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Concentration of natural radionuclides in raw water and packaged drinking water and the effect of water treatment

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ABSTRACT

The raw water (RW) samples collected from natural sources are subjected to water treatment process, including reverse osmosis (RO), and are packed in bottles as packaged drinking water (PDW). Raw water (21 samples) taken from deep wells of Chennai and Secunderabad which are used in the production of PDW, were analysed for ^{234}U, ^{235}U, ^{238}U, ^{226}Ra, ^{228}Ra and ^{210}Pb activity concentrations. Activity Concentrations of ^{234}U, ^{235}U, ^{238}U, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po in PDW were also analysed. The mean activity concentrations of ^{234}U, ^{235}U, ^{238}U, ^{226}Ra, ^{228}Ra and ^{210}Pb in RW at Chennai were 12.1, ≤1.3, 7.1, 2.6, 27.5, and 16.3 mBq/L respectively. The mean activity concentrations of ^{234}U, ^{235}U, ^{238}U, ^{226}Ra, ^{228}Ra and ^{210}Pb in RW at Secunderabad were found to be 40.9, 1.7, 41.5 84.5, 100.1, and 17.0 mBq/L respectively. The mean concentrations of ^{234}U, ^{235}U, ^{238}U, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po in PDW at Chennai were found to be ≤1.3, ≤1.3, ≤1.3, ≤0.2, ≤1.7, 28.0 and 1.2 mBq/L at Secunderabad were found to be ≤1.3, ≤1.3, 1.7, 4.3, 5.0 and 28.1 mBq/L. The study indicated a considerable reduction in the concentration of natural radionuclides due to water treatment. The reduction ratios of RW to PDW for ^{234}U, ^{238}U, ^{226}Ra, ^{228}Ra were 97, 96, 94 and 95%. In case of ^{210}Pb, the PDW showed higher concentration of ^{210}Pb than RW. This was due to its in growth from ^{222}Rn which was not removed in the RO process.

1. Introduction

The main source of drinking water consumed by the urban population of South India is from bore wells. This water often does not meet the drinking water quality standards due to high levels of total dissolved solids, chlorides and hardness and also due to poor bacteriological quality. So, among the urban population of South India, packaged drinking water (PDW) is fast becoming the primary source of drinking water. The water resources used in the production of PDW are mostly open wells and bore wells. Due to varying concentrations of uranium, thorium, and their daughter products (UNSCEAR, 1998) in ground water, the estimation of natural radionuclides in drinking water becomes relevant in assessing their contribution to internal dose by ingestion. Uranium a naturally occurring heavy and radioactive element present in the earth's crust can leach to the ground water and contribute to the internal dose through drinking water and through the local food stuff in these areas. Concentrations of uranium in groundwater depends on many factors, major one being its concentration in the aquifer rocks. Other factors like pH, redox potential, oxygen and carbon dioxide content of water as well as the presence of chelating agents can influence the concentration of uranium in natural water (Friha Hadj Ammar et al., 2010).

^{226}Ra and ^{228}Ra are considered important due to their long radiological and biological half-lives. ^{226}Ra an alpha emitter ($t_{½}$ = 1600 y) is produced by the decay of ^{238}U and ^{228}Ra a beta emitter ($t_{½}$ = 5.75 y) is produced by the decay of ^{232}Th. Both ^{226}Ra and ^{228}Ra are metabolically similar to calcium, and hence ingestion of these radium isotopes results in appreciable fraction being deposited on bone surface. About 20% of this fraction is absorbed and distributed to soft tissues and bones, but its retention is mainly in growing bone (USEPA, 1991). ^{210}Pb is beta emitter ($t_{1/2}$ = 22.3 y) produced by the decay of ^{238}U. ^{210}Po is alpha emitter ($t_{1/2}$ = 138.4 days) which is also produced by the decay of ^{238}U.

Raw water (RW) from open wells and bore wells, after filtration, is purified by reverse osmosis (RO) process, sterilised and is then marketed as PDW. Membrane techniques, such as RO or the nanofiltration used for making ground water potable are capable of removing uranium, thorium and their daughter products.

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In this study, 21 RW samples taken from deep wells of Chennai and Secunderabad, which are used in the production of PDW were analysed for ^{234}U, ^{235}U, ^{238}U, ^{226}Ra, ^{228}Ra and ^{210}Pb concentration levels. Concentrations of ^{234}U, ^{235}U, ^{238}U, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po in PDW were also analysed. Estimation of uranium isotopes, and radium isotopes and lead were not carried out in the same samples collected from the same location of Chennai and Secunderabad. The aim of the present work was to assess the reduction in concentrations for ^{238}U, ^{234}U, ^{226}Ra, ^{228}Ra and ^{210}Pb in PDW because of RO process.

2. Material and methods

Uranium in the water samples was co-precipitated with $Fe(OH)_3$ (Herranz et al., 1997). The Fe was removed by solvent extraction with Di-isopropyl ether. Uranium was separated using anion exchange method, electroplated on a stainless steel planchet and the activity was estimated by an alpha spectrometry system, Eurisys Mesures, Model No.7184. The minimum detectable limit (MDL) for uranium isotopes was 1.3 mBq/L for the counting period of 3000 s. The chemical Recovery of ^{238}U was determined in a spike experiment. In this experiment demineralised water was spiked with a natural ^{238}U standard, chemical separation was carried out using the same procedure of that of PDW. To ensure complete precipitation of $Fe(OH)_3$, the precipitation was carried out twice and taken for further separation of uranium. Ten different spiked samples were used to measure the chemical recovery. The mean chemical recovery was found to be 74.0 ± 1.5%. Hence a chemical recovery 75% was used for calculation of uranium activity concentrations in PDW.

Using the same procedure, 8 IAEA water samples were analysed for radium and uranium radionuclides. Most of the samples were within the acceptable range of IAEA.

^{226}Ra and ^{228}Ra from the water sample were preconcentrated by passing an aliquot of filtered water (≈20 L) through manganese impregnated acrylic fibre packed into a glass column. Radium isotopes along with the manganese matrix were leached from the fibre with hot HCl, evaporated to dryness, dissolved in 4N HNO_3, and loaded in a radon bubbler. ^{226}Ra in the samples were estimated by emanometry method (Iyengar et al., 1989). The minimum detection limit for ^{226}Ra was 0.2 mBq/L for a counting time of 1000 s.

Following ^{226}Ra estimation by emanometry, ^{228}Ra present in the solution was determined by co-precipitating as $BaSO_4$ and $PbSO_4$. $BaSO_4$ alone was precipitated and kept aside for it to attain equilibrium with its daughter ^{228}Ac ($t_{1/2}$ = 6.15 h). Then $BaSO_4$ was dissolved in hot perchloric acid in the presence of Lanthanum-carrier and hold back carriers lead and bismuth. Barium and lead sulphates were first precipitated from perchloric acid solution by addition of dilute sulphuric acid and then $La(Ac)F_3$ (Kannan, 2004) was precipitated in the supernatant, which was separated and counted in a low background beta counting system (LBBCS). The minimum detection limit was 1.7 mBq/L for a counting period of 3600 s for both ^{228}Ra and ^{210}Pb.

$PbSO_4$ present in the supernatant was kept aside, to attain equilibrium with its daughter ^{210}Bi. On reaching equilibrium, 200 mg calcium carrier was added. ^{210}Bi was precipitated as $Bi(OH)_3$ by the addition of NH_4OH and dissolved in 1N HNO_3. $Bi(PO_4)$ was precipitated using phosphoric acid which was counted using the LBBCS. The activity for ^{210}Pb was back calculated from the content of ^{210}Bi (Kannan, 2004). The weight of $Bi(PO_4)$ is much less (~15 mg only), so there will be little self absorption. The background and efficiency (by using KCl standard) of the detector was checked daily. The mean background of the beta counter was found to be 1.33 cpm. ^{210}Bi was measured immediately and the correction was applied for ^{210}Bi growth. ^{210}Bi was recounted again every 5 days to check for ^{210}Bi decay and it was found that the decay followed the half life of ^{210}Bi. Using the same procedure one IAEA water sample was analysed for ^{210}Pb and the error was <13%.

Estimation of ^{210}Po was carried out by co-precipitating along with $Fe(OH)_3$ and it was dissolved in 0.5 N HCl. ^{210}Po from the acidified sample was deposited on to a silver planchet by electrochemical deposition. Finally, both sides of the planchet were counted in the alpha counting system using a ZnS(Ag) scintillation detector. The minimum detection limit was 0.3 mBq/L for counting period of 5000 s (Kannan, 2004). Similar to other isotopes, the chemical recovery of ^{210}Po was also determined by spike experiment method. The mean chemical recovery obtained was 98.4 ± 1.6%. Hence a chemical recovery 99% was used in the calculation of ^{210}Po activity concentrations in PDW.

The lab participated in the IAEA-CU-2007-09 proficiency test for the determination of ^{210}Po in water samples. All the samples analysed were in the acceptable range of IAEA.

3. Results and discussion

3.1. Uranium concentration in raw and packaged drinking water

Uranium concentrations (^{234}U, ^{235}U and ^{238}U) present in both RW and PDW from Chennai and Secunderabad are given in Table 1 along with the means. The ^{234}U activity concentrations in RW ranged from ≤1.3 to 12.5 with a mean of 12.1 mBq/L at Chennai. At Secunderabad the range was 22.0–66.2 with a mean of 40.9 mBq/L. Similarly ^{235}U activity concentrations in RW were observed to be in the range of ≤1.3 and ≤1.3 to 2.9 (mean:1.7) mBq/L at Chennai and Secunderabad respectively. ^{238}U activity concentrations in RW at both locations were 1.4–10.7 (mean: 7.1) and 22.8 to 67.5 (mean: 41.5) mBq/L respectively. Uranium is ubiquitous and is present in wide ranges in different regions. It varies from 0.5 to 150,000 mBq/L in Finland 0.1–700 mBq/L in China and 0.1–1000 mBq/L in Switzerland (UNSCEAR, 2000).

In most of the reference papers uranium concentrations are given in mass unit. In order to compare with other papers, the obtained mean activity concentrations of ^{234}U, ^{235}U and ^{238}U were converted

Table 1
^{234}U,^{235}U and^{238}U in raw water and packaged drinking water.

Sr. No.	Location	Activity (mBq/L)					
		^{234}U		^{235}U		^{238}U	
		Raw water	PDW	Raw water	PDW	Raw water	PDW
1	Chennai T.N	11.7 ± 1.8	≤1.3	≤1.3	≤1.3	9.3 ± 1.6	≤1.3
2	Chennai T.N	≤1.3	≤1.3	≤1.3	≤1.3	1.4 ± 0.3	≤1.3
3	Chennai T.N	12.5 ± 1.8	≤1.3	≤1.3	≤1.3	10.7 ± 1.7	≤1.3
Mean		12.1	≤1.3	≤1.3	≤1.3	7.1	≤1.3
1	Secunderabad A.P	38.2 ± 2.4	≤1.3	2.7 ± 0.6	≤1.3	34.3 ± 2.3	≤1.3
2	Secunderabad A.P	63.1 ± 5.3	≤1.3	≤1.3	≤1.3	64.4 ± 5.3	2.2 ± 0.9
3	Secunderabad A.P	22.0 ± 2.4	≤1.3	≤1.3	≤1.3	22.8 ± 2.5	≤1.3
4	Secunderabad A.P	24.9 ± 2.6	≤1.3	≤1.3	≤1.3	26.3 ± 2.7	≤1.3
5	Secunderabad A.P	66.2 ± 5.4	≤1.3	≤1.3	≤1.3	67.5 ± 5.5	2.2 ± 0.8
6	Secunderabad A.P	45.2 ± 2.6	≤1.3	2.9 ± 0.7	≤1.3	43.7 ± 2.6	2.6 ± 0.9
7	Secunderabad A.P	26.7 ± 2.7	≤1.3	≤1.3	≤1.3	31.2 ± 2.9	≤1.3
Mean		40.9	≤1.3	1.7	≤1.3	41.5	1.7

MDA for Uranium Isotopes: 1.3 mBq/L.

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