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Weighing uncertainties in quantitative source preparation for radionuclide metrology

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Abstract

In radionuclide metrology, the assessment of activity concentrations is directly related to the mass determination in quantitative source preparation. As far as primary measurements are concerned, the associated weighing uncertainty has a non-negligible contribution in the uncertainty budget. Several parameters influencing the mass determination have to be considered in source preparation and for the evaluation of the weighing uncertainty: technical specifications of calibrated balances, environmental quantities such as air buoyancy and other disturbance sources like electrostatic effects. The accuracy of mass determination is also conditioned by good practices in source preparation. This article reviews the different parameters in the evaluation of the weighing uncertainty. Its aim is also to give some guidance on how to achieve the smallest uncertainty.

Keywords: weighing uncertainty, quantitative source preparation, radionuclide metrology

1. Introduction

In radionuclide metrology, quantitative sources prepared for activity measurements are the result of a weighing procedure applied for the sampling of a master solution. In consequence the activity concentration, given in Bq per unit of mass, relies directly on physical quantities, methods and practices involved in the mass determination. The recent comparison, CCRI(II)-S7, of uncertainty budgets for $4\pi\beta\text{-}\gamma$ coincidence counting [1] reports the analysis of two dominant uncertainty components: efficiency-extrapolation and weighing. Using the same data set, weighing relative standard uncertainties reported by each participant mainly fall between 0.05% and 0.26%. It can be concluded from this comparison exercise that the uncertainty evaluation related to the mass determination depends on the experience of each laboratory.

This article addresses the different parameters influencing the whole source preparation procedure that have to be considered in the associated uncertainty evaluation [2]. It should be stressed that source preparation must be performed by a well-trained staff in order to achieve the smallest uncertainty. For that purpose, this article intends to give some guidance to achieve standard uncertainties below 0.1% [3] for source preparation, provided some strict rules are followed through the entire procedure. Volumetric sampling is simpler but far less accurate than gravimetric sampling, which is the only method capable to achieve the smallest uncertainty for primary measurements. Whether volumetric or gravimetric sampling methods should be used depends mainly upon the final uncertainty needed.

The first part of this article is dedicated to the weighing equipment and to good practices in their use. The second part focuses on specific weighing methods in radionuclide metrology, where mass determination in source preparation involves weighing by difference from small bottles or a pycnometer. Some guidance for source preparation using the pycnometer and for source dilutions are also given. The associated uncertainty budgets are detailed afterwards in the case of a 20 mg drop deposition and a 50-fold dilution. Finally, it is shown that the uncertainty related to source preparation can only be completely assessed from a statistical analysis of primary measurements based on a set of several sources.

Throughout the paper, all stated uncertainties are given with a coverage factor equal to one ($k = 1$).

2. Electronic balances

2.1. Working principles and buoyancy correction

A contemporary electronic analytical balance does not directly measure the mass m_s of a sample s ; rather, it measures its weight force. Therefore, the display shows the so-called weighing value, w_s . In most cases, these data are not identical if the sample is weighed in air because the resulting air buoyancy gives rise to a systematic deviation. Although electronic precision balances are calibrated with reference weights whose mass is known with low uncertainty, they are also subject to buoyancy. They are made from a steel alloy with a density of 8000 kg m^{-3} . This results in the fact that the mass of a sample that has the same density can be determined without the necessity of a correction because the influence of buoyancy onto the sample and the calibration weight cancel each other.

A digital balance is a non-automatic instrument in which a load cell converts the gravitational force on a mass into an electrical signal to show the weight digitally on the display panel. A load cell is typically an electronic device (transducer) used to convert a force into an electrical signal. A load cell generally consists of four strain gauges in a Wheatstone bridge configuration. The digital balance uses the analog signal from the load cell, converted directly to a pulse with modulated output signal. It represents the weight of an object on the weighing pan and it is read by a micro-controller. This converts the signal to a weight value (usually displayed on an LCD panel). The micro-controller is responsible for calibrating the instrument.

In an electromagnetic force compensation system, the loaded weight is compensated by an electromagnetically generated force [4]. A compensation coil through which a permanent current flows is inserted in a permanent magnetic field. In the unloaded condition, current regulation ensures that the system is in the zero position. With the aid of an optoelectronic position sensor, the coil position is controlled with an accuracy better than a thousandth of a millimeter. It records vertical positional changes when the balance is loaded. This information from the controller is used to generate a compensation current in the coil that returns the weighing system to zero. The current is directly proportional to the loaded weight; its value is digitalized and sent to the display.

The indication of the balance is proportional to the force exerted by a sample of mass m_s to the load receptor according to:

$$w_s \approx m_s \cdot g \cdot \left(1 - \frac{\rho_a}{\rho_s}\right) \quad (1)$$

with g the local gravity acceleration, ρ_a the density of the surrounding air and ρ_s the density of the sample. The terms in the brackets account for the reduction of the force due to the air buoyancy of the object. If the instrument is not displaced between calibration and use, there should be no variation of g . Moreover, if weighing is performed in the same air density as at the time of adjustment, then the indication of the instrument with the sample mass m_s on its load receptor equals:

$$m_s = w_s \times \frac{1 - (\rho_a/\rho_r)}{1 - (\rho_a/\rho_s)} = w_s \cdot Bu \quad (2)$$

where the density of the inner standard weight of the balance is $\rho_r = 8000 \text{ kg m}^{-3}$ and Bu is the buoyancy correction factor. It can be seen that a variation in air density has a far greater effect than a variation in the body density [5]. For example, a variation of 0.08 mg kg^{-1} in air density results in a variation of 10 mg kg^{-1} on the balance indication.

The most accurate formula to determine the density of moist air is the one recommended by the CIPM [6], but from section E3 of the OIML report [7] the following simplified formula can be used:

$$\rho_a = \frac{0.34848p - 0.009h \times \exp(0.061T)}{273.15 + T} \quad (3)$$

with air density expressed in kg m^{-3} , barometric pressure, p , in hPa, relative humidity of air, h , in % and air temperature, T , in $^{\circ}\text{C}$. The results from this formula differ by less than $2.4 \cdot 10^{-4}$ from the formula from the CIPM under the following environmental conditions:

$$\begin{aligned} 600 \text{ hPa} &\leq p \leq 1100 \text{ hPa} \\ 20 \% &\leq h \leq 80 \% \\ 15 \text{ }^{\circ}\text{C} &\leq T \leq 27 \text{ }^{\circ}\text{C} \end{aligned} \quad (4)$$

The density of air increases with increasing pressure, decreasing temperature and decreasing relative humidity. A finer approximation would also include the influence of carbon dioxide content.

In analytical laboratories as well as in everyday life, the majority of weighed objects have a lower density than steel and their weighing value is lower than their mass; therefore, the buoyancy correction is greater than one. According to equations (2) and (3), the buoyancy correction is more important at low densities. When liquids whose density is close to water are weighed, which is usually the case for radioactive solutions used in source preparation, the buoyancy correction is close to 1.001.

This approach requires the determination of the air density and, therefore, suitable equipment to measure at least the air temperature, the air pressure and the relative air humidity within the required uncertainties. Another correction method for air buoyancy using an artefact can be used. This method, presented in Wunderli *et al* [8], has the advantage that a mass artefact is used alone to correct for air buoyancy. The associated uncertainty on buoyancy correction is higher (between 3 and 4 times) but is still low ($5 \cdot 10^{-4}$). A recent paper from Malengo [9] deals with correlations in calibration and use of electronic balances. In particular, it points out the different corrections of the buoyancy.

2.2. Best conditions of use of a balance [5]

Micro and semi-micro balances are sensitive to several physical effects because they result in measurable weight changes which analytical balances cannot suppress. For example, slow evaporation, moisture uptake, or forces which act on the weighing pan and weighing sample (e.g. magnetism, electrostatics) are interpreted by the balance as weight changes.

2.2.1. Location.

The precision and reproducibility of weighing results are closely associated with the location of the balance [10, 11]. It should be chosen in order to avoid vibrations, air currents and temperature fluctuations. Semi-micro and microbalances should be placed on a weighing bench which should be stable to transfer as few vibrations as possible. It should be antimagnetic (no steel plate) and protect against electrostatic charges (no plastic or glass). The weighing bench should be fixed either to the floor or on the wall since mounting the bench on both places at once could transfer vibrations between wall and floor. The bench should be reserved for the balance; it is better to position the balance directly over the legs of the bench, where there are the fewest vibrations. The weighing bench should be placed in a corner of a room because these are generally the most vibration-free areas of a building.

Ideally, the room should be accessed through a sliding door to reduce the influence of door movements. The temperature of the room should be kept as constant as possible since weighing results are influenced by temperature (typically $1\text{--}2 \cdot 10^{-6} \text{ }^{\circ}\text{C}^{-1}$). The bench should not be placed near radiators or windows to avoid the heat from direct sunlight. This is also true for light bulbs; fluorescent tubes should be used instead. The balance should be placed away from the air flow of air conditioners or devices with ventilators. Recent balances are often equipped with an automatic system which can compensate the remaining temperature drift virtually. Ideally, the relative humidity should be between 45% and 60%. Balances should not be operated above or below the measuring range of 20% to 80%.

Constant monitoring is advisable with micro-balances. Of course, these controls must be in place at all times (including during use) for the calibration uncertainty statement to remain valid during daily use.

2.2.2. Acclimatization

Thermal equilibrium is also useful for the balance to ensure that all circuitry and hardware have reached a stable temperature. This should be ensured by keeping the balance in an energized state (i.e. turned on) for sufficient time, at least 30 min according to the manufacturer.

It is advisable to let all objects to be used during the weighing process acclimate in the same room as the balance in order to minimize the effects of convection. Indeed, an initial temperature difference, ΔT_0 may be reduced by acclimatization over a time, Δt ; this occurs faster for smaller objects than for larger ones. When a weight is put on the load receptor, the actual difference, ΔT , will produce an air flow around the weight, leading to parasitic forces which result in an apparent change, Δm_{conv} , on its mass [12]. The sign of Δm_{conv} is normally opposite the sign of ΔT . For enclosed weights of less than 1 kg, for example, an acclimatization time of 15 h is enough; it is advisable to let the devices acclimate overnight. The effect of convection is relevant only for weights of classes E1 and E2, which are to be used for micro- and analytical balance calibration [7].

2.2.3. Static electricity.

Weighing in dry air (with less than 40% relative humidity) can drastically distort the weighing result because materials, especially those with a high degree of electrical insulation such as glass or plastic, can become electrostatically charged. For this reason, placing the object to be weighed in a metallic container can help discharge it or an external (or internal) antistatic tool can be used (U-shaped or point electrode, ionizing blower or a radioactive static charge eliminator such as an ^{241}Am source). Nevertheless, the best solution would be to use a humidifying device as soon as a relative humidity falls below 40% to prevent this additional difficulty.

Static electricity will be mentioned again later in this article since it can have an important influence during weighing of plastic pycnometers or glassware due to their high degree of electrical insulation.

2.2.4. Magnetic interaction.

Magnetic interactions should also be avoided, especially on high resolution instruments. A way to check for an observable effect of magnetism is to place a non-metallic spacer below and above a standard weight to obtain two different indications. The difference of these two indications should not be different from zero. According to NISTIR 6919 [13], magnetic effects may also be detected by making a series of mass measurements of the weight, half of the measurements with the weight in an upright position and half of the measurements with the weight inverted. Magnetism is probable if a detectable difference, greater than the repeatability of the balance, is noted between the average of the upright indications and that of the inverted indications. Practically all objects made of iron (steel) are highly permeable to magnetic forces (ferromagnetic). Since the magnetic force decreases with increasing distance, the sample can be distanced further from the weighing pan by using a non-magnetic support (e.g. beaker or aluminum stands). More information can be found in Sutton [14].

2.2.5. Good practice in handling standard weights

To perform accurate weighing with a microbalance, one should avoid dust, fingerprints and electric charges on the pycnometer and standard weights [3]. During source preparation or balance calibration, only standard weights of Class E2 [7] are used at the LNHB (Laboratoire national Henri Becquerel). Following the recommendations of the international organization of legal metrology (OIML) [7], they are made of austenitic stainless steel. These weights should be routinely dusted before use using a

clean soft haired brush; more intense cleaning should be avoided since it would affect their calibration history. Weights must always be handled with the greatest care. In particular, they should never be touched with bare hands nor breathed upon. If it is necessary to handle a weight by hand, gloves should be worn (chamois leather is ideal). Otherwise, tweezers or forceps should be used to manipulate them inside the balance (gloved hands must always be kept outside the balance to not affect its thermal stability).

2.3. Uncertainty budget associated with the use of a balance

As stated before, it is important to keep in mind that the weighing operation during source preparation is not the only uncertainty source. Therefore, the following items combine with other uncertainty components in relation to the other steps of source preparation and, for instance, the specifications of the pycnometer method (described later). These additional uncertainties are referred to as 'method uncertainty' and will be addressed in the third section of this document.

Several calibration regulations, guidelines and papers can be found in the literature in the field of mass metrology [5, 10, 13, 15–18]. Among them, the EURAMET Guide [5] seems to be the most widely adopted, especially by accreditation bodies; the DAkkS in Germany has adopted the EURAMET Guide since 2008 [19] and the guide from the COFRAC in France [18] is very similar to it. The main differences among these guides come from the modelling of the corrections due to the buoyancy effects, both during the calibration and the use of the balance [9].

The main factors associated with the weighing device to be taken into account in the uncertainty budget are: the repeatability of indications, the errors of indications, and the effects of eccentric application of a load. These elements should appear in verification certificates of the instrument. Otherwise they can be evaluated by following the recommendations given in the literature [5, 19]. One must be aware that most manufacturer specifications are based on 'idealized' weighing conditions; it is thus very important to check the available data under the laboratory conditions of use to build a realistic uncertainty budget. The uncertainty evaluation for the mass value of a sample according to ISO-GUM [20] is outlined in figure 1.

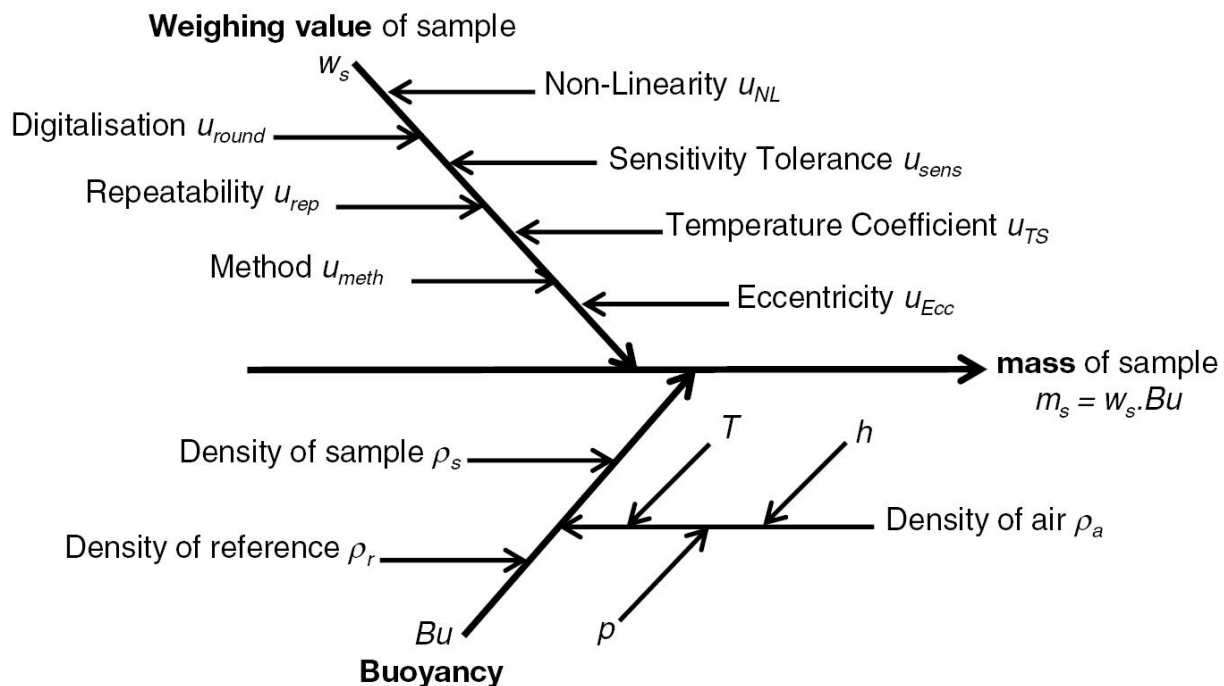


Figure 1. Cause-and-effect diagram of the main uncertainty parameters of weighing on an electronic analytical balance.

2.3.1. Reading uncertainties

Reading uncertainties should account for the rounding error of no-load indication and at load. In both cases, a rectangular indication is assumed; therefore, for a resolution or scale interval d_0 :

$$u_{round} = d_0 / (2\sqrt{3}) \quad (5)$$

On some instruments approved in accordance with OIML R76 [21], the rounding error of a zero indication is limited to $\pm d_0/4$. This uncertainty is often negligible compared to the following repeatability; however, when the balance has a limited resolution, the repeatability could be very good because of the reading uncertainty, which should not be neglected.

2.3.2. Repeatability

Varying results are not uncommon if the same object is weighed under repeatability conditions. The reasons for this behavior are of a physical nature: small air drafts present even within the draft shield, temperature non-equilibria, and electronic noise for example. From a practical point of view, the resolution is included in the experimental repeatability. Expressed as standard deviations, s , (which are identical with standard uncertainties u_{rep} in this case), the following data are typical for semi-micro or micro-balance respectively if operated carefully: 40 μg (for $m > 50$ g) and 0.9 μg (for $m > 2$ g). These data are mentioned in balance specification sheets.

If the operator works less carefully or if the environmental conditions in the laboratory are unsatisfactory, the repeatability can be worse, i.e. the value of standard deviation for repeatability will be greater and should be determined experimentally [22]. This is also true for critical weighing objects such as volatile or hygroscopic samples. Specific measurements should be performed to check if the usual repeatability criterion is met.

2.3.3. Non-linearity.

An ideal balance exhibits a perfectly linear relationship between displayed value and the load on the platform. In reality, this characteristic curve is not straight but curved in a certain shape which is not determined for each individual instrument (although this could be done in principle).

Instead of individual data, manufacturers guarantee maximum deviations from linearity which will not be exceeded. For example, for a semi-microbalance (weighing capacity up to 200 g), the limit for within 10 g is $NL_{max} = 0.03$ mg (this is also the value to be used if a sample ≤ 10 g is weighed into a tare vessel of e.g. 150 g) and for within 200 g, $NL_{max} = 0.12$ mg.

These numbers refer to the net mass and need to be considered twice for each weighing operation because the deviation from ideality may occur with both the determination of the tare and of the gross weight. The data must be described as rectangular distributions. Therefore, the non-linearity contribution of a weighing operation is:

$$u_{NL} = \frac{NL_{max}}{\sqrt{3}} \quad (6)$$

One should note that the technical data given by the manufacturer are worst-case scenarios, but they are recommended for everyday use. Non-linearity is a more complex feature of a balance as more thoroughly discussed elsewhere [23].

2.3.4. Sensitivity tolerance.

The slope of the characteristic curve (displayed mass as a function of applied mass), i.e. the sensitivity of the balance, has some tolerance or uncertainty. This deviation is expressed as a relative value (ST_{max}) and is a function of the net mass. For example, if the balance specifications mention $ST_{max} = 2 \cdot 10^{-6}$ then, if a sample of 1 g is weighed in, the sensitivity tolerance is 2 μg . The sensitivity tolerance is described by a rectangular distribution proportional to the net weighing value:

$$u_{sens} = w_{net} \cdot \frac{ST_{max}}{\sqrt{3}} \quad (7)$$

This uncertainty parameter includes the uncertainty of the built-in reference weight as well as the uncertainty of the process by which the balance adjusts its sensitivity (the slope) with the help of this reference weight. This uncertainty term is also known as u_{CAL} [8]; the mathematical treatment of u_{sens} and u_{CAL} is identical. According to Reichmuth *et al* [24], the difference is more than a linguistic one and the tendency is to avoid the use of the term 'calibration' because first, it does not cover all influences, and second, it means the comparison with a reference without any setting. However, what can be done in the laboratory is the adjustment of the balance sensitivity, i.e. the determination of the sensitivity, including its proper setting if necessary.

2.3.5. Temperature sensitivity.

Environmental conditions also have an influence on the result especially if the instrument is not adjusted immediately before its use. The slope of the characteristic curve is temperature dependent with a maximum stable deviation (meaning that the balance is in the temperature equilibrium with its surroundings). Thus, to account for a change in ambient temperature around the device, a manufacturer's specification is given (TS_{max}) expressed in 10^{-6}K^{-1} (or in $\text{ppm}^\circ\text{C}^{-1}$). This value is proportional to the net weighing value and is described by a rectangular distribution, so is the total temperature variation over the measurement and the uncertainty contribution of the temperature sensitivity is [8]:

$$u_{TS} = w_{net} \cdot \frac{|TS_{max}|}{\sqrt{3}} \cdot \frac{|\Delta T|}{\sqrt{3}} \quad (8)$$

2.3.6. Eccentricity.

The eccentricity uncertainty accounts for the error due to an off-centre position of the centre of gravity of a load or if more than one body is weighed. The effect found during calibration (see certificate) should be considered in full by:

$$\Delta m_{ECC} = (\Delta w_{ECC})_{max} / (L_{ECC} \sqrt{3}) \quad (9)$$

where $(\Delta w_{ECC})_{max}$ is the maximum deviation measured during the eccentricity test and L_{ECC} the load used. Usually L_{ecc} is at least one third of the maximum capacity of the instrument and is moved to 5 positions around the centre of gravity of the load receptor (including its centre). The zero indication should be checked after each removal of the test load. The deviation from the weighing value to the one found in the center can be as high as 0.1 mg depending on the type of semi-microbalance. However, as it can be easily avoided by the careful placement of the object centrally onto the pan, the eccentric load will not be considered in the uncertainty budgets presented in section 4 of this paper.

2.3.7. Buoyancy correction.

Following the assumptions from equation (2), the buoyancy correction is also equal to:

$$Bu = \frac{\rho_s(\rho_r - \rho_a)}{\rho_r(\rho_s - \rho_a)} \quad (10)$$

We assume that the air density and its uncertainty are identical for the calibration and the weighing derivatives as a function of ρ_s , ρ_r and ρ_a , then the uncertainty on buoyancy correction becomes [24]:

$$u(Bu) = \frac{\rho_s}{\rho_r(\rho_s - \rho_a)} \cdot \sqrt{\left(\frac{\rho_a(\rho_a - \rho_r)}{\rho_s(\rho_s - \rho_a)}\right)^2 u^2(\rho_s) + \left(\frac{\rho_a}{\rho_r}\right)^2 u^2(\rho_r) + \left(\frac{\rho_r - \rho_s}{\rho_s - \rho_a}\right)^2 u^2(\rho_a)} \quad (11)$$

For the uncertainty of the air density, it is convenient to calculate the extreme values which may occur in the laboratory and to treat these data as the boundaries of a rectangular distribution. The extremes are the combination of high atmospheric pressure, low temperature and low humidity on the one hand, and low pressure, high temperature and high humidity on the other. This evaluation was chosen because p , h and T are physically linked. It leads to a conservative value of the uncertainty on the air density compared to an application of the law of propagation of uncertainties to equation (3).

3. Performance of quantitative source preparation

3.1. Weighing techniques for quantitative source preparation

Since a wide range of processes relies on weighing, many weighing techniques can be found depending on the uncertainty requirements. The simplest method is to place a test piece on the balance and take the displayed reading as its weight. This type of measurement is suitable only for low accuracy applications. As far as source preparation is concerned, it is advisable to use a 'weighing by difference' technique or, even better, a 'substitution weighing' method for high accuracy. For example, weighing by difference involves placing a container on the balance pan, noting the reading, and then adding a substance to it. The final reading is noted and the difference between the two readings is taken to be the amount of material in the container. The 'substitution weighing' method will be described in section 3.1.2.

Radioactive sources dedicated to primary measurements have masses usually ranging from 10 mg to 30 mg. The weighing of a 20 mg drop will be discussed as an example in section 4.

3.1.1. Pycnometer method.

For this method, the mass of a vessel containing the solution is determined with a microbalance before and after dispensing an aliquot [25, 26]. Pycnometers used for this purpose are generally plastic ampoules of 5 mL made of polyethylene.

By gently heating the tip and rotating the ampoule, a capillary of about 6 cm to 8 cm length is drawn. The upper part of the capillary is cut off before the filling operation and the ampoule is filled by suction, the outside of the capillary is wiped clean, and then the pressure on the ampoule is released slowly to prevent the formation of little droplets inside the capillary and that a thin film in the capillary is formed. Before using the filled ampoule, an additional 1 cm of the capillary is cut away to assure that any dried activity on the tip will not be dissolved again [27].

3.1.2. Substitution weighing.

After weighing the pycnometer, a set of calibration weights are placed on the balance with a total mass as close as possible to the mass of the full pycnometer. This method, was the most accurate and used extensively in the 1970s to not rely to a large extent on the optical scale of balances of former generations, which were used as transfer instruments only [28]. However, with electromagnetic force compensation balances, this method has two major drawbacks: the long weighing time increases

uncertainty on losses through evaporation during preparation, and problems can arise concerning the reliability of the reference weights which are in permanent use.

3.1.3. Elimination weighing [28].

The set of calibration weights is used at the beginning of the manipulation to check the performance of the balance. Weighing by elimination is a simplification of substitution weighing. To save time and to limit the number of standard weights handled, this method consists of 3 weighing steps per source instead of 4 for the substitution technique. The pycnometer is weighed before and after dispensing the drop of solution, but knowing (by difference) the mass of the drop, a similar calibration weight is added on the balance load receptor. Thus, to derive the mass of the dispensed drop, only the first and third weighings, which are very similar, are used as well as the conventional standard weight mass (with known uncertainty). This method is detailed afterwards in the procedure for dispensing micro-drops of master solution.

3.2. Good practices for quantitative source preparation

The basic requirement when preparing and handling radioactive solutions to be characterized is to guarantee their radionuclide content, expressed in activity concentration. This content should not vary during the processes of source preparation, from dilutions to drying in the case of solid sources for example.

3.2.1. Small quantities and carrier solution.

The dependence of activity and mass can be derived from the radioactive decay law; at constant activity, the mass of a radionuclide is directly proportional to the half-life of the nuclide. For example, 1 MBq of ^{60}Co corresponds to a mass of 24 ng only. In 1 mL, this represents a molar concentration of $4 \cdot 10^{-8} \text{ mol L}^{-1}$. When one handles very small amounts of matter, there is a risk that there will be adsorption onto surfaces of the vessels. On glass surfaces, ion exchange adsorption takes place on the surface silanol groups ($-\text{Si}-\text{OH}$, [29]). This silanol group is a weak acid and its hydrogen dissociates in neutral and alkaline solutions and can bind ions. Glass surfaces typically have $10^{-10} \text{ mol cm}^{-2}$ ion exchange capacity. Thus, for example, in a 10 mL glass vial, there is about 10^{-9} mol of ion exchange capacity [30]. Placing 10 mL of the aforementioned 1 MBq $\text{g}^{-1} \text{ }^{60}\text{Co}$ solution (assuming it has a neutral pH) in such a vial could result in the adsorption of the entire radionuclide content. Adsorption also takes place on plastic surfaces, but not with ion exchange as with glass. On plastic surfaces, atoms appear with free electron pairs, such as nitrogen and oxygen, which bind metals with coordination bonds [30]. In either case, adsorption is usually a problem and must be considered.

To ensure their thermodynamic stability (i.e. avoid adsorption, hydrolysis, precipitation, colloids formation, etc), the composition of standard solutions is optimized [29]; the solvent is chosen to ensure the solubility of the chemical element and a carrier, isotopic or not, is added to control the chemical behavior of the radionuclide (there can be more than one carrier). The chemistry of stable and radioactive nuclides is the same (except at high radioactivity level where radiation may start to affect the solvent). The carrier should have the same chemical form (oxidation state especially) as the radionuclide in solution. The carrier element is present in excess compared to the radioactive element, usually between 250 and 10^5 or even more for short-lived radionuclides. The concentration of the carrier element stands usually, from the experience at the LNHB, between 10 to 100 μg of element per gram of solution, which corresponds to concentrations between $(5 \cdot 10^{-5} \text{ to } 10^{-3}) \text{ mol L}^{-1}$. It is important to note that dilutions are performed with the same solvent composition (including carrier concentration) as the master solution. Among other sources of loss of radionuclides, hydrolysis can be major. Hydrolysis is the tendency to form hydroxide with increasing pH, which results in the formation of uncharged species that are insoluble and precipitate. The presence of insoluble species must be avoided since radionuclides could adsorb on their surfaces. Fortunately, working at very acidic pH very often prevents any hydroxide formation.

As a general guide, to avoid hydrolysis, 0.1 mol L^{-1} acids are adequate although concentrations above and below this may be used successfully for specific radionuclides; it is also sometimes necessary to add complexing molecules. A table published in the handbook of radioactivity measurements procedures [31] for the preparation of standard sources section gives chemical data for various radionuclides and may serve as a guide. It should also be mentioned that, for measurement purposes, it can be important to limit the carrier quantity in solution in order to limit radiation absorption for dry sources.

Depending on the chemistry of the radionuclide, it may be necessary to adapt source preparation protocols. For example, to avoid any loss of volatile iodine during the preparation of point sources, inactive iodine (as sodium iodide) is added as carrier, but so too is a reducing agent (such as sodium thiosulfate) to prevent the formation of volatile I_2 and, eventually, silver nitrate during point source preparation to precipitate iodine (as AgI) and avoid any loss during the drying process.

It must be pointed out that, for long term storage, radioactive solutions should be stored in flame sealed glass ampoules rather than in plastic containers [32].

3.2.2. Cleaning and carrier treatment.

The basic material for preparing the solubilization medium is pure water with a constant resistivity of $18.2 \text{ M}\Omega \text{ cm}$, which must remain pure with time (impurities, CO_2 , O_2 , organic carbon content [29]). Pure acids may also be used. One must be aware that natural radionuclides, such as uranium, can be present up to $10^{-9} \text{ mol L}^{-1}$ in usual concentrated acids. Carrier solutions are prepared from commercially available pure salts. At the LNHB, we chose to use the salts with the highest hydration number so that products remain stable over time. Otherwise, hygroscopic salts can uptake water over time of use (even if stored in a desiccator), and their molar mass changes resulting in a systematic error of at least 18 g mol^{-1} . Whenever possible, for solubility reasons, concentrated carrier solutions are prepared at the LNHB; they are usually 100 times more concentrated than the carrier medium to be used and are stored at 4°C .

In preparing solutions and dilutions, care should be taken to assure cleanliness (including chemically) of all vessels used with both the radioactive and inactive solutions. At the LNHB, the first cleaning step consists in machine washing with 4.8% sodium hydroxide solution and 20% acetic acid solution. The final rinse is performed using deionized water. In order to saturate the surfaces of all the vessels (glassware, plastic pycnometers, etc), a carrier pre-treatment is systematically performed. At the LNHB, this consists of filling all laboratory ware to be used with carrier solution for at least 24 h. Indeed, it has been shown by Iroulart [29] that sorption phenomena occur quite rapidly, from 5 h to 15 h. Then, the vessels are emptied and dried in an oven at about 60°C and then cooled. They are eventually stored in a location free from dust.

3.2.3. Balance calibration.

For the first step, the levelness of the balance should be checked and corrected if necessary. Then, the instrument should be energized prior to use (and calibration) for as long as the warm-up time specified by the manufacturer (at least 30 min; manufactures often suggest longer durations up to 12 h for microbalances). The balance used for source preparation must be calibrated, which should be performed where the instrument is being used. Modern balances have a built-in and self-activated calibration routine. Others must be calibrated by laboratory personnel. The instrument should be conditioned by loading approximately up to the largest test load repeatedly at least ten times before starting measurements.

It is advisable to check the balance prior to its use. For source preparation, at least two mass points need to be checked. At the LNHB, an 8-point check is performed between 5 mg and 5 g. As the weighing procedure relies on these standard weights, they should be checked regularly (at the LNHB

standard weights are checked every 3 years, and balances are checked and adjusted by the manufacturer every 3 years as well; these two control cycles are offset from each other by 1.5 years). A calibration history of each standard weight should be kept in order to help define a suitable periodicity.

In view of buoyancy effects, the air temperature in reasonable vicinity to the instrument should be measured at least once during the measurements. In a controlled environment, the span of the temperature variation should be noted. Since weighing uncertainties below 0.1% are needed for source preparation, pressure and relative humidity of air should also be measured during the weighing.

3.2.4. Procedure for dispensing micro-drops of master solution using the weighing by elimination method.

It is essential to allow the pycnometer containing the radionuclide solution to attain thermal equilibrium with the balance but the operator should also be present in the room at least 15 min before the weighings start (in front of the balance preferably [33]) The balance performance check described in the previous section should be done. The first step is to carefully draw at least about 1 g of the solution into the pycnometer without bubbling and then to purge the capillary stem by dispensing a few drops of solution to a tissue to wipe the outside of the pycnometer stem as well. It should be recalled that pycnometers must always be handled with tweezers, preferably long ones or other equivalent remote handling equipment. For all following steps, it is crucial to dispense the solution slowly so as to leave the stem free of liquid droplets avoiding that any liquid is left in the capillary of the pycnometer since it could dry very quickly and impact the results.

The pycnometer is weighed with its full content (w_1). A suitable amount of master solution is delivered to a source mount with care (the micro-drop should not fall on the source mount but the end of the pycnometer should not be plunged into the deposited drop either). In the pycnometer method, care should be taken to avoid any drops outside the capillary of the pycnometer. They would evaporate and distort the results. After drop deposition, the pycnometer is weighed again (w_2). Depending on the mass of solution dispensed, one or several suitable standard weight(s) is/are carefully added on the balance load receptor, m_{std} . The mass recorded, w_3 , is then close to that of the previous 'full' pycnometer, w_1 , thus limiting linearity problems. The mass of the drop dispensed, w_s , is given by:

$$w_s = w_1 - w_3 + m_{std} \quad (12)$$

At the LNHB, the difference between the third and the second weight registered is used to check that it corresponds to the conventional mass(es) of the standard weight(s) added [28]. The two values should be in agreement within twice the uncertainty value on the standard weight(s) used. If not, a warning is written on the weighing sheet to stress that this measurement is questionable. There can be several causes such as evaporation in the pycnometer or drying of a drop left in the capillary stem during the weighing process, deviation of the zero of the balance, etc.

In the case of the calibration of radioactive solutions, the sources are prepared in series, which is a major source of correlations. To limit those during the preparation of a series of sources, it is advisable to record the weight of the full pycnometer before each drop deposition, even if, in theory, its mass should be the same as the 'empty' pycnometer mass from the previously deposited drop.

3.2.5. Procedure for accurate dilution of a master solution adapted from Merritt [34] and Campion [32].

Diluting radioactive solutions is a common step in source preparation in order to adapt the activity of the source to the measurement device. To perform a dilution with the lowest uncertainty, it is advisable to use two balances: one for the radioactive solution amount, usually with a maximum capacity of a few grams only and another for the mass of diluent (with a maximum capacity of a few

hundreds of grams). Again, the pycnometer, the dilution flasks and everything else needed for the process should have reached thermal equilibrium. The weighing procedure previously described for the pycnometer does not change. The addition of the diluent is performed in several steps. Indeed, the dilution flask is first weighed empty, then filled with half of the diluent needed. A suitable amount of radioactive solution is dispensed from the pycnometer (the capillary should be inserted into the flask to limit the crashing of the master solution). The dilution flask should be gently swirled to reach complete mixing; in order to achieve it effectively, the flask should not be filled to more than half its nominal capacity. The remaining quantity of the diluent is then added to the dilution flask, which is weighed. The dilution flask is closed and finally mixed by gentle swirling during at least 15 min. In practice, the weight of the dilution flask after the addition of master solution is also recorded to check the results.

If the density of the diluent is the same as that of the radionuclide, then the buoyancy corrections are cancelled in the dilution factor ratio.

In general, dilution factors should not exceed 1000, even if two suitable balances are used [3, 32]. If a larger factor is needed, it may be achieved in several stages, since the greater the dilution, the more difficult it becomes to achieve complete homogenization. If serial dilutions are needed, it is of course imperative that a different pycnometer be used for the transfer of the diluted solution from that used for the strong solution. In order to check dilution factors, it is advisable to prepare sources at each activity level to be measured by a measurement technique with a suitable measurement range (and low enough associated uncertainties).

3.2.6. Uncertainties from the weighing methods.

Possible sources of uncertainty during source preparation using a pycnometer are evaporation from the pycnometer (usually mainly evaporation of liquid on the outside of the vessel), difference in buoyancy before and after dispensing the drop or, in the worst case, the splashing of the drop when it reaches the source mount.

According to Van der Eijk and Moret [26], evaporation from a pycnometer as a function of time is about $0.5 \mu\text{g min}^{-1}$, a value which was also chosen in 2007 by Sibbens and Altitzoglou [3]. Merritt [34] considers a lower value ($20 \mu\text{g h}^{-1}$). Care must be taken to avoid liquid on the outside of the pycnometer since this evaporates at an increased rate and falsifies mass determination. In Le Gallic [28], this phenomenon was taken into account as an uncertainty of $1 \mu\text{g}$. At the LNHB, these effects are taken into account as a method uncertainty which is estimated to be $3 \mu\text{g}$ ($1.5 \mu\text{g}$ for w_1 and for w_3 , see section 4.2.).

The rate of evaporation from an open flask (dilution case) is higher than that of a pycnometer but the flask is kept stoppered as often as possible. A common evaporation rate cited by Merritt [34] or Campion [32] is 1 mg h^{-1} . At the LNHB, we consider that during the whole dilution procedure, the flask remains open for a total of no more than 1 min. The chosen absolute uncertainty value to account for the method is thus $30 \mu\text{g}$ ($15 \mu\text{g}$ per weighing, see section 4.3.).

3.2.7. Additional remarks.

If the weight display is unstable, it is advisable to check possible causes for static electricity (low humidity, use of accessories made of plastic or glass). Section 2.2.3 provides useful solutions to limit the phenomenon.

In order to lower electrostatic effects due to the radioactive solution (charge creation and accumulation), such as during source preparation with high-activity β -emitters, the control unit of the balance (containing most of the electronics) can be taken away from the weighing cell. One should also pay attention to the ground links of the different parts of the balance. Moreover, in order to lower

the accumulation of charges on the weighing pan and on the plastic pycnometer, an antistatic device can be used before each drop deposition.

Since source preparation is a sampling process where weighing is not always the last step, it is advisable to measure several sources from the same master solution in order to check for possible biases; Rytz suggests preparing at least 7 sources [35].

In order to deal with correlations, a trend study can be done by comparing the activity concentration derived from the measurement as a function of the order of preparation of the sources. Outliers should be identified. An example of a sample-to-sample trend dependent on the order of source preparation is shown in Fitzgerald *et al* [36] in the case of the standardization of ^{124}Sb . This is because no chemical reagent was added to prevent the loss of activity due to the volatility of ^{124}Sb . In order to identify potential systematic errors, a set of sources with variable masses can be prepared.

4. Uncertainty budgets for source preparation

4.1. Combined uncertainty

Any weighing operation is basically the difference between two indications: under load (with the sample) and at no-load. Each weighing is usually performed with a tare equal to zero, nevertheless, some of the uncertainty contributions described in section 2 still apply (rounding and non-linearity, [5]).

Following the cause-and-effect diagram presented in figure 1 and combining each contribution according to the law of propagation of uncertainties [20] the corresponding uncertainty expression for a balance reading w_i is given by:

$$u_{w_i}^2 = 2u_{round}^2 + u_{rep}^2 + 2u_{NL}^2 + u_{sens}^2 + u_{TS}^2 + u_{meth}^2 \quad (13)$$

Applying the law of propagation of uncertainties to equations (2) and (12), where w_s corresponds to the drop dispensed, allows to derive the following relative and absolute uncertainties on m_s :

$$\left(\frac{u_{m_s}}{m_s}\right)^2 = \frac{4u_{round}^2 + 2u_{rep}^2 + 4u_{NL}^2 + 2u_{sens}^2 + 2u_{TS}^2 + 2u_{meth}^2 + u_{Std}^2}{(w_s)^2} + \frac{u_{Bu}^2}{Bu^2}$$

$$u_{m_r} = \sqrt{Bu^2 \times (4u_{round}^2 + 2u_{rep}^2 + 4u_{NL}^2 + 2u_{meth}^2 + u_{Std}^2) + m_s^2 \times \left(\frac{2ST_{max}^2}{3} + \frac{\Delta T^2 TS_{max}^2}{9} + \frac{u_{Bu}^2}{Bu^2}\right)} \quad (14)$$

Where u_{Std} is the standard uncertainty of the standard weight(s) used.

Two common examples of source preparation operation are given in the next sub-sections: the preparation of a 20 mg source and a dilution factor calculation. The environmental conditions used in these examples are: temperature variation within $21.5\text{ °C} \leq T \leq 22.5\text{ °C}$, pressure variation is $995\text{ hPa} \leq p \leq 1005\text{ hPa}$, relative humidity variation within $40\% \leq h \leq 60\%$, and loads are centred carefully.

4.2. Example of uncertainty budget for a 20 mg drop deposition

In this example, the procedure for dispensing micro-drops of master solution using the weighing by elimination method described in section 3.2.4 is followed. The pycnometer is weighed on a microbalance of 5.1 g capacity with scale interval 1 μg . The pycnometer weighs about 2 g.

According to equation (12), 2 weighing operations are needed to know the mass of a 20 mg drop. The balance characteristics are shown in table 1 (measured repeatability for $m > 2$ g is 4 μg ; sensitivity tolerance is $1.5 \cdot 10^{-6}$ and temperature sensitivity is $10^{-6} \text{ }^\circ\text{C}^{-1}$). Following equations (10) and (11), the buoyancy correction factor is calculated with the following parameters: air density with its standard uncertainty taken as $1.181 (5) \text{ kg m}^{-3}$ (using the aforementioned environmental conditions); the density of calibration weights is $8000 (15) \text{ kg m}^{-3}$ and that of the weighed solution (carrier solution in 0.1 M HCl) is $1000 (3) \text{ kg m}^{-3}$ then the buoyancy correction is equal to 1.001034 (5).

Parameter	Value	u (mg)
Readability	1 μg	0.0003
Repeatability	4 μg	0.0040
Sensitivity tolerance	$1.5 \cdot 10^{-6} w^a$	0.0017
Temperature sensitivity	$1.0 \cdot 10^{-6} w^a$	0.0007
Method	3 μg	0.0015
Buoyancy correction (relative uncertainty)	1.001034	0.00053%
Standard weight	20 mg E2	0.0015
Combined standard uncertainty		0.0063
Combined relative standard uncertainty		0.031 %

^aW stands for the mass of the pycnometer and is around 2g.

Table 1. Example of uncertainty budget for a 20 mg drop deposition using the weighing by elimination method.

In table 1, the combined absolute and relative standard uncertainties are calculated using equation (14). Among the uncertainty components from equation (14), the non-linearity component does not need to be taken into account since w_1 and w_3 are set to be very close. The main uncertainty components are, as expected, the repeatability and the method. Using E2 class standard weight for the elimination method enables to keep the uncertainty budget low. They should be handled with great care since they are crucial in such a weighing procedure and calibrated regularly.

4.3. Example of uncertainty budget for a dilution factor calculation

In this example, we assume that 200 mg of master solution are diluted in about 10 mL of carrier solution. The protocol presented in section 3.2.5 is followed. The master solution is weighed on the same microbalance used in table 1. The carrier solution is weighed using a semi-micro balance of 220 g capacity with scale interval 0.01 mg until 80 g with performances as follows: measured repeatability for $m < 60$ g is 30 μg ; checked maximum non-linearity for $m > 10$ g is 200 μg ; sensitivity tolerance is 10^{-6} and temperature sensitivity is $10^{-6} \text{ }^\circ\text{C}^{-1}$).

The uncertainty on the mass of master solution dispensed is given by a very similar calculation to table 1, with 200 mg instead of 20 mg. The uncertainty on the standard weight used is 3 μg . This results in an absolute standard uncertainty on the mass of master solution equal to 6.9 μg , slightly higher than the previous one. This is due to the fact that the balance is operated around the same mass (the pycnometer still weighs about 2 g). Nevertheless, since the mass of the dispensed drops is one order of magnitude higher, the relative uncertainty on this value falls to $3.4 \cdot 10^{-5}$.

For the carrier solution weighing, a 20 mL glass vial is used. Its empty weight is about 28 g and will be, at last, filled with 10 g of diluted solution (including the master solution).

The combination of the uncertainty components described in table 2 has been derived from equation (13) and the weighing methodology in section 3.2.5. The absolute standard uncertainty on the diluent mass, m_{dil} is:

$$u_{m_{dil}} = \sqrt{Bu^2 \times \left(4u_{round}^2 + 2u_{rep}^2 + 4u_{NL}^2 + 2u_{sens,full}^2 + u_{sens,empty}^2 + 2u_{TS,full}^2 + 2u_{TS,empty}^2 + 2u_{meth}^2 \right) + m_{dil}^2 \times \frac{u_{Bu}^2}{Bu^2}} \quad (15)$$

Parameter	Value	u (mg) 'w _{empty} '	u (mg) 'w _{full} '
Readability	10 μ g	0.003	0.003
Repeatability	30 μ g	0.030	0.030
Non-linearity	200 μ g	0.115	0.115
Sensitivity tolerance	1.0 $10^{-6} w^a$	0.016	0.022
Temperature sensitivity	1.0 $10^{-6} w^a$	0.009	0.013
Method	30 μ g	0.015	0.015
Buoyancy correction (relative uncertainty)	1.001034	0.00053%	
Combined standard uncertainty		0.244	
Combined relative standard uncertainty		0.0024%	

^aW stands for the mass of the vial, it is either 28 g for the empty vial or 38 g for the full one.

Table 2. Example of uncertainty budget for the weighing of 10 g of solution on a semi-micro balance of 220 g capacity.

Where the subscript 'full' refers to the last weighing of the dilution flask and 'empty' refers to the weighing of the empty flask.

The uncertainty budget related to the weighing of 10 g of carrier solution (table 2) is dominated by the uncertainty on the non-linearity of the balance, which cannot be neglected in simple weighing by difference. The uncertainty on the buoyancy correction is not negligible either, whereas the uncertainty on drop deposition (table 1) depends mainly on the balance performances and the 'method' component.

The dilution factor is calculated as the ratio between the diluent mass and the master solution mass. The uncertainty on this ratio is given by the quadratic composition of the two relative uncertainties. Thus the dilution factor is 50.000 (2), the relative uncertainty being equal to $4.2 \cdot 10^{-5}$. The main contribution usually comes from the master solution mass uncertainty.

4.4. Source preparation uncertainty from statistical analysis of primary measurements

The following example aims at illustrating the fact that the uncertainty related to source preparation can only be completely assessed with the variability of activity measurements. The plots in figure 2 represent counting rates per gram obtained for the standardization of ²²Na using the $4\pi\gamma$ technique [37] at the LNHB. For activity measurements, 9 solid sources were prepared with aliquots deposited on Mylar supports designed to be placed inside the well of a large NaI(Tl) detector (almost 4π geometry). The masses are comprised between 10 mg and 25 mg. The decay-scheme of ²²Na has two main branches: (~90%) β^+ disintegration followed by a 1275 keV γ -photon and (~10%) electron capture

followed by a 1275 keV γ -photon. The detection efficiency in the well-type detector at LHNH is about 97%. For each source, the uncertainty related to counting statistics is obtained with the standard deviation of the mean of 12 individual measurements of 90 s. The relative uncertainty for counting statistics is equal to about 0.05%. Assuming that the variability of the counting rates per gram measured for the 9 sources displayed in figure 2 (standard deviation equal to 0.07%) are mainly due to counting statistics and source preparation, the uncertainty assigned to the weighing procedure is equal to about 0.05%. This value, assessed using a statistical analysis of primary measurements (type A evaluation according to the GUM [20]), is higher than the uncertainty calculated from the uncertainty budget which can be considered as a type B evaluation. The latter, based on the equipment specifications, gives only an incomplete description of the whole uncertainty components associated with source preparation. A variability lower than 0.1% as depicted in figure 2 can be commonly obtained in specialized laboratories with a well-trained staff for source preparation. Nevertheless, the experience at the LNHB has shown that higher uncertainties can also be observed.

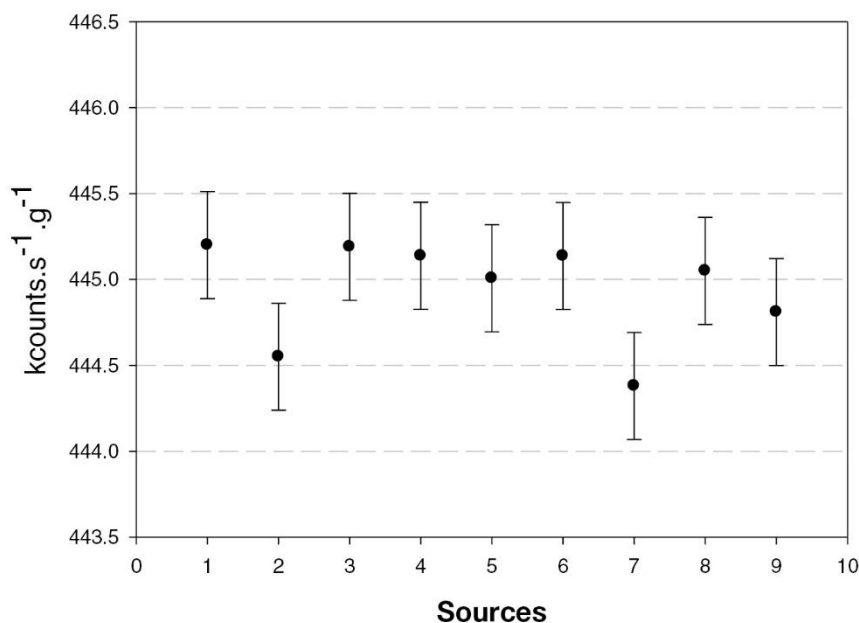


Figure 2. Counting rates per gram for the standardization of ^{22}Na using the $4\pi\gamma$ technique. The variability is mainly due to counting and to the statistical component related to source preparation.

An evidence of a correlation during source preparation (liquid scintillation) is depicted in figure 3 in the case of the standardization of ^{75}Se . This radionuclide disintegrates by electron capture mainly to the excited level of ^{75}As . For each source, the activity concentrations measured by $4\pi\beta\text{-}\gamma$ coincidence counting are displayed in the order of weighing (aliquot masses ~ 30 mg). The two consecutive sources UG6 and UG7 correspond to the extreme values of the whole distribution in figure 3. The relative uncertainty assigned to each result (0.07%) is obtained using the standard deviation of the distribution without the values corresponding to the sources UG6 and UG7 identified as correlated. This example magnifies the problem of correlations in source preparation that could be at the origin of an increase in the variability of the uncertainty. The correlation in figure 3 is difficult to explain. Selenium chemically resembles very closely sulphur, located immediately above Se in the periodic table of the elements. As a non-metal, it forms basic anions (SeO_3^{2-} at oxidation state +IV, in this example, and SeO_4^{2-} at oxidation state +VI). Nevertheless, because of the large number of possible oxidation states (including some very volatile species [30]) and their coexistence, the behaviour of Se is very complex and difficult to explore.

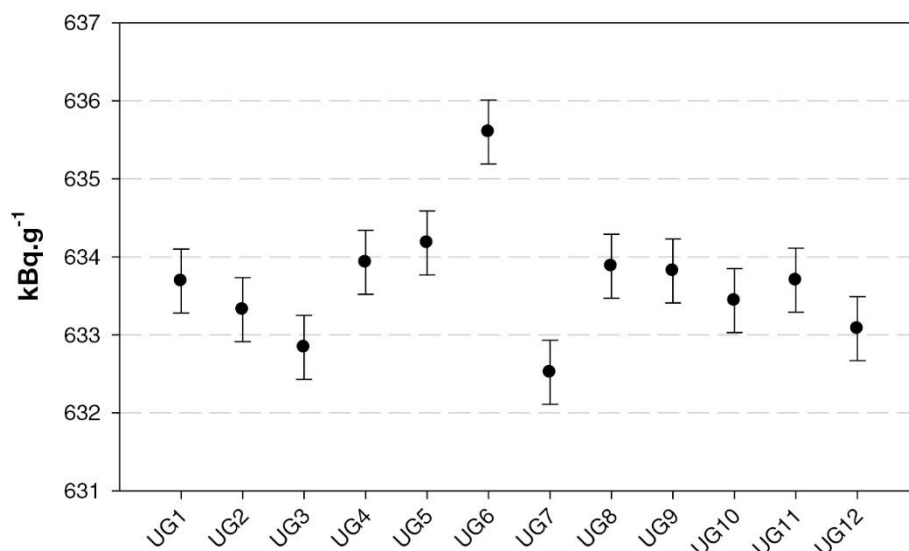


Figure 3. Observation of a correlation in source preparation for the standardization of ^{75}Se with $4\pi\beta\text{-}\gamma$ coincidence counting (liquid scintillation sources in UG scintillator displayed in the order of preparation).

5. Conclusion

When dealing with source preparation, a high accuracy is needed to avoid any bias which would directly impact the activity concentration results. Source preparation in series should be considered as a sampling step, which should be representative of the master solution. The chemical stability of the solution must be preserved from the sampling to the measurement, through the weighing step and sometimes any drying operations. These stages should be prepared and performed by skilled workers with sufficient knowledge in radiochemistry to adapt suitably the protocols of source preparation when needed. Source preparation protocols (weighing operations but also the protocols used for source preparation from carrier treatment to drying procedures) should be part of Quality Assurance documents and the staff in charge of the preparation of the sources should be trained.

Concerning the weighing step, it has been shown that uncertainties below 0.1% can be achieved on a routine basis, provided some strict protocols are followed and the personnel have been trained accordingly. It is necessary to perform the buoyancy correction in all cases where the combined relative standard uncertainty is in the 0.1% range. To achieve a complete assessment of the uncertainty in source preparation, it is highly advisable to measure a set of sources (more than 5). Indeed, experience in radionuclide measurements shows that the uncertainty in source preparation cannot be assessed considering weighing equipment only.

This uncertainty assessment gives a viewpoint on the achievable uncertainty range in source preparation in the field of radionuclide metrology. Other evaluation schemes are feasible; the most important point in uncertainty evaluation being the transparency regarding the methods or hypothesis chosen for the assessment.

References

- [1] Bailat C J, Keightley J, Nedjadi Y, Mo L, Ratel G, Michotte C and Roteta M 2014 International comparison CCRI(II)-S7 on the analysis of uncertainty budgets for 4π beta gamma coincidence counting *Metrologia* **51** 06018 (Tech. Suppl)

- [2] Collé R 2008 A primer on weighing uncertainties in radionuclidic metrology oral communication Comparisons and Uncertainties Workshop (BIPM, Pavillon de Breteuil, Sèvres, France, 17 September 2008)
- [3] Sibbens G and Altitzoglou T 2007 Preparation of radioactive sources for radionuclide metrology *Metrologia* **44** S71–8
- [4] Louvel D 2006 Balances et pesées Techniques de L'Ingénieur P1380v2
- [5] EURAMET 2011 cg-18 Guidelines on the calibration of non-automatic weighing instruments Version 3.0 (03/2011)
- [6] Picard A, Davis R S, Gläser M and Fujii K 2008 Revised formula for the density of moist air *Metrologia* **45** 149–55
- [7] OIML, R111 2004 Part 1: metrological and technical requirements Weights of Classes E1, E2, F1, F2, M1, M1-2, M2, M2-3, M3 edition (E) (Paris: International Organization of Legal Metrology) (www.oiml.org/en/files/pdf_r/r111-e04.pdf)
- [8] Wunderli S, Fortunato G, Reichmuth A and Richard P H 2003 Uncertainty evaluation of mass values determined by electronic balances in analytical chemistry: a new method to correct for air buoyancy *Anal. Bioanal. Chem.* **376** 384–91
- [9] Malengo A 2014 Buoyancy effects and correlations in calibration and use of electronic balances *Metrologia* **51** 441–51
- [10] Davidson S, Perkin M and Buckley M 2004 The measurement of mass and weight Measurement Good Practice Guide
- [11] Mettler-Toledo 2012 Guide book: weighing the right way (http://us.mt.com/dam/MT-NA/WeighMatter/Weighing_the_Right_Way_Brochure.pdf)
- [12] Gläser M 1999 Change of the apparent mass of weights arising from temperature differences *Metrologia* **36** 183–97
- [13] NISTIR-6919 2002 Recommended Guide for Determining and Reporting Uncertainties for Balances and Scales Gaithersburg, MD: National Institute of Standards and Technology
- [14] Sutton C M 2004 Magnetic Effects in Weighing (Lower Hutt: Measurement Standards Laboratory of New Zealand)
- [15] UKAS-LAB14 2006 Calibration of Weighing Machines 4th edn (Feltham: United Kingdom Accreditation Service)
- [16] G-ENAC-17 2010 Calibración de instrumentos de pesaje de funcionamiento no automático: ajustes durante el proceso de calibración. Rev1. (Entidad Nacional de Acreditación)
- [17] Sutton C M, Robinson J E and Reid G F 2010 Calibrating Balances (Lower Hutt: Measurement Standards Laboratory of New Zealand)
- [18] LAB-GTA-95 2012 Guide technique d'accréditation - Etalonnage d'instruments de Pesage à Fonctionnement Non-Automatique (Cofrac Comité Français d'Accréditation) www.cofrac.fr/documentation/LAB-GTA-95

- [19] Louvel D 2012 Étalonnage d'une balance par les utilisateurs Techniques de l'Ingénieur r1734
- [20] JCGM 100 2008 Evaluation of Measurement Data—Guide to the Expression of Uncertainty in Measurement (Sèvres: BIPM) (GUM 1995 with minor corrections)(www.bipm.org/utis/common/documents/jcgm/JCGM_100_2008_E.pdf)
- [21] OIML, R76 2006 Non-Automatic Weighing Instruments Part 1: Metrological Requirements—Tests Edition (E) (Paris: International Organization of Legal Metrology) (www.oiml.org/en/files/pdf_r/r076-1-e06.pdf)
- [22] Clark J P and Shull H 2001 Reproducibility: a major source of uncertainty in weighing Report in conjunction with Contract No.DE-AC09-96SR18500 with the U S Department of Energy
- [23] Reichmuth A 2000 Non-linearity of laboratory balances and its impact on uncertainty NCSL Workshop and Symp. (Toronto, 17–20 July 2000)
- [24] Reichmuth A, Wunderli S, Weber M and Meyer V R 2004 The uncertainty of weighing data obtained with electronic analytical balances *Microchim. Acta* **148** 133–41
- [25] Campion P J, Dale J W G and Williams A 1964 A study of weighing techniques used in radionuclide standardization *Nucl. Instrum. Methods* **31** 253
- [26] van der Eijk W and Moret H 1967 Precise determination of drop weights *Proc. of the 1966 Symp. on Standardization of Radionuclides held (Vienna, 10–14 October 1967)* 529
- [27] van der Eijk W and Vaninbrouckx R 1972 Sampling and dilution problems in radioactivity measurements *Nucl. Instrum. Methods* **102** 581–7
- [28] Le Gallic Y 1973 Problems in microweighing *Nucl. Instrum. Methods* **112** 333–41
- [29] Iroulart M G 2007 Thermodynamic Stability of Radioactivity Standard Solutions (Sèvres: Monographie BIPM-6)
- [30] Letho J and Hou X 2011 Chemistry and Analysis of Radionuclides. Laboratory Techniques and Methodology (Weinheim: Wiley)
- [31] NCRP-Report-58 1985 Techniques for the preparation of standard sources for radioactivity measurements A Handbook of Radioactivity Measurements Procedures (Report 58) 2nd edn (Bethesda, MD: National Council on Radiation Protection and Measurements)
- [32] Campion P J 1975 Procedures for Accurately Diluting and Dispensing Radioactive Solutions (Sèvres: Monographie BIPM-1)
- [33] Merritt J S 1976 Some mass measurement problems *Proc. of the 1974 Annual Conf. of the Nuclear Target Development Society* 86
- [34] Merritt J S 1973 Present status in quantitative source preparation *Nucl. Instrum. Methods* **112** 325–32
- [35] Rytz A, Colas C and Veyradier C 1969 Some experiments on the dilution of radioactive solutions Unpublished report issued by the Bureau International des Poids et Mesures Paris

[36] Fitzgerald R, Bailat C, Bobin C and Keightley J D 2015 Uncertainties in $4\pi\beta\text{-}\gamma$ coincidence counting This Metrologia Special Issue

[37] Thiam C, Bobin C, Maringer F J, Peyres V and Pommé S 2015 Assessment of the uncertainty budget associated with $4\pi\gamma$ counting Metrologia **52** S97–107