

Table S1: Comparison of Historical $\Delta^{17}\text{O}$ Data with CMAQ Simulations from 2006 and 2019

Start	End	$\Delta^{17}\text{O}_{\text{obs}}$	$\Delta^{17}\text{O}_{2006\text{sim}}$	$\Delta^{17}\text{O}_{2019\text{sim}}$	Station	Reference
2003/9/12 0:00	2003/9/25 23:00	0.800	0.568	0.921	BR	Jenkins & Bao, 2006
2003/9/25 0:00	2003/11/3 23:00	0.520	0.677	0.930		
2003/11/3 0:00	2003/11/18 23:00	0.260	0.455	0.450		
2003/11/18 0:00	2003/12/1 23:00	0.520	1.731	0.485		
2003/12/1 0:00	2003/12/15 23:00	1.230	1.019	0.874		
2003/12/15 0:00	2003/12/29 23:00	0.520	1.184	0.837		
2003/12/29 0:00	2004/1/19 23:00	0.400	1.099	1.493		
2004/1/19 0:00	2004/2/2 23:00	0.550	0.771	0.451		
2004/2/2 0:00	2004/2/11 23:00	1.040	1.813	1.315		
2004/2/11 0:00	2004/2/26 23:00	0.590	1.170	0.939		
2004/4/28 0:00	2004/5/4 23:00	0.370	0.988	1.139		
2004/5/4 0:00	2004/5/19 23:00	1.430	0.826	1.022		
2004/5/19 0:00	2004/6/7 23:00	0.560	0.781	1.079		
2004/6/15 0:00	2004/6/29 23:00	0.660	0.634	0.535		
2004/6/29 0:00	2004/9/10 23:00	0.750	0.583	0.932		
2004/11/16 0:00	2005/1/4 23:00	0.960	1.265	0.686		
2005/1/31 0:00	2005/3/29 23:00	0.385	0.985	0.860		
1997/11/10 11:00	1997/11/10 18:00	0.400	1.124	0.527	LJ	Lee & Thiemens, 2001
1997/11/13 0:00	1997/11/13 8:00	0.700	1.209	2.410		
1997/11/13 9:00	1997/11/13 13:00	1.200	0.664	2.101		
1997/11/13 12:00	1997/11/13 15:00	0.900	0.558	2.011		
1998/1/29 7:00	1998/1/29 9:00	0.800	0.575	0.407		
1998/1/29 10:00	1998/1/29 11:00	0.500	0.588	0.372		
1998/1/29 11:00	1998/1/29 13:00	0.200	0.652	0.256		
1998/1/29 12:00	1998/1/29 14:00	0.200	0.684	0.183		
1998/2/3 8:00	1998/2/3 10:00	0.600	0.553	1.229		
1998/2/3 13:00	1998/2/3 14:00	1.400	1.783	1.008		
1998/2/3 14:00	1998/2/3 15:00	1.400	1.800	1.154		
1998/2/3 15:00	1998/2/3 16:00	0.200	1.431	1.443		
1998/2/3 16:00	1998/2/3 19:00	0.800	0.916	1.859		
1998/2/6 10:00	1998/2/6 13:00	0.700	1.547	2.754		
1998/2/6 13:00	1998/2/6 15:00	0.700	0.970	2.867		
1998/2/6 14:00	1998/2/6 16:00	1.300	0.737	3.068		
1996/5/6 0:00	1996/5/6 23:00	0.800	1.788	4.005		
1996/5/31 0:00	1996/5/31 23:00	0.708	1.258	4.225		
1996/7/22 0:00	1996/7/22 23:00	0.673	1.480	1.253		
1996/7/31 0:00	1996/7/31 23:00	0.645	1.546	1.527		
1996/9/18 0:00	1996/9/18 23:00	0.707	0.807	1.115		
1996/10/16 0:00	1996/10/16 23:00	0.809	0.954	1.170		
1996/10/21 0:00	1996/10/21 23:00	0.935	0.813	0.927		
1997/1/7 0:00	1997/1/7 23:00	0.931	0.509	0.461		
1997/2/7 0:00	1997/2/7 23:00	0.694	0.509	1.186		

1997/2/21 0:00	1997/2/21 23:00	1.093	0.893	2.259	
1997/3/7 0:00	1997/3/7 23:00	1.286	0.681	1.546	
1997/3/19 0:00	1997/3/19 23:00	0.633	3.177	0.824	
1997/4/2 0:00	1997/4/2 23:00	1.021	1.057	2.416	
1997/8/5 0:00	1997/8/5 23:00	0.581	0.998	2.912	
1997/12/19 0:00	1997/12/19 23:00	0.480	1.465	1.527	
1996/8/18 0:00	1996/8/18 23:00	1.000	1.183	1.579	WMRS
1996/8/20 0:00	1996/8/20 23:00	1.100	0.867	0.973	
1996/8/23 0:00	1996/8/23 23:00	1.300	0.777	0.906	
1997/3/7 0:00	1997/3/7 23:00	1.500	4.036	4.327	
1997/3/11 0:00	1997/3/11 23:00	1.600	3.231	2.457	
1997/3/14 0:00	1997/3/14 23:00	1.400	3.100	3.338	

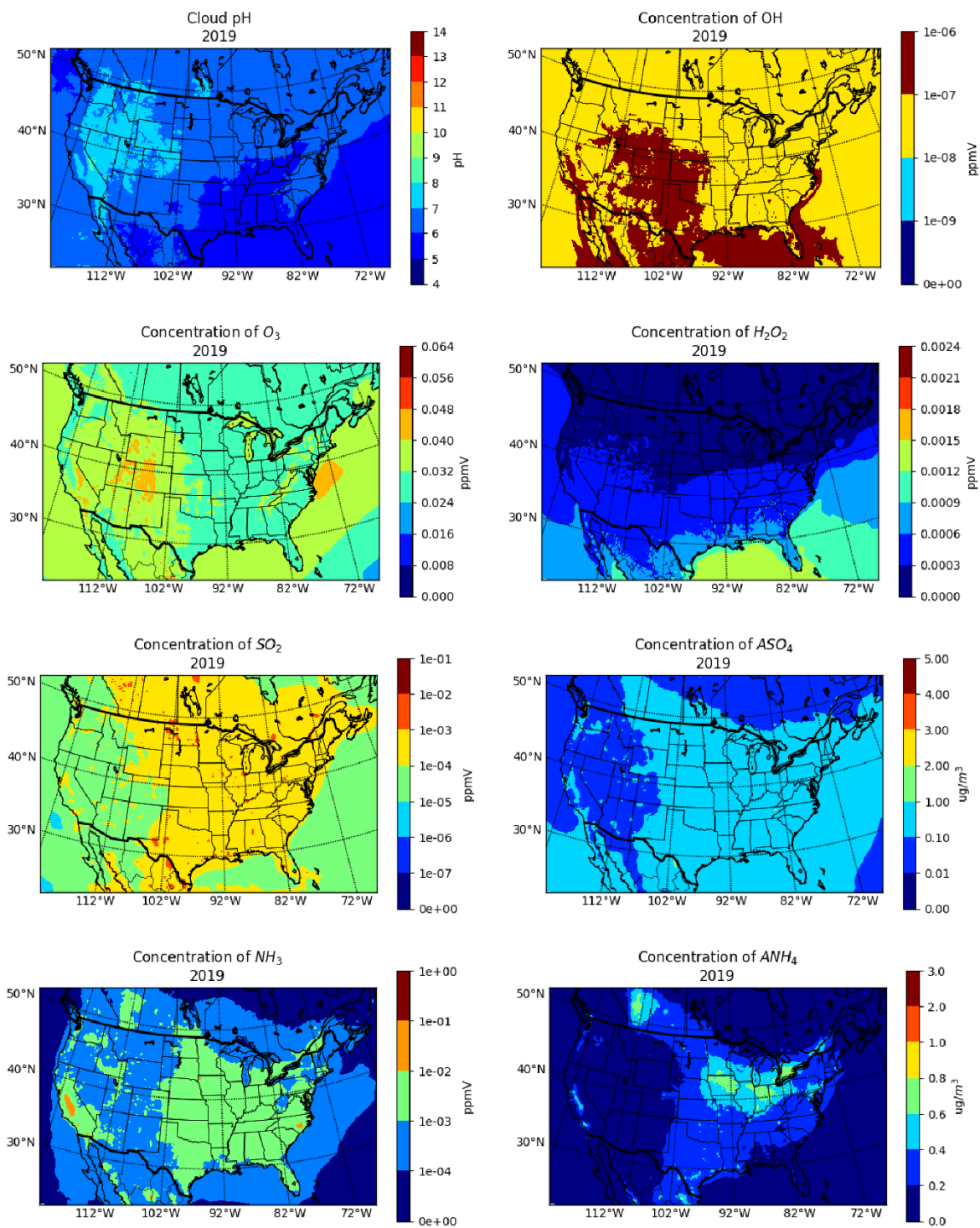


Fig. S1: The geographical distribution of the concentrations of atmospheric chemicals related to the ASO_4 formation pathways across the continental US in the year 2019.

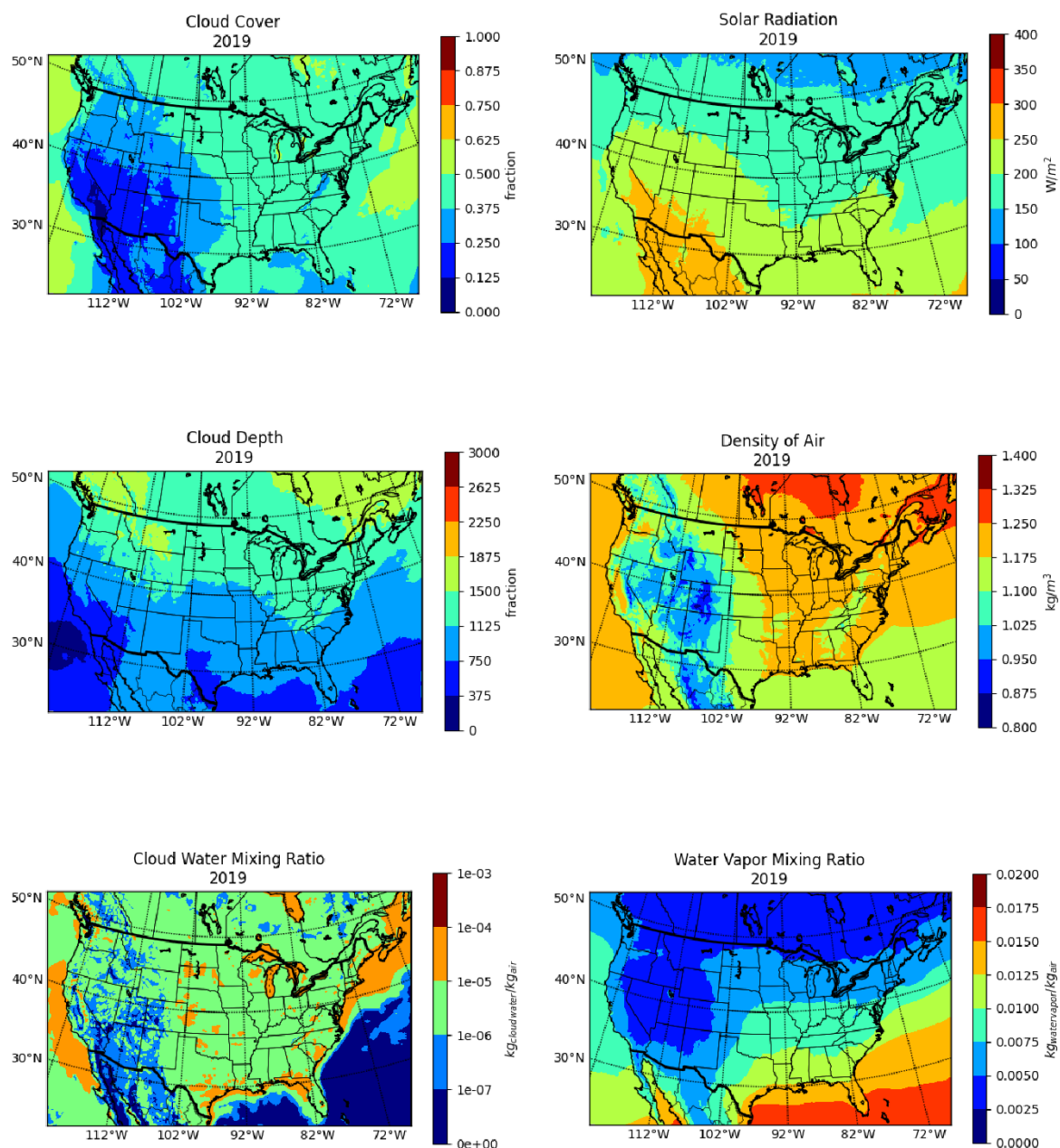


Fig. S2: The geographical distribution of the meteorological factors related to the ASO_4 formation pathways across the continental US in the year 2019.

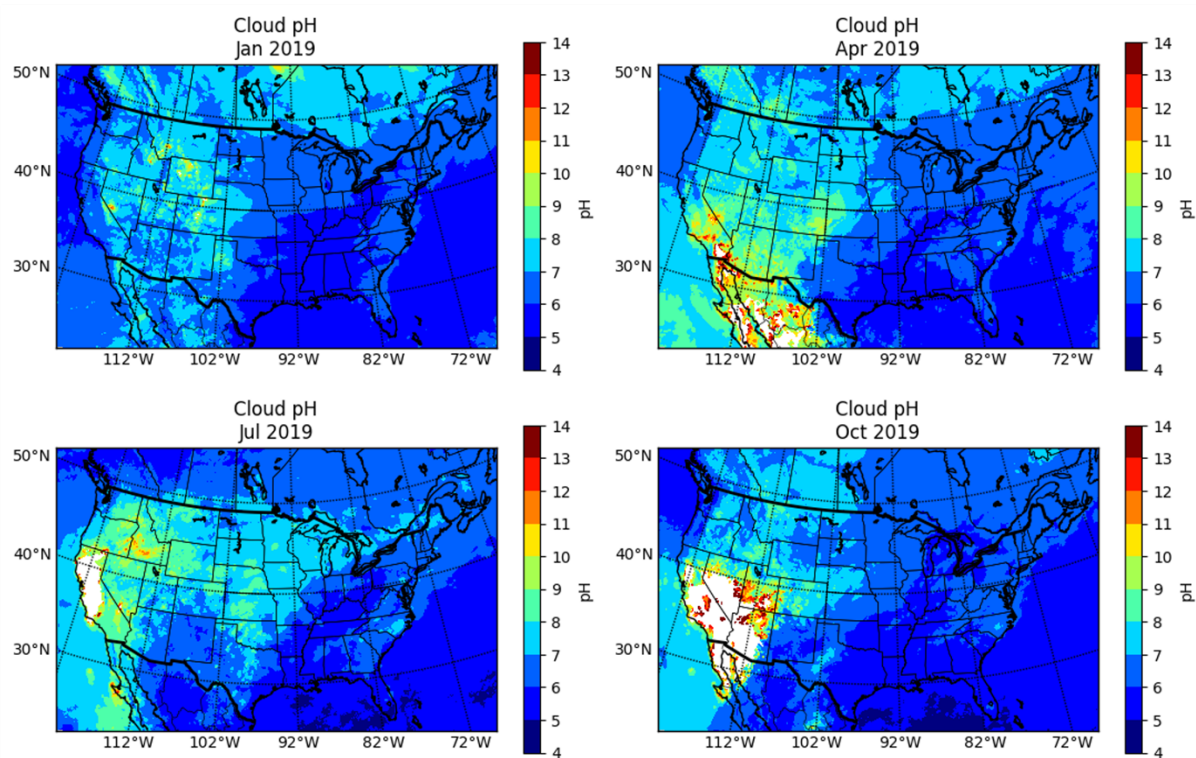


Fig. S3: The geographical distribution of cloud pH across each grid throughout the continental US for the year 2019 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

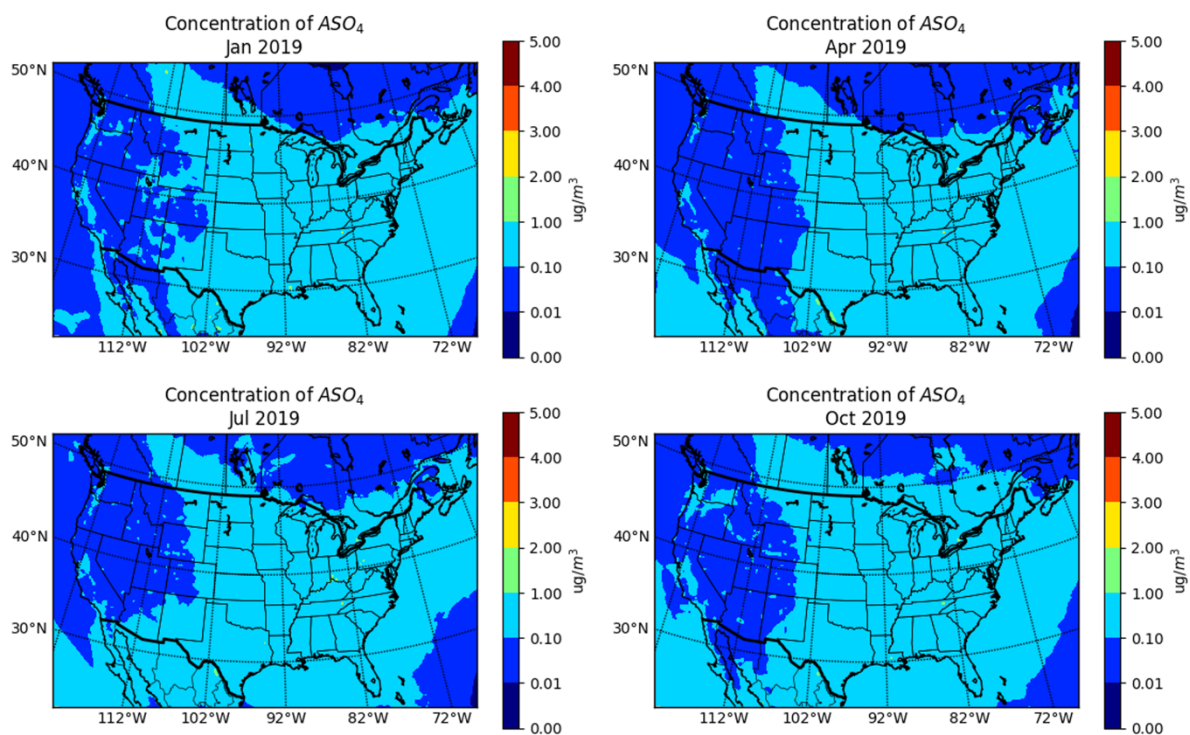


Fig. S4: The geographical distribution of cloud water ASO_4 concentration across the continental US for the year 2019 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

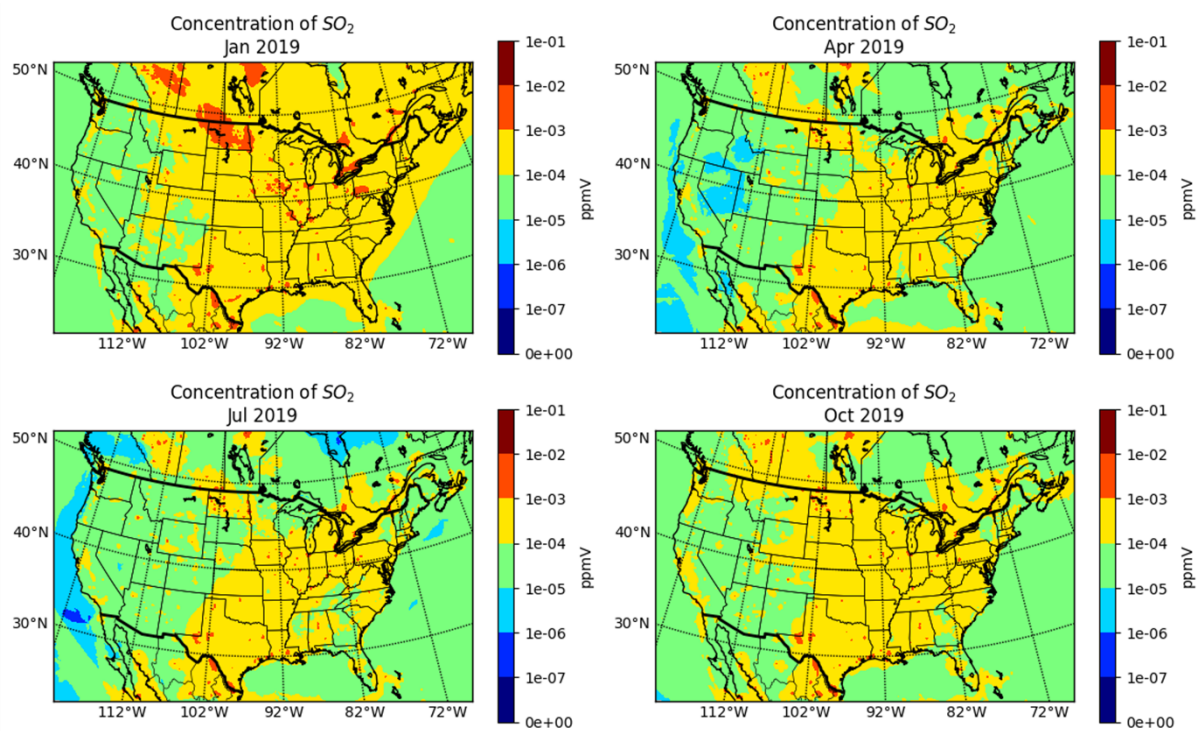


Fig. S5: The geographical distribution of SO_2 concentration across the continental US for the year 2019 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

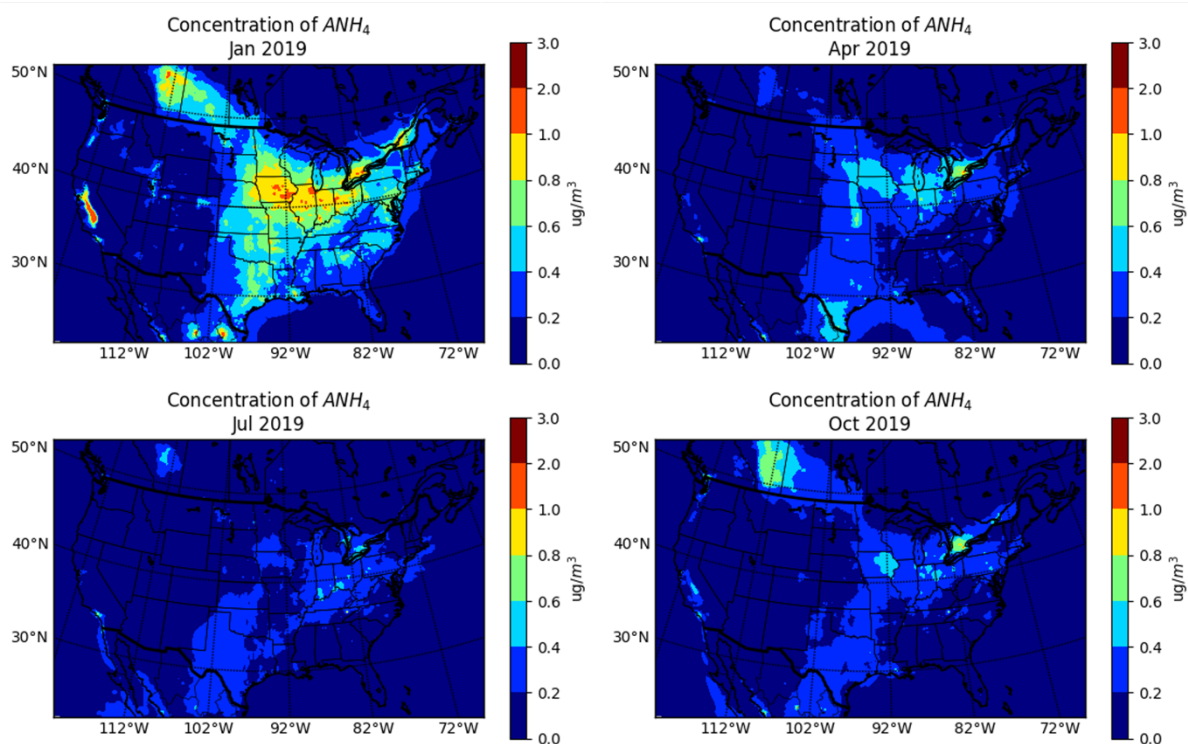


Fig. S6: The geographical distribution of cloud water ANH_4 concentration across the continental US for the year 2019 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

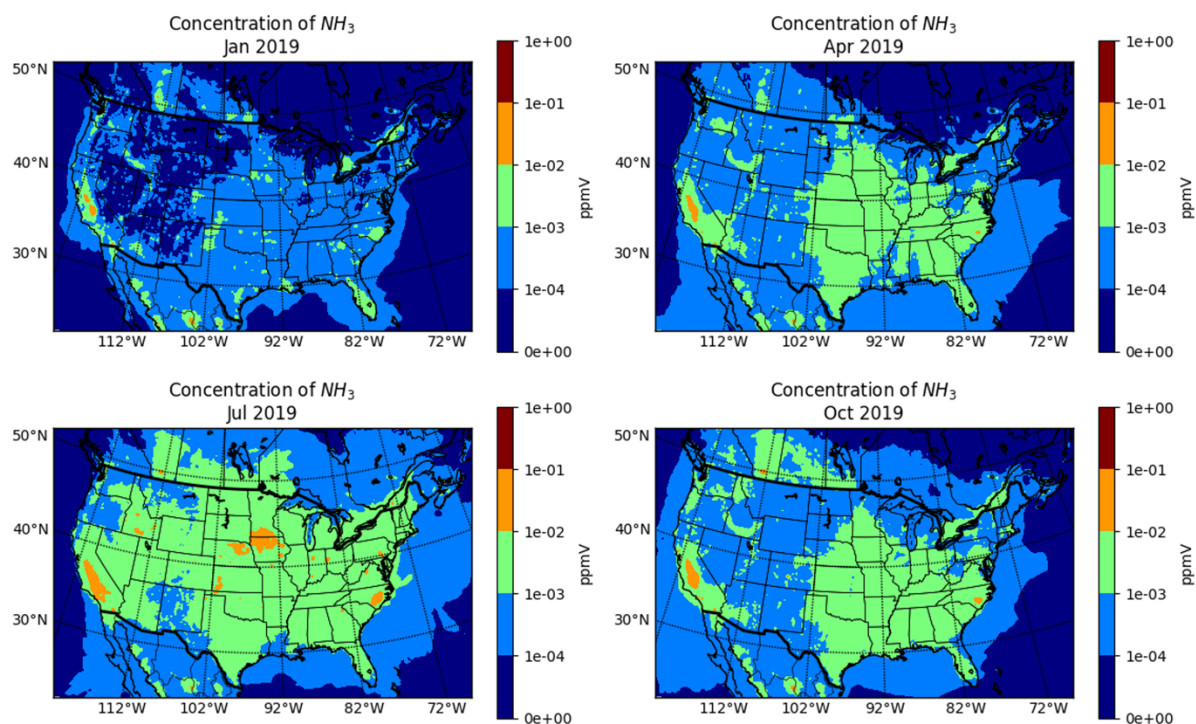


Fig. S7: The geographical distribution of NH_3 concentration across the continental US for the year 2019 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

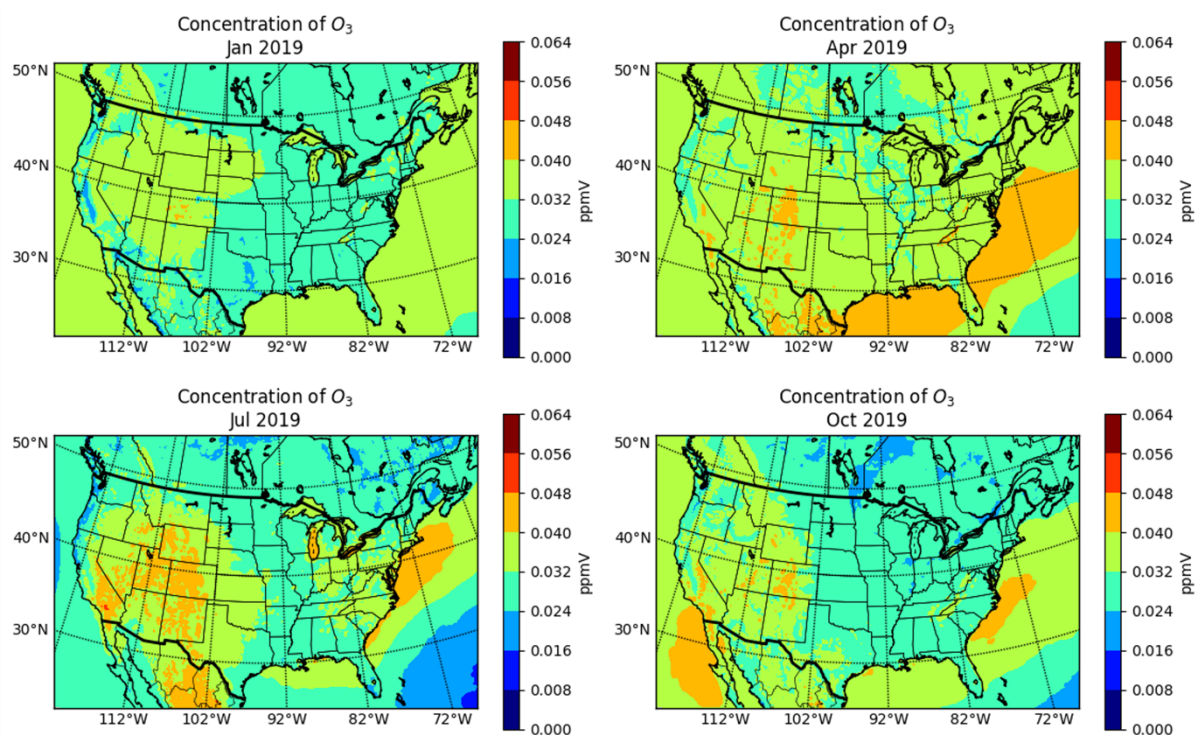


Fig. S8: The geographical distribution of O₃ concentration across the continental US for the year 2019 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

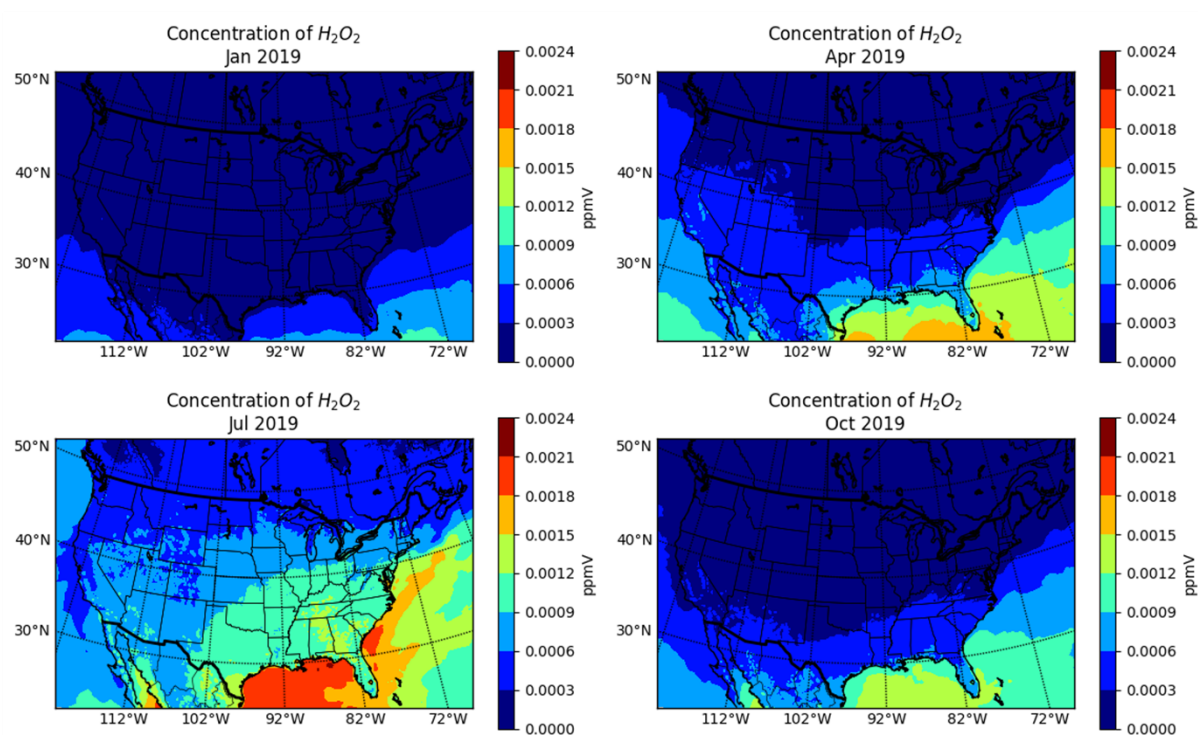


Fig. S9: The geographical distribution of H_2O_2 concentration across the continental US for the year 2019 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

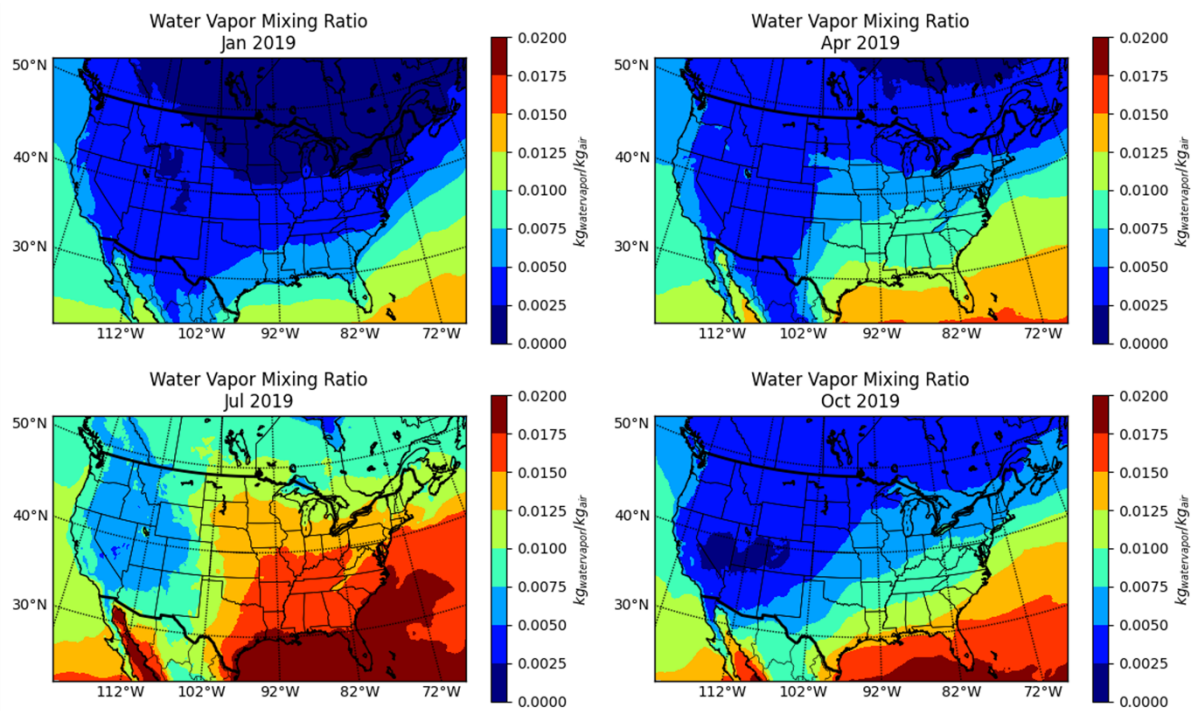


Fig. S10: The geographical distribution of water vapor mixing ratio across the continental US for the year 2019 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

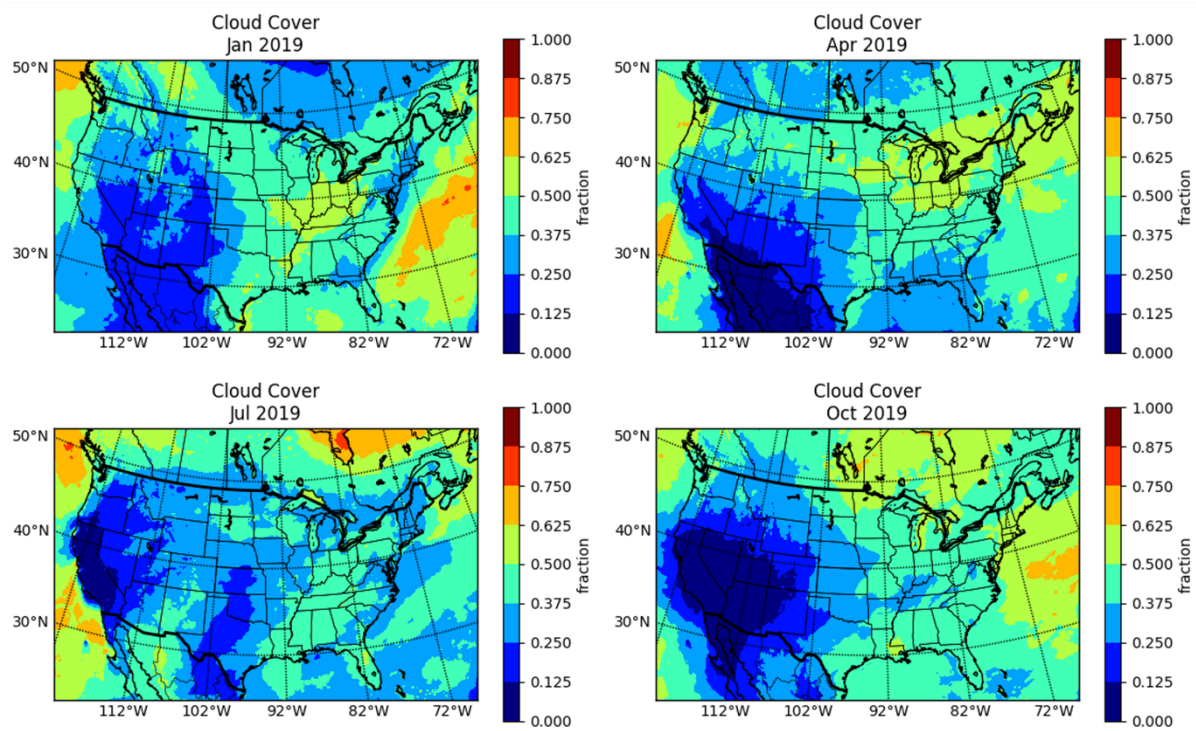


Fig. S11: The geographical distribution of cloud cover across the continental US for the year 2019 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

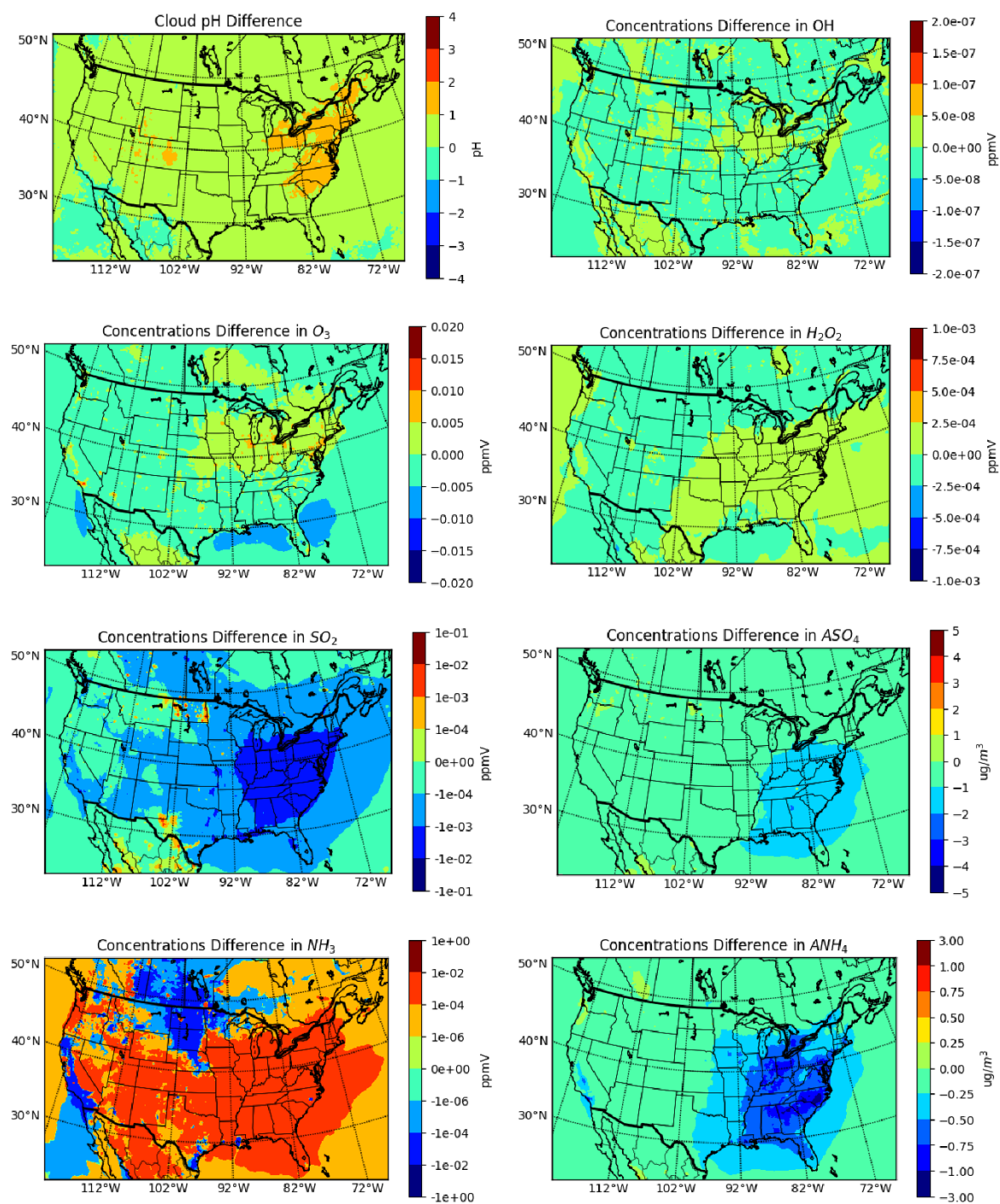


Fig. S12: The geographical distribution of the change in the concentrations of atmospheric chemicals related to the ASO_4 formation pathways across the continental US, from 2006 to 2019.

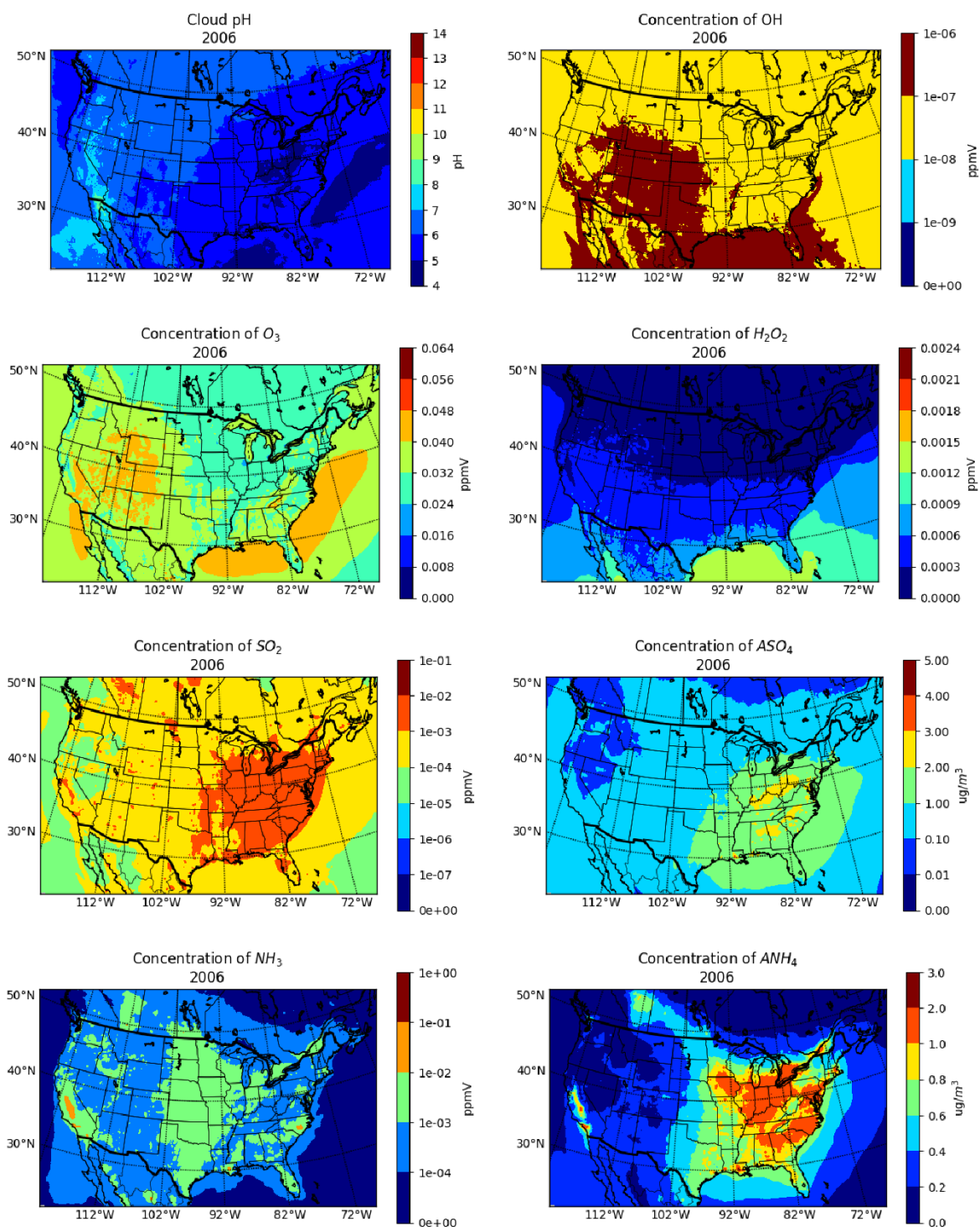


Fig. S13: The geographical distribution of the concentrations of atmospheric chemicals related to the ASO_4 formation pathways across the continental US in the year 2006.

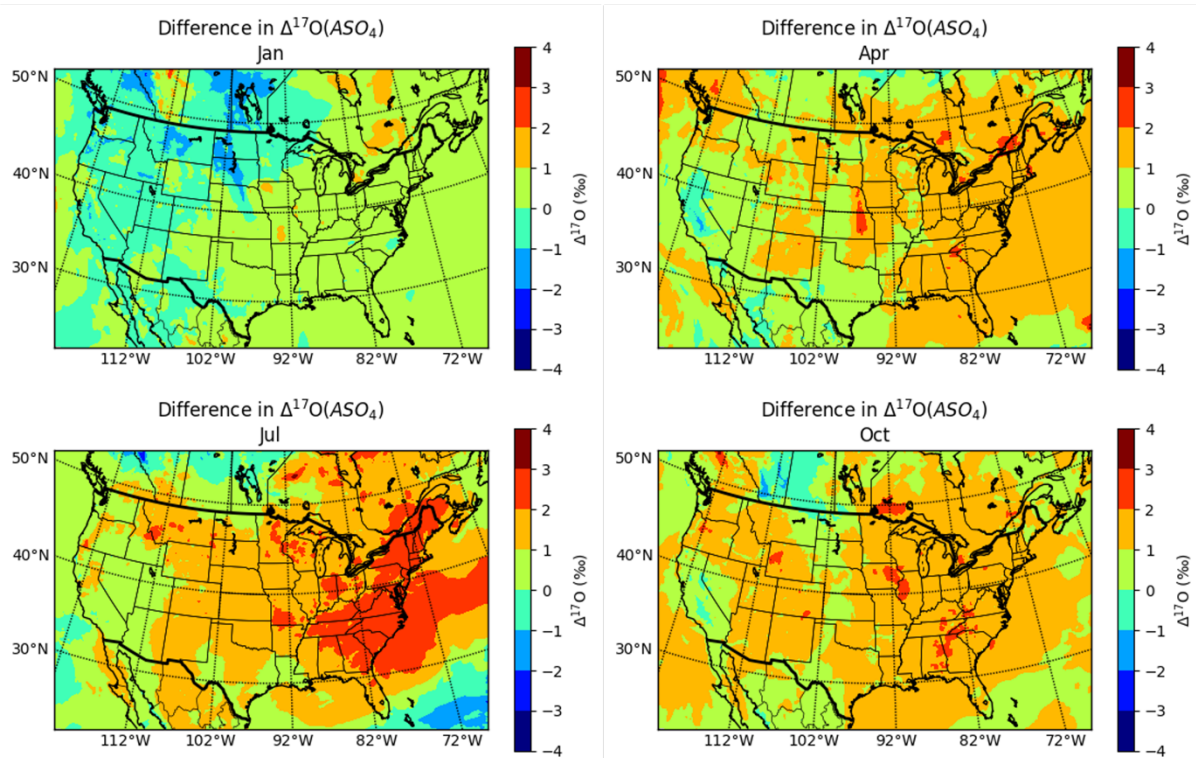


Fig. S14: The geographical distribution of the change in $\Delta^{17}\text{O}$ values across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

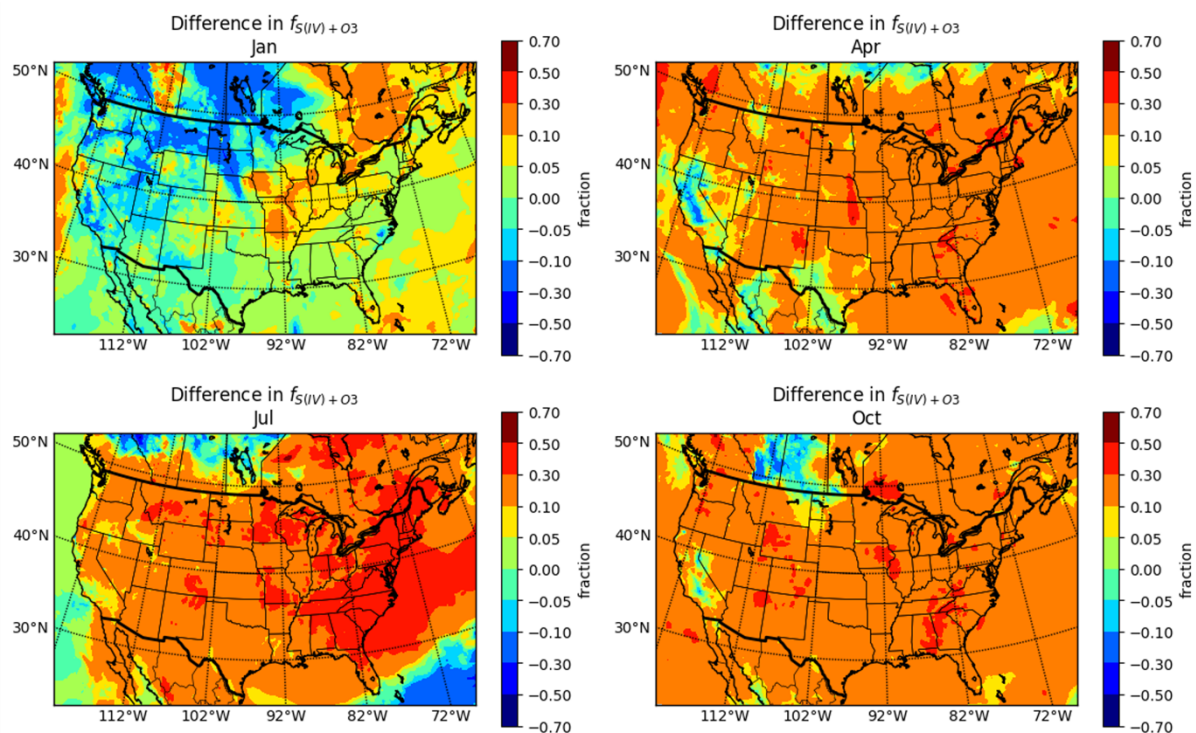


Fig. S15: The geographical distribution of the change in the fraction of ASO₄ formation from S(IV)+O₃ pathway across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

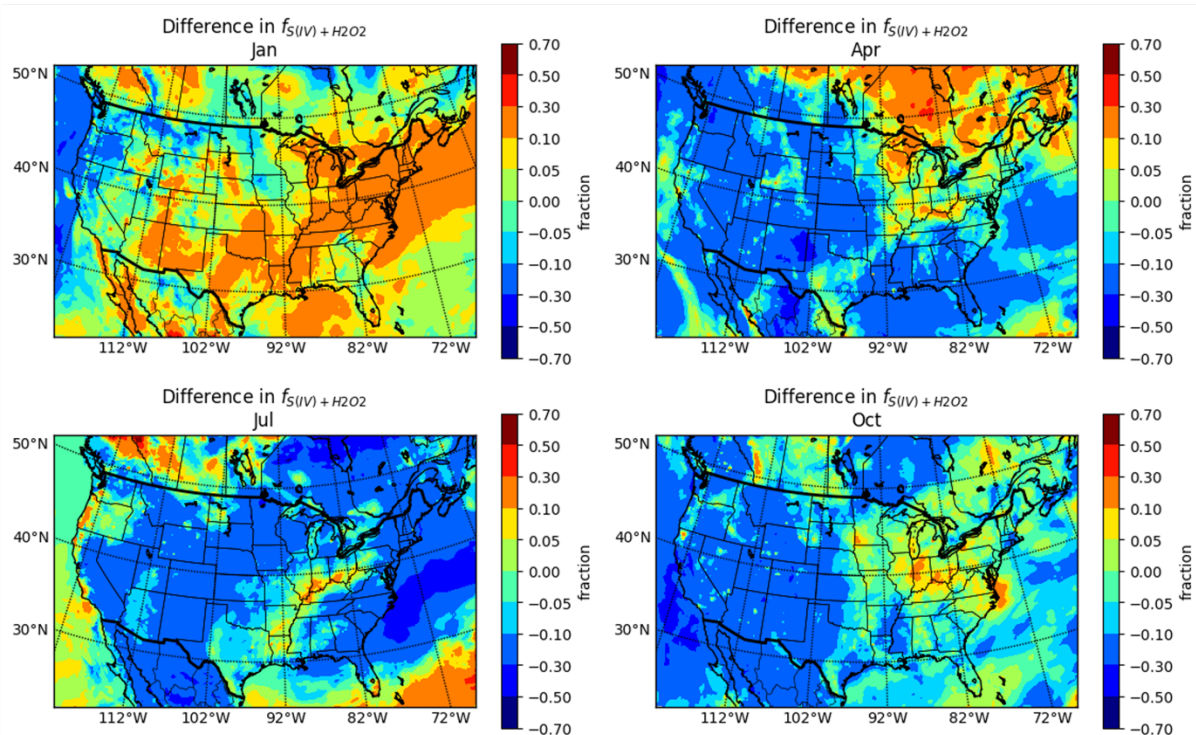


Fig. S16: The geographical distribution of the change in the fraction of ASO₄ formation from S(IV)+H₂O₂ pathway across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

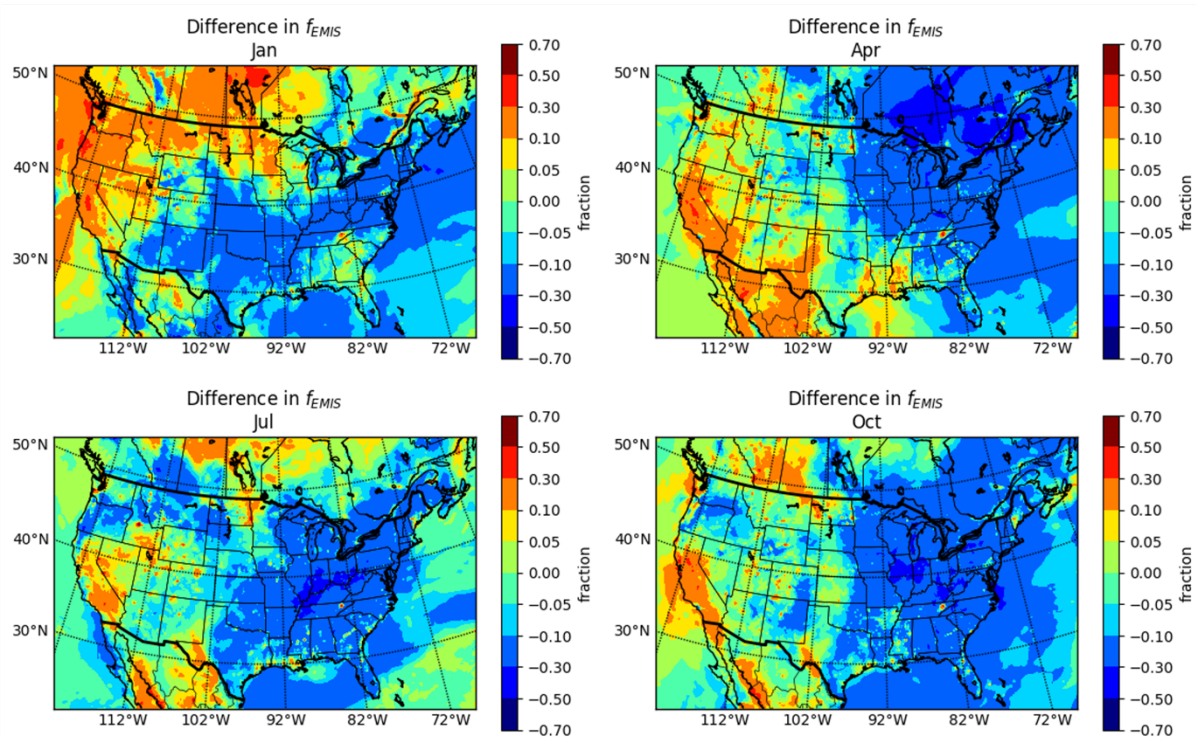


Fig. S17: The geographical distribution of the change in the fraction of ASO₄ formation from primary emission across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

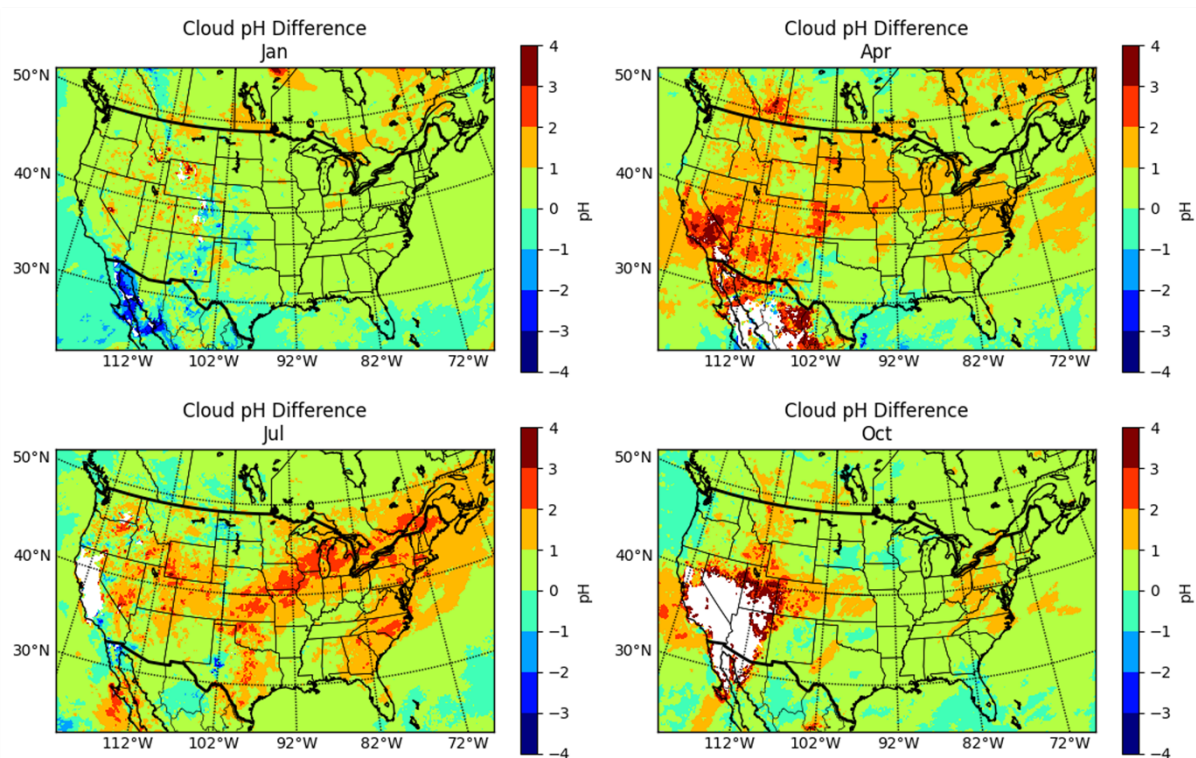


Fig. S18: The geographical distribution of the change in cloud pH across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

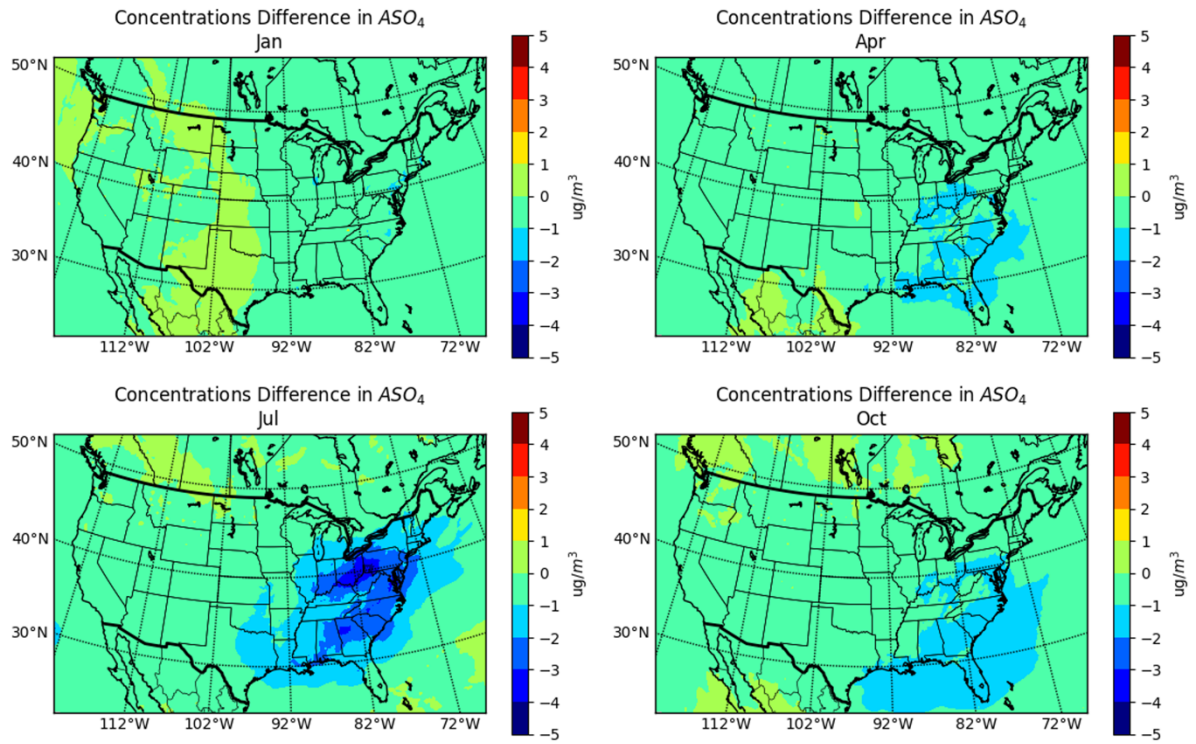


Fig. S19: The geographical distribution of the change in cloud water ASO_4 concentration across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

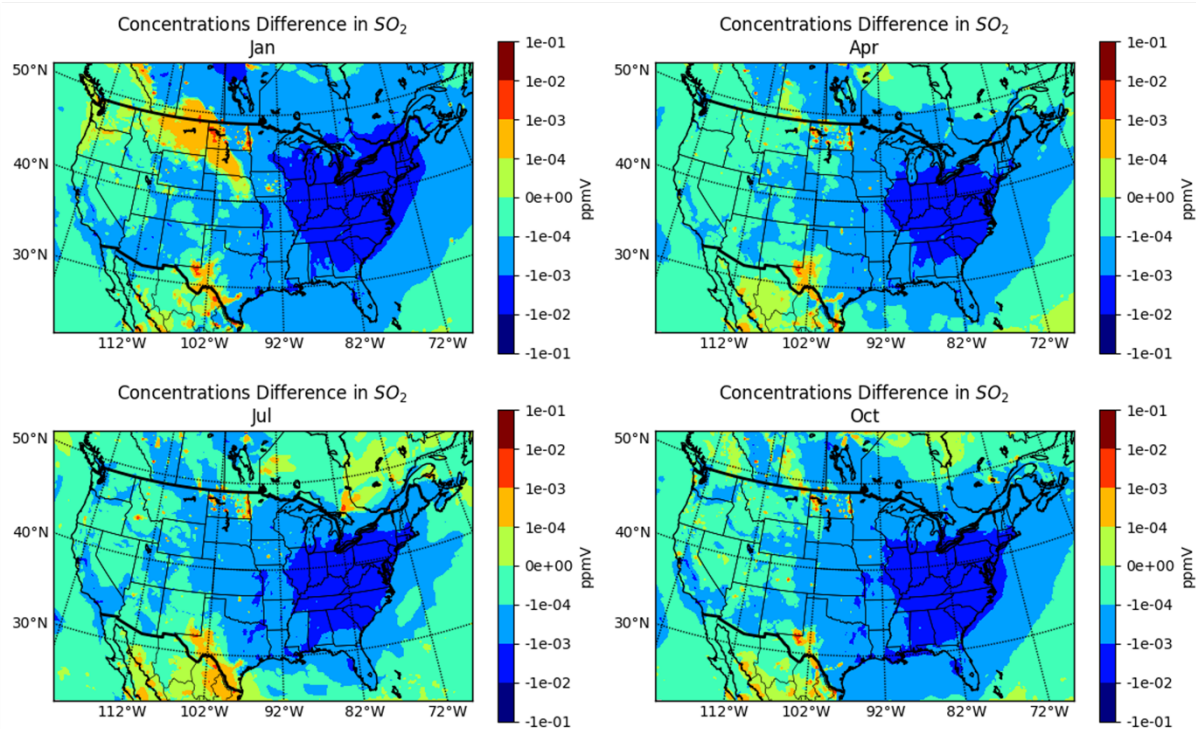


Fig. S20: The geographical distribution of the change in SO_2 concentration across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

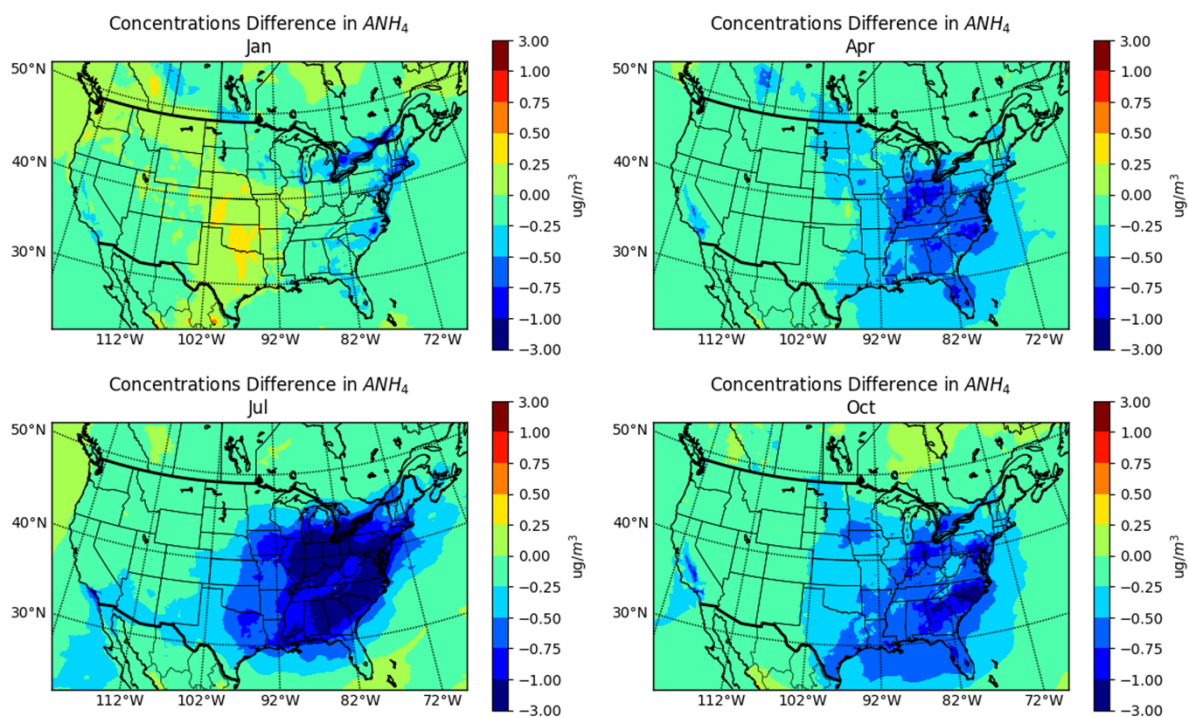


Fig. S21: The geographical distribution of the change in cloud water ANH_4 concentration across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

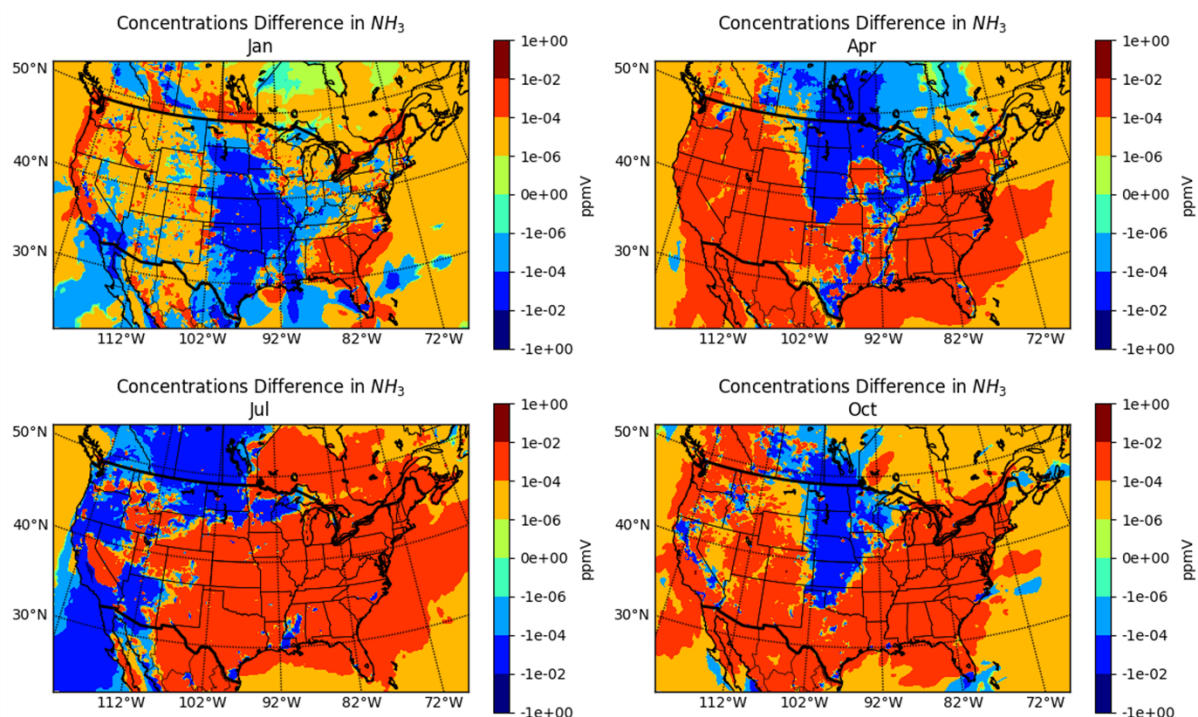


Fig. S22: The geographical distribution of the change in NH_3 concentration across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

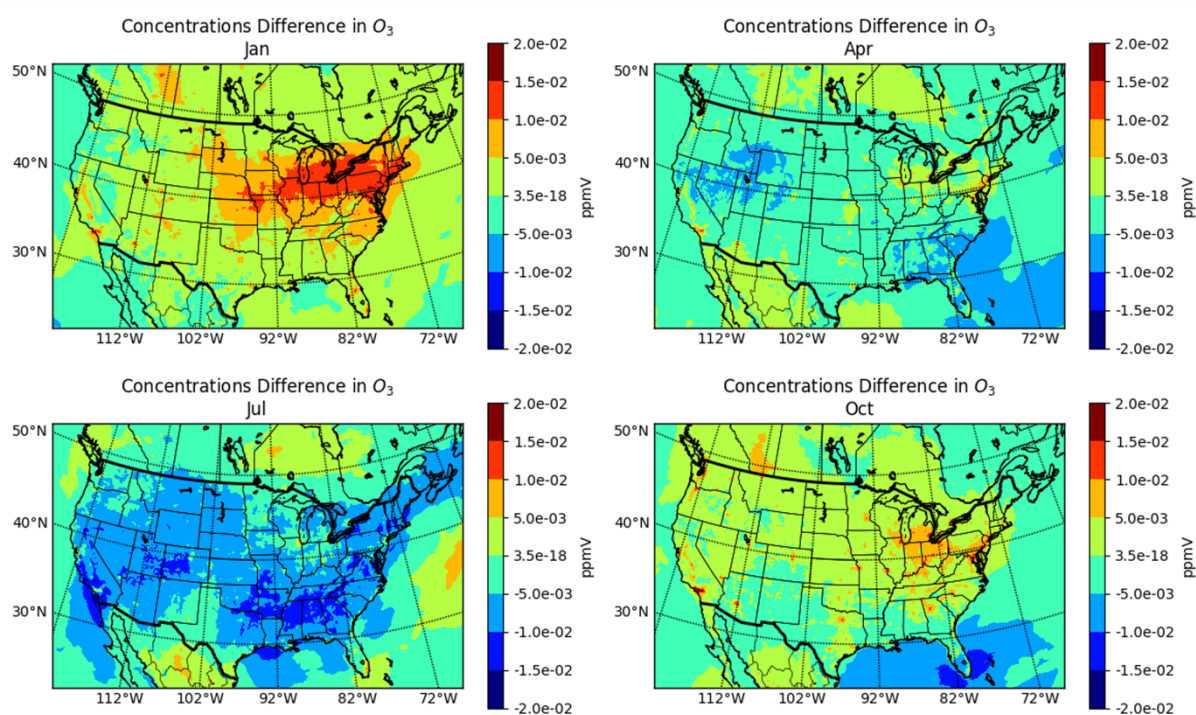


Fig. S23: The geographical distribution of the change in O₃ concentration across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

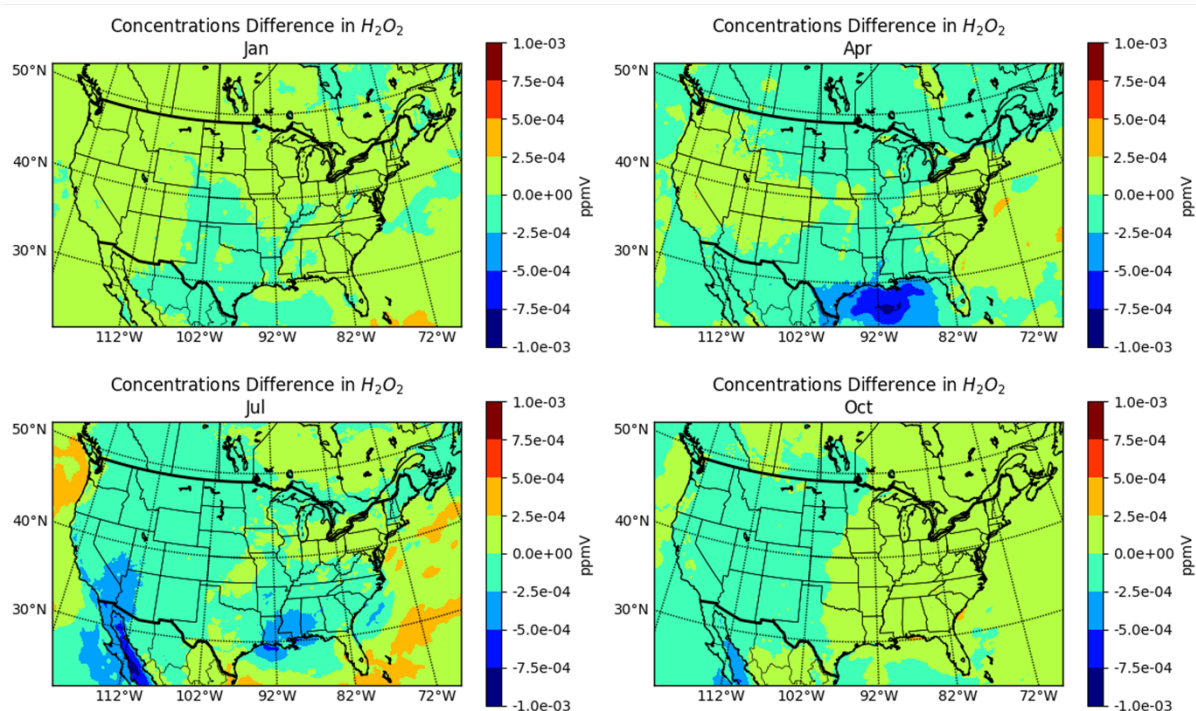


Fig. S24: The geographical distribution of the change in H_2O_2 concentration across the continental US in each season (winter: Jan, spring: Apr, summer: July, fall: Oct), from 2006 to 2019.

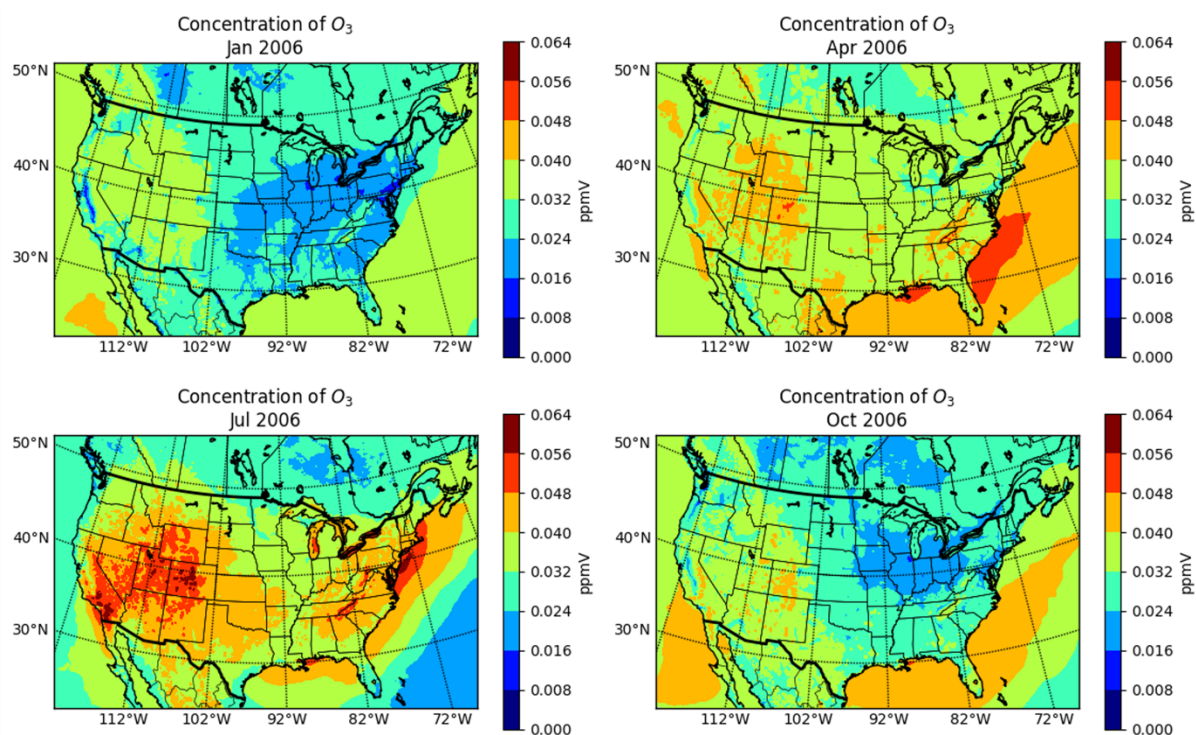


Fig. S25: The geographical distribution of O₃ concentration across the continental US for the year 2006 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).

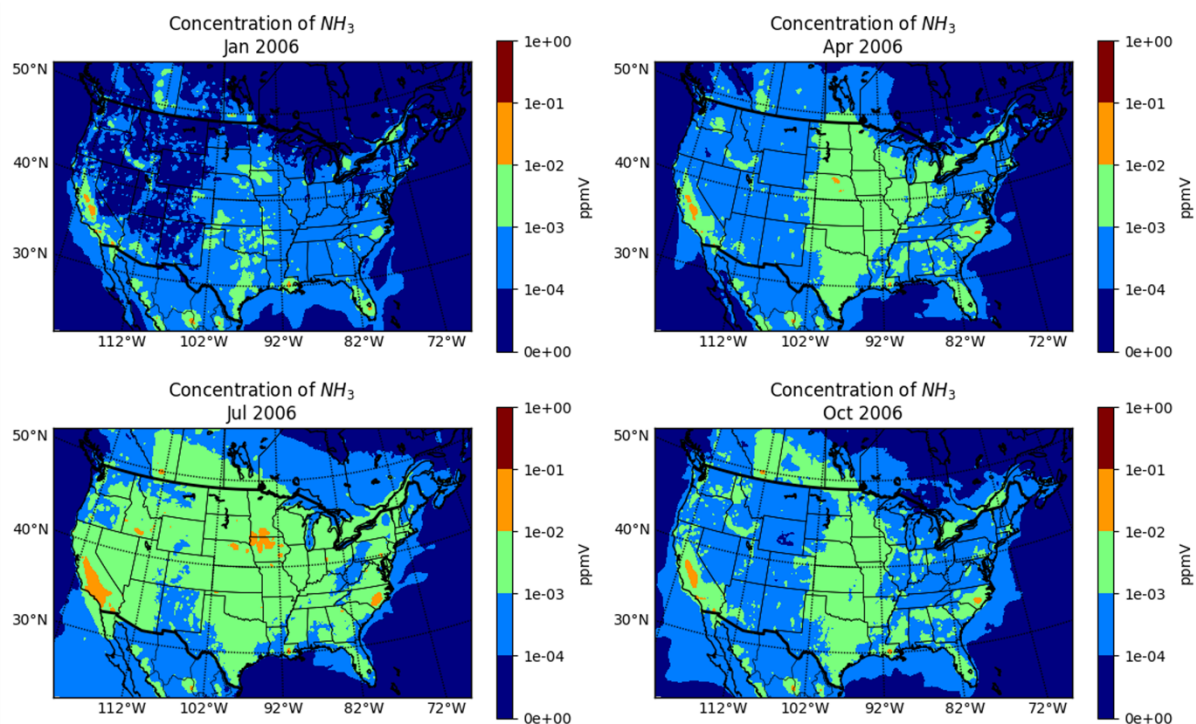


Fig. S26: The geographical distribution of NH_3 concentration across the continental US for the year 2006 in each season (winter: Jan, spring: Apr, summer: July, fall: Oct).