Glyoxal Retrieval from OMI, and Interpretation with the GEOS-Chem CTM

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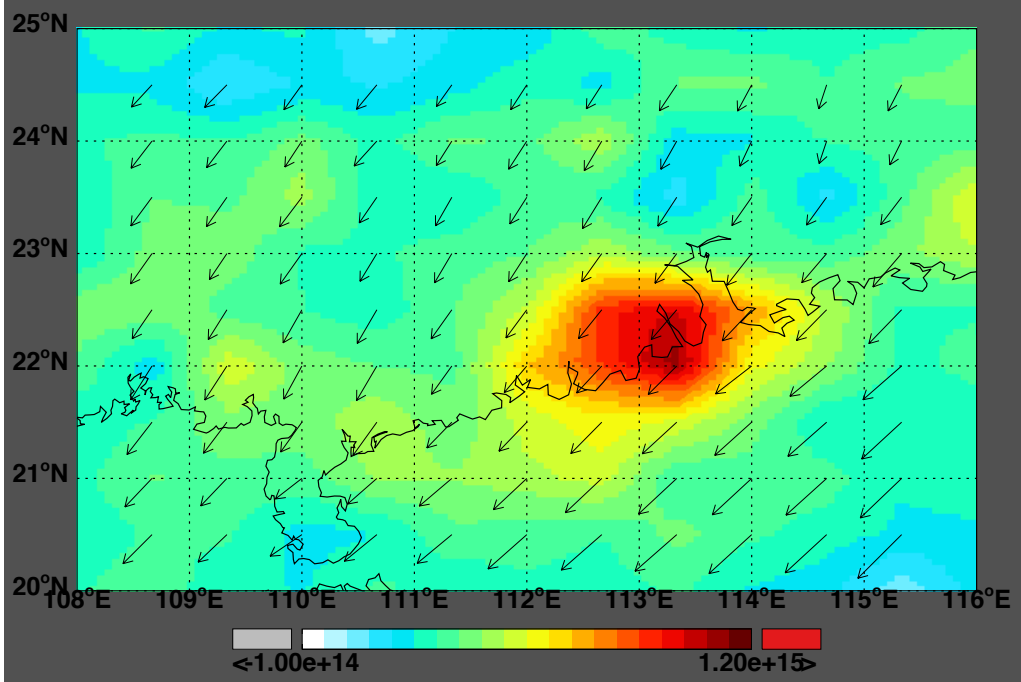


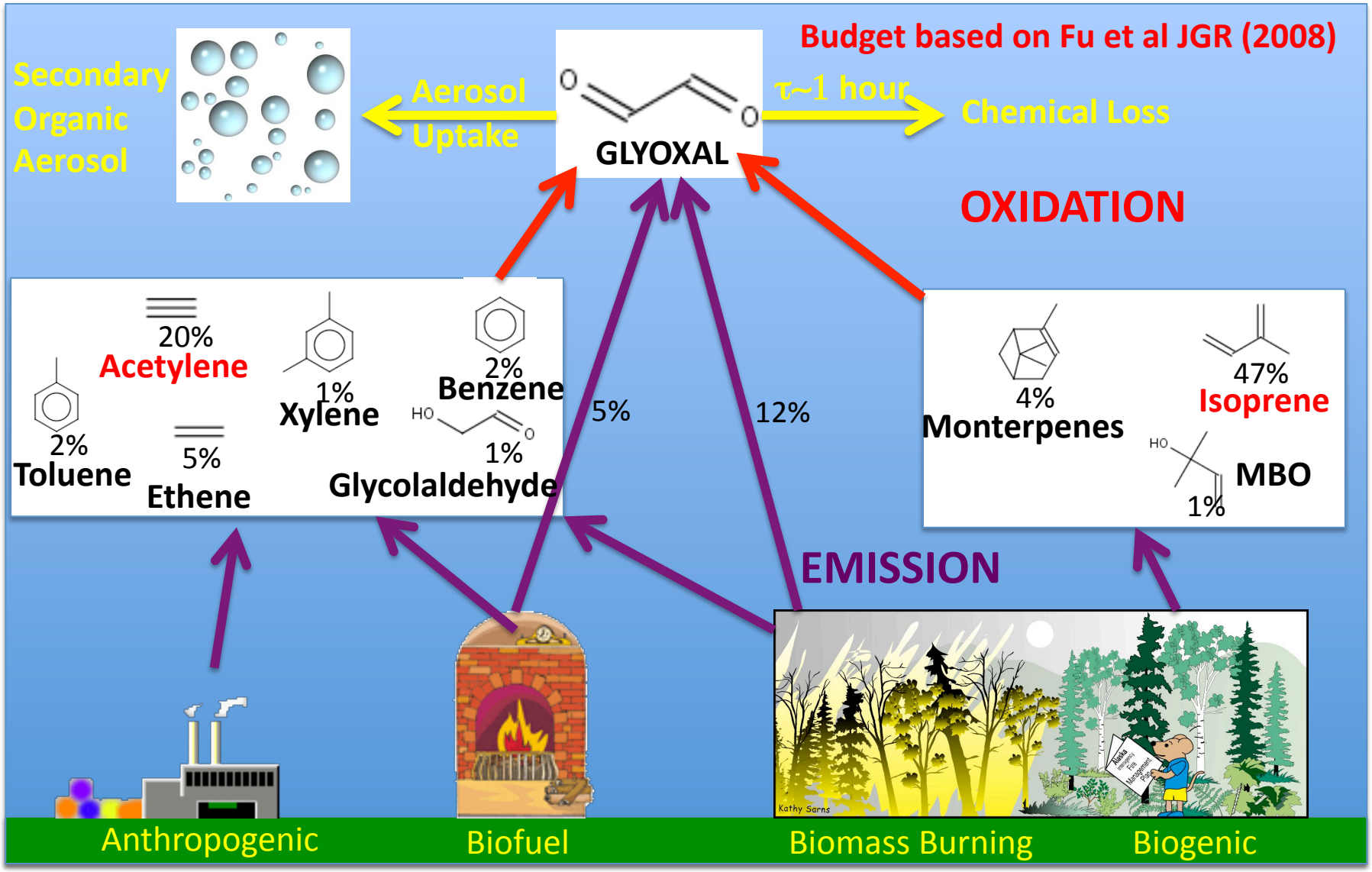
OMI Glyoxal
PRD China

HARVARD UNIVERSITY
ATMOSPHERIC CHEMISTRY MODELING GROUP

Funded by NASA/ACMAP

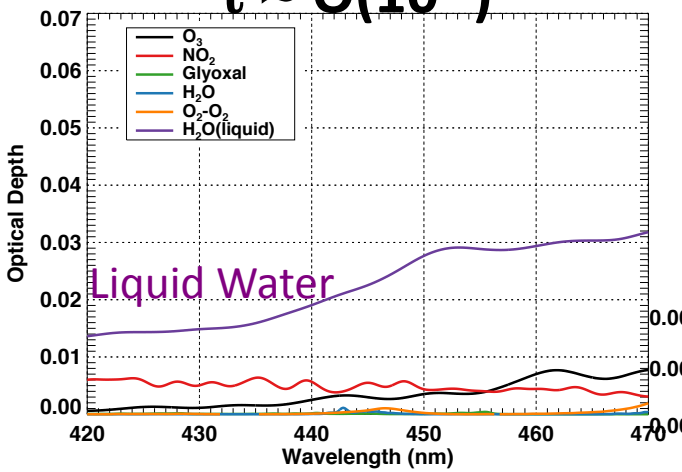
EOS Aura STM
15-18 September, 2014

Sources of Atmospheric Glyoxal



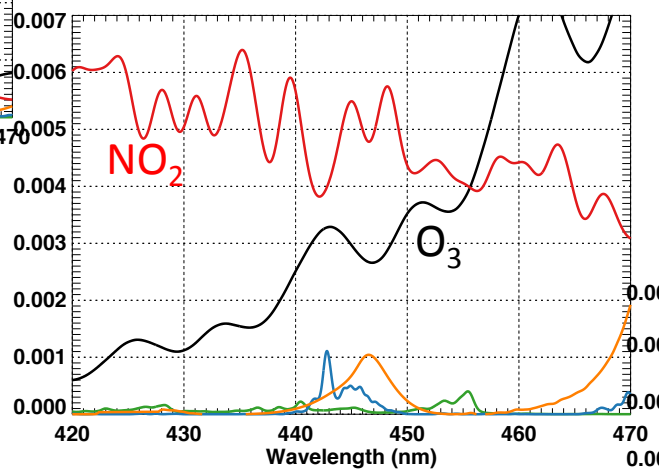
Typical Molecular Absorption Strengths in the Glyoxal Fit Region

$\tau \sim O(10^{-2})$



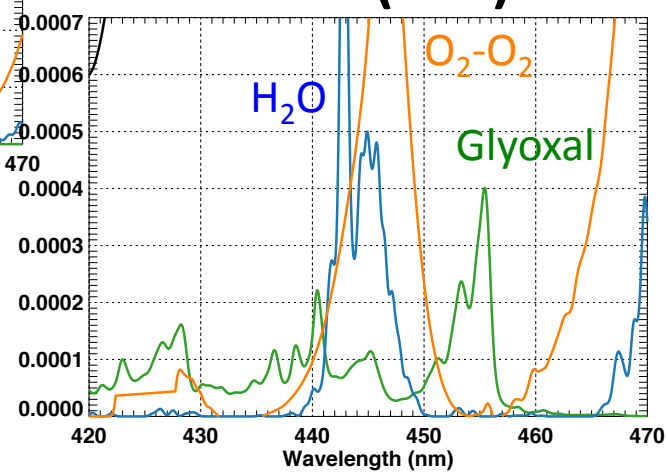
Zoom 10x

$\tau \sim O(10^{-3})$



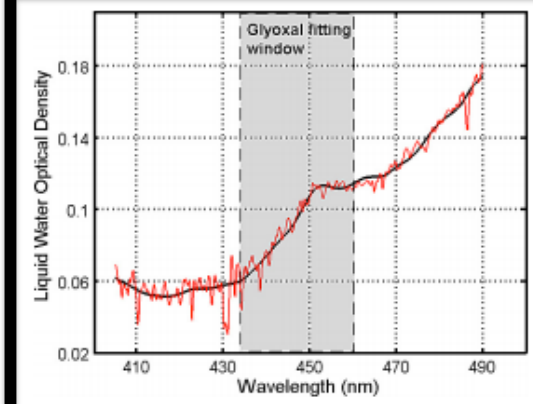
Zoom 10x

$\tau \sim O(10^{-4})$



Glyoxal absorption is at least an order of magnitude lower than interfering species

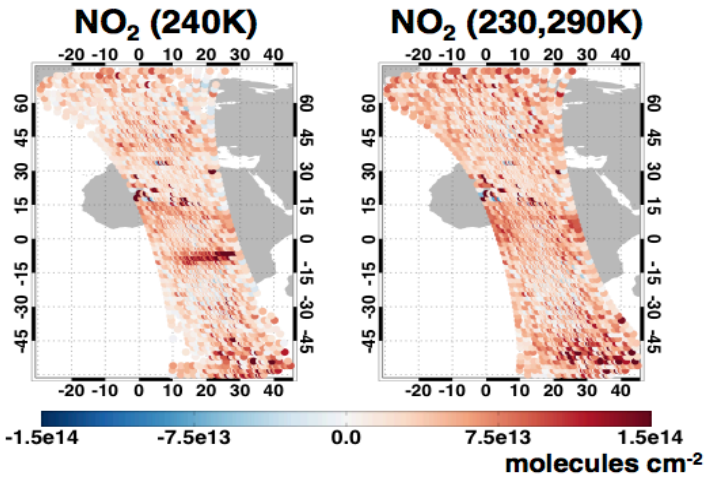
Specific Issues Resulting from Weak Glyoxal Absorption



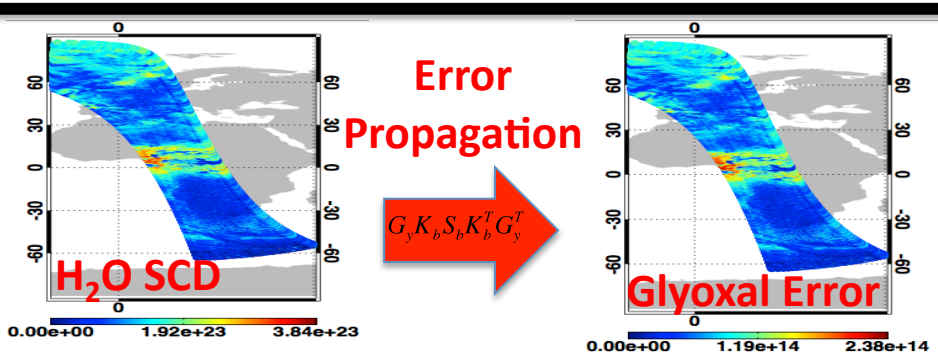
Lerot et al. (2010)

Problem: Reduced retrieval sensitivity due to strong liquid water absorption
→ Use pre-fitted liquid water optical depths

OSSE Column Error



Problem: Interference associated with cross section temperature dependence
→ Include two NO_2 cross sections in spectrum fit (Alvarado et al. 2014)

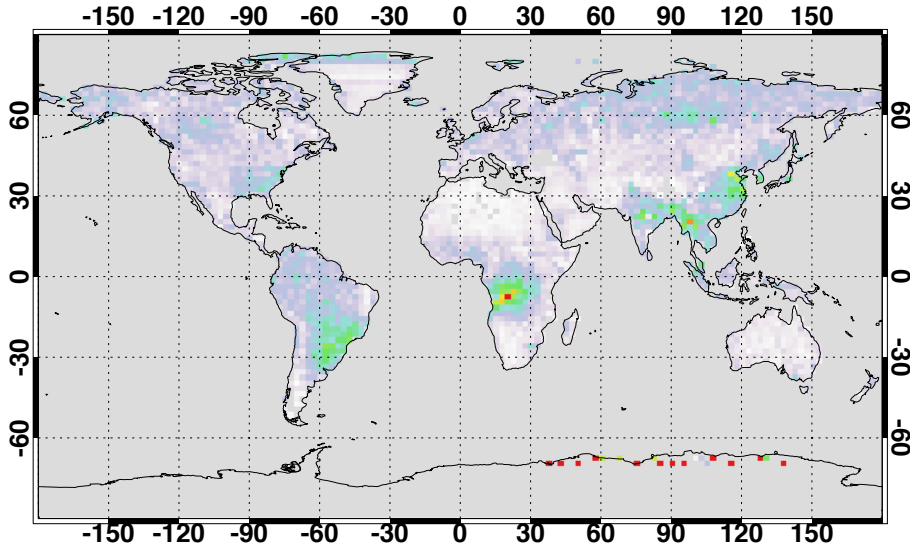


Problem: Interference associated with cross section uncertainty
→ Propagate cross section uncertainties to estimate errors

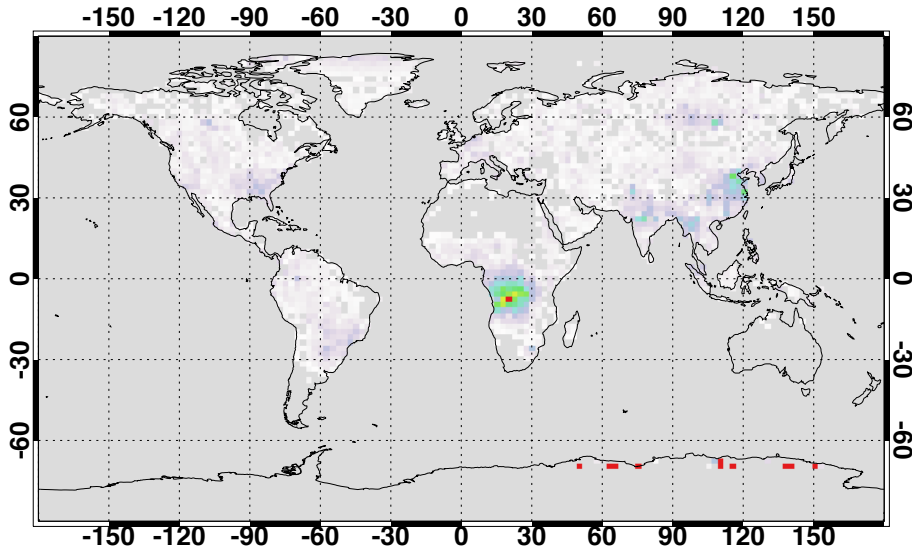
The Position of the Fit Window is a Major Cause of Discrepancy Between Algorithms

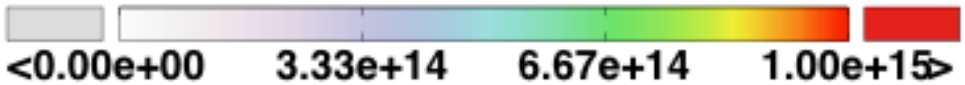
OMI Glyoxal Column Retrieved with Different Windows - July 2006

435-460nm (Lerot et al. 2010)



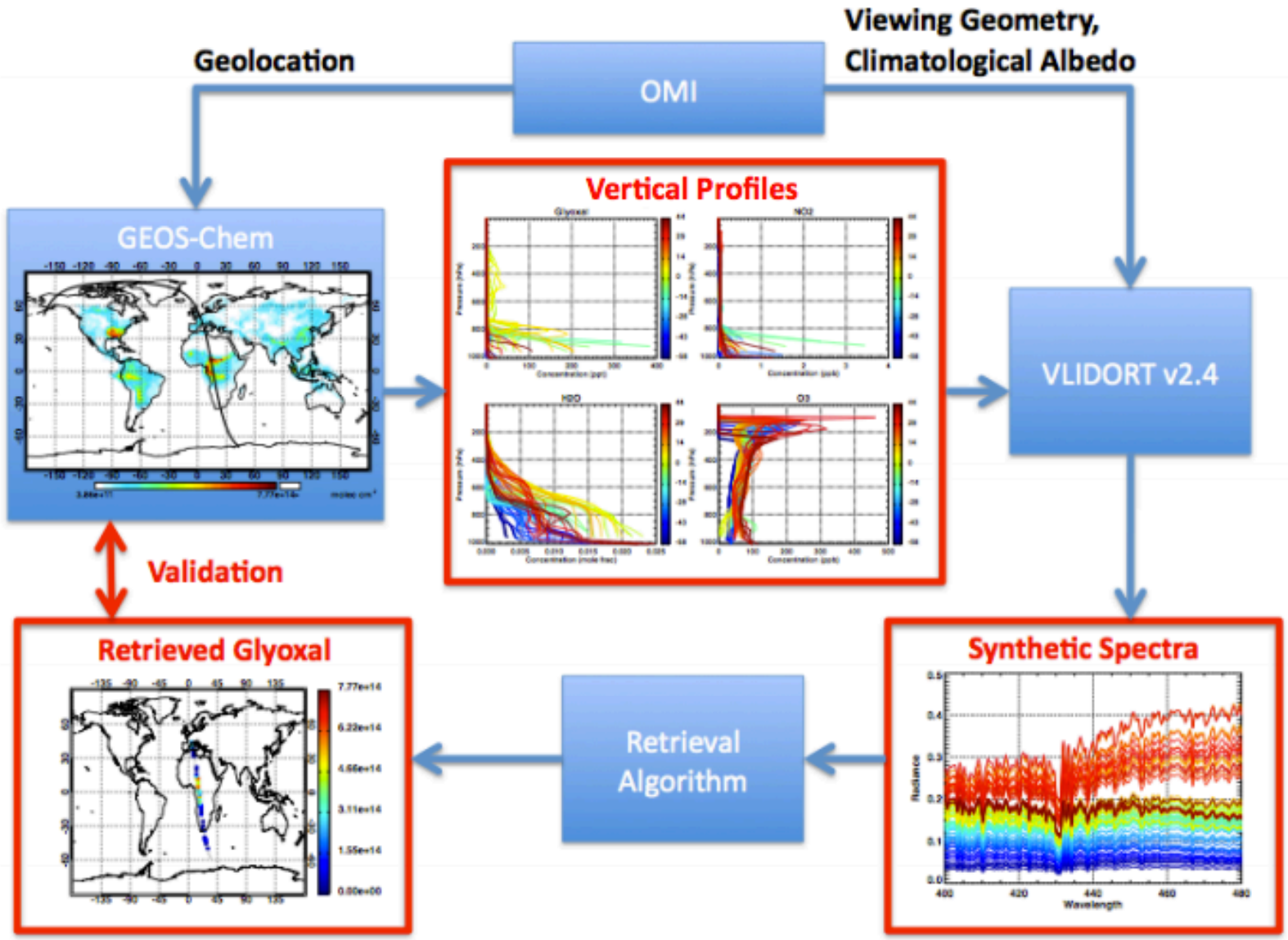
432-468nm (Kuruso et al. 2006)



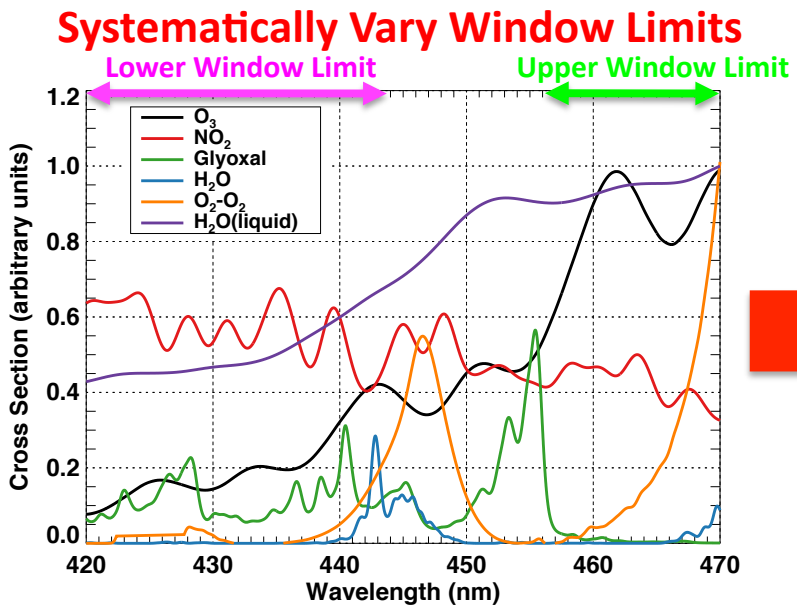


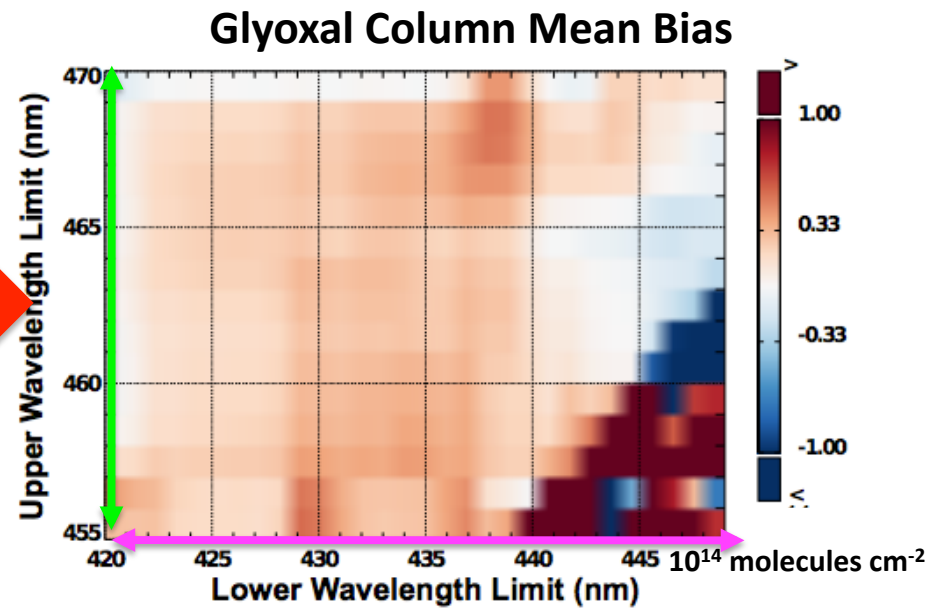
Appropriate fit window choice can be made by systematically varying upper and lower window limits

Testing Retrieval Window Sensitivity with Observing System Simulation Experiments



The Sensitivity of the Retrieval to Window Position for the OSSE is Weak

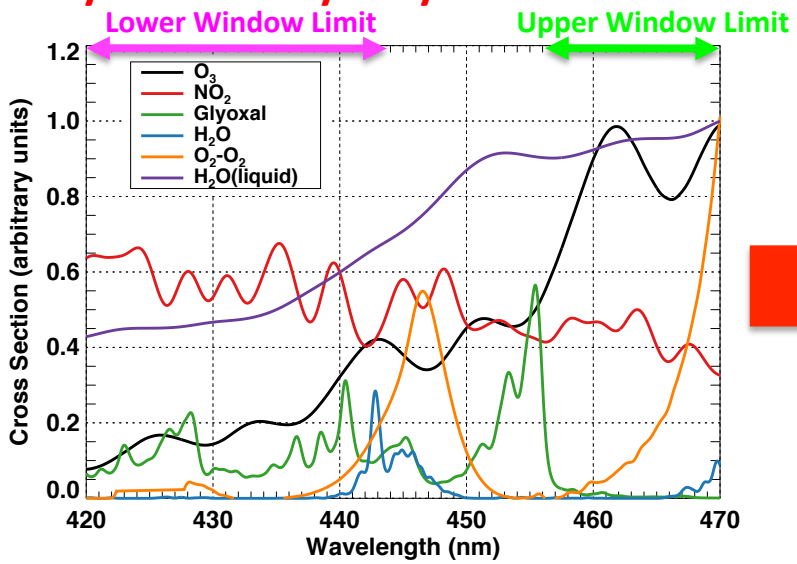




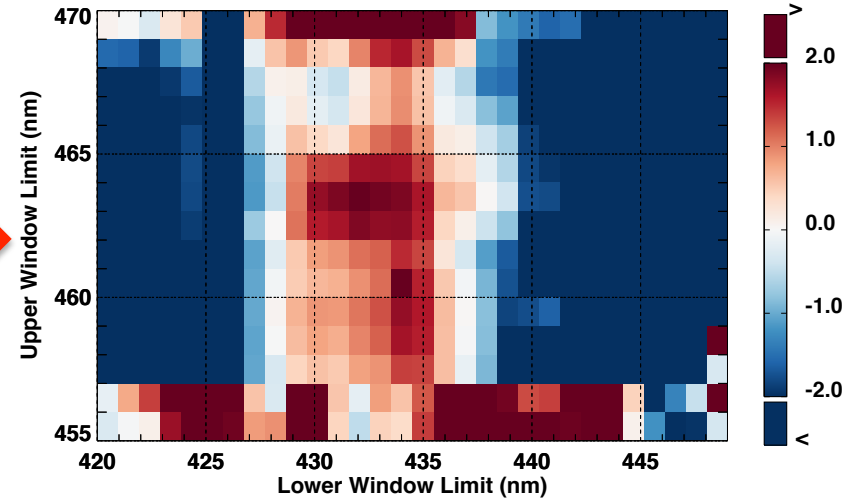
Systematic bias is small (< 10^{14} molecules cm^{-2}) for most windows

Glyoxal Window Sensitivity

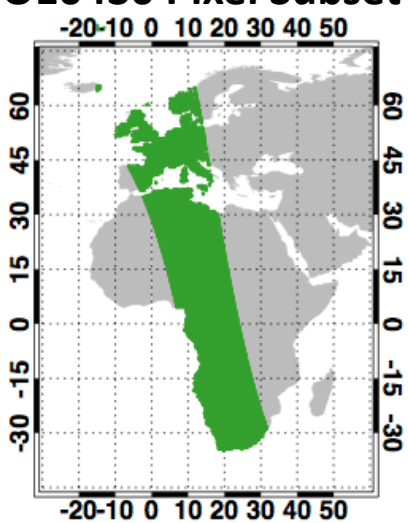
Systematically Vary Window Limits



Mean Glyoxal Slant Column (O10430 Subset)

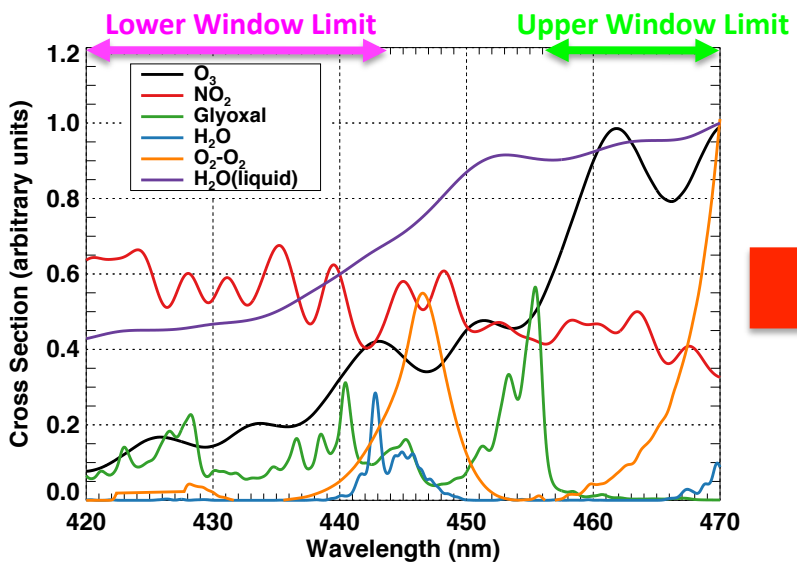


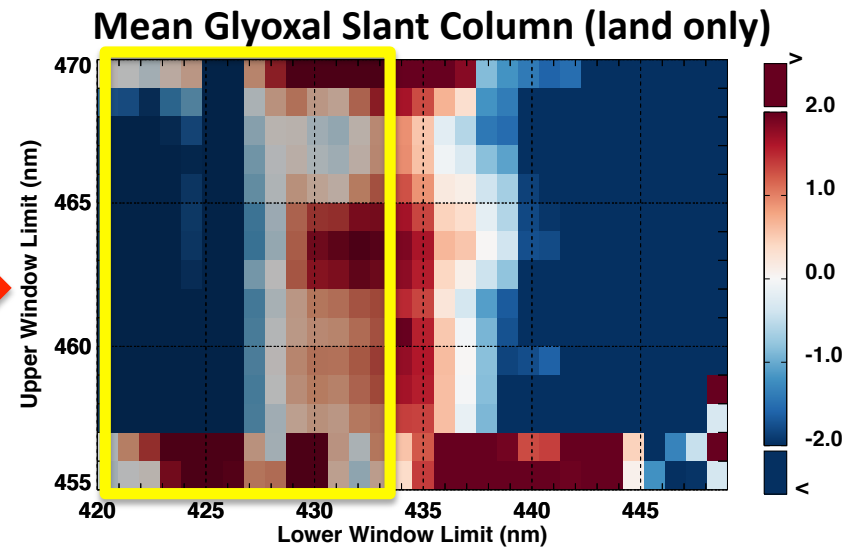
O10430 Pixel Subset



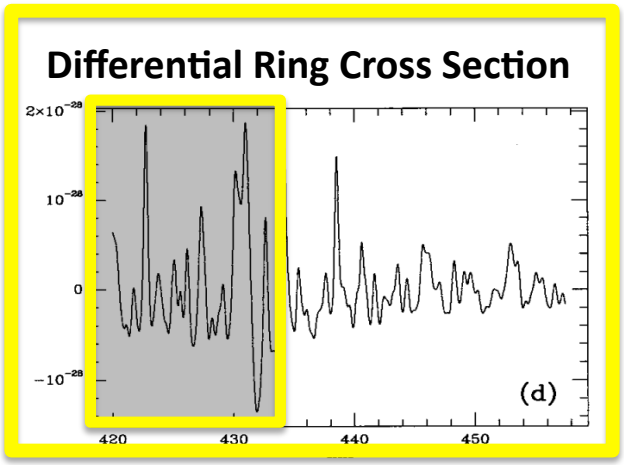
Select subset of points over land for case study orbit

Glyoxal Window Sensitivity

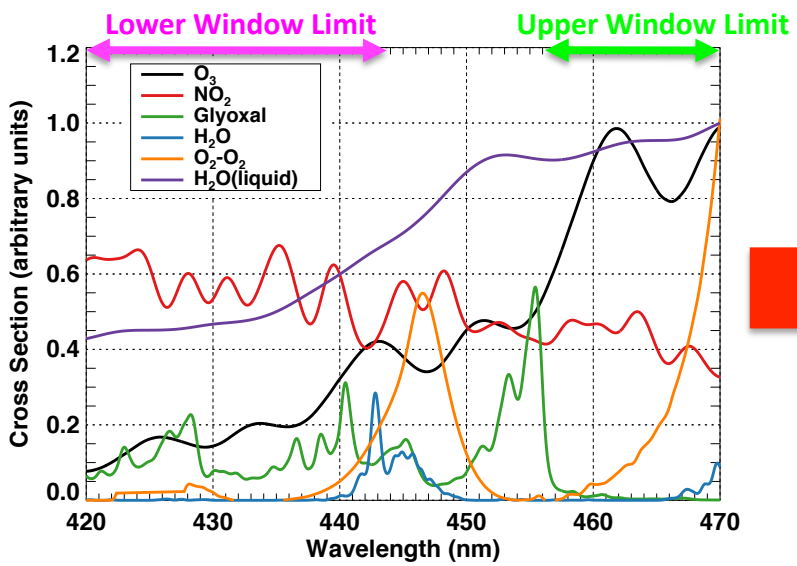


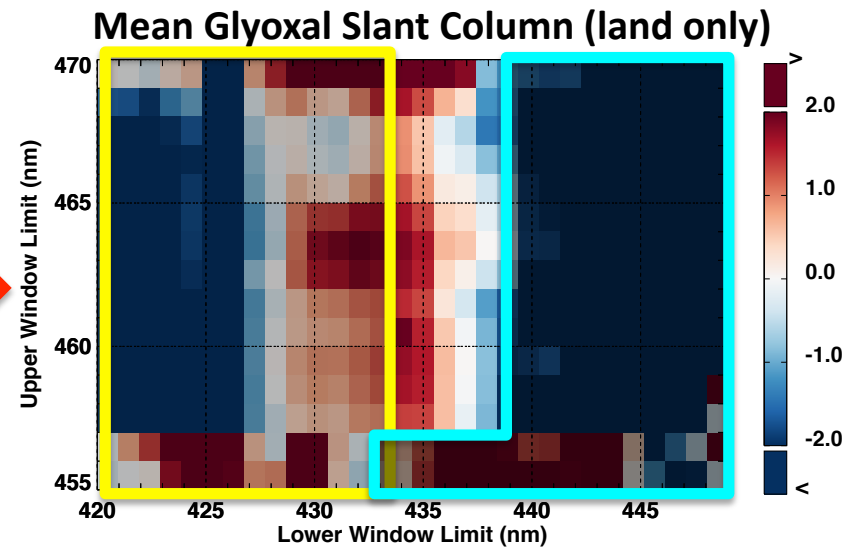


Raman Scattering

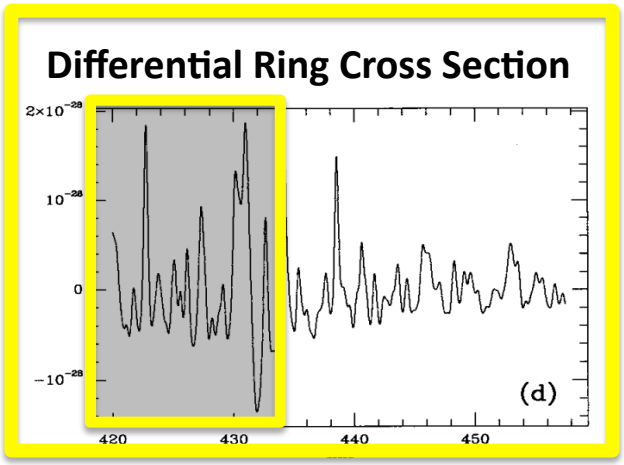


Glyoxal Window Sensitivity

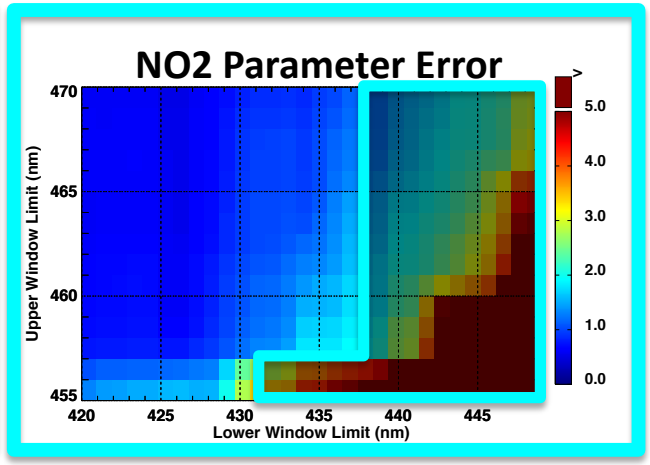




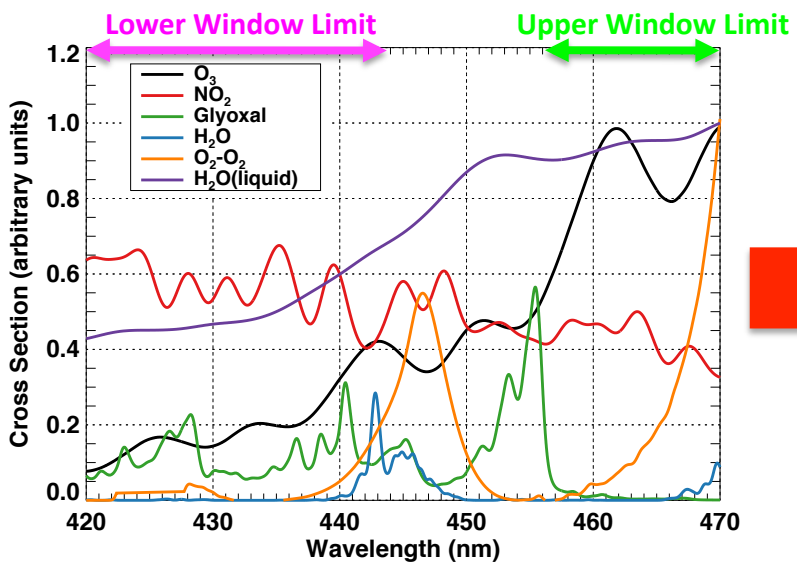
Raman Scattering

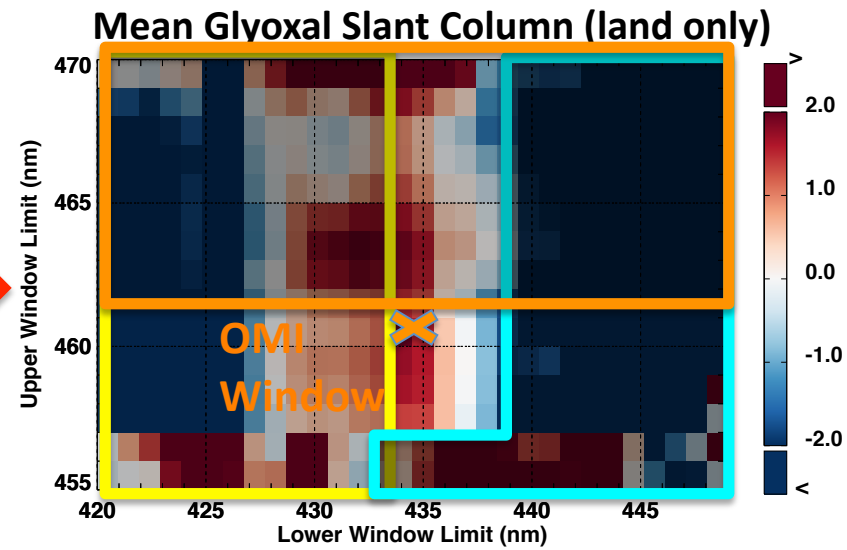


Cross Section Uncertainty

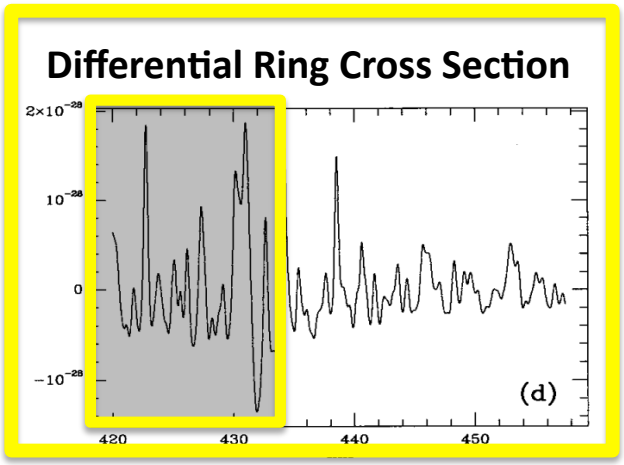


Glyoxal Window Sensitivity

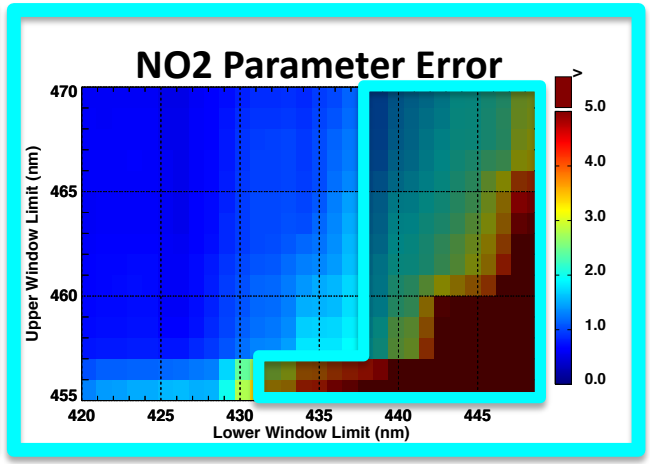




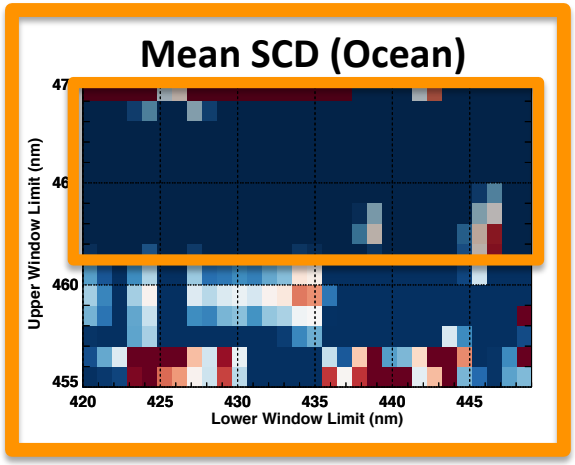
Raman Scattering



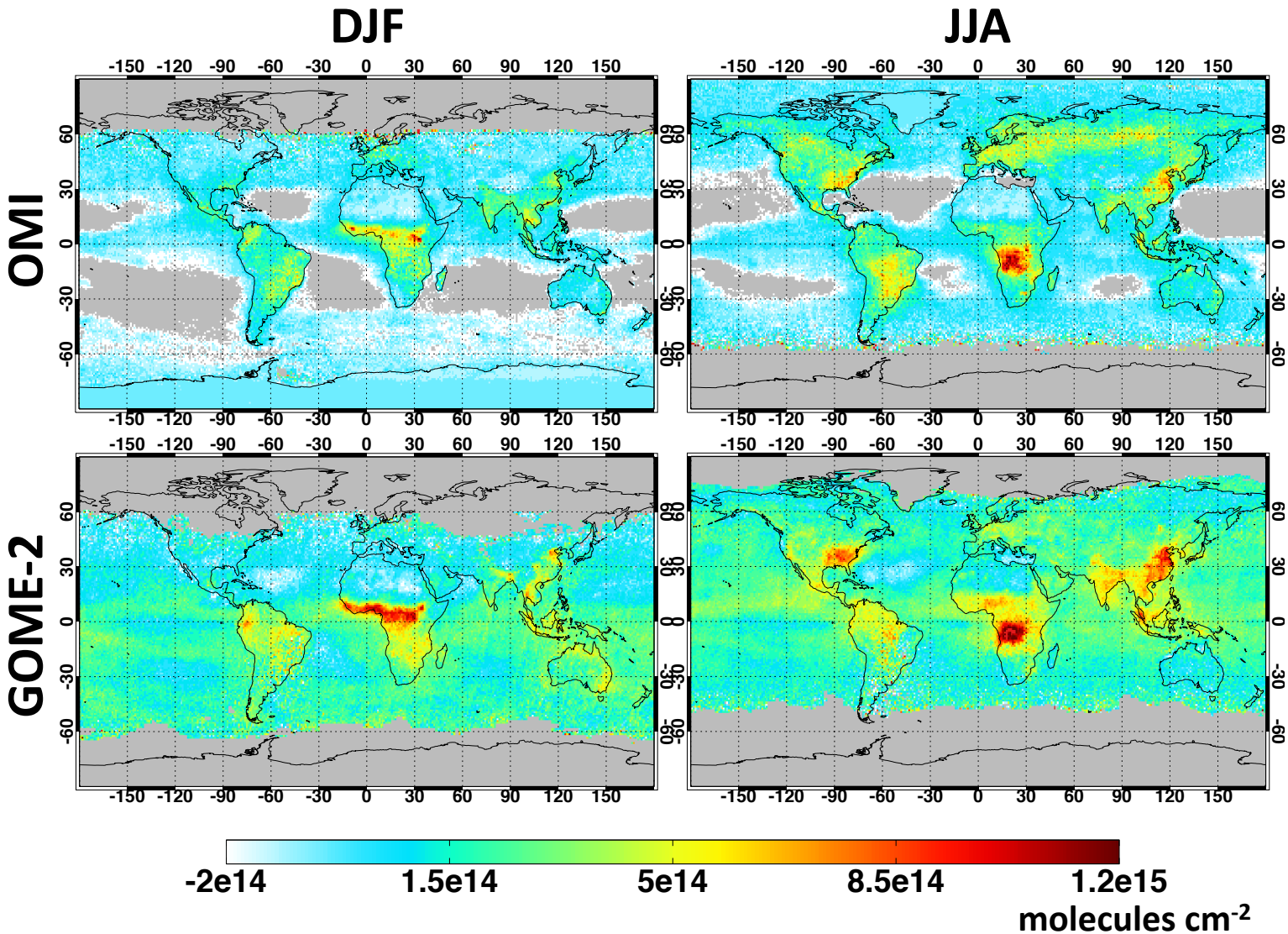
Cross Section Uncertainty



Liquid Water Interference



OMI Comparison with Lerot et al. GOME-2



GOME-2 observations are systematically higher than the OMI product

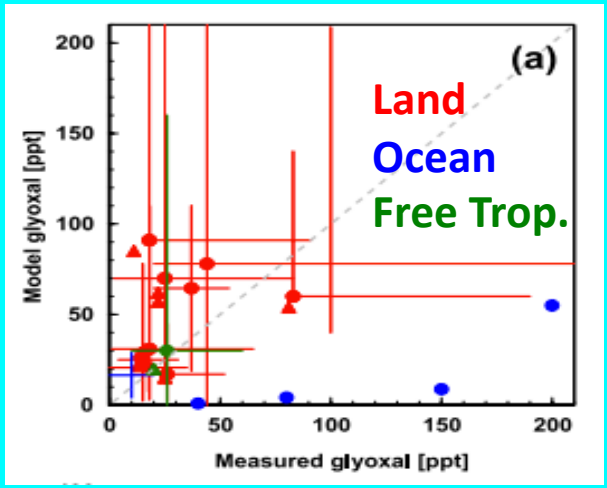
The OMI Retrieval is Consistent with Surface Observations

Current generation CTMs are not biased low relative to existing surface glyoxal observations (Fu et al. 2008)

Inversion of SCIAMACHY Glyoxal

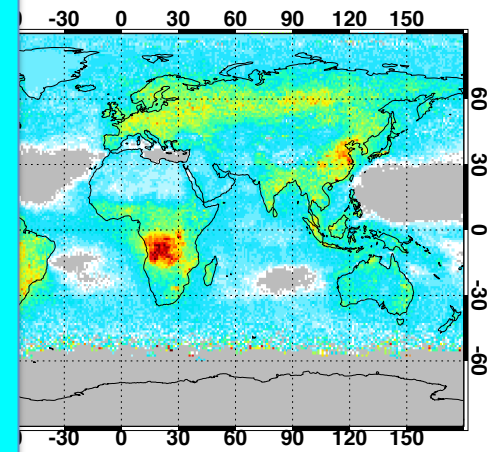
Sources	A priori	Inversion A	Inversion B
Anthopogenic			
primary	3.2	5	3.4
secondary	11.9	13.3	12.1
Pyrogenic			
primary	5.9	4.7	4.3
secondary	3.9	2.9	2.6
Biogenic	31	32.5	32
UVOC			
primary		35.6	
secondary			54
Total	56	94	108

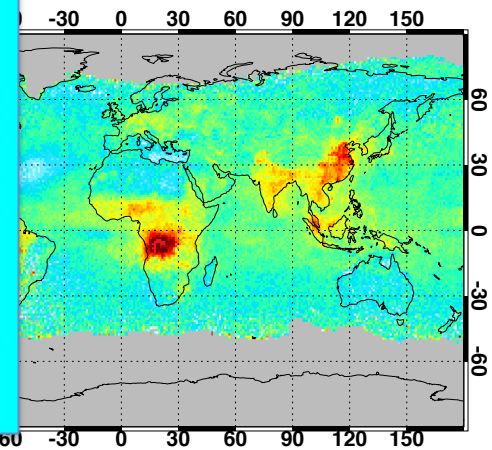
GEOS-Chem vs Meas. (Surface)

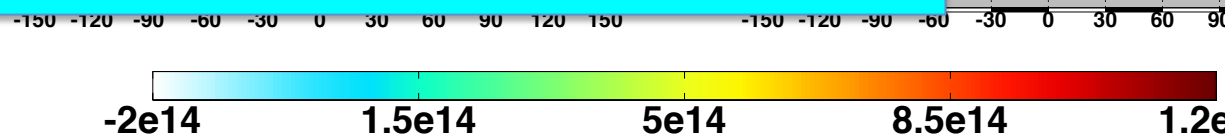


This is at odds with the satellite measurements (require doubling a-priori to match observation) (Stavrakou et al. 2009)

JJA





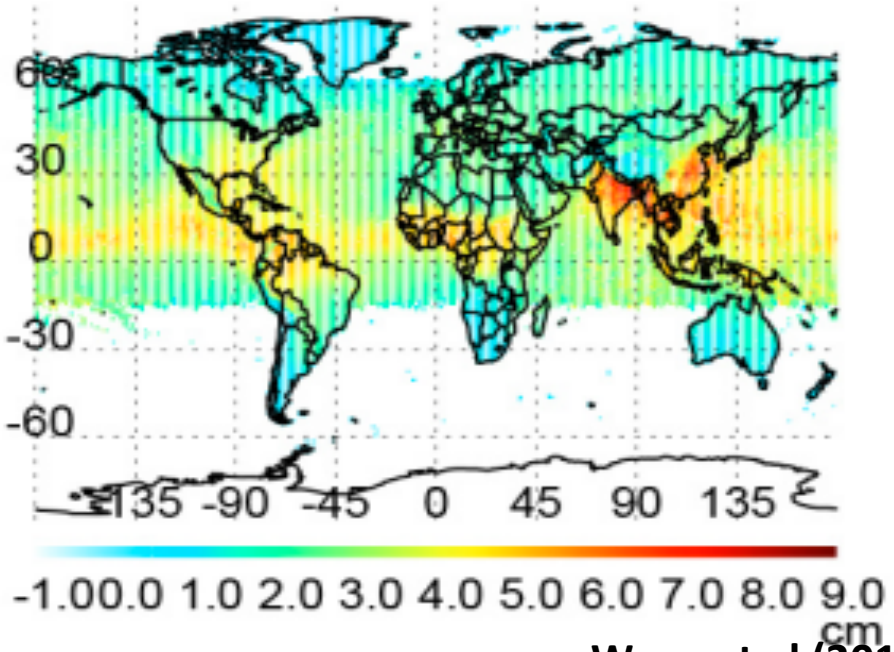


-2e14 1.5e14 5e14 8.5e14 1.2e15

molecules cm^{-2}

Glyoxal Columns in GOME-2 are Highly Correlated with Water Vapor Column Densities

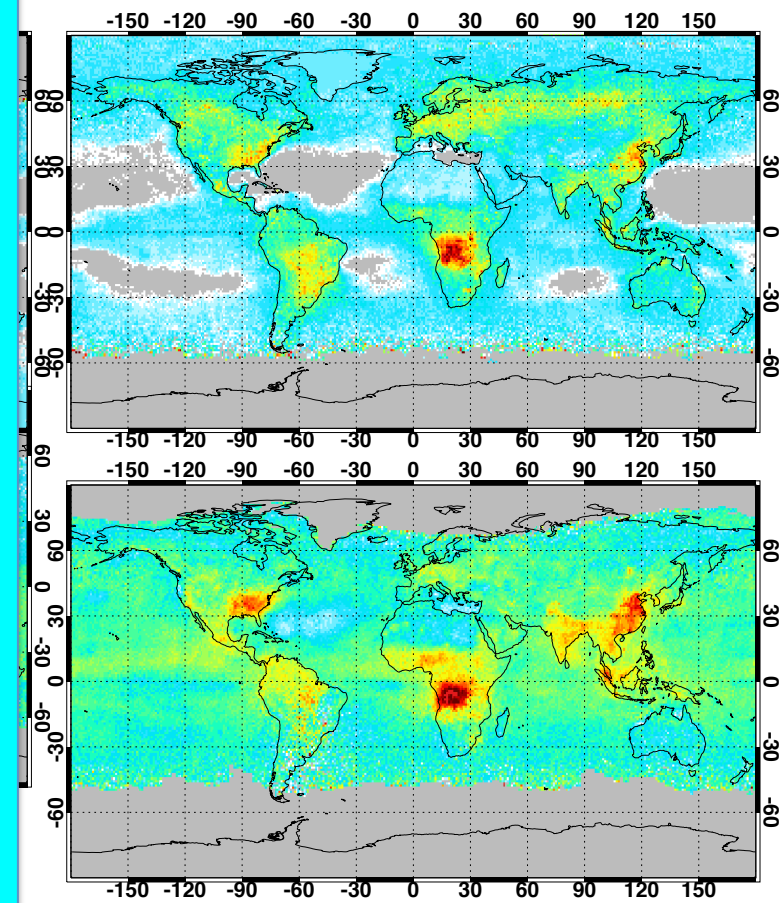
MODIS Water Vapor Columns (July 2006)



Wang et al (2014)

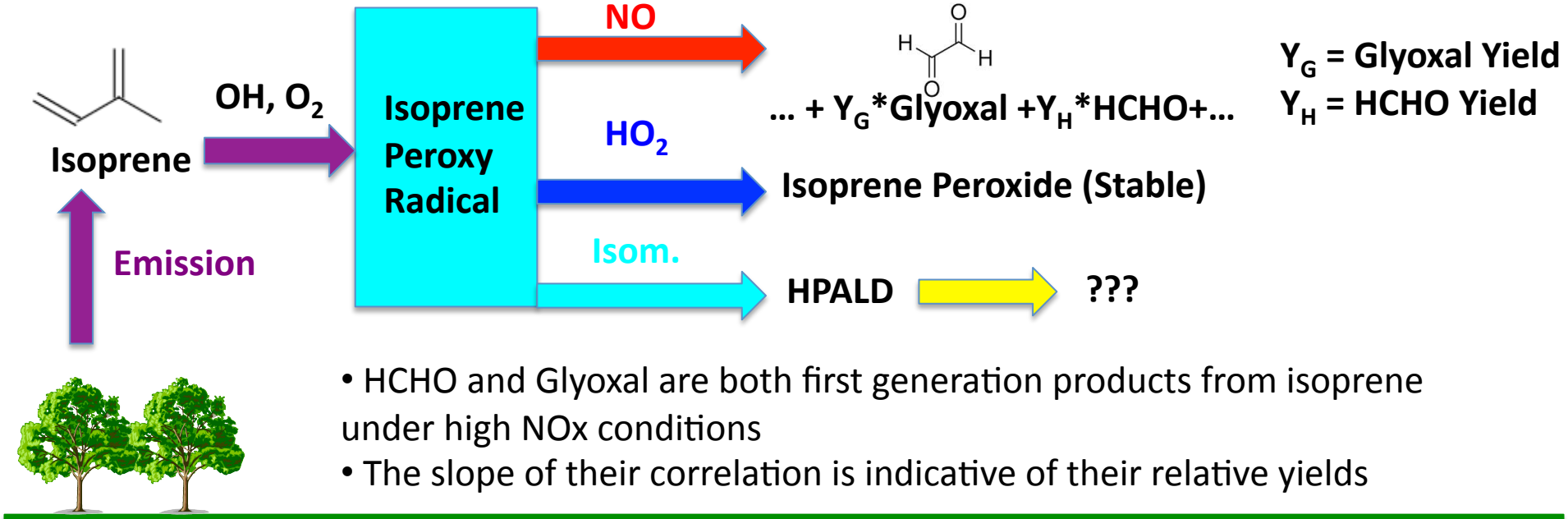
Differences may be related to HITRAN version (2005 vs 2008) and Cross Section Temperature

JJA



molecules cm^{-2}

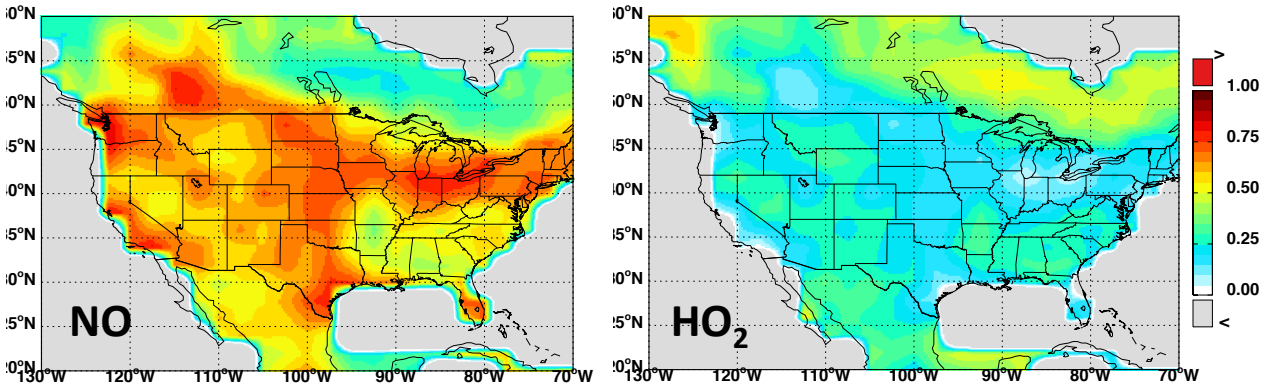
Indirect Retrieval Validation through HCHO-Glyoxal Correlations



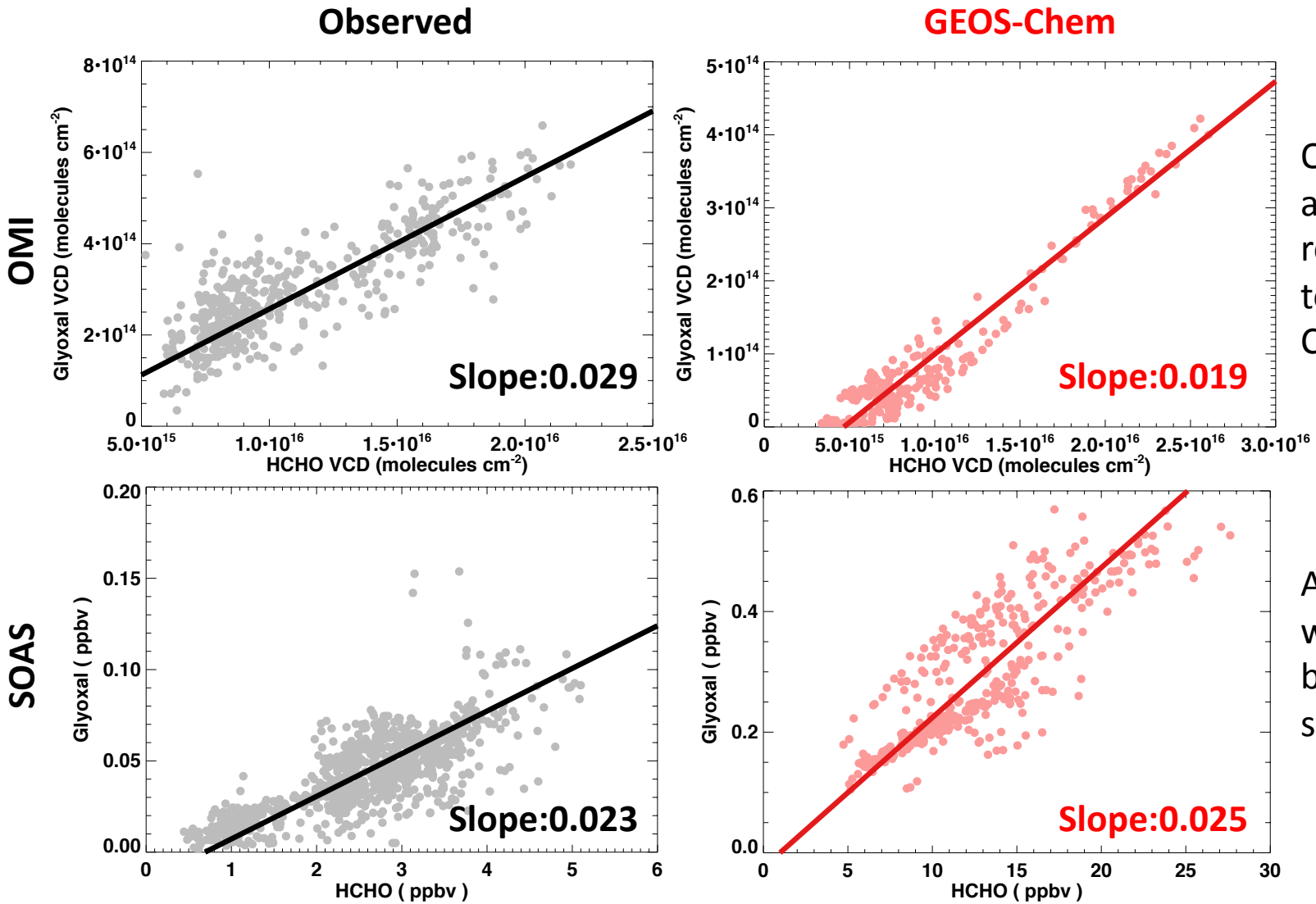
- HCHO and Glyoxal are both first generation products from isoprene under high NOx conditions
- The slope of their correlation is indicative of their relative yields

The High NO_x (NO) pathway is the dominant fate of the isoprene peroxy radical in Summer

GEOS-Chem Isoprene Peroxy Radical Branching Ratio (Summer)



HCHO-Glyoxal Correlations Over the SE United States



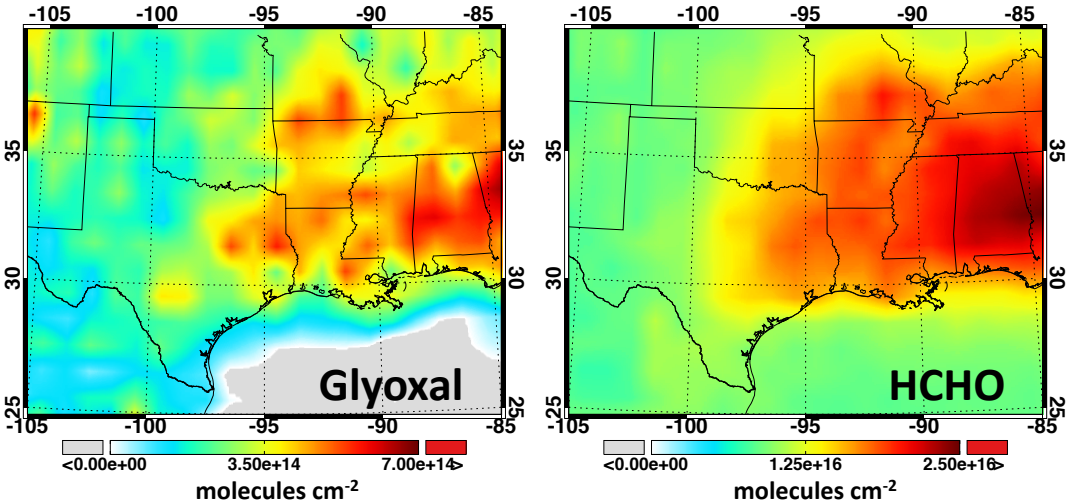
OMI suggests a higher relative yield to GEOS-Chem

Agreement with model is better at surface

Kaiser et al. (Unpublished Data)

Glyoxal Over the South East US
"Isoprene Fountain"

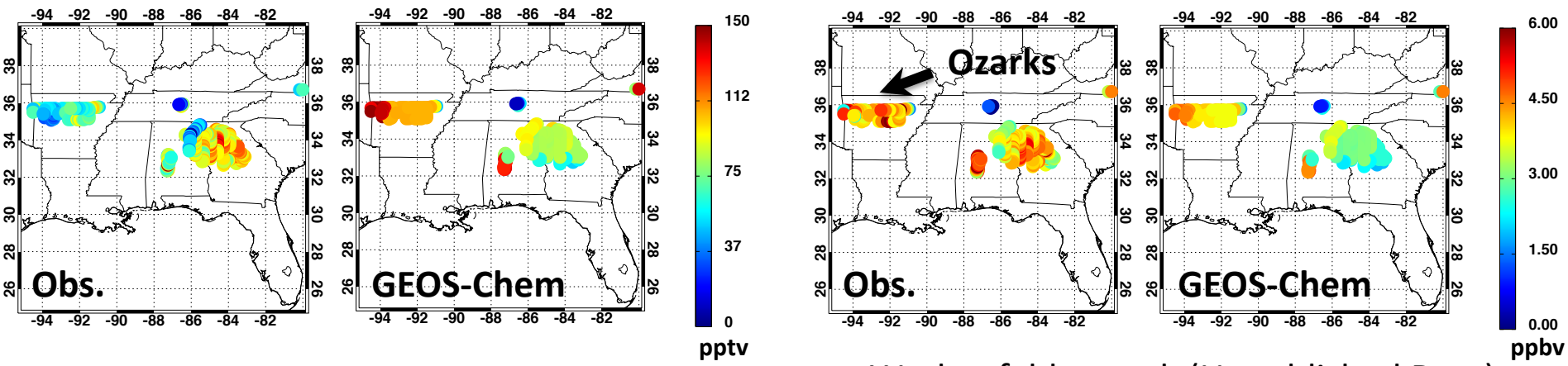
OMI VCD (Summer 2007)



The spatial distribution of glyoxal over the South East United States is more heterogenous than what should be expected from concurrent HCHO observations

SENEX Boundary Layer Observations (Summer 2013)

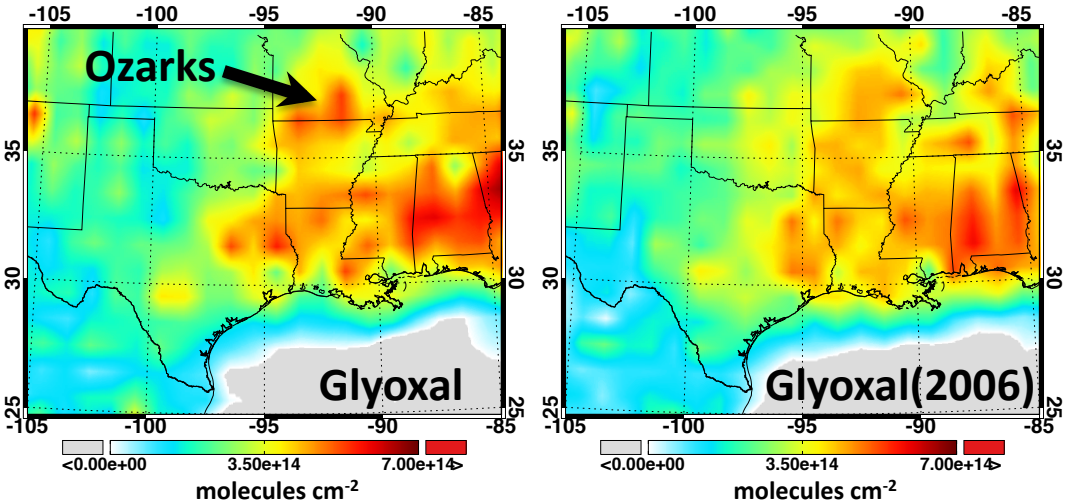
GLYOXAL **HCHO**



Washenfelder et al. (Unpublished Data)

Glyoxal Over the South East US
"Isoprene Fountain"

OMI VCD (Summer 2007)

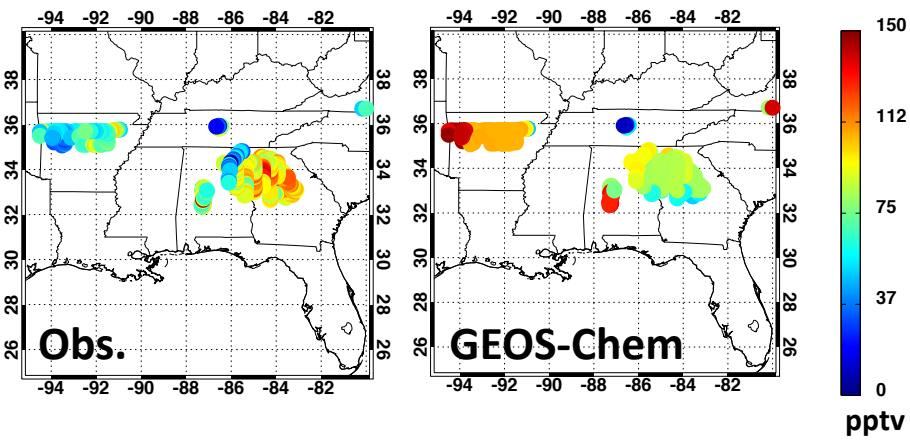


Similar glyoxal distribution for 2006 → evidence the heterogeneity is not from noise

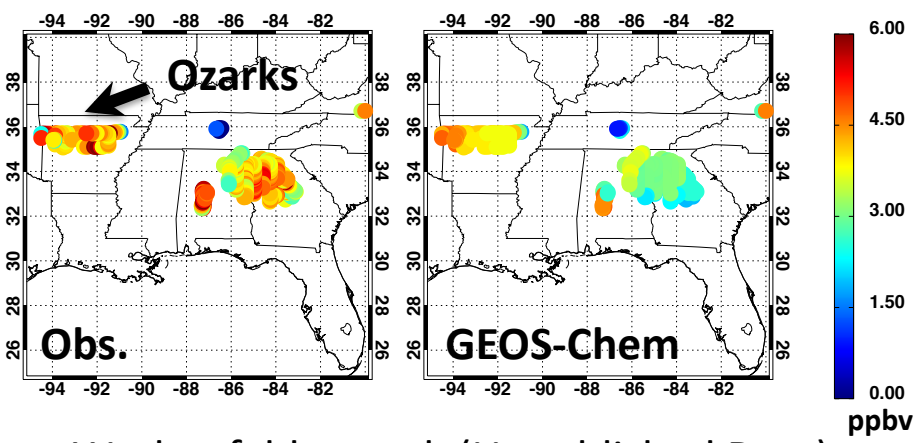
The glyoxal "hole" over the Ozarks is also present in SENEX aircraft data

SENEX Boundary Layer Observations (Summer 2013)

GLYOXAL



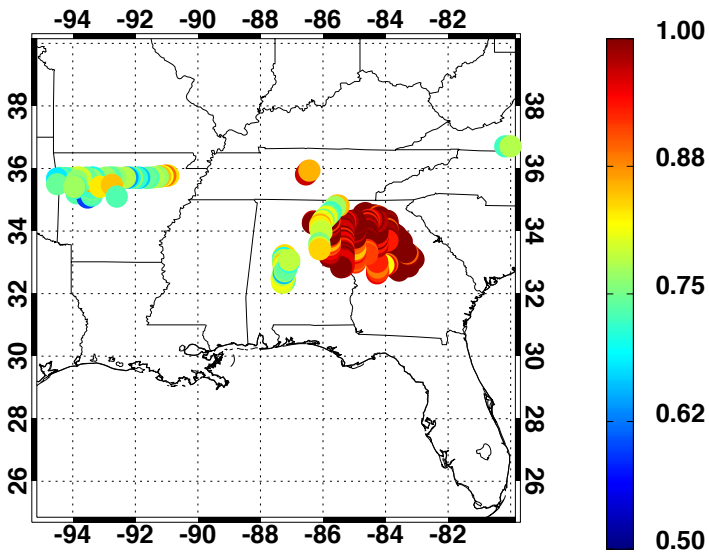
HCHO



Washenfelder et al. (Unpublished Data)

Possible Evidence for Acid Catalysed Aerosol Uptake?

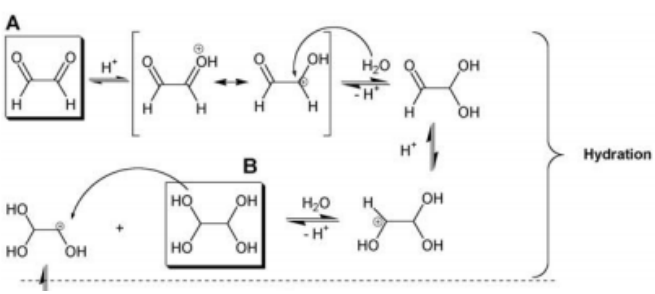
SENEX Neutralised Fraction



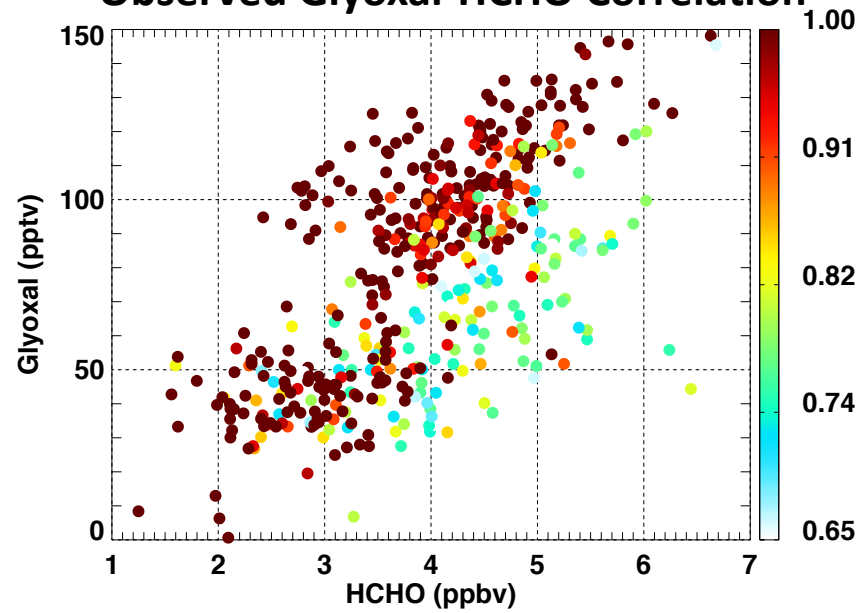
Neutralised Aerosol Fraction: $F = \frac{[NH_4^+]}{2[SO_4^{2-}]+[NO_3^-]}$

< 1.0 implies aerosol is acidic

Glyoxal:HCHO decreases with increasing aerosol acidity which may suggest acid mediated uptake e.g. Liggio et al. (2005)



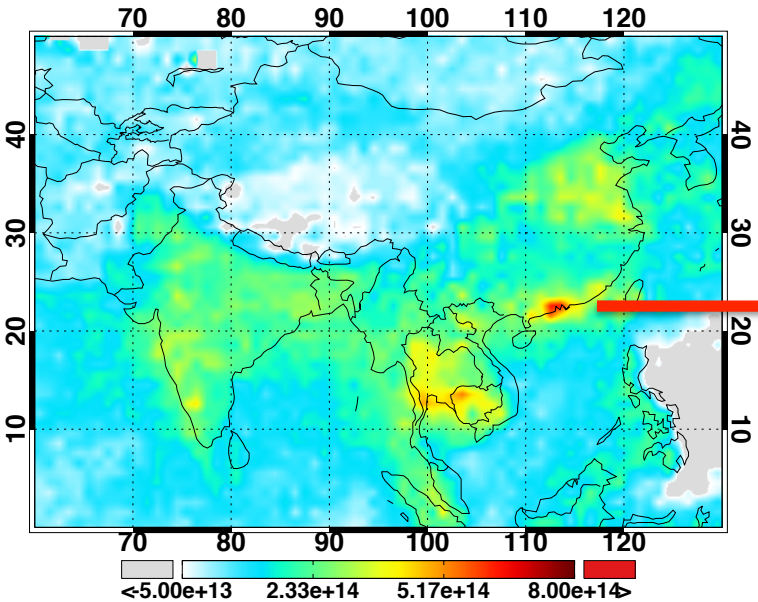
Observed Glyoxal-HCHO Correlation



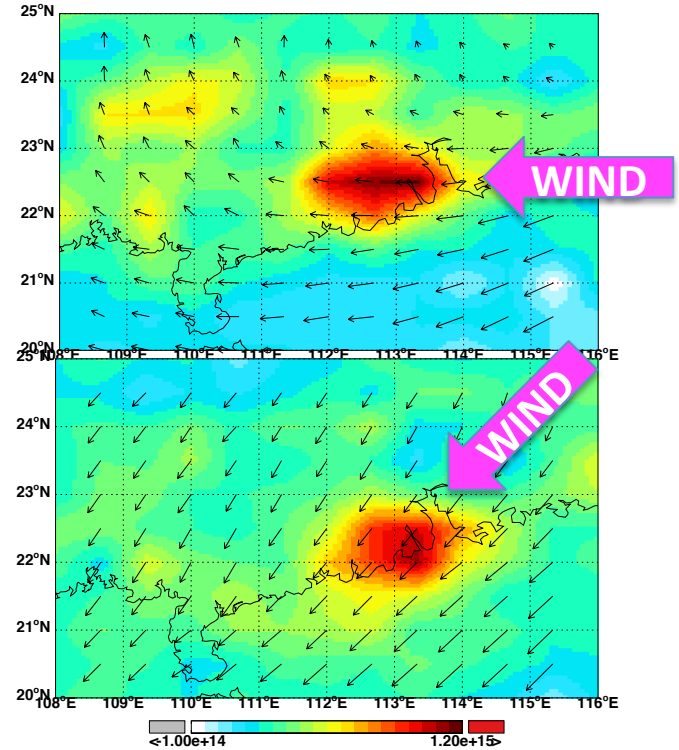
(Points color coded by neutralised fraction)

Elevated Glyoxal Levels Observed Over the Pearl River Delta

OMI Glyoxal VCD (DJF 2007)

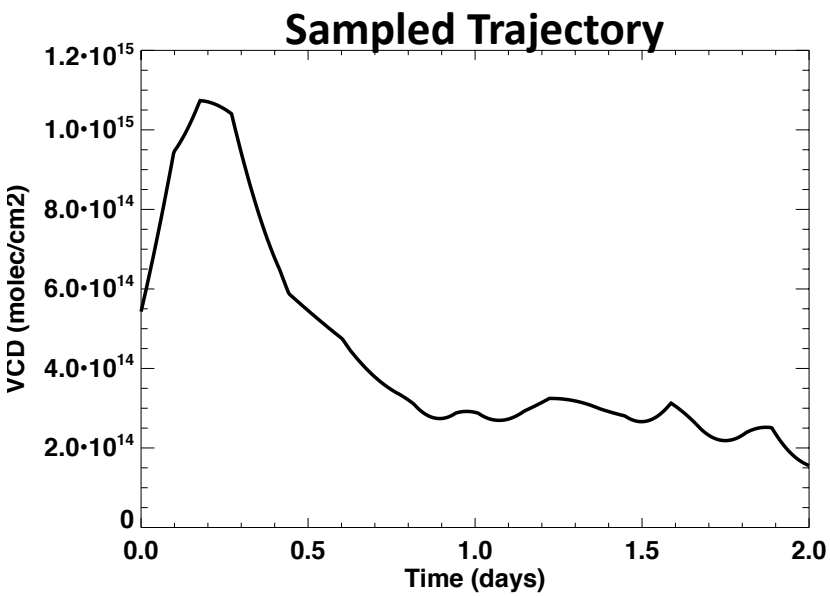


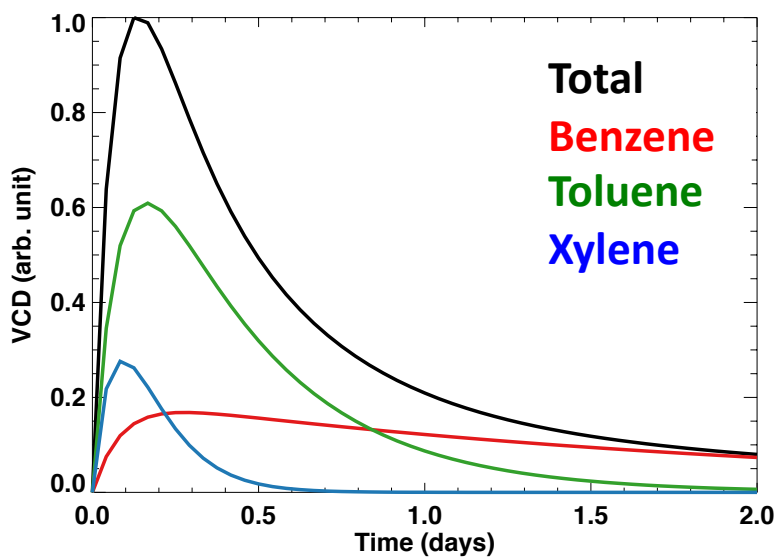
Grid by wind direction

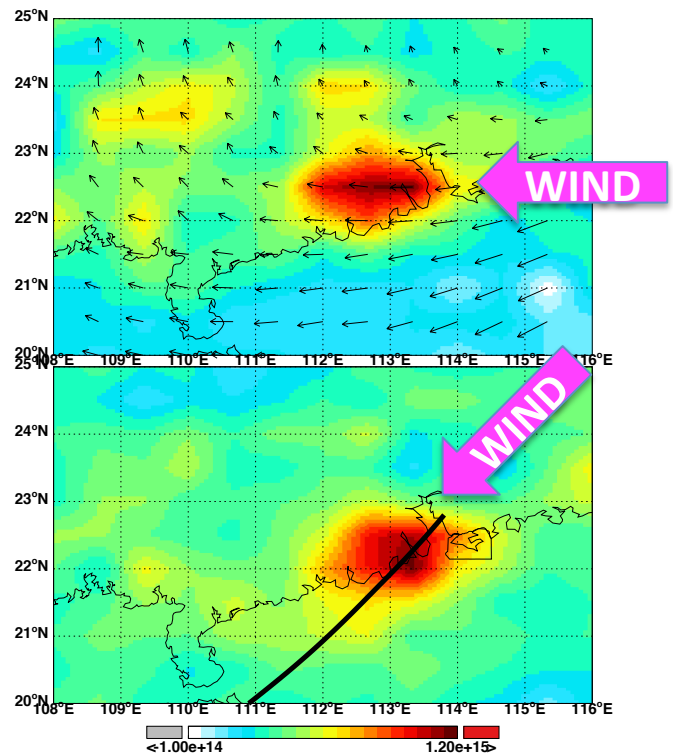


Aromatics dominate VOC sources over the PRD due to large emissions from transport, solvent use and paint fumes (Zheng et al. 2009)

Elevated Glyoxal Levels Observed Over the Pearl River Delta







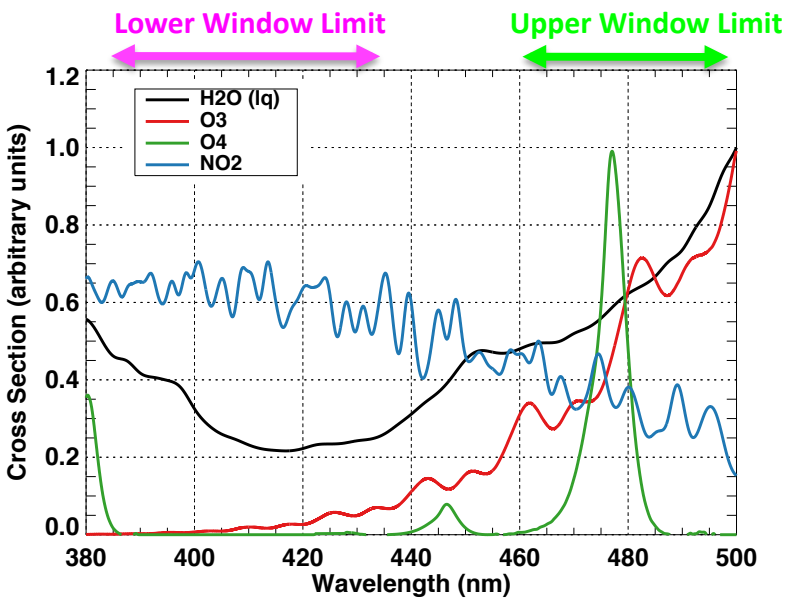
← Analytic model assuming first generation aromatic source

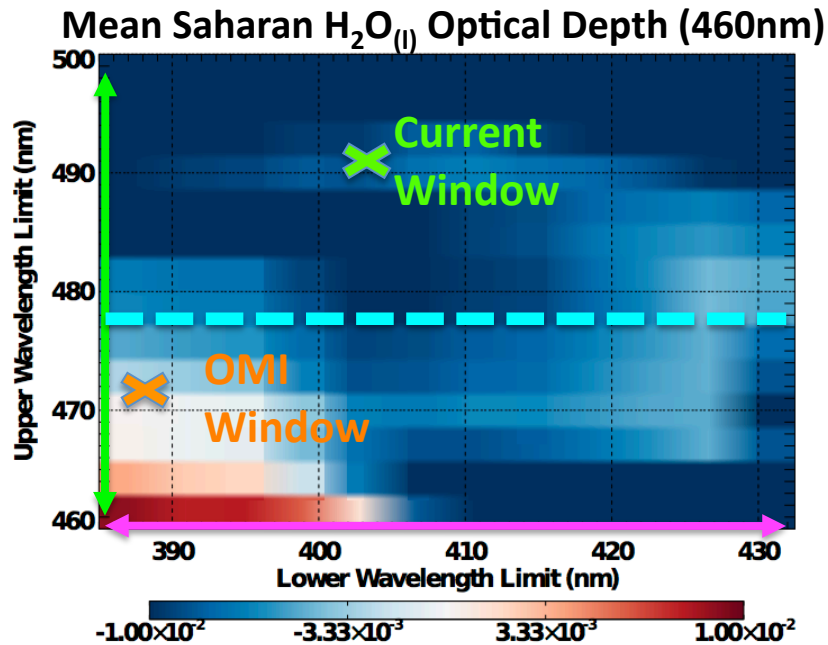
The observed plume decay is consistent with a combined Benzene/Toluene/Xylene source

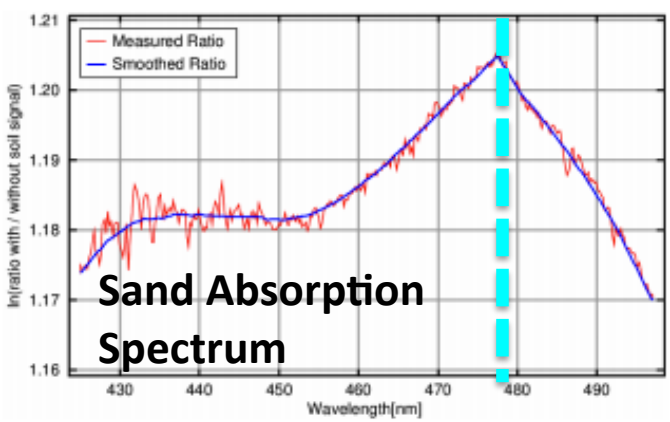
Thank you!

Supplementary

Liquid Water - Sensitivity to window position



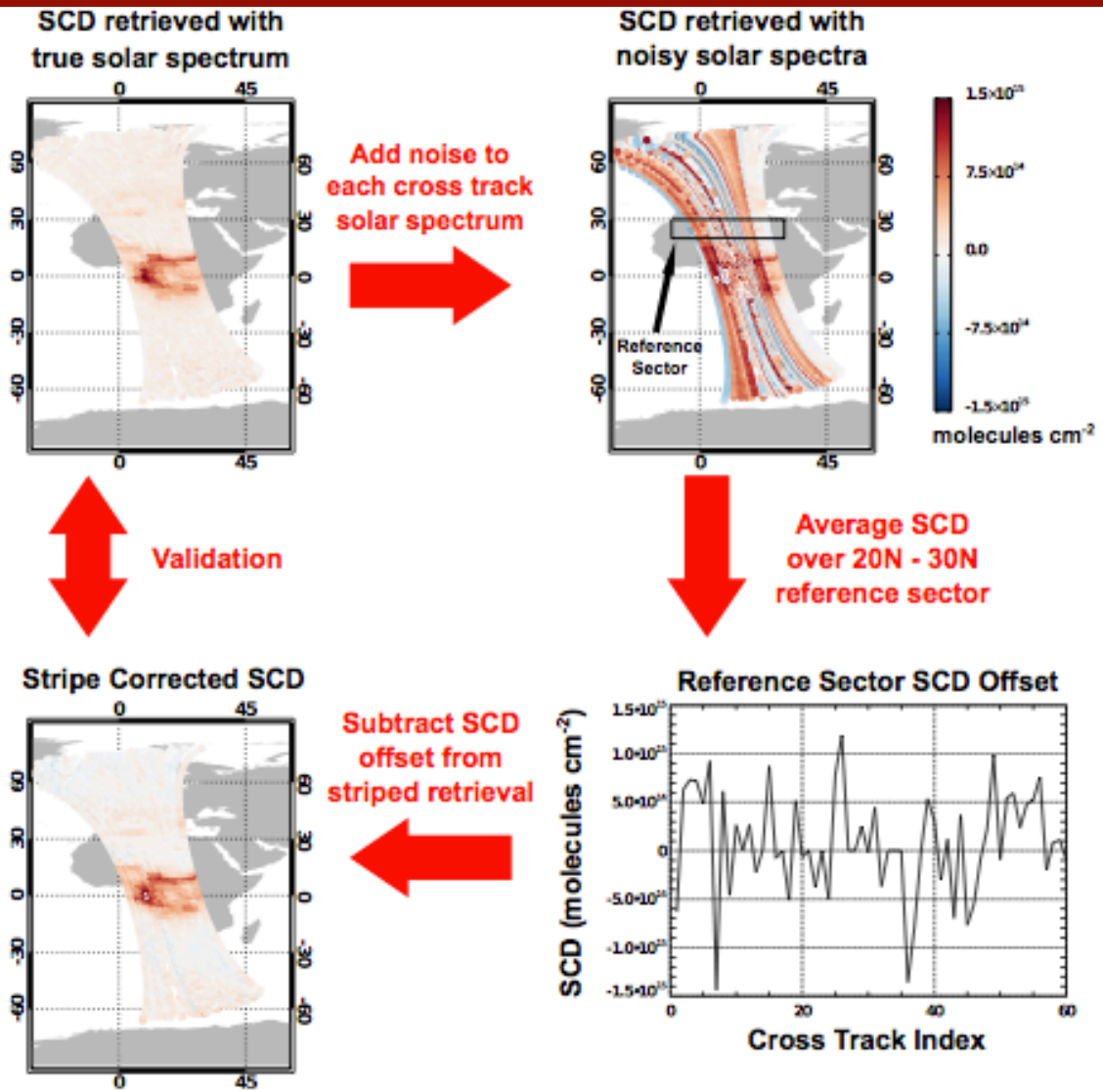




Richter et al. (2011)

Negative values over sandy surfaces may be due to sand spectrum
=> New retrieval window 405-470 nm

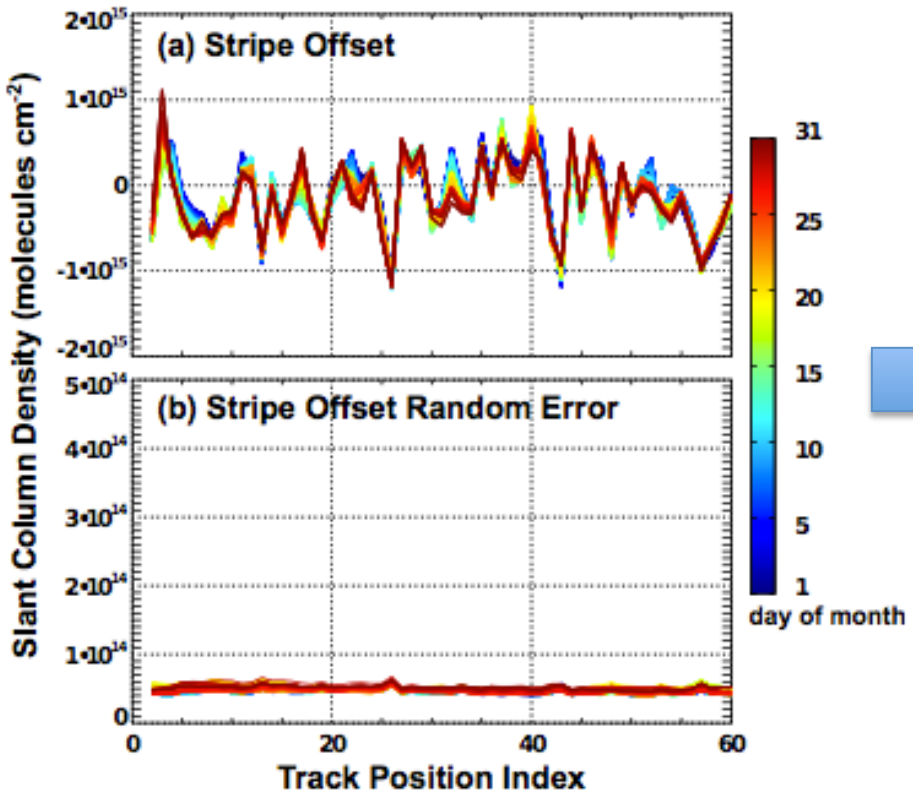
Impact of Solar Noise on Simulated Spectra

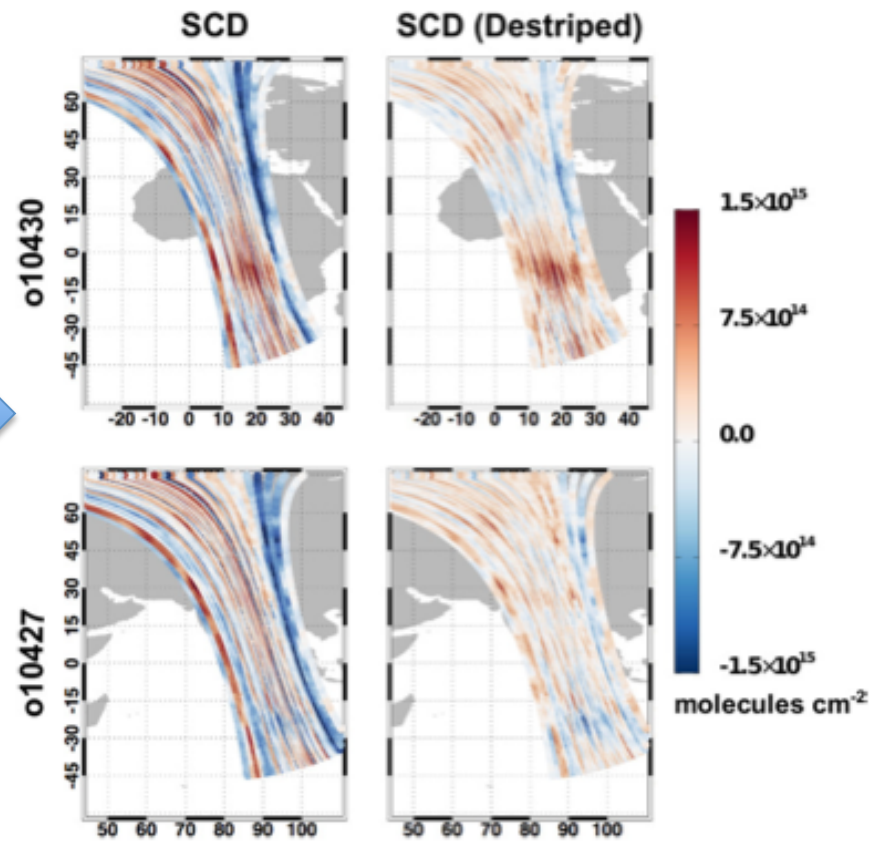


- Solar spectra with 3000 SNR introduce O(10^{15}) molecules cm^{-2} offsets
- Sahara is a good choice determining offsets (glyoxal < 1.5×10^{13} molecules cm^{-2})
- Offsets in simulation are constant with latitude => Offsets determined in Saharan reference sector can apply globally

Observed Stripe Offsets

Offsets determined through 5 day moving average





- Similar offset magnitude observed in real spectra
- Applied offset correction works for orbits outside Saharan reference sector