Toward a High Degree of Freedom Retrieval of BrO from Mountaintops

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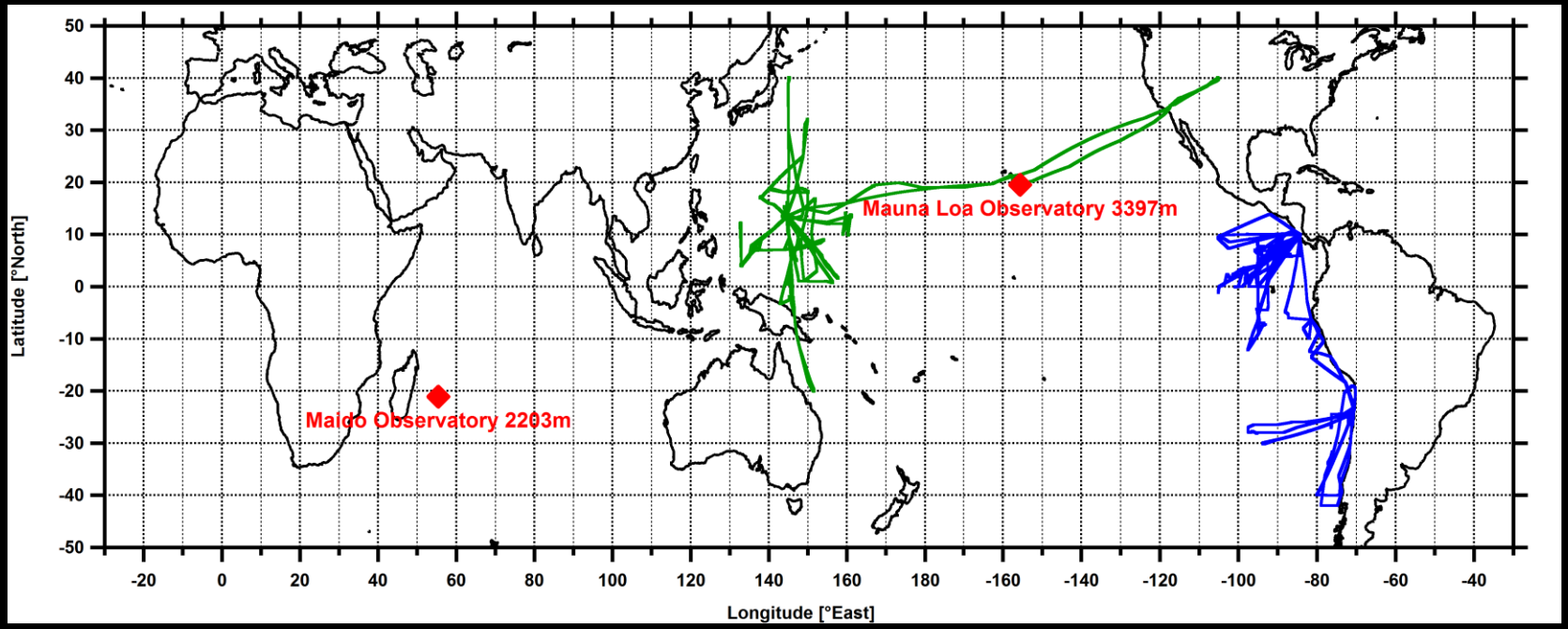
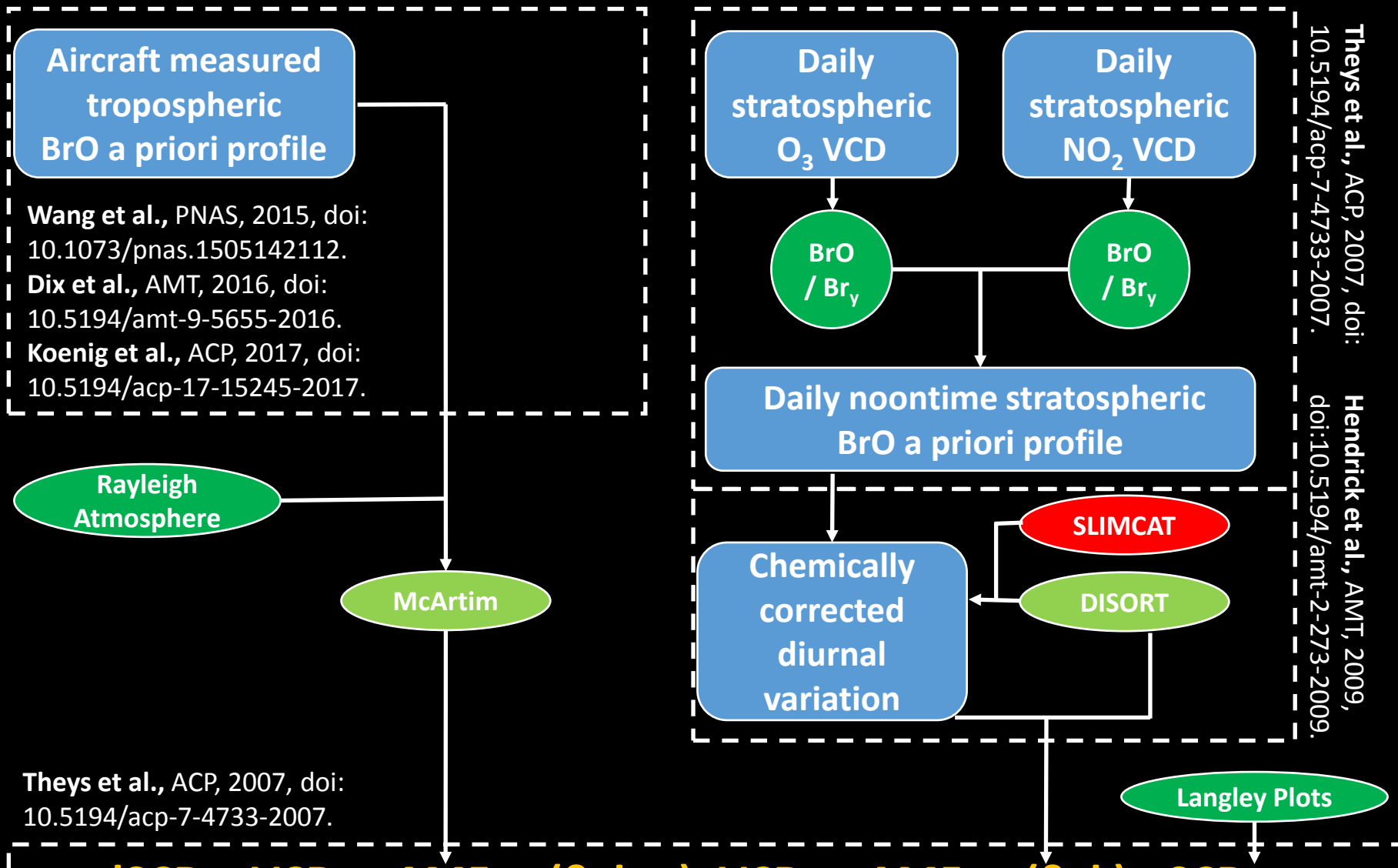
MAX-DOAS on mountaintops on tropical ocean islands

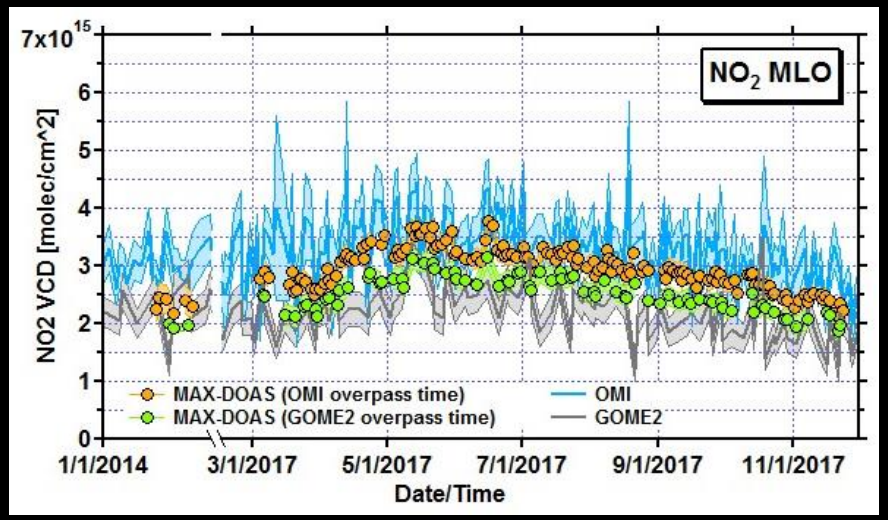


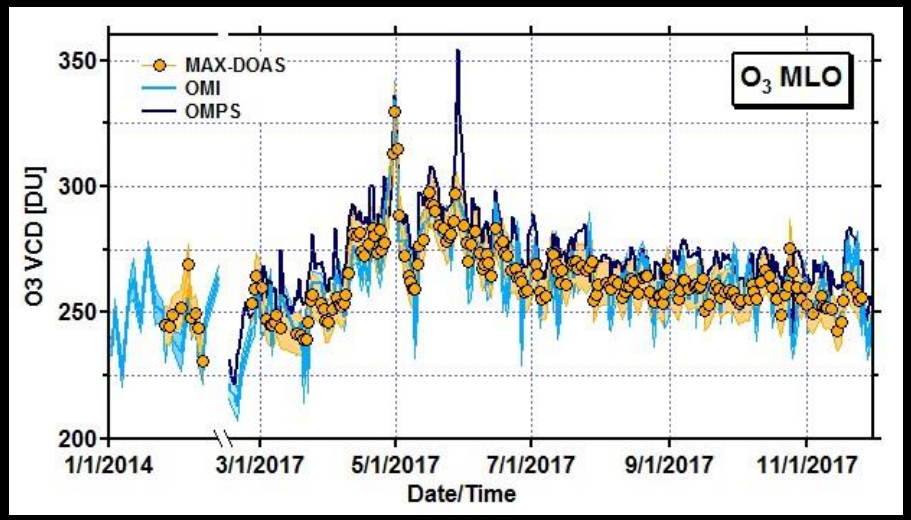
- Mountaintop Observatories at Mauna Loa on Hawai'i and Maïdo on La Réunion
- Aircraft campaigns (TORERO in blue; CONTRAST in green) used for BrO a priori profiles
  - CONTRAST included research flight near MLO during prior 2014 deployment, useful as a case study to test retrievals
- Both mountaintops operating since February 2017 with some interruption
  - Intensive Operating Period currently underway at Maïdo

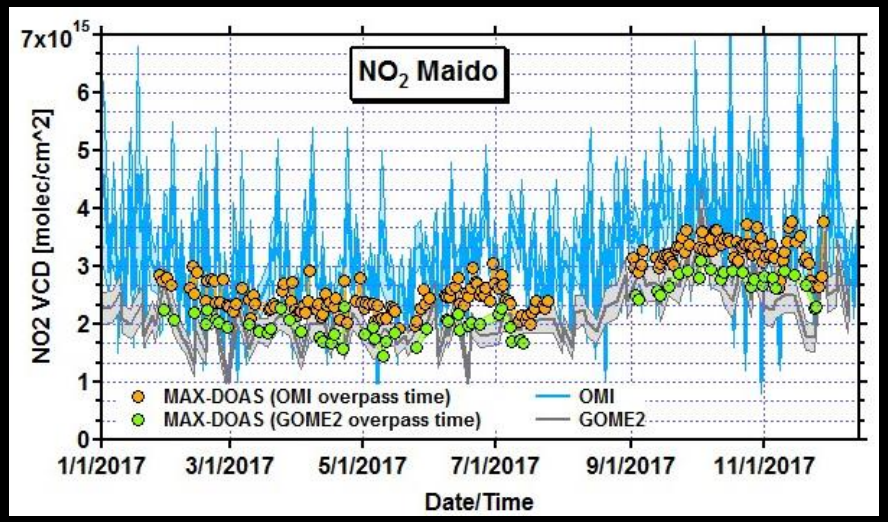
Generation 1 retrieval approach

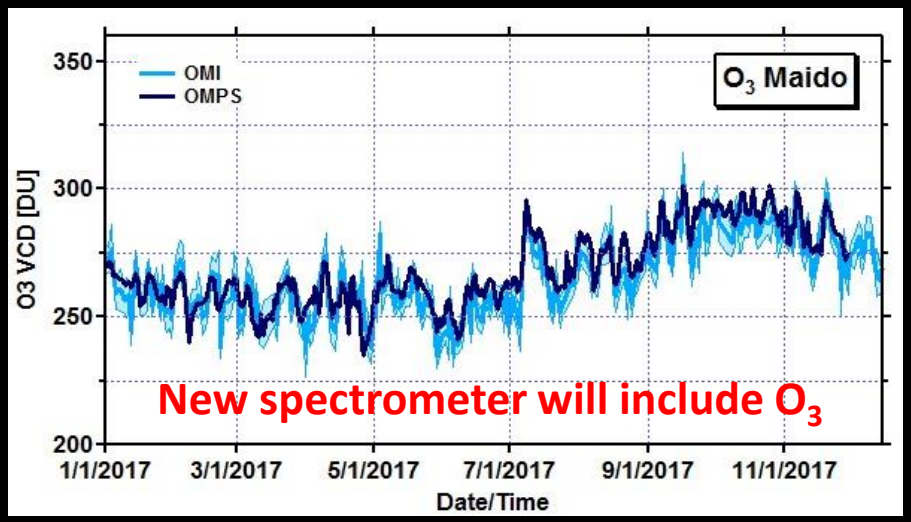


Stratospheric NO_2 and O_3 VCDs to build stratospheric BrO a priori



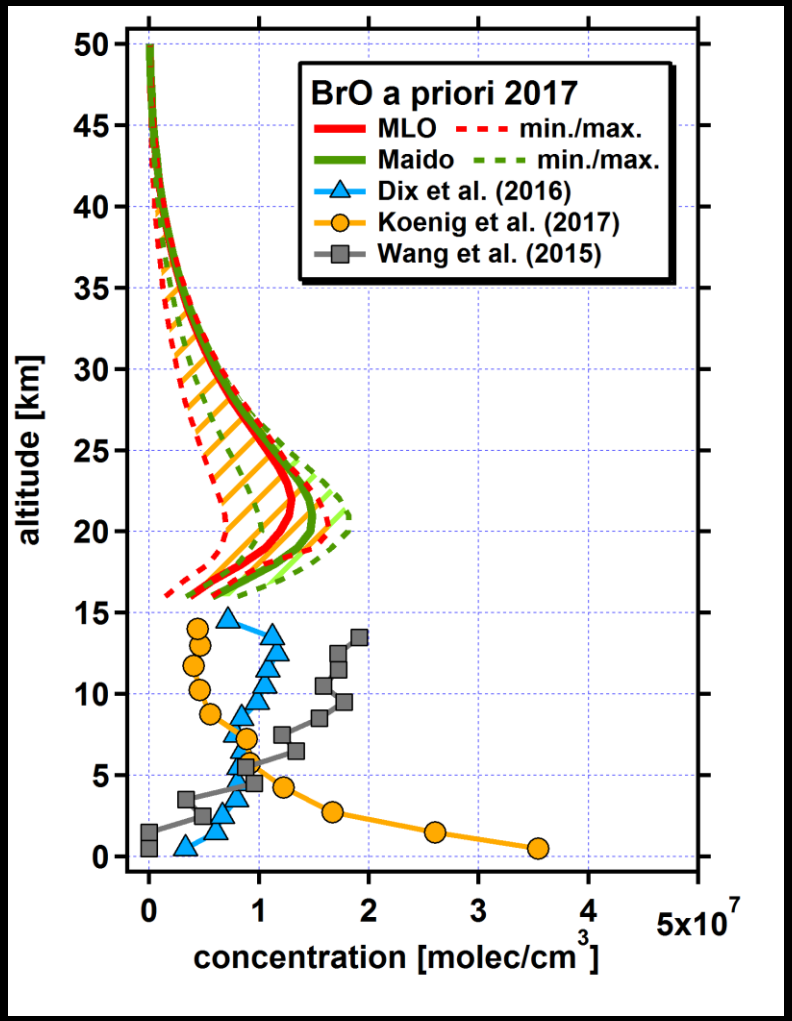






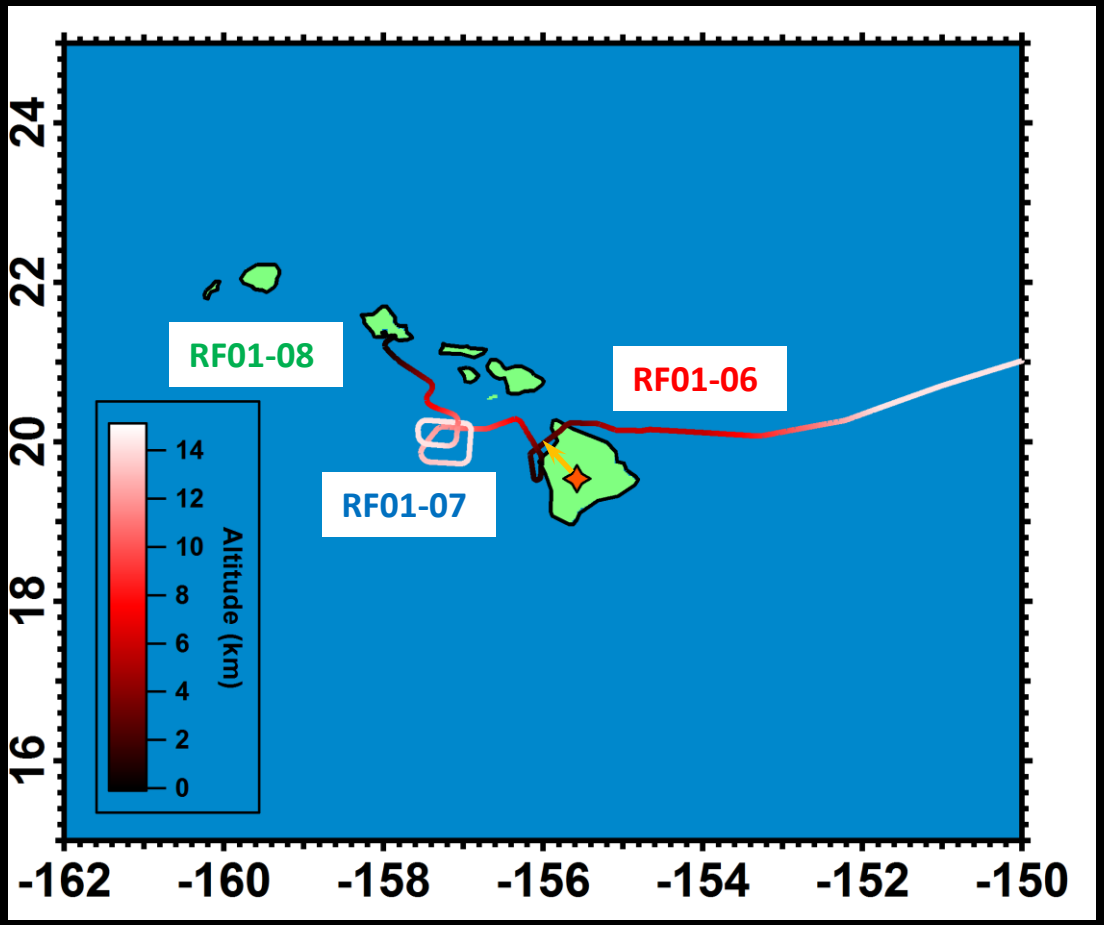
Comparison of a priori profiles

- Maximum stratospheric a priori correspond to O_3 and NO_2 maxima, late spring to early summer
- Wang a priori reflects average of optimal estimation case studies from TORERO
- Dix a priori reflects TORERO campaign average using parameterization
- Koenig a priori reflects CONTRAST optimal estimation case studies
  - Near surface concentrations reflect averaging over wide variability
- Connecting tropospheric and stratospheric profiles presents a challenge.
  - Limited information from measurements and few studies quantify BrO both below and above the tropopause



CONTRAST RF01 case study

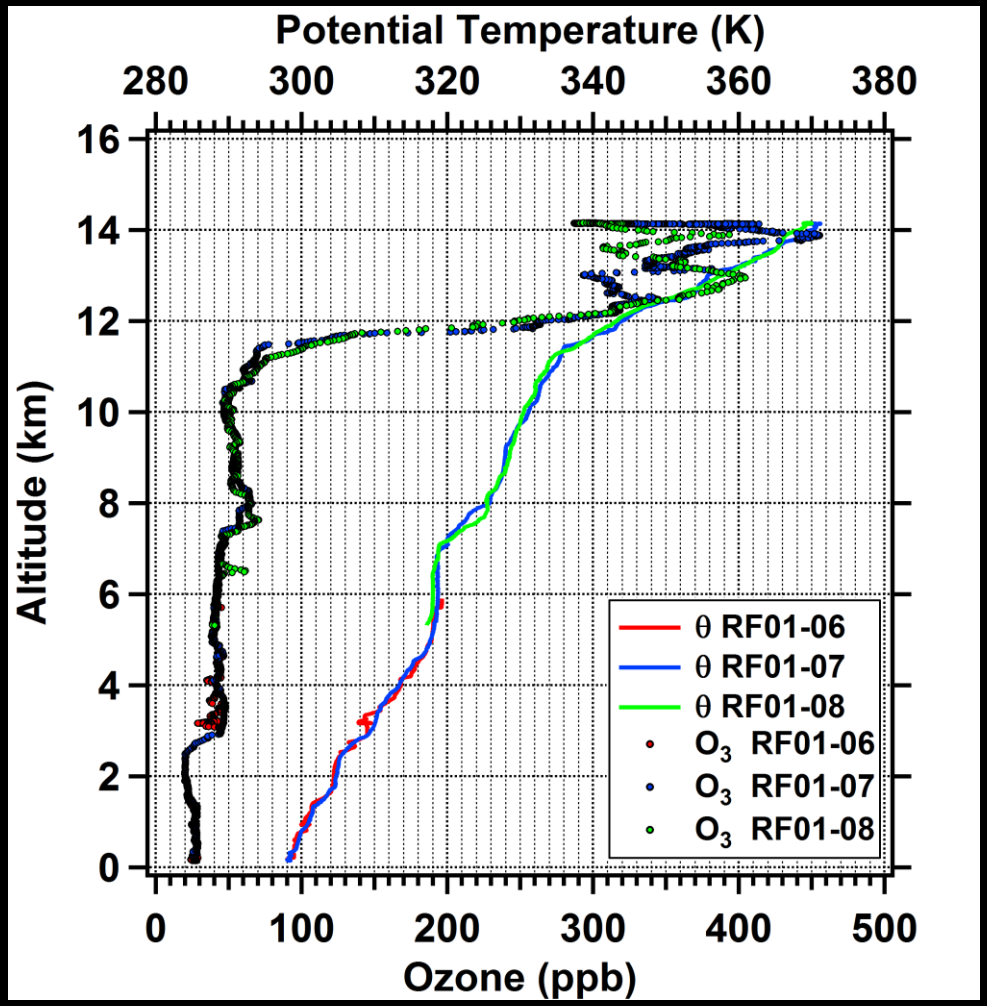
- Measurement site was cloud bound at time of flights precluding direct comparison
- Profiles are numbered sequentially for each ascent or descent including interior level legs



CONTRAST RF01: January 11, 2014

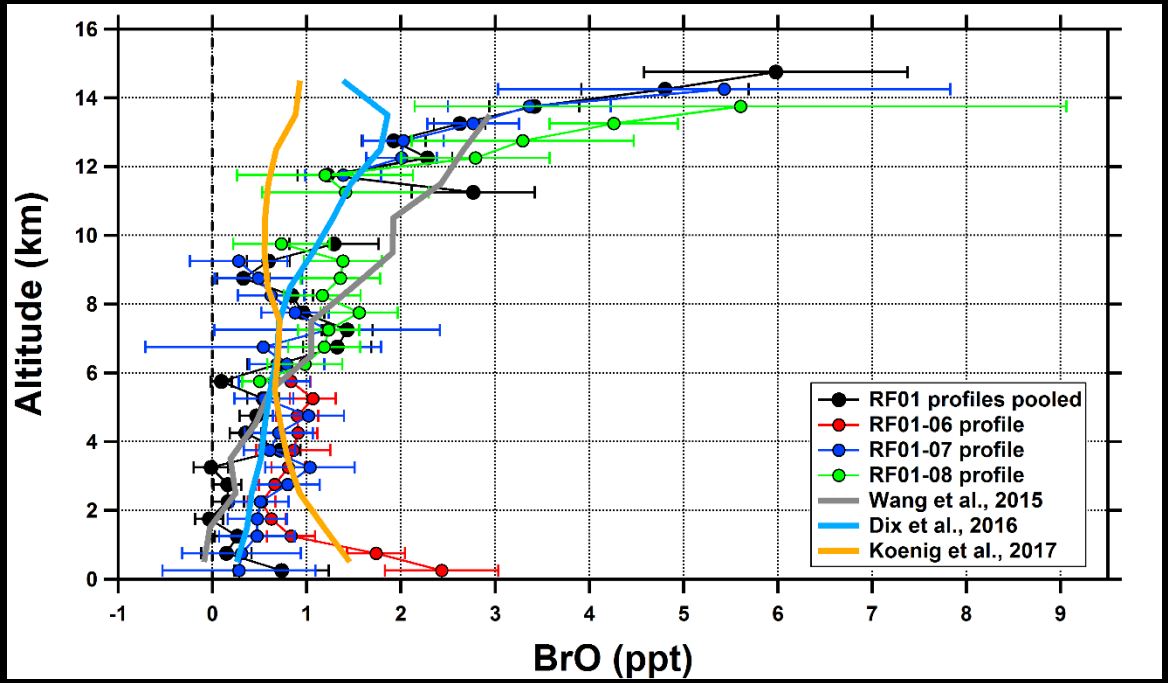
Chemical and dynamic context for case study

- Rossby wave breaking event leads to stratospheric intrusion
  - evident from increased O_3 and potential temperature
- Broadly the three profiles are similar, but
  - RF01-06 has a lofted layers around 3 -4 km
  - Layering of O_3 is different at high altitude



Comparison of case study with campaign averages

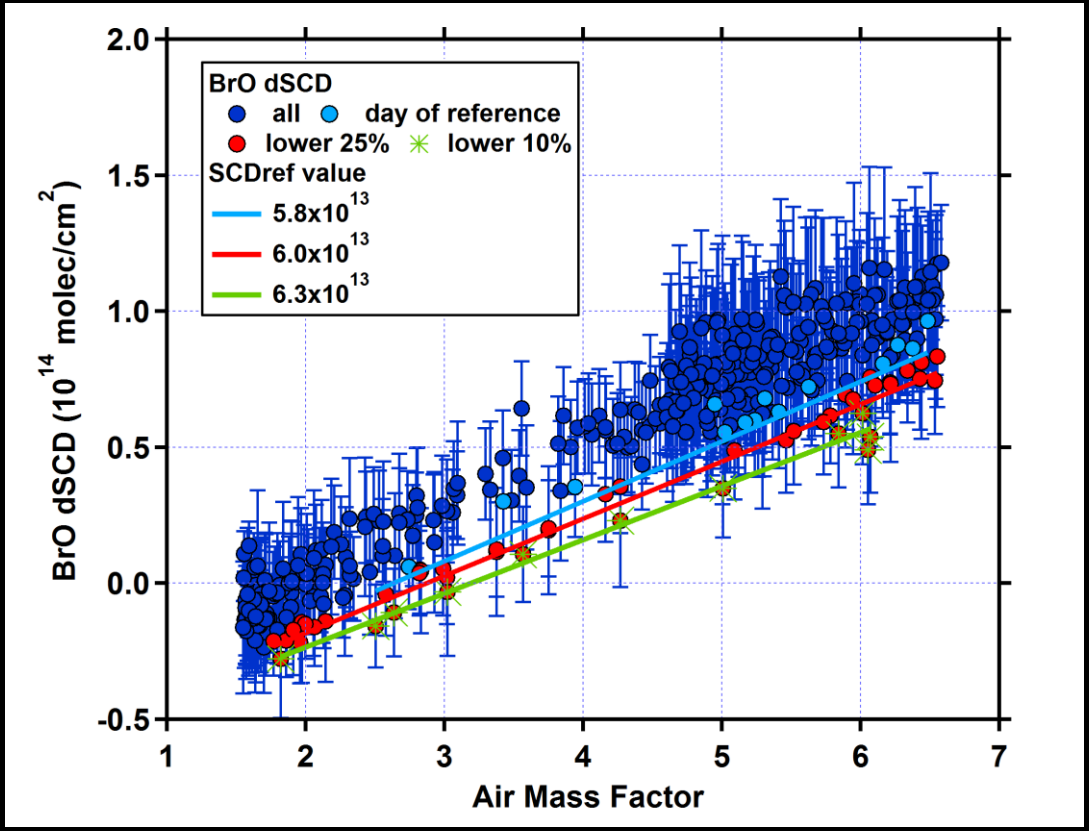
- Using data from all three profiles and flat concentration a priori run first optimal estimation
- Then use retrieved profile as a priori for optimal estimation of individual profiles



- RF01-06 has significant BrO at surface as well as enhanced BrO aloft
- Difference in Ozone layering appears smoothed over
- For mountaintop pooled profile is roughly same as Dix et al.

Langley plot determination of SCD_{Ref} for January 2014 data

- Using different subsets of data SCD_{Ref} is between 5.8×10^{13} and 6.3×10^{13} molecules cm^{-2}
- With SCD_{Ref} and a priori profiles to get Air Mass Factors have components needed for Theys et al. 2007 equation



BrO dSCDs from MLO for January 2014

Application of Theys et al. 2007 method

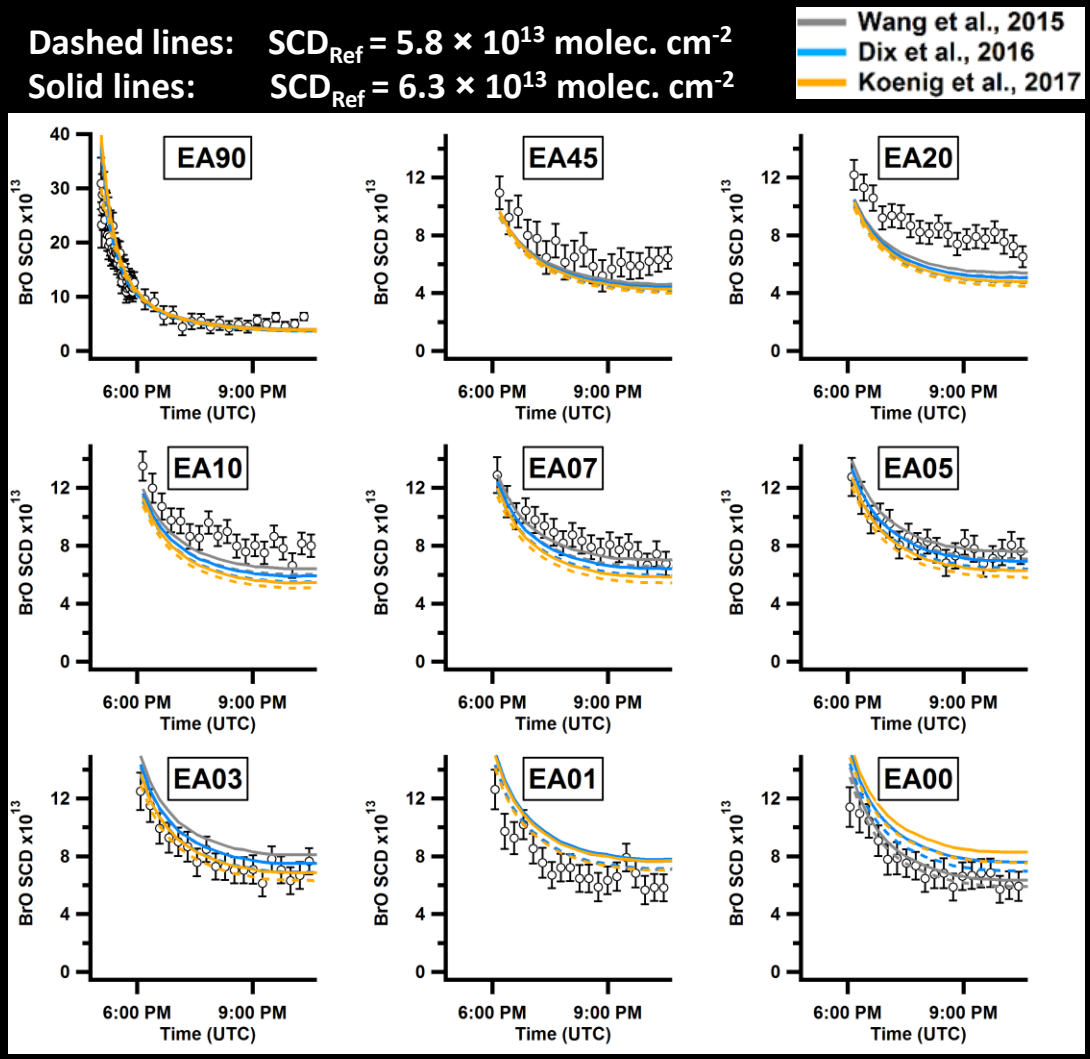
$$dSCD = VCD_{trop}AMF_{trop}(\theta,\phi,\alpha)+VCD_{strat}AMF_{strat}(\theta,\phi) - SCD_{Ref}$$

Tropospheric a priori	SCD_{ref} (molec cm^{-2})	Trop VCD (<15km) (molec cm^{-2})	Strat VCD (>15km) (molec cm^{-2})	Total VCD (molec cm^{-2})
Wang et al.	$5.8x10^{13}$	$0.98\pm0.04x10^{13}$	$1.48\pm0.04x10^{13}$	$2.46\pm0.08x10^{13}$
Dix et al.	$5.8x10^{13}$	$0.77\pm0.03x10^{13}$	$1.77\pm0.04x10^{13}$	$2.54\pm0.07x10^{13}$
Koenig et al.	$5.8x10^{13}$	$0.71\pm0.04x10^{13}$	$2.07\pm0.03x10^{13}$	$2.78\pm0.07x10^{13}$
Wang et al.	$6.3x10^{13}$	$1.10\pm0.04x10^{13}$	$1.45\pm0.05x10^{13}$	$2.55\pm0.09x10^{13}$
Dix et al.	$6.3x10^{13}$	$0.87\pm0.04x10^{13}$	$1.77\pm0.04x10^{13}$	$2.64\pm0.08x10^{13}$
Koenig et al.	$6.3x10^{13}$	$0.81\pm0.04x10^{13}$	$2.11\pm0.04x10^{13}$	$2.92\pm0.08x10^{13}$

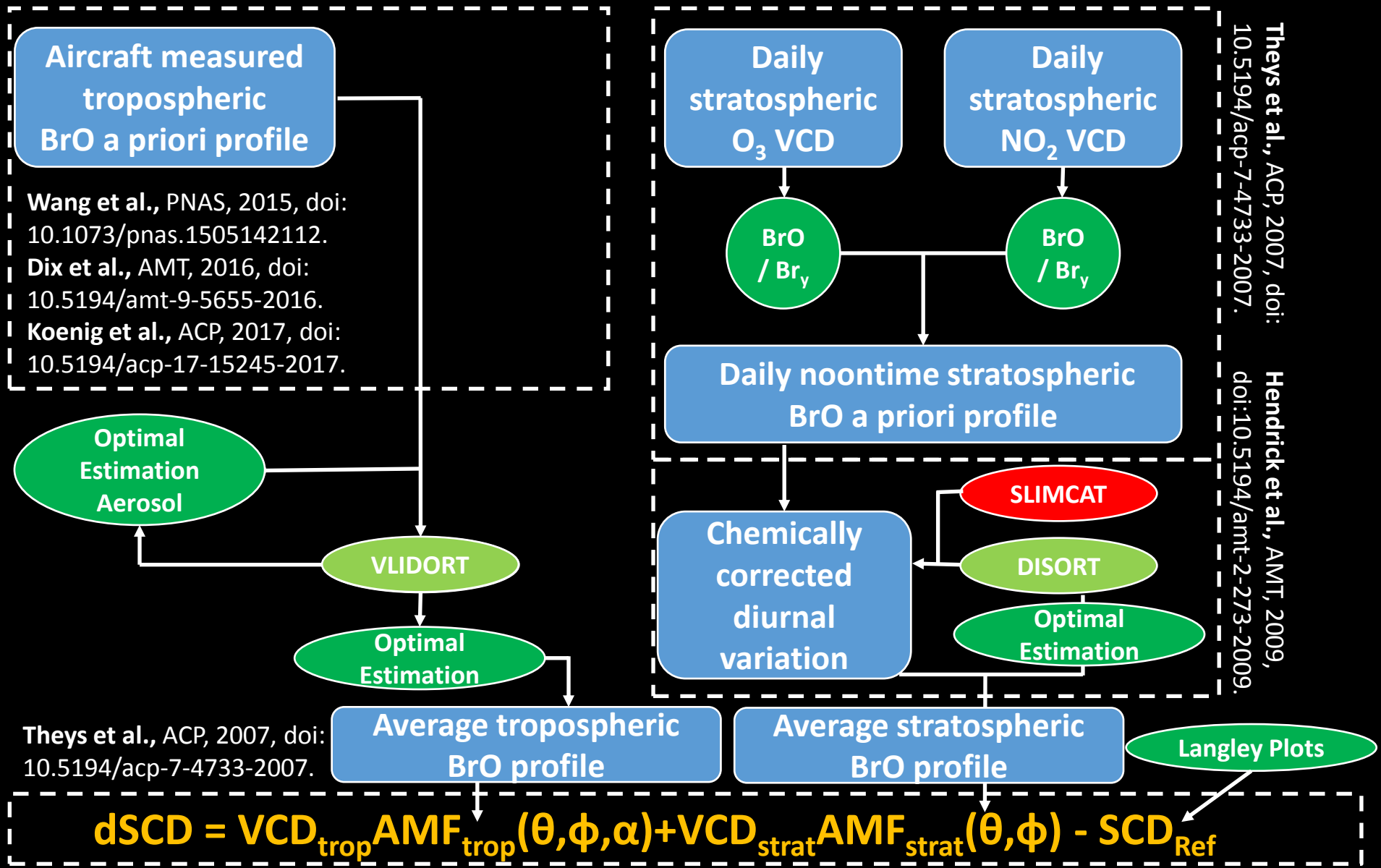
- Tropospheric a priori profiles with more BrO aloft fit a larger tropospheric VCD, a smaller stratospheric VCD, and a larger total BrO VCD
- Stratospheric VCD increases out of proportion to decrease in tropospheric VCD

Examination of fitted VCDs to measured dSCDs at different elevation angles

- Zenith data indicate that lower SCD_{Ref} is more likely to be accurate
- Wang et al. fits to low angles most closely, underestimates high angles least
- Fits suggest that even Wang et al. overestimates low altitude BrO, and underestimates BrO aloft

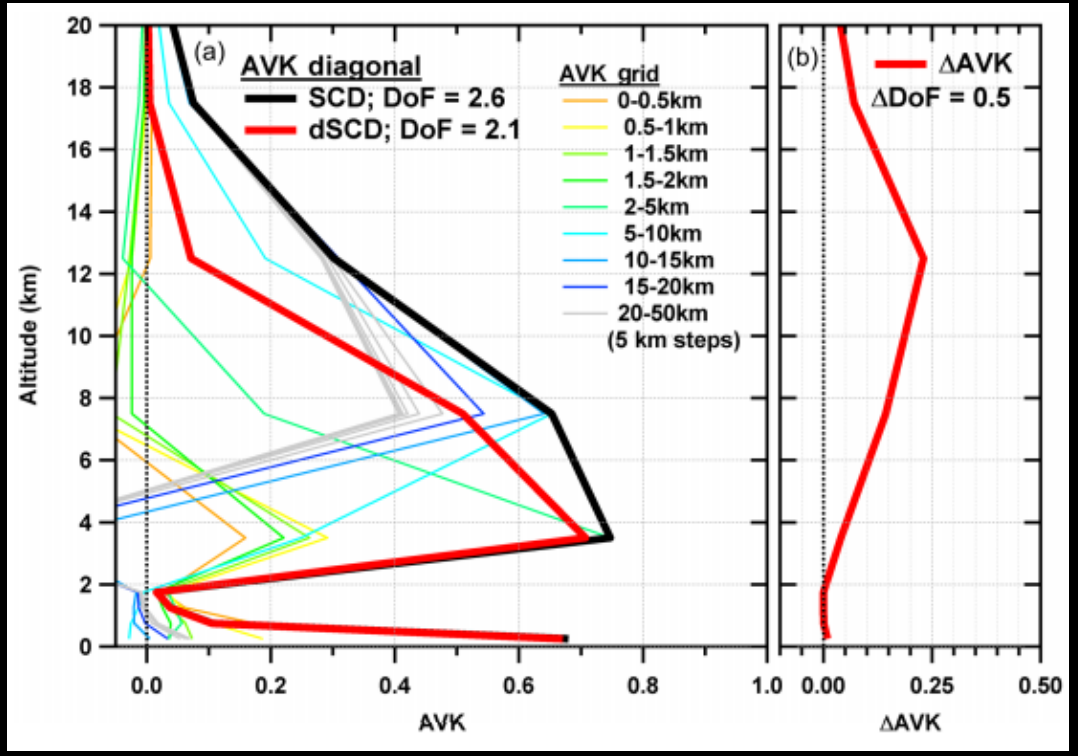


Generation 2 retrieval approach



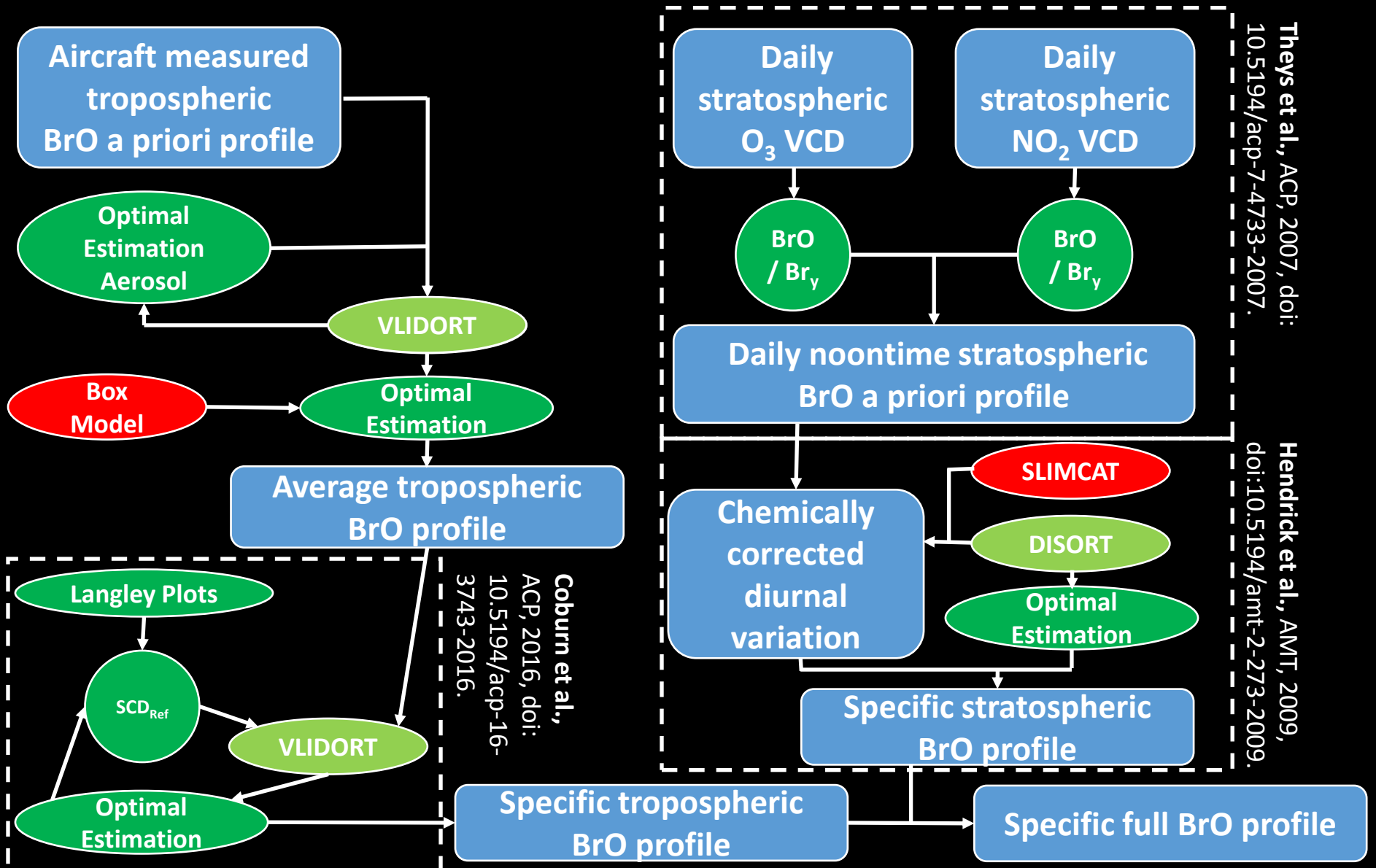
Self consistency determination of SCD_{Ref} to increase Degrees of Freedom aloft

- Scale BrO profile iteratively until a priori and a posteriori SCD_{Ref} match
- NB: altitude grid resolution is not constant
- Additional information is located 2-20 km above instrument altitude



Coburn et al., ACP, 2016, doi: 10.5194/acp-16-3743-2016.

Generation 3 retrieval approach



Outlook

- Need to efficiently capture radiative transfer
  - Examining VLIDORT as an option
    - mountaintops are near Rayleigh, explicit O_2-O_2 based optimal estimation may be practical
    - LUT approach also being examined
  - Possible parameterization approach?
- Need to define data set with which to test approaches
  - Have roughly one year of data from Maïdo and MLO
  - Generating cloud flag leveraging O_2-O_2 and color ratios