Estimating ammonia (NH_3) emissions based on IASI observations from 2008 to 2018

Zhenqi Luo[1,2], Yuzhong Zhang[2], Ying Sun[1]

[1] School of Integrative Plant Science, Soil and Crop Sciences Section, Cornell University, Ithaca, NY, US. Contact: zl725@cornell.edu

[2] Key Laboratory of Coastal Environment and Resources of Zhejiang Province, School of Engineering, Westlake University, Hangzhou, Zhejiang Province, China

Introduction

- **Motivation:**
  - Atmospheric ammonia (NH_3) plays an important role in the formation of secondary aerosol, which impacts human health, climate, and nutrition deposition.
  - Large uncertainty in the estimated fluxes of NH_3 emissions exists, largely due to inadequate understanding of ***agricultural sources***.
- **Objective**: This study aims to provide a ***top-down*** estimate of global ***NH_3 emission fluxes*** utilizing IASI (Infrared Atmospheric Sounding Interferometer) observations and GEOS-Chem chemistry transport model simulations.

Data and methods

- **IASI observations**: NH_3 total column retrievals (ANNI-NH3-v3R)
  - 2008-2018, regrid to 4°×5°, monthly, morning (9.30AM)
  - Filtered by skin temperature (from ERA5), cloud fraction and number of retrievals
- **GEOS-Chem simulations**: v12.9.3, full chemistry
  - 2008-2018, 4°×5°, 47 vertical layers, 9-10AM
  - Bottom up emission inventories (**BUI1**) include CEDS (anthropogenic), GFED4 (fire), MEGAN (biogenic VOC), and overridden by regional inventories
- **Emission flux estimation:**

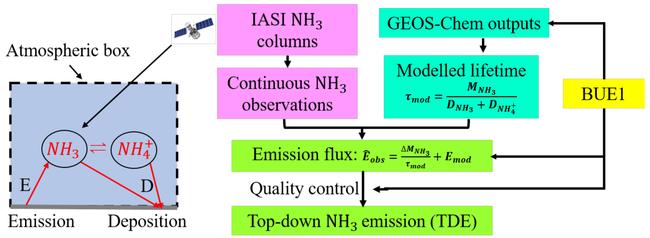


Figure 1. Flowchart of estimation framework.

NH_3 total column concentrations

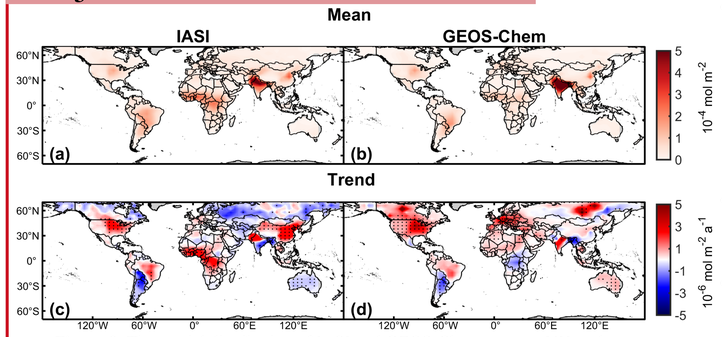
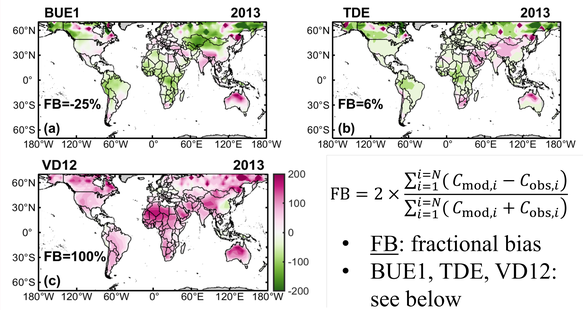


Figure 2. The spatial distribution of IASI and GEOS-Chem average NH_3 column concentrations (mol m^{-2}) and their linear trends (mol m^{-2} a^{-1}) during 2008-2018.

- **Model vs. Observation** (Fig. 2a and 2b) :
  - Good agreements (difference < 10%): the U.S., Europe, and southern South America.
  - Model underestimations (20-120%): eastern China, northern South America, and tropical Africa
  - Model overestimations (~50%): India
- **Observed positive trends** (Fig. 2c): the U.S. (3% a^{-1}), Brazil (0.5% a^{-1}), tropical Africa (2% a^{-1}), and China (5% a^{-1}),
- **Simulated positive trends** (Fig. 2d): meteorological conditions and/or fire emissions change due to invariant anthropogenic emission setting

Sensitivity test to NH_3 lifetime



$$FB = 2 \times \frac{\sum_{i=1}^{i=N}(C_{\mathrm{mod},i} - C_{\mathrm{obs},i})}{\sum_{i=1}^{i=N}(C_{\mathrm{mod},i} + C_{\mathrm{obs},i})}$$

- FB: fractional bias
- BUE1, TDE, VD12: see below

Figure 3. The comparisons with IASI NH_3 observations in 2013.

Emissions fluxes

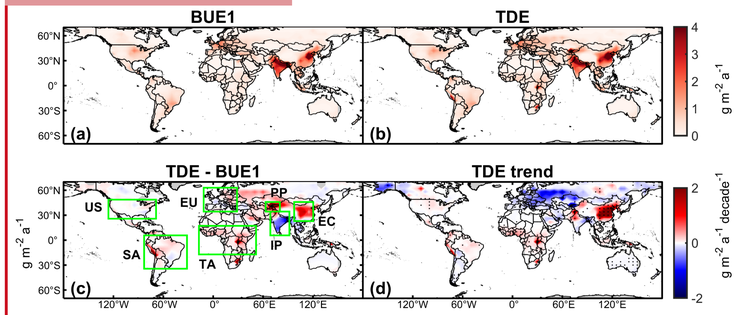
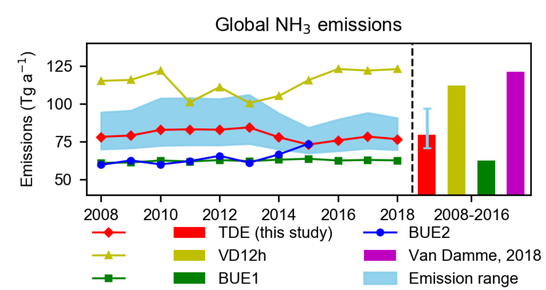


Figure 4. The spatial distribution of NH_3 emission fluxes (g m^{-2} a^{-1}) during 2008-2018.



- **Top-down emissions**:
  - TDE (this study results): 79 (71-96) Tg a^{-1}
  - VD12 (constant lifetime), following *Van Damme et al.* (2018), 121 Tg a^{-1}
- **Bottom-up inventories:**
  - BUE1: CEDS+GFED4+Natural, 62 Tg a^{-1}
  - BUE2: EDGAR+GFAS+Natural, 64 Tg a^{-1}
- TDE is about 30% higher than bottom-up inventories.

Figure 5. Comparison of NH_3 emissions with other top-down and bottom-up estimated

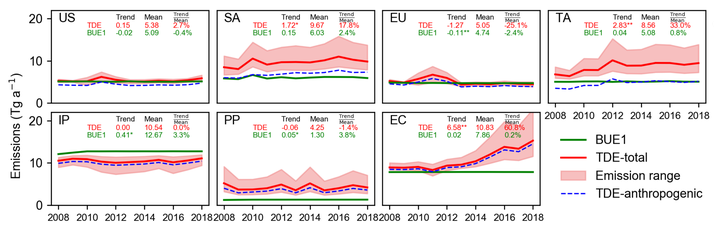


Figure 6. Annual NH_3 emissions (Tg a^{-1}) for seven regions between 2008-2018.

- **Positive emission trends**:
  - South America (SA): fertilizer input increase (5.5% a^{-1})
  - Tropical Africa (TA): manure management increase (2.8% a^{-1})
  - Eastern China (EC): inconsistent fertilizer usage change after 2014

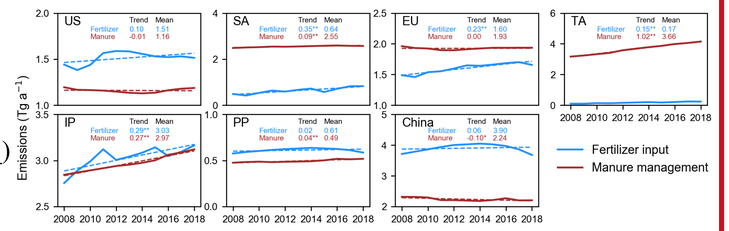


Figure 7. NH_3 emissions (Tg a^{-1}) from synthetic fertilizer and manure management based on FAO reports during 2008-2018.

Conclusion

- This study provides a top-down estimate of NH_3 fluxes constrained by IASI observations for 2008-2018. The global land NH_3 emission is 79 (71-96) Tg a^{-1}, ~30% higher than the bottom-up estimates.
- At the regional scale, the bottom-up inventory underestimates NH_3 emission over South America, tropical Africa, and eastern China by 40-70%, but reasonably agrees with our top-down estimates over the U.S. and Europe.
- There are significant increases in eastern China (61% decade^{-1}), tropical Africa (33% decade^{-1}), and South America (18% decade^{-1}), likely resulting from the intensified agricultural activities in these regions in the past decade.

Acknowledgement

- This study is supported by (Yuzhong's funding) and Cornell Institute of Digital Agriculture.
- We thank Mr. Wei Chen (Westlake University) for helping simulating the GEOS-Chem model and Dr. Peter Hess (Cornell University) for providing constructive suggestions.

Reference

- Van Damme, M., Clarisse, L., Whitburn, S., Hadji-Lazaro, J., Hurtmans, D., Clerbaux, C., & Coheur, P. F. (2018). Industrial and agricultural ammonia point sources exposed. Nature, 564(7734), 99-103.
- Evangeliou, Nikolaos, et al. "10-year satellite-constrained fluxes of ammonia improve performance of chemistry transport models." Atmospheric Chemistry and Physics 21.6 (2021): 4431-4451.
- Jacob, Daniel J. Introduction to atmospheric chemistry. Princeton University Press, 1999.