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PHYSICAL, CHEMICAL, AND RADIOLOGICAL PROPERTIES OF
SLURRY PARTICULATE FALLOUT COLLECTED DURING
OPERATION REDWING

Research and Development Technical Report USNRDL-TR-170
NS 088-001

5 May 1957

by

N. H. Farlow
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U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY

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Technical Objective
SR-2

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Chemical Technology Division
E.R. Tompkins, Head

Scientific Director
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Commanding Officer and Director
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U. S. NAVAL RADIOLOGICAL DEFENSE LABORATORY
San Francisco 24, California

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ABSTRACT

The properties of individual fallout particles produced by nuclear detonations at zero height over shallow sea water are analytically described for the first time. The particles produced during Operation REDWING were slurry masses composed of water, dissolved and crystalline sea salts, and seawater-insoluble solids from the weapon, barge, and ocean floor. Special techniques were used to measure the chloride, water, and insoluble-solids content of individual slurry particles. Autoradiography showed that the activity is primarily associated with the solids.

A table of experimental data presents particle size versus time of arrival after detonation as well as measurements of particle density and relative specific activity. Estimates of mass and relative activity of fallout per unit area for certain locations about the shot point are shown.

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SUMMARY

The Problem

Nuclear devices fired over sea water at previous weapons tests yielded fallout which was different from that associated with land surface detonations. The analytical methods previously used on dry fallout were grossly inadequate for this slurry-like material.

Certain tests at Operation REDWING yielded slurry-like fallout. Therefore, new analytical methods were required to assess the material properly.

Findings

The fallout from two seawater-surface nuclear events at Operation REDWING has been analyzed using new quantitative techniques for the measurement of chloride and slurry droplet water content. Particle size, density and radiological properties of the slurry fallout have been assessed satisfactorily for the first time.

ACKNOWLEDGMENTS

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be evaluated. Physical observation could detail the state of the material as it landed at the station. Chemical treatment would allow the separation and measurement of soluble salts and seawater-insoluble ones which were to be defined as "insoluble solids." Radiological evaluation would assess the particle radioactivity. These experimental values then would define the physical state of the particles, particle size, density, particle specific activity, total activity, and mass of fallout per unit area.

Assessment of Physical Properties

Visual observations made in the shielded laboratory confirmed the slurry droplet nature of the fallout. Crystalline materials were observed to be suspended in the liquid portion of the droplets. The characteristic cubic shape of sodium chloride was detected. After the dissolution of soluble halides, an insoluble-solids component remained on the film. It was apparent that the slurry droplets contained three major constituents: water, soluble sea salts, and insoluble solids from the environmental materials (Fig. 1).

Measurement of Halide Content

The reagent film for particle halide analysis utilized a commercial gelatin film in which colloidal red silver dichromate was precipitated by a special method.¹ Soluble halides deposited on the film were later dissolved by saturated hot water vapor and diffused into the gel structure where they were precipitated as silver salts. A microscope measurement of the reaction area yielded, by calibration, the weight of halides reported as sodium chloride. The measurable range of sodium chloride by this method is from 10^{-6} to 10^{-2} g.

Measurement of Water Content

When a droplet strikes the reagent film it spreads to a certain constant degree related to the volume of water in the droplet.⁵ Insoluble solids within each fallout slurry droplet outlined this maximum spread on the collecting film. A calibration curve² was constructed giving the volume of water in a slurry droplet as a function of the area covered by the insoluble-solids trace. This area has been termed the "slurry artifact." The smallest water volume measurable by this technique is about 10^{-10} cc.

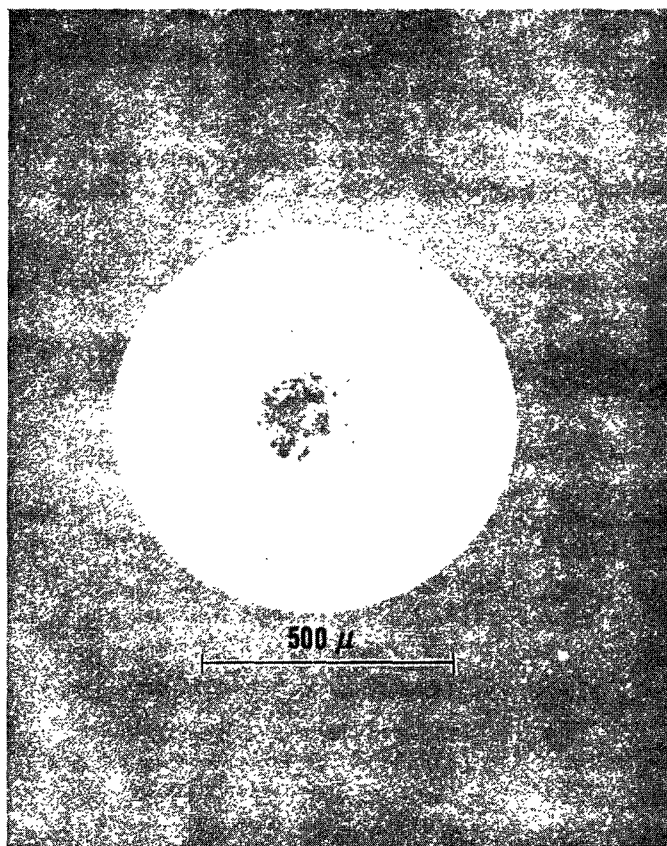


Fig. 1 Reaction of Slurry Fallout Particle on Reagent Film. The white circular area is halide reaction representing 2.9×10^{-7} g NaCl. The central elliptical area is slurry artifact of insoluble solids representing 7.2×10^{-7} cc. Particle density is 1.19 g/cc. The annular rings in the chloride reaction area are thought to be a Liesegang phenomenon.

Measurement of Insoluble-Solids Content

No known chemical method can measure the weight or volume of insoluble solids in a deposited slurry fallout particle. Therefore, a method for comparing the volume of the fallout particle with standard volumes was developed. Five standard volumes ranging from 10^{-7} to 10^{-9} cc were used for the visual comparison in the microscope. They were formed by aspirating and collecting on reagent film small slurry droplets from measured aluminum oxide suspensions. The water content of each droplet was measured by its slurry artifact. By arithmetic proportion the approximate volume of aluminum oxide was known. Five appropriately sized aluminum oxide artifacts were mounted on a microscope slide as a comparison standard. Each fallout slurry artifact was then visually compared with the standards, and estimates made of the volume of insoluble solids.

The physical-chemical composition of the fallout insoluble solids component is being investigated at this laboratory.

Measurement of Radiological Properties

After solution and diffusion of the soluble halides into the reagent film, autoradiographs were made by adaptations of the LaRiviere-Ichiki method.⁶ These studies showed the activity to be primarily centered in the insoluble-solids portion of the fallout particle (Fig. 2).

These solids were subjected to salt water leaching by the liquid phase of the droplet for at least part of their falling period. Ionic activity available for solution dissolved in this solvent. Upon striking the film the dissolved activity diffused with the water into the gelatin and there was rigidly held. It was felt that if the active solids portion of fallout could be stripped from the film and both parts counted, a rough estimate of easily soluble ionic activity could be made.

One Flathead film sample containing myriads of fallout impressions was selected for stripping and counting. The insoluble solids on the reagent film were thoroughly leached in hot water vapor at 70°C for several hours allowing further diffusion of ionic activity into the film. While the gelatin of the reagent film was still tacky from the vapor treatment, a thick layer of transparent acrylic spray was applied and allowed to dry. The assemblage was then soaked in distilled water for an hour to permit diffusing moisture to loosen the solids from the gel. The acrylic film was then stripped from the gelatin, removing most of the insoluble solids with it. This stripping process was repeated until no microscopically visible insoluble solids remained on the reagent film. The commercial gelatin of the film is so tightly bound to its substrate

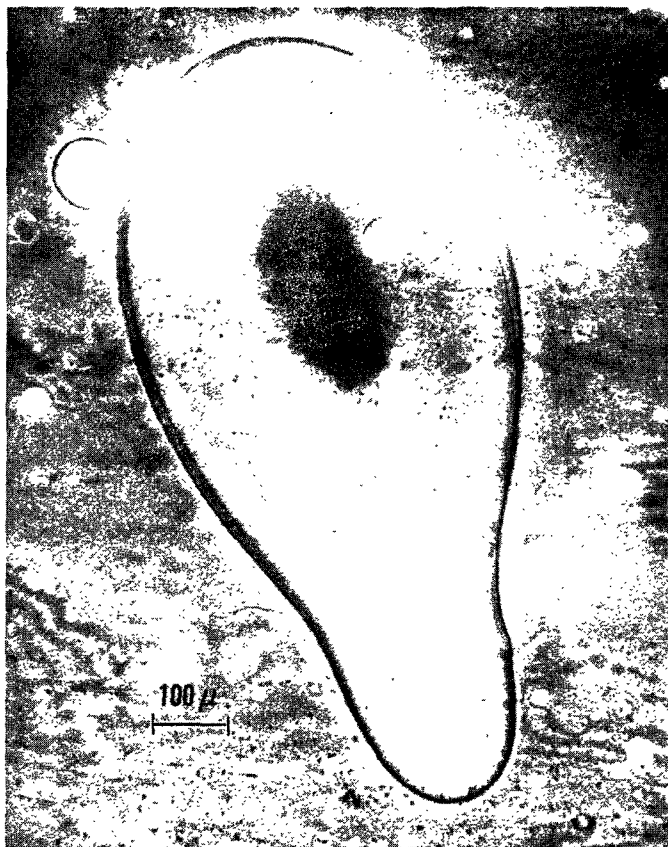


Fig. 2 Autoradiograph of Slurry Fallout Particle Trace and Its Chloride Reaction Area Showing the Radioactivity to be Concentrated in the Centrally Located Insoluble Solids Portion. The black central smear is the autoradiograph masking the solids. Patterns on the reagent film are caused by severe tropical sun reaction on the photosensitive film. Circular spots are NaCl reactions from naturally occurring airborne sea salt particles.

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Table 1 Slurry Fallout Particle Data

Time of Arrival Interval (H+hr)	Ship Station	No. of Particles Measured	Average NaCl Mass (μg)	Average H ₂ O Mass (μg)	Average Density \pm Std.Dev. (g/cc)	Average Diameter ^(a) \pm Std.Dev. (μ)	Average Specific Activity \pm Std.Dev. ($\times 10^{10}$ c/m/g) ^(b)
Flathead							
1 to 3	YFNB-29	4to10	0.06	0.08	1.28 \pm 0.1	57 \pm 6	43 \pm 8
7 to 9	YAG-39 & LST-611	50to52	0.42	0.62	1.29 \pm 0.01	112 \pm 2	282 \pm 20
11 to 12	YAG-40	10	0.94	1.20	1.35 \pm 0.05	129 \pm 16	285 \pm 160
15 to 18	YAG-40	3to4	0.50	0.69	1.34 \pm 0.08	121 \pm 6	265 \pm 90
Totals		67 to 76			1.30 \pm 0.01		282 \pm 30
Navaho							
1 to 3	YFNB-13	5to20	7.77	7.94	1.38 \pm 0.04	272 \pm 14	4 \pm 0.6
3 to 5	YAG-39	9to14	7.62	4.49	1.50 \pm 0.1	229 \pm 24	16 \pm 3
5 to 6	LST-611	14	1.61	1.83	1.41 \pm 0.04	166 \pm 6	14 \pm 2
7 to 9	YAG-40	4to10	1.25	1.08	1.45 \pm 0.04	142 \pm 22	9 \pm 3
9 to 10	YAG-40	5to23	0.44	0.60	1.31 \pm 0.02	110 \pm 5	11 \pm 2
10 to 11	YAG-40	11to15	0.66	0.50	1.43 \pm 0.03	111 \pm 4	16 \pm 4
11 to 12	YAG-40	33	0.30	0.44	1.32 \pm 0.01	94 \pm 4	26(c)
12 to 13	YAG-40	28	0.31	0.31	1.37 \pm 0.01	96 \pm 2	21(c)
13 to 14	YAG-40	6	0.17	0.27	1.28 \pm 0.02	86 \pm 7	29(c)
14 to 15	YAG-40	5	0.10	0.18	1.30 \pm 0.03	75 \pm 2	23(c)
15 to 18	YAG-40	13to14	0.06	0.32	1.15 \pm 0.02	84 \pm 4	56 \pm 7
Totals		133 to 182			1.35 \pm 0.01		21 \pm 3

(a) The diameter of the spherical slurry droplet at the time of arrival

(b) Photon count in crystal well counter at H+12

(c) Calculated value based on total tray count, number of particles per tray, and average NaCl mass per particle

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calculation of an average activity per slurry particle, and an estimate of the average volume and weight of insoluble solids per particle by the standard spot comparison method.

The two independent methods gave an average value for Flathead of 1×10^{13} c/m/g at H + 12 in the well counter. The latter method only was used for Navajo, resulting in a value of 1×10^{12} c/m/g of insoluble solids.

Table 2 presents experimental data on the total activity per unit area for the two events for various stations. These data are in terms of a count (at H + 12) in the crystal well counter. The total mass of sodium chloride per unit area is a calculated value obtained by dividing the total activity per unit area for a given station (Table 2) by the average activity per NaCl mass (Table 1). Sodium chloride represents the major solid mass of fallout. Since the water content depends on the humidity conditions through which the particle passed, the weight of water is not considered a part of fallout mass. However, if one wishes to compute the approximate weight of water, one multiplies the mass of NaCl by 1.2, which is the average weight ratio of water to NaCl. A plot of the dependent variables, activity per unit area versus NaCl mass per unit area, presents Table 2 data as a necessarily smooth curve (Fig. 3). With this plot one can roughly deduce relative activity by a measure of sodium chloride per square foot or the converse. The very high initial values of sodium chloride mass per square foot have not been plotted on the curve.

DISCUSSION

The particle densities (Table 1) cluster about a mean value which is nearly the same for both events. The mean density for the 1- to 3-hr period is close to the overall mean value. By whatever means the ratio of water to solids reaches equilibrium, this mechanism is fairly rapid. Equilibrium has been reached by the time the particle lands.

In Navajo there appears to be a sharp decrease in density after H+13. This time corresponds to the after-sunset hours when changes in ambient atmospheric conditions might be expected.

The particle size (Table 1, "Navajo") generally decreases with time, although it is noteworthy that there is little droplet size variation in 15 hr. For any given time period, one need not discuss particle size distribution since the standard deviation of the mean diameter is so small.



TABLE 2
Total Activity and Mass of Fallout

Collecting Station	Flathead		Navajo	
	Total Activity(a) $\frac{c/m}{ft^2}$	Total Mass NaCl $\mu g/ft^2$	Total Activity(a) $\frac{c/m}{ft^2}$	Total Mass NaCl $\mu g/ft^2$
YFNB-13-E-57	--		143,000,000	3,580
YFNB-29-H-78	98,400,000	229	6,500,000	31
YAG 39-C-20	14,800,000	5.25	37,400,000	178
YAG 39-C-24	3,020,000	1.07	--	--
LST 611-D-37	37,700,000	13.4	--	--
LST 611-D-50	4,850,000	1.72	--	--
YAG 40-A-1	28,800,000	10.2	28,200,000	134
YAG 40-A-2	31,500,000	11.2	--	--
YAG 40-B-7	11,900,000	4.22	--	--

(a) Photon count in crystal well counter at H+12.



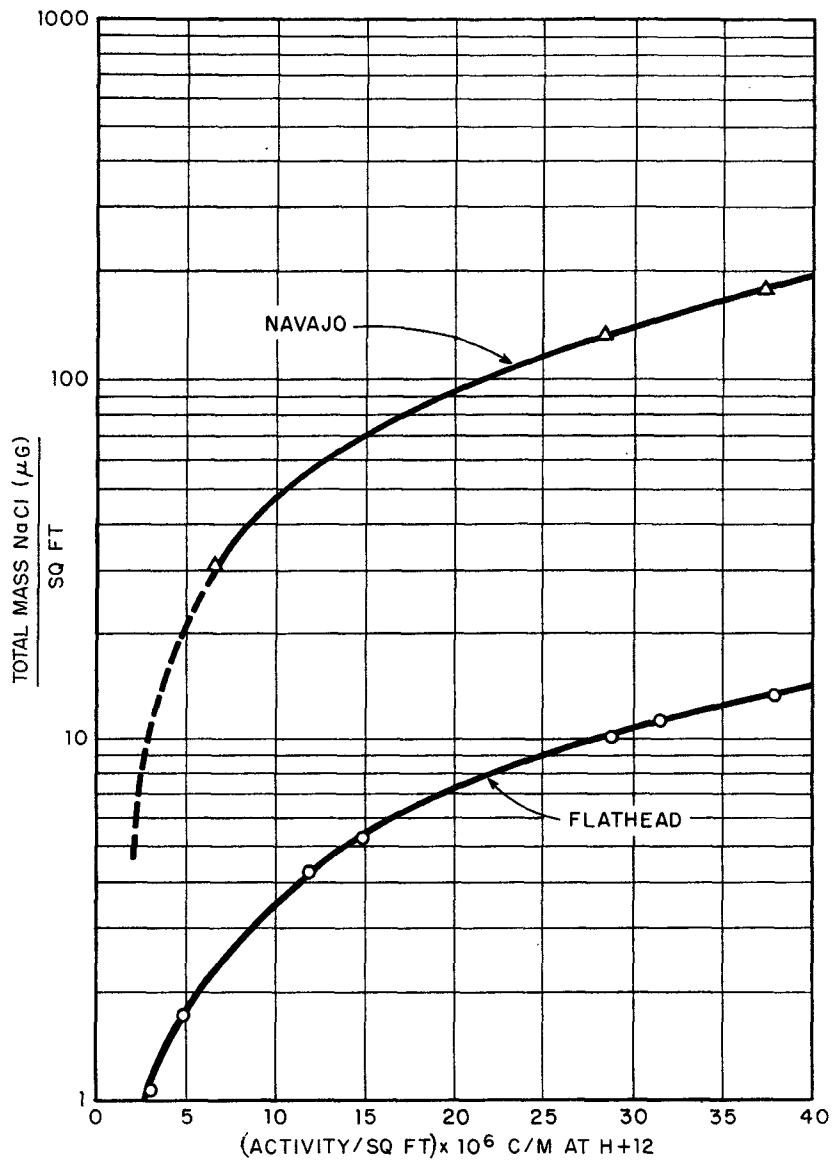


Fig.3 Plot of NaCl Mass Versus Activity Per Square Foot.

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Early arriving particles appear to have a much higher ratio of sodium chloride to activity than do later arrivals (Table 1, last column.) Such a variation might be indicative of early large droplet fallout from the cloud stem region where the concentration of activity may be less. The sampling is small, however, so caution must be exercised in the interpretation of these data. The calculations of total mean values of specific activity (Table 1) do not include these initial values, nor does the calculation of this value for Navajo include the approximate values defined by footnote (c) of the table.

A comparison of event Flathead with Navajo shows the ratio of total yields is 1:12 while the activities produced by the devices are approximately equal. Since the barge complex for each event was identical, the insoluble solids contributed by this complex are identical for both events. On the basis of activity per gram of sodium chloride, 13 times the amount of seawater was carried aloft by Navajo. Estimates of insoluble solids specific activity indicate that Navajo fallout contained about ten times more solids per activity than did Flathead. Whether this was contributed by calcium and magnesium salts from the seawater carried up by Navajo or by additional bottom material is conjectural.

It appears that the hygroscopic slurry particles can change markedly in size, density, and falling rate due to environmental influences. A detailed study of these effects is required before particle point of origin estimates can be made using measured size data.

Validity of the Data

The data of Table 1 are based on analyses which have been extensively calibrated and tested in the laboratory. The average error in the chloride analysis is about ± 5 percent. The standard deviation error of a water volume measurement is about ± 25 percent. Estimates of insoluble solids volumes are only approximate and can be subject to large errors. The number of particles sampled for each event is small, but the analyses carried out on each of these is detailed and within the errors shown. Standard deviations quoted in Table 1 are deviations of the mean value, not deviations of a single measurement from the mean.⁷ The radioactivity assay was done with counting instruments thoroughly calibrated and tested. Both standard isotopes and fission-product activities from the actual events were used to evaluate the instruments.

The tabulation of activity per square foot (Table 2) reveals sampling biases which call for caution in the use of these data. Identical adjacent collectors, YAG 39-C-20 and YAG 39-C-24, sampled the same fallout event, yet the total activities recorded for Flathead for these stations differ by a factor of 5. Samplers of somewhat different design considerably

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apart on the same ship, YAG 40-A (2 samplers) and YAG 40-B-7, collected samples differing by a factor of 3. The evaluation of these collection biases is under study at this laboratory.

Obviously, measures of total activity per unit area and perhaps of particle size are influenced by instrument and location biases. However, analyses for particle chloride content, water content, solids volume and activity values are of reasonable precision. These are parameters of individual particles and are not influenced by instrument biases.

Applications

The analytical techniques used here are directly applicable to a study of atmospheric sea salts. The similarity between the environmental interactions of naturally occurring particles and of slurry fallout is very striking. A study of the former would yield useful information on environmental reactions of slurry fallout.

Approved by:

E. R. Tompkins

E. R. TOMPKINS
Head, Chemical Technology Division

For the Scientific Director

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