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Y-12 NATIONAL SECURITY COMPLEX

MANAGED BY
BWXT Y-12, L.L.C.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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Comparison of Particulate Collection in Probes and on Filters at the U.S. DOE Oak Ridge Y-12 National Security Complex

J. M. Eaton
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June 2001

Prepared by the
Y-12 National Security Complex
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ABSTRACT

Major radionuclide emissions from the Department of Energy's Y-12 National Security Complex are nuclides of uranium which are emitted as a particulate. The radionuclide NESHAP regulation requires stack sampling to be conducted in accordance with ANSI Standard N13.1, 1969. Appendix B of this standard requires in every case where sampling delivery lines are used that an evaluation should be made of deposition in these lines. A number of Y-12 Complex stacks are fitted with continuous samplers which draw particulate laden air through a probe and across a sample filter. One approach to evaluate line loss as required by the ANSI standard is to establish a representative factor that is used for all subsequent sampling efforts. Another approach is to conduct a routine probe wash procedure on an ongoing basis to account for line losses. In 1991, Y-12 National Security Complex personnel began routine probe washes as part of their sample collection procedure. Since then, 50 - 80 stacks have been sampled on a near continuous basis and probe washes have been conducted quarterly. Particulate collection in probes versus particulate collection on filters is recorded as a probe factor and probe factor trends for a 10-year period are available.

INTRODUCTION

The Y-12 National Security Complex is located on the Department of Energy Oak Ridge Reservation immediately adjacent to the city of Oak Ridge, Tennessee. The Y-12 complex is a large industrial complex supporting 531 buildings or other facilities totaling approximately 7 million square feet of floor space. Current assignments in the Y-12 Complex include weapons dismantlement and storage, enriched uranium material warehousing and management, and nuclear weapons process technology and development support. Processing of both enriched and depleted uranium includes metal casting, forming, rolling, machining, and chemical recovery.

For a number of years continuous samplers have been operating on many of the Y-12 process stacks. The data generated from the continuous samplers is used to demonstrate compliance with the Environmental Protection Agency's Radiological National Emission Standards for Hazardous Air Pollutants (NESHAP). The radionuclide NESHAP regulation requires stack sampling to be conducted in accordance with ANSI Standard N13.1, 1969. Appendix B of this standard requires in every case where sampling delivery lines are used that an evaluation should be made of deposition in these lines. In 1987, Y-12 personnel conducted a probe wash study to determine the amount of particulate that entered the sampling probe but did not make it to the filter for collection. By 1991, Y-12 personnel began routine probe washes as part of their sample collection procedure. Since then, 50 - 80 stacks have been sampled on a near continuous basis and probe washes have been conducted quarterly. Particulate collection in probes versus particulate collection on filters is recorded as a probe factor.

The ultimate purpose of continuous stack sampling and other radiological data generation is to demonstrate compliance with the radiological NESHAP dose standard. The standard requires that emissions of radionuclides shall not exceed those amounts that would cause any member of the public to receive in any year an effective dose equivalent of 10 millirem per year. The actual annual dose contributed by the Oak Ridge Reservation has never exceeded 20% of the standard and is normally less than 10% of the standard.

METHODOLOGY

The sampling systems employed at the Y-12 Complex consist of probes fitted with multiple nozzles and a vacuum pump which pulls air through the probe and across a filter paper. A probe configuration using multiple nozzles is commonly referred to as a probe rake. Sampling procedures are based on EPA test method 40 CFR 60, Appendix A, Method 5, entitled Determination of Particulate Emissions from Stationary Sources. The sampling that is conducted is termed near-isokinetic. Velocity profiles of stack flow are conducted once per quarter and nozzle sizes are selected to maintain isokinetic conditions at a constant sampler flow rate of 1.0 cubic feet per minute (cfm). The sampling systems are calibrated on a quarterly basis. Repair action is taken whenever sampler flow is outside the acceptable range established at 1.0 cfm plus or minus 10 percent.

Filter papers are collected for analysis 1-3 times per week. Collection frequency is dependent on stack emission characteristics. Probes are changed out once per quarter. Probes removed from the stack are taken to a laboratory where the probe is washed and the sample is collected using Method 5 procedures. Notable variations from the Method 5 procedure include the use of nitric acid as the solvent and the absence of physically brushing the interior of the probe. The probe interiors are rinsed instead of brushed because the configuration of the probes do not allow access for brushing. Nitric acid is used because uranium is highly soluble in nitric acid. Currently, the probe wash technique consists of a triple nitric acid rinse, a double water rinse, a single Formula 409® (degreaser) rinse, and a single water rinse. To reduce the possibility of residual uranium remaining in the probe due to oily conditions, the Formula 409® (degreaser) rinse step was added to the sample collection procedure in July 1999. Also, during the 1990's the probe wash procedure was initially revised from a triple acid rinse to a single acid rinse and then later was revised to reinstate the triple acid rinse. This latest revision was made in the fourth quarter of calendar year 1998. All of the liquid and solids from the multiple rinses of a single probe are collected in one container for a single analysis.

For each individual stack, the data results from the multiple filters collected during each calendar quarter are totaled. This quarterly filter result is compared to the results of the single quarterly probe wash data point and a quarterly probe factor is calculated for each individual stack.

Probe factors are calculated by adding the filter paper result to the probe wash result and then dividing that total by the filter paper result. For example, a probe factor of 5.0 indicates the total sample collected consists of 1 part from the filter and 4 parts from the probe; i.e. filter result multiplied by the probe factor equals the total.

One notable exception to the sampling frequency discussed above is for Stack 11 which exhausts a foundry operation at the Y-12 Complex. In the past four years, the emissions from Stack 11 have been studied thoroughly by a team of individuals to understand cause and effects on stack emissions and the stack sampling probes have received considerable attention. Probes are collected and washed on a daily basis during material processing campaigns. All aspects of the sampling campaigns, including operational variables, analytical methods, and sampling methodologies, have been carefully monitored by experts in statistical analysis. Probe factor data results from Stack 11 are comparable to the results noted at other Y-12 Complex stacks.

RESULTS

Results of the depleted uranium process stacks and the enriched uranium process stacks are presented separately for the years 1991 - 2000. The results are presented this way because this data breakdown is a routine part of the annual radionuclide NESHAP reporting methodology and therefore is readily available. For the initial probe wash study conducted in 1987 the data for enriched and depleted processes is combined.

The results of the 1987 probe wash study are presented in Table 1. Over 50 percent of the stacks yielded probe factors less than 2, i.e. more material was recovered from the filter sample than the probe samples. Also, less than 10 percent of the stacks yielded probe factors greater than 5. It should be noted, however, that the probe wash methodology was different in 1987 than in subsequent reporting years. In particular, the solvent used in 1987 was acetone rather than the much more aggressive uranium solvent, nitric acid.

The results for1991, the first full year of probe washing all stacks on a quarterly basis, are summarized in Table 2. Eighty-one stacks were included in the stack sampling program in 1991. The results show approximately twenty-five percent of the stacks had annual probe factors less than 2, forty-five percent of the stacks had annual probe factors in the 2-5 range, and thirty percent of the stacks had annual probe factors greater than 6. In 1991, the annual mean probe factor for enriched uranium sources was 7.2 and the annual mean probe factor for depleted uranium sources was 17.1.

The results for calendar year 2000 are summarized in Table 3. Forty-eight stacks were included in the stack sampling program in 2000. The results show approximately twenty-three percent of the stacks had annual probe factors less than 2, twenty-five percent of the stacks had annual probe factors in the 2-5 range, and fifty-two percent of the stacks had annual probe factors greater than 6. In 2000, the annual mean probe factor for enriched uranium sources was 9.9 and the annual mean probe factor for depleted uranium sources was 13.1.

Data results for individual stacks indicate there is considerable variability in probe factors from quarter to quarter. Graphs of quarterly probe factor results from 1991-2000 are presented in Figures 4-7 for a few individual stacks. Stack 11, which has been studied thoroughly by a team of individuals to understand cause and effects on stack emissions, also yields probe factors worthy of note. Probe washes on this stack have been conducted daily during process campaigns since 1997. The Stack 11 probe factors for 1997-2000 are 12.5, 17.7, 7.8, 11.0 respectively.

CONCLUSIONS

In approximately 75% of the Y-12 continuously sampled stacks, the amount of material collected in the probe is greater than the amount of material collected on the sample filter. A review of the data indicates there is no single probe factor which accurately reflects the probe retention for the 50+ stacks that are currently in operation at the Y-12 Complex. Also, probe factors within the same stack do not remain constant from sampling period to sampling period and the annual average probe factors do not remain constant over time. For Y-12 stacks, the most accurate way to estimate annual radiological stack emissions is to include a routine probe wash program as part of the sampling procedure. To further understand the variability of probe retention factors there are a number of areas that could be studied. These variables include: 1) enriched uranium processes versus depleted uranium processes, 2) process types, e.g. machining versus chemical processes, 3) effects of oil and/or moisture in the exhaust stream, 4) exhaust systems with and without emission control devices, and 5) probe wash techniques.

References

1. Code of Federal Regulations, 40 CFR Part 61, Subpart H - National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities

2. ANSI N13.1-1969, American National Standard Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities

3. Code of Federal Regulations, 40 CFR Part 60, Appendix A, Method 5 - Determination of Particulate Emissions from Stationary Sources

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Table 1
1987 Probe Study - 81 Stacks
Probe Factor Ranges vs. Number of Stacks in Range

Probe Factor	#Stacks
<2	47
2-5	27
6-10	5
11-20	2
	81

Table 2
1991 Annual Probe Factor Results - 81 Stacks
Probe Factor Ranges vs. Number of Stacks Range

Annual Factor	#Stacks
<2	21
2-5	35
6-10	14
11-20	7
21-50	0
>50	4
	81

Annual Probe Factors - Enriched U Sources
Range = 1.0 - 85.8 Mean = 7.2

Annual Probe Factors - Depleted U Sources
Range = 1.0 - 208.0 Mean = 17.1

Table 3
2000 Annual Probe Factor Results - 48 Stacks
Probe Factor Ranges vs. Number of Stacks in Range

Annual Factor	#Stacks
<2	11
2-5	12
6-10	11
11-20	8
21-50	4
>50	2
	48

Annual Probe Factors - Enriched U Sources
Range = 1.2 - 55.6 Mean = 9.9

Annual Probe Factors - Depleted U Sources
Range = 1.1 - 123.8 Mean = 13.1

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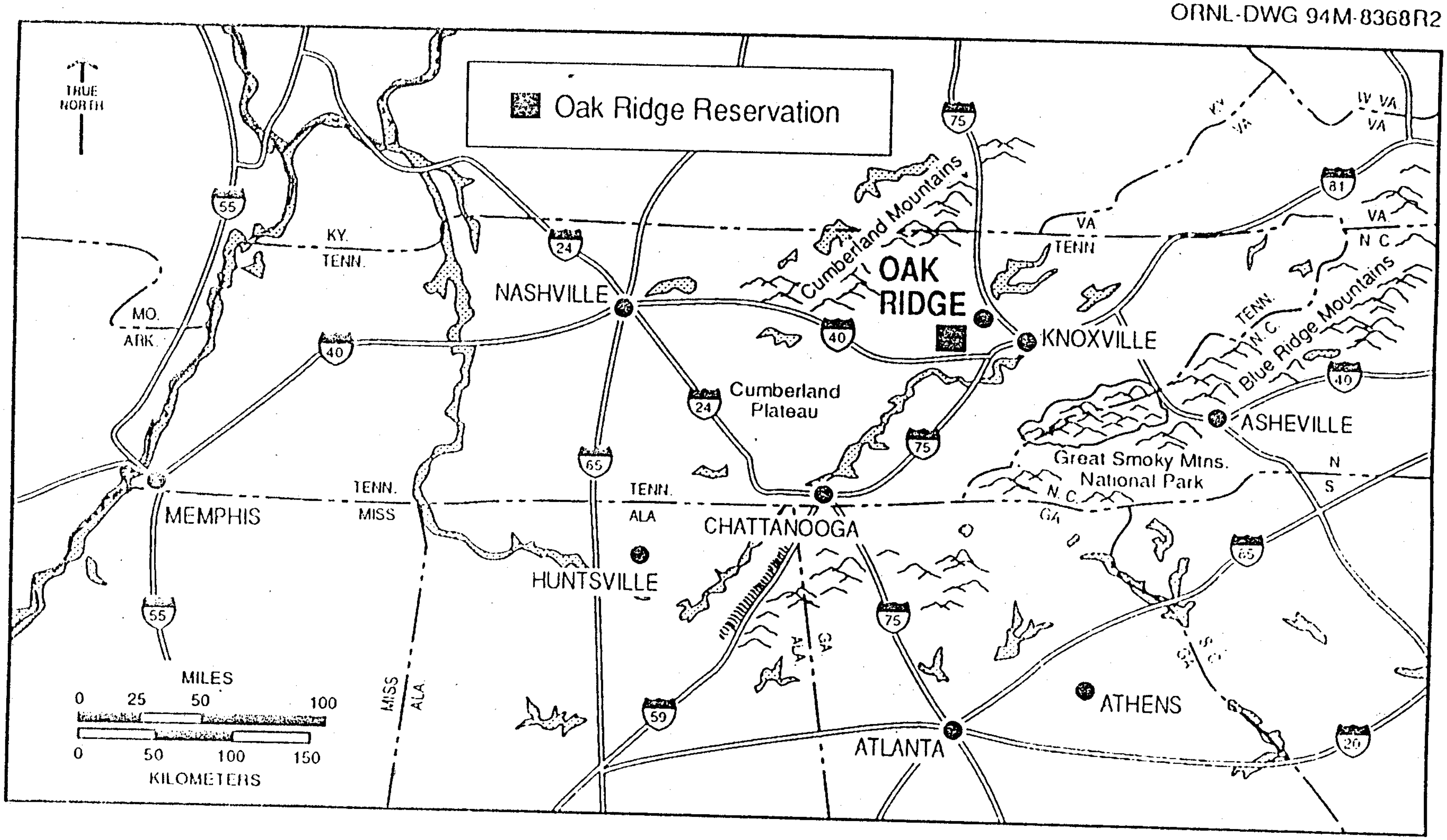

Figure 1 -
Location Map of
Oak Ridge Reservation

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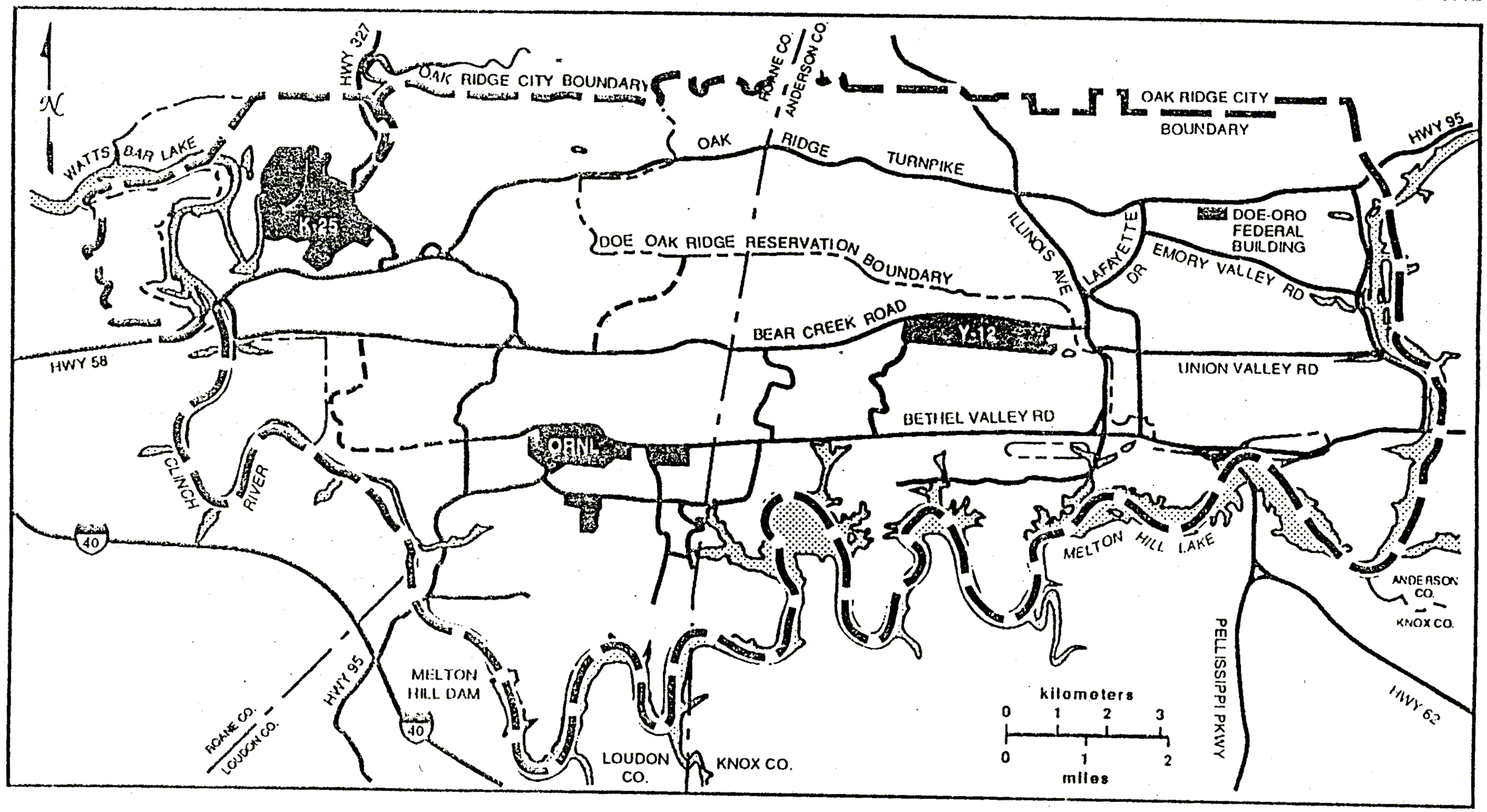
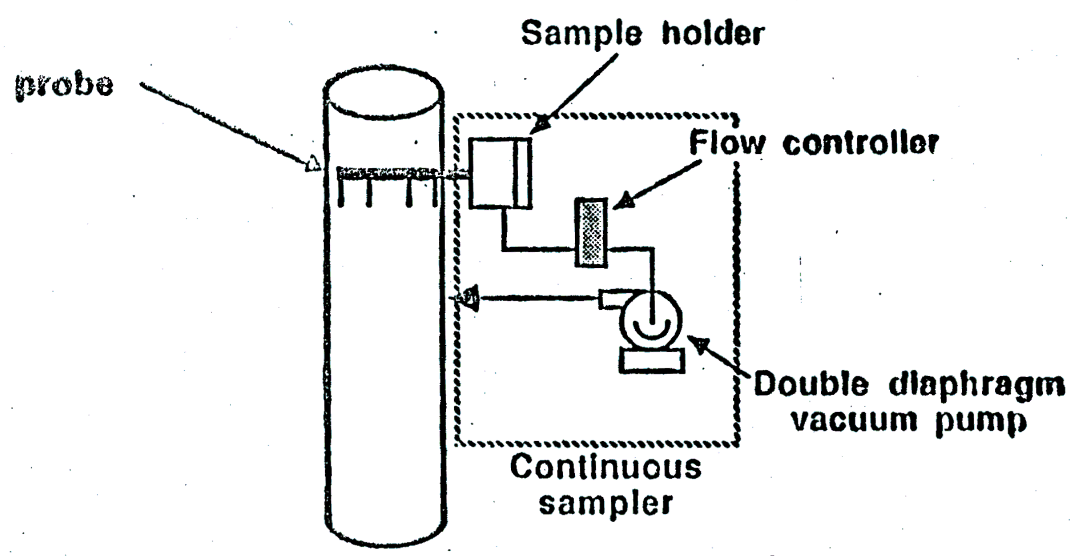


Figure 2 -
Location Map of Y-12 National
Security Complex

System Design
Continuous Sampler



- Each probe is specially designed to simulate iso-kinetic conditions
- All samplers pull 1 scfm

Figure 3 -
Schematic of Continuous Sampler.
on Stack

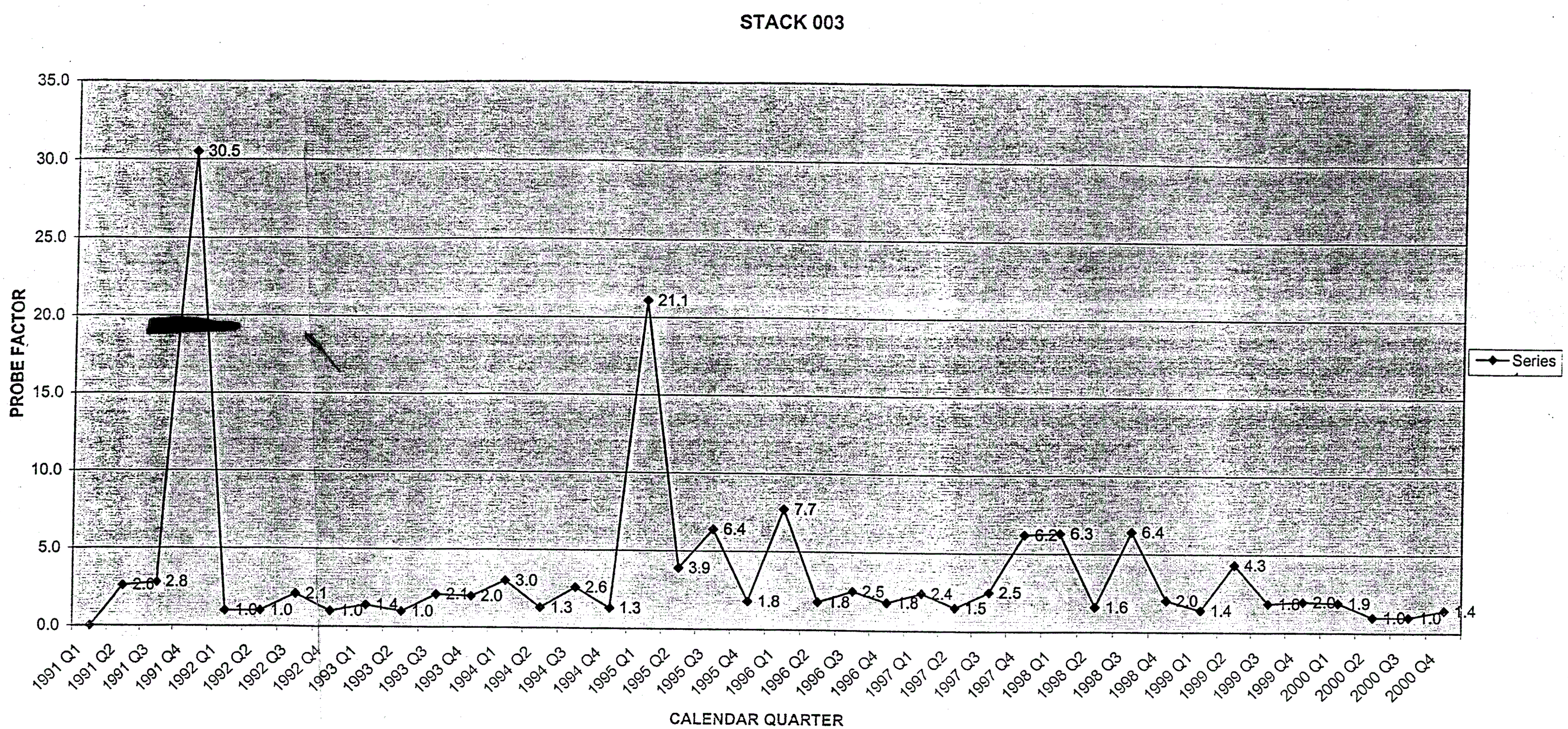


Figure 4 -
Stack 3 with HEPA filter control exhausts enriched uranium machine operations

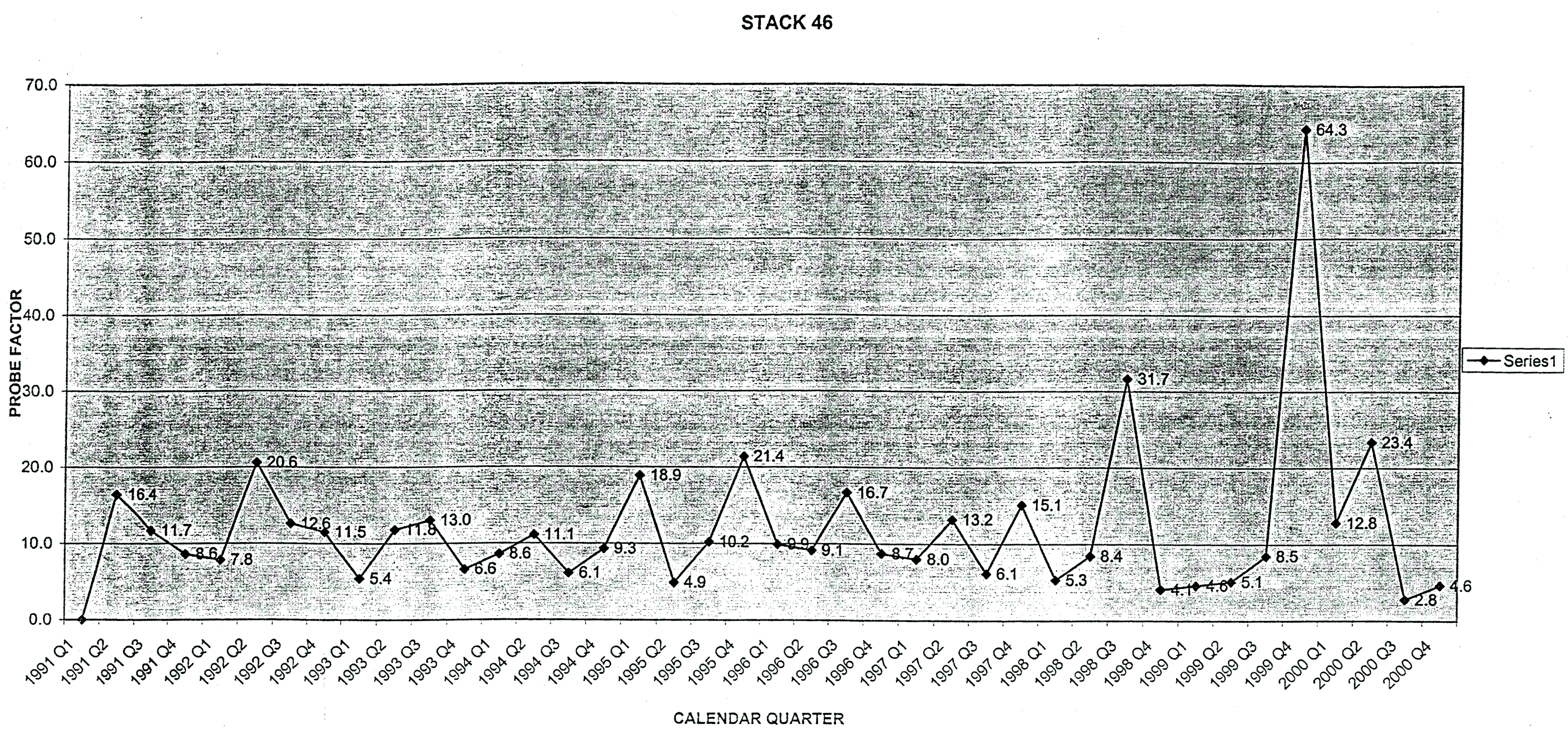


Figure 5 -
Stack 46 with no emission controls
exhausts enriched uranium
chemical recovery process

STACK 134

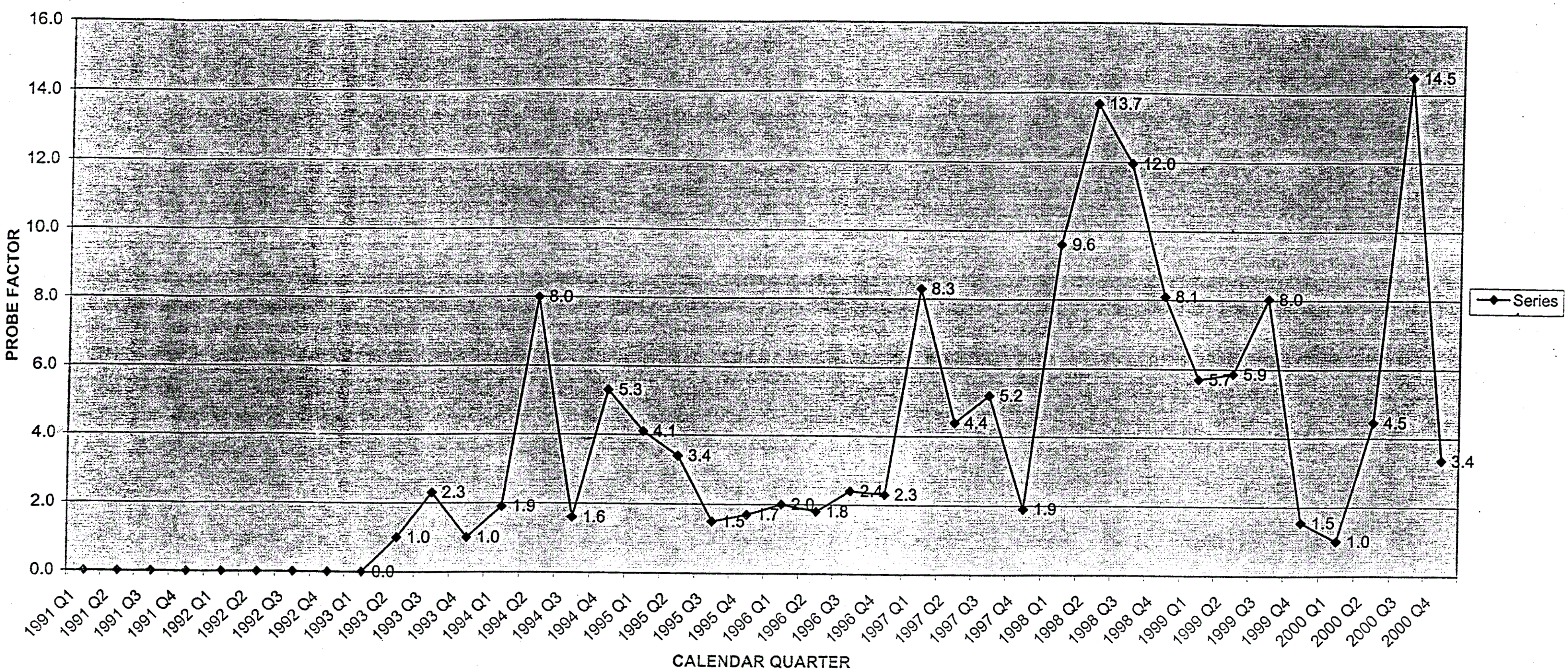


Figure 6 -
Stack 134 with HEPA filter and scrubber controls exhausts enriched uranium chemical and dry side recovery operations

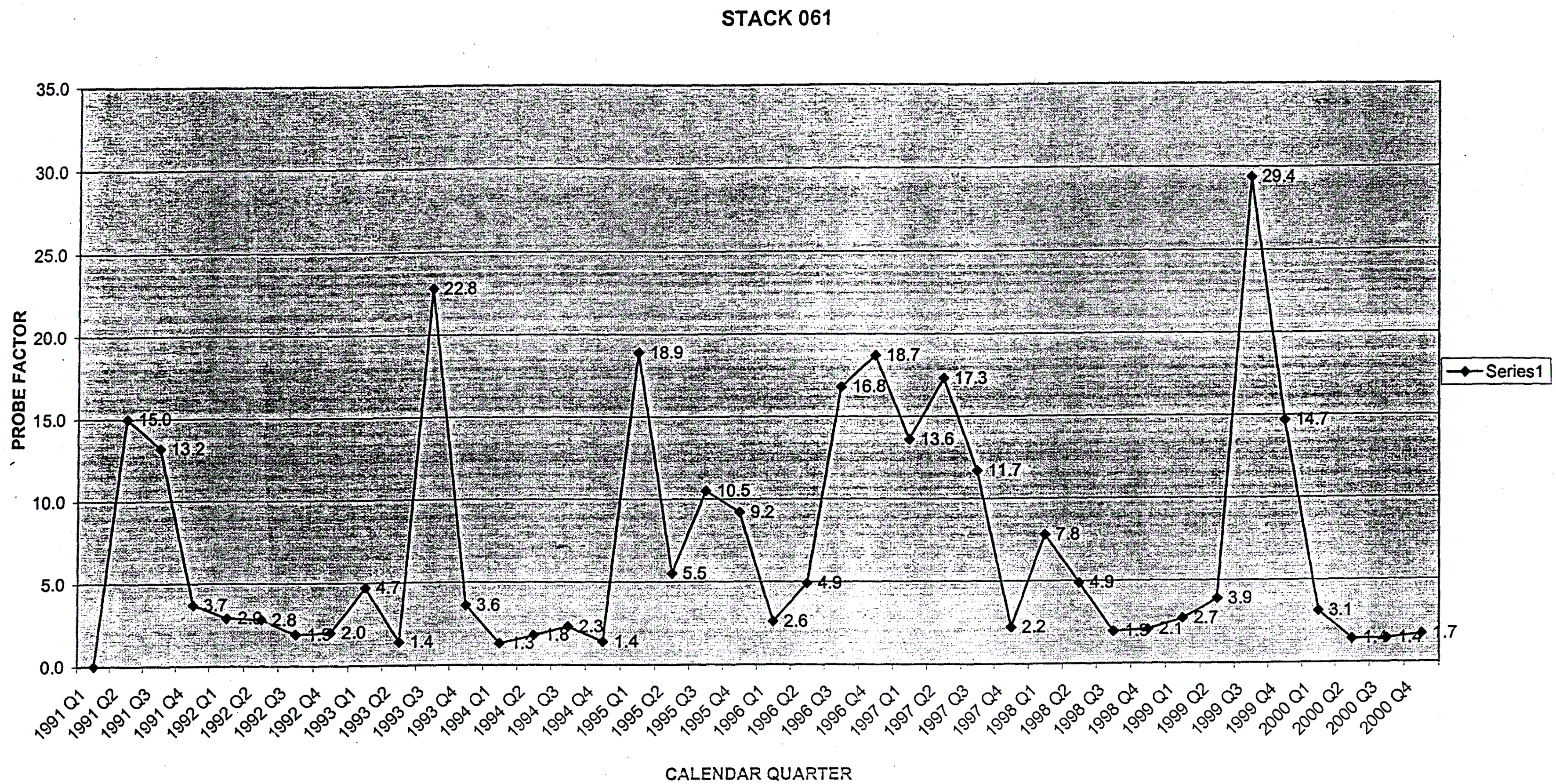


Figure 7 -
Stack 61 with filter control exhausts depleted uranium foundry operations