Evaluating mass balance methods for constraining global NO_x emissions

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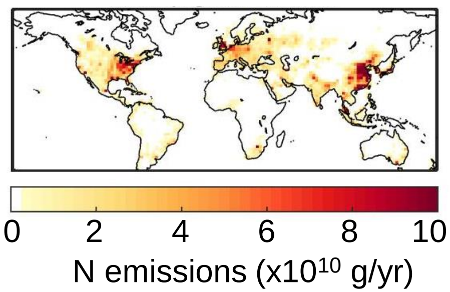
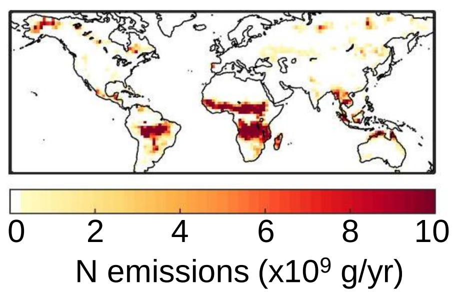
Satellite NO_2 Observations Provide Top-Down Constraints on NO_x Emission Inventories

Anthropogenic Emissions



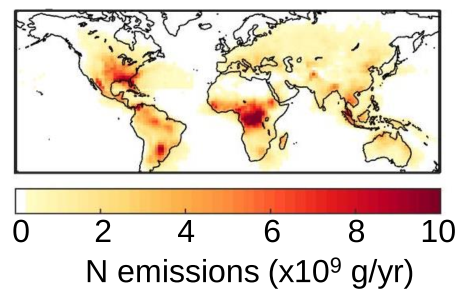
24-30Tg/yr

Biomass Burning Emissions



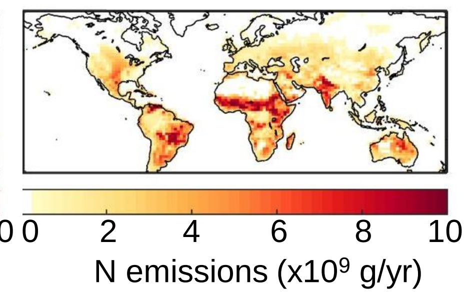
6-12 Tg/yr

Lightning Emissions



2-9 Tg/yr

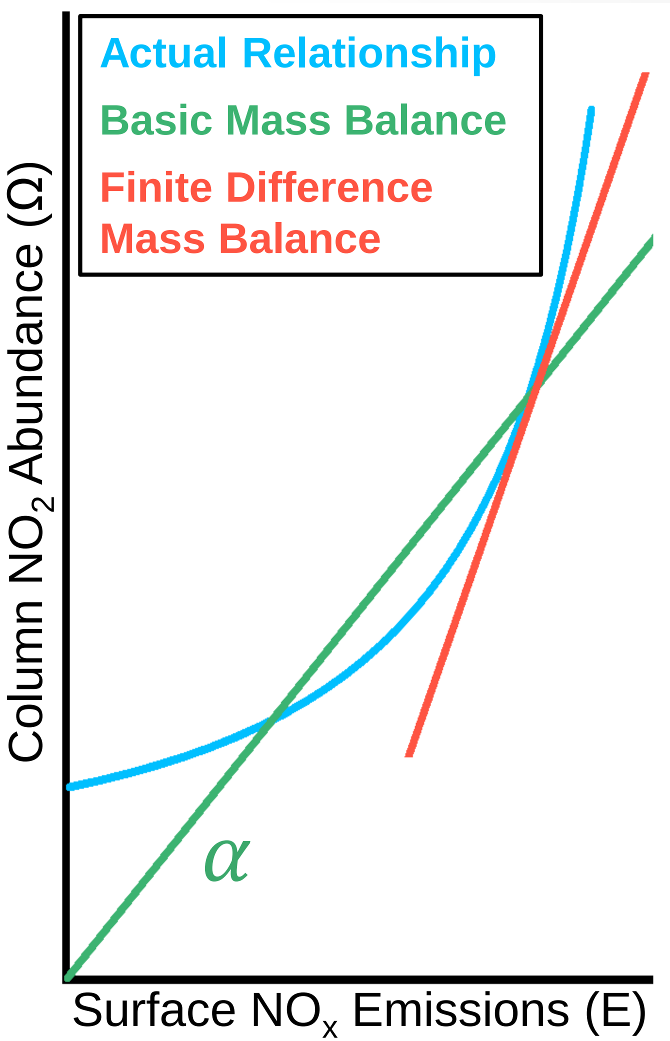
Soil Emissions



4-15 Tg/yr

- Emissions inventories have significant uncertainties
- Using satellite observations as a constraint requires inverse modeling

Improving Mass Balance Inversions with Finite Difference



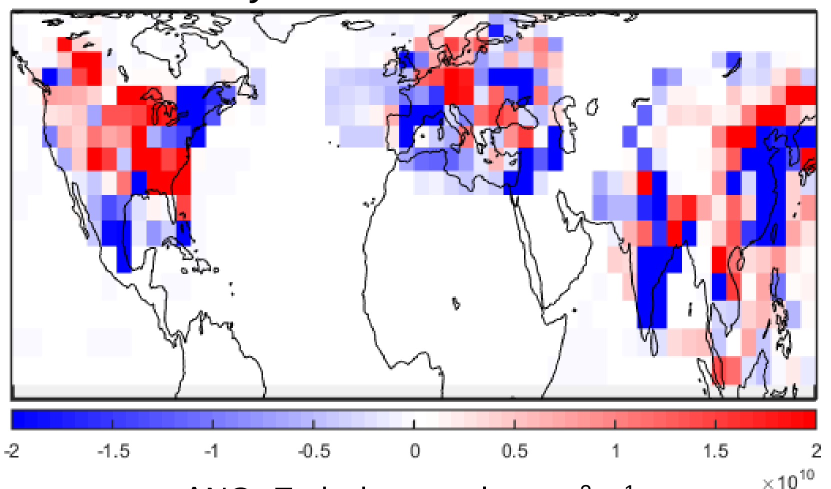
- Basic mass balance $E_{top\ down} = \alpha\Omega_{obs}$
- Actual relationship is more complicated
- Use finite difference to linearize the model around its *a priori* state
- Iterative mass balance proposed to reduce errors from transport
- Goal: To evaluate mass balance methods

Finite Difference Mass Balance Equations

$$\beta = \frac{\Delta E/_{E}}{\Delta\Omega/_{\Omega}} \qquad E_t = E_a\left(1 + \frac{\Omega_{obs} - \Omega_a}{\Omega_a}\beta\right)$$

Evaluating Mass Balance Inversions using Synthetic Observations

Sample perturbation to produce synthetic observations



ΔNO_x Emissions, molec cm^{-2} s^{-1}

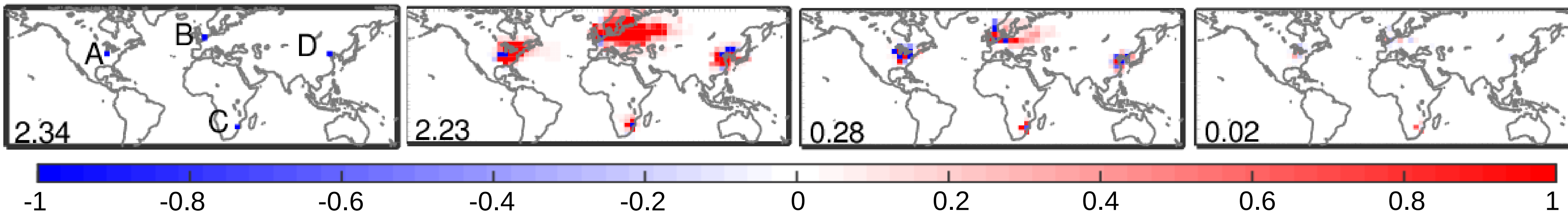
- Synthetic observations made by making specific perturbations to NO_x emissions
- Use synthetic observations to evaluate inversion methods
- Provides a known “truth” for evaluating inversion results

- GEOS-Chem Adjoint is benchmark for evaluating mass balance methods
  - Adjoint is more rigorous, but more computationally demanding

Iterative Approach Reduces Horizontal Smearing Errors

4°x5° Inversion for January 2005 (two weeks of hourly observations)

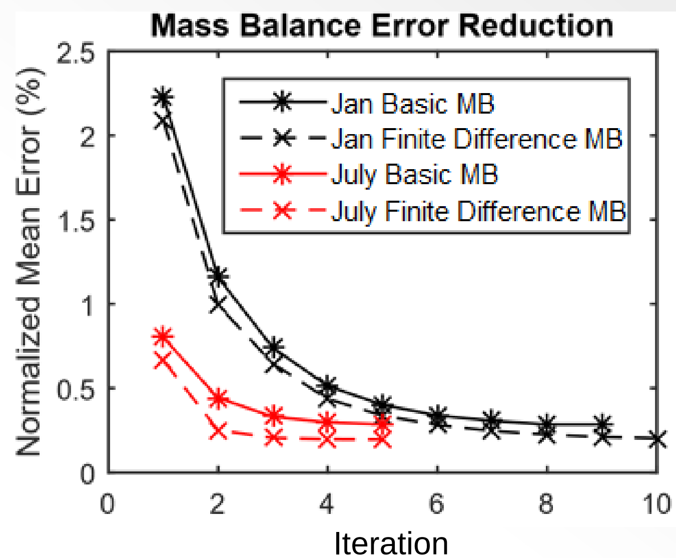
a priori – "truth"	Basic Mass Balance – "truth"	Iterative Finite Difference Mass Balance – "truth"	Adjoint – "truth"



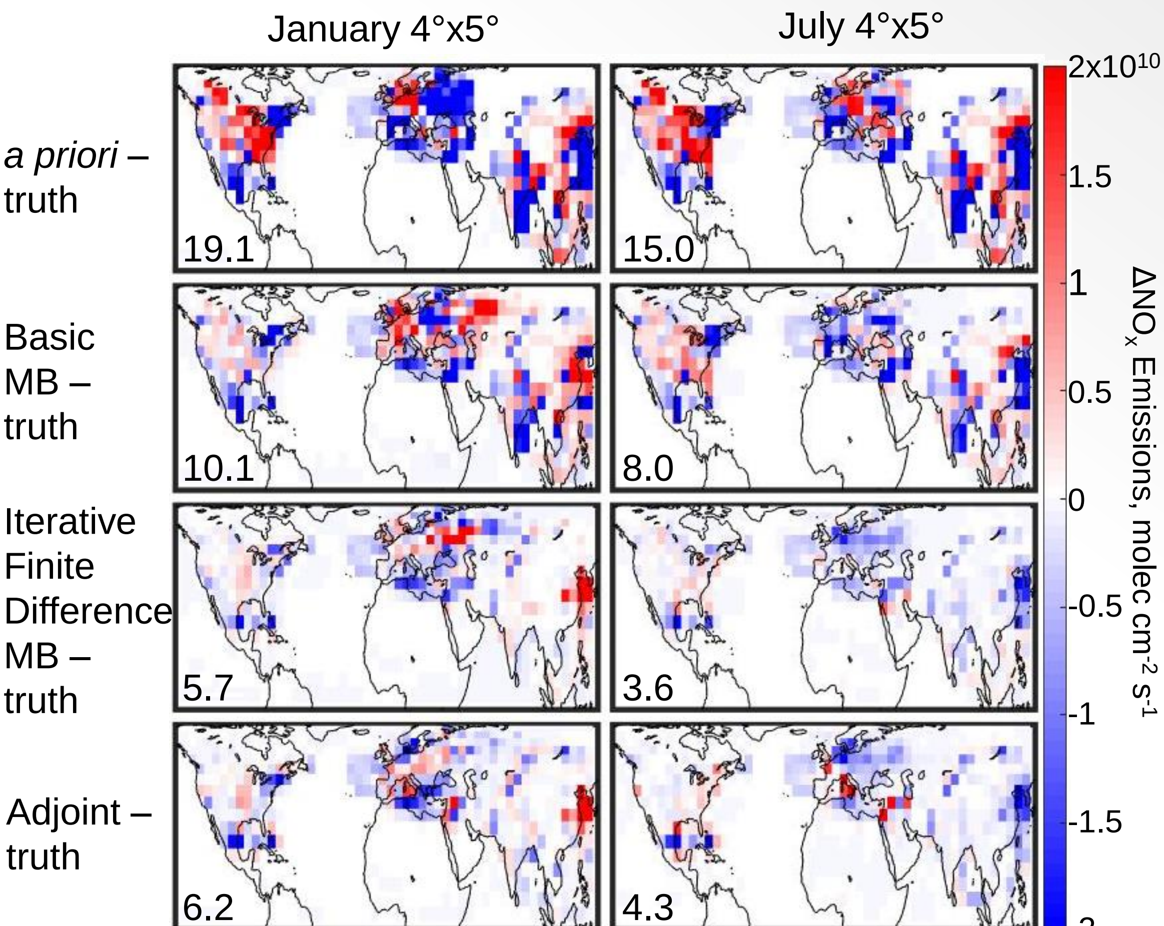
ΔNO$_x$ Emissions, molec cm^{-2} s^{-1}

Normalized Mean Error (%) inset

- Transport weakens assumptions in mass balance method
- Smearing most prevalent in winter hemisphere (longer NO$_x$ lifetime)
- Iterating the mass balance method reduces smearing
- Adjoint method best retrieves true emissions in this case



Iterative Finite Difference Mass Balance has Similar Accuracy as Adjoint for Complex Emission Perturbations



Normalized Mean Error (%) inset

- Two weeks of hourly observations
- Iterative finite difference improves basic mass balance by ~ factor of 2
- Iteration impacts January when NO_x lifetime is longer
- Finite difference impacts July when non-fossil fuel emissions contribute more to the NO_2 column

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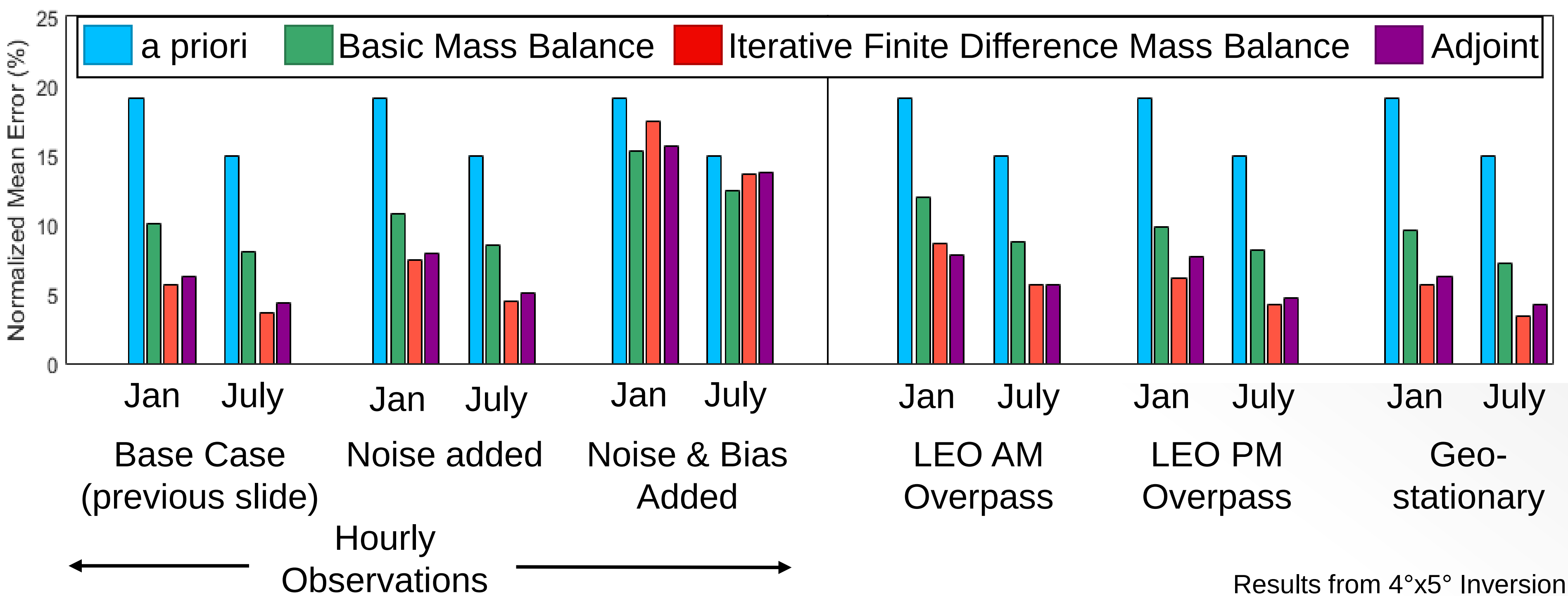
Iterative Finite Difference Mass Balance has Similar Accuracy as Adjoint

- Tested resolution: Consistent results at both 2°x2.5° and 4°x5° resolution
- Tested observational errors: Added Gaussian random noise, random bias values
- Tested sampling: Hourly observations, Synthetic LEO, Geostationary (hourly, day only)



Summary

- Iterative finite difference improves upon the basic mass balance method
- While adjoint method can better account for smearing in simplified cases, the advantage is minimal in more complex scenarios
- For more complex scenario, iterative finite difference mass balance method has same accuracy as the adjoint method

Conclusion

Iterative finite difference mass balance offers ability to estimate top-down NO_x emissions in a computationally efficient manner

Cooper et al. (2017), J. Geophys. Res. Atmos., 122, doi:10.1002/2016JD025985