8-years time evolution of stratospheric HNO3 columns: investigation of the drivers of variability and of the link to O3

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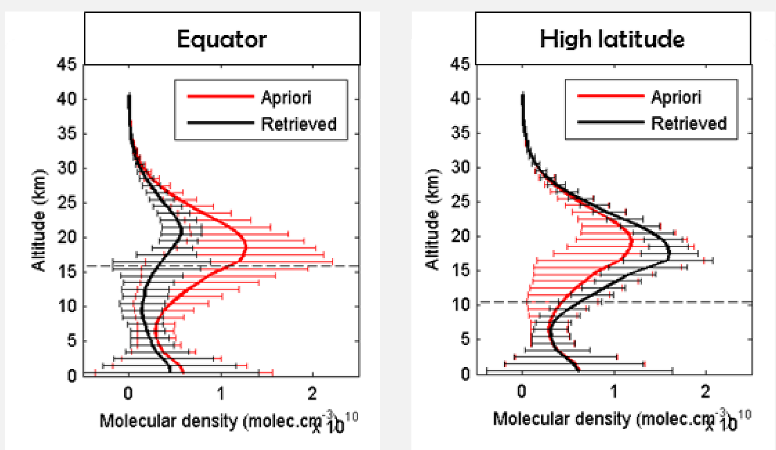
Introduction

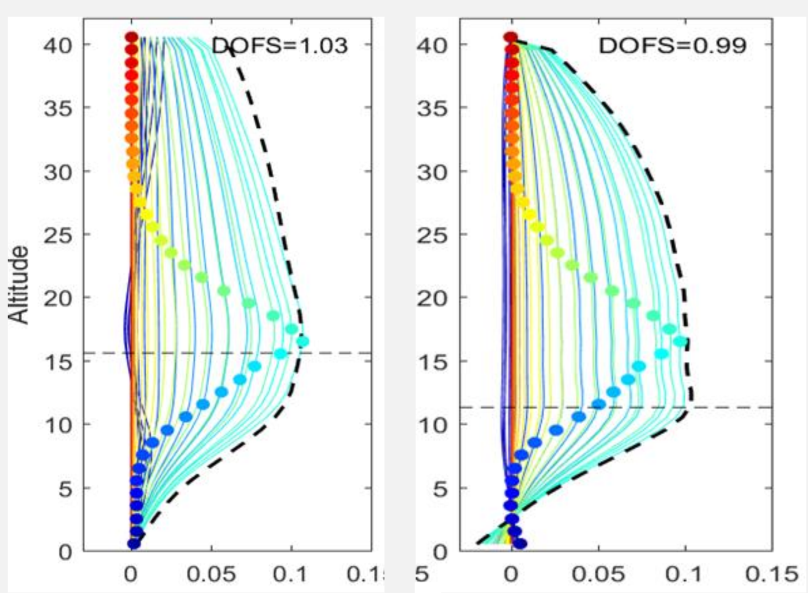
Nitric acid (HNO_3) is one of the main species involved in the stratospheric ozone cycles. Until recently, the distribution of HNO_3 in the stratosphere was available only from limb-sounding instruments, such as MLS, MIPAS or ACE-FTS. The IASI nadir looking thermal infrared instrument is now providing HNO_3 concentration distributions with unprecedented spatial and temporal sampling since 2007. Here, we briefly review the IASI observational capabilities for HNO_3 (in terms of vertical sensitivity and errors) and show the results of a validation exercise made through a comparison with ground-based FTIR measurements.

We also provide an analysis of the time evolution of HNO_3 concentrations from IASI measurements since the end of 2007, through global distributions as well as latitudinal time series of HNO_3 columns. The first results of a multivariate regression analysis are shown in order to highlight the various factors responsible for HNO_3 spatial and temporal variability. The capabilities of the IASI instrument also allows for a joint analysis between HNO_3 and O_3 evolutions throughout the years, also briefly described hereafter.

FORLI-HNO_3

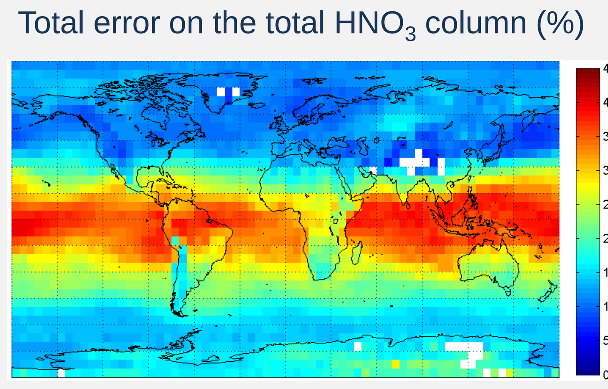
- Fast Optimal Retrieval on Layers for IASI = near-real time processing chain, at ULB
- Retrieval range: *860-900 cm^{-1}*; Retrieval scheme: *Optimal Estimation Method (Rodgers, 2000)*
- Inversion on 41 layers, with a single a priori profile and covariance matrix
- Data kept if: DOFS > 0.9;
  RMS of the spectral residual < 3×10^{-8} $W.m^{-2}.sr^{-1}.m^{-1}$;
  Cloud coverage < 25%
- See Hurtmans et al. (2012) for detailed information





- **AVK**: same shape for all layers, covering the entire altitude range from the troposphere to the stratosphere
- Around **1 level of information** in both equatorial and polar regions
- Maximum sensitivity in the **stratosphere** around 10-20 km altitude, depending on the latitude
- **DOFS**= from 0.9 (poles) to 1.2 (equator)
- **ERRORS**: vary from 0-10% at polar latitudes to 35-45% at the equator, mostly due to the interference with water vapor absorption lines

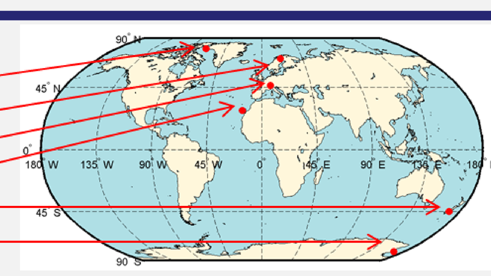
Total error on the total HNO_3 column (%)



Validation

Choice of 6 stations, all part of the NDACC network

Station	Latitude	Longitude
Thule	76.5° N	69° W
Kiruna	67.8° N	20.4° E
Jungfraujoch	46.6° N	8.0° E
Izaña	28.3° N	16.5° W
Lauder	45.0° S	170° E
Arrival Heights	77.8° S	167° E



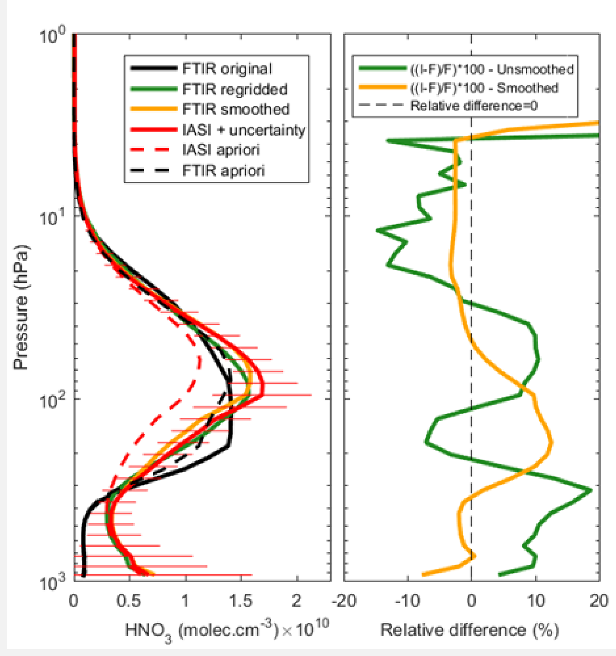
- Regridding and smoothing of the FTIR vertical profiles (bottom left figure) give good agreement with IASI profiles
- Comparison of FTIR and IASI time series (bottom right figure):
  - Retrieved columns (5-35 km) of both datasets are within the error range of one another
  - Differences between FTIR and IASI data (I-F)/F vary between -30 and 30%
  - Mean of differences (bias) = 4.0 %

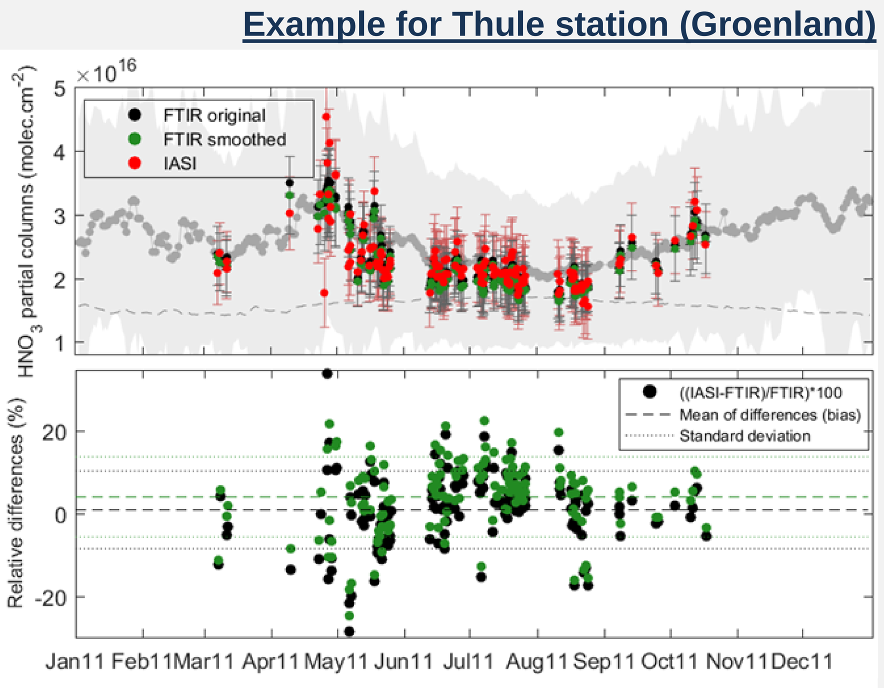
  ➔ IASI slightly overestimates concentrations compared with FTIR
  - Standard deviation = 9.7 %

  ➔ Bias > std ➔ differences not significant, compared to variability

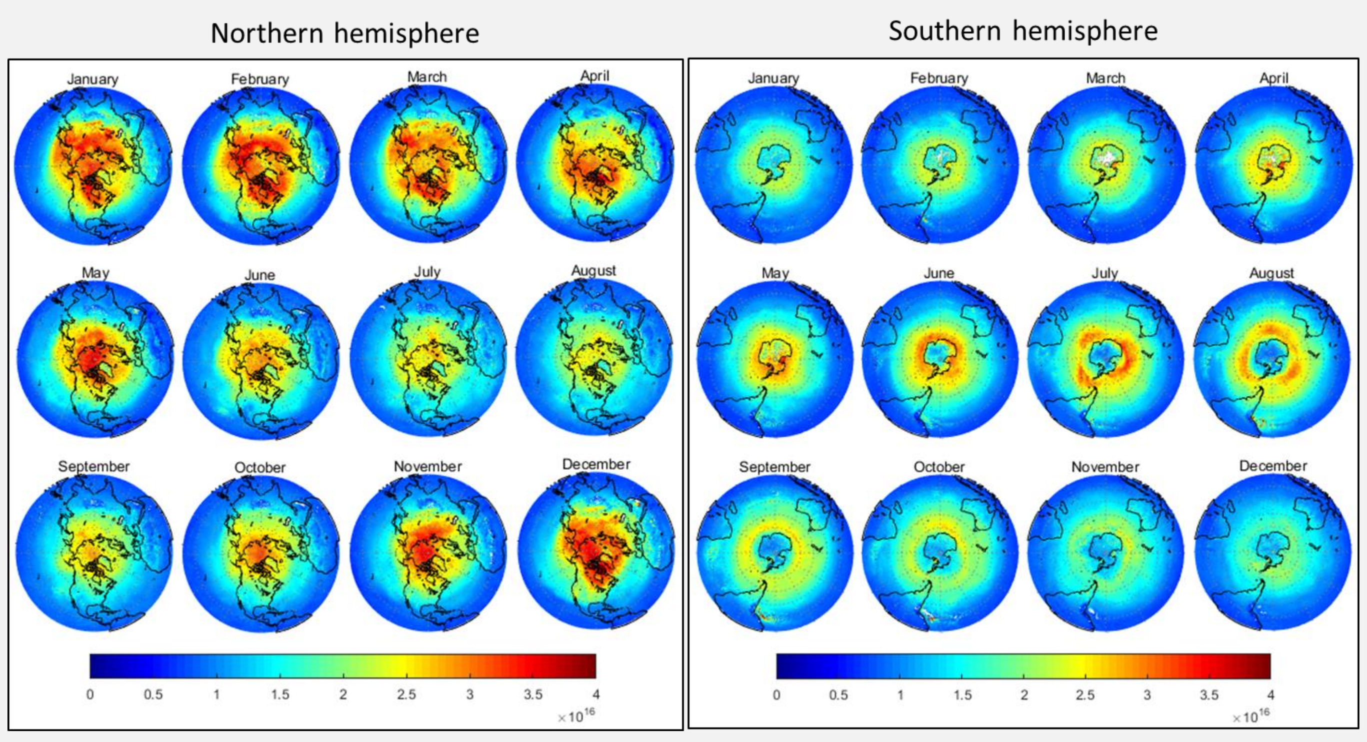
Stations	Bias (%) Smoothed FTIR	Standard deviation (%)	# pairs	R
Thule	4.0	9.7	151	0.84
Kiruna	8.6	11.9	206	0.81
Jungfraujoch	13.9	9.6	583	0.91
Izaña	9.2	9.8	256	0.74
Lauder	16.3	11.9	80	0.81
Arrival Heights	-0.8	15.9	75	0.78
All stations	10.5	11.5	1351	0.93

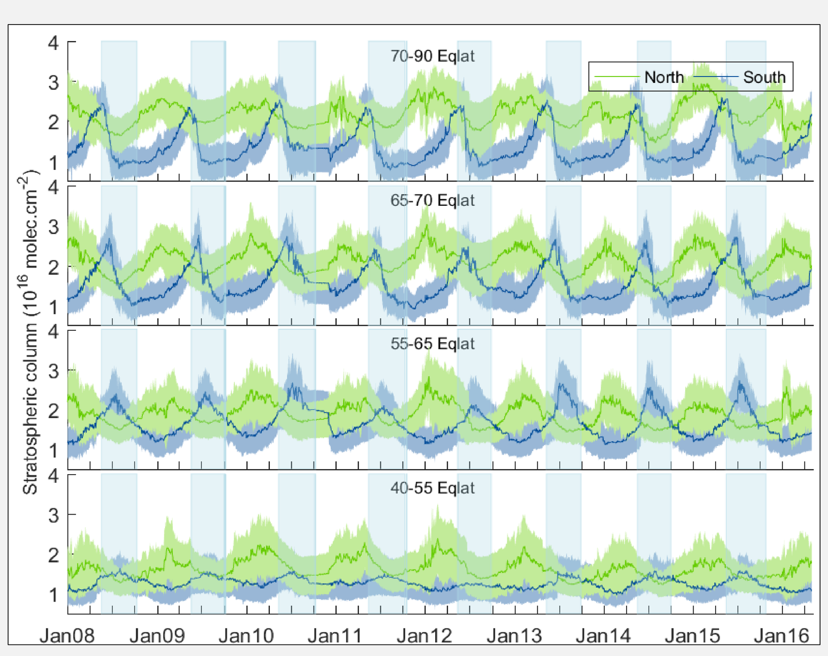
Example for Thule station (Groenland)





Spatial & temporal distributions





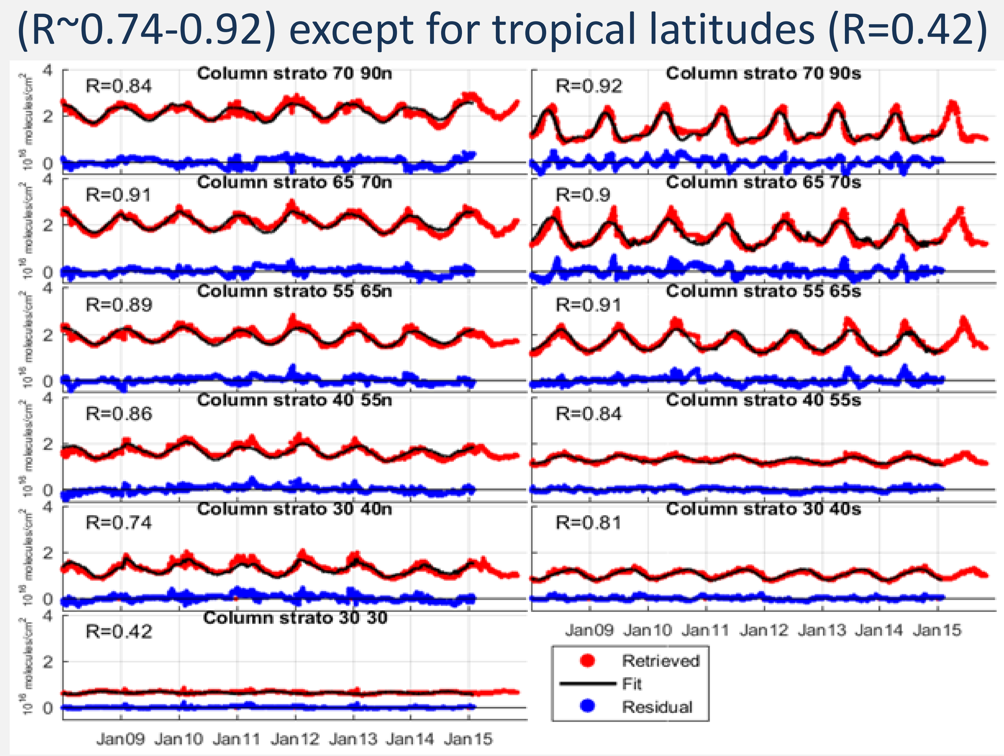
Annual cycles in stratospheric HNO_3 distributions

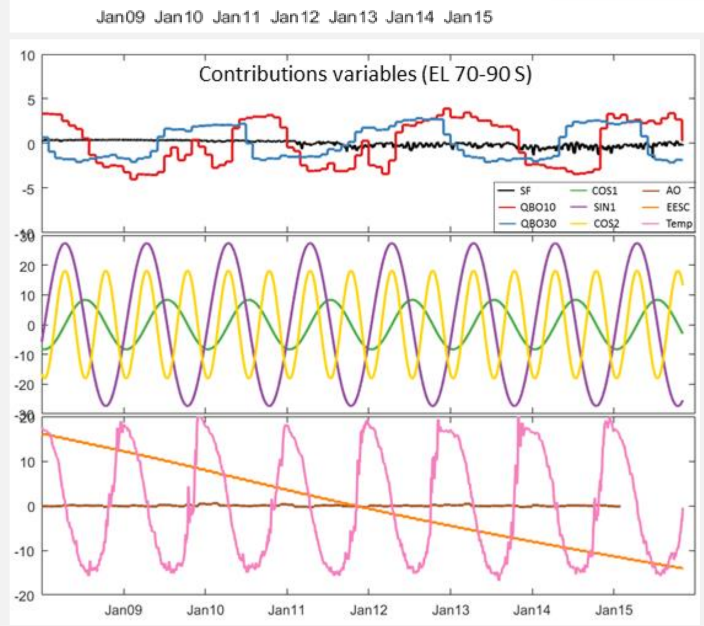
- NH: build-up of columns during winter
- SH: small build-up, then denitrification

In **equivalent latitudes**: drop in stratospheric columns when polar stratospheric T°s (taken at 50 hPa) reach 195 K (PSCs formation threshold)

Multivariate regressions

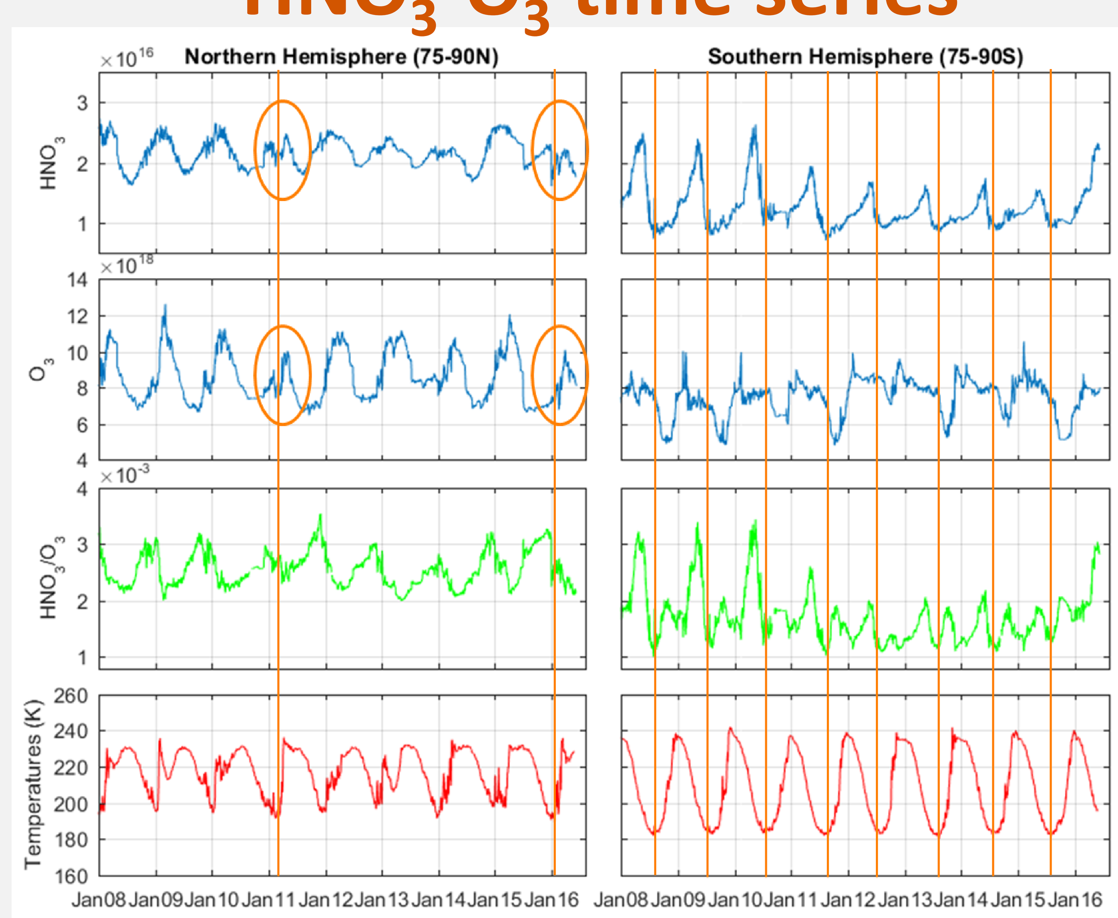
Good correlation between fit & observations (R~0.74-0.92) except for tropical latitudes (R=0.42)





- Dominance of the annual cycle (B-D circ.) in the regression
- Large influence also of stratospheric T°

HNO_3-O_3 time series



Southern hemisphere:

- systematic drop of HNO_3 columns when stratospheric T°s reach the PSCs formation threshold
  → lowest columns when lowest T°
- at the lowest HNO_3 stratospheric column: start of decrease in O_3 stratospheric column (delay of a few weeks)

Northern hemisphere:

- no such systematic pattern
- HNO_3 and O_3 depletion occur in the winters 2010-11 and 2015-16 (lower T° than usual)

Conclusions & ongoing works

- The IASI instrument shows a good sensitivity to the HNO_3 vertical profile, with its maximum in the stratosphere
- The validation with the FTIR profiles suggests good performances of IASI overall, with a mean bias of 10.5%
- The seasonal variations observed are relevant and are in agreement with chemical and dynamical processes in the atmosphere, and more specifically the stratosphere
- Multivariate regressions allow fitting the HNO_3 time evolution; some unexplained residuals deserve more attention
- The co-located IASI measurements allow for a HNO_3/O_3 co-analysis

➢ Improvement of the regression model to reduce the residuals
➢ Implementation of a chemistry-climate model to apprehend climate variables and their effects on [HNO_3]
➢ Focus on the polar regions and their specific dynamics and chemistry

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