Atmospheric Remote Sensing and Molecular Spectroscopy (Vietnam School of Earth Observation)

Quy Nhon, Vietnam, 26-31 August 2018

Ground-based FTIR measurement technique for the monitoring of atmospheric pollutants and greenhouse gases

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Station QualAir Jussieu

Introduction

Importance of the atmosphere

Why studying the atmosphere ?

where aircrafts fly

- air transport
- military applications
- space rocket

scientific research

where humans live

- meteorology / climate
- air quality
- impact on the agriculture, economy, …

Monitoring of the atmosphere

(pollution, climate, "ozone hole", greenhouse gases)

for

Understand and model atmospheric Physics and Chemistry processes

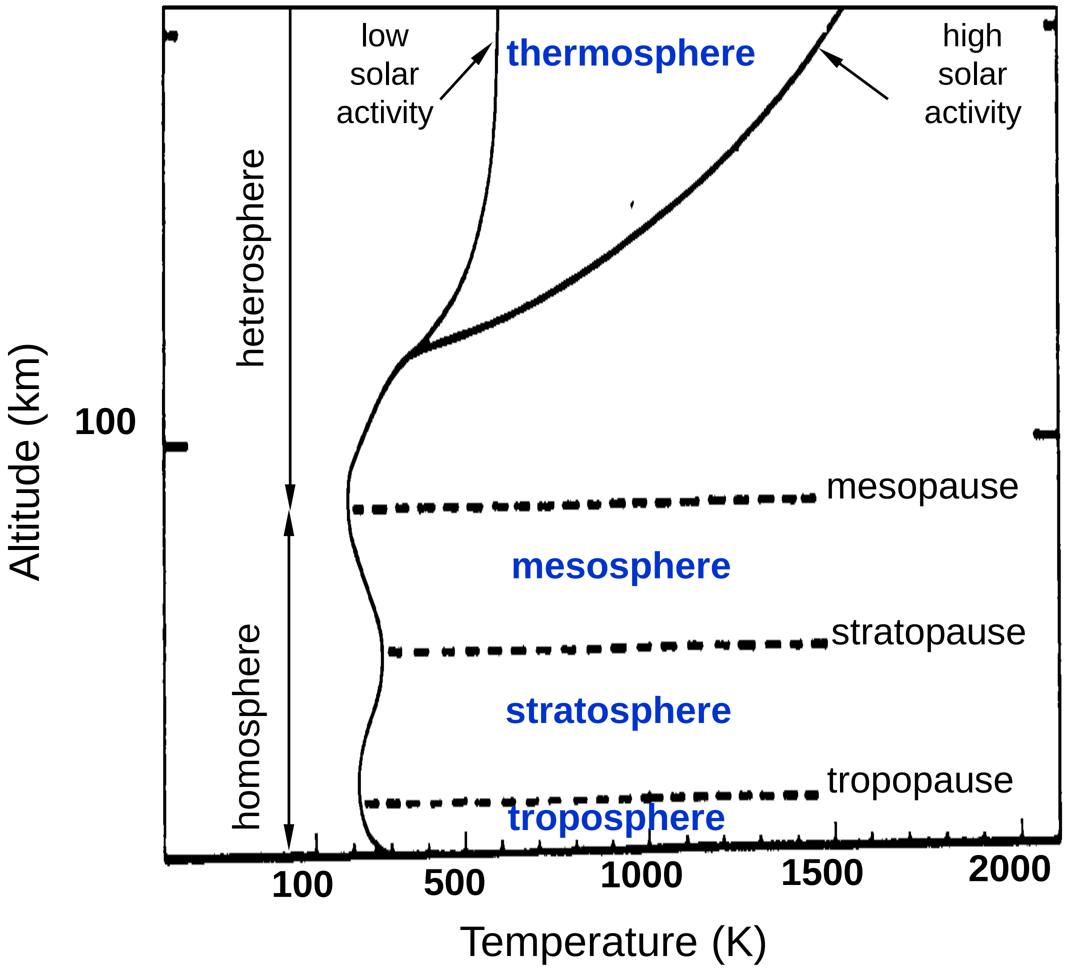
(natural variability and equilibrium, impact of **anthropogenic activities**)

need

Set-up innovating instrumentation for atmospheric observation

Atmosphere

◆ Atmospheric layers
(following temperature gradient)



- **Troposphere :**
  - from ground to ~ 12 km
  - $\dfrac{dT}{dz} < 0$

  (low atmosphere)

- **Stratosphere :**
  - from 12 to ~ 50 km
  - $\dfrac{dT}{dz} > 0$

  (middle atmosphere)

Why studying the stratosphere ?

- **Stratosphere** = layer between between the tropopause (~ 12 km) and the stratopause (~ 50 km height)

↓

where the ozone layer (maximum) is located (solar UV absorption)

↓

Study the O_3 production/destruction processes ("**ozone hole**")

Instruments for atmospheric observation :
ILAS, **GOME**, **MIPAS**, **ACE-FTS**,
LPMA (balloon), **Dobson** (ground)

Why studying the troposphere ?

◆ **Troposphere** = layer between the ground and about ten km height

↓

Zone affecting directly humans
(anthropogenic activity and natural variability influence)

Tropospheric pollution
CO, O_3, hydrocarbons, ...
(toxic and harmful atmospheric species)

Meteorology

Transport & dynamics

Greenhouse effect
CO_2, CH_4, ...
(global warming, impact on the climate)

- On-board instruments : IASI, **IASI-balloon** (balloon), AERI-X (aircraft)
- Ground-based FTIR instrument : **FTS-Paris** (**F**ourier **T**ransform **S**pectrometer)

Urban air pollution

➔ **Anthropogenic emissions are stronger in large agglomerations introducing many air pollution events**

➔ **'Great Smog' of London in 1952**

⇒ Winter pollution of SO_2 (+500%)

⇒ Related to mortality (4000 dead in 5 days)

➔ **With the increase of anthropogenic emissions due to the industrialization and the beginnings of car industry, more and more legislations appeared**

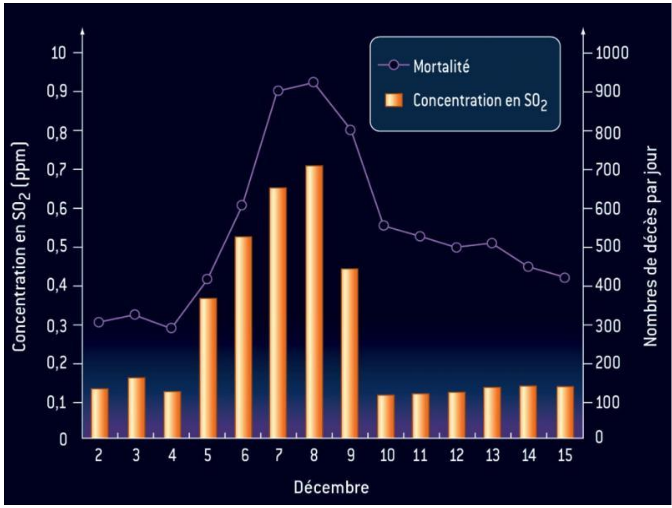
- European directive limiting SO_2 emission in 1980

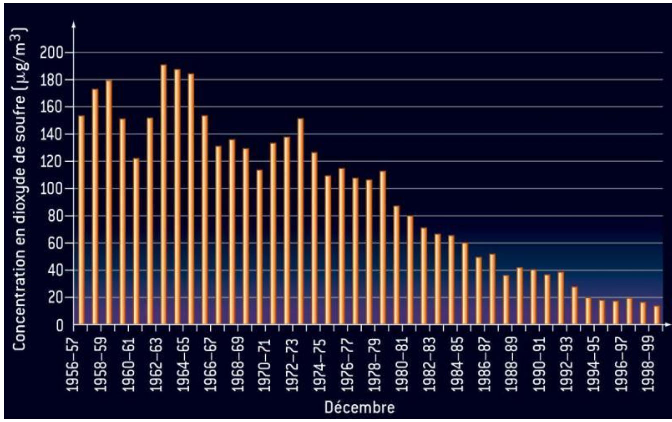
⇒ Reduction of 10 times over 40 years

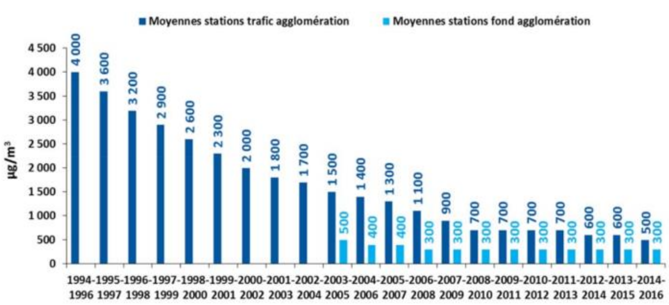
- European directive limiting CO emission in 2000

⇒ Reduction of 10 times since 2000

How about the other trace gases or pollutants ?







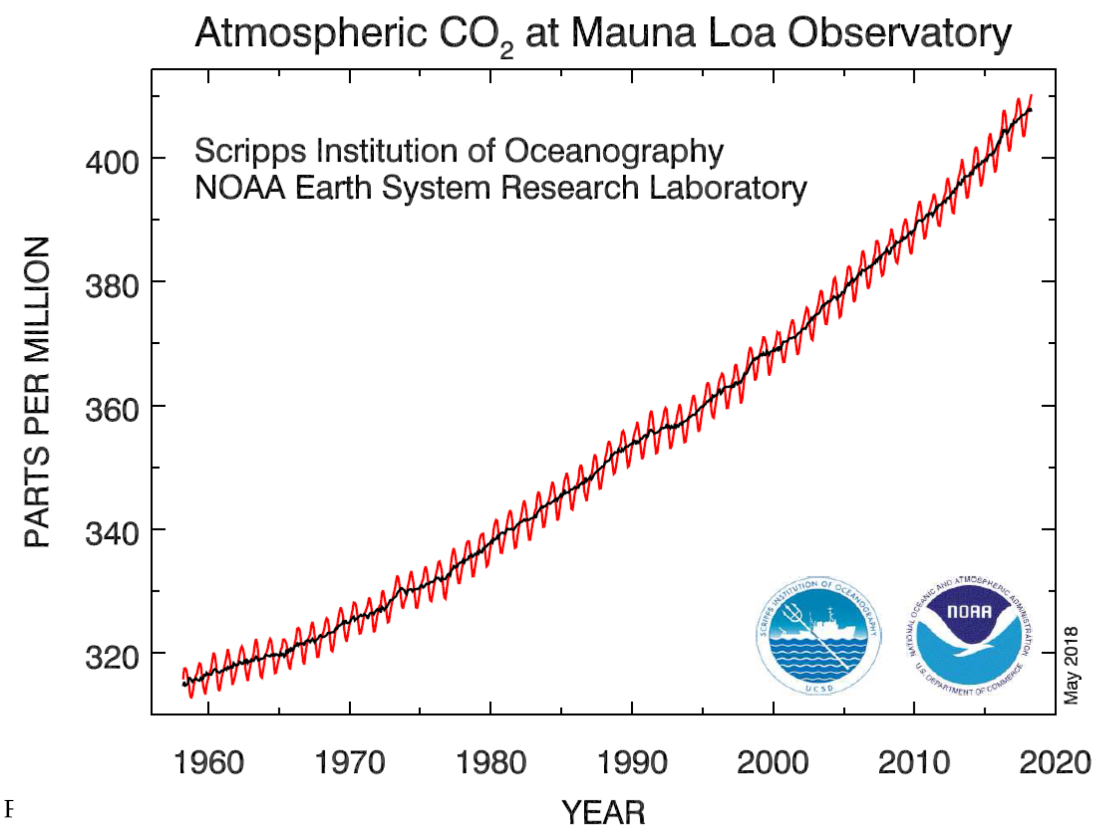
Greenhouse gas monitoring

- **World of 7 billion inhabitants**
- **Increasing need of energy**

⇨ **Anthropogenic emissions of greenhouse gas in constant increase**

These emissions are responsible of the observed global climate warming (GIEC)

It is essential to quantify precisely their source and loss



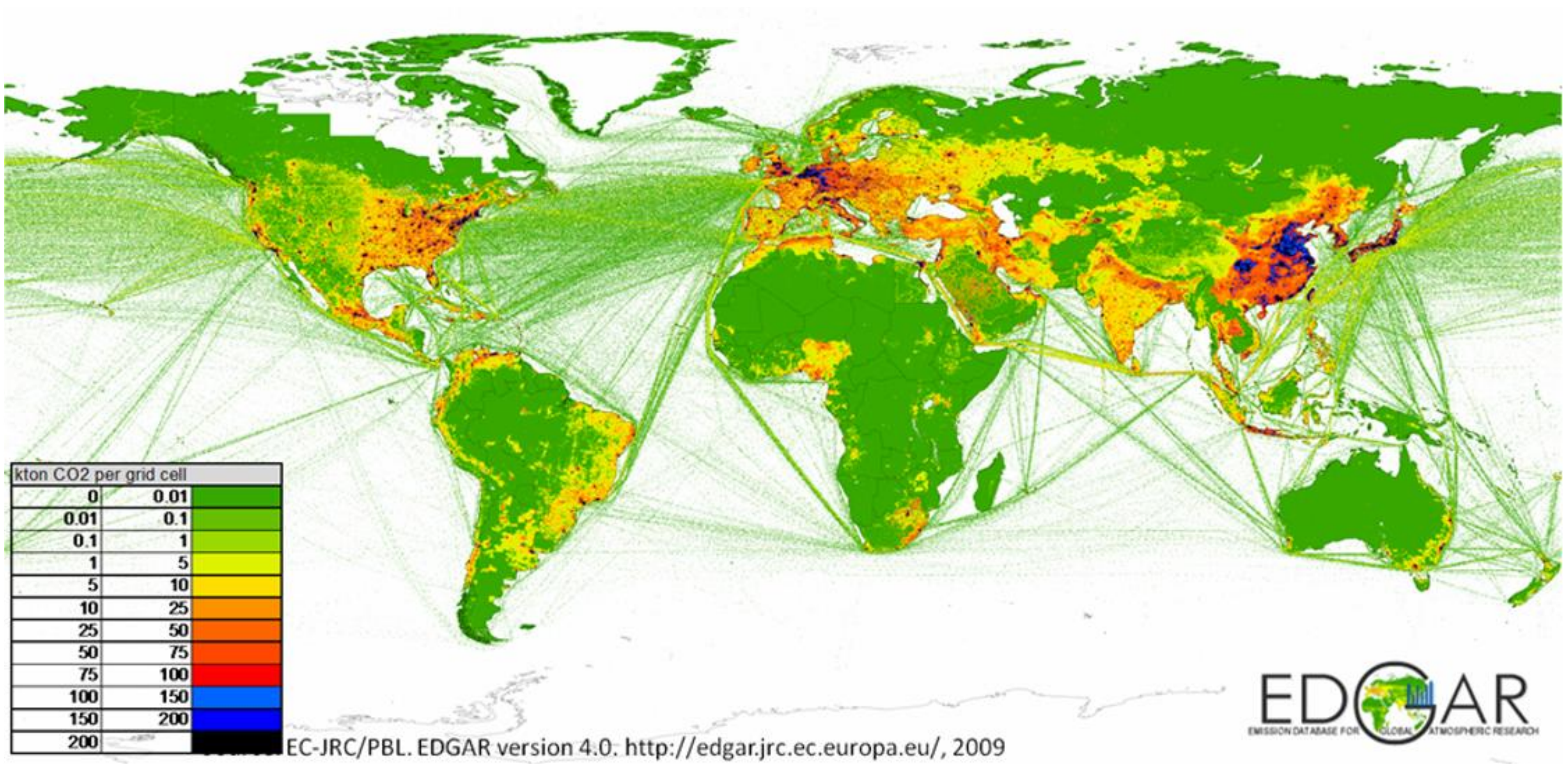
Global emissions of CO_2

➔ 70% of fossil CO_2 emissions are located in urbanized areas

➔ Installation of the measurements close to the emission sources

➔ Development of ground-based networks : ICOS, TCCON, …

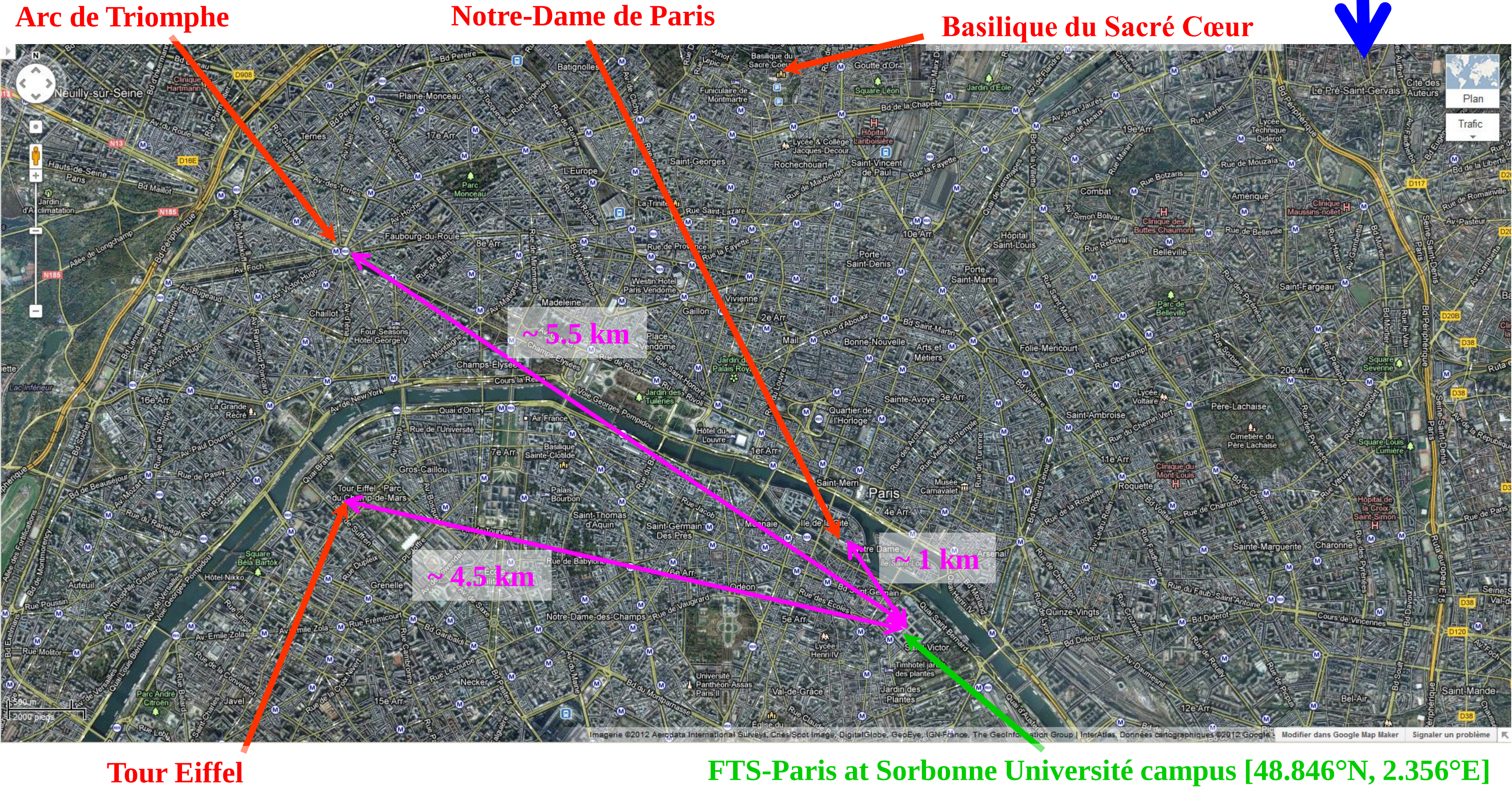
➔ Development of space missions : SCIAMACHY, GOSAT, OCO-2, TanSat, GOSAT-2, MicroCarb, MERLIN, OCO-3, …



Description of the LERMA Fourier Transform Spectrometer (FTS-Paris)

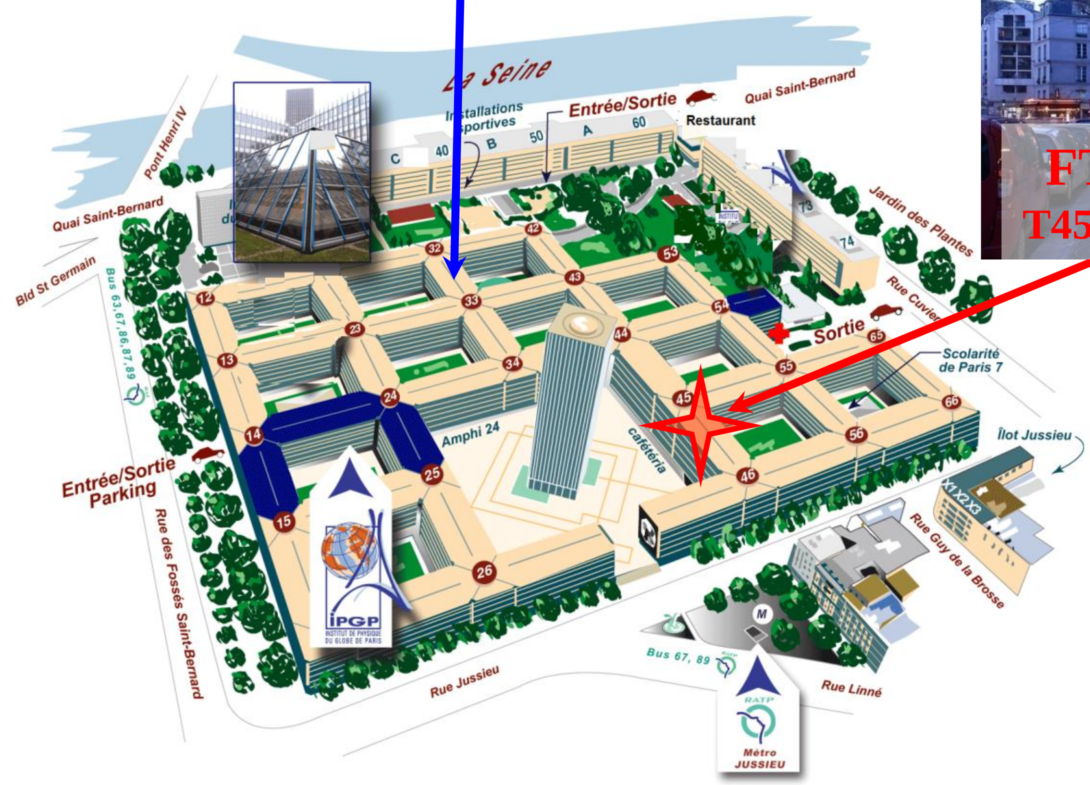
LERMA ground-based FTS at Paris megacity

- 3rd European largest megacity
- More than 2 million inhabitants in the city of Paris
- More than 10 million inhabitants in the Paris urban area

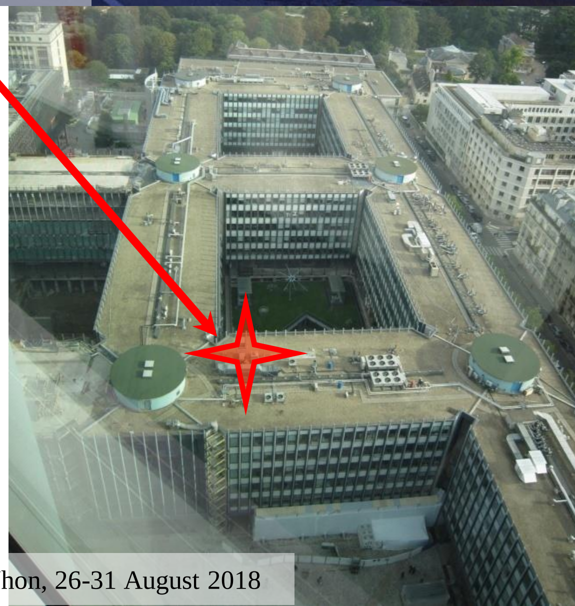


TCCON-Paris : the first TCCON site in an European megacity

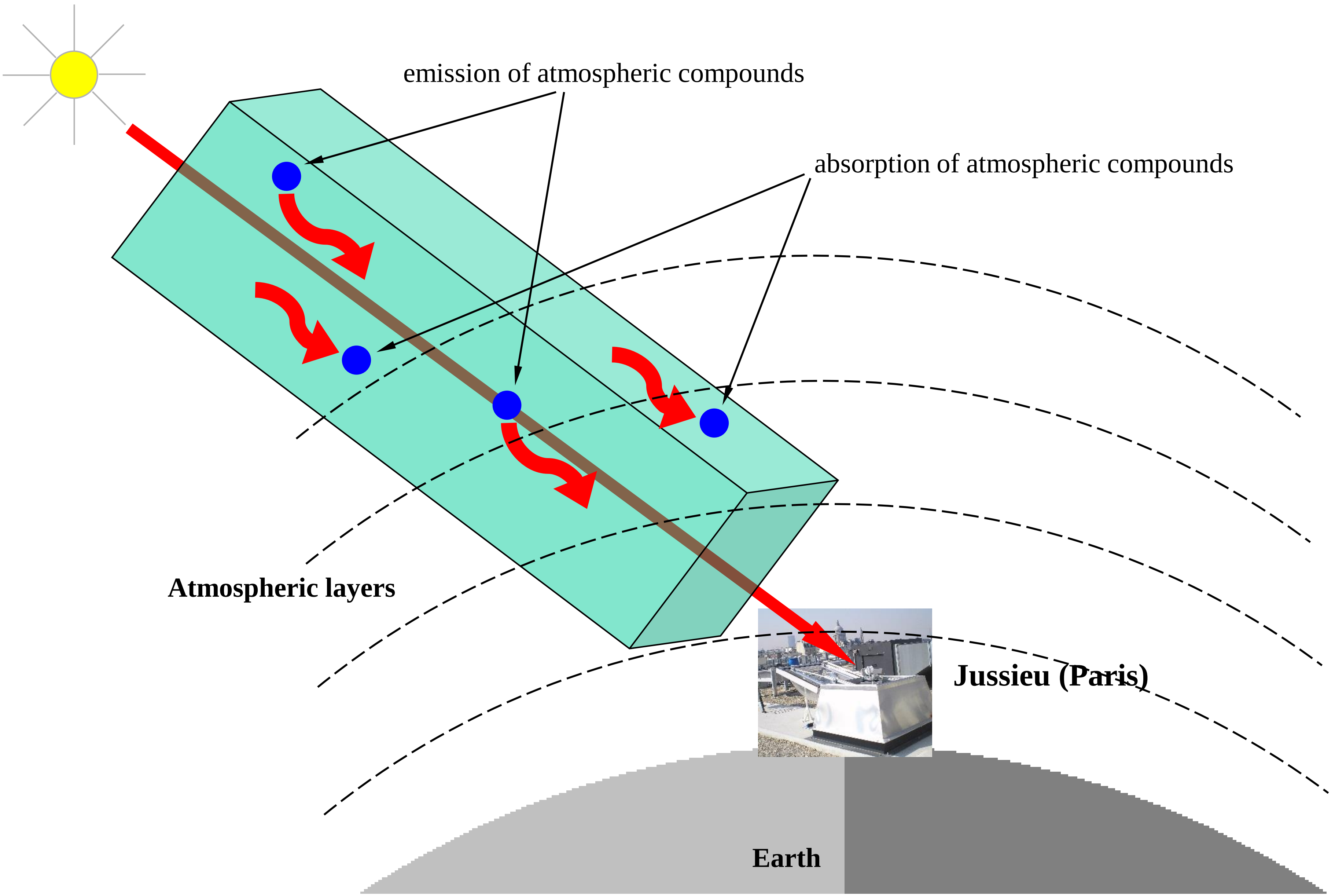
TASQ team
from LERMA-Jussieu
T32-33, Floor 2

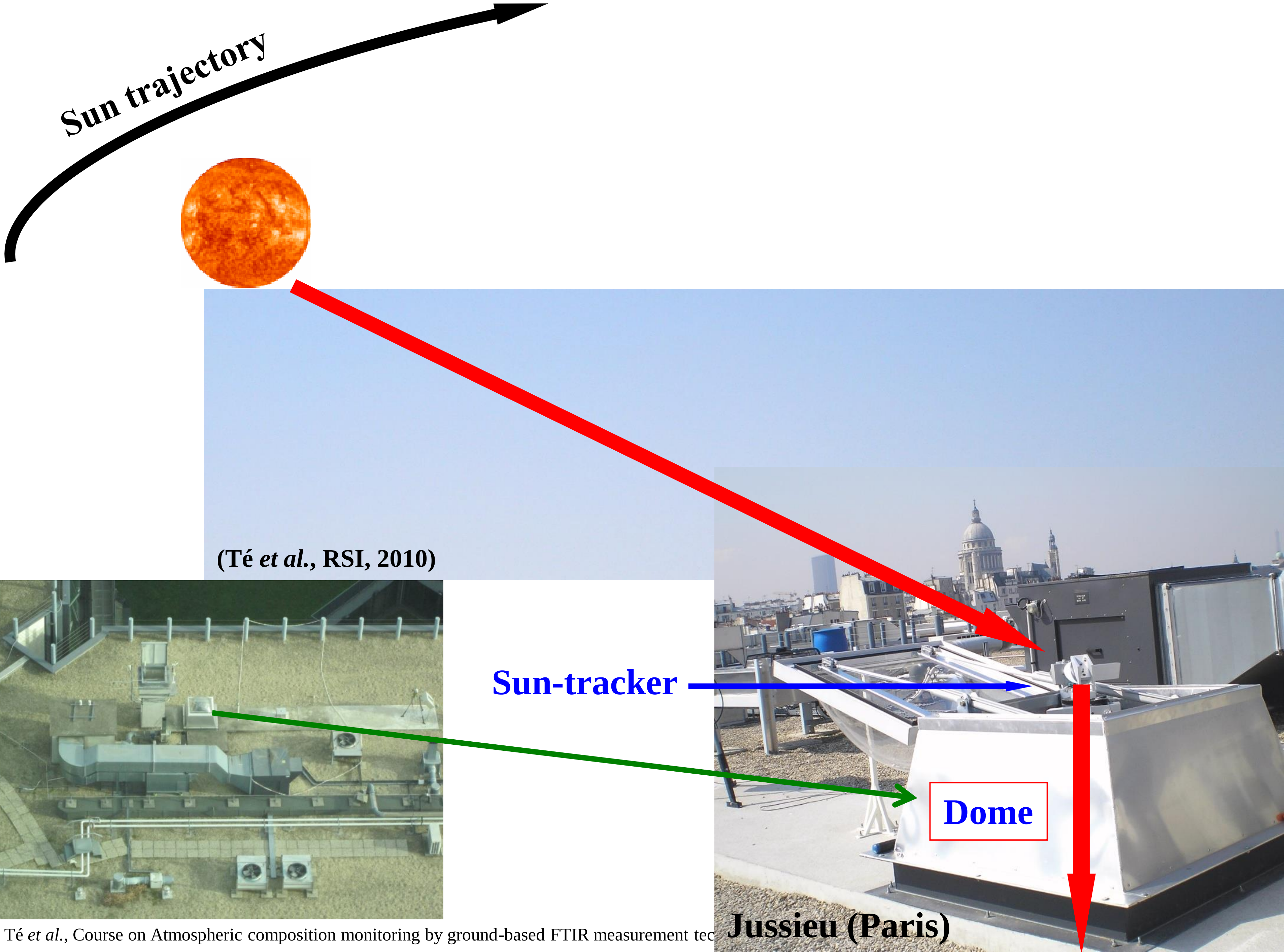


FTS-Paris
T45-46, Floor 5

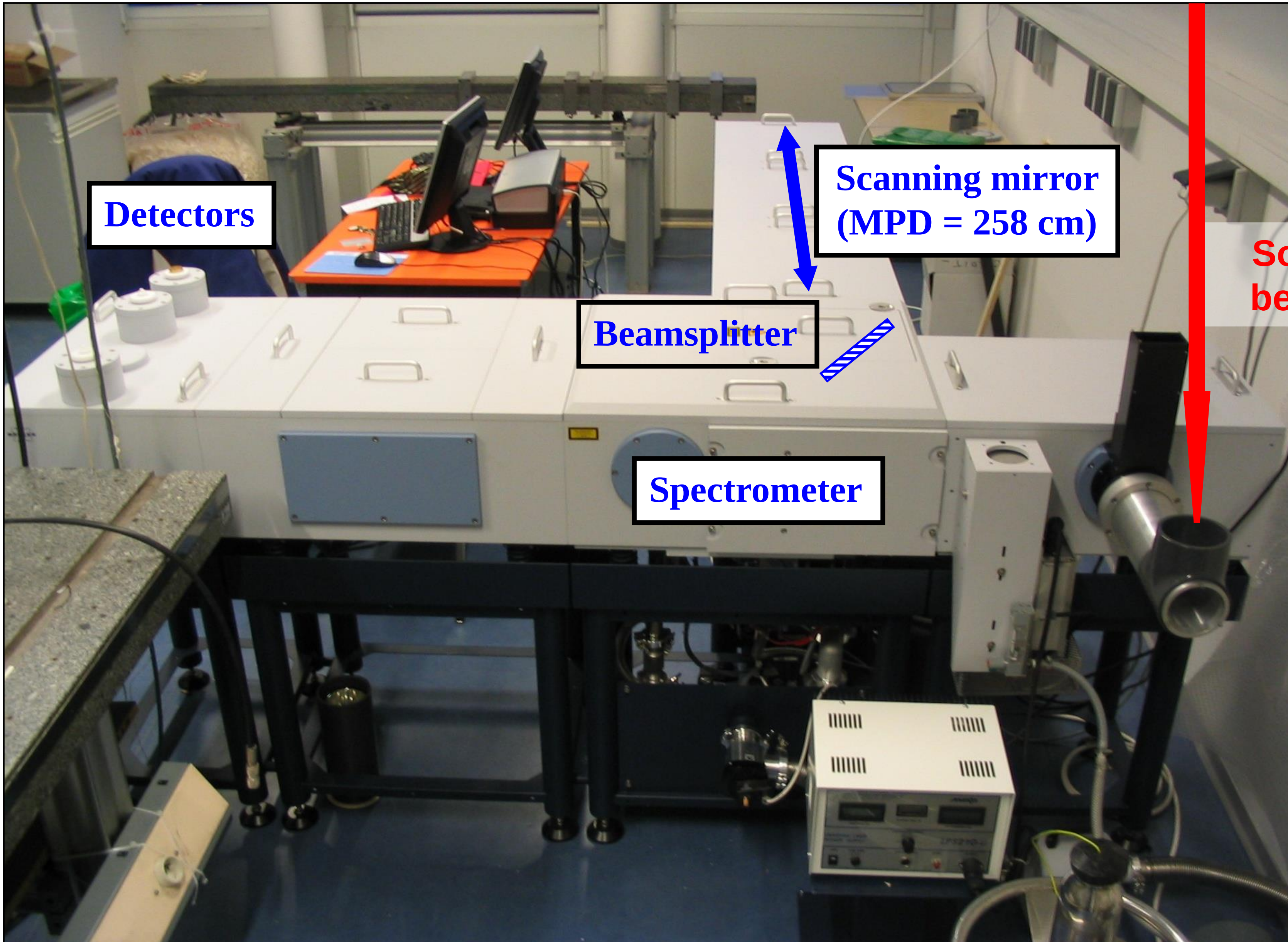


Remote sensing in solar absorption configuration

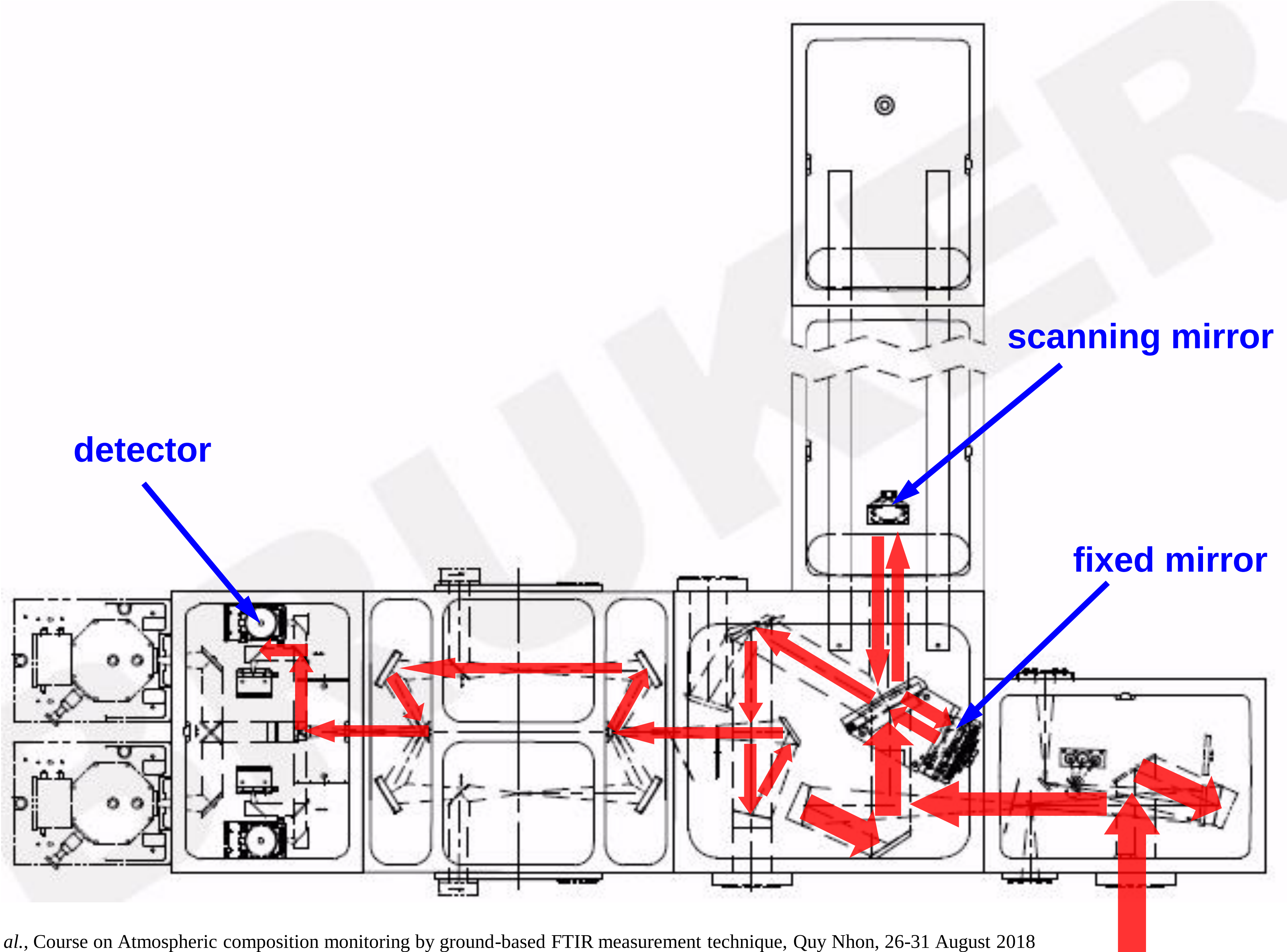



Sun trajectory
(Té et al., RSI, 2010)
Sun-tracker
Dome
Jussieu (Paris)

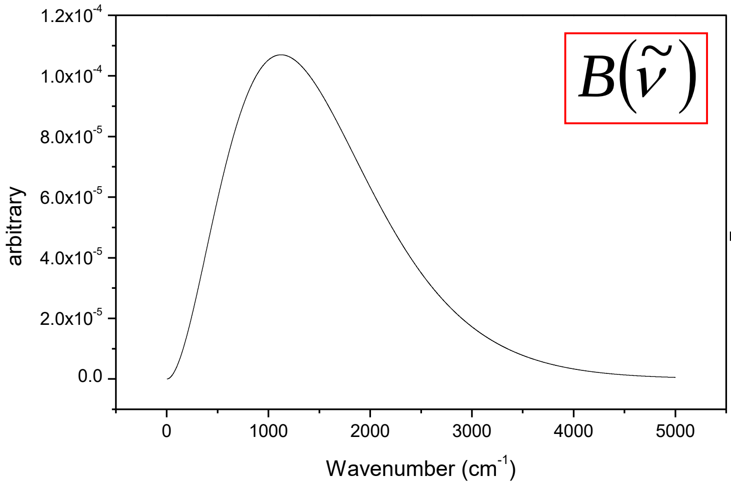
The LERMA FTS-Paris installed @ Jussieu



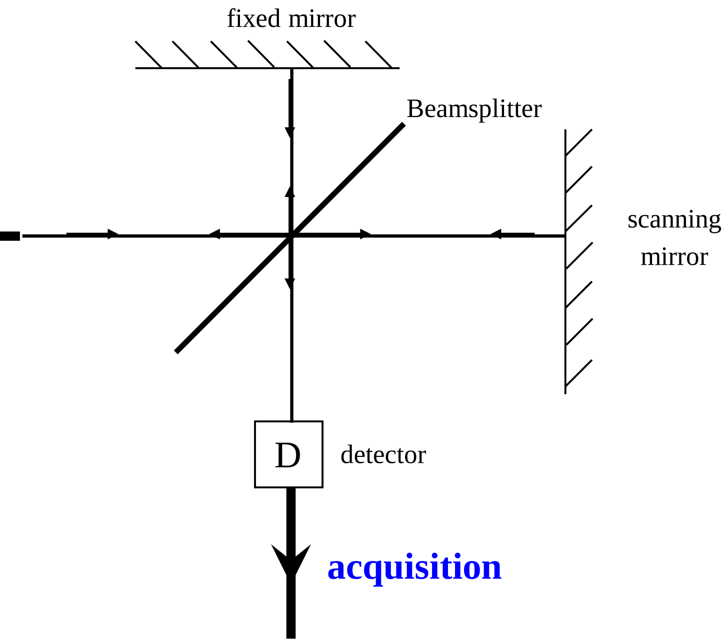
FTS-Paris optical schematic



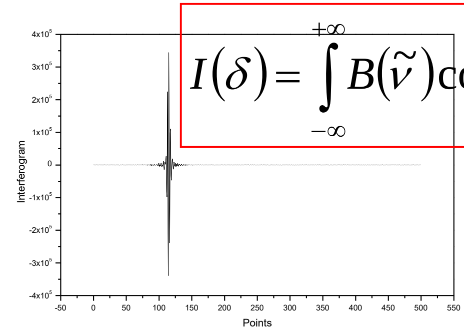
Data recorded by a Michelson interferometer



incident signal



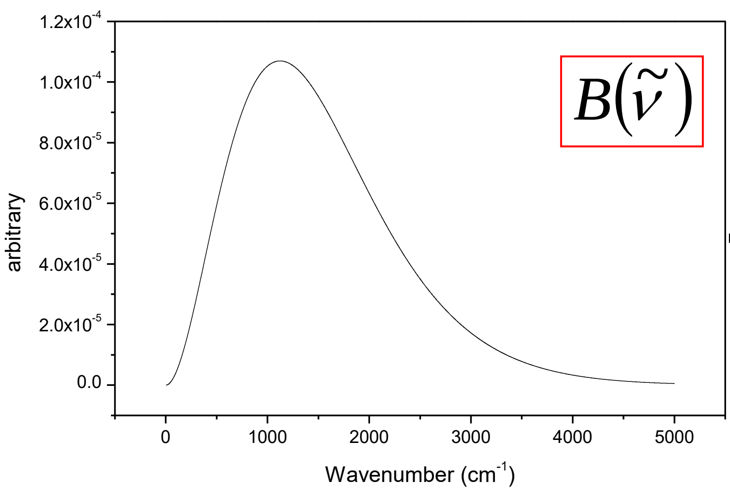
acquisition



$$I(\delta) = \int_{-\infty}^{+\infty} B(\tilde{\nu}) \cos(2\pi\tilde{\nu}\delta)\, d\tilde{\nu}$$

interferogram

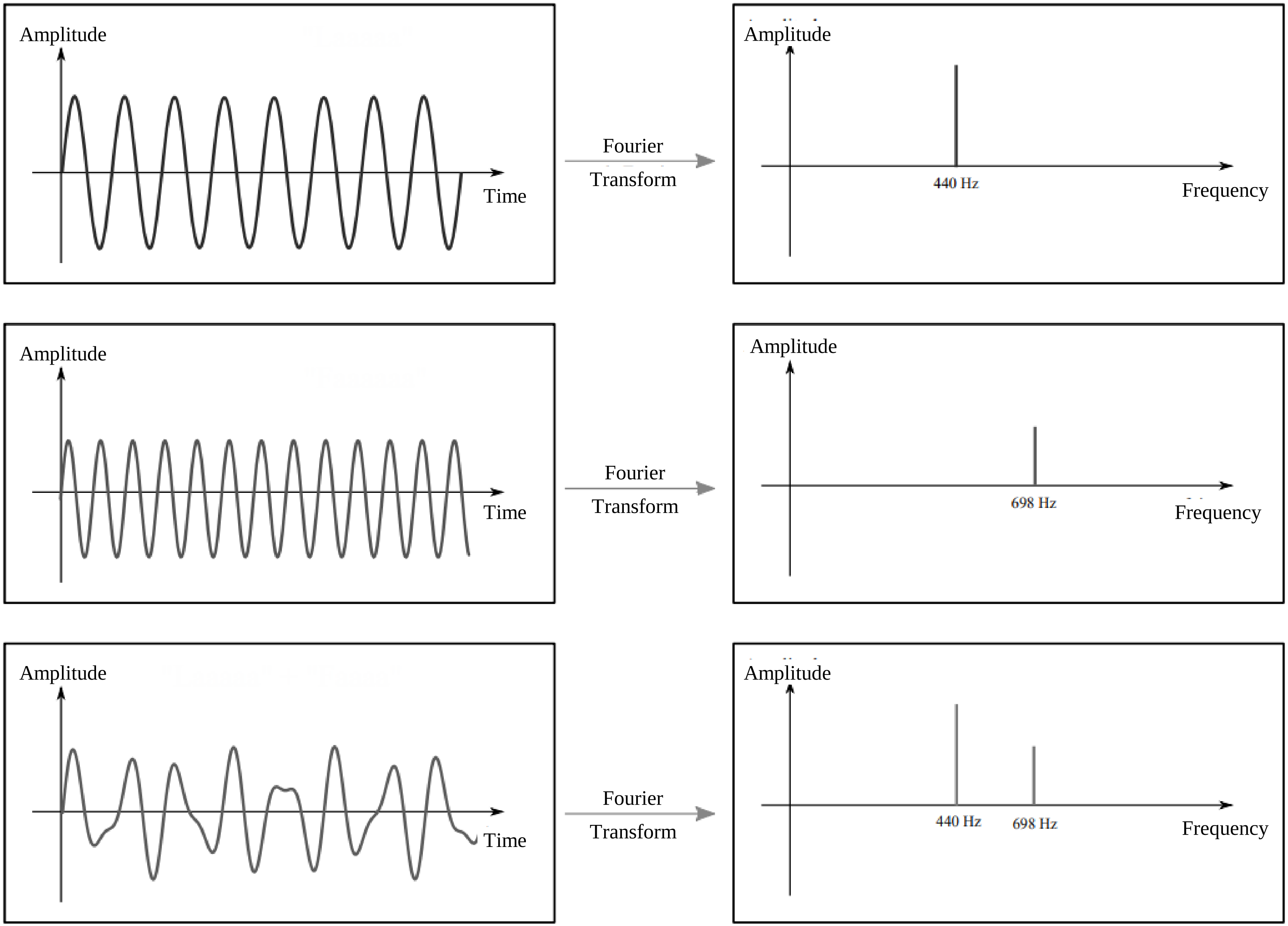
- **phase correction**
- **Fourier transformation**



measured signal

Simultaneously wide spectral range measurement

Examples of Fourier Transform function



Phase correction process

Interferogram space | **Spectrum space**

$$I(\delta)=\int_{-\infty}^{+\infty} B(\tilde{\nu})\cos(2\pi\tilde{\nu}\delta)\,d\tilde{\nu} \quad \overset{\text{FT}}{\longleftrightarrow} \quad B(\tilde{\nu})=\int_{-\infty}^{+\infty} I(\delta)\cos(2\pi\tilde{\nu}\delta)\,d\delta$$

interferogram | **spectrum**

The spectrum should be a **real function**
(for ideal and perfect instrument)

But in reality : - scanning mirror moving continuously
- beamsplitter chromatism, …

$$I_{meas}(\delta)=\int_{-\infty}^{+\infty} B(\tilde{\nu})\cos\left(2\pi\tilde{\nu}\delta+\Phi_{res}(\tilde{\nu})\right)d\tilde{\nu} \quad \overset{\text{FT}}{\longleftrightarrow} \quad B_{meas}(\tilde{\nu})=B(\tilde{\nu})\exp\left(i\Phi_{res}(\tilde{\nu})\right)$$

residual phase to be determined

phase correction

$$I(\delta)=I_{meas}(\delta)\otimes TF^{-1}\left(e^{-i\Phi_{res}(\tilde{\nu})}\right) \quad \overset{\text{FT}}{\longleftrightarrow} \quad B_{meas}(\tilde{\nu})=B(\tilde{\nu})\underbrace{\exp\left(i\Phi_{res}(\tilde{\nu})\right)\exp\left(-i\Phi_{res}(\tilde{\nu})\right)}_{1}$$

real measured spectrum

UV-VIS-IR configurations of the FTS-Paris

UV configuration

Spectral domain	28000-43000 cm^{-1}
External source	Xenon lamp
Entrance window	suprasil
Beamsplitter	UV quartz III
Detector	UV diode GAP diode

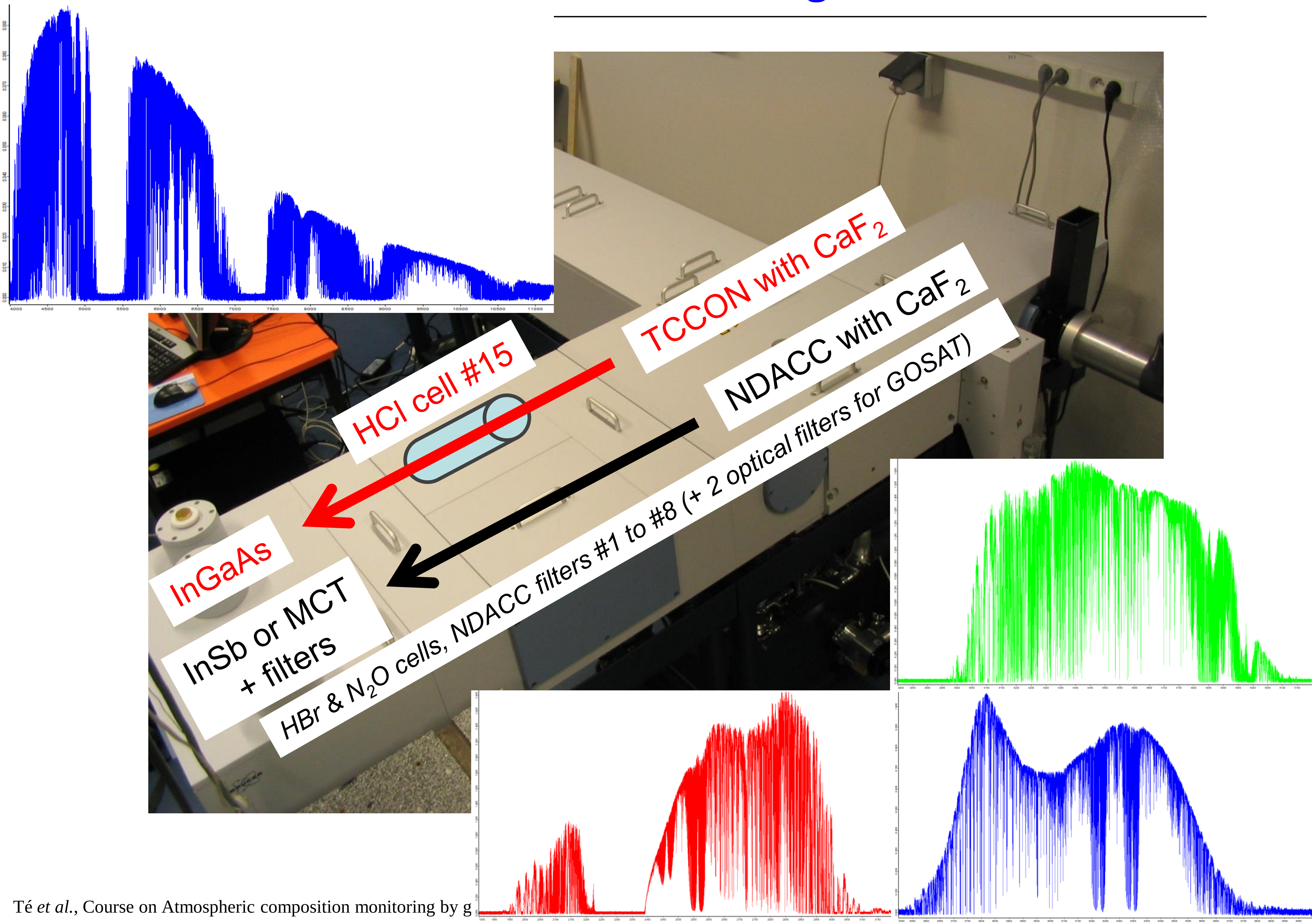
VIS configuration

Spectral domain	9500-25000 cm^{-1}
Internal source	QTH lamp NIR-VIS
Beamsplitter	Visible quartz II
Detector	Si diode

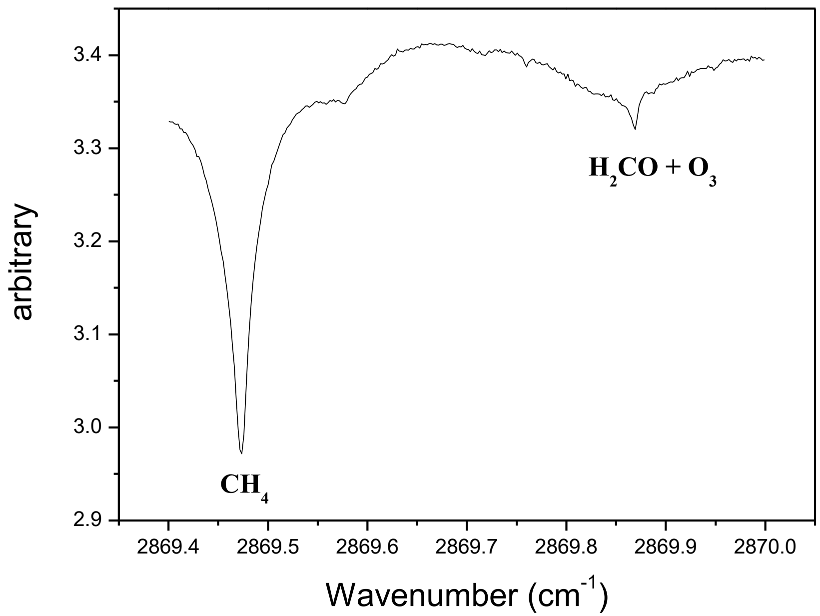
IR configuration

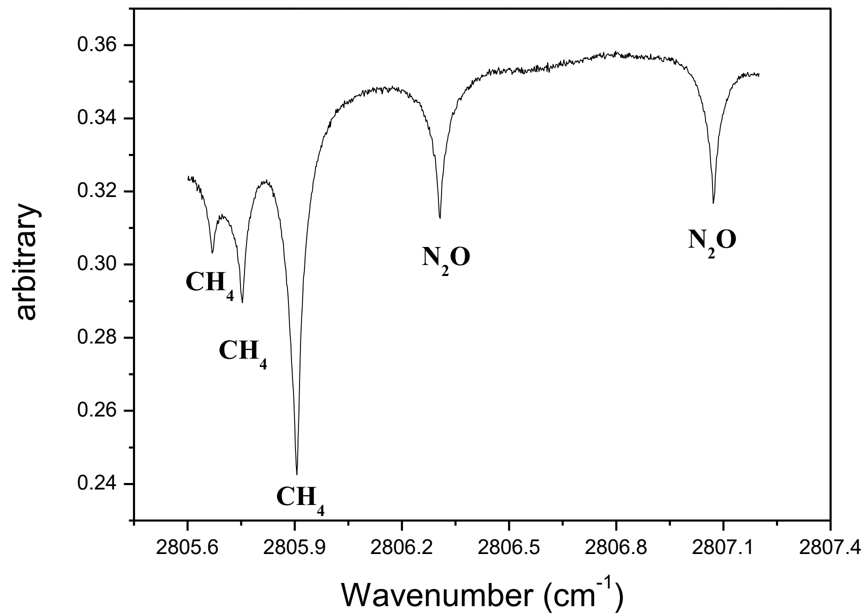
Internal source	Globar or QTH lamp
Beamsplitter	KBr : 450-4800 cm^{-1} CaF_2 : 1850-14000 cm^{-1}
Entrance window	KBr : 450-25000 cm^{-1} CaF_2 : 1850-14000 cm^{-1}
MCT detector	$D^* > 2.5\times10^{10}$ $cmHz^{1/2}W^{-1}$
InSb detector	$D^* > 1.5\times10^{11}$ $cmHz^{1/2}W^{-1}$
InGaAs detector	$NEP < 5\times10^{-12}$ $W/Hz^{1/2}$
HBr cell	NDACC Ref. #10
HCl cell	TCCON Ref. #15

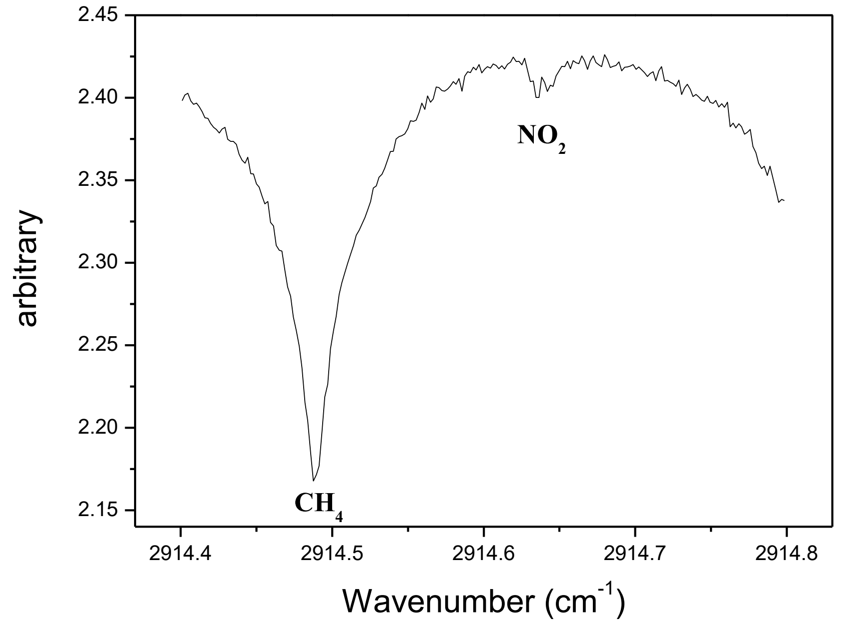
TCCON & NDACC configurations



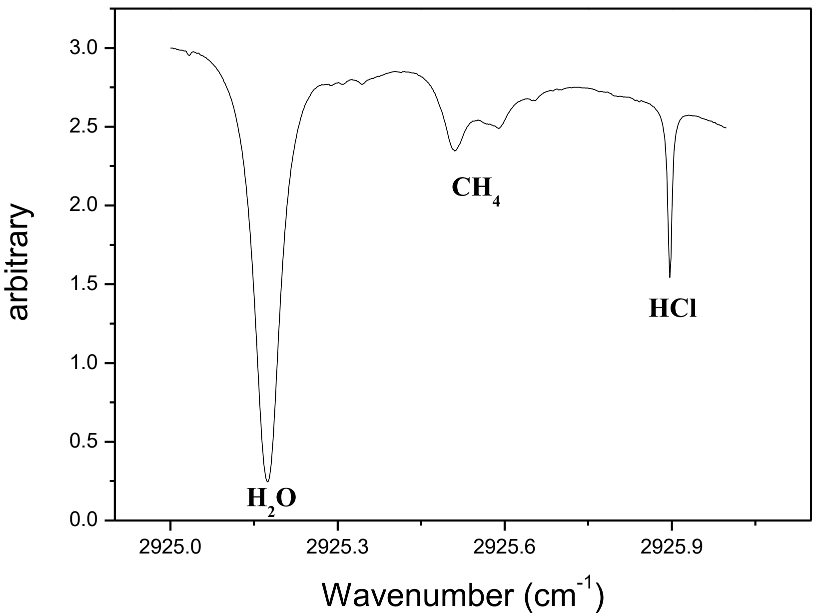
Some atmospheric species observed by FTS-Paris (1)

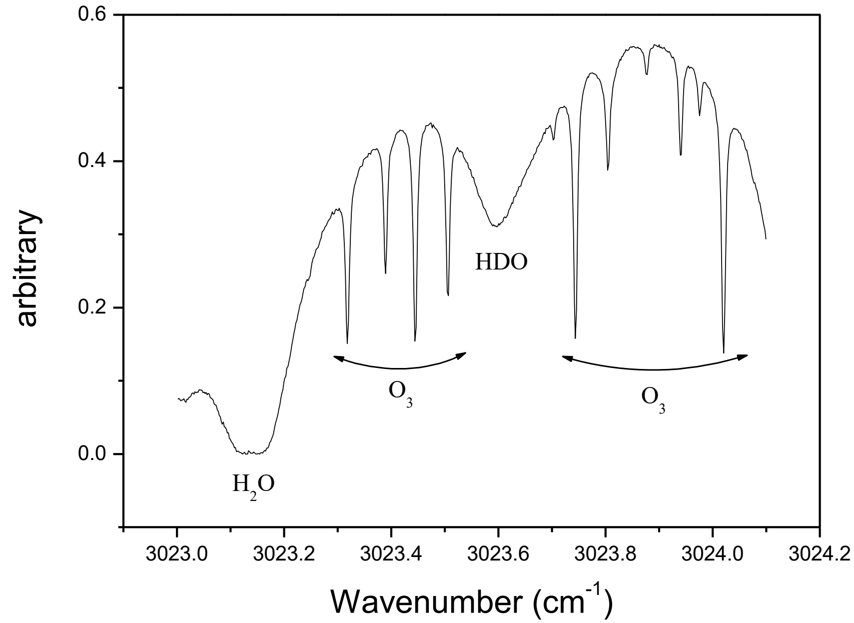


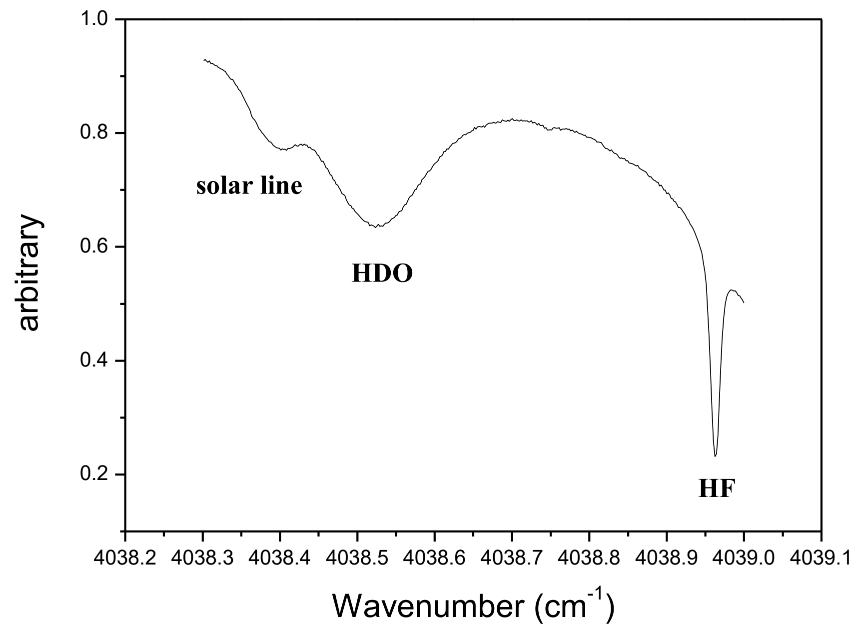




Some atmospheric species observed by FTS-Paris (2)







Radiative transfer algorithm

Radiative transfer model

$$L_{\tilde{\nu}}(M) = L_{\tilde{\nu}}(M_0).T_{\tilde{\nu}}(M_0, M) + \int_{M_0}^{M} J_{\tilde{\nu}}(s').T_{\tilde{\nu}}(s', s).\alpha_{\tilde{\nu}}(s').ds'$$

radiance at position M

incident radiance

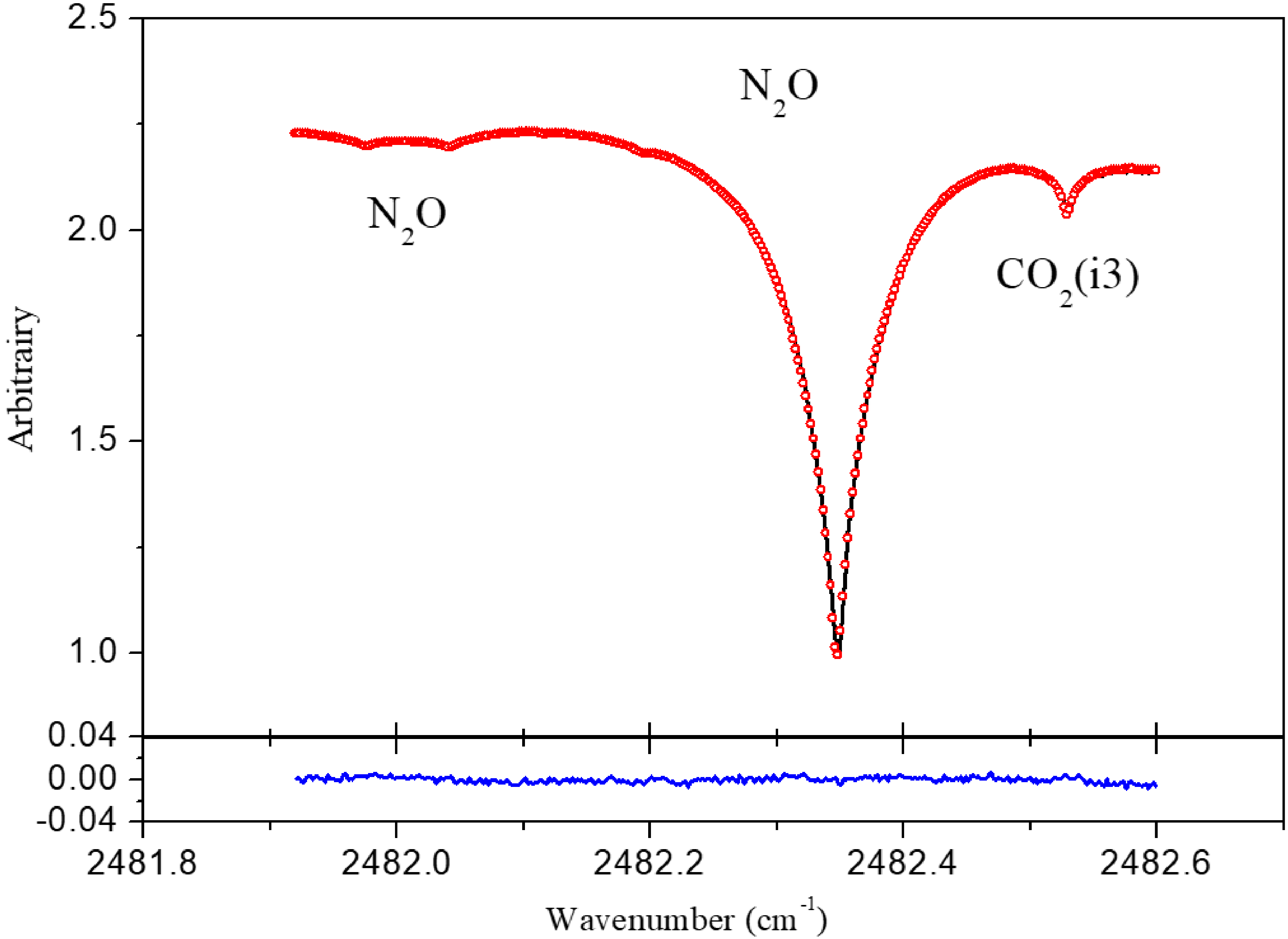
emission contribution

$$T_{\tilde{\nu}}(M_0, M) = \exp\left(- \int_{M_0}^{M} \sum_{i}^{species} \sum_{j}^{lines} S_{\tilde{\nu}}(\tilde{\nu}_j, i, T(s'))\,\Phi_{\tilde{\nu}}(\tilde{\nu}_j, i, T(s'), p(s'))\,N_i(s')\,ds' \right)$$

atmospheric transmission

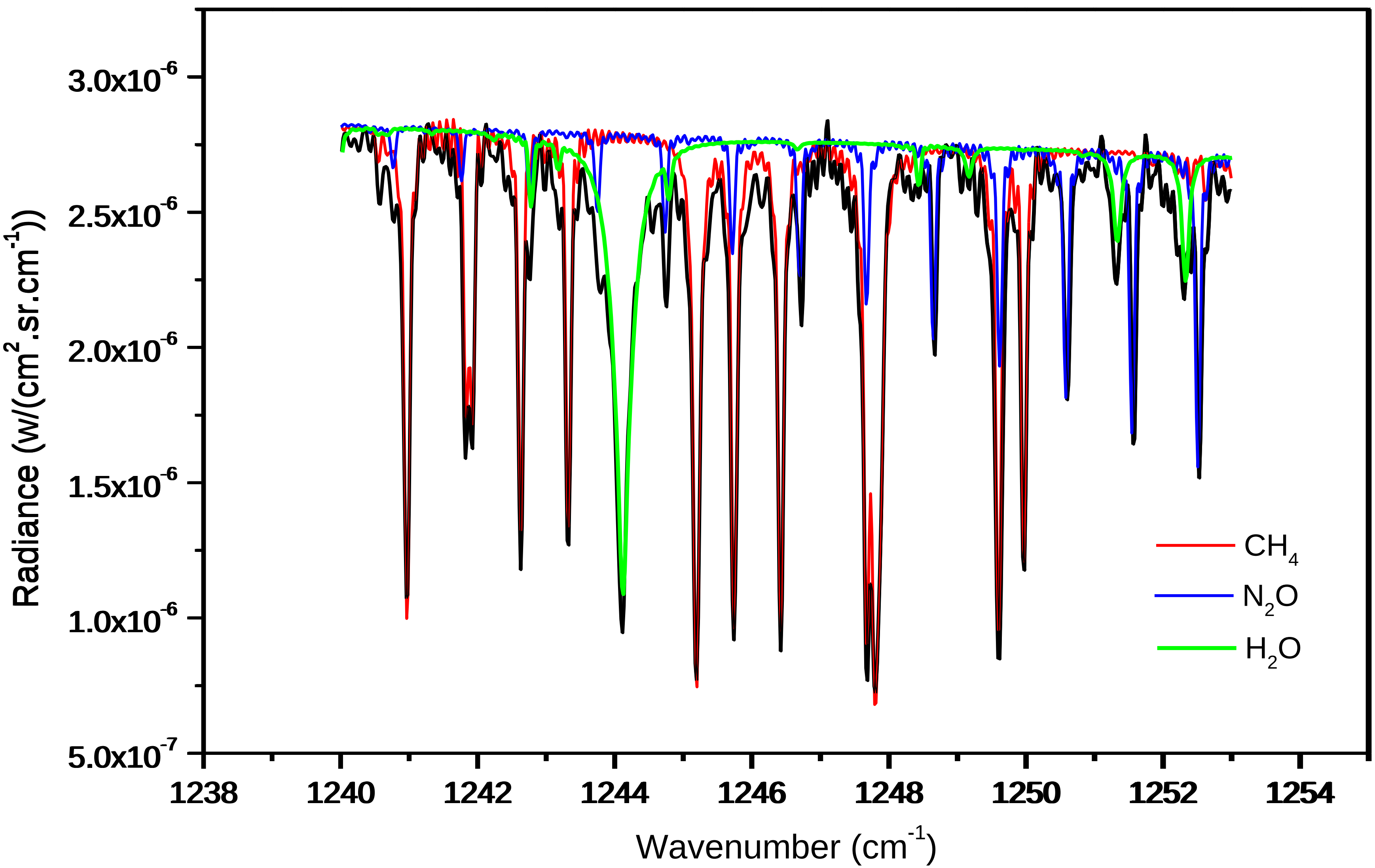
concentration

Modelling of N_2O line @2482 cm^{-1}



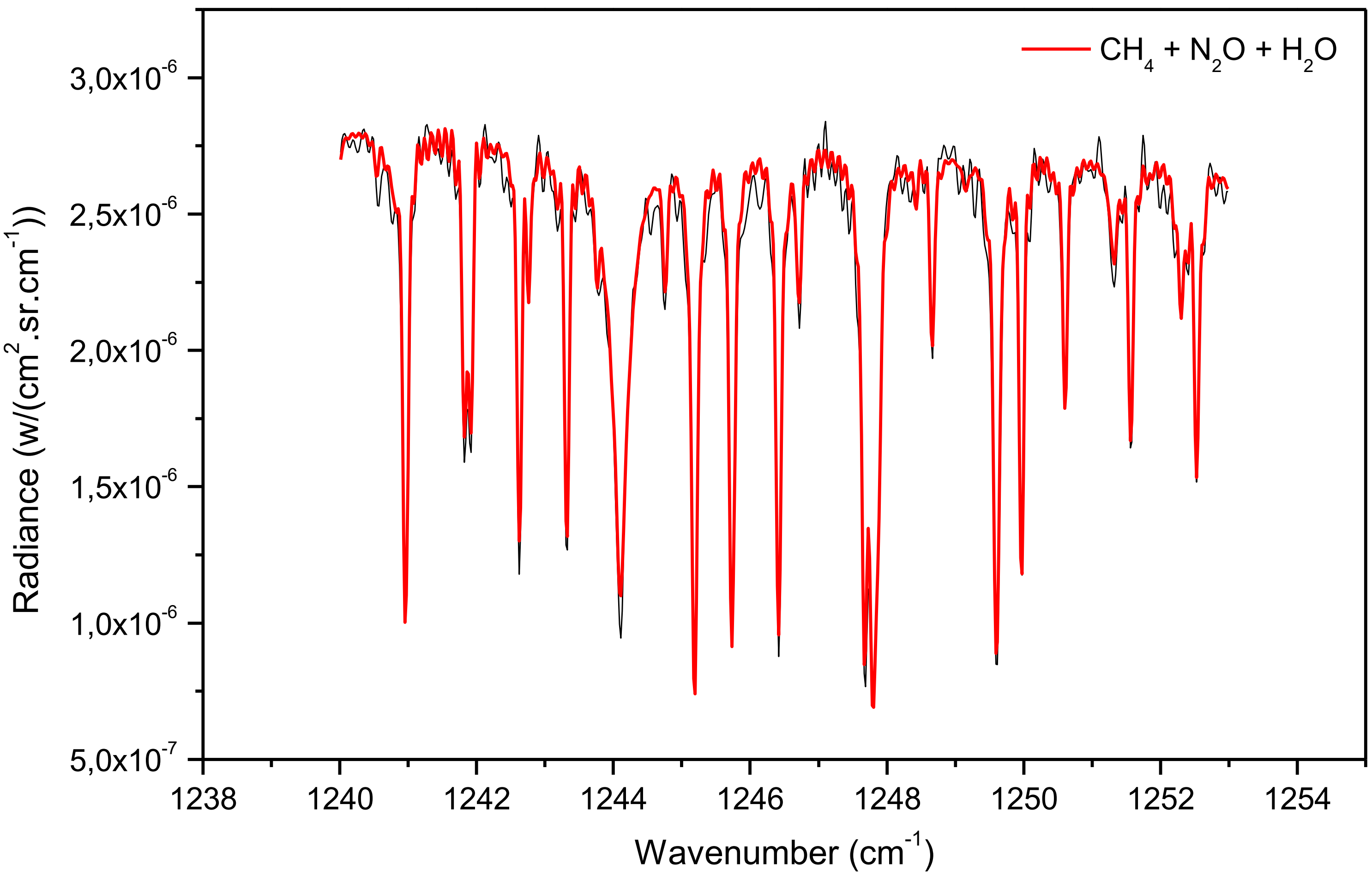
Importance of taking into account all species (1/2)

- Contribution of CH_4
- Contribution of N_2O
- Contribution of H_2O



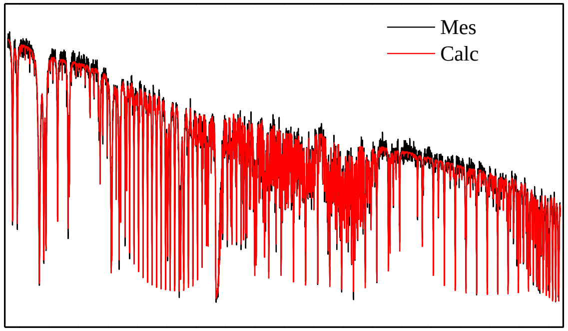
Importance of taking into account all species (2/2)

◆ Contribution of CH_4, N_2O and H_2O



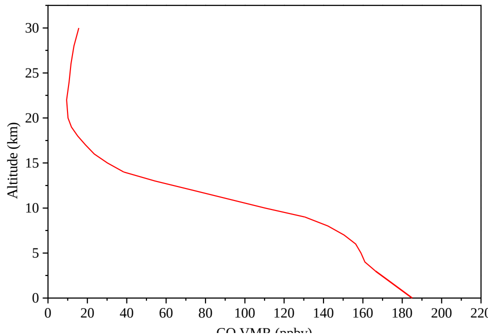
Parameters used by the radiative transfer algorithm

- Spectroscopic parameters HITRAN (line position & intensity, air-broadening, …)
- Vertical profile of temperature and pressure (NCEP, ECMWF, sounding)
- *A priori* vertical profile of studied species (WACCM, climatology)
- Taking into account the Instrument Line Shape
- Taking into account H_2O continuum effect
- Taking into account line mixing effect
- Taking into account the line of sight

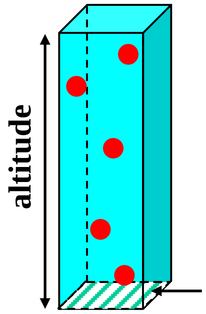


- ◆ Line by line calculation of the theoretical spectrum
- ◆ Adjustment of the theoretical spectrum to the measured one

• Species total column and/or profile



Vertical profile of VMR



Total column

Té *et al.*, Course on Atmospheric composition monitoring by ground-based FTIR measurement techni

HITRAN database: spectroscopic parameters

Molecule n° & isotope | position | intensity | air & self broadening

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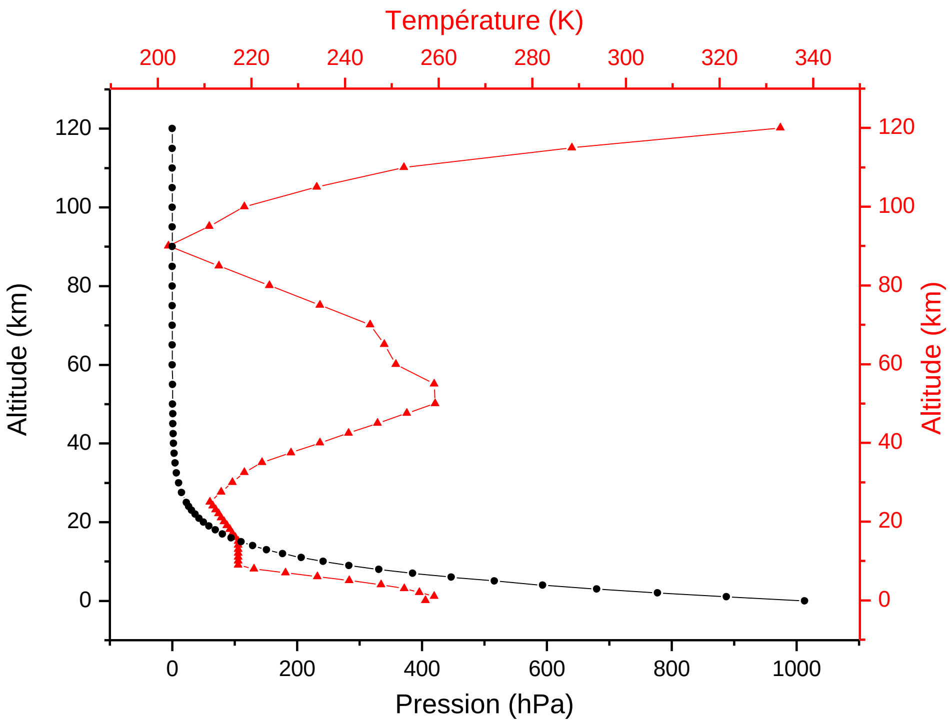
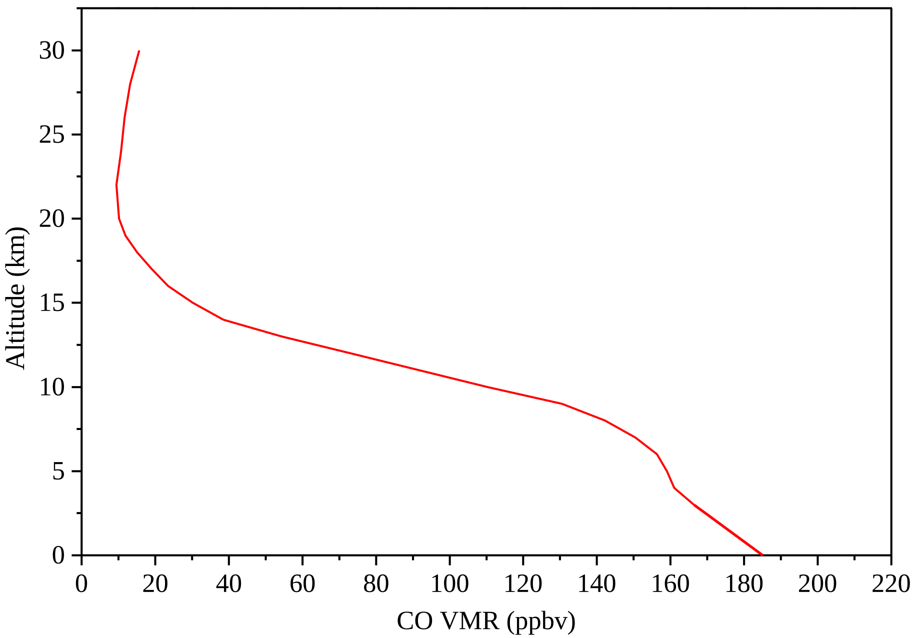
52 2099.710100 9.534E-22 1.067E+01.07970.086    0.00000.76-.001555    1    0    R  0    467664 2 2 2 2 1 6    6.0    2.0
54 2101.102700 1.086E-22 1.850E+01.06760.075   37.47690.74-.003090    1    0    P  4    467663 2 2 2 2 1 1   42.0   54.0
55 2101.264000 2.391E-24 1.528E+01.05270.057  533.92830.68-.002870    1    0    R 17    467663 2 2 2 2 1 1   74.0   70.0
55 2101.278200 1.283E-29 3.084E+01.04570.047 3256.54970.67-.003000    2    1    R 26    466623 2 2 2 2 1 1  110.0  106.0
51 2101.342400 1.798E-23 3.736E+01.06760.075 2181.36920.74-.003090    2    1    P  4    467663 2 2 2 2 1 1    7.0    9.0
53 2101.386300 5.862E-26 3.046E+01.05800.064 2255.40110.75-.001647    2    1    R  9    467664 2 2 2 2 1 6   21.0   19.0
54 2101.424200 6.301E-27 2.771E+01.07090.079 2127.43750.74-.002620    2    1    R  2    467663 2 2 2 2 1 1   42.0   30.0
52 2102.052800 3.409E-25 3.037E+01.05890.066 2227.19410.75-.001568    2    1    R  8    467664 2 2 2 2 1 6   38.0   34.0
56 2102.162400 3.799E-29 3.089E+01.05270.057 2610.70340.68-.002870    2    1    R 17    467663 2 2 2 2 1 1  444.0  420.0
56 2102.490400 1.567E-24 1.514E+01.05800.064  161.00290.75-.002550    1    0    R  9    467663 2 2 2 2 1 1  252.0  228.0
53 2102.911800 4.821E-22 1.376E+01.07090.079   10.98570.74-.001928    1    0    R  2    467664 2 2 2 2 1 6    7.0    5.0
51 2103.269700 3.250E-19 1.730E+01.05800.064  211.40410.75-.003570    1    0    P 10    467663 2 2 2 2 1 1   19.0   21.0
52 2103.320400 1.876E-21 1.287E+01.07480.082    3.67590.75-.001963    1    0    R  1    467664 2 2 2 2 1 6   10.0    6.0
55 2103.846800 8.477E-30 3.099E+01.04510.046 3349.64060.67-.003000    2    1    R 27    466623 2 2 2 2 1 1  114.0  110.0
55 2104.160400 1.864E-24 1.538E+01.05190.056  596.68130.67-.002650    1    0    R 18    467663 2 2 2 2 1 1   78.0   74.0
53 2104.650600 5.416E-26 3.075E+01.05730.063 2291.67320.75-.001578    2    1    R 10    467664 2 2 2 2 1 6   23.0   21.0
54 2104.950800 8.780E-23 1.954E+01.07090.079   22.48670.74-.002030    1    0    P  3    467663 2 2 2 2 1 1   30.0   42.0
54 2105.002900 7.972E-27 2.889E+01.06760.075 2138.57960.74-.002530    2    1    R  3    467663 2 2 2 2 1 1   54.0   42.0
56 2105.091000 2.947E-29 3.107E+01.05190.056 2674.43240.67-.002650    2    1    R 18    467663 2 2 2 2 1 1  468.0  444.0
51 2105.256600 1.455E-23 3.945E+01.07090.079 2166.13050.74-.002030    2    1    P  3    467663 2 2 2 2 1 1    5.0    7.0
52 2105.362800 3.236E-25 3.069E+01.05800.064 2259.96680.75-.001528    2    1    R  9    467664 2 2 2 2 1 6   42.0   38.0
56 2105.716500 1.451E-24 1.527E+01.05730.063  196.76960.75-.002540    1    0    R 10    467663 2 2 2 2 1 1  276.0  252.0
55 2106.381600 5.501E-30 3.113E+01.04460.045 3446.14830.67-.003000    2    1    R 28    466623 2 2 2 2 1 1  118.0  114.0
53 2106.442800 6.105E-22 1.435E+01.06760.075   21.97100.74-.001918    1    0    R  3    467664 2 2 2 2 1 6    9.0    7.0
52 2106.897800 2.720E-21 1.386E+01.07090.079   11.02760.74-.001945    1    0    R  2    467664 2 2 2 2 1 6   14.0   10.0
55 2107.024300 1.425E-24 1.547E+01.05100.055  662.90640.67-.002790    1    0    R 19    467663 2 2 2 2 1 1   82.0   78.0
51 2107.423200 3.531E-19 1.750E+01.05890.066  172.97800.75-.003590    1    0    P  9    467663 2 2 2 2 1 1   17.0   19.0
53 2107.880900 4.876E-26 3.101E+01.05670.062 2331.56740.75-.001579    2    1    R 11    467664 2 2 2 2 1 6   25.0   23.0
56 2107.985700 2.241E-29 3.125E+01.05100.055 2741.68710.67-.002790    2    1    R 19    467663 2 2 2 2 1 1  492.0  468.0
54 2108.547300 9.287E-27 2.969E+01.06500.073 2153.43510.74-.002660    2    1    R  4    467663 2 2 2 2 1 1   66.0   54.0
```

Vertical profiles : temperature/pressure & CO

Temperature/pressure



Carbon monoxide (CO)



Finite optical path difference effect

$$B(\tilde{\nu}) = \int_{-\infty}^{+\infty} I(\delta)\cos(2\pi\tilde{\nu}\delta)\,d\delta$$

finite value of δ

boxcar function

$$\Pi(\delta) = 1 \quad \text{si} \ -\delta_{max} \leq \delta \leq \delta_{max}$$

$$\Pi(\delta) = 0 \quad \text{sinon}$$

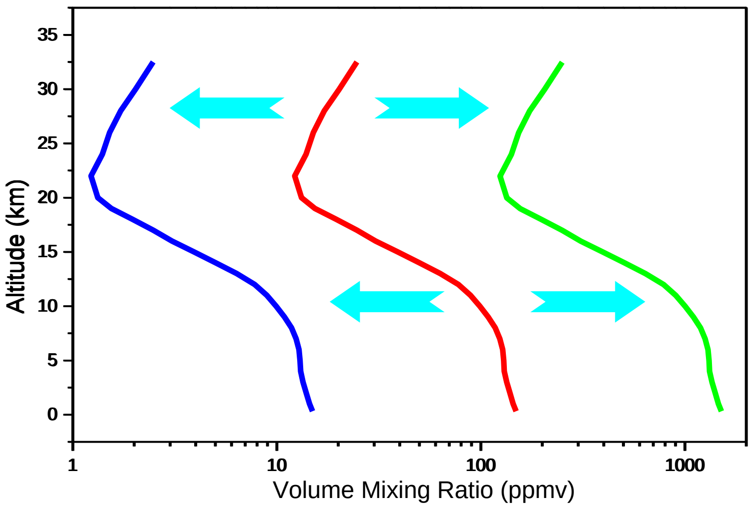
$$B_{mes}(\tilde{\nu}) = \int_{-\infty}^{+\infty} I(\delta)\Pi(\delta)\cos(2\pi\tilde{\nu}\delta)\,d\delta$$

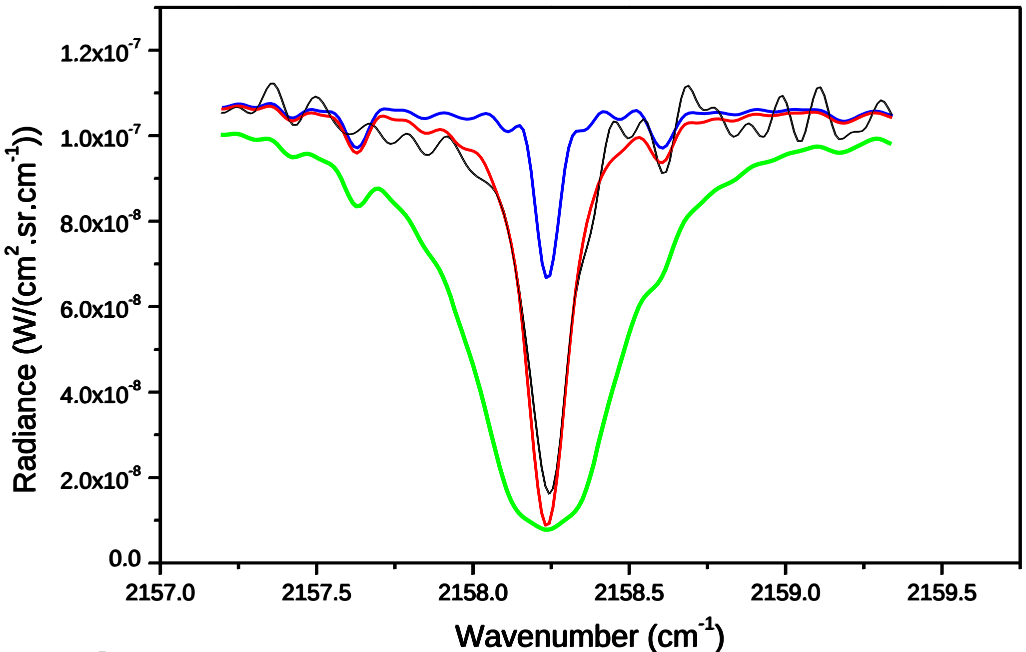
$$B_{mes}(\tilde{\nu}) = B(\tilde{\nu}) \otimes \underbrace{TF(\Pi(\delta))}_{\text{ILS}}$$

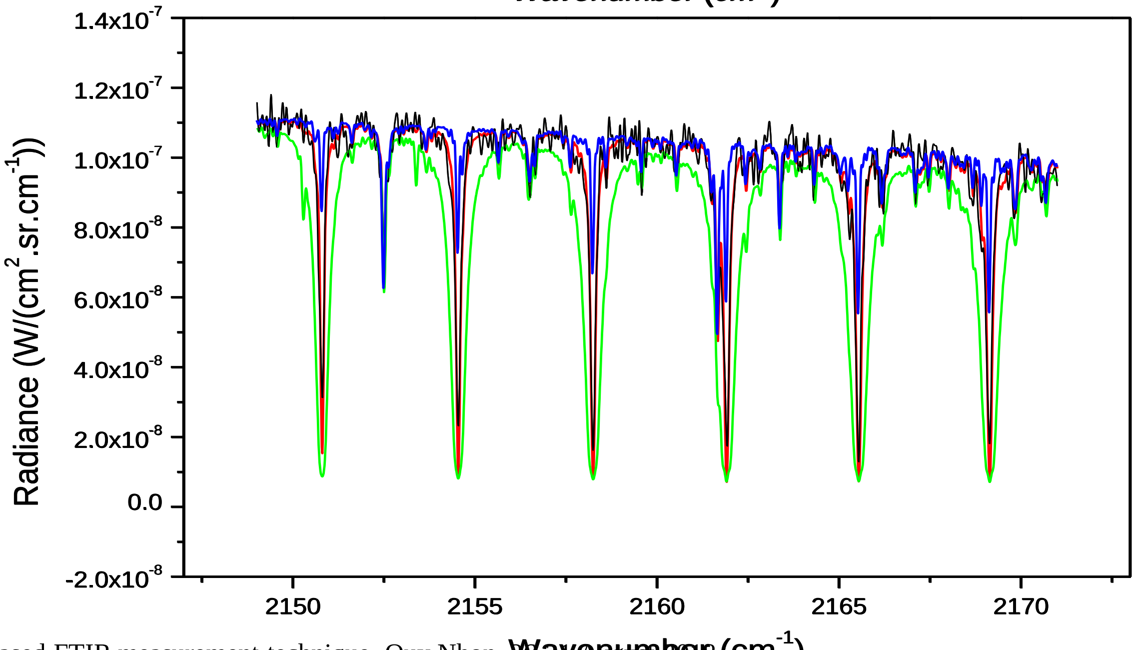
Instrument Line Shape function (ILS)

The measured spectrum is the convolution of the incident spectrum with the ILS function

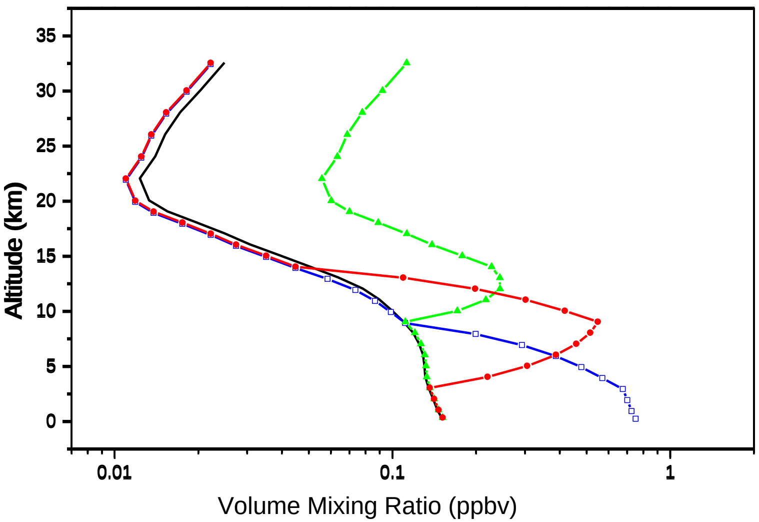
Fitting concentrations in column

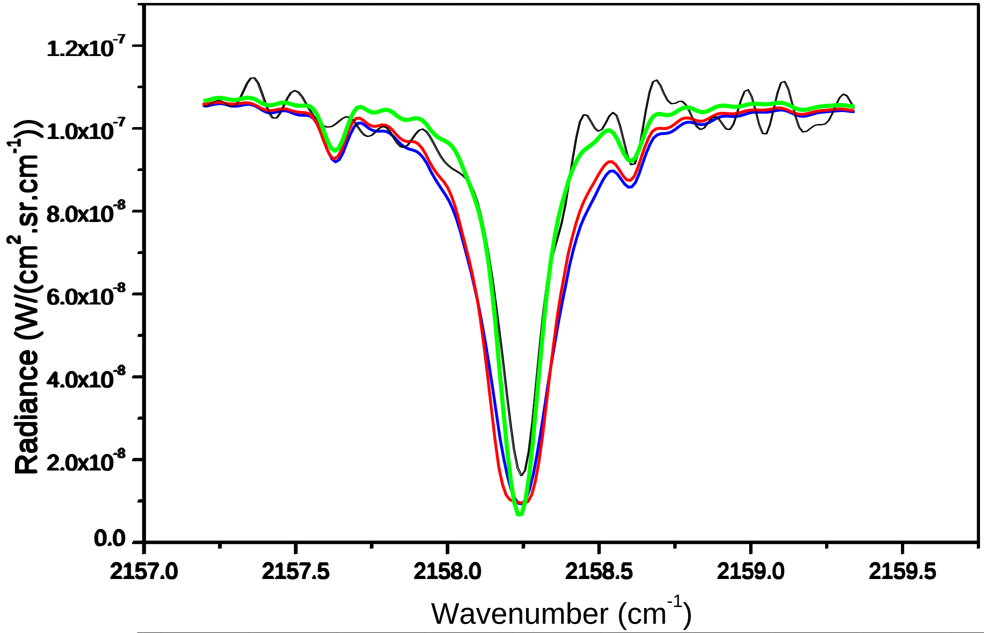


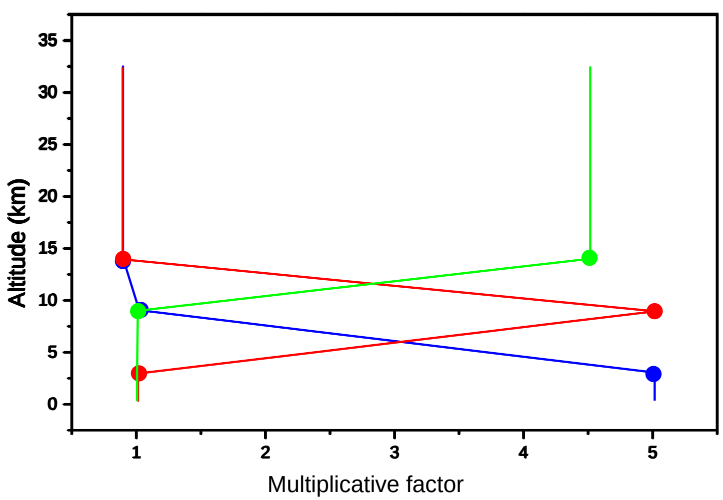


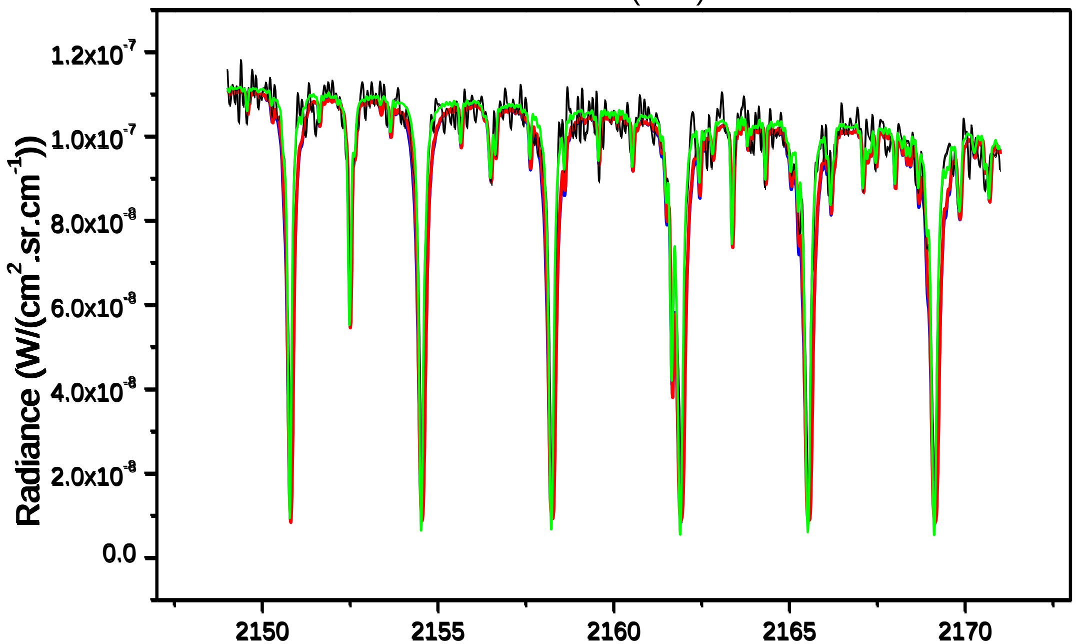


Fitting concentrations in profile









Té *et al.*, Course on Atmospheric composition monitoring by ground-based FTIR measurement technique, Quy Nhon, 26-31 August 2018

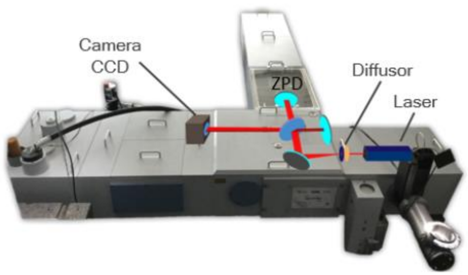
ILS function characterization

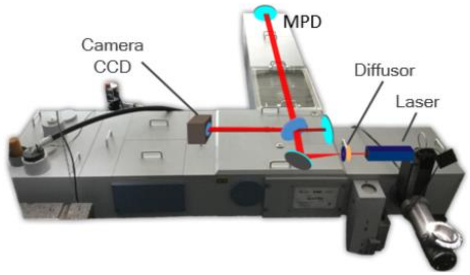
Optical alignment of FTS-Paris

➔ **Alignment of both fixed and mobile mirrors (reflectors)**

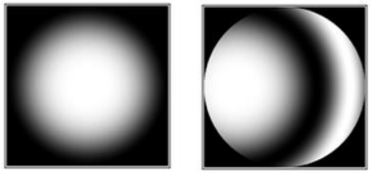
➔ **Their axis should be parallel for each instant along the optical path (between ZPD and MPD)**

➔ **Newton rings should be contrasted and centered (almost fixed in position)**



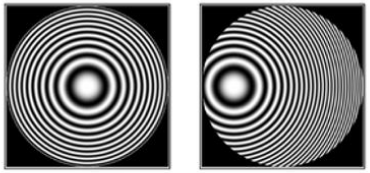


Aligned Misaligned

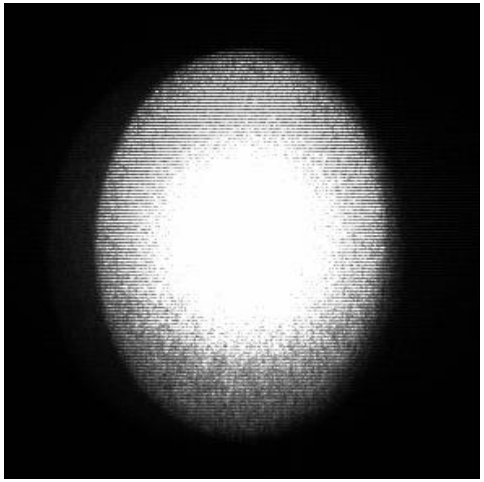


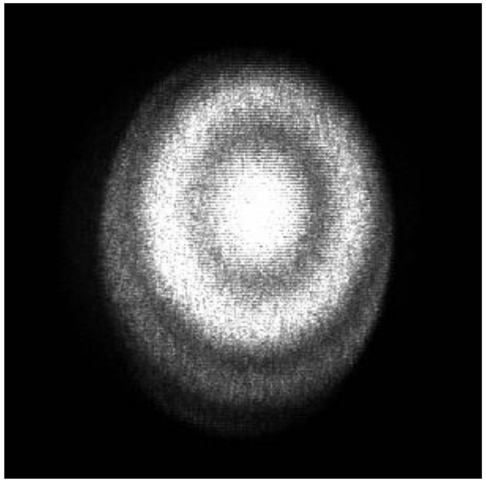
ZPD

Aligned Misaligned

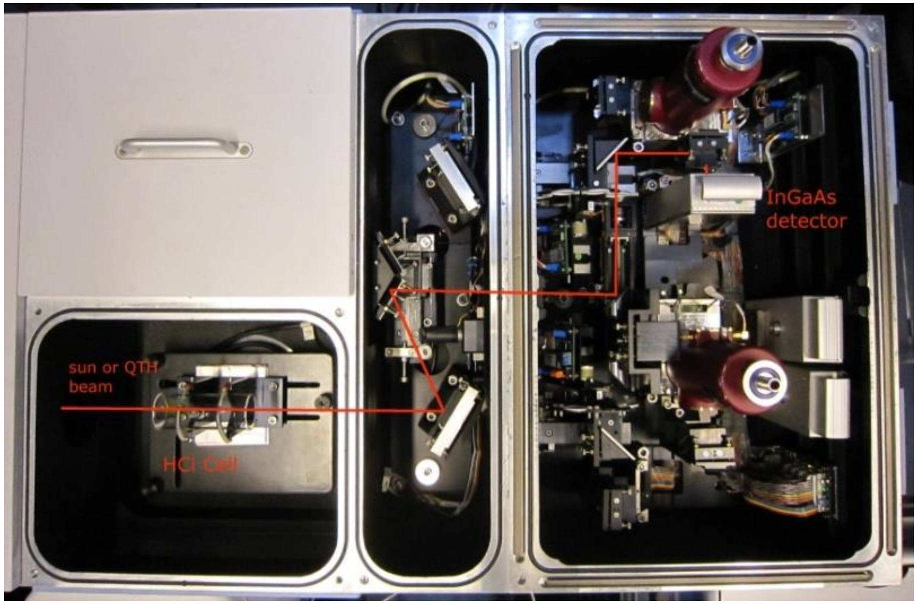


MPD



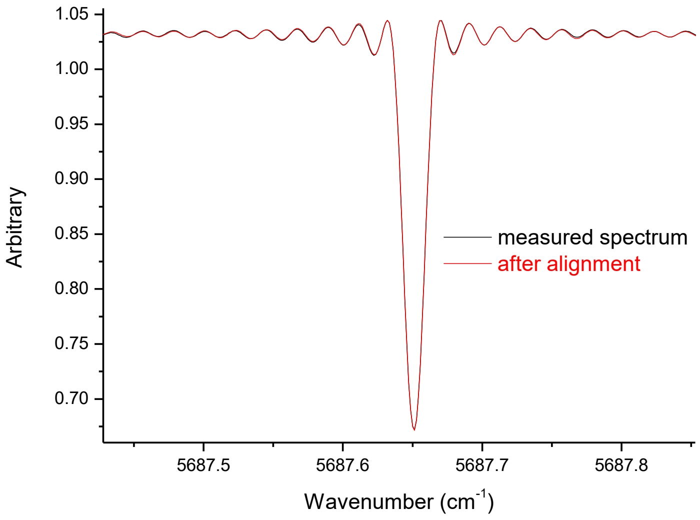


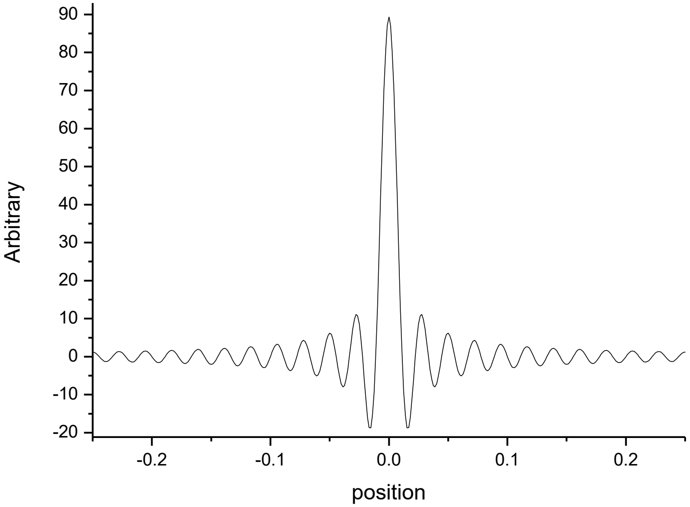
Instrument Line Shape and HCl gas cell



HCl gas cell #15 (February 2013)

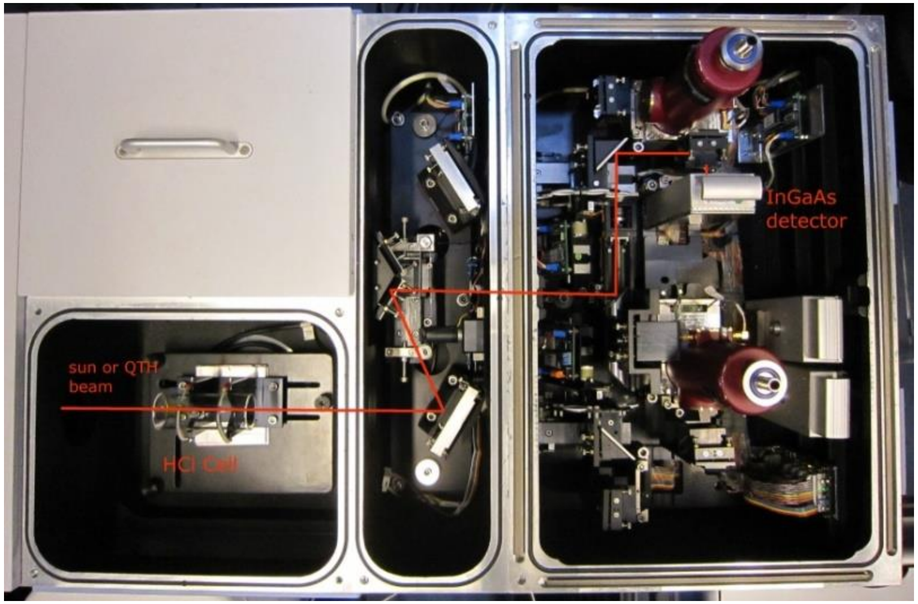
- Pressure ~ 5 mbar
- Column ~ 1.35×10^{22} molecule.m^{-2}





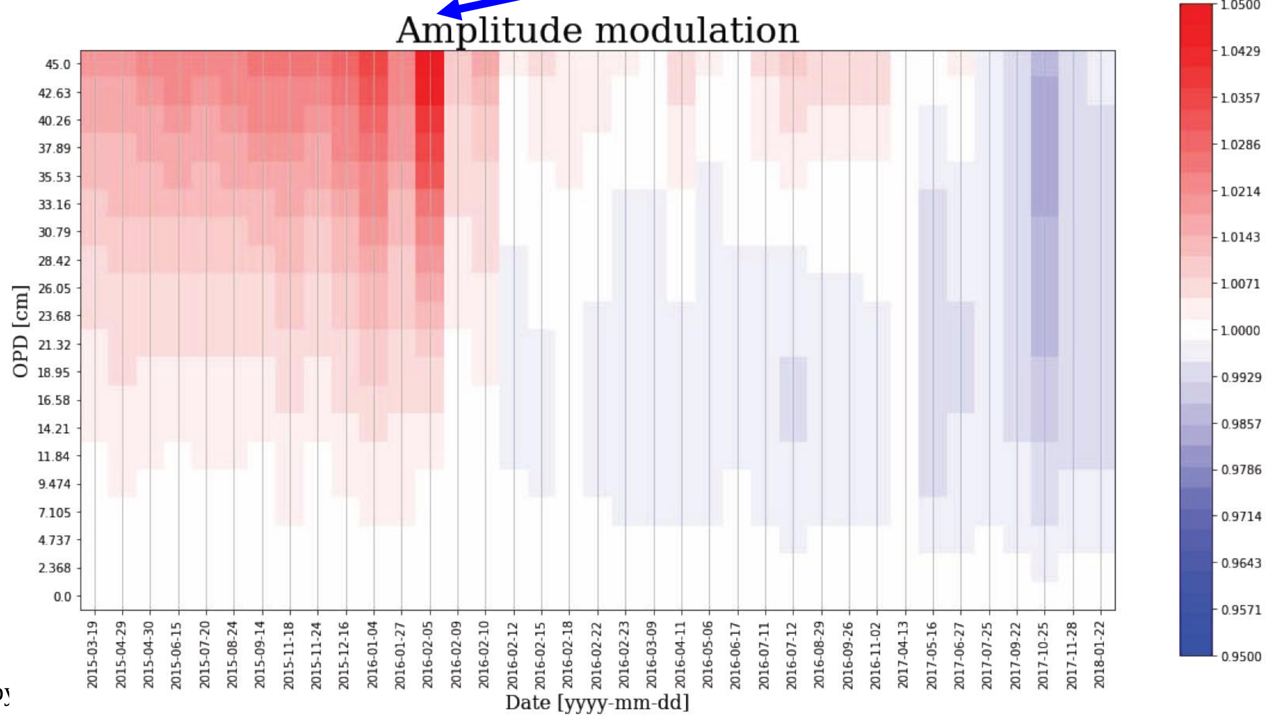
↳ Symmetrical ILS

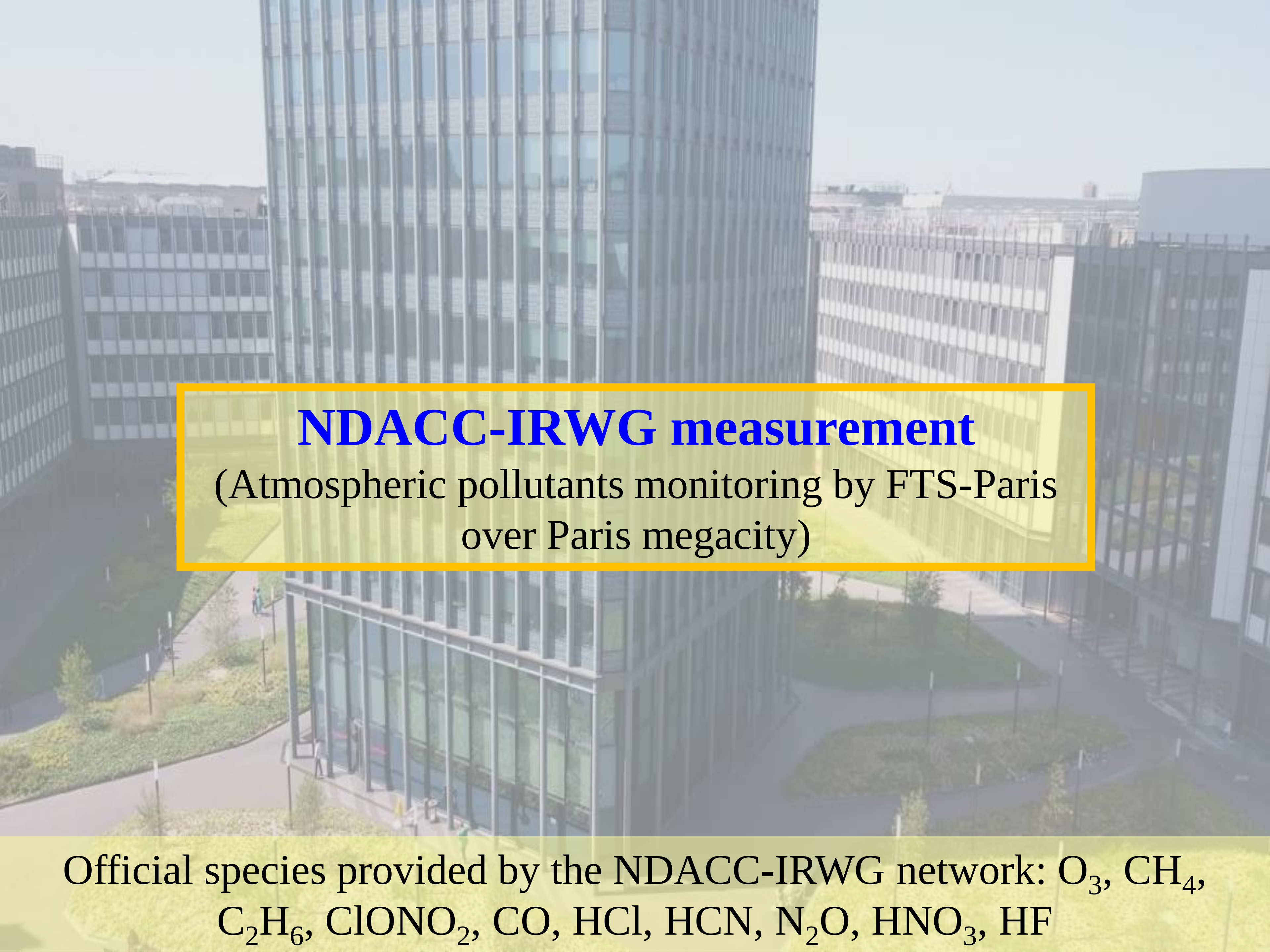
Instrument performances stability



➔ **Normalized modulation of the ILS function should be lower than 5%**

- **Modulation close to 1.05**
- **Optical alignment in Feb. 2016**





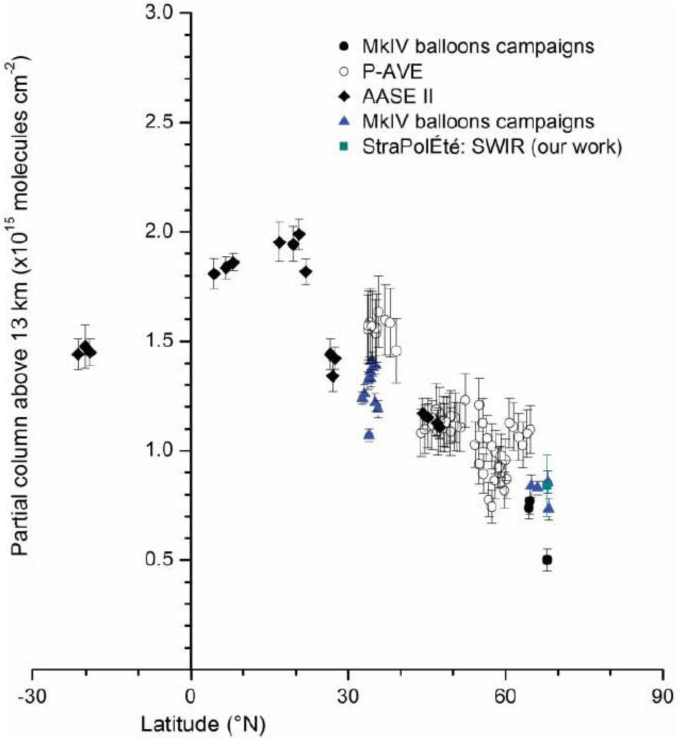
NDACC-IRWG measurement

(Atmospheric pollutants monitoring by FTS-Paris over Paris megacity)

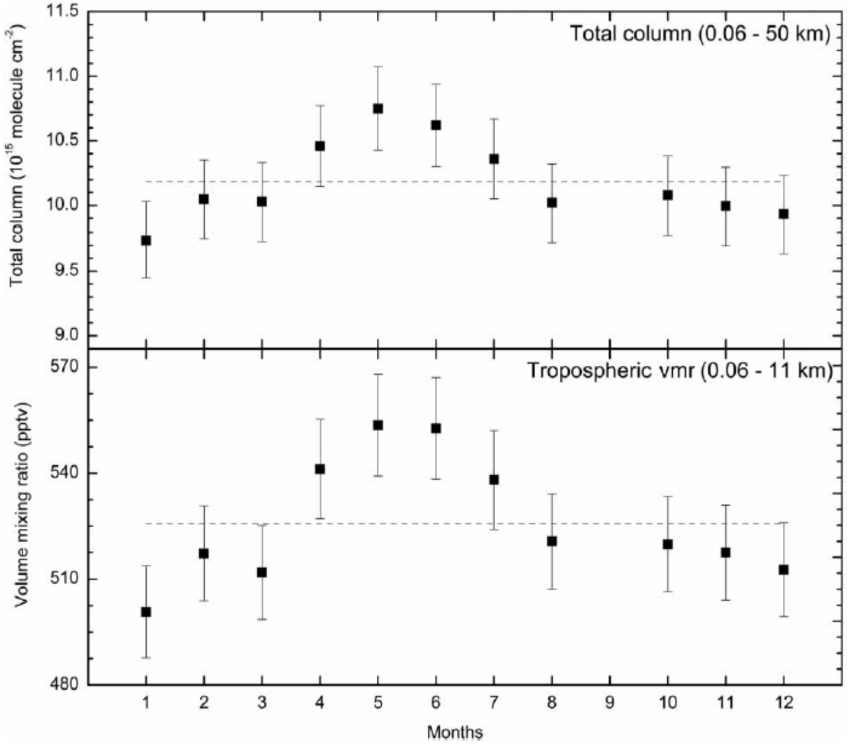
Official species provided by the NDACC-IRWG network: O_3, CH_4, C_2H_6, $ClONO_2$, CO, HCl, HCN, N_2O, HNO_3, HF

OCS (or COS) retrieval (balloon & ground-based)

➔ OCS is the most abundant sulphur compound in the atmosphere and an important precursor of sulphate aerosols



(Krysztofiak *et al.*, Atmosphere-Oean, 2014)

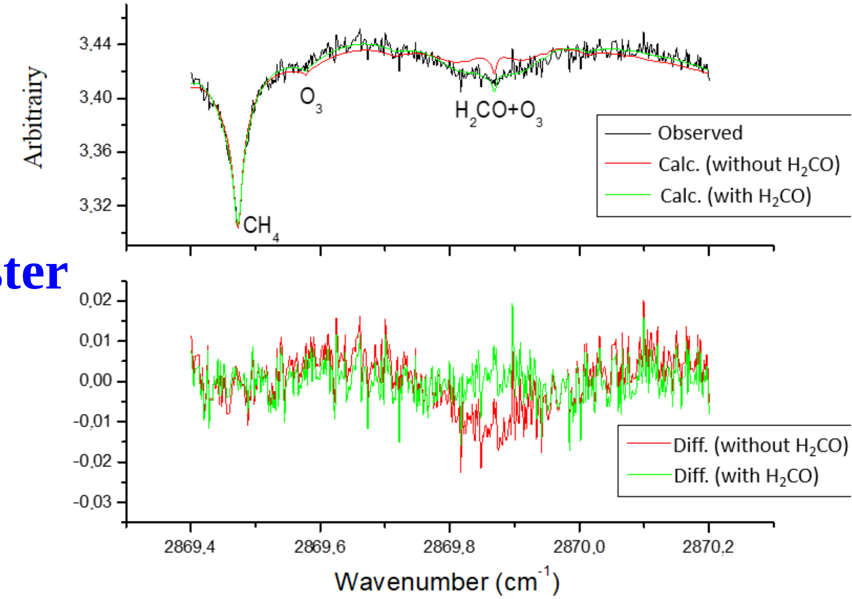


⇨ OCS seasonality over Paris obtained by the FTS-Paris instrument

H_2CO retrieval (seasonality and trends)

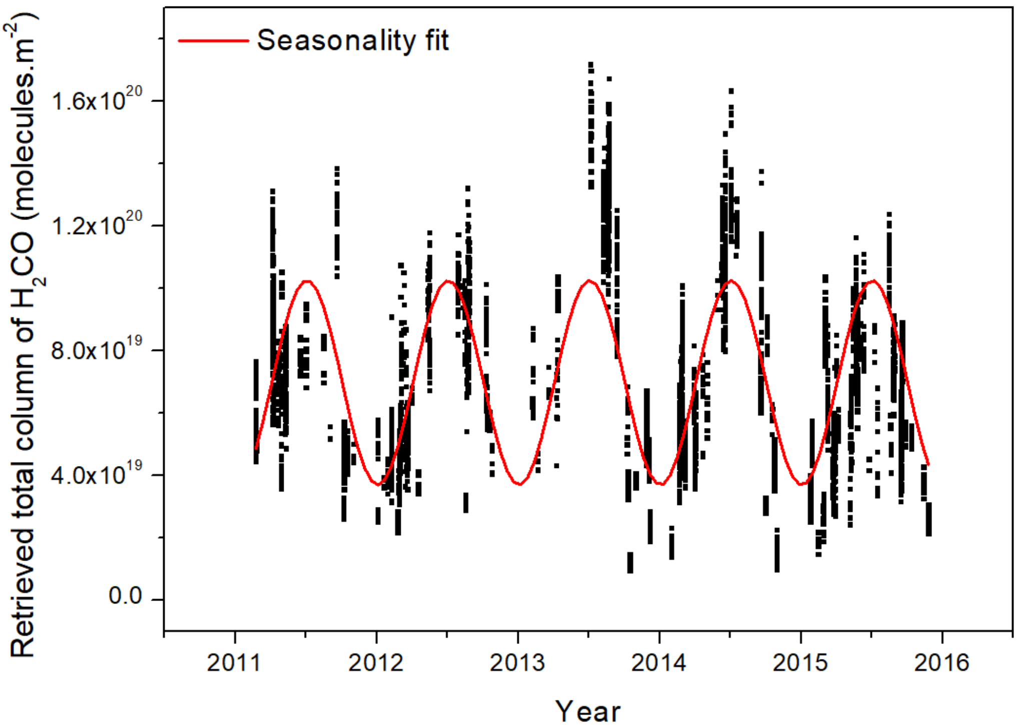
- Sources : intermediate product of hydrocarbon degradation (methane, VOC), biomass burning; combustion engine, …

- Loss : photosynthesis, OH oxidation, soil …



(Té *et al.*, ASA-HITRAN 2012)

➔ First retrieval of H_2CO using FTS-Paris data by C. Veras (master student) ⇨ Signal very small (retrieval in column)



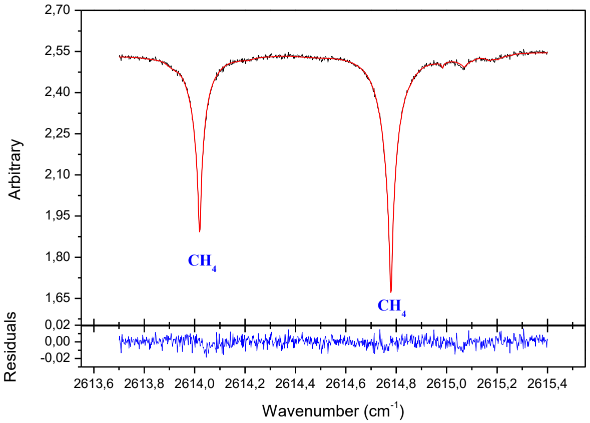
➔ New harmonized global study conducted by the NDACC-IRWG network, cf. Vigouroux *et al.* (2018)

⇨ Seasonality peak of 88%

⇨ No observed trends (fluctuation too large)

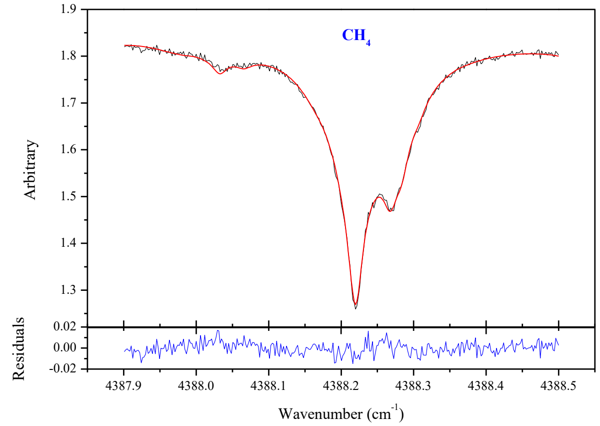
CH_4 and N_2O retrievals

~ 4 µm



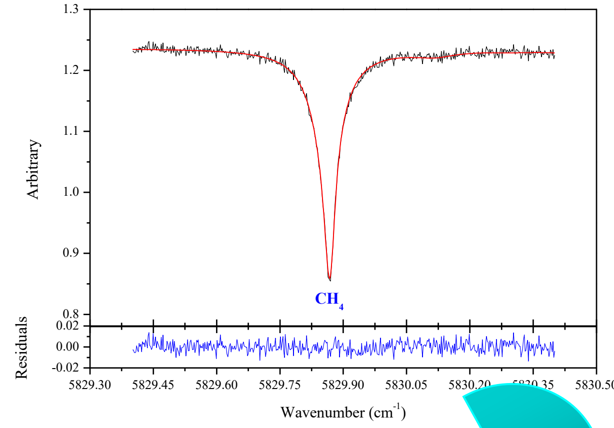
1 of 5 used µ-windows

~ 2.5 µm

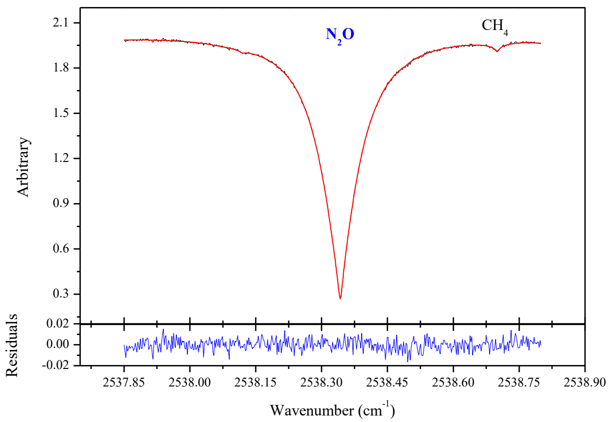


1 of 5 used µ-windows

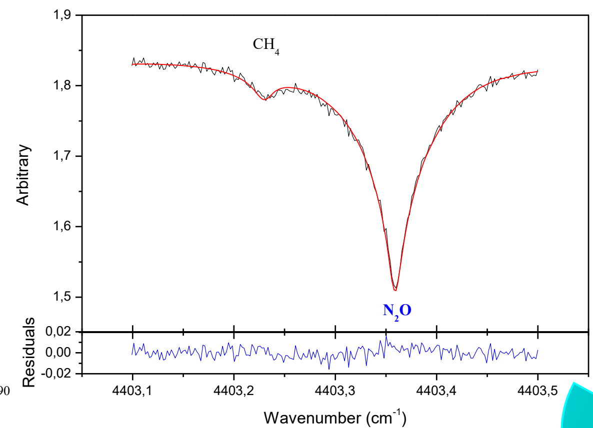
~ 1.5 µm



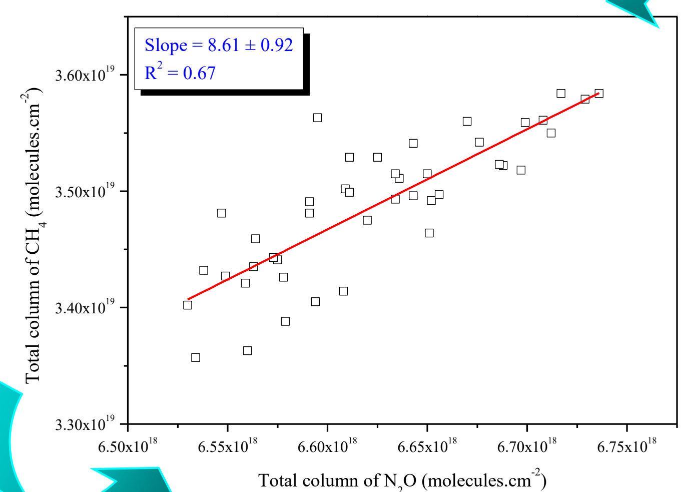
1 of 4 used µ-windows



1 of 4 used µ-windows



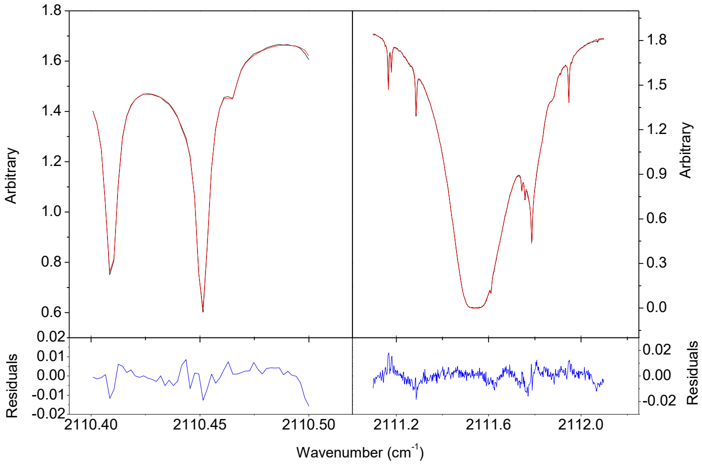
1 of 4 used µ-windows



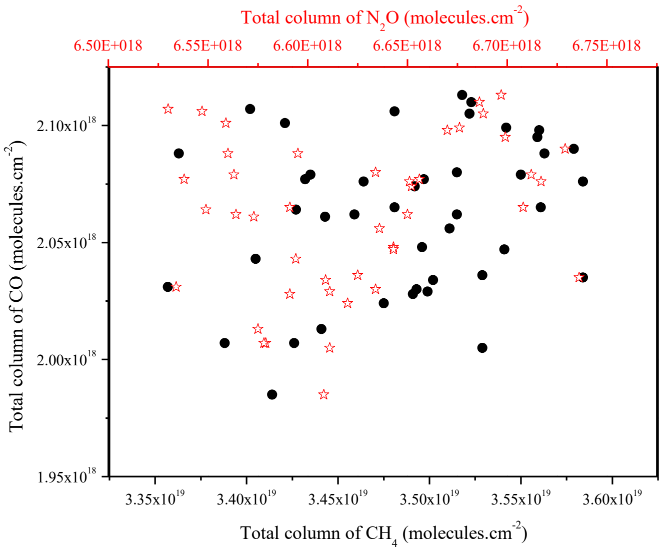
Good correlation : no strong anthropogenic local sources

CO retrieval

(Figure from Té *et al.*, JAOT 2012)



2 used µ-windows



Strong local emission of CO

Characterization of CO retrieval error

Error source	CO error (%)
Temperature	1.8
Instrument noise	<1
Solar zenith angle	1.1
Interfering solar lines	<1
Total random error	2.4
Systematic errors	
Spectroscopic parameters	3–6.8
A priori profile	<1
Instrument line shape	<1
Total systematic error	3.1–6.9

(Table from Té *et al.*, JAOT 2012)

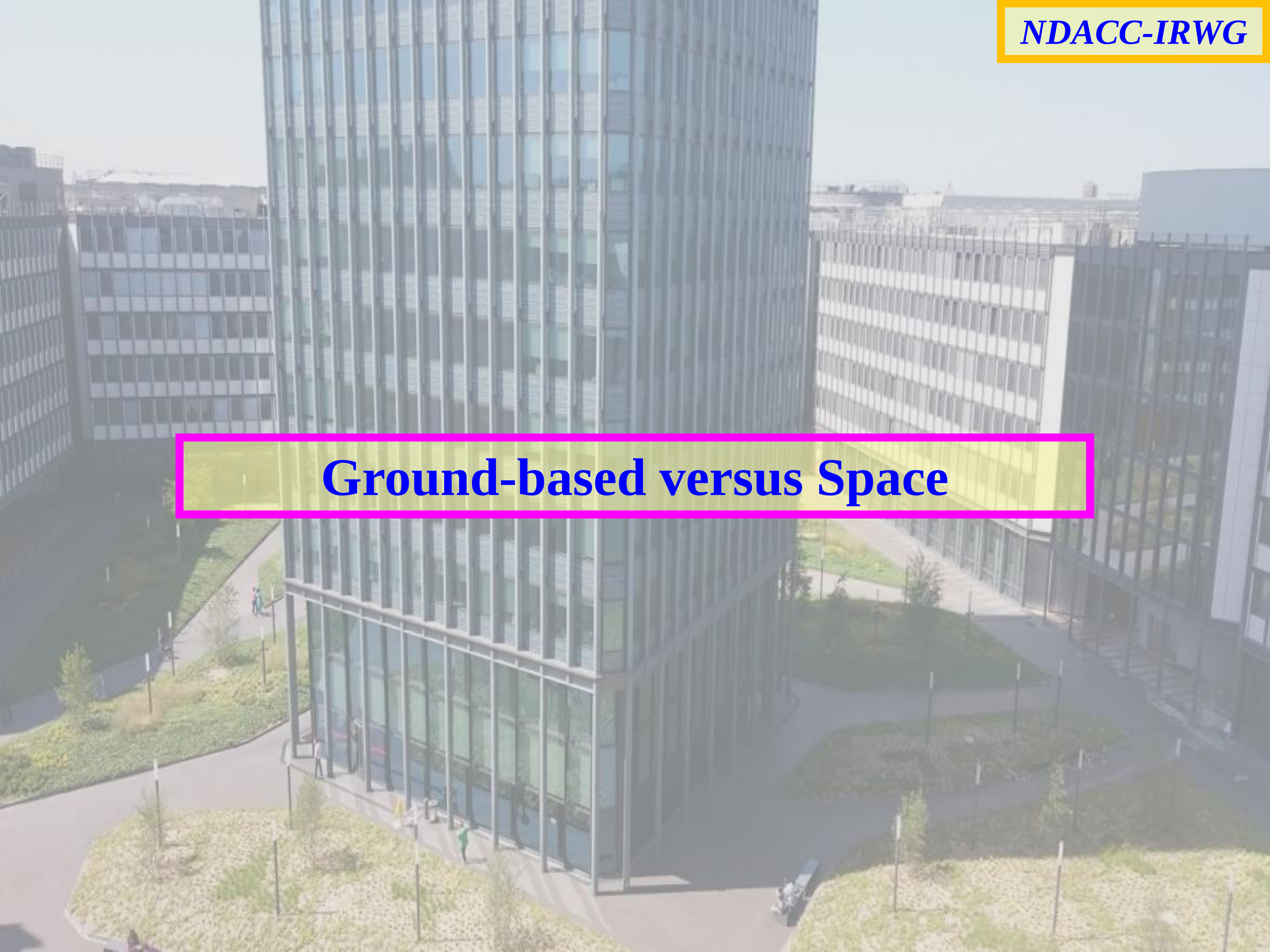
Importance of spectroscopic uncertainties

Hitran 2008:
- **Intensity uncertainties 2-5 %**
- **Air broadening coefficient uncertainties 1-2 %**

But Hitran 2016:
- **Intensity uncertainties 1-2 %**
- **Air broadening coefficient uncertainties 1-2 %**

Uncertainty below 4%

Ground-based versus Space



FTIR measurement comparison (ground versus satellite)

FTS-Paris measurement

Date	FTS-Paris	IASI-a(*)	IASI-b(**)
2009-07-01	2.06±0.04	2.05±0.16	2.11±0.09
2009-07-13	1.73±0.02	2.23±0.09	2.73±0.11
2009-07-16	1.62±0.03	1.90±0.09	2.43±0.11
2010-02-16	2.45±0.05	2.40±0.14	2.53±0.14
2010-03-02	2.57±0.05	2.68±0.11	2.63±0.10
2010-07-07	1.95±0.05	2.15±0.10	2.09±0.10
2010-10-11	1.81±0.03	1.62±0.11	none
2011-03-08	2.77±0.05	2.35±0.12	2.44±0.10
2011-04-19	2.21±0.04	1.91±0.06	2.07±0.05
2011-04-20	2.38±0.03	2.18±0.07	2.22±0.05
2011-04-21	2.23±0.03	1.94±0.06	2.07±0.05
2011-04-22	2.15±0.06	1.93±0.06	none
2011-04-26	2.54±0.04	2.10±0.06	2.22±0.05
2011-05-04	2.83±0.04	2.48±0.06	none
2011-05-05	2.13±0.02	2.36±0.06	2.38±0.05
2011-05-06	2.33±0.05	none	none
2011-05-12	2.16±0.05	none	none
2011-05-13	2.35±0.03	2.28±0.05	2.26±0.05
2011-05-25	2.06±0.03	1.97±0.05	2.04±0.05

* All morning overpasses around Ile de France (±0.5° in latitude and longitude corresponding to a 100 km × 100 km square region centered on QualAir platform location).

** Overpasses inside Paris "downtown" (<±0.15° in latitude and longitude).

IASI-MetOp measurement
~Paris downtown

IASI-MetOp measurement
~100 km × 100 km

(Té *et al.*, ESA Publications, 2012)

FTIR measurement comparison (ground versus satellite)

Date	FTS-Paris	IASI-a(*)	IASI-b(**)
2009-07-01	2.06±0.04	2.05±0.16	2.11±0.09
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2010-10-11	1.81±0.03	1.62±0.11	none
2011-03-08	2.77±0.05	2.35±0.12	2.44±0.10
2011-04-19	2.21±0.04	1.91±0.06	2.07±0.05
2011-04-20	2.38±0.03	2.18±0.07	2.22±0.05
2011-04-21	2.23±0.03	1.94±0.06	2.07±0.05
2011-04-22	2.15±0.06	1.93±0.06	none
2011-04-26	2.54±0.04	2.10±0.06	2.22±0.05
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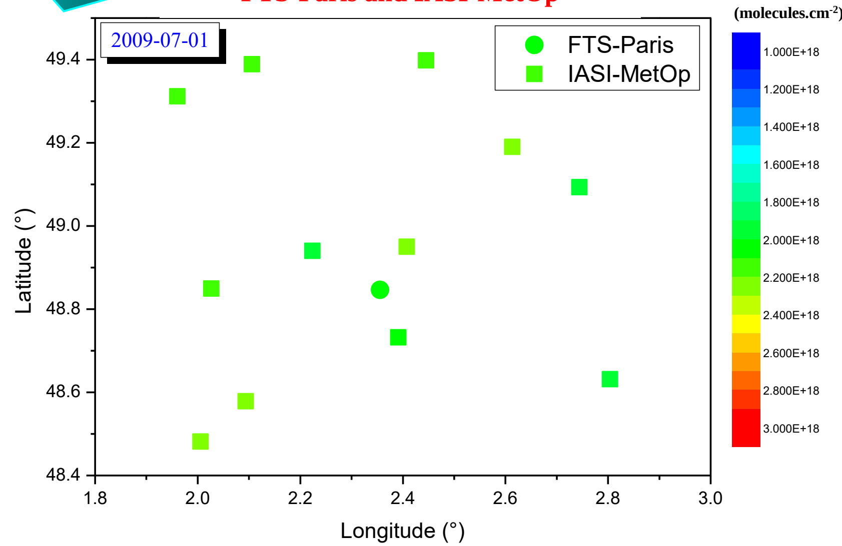
2011-05-05	2.13±0.02	2.36±0.06	2.38±0.05
2011-05-06	2.33±0.05	none	none
2011-05-12	2.16±0.05	none	none
2011-05-13	2.35±0.03	2.28±0.05	2.26±0.05
2011-05-25	2.06±0.03	1.97±0.05	2.04±0.05

* All morning overpasses around Ile de France (±0.5° in latitude and longitude corresponding to a 100 km × 100 km square region centered on QualAir platform location).

** Overpasses inside Paris "downtown" (<±0.15° in latitude and longitude).

(Té *et al.*, ESA Publications, 2012)

CO total columns comparison between FTS-Paris and IASI-MetOp



⇨ **Good agreement between ground and satellite FTIR measurements**

⇨ **Except for 5 days :**

- **13/07/2009**
- **16/07/2009**
- **11/10/2010**
- **22/04/2012**
- **04/05/2011**

FTIR measurement comparison (ground versus satellite)

Date	FTS-Paris	IASI-a(*)	IASI-b(**)
2009-07-01	2.06±0.04	2.05±0.16	2.11±0.09
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2010-07-07	1.95±0.05	2.15±0.10	2.09±0.10
2010-10-11	1.81±0.03	1.62±0.11	none
2011-03-08	2.77±0.05	2.35±0.12	2.44±0.10
2011-04-19	2.21±0.04	1.91±0.06	2.07±0.05
2011-04-20	2.38±0.03	2.18±0.07	2.22±0.05
2011-04-21	2.23±0.03	1.94±0.06	2.07±0.05
2011-04-22	2.15±0.06	1.93±0.06	none
2011-04-26	2.54±0.04	2.10±0.06	2.22±0.05
2011-05-04	2.83±0.04	2.48±0.06	none
2011-05-05	2.13±0.02	2.36±0.06	2.38±0.05
2011-05-06	2.33±0.05	none	none
2011-05-12	2.16±0.05	none	none
2011-05-13	2.35±0.03	2.28±0.05	2.26±0.05
2011-05-25	2.06±0.03	1.97±0.05	2.04±0.05

* All morning overpasses around Ile de France (±0.5° in latitude and longitude corresponding to a 100 km × 100 km square region centered on QualAir platform location).

** Overpasses inside Paris "downtown" (<±0.15° in latitude and longitude).

(Té *et al.*, ESA Publications, 2012)

⇨ For the last 3 days :

- **11/10/2010**
- **22/04/2012**
- **04/05/2011**

⇨ Meas.$_{\text{FTS-Paris}}$ > Meas.$_{\text{IASI-MetOp}}$

- Co-location less satisfying (satellite footprint beyond 30 km from the center of Paris)

- In this case, local emissions are not sounded by the satellite instrument IASI-MetOp

FTIR measurement comparison (ground versus satellite)

Date	FTS-Paris	IASI-a(*)	IASI-b(**)
2009-07-01	2.06±0.04	2.05±0.16	2.11±0.09
2009-07-13	1.73±0.02	2.23±0.09	2.73±0.11
2009-07-16	1.62±0.03	1.90±0.09	2.43±0.11
2010-02-16	2.45±0.05	2.40±0.14	2.53±0.14
2010-03-02	2.57±0.05	2.68±0.11	2.63±0.10
2010-07-07	1.95±0.05	2.15±0.10	2.09±0.10
2010-10-11	1.81±0.03	1.62±0.11	none
2011-03-08	2.77±0.05	2.35±0.12	2.44±0.10
2011-04-19	2.21±0.04	1.91±0.06	2.07±0.05
2011-04-20	2.38±0.03	2.18±0.07	2.22±0.05
2011-04-21	2.23±0.03	1.94±0.06	2.07±0.05
2011-04-22	2.15±0.06	1.93±0.06	none
2011-04-26	2.54±0.04	2.10±0.06	2.22±0.05
2011-05-04	2.83±0.04	2.48±0.06	none
2011-05-05	2.13±0.02	2.36±0.06	2.38±0.05
2011-05-06	2.33±0.05	none	none
2011-05-12	2.16±0.05	none	none
2011-05-13	2.35±0.03	2.28±0.05	2.26±0.05
2011-05-25	2.06±0.03	1.97±0.05	2.04±0.05

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** Overpasses inside Paris "downtown" (<±0.15° in latitude and longitude).

(Té *et al.*, ESA Publications, 2012)

⇨ **For the first 2 days :**
- **13/07/2009**
- **16/07/2009**

⇨ **Meas.$_{\text{IASI-MetOp}}$ > Meas.$_{\text{FTS-Paris}}$**

⇨ **Difference ↗ for a better co-location (between columns 2 and 4)**

FTIR measurement comparison (ground versus satellite)

Date	FTS-Paris	IASI-a(*)	IASI-b(**)
2009-07-01	2.06±0.04	2.05±0.16	2.11±0.09
2009-07-13	1.73±0.02	2.23±0.09	2.73±0.11
2009-07-16	1.62±0.03	1.90±0.09	2.43±0.11
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2011-05-12	2.16±0.05	none	none
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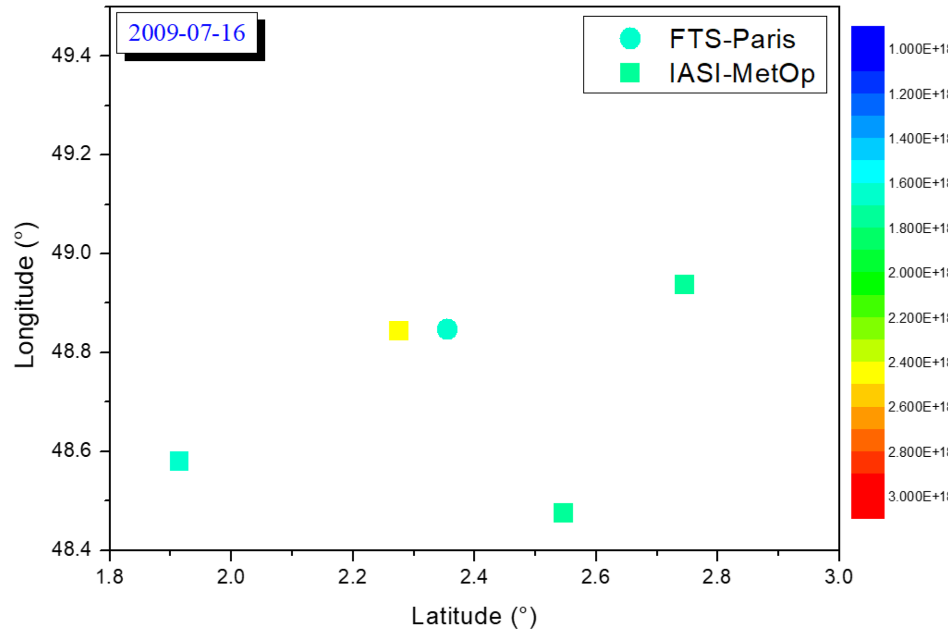
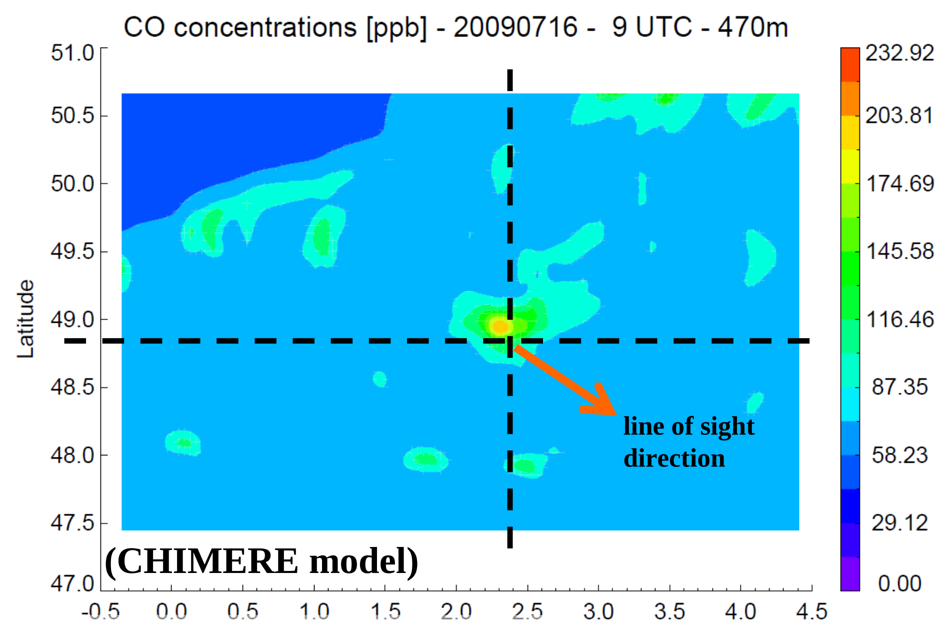
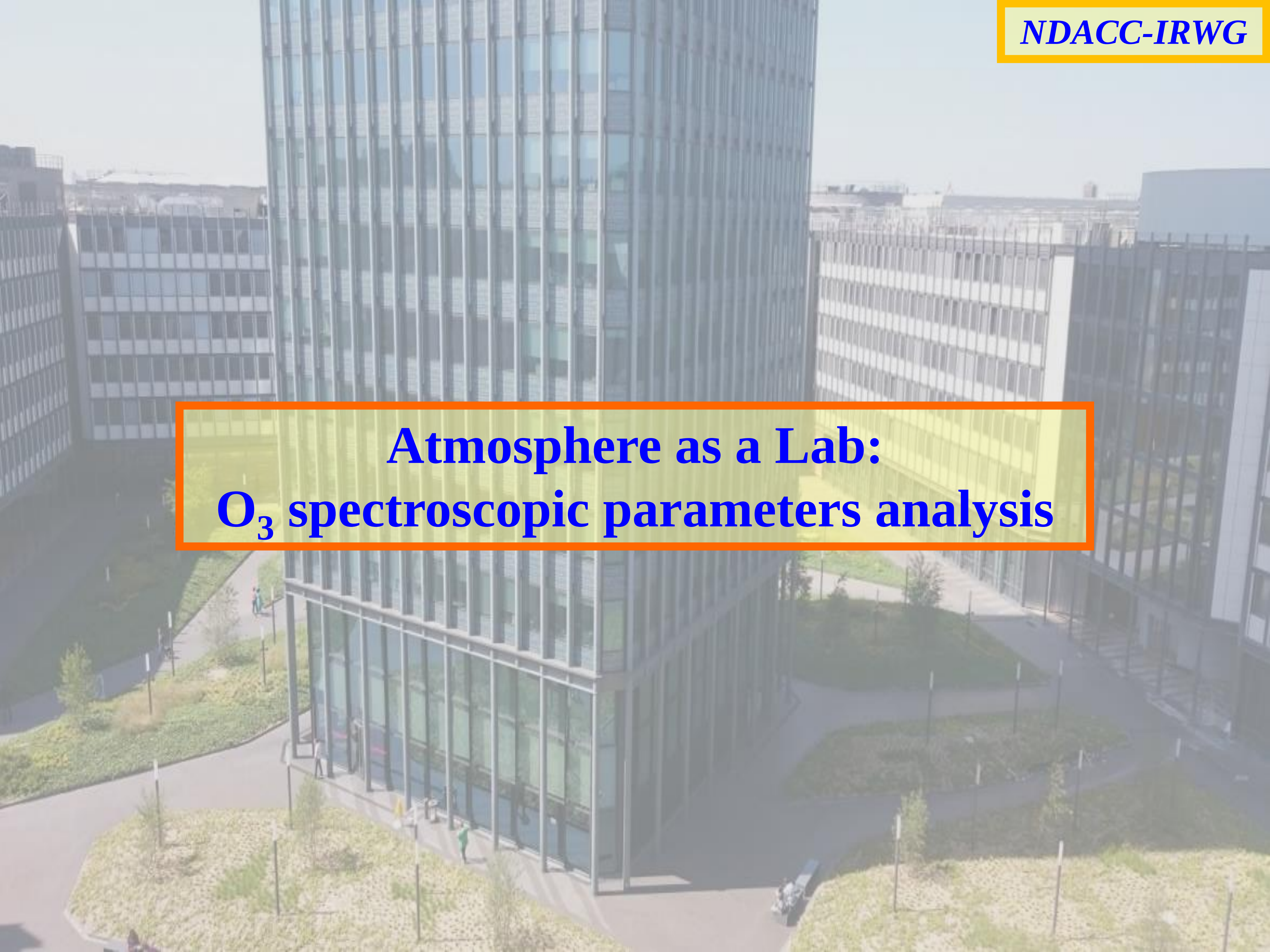
** Overpasses inside Paris "downtown" (<±0.15° in latitude and longitude).

(Té *et al.*, ESA Publications, 2012)







Atmosphere as a Lab:
O_3 spectroscopic parameters analysis

Motivations

- Atmospheric ozone concentration sounded by different instrumentations

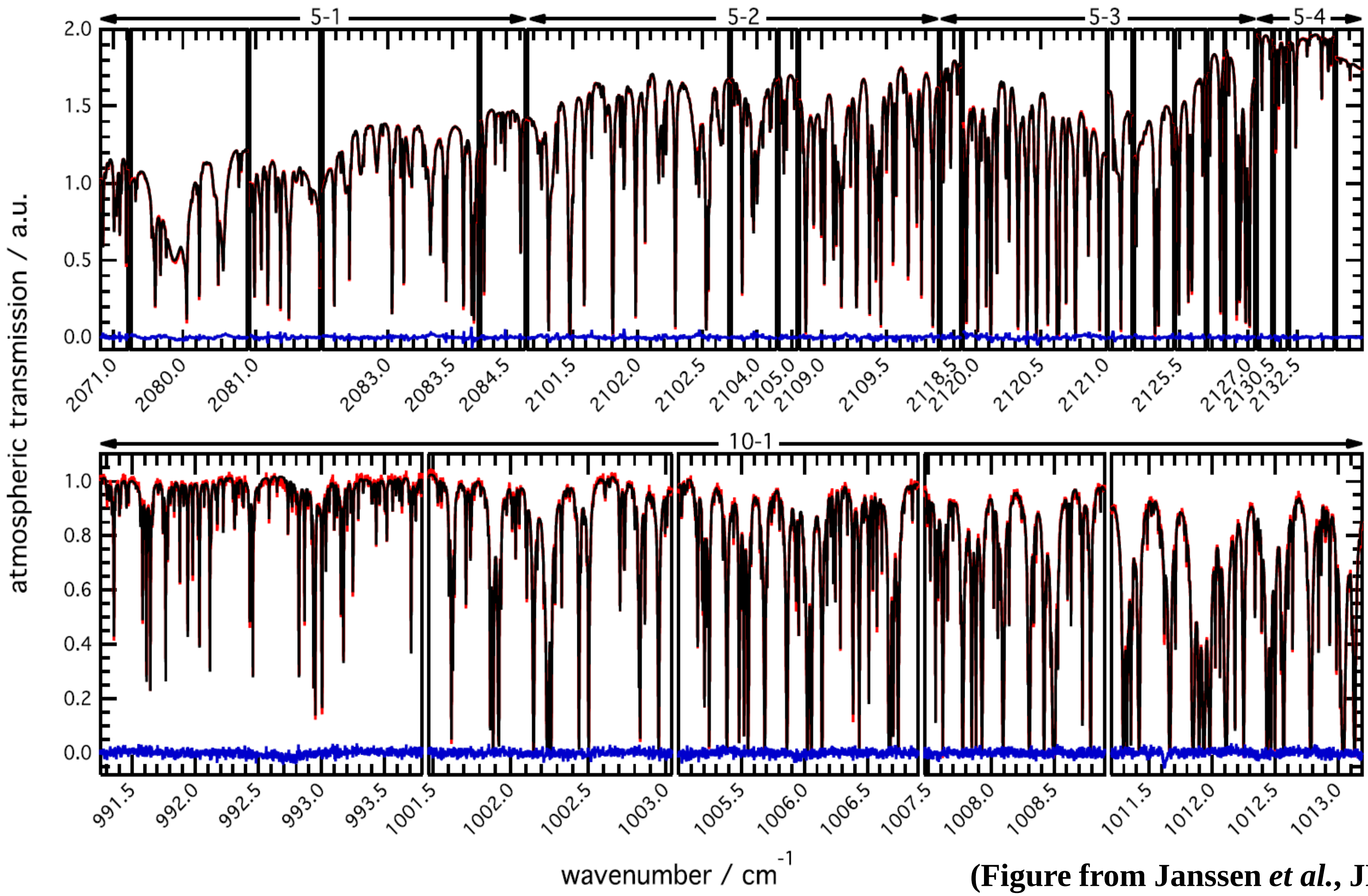
  ⇨ from different platforms (ground, balloon, satellite …)

  ⇨ in different spectral domains (UV & IR)

- Despite long years measurements efforts, 1% uncertainty in absolute line intensities not reached

=> Multispectral inter-comparison using both laboratory and atmospheric studies reveal important discrepancies in databases

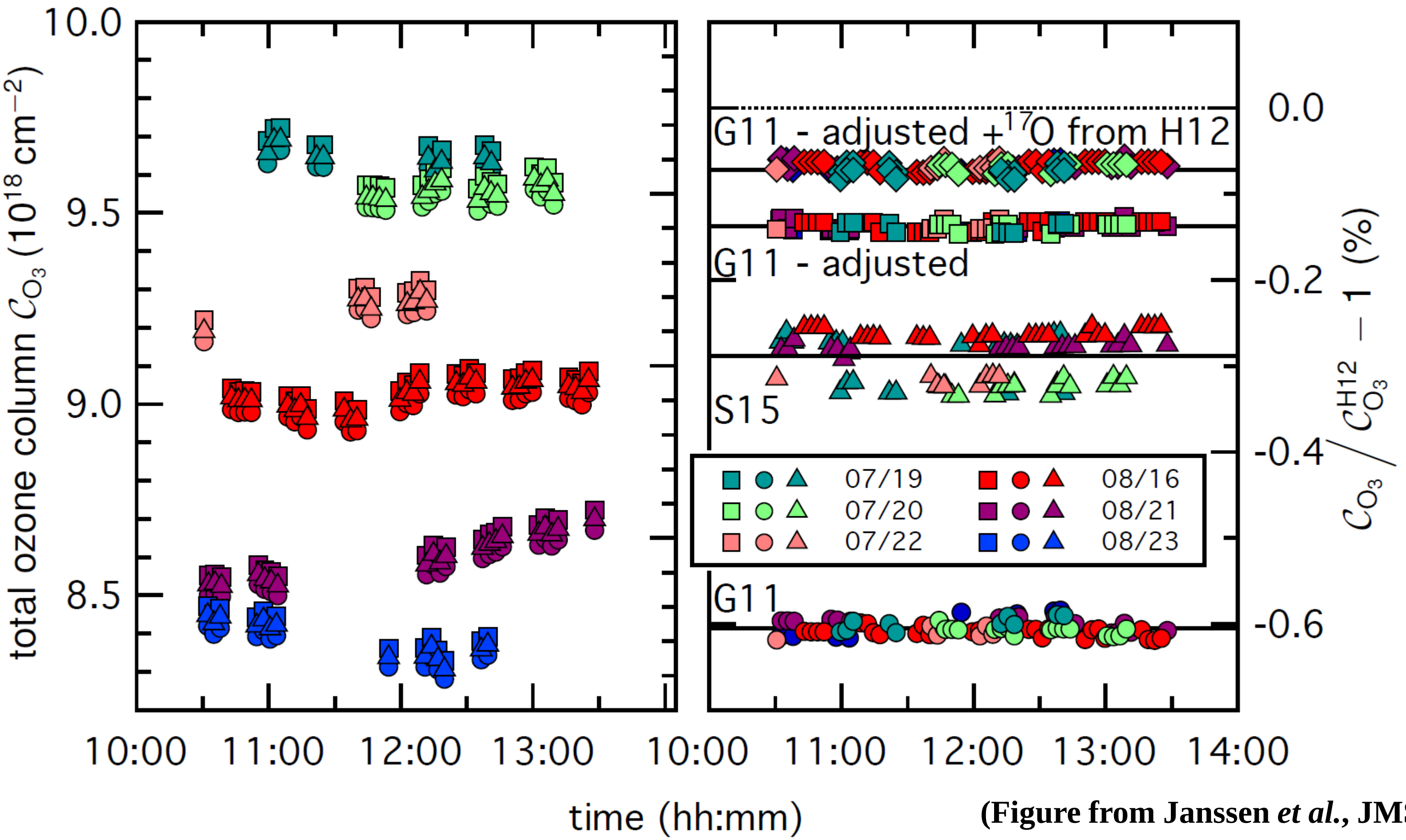
Spectral windows used for the ozone retrieval



(Figure from Janssen *et al.*, JMS 2016)

- Three spectroscopic databases studied: HITRAN2012, GEISA2011, S&MPO2015
- Six days of atmospheric measurements

Ozone retrieval @10 µm

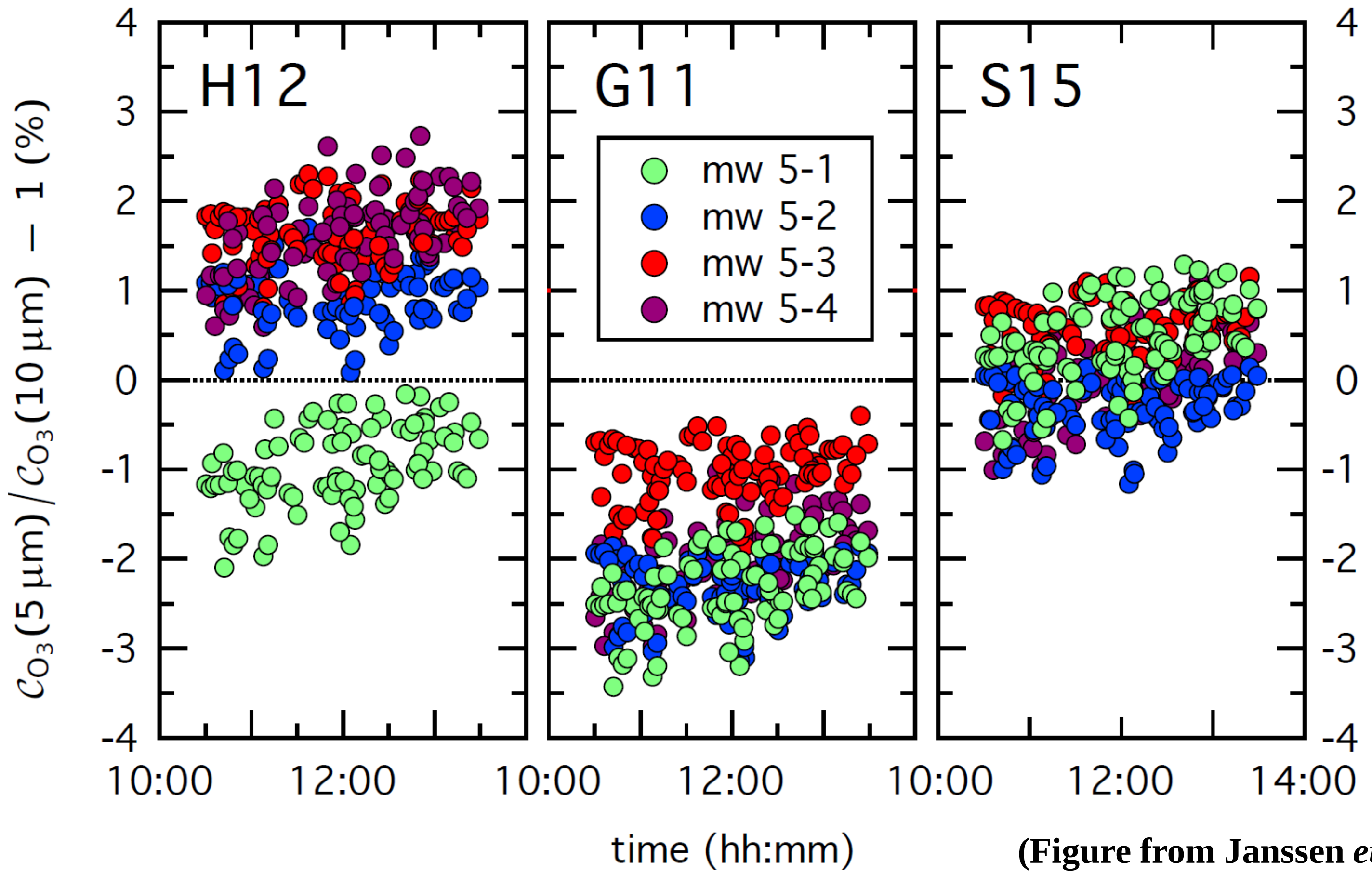


(Figure from Janssen *et al.*, JMS 2016)

⇒ Good agreement between G11 and H12 with discrepancies ~0.6%

⇒ Good agreement between S15 and H12 with discrepancies ~0.3%

Ozone retrieval @5 µm



(Figure from Janssen *et al.*, JMS 2016)

⇒ HITRAN2012: agreement with 10 µm data within ±2%, but disagreement of [5-1] vs. others

⇒ GEISA2011: good self consistency, bias of ~4% with respect to HITRAN 5 µm

⇒ S&MPO2015: best self consistency, bias within ±1 % with respect to HITRAN 5 µm

⇒ Recommendation on O_3 taken into account in HITRAN 2016

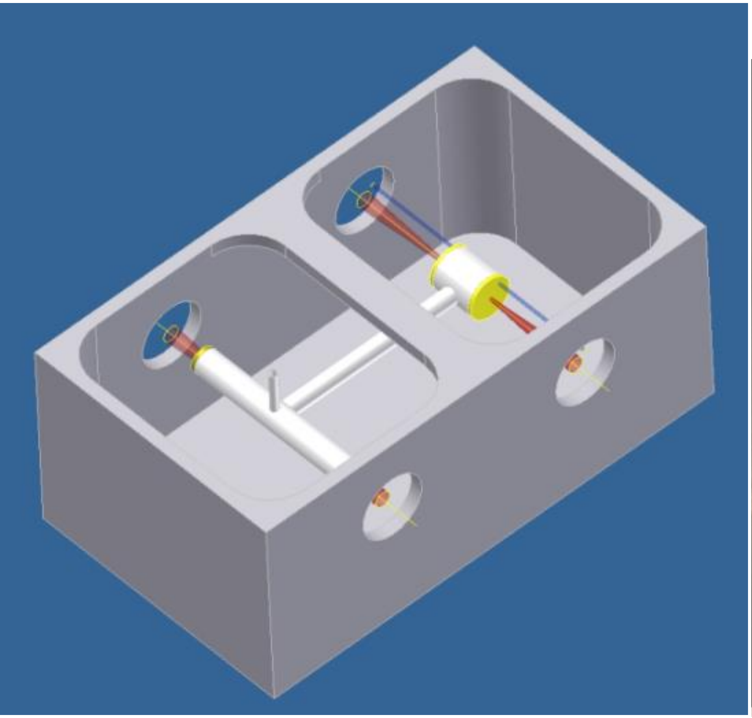
SMO_3 project

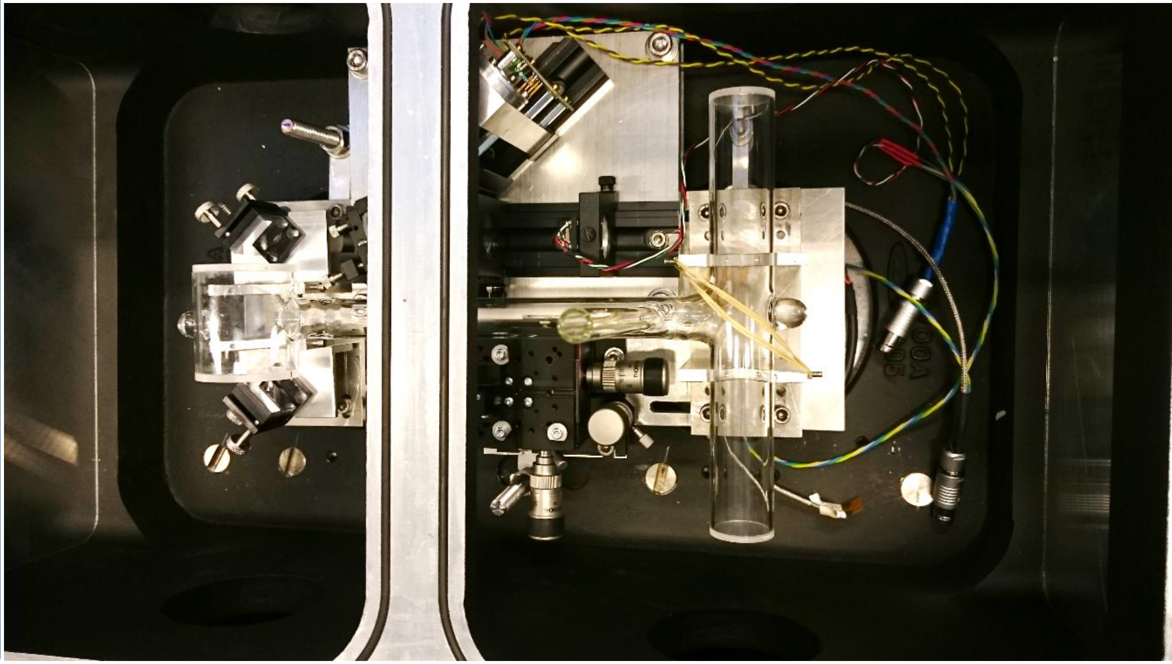
- Spectroscopie Multi-spectrale de l'Ozone (SMO_3)

- Funding from INSU LEFE-CHAT

- Relative and absolute line intensities (UV & IR)

- Lab & atmospheric measurements & ab-initio calculation (MONARIS, GSMA)





NDACC-IRWG

First evidence of a time lag between surface and free tropospheric CO seasonal variations

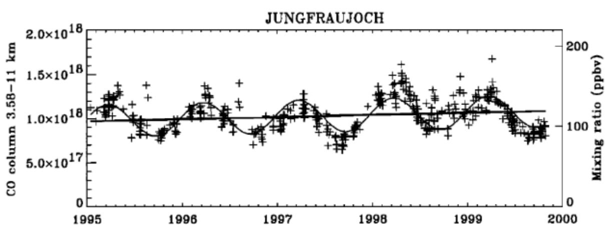
Motivations (1/2)

- CO is an important trace gas (toxicity and impact on air quality)

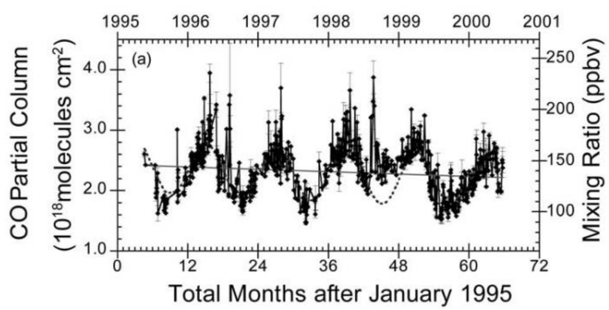
- Global increase of CO ⇨ global decrease of OH
⇨ increase of other harmful trace gases (atmospheric pollutants, greenhouse gases sensitive to oxidation as methane)

- Many scientific studies have shown the seasonal variability of CO

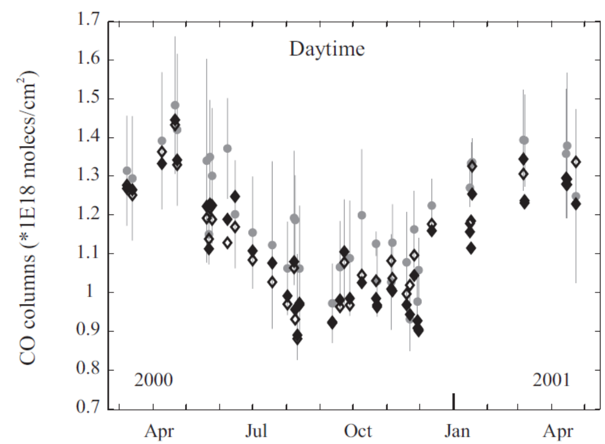
- What can be learned from still another study ?



(Rinsland *et al.*, JGR, 2000)



(Zhao *et al.*, JGR, 2002)



(Barret *et al.*, ACP, 2003)

Motivations (2/2)

<u>Here</u>

- CO seasonal variability between 2009 and 2013
  - @ Paris site (NH)
  - @ Jungfraujoch site (NH)
  - @ Wollongong site (SH)

Seasonality from CO columns obtained from FTIR measurements **VERSUS** Seasonality from surface in situ measurements

⇨ **Might not be the same**

- Comparison to satellite measurements (IASI-MetOp & MOPITT)

- Comparison to GEOS-Chem model simulations

Three ground-based FTIR sites

FTS-Paris

- Located at the UPMC University in the center of Paris
- Urban megacity site
- TCCON station

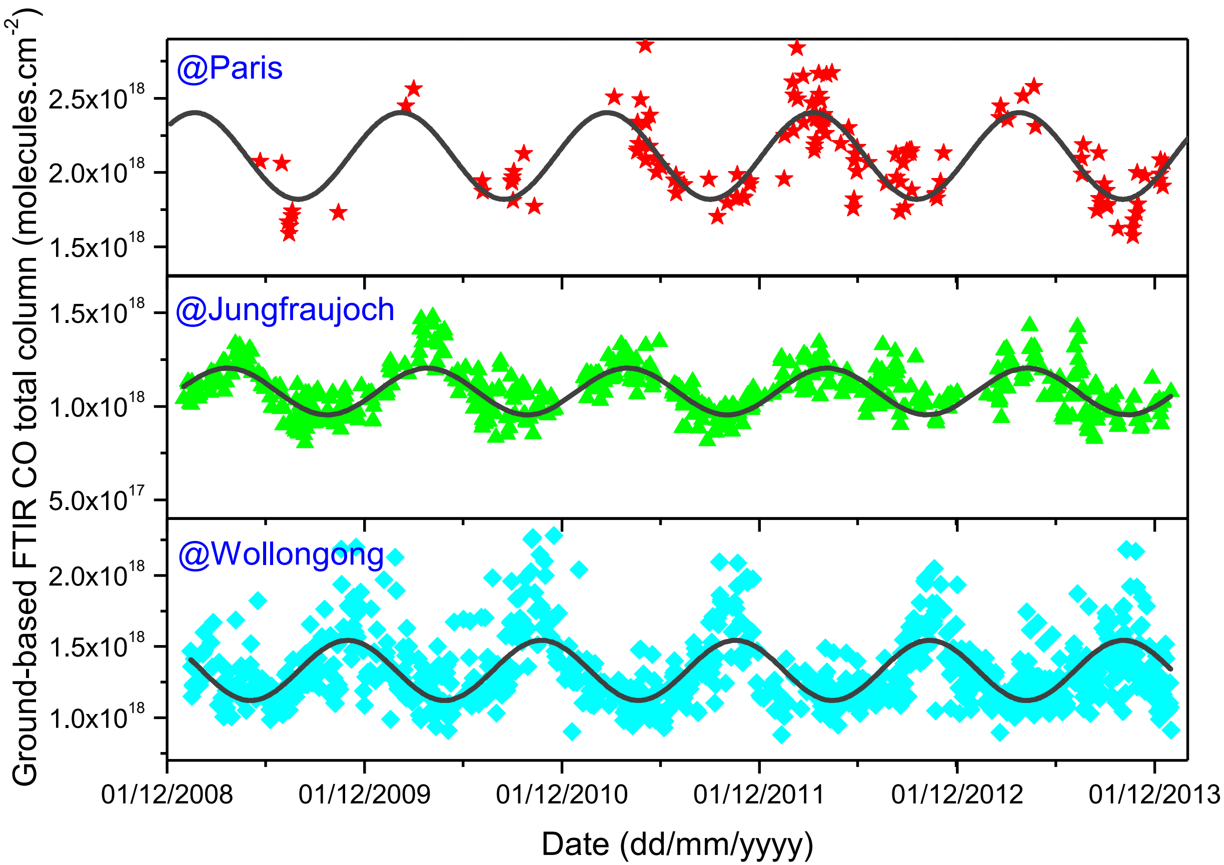
Jungfraujoch FTIR

- Located at the ISSJ
- Remote high-altitude site
- NDACC station

Wollongong FTIR

- Located at Wollongong University
- Moderate pollution site
- NDACC & TCCON station

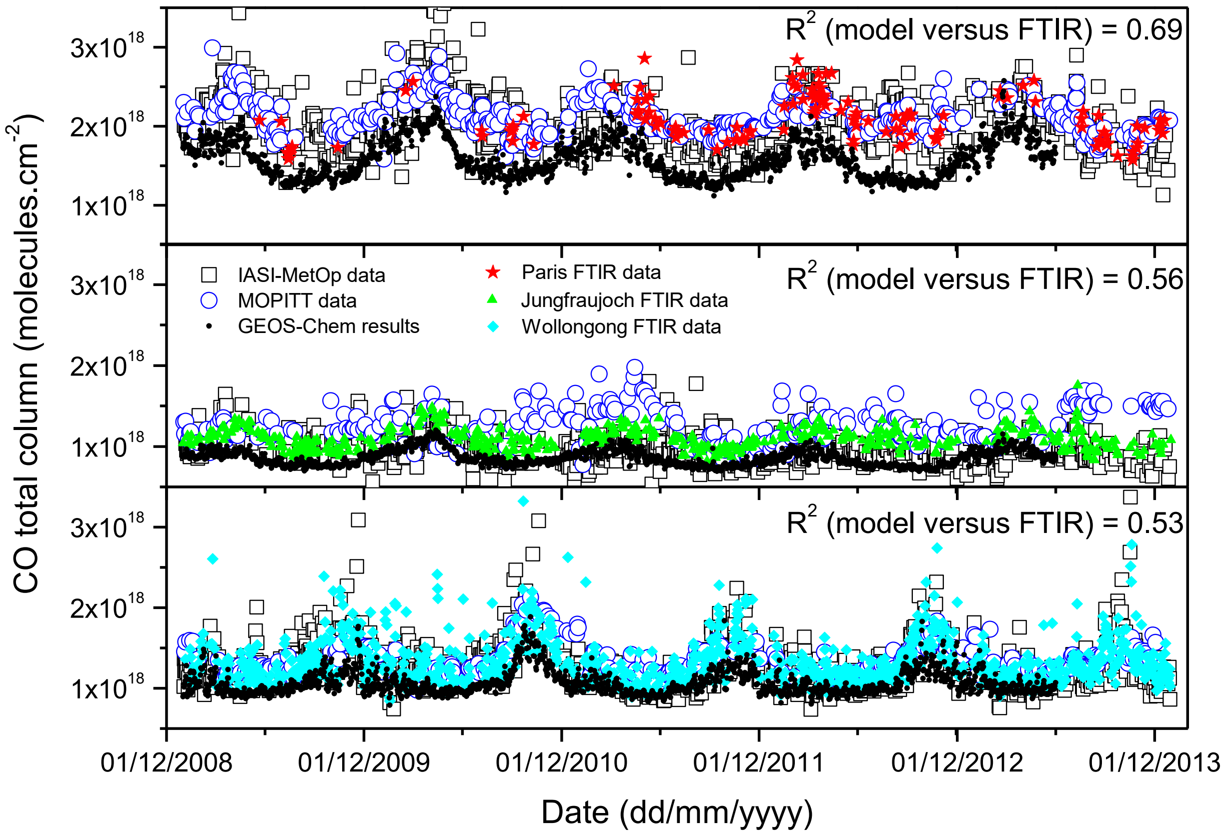
Total column seasonal variability (1/2)



Consistency of the observed CO seasonality with previous FTIR studies (Rinsland *et al.*, Zhao *et al.*, Barret *et al.*, …)

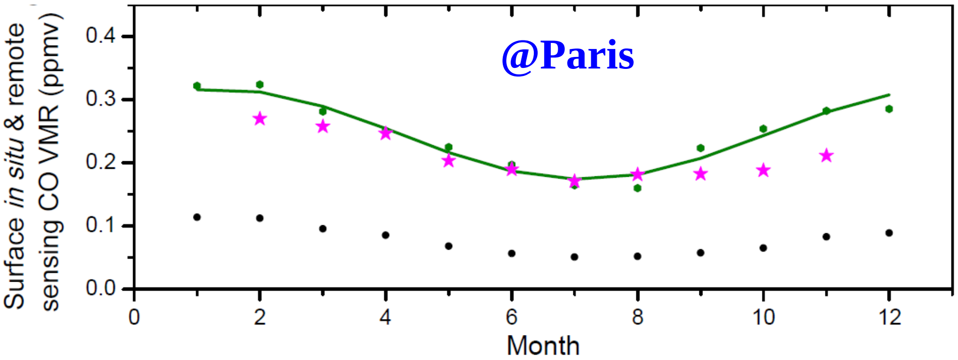
Total column seasonal variability (2/2)

(Té *et al.*, ACP, 2016)

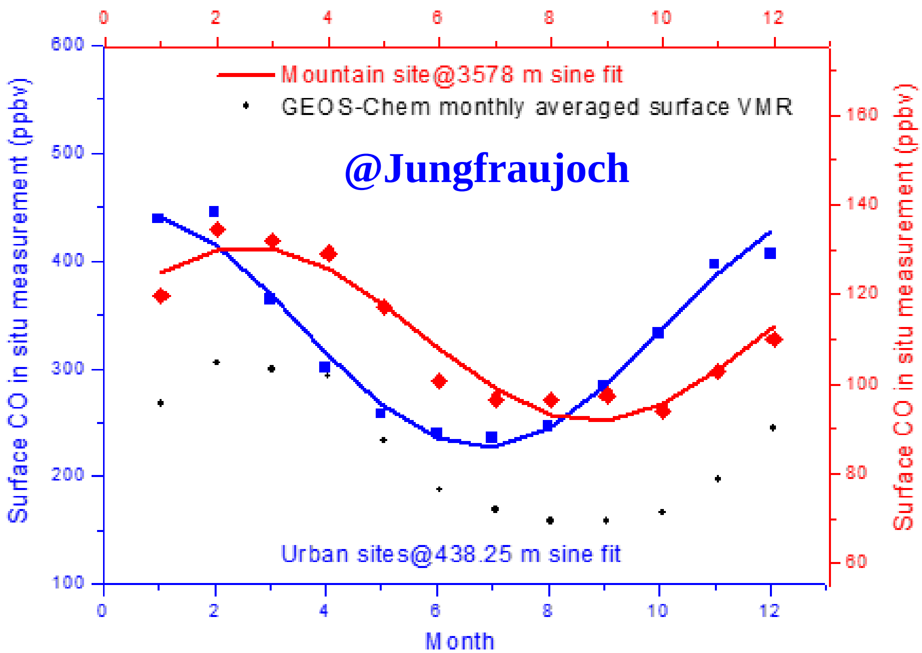


- **Good agreement between ground-based FTIR, satellites and GEOS-Chem for the CO seasonal variability**
- **But underestimation of about 20% by the model (probably due to the currently implemented inventories)**

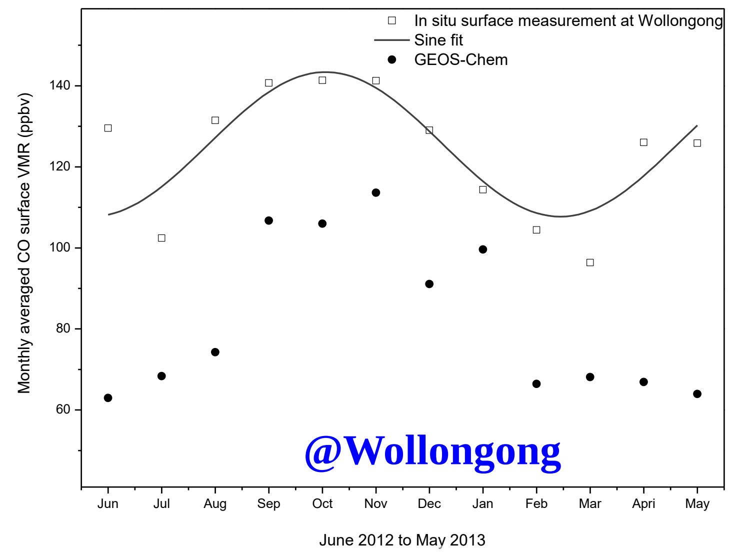
Mesure FTIR versus mesure *in situ*



[Paris & Jungfraujoch] :
Time-lag of about 2 months
between surface and column CO



[Wollongong] :
No significant time-lag observed



GEOS-Chem simulations : sources identification (1/2)

- To study the impact of the different sources of CO, three specific simulations were performed by turning off individually

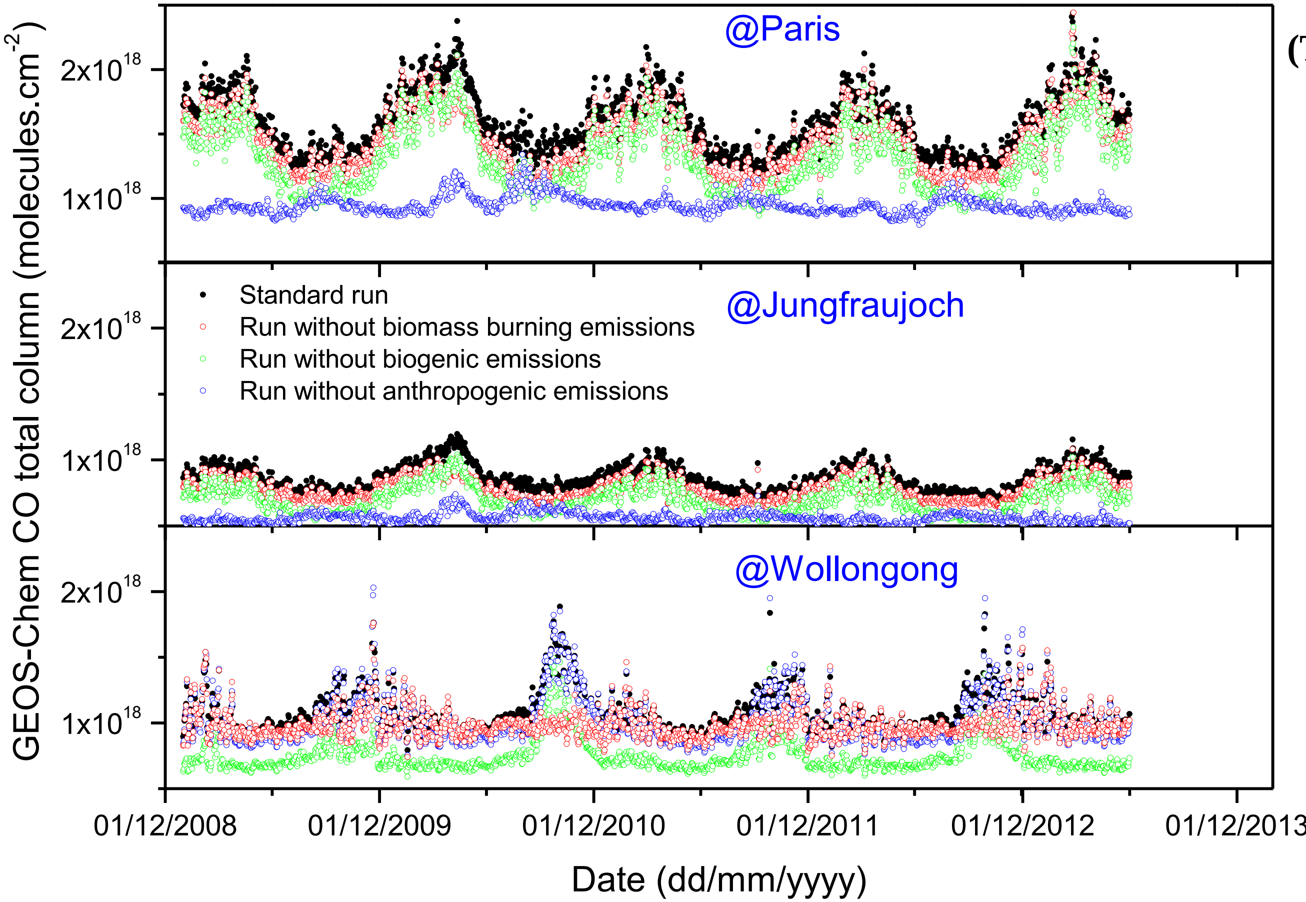
⇨ the biomass burning emission sources

⇨ the biogenic emission sources

⇨ the anthropogenic emission sources

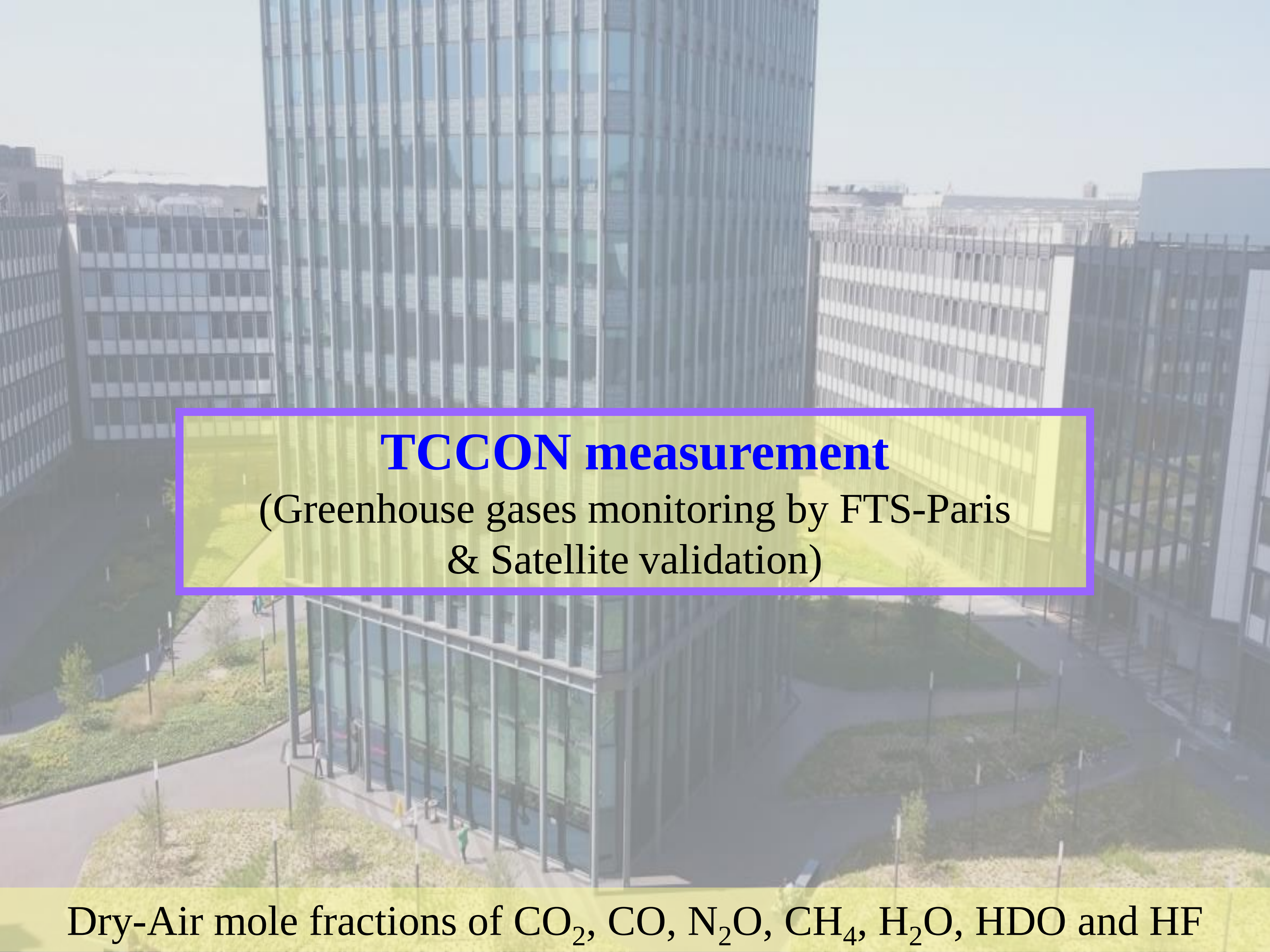
GEOS-Chem simulations : sources identification (2/2)

(Té *et al.*, ACP, 2016)



⇨ The CO seasonality at Paris and Jungfraujoch is mainly driven by anthropogenic emission ➔ Time-lag of 2 months between surface and column

⇨ The CO seasonality at Wollongong is influenced by remote or uniformly distributed biogenic and biomass burning emission sources ➔ No significant time-lag observed



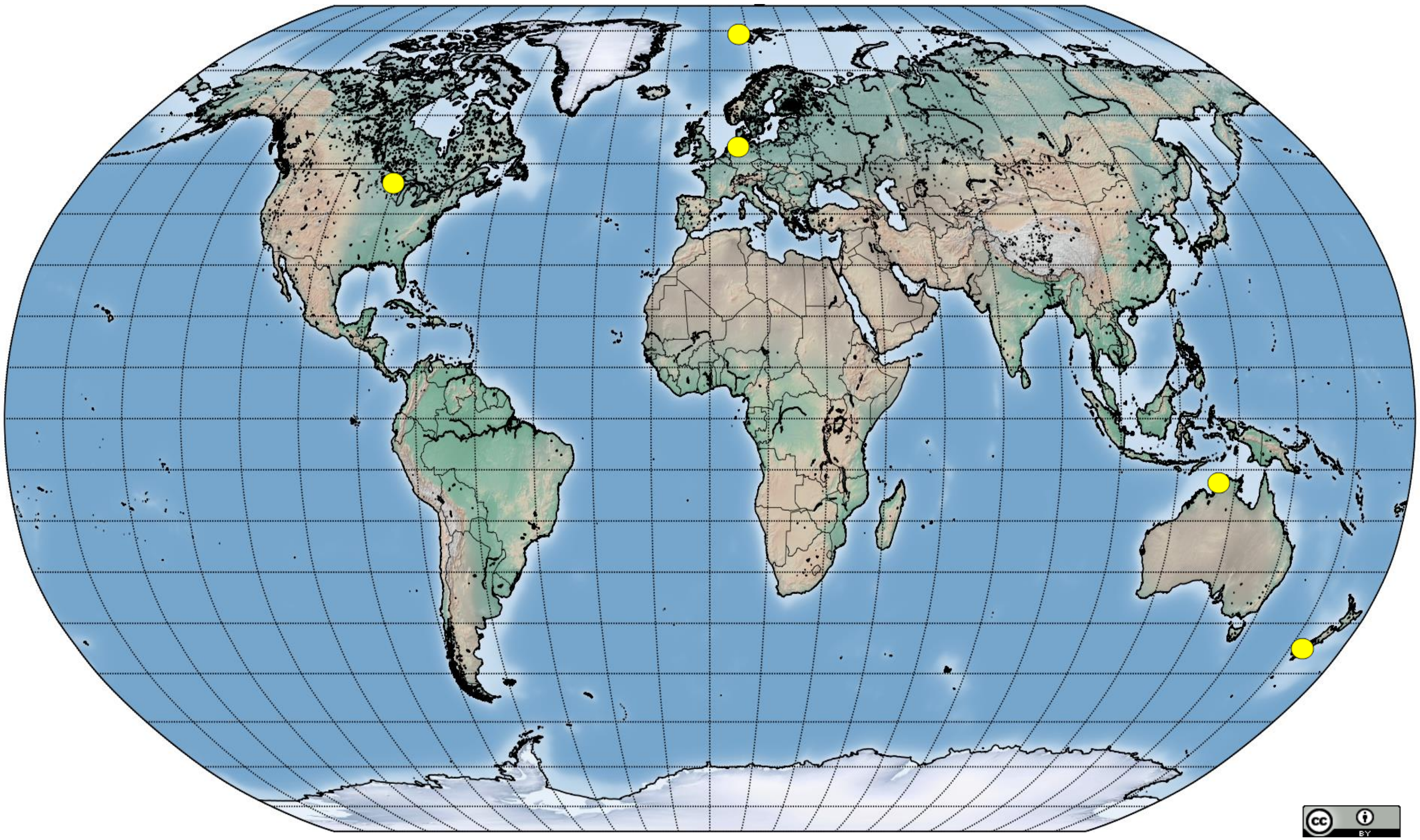
TCCON measurement

(Greenhouse gases monitoring by FTS-Paris
& Satellite validation)

Dry-Air mole fractions of CO_2, CO, N_2O, CH_4, H_2O, HDO and HF

Total Carbon Column Observing Network (TCCON) 2005

➔ TCCON network has started in 2005 with only 5 sites dedicated to greenhouse gases observation



Total Carbon Column Observing Network (TCCON) 2016

➔ TCCON network has grown strongly : 25 sites over the world

- **Absolute measurements**
- **X_{CO2} accuracy at 0.5 ppmv or 0.1% (dry air mole fraction of CO_2)**

Courtesy of D. Feist

TCCON-Paris, the first TCCON site in an European megacity

➔ **Paris agglomeration is the 3rd largest megacity in Europe**

➔ **The FTS-Paris instrument joint the network in 2014**

➔ **TCCON = international network using and /or following :**

⇨ **Same kind of instrumentation (IFS model from Bruker industry)**

⇨ **Same optical alignment method**

⇨ **Same radiative transfer code (GFIT)**

https://tccon-wiki.caltech.edu/Sites/Paris

- › Indianapolis
- › Izaña
- › Jena
- › JPL
- › Karlsruhe
- › Lamont
- › Lauder
- › Manaus
- › Ny-Ålesund
- › Orléans
- › Oxfordshire
- ˅ Paris
  - Instrument History
- › Park Falls
- › Poker Flat
- › Reunion Island
- › Rikubetsu
- › Saga
- › Sodankylä
- › Tsukuba
- › Wollongong
- › Yekaterinburg

Paris, France

TCCON Status: Provisional

48.846° N, 2.356° E, 60 meters above sea level

FTS-Paris : Bruker IFS 125HR

Operated by LERMA (Laboratoire d'Etudes du Rayonnement
Atmosphères, UMR 8112), Université Pierre et Marie Curie /
Collaborators : Yao Té (PI), Pascal Jeseck, Christof Janssen

GFIT radiative transfer code

- Actual version of GFIT : GGG2014

- Line-by-line algorithm developed by Geoff Toon (Wunch *et al.* 2011)

- Forward model and inversion model in column (scaling)

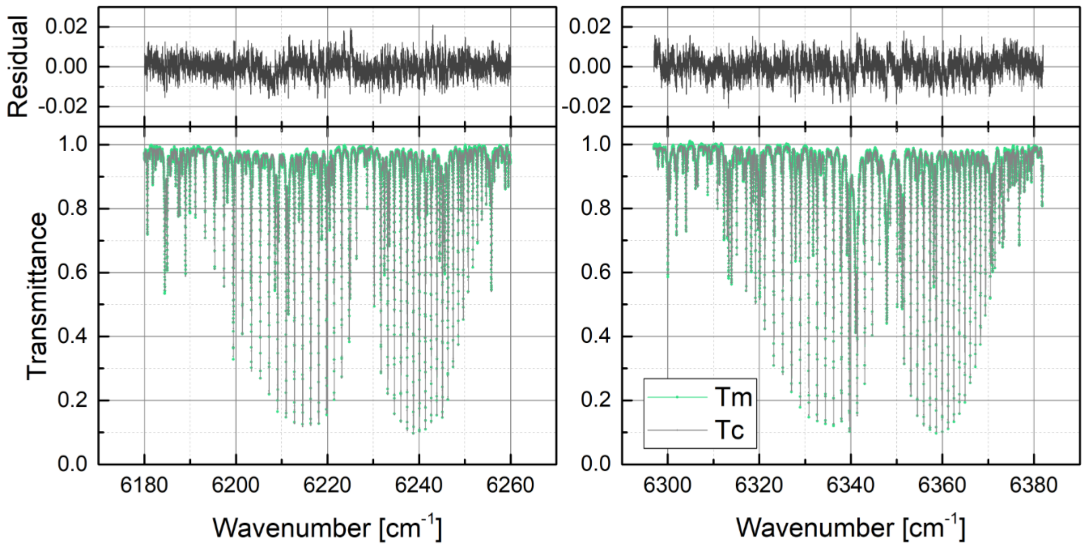
- Voigt line profile
- NCEP profiles of pressure, temperature and H_2O
- Semi-empirical *a priori* profiles, …

- Results in 'dry air mole fraction' *Xgas* defined by : $Xgas = 0.2095 \frac{colonne_{gas}}{colonne_{O2}}$

➔ XCO_2 seasonality and trends over Paris megacity

➔ TCCON data are calibrated to the WMO standard (World Meteorological Organization)

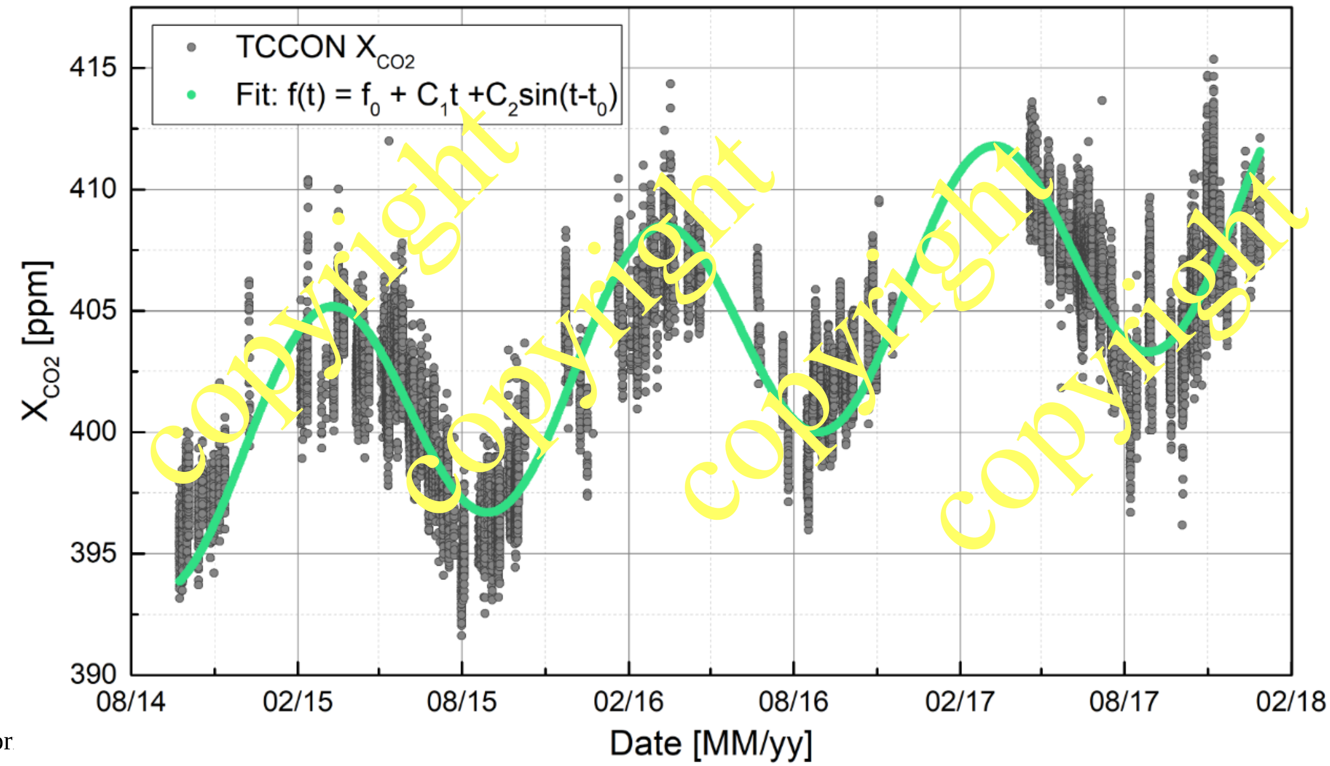
TCCON XCO_2 (seasonality & trends)



⇒ Seasonal amplitude:
~8-10 ppm (peak-peak)

⇒ Trends: **~3-4 ppm / year**

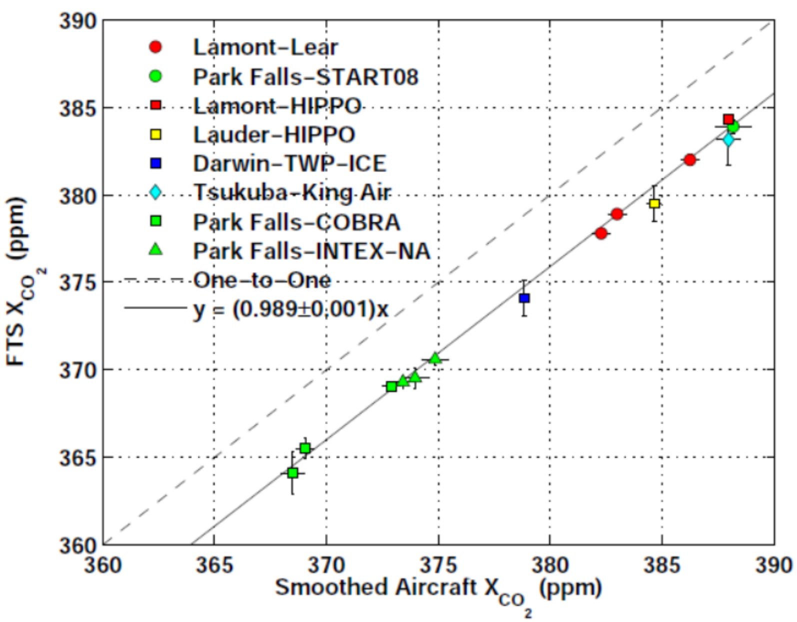
(Koshelev *et al.*, EGU, 2018)

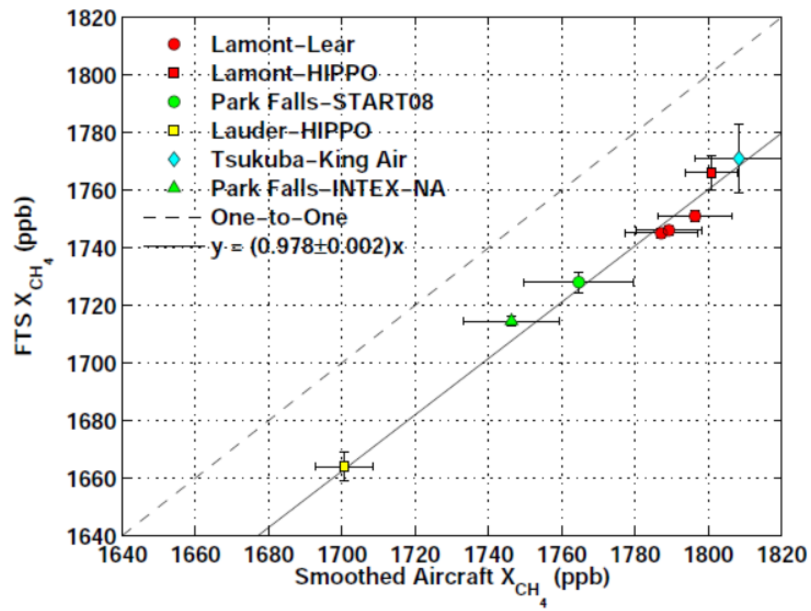


Calibration of TCCON data

➔ **Aircraft flight over TCCON stations boarding *in situ* instruments related to the WMO standard**

➔ **TCCON data compared to aircraft measurements**





(Figures from Wunch *et al.*, AMT, 2010)

Relevance of TCCON for greenhouse gas measurements by satellites

SCIAMACHY

CO_2 and CH_4

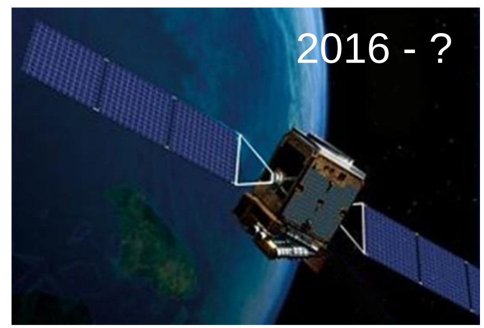
GOSAT

CO_2 and CH_4

OCO-2

CO_2

TANSAT



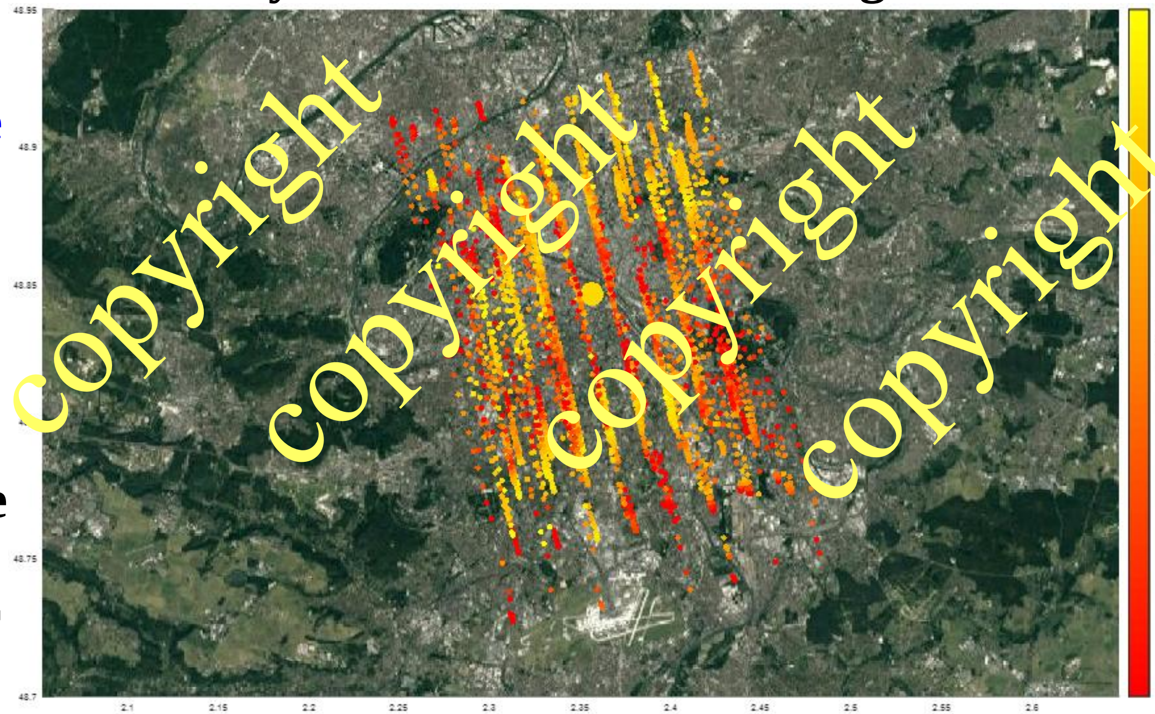
- Validation of satellite data (spatial bias, temporal drift)
- Indirect calibration of satellite data versus *in situ* WMO standard

On going satellite instruments validation

➔ TCCON-Paris station selected since July 2015 to validate the 'Target' mode of OCO-2

⇨ Characterization of possible bias related to the latitude, To the surface properties, to the aerosol scattering, …)

(Wunch *et al.*, AMT, 2017)



Date: 2016-03-11
X_{CO2} scale: max = 410 ppmv - min = 400 ppmv;
TCCON X_{CO2}: 408.2 ppmv

➔ Contribution to validate the Japanese instrument TANSO-FTS onboard GOSAT

(Uchino *et al.*, TCCON meeting, 2018)

➔ Contribution to validate the European instrument TROPOMI onboard Sentinel 5P

(Sha *et al.*, EGU, 2018), (Vigouroux *et al.*, EGU, 2018)

➔ Contribution to validate the radiative transfer algorithm RemoTeC

(Wu *et al.*, AMT, 2018)

Upcoming GHG measurement @Paris

➔ **Involvement in the preparation and the validation of coming up space missions (MicroCarb, MERLIN, IASI-NG, GOSAT-2, OCO-3, Sentinel 7, …)**

➔ **Importance of the greenhouse gas in global warming**

➔ **More and more complex atmospheric modelling**

⇒ **Increasing need of more and more precise and accurate data**

➔ **Set-up of the French COCCON consortium**

(Collaborative Carbon Column Observing Network)

⇒ **COCCON measurement using low resolution EM27/sun instrument**

⇒ **Mobile EM27/sun instrument**

⇒ **TCCON site by site bias study**

⇒ **High and unique potential when deploying few EM27/sun**

⇒ **Validation of satellite instruments**

⇒ **Contribution of better quantify CO_2 emission fluxes**

EM27/sun instrument (COCCON measurement)

- Rock Solid™ pendulum interferometer
  - 2 cube corners
  - CaF_2 beamsplitter
- MOPD: 1,8 cm; resolution: 0,5 cm^{-1}
- Double sided interferograms
- InGaAs detector
  - Spectral range: 4000 cm^{-1} – 9000 cm^{-1}
- SemiFOV: 2.36 mrad
- Standard non frequency-stabilized HeNe reference laser
- Dimensions: 35 x 40 x 27 cm
- Mass: ~25 kg with tracker
- Tracker unit including tracking software developed by M. Gisi

COCCON campaign @Paris

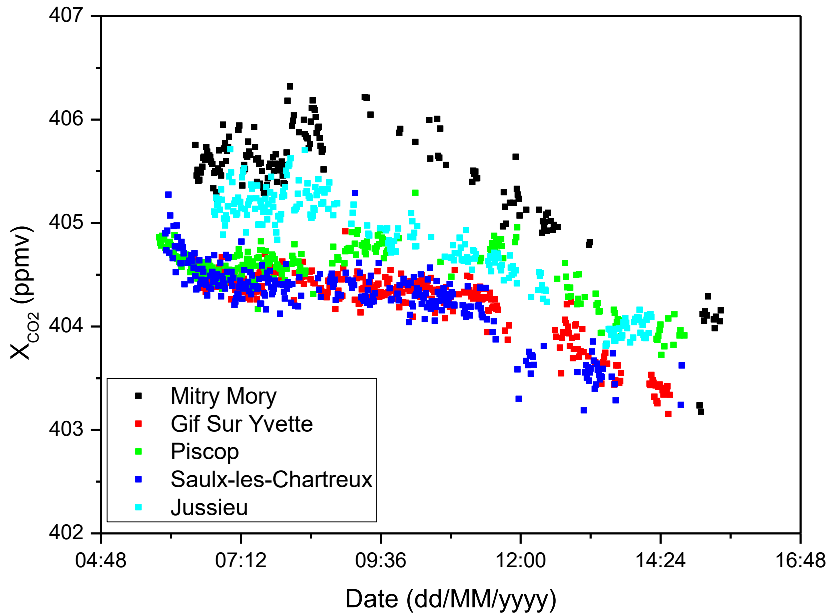
➔ **Co**llaborative **C**arbon **C**olumn **O**bserving **N**etwork

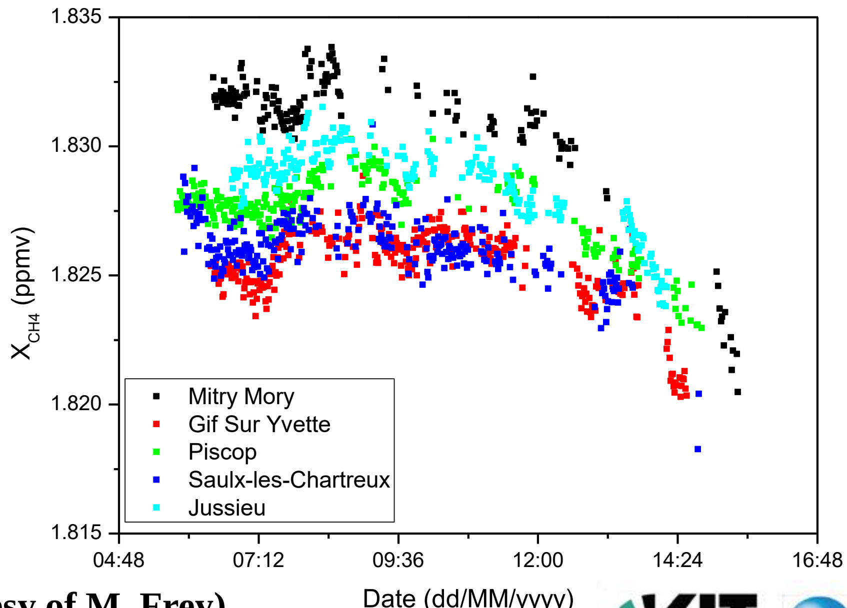
➔ Measurement during Spring 2015

➔ 5 EM27/sun deployed around Paris

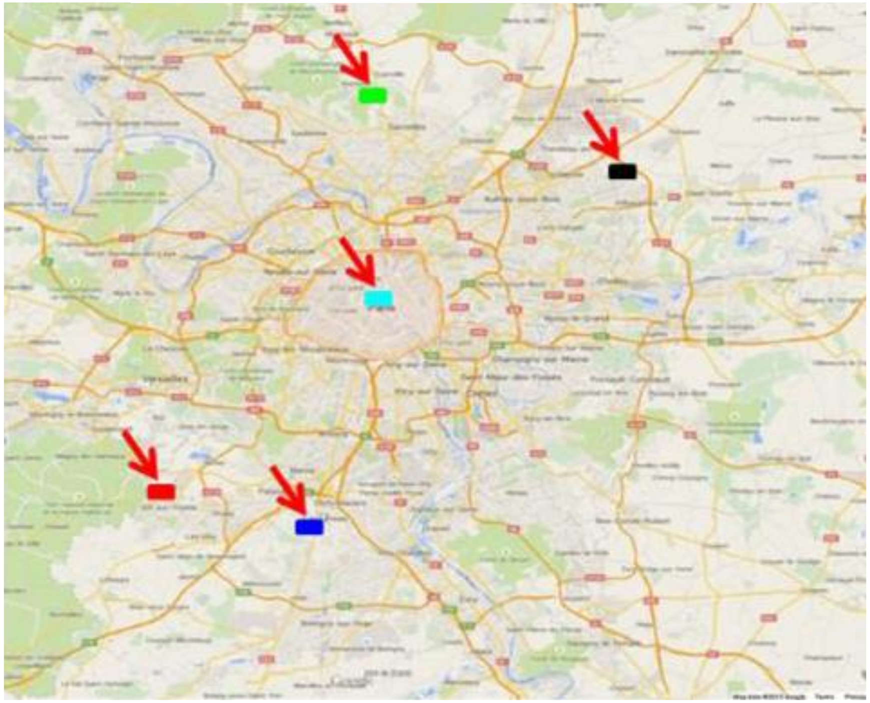
⇒ Impact of urban emission on atmospheric CO_2 and CH_4

Daily XCO_2 and XCH_4 [07/05/2015]





➔ 5 EM27/sun from KIT deployed around Paris



(Vogel *et al.*, AMTD, 2018)

(Courtesy of M. Frey)

2017 Annual Joint NDACC-IRWG & TCCON meeting hosted by the LERMA at the TCCON-Paris station

Laboratoire d'Études du Rayonnement et de la Matière en Astrophysique et Atmosphères