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Polonium and plutonium in an intertidal food chain

LARGE quantities of seaweeds (kelp) are washed ashore along the coasts of California. Estimates range from 10⁴ to 10⁶ tonnes $(10^7 - 10^9 \text{ kg})$ wet weight per yr. These algal masses provide food for insects and other invertebrates on the beaches. The plants carry with them a variety of di-, tri-, and polyvalent cations of heavy metals, largely adsorbed or chelated to superficial acidic polysaccharides and surface slimes of bacterial origin. Among the heavy metals one can detect small amounts of radioactive elements, including some that emit high-energy α particles such as the natural polonium (²¹⁰Po, half life 138 d) and the artificial plutonium nuclides $(^{239,240}$ Pu, half lives 24,360 yr and 6,580 yr respectively)¹. In surface waters of the oceans, polonium is born from the decay of ²¹⁰Pb which is continuously replenished by decay of radon released from the Earth's crust. The plutonium isotopes occur in ocean waters as a consequence of fallout from atomic test explosions some 15-20 yr ago. We have examined the extent to which these elements may be transferred from the ocean to the terrestrial biosphere by means of kelp, kelp flies and their predators. Fears have been expressed that radioactivity from the algae could be reaching man through the terrestrial food chain. Our investigations suggest that, on the basis of current radiation thresholds for human health and safety, such fears are groundless.

Samples of cast weed, largely comprising blades and stipes of Macrocystis pyrifera (L.) C.A. Ag. but containing also a few per cent of Egregia laevigata Setch. and Pelagophycus porra (Leman) Setch. and marine grasses (Phyllospadix and Zostera), were collected from the beach at La Jolla, California, in June and August 1975, at a time when kelp flies were much in evidence. Adult flies (mostly Fucilia rufitibia Stein and F. separata Stein) were caught over clumps of decaying weed by the use of a 1-mm mesh nylon net. About 30,000 flies, weighing 100 g wet weight (27 g dry weight) were collected in about 30 min. A few of the associated predatory tiger beetles (Cicindela sp.) were also netted, as individuals, on nearby sand. For comparative purposes we also obtained some 50 g (dry weight) of common houseflies (Musca domestica L) which had been raised in a laboratory (Department of Entomology, University of California, Riverside) on a commercial fly-larva medium (CSMA, Ralston-Purina Co.).

All samples were dried in an oven at 80 °C overnight. Polonium and plutonium were determined in subsamples of 1.0 g and 20.0 g respectively. The flies and beetles were 'wet-ashed' by heating in a mixture of nitric and perchloric acids. As markers, ²⁰⁸Po and ²⁴²Pu were added to monitor the chemical recoveries of ²¹⁰Po and ^{239,240}Pu from the samples. The α activities of these elements were measured with silicon surface-barrier detectors in combination with pulse-height analysers.

As shown in Table 1, the contents of polonium in laboratoryraised houseflies, and in the diet on which they were raised as larvae, were negligible (10 and 30 pCi per kg dry weight) compared with the kelp flies (250 pCi per kg dry weight). The polonium content of the kelp flies from the beach was found to be about half of that of the cast weed, on the basis of comparable dry weights. (That of laboratory-reared *Musca* was about one third that of their larval food.) This proportional reduction may be attributable to mechanical selection by the fly larvae of the softer, interior parts of the seaweeds, which have been shown to contain considerably less of the adsorbed heavy metals (including polonium) than the surface layers². Alternatively, there may have been discrimination against uptake of polyvalent metallic cations in the insect gut, selective metabolic processes in the tissues, or active excretion of such cations.

The polonium content in the predatory tiger beetles was found to be about one-half that of kelp flies, a fact which may be attributable to similar mechanical or metabolic discrimination. (It would be interesting to determine the polonium content of other predators of kelp flies, for example birds such as spotted sandpipers (*Petrochelidon pyrrhonota*) or swallows (*Hirundo rustica*), but we did not attempt this.) In neither of the two successive steps

Table 1 Polonium and plutonium contents of algae and flies from a California shore					
		²¹⁰ Po	^{239,240} Pu	Po/Pu	²¹⁰ Pb
Beach					
Kelp:	Macrocystis	480 ± 40	2.0 ± 0.3	250	380 ± 40
	and others	510 ± 25			
Flies:	Fucilia: whole adults	290 ± 15	1.1 ± 0.2	260	220 ± 30
		247 ± 18			
	hard parts	277 ± 13			
	expressed 'juice'	< 5			
Beetles	: Cicindela	164 ± 12			
Laborato	ry				
Fly-larva food (CSMA)		30 ± 3			
Flies: Musca: whole adults		10 ± 4			

Values are pCi per kg dry weight; shown as mean \pm s.e.m.

of the food chain studied here did we observe a relative enrichment of the radionuclides. On the contrary, at each step the relative polonium concentration was halved, while the plutonium (and ²¹⁰Pb) content of the kelp flies was also only half that of the content of the decayed kelp on which they fed.

A crude extract of the blood, gut contents and other soft parts of the kelp flies was prepared by homogenising a sample of the fresh flies in a Waring blender with distilled water for 45 s and centrifuging at 13,000 r.p.m. (20,000g) for 10 min. The liquid fraction was then clarified by filtration. The residues retained by the filter and at the bottom of the centrifuge tube consisted of cuticles, heads, wings, legs and bristles. This fraction contained essentially all of the polonium of the intact flies (see Table 1), indicating that most of the polonium ingested by the insect larvae with their food (the decaying kelp) had been incorporated in the cuticles of the adult flies.

In ascending the food chain from the kelp to the flies, the Po/Pu ratio remained unchanged. In contrast, corresponding values for *Macrocystis* plants and for bryozoa removed from their surfaces were respectively 230 and $1,000^{1}$, while even larger ratios, as high as 10^{5} or 10^{6} , were found in the internal organs of several species of marine fishes (V.F.H., unpublished), suggesting that in these latter systems there is a more effective discrimination against Pu, or in favour of uptake of Po, or both, than in the kelp flies.

Our observations indicated that on a warm summer day there may be as many as 10^7 flies per km of beach near the Scripps Institution of Oceanography, in an area where the estimated annual tonnage of cast weed is about 10^5 kg km⁻¹ (ref. 3). The kelp brings with it some 10^7 pCi of polonium and 4×10^4 pCi of plutonium per km of beach annually. If we assume that there are 10 generations of kelp flies per yr (in summer⁴ a generation time of kelp flies may be as short as 10 d), it can be calculated that about 0.2% of the polonium and 0.2% of the plutonium adsorbed or in some other way fixed to the major seaweeds may find its way into a terrestrial food chain.

It should be emphasised that although the kelp flies may occur in large numbers along beaches on warm days, such as those observed during the great seaweed-fly plague in southern England in $1953-54^5$, the radiation values with which we are concerned here present no obvious hazards to human health. The accidental inhalation of a few kelp flies would involve the ingestion of merely 10^{-4} pCi of 210 Po and 10^{-6} pCi of 239,240 Pu, while the deliberate ingestion of thousands of such flies would be too involved and unappetising an exercise to consider seriously.

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Free-running activity rhythm in the natural environment

RHYTHMICAL activities of plants and animals in their natural habitats have, so far, always been found to be synchronised with cyclic environmental changes¹⁻³. In the absence of appropriate environmental timing cues (usually light-dark or tidal cycles) the rhythms free-run with endogenously controlled periods other than those of the environmental cycle. These may be longer or shorter than those observed in the presence of the entraining environmental cycles. We describe here an apparently unique example of an invertebrate which seems to free-run in its natural habitat. The animal is a predatory prostigmatid mite (Bdella interrupta Evans) which inhabits marine salt marshes and is exposed to regular tidal inundation during periods of spring tides. During periods of neap tides the salt marsh is not covered by the sea. The mites showed day and night peaks of locomotory activity on the soil surface at creek edges (Fig. 1a) and, also, in isolated experimental conditions (Fig. 1b). The narrow peaks of activity imply that the individual members of the population are closely synchronised with one another.

The peaks of locomotory activity in the natural habitat did not exhibit constant relationships to the times of dawn and dusk. The interval between day peaks of activity during a sequence of non-covering tides was less than 24 h. The slope of the regression line for the first 6 d illustrated in Fig. 2 corresponds to an average separation of 23.1 h between successive day peaks during this period of non-submerging tides. During



Fig. 1 a, Numbers of mites observed walking on the soil surface on the edges of a drainage channel on Hut Marsh, Scolt Head, Norfolk, UK. Observations were made on a 25-m length of bank during a period of tidal emergence (6-7 April 1976). The times shown by broken lines. b, Surface activity of B. interrupta in isolated habitat portions (in five cylindrical containers of 15.2 cm diameter containing soil of 10 cm depth). The mites were maintained in natural light conditions.

the subsequent period of submerging tides the average separation between successive day peaks increased to 25.1 h, so that the surface locomotory activity of the mites effectively paralleled the times of the high tides for the period (Fig. 2).

Periodograms for the 1976 data illustrated in Fig. 2 yielded activity peaks at 22.9 h (for locomotory activity during nonsubmerging tides) and 24.9 h (during the subsequent period of submerging tides) (Fig. 3) for data collected during the daylight hours. Field observations made in 1977 confirmed these differences in periodicity observed during periods of tidal emergence and submergence. Since the activity peaks occur twice



Fig. 2 The timing of day activity peaks related to the time (\bullet) and the height (O) of high tides from 6 to 18 April 1976. The horizontal bars indicate the period during which mite activity was more than 50% of the maximum. The vertical lines are the midpoints of the bars and are taken as the phase reference point. The open bars represent activity during tidal emergence and the closed bars that during a subsequent period of submerging spring tides. The continuous lines are the calculated regression lines (for the period of tidal emergence: r = 0.9720; n = 6; P < 0.005; for the period of tidal submergence r = 0.9923; n = 7; P < 0.001). For these calculations the day sequence was used as the x axis.

daily and the rhythm entrains to the tidal frequency when the animal is exposed to tidal inundation the locomotory activity rhythm can be defined as a circatidal rhythm with a periodicity of about 12.5 h. As with circatidal rhythms in other organisms, the mite rhythm is insensitive to light-dark cycles. In this respect this salt-marsh mite is similar to an oribatid mite (Ameronothrus marinus Banks), which on rocky shores shows an apparently circatidal rhythm of 12.3-h periodicity⁴.

The novel feature revealed by this investigation is that during periods of non-submerging tides the period of the activity rhythm (11.5 h) is shorter than when it is entrained by the tides (12.5 h) and can be reasonably presumed to be a free-running circatidal rhythm. The maintenance of narrow peaks of activity throughout periods of tidal emergence indicates that the periods of the individuals are very similar whilst free-running. This contrasts with the breakdown of synchrony seen in free-running populations of other organisms in constant experimental conditions.

There is no conclusive evidence that the alternation of an apparently free-running rhythm, during tidal emergence, with an entrained rhythm, during periods of tidal submergence, is of adaptive value. It is conceivable, however, that this alternation could be a mechanism to achieve synchrony with the pattern of tidal coverage on the marine salt marsh. At the onset of periods of tidal coverage the activity peaks occur approximately midway between the times of successive high tides. This temporal separation is maintained during subsequent periods of tidal coverage by the circatidal rhythm of 12.5-h periodicity. The alternation between 11.5- and 12.5-h periodicities ensures