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# REMOVAL OF PLUTONIUM FROM DRINKING WATER BY COMMUNITY WATER TREATMENT FACILITIES

by

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## ABSTRACT

REMOVAL OF PLUTONIUM FROM DRINKING WATER
BY COMMUNITY WATER TREATMENT FACILITIES

Plutonium removal factors (RF) averaged 14 ±10 during a study of the effectiveness of three drinking-water treatment plants for removing plutonium from Savannah River water. Plutonium concentrations between 0.1 and 3.5 fCi/l** were measured in raw and finished water samples. From 50 to 10,000 liter samples of water were concentrated by ion exchange techniques and processed to determine the concentrations of plutonium-239, 240 and to derive plutonium RF's. The similarity between RF's observed for both plutonium and suspended solids suggests a colloidal behavior for plutonium. Plutonium RF's may be limited by low-level buildup on the treatment facility filters and subsequent bleeding into finished water, and thus may be higher during abnormal plutonium releases to the environment. Flocculation and filtration appear to be the primary factors in the water treatment process contributing to plutonium removal. The similarity between the plutonium contents of finished water from treatment facilities upstream and downstream of the Savannah River Plant (SRP) indicates that there is no measurable dose-to-man from SRP plutonium releases in the water. The 70-year bone dose commitment to an individual from consumption for one year of 1.65 liters per day of treated Savannah River water, based on the plutonium concentrations of finished waters from the three treatment facilities is $5 \times 10^{-5}$ mrem.

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** 1 fCi = 1 *femto* curie = $1 \times 10^{-15}$ curie.

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## INTRODUCTION

As part of an assessment of population exposures and potential health consequences from transuranics released to the environment, a study of the effectiveness of community water treatment facilities for the removal of plutonium in drinking water was made. Very little information on plutonium concentrations in drinking water has previously been available to allow derivation of such removal factors (RF) [1,2,3]. The extremely low concentrations of plutonium in drinking water and surface water require the concentration of large quantities of water. Ion exchange concentration techniques recently developed at the Savannah River Laboratory now permit the determination of plutonium concentrations of <1 fCi/liter, far below the heretofore routine environmental monitoring detection limits of 250 fCi/liter and the maximum permissible concentration in drinking water of $5 \times 10^6$ fCi/liter. The measured values of plutonium in the Savannah River are consistent with earlier calculated estimates of plutonium in the Savannah River derived from SRP effluents [4]. Determinations of plutonium concentrations in raw and finished water samples collected at three water treatment facilities over a period of nine months provided the necessary data to derive a working RF for plutonium across typical water treatment facilities.

## EXPERIMENTAL

Three water treatment facilities that use Savannah River water [one above and two below the Savannah River Plant (SRP)] were selected for plutonium removal studies. Their locations relative to SRP are shown in Fig. 1. The North Augusta, S. C., and Beaufort, S. C., facilities are used for municipal water supplies, whereas the Savannah, Ga., facility is primarily for industrial usage. The water treatment processes used in all three facilities are compared in Fig. 2. The processes are quite similar, varying only slightly in their methods of mixing, settling, and filtration. Flow rates for water treatment facilities are shown in Table I. Although the process times are similar, the throughput of the Savannah Industrial water treatment facility is a factor of 10 greater than the average of the two municipal facilities.

Samples of raw and finished water were collected over a period of nine months at the intake and output locations in the plants. An alternating cation-anion exchange column was used to concentrate the plutonium [5]. Volumes of water in excess of 10,000 liters were concentrated in the water treatment plants, and additional samples of 50 liters were concentrated in the laboratory under more closely controlled conditions. The on-line columns used at the water treatment plant sites are designed to equilibrate with the plutonium in large volumes (10,000 liters) of raw and finished water providing a sensitive method for directly observing a removal factor across the process over several weeks. The 50-liter laboratory samples were analyzed by spiking with $^{236}$Pu tracer, allowed to equilibrate, after which time plutonium content was quantitatively determined by ion exchange concentration, chemical separation, and ultra low-level alpha spectrometry [6,7].

## RESULTS AND DISCUSSION

### Plutonium Concentration in Water

Table II lists the mean concentrations of plutonium-239,240 in raw and finished water during the study period. Plutonium concentration of the water at the intake to the water treatment facility upstream from SRP is typical of the few concentrations in surface water reported elsewhere in the United States [2,8]. Concentrations of $^{239,240}$Pu observed in raw water downstream of SRP are higher by a factor of 2 to 6 due to low-level releases from SRP, yet these concentrations are still a factor of $10^6$ less than the maximum permissible concentration in drinking water. The lower concentration of plutonium in raw water at the Beaufort facility, in comparison to the Savannah facility, is explained by the large dilution of the actual Savannah River water transported through a 45-km canal by surface runoff from a swampy area. A similar reduction in concentration at the Beaufort facility has also been observed in tritium measurements. The plutonium-239, 240 concentrations in the finished water from all the facilities are very similar, 0.08 fCi/l, and do not reflect the higher mean concentration observed in the raw water used by the Savannah facility.

### Plutonium Removal Factors (RF's)

Table III shows the RF values derived from the measured plutonium concentrations in the raw and finished water. Although a relatively wide variation exists, the overall RF, 14 ±10, for water treatment facilities is low. The low RF found in this study compares with similar low RF's in the range of 1-12 derived from published measurements made by Rocky Flats Plant [1] and Argonne National Laboratory [2] investigators. According to Table III, there is no significant difference in the RF values determined for the large-volume samples and those determined for the 50-liter spiked samples. The variation observed in the RF values at a single location is attributed to the buildup of plutonium on the filters in the water treatment facilities. Although the filters are backwashed following loss of head pressure or every few days, fluctuations in plutonium concentrations of materials trapped in these filters cause some of the plutonium to bleed off into the finished water. This bleed off suggests th t measured RF's would be higher during abnormally high plutonium concentrations in the raw water supply. Plutonium concentrations measured in filter media from the various facilities are shown in Table IV.

## Colloidal Plutonium Behavior

In the pH range of natural waters, colloidal plutonium hydroxide forms from polymeric hydrolysis products which age slowly [9]. A wide variety of colloidal hydroxide particle sizes containing plutonium result and are the dominant plutonium species likely to be encountered in natural water [10]. The effect of the water treatment process on this colloidal plutonium hydroxide form depends on colloidal size, pH, and chemical additives. Table V shows a comparison of the water quality of the raw and finished water from the three water treatment facilities. The pH range is ideal for plutonium colloid formation, and the addition of carbonates through lime and soda ash contribute to their coagulation [11]. The mean suspended solids RF of 5 observed for all three facilities is similar to the measured plutonium RF. This similarity supports flocculation and filtration as the primary factors in the water treatment process contributing to plutonium removal. Slight changes in chemical additives during water treatment show minor differences in the water quality between the industrial supply and the municipal supplies. However, no differences in finished-water plutonium concentrations were observed as a result of chemical treatment. A higher pH of the finished water was measured for the industrial facility, and a reduction in chemical oxygen demand was observed resulting from the absence of a carbon type filter bed. Phosphates are also low since no phosphate chemicals are added to the industrial supply.

## 70-Year Bone Dose Commitment to Man

The 70-year bone dose commitment to individuals drinking Savannah River water was calculated by using ICRP recommendations, Publication 2, for the mean concentrations measured in the raw and finished water from each of the water treatment facilities. The 70-year individual bone dose commitment from one year of drinking 1.65 liters per day of treated water is 0.00005 mrem/person (Table VI). The close agreement in the plutonium concentrations in the finished water upstream and downstream show that there is no measurable dose-to-man from Savannah River drinking water by SRP plutonium releases.

## ACKNOWLEDGEMENTS

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TABLE I. WATER TREATMENT FACILITY THROUGHPUTS

| | *Water treated, $10^6$ liters/day* | *Process Time, hours* |
|---|---|---|
| Savannah Industrial | 132 | 3.5-5 |
| Beaufort Municipal | 20 | 2-3 |
| North Augusta Municipal | 6 | 4 |

TABLE II. AVERAGE PLUTONIUM CONCENTRATIONS IN RAW AND FINISHED WATER[a]

| | $^{239,240}Pu$, *fCi/l* | |
|---|---|---|
| | *Raw Water* | *Finished Water* |
| Savannah Industrial | 2.25 ±0.89 | 0.09 ±0.01 |
| Beaufort Municipal | 0.99 ±0.32 | 0.07 ±0.07 |
| North Augusta Municipal | 0.43 ±0.18 | 0.09 ±0.04 |

MPC = $5 \times 10^6$ fCi/l

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*a*. 50-liter samples.

*b*. Mean and Std. Dev. resulting from 4 measurements.

TABLE III. MEASURED PLUTONIUM REMOVAL FACTORS ACROSS THE WATER TREATMENT FACILITIES

| | *Savannah, Ga. Industrial* | *Beaufort, S.C. Municipal* | *North Augusta, S.C. Municipal* |
|---|---|---|---|
| | | *10,000-Liter Samples* | |
| Nov. 1974 | 32.0 ±10.1[a] | *b* | *b* |
| Apr. 1975 | 10.2 ±3.5 | 15.7 ±2.0 | 3.0 ±0.8 |
| May 1975 | 8.7 ±1.4 | 17.4 ±3.9 | 5.7 ±2.3 |
| Mean & Std. Dev. | 17.0 ±13.0 | 16.6 ±1.2 | 4.4 ±1.9 |
| | | *50-Liter Samples* | |
| Aug. 12, 1975 | 34.6 ±14.2 | 5.5 ±1.8 | 6.0 ±1.1 |
| Aug. 26, 1975 | 25.9 ±8.8 | 17.5 ±9.0 | 11.5 ±6.1 |
| Sept. 10, 1975 | 19.7 ±6.7 | 8.1 ±4.2 | 2.2 ±0.9 |
| Sept. 24, 1975 | 15.8 ±7.3 | 26.8 ±14.5 | 3.6 ±1.6 |
| Mean & Std. Dev. | 24.0 ±8.2 | 14.5 ±9.7 | 5.8 ±4.1 |
| Combined Mean Value | 21.0 ±10.2 | 15.1 ±7.6 | 5.3 ±3.4 |
| Overall Mean | | | 14 ±10 |

*a.* Probable error.
*b.* No sample.

TABLE IV. PLUTONIUM CONCENTRATIONS IN WATER TREATMENT FACILITY FILTER MEDIA

| | *$^{239,240}Pu$ Concentration, fCi/g (dry weight)* | | |
|---|---|---|---|
| | *Savannah, Ga. Industrial* | *Beaufort, S.C. Municipal* | *North Augusta, S.C. Municipal* |
| Sand | 0.59 ±0.20 | | 4.42 ±0.48 |
| Anthracite | | 0.27 ±0.03 | 1.01 ±0.08 |

TABLE V. VARIOUS WATER QUALITY PROPERTIES OF RAW AND FINISHED WATER [a]

| | *North Augusta, S.C. Municipal* | | *Beaufort, S.C. Municipal* | | *Savannah, Ga. Industrial* | |
|---|---|---|---|---|---|---|
| | *Raw* | *Finish* | *Raw* | *Finish* | *Raw* | *Finish* |
| pH | 6.6 | 6.8 | 6.8 | 6.8 | 6.4 | 8.5 |
| Alkalinity | 14 | 25 | 12 | 22 | 18 | 28 |
| Suspended solids | 10 | 2 | 7 | 4 | 24 | 3 |
| Total dissolved solids | 42 | 64 | 69 | 96 | 61 | 87 |
| Biochemical oxygen demand | <1 | <1 | <1 | <1 | <1 | <1 |
| Chemical oxygen demand | 9 | 7 | 9 | 6 | 14 | <5 |
| Chloride | 2.1 | 5.0 | 6.8 | 13.4 | 5.0 | 8.6 |
| Nitrate | 0.13 | 0.19 | 0.00 | 0.03 | 0.26 | 0.18 |
| Nitrite | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 |
| Sulfate | 2.1 | 7.5 | 2 | 24.8 | 3.5 | 14.0 |
| Sulfide | <1 | <1 | <1 | <1 | <1 | <1 |
| Total phosphate | <0.02 | 0.40 | 0.05 | 0.08 | 0.08 | <0.02 |
| Ortho phosphate | <0.02 | 0.15 | <0.02 | <0.02 | 0.05 | <0.02 |
| Ammonia | <0.01 | <0.01 | 0.04 | <0.01 | 0.03 | <0.01 |
| Total Kjeldahl nitrogen | <1 | <1 | <1 | <1 | <1 | <1 |

*a.* All values, except pH, are in mg/l.

TABLE VI. 70-YEAR BONE-DOSE COMMITMENT TO AN INDIVIDUAL FROM ONE YEAR OF WATER CONSUMPTION AT 1.65 LITERS PER DAY, mrem

| | *Raw Water* | *Finished Water* |
|---|---|---|
| Savannah Industrial | 0.0010 | 0.00004 |
| Beaufort Municipal | 0.0005 | 0.00003 |
| North Augusta Municipal | 0.0002 | 0.00004 |
| Mean | 0.0006 | 0.00004 |

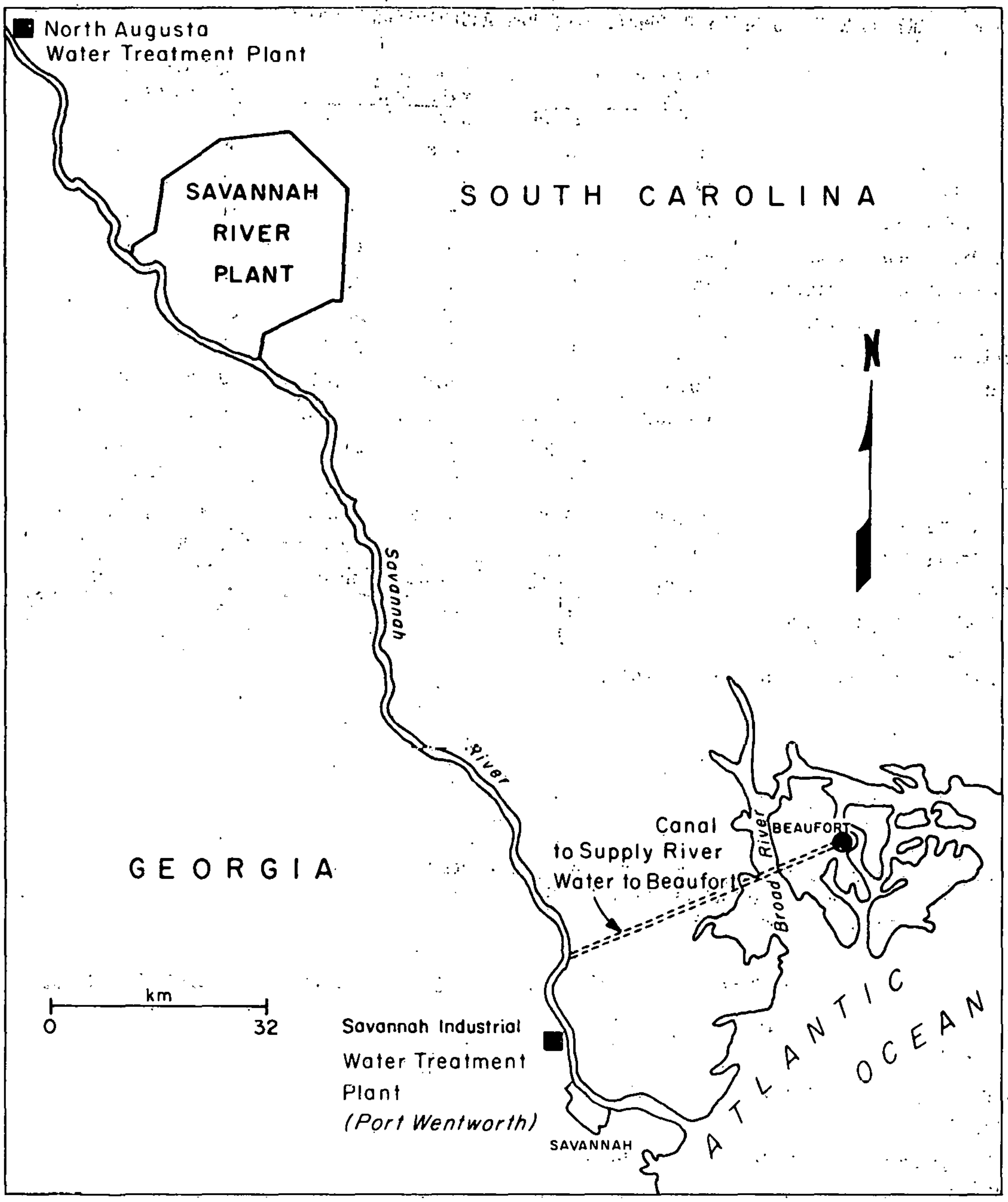


FIG. 1. Locations of water treatment plants using Savannah River Water.

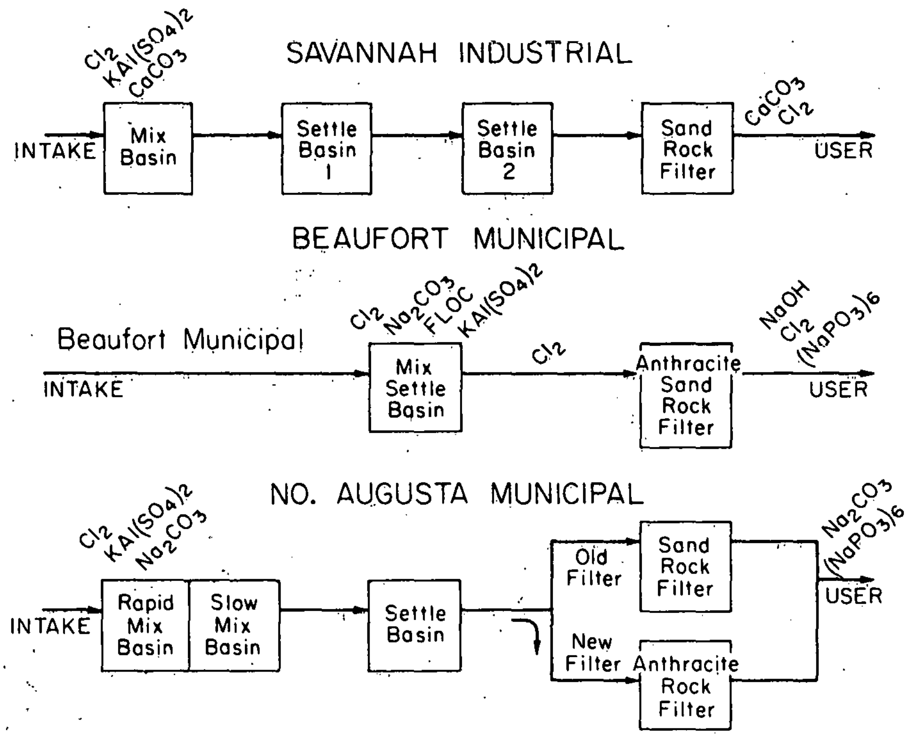


FIG. 2. Processes used for treating Savannah River Water.