Transport of NO_y from the United States

Yuanyuan Fang[1](yfang@princeton.edu), Arlene Fiore[2], Larry Horowitz[1,2], Hiram Levy[2], Yongtao Hu[3],Armistead Russell[3]

[1] Program in Atmospheric and Oceanic Sciences, Princeton University, [2] Geophysical Fluid Dynamics Laboratory, NOAA, [3] School of Civil and Environmental Engineering, Georgia Institute of Technology

1. Introduction

Motivation:

- The United States is a major source of anthropogenic nitrogen oxides (NO_x) (22% of the global anthropogenic emissions) [Olivier and Berdowski, 2001]
- Reactive nitrogen (NO_y) exported from the United States contributes to downwind O_3 production
- Powerplant NO_x emissions have decreased from 1999 to 2003 by 50% [Frost et al.,2006]
- Lightning NO_x emissions may be higher than previous studies (e.g., 0.27 rather than 0.068 TgN for the Intercontinental Chemical Transport Experiment North America (INTEX-NA) period) [Hudman et al., 2007]

Objective of this work:

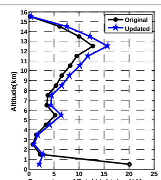
- Quantify the NO_y budget and export from the United States during Summer 2004
- Investigate the budget sensitivity of NO_y to both the surface emission change and the lightning emission change
- Examine how NO_x emission changes affect O_3 chemistry and air quality

2. Model Description

- **Model of Ozone of Related Tracers** (MOZART4) [Emmons et al., in prep] is updated from MOZART2 [Horowitz., et al., 2003] with aerosol chemistry [Tie et al., 2003]
- **Model Resolution:** 1.9° latitude by 1.9° longitude, 64 vertical levels
- **Meteorological fields** are achieved from Global Forecast System every three hours
- **Emissions:** global anthropogenic, biomass burning and natural emissions are from Horowitz., et al. [2007]

Lightning adjustment

- Increase lightning nitrogen (N) emissions over the Northern Hemispheric (NH) mid-latitude continent by a factor of 10
- Redistribute the N mass vertically by increasing the Free Troposphere (FT) N from 80% to 98%



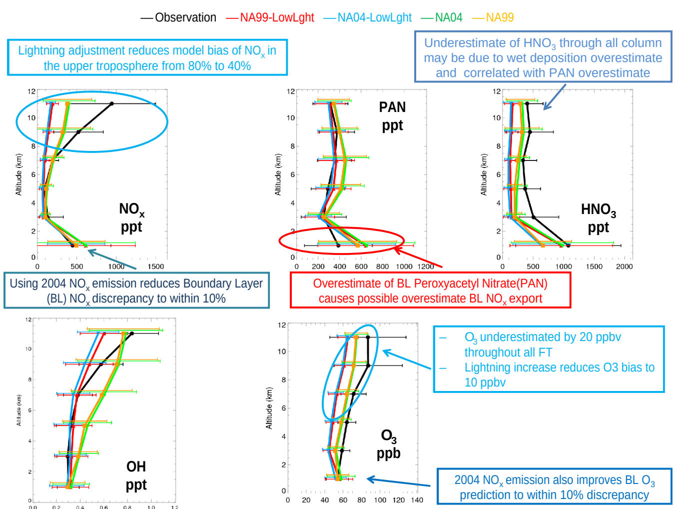
Anthropogenic emissions within the US during Summer 2004

National emission Inventory 1999 V3 (NEI99) → NEI99 with 2004 NO_x emission [Hu et al. , 2007]

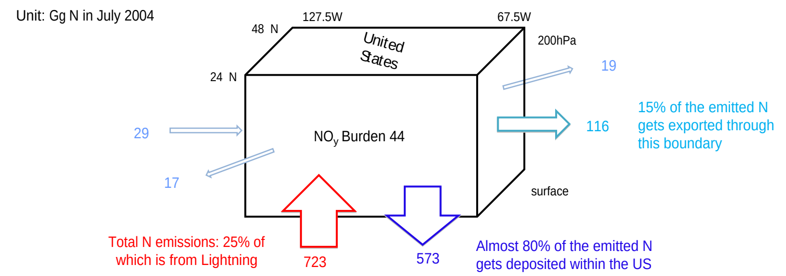
Simulation	NA99-LowLght	NA99	NA04	NA04-LowLght
Surface NO_x emission	0.68[1]	0.68[1]	0.52[2]	0.52[2]
Vertical distributed NO_x emission[3]	0.038[4]	0.20[5]	0.20[5]	0.038[4]

[1] using NEI99 [2] using 2004 anthropogenic surface NO_x emissions and emissions of other species from NEI99 [3] includes biomass burning NO_x emission, aircraft and lightning NO_x emission [4] original ightning emissions [5] lightning emissions scaled up and redistributed by a modified profile

3. Model Evaluation



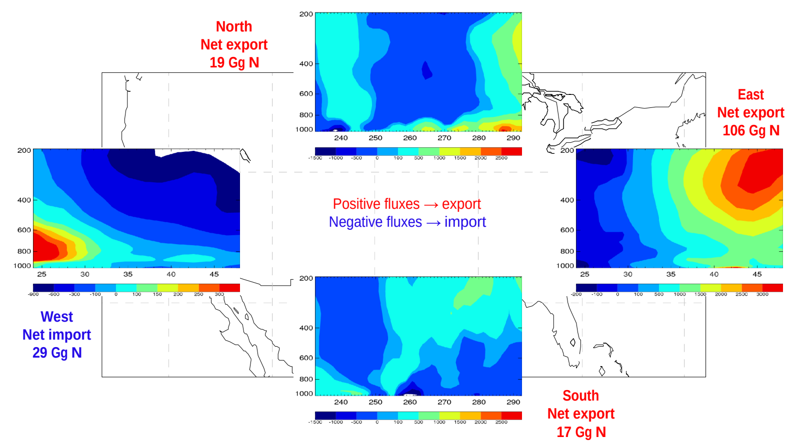
4. NO_y Budget for July 2004 (Result from NA04)



5. Major NO_y Export Pathway Pattern for July 2004 (Result from NA04)

NO_y export fluxes through different boundaries

Unit: 10^{-14}moles N cm^{-2} sec^{-1}



6. Budget Sensitivity to NO_x Emission Change

ΔExport and burden of NO_y	less than	Δ NO_x emissions (anthropogenic/lightning)
Δ deposition of NO_y	more than	

The non-linearity is caused by the NO_x-limited chemistry during summertime

For example:

Reduce NO_x emissions → Reduce Hydroxyl radicals (OH) → Decrease HNO_3 contribution to NO_y / Increase NO_y lifetime → Less reduction of NO_y burden / Less reduction of NO_y export / More reduction of NO_y deposition

The non-linearity indicates that anthropogenic NO_x emission reductions are

–Less effective to improve air quality and pollutant export

–more effective to reduce deposition

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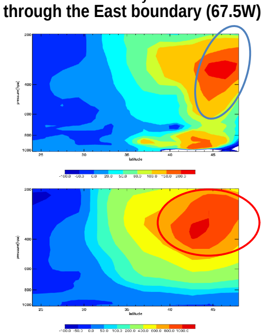
7. Sensitivity of the Major Transport Pathway to NO_x Emission Change

Changes to NO_y export flux through the East boundary (67.5W)

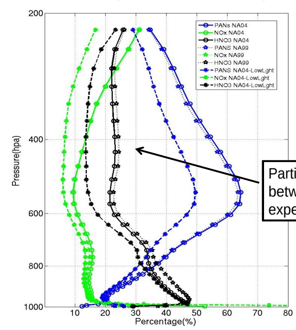
Surface emissions decrease by 23%

Lightning emission scaled up by a factor of 10



Unit: 10^{-14}moles N cm^{-2} sec^{-1}

Percentage contribution to NO_y exported through the East boundary



– Surface emissions most strongly affect export fluxes near the North boundary around 300 hPa
→ frontal lifting impacts associated with cyclone passages and strong winds
– Lightning emissions most strongly affect export at the position of the background flux maximum the
→ combined influences from strong wind and strong source change
– HNO_3 /PANs contribution to NO_y changes to the same/opposite direction of emission change
→ due to the NO_x-limited chemistry

8. Implications for O_3 Production from Lightning and Anthropogenic Emission Changes

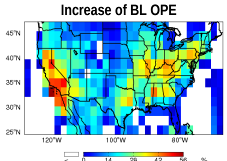
Reduce surface emissions within the U.S. (i.e., update NO_x emission from 1999 to 2004)

BL O_3 production	BL O3 Production Efficiency (OPE)	FT O_3 production	Direct O_3 export to the FT
Reduce 1.7 Gmol d^{-1}	Increase 4.8	Reduce 0.4 G mol d^{-1}	Reduce 0.55 Gmol d^{-1}

1/3 of BL O_3 production change exists as change of direct O_3 export to the FT

Similar contribution to FT O_3 change from changing export of O_3 precursor and change of direct O_3 export

Increase of BL OPE



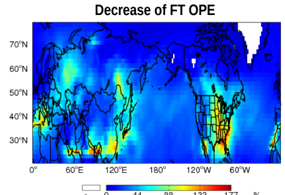
← due to

23% reduction of surface NO_x emission
↓
10% reduction of O_3 production within the BL

Adjust lightning emission over the NH mid-latitudinal continents

– Increase the FT total O_3 production from 55.45 to 73.69 Gmol d^{-1}
– Decrease the FT OPE from 56 to 39

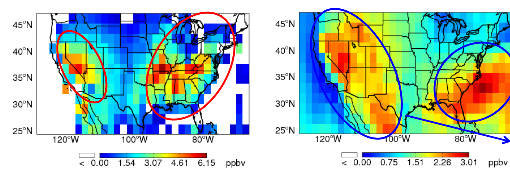
Decrease of FT OPE



If lightning NO_x is underestimated in NH middle latitude → OPE over this region is overestimated → The impact on global O_3 from the exported anthropogenic O_3 precursors is overestimated

9. Implications for Surface O_3 Concentration

Surface O_3 concentration reduction (left) /increase(right)



Update surface NO_x emission from 1999 to 2004 (23% decrease)
–Decrease of surface O_3 is strongest (as high 6 ppbv) near part of the Ohio River Valley, the Appalachia, Midwest and California

Adjust lighting emissions (a factor of 10 increase with redistribution vertically)
–Increase of surface O_3 is strongest (as high as 3 ppbv) in Southwest and over Florida

10. Conclusions

- 80% of N emitted within the United States is deposited and 15% is exported through the east boundary (as seen in Panel 4 and 5)
- NO_y export and burden respond less than proportional while NO_y deposition respond more than proportional to the NO_x emission change (as seen in Panel 6 and 7)
- Surface emission reduction causes direct O_3 export and export of O_3 precursors to decrease; these two decreases have comparable effects on FT O_3 budget (as seen in Panel 8)
- If lightning NO_x emissions is underestimated, the O_3 production efficiency is overestimated; as a result, the impact of exported anthropogenic O_3 precursors on global O_3 burden is overestimated (as seen in Panel 8)

11. References

- Fang Y, Fiore A. M., Horowitz L. W., et al (2008) Export of NOy from the United States in summer 2004 (in preparation)
- Horowitz, L. W., et al. (2003), A global simulation of tropospheric ozone and related tracers: Description and evaluation of MOZART, version 2, J. Geophys. Res., 108(D24), 4784, doi:10.1029/2002JD002853
- Horowitz L. W., et al. (2006), Observational constraints on the chemistry of isoprene nitrates over the eastern United States, J. Geophys. Res, 112, D12S08. doi:10.1029/2006JD007747
- Hudman, R. C., et al. (2007), Surface and lightning sources of nitrogen oxides over the United States: magnitudes, chemical evolution, and outflow?, J. Geophys. Res., doi:10.1029/2006JD007912

12. Acronyms and Abbreviations

BL	Boundary Layer
FT	Free Troposphere
INTEX-NA	The Intercontinental Chemical Transport Experiment
N	Nitrogen
NEI99	The National Emission Inventory 1999
NH	Northern Hemisphere
NOx	Nitrogen Oxides
NOy	Reactive Nitrogen
OPE	O_3 Production Efficiency
PAN	Peroxyacetyl Nitrate