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REVIEW OF THE STACK DISCHARGE

ACTIVE PARTICLE CONTAMINATION PROBLEM

COPY 1 OF 1, ~~2~~

By: H. M. Parker

Date: March 22, 1948

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REVIEW OF THE STACK DISCHARGE  
ACTIVE PARTICLE CONTAMINATION PROBLEM

Introduction

A report by M.L. Mickelson\*, two earlier reports by the writer\*\*, a report by R.S. Bell†, and numerous incidental references†† have defined the problem and indicated some of the actions taken to correct hazardous conditions. This report will attempt to bring this data up to date, and to include more of the studies and actions of all departments than appeared in the earlier H.I. reports. For this reason, and also because the emphasis on the various proposed tests has changed considerably, the paragraphing system of the earlier H.I. reports will be abandoned. As the contaminating agent has been variously referred to as specks, spots and active particles, it is proposed that a uniform title of "Stack Discharge Active Particle Contamination" be applied to the problem in future. The interpretation has to be broad enough to cover droplets, mist or spray as well as discrete solid particles.

1. Biological Investigations

The biological study has been limited to a literature review and to a sketchy exploratory experimental program. The literature review was essentially worthless. Reported results were concerned primarily with the biological effects of small radon implantations with either glass tubes or gold seeds. There were no data for sources substantially less than 100 activity. The search was made at a time when fairly sizeable particles (approximately 0.1 um) were considered. It is now clear that the actual problems range all the way down to ultra-fine particles - effectively aerosols. The rather extensive literature on radioactive aerosol contamination has not yet been reviewed. It is our understanding that such results have not been particularly concerned with, or successful in detecting local damage in the lungs from the deposition of fine particles. Dr. P. E. Church has indicated that the Chemical Warfare Service has made extensive investigations on the sizes of particles that can penetrate the human defense and enter the lung. This source is to be tapped, and may lead to a concentration of the biological investigation on a rather restricted range of particle size (perhaps 1 to 5 microns).

Project work on aerosols indicates that Y, Ce and Pr are in the small group of fission products which is essentially retained in the lung. As these three are responsible for most of the activity of the particles in question, it is considered certain that the limiting hazard will be the

\*"Preliminary Report on Existing Active Particle Hazard - 200 Areas".  
M.L. Mickelson - 10/22/47 (Doc. HW-7865)

\*\*"Action taken on the Spot Contamination in the Separations Plant Areas" °  
H.M. Parker 10/30/47 (Doc. HW-7920) Progress Report on "Action taken on the Spot Contamination in the Separations Plant Areas" H.M. Parker 1/20/48 (Doc. HW-8624).

†"Active Particle Investigation - 200 Areas - Establishment of Physical Control"  
R.S. Bell 11/26/47 (Doc. HW-8108)

†† The first documented reference is apparently in "H.I. Report #143 on the 200 Areas and Associated Laboratories Week Ending 10/1/47" C.M. Patterson to W.K. MacCready (Doc. HW-7693)

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local irradiation of lung tissue.\* In any case, the total activity involved is so low that assimilation into the blood stream and subsequent deposition in an organ would certainly be innocuous, unless one has to suppose that aerosols have been inhaled since the start of operations. One speculates that even this would be entirely safe, and that radioactivity would be demonstrable in the urine or feces long before a hazardous accumulation had been achieved. No such activity is found.

Biological experiments have been as follows:

a. Solubility of the active particles in human gastric juice

Solubility was negligible in the juice of each of two subjects tested (order of 1% dissolved).

b. Absorption in G.I. tract of guinea pigs, rats and rabbits

There was insignificant absorption in all trials. Both (a) and (b) are somewhat surprising in view of the known high absorptability of Ce. In both experiments the specks were broken down by the gastric juice into much smaller particles. The initial introduction of very fine particles might well have led to much greater absorption.

c. Tracheal cannulation of rabbits

The active particles were removed, presumably by ciliary action, in the four specimens.

d. Surface irradiation of rabbit ear

Four animals were exposed for times between one and four months to specks of about 0.5 to 1  $\mu$ c activity. Dubious reaction was noted in one instance, in the form of recurrent reddish brown pigmentation. The reader is entitled to note that the work was done with reddish brown specks of contaminated iron oxide. A previously reported thickening of an ear also has an equivocal status. Inactive control specks produced no irritation reactions.

e. Subcutaneous implantation - rabbits

The implants have been in place in four rabbits for 3½ months. No surface reaction is observed. The animals have not yet been sacrificed for histological study.

f. Testicular implants - rabbits

Four animals were implanted with active particles, together with 3 inactive control specks. One animal, sacrificed at two months showed no gross lesion, but there was no sperm within a radius of 2 mm. of the speck. Also there was an increase in the amount of tissue accepting an acid stain (eosin). Varying degrees of cell necrosis were noted. None of these changes occurred in the control testicle.

\* Sr is reported to be rapidly absorbed from the lung. This would remove the source of daughter Y90.

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One rabbit was given a testicular implant with a low radon seed. The animal was sacrificed after 2 months; the sections have not yet been studied. The purpose here was to correlate active particle damage with the more or less known damage from radon seed implants.

#### 6. Exposure of rabbits near the T Plant Stack

Eighteen animals so exposed have shown no activity due to particles on the fur, except on the feet. Whether mildly active particles are in the lung can only be found by autopsy. The one animal sacrificed was clean.

#### Summary of thinking on biological aspects

The available active particles can produce radiation damage. There will be no visible reaction on the skin.

The critical hazard is the inhalation and lung retention of particles. Very small particles (1 to 5 microns) will be the principal offenders. It will be impossible to define the safe permissible activity of such particles in a short term experiment.

Continued absence of radioactivity in the feces could be used as presumptive evidence of insignificant deposition following ingestion of particles. This is also a probably adequate determinant of absence of generalized lung deposition on the assumption that a significant fraction of the initial deposition would be removed and would follow the ingestion route. It is proposed to ask Dr. R. E. Zirkle and Dr. S. T. Cantrell for a more authoritative interpretation of this work, and for guidance on the future program.

## 2. Discovery and Origin <sup>of</sup> the Particles

### General

The occurrence of contamination in the form of discrete active particles on the ground in the Separations Plants was detected in late September 1947, by H. I. surveyors. It followed from the finding of activity at the base of the T Plant Canyon, from recurrent contamination of a path outside the B Plant Control Laboratory, and from shoe contamination of a patrolman, which led to finding contamination near a guard tower. By mid-October the general distribution pattern around the T & B Plant stacks was sufficiently well defined to make it certain that the particles emanated from the stacks. At that time, it was felt that the phenomenon might have existed for several months without detection. A review of the contamination data suggested that the magnitude of the effect had been increasing quite rapidly prior to its discovery.

The particles were originally separated by hand from the adjacent sand grains. It was later found that they could usually be distinguished by eye, due to their reddish brown coloration. Chemical analysis showed an approximate 50% content of  $\text{Fe}_2\text{O}_3$ , and about the same time it was realized that the

# Contaminated soil detected near the T Plant fence during the week ending 8/23/47 (Doc. EW-7435) had been proved to contain long lived fission products (principally Ce and daughter Pr) by 9/10/47 (Doc. EW-7539)

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particles could be separated by means of magnets. This greatly facilitated the preparation of samples for chemical and physical study.

#### Origin

These studies proved the presence of long-lived fission products only, together with trace amounts of alpha-emitters. Prominent inactive elements present were Bi, Fe and Si. By late November, it had been established that the carrier particles were corrosion products from black iron in the stack fan ductwork, which was supposed to have been stainless steel throughout. In early November, a program\* was initiated to install filters in the ventilation ductwork as close to the stacks as possible, to handle all process building ventilation air. The radiation hazards involved in accepting emergency design and installation of such filters was appreciated by all concerned. Consequently, when the origin of the main carrier particles was assigned to the improper ductwork, it was logical to suppose that correction of the ductwork was an adequate step for the immediate future, leaving the already proposed full study of the filtration problem to a later date. Orders were changed\*\* to postpone the installation of filters indefinitely and to proceed with the fan and duct replacement. This change was in any case urgent to avoid the risk of corrosion to the point of collapse.

The work of the Technical and H. I. Departments subsequently made it clear that although the large particles on which attention was focussed in the earlier studies were eliminated by the fan changes, numerous smaller particles were still being emitted. This was essentially known from the measurements by F. P. Seymour in February 1945. The then-assumed droplet contamination was assumed to be dispersed and diluted in the same manner as the iodine, which was then the critical contaminant. Calculations of the possible concentration of fission products at the ground (approximately  $10^{-11}$  to  $10^{-10}$  mc/liter) did not attract attention as a significant hazard. Sporadic measurements of the deposition on vegetation have been made with no more than traces located.

Specifically, the large active particles ( $>0.1$  mm.) came from the corroded ductwork. At the present time there are numerous small particles, some of which are magnetic and therefore probably from the same source, together with ultra fine particles or droplets directly from the process vessels. Such droplets may also initially contaminate dust particles pulled through the ventilation system.

#### Distribution of active particles

This problem logically divided into Technical Department studies of the stack emission, and H. I. studies of the subsequent deposition. Necessarily, there has been overlap and effective cooperation in the field between the departments. Superficially, there have been apparent conflicts in the various reports. None of these is real, and most of them have been due to a progressive increase in skill in detection of invisible particles. There has been neither time nor manpower to revise the earlier data to make it consistent in this respect.

\*Project Design Request to W.W. Fleasants from W.K. MacCreedy - 11/3/47

\*\*Stack gas decontamination - Suspense Code 10,225, W.K. MacCreedy to W.P. Overback 1/8/48.

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a. The Technical Department program on Stack emission

The Stack Gas Disposal group transferred its interest from the I131 problem to the particle problem on 11/1/48\* (Doc. HW-8074). Four principal sampling positions with filters were established in the B Plant.

1. Canyon air prior to entering the fans - through the air balance duct. Available 11/17/48.
2. Canyon air beyond the fans - through the access door in duct. Available 11/23/48.
3. Dissolver off-gas. Available 11/30/48.
4. Stack air at the 50 foot level. Available 11/17/48.

Typical sample results were:

| <u>Sample</u> | <u>Duration<br/>Hours</u> | <u>Flow<br/>CFM</u> | <u>Activity<br/>on filter<br/>mrep/hr at 2"</u> | <u>Remarks</u>                  | <u>Reference</u> |
|---------------|---------------------------|---------------------|---|---------------------------------|------------------|
| F1-1          | 5                         | 10                  | 1500  | 2 particles                     | HW-8234          |
| F1-2          | 18                        | 10                  | 1500  | 1 particle                      | 11/16/47         |
| F4-1          | 2                         | 10                  | 325   | 400-500<br>particles            |                  |
| F4-2          | 2                         | 10                  | 5 (at 4")                                       | less than F4-1                  |                  |
| F4-4          | 22½                       | 10                  | 3300  | < 25, but hot                   |                  |
| F4-5          | 5                         | 10                  | 1500  | > 100 particles                 |                  |
| F2-1*         | 4½                        | 11                  | 480   | many particles                  | HW-8235          |
| F2-2**        | 6                         | 11                  | 180   | more particles                  | 11/23/47         |
| F2-3          | 4                         | 15                  | 350   | most & hottest                  |                  |
| F4-7*         | 4½                        | 10                  | 350   | fine size hot<br>particles      |                  |
| F4-8**        | 6                         | 10                  | 300   | like F4-7                       |                  |
| F2-4          | 24½                       | 10                  | 3000  | most particles<br>yet collected | HW-8422          |
| F2-5*         | 10½                       | 10                  | 900   | fewer particles<br>than usual   | 11/30/47         |
| F4-9*         | 10½                       | 10                  | 1000  | very few particles              |                  |

\*All dates in this section are "week beginning" dates taken from the Weekly Reports  
200 Areas Technical Division, J.B. Work to O.H. Greager

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| <u>Sample</u> | <u>Duration<br/>Hours</u> | <u>Flow<br/>CFM</u> | <u>Activity<br/>on filter<br/>mrep/hr at 2"</u> | <u>Remarks</u>               | <u>Reference</u> |
|---------------|---------------------------|---------------------|---|------------------------------|------------------|
| F2-6**        | 17-1/3                    | 10                  | 480   | fewer than<br>usual          |                  |
| F4-10**       | 17-1/3                    | 10                  | 480   | pract. no<br>particle        |                  |
| F3-1          | 2                         | 10                  | 3000  | no dissolving<br>no particle |                  |
| F3-2          | 2                         | 10                  | 33,000  | first out -<br>no particle   |                  |

ductwork and one fan replaced at this point

|         |        |      |      |                               |         |
|---------|--------|------|------|-------------------------------|---------|
| F2-9*   | 23.7   | 10.5 | 7500 | many particles                | HW-8704 |
| F4-11*  | 23.8   | 10   | 3000 | no particle                   | 1/11/48 |
| F2-10** | 24     | 10   | 2500 | several small<br>particles    |         |
| F4-12   | 24     | 10   | 2500 | one large one                 |         |
| F2-11*  | 25     | 10   | 3000 | none visible<br>H.I. found 47 | HW-8705 |
| F4-13*  | 25     | 10   | 2500 | 7 visible<br>H.I. found 57    | 1/18/48 |
| F1-5    | 23 1/2 | 12   | 5000 | none visible<br>H.I. found 1  | HW-8703 |
| F1-6    | 72     | 12   | 8000 | none visible<br>H.I. found 40 | 1/25/48 |

\* and \*\* indicates simultaneous samples.

F1, F2, F3, F4 series refer to the four sampling ports defined.

The general conclusions deduced from these data were:

Prior to dust change

- Essentially all particles originated after position 1. ie: in the dust not from the process vessels.
- The correlation between number of particles and integrated flow was poor.

After Dust change

- The first samples were high despite the washing of the system.

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b. This rapidly cleaned up, and ultimately

- (1) The number of visible particles was reduced multifold
- (2) Large particles, readily visible, were practically eliminated
- (3) Small particles were present but in far smaller numbers

By 2/1/48 (Doc. HW-8954) it was clear that "Another serious problem beyond that caused by the fan dusts" was present. By 2/22/48, (Doc. HW-9068) plans had been laid to study the effects of electrostatic precipitation, of filtration and of water scrubbing.

b. Total deposition of particles on the ground

The first measurements by the E. I. Department were limited to Geiger counter surveys of the terrain of immediate interest.

The first comprehensive survey of the B Plant Exclusion Area was reported on 11/28/47.\* This showed a circular area (Zone 1) of 50,000 square feet centered about the stack with 5 to 10 particles per square foot, of activities mainly in the range 0.05 to 0.5 mc each. This was surrounded by Zone 2 of area 315,000 square feet, pulled out in fingers 1200 feet long to the southeast, as dictated by the prevailing northwest wind. Zone 2 had a concentration of 1 to 5 paf\*\* of activities from 0.01 to 0.05 mc. Zone 3, 2,300,000 square feet was elongated 2700 feet in the same direction with a width of approximately 1300 feet. Upwind from the stack it reached only 400 feet. This zone had about 0.01 paf of activity less than 0.01 mc. The total number of particles was  $1.4 \times 10^{10}$ , and the total deposited activity was 100 mc. Neither figure can be claimed accurate to better than a factor of 5.

In a repeat of this survey, completed 1/21/48, the following was found:

- Zone 1 - 180,000 square feet
- Zone 2 - 600,000 square feet
- Zone 3 - redefined to cover about 0.1 paf 2,000,000 square feet

The total deposition was  $3.4 \times 10^6$  particles and 300 mc. The additional deposition in this approximately two-month interval was therefore nominally  $2 \times 10^6$  particles if there was no appreciable loss by transport of existing particles. If one is permitted to make very sweeping generalizations from the data on stack emission, particles were being emitted at the rate of  $10^7$  to  $10^8$  particles per month.† The observed deposition was 1-10% of the emission, which at least is reasonable. Let it be emphasized that both calculations are based on scant evidence, especially as to whether the same limits on particle size applied in both cases. The reader is privileged to speculate on the present location of the 90 to 99% of the particles unaccounted for. An alternative check on the deposition is arrived at from the integrated activity.†† This shows a 1% deposition. It is of interest to note that the data predicts a removal of iron from the black iron parts at the rate of approximately 1 mil per month.

\* Particle Distribution - 200 East - L.O. Boos to File, 11/28/47 (Doc. HW-8430)

\*\* Let us use paf to avoid tiresome repetition of "particles per square foot".

† Assumption - 20 to 200 particles on a filter after a 4-hour run at 10 cfm.

†† Assumption - F-2 or F4 filters show 300 mrep/hr at 2" after a 4-hour run at 10 cfm.

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The corresponding survey of the T Plant was recorded on 12/22/47.\* Zones 1 and 2 were much more extensive, and formed approximately circular patterns centered around the stack. Zone 3 was elongated to 2700 feet downwind, but here extended 800 feet upwind.

Zone 1 - 560,000 sq. ft.,  $4 \times 10^5$  particles  
 Zone 2 - 840,000 sq. ft.,  $2.5 \times 10^5$  particles  
 Zone 3 - 4,200,000 sq. ft.,  $4 \times 10^4$  particles

Total deposition =  $6.5 \times 10^5$  particles.

The distinguishing feature of the distribution was the inclusion of all the Exclusion Area in the dangerous zones 1 and 2. This decided the eventual boundaries of defense by the use of respirators.

One month later, the deposition picture in this area had barely changed in pattern. It showed the addition of approximately  $3 \times 10^5$  particles. More recently, the area of search has been widened, principally by the examination of potential building sites. The following areas were extensively surveyed in mid-March:

#### Area A

A plot 1200' x 800' due south of the T Plant stack and about 7,000' distance. Completed 3/9/48

No particle was found in a 0.02% area sampling in a grid pattern.

The statistical value of this result is:

If the true deposition had been 0.01 pcf, there would be one chance in 10 of finding no particle.

True deposition 0.02 pcf - one chance in 100 of finding no particle.

#### Area B

A plot 1400' x 900' adjacent to and east of Area A. Completed 3/20/48

Three particles, maximum activity 0.004  $\mu$ ci in a similar sample

Statistics - nominally approximately 0.01 pcf.

True deposition 0.028 pcf - one chance in 20 of finding 3 particles

True deposition 0.033 pcf - one chance in 100 of finding 3 particles or at the opposite limit

True deposition 0.003 pcf - one chance in 20 of finding 3 particles

True deposition 0.004 pcf - one chance in 100 of finding 3 particles

#### Area C

A plot 1200' x 1200' adjacent to and north of the T Plant Canyon Completed 3/9/48

Surface concentration = 3 pcf

Maximum activity of a particle = 0.3  $\mu$ ci. Average activity = 0.007  $\mu$ ci.

\* Particle distribution 200 West - L.V. Zuercher to File, 12/22/47 (Doc. EW-8429)

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Area D

A plot 400' x 400', 3000' due east of the B Plant stack, at the location of the original C Plant. Completed 3/9/48

Surface concentration = 2.5 pcf.  
Maximum activity = 0.016  $\mu$ o  
Average activity = 0.002  $\mu$ o

Area E

A plot 800' x 2200', contiguous with Area E, and lying 2500' to 3500' S.E. of the B Plant stack. Completed 3/20/48

Surface concentration = 1 pcf  
Maximum activity = 0.035  $\mu$ o  
Average activity = 0.004  $\mu$ o

Area F

A plot 500' x 2200', S.E. of the B Plant stack, and 1000' to 2000' away. This is close to many of the servicing work areas. Completed 3/22/48

Surface concentration = 1.8 pcf  
Maximum activity = 0.2  $\mu$ o  
Average activity = 0.009  $\mu$ o

Area G

A plot 1100' x 600', due west of the B Plant stack, and 1000' to 2000' from it. Completed 3/18/48

Surface concentration = 1 pcf  
Maximum activity = 0.01  $\mu$ o  
Average activity = 0.004  $\mu$ o

Area H

A plot 3500' x 2000', N.W. of the B Plant stack, and 2000' to 5000' away. Completed 3/17/48

Surface concentration = approximately 0.003 pcf  
No activity in excess of 0.004  $\mu$ o.

Area J

A strip 5800' x 700', about 1 mile S.E. of the B Plant stack. This parallels the south side perimeter road. Completed 3/19/48

Surface concentration = 0.04 pcf  
No activity in excess of 0.004  $\mu$ o.

It will be noted that the depositions in Areas D, E, F, G greatly exceed the previously reported surface concentrations in the originally defined Zone 3. There is some evidence that apparent increase of concentration with increasing distance from the stack is real. Nevertheless, it is likely that Zone 3 is now much more widely contaminated than was reported

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on 1/21/48. If the "skip distance" effect is real\*, the required survey program reaches a magnitude beyond that originally contemplated. It will occupy at least 500 man weeks. The currently available force is 5 men; by borrowing of manpower from other departments, and by emergency purchase of instruments, the force can hardly be brought to more than 35. A comprehensive survey thus cannot be recorded in less than 3 months, by which time the overall picture may have changed completely. Radical corrective measures at source are clearly indicated.

All the above surveys have been considered in terms of detection by Geiger counters. It is known that radiocautographs may show many more particles than can be found by counter methods. The relative detection coefficient is a function of the surface concentration, mainly because the standard counters used cannot differentiate between closely spaced particles, and the radiation from weak particles is readily obscured by strong neighbors.\*\* As now operated, the following coefficients apply:

| <u>Surface density by counter</u> | <u>Detection coefficient for radiocautographs</u> |
|-----------------------------------|---|
| 15 - 30 pcf                       | 12 to 20  |
| 8 - 15 pcf                        | 5 to 12   |
| 4 - 8 pcf                         | 1 to 5  |
| < 4 pcf                           | Approximately 1                                   |

Whether the weak particles detected by refined methods are hazardous is not known. As an outright guess, the writer is not unduly concerned about activities less than 0.01 uc each. Whereas the earliest particle data covered the range 0.1 uc to 3 uc, current detection methods go down to  $10^{-3}$  uc, by counters and perhaps to  $10^{-4}$  uc by radiocautographs. It is infeasible to correct the early work to the standards of the new; the steady improvement in technique should not be overlooked in a review of the data.

c. Rate of deposition of particles

Attempts have been made to establish the rate of deposition by the exposure of rugs, and later of glass cloth surfaces for intervals of about one month. These catching surfaces were screened from the wind action by the superposition of expanded metal mesh, and supported 3 feet above the ground to minimize receipt of particles from adjacent ground. Although elimination of wind transport is not claimed, the results should give the combined deposition from the stack, plus whatever is picked up from the ground and redeposited. Until recently the deposition by this method was found to fall into a pattern quite similar to that shown by the total deposition to date. Zones of maximum deposition rate showed 4 to 8 pcf per month. During the month of December 1947, the integrated deposition in the 200 East Area Zones 1, 2 and 3 was 650,000 particles by this method. It has previously been shown by subtraction that 2 million particles appeared between 11/28/47 and 1/21/48. It is reasonable to assume that one million appeared in December. The two approaches are therefore in good agreement, within the accuracy limits of each procedure.

\* "Spot contamination on potential building sites" W.M. Parker to W.P. Overbeck 3/10/48 (Doc. HW-9141)

\*\* Counter detection could be improved by placing a metal grid over the suspected area to eliminate side radiation from one spot to another. With a counter shielded to an area equal to the grid mesh, very sensitive separation would be possible.

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The influence of the 200 East Area fan and duct change has been studied by observing the deposition rates on catchers for the month of December 1947, and for a similar period beginning 1/15/48. In the Zones 1 and 2, the total deposition of particles in these two periods was either the same or not reduced by a factor as much as two. The change did eliminate the large and usually most active particles so that the average activity of deposited particles fell by a factor of 3. Only in the apparent small reduction in the number of small particles does there appear to be some divergence from the Technical Department reports, which suggested a much more substantial reduction. In late February and early March a series of collections was made in a line extending due north from the T Plant stack. These clearly show the skip-distance, or increase in concentration with distance, and thus substantiate the suspicion of this from the total deposition data.

| <u>Distance from Stack</u><br><u>Feet</u> | <u>Rate of Deposition</u><br><u>Psf per Month</u> |
|---|---|
| 500                                       | 3 - 6   |
| 1500                                      | 6 - 12  |
| 2500                                      | 12 - 18   |

d. Concentration of active particles in the atmosphere

This has been measured by suction at 3000 cfm. through glass cloth filters in the expected location of maximum concentration in the B Plant.

Typical results were as follows:

| <u>Date</u> | <u>Duration</u><br><u>Hours</u> | <u>Particles</u><br><u>No.</u> | <u>Activity</u><br><u>Range (uc)</u> | <u>Concentration</u><br><u># per 10<sup>6</sup> cft</u> | <u>Activity</u><br><u>Concentration</u><br><u>uc/liter</u> | <u>Remarks</u> |
|-------------|---------------------------------|--------------------------------|--------------------------------------|---|--|----------------|
| 1/8/48      | 5.5                             | 2                              | 0.01-0.015                           | 2.2   | 10 <sup>-9</sup>   | pre-fan        |
| 1/16/48     | 168                             | 6                              | 0.005-0.01                           | 0.2   | 6 x 10 <sup>-11</sup>                                      | change         |
| 1/27/48     | 188                             | 1                              | 0.004                                | 0.03  | 4 x 10 <sup>-12</sup>                                      | fan<br>changed |
| 2/10/48     | 240                             | 1                              | 0.035                                | 0.023   | 3 x 10 <sup>-11</sup>                                      |                |
| 2/27/48     | 372                             | 50                             | 0.001-0.03                           | 0.75  | 2.5 x 10 <sup>-10</sup>                                    |                |
| 3/12/48     | 341                             | 56                             | 0.001-0.01                           | 0.9   | 1.8 x 10 <sup>-10</sup>                                    |                |

With the exclusion of the short run, the highest particle concentration corresponds with the expectation of a man inhaling one particle in 150 months. If this data could be accepted, at face value, the urgency of solution would be removed. It should be noted that:

- (1) There is now much less certainty about the area of maximum concentration.
- (2) The glass cloth filter is probably a poor catcher of very small particles.

Other air collections by means of the familiar Big Suckers have shown only active dust, not resolvable as particles in a total sample of 180,000 cft of air.

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Attempts to calculate the probability of inhalation of particles from the rate of deposition on the ground and from the rate of emission from the stack. These depend on a knowledge of the rate of fall, computed to be 1 foot per sec. for a particle of radius 0.03 mm. In zones of heavy concentration these methods led to inhalation rates of about one particle per week. The particles transmissible to the lung probably float in the air or at least have lower effective terminal velocities. The quoted inhalation rate is probably not conservative. The writer is unable to reconcile this with the data from the large air samples.

#### Summary of Sections 2 and 3

The active particle contamination was detected by follow-up of small deviations from the normal pattern of contamination. All the large and most active particles were separable by magnetic means. The source or at least the prime source was corrosion of black iron ductwork in the Separations Plants ventilation system. Removal of the offending ductwork eliminated the large particles, but had relatively little influence on the emission of smaller particles. The emission is estimated at  $10^7$  to  $10^8$  particles per month. Ground deposition in the Exclusion Areas is approximately  $5 \times 10^5$  to  $10^6$  particles per month. The highest known surface concentration is 50 pcf in isolated areas near the stacks or where there has been obvious wind drift. The highest concentration over an extensive area is about 10 pcf.

The contamination is by no means confined to the originally studied zones in the Exclusion Areas. Concentrations as high as 2.5 pcf have been found 3000' east of a stack.

There have been recent indications of a sharp increase in the number of particles on the ground. Whether this is due to increased emission or to subdivision by abrasion of existing particles is unknown. The increase is estimated to exceed the admitted increase in skill in detection. Radiotelescope methods are approximately ten times as effective as Geiger counter methods.

The probable inhalation rate from tests with collection on filters is one particle per 150 months. This technique may miss all very small particles. Calculations indicate an inhalation rate of one per week for small particles. It is not known whether these are hazardous.

#### 4. Defensive Measures - "S" Department Program

Close cooperation between the "S" Department, the Technical Department, and the H. I. Department was maintained at all times, with essential agreement on major policy items. Undocumented discussions immediately following the discovery of the hazard were concerned with:

- a. The feasibility of having all men in the affected areas wear respirators; the reality of the hazard was assayed against the morale damage of suddenly requiring such protection
- b. The feasibility of removing the contaminant by filtration of all stack air, including such possibilities as cleaning the stack.

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That the particles came from the stack was known before the precise origin was located.

The proposed future recommendations of the H. I. Department for various contingencies was defined in the writer's Doc. HW-7920 - 10/30/47. This included tacit agreement with the objective of further defining the hazard, before respirators were requested.

Designs to permit the following installations were requested on 11/3/47.\*

- (1) Temporary air filter to be located in the 291-T, B Buildings between the existing fans and stacks, and
- (2) A permanent filter system to be installed in the 291-T, U, B Buildings between the existing fans and new or revised stacks, the design for which is likewise to be prepared.
- (3) A permanent set of filters to be installed in 221-T U, B Buildings in the individual cell exhaust ducts and the 224-T, U, B Buildings tank vent piping.

The objectives were (1) To remove all particles originating prior to the stack immediately and at the risk of subsequent hazardous maintenance (2) To follow with a permanent installation including new stacks. (3) To filter the air from each separate cell of the process buildings. The proposed actions of the various collaborating departments were defined in line with the above intentions on 11/26/47.\*\*

By 12/9/47, it had been fully realized that the principal offending particles were corrosion debris of iron ductwork. This was primarily developed from the Technical Department sampling results, and was supported by inspections of the clean U Plant ventilation system to determine the extent of ductwork other than the prescribed stainless steel.† Plans were promptly laid to replace the duct and blowers, because this change could be completed in half the time and at twenty-five percent of the cost of the previously scheduled temporary filters.†† It was a reasonable expectation that these repairs would remove the source of the contaminated particles, and therefore minimize or eliminate the need for filters. Changes in the applicable Suspense Code to handle the revised program were documented on 1/9/48.†††

In the meantime, the impracticability of arriving at a speedy factual evaluation of the existing hazard was appreciated.

The recommendation to wear respirators in dubious zones was consummated.††† A cheap disposable unit, the Martindale Protective Mask had been located for this purpose. Although it was recognized that such filters had low efficiency for small particles (1-2 microns), namely those most likely to be inhaled and retained, there was then no evidence of important activity on the small entities. Plans were made to survey the removed

\*Installation of air filters - 200 Areas W.K. MacGready to W.W. Fleasants 11/3/47 (Doc. HW-7932)

\*\*Doc. HW-8108

†"The 291-U Duct and Fan Assemblies relative to the Particle Problem" L.C. Hode to M.L. Mickelson, 12/8/47

††"291 Fan Replacement", W.K. MacGready to G.W. Gross, 12/9/47 (Doc. HW-8245)

†††Stack Gas Decontamination - Suspense Code 10,223, W.K. MacGready to W.P. Overbeck, 1/8/48

††††"Special Regulated Area", W.K. MacGready per V.R. Chapman to 200-W Area Supervision 1/2/48

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pads at the end of each workday. This protection was applied throughout the T Plant Exclusion Area, in agreement with the existing survey data. In the B Plant, Comfo respirators were required in a smaller specially restricted zone near the stack.

The current plan of action was defined in a joint memorandum of the Technical Department and "S" Department on 1/27/48.\* This memorandum reiterated the reasons for the initial attack on the ductwork, but indicated clearly that the immediate particle problem was only a fraction of the total long-lived emission from the stacks.

A study of steps to eliminate the primary mist or droplet contamination at source was discussed, together with plans to revert to temporary filters if the ductwork change proved inadequate. Long-range plans for full decontamination were established, and the application of similar solutions to new separations installations discussed.

These procedures were amplified in a later document.\*\* A summary of studies was given, with the technical and operational criteria for successful decontamination. Intended action was defined under the following headings:

- (1) Project Engineering Department to procure and/or fabricate and install test units, including one electrostatic precipitation unit and a CMS Type #6 filter unit. Consultative assistance from the Kellier Corporation to be solicited.
- (2) Technical Department to justify or improve present sampling methods. Consultative guidance of Dr. Langmuir and Schaefer were to be obtained by Technical Department and E. I. for this, and for an analytical study of the process effluents.
- (3) Consideration of sections that could be turned over to off-site groups. Inability to reproduce separations plant ventilation air conditions elsewhere, an insurmountable barrier to off-site assignment of current local problem. The function of the Air Reduction Company to engage in long-range, overall studies.

Along with the above long-range studies, speedy and sometimes emergency measures were organized by the "S" Department. Thus, when a pessimistic view of the continued appearance of particles was submitted by the writer on 3/19/48, temporary filters were designed, fabricated and installed in each cell of the B Plant Canyon by 3/22/48. Similarly, when the surface concentrations in Area E were announced, steps were taken to remove personnel from the Garage in that region, pending further study.

Considerable attention has been focussed on the contamination of proposed sites for future expansion, and surveys of the Areas A, B, C, D and E were requested on this basis. It has been assumed that the stack emission would shortly be controlled, and that the eventual limiting hazard would be that arising from the re-injection into the atmosphere of such deposited particles as could not be cleaned off or permanently fixed. Provisionally, a current surface concentration of 0.01 pcf or less has been considered tolerable.

\*"Stack gas decontamination - Separations Plants" - O.E. Greager - W.K. MacCready to File, 1/27/48 (Doc. HW-8667)

\*\*"Stack gas decontamination, Separations Plants - Development of Decontamination Systems" - C.E. Gross to File, 3/12/48 (Doc. HW-9173)

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On this basis, one important change in the construction program has already been dictated.\* It is conceivable that further changes will be required.

### 5. Transport of Deposited Particles

While it was realized at the start of the investigation that deposited particles must be moved by the agency of winds, rain, thermal currents, etc., very little work has been done to date on this phase. Significant accumulations have been observed on the <sup>W</sup> side of buildings, near the stalks of desert plants, and even in natural gullies.

The catching surfaces to measure deposition were so designed to minimize wind transport of particles from the catcher frames and to avoid the traction load carrying already deposited particles.

A single experimental test was made. This involved the radio-autography of 10 square feet of desert surface, an exposure of the ground to wind for 7 days, and a repeat radiograph in register with the first. The initial test showed 62 particles, of which 47 remained. No particle remained in exactly the same position. As the individual particles were not distinguishable from each other, it is impossible to state how far each moved, or in the extreme case whether any of the 47 residual particles was a member of the original colony. The data were compatible with the removal of 15 small particles and a re-orientation of the remainder by inches only.

Fortuitously, the wind blew almost exclusively from the northwest throughout this period, with an average velocity of 6 mph, and an unusual peak velocity of 30 mph in the same direction. There was therefore no agent tending to shuffle the particles back and forth about a mean position. Since the behaviour of deposited particles is expected to set the residual limit on health safety in the areas, the study of the problem has been assigned to Dr. P. E. Church, Meteorological Consultant. He was invited to consider:\*\*

- (1) How far the relevant particles can roll on the ground
- (2) How high they can rise
- (3) The possible concentrations at levels at which they might be breathed.
- (4) The maximum deposition at distances up to 50 miles.

Dr. Church was pessimistic about the feasibility of answering these points, without extensive experimentation at Hanford.† Some references were made available on the experimental transport of sand grains, (Institute of Meteorology, University of Chicago), the problems of wind velocity at various heights (Geiger, E. "Das Klima der Bodennahen Luftschicht"), and the theories on boundary layer effects (New York Academy of Science "Convection in the Atmosphere and Ocean). Dilution with increasing

- \* "Location of Redox Test Unit", W.P. Overbeck to Carleton Shugg, 3/5/48 (Doc. EW-9091)
- \*\* "Proposed Problem for Meteorological Consultant", H.M. Parker to W.E. MacCreedy, 3/11/48 (Doc. EW-9170)
- † "Report of Meteorological Consultation" P.E. Church - H.M. Parker 3/19/48 (Not yet documented)

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distance of horizontal transport is best found from Dr. Church's own work at Hanford.

The only readily soluble question was that of altitude. Small particles could ascend 10,000 to 15,000 feet in summer by thermal convection, and rarely to 3,000 feet in winter by mechanical turbulence.

The consultant was of the opinion that effort should be placed on attempts to remove or to contain the particles already deposited, rather than to solve the given problem. Although this is well taken for the immediate environs of the Plants, it still appears that limiting solutions will have to be sought. If the data of Sections 2 and 3 of this report are nearly correct, there is a backlog of tens or hundreds of millions of particles unaccounted for in the Plant vicinity. The responsibilities of the Company and of the Commission require the most energetic attempts to prove that no significant radiation hazard can arise from this source, or to correct an untoward condition regardless of cost.

Another phase of the transport problem is concerned with the accumulation of particles in rain. Such particles have been sporadically observed in rain samples, but never in amounts to cause alarm. Ingestion of a single particle is almost certainly innocuous due to the demonstrated low solubility. Similarly, the deposition of casual particles in open reservoirs of drinking water can be dismissed. Deposition of contaminated rain, with a subsequent airborne career for the dried-out particles is also no more than a part of the main transport problem, as it seems unlikely that the rain cloud would travel significantly further than a particle could travel in its own right.

#### 6. Removal or fixation of deposited particles

It is exceptional to find any member of the interested groups, who has not advanced a method for the removal or safe fixation of the active particles. Equally exceptional is the mention of a scheme that could be completed for substantially less than one million dollars. It is clear that the expenditure of \$100,000 on study of the problem is in order before a final policy is accepted, provided that immediate urgency for radiation safety is not required.

For interest, and also to encourage the transmittal of better solutions, some of the more feasible local proposals are indicated:

##### Removal

a. Combing of the area by a rake and magnet truck combination. Trial runs to date have shown poor percentage recovery, even where it is known that essentially all the active particles contained iron. The method would not attack the more general species of active particle that may be present.

b. Vacuum cleaning of the area - possibly with a magnet in the system to concentrate most of the activity for better controlled disposal. Trials have been reasonably successful.

\* "Report of Meteorological Section January 1943 to August 1944", P.E. Church et al., 3/24/45

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- c. Scraping of the surface layers and disposal of the contaminated matter in collection trenches. This is presumably the method of choice in areas where construction is to be undertaken. Elsewhere, it will tend to remove the natural protective barriers, and thus leave any particles, not successfully removed, vulnerable to thermal currents and wind.
- d. Biological removal - the planting of a crop metabolically attractive to the principal fission products involved with subsequent controlled disposal of the concentrate in the plants. The apparent low solubility of the activity from the particles contra-indicates this approach.

#### Fixation

- a. Washing with water to drive the particles, presumed at least an order of magnitude smaller than adjacent sand grains, into the interstices of the sand. According to Dr. Church this would offer a high degree of resistance to pick up from the boundary layer. Preliminary tests were disappointing, but the extensive test program outlined by Dr. Church may establish optimum conditions.

The water may come from natural rain, induced rain or by sprinkling.

- b. A cement wash to bind a fairly thick layer - technique encouraged by the success in producing "Concrete" floors in Richland basements without controlled mixing of components. Costly and judged by Project Engineers to be a poor risk under the severe temperature conditions of the desert.

- c. Continued wetting of the surface, even if water cannot drive the particles down. May alter the boundary layer conditions in the favorable direction by surface densification, and would reduce the thermal gradients leading to updrafts. Costly, and not a permanent solution.

- d. Road oil as a binder. Costly, possibly a fire hazard, and would kill existing vegetation, thus decreasing natural protection.

- e. Sodium silicate or similar chemical binders.

- f. Planting of grass. Reported to be a most efficient retainer if kept upright by adequate watering. Costly, but as some recompense would improve the amenities of the desert plants. The area cooling effect could conceivably increase the available safe hours for discharging by elimination of thermal up-drafts - a valuable characteristic with a heavy production schedule.

- g. Wind breaks - logs, telephone poles or the like distributed over the ground, coupled with periodic vacuum cleaner collections from regions where the particles would accumulate.

- h. Furrowing or any other roughening of the surface. (example - cover with gravel).

#### Summary of Sections 4, 5, 6

Protection of all personnel has been assured by whatever method required, within the limits to which the hazards are understood. An early start was made on the provision of stack filters and plans for new stacks were

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drawn up, if needed. When the main source was located as corrosion of ductwork, plans were changed to correct this condition first. Although, in retrospect, this may have been unfortunate, the decision was proper and logical in the light of existing or procurable knowledge.

Plans for ideal long-range decontamination have been adequately integrated with immediate temporary corrections, and consultative aid solicited.

A program has been made for the investigation of the transport of deposited particles, judged to be the residual hazard in the Plant Areas, and probably the only potential hazard to the general public off the reservation.

Methods for the removal or fixation of the deposited particles are described.

7. Chemical, physical and allied studies of the active particles

The early and positive assignment of origin of the major offending particles removed most of the urgency from these studies. It is appreciated that a very thorough study of the process effluents is ultimately required. The formulation of this program awaits the guidance of Mr. Langmuir and Schaeffer. A brief review of the exploratory work is included here. This will be more fully documented in a forthcoming report by R.C. Thorburn.

Visible particles were small, ranging from 0.02 mm. to 1.5 mm. linear dimensions. A bar graph presenting the frequency of various areas is shown, based on 111 specks. An area distribution of 67 smaller specks is also included. Little is known of the size distribution of very small specks.

Photomicrographs of particles were compared with

- (1) Rust from ductwork in the U Plant ventilation system
- (2) Cement and brickwork from the C Plant stack
- (3) Asphalt paint from the stack
- (4) Soil

The assignment to species 1 was tolerably convincing.

Chemical analyses for carbon, calcium, iron and silicon specifically fixed the rust source. The low carbon and calcium content also virtually excludes the stack proper as a prime source. This is important, because it is feasible to filter the whole system except the stack. Adherence of soil to specks accounts for occasional high silicon content.

The gross radioactivity range of investigated particles was from 2.5  $\mu\text{Ci}$  to 3.2  $\mu\text{Ci}$ , with the conventional simplified interpretation of the "curie" for beta emitters. Alpha particle activity, principally plutonium, was found as high as 3200 disintegrations per minute. There was statistically significant correlation between activity and particle surface area. The low correlation coefficient 0.348 has impeded the definition of hazard limits. Had this correlation been absolute it might have been feasible to discount the activity of those micron-range particles that could be retained in the lung. There was poor correlation between volume and activity, thus favoring the picture of deposition of spray on a rust surface.

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**TABLE I**  
**RESULTS OF CHEMICAL ANALYSIS**

|               | Specks Collected On Ground | Specks Collected On Ground | Specks Collected On Tacky Surface (rug) | Specks Collected On Filter in Breach | Specks Collected On Filter in Stack | Bolt Head Specks | Boreings From 1st Fan in U Area | Boreings From 2nd Fan in U Area | Boreings From 3rd Fan in U Area | Paint From Fan Dust in U Area | Floor Sweepings From Dust in U Area | Duriron From Boreing Hole | Cement From Inside O Stack | Paint From Inside O Stack |
|---------------|----------------------------|----------------------------|---|--------------------------------------|-------------------------------------|------------------|---------------------------------|---------------------------------|---------------------------------|-------------------------------|-------------------------------------|---------------------------|----------------------------|---------------------------|
| Area          | 200-E                      | 200-E                      | B Plant 200-E                           | B Plant 200-E                        | B Plant 200-E                       | 200-E            | 222-U                           | 222-U                           | 222-U                           | 200-W 291-U                   | 200-W 291-U                         | 200-W                     | 200-E                      | 200-E                     |
| Area          | 23                         | 35                         | 29                                      | --                                   | --                                  | --               | 50.2                            | 51.05                           | 53.7                            | 56.05                         | 26.4                                | 57.8                      | 55.4                       | 50.4                      |
| of Specks     | 22.22                      | 17.7                       | 14.8                                    | 9.3                                  | 7.3                                 | mg               | mg                              | mg                              | mg                              | mg                            | mg                                  | mg                        | mg                         | mg                        |
| Sample Weight | 1.46                       | --                         | --                                      | --                                   | --                                  | --               | --                              | --                              | --                              | 15.3                          | --                                  | --                        | --                         | --                        |
| % C           | .61                        | --                         | --                                      | --                                   | --                                  | --               | --                              | --                              | --                              | 3.65                          | --                                  | --                        | --                         | 16.4                      |
| % H           | --                         | --                         | --                                      | --                                   | --                                  | --               | --                              | --                              | --                              | --                            | --                                  | --                        | --                         | 39.2                      |
| % Ash         | 13.05                      | 1.85                       | 7.8                                     | 12.95                                | 2.96                                | --               | 28.65                           | 8.68                            | 2.78                            | --                            | 45.0                                | --                        | --                         | 6.74                      |
| % Si          | 42.5                       | 35.5                       | 33.2                                    | 38.1                                 | 61.4                                | --               | 13.15                           | 44.5                            | 51.05                           | 9                             | --                                  | 34.8                      | 2.05                       | 2.04                      |
| % Fe          | .91                        | 3.23                       | .95                                     | 1.66                                 | 0                                   | --               | 5.64                            | 2.39                            | 60.5                            | --                            | --                                  | 92.8                      | 45.7                       | --                        |
| % Ca          | 6.22                       | --                         | --                                      | --                                   | 1.23                                | --               | --                              | --                              | 1.48                            | --                            | --                                  | 5.1                       | --                         | --                        |
| % Si          | --                         | --                         | --                                      | --                                   | --                                  | --               | --                              | --                              | --                              | --                            | --                                  | --                        | --                         | --                        |
| % Pb          | --                         | --                         | --                                      | --                                   | 0                                   | --               | --                              | --                              | --                              | 20.8                          | --                                  | --                        | --                         | --                        |

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Absorption curves of the beta activity had an end point at 3 MEV, due to praseodymium.

Decay curves of a "new" speck from stack off-gas filters, an "old" speck from the ground, and of a radioactive (dried) drop from a hand rail were compatible with a half life of approximately 250 days, in a 100-day test, with evidence of slower decay of the "old" speck.

Radiochemical analysis of particles was begun on 9/8/47, before they were recognized as such. This was a contributing factor in their initial discovery. Particles recognized as such were first assayed on 9/22/47.

These and subsequent analyses indicated cerium, yttrium, strontium, ruthenium and cesium as the principal contaminants in that order.  $\text{Pr}^{144}$ , daughter of  $\text{Ce}^{144}$  is included with the parent, because of the short half life. This praseodymium will make the largest single contribution to the radiation damage, due to the high energy beta particle emitted. The other predominant pair,  $\text{Sr}^{90}$  and  $\text{Y}^{90}$  separated in the analysis without significant growth during the run. Back scattering and self-absorption errors up to 30% were accepted in these determinations.

It is noteworthy that specks showing alpha particle activity have a ratio of plutonium activity to uranium activity of about 20:1. This is much more nearly compatible with the dissolver content than that of any later stage. Did spray from the separate dissolver line coat the already activated particles coming from the dust? Few, if any, of the particles could have originated in the dissolver off-gas line. At one time there was much interest in the "age" of the emitted particles. The elementary calculation from the fission product spectrum may be invalid, due to selective effects throughout the process. Attempts were made to define the age from the two available isotopic ratios  $\text{Sr}^{90}/\text{Sr}^{89}$  and  $\text{Ce}^{144}/\text{Ce}^{141}$ . The latter failed on account of the complex disintegration scheme of  $\text{Pr}^{144}$  daughter, which made it impossible to determine microcurie content from observed beta disintegration rate. Separated  $\text{Ce}^{141}$  and  $\text{Ce}^{144}$  standards are needed for a precise calibration. The  $\text{Sr}^{90}/\text{Sr}^{89}$  determination was moderately successful by two methods:

- (1) Deduction of  $\text{Sr}^{90}$  activity from the activity of separated  $\text{Y}^{90}$  daughter. Balance of the total Sr activity ascribed to  $\text{Sr}^{89}$ .
- (2) Change in total Sr count (after milking of  $\text{Y}^{90}$ ) with time due to relatively rapid decay of  $\text{Sr}^{89}$ .

Ages between 100 and 300 days have been reported.

Special tests made have included:

- a. Extraction of fission products from spiked feces samples, with demonstration of approximately 100% yield.
- b. Extraction of radio-cerium (90% yield), yttrium (70%) and strontium (2%) from spiked urine samples by oxalate precipitate. Alternate extraction of strontium (90%) by iron hydroxide precipitate. These are the principal contaminants to be looked for now. Ultimately,  $\text{Ce}^{137}$  might become the most promising metabolic index. No satisfactory recovery for cesium has been reported.

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TABLE II  
RESULTS OF RADIO ANALYSIS

| Particle                   | "Old" Particles<br>From 200 West | "New" Particles<br>From 200 West | "Old" Particles<br>From 200 East | "New" Particles<br>From 200 East | Particles Collected<br>From Stack Cases<br>200 East | Particles Collected<br>From Breech Cases<br>200 East |
|----------------------------|----------------------------------|----------------------------------|----------------------------------|----------------------------------|---|--|
| % Iodine                   |                                  | 6.44                             | 3.51                             | 3.2                              | $4.7 \pm 1.3$                                       | 7.2  |
| % Cerium *                 | $39.4 \pm 20.0$                  | 49.4                             | $29.1 \pm 9.0$                   | $16.8 \pm 3$                     | 18.5  | 28.5   |
| % Yttrium                  | $17.1 \pm 7.0$                   | --                               | $28.3 \pm 9.0$                   | 2.0                              | 17.1  | 23.7   |
| % Rare Earths <sup>1</sup> | --                               | --                               | --                               | 1.13                             | 5.98  | 12.1   |
| % Strontium                | $3.0 \pm 2.0$                    | 6.3                              | $10.5 \pm 4.2$                   | 3.2                              | 7.9   | 8.4  |
| % Ruthenium                | $2.4 \pm 0.6$                    | --                               | --                               | 12.5                             | --  | 13.5   |
| % Cesium                   | $0.4 \pm 1$                      | --                               | --                               | 3.74                             | 0.77  | --   |
| % Zirconium                | $1.87 \pm 1$                     | --                               | --                               | --                               | 9.1   | --   |
| % Columbium                | $0.7 \pm 0.02$                   | --                               | $0.69 \pm 0.4$                   | --                               | 2.9   | --   |
| % Barium                   | --                               | --                               | --                               | --                               | 0.1   | --   |
| % Tellurium                | --                               | --                               | --                               | --                               | 0.7   | --   |
| Alpha Activity             |                                  |                                  |                                  |                                  |   |  |
| % Plutonium                | $88 \pm 4.1$                     | --                               | $95.0 \pm 10.0$                  | 66.8                             | 73.5  | 99.5   |
| Activity %                 | $2.9 \pm 0.8$                    | --                               | $5.5 \pm 0.2$                    | 2.08                             | 5.0   | 0.5  |

Measured activity before analysis = 100% for the beta and alpha components separately

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TABLE II  
RESULTS OF RADIO ANALYSIS

| Particle                   | Scrapings From<br>Fan Duct Work<br>200 East | Filter From<br>Stack Gases | Filters From<br>Dissolver Gases | Filters From<br>Tunnel Gases<br>200 East | Dust From<br>13 R Cell<br>200 West | Hand Ball Drop<br>From 200 East | Hand Ball Drop<br>from 200 West |
|----------------------------|---|----------------------------|---------------------------------|--|------------------------------------|---------------------------------|---------------------------------|
| % Iodine                   | --  | 8.9                        | 7.0 ± 3                         | --                                       | 2.93 ± 1.5                         | 7.9                             | 6.4                             |
| % Cerium                   | 21.9 ± 1.5                                  | 25.6                       | 24.6 ± 1.0                      | 8.2                                      | 3.0 ± 2.5                          | 30.3                            | 3.0                             |
| % Yttrium                  | 8.1 ± 0.1                                   | 23.9                       | --                              | 21.7                                     | 2.1 ± 0.5                          | 10.2                            | 12.9                            |
| % Rare Earths <sup>1</sup> | 1.7 ± 1.1                                   | .01                        | 5.3 ± 1.1                       | 0.71                                     | 0.8 ± 0.7                          | 1.5                             | 3.2                             |
| % Strontium                | 3.9 ± 1.5                                   | 23.5                       | 13.6 ± 1.5                      | 3.65                                     | 3.75 ± 7.2                         | 4.5                             | 77.0                            |
| % Suthenium                | 8.32  | --                         | 4.3 ± 1.5                       | 40.6                                     | 86.0 ± 14                          | --                              | 0.0                             |
| % Cesium                   | --  | --                         | 0.3 ± .15                       | --                                       | 0.1 ± .2                           | --                              | --                              |
| % Zirconium                | --  | --                         | 8.4 ± 0.9                       | --                                       | --                                 | --                              | --                              |
| % Columbium                | --  | --                         | 1.4 ± 0.1                       | --                                       | --                                 | --                              | --                              |
| % Barium                   | --  | --                         | 1.3 ± 0.2                       | --                                       | --                                 | --                              | --                              |
| % Tellurium                | --  | --                         | --                              | --                                       | --                                 | --                              | --                              |
| Alpha Activity             |   |                            |                                 |  |                                    |                                 |                                 |
| % Plutonium                | --  | --                         | 21.6 ± 10                       | --                                       | --                                 | --                              | --                              |
| % Uranium<br>Activity      | --  | --                         | 1.5 ± 0.8                       | --                                       | --                                 | --                              | --                              |

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c. No measurable solubility of particle contamination in distilled water was observed. This is quite surprising as one assumes throughout that the particle is either a carrier for the residue from evaporated mist, or residue with no carrier. Perhaps the original mist is acidic, and this property is lost by evaporation. The activity is removed by nitric acid.

d. Some spot contamination specifically not associated with carrier particles showed fission product percentages peculiarly different from those in the particles, with a very high strontium component in one case. This could have been a "young" droplet. Unfortunately, the  $\text{Sr}^{90}/\text{Sr}^{89}$  ratio was not determined.

e. Active particles have been separated from natural rain, but none was active enough for radio-chemical analysis. The dissolved activity in other rain samples has been found more or less similar to the particle activities, by effective half-life. A higher  $\text{Sr}^{89}$  content is not excluded. Such activity, known before the particle problem arose, has always been assumed to come from emitted droplets taken up by the rain.

8. Clinical tests - applied and proposed

Contrary to the writer's first estimate, it is now fairly certain that no visible reaction will result from prolonged exposure of the skin to a particle. No particle stronger than 0.3  $\mu\text{c}$  has been detected in the past two months, and the average activity has been about 0.004  $\mu\text{c}$ . The ingestion of either one hot particle or 70 small particles would not lead to a deposition of 0.01  $\mu\text{c}$ . There would be no damage and certainly no clinical symptoms from this. Inhalation of such particles would also probably not lead to absorption and deposition in body organs of more than this amount, either directly from the lung or by the secondary ingestion of particles ejected from lung structures. The remaining possibility is of chronic irritation by irradiation of lung tissue by a retained particle or particles. No particle can destroy more than a sphere of a few millimeters radius in tissue. The expectation of clinical signs might conceivably be deduced from evidence from chemical warfare non-radioactive irritants. It is provisionally deduced that it is valueless to make direct clinical observations on potentially exposed people.

The external radiation from a lung-deposited particle is below the feasible limit of detection, nor is the use of elaborate detection instruments for insertion in the bronchus worth while. (The external radiation can be measured in small laboratory animals).

Close inspection of the clothing and body of personnel has its place in the estimation of the likelihood of exposure. Several hundred men have been so examined, with the positive results confined to shoe contamination.

Inspection of respirators worn in contaminated zones is more indicative of the inhalation hazard. Twelve thousand respirators have been tested by Geiger Counter methods. Two contaminated masks were reported. One of these had 4 particles on the inner face of the filter pad, and there is considerable doubt as to whether these arrived there by breathing. The other was a legitimate capture of a particle on the outer face. Two thousand filter pads were tested by the more sensitive radioautograph method. Suspected spots totaled 122, but only 19 were confirmed by

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repeat checks. Miscellaneous artifacts very easily simulate small specks. The capture of these 19 specks was one of the deciding factors in emphasizing the current critical nature of the situation.\* The specks which escaped detection by counters, but registered on films probably had activity not in excess of one millimicrocurie each. There is considerable doubt that these were hazardous. Calibration of the film system has not yet been defined. It requires the exposure of known activities at different depths of penetration in the filter pads. The advantages of making simultaneous radio-autographs of both sides of the pads have been considered. The relative blackening of the two film spots in register would determine the position in depth of the active particle and thus improve calibration. Also, unduplicated spots could generally be disregarded as blemishes.

Other clinical attacks are limited to the inspection of urine, feces, sputum and nasal smears. The two latter have not been employed. Very extensive programs would be needed to get significant data. They will be used if a person without respirator inadvertently enters a zone of high particle concentration.

Routine urine samples have shown no long lived activity, nor is any to be expected in view of the low elimination by this route. Fecal examination is the most promising procedure. Only four samples have been run; these were all free from radioactive contamination. Joint consideration by the Medical and H. I. Departments is being given to the practicability of large scale analysis of feces. The primary objective would be to detect the elimination of particles deposited in the lung and then swallowed. Since the commonly quoted elimination half-life of particles in the lung is two months, and since less than 1% of the force can be expected to have inhaled one particle (guessing pre-mask emission), one expects to make daily examination of feces from one hundred men, for about 6 months (3 half-lives) before a single particle would show up. This represents a tremendous and unattractive program. Such an effort would be required in the event of breakdown of morale of the force. Although this morale is unquestionably high at Hanford Works, the prolonged wearing of respirators and the removal of men from certain buildings constitute the most severe test. Hence the need for a feasibility study of this action, which currently appears to be the only promising approach.\*\*

Access to autopsy material of exposed personnel would be instructive; unfortunately it is too infrequent to be of immediate value. Nevertheless, Dr. S. T. Cantril is being asked to define a method of preparing the total lung tissue for radiation analysis. This may be done by opening all bronchioles, alveoli, etc. for inspection by counters, by gross sectioning of the organ into slices not more than 1 mm. thick, by ashing or by digestion or better means yet to be studied. At all events, the procedure will be difficult. The liver will be another organ of interest. The execution of some such plan is expected to be essential for the long-range allaying of legitimate employee or public fears.

\* For estimated use-factor of 0.8 hour per mask, this shows that a man would have inhaled one particle per 2.5 weeks, in good agreement with the one per week calculations.

\*\* Some worried employees have already appeared at the Kadlec Hospital for chest X-rays.

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9. Inferences from past data

It has been shown that a review of minor discrepancies in survey data suggests a history of evolution of particles for some six months prior to the initiation of protective measures. It is also known that mist or droplets were being emitted from the start-up of operations, and that the hazard from these may have been greater than that calculated on the assumption of uniform dilution. Applicable early data to define the time origin of possible damage are important. The sources are four in number:

1. Filters from the stack gas sampling lines taken March 1945

These were the filters that defined the emission of fission products and of plutonium at a time when the  $I^{131}$  problem was overwhelmingly greater. Fortunately, decay measurements on some of these have been maintained for the past 1000 days. The  $Co^{60}$  half-life of 275 days is the predominant presumed activity. Recalculation of stack emission at the time of the sample shows approximately  $10^{-7}$   $\mu$ c/liter of long lived fission products, on two samples totaling 4 hours coverage. Recalculation of the concurrent emission of  $I^{131}$  as shown by these filters indicated about  $5 \times 10^{-3}$   $\mu$ c  $I^{131}$ /liter stack gas. As the filters were started on counter tests one hundred days after exposure, this value has been corrected by a decay factor of 22,000, so the expected precision is low. However, the average stack emission of  $I^{131}$  in March 1945 was  $2 \times 10^{-3}$   $\mu$ c  $I^{131}$ /liter from direct iodine scrubber measurements, so that the recalculated values are reasonable, and indirectly validate the concentrations for long lived activities.

Stack sampling to better than a factor of 3 has never been claimed, and J. B. Work believes that fluctuations by a factor of 10 are possible. It will be seen later that we are practically forced to the conclusion that the above value is low.

An important observation is that no discrete particle can be found on these filters.

2. Similar filters taken February to July 1947

Filters over a total sampling period of 9 days showed a stack gas concentration of  $1.3 \times 10^{-8}$   $\mu$ c/liter. The maximum concentration in air at the ground could not have exceeded  $10^{-10}$   $\mu$ c/liter, with the average concentration no more than  $10^{-11}$   $\mu$ c/liter. Upon re-examination, one of the samples run for 7 days in April 1947 showed six particles of low activity. The total activity of the sample, including distributed activity was only  $8 \times 10^{-3}$   $\mu$ c. It follows that the corrosion debris problem had not started, or was just starting at this date. Most probable is that these particles are dust which collected a little activity in transit through the system.

3. Air sample in Richland, March 1945

Samples of air were taken in Richland as early as March 1945. The primary objective was to evaluate the then high concentration of  $I^{131}$ , but the presence of trace amounts of long lived fission products was noted. For reasons never discovered, one of these samples was at least ten times hotter than the average. This one was preserved, and it has been counted over a 860 day period. Following the decay of the  $I^{131}$  activity, the

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Sr<sup>89</sup> ( $T_{1/2}$  = 55 days) activity was prominent, and later the controlling member was the Ce<sup>144</sup> ( $T_{1/2}$  = 275 days) activity. The effective half life has been approximately 300 days for the past 500 days, representing Ce<sup>144</sup> plus small amounts of longer lived activities. The initial concentration of Ce<sup>144</sup> plus longer lived activities in air was  $2 \times 10^{-9}$  mc/liter, over a five day period. This is entirely incompatible with the quoted stack concentration of  $10^{-7}$  mc/liter, because an average dilution of  $< 100$  at Richland is impossible in view of P. R. Church's meteorological data. Either the fission product contamination did not dilute in the same manner as the smoke used for tests, or more probably the stack sampling was low by a factor of at least ten. The ratio of concentration at Richland and at the stacks will have to be more thoroughly investigated to check:

- (1) Accuracy of sampling.
- (2) Possibility of steady accumulation of activity in stagnant air conditions.

#### 4. Long lived fission products on vegetation

In the early days of operation, with short cooling time justified by the war emergency, I<sup>131</sup> was the critical environmental hazard, and it received primary attention. Nevertheless, trace amounts of longer lived activities were detected on desert vegetation at an early date. After one year of operation, this amounted to approximately 0.05% of the total activity, and this rose to 10% after about 2 years. In the meantime, the I<sup>131</sup> had been reduced by a factor of about 10, by additional cooling time, so that the long lived activity approximately doubled. There has been little further increase in this activity, although it is now comparable with the I<sup>131</sup> activity due to a further increase in cooling time equivalent to I<sup>131</sup> reduction by a factor of 15. The meagre data on long lived activity deposition is consistent with computed rise of a 300 day half-life emitter.

| <u>Operation Time (years)</u> | <u>Relative total Active Deposition</u> |
|-------------------------------|---|
| 1                             | 100                                     |
| 2                             | 145                                     |
| 3                             | 162                                     |
| 4                             | 170                                     |

It is assumed that production rate and process operation have remained constant. Certain modifications have actually tended to reduce the output of contaminated droplets, as is substantiated by the reduced concentration in stack gas shown between March 1945 and April 1947 data.

The vegetation data show that the total activity from emitted droplets has essentially reached saturation already, so that an upper limit to the environmental hazard has been established. This will persist until the weak Sr<sup>90</sup> (25 years) and Ce<sup>137</sup> (30 years) assume control of the effective half life. It will be inexcusable if the emission of such activities is not eliminated shortly.

If the environs of the plant were contaminated with a pattern of discrete particles of long lived activity, the analysis of many one-gram samples of vegetation would show wide variations. With the limited material at hand, this is not the case. In samples, taken at one mile intervals and about 2-5 miles from the stacks, the activity ranged from 0.007 to 0.08 mc/kg with only one sample

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significantly below  $0.04 \mu\text{c}/\text{kg}$ . As the deposition is a surface effect there is an expected range, depending on the use of thin leaves or thick stalks. Within the Plant Areas, 27 samples showed a range from  $0.01 \mu\text{c}/\text{kg}$  to  $0.4 \mu\text{c}/\text{kg}$ , but this spread is no greater than that found in the overall distribution pattern. It is concluded that the main picture of fission product deposition approaches a continuous distribution rather than a particulate one.

To summarize the inferences from old data, the emission of active droplets has been known from the start of operations. It has been casually studied by air sampling and by analyses of vegetation. The hazard has been known to be insignificant, and increasing at an insignificant rate, calculated on the basis of uniform dispersal. When reviewed in terms of possible concentration in discrete foci, it is clear that there is a vanishingly small probability of inhalation-induced injury to any individual outside the reservation, and no evidence that the highest conceivable concentration on a particle capable of travelling many miles would be damaging if inhaled.

#### Summary of Sections 7, 8 and 9

The exploratory chemical, physical and radiochemical studies of the active particles are outlined. These contributed to localization of the particle source. An interesting application of isotopic ratio measurement to the determination of 'age' is included for its potential use in other problems.

Studies on a much more extensive basis would be needed for a thorough analysis of the long-range problem.

The poor prospects for successful clinical studies on exposed personnel are demonstrated. Only a large-scale investigation of fecal activity is promising. Detailed analyses of available autopsy material is strongly indicated.

Limited past data is used to show approximately when the iron-bearing particles developed, and to put some limit on the magnitude of the effect from droplets.

#### 10. Medico-legal aspects

It is the intention of the Company to acquaint all employees with the full facts of the reported problem, when they have been adequately ascertained and analysed. It may be determined that damage to some individuals could have occurred prior to the application of corrective measures. In this event it would be a matter of some difficulty to prove whether a specific claim was founded on fact. The hazard question can surely be confined to the results of inhalation. If the claimed injury is death, a positive answer could be given in some cases.

- (1) If the proposed detailed lung and liver study shows the presence of radioactive particles containing active isotopes of cerium, yttrium, strontium or cesium, the claim would be indisputable, if terminal illness symptoms were suggestive of lung damage.
- (2) Absence of lung injury and absence of radioactivity is presumptive evidence of an ill-founded claim.
- (3) But if the death resulted from lung injury, without demonstrable presence of activity there would be little moral obligation to accept a claim.

This contingency leaves the organization open to many claims, due to the high normal incidence of this cause of death in men. Expert advice is

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needed as to whether it is feasible for a particle to stay in the lung long enough to do damage and then be removed before the case proceeds to autopsy.

If impairment of health is claimed, there appears to be a poor chance of obtaining definitive evidence. Perhaps only prolonged analysis of feces is applicable. Claims not supported by clinical symptoms of lung damage might be eliminated. It is assumed that the employees will act in good faith in these matters, an expectation based on the high confidence of men now required to work in potentially hazardous areas. More unreasonable claims are to be anticipated from relatives of diseased employees, or from the general public in the environs. These can arise from causes ranging from legitimate concern over problems overemphasized in the press to outright fraud. The theoretical possibility of injury developing 10 to 15 years from now poses a serious problem.

It is imperative that the best possible tolerance experiments should be conducted on particulate contamination exposure. Whether these should be done in whole or in part at Hanford should be decided by outside consultants.

#### Consultative Assistance

The following persons or organizations have been approached or will be approached for additional guidance:

- Dr. R. E. Zirkle, Radiobiological Consultant - biological studies
- Dr. S. T. Cantrill, Medical Consultant - clinical effects
- Dr. P. E. Lund, Pathology Consultant - (in process) - tissue damage
- Dr. I. Langmuir and V.J. Schaefer, - fine particle studies
- Dr. Shields Warren - radiation damage
- Dr. G. Failla - health physics
- Dr. P. E. Church - Meteorology Consultant - transport of particles
- Weather Bureau - transport of particles
- Chemical Warfare Service - particles in lung
- Dr. F. McLean - dispersal of particles
- Kellogg Corporation - stack decontamination
- Air Reduction Company - stack decontamination
- Washington State College - fixation of soil

Readers of this document are invited to advise us of other appropriate reference sources.

#### Summary of Summaries

Quantities of the order of ten million to 100 million radioactive particles per month have been emitted from the stacks over the past few months. These particles have an effective radioactive half-life of about 300 days, with trace amounts of longer lived activities. High activity in the range 0.1 - 3  $\mu$ c was probably confined to large carrier particles of corrosion debris from iron ductwork in the Separations Plant ventilation air system. Further emission of these was eliminated by replacement of the ductwork.

Residual particles may be smaller similar particles left in the system, solid particles from other exposed surfaces in the cells, or the fine specks from the mist or droplets escaping from process vessels.

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In brief, long-lived activity has been disseminated as presumed mist or droplets since the start of operations. Expected uniform dispersal of such activity was free from hazard. The unplanned provision of heavy carrier particles brought the activity to ground much closer to the stacks. Removal of the carriers will restore the status quo. In the long range view this wide dispersal of activity should be eliminated to assure unequivocal protection of the public against remotely possible environmental hazard. Possible radiation damage from these particles is poorly understood.

Significant damage probably only follows inhalation. Particles of progressively smaller activity can be found by more diligent search; at what level of activity these cease to be dangerous is vitally needed information. There is no obvious method of detecting particles in the lung. Serious medico-legal problems are thereby introduced.

Corrective steps have been applied to remove large particles from the stack air, and further measures are underway to eliminate all other particles. These steps consist of immediate temporary filtration etc., together with a formal long-range study. Personnel protection has been afforded by respirators.

A residue of a few million particles on the ground in the environs of the Separations Plants will require either removal or fixation to prevent inhalation hazard by re-entry of particles into the atmosphere. Means of effecting this appears costly and should be held to the minimum area consistent with safety. It follows that the transport of particles and possible future concentration in areas currently weakly contaminated has to be known.

Exploratory studies of chemical, physical and radiochemical properties are reported.

Some approaches to calculations of permissible exposure are given in the appendix.

#### Function and nature of this report

The report was accumulated from data developed by several departments, and many subdivisions of these departments. Although the main assignments have been indicated no attempt has been made to credit, in detail, the elements of this fine example of interdepartmental collaboration.

Also, in view of the urgency to present a broad picture of the problem to interested parties, time has not been spent to edit the report. Diffuseness, repetition and possible illogical order of topics should be accepted in the interest of speed. This is not a final report. In fact, it is closed with a paragraph on findings developed since the report was started.

#### Late Results (to 3/26/48)

1. Temporary filters were installed in the B Plant Canyon Building cells on 3/22/48. Subsequent Technical Department samples ran as follows:

| Date       | Sample | Duration<br>Hours | Flow<br>CFM | Activity<br>on filter<br>mrep/hr at 2" | Remarks<br>Visible Particles | N. I.<br>Detectable Particles |
|------------|--------|-------------------|-------------|--|------------------------------|-------------------------------|
| 3/22<br>48 | F1-12  | 4                 | 10          | 10                                     | 0                            | 308, D                        |
|            | F2-14  | 4                 | 10          | 25                                     | 0                            | 368, D                        |
|            | F4-16  | 4                 | 10          | 25                                     | 0                            | X. 508, D                     |

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| Date       | Sample | Duration<br>Hours | Flow<br>CFM | Activity<br>on filter<br>mrep/hr at 2" | Remarks<br>Visible Particles | H. I.<br>Detectable Particles |
|------------|--------|-------------------|-------------|--|------------------------------|-------------------------------|
| 3/23<br>48 | F1-13  | 18                | 10          | 500                                    | 0                            | 90S, D, 9R                    |
|            | F2-15  | 18                | 10          | 500                                    | 30-50                        | 15L 169S DD. 10R              |
|            | F4-17  | 18                | 10          | 500                                    | 0                            | 75S, D, 17R                   |
| 3/24<br>48 | F1-14  | 24                | 10          | 500                                    | 24                           | 20L, 100S, DD                 |
|            | F2-16  | 24                | 10          | 400                                    | 6                            | 16L, 350S, DD                 |
|            | F4-18  | 24                | 10          | 1100                                   | 0                            | 1L, 39S                       |
| 3/25<br>48 | F1-15  | 24                | 10          | 30                                     | 12                           | 80S, D                        |
|            | F2-17  | 24                | 10          | 30                                     | 24                           | 10L, 200S, DD                 |
|            | F4-19  | 24                | 10          | 750                                    | 0                            | 1L, 25R, D                    |
| 3/26<br>48 | F1-16  | 24                | 10          | 900                                    | 2L, 18                       | 4L, 75S, DD                   |
|            | F2-18  | 24                | 10          | 750                                    | Many S                       | 71S                           |
|            | F4-20  | 24                | 10          | 1650                                   | 0                            | 14S                           |

L = large particle (>0.5 mm) S = small particle

D = Dust - DD- = much dust - R = particle detected by radiograph

The particles were obtained by tapping the filters, the residual being measured by radioautography.

There has been a general reduction of activity from about 5000 mrep/hr per day to approximately 500 mrep/hr. Clearly, however, the problem has not been contained by the addition of these filters. Presumably there is a legacy of particles in the ventilation system, that will tend to be emitted sporadically.

2. The filters of March 1945 have been radioautographed again over a long period. One minor spot was found. Whether this was a minute speck of dust or an early corrosion particle is unknown. That it is not a dried droplet snick is inferred from the fact that the rest of the activity should have been deposited in the same form. The particles on the filters of April 1947 contain iron and presumably came from the ductwork.

3. The surveyed area in the 200 East Area has been radically extended to cover a rectangular section 7000' by 5000', comprising the S.E. quadrant of this Area, and containing the B Plant stack at its N.W. corner. The surface concentration between the previously listed Areas F and E was significantly lower than in either one of these. East of Area E, the concentration fell very rapidly to 0.06 psf. It is now suspected that particles have drifted from relatively smooth terrain between Area F and Area E on to the densely covered strip E. In all, ten million particles are accounted for in the surveyed region. There is reason to feel now that most of the problem will be contained in the area,

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especially as particle activity appears to vary more or less inversely as the distance from the stack.

## Average Activity of particles as a Function of Distance from the B Plant Stack

| <u>Distance (d)</u><br><u>feet</u> | <u>Activity A</u><br><u>μpc</u> | <u>Axd.</u><br><u>μpc - feet</u> |
|------------------------------------|---------------------------------|----------------------------------|
| 1500                               | 9                               | 13,500                           |
| 2000                               | 7                               | 14,000                           |
| 3000                               | 4                               | 12,000                           |
| 6000                               | 2                               | 12,000                           |

### Appendix

#### Calculations of permissible exposure

It is not claimed that one can calculate the active particle hazard realistically. Standard methods are adapted here to give order of magnitude of the hazard.

#### Fission product mixture, effective energy and sphere of action

Take a simplified picture in which one microcurie contains

$$0.3 \mu\text{c Ce}^{144}, 0.3 \mu\text{c Pr}^{144}, 0.2 \mu\text{c Sr}^{90}, 0.2 \mu\text{c Y}^{90}$$

This will make the radiation hazard worse than it is believed to be. The energetic beta radiations are  $\text{Pr}^{144}$  3.07 MeV and  $\text{Y}^{90}$  2.16 MeV - Take an average of 2.6 MeV as  $E_{\text{max}}$

The energy from a single particle will essentially be dissipated in a sphere of radius  $d$  defined by  $d = \frac{\text{range for } E_{\text{max}}}{2}$

$$d = \text{range in tissue for 1.3 MeV} = \text{approximately } 0.5 \text{ cm}$$

$$\text{Mass of sphere} = \frac{4}{3} \pi (0.5)^3 \text{ gm} = \text{approximately } 0.5 \text{ gm}$$

Gamma radiation may be neglected, and the effective energy of the beta radiation is

$$E = \frac{1}{3} [0.3(0.35 + 3.07) + 0.2(0.65 + 2.16)] \text{ MeV} = 0.53 \text{ MeV}$$

The most pessimistic sphere of action has a radius = range of 0.53 MeV beta radiation in tissue = 0.18 cm:

$$\text{Most pessimistic mass} = 0.024 \text{ gm}$$

Case 1. A single particle of activity  $2 \mu\text{c}$  remains in one position in the lung. By Cohn's Formula, Dose rate DR =  $62 \frac{EQ}{W}$  rep/day [see MDDC 783]

Where E = average energy of radiation in MeV

Q =  $\mu\text{c}$  deposited

W = grams of tissue in the sphere of action

$$\text{D.R.} = \frac{62 \times 0.53 \times 2}{0.5} \text{ rep per day.}$$

$$= 130 \text{ rep per day}$$

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Such a particle could hardly stay in the lung (even if it could get there in the first place) for more than one month.

The total dose would be about 4000 rep. A comparable dose in a comparable sphere of action is given by the gamma radiation from a one millicurie gold seed.

For the most pessimistic sphere of action,

$$D.R. = \frac{62 \times 0.53 \times 2 \text{ rep per day}}{0.024} = 2750 \text{ rep per day}$$

It was on some such basis that the writer predicted a skin reaction from contact with the particles. The absence of such a reaction tends to support the calculation for the larger sphere of action.

Case 2. Activity of a particle that is unequivocally safe

Such a particle will give 0.1 rep per day or less in the sphere of action.

$$\text{Permissible activity} = \frac{2000 \text{ } \mu\text{mc}}{1300} = 1.5 \text{ } \mu\text{mc}$$

Or for the most pessimistic sphere of action,

$$\text{Permissible activity} = \frac{2000 \text{ } \mu\text{mc}}{27500} = 0.07 \text{ } \mu\text{mc}$$

Case 3. Uniform lung deposition from an aerosol

It is customary to assume that 25% of the available particles are initially retained in the lung, and that the biological elimination rate has a half-life time of 2 months

The radiation half-life is effectively 300 days

$$D.R. = \frac{62 E f. Q_a \text{ rep per day}}{\lambda W}$$

where  $Q_a$  = total activity ( $\mu\text{mc}$ ) breathed in per day

$f$  = fraction retained = 0.25

$\lambda$  =  $\lambda$  radiation +  $\lambda$  biological = (0.0023 + 0.0115)

= 0.014 day<sup>-1</sup>

$W$  = total mass of lungs = 1000 gm.

For D.R. = 0.1 rep per day,

$$Q_a = \frac{0.1 \times 0.014 \times 1000 \text{ } \mu\text{mc per day}}{62 \times 0.53 \times 0.25} = 0.17 \text{ } \mu\text{mc per day}$$

Plant employees breathe 1 cubic meter of air per hour for ten hours daily exposure. In the absence of other exposure, permissible concentration of the aerosol is:

$$\frac{0.17 \text{ } \mu\text{mc/liter}}{10^4}$$

$$= 1.7 \times 10^{-5} \text{ } \mu\text{mc/liter}$$

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In public areas, where residence may be for 24 hours per day, seven days per week

$$\begin{aligned} \text{Permissible aerosol concentration} &= \frac{1.7 \times 10^{-5} \times 10 \times 5}{24 \times 7} \text{ mc/liter} \\ &= 3 \text{ mc/liter} \end{aligned}$$

Case 4. Intermediate case of exposure to numerous small particles, not sufficiently dispersed to be treated as uniform in the lung

This is the case of immediate interest, and naturally is the one that can be calculated with least certainty.

Individual particles of activity 1.5 mpc can be presumed safe. Assume n particles breathed in per day

$$\text{Total number in lung} = \frac{f.n.}{\lambda_{\text{biol}}} = \frac{0.83n}{0.0115} = 22n$$

There will be 22n spheres of action each of mass 0.5 gm in the total lung mass of 1000 gm. Provided 11n is small compared with 1000, there will be insignificant overlap of the spheres of action.

n = 5 gives a low probability of overlap

Therefore, it is safe to inhale 5 particles per day of activity 1.5 mpc or less.

From this point, calculations of the hazard of the real field conditions become more and more speculative.

There is a consensus of opinion that small isolated areas of tissue are distinctly more radioreistant than are large areas, perhaps due to the ease with which the body can marshal reparative agents to the irradiated site. Suppose this represents a factor of 5 between the whole body and a sphere of mass 0.5 gm. Then it would be safe to inhale 5 particles per day of activity 7.5 mpc or less. It is not absurd to suppose that one particle per day of activity 10 mpc could be tolerated.

Distribution of particles emitted from the stacks

This must be calculated or measured by competent experts. Provisionally, the idealized equation of Bosanquet and Pearson is applied to the case.

$$N_0 = \frac{Nt \cdot 10^{-6} (1.78 H/k_1)^{v_{tn}/k_1 v_x} e^{-H/k_1 x}}{\sqrt{2\pi} k_1 k_1 v_x x (2 + v_{tn}/k_1 v_x)}$$

- where  $N_0$  = number of particles per cubic meter of air at ground level  
 $Nt$  = number of particles emitted per day = approximately  $10^6$   
 $H$  = stack height = 60 meters  
 $k_1$  = 0.08,  $k_1 = 0.05$   
 $v_x$  = velocity of wind = 2 meters/sec., say  
 $x$  = distance from stack in direction of wind  
 $v_{tn}$  = terminal velocity of particles

Approximate terminal velocities for irregular (quartz) grains are:

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| <u>Diameter (microns)</u> | <u>V<sub>m</sub> (meter/sec)</u> |
|---------------------------|----------------------------------|
| 2.5                       | 0.01                             |
| 11                        | 0.2                              |
| 25                        | 1                                |

The following values arise from these assumptions:

| <u>Approximate Particle Size Microns</u> | <u>Terminal Velocity meter/sec</u> | <u>Peak Concentration Particles/c. meter</u> | <u>Distance of Peak from stack feet</u> | <u>Particles Inhaled per week</u> |
|--|------------------------------------|--|---|-----------------------------------|
| < 1                                      | 0                                  | $2 \times 10^{-3}$                           | 2200                                    | 10-3                              |
| 2.5                                      | 0.01                               | $2.4 \times 10^{-3}$                         | 2000                                    | $1.2 \times 10^{-3}$              |
| 11                                       | 0.2                                | $5 \times 10^{-4}$                           | 1000                                    | $2.5 \times 10^{-2}$              |
| 25                                       | 1                                  | $(5 \times 10^5)^*$                          | 320                                     | ?                                 |
| ~50                                      | 2                                  | large  | 160                                     | too large to be inhaled?          |
| ~100                                     | 4                                  | large  | 80                                      | too large to be inhaled           |

This indicates that there is no hazard for ultra fine particles. Even at much lower wind velocities, the concentration would not rise to hazardous levels. Qualitatively, it is also clear how the production of carrier particles in the system brought down showers of particles in high concentration and close to the stack, thus leading to detection. Particles in the main range of interest (1-3 $\mu$ ) behave essentially as particles with zero terminal velocities.

### Wind transport of particles

This must be solved by competent experts; in the interim, a crude order of magnitude calculation may have some value.

Dalla Valle's formula modifies to\*\*

$$N_0 = \frac{K^2 (C_1 - C_2)(V_2 - V_1)}{\ln^2(Z_2/Z_1)}$$

where  $N_0$  = number of particles picked up per sq. ft. per hour  
 $K$  = Von Karman's turbulence constant = 0.4  
 $C_1, C_2$  = number of particles per cubic foot at two levels  
 $Z_1, Z_2$  (meters)  
 $V_1, V_2$  = wind velocity (ft/hr) at these levels

For a severe dust storm, typical data are:

$C_1 = 2 \times 10^9$  at  $Z_1 = 1$   
 $C_2 = 6 \times 10^8$  at  $Z_2 = 10$   
 $V_1 = 20 \times 3280, V_2 = 17.4 \times 3280$

Hence,  $N_0 = 5.8 \times 10^{11}$  pf per hour

Now arbitrarily assume that this storm blows for 10 hours

Total pick up =  $5.8 \times 10^{12}$  pf

\* Formula breaks down for large values of  $V_m/K_1 V_x$

\*\* J. M. Dalla Valle "Micromeritics" - Pittman Publishing Corp.

# The formula gives a negative answer, but as Dalla Valle's text as a whole appears to contain several confusions of centimeters with meters, cubic cm. with cubic ft. and the like, we ignore this and hope the numerical values are correct here.

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The pick up supports  $2 \times 10^9$  p. per c. ft. at breathing height.  
If there was originally radioactive contamination of 1 paf, the concentration  
in air is now effectively  $2 \times 10^9$  =  $3.5 \times 10^{-4}$  active particles per c. ft.  
 $5.8 \times 10^{12}$

350 c. ft. are breathed in a workday.

Therefore inhalation rate = 0.1 active particles per day  
= 1 per two work weeks

This might be representative of the hazard condition in a severe wind storm.  
Such storms support particles of average diameter approximately 2  $\mu$ .

Suppose, naively, that the the ground is covered with particles of diameter 2  $\mu$  in a uniform close-packed array. There will be  $2 \times 10^{10}$  particles per sq ft on the surface layer, and 30 layers will be picked up per hour. In a more moderate storm, postulate that just the surface layer is picked up in one day. This would give a concentration at breathing level of  $7 \times 10^6$  particles per c. ft., barely recognizable as a dust cloud. The active concentration would be  $3.5 \times 10^{-4}$  particles per c. ft., and the inhalation rate again one particle in two weeks. Provisionally assume that particles of size 2  $\mu$  have activity not in excess of 2 mpc. Then 1 paf on the ground appears to be safe provided that a person stays out of readily visible clouds of dust. Similar arguments must apply to particles picked up by thermal currents. For safety, we shall take 0.1 paf (weak particles only) as a permissible value, until this type of calculation is performed by recognized experts.

H. M. Parker  
H. M. Parker

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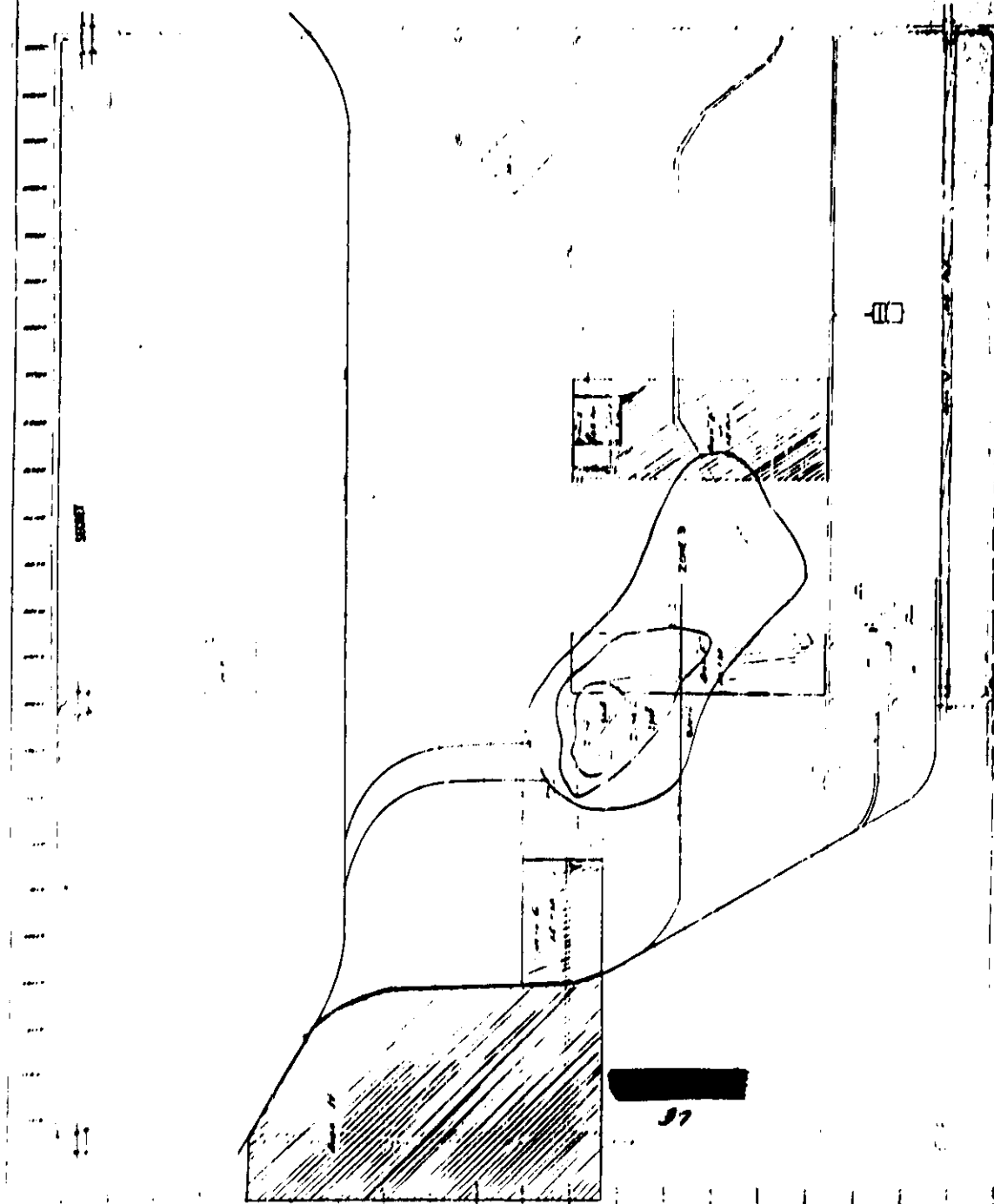
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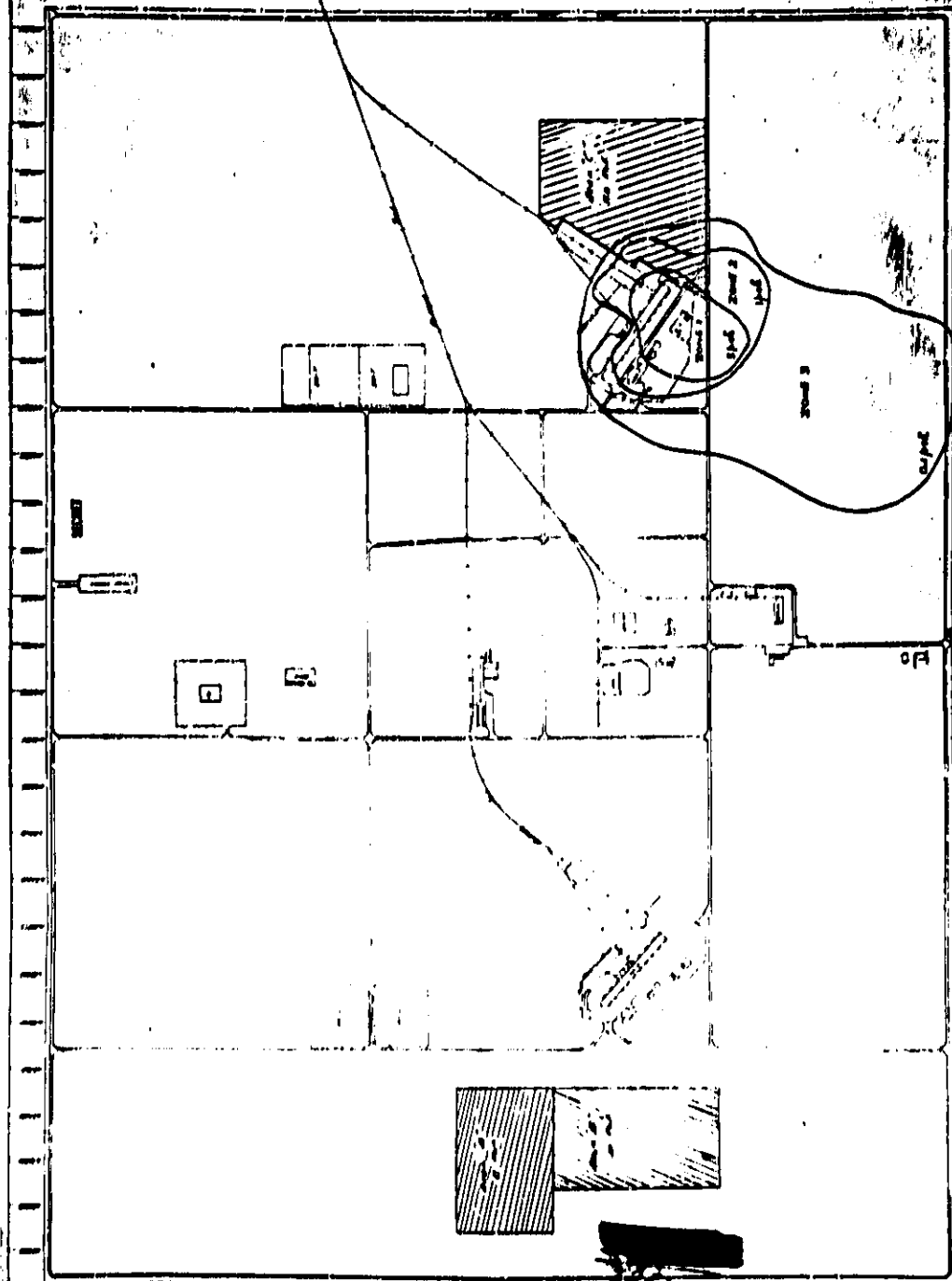
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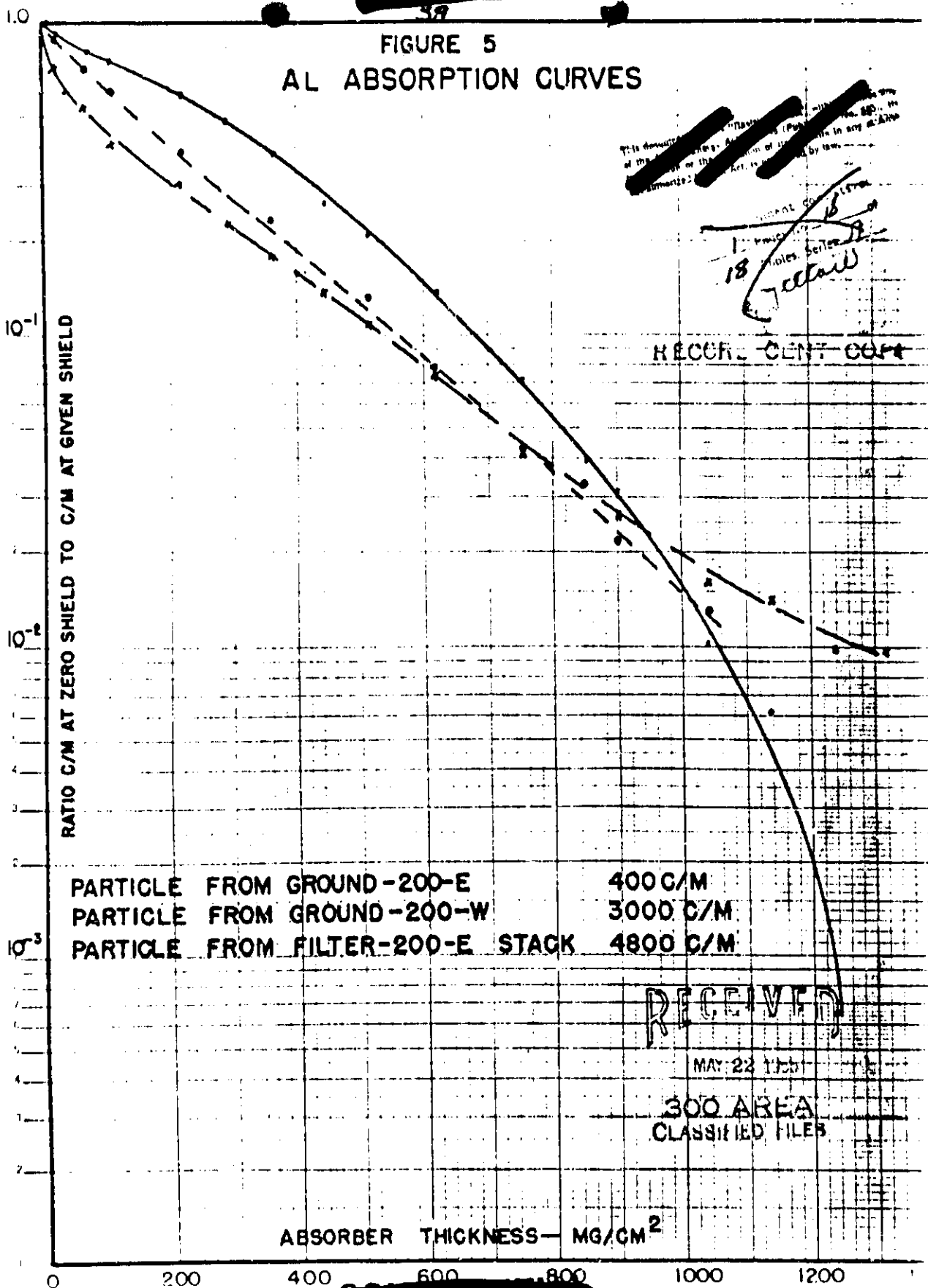
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FIGURE 5  
AL ABSORPTION CURVES



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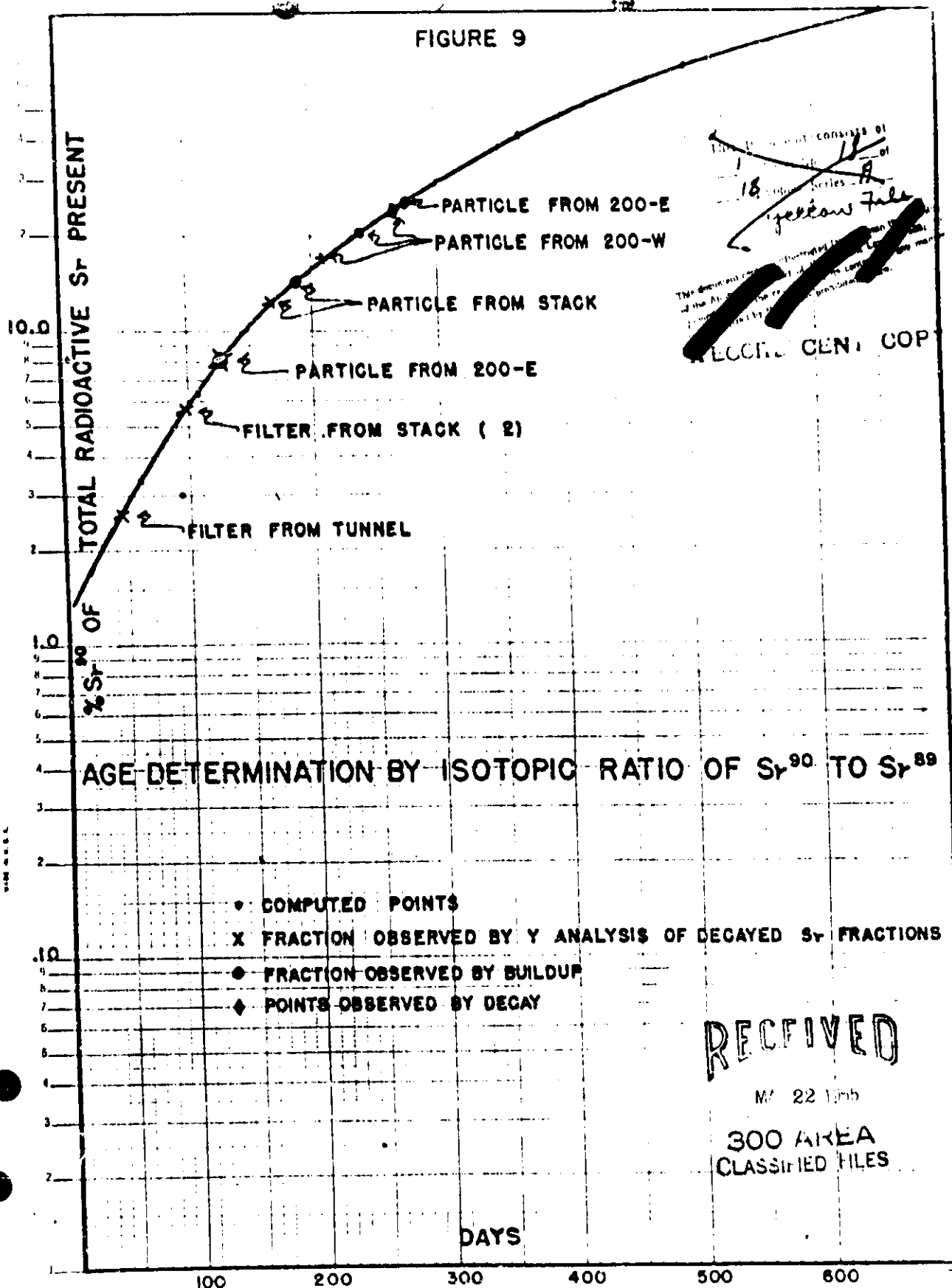
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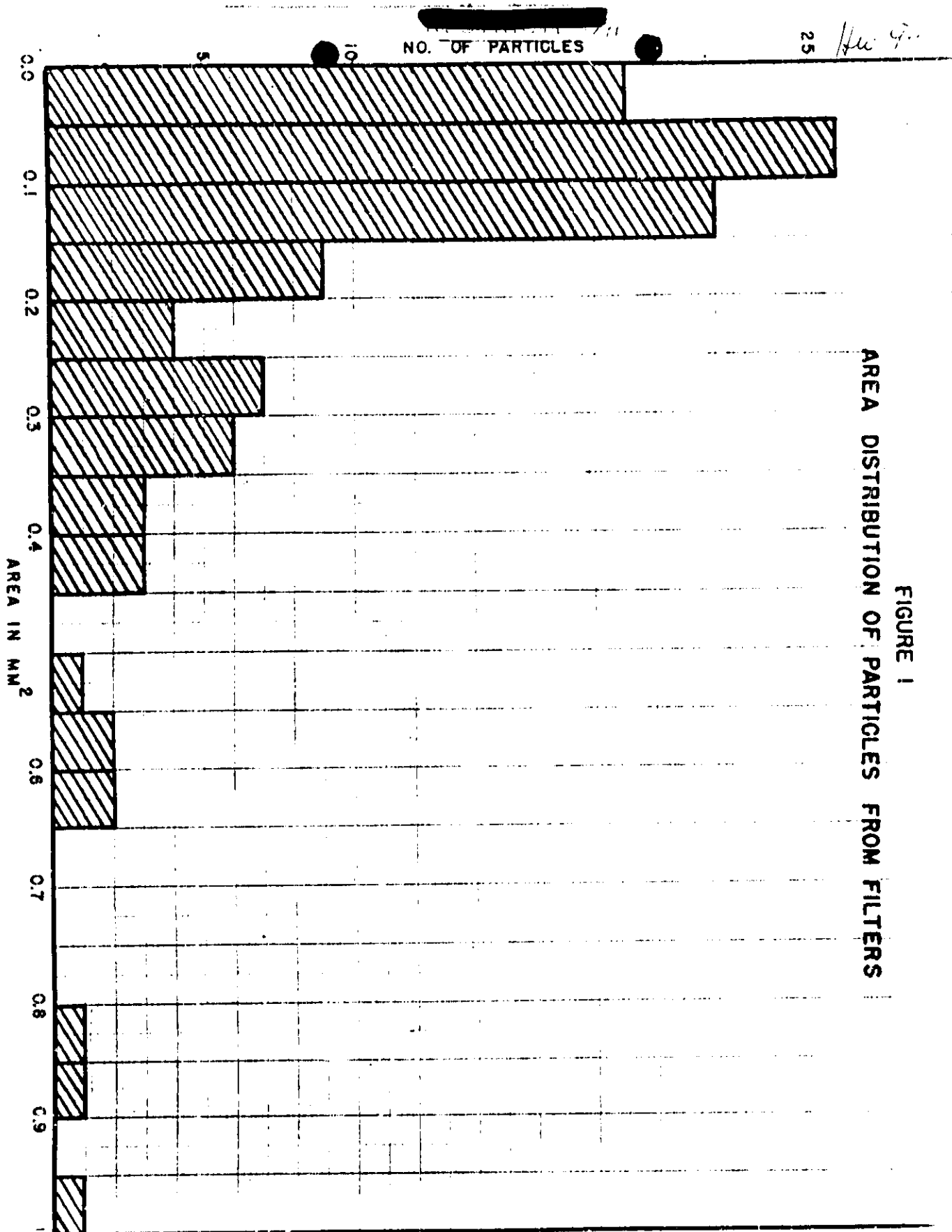
FIGURE 9



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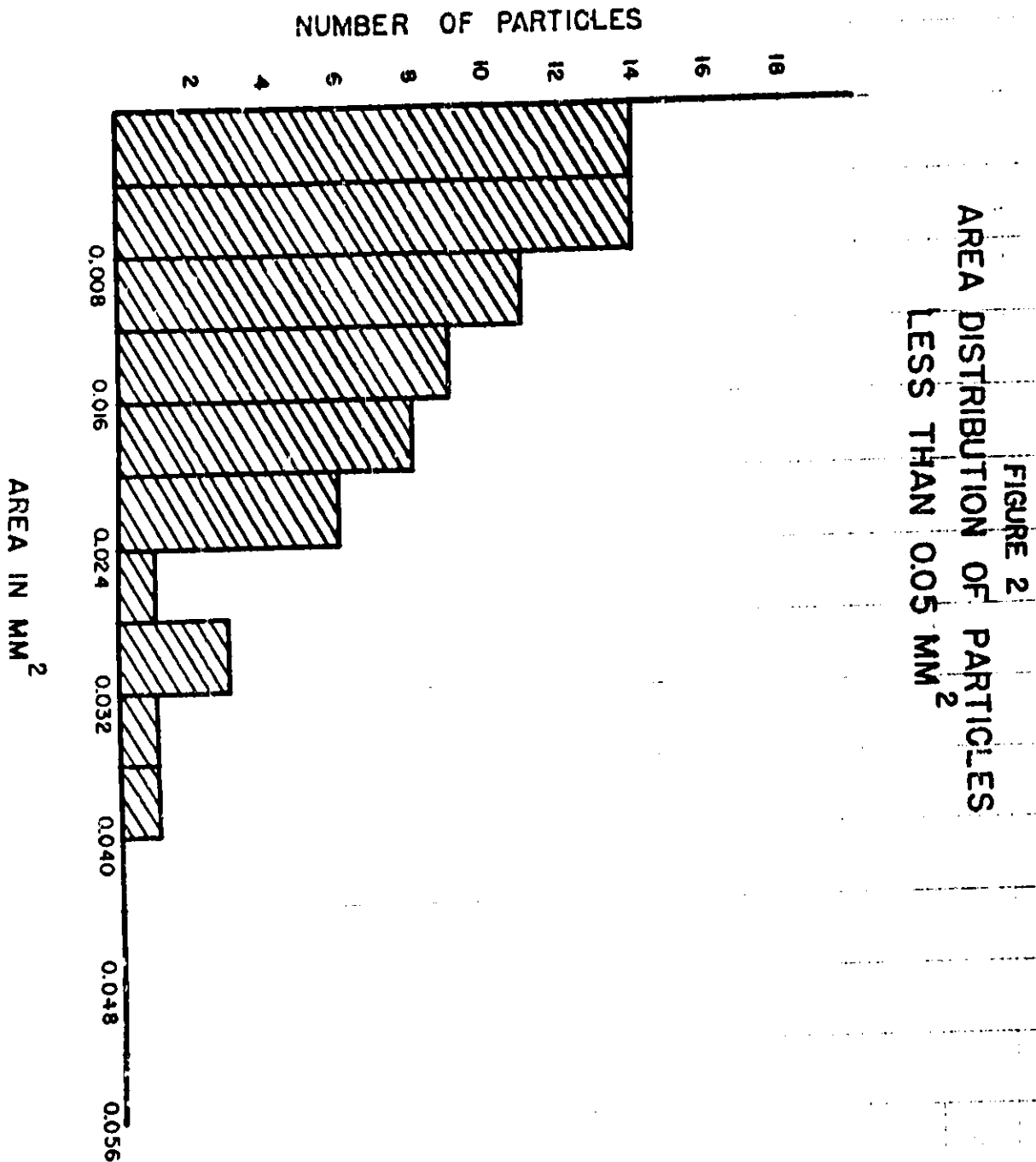


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