Modeling ammonia and its uptake by secondary organic aerosol over China

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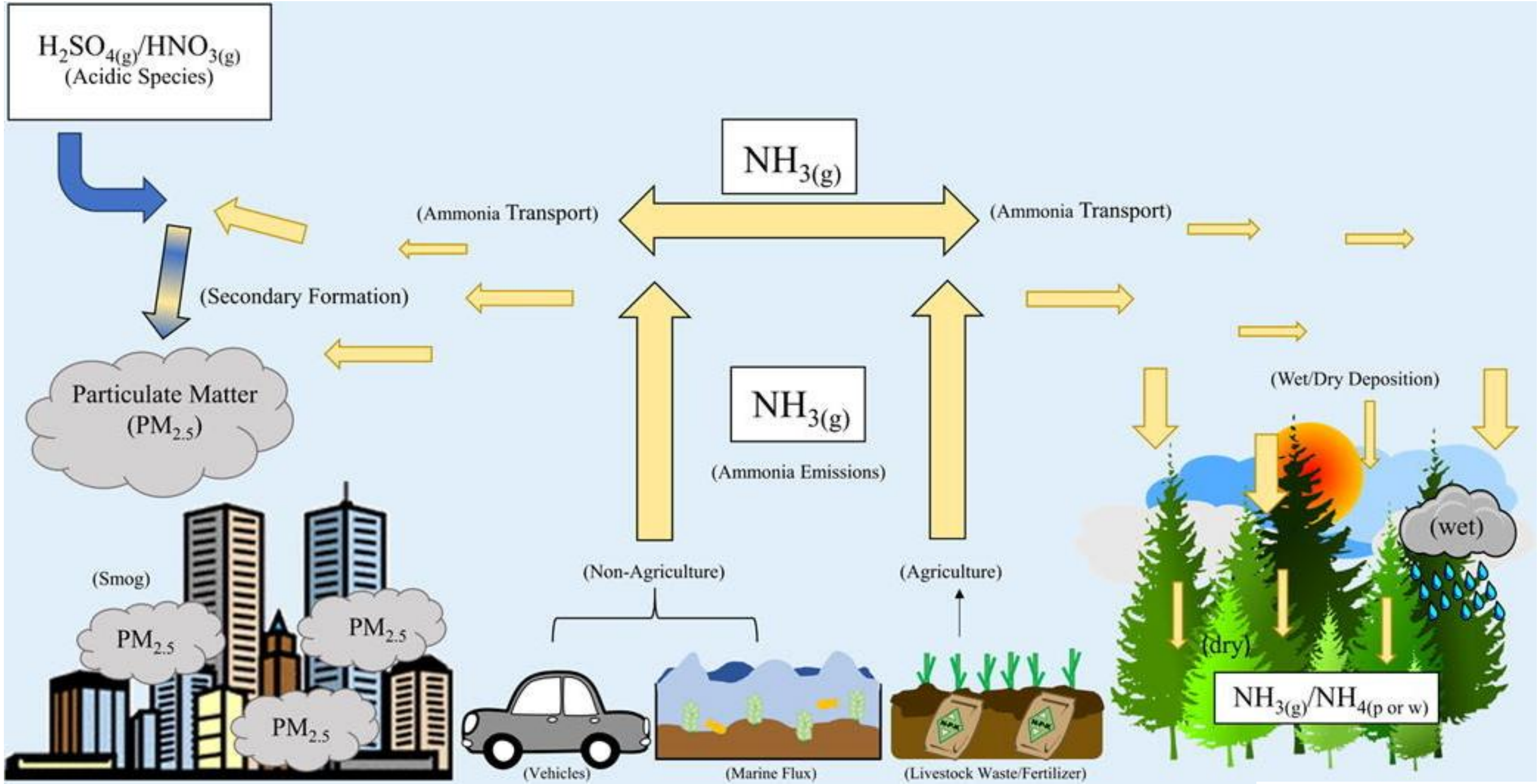
Ammonia and its transformation in the atmosphere

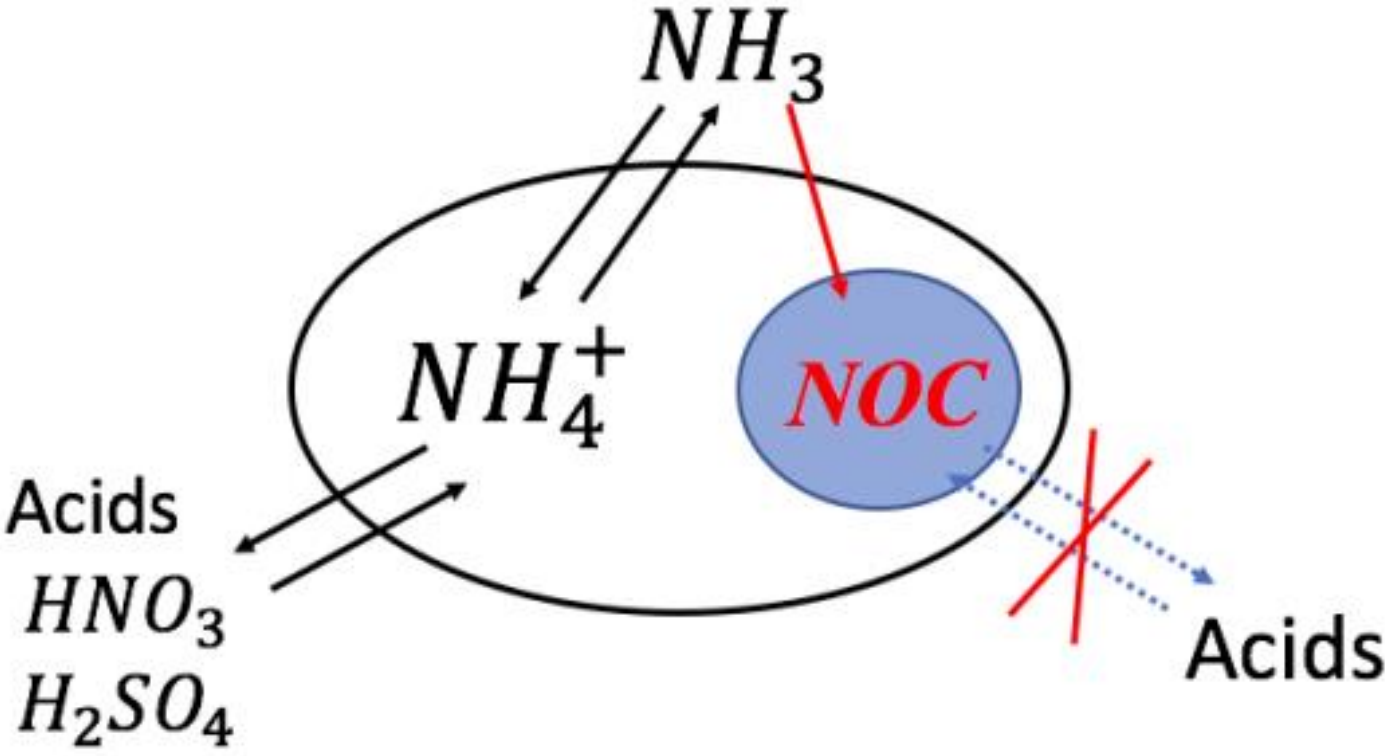


Berner *et al*. 2020, STOTEN

Major Effects of Ammonia in the Atmosphere:

- **Affect the formation and composition of tropospheric aerosols** [e.g. Jimenez et al., 2009]
- **Influence aerosol pH and total reactive nitrogen deposition** [e.g. Guo et al., 2018; Weber et al., 2016]
- **Affect urban air quality and regional climate change** [e.g. Reiss et al., 2007]

Mechanisms of interactions between NH_3 and particles in the model



- CMAQ already includes reactions between NH_3 and HNO_3 / H_2SO_4 leading to inorganic salts.
- We are adding a process that converts some (up to 10%) of SOA compounds into NOC which is not basic enough to neutralize acids.

Revisions made in AERO6 aerosol module in CMAQ model

$$S_{SOA} = \sum_{j=1}^{x} \left(S_j \times \frac{\sum_{i=1}^{y} m_{ij}}{\sum_{k=1}^{z} m_{ij}} \right) \quad (1)$$

- In equation (1), x, y, and z represent the total number of modes that contain SOA species, the total number of SOA species in mode j and the total number of aerosol species in mode j, respectively. Because SOA species only exist in the Aitken mode and the accumulation mode, the value of x is setting to 2.

$$k = \gamma \times \frac{v_{NH_3} \times S_{SOA}}{4} \quad (2)$$

- In equation (2), γ represents the uptake coefficient for ammonia and v_{NH3} represents the average speed of NH_3 molecules under the condition of 298 K. The first-order rate constant of NH_3 uptake in each grid cell is calculated by using above equations and then multiplied by the NH_3 concentration to determine the NH_3 taken up by SOA.

Scenarios settings

1. (Base case) ——without uptake process
2. (Scenario I) $\gamma = 10^{-3}$
3. (Scenario II) $\gamma = 10^{-4}$
4. (Scenario III) $\gamma = 10^{-5}$

- The parameterization used in this study assumed that one NH_3 molecule that reacts with SOA forms nitrogen-containing organic compounds (NOCs), which can further react to form heterocyclic organic compounds with two H_2O molecules as the by-product.

WRF-CMAQ model configuration

Anthropogenic Emission:MEIC 2016

Biogenic Emission: MEGANv2.1

Boundary data： NCEP-FNL

Simulation time： the year of 2017

Winter (1st Jan - 28th Feb)

Summer (1st June – 31st July)

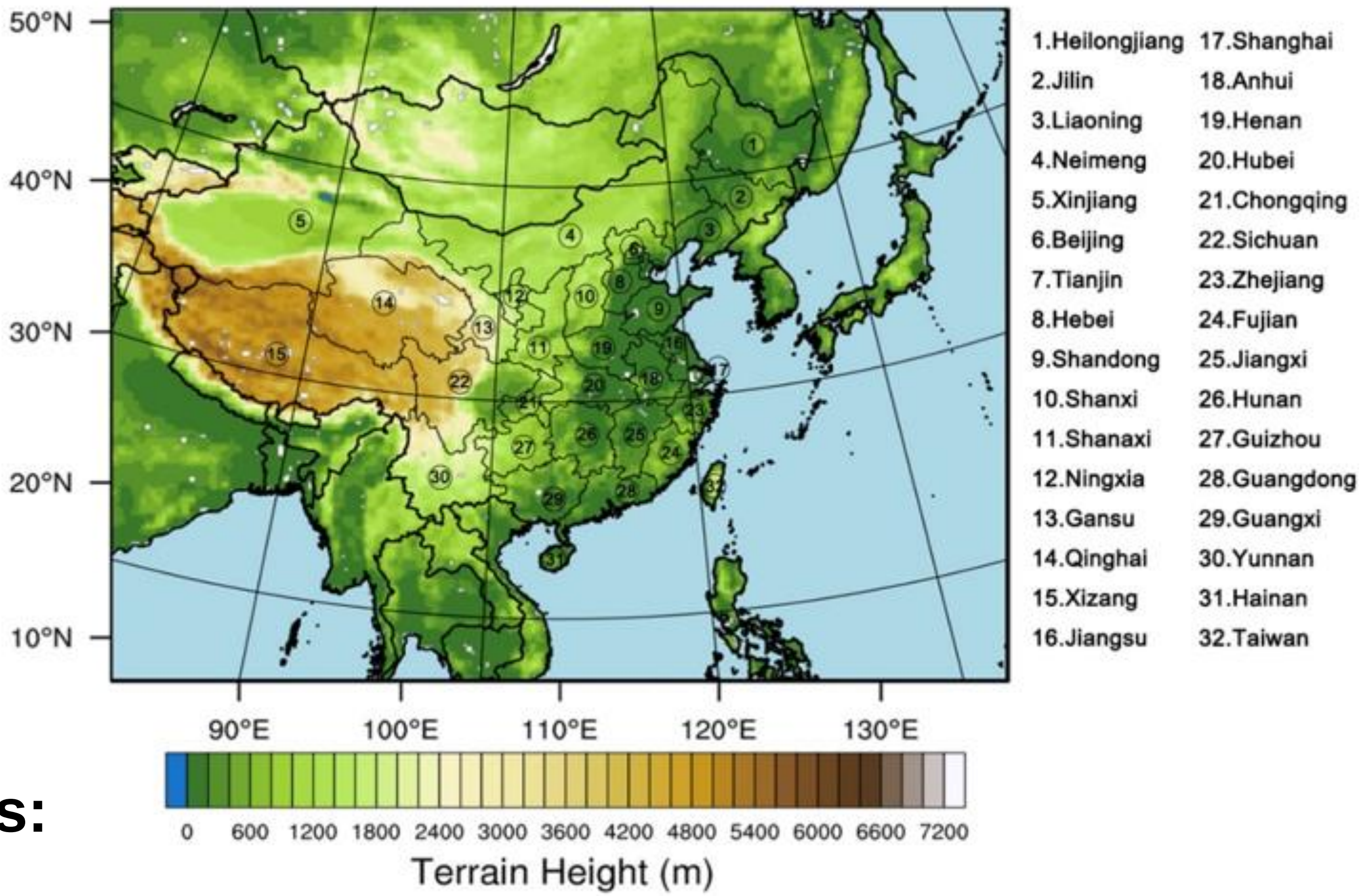
Chemical mechanism： CB06AERO6

Main WRF parameterization schemes:

Noah land surface process scheme

RRTMG radiation transmission scheme

YSU boundary layer parameterization scheme



➢ Domain of simulation

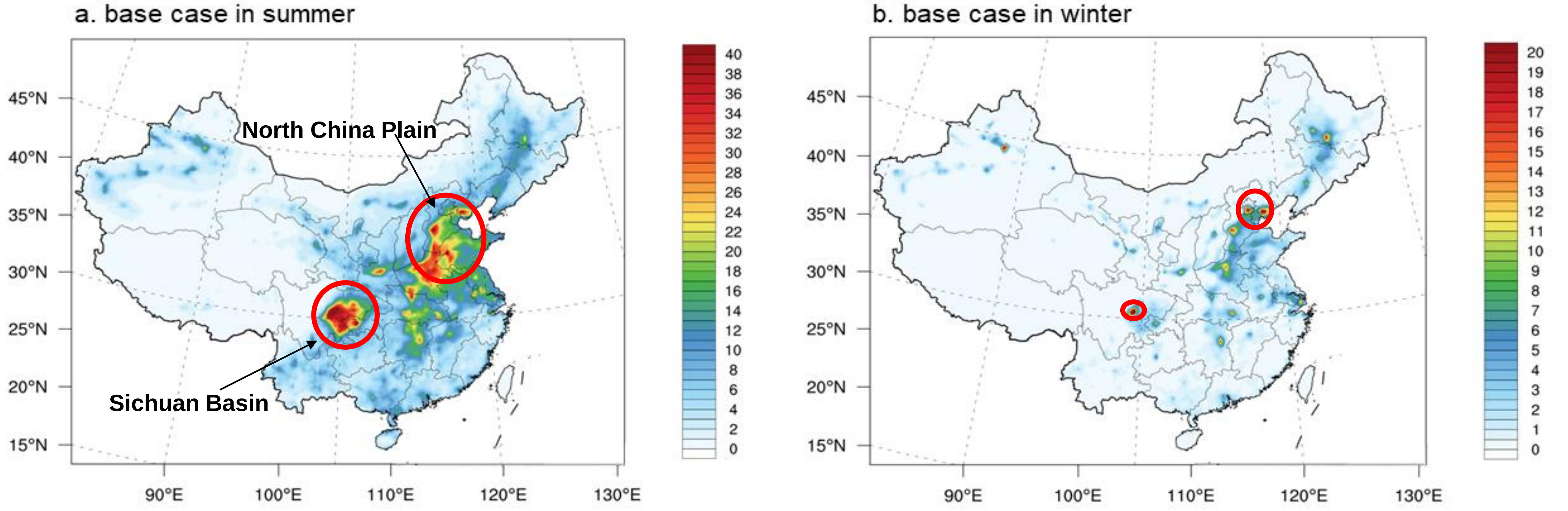
Model evaluation against surface observations

Comparison between simulation results for $PM_{2.5}$ and observations from the CNEMC monitoring network for seasonal averages.

Scenario	Period	Obs.mean $\mu g\ m^{-3}$	Sim.mean $\mu g\ m^{-3}$	MB $\mu g\ m^{-3}$	NMB %	NME %	RMSE $\mu g\ m^{-3}$
Base	Summer	28.5	30.2	1.7	10.3	36.8	22.3
$\gamma = 10^{-5}$	Summer	28.5	29.6	1.1	8.7	44.9	18.2
$\gamma = 10^{-4}$	**Summer**	**28.5**	**28.3**	**-0.2**	**-2.8**	**33.0**	**15.9**
$\gamma = 10^{-3}$	Summer	28.5	26.4	-2.1	-6.4	52.4	36.8
Base	Winter	74.0	68.5	-5.5	-12.4	42.0	30.1
$\gamma = 10^{-5}$	**Winter**	**74.0**	**67.4**	**-6.6**	**-14.9**	**46.8**	**33.2**
$\gamma = 10^{-4}$	Winter	74.0	66.9	-7.1	-17.1	49.1	36.0
$\gamma = 10^{-3}$	Winter	74.0	63.7	-10.3	-18.6	52.3	42.1

- **Overall, the base case CMAQ simulation successfully reproduced the seasonally averaged $PM_{2.5}$ concentration.**
- **When the process of NH_3 uptake to SOA was included, the simulated $PM_{2.5}$ concentrations decreased in both seasons, with the magnitude of the decrease increasing with the assumed uptake coefficient.**

Ammonia concentration simulated in base case

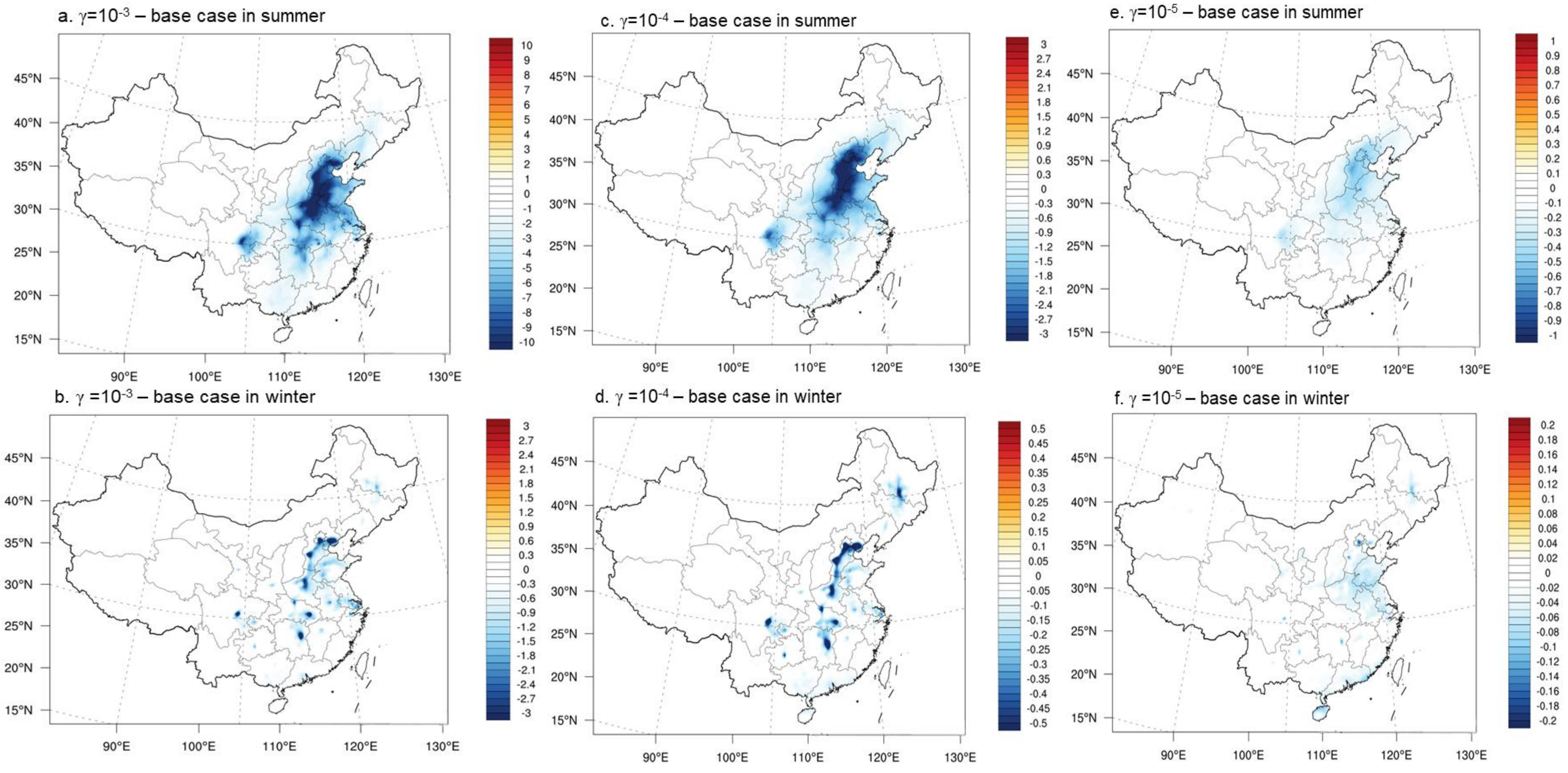


NH_3 spatial distribution

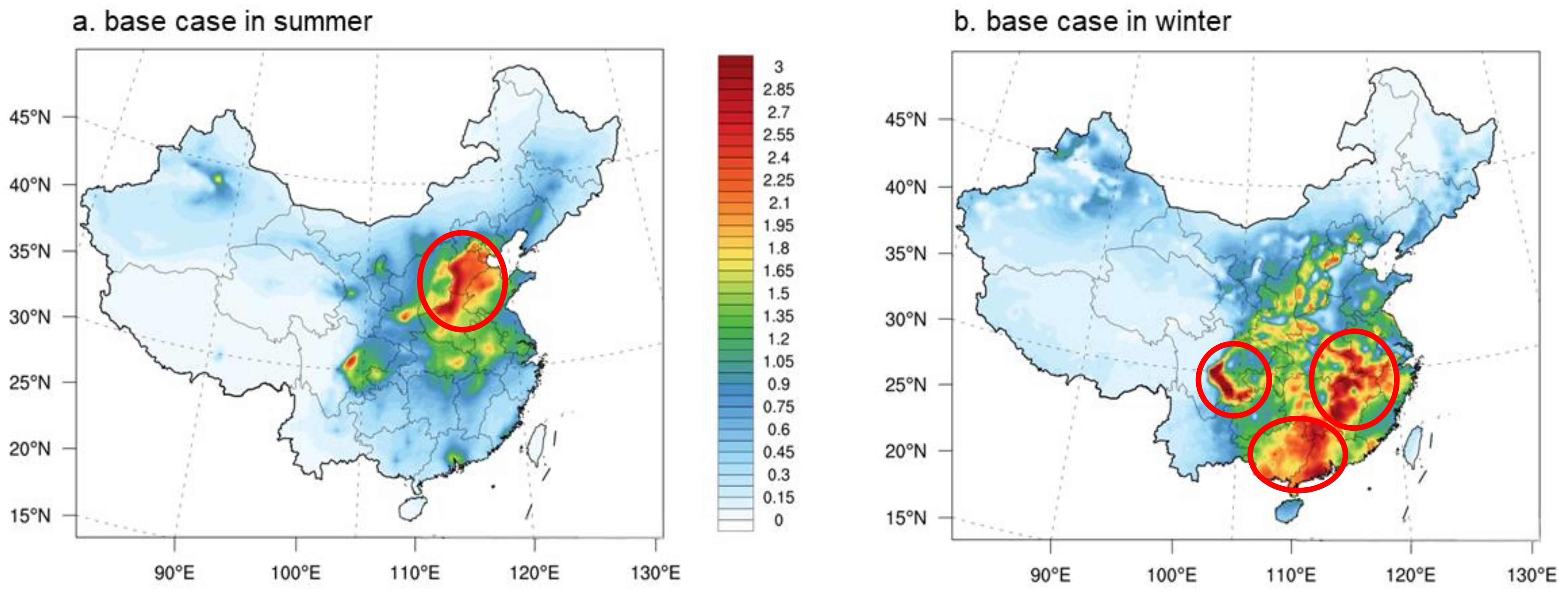
- **During summer, high NH_3 concentrations are observed over southwestern China (Sichuan Basin) and North China Plain**
- **In winter, the simulation results show some highly localized ammonia hotspots in eastern China**

➢ **How will the implementation of NH_3 uptake mechanism affect inorganic and organic aerosols?**

Spatial distribution of the difference in surface NH_3 concentrations

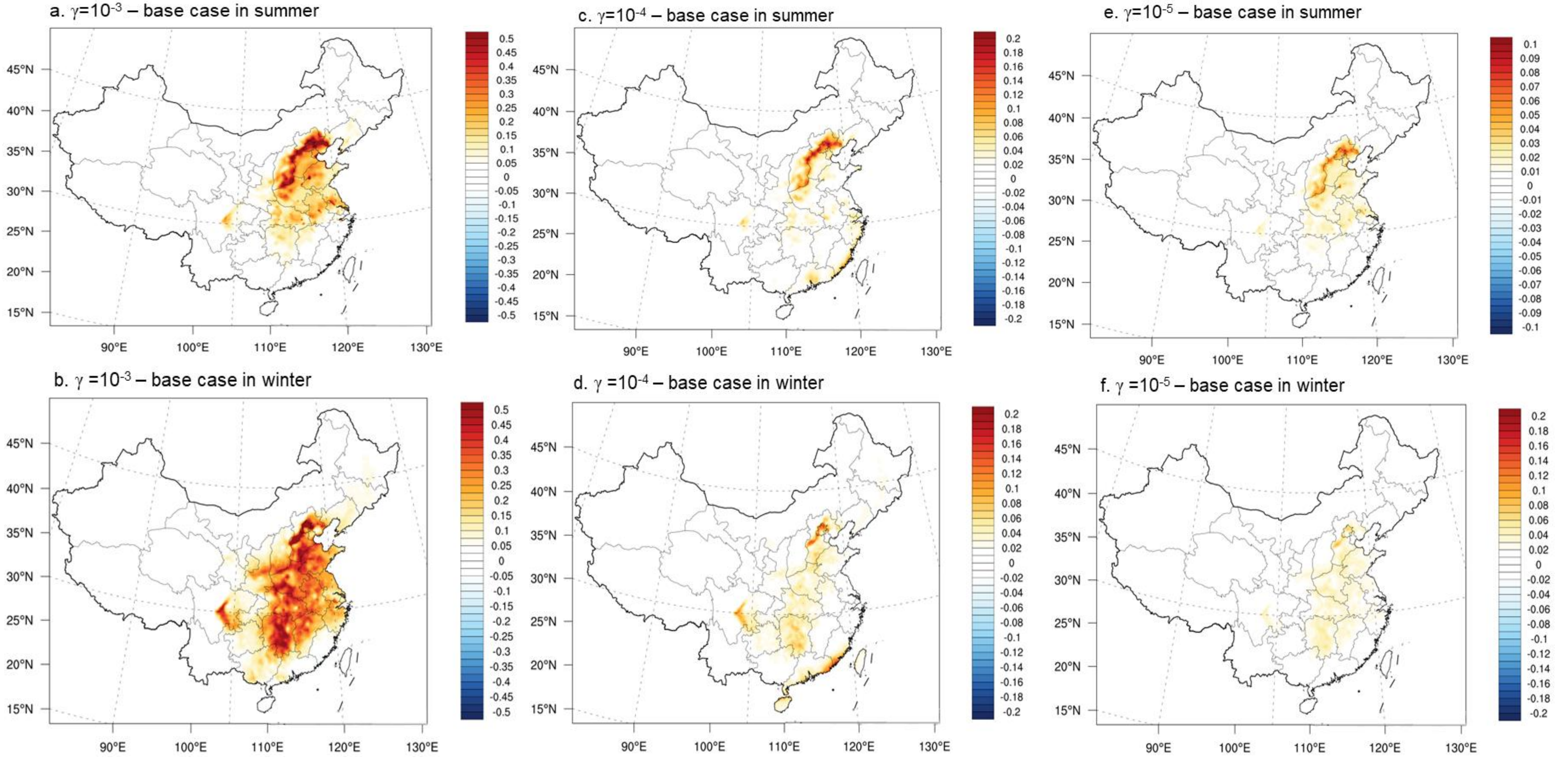


Spatial distribution of simulated surface HNO_3 concentrations



Reflecting differences in NOx emissions, production and loss pathways, and gas-particle partitioning.

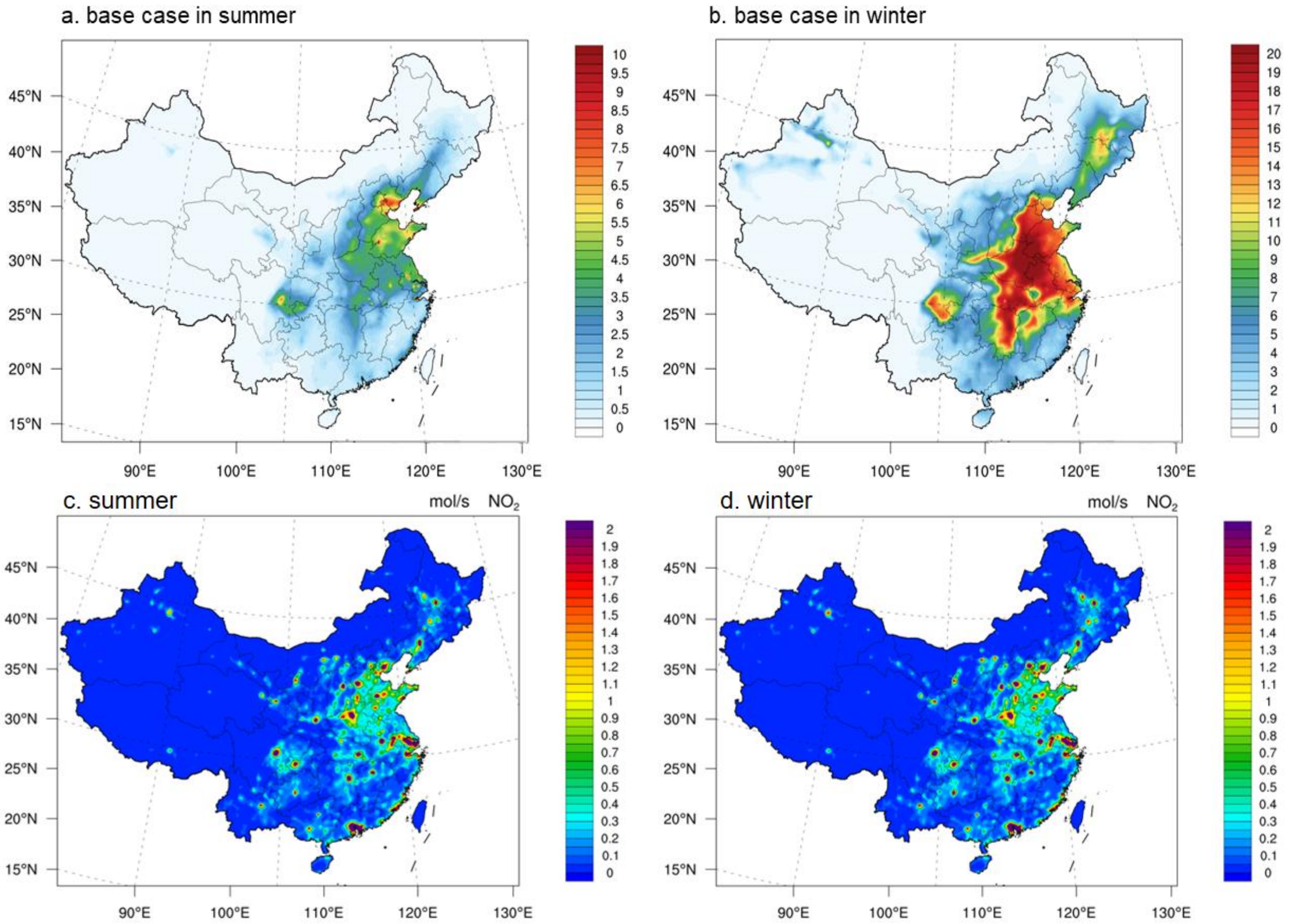
Spatial distribution of the difference in surface HNO_3 concentrations



Spatial distribution of simulated surface NO_3^- concentrations

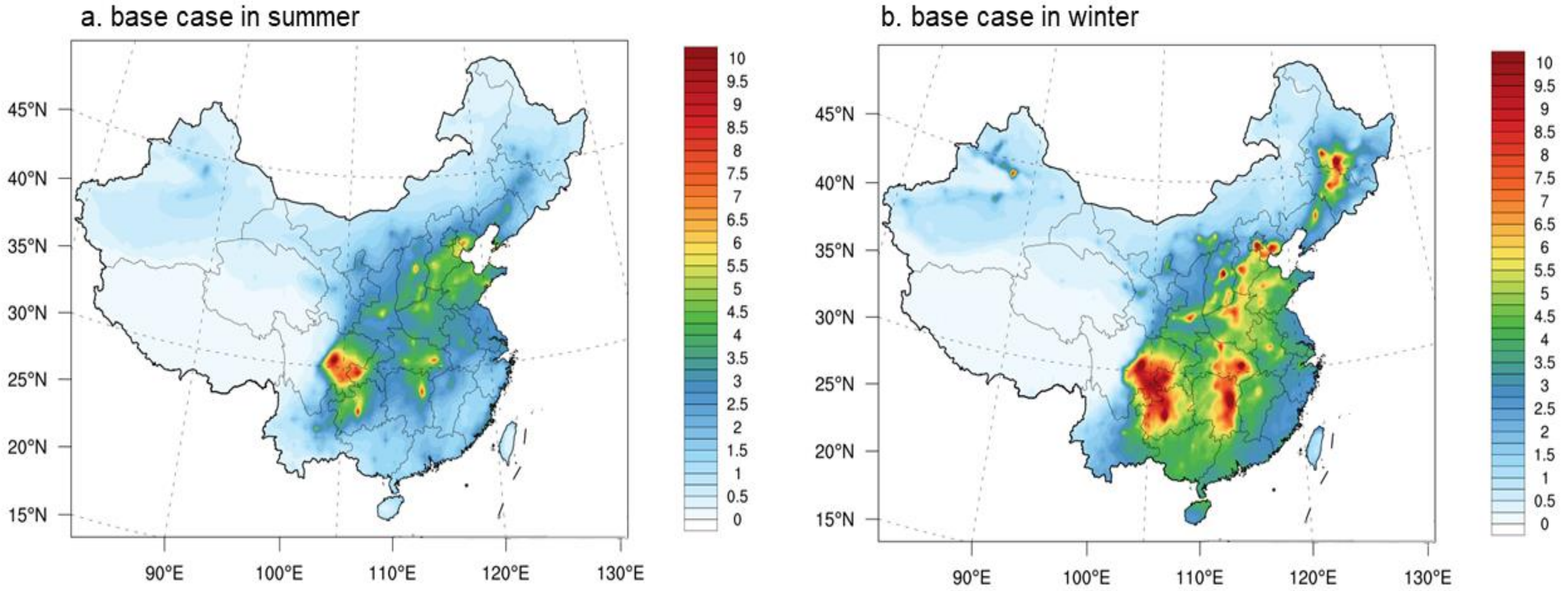
Simulated NO_3^-

Emission rate of NO_2

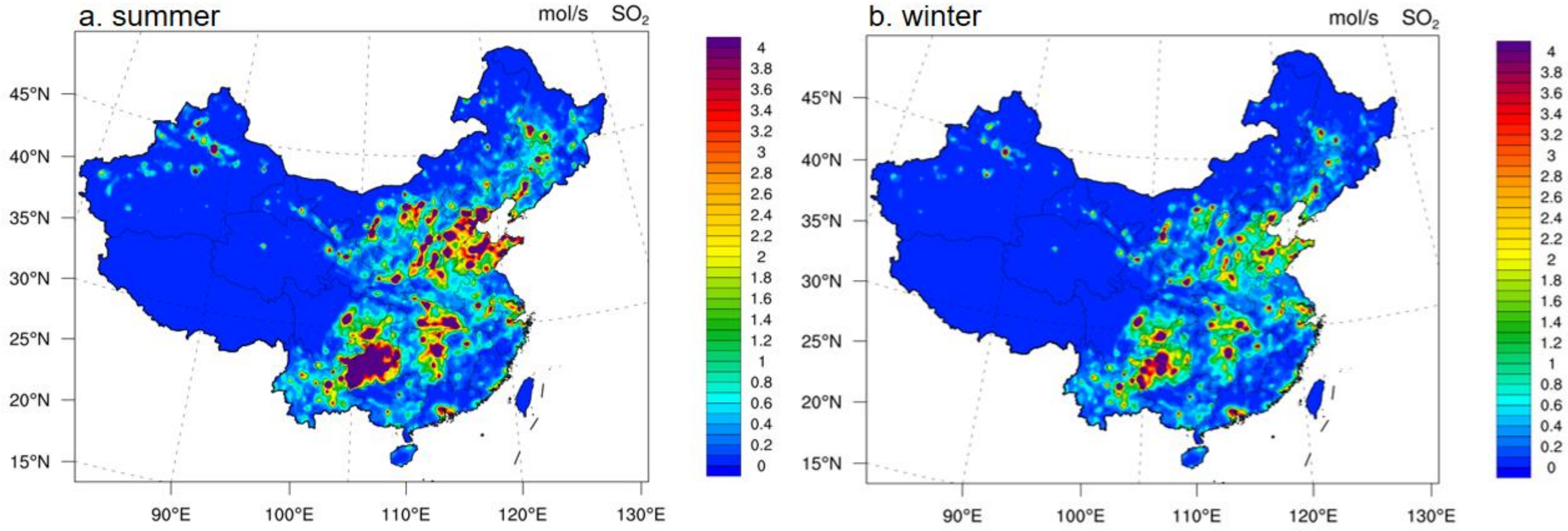


Spatial distribution of simulated surface SO_4^{2-} concentrations

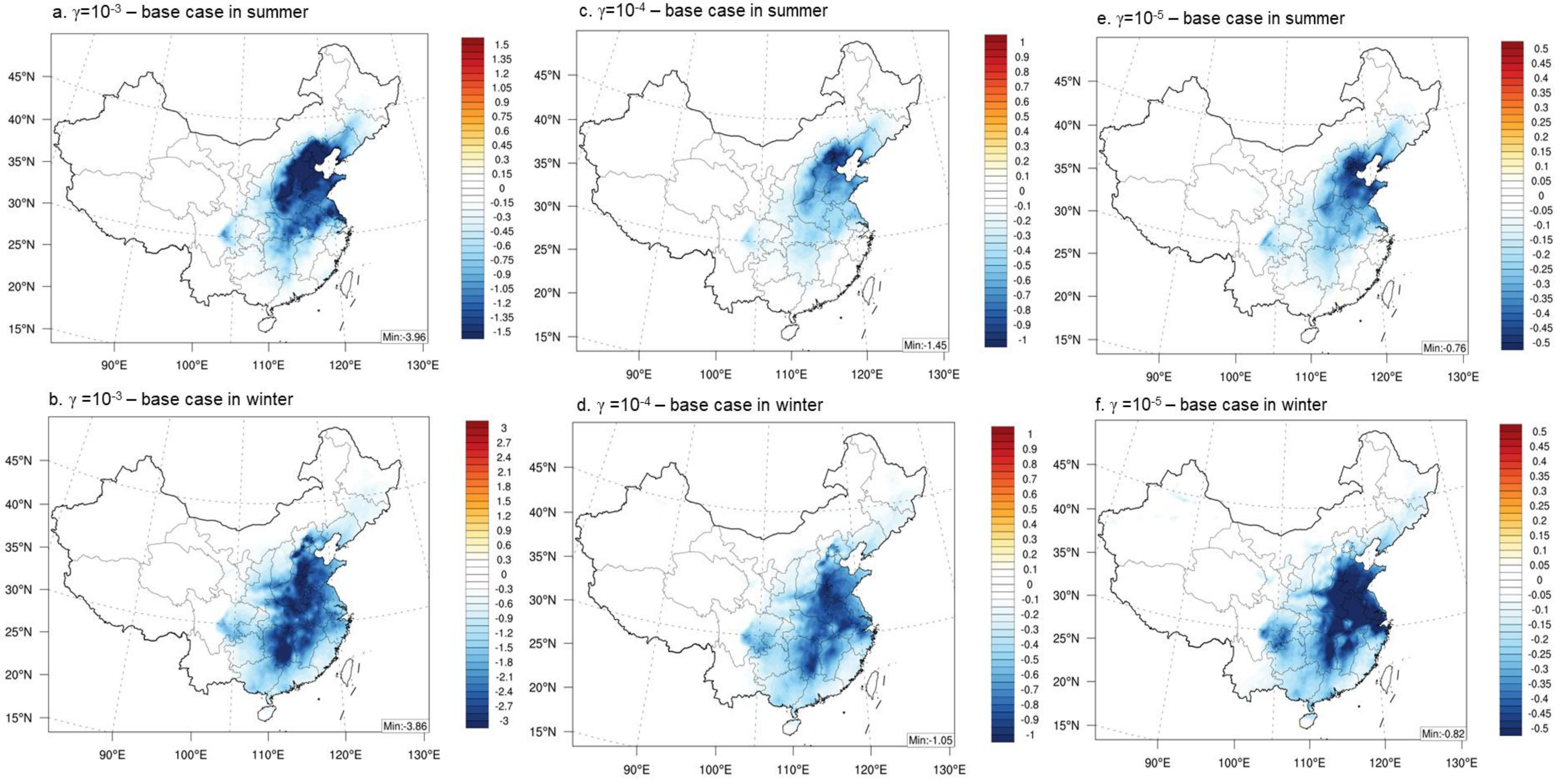
Simulated SO_4^{2-}



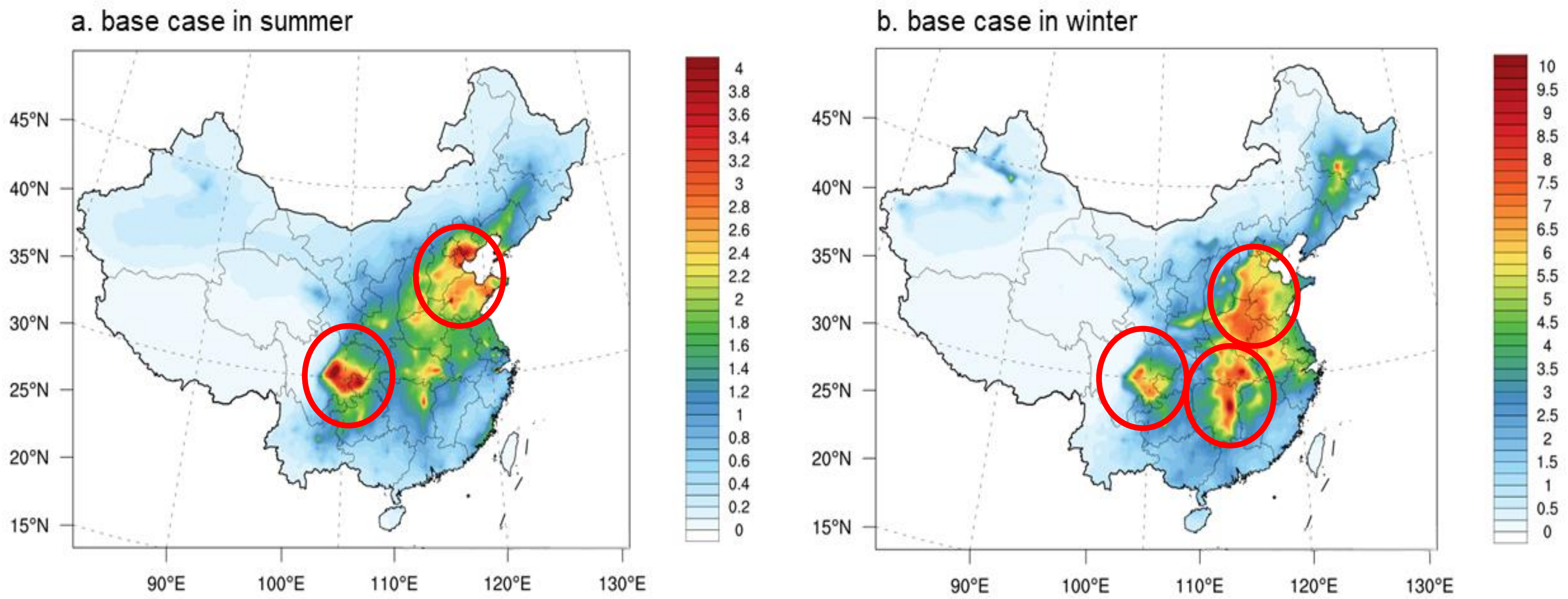
Emission rate of SO_2



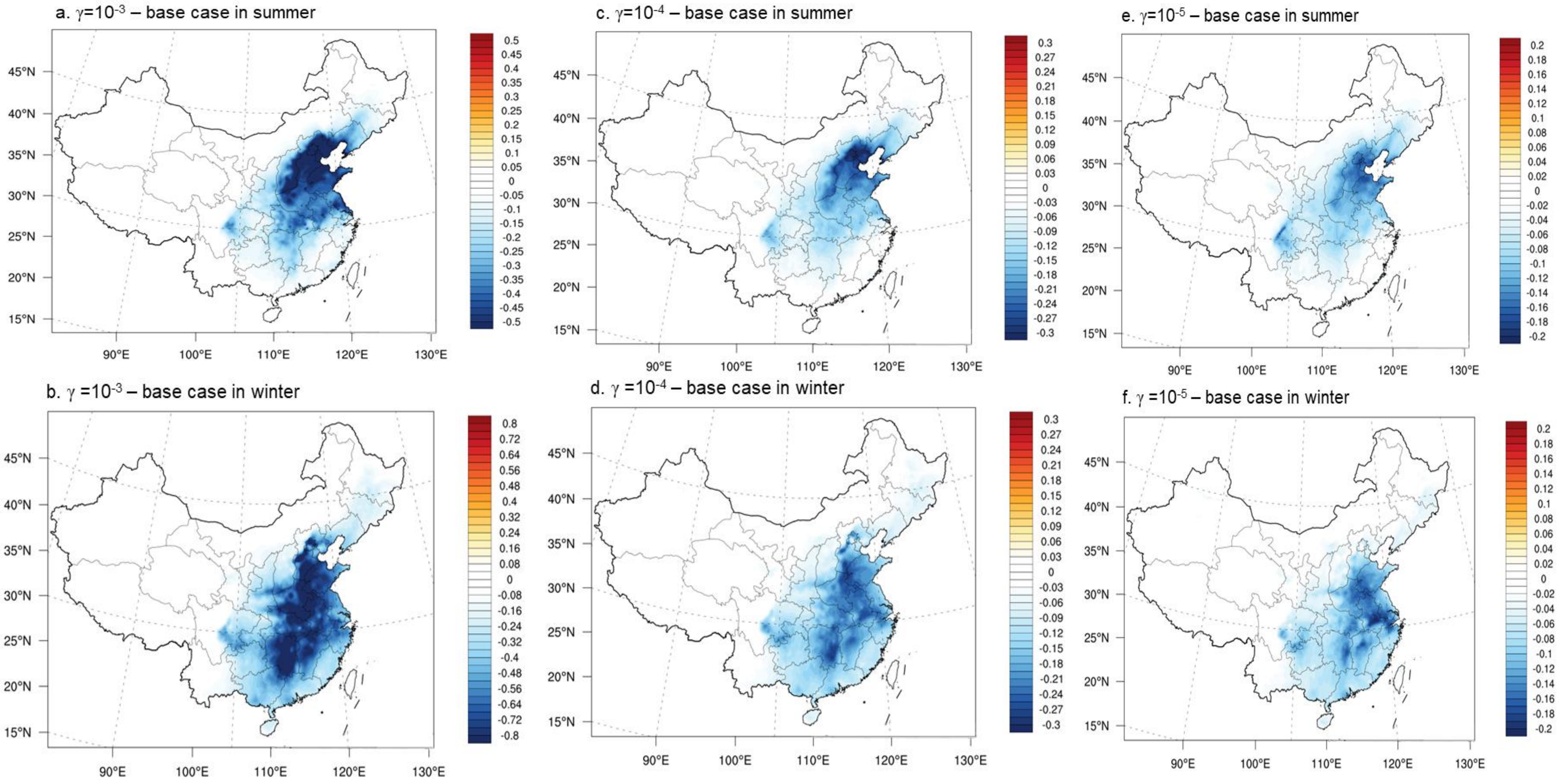
Spatial distribution of the difference in surface NO_3^- concentrations



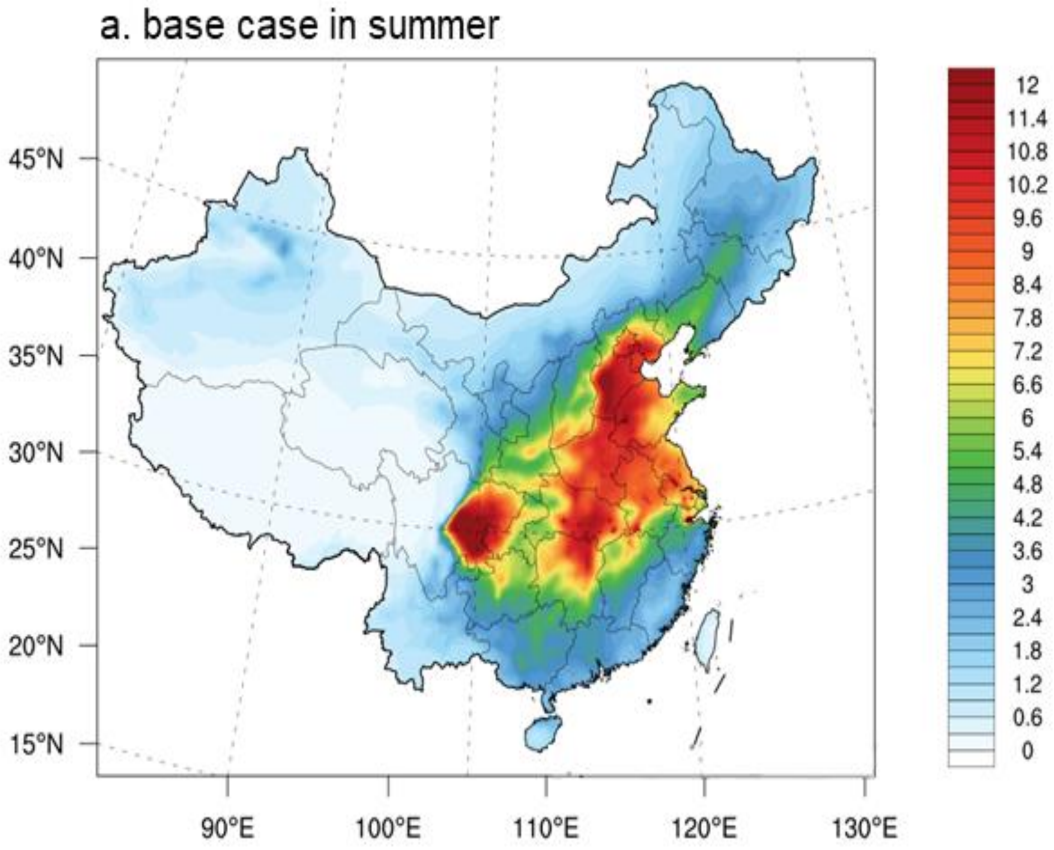
Spatial distribution of simulated surface NH_4^+ concentrations

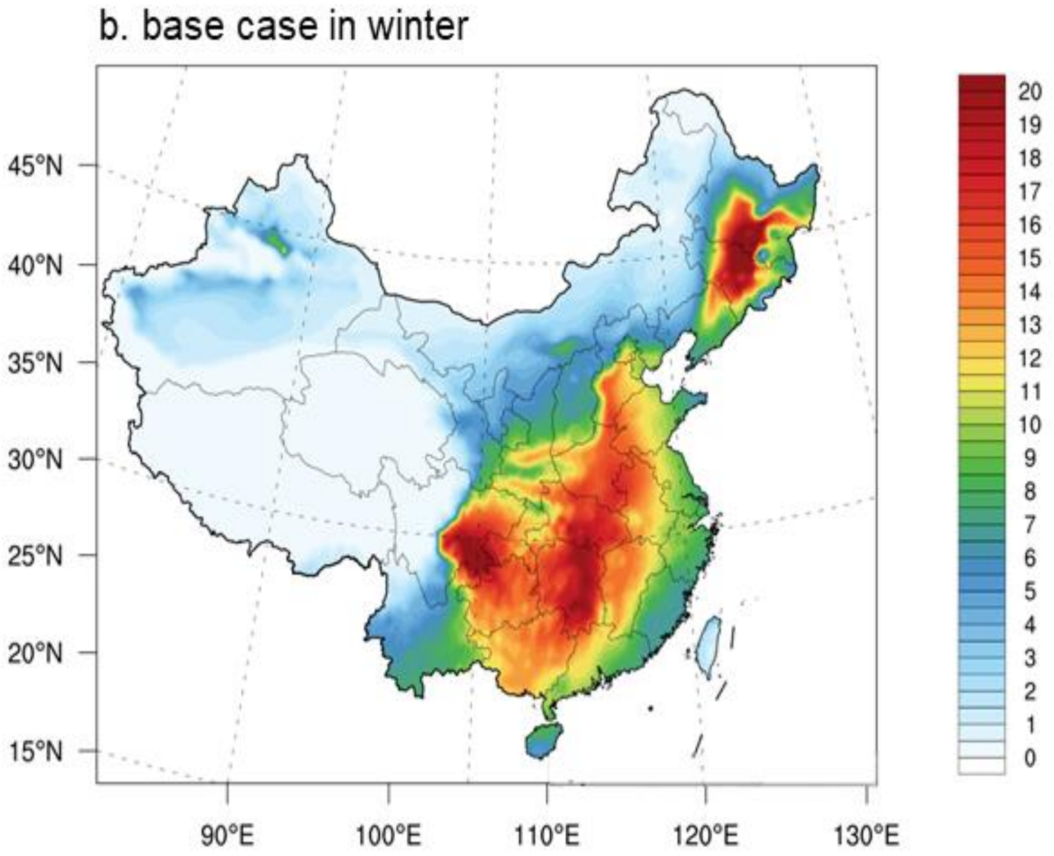


Spatial distribution of the difference in surface NH_4^+ concentrations

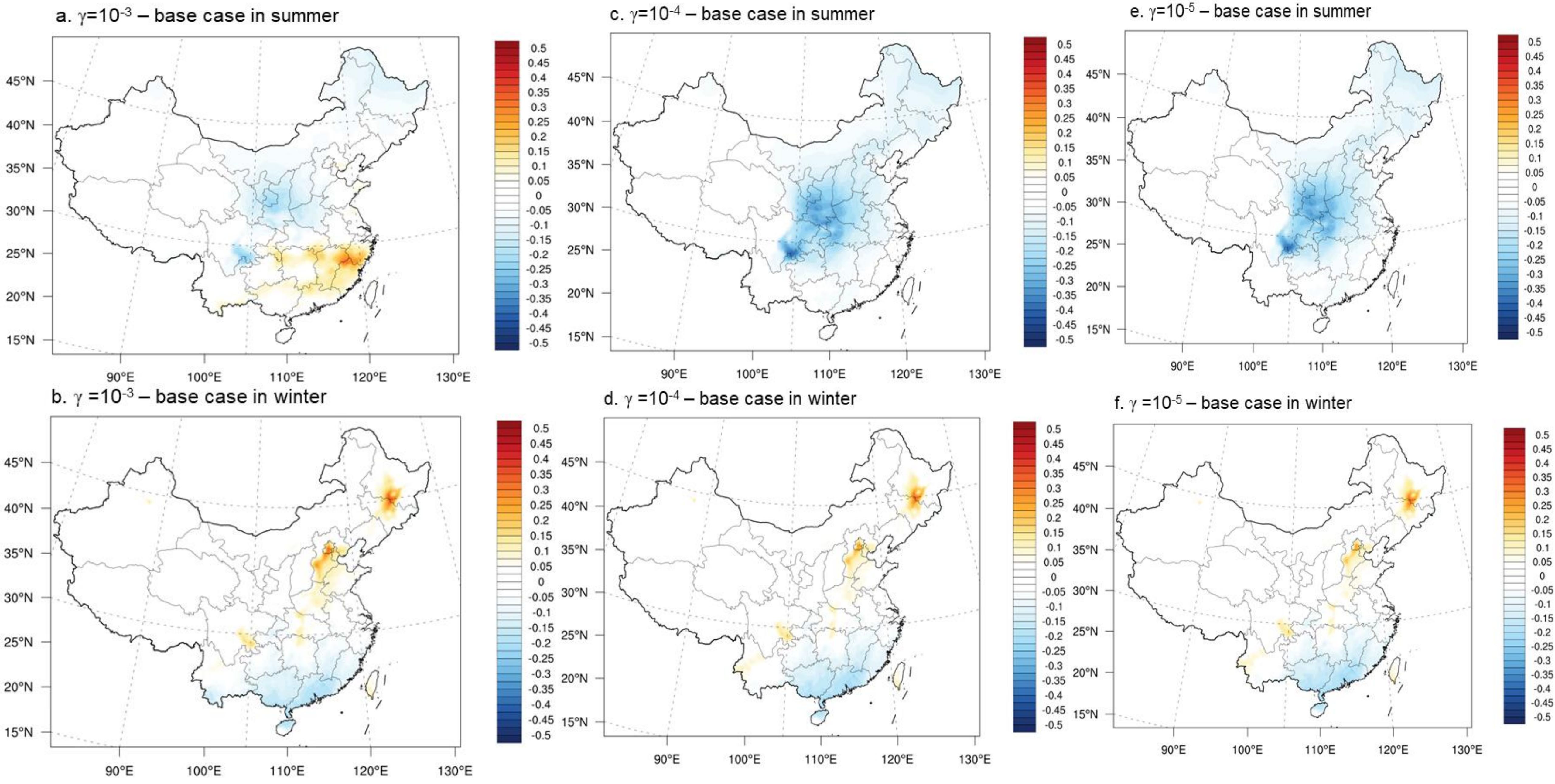


Spatial distribution of simulated surface SOA concentrations

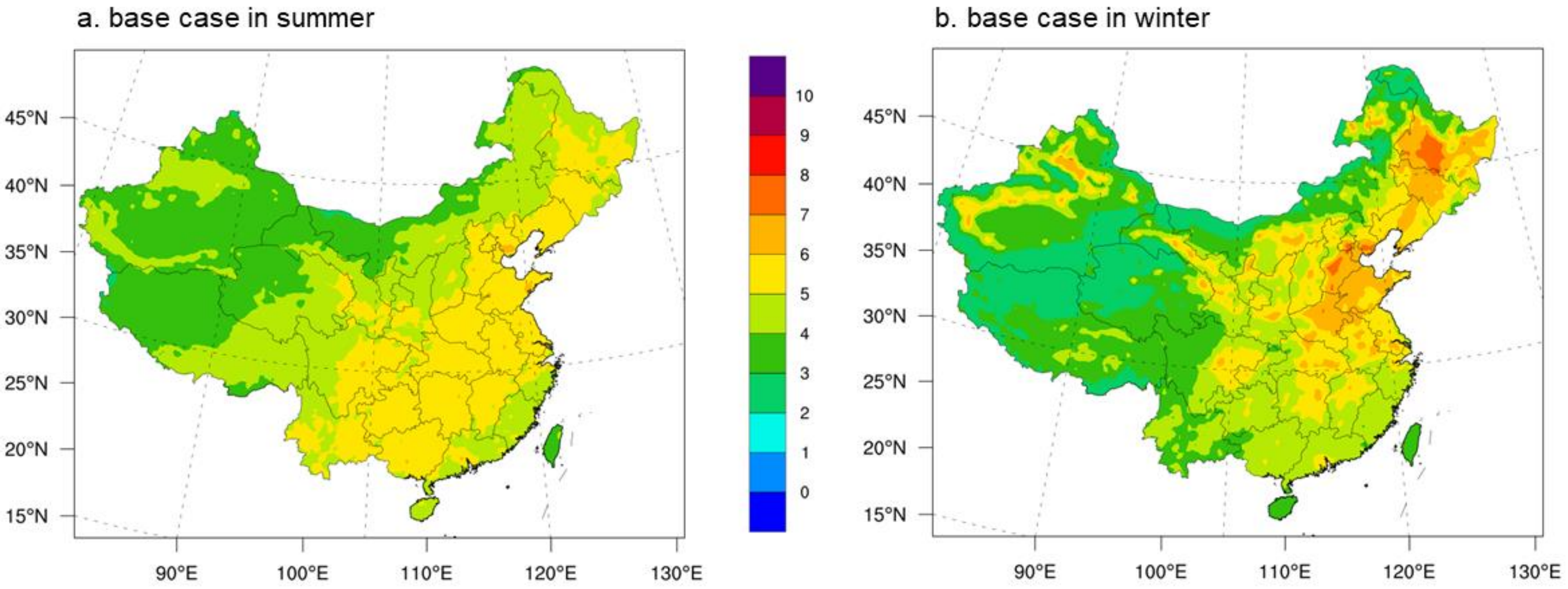




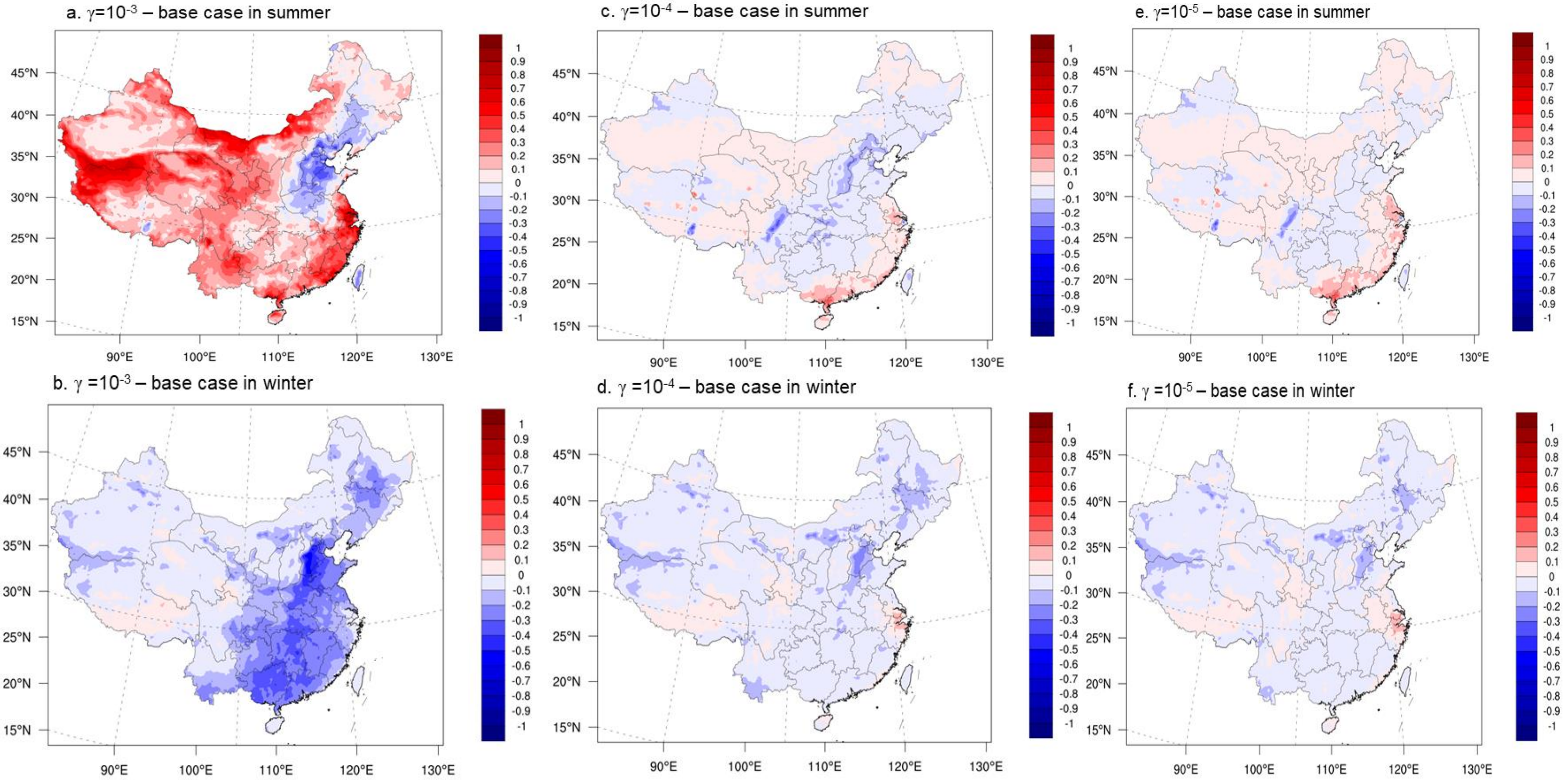
Spatial distribution of the difference in surface SOA concentrations



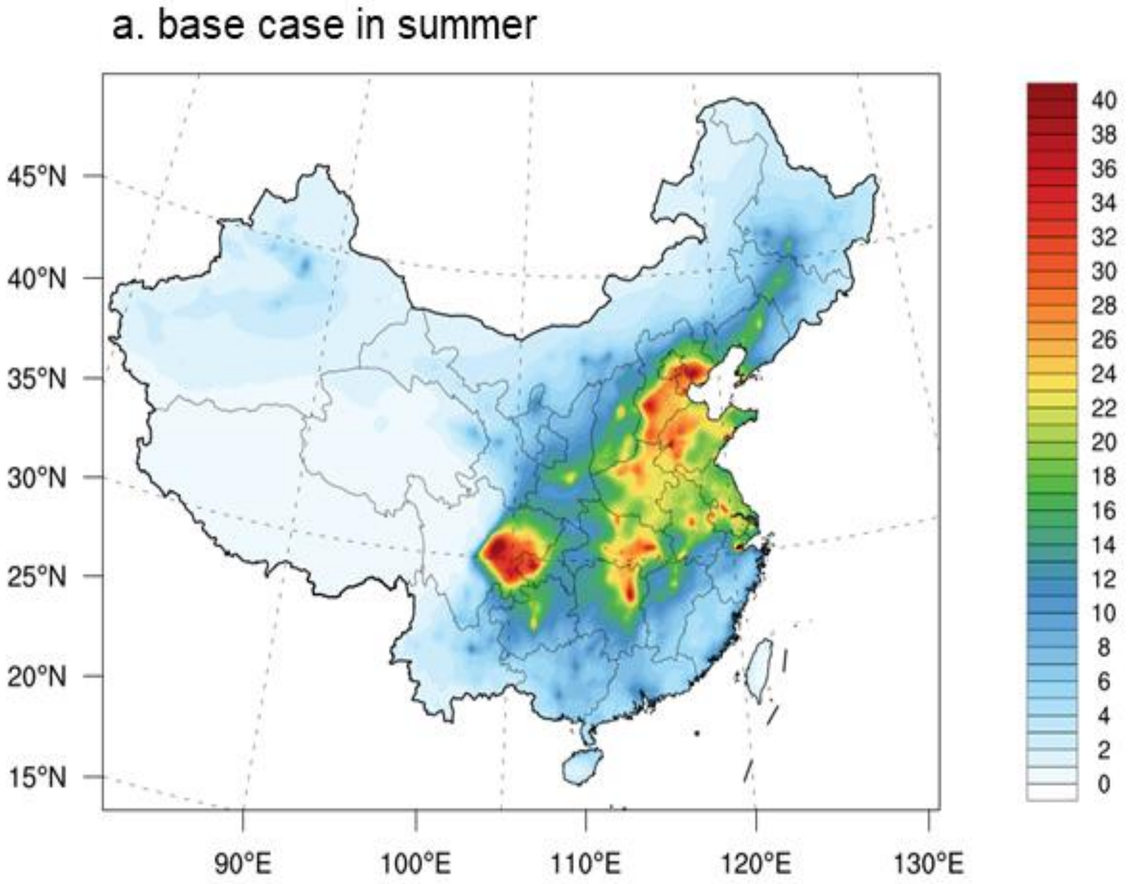
Spatial distribution of simulated aerosol pH

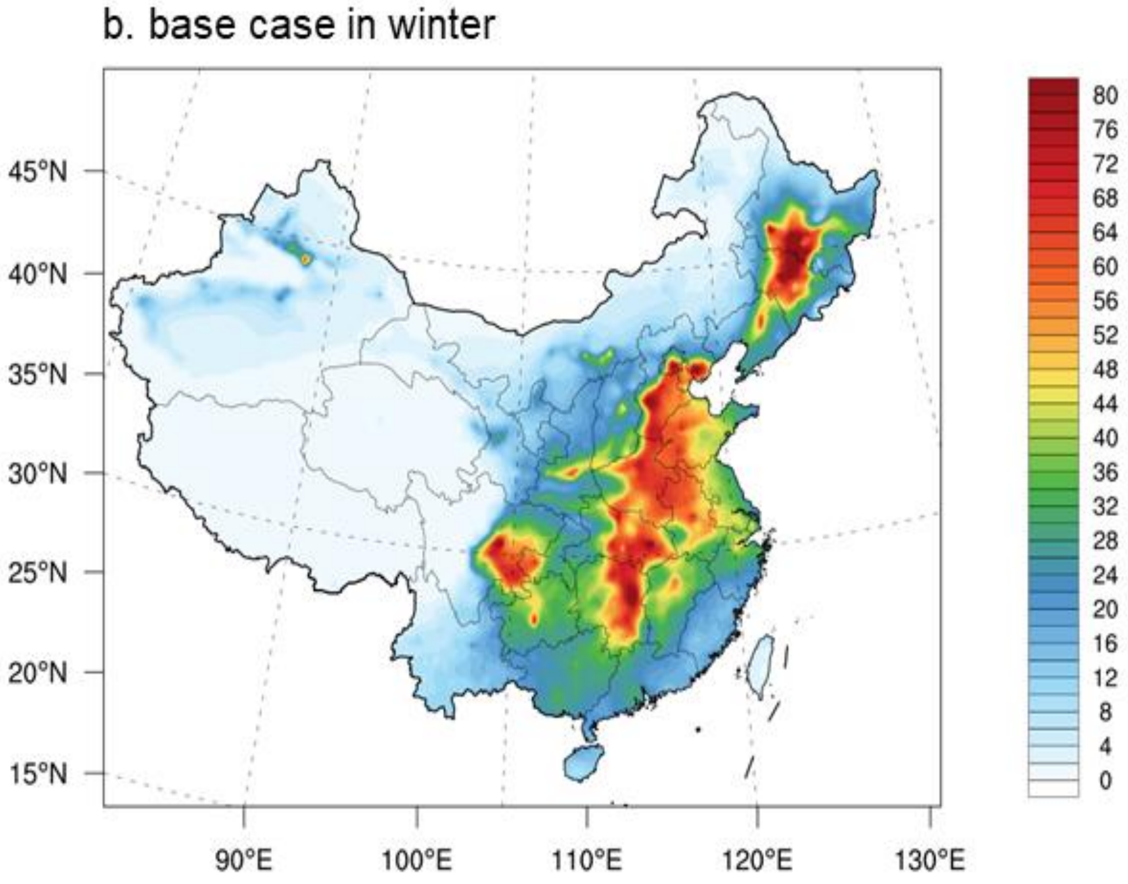


Spatial distribution of the difference in aerosol pH

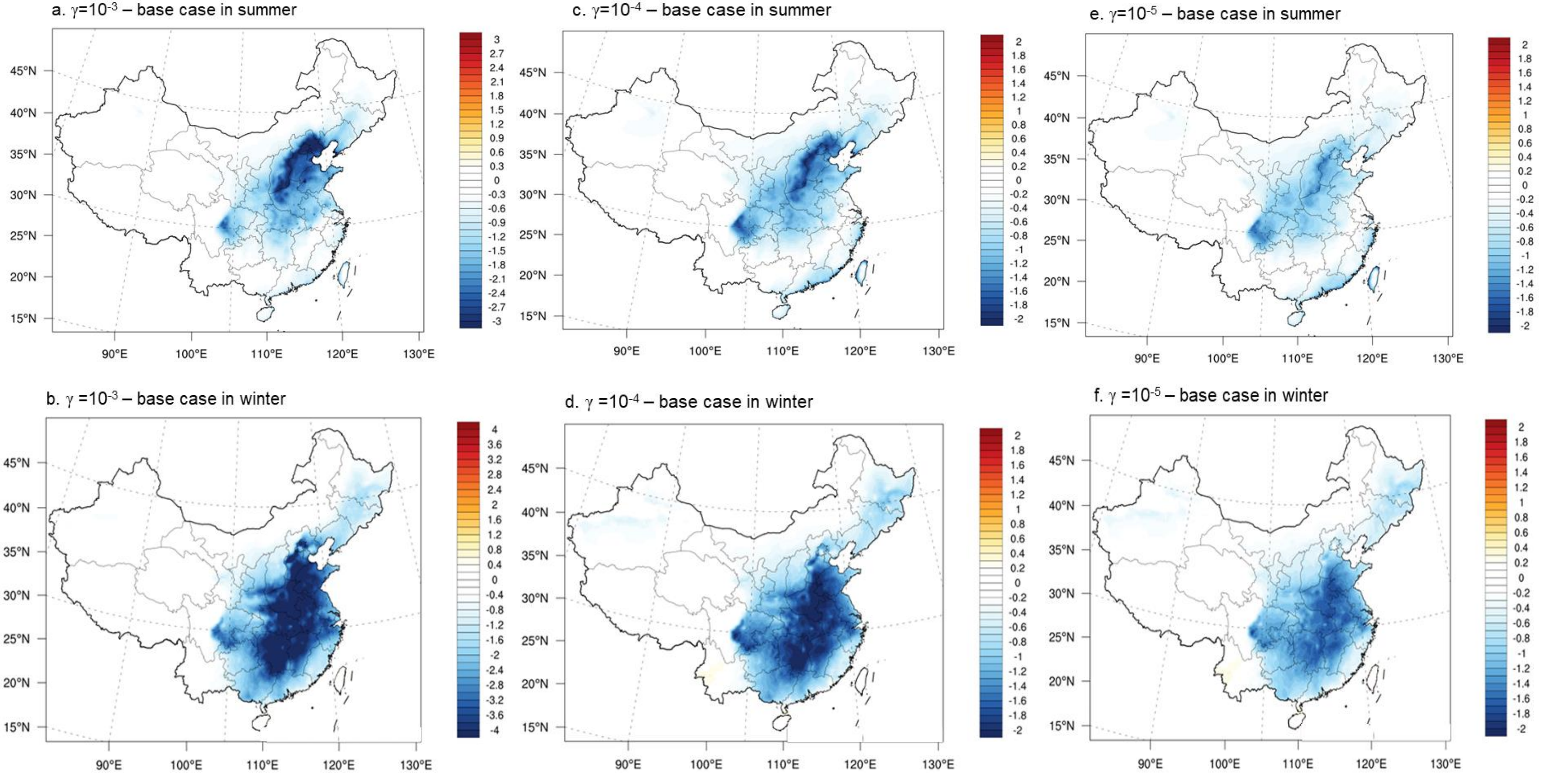


Spatial distribution of simulated $PM_{2.5}$ concentrations





Spatial distribution of the difference in $PM_{2.5}$ concentrations



Limitations And Potential Future Work For Moving Forward

- **The SOA is assumed to be well-mixed in this study then a shell of reacted SOA does not build up at the particle surface**

  → Investigating the possibility that a shell of reacted SOA could build up for particles with low viscosity under the condition of low relative humidity

- **The assumption used in this study that the aerosol had a single phase for both organic and inorganic constituents**

  → Develop phase separation mechanisms for improving the assumption

- **The real uptake coefficient of each SOA species may differ from the uptake coefficient used in this study**

  → Need further laboratory experiments for better representing the NH_3 uptake process by SOA

Thank you for listening!

Great appreciate the helpful comments from Professor Kleeman (UC Davis) and Professor Ian (UC Davis)!