New determination of the ^{3}He mixing ratio in the Earth's lower atmosphere from an international tritium intercomparison exercise

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Abstract.

The light isotope of helium, ^{3}He, is essentially a primordial substance entrapped within the Earth's interior during the formation of the planet. It is released into the atmosphere by volcanic/magmatic activity, and eventually escapes to outer space. ^{3}He is also produced by the radioactive beta-decay of tritium. Hence, measurements of ^{3}He can be used to derive the concentration of tritium. The so-called "^{3}He ingrowth" method uses a mass spectrometer to detect the amount of ^{3}He that accumulates in a sample during a given period of storage. The ^{3}He measurements are classically calibrated against an air standard. The method thus relies on the accurate knowledge of the atmospheric mixing ratio of ^{3}He. This value is based on mass spectrometric measurements with gravimetrically or volumetrically prepared ^{3}He standard mixtures. Here, we apply the ^{3}He ingrowth method in reverse, using a solution of tritiated water prepared for an international comparison of tritium activity measurements to precisely determine the ^{3}He mixing ratio of our air standard. The measured atmospheric mixing ratio of ^{3}He, based on a series of ten measurements, is $[^3He]=7.12 \pm 0.06$ ppt. This value is between 1% and 2% lower than previous determinations reported in the literature. However, all results remain statistically consistent.

Keywords: Helium-3, tritium, mass spectrometry

1. Introduction.

In the ^{3}He ingrowth method, the activity of tritium is determined via the mass spectrometric measurement of the helium-3 (^{3}He) produced by the radioactive decay of tritium (Clarke et al., 1976). Because the atmospheric residence time of helium (~10^6 years) is much longer than the mixing time of the atmosphere (~10 years only), the ^{3}He content of the atmosphere is the same over the whole globe (Glückhauf and Paneth, 1946; Jean-Baptiste and Fourré, 2012). Air is thus a convenient ^{3}He standard, that is classically used by the tritium laboratories applying the ^{3}He ingrowth technique. The value of the ^{3}He mixing ratio in atmospheric air is therefore an important parameter because any uncertainty in this value directly affects the measured tritium concentrations. This value appears to be reasonably well known : One direct determination was by Davidson and Emerson, 1990. They used mass spectrometry with gravimetrically prepared ^{3}He standards and found a value of 7.27±0.20 ppt; other relevant publications (Table 1), which concern the ^{4}He content of air (Glückauf, 1944; Glückauf, 1946; Oliver et al., 1984; Holland and Emerson, 1987) and the ^{3}He/^{4}He ratio of air (Mamyrin et al., 1970; Clarke et al., 1976; Sano et al., 1988; Hoffman and Nier, 1993) lead to a mean value of 7.21 with a standard uncertainty of the mean equal to ±0.07 ppt (calculated by a bootstrap technique). However, all these determinations rely on gravimetrically or volumetrically prepared ^{3}He mixtures. Therefore, we thought that determining the atmospheric mixing ratio of ^{3}He using a totally independent method would be of some interest. Because we routinely use the ^{3}He ingrowth method for tritium measurements (Jean-Baptiste et al., 1992; 2010), we were able to apply the ^{3}He ingrowth method in reverse, using a solution of tritiated water prepared for an international comparison of tritium activity measurements to determine the ^{3}He mixing ratio of our air standard.

2. The CCIR(II) 2009 International Tritium Intercomparison

An international comparison of activity measurements of a solution of tritiated water was organised by the Consultative Committee for Ionizing Radiation (CCIR) in 2009. The tritiated water was prepared and dispatched to the participants by the Laboratoire National Henri Becquerel (LNHB), the French National Metrology Institute for radioactivity standards. Fifteen laboratories took part in the exercise (Table 2) and reported a total of 22 independent results. Most laboratories applied the triple-to-double coincidence ratio (TDCR) method in liquid scintillation counting (LSC). The TDCR method is a primary standardization method of measuring radioactivity, mainly used by metrology laboratories. It requires a special liquid scintillation (LS) counter equipped with three photomultipliers (A, B and C) in coincidence.

Such instrumentation is generally designed and constructed by national metrology laboratories around the world. The TDCR system allows the observation of double AB, BC, and CA and triple ABC coincidences, making it possible to measure the absolute activity of the radioactive solution, independently of the detection efficiency (Broda et al., 1988 ; Broda and Pochwalski, 1992 ; Cassette and Vatin, 1992; Stanga et al., 2006).

The reference time and date used were 00:00 UTC, 31 May 2009 -. The Power-Moderated weighted Mean formalism – PMM (Pommé, 2012) was used to evaluate the Key Comparison Reference Value (KCRV). The value given by the MKEH (Budapest, Hungary) was identified as an outlier by the PMM procedure. In addition, the results of BARC, IRMM obtained by methods using ^{3}H external standards were not used to calculate the KCRV The KCRV calculated using the PMM method is thus 37.10 ± 0.19 Bq/mg, corresponding to a relative uncertainty of 0.51% (Ratel and Michotte, in prep.). A summary of the results is given in Fig. 1 and Table 2.

3. Method

3.1 Dilution Procedure

Half a liter of a tritium solution was prepared from the tritiated water distributed for the CCIR(II) 2009 International Tritium Intercomparison by two-step gravimetric dilution with “tritium-free” groundwater (0.09 ± 0.01 Bq/kg). First, in a glove box flushed with argon, a few drops (113.05±0.01 mg) of the initial tritiated water were added with a syringe to 11.02±0.02 g of “tritium-free” water. Then, in the same way, 135.52±0.01 mg of this water was in turn added to 473.20±0.02 g of “tritium-free” water, leading to a reduction factor of the initial tritium concentration of $(3.435\pm0.007)\times10^{5}$ and a final solution of ~100 Bq/kg. The weighing process consisted in weighing the empty and the filled containers and, in case of the tritium standard, the transfer container (syringe) before and after the transfer. All dilutions were performed using calibrated balances and calibrated weights traceable to the French national mass standards, checking the zero reading and making test weights before and after each weighing process. Buoyancy corrections were applied. The combined uncertainty of the weighing procedure was calculated according to the Guide to the expression of uncertainty in measurement (JCGM, 2008).

3.2 Sample preparation and ^{3}He mass spectrometry

Ten 100 ml low-helium diffusivity Corning 1724 glass bulbs were filled with approximately 40 g of the prepared standard solution. To minimize ^{3}He blank, the bulbs were

pre-baked in a flow of argon at 600°C for 24 hours to remove the helium dissolved in the glass. The samples were degassed using a routine procedure (Jean-Baptiste et al., 1992). However, to avoid any bias due to isotope fractionation that might have occurred during pumping, a special water trap at 77K was inserted between the sample and the pumping unit to trap water vapor and to transfer it back into the sample at the end of the degassing.

The storage time was about 120 days. ^{3}He which accumulated during this period of time was measured with a MAP-215-50 mass spectrometer (Jean-Baptiste et al., 2010). Classically, along with the tritiugenic ^{3}He signal, a small ^{4}He signal ($n^4He_{res} < 5\times10^{-14}$ mol) can be detected as well. This ^{4}He corresponds to the small air residue left behind at the end of the degassing step, but also, to a minor extent, to diffusion through the glass bulb (Jean-Baptiste et al., 1989) and leaking through adapter fittings. The corresponding ^{3}He signal ($n^3He_{res} < 7\times10^{-20}$ mol, i.e., representing about 0.08% of the measured ^{3}He signal) can be calculated by applying the ^{3}He/^{4}He ratio of this air residue, R_{res}. The statistics of measurements on samples analysed immediately following degassing (i.e., with no tritiugenic ^{3}He contribution) show that R_{res} is identical to the ^{3}He/^{4}He ratio of air, R_a , within $\pm$10%. Hence R_{res} can be written as follows: $R_{res} = \alpha R_a$, with α=1.0$\pm$0.1. The ^{3}He blank of the inlet system, n^3He_{blank} is less than 5×10^{-20} mol. Most of it is due to the O'ring which is used to attach the glass bulb to the inlet system. The ^{3}He/^{4}He ratio of this line blank is not statistically different from R_a , hence we assume that $R_{blank} = R_{res} = \alpha R_a$.

The measurements are calibrated against an air standard drawn from a 5 L tank filled with clean air at known pressure, temperature and relative humidity conditions, through a precisely calibrated volume V=122.2 $\pm$ 0.1 (1 σ) mm^3 (Jean-Baptiste et al., 2014). The total number of aliquots withdrawn from the 5 L tank is recorded in order to take into account the decreasing pressure in the tank. $^4He^+$ is measured on a Faraday cup. The $^3He^+$ ion beam (~250 ions/sec for an air standard with about 3.7×10^{-17} ^{3}He mole) passes through an electrostatic filter before impinging the detector (a 16-stage electron multiplier connected to a pulse counting system). The ^{3}He background is very low (< 0.05 count/sec) when employing the electrostatic filter, which constitutes the ultimate limit for the ^{3}He detection. The counting procedure consists of five series of counting of the $^3He^+$ signal interspaced by five measurements of the $^4He^+$ peak and baseline. Each series consists of ten 10-seconds integrations on the $^3He^+$ peak and on the baseline, corresponding to a total counting time of 500 sec on the peak and 500 sec on the baseline.

In routine operation, each sample measurement is followed by a standard air aliquot (Jean-Baptiste et al., 1992; 2010). A correction is routinely applied to take into account the pressure effect in the source due to the fact that the sample contains some 10^{-14} mol of gas only whereas the air aliquot contains about 10^{-10} mol of helium and neon (neon is not

retained by the charcoal trap at 77K). For this special experiment however, to avoid any additional uncertainty arising from this correction, an aliquot of the air standard was introduced in the inlet line with each sample so as to keep the pressure in the source identical for the standard and for the sample.

With this special protocol, the amount of helium-3 (n^3He_{meas}) and helium-4 (n^4He_{meas}) corresponding to the (sample+standard) measurement obeys the following equations:

$$n^3He_{meas} = n^3He_{tri} + n^3He_{std} + n^3He_{res} + n^3He_{blank} \quad (1)$$

$$n^4He_{meas} = n^4He_{std} + n^4He_{res} + n^4He_{blank} \quad (2)$$

where n^3He_{std} and n^4He_{std} are the ^3He and ^4He amount in the air standard aliquot, respectively; n^3He_{res} and n^4He_{res} are the residual amounts of ^3He and ^4He, respectively, left behind at the end of the degassing step; and n^3He_{blank}, n^4He_{blank} are the amount of ^3He and ^4He, respectively, of the blank of the inlet system. n^3He_{tri} is the amount of tritiugenic ^3He produced during the storage time

$$n^3He_{tri} \text{ (in mol)} = T \times [1-\exp(-\lambda\tau)] \quad (3)$$

where T (in mol) is the amount of tritium in the sample at the date of the bulb closure and λ is the decay constant for tritium (Lucas and Unterweger, 2000).

Using equation (2), equation (1) can be rewritten as follows:

$$n^3He_{meas} - n^3He_{std} - \alpha R_a (n^4He_{meas} - n^4He_{std}) = n^3He_{tri} \quad (4)$$

or

$$n^a_{std} [^3He] [(N3_{meas}/N3_{std} - 1) - \alpha(N4_{meas}/N4_{std} - 1)] = n^3He_{tri} \quad (5)$$

where n^a_{std} is the amount of dry air in the standard air aliquot, $[^3He]$ is the atmospheric mixing ratio of ^3He, $N3_{meas}$, $N4_{meas}$, $N3_{Std}$ and $N4_{Std}$ are the ^3He and ^4He peaks height corresponding to n^3He_{meas}, n^4He_{meas}, n^3He_{std} and n^4He_{std}. Note than in equation (5), the correction term $\alpha(N4_{meas}/N4_{std} - 1)$ is less than 0.1% of the main term $(N3_{meas}/N3_{std} - 1)$.

Finally, equations (3) and (5) give the atmospheric mixing ratio of ^3He as a function of the amount T of tritium in the sample at the date of the bulb closure, where T is given by the intercomparison exercise and the dilution factor :

$$[^3He] = T\times[1-\exp(-\lambda\tau)] / n^a_{std} / [(N3_{meas}/N3_{std} - 1) - \alpha(N4_{meas}/N4_{std} - 1)]$$

4. Results and discussion

175 The results of the ten measurements are displayed in Table 3 and Fig.2. The arithmetical
176 mean of the ten determinations of the air ^{3}He mixing ratio is [^{3}He] = 7.12 ppt. The individual
177 uncertainty on each determination (± 0.09 ppt) takes into account all existing uncertainties
178 including random uncertainties (Category A uncertainties in JCGM, 2008), i.e. counting
179 statistics and sample weight (±0.01%) as well as systematic uncertainties (Category B
180 uncertainties in JCGM, 2008) : uncertainty on the tritium value of the initial solution as
181 determined by the CCIR(II)-2009 intercomparison (±0.51%), on the dilution (±0.2%), on
182 meteorogical parameters (pressure, temperature, humidity) of the air standard (±0.25%), on
183 the volume of the standard air aliquot (±0.08%), on the value of α (±10%) and λ (±0.2%). The
184 propagation of uncertainties is calculated by combining all uncertainties using a Monte-Carlo
185 approach. The calculation shows that the systematic and random uncertainties are ± 0.05 ppt
186 and ± 0.07 ppt, respectively. This is fully consistent with the experimental standard deviation
187 of the ten measurements (±0.07 ppt – see Table 3), which only takes into account random
188 fluctuations. With ten determinations, the standard uncertainty of the mean is equal to
189 0.055 ppt, corresponding to a relative standard uncertainty of 0.8% on the determination of
190 the ^{3}He mixing ratio. Hence, our best estimate of the ^{3}He mixing ratio is [^{3}He]=7.12±0.06 ppt.
191 This is between 1% and 2% lower than the previous determinations reported in the literature,
192 7.27±0.20 ppt and 7.21±0.07 ppt (see section 1) but all values are statistically consistent
193 within their standard uncertainties (see Fig. 2). With the abundance of ^{4}He in the atmosphere
194 equal to 5.23±0.01 ppm (average of the four values in Table 1), this leads to an atmospheric
195 ^{3}He/^{4}He ratio of (1.36±0.01)×10^{-6} instead of (1.38±0.02)×10^{-6} (average of the five values in
196 Table 1). Note that the present-day ^{3}He/^{4}He value may have decrease slightly compared with
197 this value because of the increasing injection of radiogenic ^{4}He into the atmosphere by the
198 consumption of oil and natural gas. However, the magnitude of this decrease (between 0%
199 and 0.8% - see Lupton and Evans, 2004; Lupton and Evans, 2013; Brennwald et al., 2013;
200 Mabry et al., 2015) is still debated because of the difficulty to find well-preserved air archives
201 and to accurately measure such a tiny change.
202 A systematic 2% positive offset between the results based on the ^{3}He ingrowth method
203 and the Miami Tritium Laboratory beta-particle counting results (relative to the US National
204 Bureau of Standards, former name of the National Institute of Standards and Technology
205 tritiated water standard) has already been noted in the paper by Clarke et al. (1976). At the
206 time, the authors attributed this discrepancy to a possible underestimation of tritium half-life.
207 However, a later review of tritium half-life (Lucas and Unterweger, 2000) shows that this
208 explanation is unlikely. With the value of the atmospheric mixing ratio of ^{3}He determined in
209 the present study, this 2% discrepancy disappears, thus suggesting that it was indeed due to
210 an overestimation of the atmospheric mixing ratio of ^{3}He. Although the implications for ^{3}He or

^{3}H measurements are limited, we think that this new value of the ^{3}He mixing ratio in atmospheric air should be adopted by the tritium-helium community for consistency with tritium laboratories using beta-particle counting.

5. Summary and Conclusions

To re-determine the atmospheric mixing ratio of ^{3}He, we applied the ^{3}He ingrowth method in reverse, using a solution of tritiated water prepared for the international comparison of tritium activity measurements CCIR(II)-2009. This was done by preparing a tritium solution by two-step gravimetric dilution of a small aliquot of this primary solution. Compared to our routine protocol, we used special procedures both for sample preparation and mass spectrometric measurements to further increase the precision: for sample preparation, isotope fractionation due to water evaporation during degassing was suppressed by trapping the evaporated water at liquid nitrogen temperature and transferring it back into the sample at the end of the degassing step, thus avoiding any bias in the isotope fractionation correction. In the mass spectrometer, we added an aliquot of the air standard on top of each sample so as to keep the pressure in the source essentially identical for the standard and for the sample thus avoiding any additional uncertainty arising from the usual pressure correction. Our main conclusions are as follows:

- Our estimated value of the atmospheric mixing ratio of ^{3}He is [^{3}He]=7.12$\pm$0.06 ppt, corresponding to a relative standard uncertainty of 0.8%.
- This value is between 1% and 2% lower than the previous, less precise determinations reported in the literature. Nevertheless, all values are statistically consistent within their standard uncertainties.
- The adoption of this new value removes the systematic 2% positive offset noted by Clarke et al. (1976) between their results based on the ^{3}He ingrowth method and the Miami Tritium Laboratory beta-particle counting results.

Acknowledgments

Data supporting this study are available in Table 2 and Table 3 of the present manuscript.

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311

Figure captions

Fig. 1: Results of the CCIR(II)-2009 international intercomparison of activity measurements of tritiated water used in the present study. All error bars are 1-sigma standard uncertainties.

Fig. 2: Results of the ten mass spectrometric measurements of the atmospheric ^{3}He mixing ratio carried out in this study (black open circles). The two red open squares are the atmospheric ^{3}He mixing ratios reported in the literature by direct measurement (Davidson and Emerson, 1990) and by combining the determination of the atmospheric ^{4}He mixing ratio (Glückauf, 1944; Glückauf, 1946; Oliver et al., 1984; Holland and Emerson, 1987) with the $^{3}He/^{4}He$ ratio of air (Mamyrin et al., 1970; Clarke et al., 1976; Sano et al., 1988; Hoffman and Nier, 1993). All error bars are at 1-sigma.

Table captions

Table 1: Atmospheric ^{4}He mixing ratios and $^{3}He/^{4}He$ values from the literature.

Table 2 : Tritium concentrations measured by the participants of the CCIR(II)-2009 international intercomparison.

Table 3 : Results of the ten mass spectrometric measurements of the atmospheric ^{3}He mixing ratio carried out in this study.

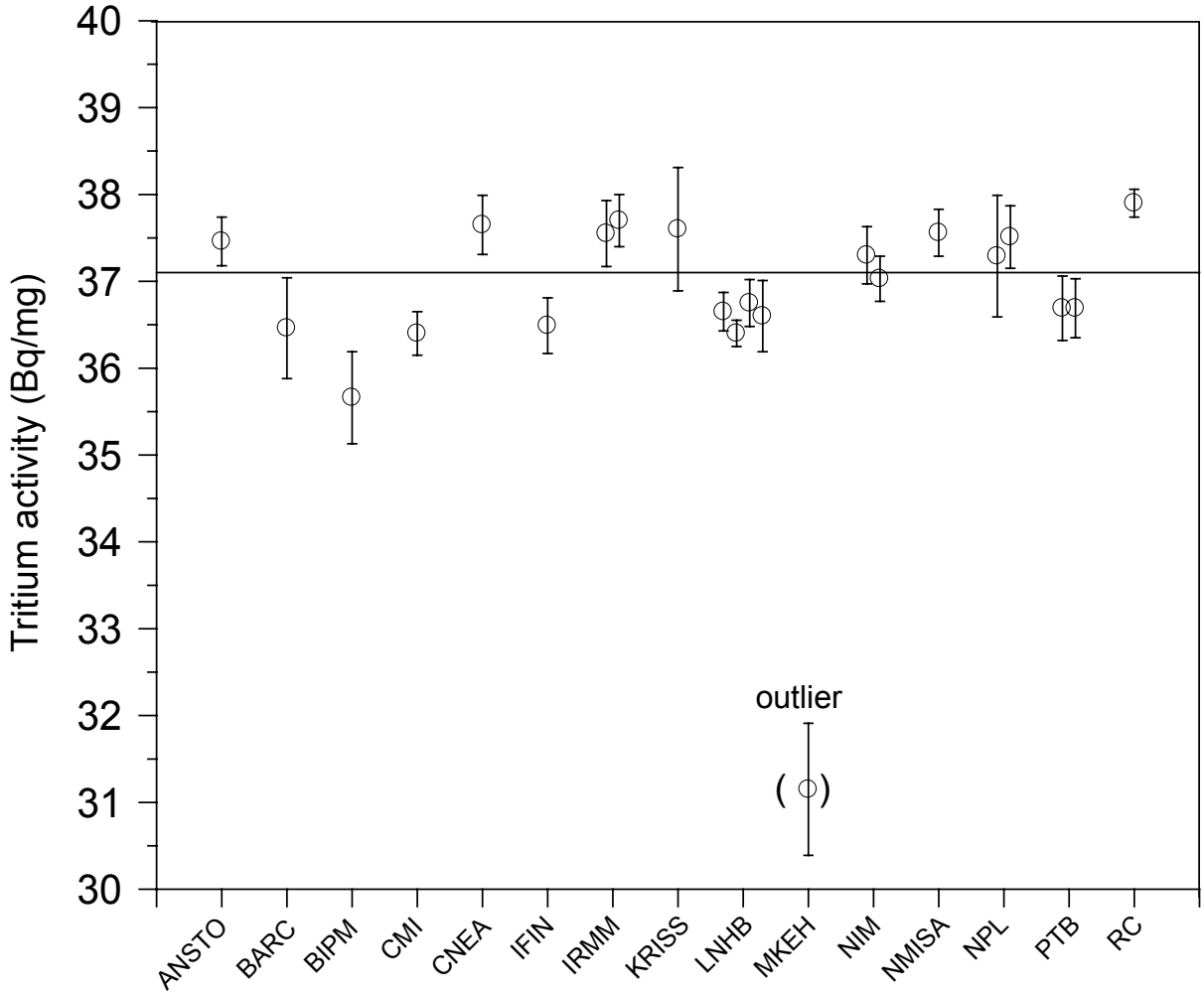

Tritium activity (Bq/mg)
40
39
38
37
36
35
34
33
32
31
30
outlier
( )
ANSTO
BARC
BIPM
CMI
CNEA
IFIN
IRMM
KRISS
LNHB
MKEH
NIM
NMISA
NPL
PTB
RC

Figure 1

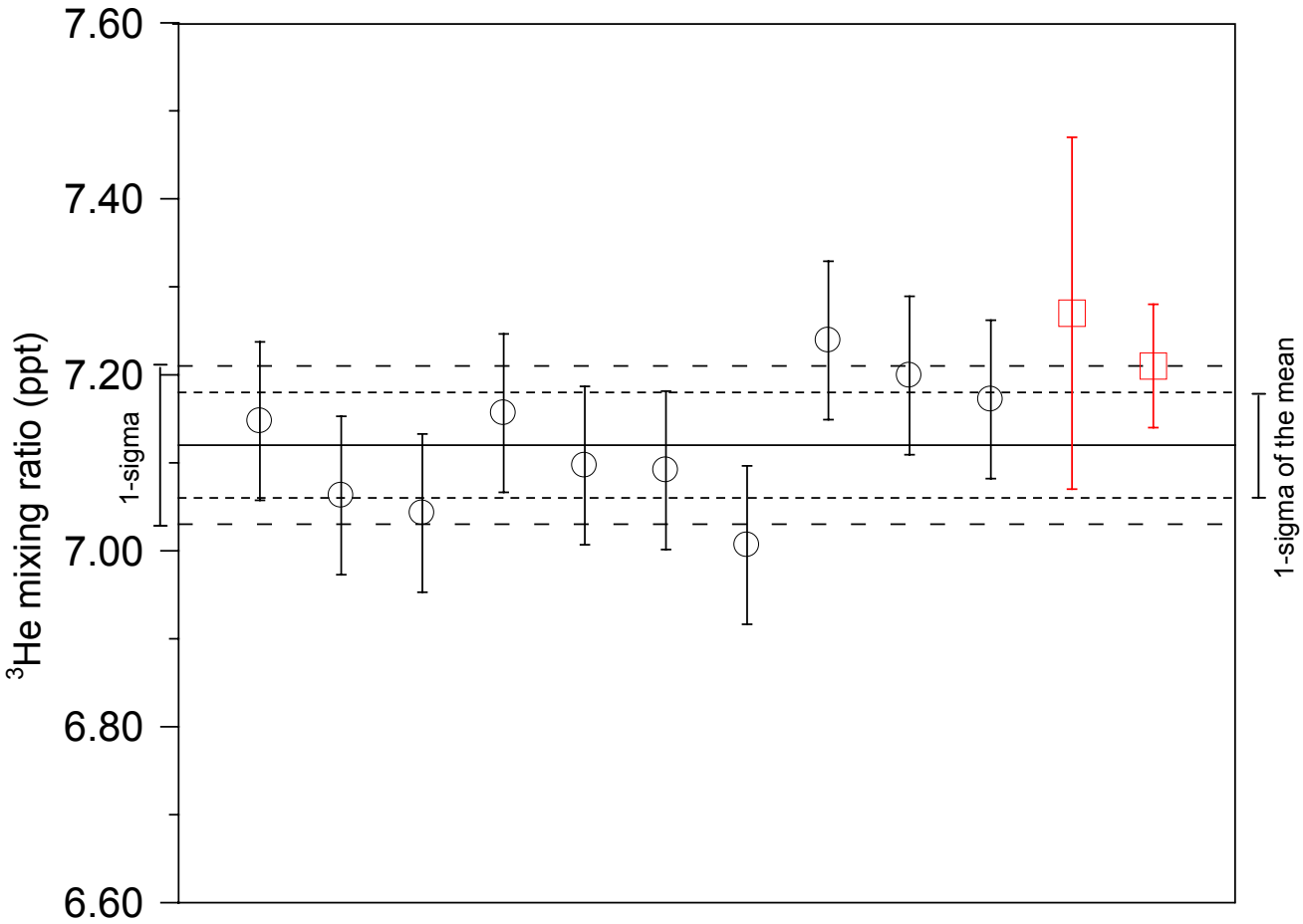

7.60
7.40
7.20
7.00
6.80
6.60
^{3}He mixing ratio (ppt)
1-sigma
1-sigma of the mean

Figure 2

References	$^3He/^4He$ ($x10^6$)
Mamyrin et al (1970)	1,399 ± 0,013
Clarke et al. (1976)	1,384 ± 0,006
Sano et al (1988)	1,343 ± 0,013
Davidson et al.(1988)	1,393 ± 0,014
Hoffman & Nier (1993)	1,371 ± 0,019
References	^4He (ppm)
Glückauf (1944)	5,24 ± 0,03
Glückauf (1946)	5,239 ± 0,004
Oliver et al (1984)	5,222 ± 0,002
Holland & Emerson (1987)	5,220 ± 0,004

Table 1

Organisation		Method	Tritium activity (Bq/mg)
ANSTO	Australian Nuclear Science and Technology Organisation, Lucas Heights, Australia	4p(LS)b TDCR	37,46 ± 0,28
BARC	Bhabha Atomic Research Centre, Trombay, Mumbai, India	4p(LS)b with 3H efficiency tracing	36,46 ± 0,58
BIPM	Bureau International des Poids et Mesures, Sèvres, France	4p(LS)b TDCR	35,66 ± 0,53
CMI	Czech Metrology Institute, Inspectorate for Ionizing Radiations, Prague	4p(LS)b TDCR	36,40 ± 0,25
CNEA	Comisión Nacional de Energía Atómica, Buenos Aires, Argentina	4p(LS)b TDCR	37,65 ± 0,34
IFIN	Institute of Research & Development for Physics and Nuclear Engineering, Bucharest	4p(LS)b TDCR	36,49 ± 0,32
IRMM	Institute for Reference Materials and Measurements, Geel, Belgium	4p(LS)b TDCR	37,55 ± 0,38
IRMM		4p(LS)b with 3H efficiency tracing	37,70 ± 0,30
KRISS	Korea Research Institute of Standards and Science, Yuseong, Daejon, Republic of Korea	4p(LS)b TDCR	37,60 ± 0,71
LNHB	Laboratoire National Henri Becquerel, CEA-Saclay, Gif-sur-Yvette, France	Differential proportional counting	36,65 ± 0,22
LNHB		4p(LS)b TDCR with CET	36,40 ± 0,15
LNHB		4p(LS)b TDCR	36,75 ± 0,27
LNHB		4p(LS)b TDCR with a digital MAC3	36,60 ± 0,41
MKEH	Magyar Kereskedelmi Engedélyezési Hivatal, Budapest, Hungary	4p(LS)b with 54Mn efficiency tracing	31,15 ± 0,76
NIM		4p(LS)b CIEMAT/NIST	37,30 ± 0,33
NIM	National Institute of Metrology, Beijing, Peoples Republic of China	4p(LS)b TDCR	37,03 ± 0,26
NMISA	National Metrology Institute of South Africa, Cape Town, South Africa	4p(LS)b TDCR	37,56 ± 0,27
NPL		Differential proportional counting	37,29 ± 0,70
NPL	National Physical Laboratory, Teddington, United Kingdom	4p(LS)b TDCR	37,51 ± 0,36
PTB		4p(LS)b CIEMAT/NIST	36,69 ± 0,37
PTB	Physikalisch-Technische Bundesanstalt, Braunschweig, Germany	4p(LS)b TDCR	36,69 ± 0,34
RC	Institute of Atomic Energy, Radioisotope Centre POLATOM, Otwock-Świerk, Poland	4p(LS)b TDCR	37,90 ± 0,16

Table 2

Sample N°	[^{3}He] ppt		
1	7,15	±	0,09
2	7,06	±	0,09
3	7,04	±	0,09
4	7,16	±	0,09
5	7,10	±	0,09
6	7,09	±	0,09
7	7,01	±	0,09
8	7,24	±	0,09
9	7,20	±	0,09
10	7,17	±	0,09
Mean	**7,12**		
1-Sigma	**0,07**		

Table 3