OBSERVATIONS OF BRO, NO_2 AND H_2O IN THE TROPICAL UTLS DURING THE 2013 ATTREX EXPERIMENT

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Thanks for NOAA O3 & UC Boulder/AMES SSFR data

University of California Los Angeles

University of Heidelberg

Introduction

Relative sensitivity of surface temperature upon unit masses of GHG

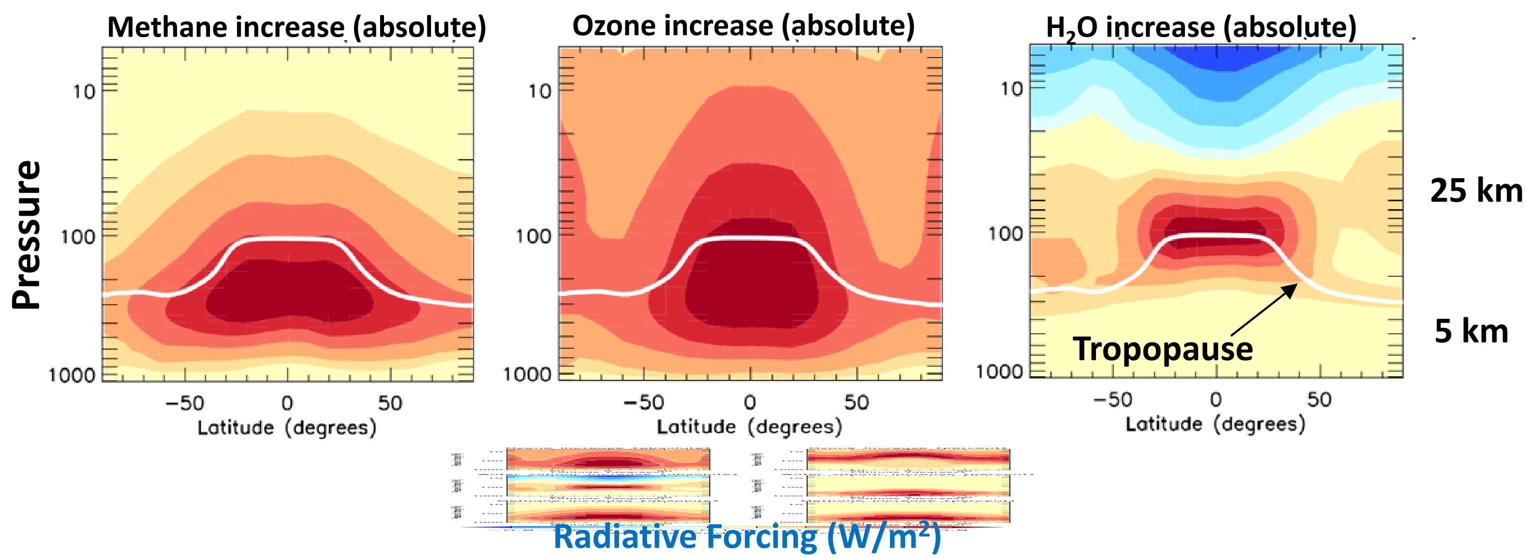


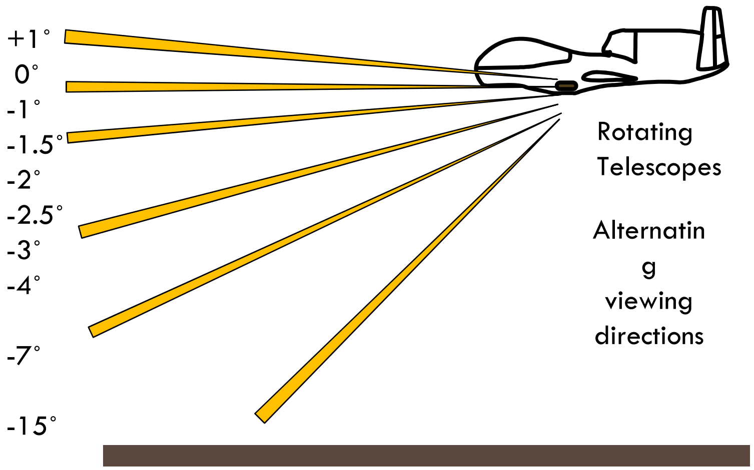
Figure: Illustration of how the surface impact depends on altitude and latitude where the CH_4, O_3 and H_2O changes take place. Shading shows relative surface impact, measured as radiative forcing from a fixed mass increase applied to different altitudes. Red shows where the increase in gas led to maximum surface warming (courtesy P. Foster).

- How does the transport of short-lived halogenated species and their decay products to the stratosphere influence the photochemistry and budget of Bromine (BrO) in the TTL?
- What is the potential of the halogen oxides to directly destroy UT/LS ozone?
- How do cirrus clouds influence chemistry in the UTLS?

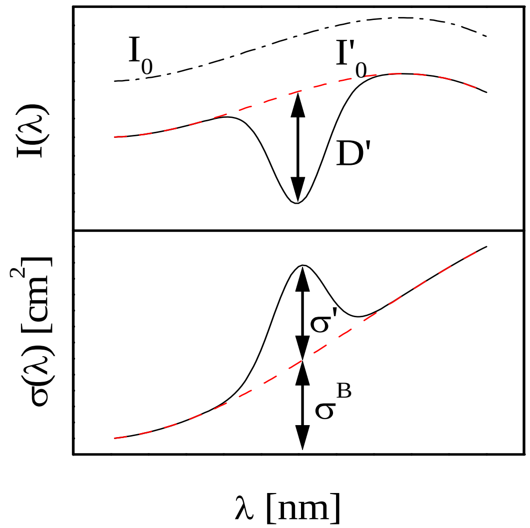
Approach

Limb Scanner

(2° off flight direction)



Differential Optical Absorption Spectroscopy



$$I(\lambda) = I_0(\lambda) \cdot e^{-\sigma(\lambda) \cdot SCD}$$

$$D' = \ln\left(-\frac{I(\lambda)}{I_0'(\lambda)}\right)$$

Observation of path-integrated trace gas absorptions using DOAS:

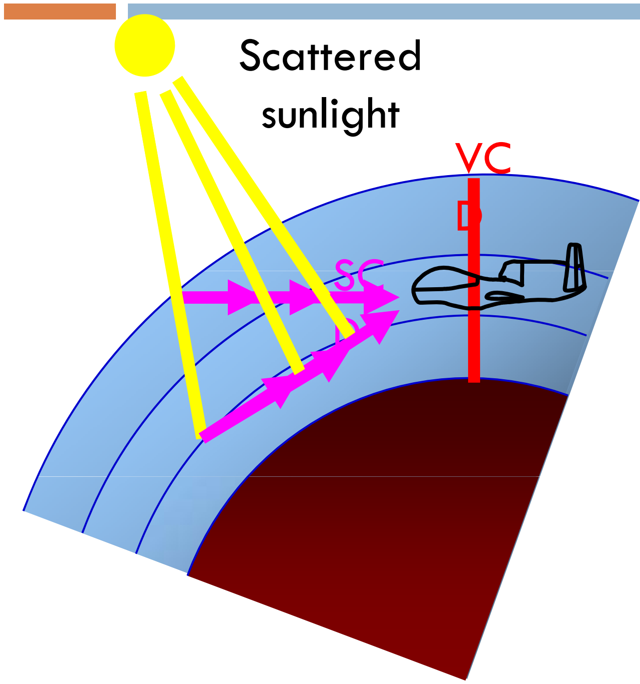
Slant Column Densities:

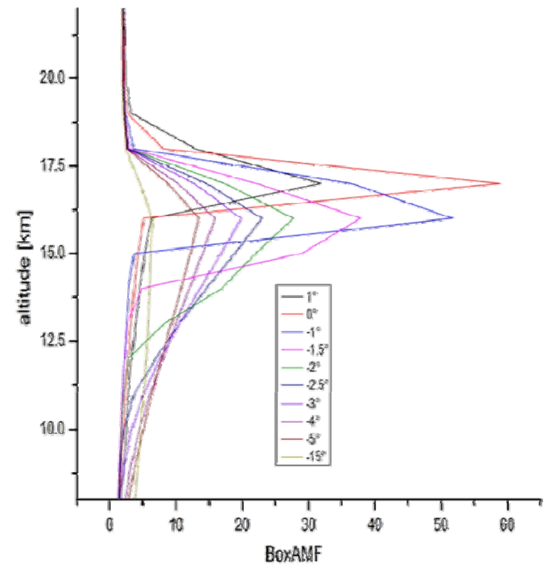
$$SCD = \int_0^s c(s)\,ds$$

$$SCD_i = \frac{\ln(D')}{\sigma_i'(\lambda)}$$

Removal of solar Fraunhofer lines: $DSCD = SCD_{Elev.Angle} - SCD_{solar}$

From SCDs to trace gas concentration





- The Airmass factor:

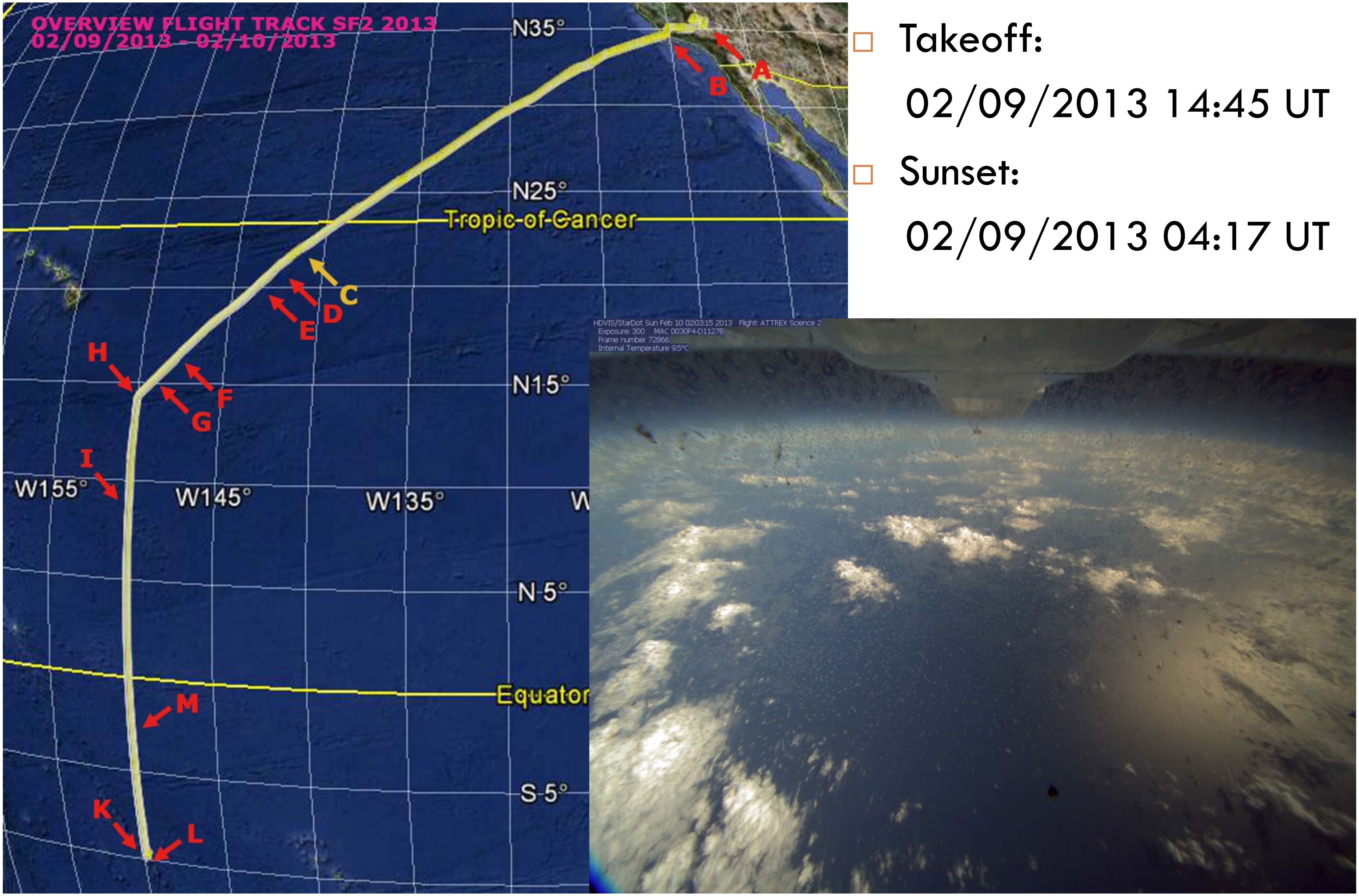
$$AMF = \frac{SCD(\lambda, Elev.angle)}{VCD} = \frac{\int_s c(s)ds}{\int_h c(h)dh}$$

- Box Airmass Factor (BAMF):

$$SCD(El.Ang.) = \sum BAMF(El.Ang.) * c(h) * dh$$

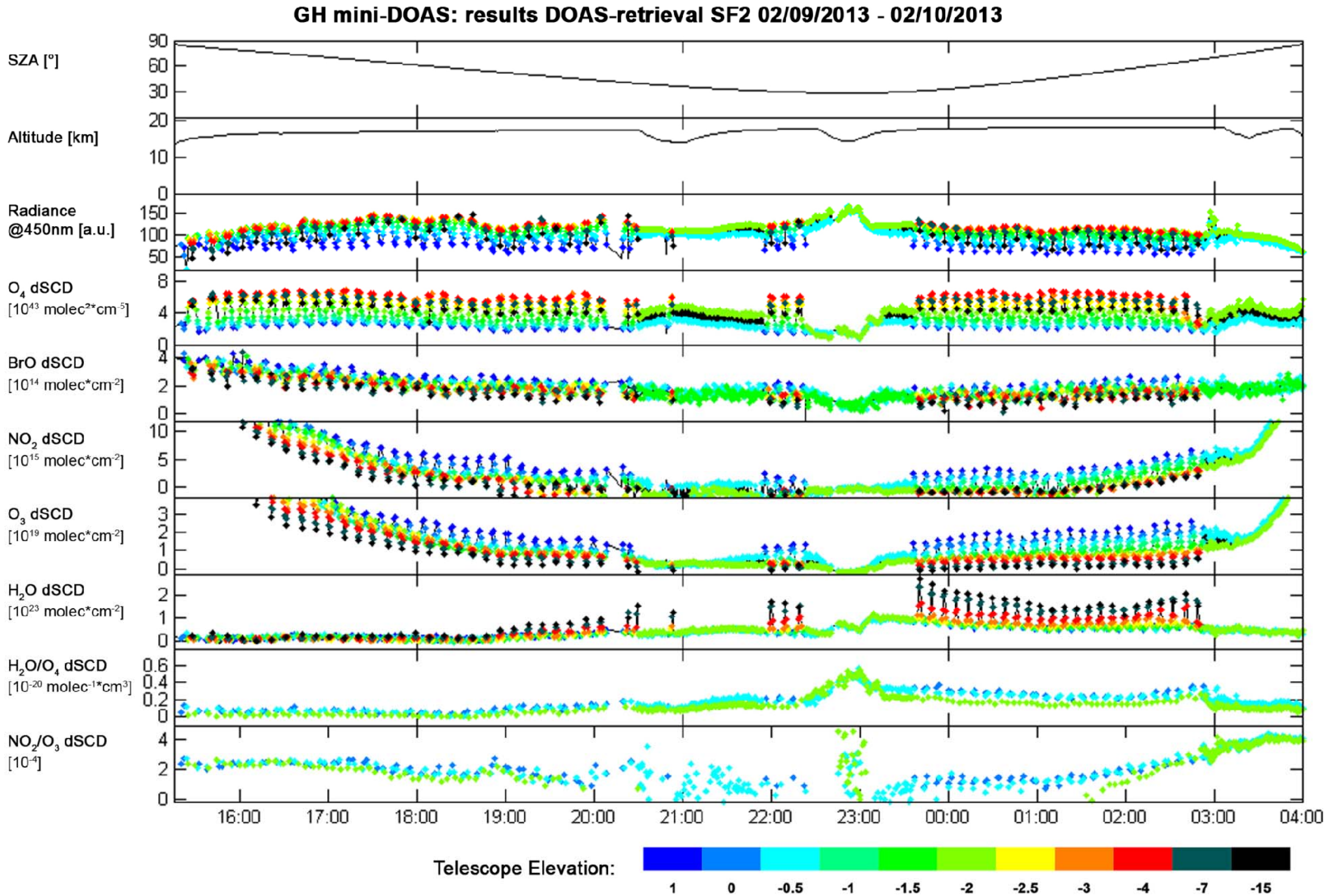
- BAMFs derived via radiative transfer calculations

Science Flight 2, 2013



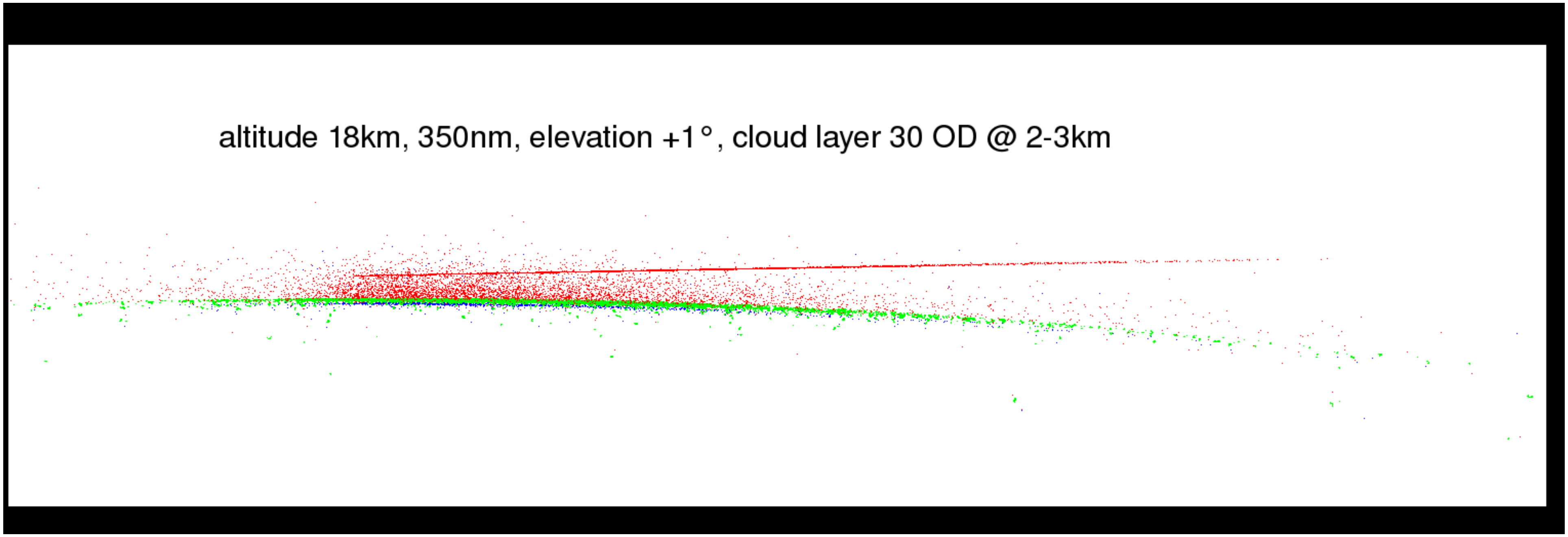
- Takeoff:
  02/09/2013 14:45 UT
- Sunset:
  02/09/2013 04:17 UT

Science Flight 2



Radiative transfer

Vertical contribution function at 350 nm



Red: Rayleigh scattering events
Green: Mie scattering events
Blue: Ground reflection

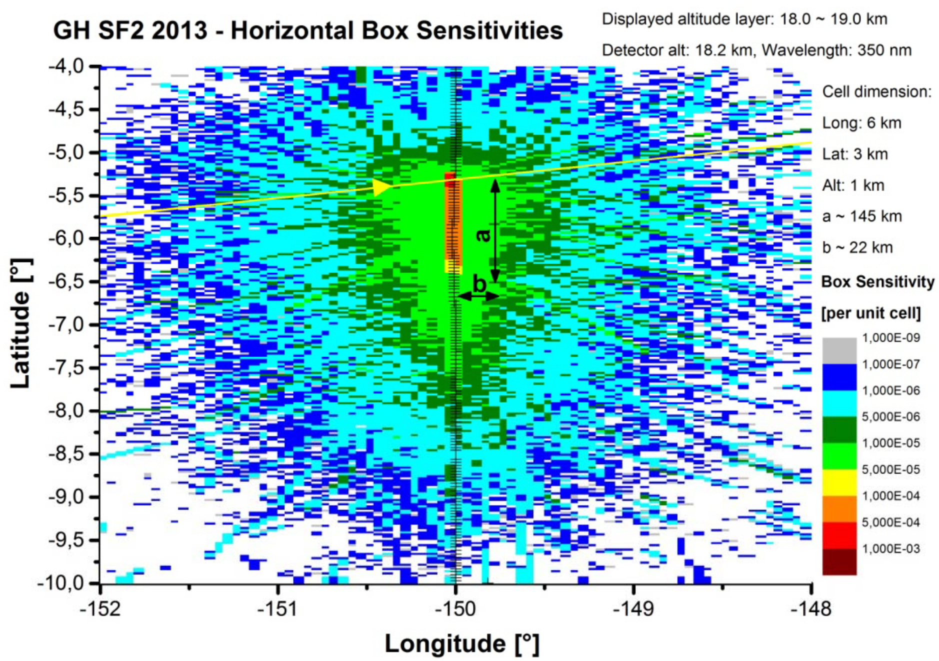
(Tim Deutschmann, PhD thesis, 2013)

Radiative Transfer Tools: McArtim & VLIDORT

Data interpretation: Flight altitude m.r.

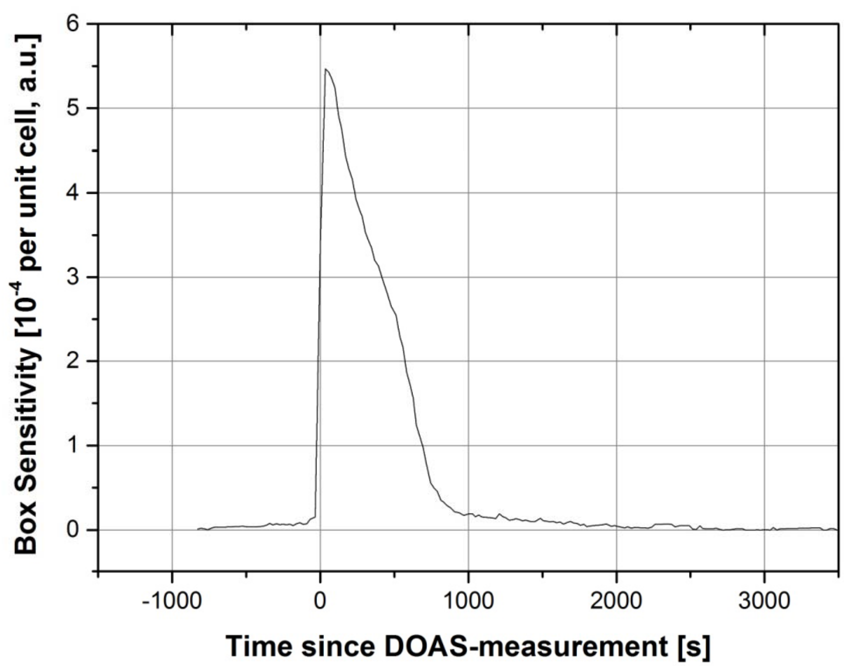
- Focus on limb (0°) and -0.5° elevation scans
- Determine the distance over which the DOAS averages
- Compare mini-DOAS O_3 SCD's with averaged NOAA in-situ O_3
- Radiative transfer calculations using SSFR albedos to determine the BAMFs & sensitivity for O_3, BrO, and NO_2
- Derive 'absorption path-length' proxy
- Calculate BrO, NO_2, and H_2O using DOAS SCD's and the 'absorption path-length' proxy

RT considerations for limb in UV



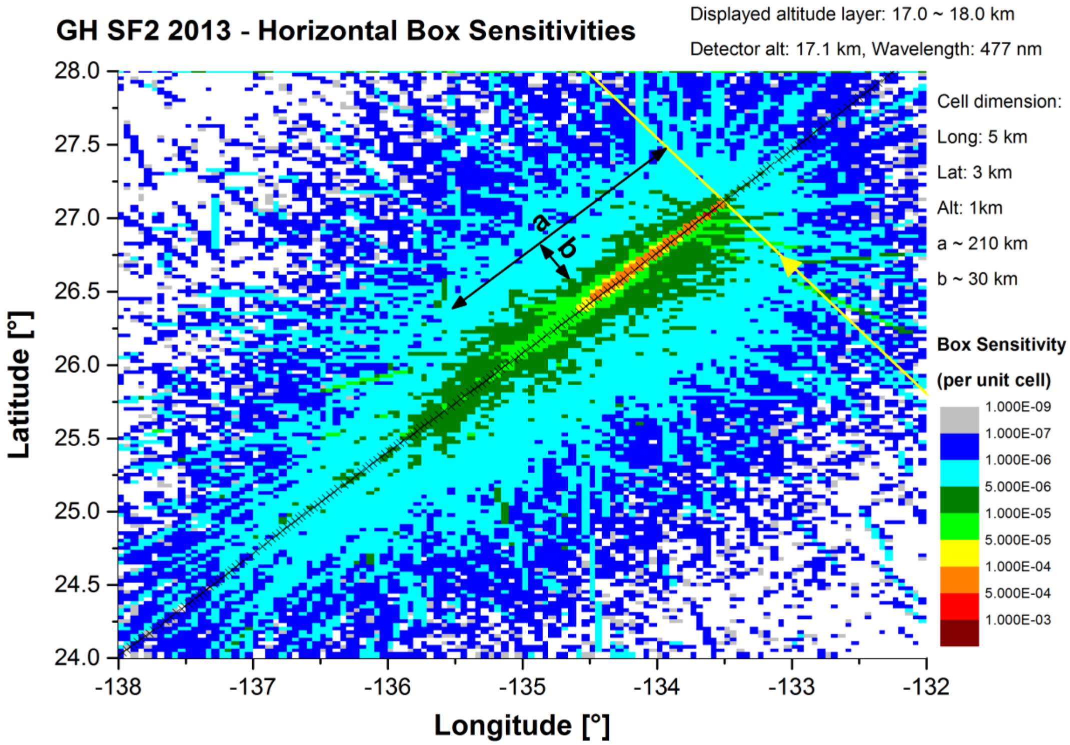
Spatial sensitivity of limb direction:

a = 145 km, b = 22 km



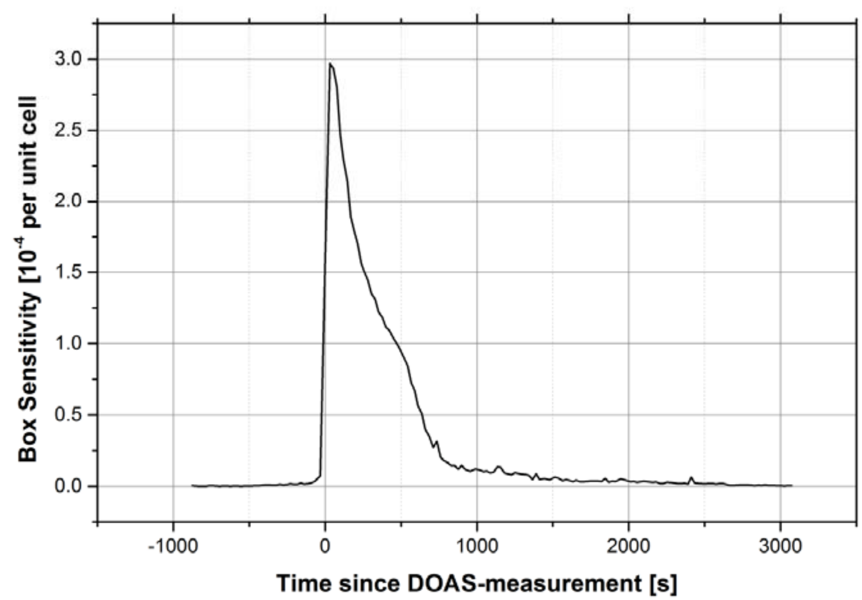
Convolution function of in-situ to path-integrated data

RT considerations for limb in visible



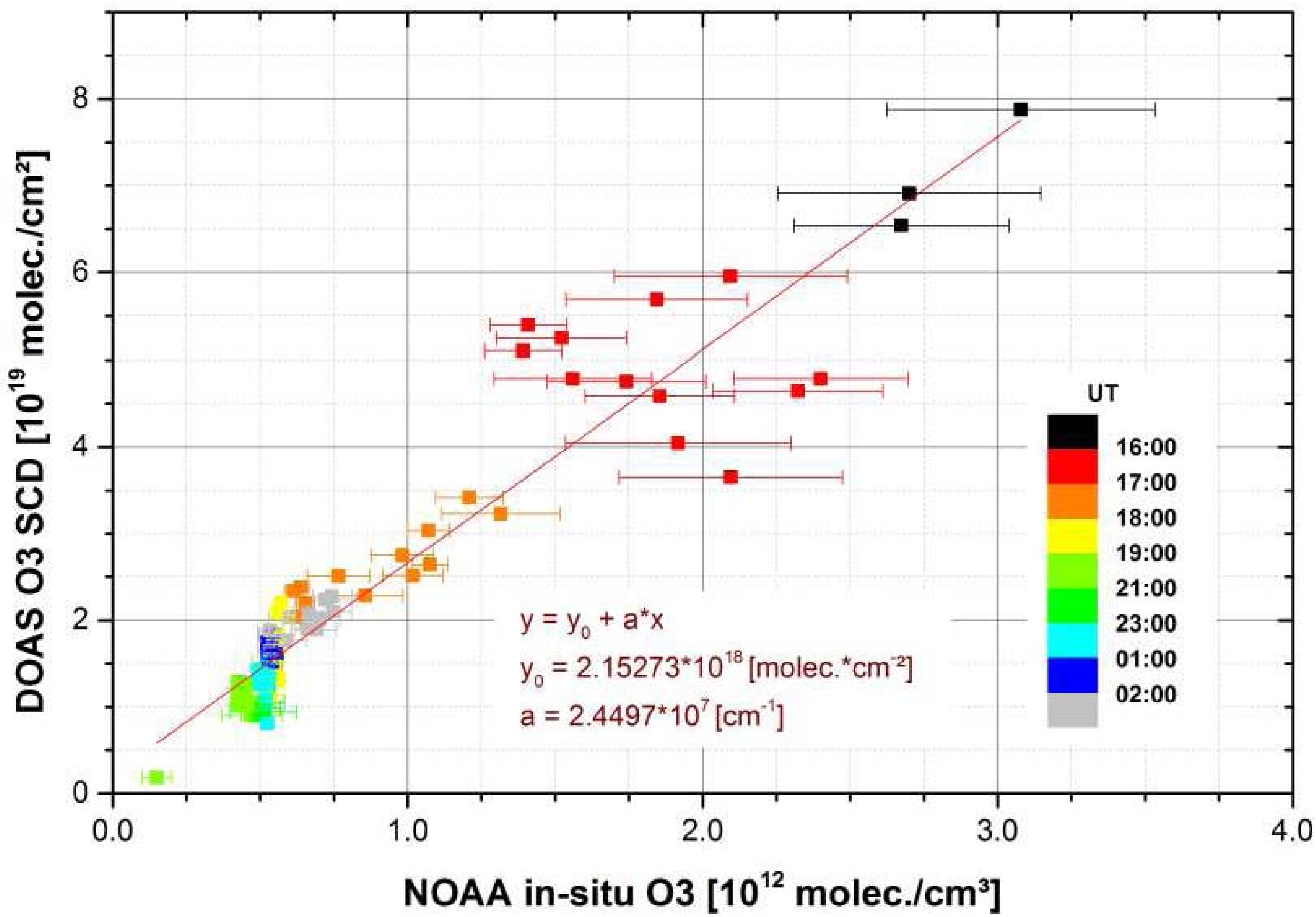
Spatial sensitivity of limb direction:

a = 188 km, b = 22 km



Convolution function of in-situ to path-integrated data

Convoluted DOAS vs in-situ O_3 in Vis

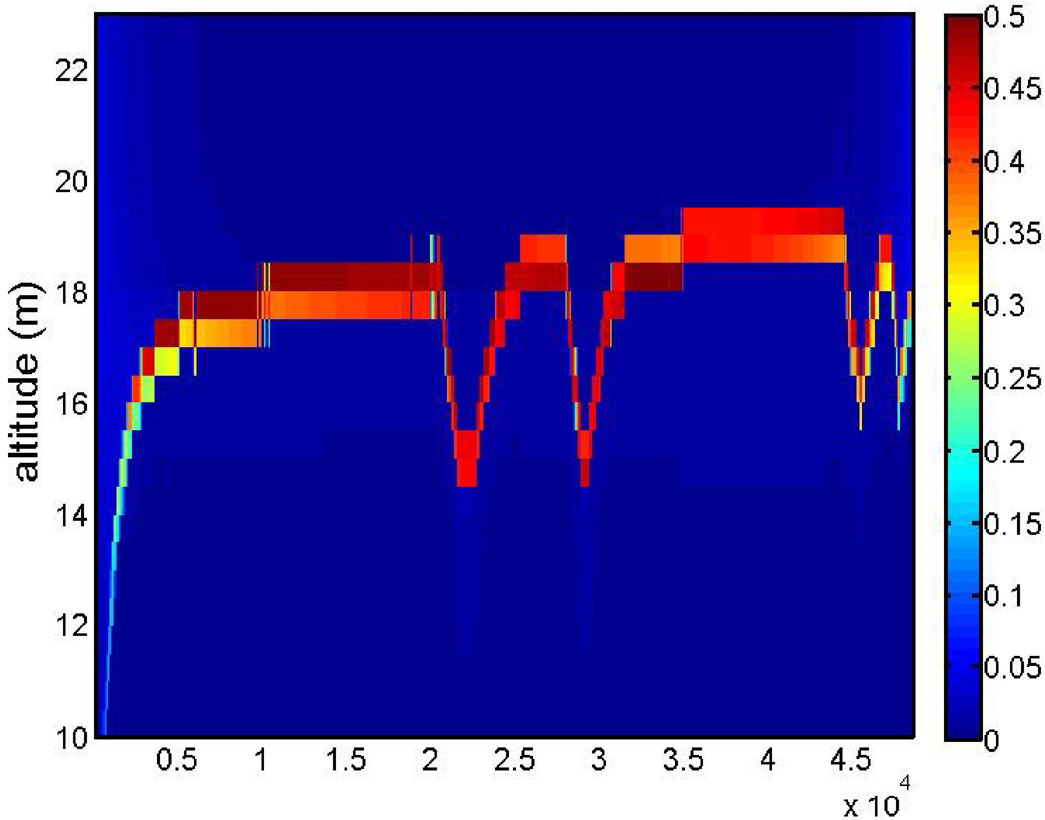


Differences due to RT effects!

Altitude sensitivity

Contribution of height intervals to BrO DSCD signal

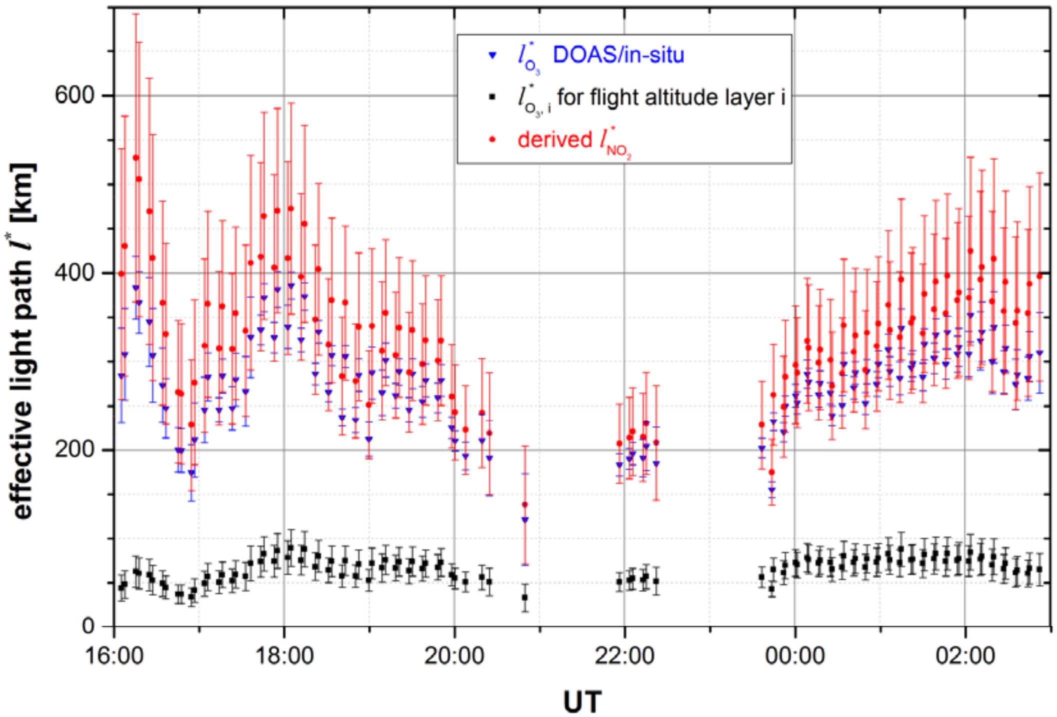
BrO sensitivity for -0.5° viewing angle SF2



- Sensitivity depends on:
  - Viewing direction
  - Trace gas profile
  - RT parameters (aerosol, albedo, etc.)
- Limb (0 °) and -0.5 ° viewing elevations can be used for direct conversion.

90% Sensitivity in a 1km layer around the GH, assuming no tropospheric BrO.

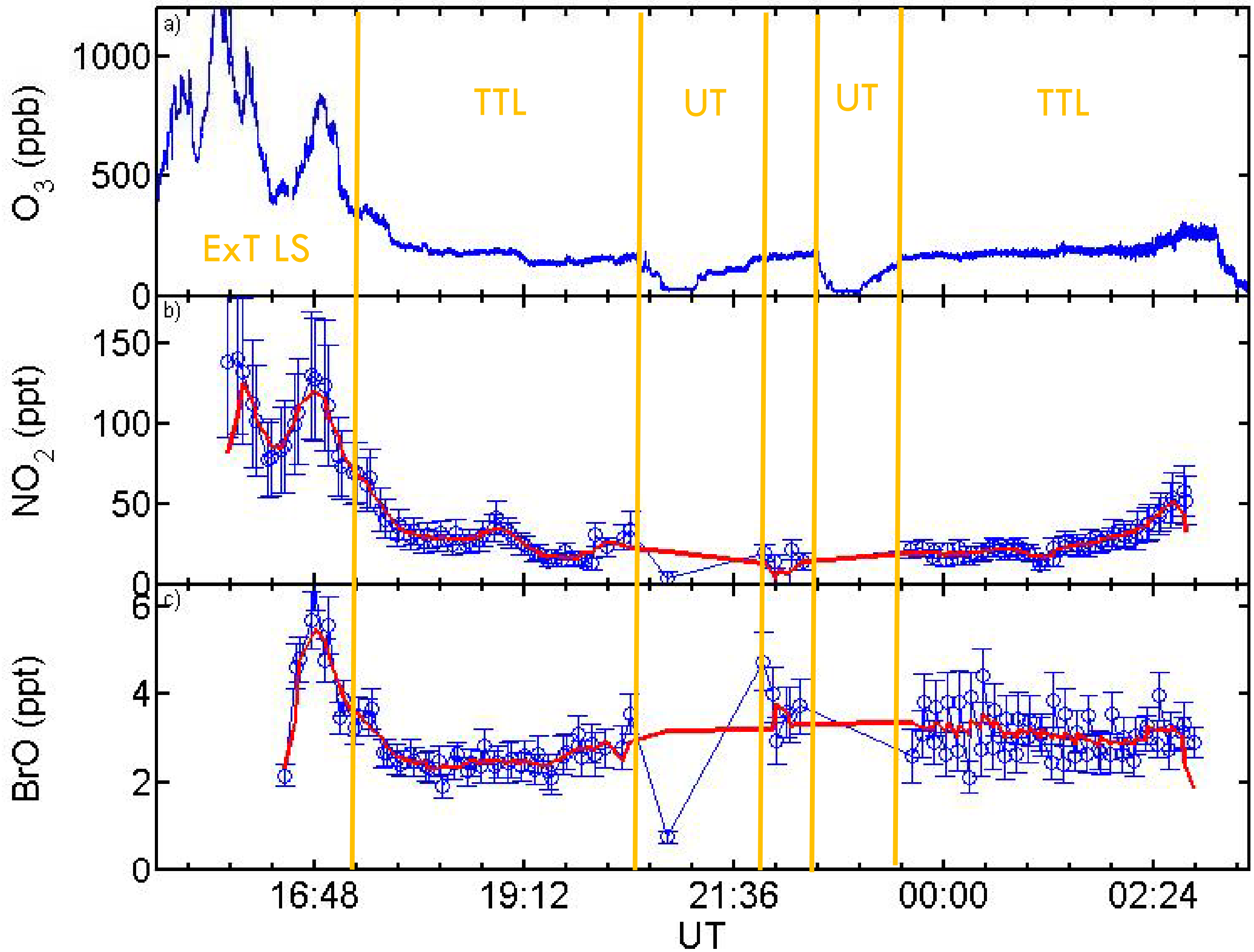
Pathlength from RT calc. and in-situ O_3 in Vis



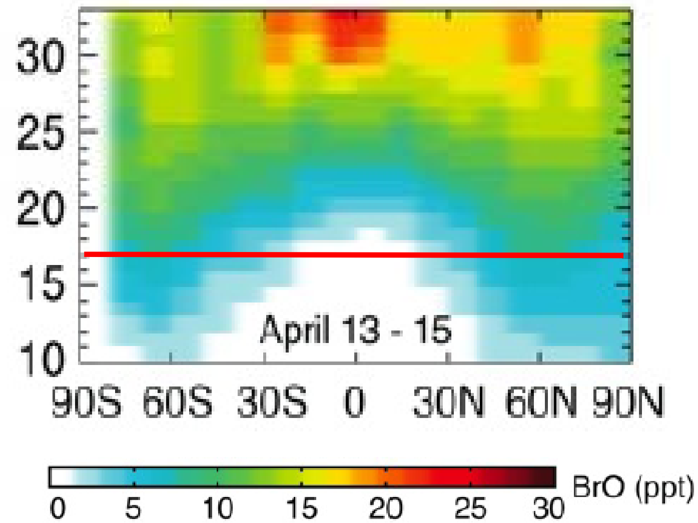
- Use in-situ and DSCD O_3 to determine absorption path-length l^*_{O3}
- Use RT calculation to relate l^*_{O3} to l^*_{NO2} and l^*_{BrO}
- Determine $[NO_2]$ and $[BrO]$ from NO_2 and BrO DSCD

$$[NO_2] = \frac{S_{NO_2}}{l^\star_{NO_2}} = \frac{S_{NO_2}}{S_{O_3}} \cdot [O_3] \cdot \frac{l^\star_{O_3}}{l^\star_{NO_2}} = \frac{\alpha_{NO_2}}{\alpha_{O_3}} \cdot \frac{S_{NO_2}}{l^\star_{O_3}}$$

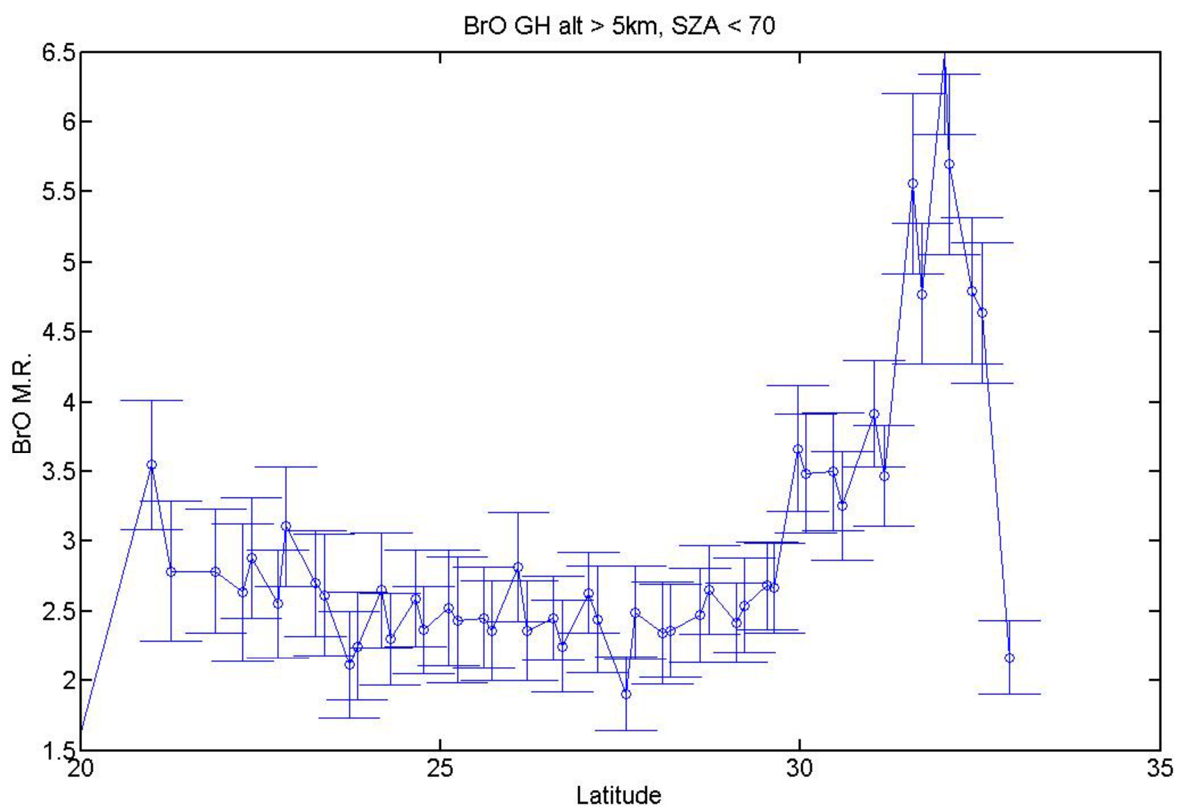
O_3 NO_2 and BrO at flight altitude SF2 2013



Comparison with literature data

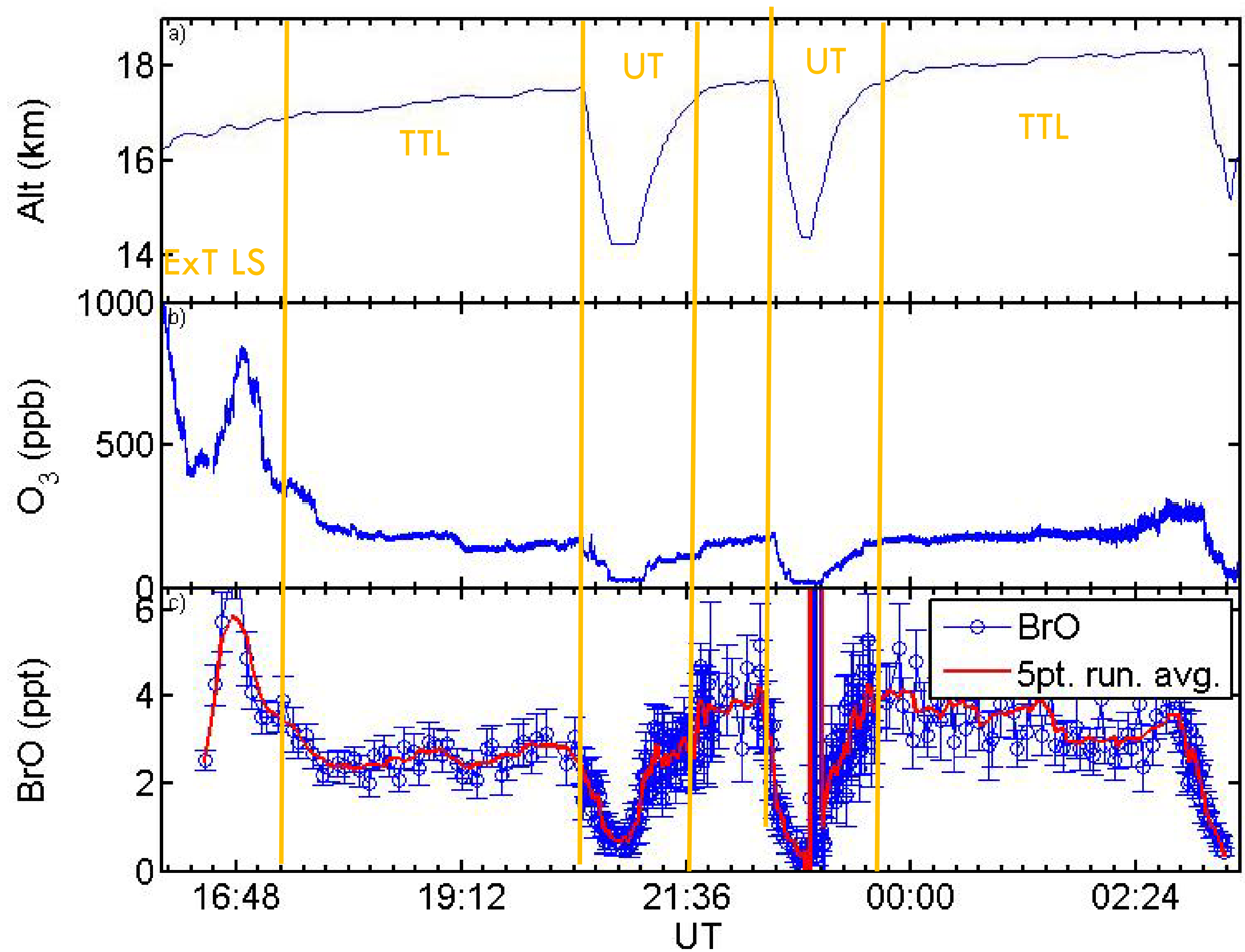


Parella et al, 2013



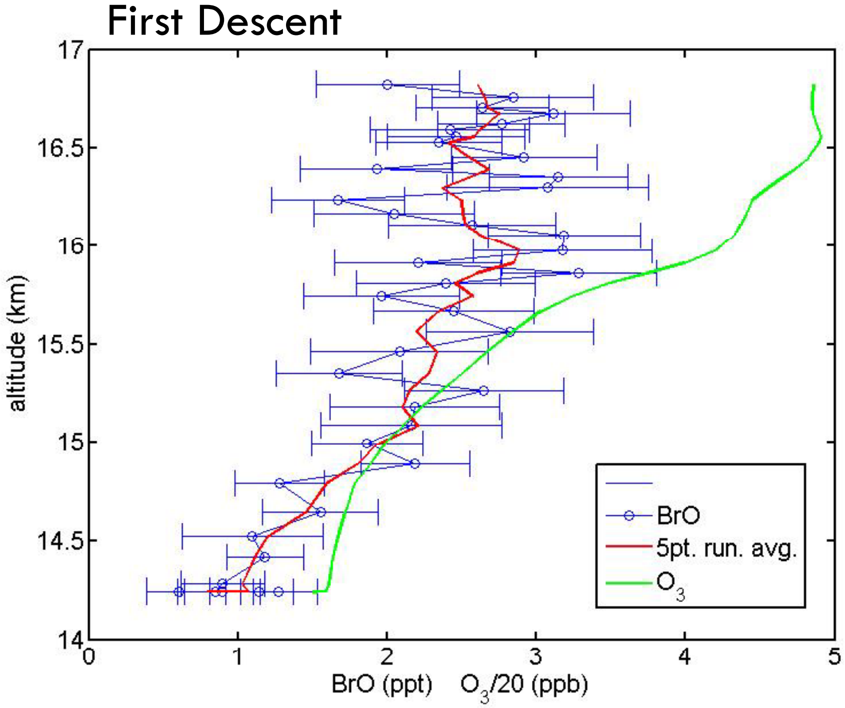
Expected latitudinal distribution seem to be captured well

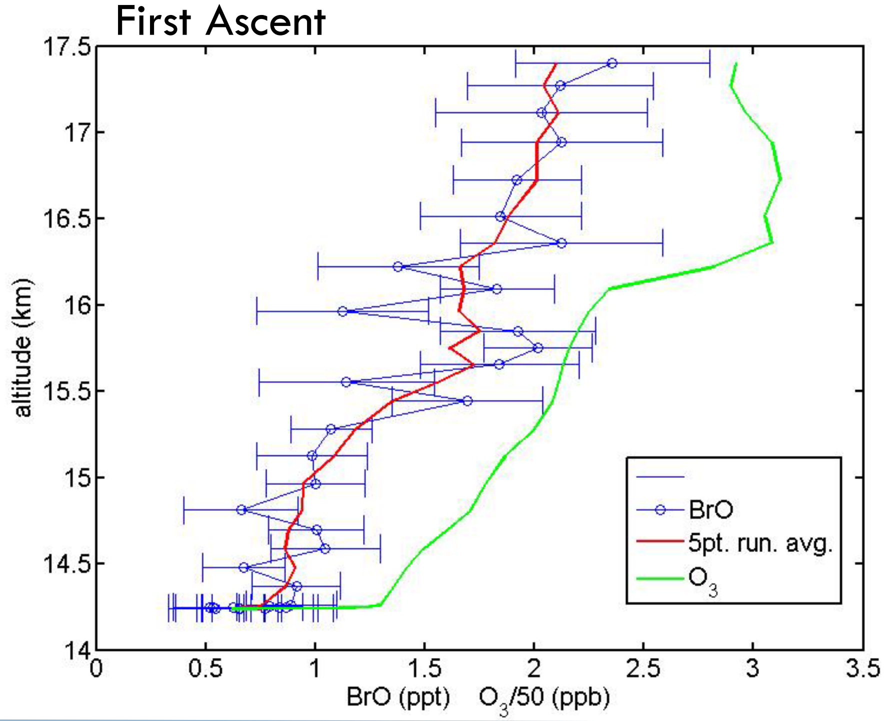
BrO in SF2 (-0.5°)

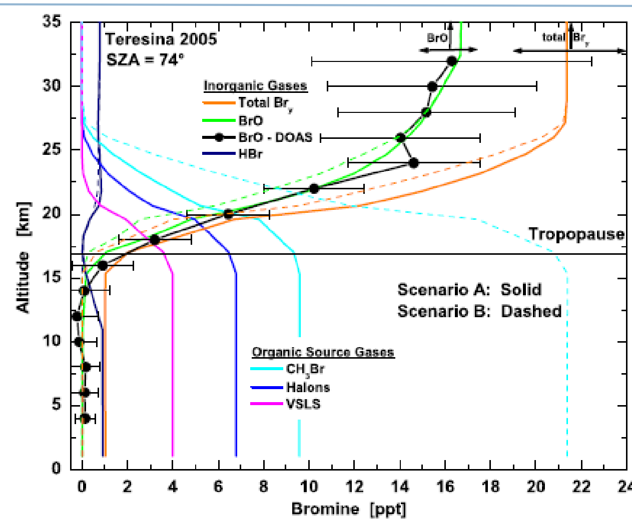


a)
Alt (km)
18
16
14
UT
UT
TTL
TTL
ExT LS
b)
O_3 (ppb)
1000
500
0
c)
BrO (ppt)
6
4
2
0
BrO
5pt. run. avg.
16:48
19:12
21:36
00:00
02:24
UT

Vertical BrO Profiles from SF2

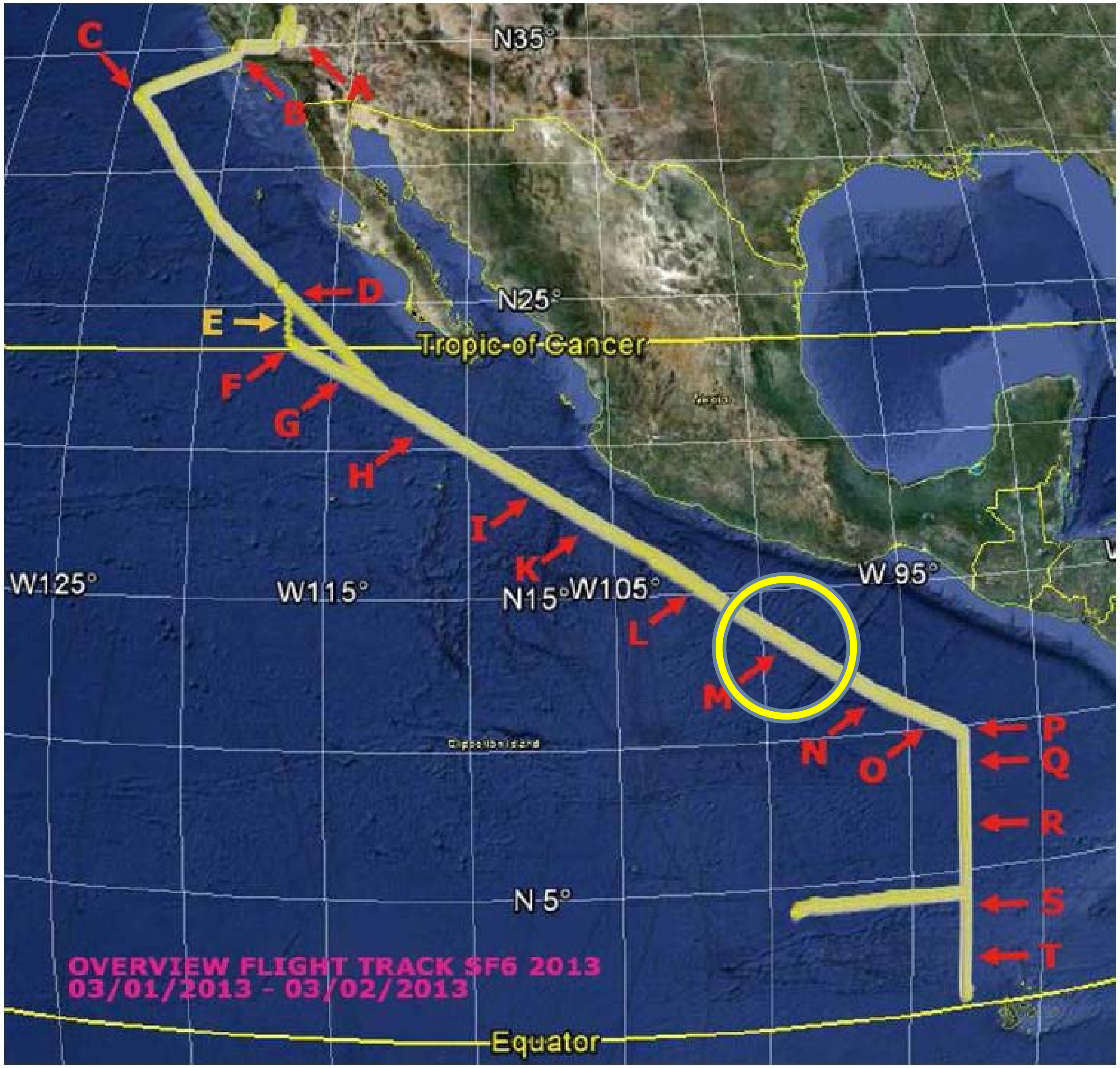




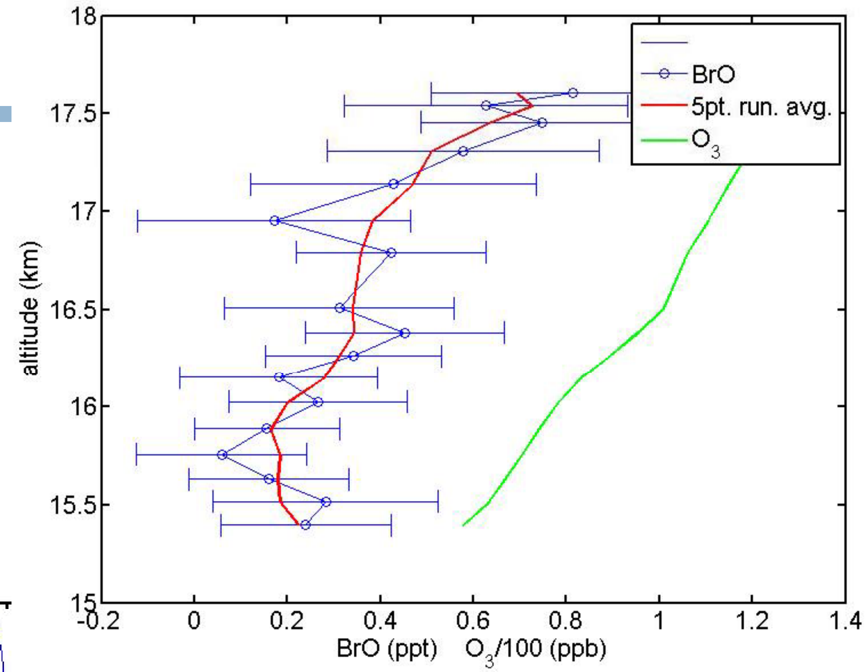


Dorf et al, 2008

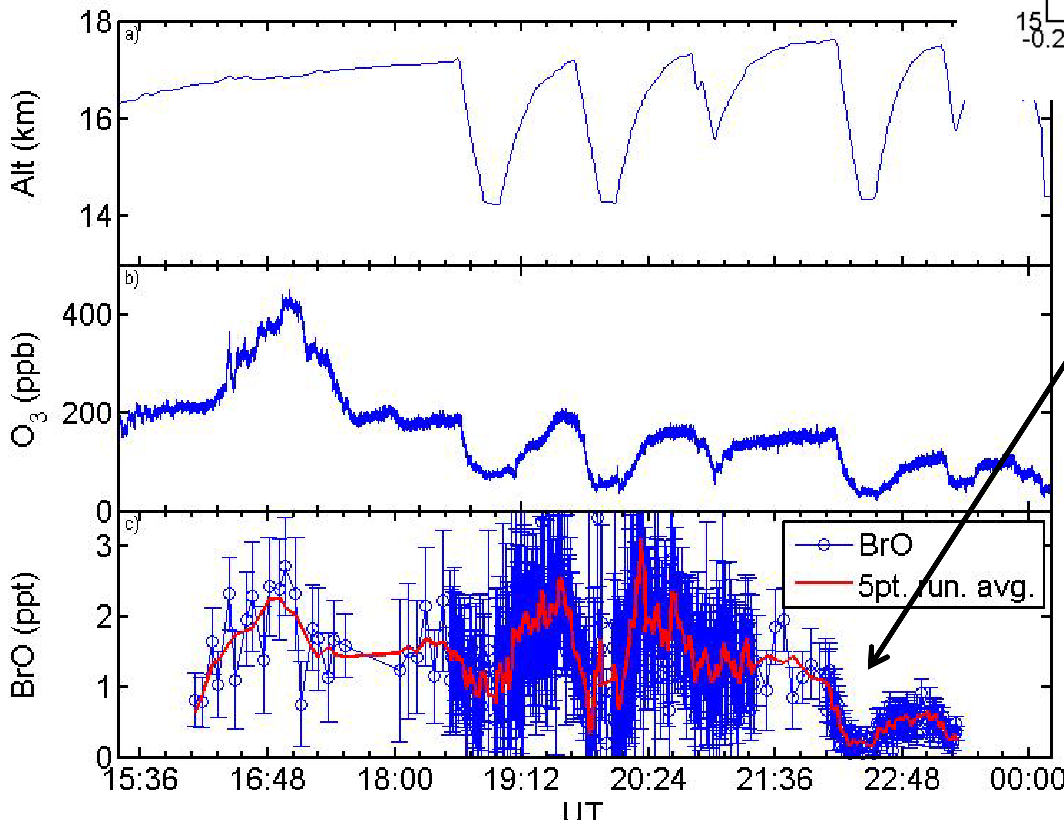
Science Flight 6, 2013



BrO in SF6

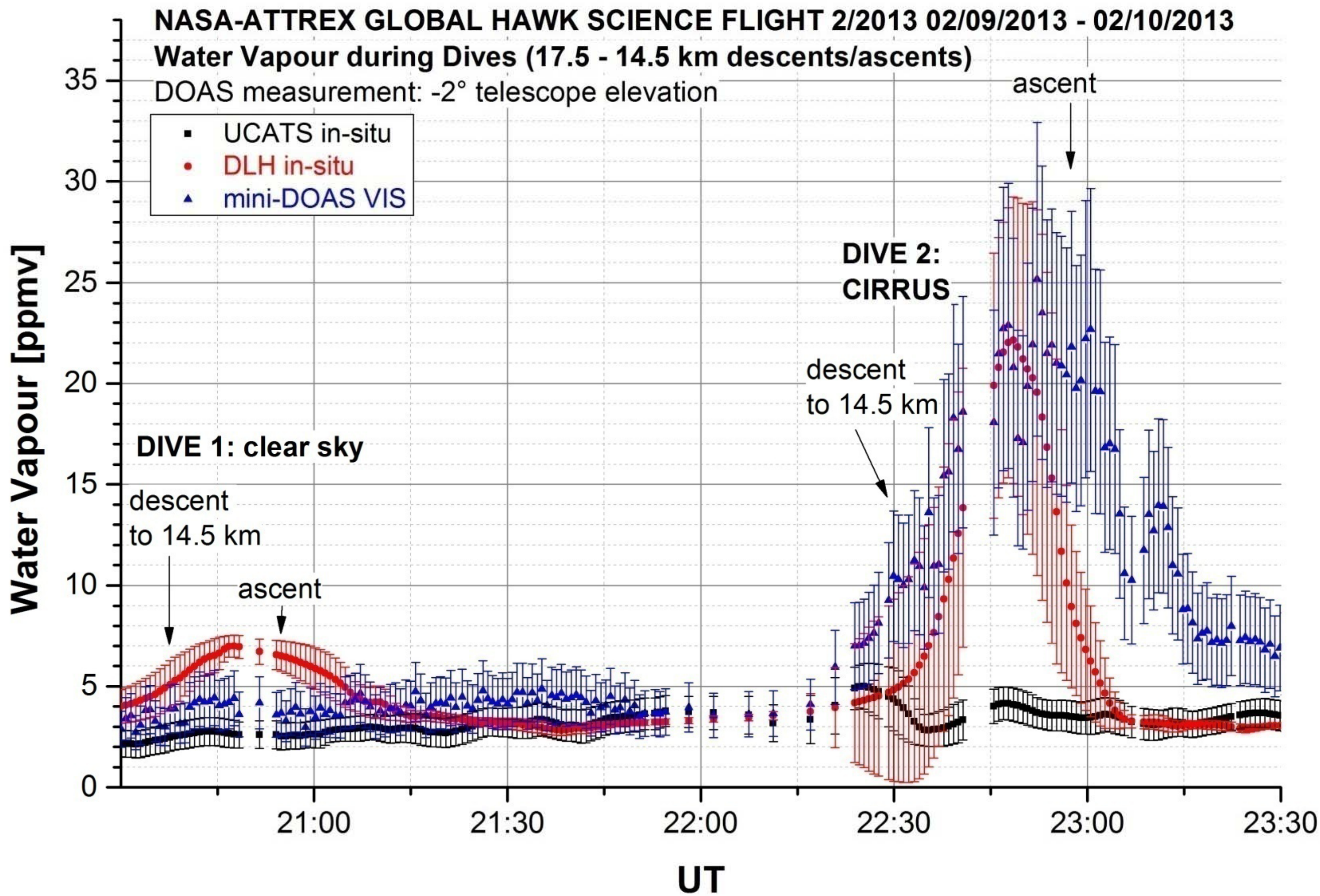


altitude (km)
BrO
5pt. run. avg.
O_3
BrO (ppt) O_3/100 (ppb)



Alt (km)
O_3 (ppb)
BrO (ppt)
BrO
5pt. run. avg.
15:36
16:48
18:00
19:12
20:24
21:36
22:48
00:00
UT

Preliminary H_2O vapor measurements

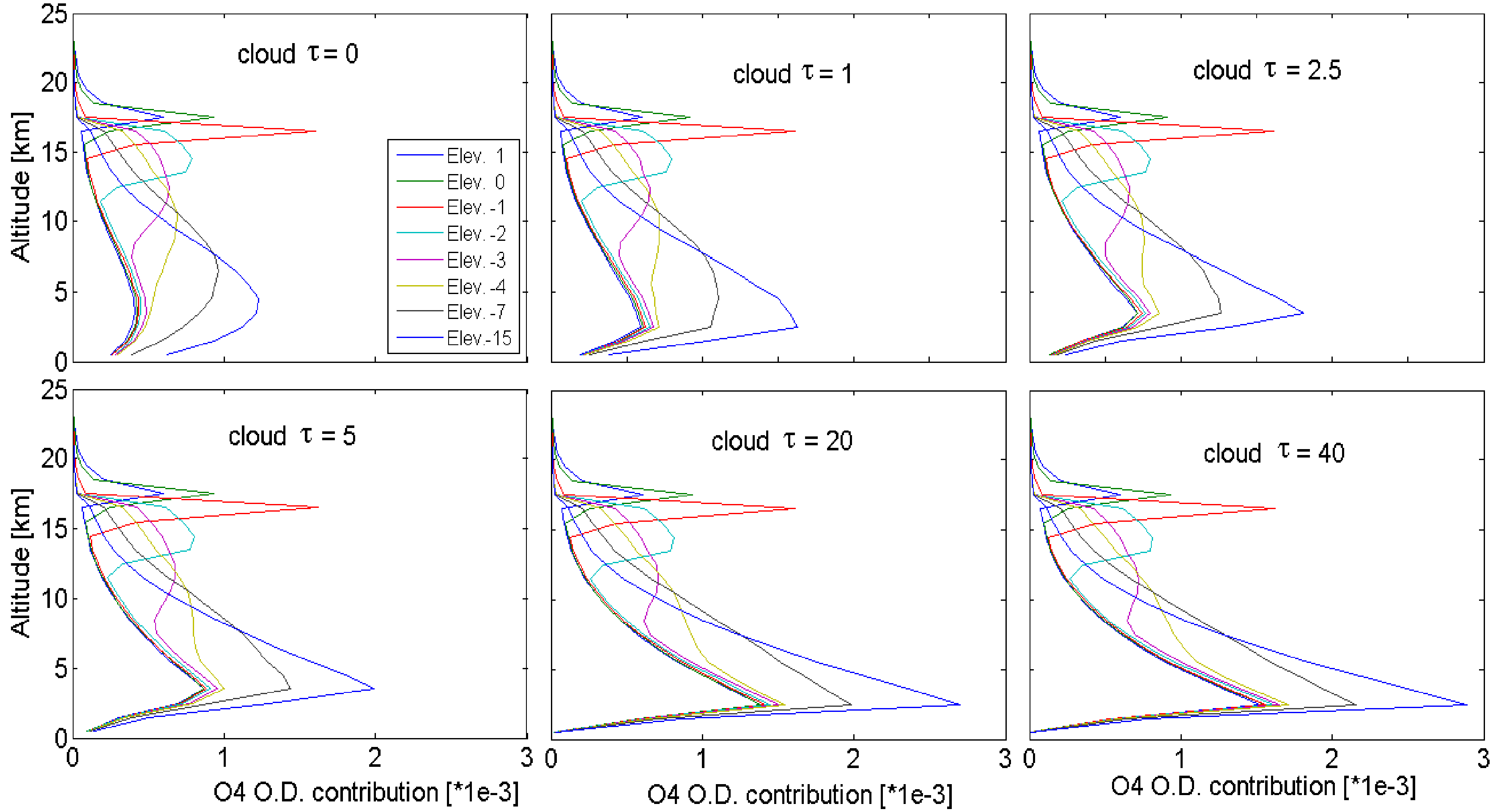


Vertical trace gas profile retrievals

- Radiative transfer models in combination with optimal estimation technique.
  - Two RT models: McArtim (Monte Carlo) and VLIDORT (Linearized)
  - Two step optimal estimation according to Rodgers:
    - Step 1: Derivation of aerosol extinction profiles using relative radiances and O_2 dimer, O_4, absorptions.
    - Step 2: Derivation of trace gas profiles based on derived aerosol profiles
- Initialization of models based on albedo / cloudiness
- Challenges:
  - Influence of trace gases in the troposphere needs to be determined accurately (in particular for O_4 and H_2O.
  - BRDF over oceans was not implemented in RT models.
  - Stratus clouds considerably change the RT.
  - Cirrus are currently not represented well in models.
  - Unknown unknowns……

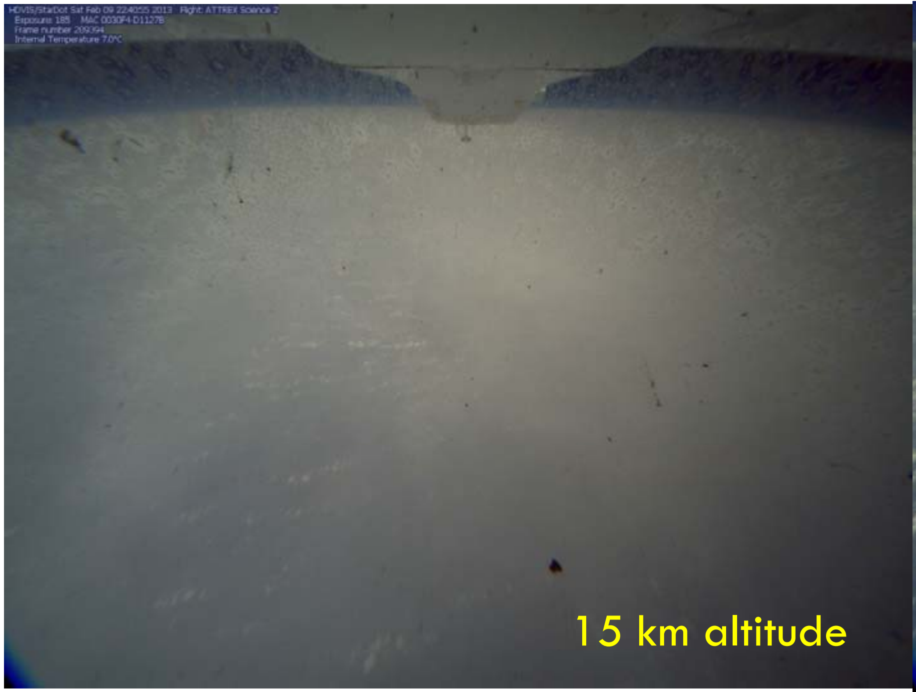
Impact of statocumulus clouds

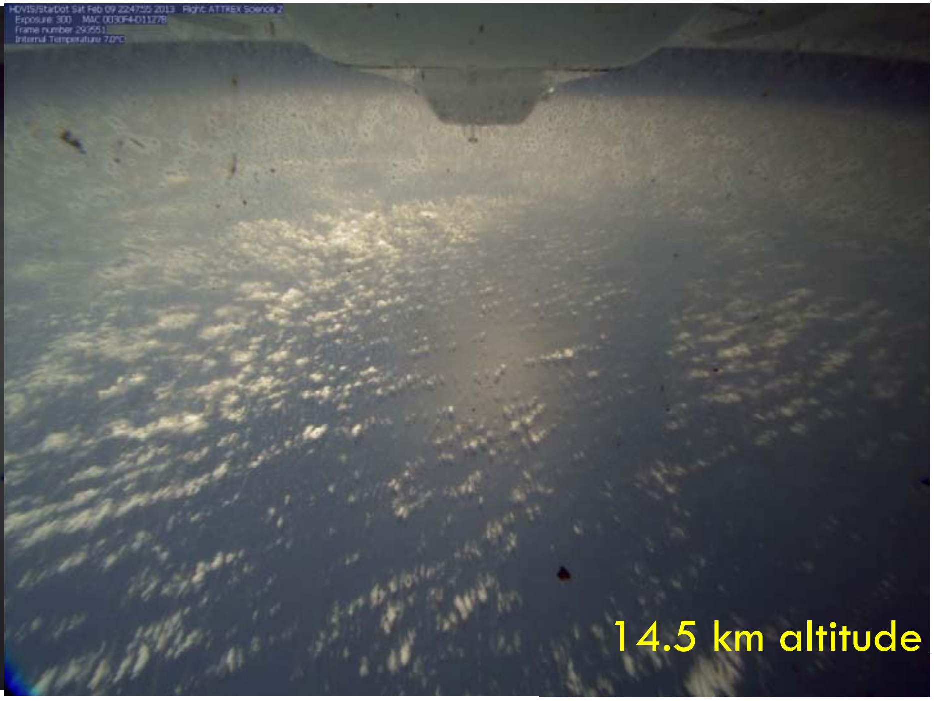
O_4 Contribution function at 361nm incl. aerosol extinctions and MSC clouds at 2km

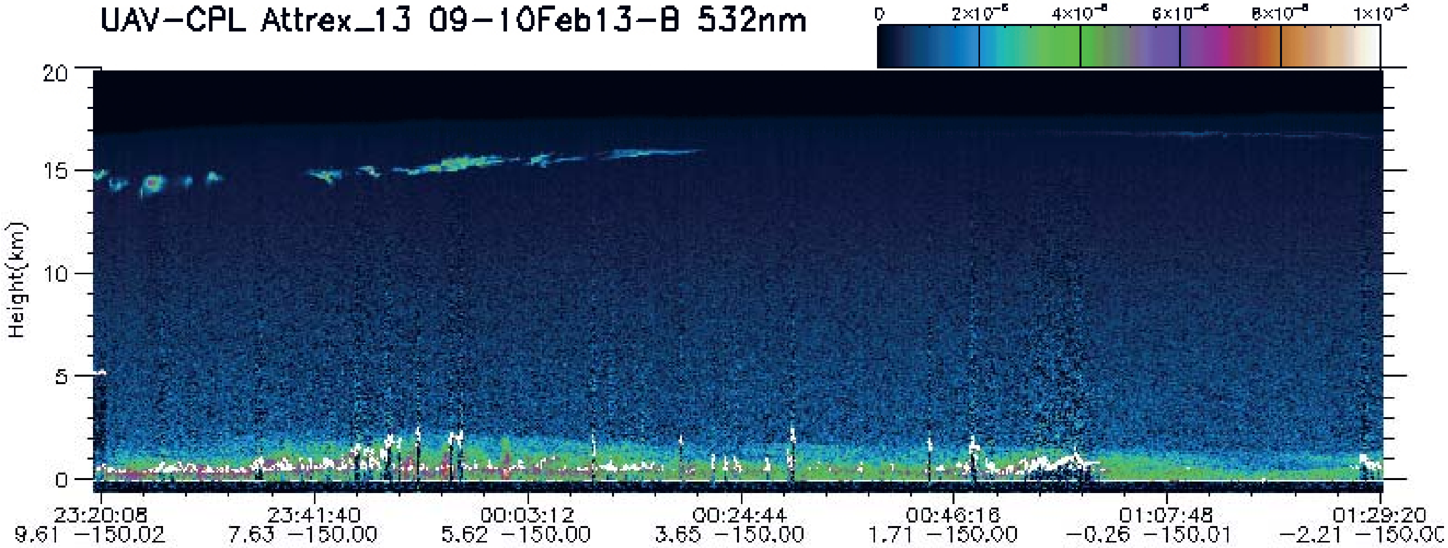


Improvement of model, retrieval, and data assimilation needed

Cirrus Clouds ……



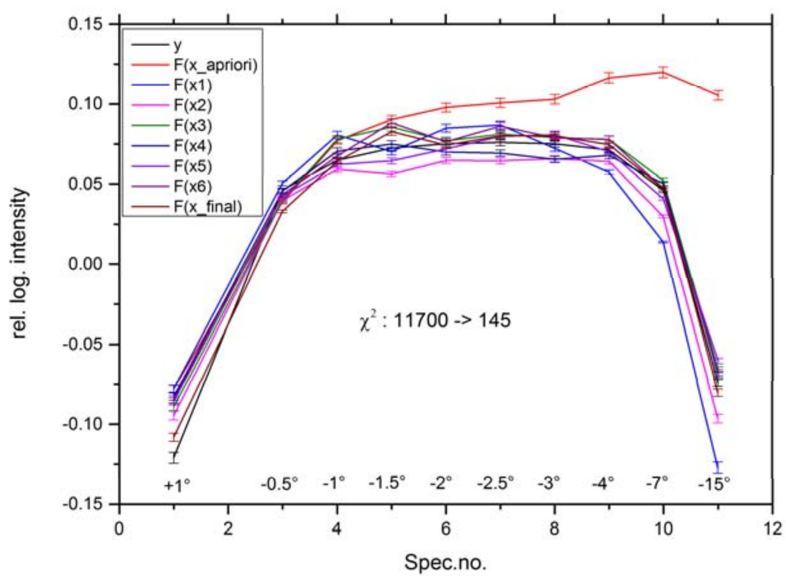




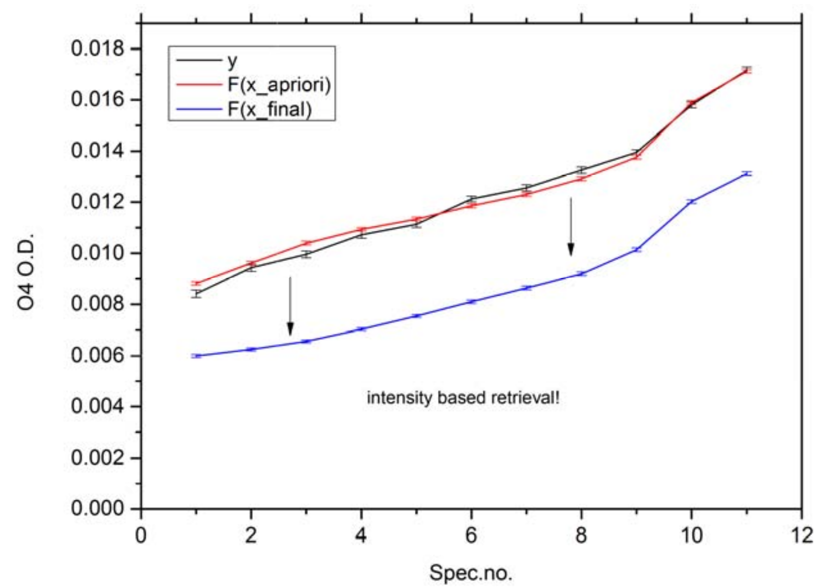
LIDAR data and Images courtesy of NASA

Status

- RT models and retrievals are operational.
- Intensive "theoretical" testing has been performed.
- But: Inconsistency in radiance and O_4 aerosol extinction retrieval!



Radiance retrieval results



Comparison of calculated O_4 optical density with observations

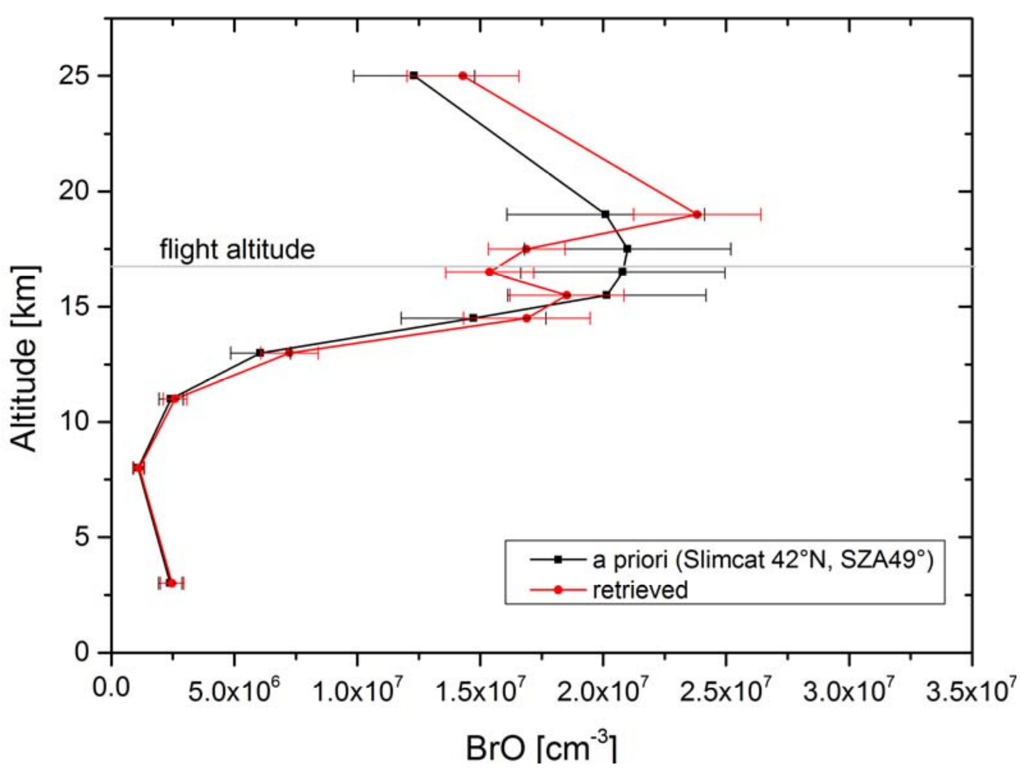
Status

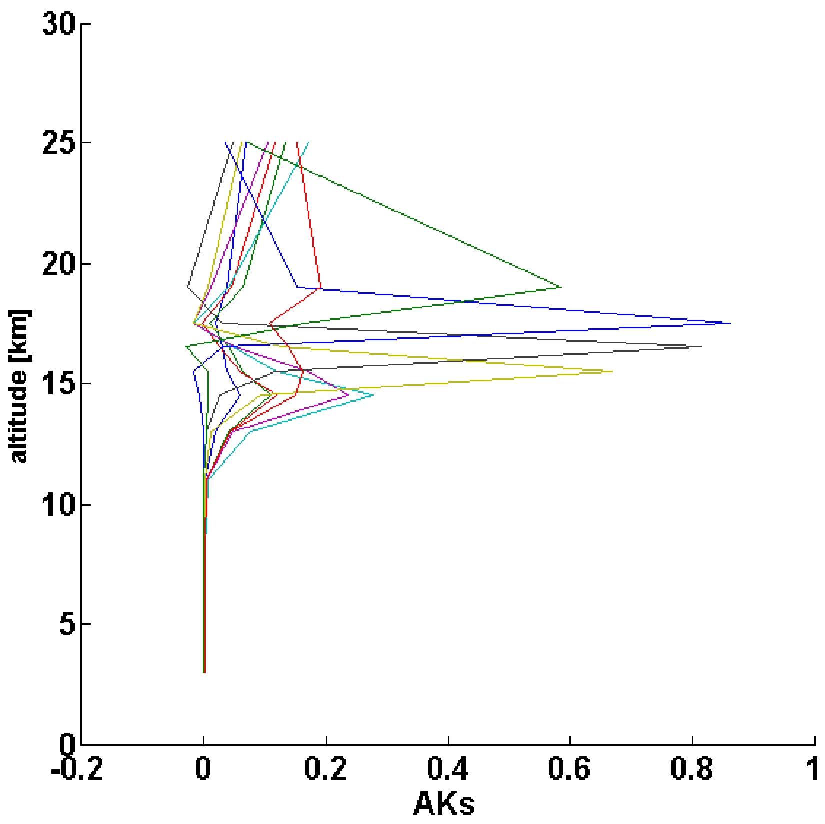
- Ideas to resolve dissagreement:
  - Model errors → comparison of two different models
  - Albedo vs. BRDF
  - Aerosol profiles
  - Impact of clouds → implementation of stratus clouds
  - *other ideas??*

- Discrepancy needs to be resolved to achieve an better accuracy of vertical concentration profiles

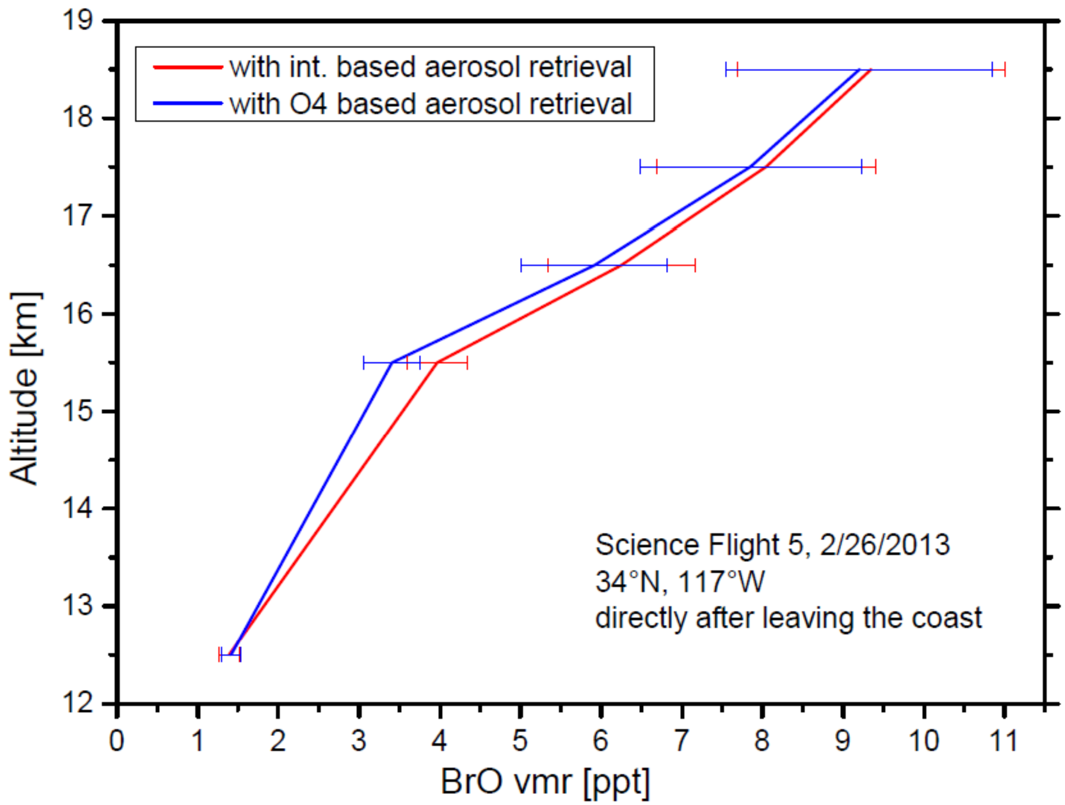
Preliminary BrO profile retrieval

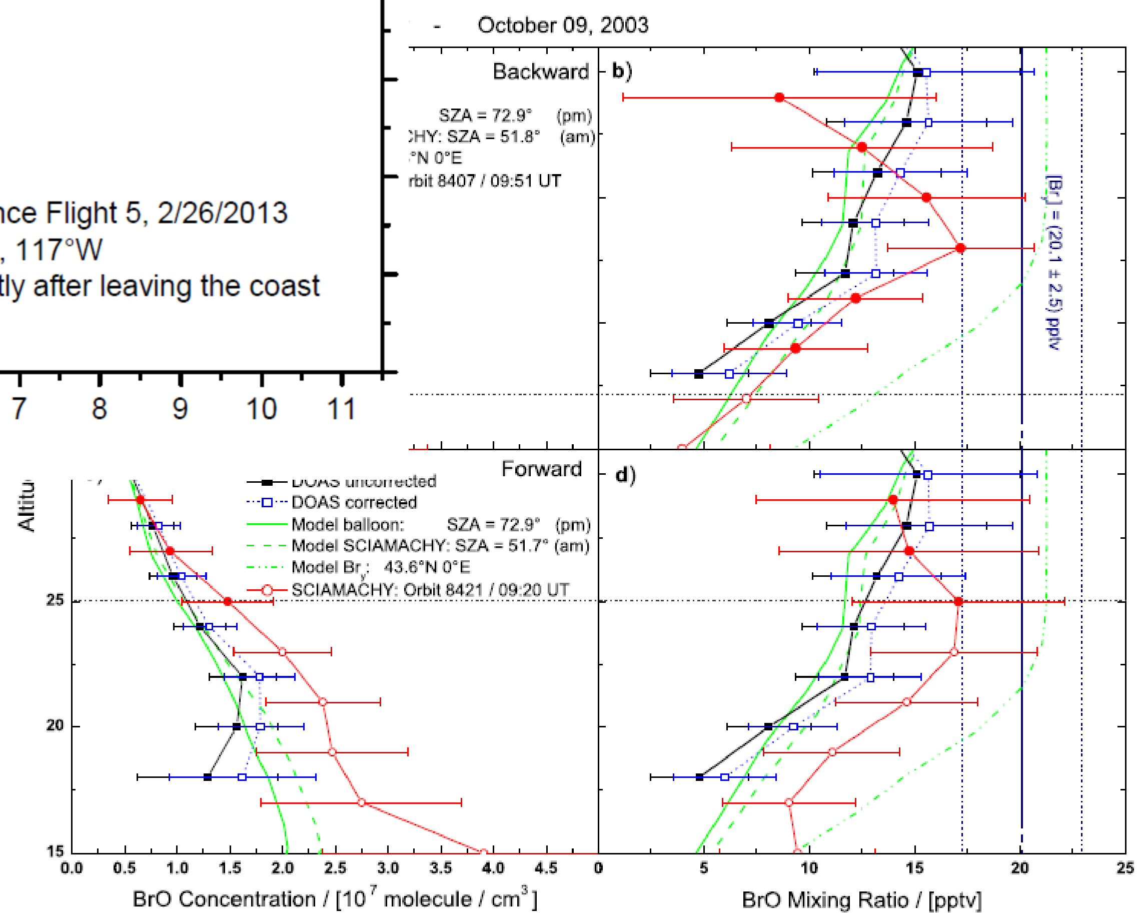
- SZA = 45°
- Aerosol profile from intensity and O_4 based retrieval using McArtim (optimal estimation with a priori constraint)
- A priori profile: SLIMCAT, 42°N, 1°W, for SZA=49°)





Preliminary BrO profile retrieval for clear sky





Dorf et al, 2006

Conclusions & Outlook

- Trace gas DSCD's have been retrieved successfully using a direct solar reference. Instrument performance in UV and Vis as good as, if not better, than expected.
  - DL(BrO)~0.5ppt and DL(NO_2)~5ppt at flight altitude
- In-flight altitude BrO, NO_2 and H_2O has been retrieved by scaling path-length with in-situ O_3.
  - Latitudinal profile looks like expected.
  - Vertical Profiles will allow analysis of bromine budget in TTL (see talk by K. Pfeilsticker….)
  - On what time-scale should we report the flight-altitude mixing ratios?
  - How do we make sure everyone understands the nature of the data, i.e. H & V averaging?
- Algorithms for concentration profile inversions (McArtim and VLIDORT) have been developed.
  - But need to resolve radiance / O_4 inconsistencies.
  - Finalize full aerosol and trace gas retrievals.
  - Better consideration of stratocumulus and cirrus clouds.

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