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Seasonal Variation of Caesium-137 from Fallout in a Clam, *Rangia cuneata* Gray

In the northern temperate zones, fallout of long lived radionuclides, including caesium-137, varies seasonally, with a maximum during May–June and a minimum during November–December[1]. The concentration of caesium-137 in caribou flesh and in Alaskan Eskimos shows a similar seasonality[2]. Because of its half life of 30 yr and the huge stratospheric reservoir of radioactive debris, the amount of caesium-137 on the surface of the Earth is still increasing, despite the moratorium on atmospheric testing[3]. Although caesium is not a biologically important trace element, chemically it resembles potassium, which is the principal cation of cytoplasm. The bioaccumulation of caesium has therefore aroused considerable interest[4,5]. Whereas the concentration of caesium in marine organisms is usually about three to thirty times higher than in the surrounding water, the concentration for similar organisms in fresh water may be higher by a factor of 100–1,000 (ref. 5). In laboratory experiments with isopods and snails from brackish water, Bryan[6] showed that the biological concentrations of caesium and potassium were directly related and that the concentration factors of both elements were inversely related to the salinity of the water. In the field study reported here, a bivalve mollusc, *Rangia cuneata* Gray, was collected from six stations in the Trent–Neuse estuary in eastern North Carolina (Fig. 1), and the amount of caesium-137 in the soft tissues was measured periodically for 18 months.

The Neuse River drains some 6,000 square miles of the central Piedmont region and coastal plain of North Carolina and receives radioactive pollution only from worldwide fallout. The Trent, a principal tributary, enters the Neuse at New Bern. *Rangia* is collected on a limited scale for human consumption in some estuaries of North Carolina; it is found fairly abundantly on sandy bottoms throughout the sampling area in salinities ranging from $<0{\cdot}1$ to >15 parts per thousand (Fig. 1). *Rangia* were collected at the stations once every 4–6 weeks, the shells were scrubbed and rinsed at the laboratory, and the clams were opened by steaming in a stainless steel bucket on a hotplate. Soft tissues were ashed at 450° C and subjected to gamma-ray spectrometry for analysis of radioactive content[7]. Caesium-137 from fallout and naturally occurring potassium-40 were determined as the sums of the counts comprising their respective photopeaks at 0·662 and 1·46 meV (five 20-keV channels) for a 200-min analysis after correction for background

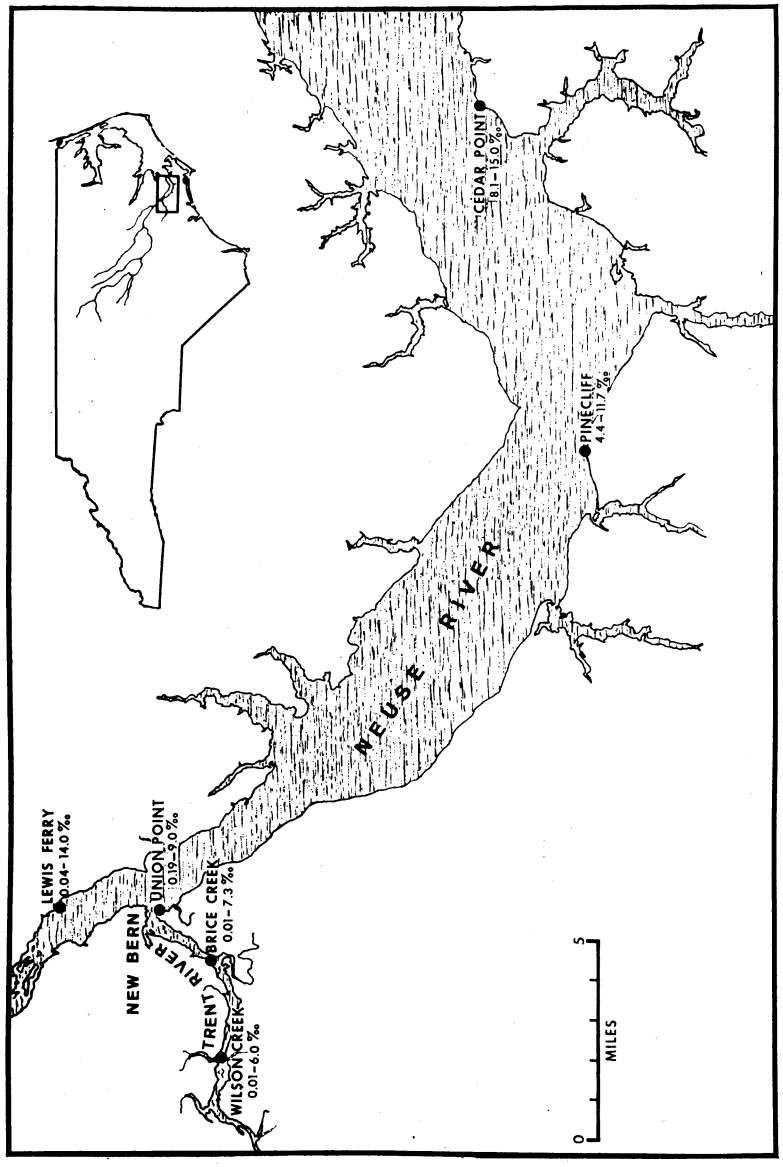

Fig. 1. Location of sampling stations for *Rangia cuneata* in the lower Trent and Neuse Rivers. Measured extremes of salinity are shown for each station.

and after spectral stripping of other gamma emitting radionuclides[8].

The ratio of caesium-137 to potassium-40 in *Rangia* at the four upstream stations exhibited a pronounced seasonal cycle which was not so apparent in clams from Pinecliff or Cedar Point. The salinity of the water, which was measured coincidentally with the collection of each sample, also varied seasonally, and reflected an inverse relationship to the caesium-137/potassium-40 ratio in *Rangia* (Fig. 2). Although the salinity, and therefore the potassium content, of the water was quite different at the six stations (usually < 1 part per thousand at Wilson and Brice Creeks and 5–15 parts per thousand at Pinecliff and Cedar Point) the concentration of potassium-40

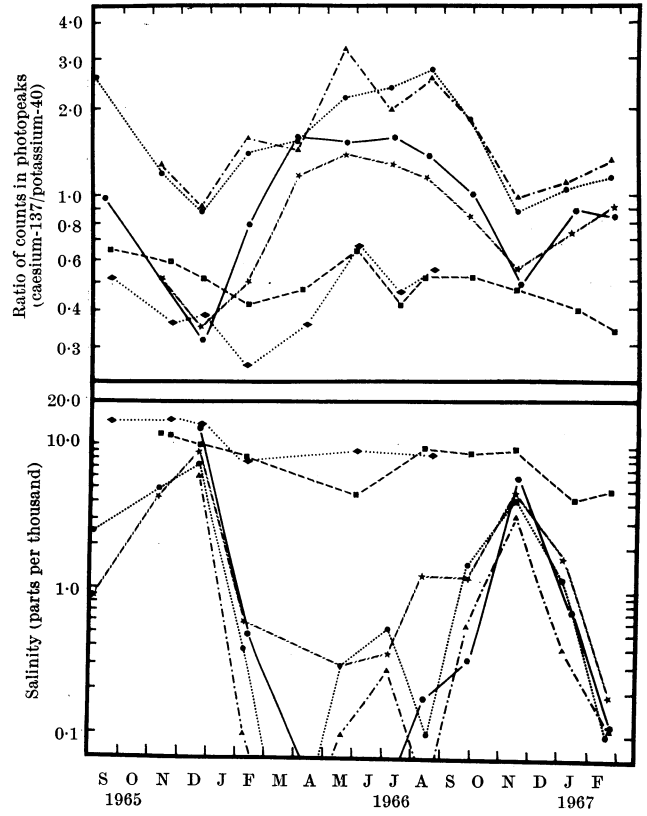

Fig. 2. Variation during 1965–67 of the caesium-137/potassium-40 ratio in *Rangia cuneata* (top) and salinity (bottom) at the stations shown in Fig. 1. Sampling at Cedar Point was discontinued in August 1966 because clams there were relatively scarce. — · —▲ —· — ·, Wilson Creek; · · · · ● · · · ·, Brice Creek; —◆—, Lewis Ferry; — · —★— · —, Union Point; – – – ■ – – –, Pinecliff; · · · ◆ · · ·, Cedar Point.

in the tissues of *Rangia* increased only two-fold with salinity (Fig. 3). In Bryan's[6] laboratory experiments, the potassium content of brackish water isopods (*Sphaeroma*) was also stable over a wide range of salinity, and the variability of potassium content at any given salinity was similar to that for *Rangia* (Fig. 3). Because concentrations of potassium are essentially constant, the variation in the ratio of caesium-137 to potassium-40 chiefly represents changes in the concentration of caesium-137 in the clam. In late spring, when the ratio of caesium-137 to potassium-40 attained its highest value at each station, the concentration of caesium-137 in *Rangia* decreased from station to station in a downstream order (Wilson Creek $\approx$ Brice Creek > Lewis Ferry > Union Point > Pinecliff $\approx$ Cedar Point). The greatest difference in caesium-137 content between clams from upstream and downstream stations was six- to seven-fold. The absolute activities of caesium-137 in *Rangia* ranged between 0·93 pc. (Cedar Point, February 9, 1966) and 8·33 pc. (Wilson Creek, May 18, 1966)/100 g wet weight, and averaged for all samples $2{\cdot}79 \pm 1{\cdot}55$ pc./100 g wet weight[8].

The seasonal fluctuations of caesium-137 content of *Rangia* may be promoted simultaneously by two phenomena, both of which are correlated with rainfall: (*a*) seasonal variation in the quantities of radioactive debris deposited in the biosphere; and (*b*) seasonal variation in the salinity (potassium content) of the river water at the sampling stations. The variation of caesium-137 in *Rangia* from upstream corresponded closely to the five-fold range reported for seasonal variation of atmospheric fallout[1]. The five-fold range of caesium-137 concentration in *Rangia* for a salinity change of more than eighty-fold (from < 0·1 to about 8 parts per thousand at Brice and Wilson Creeks) is not consistent with published results of laboratory experiments. Bryan[9] showed that the concentration of caesium-134 in the crab *Carcinus maenus* is inversely proportional to the concentration of potassium of the water. A similar relationship exists for the concentration of caesium-137 by snails and isopods from various dilutions of sea water[5]. At the nearly freshwater stations in the lower Trent and Neuse Rivers therefore the seasonal variation of caesium-137 in *Rangia* is probably caused principally by fluctuations in the deposition of fallout.

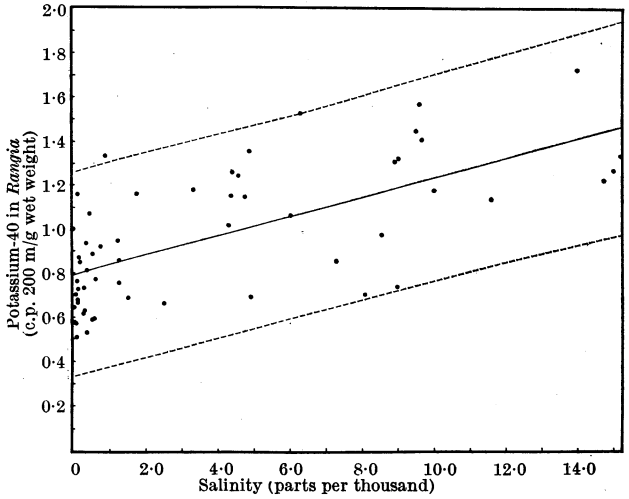

Fig. 3. Least squares regression for the relation of potassium-40 in *Rangia cuneata* to salinity of the water where the clams were collected. Dashed lines are the 95 per cent confidence limits for the regression line.

If the concentration of caesium-137 was the same in the water at all stations, the concentrations of caesium-137 in *Rangia* at Wilson Creek might be expected to be much higher in relation to those at Cedar Point than were observed. I therefore surmise that the concentration of caesium-137 is much lower in the fresh water of the Trent River than in the estuarine waters of the Neuse, and that this difference accounts for the lack of inverse proportionality between salinity and caesium-137 in *Rangia* at any one sampling time. Because the bioaccumulation of caesium-137 is influenced by the content of potassium in the water, however, estimates of potential concentrations of radiocaesium in estuarine organisms should allow for seasonal and perhaps even tidal variations in salinity.

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