Table S1. Field measurements of $ClNO_2$ and N_2O_5 from literatures

Site	Longitude	Latitude	Period	Species	Reference
Taizhou	120.00° E	32.55° N	May 23 – June 15, 2018	N_2O_5	Li et al. (2020)
Changping	116.23° E	40.22° N	May 13 – June 23, 2016	$ClNO_2$	Le Breton et al. (2018)
Beijing	116.36° E	39.97° N	June 11 – 16, 2017	$ClNO_2$	Zhou et al. (2018)
Wangdu	115.20° E	38.66° N	June 20 − July 9, 2014	$ClNO_2$ and N_2O_5	Tham et al. (2016)
Mount Tai	117.10° E	36.25° N	July 24 − August 27, 2014	$ClNO_2$ and N_2O_5	Wang et al. (2017)
Mount TaiMoShan	114.13° E	22.41° N	November 15 – December 6, 2013	$ClNO_2$	Wang et al. (2016)

Table S2. Normalized mean bias (NMB) and correlation coefficients (*r*) between observed and simulated aerosol components at different observation sites

Site	Case	SO_4^{2-}		NO_3^-		NH_4^+		Cl^-		OM	
		NMB	*r*	NMB	*r*	NMB	*r*	NMB	*r*	NMB	*r*
Dongying	Base	-33%	0.89	-41%	0.87	-40%	0.83	-36%	0.68	49%	0.77
	McDuffie	-40%	0.84	-40%	0.88	-42%	0.88	-35%	0.68	49%	0.77
	NoEm	-40%	0.84	-40%	0.86	-46%	0.85	-89%	-0.05	49%	0.77
Guangzhou	Base	-8.2%	0.19	129%	0.18	65%	0.25	39%	0.71	20%	0.28
	McDuffie	-8.4%	0.18	143%	0.16	71%	0.26	56%	0.71	21%	0.27
	NoEm	-7.0%	0.16	141%	0.16	64%	0.23	-79%	0.61	22%	0.26
Gucheng	Base	-43%	0.34	-11%	0.72	-27%	0.67	-4.7%	0.40	-11%	0.60
	McDuffie	-44%	0.33	-12%	0.73	-27%	0.67	-4.0%	0.39	-12%	0.60
	NoEm	-43%	0.33	-13%	0.73	-41%	0.66	-96%	0.10	-12%	0.60

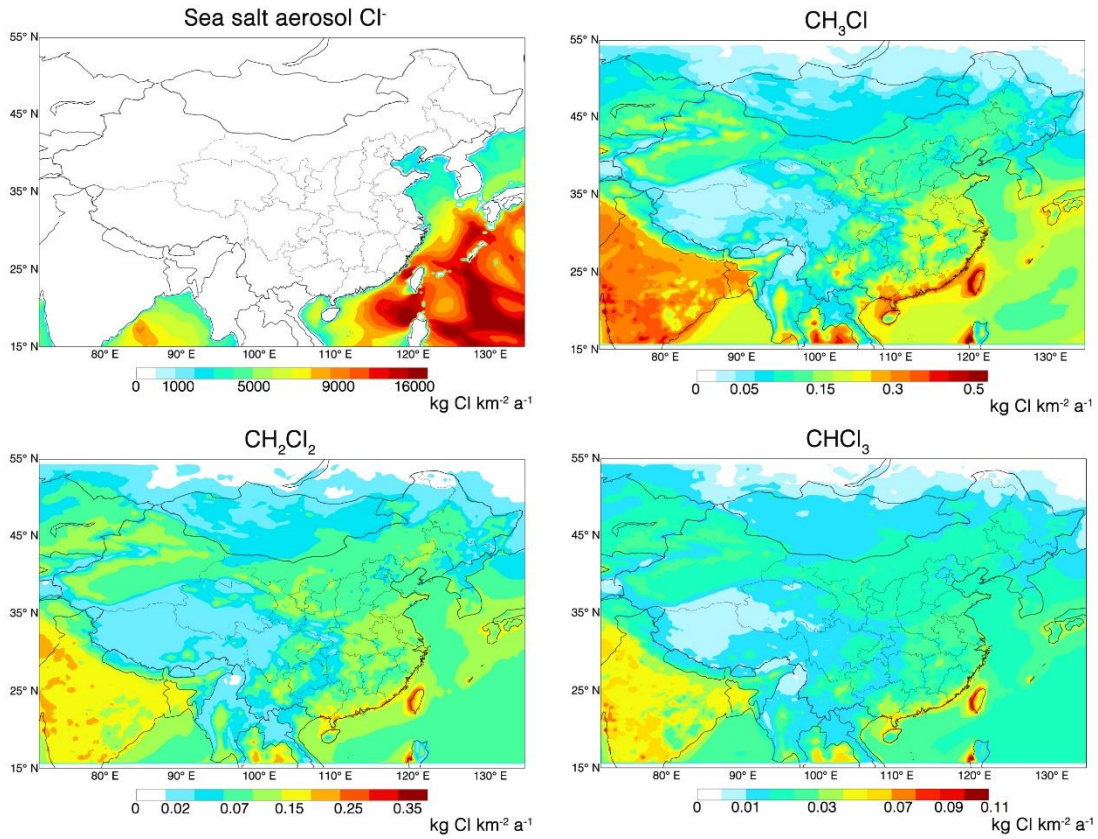

Figure S1. Spatial distribution of annual chlorine emissions released from sea salt aerosol, CH_3Cl, CH_2Cl_2 and $CHCl_3$.

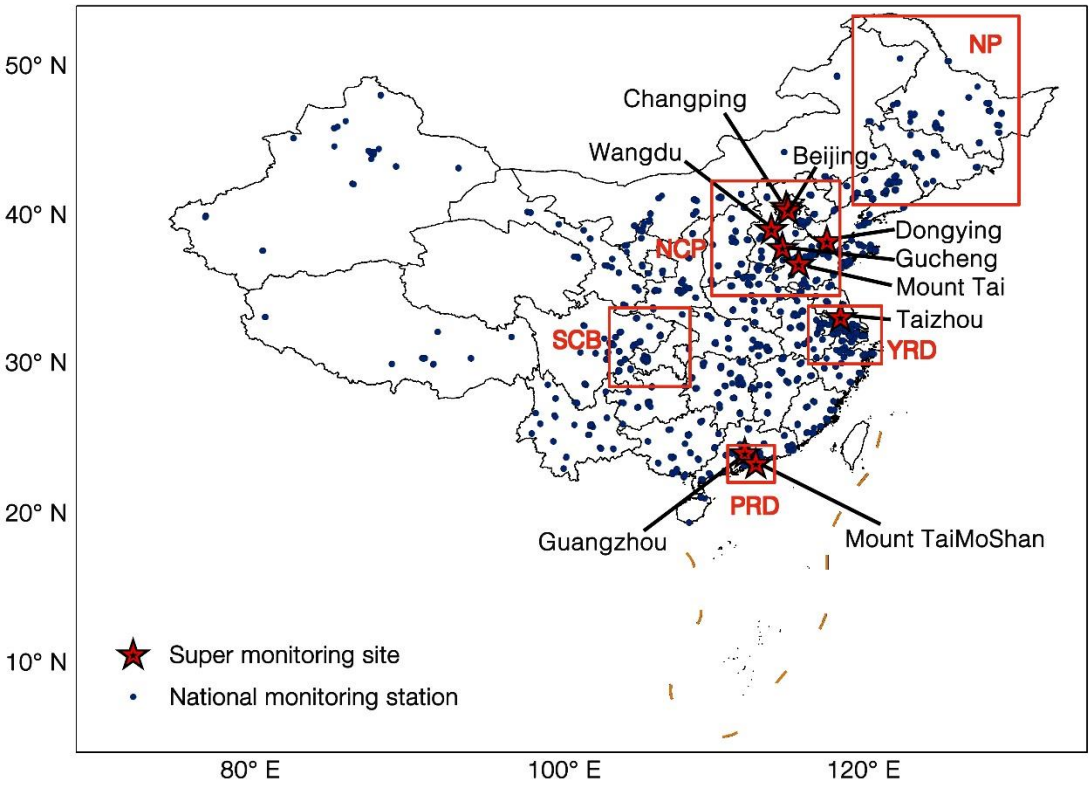

Figure S2. Spatial distribution of observation sites.

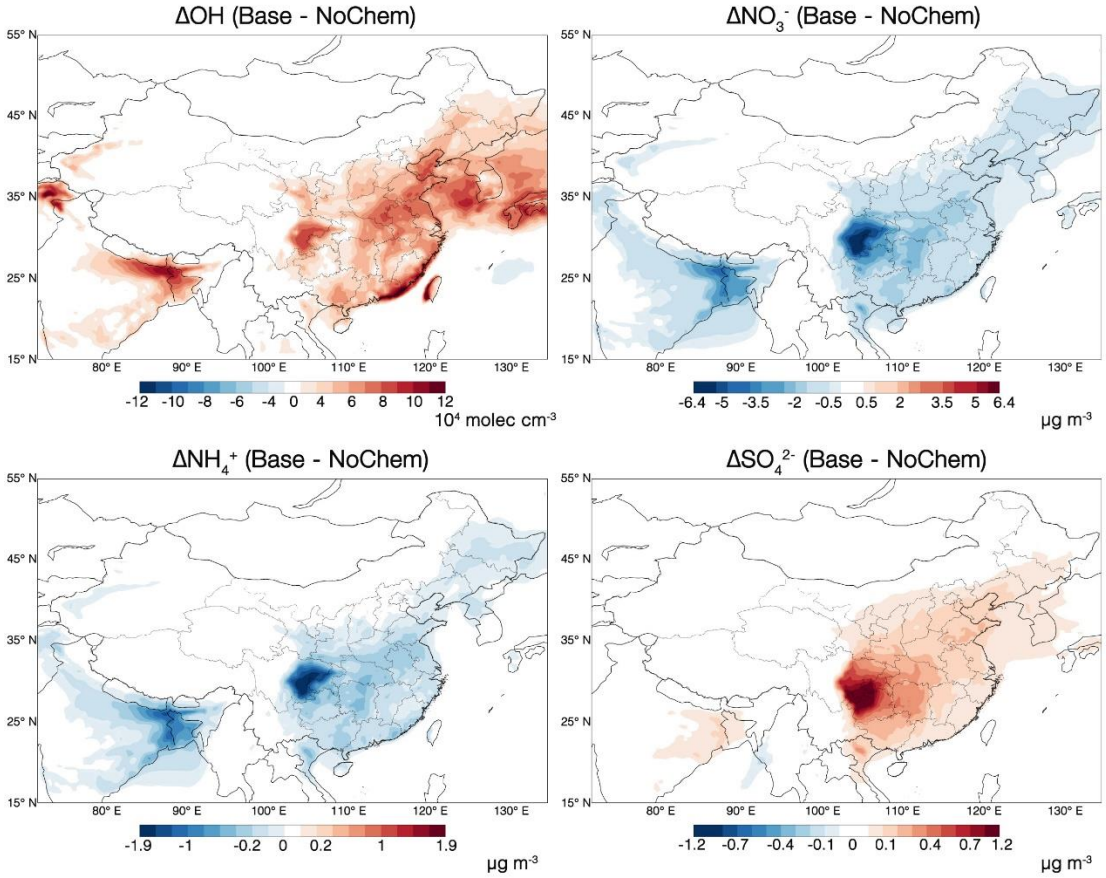

Figure S3. Effects of chlorine chemistry on annual mean surface concentrations of OH, NO_3^-, NH_4^+ and SO_4^{2-} in China.

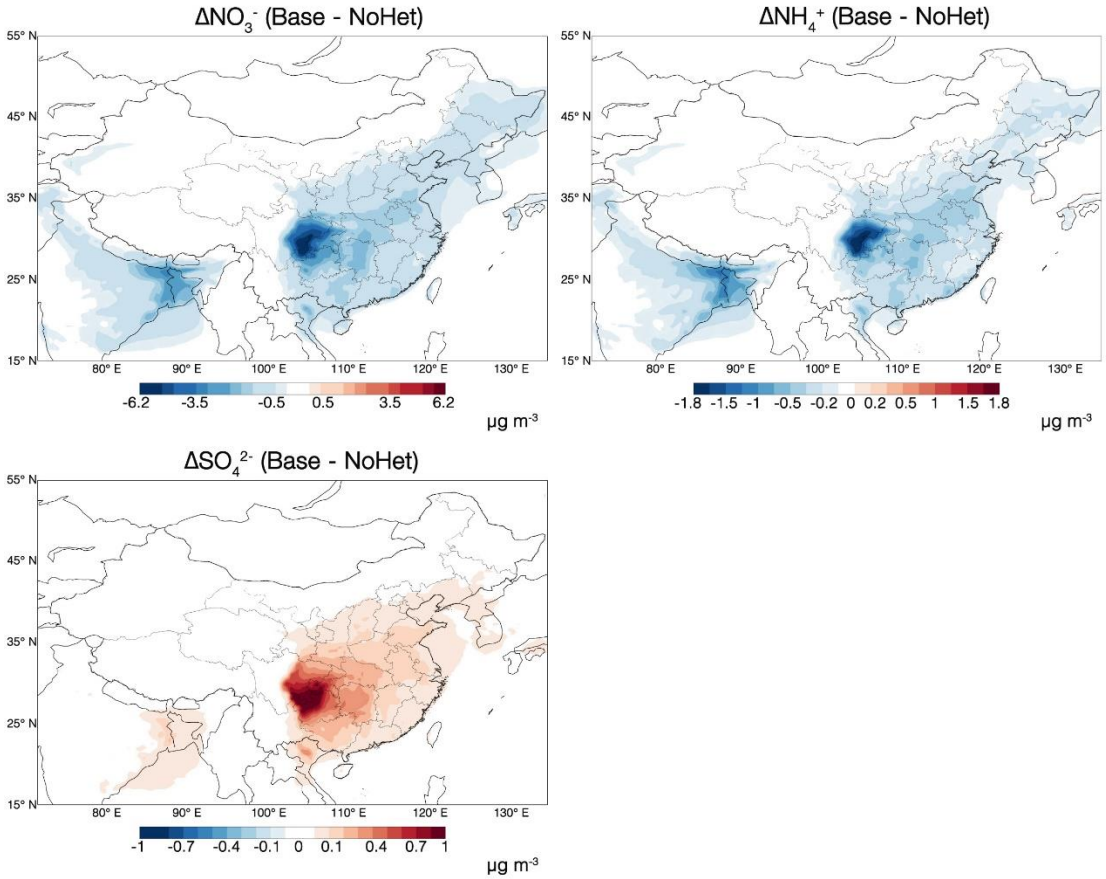

Figure S4. Effects of N_2O_5-$ClNO_2$ chemistry on annual mean surface concentrations of NO_3^-, NH_4^+ and SO_4^{2-} in China.

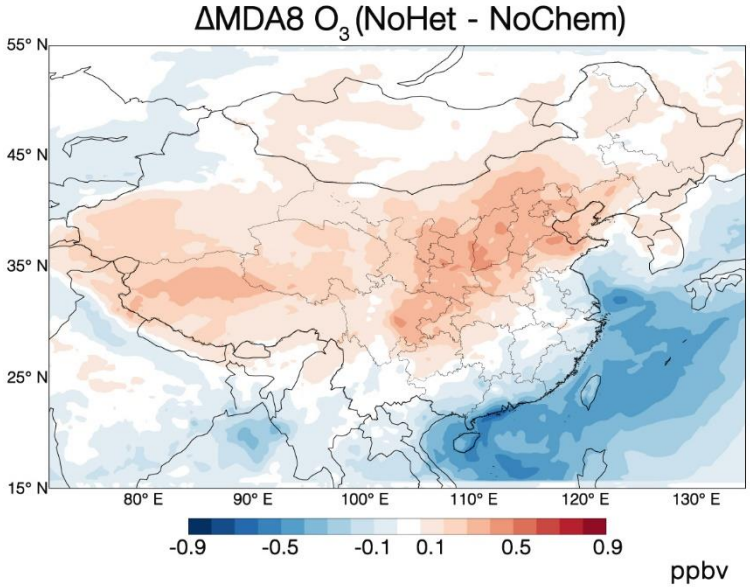

Figure S5. Impacts of chlorine chemistry other than the N_2O_5-$ClNO_2$ chemistry on annual surface mean surface concentrations of MDA8 O_3 in China.

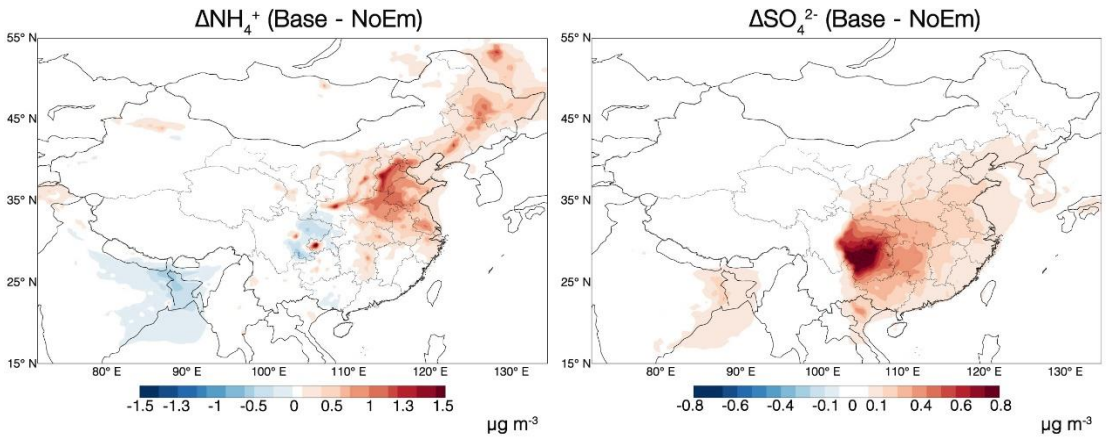

Figure S6. Effects of anthropogenic and biomass burning chlorine emissions on annual mean surface concentrations of NH_4^+ and SO_4^{2-} in China.

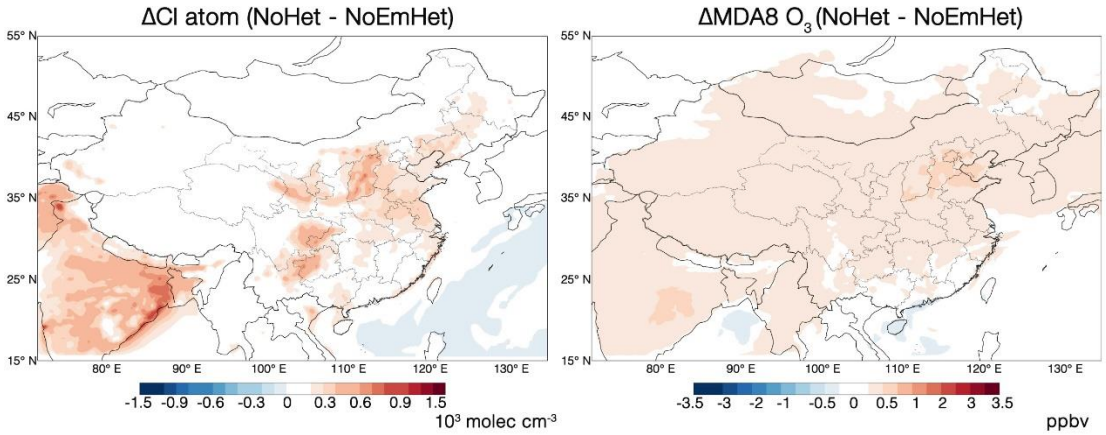

Figure S7. Effects of anthropogenic and biomass burning chlorine emissions without the N_2O_5 – $ClNO_2$ chemistry on annual mean surface concentrations of Cl atoms and MDA8 O_3 in China.

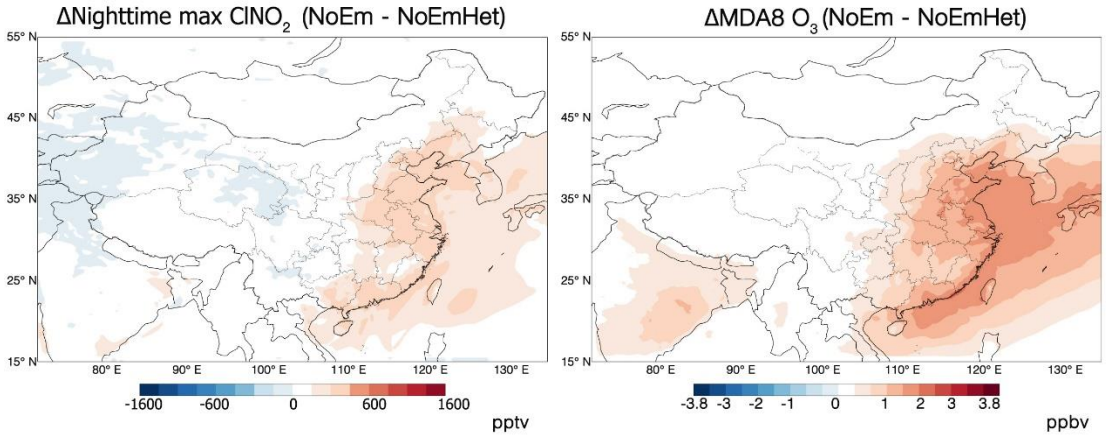

Figure S8. Effects of the N_2O_5-$ClNO_2$ chemistry without anthropogenic and biomass burning chlorine emissions on annual mean surface concentrations of nighttime max $ClNO_2$ and MDA8 O_3 in China.

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