Emissions and Deposition of Atmospheric Reactive Nitrogen over China

Lin Zhang[1] (zhanglg@pku.edu.cn)

Yuanhong Zhao[1], Youfan Chen[1]

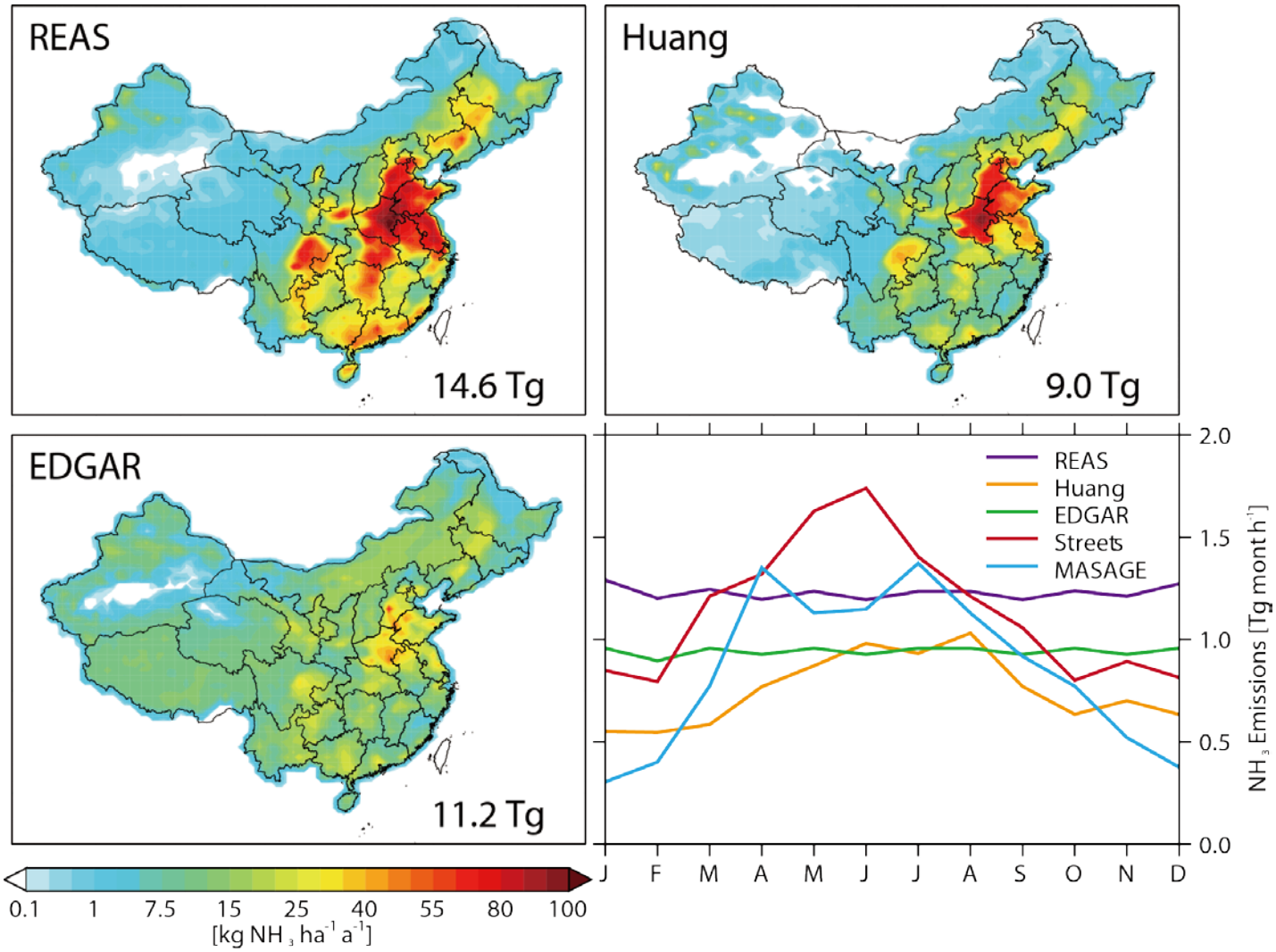
Collaborating with

Daven K. Henze[2], Fabien Paulot[3], Xuejun Liu[4],

Song Yu[5], Yuepeng Pan[6]

1. Department of Atmospheric & Oceanic Sciences, Peking University
2. University of Colorado, Boulder
3. Princeton University
4. China Agricultural University
5. Department of Environmental Science, Peking University
6. Institute of Atmospheric Physics, CAS

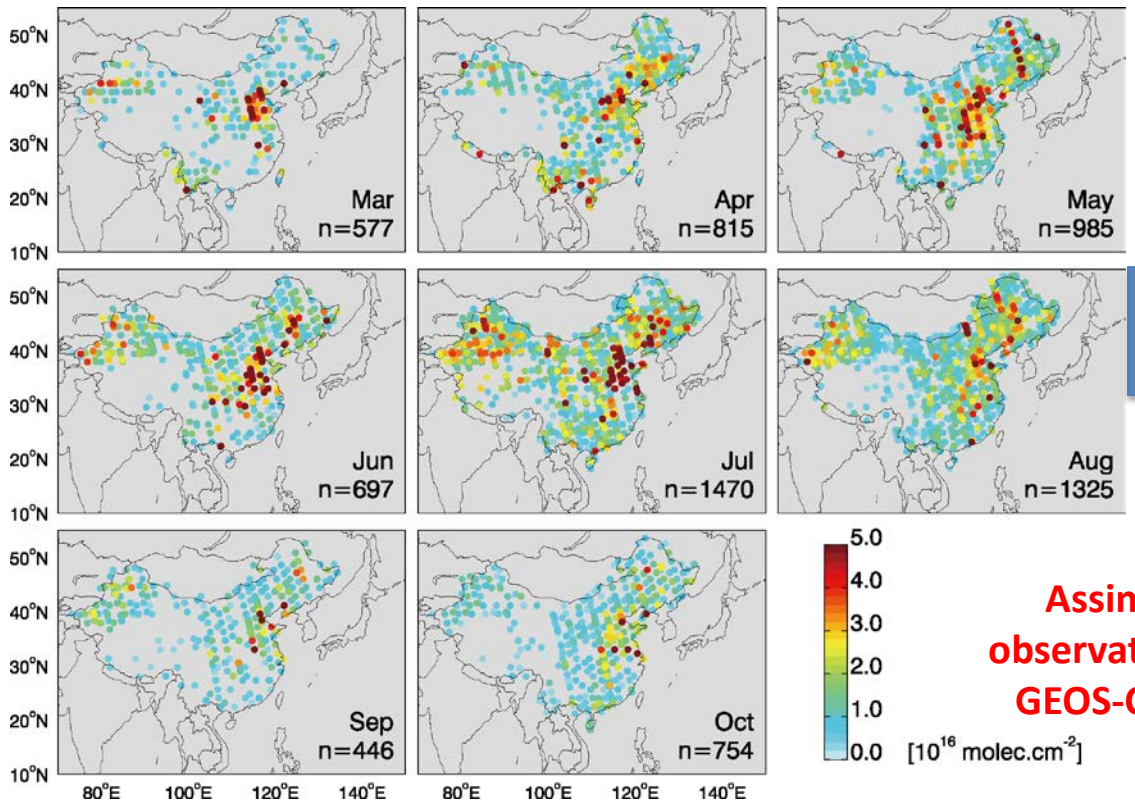
Substantial uncertainties in Chinese NH_3 emissions: spatial and seasonal variations



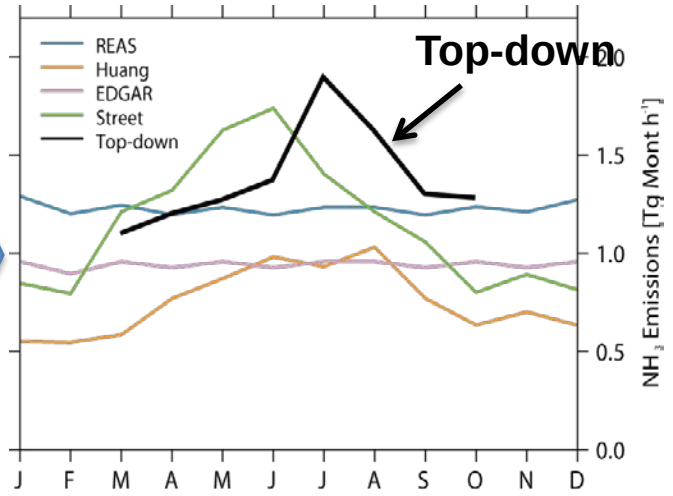
- **What can we learn from measurements?**
- **How can we improve the bottom-up estimate for consistency with measurements?**

TES satellite observed NH_3 columns as emission constraints

2005-2010 TES NH_3 observations



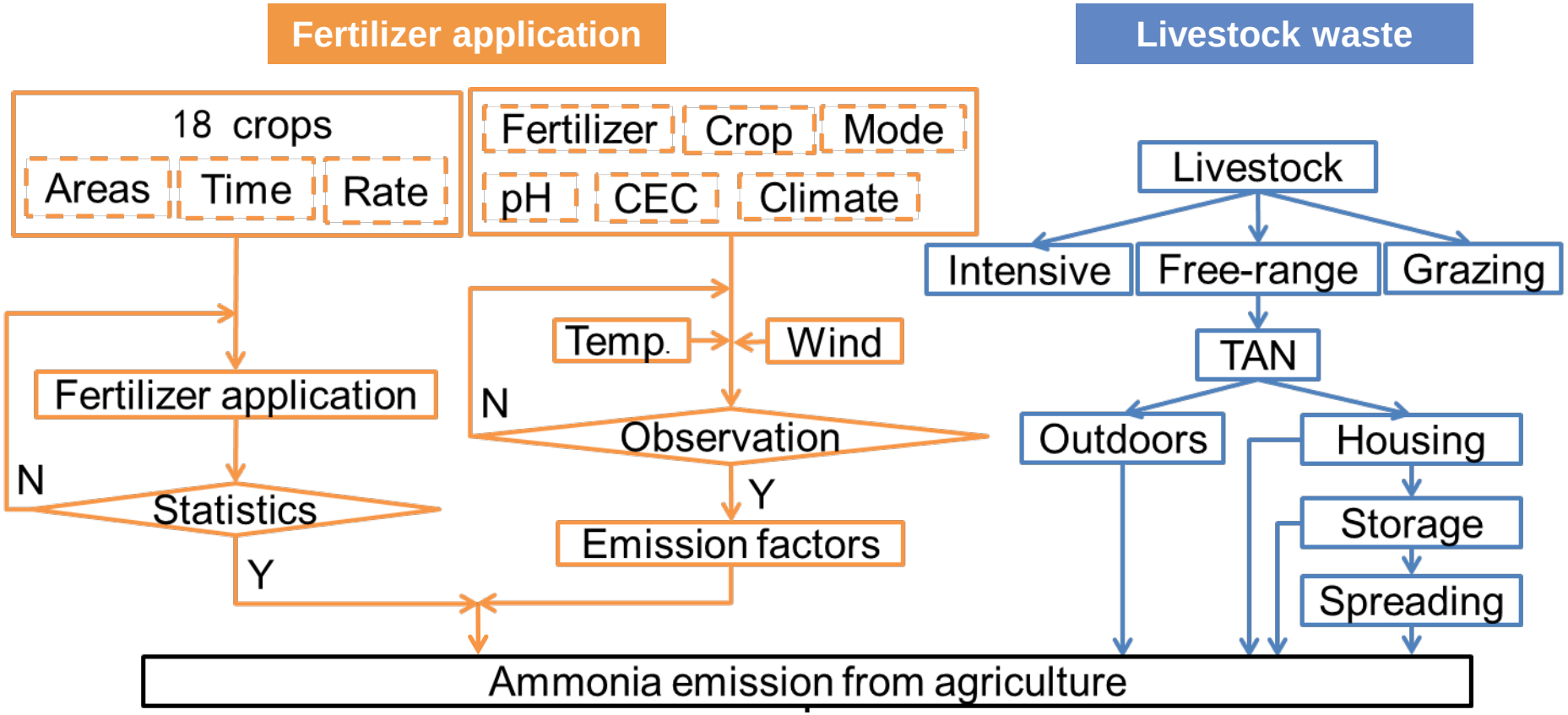
Top-down Chinese NH_3 emission seasonality



Assimilating TES observations using the GEOS-Chem adjoint

- The TES satellite instrument estimates atmospheric NH_3 concentrations from infrared spectral measurements.
- Current Chinese NH_3 emissions tend to underestimate their seasonal variations.

Revisit agricultural NH_3 emissions in China



- Better quantify how fertilizers are applied in China over 18 different agricultural crops (**more practical fertilizer application timing and magnitude**)
- Account for **influences of meteorological conditions** (near-surface temperature and wind) on the emission factors.

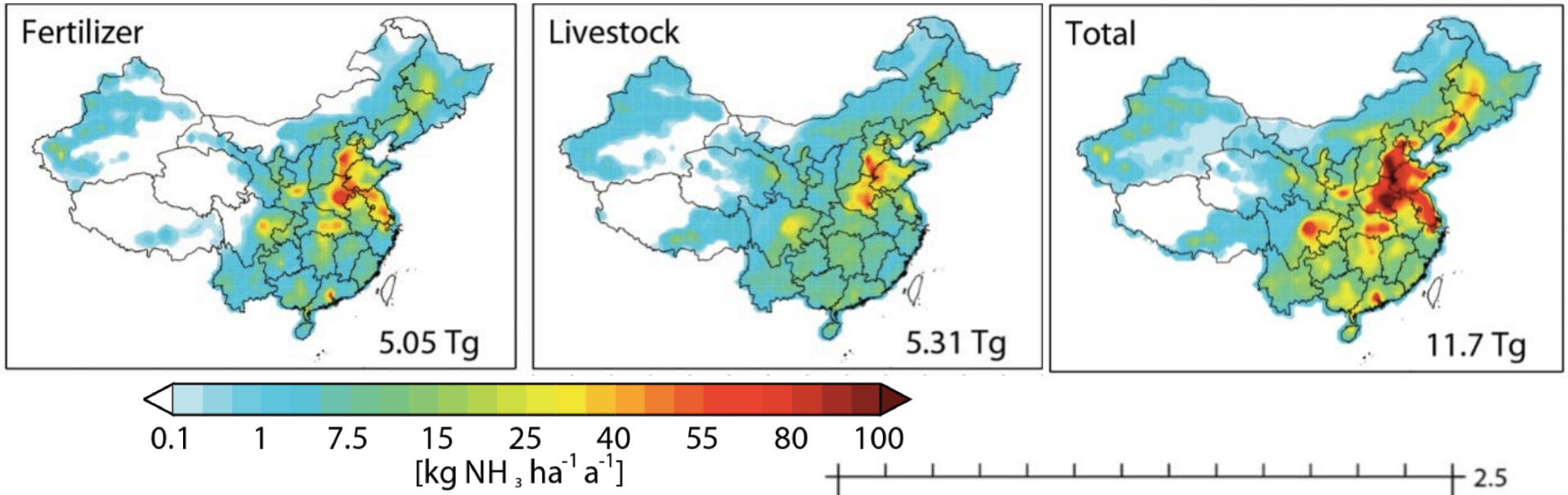
[Collaboration with F. Paulot (GFDL), Y. Song (PKU)]

An improved NH_3 emission inventory in China

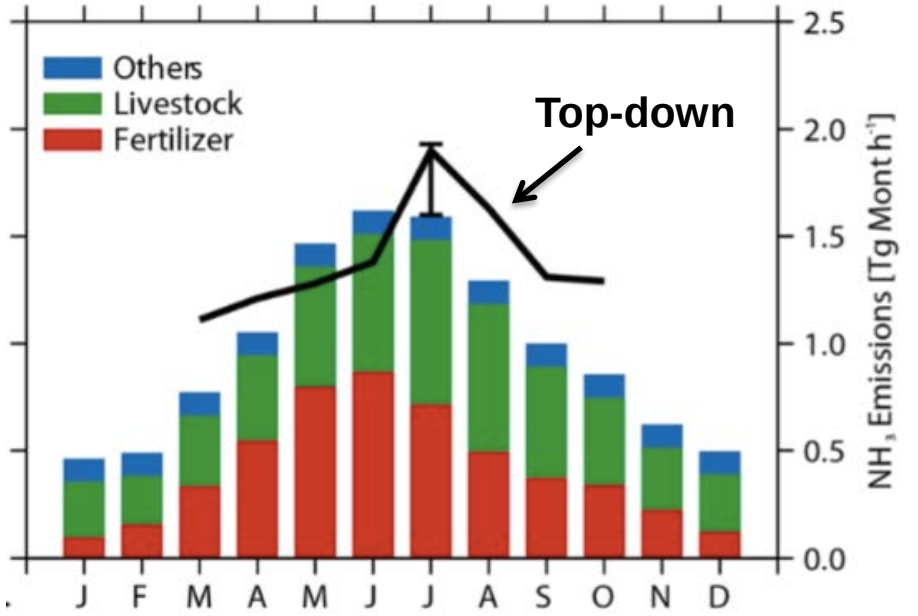
2008 NH_3 emissions from fertilizer application

Livestock waste

Anthropogenic total

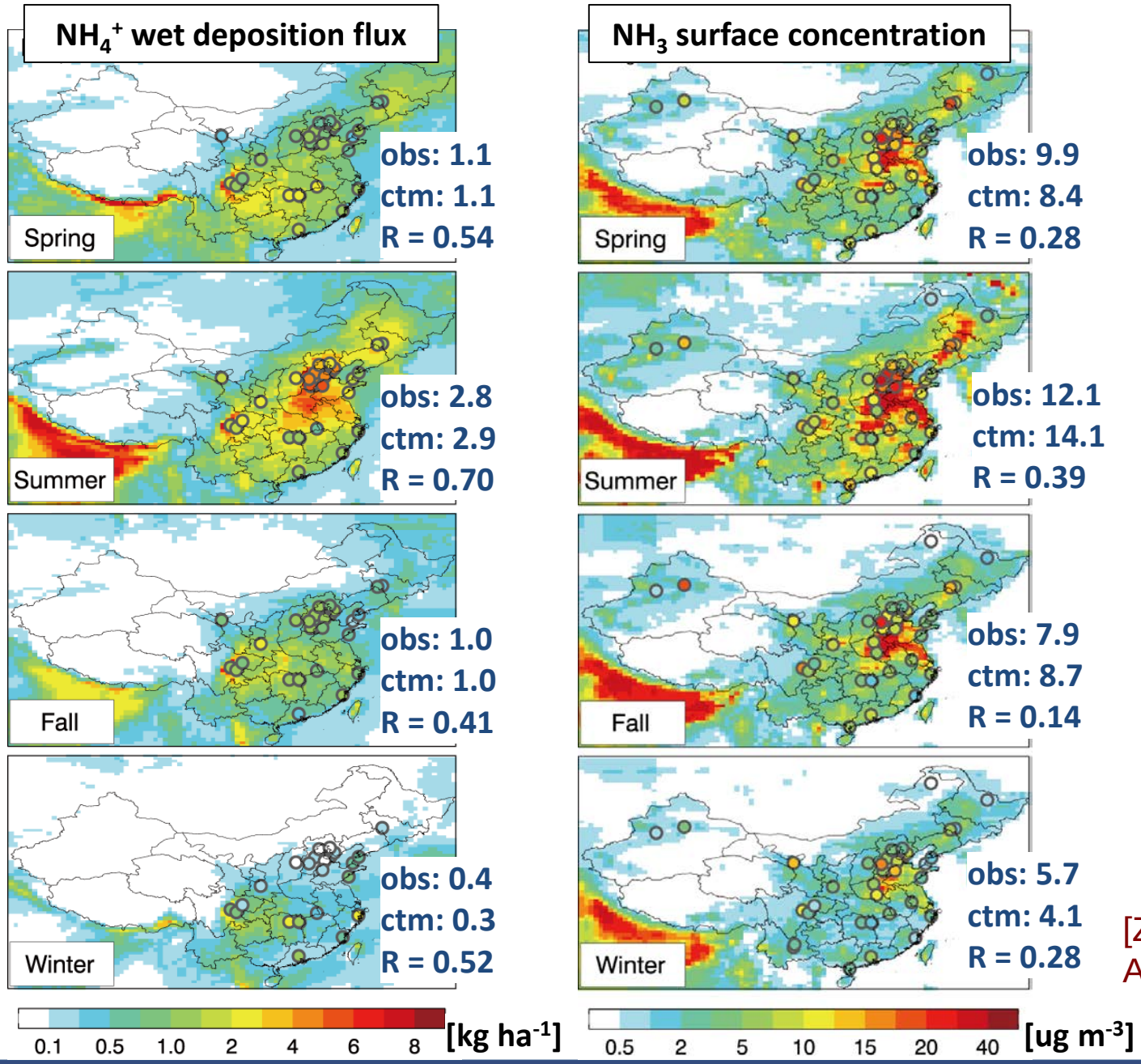


- 2008 NH_3 emissions from fertilizer application and livestock are **5.05** and **5.31 Tg**, respectively, and **11.7 Tg in total.**
- Both bottom-up and top-down show the highest NH_3 emissions in summer.



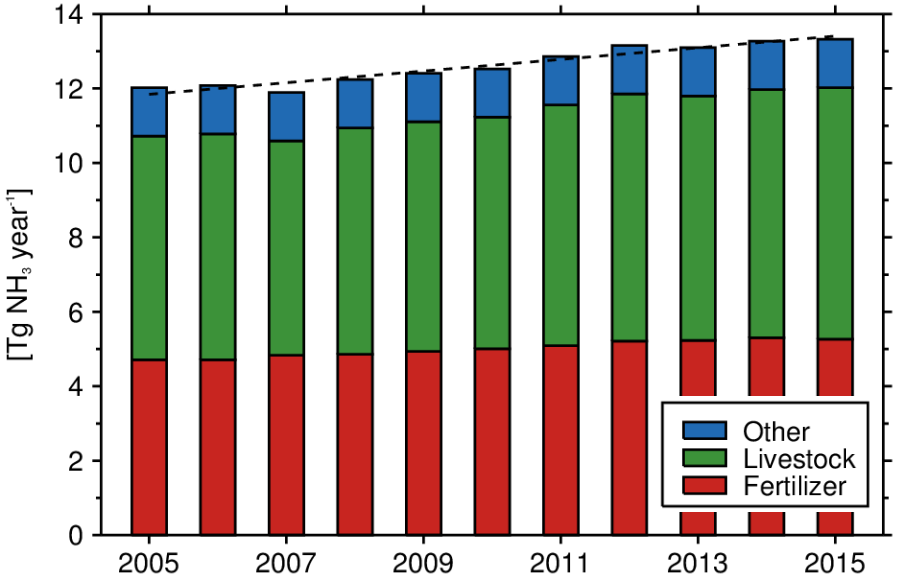
[Zhang et al. ACP, 18, 339–355, 2018]

Further support from surface measurements



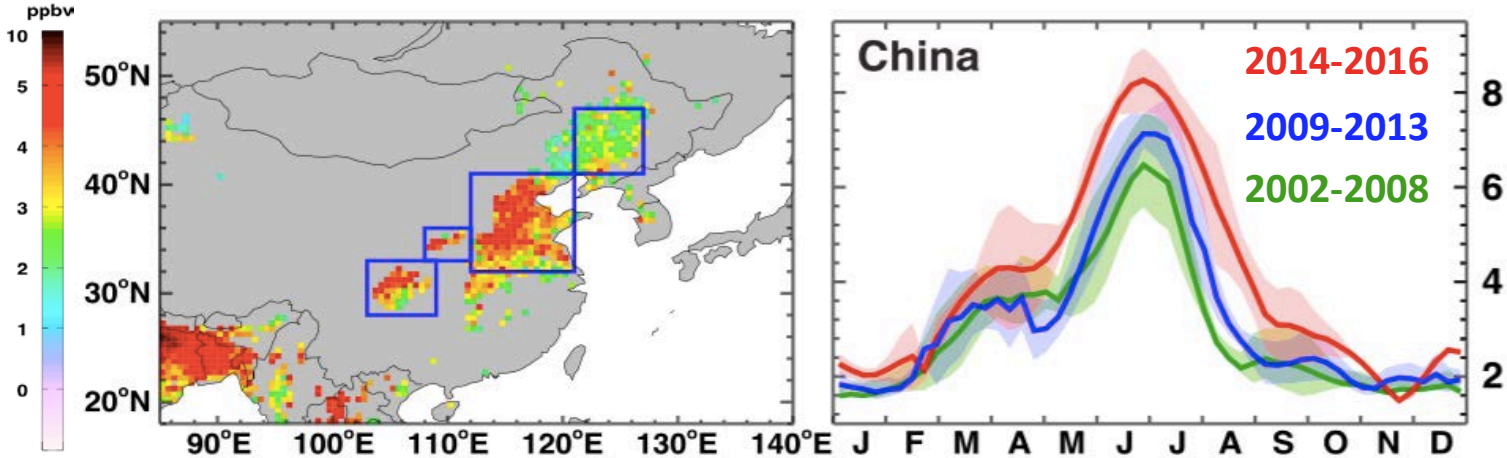
[Zhang et al. ACP 2018]

NH_3 emissions during 2005-2015 over China



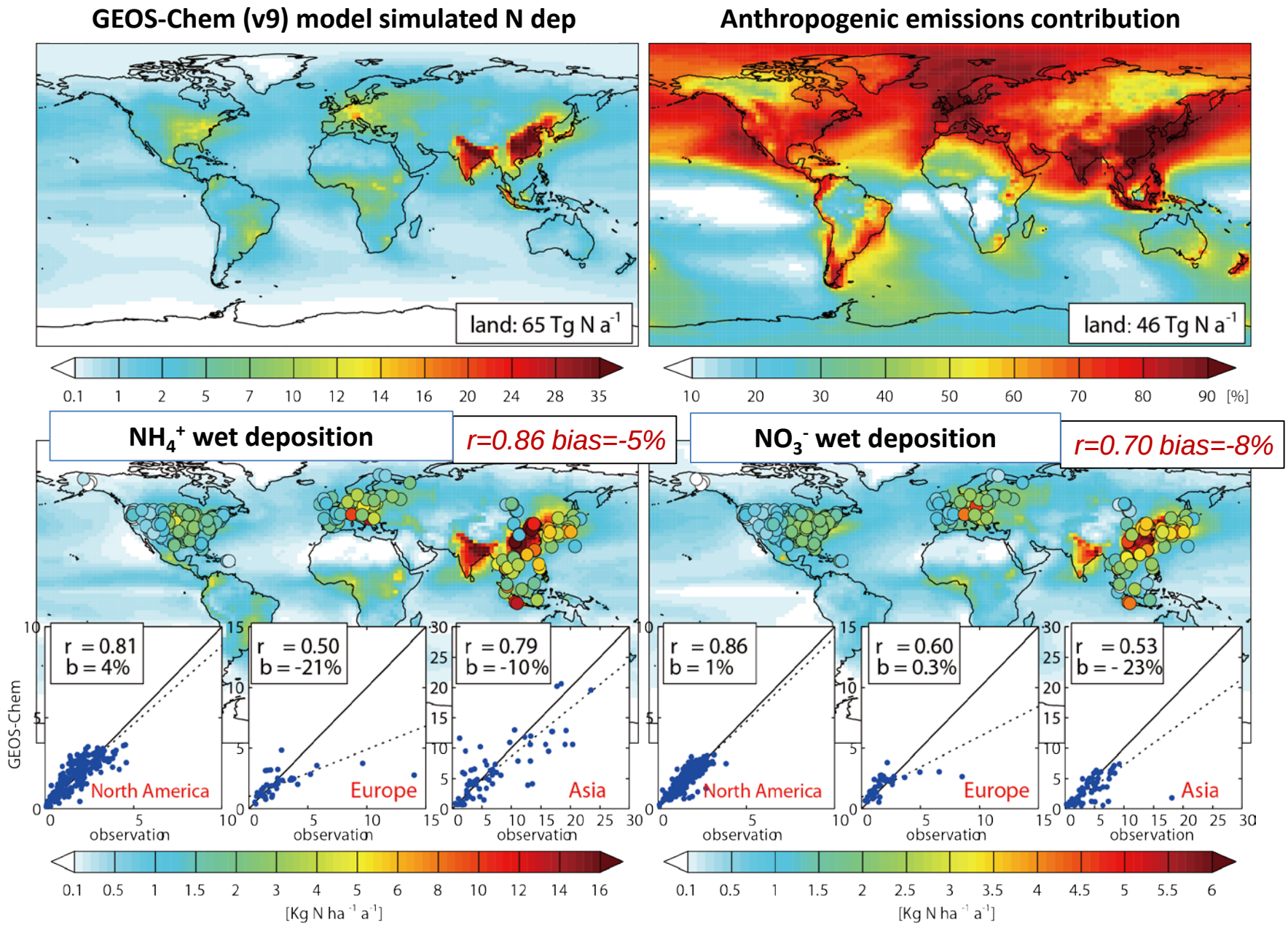
[Our work shows an increasing trend of 0.16 Tg (1.3%) year^{-1}]

Poster B5: Youfan Chen, Changes in ammonia agricultural emissions and their impact on surface PM2.5 pollution in China during 2005-2015

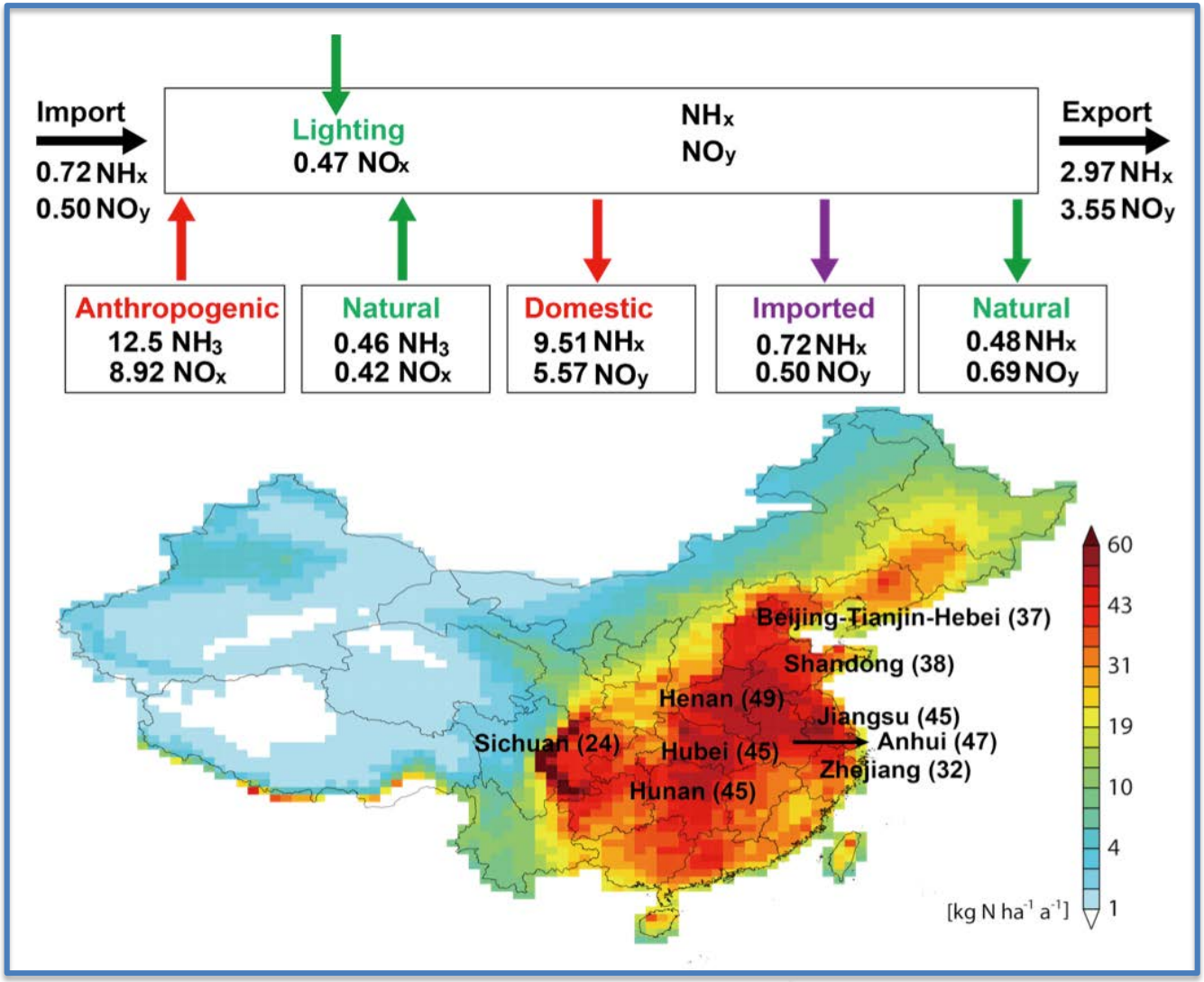


Warner et al. (2017) showed that AIRS NH_3 VMR increased at 2.27% year^{-1}.

Global nitrogen deposition fluxes and anthropogenic contributions



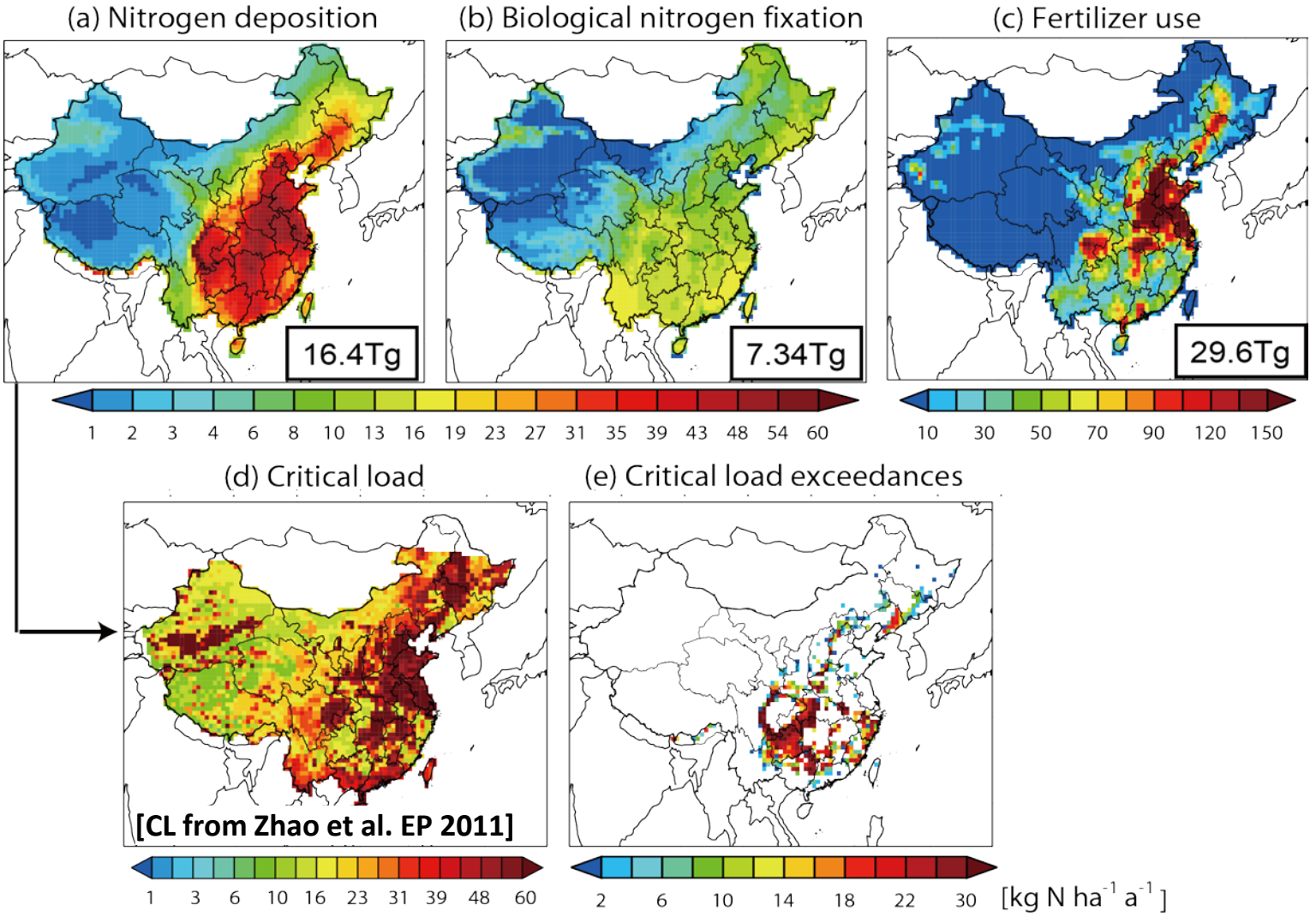
Budgets of atmospheric nitrogen deposition to China



- Annual deposition flux of 16.4 Tg N a^{-1} to China, NH_x contributing 62%;
- 24% of NH_3 emissions and 36% of NO_x emissions are exported.
- Zhang et al. (2012) estimated nitrogen deposition to the US of 6.5 Tg N a^{-1} with NH_x contributing 35%.

[Zhao, Zhang*, AE et al. 2017]

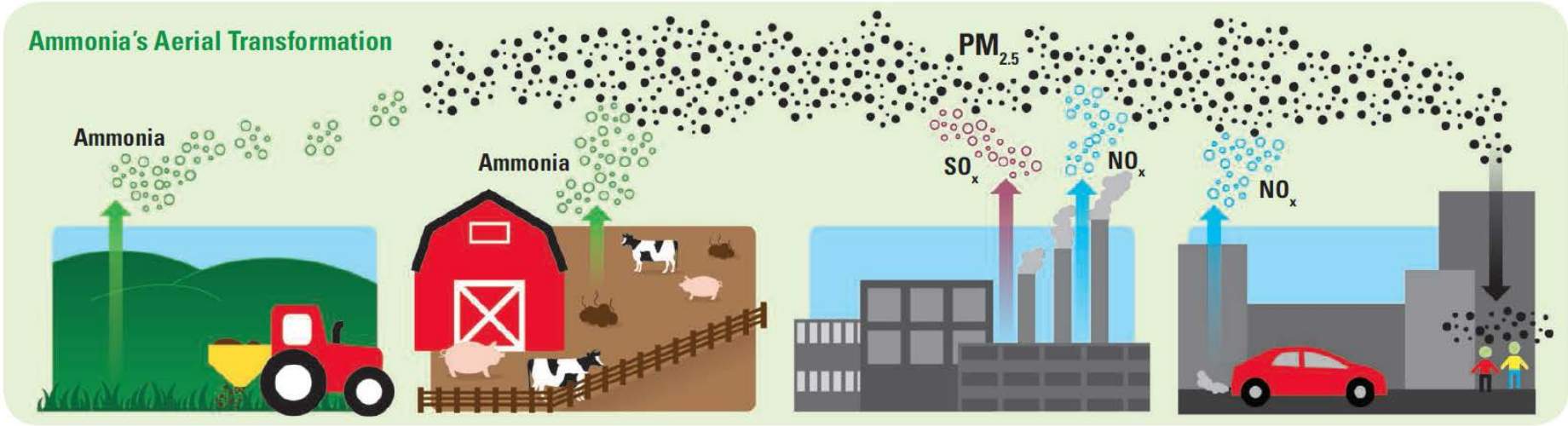
Terrestrial nitrogen input and critical load exceedance



- Nitrogen deposition is about half of fertilizer nitrogen input, and is much higher than that from natural biological fixation over China.
- About 15% of the land over China experiences eutrophication critical load exceedances.

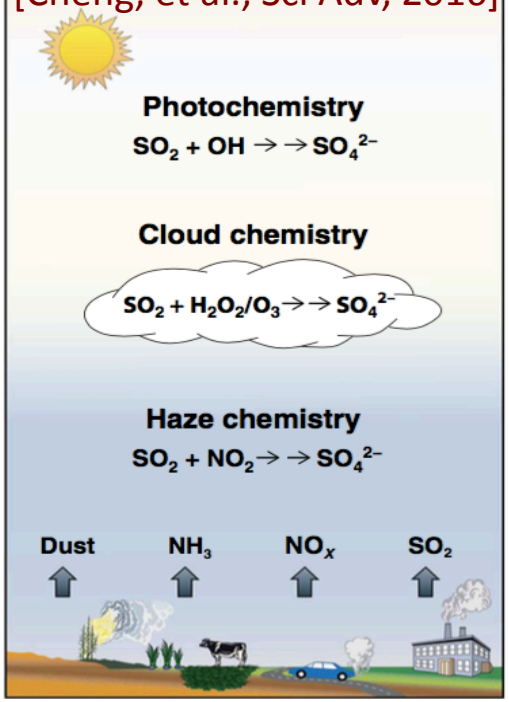
Thank you for your attention!

The role of ammonia (NH_3) on air pollution

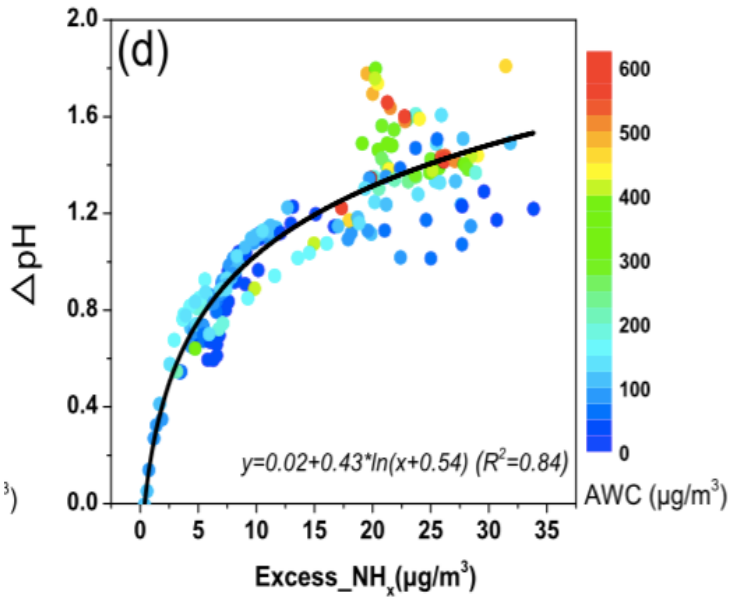


[Erik Stokstad, Science,2014]

[Cheng, et al., Sci Adv, 2016]



[Liu, Song, et al., GRL, 2017]



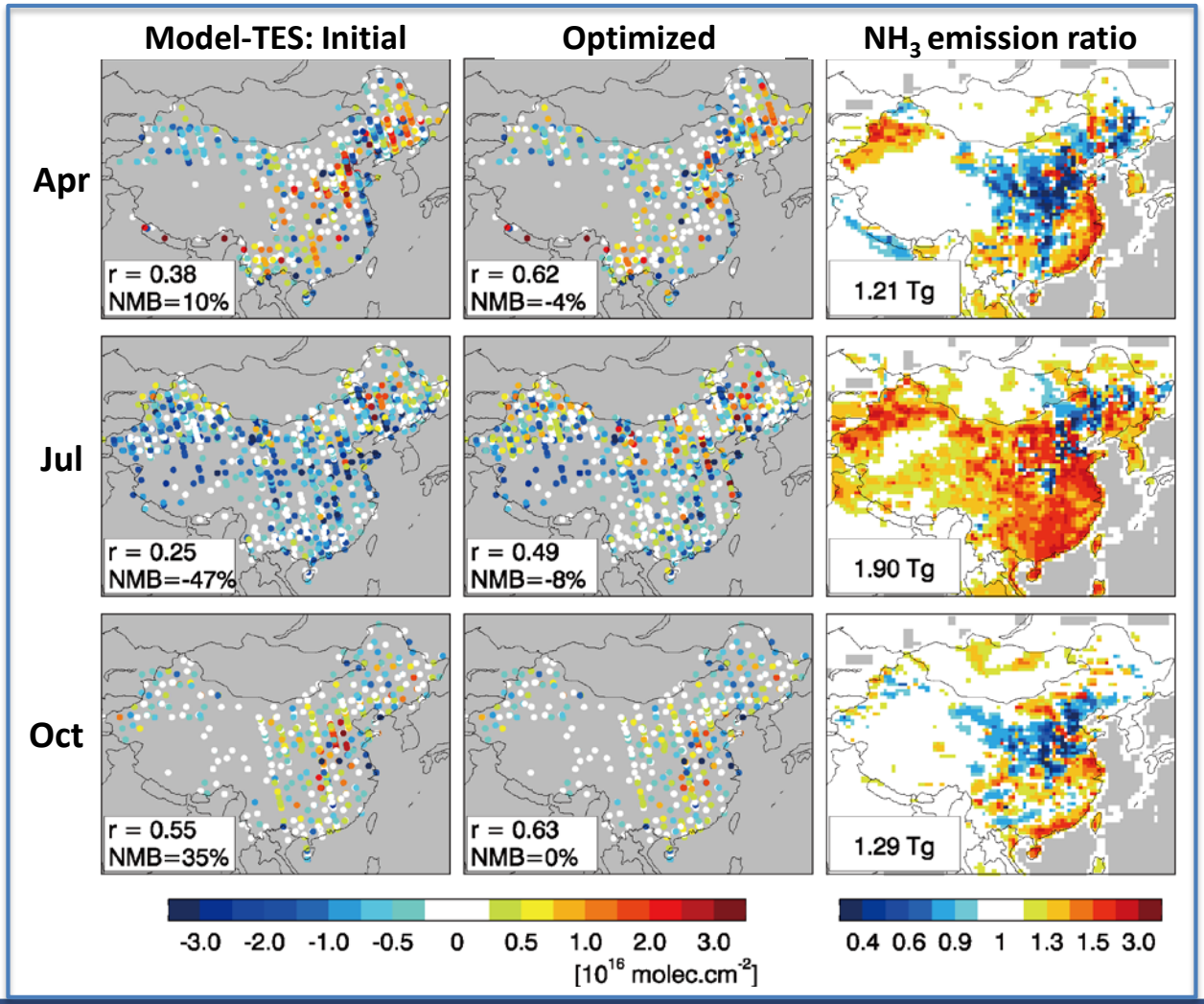
- As the main alkaline gas in the atmosphere, neutralizing H_2SO_4 and HNO_3 to form secondary inorganic aerosols;
- Affect aerosol surface pH that may change enhance formation of sulfate formation.

NH_3 columns: observations vs. prior and optimized emissions

Model observations emissions a priori emissions

$$J(\mathbf{x}) = (\mathbf{F}(\mathbf{x}) - \mathbf{y})^T \mathbf{S_e}^{-1} (\mathbf{F}(\mathbf{x}) - \mathbf{y}) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S_a}^{-1} (\mathbf{x} - \mathbf{x}_a)$$

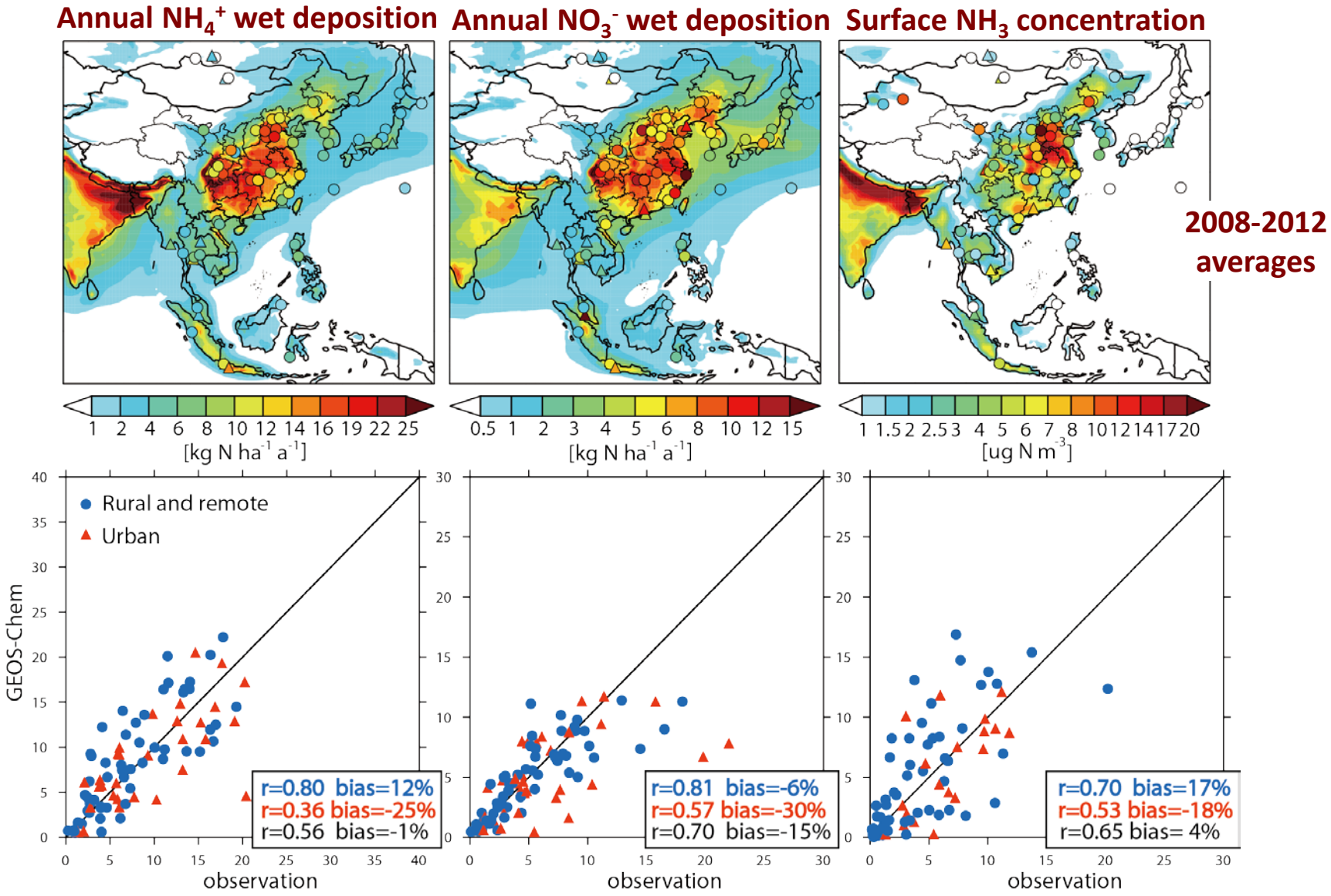
observational error a priori error



- The source inversions largely improve comparisons of model results with TES observations;
- Current Chinese NH_3 emissions tend to underestimate their seasonal variations.

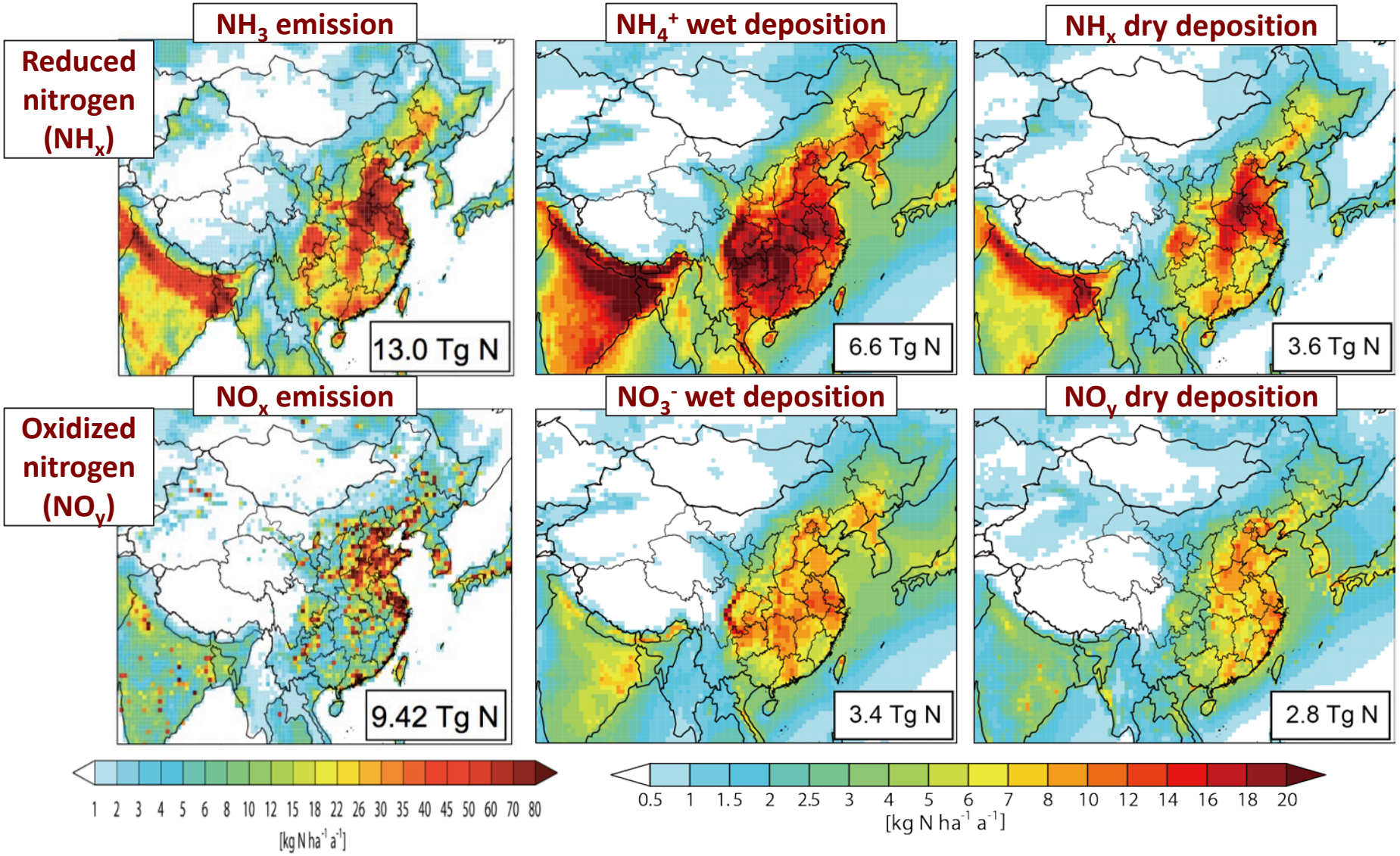
[Zhang et al. to be submitted]

Measured and GEOS-Chem simulated surface nitrogen fluxes and concentrations



[Zhao, Zhang*, et al. 2017]

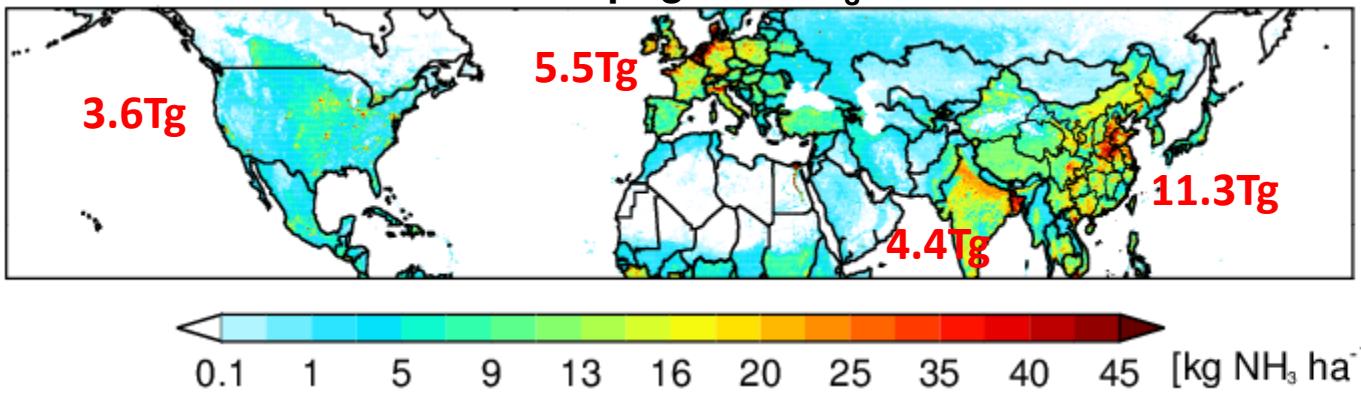
Atmospheric nitrogen deposition to China: emissions and processes



Annual total deposition flux to China **16.4 Tg N**: **wet (61%), dry (39%)**
NH_x (62%), NO_y (38%)

Substantial uncertainty in Chinese NH_3 emission estimates

EDGAR anthropogenic NH_3 emissions



References	Fertilizer	Livestock	Other	Total
Kang et al.(2016)	2.8	4.99	1.83	9.62
Zhao et al.(2013)	9.82	7.36	1.12	18.3
Paulot et al.(2014)	3.6	5.8	0.8	10.2
EDGAR(2013)	8.1	3.1	0.1	11.3
Xu et al.(2016)	3.3	3.8	1.3	8.4
REAS2.1(2013)	9.46	2.89	2.65	15.0
Huang et al.(2012)	3.2	5.3	1.3	9.8
Dong et al.(2010)	8.68	6.61	0.78	16.1
Wang et al.(2009)	4.3	8.82	0.26	13.4
Li et al.(2012)	1.82	8.30	1.88	12.0
Streets et al.(2003)	6.8	5.17	1.63	13.6
Yan et al.(2003)	4.32	2.48	0.21	7.01