

CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES

TRIP REPORT

SUBJECT: Natural Analogue Working Group (NAWG) Meeting
20-5708-561

DATE/PLACE: October 28-30, 1996
Stein am Rhein, Switzerland

AUTHOR: William M. Murphy

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PERSONS PRESENT: Approximately forty representatives from approximately fifteen nations (most are identified below); Linda A. Kovach from the US NRC

BACKGROUND AND PURPOSE OF TRIP:

The NAWG is a European Commission (EC) sponsored international organization formed to promote studies of natural analog systems in support of geologic disposal of nuclear waste, and particularly in support of performance assessments (PA) of geologic repositories. This was the seventh NAWG meeting since 1985. The Nuclear Regulatory Commission (NRC) and the Center for Nuclear Waste Regulatory Analyses (CNWRA) have been active in the NAWG for many years. The purpose of the trip was to present an invited talk on a synopsis of the Peña Blanca and Santorini natural analog projects. A poster was also presented on recently completed radioisotopic studies at Peña Blanca.

SUMMARY OF PERTINENT POINTS:

The meeting emphasized national natural analog programs, and a wide spectrum of studies was illustrated having a broad range of topics and degree of development. Presentations of NRC-CNWRA studies at Peña Blanca and Santorini were well received. Abstracts were distributed at the meeting, and an EC publication will be published with complete papers. An initiative was launched to expand natural analog studies from nuclear waste issues to issues of toxic wastes, such as mine wastes. There is also an initiative to re-organize the NAWG, dissolving the "Core Group" and creating a new "Liaison Group" comprising one representative from each country. Individual presentations are described in the following section.

SUMMARY OF ACTIVITIES:

After brief introductory statements by Andreas Gauchi, a representative of the hosting organization, NAGRA, and Henning von Maravic, the EC Secretariat, the technical meeting started with a poster session. "Isotopic constraints on radionuclide transport at Peña Blanca" authored by David A. Pickett and William M. Murphy detailed data and interpretations. In general the data indicate oxidation product (uranophane) formation at 3 Ma, small migration of U out of the ore deposit into the host rock, and

elevated U in fracture filling materials outside the U deposit associated primarily with Fe oxides and hydroxides. U migration outside the U deposit over the last hundreds of thousands of years has been characterized by both U deposition and U removal from fracture filling materials. Studies of U series isotopic systematics permits quantitative interpretation of the mechanisms and timing of U transport.

Other posters included a description of studies at the Tono U mine in Japan, native Cu occurrences at Hyrokkola, Finland, naturally occurring zirconolites as analogs of the zirconolite phase in high level waste (HLW) forms, the behavior of a clay barrier at the Cigar Lake U deposit in Canada, and the Steenkampskraal monzonite mine, South Africa. Most of these studies were amplifications of oral presentations given subsequently, and will be treated in the reports on those talks.

In a poster by M. Satir and G. Bracke it was demonstrated using isotope data that Pb environmental contamination in the vicinity of the U mining site of Schlema-Alberoda, Germany, is due to industrial and traffic sources, not uranium mining operations or wastes.

In the oral session Julio Astudillo described the El Barrocal, Spain, project, which is a large, well developed, multidisciplinary project with EC support. U transport is being studied in hydrothermally altered, fractured granite. U oxidation is apparently affected by microbiological factors. U mineralization is associated predominantly with carbonates, not Fe oxides, under ambient conditions. Automated tracer tests are being conducted to monitor groundwater flow patterns. Performance assessment aspects of the El Barrocal project were discussed by Jordi Bruno. Uraninite is considered a source term analog. Clay minerals are considered analogous to clay barriers of an engineered barrier systems (EBS). Transport processes are being studied by examination of flow characteristics, matrix diffusion, and fracture mineralogy. Public acceptance of geologic disposal of nuclear waste is also promoted by these analog studies.

William Murphy presented a synopsis of the Peña Blanca and Santorini projects. The Nopal I U deposit at Peña Blanca is an uncanny analog of the proposed repository at Yucca Mountain (YM) on the basis of geology, geochemistry, hydrology, and uraninite occurrence. Studies there have focused on the performance issues of radionuclide source term and radionuclide transport. Studies have shown that uraninite oxidation at the site is rapid relative to removal of U from the site. Secondary uranyl minerals are implicated as source term controlling phases. These results correspond closely to those obtained in laboratory studies designed to mimic conditions at YM conducted independently and reported in the literature. Comparable analog and laboratory results bracket the repository time scale. Studies of transport indicate small amounts of U migration over small distances over millions of years. Some details were given in the poster described above. The Santorini project was conceived to test modeling of contaminant transport similar to that used to support PA for unsaturated systems. Buried artifacts at the Akrotiri archaeological site provide a source of contaminants that is relatively well constrained spatially and temporally, having been buried in volcanic deposits about 3,600 years ago. Site characterization was conducted, a model for contaminant transport was generated using these data, and results were compared to measurements of the contaminant plume. Some model results were qualitatively correct, however, some aspects of the system such as persistent transience of the system, were not reflected in the models. In general, however, observations at both Peña Blanca and Santorini indicate that these systems are relatively stable on time scale of thousands to millions of years.

Russell Alexander described work at the Maqarin, Jordan, site. Here interactions of natural cements with groundwaters generate high pH solutions. Mineralogy of the cements, evolution of the water chemistry,

and sealing of fractures with mineral alteration products are regarded of analogs of repository systems containing EBS cements.

Linda Kovach described aspects of U.S. natural analog programs other than CNWRA studies. The DOE-LANL has examined U series isotopes in some U minerals from Nopal I and confirmed CNWRA results that they are in secular equilibrium. DOE-LLNL studies at Wairaki, New Zealand have focused on geochemical modeling capabilities and thermodynamic and kinetic data requirements and quality for geochemical modeling. Dr. Kovach is conducting petrographic analyses of radioactive slag materials to identify their chemistry, mineralogy, and alteration characteristics.

Jan Cramer noted that the Canadian HLW program is in a public comment stage. At the natural analog site at Cigar Lake, Canada, stable and radioisotope studies have been used to characterize the age of U mineralization. Rare isotopes ^{36}Cl , ^{99}Tc , and ^{239}Pu have been detected. Mineral precipitation on fracture surfaces has been examined. Matrix diffusion has been tested. *In-situ* experiments have been conducted under high stress conditions. A clay layer at the site effectively isolates uranium from aquifer water circulating above the deposit.

Modern hydrologic and geochemical transport processes are being studied at the natural reactors in the vicinity of Oklo, Gabon, in another EC supported study as described by John Smellie who represented the Swedish program. Metallic fission product bearing phases have also been discovered in the reactor zones and are being studied as analogs of metal inclusions in spent fuel. There is also Swedish participation in the Palmottu, Finland, project described subsequently, where among other things, experiments are being conceived to test *in-situ* K_D determinations. At Maqarin, Jordan, the stability of clay in a high pH environment is being studied. Cs release from the Varutrask, Sweden, pegmatite is being studied. Also, a 90 year old cement water tank study and other case studies are under way.

Ferrugio Gera described a study at Orciatice, Italy, where an igneous intrusion in clay is being studied to examine thermal and thermal-mechanical effects of heating on the properties of clay.

Paul Hooker described British contributions to the Maqarin project which include geochemical model testing and thermodynamic data base evaluation including zeolite properties. The British also participate in the Steenkampskraal, South Africa, project. This site hosts a REE and Th deposit in monzonite. REEs constitute an analog of fission product behavior.

Russell Alexander listed projects in which the Swiss are participating. These include the Maqarin project, where activities are focused on mineral alteration and colloid formation and transport, matrix diffusion, and the relation between earthquakes and hydrology, e.g., seismic pumping. Collaborative activities are also conducted at the Tono Mine, Japan. Natural analog studies are also being used in public relations efforts in support of geologic disposal.

Walter Steininger represented an emerging German natural analog program. They are interested in the stability of rock salt mines and of backfilled salt and potash mines. Apatite is being considered as a backfill material because its capacity to form compounds with radioelements.

Julio Astudillo noted that the Spanish are participating in the Oklo, Palmottu, and El Barrocal projects, and noted also that they are studying alteration of clay minerals in two contexts. One study is related to the stability of clay EBS materials in the presence of a saline front, and the second is related to the stability of clay minerals in potential volcanic host rocks for a repository.

Didier Louvat offered a description of the Oklo Phase II studies, which includes investigations at related natural reactor sites at Okelobondo and Bagombe. The history of the sites is: sedimentation, natural reactor activity at 2 Ga, basaltic intrusion, orogenic metamorphism, and hydrothermalism associated with the opening of the Atlantic Ocean. Fission product daughters are found associated with apatite, florencite, and clay minerals; excess ^{235}U (Pu daughter) is associated with clays. Environmental isotopes are being examined to characterize the present groundwater flow system to examine radionuclide transport.

Peter Airey described the OECD/NEA sponsored Analogue Studies of the Alligator Rivers Region, Australia, project of which the NRC is a participant. Work focuses on the Koongarra U deposit. About half of the primary uraninite deposit has been oxidized and the uranium mobilized, and much is now associated with alteration minerals. U is contained in microcrystalline torbernite in Fe nodules. New SIMS analyses show high $^{234}\text{U}/^{238}\text{U}$ activity ratios of 30 in kaolinite surrounding uraninite rich rocks. Surface complexation studies sponsored by the NRC are focused on multimineralogic systems. In mixtures of kaolinite with either anatase or ferrihydrite, the latter mineral dominates the sorption characteristics of the mixture. The Australians are now looking for a low level nuclear waste site in an arid part of the country. There was discussion of "extension" of the Alligator Rivers studies (monsoon environment) to arid systems, but how this would be done is unclear.

Hidekazu Yoshida described studies at the Tono, Japan, U deposit research facility of redox front migration from fractures into the matrix of granitic rocks. The Japanese are also studying volcanic glass alteration as an analog of glass waste forms and the corrosion of a 1,800 a bronze bell buried in silty sandstone as an analog of EBS corrosion.

Runar Blomqvist described several Finnish projects. The Hyrkkola native Cu deposit is being examined as an analog for Cu container stability. At the Palmottu U deposit a well developed project focuses on U mobility, uraninite stability, and effects of glacial events. A large water chemistry data base has been accumulated with data from 60 packed sections in 30 boreholes. Consistency tests are being conducted for multiple redox couples and Eh measurements. In a separate project glacial meltwater intrusion in an ice marginal environment at Outokumpu is being studied.

James Dodds described geochemical and hydrological issues associated with mine wastes. Tailings are the greatest problem because of acid generation and heavy metal mobilization. Groundwater flow systems tend to be diverted to flow through underground openings rather than around them. Several individual mine sites were described in various stages of operation. The period of interest with respect to mine wastes is hundreds of years at most, and little attention has been paid to use of analog systems to judge the behavior of mine waste systems. It is apparent however that because of the long history of mining in Europe (thousands of years) a record exists of long term behavior of mine wastes. Studies of old mine wastes could provide an analog of the evolution of modern wastes. However, a modern practice is to dispose mine wastes in the abandoned underground openings, for which there are apparently no historical analogs.

Thomas Brasser compared and contrasted radioactive and toxic waste management practices. The two wastes are treated differently although the objectives of environmental and health safety are the same. Radioactive waste research and development and management (e.g., regulation) is far advanced relative to mine and toxic waste studies. Heavy metals are the principal concern in toxic wastes, but in many cases appropriate methods of characterization or chemical modeling (e.g., thermodynamic properties) are unknown or undeveloped.

Bernard Come described a program to examine the possibility of using natural analogs to support management of toxic wastes. Toxic waste categories were identified including vitrified fly ash and salt based wastes. Analogs for these wastes are hyperalkaline volcanic rocks and evaporite deposits, respectively. Weathering of a hyperalkaline volcanic rock site in Italy was studied and leach tests on this rock and vitrified fly ash were compared. Identification of relatively stable phases that would contain heavy metals would be a benefit. In the fly ash heavy metals are concentrated in sulfide minerals which are unstable in oxidizing conditions.

CONCLUSIONS:

Unlike the U.S. HLW program, most countries have not identified a specific site, although some have identified likely geologic conditions. Consequently most studies are of a generic basis. Selection of the YM site has permitted site specific analog studies, and relations between the analog sites and the behavior (performance) of the proposed repository are relatively well established.

Natural analog studies are being pursued vigorously by many national programs and international organizations including the EC. In contrast, support for natural analog studies in the U.S. is waning. The US DOE was not represented at the NAWG meeting.

Many analog programs in the world offer a wealth of information from a wide variety of environments. Countries with emerging geologic disposal program or with no site selected could benefit from an examination of the NAWG experience in aiding site selection.

PROBLEMS ENCOUNTERED:

None.

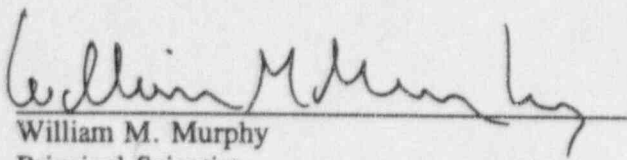
PENDING ACTIONS:

Manuscripts of the two presentations by the CNWRA are to be submitted for publication in the NAWG workshop proceedings.

RECOMMENDATIONS:

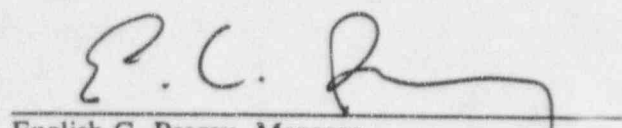
The NAWG is well established as the premier international organization serving as a forum for development of natural analog studies. Continued participation in this organization is recommended.

SIGNATURES:

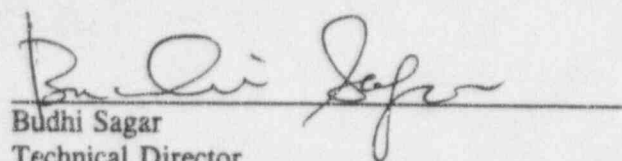

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