Retrievals of trichlorofluoromethane (CFC-11) and peroxyacetyl nitrate (PAN) from FTIR ground-based solar spectra and analysis of their long-time series above NDACC stations

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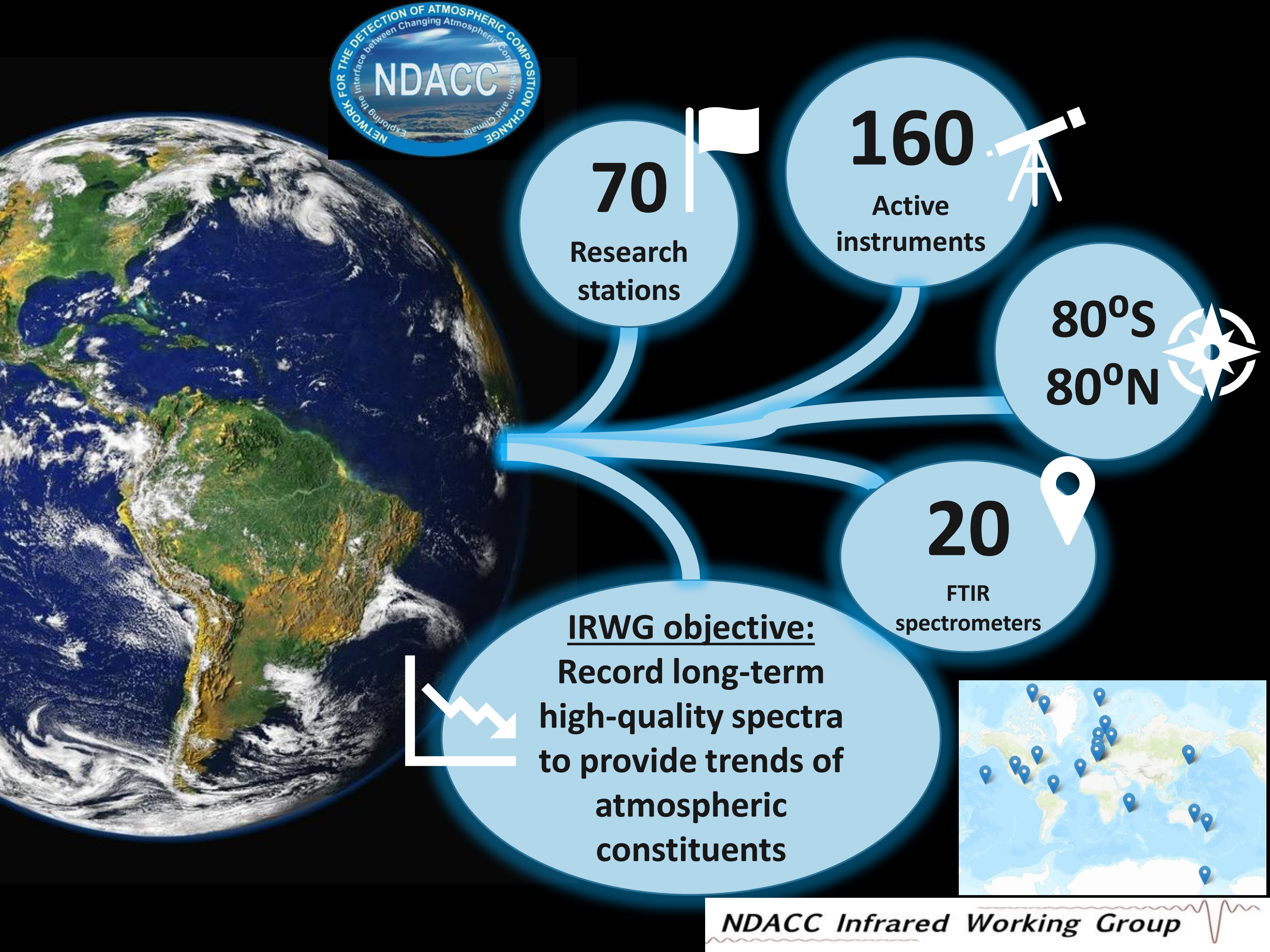
46th ACE Science Meeting (18th October 2022)


NDACC
70
Research stations
160
Active instruments
80°S
80°N
20
FTIR spectrometers
IRWG objective:
Record long-term high-quality spectra to provide trends of atmospheric constituents
NDACC Infrared Working Group

Jungfraujoch station (Swiss Alps)

Latitude: 46.55ºN
Longitude: 7.98ºE
Elevation: 3580 m a.m.s.l.

FTIR spectrometers

- 1984 – 2008: Homemade instrument
- 1994 – present: Commercial Bruker IFS-120HR improved by the team

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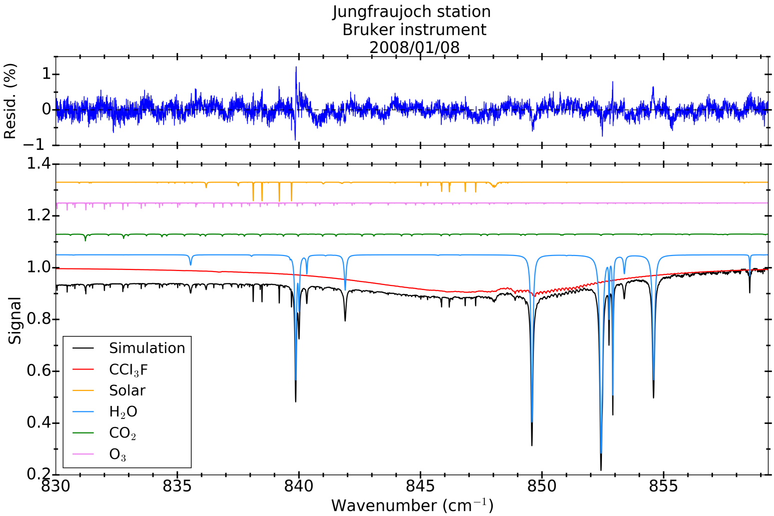
Introduction-Motivation

CFC-11

- Trichlorofluoromethane ($CC\ell_3F$, CFC-11)
- Destruction of the stratospheric O_3
- Anthropogenic origin $\longrightarrow$ Aerosol spray propellants, refrigerants, inflating agents, and solvents
- Montreal Protocol on Substances that Deplete the Ozone Layer (1987)
- CFC-11 concentration has declined since the late 1990s
- Slowing decline after 2012 (Montzka et al., 2018 - Nature)
- Delay in ozone recovery and in reduction of its contribution to radiative forcing
- Requested contribution for the UNEP report to investigate the CFC-11 trend and evolution

IR Spectra

CFC-11



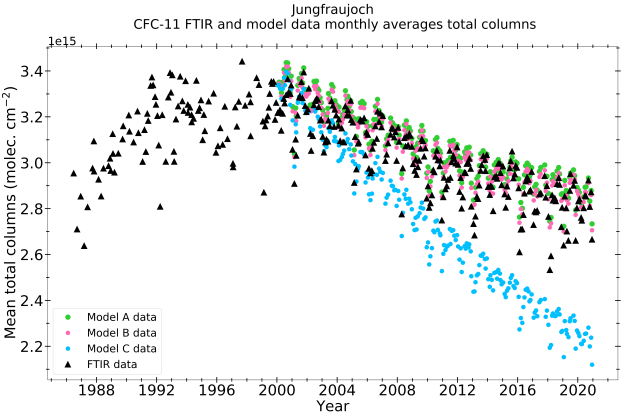
Instruments harmonisation

Jungfraujoch

- Detector change → JJB multiplied by 0.9692
  from 12/02/1999 to 09/10/2001
- Instrument change → JJL multiplied by 0.9467
- Change of one spectrometer mirror → 2016 – 2019 period data multiplied by 1.01812985
- **Harmonised series from June 1986 to Dec 2020**

Instruments harmonisation

Jungfraujoch



Model tracers:

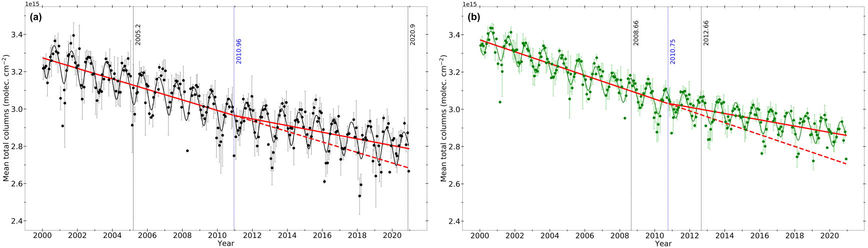
(A) Best estimate of emissions and some realistic distribution

(B) Same total emissions as (A), but equal emissions at all lat/lon

(C) Zero emissions since 2000 - Simple decay

Model and FTIR data comparison

Northern Hemisphere

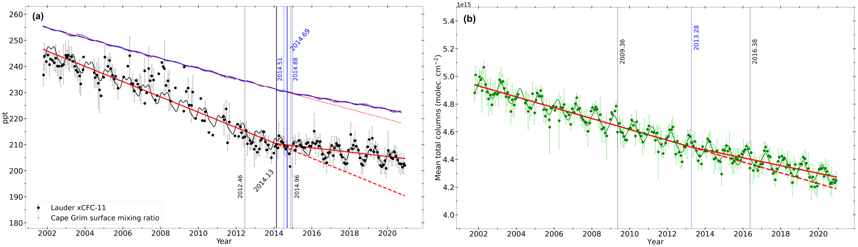


	Break location	2000 – Break	Break – 2020
CFC-11 FTIR	2010.96	-0.95 ± 0.13	-0.61 ± 0.15
CFC-11 TOMCAT	2010.75	-1.03 ± 0.08	-0.55 ± 0.09
CFC-11 ACE (global)*	2012.0	-0.8	-0.5

* Report on the Unexpected Emissions of CFC-11, 2021

Model and FTIR data comparison

Southern Hemisphere



	Break location	2001 – Break	Break – 2020
In situ CG	2014.69	-0.82 ± 0.01	-0.52 ± 0.01
xCFC-11 FTIR	2014.13	-1.34 ± 0.08	-0.39 ± 0.10
CFC-11 TOMCAT	2013.28	-0.86 ± 0.05	-0.61 ± 0.07
CFC-11 ACE (global)*	2012.0	-0.8	-0.5

* Report on the Unexpected Emissions of CFC-11, 2021

Conclusions

- Harmonised JFJ data set from June 1986 to December 2020 → NEW!
- Lauder CFC-11 time series → NEW!
- Comparison to *in situ* observations and model simulations
- Model JFJ data: break point in ≈2011
- Model LAU data: break point in ≈2014
- Concordance with ACE global trends
- Unreported emissions (ex. from eastern China (Rigby et. al, 2019))
- Decrease in global CFC-11 emissions since 2018 (Montzka et al., 2021 & Park et al., 2021)
- FTIR: complementary monitoring to surface-measurement networks

Pardo Cantos et al., 2022 (doi: 10.1039/d2ea00060a)

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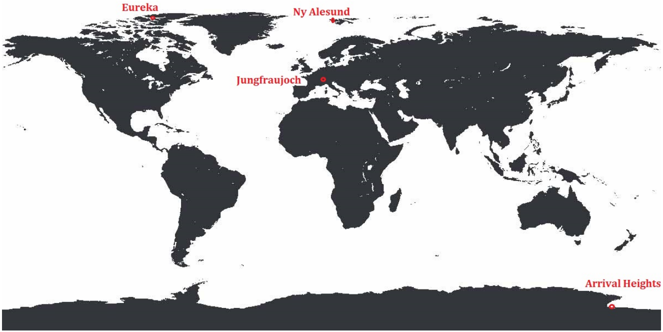
Introduction

PAN

- Peroxyacetyl nitrate ($CH_3COO_2NO_2$, PAN)
- Main tropospheric reservoir of NO_x ($NO + NO_2$)
- Anthropogenic and natural sources $\rightarrow$ fossil fuel combustion, biomass burning, lightning, NMVOCs emissions
- NO_x can travel far from the regions of emission $\rightarrow$ Formation of tropospheric ozone $\rightarrow$ Damages air quality
- FTIR total columns compared with a GEOS-Chem simulation and with total columns from IASI satellite observations

NDACC stations

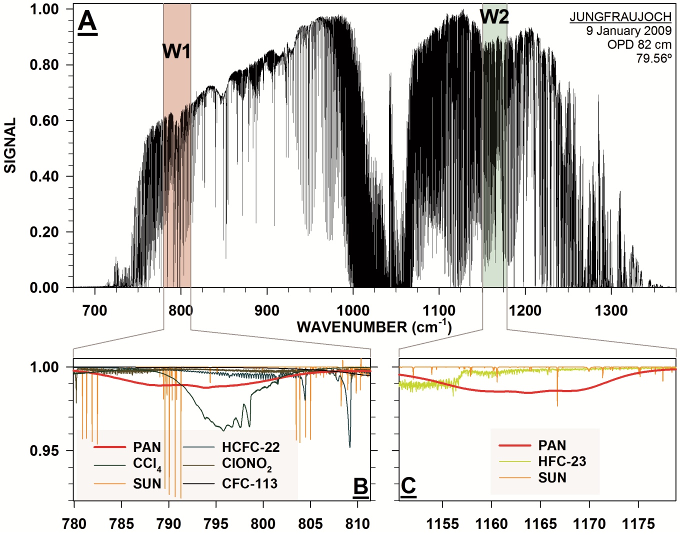
FTIR observations



Site	Latitude	Longitude	Altitude (m a.s.l.)	Team and Reference
Eureka (Canada)	$80.05°N$	$86.42°W$	610	University of Toronto (Batchelor et al., 2009)
Ny Ålesund (Norway)	$78.92°N$	$11.93°E$	24	University of Bremen (Notholt et al., 1997)
Jungfraujoch (Switzerland)	$46.55°N$	$7.98°E$	3,580	University of Liège (Zander et al., 2008)
Arrival Heights (Antarctica)	$77.83°S$	$166.67°E$	184	NIWA (Wood et al., 2002)

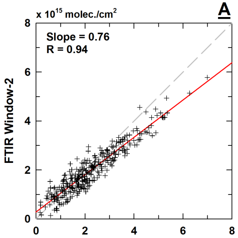
IR Spectra

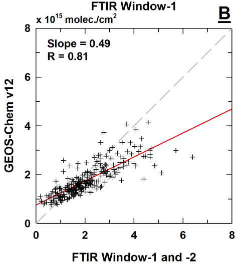
PAN

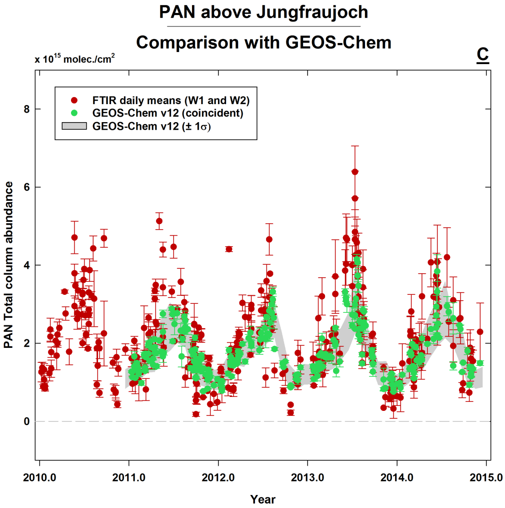


Model and FTIR data comparison

Jungfraujoch Station

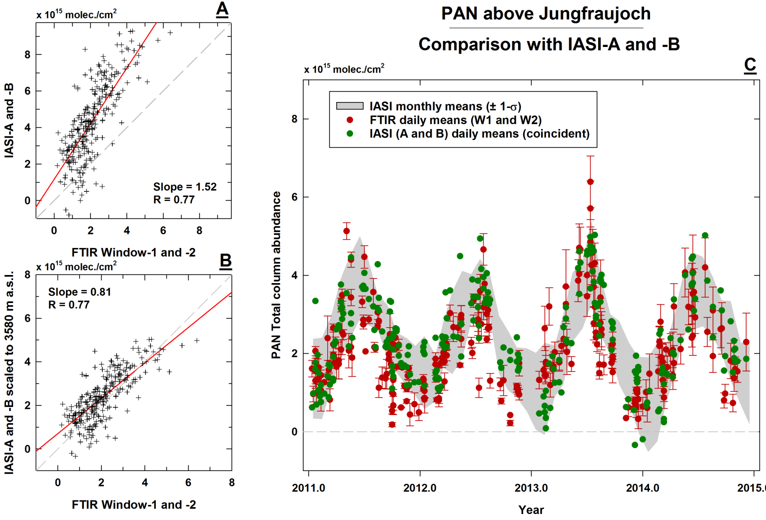






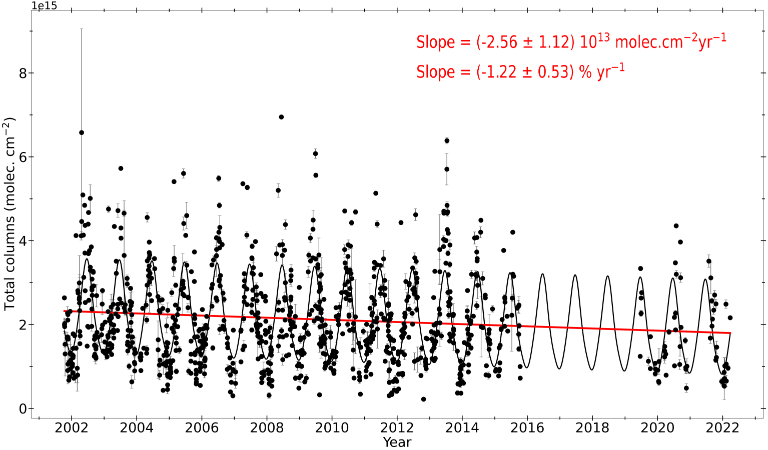
Satellite and FTIR data comparison

Jungfraujoch Station



Trend analysis

Jungfraujoch Station



Conclusions

- First retrievals of PAN from ground-based FTIR solar spectra
- Absorption features are broad, unstructured, and weak
- Total columns: essentially troposphere
- Seasonal modulation: minimun in winter and maximum in summer
- Consistent with global GEOS-Chem simulation and IASI observations
- Prospects for the production of global multidecadal time series of PAN

Mahieu et al., 2021 (doi: 10.1525/elementa.2021.00027)

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