NORS - WP4 - Task 4.2

Retrievals of formaldehyde from ground-based FTIR and MAX-DOAS observations at the Jungfraujoch station and comparisons with GEOS-Chem and IMAGES model simulations

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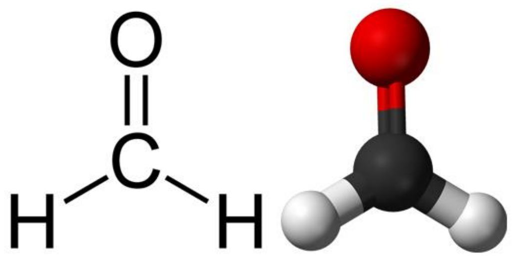
NORS/NDACC/GAW workshop, 5 to 7 November 2014, Brussels

Formaldehyde (HCHO)



- Midday lifetime of a few hours
- Sources:
  - mainly by oxidation of:
    - CH_4
    - primary NMVOCs
  - (directly from various sources)
- Sinks:
  - photolysis
  - oxidation by OH radicals
    => yield CO and HO_2
  - (dry and wet deposition)
- Involved in the VOC – HO_x – NO_x chemistry
  generating or destroying tropospheric O_3

HCHO = indicator of NMVOCs emissions from continental sources

- biogenic (≈ 85 %)
- anthropogenic (≈ 12 %)
- pyrogenic (≈ 3 %)

- **oxidative capacity of the atmosphere**
- **the global CO budget**

Key role for air quality monitoring

Formaldehyde (HCHO)

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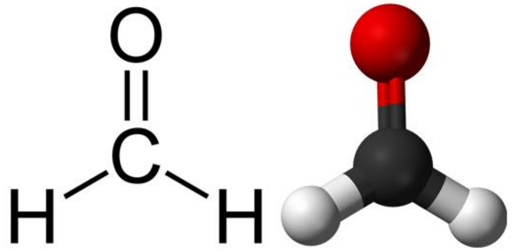


Issues

- **Can we detect background levels of HCHO in the remote troposphere from ground-based FTIR and MAX-DOAS?**

- **Is there a good consistency between both instruments regarding HCHO at a high-altitude station?**

- **Validating an optimized FTIR retrieval strategy for HCHO above Jungfraujoch as a preparation for further studies**
  e.g., multi-decadal timeseries at Jungraujoch

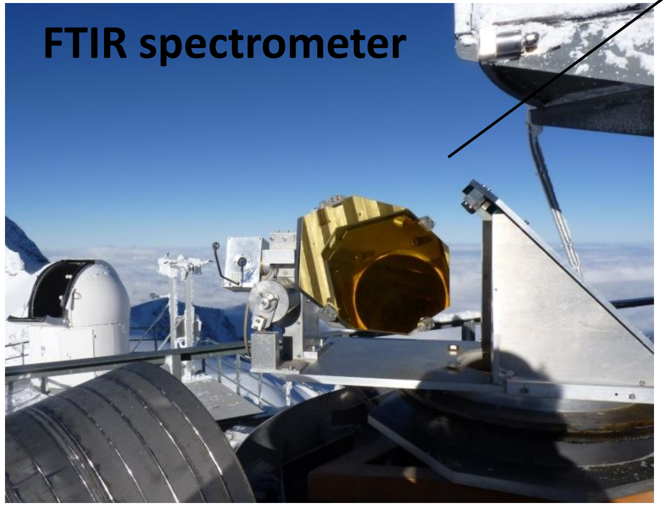
⇒ Franco et al. (2014), Atmos. Meas. Tech. Discuss., doi:10.5194/amtd-7-10715-2014

Measurement site: Jungfraujoch station (Swiss Alps, 46.5° N, 8.0° E, 3580 m a.s.l.), part of the NDACC network

- Essentially located in the free troposphere during winter
- Frequent injections of air masses from the boundary layer, especially during summer

- More than 35 years of uninterrupted IR monitoring

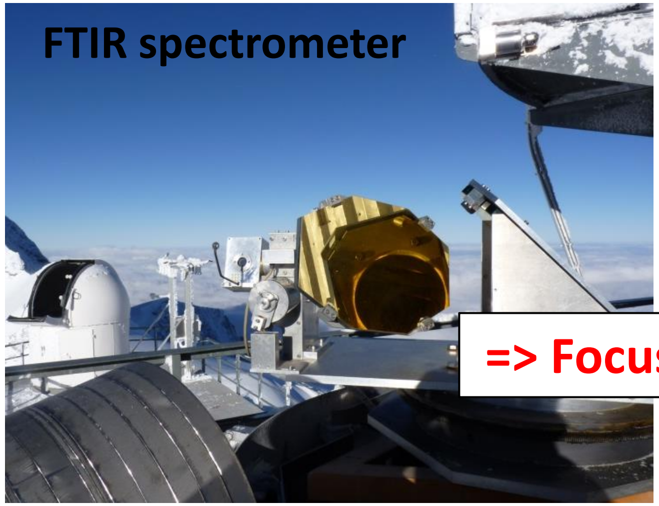
Measurement site: Jungfraujoch station (Swiss Alps, 46.5° N, 8.0° E, 3580 m a.s.l.), part of the NDACC network

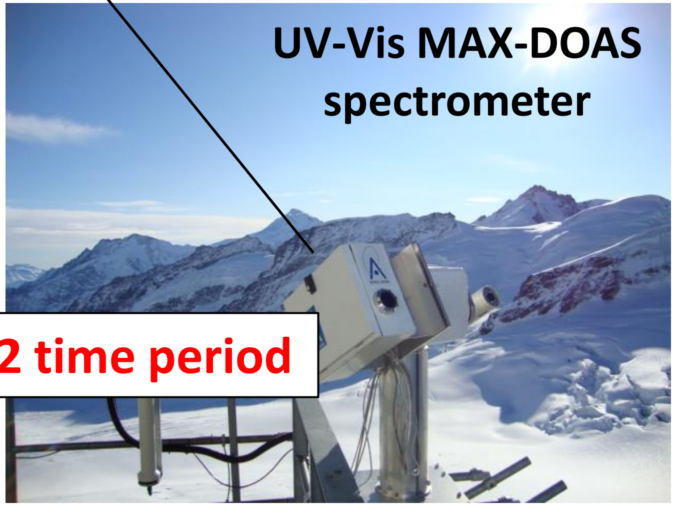


- Bruker IFS-120 HR operated by ULg
- Under clear-sky conditions
- Optical filter: 2400-3310 cm^{-1}
- Spectral resolution: 0.004 and 0.006 cm^{-1}

Measurement site: Jungfraujoch station (Swiss Alps, 46.5° N, 8.0° E, 3580 m a.s.l.), part of the NDACC network

- Operated by BIRA-IASB since 2010
- Pointing NE direction (city of Bern)
- Elevation angles used here: 0°, 1°, 3°, 4°, 5°, 8°, 10°, 12°, 15°, 30°
- Measurements from 85° SZA sunrise to 85° SZA sunset
- 20’ per scan





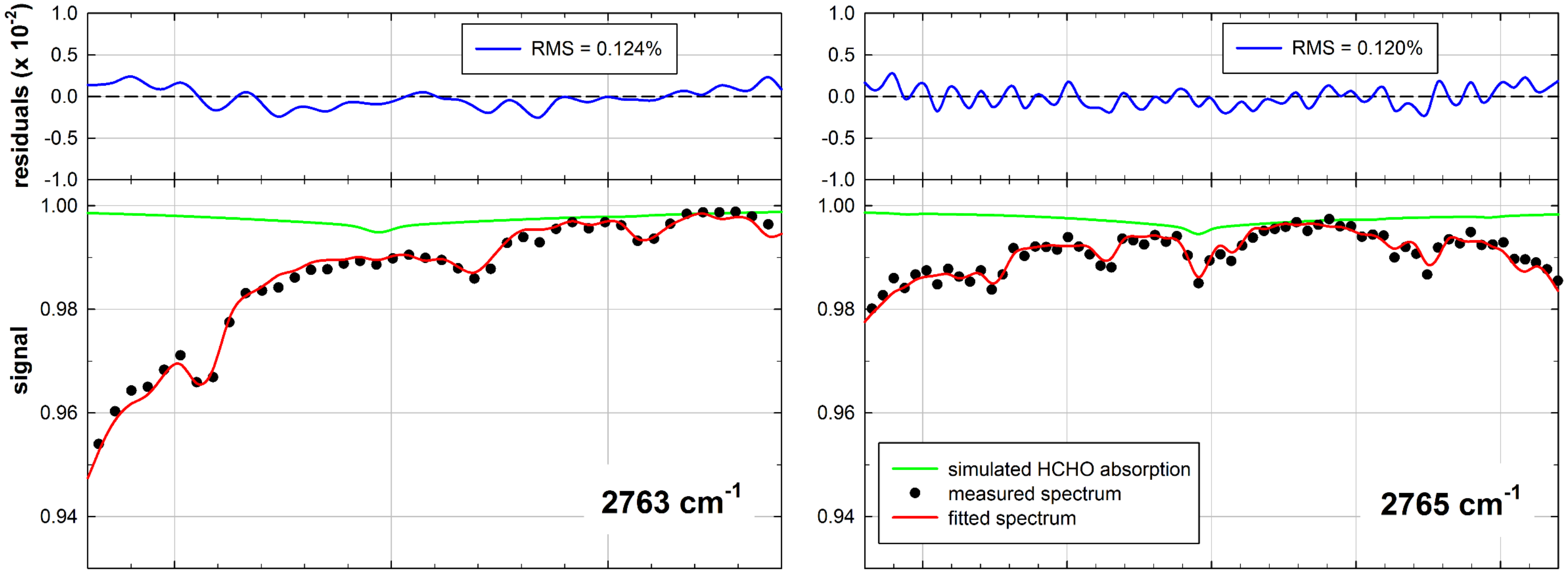
=> Focus on the mid-2010 – 2012 time period

FTIR retrieval strategy

- SFIT-2 v3.91 algorithm
- Spectroscopic line parameters from HITRAN 2008
  => updated line strength for HCHO from Perrin et al. (2009)
- A priori from 1980 – 2020 WACCM v.6 simulation
  => good consistency with 36.5 – 56.5° N zonal occultations from ACE-FTS
- Optimal Estimation Method for the retrieval process
  => covariance matrix derived from slightly « relaxed » WACCM values

Microwindows (cm^{-1})	Interfering species
2763.425 – 2763.600	HDO, CH_4, O_3, N_2O, CO_2
2765.725 – 2765.975	HDO, CH_4, O_3, N_2O, CO_2
2778.200 – 2778.590	HDO, CH_4, O_3, N_2O, CO_2
2855.650 – 2856.400	HDO, CH_4, O_3, N_2O, H_2O

Based on **Vigouroux et al. (2009)**, Atm. Chem. Phys.



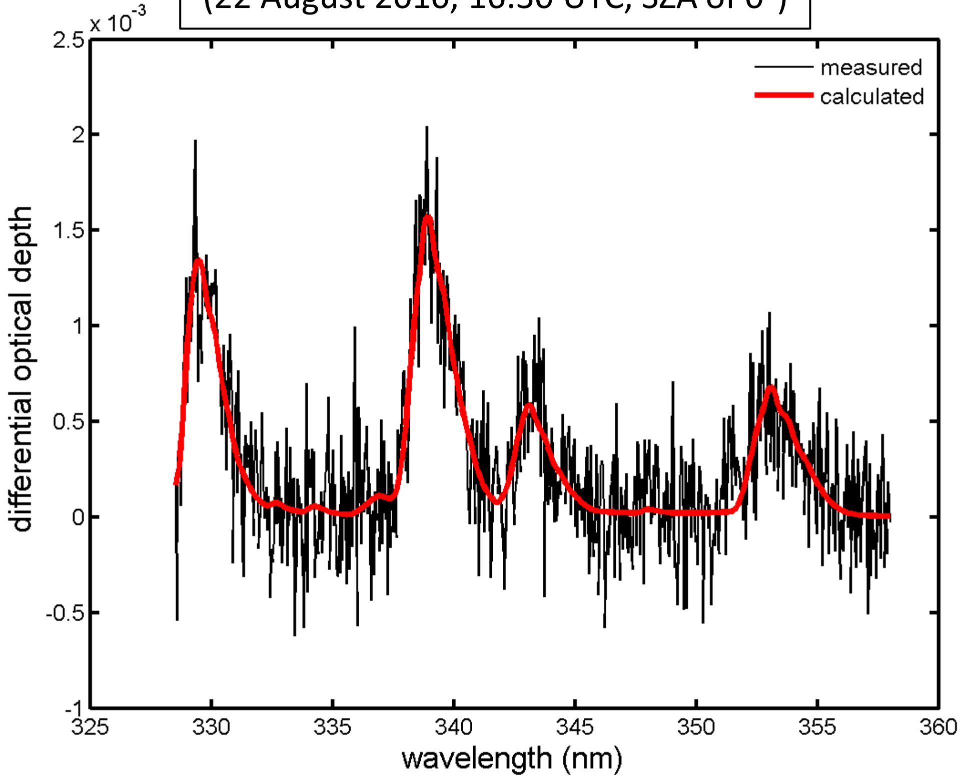
Individual FTIR fit example
(22 August 2010, 6:40 UTC, SZA of 80°)

=> Very weak solar IR absorption
+ broad spectral lines

1. DOAS spectral fitting => DSCDs

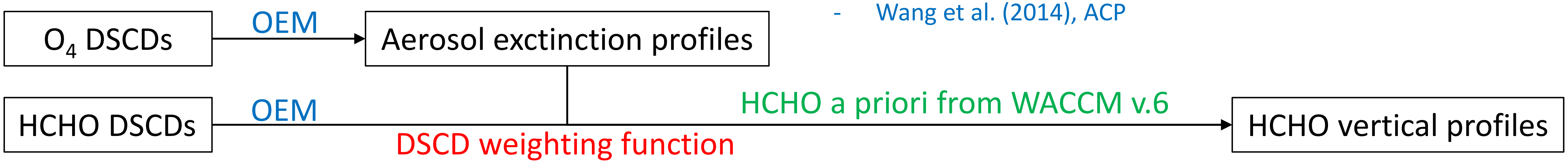
- Fitting window: 328.5 – 358.0 nm
  => minimizing the HCHO/BrO correlation
- Zenith spectrum of each scan taken as reference
  => reducing the interference by O_3
- Fitted species:
  - HCHO at 293 K
  - NO_2 at 298 K
  - O_3 at 223 and 243 K
  - O_4
  - BrO at 223 K
  - Ring effect
- 5^{th}-order polynomial fit and linear correction for off-set

Individual DOAS fit example
(22 August 2010, 16:30 UTC, SZA of 0°)

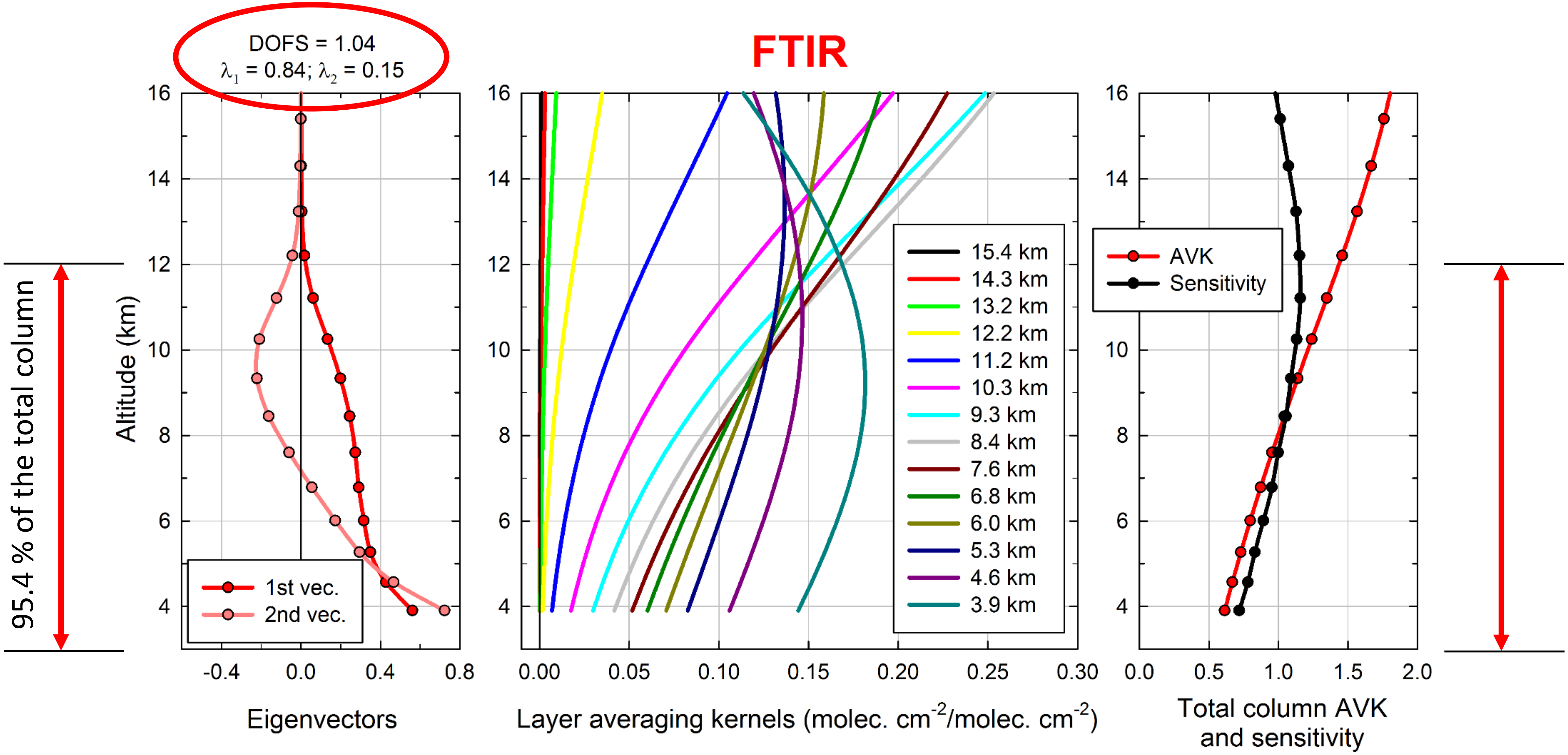


2. Profile retrieval => OEM-based profiling tool bePRO

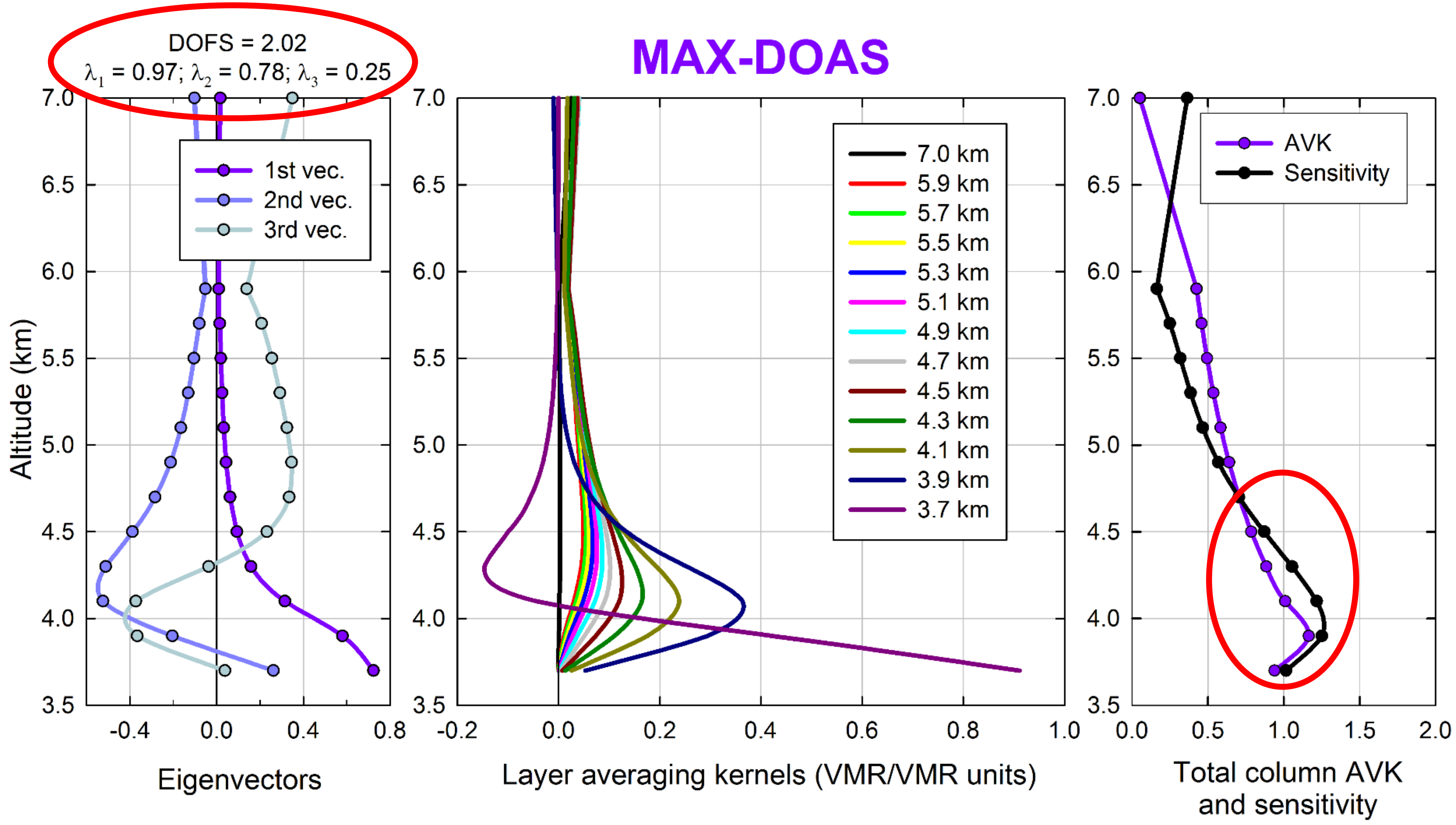
- Clémer at al. (2010), AMT
- Hendrick et al. (2014), ACP
- Wang et al. (2014), ACP



Characterization of FTIR retrievals



Characterization of MAX-DOAS retrievals



Characterization of the retrievals

- FTIR: mainly sensitive throughout the free troposphere
- MAX-DOAS: highly sensitive in the lowest layers

=> **Complementary** information content in the troposphere regarding HCHO

=> Direct comparisons between both instruments = little meaning

=> HCHO distributions from 3-D CTMs (GEOS-Chem and IMAGES) as **intermediates**

→ Smoothed by the FTIR and MAX-DOAS AVK

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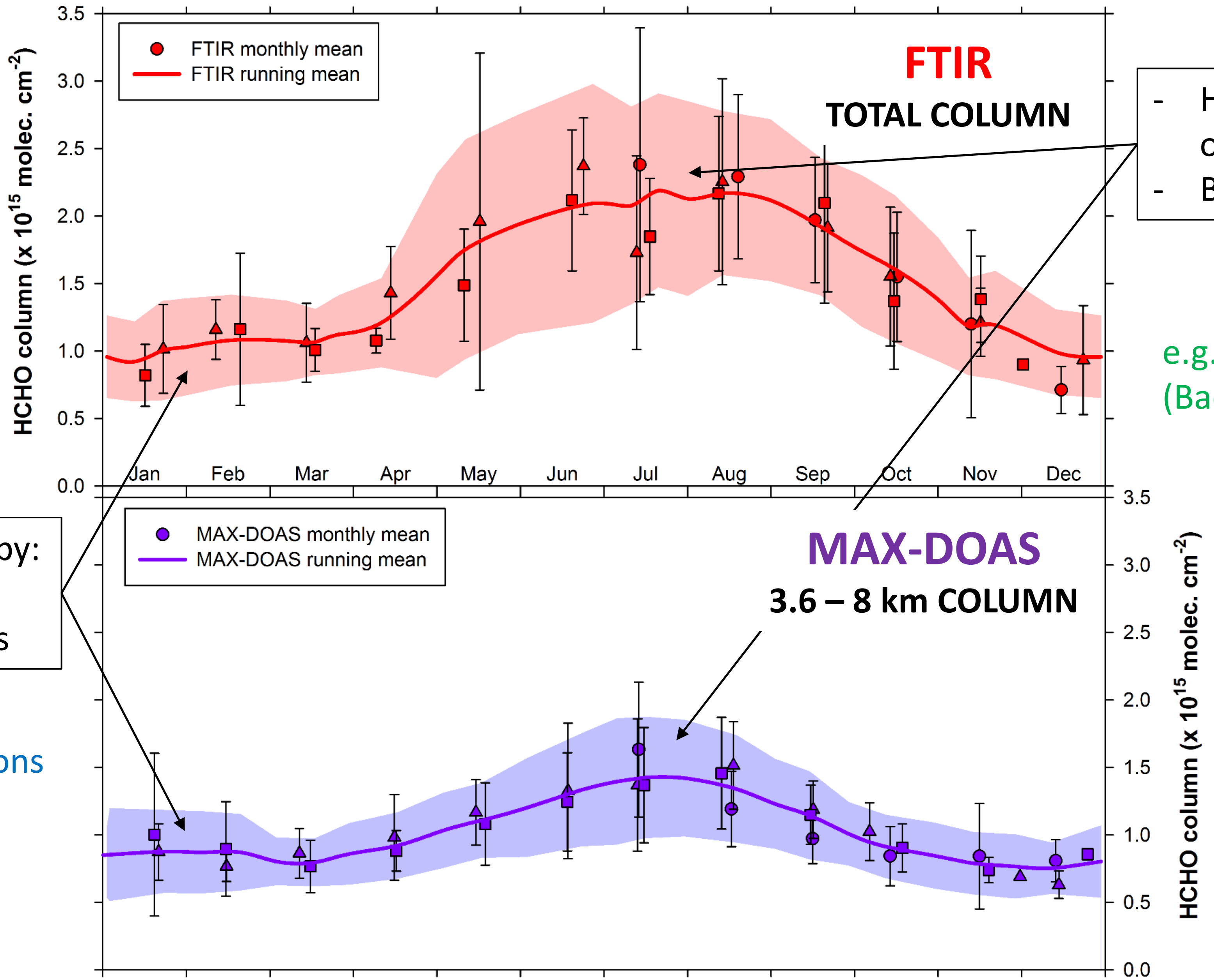
	GEOS-Chem (v9-01-03)	**IMAGES v2**
Horizontal resolution	2.0° x 2.5°	2.0° x 2.5°
Meteorological forcings	GMAO GEOS-5	ECMWF ERA-Interim
CH4 concentrations	NOAA Global Monitoring Division	NOAA Global Monitoring Division
Biogenic emissions	MEGAN v2.0	MEGAN v2.0
Biomass burning emissions	GFED v3	GFED v3
Anthropogenic emissions	EMEP (CO, NO_x, SO_x, and NH_3) RETRO and EMEP (NMVOCs)	EMEP (CO, NO_x, SO_x and NH_3) RETRO (NMVOCs)

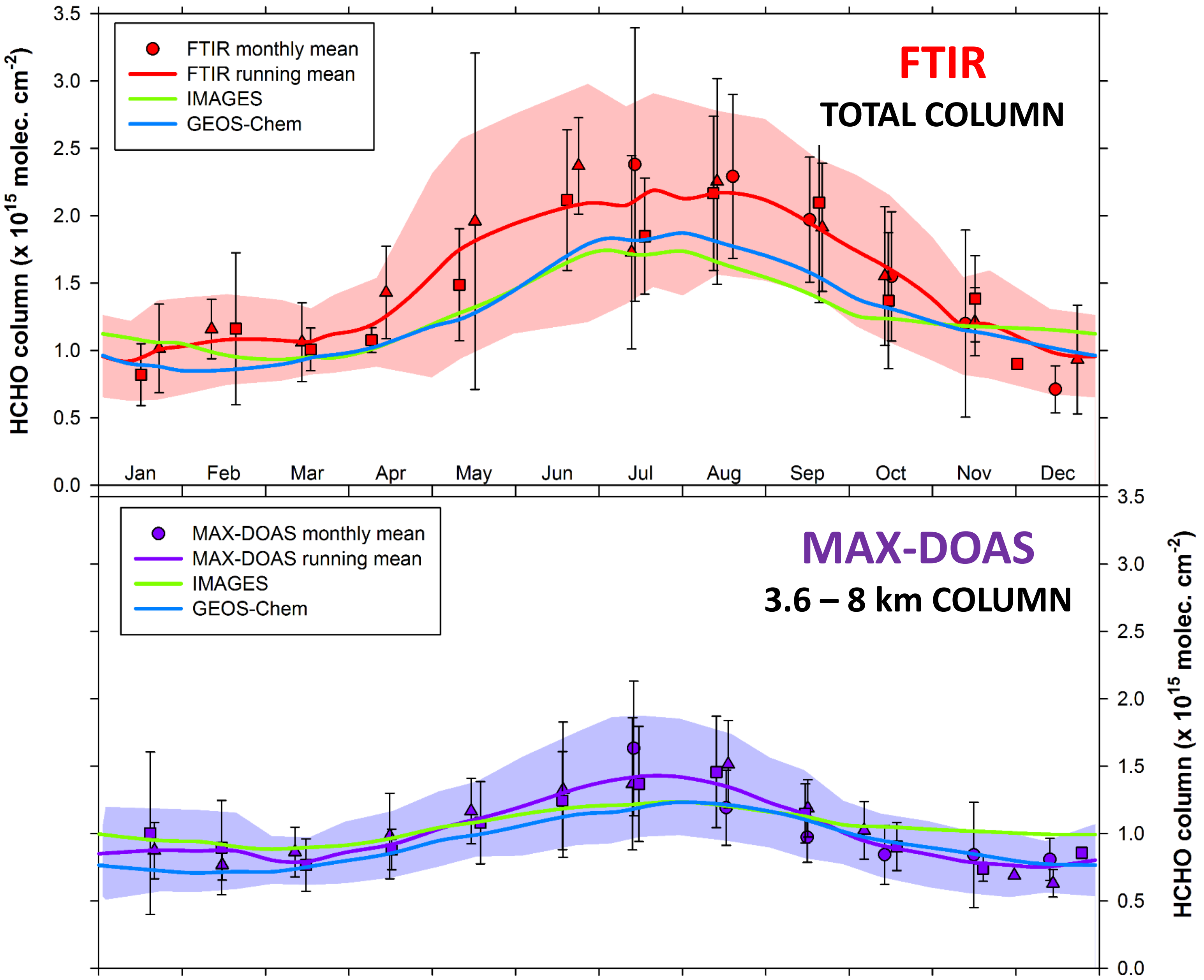
Background provided by:
- CH_4
- Long-lived NMVOCs

Anthropogenic emissions (butane, toluene, …)
- Northern Italy
- Southern France
- Southern Germany

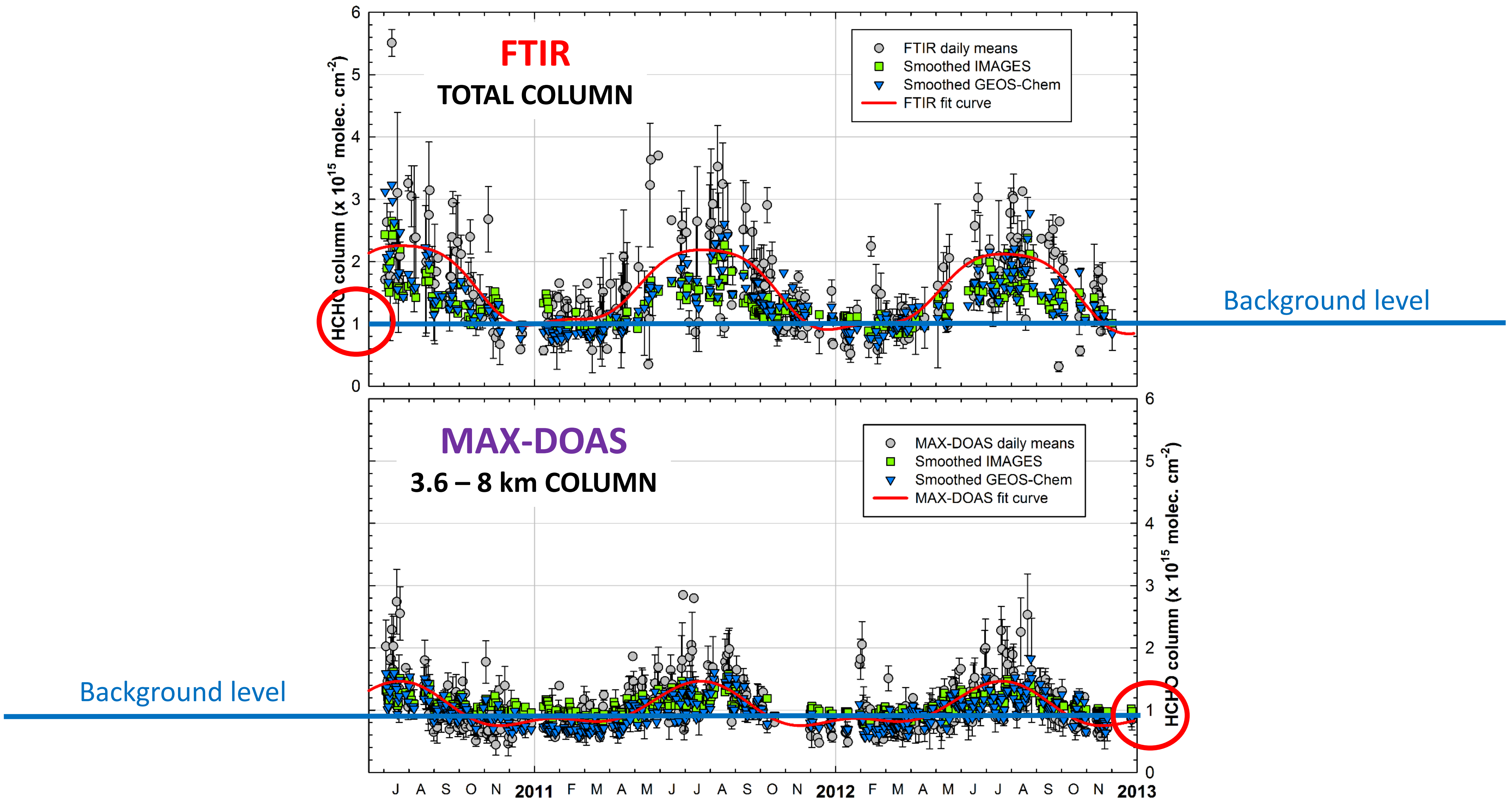


- Higher photochemical oxidation rate
- Biogenic NMVOCs

e.g., methanol (Bader et al., 2014, AMT)



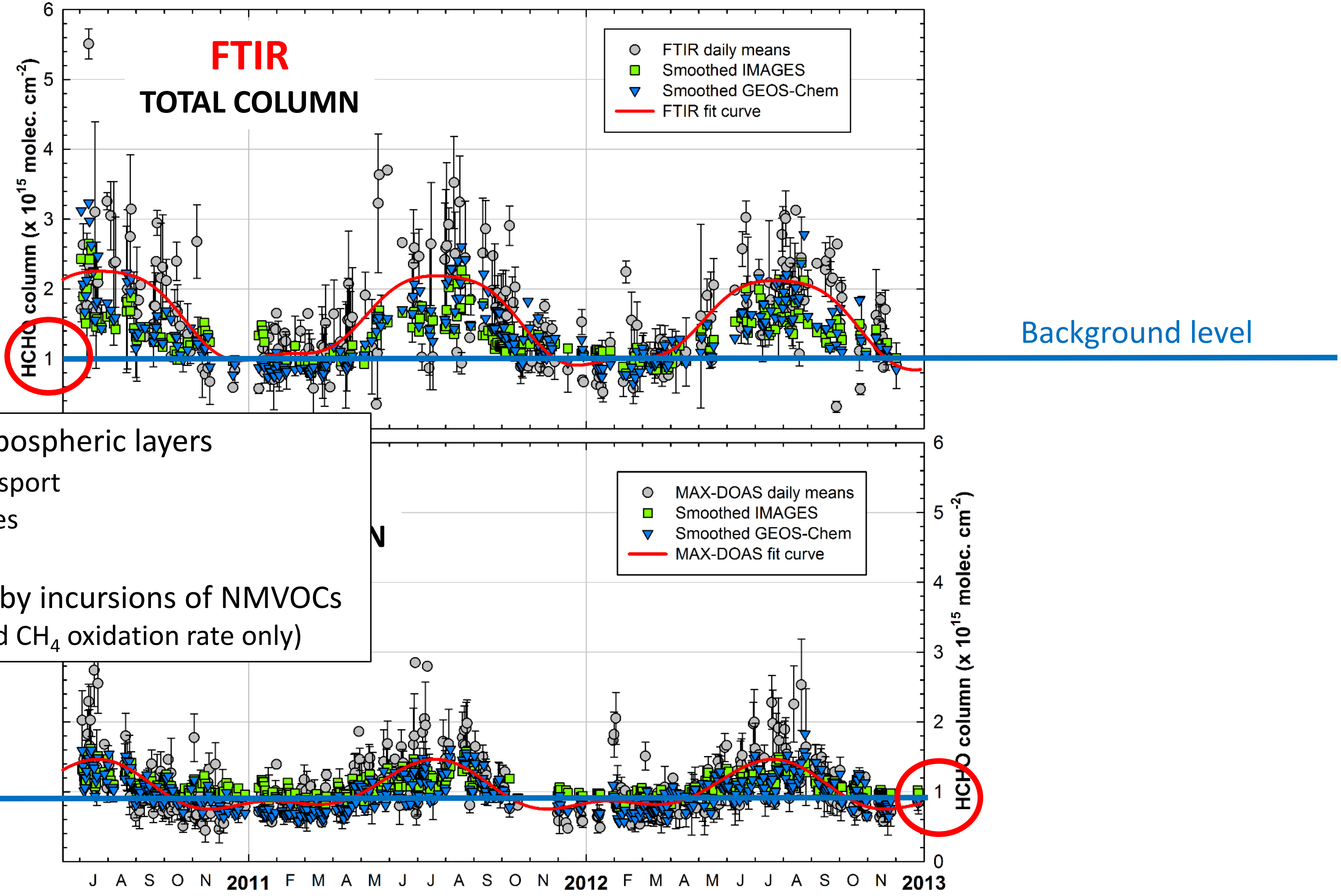
FTIR
TOTAL COLUMN
FTIR monthly mean
FTIR running mean
IMAGES
GEOS-Chem
HCHO column (x 10^15 molec. cm^-2)
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
Jan
Feb
Mar
Apr
May
Jun
Jul
Aug
Sep
Oct
Nov
Dec
MAX-DOAS
3.6 – 8 km COLUMN
MAX-DOAS monthly mean
MAX-DOAS running mean
IMAGES
GEOS-Chem


FTIR
TOTAL COLUMN
FTIR daily means
Smoothed IMAGES
Smoothed GEOS-Chem
FTIR fit curve
HCHO column (x 10^15 molec. cm^-2)
MAX-DOAS
3.6 – 8 km COLUMN
MAX-DOAS daily means
Smoothed IMAGES
Smoothed GEOS-Chem
MAX-DOAS fit curve
J A S O N 2011 F M A M J J A S O N 2012 F M A M J J A S O N 2013
Background level
Background level

- Sensitivity in the upper troposphere
- 25 % of the total column above 8 km

1. Variability in higher tropospheric layers
   - -> large-scale transport
   - -> convective fluxes

2. Seasonality also driven by incursions of NMVOCs (rather than photolysis and CH_4 oxidation rate only)

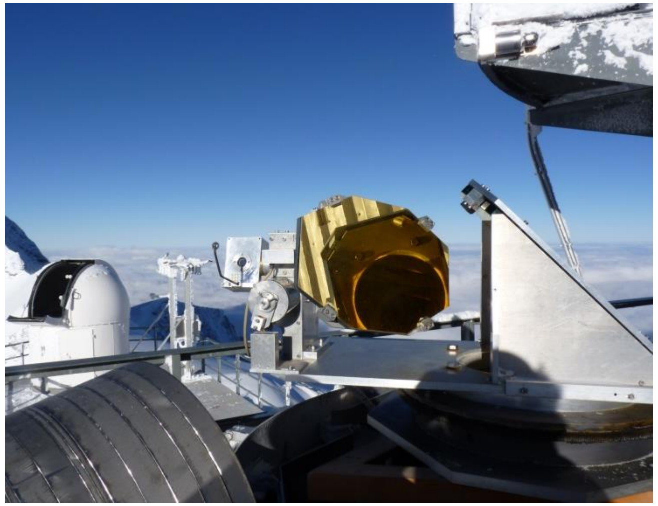


Conclusion

- HCHO amounts from ground-based FTIR and MAX-DOAS
- HCHO distributions from 3-D CTMs as intermediates

  FTIR and MAX-DOAS => **complementary** and **consistent**

- FTIR retrieval strategy available now at Jungfraujoch



Perspectives

- Contribution of the different NMVOCs to the HCHO formation
- Optimized FTIR retrieval strategy

  => multi-decadal observational time series

  - inter-annual variability
  - long-term trend
  - statistics for intra-day investigations

Thank you for your attention

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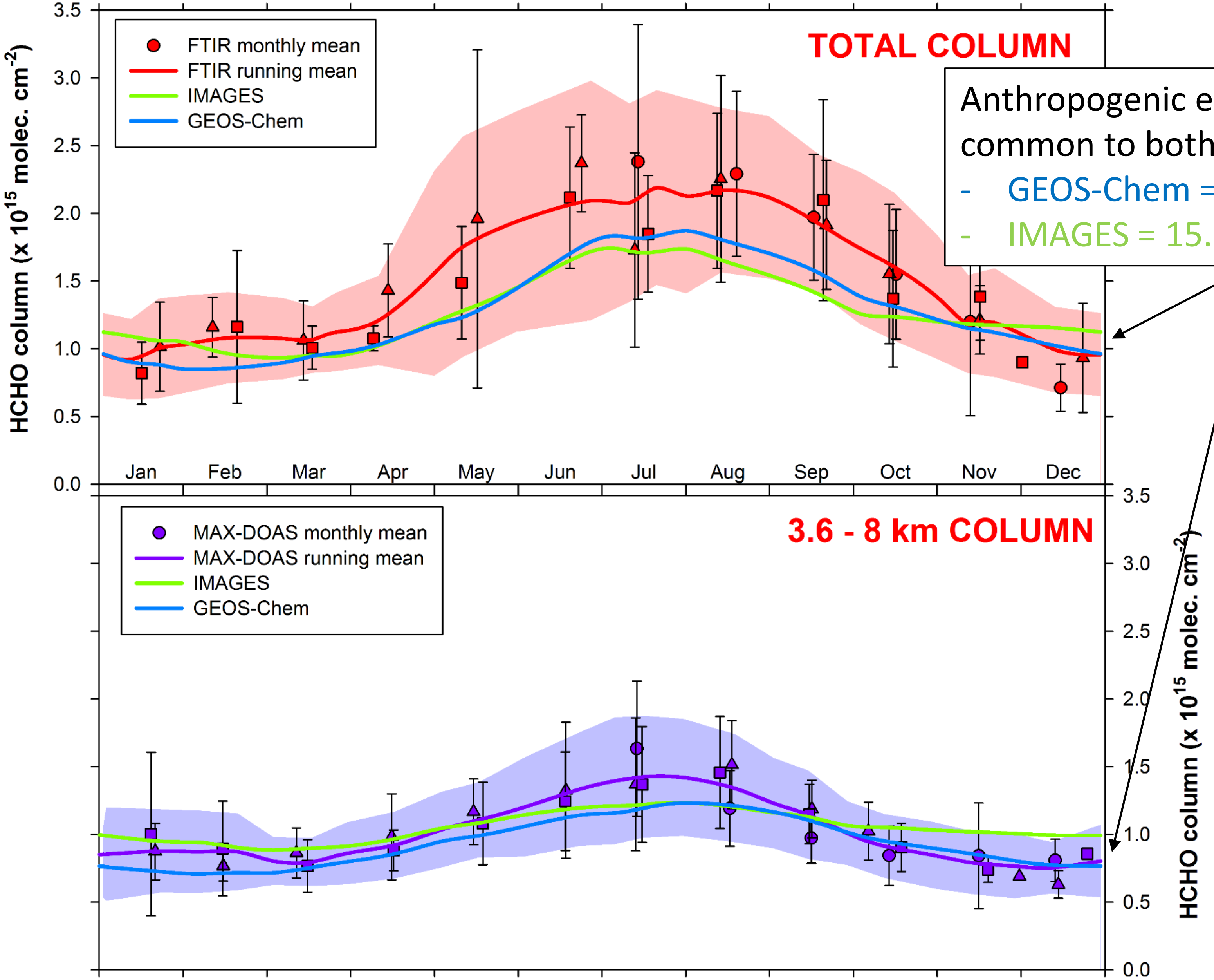
Anthropogenic NMVOCs over Europe for 2011:
- RETRO = 25.7 Tg
- EMEP = 10.3 Tg



Anthropogenic emissions of NMVOCs common to both CTMs:

- GEOS-Chem = 4.7 Tg C yr^{-1}
- IMAGES = 15.7 Tg C yr^{-1}

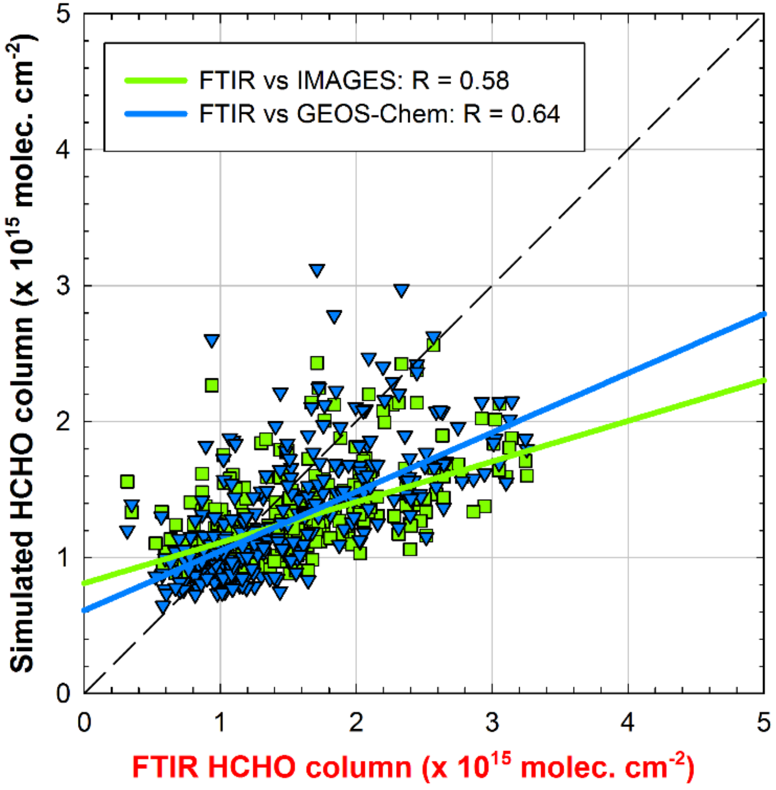
FTIR error budget

Error source	Error	Comments
Assumed variability	49.7%	WACCM variability relaxed, commensurate with ACE-FTS variability down to 6 km
Systematic errors		
Line intensity HCHO	9.7%	Assuming ±10 % uncertainties
Air-broadening coefficient HCHO	8.0%	Assuming ±10 % uncertainties
Line intensity interfering gases	5.2%	Assuming the maximal HITRAN 2008 uncertainties
ILS	2.5%	±10 % misalignment and instruments bias
Forward model	1.0%	Retrieval algorithm-related
HCHO a priori profile	3.0%	Assuming HCHO a priori profiles derived from ACE-FTS, IMAGES and GEOS-Chem
Total Systematic Error	**14.2%**	
Random errors		
Temperature profile	5.0%	±4 K around NCEP noon profile
H_2O and HDO a priori profiles	10.1%	Changes by a factor 2 in a priori slope
SZA	0.7%	Assuming ± 0.1° bias
Measurement noise	14.7%	
Smoothing	10.2%	
Model parameters	2.1%	
Total Random Error	**21.3%**	

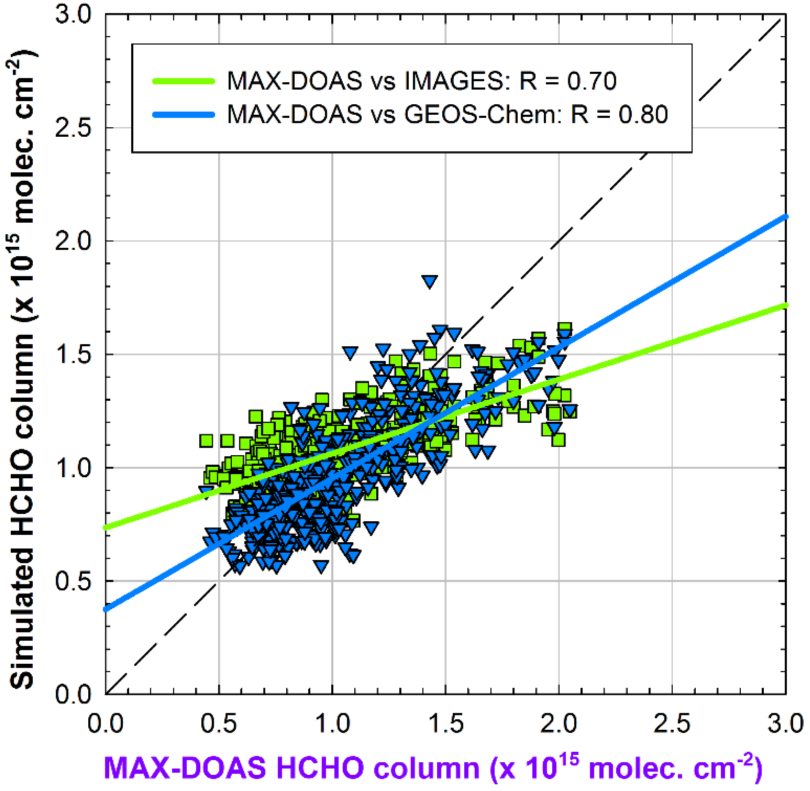
MAX-DOAS error budget

Error sources	Uncertainty on HCHO
Smoothing + noise errors	9.1%
Uncertainty related to aerosols	6.3%
Uncertainty related to the a priori	8.8%
Uncertainty related to the albedo	1.0%
Uncertainty on the HCHO cross sections	9.0%
Total uncertainty	**16.8%**

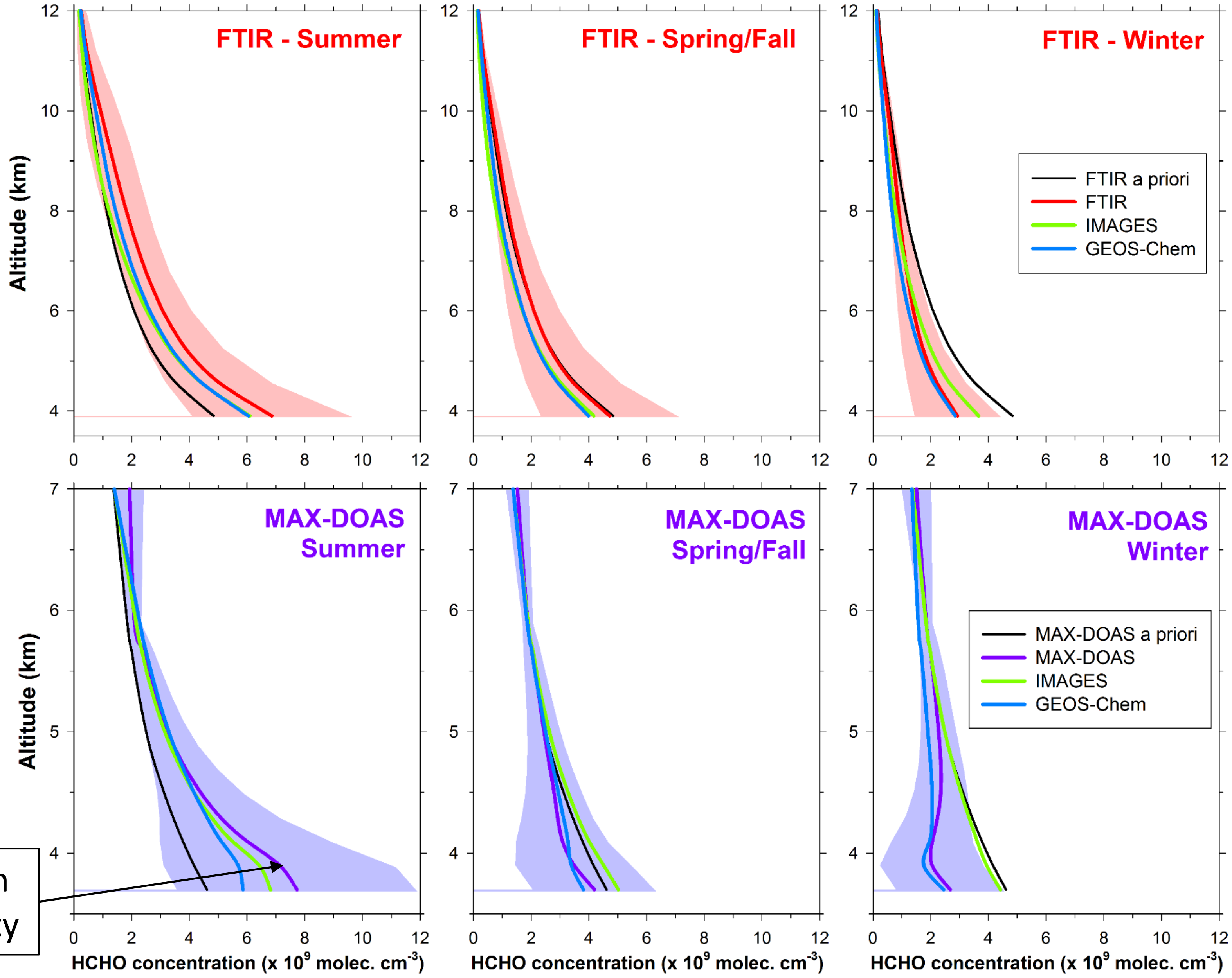
FTIR vs CTMs



MAX-DOAS vs CTMs



≈25 % of the total column above 8 km



Better representation of the HCHO variability