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INVESTIGATIONS WITH A DEVICE FOR THE PROMPT DETECTION OP RADIOACTIVE AEROSOLS

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INVESTIGATIONS WITH A DEVICE FOR THE
PROMPT DETECTION OF RADIOACTIVE AEROSOLS

C. T. Rainey

June 8, 1951

Berkeley, California
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Radiation Laboratory, Department of Physics
University of California, Berkeley, California

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ABSTRACT

A device for the prompt detection of radioactive aerosols is described. It consists of two detectors -- one measuring the activity of suspect air, the other measuring air known to contain no activity other than that due to naturally occurring airborne radioisotopes. The difference between the two channels is recorded as a measure of contamination. Alpha emitters were used in the investigation but the equipment may be converted to measure beta. The fact that contamination may be expected to occur in bursts makes possible the detection of levels below the average concentration considered permissible. The equipment lends itself to the investigation of probable sources of contamination in laboratory procedures.
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PROMPT DETECTION OF RADIOACTIVE AEROSOLS

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INTRODUCTION

The presence of short-lived alpha emitters in the air in concentrations
that are many times the maximum permissible level for long-lived alpha emitters
makes the problem of detecting the long-lived emitters difficult. In the past,
the only workable system has been to collect air samples for long periods of
time and then allow for the decay of short-lived emitters. This system has
three serious drawbacks. First, the presence of any contamination is not
known for at least one day. Second, the time of occurrence is not known.
Third, it is not known whether the contamination was a burst of high concen-
tration or a long lasting condition of low concentration. These three factors
make this system ineffectual as a means of preventing further contamination.

At the University of California Radiation Laboratory all active work is
done in gloved boxes, as shown in Figure 1. The box is maintained at slightly
negative pressure. The incoming air is filtered by a PF105 glass filter and
all air leaving the box is filtered by a train of high efficiency filters. In many cases the level of activity handled is above $10^4$ counts per minute
of alpha. If the filter system fails or if a sample is spilled when being
removed from the box a serious hazard will exist. The number of times that
such a level of activity is routinely handled in this manner makes the need
for a prompt air contamination detector urgent (Figure 2).

If we assume that the level of activity due to natural emitters is of
the same magnitude throughout a laboratory, and that a rise in activity due to contamination might be expected to occur in bursts of high concentration following particular operations, the following solution to the detection problem is suggested. Sample the air from two locations, one known to be uncontaminated (incoming ventilation system), the other suspected of being contaminated (the active work area), and count each sample. The difference in count rate will be a measure of the contamination. The obvious limitation is that to have a significant difference, the contamination would have to be of great magnitude. However, if our assumptions are correct, this limitation would be minimized. The experiments described below demonstrate the validity of our assumptions and have led to the development of a means of rapidly detecting air contamination.

EXPERIMENTAL BACKGROUND

1. Two cylindrical air proportional alpha counters were installed in the cave room at the University of California Radiation Laboratory. The walls of the cylinders were heated which caused air to be drawn through each counter. The following observations were made:

A. The activity measured by each counter was of the same order of magnitude in various parts of the room.

B. When one probe was placed immediately in front of the cave wall and the other was placed in the stream of incoming room air, a rise in activity was detected near the cave. Investigation with a smoke gun revealed a small eddy current sweeping over the chemistry area in the cave and back into the room. When this ventilation defect was corrected, the activity detected became the same on each channel.
C. Inherent noise in an air proportional counter raised the background to a point where only extremely high levels were detectable.

D. The sampler did not collect a sample but only counted the air passing through the chamber. Therefore, when a rise was detected, there was no way to recount the sample at a later time. Therefore, it was not possible to discriminate between noise and activity.

Because of the above difficulties, the equipment was discarded in favor of a low background scintillation counter and a moving filter paper.

2. Routine air monitoring procedures had revealed that contamination of the room air had occurred following the sparking of alpha emitters in the spectrographic laboratory. By means of a moving filter paper and a low background scintillation counter the room air was sampled during the operation. The results are shown in Figure 3. Before further work was accomplished, the arc-spark chamber gloved box was replaced by a new assembly. Investigations after the new box was installed revealed no further contamination of the room air.

**EQUIPMENT**

An experimental unit of the two-channel system described above has been constructed. It consisted of a paper drive assembly pulling a strip of Hollingsworth and Vose #H-70 paper across two air streams and then under two #5819 photomultiplier tubes coated with silver-activated zinc sulfide. The photomultipliers drive two scalers as well as an Esterline Angus chart recorder indicating individual pulses. Thus we have an integrated reading as well as an indication of the time occurrence of activity. The air mover
is a Filter Queen vacuum cleaner with a rheostat for controlling the air flow. The paper moves continuously over two 1-1/4 in. x 1-1/4 in. collection areas at a speed of 1/8 in. per minute. The counting area is of the same dimensions and is situated as close as possible to the collection area (approximately 1-1/4 in.). This yields a collection time and a counting time of ten minutes with about a ten minute delay before detection. The paper may be rerun through the counter if a decay plot of the filter paper is desired.

Consideration of the system yields the following observations:

1. If the activity collected by each channel is of the same order of magnitude, the counts detected will vary according to a random distribution curve.

2. If the suspect air contains contamination in addition to natural activity, the difference between the activities will indicate the magnitude of contamination. In order to be significant, this difference must be greater than the variation that might be suspected from random fluctuations.

3. To be significant, any difference in activity must be indicated by the chart recorder for at least twenty minutes which is the time required for the sample to move across the counting area.

4. A burst of activity will be indicated by the activity rising to a maximum and falling to background in twenty minutes.

5. A long lasting uniform contamination will be indicated by a flattened peak on the count rate versus time curve.

6. For bursts, the maximum count rate is a direct indication of the concentration of contamination.

7. If the activity collected is on a single particle, the count rate will be high for 10 minutes with a very sharp rise and fall.
8. The sample may be recounted if a reading is questioned or if a decay curve is desired.

9. The ultimate sensitivity will depend upon the level of activity on each channel due to radon and thoron daughters.

**EXPERIMENTS**

The sequence of the following experiments was designed to provide a semi-quantitative evaluation of the two-channel system. A more extensive investigation will be undertaken following the assembly of improved equipment.

**Procedure** Using the two-channel system, air was sampled from various locations within a glove box enclosure and in the room. Operations in the box were logged and were correlated to the chart recording of activity. The source of radioactive aerosol was contamination of the inside of the box as well as an aerosol generated by sparking microgram quantities of plutonium. The complete operation of the spark-arc gloved box is described in UCRL-1138. During all experiments the air in the laboratory was sampled by routine air monitoring procedures. In no case was contamination of room air detectable.
Experiment 1: The immediate detection of air contamination within a box. (Figure 4)

### Sample 1

<table>
<thead>
<tr>
<th>Location</th>
<th>Box air</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow Rate</td>
<td>3000 cc/minute</td>
</tr>
<tr>
<td>Aerosol</td>
<td>Three Pu samples:</td>
</tr>
<tr>
<td></td>
<td>70, 100, and 120 micrograms plus blank.</td>
</tr>
<tr>
<td>Operations</td>
<td>Introduce samples, evaporate</td>
</tr>
<tr>
<td></td>
<td>on electrodes. Open chamber, spark sample.</td>
</tr>
<tr>
<td>Results</td>
<td>1. Peak due to manipulations and evaporation.</td>
</tr>
<tr>
<td></td>
<td>Maximum of 170 c/m detected 20 minutes after starting operations.</td>
</tr>
<tr>
<td></td>
<td>Activity was high for 25 minutes with uniform rise and fall. Flat-topped for five minutes.</td>
</tr>
<tr>
<td></td>
<td>2. Activity between 1 and 2 counts/minute for 3-1/2 hours between operations.</td>
</tr>
<tr>
<td></td>
<td>3. Peak due to sparking. Maximum of 14 c/m detected. 24 minutes wide.</td>
</tr>
</tbody>
</table>

### Sample 2

<table>
<thead>
<tr>
<th>Room air</th>
<th>3700 cc/minute. Difference due to negative pressure of box.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow Rate</td>
<td>3700 cc/minute. Difference due to negative pressure of box.</td>
</tr>
<tr>
<td>Aerosol</td>
<td>Three Pu samples:</td>
</tr>
<tr>
<td></td>
<td>70, 100, and 120 micrograms plus blank.</td>
</tr>
<tr>
<td>Operations</td>
<td>Introduce samples, evaporate on electrodes. Open chamber, spark sample.</td>
</tr>
<tr>
<td>Results</td>
<td>Activity remained between 2 and 4 c/m throughout period.</td>
</tr>
</tbody>
</table>
**Experiment 2**: Difference in activity in the two units of gloved box installation. Closer identification of peaks. Activity due to chemistry procedures.

<table>
<thead>
<tr>
<th>Sample 1</th>
<th>Sample 2</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Location</strong>:</td>
<td>Spark chamber box</td>
</tr>
<tr>
<td><strong>Flow Rate</strong>:</td>
<td>3700 cc/minute</td>
</tr>
<tr>
<td><strong>Aerosol</strong>:</td>
<td>Four Pu Samples: 90.0, 92.5, 125, 15 micrograms plus blank</td>
</tr>
<tr>
<td><strong>Operations</strong>:</td>
<td>Opening chamber, Sparking, Glove manipulations</td>
</tr>
<tr>
<td><strong>Results</strong>:</td>
<td>1. Peak 18 minutes after starting to work in box. Max. 100 c/m detected. Dropped to background of approximately 2 c/m after 35 minutes. Was working in box about 12 minutes.</td>
</tr>
<tr>
<td></td>
<td>2. Remained at approximately 2 c/m while #2 went through peaks.</td>
</tr>
<tr>
<td></td>
<td>3. Opened chamber, cleaned windows and sparked samples. Activity rose and was too high for pen recorder. Average count rate was 30 c/m for 18 minutes</td>
</tr>
</tbody>
</table>
**Experiment 3:** Difference in activity in the two units of gloved box installations. Sparking operations.

<table>
<thead>
<tr>
<th>Sample 1</th>
<th>Sample 2</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Location:</strong> Spark Chamber Box</td>
<td>Chemistry box</td>
</tr>
<tr>
<td><strong>Flow Rate:</strong> 7000 cc/minute</td>
<td>7000 cc/minute</td>
</tr>
<tr>
<td><strong>Aerosol:</strong> Four samples of Pu: approximately 100 micrograms plus blank</td>
<td>Same</td>
</tr>
<tr>
<td><strong>Operations:</strong> Sparking of samples.</td>
<td>Same</td>
</tr>
<tr>
<td>Flushing chamber for varying lengths of time.</td>
<td>Same</td>
</tr>
<tr>
<td>Glove manipulations</td>
<td>Same</td>
</tr>
<tr>
<td><strong>Results:</strong></td>
<td></td>
</tr>
<tr>
<td>1. Peak due to setting up equipment.</td>
<td>1. Peak due to setting up equipment.</td>
</tr>
<tr>
<td>Approximately 150 c/m max.</td>
<td>Approximately 40 c/m max.</td>
</tr>
<tr>
<td>2. Peaks after opening chamber and sparking.</td>
<td>2. Peaks detected at same time were always consistently lower.</td>
</tr>
<tr>
<td>Flushing chamber after sparking lowered the peak by factor of 30.</td>
<td></td>
</tr>
<tr>
<td>3. No peak detected after sparking without opening chamber.</td>
<td>3. Same</td>
</tr>
<tr>
<td>4. Glove manipulations for one minute gave large peak.</td>
<td>4. Same but lower.</td>
</tr>
</tbody>
</table>
**Experiment 4:** Comparison between moving system and standard air monitoring procedure. Immediate counting, then recounting after decay period.

<table>
<thead>
<tr>
<th></th>
<th>Sample 1</th>
<th>Sample 2</th>
<th>Filter Queen 1</th>
<th>Filter Queen 2</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Location:</strong></td>
<td>Box</td>
<td>Room</td>
<td>Box</td>
<td>Room</td>
</tr>
<tr>
<td><strong>Flow Rate:</strong></td>
<td>3500 cc/min</td>
<td>3500 cc/min</td>
<td>3500 cc/min</td>
<td>4 CFM (Normal air monitoring procedure)</td>
</tr>
<tr>
<td><strong>Aerosol:</strong></td>
<td>Box contamination</td>
<td>Same</td>
<td>Same</td>
<td></td>
</tr>
<tr>
<td><strong>Operation:</strong></td>
<td>Glove manipulations</td>
<td>Same</td>
<td>Same</td>
<td></td>
</tr>
<tr>
<td><strong>Results:</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1. Immediate Count</td>
<td>Peak of 11 c/m, 20 min wide.</td>
<td>No Peak.</td>
<td>Background approx. 1.5 c/m</td>
<td>151 c/m</td>
</tr>
<tr>
<td>2. After Decay</td>
<td>Peak 9 c/m, 20 min wide.</td>
<td>No Peak.</td>
<td>Background approx. 0.25 c/m</td>
<td>73 c/m</td>
</tr>
</tbody>
</table>

See graph of Experiment 4 for greater details.

**NOTE:** The discrepancy between counting yield of each system may be accounted for by considering the lack of a representative sample as well as difficulties in regulating air flow.

**Experiment 5:** To determine the degree of airborne contamination due to the air stream in the box lifting active dust off of contaminated surfaces.

Air in the box was sampled by standard air monitoring procedures and the following counting results obtained:

- **Flow Rate:** 3500 cc/minute
- **Total Sample:** $1.26 \times 10^6$ cc
- **Operation:** no operations within box
- **Time of Run:** 6 hours
- **Filter Paper:** N-70, 4 in. x 9 in.
- **Counter:** Argon filled ion chamber

**Results:**

<table>
<thead>
<tr>
<th></th>
<th>counts/minute above background</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st Count</td>
<td>171 c/m</td>
</tr>
<tr>
<td>16 hr. decay</td>
<td>16 c/m</td>
</tr>
<tr>
<td>48 hr. decay</td>
<td>3 c/m</td>
</tr>
<tr>
<td>64 hr. decay</td>
<td>0 c/m</td>
</tr>
</tbody>
</table>
OBSERVATIONS

In all cases, a rise of activity was readily detectable following operations in the box. This rise was detectable over the background due to radon and thoron.

It was observed that the background in the box was consistently lower than in the room. The box air is prefiltered by PF105 glass wool. Further investigations as to the effect of prefiltering air supplies might be worthwhile.

In all cases, the level of activity in the box dropped rapidly to background indicating that, even in a highly contaminated area, air contamination may be expected to occur in bursts.

It is realized that the air passing through the box will sweep away a cloud of contamination in short order. However, the flow rate (10 CFM through a box volume of 14 cubic feet) is not far different than the rate the air is changed in the vicinity of a standard hood drawing 1000 CFM. The resolving time of the moving paper system was too slow to determine fall out time of a burst of activity.

It should be noted that we did not detect any air contamination due to the air stream lifting activity off the surfaces of the box.

CONCLUSION

Preliminary experiments show that bursts of contamination of a few times background are readily detectable. Though this level is many times the maximum permissible concentrations of long-lived alpha emitters, the following point should be noted. The tolerance level is an average concentration arrived at by sampling the air for long periods of time and averaging the amount collected over the total time. Results indicate that, even in a highly contaminated area, air contamination may be the result of bursts of activity
caused by particular operations. Such bursts are detectable by this system. This puts a preventive aspect to the system and would enable one to take immediate steps to prevent further contamination. Though the work described above is with alpha contaminants, the system is adaptable to beta emitters by means of replacing the zinc sulfide screen with a suitable phosphor.

The two-channel system is not intended to be a replacement for present air monitoring practices. The large volume of air sampled by present equipment makes them indispensible for detection of low concentrations.

**APPENDIX**

Calculation of concentration of contamination:

\[
\text{curies/cc} = \frac{\text{counts/minute}}{\text{flow rate x time x filter yield x geometry x } 2.2 \times 10^{12}}
\]

Filter yield is that activity level detectable from a filtered sample. It includes filter efficiency and absorption of alpha particles. We assume a value of 75 percent. Geometry is about 25 percent.

In the case of the ion chamber used for counting 4 in. x 9 in. papers, the geometry is near 50 percent.

The expected count yield is measured at 3500 cc/minute sample rate.

<table>
<thead>
<tr>
<th>Peak Count Rate</th>
<th>Concentration averaged over 10 minutes</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.135 c/m</td>
<td>(1 \times 10^{-17}) curies/cc</td>
</tr>
<tr>
<td>1.35 c/m</td>
<td>(1 \times 10^{-16}) curies/cc</td>
</tr>
<tr>
<td>13.5 c/m</td>
<td>(1 \times 10^{-15}) curies/cc</td>
</tr>
</tbody>
</table>
ACKNOWLEDGMENTS

The author wishes to thank John G. Conway for his interest in the problem and for providing the use of his spectrographic arc-spark gloved box for the generation of aerosols.

BIBLIOGRAPHY

1. Carmichael and Tunnicliffe, CRTec-326, "Measurement of Alpha Active Dust in the Atmosphere".


FIG. 1  AIR CONTAMINATION DETECTOR

MU 2168
COLLECTION AREA
1/4" x 1/4"

SCINTILLATION COUNTER
ZnS SCREEN
5819 TUBE
COUNTING AREA
1/4" x 1/4"

ALPHA COUNTER NO. 1
ALPHA COUNTER NO. 2

AIR SAMPLE
AIR SAMPLE

MANOMETER COLLEETION
FILTER PAPER
SUPPLY

PLANUM

FILTER PAPER

SPEED 1/8"/MIN.

MOTOR DRIVEN ROLLER

SPRING LOADED ROLLER

ADJUST FOR EQUAL FLOW IN EACH STREAM MEASURED BY EMIL GREINER FLOWMETER

FIG. 2 ALPHA AIR CONTAMINATION DETECTOR

MU 2189
179231
FIG. 3

PEAK 18 C/M FOR 7 1/4 MIN.

FLOW RATE 1.44 CFM

VAC ON

SPARKING

VAC OFF

TIME MINUTES

COUNTS/MIN. ON FILTER PAPER

MU 2170
Gloves are located along the front of each box.

Note: Events are plotted to coincide with peak of activity, i.e., they are moved over 20 min.
PREPARING TO WORK IN BOX. CLEAN PIPETTES, ETC.

90 µg Pu on electrode, air sample directly over electrode.

92.5 µg Pu on electrodes

Cleaning pipette under funnel

Evap. C sample: 125 µg

Removed funnel. Rest of chart shows two samples from L.H. box. Evap. 15 µg Pu. Might have spilled drop on box.

Open arg chamber, clean windows.
GOING INTO BOX. HOOK FUNNEL INTO R.H. BOX. BRING ELECTRODES INTO L.H. BOX. OPEN DOOR.

OPEN CHAMBER. PUT IN NEW ELECTRODES. SPARK. SPARK BLANK. FLUSH FOR 30 SEC. TRANSFERRED ELECTRODES FROM L.H. TO R.H. BOX.

OPEN CHAMBER. SET UP FOR SPARKING 100 µg Pu.

SPARK 100 µg Pu. FLUSH FOR 15 SEC. CHANGE ELECTRODES.

SPARK 100 µg Pu. FLUSH 30 SEC. OPEN. CHANGE ELECTRODES.

SPARKING 100 µg Pu. OPEN CHAMBER WITHOUT FLUSHING.

SPARK 100 µg Pu. NOT OPENING CHAMBER BEFORE OR AFTER SPARKING. FLUSH FOR 15 SEC.

SAMPLE 1

SAMPLE 2

WORKED HANDS IN GLOVES OF L.H. BOX.
FILTER QUEEN SAMPLE
4" BY 9" H.V. TO PAPER
COUNTED ON ARGON FILLED ION CHAMBER
ROOM BOX

SAMPLE 1
1st COUNT 198 c/m
16 HR DECAY 17
72 HR. DECAY 2

SAMPLE 2
AVER. REST OF RUN.
2 62

PAPER FROM SAMPLE 1 WHEN COUNTED IN
ION CHAMBER YIELDED 22 c/m

LEVEL DUE TO RADON AND THORON

COUNT RATE AFTER 24 HR. DECAY

FIG. 7 (EXP. 4)