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Methods of Surveying and Monitoring Marine Radioactivity

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METHODS OF SURVEYING AND MONITORING MARINE RADIOACTIVITY

REPORT OF AN AD HOC PANEL OF EXPERTS

FOREWORD

At the United Nations Conference on the Law of the Sea (1958) concern was expressed at the possibility of pollution of the sea by radioactive materials. As a result, at its Tenth Plenary Meeting, a resolution was adopted which reads in part:

".... the International Atomic Energy Agency, in consultation with existing groups and established organs having acknowledged competence in the field of radiological protection, should pursue whatever action is necessary to assist States in controlling the discharge or release of radioactive materials in the sea, in promulgating standards and in drawing up internationally acceptable regulations to prevent pollution of the sea by radioactive materials in amounts which would adversely affect man and his marine resources."

An effective control of the radioactive pollution of the sea depends, for a part, on the availability of adequate technical methods for surveying and monitoring the sea and marine products with regard to the presence of radioactive substances.

The purpose of the present manual is to offer such methods in order to help in carrying out any international or national control measures in that field.

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I INTRODUCTION

At the United Nations Conference on the Law of the Sea (1958) concern was expressed at the possibility of pollution of the sea by radioactive materials. As a result, at its Tenth Plenary Meeting, a resolution was adopted which reads in part:

"..... the International Atomic Energy Agency, in consultation with existing groups and established organs having acknowledged competence in the field of radiological protection, should pursue whatever action is necessary to assist States in controlling the discharge or release of radioactive materials in the sea, in promulgating standards and in drawing up internationally acceptable regulations to prevent pollution of the sea by radioactive materials in amounts which would adversely affect man and his marine resources."

In response to this directive an expert panel was set up under the chairmanship of Mr. Brynnielsson of Sweden. This panel carried out studies to establish a necessary set of recommendations with the purpose of ensuring that any disposal of radioactive waste into the sea involved no unacceptable degree of hazard to man. A report on the panel's activities has been published as the International Atomic Energy Agency Safety Series No.5, "Radioactive Waste Disposal into the Sea".

It is apparent from this report that any effective control of pollution must depend on the availability of adequate methods for surveying and monitoring the sea and marine products with regard to the presence of radioactive substances. Furthermore, the need for such methods of surveying and monitoring capable of being accepted as international standards for prevention of pollution exists independently of any programme of waste disposal into the sea.

The present report is the result of a study of "Methods of Surveying and Monitoring Marine Radioactivity" carried out by an ad hoc panel under the chairmanship of Dr. C. Polvani of Italy. Members of the panel were:

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A.K. Ganguly India

J.H. Harley United States of America

Y. Miyake Japan

F. Morgan United Kingdom of Great
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P. Slizewicz

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In addition, the following persons were among those who assisted in the work of the panel at various times:

J. Alinet; W. Chipman; A. Federov; T.R. Folsom;

R. Fukai; I. Hela; H. Kautsky; F. Koczy; D. Lal;

C. Michon; P. Vaissière; V. Vouk.

The secretaries of the panel were M. Saiki and C.W.C. Tait of the International Atomic Energy Agency.

The panel's purpose was to indicate or devise to a feasible extent, suitable techniques for surveying and monitoring the marine environment with respect to radioactivity. The panel, in addition, tried to indicate areas where further study was particularly necessary and to suggest the nature of studies which might be expected to be of material assistance in achieving the desired results.

Measurement of marine radioactivity

Any survey or system of monitoring of the marine environment must result in data which can be expressed in terms of quantitative units of measurement.

Measurements of marine environment radioactivity can at present be expected to fall into two rather distinct groups. One group consists of data which best indicate the radioactive state of the marine environment and which must serve as the basis of any scientific study of the processes taking place in the sea associated with its content of radioactive substances. The collection and assembly of such data are normally carried to the stage where they provide a survey of the radioactive state of the sea or some portion of the marine environment.

The other group of measurements is chosen to give the most direct answer to questions concerning the threat to man's health. The

selection of such data is more determined by the pattern of man's use of the sea than by the significance of processes taking place within the sea itself. In this case the treatment is often carried to the point of providing a warning of hazard with respect to some possible undesired consequences. That is, the total process can be considered true monitoring. However, the two situations should be identified as survey and monitoring, respectively, since the monitoring must include survey as an integral part, or as a preliminary. Further, much of the health protection data is of a type which is used as a basis to forecast possible consequences of hypothetical situations. In such a case the data collection would probably be more accurately referred to as a survey rather than as monitoring. Although the principles underlying the selection of significant measures of radioactivity are the same for the two groups specified above, the emphasis is quite different.

The first group of data can be referred to as scientific data relating to the radioactive state of the sea and the second group as data relating to man's safe use of marine products. Possible correlations between radioactive hazard and the best scientific measures of the radioactive state of the sea are perhaps premature at this stage and must depend on an extensive collection and study of data. The purpose of this study is to establish such data collection on a sound basis. This is essential if eventual correlation is to prove valid. The achievement of this purpose involves three steps.

- (a) The selection of appropriate and meaningful characteristics for measurement:
- (b) The arrangement of suitable sampling procedures to ensure representative measurements;
- (c) The setting up of reliable assay techniques to ensure reproducible determination of the quantities selected for measurement.

Criteria to be met by scientific data relating to the radioactive state of the sea

A great deal of current scientific work on radioactivity in the sea must be of an exploratory nature. Much remains to be done before we can tell which parameters will prove to be the best measures of radioactivity for application to future problems of interest. At this time the following criteria should dominate:

(a) The measurement should be meaningful in terms of some agreed definition of radioactivity;

- (b) The measurement should be reproducible:
 - (i) With regard to the same worker;
 - (ii) With regard to different workers;
 - (iii) With regard to different but equivalent techniques.
 (It should not be critically dependent on specific apparatus or techniques.)
- (c) The measurement should be representative of some definable extent, zone or component of the marine environment; and
- (d) Long period fluctuations or trends in the quantity measured should not be masked by erratic and statistically meaningless fluctuations.

Criteria to be met by data relating to man's safe use of marine products

Any measure of the radioactivity of the marine environments or marine products which is to be useful as a measure of hazard to man must be capable of interpretation in terms of the various recommendations of the ICRP. That is, the data should be presented in terms of a situation in which the final route to man is identifiable and the data can be correlated with some resultant degree of human exposure.

The interpretation of marine data to provide recommendations for human health involves many considerations outside the domain of marine sciences. Additional guidance must therefore be sought from other groups qualified to elaborate these matters.

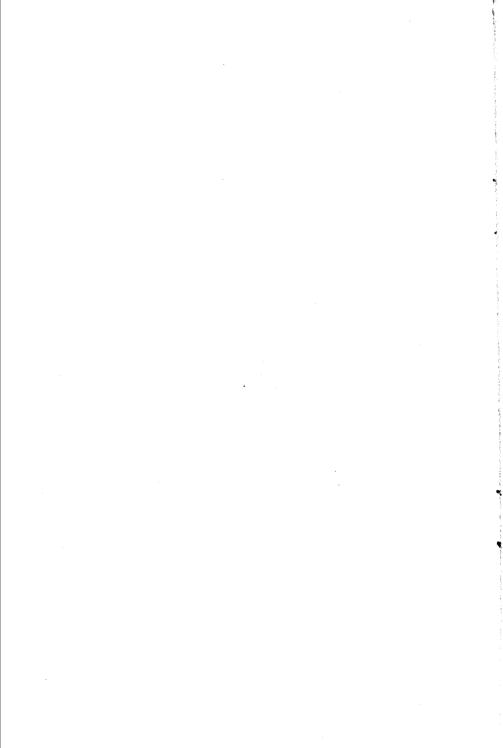
A systematic approach to measurement would commence with fundamental scientific data pertaining to the radioactive state of the marine environment. Only with an adequate understanding of the processes involved would it be desirable to develop the derived concepts such as hazard to man resulting from marine radioactivity. It is unfortunate that this ideal development of knowledge in this field is not practical. This is because of three main causes:

- (a) There is a need for an immediate answer to monitoring problems associated with the protection of man and his resources from radioactive contamination.
- (b) The available scientific staff is so small that it has to concentrate on immediate monitoring problems with resultant relative neglect of more fundamental scientific studies.

(c) The relationship between fundamental measurements of the radioactive state of the sea and measurements of applications to the protection of man is complex and even obscure. It would appear that the former study will have to reach a very high state of development before much information of direct use in monitoring can be expected.

Application of this report

In view of such considerations as those raised in the previous section, this report will be chiefly concerned with providing guidance to marine scientists engaged in survey and monitoring aimed at protecting man and marine products from adverse consequences arising from radioactive pollution. The present nature of this work requires that this report be largely directed to chemists and biologists (although hygienists and public health officials will find much in this report that will assist in the understanding of the problems of correctly monitoring marine radioactivity). This will be reflected in the presentation of material in the body of the report. Nevertheless no work in this field is likely to be sound unless based on a proper regard for physical principles basic to any measurement of radioactivity. These essential physical and mathematical considerations are therefore dealt with in appendices to the main report.



II SELECTION OF REQUIRED MEASUREMENTS AND SAMPLING

CHOICE OF CHARACTERISTIC QUANTITIES

General

The first step in any study is to decide clearly on its purpose. At present the most likely purposes are the detection of any changes in the radioactive nature of the marine environment which could pose a threat to man's interests and the expansion of basic knowledge of radioactivity in the sea. Because of the overriding need for data on which to make decisions affecting human safety and interest, an adhoc direct approach is often necessary to give immediate guidance even if it may leave much to be desired from the point of view of scientific soundness. Nevertheless the very restricted state of knowledge of radioactive processes taking place in the marine environment makes it highly desirable to take advantage of every opportunity to extend scientific knowledge.

A programme aimed at the prevention or detection of radioactive contamination significant to man's use of the marine environment could include:

- (a) A measure of radioactivity in some component of the marine environment, evaluated in terms of man's use of the component and ICRP recommendations as to the hazard of the nuclide or radiation concerned.
- (b) A measure of radioactivity resulting from some specified nuclide present in a known precursor to some component of the marine environment subject to use by man, and between which a relationship in time is known for the particular hazardous radionuclide of interest.
- (c) A measure of radioactivity for a specific nuclide in some organism showing a selective high concentration factor which will serve to indicate the presence of that nuclide when in a concentration in the environment otherwise below detection limits.
- (d) Study of dietary requirements of various populations with respect to marine products and an elementary analysis of such products.

A programme for advancing scientific knowledge could include any of the following:

- (a) A measure of the total radioactivity present, either in solution or suspension in sea-water, at significant depths;
- (b) A measure of the activity of specific nuclides present in seawater at significant depths;
- (c) A measure of the activity of specific nuclides in bottom sediments:
- (d) A measure of the activity of various nuclides in specific marine organisms;
- (e) A measure of the relative activity of various nuclides in successive members of a food chain:
- (f) A measure of the relative activity of various nuclides in different organs of organisms.

The divergence of treatment depending upon ultimate purpose is most critical at this step in which the characteristic quantities are selected for study. The subsequent steps of sampling, processing, analysis and data presentation, are subject to the same principles regardless of purpose, and difference in treatment will be largely self-evident.

Physical basis

Various physical quantities have been used in the past for reporting data on radioactivity in the marine environment. Such terms as "activity", "count-rate", "gross β count", "curies" (of unspecified substances) are frequently encountered. Serious limitations on meaningful interpretation are imposed by much of this usage. In Appendix I consideration is given to various possible physical measures of radioactivity and the implications of their use. It is apparent from the discussion in Appendix I that any serious approach to survey or monitoring directed to the protection of man is preferably based on some measure or estimate of the activity due to a specific radioactive nuclide (usually expressed as curies of a particular radionuclide). This quantity is likely to be determined either as a result of a radiochemical separation applied to the material under study followed by counting and evaluation against some standard material, or by some application of gamma spectrometry.

Certain specialized problems may require dosimetry of general external radiation in the marine environment. In such cases the appropriate physical measure would be either the ambient flux of external radiation (appropriate to fundamental scientific studies of the nature of the radiation environment) or, more probably, (in practi-

cal studies) the dose rate to some specified material expressed in rads (Appendix I).

Sea-water

The major effort in examining sea-water samples should be the determination of the concentration of specific radioactive nuclides.

In the study of hazard to man the radionuclides to be measured will be those of highest potential danger. The concentration and physico-chemical state of corresponding inactive isotopes in the sea must be considered when estimating hazard from marine sources because of the phenomenon of isotopic dilution.

In many cases biological material may be studied as a convenient indicator of radionuclides in sea-water at concentrations below those otherwise detectable. In its simplest form this practice arises from the concept that biological systems concentrate elements present in sea-water, by a constant factor dependent only on the species or organ studied. It is further presumed that the associated radionuclides remain at a constant isotopic dilution. The second assumption appears generally sound but the first is of very uncertain validity. Unless such concentration factors are well established. or there is adequate experimental data on the constancy of the isotopic dilution factor, this technique for estimating radionuclide concentration in the original sea-water is of very limited value. It may be possible, however, to set a lower limit to a particular concentration factor and thereby to state that the concentration of some radioisotope is below a certain limiting value.

In situ measurements of ambient radiation flux or dose rate have some application to monitoring of sea areas which may become contaminated as the result of major accidents or contamination incidents. Low levels of contamination will be difficult to detect by standard monitoring devices because of the background due to the natural radioactivity of potassium which, in turn, will be directly proportional to the salinity. The interfering natural radioactive nuclides are tabulated in Appendix II. Further, external radiation levels which do not double the natural background will be of little direct concern in any monitoring programme. When required, the dose rate can be measured with standard sensing devices either immersed in the water or suspended at a specified height in the air above the surface.

General dosimetry carried out in situ should be based on measurements by sensing elements with a response proportional to

energy down to low radiation levels. Such measurements will make no distinction between radioactive matter present in true solution or in suspension either as non-living matter or as small biota. Radiation measurements carried out in the air over the sea surface will be characteristic of the radiation level in the sea and the precise height at which the measurement takes place. However, the dose rate decreases rapidly with height, particularly with regard to the beta-radiation component which will be undetectable by aircraft at any feasible operating height.

The gross count or gross beta count of isolated sea-water samples is of doubtful value and may do more harm than good by suggesting that useful data on radioactivity is being collected, thereby reducing the incentive and effort to obtain other and more meaningful measures. In general there seems little point in obtaining such data unless information can thereby be obtained about specific nuclides.

Certain difficulties occur with respect to studying the distribution of radioactivity in sea-water largely because of the difficulty in providing an entirely satisfactory definition of sea-water. It is often considered necessary to separate soluble components and suspended matter. However, in view of the possibility of mobility between phases and the uncertainty of the efficiency of any such separation, it is suggested that this practice is only really justified if it assists further radiochemical separation, or if one is concerned with some particular phenomena which are dependent on particle size or phase distribution. Possibly, it would be more important in many cases to obtain knowledge of the chemical form in which a particular radionuclide appeared.

If practicable, there are advantages in separating particulates if it is known that the activity is confined to this phase, especially as this separation leads to the rejection of the bulk of the interfering potassium-40.

In addition to the application to monitoring and the direct protection of man and his marine resources, an examination of the radioactivity distribution in sea-water can prove to be of considerable scientific interest. Again, a determination of specific nuclides would be the preferred normal procedure. When one is concerned with broadly increasing scientific knowledge the preferred nuclides would normally be those most prevalent in the samples. However, certain special studies might direct attention to other nuclides. A study of the distribution between various effective reservoirs of substances held in the sea calls for the measurement of radionuclides

of half-lives commensurate with expected residence or turnover times. In the case of major sites of accumulation an examination of certain natural radionuclides is helpful. In other cases nuclides present at very low levels may prove useful investigational tools because of the existence of particularly sensitive or selective measurement methods.

In situ measurements of dose rate may also prove useful. Should it be desirable to experimentally study vertical mixing processes, measurements at various depths, following a rapid introduction of radioactive material at the sea surface, could be of interest. It is possible that in situ sensors will be devised which will be selective for radiation from particular nuclides of interest. These could be widely applied for measuring levels of specific nuclides at concentrations well below that of the interfering natural nuclides.

An examination of sea-salt samples will give information concerning any contained hazards connected with its use by man, and will also conveniently provide a concentrated sample of sea-water. However, attention must be paid to the technology of its production whereby selective crystallization or rejection of certain components is brought about. Both the salt and the rejected brines may be valuable for determining specific radionuclides with well-understood physical-chemical associations.

Sea boundaries - bottom sediments and the sea shore

Radioactive material may be present on the sea floor in the form of sediments. It may prove desirable to examine samples of bottom sediments to determine the concentration of specific nuclides which are taken up by benthos and sessile organisms. On the other hand the possibility of fishing gear and cables etc. being contaminated with bottom sediments may make it desirable to directly measure the external dose rate from such material likely to be recovered from the sea. In addition, in areas of high local contamination the external radiation to which persons using the seashore might be exposed may require in situ dose-rate determinations. Intensive contamination likely to directly affect benthic organisms by external radiation would also call for in situ dose-rate measurements. The distribution of radionuclides with depth within the sediment layer would be scientifically interesting as a source of information on the history and nature of sedimentation processes.