



Supplement of

The impacts of marine-emitted halogens on OH radicals in East Asia during summer

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Table S1. Halogen chemistry in CB6r3m mechanism. See more details in Sarwar et al. (2012) and Sarwar et al. (2019)..

Type	No.	Reaction	Rate constant / Uptake coefficient
Gaseous reactions	CL01	$\text{Cl}_2 = 2*\text{Cl}$	Photolysis
	CL02	$\text{HOCl} = \text{OH} + \text{Cl}$	Photolysis
	CL03	$\text{Cl} + \text{O}_3 = \text{ClO} + \text{O}_2$	$2.3 \times 10^{-11} e^{(-200/T)}$
	CL04	$\text{ClO} + \text{ClO} = 0.3*\text{Cl}_2 + 1.4*\text{Cl} + \text{O}_2$	1.63×10^{-14}
	CL05	$\text{ClO} + \text{NO} = \text{Cl} + \text{NO}_2$	$6.4 \times 10^{-12} e^{(290/T)}$
	CL06	$\text{ClO} + \text{HO}_2 = \text{HOCl} + \text{O}_2$	$2.2 \times 10^{-12} e^{(340/T)}$
	CL07	$\text{ClO} + \text{MEO}_2 = \text{Cl} + \text{FORM} + \text{HO}_2$	$3.2 \times 10^{-12} e^{(-110/T)}$
	CL08	$\text{OH} + \text{FMC}\text{l} = \text{Cl} + \text{CO}$	5.0×10^{-13}
	CL09	$\text{FMC}\text{l} = \text{Cl} + \text{CO} + \text{HO}_2$	Photolysis
	CL10	$\text{Cl} + \text{CH}_4 = \text{HCl} + \text{MEO}_2$	$6.6 \times 10^{-12} e^{(-1240/T)}$
	CL11	$\text{Cl} + \text{PAR} = \text{HCl} + \text{XPAR}$	
		$+ 0.06 \text{ALD}2 - 0.11 \text{PAR} + 0.76 \text{ROR} + 0.05 \text{ALDX}$	5.0×10^{-11}
	CL12	$\text{Cl} + \text{PARA} = \text{HCl} + \text{ACET} + 0.97 \text{XO}_2\text{H} + 0.03 \text{XO}_2\text{N} + \text{RO}_2$	1.4×10^{-10}
		$+ 0.06 \text{ALD}2 - 0.11 \text{PAR} + 0.76 \text{ROR} + 0.05 \text{ALDX}$	
	CL13	$\text{Cl} + \text{ETHA} = \text{HCl} + 0.991 \text{ALD}2 + 0.991 \text{XO}_2 + 0.009 \text{XO}_2\text{N} + \text{HO}_2$	$8.3 \times 10^{-11} e^{(-100/T)}$
	CL14	$\text{Cl} + \text{ETH} = \text{FMC}\text{l} + 2.0 \text{XO}_2 + \text{HO}_2 + \text{FORM}$	1.07×10^{-10}
	CL15	$\text{Cl} + \text{OLE} = \text{FMC}\text{l} + 0.33 \text{ALD}2 + 0.67 \text{ALDX} + 2.0 \text{XO}_2 + \text{HO}_2 - \text{PAR}$	2.5×10^{-10}
	CL16	$\text{Cl} + \text{IOLE} = 0.3 \text{HCl} + 0.7 \text{FMC}\text{l} + 0.45 \text{ALD}2 + 0.55 \text{ALDX} + 0.3 \text{OLE} + 0.3 \text{PAR} + 1.7 \text{XO}_2 + \text{HO}_2 + 0.3 \text{OLE} + 0.3 \text{PAR} + 1.7 \text{XO}_2 + \text{HO}_2$	3.5×10^{-10}
	CL17	$\text{Cl} + \text{ISOP} = \text{FMC}\text{l} + \text{ISPD} + 0.96 \text{XO}_2\text{H} + 0.04 \text{XO}_2\text{N} + \text{RO}_2$	4.3×10^{-10}
	CL18	$\text{Cl} + \text{FORM} = \text{HCl} + \text{HO}_2 + \text{CO}$	$8.2 \times 10^{-11} e^{(-34/T)}$
	CL19	$\text{Cl} + \text{ALD}2 = \text{HCl} + \text{C}_2\text{O}_3$	7.9×10^{-11}
	CL20	$\text{Cl} + \text{ALDX} = \text{HCl} + \text{C}_x\text{O}_3$	1.3×10^{-10}
	CL21	$\text{Cl} + \text{MEOH} = \text{HCl} + \text{HO}_2 + \text{FORM}$	5.5×10^{-11}
	CL22	$\text{Cl} + \text{ETOH} = \text{HCl} + \text{HO}_2 + \text{ALD}2$	$8.2 \times 10^{-11} e^{(45/T)}$
	CL23	$\text{OH} + \text{HCl} = \text{Cl} + \text{H}_2\text{O}$	$6.58 \times 10^{-13} (T/300)^{1.16} e^{(58/T)}$
	CL24	$\text{Cl} + \text{TOL} = \text{HCl} + 0.18*\text{CRES} + 0.65*\text{TO}_2 + 0.72*\text{RO}_2 + 0.1*\text{OPEN} + 0.1*\text{OH} + 0.07*\text{XO}_2\text{H} + 0.18*\text{HO}_2 + \text{TOLRO}_2$	6.1×10^{-11}
	CL25	$\text{Cl} + \text{XYLMN} = \text{HCl} + 0.155*\text{CRES} + 0.544*\text{XLO}_2 + 0.602*\text{RO}_2 + 0.244*\text{XOPN} + 0.244*\text{OH} + 0.058*\text{XO}_2\text{H} + 0.155*\text{HO}_2 + \text{XYLRO}_2$	1.2×10^{-10}
	CL26	$\text{Cl} + \text{NAPH} = \text{HCl} + 0.155*\text{CRES} + 0.544*\text{XLO}_2 + 0.602*\text{RO}_2 + 0.244*\text{XOPN} + 0.244*\text{OH} + 0.058*\text{XO}_2\text{H} + 0.155*\text{HO}_2 + \text{XYLRO}_2$	1.2×10^{-10}
	CL27	$\text{ClNO}_2 = \text{Cl} + \text{NO}_2$	Photolysis $k_o = 1.8 \times 10^{-31} (T/300)^{-3.4};$ $k_\infty = 1.5 \times 10^{-11} (T/300)^{-1.9};$ $F = 0.6 \text{ and } N = 1.0$
	CL28	$\text{ClO} + \text{NO}_2 = \text{ClNO}_3$	
	CL29	$\text{ClNO}_3 = \text{ClO} + \text{NO}_2$	Photolysis
	CL30	$\text{ClNO}_3 = \text{Cl} + \text{NO}_3$	Photolysis
	BR01	$\text{Br} + \text{O}_3 = \text{BrO} + \text{O}_2$	$1.6 \times 10^{-11} e^{-780/T}$
	BR02	$\text{BrO} + \text{HO}_2 = \text{HOBr} + \text{O}_2$	$4.5 \times 10^{-12} e^{460/T}$
	BR03	$\text{Br} + \text{HO}_2 = \text{HBr} + \text{O}_2$	$4.8 \times 10^{-12} e^{-310/T}$
	BR04	$\text{HBr} + \text{OH} = \text{Br} + \text{H}_2\text{O}$	$6.7 \times 10^{-12} e^{155/T}$
	BR05	$\text{BrO} + \text{BrO} = 2.0 \text{Br} + \text{O}_2$	$1.4 \times 10^{-12} e^{210/T}$
	BR06	$\text{BrO} + \text{BrO} = \text{BR}_2 + \text{O}_2$	$2.9 \times 10^{-14} e^{840/T}$
	BR07	$\text{BrO} + \text{NO} = \text{Br} + \text{NO}_2$	$8.8 \times 10^{-12} e^{260/T}$
	BR08	$\text{Br} + \text{BrNO}_3 = \text{BR}_2 + \text{NO}_3$	4.9×10^{-11}
	BR09	$\text{BR}_2 + \text{OH} = \text{HOBr} + \text{Br}$	$2.1 \times 10^{-11} e^{240/T}$
	BR10	$\text{BrO} + \text{OH} = \text{Br} + \text{HO}_2$	$1.7 \times 10^{-11} e^{250/T}$
	BR11	$\text{Br} + \text{NO}_3 = \text{BrO} + \text{NO}_2$	1.6×10^{-11}
	BR12	$\text{BrO} + \text{NO}_2 = \text{BrNO}_3$	$k_o = 5.2 \times 10^{-31} (T/300)^{-3.2};$ $k_\infty = 6.9 \times 10^{-12} (T/300)^{2.9};$ $F = 0.6 \text{ and } N = 1.0$
	BR13	$\text{Br} + \text{NO}_2 = \text{BrNO}_2$	$k_o = 4.2 \times 10^{-31} (T/300)^{-2.4};$ $k_\infty = 2.7 \times 10^{-11} (T/300)^{0.0};$ $F = 0.6 \text{ and } N = 1.0$
	BR14	$\text{BrO} + \text{ClO} = \text{Br} + \text{Cl}$	$4.7 \times 10^{-12} e^{320/T}$
	BR15	$\text{Br} + \text{FORM} = \text{HBr} + \text{HO}_2 + \text{CO}$	$1.7 \times 10^{-11} e^{-800/T}$
	BR16	$\text{Br} + \text{ALD}2 = \text{HBr} + \text{C}_2\text{O}_3$	$1.3 \times 10^{-11} e^{-360/T}$

15 **Table S1.** Continued.

Type	No.	Reaction	Rate constant / Uptake coefficient
Gaseous reactions	BR17	Br + OLE = FMBr + ALD2 + XO ₂ H – PAR + RO ₂	3.6×10^{-12}
	BR18	Br + ISOP = FMBr + ISPD + 0.96 XO ₂ H + 0.04 XO ₂ N + RO ₂	5.0×10^{-12}
	BR19	FMBr + OH = BR + CO	5.0×10^{-12}
	BR20	BrO + MEO ₂ = 0.8 HOBr + 0.2 BR + FORM	$2.7 \times 10^{-14} e^{1600/T}$
	BR21	CH ₃ Br + OH = Br + FORM	$2.9 \times 10^{-12} e^{-1230/T}$
	BR22	MB3 + OH = 3 Br + CO	$1.0 \times 10^{-12} e^{-388/T}$
	BR23	MB2 + OH = 2 Br + HO ₂ + CO	$2.0 \times 10^{-12} e^{-840/T}$
	BR24	MB2C + OH = 2 Br + Cl + CO	$9.0 \times 10^{-13} e^{-420/T}$
	BR25	MBC2 + OH = Br + 2 Cl + CO	$9.4 \times 10^{-13} e^{-510/T}$
	BR26	MBC + OH = Br + Cl + CO + HO ₂	$2.1 \times 10^{-12} e^{-880/T}$
	BR27	DMS + BrO = MEO ₂ + Br	$1.5 \times 10^{-14} e^{1000/T}$
	BR28	Br ₂ = 2 Br	Photolysis
	BR29	HOBr = OH + Br	Photolysis
	BR30	BrO = Br + O	Photolysis
	BR31	BrNO ₂ = Br + NO ₂	Photolysis
	BR32	BrNO ₃ = BrO + NO ₂	Photolysis
	BR33	BrNO ₃ = Br + NO ₃	Photolysis
	BR34	BrCl = Br + Cl	Photolysis
	BR35	FMBr = Br + CO + HO ₂	Photolysis
	BR36	MB3 = 3 Br + HO ₂ + CO	Photolysis
	BR37	MB2C = 2 Br + Cl + HO ₂ + CO	Photolysis
	BR38	MBC2 = Br + 2 Cl + HO ₂ + CO	Photolysis
	IO01	I + O ₃ = IO + O ₂	$2.1 \times 10^{-11} e^{-830/T}$
	IO02	I + HO ₂ = HI + O ₂	$1.5 \times 10^{-11} e^{-1090/T}$
	IO03	I ₂ + OH = HOI + I	2.1×10^{-10}
	IO04	HI + OH = I + H ₂ O	$1.6 \times 10^{-11} e^{440/T}$
	IO05	HOI + OH = IO + H ₂ O	5.0×10^{-12}
	IO06	IO + HO ₂ = HOI + O ₂	$1.4 \times 10^{-11} e^{540/T}$
	IO07	IO + NO = I + NO ₂	$7.15 \times 10^{-12} e^{300/T}$
	IO08	INO + INO = I ₂ + 2 NO	$8.4 \times 10^{-11} e^{-2620/T}$
	IO09	INO ₂ + INO ₂ = I ₂ + 2 NO ₂	$4.7 \times 10^{-13} e^{-1670/T}$
	IO10	I ₂ + NO ₃ = I + INO ₃	1.5×10^{-12}
	IO11	INO ₃ + I = I ₂ + NO ₃	$9.1 \times 10^{-11} e^{-146/T}$
	IO12	I + BrO = IO + Br	1.2×10^{-11}
	IO13	IO + Br = I + BrO	2.70×10^{-11}
	IO14	IO + BrO = Br + I + O ₂	$1.5 \times 10^{-11} e^{510/T}$
	IO15	IO + ClO = I + Cl	$4.7 \times 10^{-12} e^{280/T}$
	IO16	OIO + OIO = I ₂ O ₄	$1.5 \times 10^{-10} e^{0/T}$
	IO17	OIO + NO = IO + NO ₂	$1.1 \times 10^{-12} e^{542/T}$
	IO18	IO + IO = 0.4OIO + 0.4I + 0.6I ₂ O ₂	$5.4 \times 10^{-11} e^{180/T}$
	IO19	IO + OIO = I ₂ O ₃	1.5×10^{-10}
	IO20	I ₂ O ₂ = OIO + I	$2.5 \times 10^{14} e^{-9770/T}$
	IO21	I ₂ O ₄ = 2OIO	$3.8 \times 10^{-2} e^{0/T}$
	IO22	INO ₂ = I + NO ₂	$9.94 \times 10^{17} e^{-11859/T}$
	IO23	INO ₃ = IO + NO ₂	$2.1 \times 10^{15} e^{-13670/T}$
	IO24	I + NO = INO	$k_o = 1.8 \times 10^{-32} (T/300)^{-1.0};$ $k_{\infty} = 1.7 \times 10^{-11} (T/300)^{0.0};$ F = 0.6 and N = 1.0
	IO25	I + NO ₂ = INO ₂	$k_o = 3.0 \times 10^{-31} (T/300)^{-1.0};$ $k_{\infty} = 6.6 \times 10^{-11} (T/300)^{0.0};$ F = 0.6 and N = 1.0
	IO26	IO + NO ₂ = INO ₃	$k_o = 7.7 \times 10^{-31} (T/300)^{-5.0};$ $k_{\infty} = 1.6 \times 10^{-11} (T/300)^{0.4};$ F = 0.4 and N = 1.0
	IO27	CH ₃ I + OH = I + HO ₂ + H ₂ O	$4.3 \times 10^{-12} e^{1120/T}$
	IO28	IO + DMS = I + MEO ₂	$3.2 \times 10^{-13} e^{-925/T}$
	IO29	CH ₃ I = I + MEO ₂	Photolysis

Table S1. Continued.

Type	No.	Reaction	Rate constant / Uptake coefficient
Gaseous reactions	IO30	$\text{CH}_2\text{I}_2 = 2.0 \text{ I}$	Photolysis
	IO31	$\text{CH}_2\text{IBr} = \text{I} + \text{Br}$	Photolysis
	IO32	$\text{CH}_2\text{ICl} = \text{I} + \text{Cl}$	Photolysis
	IO33	$\text{I}_2 = 2.0 \text{ I}$	Photolysis
	IO34	$\text{IO} = \text{I} + \text{O}$	Photolysis
	IO35	$\text{OIO} = \text{I} + \text{O}_2$	Photolysis
	IO36	$\text{INO} = \text{I} + \text{NO}$	Photolysis
	IO37	$\text{INO}_2 = \text{I} + \text{NO}_2$	Photolysis
	IO38	$\text{INO}_3 = \text{I} + \text{NO}_3$	Photolysis
	IO39	$\text{HOI} = \text{I} + \text{OH}$	Photolysis
	IO40	$\text{ICl} = \text{I} + \text{Cl}$	Photolysis
	IO41	$\text{I}_2\text{O}_2 = \text{I} + \text{OIO}$	Photolysis
	IO42	$\text{I}_2\text{O}_3 = \text{IO} + \text{OIO}$	Photolysis
	IO43	$\text{I}_2\text{O}_4 = \text{OIO} + \text{OIO}$	Photolysis
	IO44	$\text{IBr} = \text{I} + \text{Br}$	Photolysis
Heterogeneous reactions on aerosols	HET_CLNO3_WA	$\text{CINO}_3 \rightarrow \text{HOCl} + \text{HNO}_3$	0.024
	HET_N2O5	$\text{Cl}^- + \text{N}_2\text{O}_5 \rightarrow \text{CINO}_2$	Parameterization from $1.3 \times 10^{-8} e^{4290/T}$
	HET_HBR_BR	$\text{HBr} \rightarrow \text{Br}^-$	
	HET_BRNO3_WA	$\text{BrNO}_3 (+ \text{H}_2\text{O}) \rightarrow \text{HOBr} + \text{HNO}_3$	0.03
	HET_HOBr_CL	$\text{HOBr} + \text{Cl}^- \rightarrow \text{BrCl}$	0.08
	HET_HOBr_BR	$\text{HOBr} + \text{Br}^- \rightarrow \text{Br}_2$	0.02
	HET_I2O2	$\text{I}_2\text{O}_2 \rightarrow$	0.02
	HET_I2O3	$\text{I}_2\text{O}_3 \rightarrow$	0.02
	HET_I2O4	$\text{I}_2\text{O}_4 \rightarrow$	0.02
	HET_INO3_CL	$\text{INO}_3 + \text{Cl}^- \rightarrow \text{ICl} + \text{HNO}_3$	0.005
	HET_INO3_BR	$\text{INO}_3 + \text{Br}^- \rightarrow \text{IBr} + \text{HNO}_3$	0.005
	HET_INO2_CL	$\text{INO}_2 + \text{Cl}^- \rightarrow \text{ICl} + \text{HONO}$	0.01
	HET_INO2_BR	$\text{INO}_2 + \text{Br}^- \rightarrow \text{IBr} + \text{HONO}$	0.01
	HET_HOI_CL	$\text{HOI} + \text{Cl}^- \rightarrow \text{ICl}$	0.005
	HET_HOI_BR	$\text{HOI} + \text{Br}^- \rightarrow \text{IBr}$	0.005
Heterogeneous reactions on ice crystals	HET_HOBr_HBR	$\text{HOBr} + \text{HBr} \rightarrow \text{Br}_2 + \text{H}_2\text{O}$	0.12
	HET_HOBr_HCL	$\text{HOBr} + \text{HCl} \rightarrow \text{BrCl} + \text{H}_2\text{O}$	0.30
	HET_HOCl_HBR	$\text{HOCl} + \text{HBr} \rightarrow \text{BrCl} + \text{H}_2\text{O}$	0.20

Note. First order rate constants are in units of s^{-1} , second order rate constants are in units of $\text{cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$. Temperatures (T) are in Kelvin. k_o , k_∞ , F, and N are for rate constants described by the falloff expression of the form $k = \{k_o[M]/(1 + k_o[M]/k_\infty)\} F^Z$, where $Z = \{(1/N) + \log_{10}[k_o [M]/k_\infty]^2\}^{-1}$, where [M] is the total pressure in molecules cm^{-3} .

20 ACET = acetone, ALD2 = acetaldehyde, ALDX = propionaldehyde and higher aldehydes, C2O3 = acetylperoxy radical, CRES = cresol and higher molecular weight phenols, CXO3 = higher acylperoxy radicals, ETH = ethene, ETHA = ethane, ETOH = ethanol, FMBr = formyl bromide, FMCl = formyl chloride, FORM = formaldehyde, IOLE = internal olefinic carbon bond, ISOP = isoprene, ISPD = isoprene product, MEO2 = methylperoxy radical, MECH = methanol, NAPH = naphthalene, OLE = terminal olefinic carbon bond, RO2 = peroxy radical ROR = secondary alkoxy radical, PAR = paraffin carbon bond, TOL = toluene, XLO2 = peroxy radical from XYLMN+OH reaction, XO2 = NO-to-NO2 operator, XO2N = NO-to-nitrate operator, XO2H = NO-to-NO2 with HO2 production, from alkoxy radicals, XOPN = product of aromatic ring-opening reaction, XPAR = organic nitrate production from PAR, XYLMN = xylene and other polyalkyl aromatics except naphthalene, XYLRO2 = counter species for aerosol from XYLMN+OH.

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Table S2. Global annual fluxes of very short-lived halocarbons reported in previous studies and scale factors of halocarbon emissions used in this study.

Species	Annual flux (Gg/yr)			Scale factor	
	Ordóñez et al. (2012)	WMO low	WMO high	low	high
CHBr₃	533	126	865	0.24	1.62
CH₂Br₂	67.3	62	109	0.92	1.62
CH₃I	303	176	615	0.58	2.03
CH₂BrCl	10.0	6.48	9.72	0.65	1
CHBr₂Cl	19.7	19.6	56.1	1	2.85
CHBrCl₂	22.6	16.4	22.6	0.73	1
CH₂ICl	234	--	--	0.58	2.03
CH₂IBr	87.3	--	--	0.58	2.03
CH₂I₂	116	--	--	0.58	2.03

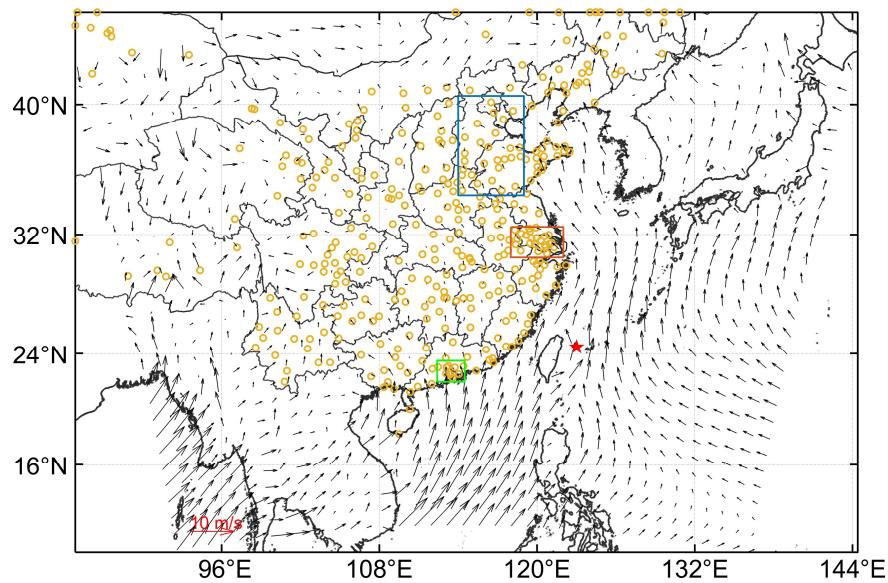
Table S3. Case design in the supplementary information.

Simulation case	Species or reactions ^a	Emission rate and ref
BASE_PHIL5	As BASE but increasing NOx and VOCs emissions in the Philippines by a factor of 5, corresponding to InorgI_chemPHIL5 below.	0
All_highI	As All_high	Replacing HOI emission with I ₂ in equivalent amount of I atom
InorgI_chemIhalf	As InorgI_chem	Decreasing I ₂ and HOI emission rates by 50%
InorgI_chemIfixed	As InorgI_chem	Fixing I ₂ and HOI emission rates over the domain and time
InorgI_chemPHIL5	As InorgI_chem but increasing NOx and VOCs emissions in the Philippines by a factor of 5.	As in InorgI_chem

Table S4. Cross reference between cases and figures.

Figures	Cases
Fig. 1	All_high
Fig. 2	All_high - BASE All_low - BASE
Fig. 3	All_high - BASE
Fig. 4	(a)-(d) SSA: SSA - BASE (e)-(h) InorgI: InorgI - BASE (i)-(l) HaloC: HaloC - BASE (m)-(p) interaction: All_high - SSA - InorgI - HaloC + 2BASE
	(a)-(d) SSA_phy: SSA_phy - BASE_phy
Fig. 5, decomposition of Figs. 4a-d	(e)-(h) SSA_chemCl: (SSA_Cl - BASE) - (SSA_phy - BASE_phy) (i)-(l) SSA_chemBr: SSA - SSA_Cl
Fig. 6	Corresponding to values in Figs. 5 and 7 divided by BASE
Fig. 7, decomposition of Figs. 4e-h	(a)-(d) O3depo: InorgI - InorgI_chem (e)-(h) InorgI_chem: InorgI_chem - BASE
Fig. 8	All_high
Fig. 9	InorgI_chem
Fig. S1	All_high, All_low, BASE
Fig. S2	All_high, BASE
Fig. S3	All_low - BASE
Fig. S4	(a) All_high - BASE (b) SSA - SSA_Cl
Fig. S5	--
Fig. S6	SSA_phy
Fig. S7	SSA_Cl, BASE
	(a)-(b) All_high - BASE
Fig. S8	(c) All_high - All_highI (d) (InorgI_chem - BASE) + (All_highI - All_high)
Fig. S9	(a) InorgI_chemIhalf - BASE (b)-(c) InorgI_chemIhalf - InorgI_chem (d) InorgI_chemIfixed - BASE (e)-(f) InorgI_chemIfixed - InorgI_chem
Fig. S10	(a) BASE_PHIL5 - BASE (b) InorgI_chemPHIL5 - BASE_PHIL5 (c)-(f) (InorgI_chemPHIL5 - BASE_PHIL5) - (InorgI_chem - BASE)

(a)



(b)

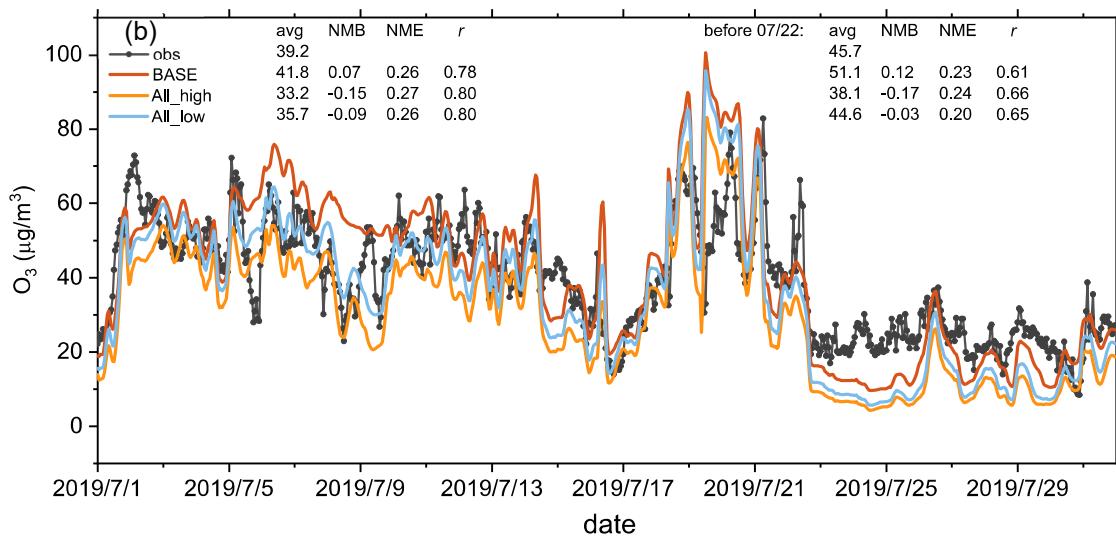
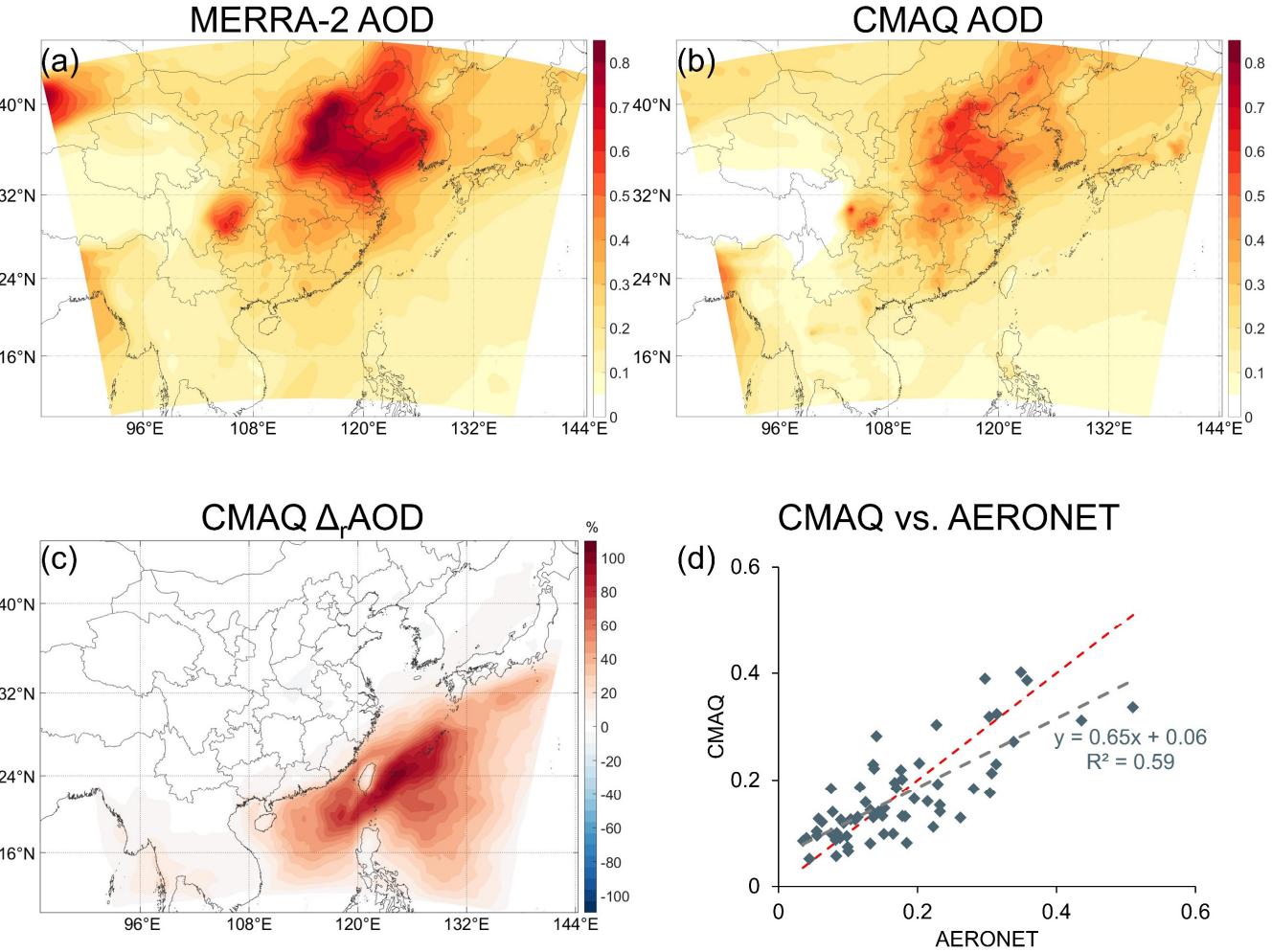


Figure S1. (a) Positions of the observation stations for model evaluation. The yellow circles are stations. The blue, orange, and green boxes indicate the North China Plain, the Yangtze River Delta, and the Pearl River Delta areas, respectively. The red star East to the Taiwan island is the location of the Yonaguni station. The arrows indicate the monthly averaged wind field. (b) Simulated and measured O₃ concentration at Yonaguni. The variation of O₃ in July 2019 here is reasonably captured by all the three simulations. Adding the halogen emissions (especially with low emission rates) can noticeably lower the bias for the high ozone concentration and improve the correlation between observation and simulations -relationship (i.e., days before July 22).



45 **Figure S2.** Spatial distribution of (a) MERRA-2 (downloaded from https://disc.gsfc.nasa.gov/datasets/M2TMNXAER_5.12/.4/summary?keywords=MERRA-2%20AOD) and (b) modelled (in All_high case) monthly average of AOD, and (c) relative change of AOD due to the emission of SSA ((3high-BASE)/BASE). (d) The comparison of daily modelled AOD in All_high case and AERONET AOD (550nm calculated from 500nm using Angstrom exponent) at four available island stations (north to south: Fukue, Okinawa_Hedo, Dongsha, and Manila, https://aeronet.gsfc.nasa.gov/cgi-bin/webtool_aod_v3?stage=2®ion=Asia). All AOD data are at 550 nm.

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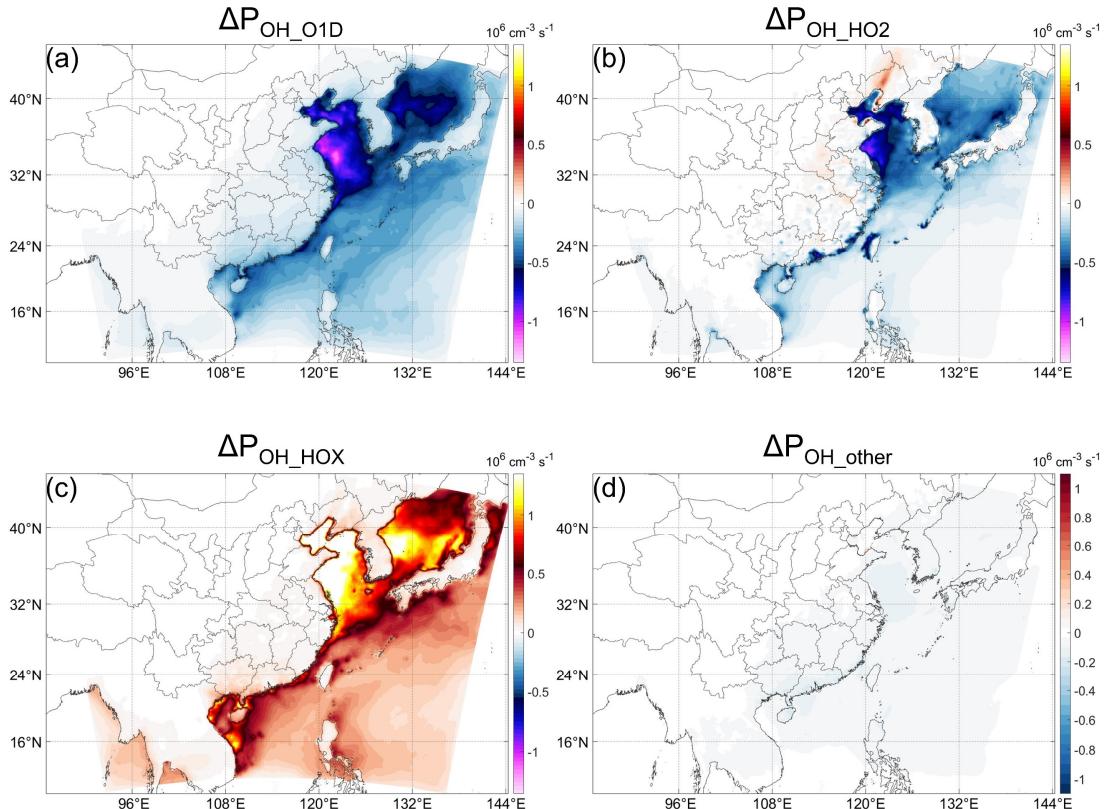
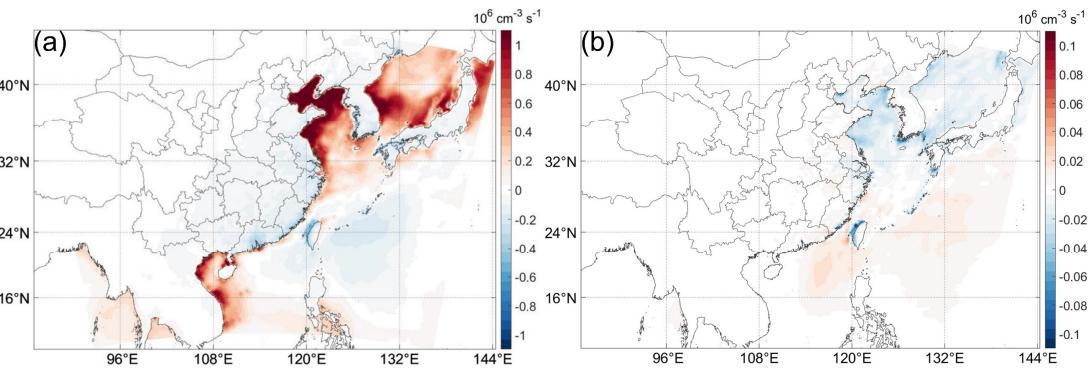


Figure S3. The same as Fig. 3 but for All_low case. The red and blue color scales are the same in (a)–(d).



55 **Figure S4.** Change of P_{OH} (a) through pathway 1 (ΔP_{OH_O1D}) and 3 (ΔP_{OH_HOX}) in All_high case compared to BASE case, and (b) through pathway 2 (ΔP_{OH_HO2}) and 3 (ΔP_{OH_HOX}) in “SSA_chemBr” case compared to BASE case.

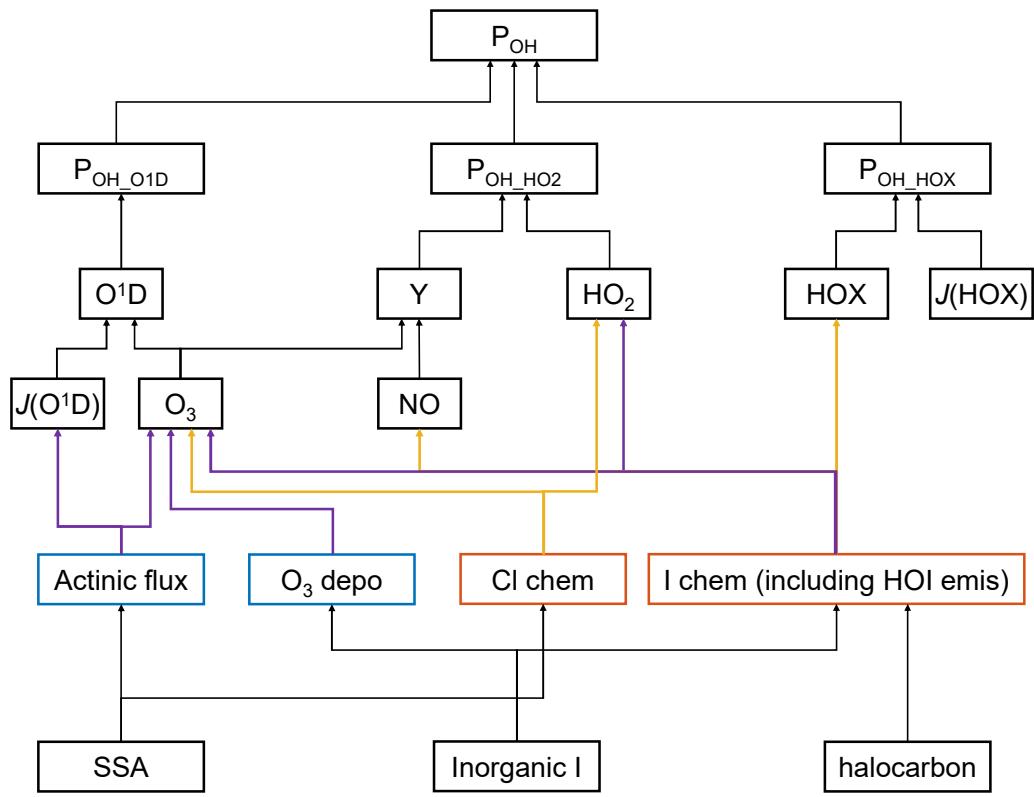


Figure S5. Schematic representation of the three main pathways that halogen species influence OH production (P_{OH_O1D} , P_{OH_HO2} , and P_{OH_HOX}) and their main controlling factors. The boxes in blue and orange represent independent and interrelated (chemical) factors, respectively. The purple and yellow arrows represent decrease and increase of the target species or parameters, respectively.

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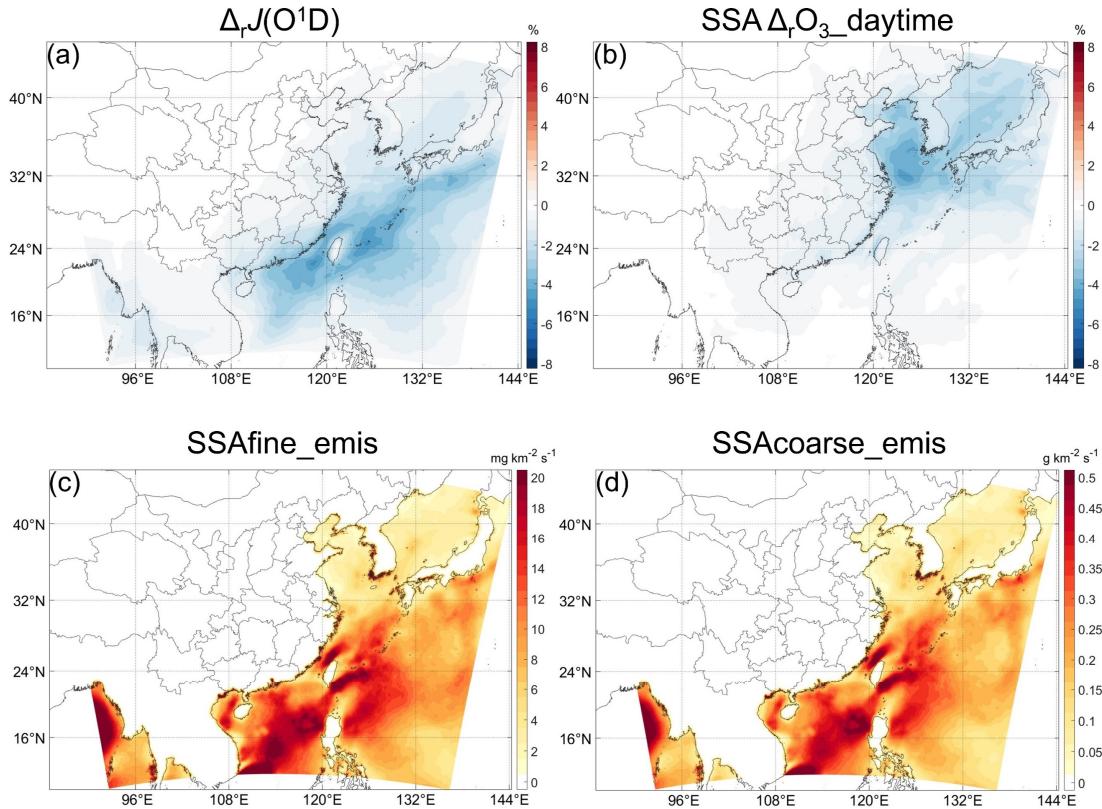


Figure S6. Relative change of (a) $J(O^1D)$ and (b) daytime O₃ caused by the extinction effect of SSA, approximated by $\Delta_r O_3_{\text{daytime}}$ in SSA_Cl without the uptake of N₂O₅ and yield of ClNO₂. (c) Monthly averaged emission rate of fine ($\leq 2.5 \mu\text{m}$) SSA. (d) Monthly averaged emission rate of coarse ($> 2.5 \mu\text{m}$) SSA.

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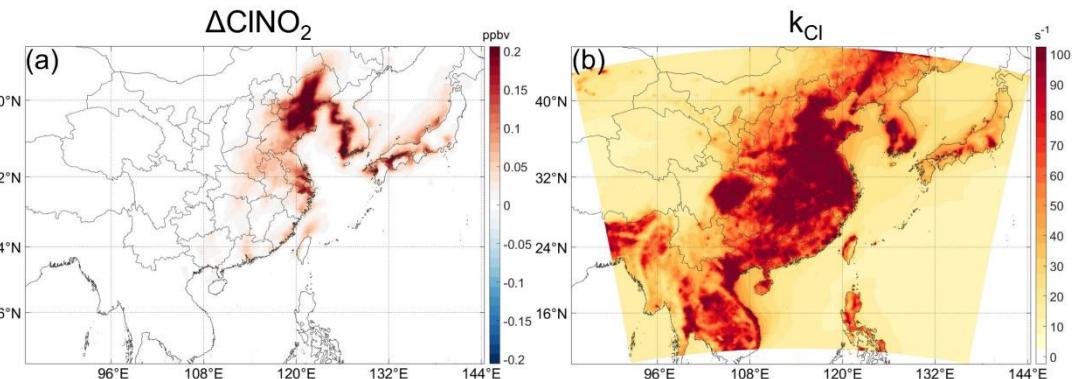
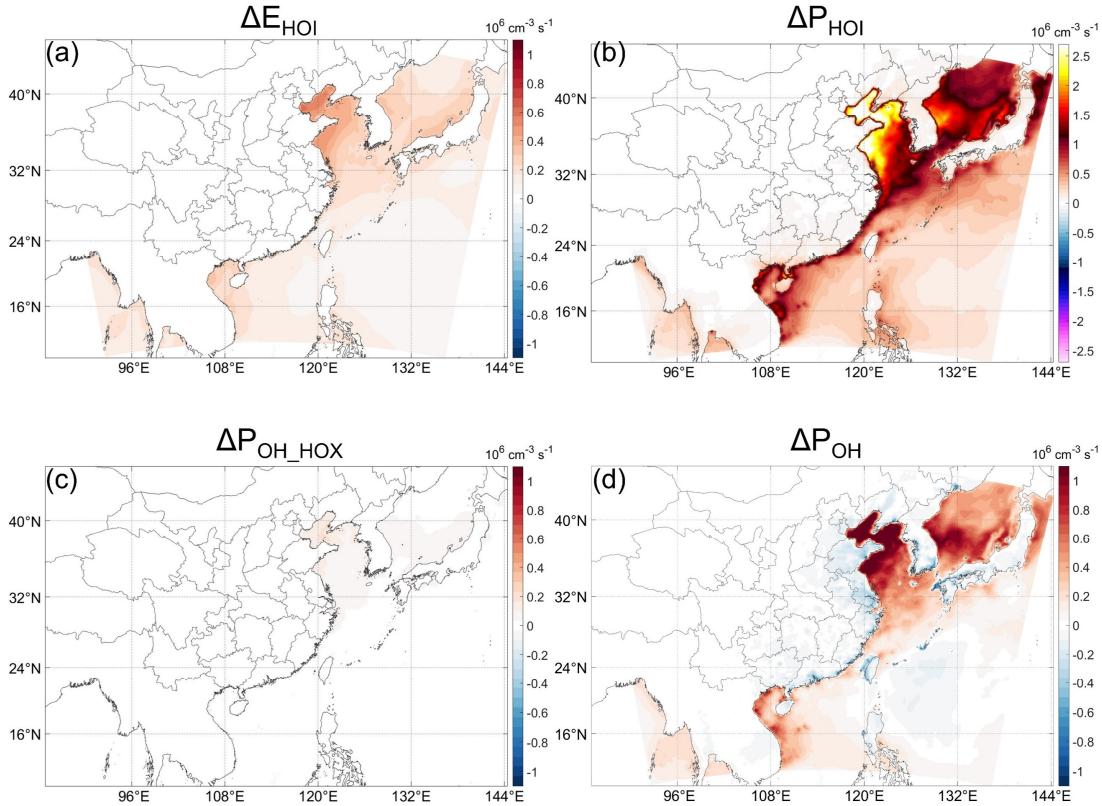


Figure S7. (a) ClNO₂ change in SSA_Cl case compared to BASE case. (b) k_{Cl} in BASE case (Δk_{Cl} is very small).



70 **Figure S8.** Change of HOI (a) emission rate and (b) production rate from $\text{HO}_2\text{+IO}$ in All_high case compared to BASE case. (c) Change of POH_{HOX} by replacing HOI emission with an equivalent amount of I_2 in All_high case, approximating the contribution of direct HOI emission to POH_{HOX} . The very small values in (c) indicates that what is more important in the iodine chemistry is the amount of reactive inorganic iodine, no matter what the form is (HOI or I_2). (d) ΔPOH induced by iodine chemistry with HOI emission replaced by I_2 in InorgI_chem case (compare with Fig. 7e), approximated using All_high data.

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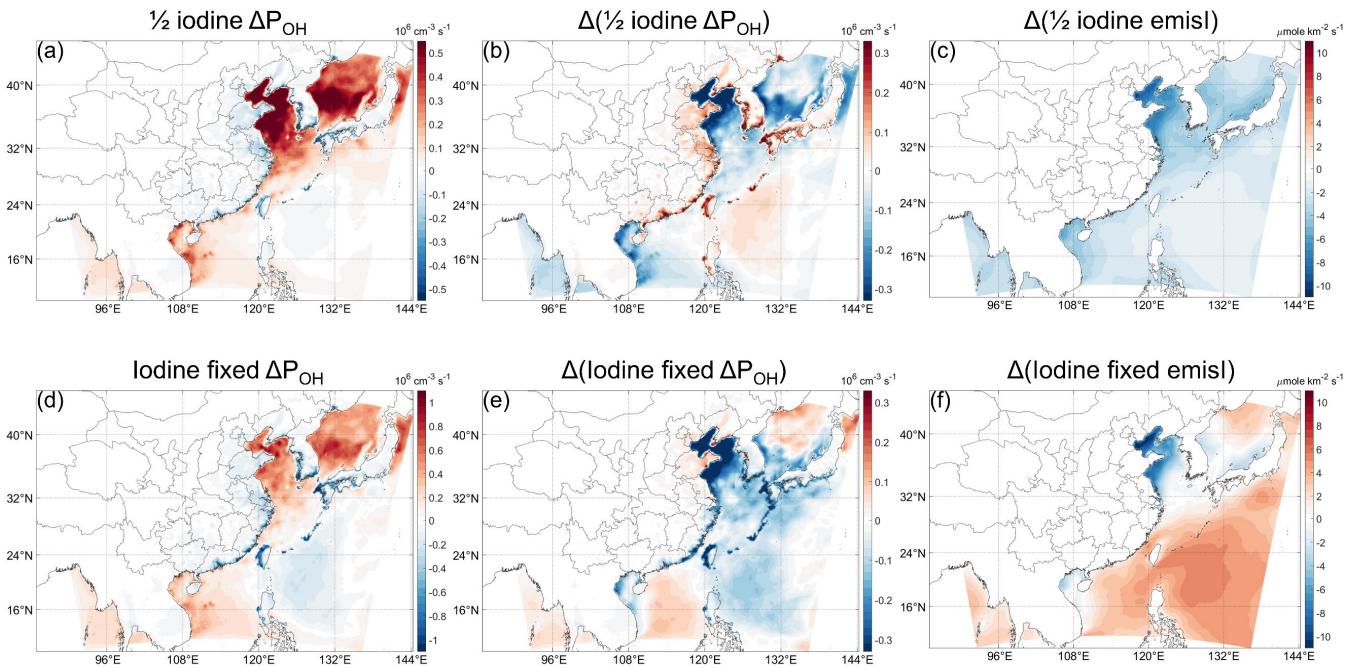
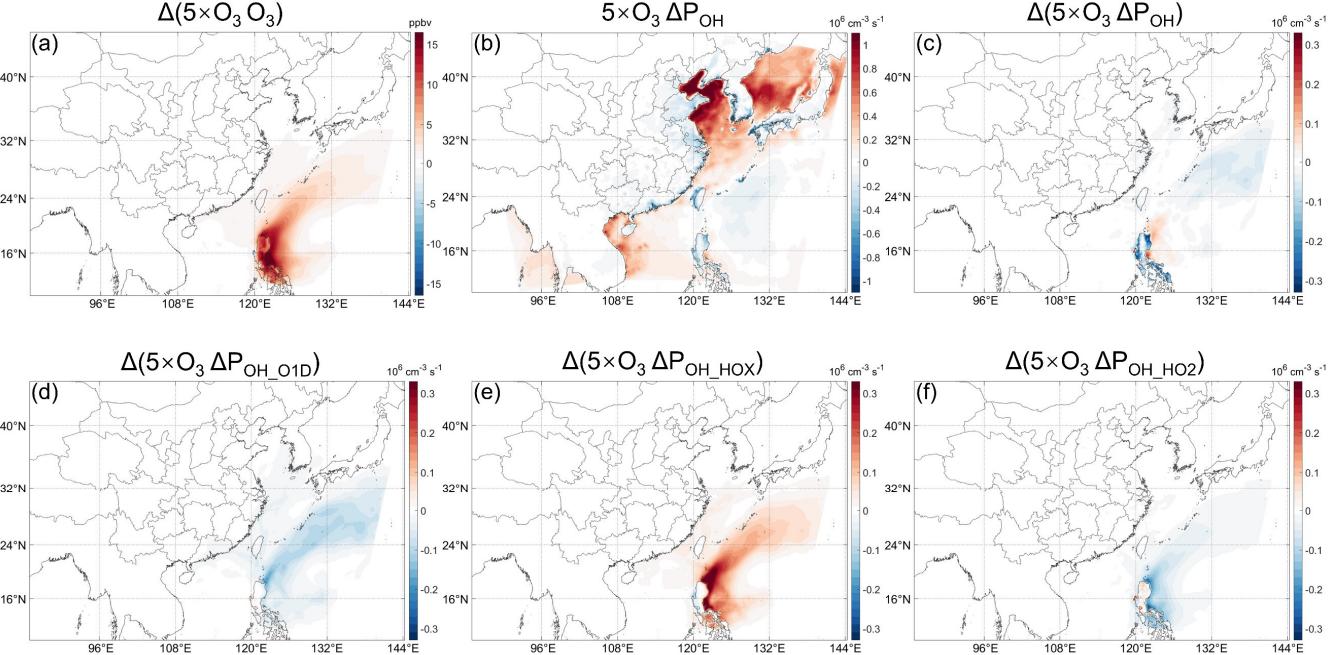


Figure S9. POH change and emission rate in a case reducing inorganic iodine emissions by 50% relative to InorgI_chem case

80 (a–c) and in a case with fixed emission rate of iodine ($6.86 = 5000/27^2 \mu\text{mole km}^{-2} \text{ s}^{-1}$ for HOI, and $1/20$ of HOI for I₂) (d–f). (a) The same as Fig. 4e but with reduced the HOI/I₂ emission rates. (b) The difference between (a) and Fig. 4e. (c) The reduction of emission rate of iodine (HOI+2×I₂). Note the different scales. (d)–(f) are the same as (a)–(c) but with fixed emission rate of iodine.



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Figure S10. (a) O₃ increase in BASE case when increasing NOx and VOCs emissions by a factor of five in the Philippines, “Δ” before the parenthesis indicated BASE with increased O₃ minus BASE. (b) P_{OH} change induced by iodine chemistry in the case with increased NOx and VOCs emissions (both BASE and InorgI_chem cases increase the emissions), the same as Fig. 4e except the NOx and VOCs emissions. (c) The difference between (a) and Fig. 4e, characterizing the further change of P_{OH} induced by iodine chemistry when increasing O₃ concentration. (d)–(f) The three main pathways decomposing (c). Note the different scales. (c) illustrates a striking characteristic of the further ΔP_{OH} over the ocean: positive close to the source, negative far away; in the continent, the negative further ΔP_{OH} is caused by the other pathways than the main three (d)–(f), which is not discussed further here.

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105