Table 1. Analyses of some silica- and iron-accumulator plants (arranged in order of decreasing silica content, dry weight).

SiO <sub>2</sub>	Ash (% of dry wt.)	Weight percent in ash*									
wt.)		${\rm SiO}_2$	$\mathrm{Al}_{2}\mathrm{O}_{3}$	$\mathrm{Fe_2O_3}$	MgO	CaO	$Na_2O$	$K_{2}O$	$P_2O_5$	$SO_8$	$\mathbf{Cl}$
Giant reed, Arundu donax (leaves)†											
9.5	12.5	76.9	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
			Comm	on grom	well, Lith	ospermu	m officina	ale‡	0.17	0	
8.11	29.30	27.68	n.d.	0.28	3.15	59.01	0.77	6.17	2.17	0.77	n.d.
F F0	10.00	00.00	,	Water cl	hestnut, 2	Trapa na	tans§	C 47		9.62	0.55
5.53	19.62	28.00	n.a.	26.51	0.30	16.28	2.06	0.4/	n.a.	2.05	0.55
E 40	17.90	21.00	Giant I	horsetail,	Equisetu	m telmai	<i>eja</i> (Ehr	h)	1.96	0.05	6 19
5.40	17.59	51.08	0.96	25.59 	2.20	15.50	1.41	9.20	1.40	0.95	0.12
2.0	5 5	70 9	G	ant reed	, Arundu	donax (	stems) Ţ	n d	n d	n d	n d
5.5	5.5	70.0	n.u. E		n.u.	1.u.	л.ц. т. ш		ma.	n.u.	mai
3 46	12 12	28:51	n d.	1.04	grass, Loi 2.34	10.15	1.26	37.80	9.64	5.78	5.57
5.10	14.14	20.01	n.u.	Phoe	more Pog	maritim	a+	07100	0101	0110	0107
3.23	6.81	47.48	n.d.	4.58	2.99	4.28	10.05	11.37	4.42	1.75	11.09
			China	laurel A	ntidesma	hulvinat	um (Hil	)**			
2.97	4.26	69.72	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
				Palm	n. Calam	us rotang	t				
2.15	3.16	67.96	n.d.	0.33	11.81	16.97	0.56	0.65	0.29	0.76	n.d.
Field horsetail. Equisetum arvense (L.)											
0.37	6.05	6.19	n.d.	37.35	6.90	19.80	7.72	4.26	2.87	6.78	5.49

<sup>\*</sup> n.d., not determined. † Collector and analyst, C. G. Engel, U.S. Geological Survey, Pasadena, Calif., Feb. 1958. ‡ E. T. von Wolff, single analysis (7, pp. 110, 111, 121). § E. T. von Wolff, av. of two analyses (7, p. 133). || G. Mariani, individual analysis (8). # E. T. von Wolff, av. of 11 analyses (7, p. 121).
\*\* L. E. Wise, wood samples from Hawaii (9).

cumulator vegetation would require  $2.5 \times$ (2000-475) years or about 3800 years to convert 1 acre foot of fresh basalt to 1 acre foot of this lateritic soil if silica were removed solely through the agency of these plant accumulators.

Not all the silica moving into plant accumulators would be lost. Nevertheless, whether the silica is present in the plant as opal, as fine-grained quartz, as inorganic compounds, or as organic compounds, it would be highly vulnerable to chemical and physical erosion while the plant matter was decomposing. In areas of high rainfall where a tropical downpour equivalent to several inches of precipitation may take place in a few hours, much of the organic debris could be washed into the nearest drainage channel. The reality of such a process is evident in the high organic content of the brown and black waters so common in the tropics.

Some of the silica-and other elements-in forest litter is recycled, and some of it is dissolved and enters groundwater circulation; under favorable conditions, however, substantial quantities of silica must be pumped out of the ground at depth, spread over the surface in organic debris, and then swept off by erosive agents.

Although I have taken silica as an example, it is, of course, evident that the accumulator plants that select aluminum in preference to silica would tend to deplete the soil of this element and leave the ground enriched in silica. Hutchinson (5) has described many aluminumaccumulator plants, several of which would apparently be competent to 22 AUGUST 1958

change materially the ratio of alumina to silica in the soil if the plant community contained a preponderance of them and a dearth of silica accumulators. The quantitative aspects of the process suggested above must remain speculative until field studies are made in suitable areas, but the entire geobiochemical history of silica-and of other elements taken up by accumulator plants -is one of great interest and seems worthy of further investigation (6).

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14 March 1958

## Long-Lived Cobalt Isotopes **Observed in Fallout**

It has generally been assumed that the long-term radiological hazards due to fallout from a nuclear detonation depend primarily upon the long-lived fission products. However, other radioactive products which are not derived from fission reactions may be present in fallout and may contribute to the hazard. These latter products would be created by neutron activation of elements in the immediate vicinity of the detonation. The presence of one such product, Co<sup>60</sup>, has been indicated by analyses of fallout material (1) and of biological specimens recovered from fallout areas (2).

The investigation discussed in this report, which was carried out to determine the amounts of Co<sup>60</sup> in fallout samples from the 1956 test series, revealed the existence of two additional cobalt nuclides, Co<sup>57</sup> and Co<sup>58</sup>. The amounts of these nuclides were determined. To permit assessment of the relative internal hazards from the cobalt isotopes, determinations of Sr<sup>90</sup> were made on the same samples. From these data the Co<sup>60</sup>/Sr<sup>90</sup>, Co<sup>58</sup>/Sr<sup>90</sup>, and Co<sup>57</sup>/Sr<sup>90</sup> ratios were derived. Since all three cobalt isotopes emit gamma radiation, they would also contribute to an external hazard. The amount of this hazard was estimated by summing the photon energy release per unit time from the observed amounts of radiocobalt and comparing this value with similar values calculated for the associated fission products.

Radiochemical analyses were carried out on three samples of fallout obtained after one detonation of the 1956 nuclear weapon test series in the Marshall Islands. The samples consisted of large trays which had been exposed at various locations within the fallout pattern. After removal of the radioactive material and solution by acid treatment, aliquots were taken for strontium and cobalt analyses.

After the addition of strontium carrier and Sr<sup>85</sup> (used as a tracer for chemical yield determinations), the strontium was separated by precipitation with fuming nitric acid and determined finally as the oxalate (3). The oxalates were dried and mounted for beta counting. The Sr<sup>90</sup> was determined by measurement of the growth of the daughter, Y<sup>90</sup>. The samples were quantitatively redissolved, and the chemical vield was determined through a gamma-ray spectrometer measurement of the remaining Sr<sup>85</sup> tracer. After application of the usual corrections, the number of Sr<sup>90</sup> atoms present at zero time was determined; a half-life value of  $27.7 \pm 0.4$  years was used (4).

Cobalt was separated by adaptation of existing procedures (3). Since Mn<sup>54</sup> was known to be present, manganese carrier

Table 1. Ratios of abundance of cobalt atoms to abundance of Sr<sup>80</sup> atoms at zero time.

Item	Co <sup>60</sup> /Sr <sup>90</sup>	Co <sup>57</sup> /Sr <sup>90</sup>	Co <sup>58</sup> /Sr <sup>90</sup>
Sample A	$0.323 \pm 0.106$	$0.051 \pm 0.017$	$0.027 \pm 0.020$
Sample B	$0.245 \pm 0.102$	$0.038 \pm 0.013$	$0.021 \pm 0.015$
Sample C	$0.255 \pm 0.090$	$0.040 \pm 0.013$	$0.022 \pm 0.016$
Avg. values	0.274	0.043	0.023
Avg. errors	$\pm 0.100$	$\pm 0.014$	$\pm 0.017$

was added (5). After a hydroxide scavenging from an ammoniacal solution, the supernatant solution was treated with  $H_2S$ . The resulting precipitate was extracted with a  $Na_2SO_4$ -NaHSO<sub>4</sub> solution to remove manganese. The cobalt was alternately precipitated several times as the hydroxide and cobaltinitrite. Chemical yield was determined by weighing the cobaltinitrite.

The cobalt yields were dissolved and analyzed for gamma activities by means of a single-channel gamma pulse-height analyzer. The presence of  $Co^{60}$ ,  $Co^{57}$ , and  $Co^{58}$  was established by identification of their respective photopeaks, by comparison with standards of known energy.

The disintegration rate of each cobalt isotope was obtained by analysis of pho-

topeak areas. After determination of the total pulse-height distribution, a standard of Co<sup>60</sup> was prepared to equal the magnitude of the Co<sup>60</sup> photopeaks of the isolated cobalt. This Co60 pulse-height distribution was subtracted from the total distribution. This subtraction of the Co<sup>60</sup> Compton continuum permitted determination of the Co57 and Co58. Their disintegration rates were computed from experimentally determined gamma-ray efficiencies and consideration of their respective decay schemes (relative photon abundance, internal conversion, and so on) (6). Figure 1 shows the pulse-height distribution of the separated cobalt and the spectrum of the Co60 standard. The 0.84-Mev Mn<sup>54</sup> photopeak is shown as an energy reference.

The quantities of the isotopes Co<sup>57</sup>,



Fig. 1. Comparison of pulse-height distribution of separated cobalt sample with distributions of reference standards.

Co<sup>58</sup>, and Co<sup>60</sup> and the fission product  $Sr^{90}$  (and its precursors) in each of the samples at zero time were in the order of  $10^{10}$  to  $10^{12}$  atoms. These values were used to compute the Co<sup>57</sup>/Sr<sup>90</sup>, Co<sup>58</sup>/Sr<sup>90</sup>, and Co<sup>60</sup>/Sr<sup>90</sup> ratios listed in Table 1. The errors associated with these values are those produced by all determinate errors present in the calculations—that is, counting error, pipetting error, errors in chemical yield, and so on. The average values show the average error of the three measurements.

The ratios shown in Table 1 indicate that significant amounts of radiocobalt were formed relative to  $Sr^{90}$ . Possible reactions for the formation of the cobalt nuclides from environmental nickel and cobalt are:

$$\begin{array}{c} \operatorname{Co}^{56}(n,\gamma)\operatorname{Co}^{60}\\ \operatorname{Ni}^{60}(n,p)\operatorname{Co}^{60}\\ \operatorname{Co}^{59}(n,2n)\operatorname{Co}^{58}\\ \operatorname{Ni}^{58}(n,p)\operatorname{Co}^{58}\\ \end{array}$$

The biological hazards resulting from fallout may be placed in two categories: (i) the internal hazard caused by ingestion and inhalation of active material and (ii) the external hazard caused by penetrating radiation the source of which is external to the body.

If material the ratios of whose atoms are those found in Table 1 were ingested or inhaled, the hazard due to the presence of the radiocobalt would be negligible as compared to that from the  $Sr^{90}$ . This is evident when, among other factors, the biological half-lives  $(3.9 \times 10^3$ days for  $Sr^{90}$ ; 8.4 days for  $Co^{60}$ ) (7) and the ionizing radiation emitted ( $\beta$  for  $Sr^{90}$ ; primarily  $\gamma$  for  $Co^{57}$ ,  $Co^{58}$ , and  $Co^{60}$ ) are compared.

External hazard in fallout may be considered to be that primarily due to gamma radiation. This hazard is a function of photon energy, scattering, absorption, and so on. The net effect, however, of these factors for fallout situations (approximated by uniformly contaminated fields) is that the hazard is roughly proportional to energy release per unit time. Values of gamma energy released per second  $(\mu_1)$  of fission products resulting from 10<sup>4</sup> fissions have been computed for times up to  $1.2 \times 10^3$  yr after time of fission (8). The values so obtained may be compared over the same time period with the energy released per second  $(\mu_2)$ by the quantities of radiocobalt found associated with 104 fissions. This ratio  $\mu_2/\mu_1$ , in percent, is found to be 0.14 at 9.8 days, 31 at 1.20 yr, 149 at 2.6 yr, 211 at 5.6 yr, and 26 at 25.7 yr. These values represent the analyses of a limited number of samples from one detonation. It is evident, however, that radiocobalt pro-

duced in the ratios given in Table 1 would constitute a considerable portion of the external hazard of fallout, particularly at times greater than 1 yr.

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7 July 1958

## Ontogeny of Hemoglobin in the Skate Raja binoculata

In all sufficiently studied mammals, the fetus has a hemoglobin that is biochemically distinct from that of the adult (1). Ontogenetic changes in the hemoglobin are found also in the chicken (2),



Fig. 1. Oxygen dissociation curves of hemoglobin solutions prepared from blood of adult (approximately 100 kg), fetal (9 g), and embryonic (1.5 g) skates. Conditions: 1.5 percent hemoglobin in potassium phosphate buffer; final ionic strength, 0.267; pH, 6.50 to 6.58; temperature, 10°C. Crosses represent a solution of adult hemoglobin subjected to a second analysis 48 hours after the original determination of the oxygen equilibrium.

the terrapin (3), the bullfrog (4), and the teleost fish Scorpaenichthys (5). The possibility that a developmental sequence of at least two hemoglobins is characteristic of the vertebrates appears to be contradicted by McCutcheon's failure to find any difference between the hemoglobin of adult and fetal viviparous rays (3). However, McCutcheon states: "The lack of early stages in the collections leaves open the possibility of an early developmental Hb [hemoglobin] with a higher affinity [for oxygen] than fetal and adult Hb." Because a few egg-cases of the barn-door skate Raja binoculata were collected during otter-trawling, it was possible to study the oxygen equilibrium of hemoglobin solutions and erythrocyte suspensions (equivalent to blood) of this oviparous elasmobranch in individuals of various ages (6).

A comparison of oxygen dissociation curves of adult, fetal, and embryonic hemoglobin is shown in Fig. 1. From such data one can calculate (7) the two constants of the Hill approximation:  $p_{50}$  is the oxygen tension at which there are equal quantities of oxygenated and deoxygenated hemoglobin; and n is a measure of the heme-heme interactions. The parameter n determines the shape of the oxygen dissociation curve, and  $p_{50}$  determines the position of the curve. Figure 2 shows the variation of these two constants as functions of pH for skate hemoglobin inside and outside the erythrocyte. From the information presented in these figures it can be concluded that fetuses (wet weight 9 to 27 g) and adults (to 100 kg) have hemoglobin with the same oxygen equilibrium.

This finding parallels the studies of McCutcheon on viviparous rays (3). However, the hemoglobin of three embryos (wet weight 1 to 1.6 g) possessed a slightly higher oxygen affinity in both erythrocyte suspensions and hemoglobin solutions; furthermore, for this possible "embryonic hemoglobin," heme-heme interaction was almost completely removed by hemolysis and subsequent dilution, whereas the same treatment did not alter "fetal and adult hemoglobin" in this respect. It is entirely possible that some adult hemoglobin was present along with the embryonic hemoglobin; however, since n for hemoglobin solutions obtained from the embryos was 1.1, as opposed to n = 1.5 to 1.8 for adult and fetal hemoglobin in solution, one can calculate, assuming that n cannot be less than 1.0 for embryonic hemoglobin and that no embryonic hemoglobin is present in adult skates, that at least 80 percent of the hemoglobin present in these embryos is of a distinct type. Further experimentation is limited by the scarcity of egg-cases and the small quantity of hemoglobin available from embryos (0.2 ml of a 1.5 percent hemoglobin solution from a 1.6-g embryo).

Since the skate egg-case, in which the embryos and fetuses remain for 1.5 to 2 years of development, is relatively isolated from the environment except for the diffusion of respiratory gases (the "cleidoic" condition) (8), it might be expected that hemoglobin with high oxygen affinity would be needed throughout not only the embryonic but also the fetal period, in contrast to observed results. However, observations on ten egg-cases indicated the egg-case is a "closed system" only during the first few months of development. When the egg-case is laid, a hole (2 by 4 mm) at each of the four corners is plugged with a very viscous mucus, which is gradually dissolved during development. In all fetuses with adult hemoglobin, the four holes were open; in the embryos weighing 1 to 1.6 g, only two of the holes were still plugged. The oxygen tension [polarographically determined (9)] inside these egg-cases was rarely less than half that of sea-water. The urea retention function of the skate egg-case (8) must take place only during the initial phases of development, although even small embryos



Fig. 2. Oxygen affinity (top) and hemeheme interaction (bottom) as functions of pH for the hemoglobin in solution and in erythrocytes for adult, fetal, and embryonic skates. Conditions: 1.5 to 2 percent hemoglobin solution in potassium phosphate buffer; final ionic strength, 0.267; erythrocyte suspensions in elasmobranch Ringer's solution; temperature, 10 to 11°C.