000-2736-2

Some Aspects of Organic and Inorganic
Particulate Transport at Rocky Flats

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Final Report on Contract EY-76-S-02-2736

Prepared for the U.S. Energy Research
and Development Administration

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Initially, this study involved the sampling of airborne particulates to discover whether organic and inorganic particles (such as pollen grains, spores, sand, silt, and clay-sized mineral fragments) were transporting smaller pieces of radionuclide material. The reasoning underlying this project was the likelihood of mineral particle transport by wind across the site, and the presence in this natural environment of astronomical numbers of pollen and spores, released seasonally, with a known ability to travel very long distances in air, and a propensity for attracting other particles (by static charge), and for carrying them attached to the elaborately-sculptured (and sometimes sticky) pollen walls. The various air samplers in use at Rocky Flats plant in 1975 were not designed to sample these relatively large light organic particles which might therefore have escaped detection, so that a subsidiary air sampling program was set up using standard aero-biological trapping techniques plus filter samples from the regular Rocky Flats air samplers.

The test for association between pollen and small radioactive particles involved the irradiation of a sampling slide containing pollen. A polycarbonate film capable of registering fission tracks was in contact with the sample during irradiation, and chemical etching of the film revealed radionuclide fission tracks. Microscopic examination of the slide searched for pollen grains which were consistently at the identical locations of the fission tracks and star bursts, to prove radionuclide transport by pollen, but the results were negative. Thus from this initial study there is no indication that out of the many large radionuclide particles transported across the site any significant number were transported by pollen grains or spores.

However, by comparison of the efficiencies of the different air samplers (both the official ERDA equipment and our own) we conclude that the lightest and largest pollen (pine), which is probably most capable of radionuclide transport, is not collected by the low-volume air sampler. This failure to collect any of the numerous pine pollen present in this environment is a clear deficiency, and raises the question of what other particulates are not being collected by the standard sampling equipment.

The program then tested soil and snow samples to examine the distribution of radionuclide particles across the Rocky Flats area. In the winter of 1975-1976 we sampled fresh snow from areas of preferred snow accumulation (snow banks) since these were clearly prime sites for concentration of the other particles, including radionuclides. Melted snow was filtered to provide two populations of particles, $> 0.5 \mu\text{m}$ and $0.5 - 0.05 \mu\text{m}$. These were irradiated in contact with polycarbonate film and the resulting etched fission tracks were counted microscopically, characterized as to track size (number of star bursts) and plotted. Star bursts indicated an irradiated radionuclide particle, and these were registered at all the sites studied, including those at Indiana Street 3 miles (5 km) from the "Hot Spot" contaminated area. The Indiana Street snowbanks contained as much or more radionuclide particles in the smallest size range ($0.5 \mu\text{m} - 0.05 \mu\text{m}$) as sites close to the "Hot Spot." It seems logical to infer that this reflects a dissemination of the finest radionuclide particles throughout the area, over a radius of several miles from the plant site, forming a background level of airborne contamination. Once airborne, these smallest particles are not noticeably reduced in numbers by gravitational settling up to 3 miles (5 km) from the apparent point of origin, and presumably reach much farther afield. Snow samples immediately east of the "Hot Spot" contained more large particles ($>0.5 \mu\text{m}$) of 100 star-track size, as did sites just west of the plant fence.

The concentration of activity at the snowbank sites may argue for snow itself as an agent in the deposition of fissile materials, possibly by snow crystal abrasion. These data show the need for any program of soil sampling for radionuclides in this area to recognize the local effects of topography and winds, and not to depend solely on a grid-pattern of sampling.

Soil sampling employed alpha-sensitive film left in contact with soils at several field sites to test by auto-irradiation whether or not the radionuclides included plutonium. Alpha tracks were barely or not detected at sites most distant from the plant, while nearer locations registered higher numbers, with peak values close to the "Hot Spot", as expected. However, this was a relatively insensitive method of testing, due to difficulties of maintaining close contact between soil and film for weeks, failure of some film to develop, etc., and the possibility that especially large particles of uranium and thorium deriving from the uranium mine west of the plant site can produce alpha tracks on the films.

SUMMARY OF THE SAMPLING PROGRAM

Low volume air sampler and Anderson Cascade Impacter

Samples of the fiberglass filters from the Low Volume Air Sampler located at AG82 (close to the "Hot Spot") were collected at weekly intervals, and were irradiated and counted for fission tracks. The largest number of fission tracks recorded from August 1975 to May 1976 fell within the period of December to March, the time of maximum winter winds, with subsidiary isolated events in September and April; (several weeks data representing late October - early November could not be obtained). These data were paralleled by the fission track totals from the Anderson collector at the same location (AG82). During the winter period there were generally one or more large particles (registered as > 100 fission track features) deposited per square centimeter per week.

Static slide sampling

Microscope slides with sticky surfaces were set up to sample airborne radio-nuclide particles at several locations known to be highly active (e.g., Y46, AC84) but very few fission tracks resulted from irradiation of these collectors, and this method of sampling was abandoned in favor of reliance on the powered samplers. This negative result was not necessarily due to a clean environment, but to the well-known difficulty of persuading small particles to impinge on a solid surface, due to the boundary layer effect.

Snow samples

During the winter of 1975/76 episodes of prolonged snowfall allowed the collection of fresh snow samples from areas of preferred snow accumulation, which after partial snow-melt were left as residual snow banks. These accumulations were due to microtopographic discontinuities, breaks of slopes, bluffs, hollows, etc., and their orientation related to the wind at the time of snow deposition. They are therefore prime sites for accumulation of other particulates, including radionuclides.

Snow samples equal to approximately one liter of water when melted were pumped through Millipore filters of 0.5 micron and 0.05 micron pore diameter.

The two populations of particle sizes (greater than 0.5 μ m, and less than 0.5 μ m and greater than 0.05 μ m) were packaged in contact with polycarbonate film and irradiated at U.S.G.S. TRIGA III reactor, Denver. Etching of fission tracks due to alpha radiation made the tracks visible at optical microscope magnifications (100 - 1000X). The tracks were counted, characterized as to track size (number of star bursts) and plotted, as exemplified by Figs. 4 to 12. The star bursts indicated an irradiated radionuclide particle, and these were registered at all the sites studied. The sites farthest away from the plant (Indiana St. sites, North Slope) are characterized by a predominance of particles between 0.5 - 0.05 μ m over the $> 0.5 \mu$ m particles. It seems logical to infer that this reflects a dissemination of the finest radionuclide particles throughout the area, over a radius of several miles from the plant site, forming a background level of contamination. This activity in the smallest size range of particles appears to be no less than near the "Hot Spot" itself, at location AF 83, implying that once airborne the particles $< 0.5 \mu$ m are not noticeably reduced in numbers by gravitational settling up to 3 miles from the "Hot Spot". These data imply radionuclide particle transport considerably beyond Indiana St., presumably to the outskirts of Denver and Boulder.

The reduction in activity at Indiana East compared to Indiana West (less than 400 yards apart) may be due to the plowed field near Indiana East, which may contribute uncontaminated topsoil to the site and thus "dilute" the radionuclide deposition. Indiana South, two miles south of these other sites on Indiana St., does show a reduction of activity. While the downwind sites are mostly characterized by a predominance of the smallest particles sampled, upwind sites have higher values of larger particles ($> 0.5 \mu$ m) than smaller ones (0.5 - 0.05 μ m). This was observed at Y 46 (near the construction site) and even at Dr. Caine's needle-ice study plot, to a diminished degree. The meteorological conditions under which there would be deposition of $> 0.5 \mu$ m radionuclide particles in greater numbers than 0.5 - 0.05 μ m particles are not clear at present.

The origin of the radionuclides in the snow may conceivably involve diffusion up through the snow from the contaminated soil, but since efforts were made to sample snow within 12 - 24 hours of the end of snowfall this is unlikely to be a major component. Indeed the concentration of activity at the snow bank sites (plus the radionuclide content of the fresh snow samples)

argues for snow itself as an agent in the deposition of fissile materials at these accumulation sites. Whether this involves the abrasion of contaminated bare soil by snow crystals and the transport of radionuclides attached to snow particles, or whether snow and soil particles are both blown independently of each other into accumulation sites, remains unclear.

A major question arising from these data are: are the fission tracks due to plutonium, americium, or beryllium, from the previous contamination at the Rocky Flats site, or are these from other radionuclides derived for example from the uranium mine about five miles away? The answer to this presumably requires a considerable effort in radio-chemical assay by Rockwell staff, such as I discussed previously with them when these findings were first made by our study.

Soil samples

Alpha-sensitive film was left in contact with a variety of soil materials at several sites to test by auto-irradiation whether or not the radionuclides were plutonium. This, however, cannot be a precise diagnostic test since especially large particles of uranium and thorium can produce alpha tracks on film, and these radionuclides may be present at the sites, derived from the uranium mine about five miles west of Rocky Flats. The most distant sites (Dr. Caine's study plot, and the Indiana Street sites) several miles from the "Hot Spot" registered few or no alpha tracks, whereas closer to the contaminated area (AJ 79, Y 46) both "upwind" to the west and "downwind" to the east there were higher values. At locations such as AG 82, AF 83, AC 84 closest to the "Hot Spot" there occurred the greatest concentrations of alpha tracks, as expected.

The use of alpha-sensitive film in contact with soil is not entirely satisfactory for registering plutonium presence, due to the difficulty of arranging close contact between soil and film for several weeks, failure of some film to develop, diffusion of radionuclides into the soil at an unknown rate, etc., so that the low numbers of alpha tracks registered three miles away at Indiana St. should not be taken as a clean bill of health, but a clear warning that even while using such sub-optimum methods we have registered what may be plutonium transport miles from the plant site.

CONCLUSIONS

We present here the summary findings from the summer 1975 sampling program and the conclusions from the 1975-76 winter sampling program.

The interim conclusions reached from the summer sampling program of 1975 were these:

- (1) Large radionuclide particles (100 star track size) were not found attached to pollen.
- (2) Sampling the airborne pollen throughout the Rocky Flats area demonstrated that the Rocky Flats air samplers were deficient in their abilities to properly collect large lightweight pollen grains (such as pine). In view of (1) above, this may not have serious consequences for the undetected transport of radionuclides by pollen, etc. However, it has implications of sampling inefficiency in some particulate size ranges, and implies that if radionuclides are carried at times by larger organic or inorganic particles then the standard Rocky Flats air samplers are not sampling these materials efficiently.
- (3) Some initial findings suggested that soil samples from downwind areas associated with winter snow accumulation had abnormally high fissile particle accumulations, and this suggested the need to sample these sites in winter to check the activity of soil and snow samples.

The winter sampling program of 1975-76 has produced these conclusions:

- (1) Areas of preferred particle accumulation such as snow banks have higher concentrations of radionuclide particles than surrounding areas which are more windswept. Any soil sampling program at Rocky Flats which seeks to monitor background radionuclide levels and movement of fissile particles across the site should take account of these topographically and wind-controlled accumulation areas, and not rely solely on a grid pattern of sampling.
- (2) The snow samples contained radionuclide material in substantial quantities. In terms of fissile particulates smaller than 0.5 microns the

snow banks on Indiana St. (three miles east of the plant site) contained as much or more radionuclides than sites close to the "Hot Spot" contaminated area. The snow samples immediately east of the "Hot Spot" contained many more large particles (>0.5 m) of 100 star-track size, as did soil samples from the upwind site next to the plant fence, at Y 46. The "Hot Spot" is clearly implicated again as the source of these materials, and it is interesting that sites next to the plant have radionuclide contamination at relatively high levels, containing large fissile particles.

Figure 1: Map of study areas showing pollen sampling sites I, II, III and IV and the soil and snow bank sampling locations of Macroplot I and Indiana West, East and South. (courtesy of Rockwell International)

Figure 2: Diagram of four sampling locations at Site III.
(courtesy of Rockwell International)

OUTSIDE AIR FILTER

Figure 3: Photograph of fission star in 10 μ m polycarbonate film.
(courtesy of J. Hayden)

Fig: 4

hot spot
AF 33

Irradiated Snow Samples

2-6-76

46 1510

KE 10 X 10 TO THE CENTIMETER 18 X 25 CM.
KEUFFEL & ESSER CO. MADE IN U.S.A.

Fig. 5

upward, concentration etc

Y46

Irradiated Snow Samples

258 plus 100's
2-6-76

total
total

0017 097 057 527 017

46 1510

K_M 10 X 10 TO THE CENTIMETER 18 X 25 CM. KEUFFEL & ESSER CO. MADE IN U.S.A.

Fig. 6 downwind

North Slope

Irradiated Snow Samples

2-6-76 (77)

46 1510

10 X 10 TO THE CENTIMETER 18 X 25 CM.
KEUFFEL & ESSER CO. MADE IN U.S.A.

Irradiated Snow Euphorbia, 3-31-76

Fig. 8

continued

Indiana East

Irradiated Snow Samples

2-6-76

46 1510

K & M
10 X 10 TO THE CENTIMETER
KEUFFEL & ESSER CO. MADE IN U.S.A.

Fig. 9. downloaded

Indiana West

Irradiated Snow Samples

2-6-76

461510

KE 10 X 10 TO THE CENTIMETER
18 X 25 CM.
KEUFFEL & ESSER CO. MADE IN U.S.A.

total
.5 M

total
.05 M

Fig. 10 upwind
in CAINE'S Plot

Irradiated Snow Samples

2-6-76

461510

10 X 10 TO THE CENTIMETER
KEUFFEL & ESSER CO. MADE IN U.S.A.

Fig. 11 downward

Indiana S.

Irradiated Snow Samples

2-6-76

46 1510

K₂S₂O₈ 10 X 10 TO THE CENTIMETER 18 X 25 CM.
KEUFFEL & ESSER CO. MADE IN U.S.A.

120
110
100
90
80
70
60
50
40
30
20
10

410 425 450 4100 7100

total
.54

total
.654

downloaded

Fig. 12.

Indiana South

Irradiated Snow Sample.

3-31-76

46 1510

10 X 10 TO THE CENTIMETER 18 X 25 CM.
KEUFFEL & ESSER CO. MADE IN U.S.A.

K&E

120
110
100
90
80
70
60
50
40
30
20
10

410

425

450

4100

7100

total
15.1

total
.0521

APPENDIX I

NUCLEAR TRACK ANALYSES

FISSION TRACK ANALYSES

The experiment authorization Parts I, II, III enclosed is written to cover the three types of sample to be packaged and sent to the USGS Triga reactor at the Federal Center at Sixth Avenue and Kipling in Denver.

If a substantially different sample is to be submitted then a further set of authorization papers should be obtained from Mr. Pat Kraker, the reactor supervisor.

Only personnel specified on the authorization may receive the irradiated materials. The names of Leigh Heart and Ann Crooimans have been added to the present authorized list. These people must stay in communication with the University of Colorado Health Physicist, Mr. Jerome Martin, and are responsible for exchanging their dosimetry badges at the beginning of each month at the Health Physics building. The Reactor Health Physicist, Mr. Bill Smith, may not deliver irradiated samples to a person not wearing a badge.

If reactor samples are collected shortly after irradiation, precautions with handling must be taken as per instructions of Health Physicist. The samples must be placed behind lead bricks of fume hood in Room 104, and left until less active before unpackaged. Most of this activity will be short-lived as can be seen by this test decay curve plotted by Mr. Mark Otis of C.U. Health Physics Laboratory from the first returned samples, using an end window Geiger tube at contact.

DATE	8-14-75	8-15-75	8-18-75
TIME	4:00 pm	8:00 am	4:00 pm
SOIL	55 m R/hr	45 m R/hr	2.5 m R/hr
FILTER	25 m R/hr	15 m R/hr	1.5 m R/hr

The three sample types submitted are:

- 1) soil
- 2) air filter
- 3) snow

1) For irradiation, less than 1 gm of surface soil is taken from undisturbed ground and sampled directly into small plastic tubes (30 mm X 14 mm diameter). The tubes are capped and labelled in the field. In the laboratory, a 25 mm square of 10 μ polycarbonate film is placed over the tube mouth and the cap replaced so that the film is stretched evenly. Inverting the tube places the soil in contact with the film. The size of each tube enables three soil samples to be placed into each Triga tube for irradiation.

2) Sections of the air filters from both the Andersen Sampler and the low volume air sampler are cut and placed on 76 mm X 22 mm plastic slides which are then labelled. Then a piece of polycarbonate film of the same size is placed over the filter section and the two pieces notched so that they may be super-imposed after development. A sandwich of four to six samples is built up: slide-filter-film-slide-filter-film, etc. Over the top film section another slide is placed and the whole sandwich bound together as tightly as possible with Scotch Tape.

3) Typically 1 liter beakers are filled with snow, transferred to plastic bags in the field and labelled. When the snow has melted, the liquid is filtered through 47 mm diameter, .5 μ and .05 μ nuclepore filters. The date of collection is recorded along with the volume of water the 1 liter of snow reduces to. Sections of the nuclepore filters are packaged in similar fashion to the air filter samples.

The samples to be placed inside each Triga tube are placed inside two heavy duty polyethylene bags, each separately heat sealed. Care is taken to limit the handling of samples, and gloves mostly worn so that the samples

do not become contaminated with sodium chloride from hands. Both these elements become "hot" during irradiation but do have short half lives.

Samples should be delivered to the reactor in sets of three tubes at \$10.00 per tube irradiation cost, their minimum order being for \$25.00 per sample. A "Radioisotope Request and Receipt Form" must accompany each set (examples of how these are to be filled out are enclosed). When the samples are received after irradiation, the form is signed and should be filed with the other receipts as record.

To develop the polycarbonate film, a solution of 6 X Normal sodium hydroxide is prepared. This is made by dissolving 6 X (23(Na) + 16(O) + 1(H)) gm in 1 liter of distilled water. This solution is then placed in a small beaker in a water bath maintained at 55-60°C. Once the film is placed in the solution the temperature of the NaOH should not be allowed to exceed this. The fission tracks in the polycarbonate will become sufficiently etched in approximately fifteen minutes. The time may need adjusting for optimum track visibility at 200 power. The film is neutralized in a very mild solution of hydrochloric acid, then thoroughly rinsed and dried. Artifacts on the film are found to be best hidden when the film is placed between the slide and coverslip and oiled with microscope immersion oil. The number of fission stars are recorded in groupings of <10, <25, <50, <100, >100 tracks.

Data Sheet #F1 is the form to be used by the individual analyser to keep a permanent record of his counts. The information is then transferred to the appropriate record sheet.

Data Sheet #F2 for multiple stage Andersen counts.

Data Sheet #F3/3A for single stage Andersen counts.

Data Sheet #F4/4A for the low Volume Air Sampler.

Data Sheet #F5 for soil or snow samples or nonpowered air samplers.

NUCLEAR TRACK DATA

Analyst _____

[illegible]