

Comparative Study on Radio Activity Associated with Common Salt Production Process of Two Salt Works in Kanya Kumari and Tuticorin Districts, Tamil Nadu

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Abstract - The manufacture of Common Salt used for the sea brine contain several chemical and radioactive elements. This study estimated the gross alpha and gross beta activities from the sea brine of two salt works at Kovalam in Kanya Kumari District and Vepalodai in Tuticorin District, Tamil Nadu. The gross activity was increasing at subsequent stages of production due to the concentration of brine. The sediment and gypsum samples collected at different stages were also subjected for estimation of different types of activity. ⁴⁰K activity was found to increase, source to bittern from 8.60 ± 0.92 to 221.006 ± 21.20 Bq l⁻¹ at Kovalam salt work and 4.006 ± 0.52 to 162.16 ± 15.94 Bq l⁻¹ at Vepalodai salt work. This study showed that the radio activity is higher in Kovalam salt work than Vepalodai salt work, indicating the influence of naturally high background radiation area.

Keywords: Alpha, Beta, Gamma activity, salt works, Sea brine

I. INTRODUCTION

Common salt (NaCl) plays critical roles in a number of life sustaining processes (M.E. Harper *et al.*, 1997). Of the world's annual production 6% is directly used for human consumption, the rest being consumed mainly by chemical industries (F.O. Wood, 1975). It has more than 14,000 ways of application either directly or indirectly (Salt Research and Industry, 1967). In temperate climate, the annual human requirement of salt is about 5kg/year and it is more in the tropical countries (D.W. Kaufmann, 1960). Manufacture of common salt from sea water or natural brine using solar energy and wind is a popular process. The brine is pumped to large reservoirs of 3-4 m depth 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 (J.P. Riley *et al.*, 1965). The original volume of the water reduces to 3%, when bittern (the supernatant liquid obtained after the precipitation of NaCl) is formed. The brine density expressed as salinity Baume (⁰Be).

The phase chemistry of solar salt production is conveniently divided into four distinct phases (H.W. Fiedelman *et al.*, 1969). The first phase is from 3 to 13⁰Be, when most of the carbonates of calcium, magnesium and iron precipitate. The second phase extending from 13 to 25.4⁰Be centres around gypsum, CaSO₄.2H₂O, which crystallizes as needle shaped

crystals from 13 to 16.4⁰Be and thereafter as anhydrous CaSO₄, 85% of CaSO₄ present is precipitated in this phase. The third phase extends between 25.6 and 30⁰Be, in this phase common salt, is precipitated out. The crystallization starts at 25.6⁰Be and its rate rapidly increases at the initial stages. 72% of the total amount is precipitated by 29⁰Be and 79% by 30⁰Be (Febvre, 1962).

Human population has always been exposed to natural radiation both externally and internally, former through terrestrial and cosmic origin and latter from radio nuclides deposited within the body. Naturally occurring radio nuclides of ²³⁸U decay series are significant contribution of radiation dose for members of the public (UNSCEAR, 1993). Three types of radiations, alpha, beta and gamma are emitted by different radioactive materials, which differ in their energy and penetrating power.

One was exposed to radiation only from natural sources until recent times, when the growth of nuclear energy has created other sources of exposure like fallout from weapon tests, radioactive releases from nuclear reactor operations and accidents, exposure due to radioactive waste disposals and industrial, medical and agricultural use of radioisotopes. Still, the major contribution to the average annual background radiation arises from natural sources. Potassium, another natural radio nuclide 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 0.20m Gy/y of radiation dose due to ⁴⁰K present in our body (S.C. Agarwal, 1956).

II. STUDY AREA

The salt work1(Kovalam) is located at a distance of 20km from Nagercoil and just 5km in the west of Kanya Kumari, where sea brine from Arabian Sea is used for the production. The salt work2 (Vepalodai) is located at distance of 40km from the Tuticorin where Bay of Bengal sea brine is used. The total area of the salt work1 is 28ha, salt work 2 is 750ha.

III. MATERIALS AND METHODS

Brine, sediments, bittern formed at different stages of manufacture of common salt and salt, gypsum samples were collected on a monthly basis for one year. The radioactivity of the brine samples was calculated after the precipitation by BaCl₂ and CaSO₄. The composite samples were dried in an electrical air oven at 120^o C, Powered in an agate mortar and suitable amount of substances were counted for alpha and beta activity. Gross alpha and gross beta activities were determined using low- background alpha counting system ZnS (Ag), ECIL, Modal RCS-4027, and gas flow beta counting system (ECIL Modal Bcs 36A). The efficiency of the alpha counter was determined using ²³⁹ Pu source of strength 542 dpm (distegration per minute) and the efficiency was estimated to be 30%. KCl with beta activity of 1000 Bq was used for estimating beta efficiency, which was calculated to be 40%. For the estimation of gamma efficiency, pure monazite sources of known strengths were used and the daughter products such as ²⁰⁸Tl, ²²⁸Ac, ²¹⁴Bi 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 paks also gives an idea of equilibrium. Th and U activity in the common salt were determined by counting 500g of samples in MCA, with 2" x 2" NaI(Tl) as detector (S.Singraravelu, 1981). ²²⁸Ac (910KeV, 29%) represents ²²⁸Ra and the peak energy of 1460 KeV(11%) was considered for ⁴⁰K. The counting time varied from 5000 to 10,000 s for maximum accuracy possible.

IV. STATISTICAL ANALYSIS

When radioactivity, particularly of low levels is measured, it is essential to make an assessment on the accuracy of the result in terms of percent error and confidence level. Hence the result presented in this work also has 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 cps, 0.212 cps, 0.101 and 0.097 cps for ²⁰⁸Tl, ²¹⁴Bi, ⁴⁰K and ²²⁸Ac respectively. With mineral monazite and KCl as standard sources, the derived sensitivities for different energies of interest were calculated to be 8.78x10⁻⁴ cps Bq⁻¹, 7.6x10⁻³ cps Bq⁻¹, 1.46x10⁻³ cps Bq⁻¹ and 4.5 x10⁻³ cpsBq⁻¹ for ²⁰⁸Tl, ²¹⁴Bi, ⁴⁰K and ²²⁸Ac respectively. Thus for ²⁰⁸Tl, 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,

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

where r_g is the counting rate of the sample, t_g is the sample counting time in seconds, r_{bg} is 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 seconds. Hence, all the samples were counted for a rounded up period of 8000 seconds. 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 5000s and an efficiency of 30%, α activity, activity of 0.001 Bq is presented with 5% error at 2σ. Similarly β activity is also presented with 5% error at 2σ, as they are counted for 3000 s, in a low background (3 counts per min) beta counting system having high efficiency (40%) (Herman cember, 1996).

V. RESULTS AND DISCUSSION

TABLE I THE MEAN AND STANDARD DEVIATION VALUES OF THE RADIOACTIVITY ASSOCIATED WITH BRINE SAMPLES AT VARIOUS STAGES OF PRODUCTION OF SALT

Salt Works	Various Stages	Activity (Bq l ⁻¹)		⁴⁰ K Activity (Bq l ⁻¹)
		gross α	gross β	
SW1 (Sea brine) Kovalam	Source	0.0018 ± 0.00009	0.0291 ± 0.001	8.60 ± 0.92
	Reservoir	0.0022 ± 0.0001	0.0352 ± 0.002	12.45 ± 1.26
	Condenser	0.0029 ± 0.0001	0.0405 ± 0.002	24.340 ± 2.82
	Crystallizer	0.0033 ± 0.0002	0.0441 ± 0.002	68.18 ± 6.82
	Bittern	0.0037 ± 0.0002	4.4930 ± 0.224	221.006 ± 21.20
SW2 (Sea brine) Vepalodai	Source	0.0009 ± 0.00005	0.0194 ± 0.001	4.006 ± 0.52
	Reservoir	0.0018 ± 0.0003	0.0241 ± 0.001	11.876 ± 1.11
	Condenser	0.0024 ± 0.0001	0.0292 ± 0.002	18.38 ± 1.73
	Crystallizer	0.0029 ± 0.0002	0.0343 ± 0.001	67.304 ± 6.37
	Bittern	0.0033 ± 0.0001	4.0405 ± 0.200	162.16 ± 15.94

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

Salt Works	Various Stages	Activity (Bqkg ⁻¹)			
		gross α	gross β	²⁰⁸ Tl	²²⁸ Ac
SW1 (Sea brine) Kovalam	Source	44.0 ± 1.2	2005.0 ± 99.25	164.4 ± 17.00	59.8 ± 6.01
	Reservoir	97.0 ± 4.85	653.0 ± 32.65	73.3 ± 7.33	34.1 ± 3.22
	Condenser	158.0 ± 6.92	3148.5 ± 157.42	169.9 ± 18.01	172.5 ± 17.52
	Crystallizer	190.0 ± 9.50	1930.0 ± 95.50	137.7 ± 11.20	69.5 ± 6.89
	Bittern	383.0 ± 20.15	2683.0 ± 130.15	126.1 ± 12.52	93.1 ± 9.22
SW2 (Sea brine) Vepalodai	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	3032.0 ± 150.16	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

TABLE III THE MEAN AND STANDARD DEVIATION VALUES OF THE RADIOACTIVITY OF GYPSUM SAMPLE FORMED AT THE CONDENSERS

Salt Works	Radio activity (BqKg ⁻¹)				
	gross α	gross β	²⁰⁸ Tl	²²⁸ Ac	²¹⁴ Bi
SW1 (Sea brine) Kovalam	298 ± 14.25	5323 ± 265.30	202 ± 21.00	2242 ± 210.55	128 ± 12.92
SW2 (Sea brine) Vepalodai	214 ± 12.72	4823 ± 240.15	194 ± 23.00	1956 ± 200.01	118 ± 12.00

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

Salt Works	Radio activity (BqKg ⁻¹)				
	gross α	gross β	²⁰⁸ Tl	²²⁸ Ac	²¹⁴ Bi
SW1 (Sea brine) Kovalam	31.7 ± 1.78	110.0 ± 6.50	12.42 ± 1.20	14.40 ± 1.54	11.20 ± 1.12
SW2 (Sea brine) Vepalodai	14.4 ± 0.81	107.0 ± 5.35	10.6 ± 1.35	13.10 ± 1.21	3.8 ± 0.20

Table I provides the details of radio activity associated with the selected salt works of brine samples at various stages of common salt production (from source to bittern stage). In SW1 salt work, the mean gross α activity varied from 0.0018 ± 0.00009 to 0.0037 ± 0.0002 Bq⁻¹ from source to bittern. In SW2 salt work, the mean gross α activity varied from 0.0009 ± 0.00005 to 0.0033 ± 0.0001 Bq⁻¹ from source to bittern. In SW1 salt work, the mean gross β activity varied from 0.0291 ± 0.001 to 4.4930 ± 0.224 Bq⁻¹ from source to bittern. In SW2 salt work, the mean gross β activity varied from 0.0194 ± 0.001 to 4.0405 ± 0.200 Bq⁻¹ from source to bittern. Gross α and gross β activity are minimum at the brine samples. The activity gets increased slowly from source to bittern stage. In gross β the activity increased drastically in the bittern stage. The concentration of ⁴⁰K activity also steadily increases from source to bittern. In SW1, the mean potassium activity varied from 8.60 ± 0.92 to 221.006 ± 21.20 Bq⁻¹ from source to bittern. In SW2, the mean potassium activity varied from 4.006 ± 0.52 to 162.16 ± 15.94 Bq⁻¹ from source to bittern. The 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. The original volume of the brine is

reduced to 3% at bittern stage (H.W. Fiedelman *et al.*, 1969). The samples of SW1 showed higher α and β activity than SW2 in all stages, indicating that the natural high background radiation area on SW1 Salt work using Arabian sea brine.

The table II gives the radio activity associated with the selected salt works of sediments collected at various stages of salt production. The mean value of gross α and β activity increased from the source to bittern stage. In SW1 salt work, the mean gross α activity varied from 44.0 ± 1.2 to 383.0 ± 20.15 Bqkg⁻¹ from source to bittern. In SW2 salt work, the mean gross α activity varied from 42.0 ± 2.50 to 362.0 ± 18.11 Bqkg⁻¹ from source to bittern. Gross beta activity is several times higher than that of gross alpha. In SW1 salt work, the mean gross β activity varied from 2005.0 ± 99.25 to 2683.0 ± 130.15 Bqkg⁻¹ from source to bittern. In SW2 salt work, the mean gross β activity varied from 1998 ± 99.21 to 2498.0 ± 124.90 Bqkg⁻¹ from source to bittern. This disproportion of values from α activity indicates the presence of sources other than Th and U. The sediments collected at the condenser stage showed maximum β activity is 3148.5 ± 157.42 , 3032.0 ± 150.16 ,

Bqkg⁻¹ and can be attributed to the precipitation of gypsum, at this stage, at salinity above 19° Be. Gypsum (CaSO₄. 2H₂O) carries similar radioactive substances like Ra, Th etc. along with it, resulting in higher radioactivity. Here again the samples from SW1 showed higher activity than SW2. The limited available information in literature indicates the presence of monazite deposits on the coastal areas of Tamil Nadu (G.Victor Rajamanickam, 2000). It is evident in the present study.

²⁰⁸Tl and ²²⁸Ac normally used for representing Th activity in gamma spectrometric analysis. But here, it can be seen that there is clear disequilibrium between these two radio nuclides. In SW1 salt work, the mean ²⁰⁸Tl and ²²⁸Ac activity varied from 164.4 ± 17 to 126.1 ± 12.52 Bqkg⁻¹ and 59.8 ± 6.01 to 93.1 ± 9.22 Bqkg⁻¹ from source to bittern. In SW2 salt work, the mean ²⁰⁸Tl and ²²⁸Ac activity varied from 151.2 ± 16.12 to 111.4 ± 12.15 Bqkg⁻¹ and 40.1 ± 4.11 to 73.4 ± 6.79 Bqkg⁻¹ from source to bittern. This also indicates that ²²⁶Ra and ²²⁸Ra have separate sources of origin other than Th and U. The reported presence of radio ferrous rocks at the sea bed is a strong possibility. The higher activity of ²²⁸Ac at the condenser stage is 172.5 ± 17.52 and 152.6 ± 15.62 Bqkg⁻¹, an indication of higher precipitation of ²²⁸Ra at this stage, due to high salinity compared to source and reservoir stage. This is probably due to the behavior of radium in the respective liquid medium.

Table III gives the distribution of activity in gypsum collected from condenser stage of two salt works. In SW1 salt work, the mean radio activity of gross α and β, ²⁰⁸Tl, ²²⁸Ac and ²¹⁴Bi were 298 ± 14.25, 5323 ± 265.30, 202 ± 21.00, 2242 ± 210.55 and 128 ± 12.92 Bqkg⁻¹. In SW2 salt work, the mean radio activity of gross α and β, ²⁰⁸Tl, ²²⁸Ac and ²¹⁴Bi were 214 ± 12.72, 4823 ± 240.15, 194 ± 23.00, 1956 ± 200.01 and 118 ± 12.00 Bqkg⁻¹. The radioactivity associated with SW1 sample is higher than that of SW2. The predominance of ²²⁸Ac in the gypsum sample shows the overwhelming presence of ²²⁸Ra in the sample due to its chemical affinity with calcium. The ratio of gross α and β activity normally observed in Th and U sources gets distorted in the present case. This can be attributed to the presence of other beta emitters, mainly radium originating from radio ferrous rocks. ²⁰⁸Tl and ²¹⁴Bi activity, which can be representative of Th and U, indicates nearly equal presence in the gypsum. An analysis of gross β and ²²⁸Ac activity indicates the presence of ²²⁸Ra also.

Table IV shows the analysis of radioactivity in the salt sample. In SW1 salt work, the mean radio activity of gross α, β, ²⁰⁸Tl, ²²⁸Ac and ²¹⁴Bi were 31.7 ± 1.78, 110.0 ± 6.50, 12.42 ± 1.20, 14.40 ± 1.54 and 11.20 ± 1.12 Bqkg⁻¹. In SW2 salt work, the mean radio activity of gross α, β, ²⁰⁸Tl, ²²⁸Ac and ²¹⁴Bi were 14.4 ± 0.81, 107 ± 5.35, 10.6 ± 1.35, 13.10 ± 1.21 and 3.8 ± 0.20 Bqkg⁻¹. Here also the gross alpha and

beta activity in the SW1 salt work is higher than SW2. This is true in the case of ²⁰⁸Tl, ²²⁸Ac and ²¹⁴Bi. The salt sample produced from SW1 salt work gamma activity is higher than that of SW2 salt work, indicating the influence of high background radiation area.

VI. CONCLUSION

An average man requires about 5 gram 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 be about 2 Kg. The annual intake of Th, Ra and U is 26.66 Bq, 36.02 Bq and 26.70 Bq respectively. With the corresponding ingestion coefficients for the above radio nuclides, the total ingestion dose works out to 13.61 μSv per year (C.G.Maniyan *et al.*, 2007). The highest contribution (12.43 μSv⁻¹) coming from ²²⁸Ra. This study showed that the radio activity in common salt is within the limit in both salt work sample. In comparing two salt work samples, the radio activity is less in SW2 (Vepalodai) salt work using Bay Of Bengal sea brine than SW1(Kovalam) salt work using Arabian sea brine.

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