Supplementary Information for

Impact of shipping emissions regulation: Urban aerosol composition changes revealed by receptor and numerical modelling

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Supplementary Tables

Supplementary Table 1. Information on study sites and observed data.

Site Information				Data information		
Site name	Longitude	Latitude	Environment type	Measurements		Temporal resolution
Buk-Hang	129.055280	35.122570	Port	AQM	6 species	hourly
Site A	129.055080	35.125586		SIFT-MS	70 trace gases	every 40 sec.
Site B	129.048766	35.114855		SIFT-MS	70 trace gases	every 40 sec.
Sin-Hang	128.834390	35.074690		AQM	6 species	hourly
				$PM_{2.5}$ speciation	8 ionic and 2 carbonaceous species	hourly
					23 metallic species	daily
Yeon-San	129.074590	35.181150	Urban background	AQM	6 species	hourly
				$PM_{2.5}$ speciation	8 ionic and 2 carbonaceous species	hourly
					23 metallic species	daily

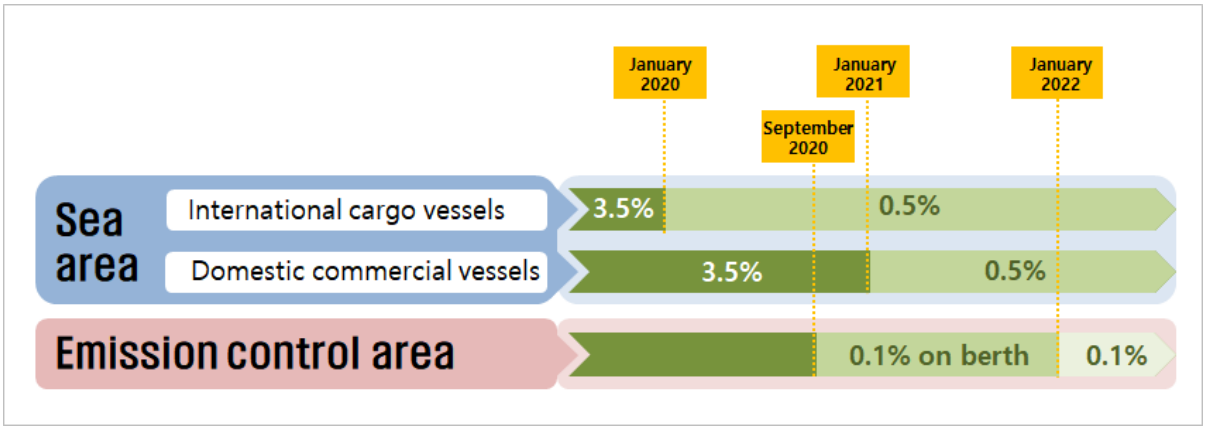
Supplementary Table 2. Hourly mean concentrations (μg m^{-3}) of trace gases measured by a mobile laboratory equipped with selected ion flow tube mass spectrometery (SIFT-MS) at near port site (*Site A*) (n = 997).

Group	Species	Concentrations (Mean ± S.D.)	Group	Species	Concentrations (Mean ± S.D.)
alkane	Methane	2874.10 ± 714.64	halogenated	Vinyl chloride; 1,2-dichloroethane	1.34 ± 0.79
	Ethane; Ethene	25.06 ± 15.56		1,1,1-trichloroethane	3.96 ± 2.31
	Propane	1036.62 ± 633.37		1,1,2-trichloroethane	12.03 ± 7.36
	Butane	39.77 ± 15.54		Ethyl chloride; 1,1-dichloroethane	8.87 ± 7.37
	Pentane (iso+n)	87.03 ± 62.65		Dichloropropene	1.07 ± 0.95
	Hexane	23.53 ± 17.68		1,2-dichloropropane	10.51 ± 4.86
	Heptane	2.69 ± 1.82		1,2,4-trichlorobenzene	1.41 ± 1.85
	Octane (iso+n)	9.79 ± 7.68		1,4-dichlorobenzene	0.55 ± 0.46
	Nonane	1.58 ± 1.40		Chlorobenzene	2.32 ± 1.61
	Decane	2.72 ± 2.15		1-chlorotoluene	2.06 ± 1.26
	Undecane	3.34 ± 3.05		Freon11; Freon113; Freon114	5.65 ± 4.12
	Dodecane	1.73 ± 3.20		Freon12; Freon114	17.39 ± 12.02
	Cyclopentane	6.27 ± 4.39		Carbon tetrachloride	4.69 ± 3.85
	Cyclohexane	11.02 ± 9.36		Tetrachloroethylene	3.51 ± 2.87
	2-methylpentane	3.45 ± 3.28	others	Hexachlorobutadiene	0.70 ± 2.82
alkene	Propene	1.69 ± 1.49		Carbonyl sulfide	3.44 ± 1.87
	2-butene	1.66 ± 1.15		Dimethyl sulfide	0.80 ± 0.46
	1-pentene	4.59 ± 3.21		Carbon disulfide	8.18 ± 6.03
	1-hexene	3.48 ± 2.26		Sulfuric acid	29.34 ± 16.43
	1,3-butadiene	0.26 ± 0.47		Methyl mercaptan	0.60 ± 0.60
alkyne	Acetylene	0.25 ± 0.52		Acetonitrile	0.79 ± 0.72
biogenic	Isoprene	0.26 ± 0.18		Ammonia	8.93 ± 8.72
	Myrcene	1.52 ± 1.54	oxygenated	Methanol	9.93 ± 6.29
aromatic	Benzene; Dimethyl sulfoxide	2.55 ± 4.19		Ethanol; Formic acid	16.96 ± 15.68
	Toluene	8.08 ± 8.29		Formaldehyde	9.23 ± 3.94
	Xylenes + Ethylbenzene	7.80 ± 13.33		Acetaldehyde	7.67 ± 5.88
	Styrene	0.76 ± 0.57		Propanal; Propylene oxide	5.24 ± 2.84
	C3-alkylbenzene	4.10 ± 3.97		Acetone	8.62 ± 3.85
	Naphthalene	1.37 ± 1.05		Butanone	3.49 ± 3.38
halogenated	Bromoform	0.89 ± 1.16		Methyl isobutyl ketone	1.56 ± 1.09
	Chloroform; Dichloromethane; Tetrachloroethane	20.61 ± 13.94		Tetrahydrofuran	6.32 ± 5.30
	Methyl bromide	11.12 ± 7.49		Methyl tert-butyl ether	8.54 ± 6.09
	1,2-dibromoethane	4.54 ± 3.61		Vinyl acetate	2.94 ± 2.39
	Trichloroethylene	1.24 ± 1.14		Acetic acid	6.45 ± 3.89
	Dichloroethene (1,1 and 1,2)	10.32 ± 6.33		Butanoic acid	1.49 ± 1.12

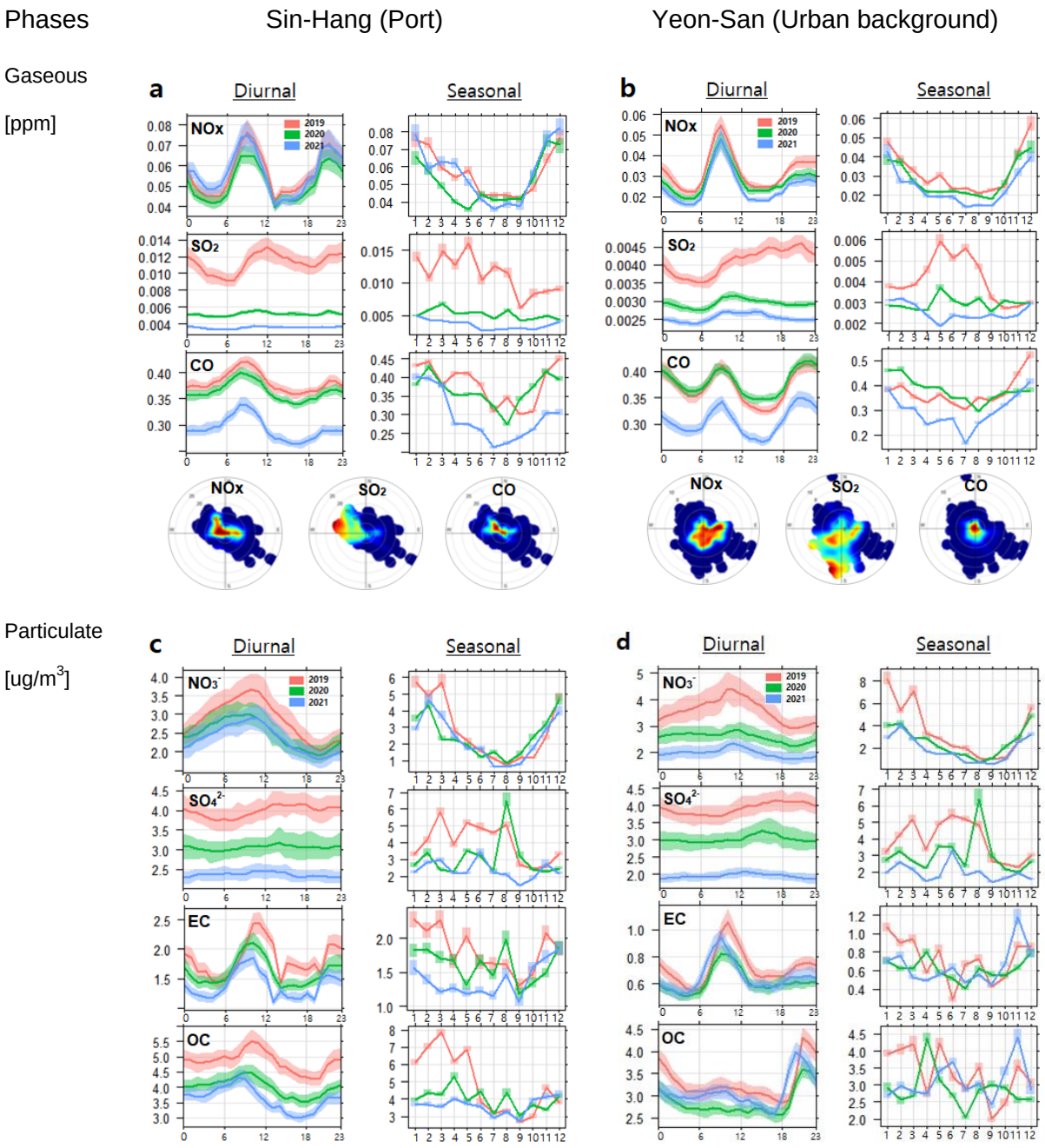
Supplementary Table 3. CMAQ scenarios for emission control in the shipping sector.

Scenarios	Emission controlling design
Case 0	Original baseline scenario
Case 1	SO_2 emissions reduction by 85.7% from switch to low-sulphur fuels (3.5% to 0.5%)
Case 2	SO_2 emissions reduction by 97.1% from switch to low-sulphur fuels (3.5% to 0.1%)
Case 3	SO_2 emissions reduction by 97.1% and shipping emission reduction of 6 species (SOx, NOx, VOC, NH_3, CO and primary $PM_{2.5}$) by 30%
Case 4	Exception of NOx emission reduction from scenario Case 3
Case 5	Exception of VOC emission reduction from scenario Case 3

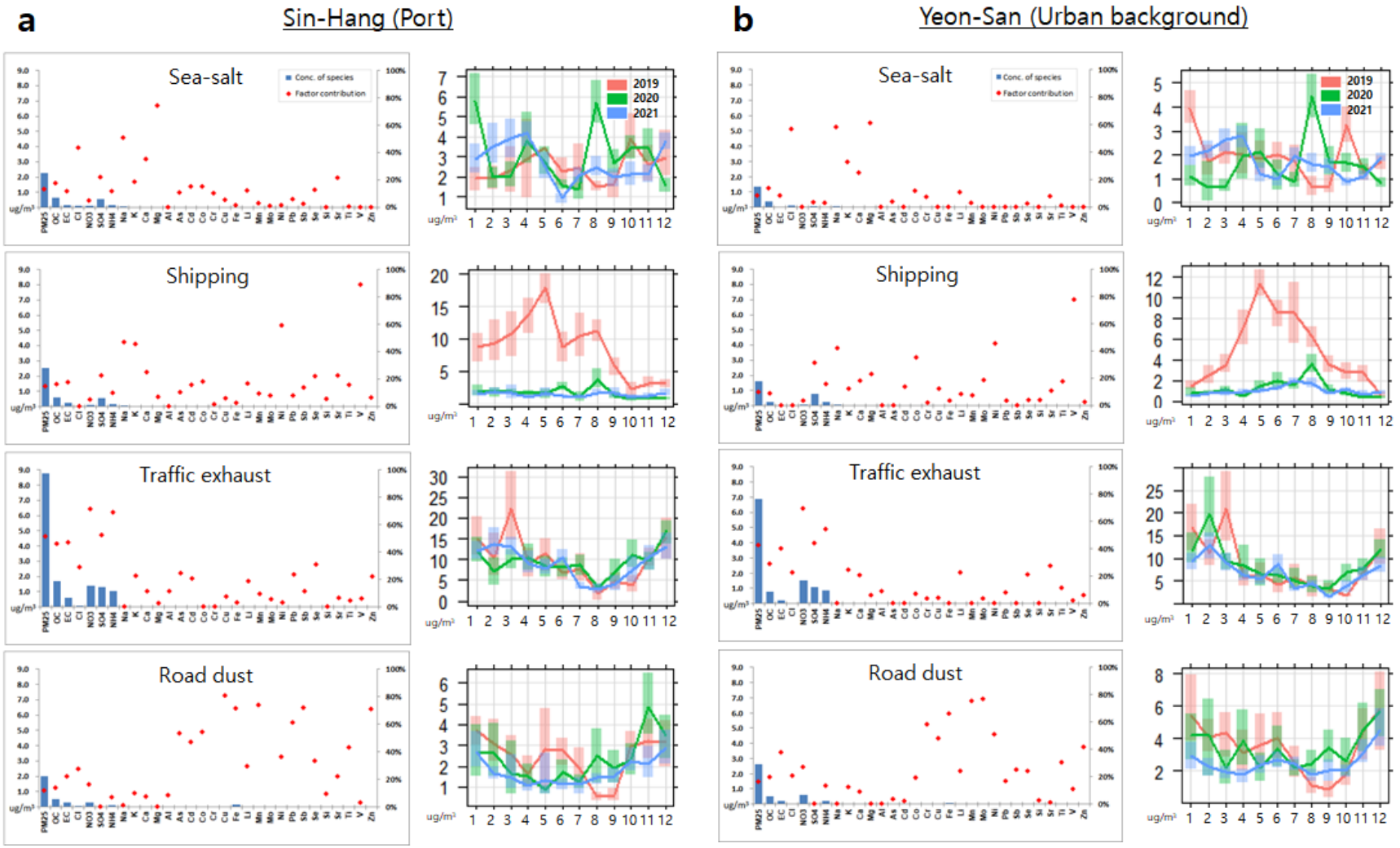
Supplementary Figures

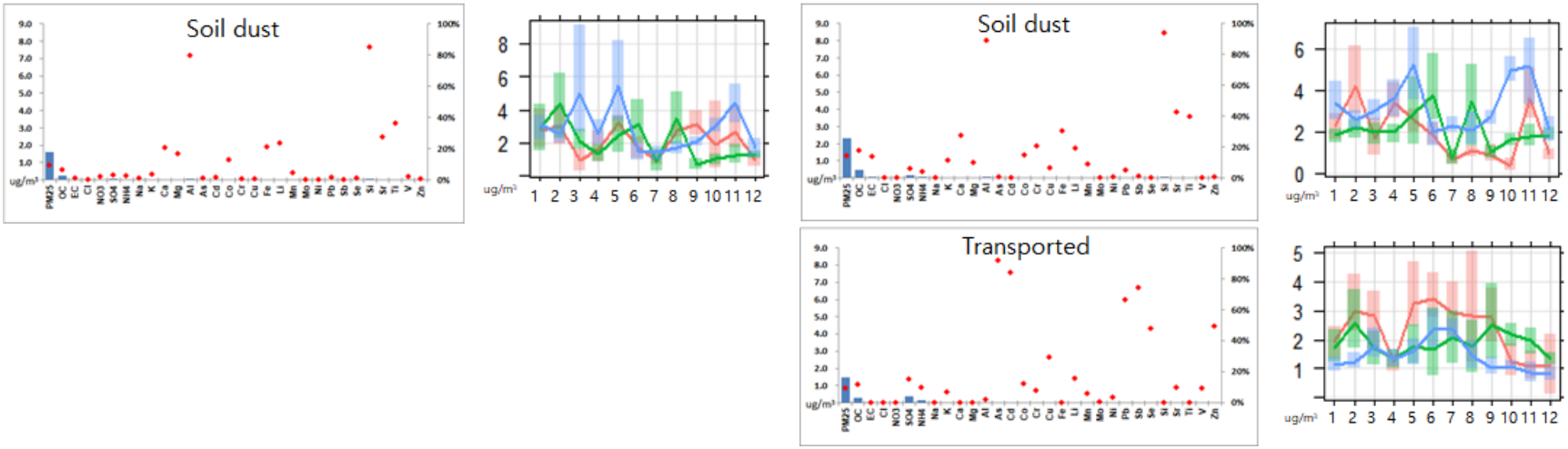


Supplementary Figure 1. History of sulphur (S) content regulations on shipping fuel oils in Busan.

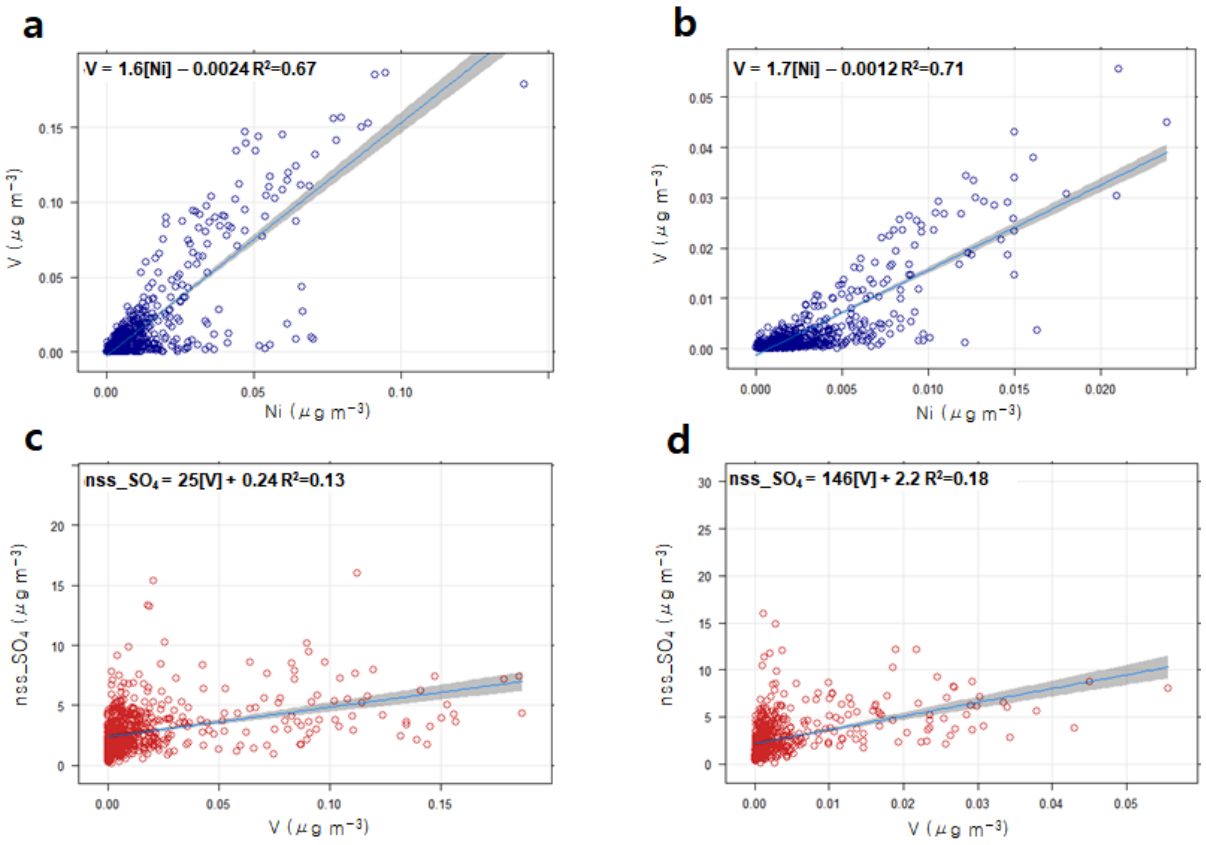


Supplementary Figure 2. Temporal/seasonal and spatial distributions of ambient pollution level. Concentrations of gaseous species derived from air quality monitoring stations (**a)** at port site (Sin-Hang) and (**b)** at urban background site (Yeon-San). Levels of $PM_{2.5}$ elements derived from $PM_{2.5}$ speciation network **(c)** at Sin-Hang and **(d)** at Yeon-San.

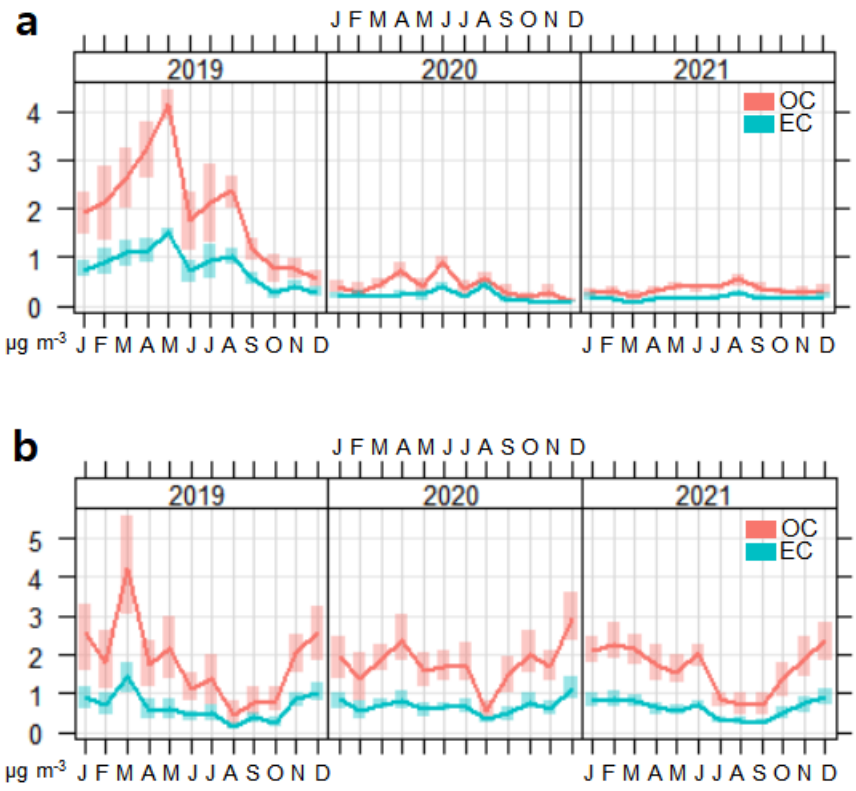

a
Sin-Hang (Port)
b
Yeon-San (Urban background)
Sea-salt
Shipping
Traffic exhaust
Road dust
Conc. of species
Factor contribution
2019
2020
2021
ug/m³



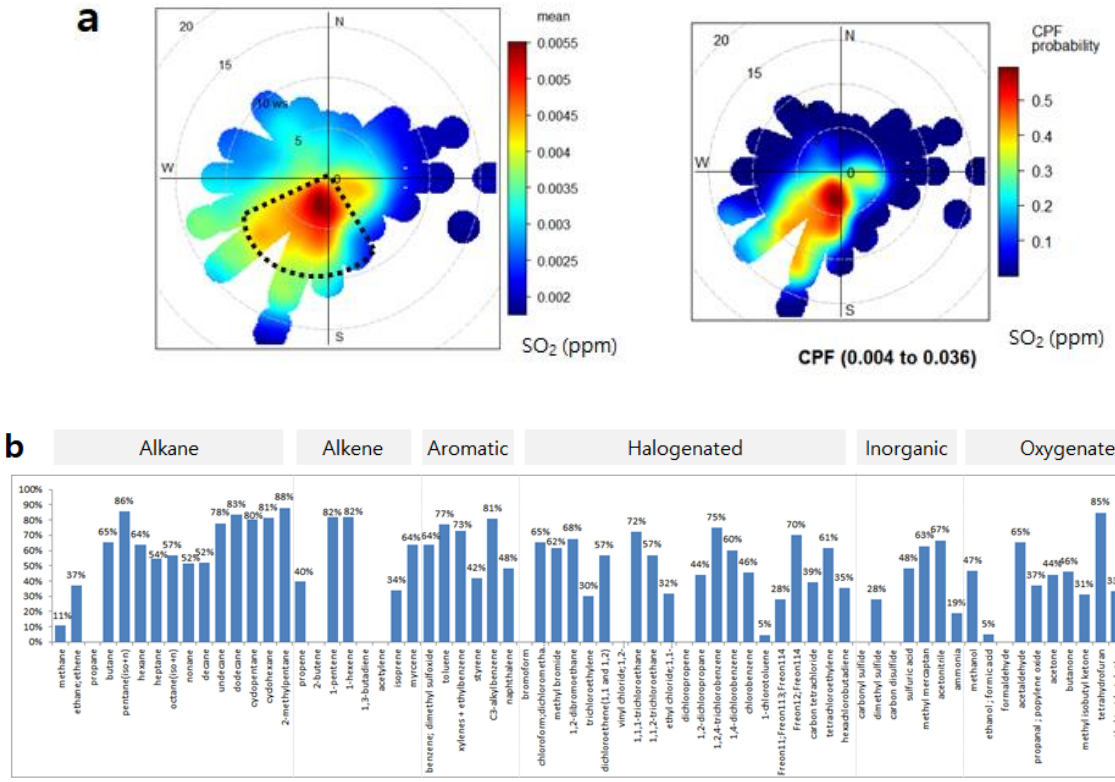
Supplementary Figure 3. PMF factor profiles for $PM_{2.5}$ and source-specific monthly variations. (a) $PM_{2.5}$ levels of the port dataset are determined by five major factors, and **(b)** the urban background dataset is explained by a six factor solution**.**



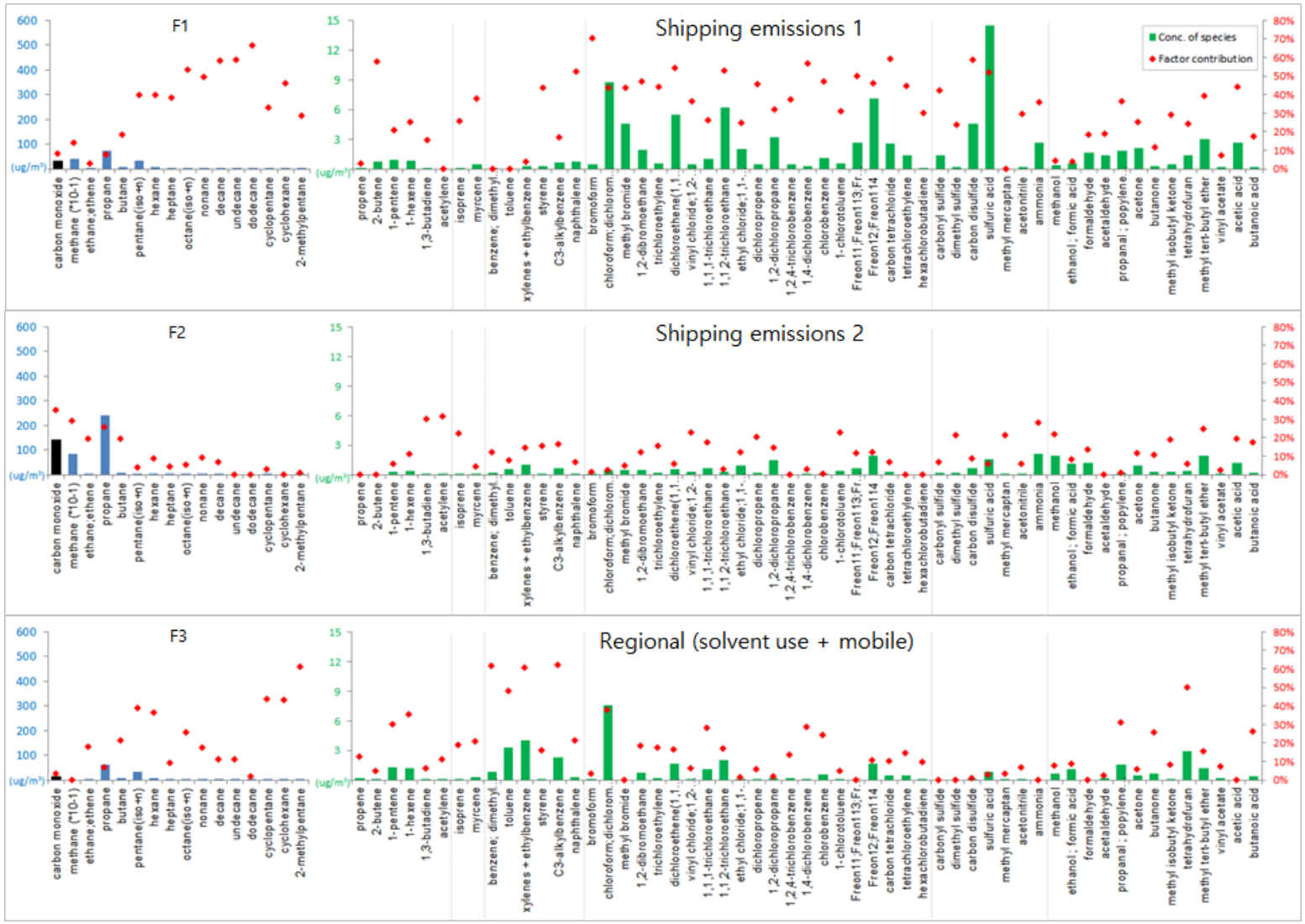
Supplementary Figure 4. Relationships between shipping emission indicators. **(a)** Correlation between V and Ni at Sin-Hang and **(b)** at Yeon-San. **(c)** Correlation between nss-SO_4^{2-} and V at Sin-Hang and **(d)** at Yeon-San.

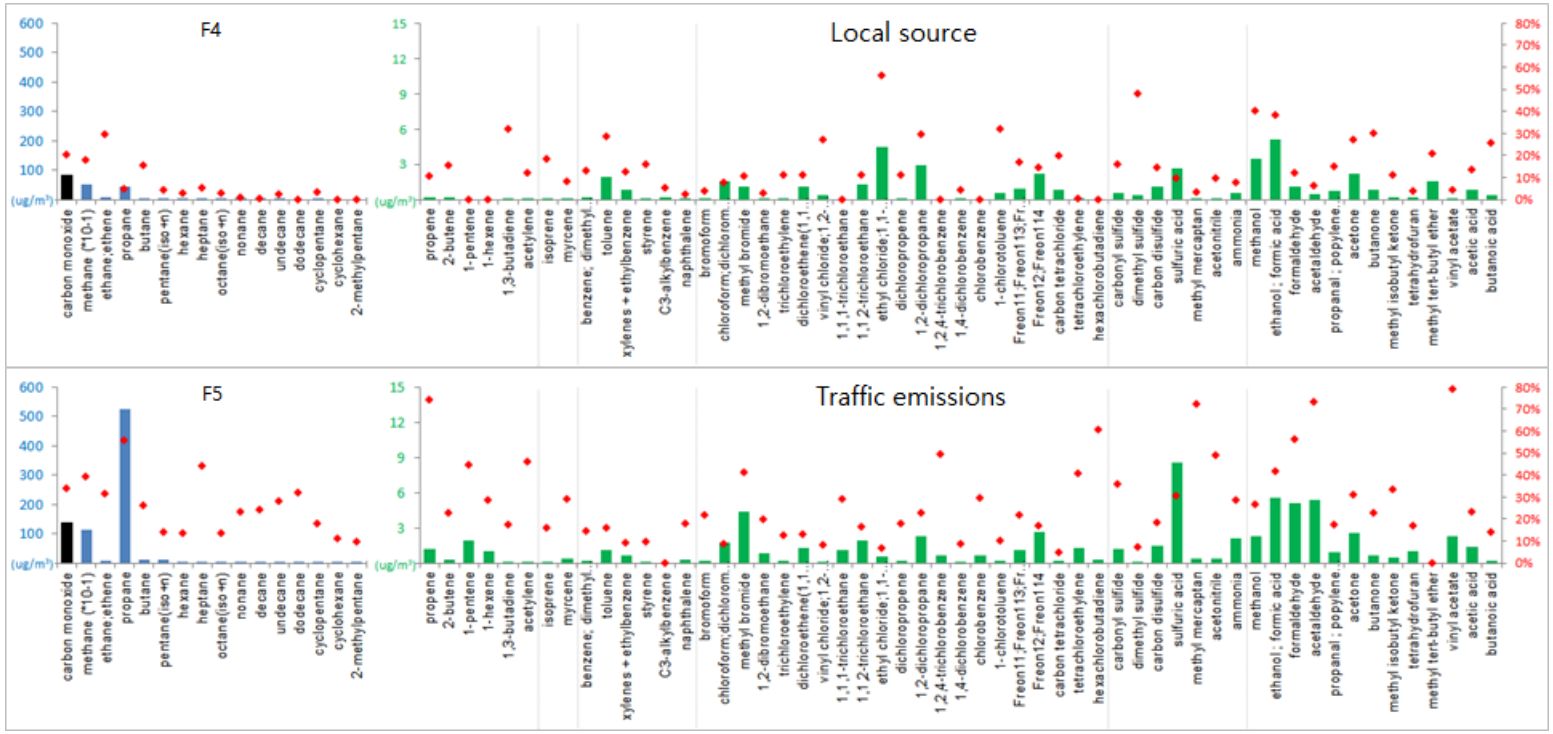


Supplementary Figure 5. Monthly variations of PMF source-specific concentrations of carbonaceous components in $PM_{2.5}$ at a port site (Sin-Hang) between 2019 and 2021. The concentration of organic carbon (OC) and elemental carbon (EC) associated with **(a)** shipping-source and **(b)** traffic-source.

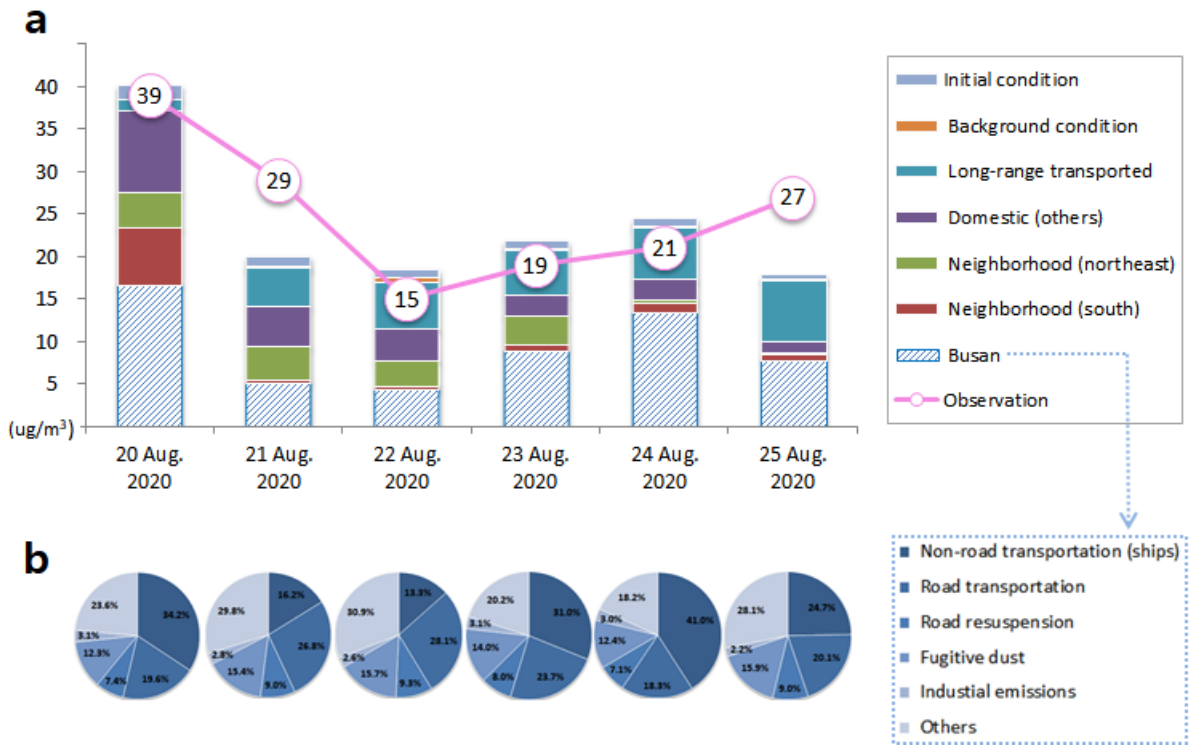


Supplementary Figure 6. Influence of air pollutions associated with shipping emissions at *Site A*. (a) Concentrations of SO_2 as a function of meteorological conditions, and a conditional probability function (CPF) plot of SO_2 being greater than the 70th percentile. **(b)** Contribution profile of net shipping emissions obtained from the cross sectional spatial analysis.


F1
Shipping emissions 1
Conc. of species
Factor contribution
F2
Shipping emissions 2
F3
Regional (solvent use + mobile)
(ug/m³)
carbon monoxide
methane (*10-1)
ethane,ethene
propane
butane
pentane(iso+n)
hexane
heptane
octane(iso+n)
nonane
decane
undecane
dodecane
cyclopentane
cyclohexane
2-methylpentane
propene
2-butene
1-pentene
1-hexene
1,3-butadiene
acetylene
isoprene
myrcene
benzene; dimethyl...
toluene
xylenes + ethylbenzene
styrene
C3-alkylbenzene
naphthalene
bromoform
chloroform;dichlorom...
methyl bromide
1,2-dibromoethane
trichloroethylene
dichloroethene(1,1...
vinyl chloride;1,2-...
1,1,1-trichloroethane
1,1,2-trichloroethane
ethyl chloride;1,1-...
dichloropropene
1,2-dichloropropane
1,2,4-trichlorobenzene
1,4-dichlorobenzene
chlorobenzene
1-chlorotoluene
Freon11;Freon113;Fr...
Freon12;Freon114
carbon tetrachloride
tetrachloroethylene
hexachlorobutadiene
carbonyl sulfide
dimethyl sulfide
carbon disulfide
sulfuric acid
methyl mercaptan
acetonitrile
ammonia
methanol
ethanol ; formic acid
formaldehyde
acetaldehyde
propanal ; popylene...
acetone
butanone
methyl isobutyl ketone
tetrahydrofuran
methyl tert-butyl ether
vinyl acetate
acetic acid
butanoic acid



Supplementary Figure 7. PMF factor profiles for trace gases measured by a mobile laboratory equipped with a selected ion flow tube mass spectrometery (SIFT-MS) at a port site (*Site A*).



Supplementary Figure 8. Estimated source-specific contributions to surface $PM_{2.5}$ mass concentrations at the urban background site (Yeon-San) based on CAMx/PSAT modelling results. (a) The bar chart represents major source regions that contribute to the daily $PM_{2.5}$ levels at Yeon-San while the line chart shows the observed $PM_{2.5}$ values at the same location. **(b)** Source-specific contributions to $PM_{2.5}$ at Yeon-San attributable to local emissions in Busan.