Energy and Technology Review

October 1980

Lawrence Livermore National Laboratory

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The cover

Test spill of 5 m^3 of liquefied natural gas (LNG) at the Naval Weapons Center, China Lake, California. Natural gas remains in liquid form only at temperatures below 113 K (-160°C). Thus, if the liquid is accidentally spilled in transport, it evaporates rapidly and expands far beyond its original volume, forming a cloud of flammable vapor that poses a serious hazard to anyone or anything in its path. Some sense of the size of the cloud shown in the cover photographs can be obtained by noting that the horizontal pipe is roughly 2 m above the surface of the pond. In tests such as this one, LLNL researchers are trying to amass the data needed to develop computer models of LNG dispersion under various atmospheric conditions. These models will help the U.S. Department of Energy provide realistic safety guidelines for the transport and storage of LNG. For further information on this project, see the article on p. 27.

About the journal

The Lawrence Livermore National Laboratory is operated by the University of California for the United States Department of Energy. The Laboratory is one of two nuclear weapons design laboratories in the United States. Today more than half of our effort is devoted to programs in magnetic and laser fusion energy, basic physics and chemistry, biomedical and environmental research, applied energy technology, and other research activities.

The *Energy and Technology Review* is published monthly to report on accomplishments in these research and applied technology efforts and on unclassified portions of the weapons program. A companion journal, the *Research Monthly*, reports on weapons research and other classified programs. Selected titles from past issues of the *Energy and Technology Review* are listed opposite the inside back cover.

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Energy and Technology Review

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LLNL Environmental Sciences Research Program

The LLNL Environmental Sciences Research Program explores ways in which developing energy technologies may affect the environment. It focuses on the flow of pollutants through the environment and the food chain to man and other target organisms. It includes studies of the composition and transport of pollutants, the physical, chemical, and biological transformations that they undergo, and their effect on aquatic organisms and terrestrial plants. It also includes development of new monitoring instruments and integrated assessments of the total environmental impact of energy operations. Its goal is to provide data that will make possible the development of benign energy technologies.

Support for our work comes largely from the the U.S. Department of Energy's Office of Environment, which has the major responsibility within DOE for evaluating the effects of DOE programs on health, safety, and the environment. Some individual projects are funded by other Federal agencies, including the Nuclear Regulatory Commission, the Department of the Interior, the Environmental Protection Agency, and the Department of Defense.

Our program addresses environmental concerns associated with nuclear, geothermal, and fossil-fuels technologies. In the nuclear area, we are studying the long-term effects of plutonium, other selected radionuclides, and copper corrosion products along the coasts and in the ocean (including their uptake and transfer through food chains). We are also assessing the potential radiation dose to natives of the Bikini and Enewetak atolls (in the Marshall Islands) who are returning to islands contaminated by nuclear explosions in the 1940s and 1950s.

In the geothermal area, we have conducted an integrated assessment of health, environmental, and socioeconomic issues raised by the prospect of geothermal development in California's Imperial Valley. At The Geysers geothermal area (also in California), we are participating in a program whose overall purpose is to assess environmental issues associated with the development of known geothermal resource areas in the United States.

Fossil fuel technologies specifically addressed in our program are direct coal combustion, oil extraction, *in situ* coal gasification, and *in situ* oil shale retorting. We are also concerned with the consequences of a large-scale liquefied-natural-gas accident.

In the September issue of *Energy and Technology Review*, we described environmental research at LLNL related to fossil fuel technologies. In this issue, we will describe recent work relating to nuclear, geothermal, and liquefied-gaseous-fuel technologies.

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The Marshall Islands project

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At the end of World War II, the U.S. entered into a Trust Territory Agreement with the six districts of Micronesia, thereby accepting responsibility for the welfare and education of their people. In 1947, the people of the Enewetak and Bikini Atolls (in the Marshall Islands) were relocated to other islands so that the U.S. could conduct nuclear tests there. In response to requests from the people for permission to return to their atolls and in anticipation of the termination of the Trust Territory Agreement in 1981, a series of field surveys and assessment studies have been conducted under the technical direction of LLNL. These studies are designed to estimate potential radiation doses and to allow the U.S. government to make reasonable recommendations on resettlement and land use. Integration of the field studies and the assessment effort has made the program more efficient.

In the 1940s and 1950s, the U.S. conducted nuclear tests at the Bikini and Enewetak Atolls in the Marshall Islands. In 1968, the people of Bikini (who had been relocated to other islands so that the tests could take place) asked to return. After a limited survey and

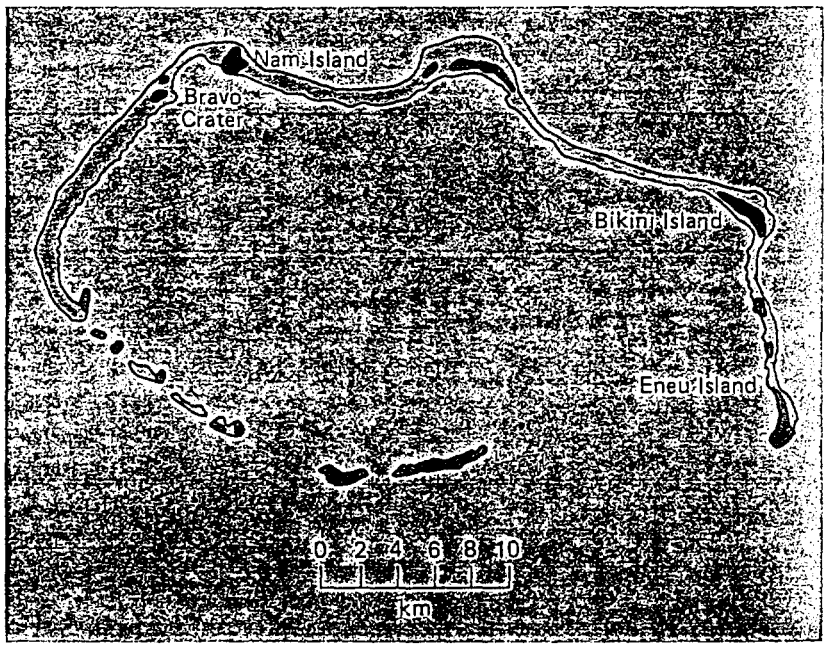

Fig. 1 *Map of the Bikini Atoll, site of nuclear testing in the 1940s and 1950s. In 1968, the people of Bikini asked to return to their home atoll, and a limited survey was conducted to evaluate the radiation doses that they might expect to receive. Resettlement of Bikini Island began in 1970. However, as locally grown subsistence foods became available (in late 1976 and early 1977), body burdens of cesium-137 began to rise. As a result, the people were moved off the atoll again in August of 1978. They have since requested to return to Eneu Island, and we are currently evaluating alternative living patterns and refining dose predictions to help in resettlement planning.*

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radiological dose assessment, resettlement began in 1970. In 1972, the people of Enewetak also asked to return to their home islands, and officials requested a detailed radiological survey and dose assessment. This dose assessment, conducted under LLNL technical direction, identified many areas where more precise data were needed. Thus, a field research project was launched in 1975.

In that same year, a second phase of housing construction was planned for Bikini Atoll, and careful planning of the location of the housing project was required to minimize external radiological exposure to the inhabitants. Accordingly, LLNL conducted a survey of Bikini and Eneu Islands to provide the data needed.

The most recent phase of our Marshall Islands research, a comprehensive survey of all the islands at Bikini Atoll (Fig. 1) and 12 other atolls downwind of the Enewetak–Bikini test sites (see Fig. 2), began in September of 1978. The purpose of this survey (and the dose assessment that it will produce) is to evaluate the radiological condition of atolls that could possibly have intercepted some fallout in excess of worldwide levels as a result of the testing program. Nearly 12 000 soil, vegetation, fish, water, sediment, and animal samples have been collected and processed, and analysis is about 60% complete.

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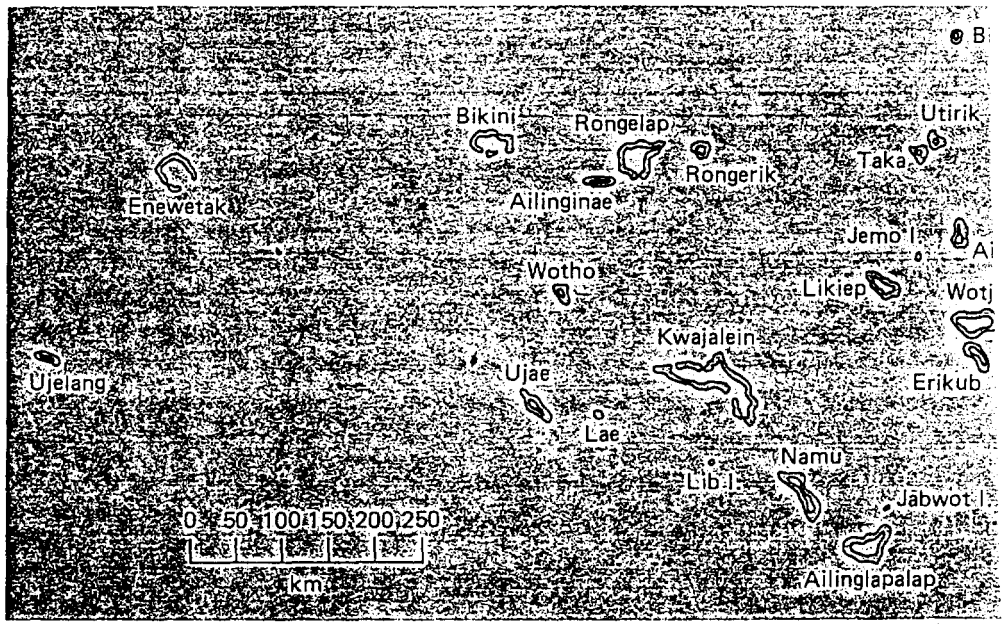

Fig. 2 *Atolls and islands of the Northern Marshall Islands. In 1946, prior to the start of the Pacific nuclear testing program, the Enewetak people were relocated to Ujelang Atoll and the Bikini people to Rongerik Atoll. From 1946 to 1958, 23 tests were held at Enewetak and 43 at Bikini. In 1948, the Bikini people were moved to temporary facilities Kili Island, a small island near M and 1973, some of the Bikini peopl and in August 1978 they were agai and several other atolls. At presen Bikini Atoll.*

plutonium-240 and americium-241 have long-term significance because they can be resuspended in the atmosphere and are present in lagoon water. (See the article on p. 8 for a discussion of the behavior of plutonium in marine environments.) However, the doses predicted for the transuranics are very low compared with those for cesium-137 and strontium-90.

A detailed data base

In 1977, we began an environmental research program on Eneu Island (Bikini Atoll) to measure concentrations of radionuclides in edible foods and thus provide more precise dose estimates. In our experiments, we were able to evaluate subsistence crops planted in 1970 by the Trust Territory government.

We are determining the concentration, rate of uptake, and residence time of strontium-90, cesium-137, plutonium-239 plus plutonium-240, and americium-241 in locally grown subsistence crops in the atoll ecosystem. We are also identifying concentration ratios (i.e., ratios of the concentration of radionuclides in various edible fruits to that in the soil) and correlation ratios (i.e., ratios of the concentration of radioactivity in different species of plants) for use in predictive environmental dose models. A comprehensive model of the transport, recycling, and ultimate disposition of long-lived radionuclides can then be developed.

To estimate the radiation dose to a given population, we measure the radionuclide concentrations in edible foods and correlate these data with appropriate dietary models, dose models for each of the radionuclides, and proposed living patterns. There were few locally grown foods at Bikini and Enewetak Atolls. We had to establish our own test plots of coconuts, Pandanus fruit, breadfruit, papaya, banana, squash, sweet potatoes, and other crops (on Enjebi Island at Enewetak Atoll and Eneu Island at Bikini Atoll).

The test plots helped us to understand the relationship between the concentration of radionuclides in edible fruit and in the soil. With accurate concentration ratios (i.e., fruit concentration/soil concentration), we can predict the concentration of radionuclides in foods that will eventually grow on islands and atolls currently devoid of food-producing plants.

The concentration ratio for cesium-137 in coconut meat (as measured in our test plots) is 6, and the concentration of cesium-137 in soil on Enjebi Island is 20 pCi/g. Thus, we can predict that coconuts planted on Enjebi island will have a cesium-137 concentration of roughly 120 pCi/g. Also, knowing that people in this area eat approximately 150 g of coconut per day, we can predict that they will ingest an average of 18 000 picocuries of cesium-137 per day (approximately 300 000 pCi/day is the limit of intake specified by the International Commission on Radiological Protection).

Our field research projects are designed to evaluate the environmental residence time of critical radionuclides in the atoll ecosystem. Cesium-137 and strontium-90 are responsible for over 95% of the total predicted doses, and both have radiological half-lives of about 30 years. However, if environmental processes are acting to reduce available cesium-137 and strontium-90 inventories so that the residence time is significantly less than 30 years, then the predicted doses will be much lower, and long-range land-use plans in the atolls could be altered.

We also study the mechanisms, processes, and controlling features of radionuclide cycling in the ecosystem. Our goal is to find ways to minimize the flow of radionuclides into subsistence foods and to reduce the available inventory in the soil.

Results

The results of our assessment at Enewetak Atoll are listed in Table 1; the ingestion dose includes that due to food and water, and the inhalation dose is negligible. Today, about 60% of the food eaten by the people of the Marshall Islands is imported. However, when such food is unavailable (because of shipping delays or weather conditions), they must subsist on locally grown foods. Hence, two types of diet are specified.

The dose rates predicted for the southern half of the atoll are much lower than the 100-mrem/yr background dose (at sea level) in the U.S. However, the maximum annual doses predicted for Enjebi Island (in the northern part of the atoll) do approach the current Federal limit for an individual (500 mrem per year). Table 2 lists the

Table 1 *Maximum dose rates for adult females at Enewetak Atoll when imported foods are available and when they are unavailable. Federal standards impose a 500-mrem/yr limit for an individual.*

Location	Type of diet	Organ	Pathway: Ingestion, mrem/yr	Pathway: External gamma, mrem/yr	Total, mrem/yr
Enjebi (Janet)	Imported foods available	Bone marrow	237	54	291
		Whole body	222	55	277
	Imported foods unavailable	Bone marrow	500	54	554
		Whole body	455	54	509
Southern islands	Imported foods available	Bone marrow	3.9	1.2	5.1
		Whole body	3.3	1.2	4.5
	Imported foods unavailable	Bone marrow	9.8	1.1	11
		Whole body	7.4	1.1	8.6

[a] An additional 22 mrem/yr is due to the natural background in the northern Marshall Islands atolls.

Table 2 *Integral doses (in rem) over 30- and 50-yr periods for adult females on Enjebi (Janet) Island. Federal standards impose a whole-body-dose limit of 5 rem over a 30-yr period.*

Nuclide	30-yr integral dose, rem: Whole body, Imported foods available	Whole body, Imported foods unavailable	Bone marrow, Imported foods available	Bone marrow, Imported foods unavailable	50-yr integral dose, rem: Whole body, Imported foods available	Whole body, Imported foods unavailable	Bone marrow, Imported foods available	Bone marrow, Imported foods unavailable
Ingestion								
^{137}Cs	4.3	8.7	4.3	8.7	6.5	13	6.5	13
^{90}Sr	–	–	0.39	1.2	–	–	0.059	1.9
$^{239+240}Pu$	–	–	0.0033[a]	0.014[a]	–	–	0.0087[a]	0.037[a]
^{241}Am	–	–	0.0046[a]	0.018[a]	–	–	0.013[a]	0.050
^{241}Pu (^{241}Am)	–	–	0.0021[a]	0.0077[a]	–	–	0.0078[a]	0.029[a]
External gamma[b]								
$^{137}Cs + {}^{60}Co$	1.4	1.4	1.4	1.4	1.9	1.9	1.9	1.9
Total	5.7	10	6.1	11	8.4	15	9	17

[a] Mineral bone dose rather than bone marrow dose; these doses are not included in the total.
[b] Natural background subtracted.

Table 3 *Maximum dose rates for full-time residents of Bikini Island or Eneu Island when imported foods are available and when they are unavailable. Federal standards impose a 500-mrem/yr limit for an individual.*

Location	Type of diet	Organ	Pathway: Ingestion, mrem/yr	Pathway: External gamma, mrem/yr	Total, mrem/yr
Bikini Island	Imported foods available	Bone marrow	905	189	1094
		Whole body	876	189	1065
	Imported foods unavailable	Bone marrow	1874	189	2063
		Whole body	1785	189	1974
Eneu Island	Imported foods available	Bone marrow	116	14	130
		Whole body	110	14	124
	Imported foods unavailable	Bone marrow	247	14	261
		Whole body	228	14	242

Table 4 *Integral doses over a 30-yr period for full-time residents of Eneu and Bikini Islands when imported foods are available. The Federal 30-yr standard is 5 rem.*

Nuclide	Eneu Island: Whole body dose, rem	Eneu Island: Bone marrow dose, rem	Bikini Island: Whole body dose, rem	Bikini Island: Bone marrow dose, rem
Ingestion				
^{137}Cs	2.5	2.5	20	20
^{90}Sr	–	0.19	–	0.97
$^{239+240}Pu$	–	0.00038[a]	–	0.00046
^{241}Am	–	0.0014[a]	–	0.0011
^{241}Pu (^{241}Am)	–	0.00058[a]	–	–
External gamma[b]				
$^{137}Cs + {}^{60}Co$	0.32	0.32	4.2	4.2
Total	2.8	3	24	25

[a] Mineral bone dose rather than bone marrow.
[b] Natural background subtracted.

predicted integral doses at Enjebi Island over 30- and 50-yr periods. The 30-yr doses are at or above the Federal guideline (5 rem whole-body dose) when imported foods are unavailable. As a result of these findings, the U.S. Government has recommended that there be no immediate resettlement of the northern islands.

It has been estimated[1,2] that the predicted doses for Enjebi Island would result in an increase of less than 0.5 case of cancer over the 75 cases expected naturally among the returning population. As for genetic defects, an increase of about 0.1 case over the 107 occurring naturally per 1000 births is expected. After comparing these risks to those that they face living on an outer atoll (typhoons, lack of medical care, sharks, etc.), the people of Enewetak wish to return to Enjebi Island.

The maximum annual dose rates and 30-yr integral doses predicted for future residents of Bikini and Eneu Islands in the Bikini Atoll are given in Tables 3 and 4. Higher concentrations of cesium-137 and strontium-90 at Bikini Island lead to doses that exceed the Federal short- and long-range guidelines. However, the predicted Eneu doses are below current federal guidelines, and the people want to return to that island.

Summary

Our dose assessments in the Marshall Islands must be as accurate as possible to protect the people from any unnecessary exposure. At the same time, we must not seriously overestimate risks that would be otherwise acceptable to people seeking to return to their home atolls. To this end, we are continuing to improve data on which the assessments are based and to make the assessments more precise.

Key words: Bikini Atoll; Enewetak Atoll; Marshall Islands; plants—radioisotope uptake; radioactive contamination.

Notes and references

1. M. A. Bender and A. B. Brill, *Assessment of Radiation Health Effects of the Resettlement of Enewetak Atoll*, National Cytogenetics, Inc., Valentine Road, Shoreham, New York (October 1979).
2. *The Enewetak Atoll Today*, U.S. Department of Energy (September 1979).

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Plutonium in the marine environment of the Marshall Islands

Between 1946 and 1958, the United States conducted 66 nuclear tests in the Pacific atolls of Enewetak and Bikini in the Marshall Islands. Since 1972, LLNL has conducted a variety of studies related to the cycling and transport of longer lived residual radionuclides in the marine environments of these atolls. Our primary concern is to collect sufficient data to develop recommendations to minimize the transfer of man-made radionuclides to people returning to the atolls. However, we have also attempted to relate our analytical findings to much wider fields of scientific endeavor and to develop models that are useful, first locally, and secondly in other aquatic environments receiving inputs from different source terms. One such program deals with the behavior of plutonium radionuclides in the marine environment of Enewetak and Bikini.

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Significant quantities of plutonium isotopes and other transuranics will be among the longer lived toxic radionuclides associated with radiological waste materials, some of which are already being disposed of in the ocean. It is, therefore, essential to understand by what processes and at what rates plutonium, under the influence of various chemical, physical, and biological phenomena on the sea floor, migrates back to the water column and is accumulated by marine food chains.

Lagoon sediment and water concentrations

After the last nuclear test at Enewetak in 1958, the residual radionuclides deposited in the lagoon water either settled to the bottom or remained as dissolved or particulate species that were eventually discharged to the North Equatorial Pacific by the prevailing exchange of water between ocean and lagoon. Fallout during and after these tests left a very heterogeneous distribution of radionuclides in lagoon sediments. Figure 1 shows the main features of plutonium-239 and plutonium-240 ($^{239+240}$Pu) activity associated with sediment components in the top 2.5 cm of Enewetak lagoon (as measured in 1972). Isolines distinguish regions with similar concentrations, and mean values are listed within regions. The number of measurements made is given in parentheses.

The highest surface concentrations are associated with sediment

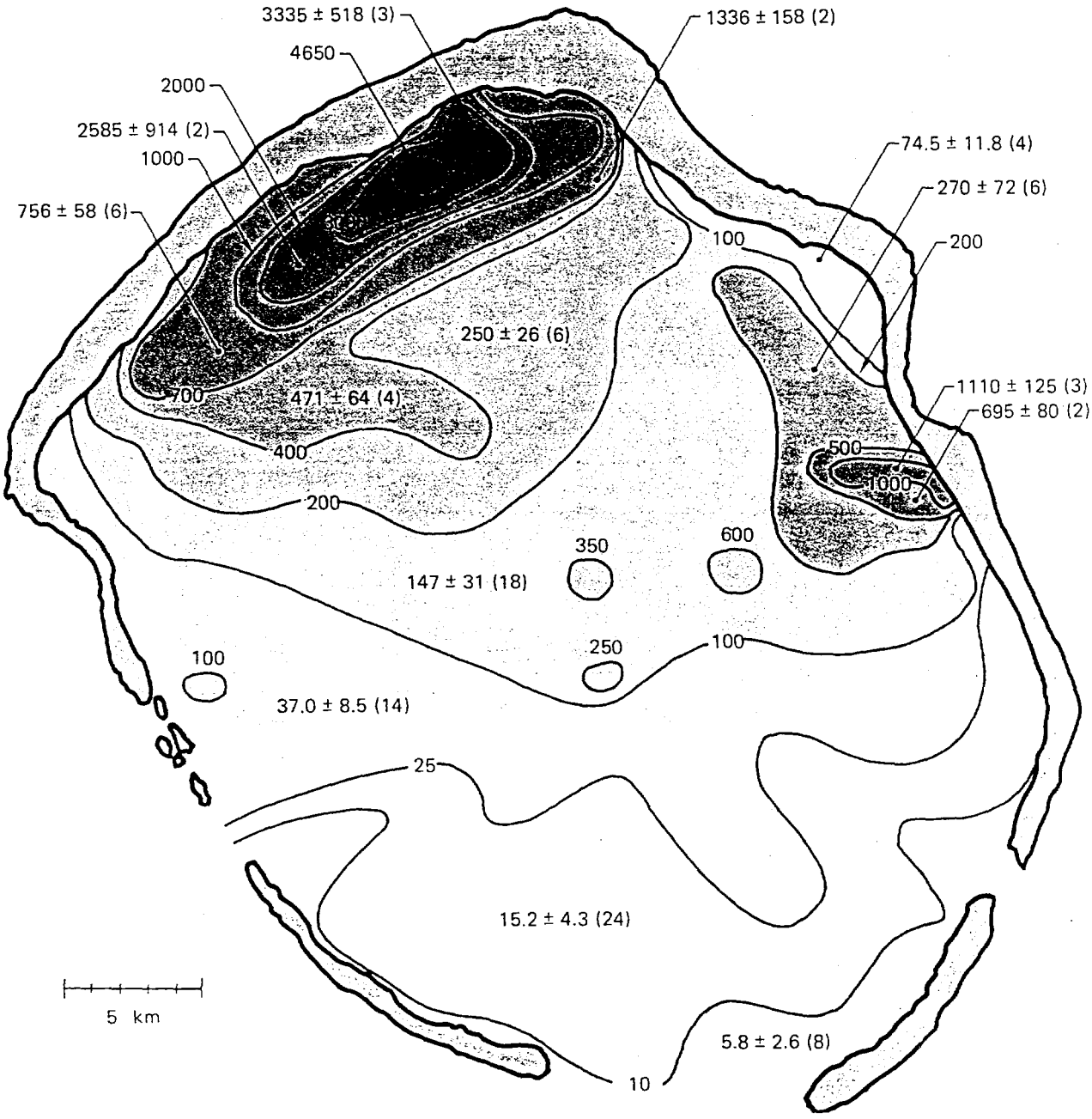

Fig. 1 *The distribution of combined plutonium-239 and plutonium-240 activity (in mCi/km^2) associated with sediment components in the top 2.5 cm of Enewetak Lagoon (measured in 1972). Isolines were constructed to distinguish regions with similar concentrations. Mean values are given within regions, and the number of measurements made is in parentheses. The highest surface concentrations are associated with sediment near (but not necessarily adjacent to) the area of the larger or more numerous tests.*

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near (but not necessarily adjacent to) the area of the larger or more numerous tests. Isolated small regions with relatively high concentrations of $^{239+240}Pu$ are evident in some of the less contaminated areas of the lagoon. The distribution of other long-lived, persistent radionuclides (including cesium-137, strontium-90, europium-155, cobalt-60, bismuth-207, and americium-241) are equally complex and unique.

It is generally believed that, following their introduction to the open ocean from any source, plutonium radionuclides become associated with particles and settle rapidly to the sea floor where they remain immobile after deposition. The concentrations of $^{239+240}Pu$ and other long-lived radionuclides in the lagoon water column during any year subsequent to 1958 should, therefore, equal the temporal fallout concentration levels in the North Equatorial Pacific surface water.

Accurate assessment of the concentration of plutonium in water requires analysis of large-volume water samples. Such analyses were facilitated by a specially developed[1] technique for the rapid separation of plutonium from 50 to 400 litres of water in the field. This new technique has effected a 100- to 800-fold reduction in sample size and has eliminated the high cost and inconvenience of transporting large-volume water samples to our home laboratory. Figure 2 shows water, collected in barrels, being processed for plutonium onboard a research vessel funded by the Department of Energy.

Table 1 summarizes the arithmetic mean of $^{239+240}Pu$ concentrations in filtered water samples collected during different periods from the regions of Enewetak and Bikini. Concentrations in surface water collected 1 to 2 miles west and south of the Bikini and Enewetak atolls and levels in the surface North Equatorial Pacific

Fig. 2 *Large-volume water samples are collected in large containers and processed onboard research ships to separate plutonium and other radionuclides. This process eliminates the high cost and inconvenience of transporting large volumes of water samples to our home laboratory.*

Table 1 *Summary of the concentration of plutonium-239 plus plutonium-240 in seawater in the Enewetak and Bikini lagoons and in the North Equatorial Pacific Ocean.*

Location sampled	Month/year sampled	Number of locations sampled	Mean $^{239+240}$Pu concentration, pCi/m^3	
			Soluble[a]	Soluble and particulate[a]
Enewetak Atoll				
Lagoon surface	11/72	29		29.0 (0.4–96)
	8/74	48	23.6 (1.4–65)	42.2
Lagoon surface (2 km off the inner atoll perimeter)	5/76	19	17.0 (2.1–31)	29.6
Lagoon bottom	11/72	6		44.0 (10–75)
	8/74	23	28.5 (3–69)	47.4
Lagoon bottom (2 km off the inner atoll perimeter)	5/76	10	14.0 (0.7–26)	29.0
Lagoon reef	11/78	5	17.0 (3–46)	
Ocean reef	10/75	9	114.0 (3–644)	
	5/76	6	26.0 (10–70)	84.0
	10/76	3	55.0 (28–29)	277.0
Bikini Atoll				
Lagoon surface	11/72	10	40.4 (3.9–79)	52.0
	2/77	18	52.0 (27–84)	
Lagoon bottom	11/72	7	40.0 (8.6–64)	111.0
	2/77	8	44.0 (13–104)	
Lagoon reef	11/78	8	29.0 (7–50)	
Equatorial Pacific				
1-8 km west and south of Bikini	10/72	4		15 ± 6
40-144 km west of Bikini	7/78	3		3.0 ± 0.8
1.6 km south of Enewetak	11/72	5		4.8 ± 3.0
	4/76	3		5.3 ± 3.0
3.2 km west of Enewetak	10/76	9		1.7 ± 0.4
North Equatorial Pacific	10/72 – 7/78	14		0.38 ± 0.12

[a]Values in parentheses represent the range in concentrations encountered at the location in question.

[redacted]

water at some distance from the atolls are shown for comparison.

Table 1 indicates that there are significant spatial and temporal differences in concentrations in the lagoon water. Wherever and whenever water was sampled in the lagoon or on the reef, the concentrations greatly exceeded the 0.3- to 0.5-pCi/m^3 fallout background levels in the Equatorial Pacific surface waters. These results are a direct indication that $^{239+240}$Pu is continuously mobilized to solution from sources within the atoll and is resuspended to the water column for subsequent redistribution both within and outside the atoll.

The mobilized $^{239+240}$Pu has solute-like characteristics and is available for uptake by organisms. Figure 3 shows that different oxidation states of $^{239+240}$Pu coexist in the lagoon water. From 75% to 94% of the dissolved $^{239+240}$Pu is in the oxidized (+5 or +6) form, while the rest is in a reduced (+3 or +4) state. The atolls have reached a chemical steady state with respect to the partitioning of $^{239+240}$Pu between the solution and solid phases of the environment. If this state persists, mobilization of the entire inventory of $^{239+240}$Pu from these atoll sediments will require more than 400 years. Although this is a long period of time compared to our life span, it is a small fraction of the radiological half-life of plutonium.

The mobilization of plutonium

A simple mass-action model employing an experimentally determined distribution coefficient (which relates the activity associated with a gram of dry sediment to the activity in a gram of water) is used to estimate the quantity of $^{239+240}$Pu that can be dissociated from the sediments into solution. The model estimates agree well with the measured average concentrations in water at both Enewetak and Bikini. Thus, our experimentally derived and field-verified distribution coefficient can be used to predict plutonium partitioning between solid–liquid phases. We are determining

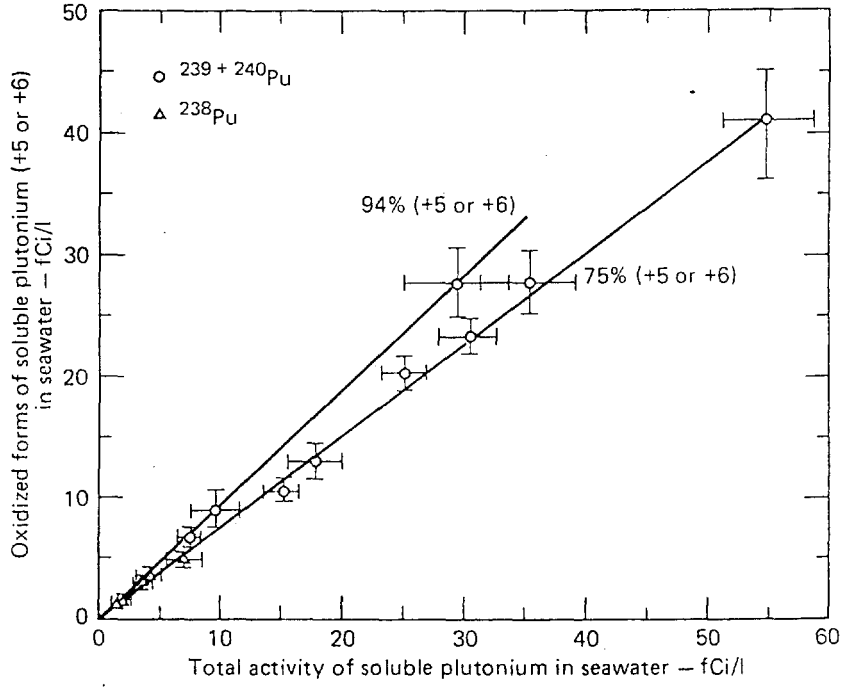

Fig. 3 *Activity of oxidized $^{239+240}$Pu in seawater at various locations in the Enewetak Lagoon compared to the total activity of plutonium in the water. From 75% to 94% of the soluble plutonium is in an oxidized (+5 or +6) form, and the rest is in a reduced (+3 or +4) state.*

whether these results can be used to predict partitioning of plutonium in other environments.

At the Farallones waste disposal area off the California coast near San Francisco, we see evidence of small amounts of plutonium-238 remobilization to bottom waters from sediments. The predicted concentration in bottom waters (based on the surface sediment concentration and the distribution coefficient) is 0.2 fCi/l (1 fCi equals 10^{-3} pCi). The measured concentration is 0.29 ± 0.14 fCi/l.

We have recently completed a study of plutonium distributions in the marine environment of San Clemente Island, off the coast of California. The mean concentration of $^{239+240}$Pu in 16 surface-sediment samples was 15 ± 6 fCi/g dry weight. Using the distribution coefficient of 2.3×10^5, determined from our studies in the Marshall Islands, we would predict that seawater in equilibrium with these sediments should contain 0.07 ± 0.03 fCi/l in solution. The average measured concentration in seawater samples was 0.09 ± 0.05 fCi/l. The agreement between the predicted and measured concentrations in these two examples is encouraging.

Summary

LLNL studies of the behavior of plutonium in the Marshall Islands are providing data relevant to the behavior of plutonium in other marine environments and results germane to problems related to the disposal of transuranics and other radioactive waste in the ocean.

Key words: Bikini Atoll; Enewetak Atoll; Marshall Islands; plutonium; radioactive wastes; water pollution.

Notes and references

1. K. M. Wong, G. S. Brown, and V. E. Noshkin, "A Rapid Procedure for Plutonium Separation in Large Volumes of Fresh and Saline Water by Manganese Dioxide Coprecipitation," *Radioanal. Chem.* **42**, 7-15 (1978).

[redacted]

Measuring transuranics in the air

For further information contact
Joe Kordas (422-6809).

The U.S. Department of Energy's Office of Health and Environmental Research is interested in improving the sensitivity and response time of air exhaust monitoring systems at nuclear facilities. At LLNL, we have developed and fielded a very sensitive alpha measurement system for determining the concentration of transuranics and other long-lived alpha emitters in the air. In tests at the Rockwell Hanford Operation's facilities in Richland, Washington, the system proved to be at least 100 times more sensitive than presently available monitors.

Large quantities of long-lived alpha-emitting substances (such as radium, thorium, uranium, plutonium, americium, and curium) are or will be handled at nuclear laboratories, fuel fabrication plants, chemical processing facilities, and future waste repositories. Online monitoring instruments play a critical role in evaluating and minimizing the release of such substances through exhaust stacks to the environment. A Transuranic Aerosol Measurement System (TAMS), recently developed at LLNL (Fig. 1), is capable of measuring extremely small quantities of transuranics (elements that have an atomic number greater than 92) or other long-lived alpha emitters in short time periods. TAMS will make it possible to assess both normal and accidental releases rapidly.

The monitoring environment

An online alpha detection system for stack effluent must be able to withstand high moisture and acidity and yet detect small quantities of long-lived alpha emitters in the presence of a much larger, natural alpha background consisting of the daughters of radon-222 and -220. Radon-222 and -220 are gases, but their daughters are charged and thus attach themselves readily to particles that are collected with long-lived alpha emitter samples. Figure 2 shows an alpha energy spectrum of this daughter background taken at atmospheric pressure. The colored area indicates where the peak caused by long-lived plutonium-239 would fall if it were present. Under normal conditions, at locations 1 metre above the ground, radon-222 and -220 concentrations range from 0.04 to 0.4 pCi per litre (20 to 200 times the 40-h maximum permissible occupational concentration for plutonium-239 in air).

Description of the TAMS system and its operation

TAMS combines a high sampling rate with high-resolution alpha spectroscopy and decay analysis to achieve extremely high sensitivity. We designed it specifically to measure concentrations of long-lived alpha emitters in corrosive stack effluent. Some of the prominent features of TAMS are:

- Separate collection and counting chambers to isolate the detectors from the effluent stream.
- A high inlet flow rate of 566 litres/min allowing frequent sampling.
- An evacuated detection chamber that improves the spectral resolution by a factor of five or more.
- Decay-scheme analysis to eliminate residual, natural alpha background caused by polonium-218. The analysis is based on the difference in the lifetimes of long-lived alpha emitters (several years) and polonium-218 (several minutes).

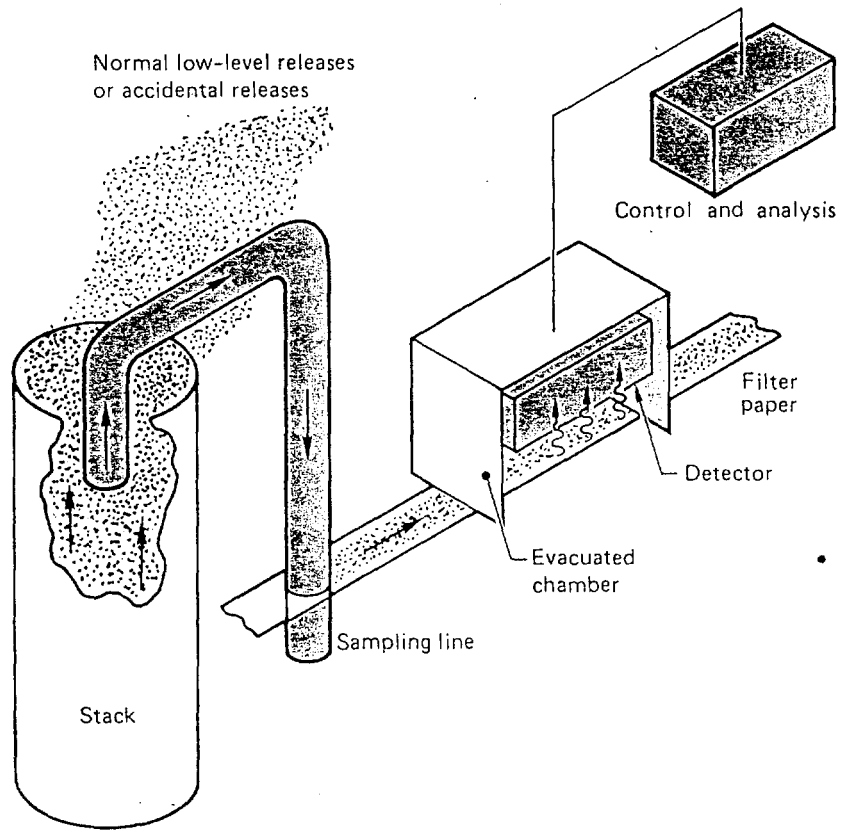

Fig. 1 *Schematic drawing of the Transuranic Aerosol Measurement System (TAMS). The system operates by drawing air from the exhaust stack of a nuclear facility, at a rate of 566 litres/min, through a membrane filter paper for a fixed period and then passing the filter containing the sampled particles in front of four ruggedized surface barrier detectors. The detector chamber is then evacuated, and the sample is analyzed while a second sample is collected. The entire process is controlled by an LSI-11 microprocessor.*

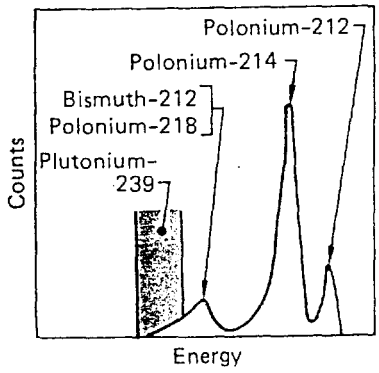

Fig. 2 *An alpha detection system for stack effluent must be able to detect small quantities of long-lived alpha emitters in the presence of a much larger natural alpha background consisting of the daughters of radon-222 and -220. Shown here is the alpha energy spectrum of that background collected on a Millipore SM filter and analyzed at atmospheric pressure. The colored area is where the peak caused by plutonium-239 would appear if it were present.*

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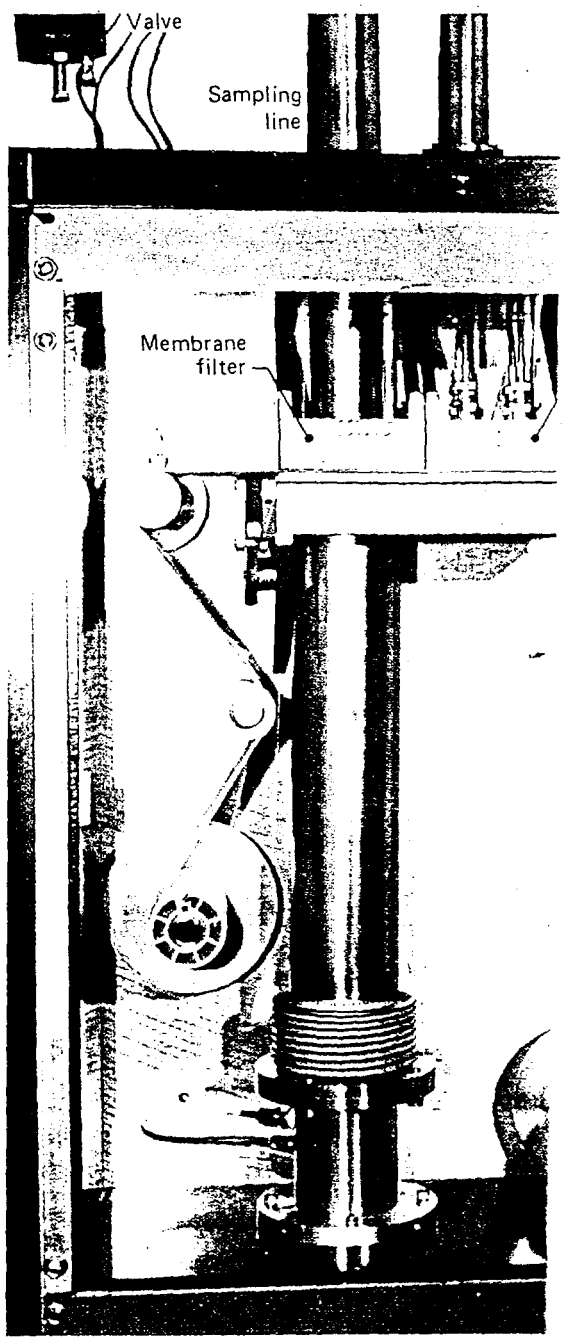


Fig. 3 *The LLNL Transuranic Aerosol Measurement System (TAMS). The flow of air down the sampling line is controlled by opening or closing the valve. The air passes through the membrane filter at a rate of 566 litres/min for a fixed period of time. The filter is then drawn into the detector chamber, and the chamber is evacuated. An LSI-11 processor then analyzes the alpha spectrum.*

The system operates by drawing air at a rate of 566 litres/min through a membrane filter paper for a fixed period and then passing the filter containing the sampled particles in front of four ruggedized surface barrier detectors. The detector chamber is evacuated to prevent the interaction of alpha particles with air (and thus produce a five-fold increase in resolution). The sample is then counted and the results analyzed while another sample is being collected. The entire process is controlled by an LSI-11 microprocessor. The system can vent the detector chamber, advance the filter paper, and pump the detector chamber back down to 20 mm of Hg in about 22 seconds. Figure 3 shows the actual sampling hardware used.

Figure 4, an alpha spectrum of a stack sample collected by TAMS at the Rockwell Hanford Operation plutonium-recovery facility, illustrates the spectral resolution of the system. The three peaks on the right are caused by the radon-222–radon-220 daughters, polonium-218, bismuth-212, polonium-214, and polonium-212. The peak resulting from polonium-214 has a full width at half maximum of about 100 keV. The first peak on the left indicates the presence of plutonium-239, and the second indicates the presence of plutonium-238 and/or americium-241.

Field results

The TAMS system was field-tested at the Rockwell Hanford Operation plutonium-recovery facility's Z-plant (in Richland, Washington) during the summer of 1978. The Z-plant is a chemical processing facility that converts plutonium nitrate to oxide or metal, recovers and dissolves scrap, and also separates americium from plutonium. The exhaust stack is about 8 m high and 36 cm in diameter with an effluent flow of about 71 000 litres/min. Our

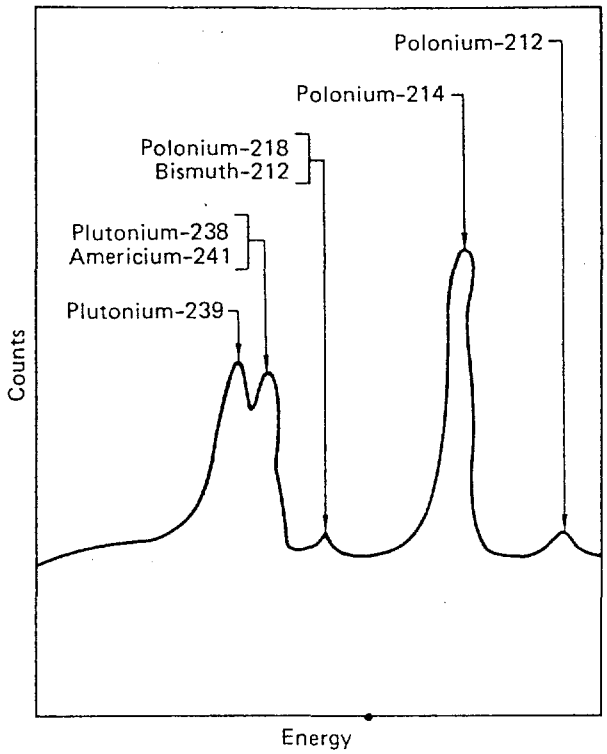


Fig. 4 *Alpha spectrum of a 60-min stack sample collected by TAMS at a plutonium facility. The three peaks on the right are caused by the radon-222-radon-220 daughters, polonium-218, bismuth-212, polonium-214, and by polonium 212. The first peak on the left indicates the presence of plutonium-239, and the second indicates the presence of plutonium-238 and/or americium-241. Note that TAMS can resolve plutonium-239 from the combined plutonium-238 and americium-241.*

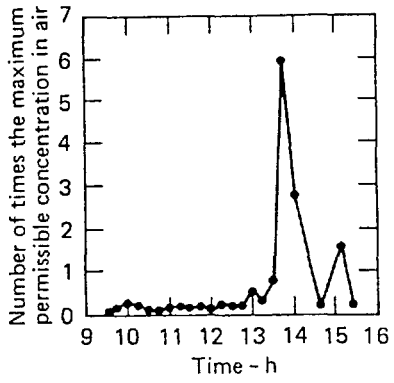


Fig. 5 *The TAMS system was fielded and tested at the Rockwell Hanford Operation plutonium-recovery facility's Z-plant (in Richland, Washington) during the summer of 1978. In these tests, it demonstrated a sensitivity 100 times that of any commercially available alpha-measurement system. Shown here is the combined concentration of plutonium-238, plutonium-239, and americium-241 in the sampled stack exhaust while a high-efficiency air particulate filter was being changed on July 10, 1978. Clearly, a low-level release is taking place. Normal dilution factors for this stack are such that a stack concentration of 50 times the maximum permissible concentration is reduced to below 2.5 before it reaches man. Man can be exposed to 100 times the maximum permissible concentration in air for up to 160 hours before the maximum allowed body dose is approached.*

sampling line (approximately 9 m long and 5 cm in diameter) was lowered approximately 1 m into the stack. The radius of curvature of the bend was about 1 m. The probe was designed for isokinetic sampling with a flow of 566 litres/min.

Figure 5 plots the combined concentrations of plutonium-238, plutonium-239, and americium-241 in the stack exhaust during the changing of a high-efficiency particulate air filter on July 10, 1978. In this test, the TAMS system demonstrated its ability to measure one-fourth of the maximum permissible concentration of plutonium-239 in air within a sampling time of only 15 minutes.

Conclusion

The TAMS field results are very encouraging. They indicate that the system has at least 100 times the sensitivity of any commercially available alpha measurement system.

We are currently designing a second-generation system (TAMS II) that will be about one-fourth of the size of the first and yet retain approximately 80% of its sensitivity. We expect it to be a prototype for a commercial unit.

Key words: air—monitoring; air—radioactivity; air pollution—monitoring; air pollution—radioactivity; nuclear power plants—environmental studies; radioactive wastes—environmental studies; transuranium elements.

The environmental impact of geothermal development in California's Imperial Valley

For further information contact David W. Layton (422-0918).

The Imperial Valley of California contains nearly one-third of the nation's identified natural hot water resources. With its 475 000 acres of irrigated lands and warm climate, it is also one of our major agricultural resources. Development of the valley's geothermal resources could be hindered if the environmental impact proves to be unacceptable or if geothermal operations are incompatible with agriculture. LLNL's Environmental Sciences Division has undertaken the Imperial Valley Environmental Project (IVEP) to measure and assess the impact of geothermal development in the area.

In 1975, the U.S. Energy Research and Development Administration (ERDA), predecessor to the Department of Energy, entered into a contract with the San Diego Gas and Electric Company to build and operate a geothermal test facility in the Salton Sea geothermal resource area. As part of this contract, ERDA agreed to sponsor field studies for the collection of baseline environmental data and to assess the potential impact of future geothermal development in the Imperial Valley. LLNL was given responsibility for fulfilling this agreement on ERDA's behalf, and the Imperial Valley Environmental Project (IVEP) was initiated. The main subject areas to be addressed were air and water quality, subsidence and induced seismicity, health and safety, ecosystems, and socioeconomics.[1]

The assessment process

We began our assessment by analyzing environmental data collected by private and public research groups. We then characterized various geothermal technologies in terms of their consumption of natural resources and their production of liquid, solid, and gaseous by-products. Finally, we assessed potential adverse environmental and socioeconomic effects, and evaluated methods of prevention and mitigation.

High-, medium-, and low-energy-production scenarios were prepared to serve as a basis for the assessment of the range of effects that might be observed. Our analysis focused on the medium-production scenario, which assumes a growth of 100 MW/yr in energy-generating capacity beginning in 1982 (Fig. 1). Under such a scenario, the total energy-production capacity would reach 3000 MW by the year 2010 (5% of California's projected peak load for that year).

The most important potential adverse effects addressed in our assessment were (a) air quality changes resulting from emissions of hydrogen sulfide, (b) increases in

the salinity of the Salton Sea resulting from use of agricultural waste waters for power plant cooling, and (c) induced subsidence and seismicity associated with the extraction and injection of geothermal fluids.

Air quality

The principal gaseous pollutant that could be released to the atmosphere from geothermal facilities is hydrogen sulfide, contained in the geothermal fluids extracted from subsurface reservoirs. It is released to the atmosphere after these fluids are "flashed" to produce steam used to run a turbine generator. The amount of hydrogen sulfide released depends on its concentration in the geothermal fluid and the quantity of fluid processed per MWh of electricity generated. Without control, power plants in the Salton Sea resource area could emit as much as 160 g/MWh, compared with a potential emission rate of 55 g/MWh for facilities in the East Mesa area.

We used computer models of atmospheric transport to simulate air quality changes resulting from 3000 MW of energy-generating capacity. The results of our simulations indicate that if hydrogen

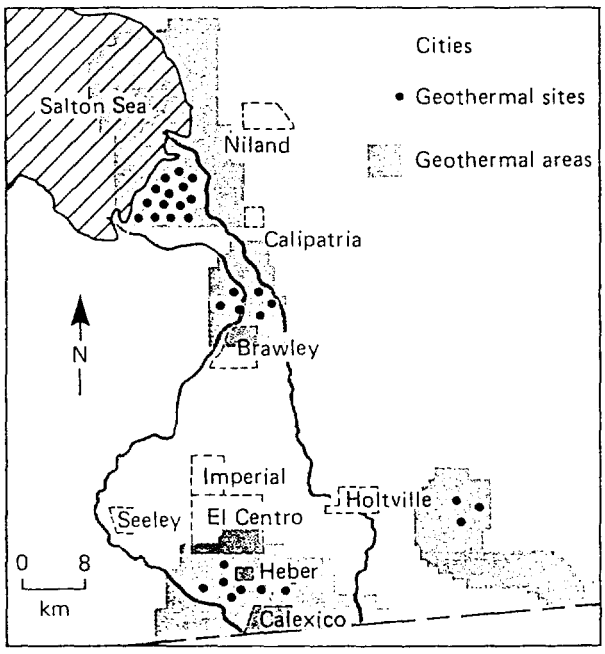


Fig. 1 *Power plant siting pattern postulated for a medium-growth scenario for geothermal development in the Imperial Valley (to 3000 MW in the year 2010). This siting pattern was prepared to enable us to carry out an assessment of the environmental impact of geothermal operations in the valley. At 3000 MW, power plants and related facilities will use, at most, only about 0.2% of the land normally irrigated in the valley.*

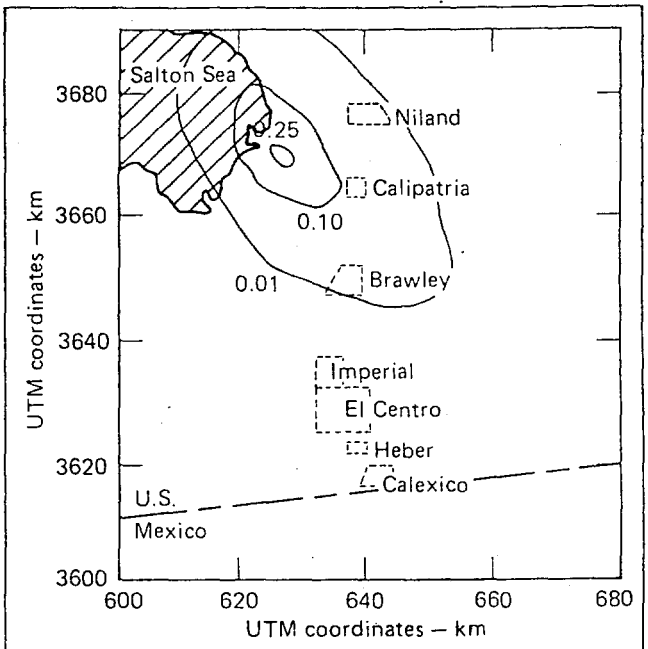


Fig. 2 *Isopleth plot showing estimates of how much of the time the California air-quality standard for hydrogen sulfide (42 $\mu g/m^3$ averaged over one hour) would be exceeded in the Imperial Valley, assuming a geothermal production capacity of 3000 MW. (Along a 1.0 isopleth, the standard is exceeded all the time, along a 0.25 isopleth the standard is exceeded one-fourth of the time, etc.) To prevent violations of the hydrogen sulfide standard at that level of energy development, facilities in the Salton Sea and Brawley resource areas would have to reduce emissions by approximately 82%.*

sulfide emissions are not abated to below 30 g/MWh, the California air quality standard (42 $\mu g/m^3$ averaged over one hour) will be exceeded over much of the valley by the year 2010 (Fig. 2).[2] This means that power plants in the Salton Sea and Brawley resource areas would have to reduce emissions by approximately 82%. Odor poses another problem (even if ambient levels of hydrogen sulfide are kept below the California standard) since 50% of the population can detect hydrogen sulfide at concentrations of 4 to 12 $\mu g/m^3$.

Hydrogen sulfide does have a beneficial fertilizing effect on crops grown in the valley but only at levels exceeding the ambient air quality standard.

Water quality

The extensive use of agricultural wastewater for power plant cooling or injection to geothermal reservoirs will lower the Salton Sea's elevation and increase its salinity. These changes have contradictory ecological effects. In the medium-growth scenario, reliance on wastewater to support geothermal development would arrest the rising elevation of the sea, which has been flooding riparian habitats in recent years. Increases in salinity, on the other hand, will have a negative impact on fish reproduction when salinities exceed 40 000 ppm total dissolved solids. Our analyses indicate that, with the medium-growth scenario and normal hydrologic conditions, toxic salinities would appear between the years 1985 and 1990.[2] Without development, toxic levels are not likely to appear until early in the 1990s (Fig. 3).

Large-scale use of wastewater would not be necessary if steam condensate were used as the sole source of cooling water for flashed-steam power plants. At present, however, a county policy favors the full injection of withdrawn geothermal fluids to protect against land subsidence. Thus, external sources of water will probably have to be used.

Subsidence and seismicity

The Imperial Valley is one of the most tectonically active areas of the United States, and the extraction and injection of large volumes of geothermal fluids could alter naturally occurring subsidence and seismicity. Our preliminary analysis indicates that the extraction of geothermal fluids could result in aquifer compaction within the reservoir, and this could eventually alter surface elevations.[2] Changes in the slope of the land could, in turn, hinder the irrigation of crops. We recommend that data on both subsurface compaction and changes in surface elevations (Fig. 4) be collected by the responsible agencies with a view to developing the ability to predict possible land deformation.

The injection of large volumes of spent geothermal fluids could alter seismicity. However, we believe that this is unlikely because fluids will be injected at low pressures (i.e., below the fracture pressure of rocks). Nevertheless, we recommend that the U.S. Geological Survey continue its monitoring of earthquakes as geothermal energy production proceeds to determine whether changes in seismic activity occur. It may be difficult to distinguish between natural and induced events in this seismically active region. Data on the location, frequency, and focal depths of earthquakes near geothermal fields will have to

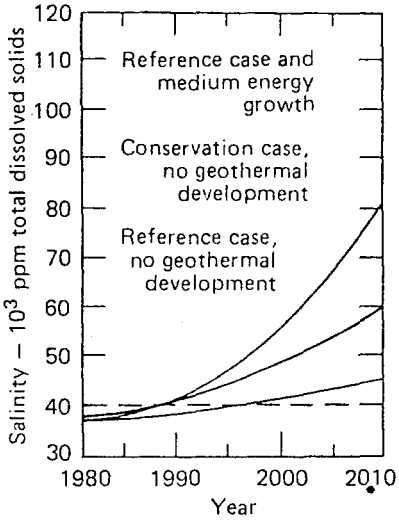


Fig. 3 *Salinity of the Salton Sea predicted for three cases: existing irrigation efficiencies (reference case), improved efficiencies (conservation case), and a reference case with growth in geothermal energy-generating capacity of 100 MW/yr and the use of agricultural waste waters for cooling. Predictions are based on normal hydrologic conditions and average evapotranspiration from crops. Toxic effects on fish reproduction are expected when the concentration of total dissolved solids exceeds 40 000 ppm (dashed line).*

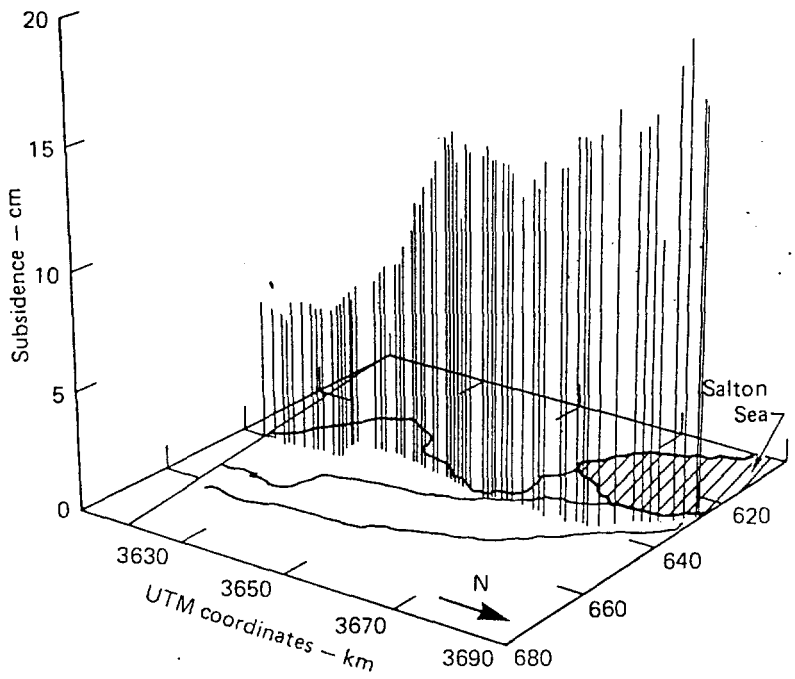


Fig. 4 *Subsidence for a 5-year period (1972–77) on a line of bench marks that runs from north to south in the central part of the Imperial Valley. We recommend that measurements of surface elevations be continued after geothermal operations begin to determine whether the extraction of geothermal fluids results in subsidence significantly greater than that occurring naturally.*

be compared with historic measurements to show whether significant changes have taken place. Since such comparisons require statistical analysis, continued monitoring by the present seismograph network is essential to enlarge the existing data base.

Conclusions

On the basis of assessments conducted so far, we believe that geothermal facilities can be operated in an environmentally acceptable manner. However, control technologies (such as hydrogen sulfide abatement) will probably be required. Continuous monitoring will play an important role in deciding what preventive actions and environmental controls are necessary.[2] We also hope to use the data so obtained to verify our predictive assessments.

Key words: geothermal energy—environmental studies; hydrogen sulfide; Imperial Valley; Salton Sea area.

Notes and references

1. A progress report on the IVEP is included in the May 1976 *Energy and Technology Review* (UCRL-52000-76-5), pp. 21-25.
2. D. W. Layton, L. R. Anspaugh, N. B. Crow, D. L. Ermack, C. H. Hall, J. R. Kercher, P. Leitner, W. F. Morris, and Y. E. Ricker, *An Assessment of Geothermal Development in the Imperial Valley of California, Vol. 1. Environment, Health, and Socioeconomics.* USDOE, Washington D.C. (in press).

Effects of geothermal effluents on ecosystems

Several environmental concerns have the potential to impede development of geothermal energy sources. Among them are hydrogen sulfide emissions (from geothermal steam) and mineral emissions from the cooling towers of geothermal power plants. Under the auspices of the Assistant Secretary for the Environment (U.S. Department of Energy), we are evaluating the effects of hydrogen sulfide and mineral emissions on surrounding ecosystems. Our goal is to determine to what extent these geothermal emissions are actually harmful.

For further information contact Joseph H. Shinn (422-6806).

Several potential pollutants are associated with the development of geothermal energy. Hydrogen sulfide gas separates from geothermal steam in a mixture consisting mostly of carbon dioxide but including traces of ammonia, methane, boric acid, mercury vapor, and other trace gases. Minerals in the hot water associated with geothermal energy production are of concern as they escape from cooling towers, infiltrate the leaves of nearby trees and plants, and enter streams and lakes. LLNL researchers are involved in studies of the effects of these materials on ecosystems in the vicinity of geothermal power plants.

The physiological response of vegetation to hydrogen sulfide

Very little was known about the effect of hydrogen sulfide on ecosystems before geothermal development began. Our experience told us that the organisms most affected by it would be plants. Woody plants will respond to sulfur dioxide when exposed to concentrations of under 0.10 ppm for a few hours. (Hydrogen sulfide begins to affect human health—by irritating nasal passages in a minor way—only at concentrations an order of magnitude above those at which harmful effects on plants are first observed.) We also knew that the uptake of gases by leaf cells is strongly controlled by the solubility of the gases in water and that all inorganic forms of sulfur gases are metabolized in the same manner. We hypothesized that foliar injury due to hydrogen sulfide, although significant, would be less than that due to sulfur dioxide and that the long-term effects could include damage to habitat and food sources in forests and damage to crops in agricultural areas.

In 1976 we first measured the physiological response of plants to various combinations of geothermal emissions. We placed open-topped chambers around rows of

lettuce and sugar beets in the field and injected a mixture of gases (common in geothermal emissions) at various concentrations through the bottom of the chambers (Fig. 1). Each chamber maintained a constant exposure condition for a period of several hours, and during that period we measured the rate of photosynthesis (milligrams of carbon dioxide fixed per gram of leaf tissue per hour) and the resistance of leaf pores (stomata) to the diffusion of gases into the leaf (inverse conductance, i.e., s/cm). We have pioneered techniques for making these measurements in the field.[1]

We found that geothermal air emissions actually stimulate photosynthesis at low concentrations (see Fig. 2) because the addition of carbon dioxide and hydrogen sulfide provides elements needed for carbohydrate and amino acid production. The first significant decrease in photosynthesis occurred when the concentration of hydrogen sulfide exceeded about 3 ppm (4200 $\mu g/m^3$) in the mixture. This value is two orders of magnitude

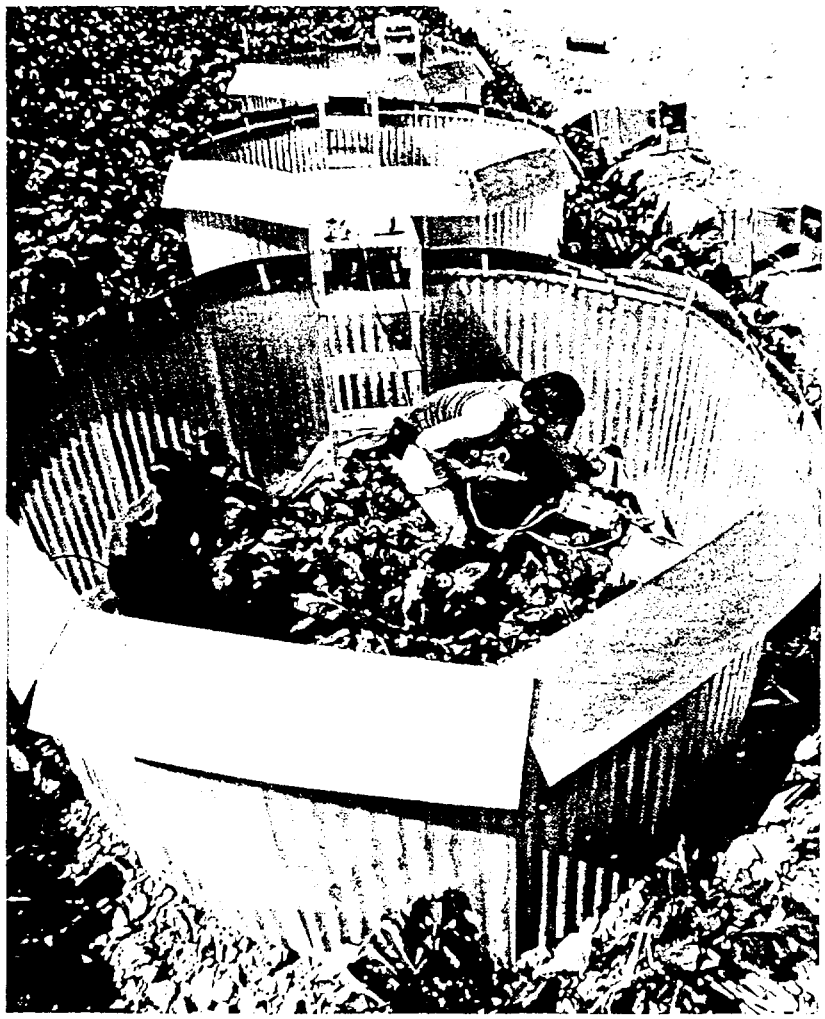


Fig. 1 *Open-topped chambers used to measure the reactions of plants to hydrogen sulfide and other gases. Potential air pollutants are injected in various concentrations into the bottom of the chamber. Each chamber maintains a constant exposure condition for a period of several hours, during which researchers make physiological measurements to evaluate effects on the plant's metabolism.*

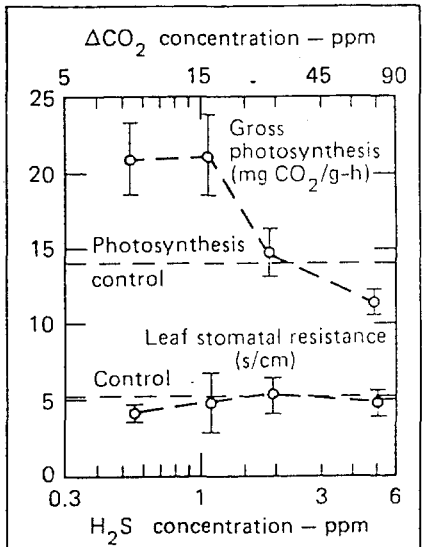
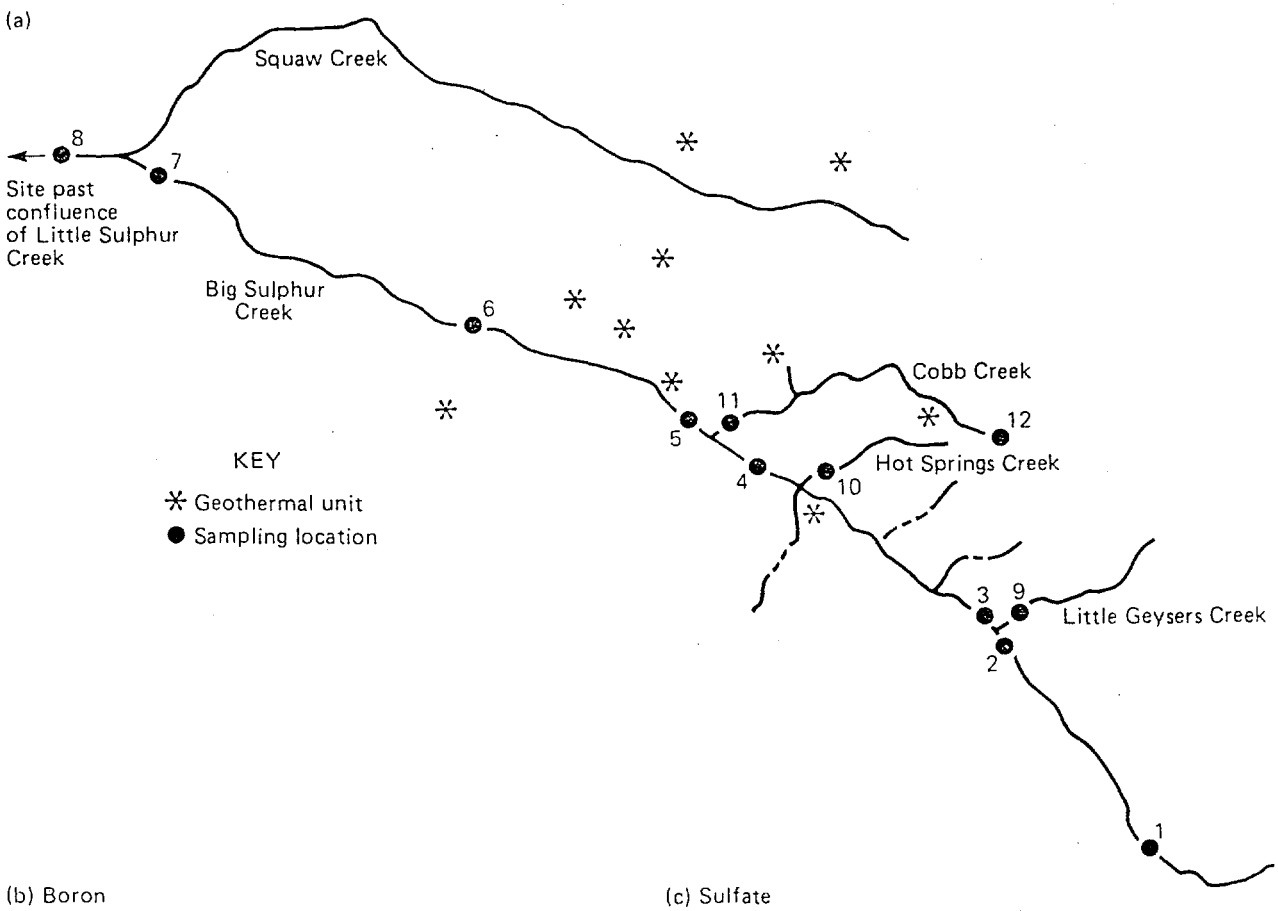
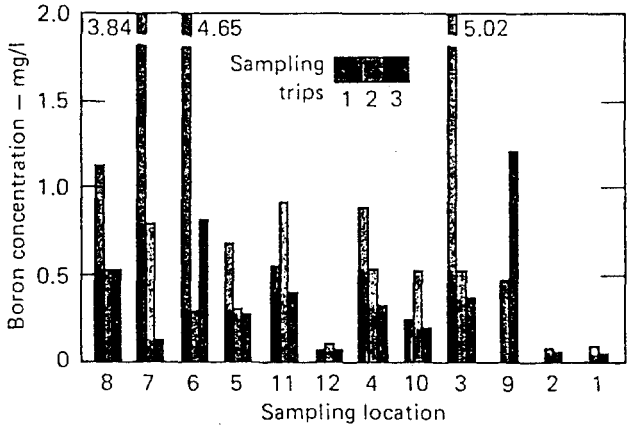


Fig. 2 *Photosynthesis rates (mg CO_2/g-h) and stomatal resistance (s/cm) in lettuce exposed (for 3 h at 30°C and high light) to a mixture of gases similar to that emitted by geothermal power plants. Geothermal emissions actually stimulate photosynthesis at low concentrations because the additional carbon dioxide and hydrogen sulfide provide essential elements for carbohydrate and amino acid production. The first significant decrease in photosynthesis occurs when hydrogen sulfide exceeds about 3 ppm in this mixture.*





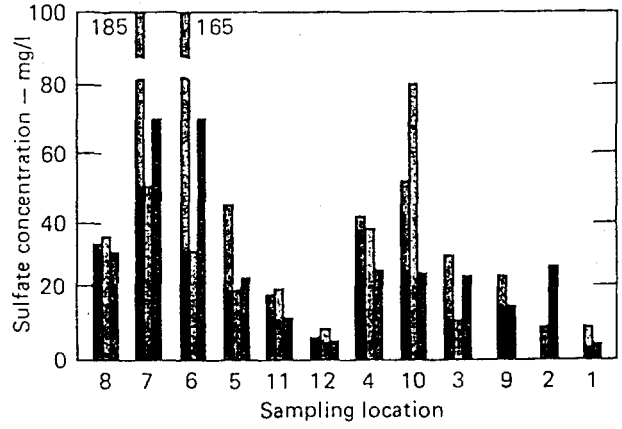


Fig. 3 *Tributaries of Big Sulphur Creek (a) show elevated levels of boron (b) and sulfate (c) where the local watersheds are affected by deposition downwind from operating power plants. Although the aquatic ecosystem at The Geysers may well be adapted to these relatively minor increases, this mode of transport and the ultimate fate of the mineral material chronically deposited at geothermal development sites must be investigated.*

[redacted]

higher than the California air quality standard.

To prove that this response was the rule rather than the exception, the Statewide Air Pollution Research Center at the University of California, Riverside, screened many crop and tree species under laboratory conditions to determine their relative sensitivity to hydrogen sulfide. Later, we used GROW1, an LLNL-developed computer simulation of the mechanisms of biochemical transformation of sulfur and energy allocation in plants, to show that the observed physiological responses do not indicate a substantial loss of growth for any reasonable, worst-case geothermal emission scenario (even when urban smog—i.e., ozone—is present). Hydrogen sulfide is thus only a problem from the point of view of odor. The California air quality standard has a more than adequate margin to protect vegetation, even when ozone is also present.

Mineralized emissions from cooling towers

Utility companies realize that heavily mineralized water (which will inevitably be associated with geothermal energy production) can kill off nearby vegetation as it escapes from the cooling towers. At the Geysers, the Pacific Gas and Electric Company has observed the death of more and more plants in deposition zones downwind from its power stations. However, the affected area is of trivial size compared to the land area (approximately 5% of the total) required for roads and well heads.

We have measured mineral deposition by positioning potted barley plants and passive aerosol collectors downwind from The Geysers power plants. Boron showed up in the barley and native plants and in animals. At 100 m downwind, deposition was primarily a result of the drift of liquid aerosols (reflected in high flux rates onto leaf tissue and the ground). However, at 1200 m and beyond, we observed higher deposition of cooling tower minerals than would be expected from liquid aerosol transport. Thus, dry transport and deposition mechanisms must also be operative. We observed significant increases in the levels of minerals in runoff water collected at the confluence of streams draining the area. Figure 3 shows boron and sulfate concentrations attributed to deposition on watersheds surrounding The Geysers.

Although aquatic ecosystems at The Geysers may well be adapted to these relatively minor increases, it is important that this mode of transport (and the ultimate fate of the mineral material chronically deposited) be investigated because of the possibility of serious effects over a period of decades. This potential problem has only recently been recognized.

Summary

Concern about hydrogen sulfide has been a substantial institutional impediment to the development of geothermal resources. However, our research shows that its actual impact on ecosystems is minor. Conversely, the long-range transport of minerals from cooling towers has been largely ignored, but it appears to be worthy of further investigation.

Key words: boron; geothermal energy; geothermal power plants; hydrogen sulfides.

Notes and references

1. See "Portable Instrumentation for Environmental Field Studies," in the September 1980 *Energy and Technology Review* (UCRL-52000-80-9), pp. 36-41.

Vapor dispersion from LNG spills

For further information contact Ronald P. Koopman (423-0163).

We have carried out a series of experiments on vapor dispersion from spills of liquefied natural gas (LNG) at the Naval Weapons Center, China Lake, California. In each of these tests, about 5 m^3 of LNG were dumped on a water pond in winds ranging from 3 to 10 m/s. Our main objective was to evaluate gas sensors to be used in future large-scale tests, but in addition, we achieved good agreement between experimental data and computer simulations. We have recently begun a series of 40-m^3 test spills. In these and planned larger scale experiments we will measure the dispersion of LNG vapor clouds with a view to developing better models of the effects of density, temperature, and turbulence on dispersion. The final product of this research will be a set of experimentally validated models that predict the consequences of a large-scale LNG accident. These models will help DOE provide improved safety guidelines for the transport and storage of LNG.

Ocean-going tankers filled with liquefied natural gas (LNG) may supply at least 10% of the natural gas consumed in the U.S. by the mid-1980s. However, this means of transport involves potential hazards far beyond those already accepted in conventional natural gas use.[1] To develop a detailed understanding of the potential hazards so that they can be controlled, the Division of Environmental and Safety Engineering of the U.S. Department of Energy has initiated a Liquefied Gaseous Fuels Safety and Environmental Control Assessment Program in which LLNL is participating.

Methane, the principal constituent of natural gas, remains liquid only at or below 113 K (−160°C), and 1 m^3 of liquid expands at atmospheric pressure to produce 600 m^3 of gas. The gas is flammable when mixed with 5 to 15 times as much air. Thus, 1 m^3 of liquid can form 10 000 m^3 of a highly flammable air–gas mixture. Because of its low temperature and high relative density, the vapor released by an LNG spill will hug the ground and spread in a cloud over a large area. Some fraction might become flammable or detonable, depending on the spill rate, the spill site, and the exact LNG composition. If a flammable

cloud covered a town or city, its almost inevitable ignition would cause widespread destruction.[2]

Although elaborate safety precautions have been proposed to minimize the chance of a large LNG spill, such an accident could still take place. Spills might result from storage tank ruptures, ship collisions, or leaks on land or at sea. They could occur during transfers from ships to storage tanks or during pipeline transfer on land. Because of the large chemical potential energy associated with the fuel in liquefied form and its ability to form a large cloud of vapor rapidly, any spill is potentially dangerous. Thus, it is vital to explore the possible consequences of such spills and how to minimize adverse effects.

When we began our research, existing models disagreed by over an order of magnitude on how far the vapor cloud from a particular large spill would travel downwind. There was also very little previous work on scaling relationships that would allow us to extrapolate from the results of small experiments to full-scale effects. There was no existing facility at which we could safely carry out large experimental LNG spills under the conditions necessary to validate our numerical models.

Since mid-1978 we have participated in a series of spill experiments at the Naval Weapons Center, China Lake, California, primarily to evaluate instruments for the larger spill experiments we plan, but also to gather data that can be used to improve our predictive ability. The first four of these tests involved spills of 5 m^3 of LNG each and produced vapor clouds that were warmer, extended farther, and contained regions more enriched in the heavier (and more detonable) hydrocarbons than expected. We are now engaged in a series of 40-m^3 spill experiments at China Lake (using the instruments and data-gathering techniques derived from the previous experiments) to further improve our models and refine our techniques for even larger experiments.

We began our experimental program with 5-m^3 spills because an existing facility at China Lake was amenable to such tests and because spills of this size were adequate to evaluate the performance of gas sensors in the field. We realized that these tests would not be large enough to permit observation of all important phenomena associated with the dispersion of large clouds of natural gas, but there was the possibility that such experiments would teach us something new about the process.

Experimental array and instrumentation

Figure 1 shows the instrument array used in our 5-m^3 spill experiments. The array consisted of eight primary stations distributed downwind from the spill point, each one equipped with a variety of grab samplers, gas sensors, and thermocouples (points 1 through 8). The primary stations were supplemented at points 9, 10, and 11 by anemometers and at point 12 by a Jet Propulsion Laboratory (JPL) gas sensor. There was also a LIDAR (laser radar) system fielded by Computer Genetics Corporation and a 12-point square array of Mine Safety Appliances (MSA) gas sensors (not shown in the figure). Cables connected each instrument station to power sources and the data-acquisition trailer. The main function of the grab samplers, gas sensors, and thermocouples was to measure the concentration of the LNG vapor in various ways at various locations.

The grab samplers consisted of evacuated bottles that were opened at prescribed intervals to collect a sample of gas for later analysis by mass spectrometer to verify the performance of our gas sensors.

The Shell gas sensors (stations 1 and 2), developed and loaned to us by Shell Research Ltd., of England, measured heat loss from a heated filament exposed to the gas stream. The TSI sensors (stations 3 and 6), manufactured by Thermo-Systems, Inc., of St. Paul, Minnesota, operated by forcing the gas through a sonic nozzle and measuring, with a thin-film anemometer, the flow velocity (which depends strongly on gas composition). The infrared gas analyzer (station 4), custom-built for us by Anarad, Inc., was capable of distinguishing methane, ethane, and propane. It gave us our first quantitative evidence of differential boiloff of the different hydrocarbons from the liquid pool.

Fig. 1 *Instrumentation array for a series of 5-m³ test spills of liquefied natural gas (LNG) at the Naval Weapons Center, China Lake, California. The array consisted of eight primary stations (points 1 through 8) distributed downwind from the spill point, each one equipped with a variety of grab samplers, gas sensors, and thermocouples. The primary stations were supplemented at points 9, 10, and 11 by anemometers and at point 12 by a Jet Propulsion Laboratory gas sensor. The radial distance from the spill point in the pond to stations 7 and 8 was 91 m. Instruments at stations 1, 2, 4, 5, 7, and 8 were positioned 0.9 m above the pond surface. Instruments at stations 3 and 6 were 0.3, 0.9, and 2.4 m above the surface. The center line of the array coincided with the prevailing wind direction. The LIDAR system, a laser-based remote gas sensor, detected Raman scattering from methane within a series of 9-m-long range gates. (Elevation contours are in feet above sea level.)*

The gas sensor at station 5 was a miniature, rapid-response, infrared system prototype that we developed for DOE and modified to detect methane.[3] It operates on the principle of differential absorption of infrared radiation by various gases. One filter allows infrared radiation of a wavelength heavily absorbed by methane to pass, and another gives us an unabsorbed wavelength. By comparing the intensities of the two beams after they have passed through the gas, we can calculate the methane concentration. The infrared sensor was the only truly portable instrument used in these tests. It operated well in environments with temperatures ranging from −20 to +40°C. Its measurement capabilities were essentially independent of the presence of water vapor, droplets, or dust in the absorption cell.

Stations 7 and 8 had, in addition to the usual grab samplers and thermocouples, Mine Safety Appliances (MSA) gas sensors. These sensors used a filament covered with a catalyst (to promote combustion) together with a means of measuring the heat evolved. They detected gas only up to the stoichiometric concentration (10 vol% for methane).

The LIDAR instrument, used only in the second test of the series, operated remotely over a line of sight that extended through the gas plume. It used a pulsed laser to excite the gas and then measured time-gated Raman-scattered return signals from the 9-m-long range gates shown in Fig. 1.

At each of stations 1 through 8 there was a thermocouple, a grab sampler, and a gas sensor measuring conditions at a standard height of 0.9 m above the ground. Most stations had an auxiliary thermocouple at 0.6 m. Stations 3 and 6 were more complex, with towers 4.5 m tall carrying thermocouples at the top and at 3.6, 2.4, and 1.5 m. There was also a grab sampler, gas sensor (TSI), and thermocouple at 2.4 m, and a separate grab sampler and thermocouple at 0.45 m.

Because the ground rises immediately downwind from the spill pond, stations 7 and 8 are 6 m above the pond surface. Stations 5 and 6 are 2.1 m, station 4 is 1.5 m, and station 3 is 0.6 m above the pond surface. Stations 1 and 2 are on the pond. The laser beam of the LIDAR was about 2 m above the surface of the pond.

Atmospheric transport modeling

In each experiment, we used the atmospheric transport model ATMAS[4] to aid in the analysis of the measured concentration data. The ATMAS code is a three-dimensional computer model that we developed to predict the transport of pollutants entering the atmosphere from extended or multiple sources. One of its major capabilities is that of simulating a three-dimensional, time-varying wind field from wind data obtained at different points within the region of interest. A time-varying wind-field model is much more realistic

Table 1 *A summary of the 5-m³ LNG spill tests at the Naval Weapons Center, China Lake, California.*

	Test number			
	1	2	3	4
Date	31 Aug	13 Sept	9 Nov	20 Nov
Time	14:56	19:37	15:26	15:11
Temperature, °C	35.8	21.1	26.8	20.1
Relative humidity, %	16	29	15	21
Spill volume, m^3	4.39	4.52	4.5	4.2
Spill duration, s	67	59	77	52
Source rate, kg/s	20.58	23.84	22.34	44.0 ($g/m^2 \cdot s$)
Spill radius, m	7.22	7.81	6.82	Variable
Boiloff time, s	90	80	85	120

than a constant-velocity wind model.

The ATMAS code models atmospheric transport by solving the advection–diffusion equation, which simply states that the concentration of pollutants at a given point is determined by two processes: diffusion (the simple spreading that takes place without wind and the effects of gusts and eddies) and advection (blowing away in the average wind). ATMAS uses the particle-in-cell method,[5] which represents the mass of the emitted pollutant by marker particles whose trajectories through space are calculated according to an equation of motion derived from the advection–diffusion equation. The array of marker particles gives us a graphic three-dimensional representation of the vapor plume.

To use the ATMAS code, we must specify boundary conditions, the characteristics of the LNG vapor source, and measured values of wind speed and direction as a function of time. For the first three experiments we assumed a constant emission rate from a round LNG pool of constant size for a fixed period of time. In the fourth test, because of significant changes in wind velocity, we found it necessary to simulate the way in which the LNG pool spreads over the water surface. We did this by turning on a series of vapor sources with successively larger areas from time to time during the calculation. The various 5-m^3 tests are described in Table 1.

The ATMAS code displays the results of its calculations in contour plots of gas concentration, graphs of concentration vs time for each of the measurement stations, and dot plots of marker-particle distributions. Figure 2 shows a contour plot of the concentration isopleths 1 m above the ground 60 s after the beginning of spill 1. The crosses indicate the measurement stations. In this experiment, the wind carried much of the plume to the left of the detector array.

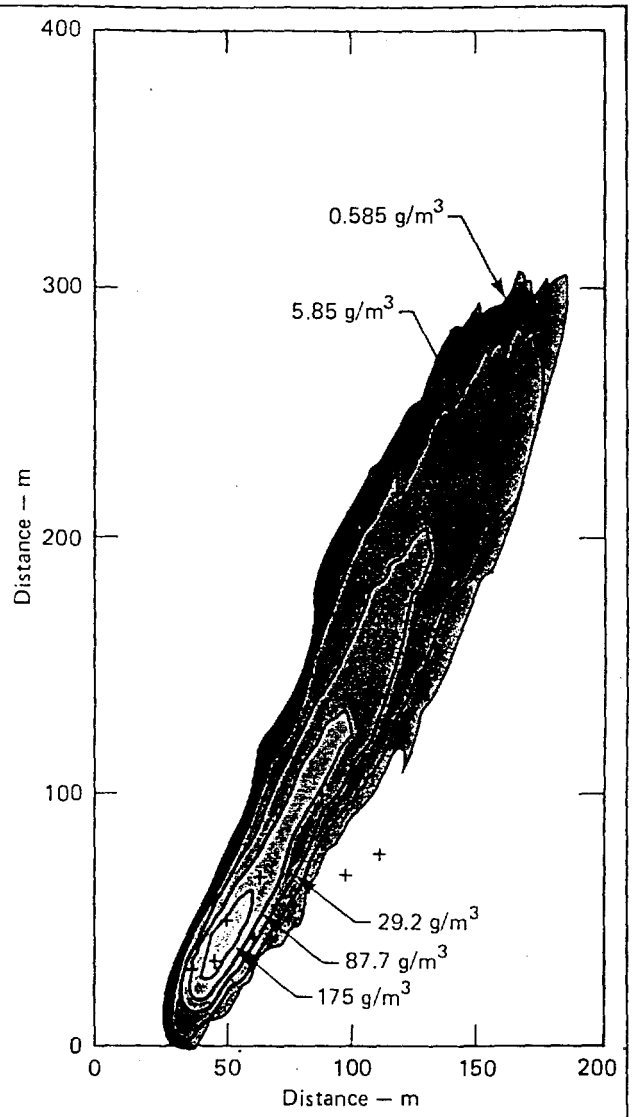


Fig. 2 *Concentration isopleths calculated by the ATMAS code for the vapor plume 60 s after the start of the first 5-m^3 spill experiment. The lower flammability limit (33 g/m^3 at 300 K) eventually extended to about 380 m.*

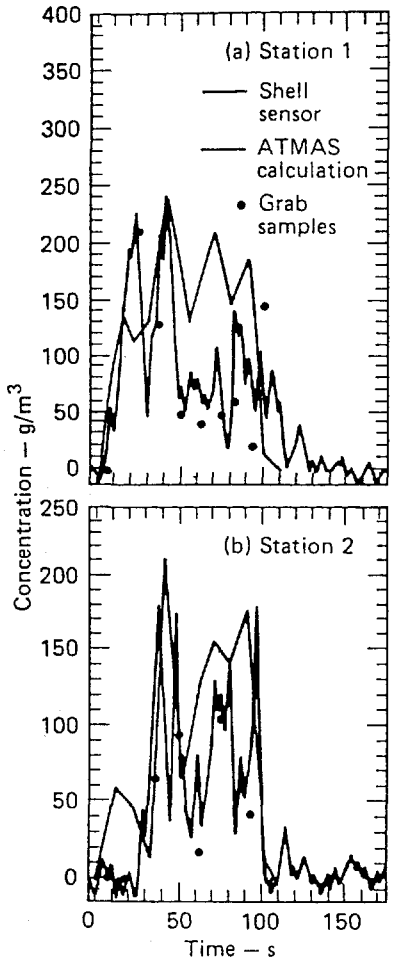


Fig. 3 *Comparison between ATMAS calculations and measured gas concentrations (a) at station 1 and (b) at station 2 in the first 5-m^3 spill experiment. Dots represent the results of mass-spectrometric analysis of grab samples taken by opening evacuated bottles at intervals during the experiment.*

Figure 3 shows the gas concentration data from stations 1 and 2 (the two stations inside the pond) for spill 1. The grab sample results (dots) agree very well with the Shell gas sensor output (black trace) in both cases, indicating that the sensors were working properly. The ATMAS calculation (colored line) agrees well, both in general and in detail, with the sensor output at station 2 (Fig. 3b). At station 1 (Fig. 3a) there was reasonably good general agreement. However, the detailed structure of the experimental data was not reproduced by ATMAS. This structure is probably due to local wind variations.

During spill 2 there was a shift in the wind direction at the far end of the detector array while the vapor cloud was dispersing. Figure 4 consists of three selected marker-particle plots from the ATMAS calculation (at 10, 50, and 90 s), showing the kink that this wind shift produced in the developing vapor plume. Although there is some uncertainty as to the exact cause of the wind shift, we believe that it was due to the presence of a hill immediately downwind from the spill pond.

The ATMAS code has significant deficiencies when applied to an LNG cloud dispersing in the atmosphere; for example, it completely neglects the effects of density and temperature variations within the cloud. However, it does attempt to simulate the time-varying wind fields encountered in the LNG spill experiments. We used this code to help overcome difficulties encountered in analyzing concentration data that were significantly affected by changes in the wind field and to evaluate the impact of these changes on plume movement.

Experimental results

The sensor array for the 5-m^3 spill experiments was designed to evaluate instruments and measurement techniques for larger spill experiments, not to reconstruct gas-cloud-concentration contours. However, we were interested in improving our computer modeling and analyzing the fragmentary data obtained to extract whatever new information it might contain on LNG boiloff and dispersion. We were able to estimate the dispersion coefficients through a point-to-point comparison with data from certain stations. We did this in the horizontal plane by repeated calculations, gradually increasing the horizontal dispersion coefficient until the gas just reached those stations that were at the edge of the plume. We found that the horizontal dispersion coefficient obtained in this way corresponded to stable conditions, while the atmosphere outside of the cold gas plume was unstable. (Unstable conditions imply a larger dispersion coefficient than do stable conditions.)

We determined the vertical dispersion coefficient in much the same way from data taken at two heights at stations 3 and 6. Vertical dispersion corresponded to stable conditions and was nearly identical

for all four spills even though the wind speed and atmospheric conditions varied considerably.

Figure 5 shows measurements made with the Anarad infrared sensor, the only instrument in the array that could distinguish among methane, ethane, and propane. These are the first field measurements ever made of differential boiloff of the various constituents of LNG. Differential boiloff is important because it affects the detonability of the vapor cloud. Detonating pure methane is difficult; detonating methane mixed with ethane or propane is much easier.

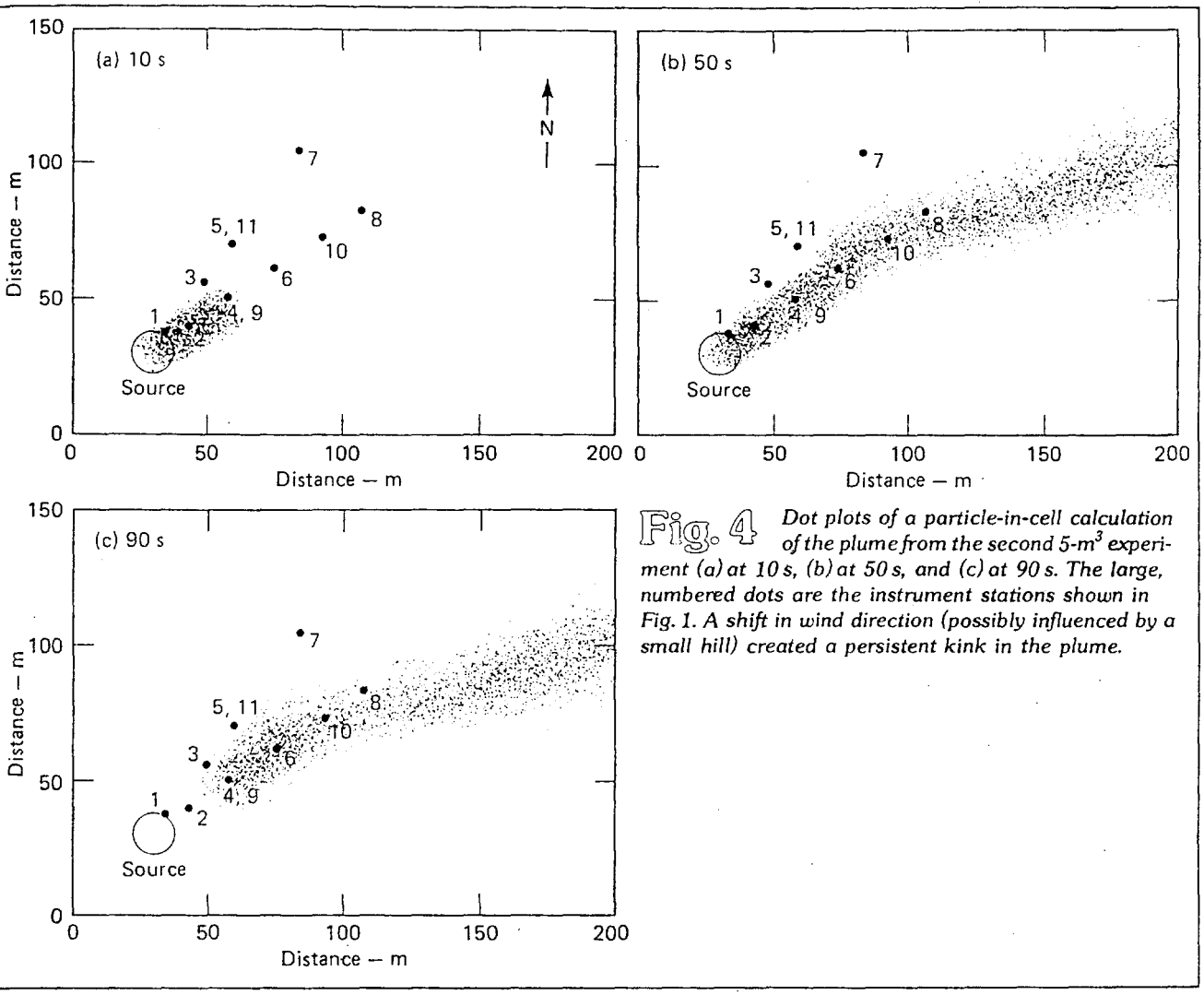


Fig. 4 *Dot plots of a particle-in-cell calculation of the plume from the second 5-m³ experiment (a) at 10 s, (b) at 50 s, and (c) at 90 s. The large, numbered dots are the instrument stations shown in Fig. 1. A shift in wind direction (possibly influenced by a small hill) created a persistent kink in the plume.*

We discovered experimentally the importance of wind variations in influencing both vapor drift downwind and the size of the LNG vapor cloud over the pond. This latter effect was large enough to significantly influence our calculations on spill 4. We had to estimate the vapor source size from photographs, since our other instrumentation was not designed to measure it.

Our data and calculations showed that the lower flammable limit of the vapor cloud (33 g/m^3) extended farther than expected and that the cloud was warmer than predicted. However, the code calculations were in reasonable agreement with the concentration data for the individual sampling stations. We believe that this agreement is due mainly to the code's ability to recreate the time and spatial variations in the wind field. To improve our predictive ability, we will need to consider the effects of the large density difference between the cold gas cloud and the air and to refine our methods of modeling the effects of terrain features such as hills and valleys.

Ongoing experiments

We are currently performing 40-m^3 spill experiments at China Lake with a much more extensive sensor array (Fig. 6). These tests began in June and will end in September. Because of the large number of stations involved, the large distances between the data-acquisition center and the stations, and the need to move stations during the experimental series, it has been impractical to link the instruments to the acquisition center with

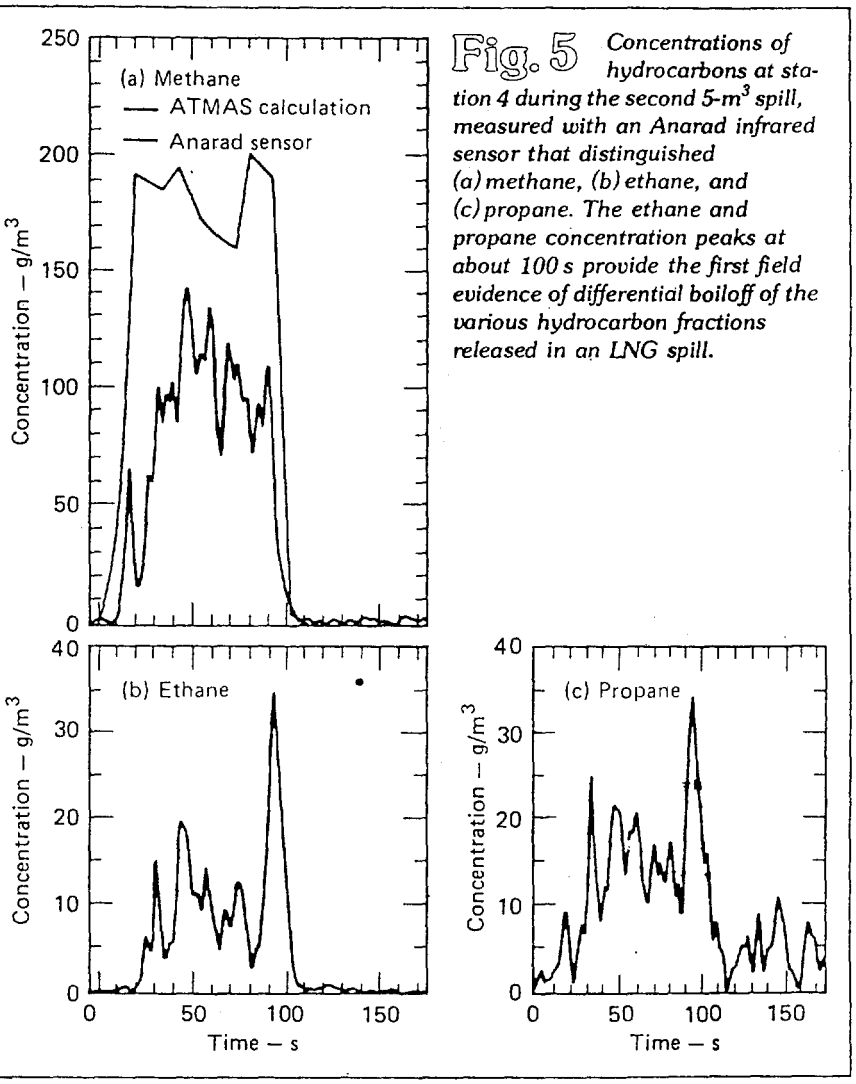


Fig. 5 *Concentrations of hydrocarbons at station 4 during the second 5-m^3 spill, measured with an Anarad infrared sensor that distinguished (a) methane, (b) ethane, and (c) propane. The ethane and propane concentration peaks at about 100 s provide the first field evidence of differential boiloff of the various hydrocarbon fractions released in an LNG spill.*

power and data cables. Instead, each station is self-contained (powered by batteries and solar panels) and connected to the center by two-way telemetry.

The greater extent of the sensor array imposes severe burdens on the instruments and the measurement stations. Both are light and portable (so that they can be tower-mounted and easily moved to accommodate shifts in the mean wind direction and changing experimental needs). The instruments require only minimum maintenance, are stable over periods of weeks, and are easy to calibrate.

The nature of the dispersing cloud affects instrument requirements. The cold gas condenses essentially all the water vapor in the air close to the pond, forming a thick fog (see front cover) that poses problems not only for optical instruments but also for sensors that rely upon the chemical or thermodynamic properties of the gas. Hence, each type of instrument must be specifically adapted to function properly within a cold, dense fog.

Because of turbulence, hydrocarbon concentrations within the gas cloud fluctuate significantly even over time intervals as short as 0.2 s. Turbulence and its effect on

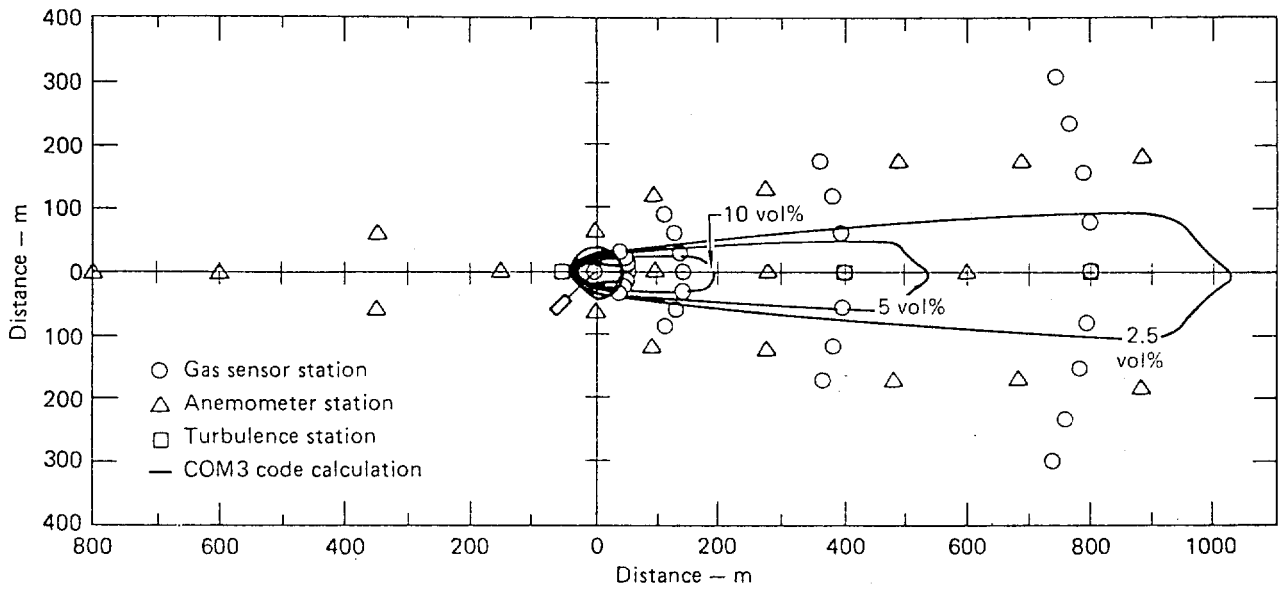


Fig. 6 *Far-field dispersion array for 40-m³ spill tests at China Lake, with calculated concentration contours for a 40-m³/min spill rate. Gas sensors, anemometers, and thermocouples are at elevations of 1, 3, and 8 m above the ground. Each of the instrument stations is self-contained (powered by batteries and solar cells) and connected to a central data-processing trailer by two-way telemetry. Each of the stations is portable so that the array can be rearranged in response to shifts in the average wind direction or changes in experimental conditions.*

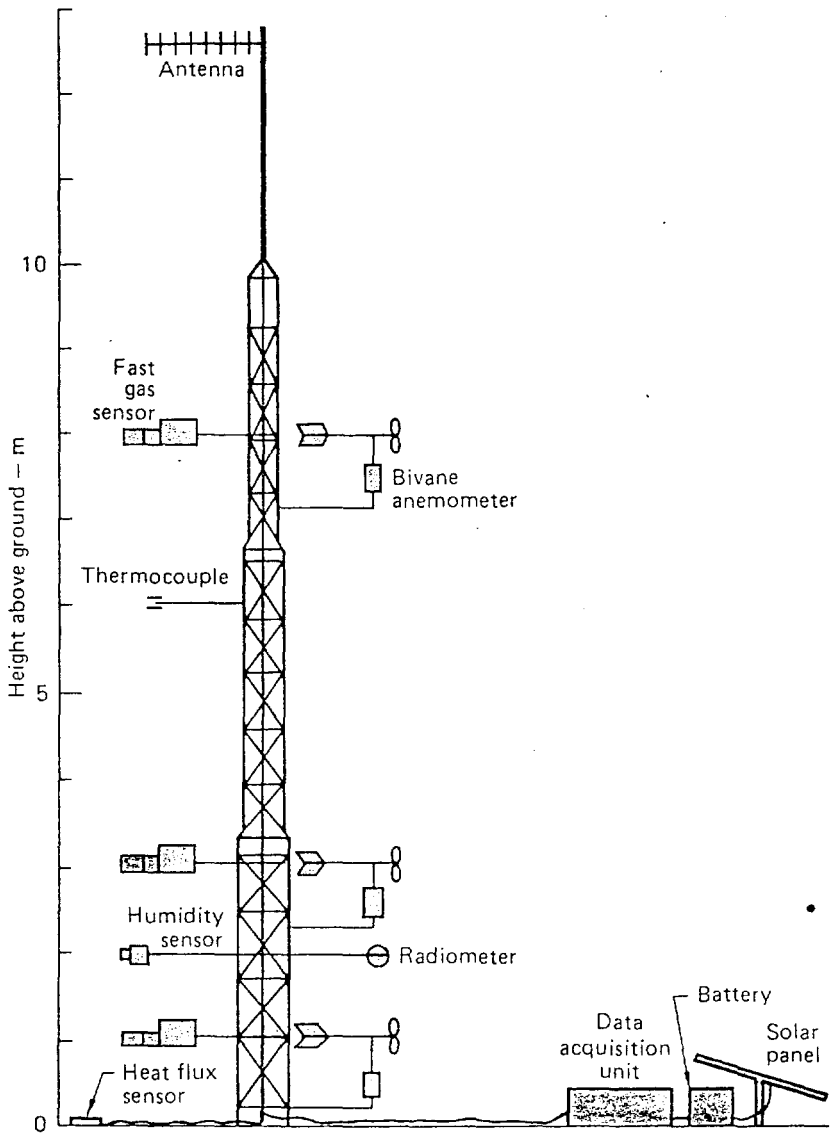


Fig. 7 *One of six turbulence stations being used in the 40-m³ spill experiments currently being performed at China Lake. The entire tower, excluding its base, weighs only about 22 kg and can easily be moved from one base to another by two men in about a half an hour. The gas stations are similar, except that they do not include the fast-response bivane anemometers.*

gas mixing and dispersion are major areas of interest in these experiments, and for this reason, we are setting up measurement stations (Fig. 7) specifically designed to measure these effects. We are equipping the turbulence stations with fast, infrared gas sensors, temperature sensors, and fast-response bivane anemometers positioned 1, 3, and 8 m above the ground. The development of a fast infrared gas sensor has been a major project for us this year.[6] This instrument, capable of measuring methane, ethane, and propane concentrations, is portable, fast (0.2 s cycle time), and able to operate in the fog. We developed it because no satisfactory commercial instruments were available.

In the 40-m³ tests, we are imaging the cloud from above by means of a helicopter-mounted infrared imaging system with optical filters centered on a band in the infrared strongly absorbed by methane. This allows us to trace the cloud downwind long after it has become invisible to the naked eye and will be of great help in reconstructing cloud movements for comparison with the computer simulation.

We plan 10 or more 40-m³ spills under a variety of wind-speed and spill-rate conditions. We would like to measure spills at very low wind speeds (1 to 2 m/s), but such conditions are rare at China Lake. When data from the current tests are analyzed, we should have enough information to proceed with the larger spills.

Summary

We have conducted a series of 5-m³ spills of LNG to evaluate gas

sensors and measurement techniques. As a result of these experiments, we now know what to measure in the cloud of dispersing natural-gas vapors and how to measure it. These tests have helped us to design a sensor array and data-acquisition system for 40-m^3 spill experiments now in progress. They have also permitted us to test our ATMAS dispersion code, to verify the importance of the code's variable-wind-field capability, and to pinpoint areas of needed improvement. These experiments provided us with the first field measurements of differential boiloff of the various components of LNG and also with preliminary estimates of the horizontal and vertical diffusion coefficients. We have learned that the lower flammability limit extends farther downwind than expected and that the vapor cloud is warmer than predicted.

In our current 40-m^3 spill experiments we are attempting to measure turbulent mixing in the gas cloud and to obtain enough gas-concentration data to generate isopleth plots for comparison with those generated by computer models. We are also attempting to measure heat transfer from various sources into the cloud as it disperses downwind. Aerial infrared imaging techniques permit us to track the methane cloud through the sensor array and beyond. All of these tests will prepare us for the larger-scale spill experiments (planned for 1982 and beyond), which will verify our computer models and help DOE to understand LNG-handling accidents and to set guidelines for minimizing adverse effects.

Key words: ATMAS; gas-concentration sensors; hydrocarbon fuels; infrared imaging; liquefied natural gas; LNG; wind-field model.

Notes and references

1. A brief discussion of LNG hazards and our plans to study them appeared in the September 1978 *Energy and Technology Review* (UCRL-52000-78-9), p. ii.
2. Studies of the detonability of methane and ethane mixtures were described in the May 1979 *Energy and Technology Review* (UCRL-52000-79-5), pp. 18–23.
3. G. E. Bingham, C. H. Gillespie, and J. H. McQuaid, *Development of a Miniature, Rapid-Response Carbon Dioxide Sensor*, Lawrence Livermore National Laboratory, UCRL-52440 (1978).
4. D. L. Ermak, R. A. Nyholm, and R. Lange, *ATMAS: A Three-Dimensional Atmospheric Transport Model to Treat Multiple Source Areas*, Lawrence Livermore National Laboratory, UCRL-52603 (1978).
5. R. Lange, "ADPIC–A Three-Dimensional Particle-in-Cell Model for the Dispersal of Atmospheric Pollutants and Its Comparison to Regional Tracer Studies," *App. Meteor.* **17**, 320–329 (1978).
6. The new sensor is described in the September 1980 *Energy and Technology Review* (UCRL-52000-80-9), pp. 36–41.

Past titles

Listed below are articles published over the last several months in the **Energy and Technology Review.** *For the most part, they are grouped in subject categories corresponding to the areas of responsibility of Assistant Secretaries in the U.S. Department of Energy. However, a few additional categories have been created for research funded by other agencies.*

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