3

PNL-3858

UC-11

Evaluation of Radon Progeny from Mount St. Helens Eruptions

E. A. Lepel
K. B. Olsen
V. W. Thomas
F. N. Eichner

September 1982

Prepared for
the U.S. Environmental Protection Agency
under a Related Services Agreement with the
U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute

Battelle

PNL-3858

DISCLAIMER

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830

Printed in the United States of America
Available from
National Technical Information Service
United States Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151

NTIS Price Codes
Microfiche A01

Printed Copy

Pages	Price Codes
001-025	A02
026-050	A03
051-075	A04
076-100	A05
101-125	A06
126-150	A07
151-175	A08
176-200	A09
201-225	A010
226-250	A011
251-275	A012
276-300	A013

PNL-3858
UC-11

EVALUATION OF RADON PROGENY FROM MOUNT ST. HELENS ERUPTIONS

E.A. Lepel
K.B. Olsen
V.W. Thomas
F.N. Eichner

September 1982

Prepared for
the U.S. Environmental Protection Agency
under a Related Services Agreement
with the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

TABLE OF CONTENTS

ACKNOWLEDGEMENTS

The authors would like to thank the following people for their permission and assistance in setting up collection units at their various localities:

Dr. James Jardee and Norma Kramer, Adams County Health District, Ritzville, WA

Lee Mellish and Ron Edgar, Air Pollution Control, County Department of Public Health, Spokane, WA

Marshall Scott, Nuclear Radiation Center, Washington State University, Pullman, WA

Robert Crossland and Tom Silva, Yakima County Clean Air Authority, Yakima, WA

Glenn Kay, U.S. Forest Service, Packwood Ranger Station, Packwood, WA

Dr. Larry Wogman, Adna, WA

Fred Prahl and Ed Nurlang, Oceanography Department, University of Washington, Seattle, WA

Lloyd Clark, Department of Natural Resources, Castle Rock, WA

Bert Reberts, Cougar, WA

Bill Locks and Monte Wilcox, U.S. Forest Service, Mt. Adams Ranger Station, Trout Lake, WA

Dr. John Cooper, Oregon Graduate Center, Beaverton, OR

Our thanks to Greg Henrie, Physical Sciences Department, PNL, for the innumerable trips around Mount St. Helens to pick up and return the samples to Richland, WA. The authors also wish to thank Patrick Heasler, Energy Systems Department, PNL, for his guidance on the statistical interpretation of the data.

LIST OF TABLES

LIST OF FIGURES

FINAL REPORT

EVALUATION OF RADON PROGENY FROM MOUNT ST. HELENS ERUPTIONS

E. A. Lepel, K. B. Olsen, V. W. Thomas
Radiological & Inorganic Chemistry Section
Radiological Sciences Department

and

F. N. Eichner
Dosimetry Technology Section
Radiological Sciences Department

Pacific Northwest Laboratory
Richland, Washington 99352

ABSTRACT

A network of twelve monitoring sites around Mount St. Helens was established to evaluate possible short-lived radioactivity in the fallen ash. Seven sites were located near major population centers of Washington and Oregon, and five sites were located within 80 km of the volcano. Each site monitored the radioactivity present by the use of thermoluminescent dosimeters which recorded the total exposure to radioactivity over the exposure period. Eruptions occurring on July 22, August 7, and October 16-18, 1980 were monitored. No statistically significant quantities of measurable radon daughters were observed.

INTRODUCTION

The cataclysmic eruption of Mount St. Helens on 18 May 1980, at 08:32 a.m., Pacific Daylight Time, generated an ashfall covering much of eastern Washington and parts of Idaho and Montana. A large number of studies have been undertaken to answer questions raised by the scientific community with respect to hazards to the general populace. One question raised concerns man's exposure to radioactivity associated with the ejected ash. Fruchter et al.[1] reported radon daughter concentrations in an ash sample collected 3 hours 43 minutes after the 18 May eruption of 212, 274 and 274 pCi/g for ^{214}Pb (a β, γ emitter), ^{214}Bi (a β, γ emitter), and ^{214}Po (an α emitter), respectively. The ejection mechanism generating these radon daughter concentrations assumed that several hundred cubic kilometers of magma were purged of radon gas which was then distributed uniformly throughout the 4 km^3 of ejecta. An alternate fallout mechanism which would generate elevated radon daughter concentration was discussed; it assumed quantitative scavenging of natural atmospheric radon daughters by the falling ash. Regardless of the mechanism responsible, the presence of radon daughter activities with the deposited ash resulted in some increased radiological dose to the population above that normally received. For this reason, it was deemed prudent to initiate an experiment whereby the radioactivity content of the ash would be studied.

A sampling network was established to measure the radon daughter content of the ash from any subsequent volcanic eruptions and thereby determine the magnitude of the increased radiological dose to the population centers and other regions of the Pacific Northwest.

SAMPLING

A sampling network consisting of twelve sites surrounding Mount St. Helens at different distances was placed in operation during June 1980. These sites are listed in Table 1, together with the sampler types. Seven of these sites were near major population centers in Washington and Oregon -- Seattle, Portland, Yakima, Richland, Ritzville, Spokane, and

TABLE 1

SAMPLE LOCATIONS

Location	Type of Samplers
Yakima	Passive radon monitor, cascade impactor, and radioactive/ nonradioactive ash collector*
Packwood	Passive radon monitor
Seattle	Passive radon monitor, cascade impactor, and radioactive/ nonradioactive ash collector*
Adna	Passive radon monitor, cascade impactor, and radioactive/ nonradioactive ash collector*
Castle Rock	Passive radon monitor
Portland	Passive radon monitor, cascade impactor, and radioactive/ nonradioactive ash collector*
Cougar	Passive radon monitor
Trout Lake	Passive radon monitor
Richland	Passive radon monitor, cascade impactor, and radioactive/ nonradioactive ash collector*
Pullman	Passive radon monitor and cascade impactor
Spokane	Passive radon monitor, cascade impactor, and radioactive/ nonradioactive ash collector*
Ritzville	Passive radon monitor, cascade impactor, and radioactive/ nonradioactive ash collector*

*Not actually deployed in the field once the program was underway.

Note: Anderson Hi-Vol cascade impactor was deployed at Yakima, WA. Sierra Instruments Hi-Vol and cascade impactor were employed at all other air sampling sites.

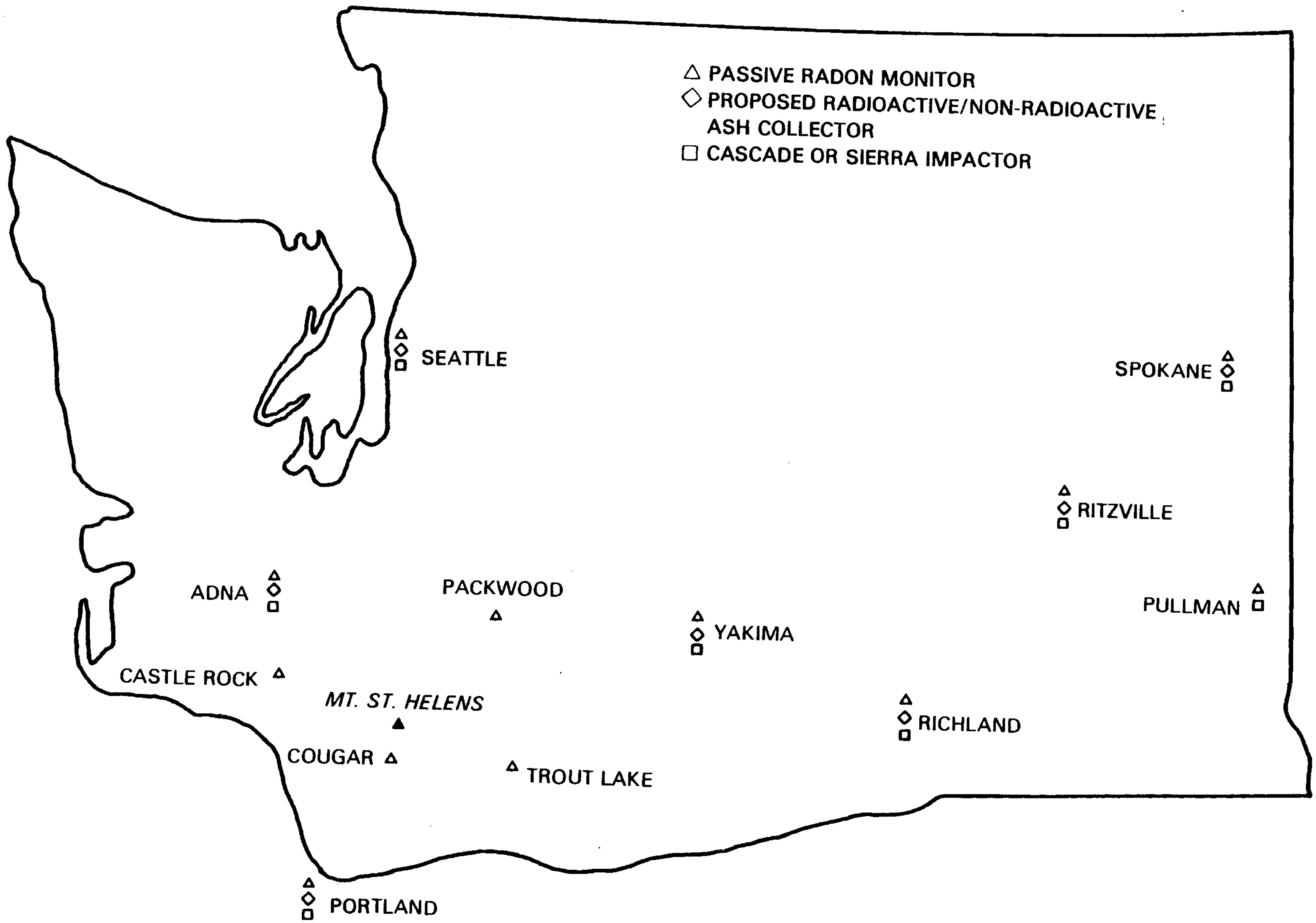

Fig. 1. Map of Sampling Sites for the Mount St. Helens Monitoring Network

Pullman. The remaining five sites were within 80 km of Mount St. Helens -- Adna, Castle Rock, Cougar, Trout Lake, and Packwood, Washington (Figure 1). Originally, it was proposed that the seven sites near major population centers would have passive radon daughter monitoring systems, ash collection systems which were to be activated by the detection of elevated levels of radioactivity, and cascade impactors to sample ashfall or resuspended ash. Passive radon daughter monitoring systems were to be placed at the remaining five sites. As will be discussed, the radioactivity triggered samplers were never deployed in the field; the prototype was designed, constructed, and is available for future projects of this type.

EXPERIMENTAL

PASSIVE RADON DAUGHTER SAMPLING

A passive radon daughter monitoring system (Figure 2) was designed to measure the integrated beta-gamma radiation dose from fresh ash fallout where the ash could not be rapidly collected and transported to radionuclide detection systems. The monitoring system consisted of a large funnel (60 cm diameter at the face) which collected and directed the ash down its sides to a small lead cave wherein thermoluminescent dosimeters (TLD's) were suspended. The inside of the cave was lined with copper to shield the TLD's from Pb X-rays produced from the interaction of cosmic rays with the lead. A small drainage hole was used to eliminate precipitation which fell into the sampler. Vials containing four 400 series[2] and four 700 series[2] TLD chips, as shown in Figure 3, were suspended within each box with the chips aligned parallel to the fallen ash. [The 400 series (CaF_2:Mn type) TLD's are approximately ten times more sensitive to radiation than are the 700 series (LiF type) dosimeters.] During ashfall, the TLD's were surrounded by the fallen ash and they absorbed the emitted radiation. An additional set of TLD's was suspended in a sealed lead cave in a configuration similar to that used for the sampling system. These TLD's measured background radiation at each site. Since samplers were placed around Mount St. Helens, data from sites not covered by ashfall generated additional atmospheric background dose data.

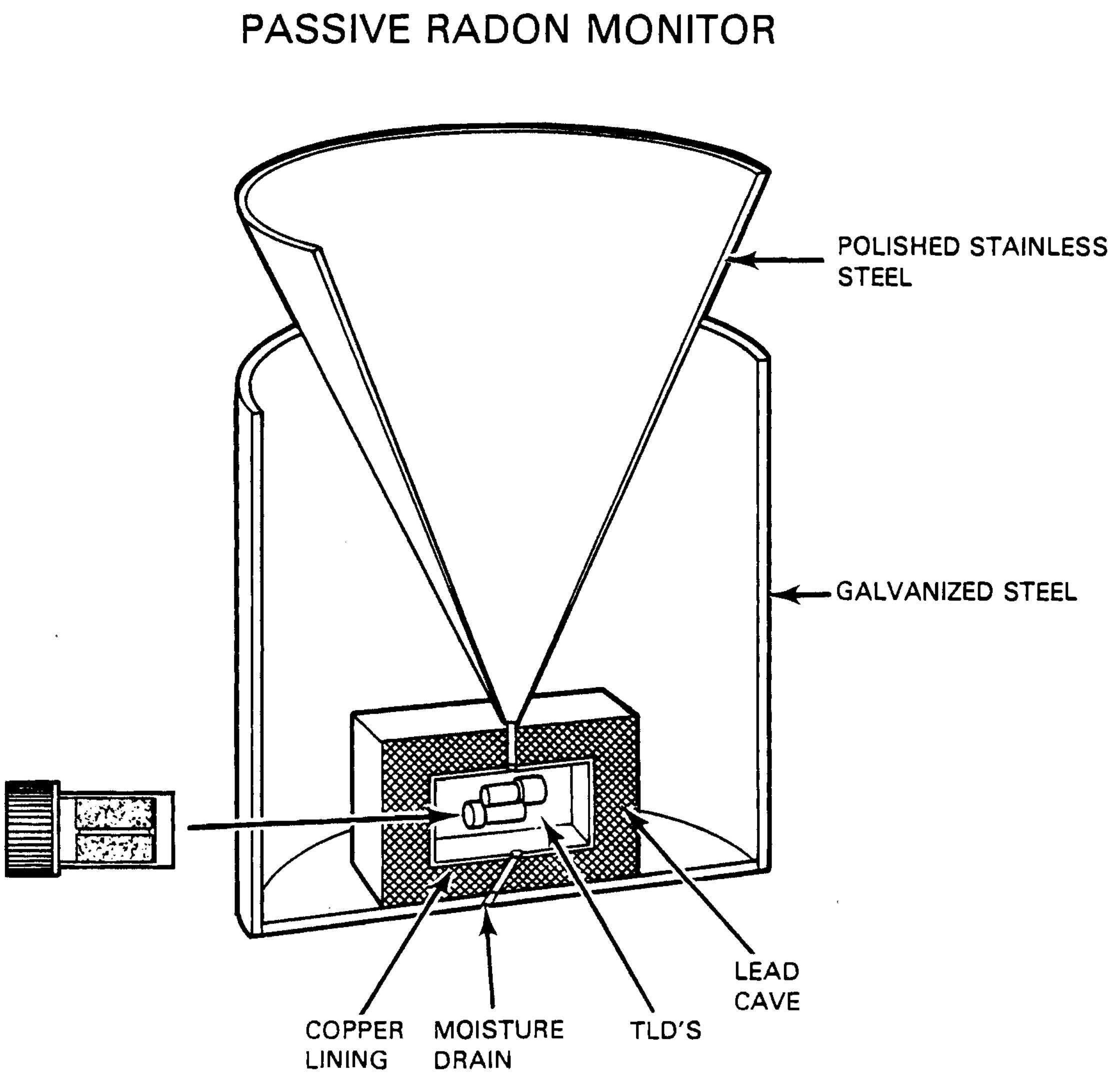

Fig. 2. Schematic of Passive Radon Daughter Monitor

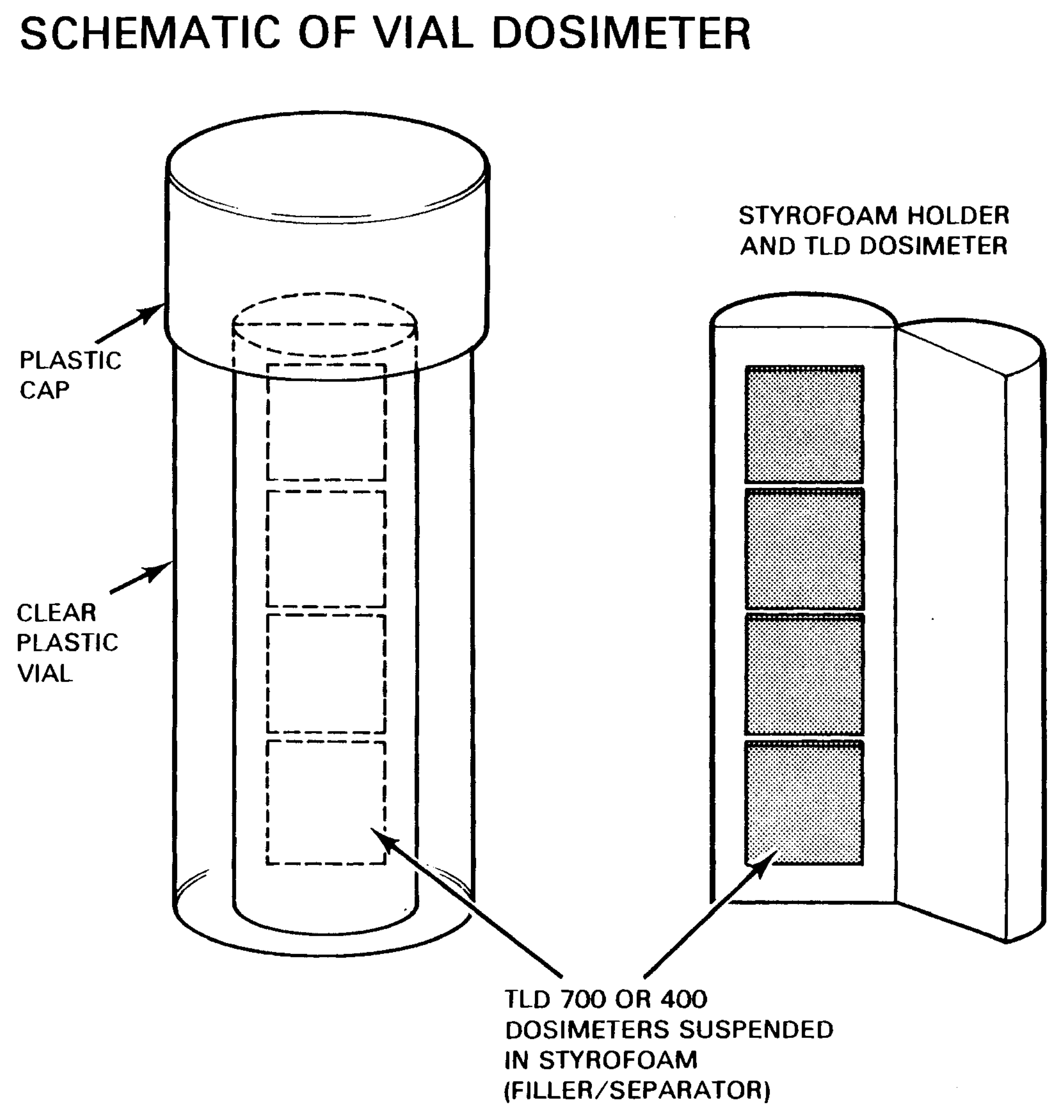

Fig. 3. Schematic of Vial Dosimeter

The radon daughter monitoring system was designed based on experience gained from a similar system being used to estimate airborne radon concentrations by measuring the radon progeny. The older system[3] was able to measure a radon daughter concentration of 1 pCi/ℓ. A calibration factor from the old system[3] was used to relate the thermoluminescent dosimeter response to the radon daughter concentration in Mount St. Helens ash. The response of the dosimeters to ash containing radon daughters was calculated to determine (1) the length of time the samplers could be left unattended and (2) whether the dose they might measure would be of significant interpretive value. The assumptions required to make these estimates include the following from the calibration of the older system[3]:

- A one-hour exposure of the older TLD system (effective volume of 705 ℓ) to an airborne radon daughter concentration of 1 pCi/ℓ results in a TLD reader response of 0.7 nanocoulomb, i.e.

  $$0.7 \text{ nC}/(1 \text{ pCi}/\ell)(705 \ \ell)(1 \text{ hr.}) = 0.00099 \text{ nC}/(\text{pCi} \cdot \text{hr}).$$

- Assuming a one-hour exposure and that the TLD chips in the old configuration[3] absorb radiation from one out of three radiation decay events, a one-hour exposure to 1 pCi/ℓ of radon daughters yields a TLD reader response of 0.00033 nanocoulombs, i.e.

  $$1/3 \ (0.00099 \text{ nC}/(\text{pCi} \cdot \text{hr})) = 0.00033 \text{ nc}/(\text{pCi} \cdot \text{hr}).$$

  Hereafter, this will be called the TLD response equivalency.

The calculation of the expected response of the new passive dosimeter system (vial TLD system) to typical monthly background radon daughter concentrations required the following assumptions:

- A maximum exposure to background radiation of one month (720 hours).
- An average total background radiation of 1 pCi/ℓ for the airborne radon daughters.
- The TLD's are continuously exposed to a maximum volume of 1.8 ℓ of ambient air (the volume of air inside the ash collector).

This criteria would generate a TLD response of 0.43 nanocoulombs, i.e.,

$$(0.00033 \text{ nc}/(\text{pCi} \cdot \text{hr}))(1 \text{ pCi}/\ell)(1.8 \ \ell)(720 \text{ hr.}) = 0.43 \text{ nC}.$$

Hereafter, this will be called the monthly TLD background response. Similarly, a two day exposure to the typical background radiation would generate an expected dosimeter response of 0.029 nanocoulombs.

The calculation of the expected response of the new passive dosimeter (vial TLD's) to ashfall following an eruption of Mount St. Helens required the following assumptions:

- Deposition of 1 gram of ash in the ashfall collector/radon monitor system should deliver 212, 274, and 274 pCi/g of ^{214}Pb, ^{214}Po, and ^{214}Bi, respectively, according to the report by Fruchter et al.[1] Thus, there are 760 pCi of measurable radon daughters (α, β, and γ emitters) or 486 pCi of beta and gamma emitters at the time of sample collection per gram of ash.
- All beta and gamma radiation from the measurable radon daughters in the ash (^{214}Pb and ^{214}Bi) was absorbed by the TLD's in four hours (approximately eight half-lives). The first half-life would result in the TLD's measuring 486 pCi, the next would be 243 pCi, then 121.5 pCi, and so on until total decay has occurred. Thus the total integrated radon daughter (^{214}Pb and ^{214}Bi) exposure would be the sum of these events or about 970 pCi.
- A geometry factor of 0.5 was used to account for ash deposited on the bottom of the cave radiating away from the suspended TLD's. This is in addition to the geometry factor incorporated into the TLD response equivalency where an hour exposure to 1 pCi/ℓ of radon daughters yields a TLD reader response of 0.00033 nanocoulombs.
- All TLD's were assumed to respond to incident radiation in a similar fashion, although the packaging of the TLD's in the two systems was not identical.

A calculation was performed to determine the vial TLD response to an exposure equivalent to the radon daughter concentration measured following the May 18 explosion. The estimate of energy absorbed by the vial TLD's due to an ashfall was based on the above mentioned assumptions where 1 g of ash was deposited, the total integrated radon daughter exposure was equivalent to 970 pCi, the geometry factor was 0.5 (since the TLD's would see 1 out of 2 radiations emitted from ash on the bottom of the container), and the passive radon daughter monitor system responded similarly to the calibrated system[3], such that a TLD response of 0.16 nanocoulombs would be expected, i.e.,

$$(970\ \text{pCi} \cdot \text{hr})(0.5)(0.00033\ \text{nC}/(\text{pCi} \cdot \text{hr})) = 0.16\ \text{nC}.$$

Thus, a radon daughter concentration (i.e., ^{214}Pb and ^{214}Bi) of 486 pCi in 1 g of ash as reported by Fruchter et al. would yield a thermoluminescent dosimeter response of 0.16 nanocoulombs. This would be easily discernible over the expected two-day background TLD response of 0.029 nanocoulombs. The ashfall response would also be discernible compared to an expected TLD response of 0.43 nanacoulombs from the background for even a month-long TLD exposure. The sensitivity of the method is directly related to the amount of background radiation measured by the TLD chips.

The passive TLD Radon Measurement System was not calibrated for dose prior to field installation. It was felt that the calibration could be done at a later date if the data gathered in the field gave evidence of highly elevated radiation dose in the TLD chips due to the ashfall.

The TLD chips were exposed for periods of one day to five weeks, depending on the eruptive activity of Mount St. Helens. Following completion of an eruptive cycle, the TLD's were changed and returned to the Laboratory for immediate analysis. For each cycle, system blanks were obtained by handling a set of TLD chips as if they were samples. However, they were not exposed as the sample TLD chips were. The dosimeters were analyzed by the use of a Harshaw[2] "TLD" system and a procedure similar to that reported by Wogman et al.[4]

SIZED PARTICULATE COLLECTION

Particulates in the respirable size range (<10 μm) were collected principally by use of a Sierra Instruments cascade impactor system[5] which was deployed at six sites. One site was supplied with an Andersen[6] Hi-Vol cascade impactor. The Sierra Instruments air sampling units (Figure 4)

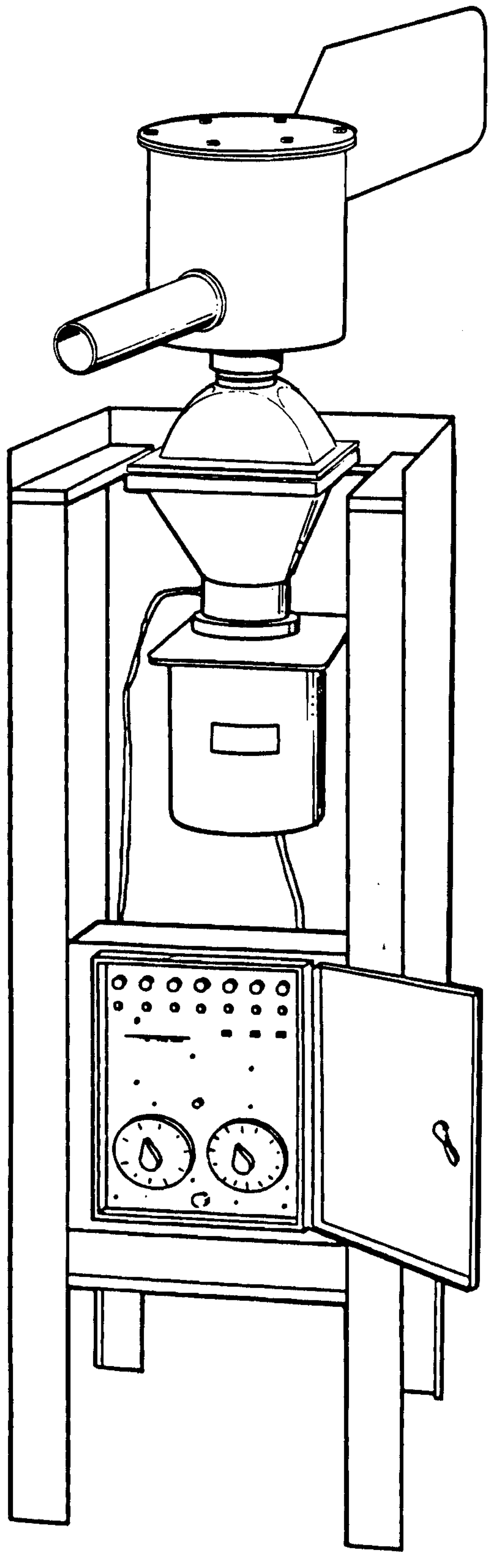

Fig. 4. Schematic of Sierra Instruments High Volume Air Sampling System

consisted of a cyclone preseparator (Model 230CP), 5-stage cascade impactor (Series 230), flow controller (Model 310A), and a high volume sampling motor. Particulates were collected on Whatman 41 cellulose paper which has relatively low concentrations of blank elements. The flow rate used for the Sierra Instruments cascade impactor was 40 SCFM while that used for the Andersen cascade impactor was 20 SCFM. These systems were calibrated against a dry gas meter before placement in the field.

The collection of air samples required electrical power and the help of a responsible individual at each site. Procedures called for operation of the impactor systems only at those times following an eruption when ashfall was visible at the sampling location. Only the Yakima, Washington site received sufficient ashfall in a short time to warrant sampling. The radioactivity content of the samples was measured with large NaI(Tl) gamma-ray spectrometers[7]. No elevated radioactivity levels above background fallout were measured in these samples.

RADIOACTIVE/NONRADIOACTIVE ASH COLLECTION

When this program was conceived, it was planned to modify wet/dry fallout collectors[8] to allow nonradioactive and radioactive ash to be collected at remote sites. The system designed used a thin window Geiger-Müller tube to monitor the radiation level of the environment. Whenever the background increased a factor of 1.5-fold above that of the five-minute average, a mechanism was actuated, by which a fresh ashfall collection bucket was exposed and the previous bucket was covered. Therefore, fresh ash would be collected which would be returned to the laboratory for radionuclide measurement. In the first month of this study, it was observed that the eruptions occurring after the May 18th eruption were of both variable strength and ash content with none approaching that of May 18. The collection devices already at the sampling sites were receiving very small ash deposits even if they were located directly in the path of the plume. It was determined that the new device would not receive sufficient ash to activate its cover mechanism. This information, with other logistical complications such as the short time allowed for transit because of the short half-lives of the radon daughters, caused the decision to be made to forego the modification of additional wet/dry collectors, and thus these samplers were not placed in field use.

DISCUSSION AND RESULTS

PASSIVE RADON DAUGHTER MONITORING

Monitoring systems employing thermoluminescent dosimeters (TLD's) were placed at the twelve sites shown in Figure 1. The vial TLD's were initially used, with a set of LiF type dosimeters (700 series) and a set of CaF_2:Mn type dosimeters (400 series) at each site. The length of exposure of the TLD's was approximately one month, unless an eruption occurred, in which case they were changed immediately. The exposure periods are shown in Table 2. The exposure periods span a time in which there were three eruptions of Mount St. Helens (July 22, August 7, and October 16-18, 1980). Table 3 lists qualitatively (and when possible, quantitatively) the amount of ash collected at each site in the passive radon daughter monitoring and ash collection systems. The ash deposition in conjunction with the ashfall maps verified whether a sample had been in the ashfall path. If the sampling site indicated deposition of ash but was not in the ashfall path (i.e., Ritzville, Yakima, and Richland), deposition of resuspended ash from the earlier eruptions or wind blown dust was assumed.

There were essentially four month-long sampling periods for all sites. The July 22 eruption occurred just after the TLD's had been changed at Yakima and Packwood. The TLD's from Yakima and Packwood were recovered and returned to the Laboratory after the ash cloud had passed these two sites and resulted in the additional exposure period for these two sites. Similarly, following the August 7 eruption, the TLD's were changed at Packwood and Adna and returned to the Laboratory for analysis. The data measured by the 400 and 700 series TLD's in the vial configuration are listed in Tables 4 and 5, respectively. The results reported are the differences between the average TLD readings (for beta and gamma) obtained in the ash collection cave and the average TLD readings (for beta and gamma) obtained in the "background" cave. The reported uncertainty is the standard deviation of the mean for the ash collection and background cave with the error propagated to account for the subtraction to obtain the net TLD reading due to ashfall. The background cave data for the 400 series TLD's were puzzling in that more dose was measured in the background TLD chips than in the ash sampler TLD chips. Recognizing that they are 10

TABLE 2

EXPOSURE PERIOD DATES FOR THE THERMOLUMINESCENT DOSIMETERS AND ASH COLLECTORS

Sample Site	Exposure Period During 1980			
	I	II	III	IV
Yakima	6/24-7/22 7/22-7/23[a]	7/23-8/25[b]	8/25-9/25	9/25-10/23
Packwood	6/24-7/22 7/22-7/23[a]	7/23-8/11[b] 8/11-8/25	8/25-9/25	9/23-10/23
Seattle	6/25-7/23	7/23-8/26	8/26-9/24	9/24-10/27
Adna	6/24-7/23	7/23-8/11 8/11-8/25	8/25-9/23	9/23-10/21
Castle Rock	6/25-7/23	7/23-8/25	8/25-9/23	9/23-10/21
Portland	6/26-7/24	7/24-8/27	8/27-9/23	9/23-10/21[c]
Cougar	6/25------------------	-8/26	8/26-9/23	9/23-10/29[c]
Trout Lake	6/26-7/24	7/24-8/27	8/27-9/22	9/22-10/20[c]
Richland	6/30-7/25	7/25-8/8 8/8-8/28	8/28-9/25	9/25-10/23
Pullman	6/20-7/31	7/31-9/5	9/5-10/2	10/2-10/28
Spokane	6/20-7/31	7/31-9/4	9/4-10/2	10/2-10/27
Ritzville	6/19-7/31	7/31-9/4	9/4-10/3	10/3-10/27

a-Sites located in eruption plume path following the July 22, 1980 eruption.
b-Sites located in eruption plume path following the August 7, 1980 eruption.
c-Sites located in eruption plume path following the October 16-18, 1980 eruption.

TABLE 3

SAMPLE DEPOSITION IN THERMOLUMINESCENT DOSIMETER CAVE OVER EXPOSURE PERIODS (MASS IN GRAMS)

Sample Site	Exposure Period During 1980			
	I	II	III	IV
Yakima	1.0 1.0[a]	2.9[b]	Trace	0.4
Packwood	None Trace[a]	9.6[b] None	None	None
Seattle	None	None	None	None
Adna	None	Trace None	None	None
Castle Rock	Trace	Trace	Trace	Trace
Portland	None	None	None	6.1[c]
Cougar	– – – – – – – →	Trace	Trace	32[c]
Trout Lake	None	None	None	6.2[c]
Richland	None	None None	None	0.5
Pullman	None	None	None	None
Spokane	None	None	None	None
Ritzville	6.2	Trace	None	None

a-Sites located in eruption plume path following the July 22, 1980 eruption.
b-Sites located in eruption plume path following the August 7, 1980 eruption.
c-Sites located in eruption plume path following the October 16-18,1980 eruption.

TABLE 4

SAMPLE RESULTS* FOR THE CaF_2:Mn (400 SERIES, NOT LIGHT SHIELDED) THERMOLUMINESCENT DOSIMETERS (TLD READER UNITS IN NANOCOULOMBS†)

Sample Site	Exposure Period I	II	III	IV
Yakima	-2.3 ± 0.2	-5.9 ± 0.4[b]	-3.0 ± 0.4	-2.3 ± 0.3
	-1.2 ± 0.2[a]			
Packwood	-0.8 ± 0.3	-2.3 ± 0.3[b]	-0.9 ± 0.2	-2.9 ± 0.3
	-1.0 ± 0.2	-0.2 ± 0.2		
Seattle	-3.5 ± 0.1	-6.0 ± 0.3	-4.6 ± 0.2	-1.9 ± 0.1
Adna	-3.2 ± 0.1	-4.0 ± 0.3	-5.4 ± 0.3	-3.8 ± 0.1
		-1.8 ± 0.2		
Castle Rock	-3.8 ± 0.2	-4.7 ± 0.4	-5.1 ± 0.5	-2.4 ± 0.1
Portland	-4.8 ± 0.2	-6.2 ± 0.3	-4.7 ± 0.2	-2.3 ± 0.1[c]
Cougar	– – – – – →	-1.9 ± 0.4	-3.8 ± 0.4	-2.4 ± 0.4[c]
Trout Lake	-2.9 ± 0.2	-4.7 ± 0.3	-4.2 ± 0.4	-3.3 ± 0.3[c]
Richland	-1.7 ± 0.3	-2.8 ± 0.2	-3.0 ± 0.4	-3.2 ± 0.4
		-2.1 ± 0.2		
Pullman	6.8 ± 0.2	-6.8 ± 0.5	-1.1 ± 0.3	-5.2 ± 0.2
Spokane	5.2 ± 0.2	-7.0 ± 0.3	-2.6 ± 0.3	-4.8 ± 0.2
Ritzville	-0.5 ± 0.2	-2.5 ± 0.1	-1.3 ± 0.1	-3.7 ± 0.2

*Reported result is the ashfall collector TLD corrected for background.

†Reported error is the standard deviation of the mean for the ash collector and background TLD result with the error being propogated when the net reading is calculated.

a-Sites located in eruption plume path following the July 22, 1980 eruption.
b-Sites located in eruption plume path following the August 7, 1980 eruption.
c-Sites located in eruption plume path following the October 16-18, 1980 eruption.

TABLE 5

SAMPLE RESULTS* FOR THE LiF (700 SERIES, NOT LIGHT SHIELDED VIAL CONFIGURATION) THERMOLUMINESCENT DOSIMETERS (TLD READER UNITS IN NANOCOULOMBS†)

Sample Site	Exposure Period I	II	III	IV
Yakima	0.07 ± 0.05 -0.06 ± 0.10[a]	0.15 ± 0.08[b]	0.09 ± 0.04	0.08 ± 0.04
Packwood	0.29 ± 0.09 -0.04 ± 0.02[a]	-0.02 ± 0.05[b] -0.002 ± 0.062	0.39 ± 0.10	0.16 ± 0.09
Seattle	-0.005 ± 0.033	-0.18 ± 0.09	-0.01 ± 0.07	-0.01 ± 0.14
Adna	0.13 ± 0.03	-0.004 ± 0.081 0.09 ± 0.07	0.13 ± 0.05	0.04 ± 0.02
Castle Rock	0.11 ± 0.03	0.26 ± 0.12	0.13 ± 0.06	0.07 ± 0.02
Portland	0.15 ± 0.07	0.08 ± 0.06	-0.04 ± 0.10	0.18 ± 0.04[c]
Cougar	--------→	0.58 ± 0.10	0.49 ± 0.08	0.30 ± 0.05[c]
Trout Lake	0.00 ± 0.06	0.13 ± 0.10	0.04 ± 0.06	0.05 ± 0.05[c]
Richland	0.10 ± 0.05	0.02 ± 0.08 0.16 ± 0.04	0.13 ± 0.05	0.15 ± 0.05
Pullman	0.18 ± 0.06	0.24 ± 0.06	0.16 ± 0.06	0.20 ± 0.02
Spokane	-0.04 ± 0.06	-0.13 ± 0.06	0.11 ± 0.05	0.23 ± 0.04
Ritzville	0.62 ± 0.08	0.32 ± 0.04	0.20 ± 0.05	0.18 ± 0.06

*Reported result is the ashfall collector TLD corrected for background.

†Reported error is the standard deviation of the mean for the ash collector and background TLD result with the error being propogated when the net reading is calculated.

a-Sites located in eruption plume path following the July 22, 1980 eruption.
b-Sites located in eruption plume path following the August 7, 1980 eruption.
c-Sites located in eruption plume path following the October 16-18, 1980 eruption.

times more sensitive than the 700 series TLD's does not account for observing large negative numbers. The possiblity of a "sun-bleaching" effect on the TLD's exposed to sunlight entering the sampling chamber through the funnel was investigated. The 700 series TLD data cannot support or reject this hypothesis. The question of sun-bleaching has been further investigated(9), and it was found that exposure to sunlight can reduce the latent thermoluminescence in a chip that was exposed to a radiation field. Regardless of the cause, an apparent error was introduced into the results of the 400 series TLD's, which eliminated their use or their data interpretation.

To circumvent the possible sun-bleaching problem, light shielded disk dosimeters (Figure 5) were also placed in the field with the vial dosimeters. They were tested and then partially deployed in exposure period III with a full set being deployed over the last exposure period. A qualitative comparison of data from Tables 5 and 6 does not show a significant difference in the data from the 700 series of LiF TLD's whether used in the vial configuration or in the light-shielded disk configuration. Thus, it seems that the 700 series TLD's in the vial configuration provided an acceptable measurement technique.

The TLD data must be interpreted in a relative sense. The vial TLD system monitored $\beta + \gamma$ radiation present in both the collector and background cave. The difference between the data recorded in the sample cave TLD's and the background cave TLD's was the $\beta + \gamma$ radiation due to whatever had been deposited in the collection cave (presumably due to radon daughters in any ashfall).

The light-shielded disk dosimeter measured both $\beta + \gamma$ and γ, individually in the ash collector cave, so that the difference calculated is energy deposited due only to β radiation in the ash collector cave. Therefore, one cannot quantitatively compare the results from the vial TLD's with those of the disk TLD's.

Again, note that the vial TLD data is the difference between the $\beta + \gamma$ measured due to ashfall in the ash collector cave and the $\beta + \gamma$ measured due to the natural background radiation in the background cave. In comparison, the disk-dosimeters reported the difference measured in the ashfall

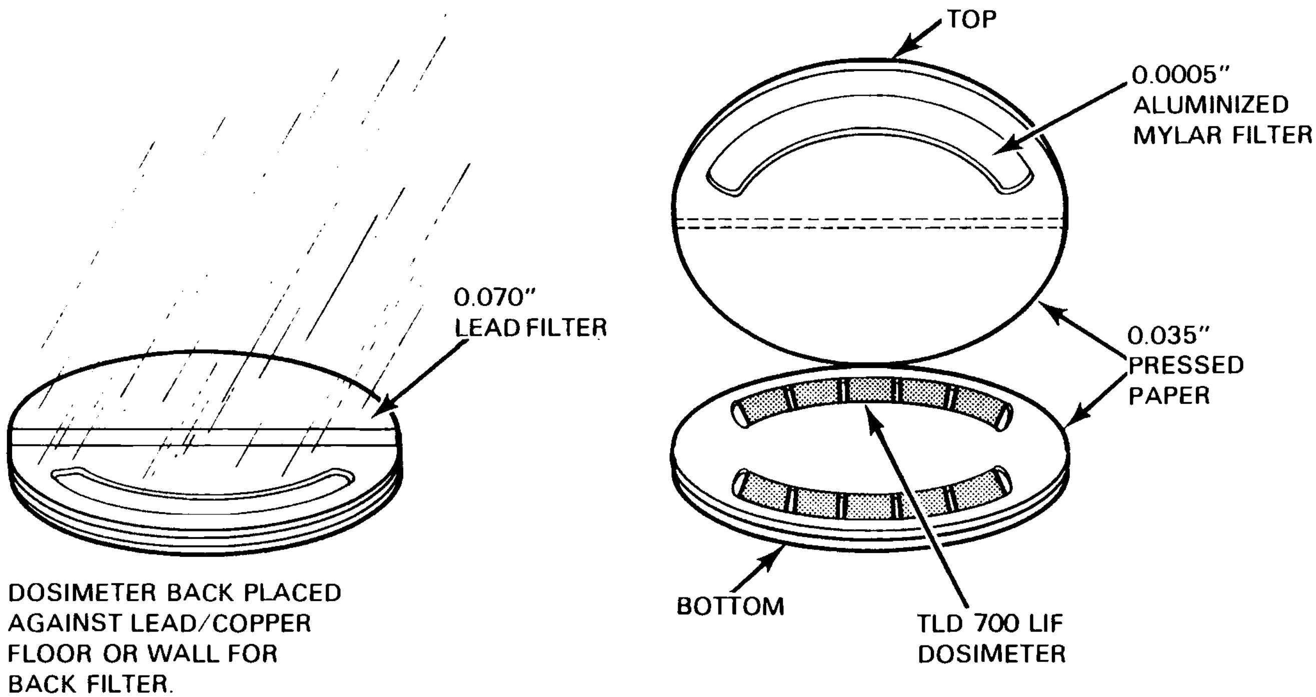

Fig. 5. Schematic of Disk Dosimeter

TABLE 6

SAMPLE RESULTS* FOR THE LiF (700 SERIES, LIGHT-SHIELDED DISK CONFIGURATION) THERMOLUMINESCENT DOSIMETERS (TLD READER UNITS IN NANOCOULOMBS†)

Sample Site	Exposure Period I	II	III	IV
Yakima	–	–	–	-0.002 ± 0.131
Packwood	–	–	–	-0.11 ± 0.04
Seattle	–	–	–	-0.06 ± 0.08
Adna	–	–	–	0.16 ± 0.05
Castle Rock	–	–	–	0.001 ± 0.056
Portland	–	–	–	0.20 ± 0.05**
Cougar	–	–	–	0.19 ± 0.08**
Trout Lake	–	–	–	0.06 ± 0.07**
Richland	–	–	–	0.14 ± 0.08
Pullman	–	–	0.18 ± 0.10	-0.15 ± 0.07
Spokane	–	–	0.05 ± 0.07	0.10 ± 0.08
Ritzville	–	–	0.02 ± 0.11	0.32 ± 0.06

*Reported result is the β present, defined as the difference between the β + γ and the γ measurement obtained with the disk TLD in the ash fall collector.

†Reported error is the standard deviation of the mean for the β + γ measurement and the γ measurement with the error being propagated when the net β is calculated.

** Sample sites located in the path of the eruption cloud from the third eruption (October 16-18, 1980).

collector cave between the $\beta + \gamma$ radiation and the γ radiation. Consequently, the measurable β radiation present in the ashfall collector cave was reported.

The eruption dates of Mount St. Helens were July 22, August 7, and October 16-18, 1980. As mentioned earlier, Table 5 presents data for the 700 series vial TLD's and Table 6 presents data for the 700 series light-shielded disk TLD's. The July 22, 1980 eruption cloud traveled to the northeast and passed over the sites at Packwood and Yakima (Figure 6). This is confirmed by the small amount of ash deposited in the container as listed in Table 3. At this time, only the vial TLD's were being used. Fortuitously, the day before the eruption, field personnel had just changed the TLD's at these two sites. Therefore, if these TLD's were exposed to radioactive ash, there should have been very little "sun-bleaching" or background accumulation, as these chips were exposed for only two days. (Note that part of the time the collector was under the ash cloud.) As the data show, no discernibly elevated radiation levels were observed relative to the levels measured by TLD's exposed during the prior sampling period.

The second eruption on August 7, 1980 had an eruption cloud that traveled to the northeast (Figure 7). This ash cloud also passed over the sites at Packwood and Yakima, as confirmed by the deposition of ash noted in Table 3. No discernibly elevated radiation levels were observed relative to the radiation levels measured by TLD's exposed at other sites.

The third eruption sequence on October 16-18, 1980 had eruptive ash clouds that traveled to the south on very light winds. The ash plume passed over the sampling sites at Cougar, Trout Lake, and Portland (Figure 8). This was confirmed by significant deposition of ash at the site of Cougar (32 g), and smaller amounts of ash at the Trout Lake site (6.2 g) and the Portland site (6.1 g) (see Table 3). The vial and disk TLD reader response data appeared significantly elevated for the Portland and Cougar sampling sites. The vial and disk TLD response data for the Trout Lake site was similar to data from non-ashfall sites and thus was not significant. The TLD reader response per gram of ash was calculated for the sampling sites of Portland and Cougar. The vial TLD reader response was 0.030 and 0.009 nanocoulombs per gram of ash for the sites of Portland and Cougar, respectively. The disk TLD response was

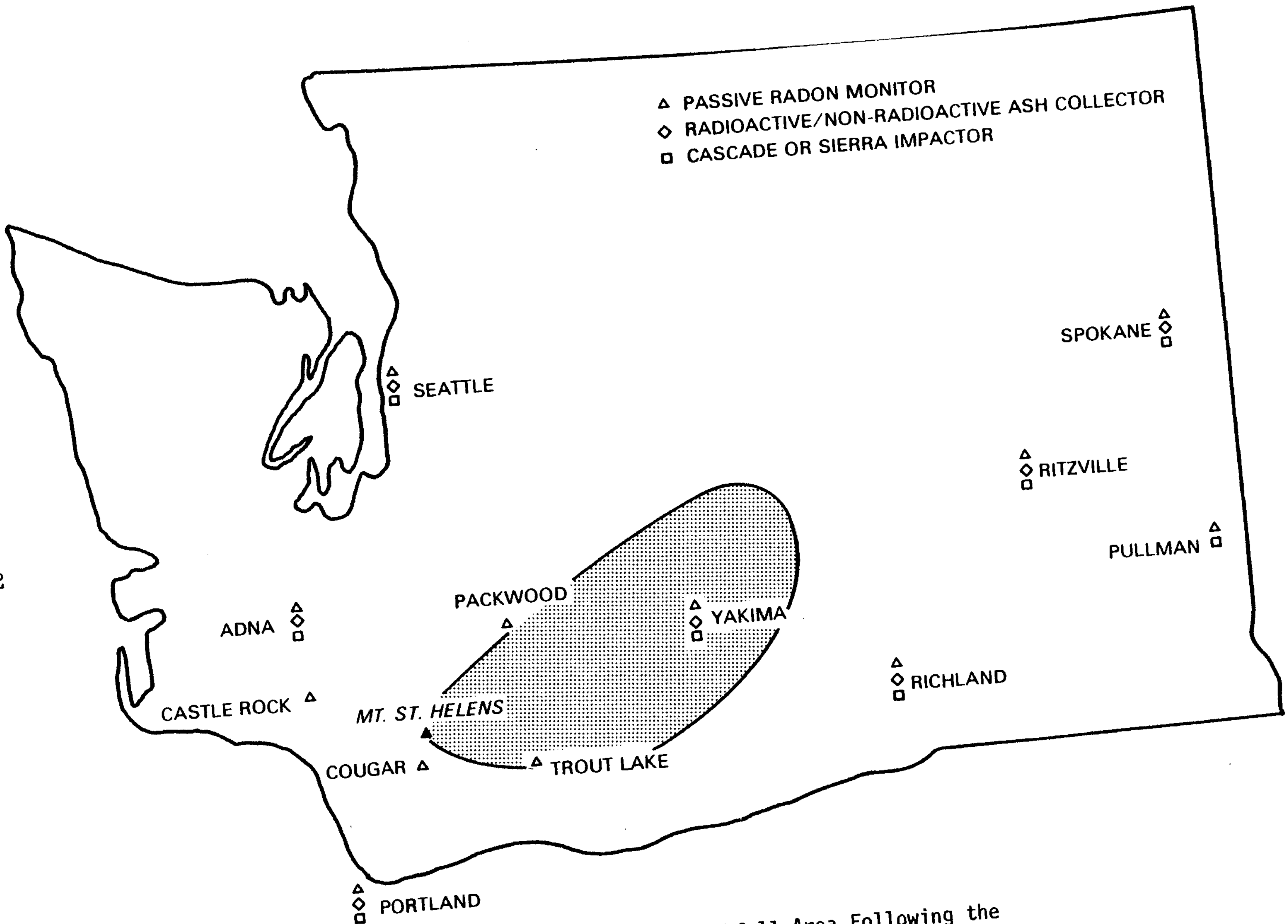

Fig. 6. Map of Ashfall Area Following the July 22, 1980 Eruption

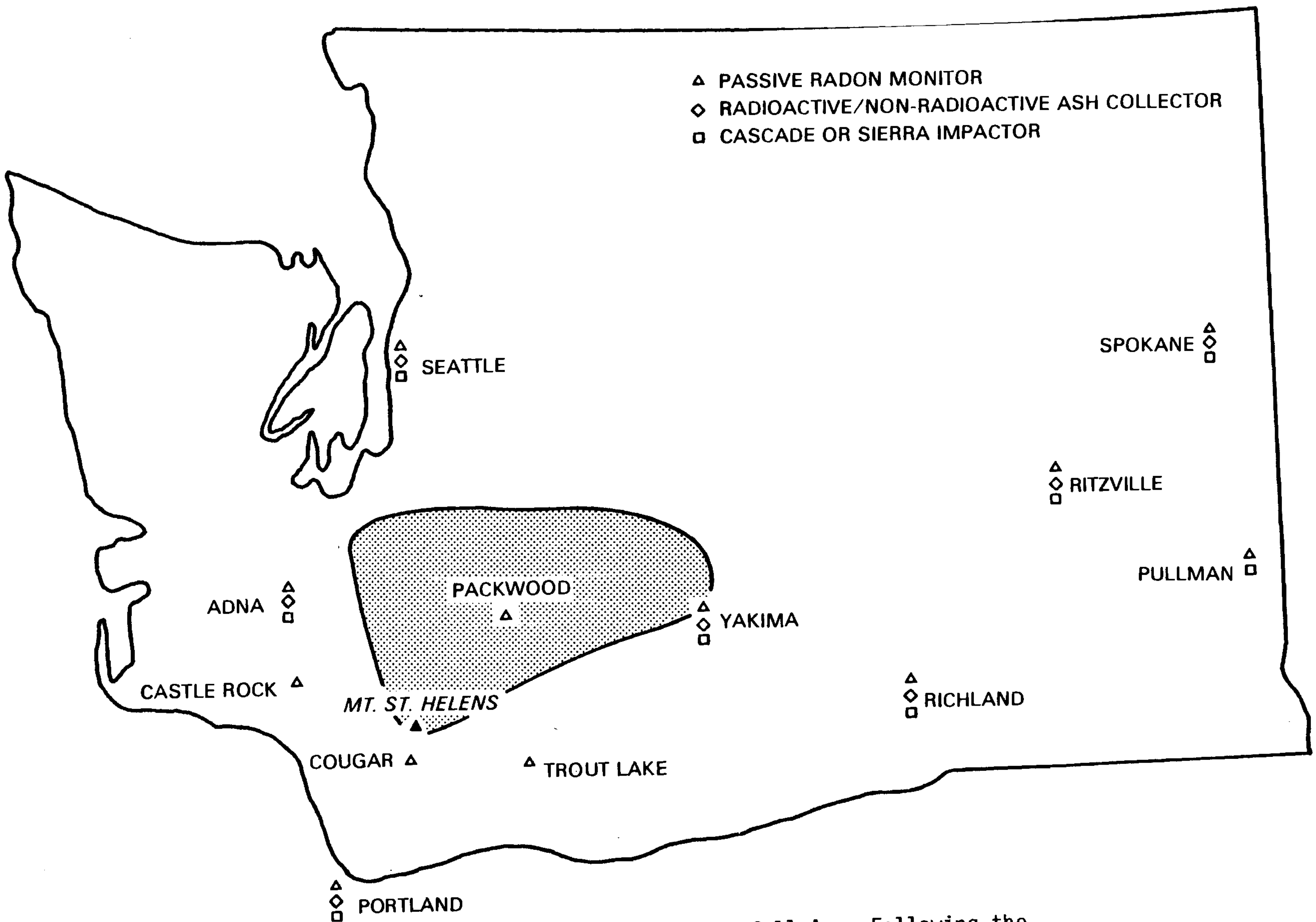

Fig. 7. Map of Ashfall Area Following the August 7, 1980 Eruption

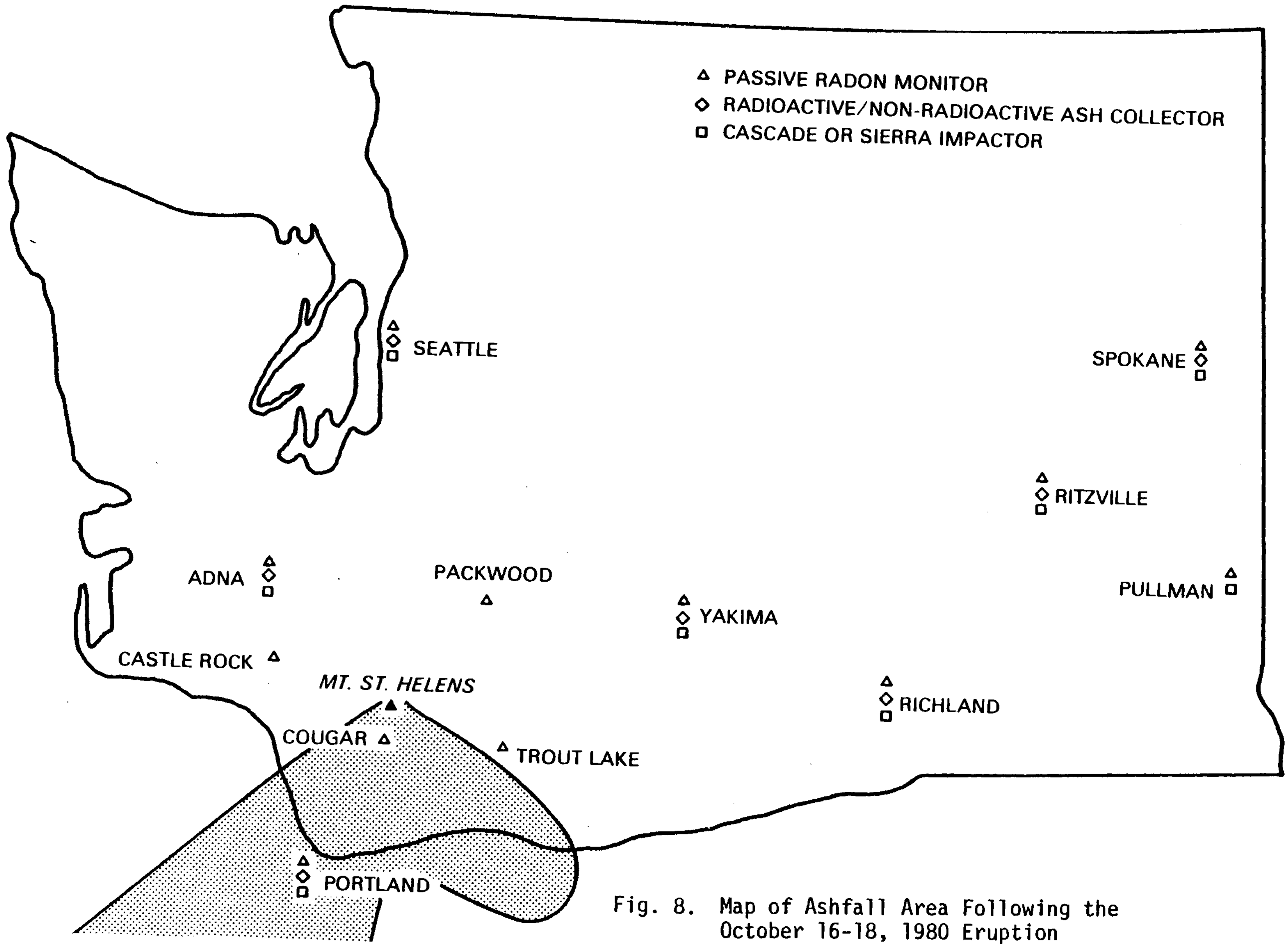

Fig. 8. Map of Ashfall Area Following the October 16-18, 1980 Eruption

0.033 and 0.006 nanocoulombs per gram of ash for the sites of Portland and Cougar, respectively.

The calculated TLD reader response expected from 1 g of ash containing 212 and 274 pCi/g of ^{214}Bi, as discussed earlier, would result in a difference of 0.16 nanocoulombs between the radiation observed in the sampler TLD and the background TLD system. However, the data for the sampling sites located in the ashfall areas do not show such a response. In particular, the TLD reader response (vial or disk) per gram of ash for the Portland and Cougar sites during period IV was much lower than the calculated TLD response of 0.16 nanocoulombs per gram of ash for the May 18 eruption.

The "t" test of significance between two sample means was applied to the data in Tables 5 and 6. That is, the null hypothesis was chosen such that H: $\mu_1 = \mu_2$. The mean, μ_1 was chosen from either the total ashfall data set or an individual value from the ashfall data set ($x_1, x_2, .., x_{n1}$). The mean, μ_2, was derived from the non-ashfall data set ($y_1, y_2, .., y_{n2}$). The unpaired variate form of the t-test[10] was used which included the pooled variance of the two sets of data to calculate a t value for $N_1 + N_2 - 2$ degrees of freedom. The calculated t value was then compared to the critical value[10] for $N_1 + N_2 - 2$ degrees of freedom and P = 5%, where the P value is the probability that t will exceed the critical value if there is no real difference between the means being tested. Thus, a confidence level of 95% was chosen.

Table 7 lists the calculated t values. Only data within a sampling period were tested against each other. Each data value was tested against the non-ashfall data points and all the ashfall data points were tested against the non-ashfall data points. The footnote at the bottom of Table 7 lists the critical values for P = 5% and the indicated degrees of freedom. This test suggests the following TLD reader results are significant for the vial TLD data - Period I: Ritzville; Period II: Cougar, and Period III: Cougar - and for the disk TLD's - Period IV: Pullman. The Period I data from the Ritzville site corresponded to a deposition of 6.2 grams of ash in the ash collector but Ritzville was not located in the path of an eruption plume during the sampling period. The Ritzville site was hard hit with several inches of ash following the May 18 eruption. The ash was still present during the exposure period and

TABLE 7

CALCULATED t VALUES[a] FOR VIAL AND DISK TLD DATA EXPOSURE PERIOD

Sample Site	I Vials vs. Non-Ashfall	II Vials vs. Non-Ashfall	III Vials vs. Non-Ashfall	IV Vials vs. Non-Ashfall	III Disk vs. Non-Ashfall	IV Disk vs. Non-Ashfall
Yakima	$-.44_{9}$	0.13_{12}	-0.43_{10}	-0.58_{7}	—	-0.38_{7}
	-1.08_{10}					
Packwood	0.82_{9}	-0.69_{12}	1.79_{10}	0.49_{7}	—	0.80_{7}
	-0.96_{10}	-0.62_{11}				
Seattle	-0.85_{9}	-1.6_{11}	-1.1_{10}	-2.0_{7}	—	-1.02_{7}
Adna	-0.08_{9}	-0.63_{11}	-0.12_{10}	-1.0_{7}	—	1.37_{7}
		-0.17_{11}				
Castle Rock	-0.20_{9}	0.70_{11}	-0.16_{10}	-0.69_{7}	—	-0.35_{7}
Portland	0.03_{9}	-0.22_{11}	-1.4_{10}	0.72_{8}	—	1.54_{8}
Cougar		3.18^{*}_{11}	3.05^{*}_{10}	2.10_{8}	—	1.43_{8}
Trout Lake	-0.82_{9}	0.06_{11}	-0.72_{10}	0.89_{8}	—	0.21_{8}
Richland	-0.26_{9}	-0.53_{11}	-0.12_{10}	0.32_{7}	—	1.06_{7}
		0.21_{11}				
Pullman	0.19_{9}	0.63_{11}	0.03_{10}	1.05_{7}	5.42_{1}	-2.53^{*}_{7}
Spokane	1.06_{9}	-1.3_{11}	-0.26_{10}	1.59_{7}	-0.36_{1}	0.48_{7}
Ritzville	5.01^{*}_{9}	1.01_{11}	0.34_{10}	0.69_{7}	-0.85_{1}	-0.04_{7}
All Ashfall Sites	-1.45_{11}	-0.39_{13}	—	0.90_{10}	—	1.74_{10}

[a]The first value is the calculated t value. The subscript is the degrees of freedom.

*Indicates significant value by t test as determined by use of crititcal values from the CRC Handbook (49th ed.) for a P value of 5%. The degrees of freedom and critical value are as follows: 1, 12.71; 7, 2.36; 8, 2.31; 9, 2.26; 10, 2.23; 11, 2.20; 12, 2.18; and 13, 2.16.

was being resuspended. Thus, the vial TLD data from the Ritzville site cannot be the result of radon daughter activity due to an eruption of Mount St. Helens during period I. It may be due to resuspended dust which scavenged radon daughters from our normal atmosphere.

The vial TLD data obtained for the Cougar sitethat spanned Period I and II and the vial TLD data for Period III are significant according to the "t" test. The Cougar site was located within 14 km of Mount St. Helens and received significant ash deposition following the May 18 eruption. The TLD data for Period I and II may be real, as it is possible for ash from the July 22 and August 7 eruption to have reached the Cougar site even though the main plume went northeast (see Figures 6 and 7). However, the positive t test value for Period III data from the Cougar site cannot be attributed to Mount St. Helens because the data was obtained during a period of non-eruptive activity. Therefore, both TLD reader response values from the Cougar site must be suspect.

The disk TLD data for Period IV suggested the TLD reader response data for the Pullman site was significant. This appeared to be a fortuitous result. The positive response was not substantiated by the vial TLD data for the Pullman site; ash deposition (Table 3) at Pullman was not noted; and, finally, the eruption plume of the October 16-18 eruption did not reach or travel in the direction of Pullman. Thus, this value should also be discounted and not considered significant.

The LiF TLD response data (shielded and not-shielded) were used to obtain average TLD exposure data for the ashfall and non-ashfall sites. The average vial and disk TLD reader response data for non-ashfall and ashfall exposures are shown in Table 8. The non-ashfall exposure was considered to be the atmospheric/ash background. The difference between the ashfall and non-ashfall average (Δ) is thus the maximum exposure observed due to ashfall from Mount St. Helens.

The vial TLD's recorded the exposure for three eruptions of Mount St. Helens. The eruption on July 22, 1980 had an ashfall exposure of -0.05 ± 0.01 nanocoulombs. The negative number resulted from inclusion of the background term in the calculation. The subtraction of the non-ashfall average yields an exposure value of -0.20 ± 0.18 nanocoulombs--clearly, not a positive response.

TABLE 8

AVERAGE DOSIMETER RESULTS FOR ASHFALL
AND NON-ASHFALL AREAS
(TLD READER UNITS IN NANOCOULOMBS)

	Exposure Period			
	I	II	III	IV
		LiF TLD's (Not Shielded-Vial)		
Non-Ashfall	0.15 ± 0.18	0.12 ± 0.20	0.15 ± 0.15	0.12 ± 0.08
Ashfall	-0.05 ± 0.01	0.07 ± 0.12	-	0.18 ± 0.13
Δ	-0.20 ± .18	-0.05 ± .23	-	0.06 ± 0.15
		LiF TLD'S (Shielded-Disk)		
Non-Ashfall	-	-	0.08 ± 0.09	0.07 ± 0.13
Ashfall	-	-	-	0.15 ± 0.08
Δ	-	-	-	0.08 ± 0.15

The August 7, 1980 eruption had an average vial TLD reading for the ashfall areas of 0.07 ± 0.12 nanocoulombs. The average vial TLD reading for the non-ashfall areas was 0.12 ± 0.20 nanocoulombs. Within uncertainty estimates, these two numbers are the same. When the non-ashfall average is subtracted as an atmospheric blank (difference is 0.05 ± 0.23), no positive response is noted.

The October 16-18 eruption had average vial TLD readings for the ashfall areas of 0.18 ± 0.13 nanocoulombs. The non-ashfall average response was 0.12 ± 0.08 nanocoulombs. The difference between these values was 0.06 ± 0.15 nanocoulombs. A positive response was obtained with an uncertainty of 250%. Disk dosimeters were also exposed during this period. The average ashfall response of the disk TLD's was 0.15 ± 0.08 nanocoulombs. The average non-ashfall response was 0.07 ± 0.13 nanocoulombs. The difference was 0.08 ± 0.15 nanocoulombs. This indicated a positive response (corroborating the vial TLD data) with a 190% uncertainty associated with the Δ value. Thus a positive response was obtained, but the associated error is extremely high and, as determined using the t test, not significant.

From the ashfall and non-ashfall results, a maximum possible activity due to the measurable radon daughters may be calculated. The maximum response for the vial TLD's would be 190 ± 490 pCi/g and for the disk TLD's 390 ± 280 pCi/g. These values may be compared to activities generated from the uranium content of natural soils (105 ppm) which have an activity of 0.3-2 pCi/g. Assuming the uranium in the soil is in equilibrium with all its daughters, the total $\beta + \gamma$ activity is 2-12 pCi/g.

The concentrations of radon progeny measured in this study are below those levels reported by Fruchter et al. for the May 18 eruption. If the concentrations of the radon progeny had been equal to, or greater than, the reported concentrations, positive TLD reader response would have been recorded by the TLD devices. Thus, there appears to be no significant dose to the Pacific Northwest population from the Mount St. Helens eruptions studied in this report.

CONCLUSIONS

The Pacific Northwest Laboratory established a network of twelve sites around Mount St. Helens to monitor ashfall for its radioactivity. Integrated dose rates were measured at the individual sites using thermoluminescent dosimeters. The doses were recorded as the amount of charge generated by the photomultiplier tube readout of the light emitted from the TLD chips.

The radiation levels measured in the ashfall from the July 22 and August 7 eruptions were no higher than background levels measured at non-ashfall sites. Measurable radon daughters were observed at the ashfall sites of Portland, Cougar, and Trout Lake following the October 16-18 eruption. The average dose reading from the ashfall areas using the vial TLD's was 0.18 ± 0.13 nanocoulombs. The non-ashfall site data ranged from -0.01 to 0.23, the average was 0.12 ± 0.08 nanocoulombs. The difference between the ashfall and non-ashfall sites, corrected for atmospheric background, is 0.06 ± 0.15 nanocoulombs -- a positive, but not statistically significant,exposure.

The t test for significance between two sample means was applied to the data. It indicated four exposure values were significant. However, the significance can be discounted in each case. Hence, there was no significant exposure to the Pacific Northwest population as determined by this data.

The data generated in this program are in agreement with data published by Leifer et al.[11] He used the Department of Energy's high altitude WB-57F aircraft to sample the Mount St. Helens plume on May 20 and 21, 1980. Plume samples were obtained from both the troposphere and the stratosphere. The measurements of radioactivity in the plume were essentially the same as those measured outside the plume.

In summary, the radioactivity measurements from the eruptions of July 22, August 7, and October 16-18 showed that no statistically significant quantities of measurable radon daughters were observed. Consequently, there was no increase in radioactivity exposure to the Pacific Northwest as a result of Mount St. Helens.

REFERENCES

1. J. S. Fruchter, D. E. Robertson, J. C. Evans, K. B. Olsen, E. A. Lepel, J. C. Laul, K. H. Abel, R. W. Sanders. P. O. Jackson, N. A. Wogman, R. W. Perkins, H. H. Van Tuyl, R. H. Beauchamp, J. W. Shade, J. L. Daniel, R. L. Erickson, G. A. Sehmel, R. N. Lee, A. V. Robinson, W. R. Moss, J. K. Briant, W. C. Cannon, and L. P. Diediker, "Mount St. Helens Ash, May 18, 1980 Eruption: Chemical, Physical, Mineralogical, and Biological Properties," Science, 209:1116, September 5, 1980.

2. The Harshaw Chemical Company, Crystal and Electronic Products Department, Solon, OH.

3. V. W. Thomas, K. K. Nielsen, N. A. Wogman, M. L. Mauch, Quarterly Reports to the NRC (RES/SAFER), TD-1045, FIN B-2279, April-June, July-September, October-December, 1979 and January-March, 1980.

4. N. A. Wogman, W. B. Silker, J. A. Glissmeyer, G. W. R. Endres, Method for Assessing Background Levels of Radiation and Radioactive Materials in the Environment Around Uranium Mills, NUREG/CR-1253, PNL-3256, April, 1980.

5. Sierra Instruments, Inc., P.O. Box 909, Carmel Valley, CA 93924.

6. Andersen Samplers, Inc., Atlanta, GA 30336.

7. N. A. Wogman, R. W. Perkins, and J. H. Kaye, "An All Sodium Iodide Anticoincidence Shielded Multidimensional Gamma-Ray Spectrometer for Low Activity Samples Nuclear Instruments and Methods, 74, pp. 197-212, 1969.

8. Wong Laboratories, Cincinnati, OH.

9. Hanford Personnel Dosimeter Supporting Studies FY 1981, PNL-3736, Pacific Northwest Laboratory, Richland, WA.

10. R. C. Weast, Handbook of Chemistry and Physics, 49th edition. The Chemical Rubber Company, 1968.

11. R. Leifer, K. G. Sommers, S. F. Guggenheim, and I. Fisenne, "Recent Developments in High Altitude Sampling: Mount St. Helens and Stratospheric Trace Gases," paper presented at the IEEE 1980 Nuclear Science Symposium, Orlando, Florida, November 5-7, 1980.

PNL-3858
UC-11

DISTRIBUTION

No. of
Copies

OFFSITE

27 DOE Technical Information Center

25 R.L. Douglas
Office of Radiation Programs
Environmental Protection Agency
P.O. Box 18416
Las Vegas, NV 89114

5 D.M. Rosenbaum
Office of Radiation Programs
Environmental Protection Agency
ANR-458
Washington, D.C. 20460

ONSITE

DOE Richland Operations Office

H.E. Ransom

24 Pacific Northwest Laboratory

F.N. Eichner
W.A. Glass
E.A. Lepel (10)
J.M. Nielsen
K.B. Olsen
R.W. Perkins
V.W. Thomas
N.A. Wogman
Technical Information (5)
Publishing Coordination (2)