Project 22-019

Refining ammonia emissions using inverse modeling and satellite observations over Texas and the Gulf of Mexico and investigating its effect on fine particulate matter

PREPARED BY

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August 2023

Outline

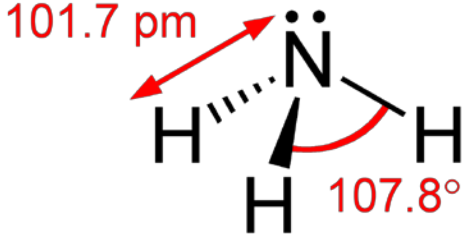
- Introduction
- Modeling Setup
- Inverse modeling technique
- Posterior Evaluation
- Results
- Conclusion

Introduction

Ammonia

The need to sustain food production, because of growing populations, causes the increased agricultural emissions of NH_3 with perturbations of:

- Nitrogen cycle and biogeochemical process
- Inorganic Fine Particulate Matter (PM)
- Climate change

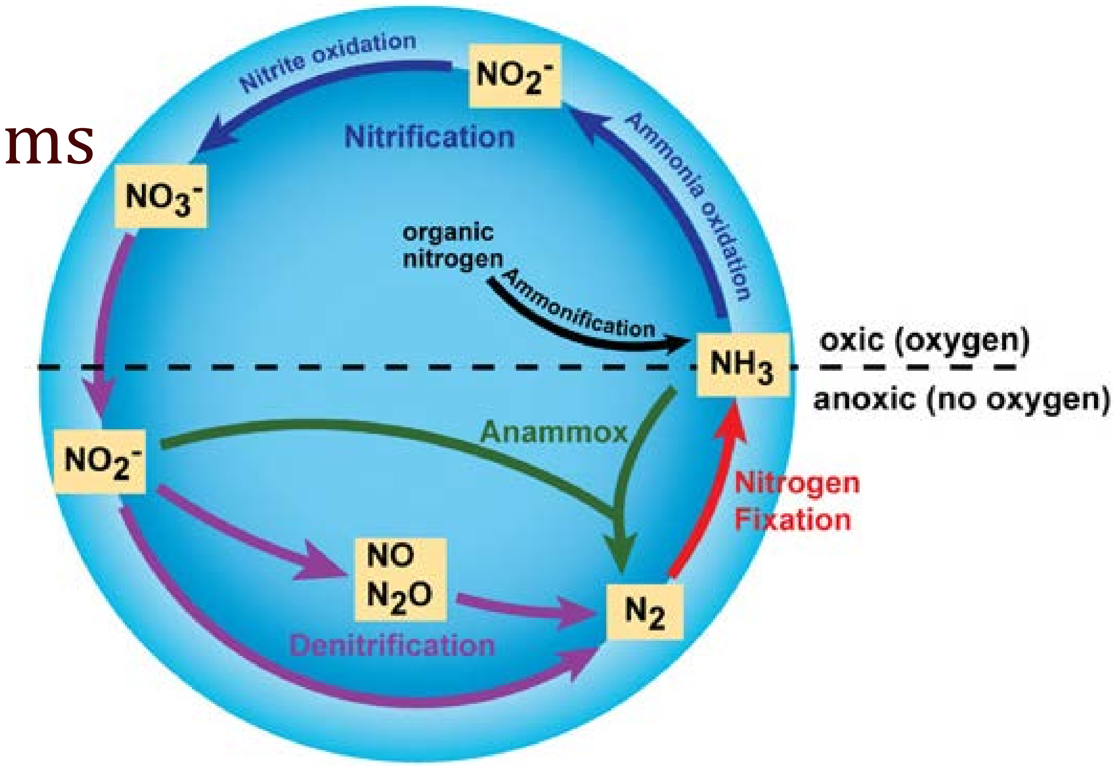


Source: Wikipedia

Introduction

Nitrogen cycle and biogeochemical process

- Acidification of terrestrial ecosystems
- Loss of biodiversity



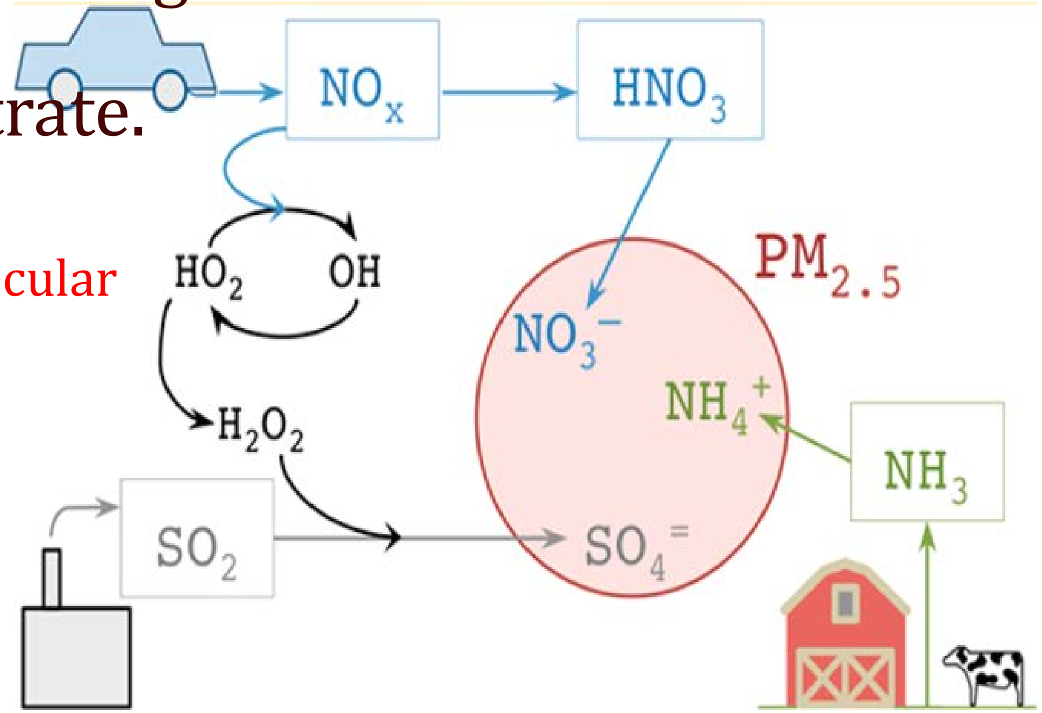
Bernhard, A. (2010) The Nitrogen Cycle: Processes, Players, and Human Impact. *Nature Education Knowledge* 3(10):25

Introduction

Inorganic Fine Particulate Matter (PM)

- A significant role in the formation of inorganic fine aerosols
- Ammonia neutralizes sulfate and nitrate.

➢ leading to various health issues such as cardiovascular disease, asthma, and respiratory problems (Cheng & Wang-Li, 2019; Pui et al., 2014)



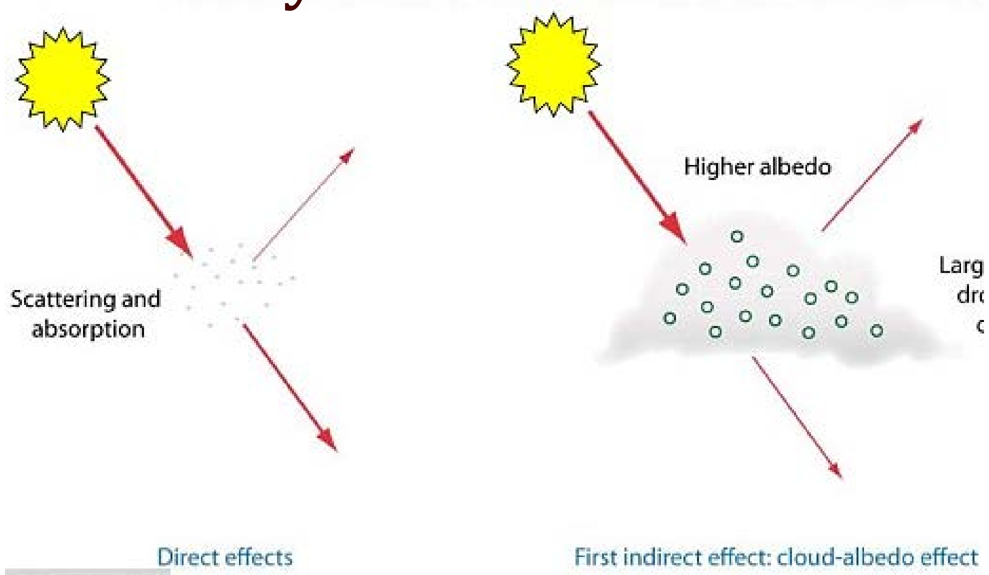
Source: Holt et al, 2015

Introduction

Climate Change (Radiative Forcing)

- Ammonia can be converted to Aerosols which have two effects upon climate:

1. They can directly reflect sunlight back away from the Earth.
2. They can interact with clouds



Source: Martin Wild

Introduction

Uncertainty in Ammonia Emissions

(Unit: Teragram N per year)

Source	Global	North America	Contiguous US	China
Schlesinger and Hartley	75			
Dentener and Crutzen	45	5.2		
Bouwman et al.	54	3.6		
EDGAR v4.2	40.6	4.1	2.9	
Moss et al.	38.5	5.1	3.4	

- **This uncertainty would be propagated in estimation of PM and nitrogen cycle.**

Introduction

Reduction in Uncertainty in Ammonia Emissions

- Inverse modeling methods:
  - A well-established approach for refining emission inventories and constraining modeling predictions.

- Observation:
  - Remote sensing data, such as NH_3 columns from Cross-track Infrared Sounder (CrIS) instrument, are commonly employed for inverse modeling techniques.

Introduction

- Remote sensing data:
  - Advantage: Contribute to our understanding of pollutant spatial patterns.
  - Limitations: inadequate spatial and temporal coverage and uncertainties in measurements.

- In contrast, chemical transport models (CTM):
  - Advantage: comprehensive data with high spatiotemporal resolution for all species.
  - Limitations: uncertainties arising from the numerical representation of chemical and physical processes in the atmosphere, as well as uncertainties in modeling inputs.

- By combining the strengths of modeling data and observations, inverse modeling techniques enhance modeling predictions by effectively addressing uncertainties present in both the predictions and observational data.

Tasks

- Task 1: Preparation of comprehensive satellite, in situ, and modeling data for iFDMB method,

- Task 2: Development of the Reduced-Complexity CMAQ Model (RCCM) for NH_3 and refinement of NH_3 emissions using iterative Finite Difference Mass Balance (iFDMB) with the combination of CMAQ model and IASI/CrIS satellite observations,

- Task 3: Investigation of $PM_{2.5}$ concentrations using the updated emission inventory.

Modeling Setup

- Weather Research and Forecasting (WRF):
  - Version 4.2
  - Domain sizes of 150×150 for the 12-km
  - The initial and boundary conditions from the North American Mesoscale Forecast System (NAM) reanalysis datasets.
- Sparse Matrix Operator Kernel Emissions (SMOKE)
  - Emissions input from National Emissions Inventory (NEI)
  - NEI point sources, NEI nonpoint sources, NEI on-road sources, and NEI nonroad sources.
  - Biogenic Emissions from the Biogenic Emissions Inventory System (BEIS3)
  - NEI 2017 employed to produce emissions for Texas throughout the entire year of 2019
  - Biogenic oceanic emissions for the Gulf of Mexico from the Emissions Database for Global Atmospheric Research (EDGAR)
- Community Multiscale Air Quality (CMAQ):
  - Chemical transport model developed and maintained by the US EPA
  - Version 5.0.1

Inverse Modeling Setup

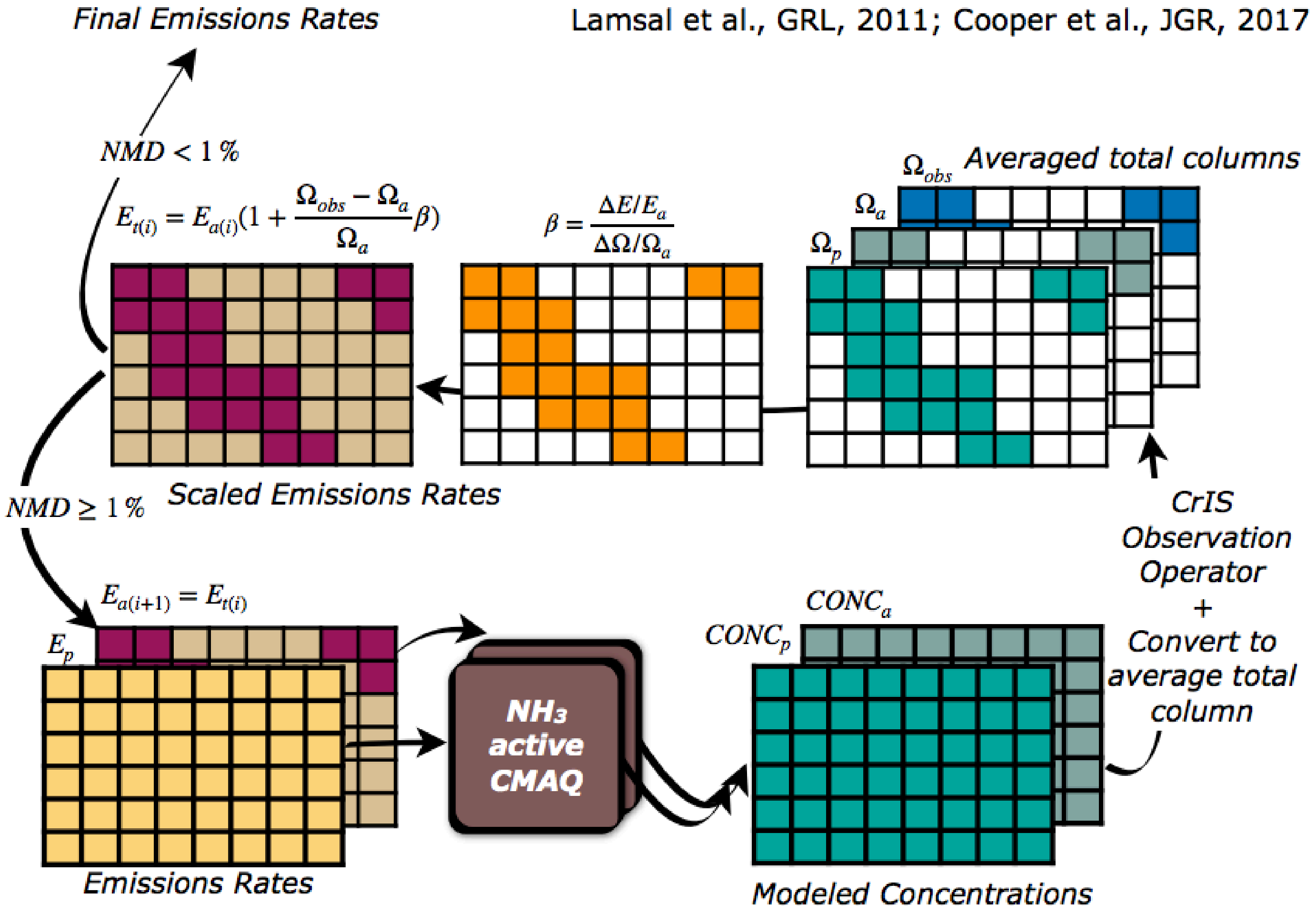
- Inverse modeling technique: iFDMB

$$E_t = E_a(1 + \frac{1}{\beta}\frac{\Omega_o - \Omega_a}{\Omega_a})$$

$$\beta = \frac{\Delta\Omega/_{\Omega}}{\Delta E/_{E}}$$

- β is the initial sensitivity by a simulation with E_a perturbed by %20
- the normalized mean difference (NMD) of 2% is has been employed

Lamsal et al., GRL, 2011; Cooper et al., JGR, 2017



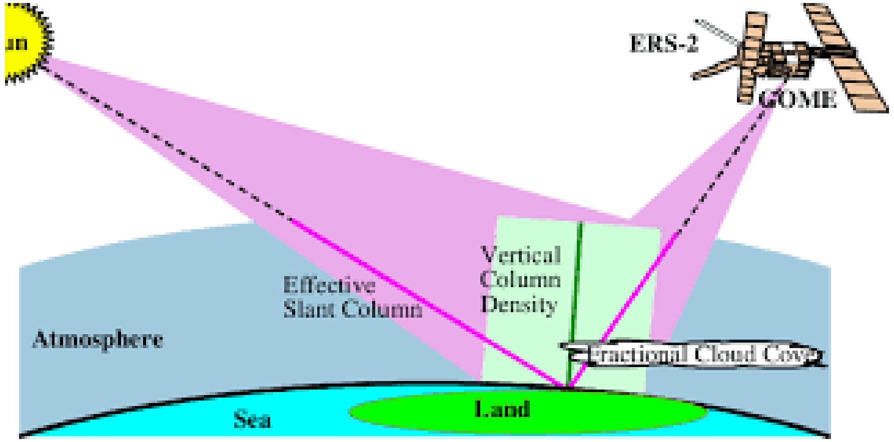
Inverse Modeling Setup

- Satellite data
  - NH_3 column density data for the year 2019 from:
    - Cross-track Infrared Sounder (CrIS)
    - Infrared Atmospheric Sounding Interferometer (IASI)

- Observation operator
  - For an apple-to-apple comparison with satellite column density, the vertical column of the model should be calculated:

$$VCD_i = -c_i \times \Delta P_i \times 2.119 \times 10^{14}$$

$$VCD = \sum_i VCD_i$$



Inverse Modeling Setup

- Reduced-Complexity CMAQ Model (RCCM) for NH_3
  - iFDMB is computationally expensive and requires numerous iterations
  - To reduce the computational cost, an RCCM was employed to simulate NH_3
  - NH_3 and NH_4^+ are considered as two tracer pollutants of the model
  - All of the chemical processes of other species are turned off
  - Include dry and wet deposition, the transport of NH_3 and NH_4^+, and NH_x partitioning
  - ISORROPIA-II in the aerosol module calculates the gas-particle partitioning of NH_3 and NH_4^+
  - To handle the chemical processes, offline files required for running RCCM
  - Offline files contain sulfate (SO_4^{2-}), nitric acid (HNO_3), nitrate (NO^{3-}), chloride (Cl), sodium (Na), hydrochloric acid (HCl) concentration in all times steps

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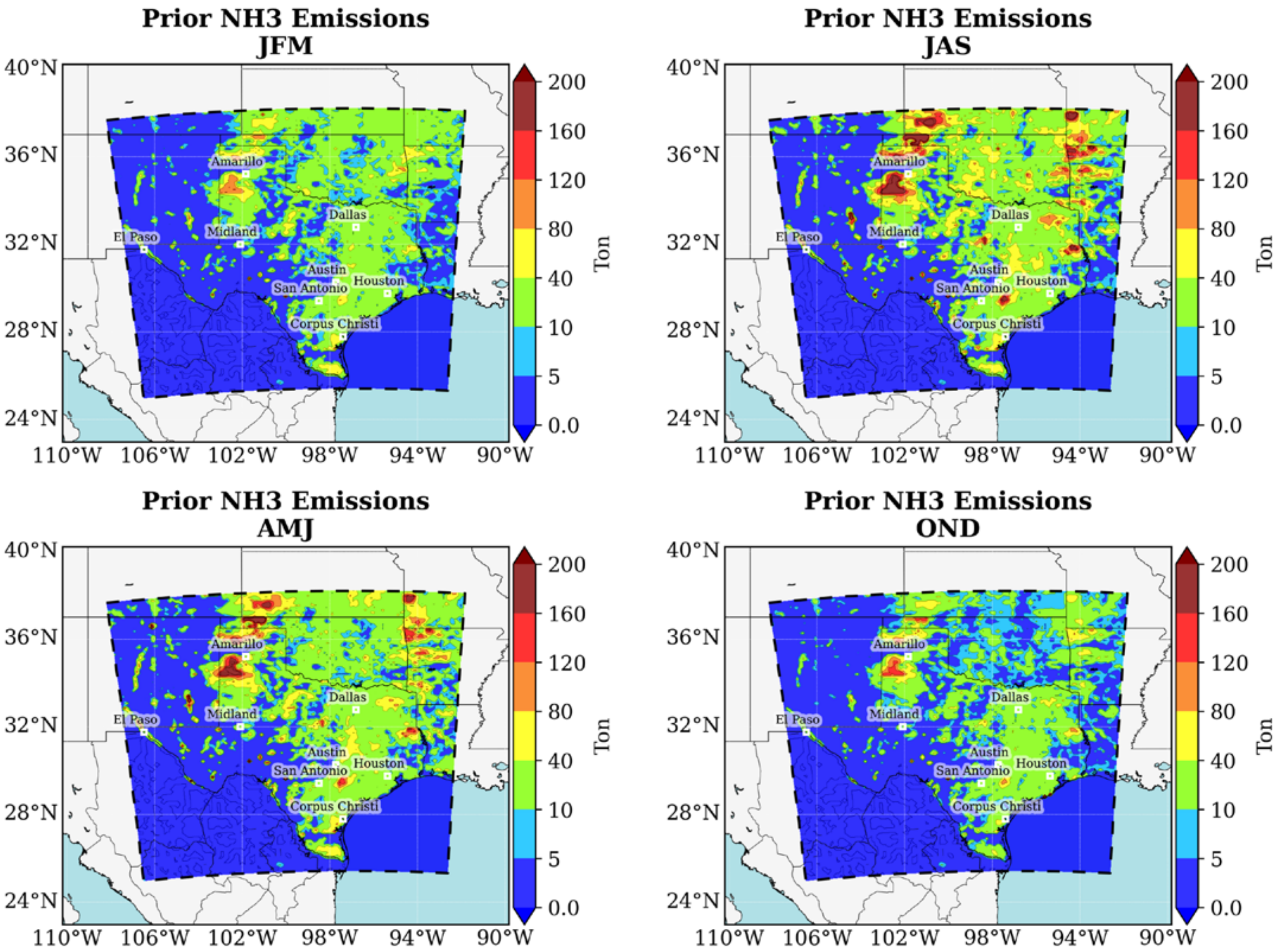
Inverse Modeling Setup

- A-priori NH_3 Emissions over the domain

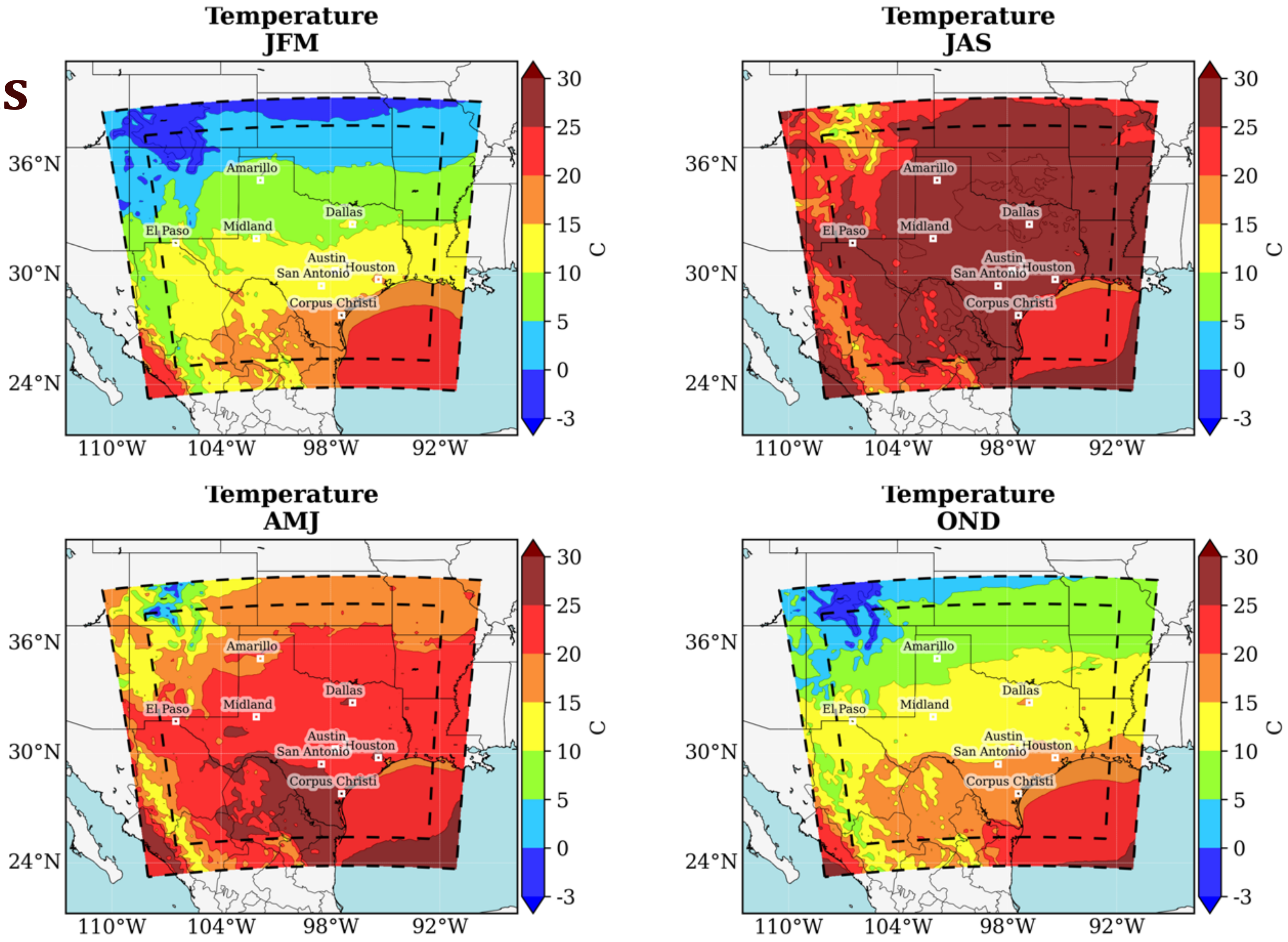


Posterior evaluation

- In order to assess the efficiency of the model:
  - Comparison between the posterior and prior estimates versus data from the CrIS/IASI satellites.
- The evaluation of the posterior emissions:
  - Comparison between the posterior simulation from updated emissions with surface measurements
  - For NH_3 concentration:
    - The Ammonia Monitoring Network (AMoN) from the National Atmospheric Deposition Program (NADP).
    - Biweekly NH_3 concentration
  - For NH_4^+ wet deposition:
    - The NADP's National Trends Network (NTN)
    - Weekly wet deposition

Results

Meteorological fields



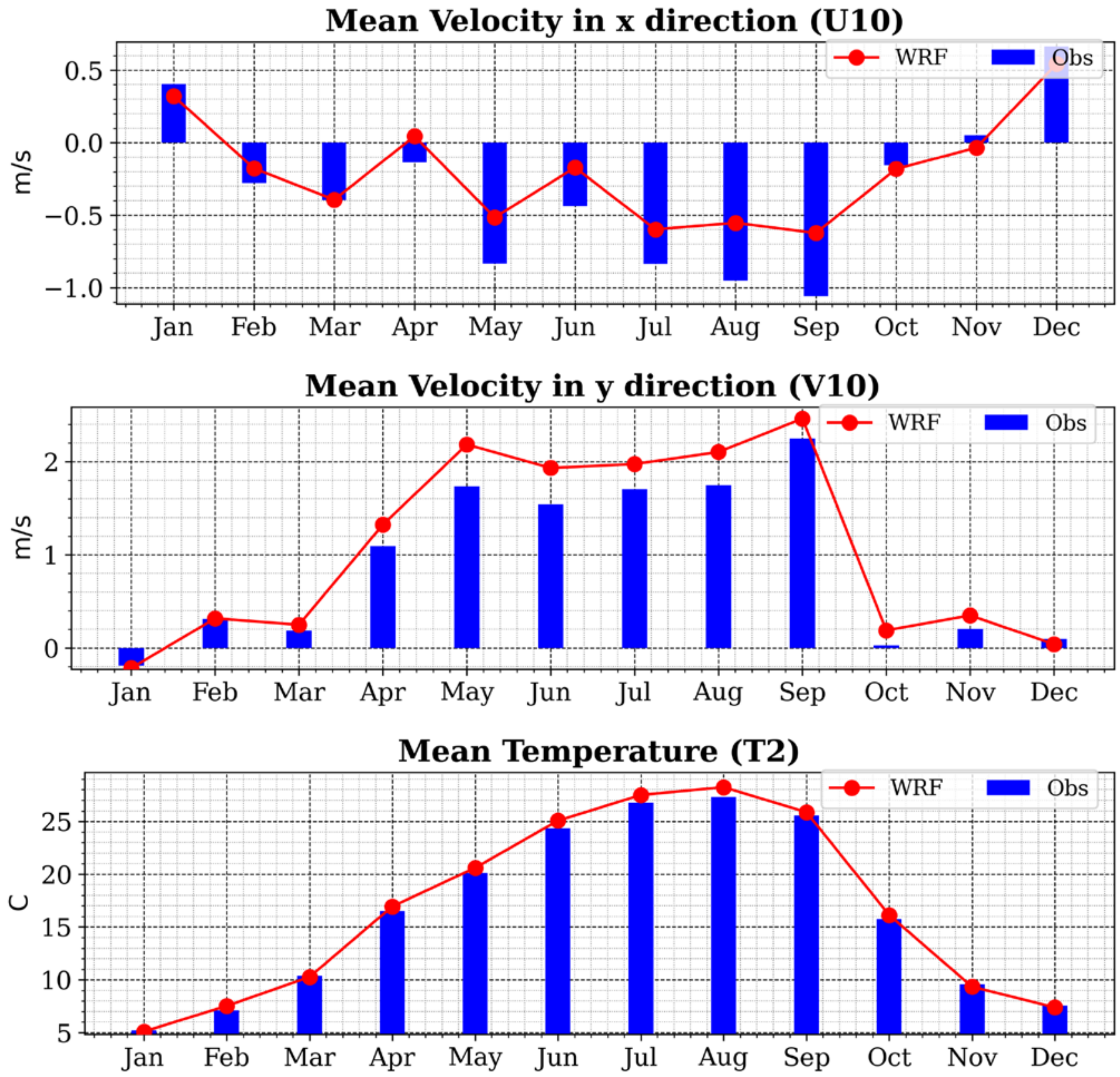
Results

Meteorological fields

- Comparison the WRF model outputs against the observed data from the MADIS stations.

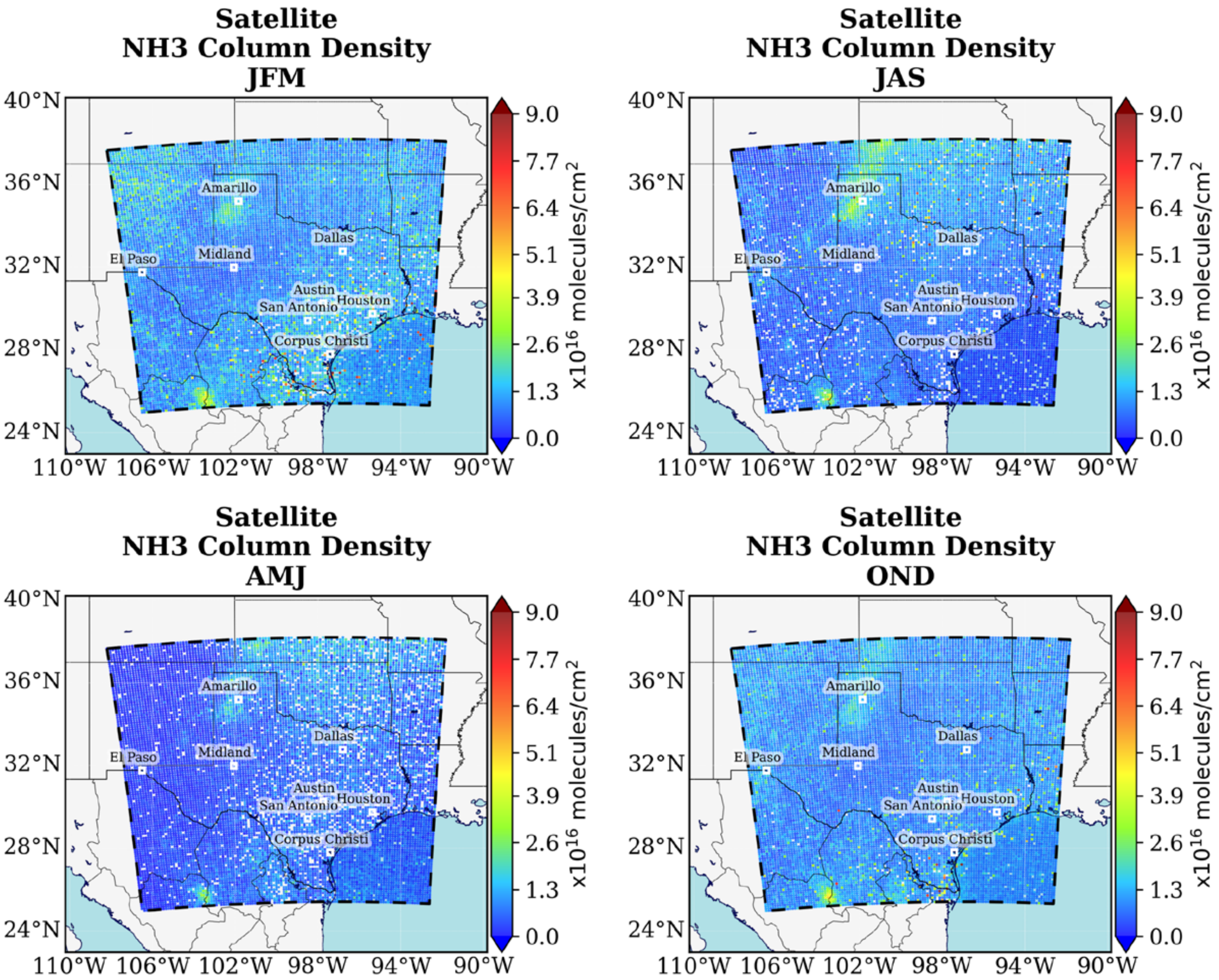
Variable	R	IOA	RMSE	MAE	Errors Unit
U10	0.66	0.79	1.89	1.51	m/s
V10	0.78	0.86	2.19	1.74	m/s
T2	0.95	0.96	2.94	2.26	C

- WRF model showed a promising ability to simulate atmospheric conditions over the region



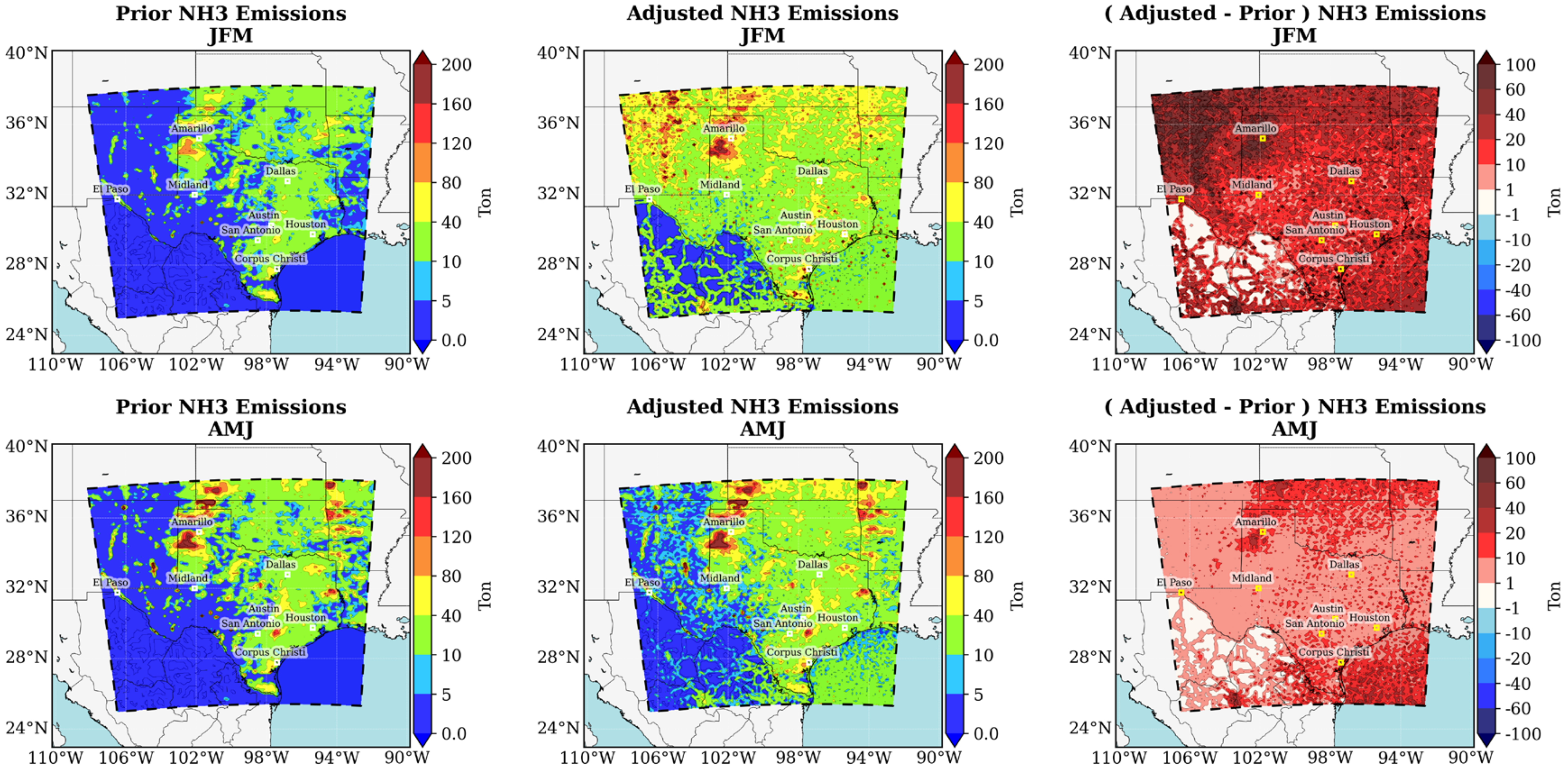
Results

Satellite data



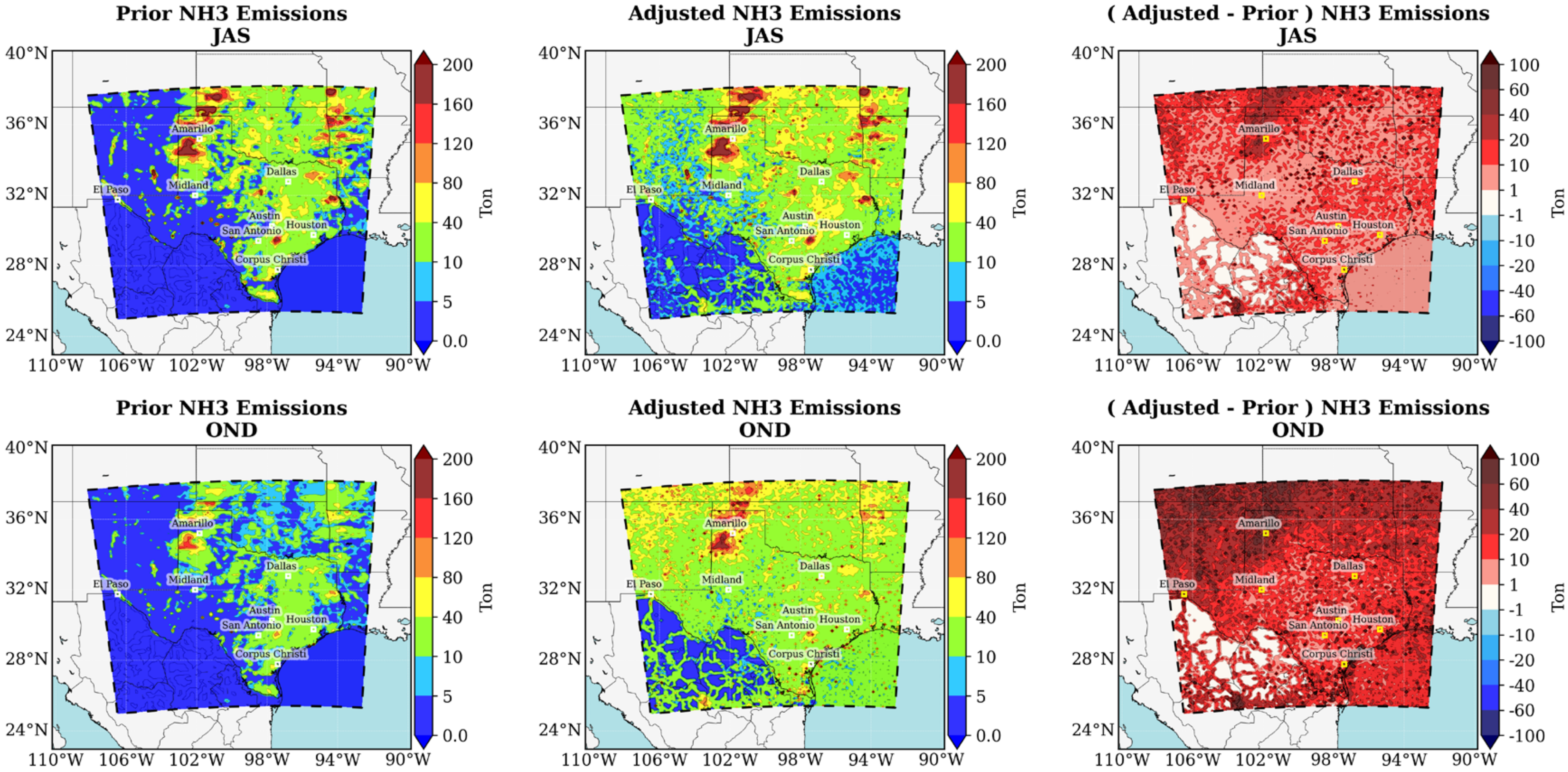
Results

Adjusted emissions



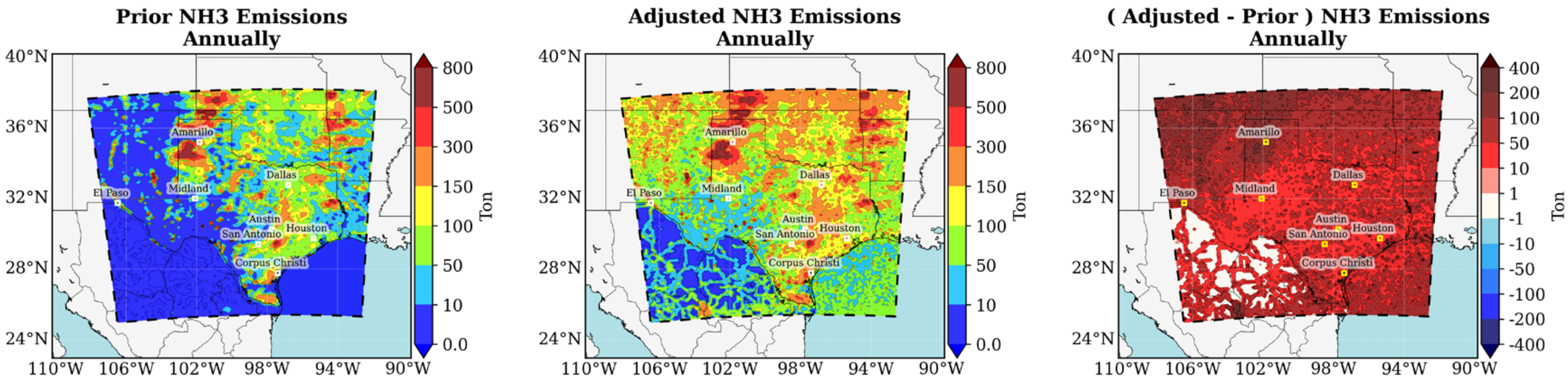
Results

Adjusted emissions



Results

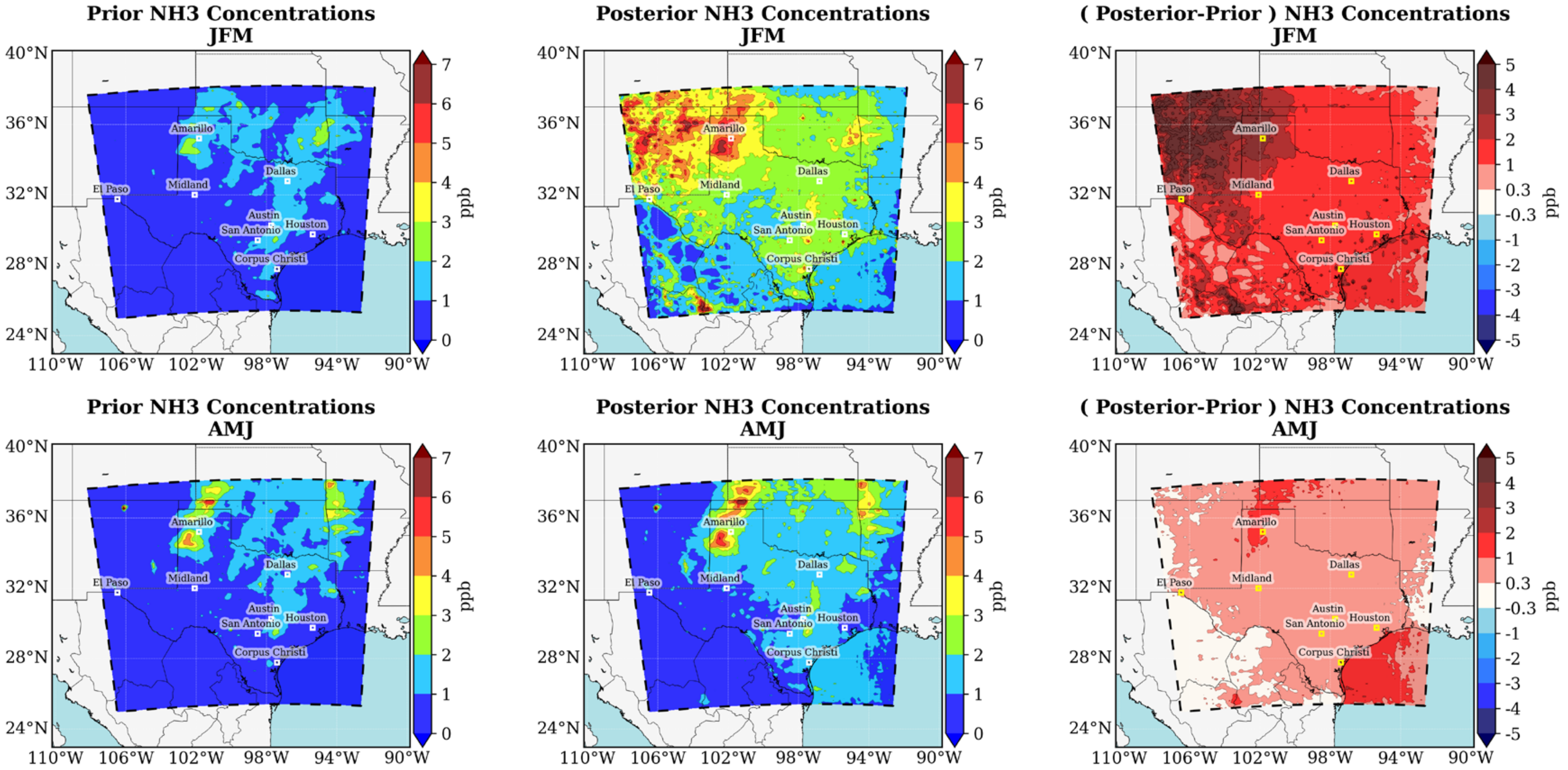
Adjusted emissions



- Across all quartiles and annual updated emissions, a noticeable increase in ammonia emission levels is observed over Texas.
- The coastline and extending over the Gulf of Mexico, demonstrates significantly elevated emission levels in comparison to the a-priori emissions.
- The updated emissions derived through our inverse modeling suggest the necessity of a thorough reevaluation of ammonia emissions from the Gulf of Mexico.

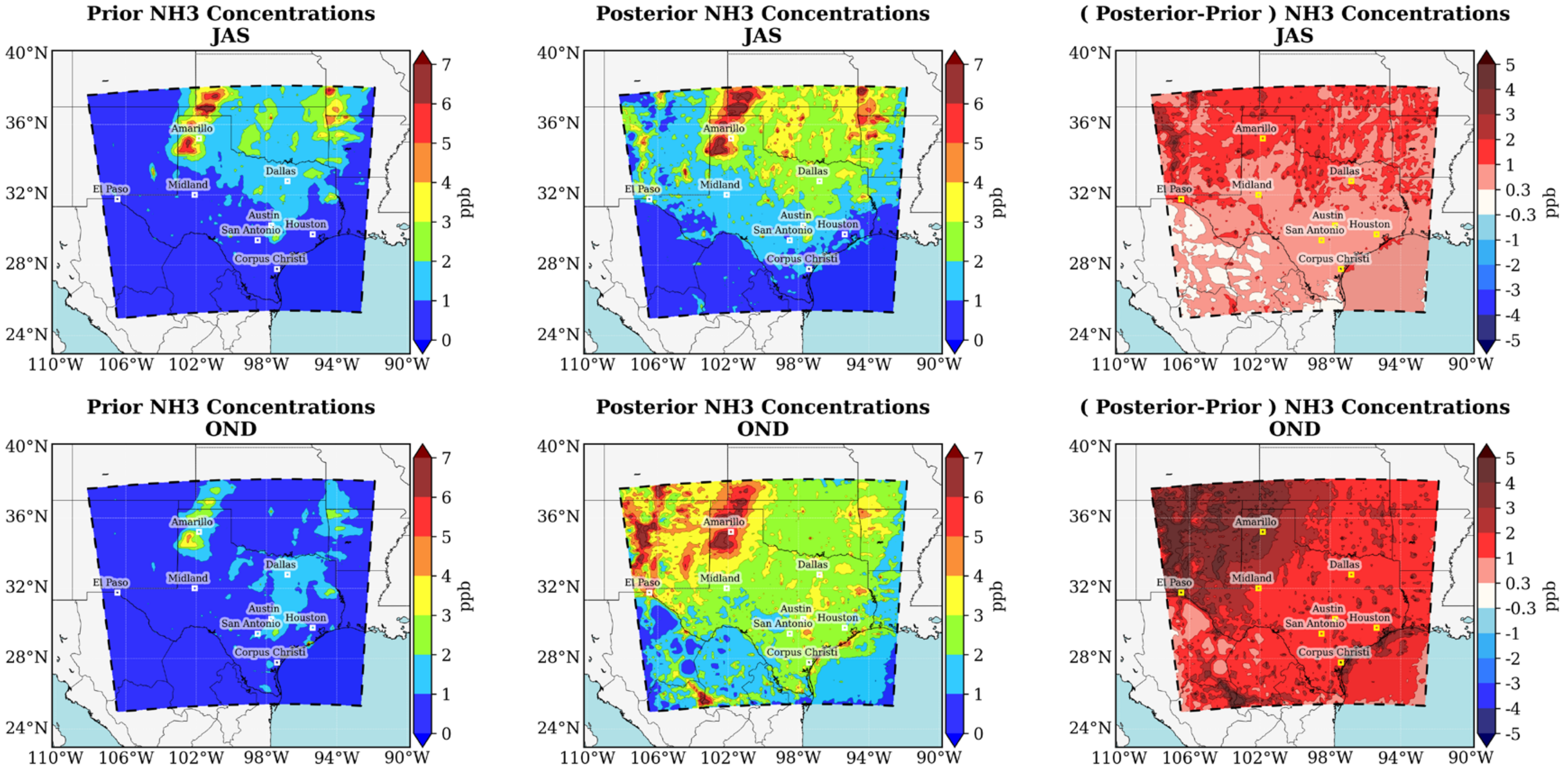
Results

Posterior NH_3 concentrations



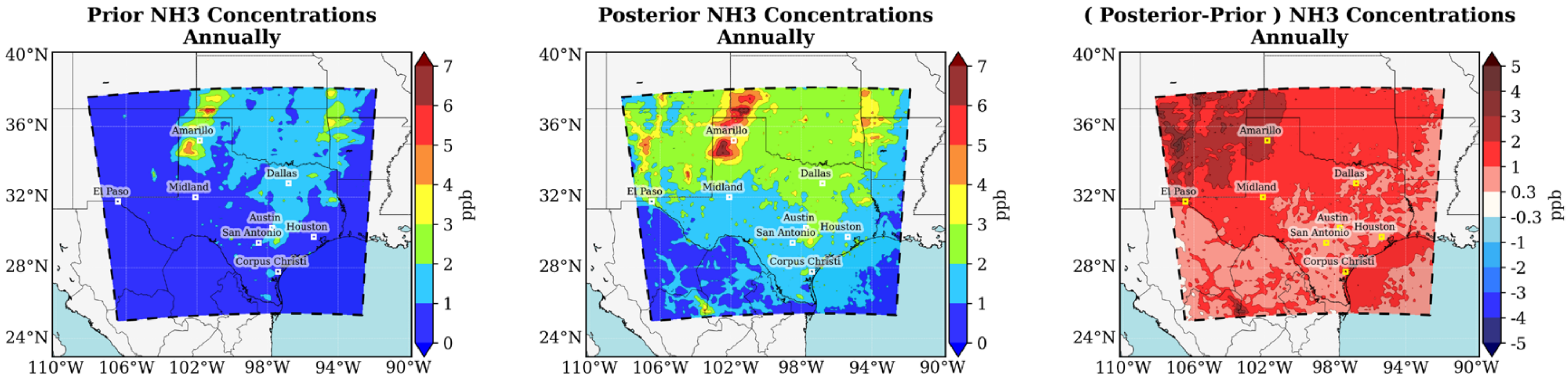
Results

Posterior NH_3 concentrations



Results

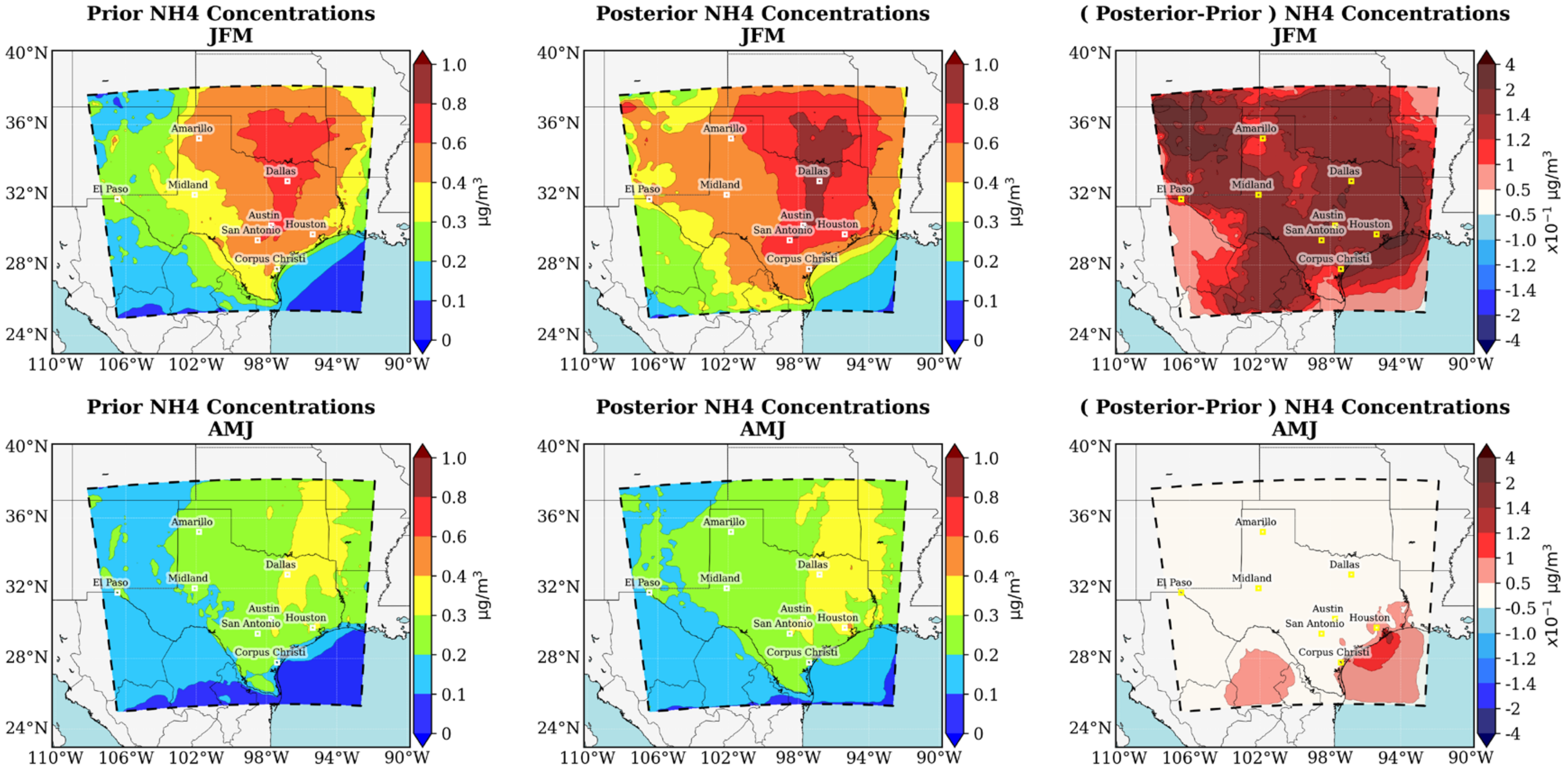
Posterior NH_3 concentrations



- NH_3 concentrations across Texas have shown an increase.
- The most substantial increase occurred in the Northwestern regions, where values rose between 3-4 ppb.
- In the rest of Texas, including most of the northern parts and some eastern areas, the increase ranged between 1-2 ppb.
- Over the Gulf of Mexico, the 1-2 ppb increase in NH_3 concentrations highlights the notable role of sources over the ocean, related to biological nitrogen fixation and maritime and oil industrial activities.

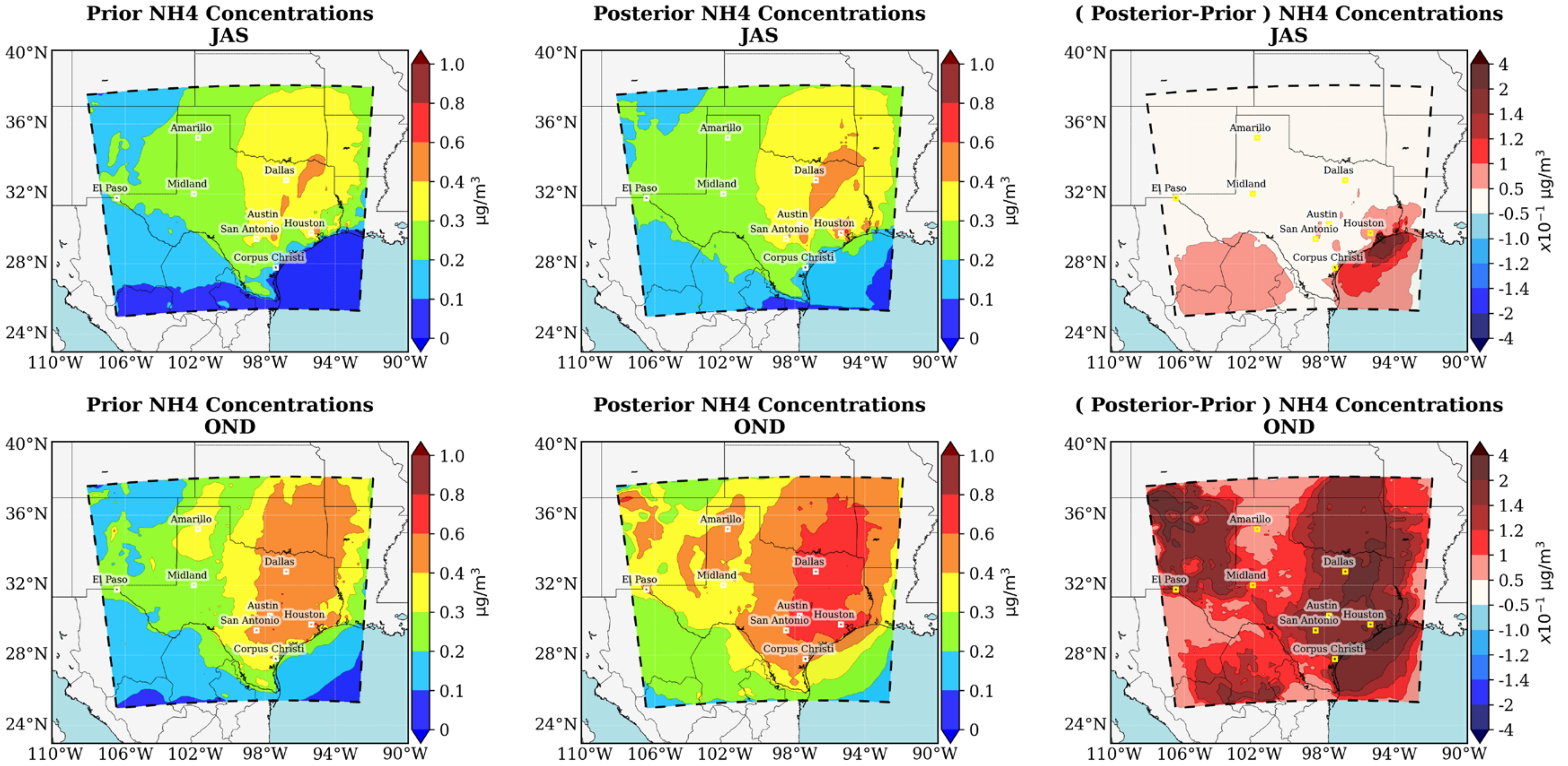
Results

Posterior NH_4^+ concentrations



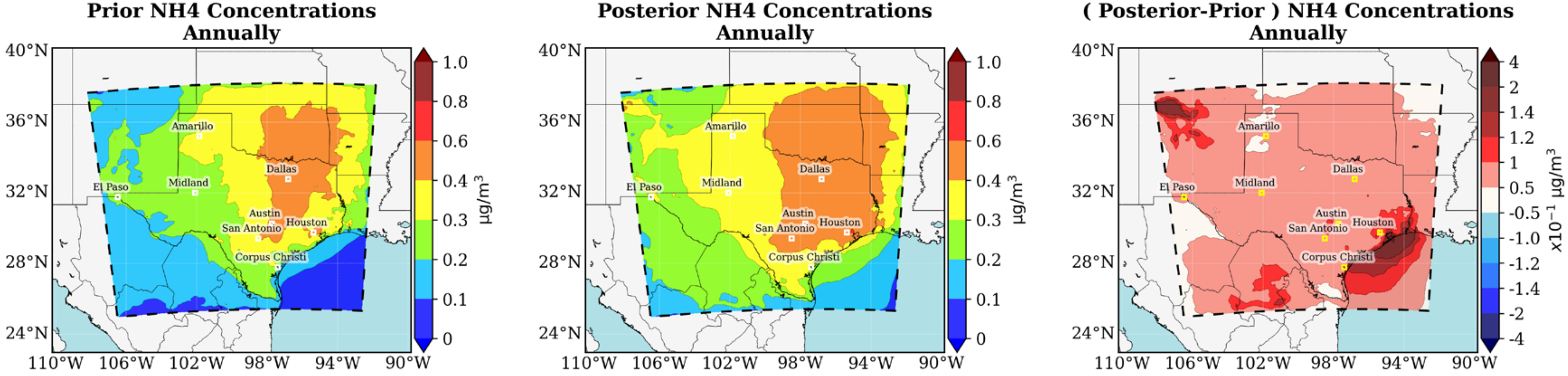
Results

Posterior NH_4^+ concentrations



Results

Posterior NH_4^+ concentrations



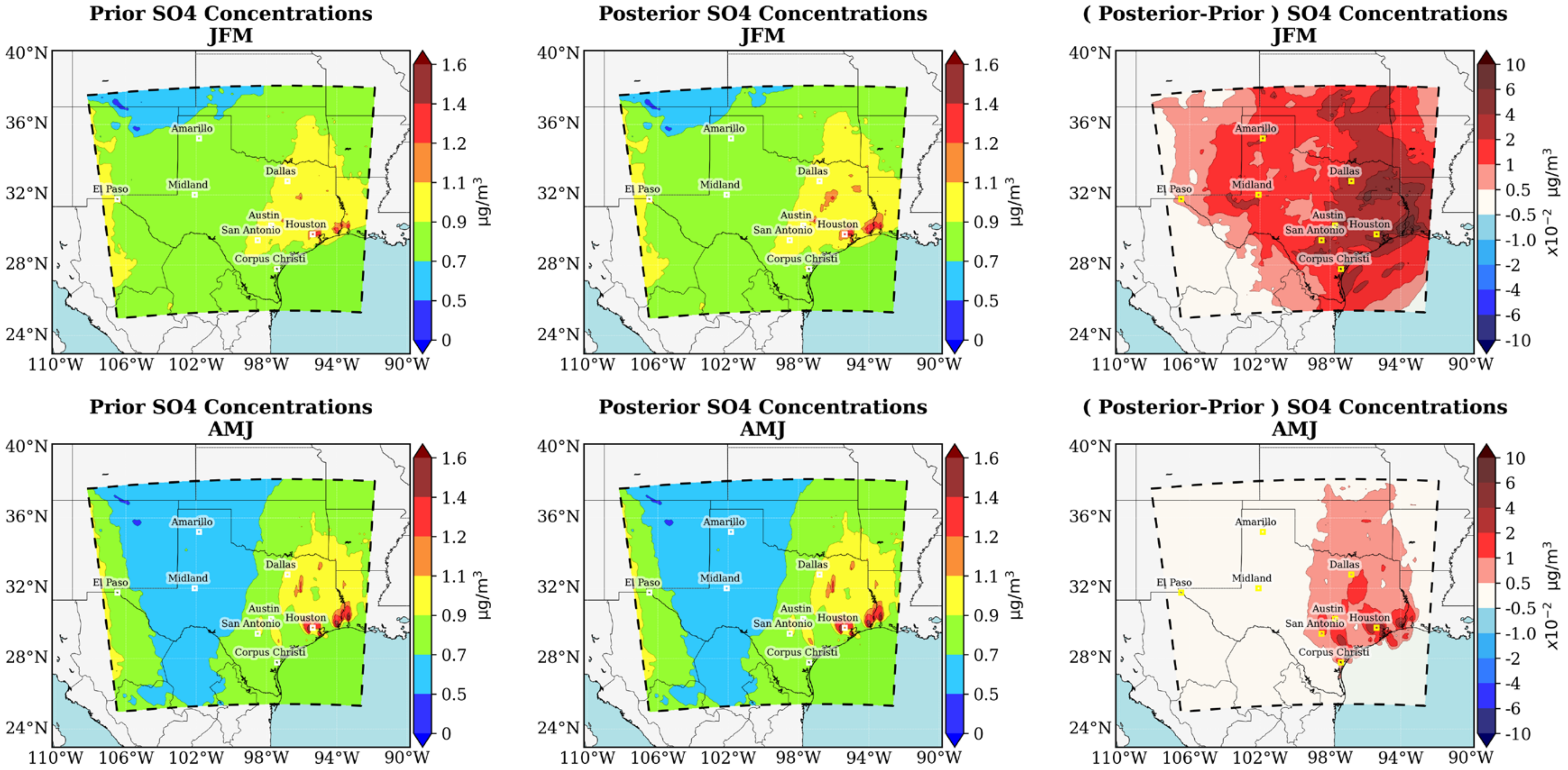
- NH_4^+ concentrations over Texas increased slightly, whereas values in the eastern part of the state increased more significantly.

- Over the Gulf of Mexico, NH_4^+ values escalated more compared to those over Texas.

- The notable hotspot over the Gulf of Mexico, particularly in the southern parts, warrants further investigation.

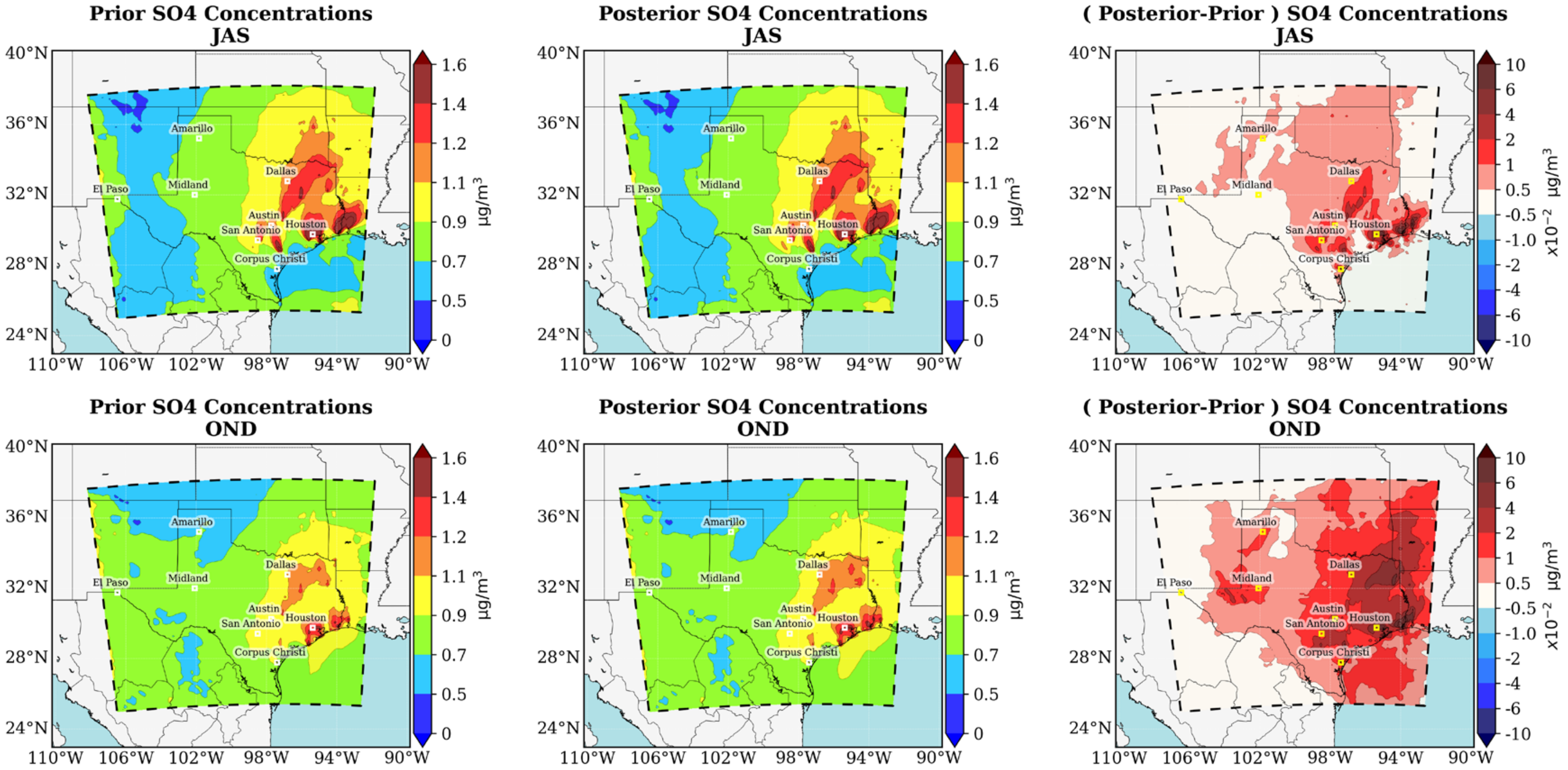
Results

Posterior SO_4^{2-} concentrations



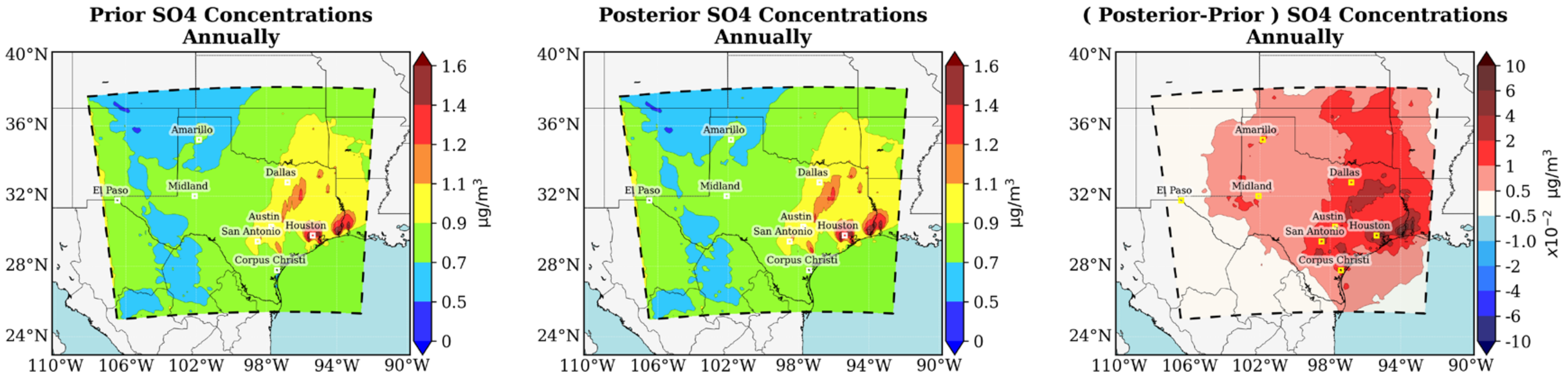
Results

Posterior SO_4^{2-} concentrations



Results

Posterior SO_4^{2-} concentrations

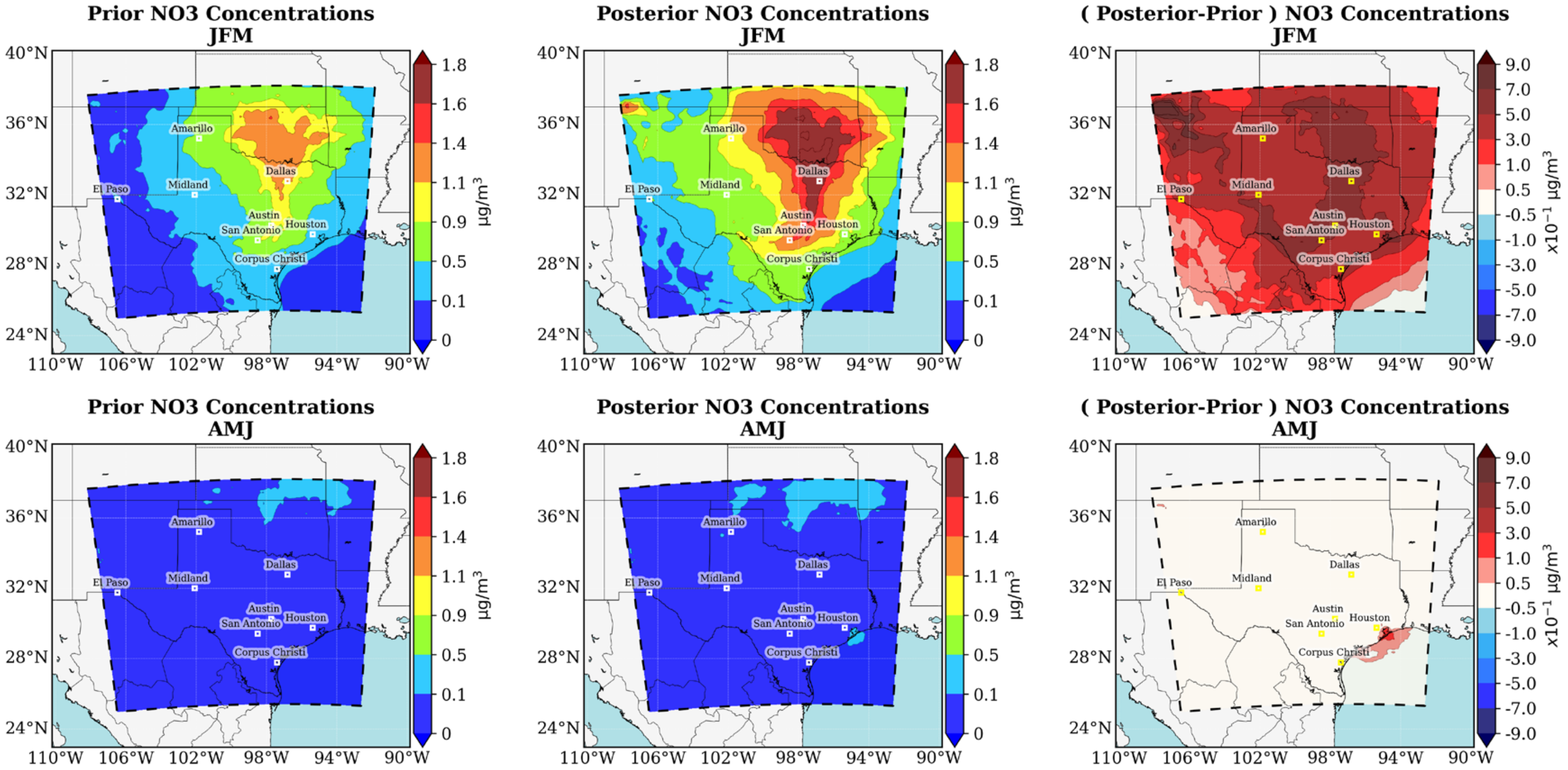


- A-posteriori SO_4^{2-} concentrations experienced a greater increase in eastern Texas, particularly in areas where a-prior SO_4^{2-} concentrations were already high.

- The significant increase in SO_4^{2-} concentrations near the Port Arthur and Galveston area, located in the southeast, emphasizes the potential importance of human activities, such as industrial operations and shipping, in contributing to the SO_4^{2-} emissions within this region.

Results

Posterior NO_3^- concentrations



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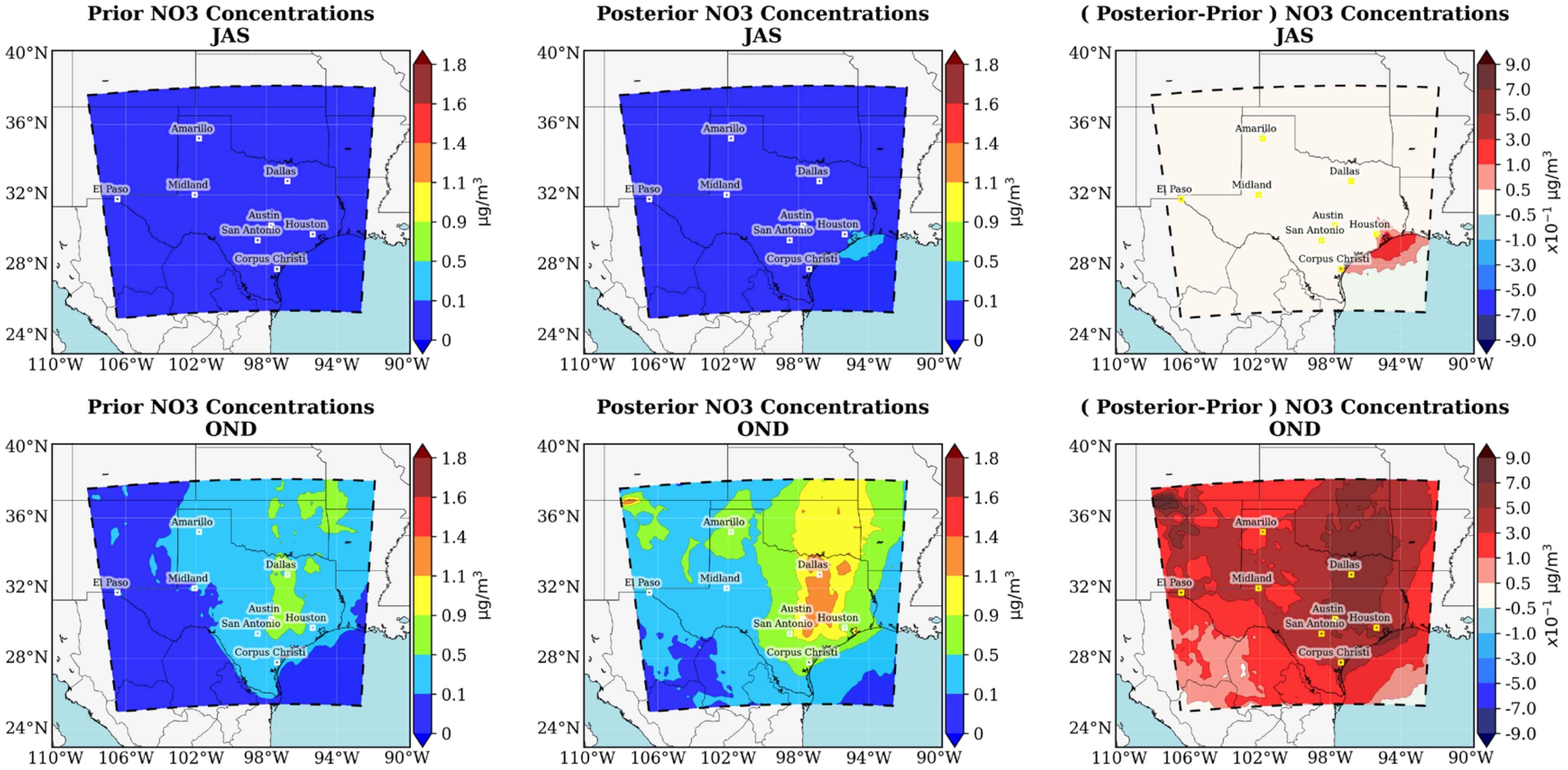
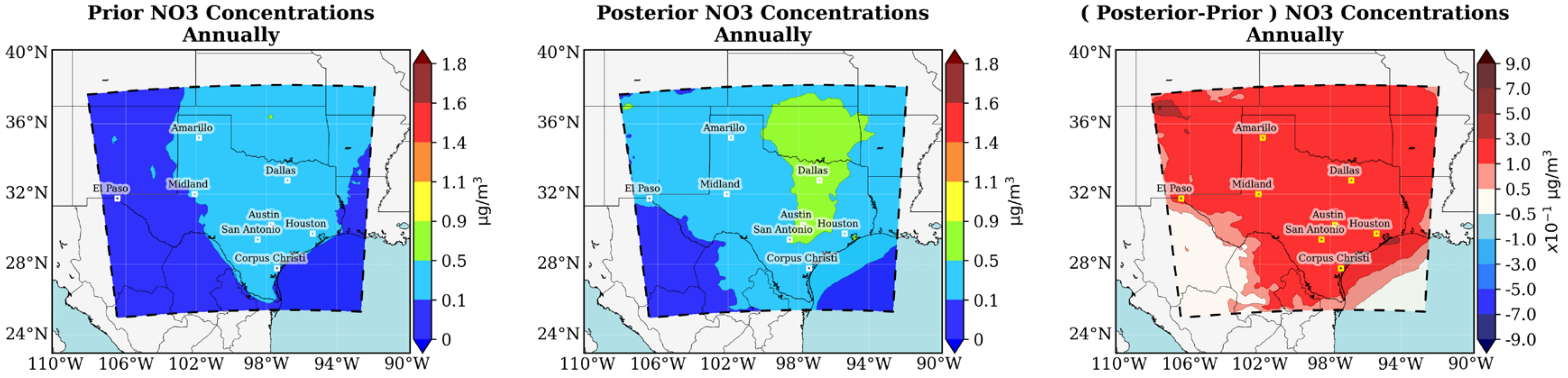
Results

Posterior NO_3^- concentrations



Results

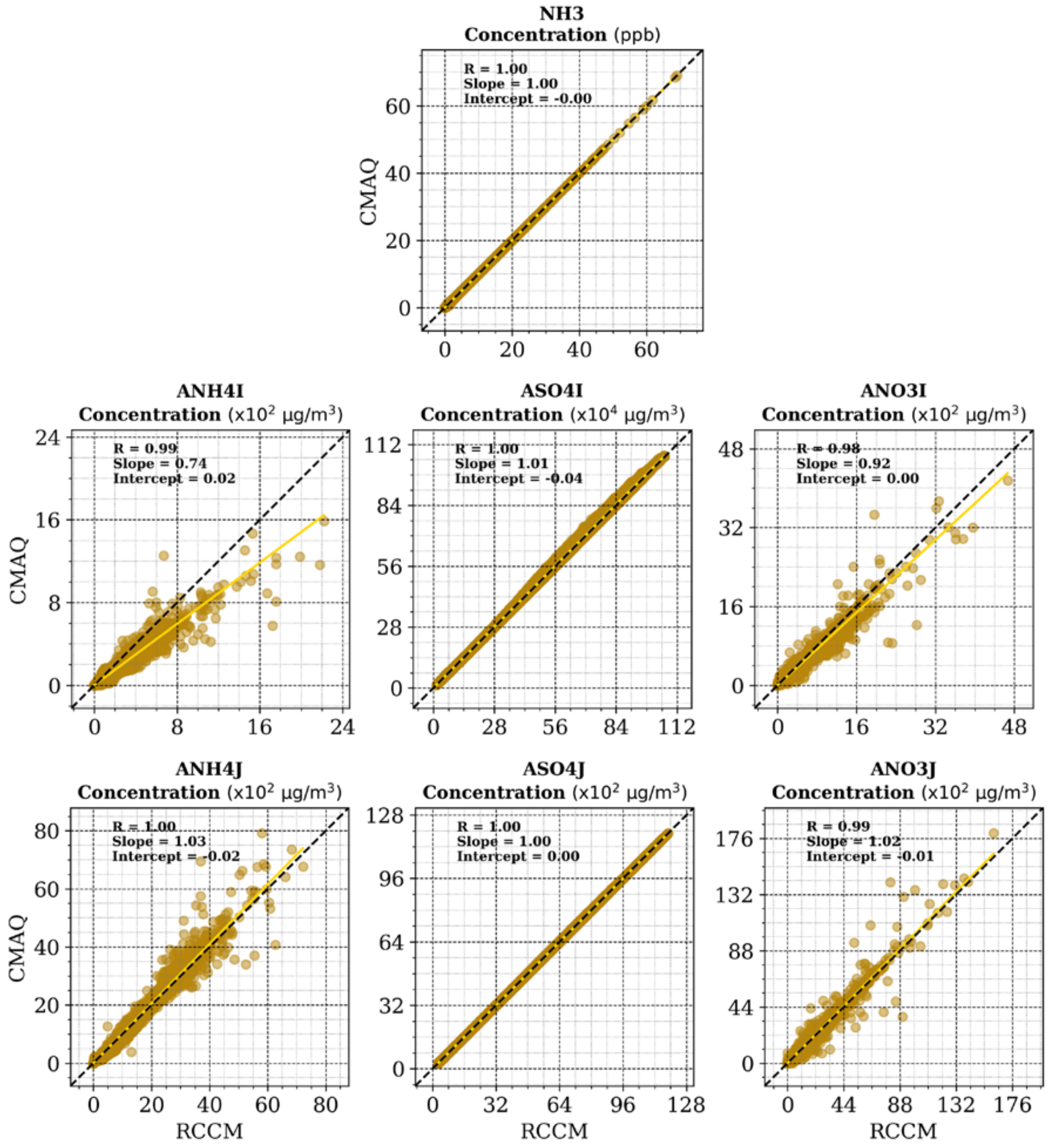
Posterior NO_3^- concentrations



- For NO_3^-, increases were simulated throughout Texas.
- The elevated NH_3 has likely resulted in an ammonia-rich regime where adequate NH_3 neutralizes SO_4^{2-} to form NO_3^- production.
- The enhanced a-posteriori NO_3^- levels along the shoreline in the Gulf of Mexico are also noteworthy. This increase may be attributable to the high levels of SO_2 emitting from shipping activities, further emphasizing the human influence on these emissions.

Evaluation

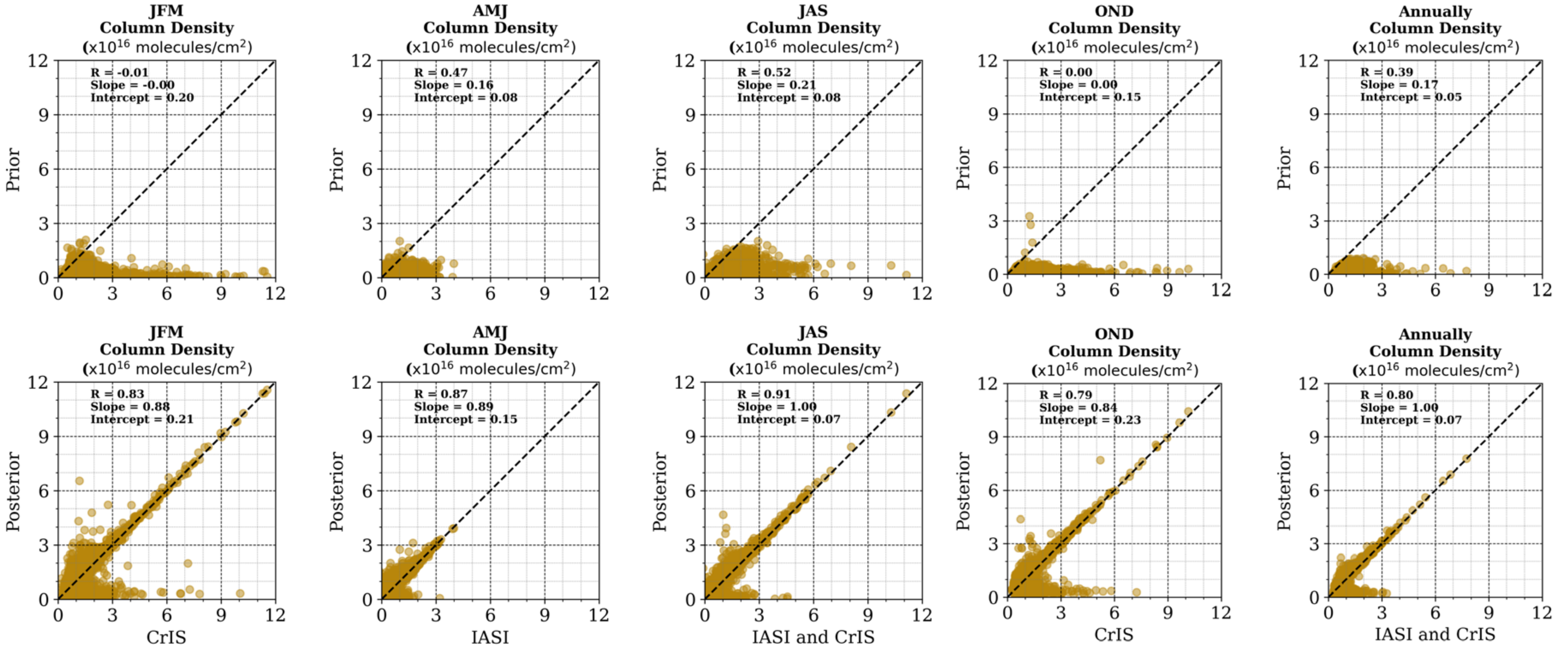
RCCM



- The results illustrate that the RCCM was in closer agreement with the standard CMAQ.

Evaluation

Posterior evaluation with satellites observations



Evaluation

Posterior evaluation with satellites observations

Quarter		R	MAE	NMSE	RMSE	IOA
JFM	Prior	-0.01	1	6.82	1.26	0.65
	Posterior	0.83	0.27	0.13	0.45	0.91
AMJ	Prior	0.47	0.42	2.99	0.55	0.39
	Posterior	0.87	0.17	0.15	0.24	0.92
JAS	Prior	0.52	0.59	2.95	0.80	0.48
	Posterior	0.91	0.19	0.10	0.28	0.95
OND	Prior	0.0002	0.88	6.95	1.04	0.67
	Posterior	0.79	0.23	0.12	0.36	0.89
Annually	Prior	0.39	0.70	3.32	0.77	0.39
	Posterior	0.80	0.20	0.09	0.28	0.89

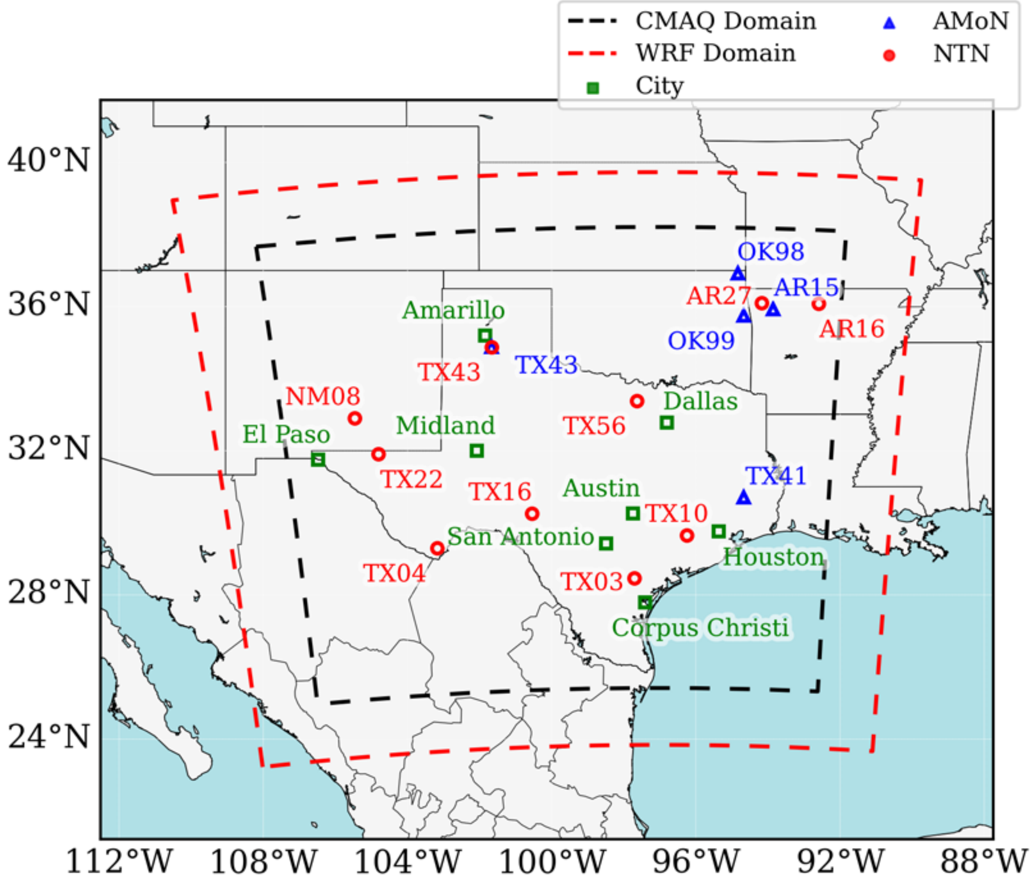
- The posterior model performs substantially better than the prior model across all quarters and on an annual basis in terms of all the measured metrics.

Evaluation

Posterior evaluation with surface measurements

The location of stations

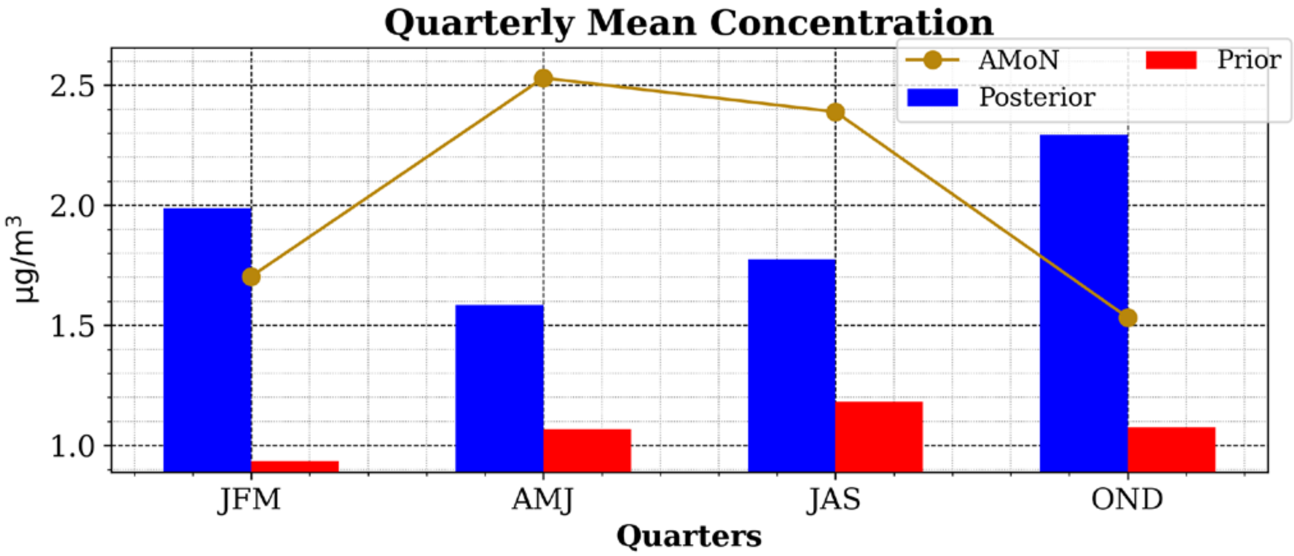
- AMoN: NH_3 surface concentration
- NTN: NH_4^+ wet deposition measurement

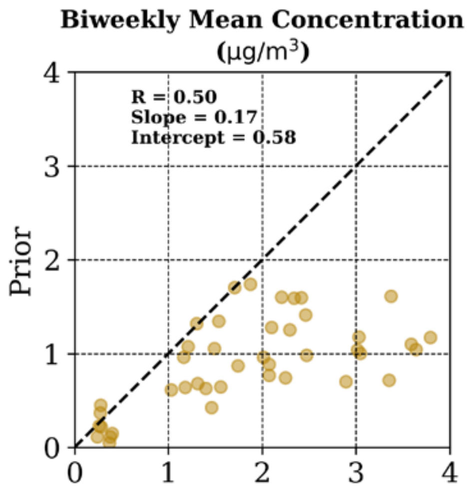


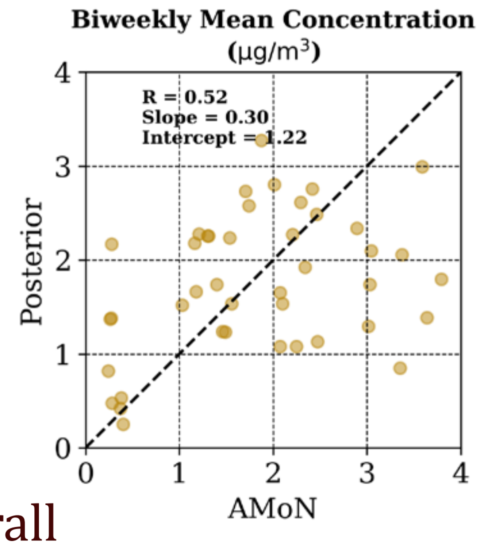
Evaluation

Posterior evaluation with surface measurements

	R	MAE	NMSE	RMSE	IOA
Prior	0.50	1.17	1.46	1.67	0.54
Posterior	0.52	0.92	0.38	1.20	0.67





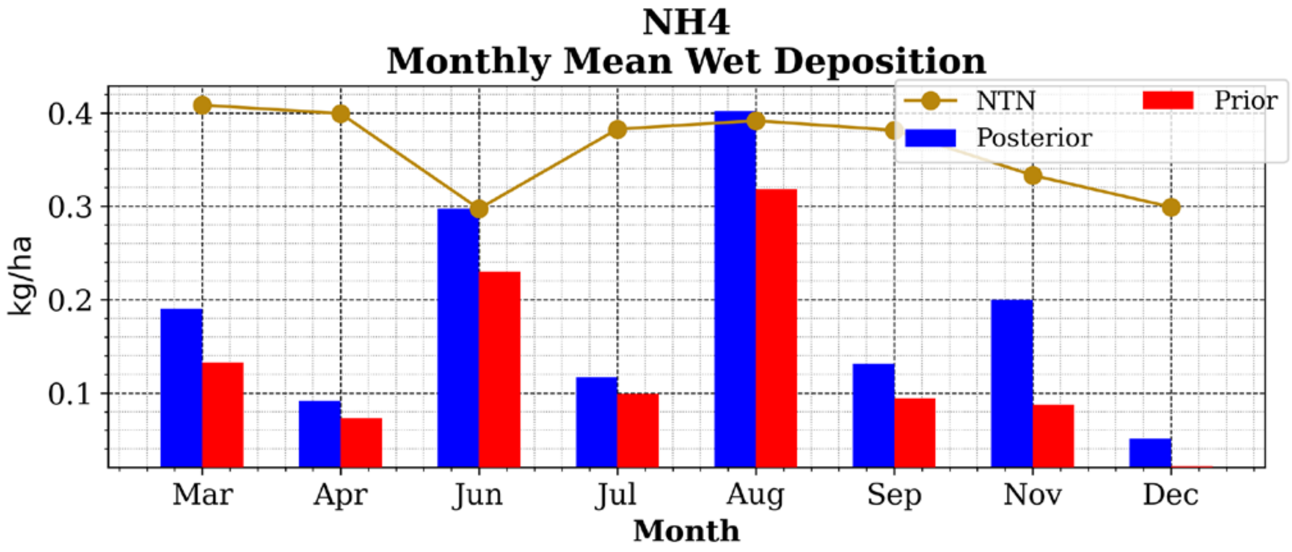


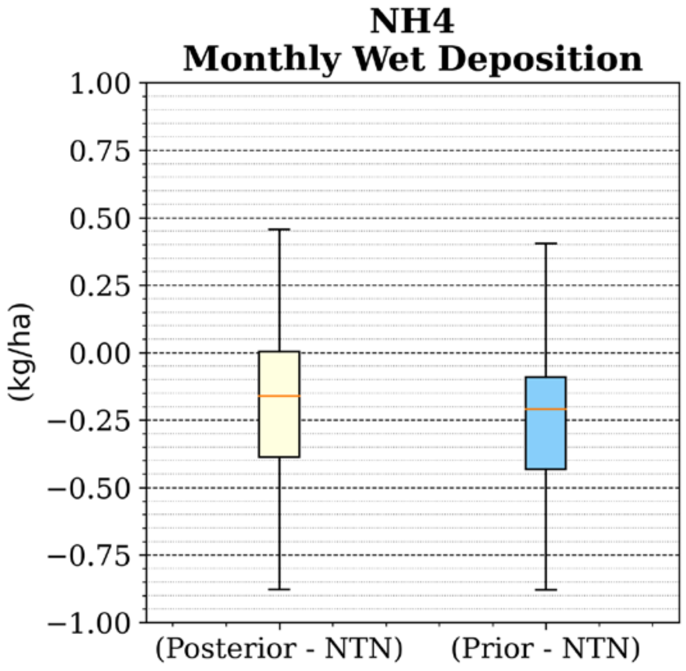
- The table shows that the updated (posterior) NH_3 emissions provide an overall improved performance over the prior NH_3 emissions, offering more accurate predictions and a better fit with observed values.

Evaluation

Posterior evaluation with surface measurements

	MAE	NMSE	RMSE	IOA
Prior	0.28	2.58	0.34	0.40
Posterior	0.26	1.71	0.33	0.37





- The posterior result seems to have improved the MAE, NMSE, and RMSE values, but the IOA has decreased slightly.

Conclusion

- The results showed a noticeable increase in ammonia emission levels is observed over Texas.
- The coastline and extending over the Gulf of Mexico, demonstrates significantly elevated emission levels in comparison to the a-priori emissions.
- The updated emissions derived through our inverse modeling suggest the necessity of a thorough reevaluation of ammonia emissions from the Gulf of Mexico.
- For NH_3 concentrations, the most substantial increase occurred in the Northwestern regions, where values rose between 3-4 ppb, while in the rest of Texas, including most of the northern parts and some eastern areas, the increase ranged between 1-2 ppb.
- For NH_4^+ concentrations, the notable hotspot over the Gulf of Mexico, particularly in the southern parts, warrants further investigation.
- For SO_4^{2-} concentrations, the significant increase in near the Port Arthur and Galveston area emphasizes the potential importance of human activities.
- For NO_3^- concentrations, increases were simulated throughout Texas.
- The posterior evaluation showed that iFDMB is able to improve NH_3 emissions with the reasonable efficiency and performance with the less run time.

Thank you!