Radiocarbon field measurements at a nuclear facility with cavity ring-down spectroscopy

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Contributors & Funding

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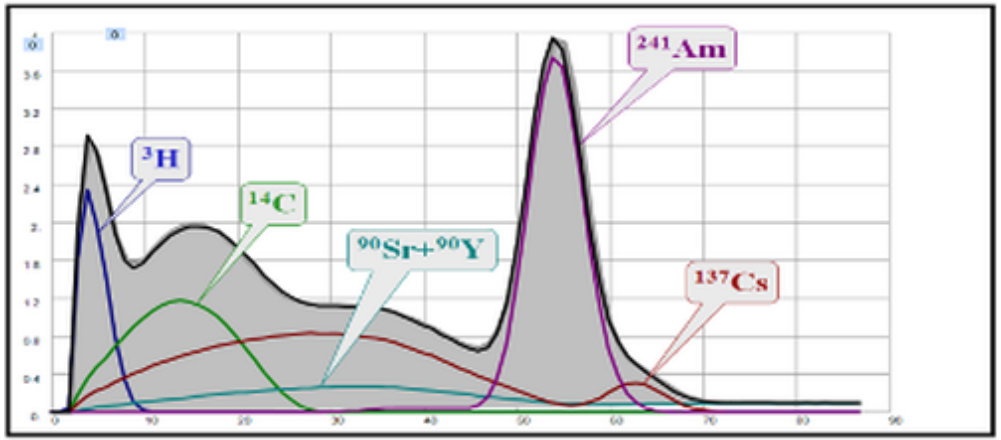
The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

Characterization of Conditioned Nuclear Waste for its Safe Disposal in Europe

Liquid scintillation counting

- require large amount of sample
- complex sample preparation
- Sensitive to all molecular forms
- produces additional waste

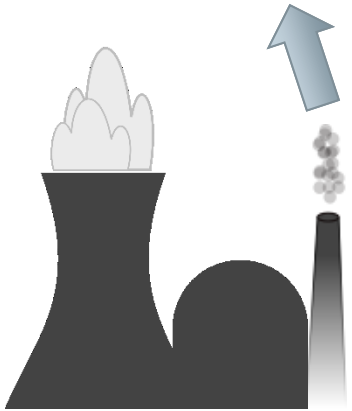
Overlapping peaks in the LSC spectrum



Scintillation peaks of commons radionuclides

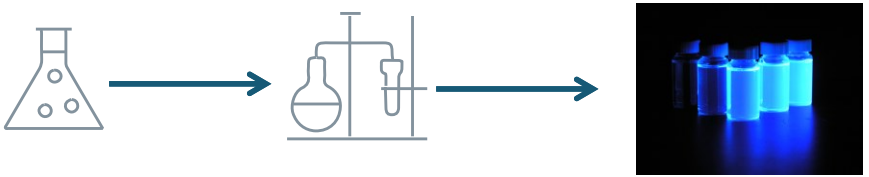
C-14 NPP stack emissions

C-14 emissions
200–400 Bq/m^3



Monitoring required by regulators

~ 1 week collection time

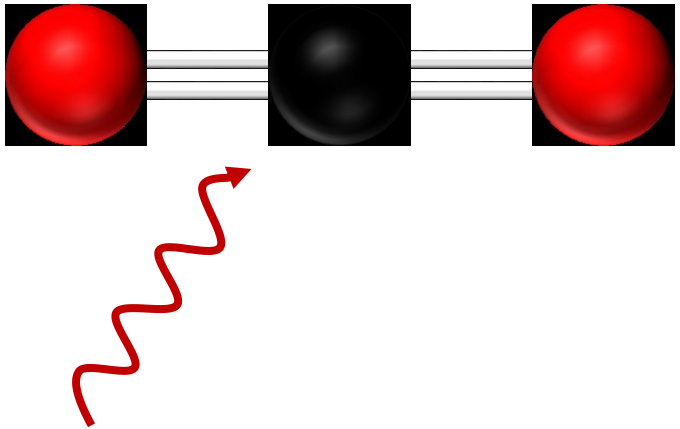


up to several days laboratory work

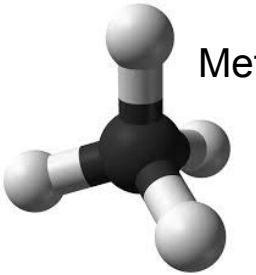
1 data point for total C-14

Laser spectroscopy for radionuclides detection

Molecular vibration modes

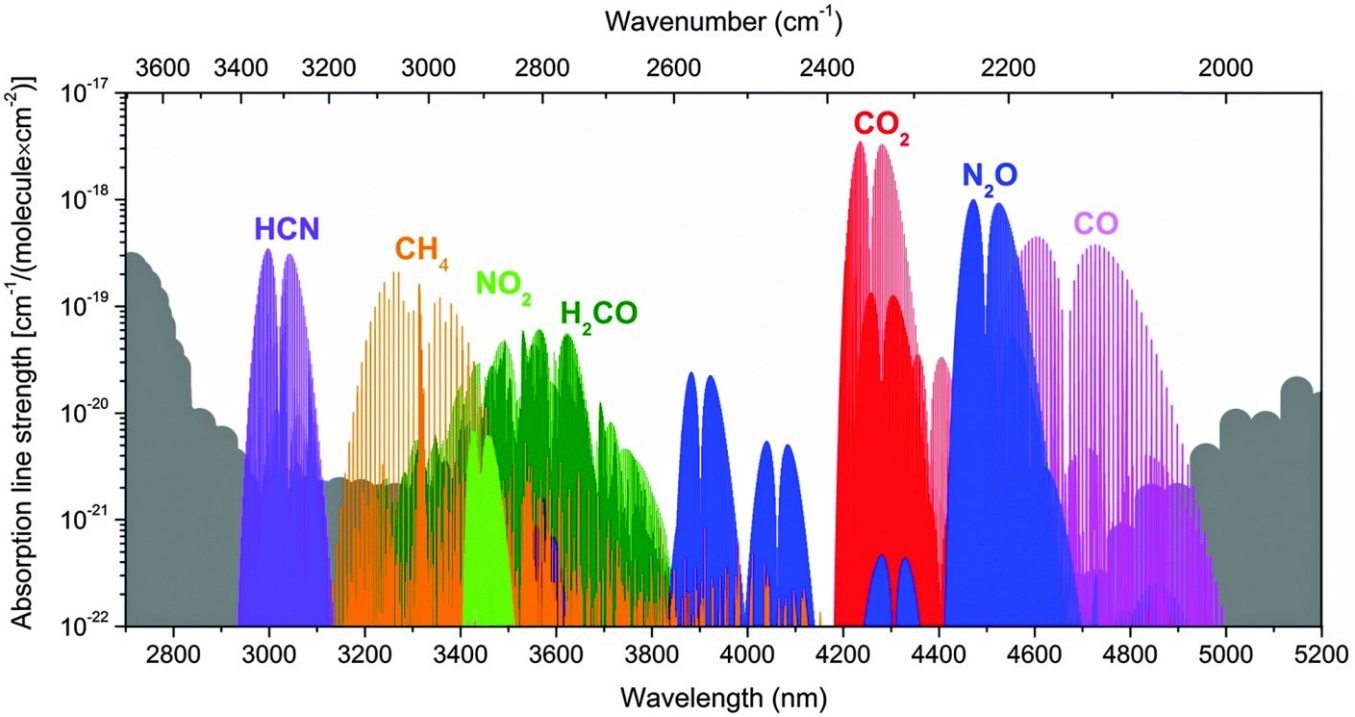


Water



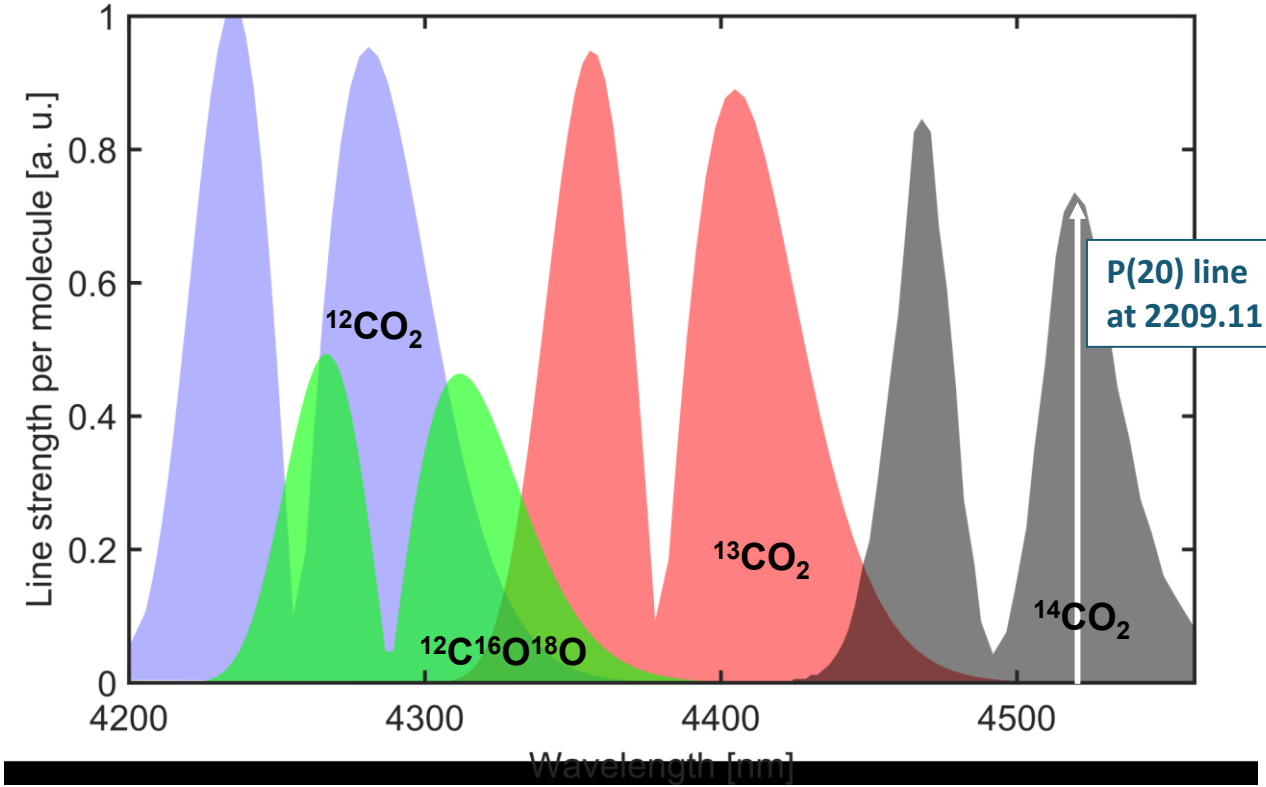
Methane

Mid-infrared spectroscopy for trace gas detection

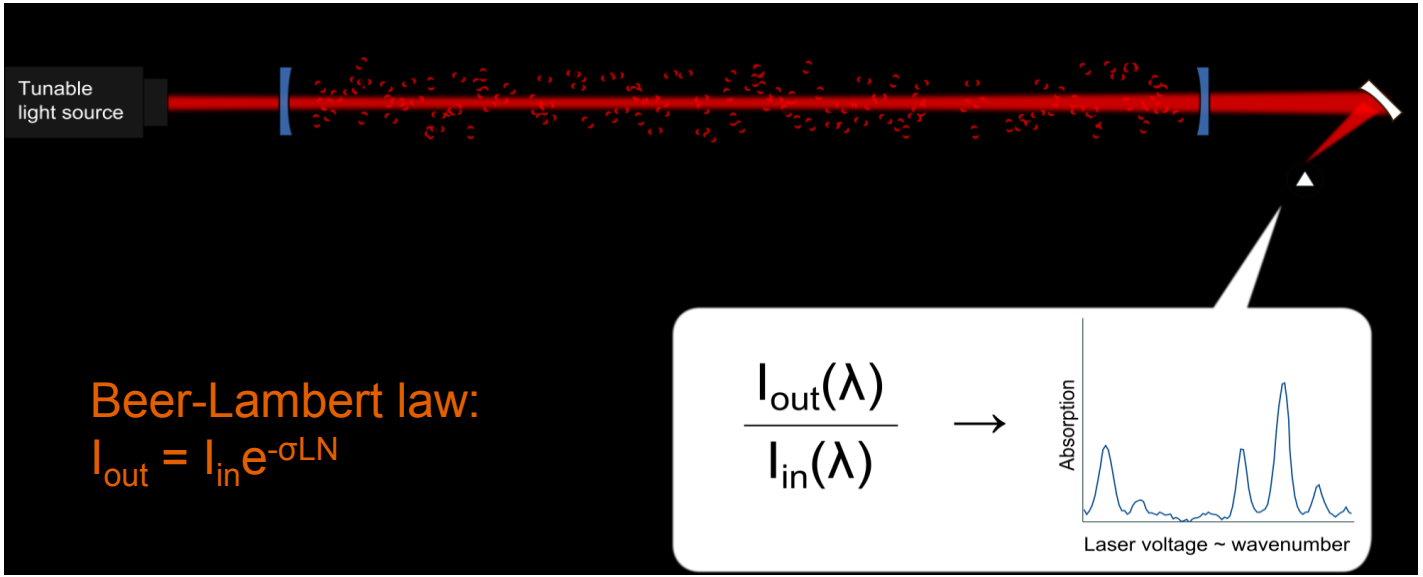


From *M. Vainio and L. Halonen, Physical chemistry Chemical Physics (PCCP)* ***18,*** *4266 (2016)*

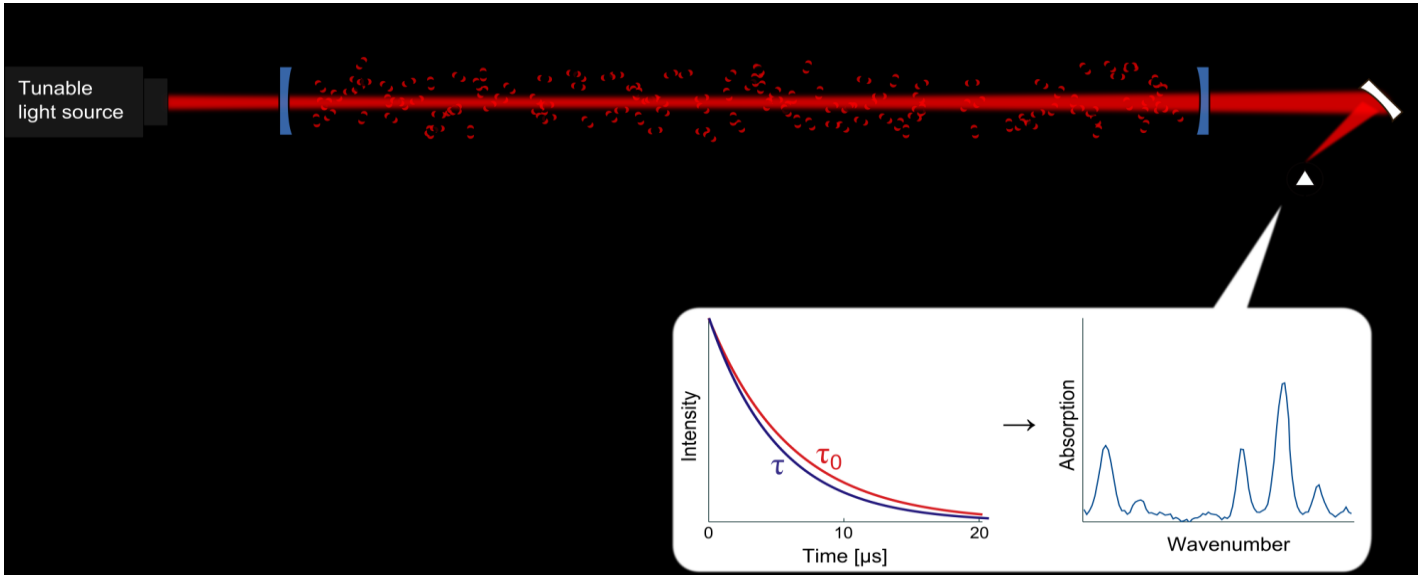
Absorption spectrum of CO_2 isotopes



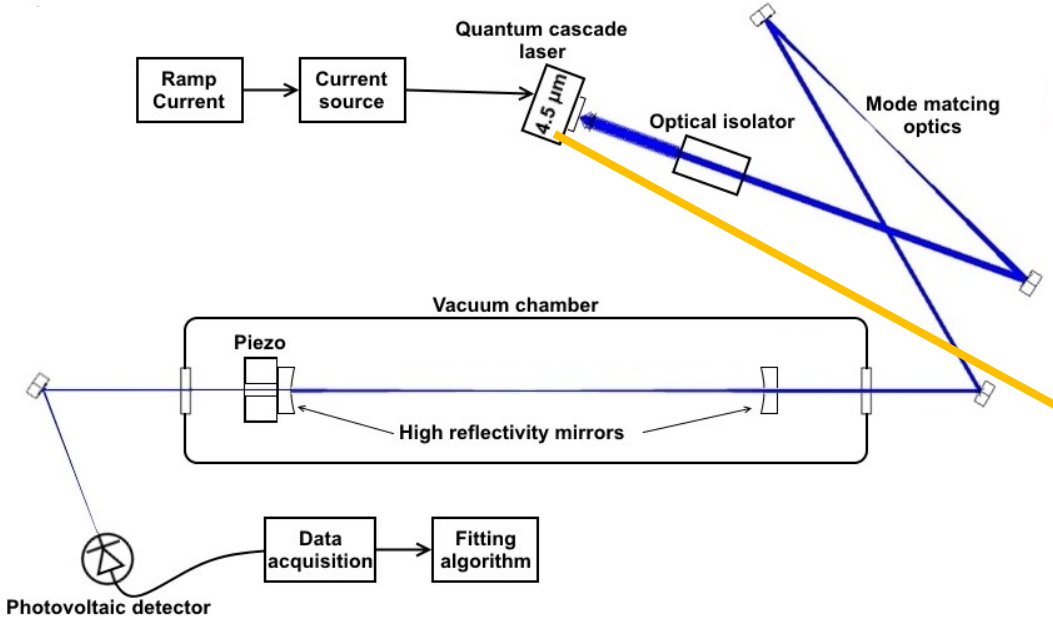
Basics of laser spectroscopy



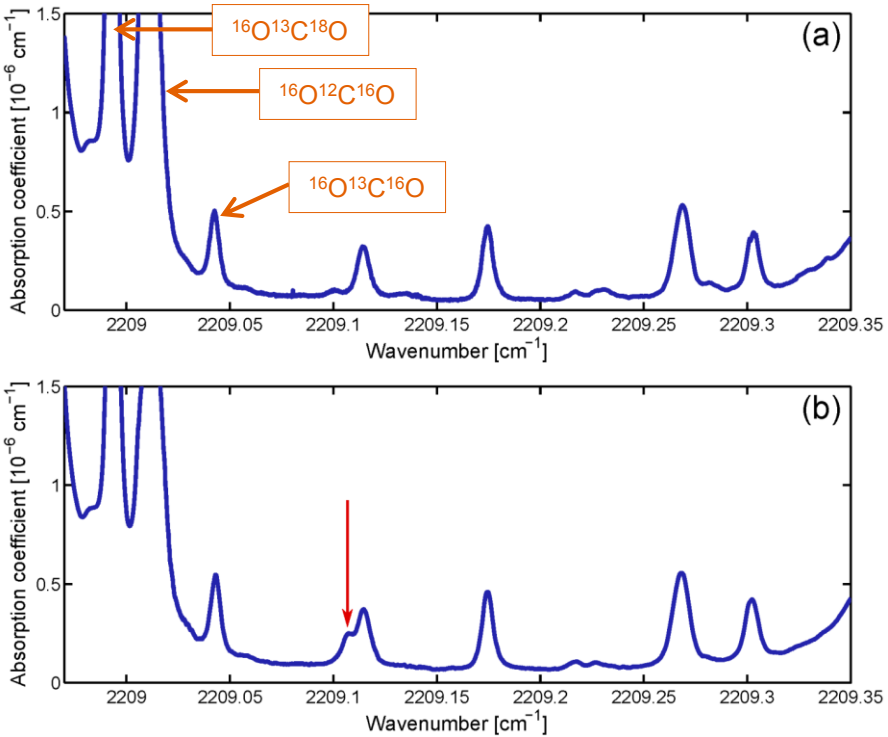
Cavity ring-down spectroscopy



Compact instrumentation



CO_2 vs $^{14}CO_2$ spectra



$^{14}C/C$=1 ppb

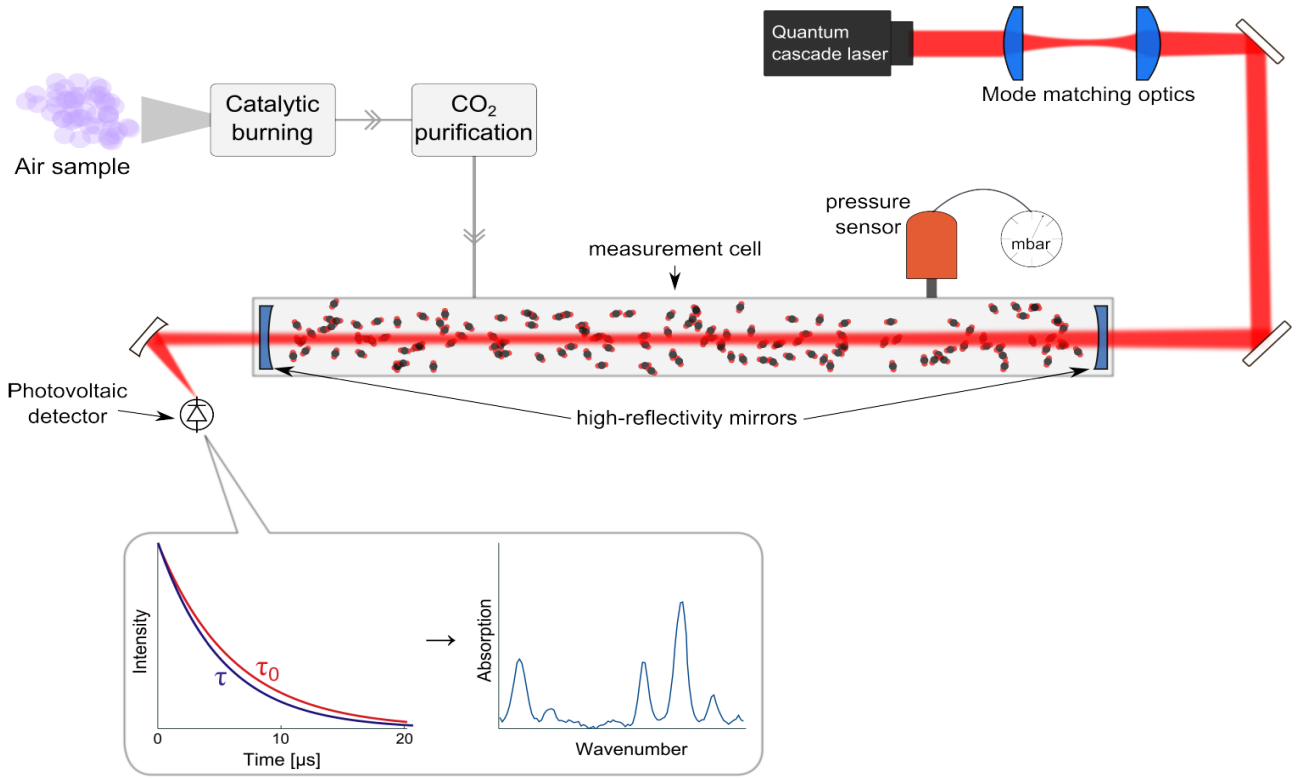
- $^{14}C/C$= 50 ppt → 5 kBq/m^3 in air
- 400 ppm of CO_2 in air

→ 2 Bq/m^3 after extraction of CO_2 from air

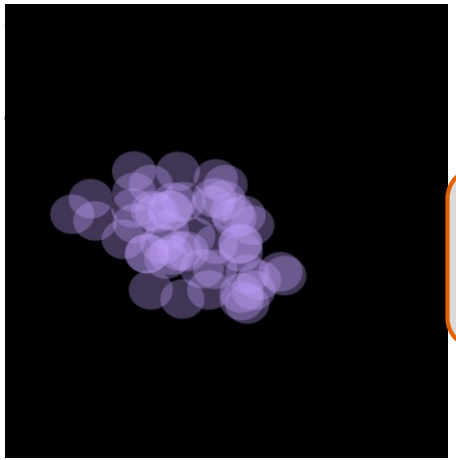
Radiocarbon Dioxide detection based on Cavity Ring-Down Spectroscopy and a Quantum Cascade Laser
*G. Genoud et al., Opt. Lett. **40**, 1342 (2015)*

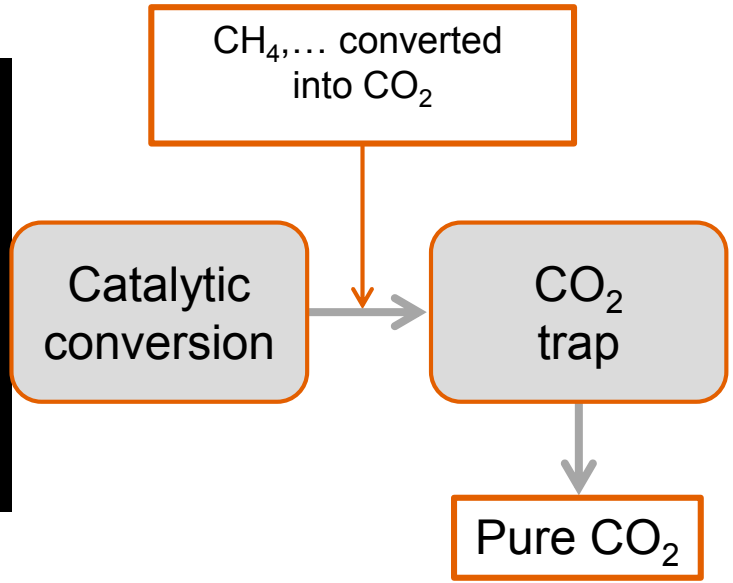
Instrumentation for field measurements

C-14 detection using cavity ring-down spectroscopy

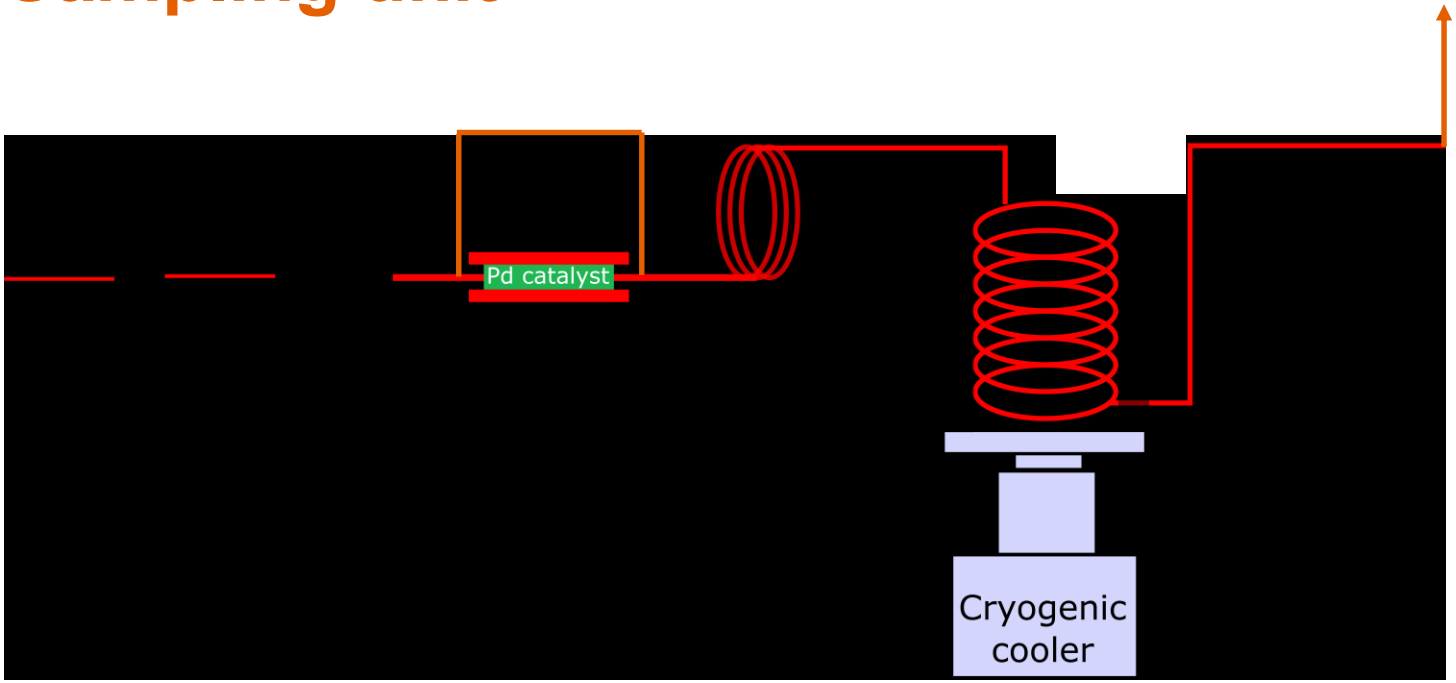


C-14 analysis from atmospheric samples

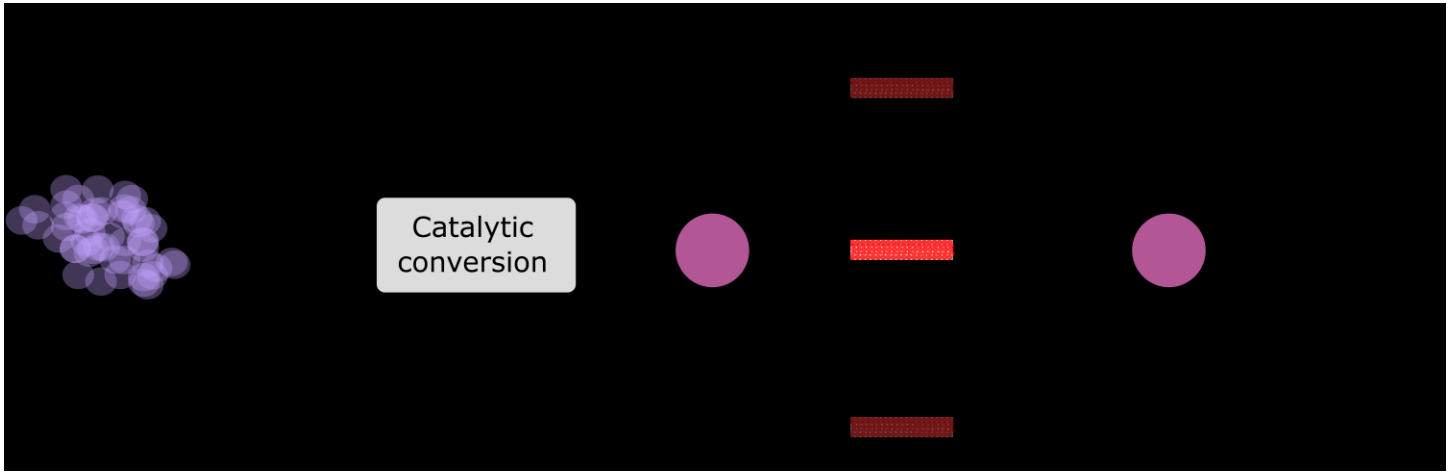




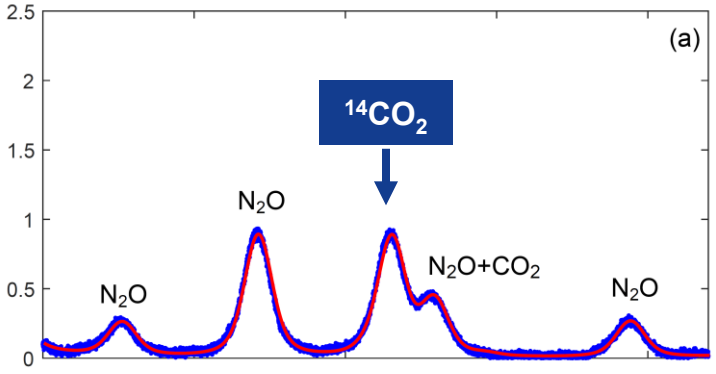
Sampling unit



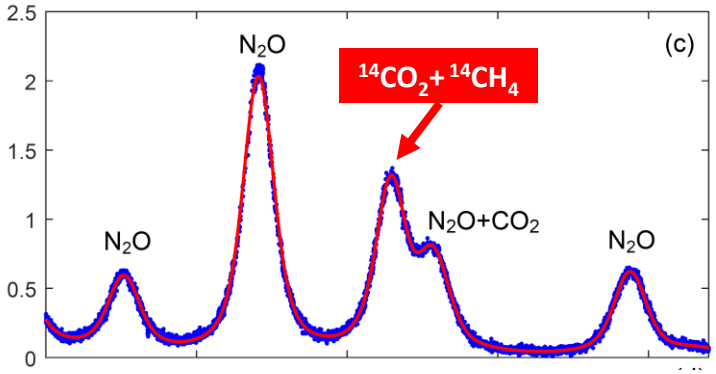
Adsorbent for CO_2 trapping



C-14 in an air matrix



No catalytic conversion 12 mbar

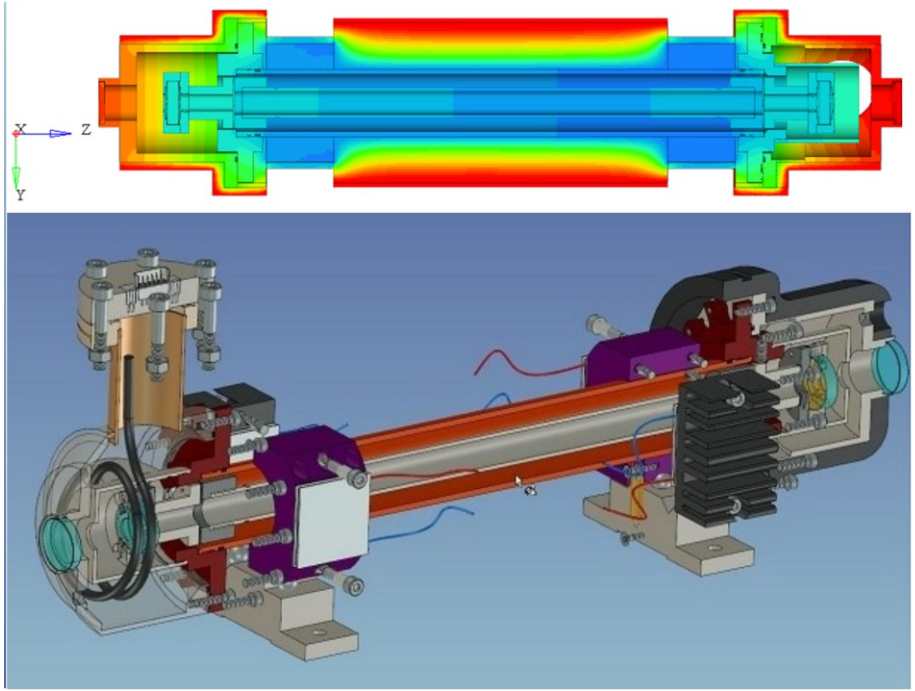


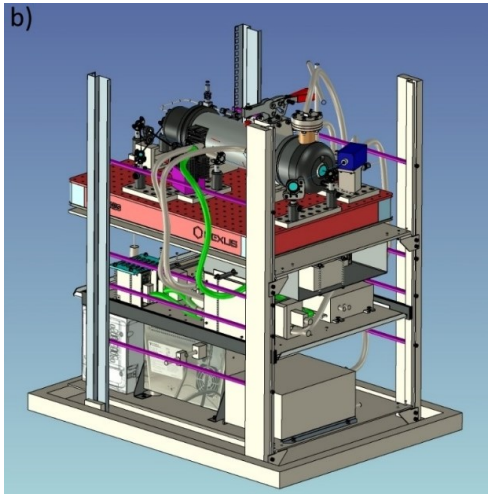
With catalytic conversion 18 mbar

Standardized samples:
- 339 Bq/m^3 of $^{14}CO_2$
- 208 Bq/m^3 of $^{14}CH_4$

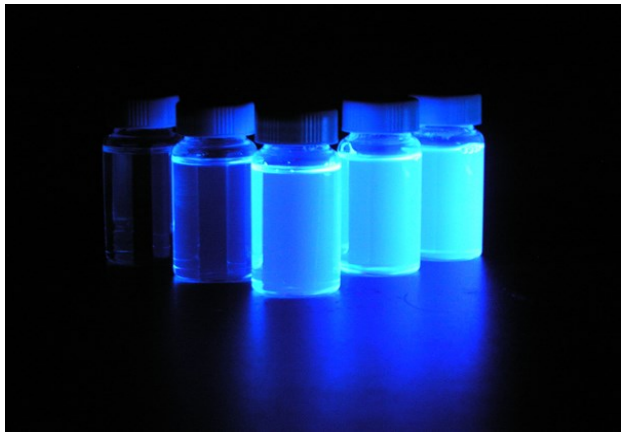
Laser Spectroscopy for Monitoring of Radiocarbon in Atmospheric Samples
G. Genoud *et al.*, *Analytical Chemistry* **2019** *91* (19), 12315-12320

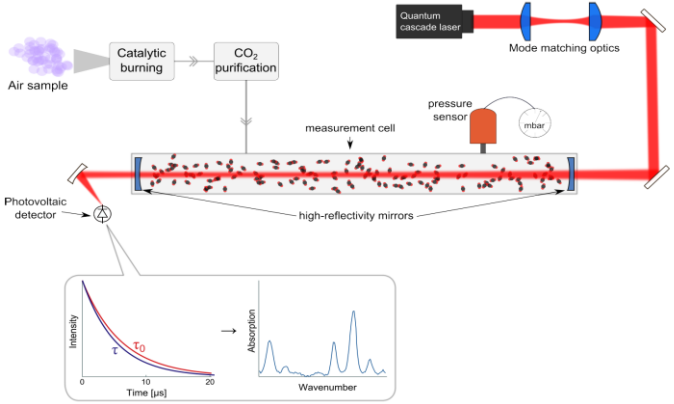
Cavity for industrial measurements





New alternative for in-situ measurements



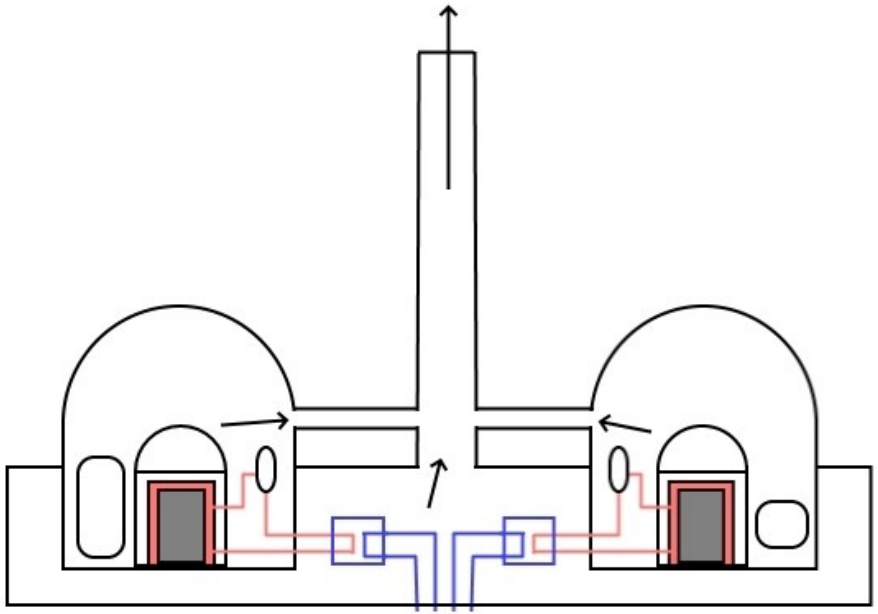


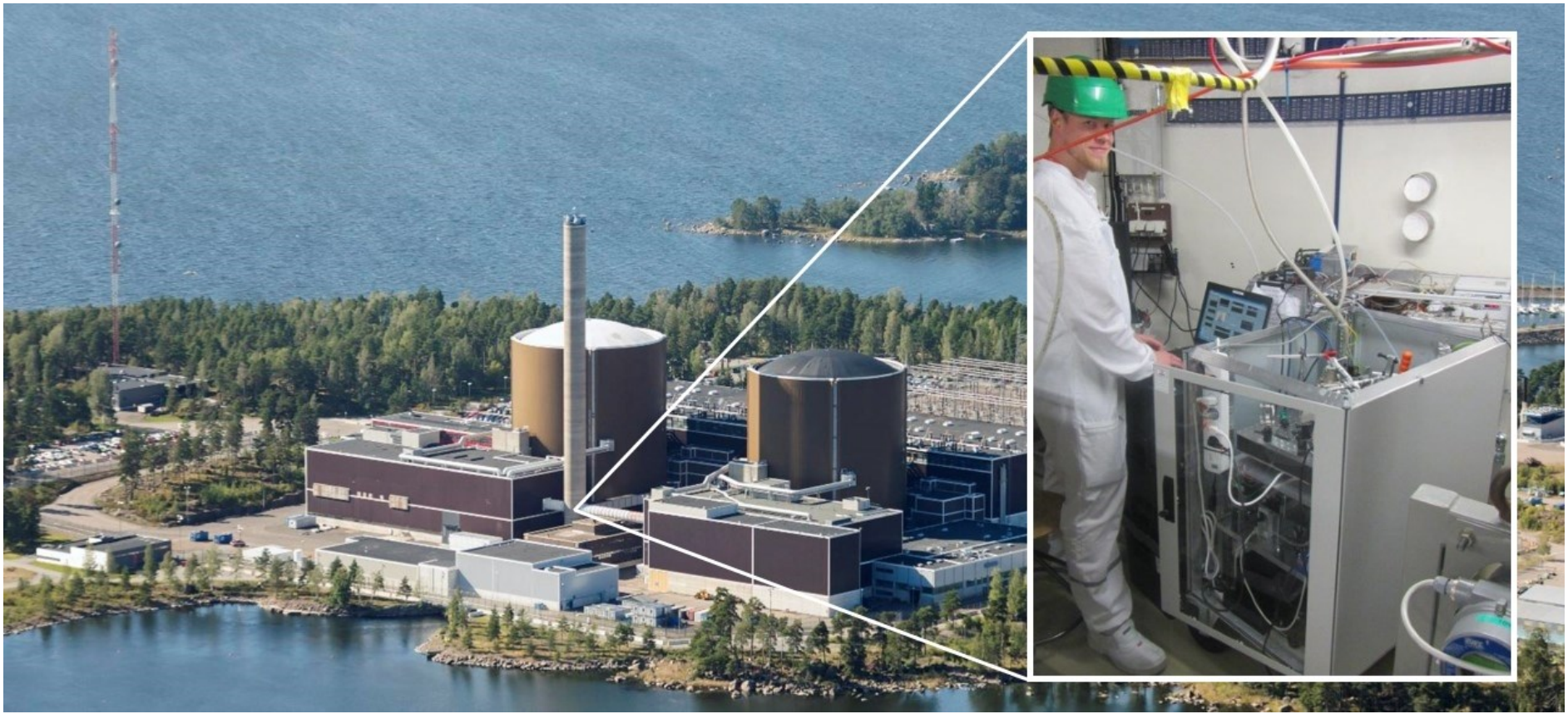
+ Compact
+ Fast measurement
+ Gas samples
+ No complex sample preparation
+ Small sample volume of a few ml
+ Only sensitive to one type of molecule
+ No additional radioactive waste

Field measurements of C-14 stack emissions

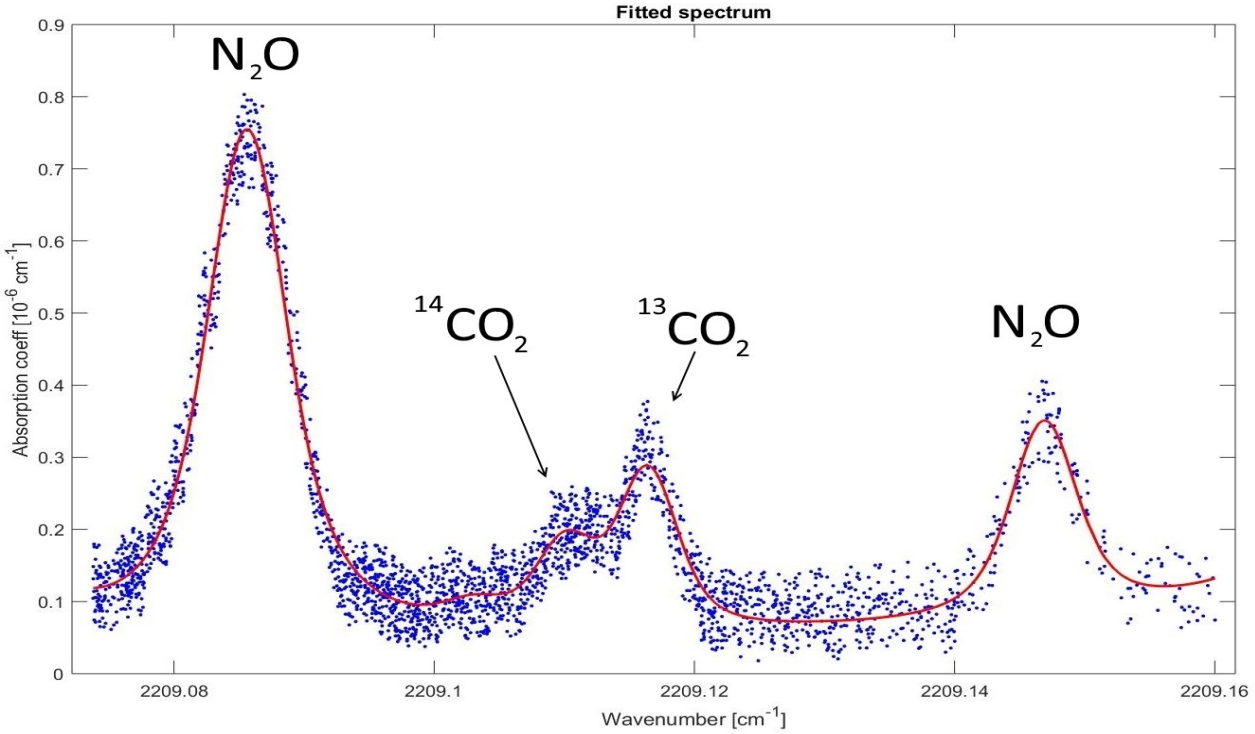
Origin of C-14 stack emissions

- Usual radiocarbon levels: 25 – 225 Bq/m^3
  - Corresponds to 0.5 – 5 ppb $^{14}C/C$

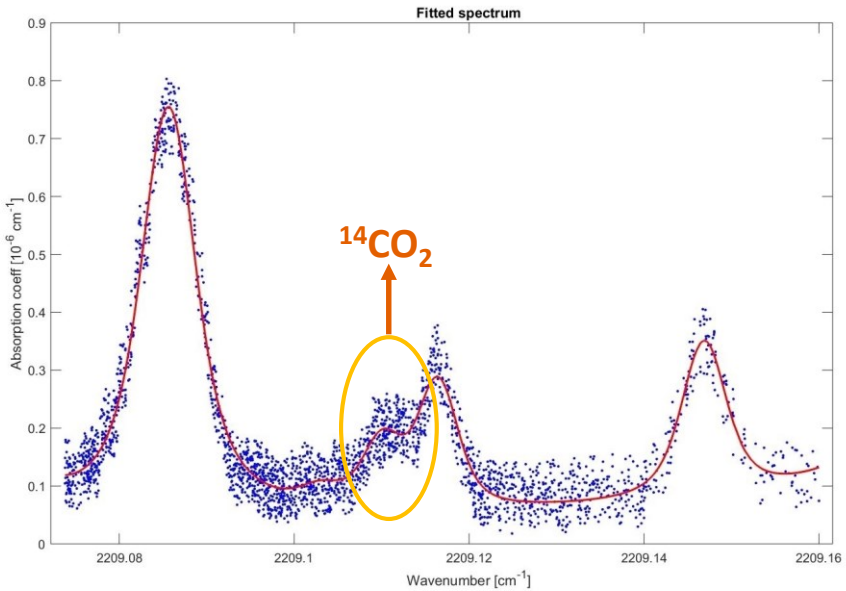




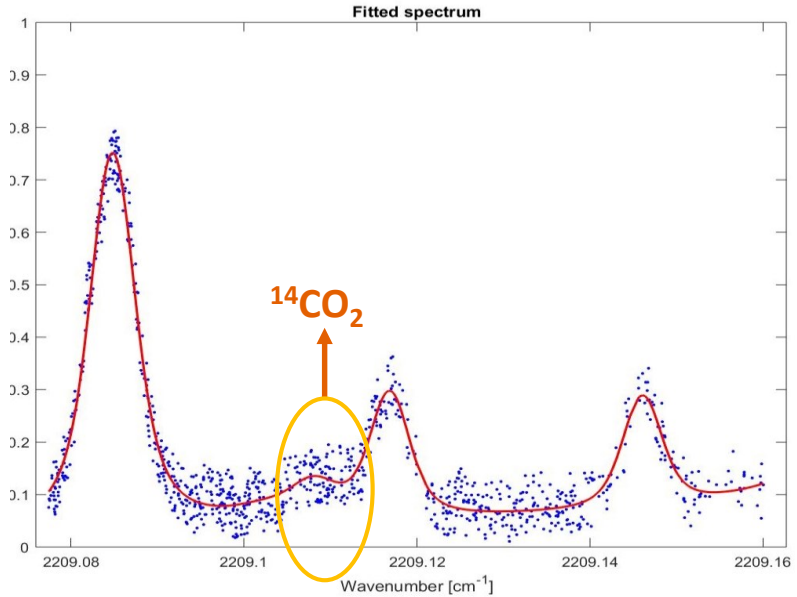
$^{14}CO_2$ from the stack measurements



Amount of $^{14}CO_2$ from line fitting



21.12.2018
122 Bq/m^3
2.7 ppb

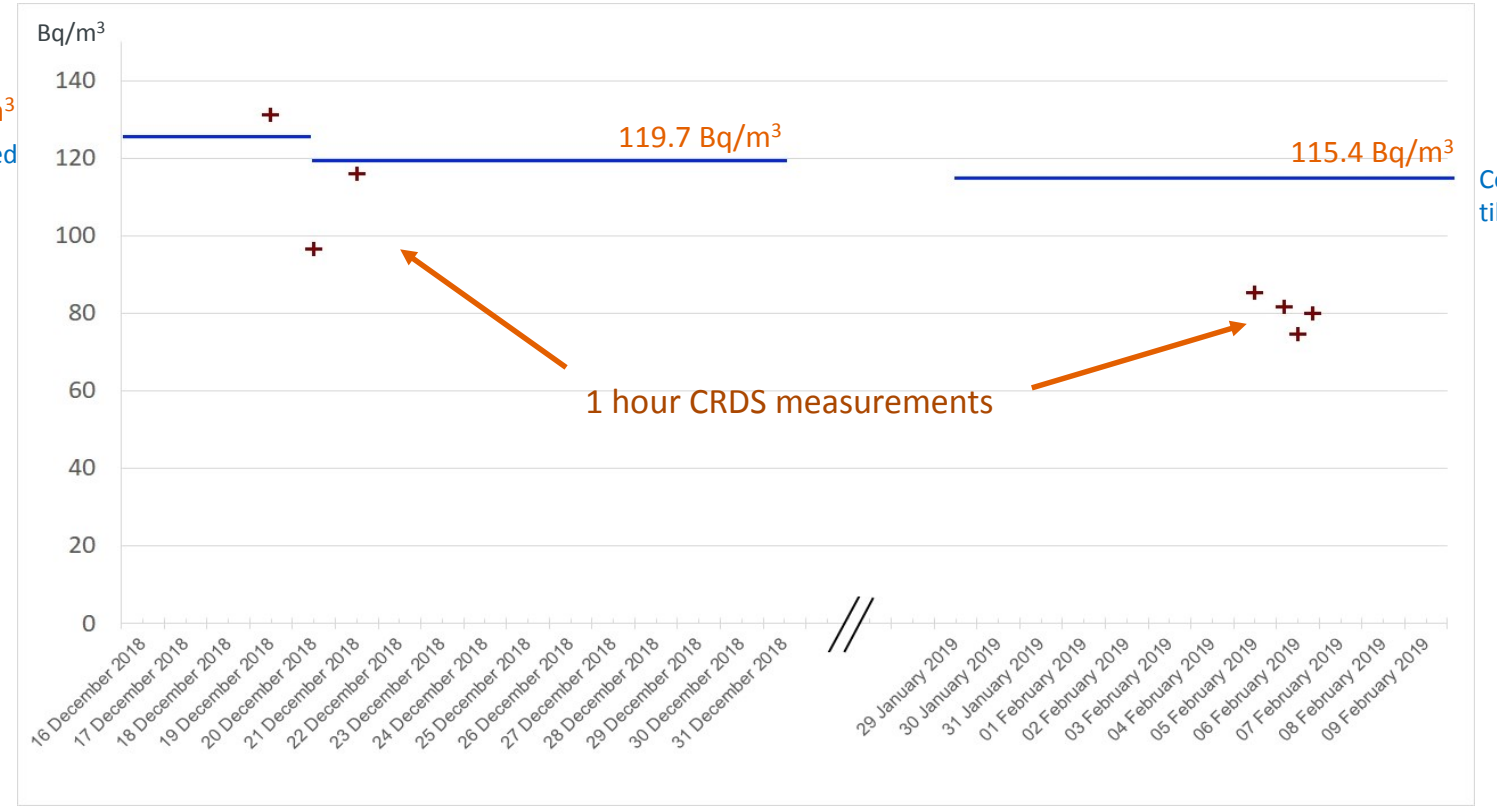


5.2.2019
85 Bq/m^3
1.8 ppb

CRDS data and LSC averages compared

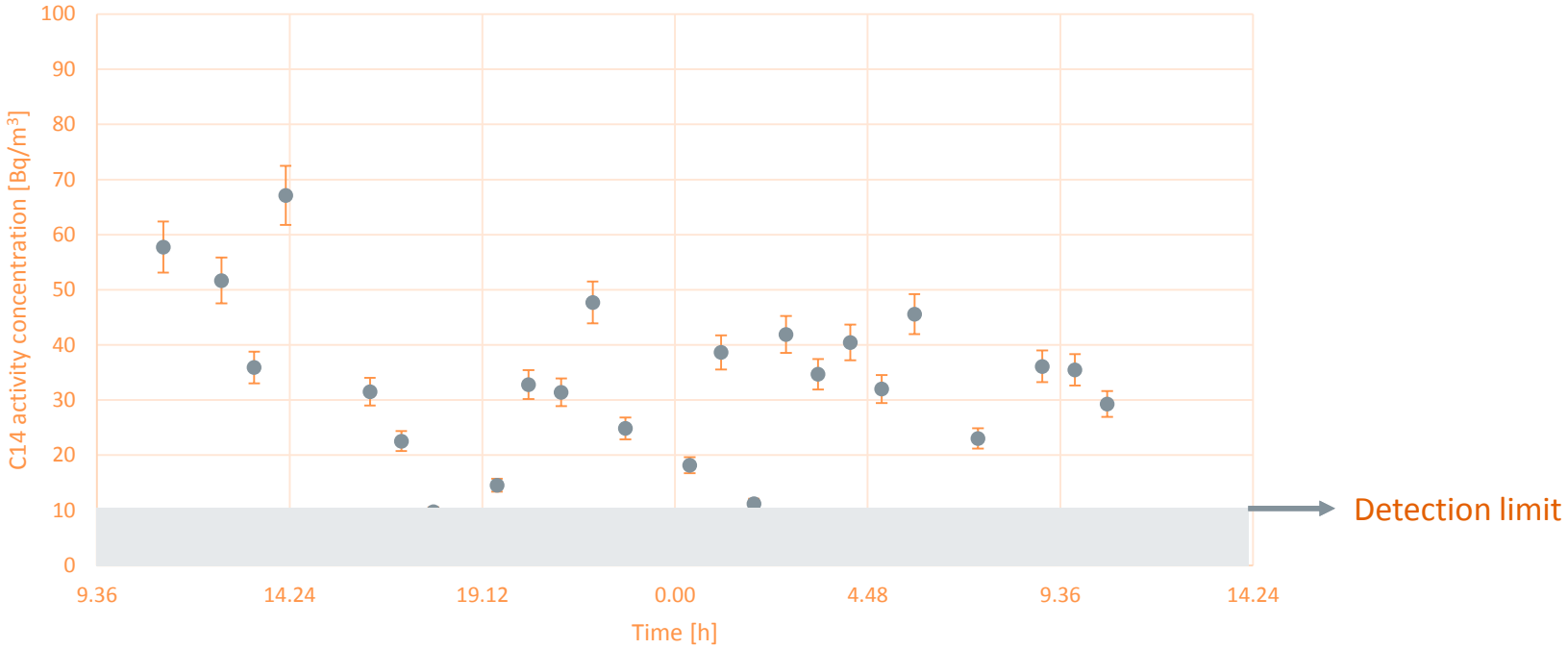
126.8 Bq/m³

Collection started on Dec 10th



Collection continued till Feb 12th

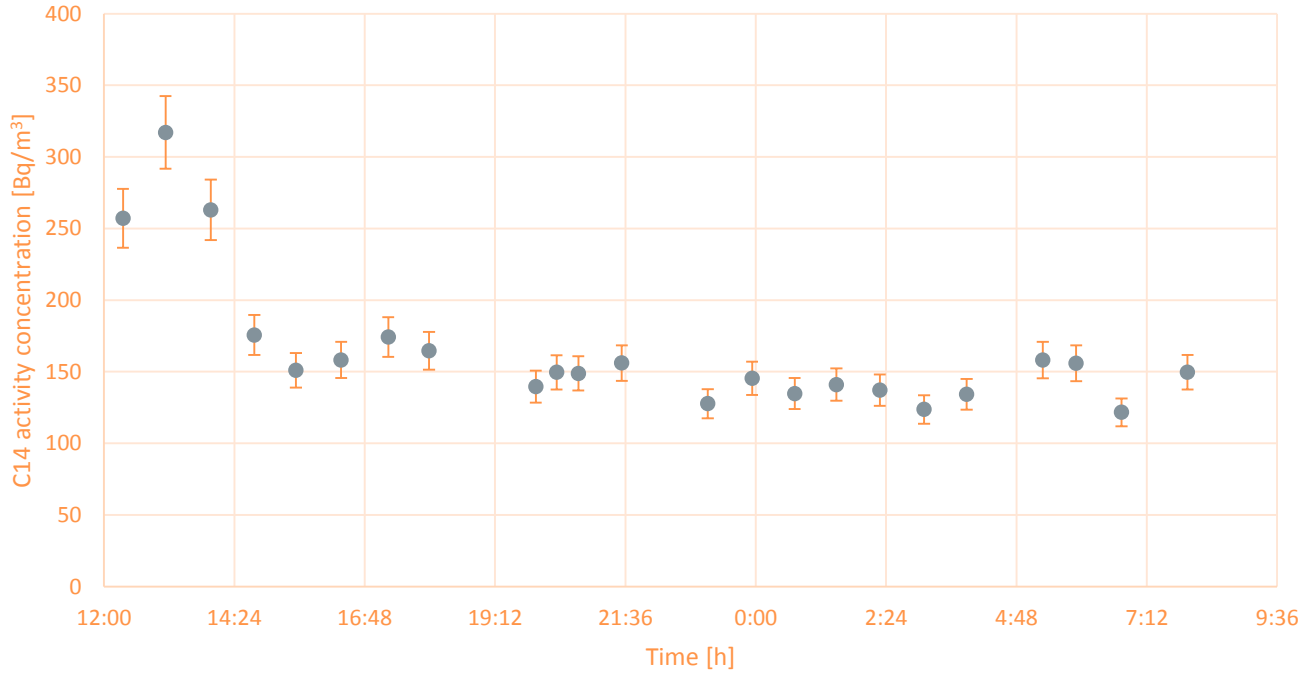
Automated continuous monitoring



24 h continuous measurement with data point every 50 min

October 2019

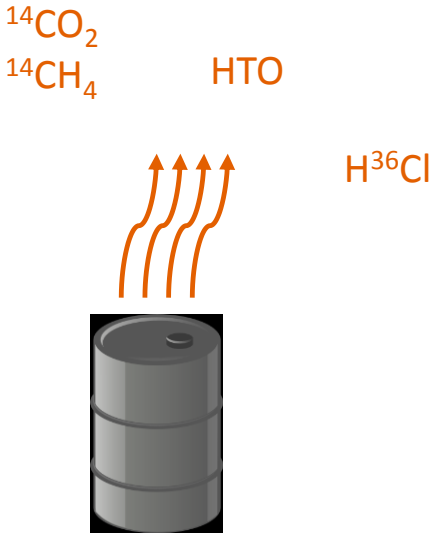
Increased time resolution



Optical methods for rapid waste characterisation

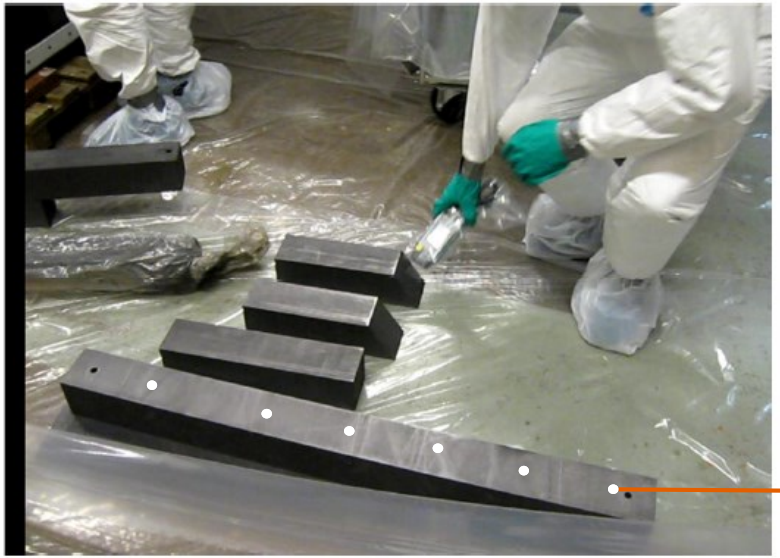
Wate outgassing waste

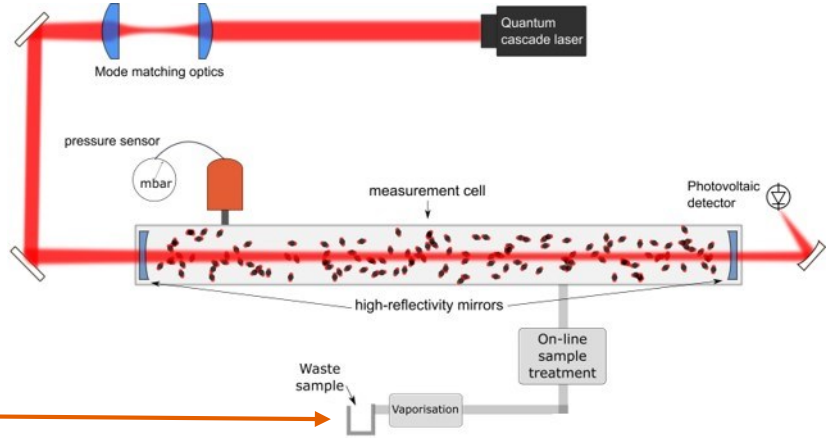
- Study outgassing (speciation) as function of storing parameters
- Test cases: graphite and spent resins
- Possibility to study a large variety of parameters



CHANCE
Characterization of Conditioned Nuclear Waste for its Safe Disposal in Europe

Rapid waste characterisation





5 mg of graphite sufficient for one CRDS measurement

No interference from other radionuclides

→ No need for complex sample preparation

→ Rapid determination of the C-14 content

Conclusions

- New method for continuous automated C-14 monitoring
- Increased time resolution can bring information that was not available before
- Potential also for other difficult to measure radionuclides
- Also suitable for nuclear waste outgassing studies and rapid waste characterisation