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DISCUSSION

G. MICHON: Could you give any further information regarding the lower sensitivity limit of the simplified method for the transfer of radon from the bubbler to the measuring cell?

D.E. RUSHING: The lower limit of detection depends upon the blanks obtainable and these depend, amongst other things, on the gas used for deemanation. The figure given in the paper is about 0.05 pg Ra, but subsequent work has enabled us to reach lower levels for blanks down to a few hundredths of a picogramme.

ENVIRONMENTAL MONITORING IN THE VICINITY OF URANIUM MILLS

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Abstract — Résumé — Аннотация — Resumen

ENVIRONMENTAL MONITORING IN THE VICINITY OF URANIUM MILLS. This paper described the extensive experience of the Public Health Service in monitoring the environment near United States uranium mills, and the highly successful pollution-control programme that has resulted. Environmental contamination from mills has involved primarily the water environment, and monitoring has been mainly for Ra^{226}, with determination of uranium and Sr^{90} as desirable. Studies have ranged from brief reconnaisance surveys to quantitative evaluation of radiation exposure of substantial population groups, depending upon degree of pollution, importance of affected water uses, and likelihood of human exposure. A Ra^{226} surveillance network covering seven States and eleven mills has recently been established to assure continued protection.

Monitoring has included river sediments, aquatic biota (insects, algae, fish), water, mill effluents, and crops and topsoils from farms irrigated with contaminated river water. This has provided understanding of the immediate and ultimate fate of Ra^{226} in the water environment, and has shown which media provide the best indication of pollution. Thus, for instance, the Ra^{226} surveillance network uses sediment as well as water sampling.

Spent ore solids, or tailings, have proved to be the most important source of environmental contamination by mill wastes. In the Animas River, for example, 95% of the dissolved Ra^{226} in the downstream river water resulted from leaching of river bed deposits of tailings, rather than from direct mill discharges of Ra^{226} in the dissolved state. Based on these findings, improved procedures for capturing and retaining the tailings at the mill yielded immediate sharp reduction of downstream dissolved Ra^{226}.

The river sediments have proved to be an excellent indicator of mill pollution. Their natural Ra^{226} content has been established at 1.3 pg. Ra^{226}/g dry sediment, and contamination at twice this level is readily detectable. Routine surveillance of their gross alpha and/or Ra^{226} content is a reliable indicator of accidental spills of mill wastes. The great reservoirs of the Colorado River Basin have been shown to be the final resting place of mill tailings released in earlier years.

Ra^{226} concentration factors for aquatic biota have been determined, and aquatic populations have been used as a reliable indicator of the presence of chemically toxic mill wastes such as organic raffinate. Ra^{226} uptake from polluted irrigation water by various crops has been measured, and has shown that feed crops such as alfalfa can become highly contaminated.

These studies and the co-operation of the United States milling industry have yielded a high degree of protection of the waters of the Colorado Basin. Earlier dissolved Ra^{226} concentrations ranged as high as 88 pg/l, whereas presently these streams contain less than 1.0 pg/l.

CONTRÔLE RADIOLOGIQUE DU MILIEU AU VOISINAGE DES USINES D'URANIUM. Le mémoire décrit l'expérience étendue acquise par le Service de santé publique dans le domaine du contrôle radiologique du milieu, au voisinage des usines d'uranium des Etats-Unis, ainsi que le programme très éfficace de lutte contre la pollution qui en est résulté. La contamination de l'environnement par les usines atteint en premier lieu le milieu aquatique. Le contrôle de la radioactivité a porté surtout sur ^{226}Ra; l'uranium et le strontium-90 ont en outre été recherchés en cas de besoin. Les études s'échelonnent entre de brèves enquêtes préliminaires et l'évaluation quantitative de l'irradiation d'importants groupes de populations, selon le degré de pollution, l'importance des emplois de l'eau compromis et la probabilité d'une exposition d'êtres humains. Un réseau de surveillance pour ^{226}Ra, couvrant sept Etats et onze usines a été récemment mis en place pour assurer une protection continue.

Le contrôle radiologique a porté sur les alluvions fluviales, la faune et la flore aquatiques (insectes, algues, poissons), l'eau, les effluents d'usines, ainsi que les cultures et terres arables des exploitations agricoles

irriguées avec de l'eau de rivière contaminée. On a ainsi compris ce que devenait ^{226}Ra dans le milieu aquatique immédiatement après son introduction dans l'eau et ultérieurement, on a pu déterminer quels éléments du milieu donnaient la meilleure indication sur la pollution: par exemple, pour ^{226}Ra ce sont les alluvions et l'eau.

Les stériles ou les produits de queue se sont révélés la principale source de contamination de l'environnement par les déchets des usines de traitement du minerai. Dans les eaux de l'Animas, par exemple, 95% du ^{226}Ra trouvé en aval de l'usine résultaient du lessivage des dépôts de stériles dans le lit de la rivière plutôt que du rejet direct d'effluents contenant ^{226}Ra en solution. Aussi a-t-on mis au point des méthodes améliorées pour retenir les stériles à l'usine et on a obtenu immédiatement une forte réduction du ^{226}Ra dissous en aval.

Les alluvions fluviales se sont révélées un excellent indicateur de la pollution provenant de l'usine. Leur teneur en ^{226}Ra naturel est de 1,3 pg de ^{226}Ra par gramme de sédiment sec, et l'on peut détecter facilement un niveau de contamination égal ou double de cette valeur. La surveillance continue de leur rayonnement alpha brut ou de leur teneur en ^{226}Ra, ou des deux ensemble, permet de déceler, à coup sûr, le déversement accidentel de déchets d'usines. On a constaté que les stériles déversés par les usines de traitement au cours des années finissaient par s'accumuler dans les grands bassins de retenue du Colorado.

On a déterminé les facteurs de concentration de ^{226}Ra pour la flore et la faune aquatiques et on a utilisé les populations aquatiques comme témoins sûrs de la présence de déchets d'usines chimiquement toxiques tels que les résidus concentrés de la séparation par solvant organique. On a mesuré l'absorption par diverses cultures de ^{226}Ra provenant d'eau d'irrigation polluée et on a constaté que les plantes fourragères telles que la luzerne peuvent être fortement contaminées.

Ces études, et la coopération des entreprises d'extraction américaines, ont permis de protéger considérablement les eaux du Bassin du Colorado contre la pollution. Les concentrations de ^{226}Ra en solution, qui atteignaient autrefois jusqu'à 88 pg/l, y sont à présent inférieures à 1,0 pg/l.

ДОЗИМЕТРИЯ ОКРУЖАЮЩЕЙ СРЕДЫ ВБЛИЗИ ПРЕДПРИЯТИЙ, ОБРАБАТЫВАЮЩИХ УРАНОВУЮ РУДУ. Дается описание богатого опыта службы здравоохранения в отношении дозиметрии окружающей среды вблизи американских урановых предприятий и очень успешная программа контроля загрязнения, которая уже завершена. Загрязнение окружающей среды отходами урановых предприятий касается в основном находящихся вблизи водных бассейнов; контролировалось, главным образом, содержание радона-226, а определение содержания урана и стронция-90 считалось желательным. Исследования были различными, начиная от непродолжительных рекогносцировочных обследований и кончая количественной оценкой облучения больших групп населения, при этом учитывались степень загрязнения, а также размеры потребления воды и вероятность облучения населения. Для обеспечения постоянной защиты недавно была создана система наблюдения за содержанием радона-226, охватывающая семь штатов и 12 обогатительных предприятий.

Дозиметрический контроль распространялся на речные отложения, водную фауну и флору (насекомые, водоросли, рыбы), воду, сбросы обогатительных предприятий, сельскохозяйственные растения и верхний слой почвы с ферм, использующих для орошения загрязненную речную воду. Это позволило установить ближайшие и окончательные места отложения радона-226 в окружающей водной среде и показало, какая среда является наиболее показательной в отношении загрязнения. Так, например, в системе наблюдения за содержанием радона-226 используется взятие проб донных отложений и воды.

Переработанные рудоносные породы, или хвосты, оказались самым важным источником загрязнения окружающей среды отходами обогатительных предприятий. Например, в нижнем течении реки Анимас 95% растворенного в воде радона-226 является следствием выщелачивания речных донных отложений хвостов, а не прямого сброса обогатительным предприятием радона-226 в растворенном состоянии. Основанные на этих выводах усовершенствованные методы улавливания и удержания хвостов на обогатительных предприятиях позволили сразу же резко уменьшить количество растворенного в воде радона-226 в нижнем течении реки.

Донные отложения в реке оказались прекрасным показателем загрязнения. Естественное содержание радона-226 составляет 1,3 мкмкг/г сухого осадка, а загрязнение превышает вдвое эту величину и поэтому легко обнаруживается. Постоянное наблюдение за полной альфа-активностью и/или содержанием радона-226 в отложениях является надежным способом обнаружения аварийной утечки отходов обогатительных предприятий. Было показано, что огромные резервуары бассейна реки Колорадо должны быть местом окончательного отложения хвостов обогатительного предприятия, сброшенных в предыдущие годы.

Были определены коэффициенты концентрации радона-226 для водной фауны и флоры, а водная фауна использовалась как надежный индикатор наличия химически токсичных отходов обогатительных предприятий, таких, например, как органический рафинат. Было измерено поглощение радона-226 из загрязненной ирригационной воды различными сельскохозяйственными растениями и показано, что кормовые культуры, например люцерна, могут оказаться очень загрязненными.

Эти исследования и сотрудничество американских обогатительных предприятий обеспечили высокую степень охраны вод бассейна реки Колорадо. Раньше концентрации растворенного радона-226 достигали 88 мкмкг/л, тогда как сейчас в воде этих рек его содержание составляет менее 1,0 мкмкг/л.

VIGILANCIA DEL MEDIO AMBIENTE EN LAS PROXIMIDADES DE LAS FÁBRICAS DE URANIO. La memoria reseña la vasta experiencia adquirida por los servicios sanitarios en el análisis del medio ambiente de las proximidades de las fábricas de uranio estadounidenses, y el programa de control de la contaminación atmosférica que se ha puesto en práctica. Las fábricas contaminan principalmente las aguas de los alrededores, así que la finalidad principal de los análisis es detectar la presencia de ^{226}Ra y, cuando se considera conveniente, de uranio y de ^{90}Sr. Los estudios comprenden desde breves análisis hasta la determinación cuantitativa de irradiación de grupos importantes de la población, según sea el grado de contaminación, la importancia del empleo de las aguas afectadas y las probabilidades de irradiación humana. Recientemente se ha establecido una red de control radiológico del ^{226}Ra que brinda protección continua a siete Estados y once fábricas.

Se han analizado sedimentos de ríos, biota acuático (insectos, algas, peces), aguas, efluentes de fábricas, y plantas cultivadas y tierra de granjas regadas con agua de los ríos contaminados. Gracias a ello se sabe lo que sucede con el ^{226}Ra del agua y de qué manera se detecta mejor la contaminación. Por ejemplo, la red de control radiológico del ^{226}Ra utiliza muestras de sedimentos y de agua.

Los desechos de las fábricas que más contaminan el medio ambiente son las menas agotadas. Por ejemplo, el 95 por ciento del ^{226}Ra disuelto en el agua del río Animas procede de la lixiviación de los desechos sólidos evacuados en el cauce del río, y no del ^{226}Ra disuelto que descarga la fábrica directamente en el agua. Gracias a los datos obtenidos se mejoraron los procedimientos empleados para capturar y retener los desechos de la fábrica, e inmediatamente se logró disminuir mucho la cantidad de ^{226}Ra disuelto en el río.

Los sedimentos fluviales proporcionan una excelente medida de la contaminación orgininada por la fábrica. Su contenido natural de ^{226}Ra se ha fijado en 1,3 pg de ^{226}Ra por gramo de sedimento seco, y es muy fácil detectar una contaminación equivalente al doble de este valor. Un análisis periódico de su actividad alfa o de su contenido de ^{226}Ra revela perfectamente cualquier contaminación accidental producida por desechos de la fábrica. Se ha visto que el lugar donde se depositan finalmente los desechos evacuados años atrás son los embalses de la cuenca del río Colorado.

Se han determinado los factores de concentración del ^{226}Ra en el biota acuático, que se ha empleado por ser indicador excelentes de la presencia de desechos de fábrica químicamente tóxicos, tales como los refinados orgánicos. Se ha medido la captación por diversos cultivos del ^{226}Ra contenido en el agua de riego, y se ha demostrado que las plantas forrajeras, por ejemplo la alfalfa, pueden resultar altamente contaminadas.

Estos estudios y la cooperación de la industria estadounidense han servido para proteger en alto grado las aguas de la cuenca del Colorado. Tiempo atrás, las concentraciones de ^{226}Ra disuelto alcanzaban hasta 88 pg/l, mientras que actualmente estos ríos contienen menos de 1,0 pg/l.

HISTORICAL BACKGROUND

In order to fill wartime and later peacetime uranium requirements, the United States uranium mining and milling industry began a sharp expansion some twenty years ago. Substantial uranium deposits were discovered in the western United States of America, and by the late 1950's uranium mills in that area were processing in excess of 20 000 t/d of ore with an average U_3O_8 content of about 0.28%. This appears to be a maximum processing rate not likely to be greatly exceeded in the near future.

During the latter 1940's the possibility of significant environmental contamination by uranium-mill wastes received little attention, primarily because little was known about the fate of such materials in the environment

and their effects on human health. In subsequent years, however, especially during the 1950's, a great deal of factual information was obtained regarding the fate of radioactive mill wastes in the water environment and the paths by which these materials may enter the human diet. Concepts of protection of the public against excessive radiation exposure also changed during this period, and associated maximum permissible concentrations (MPC's) of radioisotopes in water for human consumption have decreased accordingly [1]. Also, suitable methods of waste handling and control have been developed, and at present a high degree of environmental and public health protection has been achieved.

It was realized in the late 1940's that the most hazardous radioisotope present in the uranium mill wastes was Ra^{226}, and in 1950 the first exploratory river-water samples were collected to determine whether any real problem existed. Four water samples from river locations below uranium mills contained from 4.5 to 43 pc/l of dissolved Ra^{226}. In contrast, four companion samples of river water from above the mills showed a natural background dissolved Ra^{226} concentration averaging 0.2 pc/l [2]. Thus, the presence of stream contamination was clearly demonstrated, but the observed Ra^{226} concentrations did not appear to be unacceptably high in view of the standards for human protection that were generally accepted at that time.

Following this, however, the MPC for dissolved Ra^{226} in water for public consumption was more definitely established at 4.0 pc/l [3], and later at 3.3 pc/l [4]. During the same period it was estimated that at the anticipated uranium ore processing rate of 20 000 t/d there would result some 5000 g/yr of Ra^{226} to be disposed of as waste.

Thus, beginning in 1955 more extensive studies of the degree of surface-water contamination with Ra^{226} were conducted. They ranged from brief reconnaissance surveys all the way to full-scale studies of Ra^{226} intake by substantial population groups [5], and included in-plant studies of the characteristics of uranium-mill wastes, as well as studies of the fate of these materials in surface streams and evaluation of the chemical toxicity of the wastes to aquatic life. The detailed techniques and results of many of these studies have been published, and may be found in the references given here. A great deal has been learned about the monitoring and surveillance of various environmental media in the course of these studies, and the purpose of this paper is to bring that information together for the use of others.

MONITORING OF ENVIRONMENTAL MEDIA

Although the comments in this section of the paper reflect experience primarily with monitoring for Ra^{226}, much of the information has meaning also in regard to other radioisotopes associated with uranium-mill wastes. The gross alpha radioassay [2] has proved especially useful as a screening analysis to determine whether isotopic analysis of samples is necessary. Analysis for uranium has often been of value in accounting for most of the gross alpha activity, and, more recently, analysis for thorium and for lead-210 has proved desirable [6]. In both river surveys and in-plant mill-

waste surveys, the quantitative accounting for all Ra^{226}(dissolved and suspended) passing each successive sampling location has proved to be the essential key to successful evaluation and control of the problem.

WATER MONITORING

In contrast to other types of environmental media discussed below, single or "grab" samples of a river water or a water supply yield only an instantaneous evaluation of water quality. Thus, in exploratory surveys designed to provide estimates of the extent and magnitude of contamination, single samples suffice and have been used. In other studies such as those on the Animas River [5] where the purpose has been to evaluate human Ra^{226} intake, instantaneous samples are not adequate, and field sampling devices that collect longer term composite water samples have been used. The details of one such composite sampler have been described elsewhere [7].

Dissolved versus undissolved Ra^{226}

The MPC referred to above for Ra^{226} in water for public consumption is for "soluble" Ra^{226}[4] which, for want of a sufficiently precise definition of the term "soluble," has been taken to mean "dissolved" Ra^{226} in our work. In many situations this distinction may be unnecessary, but our early experience with uranium mill wastes indicated clearly that accurate evaluation of Ra^{226} intake required quite careful initial handling and treatment of water samples to determine the initial content of Ra^{226} in the dissolved and undissolved states.

Ra^{226} is relatively insoluble in water, and most (98%, or so) of the Ra^{226} in uranium-mill wastes occurs undissolved in the spent ore solids, or tailings. We have also found that the very fine tailings contain the highest Ra^{226} concentrations (2500 or more pc Ra^{226}/g of dry solids) [8]. These finest or lightest suspended particles are most likely to escape sedimentation basins and become a part of the effluent to a stream. Other studies have also demonstrated that some of this initially undissolved Ra^{226} is eventually leached by water and will become dissolved [9]. As a result, most mill effluents and downstream river-water samples contain at least small quantities of fine mill tailings that have relatively high Ra^{226} content.

It is common practice, in collecting ordinary water samples for Ra^{226} analysis, to add a small amount of HCl to the sample at the time of collection, in order to be sure that the Ra^{226} initially present in the dissolved state will remain dissolved until the sample is analysed. For samples containing uranium mill waste solids, this practice, of course, only adds to the problem of knowing what the dissolved Ra^{226} concentration was at the time the sample was collected.

Thus, unless the initial treatment of water samples is designed to avoid and minimize the above effects, erratic and inaccurate dissolved Ra^{226} determinations may result. Based upon these considerations, our procedure consists of filtering the sample through a millipore filter promptly after collection, to separate the suspended solids. The HCl may then be added to the filtered water to prevent Ra^{226} precipitation before analysis, and the

suspended and dissolved Ra^{226} analyses are then determined separately. In this way we hope to obtain the best possible evaluation of dissolved and suspended Ra^{226} at the time of sample collection.

Observed radioactivity concentrations

During the past seven years several thousand water samples from western United States streams have been analysed for dissolved Ra^{226} and many of these also for gross alpha radioactivity. Several hundred have also been analysed for uranium, and a small number recently for Pb^{210}. Earlier studies placed primary emphasis on particular stream sections containing relatively high levels of pollution, but more recently pollution has decreased sharply, and for the past year water monitoring has consisted primarily of the operation of a surveillance network designed to evaluate long-term radioactivity levels and to detect accidental spills of uranium mill wastes [6].

The natural or background concentration of dissolved Ra^{226} in surface waters of the western United States has proved in the majority of cases to be less than 0.1 pc/l, with a small fraction of the results ranging up to 0.2 or 0.3 pc/l. An occasional result has been as high as 0.8 or 1.0 pc/l, but this has usually been associated with local uranium mining activities or with a very small high mountain creek. The natural gross alpha radioactivity of these surface waters does not often range above 10 pc/l. The observed natural uranium content has averaged about 3 μg/l, and has ranged as high as 8 or 10 μg/l (or as high as about 6 pc/l). The very small number of samples analysed thus far for Pb^{210} indicate a natural concentration of less than 0.1 or 0.2 pc/l.

In contrast, in earlier years at stream locations downstream from uranium mills, dissolved Ra^{226} concentrations as high as 88 pc/l have been observed, and dissolved gross alpha activities have been over 2000 pc/l. Uranium and Pb^{210} data are not available for this period.

At present, our surveillance network of 24 sampling stations indicates that the co-operative programme of the States, the industry, and the federal agencies has achieved a high level of environmental protection. For the past year, dissolved Ra^{226} concentrations have averaged less than 1.0 pc/l at each of the river sampling stations, most of which are located below uranium mills. Average dissolved uranium has ranged up to only 38 μg/l at one of the sampling stations, and is generally close to natural levels at most of the others. Gross alpha activities have been at comparably low levels.

These average concentrations are substantially lower than present standards for the radioactive quality of water for domestic use. This achievement by an entire industry reflects a sincere desire to protect the water environment and minimize human radiation exposure in every practical way.

Sediment monitoring

In our studies, the Ra^{226} content of river muds and sediments has proved to be the best indicator of uranium-mill pollution and particularly of the degree to which it has occurred in the past. Spent ore solids, or tailings, released in the mill effluents settle to the river bed in pools and reaches of

low stream velocity. They continue to accumulate at such locations as long as stream flows remain reasonably low and floods do not occur. Thus, single samples from such a location can yield a cumulative estimate of the quantity of mill tailings that have deposited there since the last high flow. It was noted in early studies that relatively high dissolved Ra^{226} concentrations in the flowing stream were invariably associated with high Ra^{226} concentrations in the sediments and bottom muds.

Fig. 1 is a typical daily hydrograph for streams of the western United States. The major annual flood flows begin in late March or early April, when the snow in the high mountains begins to melt, and extend into July. Following that, an extended low flow period occurs and lasts for approximately eight months. The floods scour the river bed and cleanse it of prior accumulations of mill tailings. Settling and accumulation begins in August and continues virtually uninterrupted through the winter until the floods again begin in April.

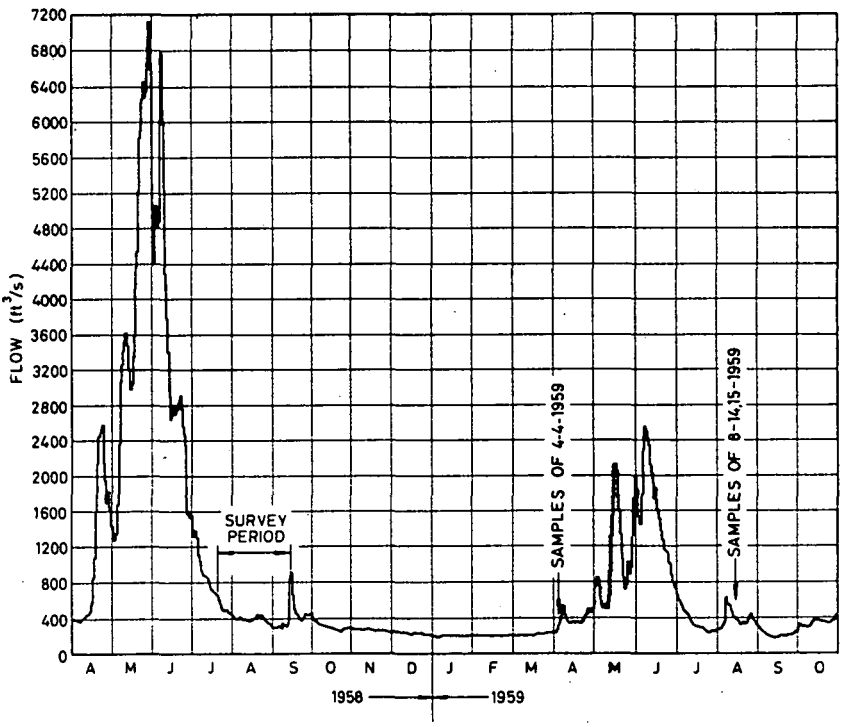

Fig. 1

Animas River at Durango, Colorado from April 1956 to October 1959

Hence, sediment samples taken below a uranium mill in mid-summer generally contain relatively low quantities of Ra^{226}, whereas those taken in the late winter contain maximum quantities. This is illustrated in Fig. 2, showing data taken from the Animas River studies. As may be seen, Ra^{226} concentrations in river muds collected in early April were about three times

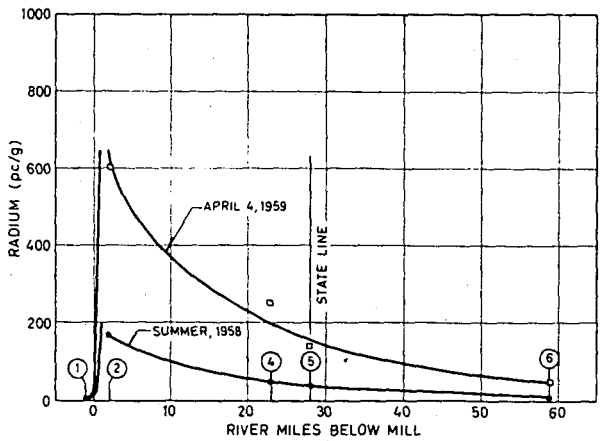

Fig. 2

Animas River: radium content of river muds in 1958-1959

as high as the concentrations observed in the preceding summer, ranging up to 600 pc Ra^{226}/g of dry sediment.

The Ra^{226} contained in such river sediments is leachable, and appears at downstream locations as dissolved Ra^{226} in the flowing water. This was first demonstrated in the field studies of the Animas River, and later in the laboratory [9]. Before pollution abatement measures were installed at the uranium mill, the dissolved Ra^{226} concentration averaged 12.6 pc/l in the Animas River two miles downstream. Treatment at the mill consisted largely of improved sedimentation to capture and retain the 14 t/d of tailings that had been escaping. The improvement of river water quality was prompt, dissolved Ra^{226} concentrations dropping quickly to about 3.0 pc/l. The subsequent spring floods flushed away much of the residual sediment contamination, and after that dissolved Ra^{226} concentrations dropped below 1.0 pc/l and have since remained at that level.

Comparable improvement of river water quality has resulted at other stream locations as the milling industry installed improved methods to prevent the escape of spent ore solids. This single treatment measure has been largely responsible for the attainment of the present high degree of protection of the radioactive quality of the surface waters of the western United States.

One other aspect of the leachability of Ra^{226} from mill tailings is worthy of mention here. In various field studies it was observed that during spring flood flows river concentrations of dissolved Ra^{226} tended to rise rather than decrease as would be expected assuming a constant pollution source and greater river dilution during floods. This confusing effect was also explained once it became understood that floods scoured and transported prior riverbed accumulations of mill tailings, making them much more readily leached by the flowing water.

Observed radioactivity concentrations

From analysis of hundreds of river sediment samples, the natural Ra^{226} content of sediments from streams of the western United States has been established at an average of 1.3 pc Ra^{226}/g of dry sediment. These data are highly consistent, and very seldom range above 2.0 pc/g at unpolluted locations. This results in an extremely sensitive yardstick for monitoring purposes, and a highly sensitive indicator of the presence of uranium-mill tailings. The Ra^{226} content of uncontaminated farm topsoils is essentially the same.

In contrast to this natural level of 1.3 pc/g, concentrations of Ra^{226} as high as 2100 pc/g were observed in earlier years in one river, and as high as 800 pc/g in Animas River muds. The Ra^{226} concentration of 2100 pc/g was associated with a gross alpha radioactivity of 10 000 pc/g and with a dissolved Ra^{226} concentration of 88 pc/l in the river water.

For the past three years a surveillance network of river sediment sampling stations has been operated in the Colorado River Basin. Its purposes include long-term evaluation of the radioactivity of these sediments and the detection of accidental releases of mill tailings. This network initially included about 100 sampling locations, mostly downstream from uranium mills. Recently it has been reduced to 30 stations. Sampling is conducted twice yearly, in August and late March. Data for the past year indicate again the great improvement that has resulted from industry-wide measures to minimize pollution. At only three locations were Ra^{226} concentrations above the natural content, and at only a single location did the Ra^{226} concentration exceed 10 pc/g. At that station a content of 50 pc/g was observed most recently, in contrast to earlier concentrations as high as 400 pc/g. The present result represents residual contamination, and will decrease as floods scour it away.

Because of the very long half-life of Ra^{226}(1620 yr) and the extremely small concentrations permissible in water for public use, the ultimate fate of mill tailings in the water environment has been of interest. The floods of spring flush prior accumulations of tailings from the river beds of the tributary streams into the main Colorado River, and from there eventually the ore solids reach Lake Mead and the great reservoirs of the Lower Colorado River Basin. This is their final resting place, for practical purposes, as these great lakes are not flushed to any appreciable extent by the annual floods. Limited sampling of Lake Mead sediments has indicated that the average Ra^{226} concentration is two to three times the natural content of sediments of the upstream river basin, and very rough estimates indicate that 2000 g or more of Ra^{226} have been added to the Lake Mead sediments during the past decade or so. These are, of course, still very low Ra^{226} concentrations, and there has been no detectable increase of dissolved Ra^{226} in the waters of Lake Mead over natural levels.

Monitoring of aquatic biota [10]

Monitoring of the biological forms found in streams receiving uranium-mill wastes was begun in 1955, and since that time a large number of samples have been collected and analysed, primarily for Ra^{226}. The samples have

included various types of filamentous algae, aquatic insects such as caddis, mayfly and stonefly larvae, and fish.

Interest in monitoring of the aquatic biota has stemmed primarily from the possibility that one or another type might serve as a particularly useful indicator of uranium-mill pollution. Like all living things, the stream biota accumulate and concentrate radioactive materials to some degree, and a single sample therefore yields a cumulative story of past pollution rather than only an instantaneous evaluation of water quality. Concentrations of radioactivity in the biota also reflect primarily radioactivity that was dissolved rather than suspended in the water, and thereby provide additional useful information not obtained from instantaneous water or sediment samples. Thus, a single sample of attached filamentous algae can yield an indication of past releases, both usual and accidental, of dissolved Ra^{226} from the uranium mills. The data gathered over the past several years have provided information as to natural Ra^{226} concentrations in the biota, as well as comparative concentrations in the flowing water, sediments and biota, and these studies have led to useful conclusions as to the value of the aquatic forms as indicators of uranium-mill pollution.

Collection methods

Filamentous algae have generally been collected by hand. Most of the streams involved have been small enough to wade, and algae samples have also been composited across the stream section. During the studies of 1955 and 1956 an attempt was made to obtain "live" weights of sample by weighing the gently blotted sample immediately after collection, but it did not prove feasible to obtain reproducible weights in this manner. The only reproducible sample weight has proved to be the ashed weight, and more recently samples have been carefully rinsed in the river to free them of attached silt and debris, then preserved in formalin for subsequent analysis.

Aquatic insects have been collected by use of a square-yard or square-foot sampler, consisting essentially of a screen attached to two posts for rigidity. Rocks and stones in a one-square-yard or one-square-foot upstream bottom area are wiped free of attached insects, which are then allowed to float downstream to be caught in the screen. The trapped insects are then picked off the screen and placed in vials for subsequent weighing, counting, identification and radioassay. Earlier samples were analysed *in toto* as collected, but beginning in 1958 all insect larvae were removed from their shells before assay.

Fish have been collected less frequently than the algae and insects. They are higher in the chain of stream life and therefore do not concentrate radioactivity to as great a degree as the lower forms. Both collection and radioassay are also more difficult. Hence, fish collections have been relatively few in number and only sufficient to provide estimates of the degree of Ra^{226} accumulation and the degree to which Ra^{226} might enter the human diet by this path.

Collection of fish samples has been largely by the use of rotenone, a fish poison. Minnows and juvenile fish have been homogenized in a blender to make a paste for radioassay *in toto*. Larger fish, most often suckers, were prepared and analysed as flesh and skeleton separately.

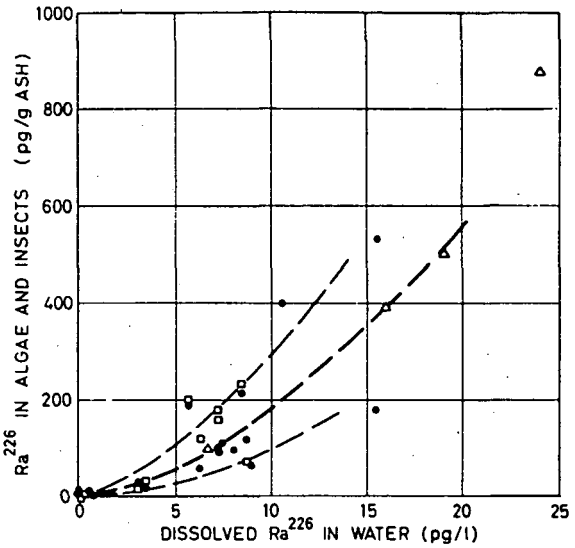

Fig. 3

Ra^{226} content of algae and insects versus water Animas River

● – Algae, 1958
□ – Insects, 1958
△ – Algae, 1959

Observed results

Detailed Ra^{226} and gross alpha concentrations observed in the stream biota have been reported elsewhere by ANDERSON [10], but a summary of these results is useful here. Fig. 3 shows the findings for algae and aquatic insects plotted as a function of the dissolved Ra^{226} content of the flowing water. All results for biological samples are reported as pc Ra^{226}/g of ash. The lines through the data have been sketched by eye.

The natural Ra^{226} content of the algae and the aquatic insects averaged about 4.0 pc Ra^{226}/g of ash, with no apparent difference for the two types of biota. At polluted locations Ra^{226} concentrations in algae as high as 3500 pc Ra^{226}/g of ash have been observed in the earlier studies [2]. Generally, the higher Ra^{226} concentrations have been associated with high Ra^{226} content of sediments as well as relatively high dissolved Ra^{226} in the flowing water. In all cases, at both polluted and clean locations, there has been a virtual one-to-one correspondence between the Ra^{226} content of algae and insects, indicating no difference in the usefulness of these forms as indicators of mill pollution. The algae are much more easily collected, however, and this decisive advantage has led to selection of the algae as the practical indicator organism.

In order to examine the relation between Ra^{226} concentrations in biota and in the associated water, a concentration factor has been defined as:

$$C = \frac{\text{pc } Ra^{226}\text{/g of live weight}}{\text{pc } Ra^{226}\text{/g of water}}$$

Factors to convert ash weights to live weights have been determined in other studies and used for these estimates of C. For both types of biota a consistent set of concentration factors between 500 and 1000 was obtained over the whole range of results.

The natural Ra^{226} content of juvenile fish analysed in toto averaged 0.5 pc Ra^{226}/g of ash, with a range of 0.1 to 0.9, against results as high as 47 pc/g in samples from polluted locations. These samples, of course, include the gut and its content of silt and sediment, hence the computation of concentration factors would be misleading. For adult fish, the data are too few to give representative average Ra^{226} concentrations in the flesh and skeleton. Flesh samples contained up to 1.4 pc Ra^{226}/g of ash, while skeleton Ra^{226} content ranged up to 8.9 pc Ra^{226}/g of ash. Concentration factors are more meaningful here, and although the data are few, the concentration factors are highly consistent. The data indicate that for sucker flesh a concentration factor of 3 is a good estimate, while a concentration factor of about 100 is associated with the skeletal material, demonstrating the bone-seeking qualities of Ra^{226}.

Chemical toxicity of mill wastes

Although not a part of the usual radioactivity monitoring system per se, studies of the populations of aquatic insects below uranium mills have yielded highly valuable information as to the chemical toxicity of the mill wastes. Fig. 4 is a graphic presentation of such studies for the Animas River [5]. As may be seen, the number of species of bottom insects and their populations were high at a location immediately upstream of the uranium mill. Within a mile after the introduction of the mill effluents, this entire bottom population was eliminated, and little improvement could be observed for the next 40 miles of river. Aside from their radioactivity, the chemically toxic quality of the mill wastes was thus clearly demonstrated. As a result, remedial treatment measures at the mill included removal of the toxic organic-bearing raffinate from the effluent, and, over a period of two years, the river-bottom insect populations gradually returned to normal numbers.

Monitoring of other media

Other media related to the problem of radioactive pollution from uranium mills have also been monitored during the foregoing studies. They include mill effluents (mentioned separately because of special sample collection problems), topsoils from farms irrigated with polluted river water, crops from such farms, and, quite recently, the fertilizers used on such farms.

Mill effluents

The effluents from uranium mills most often contain a very high concentration of tailings or suspended spent ore solids, and considerable care is necessary to assure collection of a truly representative sample. For instance, if the sample bottle is held too long in the effluent stream the solids will replace most of the liquid, and a sample of only the wet solids results.

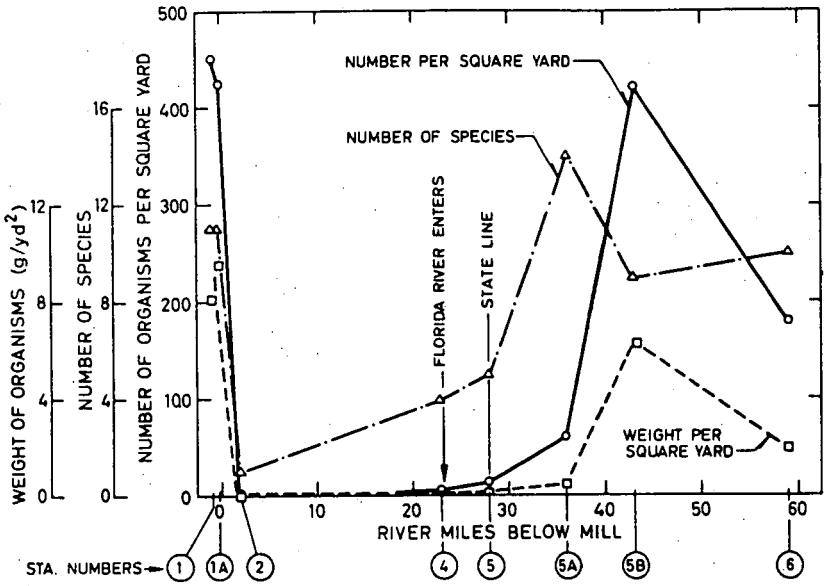

Fig. 4

Bottom fauna studies

The effluent liquors are also usually highly acid or alkaline in nature, and liquid and solid portions of the sample should be separated as soon as feasible after the sample is collected.

Dissolved Ra^{226} in the effluent liquors has ranged over 1000 pc/l in our studies, and the finer suspended spent ore solids in these effluents have often contained several thousand pc Ra^{226}/g of solids. In most cases, the receiving stream has provided relatively great dilution, hence the immediate downstream dissolved Ra^{226} concentration has been much reduced. As has been noted, the accumulation of mill tailings on the river beds has proved to be the main source of downstream dissolved Ra^{226}, and recent treatment measures at the mills have almost eliminated this problem. In addition, at locations where river dilution has been inadequate, neutralization of the effluent sometimes followed by precipitation with barite or other chemicals has been effective in reducing the dissolved Ra^{226} in the effluents.

Crops, topsoils and fertilizers

During the Animas River studies of 1958-1959 a considerable number and variety of crop samples were collected from local farms and analysed for Ra^{226}. Many were also analysed for Sr^{90} because of its bone-seeking qualities, even though it is not a constituent of uranium-mill wastes. Topsoils and irrigation water were also assayed, and since that time other limited crop assays for Ra^{226} have been conducted. Crops were generally prepared for assay in mubh the same manner as they would be prepared for human consumption. The data have been presented in detail elsewhere [5, 6].

In the Animas River case, the Ra^{226} content of crops from unpolluted locations averaged about 2.0 pc Ra^{226}/kg of live material, in comparison to about twice this figure at farms irrigating with polluted river water. The topsoils were not highly contaminated with Ra^{226}, and irrigation water Ra^{226} concentrations were usually not relatively great.

Studies at this and other locations have since indicated that the Ra^{226} content of crops is most directly related to topsoil Ra^{226} concentrations, rather than to the dissolved Ra^{226} in the irrigation water. At unpolluted farms, the topsoil Ra^{226} content has proved to be quite comparable to the natural Ra^{226} content of river sediments, or about 1.3 pc Ra^{226}/g of soil. At polluted locations, topsoil Ra^{226} has ranged as high as 40 to 50 pc/g. Quite recent studies have indicated that these higher concentrations may result in part from the use of phosphate fertilizers, and these have been found to contain Ra^{226} occasionally in excess of 30 pc/g.

Cattle feed crops, particularly alfalfa, have been found to concentrate Ra^{226} to a much greater degree than other crops. Alfalfa from uncontaminated farms contains from 10 to 15 pc Ra^{226}/kg of live material, whereas concentrations in excess of 1000 pc/kg have been observed, always in association with relatively high Ra^{226} in the topsoil. A few milk samples from cows fed on such alfalfa have also been assayed for Ra^{226}, and these were found to contain higher than normal Ra^{226} levels (concentrations of 5.0 pc Ra^{226}/l of milk have been observed).

SUMMARY AND CONCLUSIONS

Monitoring of a variety of environmental media from the vicinity of United States uranium mills has provided a consistent story of the fate of wastes released from these mills to the water environment. Natural concentrations of Ra^{226} have been established for these media and compared to observed concentrations at polluted sampling points. In recent years, the United States uranium mills have undertaken a substantial programme of pollution abatement that has sharply reduced contamination of the neighbouring environment. Presently, Ra^{226} concentrations in the surface streams are therefore substantially lower than the generally accepted limits. The uranium-milling companies, in co-operation with national and local agencies, have also undertaken a programme to eliminate the problem of accidental spills or releases of wastes normally held in the lagoons.

One further problem remains at this time, namely, the problem of long-term, or "ultimate" disposal of the mill tailings. These have accumulated at a rate in recent years of approximately 20 000 t/d. Their burden of undissolved Ra^{226} is of the order of tens of thousands of grammes. The half-life of Ra^{226} is very large (1620 yr) in terms of generations, and the permissible levels of Ra^{226} contamination of the environment are very small. Hence, it is necessary to plan well now for the very long-term confinement of the tailings.

Several solutions have been proposed for this problem. In the United States competitive economy ways are usually sought to use such waste materials, and it has thus been proposed that the mill tailings might be used as land fill

material for industrial building sites or for highway subgrade fill material. Unfortunately, aside from the possible public health hazards, the finely ground spent-ore solids are not suitable as land fill because they will not support any substantial load. Another solution to the problem of long-term disposal is to dump the tailings back into abandoned uranium mines. This, of course, involves the expense of transporting them to the mines. Even so, this may prove to be the most economical adequate solution to the problem. In the meanwhile, until the best means of long-term disposal is found, the tailings piles must be adequately confined at their present locations, and maintained in condition to prevent their dispersal to the general environment.

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