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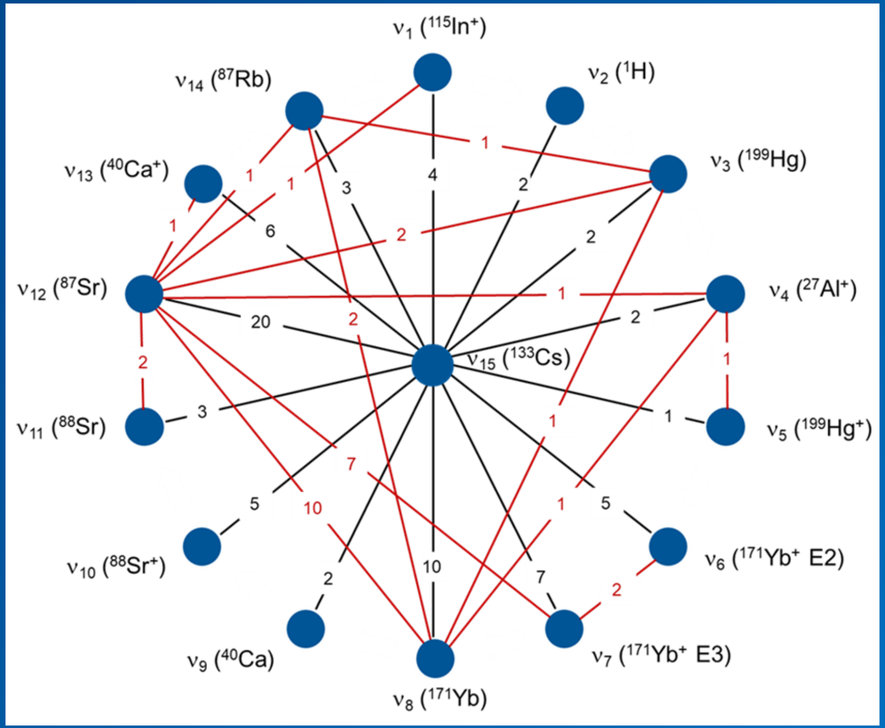
National Physical Laboratory

Least-squares analysis for optimal determination of frequency ratios



Helen Margolis

CCTF Technical Exchange on Options for Redefinition of the Second (28th April 2025)

Roadmap for redefinition of the second

CCTF

N. Dimarcq *et al*, Metrologia 61, 012001 (2024)

Achieved

In progress

- Validation that optical frequency standards (OFS) are at a level 100 times better than Cs
- Continuity with the definition based on Cs

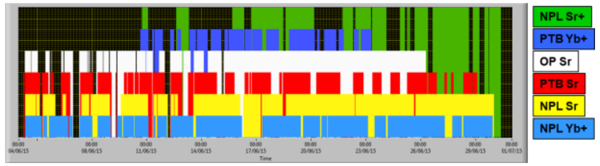
Mandatory criteria

Must be achieved before changing the definition

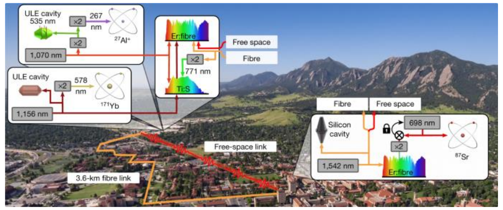
Ancillary conditions

Status should be advanced, even if not completely achieved at the time of redefinition

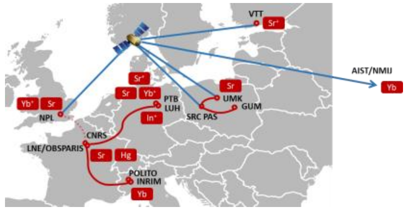
Requires comparisons between optical clocks developed independently in different laboratories around the world



ITOC collaboration
Riedel *et al*, Metrologia 57, 045005 (2020)



BACON collaboration
Nature 591, 564 (2021)

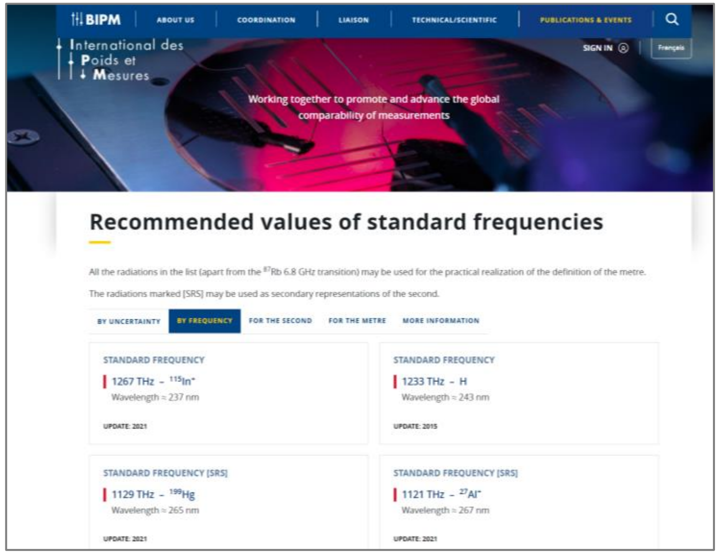


ROCIT collaboration (2022)

ICON collaboration (2023)

Recommended values of standard frequencies

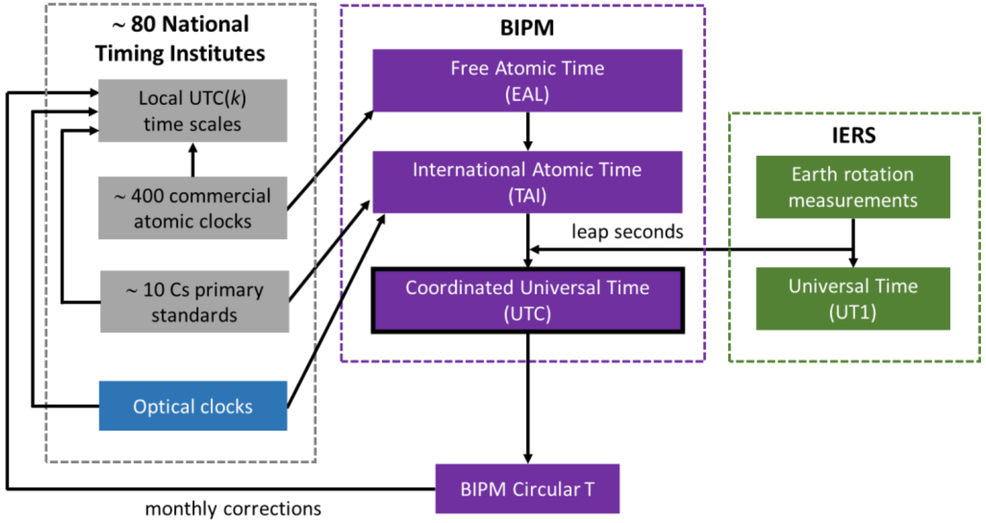
Values are periodically updated and published at
https://www.bipm.org/en/publications/mises-en-pratique/standard-frequencies



For applications including

- Practical realisation of the definition of the metre
- Secondary representations of the definition of the second (SRS)

Approved by the CCTF or CCL, based on recommendations put forward by the CCL-CCTF Frequency Standards Working Group (WGFS)



Secondary frequency standards contribute to TAI using the recommended frequency value and uncertainty of the SRS on which they are based

Clock comparisons result in an over-determined dataset

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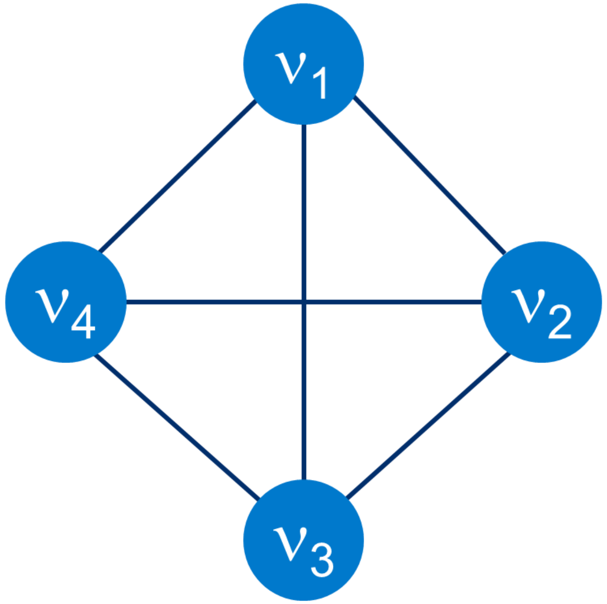
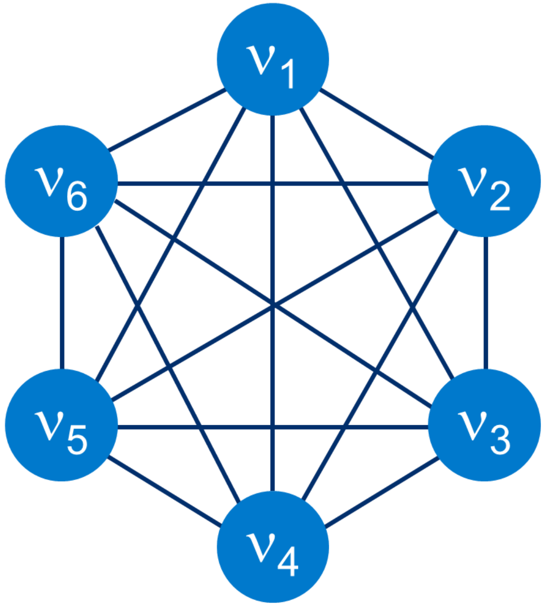
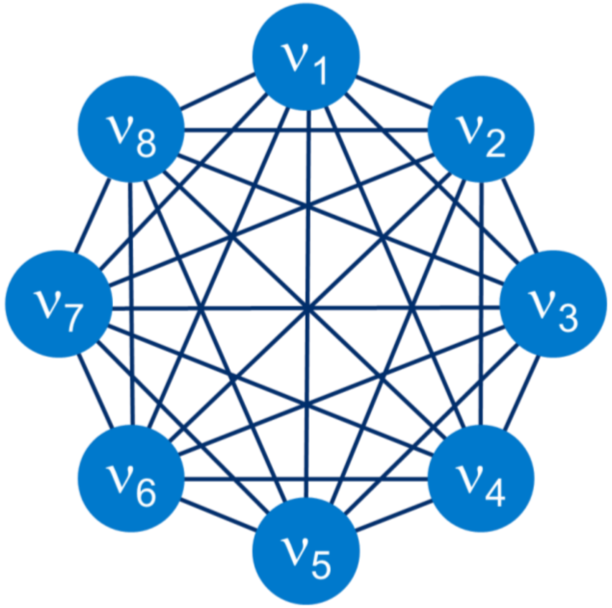


4 standards
6 ratios
3 independent ratios



6 standards
15 ratios
5 independent ratios

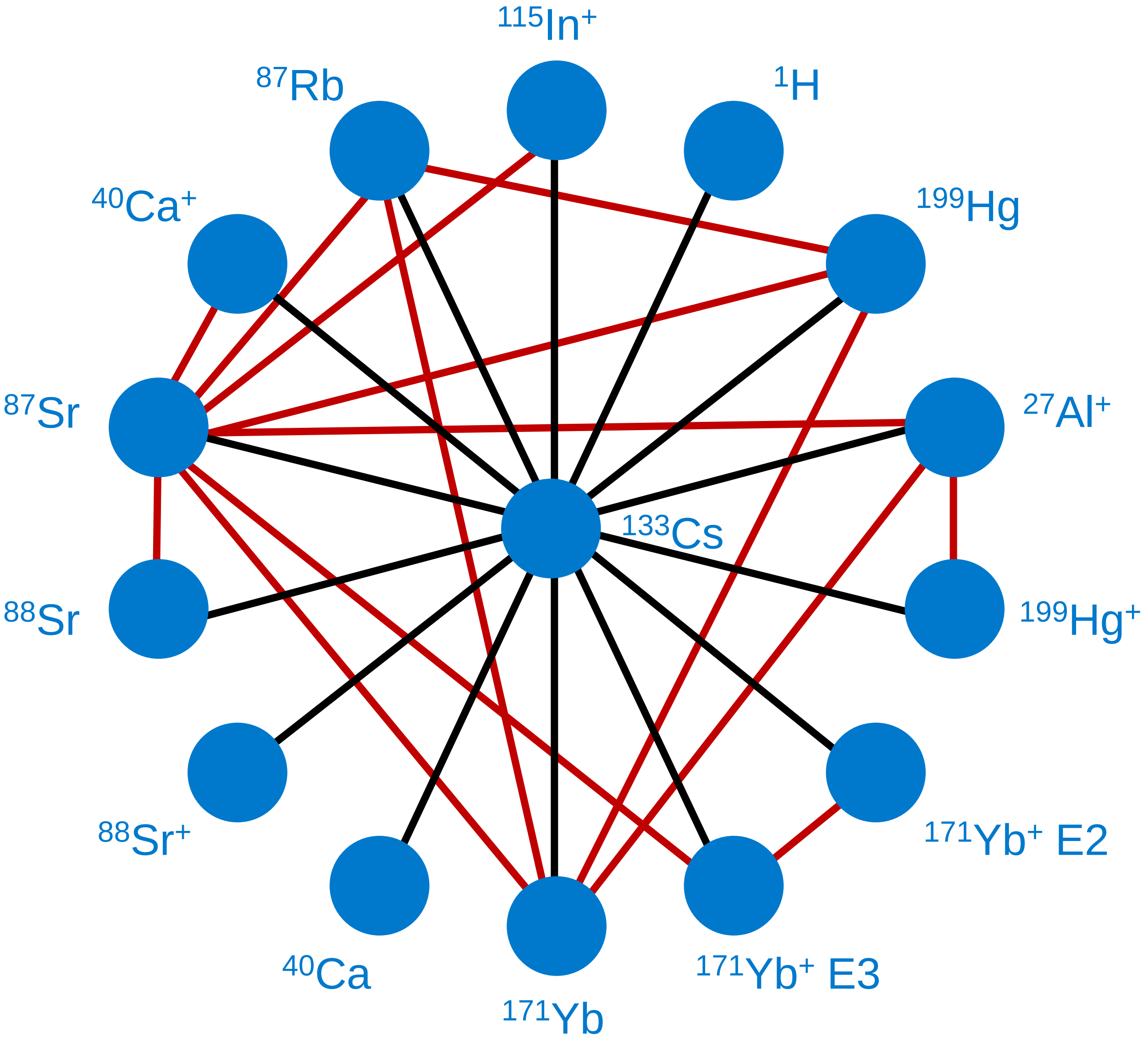


8 standards
28 ratios
7 independent ratios

For N_S different reference transitions with frequencies ν_k (k = 1, 2, … N_S),

- $N_S (N_S - 1) / 2$ different frequency ratios can be measured
- Only $N_S - 1$ of these are independent

In practice not all frequency ratios are measured



2015 — 5 directly measured optical frequency ratios

2017 — 8 directly measured frequency ratios (some more than once)

2021 — 14 directly measured frequency ratios (some more than once)

Analysis of over-determined data sets

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A new approach was needed for analysing over-determined sets of clock comparison data

a) To check the level of internal self-consistency

b) To derive optimal values for the ratios between their operating frequencies

H. S. Margolis and P. Gill, Metrologia 52, 628 (2015)

Use a **least-squares adjustment procedure**, based on the approach used by CODATA to provide a self-consistent set of recommended values of the fundamental physical constants

P. J. Mohr & B. N. Taylor, Rev. Mod. Phys. 72, 351 – 495 (2000)

All data stored as **frequency ratios**
(optical frequency ratios, microwave frequency ratios or optical-microwave frequency ratios)

Correlations between measured quantities can be included (where known)

Least-squares analysis procedure

Set of *N* measured frequency ratios, variances and covariances

N_S reference transitions

Choose set of $M = N_S - 1$ adjusted frequency ratios

Must be an independent basis set

Express measured frequency ratios in terms of adjusted frequency ratios, yielding a set of *N* (nonlinear) equations

Initial estimates of adjusted frequency ratios

Linearize equations using Taylor expansion around initial estimates of adjusted ratios

Perform least-squares adjustment

Use output from least-squares adjustment as new starting values

Best values of adjusted ratios, variances and covariances

Are adjusted frequency ratios sufficiently close to initial estimates?

no

yes

Calculate other frequency ratios and uncertainties from adjusted frequency ratios and their covariance matrix

Optimised frequency ratios

Perform self-consistency checks

Birge ratio and normalised residuals

$$R_B = \left[\frac{\chi^2}{N - M} \right]^{1/2}$$

H. S. Margolis and P. Gill, Metrologia 52, 628 (2015)

Updates to the recommended frequency values

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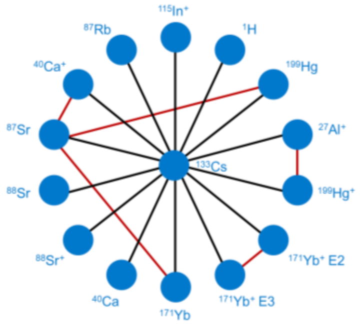
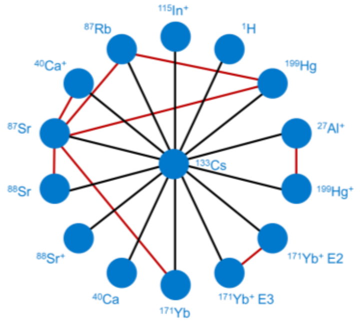
2015



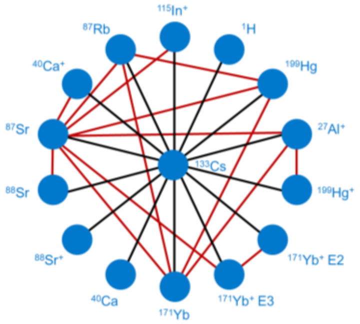
- Least-squares analysis used for the first time
- Only one algorithm / software available

2017



- 3 independent calculations using 2 different algorithms
- Correlations neglected

2021



- Correlations taken into account
- Optical frequency ratios provided as an appendix to the list
- Modified approach to assessment of input data,
  to ensure internal self-consistency of the output data set

2021 update

Analysis performed by a sub-group of the CCL-CCTF Frequency Standards Working Group (WGFS)

Sebastien Bize (LNE-SYRTE), Gianna Panfilo (BIPM), Tetsuya Ido (NICT), Gérard Petit (BIPM), Helen Margolis (NPL), Chris Oates (NIST)

+ input from Marco Pizzocaro

"The CIPM list `Recommended values of standard frequencies': 2021 update"
H. S. Margolis, G. Panfilo, G. Petit, C. Oates, T. Ido and S. Bize, Metrologia 61, 036005 (2024)

3 independent calculations using 2 different algorithms

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Algorithm 1	
Least-squares analysis H. S. Margolis and P. Gill, Metrologia 52, 628 (2015)	
Implementation A (MATLAB®)	Implementation B (Mathematica®) Chris Oates

Algorithm 2
Examination of closed loops in a graph theory framework - logarithms of frequency ratios in each closed loop should add up to zero L. Robertsson, Metrologia 53, 1272 (2016)
Implementation C (MATLAB®) Gianna Panfilo

Numerical calculations must be performed to sufficiently high precision (> 18 significant figures)

Achieved using routines designed for high precision floating point arithmetic

Implementation A v Implementation B

Differ by no more than 1 in the least-significant (24) digit of the computation

Uncertainties identical to 4 significant figures

Algorithm 1 v Algorithm 2

Frequency ratio values differ by ≤ 2 parts in 10^{21}

Uncertainties differed by ≤ 2 in the least significant digit of the 4 computed

Output correlation coefficients agreed to better than 1 part in 10^{5}

Importance of correlations

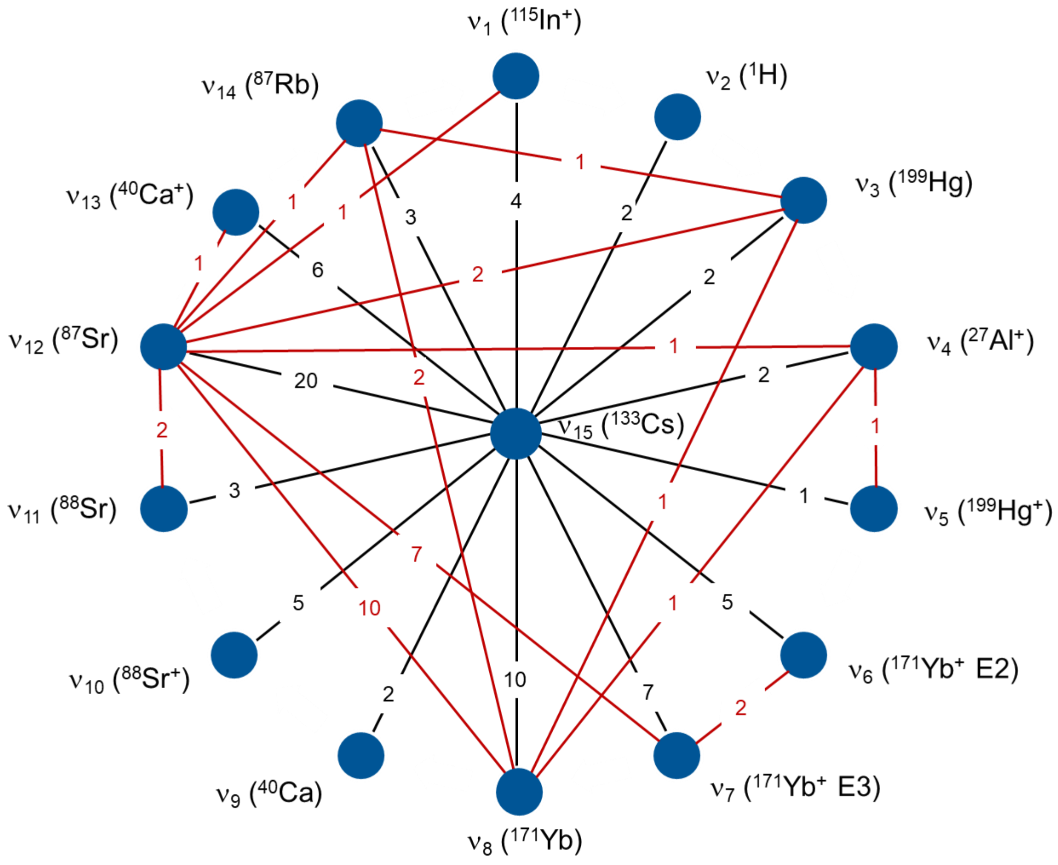
- Neglecting correlations can lead to **biased frequency values** and **underestimated uncertainties**
- Correlations can arise from both **statistical** and **systematic** uncertainties
- Consider
  a) Correlations between clocks based on the **same atomic species**
     - E.g. due to common theoretical or experimental values of atomic coefficients

  b) Correlations between **different clocks in the same institution**
     - E.g. due to common relativistic redshift correction

  c) Correlations arising from **common data**
     - Several ratio measurements involving the same clock, performed at the same time
     - Absolute frequency measurements performed using TAI as a reference,
       even if several months apart

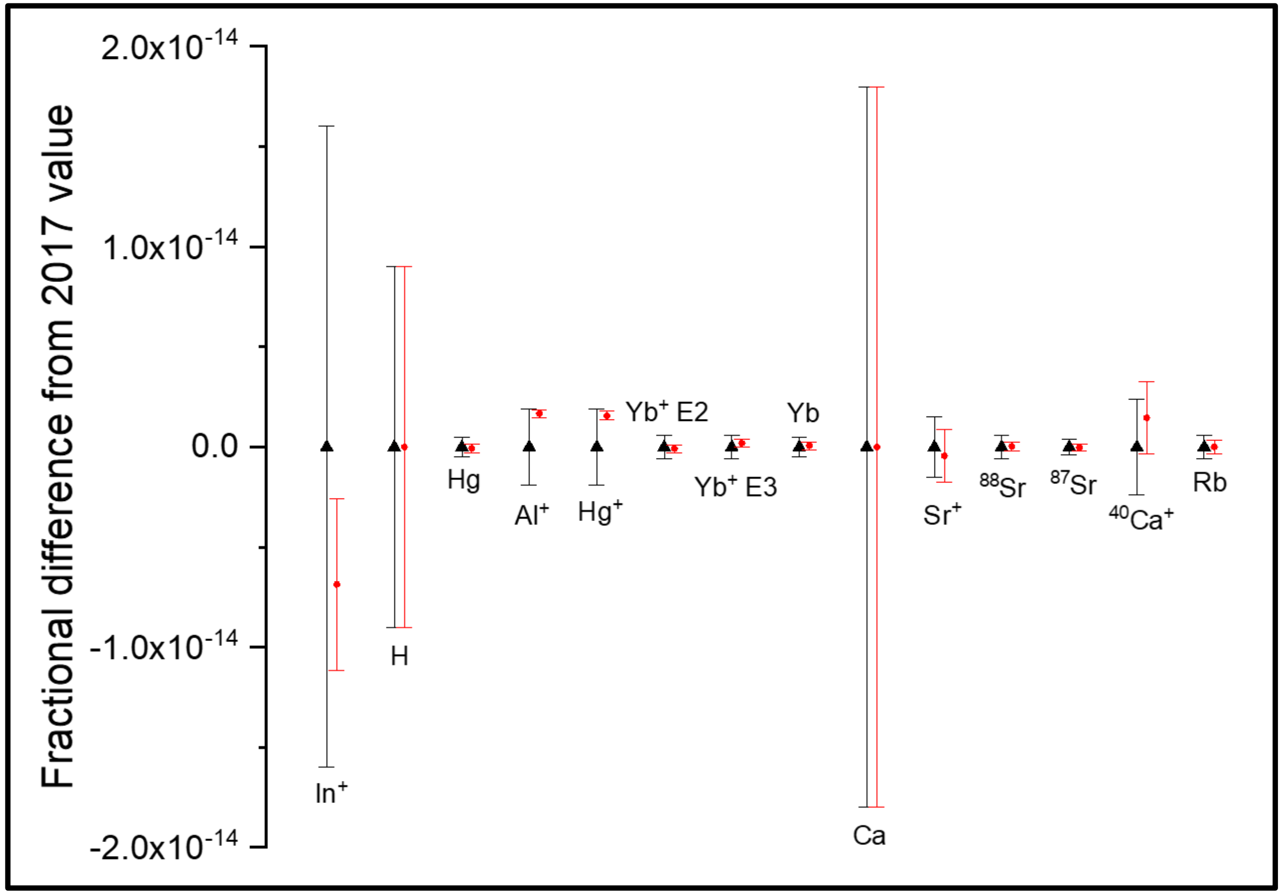
H. S. Margolis and M. Pizzocaro,
Guidelines on the evaluation and reporting of correlation coefficients between frequency ratio measurements (2020)
http://empir.npl.co.uk/rocit/project-outputs/

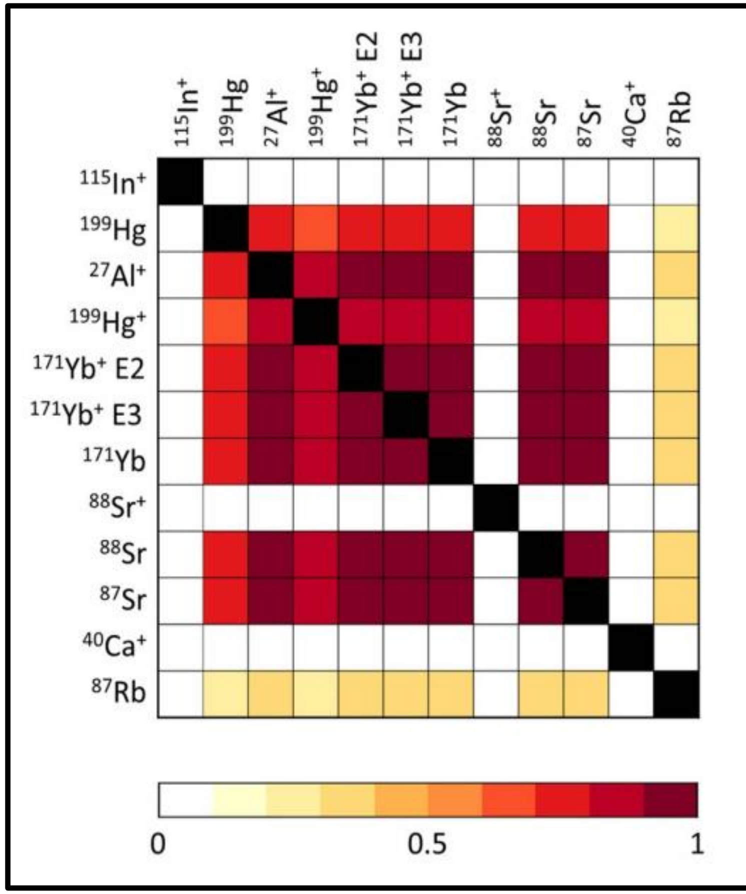
Input data for the 2021 least-squares adjustment



- 105 measurements
  (33 frequency ratios, 72 absolute frequencies)
- 483 correlation coefficients computed
  - Mostly due to use of the same primary or secondary frequency standard to access the SI second
  - 86 computed on an ad-hoc basis (common data, common coefficients…)
- Some modifications to input data compared to published results:
  - 2 data points had their uncertainty increased slightly to avoid unphysical correlation coefficients
  - 2 outliers (already present in 2017) had their uncertainty increased
  - 5 data points had their uncertainty increased due to scarcity of data in measurements that strongly influence the recommended values

2021 recommended frequency values





Correlations between recommended frequency values

- 6 secondary representations of the second now have uncertainties $\leq 2\times10^{-16}$
- Optical clocks can contribute to TAI with a similar weight to Cs primary frequency standards, if they achieve similar uptimes

Complete set of frequency ratios also provided

Consistent with the 2021 recommended frequency values, taking into account correlations between those values

Table B1. Frequency ratios consistent with the 2021 recommended frequency values, taking into account the covariance of the output matrix. (ν_2 and ν_9 are linked to the other frequencies only via Cs (ν_{15}), and hence are not included in this table.).

Clock transitions	Atomic species	Frequency ratio	Fractional uncertainty
ν_1/ν_3	$^{115}\mathrm{In}^+/^{199}\mathrm{Hg}$	1.123 010 988 476 8743(49)	4.3×10^{-15}
ν_1/ν_4	$^{115}\mathrm{In}^+/^{27}\mathrm{Al}^+$	1.130 584 343 961 8487(49)	4.3×10^{-15}
ν_1/ν_5	$^{115}\mathrm{In}^+/^{199}\mathrm{Hg}^+$	1.190 360 410 756 6604(51)	4.3×10^{-15}
ν_1/ν_6	$^{115}\mathrm{In}^+/^{171}\mathrm{Yb}^+(\mathrm{E2})$	1.841 194 044 091 2659(80)	4.3×10^{-15}
ν_1/ν_7	$^{115}\mathrm{In}^+/^{171}\mathrm{Yb}^+(\mathrm{E3})$	1.973 773 591 557 2195(85)	4.3×10^{-15}
ν_1/ν_8	$^{115}\mathrm{In}^+/^{171}\mathrm{Yb}$	2.445 326 324 126 955(11)	4.3×10^{-15}
ν_1/ν_{10}	$^{115}\mathrm{In}^+/^{88}\mathrm{Sr}^+$	2.849 510 267 459 795(13)	4.5×10^{-15}
ν_1/ν_{11}	$^{115}\mathrm{In}^+/^{88}\mathrm{Sr}$	2.952 748 322 069 815(13)	4.3×10^{-15}
ν_1/ν_{12}	$^{115}\mathrm{In}^+/^{87}\mathrm{Sr}$	2.952 748 749 874 866(13)	4.3×10^{-15}
ν_1/ν_{13}	$^{115}\mathrm{In}^+/^{40}\mathrm{Ca}^+$	3.083 388 200 597 554(14)	4.7×10^{-15}
ν_1/ν_{14}	$^{115}\mathrm{In}^+/^{87}\mathrm{Rb}$	185 436.914 199 787 30(80)	4.3×10^{-15}
ν_3/ν_4	$^{199}\mathrm{Hg}/^{27}\mathrm{Al}^+$	1.006 743 794 640 198 49(15)	1.5×10^{-16}
ν_3/ν_5	$^{199}\mathrm{Hg}/^{199}\mathrm{Hg}^+$	1.059 972 184 574 196 57(19)	1.8×10^{-16}
ν_3/ν_6	$^{199}\mathrm{Hg}/^{171}\mathrm{Yb}^+(\mathrm{E2})$	1.639 515 608 470 095 42(28)	1.7×10^{-16}
ν_3/ν_7	$^{199}\mathrm{Hg}/^{171}\mathrm{Yb}^+(\mathrm{E3})$	1.757 572 821 468 313 31(27)	1.5×10^{-16}
ν_3/ν_8	$^{199}\mathrm{Hg}/^{171}\mathrm{Yb}$	2.177 473 194 134 564 88(32)	1.5×10^{-16}
ν_3/ν_{10}	$^{199}\mathrm{Hg}/^{88}\mathrm{Sr}^+$	2.537 384 136 663 3019(34)	1.4×10^{-15}
ν_3/ν_{11}	$^{199}\mathrm{Hg}/^{88}\mathrm{Sr}$	2.629 313 828 954 238 79(40)	1.5×10^{-16}
ν_3/ν_{12}	$^{199}\mathrm{Hg}/^{87}\mathrm{Sr}$	2.629 314 209 898 909 56(39)	1.5×10^{-16}
ν_3/ν_{13}	$^{199}\mathrm{Hg}/^{40}\mathrm{Ca}^+$	2.745 643 838 071 0009(49)	1.8×10^{-15}
ν_3/ν_{14}	$^{199}\mathrm{Hg}/^{87}\mathrm{Rb}$	165 124.754 879 997 262(60)	3.6×10^{-16}
ν_4/ν_5	$^{27}\mathrm{Al}^+/^{199}\mathrm{Hg}^+$	1.052 871 833 148 990 45(11)	1.0×10^{-16}
ν_4/ν_6	$^{27}\mathrm{Al}^+/^{171}\mathrm{Yb}^+(\mathrm{E2})$	1.628 533 115 573 902 39(14)	8.3×10^{-17}
ν_4/ν_7	$^{27}\mathrm{Al}^+/^{171}\mathrm{Yb}^+(\mathrm{E3})$	1.745 799 508 102 709 104(84)	4.8×10^{-17}
ν_4/ν_8	$^{27}\mathrm{Al}^+/^{171}\mathrm{Yb}$	2.162 887 127 516 663 705(24)	1.1×10^{-17}
ν_4/ν_{10}	$^{27}\mathrm{Al}^+/^{88}\mathrm{Sr}^+$	2.520 387 163 220 7488(34)	1.3×10^{-15}
ν_4/ν_{11}	$^{27}\mathrm{Al}^+/^{88}\mathrm{Sr}$	2.611 701 053 388 596 03(10)	3.9×10^{-17}
ν_4/ν_{12}	$^{27}\mathrm{Al}^+/^{87}\mathrm{Sr}$	2.611 701 431 781 463 019(39)	1.5×10^{-17}
ν_4/ν_{13}	$^{27}\mathrm{Al}^+/^{40}\mathrm{Ca}^+$	2.727 251 811 919 3078(48)	1.8×10^{-15}
ν_4/ν_{14}	$^{27}\mathrm{Al}^+/^{87}\mathrm{Rb}$	164 018.646 808 755 766(54)	3.3×10^{-16}
ν_5/ν_6	$^{199}\mathrm{Hg}^+/^{171}\mathrm{Yb}^+(\mathrm{E2})$	1.546 753 426 486 100 05(21)	1.3×10^{-16}
ν_5/ν_7	$^{199}\mathrm{Hg}^+/^{171}\mathrm{Yb}^+(\mathrm{E3})$	1.658 131 078 387 072 22(19)	1.1×10^{-16}
ν_5/ν_8	$^{199}\mathrm{Hg}^+/^{171}\mathrm{Yb}$	2.054 273 900 601 723 59(22)	1.0×10^{-16}
ν_5/ν_{10}	$^{199}\mathrm{Hg}^+/^{88}\mathrm{Sr}^+$	2.393 821 435 684 7480(32)	1.3×10^{-15}
ν_5/ν_{11}	$^{199}\mathrm{Hg}^+/^{88}\mathrm{Sr}$	2.480 549 836 324 681 89(28)	1.1×10^{-16}
ν_5/ν_{12}	$^{199}\mathrm{Hg}^+/^{87}\mathrm{Sr}$	2.480 550 195 715 877 54(26)	1.1×10^{-16}
ν_5/ν_{13}	$^{199}\mathrm{Hg}^+/^{40}\mathrm{Ca}^+$	2.590 298 007 842 4970(46)	1.8×10^{-15}
ν_5/ν_{14}	$^{199}\mathrm{Hg}^+/^{87}\mathrm{Rb}$	155 782.158 516 102 797(54)	3.5×10^{-16}
ν_6/ν_7	$^{171}\mathrm{Yb}^+(\mathrm{E2})/^{171}\mathrm{Yb}^+(\mathrm{E3})$	1.072 007 373 634 205 473(73)	6.9×10^{-17}
ν_6/ν_8	$^{171}\mathrm{Yb}^+(\mathrm{E2})/^{171}\mathrm{Yb}$	1.328 119 831 787 671 42(11)	8.3×10^{-17}
ν_6/ν_{10}	$^{171}\mathrm{Yb}^+(\mathrm{E2})/^{88}\mathrm{Sr}^+$	1.547 642 561 958 3136(21)	1.3×10^{-15}
ν_6/ν_{11}	$^{171}\mathrm{Yb}^+(\mathrm{E2})/^{88}\mathrm{Sr}$	1.603 713 813 623 139 52(14)	8.9×10^{-17}
ν_6/ν_{12}	$^{171}\mathrm{Yb}^+(\mathrm{E2})/^{87}\mathrm{Sr}$	1.603 714 045 975 103 00(13)	8.2×10^{-17}
ν_6/ν_{13}	$^{171}\mathrm{Yb}^+(\mathrm{E2})/^{40}\mathrm{Ca}^+$	1.674 667 703 001 0606(30)	1.8×10^{-15}
ν_6/ν_{14}	$^{171}\mathrm{Yb}^+(\mathrm{E2})/^{87}\mathrm{Rb}$	100 715.573 567 538 329(34)	3.4×10^{-16}
ν_7/ν_8	$^{171}\mathrm{Yb}^+(\mathrm{E3})/^{171}\mathrm{Yb}$	1.238 909 231 832 259 428(59)	4.7×10^{-17}
ν_7/ν_{10}	$^{171}\mathrm{Yb}^+(\mathrm{E3})/^{88}\mathrm{Sr}^+$	1.443 686 489 498 3514(19)	1.3×10^{-15}
ν_7/ν_{11}	$^{171}\mathrm{Yb}^+(\mathrm{E3})/^{88}\mathrm{Sr}$	1.495 991 401 800 156 824(86)	5.8×10^{-17}
ν_7/ν_{12}	$^{171}\mathrm{Yb}^+(\mathrm{E3})/^{87}\mathrm{Sr}$	1.495 991 618 544 900 552(68)	4.6×10^{-17}
ν_7/ν_{13}	$^{171}\mathrm{Yb}^+(\mathrm{E3})/^{40}\mathrm{Ca}^+$	1.562 179 276 177 7189(28)	1.8×10^{-15}
ν_7/ν_{14}	$^{171}\mathrm{Yb}^+(\mathrm{E3})/^{87}\mathrm{Rb}$	93 950.448 518 001 415(31)	3.3×10^{-16}
ν_8/ν_{10}	$^{171}\mathrm{Yb}/^{88}\mathrm{Sr}^+$	1.165 288 345 913 1553(16)	1.3×10^{-15}
ν_8/ν_{11}	$^{171}\mathrm{Yb}/^{88}\mathrm{Sr}$	1.207 506 864 395 296 327(46)	3.8×10^{-17}
ν_8/ν_{12}	$^{171}\mathrm{Yb}/^{87}\mathrm{Sr}$	1.207 507 039 343 337 845(16)	1.3×10^{-17}

(Continued.)

Table B1. (Continued.)

Clock transitions	Atomic species	Frequency ratio	Fractional uncertainty
ν_8/ν_{13}	$^{171}\mathrm{Yb}/^{40}\mathrm{Ca}^+$	1.260 931 177 231 8993(22)	1.8×10^{-15}
ν_8/ν_{14}	$^{171}\mathrm{Yb}/^{87}\mathrm{Rb}$	75 833.197 545 114 200(25)	3.3×10^{-16}
ν_{10}/ν_{11}	$^{88}\mathrm{Sr}^+/^{88}\mathrm{Sr}$	1.036 230 104 446 0007(14)	1.3×10^{-15}
ν_{10}/ν_{12}	$^{88}\mathrm{Sr}^+/^{87}\mathrm{Sr}$	1.036 230 254 578 8345(14)	1.3×10^{-15}
ν_{10}/ν_{13}	$^{88}\mathrm{Sr}^+/^{40}\mathrm{Ca}^+$	1.082 076 536 381 8990(24)	2.2×10^{-15}
ν_{10}/ν_{14}	$^{88}\mathrm{Sr}^+/^{87}\mathrm{Rb}$	65 076.766 459 625 929(88)	1.4×10^{-15}
ν_{11}/ν_{12}	$^{88}\mathrm{Sr}/^{87}\mathrm{Sr}$	1.000 000 144 883 682 799(36)	3.6×10^{-17}
ν_{11}/ν_{13}	$^{88}\mathrm{Sr}/^{40}\mathrm{Ca}^+$	1.044 243 485 823 4592(18)	1.8×10^{-15}
ν_{11}/ν_{14}	$^{88}\mathrm{Sr}/^{87}\mathrm{Rb}$	62 801.462 899 418 361(21)	3.3×10^{-16}
ν_{12}/ν_{13}	$^{87}\mathrm{Sr}/^{40}\mathrm{Ca}^+$	1.044 243 334 529 6392(18)	1.8×10^{-15}
ν_{12}/ν_{14}	$^{87}\mathrm{Sr}/^{87}\mathrm{Rb}$	62 801.453 800 512 449(21)	3.3×10^{-16}
ν_{13}/ν_{14}	$^{40}\mathrm{Ca}^+/^{87}\mathrm{Rb}$	60 140.631 712 818 40(11)	1.8×10^{-15}

- Recommended frequency values are enough for contributions of secondary frequency standards to TAI
- Frequency ratios need to be considered when discussing options for redefinition of the second

Appendix B
H. S. Margolis *et al*. Metrologia 61, 036005 (2024)

Options for redefinition of the second

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Option 1

- Choose a single optical transition to replace the Cs hyperfine transition
- Fix the numerical value of the frequency of this transition: $\nu_{Xy} = N$ Hz, where N is the defining value

Option 2

- Create a defining constant based on several transitions rather than just a single one
- Quantity whose numerical value is used in the definition is a weighted geometric mean of the frequency of an ensemble of chosen transitions
- Unit of time set by the relation $\prod_i \nu_i^{w_i} = N$ Hz, where w_i and N are the defining values, and $\sum_i w_i = 1$

Option 2a

Fixed values of w_i and N

Option 2b

Dynamic defining values w_i and N periodically updated by the CIPM, following predefined rules adopted by the CGPM

But whichever option is selected

- One or more of the frequency ratios from a least-squares adjustment
will be used to set the defining constant or constants appearing in the new definition

- The frequency ratios from the least-squares adjustment will play a key role
in the *Mise en pratique* for the new definition

More scrutiny is needed on the evolution of the frequency ratio values

to ensure the stability of the new definition and realisation of the second

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