A novel technique for studying volcanic gas chemistry and dispersion on short time scales

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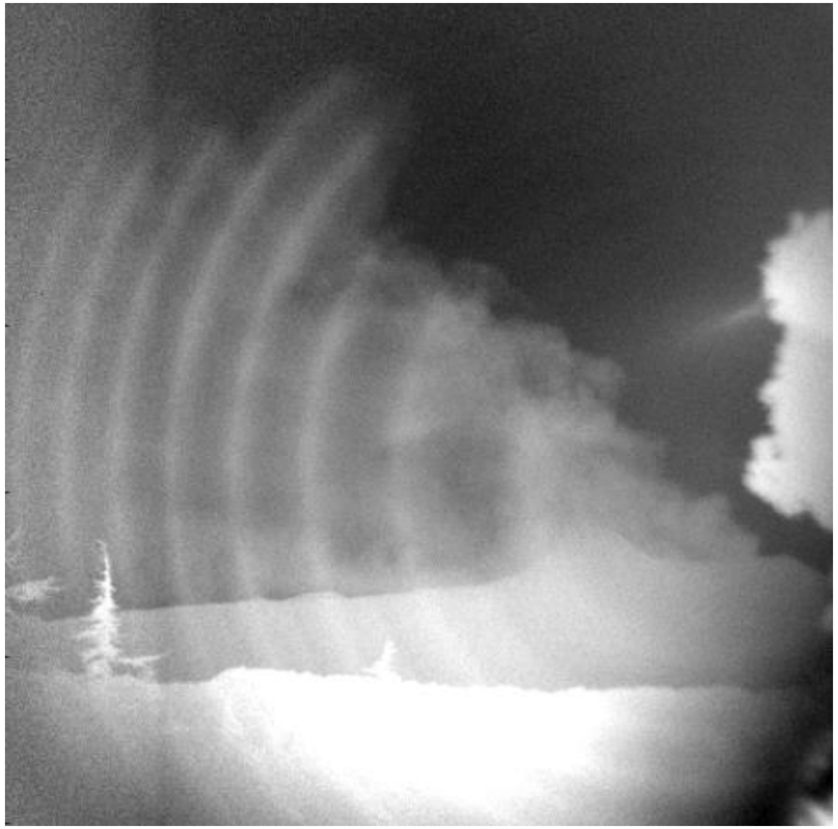

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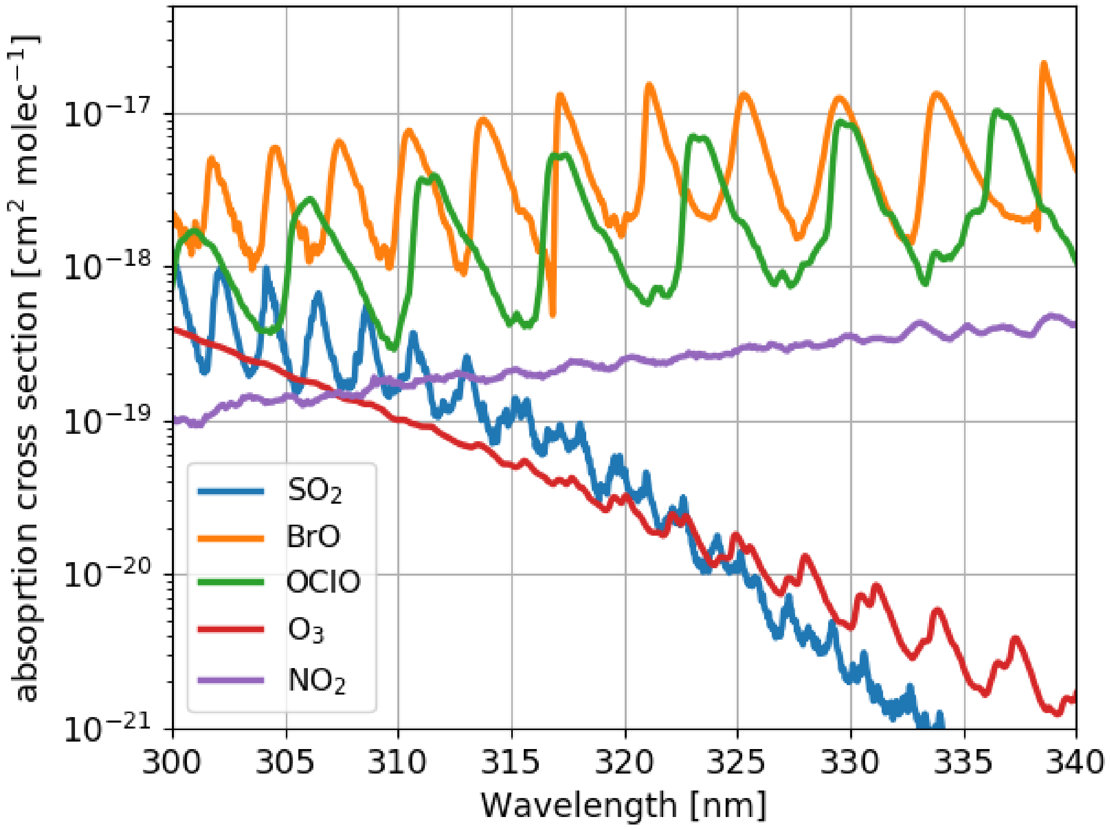
Imaging of volcanic plume emissions

Motivation: resolve plume processes at their intrinsic time and spatial scale, e.g.:

- emission fluxes of trace gases
- investigation of halogen chemistry (e.g. bromine)

Requirements:

- identification by **characteristic differential absorption structures** within a measured spectral optical density
- quantification of **column densities** (CDs, number density of trace gas along atmospheric light path)
- **‘sub nm‘ spectral resolution** required for sufficient selectivity and sensitivity



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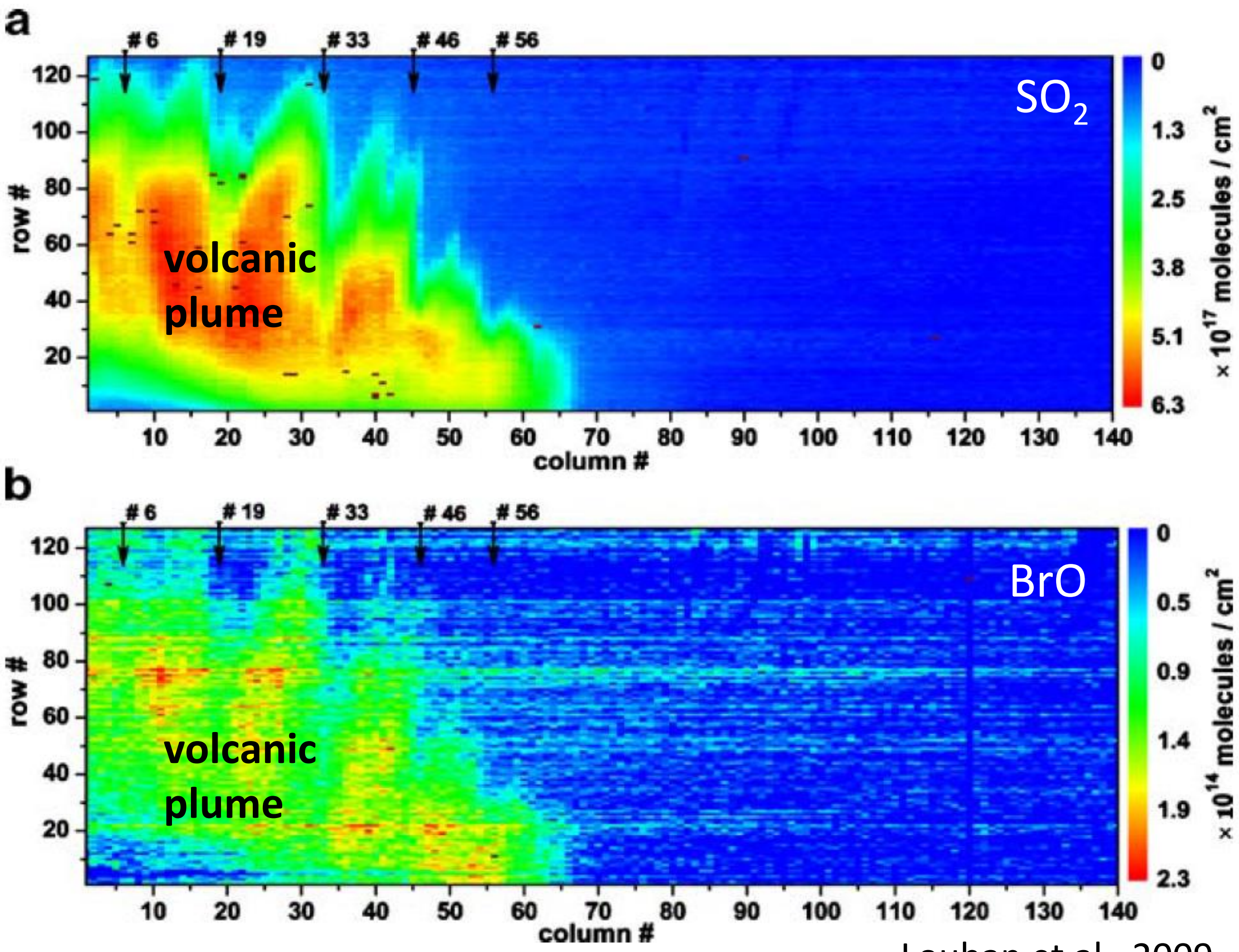
Imaging trace gases in volcanic emissions: Imaging DOAS

Imaging of volcanic SO_2 and BrO emissions (pushbroom approach, i.e. one column at a time)



Louban et al., 2009

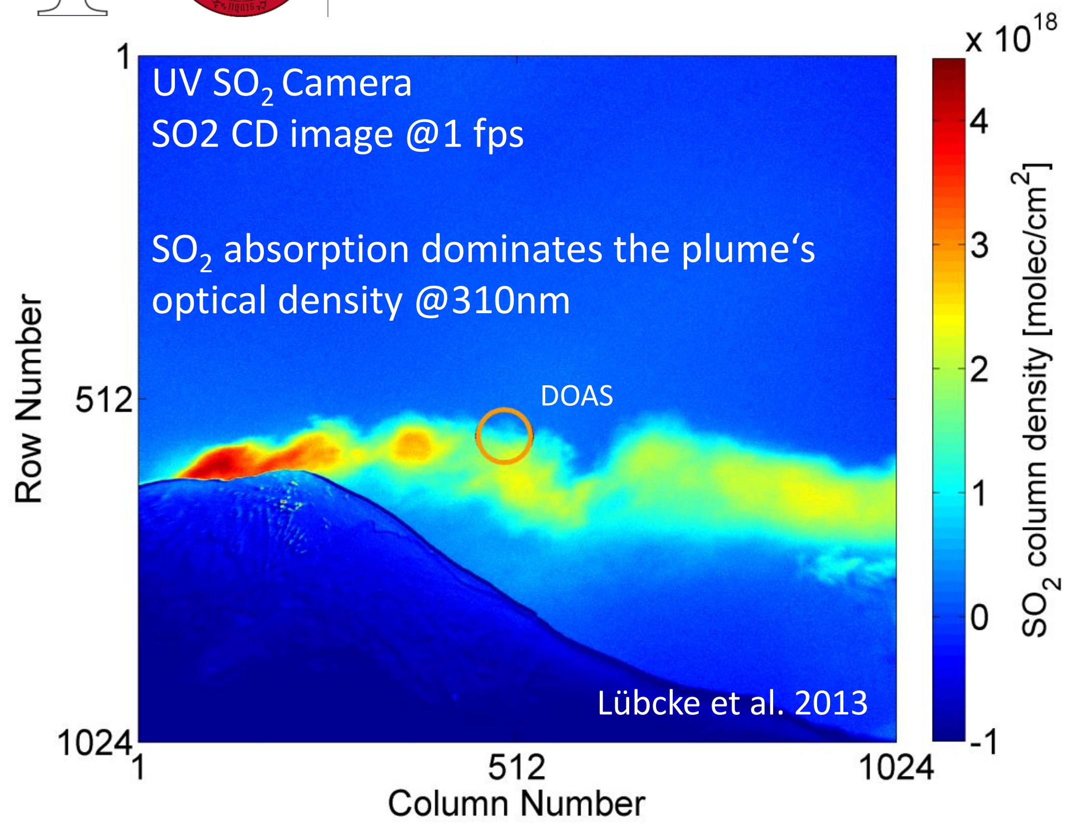
- **High spectral resolution** → detection of various different trace gas species simultaneously

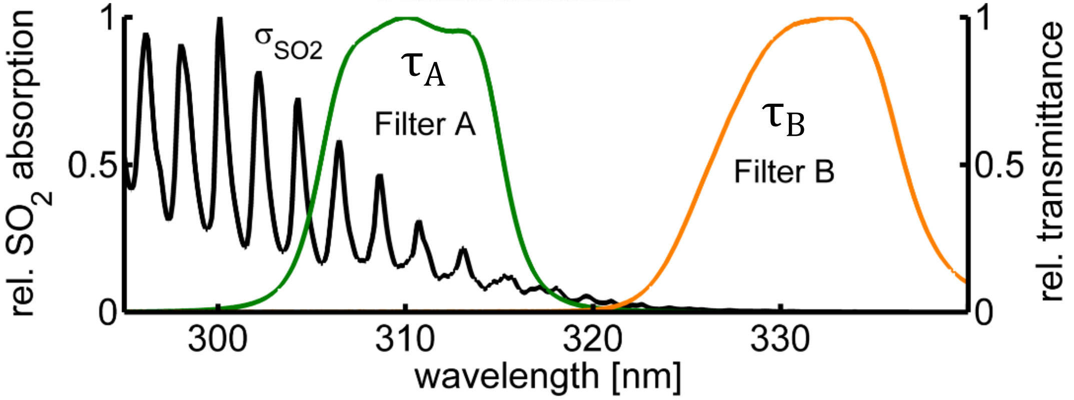
BUT: too slow!

imaging by spatial scanning:

- limited light throughput of the spectrometer
- scanning mechanisms or complex optics required

Imaging trace gases in volcanic emissions: SO_2 Camera





high spatio-temporal resolution → flux monitoring

BUT: limited selectivity & high cross interferences

→ interference to clouds, aerosol and O_3

→ Only works for high SO_2 column densities

Lambert-Beer's law:

$$dI(\lambda) = I_0(\lambda) \cdot e^{-\sigma(\lambda) \cdot S} \; d\lambda = I_0(\lambda) \cdot e^{-\tau} \; d\lambda$$

I_0: incident radiation
σ: abs. cross section
S: column density
τ: optical density

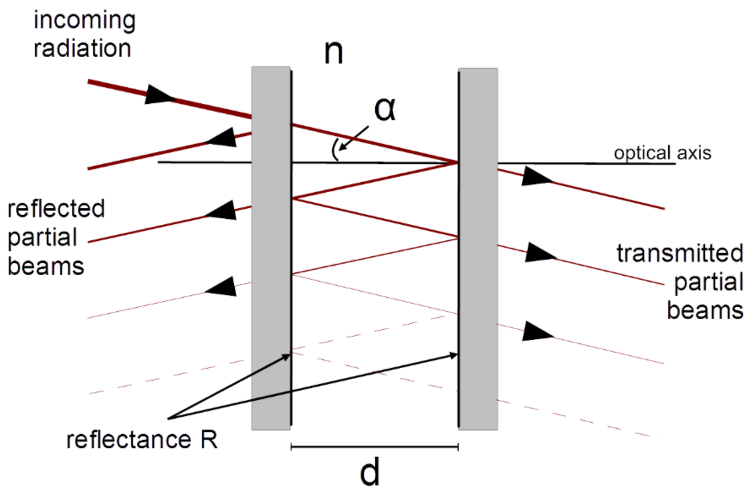
$$\tau_i = \ln\left(\frac{\int_{\lambda_{1,i}}^{\lambda_{2,i}} I_{0,i}(\lambda)\, d\lambda}{\int_{\lambda_{1,i}}^{\lambda_{2,i}} I_i(\lambda)\, d\lambda}\right) = \sigma_i \cdot S$$

$i \in$ A,B (Filter)

Apparent absorbance SO_2 Camera:

$$AA = \tau_A - \tau_B = k_{SO_2} \cdot S_{SO_2}$$

Imaging Fabry-Perot Interferometer Correlation Spectroscopy (IFPICS)

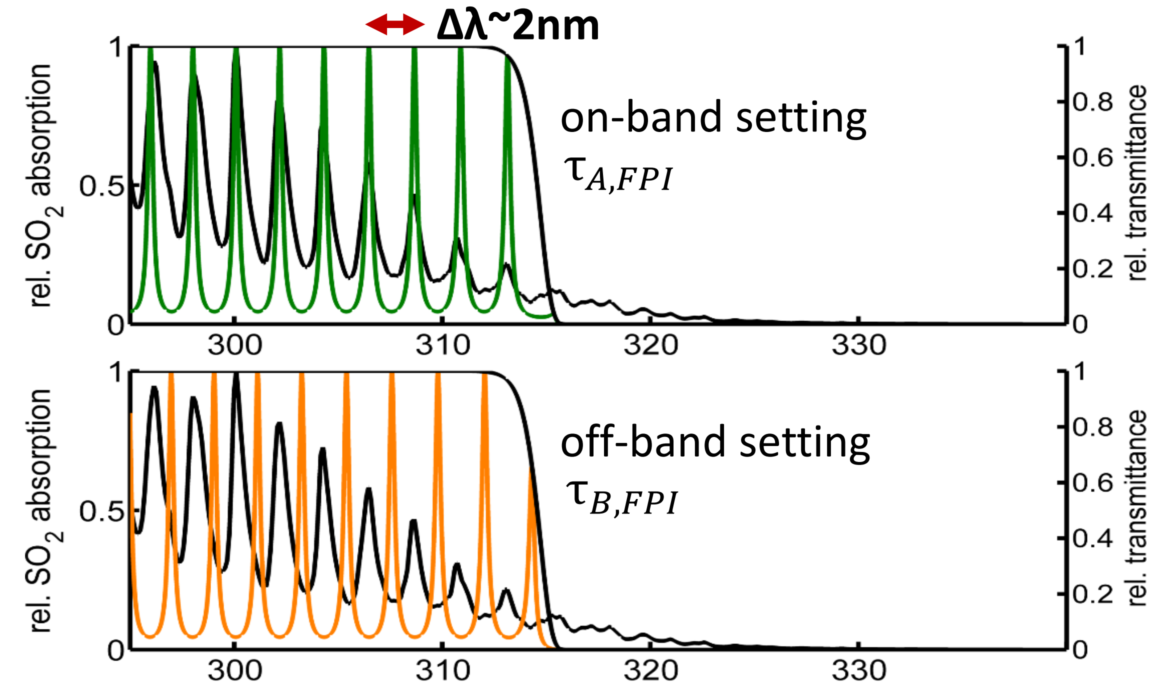


free spectral range: $\Delta\lambda \approx \frac{\lambda^2}{2 \cdot nd\cos(\alpha)}$

Apparent absorbance FPI camera:

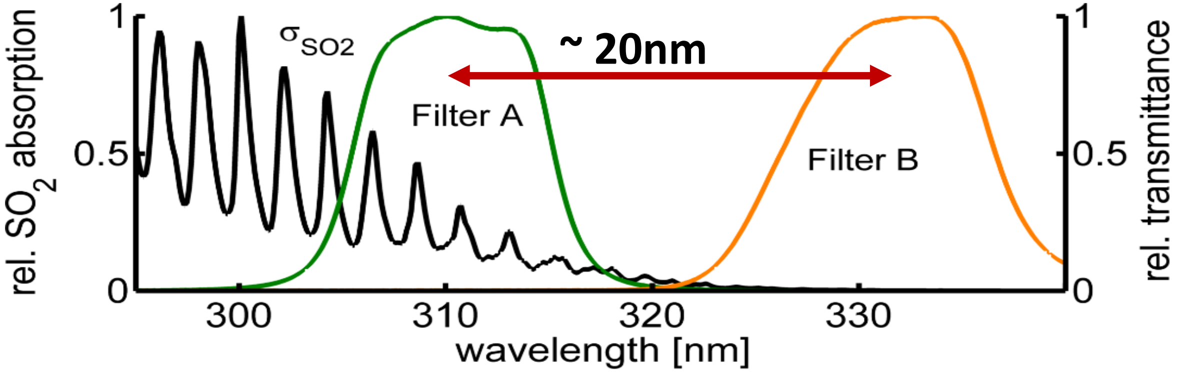
$AA = \tau_{A,FPI} - \tau_{B,FPI} = k\,S_{SO2}$

- much **more specific** to the trace gas (narrow band structure)
- broad band **interferences reduced** (spectral variation 2nm vs. 20nm)



transmission at **maximum** SO_2 absorption
→ **correlation**

transmission at **minimum** SO_2 absorption
→ **anti-correlation**



Filter based SO_2 Camera

Comments: The FPI transmission spectrum is matched to the SO_2 absorption spectrum in an on-band and off-band setting replacing the spectral filters used by SO_2 cameras. The spectral separation between the two spectral channels is thereby reduced by a factor of 10 (2 nm instead of 20 nm), thus reducing broad band interferences, e.g. to aerosols and increasing the specificity to the investigated trace gas. Tuning between on- and off-band setting is applied by tilting the FPI optical axis = variation of incidence angle α.

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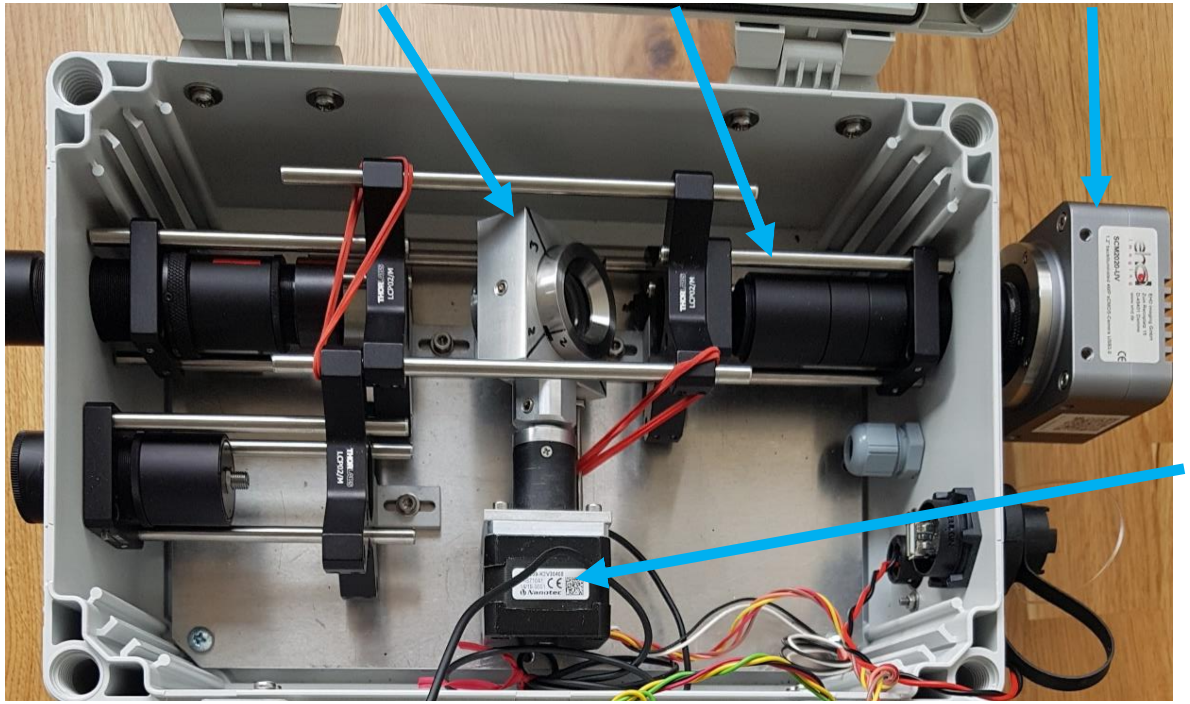
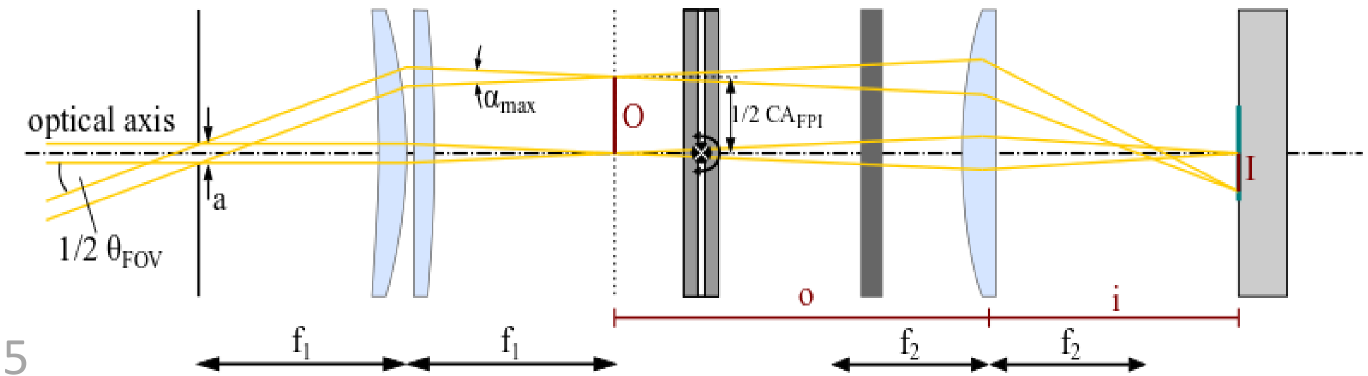
Proof of concept study for SO_2 – Imaging Camera





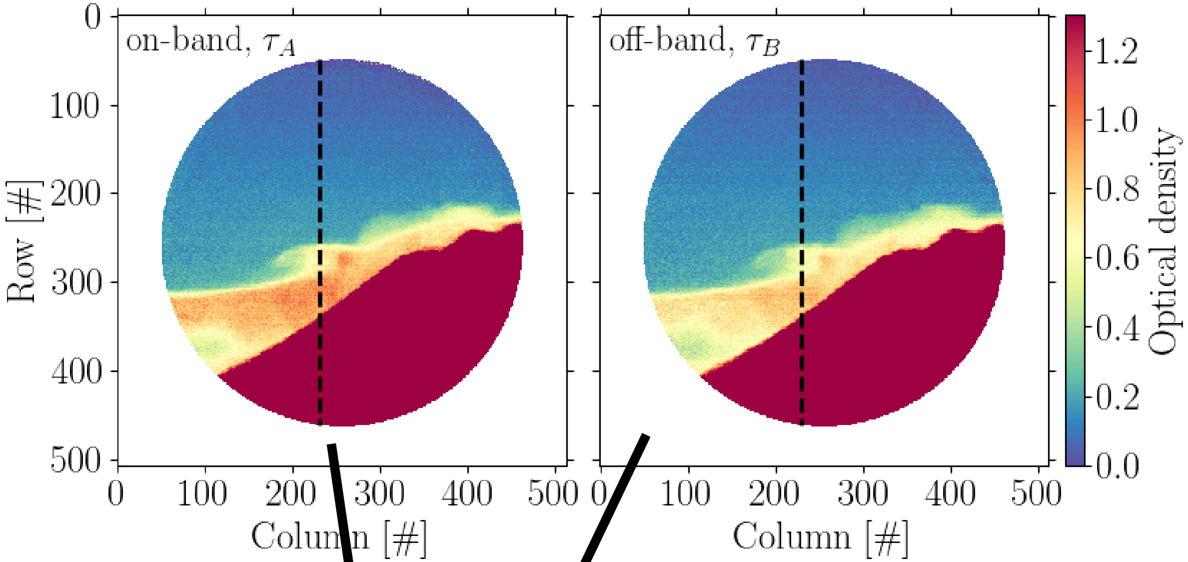
Dimensions [mm]:
200 × 350 × 130

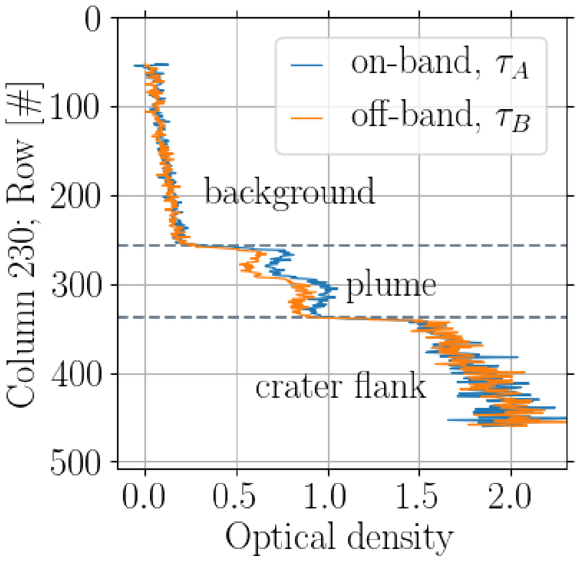
Weight: 4.8 kg

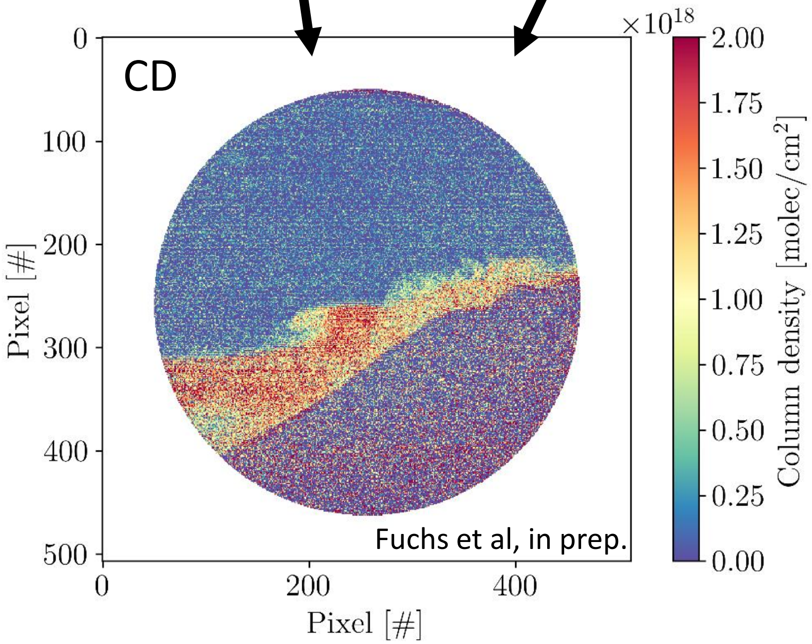
Power consumption: < 10W

Comments:
The IFPICS instrument uses an image-side telecentric optic setup (see schematic bottom left).
Light enters from the left. Light is traversing an aperture before it gets parallelised by lens 1. The aperture entrance is thereby limiting the angle of divergence α_{max}. The FPI is mounted on a motor that allows to tilt it around its optical axis to enable tuning the spectrum by varying the incidence angle onto the FPI. After traversing an band pass filter the light is imaged onto a 2D UV-sensitive detector.

Imaging of SO_2: plume of SE-crater, Mt. Etna, Sicily







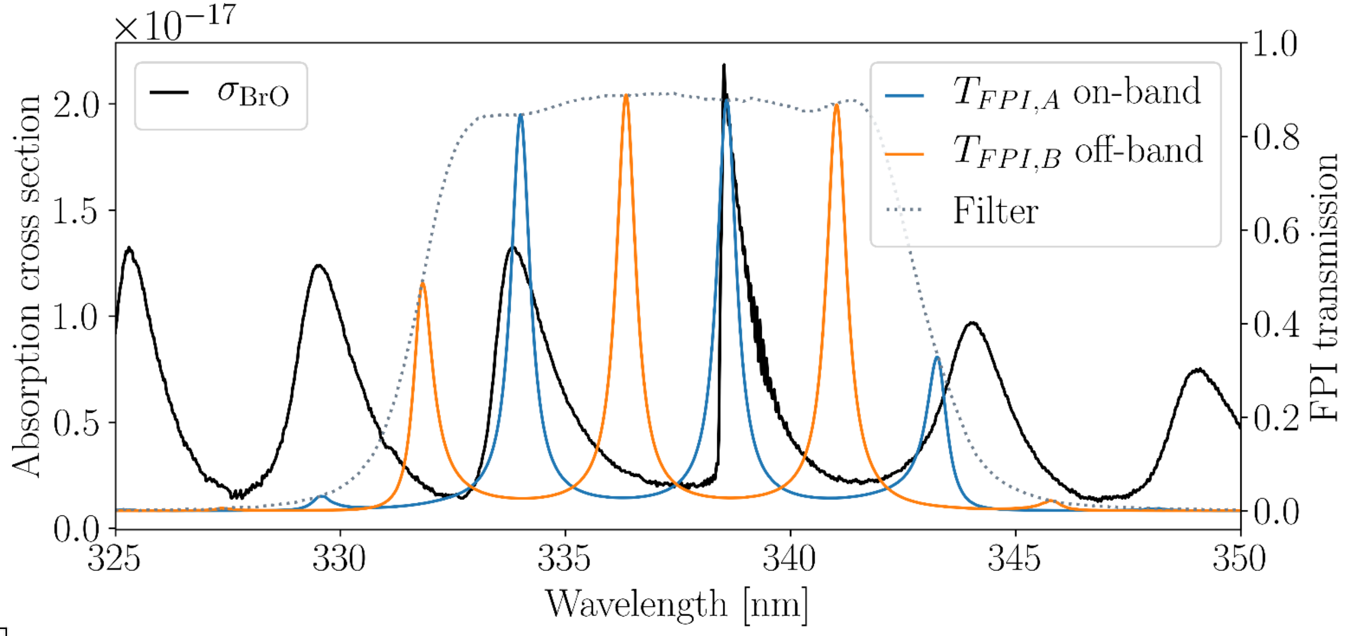
- Calibration via forward modelling the instrument response
- SO_2 sensitivity: *AA* = 10^{-19} cm^2/molec · *S*
- Detect. limit ~ 4 × 10^{17} molec cm^{-2} $s^{-1/2}$

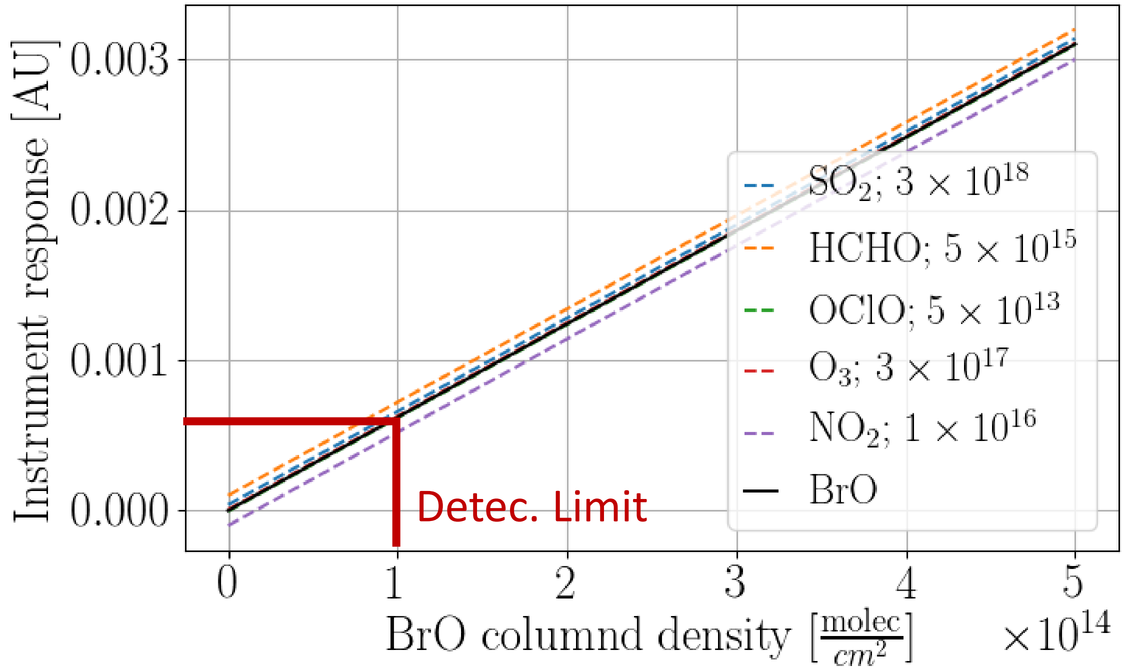
Comments:
Optical density images shown in on-band τA and off-band τB setting (top left images). Plot of the optical densities along vertical dashed lines shows that the signal differ only in the plume region (τA > τB), whereas the signal is equal in the background an crater flank area (τA = τB) (top middel). Apparent absorbance AA given by AA = τA – τB is converted into SO_2 column densities (CD) (bottom left) via an instrument forward model, which was validated by gas cell measurements.

IFPICS remote sensing of volcanic BrO – A model study

Simulated FPI transmission spectrum for atmospheric conditions (right)

Correlating and anti-correlating FPI transmission spectrum





Cross interferences of BrO measurements to other trace gas species (left)

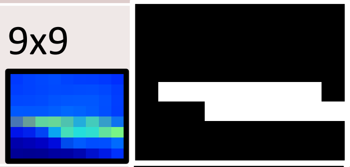
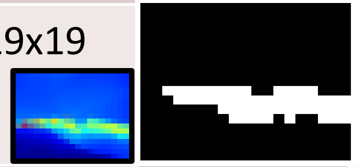
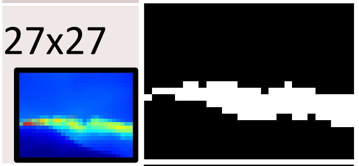
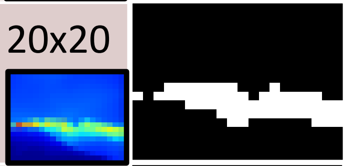
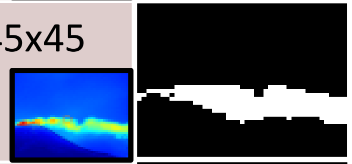
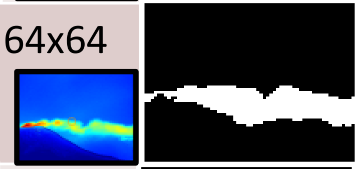
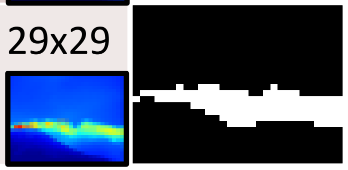
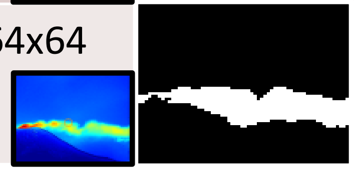
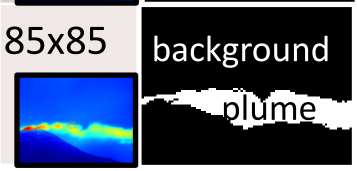
Sensitivity: $AA = 6.5 \times 10^{-18}$ cm²/molec · S

Intended detection limit for atmospheric Measurement conditions: ~ 10^{14} molec/cm²
(~ 10^{13} molec/cm² reached in laboratory)

→ Low cross interferences!

IFPICS remote sensing of volcanic BrO – A model study

- Lower signal expected: apparent absorbance ~ 10^{-3} (for SO2 ~ 10^{-1})
- Required detection limit ~10^{14} molec/cm² → achieved with an one pixel prototype under laboratory measurement conditions
- Model study:
  Low photon budget - lower signal →
  trade spatial resolution against photons
- BrO detection limits for different degrees of binning and different acquisition times (right)

BrO detect. limit [molec/ cm^{-2}]	pixel resolution		
	acquisition time 1s	acquisition time 5s	acquisition time 10s
0.5×10^{14}	9x9 	19x19 	27x27 
10^{14}	20x20 	45x45 	64x64 
1.5×10^{14}	29x29 	64x64 	85x85 

Calculations according to Kuhn et. al. 2019

Conclusion:

SO_2: First successful FPI imaging application on volcanic SO2

- Prototype is robust and small
- Good sensitivity and low cross interferences
- Inherent calibration with forward model
- No background reference images required

BrO:

- BrO model study yield promising spatio-temporal resolution
- Detection limit of ~10^{14} molec/cm² easily achieved with one-pixel prototype in laboratory

Outlook:

- Field measurements of volcanic plume BrO
- Extend the technique to further trace gases
- Field measurements of weaker SO2 point sources.

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