СОФИЙСКИ УНИВЕРСИТЕТ
„СВ. КЛИМЕНТ ОХРИДСКИ“

ФИЗИЧЕСКИ ФАКУЛТЕТ

SOFIA UNIVERSITY
ST. KLIMENT OHRIDSKI

FACULTY OF PHYSICS

Metrology and international traceability in the field of radionuclide measurement

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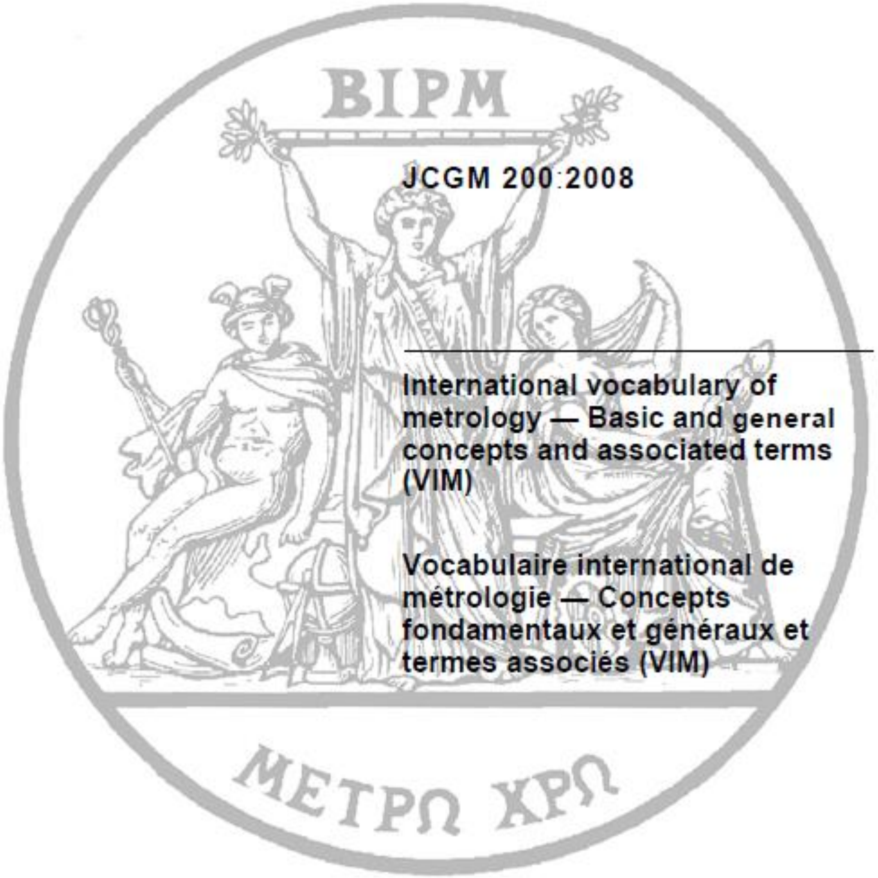
With the contribution of Carine Michotte (BIPM)

First, some useful definitions (from the VIM)



JCGM 200:2008

International vocabulary of metrology — Basic and general concepts and associated terms (VIM)

Vocabulaire international de métrologie — Concepts fondamentaux et généraux et termes associés (VIM)

https://www.bipm.org/en/committees/cc/wg/jcgm-wg2.html

Quantity

property of a phenomenon, body, or substance, where the property has a magnitude that can be expressed as a number and a reference

Measurement

process of experimentally obtaining one or more **quantity values** that can reasonably be attributed to a **quantity**

Measurand

quantity intended to be measured

Measurement unit

real scalar **quantity,** defined and adopted by convention, with which any other quantity of the same **kind** can be compared to express the ratio of the two quantities as a number

International System of Units (SI)

system of units, based on the **International System of Quantities,** their names and symbols, including a series of prefixes and their names and symbols, together with rules for their use, adopted by the General Conference on Weights and Measures (CGPM)

Measurement method

generic description of a logical organization of operations used in a **measurement**

Measurement procedure

detailed description of a **measurement** according to one or more **measurement principles** and to a given **measurement method,** based on a **measurement model** and including any calculation to obtain a **measurement result**

Primary reference measurement procedure

reference measurement procedure used to obtain a **measurement result** without relation to a **measurement standard** for a **quantity** of the same **kind**

Measurement result

set of **quantity values** being attributed to a **measurand** together with any other available relevant information

Measurement accuracy

closeness of agreement between a **measured quantity value** and a **true quantity value** of a **measurand**

Measurement trueness

closeness of agreement between the average of an infinite number of replicate **measured quantity values** and a **reference quantity value**

Measurement precision

closeness of agreement between **indications** or **measured quantity values** obtained by replicate **measurements** on the same or similar objects under specified conditions

Measurement uncertainty

non-negative parameter characterizing the dispersion of the **quantity values** being attributed to a **measurand,** based on the information used

Metrological traceability

property of a **measurement result** whereby the result can be related to a reference through a documented unbroken chain of **calibrations,** each contributing to the **measurement uncertainty**

Measurement standard (etalon)

realization of the definition of a given **quantity,** with stated **quantity value** and associated **measurement uncertainty,** used as a reference

Primary measurement standard

measurement standard established using a **primary reference measurement procedure,** or created as an artifact, chosen by convention

Metrology

Metrology is the science of measurement and its applications (VIM)

The result of a measurement result ***should*** include:

1. The numerical value of the measurand
2. The unit (in the international unit system SI)
3. The evaluated uncertainty

(The value of the quantity is the product of the numerical value by the base unit:
$L = 3\ \text{m} \quad \Leftrightarrow \quad L = 3 \times 1\ \text{m}$)

Uncertainties

The uncertainty of a measurement result expresses the reliability of that result or the confidence that we have in it

⇓

A measurement (result) is incomplete without a statement of the corresponding measurement uncertainty

The uncertainty quantifies:

The confidence we have in our values,

The confidence that others could have in our values

Metrology is all about controlling uncertainty

Units

Systems of units, historical perspective

Until the 18th century, no unified measurement system

In 1795, over 700 different "unit of measurement" in France

Many were from human morphology: the digit, the hand, the foot, the cubit, the pace, the fathom or toise…

These units of measure were not fixed: they varied from one town to another, from one occupation to another, and on the type of object to be measured

This was a source of error and fraud in commercial transactions. With the expansion of industry and trade, there was an increasing need for harmonisation.

Examples

Old French units

Mass	
Livre poids-de marc Aix	408,51 g
Livre poids-de marc Calais	509,53 g
Livre de Charlemagne	367,128 g
Livre de Paris	489,505 85 g

Length	
Pied-de-roi Rouen	270,7 mm
Pied-de-roi Calais	341,8 mm
Pied de Paris	324,839 416 7 mm
Canne Provence	2005,2 mm
Canne Troyes	792 mm

Old Bulgarian length units

Пръст	(finger)
Пѐдя	(Palm, span)
Крачка	(step)
Човешки бой	(mans height)

A coherent system of units, heritage of the French revolution

Key dates

- 16 February 1791: creation of a commission (composed of Borda, Condorcet, Laplace, Lagrange and Monge) to define uniform system of measurement
- 26 March 1791: the metre was defined as the ten millionth part of one quarter of the terrestrial meridian. The idea was to define a unit which in its determination was neither arbitrary nor related to any particular nation on the globe.

The length of the meridian was measured using the triangulation system, by Pierre-François Méchain (1744-1804) and Jean-Baptiste Delambre (1747-1822). The length of a quarter meridian, was equivalent to ten million metres (1747-1822).

The decimal metric system, a revolutionary invention!

The decimal metric system was introduced on 7 April 1795 by the law "on weights and measures". Decimalisation facilitates the calculation of areas and volumes.

For the unit of mass, the commission preferred water to any other body such as mercury or gold, due to the "ease of obtaining water and distilling it…". The kilogram was defined as being equal to the mass of a cubic decimetre of water at a given temperature.

For practical use, the first standards of the metre and the kilogram were manufactured in 1799 and deposited in the Archives of the French Republic, dedicated to "all men and all times".

METRE ETALON

La Convention nationale, afin de généraliser l'usage du système métrique, fit placer seize mètres étalons en marbre dans les lieux les plus fréquentés de Paris.

Ces mètres furent installés entre février 1796 et décembre 1797. Celui-ci est l'un des deux derniers qui subsistent à Paris et le seul qui soit encore sur son site originel.

Simple and universal, the decimal metric system started to spread outside France. After 1860, adhesion increased in many countries. But, these countries were dependent on France, as exact copies of the metre and kilogram standards were required. To overcome this problem, **the Bureau International des Poids et Mesures (BIPM)** was founded in 1875, during the diplomatic conference of the metre which led, on 20 May 1875 to the signature of the treaty known as the Metre Convention by the plenipotentiaries of 17 States.

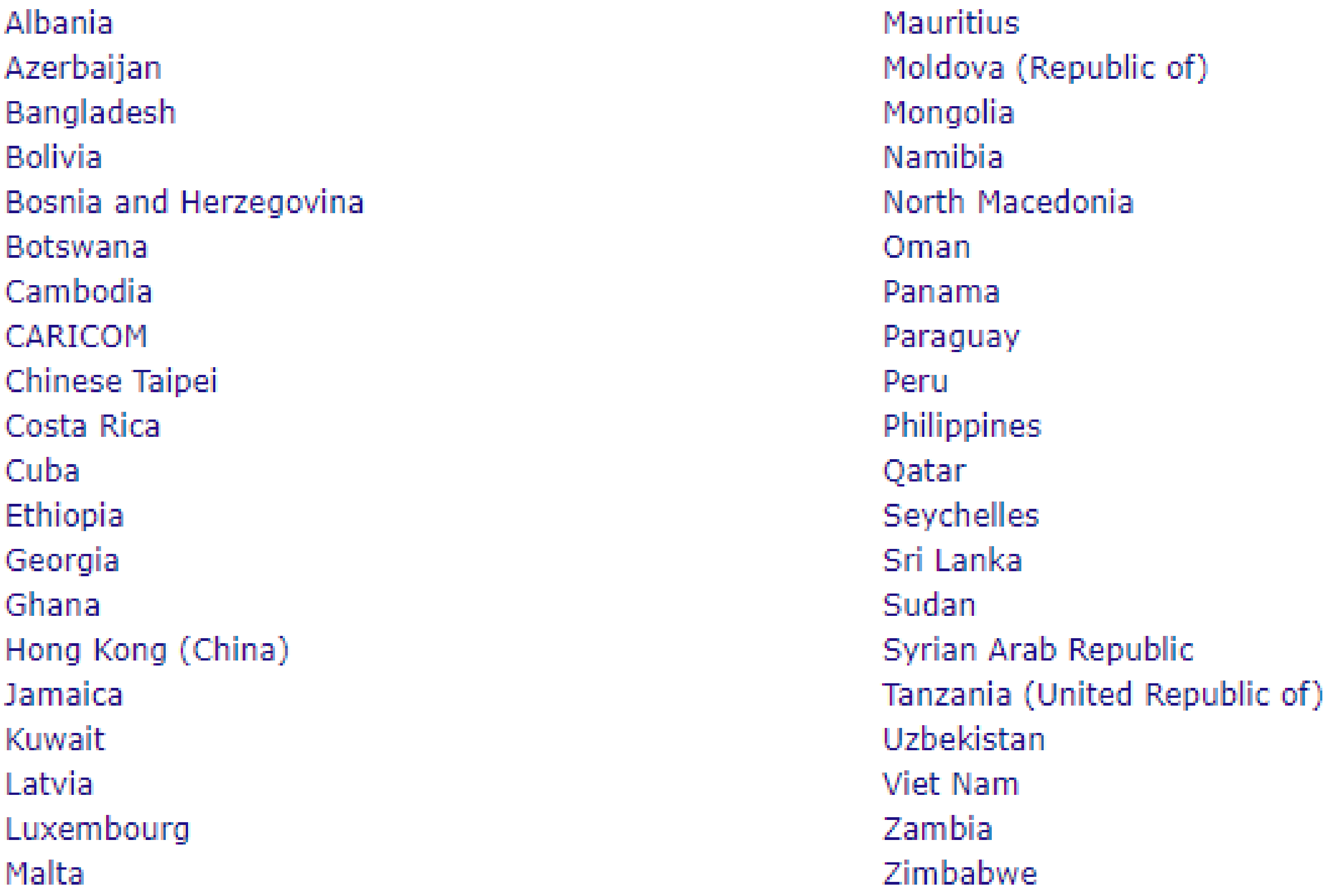
Initial signatories of the metre convention (1875)

Germany, Argentina, Austria Hungary, Belgium, Brazil, Denmark, Spain, USA, France, Italy, Peru, Portugal, Russia, Sweden and Norway, Switzerland, Turkey, Venezuela

States Parties to the Metre Convention (2020)

- Argentina
- Australia
- Austria
- Belarus
- Belgium
- Brazil
- Bulgaria
- Canada
- Chile
- China
- Colombia
- Croatia
- Czechia
- Denmark
- Ecuador
- Egypt
- Estonia
- Finland
- France
- Germany
- Greece
- Hungary
- India
- Indonesia
- Iran (Islamic Republic of)
- Iraq
- Ireland
- Israel
- Italy
- Japan
- Kazakhstan
- Kenya
- Korea (Republic of)
- Lithuania
- Malaysia
- Mexico
- Montenegro
- Morocco
- Netherlands
- New Zealand
- Norway
- Pakistan
- Poland
- Portugal
- Romania
- Russian Federation
- Saudi Arabia
- Serbia
- Singapore
- Slovakia
- Slovenia
- South Africa
- Spain
- Sweden
- Switzerland
- Thailand
- Tunisia
- Turkey
- Ukraine
- United Arab Emirates
- United Kingdom
- United States of America
- Uruguay

Associate states



Albania
Azerbaijan
Bangladesh
Bolivia
Bosnia and Herzegovina
Botswana
Cambodia
CARICOM
Chinese Taipei
Costa Rica
Cuba
Ethiopia
Georgia
Ghana
Hong Kong (China)
Jamaica
Kuwait
Latvia
Luxembourg
Malta
Mauritius
Moldova (Republic of)
Mongolia
Namibia
North Macedonia
Oman
Panama
Paraguay
Peru
Philippines
Qatar
Seychelles
Sri Lanka
Sudan
Syrian Arab Republic
Tanzania (United Republic of)
Uzbekistan
Viet Nam
Zambia
Zimbabwe

The BIPM's initial mission was to set up the Metric System throughout the world by constructing and maintaining new prototypes of the metre and the kilogram, comparing the national standards with these prototypes and perfecting the measurement methods in order to promote metrology in all fields.

The BIPM progressively focused on the study of metrological problems and physical constants which govern the precision of measurements when defining units (e.g. thermometry), then, to accompany industrial development, its scope extended to new fields : **the electrical units** (1937), **the photometric units** (1937) or **the ionising radiation measurement standards** (1960).

International organization

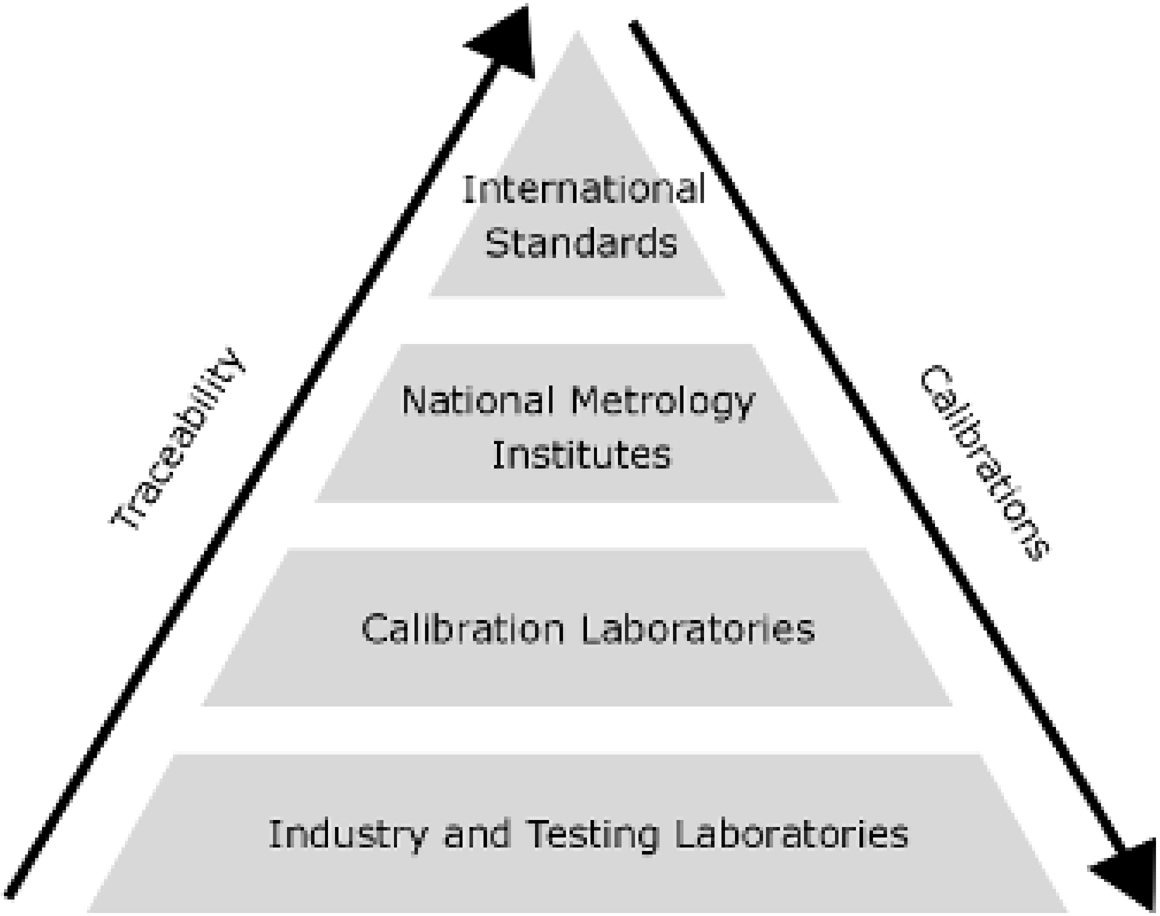
Bureau International des Poids et Mesures (BIPM) – Pavillon de Breteuil à Sèvres

"ensure and promote the global comparability of measurements, including providing a coherent international system of units "

~~Altruism, aim at the scientific perfection?~~

"...a lack of traceability and uniformization of units is a source of errors and facilitates frauds in commercial transactions "

International traceability



International Standards

National Metrology Institutes

Calibration Laboratories

Industry and Testing Laboratories

Le Système international d'unités
The International System of Units
SI

8e édition
2006

Bureau
international
des poids
et mesures

Organisation
intergouvernementale
de la Convention
du Mètre

The highest metrological authority is the General conference of weights and measurements (CGPM)

International committee for weights and measures CIPM, with the help of consultative committees

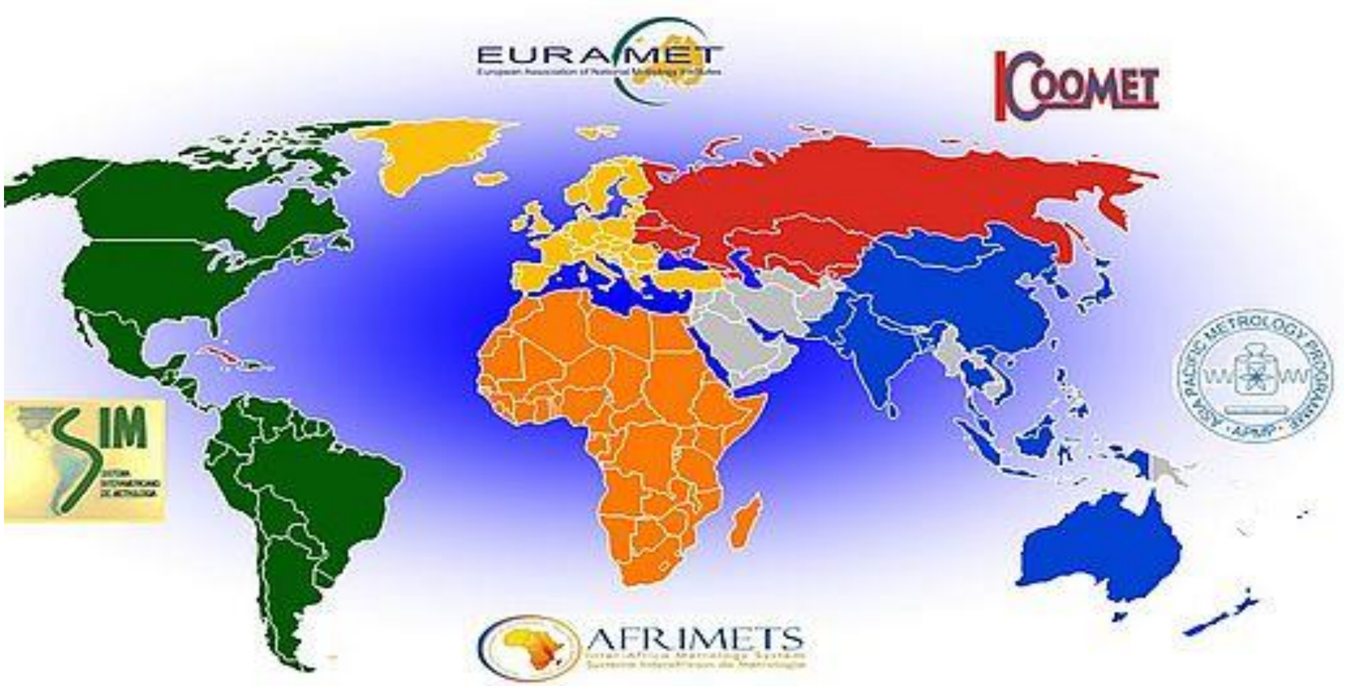
CCAUV - Acoustics, ultrasound and vibration
CCEM - Electricity and magnetism
CCL - length
CCM - mass and related quantities
CCPR - Photometry and radiometry
CCQM - Amount of substance, chemistry and biology

CCRI - Ionizing radiations
Section I: dosimetry x, γ, electrons
Section II: radionuclides
Section III: neutron metrology

CCT - Thermometry
CCTF - Time and frequency
CCU - Units

International metrology organization

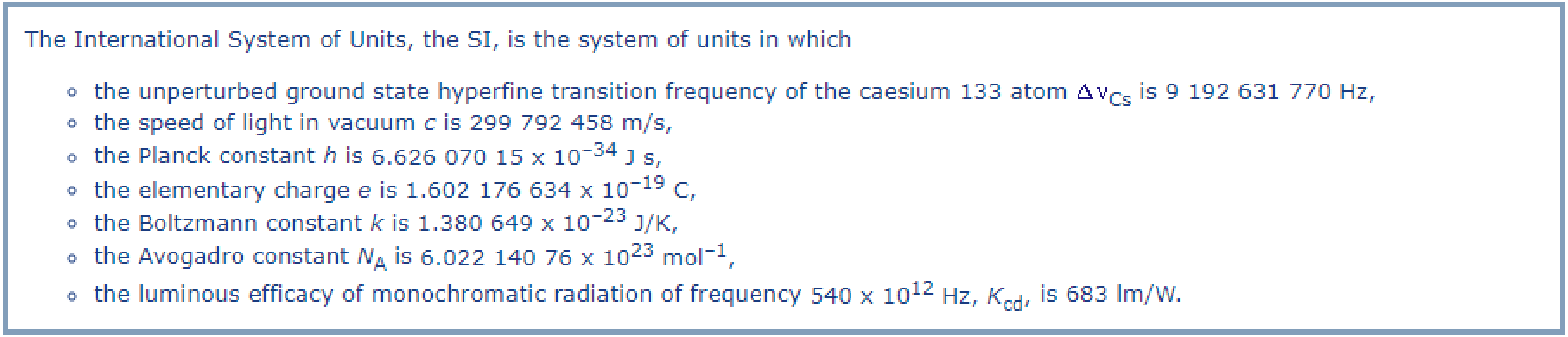
Regional organizations



The BIPM assures, with the National Metrology Institutes, the equivalence and mutual recognition of standards traceable to the SI, in the world

The new SI (2019)

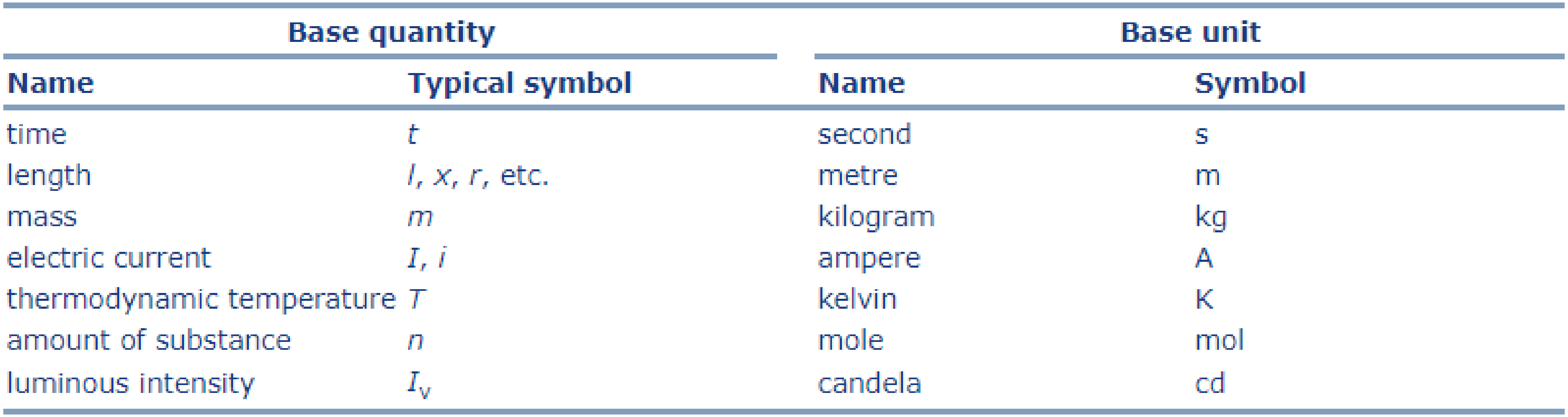
It was decided at the 26th meeting of the General Conference on Weights and Measures (CGPM), that, from 20 May 2019 all SI units are defined in terms of constants that describe the natural world. This will assure the future stability of the SI and open the opportunity for the use of new technologies, including quantum technologies, to implement the definitions.



The International System of Units, the SI, is the system of units in which

- the unperturbed ground state hyperfine transition frequency of the caesium 133 atom $\Delta\nu_{\mathrm{Cs}}$ is 9 192 631 770 Hz,
- the speed of light in vacuum c is 299 792 458 m/s,
- the Planck constant h is $6.626\ 070\ 15 \times 10^{-34}$ J s,
- the elementary charge e is $1.602\ 176\ 634 \times 10^{-19}$ C,
- the Boltzmann constant k is $1.380\ 649 \times 10^{-23}$ J/K,
- the Avogadro constant N_A is $6.022\ 140\ 76 \times 10^{23}$ mol^{-1},
- the luminous efficacy of monochromatic radiation of frequency 540×10^{12} Hz, K_{cd}, is 683 lm/W.

International System of units (SI)



Base quantity		**Base unit**	
Name	**Typical symbol**	**Name**	**Symbol**
time	*t*	second	s
length	*l*, *x*, *r*, etc.	metre	m
mass	*m*	kilogram	kg
electric current	*I*, *i*	ampere	A
thermodynamic temperature	*T*	kelvin	K
amount of substance	*n*	mole	mol
luminous intensity	I_v	candela	cd

All other SI units can be derived from these base units e.g. W, J, Bq, Gy,…

The second

The second, symbol s, is the SI unit of time. It is defined by taking the fixed numerical value of the caesium frequency $\Delta\nu_{Cs}$, the unperturbed ground-state hyperfine transition frequency of the caesium-133 atom, to be 9 192 631 770 when expressed in the unit Hz, which is equal to s^{-1}.

This definition implies the exact relation $\Delta\nu_{Cs} = 9\ 192\ 631\ 770$ Hz. Inverting this relation gives an expression for the unit second in terms of the defining constant $\Delta\nu_{Cs}$:

$$1\,\mathrm{Hz} = \frac{\Delta\nu_{Cs}}{9\,192\,631\,770}$$

or

$$1\,\mathrm{s} = \frac{9\,192\,631\,770}{\Delta\nu_{Cs}}.$$

The effect of this definition is that the second is equal to the duration of 9 192 631 770 periods of the radiation corresponding to the transition between the two hyperfine levels of the unperturbed ground state of the ^{133}Cs atom.

The metre

The metre, symbol m, is the SI unit of length. It is defined by taking the fixed numerical value of the speed of light in vacuum c to be 299 792 458 when expressed in the unit m s^{-1}, where the second is defined in terms of the caesium frequency $\Delta\nu_{Cs}$.

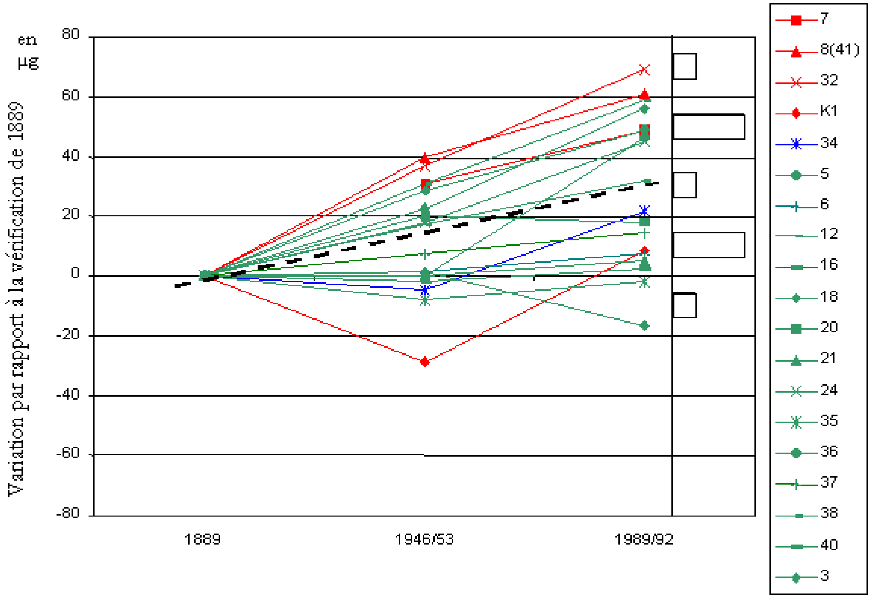
This definition implies the exact relation c = 299 792 458 m s^{-1}. Inverting this relation gives an exact expression for the metre in terms of the defining constants c and $\Delta\nu_{Cs}$:

$$1\,\mathrm{m} = \left(\frac{c}{299\ 792\ 458}\right)\mathrm{s} = \frac{9\ 192\ 631\ 770}{299\ 792\ 458}\,\frac{c}{\Delta\nu_{\mathrm{Cs}}} \approx 30{,}663\ 319\,\frac{c}{\Delta\nu_{\mathrm{Cs}}}\,.$$

The effect of this definition is that one metre is the length of the path travelled by light in vacuum during a time interval with duration of 1/299 792 458 of a second.

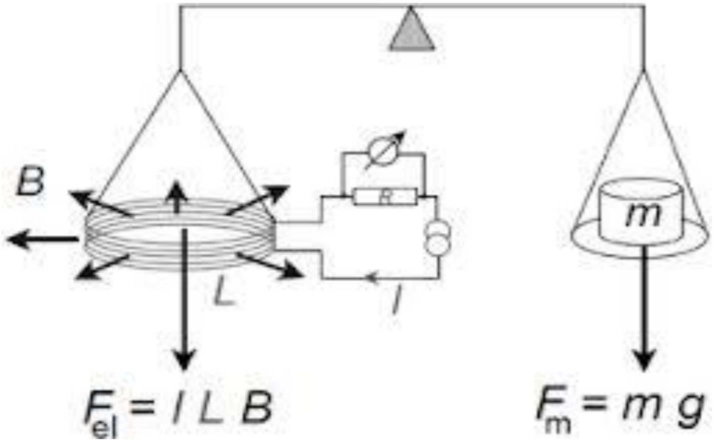
The « old » kilogram

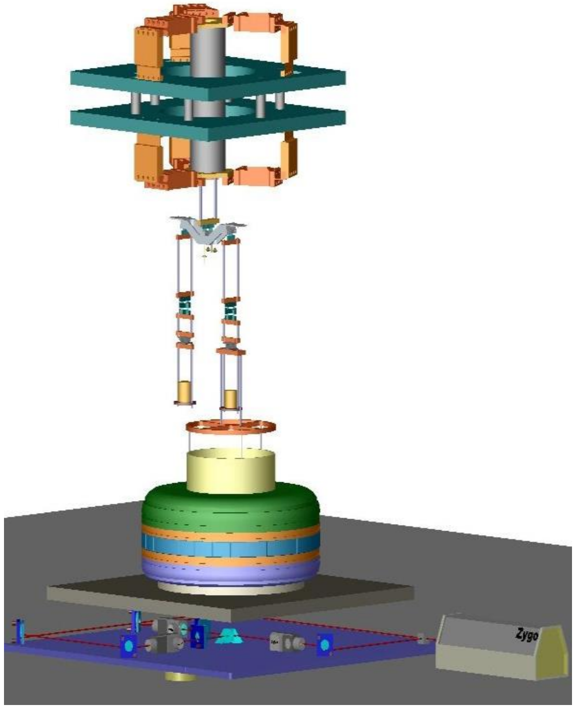
Mass of the international prototype of the kilogram (cylinder of platinum irridium) stored at BIPM since 1889



But observed variation of 50 µg over 100 years

New approach, the Kibble balance





Comparison of electrical power to mechanical power → link to *h*

Other methods: joule-balance, volt-balance and X-ray crystal density method (sphere of ^{28}Si)

The kilogram, new definition

The kilogram, symbol kg, is the SI unit of mass. It is defined by taking the fixed numerical value of the Planck constant *h* to be 6.626 070 15 x 10^{-34} when expressed in the unit J s, which is equal to kg m^2 s^{-1}, where the metre and the second are defined in terms of *c* and $\Delta\nu_{Cs}$.

This definition implies the exact relation *h* = 6.626 070 15 x 10^{-34} kg m^2 s^{-1}. Inverting this relation gives an exact expression for the kilogram in terms of the three defining constants *h*, $\Delta\nu_{Cs}$ and *c*:

$$1\,\mathrm{kg} = \left(\frac{h}{6.626\,070\,15\times10^{-34}}\right)\mathrm{m}^{-2}\mathrm{s}$$

which is equal to

$$1\,\mathrm{kg} = \frac{(299\,792\,458)^2}{(6.626\,070\,15\times10^{-34})(9\,192\,631\,770)}\frac{h\,\Delta\nu_{\mathrm{Cs}}}{c^2} \approx 1.475\,5214\times10^{40}\,\frac{h\,\Delta\nu_{\mathrm{Cs}}}{c^2}$$

The effect of this definition is to define the unit kg m^2 s^{-1} (the unit of both the physical quantities action and angular momentum). Together with the definitions of the second and the metre this leads to a definition of the unit of mass expressed in terms of the Planck constant *h*.

The ampere

The ampere, symbol A, is the SI unit of electric current. It is defined by taking the fixed numerical value of the elementary charge e to be 1.602 176 634 x 10^{-19} when expressed in the unit C, which is equal to A s, where the second is defined in terms of $\Delta\nu_{Cs}$.

This definition implies the exact relation e = 1.602 176 634 x 10^{-19} A s. Inverting this relation gives an exact expression for the unit ampere in terms of the defining constants e and $\Delta\nu_{Cs}$:

$$1\,\mathrm{A} = \left(\frac{e}{1.602\ 176\ 634 \times 10^{-19}}\right)\mathrm{s}^{-1}$$

which is equal to

$$1\,\mathrm{A} = \frac{1}{(9\ 192\ 631\ 770)(1.602\ 176\ 634 \times 10^{-19})}\Delta\nu_{\mathrm{Cs}}e \approx 6.789\ 687 \times 10^{8}\,\Delta\nu_{\mathrm{Cs}}e$$

The effect of this definition is that one ampere is the electric current corresponding to the flow of 1/(1.602 176 634 x 10^{-19}) elementary charges per second.

The kelvin

The kelvin, symbol K, is the SI unit of thermodynamic temperature. It is defined by taking the fixed numerical value of the Boltzmann constant k to be 1.380 649 x 10^{-23} when expressed in the unit J K^{-1}, which is equal to kg m^2 s^{-2} K^{-1}, where the kilogram, metre and second are defined in terms of h, c and $\Delta\nu_{\mathrm{Cs}}$.

This definition implies the exact relation k = 1.380 649 x 10^{-23} kg m^2 s^{-2} K^{-1}. Inverting this relation gives an exact expression for the kelvin in terms of the defining constants k, h and $\Delta\nu_{\mathrm{Cs}}$:

$$1\,\mathrm{K} = \left(\frac{1.380\,649}{k}\right) \times 10^{-23}\ \mathrm{kg\,m^{2}\,s^{-2}}$$

which is equal to

$$1\,\mathrm{K} = \frac{1.380\,649 \times 10^{-23}}{(6.626\,070\,15 \times 10^{-34})(9\,192\,631\,770)}\frac{\Delta\nu_{\mathrm{Cs}}h}{k} \approx 2.266\,6653\frac{\Delta\nu_{\mathrm{Cs}}h}{k}$$

The effect of this definition is that one kelvin is equal to the change of thermodynamic temperature that results in a change of thermal energy $k\,T$ by 1.380 649 x 10^{-23} J.

The mole

The mole, symbol mol, is the SI unit of amount of substance. One mole contains exactly 6.022 140 76 x 10^{23} elementary entities. This number is the fixed numerical value of the Avogadro constant, N_A, when expressed in the unit mol^{-1} and is called the Avogadro number.

The amount of substance, symbol n, of a system is a measure of the number of specified elementary entities. An elementary entity may be an atom, a molecule, an ion, an electron, any other particle or specified group of particles.

This definition implies the exact relation N_A = 6.022 140 76 x 10^{23} mol^{-1}. Inverting this relation gives an exact expression for the mole in terms of the defining constant N_A:

$$1\,\mathrm{mol} = \left(\frac{6.022\,140\,76 \times 10^{23}}{N_A}\right)$$

The effect of this definition is that the mole is the amount of substance of a system that contains 6.022 140 76 x 10^{23} specified elementary entities.

The candela

The candela, symbol cd, is the SI unit of luminous intensity in a given direction. It is defined by taking the fixed numerical value of the luminous efficacy of monochromatic radiation of frequency 540 x 10^{12} Hz, K_{cd}, to be 683 when expressed in the unit lm W^{-1}, which is equal to cd sr W^{-1}, or cd sr kg^{-1} m^{-2} s^{3}, where the kilogram, metre and second are defined in terms of h, c and $\Delta\nu_{Cs}$.

This definition implies the exact relation K_{cd} = 683 cd sr kg^{-1} m^{-2} s^{3} for monochromatic radiation of frequency ν = 540 x 10^{12} Hz. Inverting this relation gives an exact expression for the candela in terms of the defining constants K_{cd}, h and $\Delta\nu_{Cs}$:

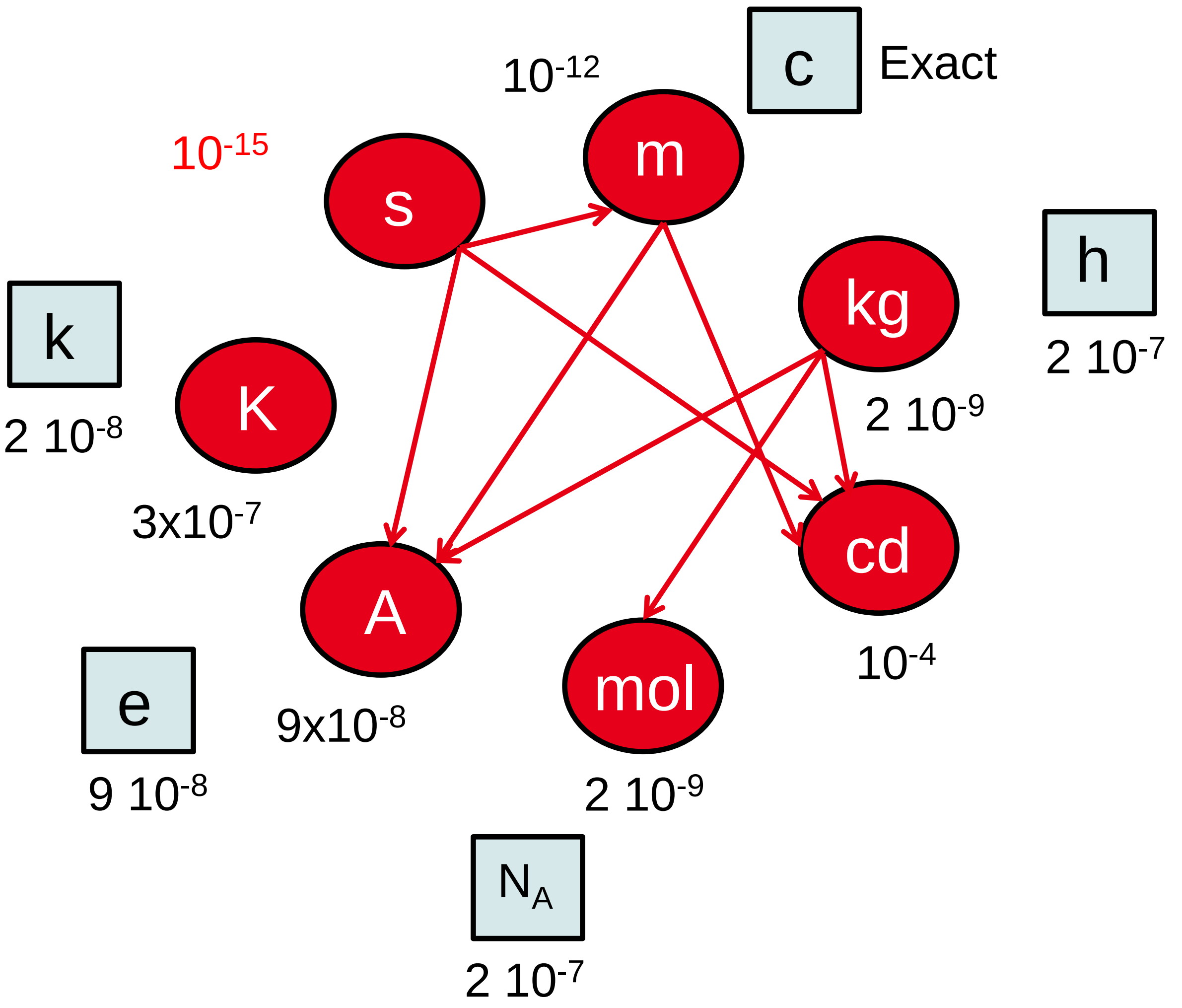
$$1\,\mathrm{cd} = \left(\frac{K_{\mathrm{cd}}}{683}\right)\mathrm{kg\,m^{2}\,s^{-3}\,sr^{-1}}$$

which is equal to

$$1\,\mathrm{cd} = \frac{1}{(6.626\,070\,15\times10^{-34})(9\,192\,631\,770)^{2}\,683}(\Delta\nu_{\mathrm{Cs}})^{2}\,h\,K_{\mathrm{cd}}$$

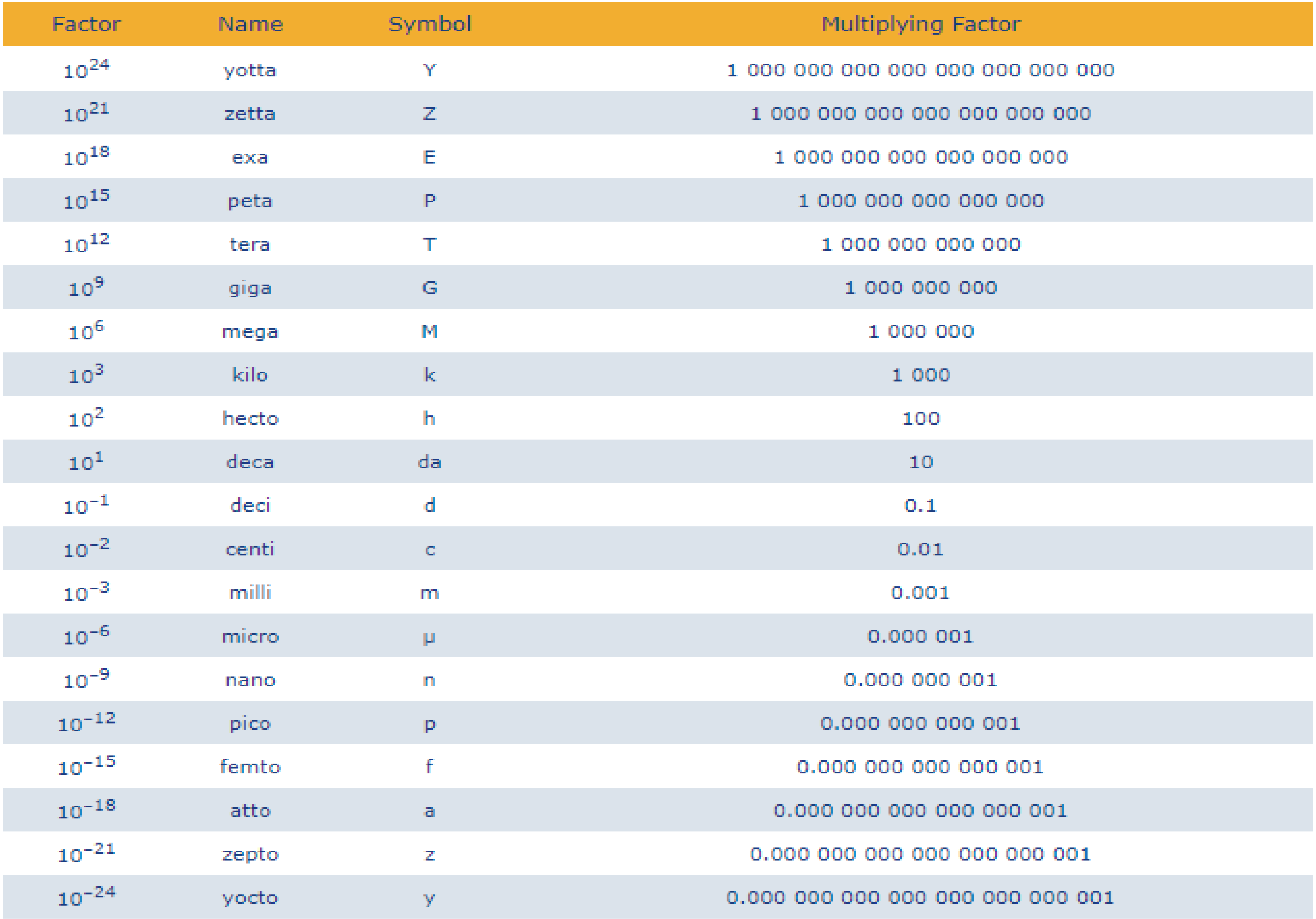
$$\approx 2.614\,830\times10^{10}\,(\Delta\nu_{\mathrm{Cs}})^{2}\,h\,K_{\mathrm{cd}}$$

Relations between the unit and the fundamental constants



Best relative standard uncertainties

Decimal multiples and submultiples of SI units, SI prefixes



Factor	Name	Symbol	Multiplying Factor
10^{24}	yotta	Y	1 000 000 000 000 000 000 000 000
10^{21}	zetta	Z	1 000 000 000 000 000 000 000
10^{18}	exa	E	1 000 000 000 000 000 000
10^{15}	peta	P	1 000 000 000 000 000
10^{12}	tera	T	1 000 000 000 000
10^{9}	giga	G	1 000 000 000
10^{6}	mega	M	1 000 000
10^{3}	kilo	k	1 000
10^{2}	hecto	h	100
10^{1}	deca	da	10
10^{-1}	deci	d	0.1
10^{-2}	centi	c	0.01
10^{-3}	milli	m	0.001
10^{-6}	micro	µ	0.000 001
10^{-9}	nano	n	0.000 000 001
10^{-12}	pico	p	0.000 000 000 001
10^{-15}	femto	f	0.000 000 000 000 001
10^{-18}	atto	a	0.000 000 000 000 000 001
10^{-21}	zepto	z	0.000 000 000 000 000 000 001
10^{-24}	yocto	y	0.000 000 000 000 000 000 000 001

Obsolete, but sometimes still used units

Ångström (Å): 10^{-10} m… so 0,1 nm

Parsec (pc): 648 000/p astronomical units (au), about 31 Pm (1 au = 149 597 870 700 m)

Carat (ct): 0,2 g

Horsepower (hp): 735,5 W (metric), 745,7 W (mechanical)

Barn (b): 10^{-24} cm^2

Units to avoid ☺

Attoparsec: about 3.086 centimetres

Barn-megaparsec: about 3 ml

Donkey power: about 250 W

Pirate-ninja: one kWh (3,6 MJ) per Martian day

Microcentury: 52 minutes and 35,7 seconds (supposed to be the maximum length of a lecture)

Examples of units problems

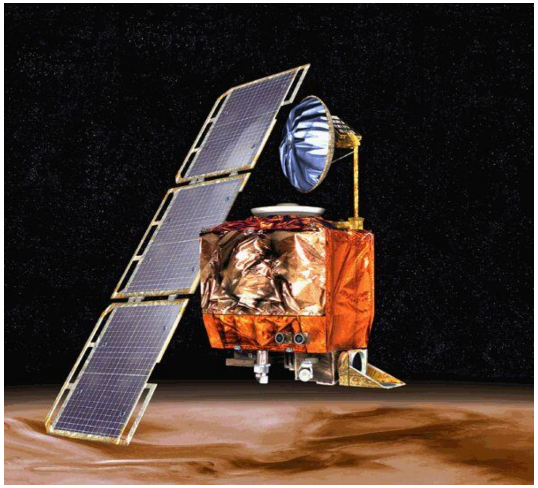
1983, Canada's aviation sector was in the process of converting from Imperial units to SI units

Air Canada Flight 143, July 23, 1983, domestic passenger flight(Boeing 767) between Montreal and Edmonton via Ottawa (61 passengers and 8 crew)

- Confusion between lb/L and kg/L
- Underestimation of the fuel quantity needed by a factor of 1,77
- Fuel shortage at an altitude of 12,500 m, midway through the flight
- The flight crew successfully glided the plane to an emergency landing at a former Military base in (Gimli, Manitoba) that had been converted to a motor racing track

Nobody was killed (only 10 minor injuries)

Mars Climate Orbiter (1999) robotic space probe



2 development teams using 2 different systems of units:

- Lockeed Martin, Imperial units: pound-force
- NASA, SI units : newton

Scale factor: 4,48

The trajectory calculation software gave a wrong result and the spacecraft was lost in the upper atmosphere of Mars

Cost: US$ 327,6 M

Units in Ionizing Radiation Metrology

Activity: mean number of disintegrations per second of a specific radionuclide (Bq)

Flux: s^{-1}/sr (e.g. emission flux of a neutron source)

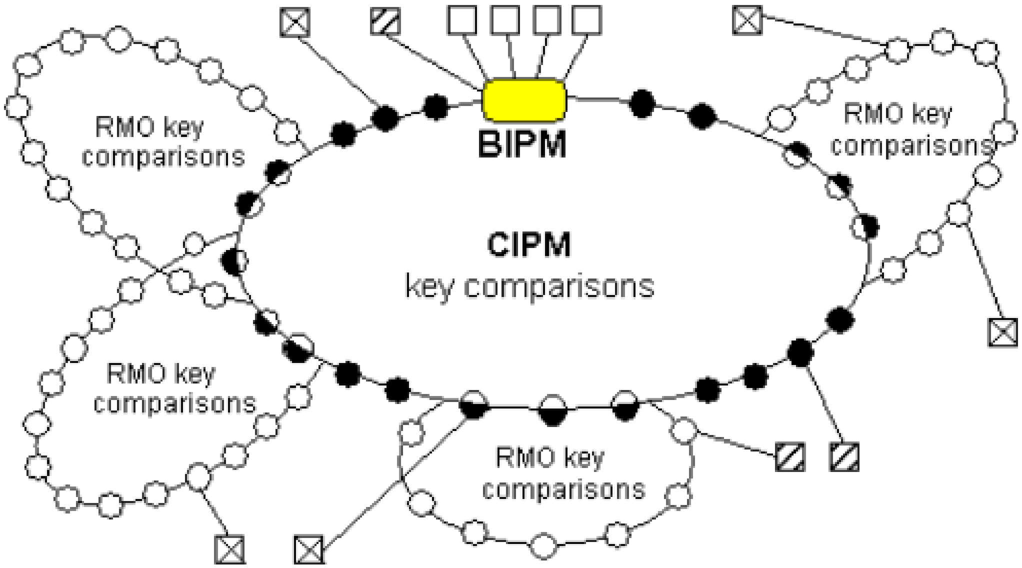
Dose: J/kg (Gy)

Equivalent dose: J/kg (Sv)

The CIPM MRA

The CIPM Mutual Recognition Arrangement (CIPM MRA) is the framework through which National Metrology Institutes demonstrate the international equivalence of their measurement standards and the calibration and measurement certificates they issue. The outcomes of the Arrangement are the internationally recognized (peer-reviewed and approved) Calibration and Measurement Capabilities (CMCs) of the participating institutes. Approved CMCs and supporting technical data are publicly available from the CIPM MRA database (the KCDB).

The CIPM MRA has been signed by the representatives of 107 institutes – from 63 Member States, 40 Associates of the CGPM, and 4 international organizations and covers a further 152 institutes designated by the signatory bodies.



●	National metrology institute (NMI) participating in CIPM key comparisons
◐	NMI participating in CIPM key comparisons and in regional metrology organization (RMO) key comparisons
○	NMI participating in RMO key comparisons
☐	NMI participating in ongoing BIPM key comparisons
☒	NMI participating in a bilateral key comparison
▨	International organization signatory to the MRA

- **CIPM key comparisons**, of international scope, are carried out by those participants having the highest level of skills in the measurement involved, and are restricted to laboratories of Member States. The CIPM key comparisons deliver "the reference value" for the chosen key quantity;
- **RMO key comparisons**, of regional scope, are organized at the scale of a region (though they may include additional participants from other regions) and are open to laboratories of Associates as well as Member States. These key comparisons deliver complementary information without changing the reference value.

Tools to assure the international traceability in the field of radionuclide metrology

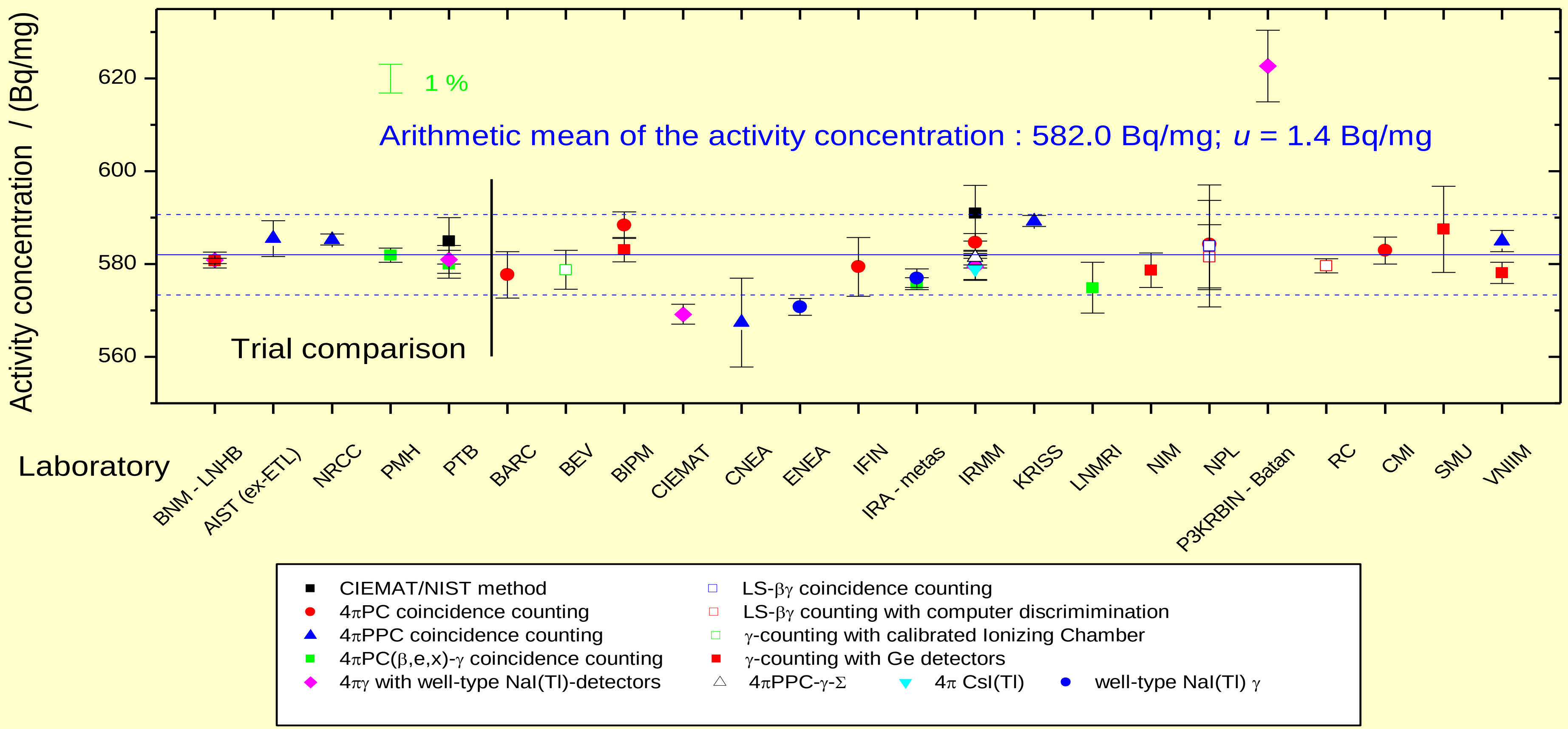
- International comparisons
- The international reference system for gamma-emitting radionuclides(SIR)

International comparisons

International activity comparisons (example 1)

^{152}Eu

Results of the full-scale international comparison of activity measurements of a solution of ^{152}Eu



International activity comparisons (example 1) ^{152}Eu

Goals of the comparison:

Define an average value of the measurand (activity per unit mass of the radioactive solution) and its associated standard uncertainty

Define equivalence between laboratories

Identify problems in the measurement or in uncertainty evaluation

How to calculate the average value?

Average, what do you mean?

$$\text{mean} = \frac{\sum x_i}{n}$$

$$\text{median} \rightarrow$$

$$\sum |x_i - M| = \min$$

$$\text{weighted mean} = \frac{\sum x_i \cdot \text{weight}}{\sum \text{weight}}$$

$$\text{weighted median} \rightarrow$$

$$\sum |x_i - M| \cdot \text{weight} = \min$$

But before calculating averages, check if the results are consistent and look for outliers

‘external’ uncertainty (weighted avg.)

$$s_{ext}^2 = \frac{1}{n-1} \frac{\sum w_i (x - <x>)^2}{\sum w_i}$$

‘internal’ uncertainty

$$s_{int}^2 = \left(\sum \frac{1}{{\sigma_i}^2} \right)^{-1}$$

for consistency between internal and external uncertainty

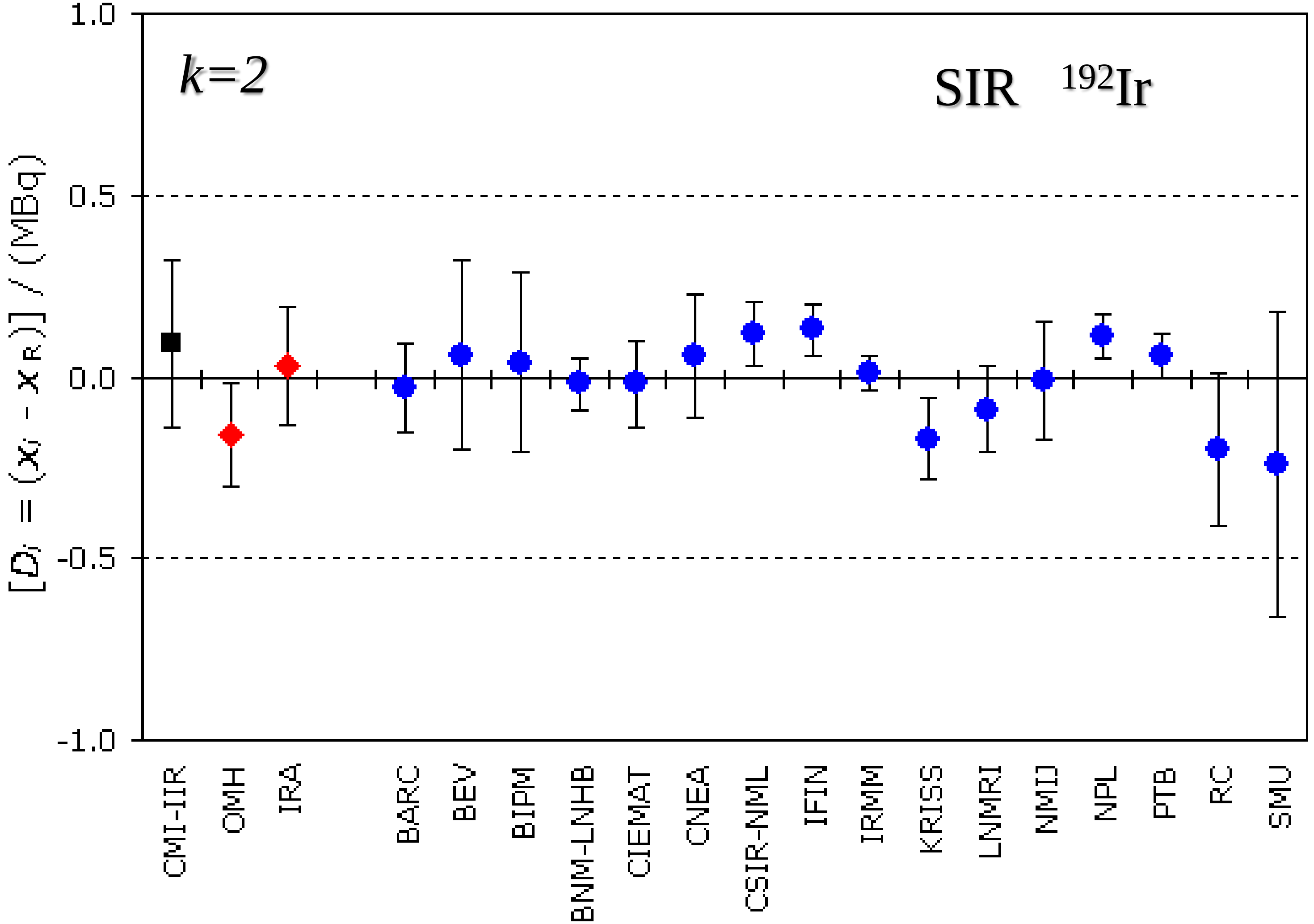
$$R_B = \frac{s_{ext}}{s_{int}} \longrightarrow 1$$

data of comparison are ‘consistent’ provided that *

$$R_B < \sqrt{1 + \sqrt{8/(n-1)}}$$

*‘*rule of thumb*’

Other example ^{192}Ir



$R_B = 1.9 > 1.3$

=> not consistent

http://kcdb.bipm.org/appendixB/KCDB_ApB_search.asp

Procedure used to analyze intercomparisons

- Degrees of Equivalence for lab *i* $D_i = x_i - x_{\text{ref}}$; U_i ($k = 2$)
- The CCRI(II) decided that D_i remains valid for 20 years
- The CCRI(II) decided that

  x_{ref} = KCRV = power moderated weighted mean

  (S. Pommé *et al, Metrologia 2015*)

$$x_{\text{ref}} = \sum_{i=1}^{N} w_i x_i \qquad \frac{1}{u^2(x_{\text{ref}})} = \sum_{i=1}^{N} \left[\left(\sqrt{u_i{}^2 + s^2} \right)^{\alpha} S^{2-\alpha} \right]^{-1},$$

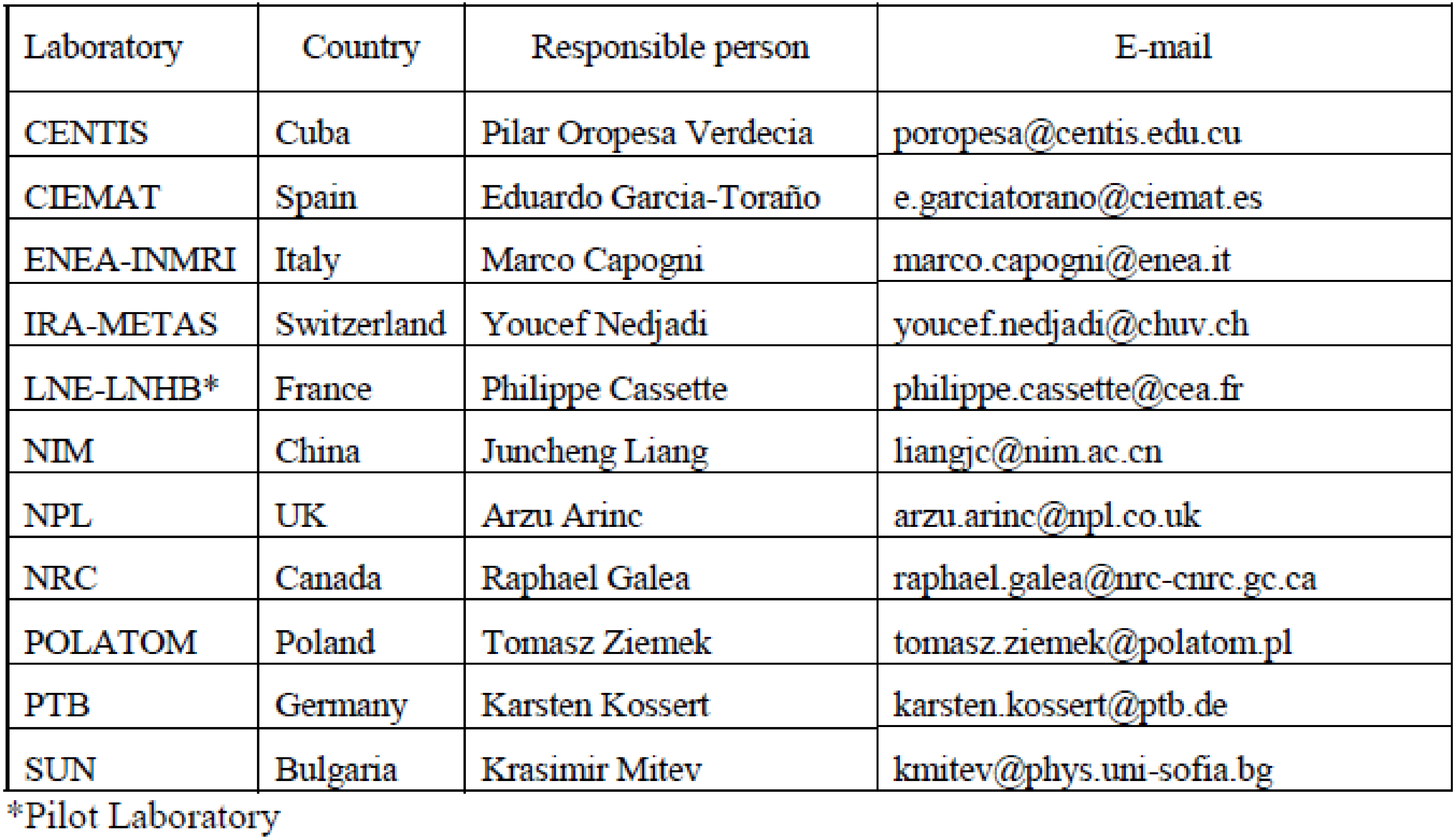
$$w_i = u^2(x_{\text{ref}}) \left[\left(\sqrt{u_i{}^2 + s^2} \right)^{\alpha} S^{2-\alpha} \right]^{-1}$$

S = ‘characteristic uncertainty per datum’

- *s* = ‘dark uncertainty’ such that $\chi^2 = 1$
- Power α : between 0 (arithmetic mean) and 2 (weighted mean)
- CCRI(II) decided that $\alpha = 2 - 3/N$

International activity comparisons (example 2)

Measurement of the activity concentration of a tritiated-water source



Laboratory	Country	Responsible person	E-mail
CENTIS	Cuba	Pilar Oropesa Verdecia	poropesa@centis.edu.cu
CIEMAT	Spain	Eduardo Garcia-Toraño	e.garciatorano@ciemat.es
ENEA-INMRI	Italy	Marco Capogni	marco.capogni@enea.it
IRA-METAS	Switzerland	Youcef Nedjadi	youcef.nedjadi@chuv.ch
LNE-LNHB*	France	Philippe Cassette	philippe.cassette@cea.fr
NIM	China	Juncheng Liang	liangjc@nim.ac.cn
NPL	UK	Arzu Arinc	arzu.arinc@npl.co.uk
NRC	Canada	Raphael Galea	raphael.galea@nrc-cnrc.gc.ca
POLATOM	Poland	Tomasz Ziemek	tomasz.ziemek@polatom.pl
PTB	Germany	Karsten Kossert	karsten.kossert@ptb.de
SUN	Bulgaria	Krasimir Mitev	kmitev@phys.uni-sofia.bg

*Pilot Laboratory

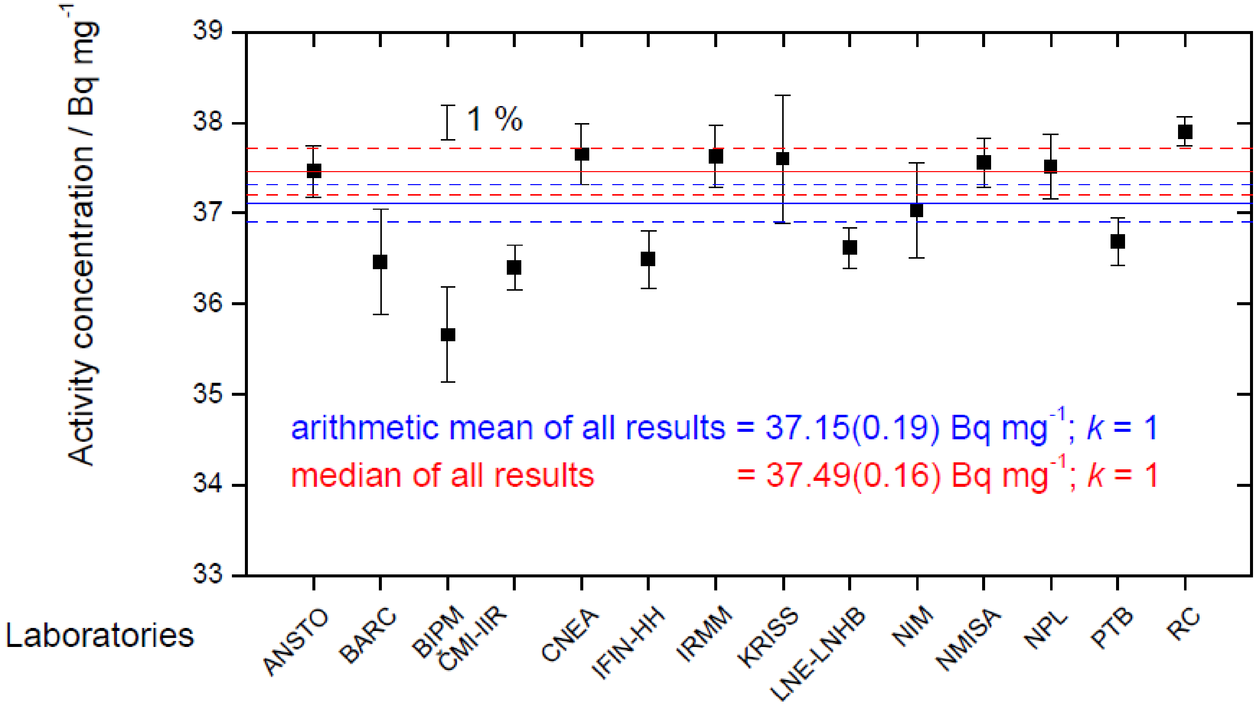
CCRI(II) H-3 2018

- **Source composition:** Tritiated water delivered in a flame-sealed glass ampoule. The volume of the solution is about 5 mL and the activity concentration about 50 kBq/g at time of shipment by LNHB (Feb 2018)
- **Activity traceable** to the solution of the CCRI(II)-K2.H-3, 2009 solution (traceability assured by comparative measurement of the 2 solutions at LNE-LNHB and PTB)
- **Measurand:** activity concentration of ^{3}H in the solution at the reference date
- **Recommended nuclear data:** Decay Data Evaluation Project
- **Reference date:** is 1st February, 2018, 12h00 UTC
- **Reporting deadline:** 1th September, 2018

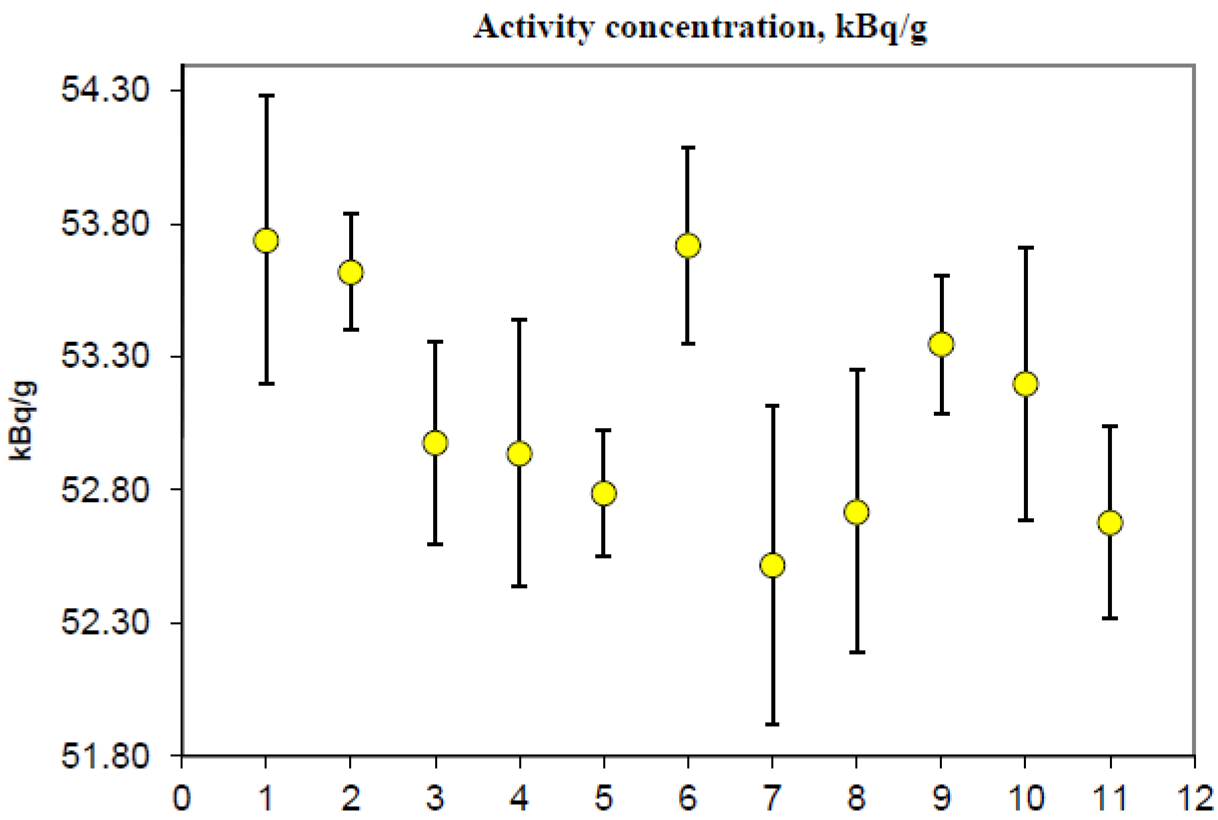
Previous comparison

CCRI(II)-K2.H-3, 2009, 14 participants

International comparison of activity measurements of a solution of ^{3}H
One result per laboratory; one result not included in the potential KCRV



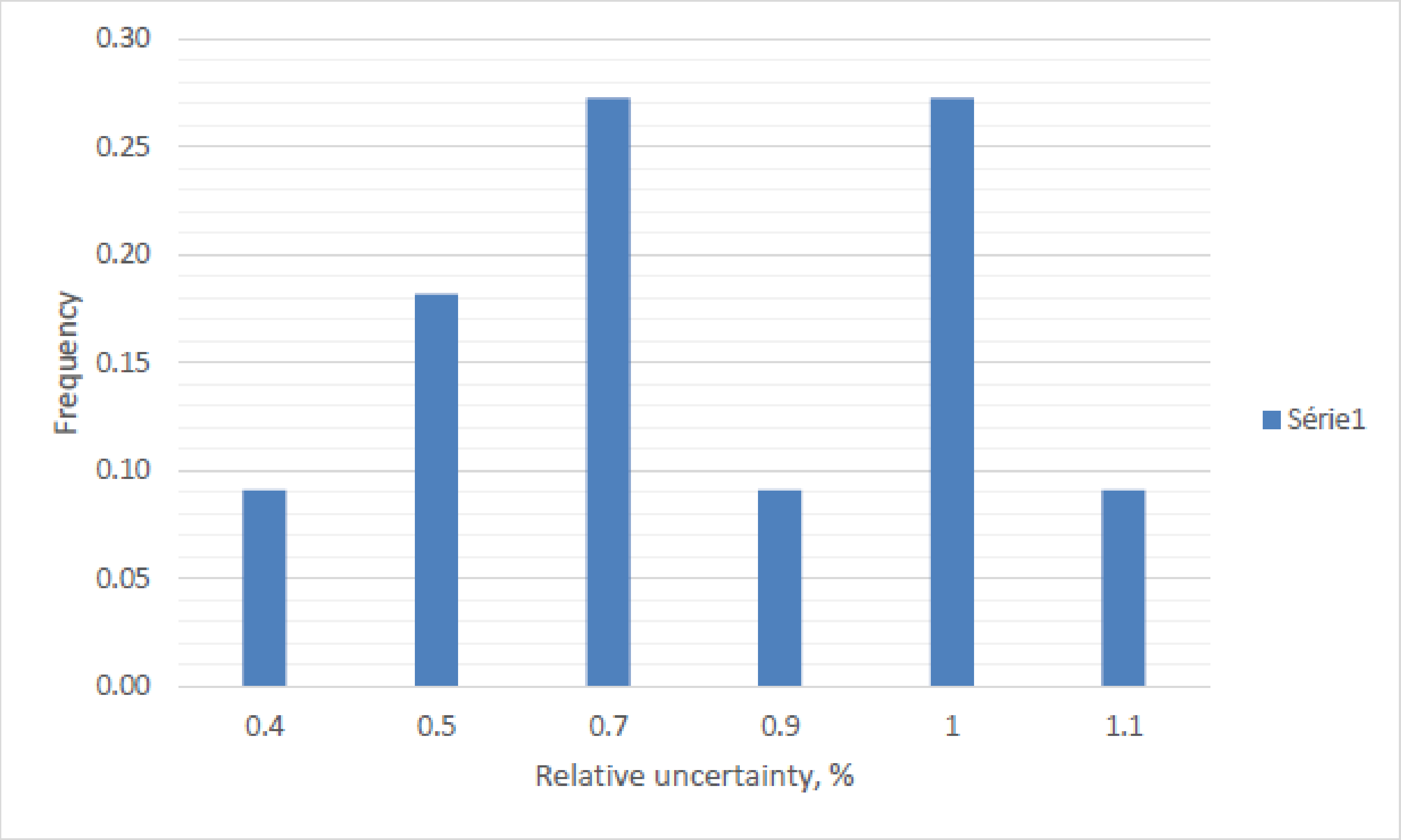
arithmetic mean of all results = 37.15(0.19) Bq mg^{-1}; $k = 1$
median of all results = 37.49(0.16) Bq mg^{-1}; $k = 1$



2018 comparison

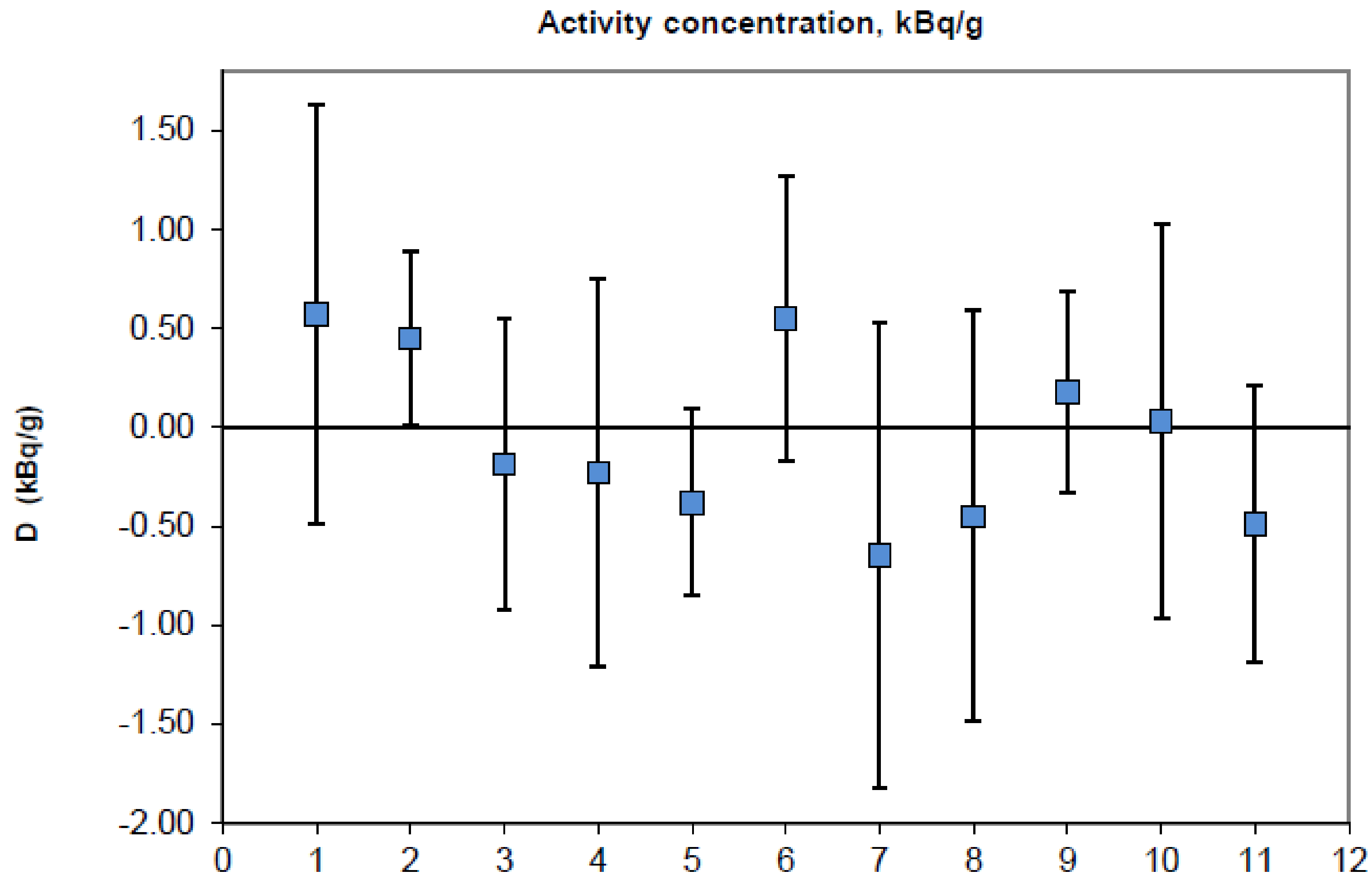
Quantity	Value, kBq/g	Standard uncertainty, kBq/g
Arithmetic mean	53.11	0.13
Weighted mean	53.17	0.13
Median	52.98	0.18
Weighted median	53.20	0.25
Power-moderated mean	**53.16**	**0.13**

Histogram of reported uncertainties



As all the laboratories used the same measurement procedure, the difference in uncertainty magnitudes deserves further investigations…

Degrees of equivalence



Conclusion of this comparison

The CCRI(II).K2.H3 comparison of tritiated water was completed successfully with the participation of eleven laboratories. No outlier was identified and the results are in good agreement with the key comparison reference value of 53.16(13) kBq/g at the reference date of 1st February 2018.

This value is traceable to the KCRV of the CCRI(II).K2.H3 comparison from 2009, with both values being in excellent agreement. This allows the comparison of 26 measurement results of a tritiated water solution.

The international reference system (SIR)

The SIR in a nutshell

BIPM.RI(II)-K1

- Highly stable comparator of activity measurements since 1976
- Total of ~ 800 SIR results for 72 different radionuclides

IG 11 – N_2 – 2 MPa

- Measurements carried out relatively to ^{226}Ra source
- SIR Result = Equivalent activity / kBq

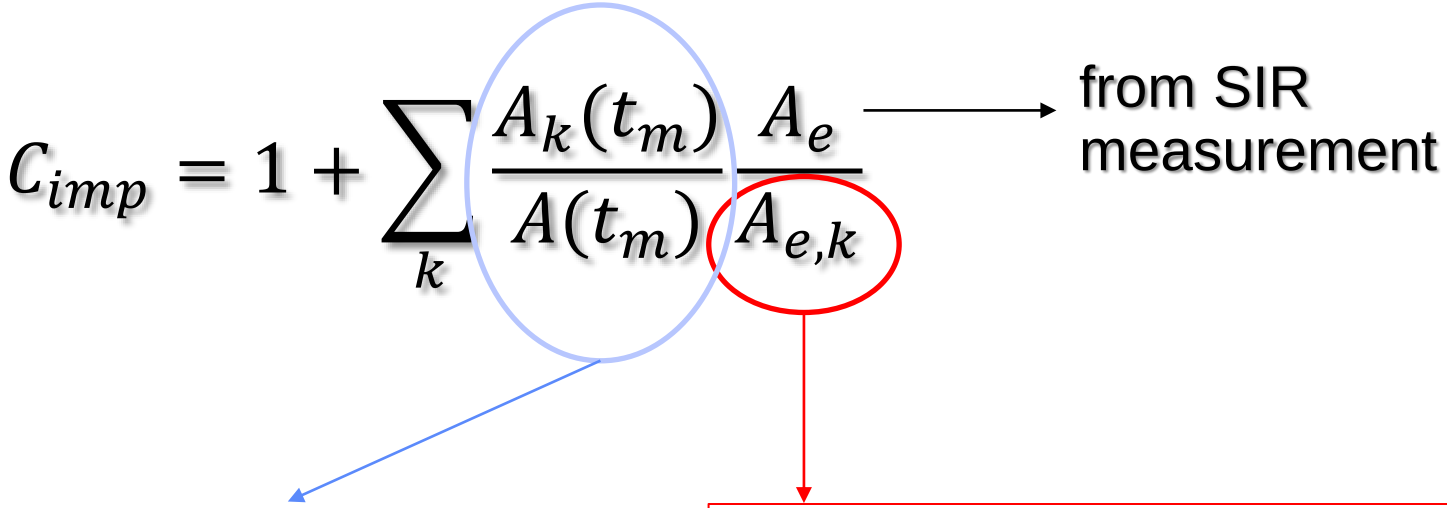
$$A_{\mathrm{e},i} = \frac{A_i(t_m)C_{\mathrm{imp}}}{I_i(t_m)/I_{\mathrm{Ra\ N°5}}(t_0)}$$

~ inverse of a calibration factor pA/MBq

To date, the following radionuclides have been measured in the SIR:

C-11 F-18 Na-22 Na-24 Sc-46 Sc-47 Cr-51 Mn-54 Mn-56 Co-56 Co-57 Co-58 Fe-59 Co-60 Cu-64 Zn-65 Ga-67 Ge-68 Se-75 Kr-85 Sr-85 Y-88 Nb-95 Mo-99 Tc-99m Ru-103 Ru-106 Cd-109 Ag-110m Ag-111 In-111 Sn-113 I-123 Sb-124 Sb-125 I-125 I-131 Ba-133 Xe-133 Cs-134 Cs-137 Ce-139 Ba-140 Ce-141 Ce-144 Eu-152 Gd-153 Sm-153 Eu-154 Eu-155 Ho-166 Ho-166m Yb-169 Lu-177 Ta-182 Re-186 Ir-192 Au-195 Tl-201 Hg-203 Pb-203 Bi-207 Rn-222 Ra-223 Th-228 Pa-231 Np-237 Am-241 Am-243.

The SIR impurity correction: how is it estimated?



$$C_{imp} = 1 + \sum_k \frac{A_k(t_m)}{A(t_m)} \frac{A_e}{A_{e,k}}$$

from SIR measurement

γ-spectrometry at NMI

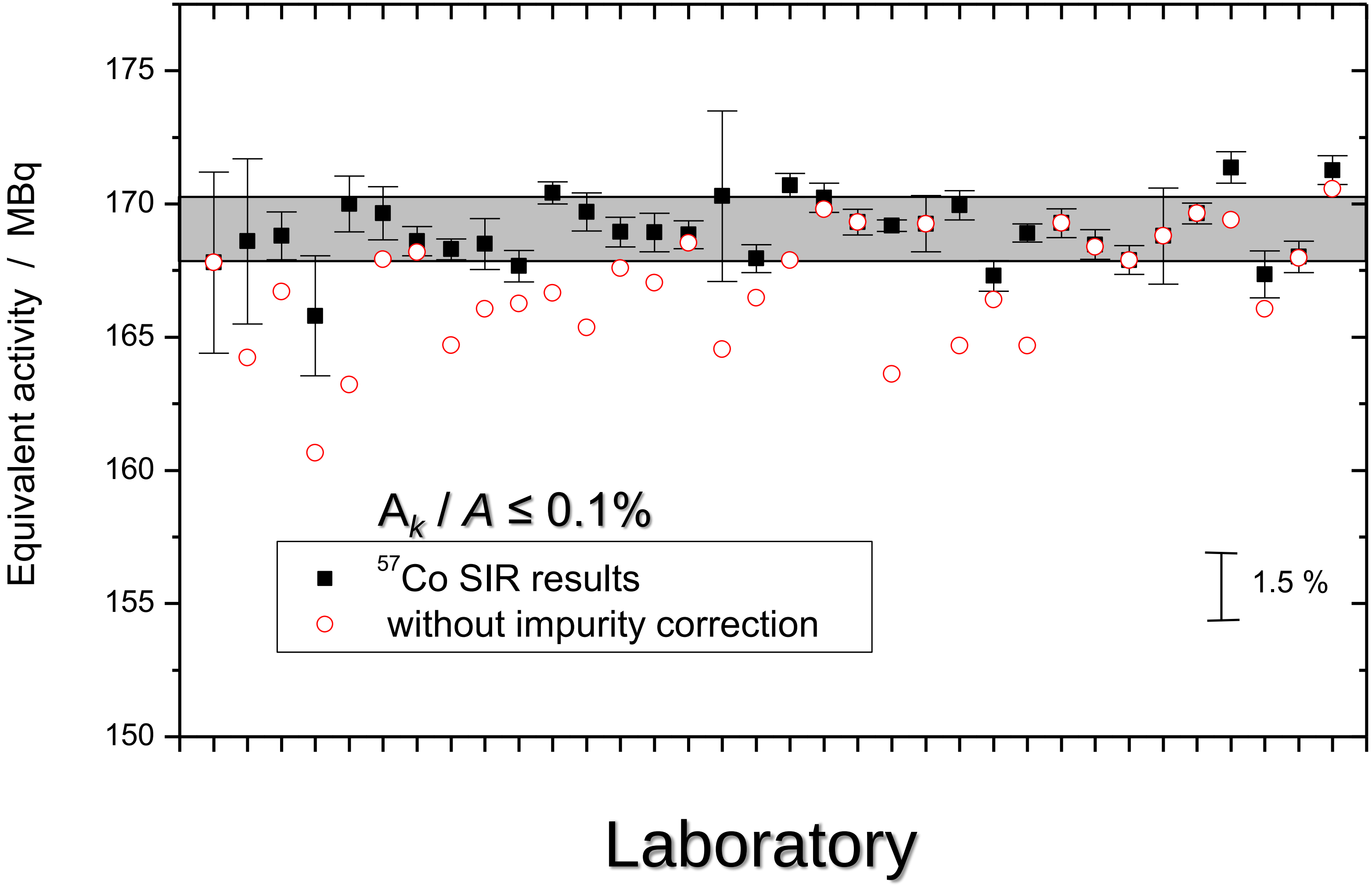
$A_{e,k}$: Equivalent activity for impurity k

obtained

- from SIR measurement
- or calculated (SIR efficiency curve + nuclear data)

The SIR impurity correction: how is it estimated?

For low-energy γ-emitter: C_{imp} reaches several (tens of) percents
precise γ-spectrometry is needed



The SIR range

- Bg current: 30 fA *u*(Bg) is supposed negligible
- SIR measurement range: from 1.5 pA to 500 pA, which corresponds to a different activity range for each RN

$$A_{\mathrm{e},i} = \frac{A_i(t_m)C_{\mathrm{imp}}}{I_i(t_m)/I_{\mathrm{Ra\ N°5}}(t_0)}$$

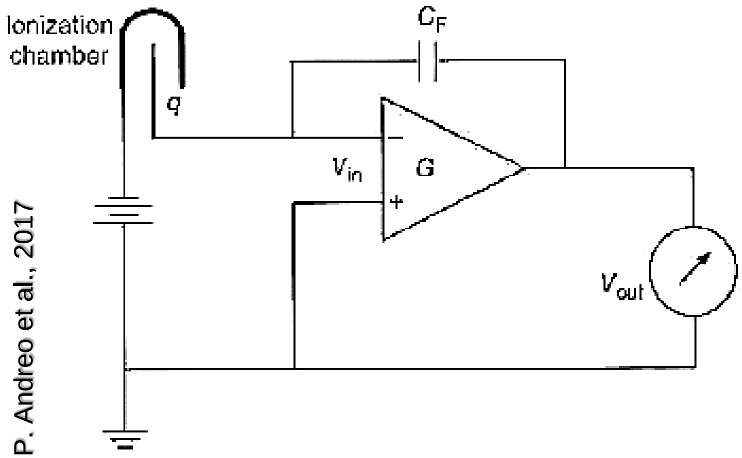
- Five ^{226}Ra sealed sources to cover the whole range
- Current ratios F_j between the 5 sources is well-known $F_j = I_{\mathrm{RaN°5}} / I_{\mathrm{RaN°j}}$

Radionuclide	Minimum A /kBq	Maximum A /MBq
Cr-51	2 600	50
Mn-54	100	29
Co-57	900	150
Co-60	40	11
Ge-68	90	20
Y-88	40	10
Cd-109	43 000	500
Cs-137	150	41
Ce-139	700	200
Eu-152	80	22
Am-241	11 000	50

Low current measurement (10 fA – 1 nA): usual techniques in IR metrology

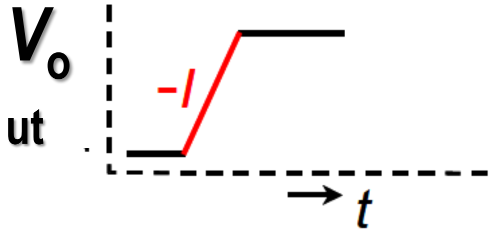
Accurate techniques are based on integrating the charge using an electrometer

Capacitors sealed-gas ,Typ. 100 pF – 20 nF



P. Andreo et al., 2017

$$\begin{cases} q = C_F \cdot \Delta V \\ I = -C_F \cdot dV/dt \end{cases}$$



2 options of measurement

Measure the time Δt to charge the capacitor until the output voltage is equal to a reference voltage ΔV

Measure the voltage versus time $V_{out}(t)$ and deduce the slope

Air conditioning is essential for high precision measurements (except for relative currents). In addition, a linear correction for Temperature dependance of C_F is usually applied.

The SIR at the BIPM: relative currents

All the IC measurements are relative to a set of reference sources (Ra-226) in order to cancel out the fluctuations ($C(T)$, V, …)

The use of a set of reference sources with activities covering the whole measurement range enables to minimize non-linearity effects which could occur when dividing currents with quite different values

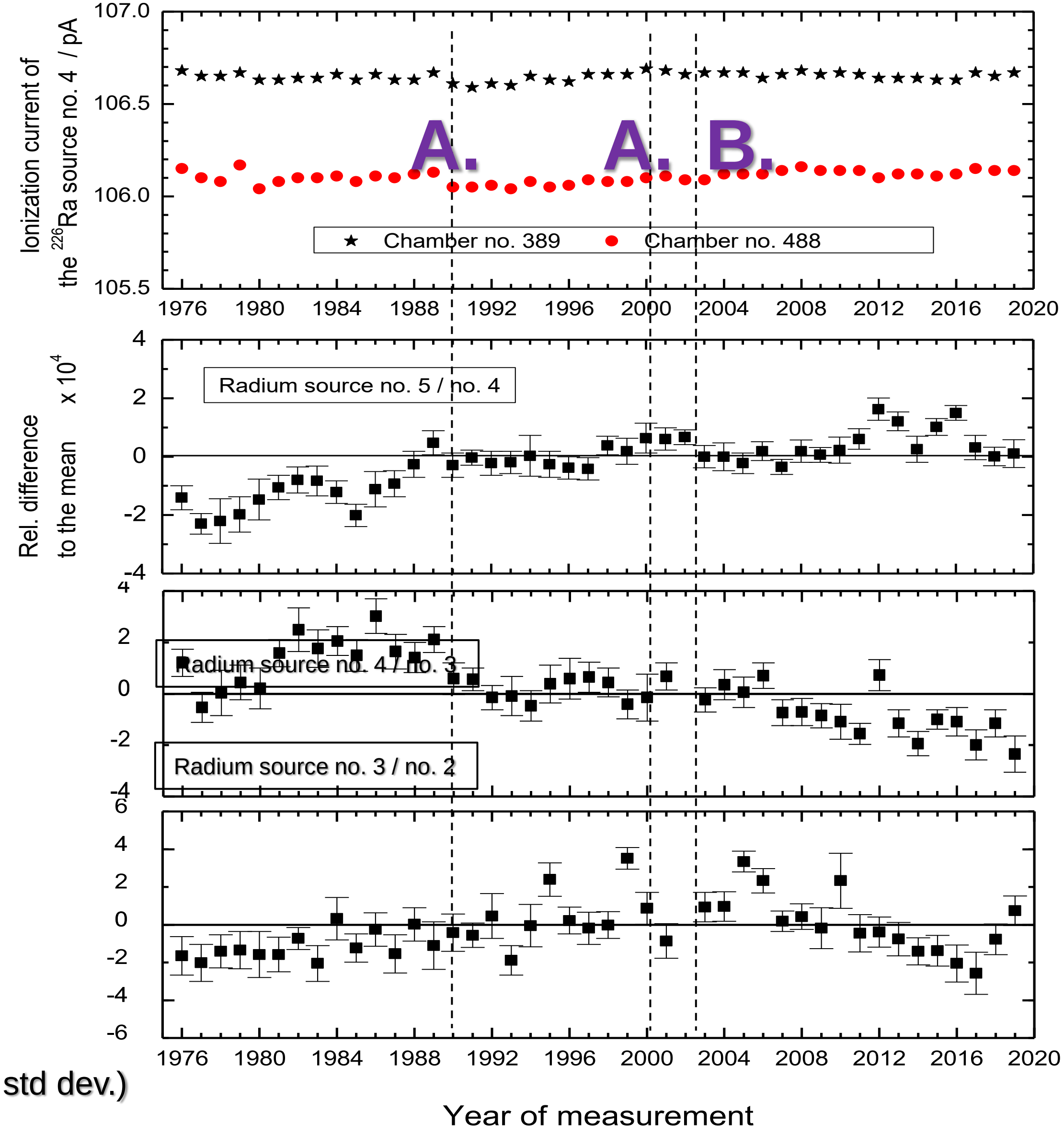
Ra-226 N° 5	I_5 300 pA
Ra-226 N° 4	I_4 100 pA
Ra-226 N° 3	I_3 30 pA
Ra-226 N° 2	I_2 10 pA
Ra-226 N° 1	I_1 3 pA

The SIR long-term stability

- ➢ ^{226}Ra current is monitored : global stability check of SIR IC, electronics and ^{226}Ra sources
- ➢ Current ratios between the ^{226}Ra sources are monitored and used in calculation of A_e

A. Move of SIR to another room
B. Complete renew of electrometer, electronics, DAQ
C. 2020 update of DAQ

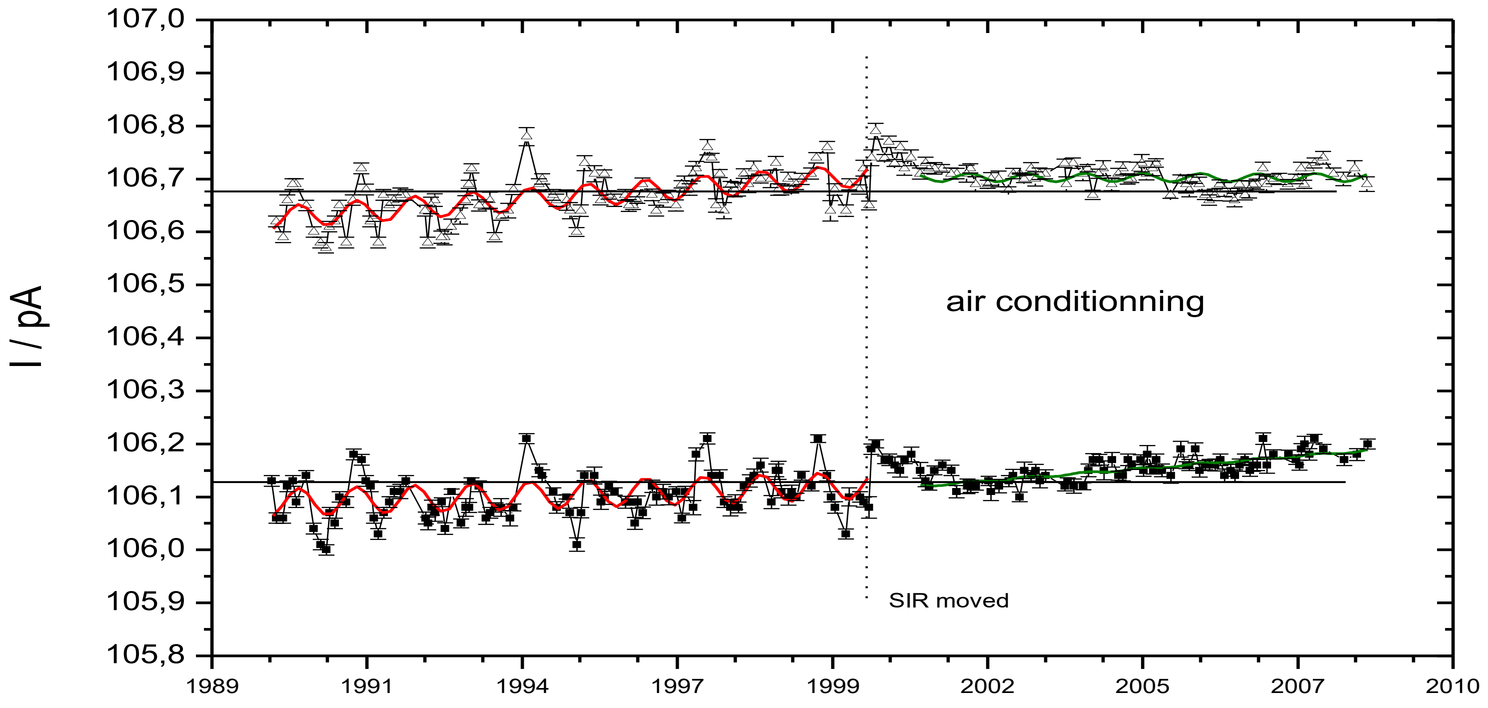
Fluctuations observed within +/- 3 x 10^{-4}



Mean over last 20 years = 106.66(3) pA (3 x 10^{-4} rel. std dev.)

Ionization chamber : stability

Annual cycles are sometimes observed and
related to the environmental conditions and/or Rn-222



Ionization chamber : stability

SHORT term instabilities related to temperature, background,… can be minimized by

- air conditioning
- short measurements (typ. < 1h)

LONG term unstabilities can be due to

- drift of electronics
- IC gas leak, geometrical drifts
- …

IC long-term stability can be monitored by

- measuring a long half-life reference source (usually Ra-226)
- checking the electrometer with an accurate electronic current source

Ionization chamber : other sources of uncertainty

- Thickness of ampoule walls for gamma energies below 50 keV :
  up to 0.1 % - 1 % depending of IC type

- Solution height and chemistry (larger effect at low energy and for β particles): the effect can reach several %

- Decay correction D: $u_{\mathrm{rel}}(D) = \ln(2)\ (t / T_{1/2}) \times u_{\mathrm{rel}}(T_{1/2})$: $10^{-5} – 10^{-3}$
  Note: for short $T_{1/2}$, the decay during measurement should be taken into account

- Impurities emitting high-energy gamma-rays may have a large impact
  => relative uncertainty on the correction up to about 1 %

The SIR uncertainty budget

Combined relative uncertainty
Typ. 0.03 – 0.15 %

Uncertainty component	Relative uncertainty / 10^{-4}	Type A/B evaluation method	Comment
Current measurement	2 - 10	A	Standard deviation of the mean
Ratio of ^{226}Ra sources F_j	0 - 7	B	Depends on ^{226}Ra source used for the measurement
Decay (RN)	Typ. < 5	B	Can reach several 10^{-3}
Decay (^{226}Ra)	0.8	B	Today
Decay (impurity)	Typ. < 1	B	
Impurity correction	Typ. < 5	A/B	Propagation of uncertainty provided by NMI Can reach several 10^{-3}
Ampoule wall thickness (liquids)	2 - 10	B	Interpolation/extrapolation of Rytz’s study using ^{241}Am and ^{60}Co
Background current	-		Negligible (within the activity range of the SIR)
Solution volume	-		Height of ampoule holder adjusted to compensate

The SIR:
case of complex decay-scheme not at equilibrium

- A_e is always calculated using the activity of the parent nuclide
- Equilibrium with daughter nuclides is assumed

- In some case, equilibrium is never reached ($A_{daughter}/A_{parent}$ is not constant), so A_e depends on time

- Specific procedure is being developed to enable comparison

- Examples: Th-227, Pa-231, Zr-95

The SIR is a reliable tool for

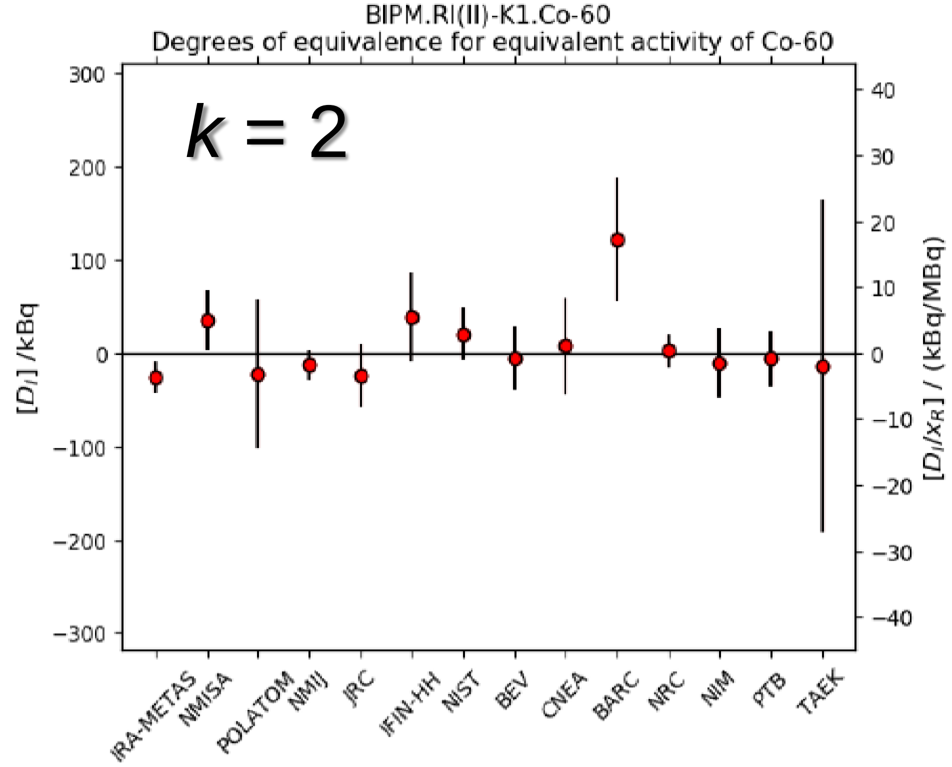
- Long-term comparison of primary/secondary activity standards
- To test/validate a new method, new nuclide…
  « Pilot comparison »: no degrees of equivalence calculated

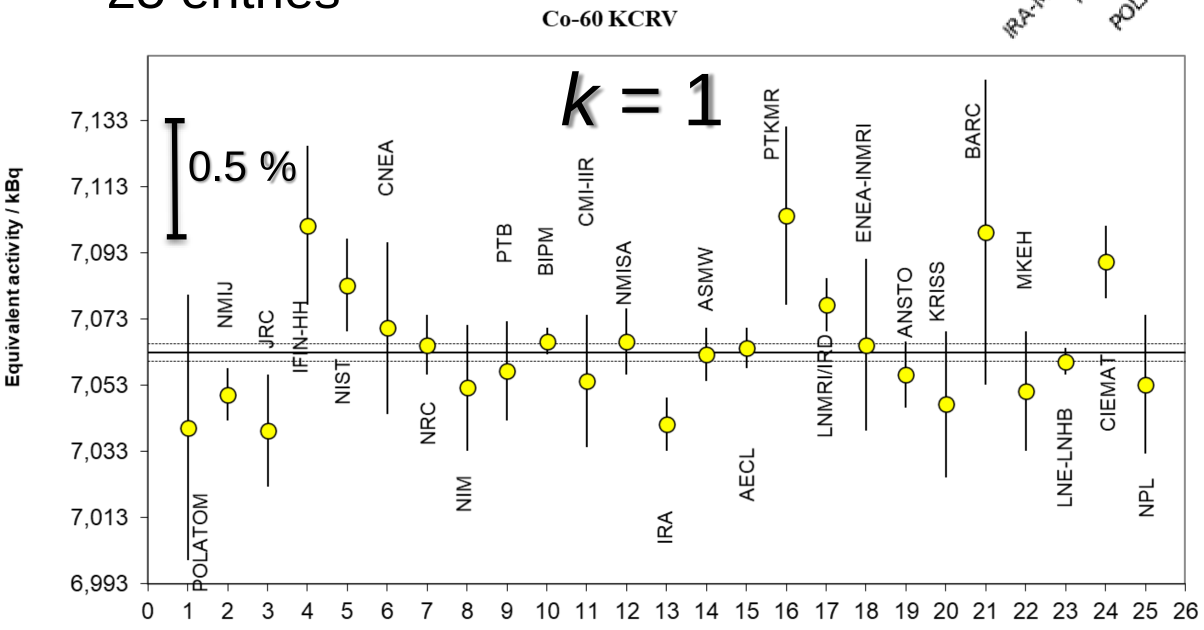
Developments

+ extension to short-lived radionuclides (SIRTI)
+ extension to beta emitters (ESIR)

Example: Co-60

- 72 ampoules measured since 1976
- 14 in the last 20 years
- KCRV: one data for each NMI
  (if primary, not outlier,…):
  25 entries





PMM	
mean	7062.7
std dev	2.7
chi	1.17
dark unc	6.4
power α	1.88

Traceability to standards

Concerns the main unit (e.g. Bq) but also all physical quantities used in the measurement:

- *mass (kg),*
- *volume (m^3),*
- *Electrical units (V or A),*
- *Time (s),*
- *Pressure (Pa),*
- *temperature (K),*
- *Etc.*

Standards

realization of the definition of a given **quantity,** with stated **quantity value** and associated **measurement uncertainty,** used as a reference *(VIM)*

Measurement methods

Examples in radionuclide metrology

- **Relative method**

$$A = A_0 \times \frac{S}{S_0}$$

A : activity of the source S

A_0 : activity of the standard

S : response of the source

S_0 : response of the standard

- **Absolute* method**

$$A_0 = \frac{S}{R}$$

R : detection efficiency

R can be:

- measured
- calculated
- extrapolated to 1

Other option: measurement independent of R

*Also called « primary » or « direct »

Activity unit history

- 1910 : from a mass 1 curie = 1 gram of radium-226
- 1934 : Hönigschmid prepared 20 standards*
- 1975 : the 15e CGPM meeting impose the becquerel
- 1982 : in France, Bq declared legal activity unit

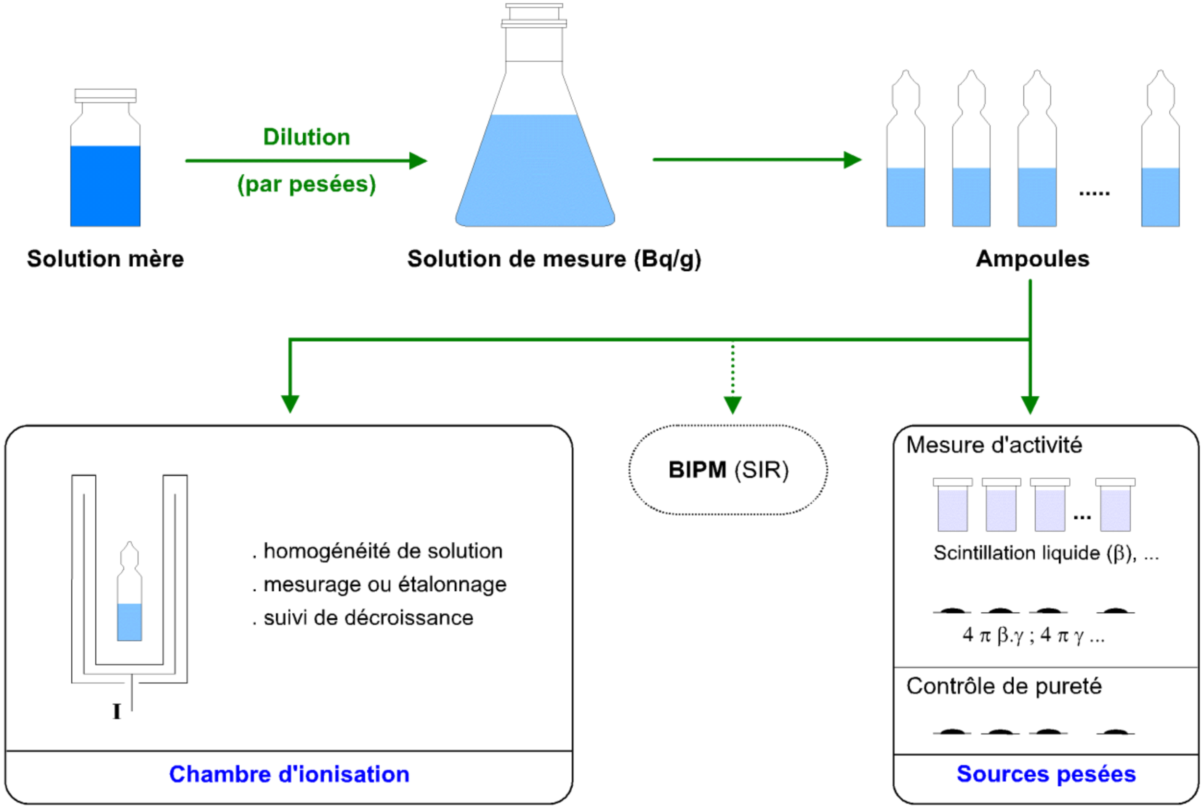
- 1993 : the Hönigschmid standard evacuated as nuclear waste!

Specificity of activity standards

- 1 Ci = 1 g of radium-226 but 5 µg of cobalt-60
- Various disintegration schemes
  - Half-life
  - Nature of emitted radiations
- Various physical forms (gas, liquid, solid)
- Problems of chemical stability
- Problems related to the dose

It is not possible to define a single activity standard

Typical standardization procedure for a solution



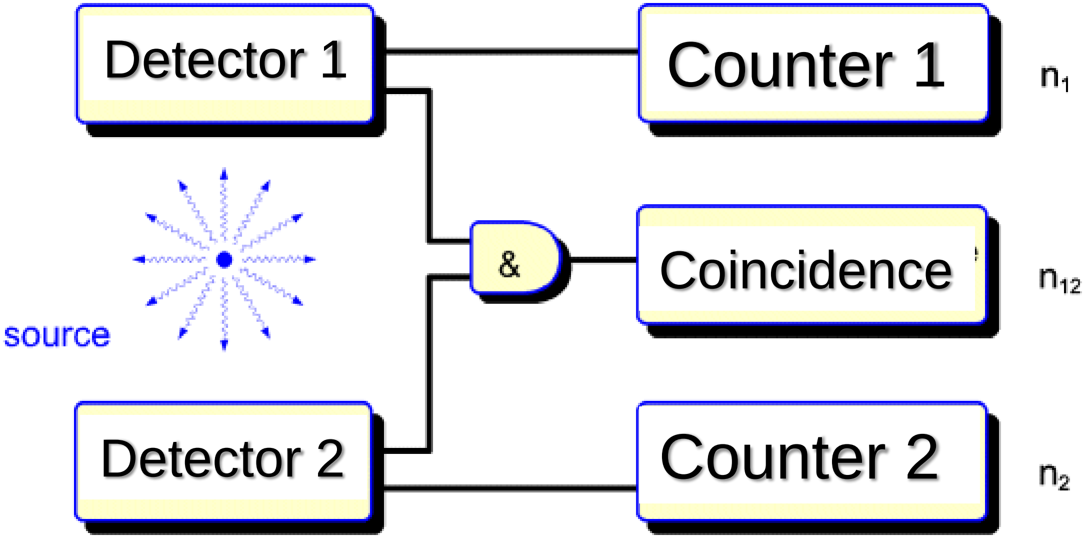
Example of absolute method: the coincidence method

For radionuclides emitting different radiations, e.g. β-γ, α-γ, …

$$n_1 = A \times R_1 \qquad n_2 = A \times R_2$$

$$n_{12} = A \times R_1 \times R_2$$

$$\frac{n_1 \times n_2}{n_{12}} = A$$


Detector 1

Counter 1

n_1

source

&

Coincidence

n_{12}

Detector 2

Counter 2

n_2

A: activity *R*: detection efficiency

More on absolute measurement methods in a next lecture

That’s all for today folks!

Благодаря за вниманието