

12/2002

18-
GC
7.1
K46
2002

**The Distribution and History of Nuclear Weapons Related Contamination
in Sediments from the Ob River, Siberia as Determined by Isotopic Ratios of
Plutonium, Neptunium, and Cesium**

by

Timothy Cope Kenna

B.A., Vassar College, 1988

Submitted in partial fulfillment of the requirements for the degree of

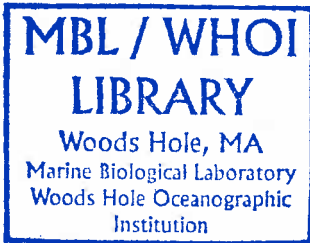
Doctor of Philosophy

at the

WOODS HOLE OCEANOGRAPHIC INSTITUTION

and the

MASSACHUSETTS INSTITUTE OF TECHNOLOGY



February 2002

© 2002 Timothy C. Kenna
All rights reserved

The author hereby grants MIT and WHOI permission to reproduce paper and electronic copies of this thesis in whole or in part and to distribute them publicly.

Signature of Author

Joint Program in Oceanography
Massachusetts Institute of Technology
and Woods Hole Oceanographic Institution

Certified by

Dr. Frederick L. Sayles
Thesis Supervisor

Accepted by

Dr. Margaret K. Tivey
Chair, Joint Committee for Chemical Oceanography
Woods Hole Oceanographic Institution

WHT

The Distribution and History of Nuclear Weapons Related Contamination in Sediments from the Ob River, Siberia as Determined by Isotopic Ratios of Plutonium, Neptunium, and Cesium

by
Timothy Cope Kenna

Abstract

This thesis addresses the sources and transport of nuclear weapons related contamination in the Ob River region, Siberia. In addition to being one of the largest rivers flowing into the Arctic Ocean, the bulk of the former Soviet Union's nuclear fuel reprocessing and weapons testing facilities (i.e. Mayak, Tomsk-7, and Semipalitinsk) are located within the Ob drainage basin. The atom ratios $^{240}\text{Pu}/^{239}\text{Pu}$, $^{237}\text{Np}/^{239}\text{Pu}$, and $^{137}\text{Cs}/^{240}\text{Pu}$, measured by magnetic-sector ICP-MS, are used to distinguish between contamination derived from global fallout and contamination derived from local sources. Deposition chronologies estimated for sediment cores are used to construct a record of weapons related contamination at the sites sampled. Contaminant records indicate that in addition to debris from atmospheric weapons tests, materials derived from local sources have also played a role in nuclear weapons related contamination of the Ob region. Isotopic data presented in this study clearly demonstrate that non-fallout contamination has been transported the full length of the Tobol, Irtysh, and Ob Rivers (i.e. the tributaries draining Mayak, Semipalitinsk, and Tomsk-7, respectively).

In several instances, unique isotopic compositions are observed in sediments collected from tributaries draining each of the suspected non-fallout sources. In such cases, these materials and their deposition ages have been used to link contamination in the Ob delta to Mayak, Tomsk-7, or Semipalitinsk. Linear transport rate estimates (km yr^{-1}) indicate that contaminated sediments transit between source tributaries and the Ob delta on time-scales of ≤ 1 year. These estimates suggest that a catastrophic release of contamination due to dam failure at one of the many reservoirs located at both Mayak and Tomsk-7 that contain high levels of radioactive waste would result in measurable levels of contamination in the delta within as little as 1 year.

Isotopic concentrations in sequentially extracted sediments containing weapons related contamination reveal that the majority of plutonium and neptunium (80 to 90

percent) behaves in a similar fashion regardless of the source and is removed by treating the sediments with citrate-dithionite. This indicates that plutonium and neptunium are not truly refractory and likely associate with redox sensitive sedimentary components. Isotopic ratios measured in extracted fractions suggest that only a minor fraction of contamination is associated with acid leachable or acid digestible sedimentary phases.

Thesis Supervisor: Frederick L. Sayles, Scientist Emeritus

Acknowledgements

Conducting high quality scientific research and writing a doctoral dissertation is not a solitary endeavor; it requires collaboration of the finest kind. Six years ago Fred Sayles took a chance on me; thanks Fred. During our time together, I have learned much and relied heavily on your advice and guidance; you have my respect and deepest gratitude. I would also like to thank my committee members, Bill Jenkins, Greg Ravizza, Hugh Livingston, John Edmond, and Harry Hemond for their insight, encouragement, and patience as I went through this process. Special thanks to Bill Jenkins for traveling across the Atlantic to be at my defense and to Harry Hemond for stepping in on rather short notice. Thanks also to Ken Buesseler not only for chairing my defense, but also for taking the time to talk with me when I would wander into his office and making available to me both laboratory facilities and reference materials.

This work would not have been possible without the efforts of Gera Panteleyev and Steven Smith who were responsible for logistics and sample collection in the Ob. It is a sad note that a tragic accident on the Ob River in the summer of 1995 took Gera's life; he is sorely missed, and I would especially like to acknowledge his contribution to this research.

I also want to acknowledge and greatly appreciate the assistance of Joanne Goudreau for setting up and over-seeing many of the sample processing and radiochemical procedures as well as putting up with my tendency to sprawl and colonize any available space in the lab.

Scot Birdwhistell was also very helpful with sample processing and method development.

Outside of the Sayles group, there are numerous scientists and technical staff, who have been of great assistance. I want to say thanks to all those who have taken the time to listen and give advice over the years. Mark Kurz, who hired me as a research assistant in 1991, and is largely responsible for my interest in isotope geochemistry. He continues to act as both friend and confidant. Dave Glover has been a constant source of information and assistance with modeling and statistical analyses and never seemed to mind taking time to help me. Special thanks to Lary Ball, the ICP-MS master, for his patient assistance, sense of humor, and allowing me to independently operate the Element. Kathleen Ruttenberg was extremely helpful with advice and information concerning selective extraction. Conversations with Ed Sholkovitz, Eric Hintsa, John Andrews, Steve Pike, Steve Manganini and Pat Lohman were also very helpful. Special thanks to both Dempsey Lott and Josh Curtis for all their technical assistance over the years. They may notice that tools and supplies last a bit longer after I am gone. I also greatly appreciate the assistance of Sheila Clifford, Joanna Ireland, Susan Casso, and Donna Mortimer in the Department of Marine Chemistry and Geochemistry as well as Julia Westwater, Marsha Bissonette in the WHOI Education Department.

Outside of WHOI, I want to acknowledge Sherrod Maxwell, Wes Efurud, Tom Beasley, and Jim Kelley for their help with method development and measurements.

I would also like to thank the many friends that I have made over the years. Fellow students, especially Mak Saito, Carrie Tuit, Payel Parekh, Allegra Hosford, Jen

Georgian, Robert Ackert, and Kirsten Laarkamp. All of my friends at the Sea Education Association, Penikese Island School, Church of the Messiah, and the Upper Cape Cod Tae Kwon Do Club. I especially want to acknowledge the friendship of Bill and Dori Mebane and thank them for all of the good times and adventures. I shall miss you all terribly.

But most importantly, I want to thank my family; my mother and father and especially my wife Laura and daughter Katie. Without their constant love, encouragement, and support, I never would have made it.

Funding for this research was provided by the Office of Naval Research under Grants N00014-93-1-1139, and N00014-1-95), and the National Science Foundation under Grant EAR-98-07590.

Table of Contents

Title Page.....	1
Abstract.....	3
Acknowledgements.....	5
Table of Contents.....	7
List of Figures.....	12
List of Tables.....	18
<u>Chapter 1. Nuclear weapons related contamination in the Ob River: Introduction, Background, Previous work, and Objectives</u>	21
The Ob River and its Drainage Basin.....	22
Sources of Radioactivity to the Ob River.....	23
Previous research on the Ob River.....	24
Research Objectives.....	26
References: Chapter 1.....	29
<u>Chapter 2. Sources of Radioactive Contamination to the Ob Watershed</u>	31
Radioactive Fallout from Nuclear Weapons Tests.....	31
Weapons Test Sites.....	35
Weapons Production and Nuclear Fuel Reprocessing Facilities.....	40
Nuclear Accident at Chernobyl.....	44
Isotopic Composition of Different contaminant Sources.....	45
End-member Isotopic Values used in this Study.....	56
References: Chapter 2.....	60

<u>Chapter 3. Characteristics of the Ob River System</u>	65
Drainage Basin.....	65
Hydrologic Regime and Sedimentation.....	65
Ice Formation.....	66
Marine/Freshwater Boundary.....	67
Water and Suspended Sediment Discharge.....	67
Irtys River.....	71
Upper Ob River.....	74
Ob/Irtys Confluence and Ob delta.....	76
References: Chapter 3.....	83
<u>Chapter 4. Sampling, Analytical Methods, and Data</u>	87
Sample Collection.....	87
Sample Analyses.....	90
Isotope dilution and yield monitors.....	91
Instrumentation and mass spectrometer analysis method.....	91
Isotope ratio and concentration calculations and uncertainty estimates.....	99
Laboratory Intercomparison Analyses.....	100
References: Chapter 4.....	111
<u>Chapter 5. Radionuclide Distributions, Sedimentation Rates, and Core Chronologies</u>	113
Radionuclide Distributions.....	113

Sedimentation Rates and Core Chronologies.....	127
Dating by the $^{210}\text{Pb}_{\text{xs}}$ Method.....	128
Dating by the Radionuclide Horizon Method.....	133
Initial Appearance of Weapons Related Contamination.....	134
Global Fallout Maximum.....	137
Chernobyl.....	141
Sedimentation Rates.....	151
Timescales.....	155
References: Chapter 5.....	156
<u>Chapter 6. Contaminant Records</u>	159
Characterization of Nuclear Weapons Related Contamination.....	159
Tobol River.....	160
Irtys River.....	164
Lower Irtys River.....	168
Upper Ob River.....	170
Ob Delta.....	174
Taz Estuary.....	183
Contaminant Record Summary.....	186
Atom Ratio Mixing Plots.....	187
Tobol and Irtys Rivers.....	188
Lower Irtys River, Upper Ob River, and Ob Delta from 1986 to 1995.....	193

Tobol River, Irtysh River, Upper Ob River and Ob delta prior to the mid-1980s.....	197
Summary Chapter Six.....	200
Chapter: 6 References.....	202

Chapter 7. The Distribution of ²³⁹Pu, ²⁴⁰Pu, and ²³⁷Np among Chemically Defined components of Sediments from the Ob River..... 205

Overview of Selective Leach Fractions and Experimental Design.....	207
Results and Discussion.....	215
Distributions of ²³⁹ Pu, ²⁴⁰ Pu, and ²³⁷ Np among Fractions.....	216
Comparison of ²³⁹ Pu, ²⁴⁰ Pu, and ²³⁷ Np measured in Bulk Sediments and Fraction Totals.....	227
Comparison of Pu and Np Isotopic Composition of Bulk Sediments and Leached Fractions.....	228
Fractionation.....	233
Conclusions.....	241
References: Chapter 7.....	242

Chapter 8. Contaminated Sediment Transport Rates and Contribution Estimates from Suspected Source Tributaries..... 245

Contaminated Sediment Transport Rates.....	245
Contaminated tributary sediment vs. global fallout contributions	258
Summary.....	265
References: Chapter 8.....	267

Chapter 9. Conclusions.....269
Future Work.....271
Appendix I. Sample Data Tables.....273
Appendix II. Radionuclide Inventories.....365

List of Figures

Chapter 1. Nuclear weapons related contamination in the Ob River: Introduction, Background, Previous work, and Objectives

Figure 1:1. Map showing Ob River drainage basin with sources of weapons related contamination and study region.....20

Chapter 2. Sources of Radioactive Contamination to the Ob Watershed

Figure 2:1. Summary of Nuclear Weapons tests by the 5 major Nuclear Powers..... 30

Figure 2:2. Latitudinal distribution of ⁹⁰Sr Fallout Measured for the Period 1958 to 1967.....32

Figure 2:3 Quarterly Deposition of ⁹⁰Sr Measured at New York City between 1954 and 1970.....34

Figure 2:4. Percent of Total Activity initially injected into the Troposphere as a Function of Total Yield for Air Bursts in the Tropical Atmosphere.....34

Figure 2:5. Summary of FSU Tests at Semipalitinsk and Novaya Zemlya.....36

Figure 2:6. Summary of USA Aboveground Tests Conducted at Low Latitude Pacific Sites.....38

Figure 2:7. Summary of PRC aboveground tests conducted at Lop Nor.....38

Figure 2:8. The Range of Isotopic Composition of Different Contaminant Sources.....57

Chapter 3. Characteristics of the Ob River System

Figure 3:1. Mean Monthly Water and Sediment Discharge observed at Salekhard Gauging Station from 1960 to 1987.....64

Figure 3:2. Map of Ob River Region with Hydrographic Stations and Potential Sources of Weapons Related Contamination.....69

Figure 3:3. Mean Annual Water and Sediment Discharge for Locations along the Iset, Tobol, and Irtysh River above its Confluence with the Ob River.....73

Figure 3:4. Mean Annual Water and Sediment Discharge for Locations along the Ob River above its Confluence with the Irtysh River.....	75
Figure 3:5. Mean Annual Water and Sediment Discharge for Locations along the Ob River Below its Confluence with the Irtysh River.....	77
Figure 3:6. Annual Water and Sediment Discharge Estimated for Khanty-Mansiysk and the Ob River above its Confluence with the Irtysh River	79
Figure 3:7. Estimated Contributions of Sediment from the Techa, Tom, and upper Irtysh River to the Suspended Sediment Load at Salekhard from 1960 to 1987.....	82

Chapter 4. Sampling, Analytical Methods, and Data

Figure 4:1. Map Showing Study Region with Sample Station Locations.....	86
Figure 4:2a. Schematic Outlining the Combustion, Acid Digestion, and Co-precipitation techniques developed at WHOI for Pu and Np ICP-MS Isotopic Analyses.....	94
Figure 4:2b. Schematic Outlining the Ion Exchange Clean-up and Purification and Sample Loading Techniques Developed at WHOI for Pu and Np ICP-MS Isotopic Analyses.....	95
Figure 4:3. Radionuclide intercomparison analyses for OB94-07B.....	101
Figure 4:4. Replicate radionuclide analyses by WHOI ICP-MS for 10g aliquots from OB94-10A.....	105
Figure 4:5. Replicate radionuclide analyses by WHOI ICP-MS for 10g aliquots from OB95-04.....	106
Figure 4:6. The $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio of excess plutonium detected in replicate analyses where concentration differences for both ^{239}Pu and ^{240}Pu were $> 3\sigma$ error.....	110

Chapter 5. Radionuclide Distributions, Sedimentation Rates, and Core Chronologies

Figure 5:1a. Depth distributions of ^{137}Cs in sediment cores from the Ob delta and Taz estuary	116
---	-----

Figure 5:1b. Depth distributions of ^{137}Cs in sediment cores from the upper reaches of the Ob, Irtysh, and Tobol Rivers.	117
Figure 5:2a. Depth distributions of ^{239}Pu in sediment cores from the Ob delta and Taz estuary	119
Figure 5:2b. Depth distributions of ^{239}Pu in sediment cores from the upper reaches of the Ob, Irtysh, and Tobol Rivers.	120
Figure 5:3a. Depth distributions of ^{240}Pu in sediment cores from the Ob delta and Taz estuary	122
Figure 5:3b. Depth distributions of ^{240}Pu in sediment cores from the upper reaches of the Ob, Irtysh, and Tobol Rivers.	123
Figure 5:4a. Depth distributions of ^{237}Np in sediment cores from the Ob delta and Taz estuary	125
Figure 5:4b. Depth distributions of ^{237}Np in sediment cores from the upper reaches of the Ob, Irtysh, and Tobol Rivers.	126
Figure 5:5a. $^{210}\text{Pb}_{\text{xs}}$ with exponential fits for sediment cores from the Ob delta and Taz estuary.....	131
Figure 5:5b. $^{210}\text{Pb}_{\text{xs}}$ with exponential fits for the upper reaches of the Ob, Irtysh and Tobol Rivers.....	132
Figure 5:6. Estimated fractions of global fallout and non-fallout ^{240}Pu in OB94-07B...	140
Figure 5:7a. $^{137}\text{Cs}/^{240}\text{Pu}$ profiles from sediment cores collected in the upper reaches of the Ob, Irtysh, and Tobol Rivers from global fallout maximum (GFM) to surface.....	143
Figure 5:7b. $^{137}\text{Cs}/^{240}\text{Pu}$ profiles from sediment cores collected in the Ob delta and Taz estuary from global fallout maximum (GFM) to surface.....	148

Chapter 6. Contaminant Records

Figure 6:1. Contaminant records in the Tobol River core, OB95-10	161
Figure 6:2. Contaminant records in the Irtysh River core, OB95-13.....	165
Figure 6:3. Contaminant records in the Lower Irtysh River core, OB95-06.....	169

