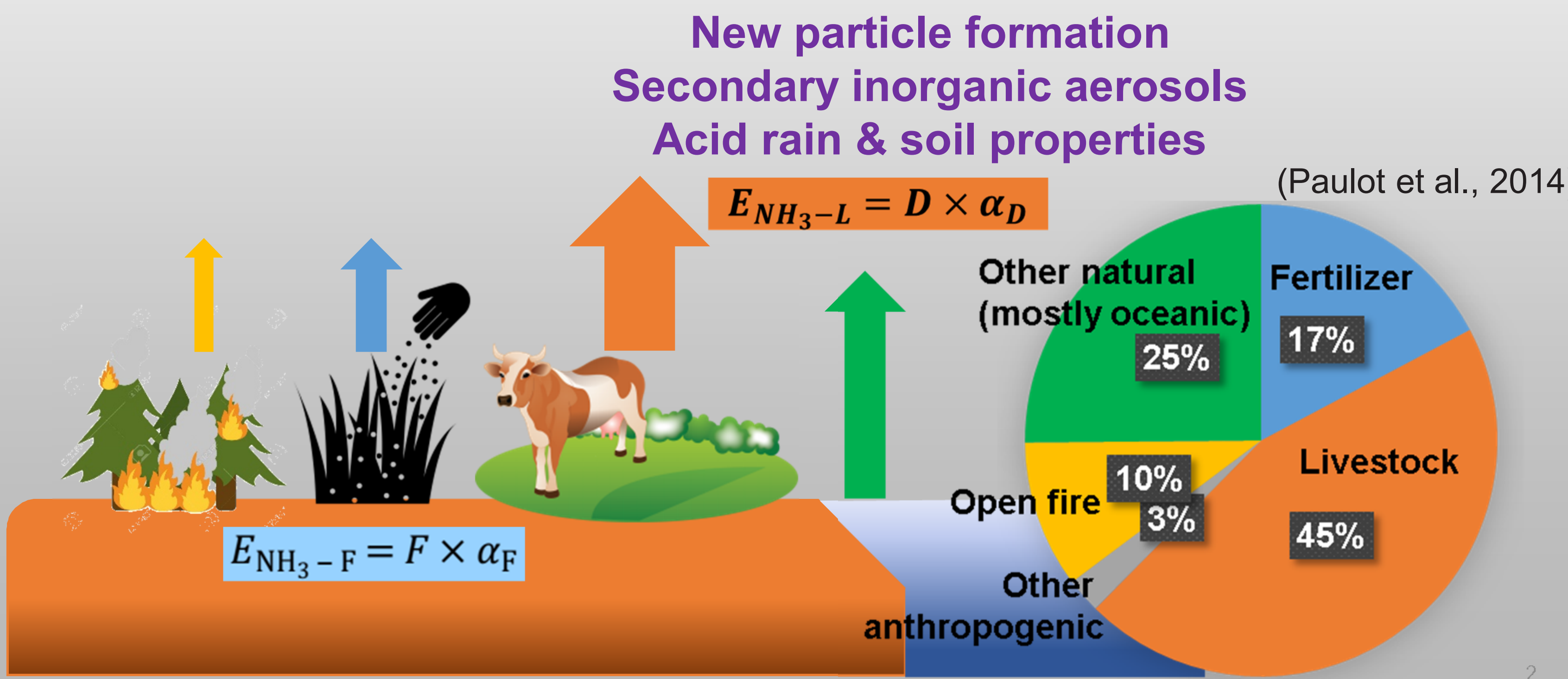
Assessing the iterative finite difference mass balance and 4D-Var methods to derive ammonia emissions over North America using synthetic observations

Chi Li, Randall V. Martin, Mark W. Shephard, Karen Cady-Pereira, Matthew J. Cooper, Jennifer Kaiser, Colin J. Lee, Lin Zhang, & Daven K. Henze

Promise of satellite observations to reduce large uncertainties in bottom-up NH_3 emission estimates



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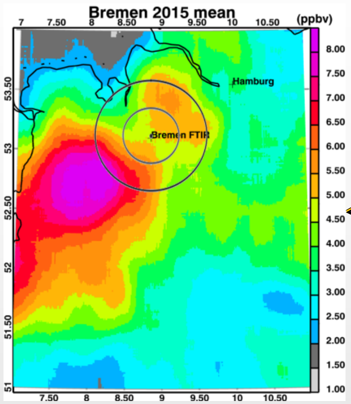
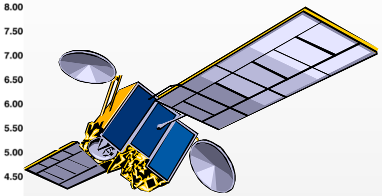
Cross-track Infrared Sounder (CrIS)

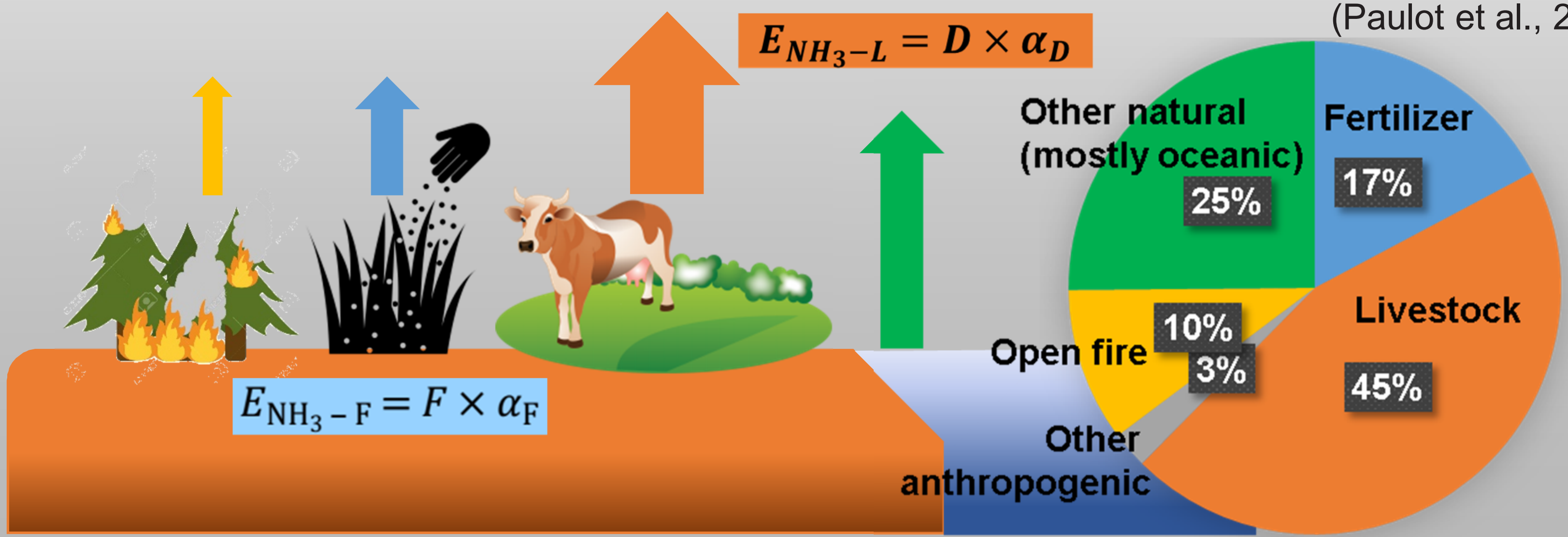




(Shepard and Cady-Pereira, 2015)

New particle formation
Secondary inorganic aerosols
Acid rain & soil properties

(Paulot et al., 2014)



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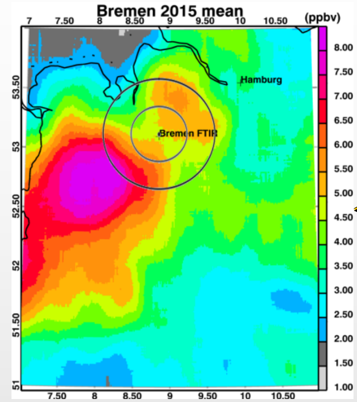

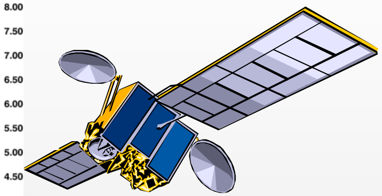

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$F(x_a)$ — Compare — $[NH_3]$ (y)

Δ

Inversion methods

adjust

E_NH_3 (x)

New particle formation
Secondary inorganic aerosols
Acid rain & soil properties

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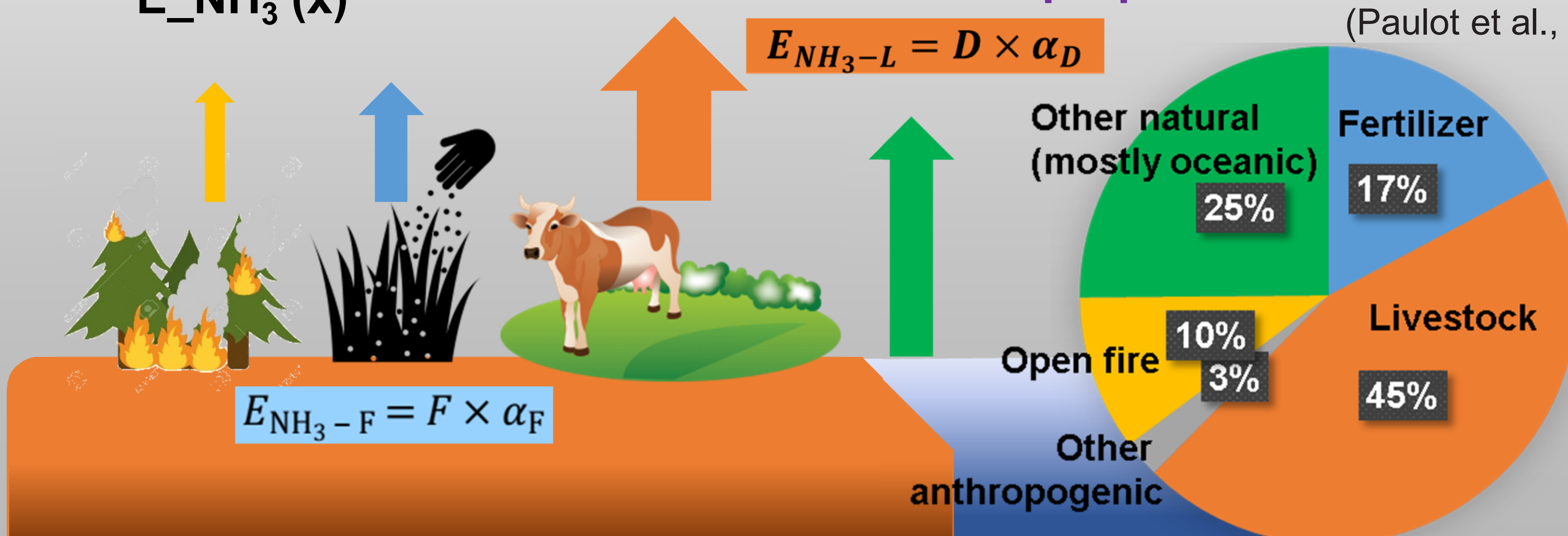
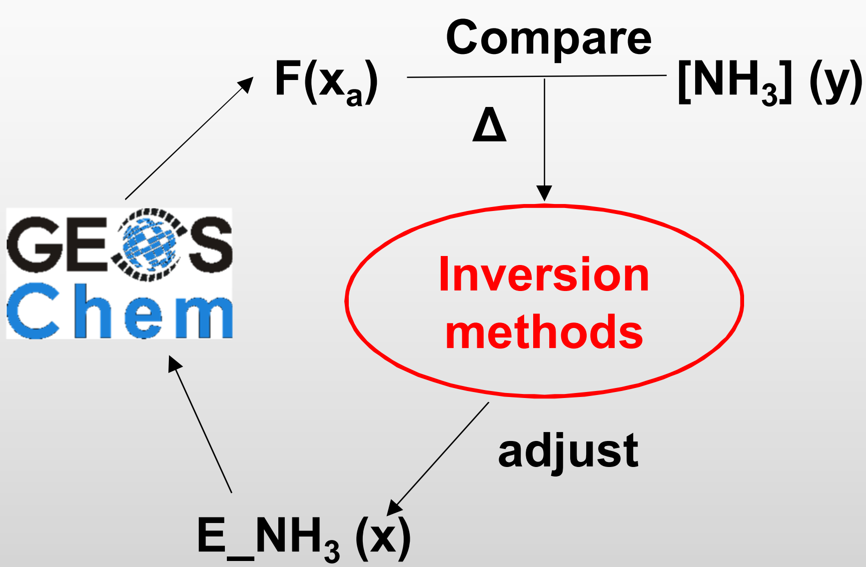


Two approaches to constrain NH_3 emissions from satellite observations



Methods	Pros	Cons
4-dimensional variational assimilation (4D-Var)	Accounts for sensitivities of Δ to emissions everywhere	Large computational cost
Iterative finite-difference mass balance (IFDMB)	Less computational complexity	Require dominant local sensitivities

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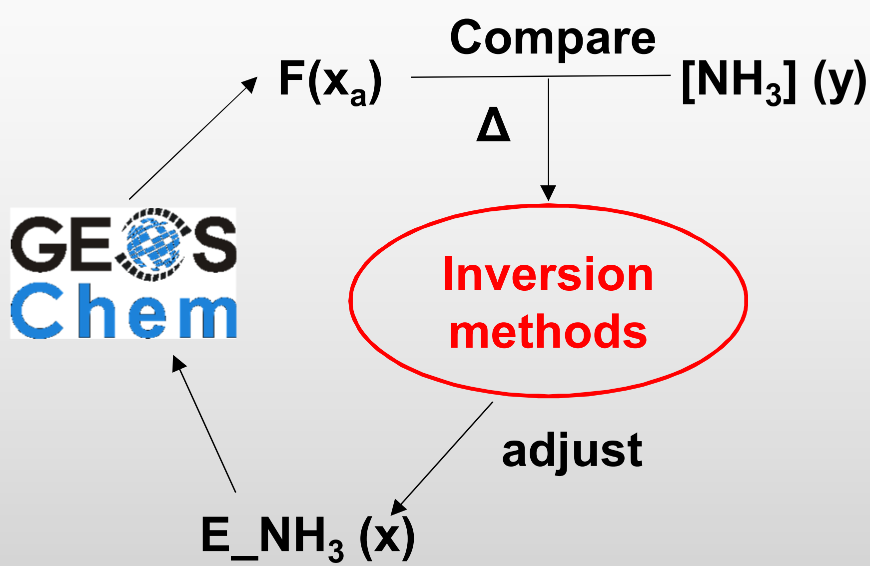

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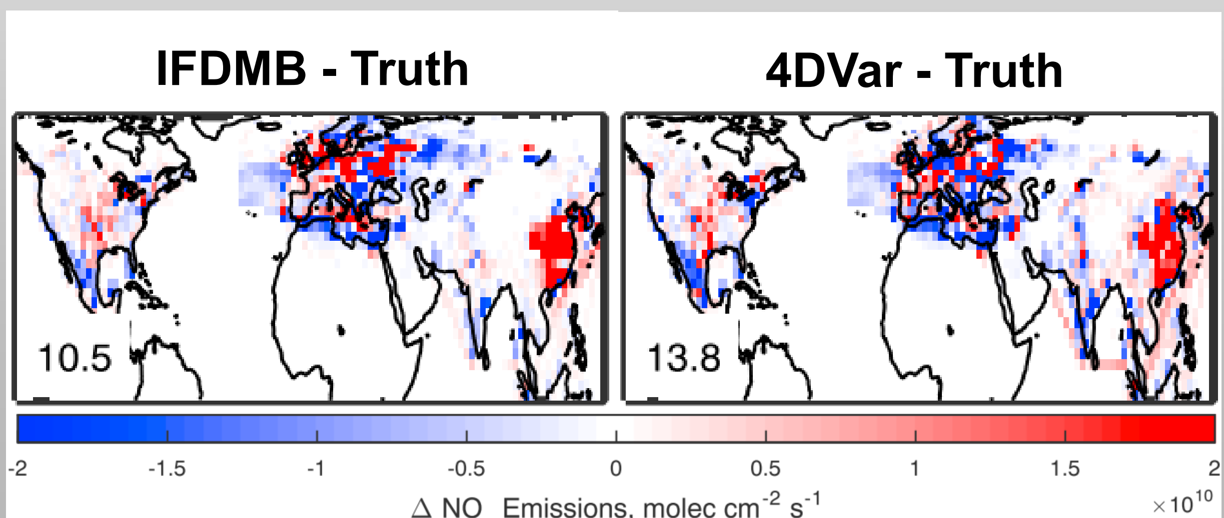

IFDMB performs similar to 4D-Var for NO_x at $2^\circ \times 2.5^\circ$ (Cooper et al., 2017)

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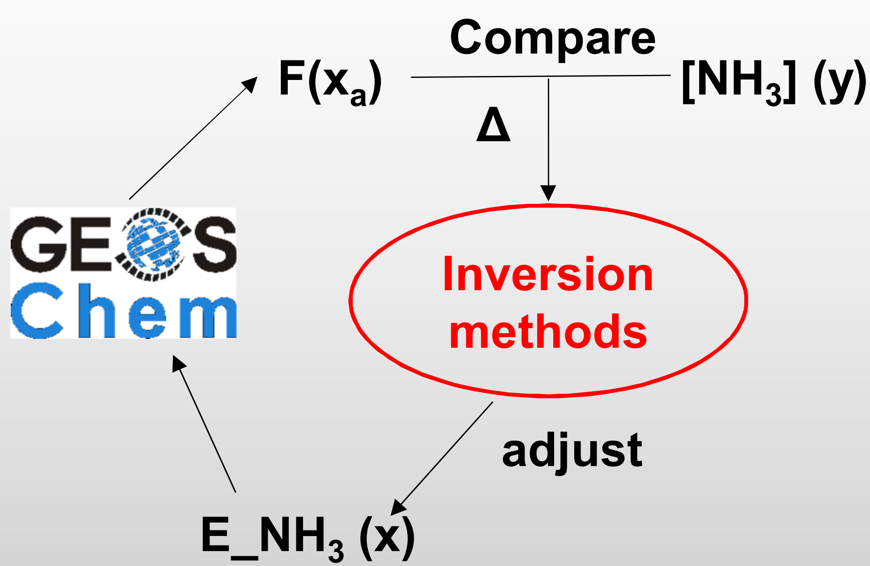

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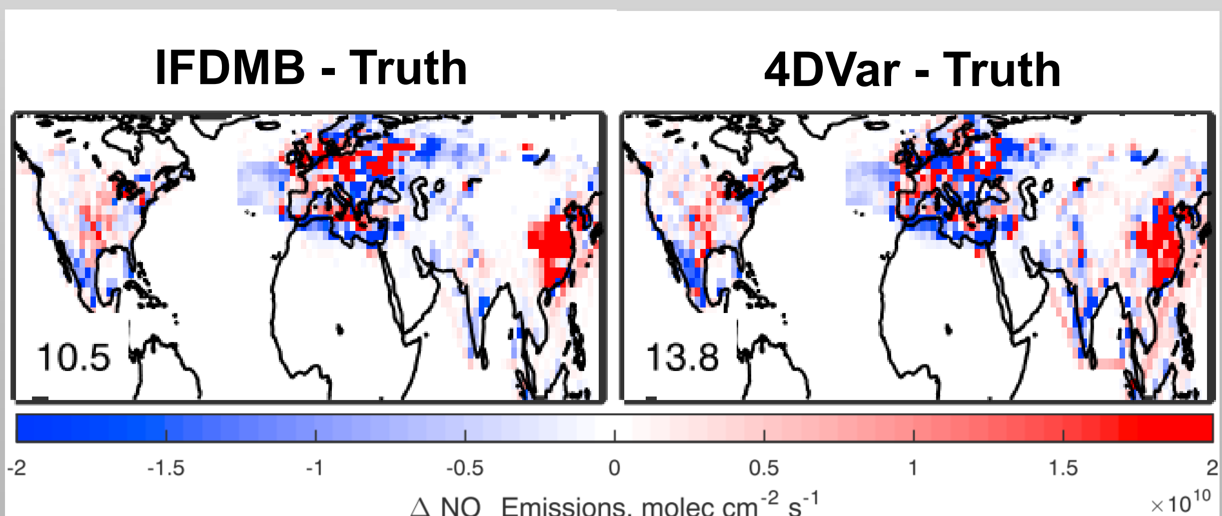

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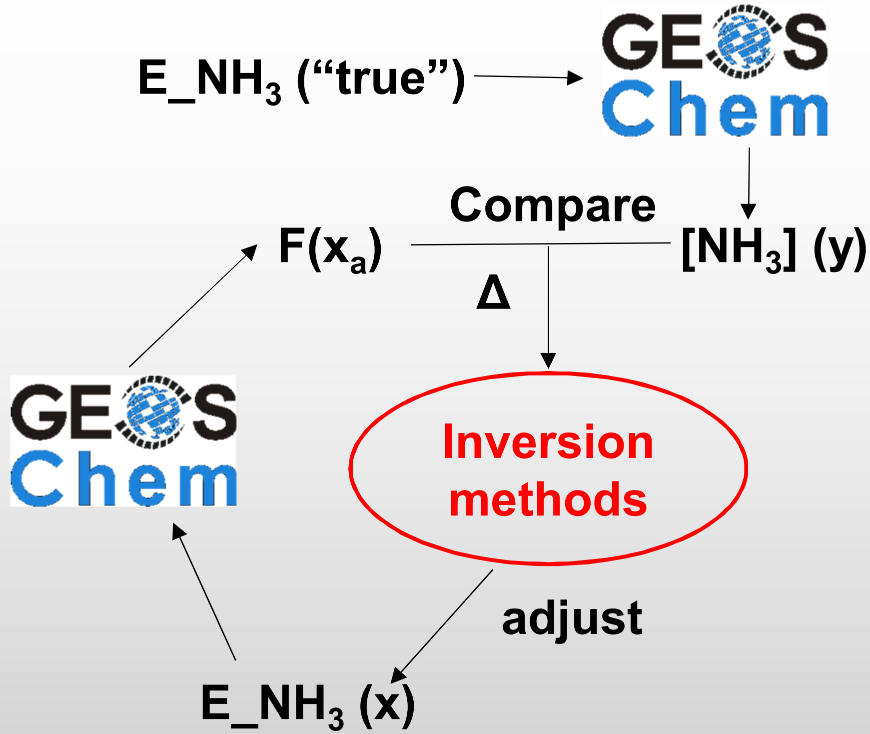

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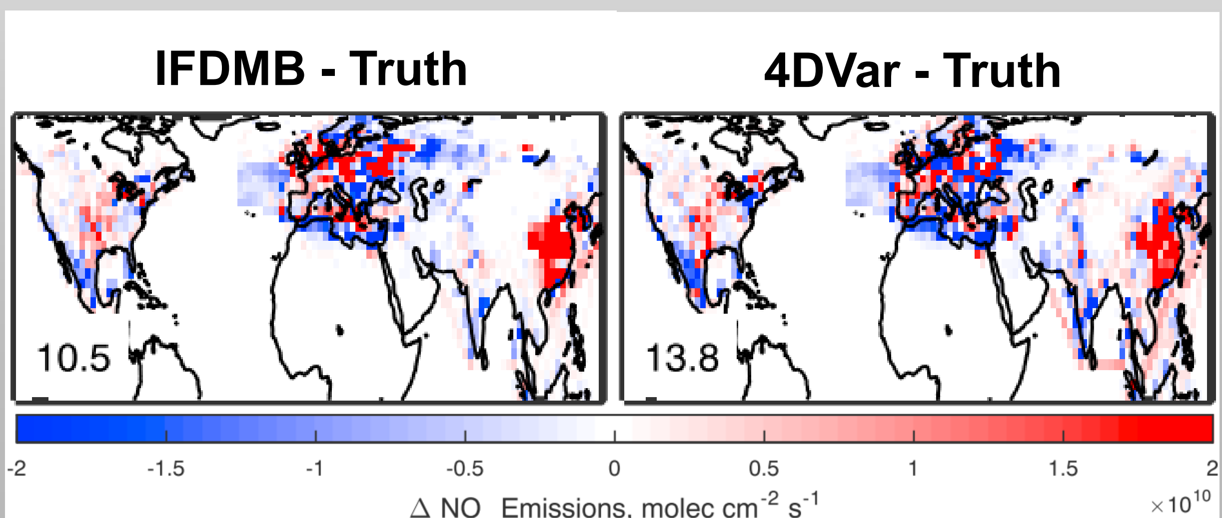

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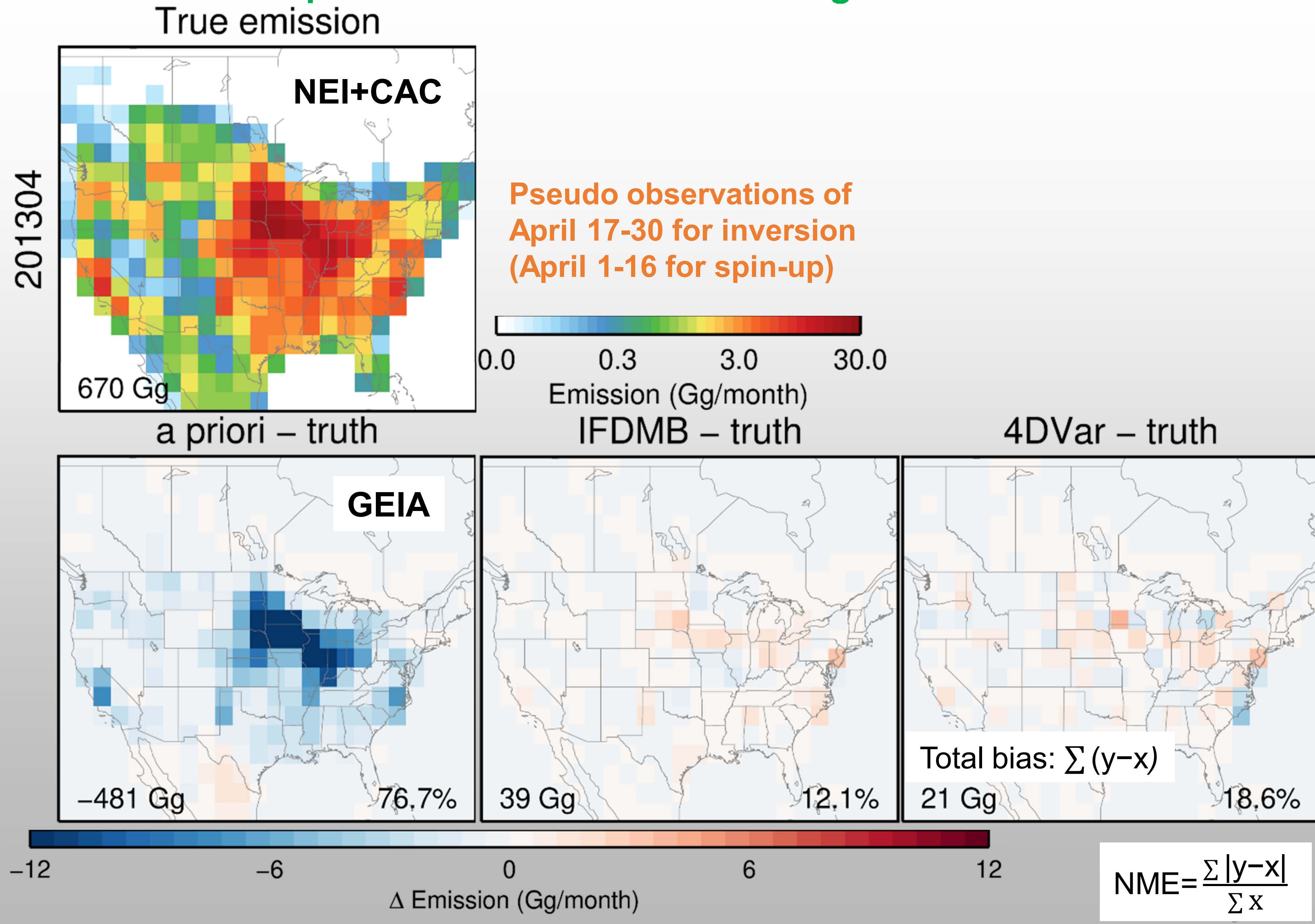
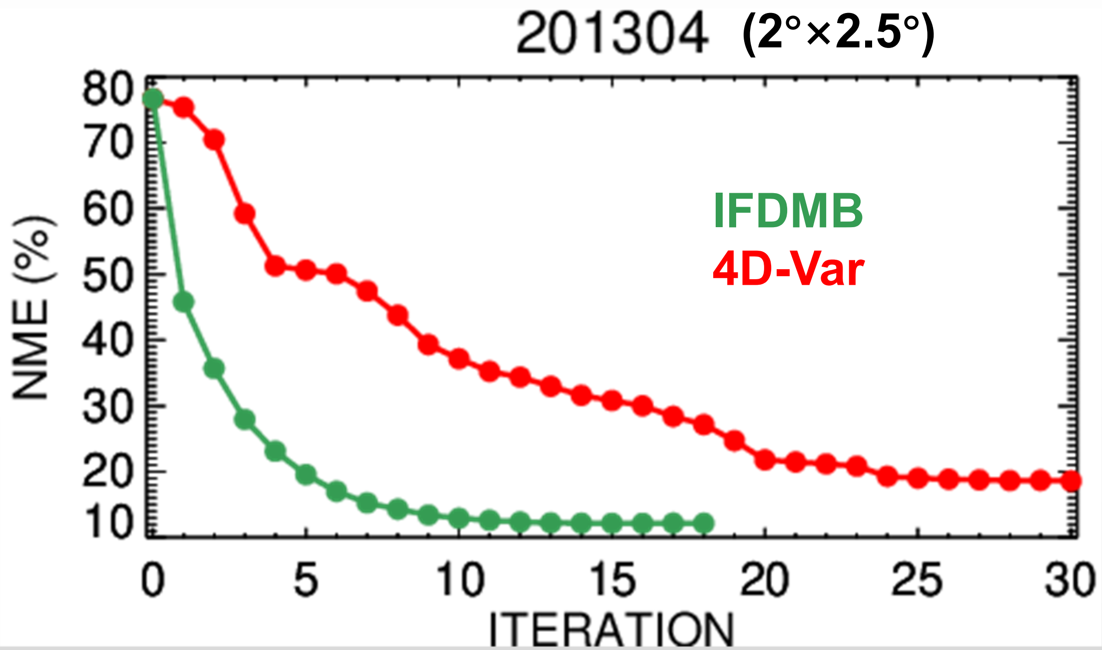
Both methods perform well if the real world grids are 2°×2.5°



IFDMB requires 3-4 times less computational cost



“Unsuccessful” 4D-Var iterations are included to reflect real computational cost.

Fewer number of iterations and wall time needed in IFDMB (~10, 50 hrs) than in 4D-Var (~20, 180 hrs)

(Tested environment: 16 CPUs, Intel(R) Xeon(R) Gold 6154, 3.00GHz)

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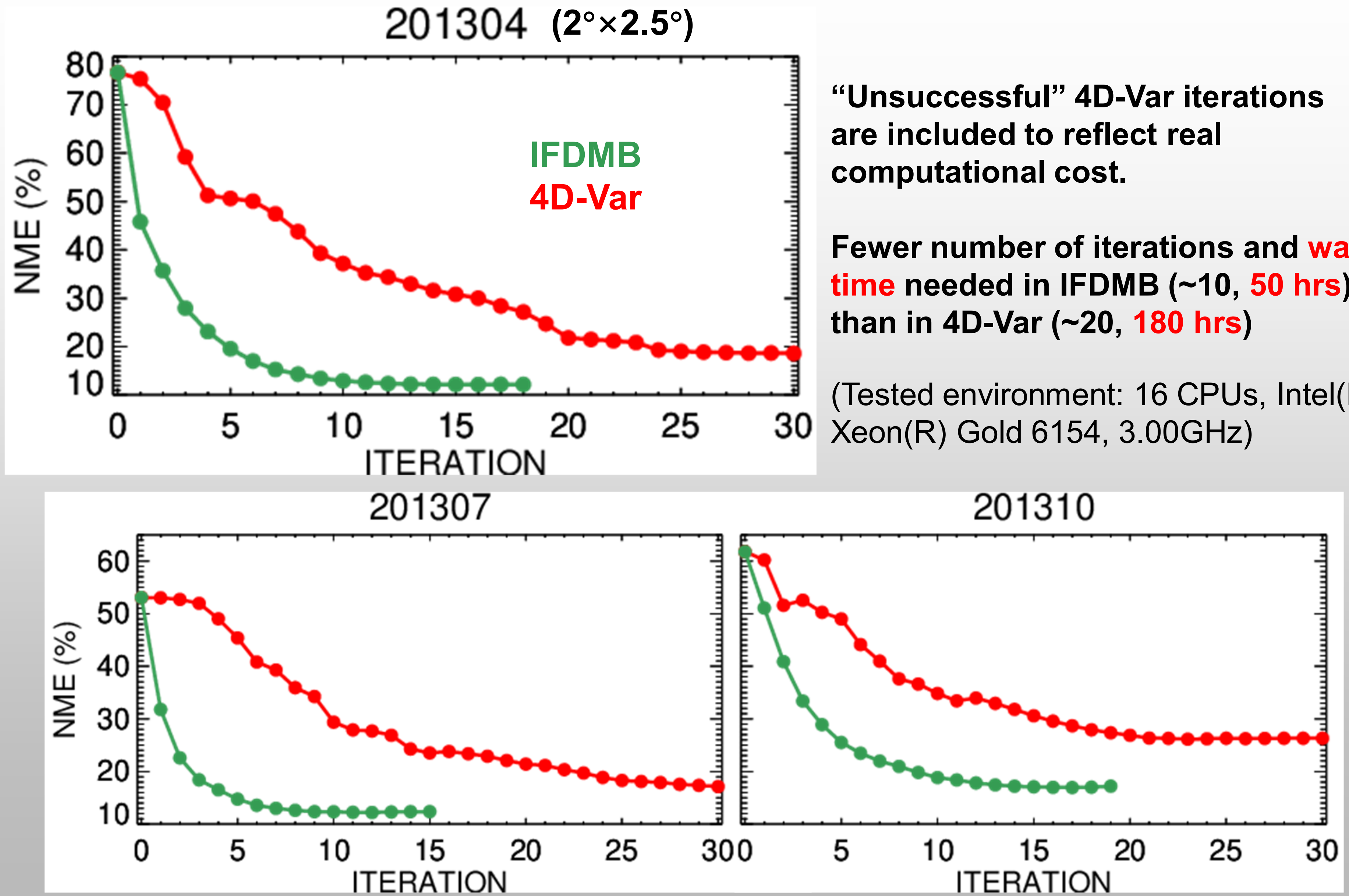

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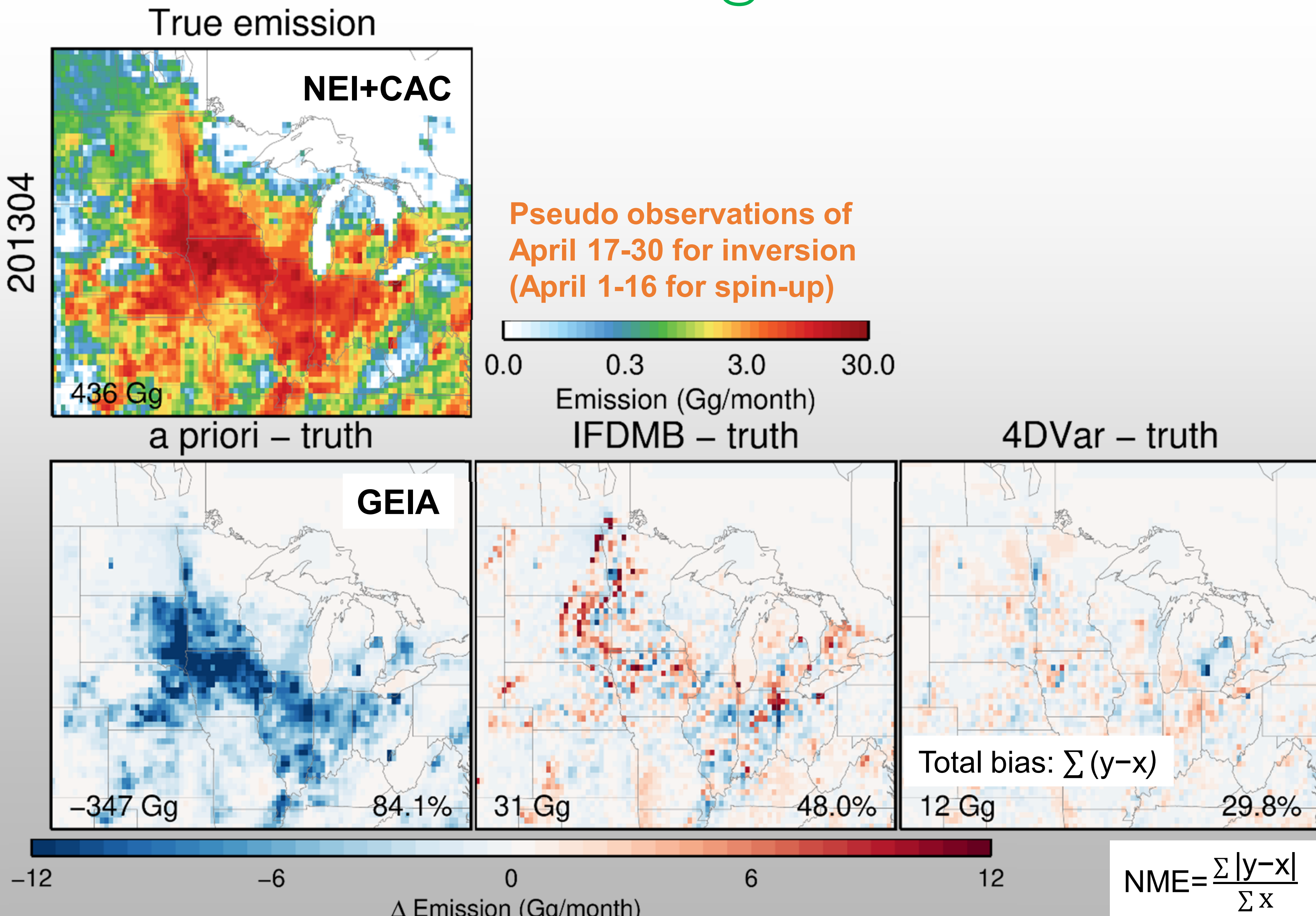
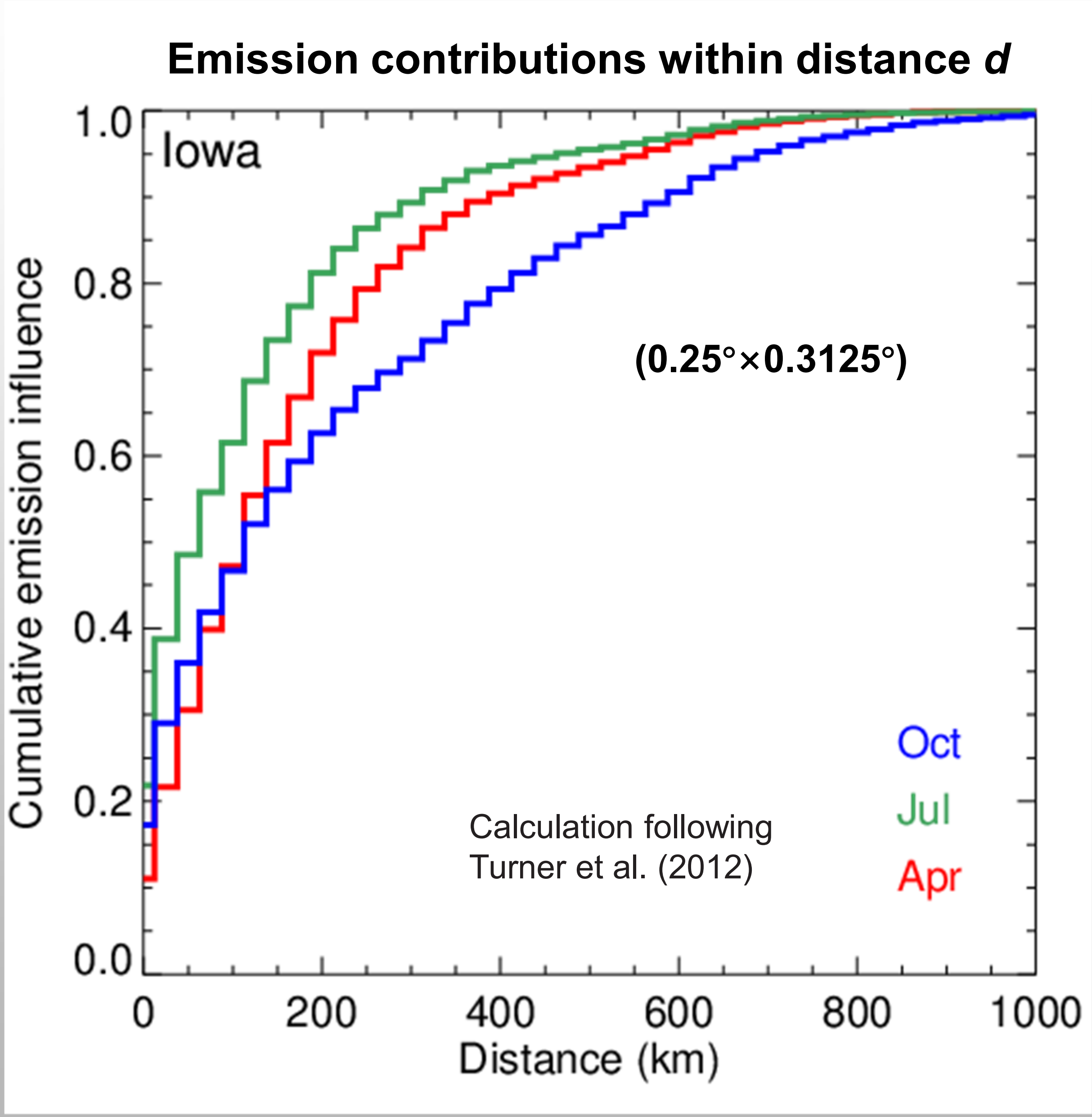
Inversion errors stand out for IFDMB @ 0.25°×0.3125°



Stronger non-local emission contribution to columnar NH_3 ("smearing effect") at finer resolution



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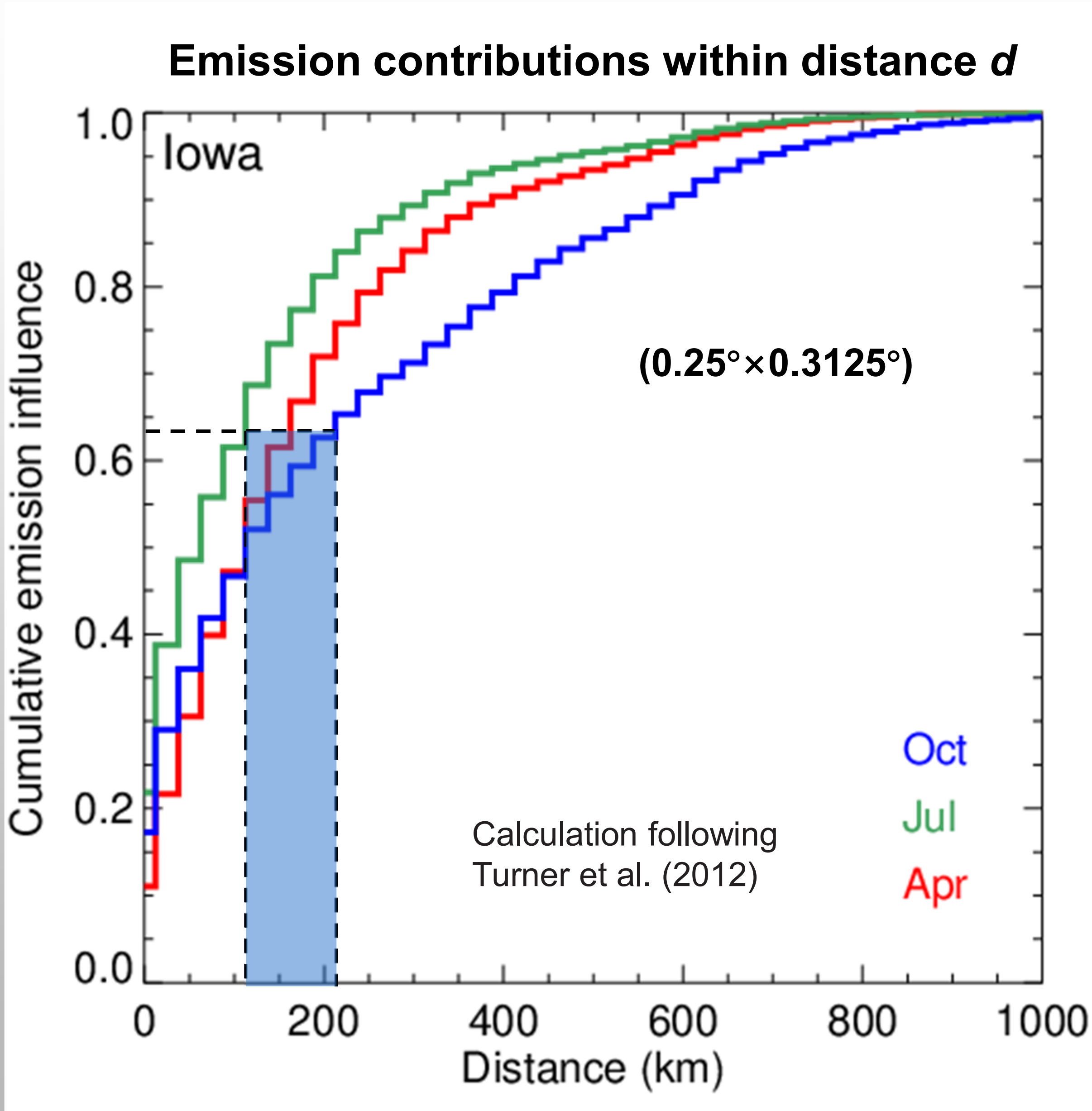

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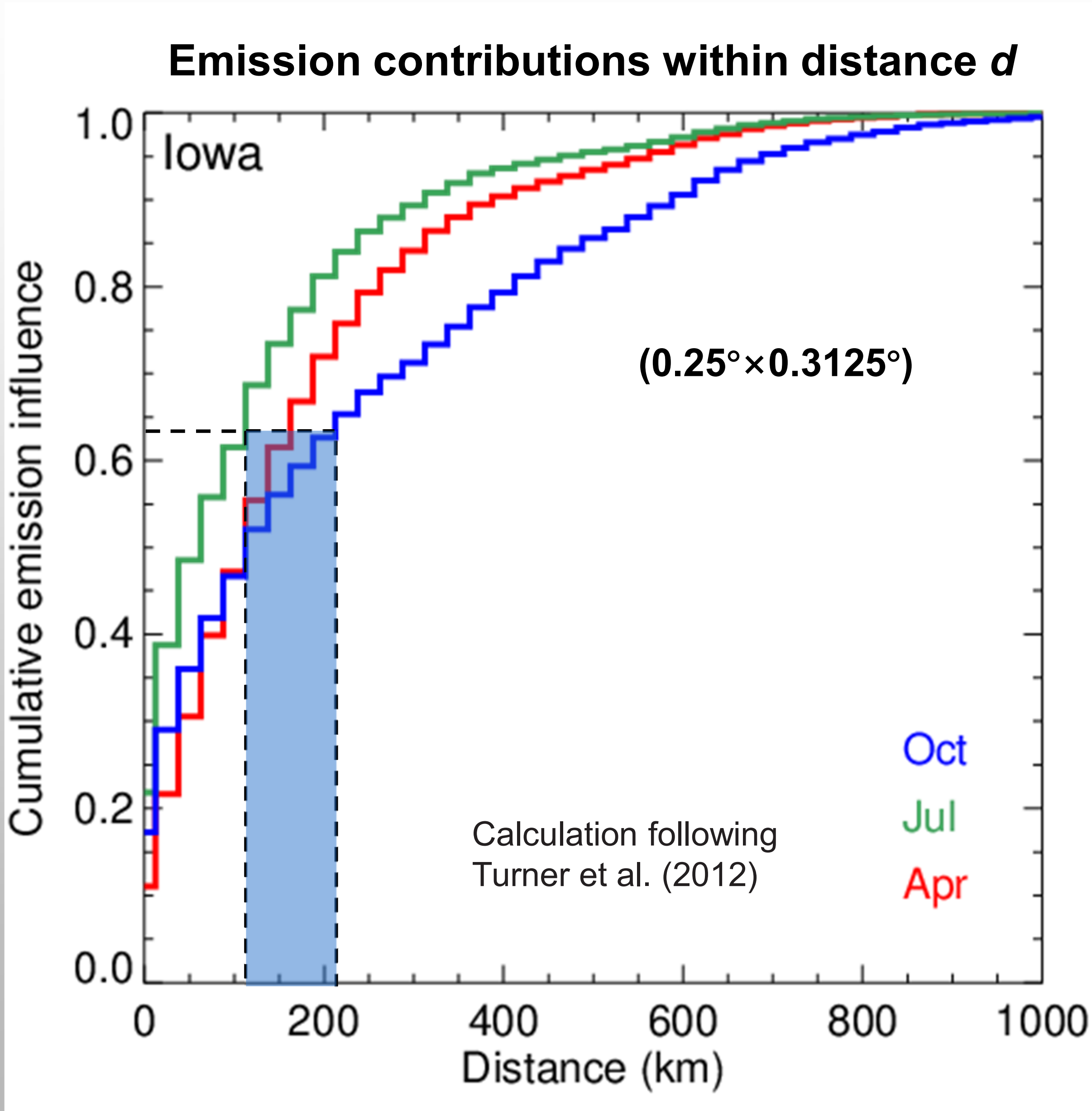

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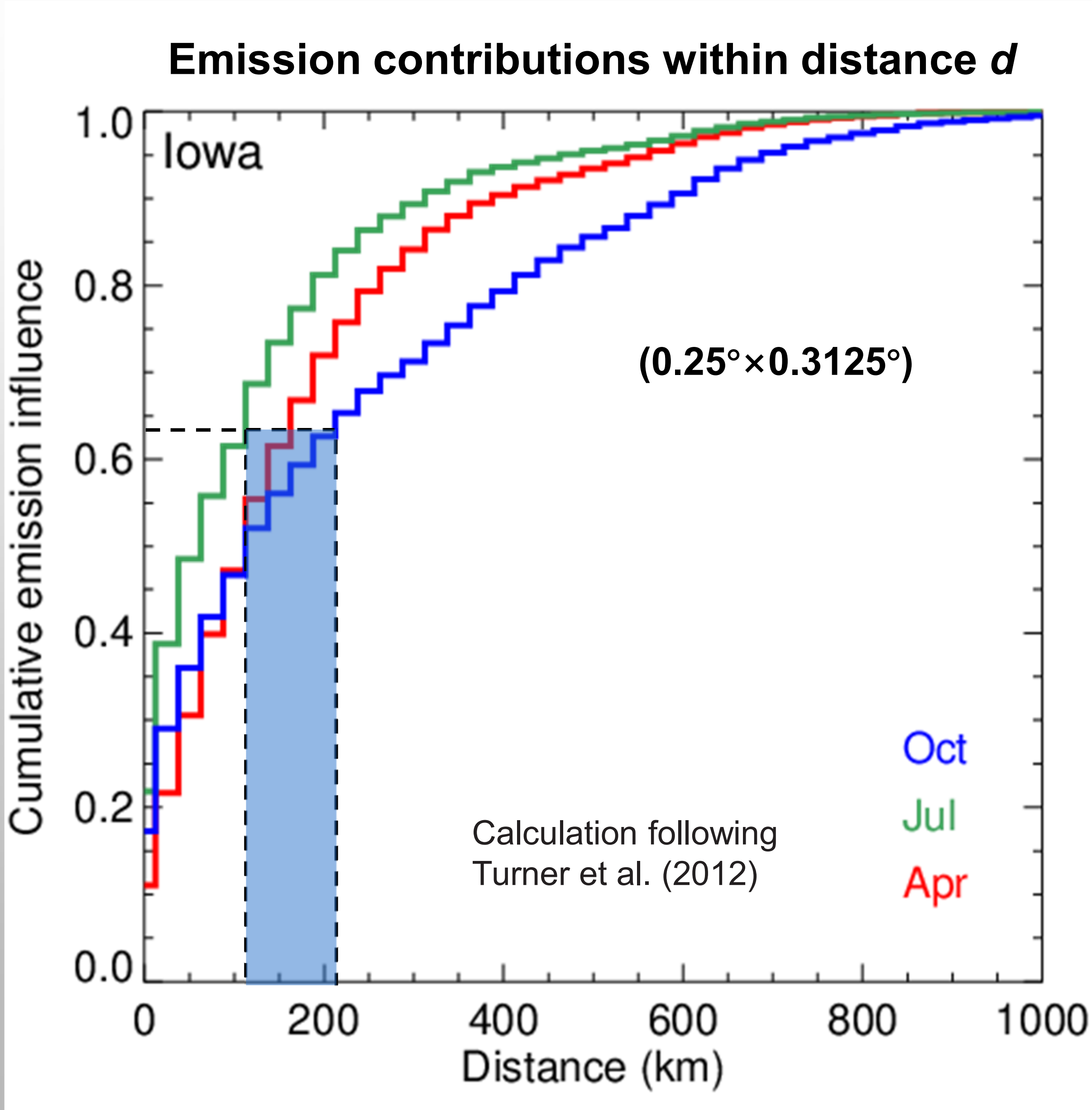

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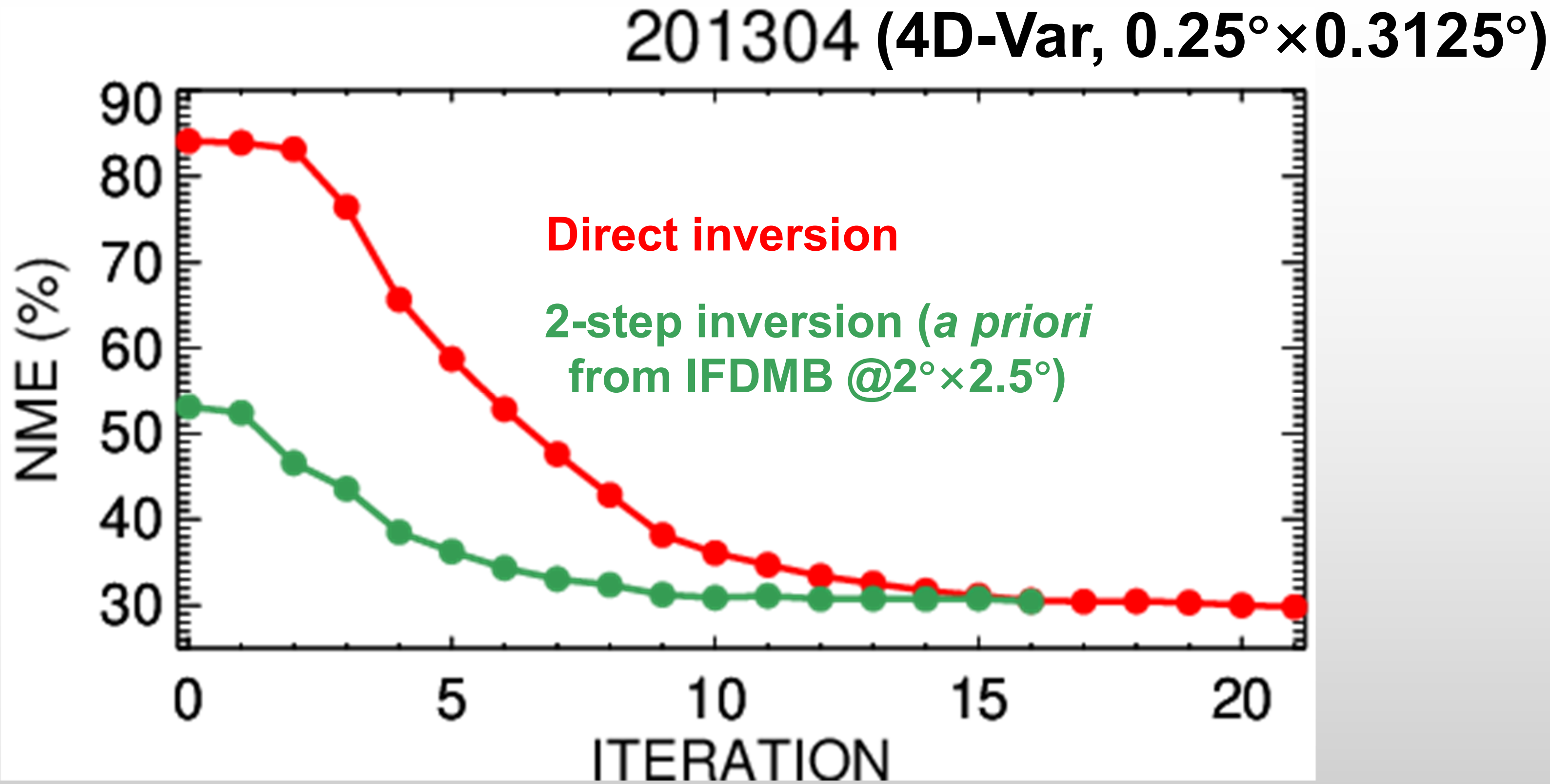

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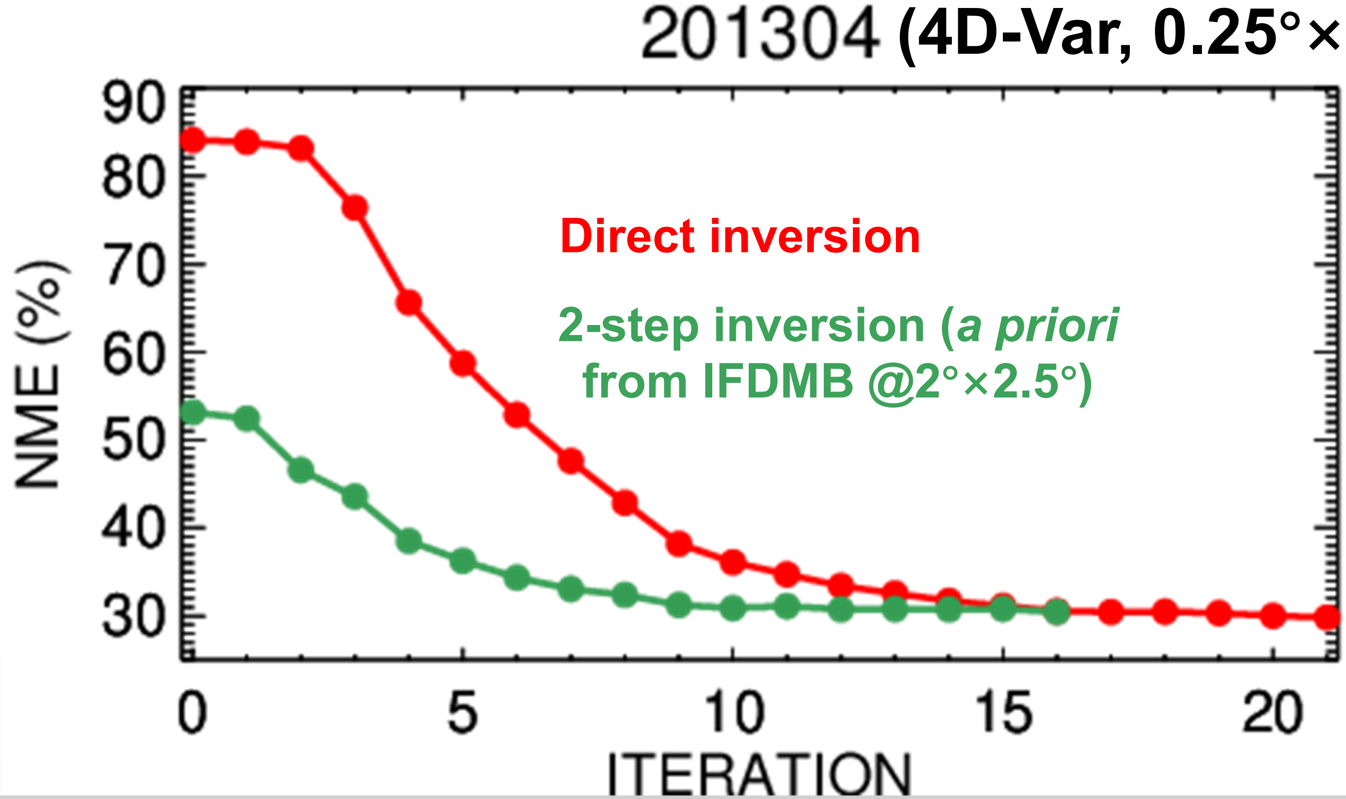
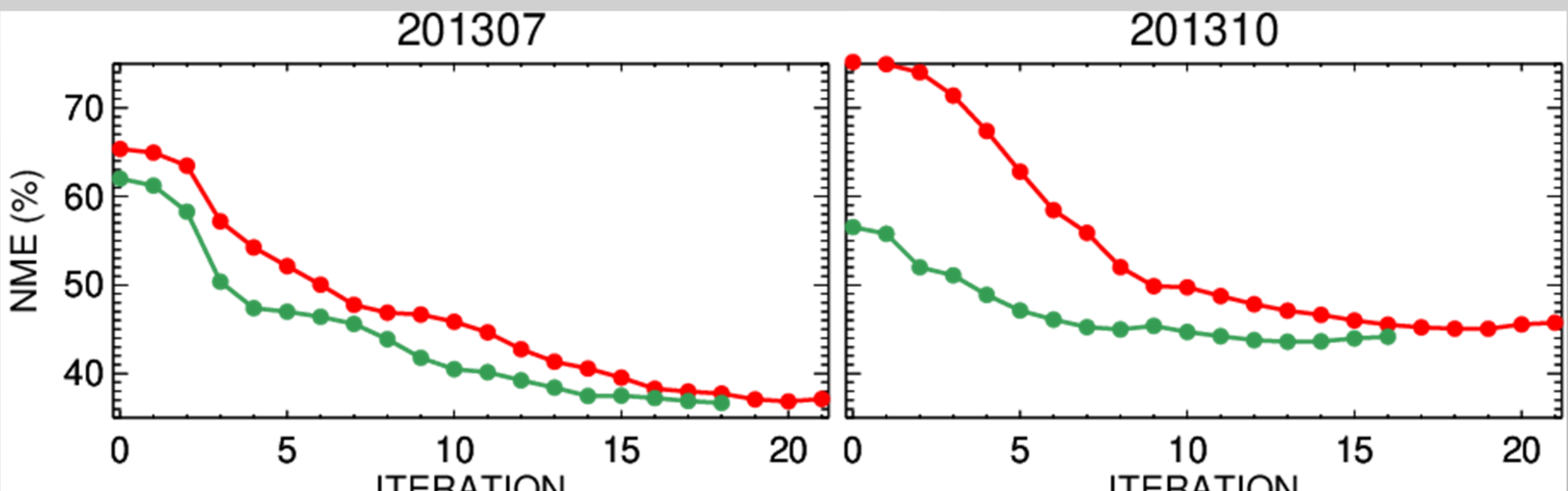


Total wall time (hrs):

420-530

320-380



Conclusion

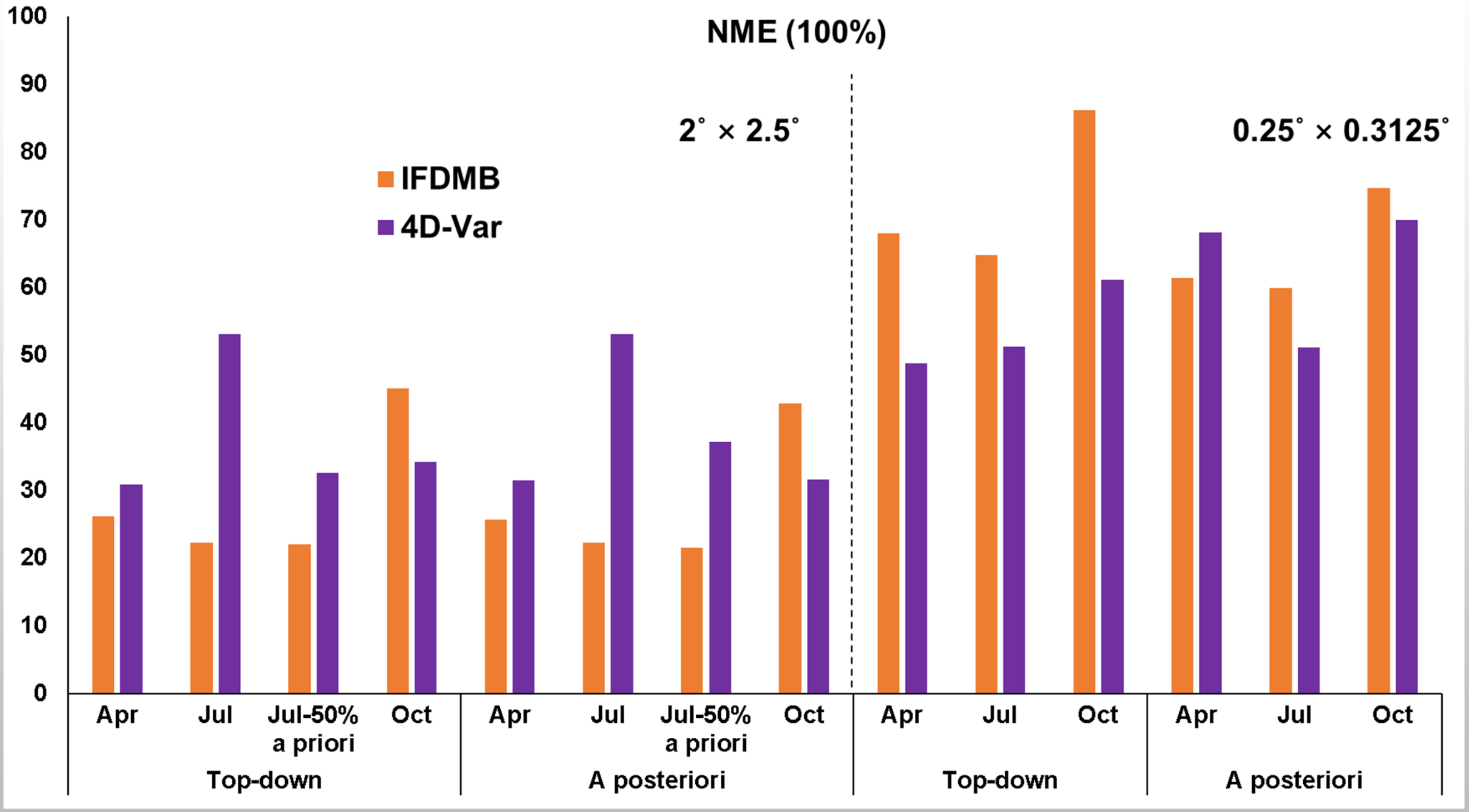
- **IFDMB approach requires 3-4 times less computational costs than 4D-Var and performs competitively at 2° × 2.5°.**
- **Strong non-local smearing at 0.25° × 0.3125° resolution (finer than the "smearing length") weakens the performance of IFDMB relative to 4D-Var.**
- **Prior IFDMB inversions at 2°×2.5° could refine the *a priori* emissions to accelerate the 4D-Var at 0.25°×0.3125°.**

Thank you!

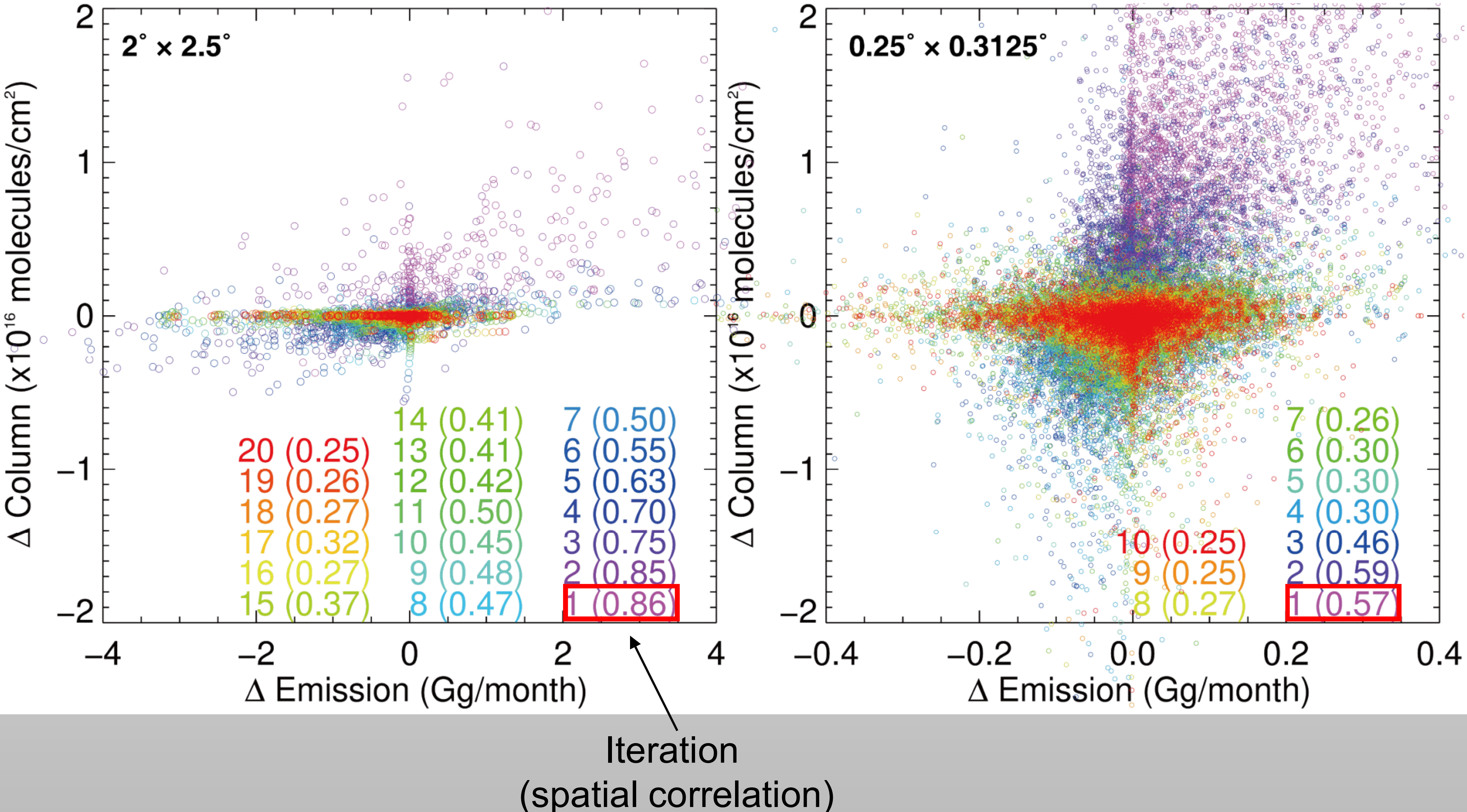
More details:
Li et al. (2019), J. Geophys. Res. Atmos., doi:10.1029/2018JD030183

Introducing errors in observations indicate similar findings

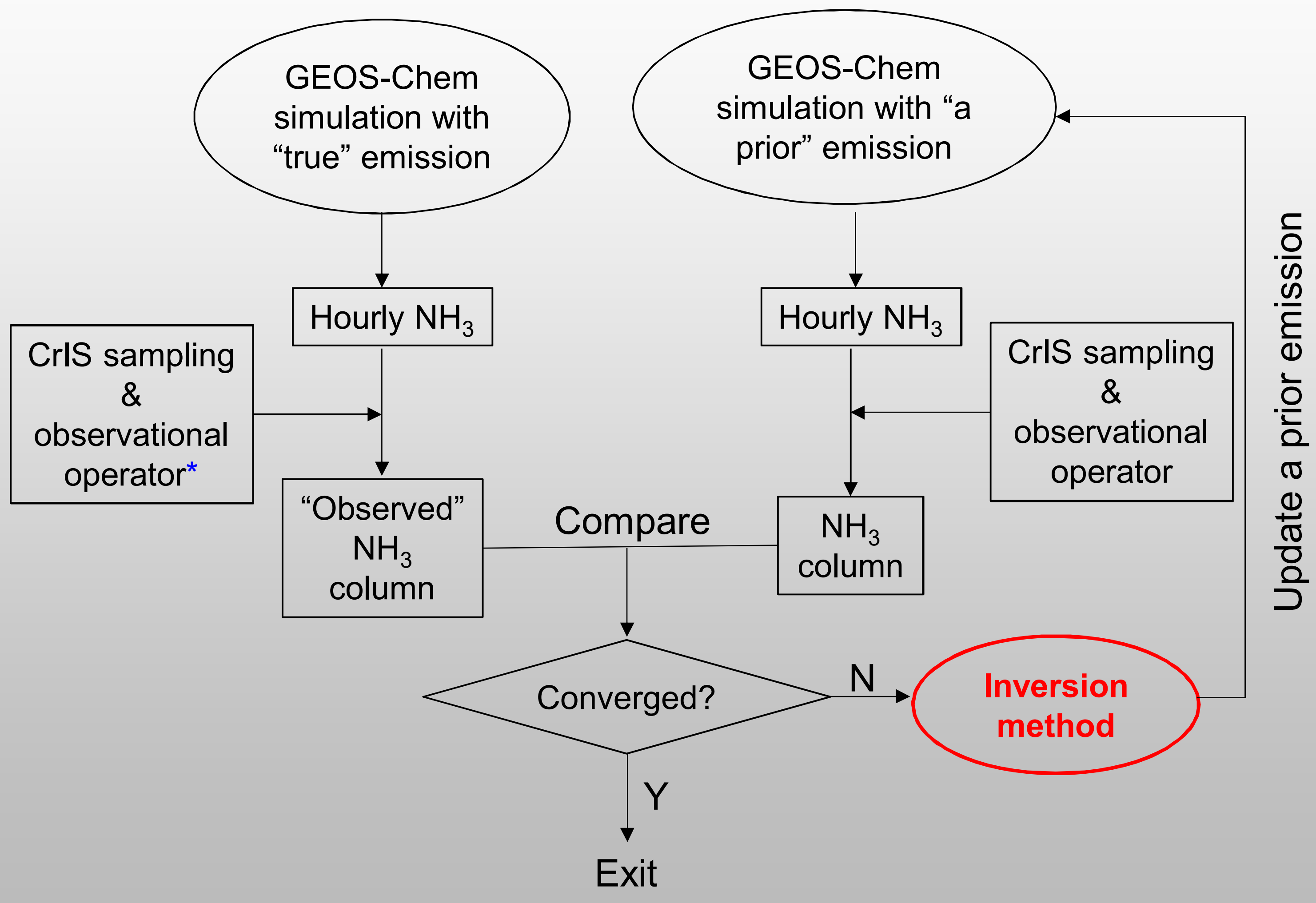
Errors: 35%*$\Omega(NH_3)$ or at least 5×10^{15} molecules/cm^2



Less "localization" of *Δy-Δx* relationship @ 0.25°×0.3125°



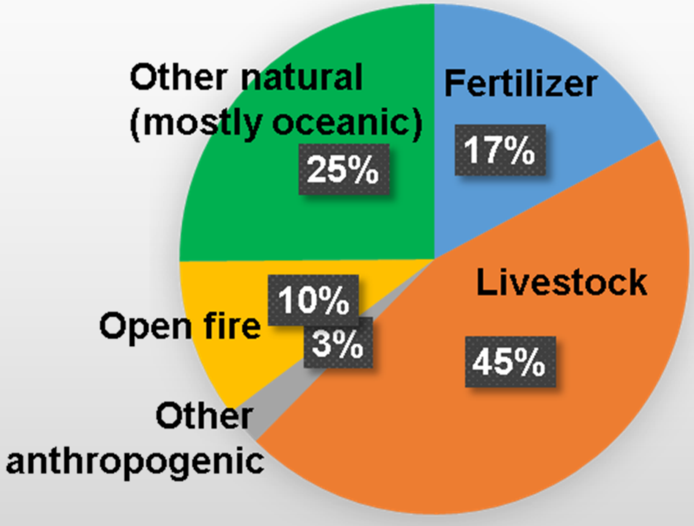
A comparison of finite difference mass balance and 4D-Var inversion methods for NH_3 emission from CrIS retrievals



Large uncertainties in (agricultural) NH_3 emissions estimates

(Paulot et al., 2014)

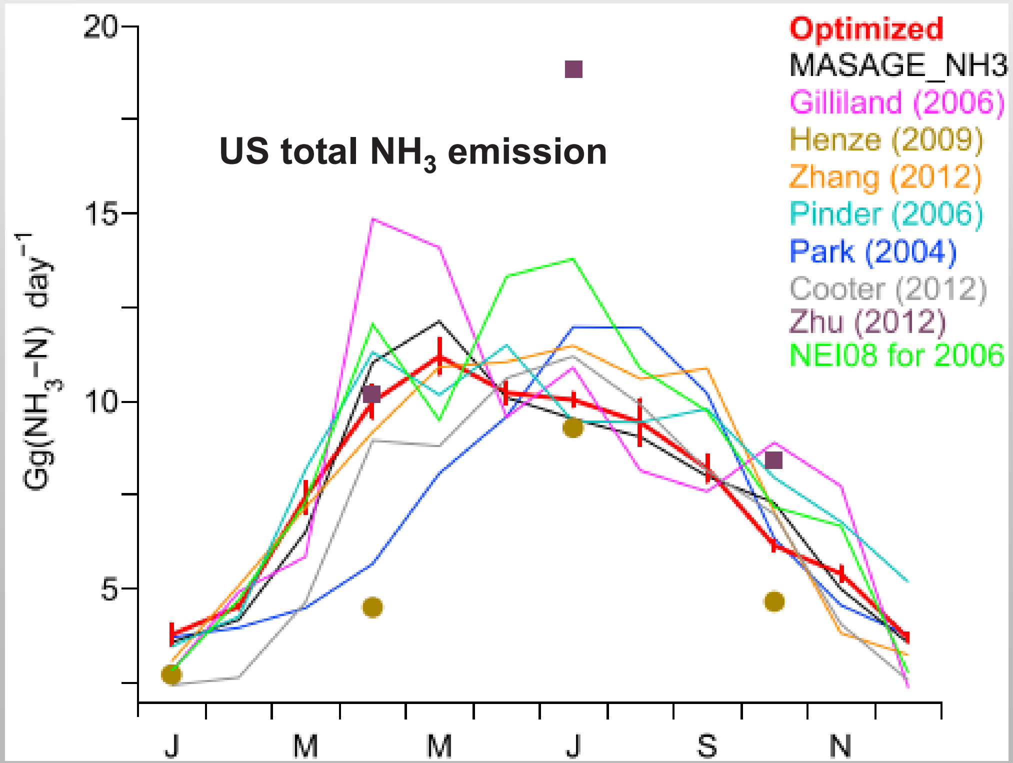
Global annual NH_3 emission (54.5 TgN)



$$E_{NH_3-F} = F \times \alpha_F$$

Emission factor

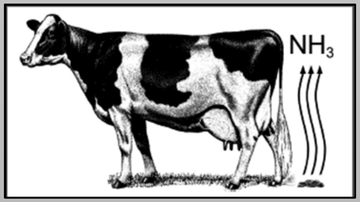
Fertilizer application magnitude



Emission factor

$$E_{NH_3-L} = D \times \alpha_D$$

Animal number



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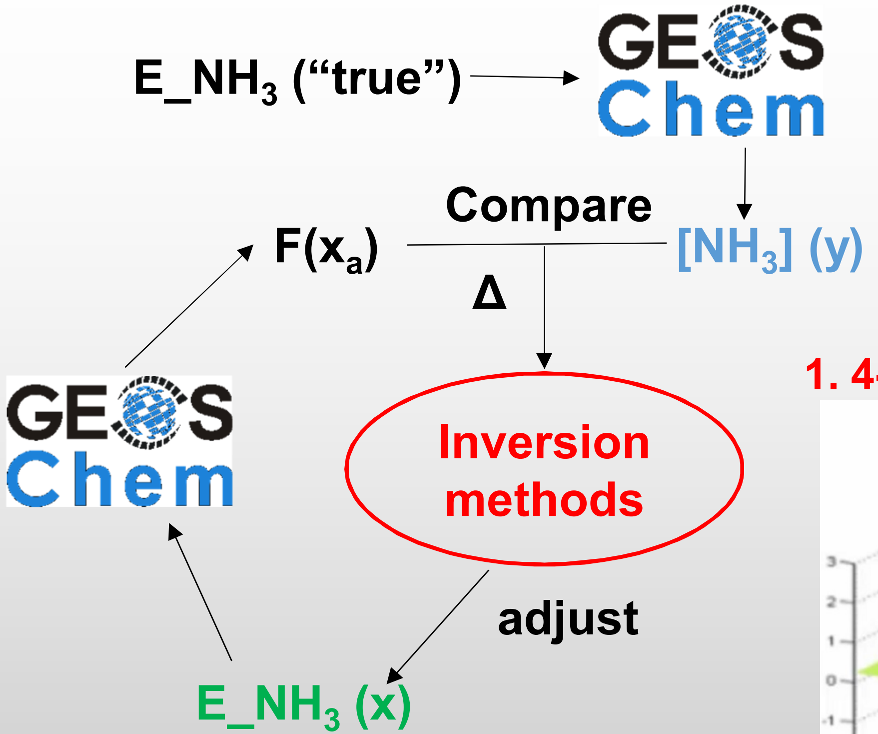

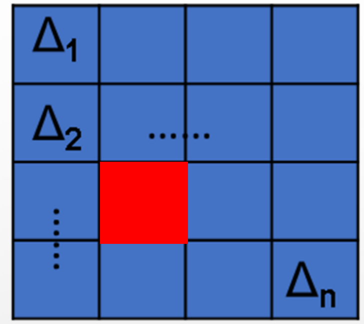

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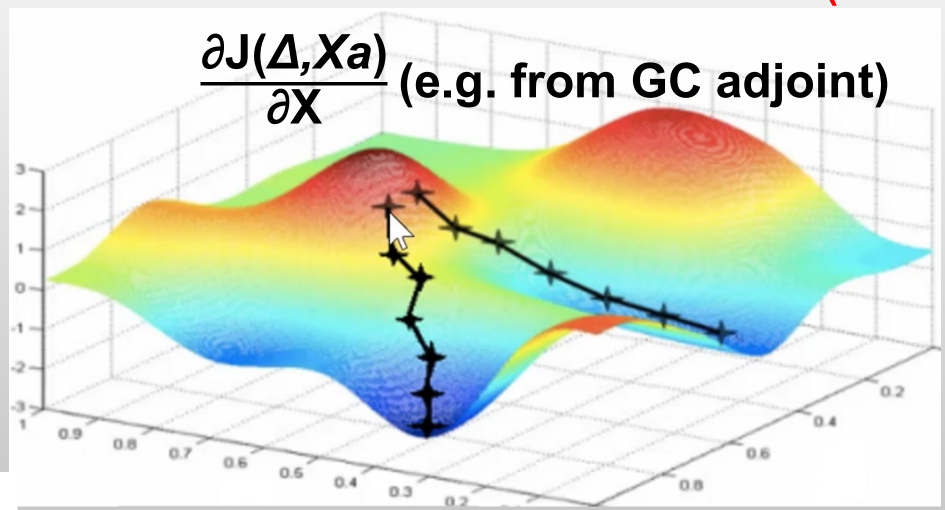

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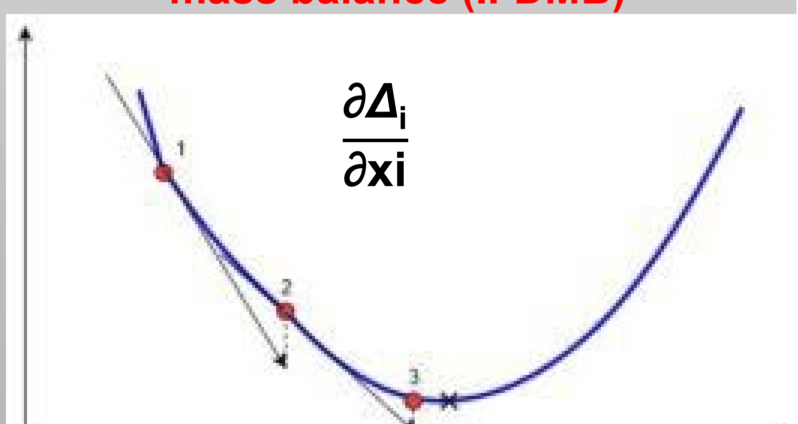

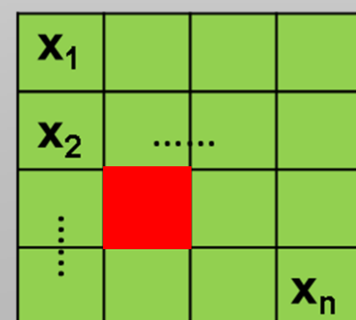

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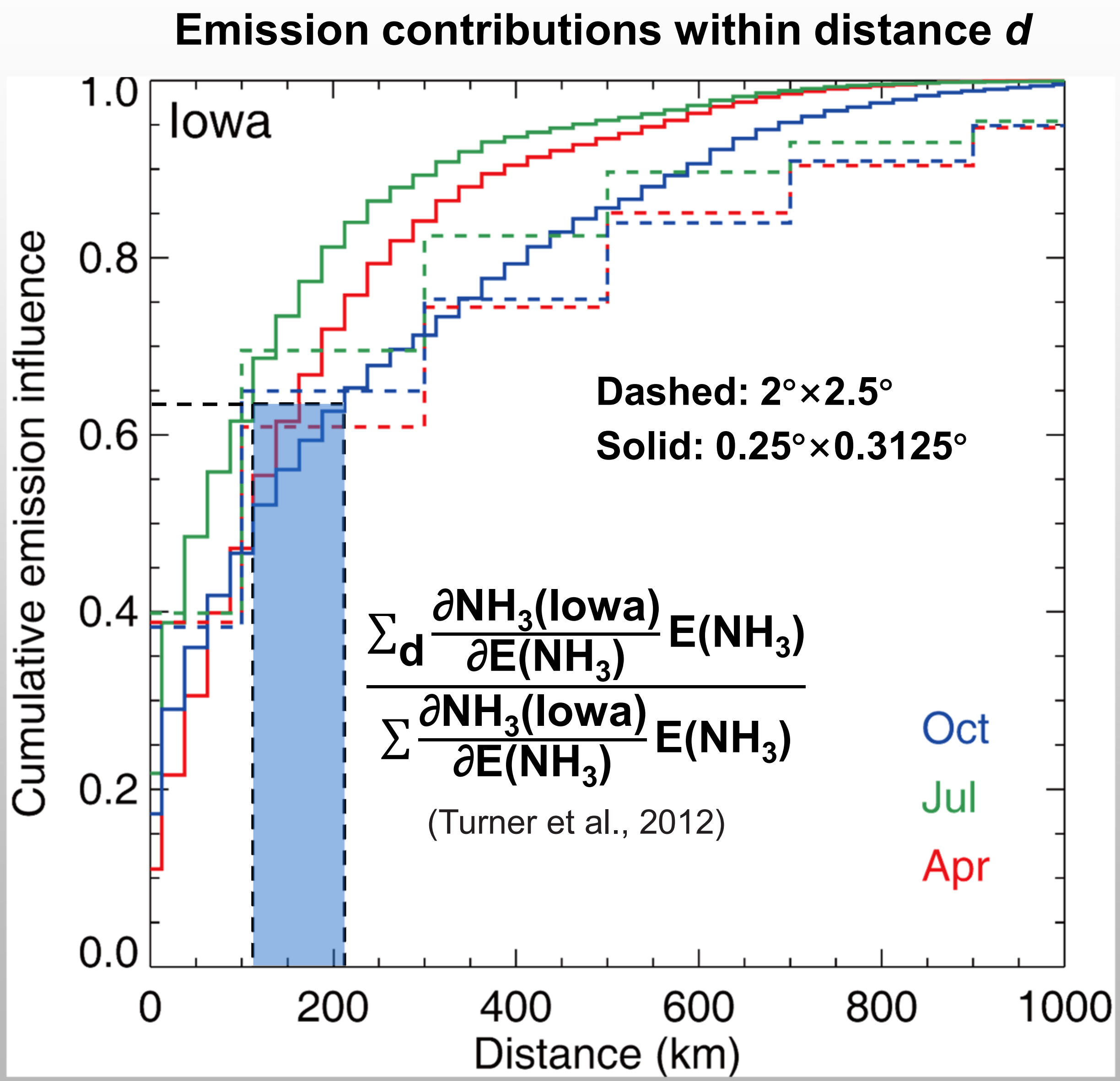

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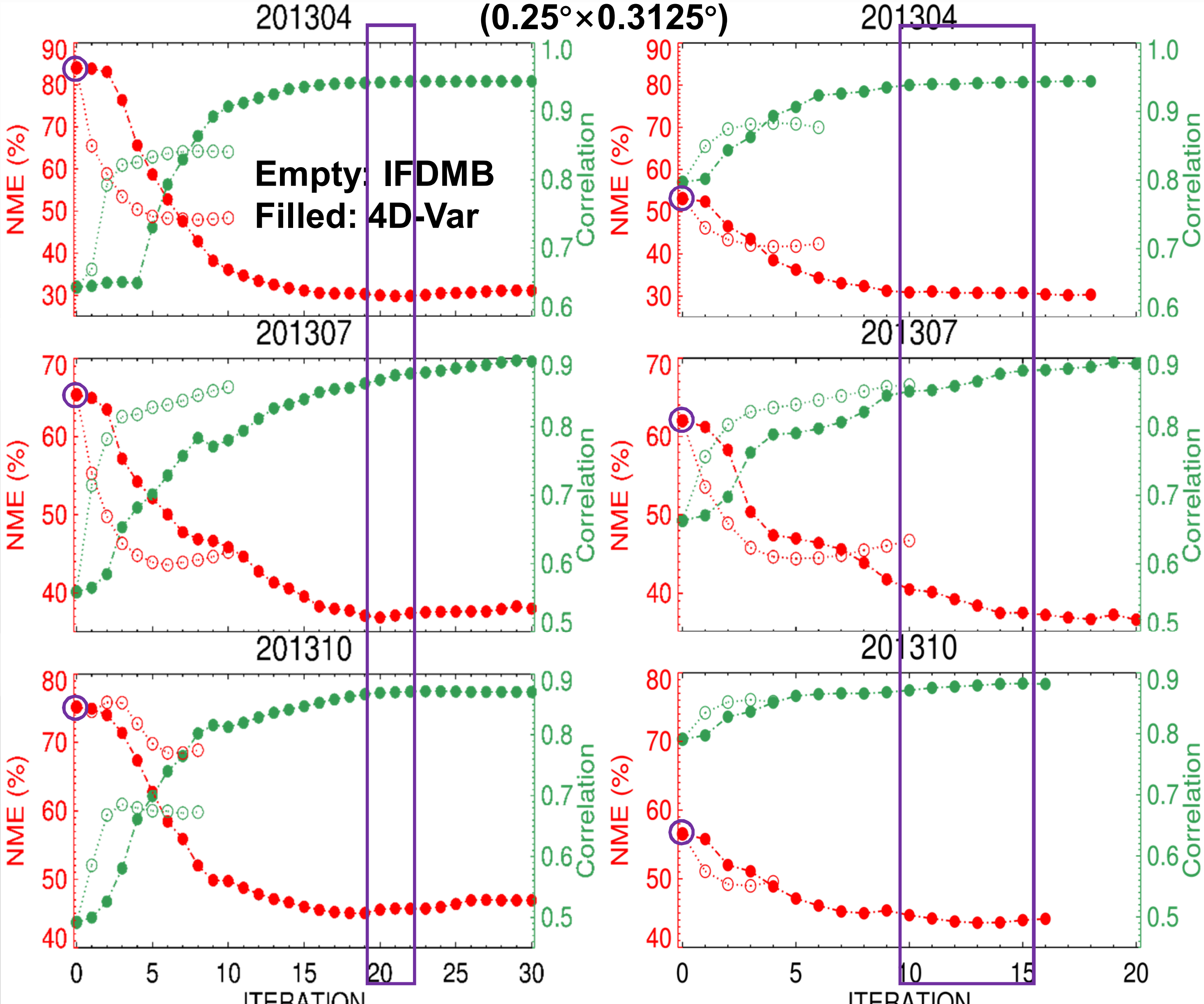
Original *a priori*

Updated *a priori* (from IFDMB @2°×2.5°)



$$\text{NME} = \frac{\sum |x-y|}{\sum y}$$

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Left: 420-530

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