Investigating lightning-driven interannual variability in the oxidative capacity of the troposphere

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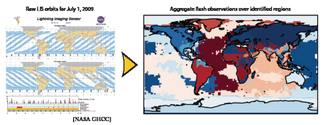
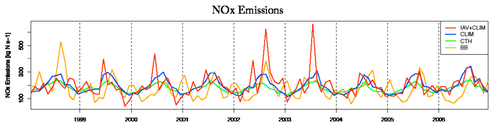
1. Background

- Lightning-sourced reactive nitrogen (LNO_x) emissions are crucial moderators for background levels of the tropospheric oxidants [e.g., *Labrador et al.*, 2004; *Sauvage et al.*, 2007; *Schumann and Huntrieser*, 2007].
- Interannual variability (IAV) in LNO_x and its influence on ozone or OH not well quantified; lightning observations networks spatiotemporally sparse.

2. Constraining interannual variability in lightning NO_x

- We constrain the LNO_x source within the **GEOS-Chem 3D chemical transport model** (v8-01-04; http://www.geoschem.org) by three different techniques for imposing variability in nine-year O_3-NO_x-CO-VOC-aerosol simulations with 6.3 Tg N a^{-1} total lightning emissions.
- (1) **(CTH) Unconstrained** physical lightning flash rate parameterization based on the cloud top height scheme of *Price and Rind* [1992]
- (2) **(CLIM) Spatial and seasonal** cycle of the parameterized flash rate at native model resolution to match the LIS/OTD High Res Monthly Climatology v2.2 from NASA MSFC (http://thunder.msfc.nasa.gov)
- (3) **(IAV+CLIM)** Continental-scale regions selected via clustering algorithm enables a more robust domain for sampling the Lightning Imaging Sensor (LIS) orbits for a given month and year to generate additional **interannual** scaling factors for the tropics

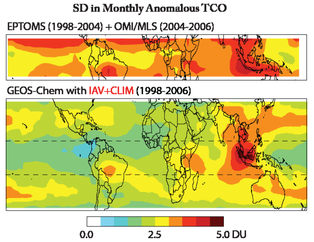


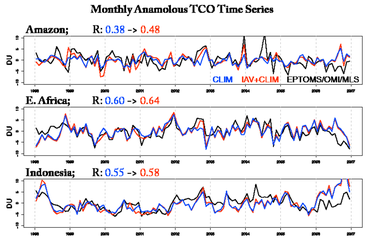


- IAV in LNO_x (σ = 0.79 Tg N a^{-1}) source is on par with biomass burning NO_x from the GFEDv2 inventory [*van der Werf et al.*, 2006]

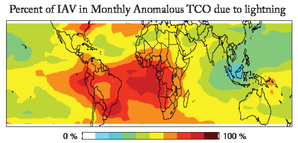
3. Ozone

- IAV apparent in the EPTOMS CCD and OMI/MLS long-term satellite obs. of tropical tropospheric ozone columns (TCO) [*Ziemke et al.*, 1998]
- Influence on tropospheric ozone disproportionately higher than surface sources because of placement in the cold, clean upper troposphere with longer lifetimes of NO_x and higher ozone production efficiencies



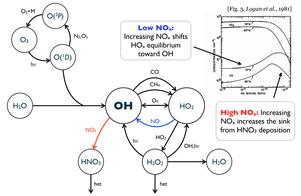


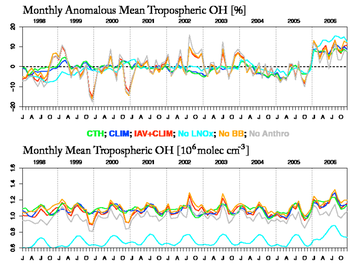
- Inclusion of IAV+CLIM constraint on lightning improves correlation of the simulation with the observations (R=**0.07** with CLIM to **0.20** with IAV+CLIM); particularly in the regions of greatest observed variability over the Amazon, Eastern Africa, and Indonesia
- The 64% increase in variability of LNO_x from CLIM to IAV+CLIM increases the variability of tropospheric ozone by 14%.
- In the IAV+CLIM simulation, lightning contributes over half the variability in Africa and the Amazon, and almost exclusively over the South Atlantic. Biomass burning and convection dominate IAV over the Maritime Continent.



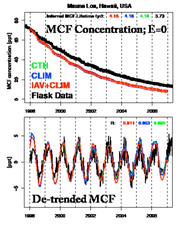
4. Hydroxyl Radical

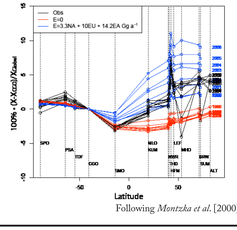
- OH extremely sensitive to variability in lightning NO_x; in low NO_x environments as the upper troposphere, OH increases with NO_x
- The 64% increase in variability of LNO_x from CLIM to IAV+CLIM increases the variability of tropospheric OH by 24%



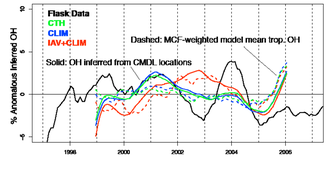


- Despite its lower absolute amounts, the location of lightning emissions allows them a **significantly higher influence on the mean and variability of OH** than either biomass burning or anthropogenic sources
- We compare against the long-term GAGE/AGAGE and NOAA CMDL methyl chloroform (MCF) observations to interpret IAV in OH by running a MCF simulation with archived OH fields with E=0 assumption
- Inferred lifetimes too short (~4.2 yr) against those inferred by station data (~5.7 yr). Combination of missing emissions and too high OH. Timeseries analysis suggests IAV+CLIM constraint is not as effective as with ozone





Following *Montzka et al.* [2000]



References

- Labrador et al., Strong sensitivity of the global mean OH concentration and the tropospheric oxidizing efficiency to the source of NOx from lightning. *Geophys. Res. Lett.* (2004) vol. 31 (6) pp. L06102
- Montzka et al., New observational constraints for atmospheric hydroxyl on global and hemispheric scales. *Science* (2000) vol. 288 (5465) pp. 500-503
- Price and Rind, A Simple Lightning Parameterization for Calculating Global Lightning Distributions. *J. Geophys. Res.* (1992) vol. 97 pp. 9919-9933
- Sauvage et al., Quantification of the factors controlling tropical tropospheric ozone and the South Atlantic maximum. *J. Geophys. Res.* (2007) vol. 112
- Schumann and Huntrieser, The global lightning-induced nitrogen oxides source. *Atmos. Chem. Phys.* (2007) vol. 7 pp. 3823-3907
- van der Werf et al., Interannual variability in global biomass burning emissions from 1997 to 2004. *Atmos. Chem. Phys.* (2006) vol. 6 pp. 3423-3441
- Ziemke et al., Two new methods for deriving tropospheric column ozone from TOMS measurements: Assimilated UARS MLS/HALOE and convective-cloud differential techniques. *J. Geophys. Res.* (1998) vol. 103 (D17) pp. 22115-22127