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Study on Radioactivity Associated with Common Salt Production Process of Two Salt Works in Tuticorin District, Tamil Nadu, India

P. PREMA KUMARI[1,*], C.G. MANIYAN[2], A.P. LIPTON[3] and M. CHANDRAN[4]

[1]Department of Chemistry, N.M.S. Kamaraj Polytechnic College, Pazhavilai-629 501, India
[2]Health Physics Unit, Indian Rare Earths Limited, Manavalakurichi-629 252, India
[3]Central Marine Fisheries Research Institute, Vizhinjam, Thiruvananthapuram-695 521, India
[4]Department of Chemistry, Vivekananda College, Agastheeswaram-629 701, India

*Corresponding author: Tel: +91 4652 264101; E-mail: prema2008phd@gmail.com

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This study estimated the gross α, gross β, ^{40}K, ^{208}Tl, ^{228}Ac and ^{214}Bi activities in the sea and subsoil brines of two salt works at Vepalodai and Sahupuram in Tuticorin District, Tamil Nadu, India. It was observed that the sea brine of at Vepalodai, the gross α activity in the brine varied from 0.0009 to 0.0033 Bq L^{-1} and the gross β activity varied from 0.0194 to 4.0405 Bq L^{-1}. At the subsoil brine of Sahupuram, the corresponding values varied from below detection limit (BDL) to 0.0032 and 0.0114 to 3.0352 Bq L^{-1}. ^{40}K activity was found to increase, source to bittern, *viz.*, 4.006 to 221.006, at Veplodai and 3.247 to 204.462 Bq L^{-1} at Sahupuram. The activity of the sea brine was higher than that of the sub-soil brine. The gross α and β activities were increasing at subsequent stages of production due to the concentration of brine. The sediments collected at different stages showed gross α activity varying from 42.0 to 362.0 Bq kg^{-1}, gross β activity varying from 1998 to 2498 Bq kg^{-1}. ^{208}Tl, ^{228}Ac activities varied from 111.2 to151.2 Bq kg^{-1} and 40.1 to 73.4 Bq kg^{-1}. The corresponding values for Sahupuram salt works were 10.2 to 98.7, 1298 to 1548, 137.4 to 175.8 and 33.8 to 55.3 Bq kg^{-1} for gross α, gross β, ^{208}Tl, ^{228}Ac, respectively. The analysis of the common salt from Vepalodai showed 14.4,107, 12.42, 13.10 and 3.8 Bq kg^{-1} of gross α, gross β, ^{208}Tl, ^{228}Ac and ^{214}Bi, respectively and that from sub soil brine of Sahupuram showed 8.7, 24.6, 7.4, 9.46 and 3.4 Bq kg^{-1}. This study shows that the radioactivity in common salt is within the limit.

Key Words: α, β activity, Salt works, Sea and Sub-soil brine, Sediments.

INTRODUCTION

Common salt is one of the most plentiful substances on earth. Common salt is one of the best known minerals and the first substances after water to have attracted human attention[1,2]. Common salt (NaCl) has several thousands of applications in the chemical industry, which consumes 94 % of the salt produced and the rest (6 %) is consumed by the human population[3]. The manufacturing of common salt from sea water or natural brine using solar energy and wind is a popular process since time immemorial. In solar salt work a series shallow ponds are used for the evaporation of brine. As evaporation proceeds, the concentration of brine goes on increasing with increasing temperature of the brine[4]. The brine from the source is pumped to large reservoirs of 3-4 m depth and made to flow through underground trenches and stored there. This brine is sent to primary, secondary and tertiary condensers, arranged in series, for evaporation. The concentrated brine from condensers is let into crystallizers, for production of salt, using solar energy[5]. The original volume of the water reduces to 3 %, when bittern (the supernatant liquid obtained after the precipitation of NaCl) is formed[6]. The brine density expressed as salinity Baume (ºBe), is a good leaching agent of radioactivity. At a salinity of 13-24 Baume, $CaSO_4 \cdot 2H_2O$ (gypsum) gets precipitated at the condensing stage[7]. Because of the chemical affinity, Ra, Th, *etc.* are also susceptible to precipitate and subsequent removal. This advantageously eliminates radioactivity from the common salt, without the addition of any external agents. Potassium, another natural radionuclide of immense importance in the body functions and metabolism, is an important constituent of sea water (1 g/L) and likely to follow a different pattern from the brine to bittern stage, due to its high solubility. It is estimated that the human receives about 0.20 m Gy/y of radiation dose due to ^{40}K present in human body[8].

In India, Tamil Nadu ranks second in the production of salt and the total production comes from solar evaporation of sea brine or subsoil brine. 15 % of the country's production is contributed by Tamil Nadu. In it, Tuticorin stands first position. It has a saltpan area of 809.7 ha under operation.

EXPERIMENTAL

The present study has been carried out on common salt manufactured from two salt works at Tuticorin District of Tamil Nadu. While Vepalodai salt work uses the sea brine from Bay of Bengal, Sahupuram salt work uses the sub-soil brine. The study estimates radioactivity associated with salt samples and the supernatant water samples, before and after the production of salt. As potassium along with sodium has several vital functions in the metabolism and electrolyte balance of human body, the concentration of potassium in common salt is also examined. This communication discusses statistical analysis of the results and minimum detectable levels of different activity. Brine, sediments, bittern and salt, formed at different stages of manufacture of common salt, were collected on a monthly basis for a period of one year.

The radioactivity of the brine samples was calculated after the precipitation by $BaCl_2$ and $CaSO_4$. The composite samples were dried in an electrical air oven at 120 °C, powdered in an agate mortar and suitable amount of substances were counted for α and β activity. Gross α and gross β activities were determined using low-background α counting system ZnS (Ag), ECIL, Model RCS-4027 and gas flow β counting system (ECIL Model BCS 36A). The efficiency of the α counter was determined using ^{239}Pu source of strength 542 dpm (disintegration per minute) and the efficiency was estimated to be 30 %. KCl with β activity of 1000 Bq was used for estimating β efficiency, which was calculated to be 40 %. For the estimation of γ efficiency, pure monazite sources of known strengths were used and the daughter products such as ^{208}Tl, ^{228}Ac, ^{214}Bi were considered assuming equilibrium with the parents. This assumption generally holds good as the minerals do not undergo any chemical processing. The comparison of different peaks also gives an idea of equilibrium. Th and U activity in the common salt were determined by counting 500 g of samples in MCA, with 2″ × 2″ NaI (T1) as detector[9]. ^{208}T1 (2614 KeV, 100 % emission) and ^{214}Bi (609 KeV, 46 % emission) peaks were taken for calculation of Th and U, respectively, assuming equilibrium. ^{228}Ac (910 KeV, 29 %) represents ^{228}Ra and the peak energy of 1460 KeV (11 %) was considered for ^{40}K. The counting time varied from 5000 to 10,000 s for maximum accuracy possible. When radioactivity, particularly of low levels is measured, it is essential to make an assessment on the accuracy of the result in terms of per cent error and confidence level. Hence, the result presented in this work has also undergone such an analysis. From a series of studies done on calibration and derivation of the sensitivity of the system, it has been concluded that the system has a background of 0.340, 0.212, 0.101 and 0.097 cps for ^{208}Tl, ^{214}Bi, ^{40}K and ^{228}Ac, respectively. With mineral monazite and KCl as standard sources, the derived sensitivities for different energies of interest were calculated to be 8.78×10^{-4}, 7.6×10^{-3}, 1.46×10^{-3} and 4.5×10^{-3} cps Bq^{-1} for ^{208}Tl, ^{214}Bi, ^{40}K and ^{228}Ac, respectively. Thus for ^{208}Tl, with the highest background and lowest sensitivity, a background of 0.34 cps works out to 380 Bq. It is quite imperative that the estimation of low level of activity involves a lot of uncertainty. Hence, the lowest activity that can be presented with relatively low error and better confidence level has been calculated based on the relation:

$$\sigma = \sqrt{\frac{r_g}{t_g} + \frac{r_{bg}}{r_{bg}} \times \frac{1}{(r_g - r_{bg})'}}$$

where r_g is the counting rate of the sample, t_g the counting time in seconds, r_{bg} the background counting rate and t_{bg} is the background counting time. It has been calculated that an activity of 10 Bq can be presented with 10 % error at 1 σ, if the sample is counted for 8078 s. Hence, all the samples were counted for a rounded up period of 8000 s. For other radionuclides, because of their better sensitivity and comparatively lower background, still lower values can be presented with better reliability. With a system background of 2 counts per 5000 s and an efficiency of 30 %, α activity of 0.001 Bq is presented with 5 % error at 2 σ. Similarly, β activity is also presented with 5 % error at 2 σ, as they are counted for 3000 s, in a low background (3 counts per min) β counting system having high efficiency (40 %; ref. 9). The MDL for γ activity can be stated to be 10 Bq/kg with 10 % error at 1 σ.

RESULTS AND DISCUSSION

Table-1 provides the details of radioactivity encountered at various stages of the production of salt from two sources at Vepalodai and Sahupuram. Gross α and gross β activities are minimum at the brine stage. The activity gets increased as the brine concentrated in the subsequent stages. At bittern stage, there is a drastic increase in the β activity, disproportional to the earlier pattern. This result was in agreement with the brine samples in kanyakumari District[10]. Subsequent analysis of the brine samples revealed the accumulation of ^{40}K in the bittern, as potassium salt is highly soluble and remains mostly with the solution, while the salt gets crystallized. This trend also indicates that radioactivity originating from Th and U preferred to stay with solid samples. There is a steady increase of potassium activity in the brine at various stages, as its concentration gets increased, the highest being at the bittern stage. The original volume of the brine is reduced to 3 % at bittern stage. This trend also indicates the high solubility of potassium salts. The samples of Vepalodai showed slightly higher activity than that of Sahupuram at all stages, indicating that the sea brine sample is more active than the sub-soil brine.

Table-2 gives the radioactivity associated with the sediments collected at different stages of production of salt at Vepalodai and Sahupuram. The sediments collected at the condenser stage showed maximum β activity and can be attributed to the precipitation of gypsum, at this stage, at salinity above 19 °Be. Gypsum ($CaSO_4 \cdot 2H_2O$) carries similar radioactive substances like Ra, Th, *etc.* along with it, resulting in higher radioactivity. Here again the samples from Vepalodai showed activity higher than that of Sahupuram, as observed in Table-1, indicating a lower activity for sub-soil brine than that of surface brine. ^{208}Tl and ^{228}Ac normally used for representing Th activity in γ spectrometric analyses. But here, it can be seen that there is clear disequilibrium between these two radionuclides. This also indicates that ^{226,228}Ra has separate sources of origin, other than Th and U. The higher activity of ^{228}Ac at the condenser stage is an indication of higher precipitation of ^{228}Ra at this stage, due to high salinity. These values are in agreement with the earlier report[10].

TABLE-1
MEAN AND STANDARD DEVIATION VALUES OF THE RADIOACTIVITY ASSOCIATED WITH BRINE SAMPLES COLLECTED AT VARIOUS STAGES OF PRODUCTION OF COMMON SALT

Salt Works	Various stages	Activity (Bq L^{-1})		
		Gross α	Gross β	^{40}K
Vepalodai (Sea brine)	Source	0.0009 ± 0.00005	0.0194 ± 0.001	4.006 ± 0.52
	Reservoir	0.0018 ± 0.00030	0.0241 ± 0.001	11.876 ± 1.11
	Condenser	0.0024 ± 0.00010	0.0292 ± 0.002	24.340 ± 2.82
	Crystallizer	0.0029 ± 0.00020	0.0343 ± 0.001	67.304 ± 6.37
	Bittern	0.0033 ± 0.00010	4.0405 ± 0.200	221.006 ± 21.20
Sahupuram (Sub soil brine)	Source	BDL	0.0114 ± 0.002	3.247 ± 0.28
	Reservoir	0.0009 ± 0.00003	0.0212 ± 0.001	10.465 ± 1.04
	Condenser	0.0012 ± 0.00006	0.0291 ± 0.002	28.480 ± 2.48
	Crystallizer	0.0022 ± 0.00010	0.0308 ± 0.001	88.796 ± 8.78
	Bittern	0.0032 ± 0.00020	3.0352 ± 0.121	204.462 ± 19.44

TABLE-2
MEAN AND STANDARD DEVIATION VALUES OF THE RADIOACTIVITY ASSOCIATED WITH SEDIMENTS COLLECTED AT VARIOUS STAGES OF PRODUCTION OF COMMON SALT

Salt works	Various stages	Activity (Bq kg^{-1})			
		Gross α	Gross β	^{208}Tl	^{228}Ac
Vepalodai (Sea brine)	Source	42.0 ± 2.50	1998.0 ± 99.21	151.2 ± 16.12	40.1 ± 4.11
	Reservoir	91.4 ± 4.12	618.0 ± 31.92	68.4 ± 7.22	24.2 ± 2.92
	Condenser	152.3 ± 7.58	3148.5 ± 157.42	149.2 ± 14.99	152.6 ± 15.62
	Crystallizer	184.5 ± 8.62	1895.0 ± 94.75	120.1 ± 13.01	49.5 ± 5.02
	Bittern	362.0 ± 18.11	2498.0 ± 124.90	111.2 ± 12.15	73.4 ± 6.79
Sahupuram (Sub soil brine)	Source	10.2 ± 0.51	1548. ± 77.40	175.8 ± 16.52	55.3 ± 6.35
	Reservoir	28.4 ± 1.24	688.40 ± 35.41	83.4 ± 8.43	28.0 ± 2.40
	Condenser	56.2 ± 3.01	1945.2 ± 97.25	175.6 ± 16.95	173.7 ± 18.12
	Crystallizer	89.2 ± 4.46	1154.0 ± 58.01	154.3 ± 15.50	49.2 ± 5.92
	Bittern	98.7 ± 3.96	1298.0 ± 65.21	137.4 ± 12.89	33.8 ± 3.83

TABLE-3
MEAN AND STANDARD DEVIATION VALUES OF THE RADIO ACTIVITY IN COMMON SALT

Salt works	Radio activity (Bq Kg^{-1})				
	Gross α	Gross β	^{208}Tl	^{228}Ac	^{214}Bi
Vepalodai (Sea brine)	14.4 ± 0.81	107.0 ± 5.35	12.42 ± 1.20	13.10 ± 1.21	3.8 ± 0.20
Sahupuram (Sub soil brine)	8.7 ± 0.53	24.6 ± 1.68	7.40 ± 6.18	9.46 ± 8.88	3.4 ± 0.10

Table-3 shows the analysis of radioactivity in the salt product. The radioactivity in the salt produced from the sea brine is higher than that of sub-soil brine. This is true in the case of ^{208}Tl, ^{228}Ac and ^{214}Bi. As the ^{208}Tl activity is not matching with ^{228}Ac activity, it can be concluded that disequilibrium exists between Th and Ra in all the cases. Hence ^{208}Tl activity can be considered that due to ^{232}Th and ^{228}Ac can be representative of ^{228}Ra, ^{214}Bi can be assumed to be due to ^{238}U. The salt sample produced from Sahupuram sub-soil brine has γ activity lower than that of Vepalodai salt work. The values are lower than Kanyakumari District sea brine using salt works because the salt work in kanyakumari District is naturally high background radiation area[10].

Basic safety series (BSS 115, IAEA) published the estimated ingestion dose to general public and radiation workers due to ingestion of various radionuclides. These values have been adopted for the estimation of ingestion dose to the general public. An average man requires about 5 g of NaCl per day, but generally consumes slightly more. Assuming a consumption of 5 g per day, the aggregate consumption per year works out to 2 kg. From the earlier report, it can be seen that the annual intake of Th, Ra and U is 26.66, 36.02 and 26.7 Bq, respectively. With the corresponding ingestion coefficients for the above radionuclides, the total ingestion dose works out to 13.61 μSv per year. The highest contribution (12.43 μSv) coming from ^{228}Ra[10].

Conclusion

The sea brine and subsoil brine used for the manufacture of common salt contain several chemical and radioactive elements. The activity of the sea brine using salt work was slightly higher than the subsoil brine using salt work. It was observed that at vepalodai, the gross α activity in the brine varied from 0.0009 to 0.0033 Bq L^{-1} and the gross β activity varied from 0.0194 to 4.0405 Bq L^{-1}. At the subsoil brine of Sahupuram, the corresponding values varied from below detection limit (BDL) to 0.0032 and 0.0114 to 3.0352 Bq L^{-1}. ^{40}K activity was found to increase, source to bittern, *viz.*, 4.006 to 221.006 Bq L^{-1} at Veplodai and 3.247 to 204.462 Bq L^{-1} at Sahupuram. The sediments collected at different stages showed gross α activity varying from 42.0 to 362.0 Bq kg^{-1}, gross β activity varying from 1998 to 2498 Bq kg^{-1}. ^{208}Tl, ^{228}Ac activities varied from 111.2 to 151.2 Bq kg^{-1} and 40.1 to 73.4 Bq kg^{-1}. The corresponding values for Sahupuram salt works were 10.2

to 98.7, 1298 to 1548, 137.4to175.8 and 33.8 to 55.3 Bq kg^{-1}, for gross α, gross β, ^{208}Tl, ^{228}Ac, respectively. The analysis of the common salt from Vepalodai showed 14.4, 107, 12.42, 13.10 and 3.8 Bq kg^{-1} of gross α, gross β, ^{208}Tl, ^{228}Ac and ^{214}Bi, respectively and that from sub soil brine of Sahupuram showed 8.7, 24.6, 7.4, 9.46 and 3.4 Bq kg^{-1}. An average man requires 5 g of NaCl per day, assuming that the total ingestion dose works out to 13.61 μSv per year. The highest contribution (12.43 μSv) coming from ^{228}Ra. This study shows that the radioactivity content in common salt is within the limit. However, the associated levels are not detrimental to health.

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