Parallel measurements of formaldehyde (H$_2$CO) at the Jungfraujoch station: Preliminary FTIR results and first comparison with MAXDOAS data

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1. INTRODUCTION

The Jungfraujoch station (Swiss Alps, 46.5° N, 8.0° E, 3580m a.s.l.)

- Site of the NDACC network
- Ground-based FTIR measurements performed year-round
- Using high-resolution FTIR spectrometer BRUKER 120HR
- Remote operation of the BRUKER instrument since late 2008
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- FTIR regular cloud-free observations since 1984, \(~120\) days \(\text{yr}^{-1}\) on average since the mid-1990s
- Grating spectra recorded from 1976 to 1989, covering narrow IR intervals

\[\Rightarrow\] more than 35 years of uninterrupted IR monitoring in the Alps
\[\Rightarrow\] more than two dozen stratospheric and/or tropospheric retrieved species
1. INTRODUCTION

Formaldehyde: $H_2CO$

- Sources → photochemical oxidation of $CH_4$ and other carbonic compounds
- Sinks → photolysis, oxidation by OH and deposition
- Lifetime → a few hours
- Absorption → very weak in IR domain ($< 1\%$)
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FTIR and MAX-DOAS observations of H$_2$CO at Réunion Island (21° S, 55° E)

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Largely influenced by biomass burning and fossil fuel combustion
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**Jungfraujoch station (46.5° N, 8.0° E, 3580m a.s.l.)**
2833.070 – 2833.350 cm$^{-1}$
Tunable optical filter improving the S/N ratio since 2005
2. RETRIEVAL STRATEGY

S/N = 3070

Broad bandpass optical filter
- from 2400 to 3000 cm$^{-1}$
- aperture = 1.1 mm
2. RETRIEVAL STRATEGY

**Broad bandpass optical filter**
- from 2400 to 3000 cm⁻¹
- aperture = 1.1 mm

**Narrow bandpass optical filter**
- from 2810 to 2850 cm⁻¹
- aperture = 1.45 mm

Enhanced S/N ratio
Simulation of formaldehyde window at the Jungfraujoch station
Jan 21st 2010 - SZA = 70°

Simulated total absorption < 1%
2. RETRIEVAL STRATEGY

Simulation of formaldehyde window at the Jungfraujoch station
Jan 21st 2010 - SZA = 70°

- Weak absorption
- Broad spectral line

Synthetic spectrum without H₂CO absorption
2. RETRIEVAL STRATEGY

Simulation of formaldehyde window at the Jungfraujoch station
Jul 11th 2010 - SZA = 80°

- Weak absorption
- Broad spectral line

Synthetic spectrum without H$_2$CO absorption
2. RETRIEVAL STRATEGY

H$_2$CO retrieval strategy at the Jungfraujoch station

- Retrieval with the SFIT-2 algorithm (v3.91)

- Microwindow: 2833.070 – 2833.350 cm$^{-1}$

- Spectral resolution: 0.005 cm$^{-1}$

- Interfering species: CH$_4$, HDO, O$_3$ and N$_2$O

- Spectroscopic parameters:
  - HITRAN 2004 for interfering species
  - HITRAN 2008 for H$_2$CO

- Data set: 2343 solar spectra from December 2005 to April 2013
2. RETRIEVAL STRATEGY

H$_2$CO PROFILES AT THE JUNGFRAUJOCH STATION

ALTITUDE (km)

H$_2$CO VOLUME MIXING RATIO (ppv)

RSD (km$^{-1}$)

WACCM v.6 simulations for Jungfraujoch (1980-2020)
2. RETRIEVAL STRATEGY

H₂CO PROFILES AT THE JUNGFRAUJOCH STATION

- WACCM v.6 simulations for Jungfraujoch (1980-2020)
- ACE-FTS v.3 36.5-56.5°N zonal average (2004-2012)
2. RETRIEVAL STRATEGY

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- Spectroscopic parameters:
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- Data set: 2343 solar spectra from December 2005 to April 2013
- A priori profile from WACCM v.6

Tikhonov type $L_1$ regularization

Constraint matrix $R = \alpha \ L_1^T \times L_1$
Determination of $\alpha$ (regularization strength) based on $\text{H}_2\text{CO}$ retrievals for 2010

**New subset:** spectra with «positive» retrieved profile only, when using $\alpha = 10$
2. RETRIEVAL STRATEGY

Simple scaling

Unphysical retrieved profile (%)

Alpha value

1e+0 1e+1 1e+2 1e+3 1e+4 1e+5

Global RMS (%)

1e+0 1e+1 1e+2 1e+3 1e+4 1e+5

H2CO total column (molec. cm⁻²)

1e+0 1e+1 1e+2 1e+3 1e+4 1e+5

Simple scaling

Simple scaling

Simple scaling
2. RETRIEVAL STRATEGY

- Fitting with Tikhonov regularization
- Scaling

α = 250
3. PRELIMINARY RESULTS

June 23rd 2010 – SZA = 77° - S/N = 17354
3. PRELIMINARY RESULTS

October 18th 2010 - SZA = 61° - S/N = 9701
3. PRELIMINARY RESULTS

H$_2$CO PROFILES AT THE JUNGFRAUJOCH STATION

- WACCM v.6 simulations for Jungfraujoch (1980-2020)
- ACE-FTS v.3 36.5-56.5°N zonal average (2004-2012)
- IMAGES simulations for Jungfraujoch (2010-2012)
- GEOS-CHEM simulations for Jungfraujoch (2007-2011)
- FTIR retrievals at Jungfraujoch (2005-2013)
3. PRELIMINARY RESULTS

EXAMPLE OF H₂CO EIGEN VECTORS AT JUNGFRAUJOCH

March 23rd 2010 spectra
SZA = 77°

July 7th 2010 spectra
SZA = 71°

DOFS = 1.533

DOFS = 1.355
3. PRELIMINARY RESULTS

TIME SERIES OF $\text{H}_2\text{CO}$ COLUMN ABOVE JUNGFRAUJOCH

FTIR retrieval from 2280 solar spectra
Daily and monthly averaged total columns
3. PRELIMINARY RESULTS

TIME SERIES OF $\text{H}_2\text{CO}$ COLUMN ABOVE JUNGFRAUJOCH

ACE-FTS occultation measurements
Partial columns above 6.5 km
3. PRELIMINARY RESULTS

**IMAGES model simulations**
Monthly average of daily columns above 3.5 km
3. PRELIMINARY RESULTS

TIME SERIES OF $\text{H}_2\text{CO}$ COLUMN ABOVE JUNGFRAUJOCH

GEOS-CHEM $4^\circ \times 5^\circ$ simulations
Monthly average of daily total columns
3. PRELIMINARY RESULTS

SEASONAL CYCLE OF H$_2$CO ABOVE JUNGFRAUJOCH

H$_2$CO ABUNDANCE (molec. cm$^{-2}$)

FTIR retrievals (2005-2013)
3. PRELIMINARY RESULTS

Assuming 3.5-6.5 km partial columns integrated from WACCM v.6
3. PRELIMINARY RESULTS

SEASONAL CYCLE OF H₂CO ABOVE JUNGFRAUJOCH

- FTIR retrievals (2005-2013)
- ACE-FTS occultations (2004-2012)
- IMAGES simulations (2010-2012)
3. PRELIMINARY RESULTS

SEASONAL CYCLE OF H$_2$CO ABOVE JUNGFRAUJOCH

![Graph of seasonal cycle of H$_2$CO abundance with different data sources and error bars for each month from January to December.]
Preliminary results from UV-visible MAX-DOAS observations
- Retrieval using a geometrical approximation

3. PRELIMINARY RESULTS

FTIR total columns appear to be low during summertime…
4. CONCLUSION AND DISCUSSION

**Forthcoming research and developments**

**Retrieval strategy with the 2833.070 – 2833.350 cm\(^{-1}\) microwindow**

- Spectroscopic parameters → contribution to the error budget?
  → HITRAN 2012 with updated H\(_2\)CO line parameters

- Two consecutive runs → first run for the interfering species
  → second run to fit H\(_2\)CO only

- Optimal Estimation Method instead of Tikhonov regularization?

**Retrieval strategy with 6 microwindows from Vigouroux et al., 2009?**

… but using broad-band spectra!
Thanks for your attention

Special thanks to Michel Van Roozendael, François Hendrick, Jenny Stavrakou, Isabelle De Smedt and Gaia Pinardi for the IMAGES and MAX-DOAS data
Investigating an Optimal Estimation Method for the H$_2$CO retrieval strategy

- Making use of climatological conditions
- Applying diagonal covariances deduced from ACE-FTS (v3) occultation measurements

Further experiments to fix the inter-layer correlation are still required
4. CONCLUSION AND DISCUSSION

Optimal Estimation Method for the H₂CO retrieval
3. PRELIMINARY RESULTS

H₂CO RETRIEVED PROFILES ABOVE JUNGFRAUJOCH FOR 2010

Tikhonov regularization - $\alpha = 25$

274 solar spectra
53 unphysical retrieved profiles (19.94 %)
3. PRELIMINARY RESULTS

H₂CO RETRIEVED PROFILES ABOVE JUNGFRAUJOCH FOR 2010

Tikhonov regularization - *alpha* = 250

- WACCM v.6 1980-2020 simulations average
- 274 solar spectra
- 8 unphysical retrieved profiles (2.92 %)