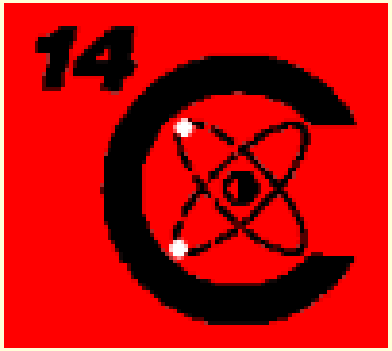
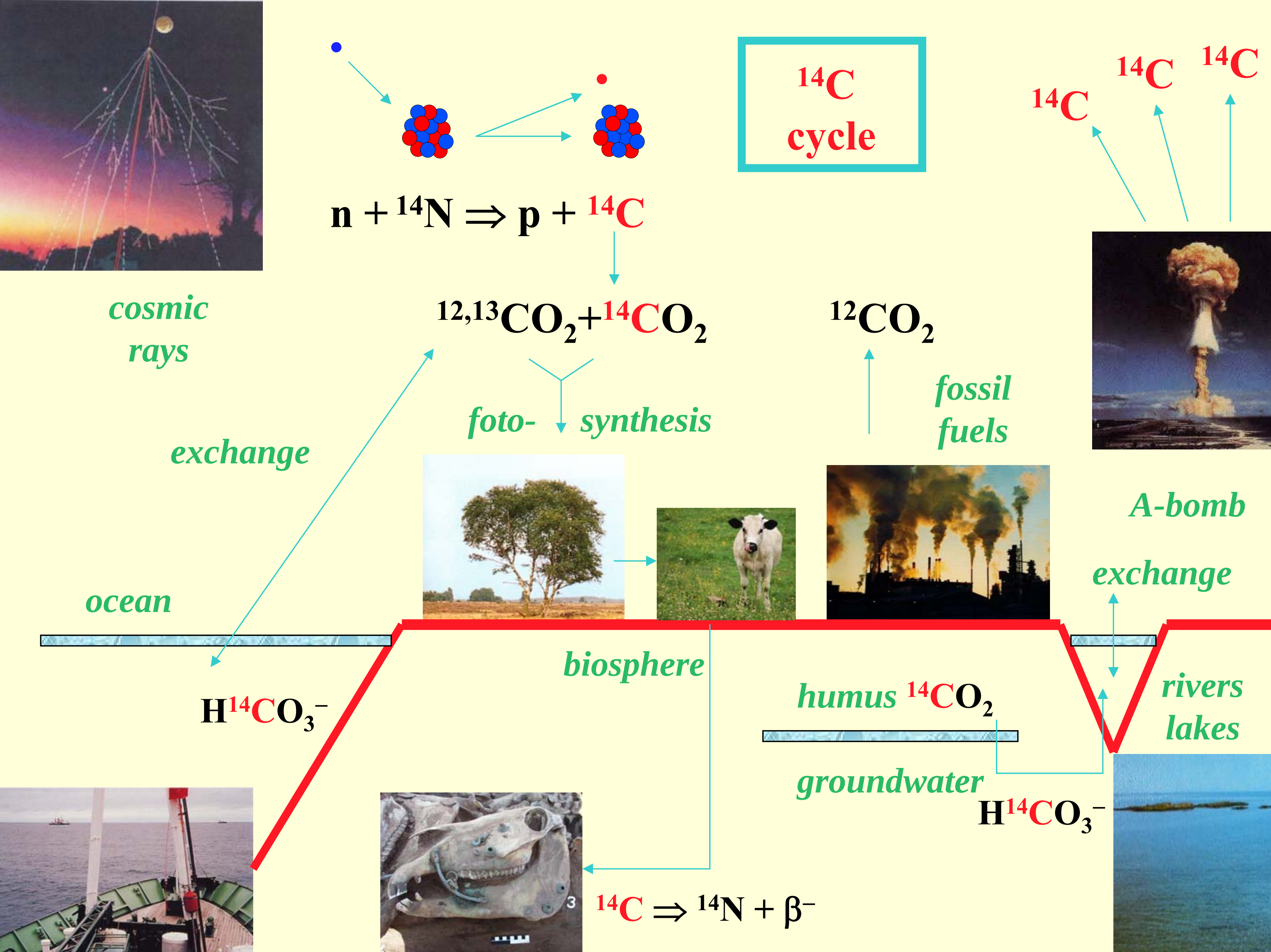
AMS: Accelerator Mass Spectrometry

- **detection of rare isotopes with ultralow abundance**
- **mass spectrometry using an accelerator**
- **application of nuclear physics into many other fields**



- *archaeology*
- *quaternary geology*
- *art*
- *ocean sciences*
- *physics*
- *atmospheric sciences*
- *cryology*
- *chemistry*
- *hydrology*
- *forensics*
- *biology*
- *environmental sciences*
- *religion*
- *astronomy*
- *medicine*
- *nuclear reactors*
- *food adulteration*
- *weapons inspection*
- *global carbon cycle*
- *planetary science*
- *sewer inspection*
- *climate*



^{14}C cycle
$n + {}^{14}N \Rightarrow p + {}^{14}C$
$^{12,13}CO_2 + {}^{14}CO_2$
$^{12}CO_2$
^{14}C
^{14}C
^{14}C
cosmic rays
foto- synthesis
fossil fuels
exchange
A-bomb
exchange
ocean
biosphere
$H^{14}CO_3^-$
humus $^{14}CO_2$
groundwater
rivers lakes
$H^{14}CO_3^-$
$^{14}C \Rightarrow {}^{14}N + \beta^-$

some ^{14}C numbers ...

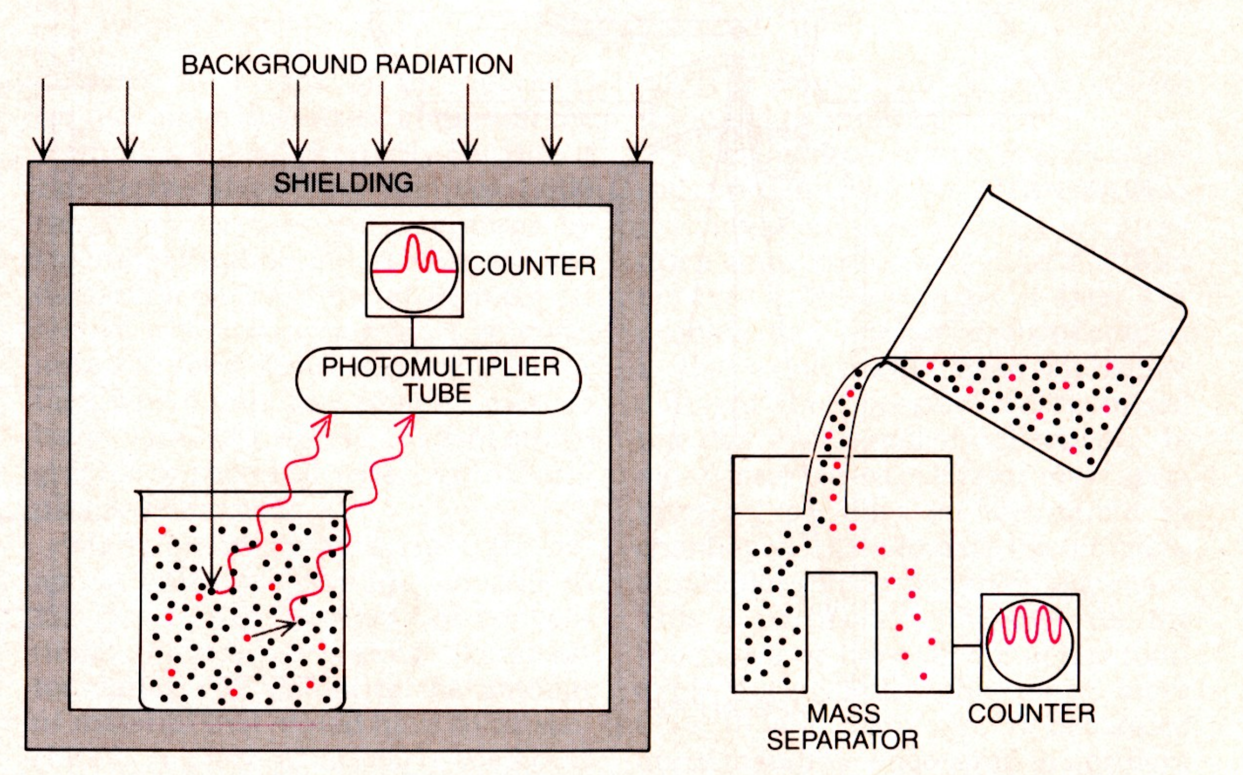
halflife	$T_{1/2} = 5730 \pm 40$ yr
natural abundance	$^{14}C/C = 1.2 * 10^{-12}$
detection limit	$^{14}C/C = 10^{-15}$
standard activity	226 ± 1 Bq/kgC $\equiv$ 13.56 dpm/gC
decay	β^-, $E_{max} = 156$ keV
natural production	2.4 ± 0.4 $^{14}C/cm^2s$

⇑

natural variation

^{14}C detection

(left)
radiometry
(right)
mass spectrometry



- **natural radioactivity is extremely low**
  < *natural background level*
- **E(β^-) is very low** ***difficult detection***
- **concentration is extremely low**
  $^{12}C:^{13}C:^{14}C = 1:0.01:10^{-12(15)}$

^{14}C - radiometry vs. AMS

$dN/dt = -\lambda N$ decay counting vs. atom counting

5‰ precision = 4.10^4 counts ⇒ $\sqrt{N}/N = 0.005$

radiometry:

15 dpm/gC, t_c = 48 hrs, 1 gC

1 mgC would take 7 yrs counting time

AMS:

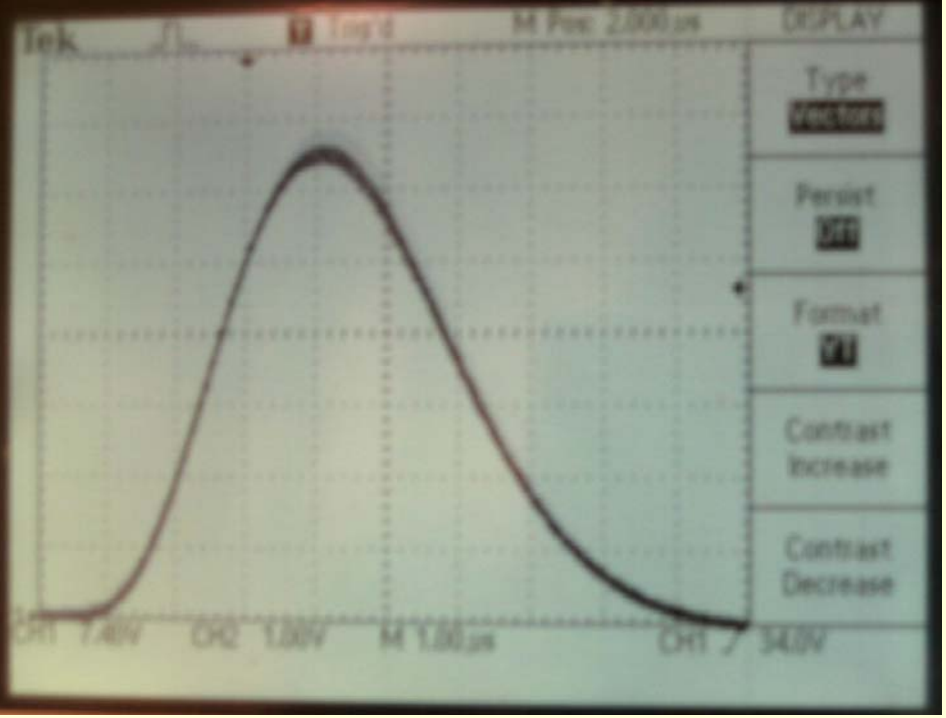
efficiency 10^{-2} ⇒ 4.10^6 atoms ^{14}C needed for 5‰

abundance 10^{-12} ⇒ 4.10^{18} atoms C = 8.10^{-5} g

10% used in source ⇒ typ. 1 mg sample size

1 hour counting time (50-100 Hz ^{14}C)

zepto (10^{-21}) to atto (10^{-18}) mol (^{14}C/mgC)

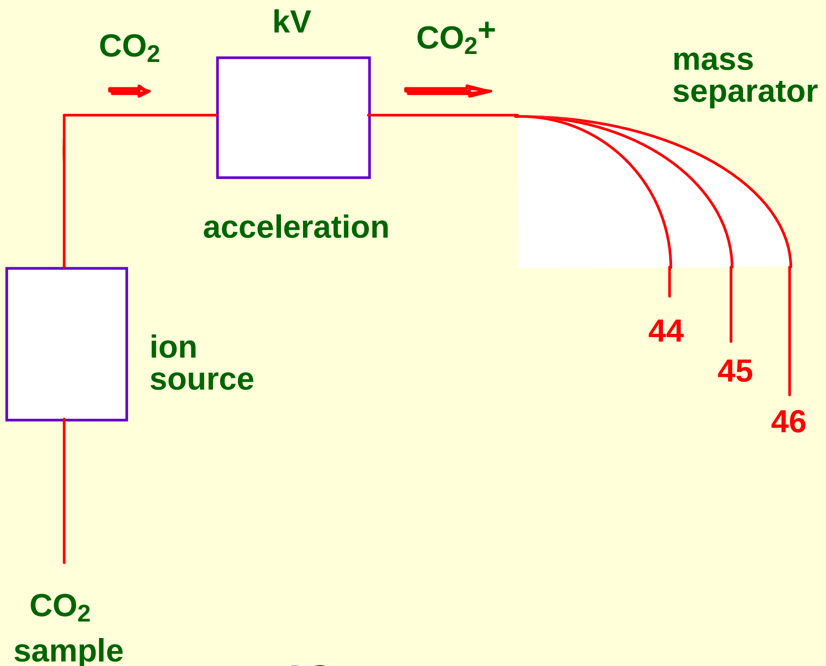


<u>AMS efficiency:</u>

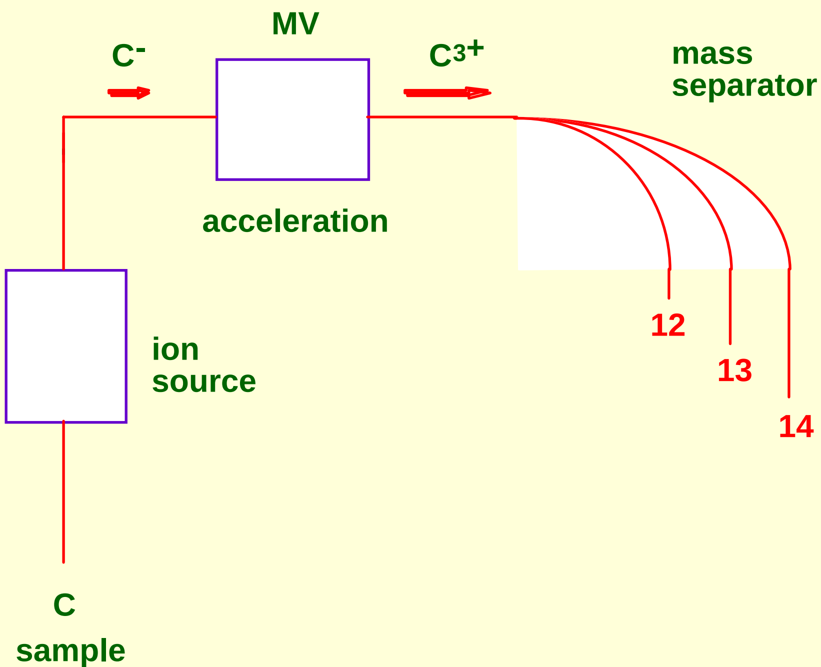
modern sample
ca. 40/sec. for 10^{-12} abundance

background
ca. 10/min. for 10^{-15} abundance

mass spectrometry basics



^{13}C mass spectrometer



^{14}C mass spectrometer

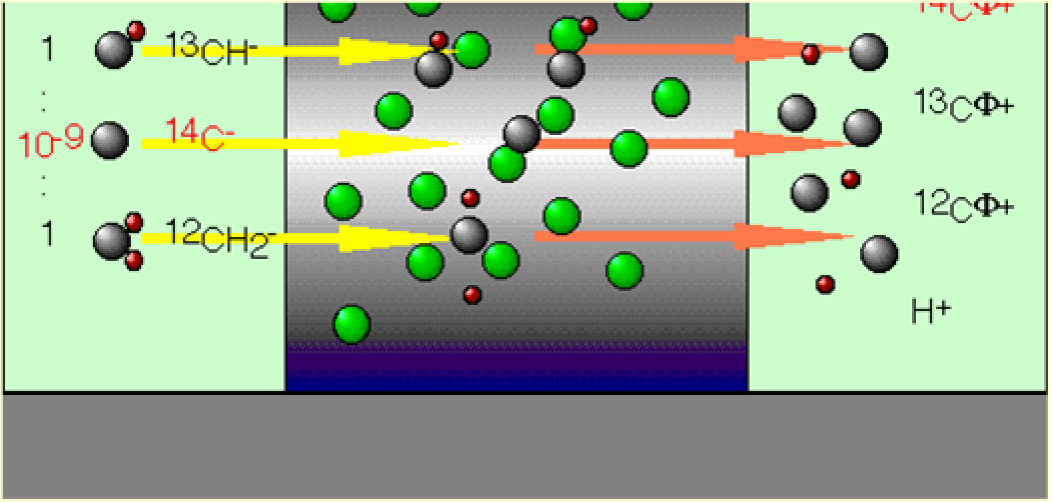
44	$^{12}C^{16}O_2$
45	$^{13}C^{16}O_2$
46	$^{14}C^{16}O_2$

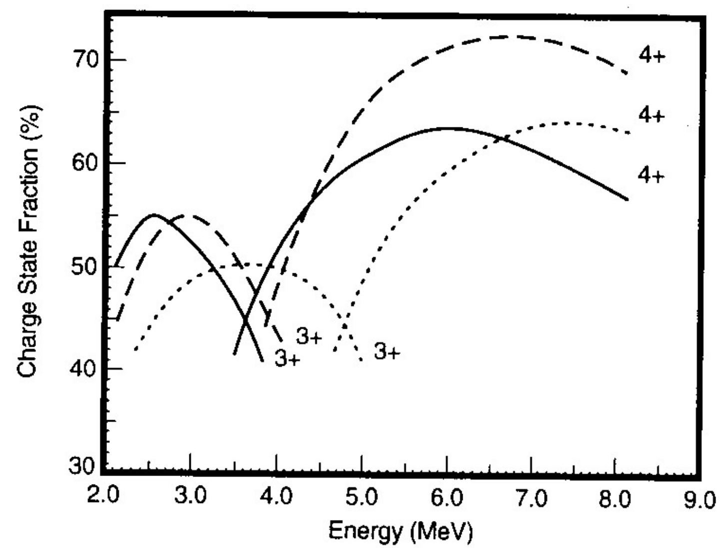
isobars: $^{12}C^{16}O^{18}O$ *etc.*

12	^{12}C
13	^{13}C
14	^{14}C

isobars: ^{14}N, $^{12}C^{1}H_2$, $^{13}C^{1}H$ *etc.*

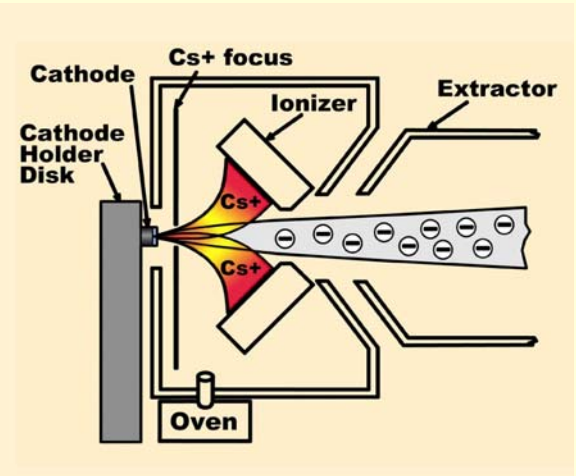
QuickTime™ en een Photo - JPEG decompressor zijn vereist om deze afbeelding te bekijken.





Ion Source & stripper

- Cs sputtering negative ions
- gasstripper optimum 2.5 MV for $^{14}C^{3+}$



solve isobar problems:

- negative ^{14}N not stable !
- stripping destroys mass 14 molecules: $^{12}CH_2$, $^{13}CH^-$, ...

measure $^{12,13,14}C$ ranging 1-10^{-15}

Injection

a) bouncing: pulse injection magnet

sequential injection - 0.5s ^{12}C, 0.5s ^{13}C, 1s ^{14}C

♦ different conditions for each isotope

b) recombinator:

simultaneous injection - ^{12}C, ^{13}C, ^{14}C

♦ removes unwanted negative ions from source

♦ allows $^{13}\delta$ measurement - *fractionation correction*

♦ ^{12}C chopped (≈1%) - *^{12}C,^{13}C beams same intensity*

♦ requires more "cleanup" after accelerator

^{14}C not alone through machine

fractionation / stability ESSENTIAL for ^{14}C

$^{14}C/^{12}C$ < 5‰ is a MUST

High Energy Mass Spectrometer

1st magnet

- separates $^{12,13,14}C$

E.S.A.

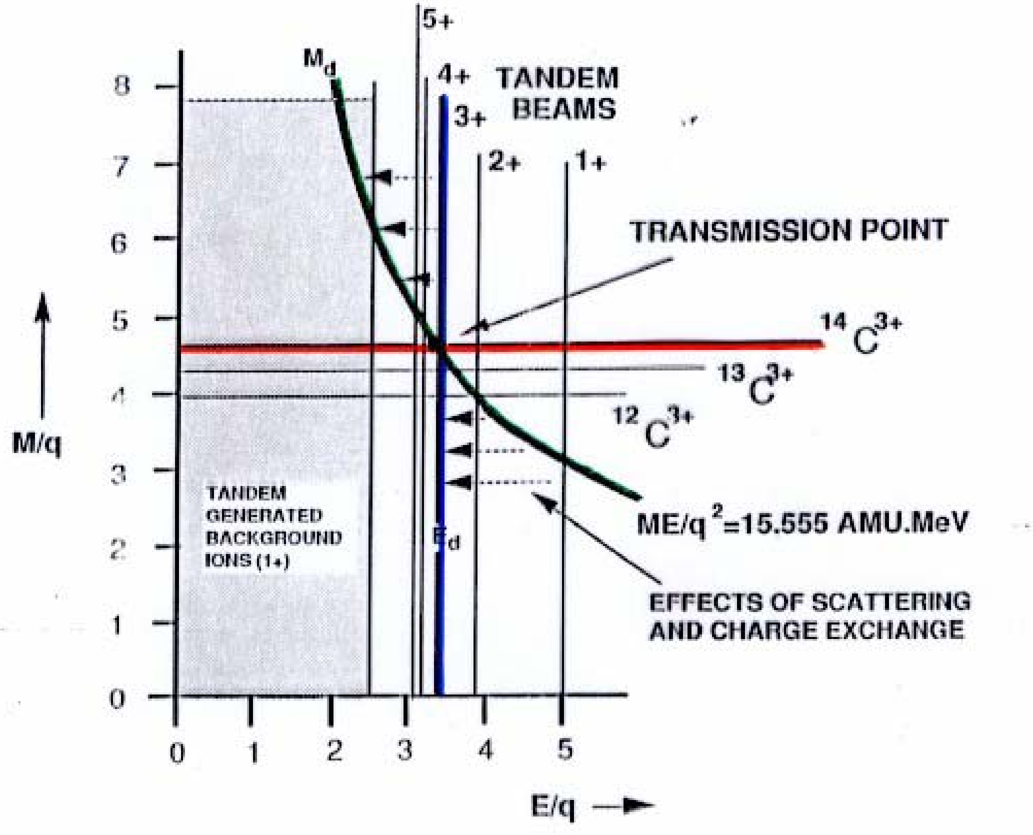
- removes $^{12}C^{3+}$,$^{13}C^{3+}$ with energies such that they en up on ^{14}C path
- removes ME/q^2 ambiguities
- $^{7}Li_2^{+}$,$^{12}CH_2^{+}$,$^{13}CH^{+}$,$^{12}C^{16}O_2^{+}$ have $M/q=14$

2nd magnet

- removes particles scattered in ESA

detector

- foil separates N and C ions ($^{14}N^{3+}$ from NH^{-})

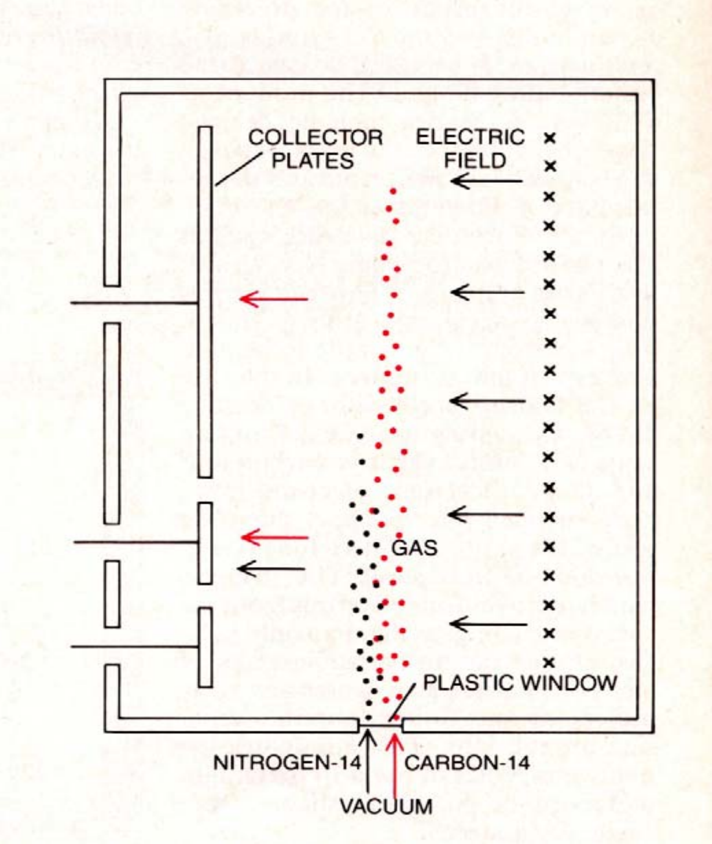


$^{14}C^{3+}$

10 MeV

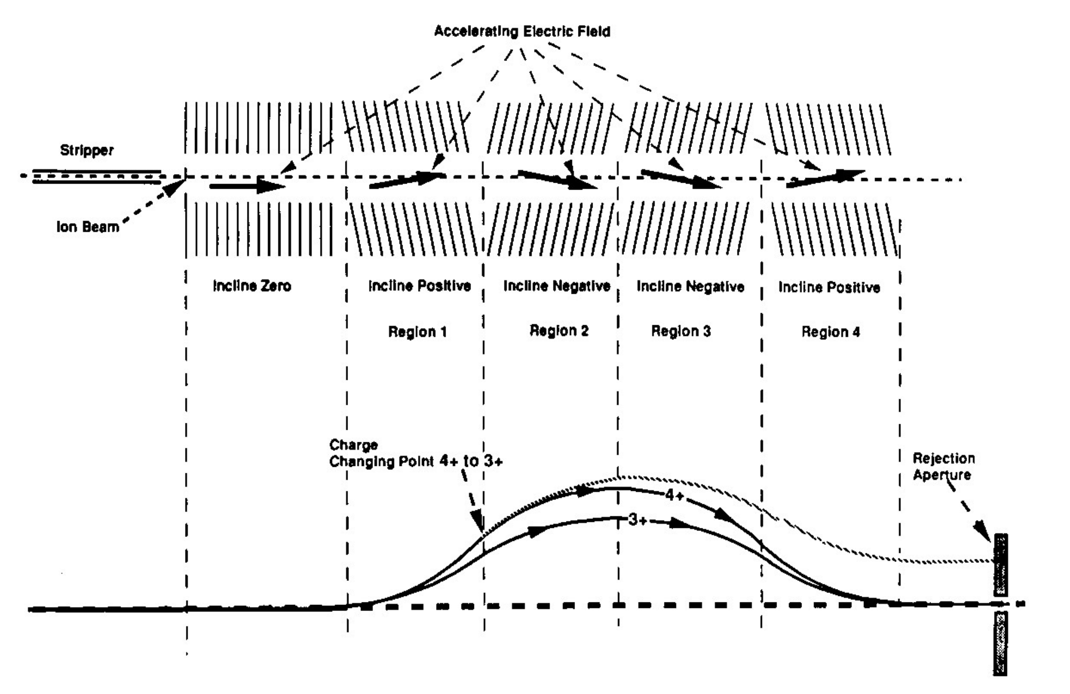
M/q = 14/3 *unique*

ME/q^2 *magnet* **E/q *electrostat***



particle detector *ionisation chamber*

the Groningen 2.5 MV ^{14}C Tandetron



er

background struggles

ME/q^2 ambiguities for 10 MeV $^{14}C^{3+}$ $^{12,13}C^{3+}$ which leave stripping canal as 4^+ and pick up electron to become 3^+ ⇒ $^{12,13}C^{3+}$ 10-12.5 MeV background reduction: electrode *inclination* in tube

Multiple charge exchange in "vacuum" residual gas $^{13}C^{3+}$ ⌫ $^{13}C^{2+}$

generation 1: large (5-15 MV)
development AMS (1978)
Tandem / VandeGraaff
all cosmogenic isotopes

generation 2: small (2-3 MV)
dedicated ^{14}C (^{10}Be) "tandetron"
2a - bouncer (1980's)
2b - recombinator (1990's)
automation: mass spectrometry practice

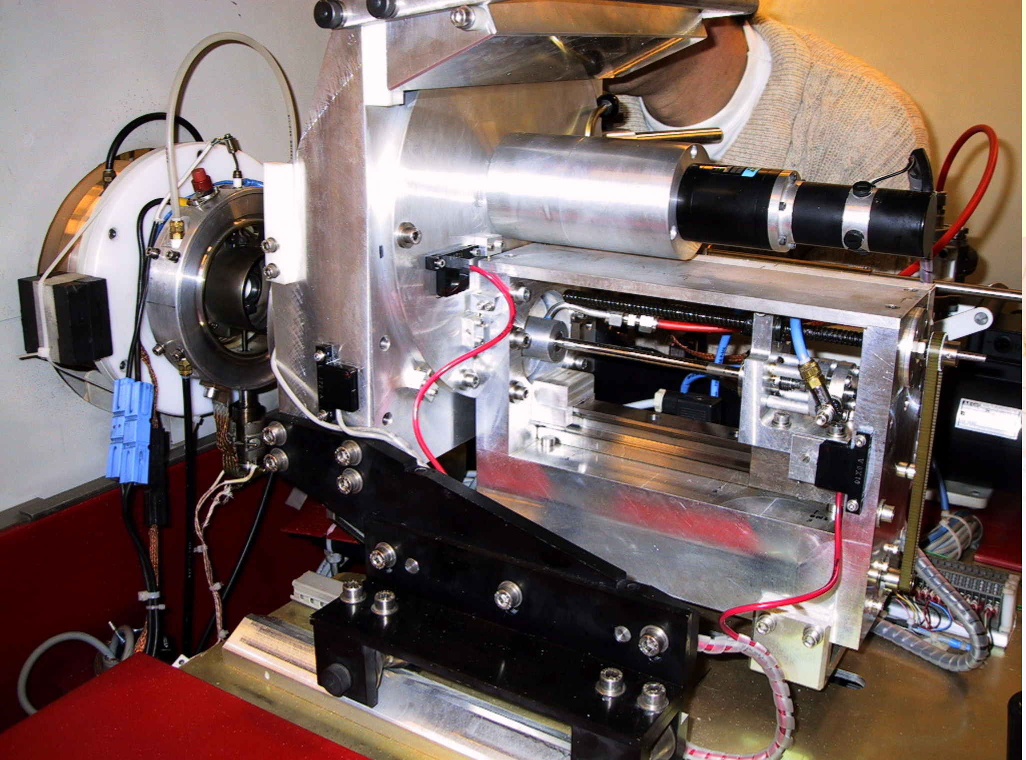
generation3 : baby (≤ 0.5 MV)
... since 2002 "tandy"

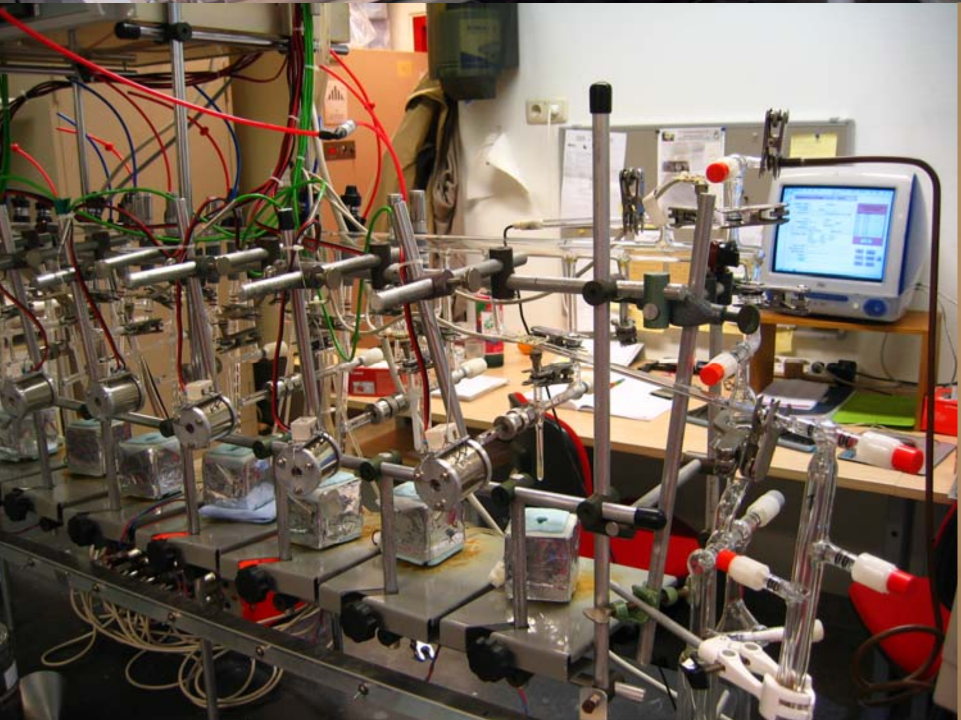
AMS accelerators

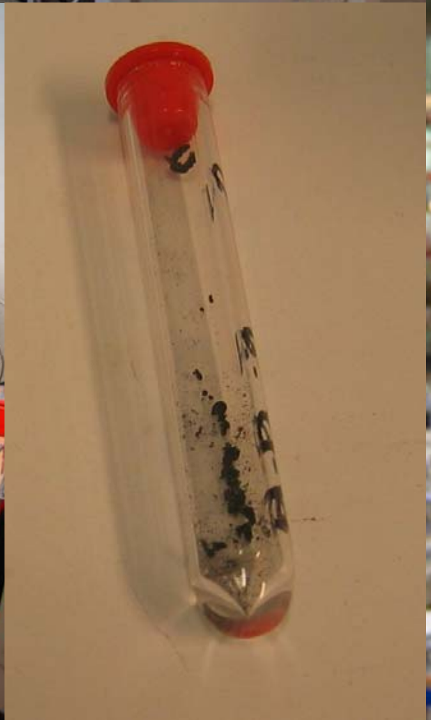
Groningen, NL ⇑

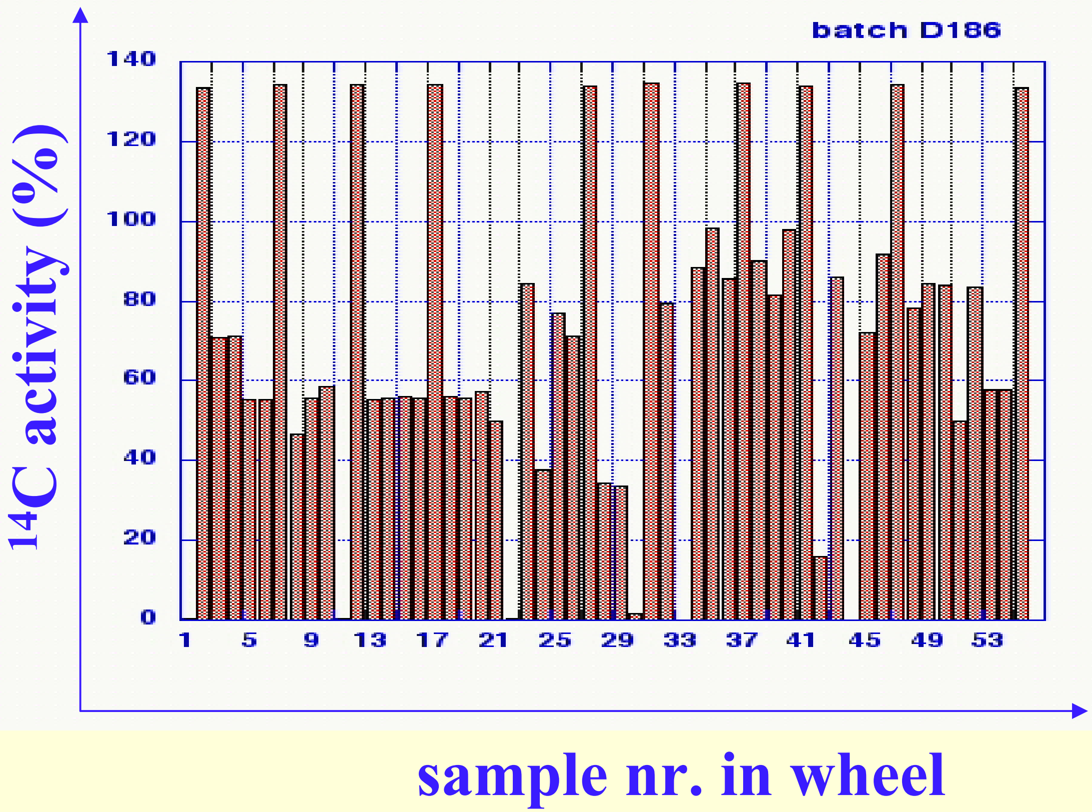
⇑ Rehovot, IL

Zürich, CH ⇒

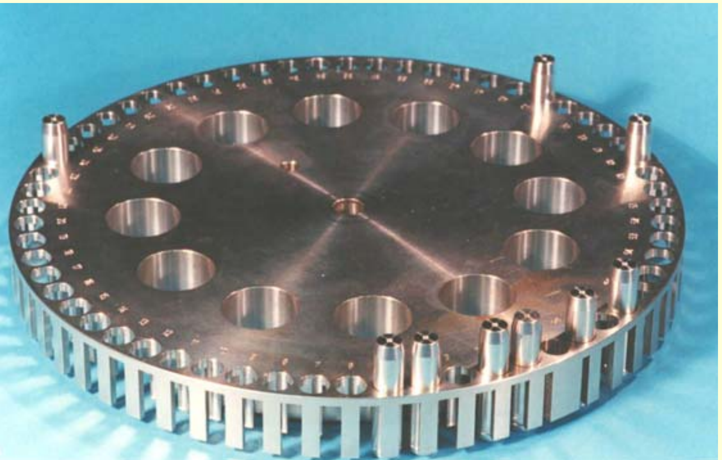


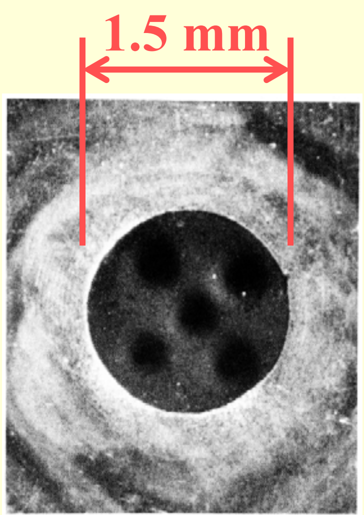


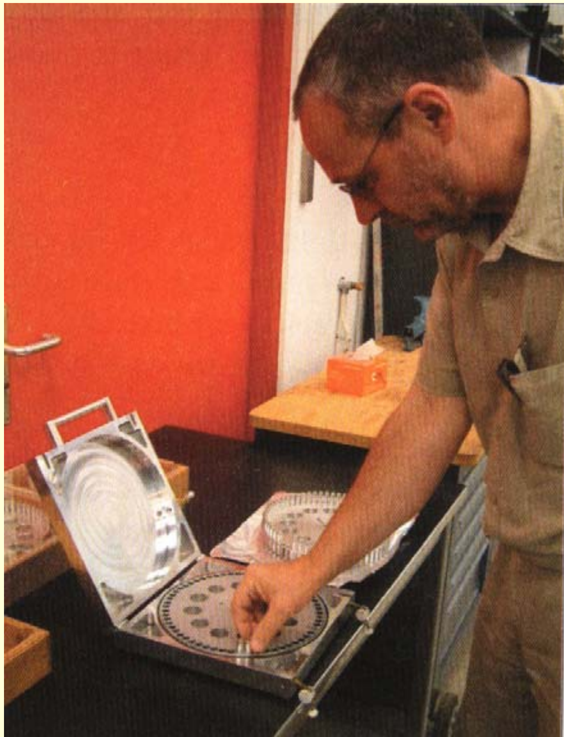


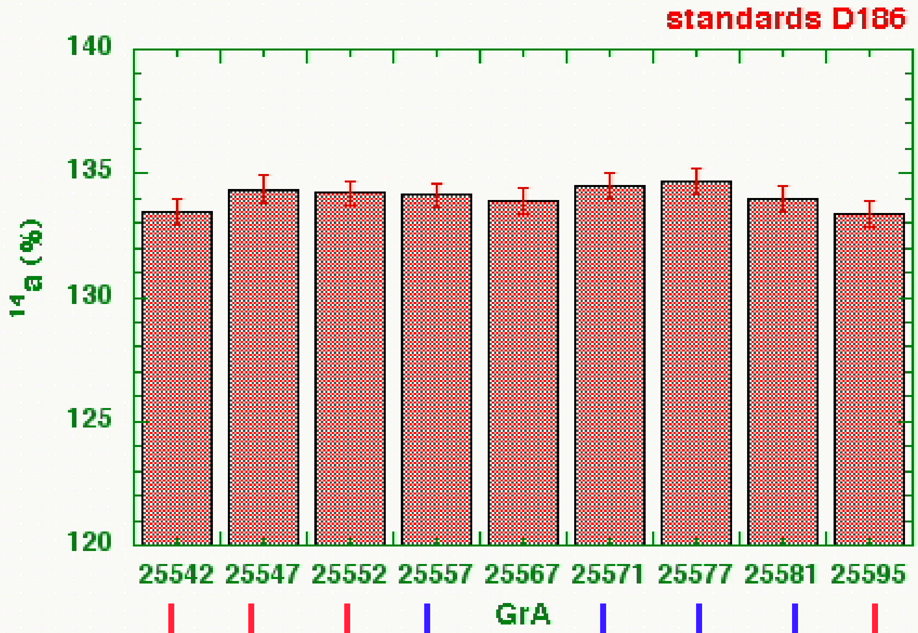


target wheel
batch D186
AMS









4‰

standards AMS

Oxalic Acid II

$^{14}a = 134.06$ %

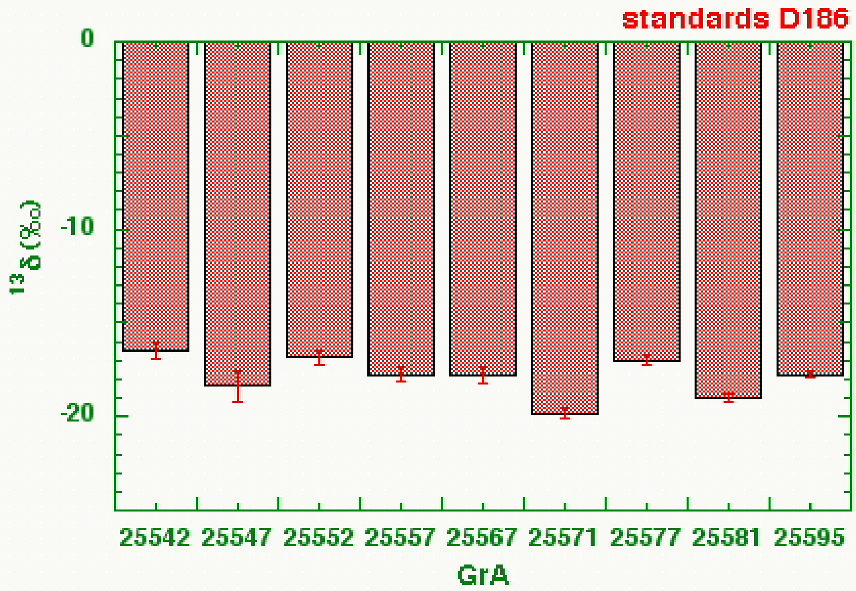
$^{13}\delta = -17.8$ ‰

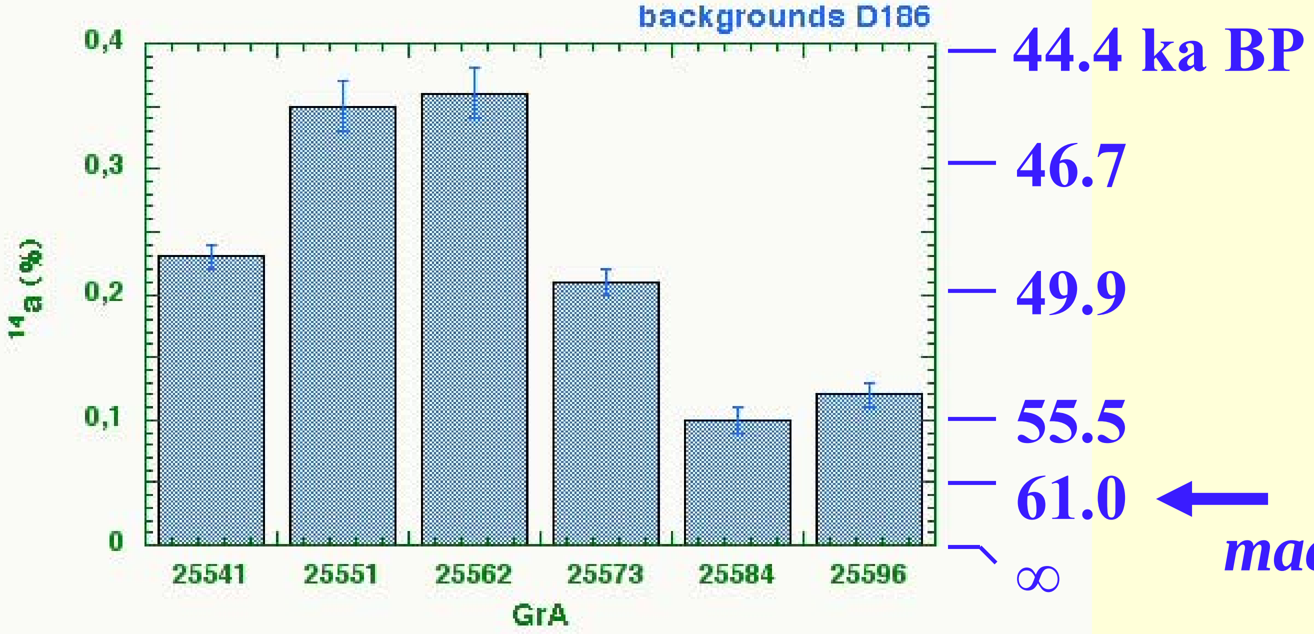
"setting" values

combustion & graphite lines <u>AMS</u>

large volume CO_2 gas <u>conventional</u> combustion AMS graphitisation

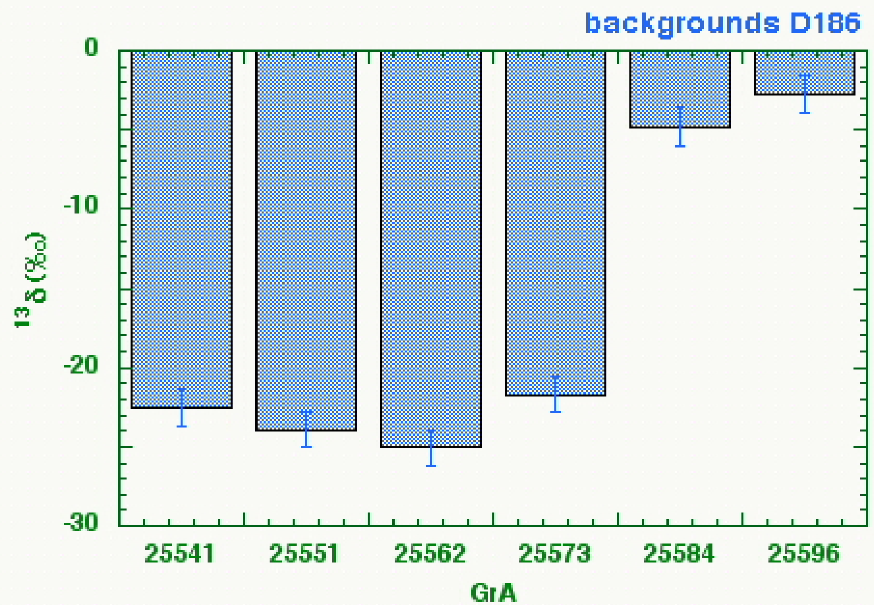
2 labs intercomparison



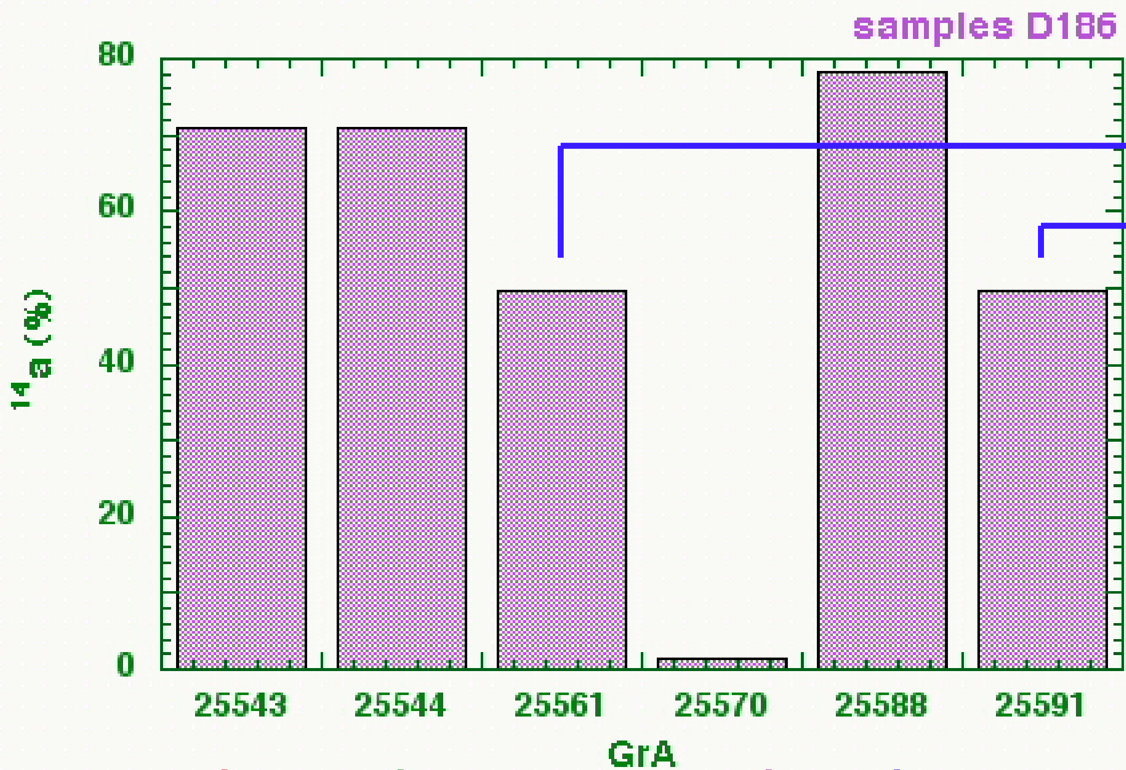


backgrounds AMS

machine blank > 60 ka (not shown)



anthracite
$^{13}\delta$ = 23.18 ‰
combustion
natural gas
$^{13}\delta$ = -3.14 ‰
graphitisation



49.67±0.26%
49.74±0.25%

known age sample
("working standard")
IAEA-C7 49.53±0.12%
5645±20 BP
quality check

samples
AMS

seeds, Iron Age, Israel	2770±35 BP
seeds, Iron Age, Israel	2740±35 BP
bone, Palaeolithic, North Sea	35160^{+330}_{-300} BP
textile, Qumran, Israel	1975±35 BP

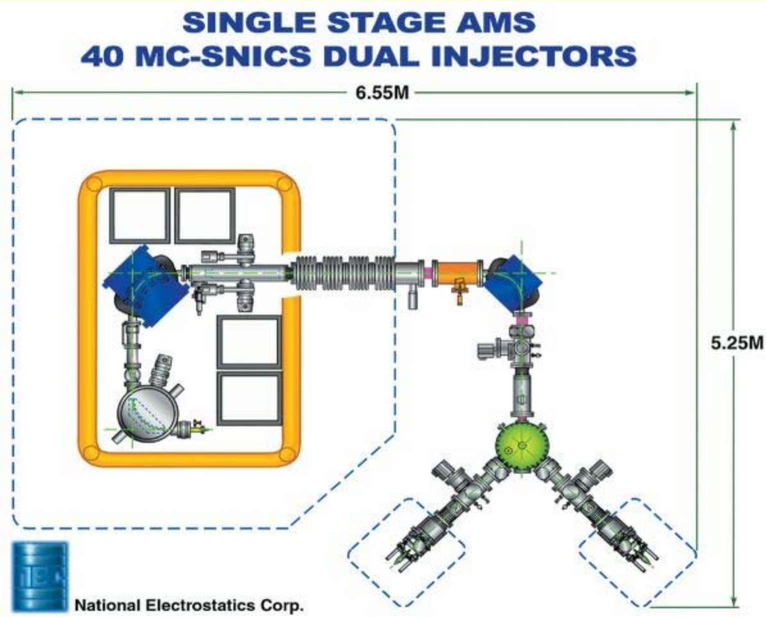
Latest development: "baby-AMS"

- single-stage AMS
- 250 kV HV deck

  AMS without the "A"
- molecular dissociation

  $^{14}C^{1+}$ background problems

  2 turbopumps 250 l/s



cosmogenic isotopes by AMS

	^{10}Be	^{14}C	^{26}Al	^{36}Cl	^{41}Ca	^{129}I
halflife (yr)	1.6×10^{6}	5730	7.0×10^{5}	3.0×10^{5}	10^{5}	16.10^{6}
origin	spallation N,O	^{14}N(n,p)	^{28}Si(μ,2n)	spallation Ar	^{40}Ca(n,γ) spall. Fe	spall. Xe
abundance	10^{-9}	10^{-12}	10^{-14}	10^{-12}	10^{-14}	10^{-12}
stable isotope	^{9}Be	^{12}C,^{13}C	^{27}Al	^{35}Cl,^{37}Cl	^{0}Ca	^{127}I
stable isobar	^{10}B	^{14}N	^{26}Mg	^{36}Ar,^{36}S	^{41}Ar	^{129}Xe
terminal (MV)	3	2.5	7.5	8	(linac)	5
charge state	3	3	7	7	10	5
energy (MeV)	12	10	60	4	200	30
chem.form	BeO	C	Al_2O_3	AgCl	CaH_3	AgI

the mother of all natural isotopes

^{14}C clock problems

1. halflife $T_{1/2}$ has been changed

$T_{1/2}$ = 5730 ± 40 yr; originally 5568 yr has been used

2. the ^{14}C content in de nature is not constant

1. ^{14}C production depends on cosmic ray flux, which depends on solar activity and earth magnetic field strength

2. changes in equilibrium between the C reservoirs
atmosphere, biosphere, ocean, soil

3. isotope effects change the ^{14}C content

***example:* photosynthesis is mass dependent - plant is depleted in ^{14}C (and therefore seems older)**

4. reservoir effects

water (sea, river) contains dissolved fossil C and is thus depleted in ^{14}C - organisms living in water are therefore older

consequence:

- *the ^{14}C clock ticks at a different pace than the calendar*

(because of halflife)

- *this pace changes continuously*

(because of changing natural ^{14}C content)

- *the ^{14}C clock starts at different moments for different materials*

(because of isotope - en reservoir- effects)

solution:

- *define the ^{14}C clock speed*

w.r.t. standard activity = 1950

use $T_{1/2}$ = 5568 jr (original)

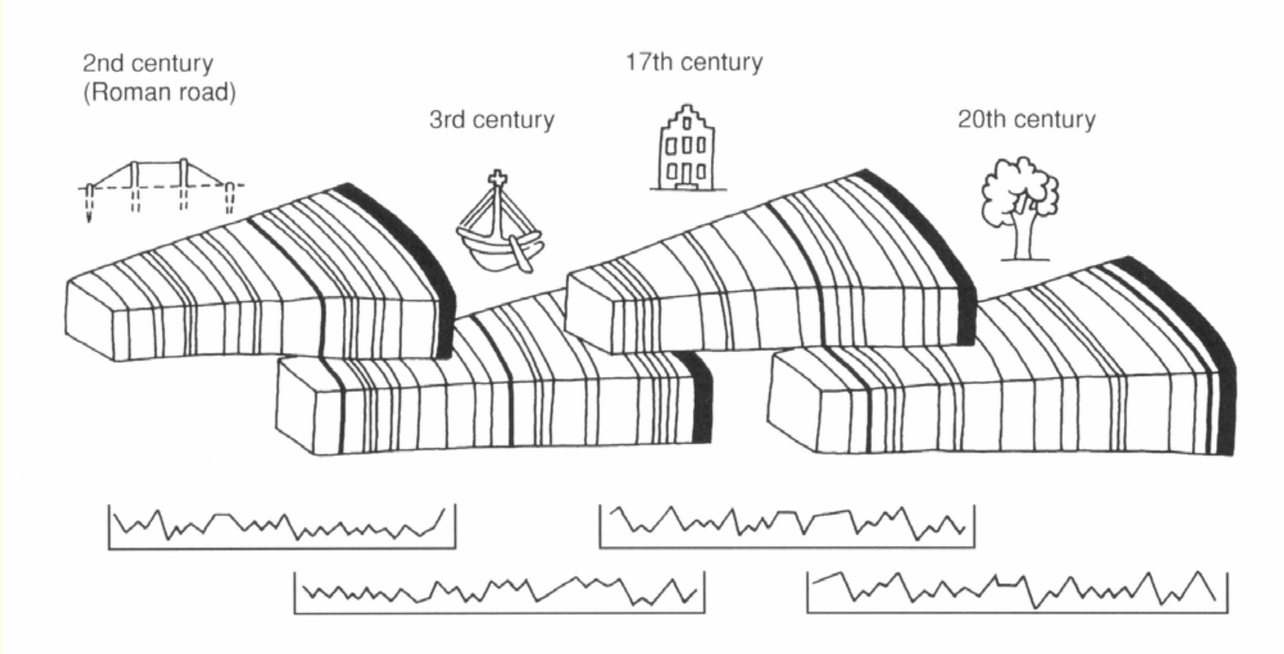
- *correct for isotope effects*

using stable isotope ^{13}C: $^{14}\delta = 2^{13}\delta$

- *express in unit "BP"*
- *calibrate the ^{14}C clock*

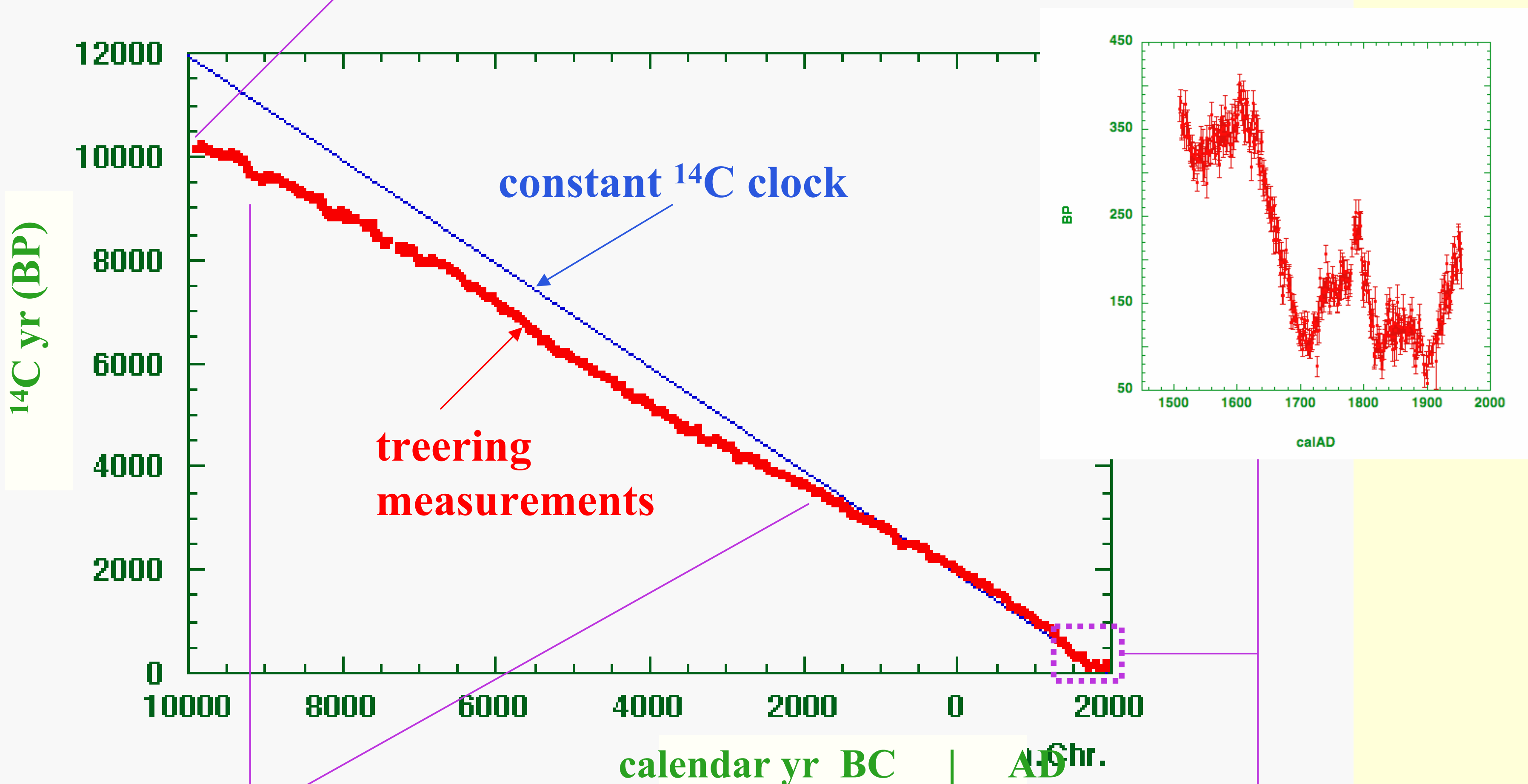
measure ^{14}C in absolutely dated materials (BP - AD/BC)

Dendrochronology



^{14}C calibration curve

10.000 ^{14}C years ago
12.000 calendar years ago
more ^{14}C in nature than present



long term trend:
geomagnetism

medium- & short term effects:
solar activity &
exchange ocean/atmosphere

intcal04

constructed curve, "decadal" (10 yr) resolution

statistic model, taking into account uncertainties in both ^{14}C and "calendric" parameters

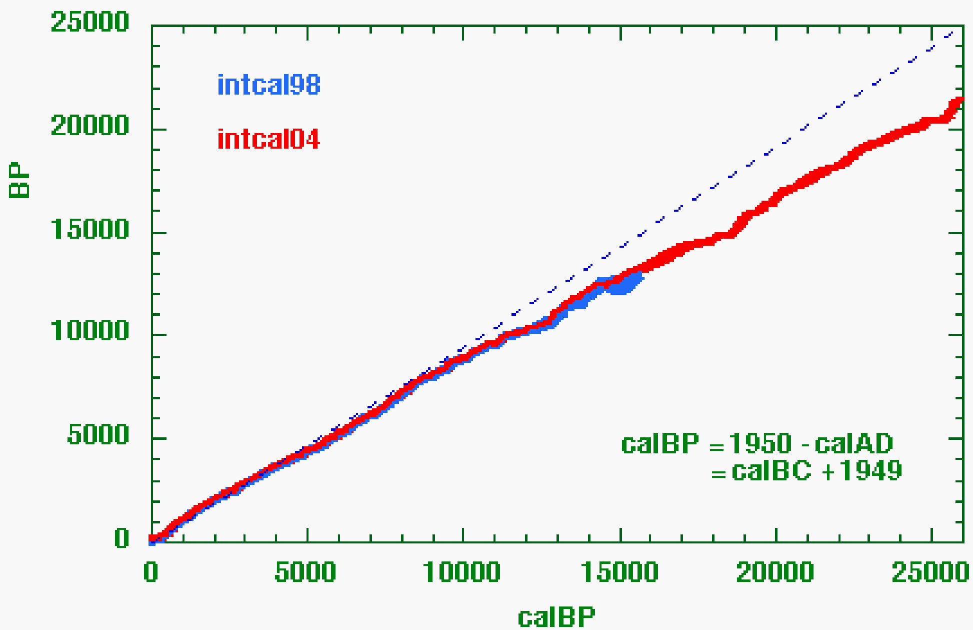
3 multi-author papers ***Radiocarbon 46, 3, 2004***

Reimer *et al.*	**intcal04**	**0-26 ka calBP terrestrial curve**
Hughen *et al.*	**marine04**	**0-26 ka calBP marine curve**
v.d.Plicht *et al.*	**notcal04**	**26-50 ka calBP comparison**

www.radiocarbon.org

- **calibration datasets**
- **computer programs**
- **articles**

  (subscription needed)



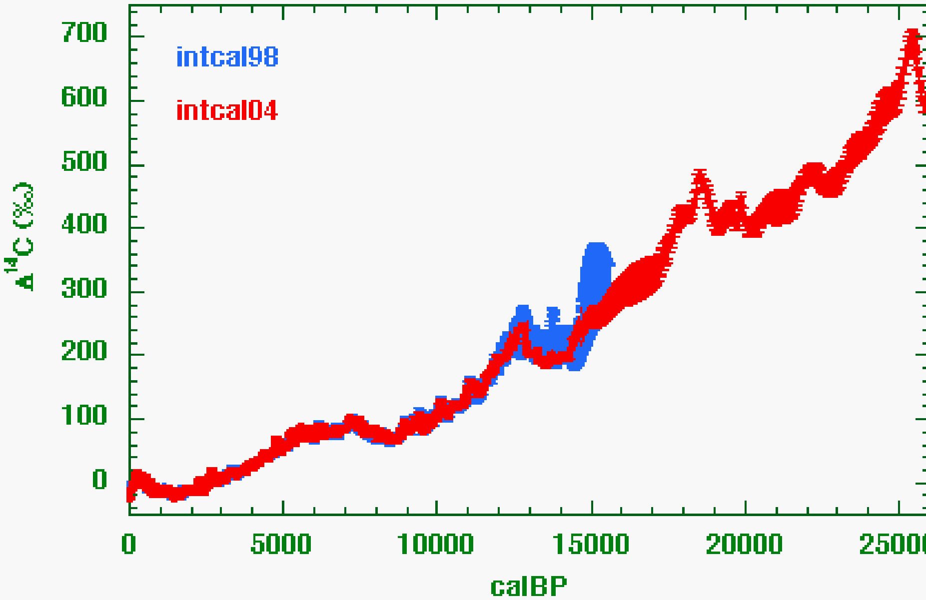
⇐ BP vs. cal BP

calibration curve

⇓ Δ^{14}C vs. cal BP

natural ^{14}C content

Wellington N.Z. 2003
^{14}C conference
intcal04
Radiocarbon 46, 3, 2004



^{14}C calibration 26-50 ka ?

0-26 ka

- **dendrochronology**

  absolute; only this is "calibration"

- **coral & marine layered sediments**

  ^{14}C reservoir effect; U-isotopes dated

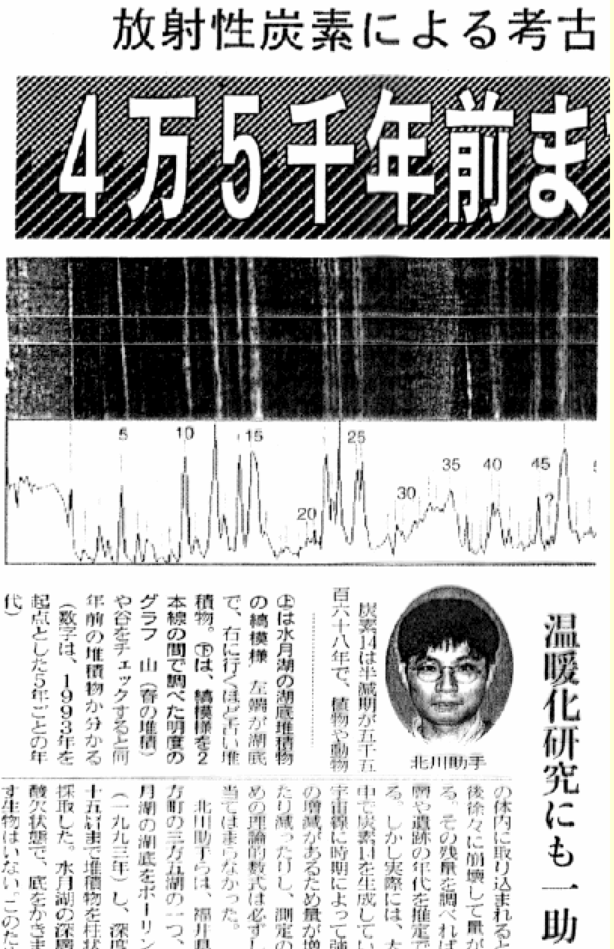
APPROVED by INTCAL working group

26-50 ka

- **layered sediments, speleothems, corals**

  each dataset has pros and cons

older $\Rightarrow$ larger measurement errors and uncertainties; data are not consistent calibration $\Rightarrow$ "comparison"


放射性炭素による考古

4万5千年前ま

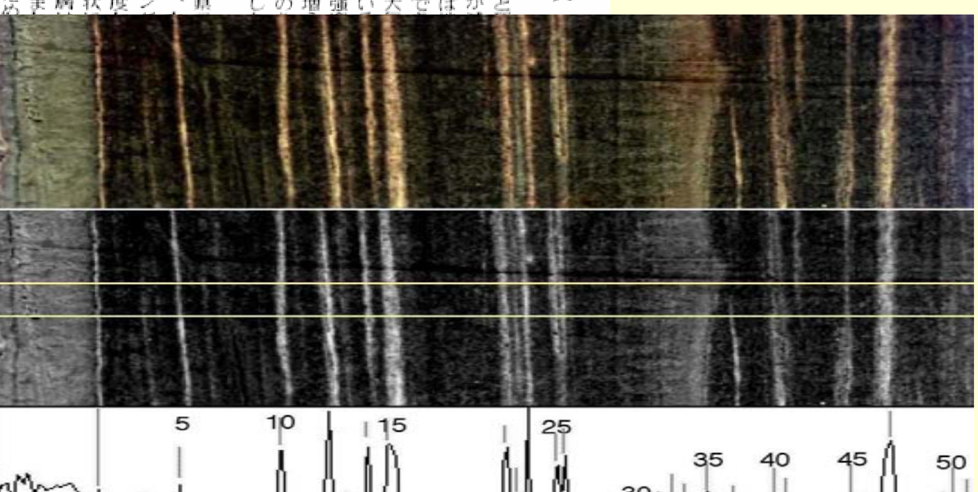
温暖化研究にも一助

炭素14は半減期が五千五
百六十八年で、植物や動物

北川助手

㊤は水月湖の湖底堆積物
の縞模様　左端が湖底
で、右に行くほど古い堆
積物。㊦は、縞模様を2
本線の間で調べた明度の
グラフ　山（春の堆積）
や谷をチェックすると何
年前の堆積物か分かる
（数字は、1993年を
起点とした5年ごとの年
代）

の体内に取り込まれると
後徐々に崩壊して量が
る。その残量を調べれば
骨や遺跡の年代を推定で
る。しかし実際には、大
中で炭素14を生成してい
宇宙線に時期によって強
の増減があるため量が増
たり減ったりし、測定の
めの理論的数式は必ずし
当てはまらなかった。
　北川助手らは、福井県
方町の三方五湖の一つ、
月湖の湖底をボーリン
（一九九三年）し、深度
十五㍍まで堆積物を柱状
採取した。水月湖の深層
酸欠状態で、底をかきま
す生物はいない。このため



Lake Suigetsu, Japan

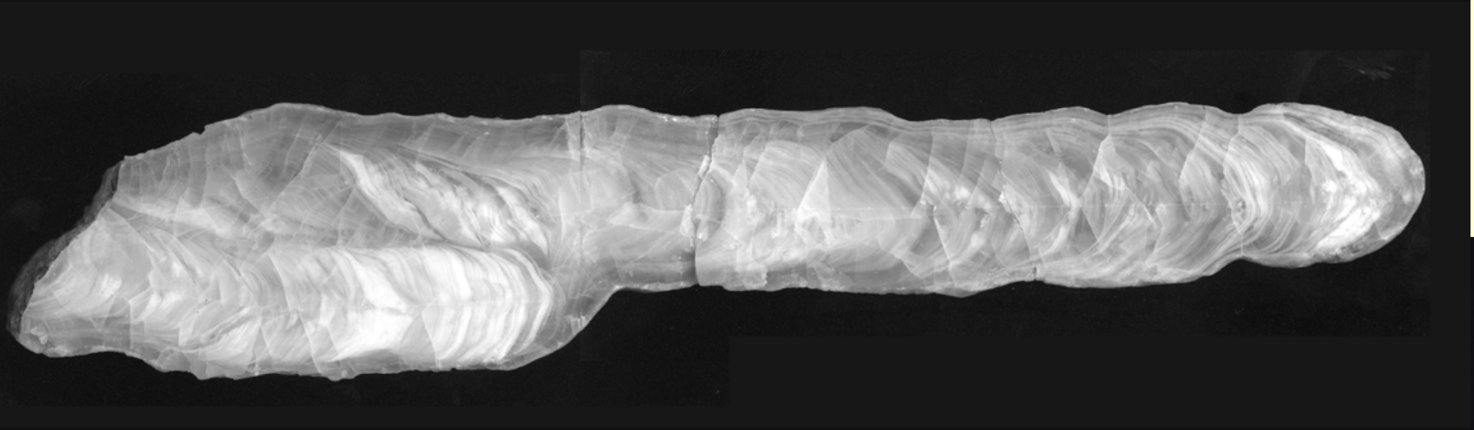
29.100 yr varved sediment

>330 AMS terrestrial samples

H.Kitagawa and J.van der Plicht

Science 279 (1998) 1187

Radiocarbon 42 (2000) 369



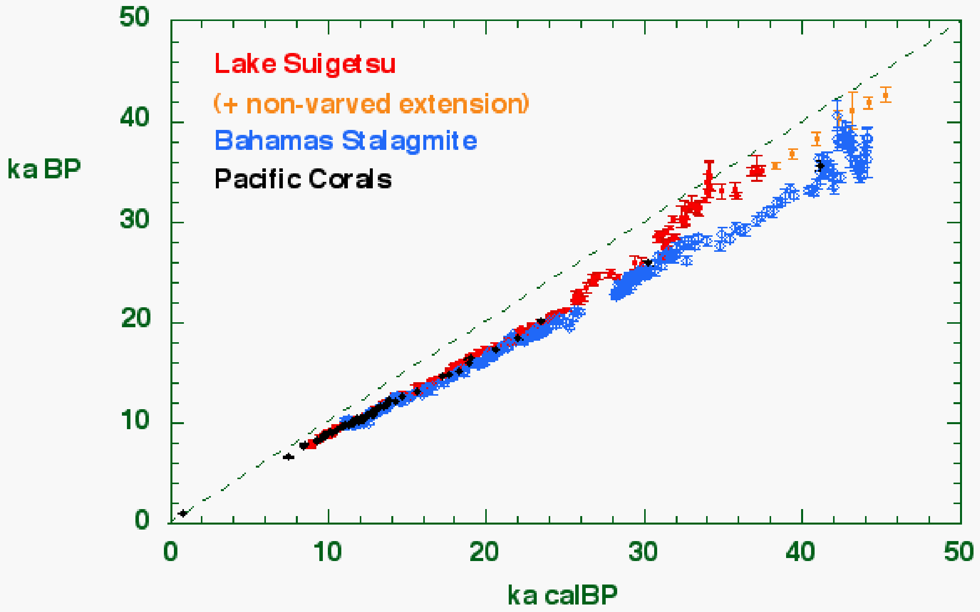
Speleothem, Bahamas

U/Th & ^{14}C dated

ca. 300 AMS carbonate samples

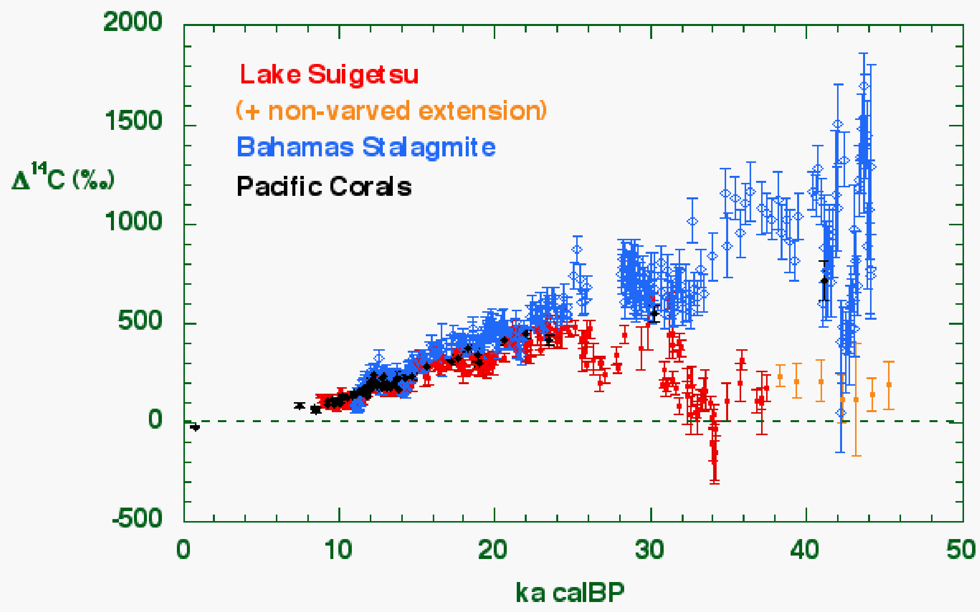
W.E.Beck et al.

Science 292 (2001) 2453



AMS-9 conference
Nagoya, Japan
september 2002
proceedings p. 353-358

4th symposium on
^{14}C & Archaeology
Oxford, UK
april 2002
proceedings p. 1-8



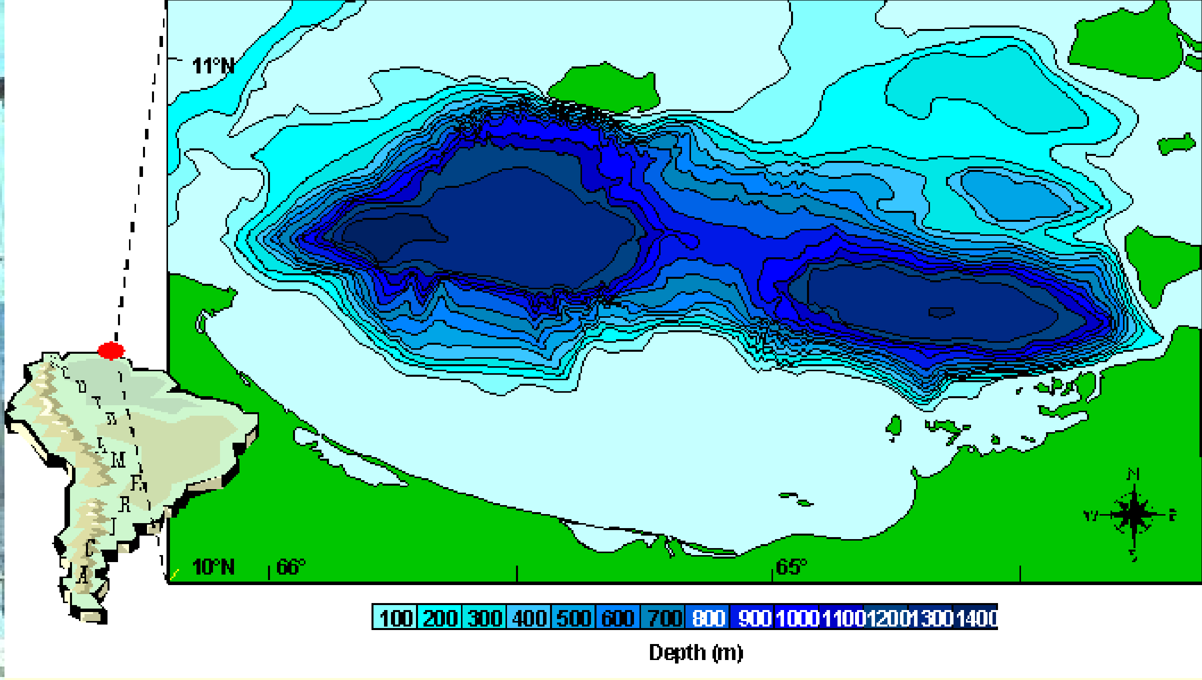
do <u>YOU</u>  believe in varves or in speleothems ?

each record has its plusses en minuses ...

Suigetsu	**BP :**	**terrestrial/atmospherih**	**plus**
	calBP :	**leyers (varves) counting**	**min**
		hiatuses, counting errors	
Bahamas	**BP :**	**reservoir correction ^{14}C**	**min**
		1470 ± 235 ^{14}C jr; constant?	
	calBP :	**U-series geochemistry**	**min**
		absolute ? hiatus at 27 ka	

- *calibration means "absolute" en "terrestrial / atmospheric"*
- *at least one of both records must be wrong*

needed: independent confirmation (or rebuttal)

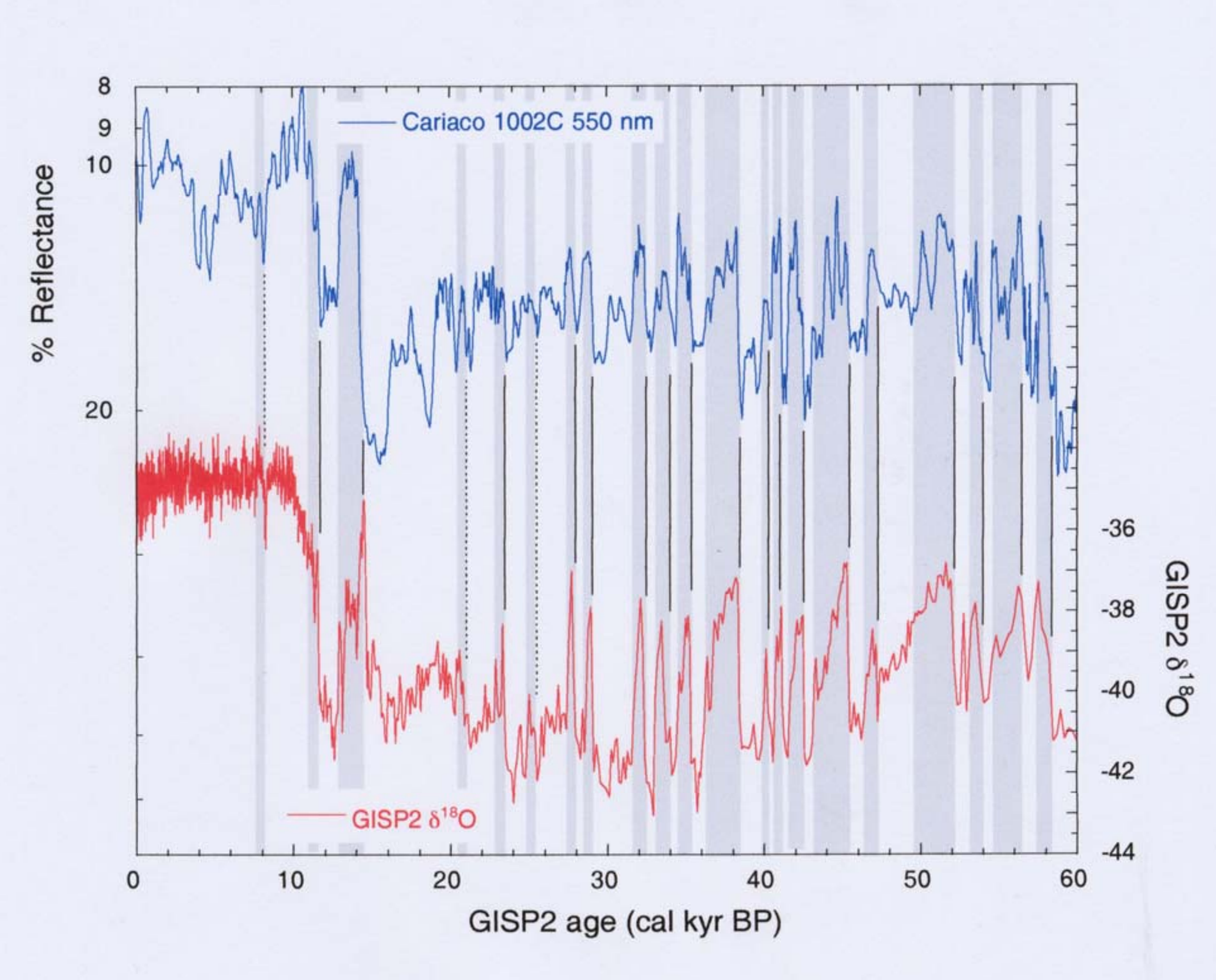


Cariaco Basin

coastal Venezuela

- **layered section (Late Glacial) used for Intcal04**
- **older part is *not* layered**

K.A.Hughen et al., Science 303 (2004) 202-207

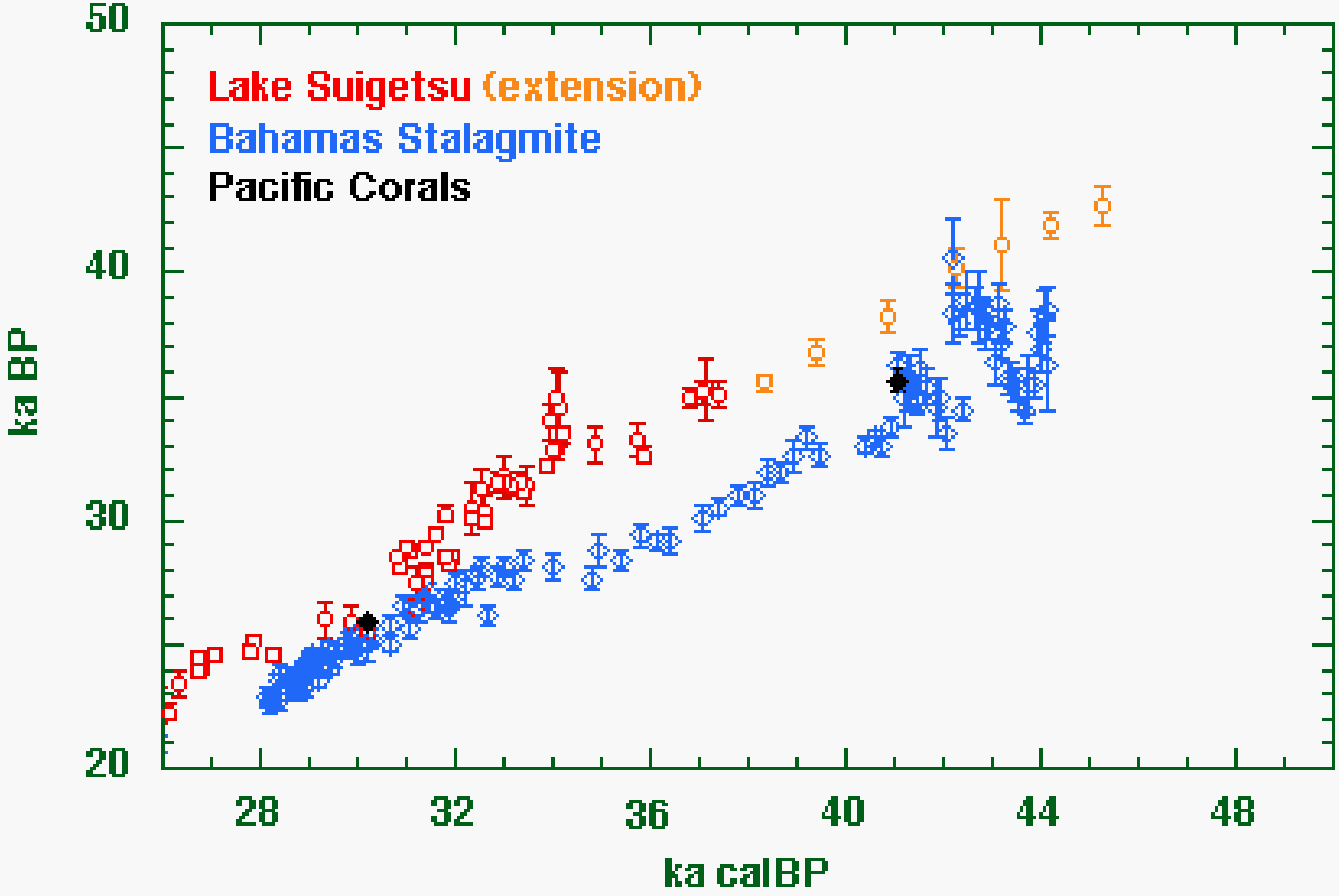


Cariaco **BP :** **foraminifera** **plus**

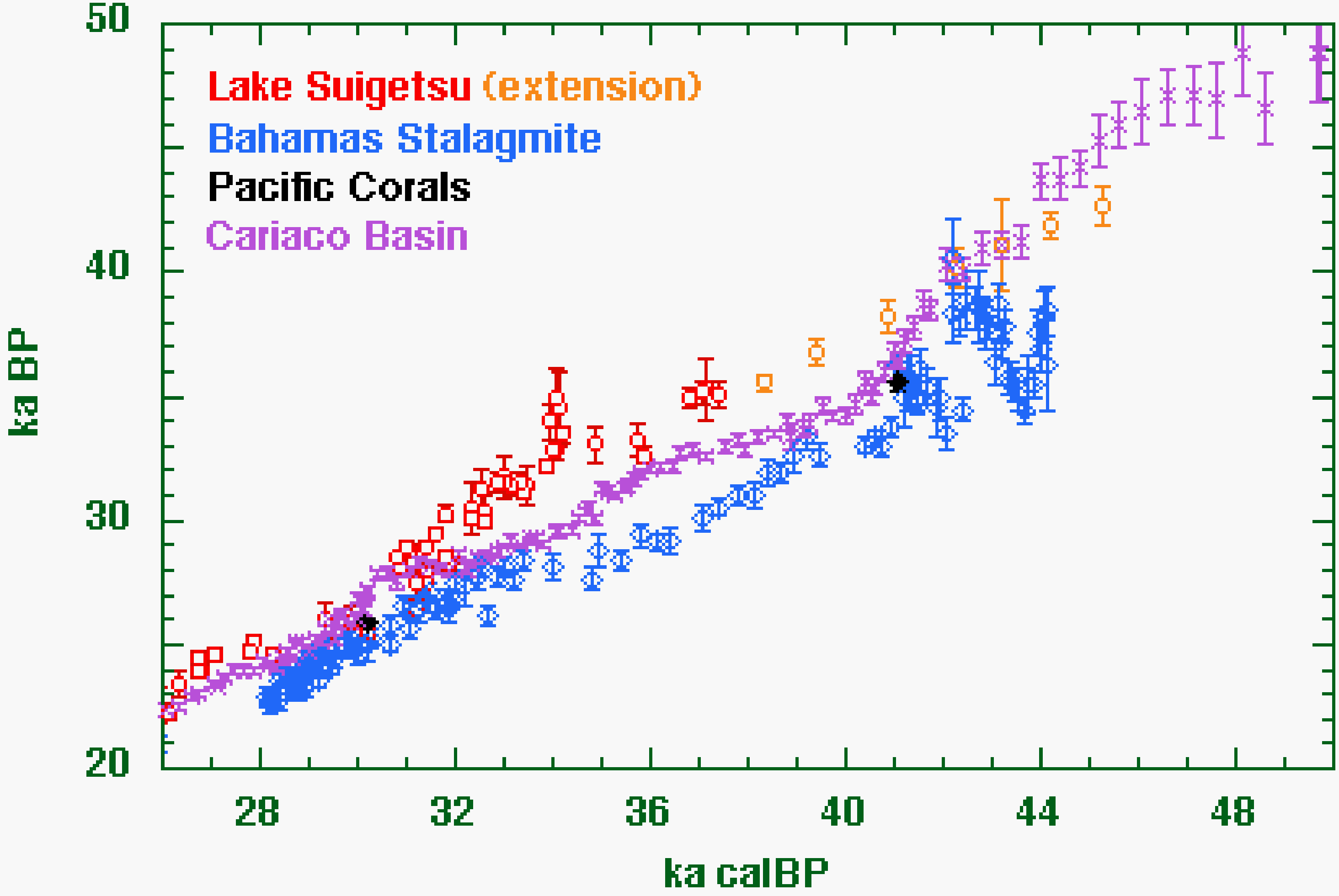
reservoir effect; constant ?

calBP : **varve counting** **min**

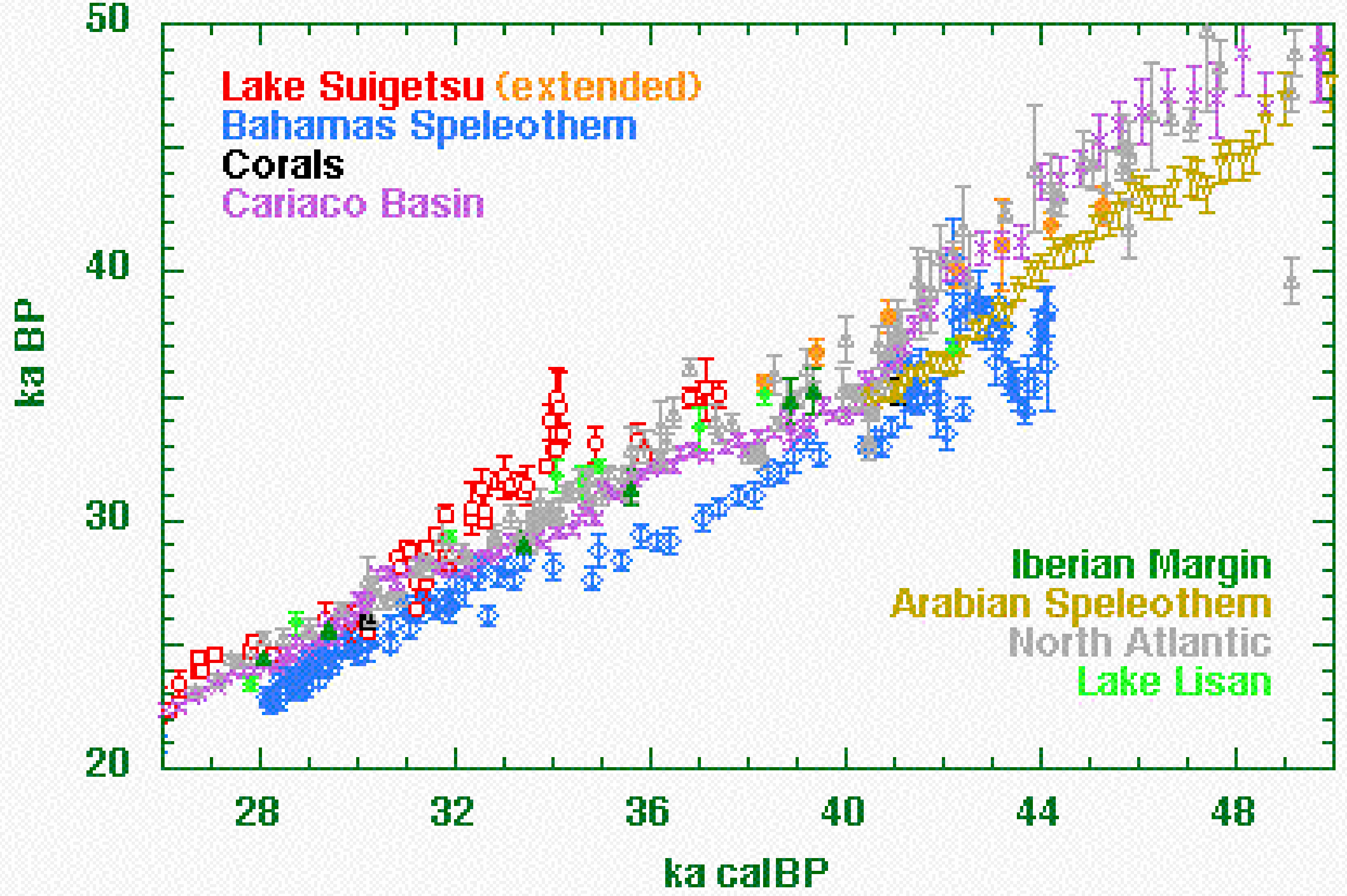
$\delta^{18}O$ correlation of climatic events with icecores

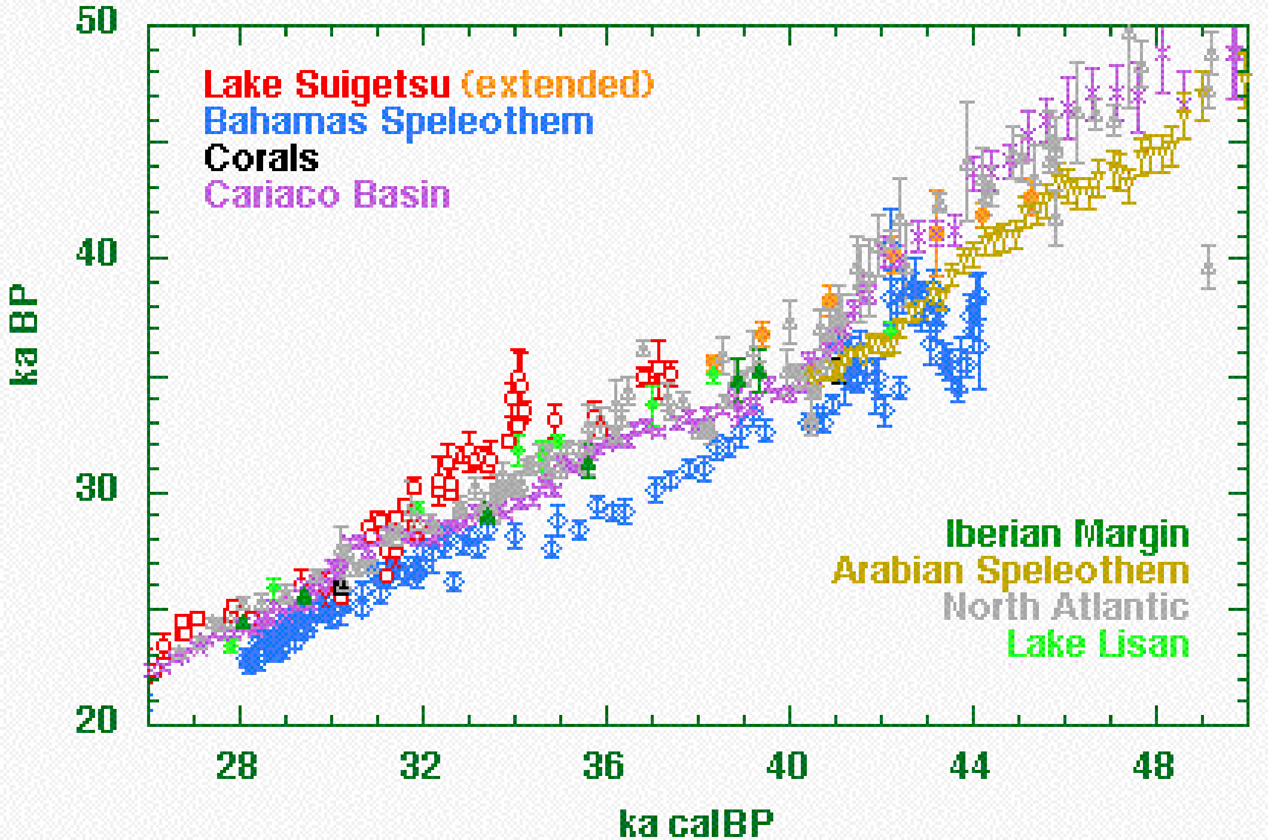


Lake Suigetsu (extension)
Bahamas Stalagmite
Pacific Corals
50
40
30
20
ka BP
28
32
36
40
44
48
ka calBP



Lake Suigetsu (extension)
Bahamas Stalagmite
Pacific Corals
Cariaco Basin
ka BP
50
40
30
20
28
32
36
40
44
48
ka calBP


Lake Suigetsu (extended)
Bahamas Speleothem
Corals
Cariaco Basin
Iberian Margin
Arabian Speleothem
North Atlantic
Lake Lisan
50
40
30
20
ka BP
28
32
36
40
44
48
ka calBP



YOUR ATTENTION PLEASE !!!

- ♦ ***errors horizontal (calBP) NOT indicated***
- ♦ **extremes “envelope” ≈ 7 millennia “absolute”**
- ♦ **extreme ^{14}C variationss Bahamas *not* confirmed by Arabian speleothem**
- ♦ **marine records use GISP2 icecore timescale**

NOTCAL04

calibration 26-50 ka impossible

1. example: 31000 BP calibrates to 32000 BC using Suigetsu
   39000 BC using Bahamas, 36000 BC using Cariaco
2. Cariaco marine data damps wiggles
3. Nobody has yet the correct record

calibration >26 ka calBP can be

1. subjective (select your favorite dataset)
2. misleading (using some averaged curve)
3. useless (using envelope extremes)

Chauvet ⇑
31000 BP

Neandertal

compare ^{14}C dating with archeology (strata, material, ...) or other dating method (TL)