

Studies in Environmental Science 43

APPLIED ISOTOPE HYDROGEOLOGY A CASE STUDY IN NORTHERN SWITZERLAND

**F.J. Pearson Jr.⁷, W. Balderer², H.H. Loosli³,
B.E. Lehmann³, A. Matter⁴, Tj. Peters⁵,
H. Schmassmann⁶ and A. Gautschi¹**

With contributions by

J.N. Andrews⁸, P. Baertschi¹, G. Däppen³, J.-Ch. Fontes⁹, M. Ivanovich¹⁰,
M. Kullin⁶, J.-L. Michelot⁹, K. Ramseyer⁴, D. Rauber³, W. Rauert¹¹, S. Soreau⁹,
W. Stichler¹¹

- 1 *NAGRA (National Cooperative for the Storage of Radioactive Waste), Parkstrasse 23, CH-5401 Baden, Switzerland*
- 2 *Ingenieurgeologie, ETH-Hönggerberg, CH-8093 Zürich, Switzerland*
- 3 *Physikalisches Institut, Universität Bern, Sidlerstrasse 5, CH-3012 Bern, Switzerland*
- 4 *Geologisches Institut, Universität Bern, Baltzerstrasse 1, CH-3012 Bern, Switzerland*
- 5 *Mineralogisch-petrographisches Institut, Universität Bern, Baltzerstrasse 1, CH-3012 Bern, Switzerland*
- 6 *Geologisches Institut Dr. Schmassmann AG, Langhagstrasse 7, CH-4410 Liestal, Switzerland*
- 7 *Ground-Water Geochemistry, 1304 Walnut Hill Lane, Suite 210, Irving, Texas 75038, USA*
- 8 *Research Institute for Sedimentology, The University, P.O. Box 227, Whiteknights, Reading RG6 2AB, UK*
- 9 *Laboratoire d'Hydrologie et de Géochimie Isotopique, Université de Paris-Sud, F-91405 Orsay Cedex, France*
- 10 *AEA Industrial Technology, Isotope Geoscience Section, Harwell Laboratory, Oxfordshire OX11 0RA, UK*
- 11 *GSF Forschungszentrum für Umwelt und Gesundheit, Institut für Hydrologie, D-8042 Neuherberg, Germany*



ELSEVIER

Amsterdam — Oxford — New York — Tokyo 1991

CONTENTS

PREFACE	vii
CONTENTS	ix
TABLES	xiv
ILLUSTRATIONS	xviii
1. INTRODUCTION	1
1.1 Overview of This Report	1
1.2 Acknowledgments	2
1.3 Scope of Data Presented	2
1.4 Reporting of Isotopic Data	4
1.5 Regional Geology and Stratigraphy	7
1.5.1 Introduction	7
1.5.2 Crystalline Basement	7
1.5.3 Sedimentary Cover	11
1.6 Hydrogeology and Hydrochemistry	15
1.6.1 General Situation	15
1.6.2 Data Base	17
1.6.3 Groundwaters in Tertiary and Upper Malm Aquifers	17
1.6.4 Groundwaters in the Dogger, Lias and Keuper Aquifers	20
1.6.5 Groundwaters in the Upper Muschelkalk Aquifer	22
1.6.6 Groundwaters of Lower Triassic and Permian Aquifers	24
1.6.7 Groundwaters in the Crystalline Basement	26
2. LIMITATIONS OF SAMPLING AND ANALYTICAL PROCEDURES .	31
2.1 Contamination of Samples	32
2.1.1 Large-Volume Gas Samples for ^{39}Ar and ^{85}Kr	33
2.1.2 Samples for Carbon Isotopes	36
2.1.2.1 Samples from Test Borehole Programme	37
2.1.2.2 Other Samples	40
2.2 Comparison of Carbon Isotope Results From Conventional and AMS Measurements	43
2.3 Evaluation of Replicate Analytical Data	44
2.3.1 Dissolved Gases	45
2.3.1.1 Oxygen	45
2.3.1.2 Noble Gases	45
2.3.2 Uranium, Thorium and Daughter Elements	48
3. INFILTRATION CONDITIONS	65
3.1 Isotopic Composition of Modern Recharge	65
3.1.1 Stable Isotope-Altitude Relationships	68
3.1.2 $\delta^{18}\text{O}$ - $\delta^2\text{H}$ Relationship	78
3.1.3 Local Altitude-Temperature Equations	80
3.1.4 Stable Isotope-Temperature Relationships	82
3.2 Isotopic Composition of Groundwater	90
3.2.1 Samples from Quaternary, Tertiary and Malm Aquifers	90

3.2.2	Samples from the Dogger and Lias	93
3.2.3	Samples from the Keuper	93
3.2.4	Samples from the Muschelkalk	94
3.2.5	Samples from the Buntsandstein, Permian and Crystalline	98
3.3	Noble Gases in Groundwater	116
3.3.1	Introduction	116
3.3.2	Calculation of Recharge Temperature Corrections	118
3.3.3	Correlation Between Recharge Temperature and $\delta^2\text{H}$ and $\delta^{18}\text{O}$ Values	125
3.3.4	Results	128
3.3.5	Discussion	129
3.3.6	Recharge Temperatures and ^{14}C Results	138
3.3.7	Summary	140
4.	DATING BY RADIONUCLIDES	153
4.1	Introduction	153
4.2	Overview of the Sources of the Individual Isotopes	154
4.3	Detection of Young Water Components With ^3H and ^{85}Kr	157
4.4	^{39}Ar Results for Waters from Sedimentary Formations	166
4.4.1	General Conclusions from the ^{39}Ar Results	167
4.4.2	Conclusions from Corrected ^{39}Ar Results about Residence Times of the "Old" Water Components	170
5.	CARBONATE ISOTOPES	175
5.1	Overview of Groundwater Carbonate Evolution	175
5.1.1	Isotope Equilibria	176
5.1.1.1	Oxygen Isotopes	177
5.1.1.2	Carbon Isotopes	177
5.1.2	Evolution of the Isotopic Composition of Dissolved Carbonate	180
5.1.2.1	Chemical Mass Balances	180
5.1.2.2	Isotope Evolution Model	183
5.1.2.3	Application of Model Equations	188
5.1.2.4	Uncertainties in Results of Modelling	191
5.1.2.5	Comparison with Other Models for ^{14}C Correction	200
5.2	Age Interpretations of Dissolved Carbonate Isotopes	204
5.2.1	Calculation of Adjusted ^{14}C Ages	204
5.2.2	Discussion of Results	207
5.2.2.1	Samples Containing Modern ^{14}C	208
5.2.2.2	Samples with Virtually No Modern ^{14}C	211
5.2.2.3	Samples Yielding Definite ^{14}C Ages Greater than c. 12 ka.	212
5.3	Isotopic Composition of Carbonate Minerals and Water	224
5.3.1	Böttstein Samples	225
5.3.2	Weiach Samples	228
5.3.3	Riniken Samples	229
5.3.4	Schafisheim Samples	230
5.3.5	Kaisten Samples	231
5.3.6	Leuggern Samples	234
5.3.7	Summary	237

6.	ISOTOPES FORMED BY UNDERGROUND PRODUCTION	239
6.1.	Introduction	239
6.2	Chlorine-36	250
6.2.1	Results	250
6.2.2	Origin of ^{36}Cl in Groundwater Samples	250
6.2.3	^{36}Cl in Recharge	252
6.2.4	Deep Subsurface Production of ^{36}Cl	256
6.2.5	Origin of Aqueous Chloride	258
6.2.6	^{36}Cl in Water from Sedimentary Formations	259
6.2.7	^{36}Cl in Water from the Crystalline	261
6.2.8	^{36}Cl Measurements on Rock Samples	263
6.2.9	Summary	264
6.3	Argon-39 and Argon-37	266
6.3.1	Introduction	266
6.3.2	^{39}Ar Results	266
6.3.3	Escape of Argon Atoms from Rock into Water	268
6.3.4	Fractional Loss Coefficients of ^{39}Ar for the Individual Formations	268
6.3.5	^{37}Ar Measurements	272
6.3.6	Comparison of ^{39}Ar and ^{37}Ar	273
6.4	^3He and ^4He	276
6.4.1	Introduction	276
6.4.2	Results and First Interpretation	276
6.4.3	Subsurface Production	278
6.4.4	Helium Accumulation Times and Water Residence Times	279
6.4.5	Helium Transport Models	281
6.4.6	Correlation Between Helium and Chloride Concentrations	284
6.4.7	Summary	286
6.5	$^{40}\text{Ar}/^{36}\text{Ar}$ Ratios	288
6.5.1	Introduction	288
6.5.2	Results	288
6.5.3	Subsurface Production of ^{40}Ar	288
6.5.4	Correlations Between $^{40}\text{Ar}/^{36}\text{Ar}$ Ratios and the Potassium Contents of Rock and Water	292
6.5.5	Correlations Between He Concentrations and $^{40}\text{Ar}/^{36}\text{Ar}$ Ratios	292
6.5.6	Summary	294
7.	FORMATION-SPECIFIC CHARACTERISTICS OF GROUNDWATERS	297
7.1	Sulphur and Oxygen Isotopes in Sulphate and Sulphide	297
7.1.1	Comparison of Sulphur and Oxygen Isotope Analytical Results	298
7.1.2	Samples from the Tertiary and Jurassic	301
7.1.3	Samples from the Keuper	305
7.1.4	Samples from the Muschelkalk	305
7.1.5	Samples from the Buntsandstein and Permian	309
7.1.6	Samples from the Crystalline	312
7.1.7	Summary of Sulphur and Oxygen Isotope Results	316
7.2	Strontium Isotopes in Groundwaters and Minerals	323
7.2.1	Introduction	323
7.2.2	Analytical Procedures	324

7.2.3	Samples Analysed	324
7.2.4	Results	325
7.2.4.1	Groundwater Data	325
7.2.4.2	Crystalline Basement	327
7.2.4.3	Sedimentary Rocks	327
7.2.5	Discussion	328
7.3	Uranium and Thorium-Series Nuclides	336
7.3.1	Origin and Significance of Decay Series Disequilibria	336
7.3.2	Analysis of Concentrations and Activity Ratios	338
7.3.3	Interpretation of Groundwater Analyses	340
7.3.3.1	Uranium Content	341
7.3.3.2	Activity Ratios	341
7.3.3.3	Water from the Tertiary, Malm and Keuper	346
7.3.3.4	Water from the Muschelkalk	346
7.3.3.5	Water from the Buntsandstein	346
7.3.3.6	Water from the Permian	347
7.3.3.7	Water from the Crystalline	347
7.3.3.8	Conclusions	350
7.3.4	Interpretation of Rock Sample Analyses	351
7.3.4.1	Lias	353
7.3.4.2	Muschelkalk	353
7.3.4.3	Buntsandstein	354
7.3.4.4	Permian and Carboniferous	354
7.3.4.5	Crystalline	355
7.3.4.6	Fracture Infillings and Minerals	356
7.3.4.7	Conclusions	357
8.	SYNTHESIS OF ISOTOPE RESULTS	375
8.1	Tertiary and Malm	375
8.1.1	Calcium-Magnesium-Bicarbonate Groundwaters	376
8.1.2	Sodium-Bicarbonate Groundwaters	377
8.1.3	Sodium-Chloride Groundwaters	378
8.1.4	Conclusion	379
8.2	Dogger, Lias, and Keuper	380
8.3	Muschelkalk	381
8.3.1	Recharge Conditions and Residence Times	381
8.3.2	Geochemical Evolution	384
8.3.3	Conclusions about Muschelkalk Groundwater	387
8.4	Buntsandstein, Permian and Crystalline	387
8.4.1	Waters of Low Total Mineralisation from the Crystalline	390
8.4.2	Waters of High Mineralisation from Permian Sediments	392
8.4.3	Waters of Mixed Origin	394
8.4.4	Conclusions about Buntsandstein, Permian and Crystalline Water	396
8.5	Inter-Aquifer and Regional Flow	397
8.6	Recommended Further Studies	398

9.	SUMMARY	409
9.1	Analytical Results and Interpretative Methods	409
9.1.1	Quality of Sampling and Analysis	409
9.1.2	Water Origin and Recharge Conditions	410
9.1.3	Water Ages	412
9.1.4	Chemical Evolution	414
9.2	Hydrogeological and Hydrochemical Conclusions	416
9.2.1	Tertiary and Malm	416
9.2.2	Dogger, Lias and Keuper	417
9.2.3	Muschelkalk	417
9.2.4	Buntsandstein, Permian and Crystalline	417
9.2.5	Inter-Aquifer and Regional Flow	418
10.	LITERATURE CITED	421
	APPENDIX: Table of samples discussed in this report	A1