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Vertical Distribution of Pollutants in GEOS-Chem and from Satellite Observations

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Introduction

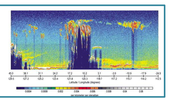
Instances of long range transport (LRT) of gas and aerosol pollutants have been well-documented in the past (Chin et al., 2007; Heald et al., 2003; Heald et al., 2006; Reidmiller et al., 2008; Yu et al., 2008), but previously limited observations of the vertical distribution during transport restricted understanding of the progression and downwind impact of these pollutants. In this study, we investigate aerosol extinction observations from the CALIPSO instrument, aerosol optical depths from MODIS, and measurements of carbon monoxide from the Tropospheric Emission Spectrometer (TES) with a global chemical transport model (GEOS-Chem) to examine the vertical distribution of pollutants in the Northern Hemisphere on seasonal timescales.

Importance of Vertical Structure

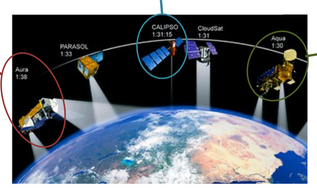
The downwind surface impact is dependent on how efficiently pollutants are transported. In general, pollutants have longer lifetimes at higher altitudes due to colder temperatures and fewer removal processes. Additionally, higher wind speeds in the free troposphere lead to faster transport. **Therefore,** pollutants that are lofted into the free troposphere are generally more efficiently transported.

Satellite Observations

CALIOP
- Aerosol Extinction (532nm)
- ~100 m footprint, 30 m vertical resolution
- Full global coverage ~16 days
- Level 2 Version 3.01 5-km Aerosol & Cloud Profiles

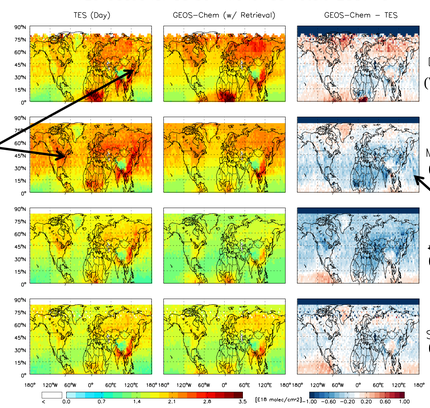


TES
- "Profiles" of CO
- Uses Optimal estimation method: $\ln\hat{x} = \ln x_a + A(\ln x - \ln x_a) + \varepsilon$
- Limited vertical resolution
- Level 2, Version 004 Day



MODIS
- AOD
- Near global coverage daily
- Combine land and ocean retrieval
- Filter along Zhang and Reid (2006)
- Collection 5, Level 3 Daily

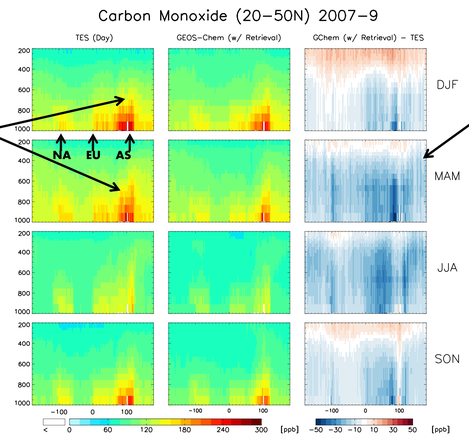
CO from Tropospheric Emission Spectrometer (TES)



Some export and long range transport is evident

GEOS-Chem simulates (most) CO source regions and seasonality

GEOS-Chem biased low (Shindell, 2007; Kopacz, 2010)



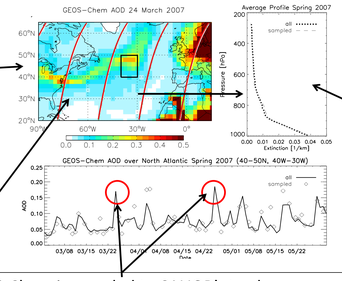
Vertical distributions show lofting over sources and strong seasonality in background concentrations.

GEOS-Chem biased low also in the vertical.

**Coarse vertical resolution of TES limits our vertical comparisons.

Note on Sampling

1. GEOS-Chem provides global coverage and can therefore simulate transported plumes.



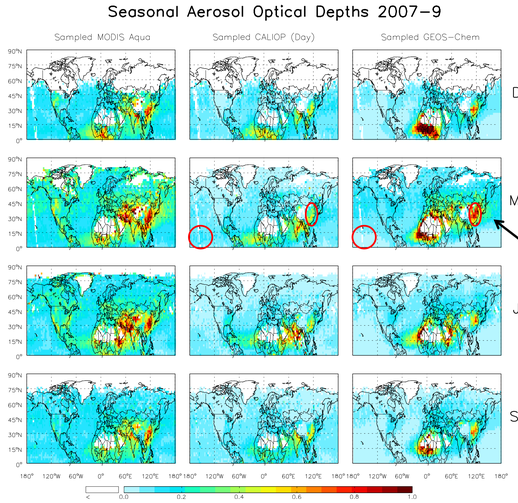
2. CALIOP has a narrow (~100m) footprint and wide distance between scans, which can result in "missed" plumes.

3. When GEOS-Chem is sampled to CALIOP's track, some transported plumes can be sampled out, and daily AOD for a region can be over- or under-estimated.

4. However, when we compare seasonal profile, there is little difference between the sampled and unsampled model extinction profiles.

5. We, therefore, make the rest of our comparisons on ****multi-year seasonal timescales.****

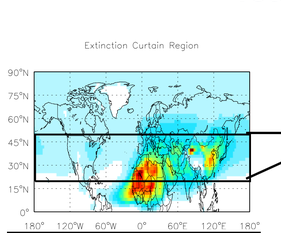
Comparing AOD



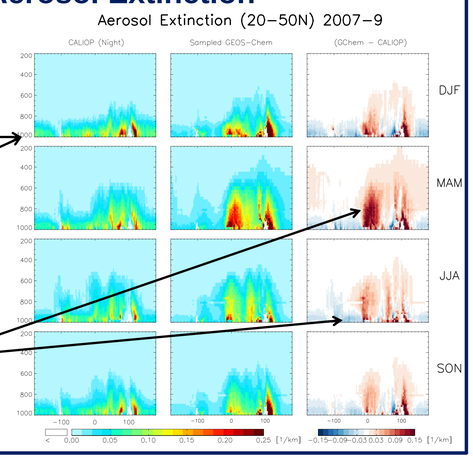
MODIS is generally biased high compared to GEOS-Chem and CALIOP.

GEOS-Chem is biased high near sources and low in remote regions compared to CALIOP.

Total Aerosol Extinction



Most LRT occurs between 20-50ºN.



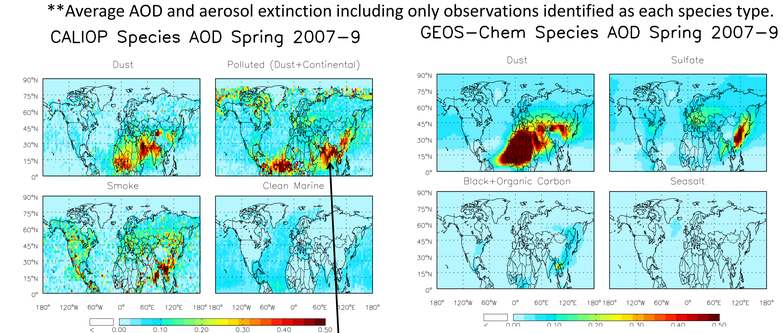
GEOS-Chem is biased high over sources and aloft, but biased low in regions of low level outflow.

Conclusions and Future Work

- GEOS-Chem is biased high near sources and low in **low-level** outflow regions. This appears to be due to both discrepancies in pollution outflow and background aerosol. The coarse vertical resolution of TES limits the ability to draw similar conclusions for gas phase pollutants.
- Future work will focus on continuing to analyze differences in aerosol species type and fractions in the boundary layer. We will also look at specific regions and mechanisms for export and lofting.

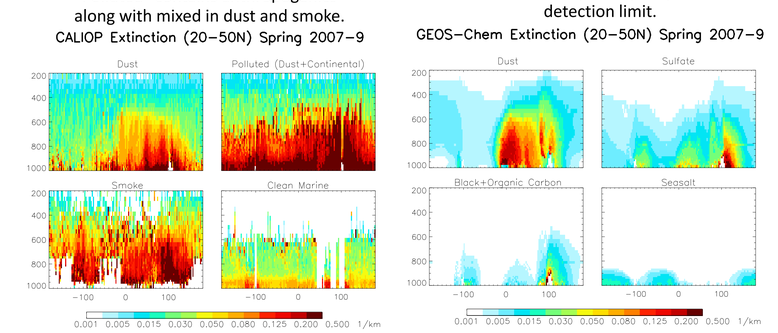
Aerosol Species

**Average AOD and aerosol extinction including only observations identified as each species type.



"Polluted" contains anthropogenics along with mixed in dust and smoke.

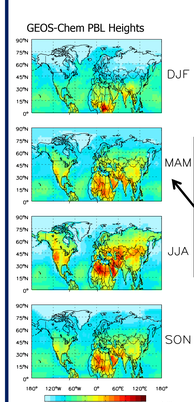
Underestimations in species partly due to CALIOP detection limit.



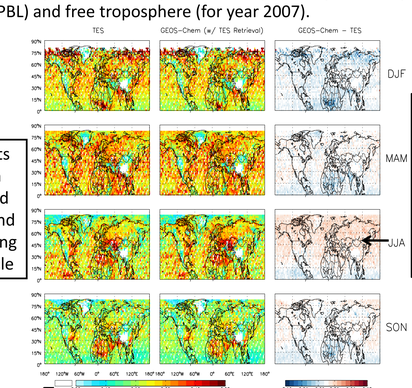
GEOS-Chem low extinction in outflow appears to be due to both less background aerosol and less pollution outflow.

Fraction in the Boundary Layer

Allows us to differentiate pollutants transported in the planetary boundary layer (PBL) and free troposphere (for year 2007).

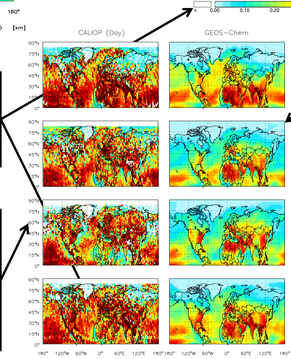


PBL heights vary with season and location, and have a strong diurnal cycle



Interaction of several factors:
- Emissions,
- PBL height
- Lofting mechanisms
- Lifetime of CO

Higher fraction of AOD in the PBL compared to CO.



Initial comparisons between CALIOP and GEOS-Chem are poor

However, CALIOP detection limit is ~2-4x10^{-4} km^{-1} sr^{-1} (Yu et al., 2010).

Applying this limit (Ext Values<0.0028/km→0 /km) to GEOS-Chem improves comparisons in remote regions, but the model still underestimates the fraction in outflow regions.