IAEA TECDOC SERIES

IAEA-TECDOC-1760

Use of Radiotracers to Study Surface Water Processes

IAEA
International Atomic Energy Agency

USE OF RADIOTRACERS TO STUDY SURFACE WATER PROCESSES

The following States are Members of the International Atomic Energy Agency:

AFGHANISTAN
ALBANIA
ALGERIA
ANGOLA
ARGENTINA
ARMENIA
AUSTRALIA
AUSTRIA
AZERBAIJAN
BAHAMAS
BAHRAIN
BANGLADESH
BELARUS
BELGIUM
BELIZE
BENIN
BOLIVIA
BOSNIA AND HERZEGOVINA
BOTSWANA
BRAZIL
BRUNEI DARUSSALAM
BULGARIA
BURKINA FASO
BURUNDI
CAMBODIA
CAMEROON
CANADA
CENTRAL AFRICAN REPUBLIC
CHAD
CHILE
CHINA
COLOMBIA
CONGO
COSTA RICA
CÔTE D'IVOIRE
CROATIA
CUBA
CYPRUS
CZECH REPUBLIC
DEMOCRATIC REPUBLIC OF THE CONGO
DENMARK
DOMINICA
DOMINICAN REPUBLIC
ECUADOR
EGYPT
EL SALVADOR
ERITREA
ESTONIA
ETHIOPIA
FIJI
FINLAND
FRANCE
GABON
GEORGIA
GERMANY
GHANA
GREECE
GUATEMALA
GUYANA
HAITI
HOLY SEE
HONDURAS
HUNGARY
ICELAND
INDIA
INDONESIA
IRAN, ISLAMIC REPUBLIC OF
IRAQ
IRELAND
ISRAEL
ITALY
JAMAICA
JAPAN
JORDAN
KAZAKHSTAN
KENYA
KOREA, REPUBLIC OF
KUWAIT
KYRGYZSTAN
LAO PEOPLE'S DEMOCRATIC REPUBLIC
LATVIA
LEBANON
LESOTHO
LIBERIA
LIBYA
LIECHTENSTEIN
LITHUANIA
LUXEMBOURG
MADAGASCAR
MALAWI
MALAYSIA
MALI
MALTA
MARSHALL ISLANDS
MAURITANIA, ISLAMIC REPUBLIC OF
MAURITIUS
MEXICO
MONACO
MONGOLIA
MONTENEGRO
MOROCCO
MOZAMBIQUE
MYANMAR
NAMIBIA
NEPAL
NETHERLANDS
NEW ZEALAND
NICARAGUA
NIGER
NIGERIA
NORWAY
OMAN
PAKISTAN
PALAU
PANAMA
PAPUA NEW GUINEA
PARAGUAY
PERU
PHILIPPINES
POLAND
PORTUGAL
QATAR
REPUBLIC OF MOLDOVA
ROMANIA
RUSSIAN FEDERATION
RWANDA
SAN MARINO
SAUDI ARABIA
SENEGAL
SERBIA
SEYCHELLES
SIERRA LEONE
SINGAPORE
SLOVAKIA
SLOVENIA
SOUTH AFRICA
SPAIN
SRI LANKA
SUDAN
SWAZILAND
SWEDEN
SWITZERLAND
SYRIAN ARAB REPUBLIC
TAJIKISTAN
THAILAND
THE FORMER YUGOSLAV REPUBLIC OF MACEDONIA
TOGO
TRINIDAD AND TOBAGO
TUNISIA
TURKEY
UGANDA
UKRAINE
UNITED ARAB EMIRATES
UNITED KINGDOM OF GREAT BRITAIN AND NORTHERN IRELAND
UNITED REPUBLIC OF TANZANIA
UNITED STATES OF AMERICA
URUGUAY
UZBEKISTAN
VENEZUELA, BOLIVARIAN REPUBLIC OF
VIET NAM
YEMEN
ZAMBIA
ZIMBABWE

The Agency's Statute was approved on 23 October 1956 by the Conference on the Statute of the IAEA held at United Nations Headquarters, New York; it entered into force on 29 July 1957. The Headquarters of the Agency are situated in Vienna. Its principal objective is "to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world''.

IAEA-TECDOC-1760

USE OF RADIOTRACERS TO STUDY SURFACE WATER PROCESSES

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2015

COPYRIGHT NOTICE

For further information on this publication, please contact:

Terrestrial Environment Laboratory, Seibersdorf
International Atomic Energy Agency
2444 Seibersdorf, Austria
Email: Official.Mail@iaea.org

Printed by the IAEA in Austria
March 2015

IAEA Library Cataloguing in Publication Data

Use of radiotracers to study surface water processes.
— Vienna : International Atomic Energy Agency, 2015.
p. ; 30 cm. — (IAEA-TECDOC series, ISSN 1011–4289 ; no. 1760)
ISBN 978–92–0–100415–4
Includes bibliographical references.

1. Radioactive tracers. 2. Radioisotopes in hydrology. 3. Radioactive tracers in water pollution research. I. International Atomic Energy Agency. II. Series.

IAEAL 15–00956

FOREWORD

Most of world's population is directly affected by the available water resources and the means to supply them. All natural or anthropogenic processes that modify the water flux and quality have a direct impact on human lives. Radioactive tracers can be extremely useful in studying such processes, and thus help to investigate the common problems faced by many States and to find adequate solutions. Problems such as water pollution, erosion, river sedimentation, and the loss of storage capability of water reservoirs, thereby reducing fish stocks, can cause great damage and negatively affect the well-being of local populations.

To address this, a consultants meeting on the use of radioactive tracers to study surface water processes was organized by the IAEA to support Member States in situations where the use of radioactive tracers is considered as a major tool for a complete understanding of the process.

This publication represents a sound knowledge base for the conduct of radiotracer studies in the environment, with papers on radiotracer methodology, radiation protection and regulation, data analysis and modelling. Environmental case histories from five Member States — Australia, Brazil, France, the Republic of Korea and Sweden — provide information on conducting studies involving the use of radioactive tracers. These case histories are not meant as guidelines for preparing a field study but can rather serve as examples of the type, caution and extent of work involved in environmental studies using radiotracers.

This publication can provide guidance for conducting potential future training events in the use of radioactive tracers in the environment and can serve as a key reference to all concerned directly or indirectly with surface water processes.

The IAEA wishes to thank all the participants in the consultants meeting for their valuable contribution, in particular C.E. Hughes (Australia) and P. Brisset (France). The IAEA officer responsible for this publication was A. Ceccatelli of the IAEA Terrestrial Environment Laboratory.

EDITORIAL NOTE

This publication has been prepared from the original material as submitted by the contributors and has not been edited by the editorial staff of the IAEA. The views expressed remain the responsibility of the contributors and do not necessarily represent the views of the IAEA or its Member States.

Neither the IAEA nor its Member States assume any responsibility for consequences which may arise from the use of this publication. This publication does not address questions of responsibility, legal or otherwise, for acts or omissions on the part of any person.

The use of particular designations of countries or territories does not imply any judgement by the publisher, the IAEA, as to the legal status of such countries or territories, of their authorities and institutions or of the delimitation of their boundaries.

The mention of names of specific companies or products (whether or not indicated as registered) does not imply any intention to infringe proprietary rights, nor should it be construed as an endorsement or recommendation on the part of the IAEA.

The IAEA has no responsibility for the persistence or accuracy of URLs for external or third party Internet web sites referred to in this publication and does not guarantee that any content on such web sites is, or will remain, accurate or appropriate.

CONTENTS

SUMMARY

1. INTRODUCTION

1.1. Background and Scope

The use of artificial radiotracers in the environment can be considered analogous to their use in the human body (i.e. diagnostic applications in medical field). In both domains they provide information on flow rates and pathways, on chemical exchange with materials and on their final discharge from the studied system. Artificially injected, radiotracers have been used to investigate flow in natural waters since the 1950's. Identification of flow paths, diagnosis of blockages or leakage, measurement of flow rates, dispersion and exchange processes, such as biological uptake or sorption, are all aspects to which a wide range of artificial radioactive tracers have been applied.

Methods for the use of radiotracers in aquatic environments have been established over the past five decades. The basic principles involve definition of the system or problem to be investigated, selection of a suitable tracer, design of tracer deployment and measurement systems and analysis of the data collected to address the initial problem. These aspects are described in detail in this publication, covering the current state of the art in tracer technology including new developments in the use of nano-particle tracers.

The translation of raw tracer data into parameters that represent the physico-chemical behaviour of the studied component is a common approach to understand and generalize results from a tracer study. Aspects of tracer data interpretation include tracer mass balance, travel time distribution, transport parameters, decoupling of tracer and water flows, modelling flow and non-conservative tracers and how to determine model parameters from tracer data.

Increasingly radiotracers are used in combination with numerical modelling to improve confidence in the predictive capacity of models used in the management of our water resources and to extend their spatial applicability. In turn this allows us to use less and less tracer and demonstrate that the human and environmental impact of modern radiotracer studies is minimal.

The use of radioactive tracers in the environment is subject to national regulation as well as international standards and guidelines. These address aspects of radiation protection during transport, deployment and in the environment following deployment and are relevant not only to radiation workers and members of the public, but also to non-human biota in the receiving environment. This publication outlines the regulatory framework and safety aspects relating to radiotracing and provides examples of how to model radiation dose from exposure to radiotracers in the environment for humans and, for the first time, non-human biota.

1.2. Objectives

In the scientific literature there are large numbers of publications on medical applications of radiotracers and some of lab-based ecological radiotracer studies. In the hydrological field there has been a recent growth in the use of stable isotopes and naturally occurring

radiotracers such as ^{14}C, ^{222}Rn, the series of U and Th, and fission products. However, there are few new publications about field applications of radiotracers to study hydrological or ecological processes in the environment. This reflects both a decrease in such studies but also, the fact that in many cases these studies remain in the unpublished literature as commercial reports.

This publication seeks to summarise the current status of radiotracer applications in surface water environments as well as addressing challenges to the use of radiotracers in the environment, such as:

- Dose assessment;
- Public perception, regulatory status and gaps.

Dose assessment: It should be highlighted that the present publication deals also with environmental assessment of radiotracer studies in water environment through some case studies. It is, in fact, no longer acceptable to consider only human dose and safety in designing radiotracer studies. However, guidelines for environmental dose assessment for radiotracing have not been established or even discussed at an expert level up to now. This report aims to raise the awareness within the regulatory agencies of the likely environmental implications of the use of radioactive tracers in the study of surface water processes.

Public perception, regulatory status and gaps: Data and studies on the environmental and human impact of using intentionally released radiotracers for hydrologic studies have rarely been published. Public and regulatory concern in the absence of such data, has led to guidelines in many countries which have the effect of reducing their applications. The present publication attempts to provide some such data in order to make the regulatory and safety basis for application of this powerful tool clearer for the future.

It is informative to compare the application of radiotracers in the environment, to their use in nuclear medicine. Nuclear medicine plays a crucial role in the diagnosis and/or treatment of illness. The technetium meta-stable (^{99m}Tc), eluted from $^{99}Mo/^{99m}Tc$ generators as pertechnetate (TcO_4^-) is the most commonly used radiotracers in nuclear medicine (>70%).

Environmental applications involve much lower concentrations of radiotracer than would be found in the human body during medical diagnostic studies. This is principally because large environmental detectors may be fully immersed in the water (4π geometry), increasing enormously the counting efficiency.

Just as the medical use of radiotracers is accepted by the public opinion and by the authorities for the benefits to the health of the patient, so their environmental applications should be accepted for the health of ecosystems on which humanity depends.

This publication aims to realistically address public and regulatory concerns about health risks and thereby reduce inhibitions to the use of artificial radiotracers in hydrology. Hopefully, this powerful tool for environmental studies will be more accepted in the near future.

1.3. Structure of the publication

The publication is structured in individual papers to specifically cover all the main aspects of the use of radiotracers for studying environmental water processes, including methodology, radiation protection and regulations, data interpretation and modelling, future trends. Case

studies are described to demonstrate how radiotracers can be successfully used in the environment in addressing water resources, contaminant transport and coastal management issues. In particular, ten case studies are presented from Korea, France, Brazil, Hong Kong, Australia, Belgium and Sweden using a variety of radiotracers including ^{99m}Tc, ^{198}Au, ^{3}H, ^{82}Br, ^{32}P, $^{175+181}$Hf, ^{160}Tb, ^{51}Cr(III), ^{65}Zn, ^{54}Mn and ^{35}S. These studies address physical transport processes such as dispersion and mixing, reactive transport and adsorption and contaminant uptake. Traced components include water, effluent, nutrients, contaminants and mud in rivers, lakes, wetlands and coastal waters.

PAPERS ON SURFACE WATER PROCESSES STUDY USING RADIOACTIVE TRACERS

ARTIFICIAL RADIOTRACER APPLICATIONS IN AQUATIC ENVIRONMENTS

J.V. BANDEIRA[*], C.E. HUGHES[**]

[*]Brazilian Nuclear Energy Commission, CNEN/CDTN, Brazil
[**]Australian Nuclear Science and Technology Organisation, ANSTO, Australia

Abstract

The use of artificial radiotracers in the environment is analogous to their use in the human body. In both domains they provide data on flow rates and pathways, on exchange with materials and on their final discharge from the studied system. Artificially injected radiotracers have been used to investigate flow in natural waters since the 1950's. Identification of flow paths, diagnosis of blockages or leakage, measurement of flow rates, dispersion and exchange processes, such as biological uptake or sorption, are all aspects to which a wide range of artificial radioactive tracers have been applied. Increasingly radiotracers are used in combination with numerical modelling to improve confidence in the predictive capacity of models used in the management of our water resources and to extend their spatial applicability. In turn this allows us to use less and less tracer and demonstrate that the human and environmental impact of modern radiotracer studies is minimal.

1. INTRODUCTION

Water sustains life and is therefore crucial to the health of the environment. With the increased human pressure on the natural world, water resources are being depleted and polluted. There is a pressing need for greater understanding of environmental processes to underpin the design of mitigation and remediation strategies.

Mathematical modelling backed up by field measurements are the basis of modern environmental hydraulic, pollutant and sediment transport studies. These measurements could include tracer experiments, which are used to study both surface and groundwater processes. Intentionally released tracers are known as artificial tracers.

Radioactive and non-radioactive tracers are important tools for investigating and quantifying surface water processes. They are able to integrate, over limited domains of space and time, the effects of all hydrodynamic agents that have acted upon the water itself, the pollutants and the sediments transported by it. Applications include the study of dispersion and advection characteristics, and flow rates. Tracers can also be used to study the rates and pathways for reactive and biologically mediated transport of contaminants.

Hydrogeological applications of tracers include measurements of: flow, velocity, interconnection of aquifers, leakage from reservoirs, total and effective porosities of water-bearing strata, the dispersion and sorption characteristics of solutes and pollutants and the location of water-transmitting fissures in boreholes [1–14]. However, the main focus of this paper will be the use of artificial radioactive tracers to study surface water processes.

The radioactive tracers are particularly useful for the in situ measurement of sediment movement [15]. There is an extensive literature [for example 16–40] on studies of:

- Sand, transported mainly on - or very near - the bottom as bed load, with velocities much lower than the ones of the water driving it; and
- Fine sediment and sludge transported in suspension with velocities similar to the ones of the driving water.

Crucial to the success of a transport study is the optimum choice of the intentionally applied tracers. A wide range of options exist [11, 35, 41–53] including:

(a) Radioactive (e.g. ^{198}Au; ^{82}Br; ^{99m}Tc in the form of TcO_4^- for water labelling [46] or in the form of $TcO(OH)_2 + 2H^+$ for fine sediment labelling [28-31]);
(b) Fluorescent (e.g. Rhodamine WT; Rhodamine B; Uranine; Fluorescein);
(c) Salt (e.g. NaCl; $Na_2Cr_2O_7$; NaI; $NaNO_2$; LiCl; KCl; KBr, $MnSO_4$).

Radioactive discharges or leachate from radioactive waste, already present in the environment, have also been used as tracers [30, 54–62].
There are fewer examples where intentionally released radiotracers have been used to study the biological uptake of contaminants or their interactions with particulates at the field scale; however, this is a particularly powerful application [63–66].
The behaviour of the environment has some analogies to that of a living being. Changes in morphology, flows and natural or anthropogenic contaminant inputs influence the health of the aquatic environment. Historical data, field measurements, water and sediment assays as well as hydraulic and mathematical modelling all provide important knowledge of the health of the environment. However, as in nuclear medicine, tracer studies of the dynamics of the environmental processes often provide the final diagnosis allowing the maintenance or return to health of the 'environmental being' through protection or remediation. The fate and behaviour of contaminants and nutrients in the aquatic ecosystem has been effectively understood by using radiotracers of heavy metals, water and nutrients to measure the time scale of uptake and transfer between water, sediments and biological compartments in fresh water ecosystems [63–67].
Radiotracers represent a powerful tool for the future understanding of the influence of the turbulence, shear stress, dead zones and other geometrical characteristics of the hydraulic flow in the transport and deposition of contaminants and fine sediments in rivers, estuaries and coastal environments. The study of fine sediment in suspension is very important as it represents about 90% of sediment transport in the rivers and is often the carrier of pollutants (e.g. heavy metals adsorbed to them) and also of nutrients and organic matter that are fundamental for the aquatic biota. In a suitable chemical (electropositive) form radiotracers can label mud, which is electronegative, and can be used to study the dynamics of fine sediments in suspension [28, 29, 31].

In summary, the success of any radiotracer investigation depends on a precise definition of the question to be addressed, a good knowledge of the system's characteristics and of the adequateness and limitations of the employed tracers.

2. MATHEMATICAL MODELLING

The tremendous improvements in computer science and computer capacity in the last two decades is enabling the development of mathematical models that can incorporate and process an almost unlimited set of parameters. Moreover, through the variation of these parameters, the models are able to produce a series of results from which the most congruent output to observed results can be selected. In this way mathematical modelling is being more and more used.
The measurements mentioned above may be applied to mathematical and/or physical modelling. In some cases the tracer experiments can be designed and optimized (e.g. reduce the number or size of tracer injections) by running the models prior to the experiment, in order to choose '*locations where the model output is particularly sensitive to the choice of input parameters*' [33].
The mathematical modelling can integrate hydrodynamic behaviour over domains of space and time, far greater than those appropriate to tracer measurements. Thus, mathematical modelling and tracer measurements are complementary. So, tracer measurements could be used as an important tool for calibrating and/or validating mathematical models. It is evident that a model which is able to present results confirmed by direct measurements in the prototype will be regarded with an increased degree of confidence by the user. In this way it is

very important that the tracer groups work in close collaboration with hydraulic and environmental institutions, in an interdisciplinary way.

3. CHALLENGES TO THE USE OF RADIOTRACERS IN THE ENVIRONMENT

In the scientific literature there are large numbers of publications on medical applications of radiotracers and some of lab-based ecological radiotracer studies. In the hydrological field there has been a recent growth in the use of stable isotopes and naturally occurring radiotracers such as ^{14}C, ^{222}Rn, the series of U & Th and fission products. However, there are few new publications about field applications of radiotracers to study hydrological or ecological processes in the environment. This reflects both a decrease in such studies but also, the fact that in many cases these studies remain in the unpublished literature as commercial reports. This publication seeks to summarize the current status of radiotracer applications in surface water environments as well as addressing challenges to the use of radiotracers in the environment, such as:

- Dose assessment;
- Public perception, regulatory status and gaps.

Dose assessment: It should be highlighted that the present report deals also with environmental assessment of radiotracer studies in water environment through some case studies. It is no longer acceptable to consider only human dose and safety in designing radiotracer studies. However, guidelines for environmental dose assessment for radiotracing have not been established or even discussed at an expert level up to now. This report aims to raise the awareness within the regulatory agencies of the likely environmental implications of the use of radioactive tracers in the study of surface water processes.

Public opinion, regulatory status and gaps: Data and studies on the environmental and human impact of using intentionally released radiotracers for hydrologic studies have rarely been published. Public and regulatory concern in the absence of such data, has led to guidelines in many countries which have the effect of reducing their applications. The present publication attempts to provide some such data in order to make the regulatory and safety basis for application of this powerful tool clearer for the future.

The following three quotes from a recent book on tracers in hydrology [52] demonstrate the prevailing attitudes within the scientific community that the regulatory hurdles facing the use of radiotracers are virtually insurmountable:

> '*Radioactive tracers were used frequently to solve hydrological problems in the past, under the guidance of International Energy Agency (IAEA). Nowadays radioactive substances may be applied to hydrological subsystems as artificial tracers only upon receipt of special permission from a local Atomic Energy Commission*'.
>
> '*The potential health and environment risks posed by radioactive substances mean that the use of artificially applied radioactive tracers is very limited nowadays….*'
>
> '*The advantages of radioactive tracers are that they are very sensitive and offer selective detection, the disappearance of the tracer from the system due to decay, and the ability to follow the flowpath of water and tracer using a Geiger counter. The disadvantages are the potential health risks of some tracers, the very high costs of measurement and materials, and the fact that it is unlikely that a permit will be granted*

for the tracer test. The disadvantages generally hinder the application of radioactive tracers in countries with modern environmental legal standards. Even so, they are very suitable for studies addressing specific problems and they disappear rapidly after their half-period has expired'.

It is informative to compare the application of radiotracers in the environment, to their use in nuclear medicine. Nuclear medicine plays a crucial role in the diagnosis and/or treatment of illness [68]. The technetium meta-stable (^{99m}Tc), eluted from ^{99}Mo/^{99m}Tc generators as pertechnetate (TcO_4^-) is the most commonly used radiotracers in nuclear medicine (>70%).
Environmental applications involve much lower concentrations of radiotracer than would be found in the human body during medical diagnostic studies. For example, studies performed at Paraopeba River [28] and Velhas River [31] used concentrations of ^{99m}Tc (Bq/mL of water) that were seven orders of magnitude lower than those typically used in nuclear medicine (Bq/mL of blood). This is principally because environmental detectors may be fully immersed in the water (4π geometry), increasing enormously the counting efficiency.
Just as the medical use of radiotracers is accepted by the public opinion and by the authorities for the benefits to the health of the patient [68], so their environmental applications should be accepted for the health of ecosystems on which humanity depends.

This paper aims to realistically address public and regulatory concerns about health risks and thereby reduce inhibitions to the use of artificial radiotracers in hydrology. Hopefully, this powerful tool for environmental studies will be more accepted in the near future.

REFERENCES

[1] PLATA BEDMAR, A., Isótopos en Hidrología, Madrid, Alhambra (1972).

[2] INTERNATIONAL ATOMIC ENERGY AGENCY, Isotope Techniques in the Hydrogeological Assessment of Potential Sites for the Disposal of High-Level Radioactive Wastes, Technical Reports Series No. 228, IAEA, Vienna (1983).

[3] GASPAR, E., Modern Trends in Tracer Hydrology – Volume II, CRC Press. ISBN: 0-8493-4321-6, (1987b).

[4] PLATA BEDMAR, A., Detection of leaks from reservoir and lakes, In: Use of Artificial Tracers in Hydrology, IAEA-TECDOC-601, 71-130, IAEA, Vienna (1991).

[5] HÖTZL, H., WERNER, A., (Eds), Tracer Hydrology. In Proceedings of The 6th International Symposium on Water Tracing, Kalsruhe, Germany, 21-26 September, 1992,Balkema/Rotterdam/Brookfield, ISBN: 90-5410-084-2 (1992).

[6] HULLA, J.; RAVINGER, R.; TURCEK, P., Artificial radio-tracers in groundwater flow measurements. In: Groundwater monitoring and management, Selected contributions to an International symposium at Dresden (Germany). 23-28 Mar 1987, IAHS Publication. No. 173 ISBN 0-947571-51-5, JONES, G.P. University Coll., London (Ed.) 484 (1990) 131-138.

[7] KÄSS, W., Tracing Techniques in Geohydrology, Publisher A.A. BALKEMA, Rotterdam, Netherlands, ISBN: 90-5410-444-9 (1998).

[8] PLATA BEDMAR, A., Manual de Fugas en Embalses, Madrid, CEDEX (1999).

[9] PLATA BEDMAR, A., ARAGUÁS, L. A., Detection and Prevention of Leaks from Dams, Publisher A.A. BALKEMA, Netherlands, ISBN 90-5809-355-7 (2002).

[10] HUGHES, C.E., STONE, D.J.M., ACWORTH, R.I., A radioisotope tracer study of estuarine groundwater movement on the eastern Australian coast, Isotope Hydrology and Integrated Water Resource Management – IAEA Conference 17 – 23 May 2003, Vienna, (2003).

[11] SINGH, B.P., KUMAR, B., Isotopes in Hydrology, Hydrogeology, and Water Resources, NAROSA Publishing House, New Delhi (2005).

[12] BIANCHIN, M., SMITH, L., BARKER, J. F., BECKIE, R., Anaerobic degradation of naphthalene in a fluvial aquifer: A radiotracer study, Journal of Contaminant Hydrology **84** (2006) 178–196.

[13] HUGHES, C.E., CRAWFORD, J., ACWORTH, R.I., A radiotracer technique to track tidally induced groundwater movement in 3D. In: 4th International Conference on Tracers and Tracing Methods – TRACER 4, Autrans/Grenoble, France (2006a).

[14] ACWORTH, R. I., HUGHES, C. E., TURNER, I. L., A radioisotope tracer investigation to determine the direction of groundwater movement adjacent to a tidal creek during spring and neap tides, Hydrogeology Journal **15** (2007) 281–296.

[15] INTERNATIONAL ATOMIC ENERGY AGENCY, Radiotracer and sealed source techniques for sediment management, Report of the CM, held in Vienna, Austria, 21-25 April 2008.

[16] COURTOIS, G., Possibilité d'un nombre limité de grains radioactifs dans les etudes quantitatives de mouvements de sédiments, International Journal of Applied Radiation and Isotopes, (IJARI), **15** (1964) 655-663

[17] COURTOIS, G., SAUZAY, G., Les masses de sédiments à injecter dans une expérience de traceurs radioactifs en sedimentology dynamique, Isotope Hydrology - AIEA SM 129/36 (1970) 581-616

[18] SAUZAY, G., Méthode du bilan des taux de comptage d'indicateurs radioactifs pour la détermination du débit de charriage de litssableux. Thèse, Université de Toulouse, 20 Novembre (1967).

[19] Anonymous, L'application des radioéléments en sedimentology, La Houille Blanche **7**, **8** (1970)
[20] INTERNATIONAL ATOMIC ENERGY AGENCY, Tracer Techniques in Sediment Transport, Technical Reports Series No. 145, Vienna: IAEA (1973).
[21] TOLA, F., The use of radioactive tracers in dynamic sedimentology. In: IAEA Regional Seminar on the Use of Isotope Techniques in Water Resources Development, Athens. Proceedings, IAEA, Vienna, (1981).
[22] WILSON G., Étude du transport et de la dispersion des sédiments en tant que processus aléatoire. Thèse doctorat état, Paris (1987).
[23] CRICKMORE, M.J., TAZIOLI, G.S., APPLEBY, P.G., OLDFIELD, F., The use of nuclear techniques in sediment transport and sedimentation problems, Technical Documents in Hydrology, International Hydrological Programme, IHP-III Project 5.2. UNESCO – Paris (1990).
[24] BECK, C., et al., Sand bodies and sand transport paths at the English Channel-North Sea border: morphology, hydrodynamics and radioactive tracing, Océanologicaacta **11** (1991) 111-121.
[25] ROMANA, L.A., et al., Use of radioactive tracing techniques to identify particulate deposits and biological effects of urban effluents discharges from sewage outfalls, Water Science and Technology **25** 12 (1992) 115-122.
[26] PRITCHARD, T.R., LEE, R.S., DAVISON, A., Sydney Deep Ocean Outfalls; In situ Observations of Plume Characteristics. In Proceedings of the Eleventh Conference on Coasts and Ocean Engineering, Townsville (1993) 53-58.
[27] AUN, P.E.; BANDEIRA, J.V., The role of nuclear techniques in sedimentological studies and some applications in Latin America. Proceedings of an Advisory Group meeting held in Vienna, 26-29 April 1993. In: International Atomic Energy Agency. Use of nuclear techniques in studying soil erosion and siltation (IAEA-TECDOC-828),Vienna (1995) 29-97.
[28] BANDEIRA, J.V., PINTO, G.G., SABINO, C.S., AGUDO, E.G., The use of ^{99m}Tc as an adsorbable tracer for studying the dynamics of fine sediments in suspension. In: Proceedings of the International Symposium on Isotope Techniques In Water Resources Development and Management, 10-14 May 1999 Vienna: IAEA (1999) 1 CD-ROM.
[29] BANDEIRA, J.V., at al., Development of a technique for using ^{99m}Tc as an adsorbable tracer for hydrodynamic studies of fine sediments in suspension, Journal of Applied Radiation and Isotopes,.**57** (2002) 85-92.
[30] BANDEIRA, J. V. et al., The use of tritium content as an indicator of the groundwater contamination by sanitary landfills leachates in the region of Belo Horizonte City, Brazil. Water Science and Technology, **57** (2008) 1915.
[31] BANDEIRA, J.V., Development of Nuclear and Correlate Techniques for Urban Hydrology Studies - Applications in Pampulha Hydrographic Basin and Velhas River, MG. Doctorate Thesis in Sanitation, Environment and Hydric Resources – Federal University of Minas Gerais - UFMG, Belo Horizonte, Brazil. (2004)(In Portuguese).
[32] NIELSEN, A.F. et al**.,** Field verifications of formulations for sand transport under wave and current action, In: Proceedings of the Coasts and Ports Conference, Surfers Paradise, Qld., September (2001) 481–486.
[33] AIREY, P., HUGHES, C., KLUSS, T., DURAN, E., MILLER, B., CHIUENTA, S., NIELSEN, A.F., HOLLINS, S., The evolving role of radiotracers in coastal zone studies, Applied Radiation and Isotopes, **58** (2003) 401-406.
[34] INTERNATIONAL ATOMIC ENERGY AGENCY, Fluvial sediment transport: Analytical techniques for measuring sediment load, IAEA-TECDOC-1461, Vienna: (2005).

[35] HUGHES, C.E., et al., Field scale coastal zone radiotracer applications conducted under IAEA/RCA projects. In: 4th International Conference on Tracers and Tracing Methods – TRACER 4, Autrans/Grenoble, France (2006b).
[36] JUNG, S.H., Kim, J.B., KIM, J.S., Sediment Transport Study at Haeundae Beach in Korea by using a ^{192}Ir Labeled Compound. In: 4th International Conference on Tracers and Tracing Methods – TRACER 4, Autrans/Grenoble, France (2006).
[37] JUNG, S.H., et al., Investigation into the dispersion coefficients for natural rivers by using a radiotracer method, Journal of Nuclear Science and Technology (2008).
[38] SAVELLI CIATTEO, R., et al., Management of Sediments throughout the Navigation Channel of the Orinoco River at Guarguapo–YaYa Sector: Radioactive Tracer Study. In: 4th International Conference on Tracers and Tracing Methods – TRACER 4, Autrans/Grenoble, France (2006).
[39] MACHADO, M.L. et al., Orinoco River suspended sediment studies using ^{99m}Tc – Venezuela. In: Proceedings of The 5th International Conference on Tracers and Tracing Methods – TRACER 5, Tiradentes, Brazil, Belo Horizonte: CDTN (2008) CD-ROM
[40] CAILLOT, A. La Radioactivité pour l'Étude et la Mesure des Transports Solides. CEA Internal Report, Saclay, France (2009)
[41] GASPAR, E., Modern Trends in Tracer Hydrology – Volume I. CRC Press. ISBN: 0-8493-4320-8. (1987a)
[42] MOREIRA,R.M., et al., Hydrological and sedimentological surveys in the design of outfalls for industrial effluents, In: International Conference on Coastal and Port Engineering in Developing Countries (COPEDEC III), Mombasa, Kenya, Sept. (1991).
[43] BORROTO, J., et al., Behavior of ^{99m}Tc in highly polluted surface waters: field and laboratory studies. In Proceedings of The International Symposium on Isotope Techniques in Water Resources Development and Management, Vienna, 10-14 May 1999.IAEA (1999) 1 CD-ROM.
[44] LOWENTHAL, G.C., AIREY, P.L., Practical Applications of Radioactivity and Nuclear Radiations (Chapters 8 & 9), Cambridge University Press (2001).
[45] CAILLOT, A., La Radioactivité au Service de l'Industrie et de l'Environnement, EdsTEC & DOC, Paris (2002).
[46] BORROTO, J., Comportamiento del ^{99m}Tc como radiotrazador en aguas superficiales y residuales, Tesis presentada en opción al grado científico de Doctor en Ciencias Químicas. Instituto Superior de Tecnologías y Ciencias Aplicadas. Facultad de Ciencias y Tecnología Nucleares (INSTEC), Departamento de Radioquímica, La Habana, Cuba (2004) (In Spanish).
[47] HUGHES, C.E. et al., Using radiotracer techniques for coastal hydrodynamic model evaluation, Journal of Environmental Radioactivity, **76** (2004) 195-206.
[48] MOSER, H.; RAUERT, W., Injected isotopic tracers for obtaining hydrologic parameters. In: Isotopes in Water Cycle – Past, Present and Future of a Developing Science. IAEA Publication, AGGARWAL, P. K., GAT, J. R., FROELICH K.F.O. (Eds) Springer Edition (2005).
[49] BORROTO, J.; GRIFFITH, J., Validation of Tc-99m as a radiotracer for hydrological studies of natural waters. In: 4th International Conference on Tracers and Tracing Methods – TRACER 4, Autrans/Grenoble, France (2006).
[50] RAO, S.M., Practical Isotope Hydrology. New India Publishing Agency, New Delhi (2006).
[51] SEO, I.W., BAEK, K.O., JEON, T.M., Analysis of transverse mixing in natural streams under slug tests, J. Hydraulic Res. **44** 3 (2006) 350–362.
[52] LEIBUNDGUT, C.; MALOSZEWSKI, P., KÜLLS, C., Tracers in Hydrology, WILEY-BLACKWELL (Ed), ISBN: 978-0-470-51885-4 (2009).

[53] SUH, K.S., et al., Determination of dispersion coefficients using radioisotope data in river environment, Journal of Applied Radiation and Isotopes 67 (2009) 1499-1502.
[54] DAHLGAARD H., Radioactive tracers as a tool in coastal oceanography - An overview of the MAST-52 Project, Journal of Marine Systems **6** (5-6) (1995) 381-389.
[55] NIELSEN, S.P., A box model for north-east Atlantic coastal waters compared with radioactive tracers. Journal of Marine Systems. **6** (5-6) (1995) 545-560.
[56] ROBINSON, H.D., GRONOW, J.R., Tritium levels in leachate from domestic wastes in landfill sites. In: Proceedings of The International Symposium CISA, 5, Cagliari (1995).
[57] PERIANEZ, R., MARTINEZ-AGUIRRE, A., Uranium and thorium concentrations in an estuary affected by phosphate fertilizer processing - Experimental results and a modelling study, Journal of Environmental Radioactivity **35** 3 (1997) 281-304.
[58] VERHAGEN, B.T., et al., High level tritium in leachate from landfill sites in Republic of South Africa with emphasis on its distribution and value as an environmental tracer. In: Proceedings of the WISA Conference, Cape Town (1998).
[59] PUJOL, L., SANCHEZ-CABEZA, J.A., Determination of longitudinal dispersion coefficient and velocity of the Ebro River waters (Northeast Spain) using tritium as a radiotracer, Journal of Environmental Radioactivity **45** 1 (1999) 39-57.
[60] SANCHEZ-CABEZA, J.A., PUJOL, L., Study on the hydrodynamics of the Ebro river lower course using tritium as a radiotracer, Water Research **33** 10 (1999) 2345-2356.
[61] VERHAGEN, B.T., et al., Tritium in waste as a tracer of landfill leachate in surface and ground water in South Africa. In: Proceedings of the International Symposium on Isotope Techniques in Water Resources Development and Management, 10-14 May 1999, Vienna.(1999). 1 CD-ROM.
[62] BANDEIRA, J.V., et al., ^{99m}Tc mud labelling and its application in hydrodynamic studies of fine suspended sediment in Brazil. Proceedings of the 5th International Conference on Tracers and Tracing Methods – TRACER 5, Tiradentes, Brazil, 2008, Belo Horizonte: CDTN (2008). CD-ROM.
[63] KLAVERKAMP, J.F., et al., Fates of metal radiotracers added to a whole lake: accumulation in slimy sculpin (Cottuscognatus) and white sucker (Catstomuscommersoni), Sci Tot Env, **28** (1993) 119-128.
[64] HESSLEIN, R.H., BROECKER, W.S., SCHINDLER, D.W., Fates of metal radiotracers added to a whole lake: sediment-water interactions. Can, J. Fish. Aquat. Sci. **37** (1980a) 378-386.
[65] HESSLEIN, R.H., BROECKER, W.S., QUAY, P.D., Whole-lake radiocarbon experiment in an oligotrophic lake at the Experimental Lakes Area, north-western Ontario, Can. J. Fish. Aquat. Sci. **37** (1980b) 454-463.
[66] QUAY, P.D., et al., Vertical diffusion rates determined by tritium tracer experiments in the thermocline and hypolimnion of two lakes, Limnology and Oceanography, **25** 2 (1980) 201-218.
[67] KJELLIN, J., WÖRMAN, A, JOHANSSON, H, LINDAHL, A, Controlling factors for water residence time and flow pattern in Ekeby treatment wetland, Sweden, Advances in Water Resources **30** (2007) 838-850.
[68] WILLIAMS, L.E., Anniversary Paper: Nuclear medicine: Fifty years and still counting, Medical Physics **35** 7 (2008).

RADIOTRACER METHODOLOGY

P. BRISSET[*], P.L. AIREY[**], S.H. JUNG[***], C.E. HUGHES[**]

[*]French Alternative Energies and Atomic Energy Commission, CEA, France/IAEA
[**]Australian Nuclear Science and Technology Organisation, ANSTO, Australia
[***]Korea Atomic Energy Research Institute, KAERI, Republic of Korea

Abstract

Methods for the use of radiotracers in aquatic environments have been established over the past five decades. The basic principles involve definition of the system or problem to be investigated, selection of a suitable tracer, design of tracer deployment and measurement systems and analysis of the data collected to address the initial problem. These aspects are covered in detail in this paper covering the current state of the art in tracer technology. New developments in the use of nano-particle tracers are also addressed.

1. PRINCIPLES OF TRACER STUDY DESIGN

'Tracers may be defined as substances that behave identically in essential respects to the selected component of a complex system and which can be measured independently of the other components and with high sensitivity' [1]. In developing a strategy for an investigation, the practitioner must select the key component of the system for labelling, choose the optimum radionuclide as a tracer and apply the most favourable injection procedure and measurement techniques.

The principle underlying most radiotracer experiments is illustrated in Fig. 1. In the simplest case, a small quantity of a radiotracer is injected at point A, and disperses through the system past a strategically located detector at B.

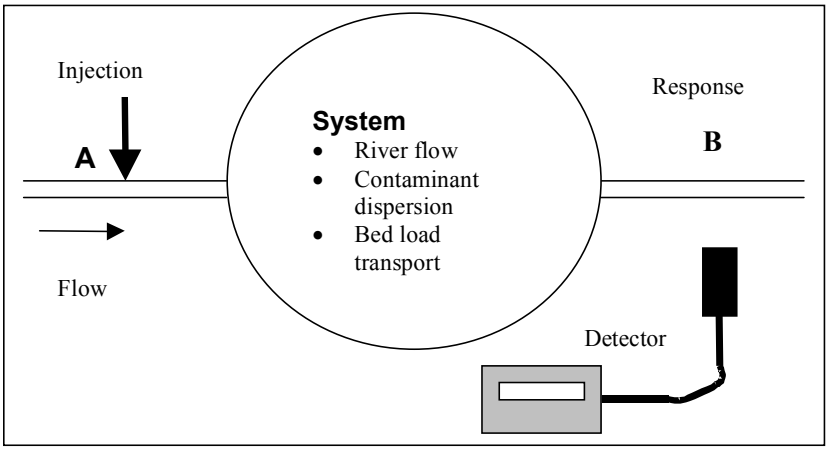


FIG. 1. General principle of the tracer study. The tracer is injected at A, is transported through the system, and the response monitored at B.

The success of a tracer experiment depends upon proper planning, execution and analysis of the primary data. The accurate measurement of the response curve to a tracer injection pulse is crucial for further processing and interpretation. The response curve can be obtained either by *in situ* measurement of the radioactivity, or by collection and analysis of samples in laboratory.

Many publications have addressed tracer methodology and application as referenced in the introduction. In particular the IAEA has published a number of reports covering technical aspects of radiotracing for industrial and environmental applications [2–7]. The objective of

this section is to focus on specific points or aspects of radiotracer methodology, to complement the current literature.

Radiotracer applications are a very dynamic area of research and technology development. On the one hand they enable the collection of data which cannot be obtained by other investigative techniques, and on the other hand there are enormous opportunities for the refinement of field data acquisition. This is because radiotracer methodology comprises many interrelated aspects including: tracer selection and injection, radiation detection and measurement as well as interpretation and modelling which benefit from new procedures for data acquisition, analysis and imaging (Fig. 2). Advances in software have facilitated the widespread applications of Monte-Carlo simulations, RTD calculations and CFD modelling. Finally there are repercussions from the developments in medical and biological sciences, environmental engineering and elsewhere.

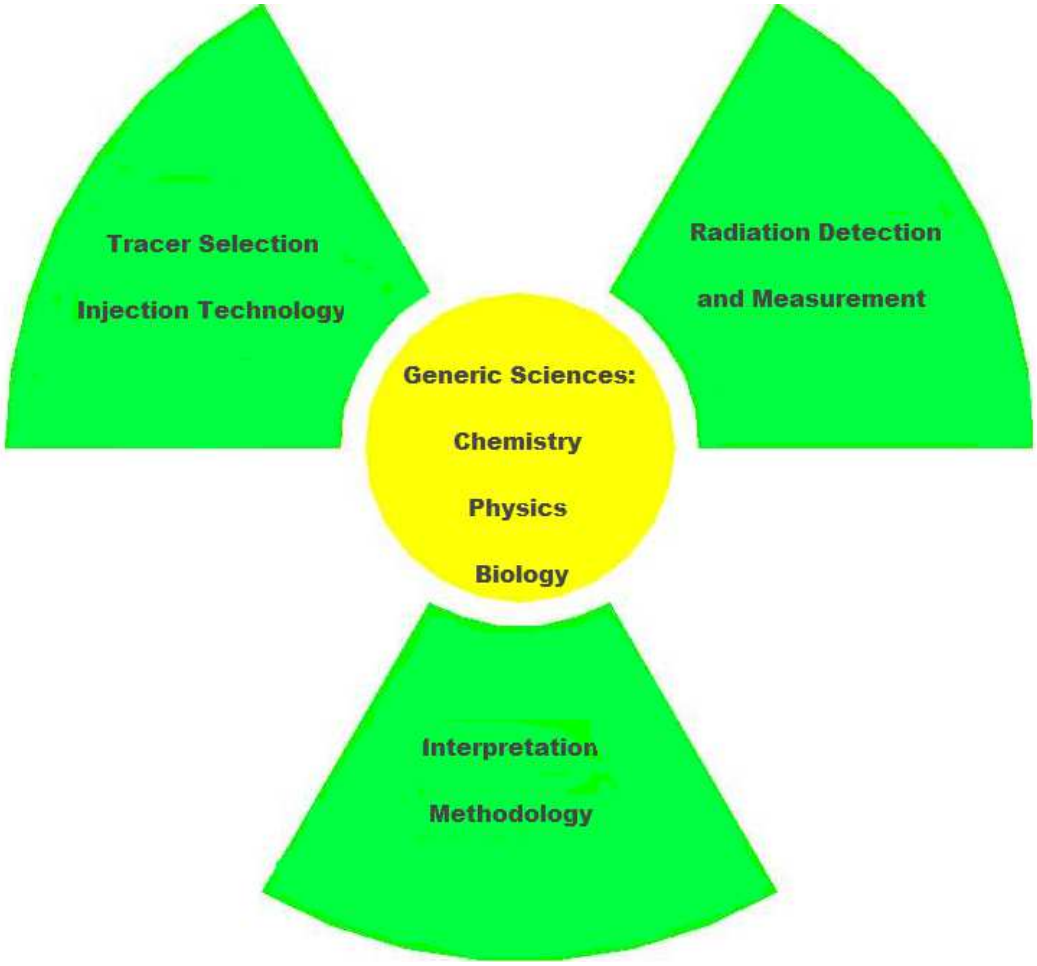
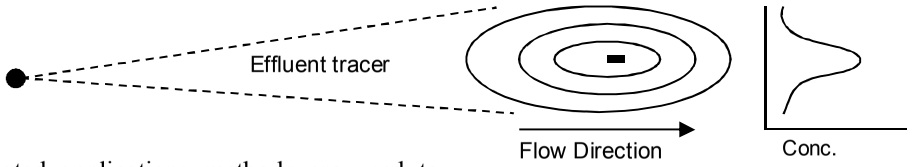
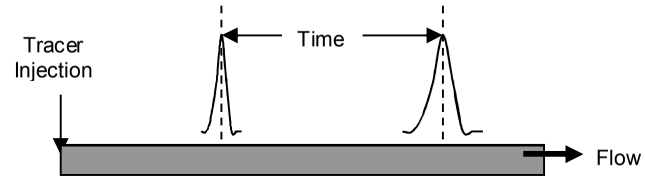
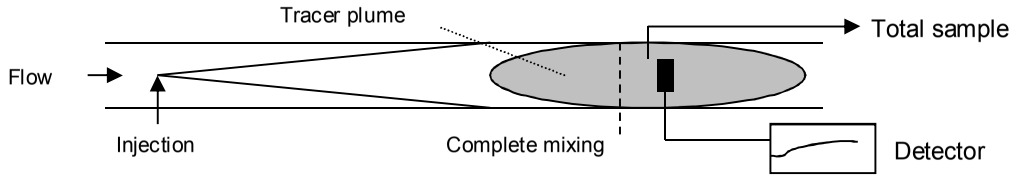
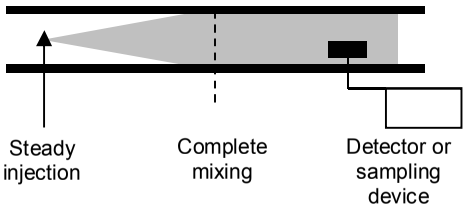


FIG. 2. Radiotracer methodology interrelated aspects [5].

Table 1 provides a summary of the range of measurements, techniques and applications of radiotracers in surface water environments.

TABLE 1. APPLICATIONS OF ARTIFICIAL RADIOTRACER TECHNIQUES FOR STUDIES OF WATER SURFACE PROCESS

Measurement	Technique	Application	Comment
Dilution factors	Samples are taken downstream of the injection point and the tracer concentrations expressed as fractions of those at a reference location.	Environmental Eng: Assessment of the effectiveness of engineering structures (e.g. sewage outfalls) in diluting effluent into, e.g. coastal waters.	
Dispersion	Tracer techniques can be used to measure the capacity of receiving waters to dilute and disperse effluent. If the tracer is released as a point source, say from an ocean outfall, the tracer concentration profiles frequently approximate Gaussian curves. An analysis of these curves yields dispersion coefficients in the longitudinal, transverse and depth directions.	 In the most sophisticated applications methods are used to evaluate environmental transport models	
Flow rates Pulse velocity	An isotope pulse is injected into a pipeline, a channel or a river and the flow rate measured by noting the time for the passage of the pulse between two points. To calculate the volume flow, the average cross-section must be known.	Because of its simplicity, the method is widely used for the measurement of flows in industry and the environment. For the most accurate measurements, an analysis of the pulse shape is necessary under conditions of complete mixing.	
Flow rates Tracer dilution methods	A tracer is injected into a pipeline, channel or river, either at a steady rate, or as a pulse. After complete mixing has been achieved, the dispersing plume is monitored. Measurement of the cross sectional areas are not necessary to calculate volume flows.		

Measurement	Technique	Application	Comment
Steady state injection	The tracer is injected at a steady rate for a period necessary to establish a steady concentration of tracer at the measurement point (see diagram).	Although the most accurate method, it is not widely used because considerably more tracer is required than with the pulse methods.	
Pulse injection	The tracer is injected as an instantaneous pulse and monitored after achievement of complete mixing.	▪ Since volume flow rates do not require cross-sectional area measurements, they are ideally suited for channels etc. with rough profiles. ▪ Using the total sample method, the mass flow of gas may be measured knowing the density of gas under ambient conditions after sampling.	
Total sample method	A sample is pumped from a given point at a constant rate for a measured time during the whole pulse.		
Total count method	The integrated net count rate from a calibrated detector at a fixed location is measured.		

2. TYPES/CHARACTERISTICS OF TRACERS

2.1. Selecting a suitable tracer

Selecting an appropriate radiotracer involves balancing the following:

- The integrity of the tracer: Does the radiotracer behave identically in essential respects to the component of the system which is under study?
- The nuclear properties of the tracer. Does the tracer emit a gamma ray with adequate efficiency and of adequate energy for efficient detection? Other factors having been considered, the radiotracer with the shortest half-life is favoured being more efficiently removed by decay from the environment.
- Radiological and environmental aspects. Does the handling and transport of the radiotracer lead to a radiological impact on operators or the critical group within the community which is within regulations and, in addition, is as low as reasonably achievable (the ALARA principle)? Are there any other environmental impacts, which are not adequately reflected in human health assessments?

A wide variety of radiotracers are potentially available. Over 3000 radionuclides are listed in the chart [8, 9]. In practice, about 100 are commonly used in lab, field and industrial settings, some of which are listed in Tables 2, 3 and 4.

TABLE 2. RADIONUCLIDES COMMONLY USED AS ENVIRONMENTAL TRACERS (WATER COLUMN)

Tracer	Half life	Nuclear properties	Comments
Tritium HTO	12.33 y	β- emitter Emax 18.6 keV	▪ 'Perfect' tracer for water (chemically); ▪ Cannot be detected in situ with deployed detectors (major problem); ▪ Often used with gamma emitters for 'internal' system calibration.
Br-82	35.3 h	Emits 6 gamma rays in the range 554 to 1044 keV	▪ Readily monitored by deployed gamma detectors; ▪ 'Simple' chemistry; ▪ Relatively short half- life.
Tc-99m	6.0 h	141 keV(89%)	▪ Readily monitored by deployed detectors; ▪ Eluted from a Mo-99 generator ($T_{1/2}$ 66 h); ▪ Medical generators have very refined engineering and safety features; ▪ However, the chemistry is complex and not well understood in environmental systems.
Au-198	2.69 d	412 keV (96%)	▪ Used in offshore effluent studies; ▪ Gold does not dissolve in environmental water but adsorbs on sediment fines – in practice, transport properties normally reflect those in the water column

TABLE 3. RADIONUCLIDES COMMONLY USED AS ENVIRONMENTAL TRACERS (BED LOAD AND SUSPENDED PARTICLES TRANSPORT)

Tracer	Half life	Major gamma rays (intensity%)	Comments
Au-198	2.69 d	412 keV (96%)	▪ Labelling involves surface adsorption from aqueous phase or incorporation into glass; ▪ Duration of investigation, 2 weeks.
Cr-51	27 d	320 keV (9.85%)	▪ Surface labelling of sand particles by sorption – stabilization by baking in a muffle furnace; ▪ Duration of investigation, 3 months.
Ir-192	74 d	468 keV (48%); 316 keV (83%) + others	▪ Ir-192, Sc-46 and Ag-110m are normally incorporated into glass, and prepared in a matching particle size distribution.
Sc-46	80 d	1121keV (100%), 889 keV (100%)	
Ag-110m	249.8 d	1505 keV (13%); 1384 keV (24%) + others	

TABLE 4. RADIONUCLIDES COMMONLY USED AS ENVIRONMENTAL TRACERS (BIOLOGICAL OR REACTIVE PROCESSES)

Tracer	Half life	Major gamma rays (intensity%)	Comments
Zn-65	244.2 d	1115.55keV (50.6%)	▪ Uptake of heavy metals by biota can be monitored in lab and field environments; ▪ Adsorption of heavy metals to sediments or particulates and retardation of metal constituents in contaminant plumes may be studied; ▪ Transfer through the food chain or transfer from sediment to biota may be measured due to the longer half- life of these tracers enabling studies in the order of one year or more in closed systems.
Fe-59	44.5 d	1291.3 keV(43.2%) 1099.25keV (56.5%) + others	
Hg-203	46.6 d	279.2 keV (81%)	
Cd-109	462.6 d	88 keV (3.6%); β^-Emax 126 keV	
Se-75	119.8 d	400.7 keV (11.5%); 279.2 keV (25%); 264.7 keV (58.9%); 132 keV (58.3%); 121.1 keV (17.2%) + others	
Cs-134	2.065 y	1365.19 keV (3%); 795.86keV (85.5%); 604.72 keV (97.6%) + others	
Co-60	5.27 y	1332.5 and 1173.25 keV (100%)	
H-3	12.33 y	β- Emax 18.6 keV	▪ Used to label organic compounds or as nutrients; ▪ Transport, uptake and degradation studies; ▪ Beta emitters require sampling and measurement in the lab.
C-14	5730 y	β- Emax 156.48 keV	
P-32	14.26 d	β- Emax 1.709 MeV	

2.2. Selecting a suitable tracer for tracing water

On the face of it, the use of HTO (tritiated water) as a tracer for water or aqueous systems would appear to be an obvious choice. However, tritium is a very low energy β particle emitter (Emax = 18.6 keV) and hence its activity cannot be measured in situ. Samples must be collected and returned to the laboratory for analysis.

Other tracers most commonly used in aqueous solutions, if data is required in real time, are gamma ray emitting isotopes such as ^{51}Cr-EDTA complex, ^{113m}In-EDTA complex, Na^{131}I, K^{131}I, $^{24}Na_2CO_3$, $^{24}NaHCO_3$, $NH_4{}^{82}Br$, $H^{198}AuCl_4$ and pertechnetate, $^{99m}TcO_4^-$.

3. ADVANTAGES AND DISADVANTAGES OF DIFFERENT TRACERS

In attempting to solve environmental problems, and determine the factors above, radioisotope tracers have a number of advantages over chemical, visible or fluorescent materials. In some cases such as in very turbid waters or very large deep water diffuser systems where dilutions are high and effluent discharge rates can be 1000 x 10^6 L/d and dyes simply do not have the necessary sensitivity. However the principal advantages of radioisotope tracers are as follows:

Uniqueness: Rarely will any type of radionuclide, let alone the specific one chosen for a particular investigation, be present in the system under examination. The radionuclide will uniquely and unambiguously define material originating from a particular source.

Chemical analogue: In processes where a particular chemical species needs to be monitored, use of the appropriate radionuclide analogue will ensure that the tracer behaves exactly like the chemical under investigation because, with few exceptions, all the isotopes of an element, whether radioactive or stable, have the same chemical behaviour. Where only a particular phase needs to be tagged, there is more freedom of choice for the most appropriate radionuclide label and also because of the high sensitivity and stability of modern electronic nuclear radiation detection equipment.

External monitoring: Many radionuclides emit γ-rays which can be readily monitored using a variety of detectors. Two benefits accrue from this feature. One, the behaviour *of* a system can be monitored externally and the perturbations caused by sampling can be avoided and two, the movement *of* the tracer can be followed directly thus allowing 'real time' measurements. This latter feature enables complex systems to be studied in the most cost effective way.

Limited 'memory effect': Since each radionuclide is unstable and decays at a known constant rate, the time interval for it to become effectively 'dead' can be readily calculated. Thus careful selection of a radionuclide tracer can allow multiple experiments to be undertaken over a period without cross-contamination or 'memory effect'.

Minimal tracer mass: In general, radionuclide tracers are required in only physically small amounts which do not constitute a major perturbation (milligrams of solids or millilitres of liquids). This is because a large amount of radioactivity can be induced into many elements through nuclear reactor or cyclotron irradiation.

4. RADIOTRACER DEPLOYMENT METHODS

4.1. Radiotracer fabrication

The amount of activity and mass (number of particles) required for an investigation should be sufficiently high for detection during the experiment but at the same time, should be minimized on the grounds of the radiological protection for the operator, public and the environment.

The amount of activity usually depends mainly upon efficiency of the detector used for detection, expected spread of the tracer, the level of background radiation and duration of the study.

Ultimately the activity is limited by the regulatory approval in each in each country.

After estimating the activity and mass of the tracer, the target is irradiated in a reactor (or cyclotron) and brought back to the hot cell. In hot cell the activated target is removed from the irradiation container (usually aluminium or silica), chemically treated, if required, and transferred to an approved transport container. In some countries (e.g. Republic of Korea) the transport container itself is designed and used to inject the tracer. This avoids the unnecessary exposure during injection procedure.

In some cases the radiotracer can be obtained from a radionuclide generator. A radionuclide generator is a chemical/physical/mechanical device that is based on a nuclear mother-daughter genetic relationship and which allows for the separation and extraction (elution) of the short-lived daughter in pure form from the longer-lived (stationary) mother. The elution may be repeated at intervals.

The most common radionuclide generators commercially available and usable for environmental applications are listed in the Table 5.

TABLE 5. RADIONUCLIDE GENERATORS COMMERCIALLY AVAILABLE

Generator	**Half-lives**	**Radiation type and energy**	**Comment**
$^{99}Mo \rightarrow ^{99m}Tc$	$T_{1/2}$ (^{99}Mo) = 66 h $T_{1/2}$ (^{99m}Tc) = 6 h	Mother: β^-, γ 740 keV Daughter: IT 141 keV	Energy is not ideal (too low for many studies), used because of generally good availability due to extensive use in hospitals. Eluate is $^{99m}TcO_4^-$ in weakly saline aqueous solution
$^{113}Sn \rightarrow ^{113m}In$	$T_{1/2}$ (^{113}Sn) = 115 d $T_{1/2}$ (^{113m}In) = 99.5 min	Mother: EC, γ 255 keV Daughter: IT 392 keV	Commercially available with regular production at several irradiation facilities, may be distributed by IAEA to developing countries for a reasonable price. Eluate is In^{3+} in weak hydrochloric aqueous solution
($^{137}Cs \rightarrow ^{137m}Ba$)	$T_{1/2}$ (^{137}Cs) = 30 y $T_{1/2}$ (^{137m}Ba) = 2.55 min	Mother: β^-, no γ Daughter: IT 661 keV	Is not commercially available in industrial strength at a regular basis, may be produced on special order. Eluate is Ba^{2+} in weak acidic aqueous solution

4.2. Transportation of the radiotracer

All the safety measures requested within the international and national regulations for dangerous materials (Category 7: radioactive) transportation are followed during the transport of the tracer from reactors to the hot cell and subsequently from hot cell to the experimental site. The dose rate at the surface of the transport container should not exceed 2 mGy/hr [10]. Fig. 3 shows an example of a radioactive transport container.

FIG. 3. Example of a transportation container.

4.3. Injection of the tracer

Injection systems are generally laboratory crafted and adapted for specific applications. A range of designs for industrial and environmental applications are illustrated in this section. They vary considerably in concept from the simplest (a syringe or a reservoir with a peristaltic pump e.g. Fig. 4) to the most complex (devices for remote injection into pressure vessels). A multi-purpose device for pulse injection is presented in Fig. 5. The red arrows indicate the fluid flow inside this system from tracer storage house to the industrial reactor.

The system shown in Fig. 5 is designed for industrial applications. The tracer is initially contained in a quartz ampoule, which is placed in an internal steel cylinder inside the dilution chamber (1). The chamber is then filled with the fluid being traced (gas, liquid or particles).If required this volume is connected to a pressurized container (6). The pneumatic actuator (2) releases a hammer (3). Its initial top position in the chamber and running length can be adjusted in order to efficiently break the quartz ampoule into pieces that are retained by a grid. The tracer is then diluted by the pressurized fluid contained in the chamber. The pneumatic three way valve (5) facilitates the injection of the labelled volume of fluid into the industrial reactor. Once the tracer has been injected, it is possible to extract the internal cylinder containing the broken quartz ampoule.

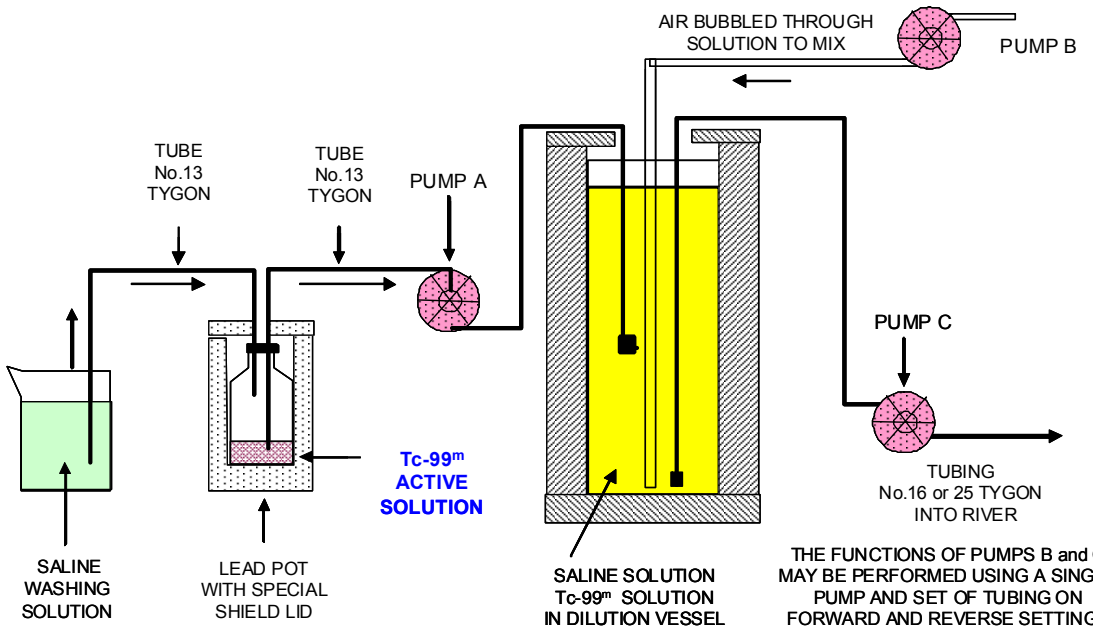


FIG. 4. Schematic of simple peristaltic pump driven injection setup for continuous injection. The active solution is flushed into a mixing vessel, mixed with air bubbles then metered into the receiving waters.

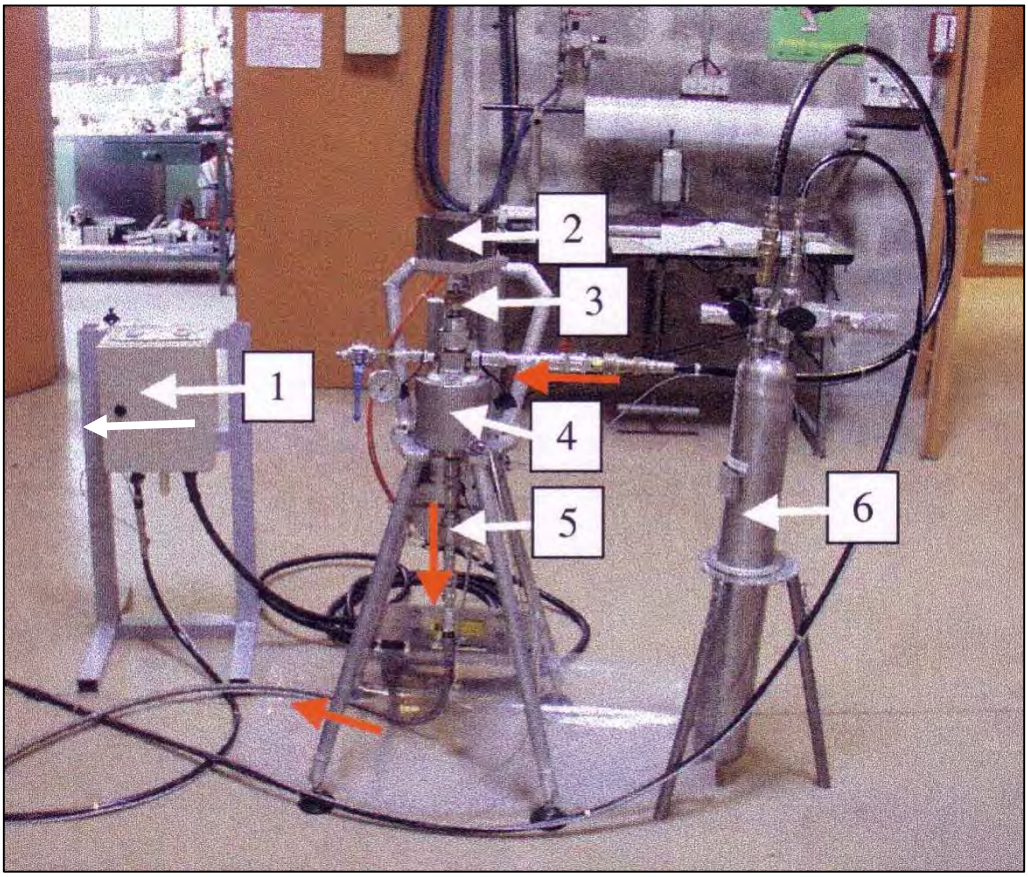


FIG. 5. Photograph of a multi-purpose injection device.

After the tracer ampoule has been placed in the dilution chamber, every operation is remote controlled so as to minimize exposure to radiation. This system injects the radiotracer as a pulse (Dirac type) signal into the flow under investigation. It is worth noting that other types

of injection (continuous flow rate, sinusoidal, random) can be of great interest in the understanding of certain complex processes. Nevertheless, most industrial processes can be perfectly characterized using the above proposed injection device provided that the time duration of the injected signal is about 100 times shorter than the MRT of the studied flow. Fig. 6 depicts a typical injector that is suitable for a variety of applications.

FIG. 6. A conventional injection device for chemical industry applications.

Fig. 7 illustrates the principles of the injection systems for pulse and continuous injection of tracer, as well as simple practical injectors for laboratory scale tracer experiments.

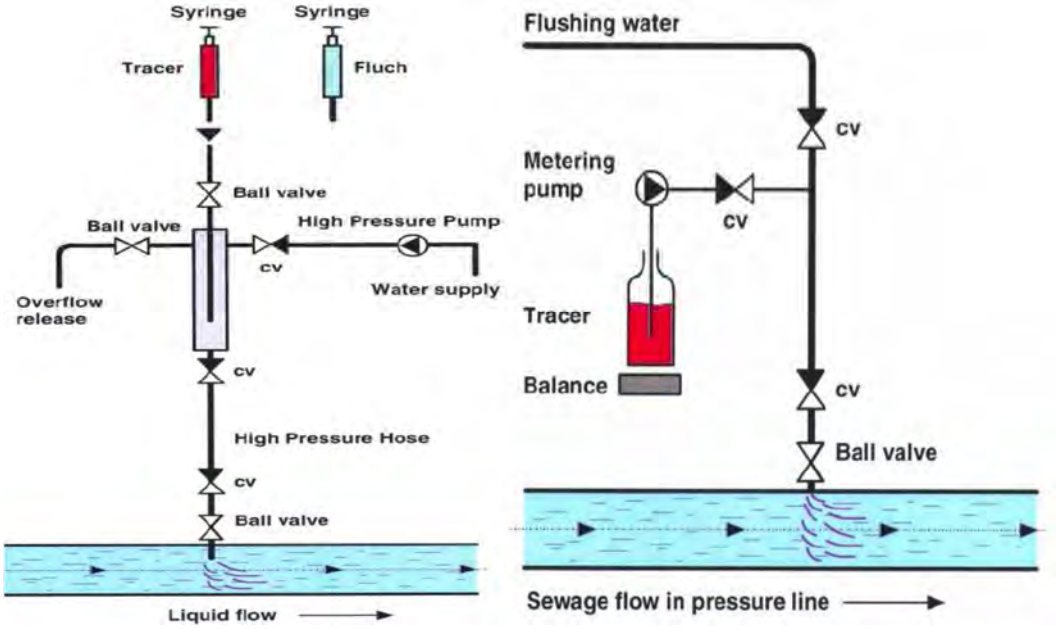


FIG. 7. Principle of tracer injections, left pulse injection, right continuous injection.

In many cases particles have to be labelled by the tracer(s) for suspended sediment transport or sewage outfalls experiments. In some cases a glass containing the tracer will be produced (Table 3), however in some instances the natural particles are labelled with the selected radioisotope. The radioisotope is adsorbed onto the surface of the particle after a suitable treatment. Particle of size less than 0.04 mm should be labelled under careful conditions in order not to modify the surface properties and thus their hydrodynamic behaviour. Care should be taken to ensure that the radioactivity does not get released under the severest field conditions.

The uptake of the label on mud or sludge particles is proportional to the mass of the particle. The labelling for sand particles is superficial and hence the activity labelled will be proportional to the surface area of the grains.

Gold-198 (^{198}Au) and scandium-46 (^{46}Sc) are the two most commonly used radioisotopes used for labelling particulate material. The half-life of ^{198}Au is 2.7 days and thus is used for short term studies whereas the half-life of ^{46}Sc is 84 days and thus is used for long term investigations.

The particulate material is directly labelled with the desired radioisotope at the site and hence the operators are likely to receive some radiation dose.

The labelling efficiency is a function of various parameters such as concentration of radioisotope to be labelled, pH, amount of particulate material and its particle size distribution, equilibrium time etc. and needs to be investigated beforehand. In addition, the uniformity of labelling and leaching of the radioisotope is also assessed under simulated experimental conditions. After optimizing labelling conditions, one knows the amount of activity require for prepare the tracer. Generally the labelling efficiency is from 80% (sand) to 99% (mud).

Figures 8 and 9 show different types of injection and labelling systems. These systems allow instantaneous or continuous injection.

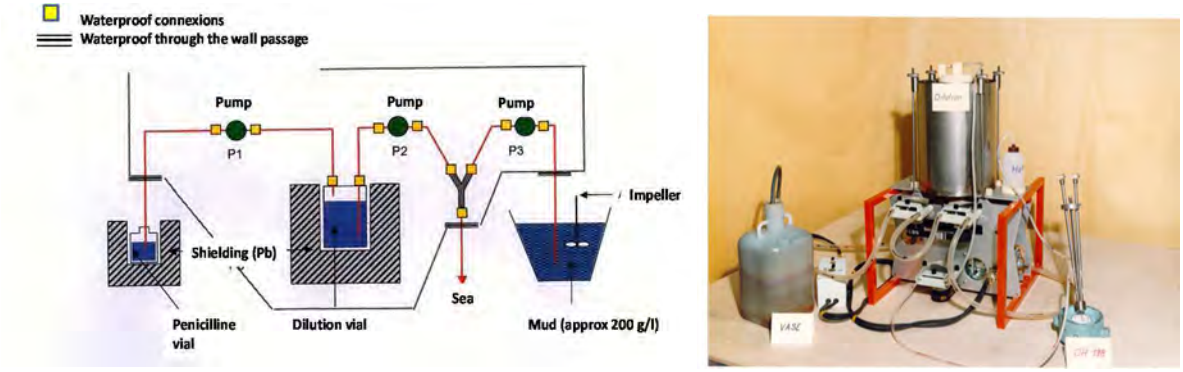


FIG. 8. Labelling and Injection system used in France (this system is used in a glove box).

FIG. 9. Labelling and injection system used in Brazil and some countries in Latin America.

During injection operations, radiation doses to the workers have to be monitored. These doses have to be estimated before the experiment and minimized as required by the ALARA principle. Precautions have also to be taken in order to avoid any incidental contamination of

the injection place using plastic covers for example. These points are treated in detail in the paper from Hughes and Brisset titled “Radiation protection and regulation for environmental radiotracer studies”, included in this publication.

5. MEASUREMENT TECHNIQUES – IN SITU AND LABORATORY (SAMPLING) EQUIPMENT

After injection, one proceeds to the detection which could be static (in narrow rivers) or dynamic (large rivers or sea).

5.1. Static detection

Detectors are placed along the river at fixed points to measure the count rate versus time. They can be placed directly on the river bed or placed in the water layer from bridge or from a rope strained between the banks as shown in Figs 10 and 11. A calibration factor, taking into account the detection geometry is needed to convert the raw data (counts per second) to concentration units (specific activities, Bq/L). Assuming that the distance between the tracer injection point and the location of the detector is such that complete mixing is achieved, the detector response can be readily interpreted in terms of RTD theory.

FIG. 10. Installation of NaI scintillation detectors for the radiotracer experiment in a river.

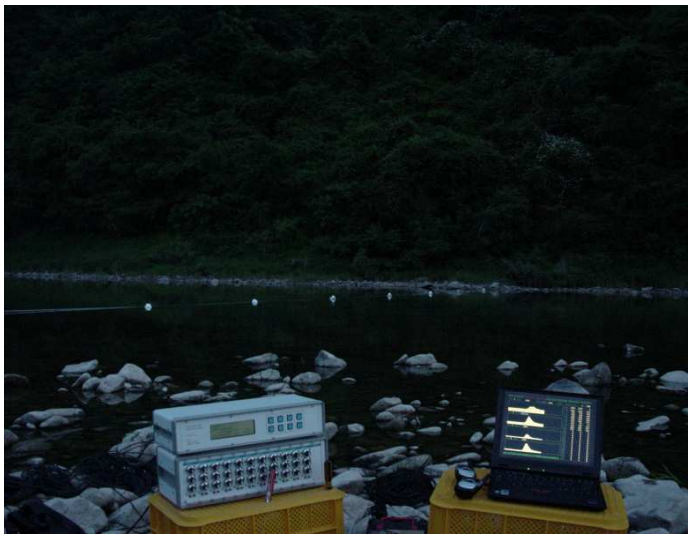


FIG. 11. Radiotracer data acquisition from detectors installed in a river.

5.2. Dynamic detection

Detectors are placed on a boat which follows the tracer cloud to measure its main characteristics. The detectors are placed in the water column on a cable strained vertically by a lead fish. (Fig. 12)

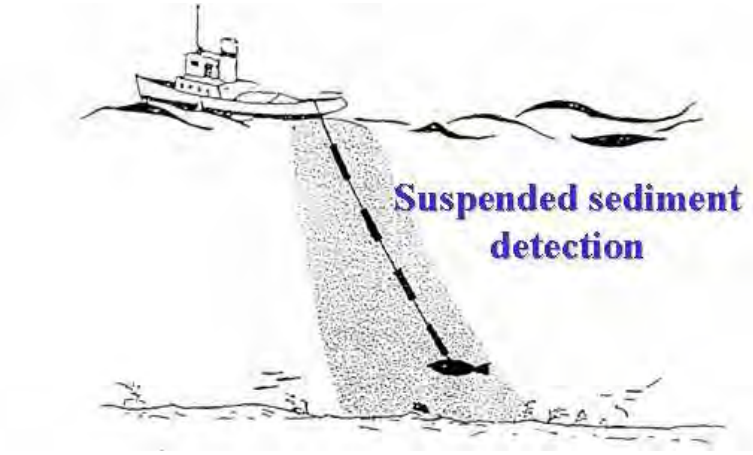


FIG. 12. Detector placed on a vertical cable.

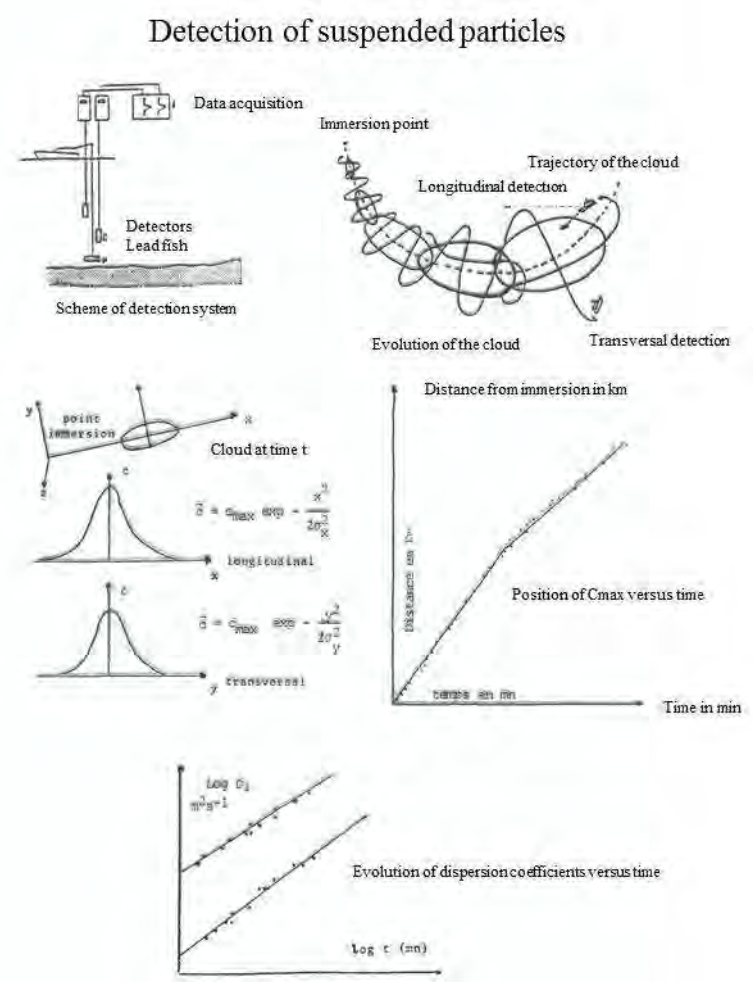


FIG. 13. Principle of the detection of a cloud of suspended particles.

In this case the number of suspended detectors relies on the site conditions and objective of the work (typically up 5 for 20 m water depth). Vertical profiles may also be obtained by lowering and raising a detector and depth sensor through the water column. At least 1 boat is used to criss-cross the cloud mainly at right angles to the direction of transport (transverse profiles). Preferably a second boat would be recording longitudinal profiles.

Nowadays, the spatial positioning of the boats is undertaken with differential global positioning system (DGPS) technology (precision typically 1 m).It is important to correct for the offset between the detector position and that of the boat's DGPS antenna.

It is also important to correct the variations of the detectors depth which can vary according to the relative velocity between the boat and the water. Some detectors are equipped with pressure sensors allowing to measure simultaneously the tracer concentration and the depth of the detector as shown in Fig. 13.

In many studies of the transport of suspended particles a boat equipped of a sledge detection system is used to explore the bottom of the experimental area to map the (potential) deposit.

Depending on the requirements of the investigation, the data may be interpreted in terms of the following parameters:

(a) Trajectory and advection velocity of the tracer cloud;
(b) Horizontal dispersion coefficients;
(c) Dilution along the cloud trajectory (vs. space and time);
(d) Settling velocity (for particles);
(e) Map of the potential deposit (sewage outfalls or dumping of dredged materials).

5.3. In situ detection equipment

These systems comprise radiation detectors, rate meters and either data loggers or output devices. When working with X or γ rays, NaI(Tl) crystals optically coupled to a photomultiplier tube and associated electronics are almost universally used as radiation detectors because they are relatively cheap, efficient and robust. Either 50 × 50 mm, 38 × 50, 38 × 25 or 25 × 25 mm crystals are used as shown in Fig. 14. There is little incentive to use larger crystals because the bulk of the signal is scattered radiation with energies below 150 keV. At these energies, the absorption efficiencies approach 90% even when using 25 × 25 mm crystals.

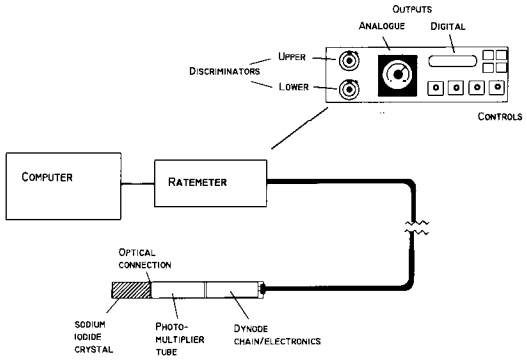


FIG. 14. Field detection system, comprising a probe (Na(Tl) crystal, PM tube and electronics, a ratemeter and computer.

The radiation probe in Fig. 15 is mounted in a cylindrical aluminium or stainless steel case. Although more rugged, the stainless steel housing absorbs a higher proportion, especially of the low energy radiation, than aluminium. The loss of efficiency could exceed 50%, and practitioners need to choose between enhanced efficiency and reduced ruggedness.

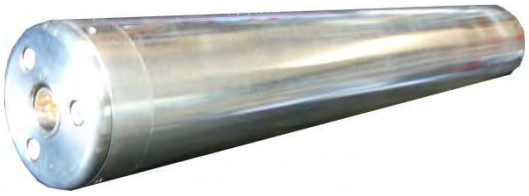


FIG. 15. Example of a waterproof NaI(Tl) ф 38 × 50 mm detector, waterproof 80 m water depth, aluminium or stainless steel housing.

With field equipment, the electronics are usually stand-alone units including amplifiers, discriminators and ratemeters operated by a battery. The data is frequently stored for 'on-line' or later transfer to computers or other output devices.

In accordance with the purpose of the experiment, radioisotope concentration is recorded along with the position of the radiation sensor fixed at specific depth in the water column. Data acquisition systems or software may be used to receive data from DGPS system and radiation counters concurrently. The counts rate recorded by a nuclear counter and the position of the detector recorded by a DGPS can be integrated and stored in a DAS in order to make it easy to install and operate them on board. An example of this is shown in Fig. 16. The counts (counts/second) recorded are corrected for natural background and decay. The net counts are plotted as a function of latitude and longitude on a survey chart of the respective sites.

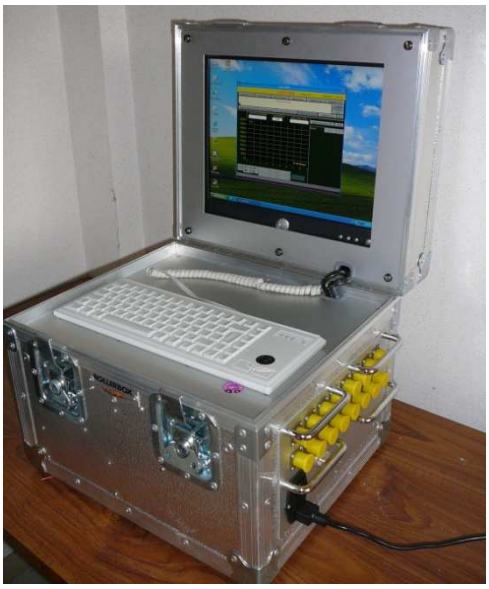


FIG. 16. Multi probes (15) data acquisition system.

Fig. 17 shows data acquisition software developed by ANSTO in Australia using Labview which is able to receive and store DGPS, radiation detector, depth and water quality data in real time displaying it onscreen in plan or sounding view to assist in navigation. This software also corrects for the position of the detector with respect to the DGPS antenna location which is valuable for small plumes or long cable lengths.

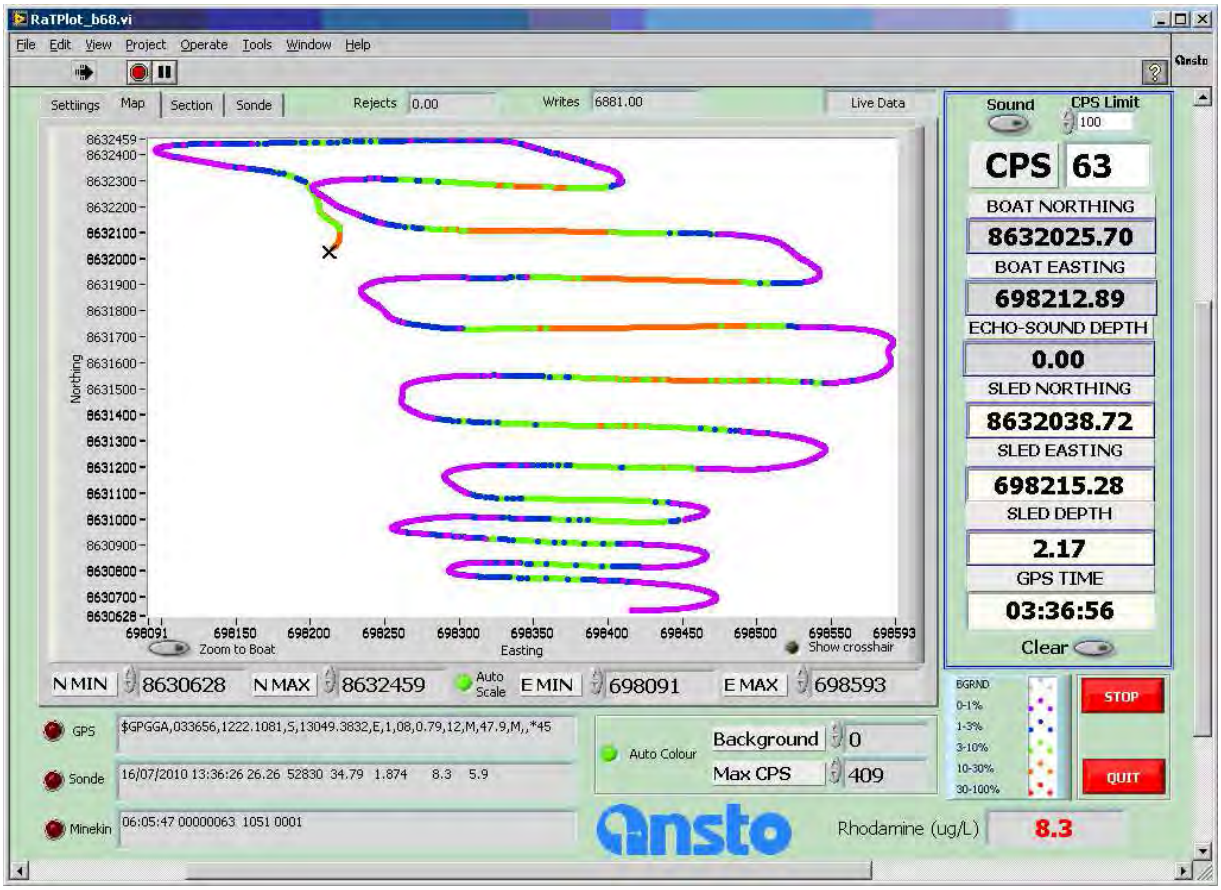


FIG. 17. Screenshot of Labview based radiotracer data acquisition and display module in operation.

5.3.1. Accurate field measurements

For accurate field measurements, system checks and calibrations should be undertaken under conditions as close as possible to those in the field.

The accuracy of such measurements depends on the quality of the calibration of the detector; and this is often limited by the need to reproduce in the laboratory the counting geometry that is found in the field.

Three classes of counting geometry may be distinguished:

- Quasi infinite geometry, when the detector is completely immersed in a large volume of liquid such as industrial tanks, rivers or lakes;
- Quasi planar geometry, when the detector is mounted, for instance, on a sled a few centimetres above labelled sediment on the bed of an estuary;
- Quasi linear geometry when the detector is mounted external to a pipeline.

An example of detector calibration for measurements under quasi infinite conditions is shown in Fig. 18.

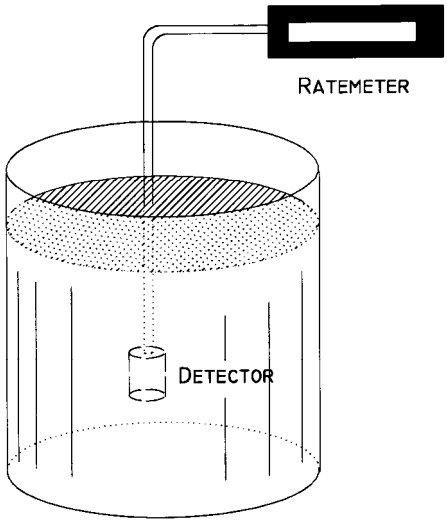


FIG. 18. Tank for detector calibration under 'quasi infinite geometry' conditions.

5.4. Sampling and laboratory measurement, equipment

5.4.1. Sampling measurement

Measuring techniques for samples depend on which kind of radiotracers (beta or gamma) is used. From the outset, attention is brought to the paramount need for representative samples.

When counting a radioactive sample it is well known that instrument reading is a measure of sample activity plus background. The latter must be subtracted in order to evaluate the net sample activity. The background is usually measured on blank samples taken before the injection.

The following criterion is often used to calculate minimum detectable concentration (MDC) of radiotracer taking background into account:

$$n > 2\,\sigma\,(n)$$

i.e. the measured count rate n (including background) should be at least two times the standard deviation.

The standard deviation is given by the expression (1):

$$\sigma(n) = \sqrt{\frac{n_g}{t} + \frac{n_b}{t}} \tag{1}$$

Where:

$\sigma\,(n)$ is the standard deviation for the net count rate (counts/s)
n_g is the gross count rate (counts/s)
t is the measuring time (s)
n_b is the background count rate (counts/s)

On this basis, the following expression (2) is obtained for MDC of a sample.

$$MDC = \frac{2.8}{e\,V_m}\sqrt{\frac{n_b}{t}} \qquad (2)$$

Where:

MDC is the minimum detectable concentration (Bq/L)
n_b is the background count rate (counts/s)
t is the measuring time (s)
e is the efficiency (counts/disintegration)
V_m is the volume of the sample (L)

5.4.2. Beta tracers

The most common beta-radioactive tracer for water transfer studies is tritium (in the form of tritiated water: ^{3}HHO).Some compounds labelled with tritium, carbon-14 (^{14}C) or sulphur-35 (^{35}S) can also be used in special cases. All of them are usually measured by means of liquid scintillation technique.

Liquid scintillation counting: A small volume of a liquid sample is mixed with a special solution known as scintillation cocktail, commonly in a 20 ml light transparent (glass, polypropylene, Teflon) vial. Beta particles cause emission of light when passing through and slowing down in the scintillation cocktail. These light pulses are registered by photo-multipliers (PMT) suitable for that particular photon wavelength. The light output in a pulse (light intensity) is proportional to the energy of the beta particle. This process is called scintillation, and since it happens in liquid media, it is known as liquid scintillation. The vial is placed inside an instrument, a liquid scintillation counter (LSC), which has normally two PMTs operated in co-incidence to reduce background. The LSC analyses the pulses from the PMTs and provides information about the energy of the beta particles and the rate of beta emission (activity) in the sample.

Various processes may perturb the beta-spectrum obtained in a liquid scintillation process. The most important are chemical and physical quenching which result from the fact that some chemicals may absorb the energy and release it in the form of heat rather than light pulses. Heavy chemical quenchers include organic compounds containing oxygen and in particular chlorine. All quenching results in a shift of the energy spectrum towards lower channel numbers because the number of photons detected by the PMTs per beta decay is reduced. Evaluation of the quenching effect which may vary from sample to sample is necessary to calculate counting efficiency.

Liquid scintillation counting requires careful sample preparation. Most often, chemical separations are involved. When these procedures are optimized, very low detection limits may be obtained ranging from 2 Bq/L for HTO to <0.02 Bq/L for $S^{14}CN^{-}$.

5.4.3. Gamma tracers

Gamma tracers are commonly measured using either solid scintillation detectors or semiconductor detectors.

Solid scintillation detectors: These are of different types, but the most generally applicable is the detector based on a single crystal of sodium iodide doped with traces of thallium, the so-called NaI(Tl)-detector. The crystal is optically coupled to a photomultiplier (PMT). Interaction of a gamma photon with the scintillation crystal results in emission of light, which is detected by the PMT. The light output is proportional to the gamma energy. The electronic system associated to the PMT analyses the pulses according to pulse amplitude (energy) and stores the results in a multichannel analyzer (MCA). Thus, energy and intensity are recorded, and the result is the gamma energy spectrum of the radiation source.

The NaI(Tl)-detector has a high intrinsic efficiency but a limited energy resolution. The scintillation crystals are provided in different sizes. The efficiency for high gamma energies increases with detector volume. Common counting equipment has cylindrical crystal sizes of 2" × 2" to 5" × 5" (height × diameter). Fig. 19 shows a NaI(Tl) detector 4" × 4". The larger the crystal the higher the price is. The detectors can be made quite rugged, and are suitable in field instrumentation.

FIG. 19. NaI(Tl) detector 4" × 4".

Semiconductor detectors: Modern detectors are mainly based on high purity germanium HPGE crystals, where a semiconductor junction is created by suitable elemental dopants on the crystal surface. A gamma ray interacting with the detector will result in an excitation of electrons from the valence band to the conduction band in the crystal, and a small electric pulse is created in a high-voltage field. Pulse height is proportional to gamma energy. The pulses are sorted and stored in a MCA.

The intrinsic efficiency of semiconductor detectors has, for many years, been lower than for NaI(Tl)-detectors. Today, it is however possible to purchase detectors with efficiencies 100% relative to that of a 3" × 3" NaI (Tl)-detector, but prices are very high. The main advantage of an HPGE-detector (Fig. 20) is its excellent energy resolution. This property may be indispensable for analysis of complex radiation sources. HPGE-detectors need cooling to liquid N_2 temperature during operation, are delicate and cannot generally be used in the field. Because of the high energy resolution, gamma tracer detection requires little sample preparation except for the extreme low-energy emitters' (i.e. ^{125}I).

FIG. 20. HPGe detector.

There are several ways to reduce the minimum detectable concentration in gamma detection:

- Increase the detector intrinsic efficiency: This is a matter of cost.
- Increase counting sample volume (constant activity concentration in the sample leads to higher total activity in the sample): There is a practical limit to the sample size.
- Optimize the counting geometry by shaping the counting sample: For a given radionuclide, a selected detection set-up and a certain sample volume there is an optimum shape of the sample volumes. For practical reasons these are most often cylindrical-like shapes.
- Enrich the tracer from a large into a smaller sample volume (increased total activity for a better sample counting geometry): This requires sample treatment either by liquid evaporation or by chemical separation. Sample treatment time and cost increase.
- Reduce the background level by effective detector shielding: This is most often done by passive shielding with lead walls (5-10 cm thickness) around the detector and sample.

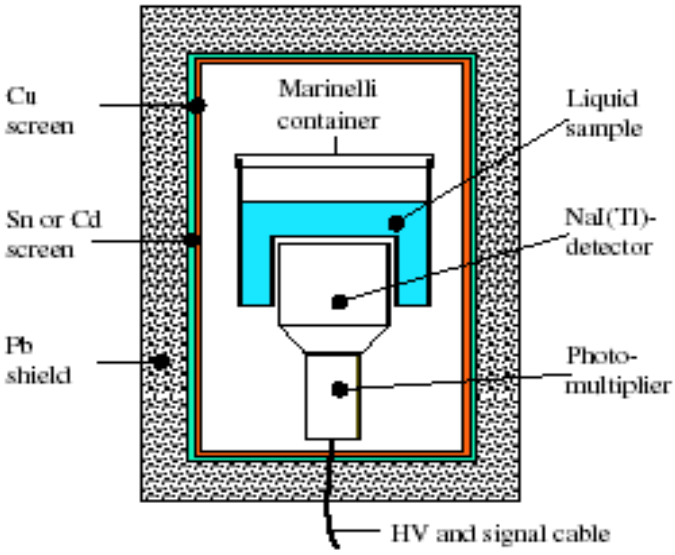


FIG. 21. Sketch of a common set-up for counting of gamma active liquid sample. 1000 ml Marinelli sample container, 3" × 3" NaI(Tl)-detector, Pb-shield (5-10 cm), a Sn (or Cd) screen to filter away Pb X rays generated by the sample activity in the Pb-shield, Cu filter screen to filter away Sn (or Cd) X rays generated by the Pb x-rays in the Sn (or Cd) screen.

A typical counting set-up for a NaI(Tl)-detector and a liquid sample in a Marinelli beaker is shown in Fig. 21.

With NaI(Tl)-detector based analytical equipment, detection limits < 0.2 Bq/L can be obtained using Marinelli beakers and reasonable counting times for common radionuclides like ^{22}Na, ^{60}Co and ^{125}I. For HPGE-detectors, the corresponding detection limits are < 0.1 Bq/L.

6. DATA TREATMENT

6.1. Data treatment for static measurement – RTD approach

The basic radiotracer methodology comprises accurate recording of a residence time distribution (RTD) experimental curve and its utilization for troubleshooting and diagnosis.

A sharp pulse of radioactive tracer is injected upstream of the vessel and a detector located at the inlet marks time-zero (Fig. 22). A second detector, located at the outlet, records the passage of the tracer from the vessel. The response of this detector is the residence time distribution (RTD). The experimental RTD is calculated from the count rate distribution at the outlet of the system I(t). A practical example is illustrated in Fig. 22.

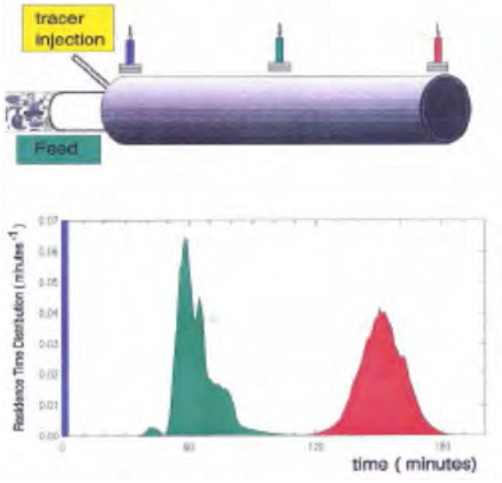


FIG. 22. Principle of RTD method.

The RTD function *E(t)*, is represented by equation (3):

$$E(t) = \frac{C(t)}{\int_0^\infty C(t)dt} \tag{3}$$

Where *C(t)* is the tracer concentration versus time at the outlet of the system.

The instantaneous injection (Dirac pulse) of tracer is normally applied in practice because is simple, less costly and rich in information. Fig. 23 presents the exemplary *E(t)* function; mean residence time (MRT) and its standard deviation (SD) are depicted.

MRT(τ) and SD (σ) have the following physical interpretation in relation to flow systems: MRT is directly related to flow rate Q and the effective flow volume V of the system $\tau = V/Q$, where V is the effective volume of the system and Q is a constant, volumetric flow rate.

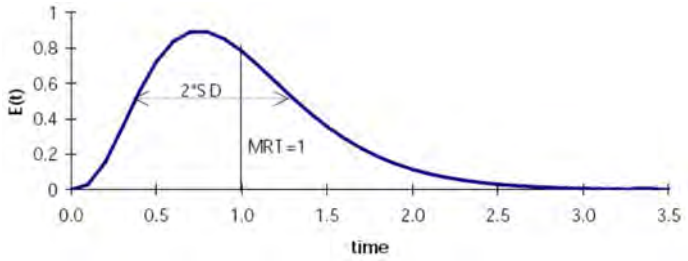


FIG. 23. The exemplary E (t) function with MRT and SD parameters shown.

6.2. Data preparation

The main steps in the analysis of the experimental response curve to obtain the correct RTD curve are presented in Fig. 24.

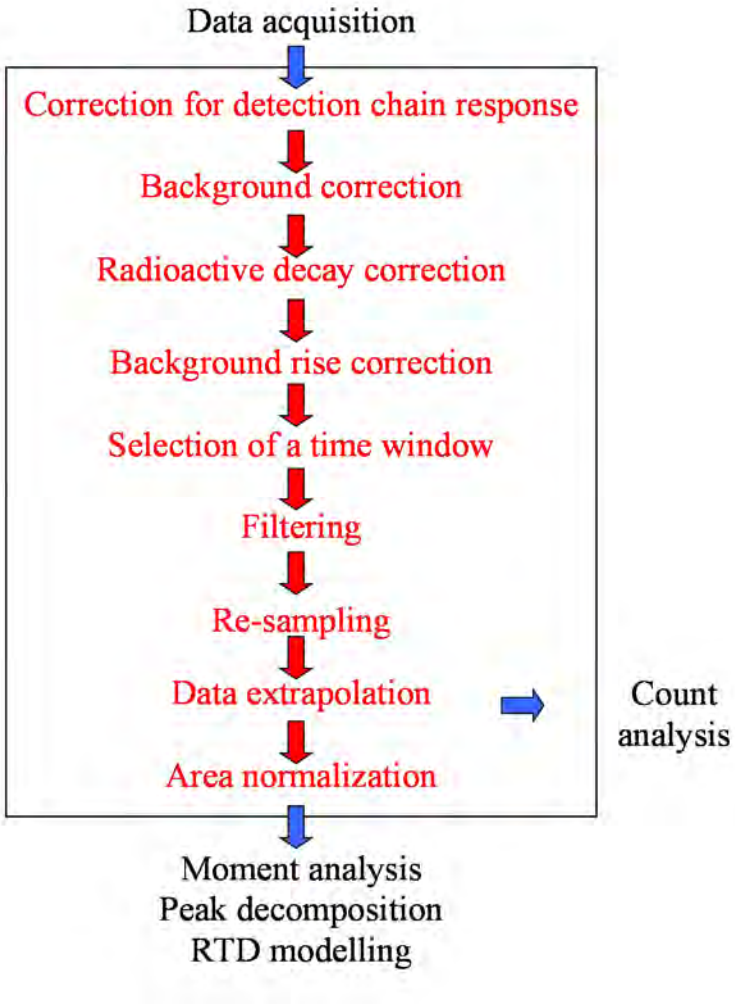


FIG. 24. Protocol analysis for experimental response curve.

6.2.1. Background correction

Prior to the injection of radiotracer into a system, it is necessary to measure the background radiation level, which is subtracted from the experimental data (Fig. 25).

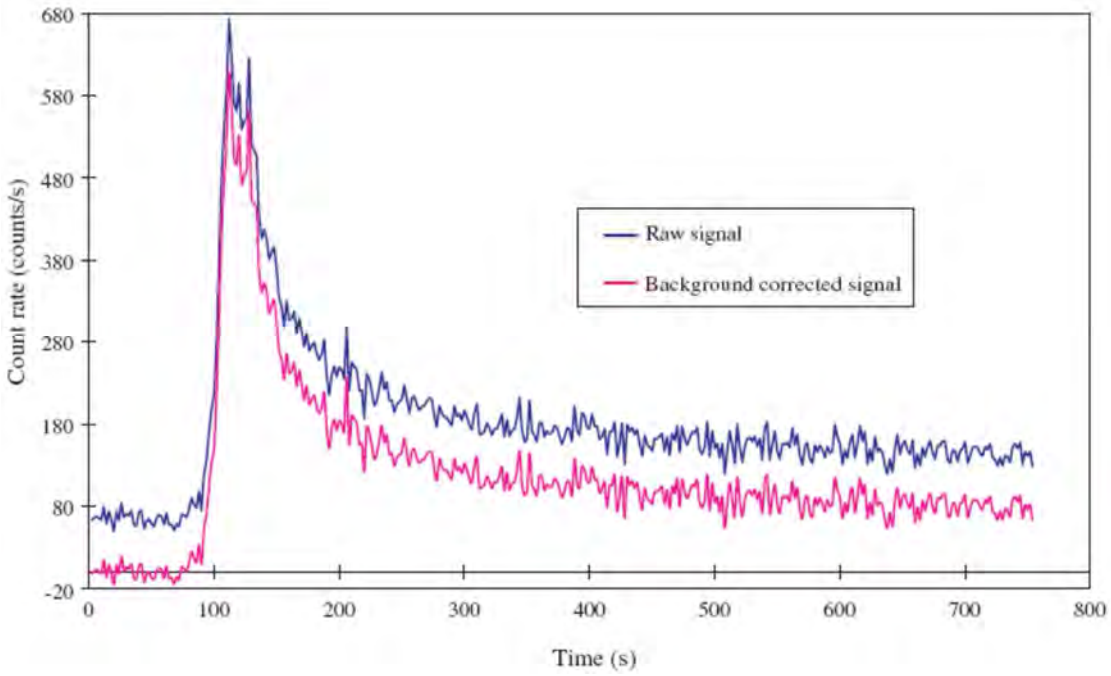


FIG. 25. Raw signal and background corrected signal.

6.2.2. Radioactive decay correction

Source activity at any time '*t*' may be calculated from the activity at some original or zero time using the following formula:

$$A = A_o e^{-0.693(\frac{t}{t_{1/2}})} \quad (4)$$

Where:

A is the activity at time '*t*'; A_0 the activity at time zero; t the time elapsed (same units as half-life) and $t_{½}$ the half-life.

Example: What is the activity of ^{32}P after 45 days? The initial activity is 10,000 Bq and the half-life is 14.3 days.

Solution: From equation (4)

A=10,000 × $e^{-[0.693 \times 45 / 14.3]}$

A= 10,000 × 0.11295

A= 1129.5 Bq

Fig. 26 shows the correction curve for radioactive decay.

6.2.3. Background rise correction

Sometimes, particularly in the case of particles transfer studies, the tracer can paste on the probes. This leads to an increase in the baseline radioactivity level as illustrated for three values in Fig. 27(a). It is necessary to correct the phenomenon (Fig. 27(b)).

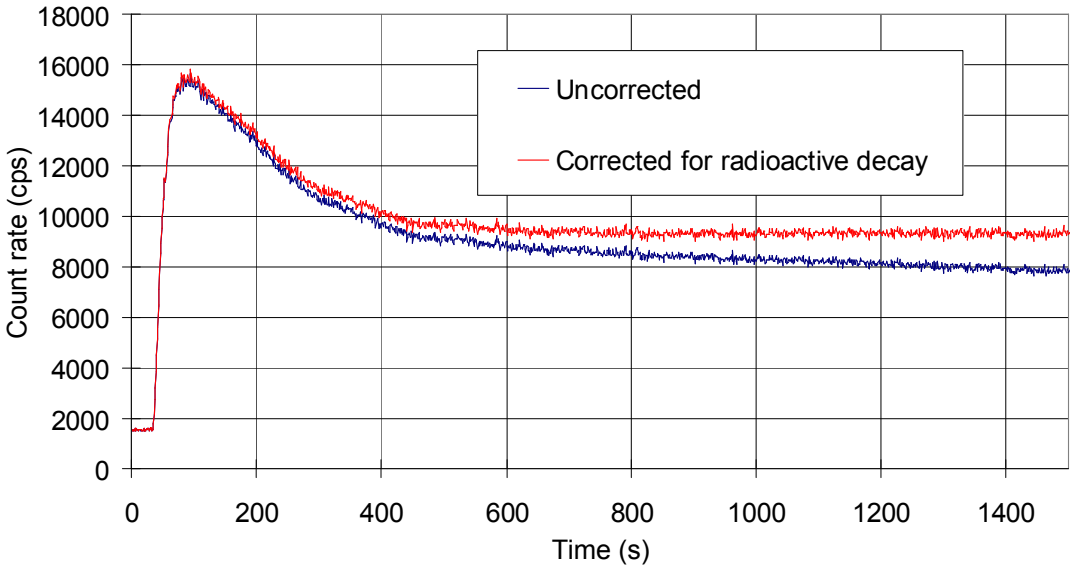


FIG. 26. Count rate correct for radioactive decay.

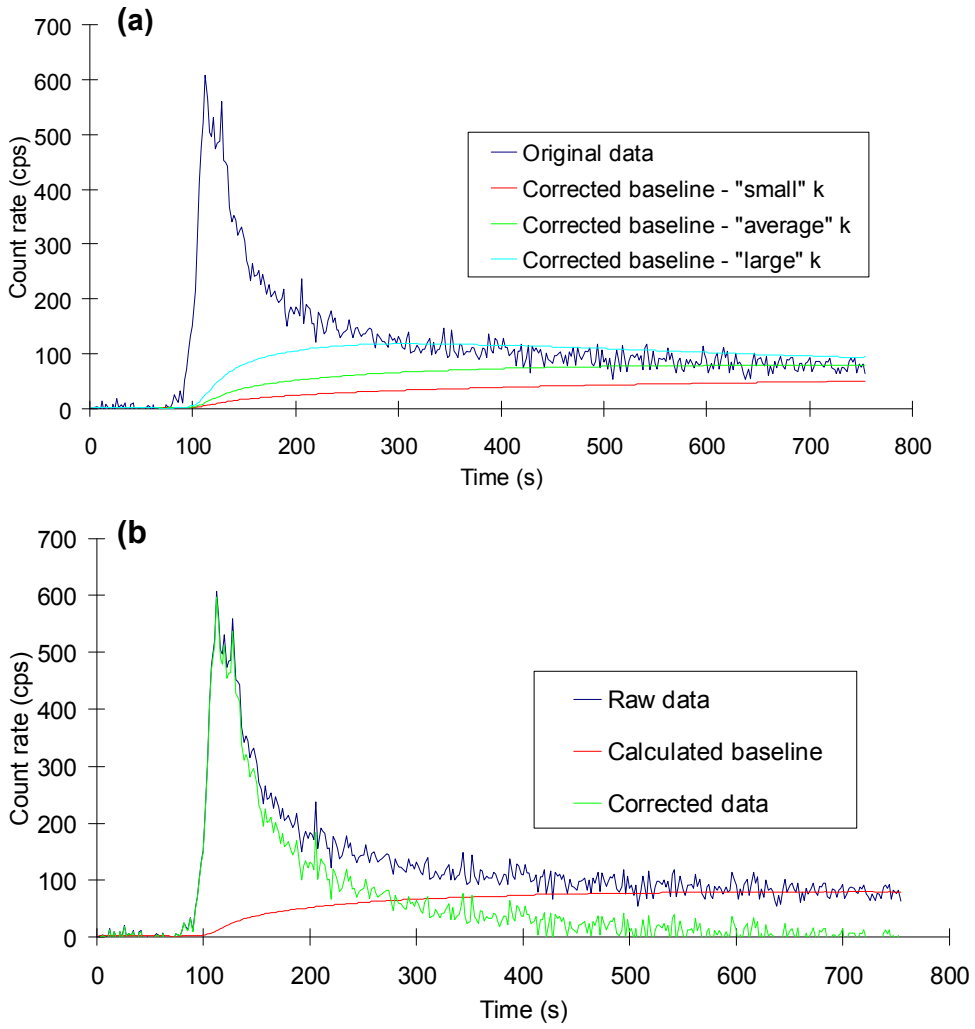


FIG. 27 (a) Original data and the baselines; (b) Original and corrected data.

6.2.4. Filtering (or smoothing)

The aim of filtering is to eliminate, or at least decrease, fluctuations due to counting statistics or electronic noise (Fig. 28).

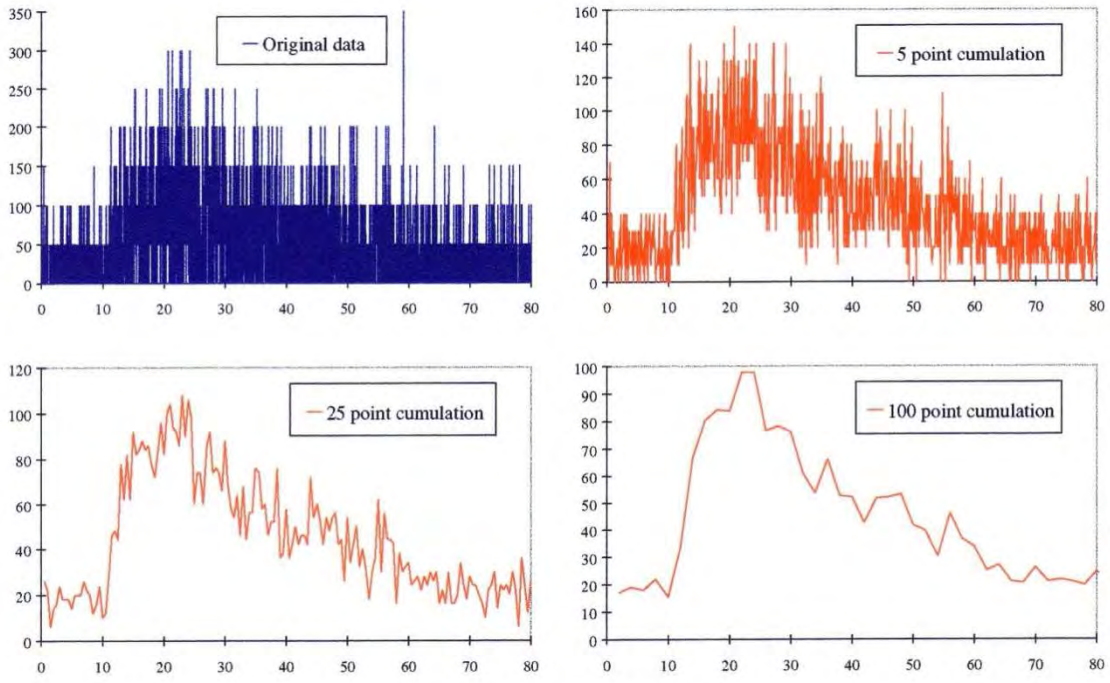


FIG. 28. Filtering (or smoothing) of fluctuations and noises in the experimental RTD curve by count cumulating.

Several methods are available. The Fourier transform is very effective; many high frequencies can be filtered without altering the general shape of the RTD experimental curve. The Fourier method requires that the data be sampled at equidistant (regular) intervals. Cumulating or re-sampling counts is simpler technique for smoothing fluctuations. Counts are cumulated by groups of 5, 25 and 100.

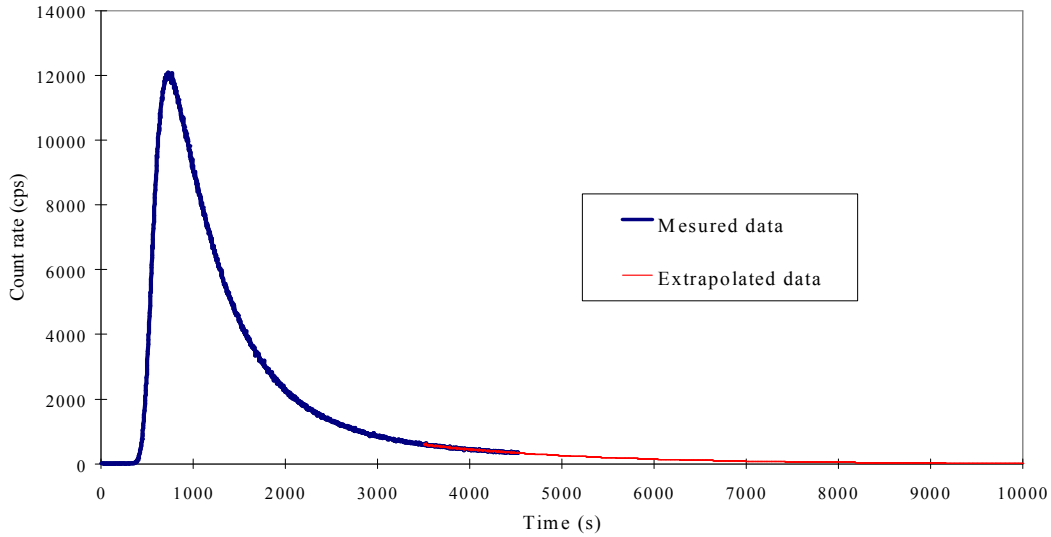


FIG 29. Incomplete experimental curve from tracer test.

6.2.5. Data extrapolation

Data extrapolation is needed when the end of the tracer curve is missed, or, in other words, count rates go back to zero after the end of the data acquisition sequence, as illustrated in Fig. 29. Mostly extrapolation is performed mathematically using exponential decay function.

The aim of data extrapolation is to extend the tracer curve in some plausible way. The most common procedure is to check that count rates decrease exponentially at the end of the experiment; this is easily done by plotting the logarithm of count rates versus time, which should exhibit a linear behaviour towards the end (Fig. 29). A decaying exponential function should then be adjusted on that part of the curve, and the data extended with this function until count rates are negligibly small. The number of extrapolated points should obviously be 'reasonable' (the meaning of 'reasonable' depends very much on available data and level of precision desired).

6.2.6. Area normalization

After all the previous corrections have been made, it is possible to exploit the data by time analysis or count numbers analysis. These methods provide valuable information on arrival time, modal time and transit time on the one hand, and dilution, flow rates, flow distributions, tracer mass balance on the other.

One last operation can then be performed, i.e. area normalization. This operation has several benefits. First of all, the influence of all the factors that affect the area of the curves but not their shape (injected activity, radiation attenuation by walls) is eliminated. It is then possible to compare readings from two experiments with different injected activities, or at two points with different wall thickness.

Secondly, the calculation of moments is simpler with area-normalized data. Thirdly, area-normalized curves are in certain cases RTD curves that have a precise meaning in terms of fluid population balance.

Area normalization is compulsory when doing model adjustment with software. The tracer concentration curve is normalized by dividing each data point by the area under the curve (i.e. the total count number) Eq.(5):

$$E(t) = \frac{\dot{n}_c(t)}{\int_0^\infty \dot{n}_c(t)dt} \tag{5}$$

where

$\dot{n}_c(t)$ is the corrected count rate (i.e. the result of all the previous operations),

$E(t)$ is the normalized data function.

Since count rates are actually known at discrete intervals Δt, values of function $E(t)$ can only be calculated at the same intervals, Eq.(6):

$$E_i = \frac{\dot{n}_{c,i}}{\sum_{1}^{N} \dot{n}_{c,i} \Delta t} \tag{6}$$

where

$\dot{n}_{c,i}$ is corrected count rate at time i.Δt.

The area under the new curves is therefore unity. If the tracer injection can be considered as a Dirac pulse, function $E(t)$ is the RTD at the measurement point. It is also customary to define the cumulative RTD function $F(t)$. This function can be useful when analyzing the behaviour of a system, Eq.(7):

$$F(t) = \int_0^t E(u)du \tag{7}$$

6.3. Data treatment for dynamic measurement

The picture here below (Fig. 30) shows a typical dynamic detection of tracer cloud during a dispersion study [11].

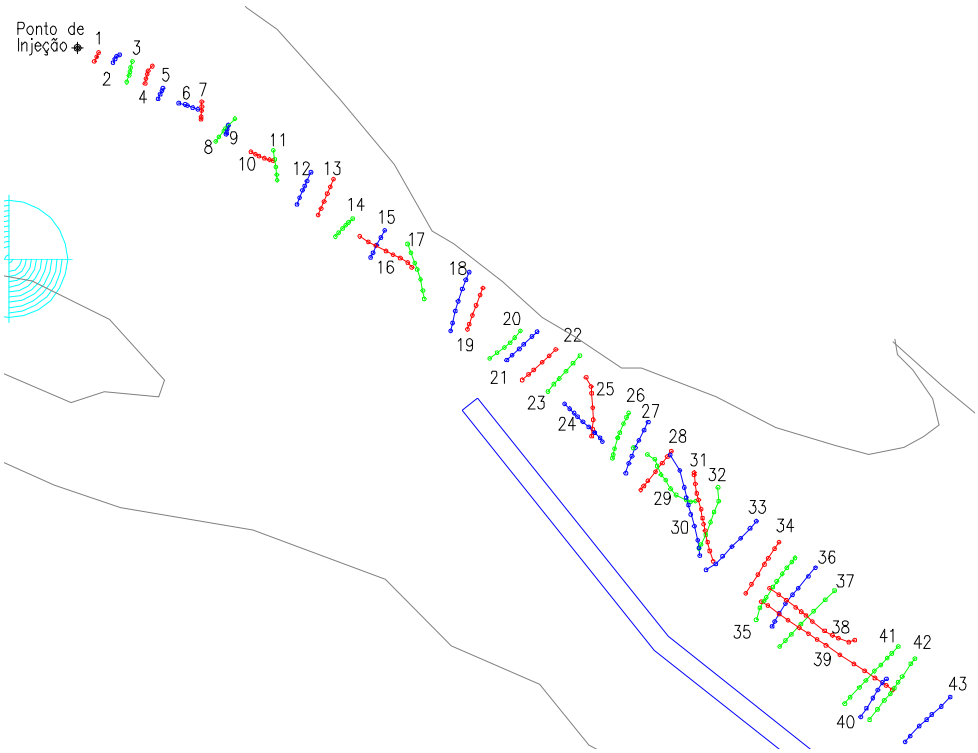


FIG. 30. Typical dynamic detection during a dispersion study.

The boat follows the tracer cloud performing criss-cross detection. The probes measure along each profile (transversal or longitudinal) the tracer concentration in counts per second (cps) versus time. These profiles have first of all to be corrected for background, decay and plating (pasting) as described in Section 6.2.

Additional corrections are needed to convert the temporal into spatial measurements:

- Offset corrections to take into account the horizontal distance between DGPS antenna and the detectors position;
- Corrections to take into account the variations in the probes depth. This correction is difficult in the absence of vertical mixing and is generally neglected if the probes are not equipped with pressure sensors.

7. FUTURE DEVELOPMENT IN RADIOTRACER TECHNOLOGY

The fundamental aspects of radiotracer technology have been in place since the 1960s. Since this time developments have been mainly due to improvements in computing including the use of DGPS for positioning, real time data acquisition, processing and visualization systems and the capacity to model tracer movement using finite difference or finite element models.

An important area of development for the future improvement of radiotracer technology is the development of new tracer forms or generators. However, there is an increasing challenge to obtain even the standard suite of tracers as fewer and fewer reactors and radiochemical production facilities are able to provide non-routine irradiations or isotope handling services.

7.1. Nano particle tracers

A radiotracer needs to be of an appropriate physical and chemical form so that it will behave and move in the same way as the process being studied. A tracer is selected from various radionuclides that can be produced in a relatively easy and low cost way from a nuclear reactor or accelerator. The chemical form is very important to ensure that the behaviour of the tracer is identical in essential respects to the component of the system under study.

Nano technology offers new possibilities obtaining the optimal tracers for the environmental hydrodynamics investigations. One of the examples is the radioactive gold (0.412~1.088MeV) nano particles coated with silica that can be chemically processed in such a way to track selectively water or suspended sediments (Fig. 31). Potentially, nano particles can be produced with the nuclides that have relatively large neutron cross section values such as Eu and Dy. These non-radioactive tracers are introduced into the water environmental system and quantitatively analyzed by the neutron activation technique after sampling.

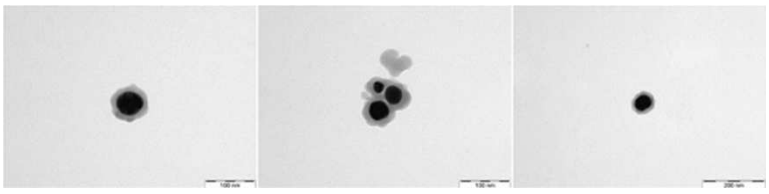


FIG. 31. TEM images of $^{198}Au@SiO_2$ nano particles with approximately 120 nm in diameter.

Additionally to the radioactive safety aspect, these nano particles need also to be assessed from the safety aspect due to their size itself. At this moment many studies are in progress to evaluate the risks coming from these nano particles. This risk has to be particularly evaluated because the size of the particles is a conservative parameter unlike the radioactivity which will disappear with the half-life of the nuclide. Radioactively labelled nano particles may also be used to study the fate and pathways of nano particles in the environment and their uptake into biota to evaluate their environmental impacts. This is already being done at the lab scale but could potentially be done in natural environments.

REFERENCES

[1] LOWENTHAL, G.C., AIREY, P.L., Practical Applications of Radioactivity and Nuclear Radiations (Chapters 8 & 9), Cambridge University Press (2001).

[2] INTERNATIONAL ATOMIC ENERGY AGENCY, Isotope Techniques in the Hydrogeological Assessment of Potential Sites for the Disposal of High-Level Radioactive Wastes, Technical Reports Series No. 228, IAEA, Vienna (1983).

[3] INTERNATIONAL ATOMIC ENERGY AGENCY, Radiotracer and sealed source techniques for sediment management, Report of the CM, held in Vienna, Austria, 21-25 April 2008.

[4] INTERNATIONAL ATOMIC ENERGY AGENCY, Fluvial sediment transport: Analytical techniques for measuring sediment load, IAEA-TECDOC-1461, Vienna (2005).

[5] INTERNATIONAL ATOMIC ENERGY AGENCY, Radiotracer Applications in Industry - A Guidebook, Technical Report Series TRS**,** 423**,** IAEA, Vienna (2004).

[6] INTERNATIONAL ATOMIC ENERGY AGENCY, Guidebook on Nuclear Techniques in Hydrology, Technical Reports Series 91, IAEA, Vienna (1983).

[7] INTERNATIONAL ATOMIC ENERGY AGENCY, Use of Artificial Tracers in Hydrology. IAEA-TECDOC-601, 71-130, IAEA, Vienna (1990).

[8] AIREY P L., HOWARD C. J., Comment on a General Property of Radionuclides, Journal of Radioanalytical and Nuclear Chemistry (JNRC) 257, **1-3** (2003).

[9] MAGILL, J., Nuclides 2000 – An Electronic Chart of Nuclides on CD – ROM(The European Commission, Institute for Transuranium Elements, Karlsruhe.)

[10] INTERNATIONAL ATOMIC ENERGY AGENCY, Regulations for the safe transport of radioactive material, Safety Standards Safety Requirements No. TS-R-1, Vienna (2009).

[11] MACHADO, M.L. et al., Orinoco River suspended sediment studies using ^{99m}Tc – Venezuela. In: Proceedings of The 5th International Conference on Tracers and Tracing Methods – TRACER 5, Tiradentes, Brazil, Belo Horizonte: CDTN (2008) CD-ROM

RADIATION PROTECTION AND REGULATION FOR ENVIRONMENTAL RADIOTRACER STUDIES

C.E. HUGHES[*], P. BRISSET[**]

[*]Australian Nuclear Science and Technology Organisation, ANSTO, Australia
[**]French Alternative Energies and Atomic Energy Commission, CEA, France/IAEA

Abstract

The use of radioactive tracers in the environment is subject to national regulation as well as International Standards and Guidelines. These address aspects of radiation protection during transport, deployment and in the environment following deployment and are relevant not only to radiation workers and members of the public, but also to non-human biota in the receiving environment. This paper outlines the regulatory framework and safety aspects relating to radiotracing and provides examples of radiation dose modeling for humans and non-human biota.

1. REGULATORY FRAMEWORK

The regulatory requirements for the conduct of radiotracer experiments are determined at a national level. These vary from country to country, but are commonly determined with reference to safety standards and guidelines issued by the International Atomic Energy Agency (IAEA) and the International Commission on Radiological Protection (ICRP). At the highest level IAEA Fundamental Safety Principles [1] are relevant to environmental radiotracer applications in requiring that:

- The benefits of an activity must outweigh the radiation risk (Principle 4);
- Activities should be optimized to ensure exposures are as low as reasonably achievable ('ALARA)' (Principle 5);
- Radiation exposure to ecosystems should be limited to ensure protection of populations of a species (as distinct from individual organisms) (Principle 7).

These principles are binding on the IAEA for its own operations, are applied by other sponsoring organizations for their own operations, and are recommended for use by States and national authorities in relation to their own activities.

The IAEA *International Basic Safety Standards* [2] lists the requirements for the use of uses of radiation and radioactive material. An environmental radiotracer application can be considered to be a planned exposure, as defined in this publication, which includes the practices of industrial, agricultural and research uses of radiation and radioactive material in its scope. In this publication the IAEA specifies dose limits for occupational and public exposure and defines the following requirements relevant to radiotracing:

- To assess the nature, magnitude and likelihood of radiation exposures;
- To comply with the requirements of the relevant regulatory body including seeking approval where potential exposure exceeds regulatory exemption levels and to assess potential impact on the environment as required.

1.1. National regulations and licensing of radiotracer studies

National regulations covering the use of radiotracers in the environment vary from jurisdiction to jurisdiction and we do not intend to address these comprehensively here. Generally regulation of radiotracing may cover three aspects:

- Licensing of an organization to possess and use radioisotopes;
- Licensing of individuals to handle radioisotopes;

- Approval for the release of a radioisotope tracer into the environment.

The injection of a radiotracer into the environment for the purposes of studying environmental processes is, in effect, a radioactive discharge and is considered by many national jurisdictions to be a form of radioactive waste disposal. In some jurisdictions the limits applied to environmental radiotracer studies are those derived for waste discharges.

In all known examples there is a requirement for pre-approval of each radiotracer release either by the national regulator (e.g. France, Sweden, Indonesia, Uruguay) or in some cases by a radiation safety body within the applicants organization (e.g. Australia, Brazil) where general licensing for radiotracer studies has already been obtained. The application for approval requires a radiation safety assessment of the release to be carried out addressing the following aspects:

- Purpose of the experiment and justification for the use of radiotracers;
- Location of the experiment and predicted radioactivity levels in the environment during and following the release;
- Radiological dose calculations for planned radiation exposure of workers and the public and potential unintended exposures;
- Safety measures and monitoring to be employed to minimize radiological dose and risk of injury to workers and the public during and following the experiment.

In the majority of jurisdictions a radiological risk assessment is only required for humans potentially exposed to the radiotracer. An underlying assumption is that the environmental impact of radiotracer releases is negligible due to the rapid dispersion of the radiotracer in the receiving environment. However, in some jurisdictions, particularly Australia, it is required also to address the environmental impact of the radiotracer study in conducting the risk assessment.

2. SAFETY ASPECTS

Radiological safety aspects considered when conducting field radiotracer investigations include:

- Safe transport of radioactive material to the experimental site;
- Methodology and risk assessment for handling of radioactive material during injection into the environment, including predicted radiation dose to the operators under normal conditions;
- Risk assessment and mitigation planning in the case of an abnormal situation during injection, including potential radiation dose to the operators;
- Post injection hazards and radiation dose to the public and environment.

The general guidelines for radiological safety including handling of of radioisotopes have been issued by the International Commission on Radiological Protection (ICRP) and the International Atomic Energy Agency (IAEA) from time to time [e.g. 1, 3, 4]. Navada [5] also outlined considerations specifically for the safe use of artificial radioactive tracers in hydrology.

Generally safety regulations include justification of the use of radiotracer, optimization of radiation exposures and annual dose limits in order to prevent unnecessary exposures. The

justification implies that the competent authority should not allow the use of radiation unless there arises a net positive benefit from its use.

The design of a radiotracer experiment has to ensure optimization of radiation exposures. All the exposures have to be as low as possible (ALARA). The optimization of radiation exposures primarily depends upon distance, time and shielding.

- The dose rate at a point varies inversely proportional to the square of the distance between the source and the point. Therefore a radiation worker has to maintain maximum possible distance from a radiation source.
- The dose received is directly proportional to the time spent in handling the source. Thus the time of handling of the source should be minimized.
- The radiation intensity at a point varies exponentially with the thickness of shielding material. Thus an optimum thickness of the shielding material has to be used between the source and a radiation worker

The annual dose limits have to be taken in account and no individual should be exposed more than the prescribed limit. The dose limits recommended by ICRP are 1 mSv/year for public and 20 mSv/year for a radiation worker [6].

The packaging and transportation of radioactive material is governed by stringent rules and regulations. The first set of rules and regulations for safe transport of radioactive material was published by International Atomic Energy Agency (IAEA) in 1961 based on ICRP recommendations, which now forms basis for all national and international regulations. These regulations are constantly reviewed, revised and updated from time to time by ICRP and IAEA. Recently IAEA has published revised regulations for safe handling and transport of radioactive material [7].

3. HUMAN RADIOLOGICAL DOSE ASSESSMENT FOR RADIOTRACER RELEASE

The methodology for dose assessment covers the following aspects for routine operational exposure:

- Calculation of external dose based on time of exposure, distance and shielding;
- Calculation of internal dose based on ingestion or inhalation.

The risk assessment approach also requires assessment of realistic scenarios for non-routine operation. Examples of such scenarios include:

- Loss of shielding;
- Failure of containment resulting in spill, operator contamination or clean-up requirement;
- Failure of deployment e.g. loss of intact source overboard, or failure of deployment or partial deployment. This may result in additional exposure during retrieval or redeployment.

Dose to the public is also considered, generally for the maximally exposed person or critical group. The same methodology is used as for dose to the operator, however different dose limits generally apply. Possible scenarios for exposure of members of the public to a radiotracer include:

- Immersion in near-field of radiotracer injection - child or adult;
- Ingestion of water from plume (based on ingestion amounts used to calculate ALI);

- Ingestion of plant or animal from tracer plume (food chain transfer).

3.1. Dose to the operators under normal operation conditions

In this section we will consider only exposure in normal working conditions (calculation of external dose based on time of exposure, distance and shielding), that means we will not consider accidental situations such as loss of shielding, failure of containment with the associated external or internal contamination, etc.). The dose assessment will be done using Microshield software but the same results can be obtained using the Beer-Lambert equation.

3.1.1. General aspects of dose assessment to operators

We will consider here two types of experiments, dispersion studies and bed-load transport studies and the most typical radiotracers used to perform these studies.

- dispersion studies : ^{99m}Tc, ^{198}Au, ^{131}I, ^{181}Hf
- bed-load transport studies: ^{192}Ir

The transportation of the tracer is not taken into account here and is assumed to be conducted under safe conditions according to the transport regulations.

Case 1. Dose from radiotracer injection for dispersion studies using French CEA apparatus:

The scenario is based on the French CEA injection and labelling device, shown in Fig. 5 of the paper titled "Radiotracer methodology", from Brisset et al., included in this publication.

The device can be modelled by the following geometry (using Microshield software).

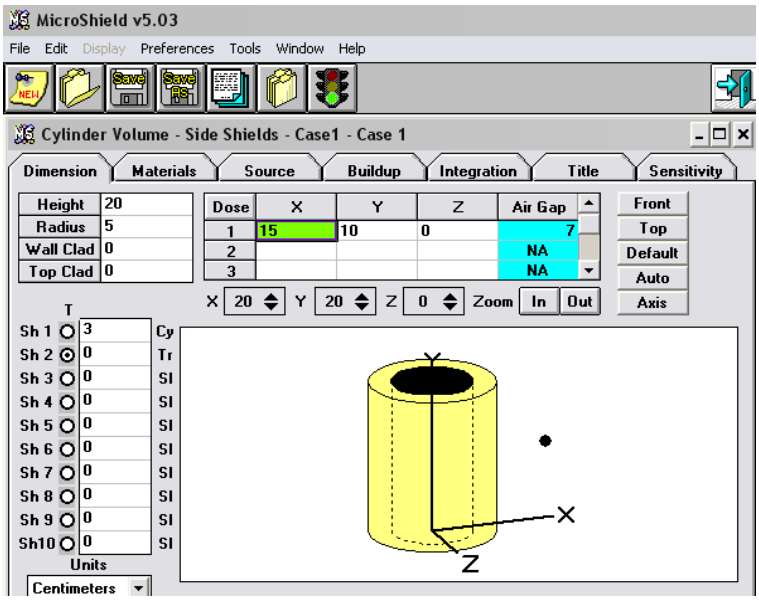


FIG. 1. Schematic for operator dose Case 1.

The main parameters of the calculation (Fig. 1) are:
- activity of the nuclide: 37 GBq
- distance of the operator to the axis of the source: 15 cm and 2 m
- duration of the injection operation: 1 min
- shield: lead 3 cm thick

The main results are presented in Table 1.

TABLE 1. DOSE FROM 37 GBq RADIOTRACER INJECTION FOR FOUR RADIONUCLIDES

Nuclide Activity 37 GBq	Dose rate at 15 cm from the source axis i.e. 7 cm from the shield wall (mSv/h)	Dose rate at 2 m of source axis (mSv/h)	Dose for operation 1 min (μSv)
^{99m}Tc	$3.8\ 10^{-20}$	$9.8\ 10^{-23}$	$16\ 10^{-22}$
^{198}Au	0.2	$0.9\ 10^{-3}$	$15\ 10^{-3}$
^{131}I	0.26	$2\ 10^{-3}$	$33\ 10^{-3}$
^{181}Hf	0.22	$1.7\ 10^{-3}$	$28\ 10^{-3}$

We can conclude that in normal conditions, the dose to operators is very low and always lower than 1 μSv per injection.

Case 2. Safety assessment for radiotracer tests in a river in Korea – ^{131}I – 370 MBq

In order to get permission for radiotracer experiments in the field, the radiation safety assessment should show that there will be no environmental impact in the area where the experiment will be performed. On the basis of the geological and hydrological information, the travelling time of the radiotracer and its concentration can be roughly predicted. At the stage of the experiment planning, uninhabited area is preferred as long as the experiment condition is acceptable in order to avoid any unnecessary exposure to the public. It needs to be sure that no water is taken from the site for either drinking or irrigation purpose during the investigation.

As an example, when 370 MBq of ^{131}I is injected in a river with 0.5 m/sec of the average velocity and 5.66 m^3/sec of the average flow rate and the detection is made 3 km downstream, the diluted concentration of the radioisotope would be 10.9 Bq/L which is lower than the maximum permissible concentration to the public in the regulations. Since the radioactivity decays as a function of its decay constant in addition to the dilution, the concentration would be much lower than the calculation.

The dose rate from 370 MBq of ^{131}I (gamma dose rate constant, $\Gamma = 5.72 \times 10^{-2}$ mSv·m^2·GBq^{-1}·h^{-1}) at 1 m distance is 21.2 μSv·h^{-1}. When the radioisotope is kept in a container with 15 mm thick Pb the dose rate reduces to 1 μSv·h^{-1} (R_1) (Eq.(1)).

$$R_1 = \Gamma \times Q \times d^{-2} \times B \times e^{-\mu\rho x} \quad (1)$$

$= 5.72 \times 10^{-2}$ mSv·m^2·GBq^{-1}·h^{-1} × 0.37 GBq × 1 m^{-2} × (1.5 × exp(-0.2 cm^2/g × 11.3 g/cm^3 × 1.5 cm))

$= 1$ μSv·h^{-1}

(d: distance, B: Build-up factor, μ: mass attenuation coefficient, x: thickness of Pb, ρ: density of Pb) [1]

It takes approximately 3 minutes to perform the radiotracer injection including taking the radiotracer from a vial and mixing it with water and the distance between the source and the body is 0.75 m. The exposure dose to the body from one injection procedure would be 0.09

[1] Some measurement units used in this paragraph are not converted to SI units, as in many national regulations still these non-SI units are commonly applied.

μSv (R_2) while the exposure to the hand at 0.25 m away from the source for 10 seconds would be 0.04 μSv (R_3). It is clearly understood that the total dose from a series of radiotracer injections would be much lower than the regulations.

$R_2 = 1\ \mu Sv \cdot h^{-1} \times (1\ m)^2/(0.75\ m)^2 \times 3\ min \times 1\ hr/60\ min = 0.09\ \mu Sv$
$R_3 = 1\ \mu Sv \cdot h^{-1} \times (1\ m)^2/(0.25\ m)^2 \times 10\ sec \times 1\ hr/3600\ sec = 0.04\ \mu Sv$

Case 3. Bed load transport studies

Here the calculations are based on the French injection system shown in Fig. 2.

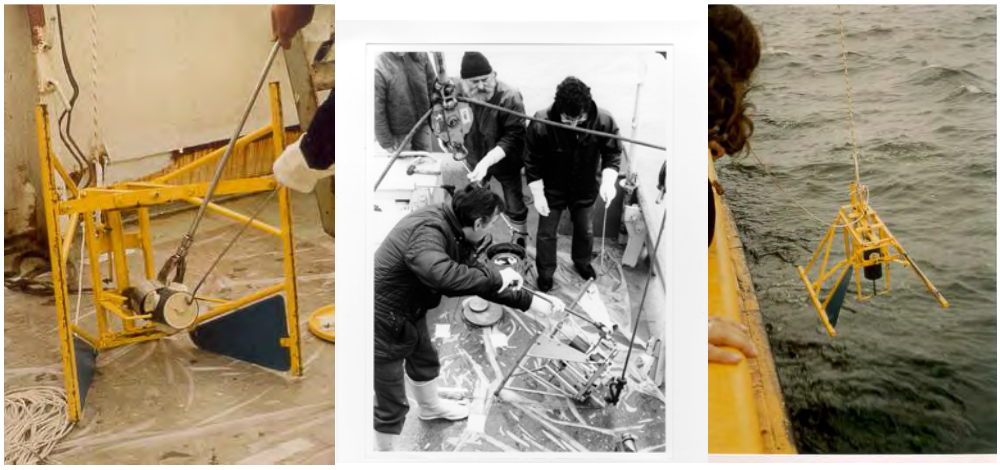


FIG. 2. Bed load transport studies.

In this system the source is typically 37 GBq of ^{192}Ir glass and is not shielded.

The system can be simulated by the following geometry (Fig. 3) using Microshield software.

With these conditions (37 GBq^{192}Ir, distance of operator to the source 1m) the dose rate to the operator is 1.25 mSv/h thus the dose received by the operator is 10 μSv for the duration of the injection operation (30 sec).This value has been many times confirmed by experience.

As a conclusion of this section we can say that the dose to the operator is less than 10 μSv per injection of 37 GBq tracer in any case in normal conditions.

3.2. Dose to the public

For the public the first safety precaution is to define an exclusion area around the tracer manipulators during injection. The presence of members of the public has to be strictly prohibited inside this exclusion area.

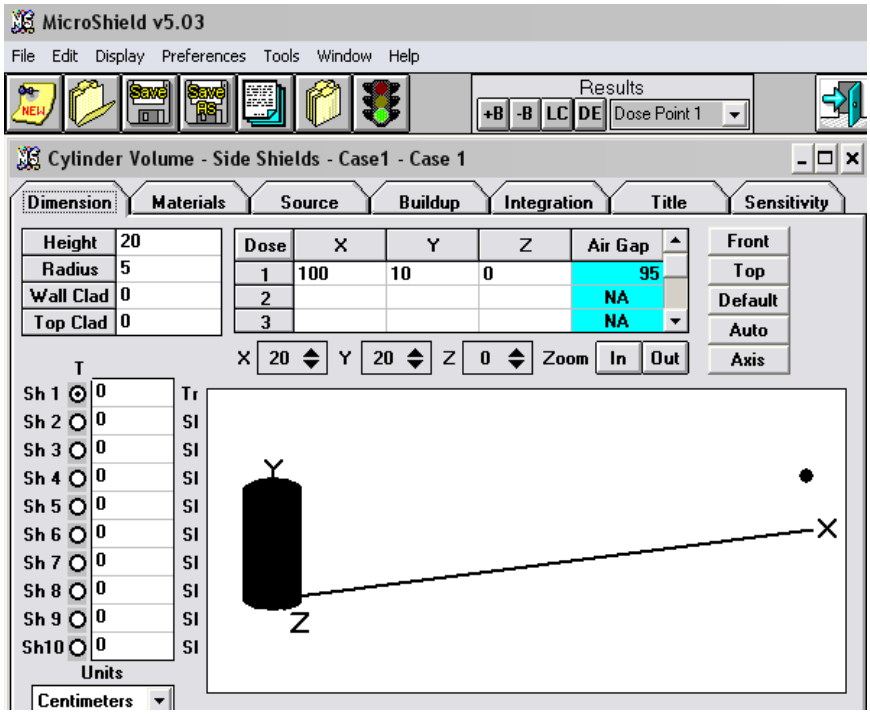


FIG. 3. Schematic for operator dose Case 3.

Thus the scenarios for exposure of members of the public to radiotracer can be limited to:

- Case 1: Immersion in near-field of radiotracer injection, only for dispersion studies;
- Case 2: Ingestion of water from near-field, only for dispersion studies;
- Case 3: Immersion on the sea bed in the near-field deposit area, only for bed-load studies;
- Case 4: Ingestion of sand grains, only for bed-load studies.

Ingestion of plant or animal from tracer plume (food chain transfer) will not be considered here because of the short half-life of the tracers used in dispersion studies and because the tracer grains are in glass matrix for bed-load transport studies (and thus not able to be metabolised). However, such a scenario has been considered in many risk assessments and the dose found to be infinitesimal.

Case 1. Immersion in near-field of radiotracer injection (Table 2)

We will consider here somebody immersed at the centre of a cylinder of tracer labelled water (dimensions diameter 10 m, height 10 m, Fig. 4) containing 37 GBq of radiotracer.

TABLE 2. DOSE FOR IMMERSION IN NEAR-FIELD OF RADIOTRACER INJECTION

Nuclide **Activity 37 GBq**	**Dose rate at the centre of tracer volume** **(μSv/h)**	**Dose for 1 hour exposure** **(μSv)**
^{99m}Tc	0.83	0.83
^{131}I	2.2	2.2
^{198}Au	2.4	2.4
^{181}Hf	3.2	3.2

Radioactive decay and dispersion are not taken into account for these calculations.

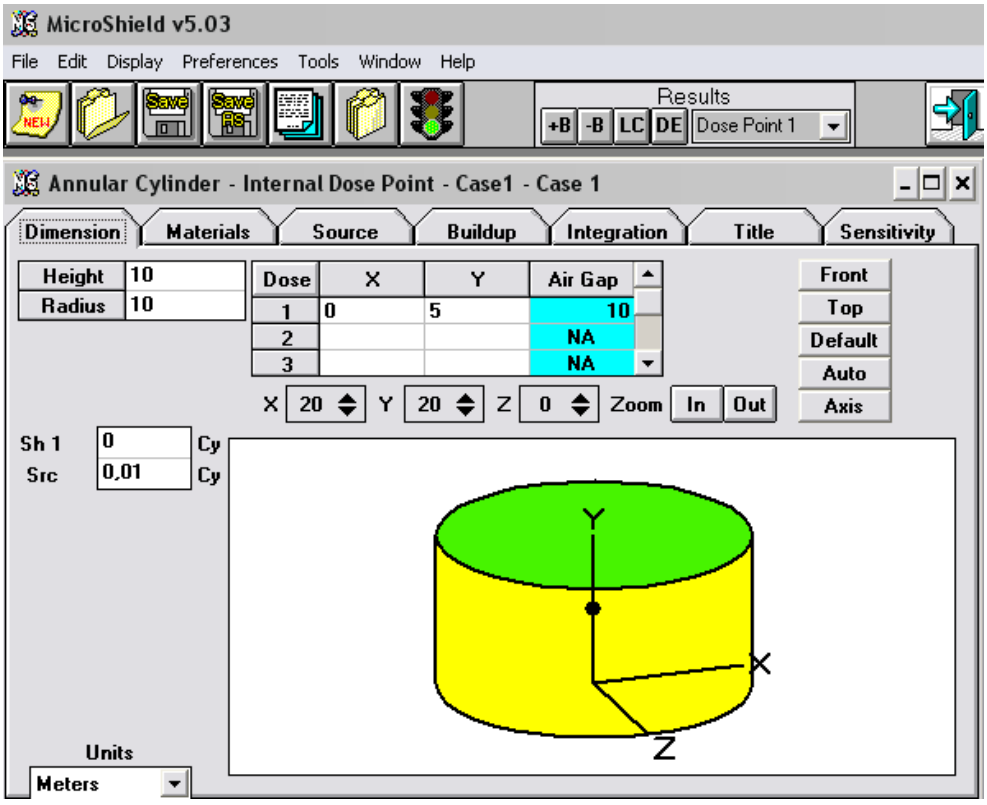


FIG. 4. Schematic for public dose Case 1.

Case 2. Ingestion of water from the cylinder defined in the previous case (Table 3).

TABLE 3. NUCLIDE ACTIVITY AND INGESTION DOSE

Nuclide Activity 37 GBq	**Max activity for ingestion All compounds (Bq)**	**Concentration in the 850 m^3 volume (Bq/L)**	**Volume to be ingested for max activity (litres)**
^{99m}Tc	9 x10^8	43500	20700
^{198}Au	2 x10^7	43500	460
^{181}Hf	1.8 x10^7	43500	414

The data for the maximum activity for ingestion are from Delacroix et al [3]. This activity corresponds to the activity resulting in the dose limit to the people. We consider here the lower acceptable activity (i.e. the most dangerous compound). In these examples it is not physically possible the reach the dose limit in any case and by many orders of magnitude.

Case 3. Immersion on the sea bed in the near-field deposit area

Practically we consider here a diver staying on the sea bed at 10 cm from the centre of a disc of 10 m diameter where 37 GBq of iridium-192 glass is homogeneously dispersed (Fig. 5). We speak here of a diver because in such experiment the tracer is released at the sea bottom under typically 20 to 30 m water depth. The tracer is always submersed and never accessible without special means such as diving tools.

This geometry can be modelled as shown in the following scheme (from Microshield software, Fig. 5).

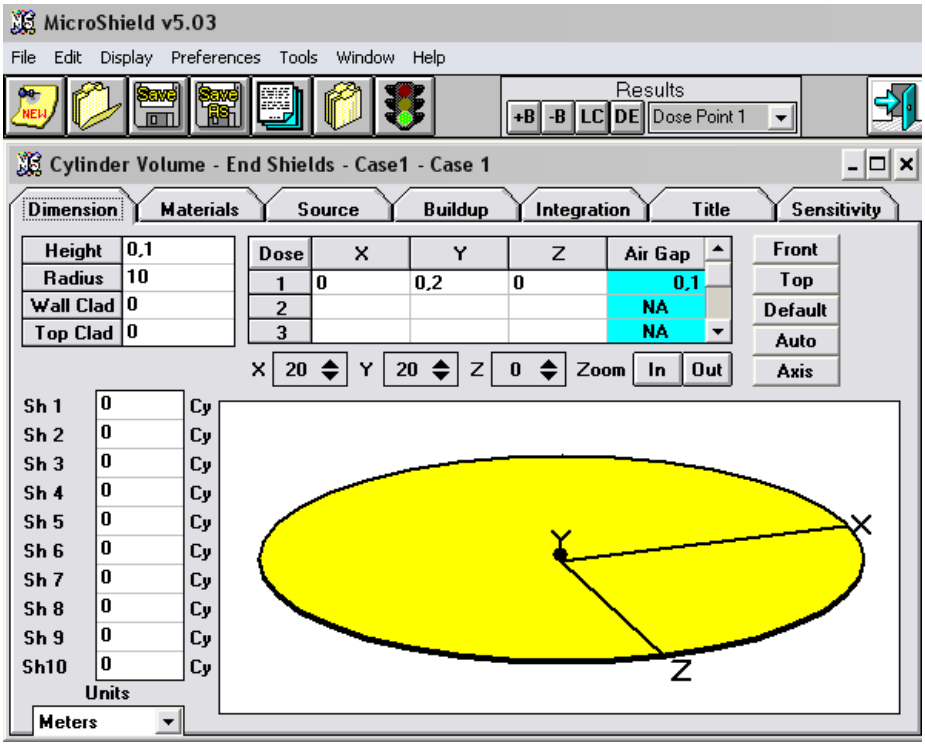


FIG. 5. Schematic for public dose Case 3.

The dose rate calculated at the centre of the disk is 8 µSv/h. That means that the diver would have to stay for 125 hours to reach the 1 mSv dose limit for the public, which is physically not possible.

Case 4. Ingestion of iridium-192 glass radioactive grains

We consider a typical experiments using 37 GBq in 450 grams of ^{192}Ir glass (particles size 155 µm). The maximum activity acceptable by ingestion is 24 MBq for ^{192}Ir.

The activity of 1 grain of 155 µm diameter (for 37 GBq in 450 grams) is 385 Bq.

It is thus necessary to eat 63 000 grains (or 295 mg) of the undispersed source to reach the limit of activity. This not possible in any case because the undispersed source is accessible only in case of accident during injection and the public is not allowed in the exclusion area.

After injection we can consider that the tracer is mixed with natural sand on a 1 cm thickness disk of 10 m diameter. The volume of traced sand is thus: 0.78 m^3 and the concentration is 18000 Bq/g.

With this hypothesis it is thus necessary to eat 1340 grams of sand from the tracer plume to reach the activity limit. This is not physically possible.

4. ENVIRONMENTAL RADIOLOGICAL DOSE ASSESSMENT FOR RADIOTRACER RELEASES

4.1. Assessing the impact of radiation on non-human biota

Only recently has the requirement to assess the impact of ionising radiation on non-human species started to be introduced into national regulations and standards. Previously it was

considered that the requirement to protect humans from radiation this would be sufficient to protect other species. In 1991 the International Commission on Radiological Protection [6] stated that:

'The Commission believes that the standard of environmental control needed to protect man to the degree currently thought desirable will ensure that other species are not put at risk. Occasionally, individual members of non-human species might be harmed, but not to the extent of endangering whole species or creating imbalance between species.'

In many circumstances this may be true as mammals have been shown to be the most radiosensitive taxa, as shown in Fig. 6 [9]. However, the characteristics of non-human species and their response to radiation are far more variable and less well defined than those for humans. In addition the exposure pathways for humans cannot be assumed to apply for non-human species as humans are often not present in parts of the environment where other species may be exposed to radiation.

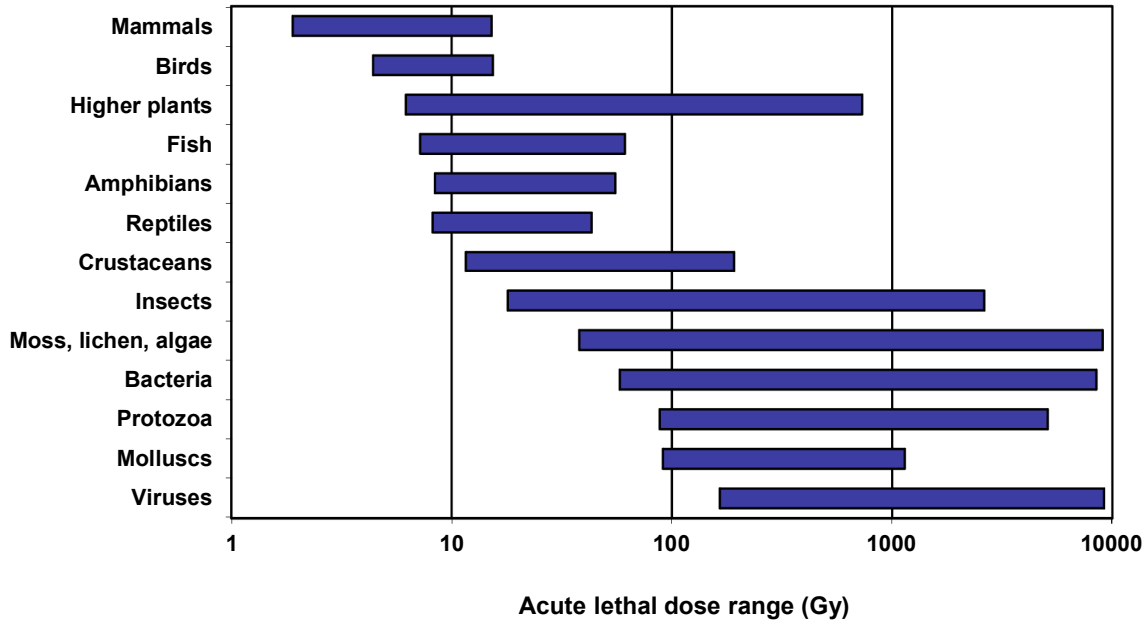


FIG. 6. Radiosensitivity of various taxa [9].

In 2003 the ICRP [10] responded to increased concern about environmental impacts of radioactive releases and the increasing pressure to develop policy in this area and issued ICRP Publication 91 *A framework for assessing the impact of ionising radiation on non-human species*. This recommended the development of a systematic framework *'to assess the relationships between exposure and dose, and between dose and effect, and the consequences of such effects, for non-human species, on a common scientific basis'* using the concept of reference animals and plants. Since then the ICRP [11] has developed a small set of reference organisms for which the relationship between radiation exposure, dose and effect have been described.

The following groups have been active in the development of environmental dose assessment methodologies, tools and guidelines:

- EC Euratom Projects EPIC, FASSET, ERICA and, most recently, PROTECT have developed a framework and integrated approach for the assessment of environmental impact of ionising radiation in European ecosystems which have produced the EPIC impact assessment framework [12], FRED dose effects database and ERICA dose assessment tool (http://wiki.ceh.ac.uk/display/rpemain/Radiological+protection+of+the+environment+-+sharing+knowledge).
- US Dept of Energy have developed a technical standard [13] and dose assessment tool, RESRAD-BIOTA (http://web.ead.anl.gov/resrad/).
- UK Environment Agency R&D 128 Publication *Impact Assessment of Ionising Radiation on Wildlife* [14] developed an easy to use spreadsheet based dose assessment tool for a limited range of organisms and radionuclides (http://wiki.ceh.ac.uk/pages/viewpage.action?pageId=115016183).
- French agency for radiological and nuclear safety IRSN have developed the EDEN software package [15].
- IAEA EMRAS I and II Programmes (http://www-ns.iaea.org/projects/emras/emras2/default.htm) and Coordination Group on Radiation Protection of the Environment (http://www-ns.iaea.org/tech-areas/waste-safety/enviro-protection.asp#2).
- ICRP has established a new ***Committee 5*** which is concerned with radiological protection of the environment (http://www.icrp.org).

4.2. Dose assessment methodology

Non-human biota dose assessment typically involves a number of steps as outlined in Fig. 7 by Copplestone et al [14].

Methods to calculate dose once the concentrations and uptake are determined are based on standard human dosimetry techniques. Dose per unit intake or concentration (or dose conversion coefficient, DCC) for a range of organism geometries and radionuclide energies is determined using Monte Carlo modelling. Internal dose from ingestion of radionuclides and external dose from immersion in or proximity to contaminated media can then be calculated.

Dose conversion coefficients for many isotopes have been determined for a variety of organisms based on their geometry by the ICRP [11], Copplestone et al [14] and are calculated for any isotope or geometry by the ERICA software. Concentration factor data to determine the uptake of radionuclides biota is also incorporated in existing software and some values have been published by the IAEA [16, 17]. However, available data cover only a small number of organisms and isotope. The flow diagram of the impact assessment approach is represented in Fig. 7.

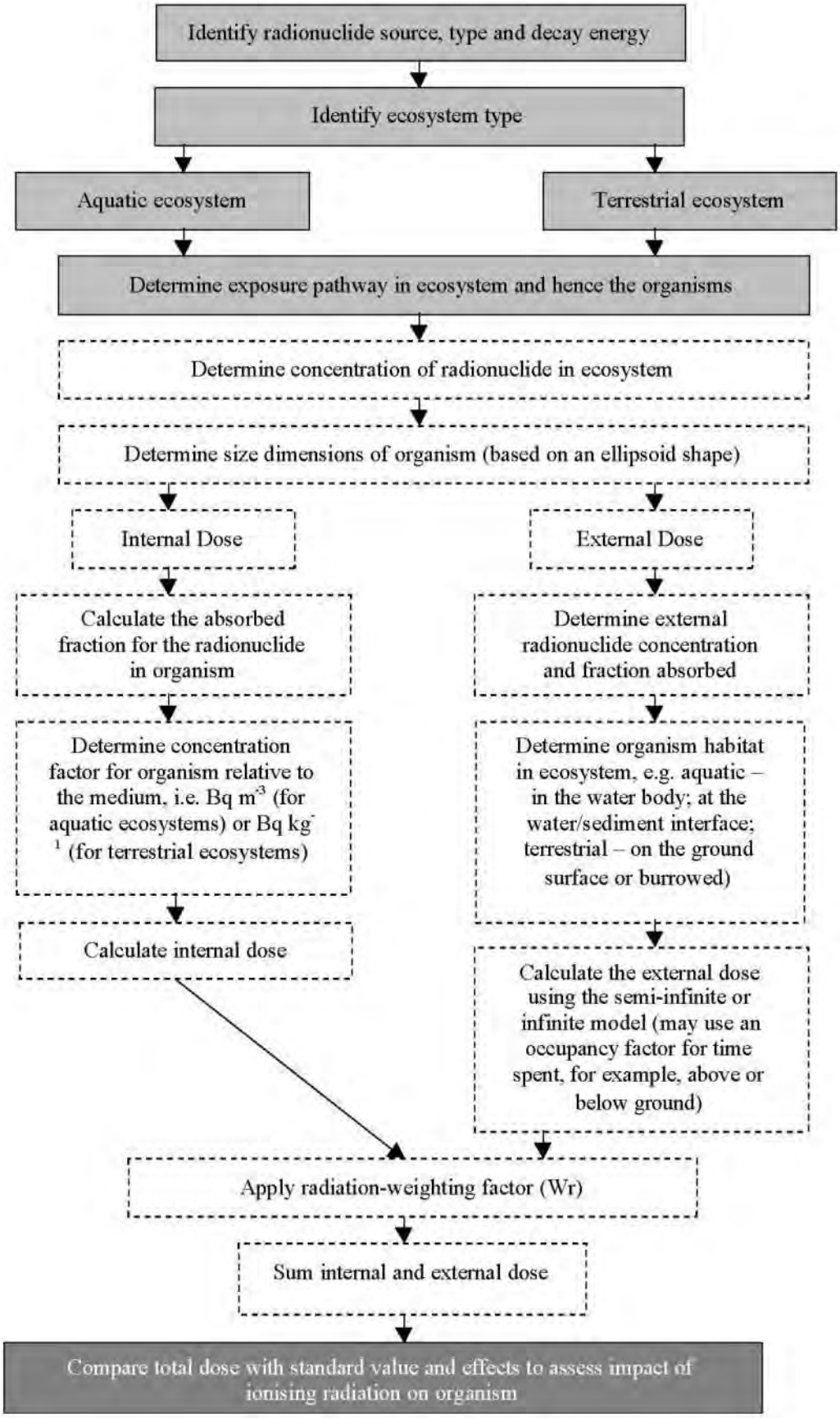


FIG. 7. Flow diagram of the impact assessment approach (from Copplestone, [14]).

A tiered approach to dose assessment is followed in both the RESRAD-BIOTA and ERICA models. After determining potential exposure scenarios and obtaining data on actual or possible radionuclide concentrations and species present Tier 1 assessments simply compare radionuclide concentrations with screening levels for each organism selected. If concentrations exceed the screening level then a Tier 2 assessment allows you to define the organisms present, their occupancy, and parameters such as the concentration factors, radiation weighting factors and K_ds. Doses calculated are then compared with dose guidelines (e.g. Table 4 or ICRP [11]) selected by the user. Finally Tier 3 provides a probabilistic dose assessment incorporating uncertainty and sensitivity analyses, temporal and spatial averaging and the ability to incorporate extra dose-effects data.

4.3. Dose guidelines for non-human species

No generalized dose limits have been set for exposure of non-human species to ionising radiation. It is important, however, to have some guidelines with which to compare any dose assessment outcomes in order to establish whether an exposure is acceptable. A number of reports have sought to establish guide dose levels (Table 4) and most recently the ICRP [11] have proposed a set of derived consideration reference levels (DCRLs) relevant to their set reference organisms. These levels vary from 0.1-1 mGy d^{-1} (4-40 μGy hr^{-1}) for mammals and birds, 1-10 mGy d^{-1} (40-400 μGy hr^{-1}) for fish and amphibians and 10-100 mGy d^{-1} (400-4000 μGy hr^{-1}) for invertebrates and seaweed, showing a similar relative radiosensitivity to that proposed by Whicker and Schultz [9, Fig. 6].

TABLE 4. GUIDE DOSE-RATE LEVELS BELOW WHICH NON-HUMAN ORGANISMS ARE PROTECTED

Units μGy hr^{-1}	**IAEA [18, 19])**	**NCRP [20]**	**UNSCEAR [21]**	**ICRP [11]**	**US DOE [22]**	**Canada [23]**	**France - IPSN [24]**
Mammals			400	4-40			
Plants			400	4-4000			
Freshwater organisms	400	400	400		400		10
Benthic invertebrates			400	400-4000		200	
Fish			400	40-400		20	
Deep ocean organisms	1000		400				

4.4. Environmental dose assessment for radiotracer studies

The earliest known attempts to determine the dose to biota from radiotracer studies were conducted for mammals in the late 1990s: a cow in a stream in Norway labelled with ^{51}Cr and ^{3}H and a white dolphin swimming in a ^{198}Au and ^{3}H labelled sewage plume in Hong Kong Harbour. In both examples (treated in detail in the paper titled “Radiotracer Applications: Case studies from four continents”, from Jung et al., included in this publication) dose coefficients and dose limits for reference man were used as none were available for the species potentially exposed.

In the following decade changes in the licensing of radiotracing in Australia led to a variety of attempts to determine dose to biota from exposure to radiotracer sources, in particular ^{192}Ir labelled glass used in sediment transport studies.

At that time and continuing into the present day a number of significant problems are faced in attempting to apply the available non-human biota dose assessment tools and methodologies to radiotracer studies. The major limitations are with the assumptions underlying the dose assessment methodologies and the lack of biological uptake data available for the majority of important radiotracer elements.

The new ICRP [11] document reports dose conversion coefficients for 12 organisms and 70 radionuclides. Many key radionuclides for radiotracer studies are not included in this list (e.g. ^{198}Au, ^{99m}Tc) because they are not commonly found in the environment or have sufficiently short half-lives that they do not persist in the environment. These factors demonstrate the assumptions underlying the newly developed guidelines and tools for environmental dose assessment that are not valid radiotracer applications. The known limitations with using existing tools and guidelines for dose to non-human biota for radiotracer studies are:

Missing input data for radiotracer sources – Even though some of the non-human biota dose assessment tools include the ability to add new isotopes, biota concentration factor (CF) or sediment partition coefficient (K_d) data are not available for many elements of interest for radiotracing because they are not contaminants of concern in the environment. Examples of 'missing' elements include: Au, Br and Hf.

Assumption of steady state exposure – the use of a simple time independent concentration factor assumes that radioisotopes in the biota and adsorbed to sediments are in equilibrium with the radioisotopes in the water which are infinitely replenished. This poses two problems for application to radiotracer studies, firstly that the short time frame of exposure to a moving tracer plume may be insufficient to allow biota or sediment concentrations to come to equilibrium, and secondly, that for the very small mass of radiotracers uptake of the radiotracer by sediments and biota may lead to a decrease in the concentration in the water. Both of these factors may result in the model overestimating dose rates during the study. However, retention of the radioisotope by the organism after the tracer plume has passed may result in ongoing dose to that organism for a period of hours to months depending on the half-life of the radioisotope.

Dose guidelines for chronic not acute exposure – Available guidelines are in units of say $mGy \cdot d^{-1}$ or hr^{-1}, and assume a chronic exposure for the organism which, in the case of radiotracer studies, is commonly not the case. No acute dose guidelines are available for dose to non-human biota.

Assumption of infinite extent and uniform spatial and temporal concentration – in a dispersing and decaying tracer plume the radioisotope concentration varies rapidly both spatially and temporally. In conducting a dose assessment it is difficult to determine what concentration to use – e.g. the maximum immediately following injection, the average over the study duration. It is possible in a Tier 3 probabilistic assessment to use the frequency distribution of the tracer concentration as an input. In addition dose guidelines also assume infinite extent and are aimed at protecting populations, not individuals of a species. Most radiotracer plumes only interact with a very small proportion of any population being assessed.

Assumption of bioavailability – all models assume that the radioisotope is in a dissolved and bio-available form. This is not valid for many tracers which are encapsulated in glass or other forms, and bioavailability is reduced for surface coated sediment tracers. This leads to over estimation of ingestion dose.

No capacity to deal with hot particles – related to the previous point is that existing tools have no capacity to model dose from concentrated particles of radiotracer (e.g. labelled glasses) which have the potential to provide a higher internal dose when ingested than a more bio-available form, but have a lower probability of receiving that dose determined by particle numbers.

As outlined above, the approach given in Fig. 7 is very complex, but in fact not complex enough to adequately model dose from radiotracer releases. The tools available essentially consider continuous release of nuclides from hospitals and nuclear industry activities (i.e. steady state phenomena) and thus are not well suited to tracer applications, which use short half-life nuclides and instantaneous injections (i.e. transient phenomenon).A number of approaches have been taken in order to conduct non-human biota dose assessment for a radiotracer release either by adapting existing tools or doing calculations based on first principles for particular exposure scenarios.

4.5. Practical approaches for environmental dose assessment for radiotracing

4.5.1. Human dose assessment method applied to non-human biota

A first pragmatic dose assessment step follows the procedure and uses the tools and results obtained for humans in Section 3.

Case 1: an organism in the water column placed, as for the public, at the centre of a cylinder of traced water 10 m diameter, 10 m height containing 37 GBq. We don't take into account radioactive decay or dispersion. The maximum external dose rate is obtained for ^{181}Hf: 3.2 μGy/h. This value is 3 times lower than the 10 μGy/h dose rate considered to have no effect on any species considered [24] and used a the lowest screening value in the ERICA methodology. It is important to note that this screening value as been defined for a continuous and constant exposure. The exposure to a tracer test nuclide is transient due to both radioactive decay and dispersion. The external dose calculated in these examples is thus highly over-estimated.

Case 2: an animal at the centre of a disk 10 m diameter equivalent to a diver staying on the sea bed. The external dose is 8 μGy/h again less than the 10 μGy/h screening value.

After that which shows that the potential impact, if any, is very low, we have to consider two particular cases:

Case 3: this case considers the ^{99}Tc (213000 years half-life) which is produced by decay from ^{99m}Tc. This is the only case where a nuclide remains in the environment more or less as a conservative product. 37 GBq of ^{99m}Tc will produce 119 Bq of ^{99}Tc.

We can assume that all this activity will concentrate as a point source (although is not physically possible because the tracer is dispersed into at least million of m^3 of water). Anyway we can calculate the dose rate delivered by a point source of 119 Bq of ^{99}Tc placed at the contact of an animal or inside the animal. The result given by the Microshield modelling

is 2.2 µGy/h. This value is again 5 times lower than the ERICA screening value accepted to have no impact.

Case 4: this case considers an oyster or a mussel which has incorporated 1 grain of 155 µm diameter iridium glass (activity 385 Bq as in Section 3.2 Case 4). The internal dose rate is 0.4 µGy/h and again 25 times lower than the ERICA screening value.

Case 5: an organism has by any mechanism incorporated into its whole body a tracer (Fig. 8). The total activity in the organism is assumed to be 37 kBq. The dose is calculated at the centre of the organism modelled by a cylinder of 10 cm long and 10 cm diameter.

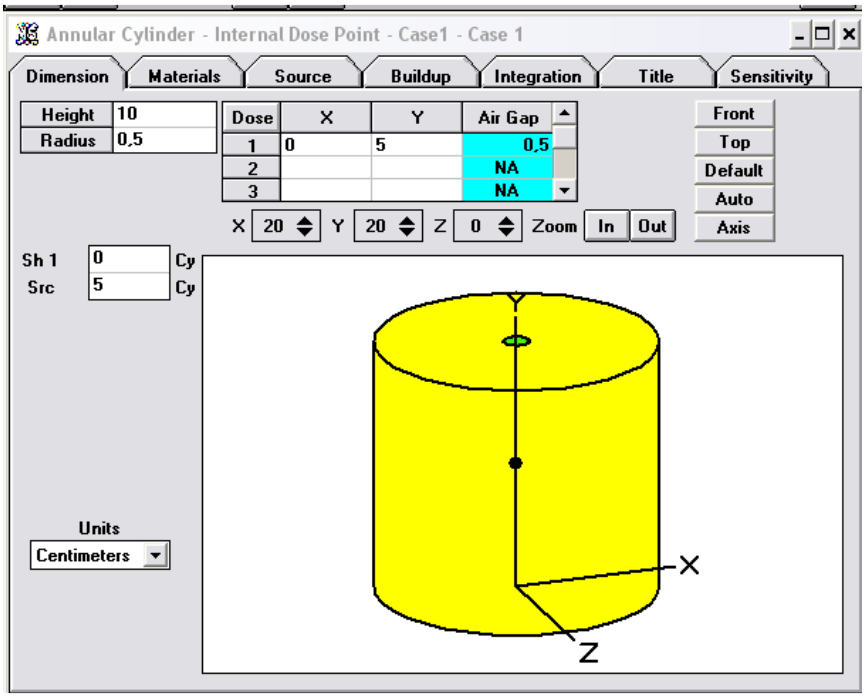


FIG. 8. Schematic for biota dose Case 5.

Typical results are given in Table 5.

TABLE 5. NUCLIDE ACTIVITY AND DOSE RATE CONSIDERING THE RADIOTRACER INCORPORATED IN A WHOLE ORGANISM

Nuclide Activity in the organism 37 kBq	**Dose rate at the centre of the organism µGy/h**
^{99m}Tc	0.48
^{131}I	1.3
^{198}Au	1.4
^{181}Hf	2.0

Again we see that these values are very lower than the ERICA screening value (10 µGy/h).

Obviously this approach provides only a preliminary and pragmatic approach. However, as a first step, these cases show that despite calculation assumptions of a highly conservative nature (e.g. re-concentration of ^{99}Tc as a point source after dispersion in millions cubic meters of water) we can assume that most radiotracer experiments have a very low impact or no impact on the environment.

4.5.2. Environmental dose assessment for Au-198 radiotracer study in Hong Kong Harbour.

The Tolo Effluent Export Scheme study conducted in Hong Kongin 1999, is described in more details in the paper titled "Radiotracer Applications: Case studies from four continents", from Jung et al., included in this publication. During this study 2000 GBq of ^{198}Au was dissolved in aqua regia to produce $AuCl_4$ (tetrachloroaurate) ions. This was injected over a period of 18.5 h into a treated sewage effluent stream.

The maximum gold-198 activity was found to be 4658 Bq/L in the channel and 1748 Bq/L at Station 1. Water depth was 1-1.5 m at Station 1A and 2-2.5m at Station 1. Water body was ~200 m wide and 2 m deep at Station 1 and narrower channel at Station 1A. At Station 1A the water was flowing in a man-made channel as discharged from the sewage treatment plant. This is a positive flow of ~3 m^3/s in one direction. Apart from alga growing on the channel walls the majority of phytoplankton in this zone will have been sourced from the sewage treatment plant. Once the water is discharged into the upper section of the Kai Tak Nullah (Station 1) it starts to dilute. At both of these locations some loss of ^{198}Au bound to settling sediments has occurred and is estimated to be 33% and 50% at Stations 1A and 1 respectively.

Because ^{198}Au at the bottom is believed to be adsorbed to particulates which have settled the bottom values are not used for phytoplankton dose assessment as the gold is not available for uptake. The average suspended solids concentration was 6.6 mg/L, average pH 8.0, and chlorophyll *a* 3.5 µg/L.

Greene et al [25] studied uptake of gold by algal species *Chlorella vulgaris* and found that:

- The interactions between the tetrachloroaurate ion and algae *Chlorella vulgaris* are not pH dependant

- Uptake rate is very fast – e.g. for 0.1mM $AuCl_4$with 5 mg/mL algae 100% bound within 2-3 minutes

- The bound fraction increased with increasing algal density with 100% bound at 1 mg/mL.

In this field experiment the maximum $AuCl_4$ concentration was 3 x 10^{-15}mol/L which is 11 orders of magnitude less than values used by Greene et al [25] and even assuming all the suspended solids in the Kai Tak Nullah are algae then the density is 3 orders of magnitude less. The 95th percentile suspended solids standard for the effluent stream is 30 mg/L. On this basis it is reasonable to assume that all of the available ^{198}Au is adsorbed to algae or other organic matter as soon as the tracer mixes with the effluent stream and that little free $AuCl_4$ is left in the plume when it mixes with seawater in the Kai Tak Nullah (Fig.9).

Dose rate was calculated using the methodology outlined by Copplestone et al [14] (Fig. 7). External doses were lower than US DOE & UNSCEAR dose limit of 400 µGy/hr but higher than FASSET guideline of 100 µGy/hr and no-effect level of 10 µGy/hr [24]. Fig. 10 represented the estimated maximum doses rates from ^{198}Au tracer studying Hong Kong, 1999.

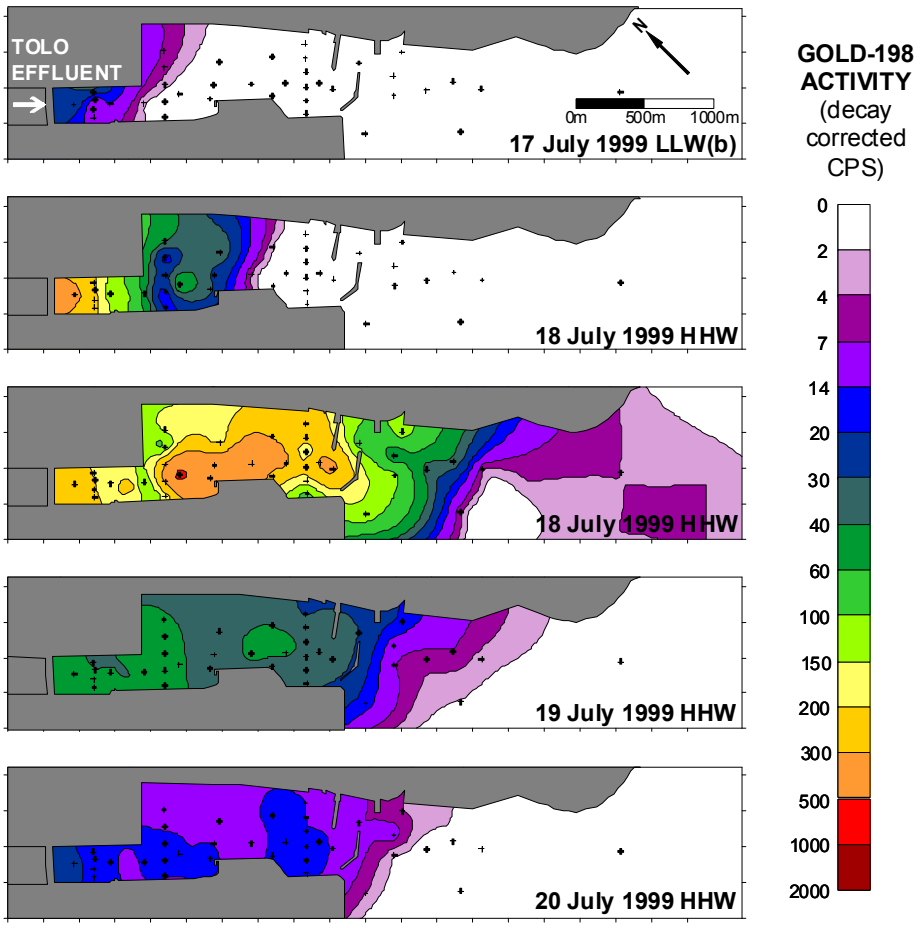


FIG. 9. ^{198}Au distribution in the Kai Tak Nullah during the radiotracer study.

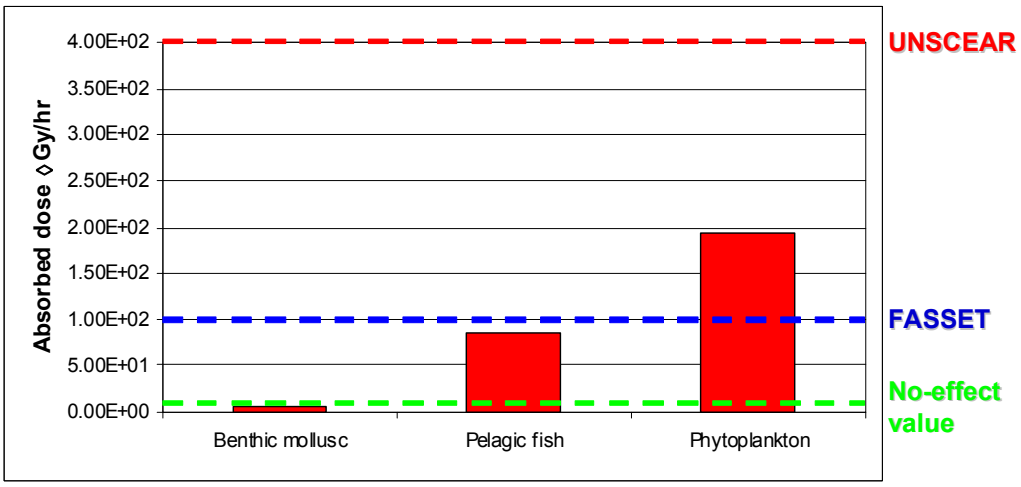


FIG. 10. Estimated maximum dose rates from Au-198 tracer study in Hong Kong, 1999.

4.5.3. Non-human biota dose assessment for Canadian Experimental Lake 224 using ERICA

Taking data from the literature, the ERICA assessment tool was used to conduct a non-human biota dose assessment for the radiotracer studies conducted in Canadian Experimental Lake 224. The studies looked at the distribution of a range of heavy metals and nutrients through the water column, their adsorption onto sediments and uptake into biota over the period of a

year [26-29]. Because of the long term nature of the lakes experiments the steady state assumption underpinning the ERICA methodology is more closely approximated, in contrast to most radiotracer applications which are transient.

A radiotracer injection into the epilimnion of Lake 224 was conducted on 20/7/76 [26]. The tracers were 200-500 mCi [7.4 -18.5 GBq] each of ^{59}Fe, ^{60}Co, ^{65}Zn, ^{75}Se. ^{134}Cs and ^{203}Hg. The tracer was mixed through the epilimnion within 24 hours and the authors state that the activities were only 10% of the IAEA limits at that time.

A tritium study of vertical dispersion was carried out in the same year [29].The depth of the epilimnion on 27/6/76 was found to be approximately 5-6 m, the surface area of the lake is 25.9 ha, maximum depth is 27 m, mean depth is 11.6 m, and volume is 30.1×10^5 m^3. The volume of water in the top 6 metres is 12.96×10^5 m^3 [29].

Assuming that the maximum was injected of each isotope (500 mCi = 18500 MBq) the concentration of each tracer in the epilimnion would be:

$$1.85 \times 10^{10} \text{ Bq}/(12.96 \times 10^5 \text{ m3} \times 1000 \text{ L/m}^3) = 14.3 \text{ Bq/L}$$

Assumed to be mixed over the entire water column it would be 6.15 Bq/L.

In order not to overestimate the adsorption of the radiotracers to the sediment instead of using a K_d approach within ERICA we have made an assumption of the whole of the tracer amount being adsorbed to the sediments. According to Hesslein et al [28] the various isotopes have different affinities to the sediments but the majority of adsorption was to bottom sediments from 1-3 cm deep within the epilimnion zone down to about 8-10 m depth. From Quay et al [29] the bottom area of the lake from 0-9 m is calculated to be 11.29×10^4 m^2. Assuming an average depth of 2 cm and a sediment bulk density of 1 g/cm^3 the maximum activity on the sediments would be 8.19×10^3 Bq/kg.

Hesslein et al [27] estimated the proportion of each tracer to have been incorporated into bottom sediments over the course of a year to be as represented in Table 6.

TABLE 6. PROPORTION OF TRACER INCORPORATED INTO BOTTOM SEDIMENTS OF THE COURSE OF A YEAR

	Fe-59	**Co-60**	**Zn-65**	**Se-75**	**Cs-134**	**Hg-203**
Adsorption %	98%	80%	80%	50%	1%	80%
Sediment activity	8.03E+03 (Bq/kg)	6.55E+03 (Bq/kg)	6.55E+03 (Bq/kg)	4.10E+03 (Bq/kg)	8.19E+01 (Bq/kg)	6.55E+03 (Bq/kg)

For this assessment we will take all other default parameters and values from the ERICA Assessment tool. The estimated dose rates are given in Fig. 11. For all organisms the dose rates exceed the ERICA screening value, however, they are all below the derived consideration reference levels proposed by the ICRP [11]. This demonstrates that even a longer term radiotracer study of a closed ecological system can be conducted without radiation dose impacting on the ecosystem.

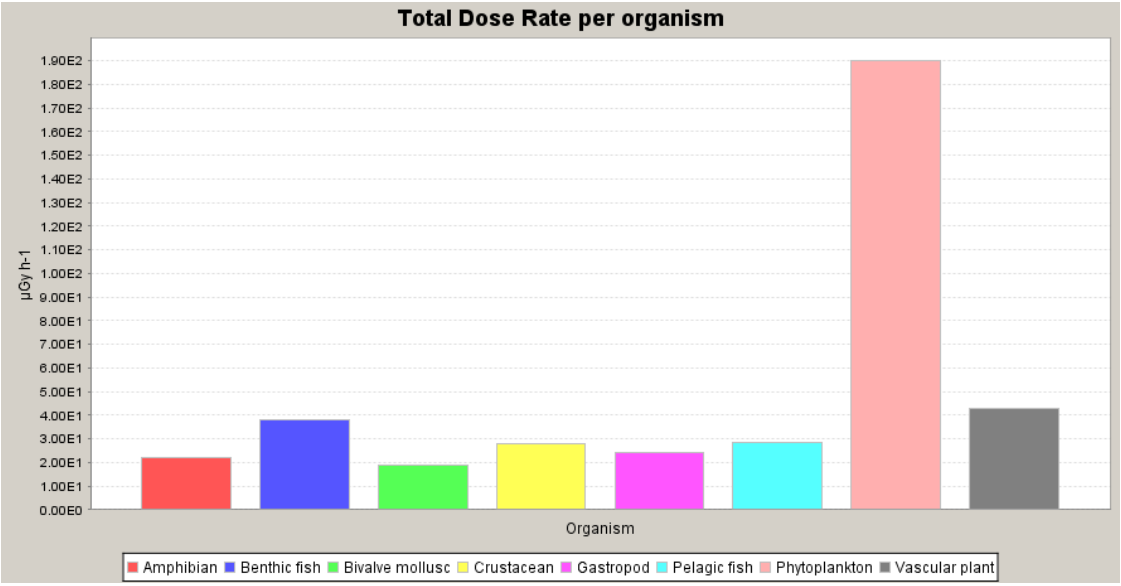


FIG. 11. Dose rate to aquatic organisms from Experimental Lake 224 heavy metal radiotracer using ERICA Assessment tool.

4.5.4. Dynamic versus steady state modelling for ^{131}I tracer study

Where pulse or instantaneous radiotracer injections are made into flowing waters the tracer rapidly disperses. In such a case no steady state concentration is ever achieved. To demonstrate the impact of this we use as a case study a radiotracer release of 600 mCi of ^{131}I conducted over 30 minutes into the Bay of Chimbote in Peru [30]. The activity was monitored as it dispersed over a period of up to four hours. The peak activity of the plume over time was used to assess the dose rate to marine organisms from this tracer study.

TABLE 7. STEADY STATE AND DYNAMIC MODEL INPUTS AND DOSE RATES

Input and model scenario	Input activity (Bq/L)	Macrophyte dose rate (µGy/h)	Zooplankton dose rate (µGy/h)
ERICA: Study maximum	1.06×10^5	52200	31200
ERICA: Study average	1.11×10^4	5430	3270
ERICA: Daily average	1.69×10^3	832	497
Dynamic model	Time varying	1464 (max)	630 (max)

When using the currently available steady state dose assessment tools it is difficult to determine which input activity to use, and the result is commonly that the dose is overestimated. With the assistance of Richard Wilson and Jordi I Vives a new dynamic dose assessment model [31] was adapted to run on an hourly time step and applied to a case study from the Bay of Chimbote in Peru [30]. This model allows the concentration of the surrounding tracer plume to be reducing over time and models the uptake and depuration of the tracer by the modelled organism over time. The results from this dynamic model were compared with outputs from an ERICA Tier 2 model using a range of input tracer activities (Table 7). The steady state and dynamic results are compared in Fig. 12. It can be clearly seen that using either the maximum tracer concentration or even averaging it over the short study

duration leads to a dramatic overestimate of the dose rate to the exposed organisms. However, the dynamic results indicate that the effect of radioisotope continues beyond the duration of the study as the exposed organisms retain the isotope. The short duration of the peak concentration is clearly insufficient to allow full uptake of the radioisotope by the organism before the surrounding concentration has diluted.

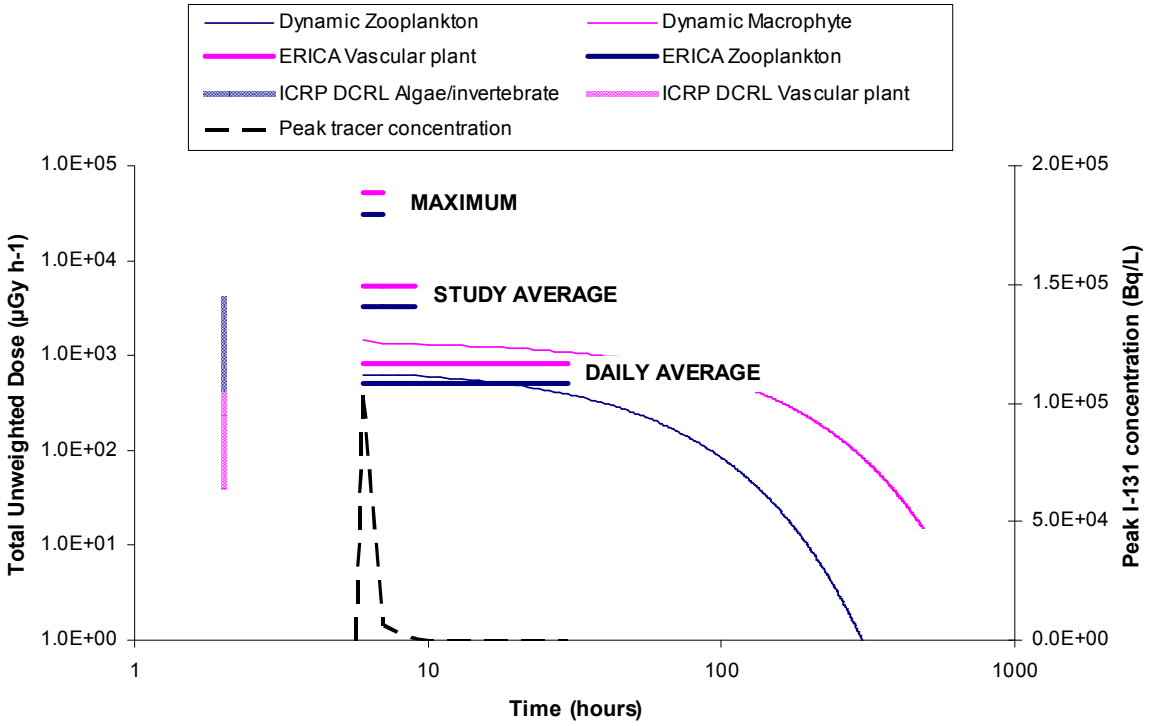


FIG. 12. Dynamic and steady state model of ^{131}I dose to non-human biota from the Bay of Chimbote study, Peru.

REFERENCES

[1] INTERNATIONAL ATOMIC ENERGY AGENCY. Fundamental Safety Principles. Safety Standards Series No. SF-1. IAEA, Vienna (2006) 21 pp.

[2] INTERNATIONAL ATOMIC ENERGY AGENCY, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards. IAEA Safety Standards Series No GSR Part 3 (Interim) (2011).

[3] INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION, Dose Coefficients for Intakes of Radionuclides by Workers. ICRP Publication 68. Ann. ICRP **24** 4 (1994).

[4] INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION General Principles for the Radiation Protection of Workers. ICRP Publication 75. Ann. ICRP **27** 1 (1997).

[5] NAVADA, S.V., Safety aspects of the use of artificial radioactive tracers in hydrology. In In: Use of Artificial Tracers in Hydrology, IAEA-TECDOC-601, 179-200, Vienna (1990).

[6] INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION, Recommendations of the International Commission on Radiological Protection. ICRP Publication 60 (Users Edition) (1990).

[7] INTERNATIONAL ATOMIC ENERGY AGENCY, Regulations for the Safe Transport of Radioactive Material – 2012 Edition. IAEA Safety Standards Series SSR-6 (2012).

[8] DELACROIX D, GUERRE J-P, LEBLANC P., Guide Pratique Radionucléides et Radioprotection. Radioprotection, **39** (2004). CEA et EDP Sciences.

[9] WHICKER, P.W., SCHULTZ, V., Radioecology: Nuclear Energy and the Environment, Vol. I. CRC Press, Boca Raton, FL (1982).

[10] INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION, A framework for assessing the impact of ionising radiation on non-human species. ICRP Publication 91 (2003).

[11] INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION Environmental Protection: the Concept and Use of Reference Animals and Plants, ICRP Publication 108, Ann ICRP, **38** 4-6 (2008) 242 pp.

[12] BROWN, J.E., THØRRING, H. & HOSSEINI, A. (Eds), The ‘‘EPIC’’ impact assessment framework. Towards the protection of the Arctic environment from the effects of ionising radiation. Deliverable Report for the EC Project EPIC. EC Inco-Copernicus project ICA2-CT-2000-10032. Statens Stralevern, Oslo (2003).

[13] US DEPARTMENT OF ENERGY, A Graded Approach for Evaluating Radiation Doses to Aquatic and Terrestrial Biota. Technical Standard DOE-STD-1153-2002 (2002). http://www.eh.doe.gov/techstds/standard/std1153/1153.htm

[14] COPPLESTONE, D., et al., Impact Assessment of Ionising Radiation on Wildlife. R&D Publication 128, Environment Agency, Bristol, UK (2001) 222 pp.

[15] BEAUGELIN-SEILLER, K, et al., E.D.E.N.: A tool for the estimation of dose coefficients for non-human biota, Radioprotection **40** (2005) S921-S926.

[16] INTERNATIONAL ATOMIC ENERGY AGENCY, Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments IAEA Technical Report Series TRS 472, IAEA, Vienna (2010).

[17] INTERNATIONAL ATOMIC ENERGY AGENCY, Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment, Technical Reports Series No. 422, IAEA, Vienna (2004).

[18] INTERNATIONAL ATOMIC ENERGY AGENCY, Effects of ionising radiation on plants and animals at levels implied by current radiation protection standards, IAEA Vienna, Technical Report Series No 332 (1992).

[19] INTERNATIONAL ATOMIC ENERGY AGENCY, Principles for the Exemption of Radiation Sources and Practices from Radiological Control, IAEA Report Safety Series No. 89, IAEA, Vienna (1988).
[20] NUCLEAR COMMISION ON RADIATION PROTECTION, Report No. 109 - Effects of Ionizing Radiation on Aquatic Organisms (1991).
[21] UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION, Effects of radiation on the environment. Annex in: *Sources and Effects of Ionizing Radiation*. United Nations Scientific Committee on the Effects of Atomic Radiation 1996. Report to the General Assembly, with Annexes. pp 1-86. United Nations, New York (1996).
[22] US DEPARTMENT OF ENERGY, RESRAD-BIOTA: A Tool for Implementing a Graded Approach to Biota Dose Evaluation. User's Guide, Version 1. DOE Report No. DOE/EH-0676; ISCORS Technical Report 2004-02 (2004).
[23] BIRD, G.A. et al., Ecological risk assessment approach for the regulatory assessment of the effects of radionuclides released from nuclear facilities. In: Protection of the Environment from Ionising Radiation The Development and Application of a System of Radiation Protection for the Environment, IAEA-CSP-17, IAEA (2003) 241–247.
[24] GARNIER-LAPLACE, J., et al., First derivation of predicted no effect values for freshwater and terrestrial ecosystems exposed to radioactive substances, Environ. Sci. Technol.**40** (2006) 6498-6505.
[25] GREENE, B., et al., Interaction of Gold (I) and Gold (III) Complexes with Algal Biomass. Environ. Sci. Technol. **20** (1986) 627-632.
[26] KLAVERKAMP, J.F., et al., Fates of metal radiotracers added to a whole lake: accumulation in slimy sculpin (Cottuscognatus) and white sucker (Catstomuscommersoni), Sci Tot Env, **28** (1993) 119-128.
[27] HESSLEIN, R.H., BROECKER, W.S., SCHINDLER, D.W., Fates of metal radiotracers added to a whole lake: sediment-water interactions. Can, J. Fish. Aquat. Sci. **37** (1980a) 378-386.
[28] HESSLEIN, R.H., BROECKER, W.S., QUAY, P.D., Whole-lake radiocarbon experiment in an oligotrophic lake at the Experimental Lakes Area, north-western Ontario, Can. J. Fish. Aquat. Sci. **37** (1980b) 454-463.
[29] QUAY, P.D., et al., Vertical diffusion rates determined by tritium tracer experiments in the thermocline and hypolimnion of two lakes, Limnology and Oceanography, **25** 2 (1980) 201-218.
[30] SEBASTIÁN, C., MAGHELLA, G., MAMANI, E. Estudios de Dispersión de Contaminantes y Declinación Bacteriana en la Bahia del Mar de Chimbote - Perú. Instituo Peruano de Energía Nuclear, Report IPEN-TR--075, pp.20 (2000).
[31] VIVES I BATLLE J., WILSON, R.C., WATTS, S.J., JONES, S.R., MCDONALD, P., VIVES-LYNCH, S. Dynamic model for the assessment of radiological exposure to marine biota. Journal of Environmental Radioactivity **99** 11 (2008) 1711-1730.

TRACER DATA INTERPRETATION AND MODELLING

A. WÖRMAN

Royal Institute of Technology, KTH, Sweden

Abstract

The translation of raw tracer data into parameters that represent the physico-chemical behaviour of the studied component is a common approach to understand and generalize results from a tracer study. This paper covers aspects of tracer data interpretation including tracer mass balance, travel time distribution, transport parameters, decoupling of tracer and water flows, modelling flow and non-conservative tracers and how to determine model parameters from tracer data.

1. RADIOACTIVITY BALANCES AND TRAVEL TIME DISTRIBUTIONS

Tracer data can be interpreted in various ways depending on purpose of the investigation, but in flowing surface water mass balance is a key variable. In a multidimensional water flow, such as in an estuary, a lake or a wetland, it can be difficult to obtain representative estimates of the radioactive inventories. However, in this paper we will assume that we can identify two control sections through which all water is flowing and an intermediate volume in which the processes subject to the tracer study occur. The tracer injection is performed some appropriate (small) distance outside the up-stream control section and measurements of activity in the water are performed in the control sections.

In a stream or river the control sections can easily be identified. The activity A passing the control section can be written on the form of Eq. (1)

$$A = \int_0^\infty C(t)Q(t)dt \approx \sum_{i=1}^{N} C_i Q_i \Delta t_i \tag{1}$$

in which $Q(t)$ is the water discharge variation [m^3/s], $C(t)$ is concentration variation with time [Bq/L], t is time [s], Δt is a time interval selected for numerical treatment of data [s], and i is an index of the time discretization. Here it will be assumed that all concentrations as well as activities are compensated for decay, as discussed in the paper titled “Radiotracer methodology“, from Brisset et al., include in this publication. Thus, the activity corresponds to the initial activity A_0. The activity balance can be written as:

$$A_{up} - A_{down} + A_{acc} = 0 \tag{2}$$

where subscripts *up* = upstream control section, *down* = downstream control section and *acc* = accumulation within water volume.

Since the integrated flux of activity $F(t) = C(t)Q(t)$, the right-hand side of (1) is the zeroth moment of the flux, $n_{F,0}$. The flux weighted mean travel time of tracer is obtained by use of the first moment of the activity flux:

$$n_{F,1} = \int_0^\infty tC(t)Q(t)dt \tag{3}$$

Thus, the mean travel time becomes:

$$\mu_t = \left(\frac{n_{F,1}}{n_{F,0}}\right)_{down} - \left(\frac{n_{F,1}}{n_{F,0}}\right)_{up} \tag{4}$$

In case the up-stream location is considered to be the site of injection and the injection is sudden, the second term on the right-hand side of (4) can be neglected. Further, if the discharge is constant we can obtain the mean residence time directly from the concentration breakthrough curve $\mu_t = n_{C,1} / n_{C,0}$.

Fig. 1 exemplifies a breakthrough curve obtained at the outlet of a wetland due to injection of tritiated water at the inlet to the wetland. The grey bar corresponds to the mean residence time calculated from Eq. (4). Since, the breakthrough curve is considerably spread out the water has a distribution of travel times through the wetland (Fig.1). Especially, the normalized concentration breakthrough curve $C/n_{C,0}$ can be interpreted as a travel time probability density function (pdf) from the site of (sudden) injection. This normalization is discussed and utilized in the paper titled "Radiotracer methodology", from Brisset et al., included in this publication. Such a travel time pdf is an attractable measure if the tracer recovery is incomplete and, hence, the estimation of the mean travel time is unreliable. There can be significant problems with insufficient tracer recovery for sorbing and reactive tracers.

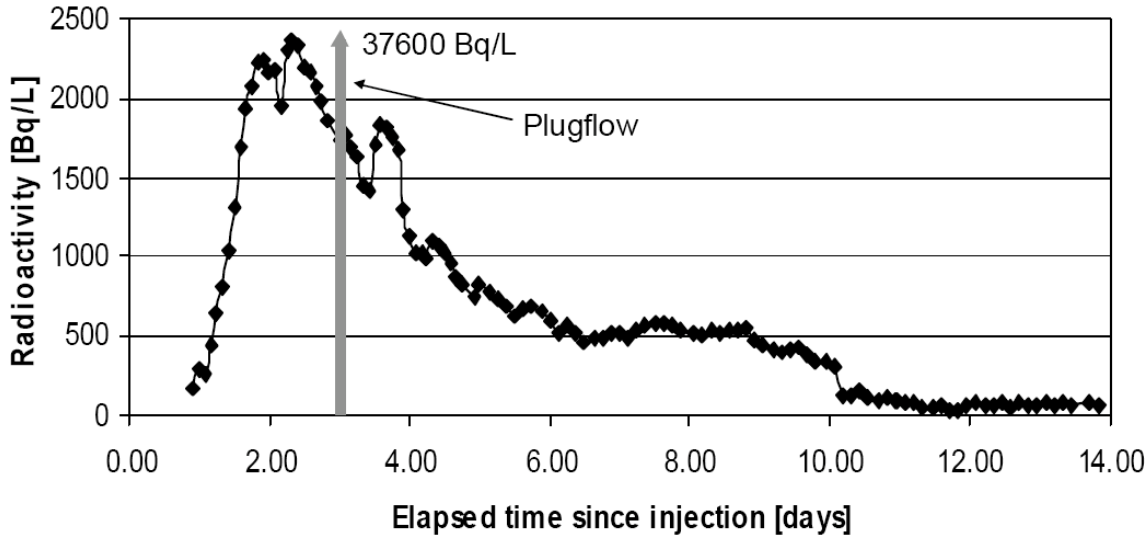


FIG. 1. Breakthrough curve at the outlet of a treatment wetland (blue curve and dots. The light-blue vertical bar indicates the time interval corresponding to the mean travel time according to Eq. (4). The width of the bar equals that of the injection and, thus, is the concentration response if the wetland had a plug flow (the height is decreased for visualization reasons).

2. TRANSPORT PARAMETERS AND FUNDAMENTAL MODEL PRINCIPLES

Modelling of flow and solute transport in the environment is based on conservation statements for extensive properties, such as mass, heat and momentum, and constitutive relationships for fluxes of the extensive properties. In addition, the problem formulation may recognize equations of state expressing state variables, like density or viscosity, as function of the extensive properties.

The conservation statements can be formulated for control volumes representing suitable (arbitrary) zone of the environment, such as a lake, or for specific species, such as dissolved solute or solute taken up in biota. Fluxes occur between the control volumes and are represented by the flux equations. These models are often referred to as compartmental or box models. An alternative statement of conservation recognizes a continuous variation of the extensive properties in space, but to formulate these separately for each of the species as demonstrated in the paper titled "Radiotracer Applications: Case studies from four

continents", from Jung et al., included in this publication. These continuous conservation equations also contain constitutive flux relationships for the spatial redistribution (transport) of the extensive properties. Because the formulation is continuous in space it is said to be differential form of conservation model.

Both the compartmental and differential model and a combined approach open the possibility of relating the observed tracer response behaviour, such as the breakthrough curve, to the recognised transport parameters. This generally involves some type of optimization between model and data, in which the considered optimization criteria can have a major role.

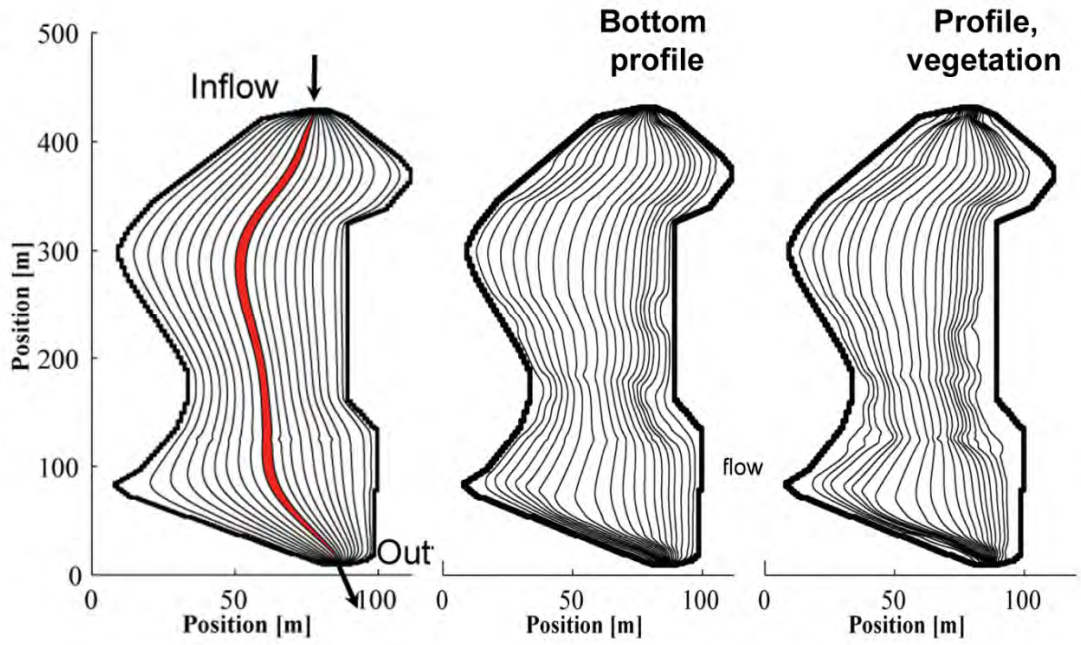


FIG. 2. Water stream lines through Ekeby wetland according to flow models with various assumptions of bottom profile and distribution of vegetation. The left-hand side figure is based on the assumption of a flat bed and no vegetation. By adding increasing complexity in terms of measured bottom profile (mid figure) and vegetation (right-hand side figure) we find increasing disturbance of the stream-lines and a wider residence time distribution.

3. DECOUPLING OF WATER FLOW AND TRACER TRANSPORT

The tracer transformations in surface water can be studied using a model framework that decouples hydraulics and chemical transformation of solutes in two separate functions that can be evaluated independently using tritiated water or another inert tracer and reactive tracers [1-3]. The decoupling can be performed under the assumption of no mixing between flow paths. Fig. 2 shows three modelling results for the flow in a wetland with free water surface. In the left-hand side of Fig. 2 a flow tube is indicated in red. In a steady flow, each of those flow tubes, contained between stream-lines, can be ascribed a residence time τ for water between the inlet to the outlet. If $C(t, \tau)$ is the variation of the concentration with time t for a specific transport pathway (denoted by τ) one can write the effluent concentration as:

$$C_{out}(t) = \sum_{i=1}^{N} C(t,\tau_i) F(\tau_i) = \int_0^{\infty} C(t,\tau) f(\tau) d\tau \tag{5}$$

in which $F(\tau_i)$ is the probability for flow path with residence time τ_i and $f(\tau)$ is the probability density function (PDF) of the residence time. The method can be regarded as an evaluation of the average response at the exit concentrations of several pathways of different residence time τ.

The advantage of Eq. (5) is that it allows an evaluation of the reactive tracer response decoupled from water flow. In the paper titled "Radiotracer Applications: Case studies from four continents", from Jung et al., included in this publication, a simultaneous tracer test using tritiated water and ^{32}P marked phosphate in a wetland is described. By the measured water residence times in terms of $f(\tau)$ it is possible to evaluate the phosphorus concentration at effluent C_{eff} the as function of inlet concentration C_{in} and the residence time $C_{eff} = C_{in} \exp(-k\tau)$ as demonstrated in Fig.2. Another possibility is to use flow modelling in several dimensions to represent the water residence times.

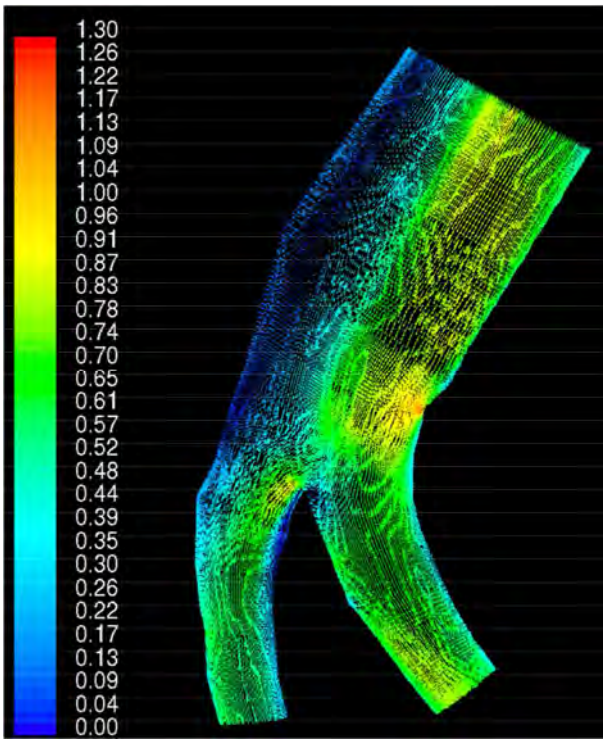


FIG. 3. Two-dimensional flow field in River Kalrälven, Sweden, according to numerical simulation [Results after: Bijan Dargahi].

4. MODELS FOR FREE SURFACE FLOWS

The mathematical statement of the water flow with free surface is based on the Navier-Stokes equations and thermodynamic equations for accounting of turbulent momentum exchange [4]. Generally, we can accept various kinds of simplifications, such as depth or cross-sectional averaging for river flows [5] or neglect inertia terms of the momentum equations in slowly flowing wetlands [6]. The two-dimensional flow fields illustrated Fig.3 are based on depth averaging, steady-state and negligible inertia terms.

An important problem for producing plausible simulation flow fields involves the energy loss, a problem that has different character in different applications and model approaches (model simplifications). In three-dimensional the time averaged momentum equations give rise to the so called 'closure problem' due to turbulent diffusion, whereas in cross-sectional averaged

flows all energy losses are accounted for towards boundaries and represented in terms of constitutive equations, like the Darcy-Weisbach equation.

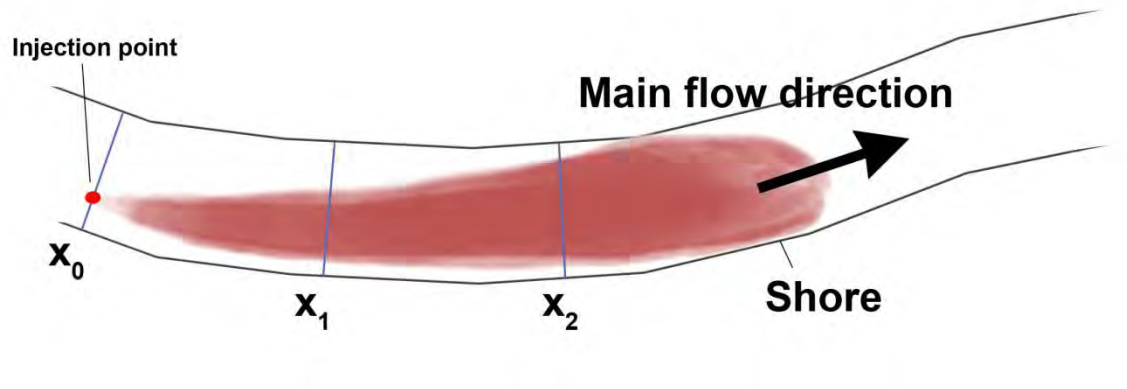


FIG. 4. Sketch of an instantaneous 'freeze' of tracer injected in a water mass that spreads both along the main flow direction and transversally.

5. TRANSPORT AND REACTION MODELS

5.1. Model dimensionality and longitudinal vs. transversal dispersion

When a tracer is injected into a water mass, such as a river, the tracer starts to spread both along the flow and transversal to the flow direction. As long as there is a significant gradient in concentration across the river, the tracer data collected at those sections (e.g. x_1 and x_2 in Fig. 4) can be used to evaluate the transverse dispersion coefficient. One has to consider a two-dimensional formulation of the tracer transport in order to separate out the transversal and longitudinal mixing. Correspondingly, after full mixing is achieved, the tracer data can be evaluated based on a one-dimensional formulation. According to Rutherford [7] complete mixing of a stream or river prevails for a transport distance, X, that satisfies the condition $X/h >> 5\ (U\ h)/D_T$, in which D_T = transversal dispersion coefficient of the main stream [m^2/s], h = hydraulic mean depth of the stream (m) [7]. For shorter distances, we can apply the following statement for conservation of solute mass:

$$\frac{\partial hc}{\partial t} + \frac{\partial}{\partial x}\left[uhc - D_L h \frac{\partial c}{\partial x}\right] + \frac{\partial}{\partial y}\left[vhc - D_T h \frac{\partial c}{\partial y}\right] = 0 \qquad (6)$$

in which x and y are longitudinal and transversal coordinates, respectively, u is longitudinal velocity (m/s), v is transversal velocity (m/s) and D_L is longitudinal dispersion coefficient (m^2/s).

5.2. Statement of solute mass conservation

In reactive transport models we have to consider the various phases in which the solute may exist. Those phases can represent either a chemical compound or an adsorbed state that is discriminated from the freely dissolved state of the solute substance. In addition, in a one- or two-dimensional averaged form of mathematical statement, there can be a need to account for various mobility states in the cross-section, such as dissolved solute in the flowing water column in a river and dissolved solute accumulated in the hyporheic zone of the same river). The differentiation of mass phase species and mobility state should be adequate for analyzing

transport and retention of the tracer substance in use. Mass phase species and mobility states are represented by the red compartments (rectangles) in Fig. 5 and the green arrows are the possible transfer rates between compartments. Not all transfer rates may be considered 'active' in a mathematical statement, but are included here for completeness. Consequently, the evaluation of a tracer test can focus on either establishing the activity inventories within compartments or transfer rates between compartments or both.

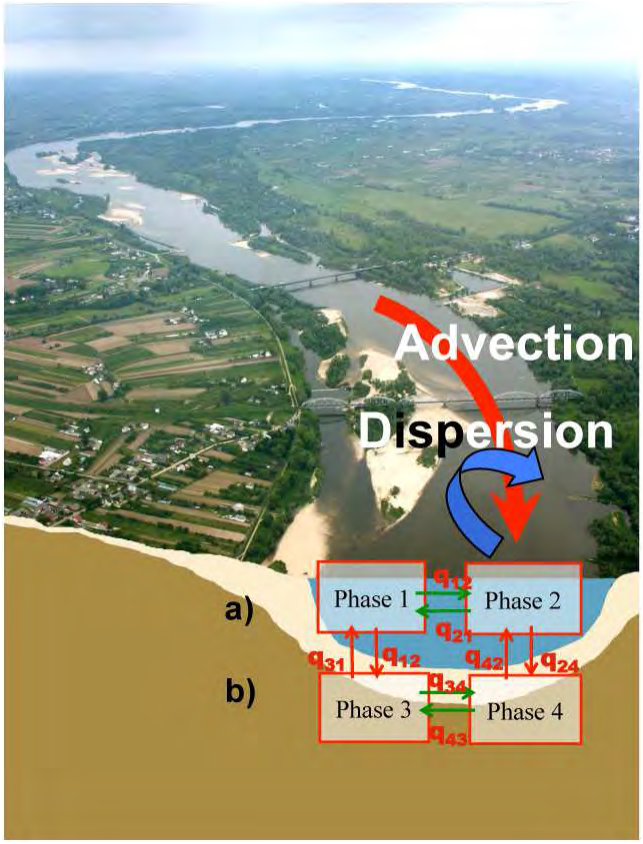


FIG. 5. Schematic of transport and reaction mechanisms (arrows) and mass phase species (boxes) of a radioactive tracer. In this example, phase 1 and phase 2 are dissolved and adsorbed phase concentrations in the flowing water and phase 3 and phase 4 are the dissolved and adsorbed phase concentrations in hyporheic storage zones. The storage mechanism strongly affects the mobility of the tracer and the exchange intensity (rate) in adsorbed/particulate or dissolved state can be markedly different. The picture shows Vistula River outside Warsaw, Poland.

Each of the solute mass phase species and mobility states can, in principle, undergo advection and dispersion/diffusion. In a one dimensional form the statement of solute mass conservation for the four compartments included in Fig. 5 becomes:

$$\frac{\partial A_a c_1}{\partial t} + \frac{\partial}{\partial x}\left[u_a A_a c_1 - D_{L,a} A_a \frac{\partial c_1}{\partial x}\right] = \left[-q_{12} + q_{21} - q_{13} + q_{31}\right] \tag{7}$$

$$\frac{\partial A_a c_2}{\partial t} + \frac{\partial}{\partial x}\left[u_a A_a c_2 - D_{L,a} A_a \frac{\partial c_2}{\partial x}\right] = \left[q_{12} - q_{21} - q_{24} + q_{42}\right] \tag{8}$$

$$\frac{\partial A_b c_3}{\partial t} + \frac{\partial}{\partial x}\left[u_b A_b c_3 - D_{L,b} A_b \frac{\partial c_3}{\partial x}\right] = \left[q_{13} - q_{31} - q_{34} + q_{43}\right] \tag{9}$$

$$\frac{\partial A_b c_4}{\partial t} + \frac{\partial}{\partial x}\left[u_b A_b c_4 - D_{L,b} A_b \frac{\partial c_4}{\partial x} \right] = [q_{34} - q_{43} - q_{42} + q_{24}] \tag{10}$$

in which subscript *a* refers to a mobility state in the flowing surface water, subscript *b* refers to a mobility state in the hyporheic zone, A is cross-sectional area (m^2), c is concentration [Bq/m^3] q denotes the mass transfer rate [$Bq/(m^3\ s)$], D is the longitudinal dispersion coefficient (m^2/s) and numbering denotes the various compartments. The subscripts of the mass transfer rates *i, j* implies flux 'from' compartment *i* 'to' compartment *j*.

5.3. Transport and storage (retention) of inert solute using first-order approximations

A specific application of the mathematical statement described in the former section comprises inert solute transport and uptake in the hyporheic zone. For such conditions, we only have to consider one phase in the flowing water and one phase in the hyporheic zone (Fig. 6). Mass transfer rates, like q_{12}, q_{21}, q_{42}, etc., are zero and it is assumed that the advection velocity in the hyporheic zone is zero. Introducing these simplifications in Eqs. (7) – (10) yields:

$$\frac{\partial c_1}{\partial t} + u_a \frac{\partial c_1}{\partial x} - \frac{1}{A_a}\frac{\partial}{\partial x}\left[D_{L,a} A_a \frac{\partial c_1}{\partial x} \right] = \frac{1}{A_a}[-q_{13} + q_{31}] = \alpha[c_3 - c_1] \tag{11}$$

$$\frac{\partial c_3}{\partial t} = \frac{1}{A_b}(q_{13} - q_{31}) = \alpha \frac{A_a}{A_b}[c_3 - c_1] \tag{12}$$

in which α is the first-order rate coefficient defined by the right-most-hand sides of Eqs. (11) and (12) to represent the considered fluxes between the flowing water and the hyporheic zone [8, 9]. These equations can be replaced with other types of formulations using one dimensional (transversal) uptake in the hyporheic zone based on either diffusion [10], advection [11] or both [12]. Essentially, these different statements associate with the same temporal moment equations [12], which means that Eqs. (11) and (12) demonstrate the general principle of formulation.

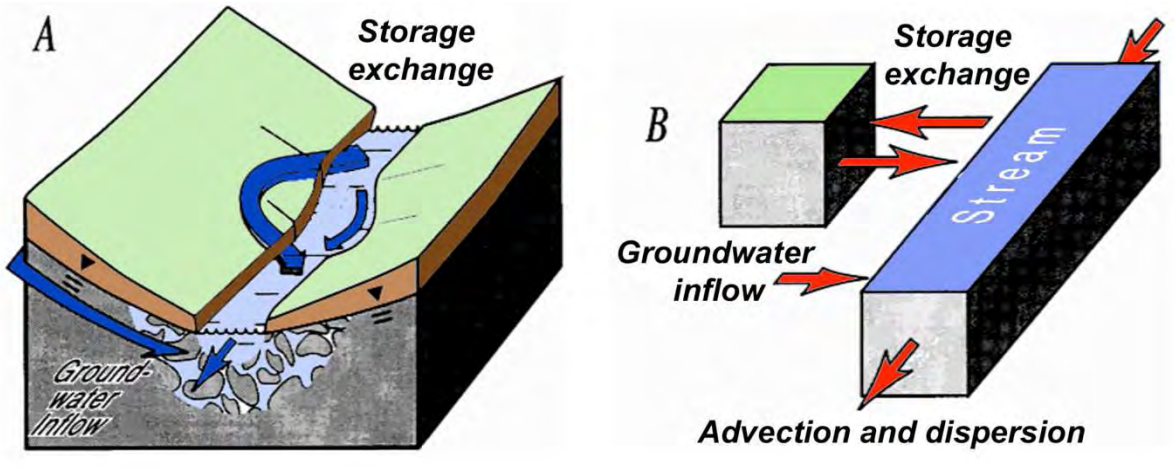


FIG. 6. .Schematic of solute transport in streams (After Harvey and Wagner [9]).

6. EVALUATING MODEL PARAMETERS FROM THE SHAPE OF TRACER BREAKTHROUGH CURVES

6.1. Selection of criteria for parameter evaluation from breakthrough curves

There are a number of ways to utilise tracer data to evaluate transport parameters defined by the physical model in which they were introduced. All are based on fitting model characteristics to data in terms of either temporal moments of the breakthrough curve (concentration vs. time), the entire breakthrough curve or some other sensible measure. The choice of criteria is important for the accuracy of the evaluated parameter regardless of its phenomenological nature (chemical, biological or physical). Fig. 7 demonstrates that specific parameters have higher sensitivity to certain parts of the model breakthrough curve, indicating that accurate evaluations may have to focus on certain parts of a breakthrough curve. For instance, to determine the exchange rate coefficient α it is necessary to consider primarily the slope of the tail of the breakthrough curve (Fig. 7). The exchange rate coefficient affects also the general shape (roundness) of the curve, but can easily be confused with other mixing parameters, like the dispersion coefficient, in other parts of the breakthrough.

The specific sensitivity of the model breakthrough curve to certain parameters is especially important to recognize in a fit-by-eye procedure. However, in an automated procedure the weighting of different parts of the breakthrough has to be provided in a synthetic way inherent in the object function subject to optimization. One example of different weighting can be obtained by log-transforming the modelled and observed breakthrough, whereby a higher weight is given to the lower values, such as the tail [13].

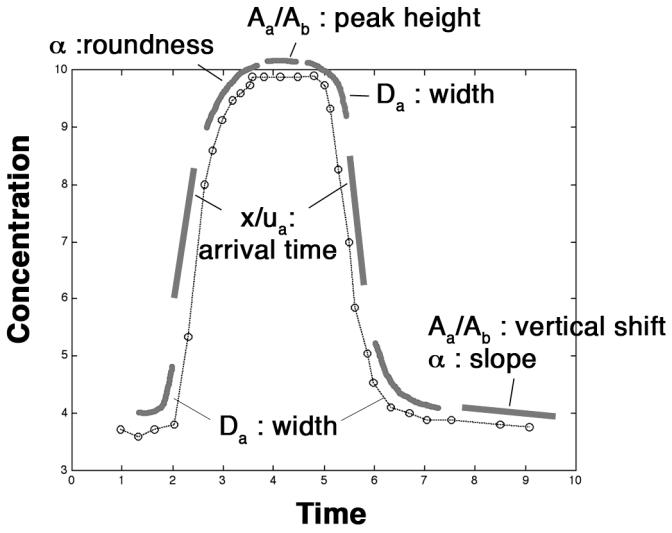


FIG. 7. Breakthrough curve from solute injection in a small mountain stream with marks for parts most sensitive to transport parameters included in the model (11) – (12) [After Wörman and Przemyslaw, [13]].

6.2. Optimization using numerical or exact solutions

Model parameters can be determined from tracer breakthrough curves by optimization methods. In a straightforward approach we search for the parameters contained in the vector θ so that the 'optimized' set of parameters minimizes the sum of squared deviations between model and observation $(\mathbf{O} - \mathbf{M}(\theta))^T (\mathbf{O} - \mathbf{M}(\theta))$, where $\mathbf{O}$ is a vector containing observed concentration values and $\mathbf{M}$ is the corresponding predicted values. These vectors of observed

and predicted values can be assembled for different times and spatial locations and can also be transformed in different manners. The exact approach will give different results in terms of the optimized set of parameters θ.

A common approach is to derive numerical to the model statements, i.e. (7) – (10) or (11) – (12), and to use these as a basis for the optimization. There are also several exact solutions to the advective-diffusive equation with reactions available in the literature, specifically in Crank [14] and Carslaw and Jaeger [15]. The latter reference book is concerned with heat transport, but since this is an analogous problem to solute transport, most solutions are directly convertible. The intention of this report is not to describe all relevant solutions applicable to tracer tests, but rather to point at the fact that exact solutions are available for certain problems and under certain assumptions. Use of such solutions provides a quick means to optimize the model fit and gives an overview of the importance of different processes to the transport of radionuclides.

De Smedt [16] derived a solution to (11) – (12) for the case of an initial concentration spike defined as $c(x,t=0) = A/A_a\delta(t)$; A being the activity, A_a the cross sectional area of the stream and δ is the Dirac delta function. The solution is on an integral form that requires some numerical operations that the author implemented in a Matlab® environment. If the first-order reaction is replaced by a Fickian diffusion term that represents the transient storage (i.e. Eq. (12) is replaced), Wörman [17] derived an exact solution for the case when in-stream dispersion can be neglected in comparison to the transient storage. A similar solution was derived by Maloszewski and Zuber [18] for an analogous formulation of transport of radionuclides in rock fractures and with account to longitudinal dispersion. Both the latter solutions were derived for a defined boundary condition $c(x=0,t) = A/A_a\delta(x)$. If the transient storage term can be neglected the solution can be derived for the same boundary condition on the form of [15]:

$$c(x,t) = \frac{A/A_a}{2\sqrt{\pi D_L t}} \exp\left[-\frac{(x-ut)^2}{4D_L t} \right] \tag{13}$$

This solution implies that the activity distribution is Gaussian in a fixed system (if $u = 0$), but also floats along the river with the advection velocity u.

6.3. Matching of temporal moments in one dimension along flow direction

A number of papers have made use of the temporal moment method to characterize the solution [12, 19-21]. The basic idea is to describe quantities like the mean transport time μ_t and the variance σ_t^2 (cf. Fig. 8) or Skewness S_t in terms of the transport parameters. Hence, the parameters can be determined by fitting the model prediction of the temporal moments versus the observed moments of the breakthrough. Following Schmid [22] we can express the temporal moments of (11) – (12) for spatially constant parameters of the form of

$$\mu_t = \frac{x}{u}(1+\varepsilon) + \mu_0[t] \tag{14}$$

$$\sigma_t^2 = 2\frac{x}{u}\left[\varepsilon T + \frac{D_{L,a}}{u^2}(1+\varepsilon)^2\right] + \sigma_0^2[t] \tag{15}$$

$$S_t = 6\frac{x}{u}\left[\varepsilon T^2 + 2\frac{D_{L,a}}{u^2}\varepsilon T(1+\varepsilon) + 2\frac{D_{L,a}^2}{u^4}(1+\varepsilon)^3\right] + S_0[t] \tag{16}$$

where $T = (1/\alpha)\ A_b/A_a$, $\varepsilon = A_b/A_a$ and subscript '0' denotes temporal moment of the input concentration-vs-time relationship at $x = 0$. An application of this approach to determine the transport parameters (T, ε, D_a and u) in a tracer test in a stream is given in the paper titled "Radiotracer Applications: Case studies from four continents", from Jung et al., included in this publication.

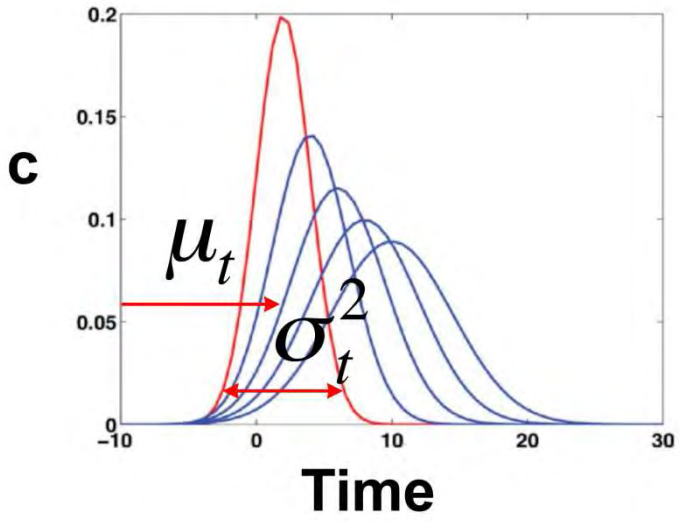


FIG. 8. Schematic graph of breakthrough curves and the first temporal moment (μ_t) and the second central temporal moment (σ_t).

One option to evaluate the model parameters is to optimize between predicted and observed temporal moment using similar optimization principles as discussed in § 6.2. If the number of temporal moment equations equal the number of unknown model parameters we have a determined system that can be determined exactly. Wörman [12] derived the following formulas for model parameters of (14) – (16), which applies when the longitudinal dispersion can be neglected in comparison to the effect of transient storage:

$$T = \frac{1}{3}\frac{dS}{d\sigma^2} \tag{17}$$

$$u = \left[\frac{d\mu_t}{dx} - \frac{3}{2}\frac{d\sigma^2}{dx}\frac{d\sigma^2}{dS}\right]^{-1} \tag{18}$$

$$\varepsilon = \left[\frac{4}{6}\frac{d\mu_t}{d\sigma^2}\frac{dS}{d\sigma^2} - 1\right]^{-1} \tag{19}$$

6.4. Two-dimensional moment matching technique for transversal dispersion

For the initial mixing phase immediately after injection as discussed in § 5.1, we can integrate Eqn. (6) over time and solve for the property $\theta(x,y) = \int_0^t c(x,y)dt$ under the assumption that the transverse velocity $v = 0$. Hence, one can derive the following formula for the transverse dispersion coefficient [23]:

$$D_{T,a} = \frac{u}{2}\frac{d\sigma_y^2}{dx} \tag{20}$$

in which σ_y is the standard deviation of the activity distribution transversal to the stream direction.

REFERENCES

[1] KADLEC, R. H., KNIGHT, R. L., Treatment Wetlands, CRC Press LLC, USA (1996).
[2] LEVENSPIEL, O., Chemical Reaction Engineering, third edition, John Wiley & Sons Inc., USA (1999).
[3] FOGLER, H. S., Elements of Chemical Reaction Engineering, third edition, Prentice Hall PTR, USA (1999)
[4] CENGEL, Y., CIMBALA, J., Fluid Mechanics: Fundamentals and Applications with OLC, McGraw Hill. SBN: 9780071117203 (2005).
[5] FRENCH, R., Open channel hydraulics, McGraw-Hill International Editions (1994).
[6] KJELLIN, J., WÖRMAN, A, JOHANSSON, H, LINDAHL, A, Controlling factors for water residence time and flow pattern in Ekeby treatment wetland, Sweden, Advances in Water Resources **30** (2007) 838-850.
[7] RUTHERFORD, J.C., River Mixing, John Wiley, New York, USA (1994).
[8] BENCALA, K.E., WALTERS, R.A., Simulation of solute transport in a mountain pool-and-riffle stream: a transient storage model, Water Resources Res. **19** 3 (1983) 718-724.
[9] HARVEY, J.W., WAGNER, B.J., Experimental design for estimating parameters of rate-limited mass transfer: Analysis of stream tracer studies, Water Resour. Res. **33** 7 (1997) 1731-1741.
[10] JACKMAN, A.P., WALTERS, R.A., KENNEDY, V.C., Transport and Concentration Controls for Chloride, Strontium, Potassium and Lead in Uvas Creek, a Small Cobble-Bed Stream in Santa Clara County, California, USA., J. Hydrology **75** (1984) 111–141.
[11] WÖRMAN, A., et al., Effect of Flow-Induced Exchange in Hyporheic Zones on Longitudinal Transport of Solutes in Streams and Rivers, Water Resources Research: **38** 1 (2002) 2:1-15
[12] WÖRMAN, A., Comparison of Models for Transient Storage in Small Streams, Water Resources Research: **36** 2 (2000) 455-468.
[13] WÖRMAN, A., PRZEMYSŁAW, W., Reach scale and evaluation methods as limitations for transient storage properties in streams and rivers, Water Resour. Res. **43** 10 (2007).
[14] CRANK, J., Mathematics of Diffusion, Oxford Science Publications (1973).
[15] CARSLAW, JAEGER, Conduction of heat in solids, Oxford Science Publications (1959).
[16] DE SMEDT, Analytical solution and analysis of solute transport in rivers affected by diffusive transfer in the hyporheic zone, Journal of Hydrology **339** (2007) 29– 38.

[17] WÖRMAN, A., 1998. Analytical Solution and Time Scale of Solute Transport in Streams and Rivers, Water Resources Research **34** 10 (1998) 2703-2716.
[18] MALOSZEWSKI, P., ZUBER, A., Mathematical modelling of tracer behaviour in short-term experiments in fissured rocks, Water Res. Res. **26** 7 (1990) 1517-1528.
[19] NORDIN, C. F., TROUTMAN, B. M., Longitudinal dispersion in rivers: The persistence of skewness in observed data, Water Resour. Res., **16** (1980) 123–128.
[20] SCHMID, B.H., On the transient storage equations for longitudinal solute transport in open channels: temporal moments accounting for the effect of first-order decay, J.Hydr.Res., **33** 5 (1995) 595-610.
[21] CZERNUSZENKO, W., ROWINKSI, P. M. Properties of the dead-zone model of longitudinal dispersion in rivers, J. Hydraul. Res., **35** (1997) 491– 504.
[22] SCHMID, B. H., Temporal moments routing in streams and rivers with transient storage, Advances in Water Resources **26** (2003) 1021–1027.
[23] SEO, I.W., BAEK, K.O., JEON, T.M., Analysis of transverse mixing in natural streams under slug tests, J. Hydraulic Res. **44** 3 (2006) 350–362.

RADIOTRACER APPLICATIONS: CASE STUDIES FROM FOUR CONTINENTS

S.H. JUNG[*], J.V. BANDEIRA[**], P. BRISSET[***], A.WÖRMAN[****], P.L. AIREY[*****], C.E. HUGHES[*****]

[*]Korea Atomic Energy Research Institute, KAERI, Republic of Korea
[**]Brazilian Nuclear Energy Commission, CNEN/CDTN, Brazil
[***]French Alternative Energies and Atomic Energy Commission, CEA, France/IAEA
[****]Royal Institute of Technology, KTH, Sweden
[*****]Australian Nuclear Science and Technology Organisation, ANSTO, Australia

Abstract

Case studies are a simple way to demonstrate how radiotracers can be successfully used in the environment in addressing water resources, contaminant transport and coastal management issues. This paper presents ten case studies from Korea, France, Brazil, Hong Kong, Australia, Belgium and Sweden using a variety of radiotracers including ^{99m}Tc, ^{198}Au, ^{3}H, ^{82}Br, ^{32}P, $^{175+181}$Hf, ^{160}Tb, ^{51}Cr(III), ^{65}Zn, ^{54}Mn and ^{35}S. These studies address physical transport processes such as dispersion and mixing, reactive transport and adsorption and contaminant uptake. Traced components include water, effluent, nutrients, contaminants and mud in rivers, lakes, wetlands and coastal waters.

1. PHYSICAL TRANSPORT PROCESSES – HYDRODYNAMICS, SUSPENDED SEDIMENT DYNAMICS

When a pollutant is introduced into rivers as a point source, it gets mixed in three directions as it moves with the river flow. Since, in most rivers, width is much larger than depth, the pollutant becomes well mixed vertically in a short time before it is mixed transversely. Therefore, the vertical mixing is only important close to the contaminant source. Thus, in the mid-field, the vertical concentration gradients can be neglected, and our attention can be focused on the transverse and longitudinal changes of the depth-averaged concentrations of the pollutant. Transverse mixing is more important in water quality management than either vertical or longitudinal mixing, especially when dealing with the discharge of wastes from point sources or the mixing of tributary inflows [1].

When the two-dimension model is applied to predict the concentration variations of pollutants in natural streams, dispersion coefficients must be properly selected. To obtain the observed dispersion coefficients, many investigators conducted tracer tests in natural streams.

1.1. Case Studies 1-3: Determining dispersion coefficients in Korean rivers

1.1.1. Case Study 1. Measurement of a dispersion coefficient for a river with $^{99m}TcO_4^-$ as a tracer

The dispersion process in a natural river with many curvatures is so complicated that its dispersion coefficients should be evaluated carefully. Unfortunately, they have seldom been obtained from the field. In many cases, the dispersion coefficients for simulating the dynamic behaviour of rivers are cited from the research found in the literatures that were carried out on their own rivers with much different conditions in terms of the hydrodynamic and geological parameters. This causes a potential risk that the self-purification capacity of a river could be overestimated. The in-situ measurement of a dispersion coefficient can supply numerical modellers with practically useful data and contribute to the sustainable development of natural resources [2].

The mixing characteristics of natural rivers were investigated by analyzing their radioisotope concentration curves for transverse dispersion coefficients. A short half-life radionuclide was

injected instantaneously into a flow as a point source by means of an underwater glass-vial crusher. The detection was made with 2" NaI(Tl) scintillation detectors bound to the transverse lines at a downstream position.

The river was assumed as a two-dimensional system for a coefficient evaluation due to the low water depth (< 0.5 m) in most of the sample areas when compared to their lateral and longitudinal size. Transverse dispersion coefficient was calculated by the moment method.

The two-dimensional advection-dispersion equation (1) can be expressed as below.

$$\frac{\partial C}{\partial t}+u\frac{\partial C}{\partial x}+v\frac{\partial C}{\partial y}=\frac{1}{h}\frac{\partial}{\partial x}\left(hD_L\frac{\partial C}{\partial x}\right)+\frac{1}{h}\frac{\partial}{\partial y}\left(hD_T\frac{\partial C}{\partial y}\right) \quad (1)$$

Where C = depth-averaged concentration; t = time; x = longitudinal distance measured from the injection point; y=transverse distance measured from the left bank; u, v = depth-averaged longitudinal and transverse velocities, respectively; h = depth; and D_L, D_T = longitudinal and transverse dispersion coefficients, respectively.

The moment equation for the transverse dispersion coefficient in the simple moment method is as follows (2):

$$D_T=\frac{u}{2}\frac{\partial}{\partial x}\left(\frac{\int_0^w \theta y^2 dy}{\int_0^w \theta dy}\right)=\frac{u}{2}\frac{d\sigma_y^2}{dx} \quad (2)$$

where σ^2_y is the variance of the transverse distribution of the depth-averaged concentration. In this study, D_T was obtained from the slope of the straight line that was fitted to the data in the plot of σ^2_y versus x.

As a result, the coefficients ranged from 0.0194 to 0.0097 m^2/s. It was also seen that the number of measurement sections definitely affects the accuracy of the estimation by the simple moment method.

In Fig. 1 are represented the results of radiotracer measurement after two separate injections

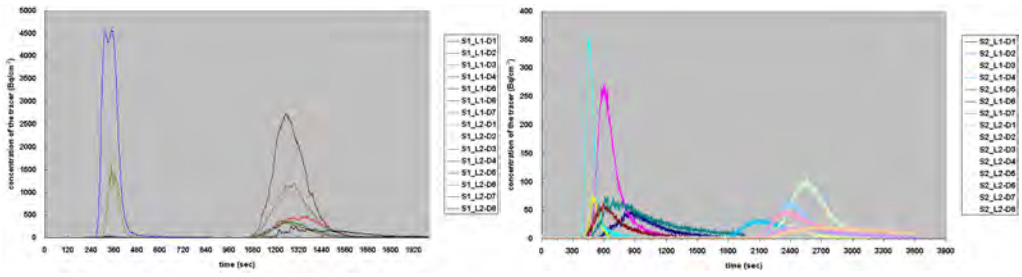


FIG. 1. Radiotracer Measurement Results from the two separate radiotracer injections.

1.1.2. Case Study 2. Analysis of transverse mixing in natural streams by the stream-tube routing procedure

Tracer tests were conducted at several different sites in tributaries of a main river in order to investigate the effects of the stream geometry on the transverse mixing of the pollutant.

Transverse dispersion coefficients were calculated from the observed concentration data obtained by the radiotracer experiments. The field data showed that, in most tests on the curved reaches of the streams, the high tracer concentration as well as the high velocity occurred following the thawed line. However, the local irregularities inside the stream cross-section significantly affected the transverse distributions of stream velocity and tracer concentration as much as the channel meander did. The analysis of the relations between the transverse dispersion coefficients and the basic hydraulic parameters showed that as the ratio of width to depth increased, the transverse dispersion coefficients tended to increase. The transverse dispersion coefficients were also proportional to the sinuosity of the stream. [3]

The difficulty with the moment method is that the skewed observed distribution due to irregular water depth and reflection at the bank makes it difficult to computer a meaningful accurate value of the variance. Thus, in the study, the routing procedure combined with the stream-tube concept was developed. In the routing procedure proposed by Fischer [4], the dispersion coefficient is calculated by matching the downstream observation of a passage of a tracer cloud to the prediction based on an upstream observation.

The routing procedure combined with the stream-tube concept was developed for the analysis of the dispersion data collected in the natural stream with irregular geometry by slug test. The results (Table 1) were compared with the results by the conventional moment methods.

TABLE 1. COMPARISON OF OBSERVED TRANSVERSE DISPERSION COEFFICIENTS

Test case	D_T/HU_*			
	Yotsukura and Sayre (1976), Eq. (17)	Simple MM, Eq. (6)	Stream-tube MM, Eq. (11)	Stream-tube RP, Eq. (14)
S-Expt 1	0.43	0.45	0.43	0.46 (Secs. 3–6)
S-Expt 2	0.36	0.76	0.85	1.21 (Secs. 2–4)
S-Expt 3	—	0.27	0.32	0.30 (Secs. 1–5)
C-Expt 1	0.08	0.24	0.34	0.27 (Secs. 2–5)
H-Expt 1	1.29	0.47	0.85	0.64 (Secs. 2–6)
H-Expt 2	0.08	0.24	0.24	0.23 (Secs. 2–6)
H-Expt 3	0.20	0.33	0.29	0.32 (Secs. 3–6)

1.1.3. Case Study 3. Determination of dispersion coefficients using radioisotope data in river environment

Field tracer experiments using radioisotope labelled chemicals, $^{99m}TcO_4^-$, $Na^{131}I$ and $NH_4{}^{82}Br$ were performed to estimate the dispersion characteristics of pollutants in river environment. The dispersion coefficients in the longitudinal and transverse directions were determined by using the measured concentration of a radioisotope. Numerical models were applied to calculate the flow and concentration fields at the experimental site [5, 6].

The computational domain for the hydrodynamic and dispersion simulations is composed of 1077 elements and 3540 nodes. It has two open boundaries. Boundary conditions for the simulation were established by measured discharge at upstream and water surface elevations at downstream. Analytical solution of a two-dimensional advection-diffusion equation is used to determine the dispersion coefficients by using the measurement concentrations of the

radioisotope. Analytic solution of a two-dimensional advection-diffusion equation is as follows:

$$c = \frac{M}{4\pi Ht(D_x D_y)^{1/2}} \exp\left(-\frac{(x-ut)^2}{4D_x t} - \frac{(y-vt)^2}{4D_y t} - \lambda t\right) \quad (3)$$

Here M is the total tracer amount, H is the water depth, u and v are the flow velocities, λ is the decay constant, D_x and D_y are the longitudinal and transverse dispersion coefficients, respectively.

A least square method for the experimental data was used to obtain the best values for the dispersion coefficients in the longitudinal and transverse directions. The values of D_x and D_y obtained by the measured concentration data of the radioisotope were 0.27 and 0.042m^2/sec, respectively. Sensitivity analysis was performed to investigate the effects of the numerical results according to variations of the dispersion coefficients. The calculated results for several runs were compared with the measured ones by using statistical methods as shown in Fig. 2.

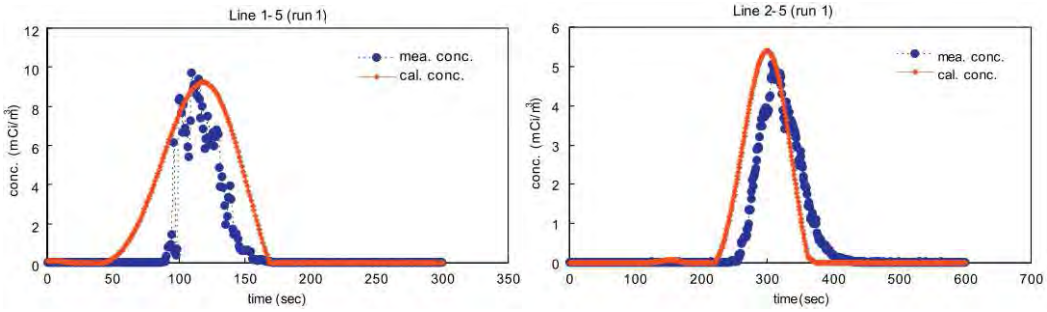


FIG. 2. Comparison of the calculated and measured concentrations at the centre point of detection line 1 and 2 in Run (1) condition.

1.2. Case Study 4. Urban wastes release in the Mediterranean Sea program, Toulon, France

These experiments (Table 2) are performed to study the behaviour of the contaminants and effluents discharged by the Toulon – Sainte Marguerite WWTP in the Bay of Toulon. The general idea is to improve knowledge on the dispersion phenomenon in order to develop means and methodology helping to a better management of the coastal waters of the Mediterranean Sea facing the contaminants brought into the medium by urban discharges [7-12].

The release is made through a pipe 1800 m offshore at a depth of 45 m. The average concentration is 45 mg/L and the average flow-rate is 16800 m^3/day (200 000 eq. population).

The experimental methodology consists in injecting radiotracers at the outlet of the WWTP and following their dispersion and behaviour in the Bay of Toulon:

- Water tracing experiment to study water and conservative solute contaminants dispersion in the near-field water column, using ^{99m}Tc in technetium pertechnetate form;
- Sludge particles experiment to study particles (considered as vectors of pollutants) dispersion in the near-field water column, using ^{198}Au in chloride solution to label natural particles;

- Sludge particles experiment to study the behaviour of the particles in the far field (i.e. the entire Bay of Toulon) to assess their deposit and potential accumulation on the sea bed, using $^{175+181}$Hf in chloride solution to label natural particles.

TABLE 2. EXPERIMENTAL RADIOTRACERS INJECTION IN THE BAY OF TOULON

Experiment	Area of interest	Tracer	Injected Activity	Chemical form
Water tracing without and without thermocline	Water column near-field	^{99m}Tc 140 keV, %	37 GBq	Technetium pertechnetate
Particles tracing with and without thermocline	Water column near-field	^{198} Au	333 GBq Instantaneous injection	Gold chloride
Particles tracing for far field study	Far field sea bottom	$^{175+181}$Hf	333 GBq	Hafnium chloride

The tracer detection is performed both through static and dynamic methods for near-field experiments, through dynamic method for far field experiment.

The scheme of the near-field experiments is shown in Fig. 3 below.

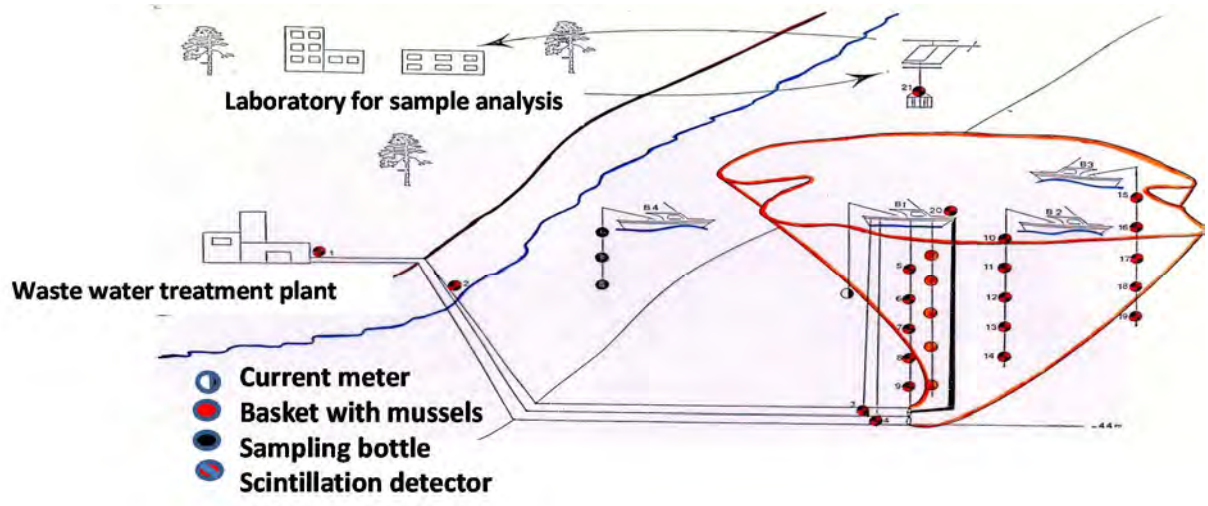


FIG. 3. Far field experiment.

In this experiment the natural sludge particles are labelled by ^{181}Hf initially in chloride solution. Hf is fixed on the particles through a oxidation-reduction reaction in an indelible way. The labelled particles are injected in the outfall pipe with a micro-peristaltic pump continuously during 35 days in order to integrate various hydrodynamical and meteorological conditions and thus to obtain an average value of the deposit along a representative period of time. Periodically a boat equipped of e sledge and radiation scintillation detector scan the sea bottom around the outfall exit to map the particles deposits and thus to evaluate quantitatively

the impacted area. This mapping is a support for and is completed by samplings for pollutants analysis.

Environmental impact assessment - Input data:

- Total activity injected: 333 GBq at the end of injection (decay not considered)
- Duration of the injection: 35 days
- Average flow rate: 16800 m^3/day
- Average concentration in the outfall pipe: 6000 Bq/L
- Surface of deposit measured on site: about 6 km x 4 km = 24 x 106 m^2
- Superficial concentration (hyp. 100% of the tracer is deposited): 13 875 Bq/m^2
- Maximum concentration measured: 250 cps i.e. Bq/m^2
- Supposing 50 cps per µCi/m^2 > 250 cps correspond 5 µCi/m^2 from 5 x 37000 Bq/m^2 to 185000 Bq/m^2
- NB this value is observed only on a small area during the 2nd detection i.e. at the end of injection

Figs 4, 5 and 6 show the map of the tracer deposit on the sea bed 33, 63 and 104 days after the beginning of the injection.

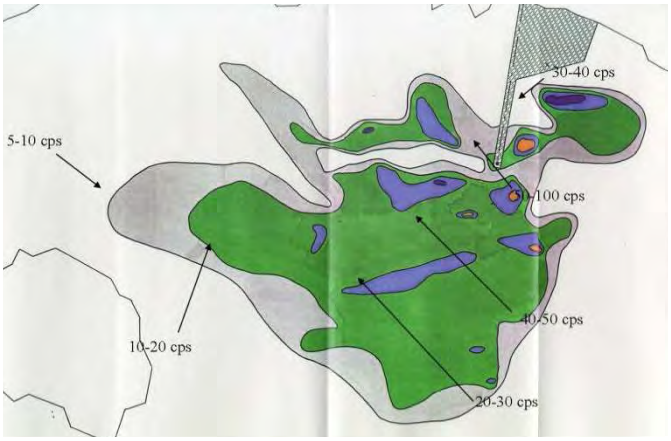


FIG. 4. Map of the tracer deposit on the sea bed 33 days after the beginning of the injection i.e. 2 days before its end (det 2).

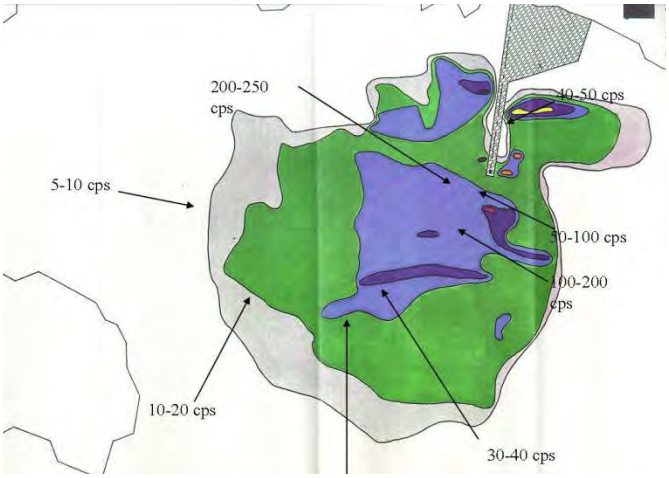


FIG. 5. Map of the tracer deposit on the sea bed 63 days after the beginning of the injection i.e. 28 days after its end (det 3).

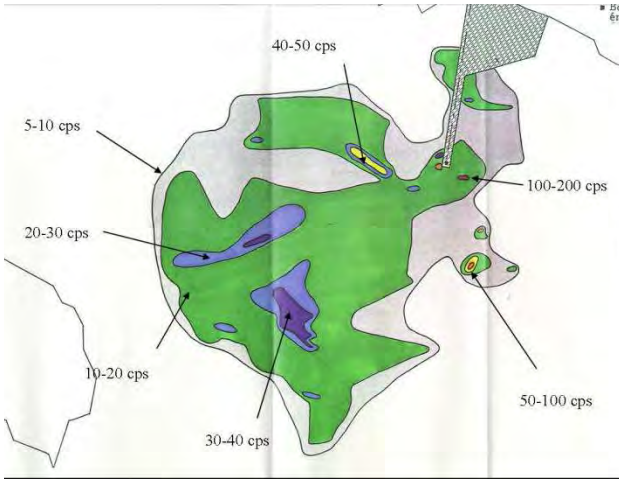


FIG. 6. Map of the tracer deposit on the sea bed 104 days after the beginning of the injection i.e. 69 days after its end (det 4).

Fig. 7 illustrates the see balance of deposit and the conversion cps Bq/m^2 for Hf.

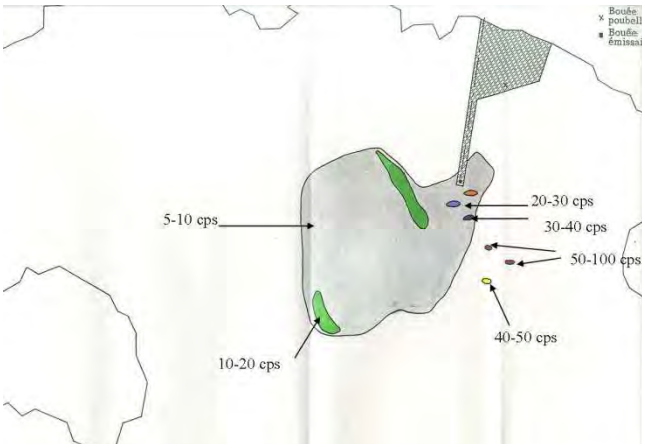


FIG. 7. The balance of deposit, conversion cps Bq/m2 for Hf.

1.3. Case Study 5. Labelling of mud in reservoir dredging studies using ^{99m}Tc (Brazil)

The study was performed to evaluate the viability of an environmental and perennial solution to dredge the fine sediment that accretes (400,000 m^3/year) the Pampulha reservoir, in Belo Horizonte, Brazil, which is in an accelerated process of reduction of its liquid volume and water surface (Fig. 8). With this trend, the reservoir could lose, in a near future, two of the main purposes for which it was built: flood damping and leisure region, with its water surface indubitably linked to the buildings designed in the 1940 decade by the famous Brazilian architect Oscar Niemeyer, forming the Architectonic Complex which is the landmark of the modern Brazilian Architecture.

There is no available area inland to dump the dredged material from the reservoir. The watercourses downstream the dam are the natural way for the sediment that accretes the reservoir if the dam were not constructed. In this way, field experiments, with simultaneous and instantaneous injections of sediment and water, labelled, respectively, with ^{99m}Tc and Rhodamine WT (Table 3), were performed, in dry season, in 2000 and 2001, to measure the hydro-transport capability downstream the dam, in a stretch of 25 km (Fig. 9). They allowed comparing the different hydrodynamic behaviours of the mud in suspension and the water transporting it [13].

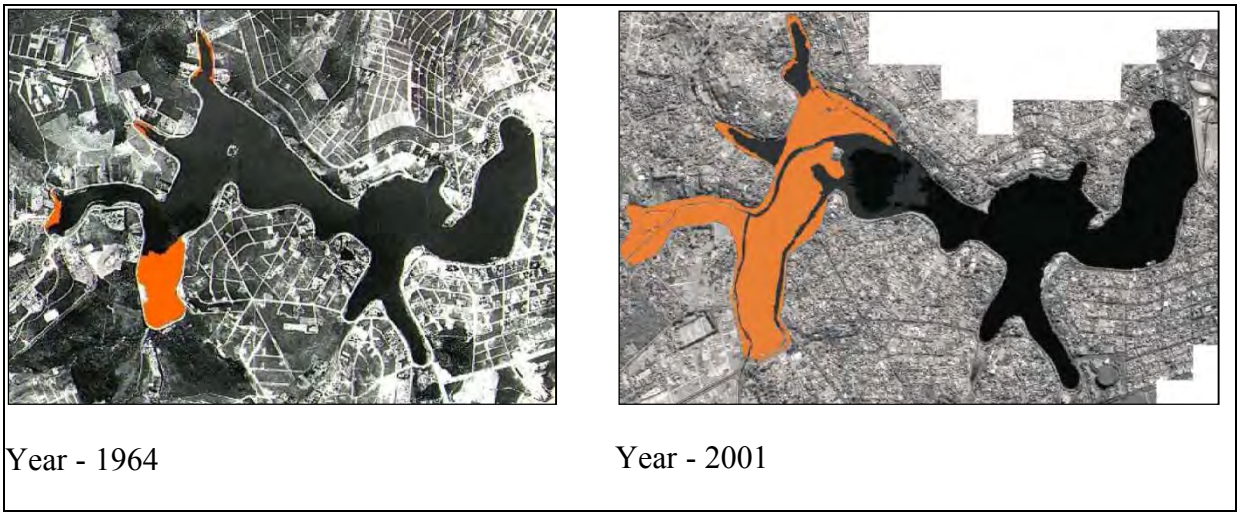


FIG. 8. Pampulha Reservoir accretion (orange), Belo Horizonte, Brazil[13].

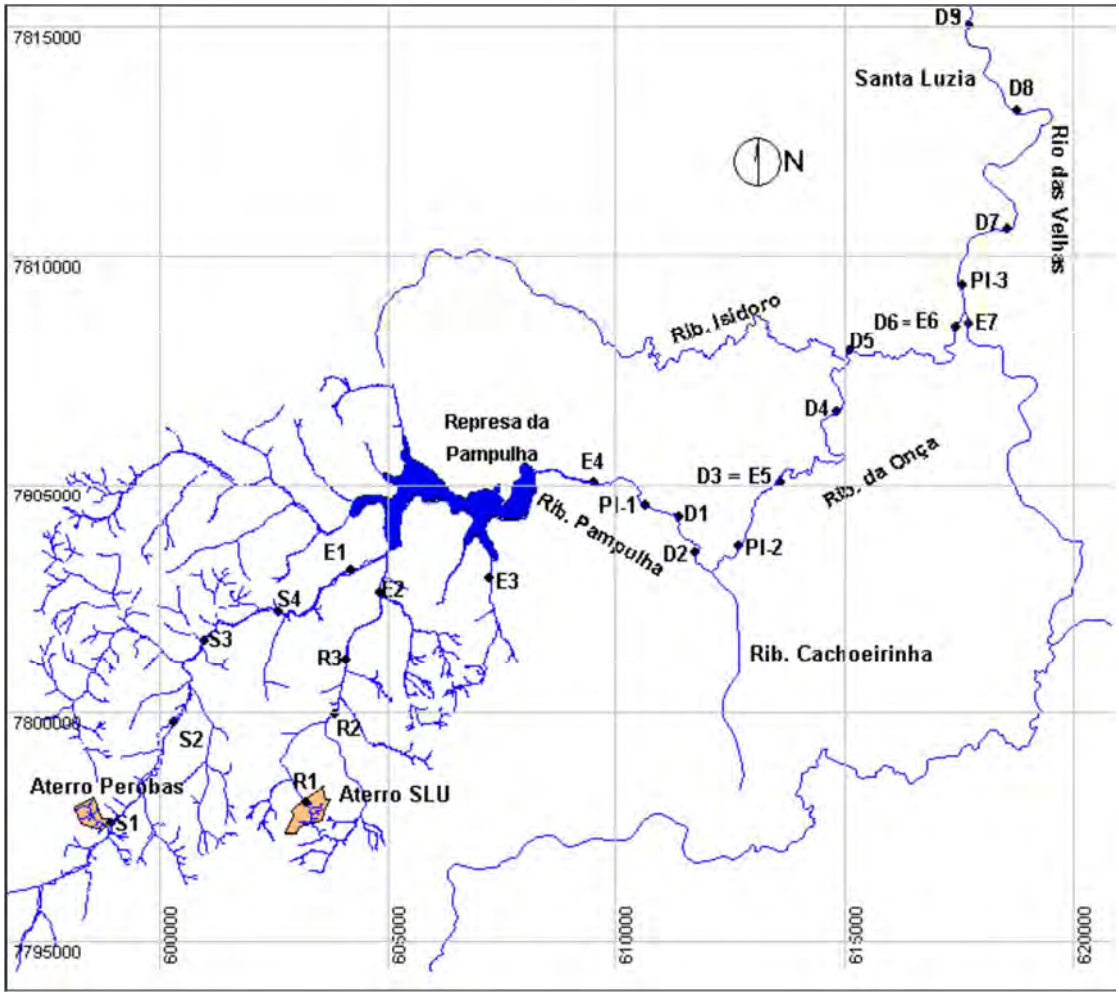
TABLE 3. FIELD EXPERIMENTS WITH COMBINED LABELLING FINE SEDIMENT WITH ^{99m}Tc & WATER WITH RHODAMINE WT

Date	Injection Point[(a)]	Site	Detection Station [(a)]	Initial activity of ^{99m}Tc (GBq)	Initial sediment conc. (g/L)	Rhodamine WT at 20% volume (L)
09/27/2000	PI-1	Pampulha& Onça creeks	D1, D2 & D3	13.3	41.9	2
10/03/2000	PI-2	Onça Creek	D3, D4, & D6	12.2	72.4	2
07/03/2001	PI-2	Onça Creek	D3, D4, D5 & D6	19.5	54.4	4
06/26/2001	PI-3	Velhas River	D7, D8 & D9	50.3	47.1	6

(a) See Fig. 9.

A mathematical model [15] was applied and calibrated to the data obtained: advection velocity and longitudinal dispersion. Through convolution, the sediment dumping in suspension using hydraulic dredging system was simulated, calculating also, the physical environmental impacts: increase of sediment concentration and the possibility of deposition. The measurement of physical-chemical parameters of the water allowed to evaluate the possibility of desorption of the metals adsorbed in the sediment to be dredged and dumped downstream. It was concluded that there is no impairment for the dumping of the dredged fine sediment in the watercourses downstream [13].

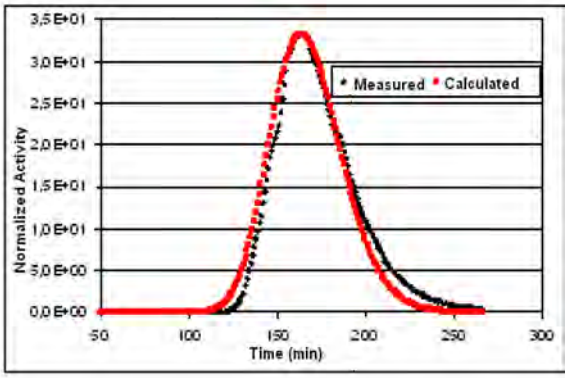
As pointed out before, the simultaneous use of two tracers allowed comparing the different hydrodynamic behaviours of the mud in suspension and the water transporting it. The model of Singh & Beck [15] considers that water and the component being transported (solute or tracer) have the same density. While it did not occur, in the creeks, the presence of dead zones able, by its geometry and situation in the water flow, to temporarily detect the sediment in suspension, with density higher than that of the water, the model represented very well both phases (Fig. 10– stretches before D5). Fig. 11 shows the simultaneous propagation of the fine sediment and the water. Just upstream of the station D5 (experiment of 10/03/2000) there is a dead zone (Fig. 12) and the behaviour of the sediment in suspension, as detected in the station D6, differed from the one foreseen by the model and also in relation to the behaviour of the water (higher longitudinal dispersion coefficient), considering also the results of the experiment of 07/03/2001 [13].



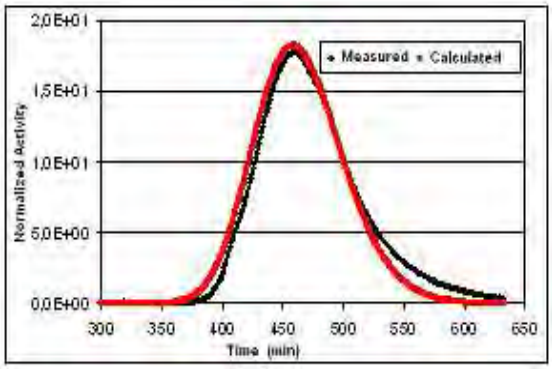
PI-1 to PI-3: Injection points (instantaneous injection of fine sediment labelled with ^{99m}Tc.
Stations D1 a D9: Static detection of the radioactive & fluorescent tracer clouds.
Obs.: Represa da Pampulha = Pampulha Reservoir; Rib. Pampulha = Pampulha Creek; Rib. da Onça = Onça Creek; Rio das Velhas = Velhas River, Aterro = Landfill

FIG. 9. Studied region in Pampulha Hydrographic Basin and Velhas River [13].

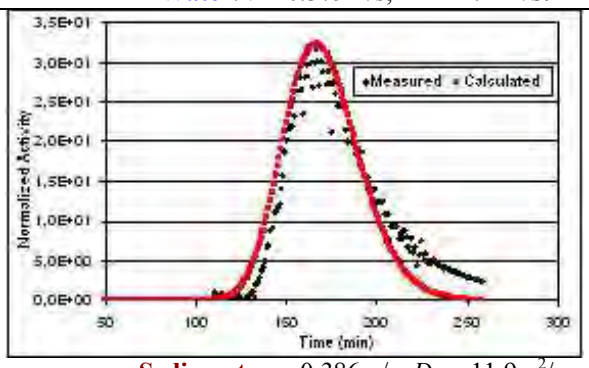
As pointed out before, the simultaneous use of two tracers allowed comparing the different hydrodynamic behaviours of the mud in suspension and the water transporting it. The model of Singh & Beck [15] considers that water and the component being transported (solute or tracer) have the same density. While it did not occur, in the creeks, the presence of dead zones able, by its geometry and situation in the water flow, to temporarily detect the sediment in suspension, with density higher than that of the water, the model represented very well both phases (Fig. 10– stretches before D5). Fig. 11 shows the simultaneous propagation of the fine sediment and the water. Just upstream of the station D5 (experiment of 10/03/2000) there is a dead zone (Fig. 12) and the behaviour of the sediment in suspension, as detected in the station D6, differed from the one foreseen by the model and also in relation to the behaviour of the water (higher longitudinal dispersion coefficient), considering also the results of the experiment of 07/03/2001 [13].



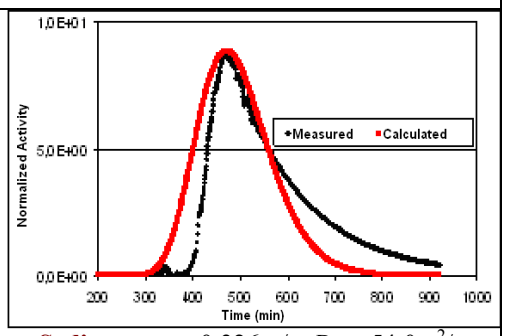
Water: $v = 0.379$m/s; $D = 11.2$m^2/s.



Water:$v = 0.344$m/s; $D = 10.2$m^2/s.

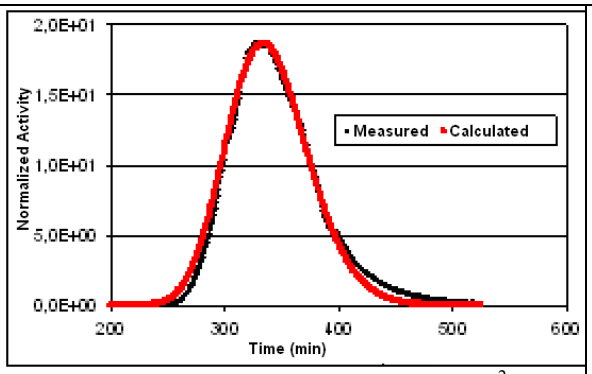


Sediment: $v = 0.386$m/s; $D_L = 11.9$m^2/s.

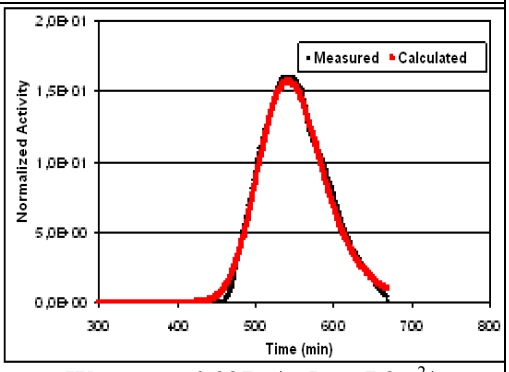


Sediment: $v = 0.326$m/s; $D_L = 54.0$m^2/s.

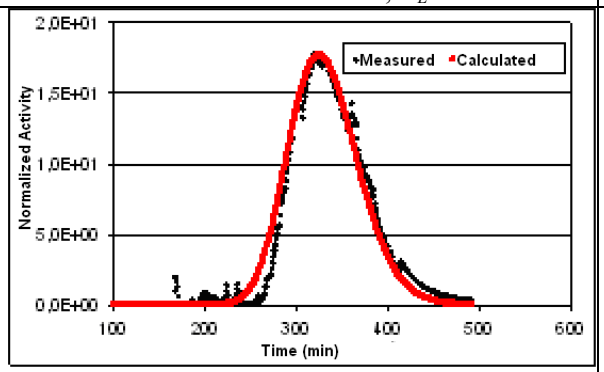
Stretch D3-D4 **Day 10/03/2000** **Stretch D4-D6**



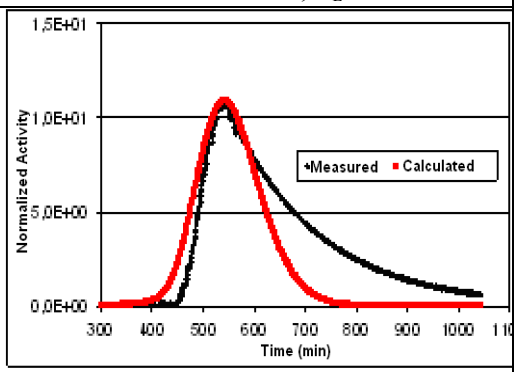
Water: $v = 0.282$m/s; $D_L = 9.4$m^2/s.



Water: $v = 0.327$m/s; $D_L = 7.2$m^2/s.



Sediment: $v = 0.290$m/s; $D_L = 11.4$m^2/s.



Sediment: $v = 0.320$m/s; $D_L = 30.6$m^2/s.

Stretch D3-D5 **Day 07/03/2001** **Stretch D5-D6**

FIG. 10. Detection & mathematical model curves [13].

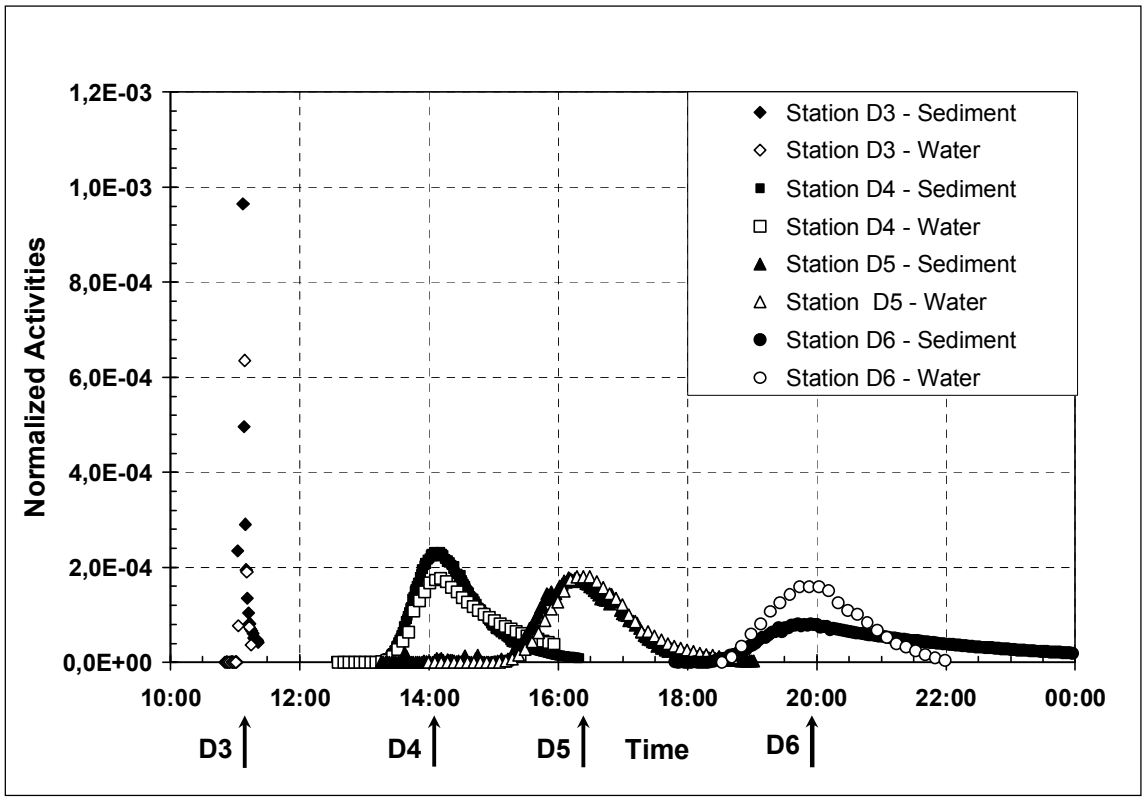


FIG. 11. Fine sediment & water experiment of 07/03/2001 [13].

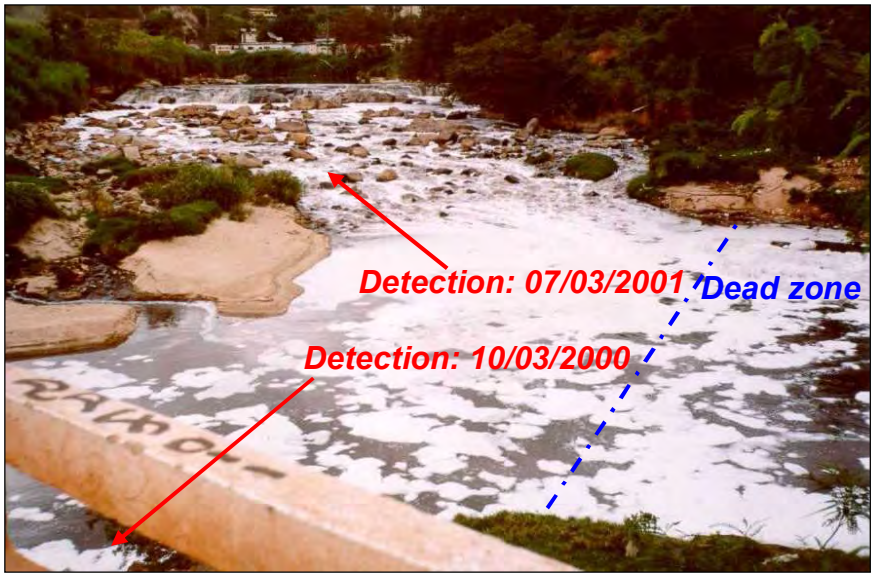


FIG. 12. Station D5 (Onça Creek).

The hydrodynamic behaviour, unequal for water and the fine sediment in suspension, in the presence of dead zones, can be studied in more detail with these tracer techniques, in controlled situations, aiming at, for instance, to obtain a parameter to be introduced in the model of Singh & Beck [15], to take into account the density of the sediment. This double tracer technique (simultaneous labelling of water and fine sediment in suspension) could be used for in situ and physical model studies of the influence of dead zones. These studies encompass the siltation mechanisms and the exchange of flow and sediment between the main

river and harbour basins aiding also in the design of sills and deflecting walls to hamper the access of fine sediment to the harbour basin. This technique is also a powerful tool for scientific research of the behaviour of fine sediment in suspension, in water environment, reactive transport and retention.

1.4. Case Study 6. Effluent dispersion from the Tolo Effluent Export Scheme, Hong Kong

A radiotracer study of the transport of sewage effluent diverted from the Tolo Harbour Water Control Zone through the Kai Tak Nullah (typhoon shelter) was undertaken in July 1999. The diversion project hoped to achieve two major environmental goals:

- The reduction of nitrogen loads to Tolo Harbour;
- An increase in flows in the upper sections of the Kai Tak Nullah to increase the flushing rate of the nullah and improve water quality.

Particulates were labelled using ^{198}Au and water was labelled with tritium. The mean effluent flow rate during the injection period was 3.5 m^3/s. The labelled effluent was tracked within the nullah and in Victoria Harbour for 6 days using (Fig. 13). ^{198}Gold, with a half-life of 2.69 days, exhibited considerable decay during the course of the investigation so results are decay corrected to allow comparisons between data collected on different days.

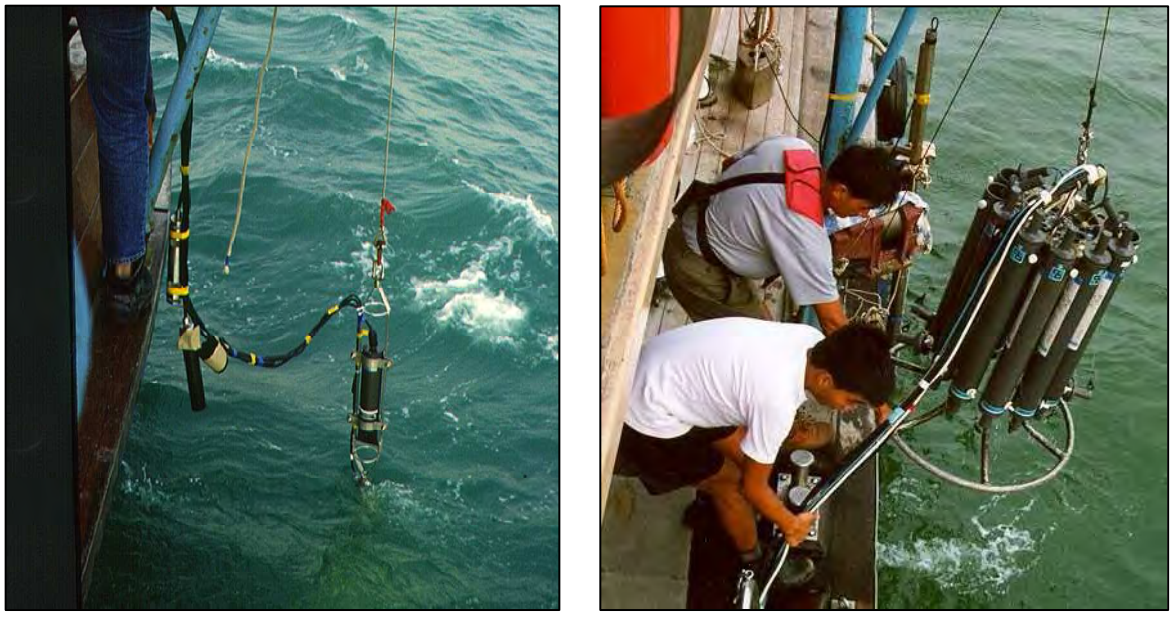


FIG. 13. Deployment of scintillation counters for in-situ measurement of ^{198}Au and sampler for tritium analysis.

The movement of the radiotracer plume over four days was studied. The observed mean residence time for near surface effluent was about 1 day with tides exerting little influence on net transport. In contrast the mean residence time upstream of the typhoon shelter has been estimated to be about 5 days.

Considerable flow stratification was observed (e.g. Fig. 14, 18 July HHW) with the near surface accounting for about 66% of the ^{198}Au activity (particle fines) at Station 1A and about 50% at Station 1. Fig. 15 shows the change in surface and bottom concentrations with time of

both ^{198}Au and HTO (where 2.5 Bq/L of HTO is equivalent to 1 count per second of ^{198}Au) at Site 1 at the entrance to the nullah. The settling of particulates can be clearly seen at this location as the surface concentration of ^{198}Au at the surface decreases and at the bottom increases with time, until the tritium plume has passed.

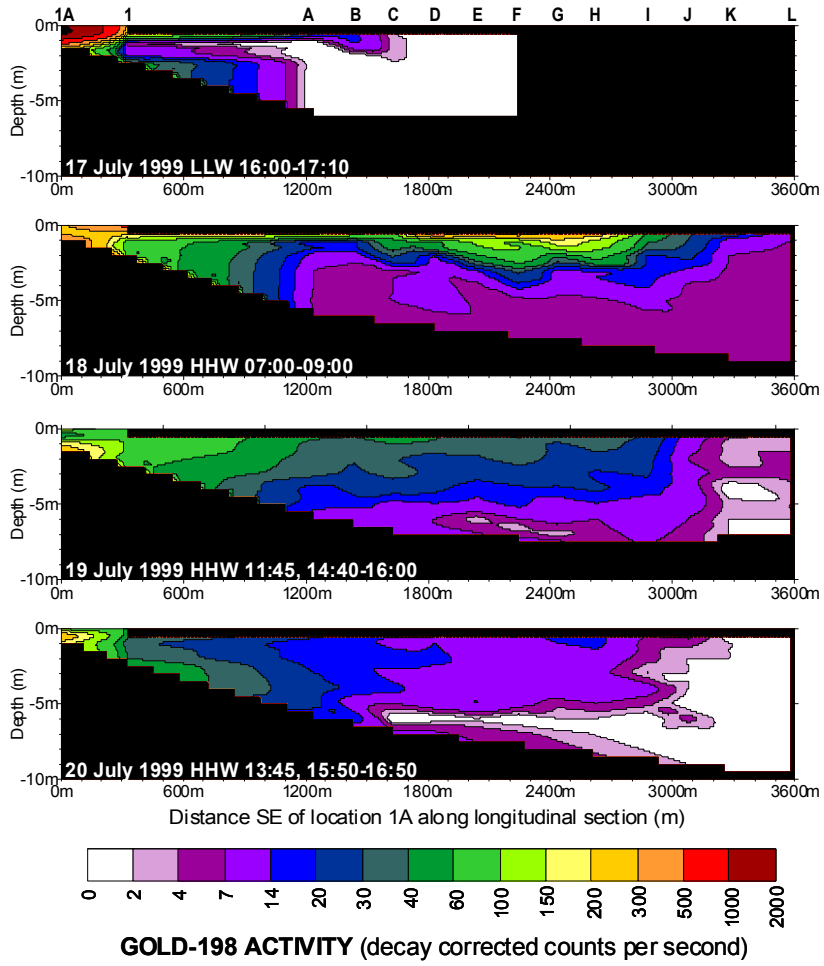


FIG. 14. Vertical profiles along the flow path from Site 1A to L (Shown at the top of the figure).

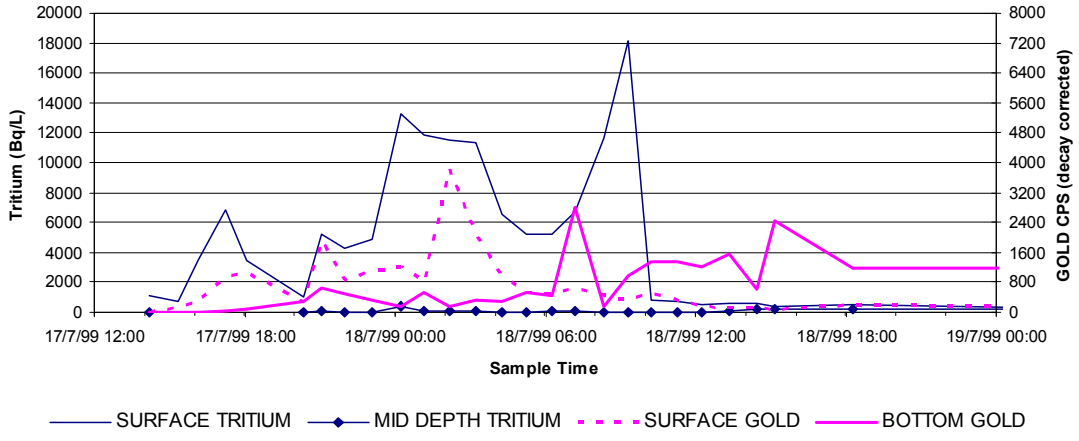


FIG. 15. Concentration of ^{198}Au and tritium at Site over the first 36 hours.

In the more open parts of the nullah during flood tides a wedge of higher density marine water, which is relatively low in Tolo effluent, forms near the bottom whilst the effluent continues to flow out at the surface. While the bulk of the activity was close to the surface due to density stratification, there was also an accumulation close to the bottom (e.g. Fig. 14, 20 July) resulting from on-going settlement of the heavier ^{198}Au labelled particles.

The Tolo radiotracer study was used as an independent method of assessing effluent retention times in the nullah and dilution in Victoria Harbour. In this case tracer studies were not used for model calibration.

2. REACTIVE TRANSPORT AND RETENTION

Most common radioactive tracers undergo reactions and this may have to be considered in the evaluation of a tracer test as well as in selection of tracer. The case studies in this paper consider several reactive, radioactive tracers, such as $^{99m}TcO_4^-$, $Na^{131}I$ and $NH_4{}^{82}Br$, ^{32}P and ^{51}Cr(III). Most cationic tracers, like heavy metals, have a sorption affinity to particulate matter that depends on factors like pH, redox and particle concentration [16]. Specifically, tracers adsorbed to deposited sediments are practically immobile for some time, which provides a significant retardation of the transport process. Such retardation can be caused by sorption to particulate matter in the flowing water column and subsequent deposition on the stream-bed or filtering of colloidal matter in the stream-bed. Adsorption of the tracer to 'carrier' particles generally implies a retardation of the solute transport, but in some cases where alternative fixation mechanisms are present, the adsorption to colloidal particles may increase the mobility. Retardation can be caused also by direct exchange of the flowing water due to pressure variability along the bed surface caused by bed-roughness or stream curvature [17-19]. The flow of water and solute into the hyporheic zone facilitates adsorption of the tracer substance to solid surfaces [20].

In many cases it is important to select tracers that are representative for the transport process of interest, preferentially the same molecule. For example in order to study phosphate transformation in a hydrological system it can be suitable to use either $^{32}PO_4^-$ or $^{33}PO_4^-$ [21]. The identical molecule structure with naturally occurring phosphate (involving the stable isotope ^{31}P) suggests that its transformation would be analogous and the tracer test would be representative to a 'natural' transformation of phosphate that involves biological uptake and sorption. Similarly, the radioactive tracer molecule $^{51}CrCl_3$ is suitable for studying transport of ion forms of chromium in solution. The positively charged chromium ion is adsorbed to mineral surfaces especially as pH increases [22].

2.1. Case Study 7. Radiotracer Studies in the Magela Flood Plain, NT, Australia, 1977 to 1987

Over the decade 1977-87 a program of tracer studies were undertaken to assess the environmental impact of heavy metals and sulphate across the Magela flood plains. The Ranger Uranium Mine and the Jabiluka deposit lie within the catchment of the Magela Creek of the Northern Territory which flows into the East Alligator River and floods annually during the summer monsoon (Fig. 16). The levels of manganese and sulphate in the retention ponds at Ranger were sufficiently high to be limiting factors in any controlled release. These studies complement extensive investigations of the transport of uranium series nuclides downstream of the deposits.

A summary of the principal aims of the investigation and the results are listed in Table 4.

TABLE 4. PRINCIPAL AIMS OF INVESTIGATION AND RESULTS OF THE TRACER EXPERIMENT ACROSS THE MAGELA FLOOD PLAIN

No	Aim	Strategy	Results
1	To study the fate and behaviour of Zn released into the Magela Creek system	^{65}Zn (30 GBq) and HTO (3700 GBq) injected into the Magela Creek at IR and IPC at a steady rate over 32 h.	1) The ratio R of dissolved and 'colloidal' Zn tracer averaged 0.38[(1)]. 2) Zn/T ratio decreases systematically with distance due to Zn uptake on sediment and vegetation. The ratio fell from 0.9 10^{-2} (activity ratio at injection 0.8 10^{-2}) to a minimum 0.15 10^{-2} at the end of the main pulse before increasing to a max of 21 10^{-2} due to the mobilization of the retarded tracer which was subsequently followed onto the flood plain. 3) The half- life of the ^{65}Zn in the vegetation in the Jabiluka Billabong was about a two weeks.
2	To monitor the fate of the ^{65}Zn tracer in the subsequent dry season	Sediment and vegetation samples were collected on 11 July 1978 and monitored.	^{65}Zn levels were surveyed on sediment samples at three depths on the flood plain and in the new season vegetation. Soil samples taken at 0-8; 8-13 and >13 cm. In the upper level the average ^{65}Zn (decay corrected) was 33± 22 Bq/g. Three samples of new season growth collected on 11/7 and 14/8 recorded values between 5 & 37 Bq/g.
3	To investigate variations in chemistry on the uptake of Zn on flood plain vegetation.	^{69}Zn (with some ^{65}Zn 'impurity') and isotopically pure ^{65}Zn were used. The chemistry of the ^{65}Zn was modified by releasing an excess of either humic acid, or EDTA, or tannic acid, or KOH (zincate) in close proximity. The chemically altered plume did not overlap the ^{69}Zn.	The ^{69}Zn/^{65}Zn ratios observed from vegetable samples in the vicinity of the 4 impulse injection points on the floodplain north of the Jabiluka billabong were statistically the same as those at injection. It was concluded that altering the chemistry of the tracer had no major effect on vegetation uptake. However, the possibility of isotopic exchange between the two zinc isotopes cannot be completely excluded.
4	To study the behaviour of Mn relative to that of Zn.	A series of pulse releases of ^{54}Mn and ^{65}Zn in the Magela Creek (Georgetown Gauging Station) and the Jabiluka Billabong were made.	There was enhanced adsorption on the sediment bottom, vegetation and suspended sediment of ^{54}Mn and ^{65}Zn species not found in the natural water systems. The systematic variation of the experimental parameters ^{65}Zn (or ^{54}Mn)/HTO. ^{54}Mn/^{65}Zn with distance from the injection point is associated with the rate of establishment of equilibrium with the natural water system. The time scales are at least of the order of hours.

No	Aim	Strategy	Results
5	To study the comparative uptake of sulphate and manganese (Alligator Rivers Research Institute initiative)	A three hour injection was made into the Magela Creek near Ranger on 10 April 1986 comprising ^{35}S, ^{54}Mn as well as HTO, ^{99m}Tc and ^{198}Au to normalize for dilution/ dispersion of water, solute and suspended sediment respectively.	Downstream of the injection point, dilution and dispersion alone leads to a decrease in SO_4 concentration of about 50% every 4 km. In addition sorption phenomena led to a loss of 24 ± 6% of sulphate in the first 12 km. The $^{54}Mn/^{35}S$ ratio, after correction for sulphate sorption suggested that 85% of the first 12 km. As with Zn a fraction of the Mn may have been retained by sediment/ vegetation and re-released. Widespread distribution of ^{54}Mn was found on the bed of the Creek, although there was some penetration to depths of >500 mm.
6	To gauge the Magela Creek in flood using tritium dilution techniques.	Volume flow of the flooded Magela using tritium dilution were consistent with estimates from a string of gauges across a flood plain transect. New insights into the scaling of dispersion across broad flood plains were obtained.	1) The tracer dilution flow was 53 $m^3 s^{-1}$. Conventional gauging indicated that the flow rate fell from 90 to 45 $m^3 s^{-1}$ during the passage of the plume (10 to 21 March 79). 2) The measured flow rate corresponds to the time at which the first moment of the concentration profile is zero (14 Mar 79). At this point the flow rate was 70 $m^3 s^{-1}$. 3) The lateral dispersion coefficients are independent of the size of the pulse (10 m to 1 km). It was concluded that conditions of isotropic turbulence were reached on the shallow flood plain with a Prandtl mixing length determined by the depth. 4) The velocity of the tritium pulse relative to the wave front was retarded by a factor of 2 in acceptable agreement with a theoretical value of 1.67. Conclusion: The transport of non-interacting contaminants on shallow flood plains can be predicted from the velocity of the wave fronts and the dispersion coefficients close to the release point.

Notes (1): Dissolved ^{65}Zn passed through a 0.45μ filter.

The value of tracer studies to environmental protection relates to the additional insights which can be obtained which are of regulatory interest. They include the following:

- If heavy metals (Zn, Mn) are discharged into the Magela system, chemical equilibrium within the receiving waters, sediments and vegetation is achieved only after a number of hours;
- It is possible to separate decrease in concentration of contaminants in the water column into two categories: dilution/dispersion and adsorption onto sediments/ vegetation;
- It is possible to estimate the dispersion and dilution across a shallow flood plain from dispersivities measured close to the release point and the rate of migration of a wave front (caused e.g. by upstream rainfall) across the flood plain. Both parameters are relatively easy to acquire.

The time scales for distribution of heavy metals across the sediments and through the vegetation may be measured. Such studies may extend through a wet and a dry season if (as in the case of ^{65}Zn and ^{54}Mn) the half-lives are suitable. Such observations are of direct relevance to the long term assessment of the impact of a discharge.

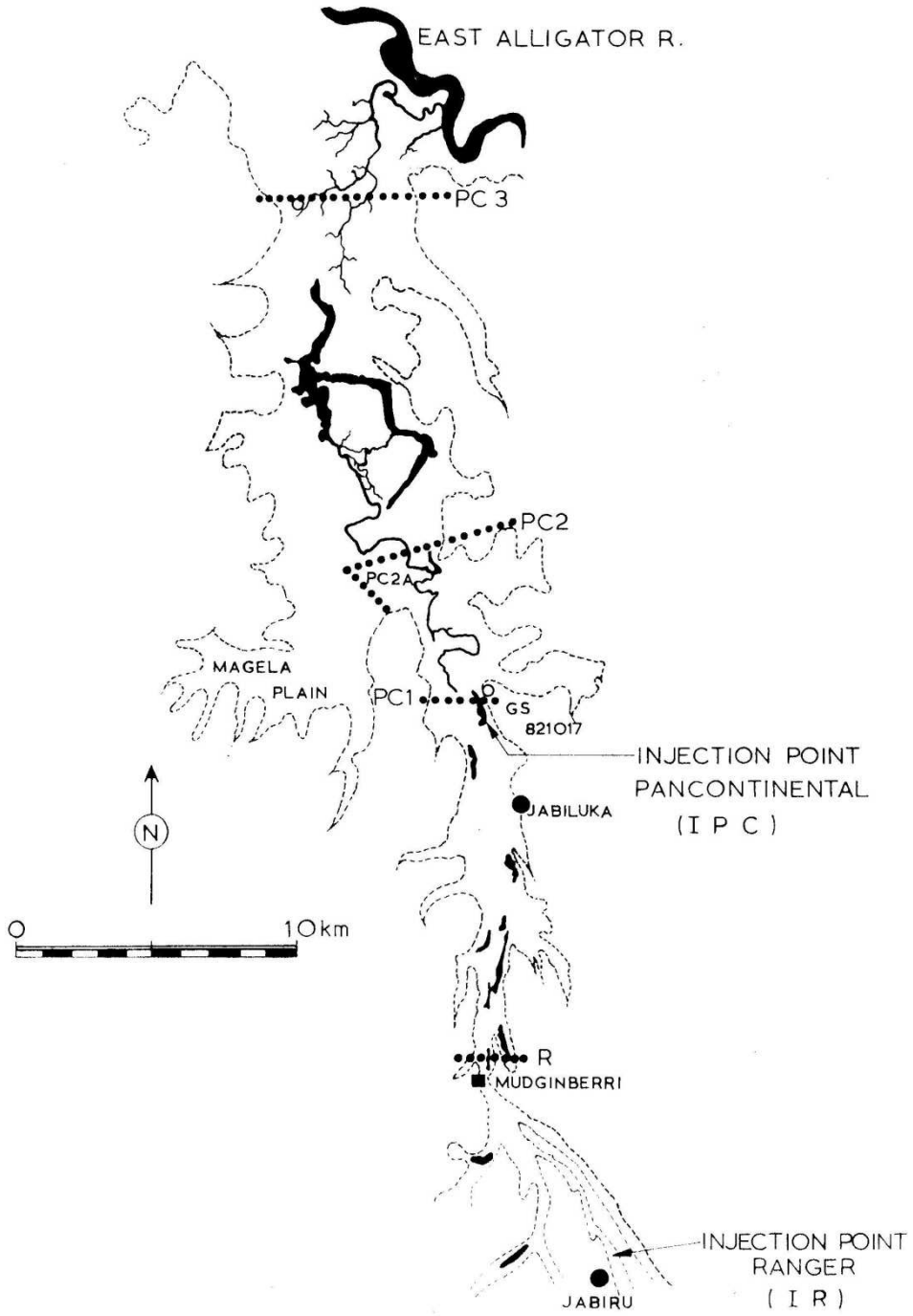


FIG.16. The Magela Flood plain region where the experiment was conducted.

2.2. Case Study 8. Heavy metal retention in stream-bed sediments

Exchange of water in streams between flowing (open) water mass and the underlying sediment is an important process determining the fate of contaminants. Pressure-driven advection, arising due to irregularities on the sediment surface, is believed to be an important hydraulic mechanism that causes the stream water to flow into and out of stream-beds [6]. In

addition to the filtration of water through bed sediment, reactive contaminants can be retained within the sediment due to adsorption on to particulate matter. To quantify the relative importance of retention in the hyporheic zone the Säva Stream Tracer Experiment 1998 was performed on 19th of May 1998 along a 30-km-long reach of the Säva Stream, Uppland County, Sweden. In the upper ~5 km of the studied reach, the surrounding land consisted of forest vegetation whereas, the lower part flows through agricultural land. The discharge was around 0.09 m^3/s at the injection point and 0.43 m^3/s at the measuring station furthest downstream. A simultaneous injection with a constant rate was performed during 5.3 h using the tracers tritium (74 GBq) and chromium-51 (92.5 GBq). During the injection, the chromium concentration (including the added non-radioactive Cr(III) carrier substance) in the stream water increased by 0.0001 nM.

Further, a transient storage model is evaluated using results from a tracer experiment, where a conservative and a reactive tracer (^3H and $^{51}Cr(III)$) were injected simultaneously and monitored in stream water and bed sediment. About 76% of the chromium was lost from the stream water on the reach 30 km downstream of the injection point directly after the passage of the pulse in the flowing water (Fig. 17). The bed sediment hosted the main part of the retained chromium. The time to washout 75% of the maximum solute uptake in the sediment was ~85 times longer for chromium than for tritium (i.e. ~45 days). This experiment revealed further that observations of chromium concentrations in the sediment were essential for the quantifying of sorption properties, as it was not possible to catch accurately the time scale of sorption within the duration of the breakthrough curves in the stream water. There was a clear need for a rate-limited description of the sorption of chromium in the sediment. We found that a first-order kinetic description of the sorption process could acceptably describe the breakthrough curves in both the stream water and the bed sediment.

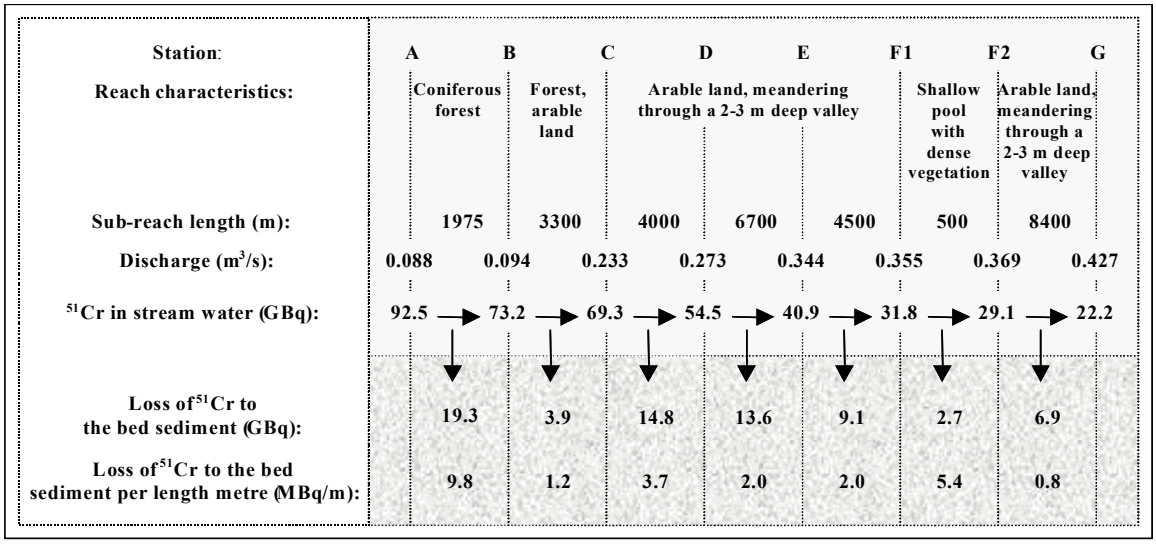


FIG. 17. Activity balance using the approach described in the paper titled "Tracer Data interpretation and modelling", from Wörman, included in this publication.

3. AQUATIC SEDIMENTS AND HYPORHEIC ZONE OF RIVERS

Aquatic sediments play a major role for accumulating contaminants and natural solute elements. The pools of carbon, phosphorus and nitrogen contained in the sediments of the oceans completely dominate the pools of the biogeochemical cycles. The sediment of the oceans act as 'end-stations' of solute elements, but aquatic sediments in general can be seen as a terminal accumulation on some relatively short time scale specific to the application of

interest. However, there is also a significant redistribution of solutes by erosion and hydro-mechanical exchange, which can be important both in rivers and estuaries. Such an accumulation and redistribution process is exemplified by the mud tracer labelling study in the Garonne – Gironde estuary and heavy metal transport in rivers. The retention in a fast flowing system like a river has often been considered to be 'inert' in the sense that soluble elements follow the flowing water and has a relatively short residence time in the river system. However, detailed tracer tests using specific adsorbing tracers and tritiated water indicate that the retention can significantly retard and even accumulate reactive tracers in aquatic sediments in flowing surface water. Other tracer studies with metals and radiocarbon have been performed in lakes to show the turnover processes as well as the accumulation in aquatic sediments [23, 24].

3.1. Case Study 9. Recycling of dredging product – The case of Zeebrugge Harbour

Previous research programs have shown that the global efficiency of dredging works is depending on the efficiency of the discharge on site, meaning by the fraction of particles staying on site a long time compared to the recycling fraction to the dredged areas. In-situ experiments using radiotracers have allowed to quantify this recycling. A first experiment performed on site 1 has allowed to better understand the recycling mechanism [6-12].

Studies have been performed through 3 different approaches:

- Morphological analysis to determine variations in deposited volumes;
- Determination of the bed characteristics through sampling and measurement of natural radioactivity;
- Tracer experiments.

The method requires to be able to distinguish the tracer from the natural background during some months. The main criteria to choose the tracers have been:

- The tracer has to label definitively the natural particles without alteration of their hydrodynamic properties;
- Its detection limit has to be very low;
- It must have no impact on environment;
- Its half-life has to be compatible with the duration of the study.

The selected tracers (^{181}Hf and^{160}Tb) are measured by low background gamma-ray spectrometry on natural sediment samples. Four large scale experiments have been performed. More than 200 samples have been taken on 66 stations along the Belgian coast and analyse during each experiment.

The program consists of 6 tracer injections over 3 years.

- Experiment n° 1: injection on site " ZB-Oost" (Hafnium, distance to the coast ± 2.8 km) and S2 (Terbium; distance to the coats ±12.5 km).

- Experiment n° 2: injection on northern part of the Akkaert sand bank (Fig. 18, Hafnium, distance to the coast ±21 km) and near the channel between S1 andS2 (Terbium, distance to the coast ± 15 km).

- Experiment n° 3: injection near Negenvaam (Hafnium, distance to the coast ± 17 km) and on the northern side of Thornton bank (Terbium, distance to the coast ± 30 km).

After injection, samples are taken versus time in the study area (between Terneuzen and Newport) and analysed.

Field experiments allow obtaining data on the dispersion of labelled sediments and on the temporal evolution of their concentration. They have shown that the particles migrate on the sea bed inside a low concentration layer of about 0.5 m thickness. These particles are deposited inside a narrow area (less than 10 km width) along the Belgian coast between Terneuzen and Newport.

These field experiments are thus a proof of particles recycling onto the coastal area and that the mud remains trapped inside a hydraulic vortex, at least for the duration of the experiment, about 4 months.

Tracers have been found in the harbours of Zeebruggen Blankenbergen Ostend, Newport, Breskens and Terneuzen but also inside the access channels to Zeebrugge Harbour (Scheur West, ScheurOost et Pas van het Zand).

These radiotracers recirculation experiments confirm the hypothesis of the sediment recirculation inside the area; this area is characterized by a low exchange rate of the sediments with the other part of the North Sea and a migration upstream inside the western Scheldt Estuary

The recycling process seems to be very fast. The arrival on the coast can take less than 2 days depending of the injection point and meteorological conditions. The concentration moves to a constant value versus time. The 2 tracers are recycled at different levels depending upon the position of the injection point.

The detected activity is slightly modified by dredging work but storms lead to a fast dilution of the tracers.

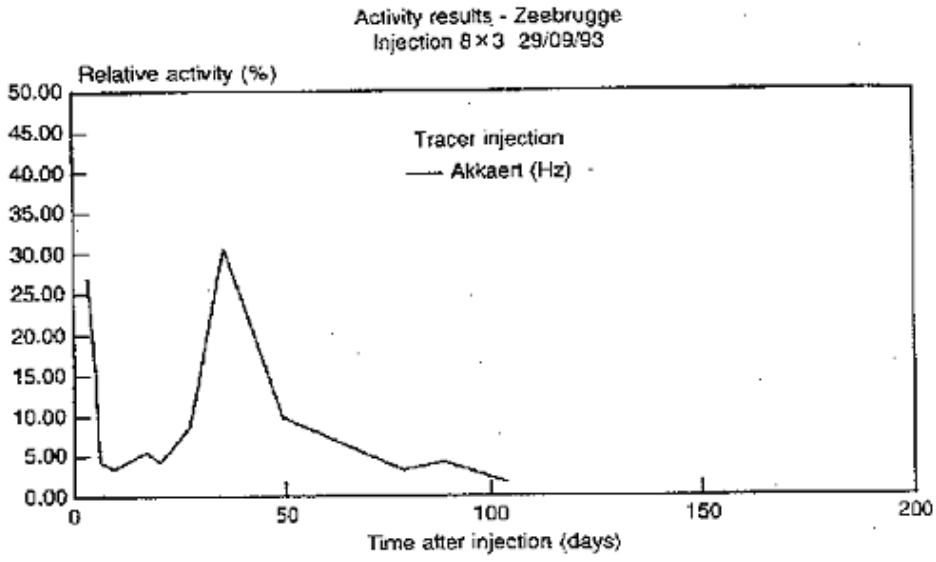


FIG. 18. Time evolution of relative activity at the harbour of Zeebrugge.

Fig. 19 shows the time evolution of tracer concentration near the Zeebrugge Harbour.

In Ostend Harbour the concentration observed during the second campaign is significantly lower than during the first one. That can be due to the distance of the injection point and thus to a repartition of the tracer on a bigger surface. These observations indicate that the system is near to be closed and that it is always the same sediment which is deposited, transported and recirculated both by natural processes and by dredging works. Both tracers can be found with the same rapidity in the study area. Even released far offshore they come back to the coast within about 2 days. The high concentrations observed 2 days after injection particularly inside the Scheur-Zand navigation channel show that the transport of the traced sediment takes place mainly through the channels onto the coast.

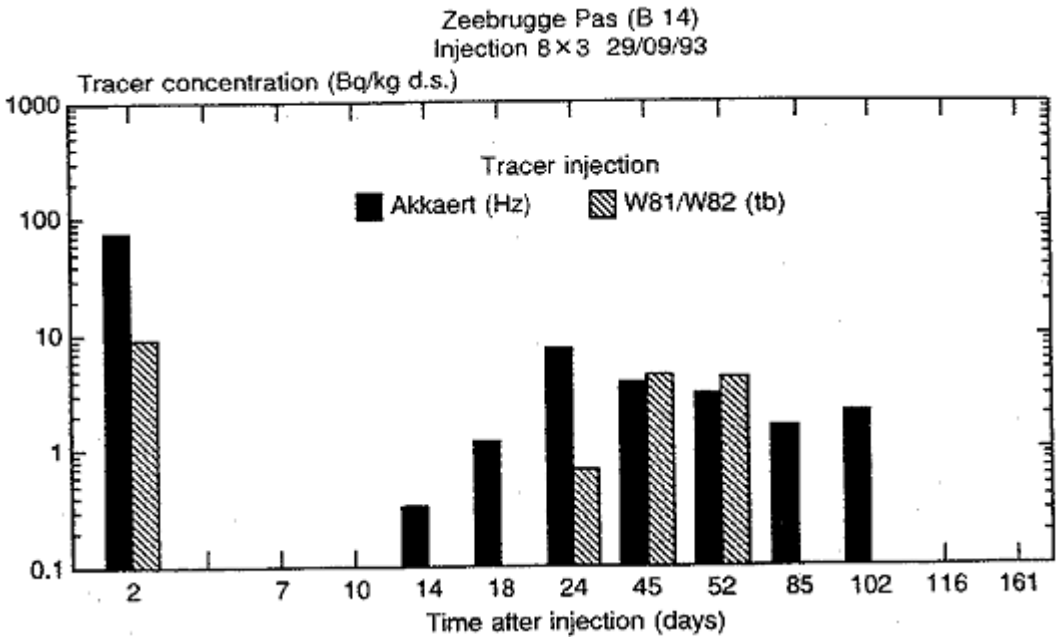


FIG.19. Time evolution of tracer concentration near the harbour of Zeebrugge.

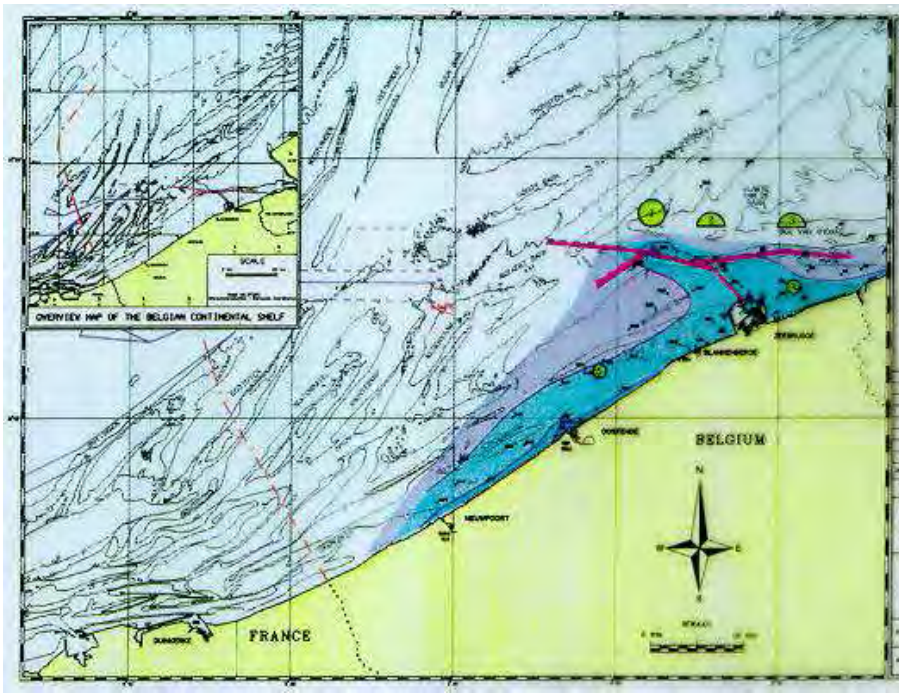


FIG. 20. Example of a tracer (Terbium) dispersion map. Dumping sites positions are figured in green

After this experiment can be concluded that the coastal zone appears as a trap for fine sediments. The dredging works products discharged inside this zone are recycled to the coast. A discharge outside this zone, more than 25 km offshore, is necessary to eliminate the recycling risk.

4. BIOLOGICAL UPTAKE OF CONTAMINANTS AND NUTRIENTS

Not only are tracers potentially adsorbed to sediments, they are also taken up by aquatic organisms and passed through the food chain, potentially to humans. Radiotracers have been used in both laboratory and field environments to study the rates of contaminant and nutrient uptake and depuration, to look at organ specific uptake and transfer through the food chain. While most of this work is conducted in laboratory settings, it is clear that the complexity of the natural environment cannot be replicated in a tank environment. The most extensive field program was a series of experiments conducted in the 1970s in the Canadian Experimental lakes [23-26]. These studies used a suite of radiotracers of water, nutrients and heavy metals to study mixing, adsorption to lake sediments and uptake by biota over the period of a year. The tracers used in the various studies included HTO, ^{54}Mn, ^{65}Zn, ^{133}Ba, ^{75}Se, ^{134}Cs, ^{48}V, ^{203}Hg, ^{59}Fe, ^{60}Co, $NaH^{14}CO_3$ and the compartments monitored included water, sediments, zooplankton, phytoplankton, macrophytes, invertebrates and fish. A number of studies have focussed on the behaviour of phosphorous using ^{32}P or ^{33}P [27]. Generally this type of study can only be conducted in closed or slow flowing water bodies as the time scale for uptake of the tracer needs to be shorter than the residence time of the tracer.

4.1. Case Study 10. Phosphate retention in a treatment wetland

4.1.1. Tracer tests in treatment wetlands

Treatment plant and treatment wetlands play an important role in reducing the nutrient content of recipient waters both for municipal waste water and runoff from agricultural land [15]. Particularly wetlands have the potential to offer sufficiently long residence times needed for reducing both the phosphorus and nitrogen load [28-30]. A crucial point in design model development is to access high quality data of the functional characteristics of wetlands. This is not always possible to achieve with standard monitoring data, due to the significant fluctuations in the in- and out-put loads of nutrients to the wetland and the internal circulation of nitrogen and phosphorus, which also varies temporally with changes in whether conditions as well as spatially. Tracer experiments, on the other hand, offer a possibility of artificial production of in- and output signals (of the tracers) that are more clearly defined and, therefore much easier to interpret in terms of model parameters. The experimental data can also reveal the mass fractions of nutrients that are removed in the wetland.

The overall objective of this paper is to present a simultaneous tracer experiment in Ekeby treatment wetland in Eskilstuna, Sweden, with tritiated water (^3H_2O), ^{15}N-enriched nitrate and ^{32}P-labelled phosphate. The use of both the conservative tracer and reactive tracers, ^{15}N and ^{32}P, facilitate a discrimination of the hydraulic processes and biogeochemical reactions. The wetland consists of several parallel series of treatment ponds and the test was carried out in one of the ponds. This paper describes mainly the results related to tritiated water and ^{32}P.

4.1.2. Ekeby treatment wetland

Ekeby wetland (Fig. 21) is a constructed wetland that receives the effluent water from the municipal sewage treatment plant in Eskilstuna 120 km west from Stockholm, Sweden. The

annual flow through the treatment plant and the wetland is 15,400,000 m^3 and serves 76 500 connected persons. The annual load to the wetland is 66.3 tons of total phosphorus, 420 tons of total nitrogen and the total open water area is about 30 hectares. Main vegetation in the wetland consists of emergent Reed Grass (*Glyceria maxima*) and submersed Canadian Pond Weed (*Elodera Canadensis*).

FIG. 21. Ekeby treatment wetland, Eskilstuna, Sweden. The wetland is excavated in natural clay deposits and used as a final 'polishing' step in the wastewater treatment plant in the municipality of Eskilstuna.

The wetland consists of 8 ponds. The treated wastewater from the treatment plant flows into a channel that distributes the water flow on five parallel ponds. Thereafter, the water flows into another distribution channel that separates the water on another three ponds. After the three downstream ponds the water ones again are collected in a channel before it flows into the Eskilstuna River. Eskilstuna River ends in Lake Mälaren, a lake that is connected with the Baltic Sea, via Stockholm.

4.1.3. Test procedure

On the 22nd November 2002 a tracer mixture was injected in the inlet pipe that connects the distribution channel with pond one in Ekeby wetland (Fig. 22). Pond one has a surface area of 2.6 hectares and is the first of the first five parallel ponds and is situated closest to the sewage treatment plant. The second inlet tube was closed during the injection and the discharge through the pond increased from 84 L/s at the time of the injection to 150 L/s on the 26th of November and then decreased to 84 L/s on the 10th of December. The mixture consisted of 18 GBq of ^{32}P, 3 kg ^{15}N (10Atom% KNO_3-^{15}N) and 74 GBq of HTO. ^{32}P was injected in ionic form (PO_4^{3-}) and ^{15}N as nitrate (NO_3^-). The injection was performed at a fairly constant rate during 5 h and 15 min.

Water samples were taken by means of auto-samplers at five points in the pond, at the inlet and outlet and at the three small islands that lie in a transversal line across the pond about 1/3 of the way from the inlet to the outlet. The last water sample was taken to weeks after the injection at the outlet. The analyses of ^{32}P and HTO were performed in a beta counter and corrected for natural background and radioactive decay.

After the tracer pulse had passed through the pond, samples from sediment and vegetation were collected. Sediment samples were taken from pond bed sediment before and after the small islands and vegetation samples from selected locations near two of the islands.

Filtration of water samples was performed in the laboratory for determination of the partitioning of particulate and dissolved (< 0.45 μm) fractions of ^{15}N and ^{32}P.

4.1.4. Results and discussion

From the breakthrough curves established at the three islands on about one third of the distance between inlet and outlet we can see a fairly well distribution of tracers on the flow pathways through the wetland (Fig. 22). The concentration in the central pathway is slightly lower than at the two others, but only moderately. From the breakthrough curve of tritiated water obtained at the outlet (Fig. 23), the mean residence time of water in pond 1 can be estimated to be about 3 to 3.5 days. This agrees well with the mean residence time estimated as the volume divided by the through flow, which results in about 3.2 days. Overall, these results indicate that the volume of water in the wetland is active in the through flow of wastewater, which is a prerequisite for a well functioning of the treatment process.

The total injected activity of ^{32}P was 18.0 GBq and about 13.7 GBq was recovered at the outlet during the investigation period ending 10 days and 16 hours after the start of the injection. This implies that 24% of the phosphate solution was removed in the November – December period in which the experiment was performed. An analysis of regular monitoring data shows that the annual removal rate of total phosphorus in the entire wetland for year 2002 (each flow line passes two ponds in series) is 57%.

Tentative results from the bed core samples indicate that a major fraction of the ^{32}P was removed by accumulation in the bed sediments. Probably, the most important mechanism for this removal is adsorption onto particulate matter and deposition as well as a certain exchange in dissolved phase directly with the bed sediment and adsorption onto the bed matrix. Analyses of vegetation material also show that a certain degree of ^{32}P accumulation onto vegetation and suspended solids attached to vegetation has occurred. On a short time-scale, the removal of ^{32}P can be seen as irreversible. A simple representation is to use a first order reaction for the removal rate from the water to the bed sediment under the water travelling through the wetland. The result in terms of the through flow of ^{32}P can be expressed as $M_{eff}/M_{in}=exp(-k<\tau>)$, where k is a rate coefficient [s^{-1}] and $<\tau>$ is the mean residence time (~3.2 days). Since, M_{eff}/M_{in}= 0.76, we find that k = 0.086 $days^{-1}$.

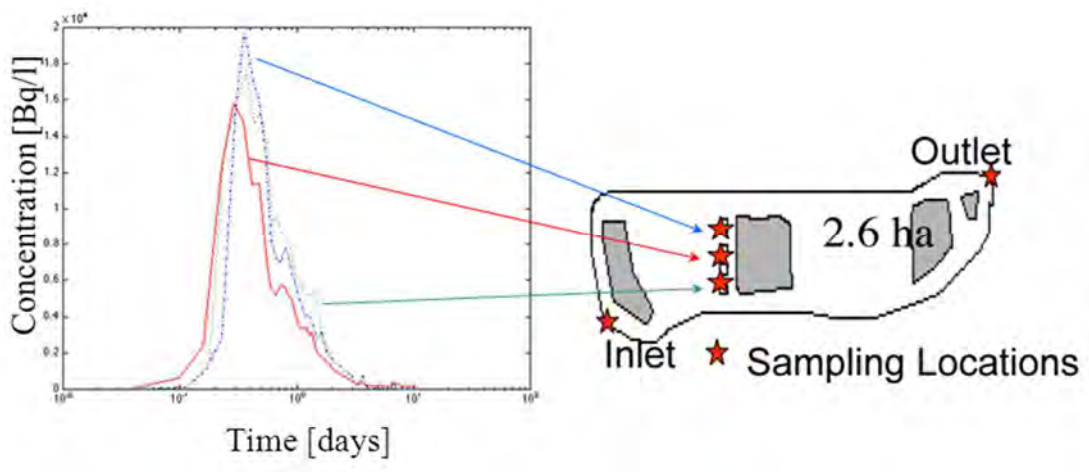


FIG. 22. Tritium breakthrough curves obtained at three islands inside the treatment wetland at Ekeby, Eskilstuna, Sweden.

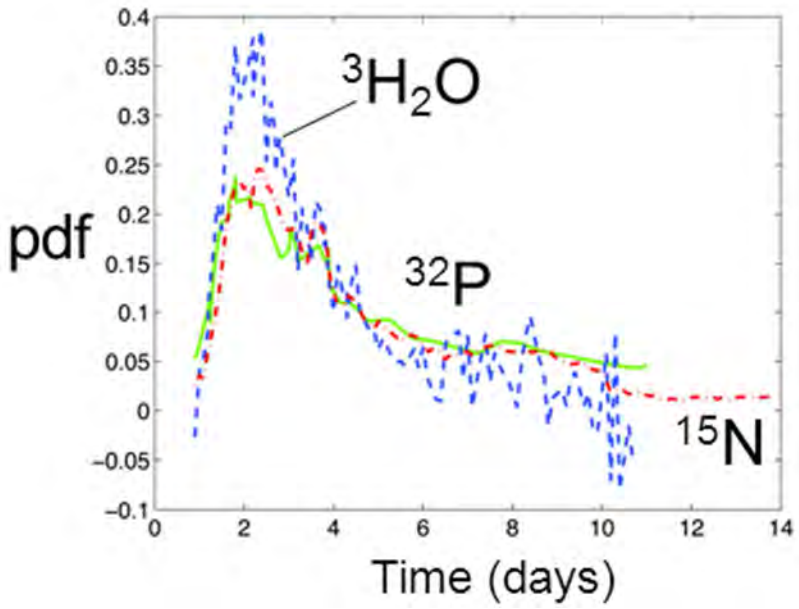


FIG. 23. Breakthrough curves of ^{32}P, ^{15}N and ^3H at the outlet of Ekeby treatment wetland, basin 1. These breakthrough curves have been normalized in a form of probability density functions. The tritium breakthrough curve can be interpreted as a transit time probability density function for water. The mean value is found to be 3.2 days with some uncertainty due to the exact background value.

REFERENCES

[1] RUTHERFORD, J.C., River Mixing, John Wiley, New York, USA (1994).

[2] BUTCHER, S.S., et al., Global Biogeochemical Cycles, Academic Press Limited, London (1992).

[3] SEO, I.W., BAEK, K.O., JEON, T.M., Analysis of transverse mixing in natural streams under slug tests, J. Hydraulic Res. **44** 3 (2006) 350–362.

[4] FISCHER, H.B., Dispersion Predictions in Natural Streams. J. Sanit. Eng. Div. ASCE **94** (SA5), (1968) 927-943.

[5] JUNG, S.H., et al, Measurement of a Dispersion Coefficient for a River with $^{99m}TcO_4^-$ as a Tracer, Journal of Nuclear Science and Technology (2008).

[6] SUH, K.S., et al., Determination of dispersion coefficients using radioisotope data in river environment, Applied Radiation and Isotopes (2009).

[7] MIGNIOT, C., Tassement et rhéologie des vases, La Houille Blanche 1/1989; 2/1989.

[8] DE MEYER, C.P., MALHERBE, B., Optimisation of maintenance dredging operations in maritime and estuarine areas, Terra et Aqua **35** (1987) 25- 40.

[9] BROSSARD, C., et al., Sécurité de la navigation dans les chenauxenvasés, Congrèsmondial, Association Internationale des ports et chenauxnavigables, section SQII.1, Osaka (1989).

[10] MEYER, G., et al., Utilisation des jaugesnucléaires de densitépourétudier et mesurer au laboratoire et in situ la concentration et la formation des dépôts de vase . Symposium Isotope Hydrology, IAEA, Vienna (1983).

[11] CAILLOT, A., Bed load transport. Guidebook on nuclear techniques in hydrology, (1983).

[12] TOLA F., CAILLOT A., COURTOIS G., Study of the evolution of dredged materials discharges by means of radioactive tracers . Coastal engineering Vol. 2, Houston, Texas (1984).

[13] BANDEIRA, J.V., Development of Nuclear and Correlate Techniques for Urban Hydrology Studies - Applications in Pampulha Hydrographic Basin and Velhas River,

MG. Doctorate Thesis in Sanitation, Environment and Hydric Resources – Federal University of Minas Gerais - UFMG, Belo Horizonte, Brazil. (2004)(In Portuguese).
[14] MALHERBE B., A case study of dumping of dredged material in open areas, Terra et Aqua **45** (1991).
[15] SINGH, S.K.; BECK, M.B. Dispersion coefficient of streams from tracer experimental data. Journal of Environmental Engineering **129** 6 (2003) 539-546.
[16] SAYRE, W.W., CHANG, F.M.,A laboratory investigation of the open channel dispersion process of dissolved, suspended and floating dispersants, US Geological Survey, Professional paper, 433E (1968).
[17] PACKMAN, A.I., BROOKS, N.H., MORGAN, J.J., A physicochemical model for colloid exchange between a stream and a sand streambed with bed forms, Water Resources Res. **36** 8 (2000) 2351-2361.
[18] THIBODEAUX, L.J., BOYLE, J.D., Bedform-generated convective-transport in bottom sediments, Nature **325** (1987) 341-343.
[19] ELLIOTT, A.H., Transport of solutes into and out of stream beds, Report No. KH-R-52, W.M. Keck Laboratory of Hydraulics and Water Resources, California Institute of Technology, USA (1990).
[20] WROBLICKY, G.J., et al., Seasonal variation in surface-subsurface water exchange and lateral hyporheic area of two stream-aquifer systems, Water Resources Research, **34** 3 (1998) 317-328.
[21] JONSSON, K., JOHANSSON, H., WÖRMAN, A., Hyporheic Exchange of Reactive and Conservative Solutes in Streams – Tracer Methodology and Model Interpretation, Journal of Hydrology **278** (2003) 153-171.
[22] MULHOLLAND, P.J., et al., Evidence that hyporheic zones increase heterotrophic metabolism and phosphorus uptake in forest streams. Limnology and Oceanography **42** 3 (1997) 443-451.
[23] HESSLEIN, R.H., BROECKER, W.S.,SCHINDLER, D.W., Fates of metal radiotracers added to a whole lake: sediment-water interactions. Can, J. Fish. Aquat. Sci. **37** (1980a) 378-386.
[24] HESSLEIN, R.H., BROECKER, W.S., QUAY, P.D., Whole-lake radiocarbon experiment in an oligotrophic lake at the Experimental Lakes Area, north-western Ontario, Can. J. Fish. Aquat. Sci. **37** (1980b) 454-463.
[25] KLAVERKAMP, J.F., et al., Fates of metal radiotracers added to a whole lake: accumulation in slimy sculpin (Cottuscognatus) and white sucker (Catstomuscommersoni), Sci Tot Env. **28** (1993) 119-128.
[26] QUAY, P.D., et al., Vertical diffusion rates determined by tritium tracer experiments in the thermocline and hypolimnion of two lakes, Limnology and Oceanography **25** 2 (1980) 201-218.
[27] KJELLIN, J.,WÖRMAN, A., JOHANSSON, H., LINDAHL, A., Controlling factors for water residence time and flow pattern in Ekeby treatment wetland, Sweden, Advances in Water Resources **30** (2007) 838-850.
[28] KADLEC, R. H., KNIGHT, R. L., Treatment Wetlands, CRC Press LLC, USA (1996).
[29] PELL, M., WÖRMAN, A., Biological Wastewater Treatment Systems. In Encyclopedia of Ecology, Ed. Brian Kronan, Elsevier, The Netherlands (2008).
[30] KADLEC, R.H., Chemical, Physical and Biological Cycles in Treatment Wetlands, Wat. Sci. Tech. **40** 3 (1999) 37-44.

THE FUTURE OF RADIOTRACING

P.L. AIREY, C.E. HUGHES

Australian Nuclear Science and Technology Organisation, ANSTO, Australia

Abstract

Radiotracing has enormous potential to provide data to underpin environmental management in aquatic ecosystems. The use of tracer data to validate numerical models, improvements in data acquisition and new tracer forms provide the basis for the future of radiotracing. However, an increasing regulatory burden, reduction in tracer availability and a loss of technical expertise threaten this field of study. Standardisation of tracer approaches where possible and new work on assessing the impact of radiotracers on non-human biota are two areas that may enable the future use of radiotracers at a field scale in engineering and research applications.

1. INTRODUCTION

Pressures on aqueous ecosystems including wetlands, riverine, lacustrine and coastal systems will continue to increase. Natural population growth exacerbated by the movement of peoples will contribute to the growing demands for food and fresh water from largely degraded environmental resources. The consequences of these pressures can only be mitigated over the long term by sophisticated ecosystem management. Underpinning good management is a clear understanding of the flow and contaminant dynamics, hydro-geochemistry as well as the biology of these complex systems.

The tracer approach is exceptionally powerful as it enables the separate study of individual components of complex systems as well as the interaction between compartments. The range of tracers is such that a good match between physico-chemical properties of the component of interest and the tracer is often possible. For instance, tracers can be chosen to study the movement of bulk water, of suspended sediment, of sands and silts of heavy metal, nutrient or hydrocarbon contaminants. Examples are provided in this paper.

2. APPLICATION OF TRACERS TO THE EVALUATION OF MATHEMATICAL MODELS

Tracer studies are expensive to implement and only provide direct information on limited temporal and spatial domains around the injection point. Further, their predictive power is limited to the prevailing conditions at injection.

These limitations do not apply to distributed mathematical models of the environmental systems. There has been an enormous increase in the availability of these models in recent times and this trend is set to continue.

This has led to a paradigm shift in the role of tracer studies over the past years. Tracer studies are now primarily designed to evaluate and refine predictive models. The refined models are more robust and can be used with greater confidence in making the types of predictions on which good management decisions are based.

The best studies involve a close interaction between the tracer practitioner and the mathematical modeller. Preliminary models of a study area can be made and their outputs used to define the most sensitive areas for the tracer study.

Relatively speaking the cost of mathematical modelling is decreasing and will continue to do so. However, it is extremely important that decision makers reliant on model output are not too far removed from nature. If parameters such as dispersivity, mixing, sediment exchange or

bed load transport are important, some level of model validation based on tracing is often recommended.

3. STANDARDIZATION OF RADIOTRACER APPROACH

Radiotracer studies are highly regulated; indeed the cost of regulation is a major factor inhibiting their widespread use. This issue can be addressed in a number of ways:

- *Enhanced understanding of the impact radiotracers on the environment:* The impact of radiation dose on humans has been widely studied and is directly transferable to tracer studies. Less well understood is the impact of radiation on elements of the ecosystems. Presentation of the current understanding and results of specific studies in the field will provide regulators with the confidence needed to make judgments on any impact of the release of radiotracers to the environment.
- *Standardization of procedure:* The increasing difficulty of sourcing a wide range of isotopes for tracer studies has been noted. Radiotracer practitioners often use materials such as ^{99m}Tc designed primarily for nuclear medical applications. On the positive side, advantage can be taken of the major investment in the manufacture of the generator and the field dispensing systems. Regulators often feel more comfortable in assessing experimental procedures which are closely analogous to those routinely used in nuclear medicine.

4. ALTERNATIVES TO RADIOTRACERS

Generally speaking, alternatives to radiotracers tend to be used where possible. In the case of water tracers, fluorescent tracers are most commonly used. In times gone by, aqueous (gamma emitting) radiotracers had the enormous advantage that could be measured in situ by detectors deployed overboard from monitoring vessels. Now-a-days, easily available pulsed laser fluorimeters can also be deployed for real time monitoring. Generally speaking the limit sensitivities of radio- and fluoro-tracers are comparable for deployed monitors. Fluorescent tracers have the advantage that they are not subject to counting statistics uncertainty or enhanced backgrounds near the bed of the water bodies and they can be seen to assist in tracking in surface waters. However, unlike radiotracers, they are subject to variable background in polluted water and to the leakage of light to the fluorimeter near the water surface. In the past the use of fluorescent tracers in the environment was not subject to significant regulatory control which provided a practical advantage to their use. However, tightening of environmental controls in many jurisdictions means that their use now often requires a similar level of risk assessment as the use of radiotracers. In some applications fluorescent tracers may be considered more toxic to the environment than radiotracers.

Radiotracers have an enormous advantage in the study of sand and particulate transport, particularly if the movements are slow and studies are needed over weeks or months. Whilst fluorescent tracers have been used to trace sand transport, they cannot be detected in-situ when buried, and detection by sampling is inefficient and requires larger tracer volumes. Magnetic sand tracers have been developed, however, this techniques is not yet well established.

A wide range of chemical and activatable tracers are available including the relatively recent development of tracers with DNA markers. However, sampling is generally required and this adds significant costs to the investigation whilst reducing the spatial and temporal resolution of the data.

5. NEW TECHNOLOGIES

As indicated in the whole publication, technologies are advancing and many are directly applicable to radiotracers. They include:

- DGPS and depth sensor positioning of the detectors in three dimensions;
- Real time data accumulation and processing;
- The development of new detectors especially ambient temperature probes with spectral capabilities;

The development of new tracer forms such as nano particle tracers is discussed in the paper titled “Radiotracer methodology”, from Brisset et al., included in this publication. Radiotracing is a mature science and hence incremental improvements only can be expected. Its future will depend on defining applications in which the radiotracer approach has an overwhelming advantage given the regulatory hurdles which will remain.

LIST OF PARTICIPANTS AND CONTRIBUTORS

Airey, P.L.	Australian Nuclear Science and Technology Organization, ANSTO, Australia
Bandeira, J.V.	Brazilian Nuclear Energy Commission, CNEN/CDTN, Brazil
Brisset, P.	French Alternative Energies and Atomic Energy Commission, CEA, France
Hughes, C.E.	Australian Nuclear Science and Technology Organization, ANSTO, Australia
Jung, S.H.	Korea Atomic Energy Research Institute, KAERI, Republic of Korea
Martin, P.	International Atomic Energy Agency
Sansone, U.	International Atomic Energy Agency
Wörman, A.	Royal Institute of Technology, KTH, Sweden

Consultants Meetings

Vienna, Austria; 20 - 24 Jun 2009; 22 - 26 Feb 2010

IAEA
International Atomic Energy Agency

No. 23

ORDERING LOCALLY

In the following countries, IAEA priced publications may be purchased from the sources listed below or from major local booksellers.

Orders for unpriced publications should be made directly to the IAEA. The contact details are given at the end of this list.

AUSTRALIA
DA Information Services
648 Whitehorse Road, Mitcham, VIC 3132, AUSTRALIA
Telephone: +61 3 9210 7777 • Fax: +61 3 9210 7788
Email: books@dadirect.com.au • Web site: http://www.dadirect.com.au

BELGIUM
Jean de Lannoy
Avenue du Roi 202, 1190 Brussels, BELGIUM
Telephone: +32 2 5384 308 • Fax: +32 2 5380 841
Email: jean.de.lannoy@euronet.be • Web site: http://www.jean-de-lannoy.be

CANADA
Renouf Publishing Co. Ltd.
5369 Canotek Road, Ottawa, ON K1J 9J3, CANADA
Telephone: +1 613 745 2665 • Fax: +1 643 745 7660
Email: order@renoufbooks.com • Web site: http://www.renoufbooks.com

Bernan Associates
4501 Forbes Blvd., Suite 200, Lanham, MD 20706-4391, USA
Telephone: +1 800 865 3457 • Fax: +1 800 865 3450
Email: orders@bernan.com • Web site: http://www.bernan.com

CZECH REPUBLIC
Suweco CZ, spol. S.r.o.
Klecakova 347, 180 21 Prague 9, CZECH REPUBLIC
Telephone: +420 242 459 202 • Fax: +420 242 459 203
Email: nakup@suweco.cz • Web site: http://www.suweco.cz

FINLAND
Akateeminen Kirjakauppa
PO Box 128 (Keskuskatu 1), 00101 Helsinki, FINLAND
Telephone: +358 9 121 41 • Fax: +358 9 121 4450
Email: akatilaus@akateeminen.com • Web site: http://www.akateeminen.com

FRANCE
Form-Edit
5 rue Janssen, PO Box 25, 75921 Paris CEDEX, FRANCE
Telephone: +33 1 42 01 49 49 • Fax: +33 1 42 01 90 90
Email: fabien.boucard@formedit.fr • Web site: http://www.formedit.fr

Lavoisier SAS
14 rue de Provigny, 94236 Cachan CEDEX, FRANCE
Telephone: +33 1 47 40 67 00 • Fax: +33 1 47 40 67 02
Email: livres@lavoisier.fr • Web site: http://www.lavoisier.fr

L'Appel du livre
99 rue de Charonne, 75011 Paris, FRANCE
Telephone: +33 1 43 07 50 80 • Fax: +33 1 43 07 50 80
Email: livres@appeldulivre.fr • Web site: http://www.appeldulivre.fr

GERMANY
Goethe Buchhandlung Teubig GmbH
Schweitzer Fachinformationen
Willstätterstrasse 15, 40549 Düsseldorf, GERMANY
Telephone: +49 (0) 211 49 8740 • Fax: +49 (0) 211 49 87428
Email: s.dehaan@schweitzer-online.de • Web site: http://www.goethebuch.de

HUNGARY
Librotade Ltd., Book Import
PF 126, 1656 Budapest, HUNGARY
Telephone: +36 1 257 7777 • Fax: +36 1 257 7472
Email: books@librotade.hu • Web site: http://www.librotade.hu

INDIA
Allied Publishers
1st Floor, Dubash House, 15, J.N. Heredi Marg, Ballard Estate, Mumbai 400001, INDIA
Telephone: +91 22 2261 7926/27 • Fax: +91 22 2261 7928
Email: alliedpl@vsnl.com • Web site: http://www.alliedpublishers.com

Bookwell
3/79 Nirankari, Delhi 110009, INDIA
Telephone: +91 11 2760 1283/4536
Email: bkwell@nde.vsnl.net.in • Web site: http://www.bookwellindia.com

ITALY
Libreria Scientifica "AEIOU"
Via Vincenzo Maria Coronelli 6, 20146 Milan, ITALY
Telephone: +39 02 48 95 45 52 • Fax: +39 02 48 95 45 48
Email: info@libreriaaeiou.eu • Web site: http://www.libreriaaeiou.eu

JAPAN
Maruzen Co., Ltd.
1-9-18 Kaigan, Minato-ku, Tokyo 105-0022, JAPAN
Telephone: +81 3 6367 6047 • Fax: +81 3 6367 6160
Email: journal@maruzen.co.jp • Web site: http://maruzen.co.jp

NETHERLANDS
Martinus Nijhoff International
Koraalrood 50, Postbus 1853, 2700 CZ Zoetermeer, NETHERLANDS
Telephone: +31 793 684 400 • Fax: +31 793 615 698
Email: info@nijhoff.nl • Web site: http://www.nijhoff.nl

Swets Information Services Ltd.
PO Box 26, 2300 AA Leiden
Dellaertweg 9b, 2316 WZ Leiden, NETHERLANDS
Telephone: +31 88 4679 387 • Fax: +31 88 4679 388
Email: tbeysens@nl.swets.com • Web site: http://www.swets.com

SLOVENIA
Cankarjeva Zalozba dd
Kopitarjeva 2, 1515 Ljubljana, SLOVENIA
Telephone: +386 1 432 31 44 • Fax: +386 1 230 14 35
Email: import.books@cankarjeva-z.si • Web site: http://www.mladinska.com/cankarjeva_zalozba

SPAIN
Diaz de Santos, S.A.
Librerias Bookshop • Departamento de pedidos
Calle Albasanz 2, esquina Hermanos Garcia Noblejas 21, 28037 Madrid, SPAIN
Telephone: +34 917 43 48 90 • Fax: +34 917 43 4023
Email: compras@diazdesantos.es • Web site: http://www.diazdesantos.es

UNITED KINGDOM
The Stationery Office Ltd. (TSO)
PO Box 29, Norwich, Norfolk, NR3 1PD, UNITED KINGDOM
Telephone: +44 870 600 5552
Email (orders): books.orders@tso.co.uk • (enquiries): book.enquiries@tso.co.uk • Web site: http://www.tso.co.uk

UNITED STATES OF AMERICA
Bernan Associates
4501 Forbes Blvd., Suite 200, Lanham, MD 20706-4391, USA
Telephone: +1 800 865 3457 • Fax: +1 800 865 3450
Email: orders@bernan.com • Web site: http://www.bernan.com

Renouf Publishing Co. Ltd.
812 Proctor Avenue, Ogdensburg, NY 13669, USA
Telephone: +1 888 551 7470 • Fax: +1 888 551 7471
Email: orders@renoufbooks.com • Web site: http://www.renoufbooks.com

United Nations
300 East 42nd Street, IN-919J, New York, NY 1001, USA
Telephone: +1 212 963 8302 • Fax: 1 212 963 3489
Email: publications@un.org • Web site: http://www.unp.un.org

Orders for both priced and unpriced publications may be addressed directly to:

IAEA Publishing Section, Marketing and Sales Unit, International Atomic Energy Agency
Vienna International Centre, PO Box 100, 1400 Vienna, Austria
Telephone: +43 1 2600 22529 or 22488 • Fax: +43 1 2600 29302
Email: sales.publications@iaea.org • Web site: http://www.iaea.org/books

International Atomic Energy Agency
Vienna
ISBN 978–92–0–100415–4
ISSN 1011–4289