# Refining ammonia emissions estimates with satellite-based observations using a novel framework and an air quality model

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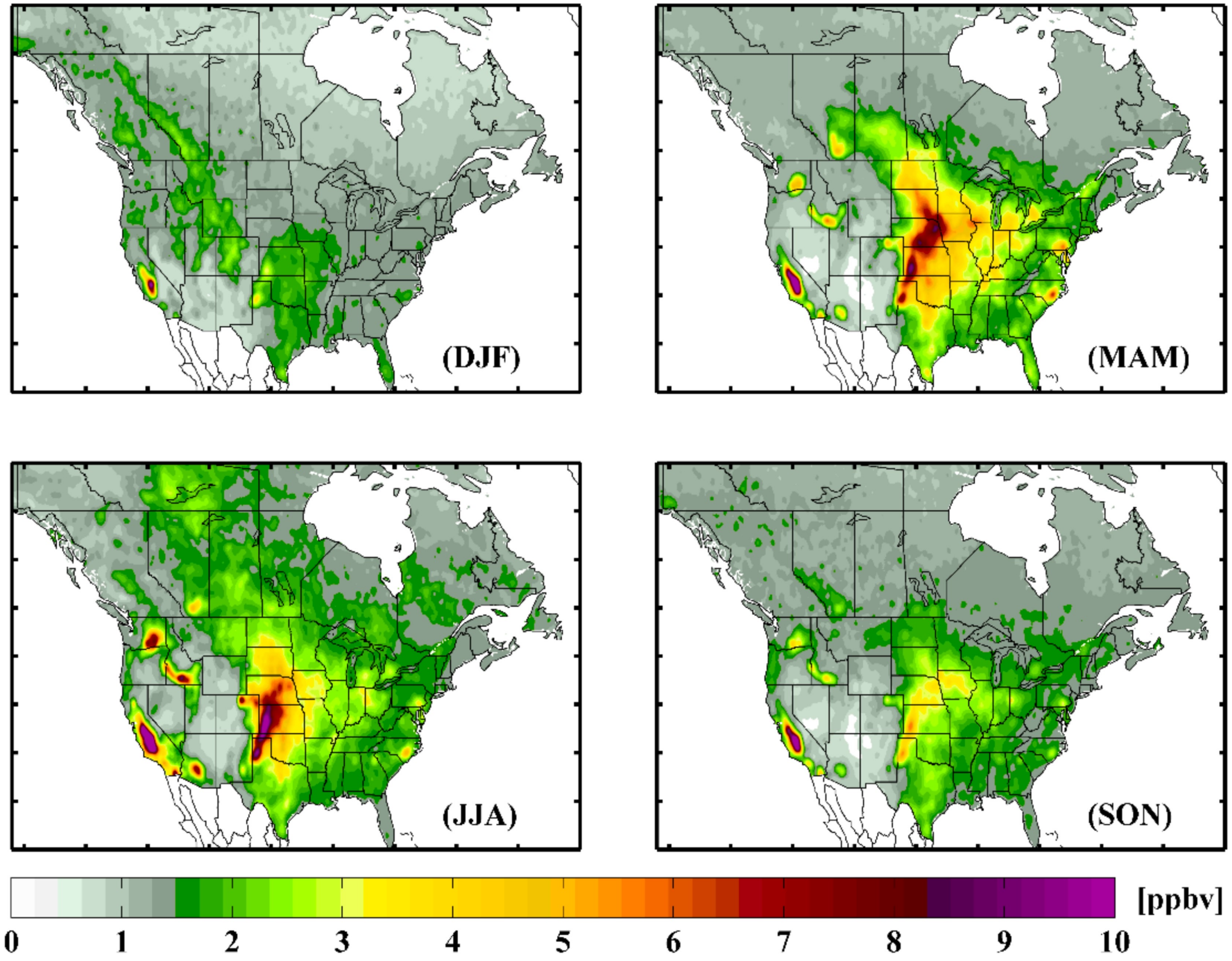
in prep.

# Ammonia in the atmosphere

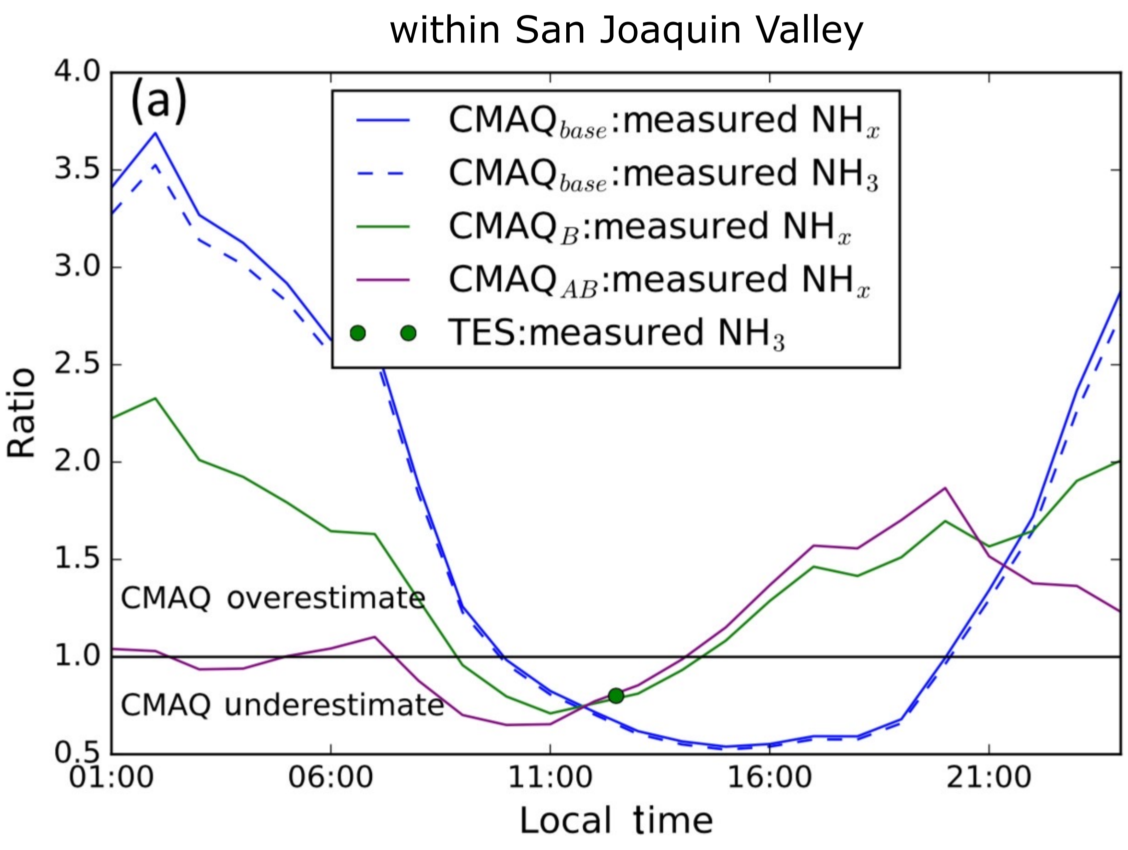
## 5-year mean of surface $NH_3$ from CrIS (2013-2017)




Shephard et al., ACP, 2020

# Challenges modeling ammonia

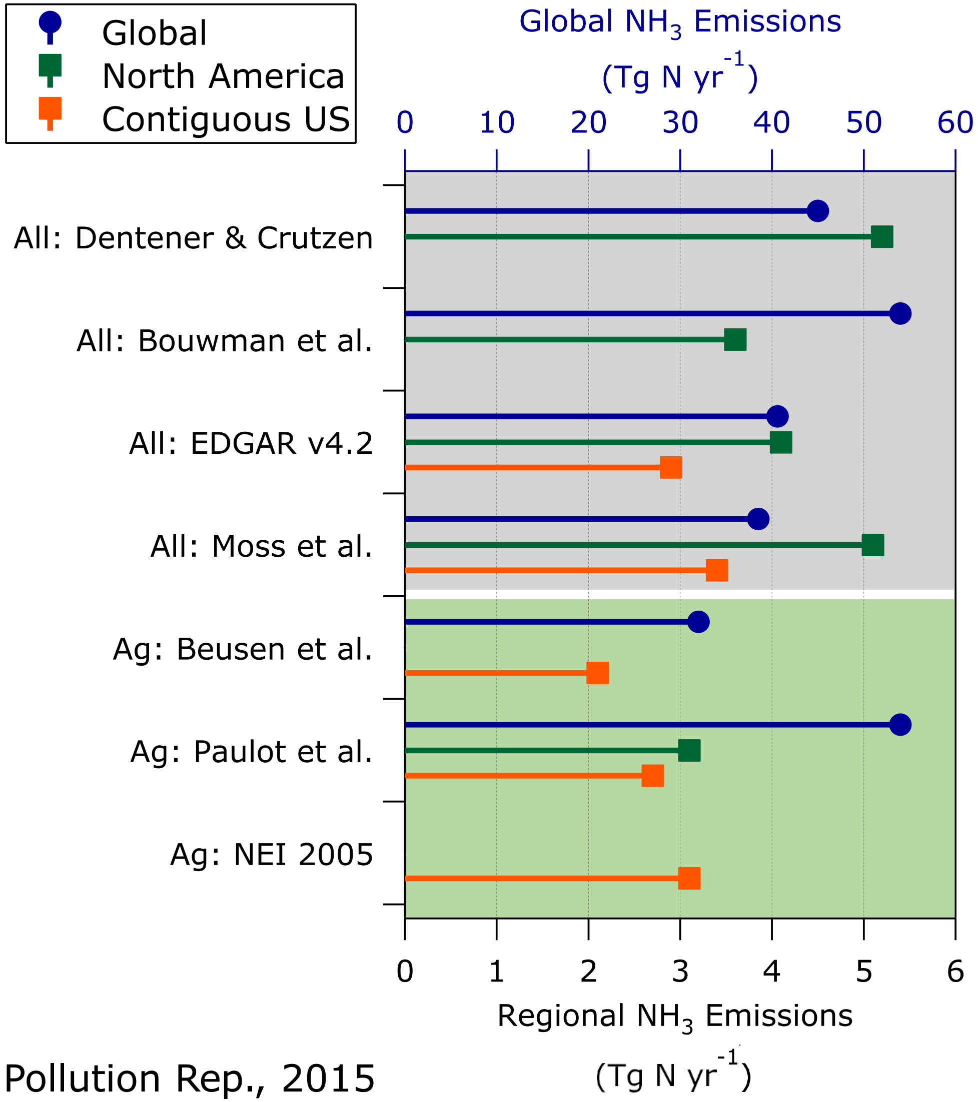
- Emissions estimation from variable sources
- Volatility of gas
- Potential for bidirectional flux




Lonsdale et al., ACP, 2017

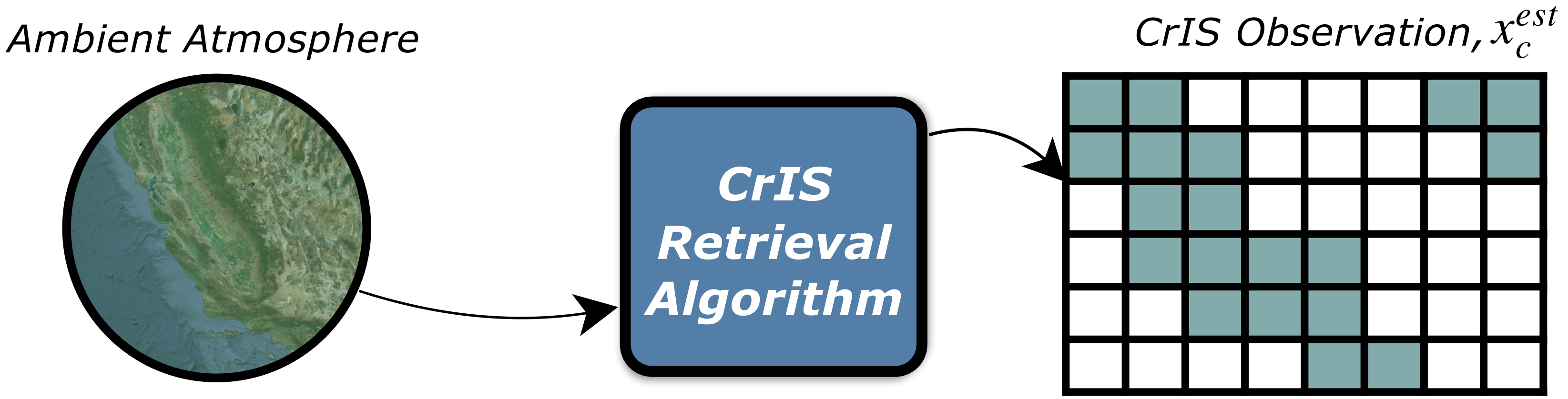
# Uncertainty in ammonia emissions

- Between 25% and 50% spread in emissions estimates exists across inventories.
- Select agricultural contributions are equivalent to other estimates of total emissions.




Zhu et al., Curr. Pollution Rep., 2015

# CrIS satellite-based observation



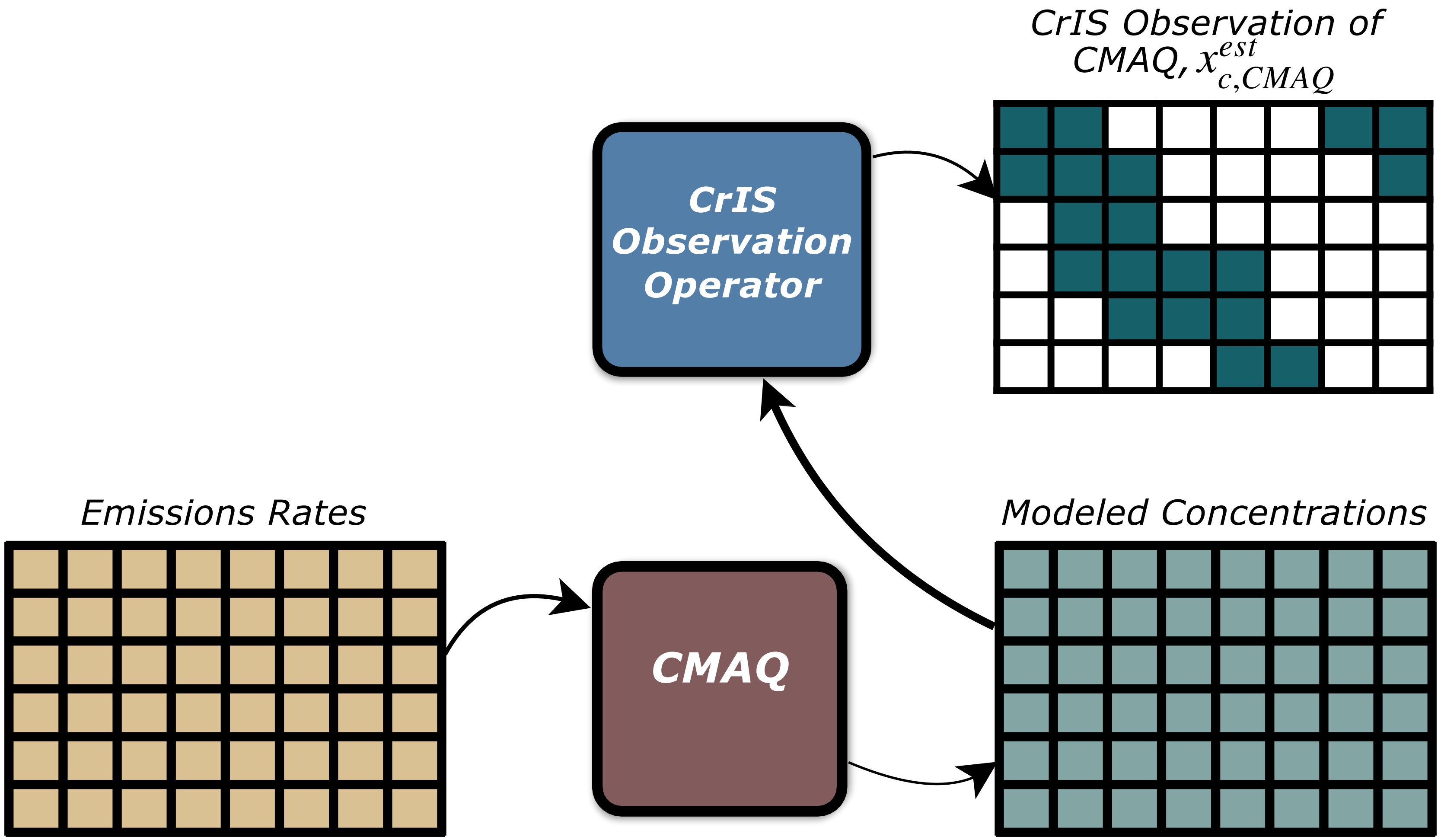

$$x_c^{est} = x_a + A(x_c^{mapped} - x_a)$$

where $x_a$ is a profile based on
clean, moderate, or polluted conditions



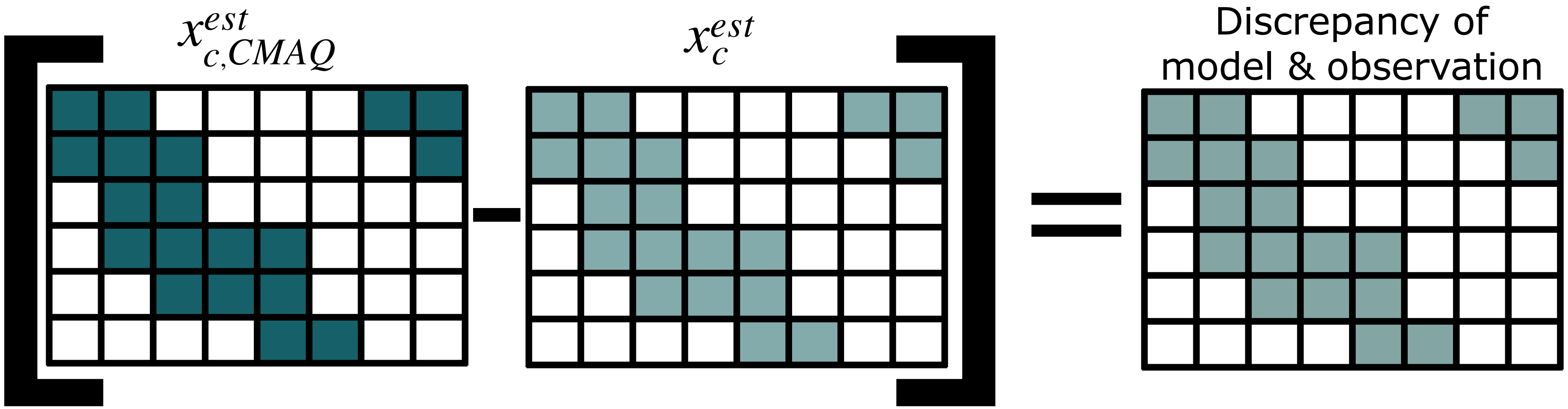
Shephard and Cady-Pereira, Atmos. Meas. Tech., 2015

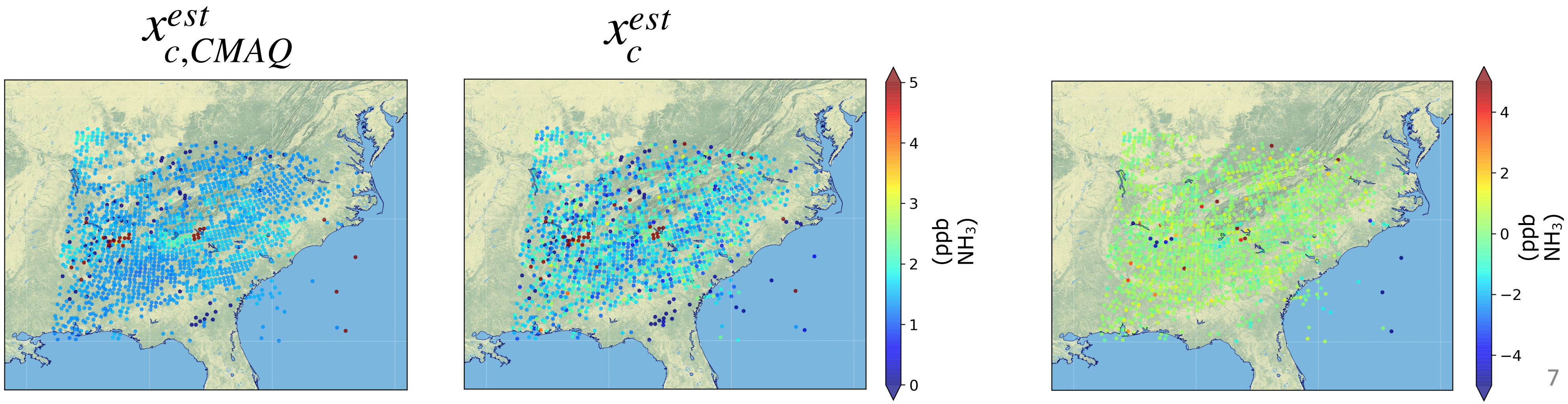
# Simulating CrIS observations



# Comparing CrIS observations of ambient & CMAQ ammonia

$$J = \frac{1}{2}\sum\left(x^{est}_{c,CMAQ} - x^{est}_{c}\right)^{T} S^{-1}_{obs}\left(x^{est}_{c,CMAQ} - x^{est}_{c}\right) + \frac{1}{2}\gamma(\sigma - \sigma_a)^{T} S^{-1}_{a}(\sigma - \sigma_a)$$

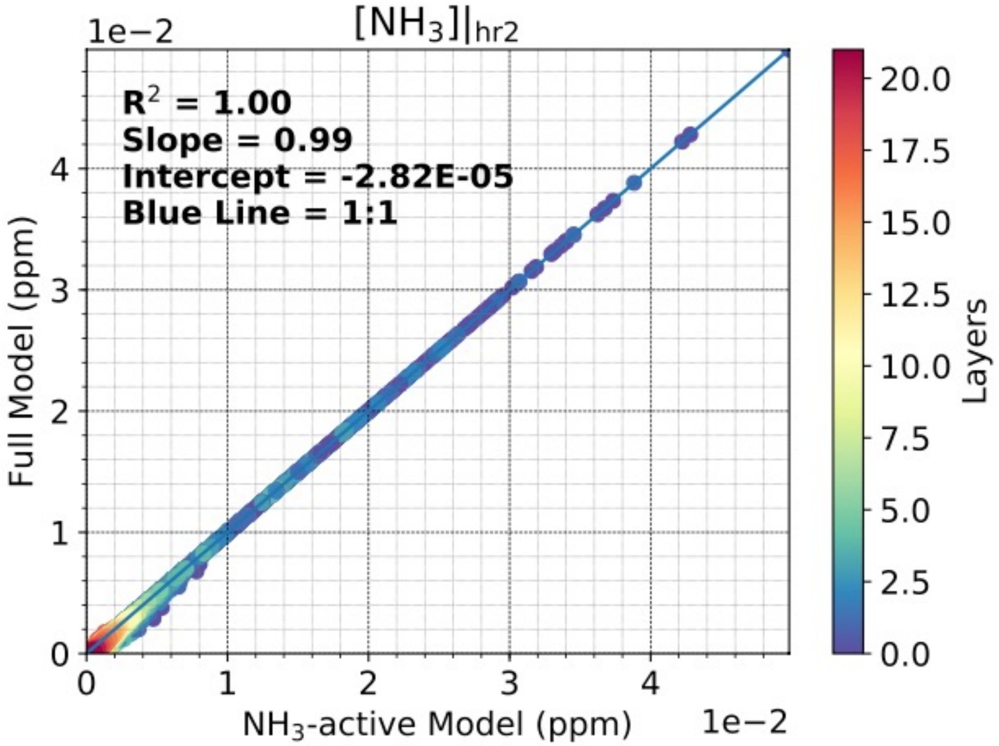


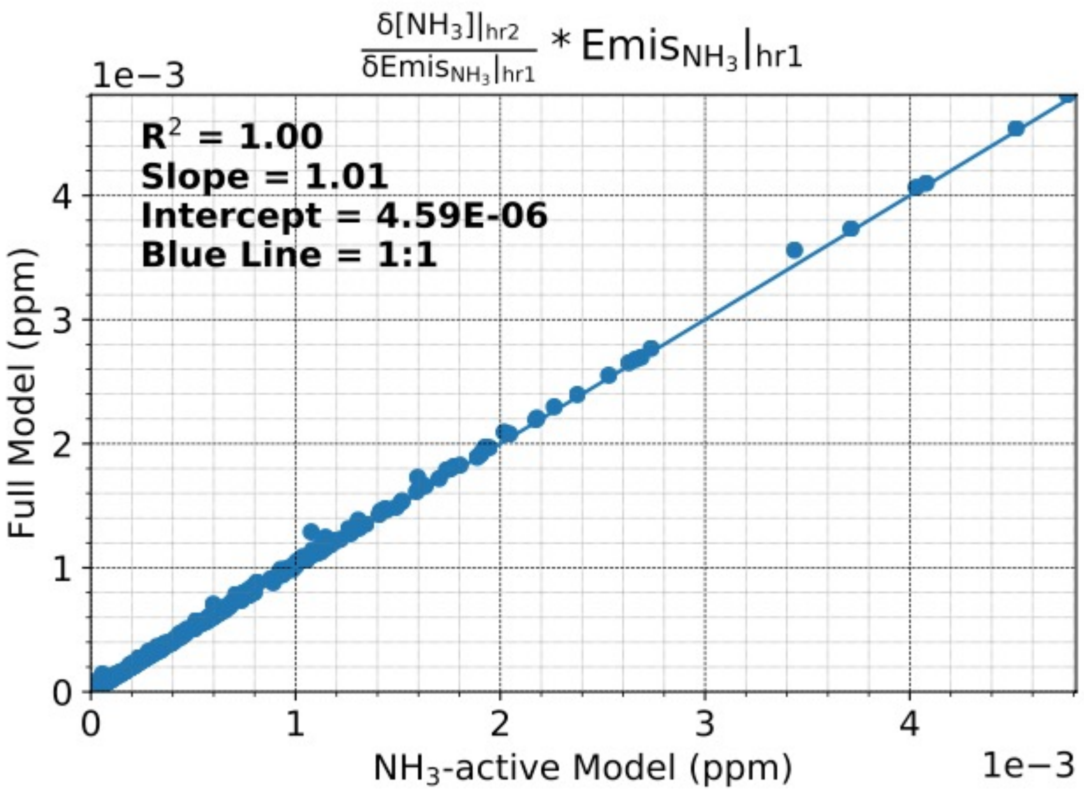



# Development of an ammonia-active CMAQ-adjoint model

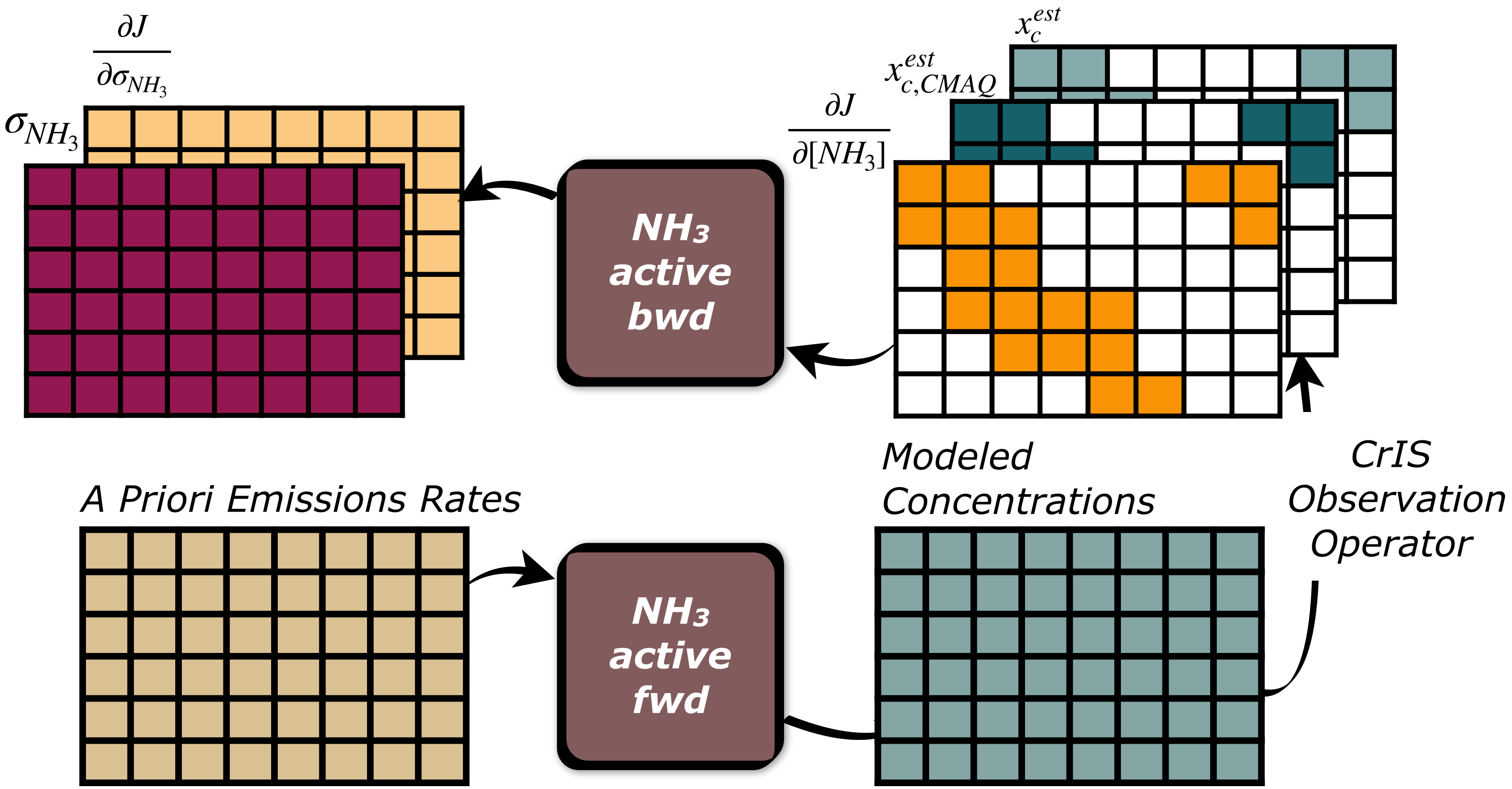
run time changes

- Forward sweep:
  $NH_3$-active forward = 67% full forward
- Backward sweep:
  $NH_3$-active backward = 8% full backward



# Refining emissions with CrIS observations



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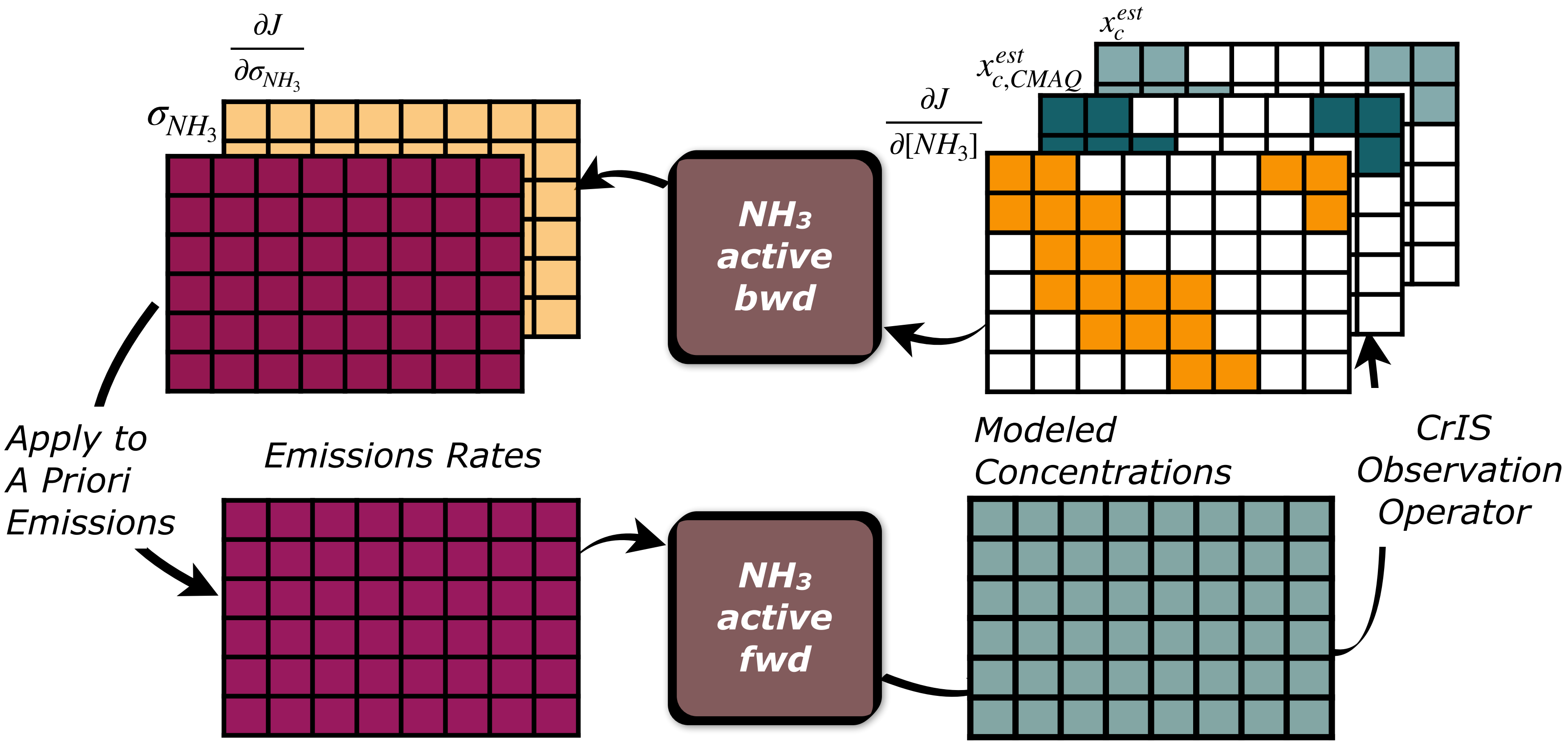
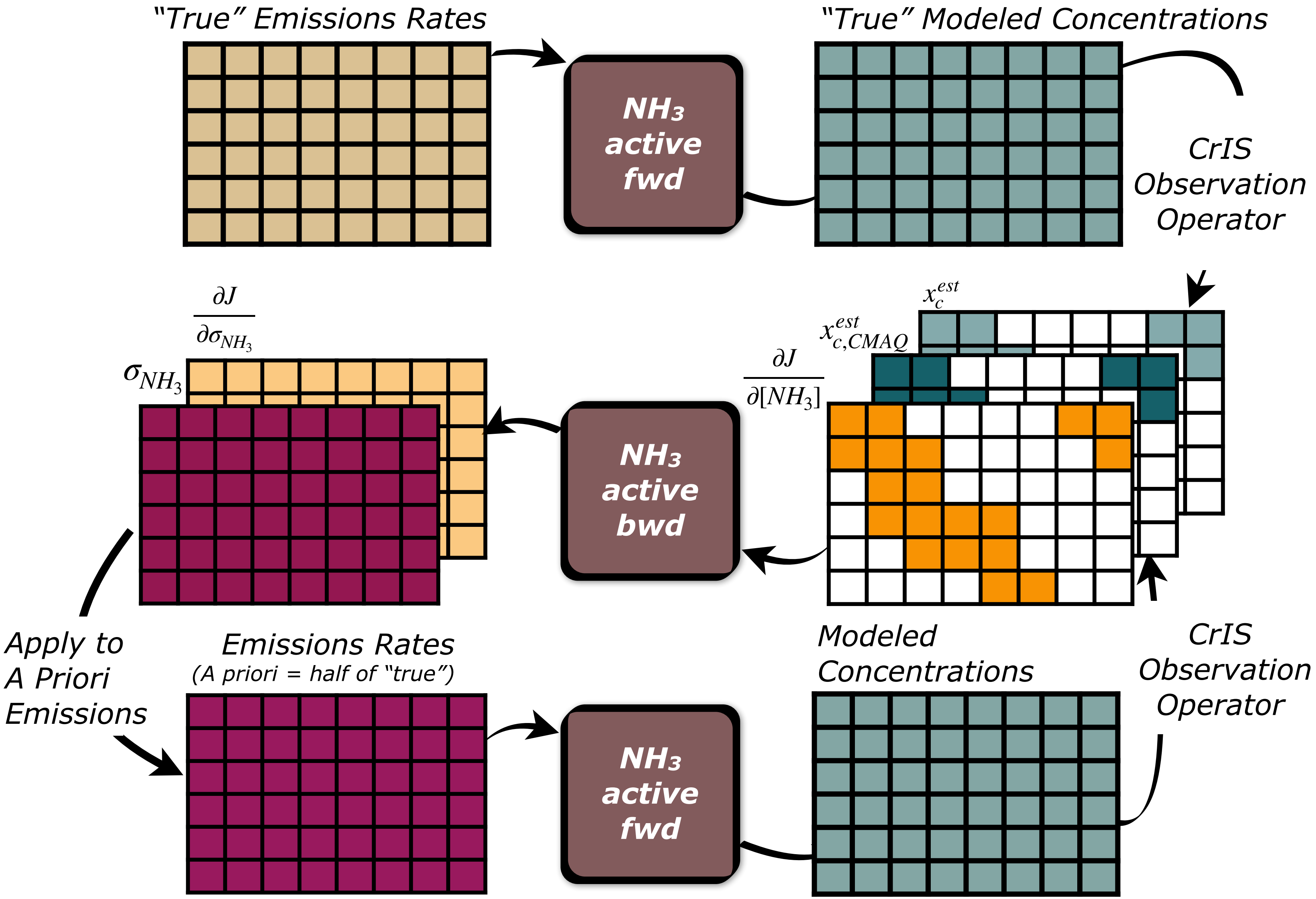


# Observing system simulation experiment (OSSE)



# Py4D-Var OSSE results

- Modeling domain: 12-km*12-km resolution Georgia benchmark domain
- Three days simulation (06/10/2007 – 06/12/2007)
- CrIS data v1.5, 2016
- 16 cores
- 22 minutes to finish one forward sweep and one backward sweep for a line-search
- Up to 20 line-searches for an iteration
- Time consuming

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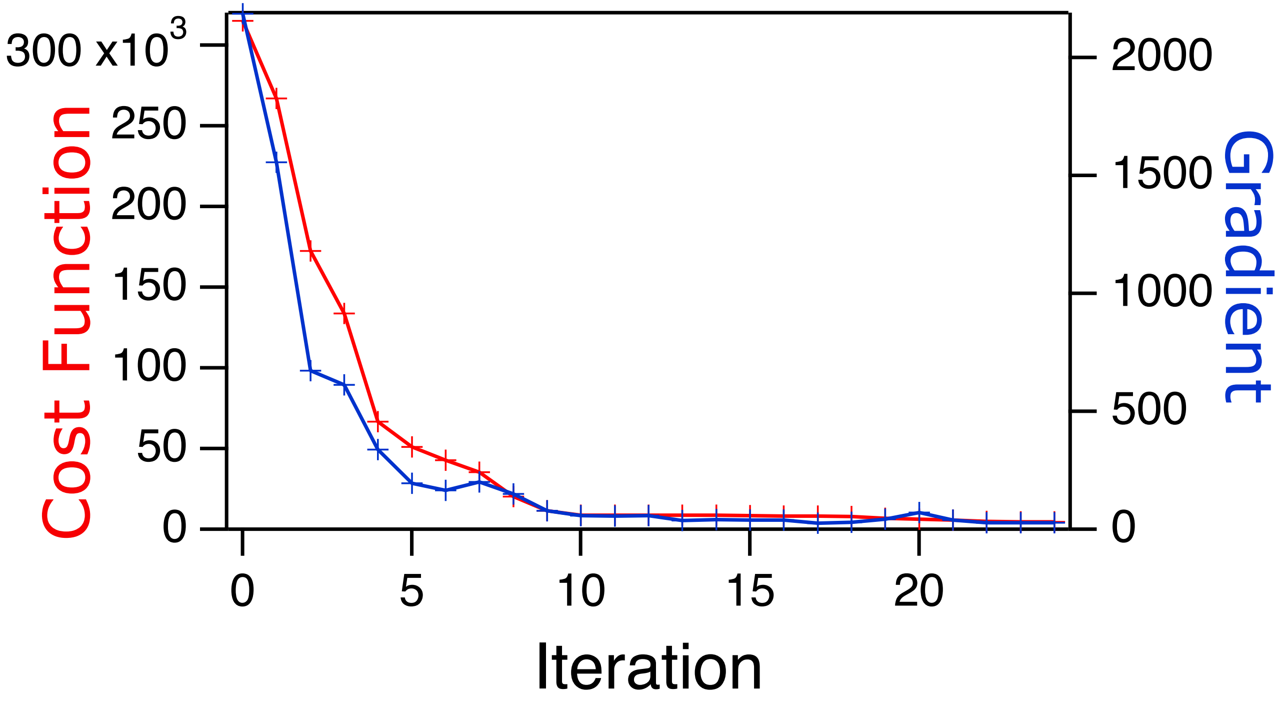
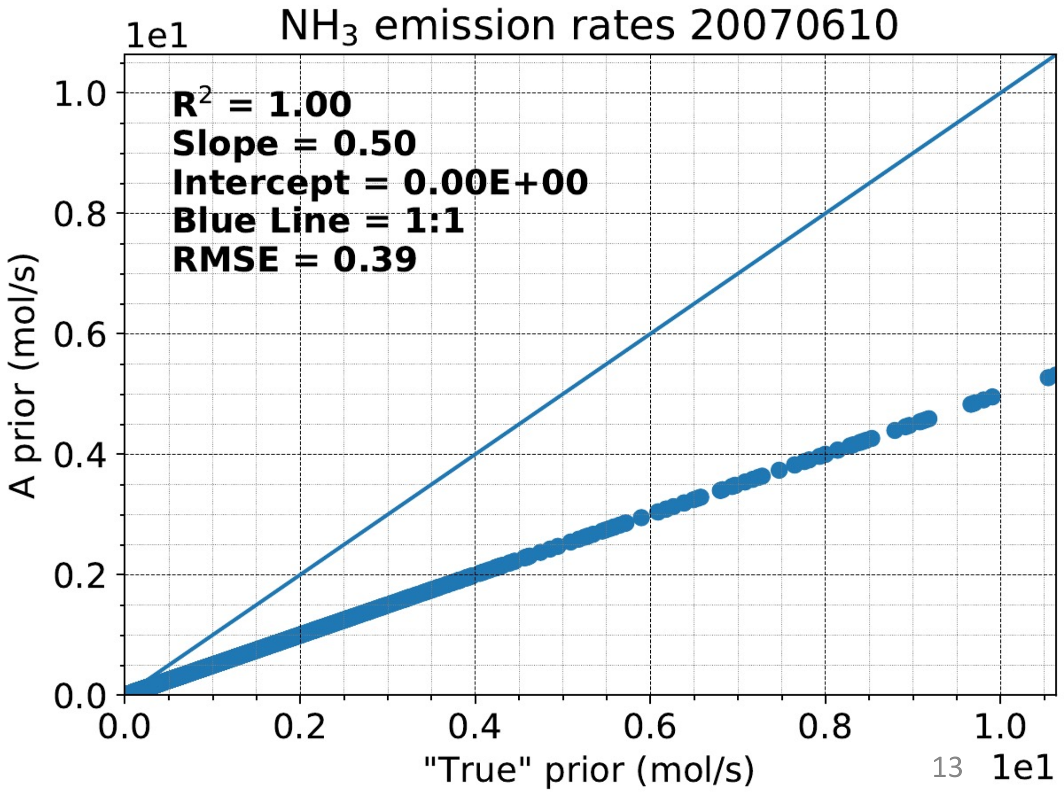
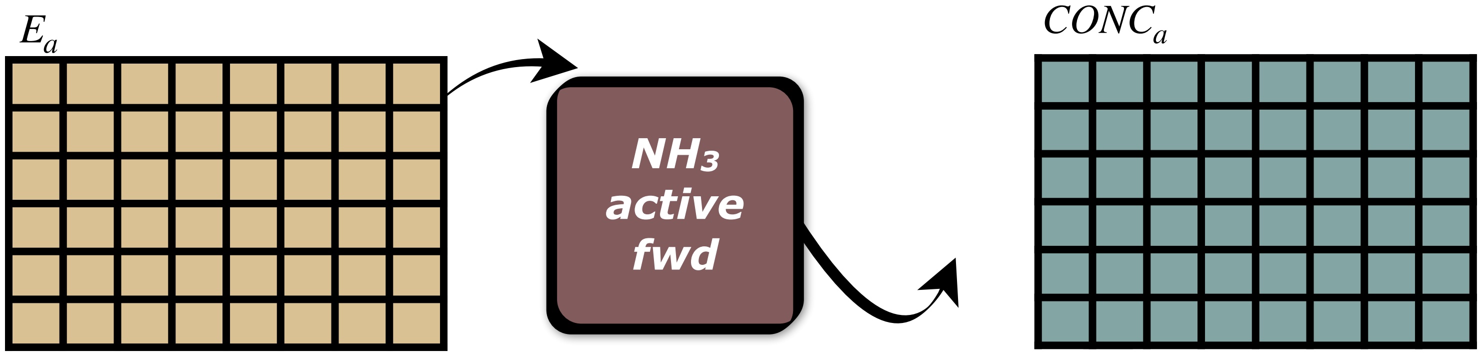


# Finite difference mass balance (FDMB)




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

*Emissions Rates*

*Modeled Concentrations*

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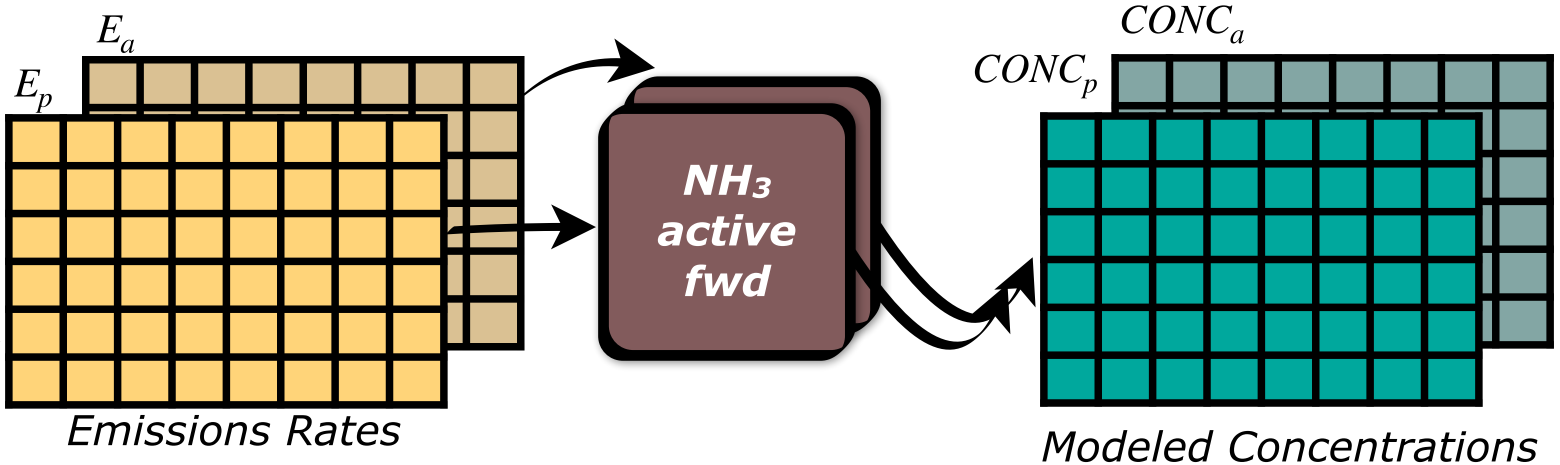


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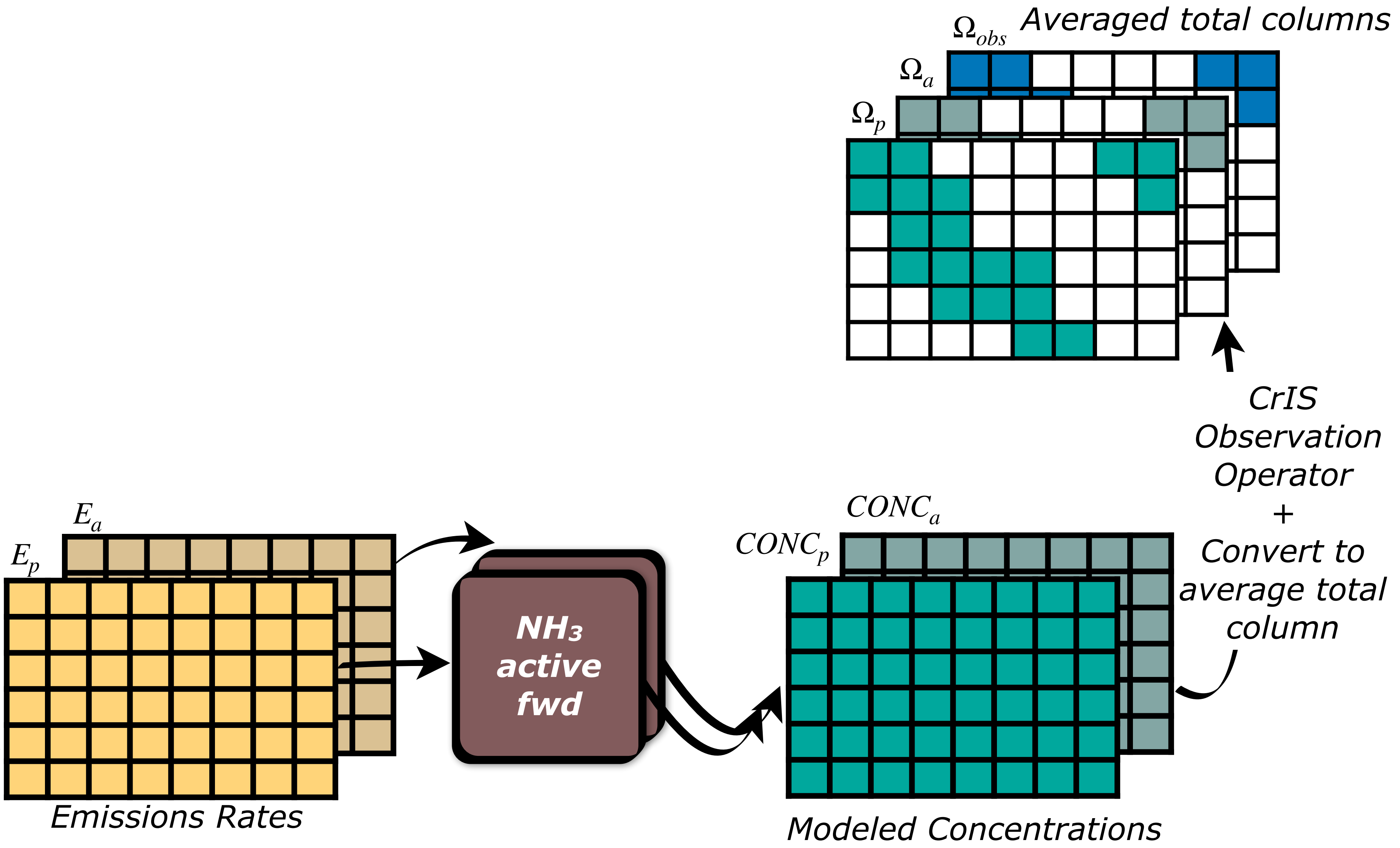


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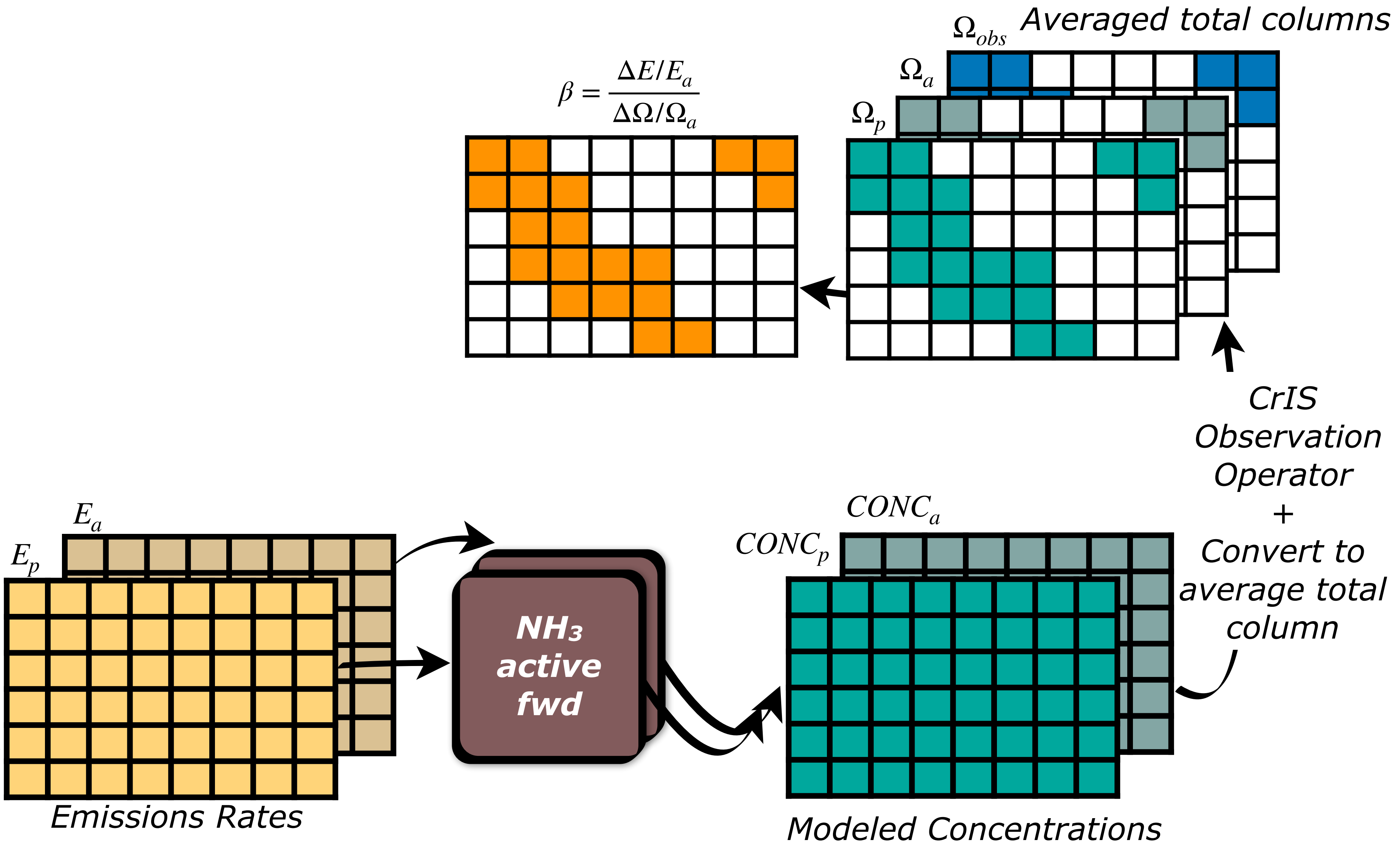


Lamsal et al., GRL, 2011;
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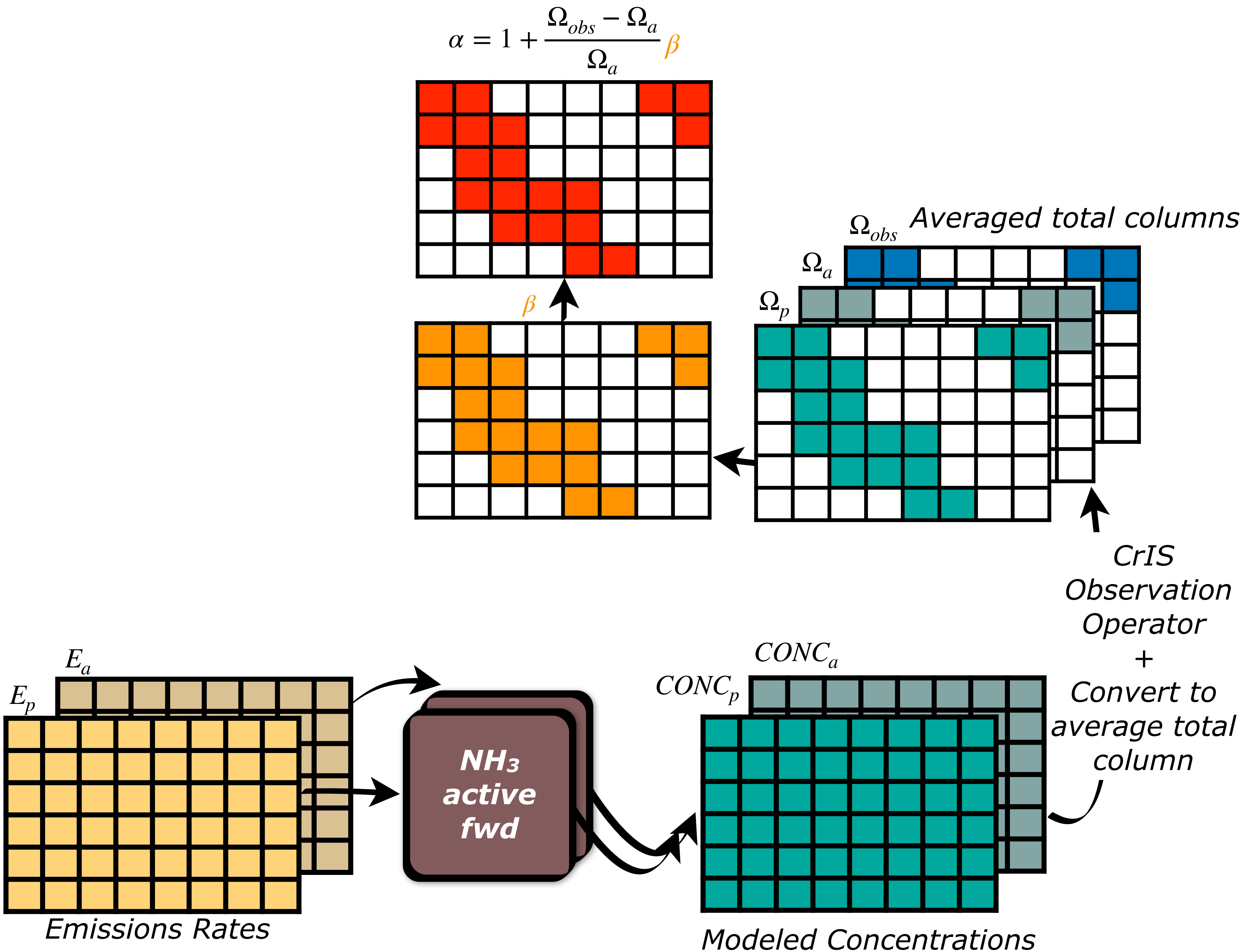


Lamsal et al., GRL, 2011;
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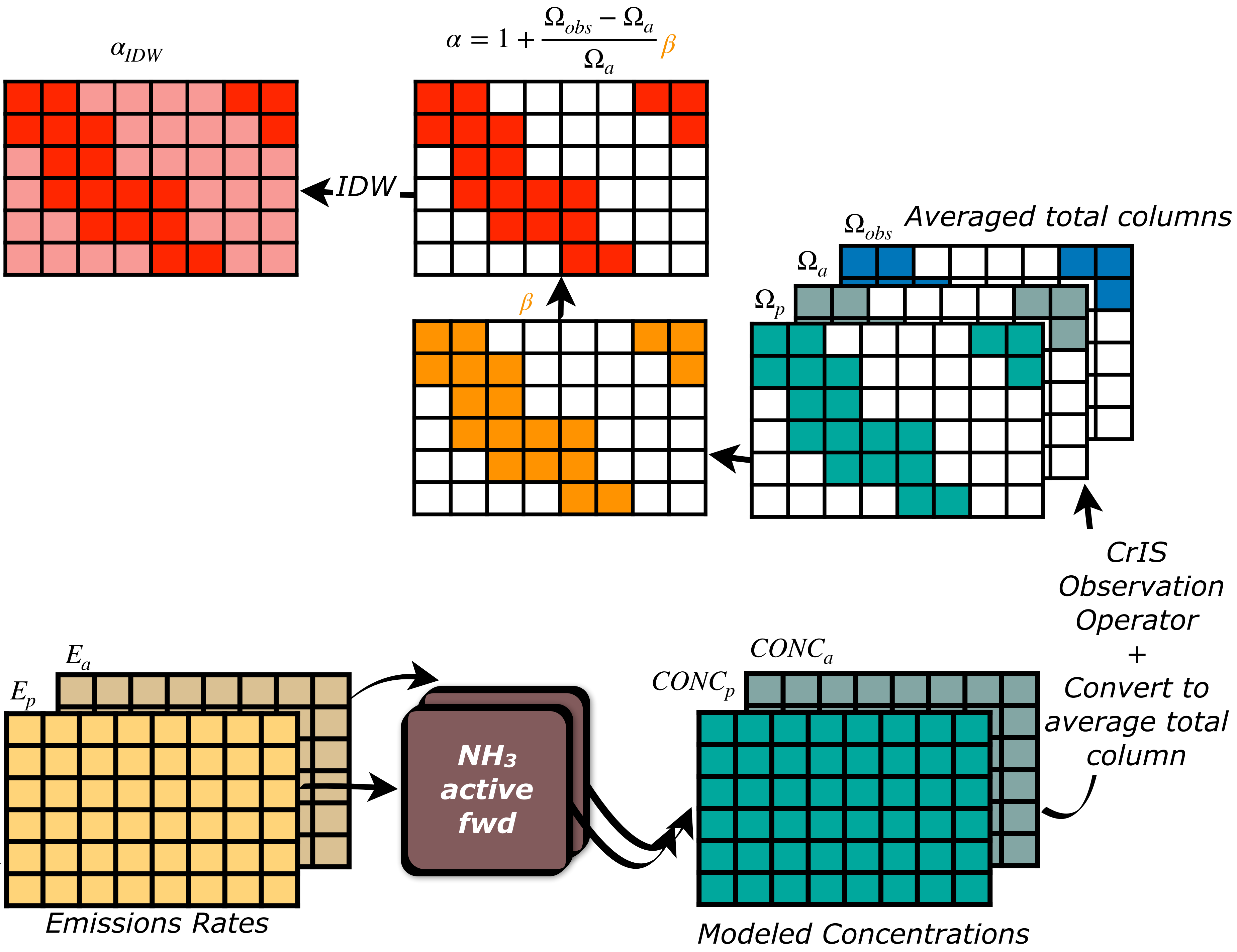


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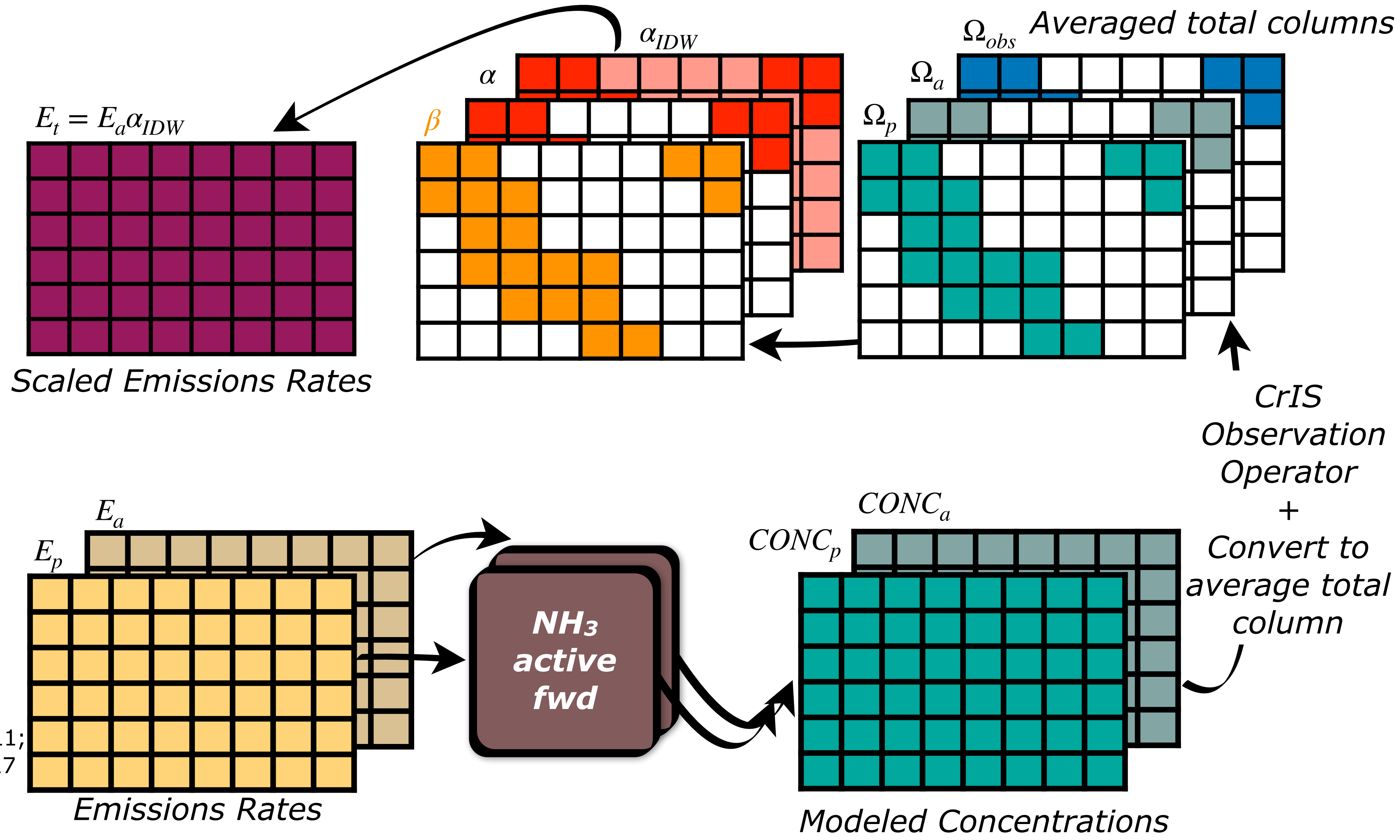


Lamsal et al., GRL, 2011;
Cooper et al., JGR, 2017

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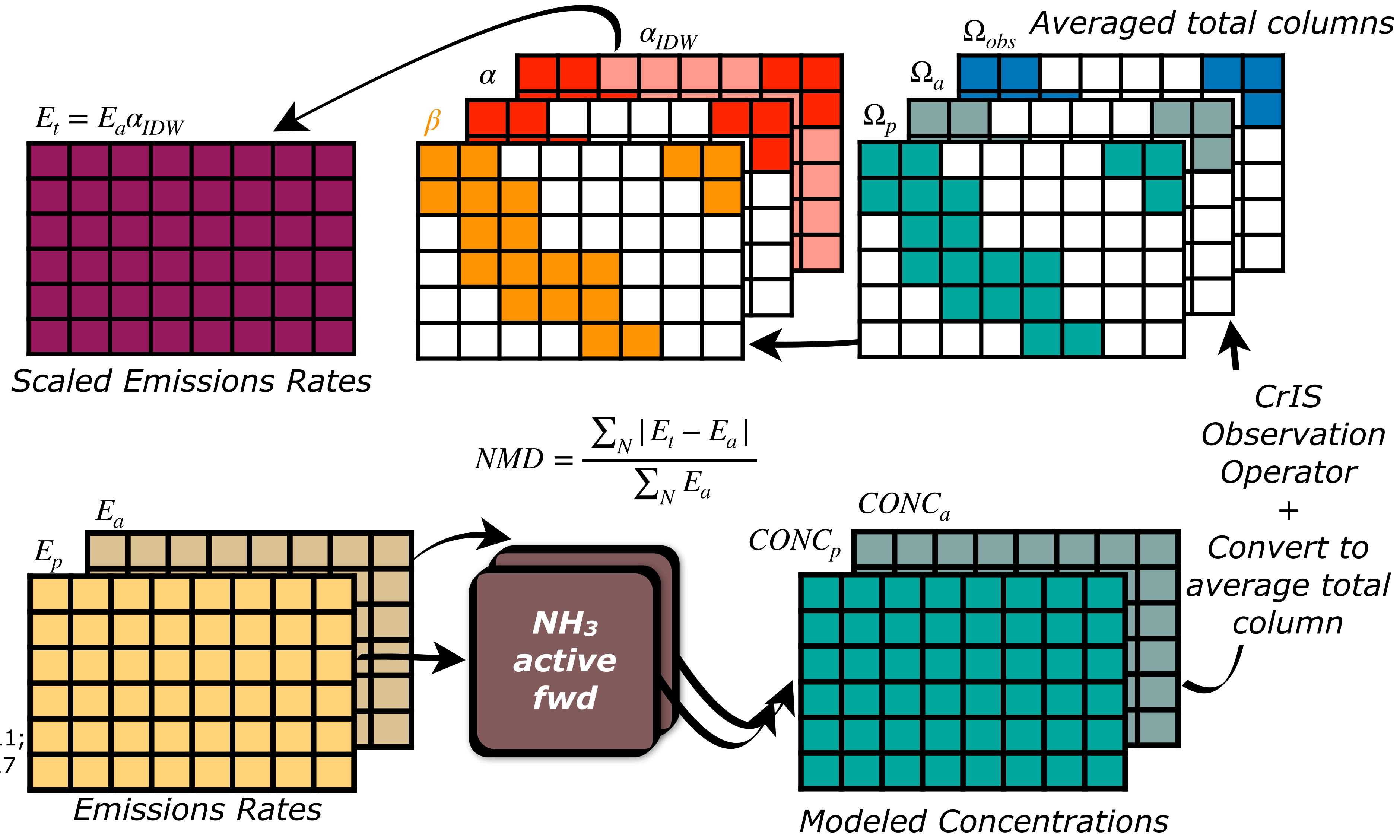


Lamsal et al., GRL, 2011;
Cooper et al., JGR, 2017

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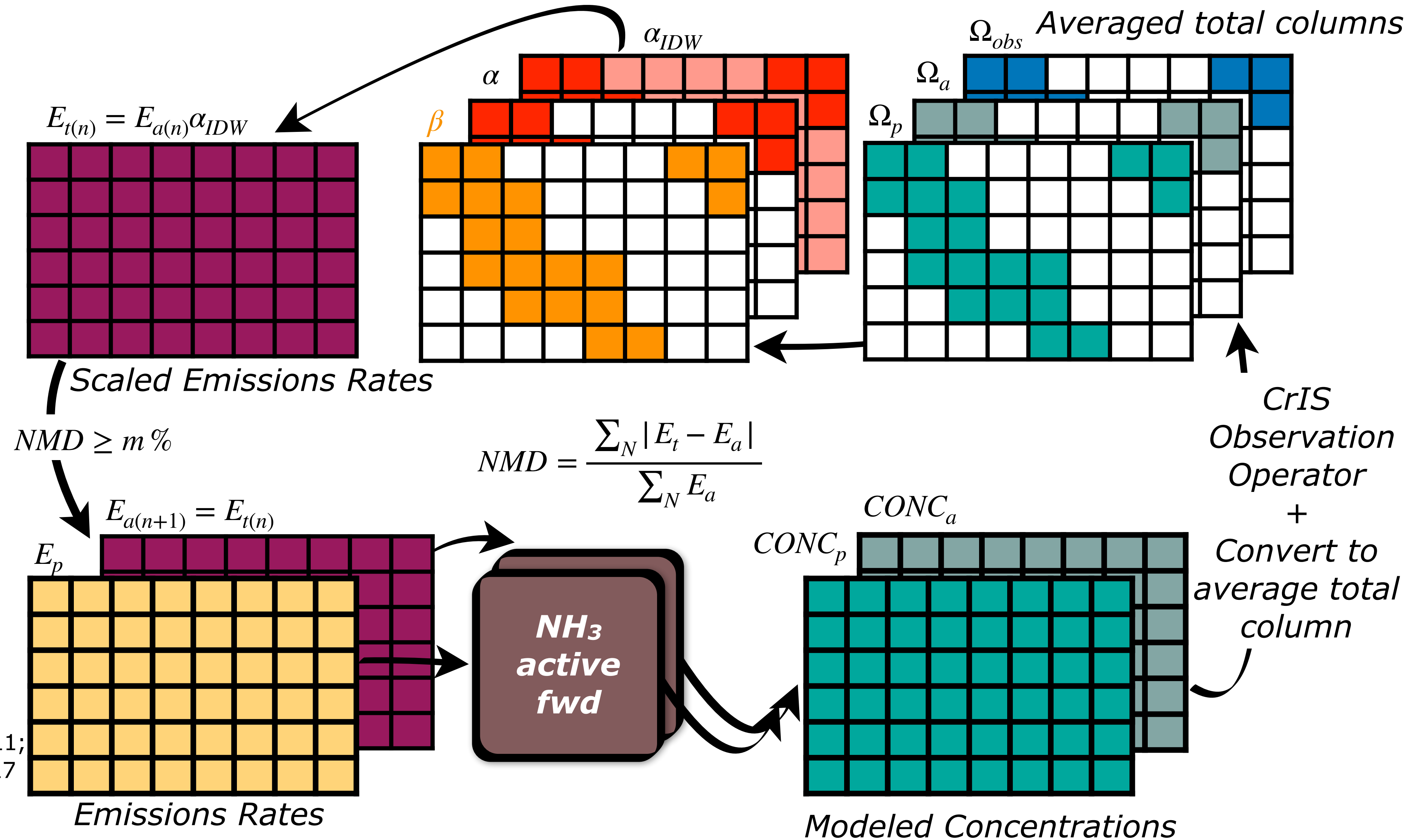
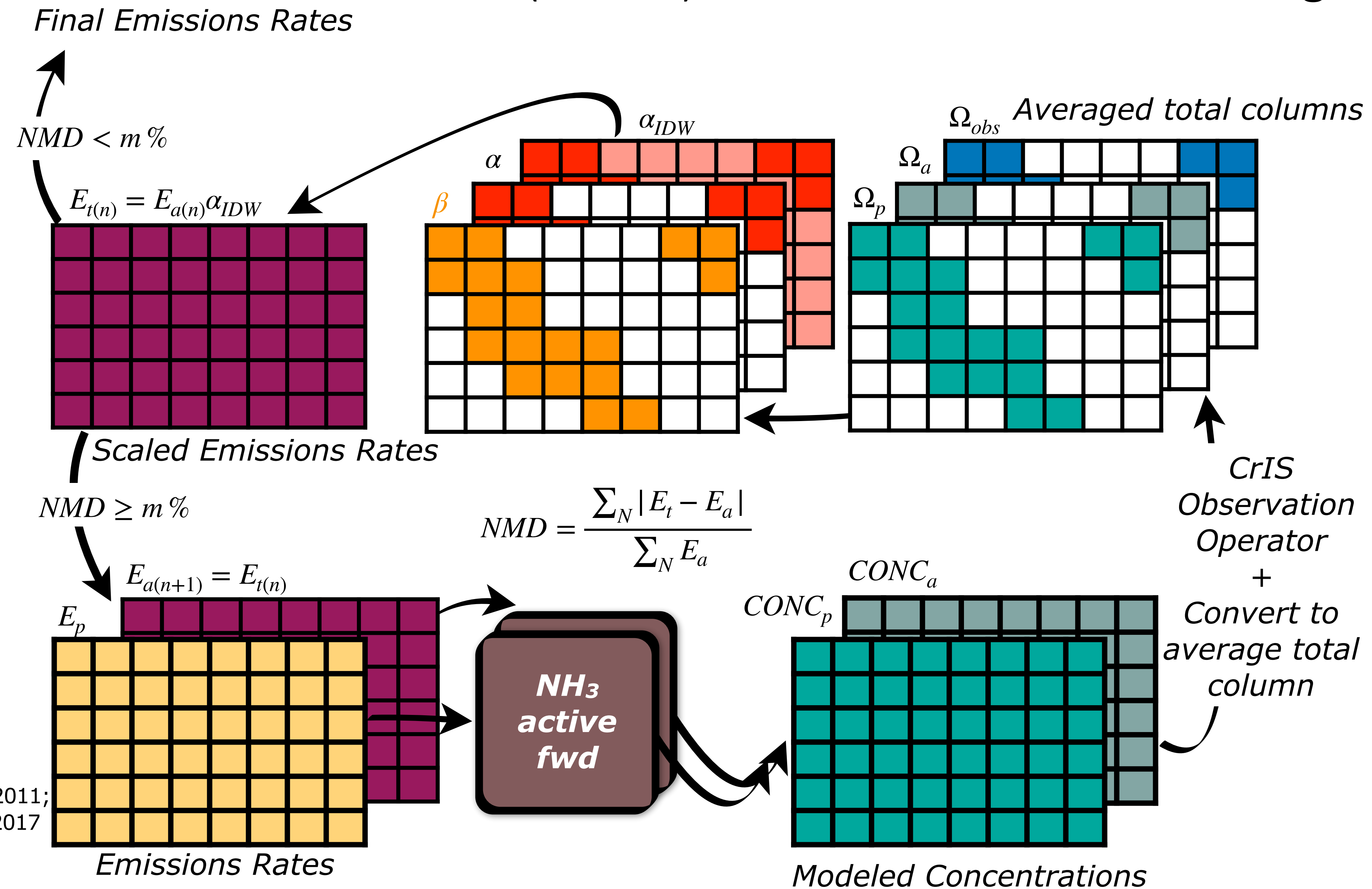



Lamsal et al., GRL, 2011;
Cooper et al., JGR, 2017

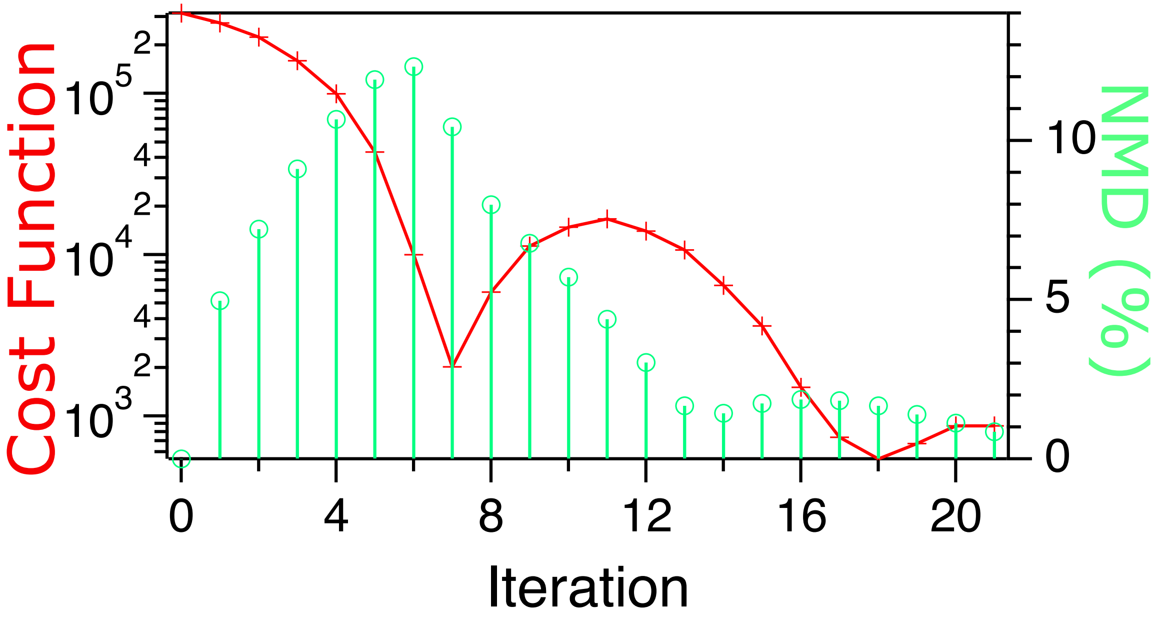
# Finite Difference Mass Balance (FDMB) with Inverse Distance Weighting (IDW)

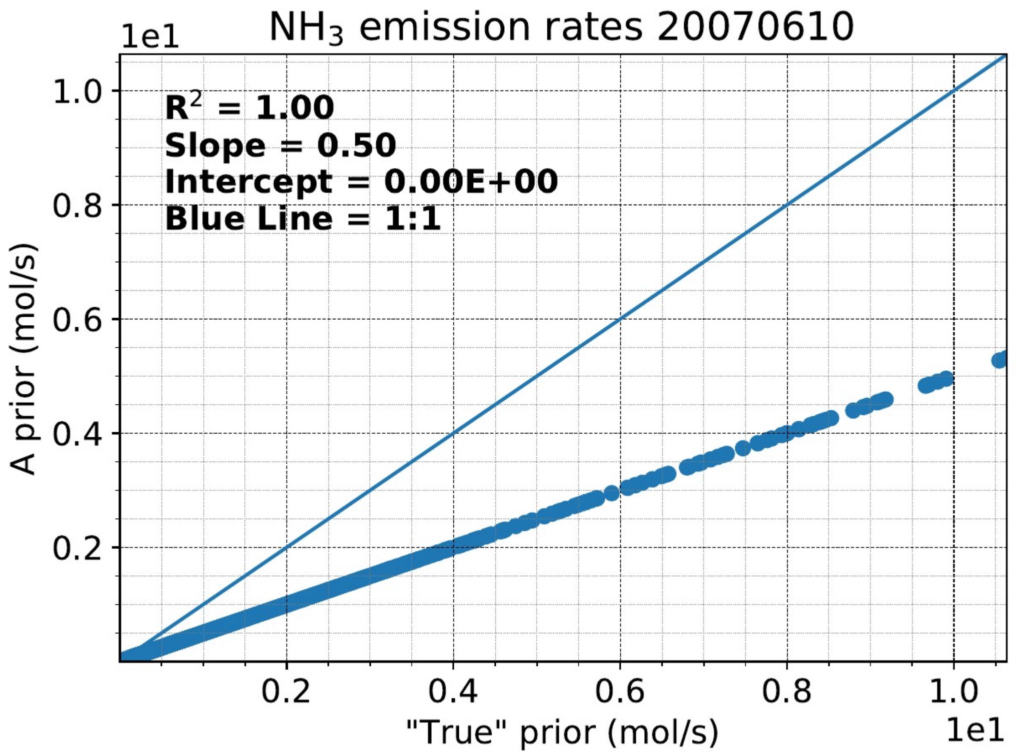



Lamsal et al., GRL, 2011;
Cooper et al., JGR, 2017

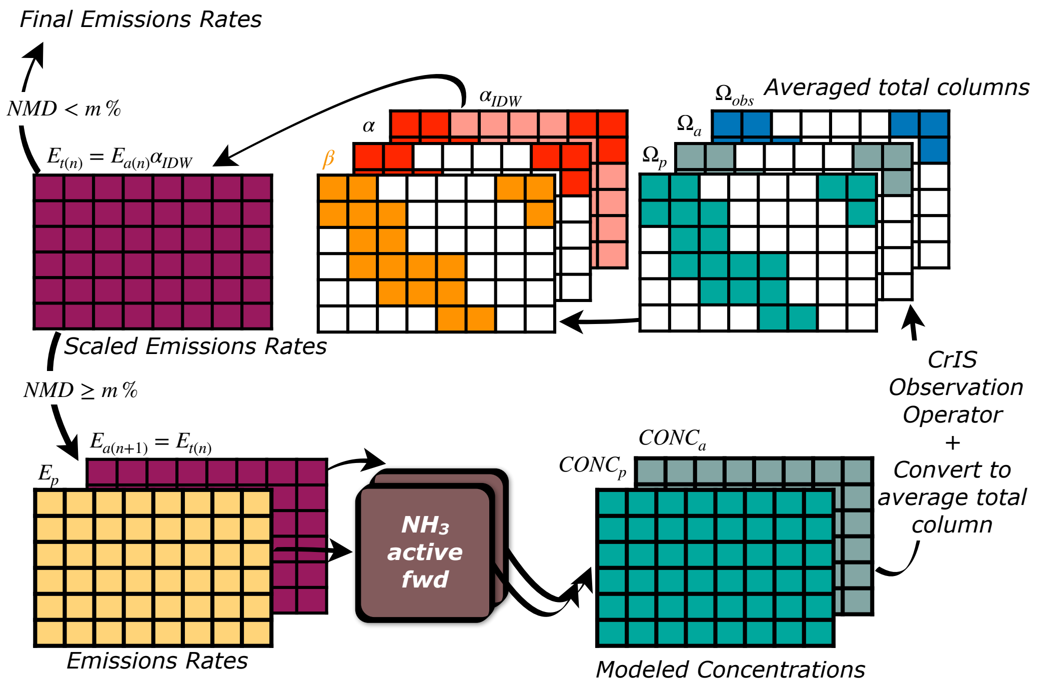
# FDMB with IDW OSSE results

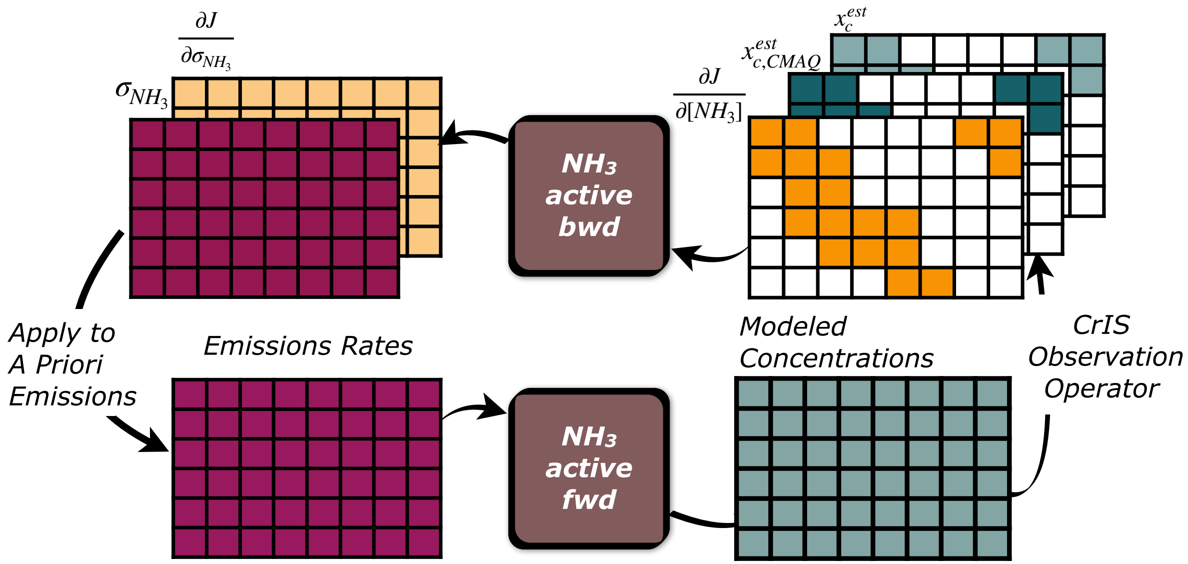
- Modeling domain: 12-km*12-km resolution Georgia benchmark domain
- Three days simulation (06/10/2007 – 06/12/2007)
- CrIS data v1.5, 2016
- 16 cores
- 10 minutes to finish two forward sweeps for an iteration
- Time saving



# FDMB vs. 4D-Var



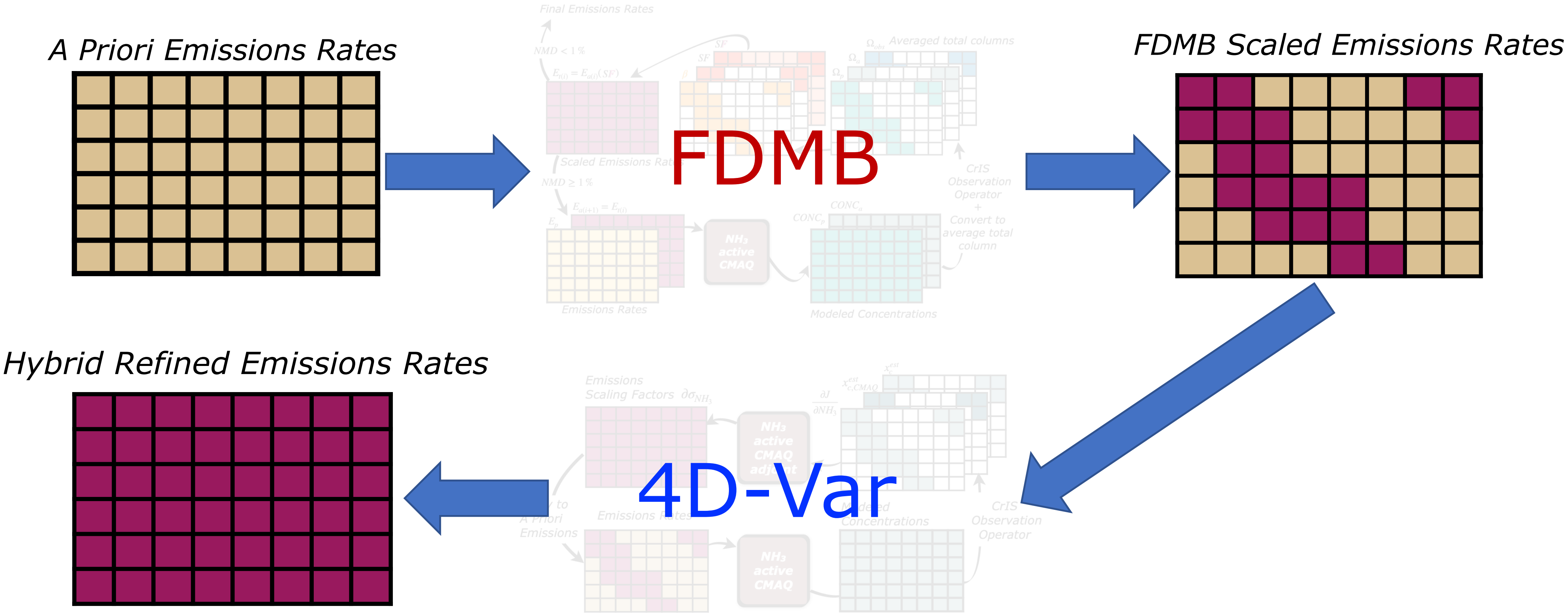




- Fast: two forward sweeps per iteration
- Lower accuracy: scaling for grid cells without observations are estimated by IDW, which tends to introduce uncertainties
- Local influence: assumes a linear relationship between local emissions and concentrations

- Slow: one forward sweep and one backward sweep per line-search, multiple line-searches per iteration
- Higher accuracy: constrains emissions in the whole domain using heterogeneous sensitivities
- Grid-to-grid transportation

# FDMB + 4D-Var hybrid framework

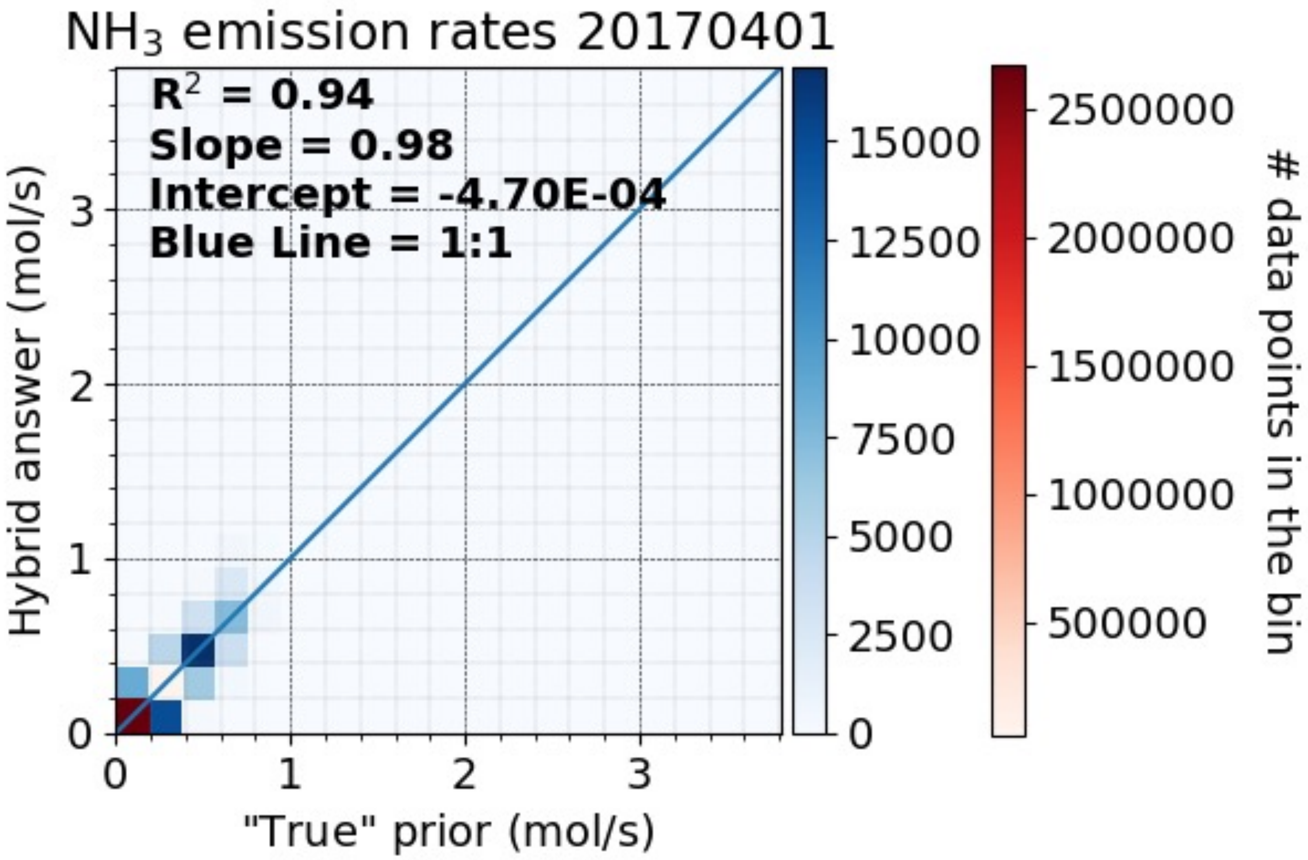
$$J = \frac{1}{2}\sum\left(x^{est}_{c,CMAQ} - x^{est}_{c}\right)^{T} S^{-1}_{obs}\left(x^{est}_{c,CMAQ} - x^{est}_{c}\right) + \frac{1}{2}\gamma(\sigma - \sigma_a)^{T} S_a^{-1}(\sigma - \sigma_a)$$

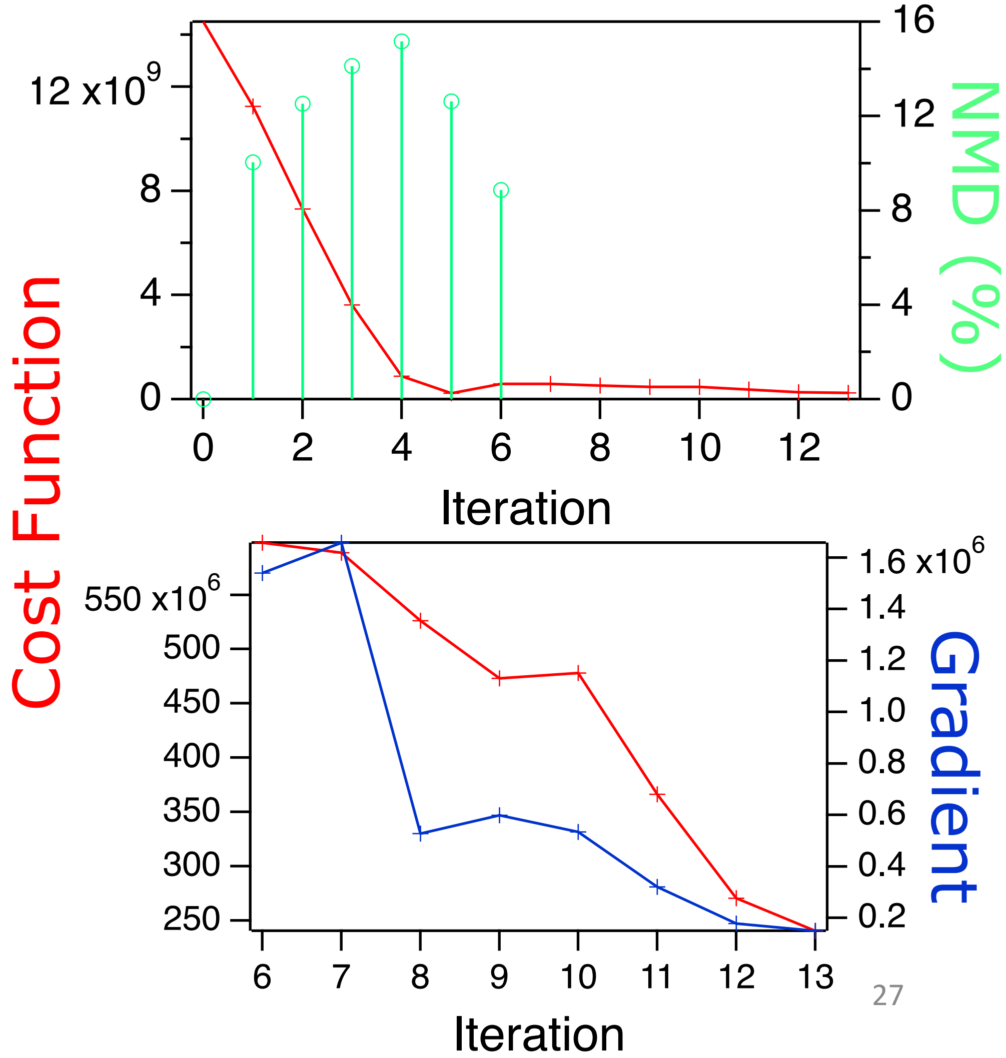



Li et al., JGR Atmospheres., 2019

# FDMB + 4D-Var hybrid framework OSSE

- Modeling domain: 12-km*12-km resolution CONUS domain
- Six days simulation (04/01/2017 – 04/06/2017)
- CrIS data v1.5, 2017



# Future work

- Assimilating CrIS v1.6 observations (The first weeks of April through June 2017 are selected for assimilation)
- Determining the $\gamma$ regulation parameter in the cost function through an L-curve approach
- Evaluating posterior modeled concentrations against independent surface measurements

# Conclusions

- We designed/evaluated a hybrid framework of FDMB with IDW and Py4DVar
- Py4DVar performs well but it is time consuming especially on high-resolution large domain applications
- FDMB is at least two times faster than Py4DVar
- A hybrid approach of the two can take advantage of the shorter run time of the FDMB and still using 4DVar to do the refinement at the end

# Acknowledgements


- CMAQ Adjoint Development Team
- NASA New Investigator Program Award #80NSSC18K1302


**Thanks!**

**Questions? Comments?**