How a small accelerator can be useful for interdisciplinary applications
Part II: cultural heritage studies

Mariaelena Fedi
CHNet (Cultural Heritage Network)
INFN Sezione di Firenze

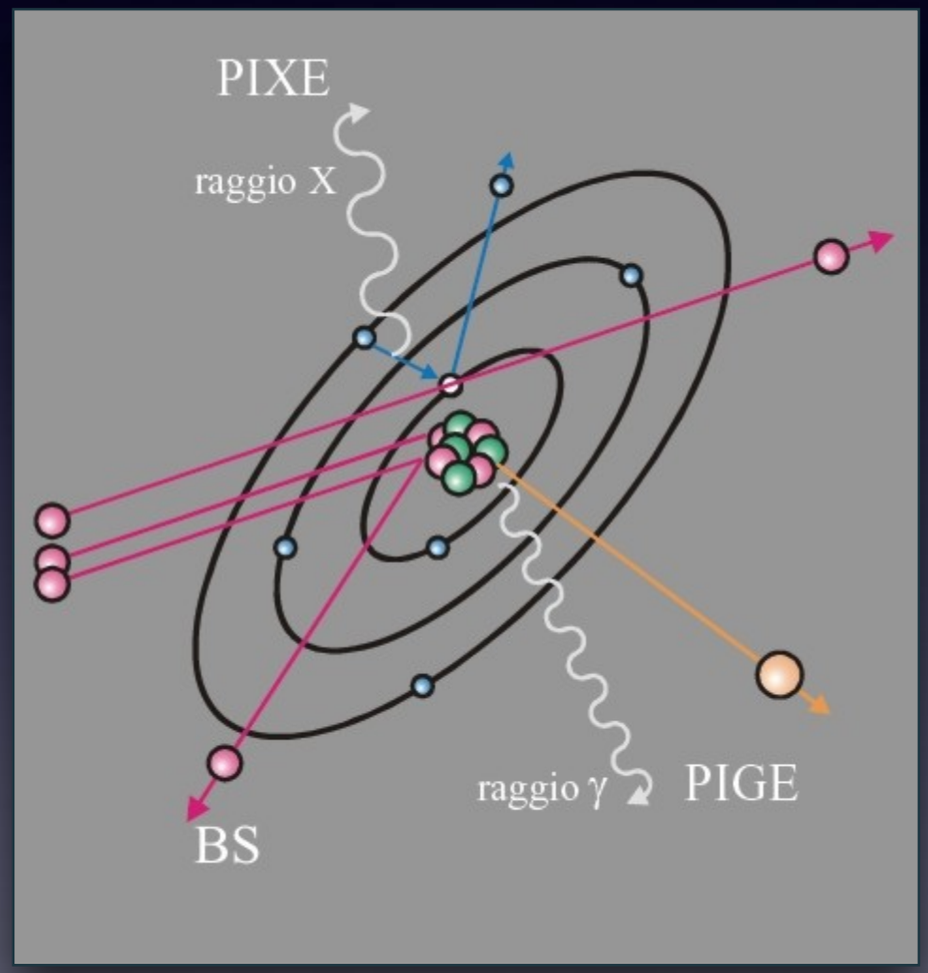
Rewriting nuclear physics textbooks: one more step forward
Pisa July 22th-26th, 2019

An example of small accelerator

Laboratorio di tecniche nucleari per l'Ambiente e i Beni Culturali - Firenze

What you have already learnt

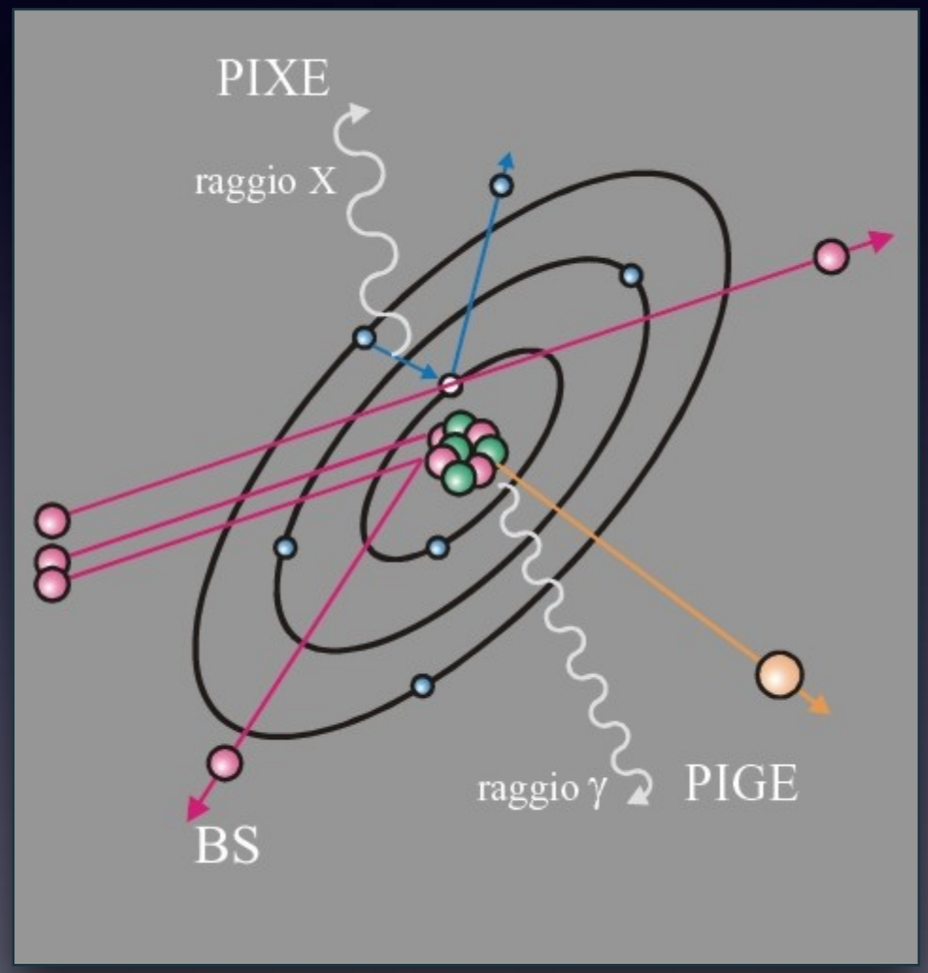
- IBA (Ion Beam Analysis) measurements can allow us to study the composition of a sample
  - PIXE (Particle Induced X-ray Emission) –> information on Z
  - PIGE (Particle Induced γ-ray Emission) –> information on the isotope
  - BS (Back Scattering) –> information on A



What you have already learnt

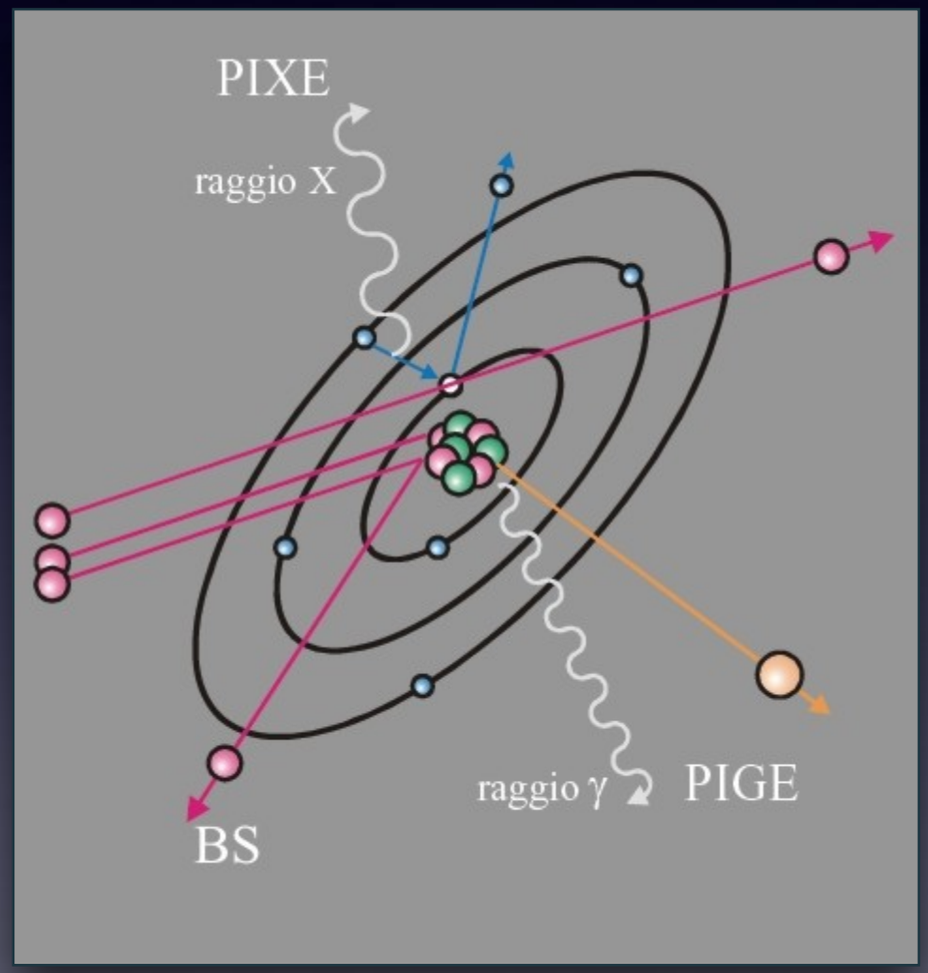
- IBA (Ion Beam Analysis) measurements can allow us to study the composition of a sample
  - PIXE (Particle Induced X-ray Emission) –› information on Z
  - PIGE (Particle Induced γ-ray Emission) –› information on the isotope
  - BS (Back Scattering) –› information on A

But… what about these nuclear techniques and Cultural Heritage?



What you have already learnt

- IBA (Ion Beam Analysis) measurements can allow us to study the composition of a sample
  - PIXE (Particle Induced X-ray Emission) –> information on Z
  - PIGE (Particle Induced γ-ray Emission) –> information on the isotope
  - BS (Back Scattering) –> information on A



But… what about these nuclear techniques and Cultural Heritage?

or… more in general… why mixing nuclear techniques, material analysis and CH?

The discipline of Archaeometry

The use of science and modern technology in archaeology
to examine and interpret archaeological remains

(Collins dictionary def.)

The discipline of Archaeometry

The use of science and modern technology in archaeology to examine and interpret archaeological remains

(Collins dictionary def.)

- Materials studies

The discipline of Archaeometry

The use of science and modern technology in archaeology to examine and interpret archaeological remains

(Collins dictionary def.)

- Materials studies
  - to study the preservation state of artworks and findings
  - to support restoration (to find the most appropriate materials which are compatible with the original ones)
  - to study the artistic techniques
  - to date (although in an indirect way)

The discipline of Archaeometry

The use of science and modern technology in archaeology to examine and interpret archaeological remains

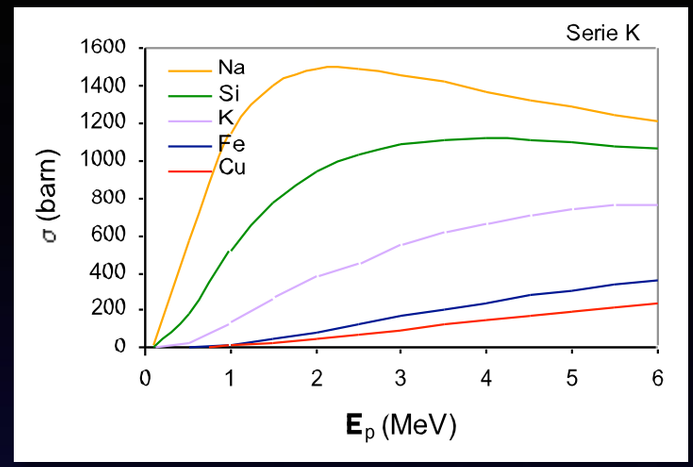
(Collins dictionary def.)

- Materials studies
  - to study the preservation state of artworks and findings
  - to support restoration (to find the most appropriate materials which are compatible with the original ones)
  - to study the artistic techniques
  - to date (although in an indirect way)
- Dating

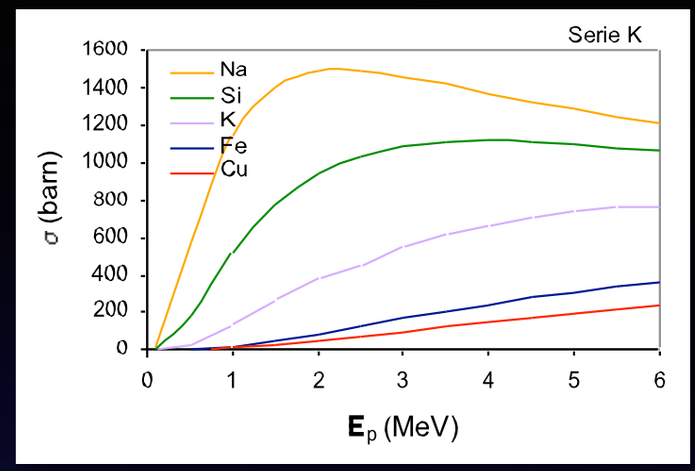
PIXE: the most used IBA technique

PIXE: the most used IBA technique

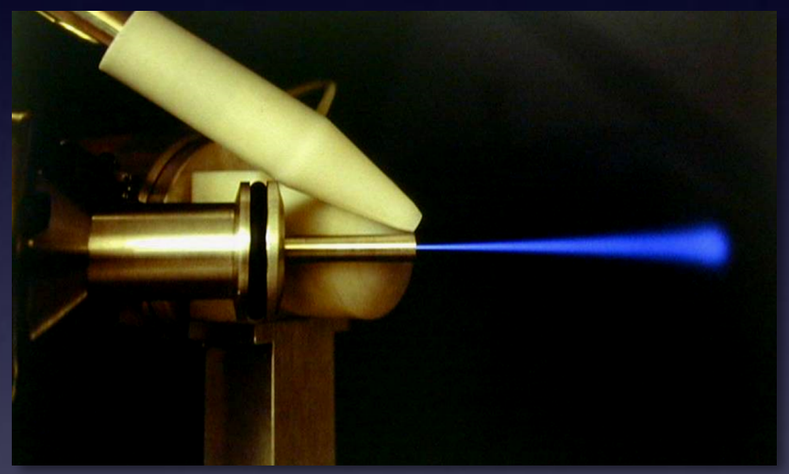
- X-ray production cross sections are pretty high –> we can keep the incident ions current as low as possible, still allowing for fast acquiring times



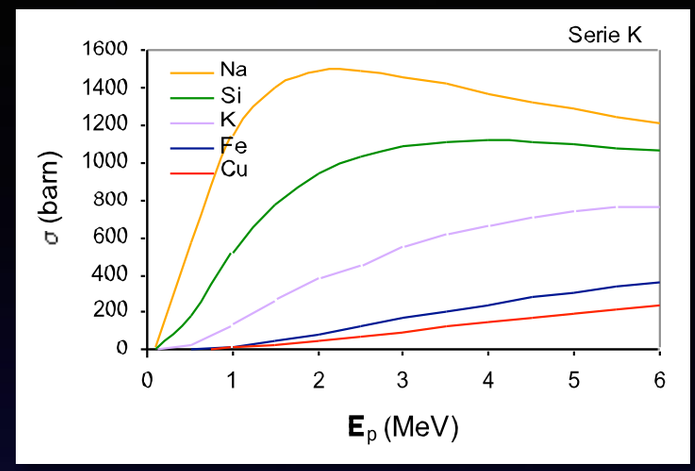
PIXE: the most used IBA technique



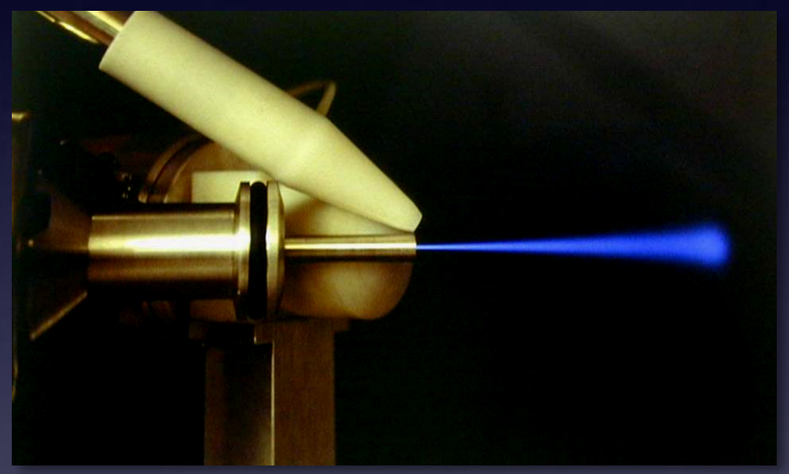
- X-ray production cross sections are pretty high –> we can keep the incident ions current as low as possible, still allowing for fast acquiring times
- Incident ions can be extracted into atmosphere –> no sampling is needed



PIXE: the most used IBA technique

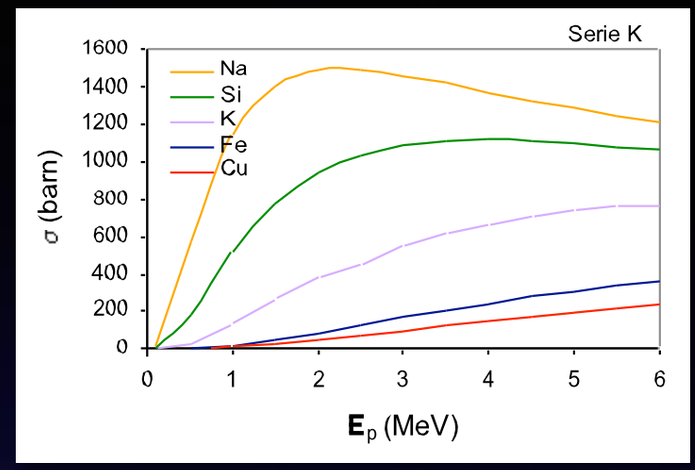


- X-ray production cross sections are pretty high –> we can keep the incident ions current as low as possible, still allowing for fast acquiring times
- Incident ions can be extracted into atmosphere –> no sampling is needed

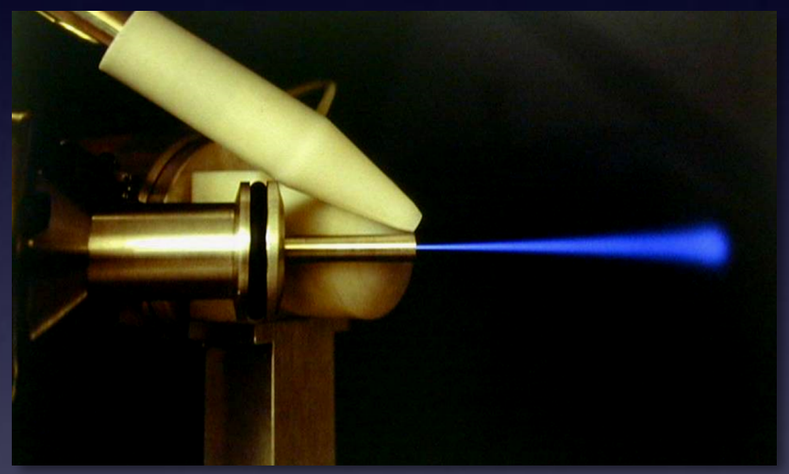


- PIXE is multielemental

PIXE: the most used IBA technique



- X-ray production cross sections are pretty high –> we can keep the incident ions current as low as possible, still allowing for fast acquiring times
- Incident ions can be extracted into atmosphere –> no sampling is needed



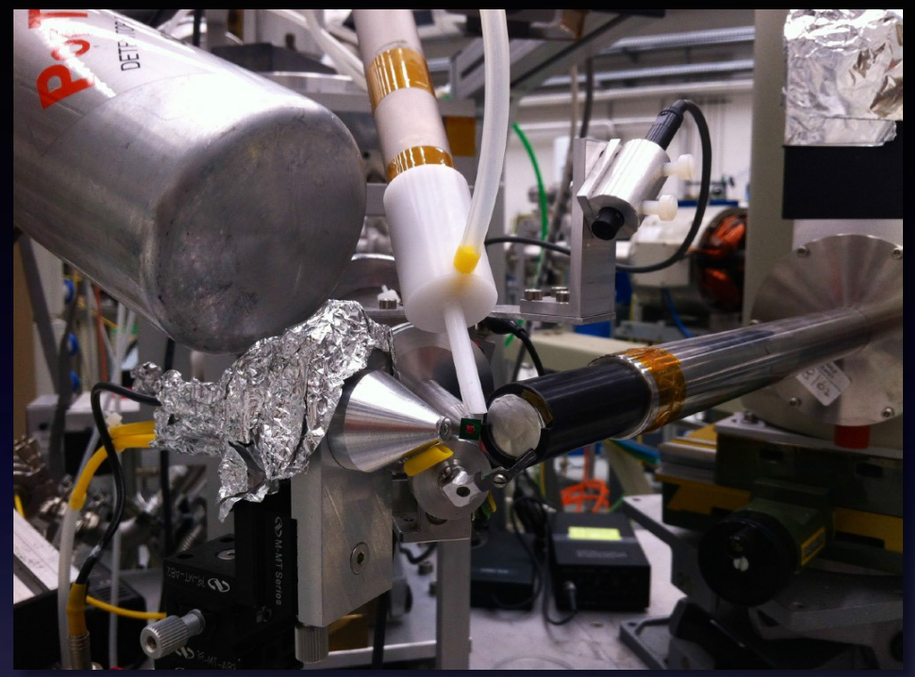
- PIXE is multielemental

Thus, PIXE measurements (IBA in general) are:
non invasive and non destructive
quick

PIXE: some drawbacks

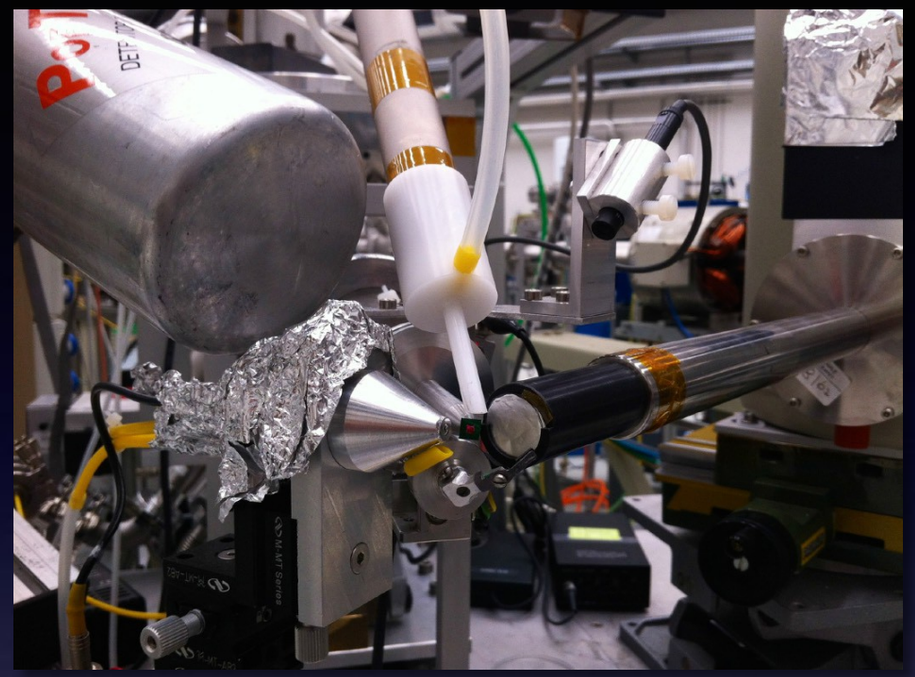
PIXE: some drawbacks

- In external beam set-ups, X-rays with energies below 1 keV are not collected –› no information on elements with Z‹11



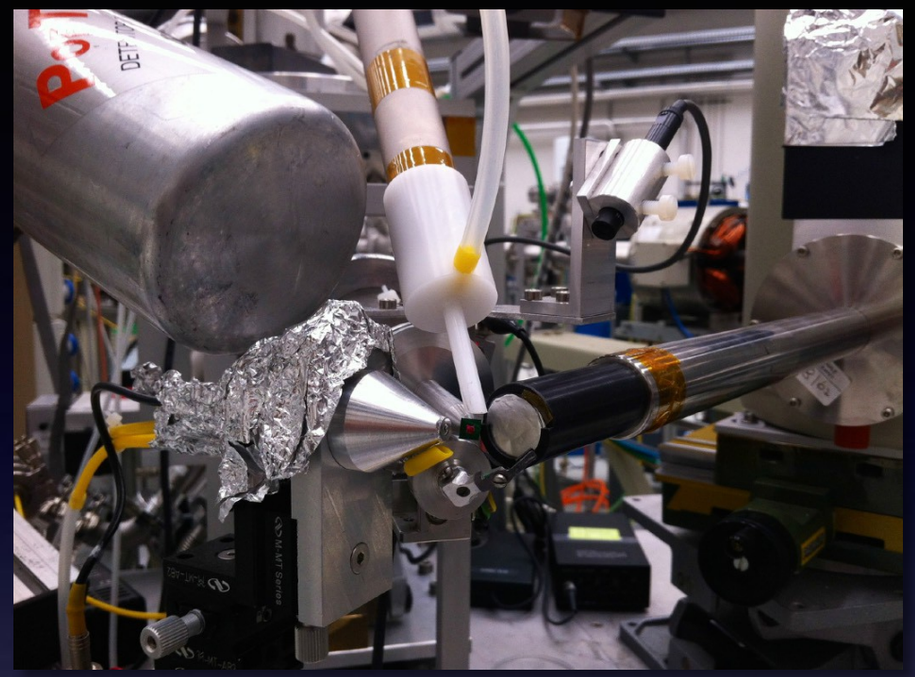
PIXE: some drawbacks

- In external beam set-ups, X-rays with energies below 1 keV are not collected –› no information on elements with Z‹11
- Emitted X-rays depend on the allowed atomic transitions –› information on the element and not on possible molecular structure and compounds

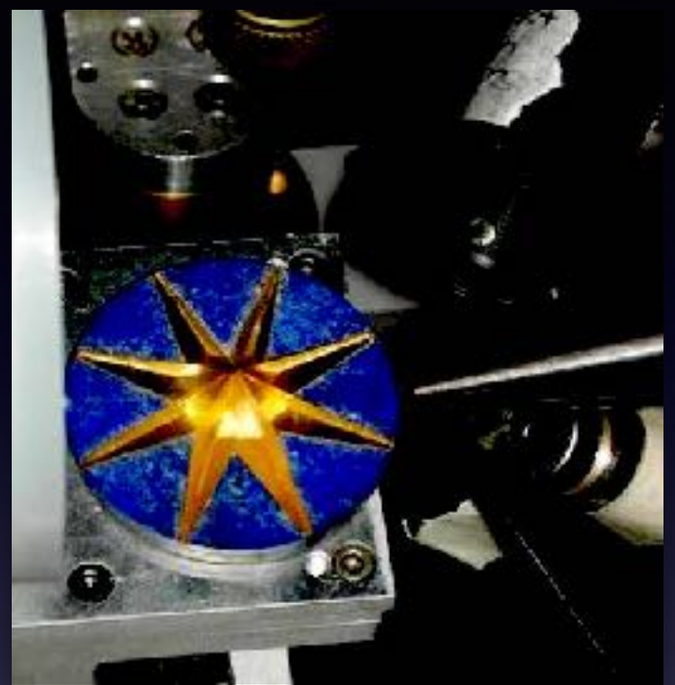


PIXE: some drawbacks

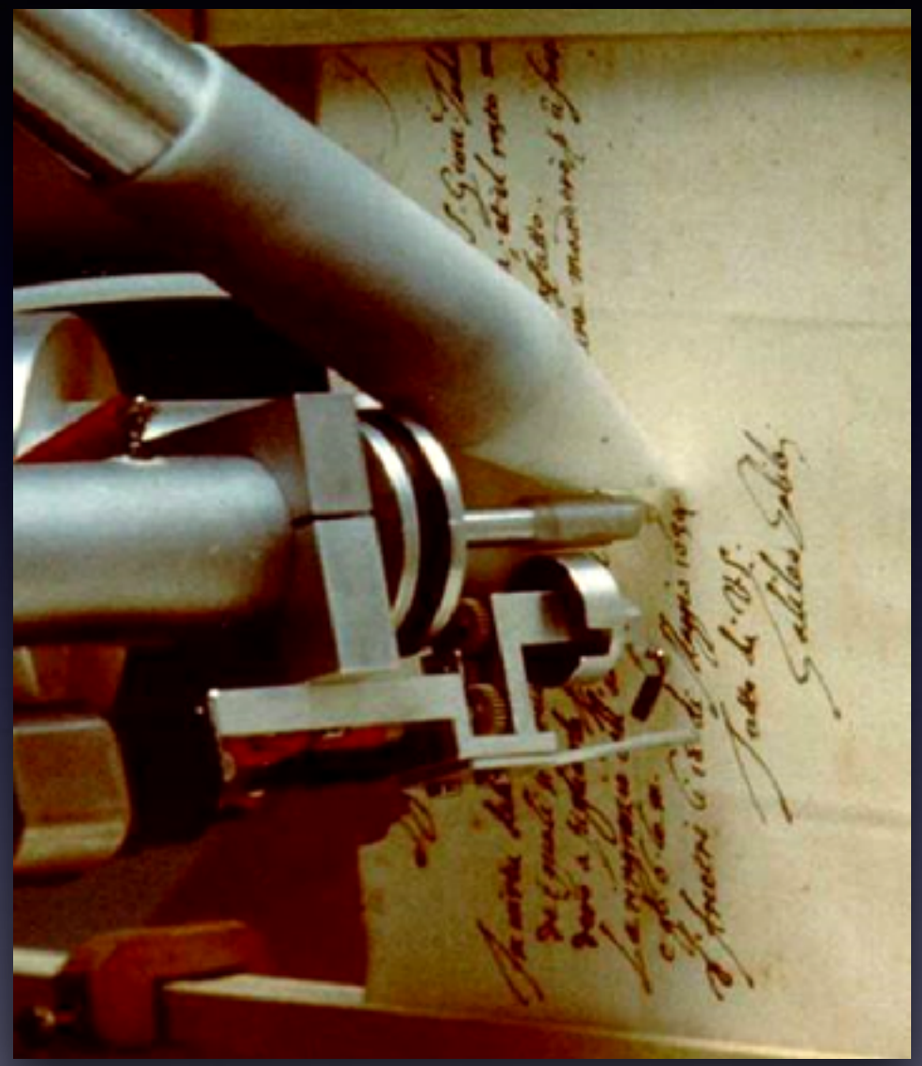
- In external beam set-ups, X-rays with energies below 1 keV are not collected –› no information on elements with Z‹11
- Emitted X-rays depend on the allowed atomic transitions –› information on the element and not on possible molecular structure and compounds
- In-situ measurements are not typically possible



Some examples of artworks to be analyzed



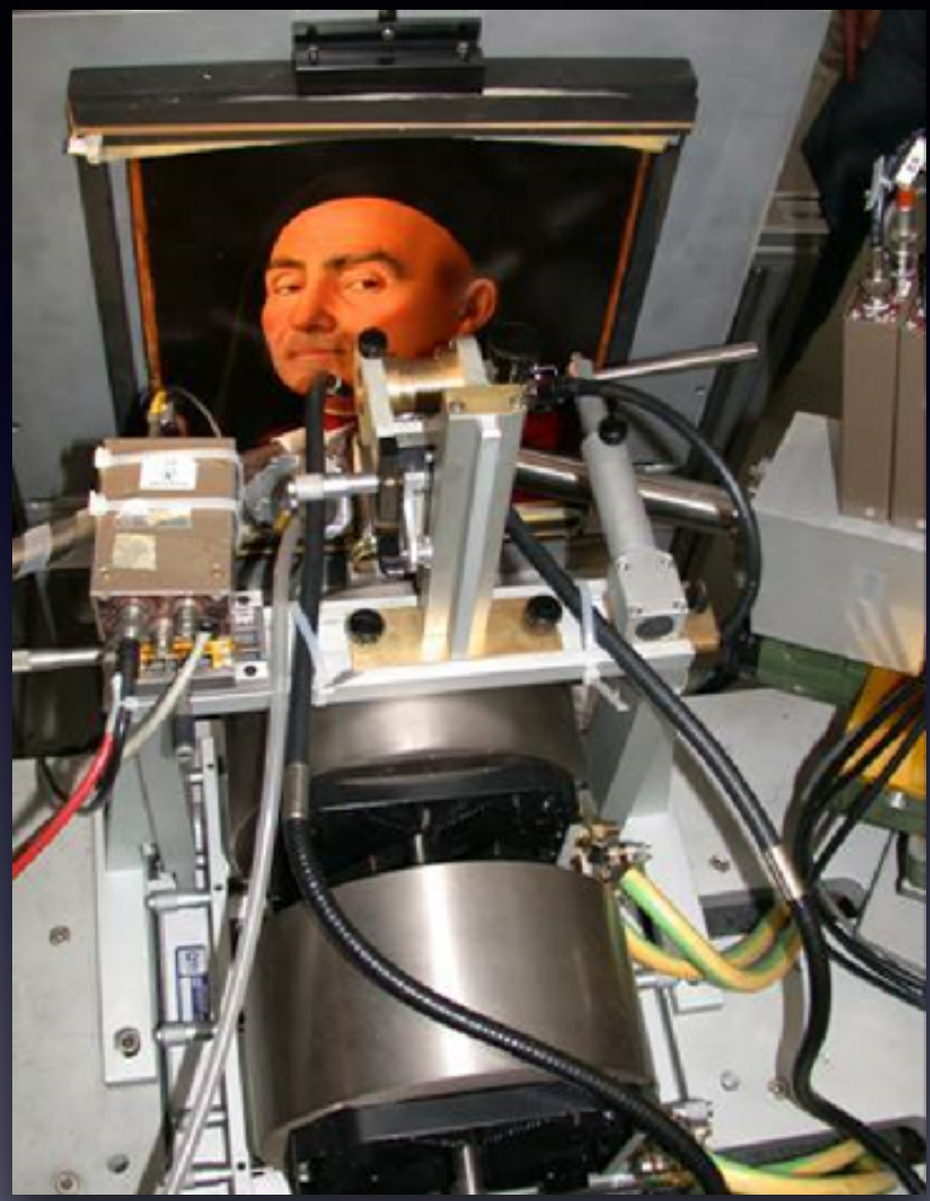
"Collezione Medicea di Pietre Ornamentali", Museum of Natural History, University of Florence



Galileo Galilei, hand-written manuscript, Biblioteca Nazionale, Florence

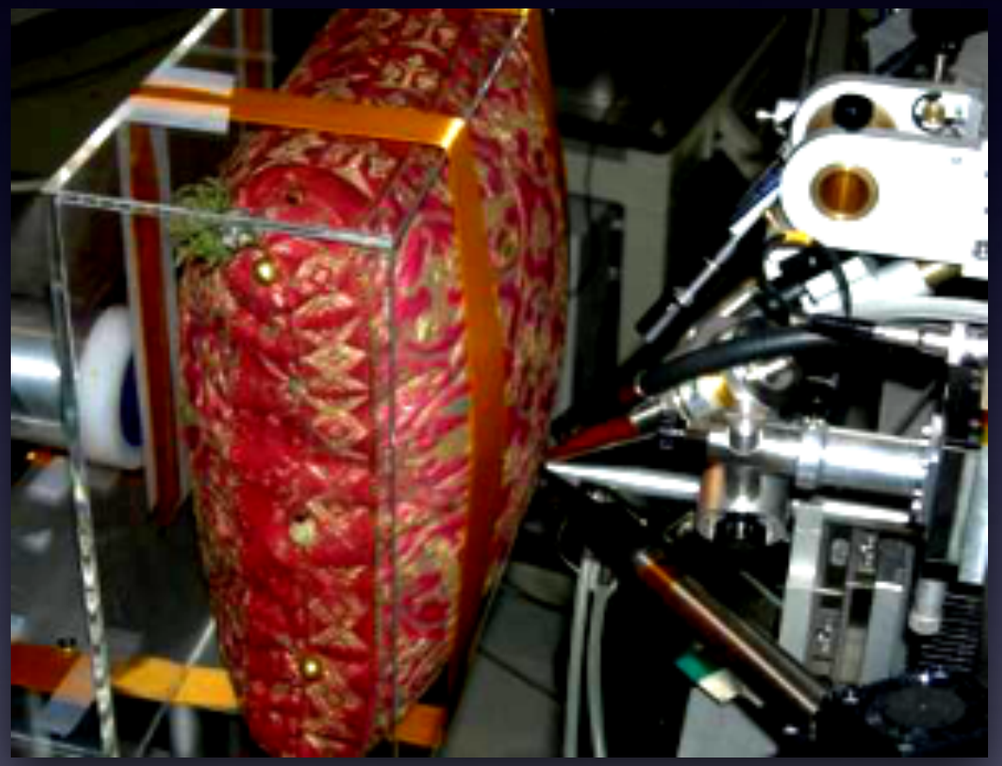
Some examples of artworks to be analyzed

Leonardo da Vinci, Madonna dei Fusi
(private collection)

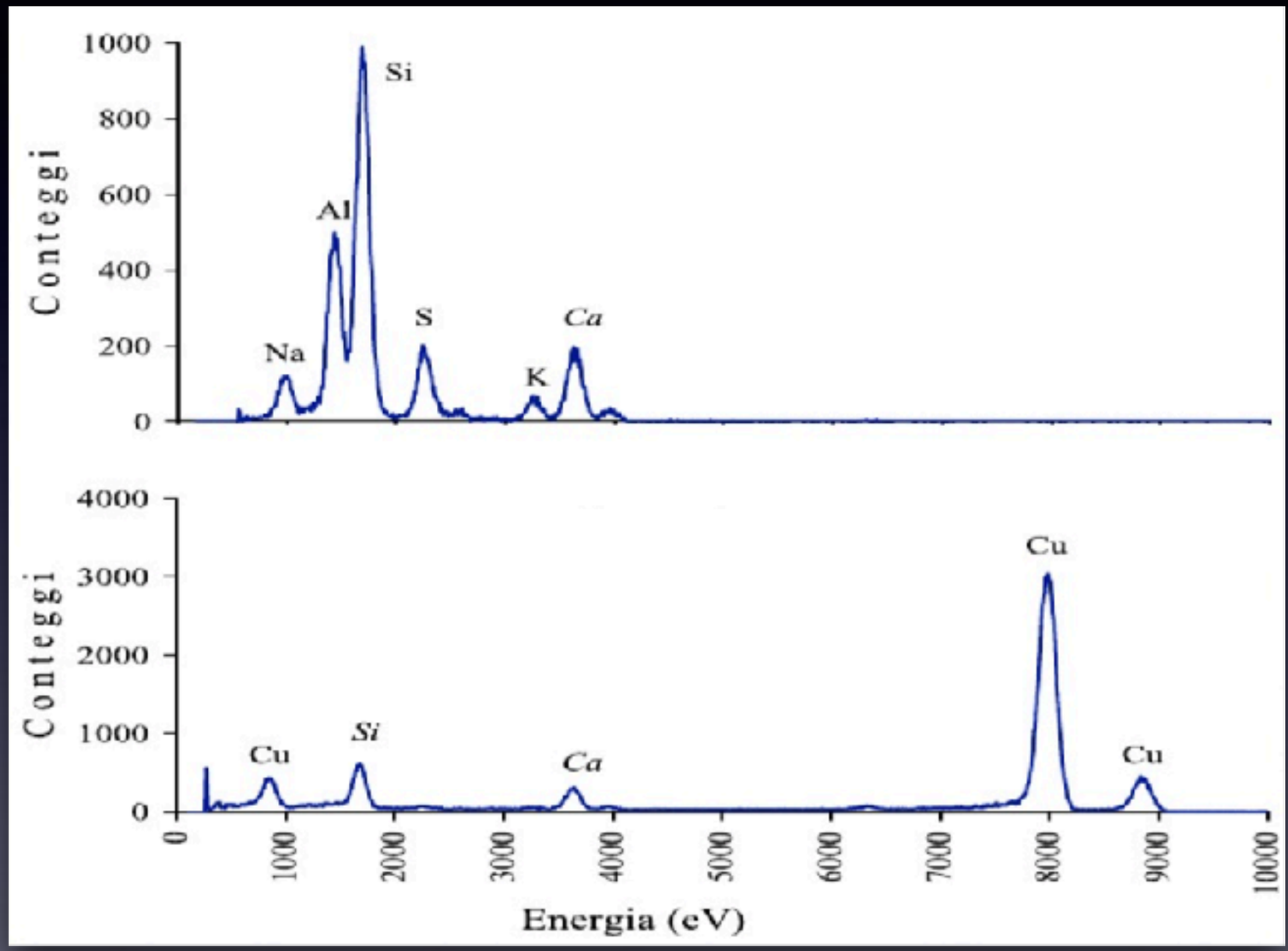


Antonello da Messina, Ritratto
Trivulzio, Palazzo Madama, Turin

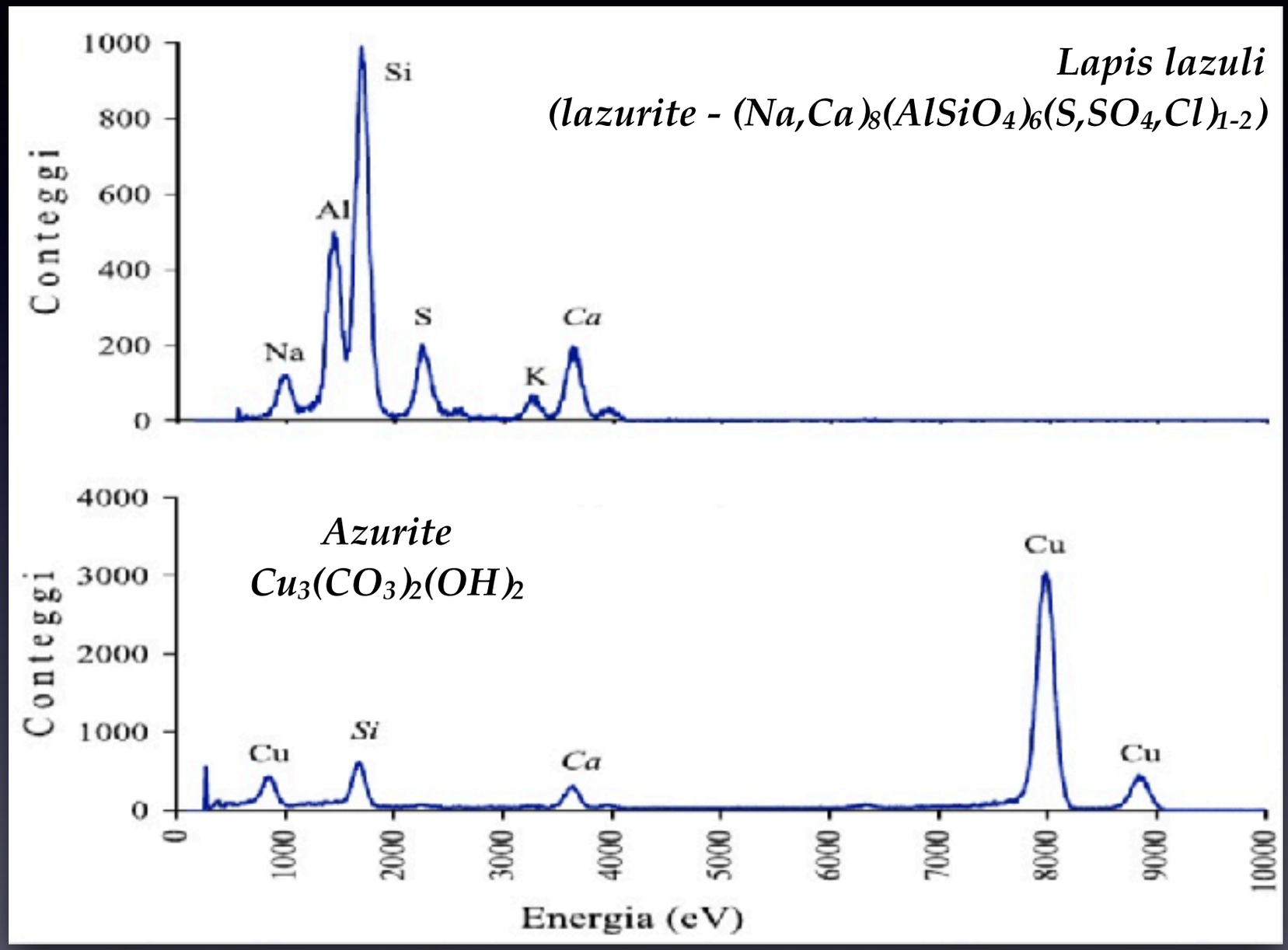
Some examples of artworks to be analyzed



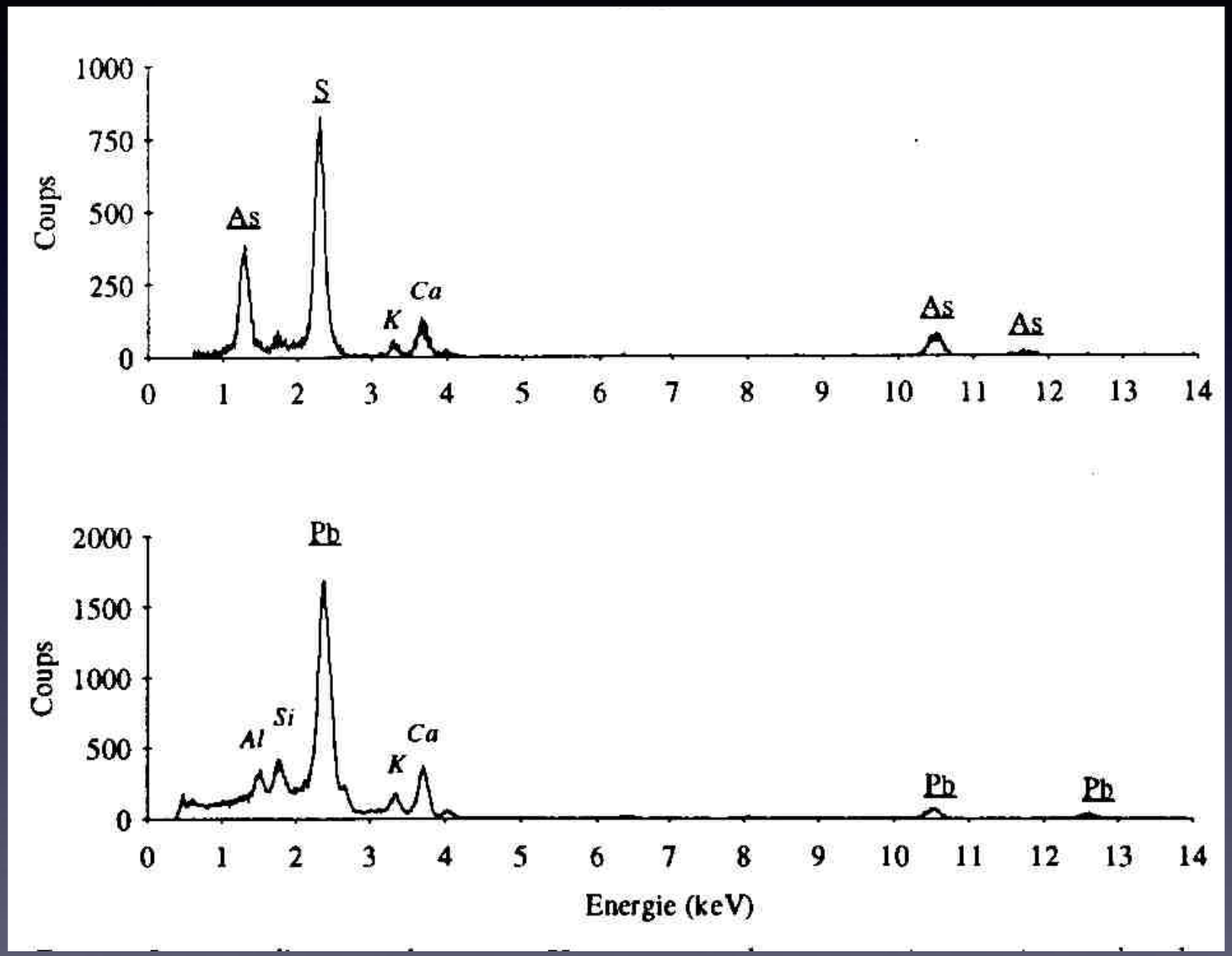
Spectra from pigments



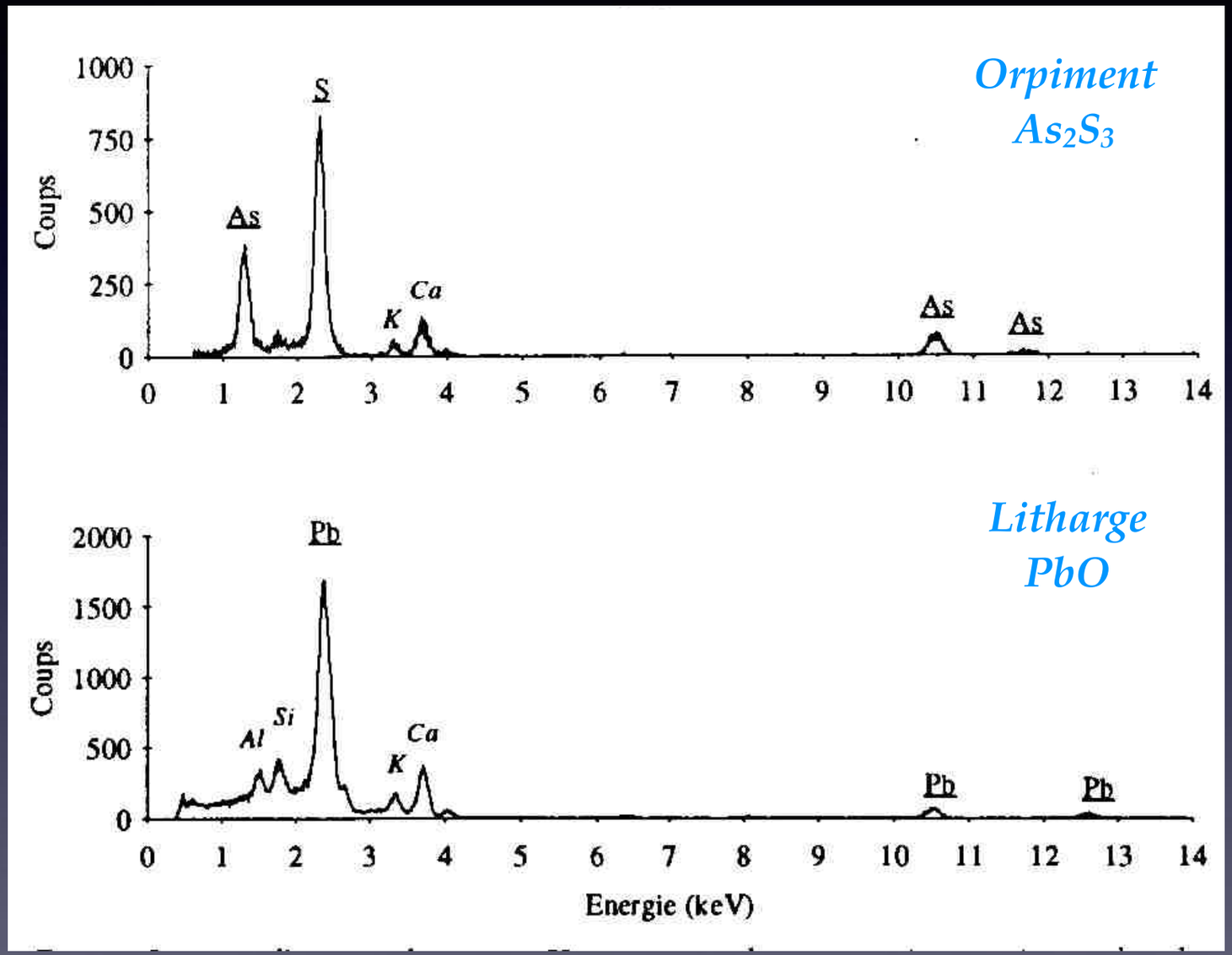
Spectra from pigments



Spectra from pigments



Spectra from pigments



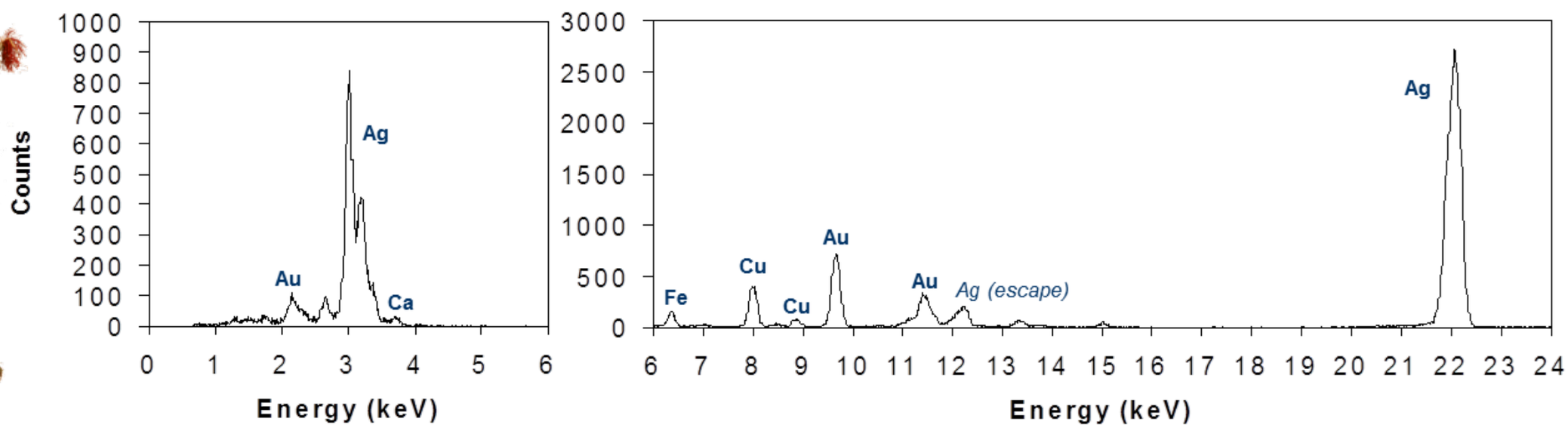
Scanning PIXE –› imaging

- We can exploit very focused ion beams through strong focusing by magnetic lenses –› about 10 μm FWHM on target (as order of magnitude)
  - Scan of the target in front of the beam
    or
  - Scan of the beam in front of the target (in this case, the scanning area is limited to the exit beam window dimensions)

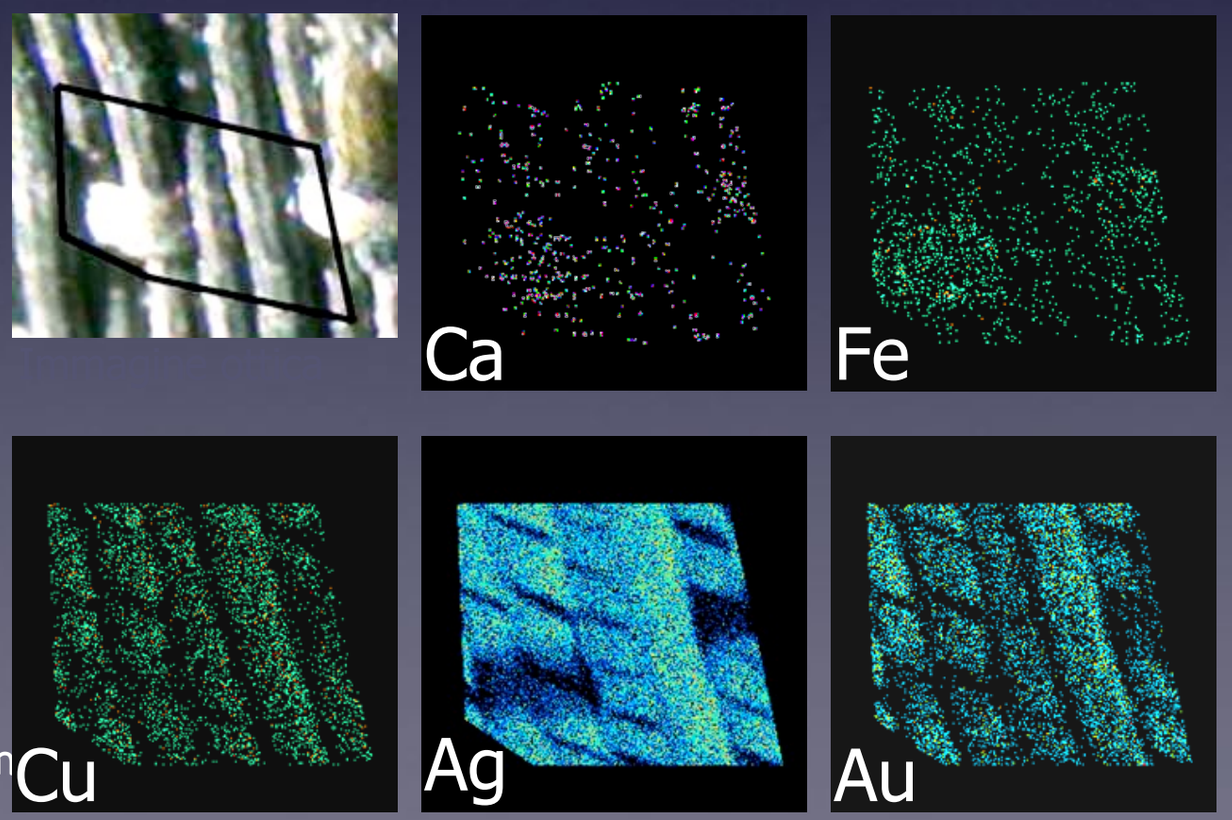
✓ *For each of the analyzed point we can acquire both the X-rays energies and the point coordinates*

✓ ***(x_i, y_i, E_z) to represent how the elements are distributed over the scanned area: elemental maps***

Golden embroideries



1 mm

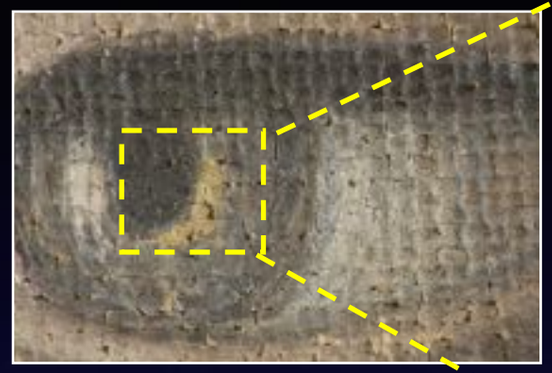


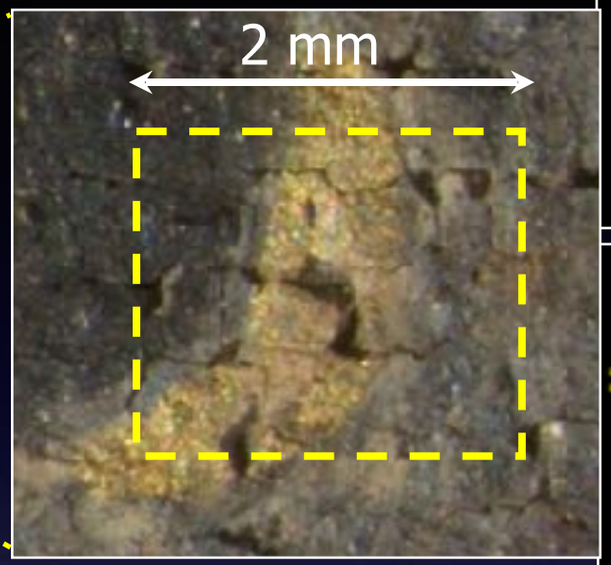
“Madonna con il Bambino” by Mantegna

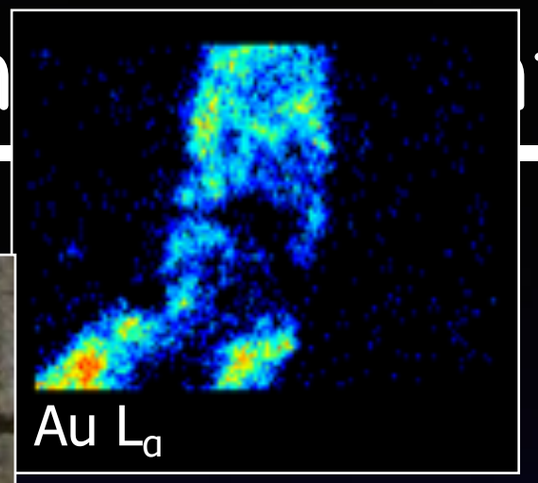
Accademia Carrara, Bergamo

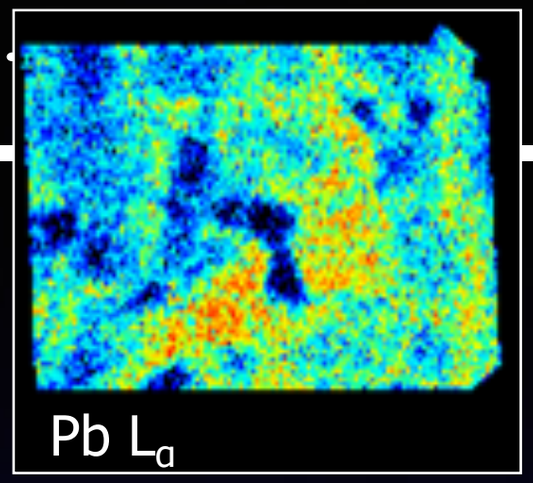
“Madonna con il Bambino” by Mantegnaino

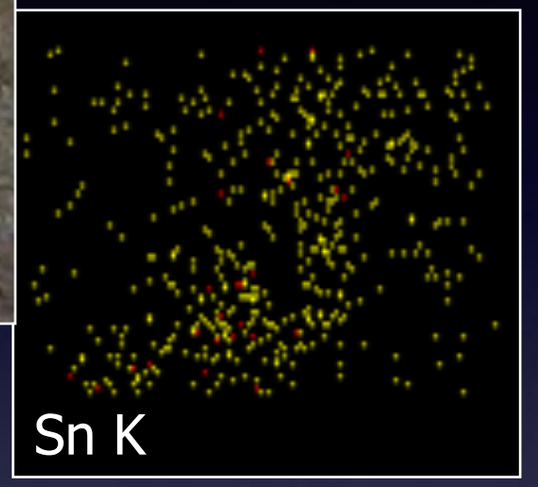
“Madonna con il Bambin

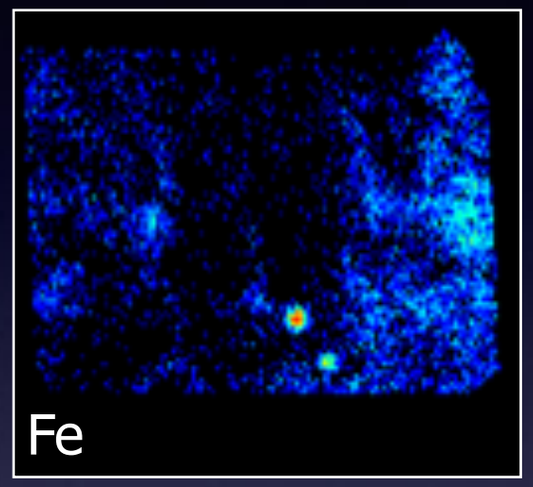




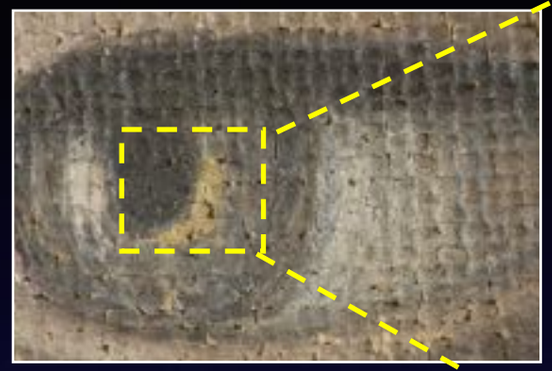


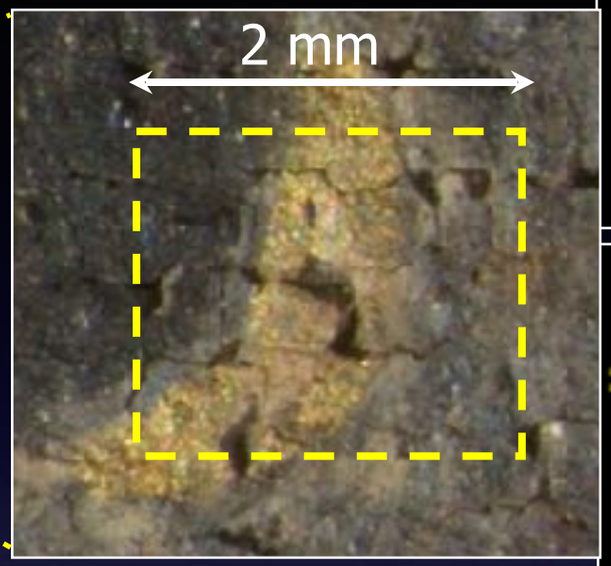


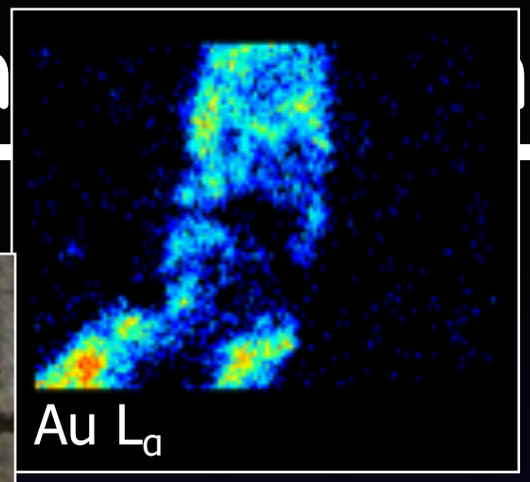


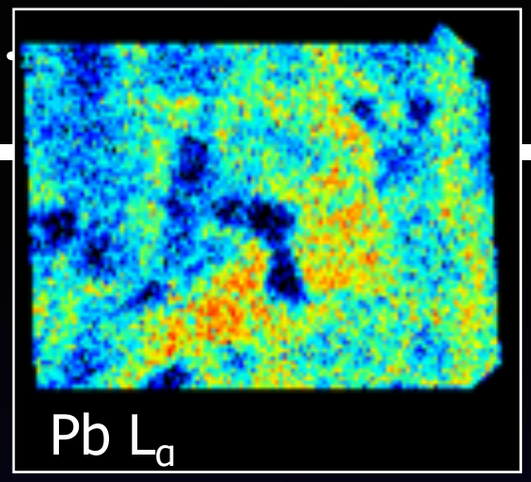


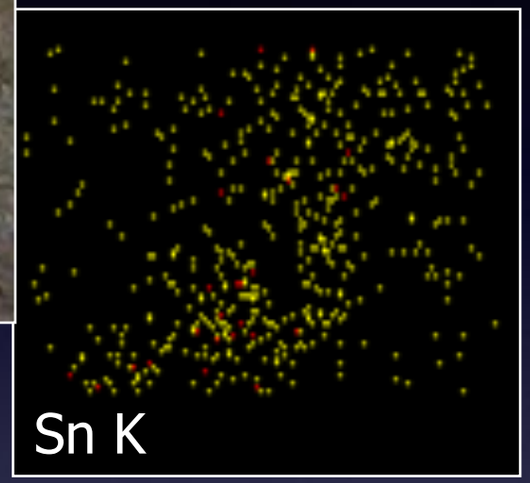
“Madonna con il Bambin

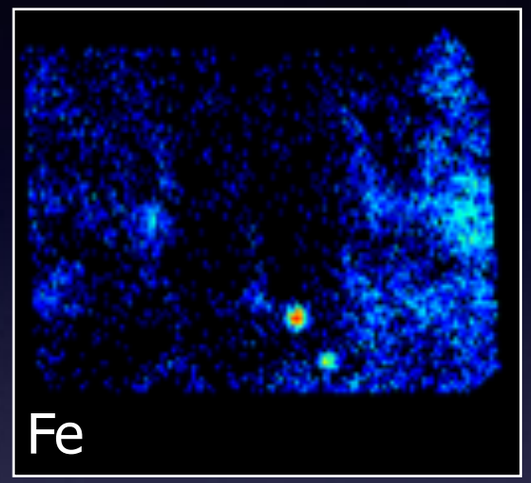


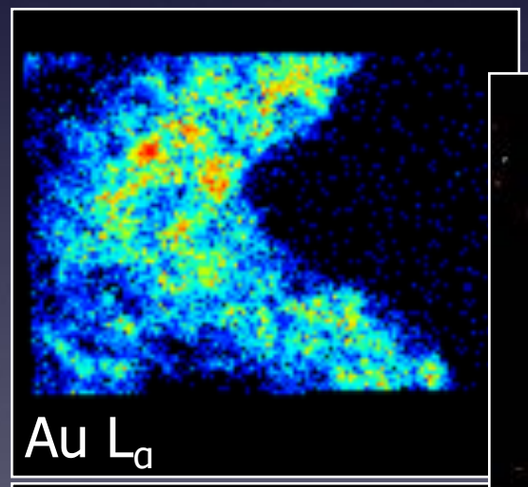


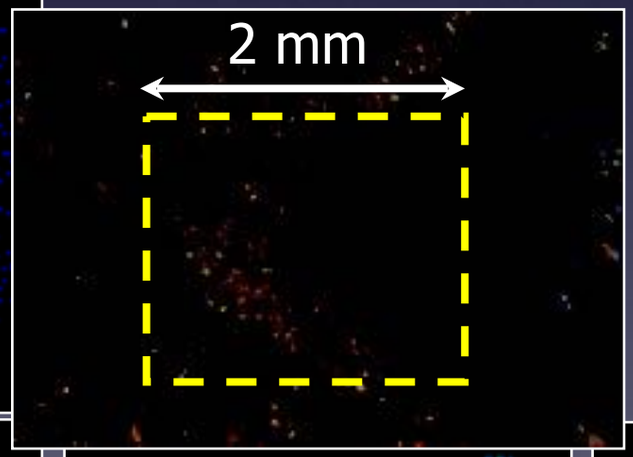


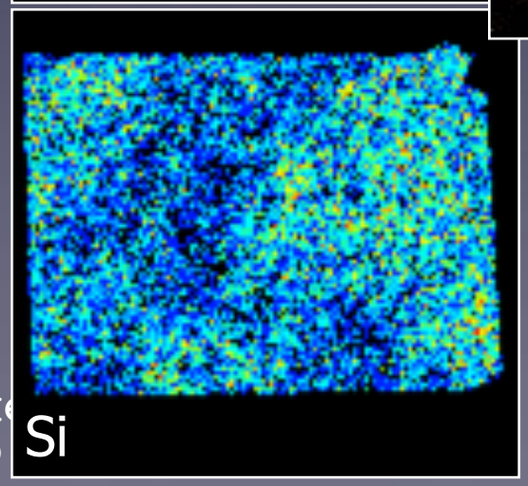


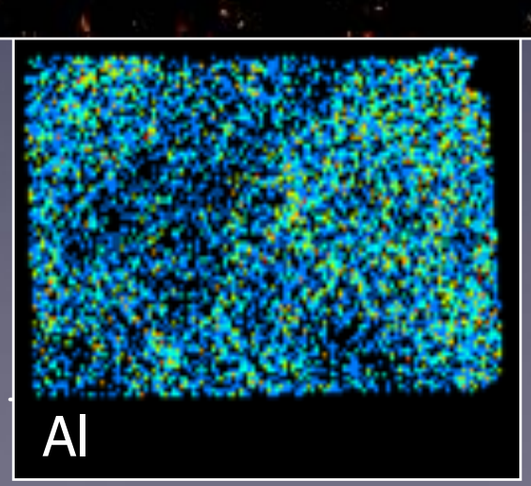


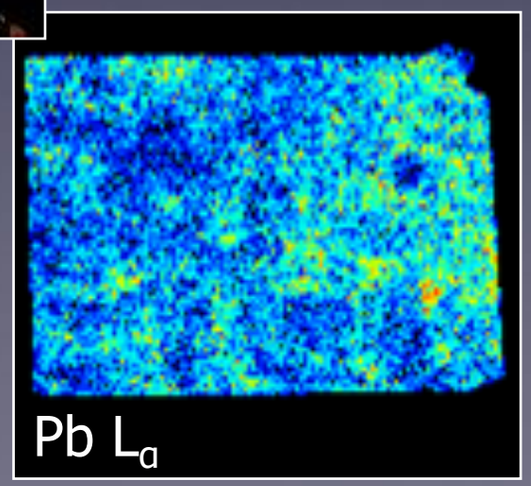












Going into the deep: differential PIXE

Going into the deep: differential PIXE

- Maximum analyzed depths depend on:
  - Incident ions energy
  - Sample composition
  - Z of the analyzed element (i.e. collected x-ray energy)

Going into the deep: differential PIXE

- Maximum analyzed depths depend on:
  - Incident ions energy
  - Sample composition
  - Z of the analyzed element (i.e. collected x-ray energy)

Analyzed thicknesses up to about 100 μm in light matrix for medium-high energy x-rays

Going into the deep: differential PIXE

- Maximum analyzed depths depend on:
  - Incident ions energy
  - Sample composition
  - Z of the analyzed element (i.e. collected x-ray energy)

Analyzed thicknesses up to about 100 μm in light matrix for medium-high energy x-rays

The information we get is integrated over the whole analyzed thickness

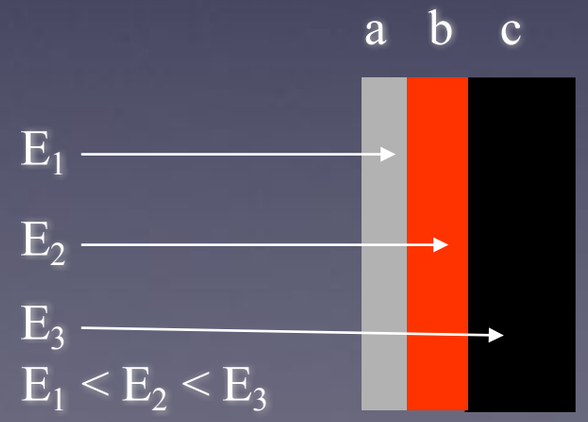
Going into the deep: differential PIXE

- Maximum analyzed depths depend on:
  - Incident ions energy
  - Sample composition
  - Z of the analyzed element (i.e. collected x-ray energy)

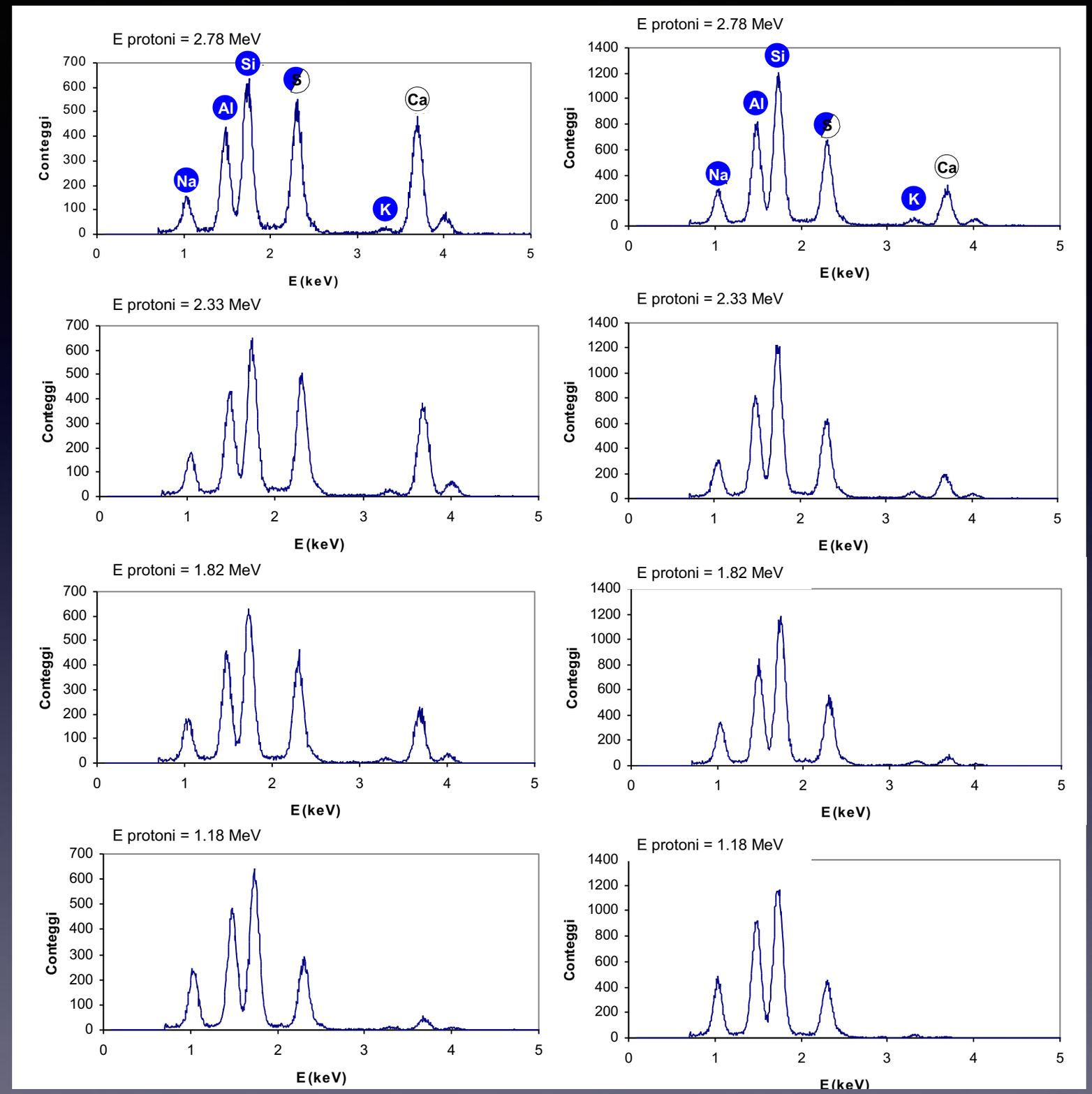
Analyzed thicknesses up to about 100 μm in light matrix for medium-high energy x-rays

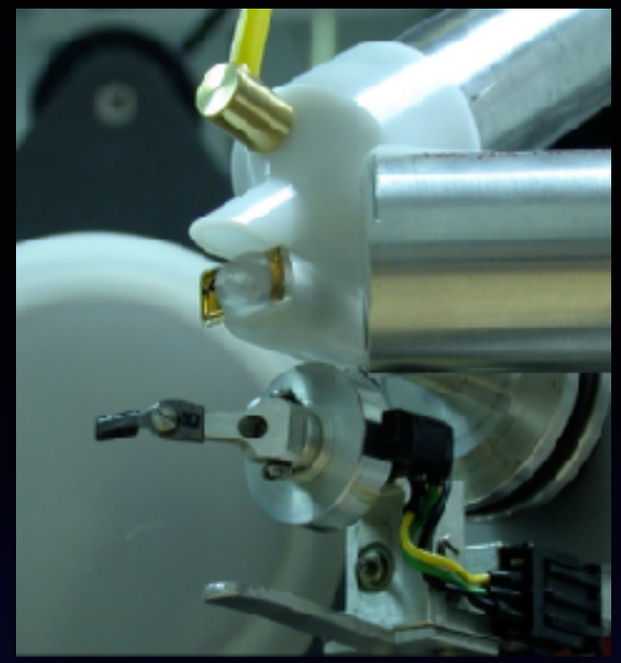
The information we get is integrated over the whole analyzed thickness

- If we change the incident proton energy:
  - Protons range inside the target changes
  - X-ray production cross section changes

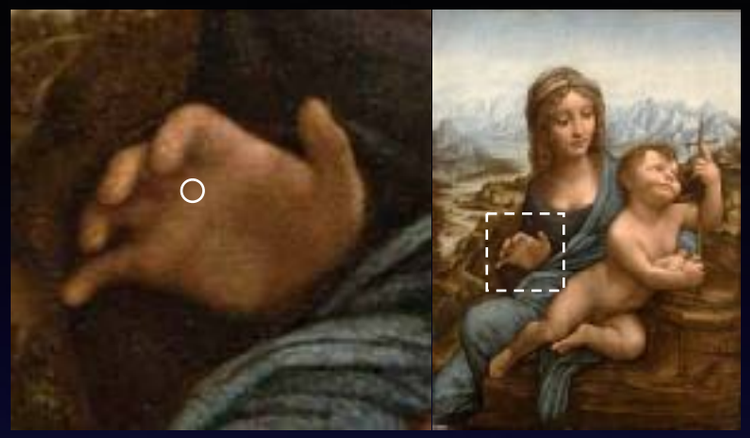


Lapis lazuli on chalk (wooden table)





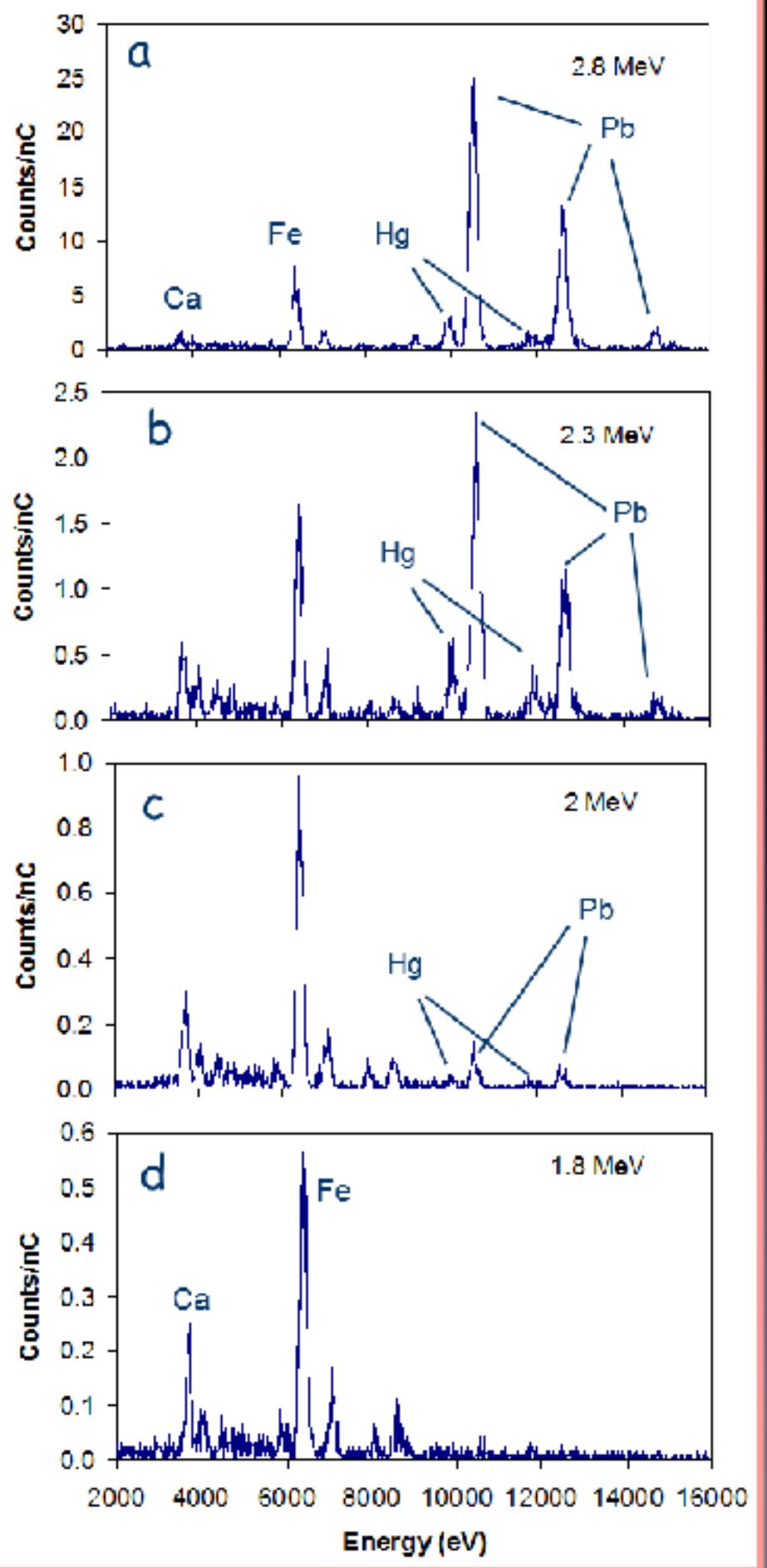
An example from Leonardo



- Hg –> cinnabar (HgS)
- Pb –> white lead ($2PbCO_3 \cdot Pb(OH)_2$)

  In b) and c), **Hg/Pb** increases with respect what happens in a): the preparation layer underneath is not hit by incident protons anymore –> no contribution by this layer to the total Pb counts.
  In d), neither Hg or Pb are evident: protons stop in the superficial varnish layer

- Fe, Ca –> varnish

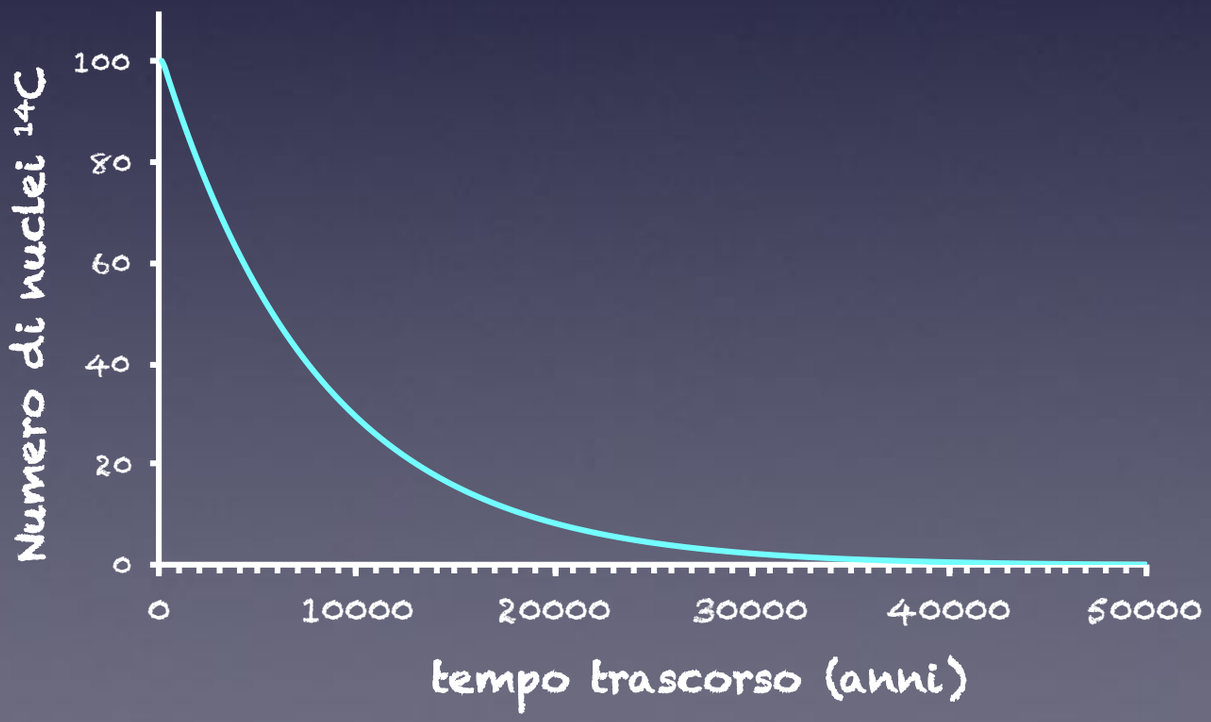


A small accelerator and the dating issue

- ^{14}C can be used as a natural chronometer to date organic materials, or - in more generally speaking - those systems that have been directly (or indirectly) in equilibrium with atmosphere, acquiring ^{14}C from it, and that have ceased that equilibrium condition in a well defined moment

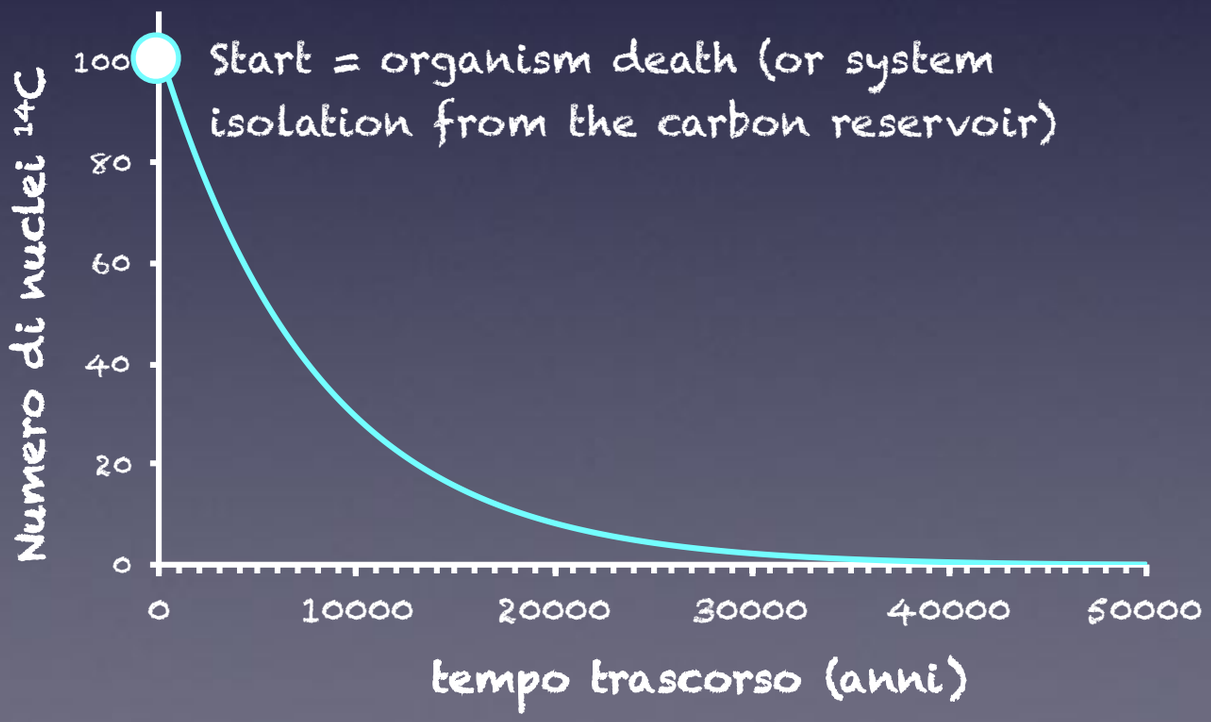
A small accelerator and the dating issue

- ^{14}C can be used as a natural chronometer to date organic materials, or - in more generally speaking - those systems that have been directly (or indirectly) in equilibrium with atmosphere, acquiring ^{14}C from it, and that have ceased that equilibrium condition in a well defined moment



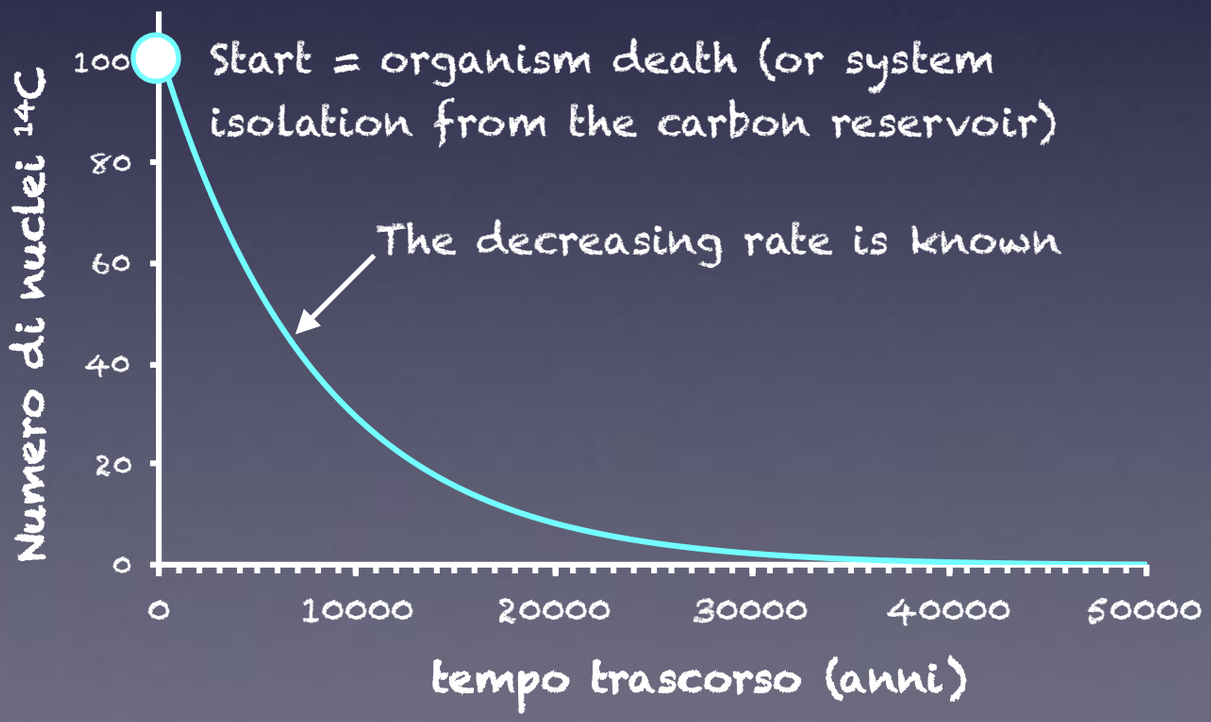
A small accelerator and the dating issue

- ^{14}C can be used as a natural chronometer to date organic materials, or - in more generally speaking - those systems that have been directly (or indirectly) in equilibrium with atmosphere, acquiring ^{14}C from it, and that have ceased that equilibrium condition in a well defined moment



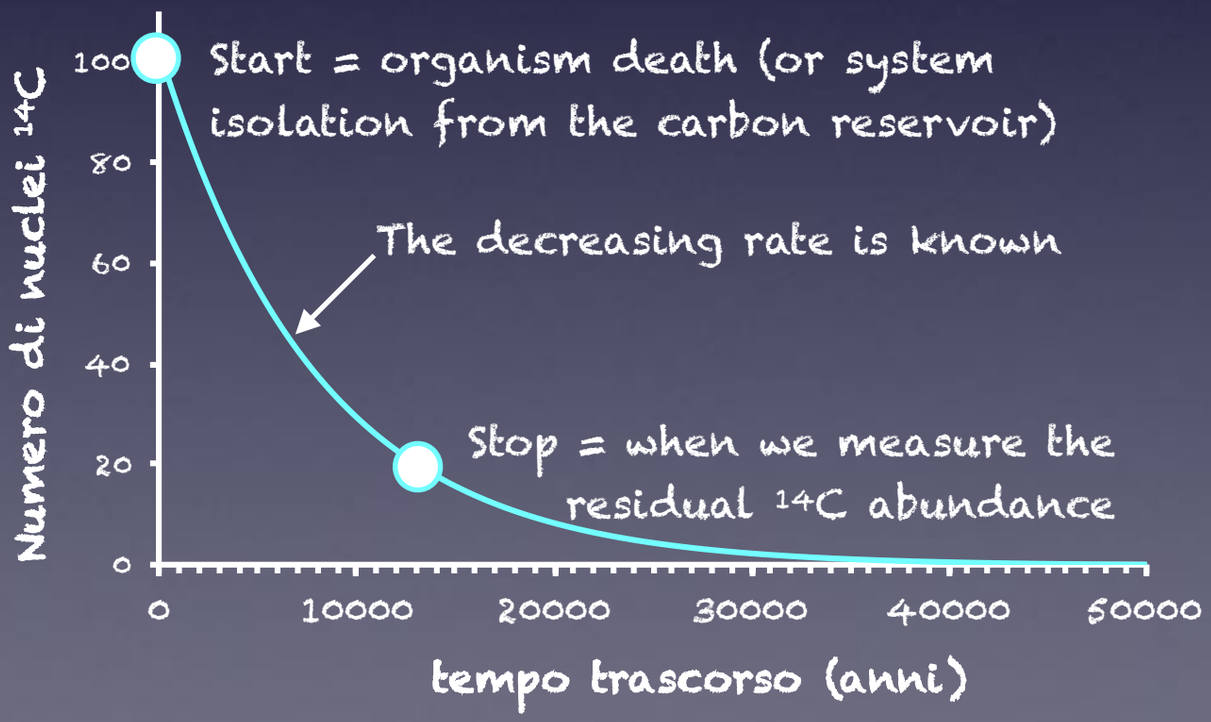
A small accelerator and the dating issue

- ^{14}C can be used as a natural chronometer to date organic materials, or - in more generally speaking - those systems that have been directly (or indirectly) in equilibrium with atmosphere, acquiring ^{14}C from it, and that have ceased that equilibrium condition in a well defined moment



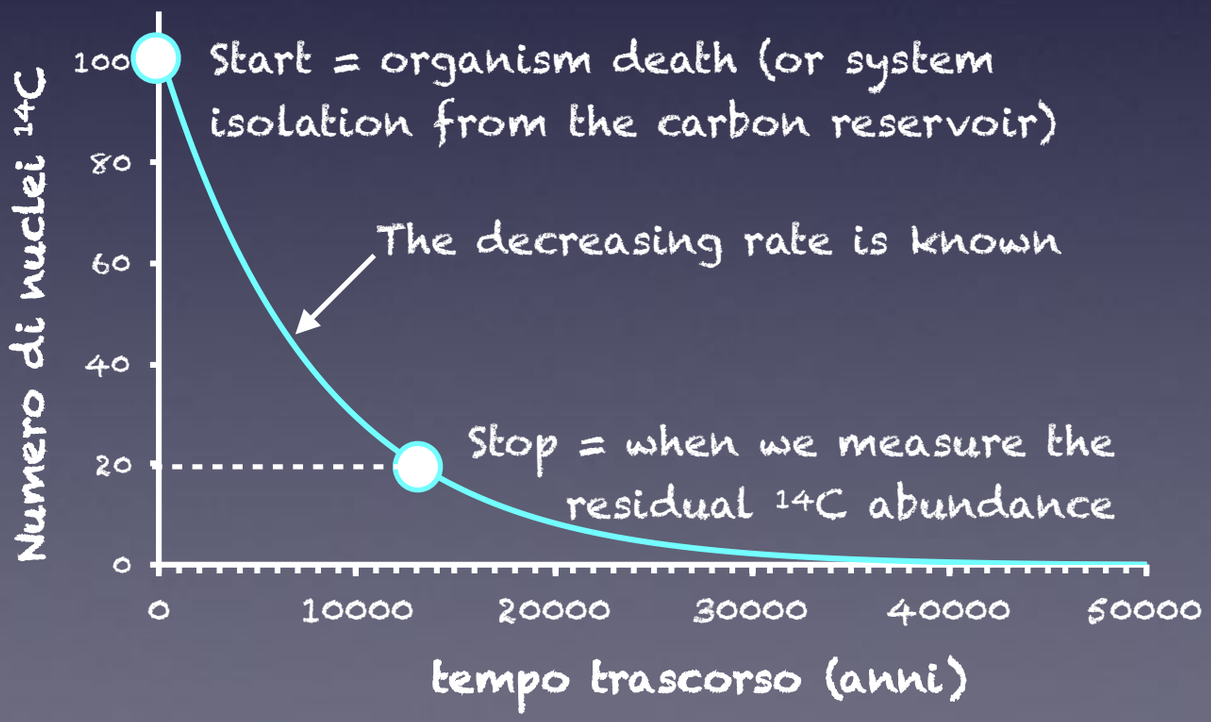
A small accelerator and the dating issue

- ^{14}C can be used as a natural chronometer to date organic materials, or - in more generally speaking - those systems that have been directly (or indirectly) in equilibrium with atmosphere, acquiring ^{14}C from it, and that have ceased that equilibrium condition in a well defined moment



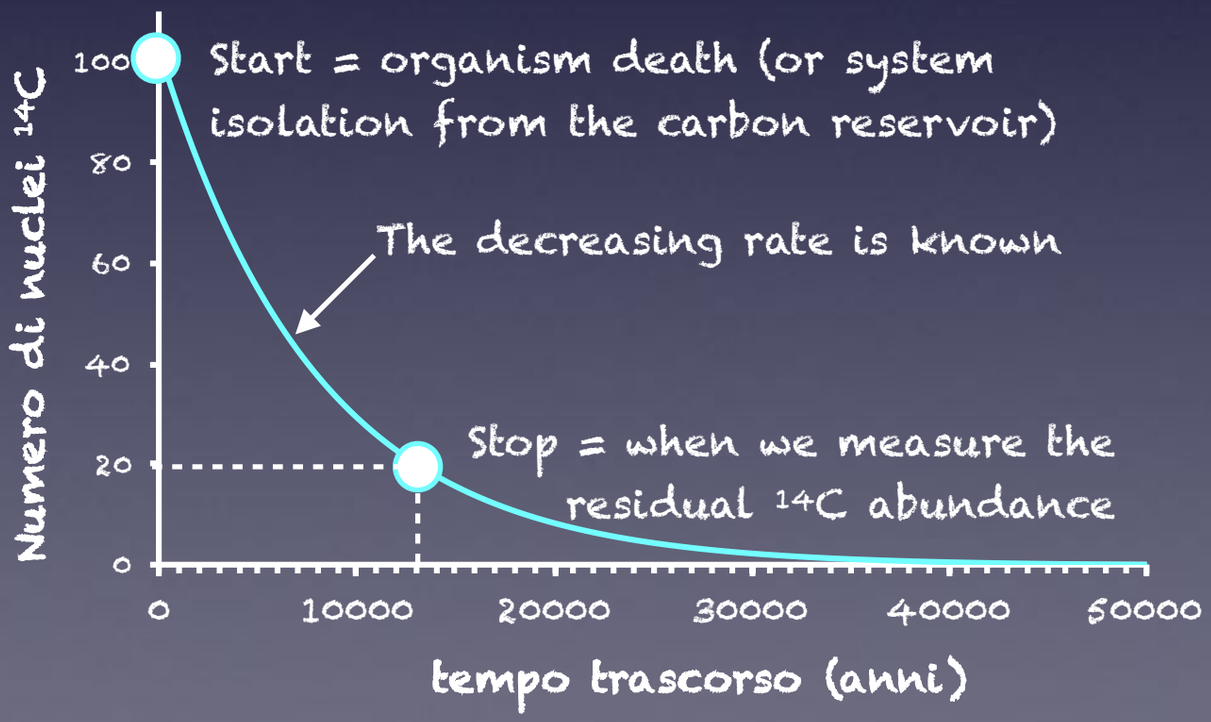
A small accelerator and the dating issue

- ^{14}C can be used as a natural chronometer to date organic materials, or - in more generally speaking - those systems that have been directly (or indirectly) in equilibrium with atmosphere, acquiring ^{14}C from it, and that have ceased that equilibrium condition in a well defined moment



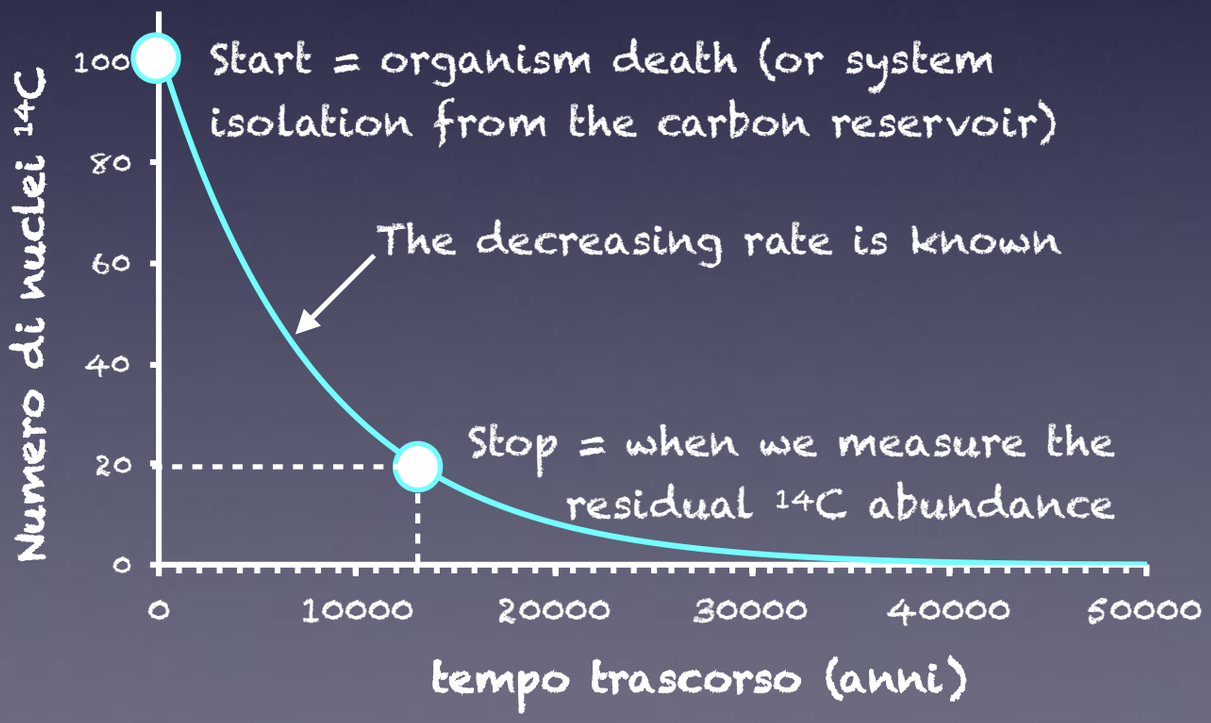
A small accelerator and the dating issue

- ^{14}C can be used as a natural chronometer to date organic materials, or - in more generally speaking - those systems that have been directly (or indirectly) in equilibrium with atmosphere, acquiring ^{14}C from it, and that have ceased that equilibrium condition in a well defined moment



A small accelerator and the dating issue

- ^{14}C can be used as a natural chronometer to date organic materials, or - in more generally speaking - those systems that have been directly (or indirectly) in equilibrium with atmosphere, acquiring ^{14}C from it, and that have ceased that equilibrium condition in a well defined moment



Thanks to Nicolas Alamanos for having explained the principles of radiocarbon dating in his introductory lecture

Few pills of radiocarbon dating

- From the measurement we obtain the conventional radiocarbon age (measured in years BP):

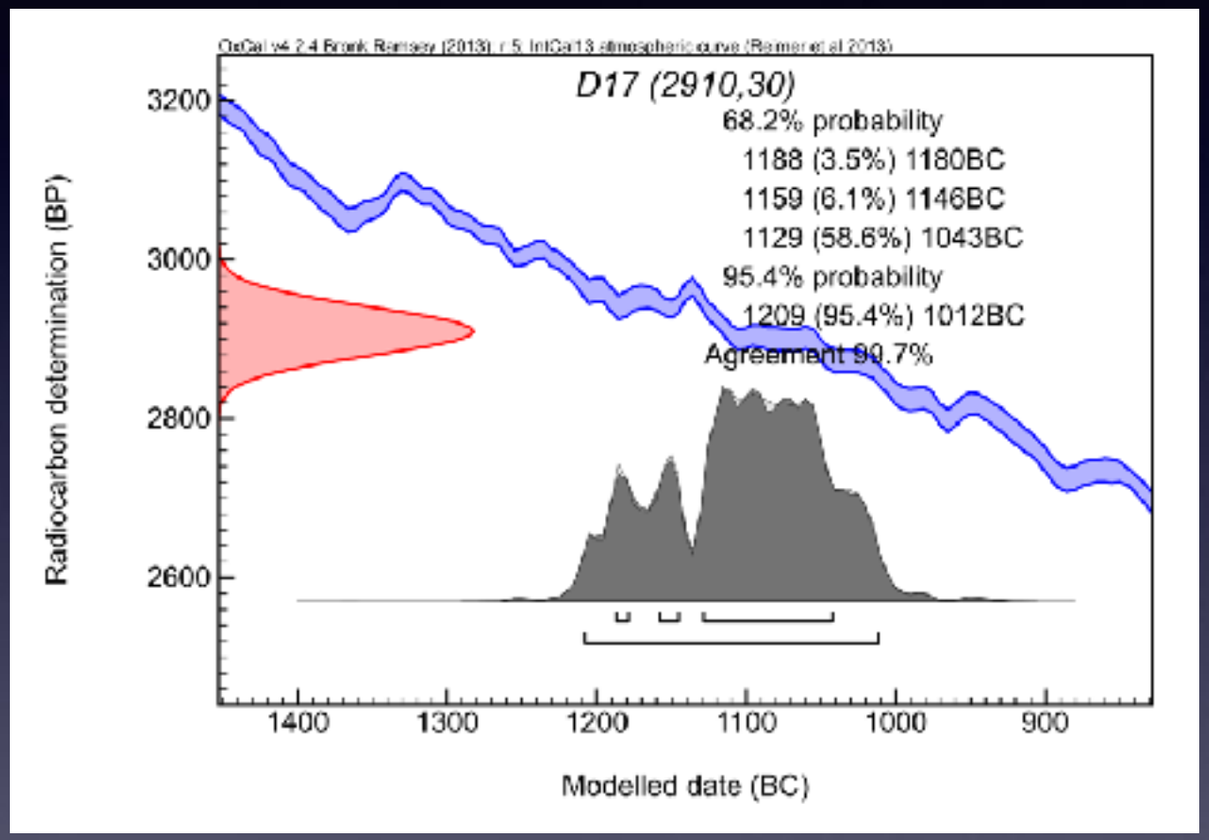
$$t_{rc} = \tau \cdot ln \frac{^{14}R}{^{14}R_0}$$

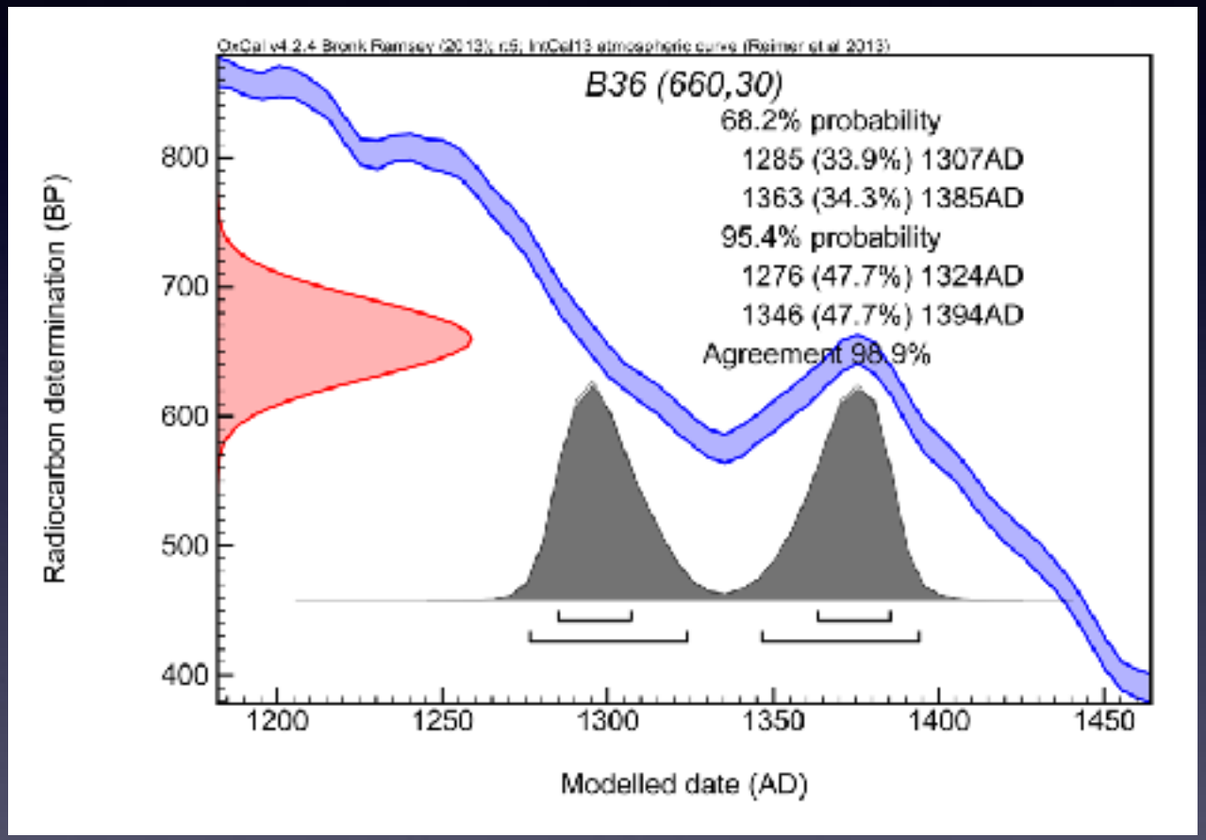
  - T = 8033 years (Libby half life)
  - According to the international agreement, $^{14}R_0$ is basically chosen as the radiocarbon concentration in 1950

- Pay attention to correct for isotopic fractionation
- Pay attention to calibrate the measured conventional radiocarbon age

We can date organic materials (or systems once in equilibrium with the atmosphere that have become close systems)
We always date the moment of the "death" of the organism

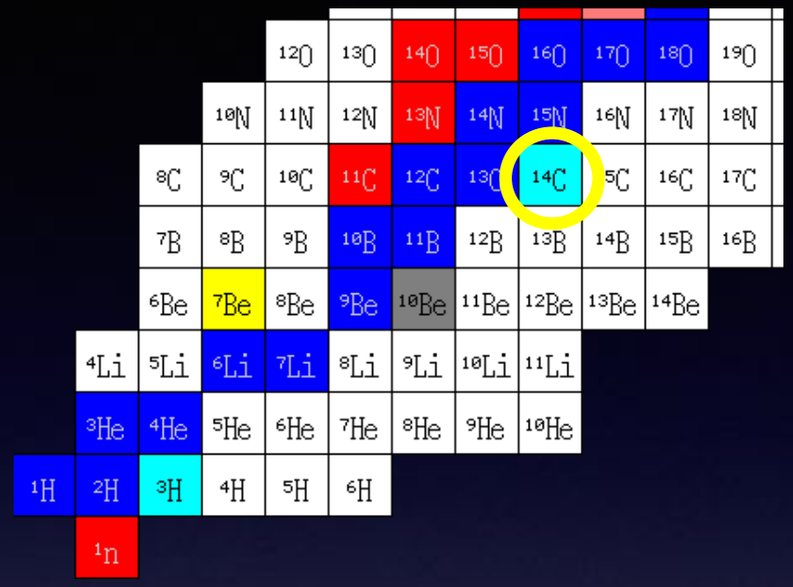
How a radiocarbon result looks like





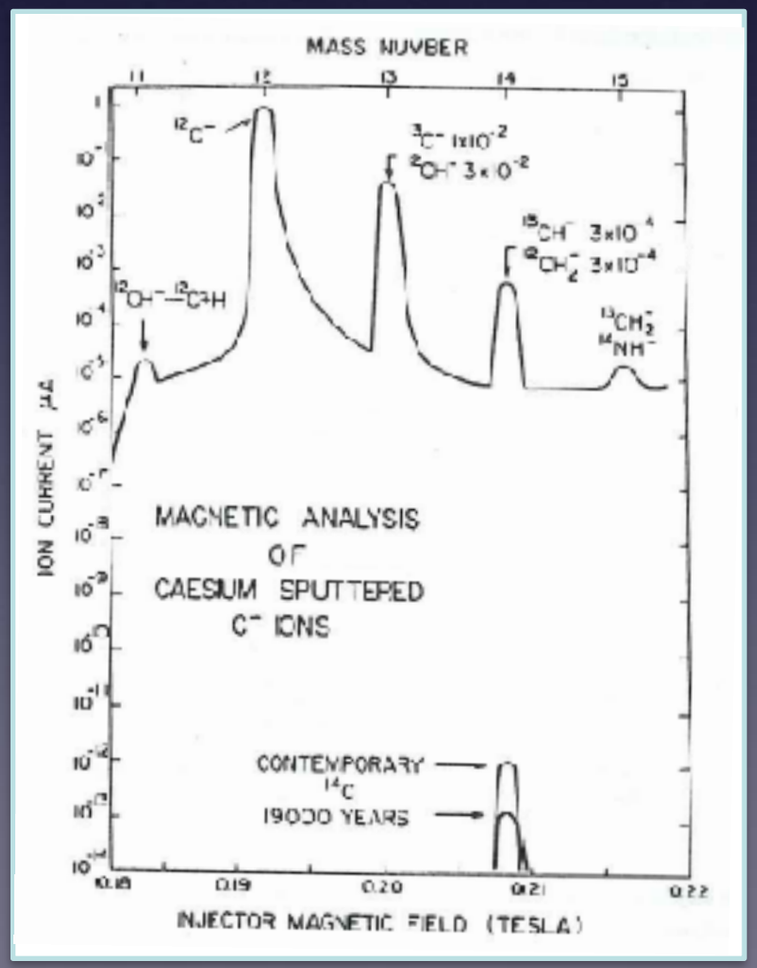
Why the accelerator

- ^{14}C is a rare isotope
  - Low activity –> counting emitted beta is not favored
  - Low natural abundance (~10^{-12} in the atmosphere) –> in principle measurable by mass spectrometric techniques
- Presence of highly abundant isobars



	^{14}N	$^{12}CH_2$	^{13}CH
Δm/m	0.000012	0.0009	0.0003

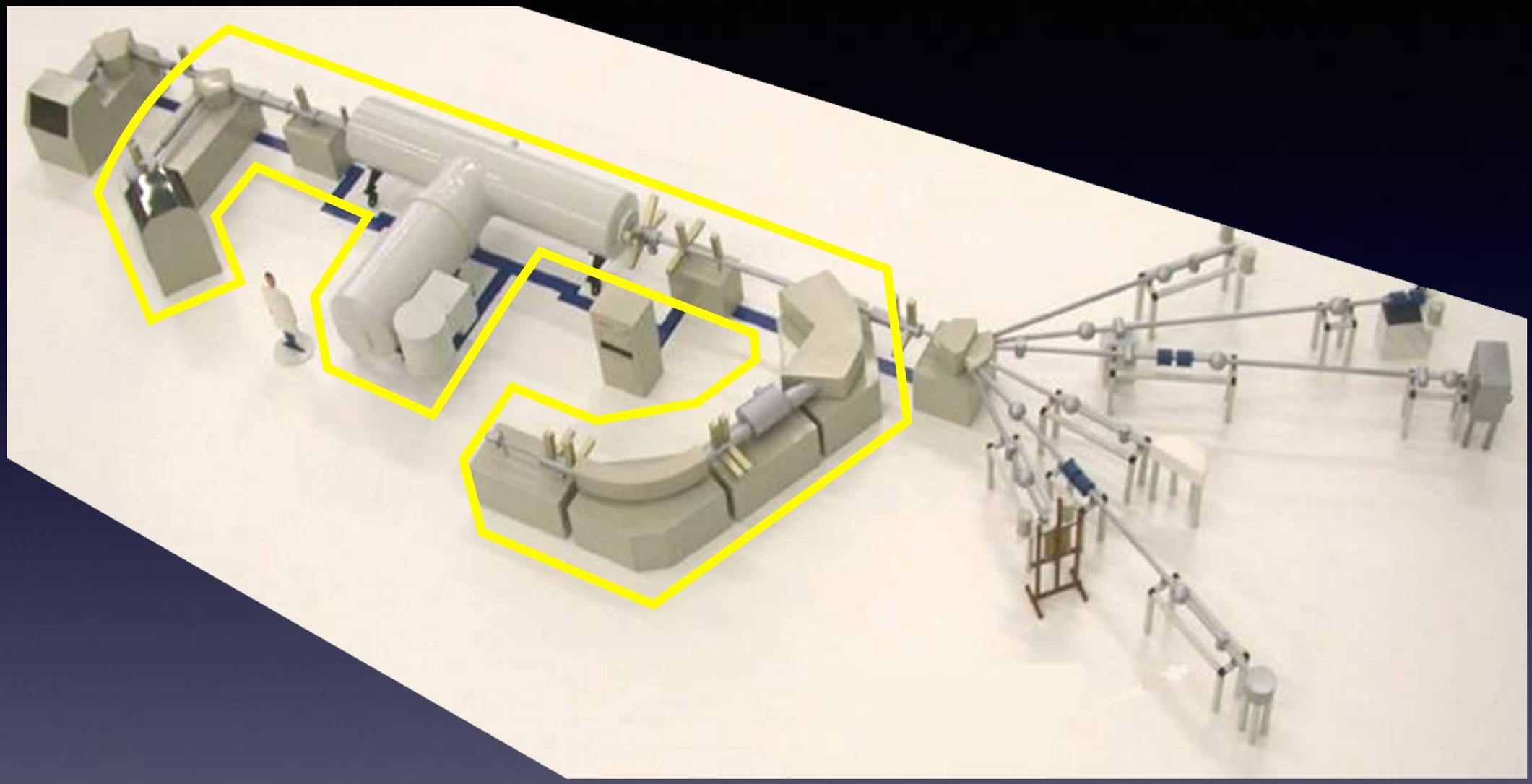
Radiocarbon is a sort of "needle in a stack"
We need a highly sensitive technique even when the sample is very small (few milligrams)



Accelerator Mass Spectrometry

- The key: using a tandem electrostatic accelerator to suppress the isobars abundance (both elemental and molecular interferences)

Accelerator Mass Spectrometry



- The key: using a tandem electrostatic accelerator to suppress the isobars abundance (both elemental and molecular interferences)

The importance of being a tandem

- Negative Ion Source:
  –> some elements do not form stable negative ions –> suppression of elemental isobars

AN ATTEMPT TO DETECT STABLE N^- IONS FROM A SPUTTER ION SOURCE AND SOME IMPLICATIONS OF THE RESULTS FOR THE DESIGN OF TANDEMS FOR ULTRA-SENSITIVE CARBON ANALYSIS

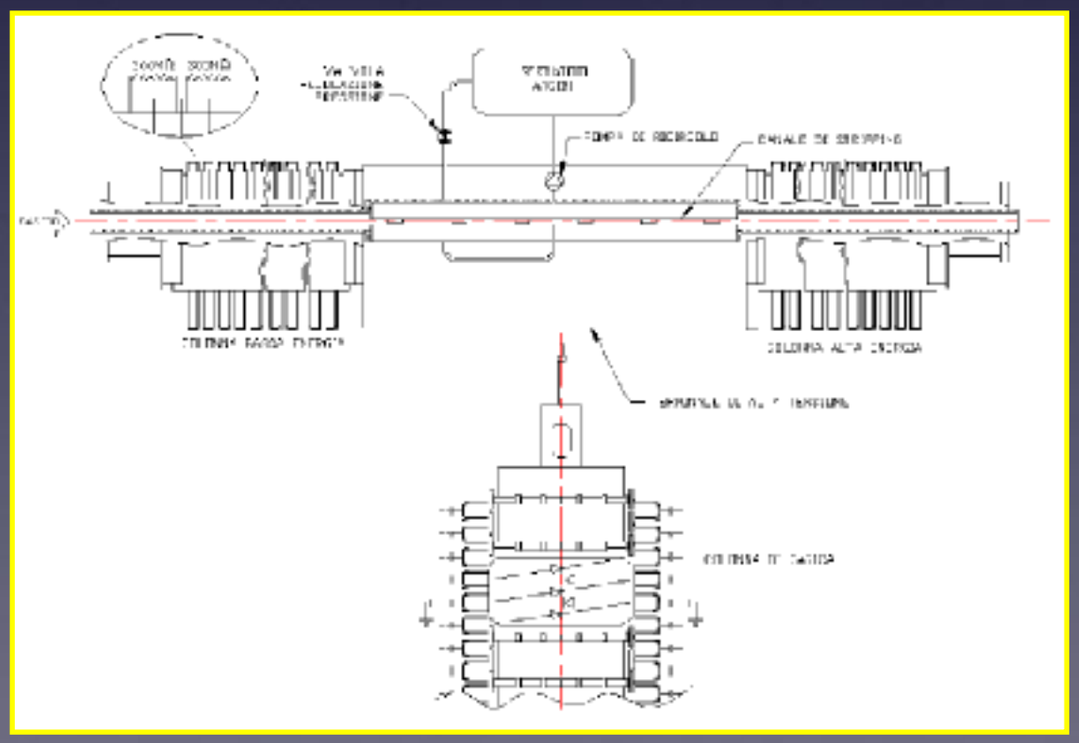
K. H. PURSER and R. B. LIEBERT
General Ionex Corporation
Ipswich, MA 01938, U.S.A.

A. E. LITHERLAND and R. P. BEUKENS
University of Toronto(*)
Toronto, Ont., Canada

H. E. GOVE, C. L. BENNETT, M. R. CLOVER and W. E. SONDHEIM
Nuclear Structure Research Laboratory(**)
University of Rochester, Rochester, NY 14627, U.S.A.

Résumé. — Dans cette communication on discute d'une expérience où l'on fait une recherche d'ions $^{14}N^-$ en présence d'un faisceau intense de carbone. On utilise une source d'ions Middleton à *sputtering*. Malgré un champ accélérateur de 1.5 MV/m nous n'avons pas trouvé d'ions azote négatifs avec des intensités détectables. La publication indique aussi les conditions que doivent remplir l'accélérateur et son tout bénéficier pleinement de ce résultat négatif afin de faire des mesures avec des faisceaux très faibles de ^{14}C.

Abstract. — The present paper discusses an experiment, using a Middleton sputter ion source, where a sensitive search was made for $^{14}N^-$ ions in the presence of an intense carbon beam. It was found that negative nitrogen ions, capable of withstanding acceleration fields of 1.5 MV/m, were not produced in detectable intensities. The paper also points out some design requirements of tandem facilities that must be satisfied if full advantage is to be taken of this negative result to permit the measurement of very weak ^{14}C beams.

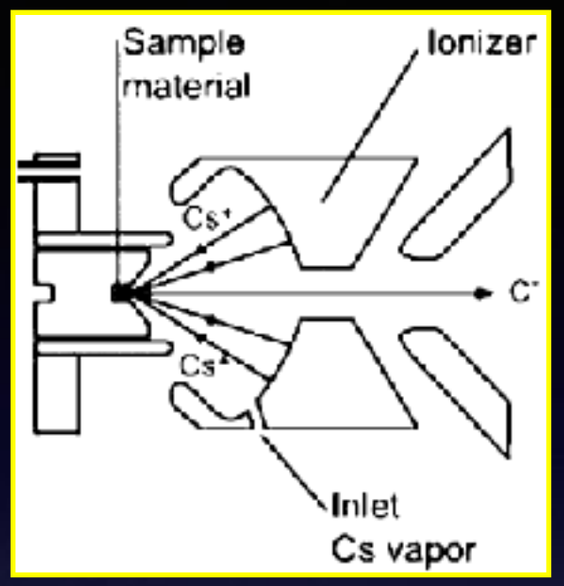


- Stripping:
  –> after stripping at the high voltage terminal, molecules are not stable (if we consider charge >1+) –> suppression of molecular isobars

AMS: the general scheme

- Sensitivity on concentration measurement down to ~10^{-15} –> dating samples as older as 40000 years

About the negative ion source



- It is based on sputtering by Cs^+ ions
- Negative ions (elemental, molecular, cluster…) are extracted through a voltage of few tens of keV
- Not all the elements can be extracted as negative ions

Ionization Potentials and Electron Affinities of the Elements

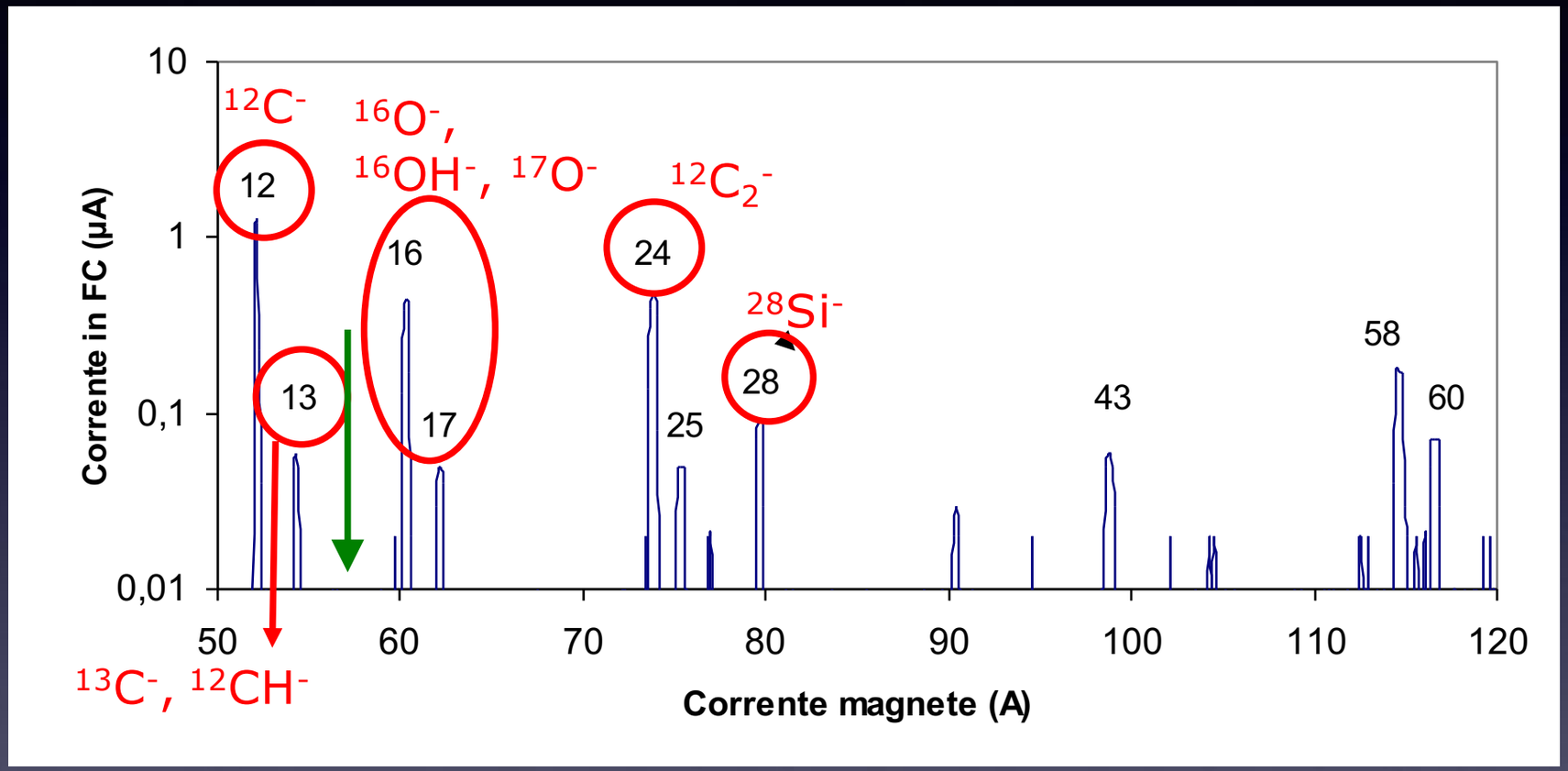
IA	IIA	IIIA	IVA	VA	VIA	VIIA	VIIIA
$_1$H 13.59 0.754			Ionization Potential Electron Affinity				$_2$He 24.48 0.078*
$_3$Li 5.39 0.618	$_4$Be 9.32 0.195*	$_5$B 8.30 0.277	$_6$C 11.26 1.263	$_7$N 14.53 0.07	$_8$O 13.61 1.461	$_9$F 17.42 3.399	$_{10}$Ne 21.56 < 0
$_{11}$Na 5.14 0.548	$_{12}$Mg 7.64 < 0	$_{13}$Al 5.98 0.441	$_{14}$Si 8.15 1.385	$_{15}$P 10.48 0.747	$_{16}$S 10.36 2.077	$_{17}$Cl 13.01 3.617	$_{18}$Ar 15.76 < 0
$_{19}$K 4.34 0.501	$_{20}$Ca 6.11 0.043	$_{31}$Ga 6.00 0.30	$_{32}$Ge 7.90 1.2	$_{33}$As 9.81 0.81	$_{34}$Se 9.75 2.021	$_{35}$Br 11.81 3.365	$_{36}$Kr 14.00 < 0
$_{37}$Rb 4.18 0.486	$_{39}$Sr 5.70 < 0	$_{49}$In 5.79 0.3	$_{50}$Sn 7.34 1.2	$_{51}$Sb 8.64 1.07	$_{52}$Te 9.01 1.971	$_{53}$I 10.45 3.059	$_{54}$Xe 12.13 < 0
$_{55}$Cs 3.89 0.472	$_{56}$Ba 5.21 < 0	$_{81}$Tl 6.11 0.2	$_{82}$Pb 7.42 0.364	$_{83}$Bi 7.29 0.946	$_{84}$Po 8.42 1.9	$_{85}$At 9.5 2.8	$_{86}$Rn 10.75 < 0

*Metastable

^{14}N is suppressed

R. Middleton, A negative-ion cookbook (www.pelletron.com/cookbook.pdf)

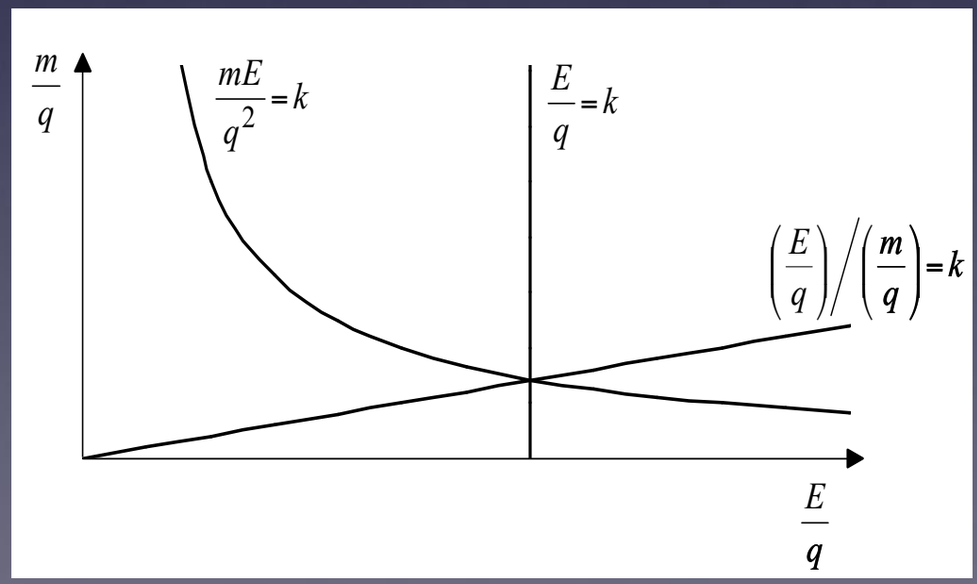
Which ions inside the beam?



- We need some elements to discriminate the particles according to their mass, energy and charge

Beam analysis

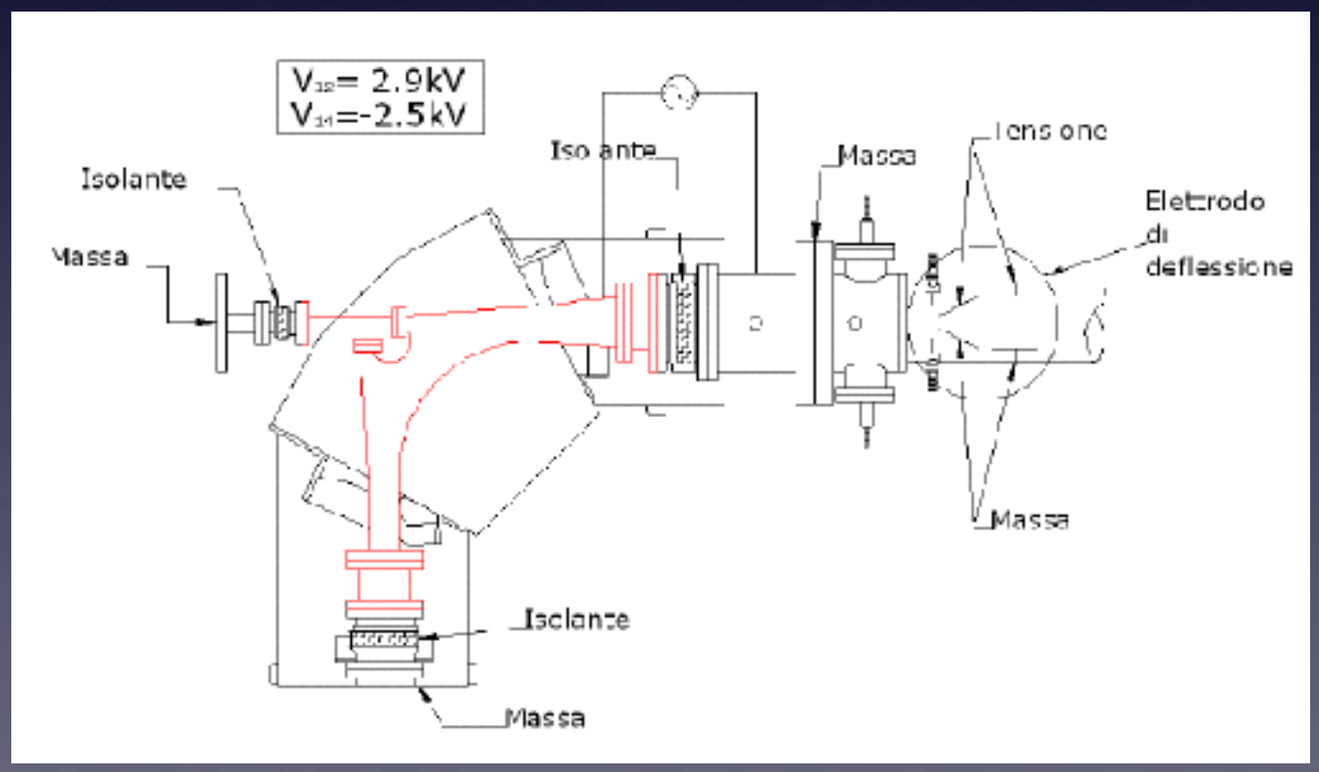
- Electrostatic analysis: $m\frac{v^2}{r} = q\mathscr{E} \rightarrow \mathscr{E}r = 2\frac{E}{q}$
- Magnetic analysis: $m\frac{v^2}{r} = qvB \rightarrow Br = \frac{\sqrt{2mE}}{q}$
- Wien filter: $q\mathscr{E} = qvB \rightarrow \left(\frac{\mathscr{E}}{B}\right)^2 = 2\frac{E}{m}$

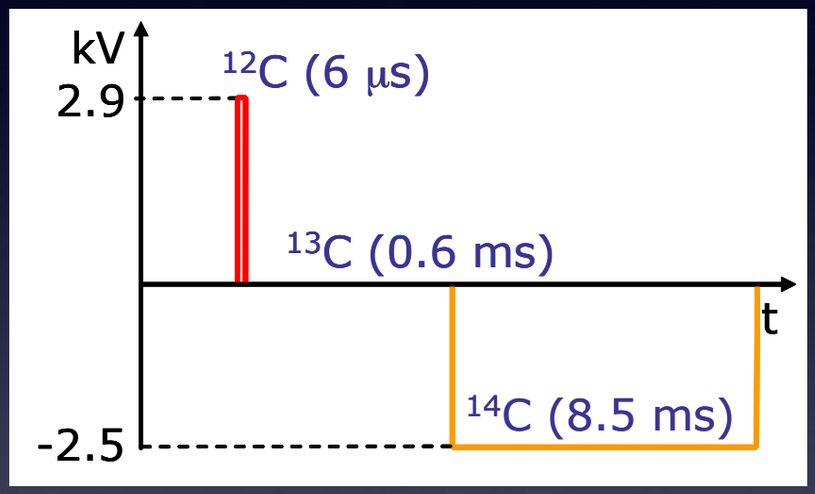


Each of the analysis identifies a locus of points

Bouncing injection

- Magnet kept fixed to transmit mass 13
- Other masses injected thanks to different beam energies inside the magnet chamber



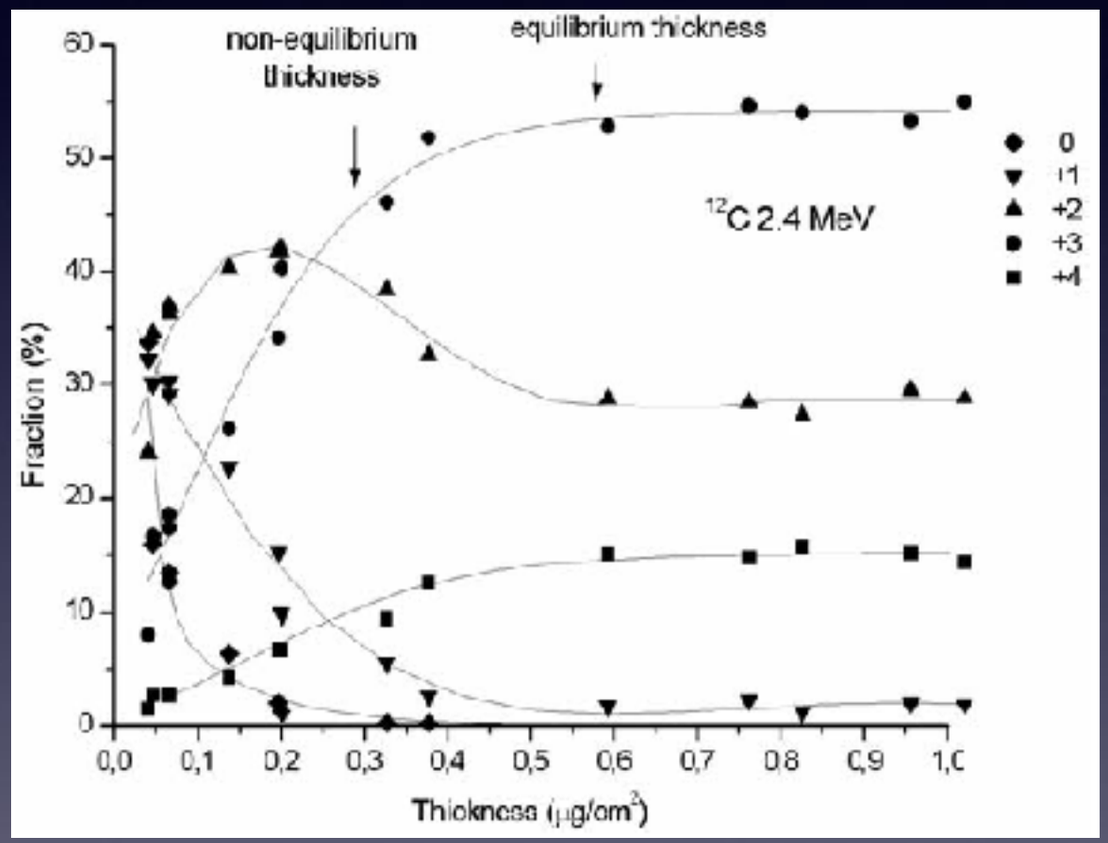


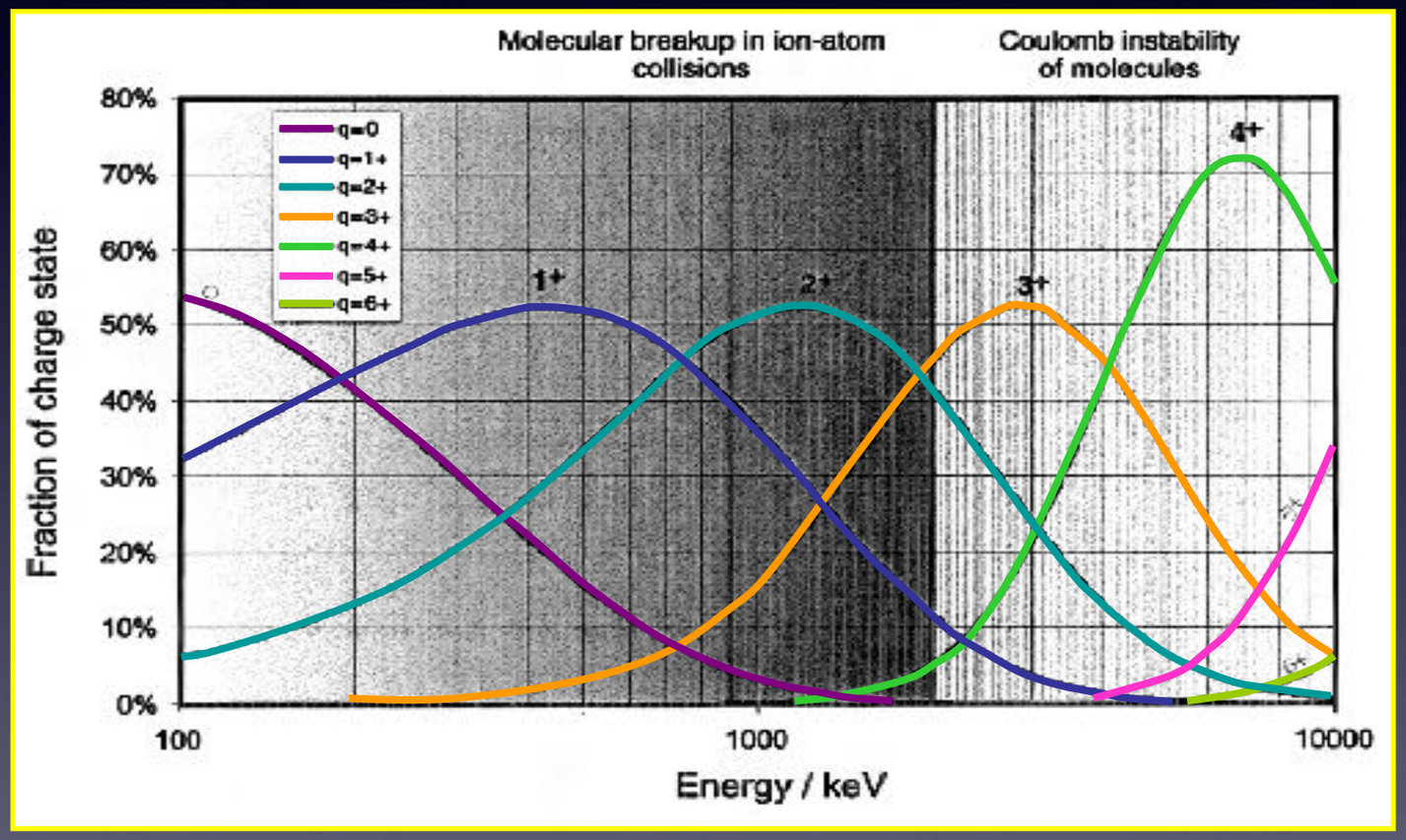
Injection times adjustable according to the average currents we would like to measure on the high energy side

Molecular isobars are injected as well

The stripping process

- Distribution of charge state after stripping depends on stripper thickness and incident ion velocity





$$E_{fin} = \left(E_{inj} + eV_t\right)\frac{m_i}{m_{tot}} + qeV_t$$

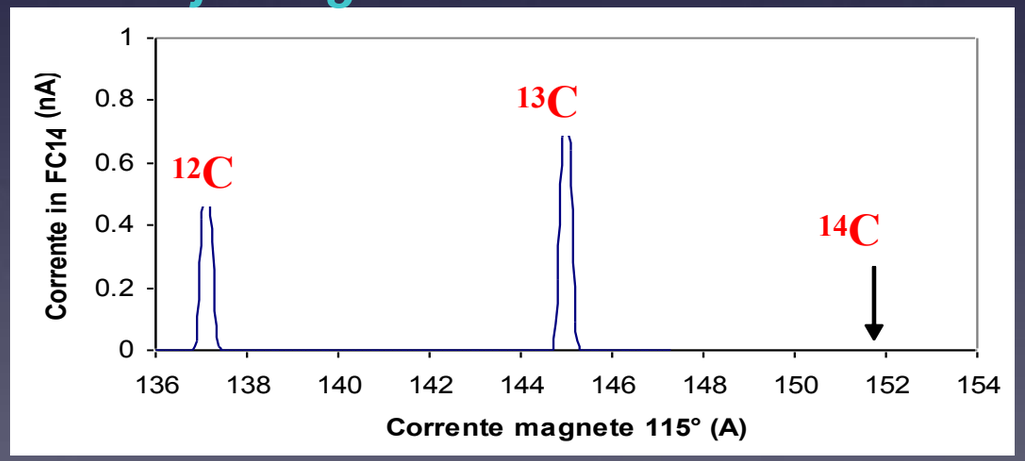
What happens to molecular isobars

- After stripping, the probability to find molecules characterized by a high charge state is low (Coulomb explosion) –> the capability to suppress isobars is due to the combination of terminal voltage, stripper thickness and charge state, which is analyzed on the high energy side

What happens to molecular isobars

- After stripping, the probability to find molecules characterized by a high charge state is low (Coulomb explosion) –> the capability to suppress isobars is due to the combination of terminal voltage, stripper thickness and charge state, which is analyzed on the high energy side

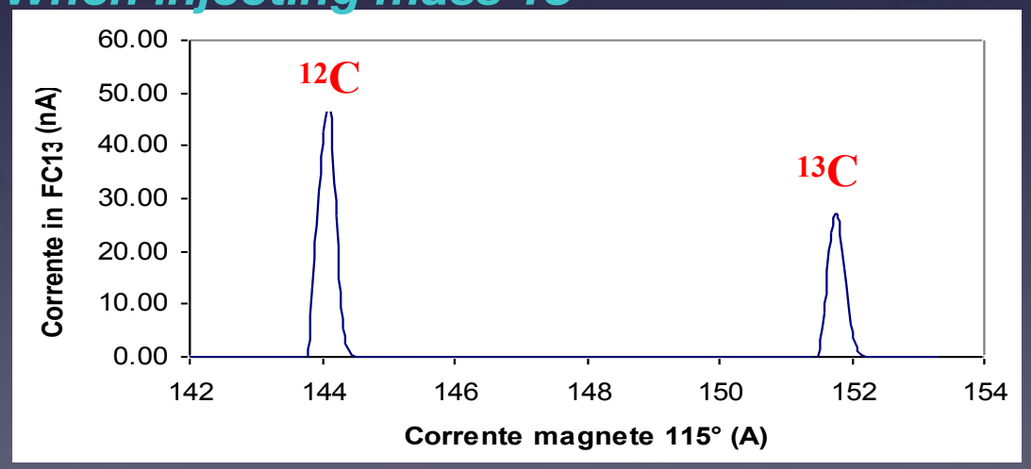
When injecting mass 14



What happens to molecular isobars

- After stripping, the probability to find molecules characterized by a high charge state is low (Coulomb explosion) –> the capability to suppress isobars is due to the combination of terminal voltage, stripper thickness and charge state, which is analyzed on the high energy side

When injecting mass 13

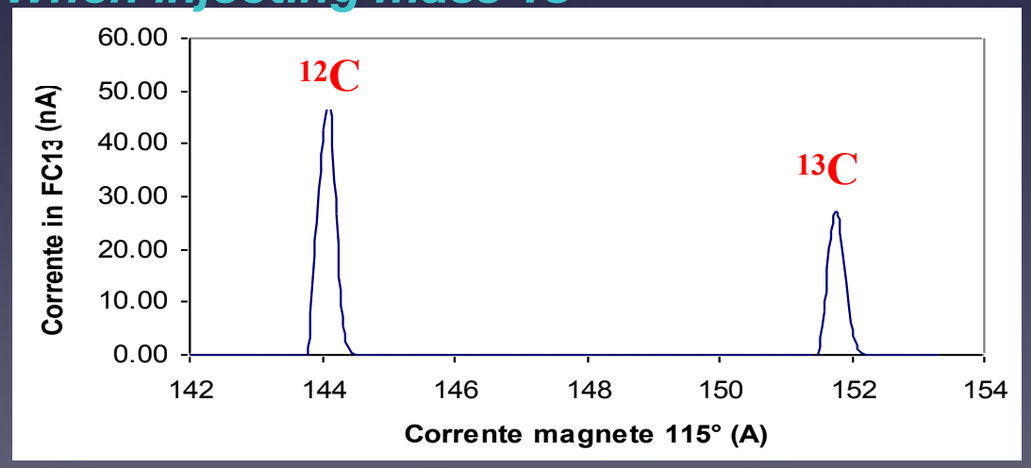


What happens to molecular isobars

- After stripping, the probability to find molecules characterized by a high charge state is low (Coulomb explosion) –> the capability to suppress isobars is due to the combination of terminal voltage, stripper thickness and charge state, which is analyzed on the high energy side

When working at 2.5 MV, the charge state which is analyzed at the high energy side is the most probable i.e. 3+
Working at lower voltages, we should choose lower charge state → decreased capability of isobars suppression (increase of the equivalent stripper thickness)

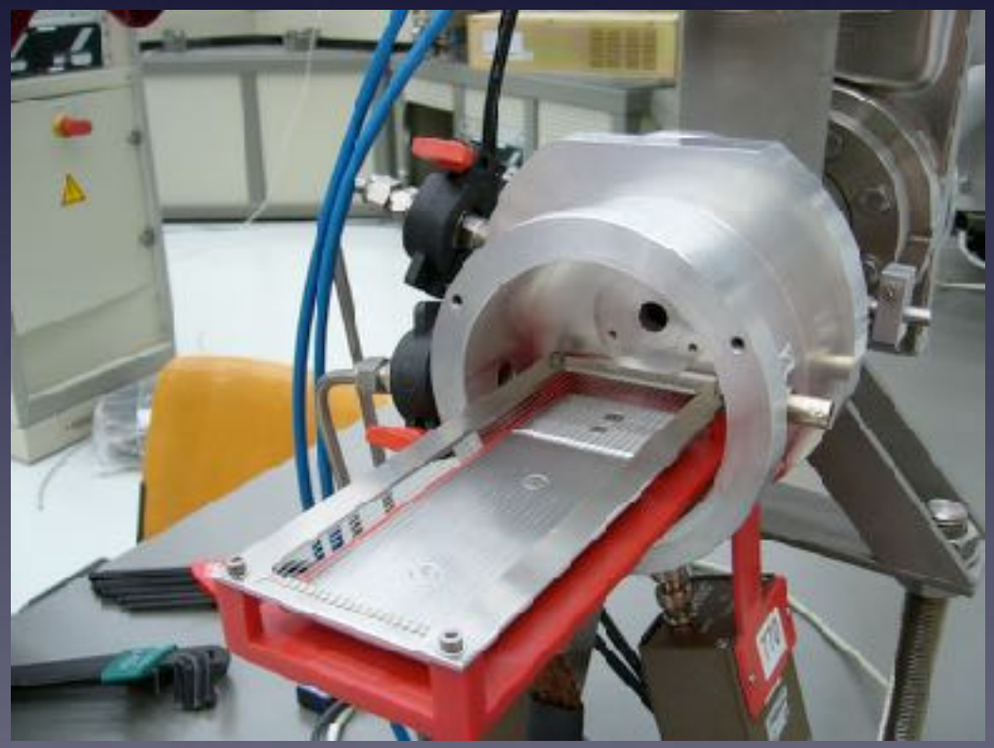
When injecting mass 13



About the rare isotope detector

- Possibility to measure the energy of the ions
- Possibility to use the detector just as a simple counter

Ionization chamber

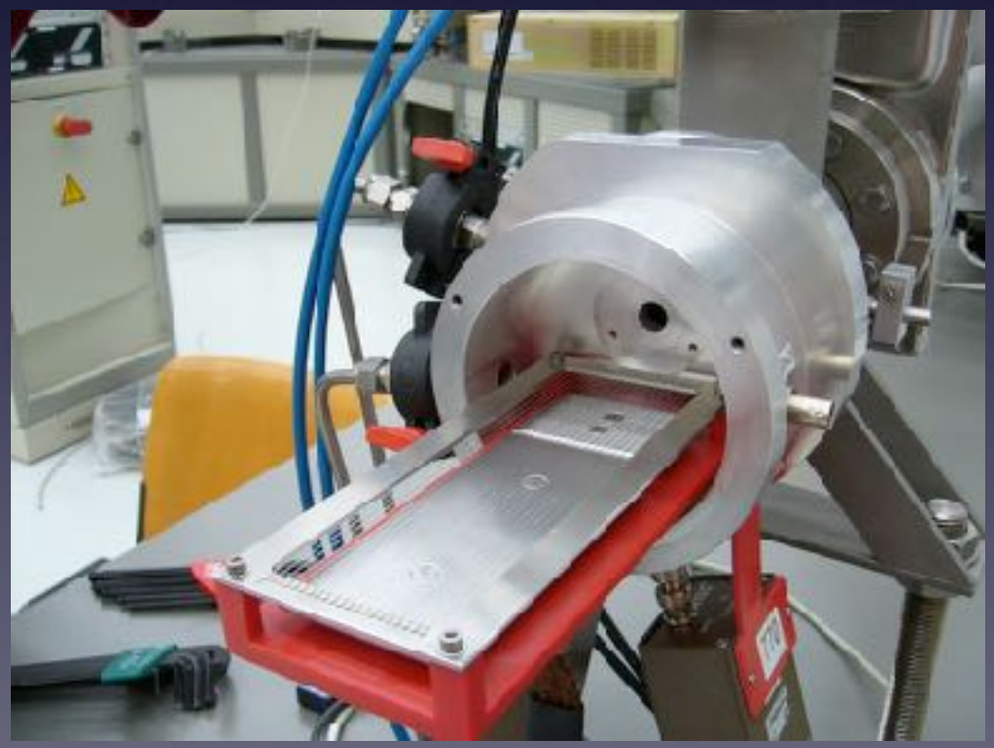


Silicon diode

About the rare isotope detector

- Possibility to measure the energy of the ions
- Possibility to use the detector just as a simple counter

Ionization chamber



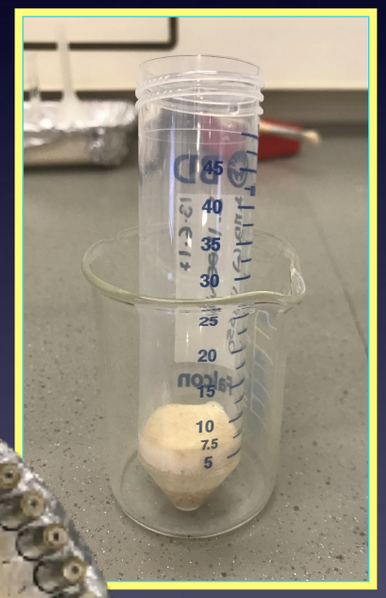
Silicon diode

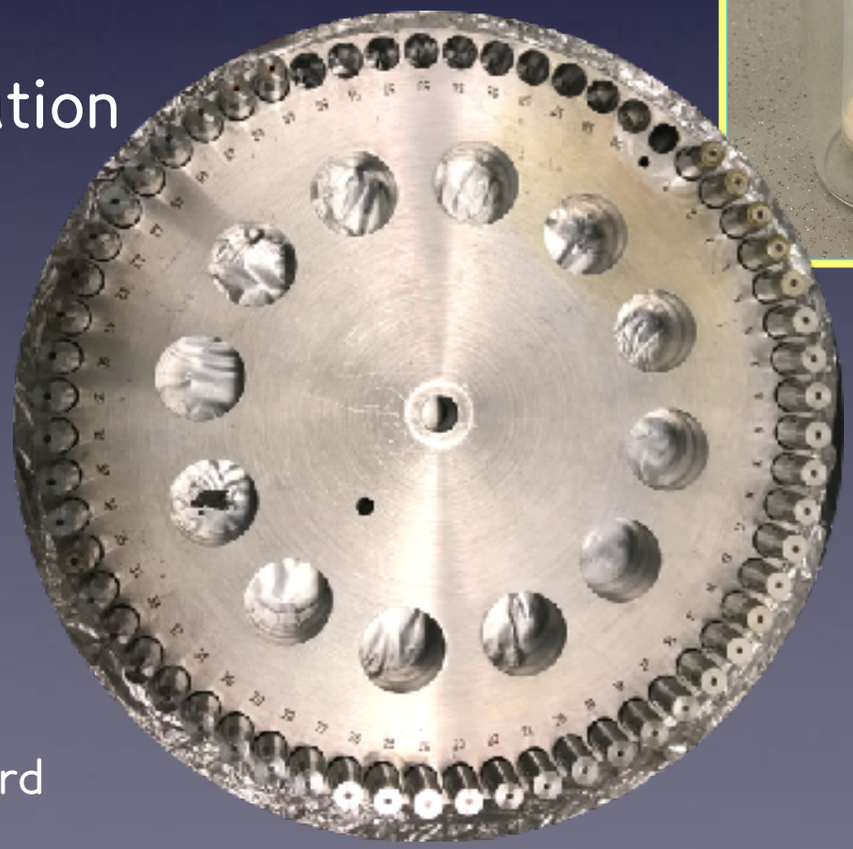
Counting rate < 20 Hz!

No appreciable radiation damage!

How to run a dating measurement

- Sample selection:
  - Identification of the material
  - Identification of the finding context (e.g. the archaeological excavation)
  - Information on its preservation state
- Sample preparation
  - To remove any possible contamination
  - To convert the cleaned material to the most appropriate chemical form (e.g. graphite)
- AMS measurement and data analysis





Archaeological sites

Archaeological sites

Baratti, Toscana – Etruscan and Roman-imperial periods

Erimi Laonin Tou Porakou, Cipro – Bronze Age and Hellenistic Age

Artworks

Artworks

Trittico di Badia a Rofeno - frame, Ambrogio Lorenzetti

Artworks

F. Léger (attr.), Contraste des Formes

Trittico di Badia a Rofeno - frame, Ambrogio Lorenzetti

Artworks

F. Léger (attr.), Contraste des Formes

Trittico di Badia a Rofeno - frame, Ambrogio Lorenzetti

U. Boccioni, L'Idolo Moderno

Do you remember Léger?

Contraste de formes
Peggy Guggenheim Collection
(never shown to the public)

Do you remember Léger?

Contraste de formes
Peggy Guggenheim Collection
(never shown to the public)

Do you remember Léger?

^{14}C = (129.05 ± 0.68) pMC

Contraste de formes
Peggy Guggenheim Collection
(never shown to the public)

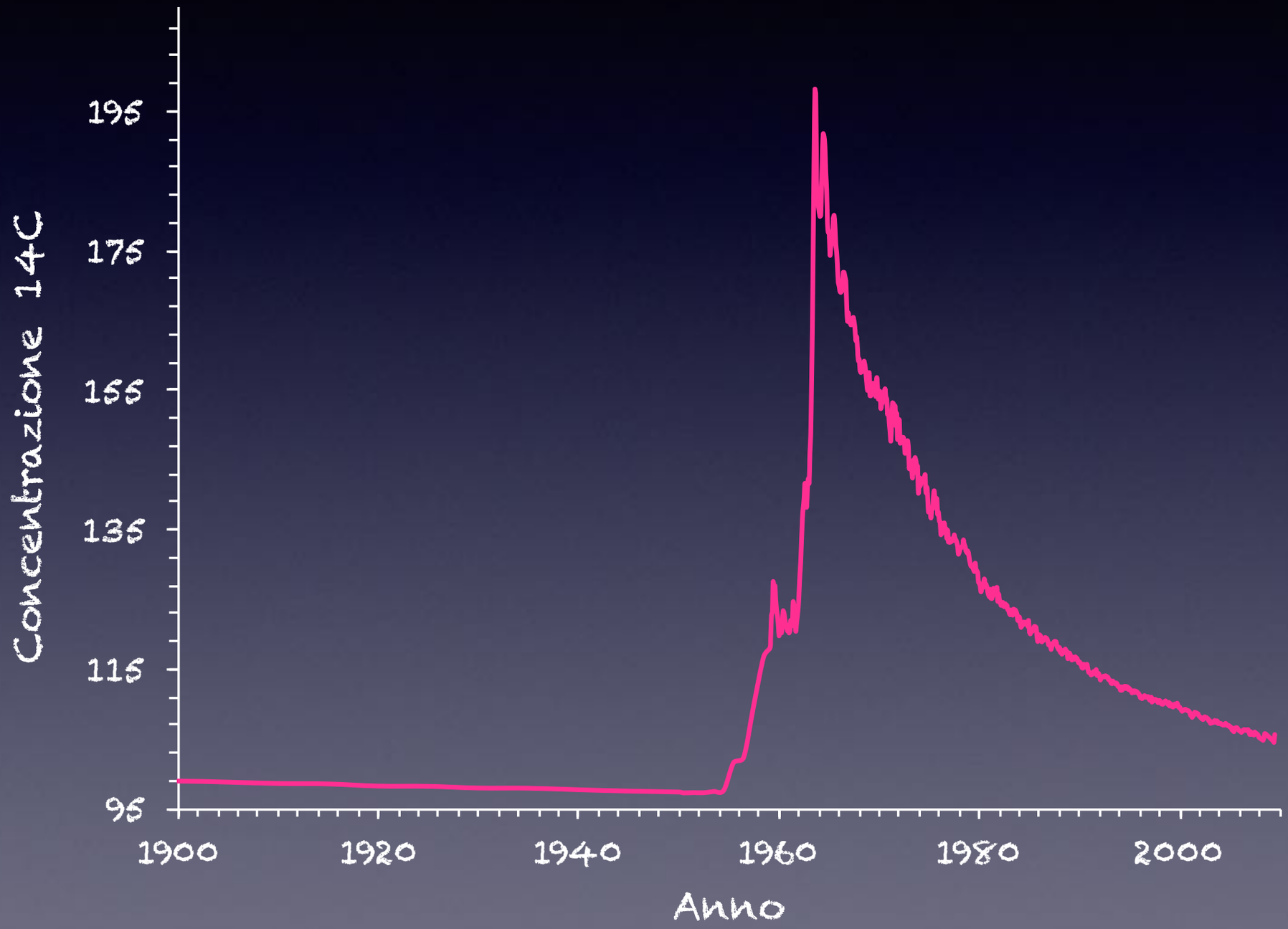
Do you remember Léger?

^{14}C = (129.05 ± 0.68) pMC

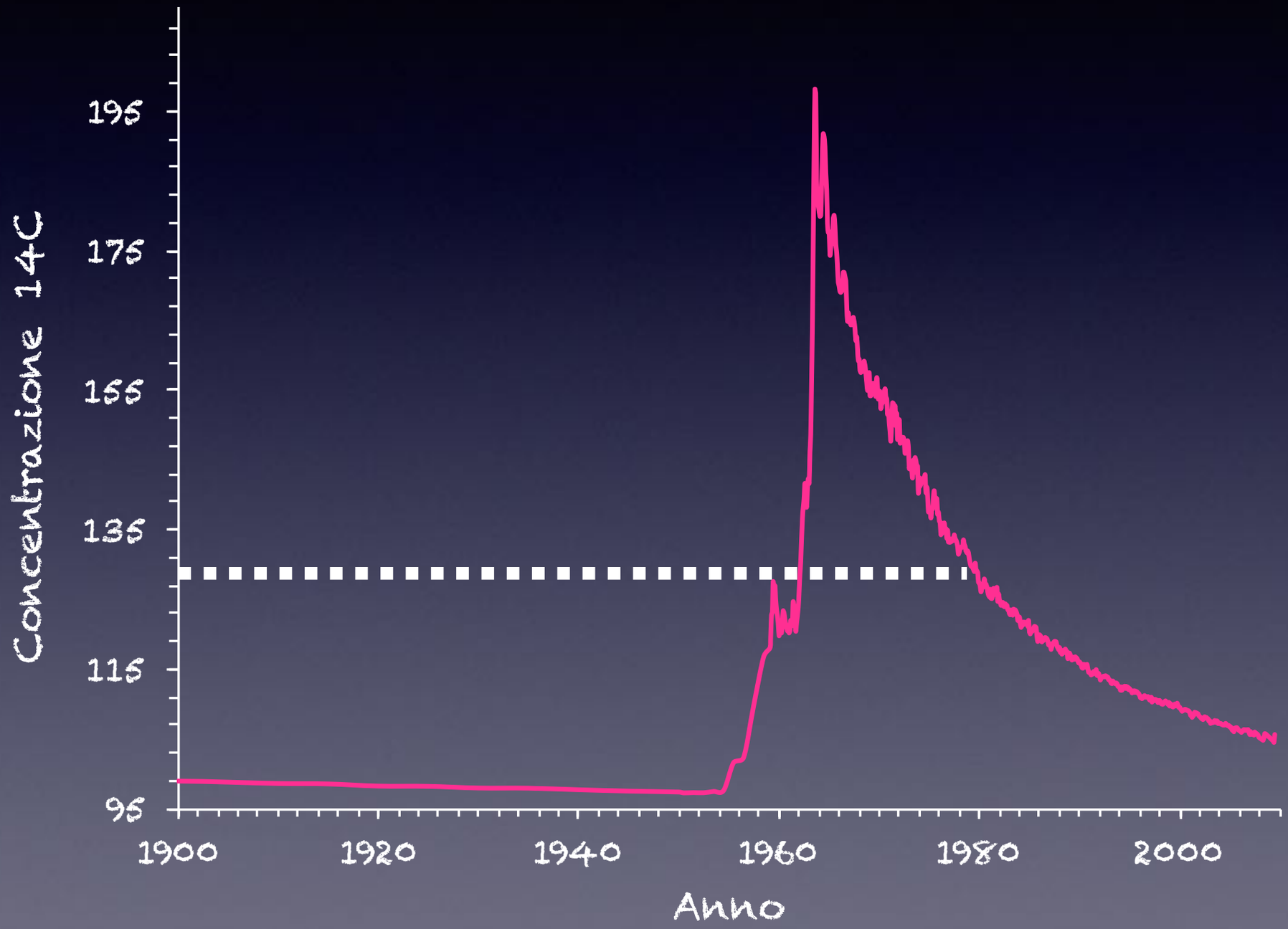
Contraste de formes
Peggy Guggenheim Collection
(never shown to the public)

Ma per un campione moderno
^{14}C ~ 100 pMC…

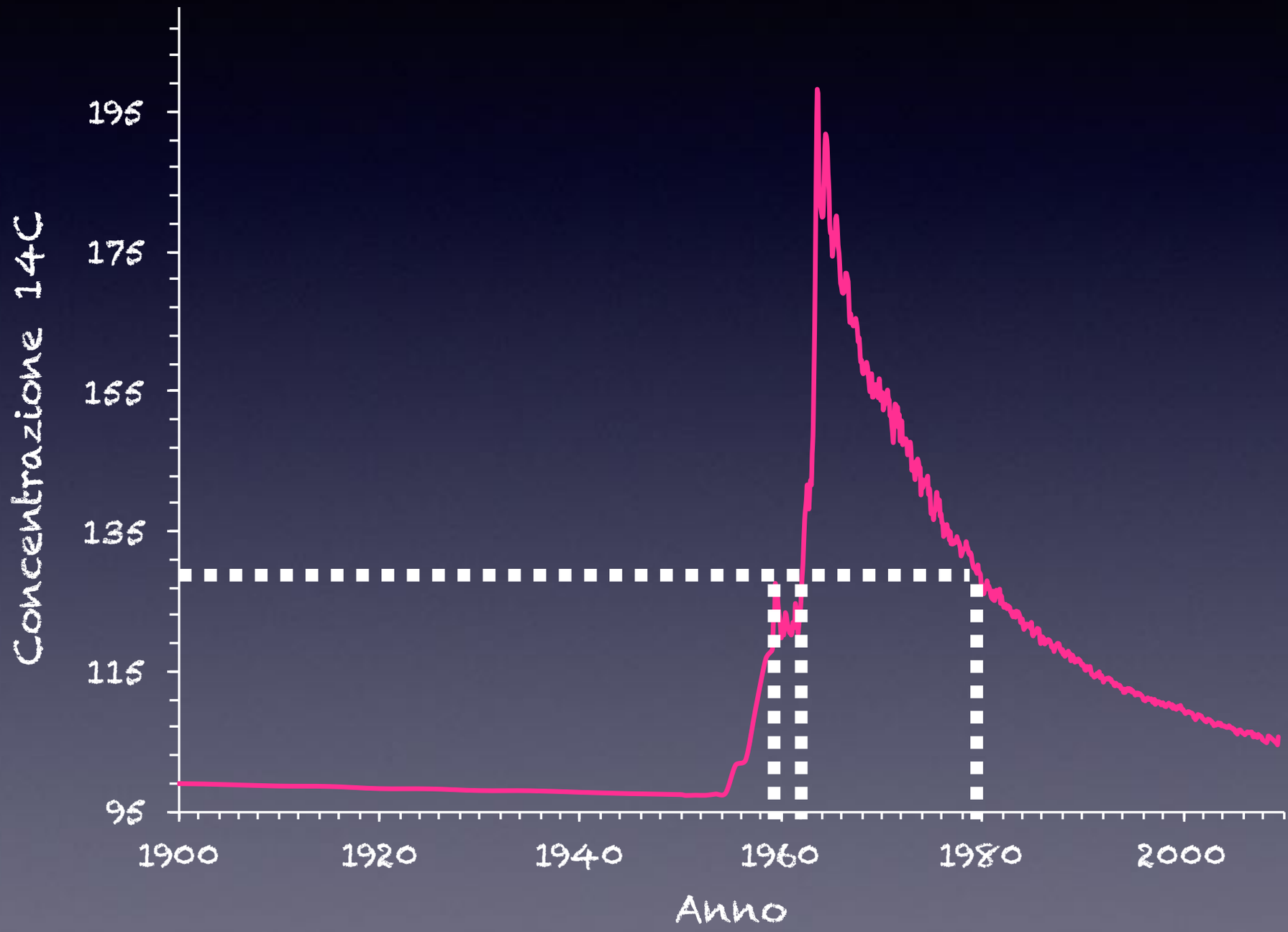
Look at the “Bomb Peak”!



Look at the “Bomb Peak”!



Look at the “Bomb Peak”!



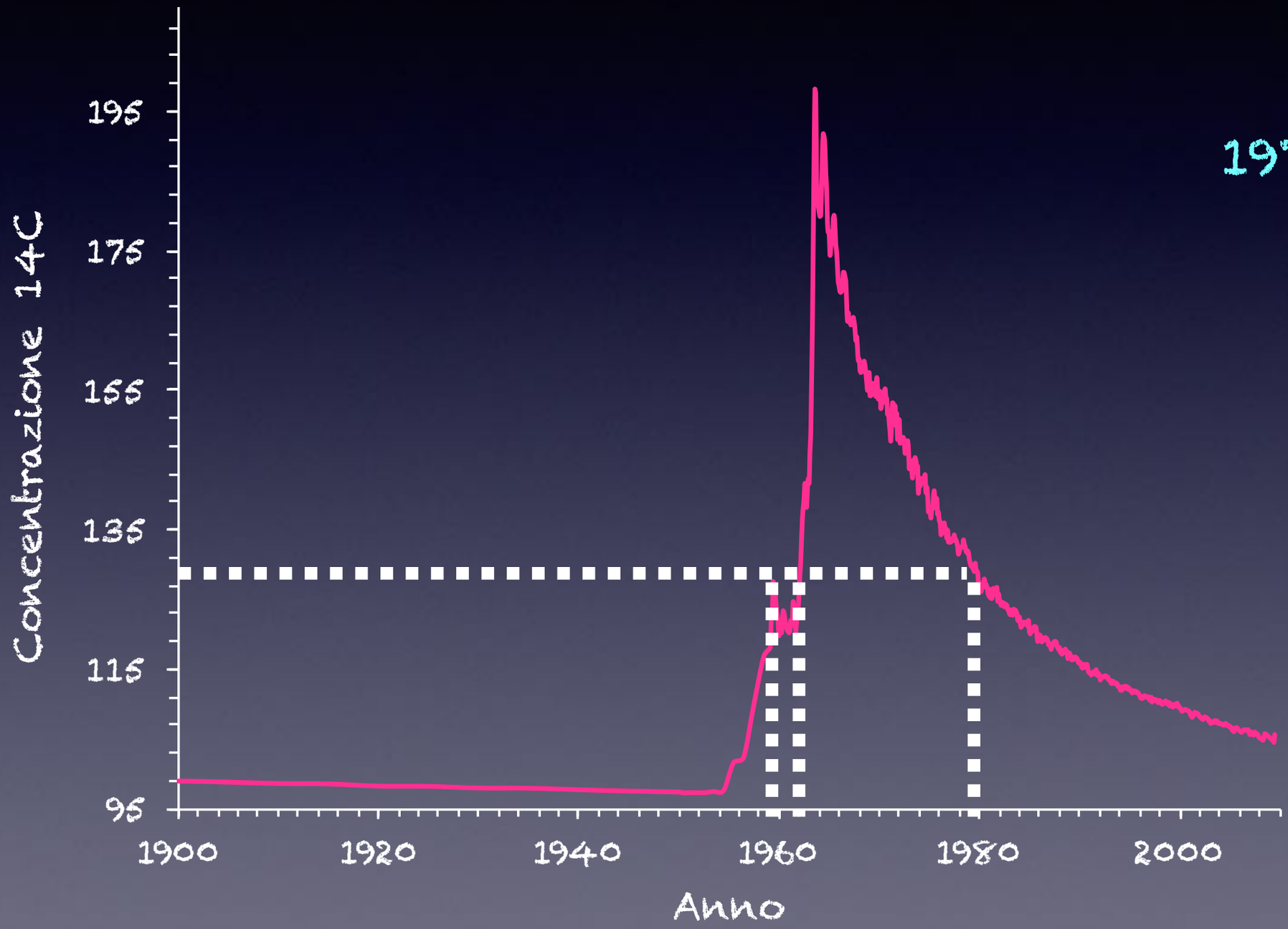
Look at the “Bomb Peak”!

Three probable ranges of calibrated age:

1959

1962

1979–1980



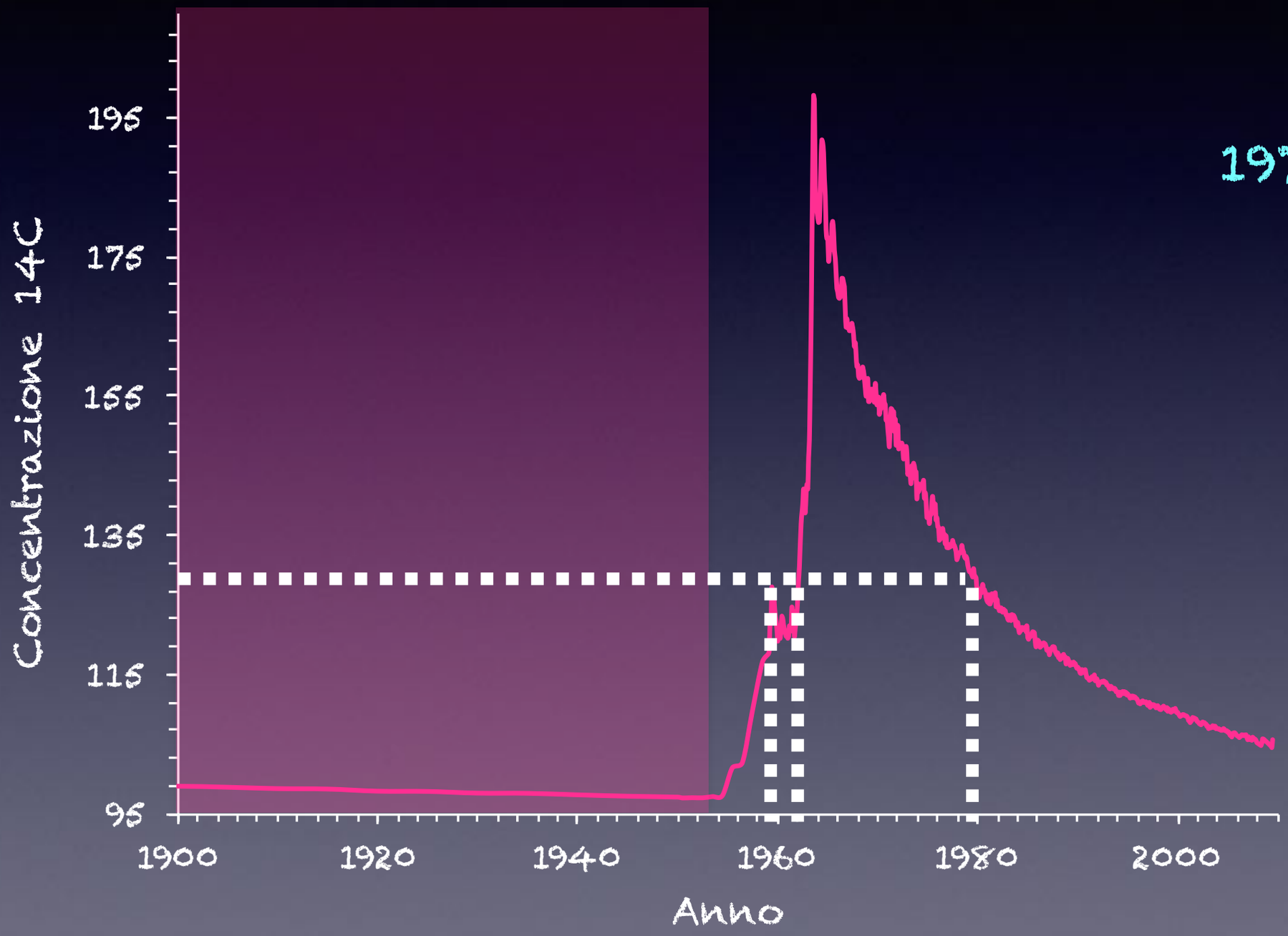
Look at the “Bomb Peak”!

Three probable ranges of calibrated age:

1959

1962

1979–1980



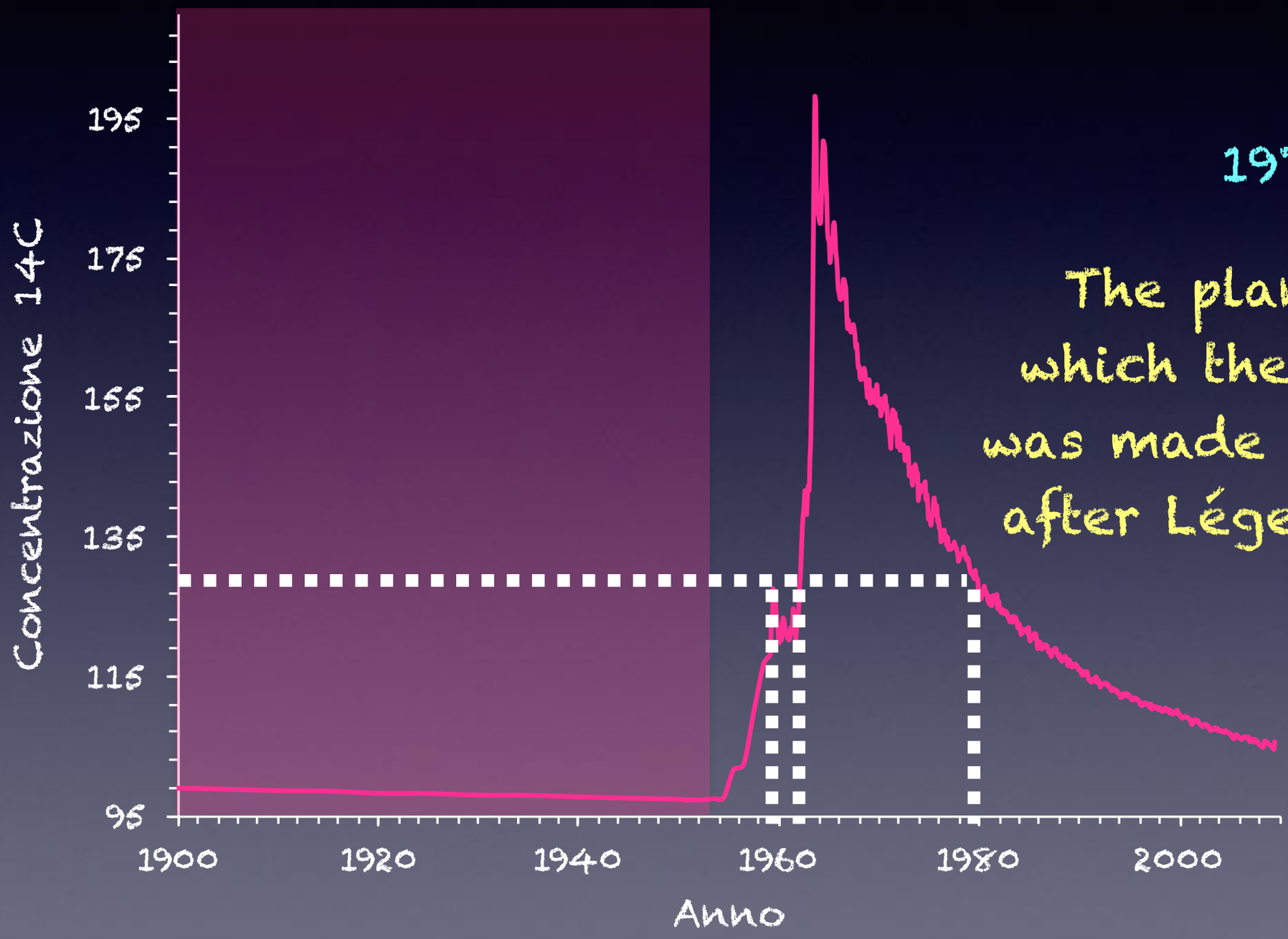
Look at the “Bomb Peak”!

Three probable ranges of calibrated age:

1959

1962

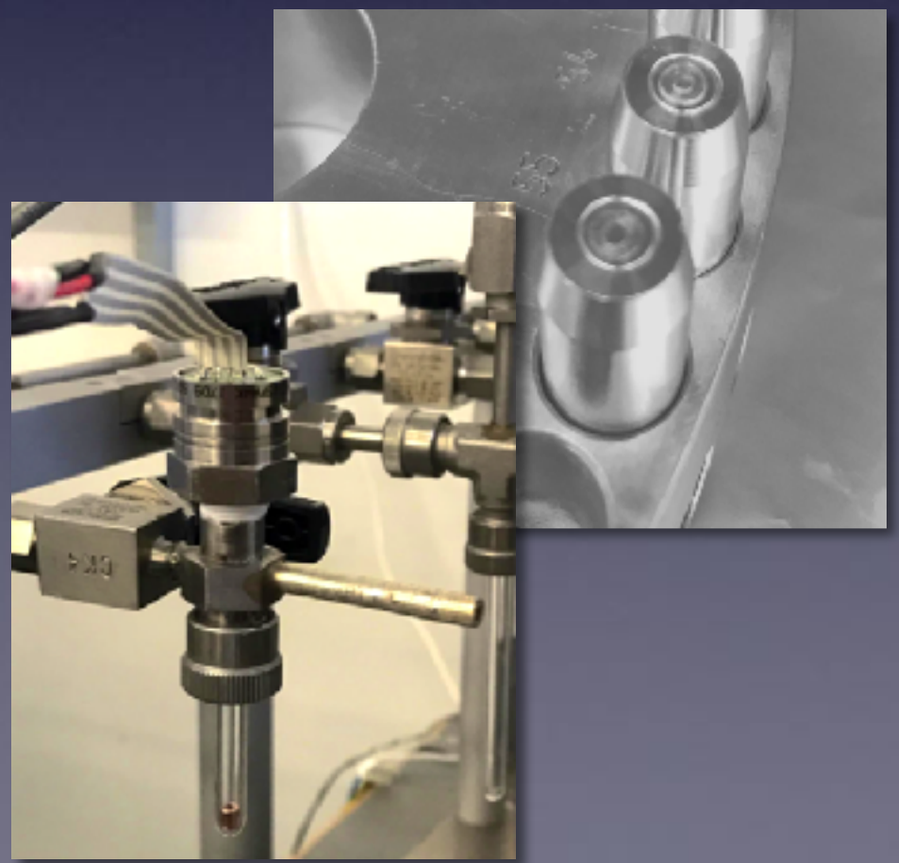
1979–1980



The plant from which the canvas was made was cut after Léger death

Where the research is going to?

- IBA and PIXE in particular:
  - Small and compact accelerators to be transportable (also to be installed in national restoration facilities)
    –> MACHINA project (INFN & CERN):
    acceleration is achieved through radio-frequency quadrupole technology
    (giuntini@fi.infn.it - ftaccetti@fi.infn.it)
- ^{14}C-AMS:
  - Smaller terminal voltages
  - Single stage AMS
  - Smaller samples
    –> graphite samples down to few tens of μg
  - Handling of big data –> Ariadne+



Thanks to all colleagues from LABEC and CHNet
(in particular Lucia Liccioli e Serena Barone)

...and many thanks to you!

fedi@fi.infn.it
https://chnet.infn.it