Il set-up per misure ambientali

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Why PIXE for aerosols analysis?

+

- 1-5 min bombardment to detect up to 20 elements (Na to Pb)
- Good detection limits down to µg/g (ng/m^3)
- Possibility to analyze sample with little mass
- Non-destructive analysis (further analysis by other techniques possible!) without any sample preparation
- It can be complemented with other IBA techniques

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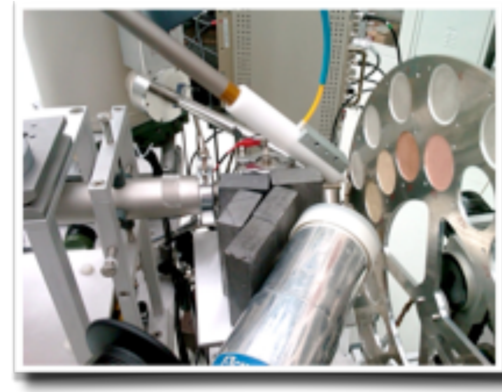
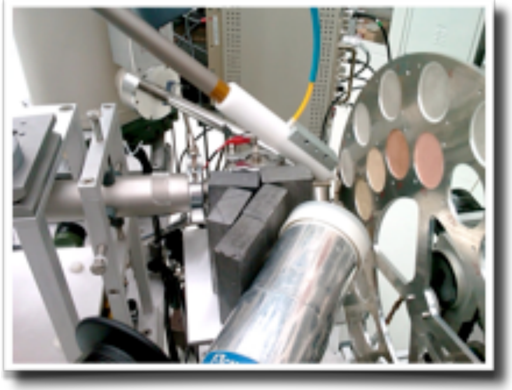
- PIXE provides only part of the desired information
- Other competitive techniques: ICP-MS
XRF
SR-XRF

A dedicated set-up



- Not a "multi-purpose" line
- Main differences with Cultural Heritage samples:
  - Beauty
  - Small samples (10-300 μg/cm^2) on filters/foils
  - Damage is less important
  - High troughput analysis is needed: we need high statistics!
- Important differences in beam currents, counting rates and measuring times

External set-up

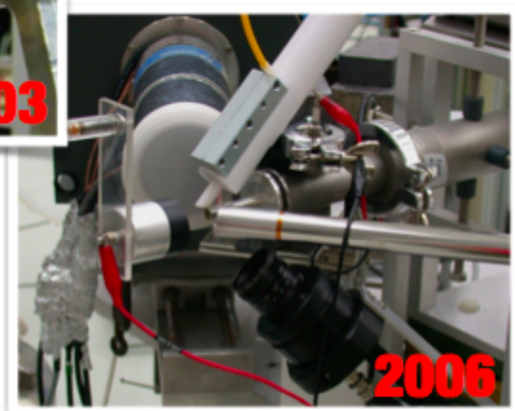


- Easy handling, positioning, changing and/or scanning of the samples
- Good heat dissipation: reduced loss of aerosol volatile components (Cl, Br) under beam irradiation
- No mechanical stress for transients to/from in-vacuum conditions
- External beam set-up based on two detection systems for low-Z and medium-high-Z elements

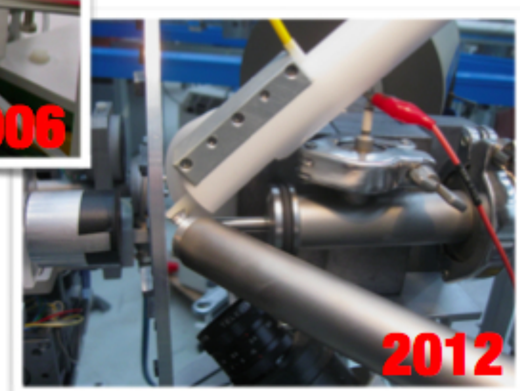
Target	X-rays	What is needed	Detector features
Low-Z elements	Low energy High cross sections	Minimum dead layers Small solid angles	Thin entrance window Small active area
Medium-high-Z elements	High energy Low cross sections	Efficiency Large solid angles	Large active thickness Large active area

Evolution of PIXE set-up for aerosols

2 Si(Li):
10 mm^2, 3 mm
80 mm^2, 5 mm

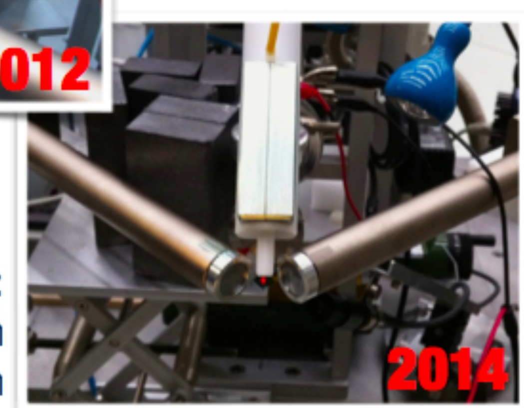


SDD: 7 mm^2, 0.3 mm
Si(Li): 80 mm^2, 3 mm



2 SDDs: 7 mm^2, 0.3 mm
80 mm^2, 0.5 mm

Funded by INFN grants (NUTELLA, NUMEN, MASAI) and EU LIFE+ AIRUSE project



3 SDDs:
30 mm^2, 0.5 mm
2x 80 mm^2, 0.5 mm

Calzolai et al., NIM B 249 (2006) 928
Lucarelli et al., NIM B 318 (2014) 55
Calzolai et al., NIM B 363 (2015) 99

Silicon Drift Detectors

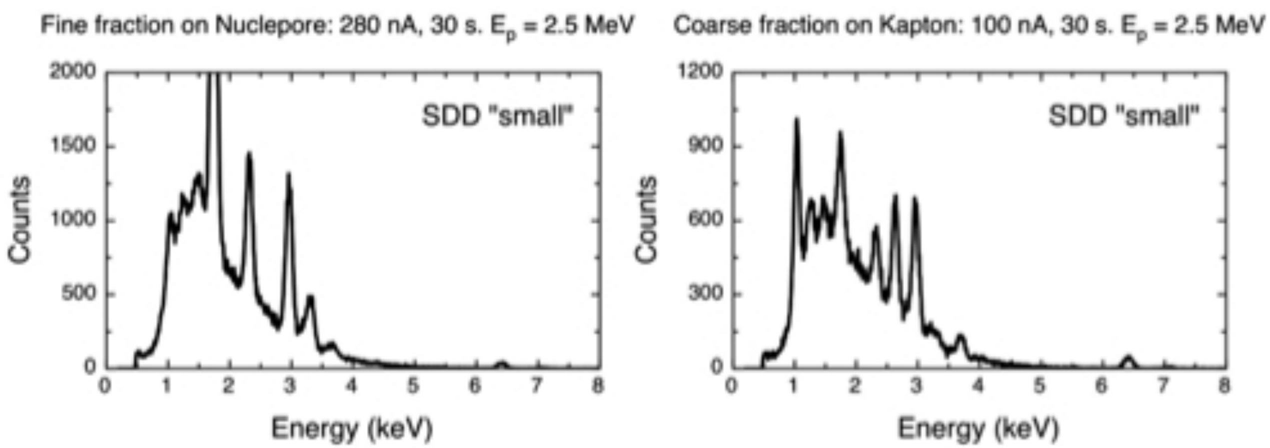
SDDs for IBA analysis:

- better resolution with respect to Si(Li)
- moderate cooling (-10/-40°C) achievable with Peltier cells
- high counting rates (up to ~50 kHz at 0.5 μs shaping time)

BUT

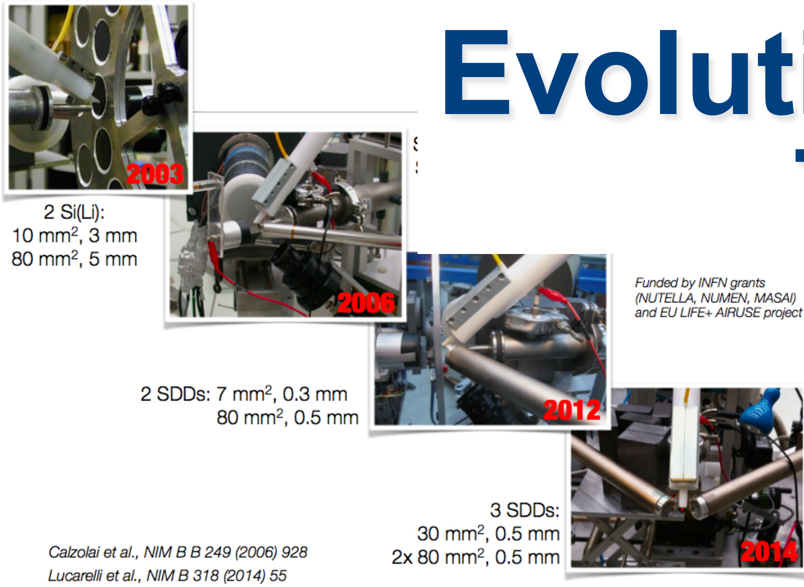
their use for a complete characterization of X-rays was limited by the small thickness and surface areas available

→ *In the beginninng SDD was introduced only for low-energy X-rays detection*



Evolution of PIXE set-up for aerosols

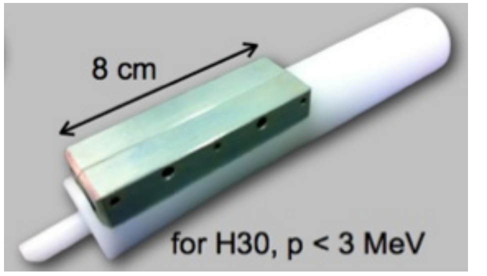
in INFN CSN5



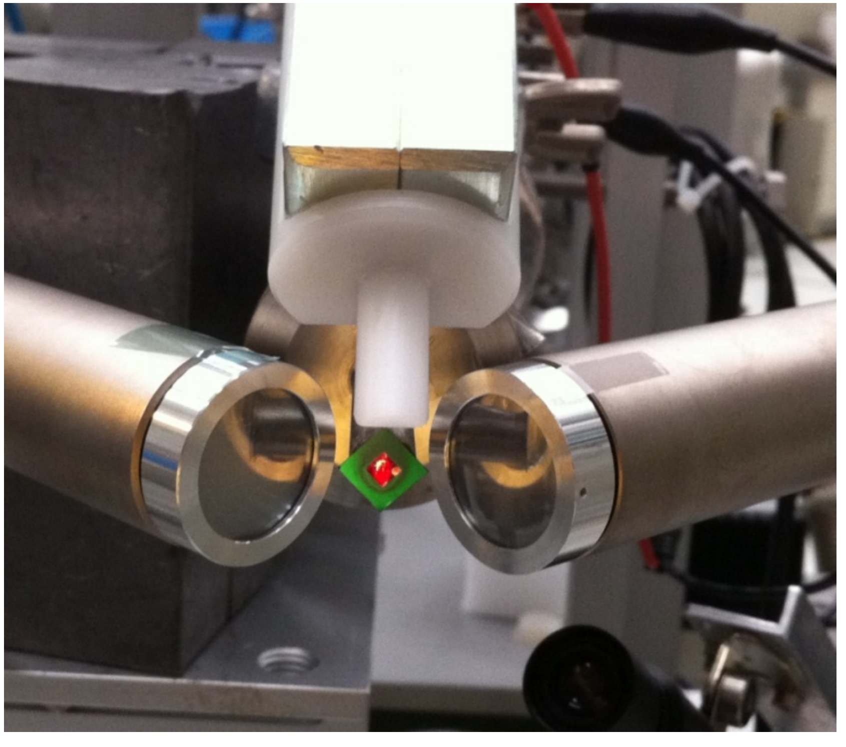
- 2017-2019: TRACCIA (Time Resolved Aerosol Characterisation: Challenging Improvements and Ambitions).
- 2015-2016: DEPOTMASS (DEtermining Particulate Organic and Total Mass in Aerosol Streaker Samples).
- 2012-2014: MANIA (Metodologie Analitiche Nucleari per Indagini Ambientali).
- 2009-2011: NUMEN (Nuclear Methods for the Environment).
- 2006-2008: NUTELLA (NUclear Techniques for Environmental PoLLoution Analysis).
- 2003-2005: MASAI (Metodologie Applicative per Studi di Arte e Inquinamento).

The current set-up for aerosols

small



- 140 eV (FWHM at 5.9 keV)
- 450 μm thick, 30 mm^2 active area
- ultrathin window (8 μm Be)
- + He flow & magnetic p deflector



Backscattering of protons from the target causes:

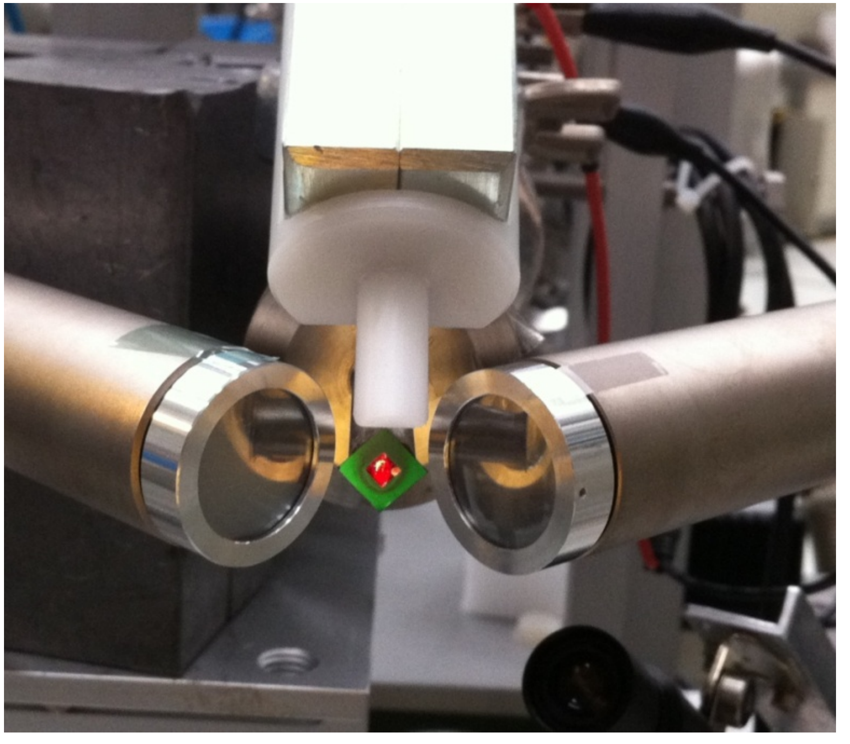
- pile-up increase
- energy resolution deterioration
- damage of the detector

The limit dose of 10^9 protons/cm^2 (=significant deterioration of the detector performance due to radiation damage) would be almost reached during the measurement of only 1 streaker sample!

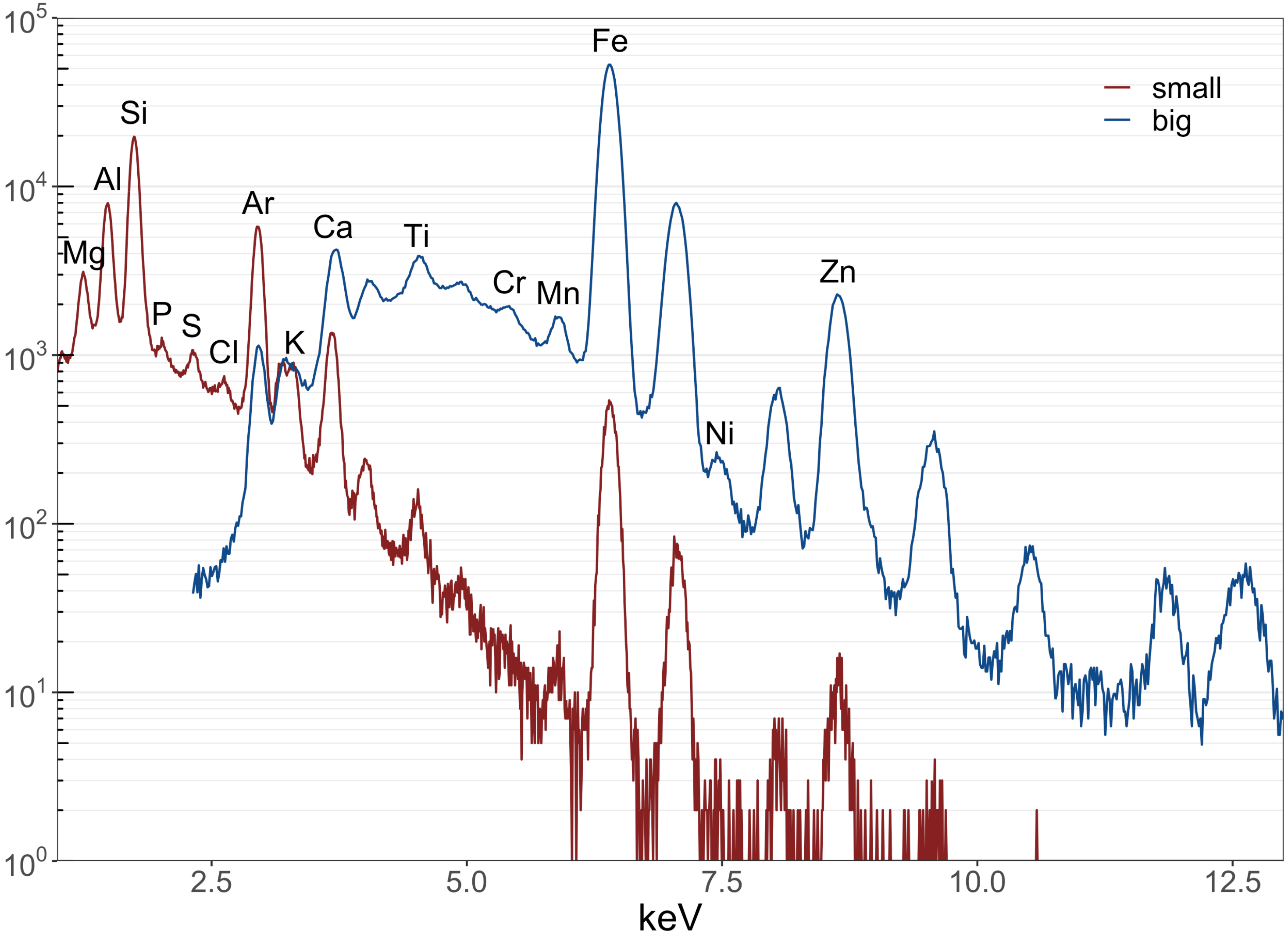
The current set-up for aerosols
big

- 165 eV (FWHM a 5.9 keV @ τ=1 μs)
- 450 μm thick, 80 mm^2 active area
- with absorber (450 μm Mylar) to attenuate low-energy X-rays

X 2



The current set-up for aerosols

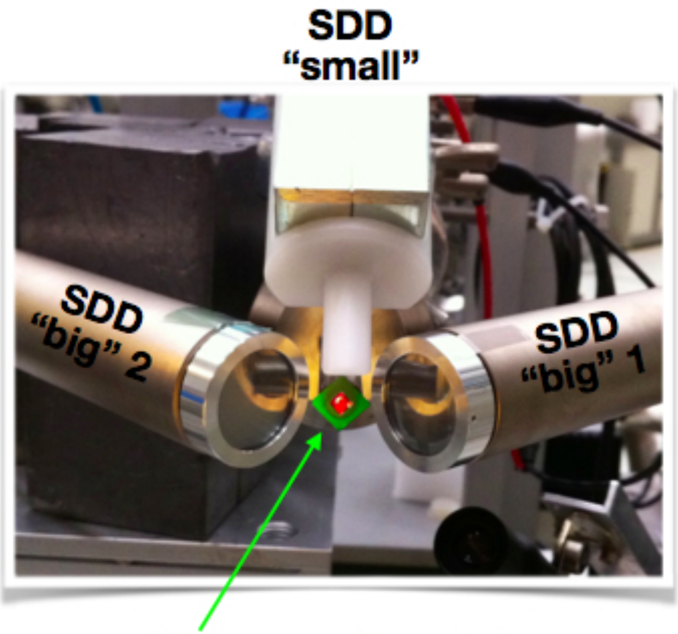


The beam extraction window

Up to 2013, 7.5 μm Upilex window:

• Progressive thinning of the window, breaking after about 800-900 μC of integrated charge.

• Measuring with ~100 nA, replacement needed every ~ 2-3 h.



Si_3N_4 extraction window
5x5 mm^2, 500 nm

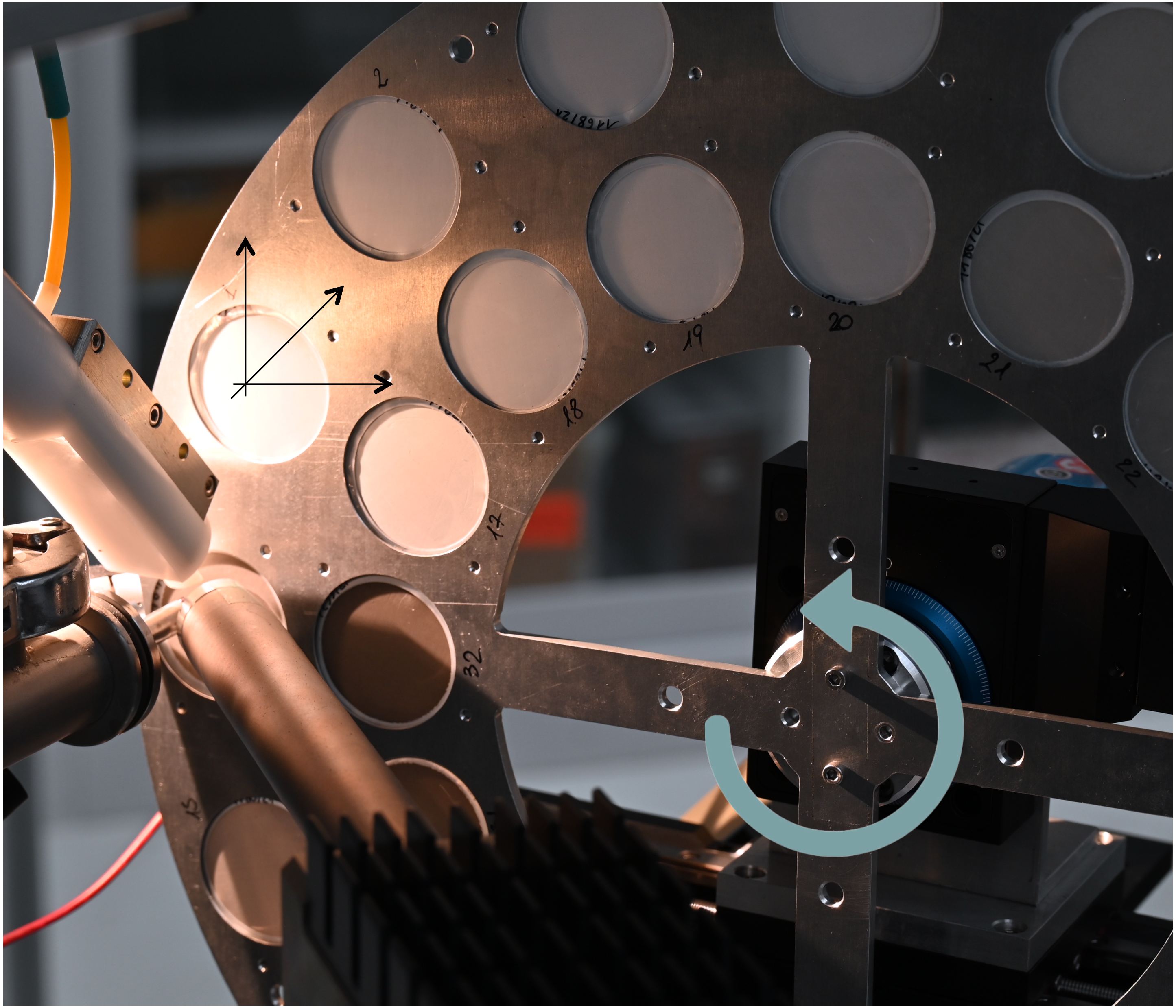
Since 2013, 500 nm thick Si_3N_4 window by Silson:

BS tests after 4600 μC showed no thinning

No need to change it for many years!

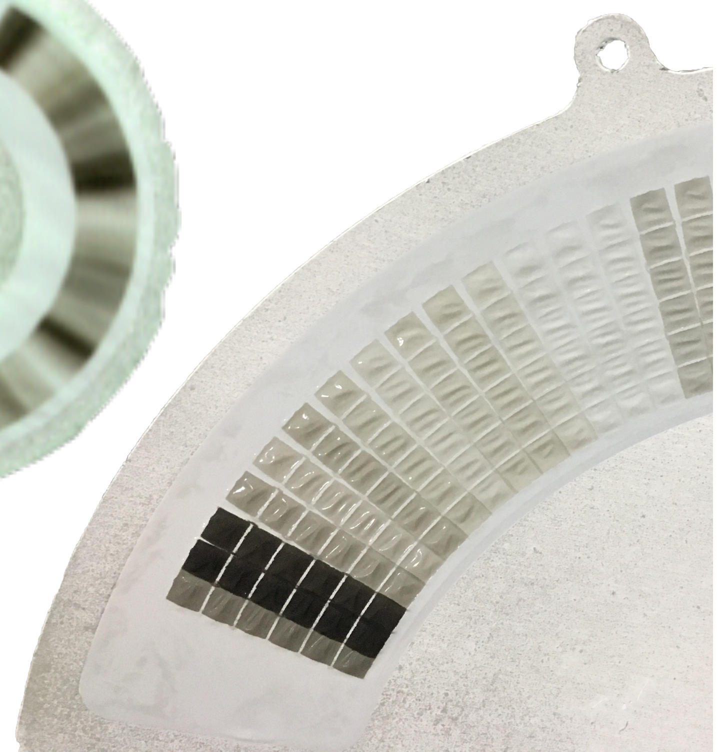
(unfortunately it was victim of a black-out...)

Stepper motors controls



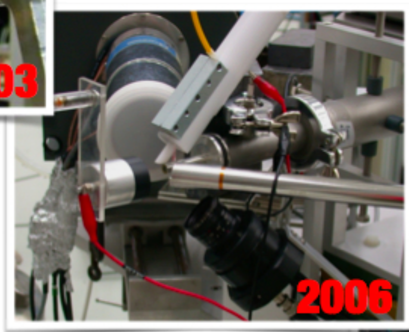
Special thanks to L. Carraresi

Stepper motors controls

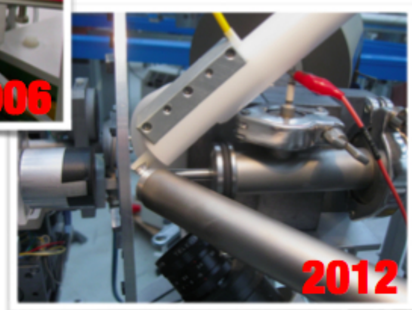


Evolution of PIXE set-up for aerosols – towards the future

2 Si(Li):
10 mm², 3 mm
80 mm², 5 mm



SDD: 7 mm², 0.3 mm
Si(Li): 80 mm², 3 mm



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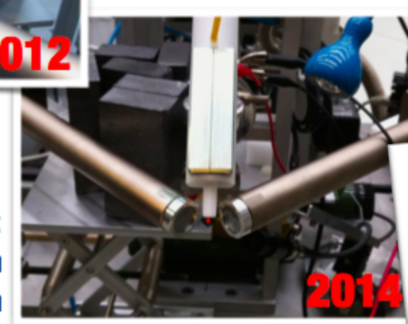
Funded by INFN grants (NUTELLA, NUMEN, MASAI) and EU LIFE+ AIRUSE project



3 SDDs:
30 mm², 0.5 mm
x 80 mm², 0.5 mm

Calzolai et al., NIM B 249 (2006) 928
Lucarelli et al., NIM B 318 (2014) 55
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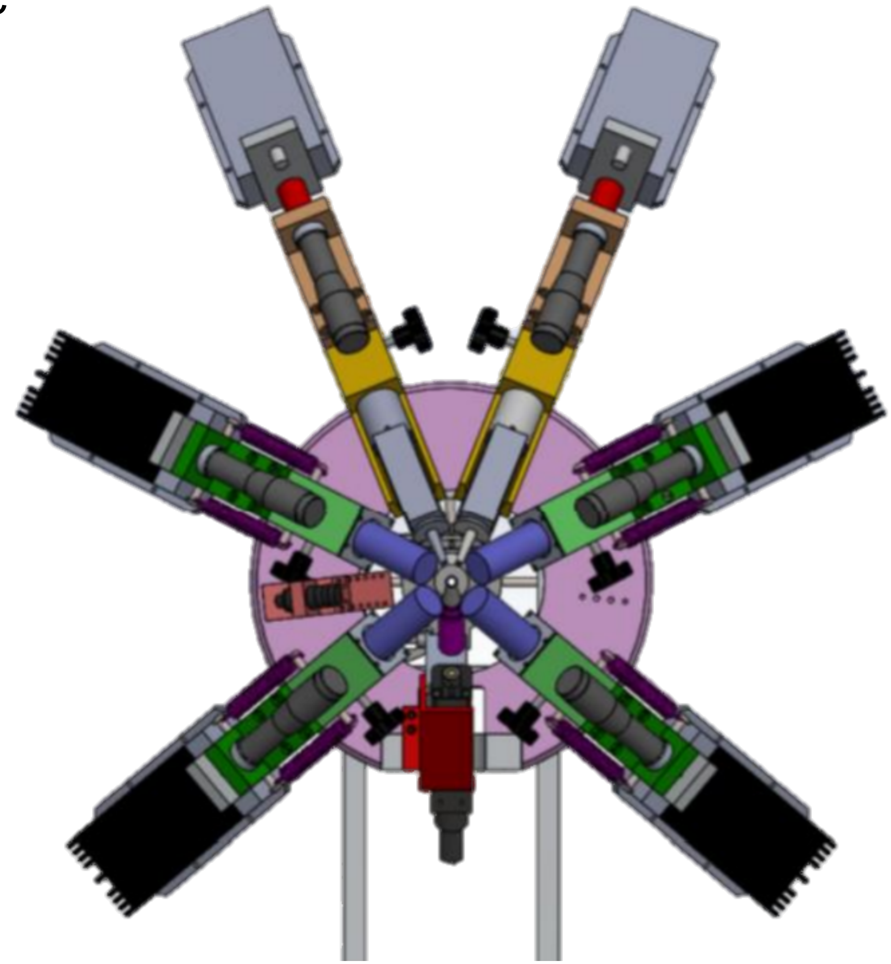
The forthcoming set-up

- Four 80 mm2 SDDs for medium-high Z elements
- Two 30 mm2 SDDs with proton magnetic deflector for low Z elements

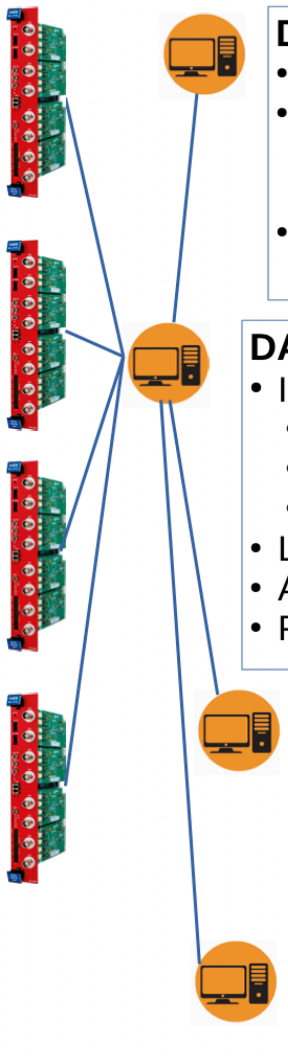
- Beam current monitoring through the detection of the Si X-rays from the extraction window



Special thanks to C. Cialdai and the mechanical workshop

The forthcoming set-up

DAQ Software



DAQ User Interface (GUI)
- ADC Parameter editing (with presets)
- Display of Real time statistics:
  - Channel counting rates (raw, pile-up, saturated)
  - Dead-time estimation
- Acquisition start (manual) and stop (manual or timed)

DAQ Server
- Implements the ADC interface (Optical Link)
  - Through dedicated CAEN libraries
  - Parameter Write/Read (Measure Setup)
  - Data Readout and storage
- Local storage of used setups
- Acquisition Coordination (manual/timed)
- Provides real-time spectra for inspection

Real Time Data Viewer (ROOT based)
- On-line spectra inspection and manipulation
- Definition of ROIs for statistics estimation (counts for selected elements) and automatic calibration
- Real-time statistics (ROI counts) viewing

Real Time Browser Data Viewer
- On-line spectra inspection via Browser
- Display of measurement statistics

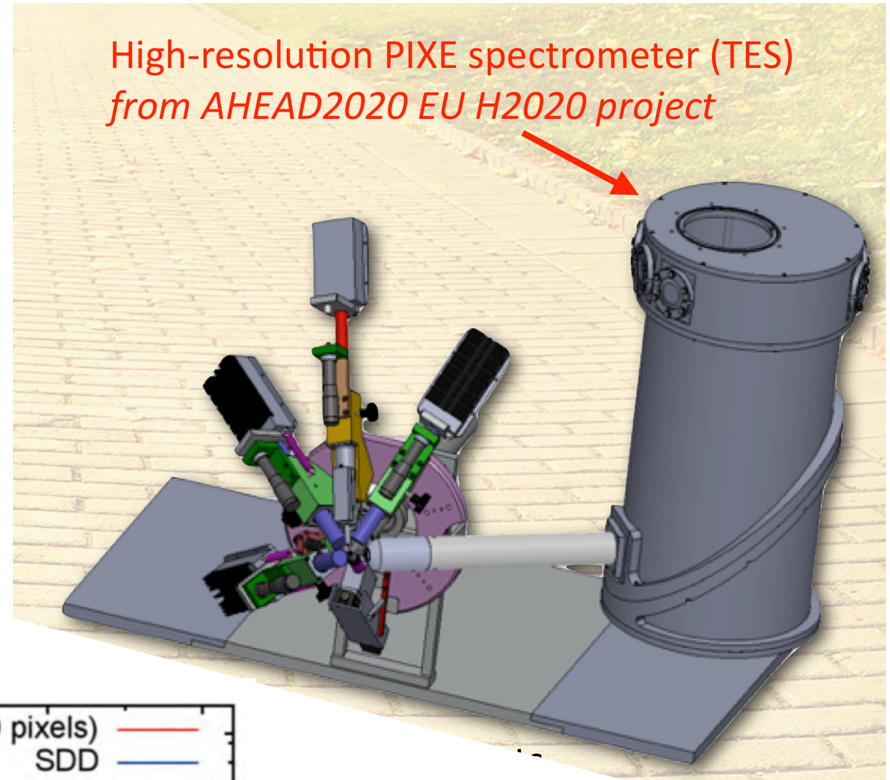
The new digital acquisition handles up to 8 different channels

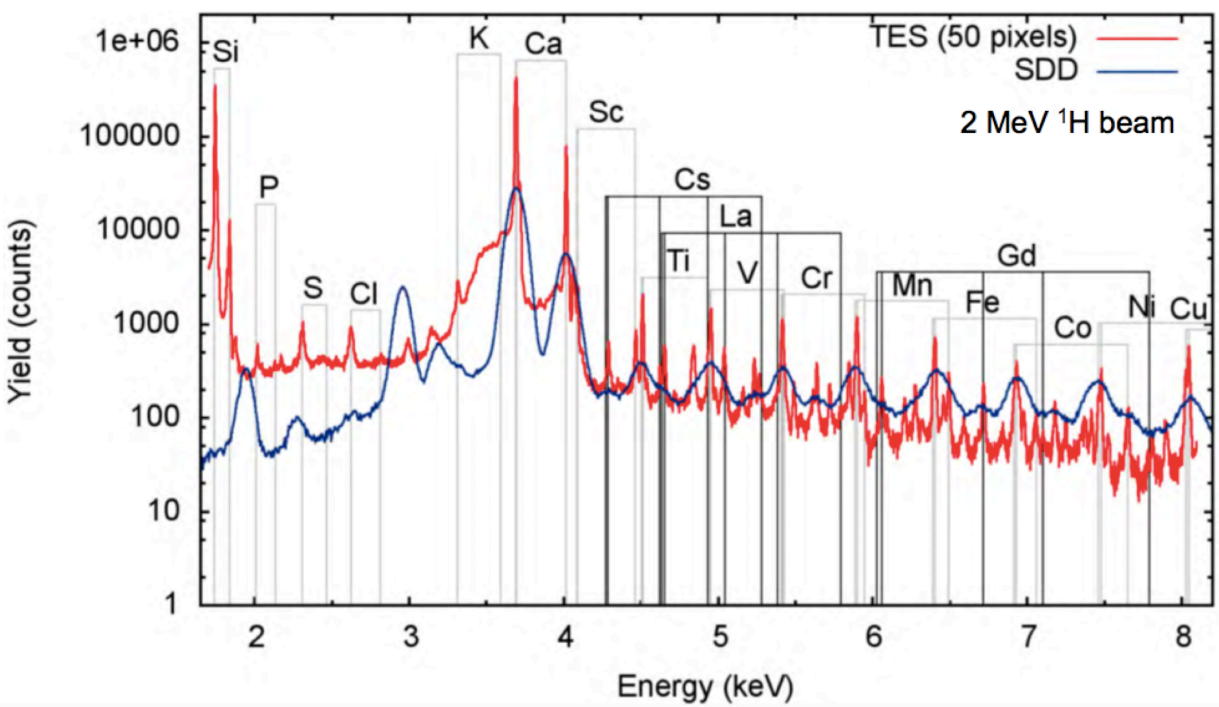
Special thanks to P. Ottanelli

New possibilities

Test of a High-resolution imaging X-ray spectrometer based on superconducting transition-edge sensor

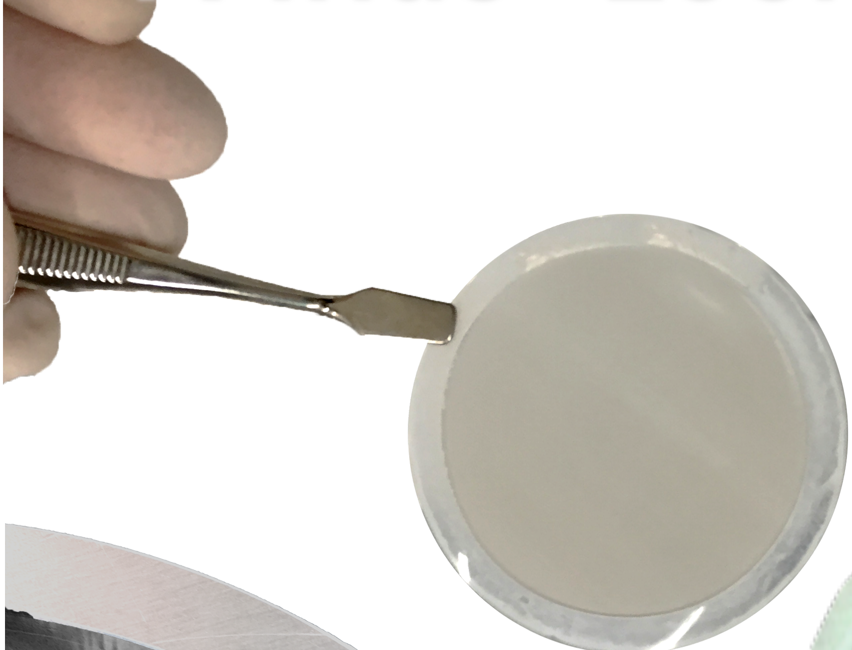
developed for astrophysics for PIXE purposes

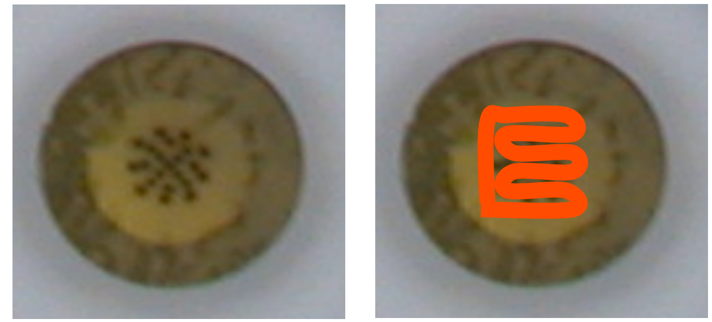




Comparison of PIXE measurements with 2 MeV protons on a glass reference standard with a TES detector and with a traditional Silicon Drift Detector (SDD)

A wide “zoology” of samples:





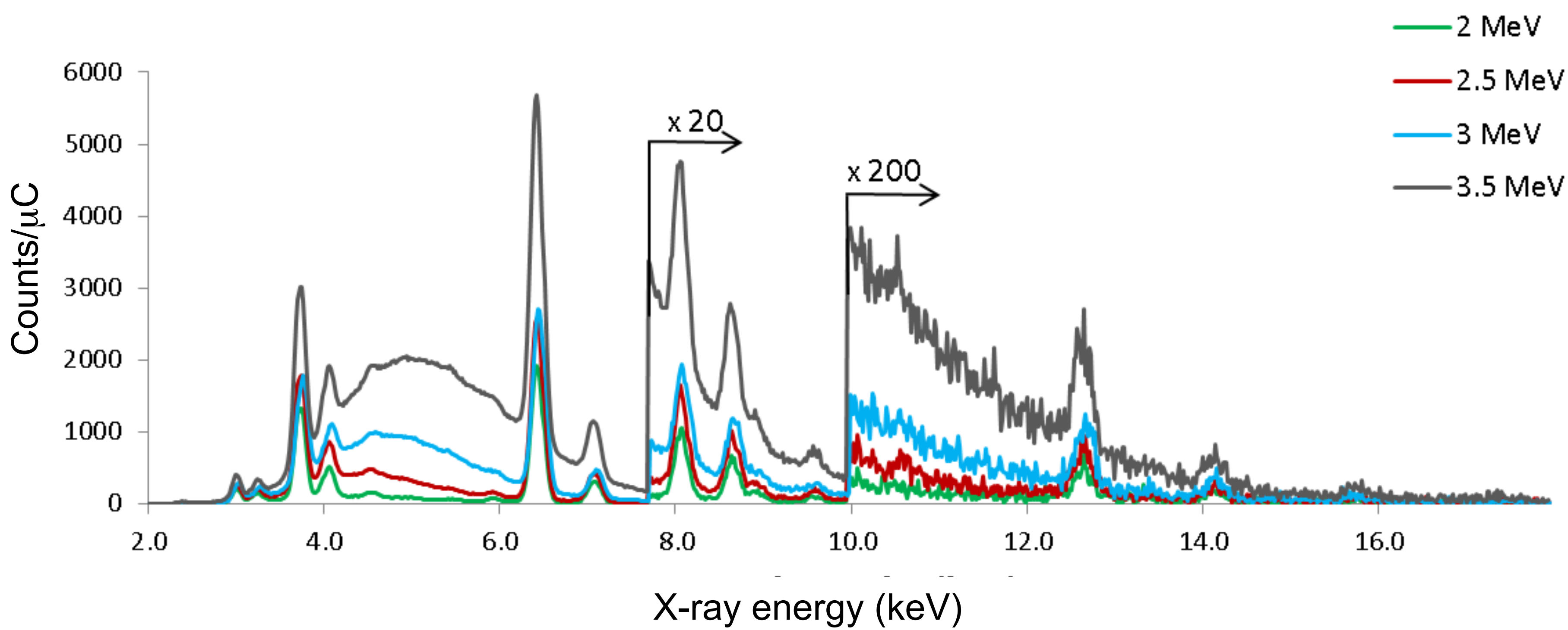
best measuring conditions?

Hourly samples: which energy?

Hourly samples collected on: Policarbonate filters, Kapton/ Polipropilene folis

Increasing the beam energy:

- X-ray emission cross-sections increase
- BUT: also the background increases



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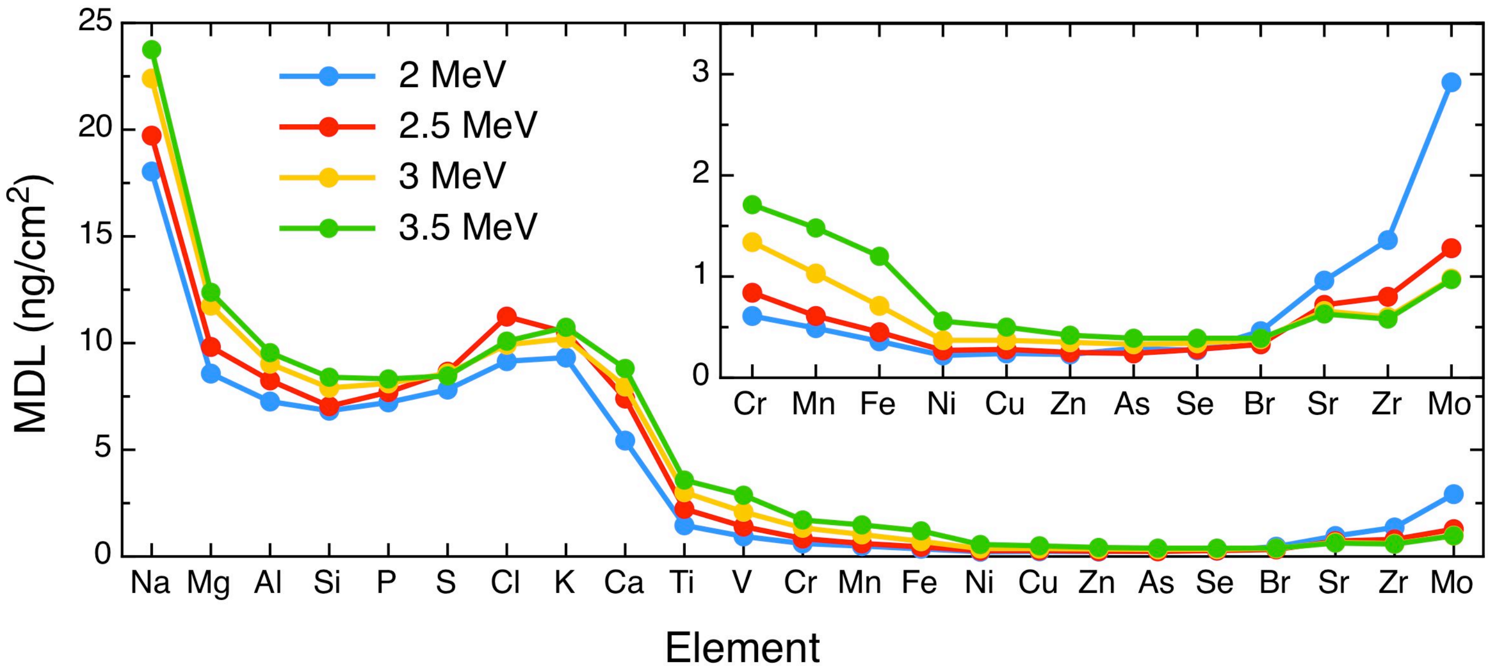
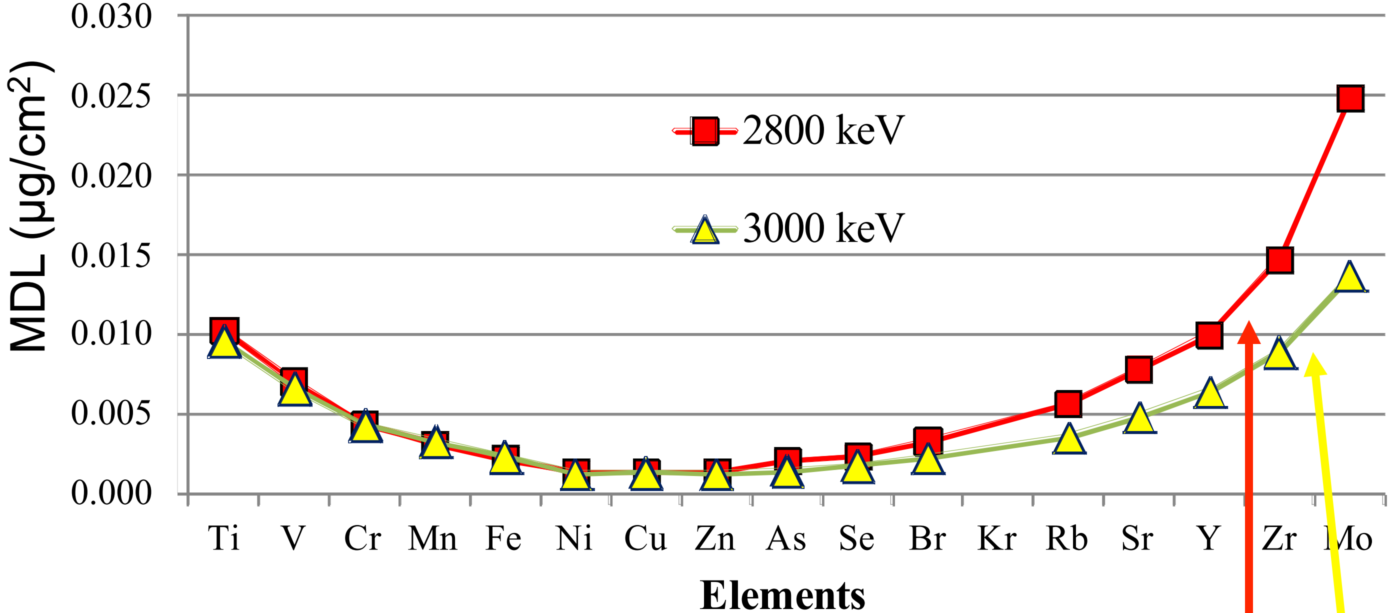
→ MDL study

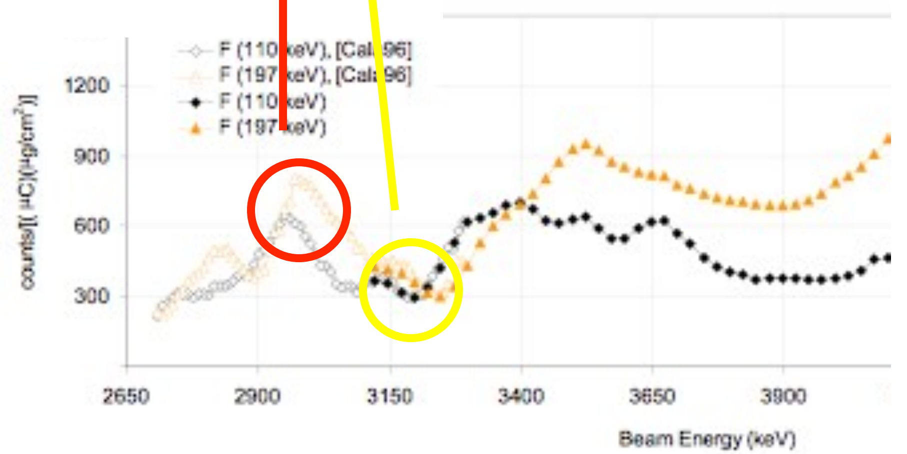


Taking into account the importance of the high-Z elements in aerosol studies and the average aerosol composition, with prevailing low-Z elements, energies between 2.5 and 3.0 MeV appear to provide the best compromise

Daily samples on Teflon (CF_2)



The F of Teflon gives rise to a strong Compton background in PIXE spectra due to the emission of several γ-rays from the $^{19}F(p,p'\,\gamma)^{19}F$ and $^{19}F(p,\alpha\gamma)^{16}O$ reactions



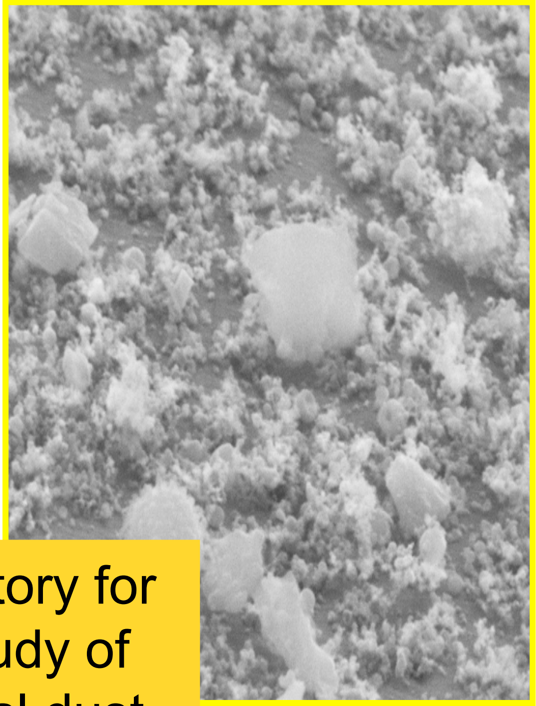
PIGE: why? Matrix effects for PIXE

ΔE ≤ 100 keV for few MeV p in a PM10 deposit:

✓ Small PIXE cross section variations within energy loss inside the sample

✓ Self-absorbtion of the emitted X-rays inside the aerosol particles:

- a problem for a quantitative PIXE analysis on low-Z elements: Na, Mg, Al, Si
- it can not be calculated considering an average matrix composition



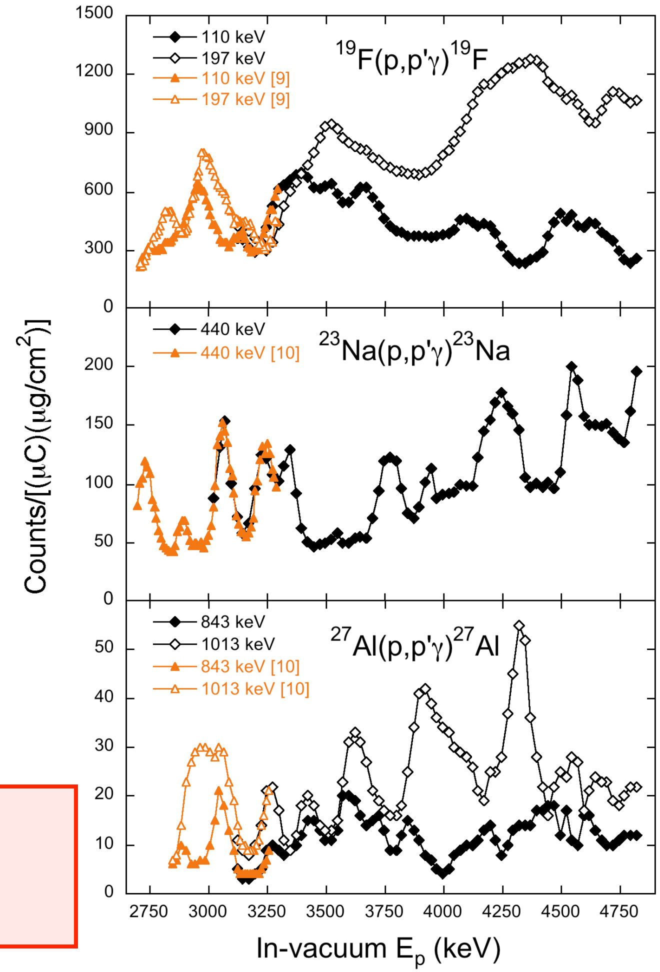
mandatory for the study of mineral dust

PIGE: which energy? Matrix effects for PIGE

ΔE ≤ 100 keV for few MeV p in a PM10 deposit:

- ✓ Self-absorbtion of the emitted γ-rays inside the aerosol particles: negligible.
- ✓ PIGE cross section strongly energy dependent:
  need to find *plateaux* where the cross sections are constant over ΔE! So: analysis by comparison with thin standards is possible.

External set-up: fluctuations in the PIGE cross sections smoothed: easier to find a *plateau*!!

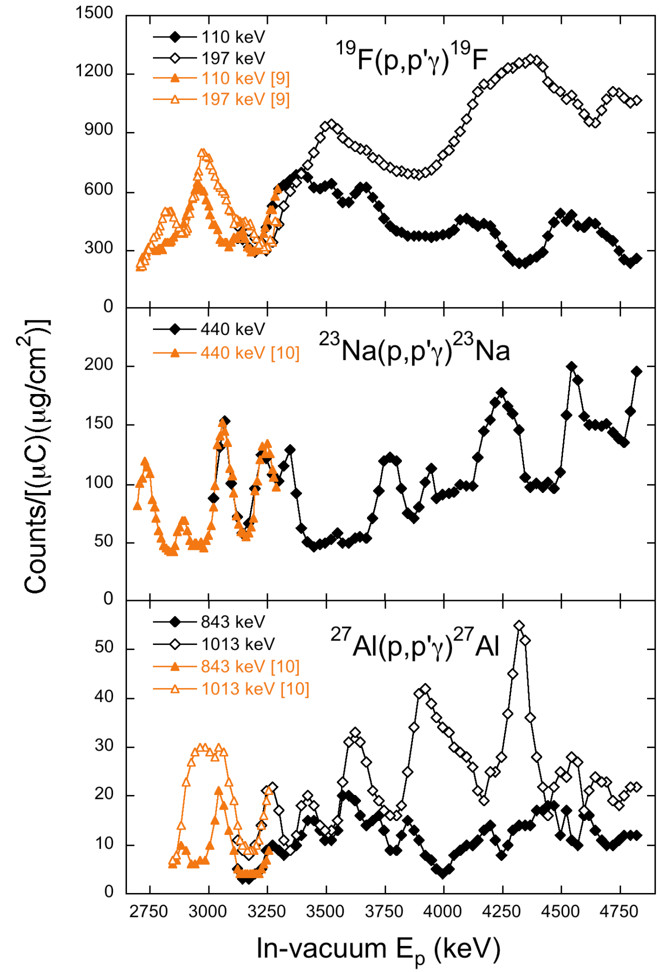


G. Calzolai et al., NIM B 268 (2010)

PIGE: which energy? Matrix effects for PIGE

Reaction	E_γ (keV)	E_p (keV)	E_p on target[a] (keV)	Width (keV)	Average yield [counts/(μC·μg/cm²)]
$^{19}F(p,p'\gamma)^{19}F$	110	4020	3863	175	380
		4395	4248	100	250
	197	3945	3785	125	700
		4420	4274	150	1250
$^{23}Na(p,p'\gamma)^{23}Na$	440	2990	2791	70	50
		3520	3346	100	50
		3670	3501	100	55
		4045	3889	75	90
		4470	4326	100	100
		4695	4556	100	150
$^{27}Al(p,p'\gamma)^{27}Al$	843	4370	4223	75	14
		4745	4607	75	10
	1013	3065	2870	120	29
		3820	3657	75	18
		4695	4556	75	23

[a] The proton beam energy E_p on target is calculated taking into account the energy loss in the 7.5 μm Upilex exit window and in the external path of 5 mm of air and 5 mm of He.

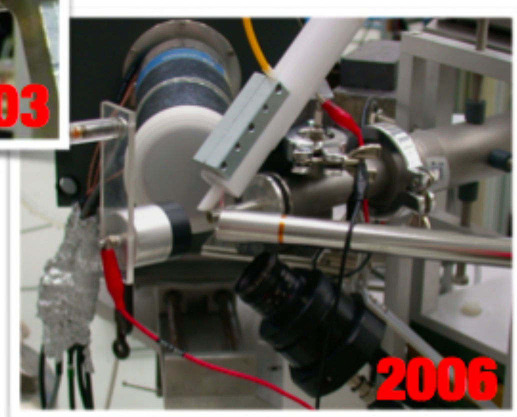


G. Calzolai et al., NIM B 268 (2010)

- ✓ Interelemental ratios (often with respect to Al) used to differentiate aerosol sources/ potential source areas
- ✓ Uncorrected interelemental ratios may prevent the possibility of differentiation.

Conclusions

2 Si(Li):
10 mm^2, 3 mm
80 mm^2, 5 mm



SDD: 7 mm^2, 0.3 mm
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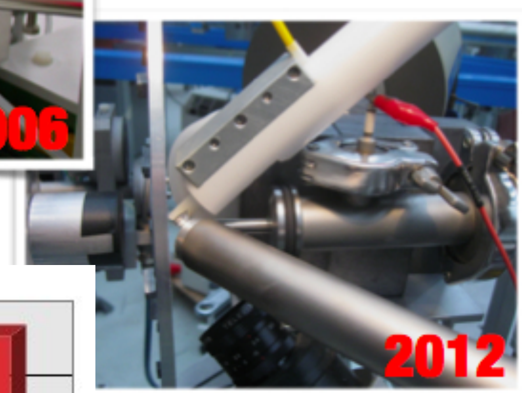

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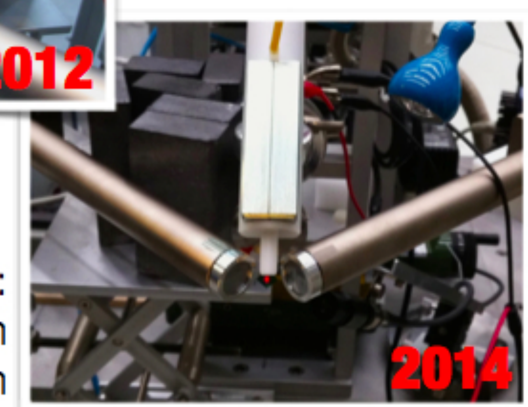
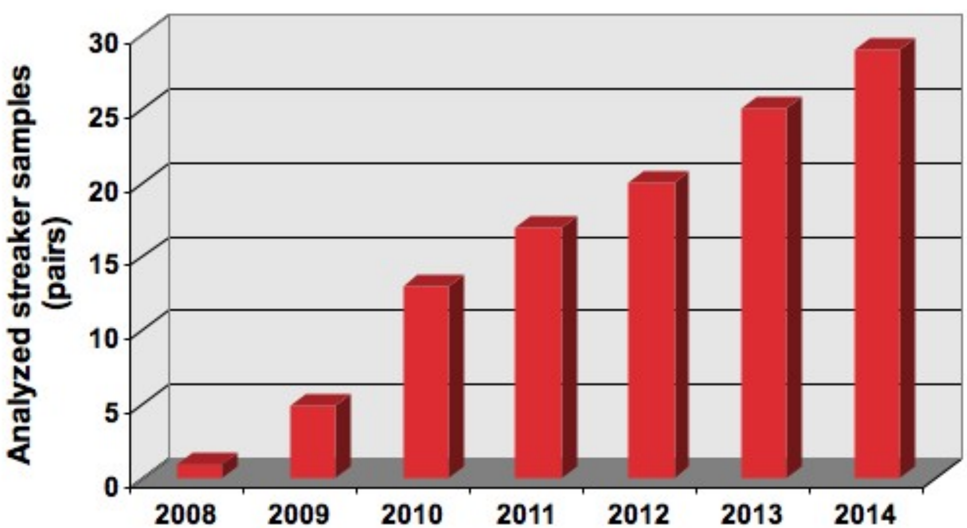


3 SDDs:
30 mm^2, 0.5 mm
x 80 mm^2, 0.5 mm

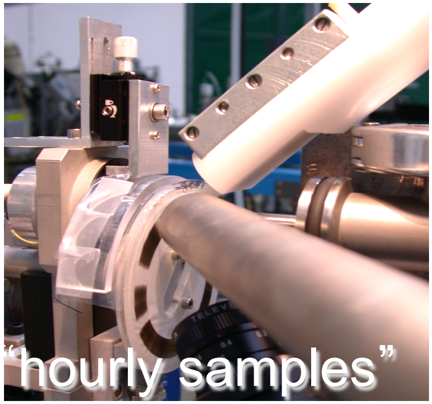


Conclusions

i ~ 50-100 nA

Measuring time: ~ 0.5 - 3 minutes

Daily samples collected in 1 year → 1-2 days



i ~ 100 - 300 nA

Measuring time: ~ 1-1.5 minutes/spot

1 sampling week with 1 h resolution: 168 spots!

1 week sampling → ~ 3-4 hours

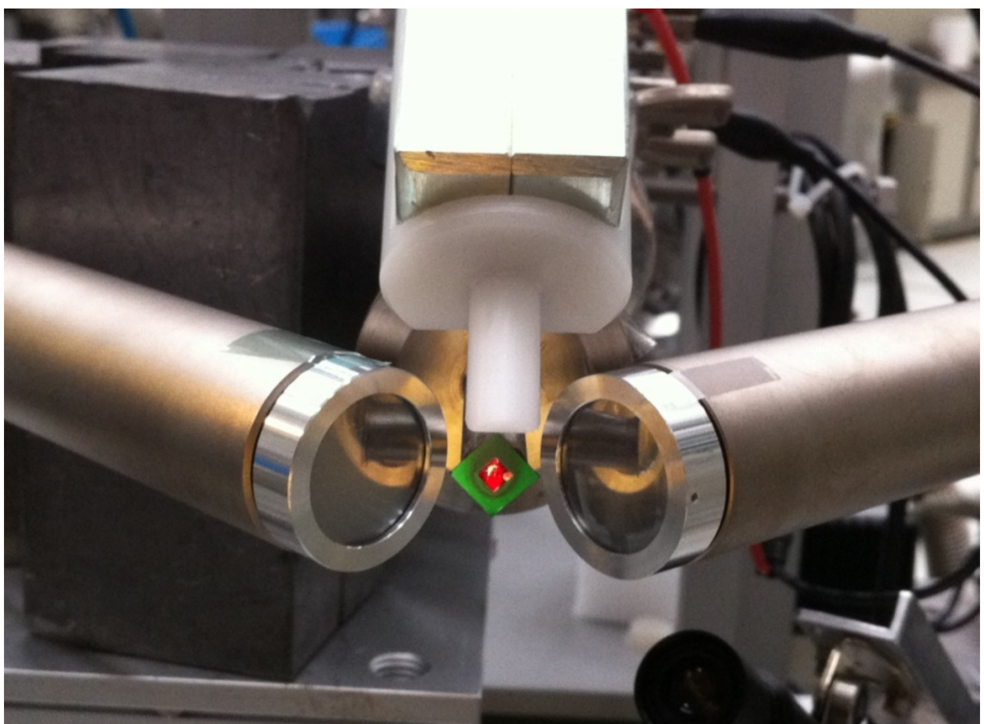
Is it everything about QUANTITY?

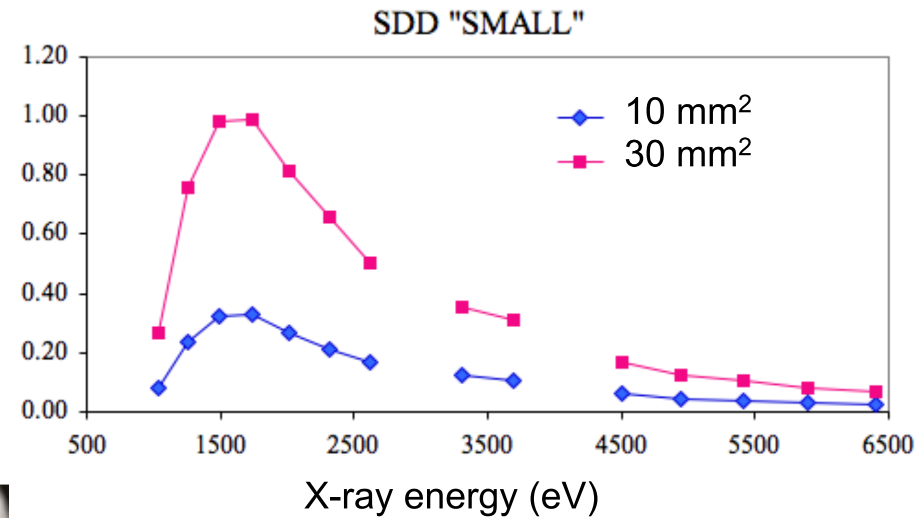
No! QUALITY is mandatory, but environmental sciences need a huge amount of data to make a study "representative"!

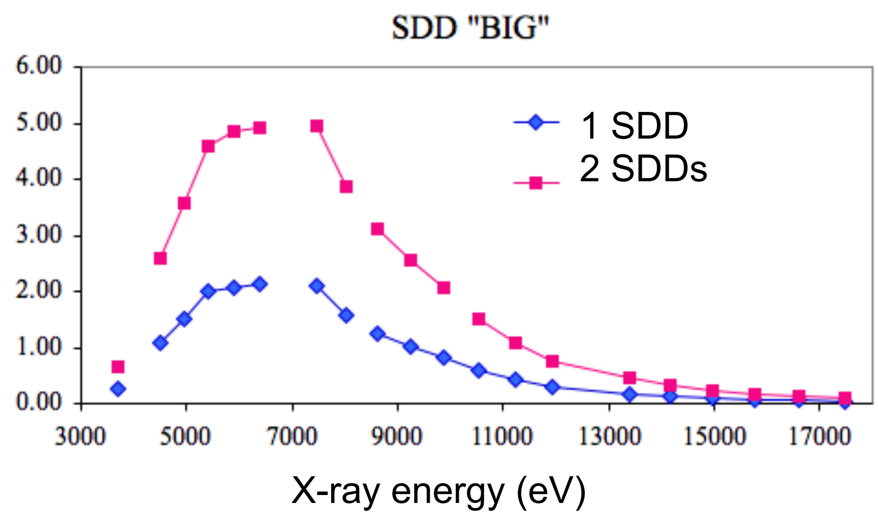
Thanks for your attention!

3-SDD set-up

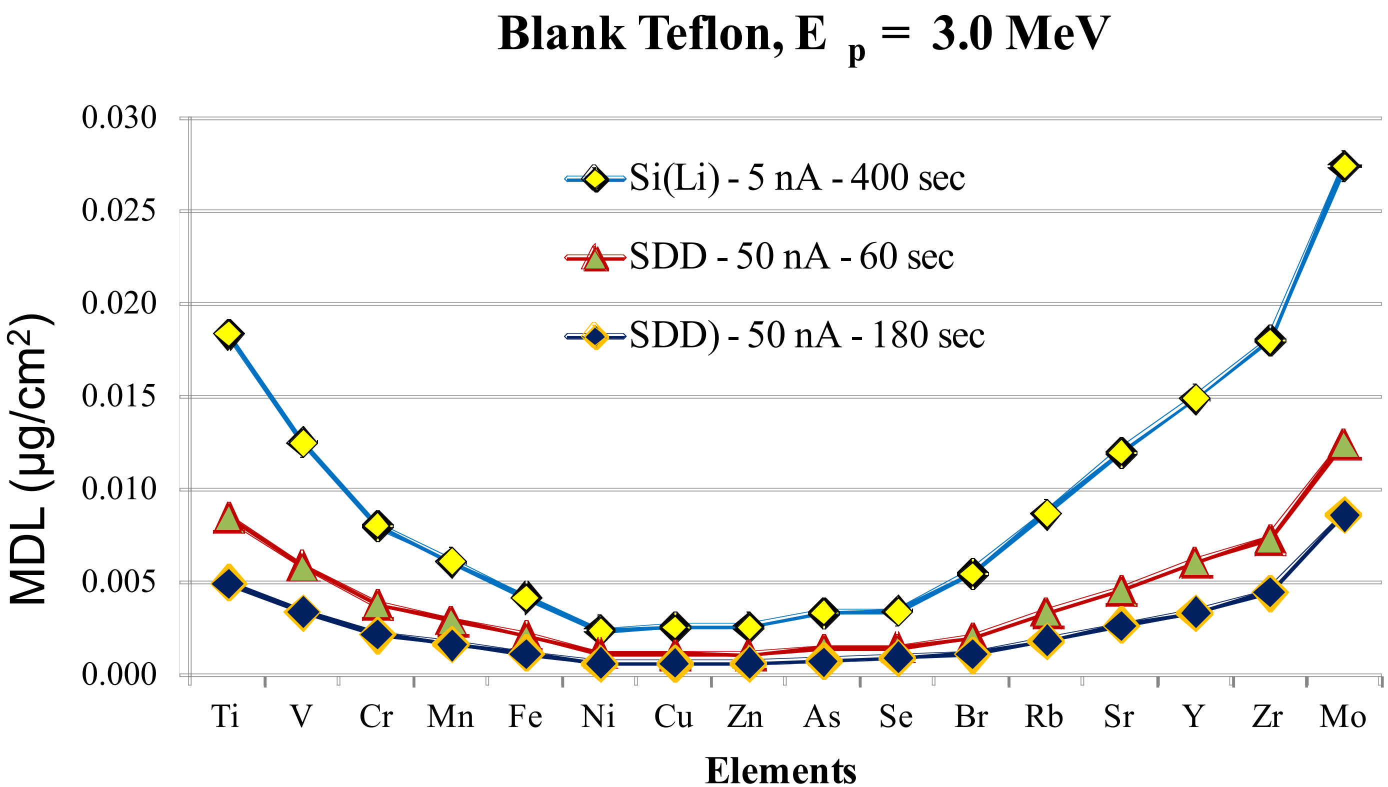
- Small: from a 10 mm^2 SDD to a 30 mm^2 SDD
- Big: a new SDD with same characteristics as the previous one in order to get a double statistics







MDLs “Big”: SDD vs. Si(Li)



PIXE analysis of 1 daily sample: from 400 s to 60 s!

Hundreds of samples may be analyzed in 1 day!

PIXE outstanding technique for the elemental analysis of aerosol samples

Collection substrata