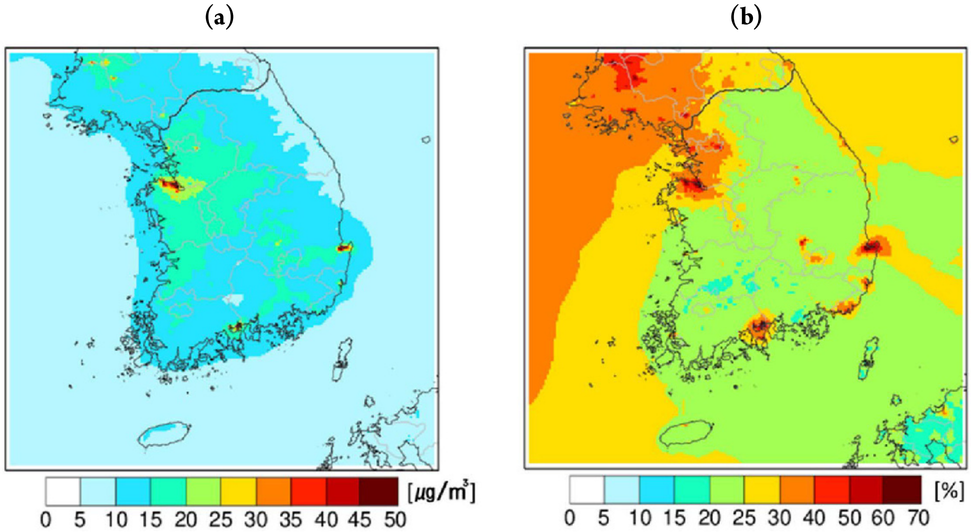


Fig. 11. (a) Simulated annual mean concentrations of $PM_{2.5}$ in 2018 and (b) the spatial distribution of primary $PM_{2.5}$ components' relative proportion.

long-range transport affected the errors between simulated and observed concentrations was determined to be low because it affects the entire country rather than specific regions (Bae *et al.*, 2021). The components generated by the secondary formation caused by precursors accounted for 50–60% of the domestic $PM_{2.5}$ concentrations (Kim *et al.*, 2020b). This explains why it is necessary to analyze the uncertainty of precursor emissions in addition to the uncertainty of the air pollutants directly emitted as $PM_{2.5}$.

Comparing observed and simulated concentrations on the basis of the concentrations of PM's detailed components would be the most direct way to distinguish the impacts of secondary formation from those of direct emission in the process of analysis. However, since the number of monitoring stations measuring the concentrations of $PM_{2.5}$ components is extremely limited, simulated concentrations were analyzed based on the following two assumptions: 1) If the uncertainty of primary PM emissions is large, the error will be relatively large in regions adjacent to emission sources due to their direct impacts from emission sources, and the proportion of primary PM components will be relatively high in the simulated $PM_{2.5}$ concentrations; 2) If the uncertainty of precursor emissions is higher, on the other hand, it takes some time for $PM_{2.5}$ to be generated after precursors come from emission sources. Therefore, the errors are likely to be larger in the downwind region relatively far from emission sources, and the proportion of secondary components (such as NO_X and SO_X), will be high in the simulated concentrations.

Figure 11 shows the spatial distribution of the relative proportion of primary PM components in the simulated $PM_{2.5}$ concentrations. For the Pohang, Yeosu, Gwangyang, and Dangjin regions mentioned above, the proportion of primary PM was $>$70%, which was relatively high compared to that in other regions. Based on this, the main cause of the error in $PM_{2.5}$ simulation for Pohang, Yeosu, Gwangyang, and Dangjin was determined to be the uncertainty of emissions (primary PM). And the major emission sources for the areas were analyzed reflecting this conclusion.

For the five regions where the simulated concentration was distinctively higher than the observed concentrations (Pohang, Yeosu, Gwangyang, Dangjin, and Ulsan), the manufacturing (first-level category)-others (second-level category), industrial process (first-level category)-iron and steel making (second-level category), industrial process (first-level category)-petroleum industry (second-level category), and non-road transport (first-level category)-ships (second-level category) sectors were major air pollutant emission sources. Among them, four emission sources at the second-level category level accounted for 57% (NO_X), 78% (SO_2), and 88% ($PM_{2.5}$) of the total air pollutant emissions in the five regions. Major emission sources were slightly different by region. In Pohang, Gwangyang, and Dangjin, manufacturing (first-level category)-others (second-level category) and industrial process (first-level category)-iron and steel making (second-level category) were major emission sources. Meanwhile, in Yeosu and Ulsan, industrial process (first-level category)-petroleum industry (second-level category) and non-road transport (first-level category)-cars (second-level category) were major emission sources. In particular, manufacturing (first-level category)-others (second-level category) emission sources produces the large amounts of emissions of all the target air pollutants of this study (NO_X, SO_2, and $PM_{2.5}$). In detail, the emission source of manufacturing (first-level category)-others (second-level category)-primary metal industry (third-level category), in which non-public anthracite is used as fuel, represented >99% of the emissions from manufacturing (first-level category)-others (second-level category). Thus, to improve the accuracy of emissions, it is necessary to first examine the uncertainty that may occur in the process of estimating emissions from corresponding emission sources. The uncertainty ahead, however, does not mean the uncertainty of emissions from point sources. When it comes to point sources of large-scale places of business, errors in emissions are not likely to occur because their emissions are estimated on the basis of TMS data. NAIR estimates national air pollutant emissions and has identified the problems with the activity data and the process of estimating emissions from corresponding emission sources. Accordingly, NAIR is conducting research on the improvement of the emission estimation method and the results of improvement to address these problems. The details will be presented in a follow-up paper.

In summary, in this study, air quality modeling was conducted using CAPSS 2018 emissions, and the uncertainty of the current emissions was examined through comparison between observed and simulated concentrations. It was determined that emissions from five regions (Pohang, Yeosu, Gwangyang, Dangjin, and Ulsan) need to be improved. Most of all, it is necessary to examine the emissions form point sources using non-public anthracite as a fuel in manufacturing (first-level category)-others (second-level category)-primary metal industry (third-level category).

5. CONCLUSIONS

According to the 2018 NEI, air pollutant emissions in the Republic of Korea, estimated using CAPSS, comprised 808,801 tons of CO; 1,153,265 tons of NO_X; 300,979 tons of SO_X; 617,481 tons of TSP; 232,993 tons of PM_{10}; 98,388 tons of $PM_{2.5}$; 15,562 tons of BC; 1,035,636 tons of VOCs; and 315,975 tons of NH_3, and CO, NO_X, SO_X, VOCs emissions decreased by 1.1%, 3.1%, 4.6%, and 1.1% respectively, while TSP, PM_{10}, $PM_{2.5}$, BC, NH_3 emissions increased by 4.2%, 6.6%, 7.3%, 0.04% and 2.5% respectively.

Emissions of primary $PM_{2.5}$ as well as $PM_{2.5}$, SO_X, VOCs, and NH_3, which contribute to the formation of secondary $PM_{2.5}$ were assessed in this study. For $PM_{2.5}$, SO_X, VOCs, and NH_3, Yeongnam region (33.5, 37.7, 33.3, and 25.9%, respectively) produced the largest amounts of emissions compared to other regions. Meanwhile, for NO_X, the largest amounts of emissions occurred in SMA (27.9%). In SMA, the large amounts of $PM_{2.5}$, VOCs, and NH_3 emissions were observed in the everyday activities and others sector (49.1, 74.2, and 85.7%, respectively), and the large amounts of SO_X emissions were observed in the industry sector (42.6%), and the large amounts of NO_X emissions were observed in the road sector (49.1%). In the Gangwon region, the large amounts of $PM_{2.5}$, VOCs, and NH_3 emissions occurred in the everyday activities and others sector (55.2, 74.7, and 84.9%, respectively) and the large amounts of SO_X and NO_X emissions occurred in the industry sector (55.0 and 54.1%, respectively). In the Chungcheong region, the large amounts of $PM_{2.5}$, SO_X, and NO_X emissions occurred in the industry sector (59.0, 68.0, and 34.7%, respectively) and the large amounts of VOCs and NH_3 emissions occurred in the everyday activities and others sector (64.2 and 83.5%, respectively). In the Honam region, the large amounts of $PM_{2.5}$ and SO_X emissions occurred from the industry sector (49.3 and 77.3%, respectively), and the large amounts of NO_X emissions occurred from the road sector (31.0%), and the large amounts of VOCs and NH_3 emissions occurred from the everyday activities and others sector (48.3 and 86.8%, respectively). In the Yeongnam region, large amounts of $PM_{2.5}$ and SO_X emissions occurred from the industry sector (53.0 and 69.6%, respectively), and the large amounts of NO_X emissions occurred from the road sector (37.1%), and the large amounts of VOCs and NH_3 emissions occurred from the everyday activities and others sector (64.5 and 77.6%, respectively).

The method of utilizing 3D chemical transport modeling was used to examine the uncertainty of national air pollutant emissions based on the NEAS. In this study, air quality was simulated using CAPSS 2018, and the uncertainty of the current emissions was examined through comparison between the simulated and observed concentrations. The results indicate that the proportion of primary PM in the simulated $PM_{2.5}$ concentrations was >70% for Pohang, Yeosu, Gwangyang, and Dangjin, which was relatively high compared to that for other areas. Based on this, the main cause of the errors in $PM_{2.5}$ simulation for Pohang, Yeosu, Gwangyang, and Dangjin was determined to be the uncertainty of emissions (primary PM) In addition, it is necessary to examine the emissions from places of business that use anthracite, a major emission source of $PM_{2.5}$, as fuel in these si (cities).

To improve the uncertainty of air pollutant emissions, NAIR of Republic of Korea has been conducting research as follows: development of emission factors for facility using SRF (Solid Refuse Fuel), asphalt concrete manufacturing facility, SRU (Sulfur Recovery Unit), latest car models; improvement of activity data on anthracite consumption, traffic volumes of cars, vacant land, and barbecue grilling; identification of missing emission sources such as CHE (Cargo Handling Equipment), military equipment, GSE (Ground Support Equipment). Based on these research efforts, NAIR aims to establish and implement air quality improvement policy, including highly effective PM reduction policies whose impacts can be felt by people, so that it can contribute to improve air quality and promote public health.

REFERENCES

Bae, C., Kim, H.C., Kim, B.U., Kim, Y., Woo, J.H., Kim, S. (2020a) Updating Chinese SO_2 emissions with surface observations for regional air-quality modeling over East Asia. Atmospheric Environment, 228, 117416. https://doi.org/10.1016/j.atmosenv.2020.117416

Bae, C., Kim, H.C., Kim, B.U., Kim, S. (2020b) Surface ozone response to satellite-constrained NO_X emission adjustments and its implications. Environmental Pollution, 258, 113469. https://doi.org/10.1016/j.envpol.2019.113469

Bae, C., Kim, E., Yoo, C., Kim, H.-C., Kim, Y.-M., Kim, S. (2021) Prioritizing Local Authorities Effective to Lower the Nationwide $PM_{2.5}$ Concentrations and the Personal Exposure Based on the Source Apportionment with the CAPSS 2016 Emissions Inventory. Journal of Korean Society for Atmospheric Environment, 37(4), 410–428. https://doi.org/10.5572/KOSAE.2021.37.3.410

Choi, S.-W., Kim, T., Lee, H.-K., Kim, H.-C., Han, J., Lee, K.-B., Lim, E.-H., Shin, S.-H., Jin, H.-A., Choi, E., Kim, Y.-M., Yoo, C. (2020) Analysis of the National Air Pollutant Emission Inventory (CAPSS 2016) and the Major Cause of Change in Republic of Korea. Asian Journal of Atmospheric Environment, 14(4), 422–445. https://doi.org/10.5572/ajae.2020.14.4.422

Choi, S.-W., Bae, C.-H., Kim, H.-C., Kim, T., Lee, H.-K., Song, S.-J., Jang, J.-P., Lee, K.-B., Choi, S.-A., Lee, H.-J., Park, Y., Park, S.-Y., Kim, Y.-M., Yoo, C. (2021) Analysis of the National Air Pollutant Emissions Inventory (CAPSS 2017) Data and Assessment of Emissions based on Air Quality Modeling in the Republic of Korea. Asian Journal of Atmospheric Environment, 15(4), 2021064. https://doi.org/10.5572/ajae.2021.064

Gong, J., Shim, C., Choi, K.-C., Gong, S. (2021) The Characteristics of $PM_{2.5}$ Pollution and Policy Implications in Chungcheong Region. Korean Society of Environmental Engineers, 43(6), 407–418, (in Korean with English abstract). https://doi.org/10.4491/KSEE.2021.43.6.407

Hwang, I., Lee, T.-J., Kim, T., Bae, G.-N. (2021) Characteristics of Air Pollutant Emissions and Distribution for Particulate Matter Concentration of Air Pollution Networks in Gyeongsangbuk-do. Korean Society for Atmospheric Environment, 37(3), 536–551. https://doi.org/10.5572/KOSAE.2021.37.3.536

Kim, J., Jang, Y.-K. (2014) Uncertainty Assessment for CAPSS Emission Inventory by DARS. Journal of Korean Society for Atmospheric Environment, 30(1), 26–36. https://doi.org/10.5572/KOSAE.2014.30.1.026

Kim, H.C., Kim, S., Kim, B.U., Jin, C.S., Hong, S., Park, R., Son, S.-W., Bae. C., Bae. M., Song. C.-K., Stein, A. (2017a) Recent increase of surface particulate matter concentrations in the Seoul Metropolitan Area, Korea. Scientific Reports, 7(1), 1–7.

Kim, S., Bae, C., Yoo, C., Kim, B.U., Kim, H.C., Moon, N. (2017b). $PM_{2.5}$ simulations for the Seoul Metropolitan Area: (II) estimation of self-contributions and emission-to-$PM_{2.5}$ conversion rates for each source category. Journal of Korean Society for Atmospheric Environment, 33(4), 377–392. https://doi.org/10.5572/KOSAE.2017.33.4.377

Kim, O., Bae, M., Kim, S. (2020a) Evaluation on Provincial NO_X and SO_2 Emissions in CAPSS 2016 Based on Photochemical Model Simulation. Journal of Korean Society for Atmospheric Environment, 36(1), 64–83. https://doi.org/10.5572/KOSAE.2020.36.1.064

Kim, J.A., Lim, S., Shang, X., Lee, M., Kang, K.S., Ghim, Y.S (2020b). Characteristics of $PM_{2.5}$ chemical composition and high-concentration Episodes observed in Jeju from 2013 to 2016. Journal of Korean Society for Atmospheric Environment, 36(3), 388–403. https://doi.org/10.5572/KOSAE.2020.36.3.388

Kim, S., You, S., Kang, Y.-H., Kim, E., Bae, M., Son, K., Kim, Y., Kim, B.-U., Kim, H. (2021a) Municipality-Level Source Apportionment of $PM_{2.5}$ Concentrations based on the CAPSS 2016: (II) Incheon, Journal of Korean Society for Atmospheric Environment, 37(1), 144–168. https://doi.org/10.5572/KOSAE.2021.37.1.144

Kim, S., You, S., Kim, E., Kang, Y.-H., Bae, M., Son, K. (2021b) Municipality-Level Source Apportionment of $PM_{2.5}$ Concentrations based on the CAPSS 2016: (III) Jeollanamdo. Journal of Korean Society for Atmospheric Environment, 37(2), 206–230. https://doi.org/10.5572/KOSAE.2021.37.2.206

Kim, E., You, S., Bae, M., Kang, Y.-H., Son, K., Kim, S. (2021c) Municipality-Level Source Apportionment of $PM_{2.5}$ Concentrations based on the CAPSS 2016: (IV) Jeollabuk-do. Journal of Korean Society for Atmospheric Environment, 37(2), 292–309. https://doi.org/10.5572/KOSAE.2021.37.2.292

Kim, E., Kim, B.U., Kim, H.C., Kim, S. (2021d) Sensitivity of fine particulate matter concentrations in South Korea to regional ammonia emissions in Northeast Asia. Environmental Pollution, 273, 116428. https://doi.org/10.1016/j.envpol.2021.116428

Kim, M., Kim, J., Lee, Y., Park, S., Oh, B., Cha, J.-D., Kim, J.-B. (2022) Analysis of Emission Characteristics and Estimation of Air Pollutants Emitted from Small Ship. Journal of Korean Society for Atmospheric Environment, 38(2), 258–268. https://doi.org/10.5572/KOSAE.202.38.2.258

Lee, H.J., Song, M.G., Kim, D.K. (2019) Estimation of emissions and emission factor of volatile organic compounds from small-scale dry cleaning operations using organic solvents. Journal of Korean Society for Atmospheric Environment, 35(4), 413–422. https://doi.org/10.5572/KOSAE.2019.35.4.413

NAIR (National Air Emission Inventory and Research) (2021) 2018 national air pollutant emissions, https://www.air.go.kr/jbmd/sub90_detail.do?tabPage=1&detailKey=61027P06&inputSchTxt=&typeSchOption=titleNm&menuId=POT027

NAIR (National Air Emission Inventory and Research) (2022) Handbook of estimation methods for national air pollutant emissions (V), https://www.air.go.kr/jbmd/sub90_detail.do?tabPage=2&detailKey=62017P06&inputSchTxt=&typeSchOption=titleNm&menuId=POT027

Yeo, S.-Y., Lee, H.-K., Choi, S.-W., Seol, S.-H., Jin, H.-A., Yoo, C., Kim, J.-H., Kim, J.-S. (2019) Analysis of the National Air Pollutant Emission Inventory (CAPSS 2015) and the Major Cause of Change in Republic of Korea, Asian Journal of Atmospheric Environment, 13(3), 212–231. https://doi.org/10.5572/ajae.2019.13.3.212

You, S., Bae, C., Kim, C., Yoo, C., Kim, S. (2020) Municipality-Level Source Apportionment of $PM_{2.5}$ Concentrations based on the CAPSS 2016: (I) Gyeonggi Province. Journal of Korean Society for Atmospheric Environment, 36(6), 785–805. https://doi.org/10.5572/KOSAE.2020.36.6.785

APPENDIX

Appendix 1. National Air Pollutant Emission.

(a) Trends in CO emissions (Unit: metric tons/year)

Emission source category		2014	2015	2016	2017	2018	Change (%) (2018–2017)
First-level	Second-level						
Energy production	Public power generation	41,534	33,425	35,515	33,924	36,979	9.0%
	District heating	3,675	3,365	4,242	5,306	7,271	37.0%
	Oil refining	2,320	2,136	1,605	1,862	1,788	−4.0%
	Private power generation	10,327	16,212	17,217	18,212	23,934	31.4%
	Subtotal	57,856	55,138	58,579	59,304	69,972	18.0%
Non-industry	Commercial and public facilities	16,227	16,956	18,896	19,320	19,742	2.2%
	Residential facilities	59,341	54,445	47,997	42,612	37,687	−11.6%
	Agricultural · livestock · fishery facilities	1,026	898	842	784	743	−5.2%
	Subtotal	76,594	72,299	67,735	62,716	58,172	−7.2%
Manufacturing industry	Combustion facilities	1,389	1,608	3,265	3,129	3,505	12.0%
	Process furnaces	6,587	6,607	7,138	7,043	7,070	0.4%
	Others	10,740	8,639	7,767	8,092	9,484	17.2%
	Subtotal	18,716	16,854	18,170	18,263	20,060	9.8%
Industrial process	Petroleum industry	11,545	12,069	12,643	12,879	12,962	0.6%
	Iron and steel industry	5,638	5,761	5,760	5,745	5,834	1.5%
	Inorganic chemical industry	485	487	510	605	520	−14.1%
	Organic chemical industry	5,316	5,011	5,661	5,889	6,000	1.9%
	Pulp and paper industry	2,604	2,469	2,495	2,426	2,351	−3.1%
	Others	267	272	271	205	200	−2.6%
	Subtotal	25,855	26,069	27,340	27,750	27,866	0.4%
Road transport	Passenger cars	136,451	123,534	118,777	114,450	92,483	−19.2%
	Taxis	1,757	1,151	740	639	571	−10.7%
	Vans	3,730	3,203	4,430	3,966	3,724	−6.1%
	Buses	9,451	6,805	6,964	6,825	6,764	−0.9%
	Freight cars	49,976	48,379	49,643	48,360	48,631	0.6%
	Special cars	1,035	830	1,057	968	1,032	6.6%
	RVs	26,634	21,349	22,342	21,104	19,342	−8.3%
	Two-wheeled vehicles	52,190	40,265	40,604	40,840	41,021	0.4%
	Subtotal	281,225	245,516	244,556	237,152	213,568	−9.9%
Non-road transport	Railroads	3,057	2,734	2,426	2,360	2,379	0.8%
	Ships	54,535	60,491	62,632	102,179	118,043	15.5%
	Aircraft	7,117	7,838	8,865	10,370	10,454	0.8%
	Agricultural machinery	7,165	7,097	7,076	7,090	7,038	−0.7%
	Construction machinery	54,229	57,540	55,614	54,456	57,105	4.9%
	Subtotal	126,103	135,700	136,612	176,455	195,020	10.5%
Waste	Waste incineration	1,645	1,548	2,008	2,051	1,954	−4.7%
Others	Forest fires and other fires	6,459	7,197	6,977	8,656	7,556	−12.7%
Biomass burning	Open burning	4,498	4,200	4,080	3,959	3,784	−4.4%
	Crop residue incineration	155,437	157,616	159,196	152,427	143,048	−6.2%
	Grilled meat and fish	12	13	9	11	10	−15.0%
	Wood stoves and boilers	58,938	57,772	57,029	56,066	55,298	−1.4%
	Traditional fireplaces	6,031	5,856	5,750	5,609	5,493	−2.1%
	Charcoal kilns	7,000	7,000	7,000	7,000	7,000	0.0%
	Subtotal	231,917	232,455	233,066	225,073	214,632	−4.6%
Total		826,370	792,776	795,044	817,420	808,801	−1.1%

Appendix 1. Continued.

(b) Trends in NO_X emissions (Unit: metric tons/year)

Emission source category		2014	2015	2016	2017	2018	Change (%) (2018–2017)
First-level	Second-level						
Energy production	Public power generation	127,456	116,250	109,721	77,296	64,830	−16.1%
	District heating	4,651	4,116	4,075	4,349	4,979	14.5%
	Oil refining	8,066	7,818	7,701	8,547	7,881	−7.8%
	Private power generation	22,644	22,634	23,948	24,001	26,731	11.4%
	Subtotal	162,818	150,818	145,445	114,192	104,420	−8.6%
Non-industry	Commercial and public facilities	29,871	32,630	34,249	34,610	34,120	−1.4%
	Residential facilities	47,055	46,605	48,101	48,983	50,447	3.0%
	Agricultural · livestock · fishery facilities	4,216	3,712	3,474	3,210	3,032	−5.5%
	Subtotal	81,143	82,948	85,824	86,803	87,599	0.9%
Manufacturing industry	Combustion facilities	13,612	13,955	17,137	16,201	17,294	6.7%
	Process furnaces	95,197	94,326	98,494	99,775	89,771	−10.0%
	Others	64,852	60,858	59,702	53,814	61,902	15.0%
	Subtotal	173,660	169,139	175,332	169,790	168,967	−0.5%
Industrial process	Petroleum industry	4,478	4,799	4,932	4,322	4,690	8.5%
	Iron and steel industry	38,485	43,671	43,352	42,849	46,077	7.5%
	Inorganic chemical industry	4,284	4,882	2,752	3,353	3,050	−9.0%
	Organic chemical industry	23	16	19	24	29	19.8%
	Others	6,042	6,462	4,877	3,070	3,175	3.4%
	Subtotal	53,311	59,830	55,932	53,618	57,020	6.3%
Road transport	Passenger cars	34,036	36,193	41,190	41,023	36,431	−11.2%
	Taxis	487	363	249	238	221	−7.0%
	Vans	15,346	13,121	17,350	15,451	14,428	−6.6%
	Buses	31,365	34,097	32,011	28,981	25,013	−13.7%
	Freight cars	204,086	206,915	239,450	226,640	210,361	−7.2%
	Special cars	2,482	2,479	2,833	2,494	2,618	4.9%
	RVs	70,509	73,506	116,938	116,175	114,061	−1.8%
	Two-wheeled vehicles	2,919	2,911	2,974	3,037	3,094	1.9%
	Subtotal	361,230	369,585	452,995	434,038	406,227	−6.4%
Non-road transport	Railroads	7,476	6,688	5,932	5,771	5,819	0.8%
	Ships	144,030	151,735	161,826	162,514	155,381	−4.4%
	Aircraft	7,323	8,058	9,104	10,621	10,713	0.9%
	Agricultural machinery	16,288	16,209	16,190	16,351	16,249	−0.6%
	Construction machinery	116,053	121,686	116,934	114,053	119,780	5.0%
	Subtotal	291,171	304,376	309,986	309,309	307,942	−0.4%
Waste	Waste incineration	12,257	11,977	13,570	12,994	12,492	−3.9%
Others	Forest fires and other fires	153	172	167	214	184	−14.0%
Biomass burning	Open burning	590	550	535	519	496	−4.4%
	Crop residue incineration	5,423	5,606	5,816	5,634	5,247	−6.9%
	Grilled meat and fish	9	9	7	8	7	−15.3%
	Wood stoves and boilers	2,205	2,195	2,188	2,179	2,172	−0.3%
	Traditional fireplaces	528	513	504	491	481	−2.1%
	Charcoal kilns	10	10	10	10	10	0.0%
	Subtotal	8,765	8,883	9,059	8,841	8,413	−4.8%
Total		1,144,508	1,157,728	1,248,309	1,189,800	1,153,265	−3.1%

Appendix 1. Continued.

(c) Trends in SO_X emissions (Unit: metric tons/year)

Emission source category: First-level	Emission source category: Second-level	2014	2015	2016	2017	2018	Change (%) (2018–2017)
Energy production	Public power generation	73,506	71,515	71,497	58,900	51,555	−12.5%
	District heating	1,920	1,531	1,425	1,173	669	−42.9%
	Oil refining	13,071	12,405	12,917	12,308	8,984	−27.0%
	Private power generation	6,065	5,791	5,856	5,194	4,659	−10.3%
	Subtotal	94,562	91,243	91,696	77,574	65,868	−15.1%
Non-industry	Commercial and public facilities	6,328	12,015	9,744	8,202	6,045	−26.3%
	Residential facilities	17,111	15,471	13,204	11,500	9,694	−15.7%
	Agricultural · livestock · fishery facilities	1,229	1,249	1,067	1,012	827	−18.3%
	Subtotal	24,668	28,736	24,015	20,714	16,566	−20.0%
Manufacturing industry	Combustion facilities	3,232	2,441	2,727	2,223	2,066	−7.1%
	Process furnaces	19,456	18,811	18,505	16,878	15,955	−5.5%
	Others	60,294	63,847	65,362	53,226	60,845	14.3%
	Subtotal	82,982	85,098	86,593	72,327	78,867	9.0%
Industrial process	Petroleum industry	57,572	57,789	61,756	57,958	58,732	1.3%
	Iron and steel industry	29,600	35,538	39,451	39,024	39,757	1.9%
	Inorganic chemical industry	1,915	1,706	1,178	1,266	1,440	13.8%
	Organic chemical industry	375	448	463	449	455	1.4%
	Pulp and paper industry	129	122	123	120	116	−3.1%
	Others	9,337	9,781	9,762	7,914	6,853	−13.4%
	Subtotal	98,927	105,385	112,734	106,730	107,353	0.6%
Road transport	Passenger cars	63	67	82	97	78	−19.3%
	Taxis	5	7	4	4	4	−4.3%
	Vans	5	5	5	6	4	−24.0%
	Buses	9	11	12	15	11	−26.0%
	Freight cars	69	82	85	101	76	−25.0%
	Special cars	2	2	2	2	2	−1.0%
	RVs	23	27	31	40	32	−19.8%
	Two-wheeled vehicles	8	8	10	12	9	−22.5%
	Subtotal	183	209	231	277	217	−21.7%
Non-road transport	Railroads	191	171	151	147	149	0.9%
	Ships	39,074	38,467	40,429	34,610	28,711	−17.0%
	Aircraft	678	729	802	876	905	3.2%
	Agricultural machinery	4	4	4	6	4	−24.3%
	Construction machinery	45	53	56	71	62	−12.5%
	Subtotal	39,991	39,424	41,443	35,710	29,831	−16.5%
Waste	Waste incineration	1,846	2,119	2,161	2,120	2,202	3.9%
Biomass burning	Grilled meat and fish	2	2	1	2	1	−15.2%
	Wood stoves and boilers	62	60	60	59	58	−1.2%
	Traditional fireplaces	9	9	9	9	8	−2.1%
	Charcoal kilns	8	8	8	8	8	0.0%
	Subtotal	80	79	78	77	76	−1.5%
Total		343,241	352,292	358,951	315,530	300,979	−4.6%

Appendix 1. Continued.

(d) Trends in TSP emissions (Unit: metric tons/year)

Emission source category		2014	2015	2016	2017	2018	Change (%) (2018–2017)
First-level	Second-level						
Energy production	Public power generation	3,976	3,812	3,337	3,147	3,106	−1.3%
	District heating	108	132	149	168	181	7.9%
	Oil refining	169	182	157	148	215	45.0%
	Private power generation	481	565	630	646	803	24.3%
	Subtotal	4,733	4,692	4,273	4,109	4,305	4.8%
Non-industry	Commercial and public facilities	121	184	165	154	128	−17.4%
	Residential facilities	1,447	1,349	1,238	1,152	1,068	−7.4%
	Agricultural · livestock · fishery facilities	340	308	291	265	244	−7.9%
	Subtotal	1,908	1,841	1,694	1,572	1,439	−8.4%
Manufacturing industry	Combustion facilities	449	445	408	237	239	1.2%
	Process furnaces	3,771	3,825	3,196	3,044	3,463	13.7%
	Others	98,518	117,399	119,533	92,535	113,448	22.6%
	Subtotal	102,738	121,668	123,138	95,815	117,150	22.3%
Industrial process	Petroleum industry	466	459	502	511	482	−5.6%
	Iron and steel industry	7,617	7,740	7,797	7,801	7,990	2.4%
	Inorganic chemical industry	635	620	634	771	705	−8.5%
	Organic chemical industry	1,558	1,844	1,911	1,859	1,884	1.4%
	Pulp and paper industry	44	44	44	43	41	−6.0%
	Others	1,847	1,168	1,168	1,111	872	−21.5%
	Subtotal	12,167	11,876	12,056	12,096	11,975	−1.0%
Road transport	Passenger cars	81	88	158	169	137	−19.4%
	Taxis			2	2	2	−4.5%
	Vans	435	328	437	394	377	−4.3%
	Buses	223	234	222	195	181	−7.2%
	Freight cars	6,839	6,694	7,296	6,483	6,178	−4.7%
	Special cars	74	58	97	65	63	−2.9%
	RVs	2,367	2,182	2,307	2,085	1,840	−11.8%
	Motorcycles			78	79	80	1.3%
	Subtotal	10,019	9,583	10,596	9,473	8,858	−6.5%
Non-road transport	Railroads	484	433	384	374	377	0.9%
	Ships	6,983	7,091	7,589	8,290	8,973	8.2%
	Aircraft	89	94	103	109	109	0.4%
	Agricultural machinery	1,364	1,348	1,342	1,340	1,330	−0.8%
	Construction machinery	5,945	6,354	6,173	6,086	6,448	5.9%
	Subtotal	14,865	15,320	15,592	16,198	17,236	6.4%
Waste	Waste incineration	335	340	406	377	338	−10.5%
Others	Forest fires and other fires	428	498	481	679	560	−17.4%
Fugitive Dust	Paved roads	140,840	143,644	152,599	161,824	163,640	1.1%
	Construction	40,356	55,714	51,005	53,284	55,488	4.1%
	Vacant lands	27,519	27,403	24,712	20,979	21,645	3.2%
	Loading and unloading	25	26	26	27	25	−5.0%
	Agricultural production	29,553	29,072	28,549	27,845	27,778	−0.2%
	Livestock production	29,745	30,524	31,898	32,734	33,898	3.6%
	Waste disposal	12,655	14,414	15,498	15,902	16,585	4.3%
	Unpaved roads	115,250	107,445	108,400	109,825	108,856	−0.9%
	Subtotal	395,944	408,242	412,686	422,420	427,916	1.3%
Biomass burning	Open burning	1,438	1,342	1,304	1,265	1,209	−4.4%
	Crop residue incineration	22,085	22,126	22,832	22,079	20,139	−8.8%
	Grilled meat and fish	606	626	461	565	491	−13.2%
	Wood stoves and boilers	4,173	4,072	4,008	3,924	3,857	−1.7%
	Traditional fireplaces	173	168	165	161	157	−2.1%
	Charcoal kilns	1,849	1,849	1,849	1,849	1,849	0.0%
	Subtotal	30,323	30,183	30,618	29,843	27,703	−7.2%
Total		573,460	604,243	611,539	592,582	617,481	4.2%

Appendix 1. Continued.

(e) Trends in PM_{10} emissions (Unit: metric tons/year)

Emission source category: First-level	Emission source category: Second-level	2014	2015	2016	2017	2018	Change (%) (2018–2017)
Energy production	Public power generation	3,854	3,681	3,194	3,041	3,011	−1.0%
	District heating	85	113	133	152	171	12.2%
	Oil refining	104	57	53	53	69	30.5%
	Private power generation	465	544	571	583	724	24.2%
	Subtotal	4,508	4,394	3,951	3,829	3,975	3.8%
Non-industry	Commercial and public facilities	112	170	152	144	119	−17.2%
	Residential facilities	1,206	1,129	1,048	987	926	−6.2%
	Agricultural · livestock · fishery facilities	312	283	267	243	224	−7.9%
	Subtotal	1,629	1,582	1,468	1,374	1,269	−7.7%
Manufacturing industry	Combustion facilities	323	249	240	132	134	2.0%
	Process furnaces	2,282	2,290	1,955	1,855	2,122	14.4%
	Others	57,370	68,354	69,599	53,886	66,058	22.6%
	Subtotal	59,975	70,893	71,794	55,872	68,315	22.3%
Industrial process	Petroleum industry	135	133	145	148	139	−5.6%
	Iron and steel industry	4,755	4,833	4,856	4,856	4,981	2.6%
	Inorganic chemical industry	359	348	356	435	399	−8.3%
	Organic chemical industry	795	940	975	948	961	1.4%
	Pulp and paper industry	27	27	26	26	25	−6.0%
	Others	337	377	373	346	253	−26.7%
	Subtotal	6,407	6,658	6,731	6,759	6,758	0.0%
Road transport	Passenger cars	81	88	158	169	137	−19.4%
	Taxis			2	2	2	−4.5%
	Vans	435	328	437	394	377	−4.3%
	Buses	223	234	222	195	181	−7.2%
	Freight cars	6,839	6,694	7,296	6,483	6,178	−4.7%
	Special cars	74	58	97	65	63	−2.9%
	RVs	2,367	2,182	2,307	2,085	1,840	−11.8%
	Motorcycles			78	79	80	1.3%
	Subtotal	10,019	9,583	10,596	9,473	8,858	−6.5%
Non-road transport	Railroads	484	433	384	374	377	0.9%
	Ships	6,983	7,091	7,589	8,290	8,973	8.2%
	Aircraft	85	90	99	104	105	0.4%
	Agricultural machinery	1,364	1,348	1,342	1,340	1,330	−0.8%
	Construction machinery	5,945	6,354	6,173	6,086	6,448	5.9%
	Subtotal	14,861	15,317	15,588	16,194	17,232	6.4%
Waste	Waste incineration	247	246	295	274	245	−10.6%
Others	Forest fires and other fires	272	317	306	431	356	−17.4%
Fugitive Dust	Paved roads	27,034	27,573	29,291	31,062	31,411	1.1%
	Construction	27,685	38,221	34,990	36,553	38,065	4.1%
	Vacant lands	10,733	10,687	9,638	8,182	8,442	3.2%
	Loading and unloading	9	9	9	9	9	−5.0%
	Agricultural production	10,141	9,961	9,791	9,596	9,572	−0.2%
	Livestock production	9,939	10,200	10,658	10,938	11,325	3.5%
	Waste disposal	3,416	3,926	4,220	4,335	4,473	3.2%
	Unpaved roads	9,715	9,057	9,137	9,257	9,176	−0.9%
	Subtotal	98,671	109,633	107,735	109,932	112,472	2.3%
Biomass burning	Open burning	984	919	893	866	828	−4.4%
	Crop residue incineration	9,121	9,183	9,474	9,150	8,471	−7.4%
	Grilled meat and fish	606	626	461	565	491	−13.2%
	Wood stoves and boilers	2,002	1,958	1,930	1,893	1,864	−1.5%
	Traditional fireplaces	114	111	109	106	104	−2.1%
	Charcoal kilns	1,757	1,757	1,757	1,757	1,757	0.0%
	Subtotal	14,583	14,552	14,623	14,338	13,514	−5.7%
Total		211,172	233,177	233,085	218,476	232,993	6.6%

Appendix 1. Continued.

(f) Trends in $PM_{2.5}$ emissions (Unit: metric tons/year)

Emission source category		2014	2015	2016	2017	2018	Change (%) (2018–2017)
First-level	Second-level						
Energy production	Public power generation	3,162	2,989	2,593	2,470	2,454	−0.7%
	District heating	63	99	120	140	166	18.4%
	Oil refining	46	23	23	25	29	16.6%
	Private power generation	407	496	517	526	659	25.2%
	Subtotal	3,679	3,607	3,253	3,162	3,308	4.6%
Non-industry	Commercial and public facilities	72	109	98	96	81	−15.5%
	Residential facilities	782	745	721	694	673	−3.1%
	Agricultural · livestock · fishery facilities	191	171	159	144	135	−6.4%
	Subtotal	1,045	1,025	978	935	890	−4.9%
Manufacturing industry	Combustion facilities	165	121	148	101	102	0.3%
	Process furnaces	1,245	1,226	1,059	1,006	1,155	14.8%
	Others	28,912	34,971	35,577	27,393	33,842	23.5%
	Subtotal	30,322	36,317	36,785	28,501	35,099	23.2%
Industrial process	Petroleum industry	30	29	32	32	31	−5.6%
	Iron and steel industry	3,636	3,705	3,730	3,729	3,825	2.6%
	Inorganic chemical industry	202	194	199	244	224	−8.2%
	Organic chemical industry	715	846	877	853	865	1.4%
	Pulp and paper industry	17	18	17	17	16	−8.0%
	Others	303	340	336	311	229	−26.4%
	Subtotal	4,903	5,132	5,191	5,186	5,189	0.1%
Road transport	Passenger cars	75	81	145	156	126	−19.4%
	Taxis			2	2	2	−4.5%
	Vans	400	302	402	363	347	−4.3%
	Buses	205	215	204	179	166	−7.2%
	Freight cars	6,292	6,159	6,712	5,964	5,683	−4.7%
	Special cars	68	53	89	60	58	−2.9%
	RVs	2,178	2,008	2,123	1,918	1,693	−11.8%
	Motorcycles			72	73	74	1.3%
	Subtotal	9,218	8,817	9,748	8,715	8,149	−6.5%
Non-road transport	Railroads	446	399	354	344	347	0.9%
	Ships	6,423	6,539	6,995	7,731	8,383	8.4%
	Aircraft	78	83	91	96	96	0.4%
	Agricultural machinery	1,255	1,240	1,235	1,233	1,223	−0.8%
	Construction machinery	5,469	5,846	5,679	5,599	5,932	5.9%
	Subtotal	13,671	14,106	14,354	15,002	15,981	6.5%
Waste	Waste incineration	204	209	252	234	209	−10.5%
Others	Forest fires and other fires	245	285	275	388	320	−17.4%
Fugitive dust	Paved roads	6,541	6,671	7,087	7,515	7,599	1.1%
	Construction	2,769	3,822	3,499	3,655	3,807	4.1%
	Vacant lands	1,610	1,603	1,446	1,227	1,266	3.2%
	Loading and unloading	1	1	1	1	1	−5.0%
	Agricultural production	2,028	1,992	1,958	1,919	1,914	−0.2%
	Livestock production	1,840	1,861	1,960	2,013	2,073	3.0%
	Waste disposal	342	393	422	433	447	3.2%
	Unpaved roads	971	906	914	926	918	−0.9%
	Subtotal	16,101	17,248	17,286	17,690	18,025	1.9%
Biomass burning	Open burning	873	815	792	768	734	−4.4%
	Crop residue incineration	7,563	7,621	7,878	7,627	7,046	−7.6%
	Grilled meat and fish	556	574	423	518	451	−13.0%
	Wood stoves and boilers	1,326	1,298	1,280	1,257	1,238	−1.5%
	Traditional fireplaces	92	89	87	85	83	−2.1%
	Charcoal kilns	1,664	1,664	1,664	1,664	1,664	0.0%
	Subtotal	12,073	12,060	12,124	11,919	11,217	−5.9%
Total		91,460	98,806	100,247	91,731	98,388	7.3%

Appendix 1. Continued.

(g) Trends in Black Carbon emissions (Unit: metric tons/year)

Emission source category: First-level	Emission source category: Second-level	2014	2015	2016	2017	2018	Change (%) (2018–2017)
Energy production	Public power generation	219	146	151	128	148	15.9%
	District heating	17	28	36	45	63	40.0%
	Oil refining	5	1	2	3	2	−36.5%
	Private power generation	83	132	141	143	192	34.6%
	Subtotal	324	307	330	319	405	27.2%
Non-industry	Commercial and public facilities	9	13	13	15	13	−9.6%
	Residential facilities	130	128	136	140	147	4.8%
	Agricultural · livestock · fishery facilities	16	14	13	12	11	−5.9%
	Subtotal	156	155	161	167	172	2.8%
Manufacturing industry	Combustion facilities	20	14	35	30	29	−2.0%
	Process furnaces	74	60	62	64	75	16.3%
	Others	554	666	679	526	649	23.4%
	Subtotal	648	741	776	620	753	21.4%
Industrial process	Petroleum industry	0.02	0.02	0	0.02	0.02	−5.6%
	Iron and steel industry	11	11	11	11	11	2.5%
	Pulp and paper industry	0.1	0.1	0.1	0.1	0.04	−33.3%
	Others	4	5	6	6	4	−32.0%
	Subtotal	15	16	17	17	15	−9.7%
Road transport	Passenger cars	33	39	60	66	48	−26.2%
	Vans	240	183	237	214	204	−4.6%
	Buses	158	166	157	138	128	−7.2%
	Freight cars	3,939	3,873	4,187	3,749	3,538	−5.6%
	Special cars	52	41	69	46	45	−2.9%
	RVs	1,252	1,154	1,219	1,102	971	−11.8%
	Subtotal	5,674	5,456	5,930	5,315	4,935	−7.1%
Non-road transport	Railroads	344	308	273	265	267	0.9%
	Ships	1,004	1,042	1,105	1,141	1,154	1.1%
	Aircraft	61	64	70	74	74	0.4%
	Agricultural machinery	968	956	953	951	943	−0.8%
	Construction machinery	4,218	4,509	4,380	4,318	4,575	5.9%
	Subtotal	6,594	6,879	6,781	6,749	7,014	3.9%
Waste	Waste incineration	3	3	4	4	3	−10.5%
Others	Forest fires and others	11	15	14	24	19	−24.1%
Fugitive Dust	Paved roads	68	70	74	79	79	1.1%
	Vacant lands	0.3	0.3	0.3	0.2	0.3	3.2%
	Loading and unloading	0.04	0.04	0.04	0.04	0.04	−5.0%
	Agricultural production	0.4	0.4	0.4	0.4	0.4	−0.2%
	Livestock production	28	27	30	30	31	1.8%
	Unpaved roads	11	10	10	10	10	−0.9%
	Subtotal	108	108	115	120	121	1.1%
Biomass burning	Open burning	37	34	33	32	31	−4.4%
	Crop residue incineration	1,707	1,709	1,738	1,687	1,599	−5.2%
	Grilled meat and fish	23	23	17	21	18	−13.0%
	Wood stoves and boilers	219	213	210	206	202	−1.7%
	Traditional fireplaces	13	13	13	12	12	−2.1%
	Charcoal kilns	263	263	263	263	263	0.0%
	Subtotal	2,261	2,255	2,274	2,221	2,125	−4.3%
Total		15,795	15,934	16,401	15,555	15,562	0.04%

Appendix 1. Continued.

(h) Trends in VOCs emissions (Unit: metric tons/year)

Emission source category		2014	2015	2016	2017	2018	Change (%) (2018–2017)
First-level	Second-level						
Energy production	Public power generation	5,486	4,497	4,832	4,327	4,774	10.3%
	District heating	509	472	591	732	995	35.9%
	Oil refining	318	327	296	269	208	−22.8%
	Private power generation	1,384	2,169	2,282	2,425	3,185	31.3%
	Subtotal	7,697	7,464	8,001	7,753	9,161	18.2%
Non-industry	Commercial and public facilities	722	795	810	820	817	−0.4%
	Residential facilities	1,777	1,773	1,879	1,963	2,075	5.7%
	Agricultural · livestock · fishery facilities	59	53	51	47	44	−6.3%
	Subtotal	2,558	2,622	2,740	2,830	2,936	3.7%
Manufacturing industry	Combustion facilities	193	222	447	428	476	11.2%
	Process furnaces	1,134	1,079	1,176	1,166	1,182	1.3%
	Others	1,953	1,800	1,719	1,606	1,922	19.7%
	Subtotal	3,280	3,101	3,342	3,199	3,579	11.9%
Industrial process	Petroleum industry	53,588	56,021	58,686	59,780	60,165	0.6%
	Iron and steel industry	19,325	19,408	19,546	19,756	20,117	1.8%
	Inorganic chemical industry	579	564	613	741	679	−8.4%
	Organic chemical industry	44,050	44,417	45,508	45,856	45,457	−0.9%
	Pulp and paper industry	1	1	1	1	1	0.0%
	Food and beverage industry	62,275	61,943	61,206	61,780	61,429	−0.6%
	Others	534	544	543	410	399	−2.6%
	Subtotal	180,351	182,899	186,104	188,324	188,247	0.0%
Energy transport and storage	Gasoline supply	27,645	29,137	30,160	30,695	30,770	0.2%
Solvents use	Painting facilities	339,582	344,671	347,608	348,822	334,364	−4.1%
	Cleaning facilities	27,701	28,144	27,740	27,442	27,074	−1.3%
	Laundry facilities	21,304	20,407	20,390	20,250	20,464	1.1%
	Other solvent use	160,731	162,137	162,266	167,134	165,451	−1.0%
	Subtotal	549,318	555,359	558,004	563,648	547,353	−2.9%
Road transport	Passenger cars	18,045	16,071	15,877	15,315	13,984	−8.7%
	Taxis	89	61	38	33	28	−13.7%
	Vans	632	531	669	629	576	−8.5%
	Buses	12,134	12,366	11,936	11,447	10,833	−5.4%
	Freight cars	11,436	11,514	12,700	12,149	11,899	−2.1%
	Special cars	266	246	317	285	278	−2.5%
	RVs	2,610	2,384	3,017	3,027	2,999	−0.9%
	Two-wheeled vehicles	4,255	2,973	3,008	3,036	3,061	0.8%
	Subtotal	49,468	46,145	47,561	45,920	43,658	−4.9%
Non-road transport	Railroads	1,225	1,095	973	948	954	0.7%
	Ships	18,340	20,970	22,185	41,064	48,961	19.2%
	Aircraft	672	700	749	834	789	−5.4%
	Agricultural machinery	1,955	1,933	1,925	1,917	1,902	−0.8%
	Construction machinery	14,681	15,613	14,984	14,645	15,261	4.2%
	Subtotal	36,873	40,311	40,816	59,407	67,867	14.2%
Waste	Waste incineration	44,612	53,173	55,520	55,366	54,770	−1.1%
	Others	3,449	3,901	3,468	3,039	2,965	−2.4%
	Subtotal	48,061	57,074	58,988	58,405	57,735	−1.1%
Others	Forest fires and other fires	551	648	624	901	737	−18.2%
Biomass burning	Open burning	4,807	4,488	4,361	4,231	4,044	−4.4%
	Crop residue incineration	61,154	61,408	63,497	62,729	60,279	−3.9%
	Grilled meat and fish	147	154	110	137	119	−13.3%
	Wood stoves and boilers	17,406	17,071	16,858	16,581	16,361	−1.3%
	Traditional fireplaces	1,687	1,638	1,608	1,569	1,536	−2.1%
	Charcoal kilns	1,254	1,254	1,254	1,254	1,254	0.0%
	Subtotal	86,454	86,012	87,687	86,500	83,592	−3.4%
Total		992,256	1,010,771	1,024,029	1,047,585	1,035,636	−1.1%

Appendix 1. Continued.

(i) Trends in NH_3 emissions (Unit: metric tons/year)

Emission source category		2014	2015	2016	2017	2018	Change (%) (2018–2017)
First-level	Second-level						
Energy production	Public power generation	798	557	708	440	534	21.4%
	District heating	145	128	158	192	263	37.1%
	Oil refining	174	198	177	151	106	−30.1%
	Private power generation	308	496	516	547	723	32.2%
	Subtotal	1,425	1,379	1,559	1,330	1,626	22.3%
Non-industry	Commercial and public facilities	498	567	582	580	538	−7.3%
	Residential facilities	618	641	698	723	757	4.6%
	Agricultural · livestock · fishery facilities	164	143	134	125	119	−5.2%
	Subtotal	1,280	1,351	1,415	1,429	1,414	−1.1%
Manufacturing industry	Combustion facilities	57	67	130	122	134	9.6%
	Process furnaces	229	233	254	245	250	1.9%
	Others	431	327	288	320	353	10.2%
	Subtotal	717	627	672	688	737	7.1%
Industrial process	Petroleum industry	22,368	23,384	24,496	24,953	25,113	0.6%
	Iron and steel industry	1,691	1,728	1,728	1,724	1,750	1.5%
	Ammonia consumption	13,984	14,320	16,265	16,301	19,117	17.3%
	Subtotal	38,043	39,432	42,489	42,977	45,981	7.0%
Road transport	Passenger cars	9,906	9,863	4,554	3,914	2,800	−28.5%
	Taxis			102	104	99	−4.3%
	Vans	8	7	18	16	14	−13.6%
	Buses	12	14	27	29	29	2.8%
	Freight cars	83	88	162	160	157	−2.0%
	Special cars	2	2	3	3	4	58.7%
	RVs	52	56	154	160	166	3.8%
	Two-wheeled vehicles	49	50	51	52	52	1.4%
	Subtotal	10,113	10,078	5,071	4,437	3,322	−25.1%
Non-road transport	Railroads	14	12	11	10	11	1.5%
	Ships	13	14	14	15	14	−3.8%
	Agricultural machinery	53	53	53	54	53	−0.6%
	Construction machinery	36	38	39	41	47	15.0%
	Subtotal	116	117	117	120	126	4.5%
Waste	Others	23	22	22	22	22	−1.9%
Agriculture	Fertilizer use	20,172	19,901	19,553	17,754	19,566	10.2%
	Livestock manure management	207,781	211,362	217,464	226,582	230,211	1.6%
	Subtotal	227,953	231,263	237,017	244,335	249,777	2.2%
Others	Animals	12,832	12,882	12,924	12,945	12,957	0.1%
Biomass burning	Open burning	2	2	2	2	2	−4.4%
	Crop residue incineration	5	5	5	5	5	−6.0%
	Wood stoves and boilers	6	6	6	6	6	−1.2%
	Traditional fireplaces	2	2	2	2	2	−2.1%
	Subtotal	16	15	15	15	14	−3.4%
Total		292,517	297,167	301,301	308,298	315,975	2.5%

SUPPLEMENTARY MATERIALS

Table S1. Configurations of (A)WRF and (B)CMAQ models in this study1

(A) WRF	Description
Version	WRF v3.4.1
Microphysics	WSM6
Short wave radiation	Dudhia
Land-Surface Model	NOAH
PBL scheme	YSU

(B) CMAQ	Description
Version	Version 4.7.1
Chemical Mechanism	SAPRC99
Chemical Solver	EBI
Aerosol Module	AERO5
Boundary Condition	Default profile
Advection Scheme	YAMO
Horizontal Diffusion	Multiscale
Vertical Diffusion	Eddy

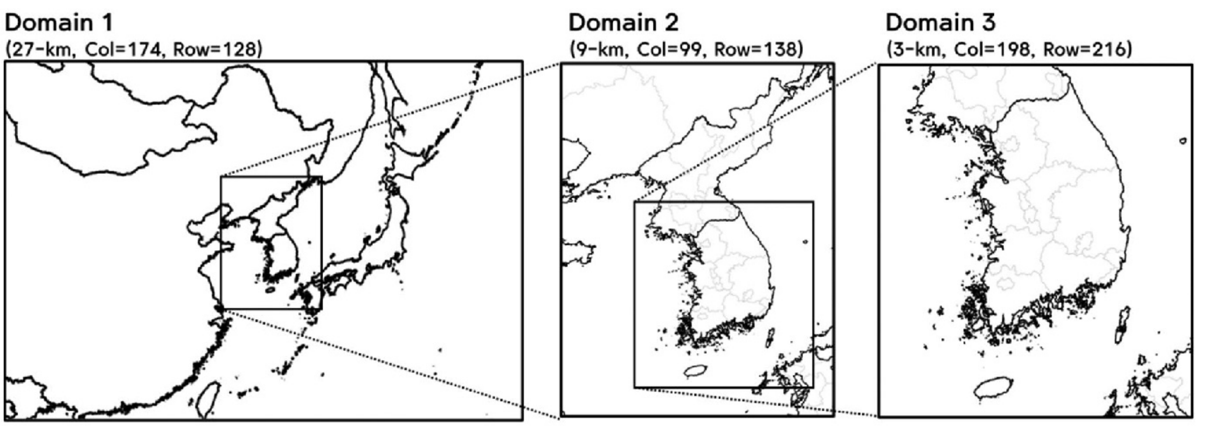


Fig. S1. Horizontal resolutions for the simulation by domain were as follows: 27 km (Domain 1), 9 km (Domain 2), and 3 km (Domain 3).