Tropospheric Aerosols Over the Western North Atlantic Ocean During May-June ACTIVATE 2021 and 2022: Transport, Distribution, and Sources

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ACTIVATE Science Team Meeting
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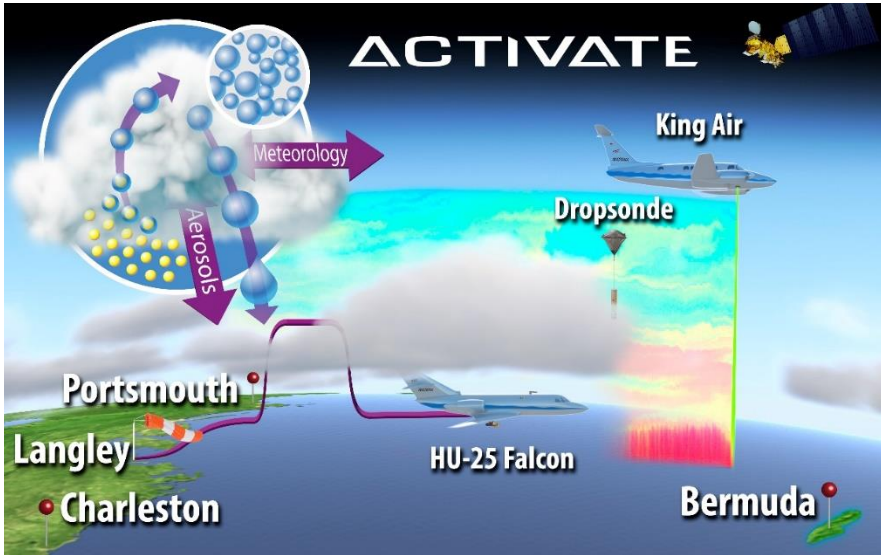
Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment



- PI: Armin Sorooshian (U. Arizona)
- Project Scientist: John Hair (NASA LaRC)
- NASA Earth Venture Sub-orbital (EVS-3) Mission
- Partnering Institutions: U. Arizona, NASA LaRC, NASA GISS, NASA WFF, NASA ARC, NCAR, RSES, NIA, PNNL, BNL, BAERI, U. Miami, USC, UW, NOAA CSL, College of William & Mary, Yulista, Bermuda Institute of Ocean Sciences, DLR (Germany)

- Platforms: **HU-25 Falcon** + **UC-12 King Air**
- 162 joint flights (~580 hrs per plane) over the western North Atlantic
- Based out of **NASA LaRC** (Hampton, Virginia) **and Bermuda**
- Measurements: In situ and remote sensing measurements of aerosol and cloud distributions and properties, atmospheric state

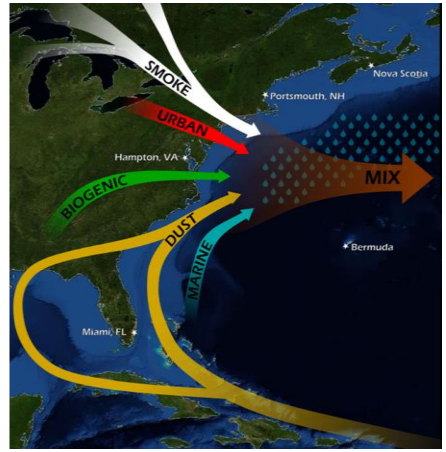
Mission Goal: **"**Robustly characterize aerosol-cloud-meteorology interactions using extensive, systematic, and simultaneous in situ and remote sensing airborne measurements with two aircraft and a hierarchy of models" (https://activate.larc.nasa.gov/science/).

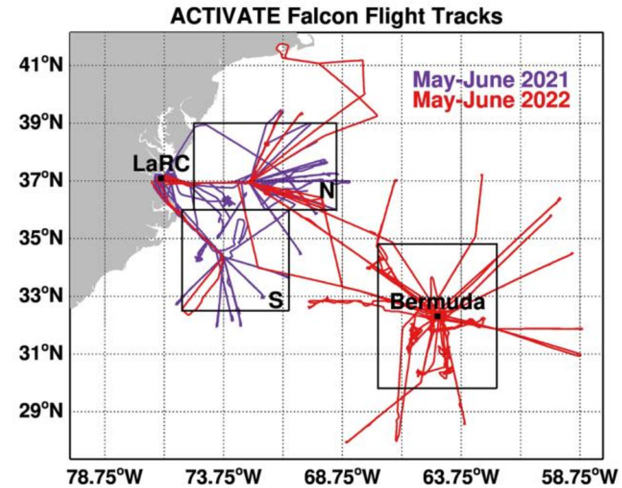
Characterization of aerosol-cloud-meteorology interactions requires understanding of aerosol sources, transport pathways, composition, and distribution.

Modeling tropospheric aerosols over the Western North Atlantic Ocean (WNAO)

ACTIVATE #1: Feb. 14 – Mar. 12, 2020
#2: Aug. 13 – Sep.30, 2020
#3: Jan. 27 – Apr. 2, 2021
#4: May 13 – Jun. 30, 2021
#5: Nov. 30 – Mar. 29, 2022
#6: May 3 – Jun. 18, 2022

Objective: to improve understanding of aerosol sources, transport pathways, composition, and distribution over the WNAO (25-50°N, 60-85°W).





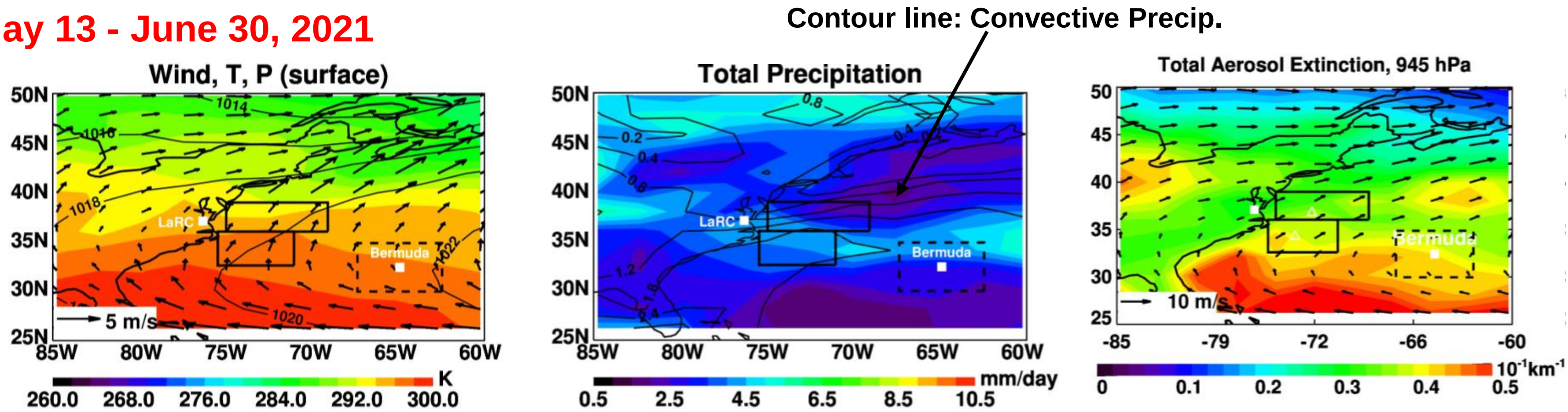
Major sources of tropospheric aerosols and their precursors over the WNAO: Urban, smoke, biogenic emissions, dust, and marine emissions (Sorooshian et al., JGR 2020).

Model: GEOS-Chem v11-01 (MERRA-2, 2°×2.5°)

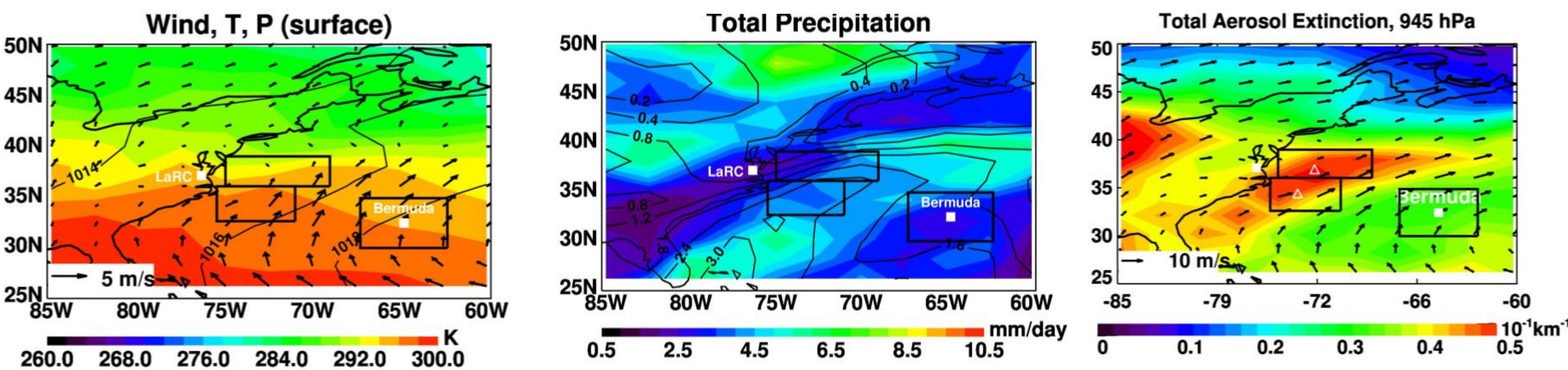
- Sulfate-nitrate-ammonium (SNA) aerosol thermodynamics (**ISORROPIA**; Fountoukis and Nenes, ACP 2007) coupled to gas-phase chemistry
- Mineral dust in four size bins (Fairlie et al., AE 2007)
- **Sea salt in two size bins** (Jaegle et al., ACP 2011)
- Elemental and organic carbon aerosols (Q. Wang et al., JGR 2014; Pye et al., ACP 2010)
- EDGAR v4.2 FF emis. (NEI 2011 scaled to YR 2018)
- **QFED biomass burning emis. v2.6r1** (Darmenov & da Silva, GMAO 2015)
- Biogenic emissions (MEGAN)
- Marine DMS (Lana et al., GBC 2011)
- **Aerosol wet deposition scheme**
  1) MERRA-2 cloud water content (CWC) (Luo et al., 2019);
  2) Fixed CWC value (Liu et al., 2001).

Meteorological settings & aerosol distribution

May 13 - June 30, 2021

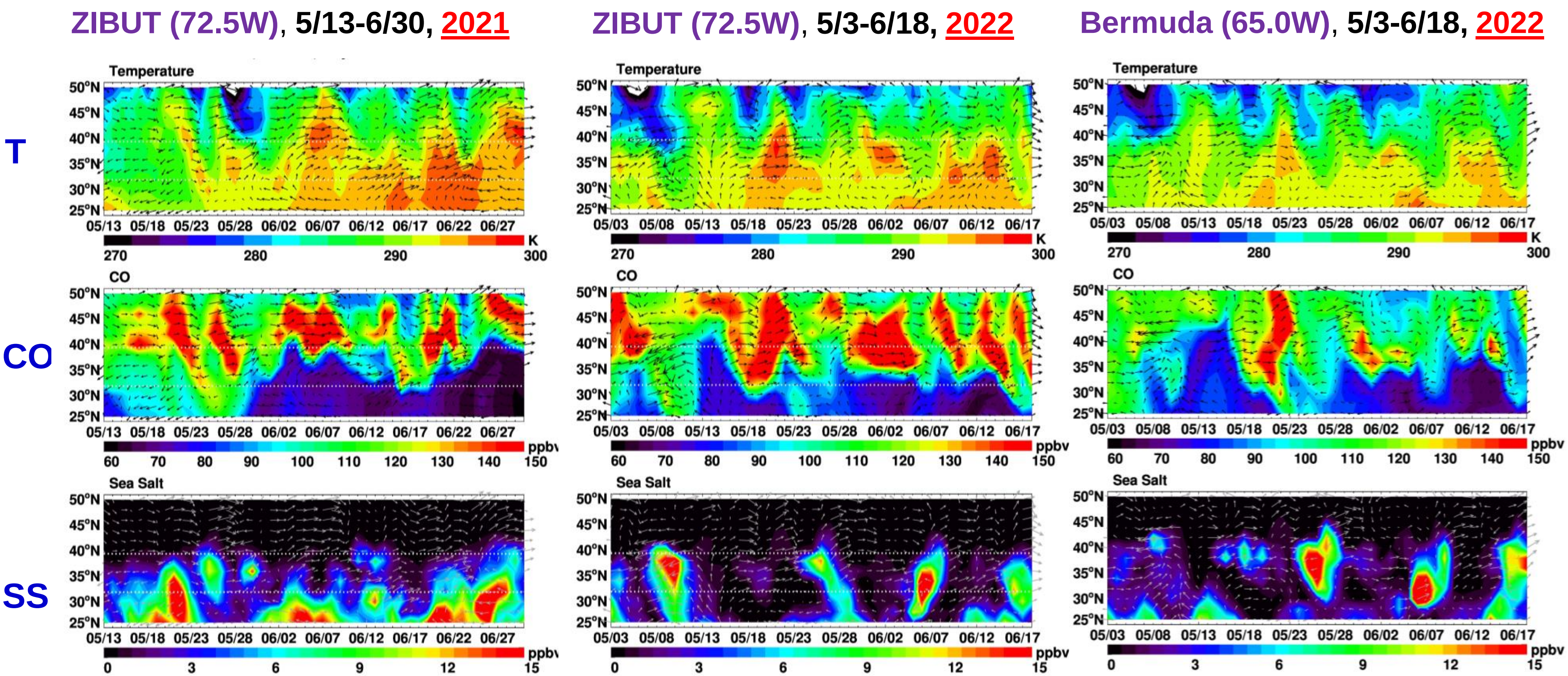


May 3 - June 18, 2022



- Midlatitude cyclones driving NA outflow to the WNAO (westerly winds)
- Bermuda High shifts westward towards summer (SW winds over the ACTIVATE near-coast domain), with stronger NE-ward transport in 2022
- Significant impact of precipitation on the distribution of BL aerosol extinction
- Stronger convection within the southern box area (near-coast)

Spatiotemporal evolution of BL outflow to the WNAO

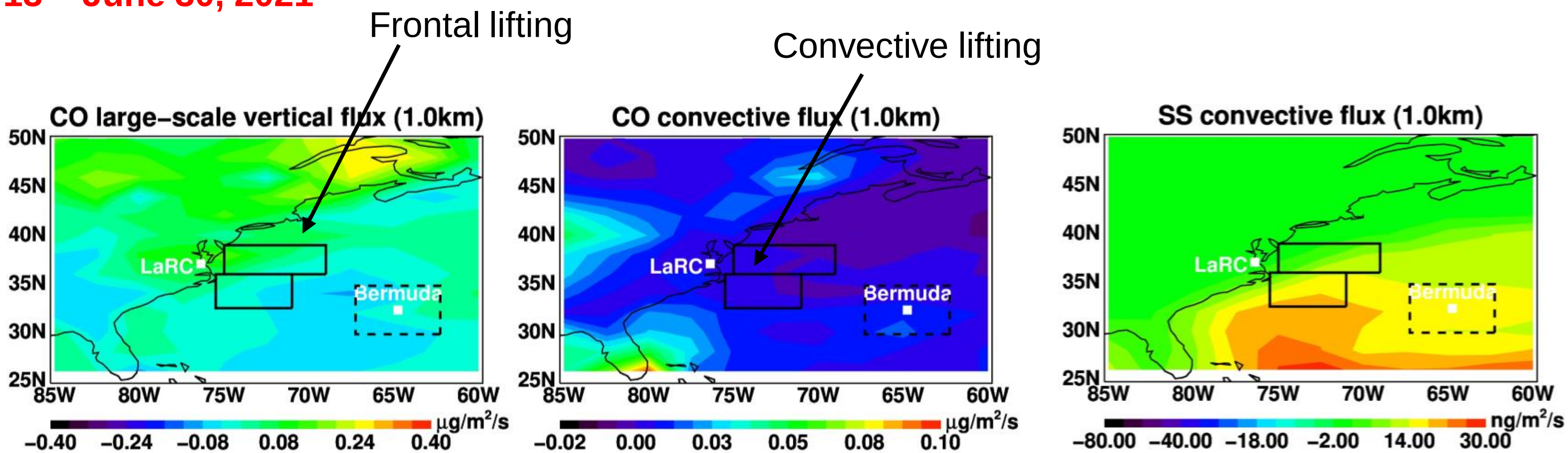


Hovmöller diagram of GEOS-Chem daily mean **950-hPa** air temperature, winds, CO and sea salt mixing ratios along 72.5W (ZIBUT waypoint) and 65.0W (Bermuda).

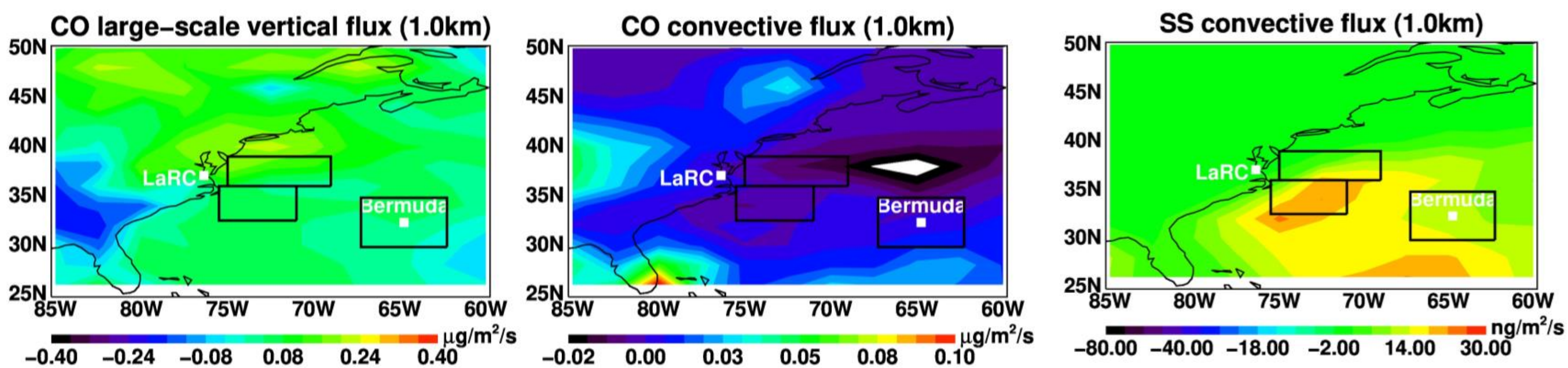
- WNAO is strongly influenced by BL outflow of North American pollution behind cold fronts, but the influence weakens starting early June.

Vertical transport of carbon monoxide (CO) and sea salt (SS)

May 13 – June 30, 2021



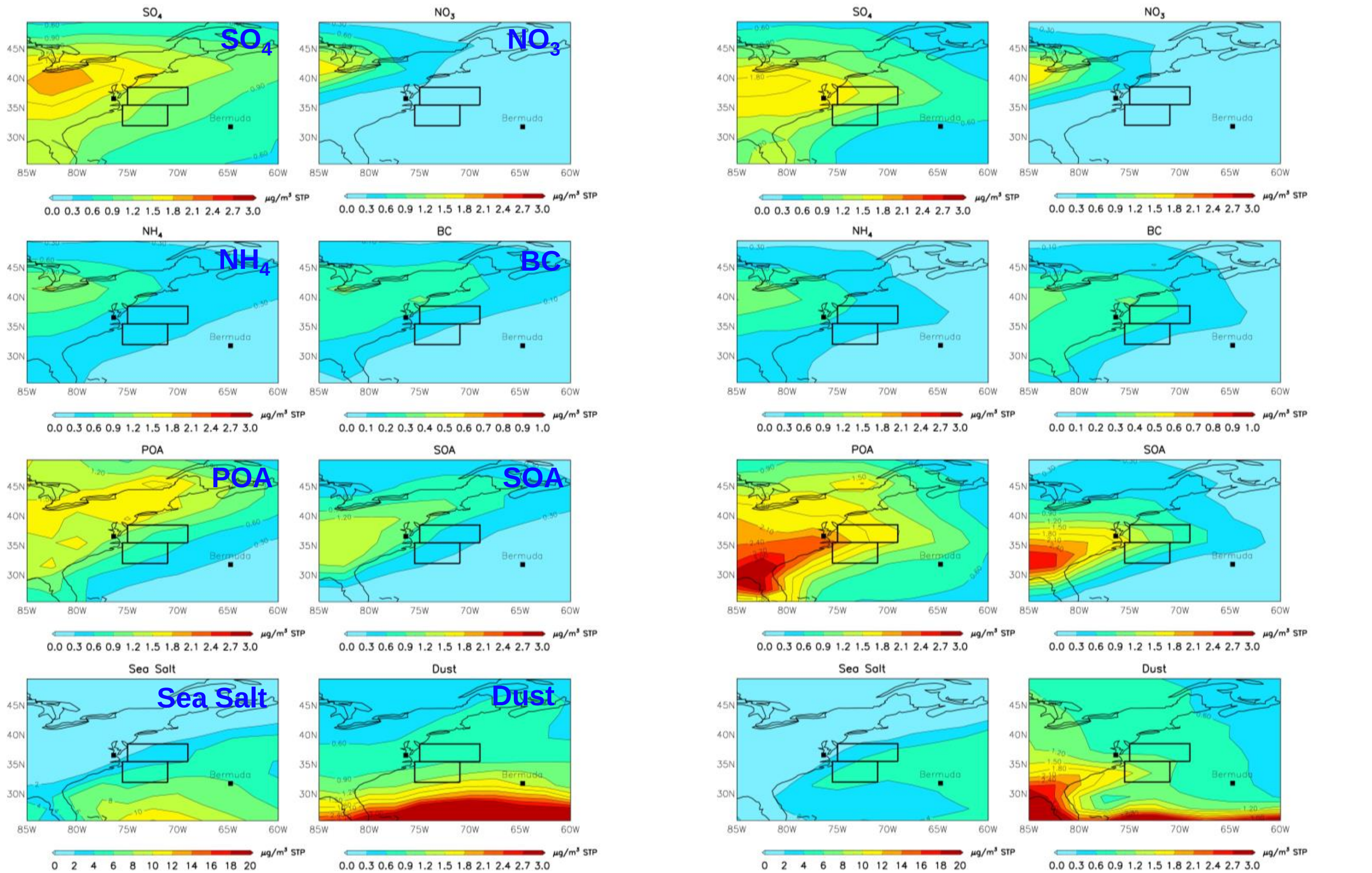
May 3 – June 18, 2022



- CO (pollution tracer): Large-scale vertical advection is the major upward transport pathway; convective lifting becomes important towards summer
- Convection (vs. PBL turbulent mixing in winter) is the dominant process responsible for upward transport of sea salt within and ventilation out of the PBL during May-June

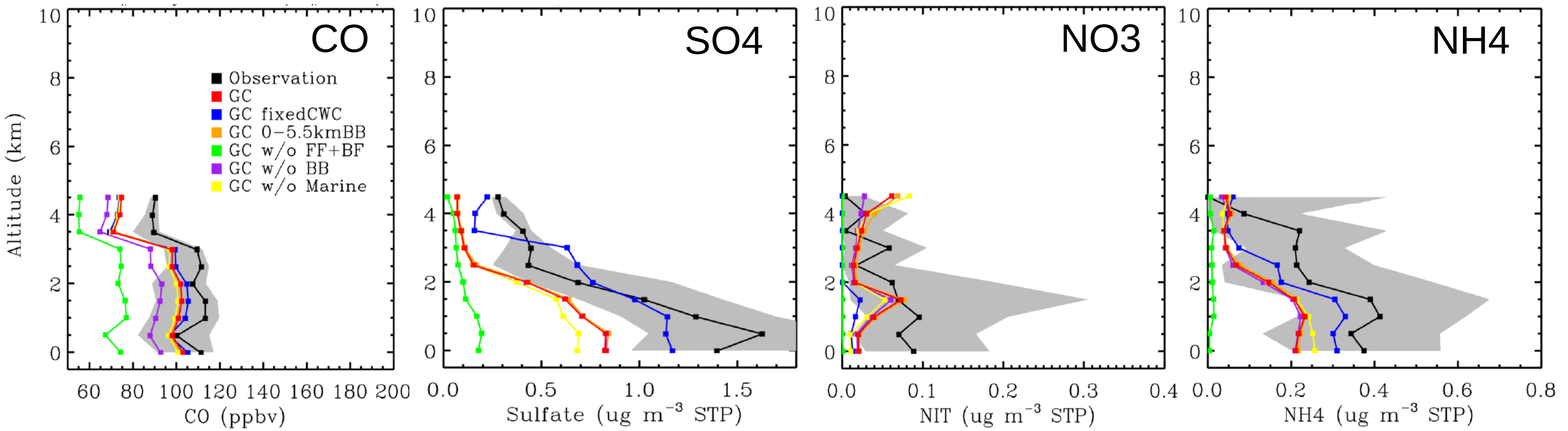
Model simulated distribution of aerosol concentrations at 0.7-km altitude

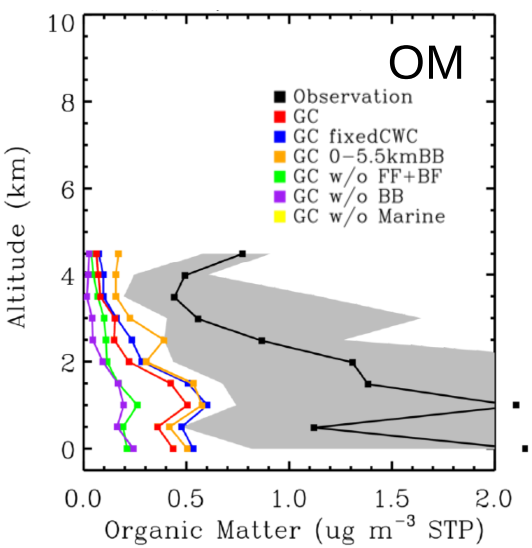
May 13 – June 30, 2021 | **May 3 – June 18, 2022**



- Organic aerosols: Much higher POA & SOA in 2022 due to stronger biomass burning influence
- Sea salt: higher in 2021, especially in the proximity of Bermuda
- Dust: African dust transported to Florida and the Gulf of Mexico followed by N- or NE- ward transport to the eastern U.S. coast and WNAO in 2022.

Evaluation with aircraft (Falcon) in situ measurements – May-June ACTIVATE 2021

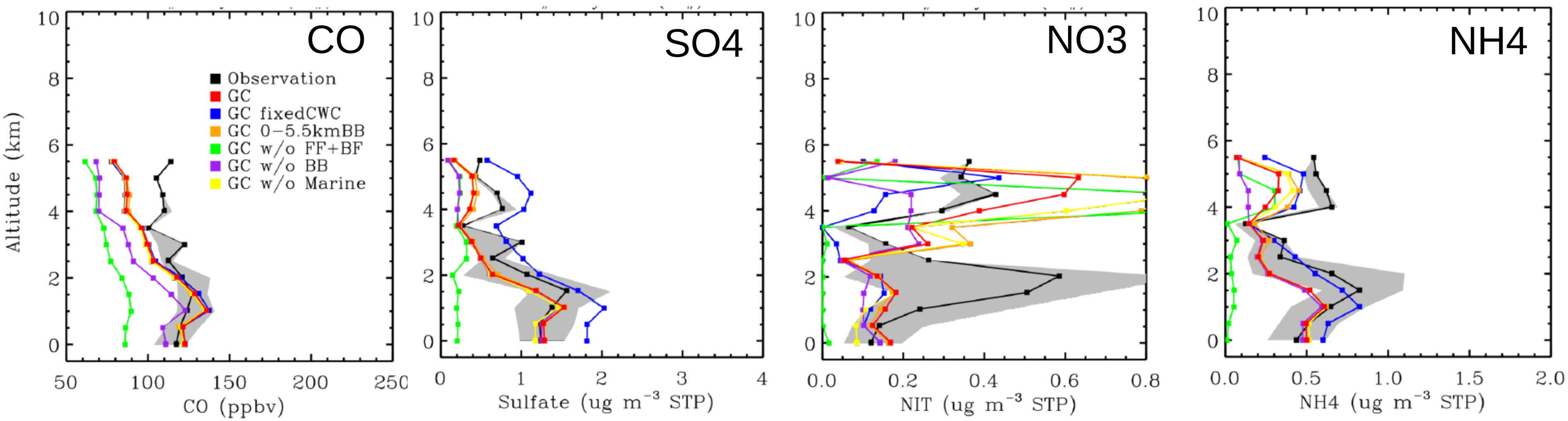


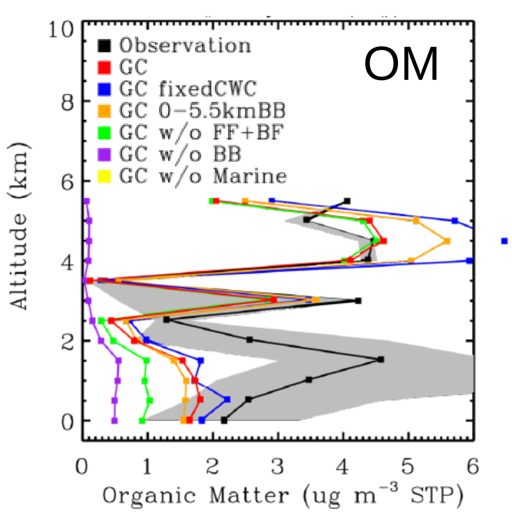


May-June 2021:

- Model generally underestimates BL aerosol observations
- A distinct layer of enhancement at ~1-2 km in both the model and measurements.
- Strong aerosol (vs CO) vertical gradients suggest the role of precipitation scavenging
- Anthropogenic emission is the dominant source for CO, SO_4, NO_3, and NH_4 aerosols in the near-coast study domain
- Biomass burning emissions contribute to >30% of organic aerosols in the lower troposphere

Evaluation with aircraft (Falcon) in situ measurements – May ACTIVATE 2022

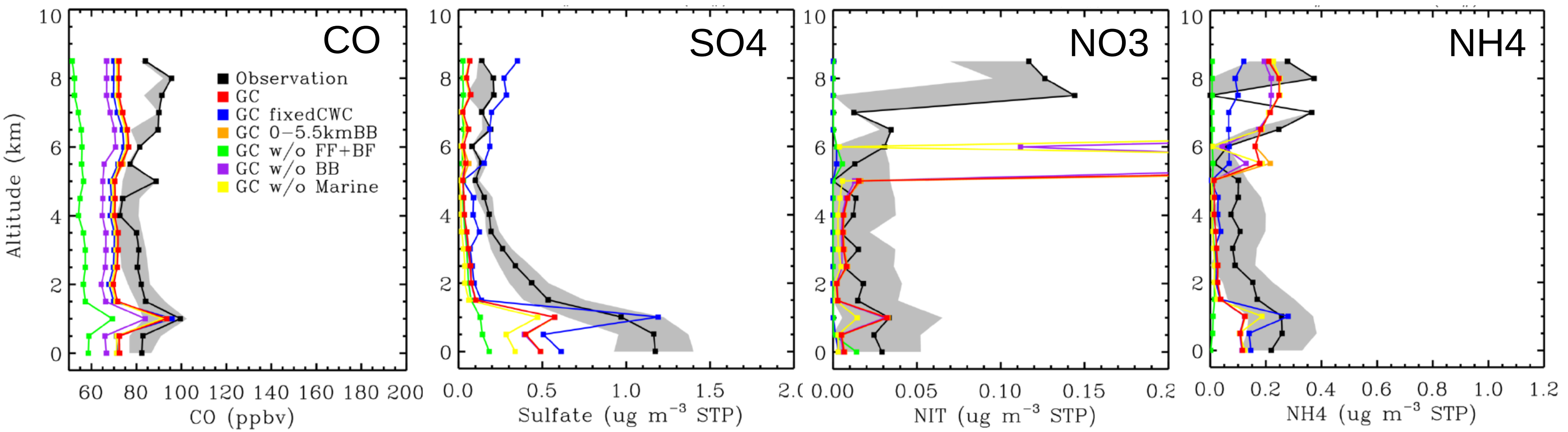


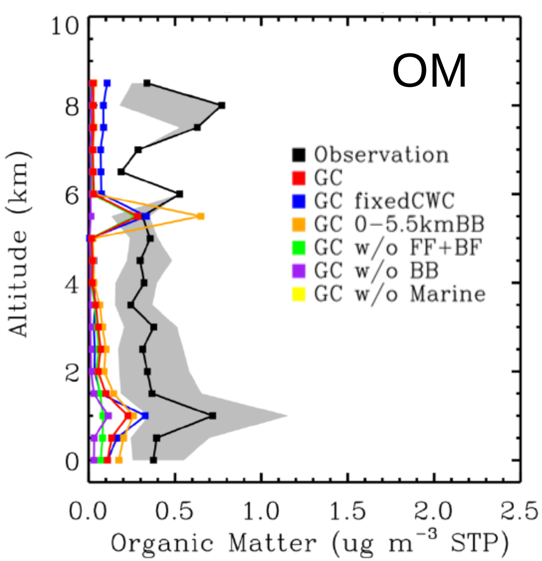


May 2022:

- Model reasonably simulates BL CO, SO4, and NH4 observations, but underestimates BL NO3 and OM.
- A distinct layer of enhancement at ~1-2 km in both the model and the measurements.
- Anthropogenic emission is the dominant source for BL CO, SO_4, NO_3, and NH_4 aerosols in the near-coast study domain
- Biomass burning emissions dominate organic aerosols in the model

Evaluation with aircraft (Falcon) in situ measurements – June ACTIVATE 2022

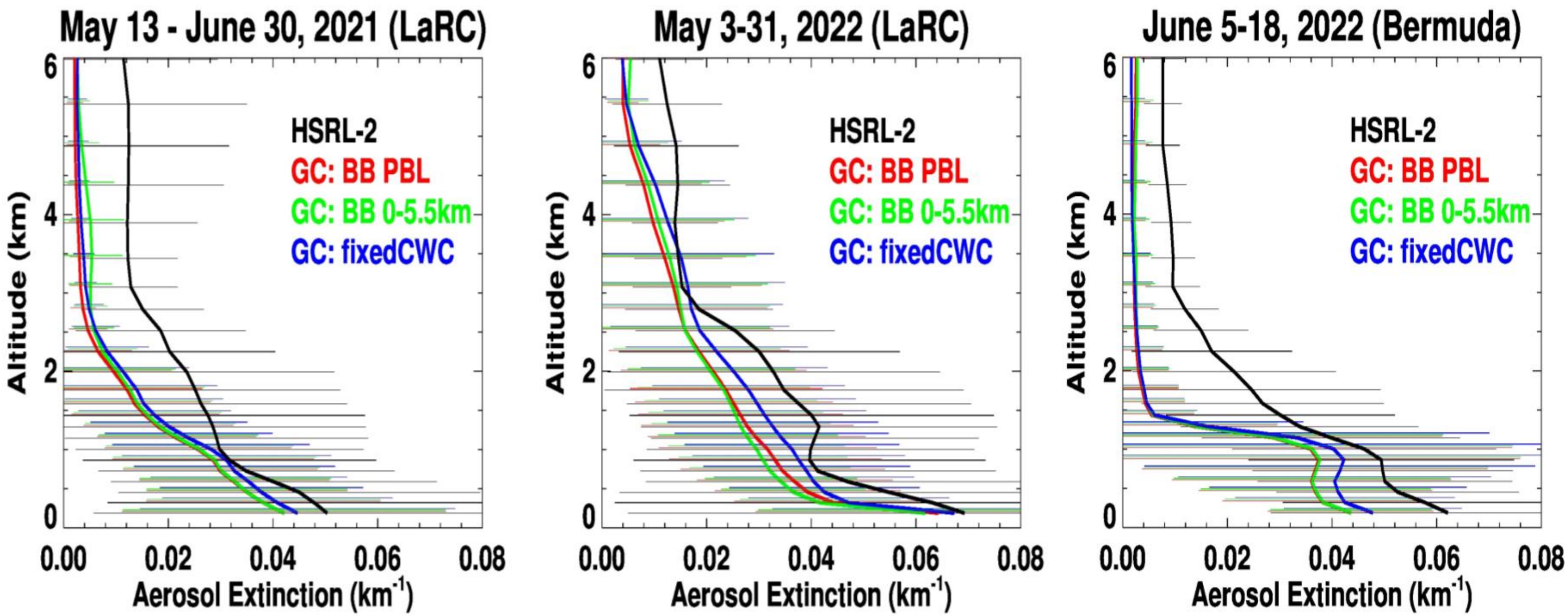




Bermuda June 2022:

- Flight 6/17/2022 excluded (volcanic influence on SO4 in the model)
- CO and aerosol layer enhancements at ~1.0 km (above BL top).
- Model generally underestimates Falcon AMS aerosol concentrations

Evaluation with aircraft (King Air) HSRL-2 lidar measurements

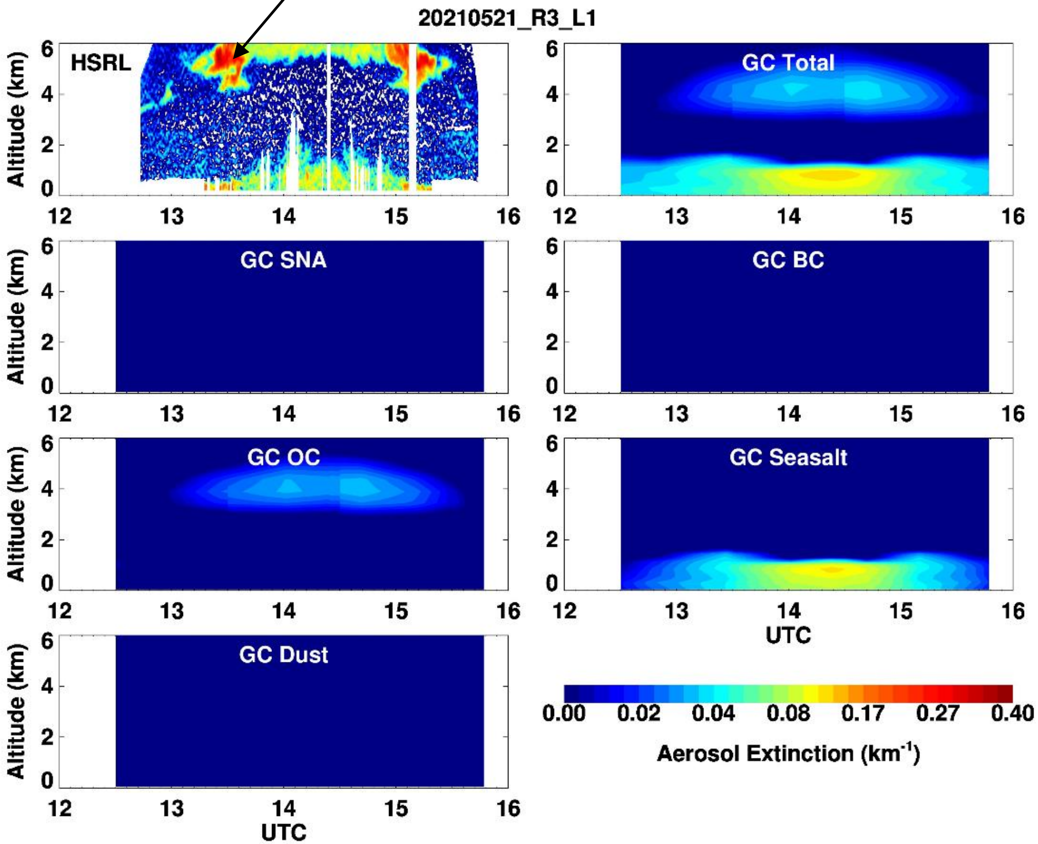


Comparison of the model aerosol extinction (550nm) vertical distribution with aircraft HSRL-2 lidar measurements (532nm) averaged over all flights for each campaign.

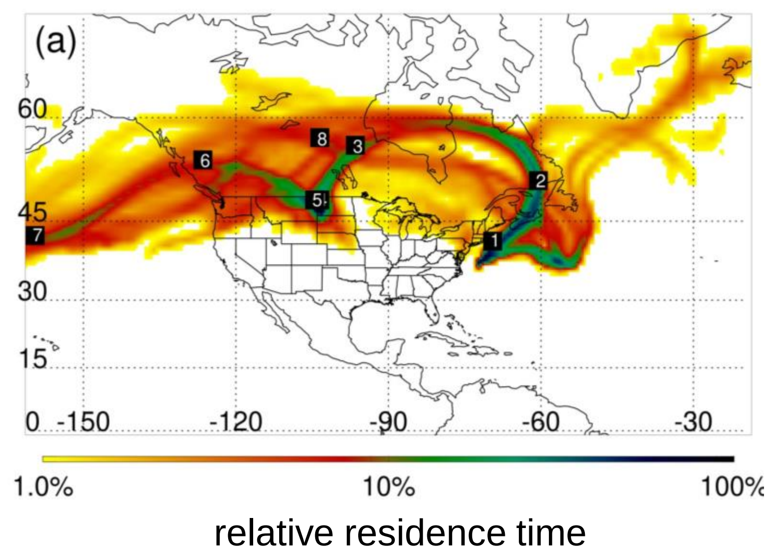
- LaRC: Model reasonably simulates NA boundary-layer outflow of aerosols
- LaRC: Model slightly underestimates HSRL-2 aerosol extinctions in the free trop during 5/13–6/30, 2021, with better model-obser agreements during 5/3-31, 2022
- Bermuda: Model underestimates HSRL-2 aerosol extinctions over the Bermuda area

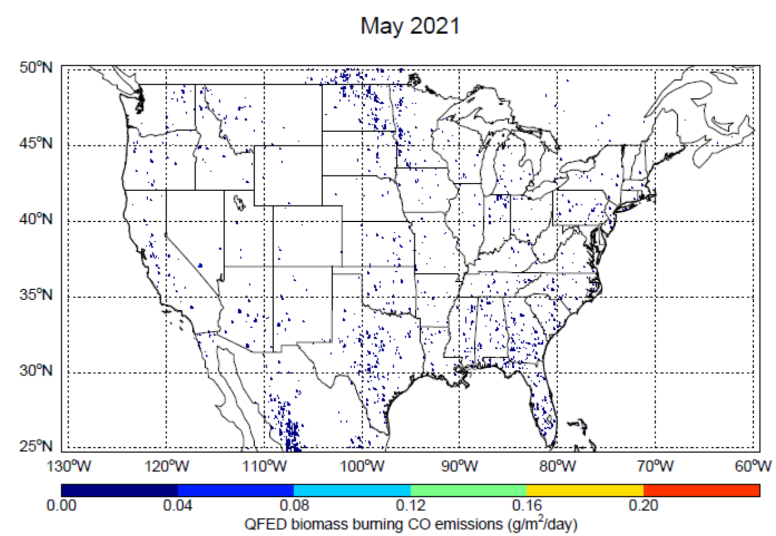
0-5.5 km fire emission injection height improves the simulations of HSRL-2 aerosol extinction curtains on May 21, 2021

Canadian fire smoke



FLEXPART Trajectory 2021-05-21, 13:24Z



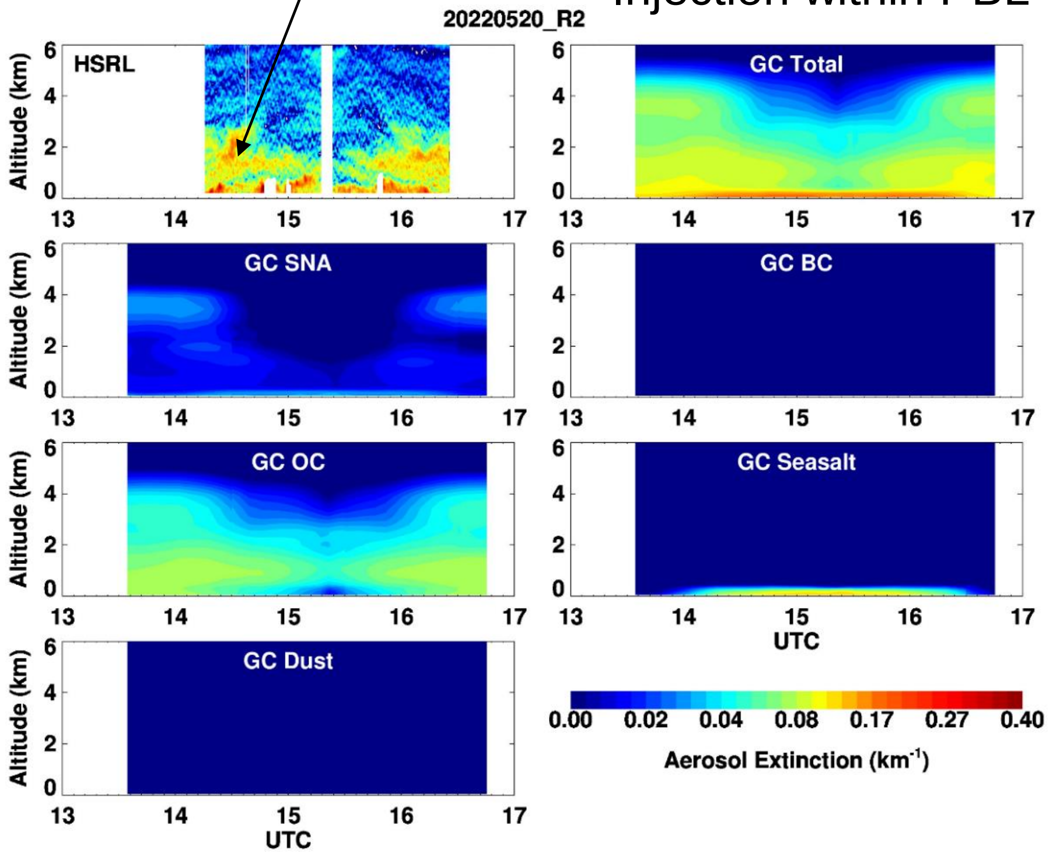


Time-height cross section of aerosol extinctions observed by aircraft HSRL-2 lidar (532 nm) compared to model aerosol extinctions (550 nm) during the morning flight on May 21, 2021. Curtain plots of model speciated aerosol extinctions along the flight track are also shown. SNA = sulfate + nitrate + ammonium

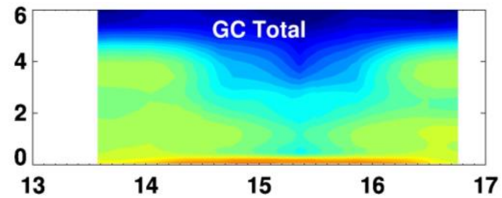
Model simulation with fire emissions injected within PBL was able to reasonably reproduce HSRL-2 aerosol extinction curtains on May 20, 2022

South U.S. fire smoke

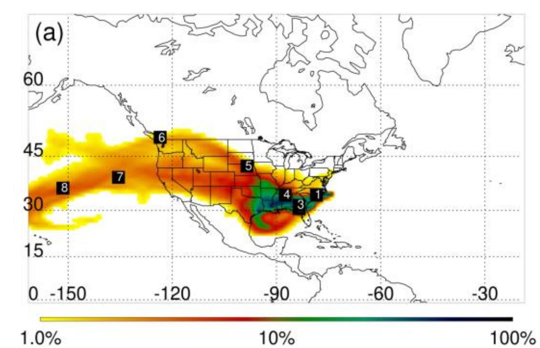
Injection within PBL



0-5.5km injection height



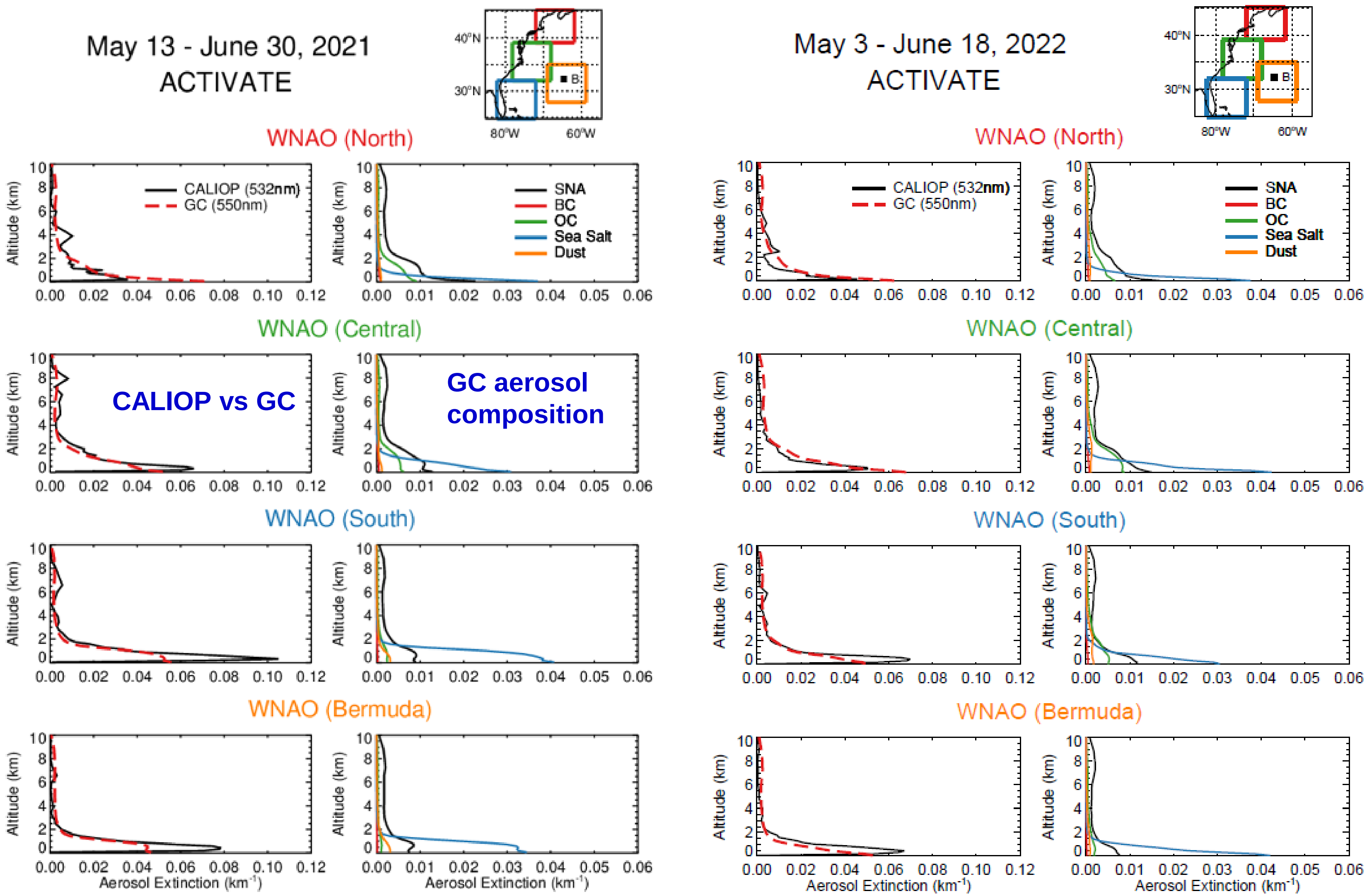
FLEXPART Trajectory 2022-05-20, 14:30Z



relative residence time

Time-height cross section of aerosol extinctions observed by King Air HSRL-2 lidar (532 nm) compared to model results (550 nm) during the flight on May 20, 2022. Curtain plots of model speciated aerosol extinctions along the flight track are also shown. SNA = sulfate + nitrate + ammonium

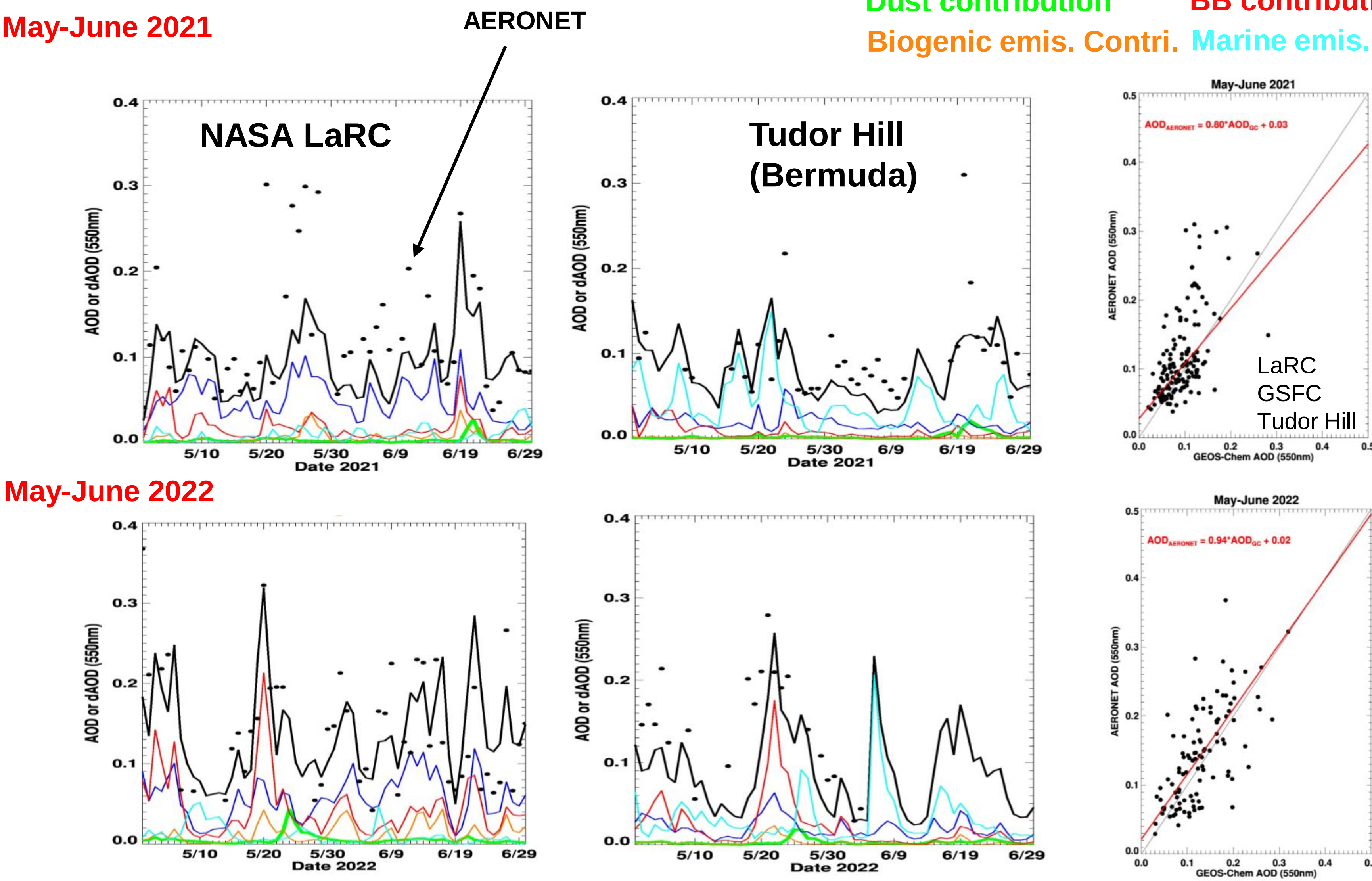
Vertical distribution of aerosol extinction: CALIOP vs GEOS-Chem



- The model underestimates CALIOP aerosol extinction in 2021 but agrees better with CALIOP in 2022.
- The model largely underestimates near-surface aerosol extinction at <32°N.

Model analysis of AERONET AODs

GC w/ BB PBL
Anthrop. contribution
Dust contribution
BB contribution
Biogenic emis. Contri.
Marine emis. contri.

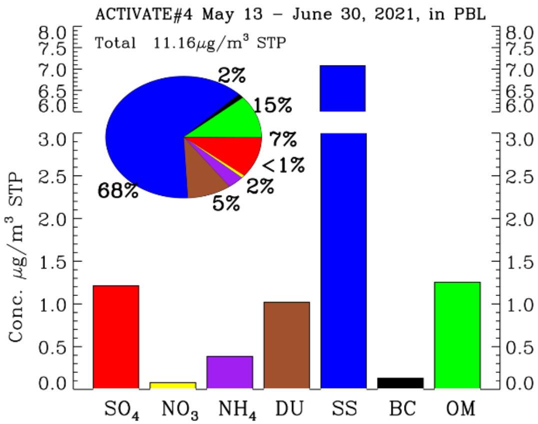


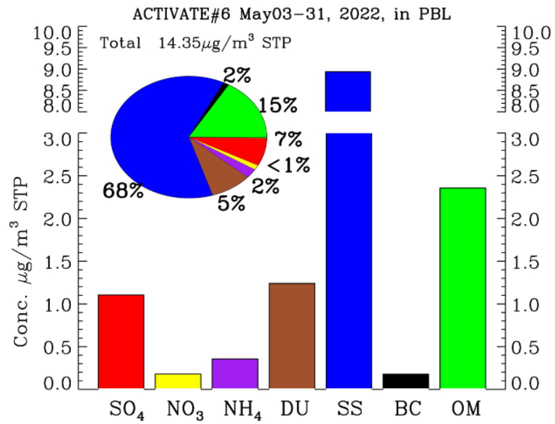
- ➢ Model simulates reasonably well the day-to-day variation of AOD & most of the biomass burning AOD events
- ➢ Bermuda AOD: large contribution from sea salt in 2021; impact of a strong fire event in 2022

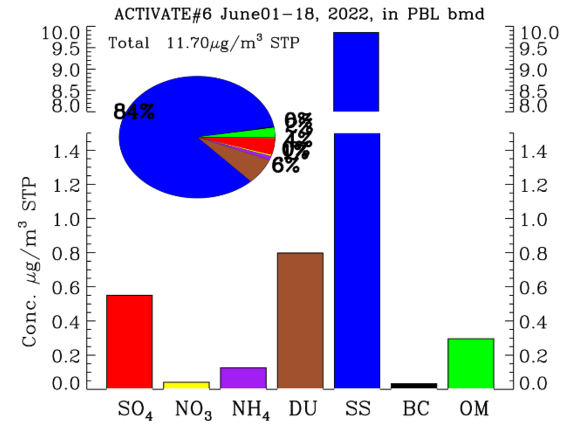
Simulated BL aerosol composition & AOD over the ACTIVATE domain

May 13 – June 30, 2021 | **May 3-31, 2022** | **June 1-18, 2022 (Bermuda)**

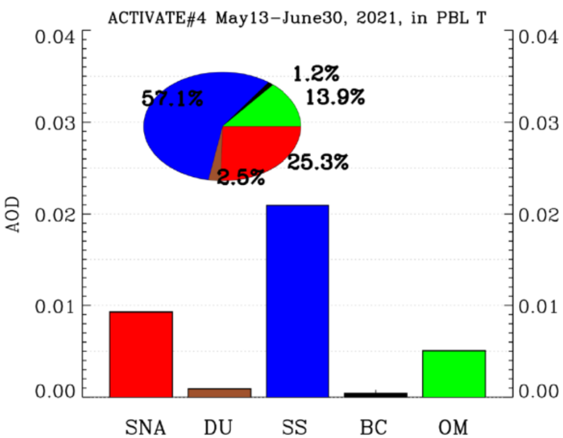
Mass conc.

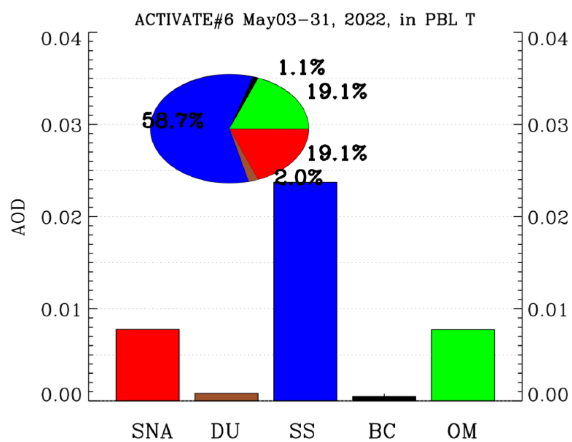


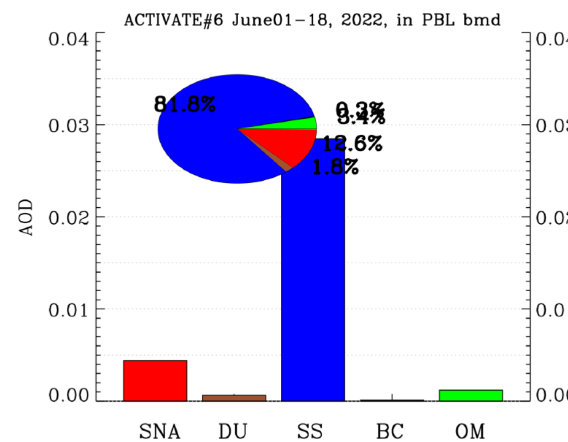




AOD







- ➢ Aerosol mass & AOD are dominated by sea salt, followed by organic matter and sulfate.
- ➢ Dust is the 2nd most abundant aerosol composition over the Bermuda domain during June 1-18, 2022.

Summary

- GEOS-Chem CTM is used to simulate tropospheric aerosols over the WNAO during May-June ACTIVATE 2021 and 2022, aiming to understand aerosol sources, transport, composition, and distribution in the region.

- BL outflow of NA pollution behind cold fronts weakened in early June as the Bermuda High shifted westward. Frontal and convective lifting are the major mechanisms for vertical transport of CO pollution. Convection (vs. PBL turbulent mixing in winter) is the dominant process responsible for upward transport of sea salt within and ventilation out of the PBL.

- <u>Near-coast domain</u>: BL aerosol composition & AOD are dominated by sea salt, followed by organic aerosol, sulfate, and dust.

- <u>Bermuda domain</u>: BL aerosol composition is dominated by sea salt followed by dust, sulfate, and organic aerosol (June 1-18, 2022). African dust: LRT to the remote North Atlantic & transported to Florida and the Gulf of Mexico followed by N- or NE- ward transport to the eastern U.S. coast and WNAO.

- The model generally captures the MODIS AOD enhancements (not shown) due to BB emissions over South U.S. and Southwest U.S. during May-June 2021 and 2022. It also simulates reasonably well the day-to-day variation of AERONET AOD & most of the BB AOD events.

Summary

- Relative to May-June 2021, May-June 2022 experienced much stronger continental outflow of biomass burning (BB) pollution to the WNAO. BB emissions contribute to >30% of organic aerosols in the lower troposphere during May-June 2021 while dominating organic aerosols during May 2022.

- A distinct layer of CO & aerosol enhancements at ~1-2 km altitude, resulting from trace species (precursors) uplifted over land followed by eastward advection and/or aerosol formation above the BL. Also seen at ~1.0 km altitude over the Bermuda area.

- Near-coast domain: The model reasonably simulates N American BL outflow of aerosols observed by HSRL-2 but tends to underestimate HSRL-2 aerosol extinctions in the free troposphere in 2021 as well as CALIOP near-surface aerosol extinctions (<32°N). Bermuda domain: Model generally underestimates Falcon AMS aerosol concentrations & HSRL-2 aerosol extinctions (especially in the free trop) around Bermuda.

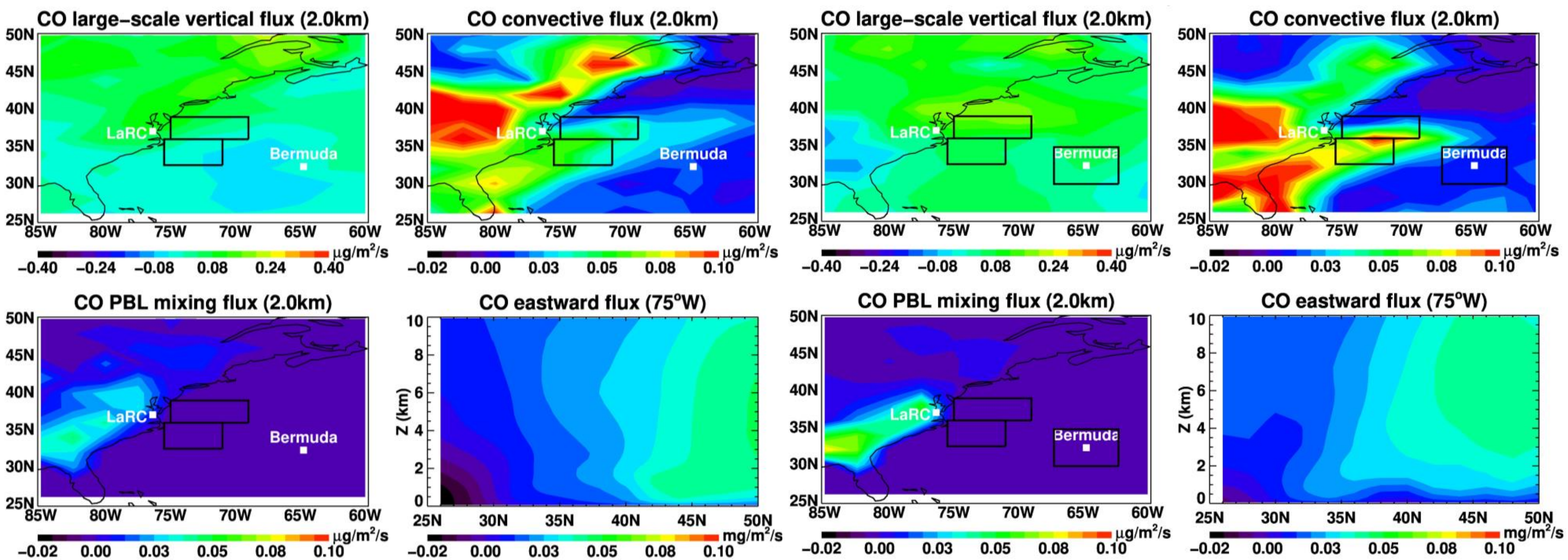
- Injecting biomass burning emissions to the 0-5.5 km altitude range in the model often helps capture the lidar-observed smoke plumes at ~5-6 km altitude.

Acknowledgements: NASA EVS-3 ACTIVATE mission; AERONET PIs and staff; GEOS-Chem support team; NASA Center for Climate Simulation (NCCS).

Backup Slides

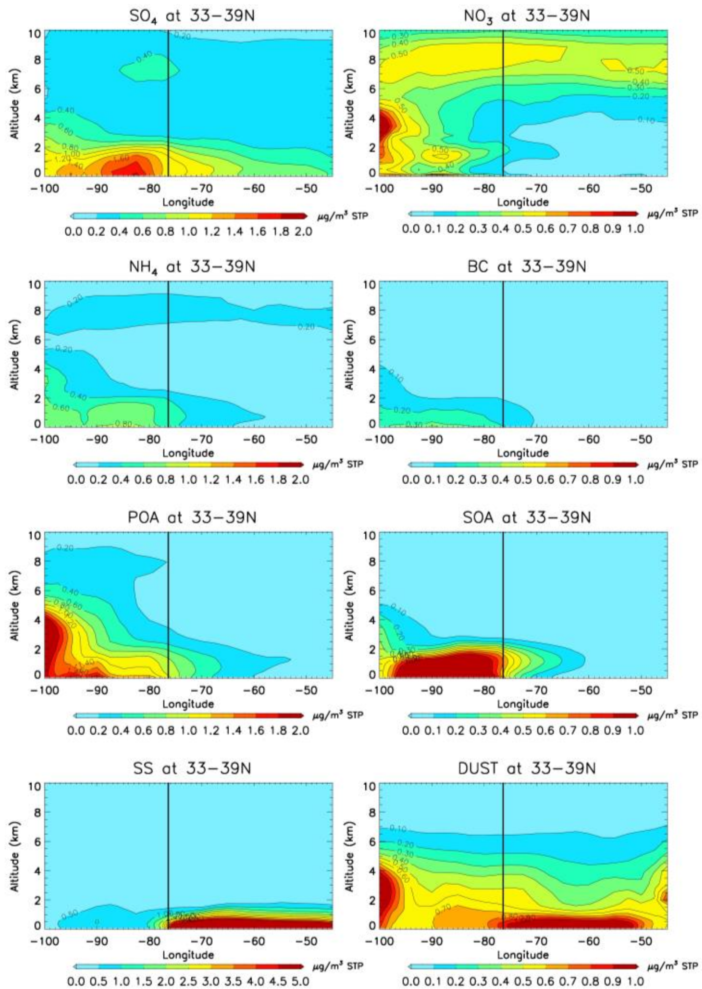
May 13 – June 30, 2021

May 3 – June 18, 2022

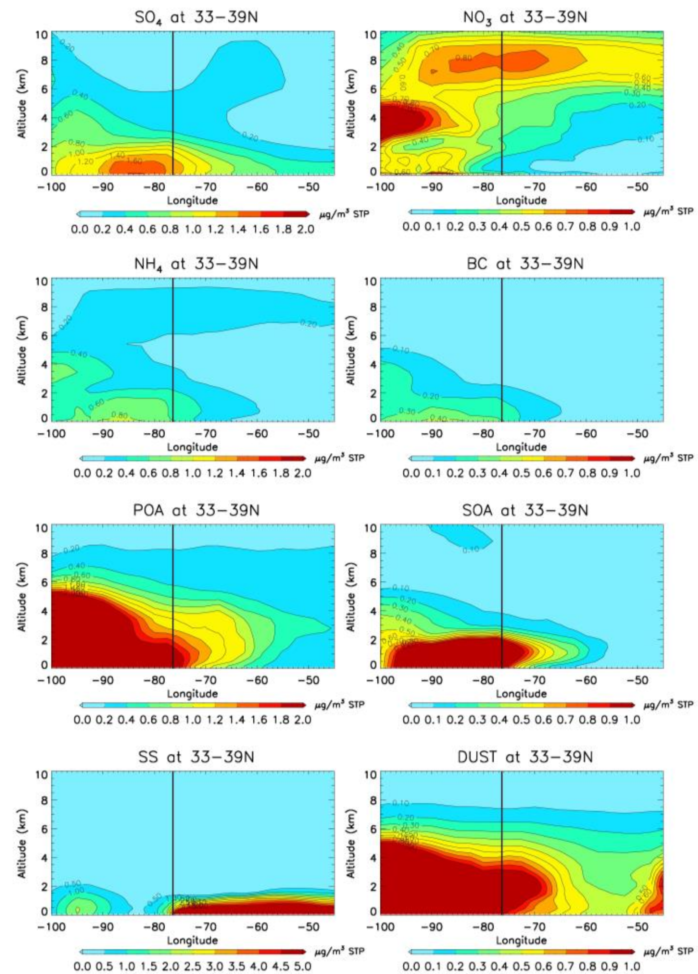


Model simulated large-scale (resolved) vertical fluxes, convective fluxes, and PBL turbulent mixing fluxes of CO at 2 km. Also shown is the latitude-height cross-section of CO eastward fluxes at 75°W. The three rectangular boxes denote major flight areas.

May 13 – June 30, 2021

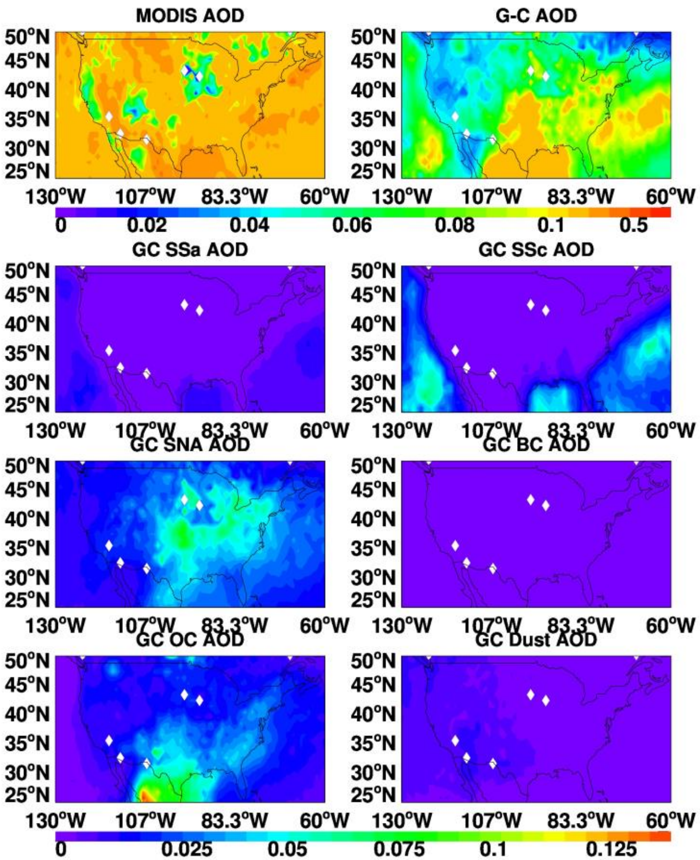


May 3 – June 18, 2022

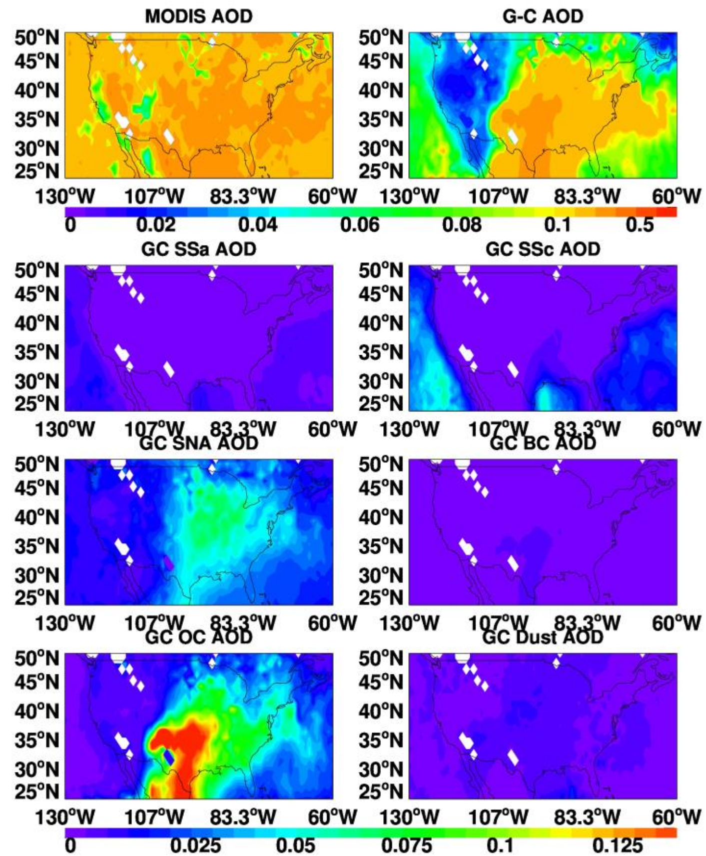


Model simulated concentrations of sulfate-nitrate-ammonium (SNA), black carbon (BC), primary organic aerosol (POA), secondary organic aerosol (SOA), sea salt, and dust. The plots show longitude-altitude cross-sections averaged over 33-39°N. Note different color scales among panels. The vertical lines indicate the longitude (76.4°W) of LaRC.

May 2021

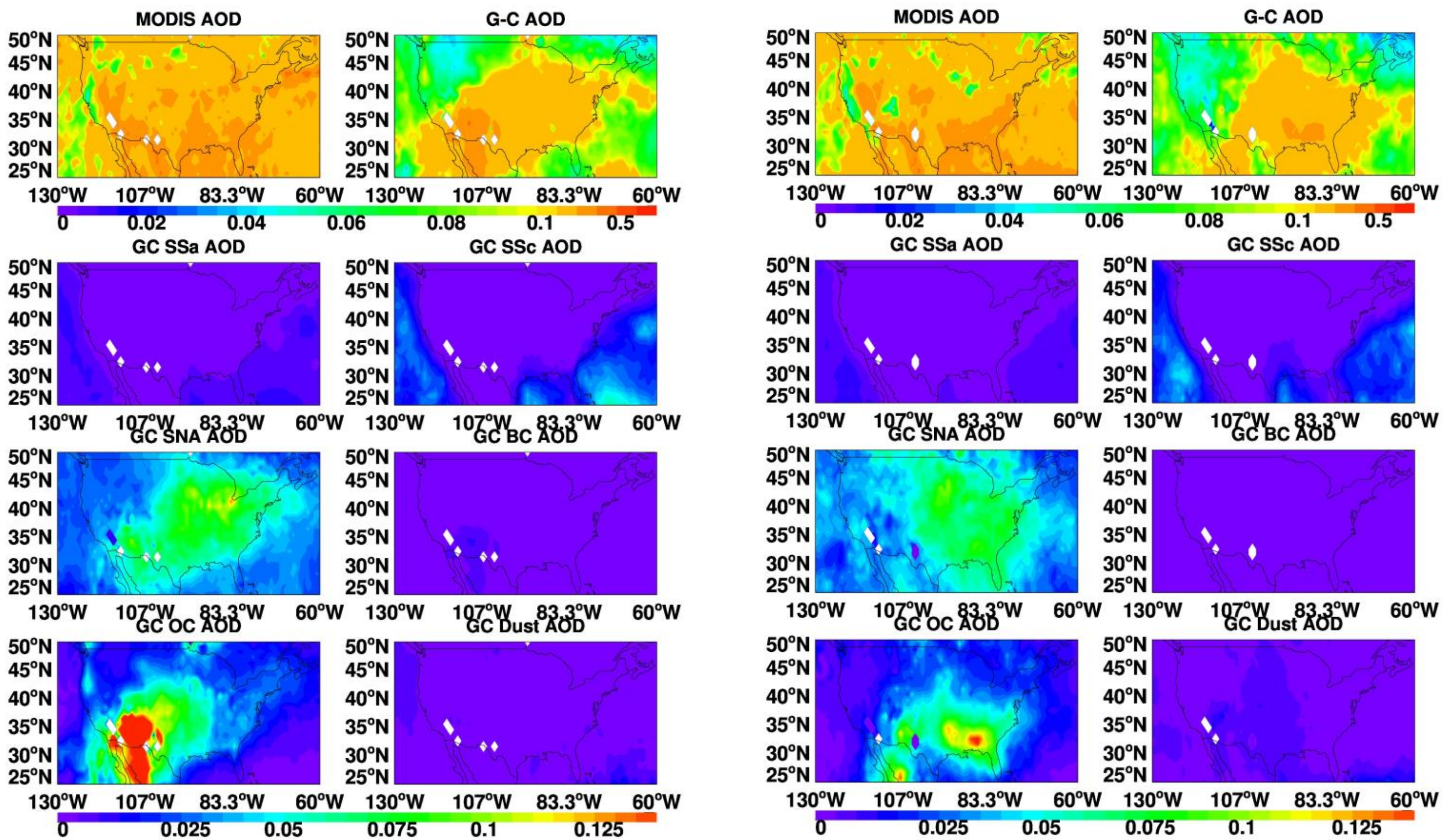


May 2022



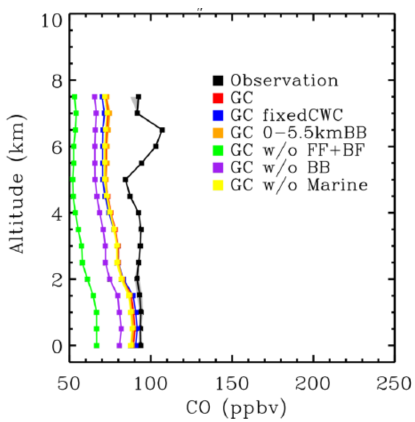
Evaluation of model monthly mean AODs with MODIS/Aqua retrievals (at 550nm) over North America and North Atlantic. Model output is sampled daily at 1:30 pm local time along the Aqua satellite orbit track. Also shown are contributions to the total AOD in the model from accumulation mode sea salt (SSa), coarse mode sea salt (SSc), sulfate-nitrate-ammonium (SNA), black carbon (BC), organic carbon (OC), and dust.

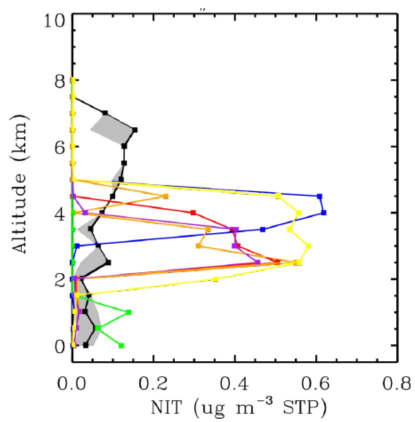
June 2021 June 2022

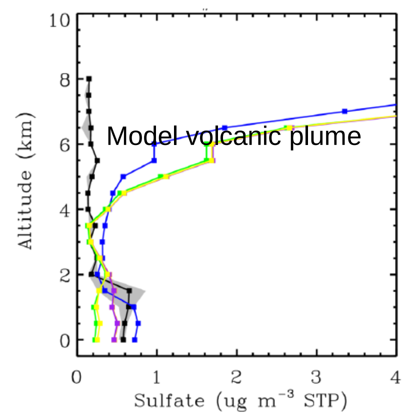


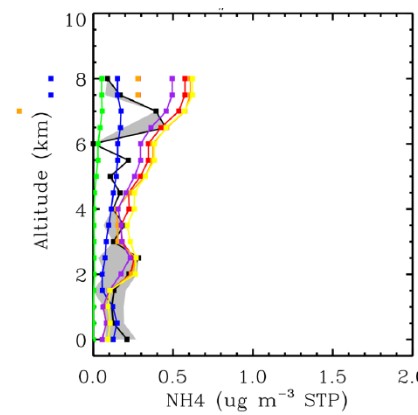
Evaluation of model monthly mean AODs with MODIS/Aqua retrievals (at 550nm) over North America and North Atlantic. Model output is sampled daily at 1:30 pm local time along the Aqua satellite orbit track. Also shown are contributions to the total AOD in the model from accumulation mode sea salt (SSa), coarse mode sea salt (SSc), sulfate-nitrate-ammonium (SNA), black carbon (BC), organic carbon (OC), and dust.

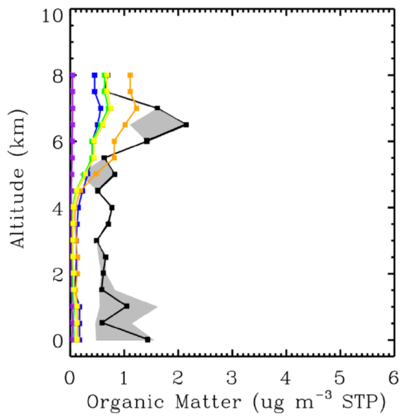
Bermuda 6/17/2022

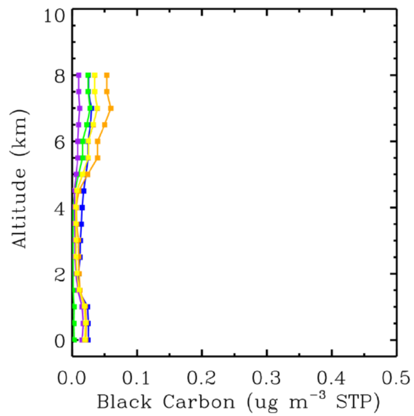




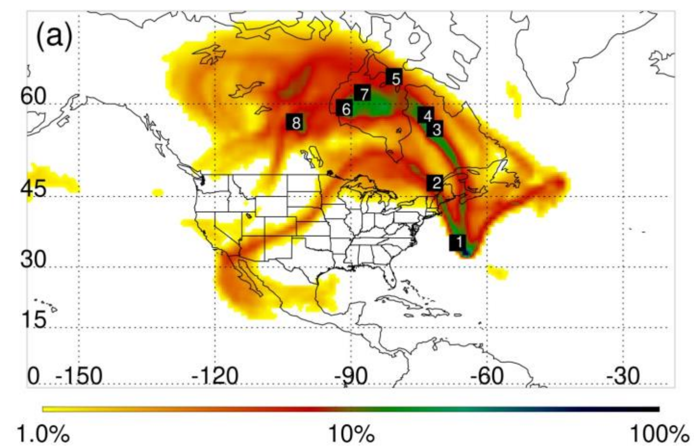






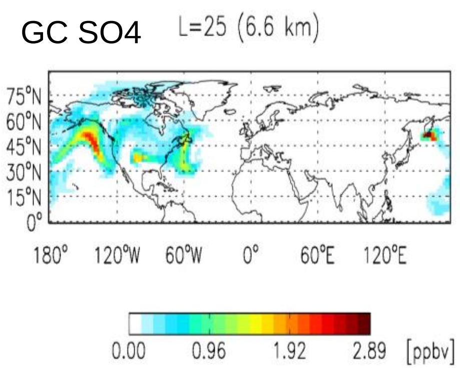


FLEXPART Trajectory 2022-06-17, 13:24Z, arriving altitude 6.5km



relative residence time

GC SO4



Need to use appropriate volcanic SO_2 emissions for 2022.