BrO in the Tropical and Subtropical UTLS

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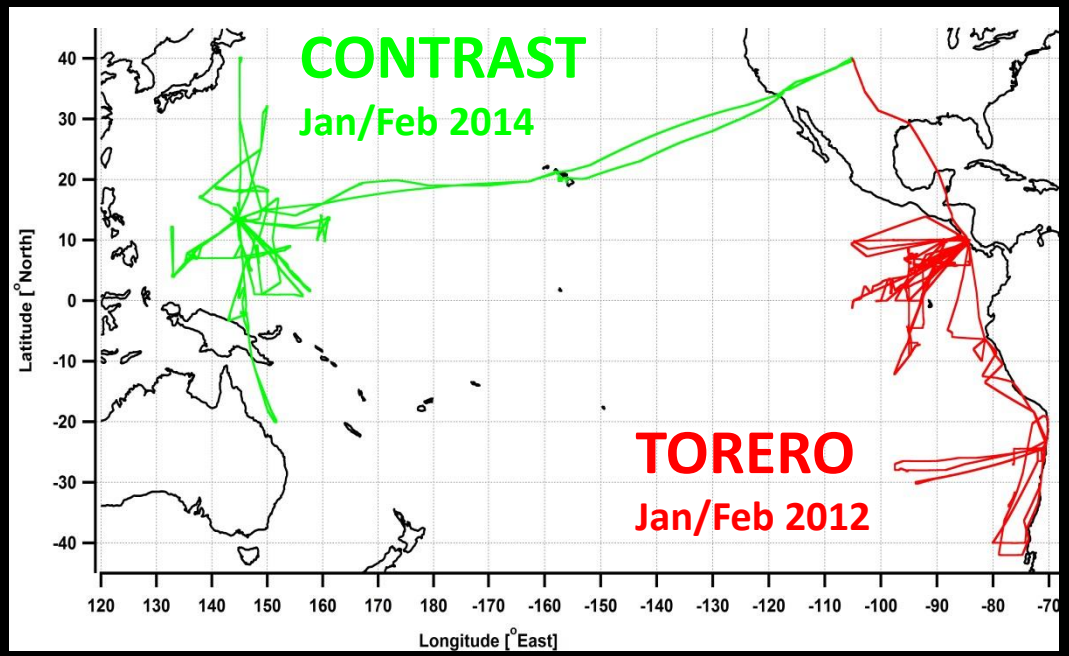
- Introduction BrO conundrum
- CU AMAX-DOAS (BrO, IO, NO_2, H_2O, O_4) & data status
- Robustness of NO_2 and BrO VMR conversion
- Comparison of VMRs from remote-sensing with in-situ and model data



CIRES
Cooperative Institute for Research in Environmental Sciences

University of Colorado Boulder

BrO overview: observations and models

Satellite: 1-3 x10^{13} molec cm^{-2}

(Chance et al., 1998; Wagner et al., 2001; Richter et al., 2002; Van Roozendael et al., 2002; Theys et al., 2011)

Ground : 0.2-3 x10^{13} molec cm^{-2}

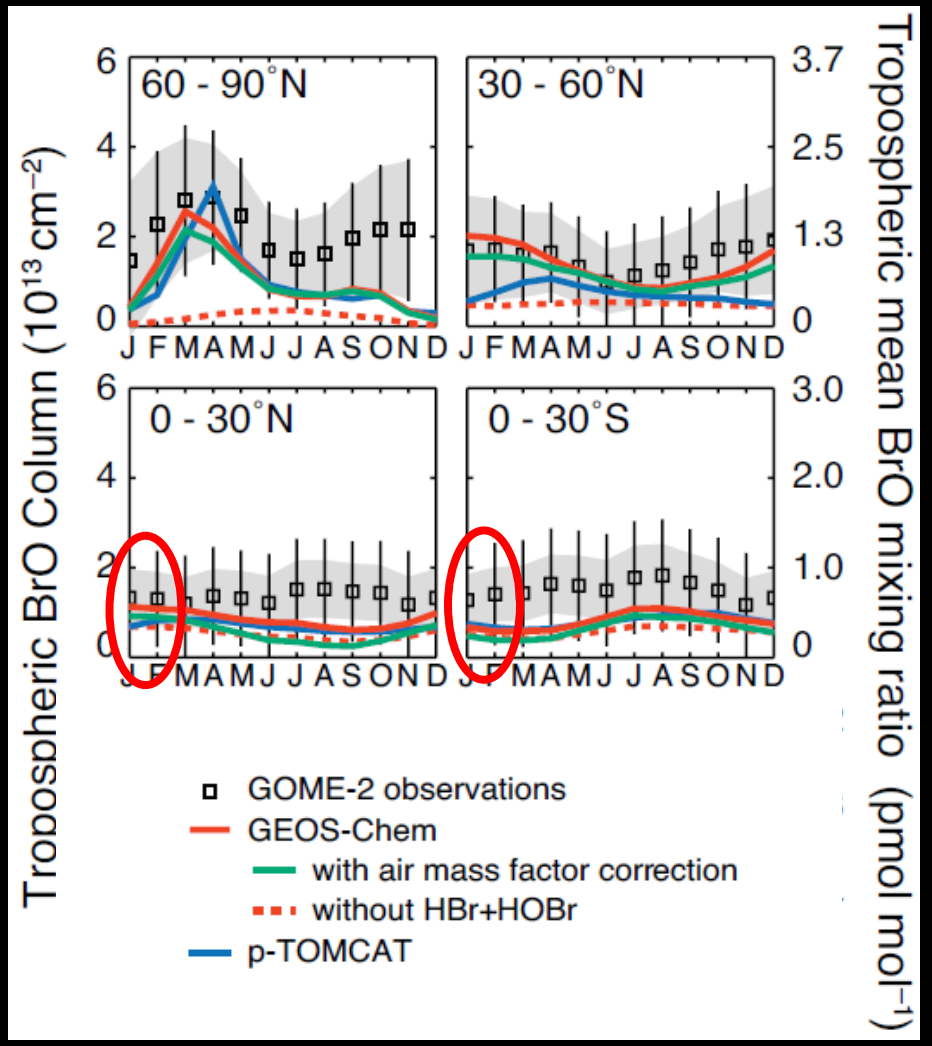
(Schofield et al., 2004 , Hendrick et al., 2007; Theys et al., 2007; Coburn et al., 2011; Coburn et al., 2014, in prep.)

Balloon: 0.2-0.3 x10^{13} molec cm^{-2}

(Pundt et al., 2002; Dorf et al., 2008)

Models: 0.2-1.0 x10^{13} molec cm^{-2} (~ 0.2-0.5 ppt)

(Saiz Lopez et al., 2012; Parrella et al., 2012) – in the tropics



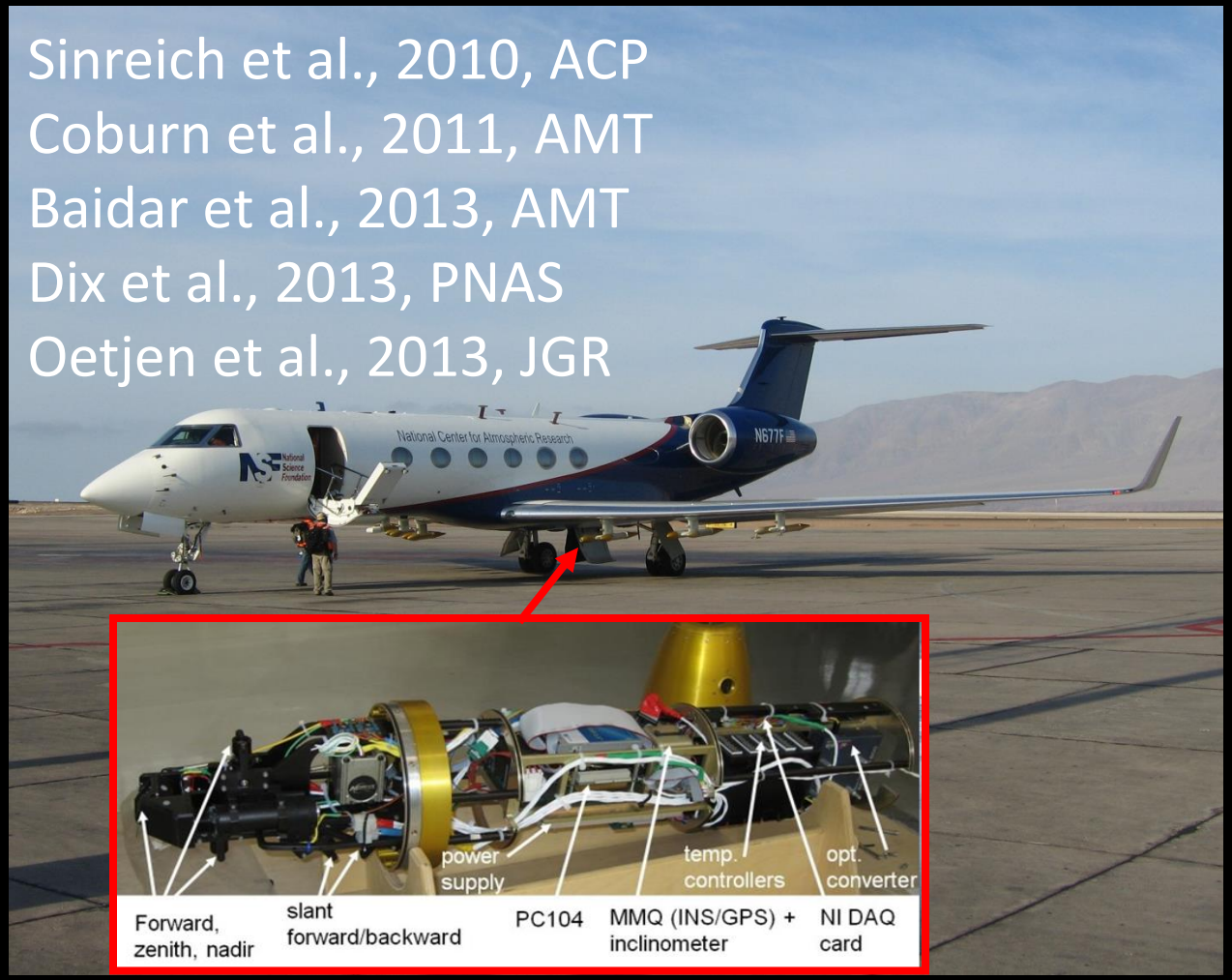
Theys et al. [2011]

Halogens deplete the O_3 column by ~10% in the tropics (Saiz-Lopez et al., 2012)

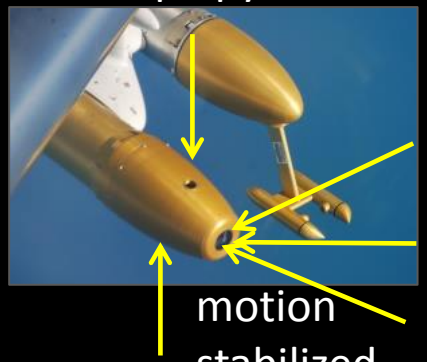
CU-AMAX-DOAS instrument aboard NSF/NCAR GV

University of Colorado Airborne Multi-AXis Differential Optical Absorption Spectroscopy

Sinreich et al., 2010, ACP
Coburn et al., 2011, AMT
Baidar et al., 2013, AMT
Dix et al., 2013, PNAS
Oetjen et al., 2013, JGR



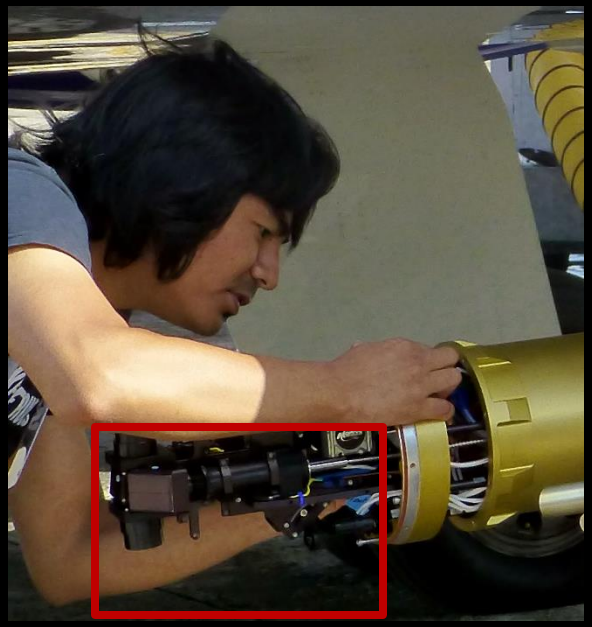
Telescope pylon

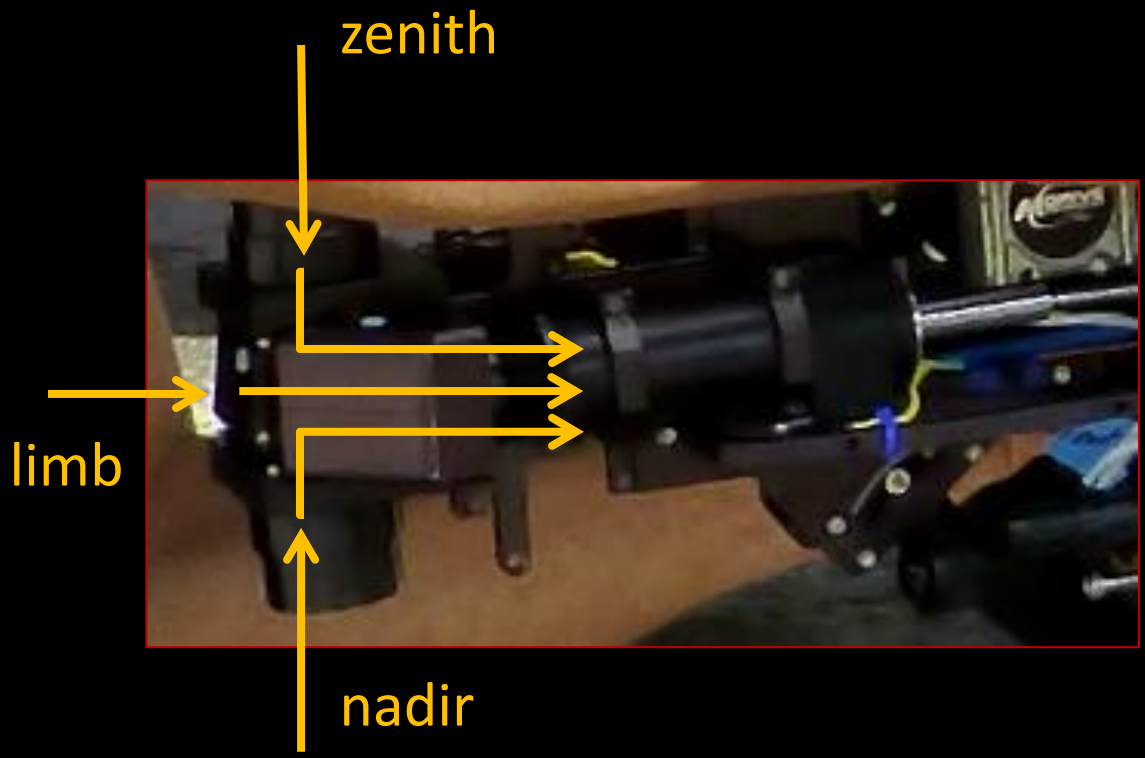


spectrographs/detectors

CU-AMAX-DOAS instrument aboard NSF/NCAR GV

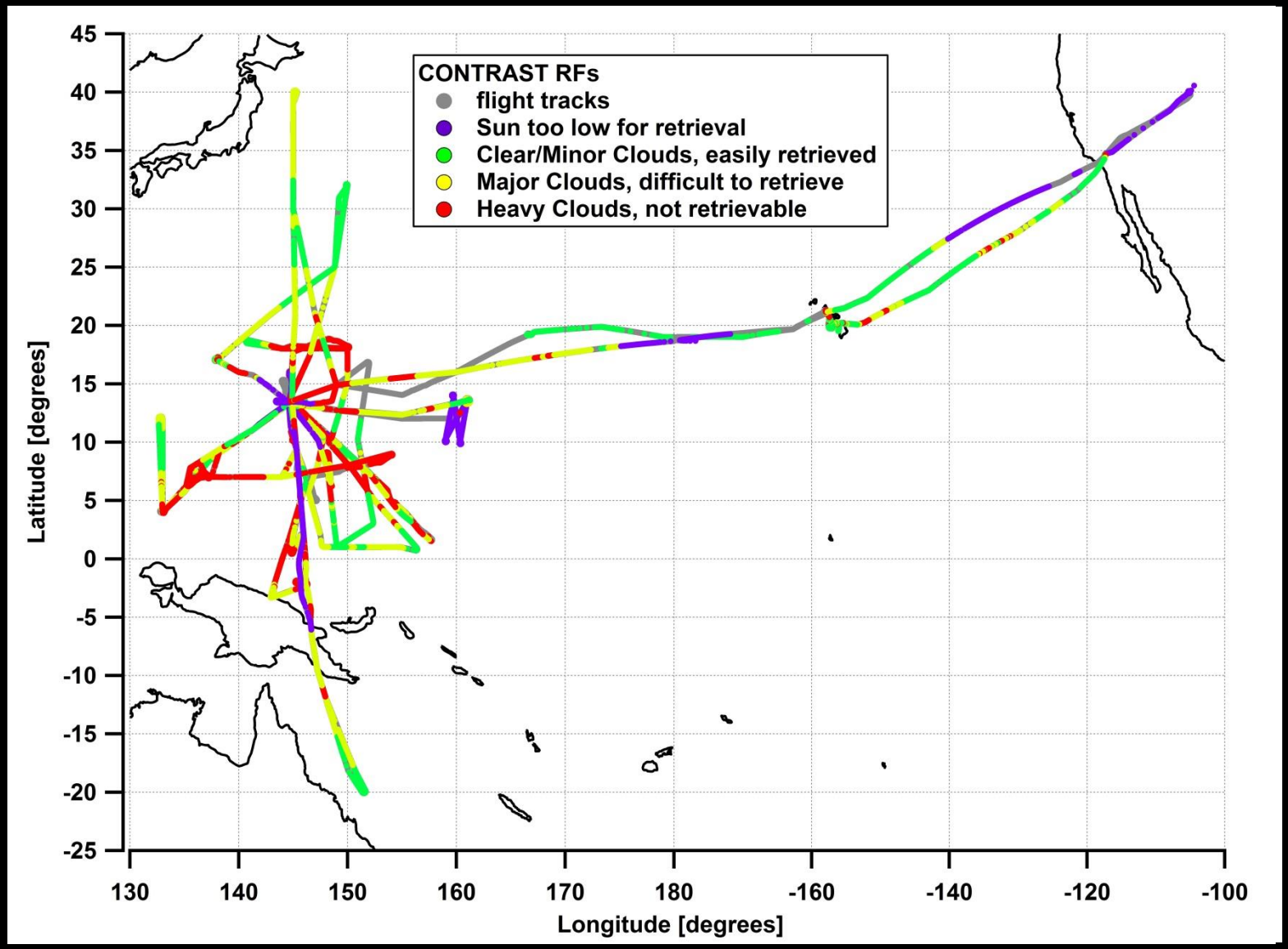
Hardware: new telescope design implemented for CONTRAST
Software: Autonomous deployment on the NSF/NCAR GV





→ Successful: more flexibility to record reference spectra
→ Successful: remote control in flight (RF07)
→ **Primary benefit is added flexibility**

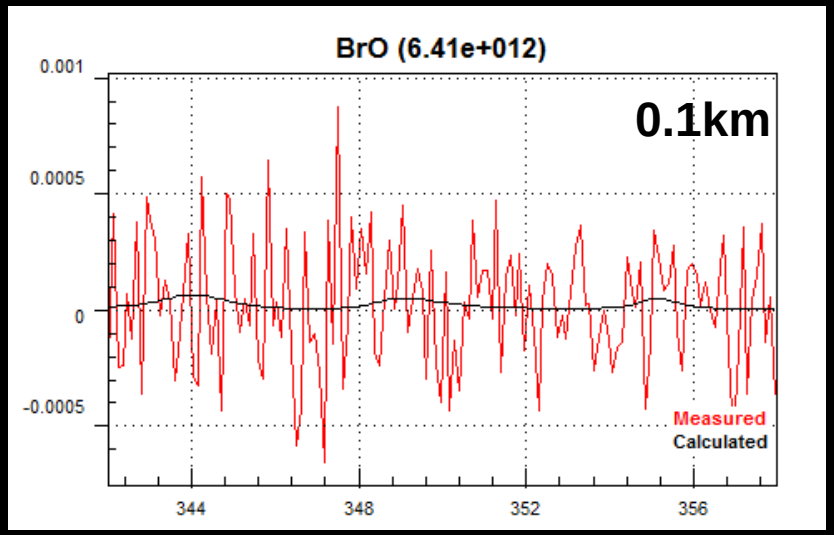
CU-AMAX-DOAS data status
- premise -

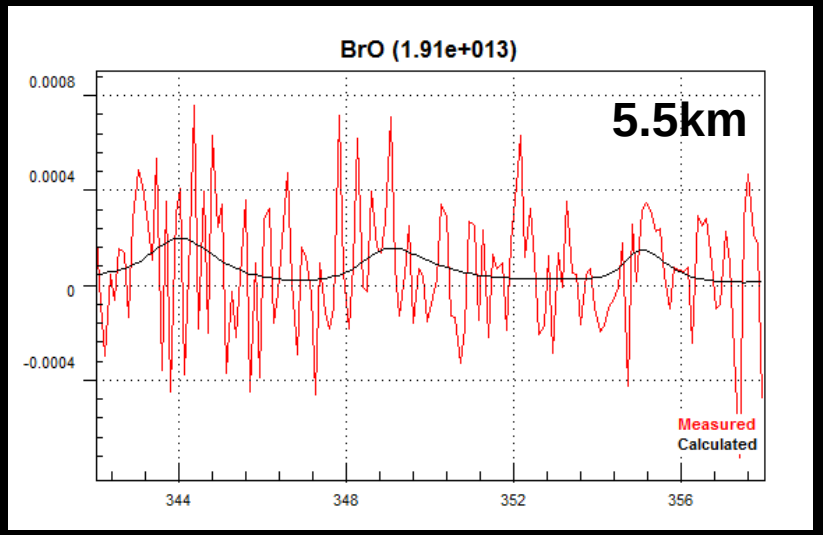


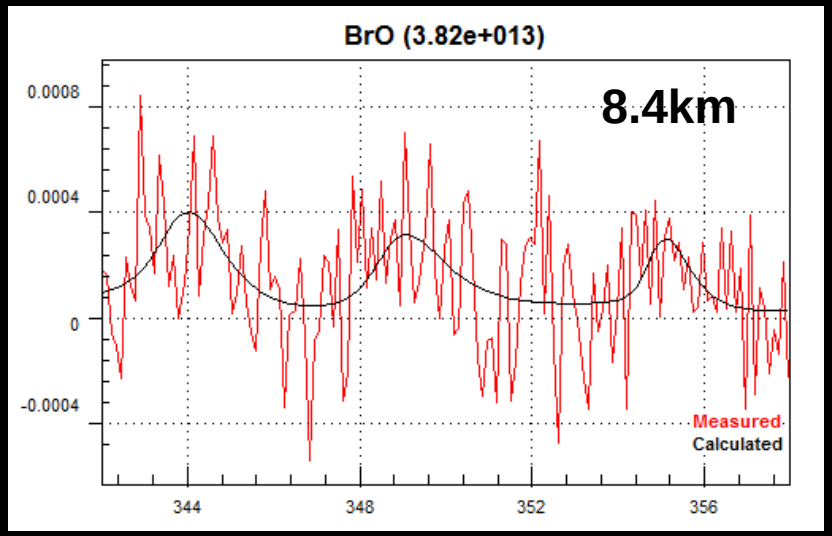
CU-AMAX-DOAS data status

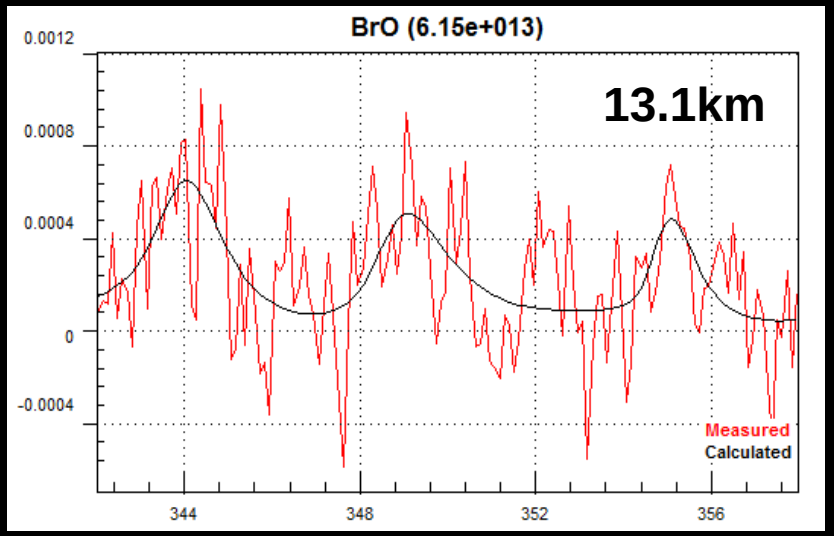
Flight number	AMAX-DOAS data	Final dSCDs	mixing ratios	major/heavy clouds [%]
RF01	yes	yes	BrO	39.1
RF02	partially	yes	BrO	17.5
RF03	yes	yes	BrO	59.3
RF04	yes	yes	BrO,IO,NO_2	51.2
RF05	yes	yes		68.6
RF06	yes	yes	BrO	50.2
RF07	yes	yes		87.1
RF08	no	-	-	-
RF09	yes	yes		79.6
RF10	yes	yes		45.2
RF11	yes	yes		62.6
RF12	partially	yes		100
RF13	yes	yes		45.8
RF14	yes	yes		61.6
RF15	yes	yes	BrO, IO, NO_2	62.8
RF16	yes	yes		70.5
RF17	yes	yes	BrO,IO	13.8

BrO detection during CONTRAST (RF04)



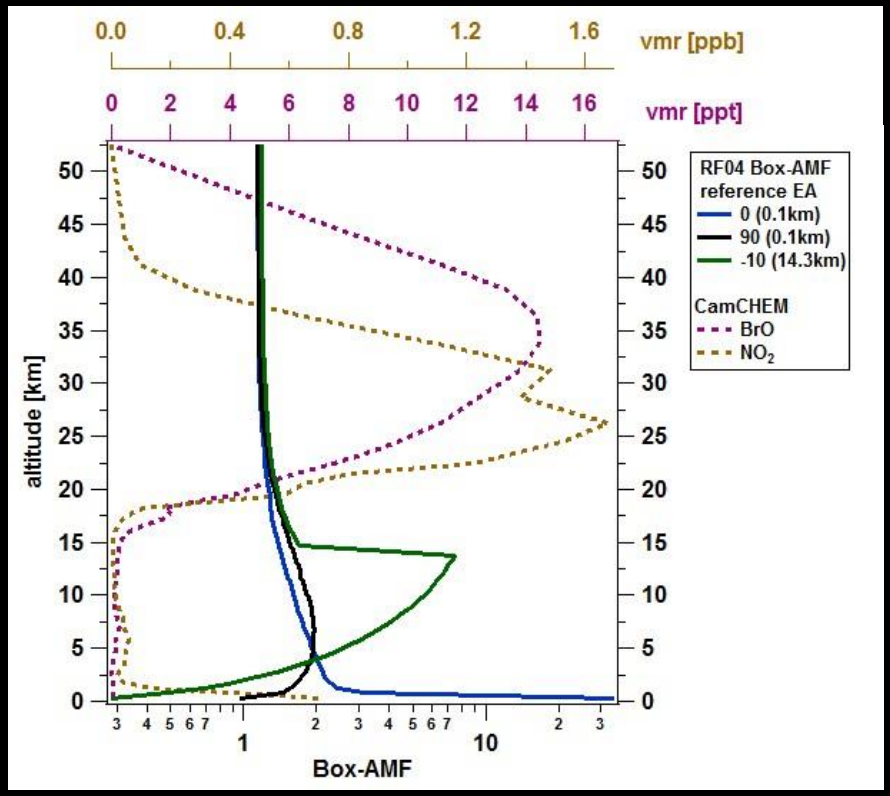




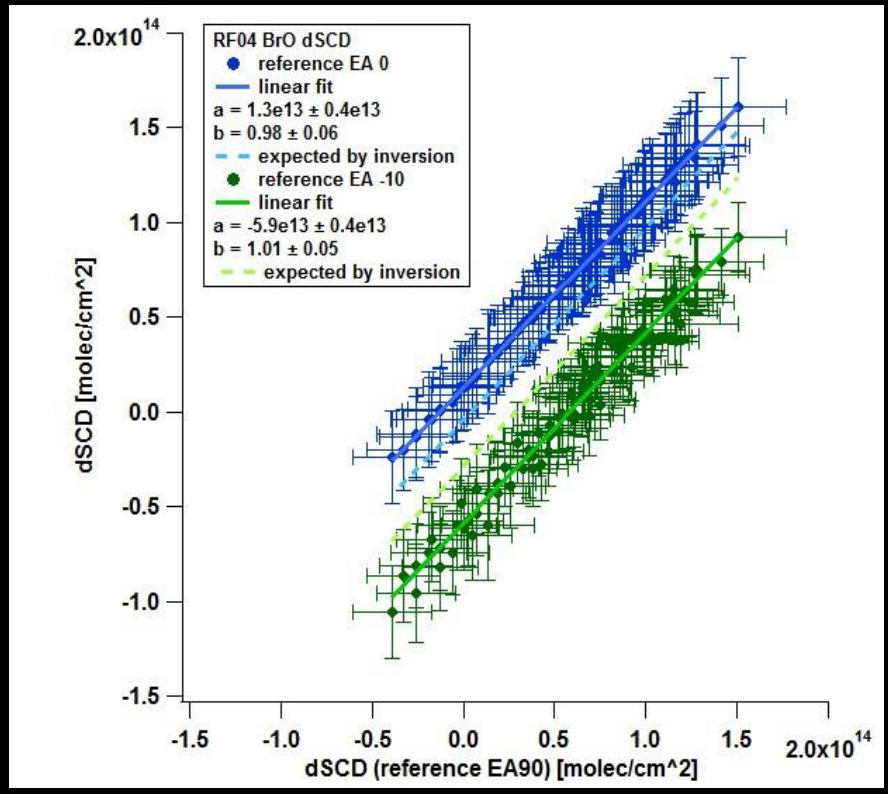


→ significant BrO detection above 8km

BrO retrieval - robustness



stratospheric correction

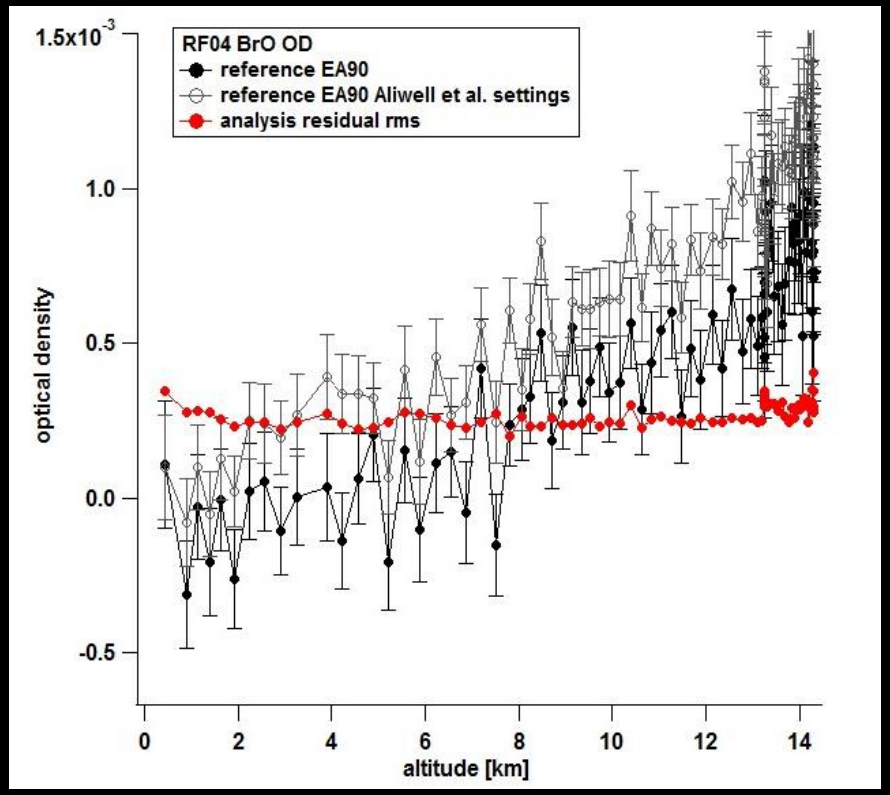


consistency between references

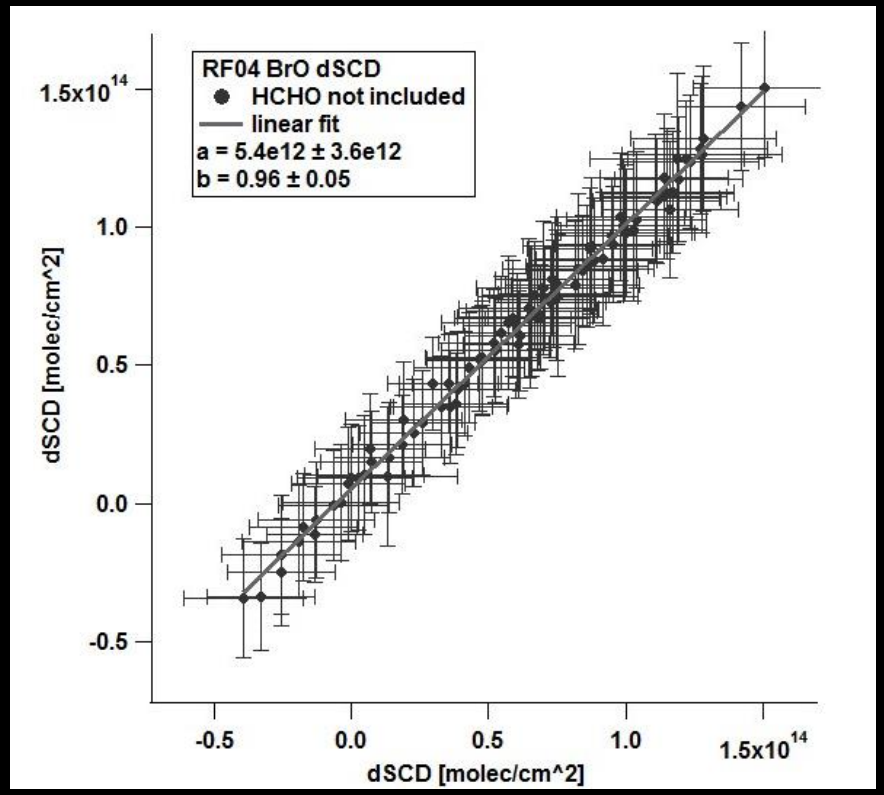
→ **stratospheric contributions are reliably cancelled out**

→ **consistent dSCD offset between different reference geometries**

BrO retrieval - robustness



comparison with “Aliwell” settings

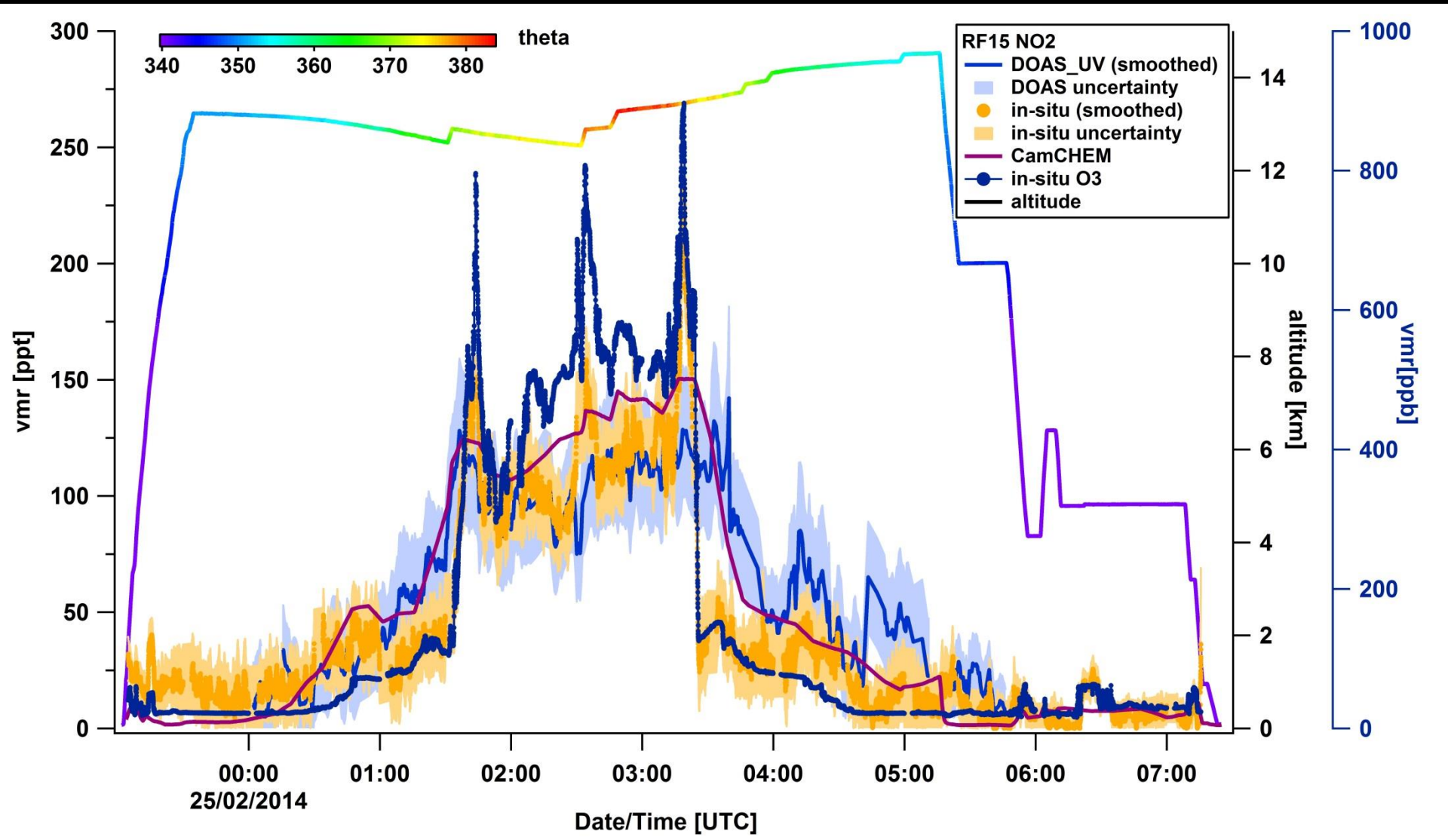


sensitivity to HCHO cross section

→ **BrO fit settings: 3-band analysis; BrO is conservatively bound**

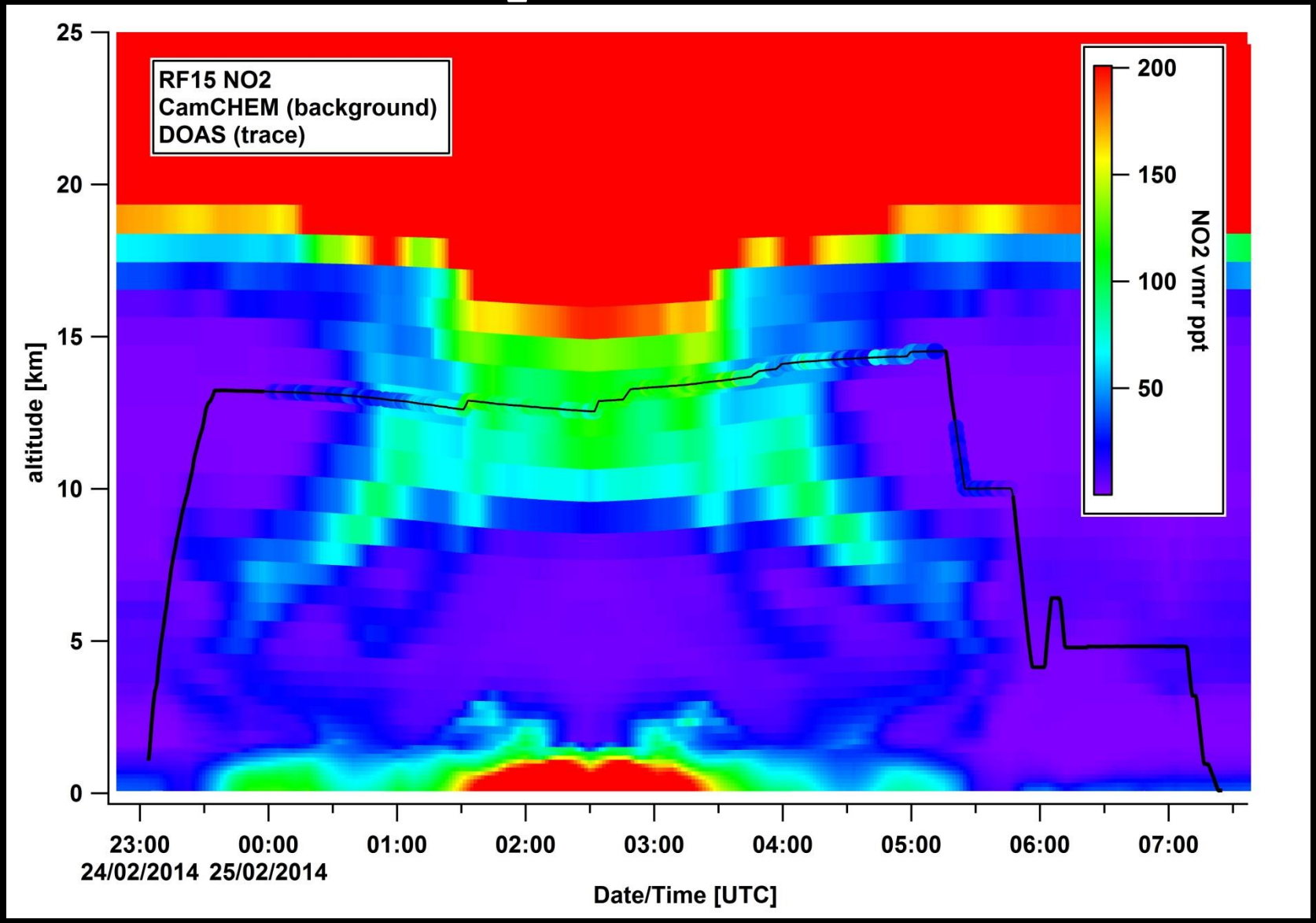
→ **Including/excluding HCHO has no effect on BrO dSCDs**

RF15 NO_2 – comparing column and in-situ vmr



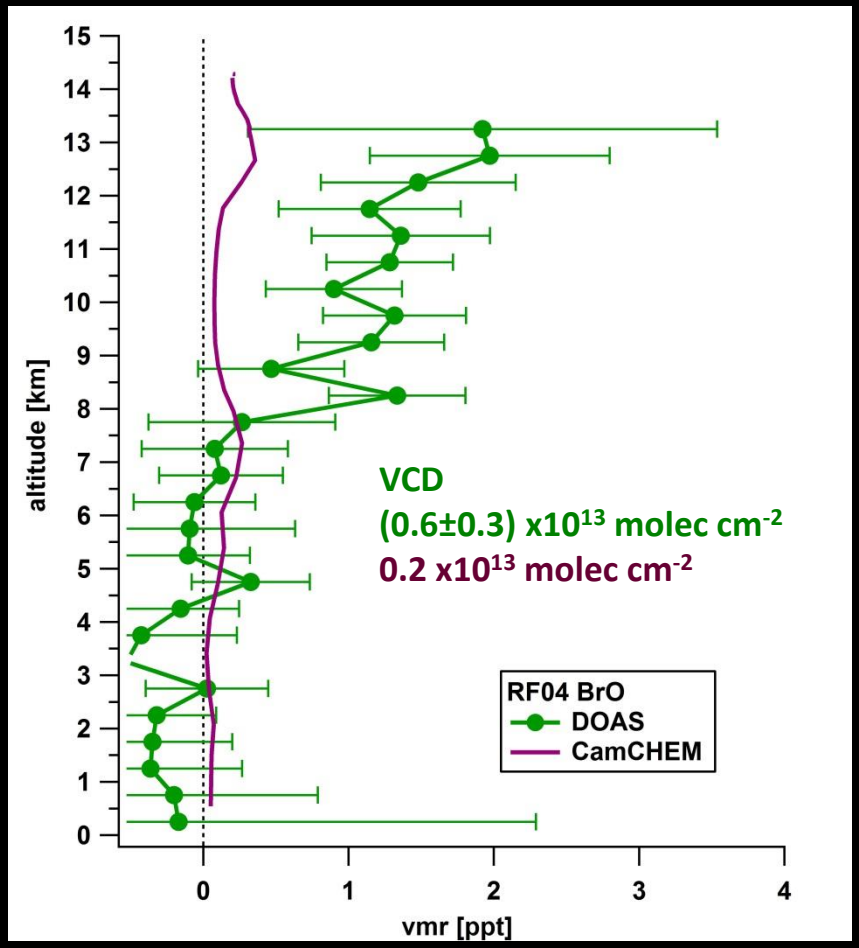
very good agreement: → RTM control → Homogeneity

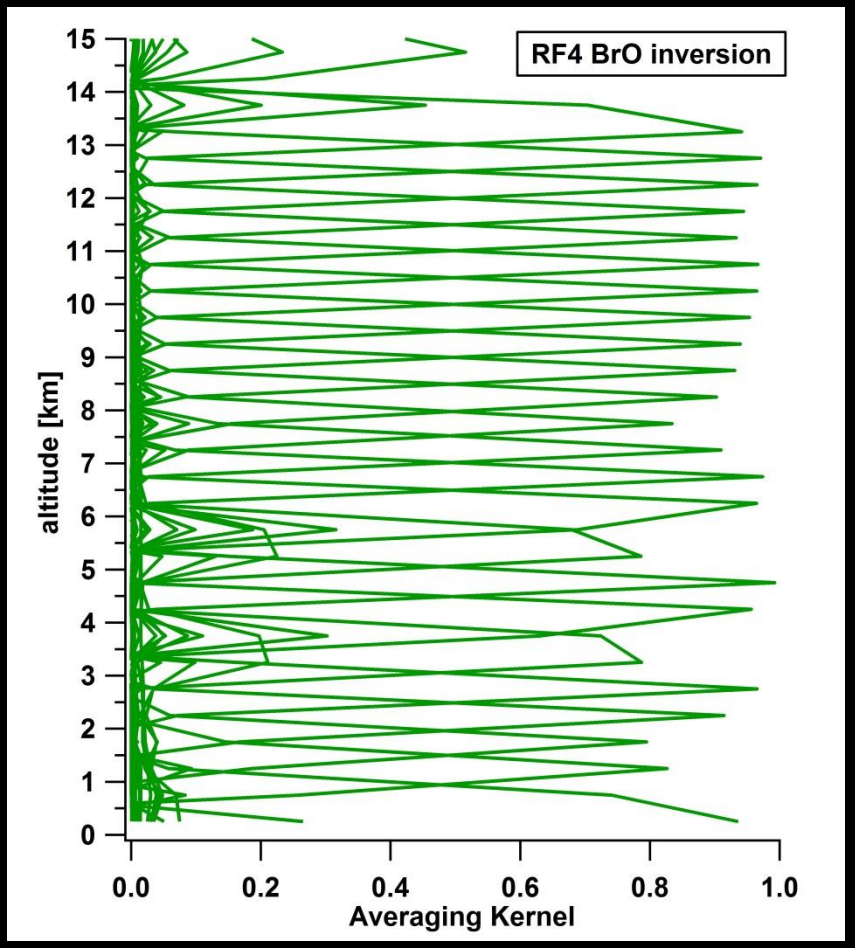
RF15 NO_2 DOAS/CamCHEM



very good agreement: → RTM control → Homogeneity

RF04 BrO vertical profile

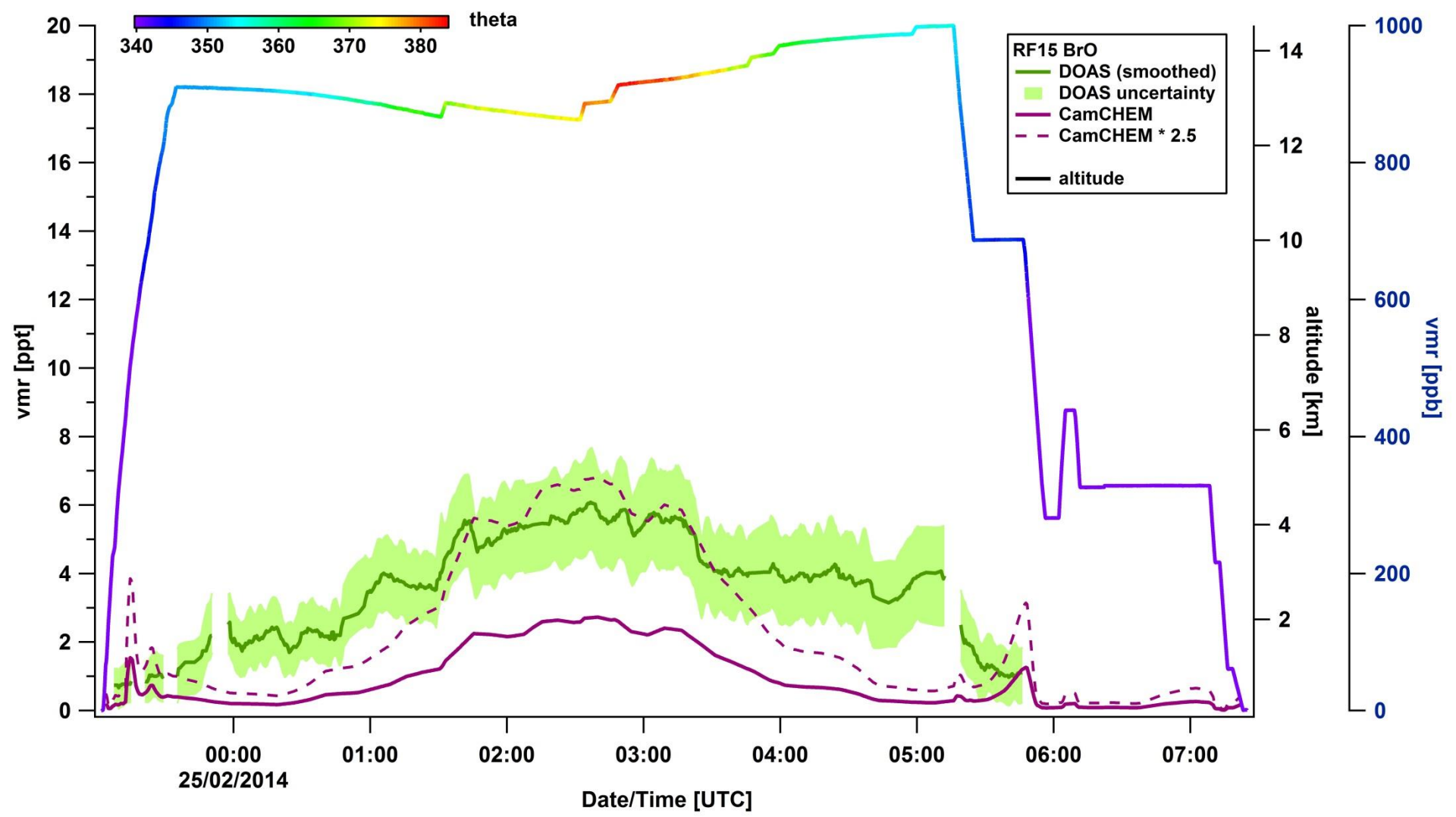




→ high DoFs; inversion is fully constrained by measurements

→ observed BrO underestimated in upper FT by model

RF15 BrO DOAS/CIMS/CamCHEM along flight track

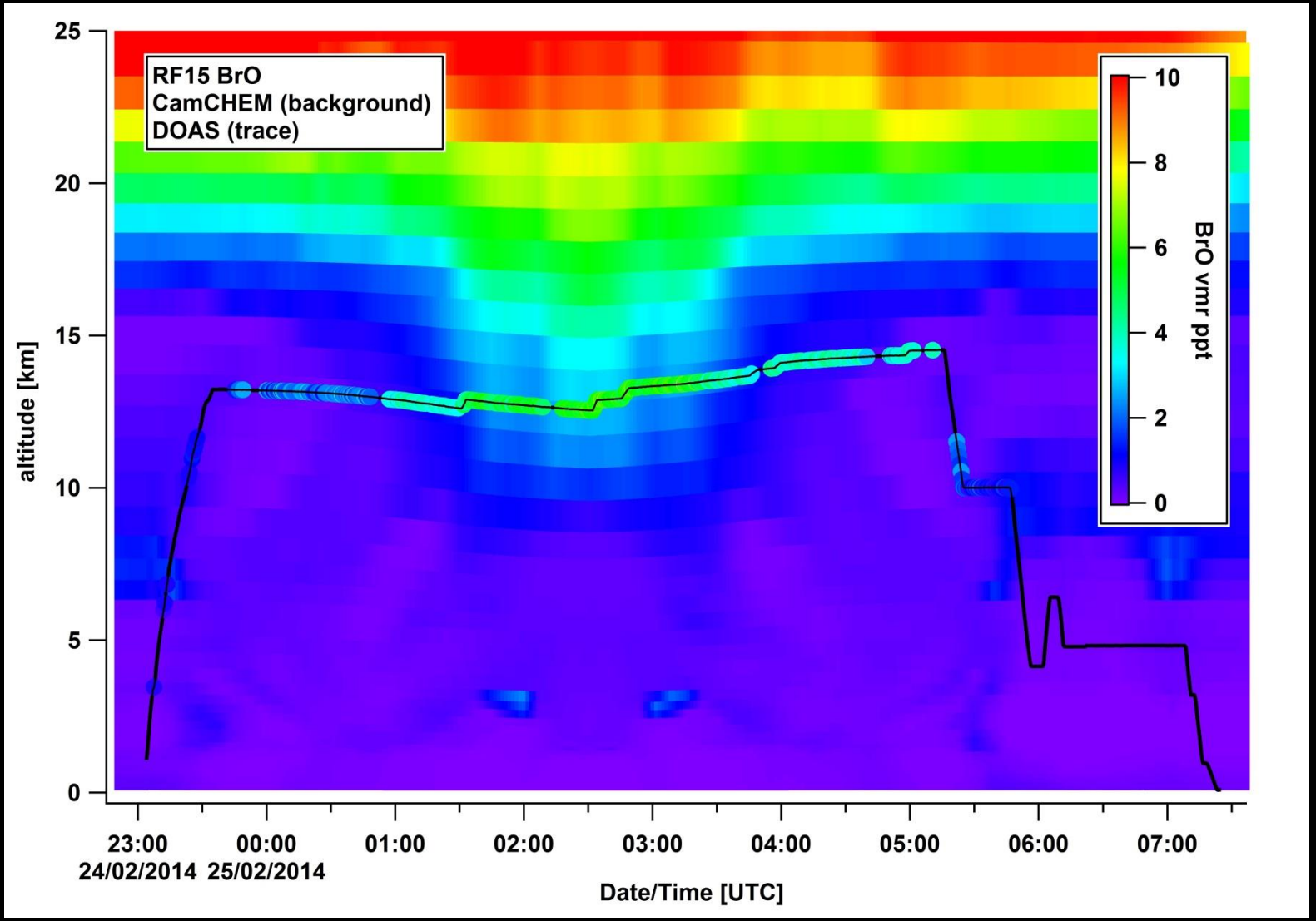


→ DOAS and CIMS agree at theta(max)

→ DOAS BrO reproduces model gradients

→ observed BrO ~factor 2.5 higher in stratosphere and >2.5 outside

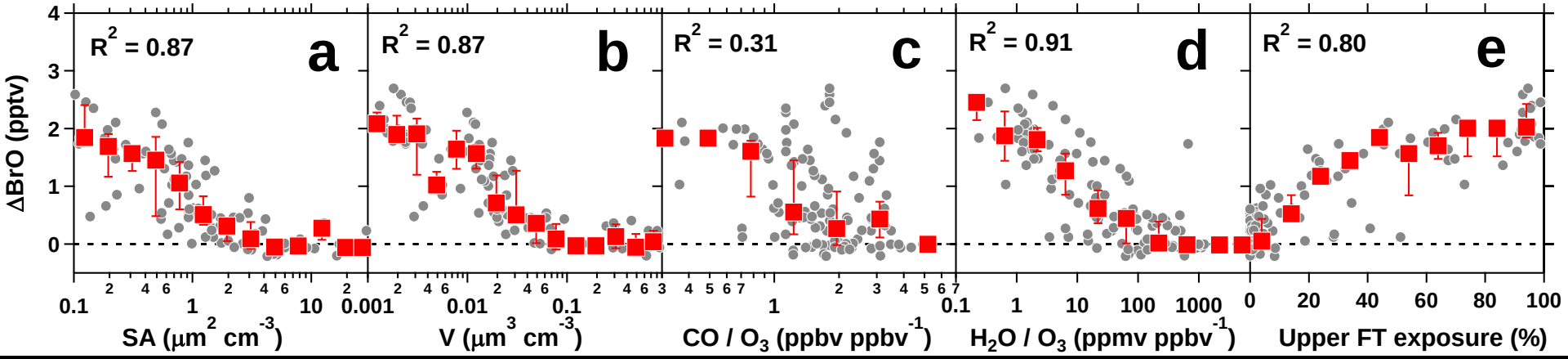
RF15 BrO: comparing DOAS & CamCHEM



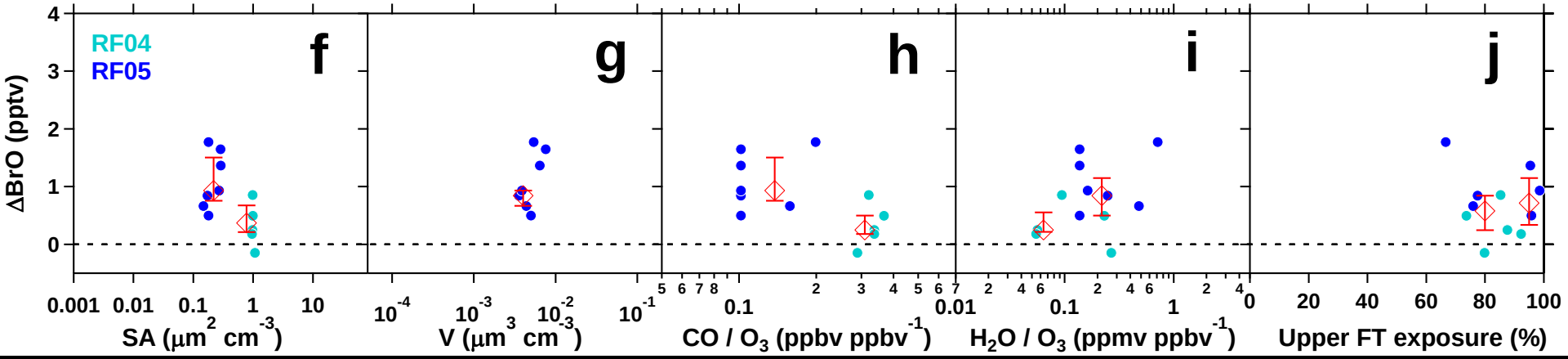
→ DOAS BrO follows model gradients but shows higher BrO, particularly in upper FT

TORERO ΔBrO (unexplained BrO) – correlations

Tropospheric air



Lower stratospheric air



- Unexplained BrO in the upper tropical FT:
  - correlates with uFT exposure, decreasing H_2O/O_3 ratios (stratospheric tracer)
  - Is anti-correlated with aerosol SA
  - BrO in the lower stratosphere seems underestimated

Conclusions

- **BrO is significantly detected above 6 km during RF04 and RF15.**
  - **Retrievals are robust**
  - **NO_2 shows RTM control and homogeneity for RF15**
- ***Western Pacific: BrO in UT is lower than over the Eastern Pacific, but higher than predicted by models (Western and Eastern Pacific)***
  - ***BrO VCD is 60% /12% lower than GOME-2, consistent with ground-based MAX-DOAS data (Theys et al., 2011)***
  - ***BrO in the lower stratosphere is higher than predicted***
- ***Eastern Pacific: stratospheric sources are underestimated***
  - ***Elevated BrO is sensitive to BrY in the LS (injected as bromocarbons over Western Pacific?), and UTLS dynamics (GEOS4/GEOS5 sens. studies).***
  - ***Stronger convection (GEOS4) leads to improvements in O_3 profiles, and invigorates UTLS transport***
- **Comparison of RF01/RF17 BrO with ground based MAX-DOAS at MLO presented at AGU 2014**

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