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Variations in the gross alpha and beta activity in surface waters at the Atomic Weapons Establishment Aldermaston (UK)

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Abstract Statistical analysis has been performed on the gross alpha- and beta-activity measurements of surface waters collected at the Atomic Weapons Establishment at Aldermaston (UK) during the period January 2002–September 2005. The results have been found to follow a lognormal distribution and this has important applications when considering gross activity exemption thresholds. This implies that the gross activity is the multiplicative product of many small independent factors, such as meteorology, flow conditions and site operations. The influence of meteorological parameters has been investigated using linear regression, and some correlation has been identified between gross beta-activity and parameters indicative of fine weather. Variations in gross activity have been considered on monthly, weekly and daily timescales and characterised using the geometric mean and geometric standard deviation in accordance with the properties of the lognormal probability density function.

Keywords Gross activity · Surface waters · Radioactivity

Introduction

The radioactivity of surface waters is dominated by naturally occurring radionuclides (NOR) from the uranium (^{238}U) and thorium (^{232}Th) decay series together with a contribution from the primordial radionuclide ^{40}K. The concentrations of these alpha- and beta- emitters show considerable variation in the environment [1] and this has important implications for environmental monitoring programmes for anthropogenic radionuclides [2–4]. At the Atomic Weapons Establishment (AWE) at Aldermaston (UK), surface water samples that are collected on site are screened for isotopes of uranium and plutonium (^{238}U, ^{235}U, ^{234}U, ^{238}Pu, ^{239}Pu and ^{240}Pu) by means of a gross activity evaporation technique. The purpose of this monitoring is to demonstrate that site emissions, whether planned or inadvertent, have an insignificant impact on the surrounding environment, and that any possible radiation dose to either AWE employees or the public is negligible compared to that from natural background radiation. This demands an understanding of the variations in gross activity in order to assess the current activity thresholds of 40 Bq m^{-3} for alpha-activity and 1,000 Bq m^{-3} for beta-activity [5, 6]. Above these limits, samples must be either recounted until the activity decreases or analysed further by radiochemical techniques. Both options delay the sentencing of waters collected on site and occupy additional laboratory resources.

Methodology

Reagents and equipment

All reagents used were analytical grade. All reagents and equipment were supplied by Fisher Scientific, Loughborough, UK.

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Sample collection and preparation

Surface water samples (2L) were collected daily from a drainage facility at AWE Aldermaston during January 2002–September 2005 and analysed for gross activity. The samples were collected from an inlet to the drainage facility at 0700, 1100 and 1900 h and from a holding tank (HT) before discharge. Samples were prepared for counting by taking a 125 ml aliquot of the collected water sample and adding this to a borosilicate glass beaker. The water sample was vigorously shaken before this fraction was taken, and the counting source prepared in triplicate when taken from a holding tank sample. A few drops of methyl orange indicator solution were added and 1 M nitric acid added dropwise until the pH was 4–6. The sample was then evaporated on a hot plate to near dryness and care was taken to avoid baking solids. This was then transferred to a tared stainless steel counting pan ($\phi = 47$ mm) with the aid of a rubber stopper and distilled water from a wash bottle. Using the rubber stopper, the walls of the beaker were thoroughly wetted with a few drops of 1 M nitric acid and the washings transferred to the counting pan. The pan was then placed under an infra red lamp until completely dry. Once dry, the sample was cooled in a desiccator, weighed, and counted promptly for gross alpha- and beta-activity.

Gross activity counting

Measurements of gross activity were performed using a gas flow proportional counter (GFPC) of the low background multiple detector type with 10 sample detectors (Berthold LB770). The instrument is able to discriminate between alpha- (4–10 MeV) and beta-radiations (>0.2 MeV), although no resolution of the energy spectrum is recorded. Each sample was counted for 12 h to provide an average detection limit of 11.1 Bq m^{-3} for gross alpha-activity and 30.0 Bq m^{-3} for gross beta-activity. All calculations were made using the appropriate density thickness corrections for efficiencies to convert the gross alpha-activity (based on ^{241}Am) and gross beta-activity (based on ^{90}Sr) measurements to specific activities in Bq m^{-3} with estimates of error at $\pm 2\sigma$.

Statistical analysis

Statistical analysis was undertaken on the measurements to characterise their distributions, and in-depth analysis undertaken on measurements recorded during July 2002–July 2003. This time interval was chosen as meteorological data was available from a weather station situated close to the sampling location. This station recorded a range of standard parameters, including measurements of air temperature and pressure, wind speed and direction, incoming and outgoing radiation, soil conditions, relative humidity and rainfall. During this period, gross activity measurements and meteorological conditions were converted into daily, weekly and monthly measurements and correlations investigated using a linear regression model.

Results

Statistical analysis

The measurements of gross alpha- and beta-activity recorded during January 2002–September 2005 were statistically analysed to provide information on their distributions (Table 1). The arithmetic mean (AM) ranged from 30.6 to 35.9 Bq m^{-3} for gross alpha-activity and from 168.9 to 183.1 Bq m^{-3} for gross beta-activity. Similarly that of the geometric mean (GM) ranged from 28.0 to 32.6 Bq m^{-3} and 162.8 to 172.4 Bq m^{-3}. Within the confines of measurement uncertainty (typically 38% for

Table 1 Gross activity statistical parameters

Activity	Parameter	0700 h	1100 h	1900 h	Holding tank
Gross alpha	AM	34.0 ± 12.2	35.9 ± 12.4	30.6 ± 12.0	31.8 ± 11.8
	GM	31.1 ± 12.1	32.6 ± 12.3	28.0 ± 11.9	28.8 ± 11.7
	Min	3.3 ± 11.0	7.4 ± 7.4	2.2 ± 8.6	1.9 ± 9.4
	Max	197.9 ± 23.3	216.3 ± 28.0	189.3 ± 22.0	239.7 ± 24.5
	N	1,148	1,143	1,117	2,335
Gross beta	AM	178.7 ± 29.4	178.2 ± 29.9	183.1 ± 30.8	168.9 ± 29.6
	GM	172.3 ± 29.3	172.3 ± 29.8	172.4 ± 30.6	162.8 ± 29.4
	Min	28.1 ± 32.0	33.4 ± 42.0	48.4 ± 27.9	22.4 ± 41.8
	Max	723.6 ± 43.4	523.1 ± 41.8	1,967.5 ± 64.4	764.4 ± 45.7
	N	1,194	1,186	1,189	2,466

Values are given in Bq m^{-3}

alpha- and 17% for beta-activity) these variations were not significant at different collection times or locations. The minimum and maximum values of gross alpha-activity activity were also similar between collection times and locations, with values ranging from 2.2 to 239.7 Bq m^{-3}. For gross beta-activity measurements, minimum values were similar, although the maximum activity showed greater variation with values of 523.1 to 1,967.5 Bq m^{-3}. The number of samples analysed (N) ranged from 1,117 to 2,335 for alpha- and from 1,186 to 2,466 for beta-activity. This sample size was considered sufficient to be representative of the population of gross activity measurements.

The frequency distribution of gross alpha-activity measurement showed a highly skewed distribution with an asymmetrical tail that extended towards higher activity values (Fig. 1). Skewness values were calculated as 3.4 (0700), 2.7 (1100), 3.2 (1900) and 3.7 (HT). A more symmetrical distribution was evident for gross beta-activity measurements, with skewness values of 2.8 (0700), 1.8 (1100), 9.2 (1900) and 2.5 (HT). The value of 9.2 was offset by a singularly high gross beta-activity measurement of 1,967.5 ± 64.4 Bq m^{-3}. Both distributions had a high level of positive kurtosis, or peakedness compared with the normal distribution, with values of 23.9 (0700), 17.4 (1100), 21.6 (1900) and 29.8 (HT) for alpha-activity and 21.4 (0700), 8.8 (1100), 122.6 (1900) and 18.6 (HT) for beta-activity. Kurtosis measurements of alpha-activity were generally higher, and the beta-activity kurtosis value of 122.6 was again offset by the anomalous measurement.

Probability plots and the Anderson–Darling goodness-of-fit-statistic were used to evaluate the optimum distribution of the gross activity results [7]. Out of a possible 14 distributions that were examined, the gross alpha- and beta-activity measurements were found to follow a lognormal distribution for a significance level of 0.05 (Eq. 1). The gross alpha-activity was found to most strongly follow this type of distribution, as indicated by a lower Anderson–Darling goodness-of-fit-statistic. This hypothesis was confirmed using the least squares estimation method, and comparing how closely the gross alpha- and beta-activity measurements lay on the best-fit lines of a lognormal probability plot. By following this distribution, it implies that the gross activity is the multiplicative product of many small independent factors. This has been demonstrated using Monte Carlo simulation, such that when a variable (e.g. gross activity) is acquired through repeated experimentation under the influence of random, multiplicative perturbations, the distributed empirical outcomes tend to be well approximated by the lognormal probability density function (PDF) for a wide range of conditions [8, 9]. It is hypothesised that such perturbations might include variations in meteorology, flow conditions and site operations. Assuming this distribution, then it is appropriate to use the GM for characterising the average gross alpha- and beta-activity of the surface waters at AWE Aldermaston.

$$F[x, m, \sigma] = \frac{1}{\sqrt{2\pi\sigma}} \int_0^x \frac{1}{a} \mathrm{e}^{\left\{\frac{(\ln a - m)^2}{2\sigma^2}\right\}} \mathrm{d}a \qquad (1)$$

Equation 1. The probability density function of the lognormal distribution. M and σ are the parameters of the normal distribution of the logarithm of the measured gross activity. They have been estimated by the maximum likelihood method, where GM = e^m and GSD = e^{σ}.

Monthly variations

There were no significant changes or evidence of seasonal effects in the monthly GM concentrations during January 2002–September 2005 (Table 2). Values ranged from 28.2 to 35.4 Bq m^{-3} for gross alpha-activity and from 153.5 to 188.3 Bq m^{-3} for gross beta-activity. All values were

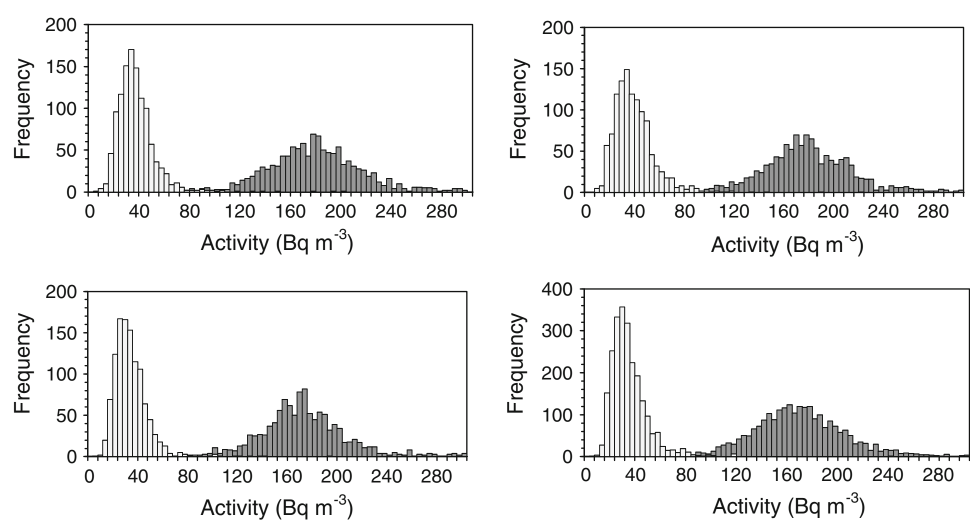

Fig. 1 Frequency histograms of gross alpha- (*light grey*) and beta- (*dark grey*) activity for surface water samples taken from the inlet channel at 0700 h (*top left*), 1100 h (*top right*), 1900 h (*bottom left*) and from the holding tank (*bottom right*)

Table 2 Mean monthly gross alpha- and beta-activity measurements averaged over all sampling years

Month	Gross alpha-activity (Bq m^{-3})			Gross beta-activity (Bq m^{-3})		
	GM	GSD	N	GM	GSD	N
Jan	29.8 ± 12.3	1.5	250	164.3 ± 29.5	1.3	250
Feb	33.3 ± 12.2	1.4	244	171.9 ± 29.5	1.2	244
Mar	30.0 ± 11.9	1.5	269	167.0 ± 29.4	1.3	263
Apr	31.7 ± 12.7	1.6	286	171.8 ± 29.9	1.3	292
May	28.5 ± 12.2	1.5	333	161.2 ± 29.5	1.3	348
Jun	28.2 ± 12.1	1.5	338	185.6 ± 30.2	1.4	341
Jul	30.1 ± 11.8	1.4	355	188.3 ± 30.2	1.4	358
Aug	30.6 ± 11.9	1.6	358	181.8 ± 30.3	1.3	352
Sep	30.8 ± 12.0	1.6	306	181.0 ± 30.3	1.3	299
Oct	35.4 ± 12.6	1.7	278	171.3 ± 30.6	1.4	267
Nov	30.6 ± 11.9	1.5	219	156.5 ± 30.0	1.3	206
Dec	29.0 ± 11.4	1.5	172	153.5 ± 29.7	1.5	167

similar within the confines of measurement uncertainty, although gross beta-activity measurements did appear slightly higher during June–October. The gross alpha-activity was not significantly correlated to the beta-activity. The geometric standard deviation (GSD) was highest for gross alpha-activity measurements with a value of 1.7 Bq m^{-3}, compared to a value of 1.4 Bq m^{-3} for gross beta-activity measurements, indicating a higher dispersion of measurements relative to the average value.

The influence of meteorological conditions on these gross activity values was investigated using a linear regression model for data collected during July 2002–July 2003. Linear regression techniques can be justified as the natural logarithm of a lognormal series of measurements is itself normally distributed [10]. Thus if a series of multiplicative factors form the basis of the lognormal distribution of gross activity measurements, it is reasonable that a series of additive disturbances form the basis of the natural logarithm of gross activity measurements and are therefore suitable for linear regression analysis [8, 11]. No significant correlations were found between gross alpha-activity and different meteorological variables with regression coefficient (r^2) values of 0.03–0.15. Some indications of a positive correlation were evident between gross beta-activity and indicators of fine weather, with r^2 values of 0.43 for air temperature and 0.39 for relative humidity. This may support the suggestion that gross beta-activity is highest during the summer months.

Weekly variations

The weekly GM varied from 19.5 to 45.5 Bq m^{-3} for gross alpha-activity and from 127.4 to 216.5 Bq m^{-3} for gross beta-activity during the period July 2002–July 2003 (Fig. 2). The correlation between gross alpha-activity and gross beta-activity appeared more evident with an r^2 value of 0.28 which was significant at the 0.01 probability level. The GSD was 1.2 Bq m^{-3} for both gross alpha-activity and gross beta-activity and was similar to that calculated for monthly values. Seasonal variations in gross alpha-activity were not apparent, and the influence of meteorological parameters was not significant. Weekly gross beta-activity measurements mimicked those on a monthly scale, with values generally highest during the summer months. This was again supported by regression analysis with recorded meteorological data, with r^2 values of 0.23–0.30 for indicators of fine weather.

Daily variations

Daily measurements ranged from 11.3 to 65.5 Bq m^{-3} for gross alpha-activity and from 88.0 to 377.8 Bq m^{-3} for gross beta-activity during July 2002 to July 2003 (Fig. 3). Gross alpha-activity continued to be correlated to gross beta-activity with an r^2 value of 0.29 which was significant at the 0.01 probability level. Variations within the results

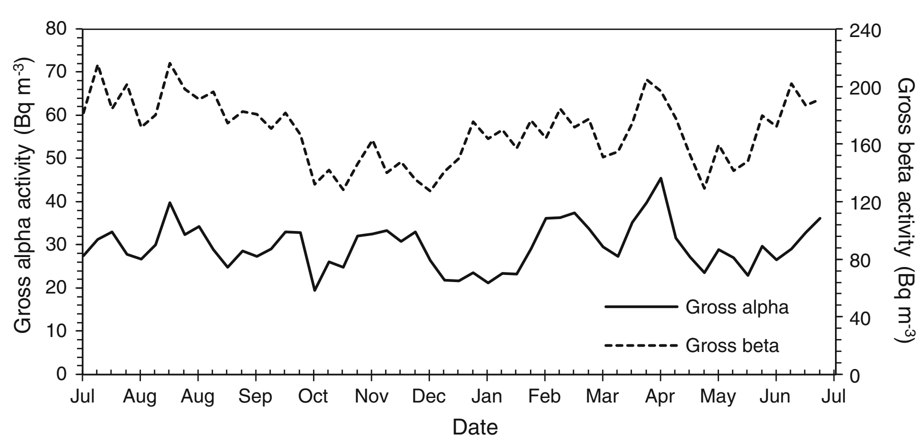

Fig. 2 Mean weekly variations of gross alpha- and beta-activity during July 2002–July 2003

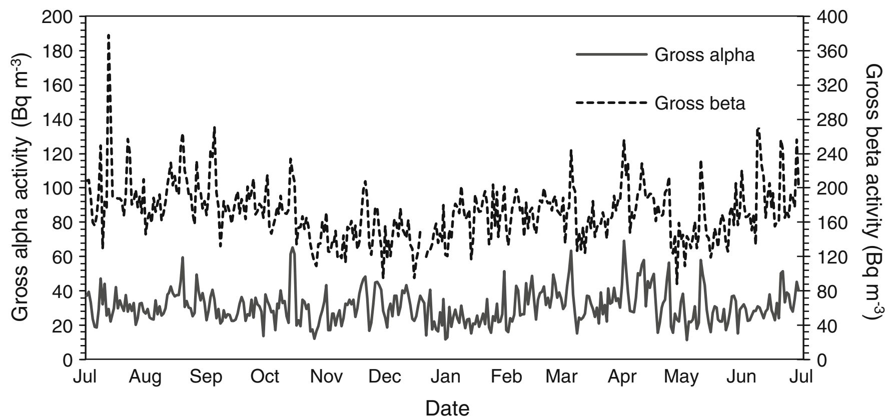

Fig. 3 Mean daily variations of gross alpha- and beta-activity during July 2002–July 2003

were highest for gross alpha-activity measurements with a GSD of 1.4 Bq m^{-3} compared to that of 1.2 Bq m^{-3} for gross beta-activity measurements. However, unlike for weekly averaged measurements, daily measurements were not well correlated to daily weather conditions, with r^2 values for each meteorological parameter below 0.03 for gross alpha-activity and below 0.11 for gross beta-activity.

Discussion

Consideration of exemption thresholds

The GM for gross alpha-activity is comparable to the value of 36.1 Bq m^{-3} reported as the typical global activity of lakes and rivers [12]. This is despite there being considerable variability in gross activity depending on the local geology and mineral types. Calculation of the GM for the entire dataset (inlet and HT) gives a value of 30.5 ± 12.1 Bq m^{-3} for gross alpha-activity and 172.3 ± 29.9 Bq m^{-3} for gross beta-activity. Both of these values are below the 40 Bq m^{-3} (alpha) and 1,000 Bq m^{-3} (beta) exemption thresholds of AWE, in addition to the 100 Bq m^{-3} (alpha) and 1,000 Bq m^{-3} (beta) thresholds recommended by the World Health Organisation (WHO) and guidelines of the EC Drinking Water Directive (98/83/EC) [13]. It is of additional importance to note, that all samples with a gross alpha-activity exceeding the 40 Bq m^{-3} value, have decayed to below this threshold since automatic recounting of samples was introduced in 1998. Any gross beta-activity values exceeding the 1,000 Bq m^{-3} threshold have also been found to be attributable to natural ^{40}K.

Predictions based on the lognormal distribution

It has been demonstrated from earlier statistical analysis that the distribution of gross activity measurements can be approximated by the lognormal PDF (Eq. 1). This concept is additionally supported by the Principle of Maximum Entropy and the arguments of Bayesian statistics. This suggests that the distribution is considered suitable under circumstances of a positive random variable, and when information on the mean and standard deviation (but no other data) is available [8, 14–18]. Under these assumptions we can therefore predict the probability and

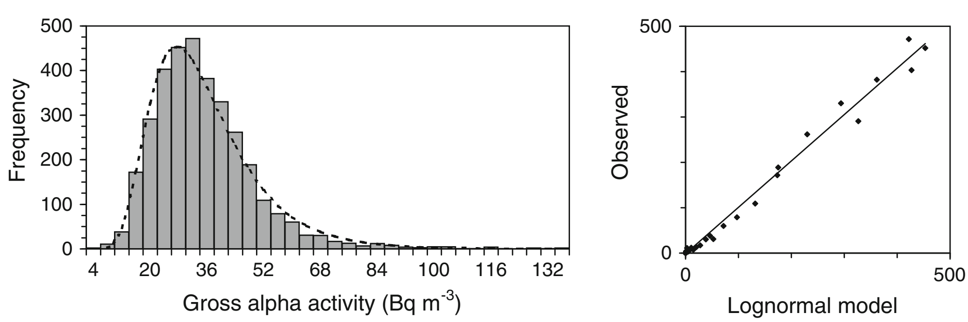

Fig. 4 Comparisons between measured gross alpha-activity (*grey* histogram) and the predicted lognormal PDF (*dashed line*) for all measurements taken during January 2002–September 2005. The intervals of the histogram (bins, given in Bq m^{-3}) are 0–4, 4–8, 8–12, etc. The correlation diagram (*right*) compares the number of measurements predicted by the lognormal PDF and the number observed at different gross alpha-activity bins

occurrences of gross activity measurements using the lognormal PDF. This has been demonstrated for measurements of gross alpha-activity during January 2002–September 2005 (Fig. 4). The correlation between the lognormal model (GM = 30.5, GSD = 1.5 Bq m^{-3}) and the observed measurements was very high with an r^2 value of 0.99. An ANOVA test was used to compare the data, and this showed a significant correlation at a probability level of 2.2×10^{-49}. Thus the lognormal model provides a very good fit to the measured gross alpha-activity. However, the lognormal PDF does appear to underestimate the occurrence of higher gross alpha-activity measurements within the extremities of the model, predicting a probability of 0.003 for measurements above 100 Bq m^{-3}. Based on the number of samples analysed during January 2002–September 2005, this equates to approximately 10 measurements (0.28% of total) although 26 were actually observed (0.76% of total). Nevertheless, this indicates that an inherent variation of measurements should be expected and serves as a first approximation to their distribution characteristics.

Conclusions

Statistical analysis and theoretical considerations indicate that the gross alpha- and beta-activity of surface waters at AWE Aldermaston follow a lognormal distribution. This implies that the gross activity is the multiplicative product of many small independent factors, such as meteorology, flow conditions and site operations. In accordance with this distribution, the GM has been calculated as 30.5 ± 12.1 Bq m^{-3} for gross alpha-activity and as 172.3 ± 29.9 Bq m^{-3} for gross beta-activity. Linear regression indicates a positive correlation between gross beta-activity and fine weather over monthly and weekly timescales. Gross alpha- and beta-activity measurements are not strongly correlated, and this may be indicative of variations in their NOR composition, particularly the contribution of ^{40}K to gross beta-activity.

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