Investigating the impacts of wet scavenging on tropospheric aerosols during ACTIVATE using FLEXPART air mass history and GEOS-Chem simulations

Bo Zhang (bo.zhang@nasa.gov)[1,2], Hongyu Liu[1,2], Gan Luo[3], Luke Ziemba[2], Richard Moore[2], Michael Shook[2], Glenn Diskin[2], Josh DiGangi[2], Yonghoon Choi[2,4], and Armin Sorooshian[5]

1. National Institute of Aerospace, Hampton, VA 2. NASA Langley Research Center, Hampton, VA 3. State University of New York at Albany, Albany, NY

4. Analytical Mechanics Associates, Inc., Hampton, VA 5. University of Arizona, Tucson, AZ

Introduction

Interpreting airborne aerosol measurements can benefit from analysis based on the cloud and precipitation regimes encountered during aerosol air mass transport. This study aims to characterize aerosol composition measurements obtained during the NASA EVS-3 ACTIVATE aircraft campaign, using a suite of simulated air mass transport history indicators. These indicators include accumulated precipitation during transport (APT), source region, transport timescale and altitude (as illustrated in **Fig. 1**). By incorporating these parameters, we enhance our ability to interpret the measurements and gain a deeper understanding of the impacts of wet scavenging on the abundance and composition of tropospheric aerosols.

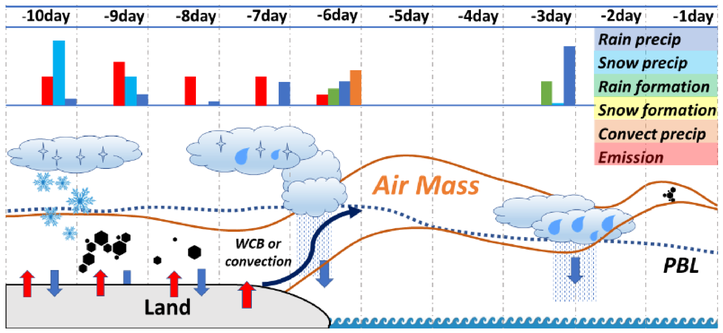


Figure 1. *A schematic of the Lagrangian Particle Dispersion Model (LPDM) method to determine air mass history. Air mass indicators we consider include precipitation flux and formation for rain (liquid) and snow (ice), emission, transport height, and lifting due to Warm Conveyor Belts (WCBs) / convection.*

ACTIVATE is a NASA Earth-Venture Suborbital-3 mission to characterize aerosol-cloud-meteorology interactions over the western North Atlantic using extensive in situ and remote sensing airborne measurements (Sorooshian et al., ESSD 2023).

Methodology

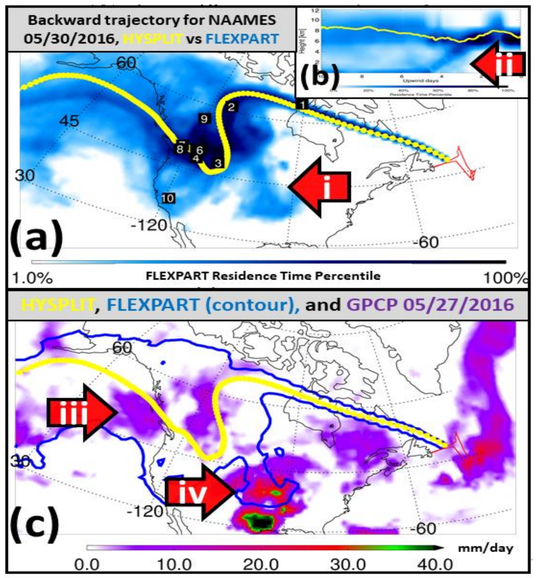


Figure 2. *Horizontally (a) and vertically (b) integrated FLEXPART residence time (blue) overlapped with the HYSPLIT single trajectory (yellow). (c) is the same with Global Precipitation Climatology Project (GPCP) precipitation. Arrows indicate transport features shown in FLEXPART: (i) air mass mixing/convection, (ii) mass portion lifted from PBL, (iii) and (iv) precipitation influence missed by HYSPLIT but captured by FLEXPART.*

- Traditional approaches to using airborne data often present averaged profiles, making it difficult to interpret aerosol observations due to various factors that affect them (e.g., RH, precipitation, cloud, emission).
- A Lagrangian Particle Dispersion Model (LPDM), FLEXPART (Pisso et al., 2019) is used to quantify RH, accumulated precipitation, and cloud fraction along transport pathway (**Fig. 2**).
- FLEXPART source-receptor sensitivity (SRS) function can be combined with emission inventories and reanalysis data (MERRA-2) to quantify sources and meteorological conditions along the transport history.

$$APT = \sum_t \sum_j (S_{j,t(j_0,t_0)} \cdot P_{j,t})$$

- FLEXPART calculates source–receptor sensitivity functions S(j, t, (j_0, t_0)), indicating how air masses from an upwind cell (j) at an earlier time (t) contribute to the entire plume released at the receptor (j_0) at time t_0.
- These sensitivities S(t, (j_0, t_0)) form a 3-D matrix that can estimate the impact of emissions and meteorological conditions from upwind air masses. For example, APT is derived by multiplying the sensitivity matrix S(t,(j_0, t_0)) by the 3-D precipitation flux field P(j,t).

FLEXPART-derived Air Mass Indicators

Table 1. FLEXPART air mass indicators.

Indicators	Full name	Data coupled with FLEXPART
Accumulated Precipitation along Transport (APT)		
FP_PFLLSAN *	AP** from large-scale and anvil rain flux	MERRA-2 (0.5° × 0.625°)
FP_PFILSAN	AP from large-scale and anvil snow flux	
FP_DQRLSAN	AP from large-scale and anvil precipitation formation	
FP_DQRCU	AP from convective precipitation formation	
FP_PFLCU	AP from convective rain flux	
FP_PFICU	AP from convective snow flux	
Source, transport, and meteorology indicators		
FP_COf	CO from wildfire emissions	GFAS v1.2
FP_COanth	CO from anthropogenic emissions	EDGAR v6.1
***FP = FLEXPART; **AP = Accumulated Precipitation**		

GEOS-Chem simulations:

- GC v11-01 & v12.6.0 (MERRA-2, 2°×2.5°).
- Sulfate-nitrate-ammonium aerosol thermodynamics coupled to gas-phase chemistry, mineral dust, sea salt, organic carbon.
- EDGAR v4.2 for anthropogenic, QFED biomass burning, biogenic emissions (MEGAN), and marine DMS.
- GC v12.6.0 provides production and loss terms of aerosols for coupling with FLEXPART.
- Simulations conducted for the winter (Feb-Mar) & summer (Aug-Sep) deployments of ACTIVATE 2020.

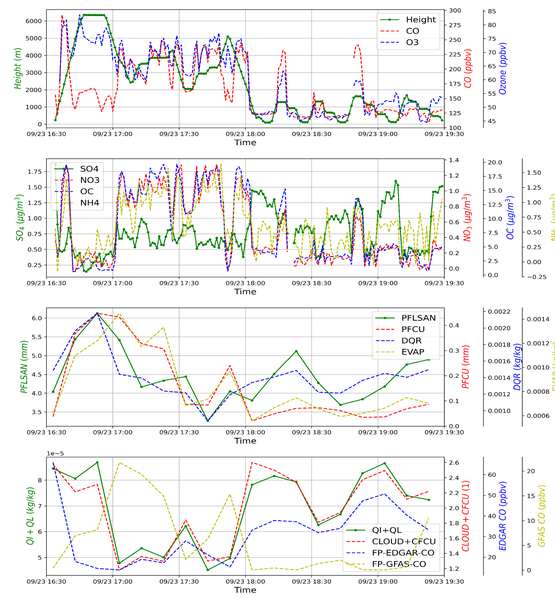


Figure 3*. A time series of measured trace gases (1st panel), aerosols (2nd panel), and FLEXPART-simulated accumulated precipitation along transport (APT) (3rd panel), and sources of carbon monoxide (CO; 4th panel) for air masses encountered during the ACTIVATE Falcon flight on September 23, 2020.*

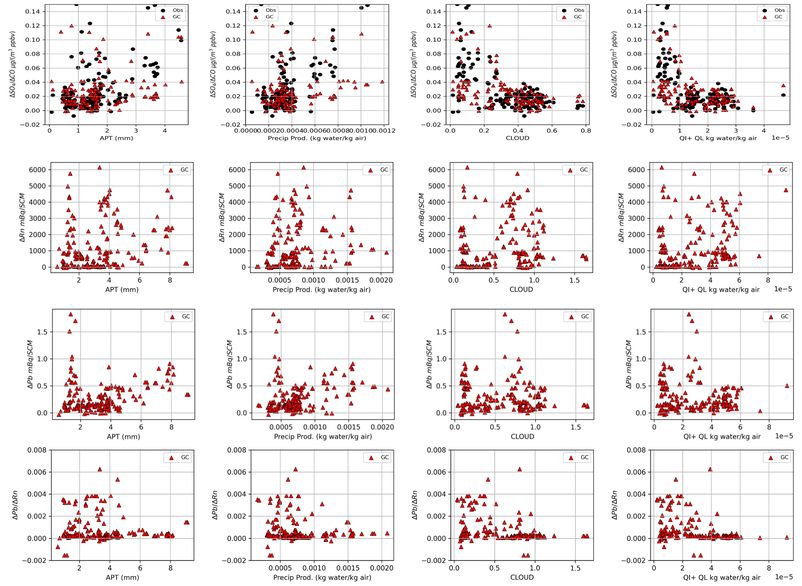


Figure 4*. Relationships between sulfate aerosol or radionuclide tracer enhancements (ΔSO$_4$, ΔPb-210, and ΔRn-222) and accumulated precipitation/cloud quantities during transport. Hourly GEOS-Chem outputs (red triangles) are sampled at the Falcon AMS aerosol measurements (every 10 min; black dots). Radionuclide tracers are from GEOS-Chem only. Aerosol enhancement is calculated by subtracting background levels (average of the lowest 5% measurements) from measured concentrations and then dividing it by the corresponding enhancement of CO or Rn-222 (e.g., ΔSO$_4$/ΔCO). The x-axis in each panel displays different FLEXPART air mass indicators, including total APT (PFLLSAN + PFLCU + PFILSAN + PFICU), precipitation formation rate (DQRLSAN + DQRCU), cloud fraction (CLOUD), and cloud water content (QL+QI). See Table 1 for MERRA-2 meteorological variables.*

- APT includes large-scale and anvil precipitation (PFLLSAN), convective precipitation (PFLCU), large-scale and anvil snow (PFILSAN), and convective snow (PFICU) fluxes (see **Table 1**). These APT terms are used to study aerosol wet scavenging by each precipitation type and its impact on aerosol composition (**Fig. 3 & 4**).
- CO contributions indicate CO emission impacts due to biomass burning (GFAS v1.2) and anthropogenic (EDGAR v6.1) sources, respectively (**Fig. 3**).
- Decrease in simulated ΔPb/ΔRn with increasing accumulated precipitation (or cloud water) suggests the impact of wet scavenging on aerosols. ΔSO$_4$ increases with increasing accumulated precipitation but decreases with accumulated cloud water, suggesting the effects of factors other than wet scavenging (e.g., in-cloud production).

Summary

- FLEXPART indicators provide valuable insights into air mass distribution and source attributions, enhancing our understanding of air mass composition.
- Accumulated precipitation and cloud quantities during transport exhibit significant variations across different aerosol samples, reflecting diverse atmospheric conditions along transport.
- Initial results suggest that different cloud and precipitation processes during air mass transport have varying impacts on aerosol composition.
- Ongoing research aims to investigate these impacts and establish the relationships between wet scavenging processes and changes in aerosol compositions.

References

Sorooshian, A., et al., Spatially-coordinated airborne data and complementary products for aerosol, gas, cloud, and meteorological studies: The NASA ACTIVATE dataset. Earth Syst. Sci. Data, 15, no. 8, 3419-3472, doi:10.5194/essd-15-3419-2023.

Pisso, I., Sollum, E., Grythe, H., Kristiansen, N. I., Cassiani, M., Eckhardt, S., ... & Stohl, A., The Lagrangian particle dispersion model FLEXPART version 10.4. Geoscientific Model Development, 12(12), 4955-4997, 2019.

Acknowledgement: This work is supported by the NASA's EVS-3 ACTIVATE mission and ACCDAM Program. High-end computing resources are provided by the NASA Center for Climate Simulations (NCCS).