High Radium-226 Concentrations in Public Water Supplies

JOHN L. S. HICKEY and SAMUEL D. CAMPBELL

TO IDENTIFY population groups consuming water with a relatively high radium content (more than 3.0 picocuries per liter), selected public water supplies were analyzed for radium-226. (The term "radium" indicates radium-226 unless otherwise specified.) The principal objective of the study was to identify compact populations which could serve as a base for epidemiologic studies of the effects of chronic radium ingestion at low levels.

Marinelli (*1*) pointed out earlier the possibilities of using such populations to study the effects of radium ingestion. One such study by Petersen and co-workers (*2*) attemped to relate the mortality rates from bone neoplasms to the level of radium in drinking water for 111 midwestern cities. These authors reported that a retrospective analysis of death certificates indicated that the mortality rate from bone neoplasms in a population ingesting an average of 4.7 picocuries (pCi) of radium per liter from its water supplies was higher than in a matched control group with almost no radium in its water supplies. Although their study included a population of 900,000, few cases of bone neoplasm were found because incidence of this disease is very low. The small observed differences in the mortality rates between the two groups were therefore not statistically significant. Before a difference of the magnitude found by Petersen and co-workers could be considered significant, many more cases would be needed for study; hence, a larger study population or many more years of observation of the same population would be required.

Mr. Hickey, who is now in the Office of Research and Development of the Bureau of Disease Prevention and Environmental Control, Public Health Service, at the time of the study was deputy chief for operations of the Environmental Surveillance and Control Program, National Center for Radiological Health. Mr. Campbell is a staff officer in that program.

Petersen and co-workers (*2*) concluded that the identification of additional population groups exposed to relatively high radium concentrations offered the greatest hope for a more meaningful study. They observed that the feasibility of such a study might be limited by the lack of a sufficiently large exposed population. The National Center for Radiological Health undertook the present study to identify additional population groups who might be exposed to relatively high radium concentrations in their drinking water. Also, the study was viewed as a means of locating water supplies that would require corrective action should epidemiologic studies show that low-level radium ingestion is harmful to human beings.

Selection of Water Supplies

There are more than 19,000 community water supplies in the United States, serving more than 150 million people (*3*). The laboratory facilities available for the present study permitted radium analyses of only approximately 1,000 water samples. It was therefore necessary to select those supplies most likely to contain relatively high amounts of radium.

A literature search was made to locate geographic areas with evidence of relatively high radium levels in water (*4–10*). Smith and coworkers (*6*) reported mean values of up to 160 pCi of radium per liter in some Maine well waters and up to 8.4 pCi per liter in some New Hampshire well waters. Scott (*8*) found radium levels exceeding 3.3 pCi per liter in a large number of fresh water wells in several midwest and Rocky Mountain States and in Kansas and Texas. Lucas (*5*) found that approximately 37,000 persons were consuming water containing more than 10 pCi of radium per liter in 21 communities in Illinois, Iowa, and Missouri. Ground waters in Illinois and Iowa have been sampled extensively by several investigators, who reported elevated radium levels in many municipal well water supplies in these States (*2*, *5*, *7*, *8*, *10*).

Analyses of the water supplies (*9*) of the 100 largest cities in the United States, serving 60 million people, showed that only three of these supplies contained more than 1 pCi of radium per liter; the supply of Rockford, Ill., had 2.5 pCi per liter; that of Houston, Tex., 1.3 pCi per liter; and that of Lubbock, Tex., 1.9 pCi per liter. Several State health departments provided data on radium or alpha levels in ground water; the U.S. Geological Survey provided additional data (unpublished report by R. C. Scott entitled "Known and suspected high-radium aquifers in the United States, March 1966).

With the foregoing information as a guide, 20 States were selected in which to sample water supplies for radium content (see map). Illinois, Iowa, and Wisconsin were not included in the study because the well water supplies in these States had already been systematically sampled by others. Because radium levels in well water have been reported to vary over time (*10*), several water supplies (principally in Minnesota) with previously reported high radium or alpha levels were sampled to determine if these levels still existed. Although there was no history of high radium levels in water in Alaska and Hawaii, these States were included on the presumption that some unusual radium levels might be found as a result of such features as volcanic and glacial activity.

Within the defined geographic area, three basic criteria were used in selecting water supplies for radium analysis.

1. Only ground water supplies were selected. Other investigators (*4*, *5*, *7*) have shown that surface waters are unlikely to contain significant amounts of naturally occurring radium.

2. Supplies receiving aeration, flocculation, settling, softening, or a combination of these, were not selected. Some types of water treatment have been shown to remove the bulk of the radium (*4*, *7*, *10*). Since the principal interest was in water with high radium levels at the point of consumption by the population, treated supplies were not sampled.

3. Supplies serving fewer than 1,000 persons were usually not selected. The 7,850 ground water supplies in this classification serve only 10 percent of the population that consumes ground water (*3*), and omission of these supplies permitted a broader population coverage with the available analytical facilities. However, 80 water supplies serving fewer than 1,000 persons were sampled in several areas having particular evidence of elevated radium levels in ground water.

Information was obtained for each community on the source of water, the treatment it received, and the population it served, principally from the 1963 inventory of municipal water facilities (*11*). Both Lucas (*7*) and Scott (*8*) have pointed out that aquifers in certain types of geologic formations are more likely to contain radium than those in other formations. However, there was not sufficient geologic information readily available on specific water supplies to justify using this factor as one of the criteria for selection.

In areas where several water supplies suitable for study were clustered, only one or two were selected in order to obtain broader geographic coverage with the limited laboratory facilities. If any of the supplies selected contained elevated radium levels, it was assumed that the nearby supplies could be more knowledgeably sampled in the future.

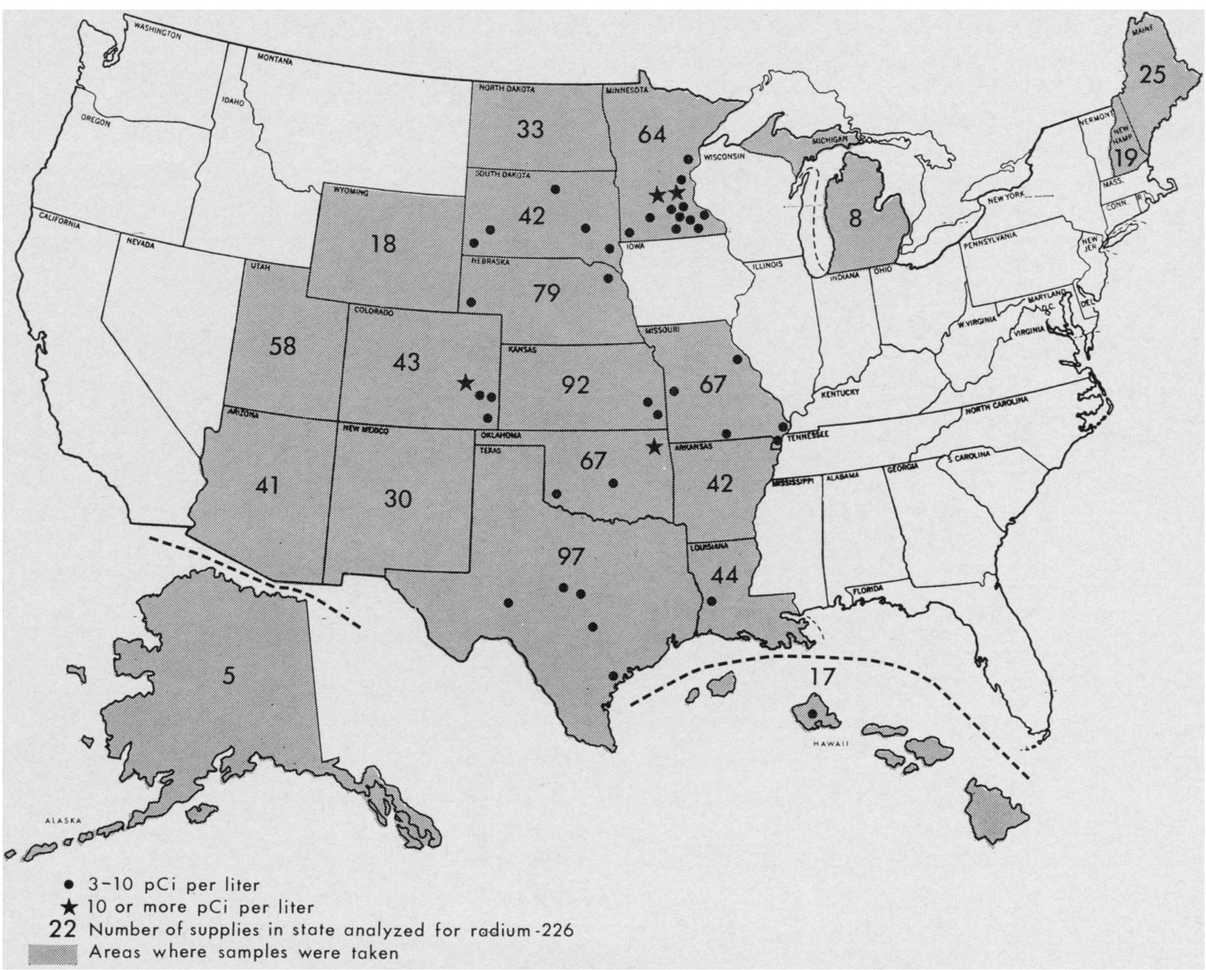
Sample Collection

Before the sampling, permission to make the study was obtained from the 20 State health departments involved, through the Public Health

Service Regional Offices. State health departments were notified several days in advance when and where samples would be taken. Collapsible plastic containers and return-addressed mailing cartons were mailed directly to the superintendent or manager of each city or community water department. Each envelope contained a request that a water sample be collected and instructions for its collection and mailing. Samples were numbered with the post office ZIP code of the community served.

The requests for water samples were mailed to the 976 communities selected for radium analysis over a period of several weeks in May and June of 1966. Within a few weeks of the requests, 923 samples were received at the laboratory—a response of more than 94 percent. A few of the first samples received did not contain enough water for analysis because the plastic containers had not been expanded sufficiently before filling, and the samples had to be discarded. Brief illustrated instructions were inserted in later requests for samples, thereby eliminating this problem. Of the 923 samples received, 32 could not be analyzed because of insufficient volume or loss of the sample through laboratory error. In one State, the health department selected the communities to be sampled; in two States, the health departments collected and mailed the samples.

Upon completion of the analyses, sample results were sent to each State health department (through the Public Health Service Regional Offices) with a request that the department notify the communities of the results. It was anticipated that questions would arise



Radium-226 in public ground water supplies, 1966

regarding interpretation of the results. Therefore, concurrently with the study, a digest was prepared of various protection standards and State regulations for radium in water (*12*). Reprints of this digest accompanied sample results sent to the States.

Results of Sample Analysis

A total of 891 water supplies serving 3,864,500 people were analyzed for radium. Seventy-two water supplies, serving 329,000 people, were found to contain from 1 to 3 pCi of radium per liter. Forty-one water supplies, serving 144,000 people in 10 States, contained 3 or more pCi of radium per liter. Four of the 41 water supplies, serving 3,000 people, contained 10 or more. The highest radium level found was 24.1 pCi per liter in a supply serving 360 people. The remaining 778 samples contained less than 1 pCi of radium per liter of water. These results are summarized in tables 1 and 2. The locations of the water supplies having 3 or more pCi of radium per liter (including the positive confidence interval) are shown in the map. The number of samples analyzed from each State is also shown.

Analytical Methods

Radium analysis was performed under contract during 1966–67 by the New Mexico Department of Public Health. The majority of the samples were radiochemically analyzed for radium-226 using published methods with minor modifications (*13*). The radium-226 recovery, based on 26 standards of radium-226 (1.0 pCi), was 95±15 percent at the 95 percent confidence level. Early in the study, the laboratory staff had attempted to analyze samples for both radium-226 and radium-228 simultaneously by combining methods described by Petrow (*14*) and others (*13*). Analysis for radium-228, however, proved too time consuming for the period allowed for this project, and it was abandoned. The radium-226 recovery with the combined method was 80±20 percent at the 95 percent confidence level.

The mean recovery values found for radium-

Table 1. Summary of radium-226 analyses of public water supplies, 1966

State	Number of water supplies analyzed	Total population served 1963 [1]	Supplies with more than 3 pCi Ra-226 per liter		Highest Ra-226 level observed (pCi per liter)
			Number	Population served 1963 [1]	
Alaska	5	9, 840	0	0	0. 1
Arizona	41	241, 780	0	0	2. 0
Arkansas	42	178, 365	0	0	1. 4
Colorado	43	106, 750	4	3, 110	15. 7
Hawaii	17	427, 780	1	16, 000	8. 9
Kansas	92	286, 840	2	1, 130	5. 0
Louisiana	44	299, 865	1	9, 000	4. 0
Maine	25	73, 920	0	0	. 3
Michigan	8	115, 300	0	0	. 9
Minnesota	64	304, 200	13	46, 175	24. 1
Missouri	67	175, 030	5	17, 815	8. 7
Nebraska	79	225, 820	2	9, 660	3. 8
New Hampshire	19	59, 575	0	0	2. 5
New Mexico	30	240, 880	0	0	2. 3
North Dakota	33	60, 740	0	0	. 7
Oklahoma	67	202, 725	3	6, 830	10. 3
South Dakota	42	88, 930	5	11, 550	6. 9
Texas	97	539, 590	5	23, 100	9. 7
Utah	58	142, 705	0	0	. 9
Wyoming	18	83, 865	0	0	. 8
Total	891	3, 864, 500	41	144, 370	----------

[1] SOURCE: reference 3.

Table 2. High radium-226 concentrations observed in public water supplies, 1966

State and community	Population served, 1963 [1]	Ra-226 (pCi per liter)	±95 percent confidence level [2]
Colorado			
Granada	600	3. 14	0. 07
Holly	1, 110	5. 65	. 06
Walsh	1, 000	3. 00	. 07
Wiley	400	15. 71	. 15
Wahiawa, Hawaii	16, 000	8. 90	. 08
Kansas			
Mulberry	750	[3] 2. 98	. 05
Walnut	380	4. 97	. 06
DeRidder, La	9, 000	3. 95	. 08
Minnesota			
Anoka	10, 560	7. 38	. 10
Claremont	465	9. 83	. 12
Courtland	240	4. 82	. 09
Faribault	16, 925	3. 19	. 07
Hardwick	330	4. 24	. 08
Howard Lake	1, 005	15. 20	. 15
Janesville	1, 425	7. 02	. 09
Jordan	1, 480	7. 12	. 11
Montrose	360	24. 11	. 20
New Market	210	8. 83	. 12
Red Wing	10, 530	4. 65	. 08
Sandstone	1, 550	4. 05	. 08
Savage	1, 095	9. 60	. 12
Missouri			
Charleston	5, 600	4. 04	. 06
Clarkton	1, 440	5. 64	. 08
Rich Hill	1, 555	8. 67	. 10
Troy	1, 650	3. 47	. 06
West Plains	7, 570	4. 83	. 07
Nebraska			
Kimball	5, 160	3. 84	. 07
Wayne	4, 500	3. 34	. 06
Oklahoma			
Afton	1, 250	10. 29	. 12
Mangum	4, 200	7. 35	. 11
Maud	1, 380	5. 24	. 09
South Dakota			
Canton	2, 500	6. 93	. 11
Edgemont	1, 770	5. 33	. 10
Hot Springs	4, 950	4. 82	. 09
Howard	1, 200	3. 52	. 08
Ipswich	1, 130	3. 02	. 07
Texas			
Big Lake	2, 750	9. 71	. 12
Brady	5, 350	7. 72	. 10
Eden	1, 600	6. 21	. 10
Kerrville	12, 000	3. 73	. 07
Sea Drift	1, 400	3. 45	. 07

[1] SOURCE: reference 3.
[2] 95 percent confidence level based on counting errors only.
[3] Included because confidence limit range includes 3.0.

226 were used in calculating the radium levels reported. Samples for radium-226 analysis were counted for 20 hours. Counting errors were calculated, and the 95 percent confidence limits are reported individually for those samples appearing in table 2.

Discussion

Unpublished information provided by several State health departments and the U.S. Geological Survey was particularly helpful in selecting sampling locations. Two of the four samples showing more than 10 pCi of radium per liter (table 2) confirmed results that the Minnesota Department of Health had previously reported for these water supplies. Most of the other water supplies showing 3 or more pCi per liter are located in or near areas with previously reported elevated radium or alpha activity in well waters. Since sampling was generally limited to States having a previous history of elevated radium levels, this result was expected.

Seven widely scattered water supplies in South Dakota and Nebraska showed levels of radium of more than 3 pCi per liter; only two of these, in the southwest corner of South Dakota, were in areas from which elevated radium levels had previously been reported (*8*). Samples were collected from 44 well water supplies in Maine and New Hampshire; most of these sources were within 50 miles of wells previously reported to have high radium levels (*6*). However, the highest radium levels found in these 44 water supplies were 0.3 pCi per liter in Maine and 2.5 in New Hampshire.

In southern Missouri, 13 water supplies were found to contain more than 1 pCi of radium per liter; in addition, 15 other supplies in this area had previously been reported by Lucas (*5*) to contain more than 1 pCi per liter. These results support Scott's postulation (*8*) that the known high-radium formations of the upper Mississippi valley may extend through southern Missouri. Therefore, this locality may be considered as a promising area for more extensive sampling of ground water supplies for radium.

The next step toward identification of additional populations ingesting high levels of radium from water would be a sampling program for previously unsampled water supplies in the vicinity of the high radium water supplies observed in this study. This step was not possible in this study because of the limited time.

Only those water supplies found to contain 3 or more pCi of radium per liter are named in table 2. This level was used in Petersen's study (*2*) as the lower limit of radium ingestion from drinking water in defining the "exposed" population. A large number of the water supplies sampled showed virtually zero radium content, and the communities they serve may be suitable control populations in studies of the effects of radium ingestion. Sampling results for all communities included in the study are available from the authors through the National Center for Radiological Health.

The sampling instructions requested that samples be collected on the water distribution systems but not from fire hydrants, deadends, or water taps on which home softeners were in use. This request was made so that the samples would be representative of the water consumed by the population. Because of the previously observed variable levels of radium within a water system (*2*), several samples from various points on a water supply should be analyzed for radium before a community is selected for an epidemiologic study of the health effects of low-level radium ingestion.

The sample containers and the general method for collecting the samples and getting them to the laboratory proved satisfactory. A large degree of credit should be given to the State health departments for their cooperation in notifying the communities to be sampled in advance and for generally maintaining an overall rapport with their communities, which made the individual water plant managers willing to take the time to collect and mail a sample promptly. This State-city relationship is believed to have been responsible for the high response.

Summary

To identify population groups in the United States having relatively high chronic intakes of radium from drinking water, samples of approximately 900 public ground water supplies serving 3.8 million people in 20 States were analyzed in 1966–67.

After the National Center for Radiological Health, Public Health Service, had arranged for the water sampling with the health departments of the 20 States, personnel of these departments or of community water departments collected 1-liter samples of the selected water supplies and mailed them to the laboratory performing the radium analyses. The response to the request for water samples was more than 94 percent.

Forty-one water supplies, serving 144,000 people in 10 States, were found to have 3 or more pCi (picocuries) of radium-226 per liter. Four supplies, serving 3,000 people, contained 10 or more pCi per liter. The highest concentration observed was 24.1 pCi per liter in a supply serving 360 people.

REFERENCES

(*1*) Marinelli, L. D.: Radioactivity and the human skeleton. Amer J Roentgen: 80: 729–739 (1958).

(*2*) Petersen, N. J., Samuels, L. D., Lucas, H. F., and Abrahams, S. P.: An epidemiologic approach to the problem of low-level radium 226 exposure. Public Health Rep 81: 805–814, September 1966.

(*3*) U.S. Public Health Service: Statistical summary of municipal water facilities in the United States, January 1, 1963. PHS Publication No. 1039. U.S. Government Printing Office, Washington, D.C., 1965.

(*4*) Hursh, J. B.: Radium content of public water supplies. J Amer Water Works Assoc 46: 43–54 (1954).

(*5*) Lucas, H. F., Jr.: Study of radium-226 content of midwest water supplies. Radiol Health Data 2: 400–401 (1961).

(*6*) Smith, B. M., Grune, W. N., Higgins, F. B., Jr., and Terrill, J. G., Jr.: Natural radioactivity in ground water supplies in Maine and New Hampshire. J Amer Water Works Assoc 53: 75–88 (1961).

(*7*) Lucas, J. F., Jr., and Ilcewicz, F. H.: Natural radium-226 content of Illinois water supplies. J Amer Water Works Assoc 50: 1523–1532 (1958).

(*8*) Scott, R. C.: Radium in natural waters in the United States. Radioecology. Proceedings of the first national symposium on radioecology, 1961. Reinhold Publishing Corp., New York City, 1963.

(*9*) Durfor, C. N., and Becker, E.: Public water supplies of the 100 largest cities in the United States, 1962. U.S. Geological Survey Water Supply Paper 1812. U.S. Department of the Interior. U.S. Government Printing Office, Washington, D.C., 1964.

(*10*) Ball, A. D.: Protocol for proposed plan of study of the causes of variability in radium concen-

tration in municipal water systems. State Hygienic Laboratory, State University of Iowa, Iowa City, September 1964.
(*11*) U.S. Public Health Service: Municipal Water Facilities, 1963 Inventory. PHS Publication No. 775. U.S. Government Printing Office, Washington, D.C., 1964, vols. 1–9.
(*12*) Hickey, J. L. S.: Digest of protection standards and State regulations for radioactivity in water. Radiol Health Data Rep 7: 549–554 (1966).
(*13*) Radionuclide analysis of environmental samples. A laboratory manual of methodology. Robert A. Taft Sanitary Engineering Center Technical Report R59–6. U.S. Public Health Service, Cincinnati, Ohio, 1966, pp. 42–56.
(*14*) Petrow, H. G., Cover, A., Schiessle, W., and Parsons, E.: Radiochemical determination of radium-228 and thorium-228 in biological and mineral samples. Anal Chem 36: 1600–1603 (1964).

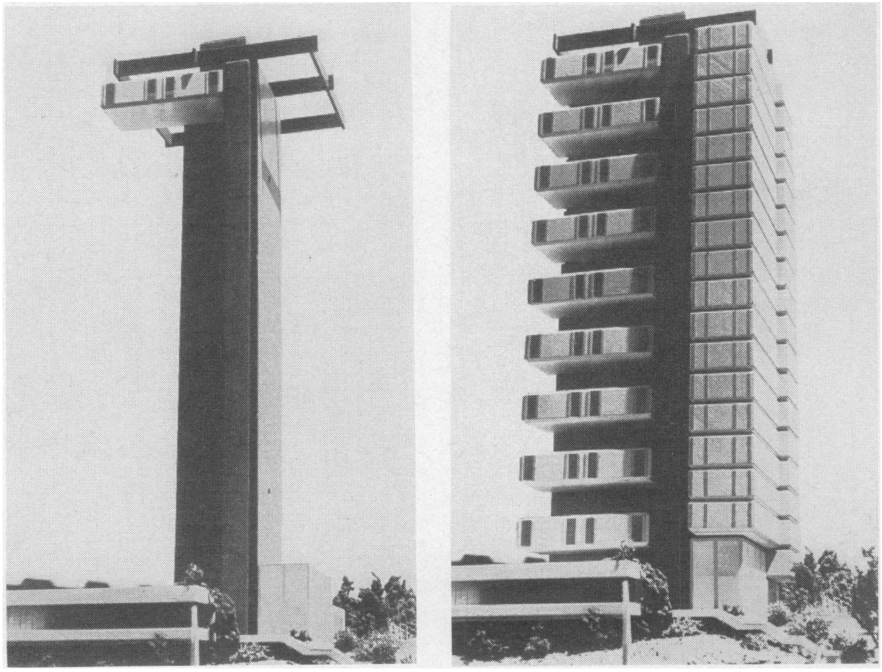
Sodium Chlorate—A Fire Hazard

Persons who use a weed killer and defoliant known as sodium chlorate, used mainly in southern States and California, may be seriously burned by clothing fires unless they take proper precautions.

The chemical is used to kill a weed known as Johnson grass and as a defoliant by cotton growers. The danger of misusing sodium chlorate was reported to the Public Health Service by investigators in a State health project in southeast Missouri to prevent burns. The project grant is directed by the Injury Control Program of the National Center for Urban and Industrial Health in Cincinnati. Project investigators have been studying burns from various causes in the counties of New Madrid, Pemiscot, Butler, Stoddard, Scott, and Mississippi.

Sodium chlorate may be made less dangerous by mixing it with fire retardant solutions such as sodium metaborate. Sodium chlorate is sold in many compounds containing varying strengths of the chemical and under many trade names. The fire hazard varies with the strength of the solution. Users of the spray should remove any clothing contaminated by the spray and have it laundered as soon as possible. The directions on the container should be read before the chemical is used.

Sodium chlorate itself will not burn and is moderately toxic to a human being; however, when heated the material gives off oxygen, adding greatly to the flammability of any combustible material present. In one incident a child carried pellets of sodium chlorate in his pocket, and friction between materials in the pocket and the pellets ignited his trousers. When used as a spray, any clothing that comes in contact with the chemical becomes highly flammable.



Encapsulated building. Before and after

Left: before, the tower which contains elevators, stairs, and utility lines. Right: after, with alternate capsules to demonstrate varying patterns that can be achieved in design.

Experiment in Hospital Construction

Instant modernization and reduction in building costs are among the ultimate objectives of a research and experimental construction project at Michigan State University, East Lansing, developed by architect Christian Frey, consultant to Suspended Structures, Inc., in collaboration with Lester Gorsline Associates of San Francisco.

The university has received a Public Health Service grant of $437,572 for the construction of an addition to the Olin Health Center Hospital, the health care unit for students.

The structure will be built from modular subunits, complete with finishes and mechanical services. These units which may be constructed onsite or offsite in assembly line fashion could result in considerable reduction in building costs.

The subunits, or rooms, will be lifted into place and attached to a permanent vertical support structure. The only onsite construction necessary would be the reinforced concrete shaft or towers containing elevators, stairs, and utility lines.

The rooms and hallways will be suspended from and plugged into the utility services in the support towers, permitting erection of a large building in far less time than is now required.

Under the proposed project, the sponsor will test the feasibility of changing the use of modules from time to time. This new use may require complete revision of the mechanical and electrical services, which could be accomplished easily without demolition or major alteration of the structure. Moreover, activities in adjoining areas would not have to be suspended during the changeover period.

If the project is successful, this suspended structural system of construction could lead to even greater innovation and versatility in the construction of health facilities and could prove a boon to the hospital field in light of the mounting construction needs throughout the country.

Construction based on some of these new design principles may make it possible to replace quickly entire sections of a structure whenever modernization is needed.