Journal of Environmental Radioactivity 138 (2014) 289–301

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

A statistical approach to investigating enhancement of polonium-210 in the Eastern Irish Sea arising from discharges from a former phosphate processing plant☆

Alastair Dewar [a, *], William Camplin [a], Jon Barry [a], Paul Kennedy [b]

[a] Centre for Environment, Fisheries and Aquaculture Science, Lowestoft NR33 0HT, United Kingdom
[b] Food Standards Agency, London WC2B 6NH, United Kingdom

ARTICLE INFO

Article history:
Received 3 April 2014
Received in revised form
21 August 2014
Accepted 22 August 2014
Available online

Keywords:
Polonium
Natural radionuclides
Dose assessments
Statistical testing
Irish Sea

ABSTRACT

Since the cessation of phosphoric acid production (in 1992) and subsequent closure and decommissioning (2004) of the Rhodia Consumer Specialties Limited plant in Whitehaven, the concentration levels of polonium-210 (^{210}Po) in local marine materials have declined towards a level more typical of natural background. However, enhanced concentrations of ^{210}Po and lead-210 (^{210}Pb), due to this historic industrial activity (plant discharges and ingrowth of ^{210}Po from ^{210}Pb), have been observed in fish and shellfish samples collected from this area over the last 20 years. The results of this monitoring, and assessments of the dose from these radionuclides, to high-rate aquatic food consumers are published annually in the Radioactivity in Food and the Environment (RIFE) report series. The RIFE assessment uses a simple approach to determine whether and by how much activity is enhanced above the normal background.

As a potential tool to improve the assessment of enhanced concentrations of ^{210}Po in routine dose assessments, a formal statistical test, where the null hypothesis is that the Whitehaven area is contaminated with ^{210}Po, was applied to sample data. This statistical, modified "green", test has been used in assessments of chemicals by the OSPAR commission. It involves comparison of the reported environmental concentrations of ^{210}Po in a given aquatic species against its corresponding Background Assessment Concentration (BAC), which is based upon environmental samples collected from regions assumed to be not enhanced by industrial sources of ^{210}Po, over the period for which regular monitoring data are available (1990–2010). Unlike RIFE, these BAC values take account of the variability of the natural background level. As an example, for 2010 data, crab, lobster, mussels and winkles passed the modified "green" test (i.e. the null hypothesis is rejected) and as such are deemed not to be enhanced. Since the cessation of phosphoric acid production in 1992, the modified "green" test pass rate for crustaceans is ~53% and ~64% for molluscs. Results of dose calculations are made (i) using the RIFE approach and (ii) with the application of the modified "green" test, where samples passing the modified "green" test are assumed to have background levels and hence zero enhancement of ^{210}Po. Applying the modified "green" test reduces the dose on average by 44% over the period of this study (1990–2010).

1. Introduction

The radiological significance of naturally occurring radionuclides in marine foodstuffs, particularly polonium-210 (^{210}Po, $t_{1/2} = 138$ days), has been recognised since the 1980s (McDonald et al., 1986; Pentreath et al., 1989). In the eastern Irish Sea, it has been shown in a number of studies that the concentrations of many naturally occurring radionuclides were enhanced above levels observed in environmental samples from further afield (McCartney et al., 1990, 1992; McDonald et al., 1991; Rollo et al., 1992). This was due to the discharge of liquid waste, containing technologically enhanced concentrations of naturally-occurring radionuclides, at Saltom Bay, near Whitehaven. These discharges were from the Rhodia Consumer Specialties Limited (formerly Albright and Wilson) phosphoric acid production plant, over the period 1954–2004 and authorised under the Radioactive Substances Acts 1960, 1993.

☆ This has been presented in poster form at the 14th International Conference on the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere, Brighton 2013.

* Corresponding author. Tel.: +44 1502521346.
E-mail address: Alastair.Dewar@Cefas.co.uk (A. Dewar).

http://dx.doi.org/10.1016/j.jenvrad.2014.08.016
0265-931X/

The discharges resulted in significantly enhanced concentrations of naturally occurring radionuclides in the local shellfish and elevated dose to the local seafood consumers (Rollo et al., 1992; Camplin et al., 1996; Pollard et al., 1998). Since 1992, concentrations of naturally-occurring radionuclides in the local environment have decreased concomitant with changes in discharge practices, with a change in raw materials from phosphate ore to crude phosphoric acid and the commissioning of a raffinate treatment plant, as described in Poole et al. (1995). Polonium-210 currently observed in the eastern Irish Sea is a result of natural sources and legacy of the effects of discharges from the phosphate production plant.

The results of regular monitoring, to determine the concentrations of ^{210}Po (and other naturally occurring radionuclides, such as lead-210 (^{210}Pb), thorium and uranium isotopes) in environmental samples near Whitehaven, and the resultant dose from the consumption of foodstuffs containing naturally occurring radionuclides, are reported annually (see Environment Agency et al., 2012 and predecessor reports). As levels have reduced, it has become more difficult to identify the small enhancements of naturally occurring radionuclides (from historical discharges) against the range of concentrations normally expected from naturally sourced radioactivity (background) due to the extent of environmental fluctuations. In this study, the aim was to assess whether there is enhanced ^{210}Po contamination following the reduction in discharges to the eastern Irish Sea, by means of a statistical test. This test has compared the observed ^{210}Po concentrations in the Whitehaven area to Background Assessment Concentrations (BACs) and takes account of the natural variability of test and background samples. These BACs were calculated using ^{210}Po data remote from the eastern Irish Sea presented in Young et al. (2002), which collected and analysed seafood samples from around the United Kingdom. Retrospective dose assessments were performed to determine the potential implications of the results of the statistical test upon dose.

2. Methods and sources of data

2.1. Statistical testing

This study used the modified "green" test outlined by the OSPAR Working Group on Statistical Aspects of Environmental Monitoring (WGSAEM) (OSPAR, 2008). This test was previously used to study the "close to zero" problem (where it is difficult to distinguish concentration levels of man-made substances from zero concentrations or natural substances from their natural background levels) for toxic metals (Nicholson and Fryer, 2003) and other contaminants, such as total oxidized nitrogen and chlorophyll-A (Heffernan et al., 2010). OSPAR (2008) also describes two alternate tests; the unmodified "green" (i.e. in its non-modified form) and "brown" tests. These were not considered in this paper because they were regarded by the authors to be overly or insufficiently cautious.

The modified "green" test employed in this study is made more effective by the application of a screening level, which is above the background level. Samples, where the mean concentration is between the screening and background levels can be identified as being not enhanced. In the modified "green" test the null hypothesis is that the mean concentrations of ^{210}Po in seafood samples are enhanced above the BAC level and the data from the test area are used to confirm or reject this. The modified "green" test is appropriate to ^{210}Po enhancement as it includes the precautionary principle, where measures are taken to prevent harmful consequences of industrial processes. This is advocated by OSPAR as the null hypothesis is that the test samples will fail and the data have to show otherwise. This is different to many standard hypothesis tests in, for example, epidemiology where the null hypothesis might be that disease levels near and far from a source of pollution are the same – and the data has to demonstrate any difference. Further details of the modified "green" test can be found in Fryer (2004a,b) and OSPAR (2008). Formally, the modified "green" test is of the form:

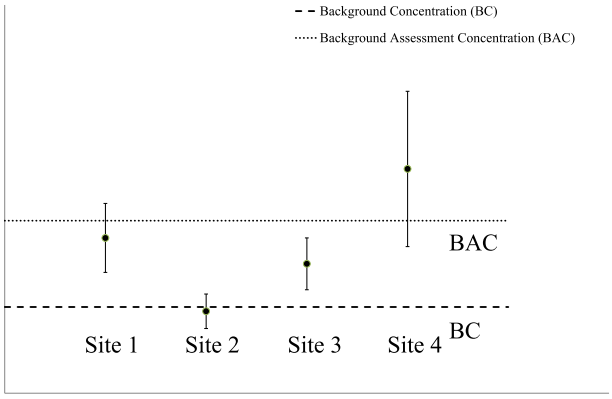

Fig. 1. Stylised explanation of the modified "green" Test with Upper Confidence Limits for the mean value at four hypothetical sites. The second and third sites pass the modified "green" test, i.e. they are regarded as being not enhanced, that is the Upper Confidence Limit is less than the Background Assessment Concentration.

$$H_0: \quad \mu \geq \text{BAC} \tag{1}$$

$$H_1: \quad \mu < \text{BAC} \tag{2}$$

where H_0 is the null hypothesis, H_1 is the alternate hypothesis and μ is the arithmetic mean concentration in the target environment. The environment is only declared "healthy" if the null hypothesis is rejected and as such is deemed a modified "green" test pass. Where the null hypothesis is not rejected, this is deemed a modified "green" test fail, i.e. enhancement of ^{210}Po exists. Fig. 1 shows a

Table 1
Consumption rates of edible fraction of fish, crustaceans and molluscs (kg y^{-1}) in the Whitehaven area.

Year (report)	Fish	Crustaceans			Molluscs	
	Cod	Crabs	Lobsters	Nephrops	Winkles	Mussels
1990 (AEMR 29)[a]	18.5	3.6	2.4	0.0	8.3	0.0
1991 (AEMR 34)	18.5	3.6	2.4	0.0	8.3	0.0
1992 (AEMR 38)	18.5	3.6	2.4	0.0	8.3	2.8
1993 (AEMR 42)	14.0	12.0	3.0	0.0	6.2	1.6
1994 (AEMR 45)	13.0	7.8	4.2	0.0	8.2	1.5
1995 (RIFE 1)[b]	13.0	6.5	2.2	0.0	6.0	6.0
1996 (RIFE 2)	12.5	7.2	4.8	0.0	7.2	4.8
1997 (RIFE 3)	27.8	8.5	6.8	1.7	1.7	2.5
1998 (RIFE 4)	22.5	23.8	4.2	0.0	4.5	10.5
1999 (RIFE 5)	21.5	19.2	4.8	0.0	12.5	12.5
2000 (RIFE 6)	12.4	8.0	8.0	4.0	8.5	8.5
2001 (RIFE 7)	12.4	8.0	8.0	4.0	8.5	8.5
2002 (RIFE 8)	20.4	8.0	4.8	3.2	17.4	11.6
2003 (RIFE 9)	24.6	21.6	2.7	2.7	13.6	20.4
2004 (RIFE 10)	24.6	12.5	10.0	2.5	17.0	17.0
2005 (RIFE 11)	24.6	12.0	4.0	4.0	19.8	13.2
2006 (RIFE 12)	24.6	12.0	4.0	4.0	20.0	20.0
2007 (RIFE 13)	24.6	10.2	6.1	4.1	17.3	11.6
2008 (RIFE 14)	10.0	11.8	3.4	1.7	15.7	15.7
2009 (RIFE 15)	10.0	4.8	8	3.2	16.8	11.2
2010 (RIFE 16)	10.0	11.0	6.6	4.4	4.4	17.6
Mean	18.0	10.3	4.9	1.9	11.0	9.4

[a] For example Camplin (1992).
[b] For example Ministry of Agriculture, Fisheries and Food (1996).