

# Determination of $^{210}\text{Pb}$ at ultra-trace levels in water by ICP-MS

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## Abstract

$^{210}\text{Pb}$  ( $t_{1/2} = 22.26$  years) is one of the most difficult naturally occurring radionuclides to analyze directly via radiometric measurement due to its low-energy radioactive decay. In this work, inductively coupled plasma mass spectrometry (ICP-MS) was investigated as a possible alternative to radiometric counting for the determination of  $^{210}\text{Pb}$ . The formation of isobaric and polyatomic (molecular) interferences was studied and various strategies (i.e. co-precipitation, extraction chromatography, and derivatization) were tested to assess their usefulness in reducing these interferences. In addition, the potential of these strategies as pre-concentration methods was demonstrated. A  $^{210}\text{Pb}$  pre-concentration of >300-fold and recoveries of 63–73% were obtained using a combined co-precipitation and extraction chromatography protocol followed by derivatization using an ethylating reagent. The abundance sensitivity at  $m/z = 210$  in the presence of stable Pb was also investigated for three types of mass spectrometers (sector-field (ICP-SFMS), quadrupole-based (ICP-QMS), and quadrupole-based with a dynamic reaction cell (ICP-QMS (DRC))). Finally, the method was applied to determine  $^{210}\text{Pb}$  in water samples and a detection limit of  $90 \text{ mBq L}^{-1}$  ( $10 \text{ pg L}^{-1}$ ) was obtained.

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## 1. Introduction

$^{210}\text{Pb}$  belongs to the  $^{238}\text{U}$  decay series and has a half-life ( $t_{1/2}$ ) of 22.26 years. Because it originates from the successive decay of many short-lived intermediates starting with  $^{222}\text{Rn}$ ,  $^{210}\text{Pb}$  is of interest in both environmental and health studies. Applications of radioisotopes of Pb as an environmental tracer have been demonstrated in many scientific disciplines. The radiotoxicity of  $^{210}\text{Pb}$ , mainly resulting from its relatively long radiological half-life in comparison to environmental processes, its chemical similarities with alkaline earth elements, and the high energy associated with the decay of its progeny (see Fig. 1), has pushed many governmental organizations to include in their legislation strict regulations for exposure to this radionuclide. One example is the

maximum acceptable concentration (MAC) in drinking water of  $0.1 \text{ Bq L}^{-1}$  for  $^{210}\text{Pb}$ ; the lowest for any of the naturally occurring radionuclides [1,2]. Thus, it would be expected that  $^{210}\text{Pb}$  measurements must be routinely performed for radiological safety. However, this is not the case, mainly due to analytical limitations which arise from the nature of  $^{210}\text{Pb}$  decay.

$^{210}\text{Pb}$  decays into  $^{210}\text{Bi}$  through the emission of a low energy  $\beta$ -particle (64 keV) (Fig. 1), which is rapidly reabsorbed by the sample matrix before reaching the detector. An alternative for the direct measurement of  $^{210}\text{Pb}$  is through  $\gamma$ -spectrometry of the ray emitted with an energy of 47 keV; with an average only four times per 100 decays. However, the low energy and rate of emission associated with this mode of decay make this type of measurement time-consuming, especially at low activities (up to several days of counting per sample) [3]. Other measurement alternatives, based on the achievement of secular equilibrium between  $^{210}\text{Pb}$  and its progeny ( $^{210}\text{Bi}$  or  $^{210}\text{Po}$ ) are effective for the indirect determination of  $^{210}\text{Pb}$  activity [4–6]. Nevertheless, these approaches

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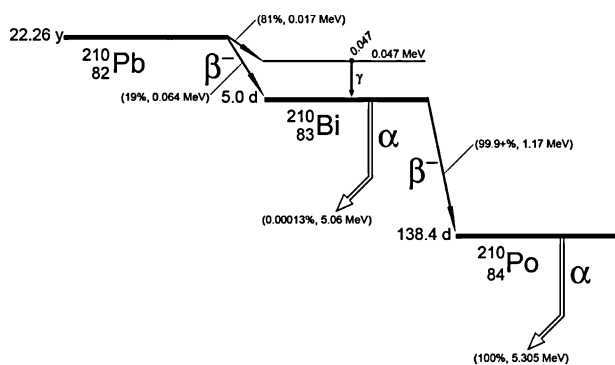


Fig. 1.  $^{210}\text{Pb}$  radioactive decay scheme.

necessitate long storage periods (up to 2 months) and complex separation procedures. Consequently, an alternative to radiometric measurements that would provide similar analytical performance would be beneficial.

Recently, inductively coupled plasma mass spectrometry (ICP-MS) has been used for the determination of many radioisotopes including  $^{99}\text{Tc}$ ,  $^{226}\text{Ra}$ ,  $^{235,236,238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{239,240}\text{Pu}$ , based on their masses instead of their radioactive properties [7–13]. It is accepted that, for radioactive isotopes with short half-lives, radiometric techniques generally offer better and faster analyses than ICP-MS since the number of disintegrations per mole of atoms is higher than for long-lived radionuclides [14]. Nevertheless, for some of these short half-life radionuclides such as  $^{210}\text{Pb}$ , determination by conventional radiometric techniques is tedious and time-consuming and consequently, the detection of this radionuclide by mass spectrometry might be a viable alternative.

The direct determination of  $^{210}\text{Pb}$  by  $\gamma$ -spectrometry requires a knowledge of the sample geometry and composition, and has a small sample throughput (<1 sample per day), while sample geometry is unimportant for ICP-MS and the analysis time is rapid (i.e. within a minute). Despite these clear advantages, mass spectrometry has two major issues that must be addressed before  $^{210}\text{Pb}$  can be determined at environmental levels. The first is the presence of isobaric and possible molecular interferences that can significantly influence the measurement of  $^{210}\text{Pb}$  by generating a signal

at  $m/z = 210$ . Second,  $^{210}\text{Pb}$  is found in the environment at activities ranging from a few millibecquerels to several Bq, depending on the medium studied [15]. These activities, converted to masses, represent only a few femtograms and are significantly lower than the typical detection limits of most ICP-MSs using direct aspiration nebulization. This suggests that a pre-concentration of the sample is necessary in order to increase the number of atoms detected. The purpose of this study is to investigate strategies to eliminate/reduce spectral interferences and to develop a protocol for pre-concentrating and isolating  $^{210}\text{Pb}$  from aqueous matrices.

## 2. Experimental

### 2.1. Instrumentation

Two types of ICP-MS were used to investigate  $^{210}\text{Pb}$  determination by mass spectrometry: a sector-field double focusing (SF) ICP-MS and a quadrupole (Q) ICP-MS. An Element2 SF-ICP-MS (ThermoFinnigan, Bremen, Germany) was used to determine spectral interference and abundance sensitivity, while a Perkin-Elmer Elan-5000 and Elan-6100 (Dynamic Reaction Cell), both ICP-QMS, were used for abundance sensitivity and sample measurements. The conditions used are presented in Table 1. In order to evaluate the precision and accuracy of the proposed method, samples were measured using both ICP-MS and  $\gamma$ -spectrometry. Radiometric measurements were performed using a germanium detector (BE5030, Canberra, Concord, ON, Canada) protected by a carbon window. The samples were counted, after ammonium pyrrolidine dithiocarbamate (ADPC) co-precipitation (solution redissolved in 25 mL 3 M  $\text{HNO}_3$ ) in a 7.16 i.d.  $\times$  0.62 cm plastic container with a wall thickness of 0.226 cm. The 46.5 keV ray (4.25% intensity) was used to determine the sample activity. Counting was performed for a period longer than 2 days. The efficiency of the counting geometry was determined using the Virtual Gamma Spectroscopy Laboratory software (VGSL, Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization, Vienna, Austria).

Table 1

Experimental parameters for the Perkin-Elmer SCIEX Elan-5000 and 6100 (DRC) for the measurement of  $^{210}\text{Pb}$

| Instrument            | Elan-5000 ICP-QMS                           | Elan-6100 (DRC) ICP-QMS                     |
|-----------------------|---|---|
| Forward power         | 1050 W                                      | 1050 W                                      |
| Coolant gas (Ar)      | 15 L $\text{min}^{-1}$                      | 15 L $\text{min}^{-1}$                      |
| Auxiliary gas (Ar)    | 0.8 L $\text{min}^{-1}$                     | 1.2 L $\text{min}^{-1}$                     |
| Nebulizer gas I (Ar)  | 800 mL $\text{min}^{-1}$                    | 1110 mL $\text{min}^{-1}$                   |
| Nebulizer gas II (Ar) | 150 mL $\text{min}^{-1}$                    | 150 mL $\text{min}^{-1}$                    |
| Torch position        | Optimized daily on $^{208}\text{Pb}$ signal | Optimized daily on $^{208}\text{Pb}$ signal |
| Sampling cone         | 1.0 mm nickel or platinum                   | 1.0 mm nickel                               |
| Skimmer cone          | 0.8 mm nickel                               | 0.8 mm nickel                               |
| Lens voltage          | Optimized daily on $^{208}\text{Pb}$ signal | 6.5 V                                       |
| Mass resolution       | 500   | 500   |
| Dwell time            | 30 ms                                       | 30 ms                                       |
| Mass monitored        | 129 and 210                                 | 129 and 210                                 |

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