EML-599

HEAVY ELEMENT RADIONUCLIDES (Pu, Np, U) AND ^{137}Cs IN SOILS COLLECTED FROM THE IDAHO NATIONAL ENGINEERING AND ENVIRONMENTAL LABORATORY AND OTHER SITES IN IDAHO, MONTANA, AND WYOMING

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ABSTRACT

The isotopic composition of Pu in soils on and near the Idaho National Engineering and Environmental Laboratory (INEEL) has been determined in order to apportion the sources of the Pu into those derived from stratospheric fallout, regional fallout from the Nevada Test Site (NTS), and facilities on the INEEL site. Soils collected offsite in Idaho, Montana, and Wyoming were collected to further characterize NTS fallout in the region. In addition, measurements of ^{237}Np and ^{137}Cs were used to further identify the source of the Pu from airborne emissions at the Idaho Chemical Processing Plant (ICPP) or fugitive releases from the Subsurface Disposal Area (SDA) in the Radioactive Waste Management Complex (RWMC).

It is estimated that the non-fallout Pu in INEEL soils is approximately 5.7 GBq (~ 150 mCi) and that the majority of this site-derived Pu has its origin from releases of Pu at the SDA in the early and late 1960s. Stratospheric fallout on the INEEL is approximately one-half of that measured elsewhere in the 40-50°N latitude band such that the 5.7 GBq of Pu originating from the SDA represents an approximate 14 percent increase over that expected from stratospheric fallout.

There is convincing evidence from this study that ^{241}Am , in excess of that expected from weapons-grade Pu, constituted a part of the buried waste at the SDA that has subsequently been released to the environment.

Measurements of ^{236}U in waters from the Snake River Plain aquifer and a soil core near the ICPP suggest that this radionuclide may be a unique interrogator of airborne releases from the ICPP. Neptunium-237 and ^{238}Pu activities in INEEL soils suggest that airborne releases of Pu from the ICPP, over its operating history, may have recently been overestimated. Future ^{236}U measurements in INEEL soils would provide data on the geographical influence of ICPP emissions as well as confirmatory information concerning past releases of Pu from this facility.

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INTRODUCTION

Beginning in April, 1954, transuranic waste has been stored at the Radioactive Waste Management Complex (RWMC) at the Idaho National Engineering and Environmental Laboratory (INEEL). These wastes were generated, principally, at the Rocky Flats Environmental Technology Site , near Marshall, CO as part of the production process for nuclear device components. Between 1954 and 1970, ~ 65 x 10^3 m^3 of solid waste was buried in the Subsurface Disposal Area (SDA), an area within the RWMC; additional waste has been stored at the RWMC since 1970, but this material is in above-ground, retrievable storage in the Transuranic Storage Area (Markham et al.1978). The activity of the buried transuranic waste for $^{239+240}$Pu, ^{241}Pu, ^{241}Am, and ^{238}Pu has been estimated at 0.78 PBq, 6.3 PBq, 1.8P Bq and 21 TBq, respectively (Markham et al. 1978).

The Department of Energy plans to demonstrate the retrievability and treatment of buried transuranic waste from one portion of the SDA (Pit 9). It was decided that prior to this demonstration project that the isotopic composition of Pu in INEEL soils should be determined. This decision was based on three principal considerations. They were: 1] fugitive releases of transuranic radionuclides from the RWMC that occurred in the early and late 1960s (the translocation of this contamination away from the RWMC had not been systematically investigated since 1978); 2] the isotopic composition of the Pu in the SDA is characteristic of "low-burn up" Pu , i.e., Pu whose ^{240}Pu/^{239}Pu atom ratio ($\leq$0.05) makes it easily distinguishable from that generated by global fallout (^{240}Pu/^{239}Pu atom ratio ~ 0.18,) if high-sensitivity, thermal ionization mass spectrometry (TIMS) techniques are used for the analysis, and; 3] in the unlikely event of a release of transuranic radionuclides during retrieval and treatment of the buried wastes, the most sensitive method of detecting significant areal contamination from such an event (especially to distances > 1-2 km) would be to track its isotopic Pu "footprint" in the surrounding soils. Such an approach requires that the existing isotopic Pu composition (and inventories) be known.

This report contains the results and interpretation of transuranic radionuclide (Pu, ^{237}Np, ^{241}Am) and ^{137}Cs measurements in 123 soil cores collected on the INEEL site and in selected areas of Idaho, Montana and Wyoming between 1993 and 1995. The principal focus was to map the existing isotopic Pu composition in INEEL soils and to determine the extent to which material released at the RWMC in the 1960s had been redistributed away from this facility. Consequently, the majority of the core samples (96) were taken at, or near, the INEEL. The remaining cores were collected at selected sites in Idaho, Montana and Wyoming to supplement previously published data on the trajectories and deposition of fallout from the Nevada Test Site (NTS). Finally, water from selected monitoring wells down-gradient from the Idaho Chemical Processing Plant (ICPP) were analyzed, again by thermal ionization mass spectrometry techniques, for uranium isotopes and ^{237}Np. This was done to confirm their presence in wastes discharged to the Snake River Plain aquifer and to assess the potential for their atmospheric release from the ICPP. The ^{237}Np/ ^{239}Pu atom ratio in global fallout and in NTS fallout has recently been reassessed (Beasley et al.1997) so that this ratio is a potentially sensitive interrogator of releases of both isotopes from sources other than fallout.

BACKGROUND

Transuranic radionuclides in soils at the INEEL have five principal sources. These are: 1] releases at the RWMC; 2] integrated global fallout from the testing of above-ground nuclear devices by the United States, the Former Soviet Union (FSU), England, France, and the People's Republic of China; 3] ^{238}Pu injected into the stratosphere by the burn-up of a failed radioactive thermal generator (RTG) in 1964 (Krey 1967); 4] regional fallout from above-ground testing at the NTS , and; 5] stack releases from the ICPP. Each of these sources have characteristic radionuclide signatures and/or abundances that can, in principle, be used to identify their presence in the soils and to estimate their concentrations. Consequently, each of these sources is reviewed below as they relate to assigning provenance to the transuranic radionuclides discussed in this report.

TRANSURANIC RELEASES FROM THE RWMC

Markham et al. (1978) presented data from soil measurements collected in 1974 that showed the presence of SDA-derived ^{238}Pu, $^{239+240}$Pu, and ^{241}Am beyond the perimeter of the RWMC. Flooding events in both 1962 and 1969 remobilized shallow buried transuranic waste which was then believed to be redistributed in subsequent years owing to the strong monthly peak wind gusts at the site (between 97 and 126 km hr^{-1}) and the semi-arid climate of the region (22 cm yr^{-1}precipitation). Figure 1 shows the location of the INEEL and RWMC in geographical prospective to the State of Idaho as well as the $^{239+240}$Pu activity isopleths (nCi m^{-2}) around the RWMC as measured in 1974. Integration of the isopleths shown gives a 1974 soil inventory, outside the RWMC, of between 60 and 70 mCi (2.2-2.6 GBq). It should be mentioned that the scale lengths shown in Markham et al. (1978) are incorrect and should range between 0 and 600 m rather than 0 and 400 m as indicated.

INTEGRATED GLOBAL FALLOUT

It must be emphasized that the global fallout inventories discussed below are expected to be found in "undisturbed" soils, i.e., soils where remobilization of the deposited radionuclides from human activities, overland runoff, and/or wind-forced resuspension of soil are minimal. As discussed above, such conditions cannot be expected to apply to the semi-arid landscape of the INEEL and its environs. Moreover, because fallout deposition has been shown to be strongly correlated with annual average rainfall (Toonkel 1977), inventories on and near the INEEL are expected to be lower than those recorded in wetter climates at the same latitude.

From soil core measurements made in 1970, Hardy et al. (1973) estimated the average areal inventory of fallout-derived $^{239+240}$Pu in the 40-50°N latitude band to be 81 ± 19 Bq m^{-2} with a pre-SNAP 9A ^{238}Pu/$^{239+240}$Pu activity ratio of 0.024 ± 0.007. Following the burn-up of the SNAP-9A ^{238}Pu thermal generator in the southern hemisphere (Krey 1967), ^{238}Pu/$^{239+240}$Pu activity

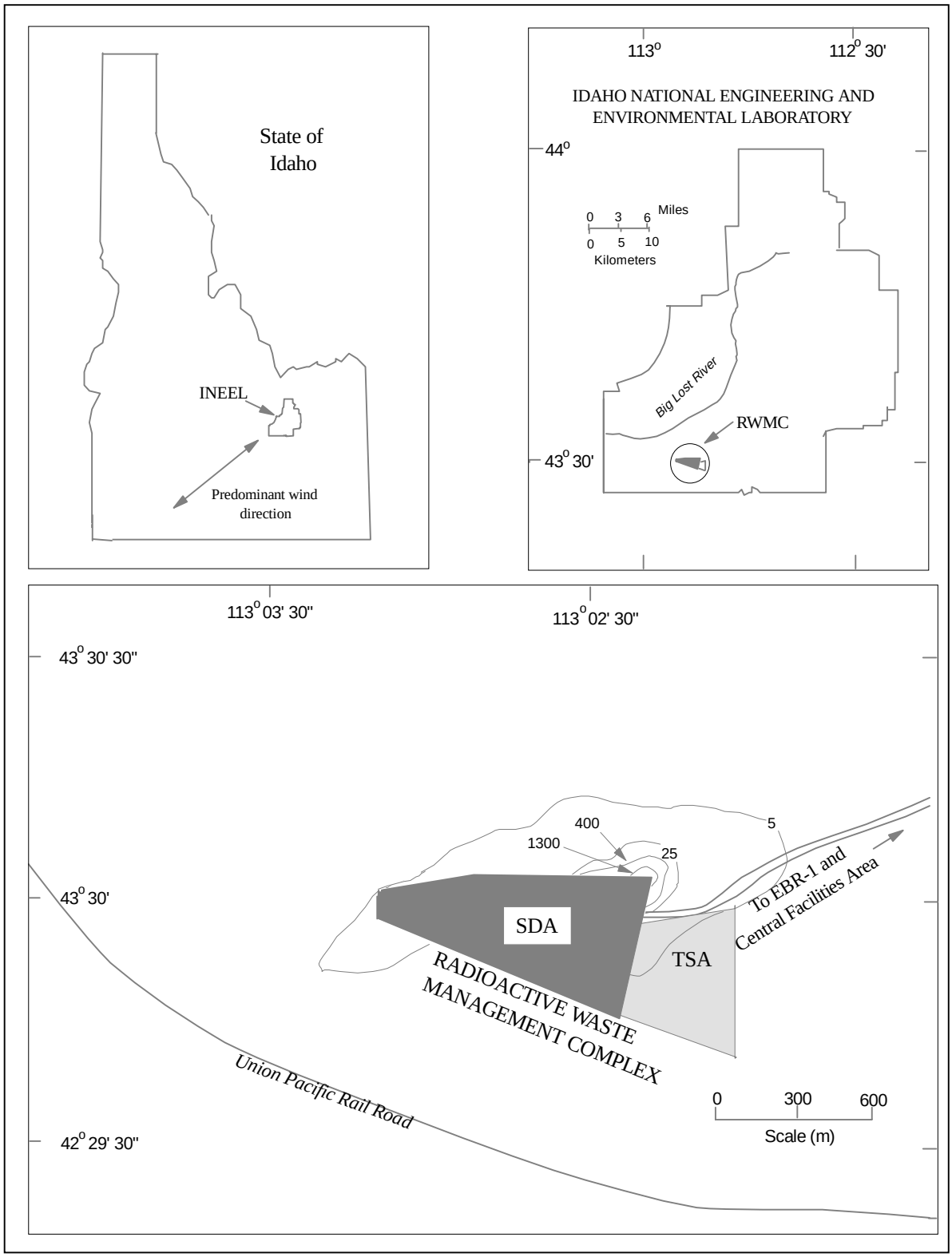

Figure 1. INEEL showing the principal areal extent to which $^{239+240}Pu$ moved beyond the perimeter at the RWMC by 1974. Some radioactivity was believed to have reached 2.5 km NE and 2.0 km SE of the RWMC. SDA (Subsurface Disposal Area). TSA (Transuranic Storage Area; above ground). Adapted from Pittman (1989) and Markham et al. (1978). Isopleth units: nCi m^{-2} .

ratio at this latitude increased to 0.036 ± 0.011. Therefore, in the intervening 24-year period (1970-1994), the expected, decay-corrected $^{238}Pu/^{239+240}Pu$ activity ratio at the INEEL is calculated to be 0.030 ± 0.009 or ~ 0.03 ± 0.01($T_{½}$ ^{238}Pu = 87.74 yr; Lederer and Shirley 1978). However, other Pu measurements at mid-latitudes in the northern hemisphere during the late 1970s showed $^{238}Pu/^{239+240}Pu$ activity ratios increasing to between 0.05 and 0.06 from the SNAP-9A event (Beasley et al.1986; Krey and Beck 1981) which, when decay corrected to 1994, would give $^{238}Pu/^{239+240}Pu$ activity ratios between 0.04 and 0.05. For the purposes of this report, we will consider these latter values as representative of integrated global fallout in the mid-1990s.

The $^{240}Pu/^{239}Pu$ atom ratio in northern hemisphere fallout is ~ 0.18 (Krey et al. 1976) and the associated $^{237}Np/^{239+240}Pu$ activity ratio has recently been estimated at 0.0031 ± 0.0003 (Beasley et al.1997). The corresponding $^{237}Np/^{239}Pu$ *atom ratio* is northern hemisphere fallout is 0.45 and therefore the areal inventory of global fallout-derived ^{237}Np at the INEEL is calculated to be 0.25 ±0.06 Bq m^{-2}. The $^{137}Cs/^{239+240}Pu$ activity ratio in northern hemisphere fallout was 53 ± 2 in 1979 (Beck and Krey 1983). This same ratio, decay-corrected to 1994 ($T_{½}$ ^{137}Cs = 30.17 yr; Lederer and Shirley 1978), is 37.6± 1.4 or ~ 38 ± 2 giving a predicted ^{137}Cs global fallout inventory of 3078 ± 740 Bq m^{-2} at the INEEL. Finally, the present-day (1998) $^{241}Am/^{239+240}Pu$ activity ratio at mid-latitudes is ~ 0.4 (0.35 ± 0.02; Yamamoto et al.1990).

NTS FALLOUT

Regional fallout from testing at the NTS is characterized by significantly lower $^{240}Pu/^{239}Pu$ and $^{237}Np/^{239}Pu$ atom ratios than those present in integrated global fallout because of the size of the nuclear devices tested there (≤ 100 kilotons). The neutron fluxes produced by these devices were much less than those of the megaton devices that generated the majority of global fallout such that successive neutron capture to produce higher mass number transuranic radionuclides was greatly reduced in the NTS tests. For example, Table 1 lists the nuclear tests carried out at NTS, their yields, and the $^{240}Pu/^{239}Pu$ atom ratios (30 days post shot) measured in their fallout debris (Hicks and Barr 1984). It is important to note (Figure 2) that there is no correlation between the yield of the device and the resultant measured $^{240}Pu/^{239}Pu$ atom ratios. Moreover, the $^{237}Np/^{239}Pu$ atom ratio in NTS debris, while less well characterized than that in global fallout, is most likely ≤0.03 (Beasley et al.1997). Thus, as a first order approximation, it can be assumed that the $^{240}Pu/^{239}Pu$ atom ratio in fallout from early NTS tests was very near that of the weapons-grade Pu produced in the production reactors at the Hanford Complex (Washington State) from the early-to-middle 1950s. This assumption is supported by recent measurements of isotopic transuranic signatures in soils contaminated by the waste storage tank explosion which occurred at the Mayak Complex in Chelyabinsk Province, FSU, in late 1957 (Beasley et al.1998). Greater than 98 percent of the Pu in the soil came from this contamination event and its $^{240}Pu/^{239}Pu$ atom ratio was 0.0283, a value not unlike those seen for early NTS tests (Table 1). Given that the production reactors of both countries in that period were graphite-moderated, water-cooled reactors, it is not unreasonable to expect that to produce weapons-grade material of low ^{240}Pu content, fuel irradiation cycles in both countries were comparable.

With time, experience showed that greater amounts of ^{240}Pu in weapons-grade Pu did not compromise explosive yield and by the late 1950s, $^{240}Pu/^{239}Pu$ atom ratios as high as 0.07 - 0.08 were observed in fallout debris from several devices tested at NTS (Table 1: Figure 2). Thus, the

TABLE 1

NUCLEAR DEVICES TESTED AT THE NEVADA TEST SITE, APPROXIMATE YIELD, AND THE $^{240}Pu/^{239}Pu$ ATOM RATIOS MEASURED P0ST SHOT (30 DAYS)

OPERATION	Date	Kilotons	$^{240}Pu/^{239}Pu$	OPERATION	Date	Kilotons	$^{240}Pu/^{239}Pu$
BUSTER- JANGLE				**UPSHOT-KNOTHOLE**			
Baker	10/28/51	3.5	0.0326	Simon	04/25/53	43	0.0267
Charlie	10/30/51	14	0.0283	Encore	05/08/53	27	0.0517
Dog	11/01/51	21	0.0282	Harry	05/19/53	32	0.0375
Easy	11/05/51	31	0.0355	Climax	06/04/53	61	0.0342
TUMBLER-SNAPPER				**OPERATION TEAPOT**			
Charlie	04/22/52	31	0.0506	Wasp	02/18/55	1	0.0533
Dog	05/01/52	19	0.0346	Moth	02/22/55	2	0.0778
Easy	05/07/52	12	0.0236	Turk	03/07/55	43	0.0326
Fox	05/25/52	11	0.0236	Hornet	03/12/55	4	0.0577
George	06/01/52	15	0.0257	Bee	03/22/55	8	0.0853
How	06/05/52	14	0.0271	Apple	03/29/55	14	0.0245
UPSHOT- KNOTHOLE				Wasp Prime	02/29/55	3	0.0522
Anne	03/17/53	16	0.0246	HA	04/06/55	3	0.0510
Nancy	03/24/53	24	0.0283	Post	04/09/55	2	0.0194
Dixie	04/06/53	11	0.0217	Met	04/15/55	22	0.0068
Badger	04/18/53	23	0.0342	Apple II	05/05/55	29	0.0313

TABLE 1 (cont'd)

NUCLEAR DEVICES TESTED AT THE NEVADA TEST SITE, APPROXIMATE YIELD, AND THE $^{240}Pu/^{239}Pu$ ATOM RATIOS MEASURED P0ST SHOT (30 DAYS)

OPERATION	Date	Kilotons	$^{240}Pu/^{239}Pu$	OPERATION	Date	Kilotons	$^{240}Pu/^{239}Pu$
OPERATION-TEAPOT				**OPERATION-PLUMBOB**			
Zuchinni	05/15/55	29	0.0319	Wheeler	09/06/57	0.2	0.0376
OPERATION-PLUMBOB				Fizeau	09/14/57	11	0.0630
Boltzmann	05/28/57	12	0.0787	Newton	09/16/57	12	0.0716
Wilson	06/18/57	10	0.0818	Whitney	09/23/57	19	0.0734
Priscilla	06/24/57	37	0.0108	Charleston	09/28/57	12	0.0735
Hood	07/05/57	74	0.0673	Morgan	10/07/57	8	0.0773
Diablo	07/15/57	17	0.0624	**HARDTACK-II**			
John	07/19/57	~ 2	0.0591	Eddy	09/19/58	0.083	0.0503
Kepler	07/24/57	10	0.0722	Mora	09/29/58	2	0.0544
Owens	07/25/57	~ 10	0.0702	Lea	10/13/58	1.4	0.0542
Stokes	08/07/57	19	0.0074	Socorro	10/22/58	6	0.0568
Doppler	08/23/57	11	0.0700	De Baca	10/26/58	2.2	0.0637
Franklin Prime	08/30/57	~ 5	0.0029	Santa Fe	10/30/58	1.3	0.0538
Smoky	08/31/57	44	0.0058				
Galileo	09/02/57	11	0.0753				

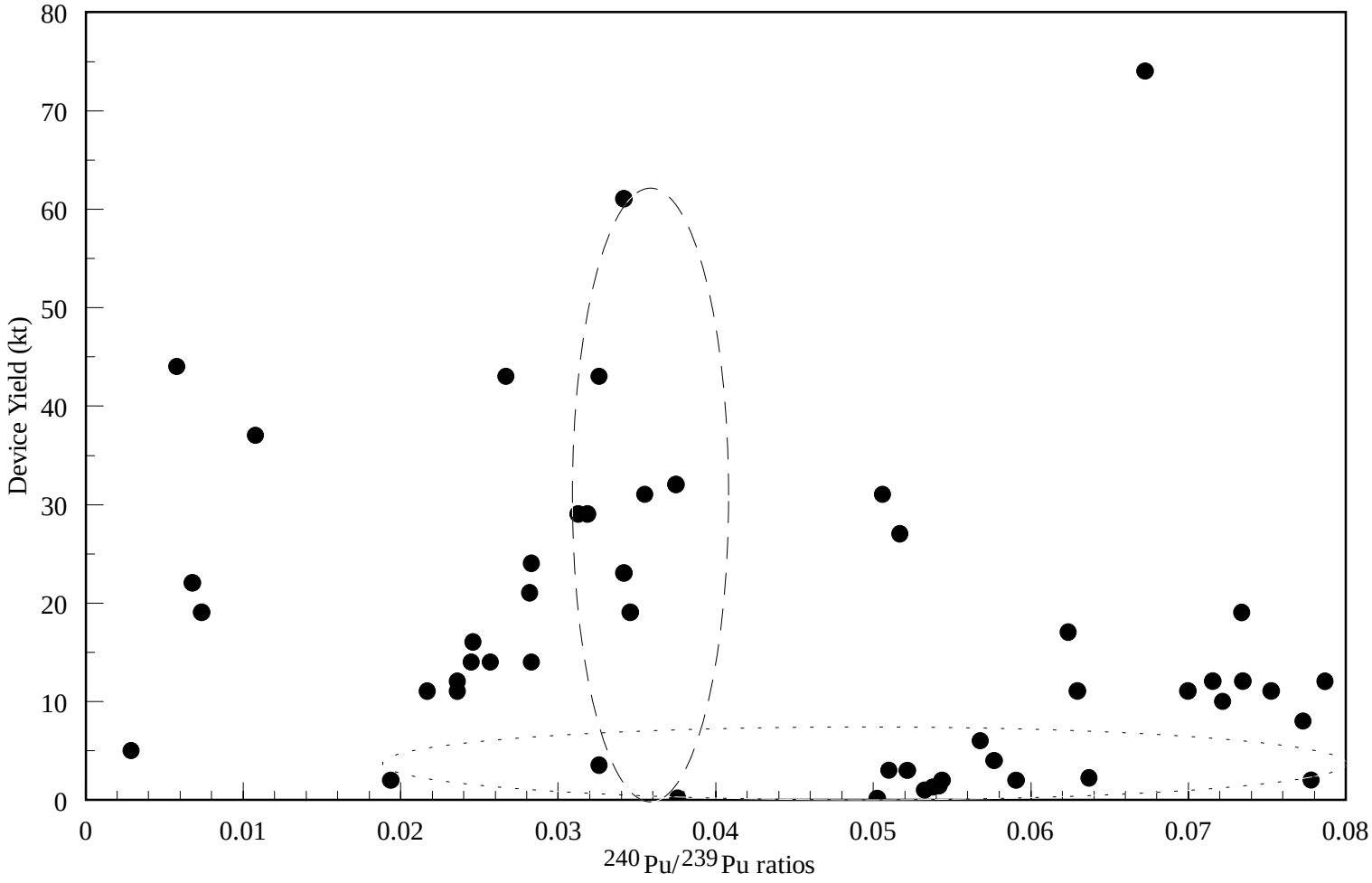

Figure 2. Nuclear device yield and the $^{240}Pu/^{239}Pu$ atom ratio measured in NTS fallout debris 30 days postshot. For very low yield tests, 2-6 kt (·····), atom ratios ranged between ~ 0 .02 and ~ 0.08. For some tests whose yields ranged between 4 and 61 kt (– – –), atom ratios ranged between ~ 0.032 and ~0.038.

range of $^{240}Pu/^{239}Pu$ atom ratios in material buried at the SDA, through 1969, conceivably spans values between 0.03 and 0.08. Therefore, it is not possible, *a priori*, to assign a single $^{240}Pu/^{239}Pu$ atom ratio to the material released from the SDA in the flooding events of 1962 and 1969.

As discussed by Beck and Anspaugh (1991), although the $^{137}Cs/^{239+240}Pu$ activity ratios from global fallout in the northern hemisphere are well correlated, the same cannot be said for regional fallout from the NTS. First, because of the yields of the devices tested (Table 1), $^{137}Cs/^{239+240}Pu$ activity ratios differed widely in time, especially in those periods when uranium-fueled devices produced significant quantities of ^{137}Cs and virtually no Pu. Secondly, the trajectories of fallout from the NTS varied such that no regional locality was uniformly contaminated by all tests conducted there. In general, however, the $^{137}Cs/^{239+240}Pu$ activity ratios from NTS fallout debris were less than that observed from global fallout as would be expected from the testing of low-yield, Pu-fueled devices (Beck and Anspaugh 1991).

PLUTONIUM AND NEPTUNIUM EMISSIONS FROM THE ICPP

As discussed by Benedict et al. (1981), the ICPP was designed as a multipurpose facility to recover highly-enriched uranium ($\leq$ 100 percent) from different fuel types used in naval propulsion, research and test reactors. Indeed, the great majority of reactors built and operated at the INEEL have been fueled with ~ 93 percent ^{235}U (Nucleonics 1960). Plutonium-239 is produced by n,γ reactions on ^{238}U to produce ^{239}U which then decays by β-particle emission to form ^{239}Pu. Therefore production of ^{239}Pu (and other Pu isotopes) in reactors fueled with > 90 percent ^{235}U is much less than in those fueled with natural uranium or 3-5 percent enriched ^{235}U as is their isotopic composition. Characteristic of highly-enriched fuels is the enhanced production of other transuranic radionuclides such as ^{236}U, ^{237}Np, and ^{238}Pu through the following nuclear reactions and decays:

$$^{235}U\ (n,\gamma)\ ^{236}U;\ \ ^{236}U(n,\gamma)\ ^{237}U \qquad (1)$$

$$^{237}U \xrightarrow{\beta} {}^{237}Np\ (T_{½} = 6.5\ d) \qquad (2)$$

$$^{237}Np\ (n,\gamma)\ ^{238}Np \qquad (3)$$

$$^{238}Np \xrightarrow{\beta} {}^{238}Np\ (T_{½} = 2.12\ d) \qquad (4)$$

where the cross sections for neutron capture in equations (1) and 3) are ~ 100, ~ 5, and ~ 400 barns, respectively (Lederer and Shirley 1978).

In 1991 the Department of Energy published the Idaho National Engineering Laboratory Historical Dose Evaluation (DOE 1991). As discussed in this report, the earliest measurements of Pu stack releases from the ICPP began in 1974 and releases prior to this time were largely estimated by calculation. With the exception of several episodic releases of transuranics on the site, stack releases from the ICPP constitute the principal source of site-derived Pu to the INEEL.

ICPP stack monitoring data, from 1974 to the present, is contained in the INEEL Radioactive Waste Management Information System (RWMIS) data base. The mean $^{238}Pu/^{239+240}Pu$ activity ratio for ICPP releases between 1974 and 1986 was 6.8 (standard deviation = ± 3.5) confirming the neutron activation pathways of highly-enriched ^{235}U discussed above. There are also entries in the RWMIS for ^{237}Np releases at the ICPP in 1975 (annual), and 1979 (October and November) equaling 1.44 MBq (1.4×10^{20}atoms) in 1975 and 32.2 kBq (3.1×10^{18} atoms) in 1979. Moreover, for the years 1978, 1979, and 1986 there are data for both ^{239}Pu and ^{240}Pu releases from the ICPP, but it is not entirely clear whether these data represent actual mass spectrometric measurements or whether they are based on reactor burn up code calculations. For example, in October, 1979 an estimated 7.96 kBq of ^{239}Pu and 7.14 kBq of ^{240}Pu were released from the ICPP stack representing a $^{240}Pu/^{239}Pu$ atom ratio for this material of ~ 0.25 ($T_{½}$ $^{239}Pu = 2.4119 \times 10^4$ yr; $T_{½}$ $^{240}Pu = 6.564 \times 10^3$ yr) a value characteristic of high burn up fuel. In November, 1979 the ^{239}Pu and ^{240}Pu stack releases were reported as 41 kBq and 7.2 kBq, respectively, representing a $^{240}Pu/^{239}Pu$ ratio of 0.048. The corresponding $^{237}Np/^{239}Pu$ atom ratios for October and November are calculated to be 148 and 41, respectively, again showing the enhanced production of ^{237}Np from the irradiation of highly-enriched U fuels. For the entire year 1978, $^{240}Pu/^{239}Pu$ atom ratios calculated from the stack emission data were invariant at 0.048 suggesting that only fuels (or stored wastes) of identical irradiation history and composition were processed during that time. For much of 1979, the calculated $^{240}Pu/^{239}Pu$ atom ratios were invariant at 0.048, but there were notable exceptions in April, May and October. In April, $^{240}Pu/^{239}Pu$ atom ratios rose to 0.287, but declined to 0.054 in May. In October, the $^{240}Pu/^{239}Pu$ atom ratios were again elevated at 0.244, but declined to 0.048 by November. For 1986, the RWIMS contains the summed annual release of ^{239}Pu (44.4 kBq) and ^{240}Pu (10.8 kBq) giving a mean $^{240}Pu/^{239}Pu$ atom ratio of 0.066. The significance of these isotopic data, especially the $^{237}Np/^{239}Pu$ atom ratios mentioned above, are elaborated in greater detail in this report where the provenance of the Pu measured in INEEL soils is discussed.

The combined measured and reconstructed airborne $^{239+240}Pu$ release estimates from the ICPP through 1994, as reported in the Historical Dose Evaluation report, total to some 3.5 TBq (~ 94 mCi). However, the assumptions used in reconstructing the Pu releases for the period 1953-1973 were intended to provide conservative dose estimates to populations near the INEEL with the result that the actual $^{239+240}Pu$ releases were likely much smaller than this amount. For example, the majority of airborne radioactivity released from the ICPP occurred between 1957 and 1965. During those years, isotope-specific measurements were not made for ^{238}Pu or $^{239+240}Pu$; the recorded data for all ICPP stack releases from 1952 to 1961 specify the total quantity of radioactivity released (Ci) but not its isotopic composition (DOE 1991). Moreover, in those cases where stack effluent data are recorded as “total” or “unidentified” alpha, the dose reconstruction calculations ascribe this activity entirely to Pu that is then partitioned into ^{238}Pu and $^{239+240}Pu$ using the measured 1974-1986 stack effluent ratios cited above.

Given the uncertainties in the stack monitoring data and the assumption of a constant $^{238}Pu/^{239+240}Pu$ activity ratio in stack effluents over nearly 20 years of the ICPP’s operating history it is difficult to judge the extent to which calculated and actual releases might agree. Overall, however, it would not be unrealistic to assume that the actual releases were lower than those estimated to have occurred, perhaps by as much as a factor of 2 or more.

OTHER TRANSURANIC RELEASES AT THE INEEL

Besides stack emissions from the ICPP, there are only two other principal transuranic releases of operational origin known to have occurred at the INEEL. One involved the release of ~ 3.7 GBq (~ 100 mCi) of Pu at the ICPP as a result of burning contaminated waste solvent at the facility in 1959 and the second a release of ~ 3.7 TBq (~ 100 Ci) of "long-lived" particulate radioactivity at the Fuel Element Cutting Facility (FECF) in 1958 (AEC 1960). In the latter case, it must be assumed that some fraction of the particulate radioactivity released contained Pu and Np, but the specific isotopic composition of the released material was not detailed. For both releases the initial contamination was believed constrained to the immediate vicinity of the ICPP. However, without remediation, contamination from both incidents could be remobilized away from the original deposition sites by winds, but there appears to be no information in recent site environmental survey reports on the extent to which this may have occurred.

While it can be reasonably assumed that ^{237}Np airborne releases have accompanied those of Pu over the operating history of the ICPP, confirmation that this radionuclide has been a *long-term* component of waste discharges there have not been published. Such a confirmation can be obtained from measurements of this radionuclide in monitoring wells downgradient from the ICPP because of the discharge of low-level liquid wastes directly to the Snake River Plain aquifer between 1952 and 1984 (Beasley et al. 1993 and references therein). Figure 3 shows the location of monitoring wells that were sampled for both U isotopes and ^{237}Np and Table 2 shows the results of the TIMS measurements following isolation and purification of these radionuclides from $\leq$ 1-liter water samples collected in 1995.

The data of Table 2 illustrate two important points. First, within the uncertainties shown (1 σ), waters from well 45 and downgradient to well CFA-1 show elevated ^{235}U/^{238}U atom ratios over that expected from natural uranium. Second, measurable concentrations of ^{236}U exist in all wells sampled, while ^{237}Np was detectable only to distances represented by well 112. However, the decrease in ^{236}U between wells 112 and CFA-1, by a factor of ~ 44, suggests that if a similar decrease occurred in ^{237}Np, its quantitation would be doubtful which is exactly what the data shows. The use of larger sample sizes (~ 10 L) would likely have confirmed the presence of ^{237}Np in well 106. The data further emphasize the potential utility of ^{236}U as a unique tracer at the INEEL for both above and below-ground processes.

METHODS

SAMPLE COLLECTION AND PREPARATION

Between September, 1993 and September, 1995, a total of 123 soil samples were collected at the INEEL and other locations in Idaho, Montana, and Wyoming to measure the isotopic composition of transuranic radionuclides in the soils. The majority of samples (100) were

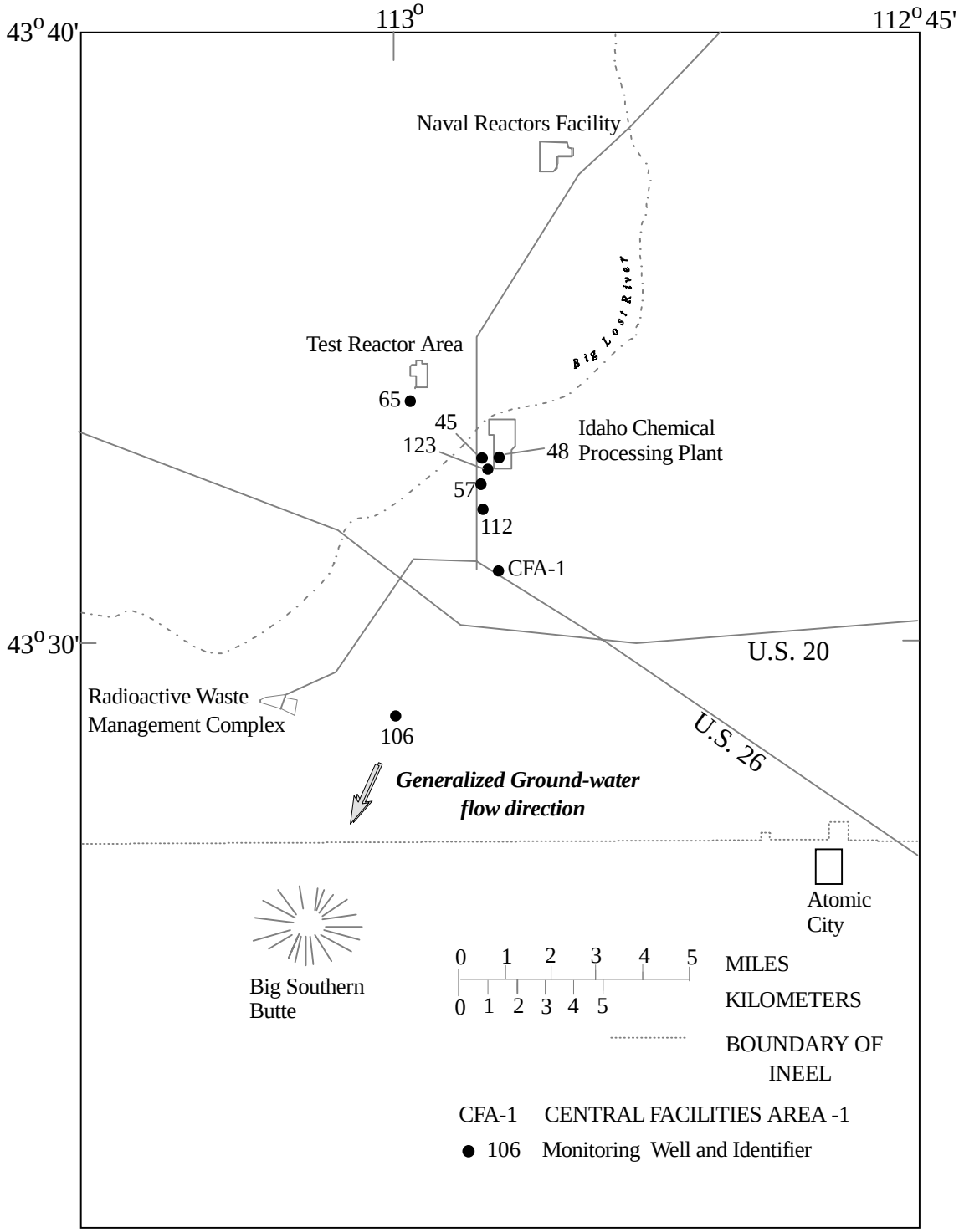

Figure 3. USGS monitoring wells sampled for measuring uranium isotopes and ^{237}Np.

TABLE 2

URANIUM AND NEPTUNIUM ATOM CONCENTRATIONS AND RATIOS IN SELECTED USGS MONITORING WELLS AT THE INEEL

Well Number	$^{234}U/^{238}U$	$^{235}U/^{238}U$¶	^{236}U (atoms L^{-1})	^{237}Np (atoms L^{-1})
65	0.00011384 ± 0.00000068	0.007269 ± 0.000044	1.39 ± 0.38 (10^{8})†	5.65 ± 0.80 (10^{5})
45	0.00011751 ± 0.00000071	0.007394 ± 0.000044	9.91 ± 0.22 (10^{10})	3.61 ± 0.08 (10^{8})
48	0.00012120 ± 0.00000073	0.007987 ± 0.000048	1.94 ± 0.04 (10^{11})	7.67 ± 0.17 (10^{8})
57	0.00011248 ± 0.00000067	0.007693 ± 0.000046	1.94 ± 0.04 (10^{11})	9.19 ± 0.21 (10^{8})
123	0.00011541 ± 0.00000038	0.007984 ± 0.000018	2.65 ± 0.04 (10^{11})	1.09 ± 0.04 (10^{9})
112	0.00010090 ± 0.00000061	0.007602 ± 0.000046	2.02 ± 0.05 (10^{11})	3.03 ± 0.08 (10^{8})
CFA-1	0.00011573 ± 0.00000069	0.007320 ± 0.000044	4.58 ± 0.15 (10^{9})	< 3 (10^{5})
106	0.00014507 ± 0.00000087	0.007254 ± 0.000044	2.05 ± 0.30 (10^{8})	<6 (10^{5})

¶ The $^{235}U/^{238}U$ atom ratio in natural uranium is 0.007253 while that for $^{234}U/^{238}U$ is 0.0000552 (De Bievre 1985). This latter ratio is highly variable due to isotopic fractionation of ^{234}U from ^{238}U in natural systems (Osmond 1980); †read as 1.39 ± 0.38 x 10^{10}

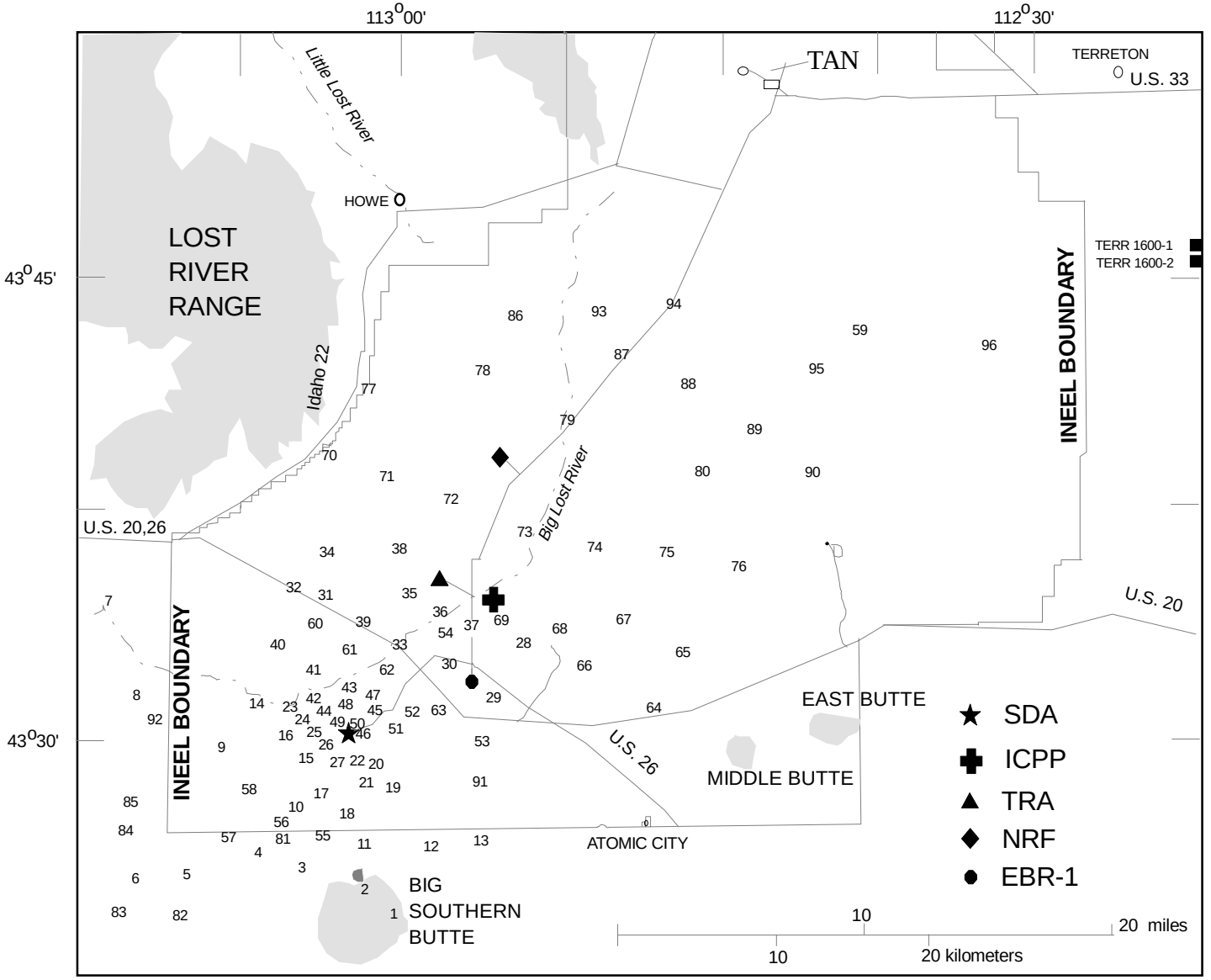

Figure 4. Soil core locations relative to INEEL facilities. Subsurface Disposal Area (SDA), Idaho Chemical Processing Plant (ICPP), Test Reactor Area (TRA), Naval Reactors Facility (NRF), Test Area North (TAN), and Experimental Breeder Reactor-1 (EBR-1). Tables in text carry W and E designations indicating samples collected in a generally western or eastern direction from the SDA.

collected on or near the INEEL and the latitude and longitude of these sample locations were determined using Global Positioning System (GPS) techniques (Ashtech 1993; Trimble 1993). Five National Geodetic Survey bench-marks were used for horizontal control and all sample locations were determined to within an accuracy of 1 meter. Table A of Appendix 1 shows the exact coordinates of each sampling site as determined by GPS measurements.

Figure 4 shows the onsite, and near site, sampling locations in relation to major facilities at the INEEL. Because the predominant prevailing wind direction is SW-NE (Figure 1), a larger proportion of the samples collected were radially away from the RWMC in that direction. The general area at each site was inspected prior to coring to reject those areas that showed obvious disturbance viz., human or animal activity, or evidence of wind/water erosion. At the chosen sampling sites, four soil cores were taken within an approximate 1-m square area using a drive core sampler 7.6 cm in diameter and 15.2 cm long. The four cores were then combined to form a single sample.

The coring depth (10 cm) was chosen based on results obtained from an earlier sample collection specifically designed to determine the depth to which Pu had penetrated the soil column at three different locations on the INEEL site. The location of these early coring sites are shown in Figure 5 and the Pu distribution over a depth interval of 20 cm are listed in Table 3. In all three cases, coring to 10 cm captured virtually the entire Pu inventory in the soil column, in agreement with previous experience observed in Pu soil measurements at the site (Bernabee 1993).

The coring sites were marked with a metal rod, centered in the sampling grid, and fluorescent flagging tape was attached to surrounding vegetation to aid in revisiting the site during the course of the three-year sampling. At the INEEL Lithologic Core Storage Laboratory, samples were weighed to the nearest 0.1 g and then oven dried (110°C) to constant weight. The samples were sieved using a # 10 (2mm) U.S. Standard Testing Sieve to remove vegetation and pebbles and this > 2mm fraction placed in curation. The < 2mm fraction was returned to the container and fifteen to twenty, 21 mm diameter ceramic jar mill pellets were added to aid in sample homogenization. The sample was placed on a jar mill at 30 revolutions min^{-1} and milled for 20-24 hours. The sample was then sieved using a # 35 (0.5mm) U.S. Standard Testing sieve, and the < 2mm and > 0.5mm fraction was weighed and curated; the < 0.5 mm fraction was weighed to the nearest 0.1g and constituted the sample ultimately aliquoted for transuranic analyses. All sieves, ceramic pellets, and other laboratory instruments were thoroughly cleaned between samples to prevent cross-contamination.

Table B, Appendix 1, shows that for the great majority of cores, ≥ 90% of the processed core material is contained in the < 0.5mm fraction ultimately retained for analysis. In cases where this percentage is < 90, the preponderant screened material (by weight) consisted of “pebble-sized” rock and, to a lesser degree, detrital vegetation. Exclusion of this material from analysis is unlikely to affect any of the conclusions reached in this report because: 1] there is a substantial body of evidence showing that Pu from sources such as those discussed in the BACKGROUND section of this report are associated with the fine particulate matter in soils (Dahlman et al. 1980 and references therein; Hakonson and Nyhan 1980 and references therein), and; 2] transfer of transuranics from soil to vegetation is not an efficient process and > 99 percent of the transuranic

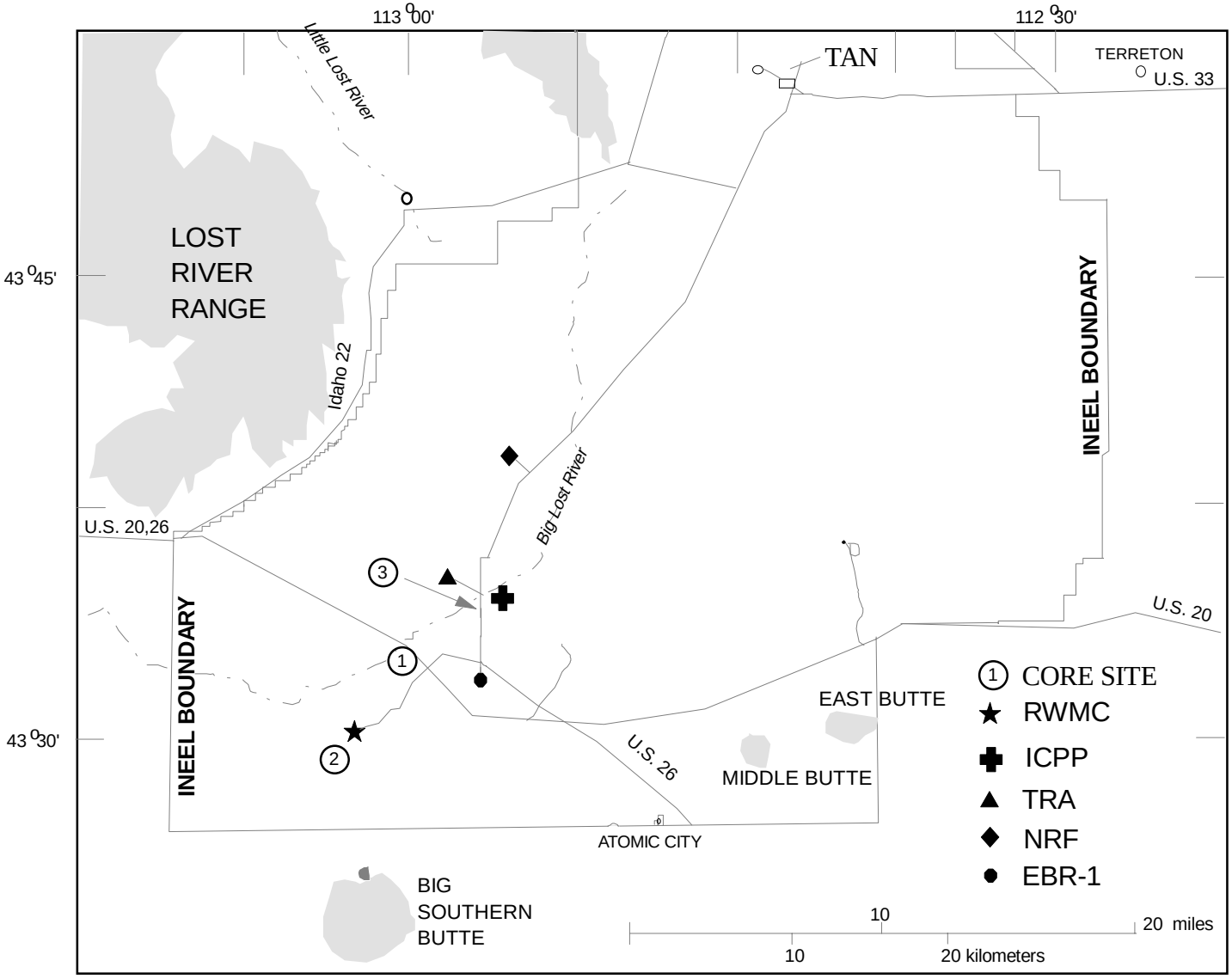

Figure 5. Location of initial coring sites to determine the penetration depth of Pu in INEEL soils.

TABLE 3

PENETRATION DEPTH OF Pu IN SOILS AT SELECTED SITES AT THE INEEL

Location	Depth (cm)	$^{239+240}$Pu (Bq kg $^{-1}$)	Percent total activity	^{238}Pu/$^{239+240}$Pu†	^{240}Pu/ ^{239}Pu ‡
Site 1	0 - 5	0.27 ± 0.06	~ 82	0.04 ± 0.01	0.165 ± 0.002
	5 - 10	0.06 ± 0.03	~ 18	-	-
	10 - 20	0.05 ± 0.05	-	-	-
Site 2	0 - 5	0.68 ± 0.07	100	0.05 ± 0.01	0.123 ± 0.004
	5 - 10	< 0.04	-		
	10 - 20	< 0.04	-	-	-
Site 3	0 - 5	0.70 ± 0.09	~ 93	0.84 ± 0.08	0.136 ± 0.005
	5 - 10	0.05 ± 0.01	~ 7	-	-
	10 - 20	<0.04	-	-	-

†Activity ratio; ‡atom ratio; - not measured

inventory present in a vegetated landscape is found in the soil column (Dahlman et al.1980; Hakonson and Nyhan 1980).

SAMPLE HOMOGENEITY

Sample homogeneity of the 0.5 mm fraction from the soil preparation activities was tested by analyzing selected samples for their ^{137}Cs content. The Radiological and Environmental Sciences Laboratory (RESL) at the INEEL analyzed 18 core samples, in duplicate, to determine the consistency of their ^{137}Cs content; sample sizes ranged between 10 and 15 g dry weight. Additionally, larger aliquots of these same cores (40-60g dry weight) were analyzed at EML to corroborate the absolute ^{137}Cs activities in the samples. Both laboratories use ^{137}Cs standards for instrument calibration that are traceable to the National Institute of Science and Technology (NIST), and the ^{137}Cs was determined by gamma-ray spectrometry techniques using Ge(Li) detectors. Table 4 shows, with few exceptions, that the processing procedures described above produces material whose homogeneity at the 10 to 15 g level, with respect to ^{137}Cs, is perfectly acceptable. Moreover, the agreement between RESL and EML for quantitation of ^{137}Cs is excellent. Other core samples, counted in duplicate at EML (40-60 g dry), consistently delivered ^{137}Cs results that were within ± 2 percent of one another giving confidence that the final archived material was well blended.

As described elsewhere (Beasley et al.1997), the mass spectrometry facility at the Pacific Northwest National Laboratory (PNNL) is maintained as an ultra-low level laboratory and samples destined for transuranic analysis there are carefully controlled to ensure that $^{239,240}Pu$ atom concentrations do not exceed 10^9 atoms sample^{-1}. This limit constrained the sample sizes chosen for thermal ionization mass spectrometry (TIMS) analyses to dry weights ranging between 1 and 2 g. Homogeneity at this sample size was determined by analyzing a selected number of core samples (in duplicate) by TIMS to determine the consistency of their $^{240}Pu/^{239}Pu$ ratios. Table 5 shows the results of these measurements and indicate that for samples where the total area sampled is ~ 182 cm^2 (> 1500 g dry), viz., the composited samples representing the great majority of cores described here, the homogeneity is comparable to that observed in larger aliquots as determined by ^{137}Cs measurements. However, when the area sampled is ~ 45 cm^2 (≤ 500 g dry), the detection of discrete particles becomes more probable. This can be seen from the results of individual core samples collected in the near vicinity of the composite core taken at site 54E. In both the R-1 and R-3 samples, the $^{240}Pu/^{239}Pu$ ratio in one of the aliquots analyzed is significantly different from that observed in the composite sample. Excluding these two values gives a weighted mean of 0.159 for the $^{240}Pu/^{239}Pu$ ratio in the remaining soils sampled, a value within ~ 4 percent of the mean ratio observed from duplicate analyses of the composite sample. Interestingly, in none of the measurements associated with the 54E location do $^{237}Np/^{239}Pu$ ratios exceed 0.45, the value expected for integrated global fallout in the northern hemisphere. This suggests that the low $^{240}Pu/^{239}Pu$ ratio material observed in the R-1 and R-2 samples does not have its origin at the ICPP where $^{237}Np/^{239}Pu$ ratios would be expected to be much higher than 0.45. As discussed later, however, such an interpretation is inconsistent with observations of both elevated ^{137}Cs inventories and enhanced $^{238}Pu/^{239+240}Pu$ activity ratios in soils at this particular site, unless Pu contributions from the ICPP are characterized by elevated $^{240}Pu/^{239}Pu$ ratios.

TABLE 4

HOMOGENEITY OF SOIL CORES AS DETERMINED BY ^{137}Cs MEASUREMENTS

Sample	^{137}Cs activity (Bq g^{-1}) RESL[a]	^{137}Cs activity (Bq g^{-1}) EML[a]
Carey, ID	0.033 ± 0.003[†]	0.029 ± 0.002
	0.030 ± 0.003	
Howe, ID	0.036 ± 0.003	0.037 ± 0.002
	0.043 ± 0.003	
Richfield, ID	0.029 ± 0.002	0.026 ± 0.002
	0.029 ± 0.002	
Terreton, ID	0.017 ± 0.002	0.013 ± 0.001
	0.013 ± 0.002	
Thermopolis, WY	0.023 ± 0.003	0.024 ± 0.003
	0.031 ± 0.003	
2W	0.038 ± 0.003	0.031 ± 0.002
	0.031 ± 0.002	
3W	0.021 ± 0.003	0.019 ± 0.001
	0.021 ± 0.003	
4W	0.019 ± 0.002	0.024 ± 0.001
	0.027 ± 0.002	
6W	0.023 ± 0.002	0.019 ± 0.001
	0.022 ± 0.003	
11W	0.026 ± 0.002	0.024 ± 0.002
	0.025 ± 0.003	
12W	0.026 ± 0.002	0.023 ± 0.002
	0.024 ± 0.003	
14W	0.016 ± 0.002	0.017 ± 0.001
	0.020 ± 0.002	
16W	0.015 ± 0.001	0.012 ± 0.001
	0.013 ± 0.003	

TABLE 4 (cont'd)

HOMOGENEITY OF SOIL CORES AS DETERMINED BY ^{137}Cs MEASUREMENTS

Sample	^{137}Cs activity (Bq g^{-1}) RESL[a]	^{137}Cs activity (Bq g^{-1}) EML[a]
17W	0.019 ± 0.002 0.021 ± 0.002	0.020 ± 0.001
18W	0.027 ± 0.003 0.029 ± 0.002	0.026 ± 0.001
19W	0.013 ± 0.001 0.015 ± 0.003	0.013 ± 0.001
23W	0.014 ± 0.001 0.017 ± 0.002	0.014 ± 0.001
25W	0.016 ± 0.003 0.014 ± 0.002	0.015 ± 0.001
26W	0.023 ± 0.003 0.016 ± 0.003	0.016 ± 0.001
34E	0.015 ± 0.002 0.018 ± 0.003	0.020 ± 0.001
37E	0.127 ± 0.006 0.120 ± 0.006	0.128 ± 0.007
41E	0.019 ± 0.003 0.019 ± 0.003	0.018 ± 0.001

[a]Sample sizes for RESL analyses ranged between 10-15g; those for EML ranged between 40-60g. Counting times for both sample sets was ~ 1000 minutes. Reference date: 1 January 1994.
All reported ^{137}Cs measurements were made by gamma-ray spectrometry using Ge(Li) detectors.
[†]Uncertainties at 1σ.

TABLE 5

HOMOGENEITY OF SELECTED SOIL CORES AS DETERMINED BY $^{240}Pu/^{239}Pu$ ATOM RATIO MEASUREMENTS

Core	$^{240}Pu/^{239}Pu$ atom ratio	Core	$^{240}Pu/^{239}Pu$ atom ratio	Core	$^{240}Pu/^{239}Pu$ atom ratio
Worland, WY	0.150 ± 0.001 0.155 ± 0.004	62W	0.149 ± 0.001 0.136 ± 0.001	90E	0.154 ± 0.001† 0.157 ± 0.001
Thermopolis, WY	0.163 ± 0.001 0.162 ± 0.001	81W	0.160 ± 0.001 0.132 ± 0.001	95E	0.161 ± 0.001 0.160 ± 0.001
3W	0.169 ± 0.001 0.172 ± 0.001	35E	0.153 ± 0.001 0.156 ± 0.003	54E (Composite)	0.162 ± 0.002 0.166 ± 0.001
4W	0.165 ± 0.001 0.164 ± 0.001	87E	0.172 ± 0.001 0.173 ± 0.001	54E R-1	0.153 ± 0.002 0.162 ± 0.002 0.109 ± 0.001
10W	0.140 ± 0.001 0.147 ± 0.001	88E	0.157 ± 0.001 0.130 ± 0.001	54E R-2	0.153 ± 0.002 0.162 ± 0.002
60W	0.152 ± 0.001 0.149 ± 0.001	89E	0.152 ± 0.001 0.156 ± 0.001	54E R-3	0.152 ± 0.002 0.113 ± 0.001
				54E R-4	0.160 ± 0.002 0.166 ± 0.001

† Uncertainties shown are 1 σ. Sample weights ranged between 1 and 2 g dry.

CHEMICAL SEPARATION/PURIFICATION OF ACTINIDES AND TIMS MEASUREMENTS

The analysis techniques used to isolate and purify the actinides reported here and the TIMS measurements that lead to their quantitation have been recently described by Beasley et al. (1997) and will not be described here. For the purposes of this report, it is sufficient to emphasize two important points: 1] all soils were totally dissolved in a mixture of HCL and HF to insure that refractory transuranic radionucildes were brought into solution, and; 2] that during the course of the soil core measurements described, repeated analyses of soils from EML's curatorial collection delivered reproducible Pu isotopic ratios and Pu abundances as measured in these same soils by other mass spectrometry facilities (Beasley et al. 1997).

RADIONUCLIDE INVENTORY ESTIMATE

The radionuclide inventories reported here were calculated as the product of the activity per unit weight of soil and the total sample weight collected divided by the area of the soil column sampled. It has been estimated that a precision of ± 9 percent is attainable in the sampling and analysis of fallout ^{137}Cs and $^{239+240}$Pu when the total area sampled is ~ 600 cm^2 (Shebell and Hutter 1996). However, when the total area sampled approximates 180 cm^2, the collection and analysis precision for ^{137}Cs sum to ± 15% (Shebell and Hutter 1996). Thus, while the *analytical* uncertainties associated with inventory estimates for $^{239+240}$Pu and ^{237}Np discussed here are generally < 5 percent (because of the high sensitivity afforded by the TIMS measurements), a more conservative estimate of their overall inventory uncertainties is probably in the range of ± 15 percent.

ISOTOPE SYSTEMATICS

It is possible to resolve the fractional contribution of two components in a mixture if their isotopic ratios differ significantly (Faure 1977). Using this approach, Krey et al. (1976) showed that the ratio of $^{239+240}$Pu activities from two different sources can be expressed as:

$$\frac{\left(Activity\right)_1}{\left(Activity\right)_2} = \frac{\left(R_2 - R\right)}{\left(R - R_1\right)} \frac{\left(1 + 3.67 R_1\right)}{\left(1 + 3.67 R_2\right)} \qquad (5)$$

where:
$(Activity)_1$ = activity from source 1;
$(Activity)_2$ = activity from source 2;
R_1, R_2, R = ^{240}Pu/^{239}Pu isotope ratio in source 1, 2 and in the mixture, respectively;
and the constant 3.67 is the ratio of the half lives of ^{239}Pu to ^{240}Pu. Given the further identity that the activity measured in the mixture must equal the sum of the activities from sources 1 and 2, viz.:

$$(Activity)_1 + (Activity)_2 = (Activity)_{mix} \qquad (6)$$

it is possible to calculate the activity from sources 1 and 2 in the sample. The degree of specificity in source allocation depends on the numerical differences in R_1 and R_2, and the uncertainties in the isotopic measurements which are propagated through the calculations.

RESULTS AND DISCUSSION

$^{240}Pu/^{239}Pu$ ATOM RATIOS FOR NTS AND SDA/ICPP SOURCES

NTS

Because of the multiple sources of transuranic radionuclides at the INEEL site, it is first necessary to estimate the isotopic signatures of these radionuclides that can be assumed to constitute "background". Figure 6 shows offsite locations in Idaho, Montana and Wyoming for a number of soil cores collected for this purpose as well as several locations for which $^{240}Pu/^{239}Pu$ atom ratios are available from other studies. Tables 6 and 7 list the values of the transuranic radionuclide atom ratios (one to another), and their concentrations, which indicate that, in general, lower $^{240}Pu/^{239}Pu$ atom ratios are observed to the east of the Idaho/Wyoming border than for sites to the west of Yellowstone, WY. This observation undoubtedly reflects the differing trajectories followed by NTS fallout over the period of testing.

From the analysis of a large number of soil cores collected in Utah, Krey and Beck (1981) concluded that the most representative $^{240}Pu/^{239}Pu$ atom ratio for NTS fallout debris deposited there was 0.0321. Figure 7 shows a correlation plot ($r^2 = 0.67$) of the $^{240}Pu/^{239}Pu$ versus $^{242}Pu/^{239}Pu$ atom ratios observed at the offsite sampling locations set out in Table 6 that are assumed to contain only mixtures of global and regional NTS fallout, i.e., a two-component mixing curve. Regression of the data gives a $^{240}Pu/^{239}Pu$ atom ratio of 0.038 for the NTS end member, in good agreement with the Utah data of Krey and Beck given the fact that fallout deposition in Utah from certain of the NTS tests may not have reached the locations sampled in Idaho, Montana, and Wyoming. Assuming that integrated global fallout is characterized by $^{240}Pu/^{239}Pu$ atom ratios of 0.180, and that this same ratio for NTS fallout at the sampling sites shown is ~ 0.0400, the calculated percentage of NTS fallout $^{239+240}Pu$ in the offsite soils sampled is shown in Table 8. The $^{240}Pu/^{239}Pu$ atom ratio measured in Fontenelle, WY soil in 1993 is ~ 20 percent lower than the same ratio measured by Beck and Anspaugh (1991) in soils at Kemmerer, WY in 1982/1983 (Figure 6). The mean and standard deviation of the $^{240}Pu/^{239}Pu$ atom ratio observed for *all* locations sampled west of Yellowstone, WY (Table 6) is 0.159 ± 0.012, a value close to that for Idaho Falls, ID (0.166) reported by Beck and Anspaugh (1991).

There are several points to be emphasized from the data shown in Tables 6 and 8. First, in only a few cases do $^{237}Np/^{239}Pu$ atom ratios approach the estimated northern hemisphere fallout value of 0.45 (Table 6); where they do, the $^{240}Pu/^{239}Pu$ atom ratios agree, within the measurement uncertainties, with those expected from global fallout. Second, at least two of the sites sampled (Daniel and Ethete, WY) show radionuclide inventories (Table 8) that are significantly lower than

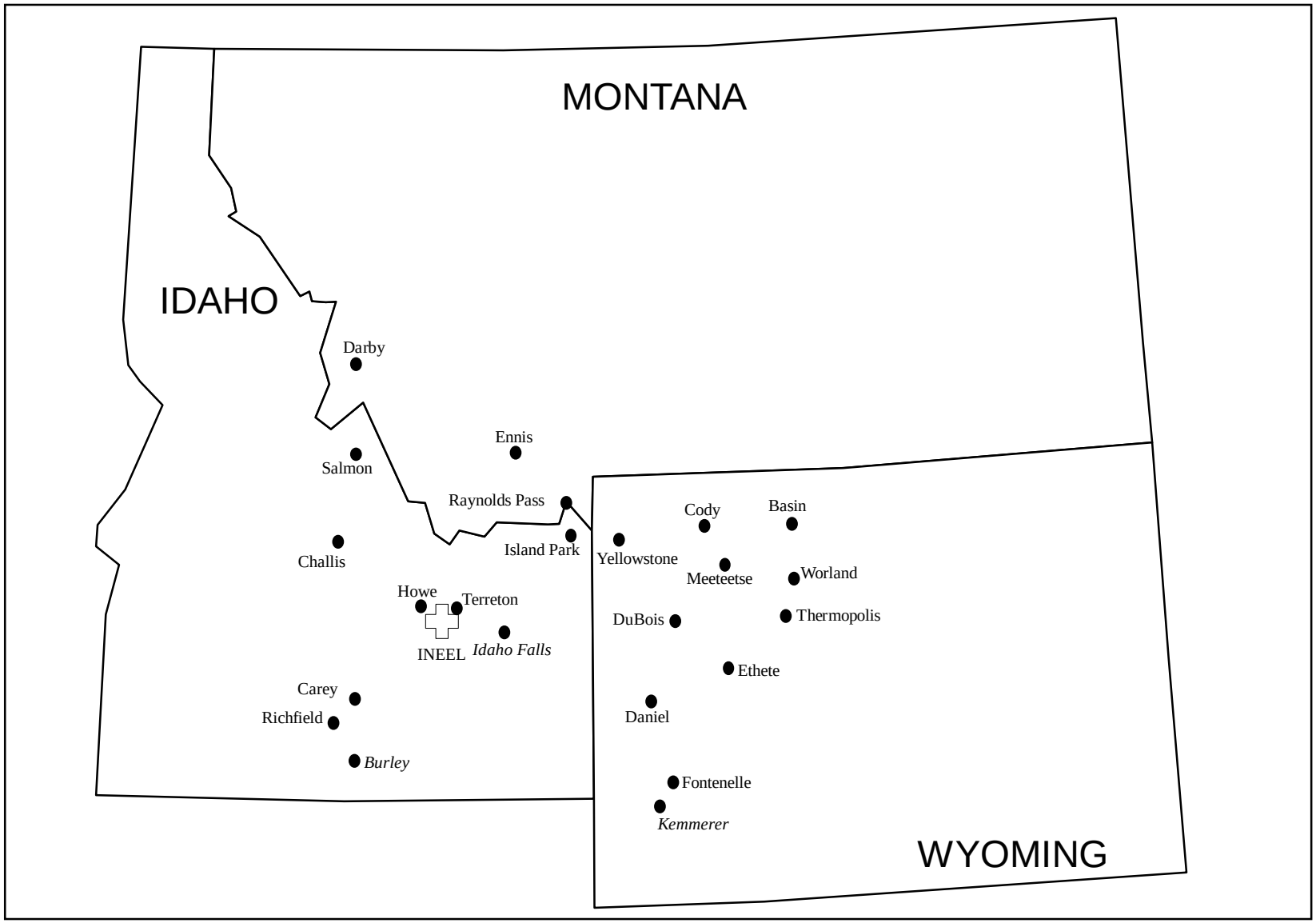

Figure 6. Location of offsite core collections in Idaho, Montana, and Wyoming. Cities in italics represent sites where other isotopic Pu measurements have been made to determine the presence of Nevada Test Site debris (see text). Locations are approximate and are only intended to show the general spatial relationship of the sampling sites.

TABLE 6

RATIOS OF NEPTUNIUM AND PLUTONIUM ISOTOPES IN OFFSITE SOILS COLLECTED IN ID, MT, and WY

Location	Atom Ratio ± 1σ Uncertainty				Activity Ratio ± 1σ
	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$	$^{237}Np/^{239,240}Pu$[†]
Carey, ID	0.357 ± 0.010	0.158 ± 0.001	0.00217 ± 0.00005	0.00327 ± 0.00006	0.00254 ± 0.00004
Challis, ID	0.420 ± 0.012	0.168 ± 0.001	0.00246 ± 0.00005	0.00340 ± 0.00007	0.00292 ± 0.00008
Howe, ID	0.385 ± 0.011	0.161 ± 0.001	0.00240 ± 0.00004	0.00328 ± 0.00005	0.00273 ± 0.00009
Island Park, ID	0.399 ± 0.010	0.176 ± 0.001	0.00256 ± 0.00004	0.00396 ± 0.00005	0.00273 ± 0.00007
Richfield, ID	0.294 ± 0.008	0.160 ± 0.001	0.00229 ± 0.00004	0.00331 ± 0.00005	0.00186 ± 0.00005
Salmon, ID	0.351 ± 0.013	0.167 ± 0.001	0.00248 ± 0.00010	0.00291 ± 0.00010	0.00245 ± 0.00009
Terreton, ID (1)	0.248 ± 0.007	0.128 ± 0.001	0.00172 ± 0.00005	0.00229 ± 0.00006	0.00190 ± 0.00006
Terreton, ID (1[§])	0.335 ± 0.016	0.148 ± 0.001	0.00180 ± 0.00023	0.00282 ± 0.00021	0.00244 ± 0.00012
Terreton, ID (2[¶])	0.406 ± 0.011	0.156 ± 0.001	0.00216 ± 0.00007	0.00333 ± 0.00008	0.00291 ± 0.00008
Darby, MT	0.433 ± 0.014	0.176 ± 0.001	0.00255 ± 0.00005	0.00383 ± 0.00008	0.00296 ± 0.00009
Ennis, MT	0.310 ± 0.016	0.154 ± 0.001	0.00221 ± 0.00003	0.00315 ± 0.00004	0.00224 ± 0.00012
Ennis, MT[‡]	0.309 ± 0.012	0.149 ± 0.001	0.00206 ± 0.00005	0.00295 ± 0.00005	0.00223 ± 0.00009
Raynolds Pass, MT	0.359 ± 0.014	0.164 ± 0.001	0.00241 ± 0.00002	0.00344 ± 0.00002	0.00252 ± 0.00011
Basin, WY	0.279 ± 0.011	0.139 ± 0.001	0.00168 ± 0.00005	0.00317 ± 0.00005	0.00209 ± 0.00009
Cody, WY	0.307 ± 0.012	0.140 ± 0.001	0.00195 ± 0.00003	0.00262 ± 0.00003	0.00230 ± 0.00010

TABLE 6 (cont'd)

RATIOS OF NEPTUNIUM AND PLUTONIUM ISOTOPES IN OFFSITE SOILS COLLECTED IN ID, MT, and WY

Location	Atom Ratio ± 1σ Uncertainty				Activity Ratio ± 1σ
	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$	$^{237}Np/^{239,240}Pu^{\dagger}$
Daniel, WY	0.330 ± 0.010	0.156 ± 0.001	0.00270 ± 0.00021	0.00299 ± 0.00019	0.00238 ± 0.00007
DuBois, WY	0.406 ± 0.013	0.156 ± 0.001	0.00233 ± 0.00005	0.00279 ± 0.00007	0.00291 ± 0.00010
Ethete, WY	0.336 ± 0.009	0.137 ± 0.001	0.00182 ± 0.00007	0.00225 ± 0.00007	0.00252 ± 0.00008
Fontenelle, WY	0.282 ± 0.017	0.129 ± 0.001	0.00185 ± 0.00007	0.00285 ± 0.00007	0.00227 ± 0.00014
Meeteetse, WY	0.341 ± 0.011	0.153 ± 0.001	0.00216 ± 0.00005	0.00290 ± 0.00006	0.00247 ± 0.00008
Thermopolis, WY	0.472 ± 0.018	0.162 ± 0.001	0.00258 ± 0.00006	0.00307 ± 0.00005	0.00335 ± 0.00015
Worland, WY	0.342 ± 0.015	0.155 ± 0.001	0.00217 ± 0.00005	0.00289 ± 0.00005	0.00247 ± 0.00011
Yellowstone, WY	0.189 ± 0.005	0.159 ± 0.001	0.00231 ± 0.00003	0.00322 ± 0.00003	0.00135 ± 0.00005

[§]Represents repeat sampling of coring site (1) one year later; [¶] sample from second coring site (2) ~ 1 km from first coring site; [‡] second core from Ennis ~ 20 km from first Ennis coring site; [†] radioactivity ratios; * decay corrected to 1 January 1995; [†] the $^{237}Np/^{239+240}Pu$ alpha activity ratio calculated from mass spectrtometrically determined atom ratios and known half lives. Values used for the half lives of ^{237}Np, ^{239}Pu, ^{240}Pu, and ^{241}Pu were (2.14 ± 0.01) x 10^6 yr, (2.4119 ± 0.0027) x 10^4 yr, (6.564 ± 0.011) x 10^3 yr, and 14.33 ± 0.02 yr, respectively.

TABLE 7

PLUTONIUM AND NEPTUNIUM CONCENTRATIONS IN OFFSITE SOILS COLLECTED IN ID, MT, and WY

Location	Concentration ± 1σ Uncertainty (atoms g^{-1})				
	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Carey, ID	2.16 ± 0.02 (10^{8})♦	6.06 ± 0.12 (10^{8})	9.58 ± 0.20 (10^{7})	1.31 ± 0.04 (10^{6})	1.98 ± 0.05 (10^{6})
Challis, ID	1.06 ± 0.03 (10^{8})	2.53 ± 0.04 (10^{8})	4.26 ± 0.07 (10^{7})	6.24 ± 0.17 (10^{5})	8.60 ± 0.22 (10^{5})
Howe, ID	2.92 ± 0.08 (10^{8})	7.62 ± 0.16 (10^{8})	1.22 ± 0.03 (10^{8})	1.83 ± 0.05 (10^{6})	2.50 ± 0.08 (10^{6})
Island Park, ID	2.55 ± 0.07 (10^{8})	6.40 ± 0.11 (10^{8})	1.13 ± 0.02 (10^{8})	1.64 ± 0.04 (10^{6})	2.54 ± 0.05 (10^{6})
Richfield, ID	1.57 ± 0.04 (10^{8})	5.33 ± 0.10 (10^{8})	8.53 ± 0.17 (10^{7})	1.22 ± 0.03 (10^{6})	1.76 ± 0.04 (10^{6})
Salmon, ID	6.47 ± 0.21 (10^{7})	1.84 ± 0.03 (10^{8})	3.07 ± 0.06 (10^{7})	4.57 ± 0.20 (10^{5})	5.36 ± 0.20 (10^{5})
Terreton, ID (1)	1.01 ± 0.03 (10^{8})	4.08 ± 0.08 (10^{8})	5.23 ± 0.11 (10^{7})	7.02 ± 0.24 (10^{5})	9.35 ± 0.30 (10^{5})
Terreton, ID (1§)	1.17 ± 0.05 (10^{8})	3.50 ± 0.11 (10^{8})	5.18 ± 0.17 (10^{7})	6.31 ± 0.81(10^{5})	9.86 ± 0.79 (10^{5})
Terreton, ID (2¶)	1.62 ± 0.05 (10^{8})	4.00 ± 0.07 (10^{8})	6.25 ± 0.11 (10^{7})	8.65 ± 0.32 (10^{5})	1.33 ± 0.04 (10^{6})
Darby, MT	2.84 ± 0.08 (10^{8})	6.57 ± 0.11 (10^{8})	1.16 ± 0.02 (10^{8})	1.68 ± 0.04 (10^{6})	2.52 ± 0.07 (10^{6})
Ennis, MT	2.94 ± 0.16 (10^{8})	9.50 ± 0.16 (10^{8})	1.46 ± 0.03 (10^{8})	2.10 ± 0.05 (10^{6})	2.99 ± 0.06 (10^{6})
Ennis, MT‡	1.95 ± 0.07 (10^{8})	6.31 ± 0.11 (10^{8})	9.41 ± 0.17 (10^{7})	1.30 ± 0.04 (10^{6})	1.86 ± 0.05 (10^{6})
Raynolds Pass, MT	9.45 ± 0.38 (10^{8})	2.64 ± 0.04 (10^{9})	4.33 ± 0.08 (10^{8})	6.34 ± 0.12 (10^{6})	9.07 ± 0.16 (10^{6})
Basin, WY	7.83 ± 0.32 (10^{7})	2.80 ± 0.05 (10^{8})	3.90 ± 0.07 (10^{7})	4.74 ± 0.15 (10^{5})	8.89 ± 0.20 (10^{5})

TABLE 7 (cont'd)

PLUTONIUM AND NEPTUNIUM CONCENTRATIONS IN OFFSITE SOILS COLLECTED IN ID, MT, AND WY

Location	Concentration ± 1σ Uncertainty (atoms g^{-1})				
	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Cody, WY	1.69 ± 0.07 (10^8)	5.49 ± 0.09 (10^8)	7.68 ± 0.13 (10^7)	1.07 ± 0.02(10^6)	1.44 ± 0.03 (10^6)
Daniel, WY	1.52 ± 0.04(10^7)	4.60 ± 0.10 (10^7)	7.17 ± 0.16 (10^6)	1.24 ± 0.10 (10^5)	1.38 ± 0.09 (10^5)
DuBois, WY	2.03 ± 0.06 (10^8)	5.00 ± 0.11 (10^8)	7.81 ± 0.18 (10^7)	1.17 ± 0.04 (10^6)	1.40 ± 0.05 (10^6)
Ethete, WY	5.83 ± 0.16 (10^7)	1.74 ± 0.04 (10^8)	2.38 ± 0.05 (10^7)	3.15 ± 0.14 (10^5)	3.90 ± 0.14 (10^5)
Fontenelle, WY	2.15 ± 0.13 (10^8)	7.62 ± 0.17 (10^8)	9.87 ± 0.22 (10^7)	1.42 ± 0.06 (10^6)	2.17 ± 0.07 (10^6)
Meeteetse, WY	3.68 ± 0.11 (10^8)	1.08 ± 0.02 (10^9)	1.64 ± 0.03 (10^8)	2.32 ± 0.07 (10^6)	3.11 ± 0.09 (10^6)
Thermopolis, WY	1.87 ± 0.08 (10^8)	3.96 ± 0.07 (10^8)	6.40 ± 0.11(10^7)	1.02 ± 0.03 (10^6)	1.22 ± 0.03 (10^6)
Worland, WY	2.45 ± 0.11 (10^8)	7.14 ± 0.12 (10^8)	1.10 ± 0.02 (10^8)	1.55 ± 0.04 (10^6)	2.06 ± 0.05 (10^6)
Yellowstone, WY	1.98 ± 0.06 (10^8)	1.05 ± 0.02 (10^9)	1.67 ± 0.03 (10^8)	2.42 ± 0.05 (10^6)	3.38 ± 0.07 (10^6)

♦Read as (2.16 ± 0.02) x 10^8; § represents repeat sampling of coring site (1) one year later; ¶ sample from coring site (2) collected ~ 1 km from first coring site; ‡second core from Ennis ~ 20 km from first Ennis coring site; decay corrected to 1 January 1995 using a $T_{½}$ of 14.33 ± 0.02 yr.

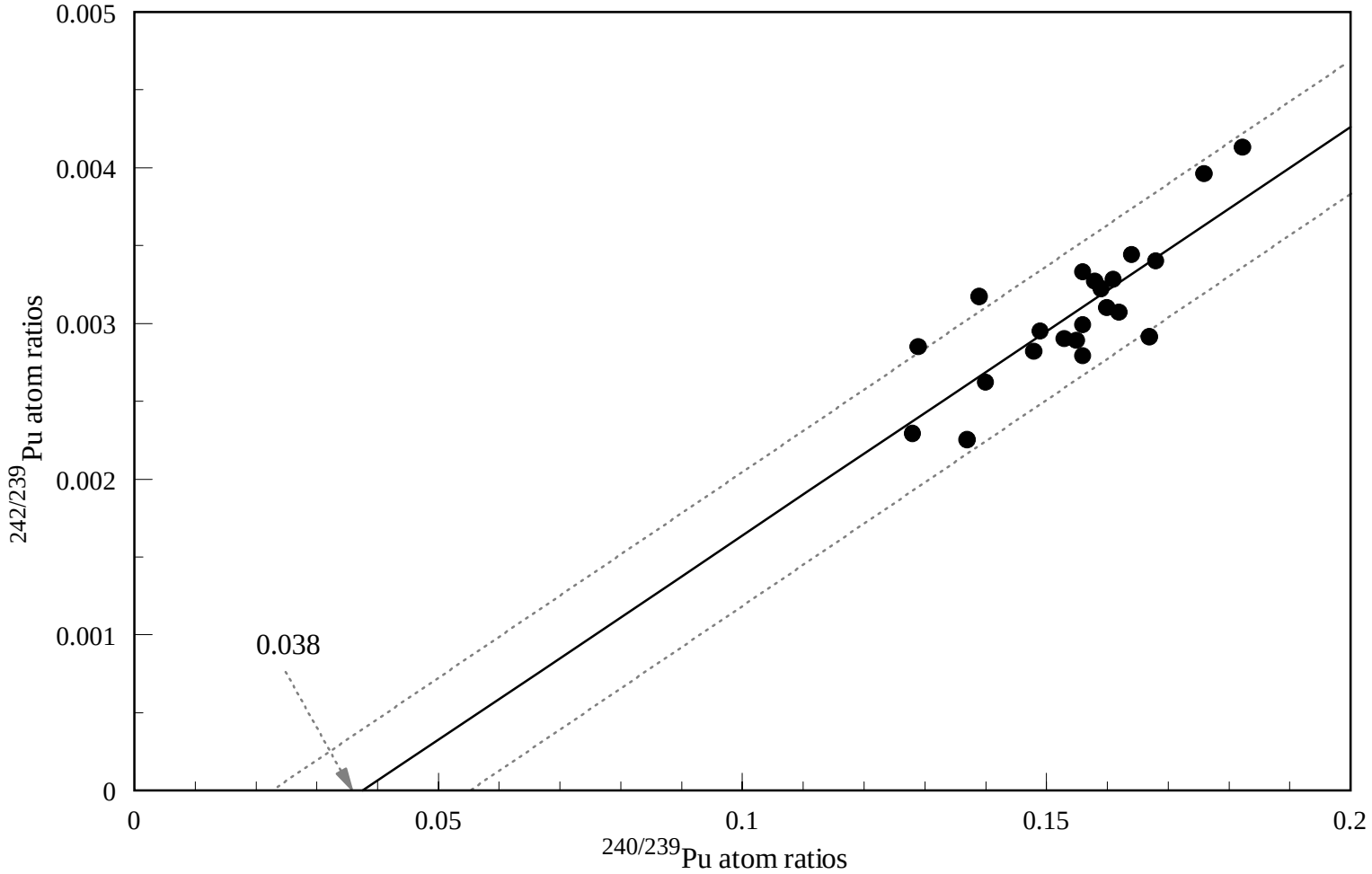

Figure 7. Plutonium isotope mixing curve showing the mean $^{240}Pu/^{239}Pu$ atom ratio of NTS-derived Pu insoils from Idaho, Montana, and Wyoming.The dotted lines reasonably bracket a range of ratios characteristic of post- shot NTS debris.

TABLE 8

^{137}Cs, ^{237}Np, AND $^{239+240}Pu$ INVENTORIES AND $^{137}Cs/^{239+240}PU$ ACTIVITY RATIOS IN IDAHO, MONTANA, AND WYOMING SOILS. UNCERTAINTIES AT 1σ

Location	^{137}Cs (Bq m^{-2})	^{237}Np (Bq m^{-2})	$^{239+240}Pu$ (Bq m^{-2})	Cs/Pu	% NTS
Carey, ID	1677 ± 104	0.122 ± 0.001	47.90 ± 0.96	35 ± 2	12
Challis, ID	826 ± 70	0.083 ± 0.002	28.33 ± 0.45	29 ± 3	6
Howe, ID	2027 ± 114	0.153 ± 0.004	56.43 ± 1.19	36 ± 2	10
Island Park, ID	4021 ± 238	0.259 ± 0.007	94.80 ± 1.65	42 ± 3	2
Richfield, ID	1710 ± 104	0.098 ± 0.003	46.95 ± 0.89	36 ± 2	10
Salmon, ID	1027 ± 73	0.068 ± 0.002	27.58 ± 0.45	37 ± 3	7
Terreton, ID (1)	1117 ± 69	0.084 ± 0.003	44.49 ± 0.88	25 ± 2	29
Terreton, ID (1§)	1694 ± 123	0.148 ± 0.006	60.50 ± 1.91	28 ± 2	17
Terreton, ID (2¶)	2559 ± 167	0.198 ± 0.006	68.20 ± 1.21	38 ± 2	13
Darby, MT	2213 ± 132	0.199 ± 0.006	67.25 ± 1.14	33 ± 2	2
Ennis, MT	3920 ± 212	0.243 ± 0.013	109.0 ± 1.86	36 ± 2	14
Ennis, MT ‡	2725 ± 153	0.214 ± 0.008	95.28 ± 1.68	29 ± 2	16
Raynolds Pass, ID	3760 ± 181	0.236 ± 0.009	93.93 ± 1.44	40 ± 2	8
Basin, WY	1077 ± 49	0.075 ± 0.003	35.78 ± 0.65	30 ± 2	22
Cody, WY	2228 ± 117	0.145 ± 0.006	63.20 ± 1.05	35 ± 2	22
Daniel, WY	110 ± 12	0.014 ± 0.0004	5.85 ± 0.13	19 ± 2	13
DuBois, WY	2367 ± 134	0.185 ± 0.005	63.54 ± 1.41	37 ± 2	13
Ethete, WY	595 ± 30	0.045 ± 0.001	17.77 ± 0.41	33 ± 2	23
Fontenelle, WY	1791 ± 107	0.144 ± 0.009	66.59 ± 1.50	27 ± 2	28
Meeteetse, WY	2626 ± 146	0.218 ± 0.006	88.54 ± 1.65	30 ± 2	14
Thermopolis, WY	1193 ± 66	0.092 ± 0.004	27.46 ± 0.49	43 ± 3	9
Worland, WY	2339 ± 131	0.162 ± 0.007	65.77 ± 1.12	36 ± 2	13
Yellowstone, WY	1407 ± 78	0.063 ± 0.002	46.69 ± 0.90	30 ± 2	11

§Repeat sampling of coring site (1) one year later; ¶sample from second coring site (2) ~ 1 km from first coring site; ‡second core from Ennis ~ 20 km from first Ennis site. The 1994 expected Cs/Pu activity ratio in global fallout is 38 ± 2 (Beck and Krey 1983). $^{240}Pu/^{239}Pu_{(global)}$ and $^{240}Pu/^{239}Pu_{(NTS)}$ used were 0.1800 and 0.0400, respectively.

those observed elsewhere, probably from wind erosional losses, making the utility of the data from these sites marginal. Third, the majority of the sampling sites show an inventory deficit for ^{137}Cs, ^{237}Np, and $^{230+240}Pu$ from that expected at the latitudes of sampling again reinforcing the observation that in semi-arid environments, low annual precipitation during the years of above-ground testing led to reduced fallout scavenging. Finally, it is instructive to show how the calculated percentage of NTS fallout changes with the magnitude assigned to the NTS $^{240}Pu/^{239}Pu$ atom ratio. Using a $^{240}Pu/^{239}Pu$ atom ratio of 0.0400, the calculated percentages of NTS fallout in soils at Carey, ID and Terreton, ID (1) are approximately 12 and 29 percent, respectively (Table 8). Assigning a value of 0.0321 to the NTS $^{240}Pu/^{239}Pu$ atom ratio, as for Utah soils, changes these same percentages to approximately 11 and 27.

SDA and ICPP

The same technique described above can be used to estimate the $^{240}Pu/^{239}Pu$ atom ratio of "non-fallout" Pu at the INEEL. The choice of a fallout value (integrated global fallout + NTS) for this ratio was determined by averaging the $^{240}Pu/^{239}Pu$ atom ratios of thirteen soil cores taken both offsite (Table 7) and onsite (Table C, Appendix 1). The cores selected were those collected at sites 1W, 2W, 3W, 4W, 87E, 94E, 95E, and 96E (Figure 4) and those at Carey, Challis, Salmon, and Richfield, ID, and Raynolds Pass, MT (Figure 6). The mean $^{240}Pu/^{239}Pu$ atom ratio measured at these locations is 0.166 ± 0.004 where the standard deviation shown is at the 67 percent confidence interval (1σ), once again confirming the $^{240}Pu/^{239}Pu$ atom ratio measured at Idaho Falls, ID in the early 1980s (Beck and Anspaugh 1991).

Figure 8 shows a plot of the $^{242}Pu/^{239}Pu$ versus $^{240}Pu/^{239}Pu$ atom ratios for all cores collected onsite (Table C, Appendix 1) whose measured $^{240}Pu/^{239}Pu$ atom ratios are ≤ 0.166, the value assigned to the onsite "fallout" end member of the two component mixing curve. As indicated, the estimated $^{240}Pu/^{239}Pu$ atom ratio of the "non-fallout" Pu measured in these soils is ~ 0.055. While the majority of values shown fall close to the mixing curve, a number of cores are displaced from the regression line suggesting that these soils contain "non-fallout" Pu whose $^{240}Pu/^{239}Pu$ atom ratios are significantly larger and smaller than 0.055, a finding not unexpected given the multiple onsite Pu source terms. However, as discussed in greater detail below, this value does represent the best *mean estimate* of the non-fallout $^{240}Pu/^{239}Pu$ atom ratio component on the INEEL site.

ORIGIN OF ONSITE RADIONUCLIDES

Table 9 shows the inventories of ^{237}Np, $^{239+240}Pu$, and ^{137}Cs as well as Cs/Pu activity ratios, the percentage of non-fallout Pu at the coring site, and the estimated non-fallout $^{239+240}Pu$ inventory as reconstructed from the measurement data of Tables C and D, Appendix 1. The percentage of non-fallout Pu was calculated using a $^{240}Pu/^{239}Pu$ atom ratio of 0.055; where the measured $^{240}Pu/^{239}Pu$ atom ratio is ≥ 0.166, the non-fallout Pu contribution is considered to be zero. Because relationships between the different radionuclide inventories are useful in determining provenance, several approaches are presented below to better understand the contribution of SDA and ICPP-derived material to the measured inventories.

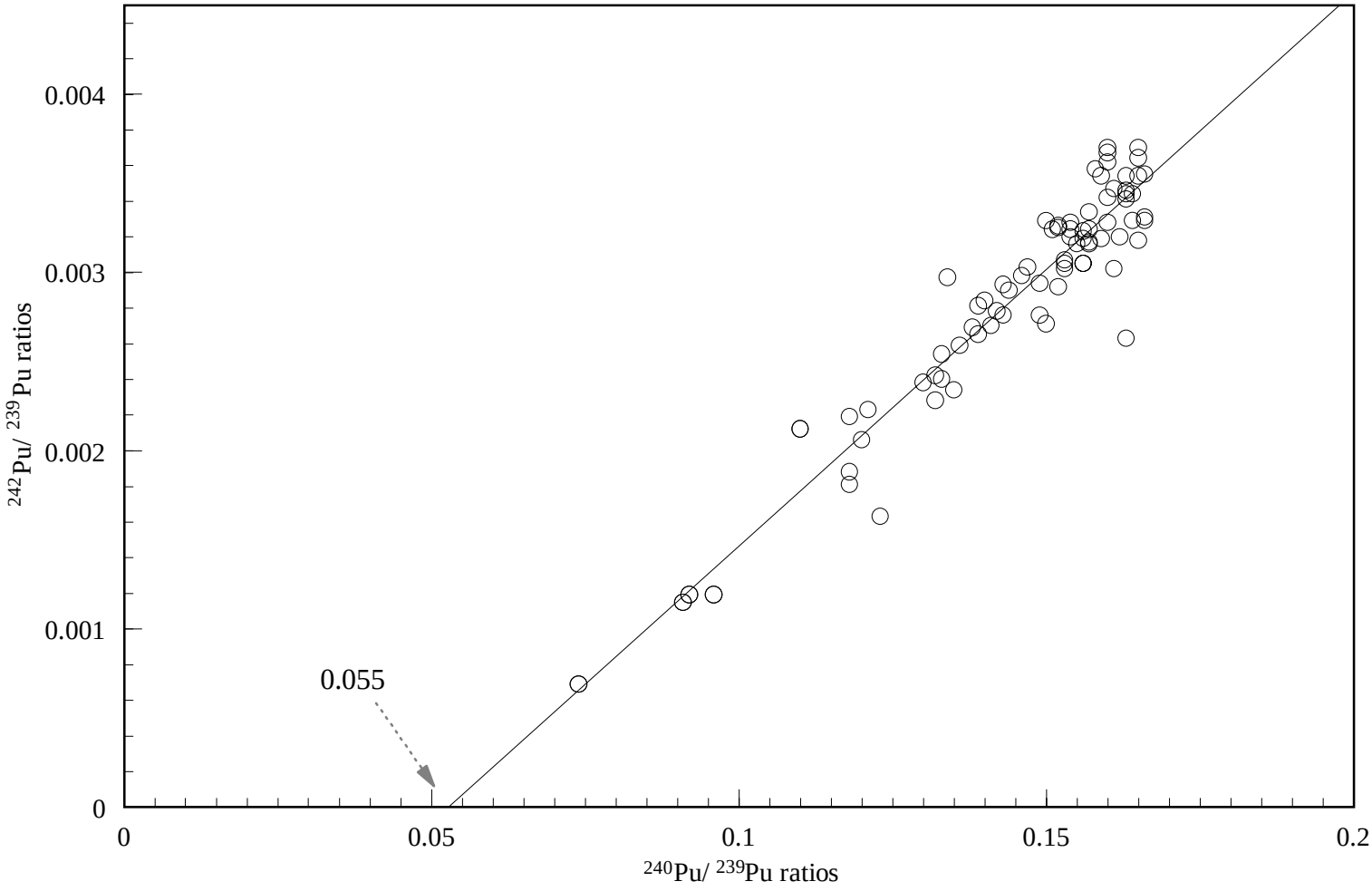

Figure 8. Correlation plot of ^{242}Pu/ ^{239}Pu versus ^{240}Pu/ ^{239}Pu atom ratios for INEEL cores whose ^{240}Pu/^{239}Pu atom ratios ≤ 0.166. A number of onsite cores are significantly displaced from the regression line (r^2 = 0.93) suggesting Pu whose 240/239 atom ratios are both larger and smaller than 0.055.

TABLE 9

AREAL INVENTORIES OF ^{137}Cs, ^{237}Np, $^{239+240}Pu$, AND $^{137}Cs/^{239+240}Pu$ ACTIVITY RATIOS IN INEEL SOILS, AND PERCENTAGE OF NON-FALLOUT (NF) Pu[†] AT THE CORING SITES

Core	Areal Inventories ± 1σ uncertainty (Bq m^{-2})					
	^{137}Cs*	^{237}Np	$^{239+240}Pu$	Cs/Pu*	% NF Pu	$^{239+240}Pu_{(NF)}$
1W	2830 ± 110	0.184 ± 0.005	72.77 ± 1.54	39 ± 2	0	0
3W	1970 ± 110	-	43.93 ± 0.64	45 ± 3	0	0
4W	2450 ± 110	-	59.26 ± 1.06	41 ± 2	0	0
5W	1880 ± 120	0.143 ± 0.004	54.17 ± 0.88	35 ± 2	10 ± 3	5.4 ± 1.6
6W	2040 ± 130	0.154 ± 0.010	47.15 ± 0.79	43 ± 3	0	0
7W	2060 ± 90	-	55.50 ± 1.17	37 ± 2	5 ± 3	2.8 ± 1.7
8W	2350 ± 110	-	60.64 ± 0.99	39 ± 2	4 ± 3	2.4 ± 1.8
9W	1300 ± 60	0.109 ± 0.005	42.94 ± 0.81	30 ± 2	19 ± 3	8.2 ± 1.3
10W	2000 ± 90	-	57.25 ± 0.15	35 ± 2	19 ± 3	10.9 ± 1.7
11W	2590 ± 170	0.158 ± 0.008	60.68 ± 1.25	43 ± 3	0	0
12W	2560 ± 150	0.155 ± 0.002	59.21 ± 0.97	43 ± 3	4 ± 3	2.4 ± 1.8
13W	2010 ± 110	0.155 ± 0.004	53.70 ± 0.90	37 ± 2	2 ± 3	1.1 ± 1.9
14W	2160 ± 120	0.158 ± 0.006	55.64 ± 0.92	39 ± 2	2 ± 3	1.1 ± 2.0
15W	1730 ± 80	-	50.31 ± 0.61	34 ± 2	23 ± 3	11.6 ± 1.5
16W	1420 ± 110	0.122 ± 0.005	37.29 ± 0.71	38 ± 3	0	0
17W	2380 ± 170	0.152 ± 0.006	51.12 ± 0.90	47 ± 3	0	0
18W	2800 ± 170	0.171 ± 0.008	85.50 ± 1.43	33 ± 2	32 ± 3	27.4 ± 2.6
19W	1610 ± 120	0.121 ± 0.005	48.78 ± 0.83	33 ± 3	24 ± 3	11.7 ± 1.5
20W	2460 ± 160	0.103 ± 0.010	52.74 ± 3.70	47 ± 5	2 ± 3	1.1 ± 1.9
21W	2000 ± 140	0.122 ± 0.007	56.26 ± 1.01	36 ± 3	10 ± 3	5.6 ± 1.7
22W	1600 ± 120	0.093 ± 0.005	46.51 ± 0.72	36 ± 3	22 ± 3	10.2 ± 1.4
23W	1790 ± 130	0.133 ± 0.004	43.94 ± 0.76	41 ± 3	2 ± 3	0.9 ± 1.6

TABLE 9 (cont'd)

AREAL INVENTORIES OF ^{137}Cs, ^{237}Np, $^{239+240}Pu$, AND $^{137}Cs/^{239+240}Pu$ ACTIVITY RATIOS IN INEEL SOILS, AND PERCENTAGE OF NON-FALLOUT (NF) Pu[†] AT THE CORING SITES

Core	Areal Inventories ± 1σ uncertainty (Bq m^{-2})					
	^{137}Cs	^{237}Np	$^{239+240}Pu$	Cs/Pu	% NF Pu	$^{239+240}Pu_{(NF)}$
24W	2430 ± 160	0.114 ± 0.043	81.20 ± 2.75	30 ± 2	26 ± 3	21.1 ± 2.5
25W	1770 ± 130	0.115 ± 0.003	39.27 ± 0.68	45 ± 3	0	0
26W	1680 ± 90	0.117 ± 0.004	54.46 ± 0.97	31 ± 2	25 ± 3	13.6 ± 1.7
27W	1910 ± 140	0.127 ± 0.018	57.60 ± 1.79	33 ± 3	19 ± 3	10.9 ± 1.8
55W	2480 ± 170	0.191 ± 0.006	57.03 ± 1.01	44 ± 3	0	0
56W	1830 ± 120	0.132 ± 0.003	62.51 ± 0.90	29 ± 2	34 ± 3	21.3 ± 1.9
57W	1250 ± 90	0.098 ± 0.003	34.05 ± 0.59	37 ± 2	5 ± 3	1.7 ± 1.0
58W	1940 ± 110	0.141 ± 0.004	43.80 ± 1.08	44 ± 3	0	0
60W	820 ± 80	0.069 ± 0.002	28.07 ± 0.58	30 ± 3	10 ± 3	2.8 ± 0.8
61W	1850 ± 120	0.126 ± 0.004	43.30 ± 0.73	43 ± 3	0	0
62W	2280 ± 130	0.149 ± 0.004	62.55 ± 1.55	37 ± 2	16 ± 3	10.0 ± 1.9
63W	2350 ± 150	0.113 ± 0.003	42.58 ± 0.89	55 ± 4	2 ± 3	0.9 ± 1.5
81W	1890 ± 110	0.140 ± 0.003	65.08 ± 0.87	29 ± 2	25 ± 3	16.3 ± 2.0
82W	2020 ± 120	0.140 ± 0.003	55.48 ± 0.79	36 ± 2	9 ± 3	5.0 ± 1.7
83W	1790 ± 110	0.140 ± 0.003	53.77 ± 0.71	33 ± 2	8 ± 3	4.3 ± 1.6
84W	2460 ± 130	0.120 ± 0.003	37.40 ± 0.45	66 ± 4	0	0
85W	610 ± 60	0.044 ± 0.001	27.31 ± 0.44	34 ± 2	36 ± 2	9.8 ± 0.6
92W	1400 ± 90	0.109 ± 0.002	47.04 ± 0.58	30 ± 2	16 ± 3	7.5 ± 1.4
28E	2150 ± 280	0.130 ± 0.016	63.02 ± 2.82	34 ± 5	11 ± 3	6.9 ± 1.9
29E	1890 ± 130	0.125 ± 0.007	44.44 ± 0.85	43 ± 3	4 ± 4	1.8 ± 1.6
30E	4330 ± 220	0.089 ± 0.005	38.80 ± 0.73	112 ± 6	20 ± 3	7.8 ± 1.2
31E	2100 ± 140	0.123 ± 0.009	61.73 ± 1.47	34 ± 2	6 ± 3	3.7 ± 1.9

TABLE 9(cont'd)

AREAL INVENTORIES OF ^{137}Cs, ^{237}Np, $^{239+240}Pu$, AND $^{137}Cs/^{239+240}Pu$ ACTIVITY RATIOS IN INEEL SOILS, AND PERCENTAGE OF NON-FALLOUT (NF) Pu[†] AT THE CORING SITES

Core	Areal Inventories ± 1σ uncertainty (Bq m^{-2})					
	^{137}Cs	^{237}Np	$^{239+240}Pu$	Cs/Pu	% NF Pu	$^{239+240}Pu_{(NF)}$
32E	1590 ± 90	0.109 ± 0.005	34.97 ± 0.68	45 ± 3	0	0
33E	2130 ± 120	0.131 ± 0.005	45.67 ± 0.74	47 ± 3	0	0
34E	2150 ± 120	0.154 ± 0.006	49.44 ± 0.96	44 ± 3	0	0
35E	2330 ± 140	0.139 ± 0.003	59.95 ± 1.06	39 ± 2	9 ± 3	5.4 ± 1.8
36E	2210 ± 140	0.116 ± 0.009	43.23 ± 0.86	51 ± 3	9 ± 3	3.9 ± 1.3
37E	12540 ± 680	0.171 ± 0.006	50.89 ± 0.72	246 ± 14	0	0
38E	1470 ± 90	0.119 ± 0.010	53.07 ± 1.14	28 ± 2	43 ± 2	22.8 ± 1.2
39E	2140 ± 140	0.113 ± 0.004	48.77 ± 0.72	44 ± 3	14 ± 3	6.8 ± 1.5
40E	1090 ± 90	0.090 ± 0.003	36.58 ± 0.65	30 ± 3	22 ± 3	8.0 ± 1.1
41E	2110 ± 130	0.165 ± 0.005	59.32 ± 0.98	36 ± 2	7 ± 3	4.2 ± 1.8
42E	1620 ± 120	0.097 ± 0.003	36.06 ± 0.61	45 ± 3	0	0
43E	1820 ± 130	0.136 ± 0.005	65.73 ± 0.98	28 ± 2	18 ± 3	11.8 ± 2.0
44E	1900 ± 120	0.128 ± 0.009	49.06 ± 0.97	39 ± 3	12 ± 3	5.9 ± 1.5
45E	2290 ± 130	0.152 ± 0.004	167.2 ± 2.6	14 ± 1	61 ± 2	102 ± 4
46E	2840 ± 160	0.163 ± 0.004	173.4 ± 2.6	16 ± 1	60 ± 2	104 ± 4
47E	1900 ± 120	0.121 ± 0.005	54.02 ± 0.84	35 ± 2	8 ± 3	4.3 ± 1.6
48E	1490 ± 130	0.112 ± 0.005	42.14 ± 0.65	35 ± 3	16 ± 3	6.7 ± 1.3
49E	1940 ± 130	0.144 ± 0.003	54.02 ± 0.84	36 ± 3	0	0
50E	2060 ± 120	0.153 ± 0.004	64.45 ± 0.94	32 ± 2	13 ± 3	8.4 ± 2.0
51E	1510 ± 100	0.087 ± 0.003	35.21 ± 0.49	43 ± 3	7 ± 3	2.5 ± 1.1
52E	2480 ± 160	0.131 ± 0.003	49.21 ± 0.80	51 ± 3	5 ± 3	2.5 ± 1.5
53E	1810 ± 130	0.118 ± 0.003	44.73 ± 0.65	41 ± 3	3 ± 3	1.2 ± 1.6

TABLE 9 (cont'd)

AREAL INVENTORIES OF ^{137}Cs, ^{237}Np, $^{239+240}Pu$, AND $^{137}Cs/^{239+240}Pu$ ACTIVITY RATIOS IN INEEL SOILS, AND PERCENTAGE OF NON-FALLOUT (NF) Pu† AT THE CORING SITES

Core	Areal Inventories ± 1σ uncertainty (Bq m^{-2})					
	^{137}Cs	^{237}Np	$^{239+240}Pu$	Cs/Pu	% NF Pu	$^{239+240}Pu_{(NF)}$
54E	10920 ± 570	0.220 ± 0.012	71.43 ± 1.65	154 ± 9	0	0
59E	1510 ± 90	0.130 ± 0.036	48.02 ± 0.91	31 ± 2	7 ± 3	3.4 ± 1.4
64E	1630 ± 120	0.105 ± 0.003	42.59 ± 0.92	38 ± 3	17 ± 3	7.2 ± 1.3
65E	1870 ± 120	0.118 ± 0.003	66.02 ± 1.07	28 ± 2	36 ± 2	23.8 ± 1.4
66E	2080 ± 120	0.145 ± 0.004	98.16 ± 1.40	21 ± 1	36 ± 2	35.3 ± 2.0
67E	1420 ± 110	0.111 ± 0.006	53.11 ± 0.87	27 ± 2	24 ± 3	12.7 ± 1.6
68E	1620 ± 120	0.110 ± 0.005	45.81 ± 0.69	35 ± 3	8 ± 3	3.7 ± 1.4
69E	3720 ± 210	0.147 ± 0.005	56.49 ± 0.79	66 ± 4	4 ± 3	2.3 ± 1.7
70E	6830 ± 120	0.588 ± 0.019	186.4 ± 3.2	37 ± 1	0	0
71E	1970 ± 130	0.149 ± 0.005	53.81 ± 0.77	37 ± 3	0	0
72E	2350 ± 150	0.164 ± 0.005	64.24 ± 1.02	37 ± 2	7 ± 3	4.5 ± 1.9
73E	4520 ± 240	0.217 ± 0.005	90.12 ± 1.38	50 ± 3	10 ± 3	9.0 ± 2.7
74E	2470 ± 140	0.155 ± 0.005	141.6 ± 2.00	17 ± 1	56 ± 2	79.3 ± 3.0
75E	1550 ± 120	0.108 ± 0.003	44.26 ± 0.65	35 ± 3	11 ± 3	4.9 ± 1.3
76E	1550 ± 120	0.115 ± 0.003	70.92 ± 0.93	22 ± 2	35 ± 3	24.8 ± 2.2
77E	1770 ± 110	0.140 ± 0.003	212.7 ± 2.50	8 ± 1	79 ± 1	167 ± 2
78E	2290 ± 120	0.190 ± 0.007	57.97 ± 0.90	40 ± 2	0	0
79E	1540 ± 110	0.127 ± 0.003	42.44 ± 0.64	36 ± 3	0	0
80E	2560 ± 150	0.206 ± 0.005	79.59 ± 1.15	32 ± 2	6 ± 3	4.8 ± 2.4
86E	1270 ± 100	0.117 ± 0.002	36.46 ± 0.56	35 ± 3	0	0
87E	2440 ± 170	0.191 ± 0.005	66.61 ± 1.29	37 ± 3	0	0
88E	2640 ± 150	0.195 ± 0.005	82.84 ± 1.56	37 ± 2	6 ± 3	5.0 ± 2.5

TABLE 9 (cont'd)

AREAL INVENTORIES OF ^{137}Cs, ^{237}Np, $^{239+240}Pu$, AND $^{137}Cs/^{239+240}Pu$ ACTIVITY RATIOS IN INEEL SOILS, AND PERCENTAGE OF NON-FALLOUT (NF) Pu[†] AT THE CORING SITES

Core	Areal Inventories ± 1σ uncertainty (Bq m^{-2})					
	^{137}Cs	^{237}Np	$^{239+240}Pu$	Cs/Pu	% NF Pu	$^{239+240}Pu_{(NF)}$
89E	1290 ± 90	0.104 ± 0.002	38.46 ± 0.77	34 ± 2	10 ± 3	3.8 ± 1.2
90E	2020 ± 120	0.185 ± 0.005	68.32 ± 1.33	30 ± 2	8 ± 3	5.5 ± 2.0
91E	3700 ± 200	0.159 ± 0.006	54.17 ± 0.89	68 ± 4	6 ± 3	3.3 ± 1.6
93E	810 ± 80	0.066 ± 0.002	24.32 ± 0.36	33 ± 3	3 ± 3	0.7 ± 0.7
94E	980 ± 60	0.082 ± 0.002	29.40 ± 0.36	33 ± 2	0	0
95E	2310 ± 150	0.165 ± 0.004	63.82 ± 1.15	36 ± 2	3 ± 4	1.9 ± 2.6
96E	2030 ± 120	0.179 ± 0.009	69.01 ± 4.05	29 ± 2	3 ± 4	2.1 ± 2.8

[†] Calculated using a $^{240}Pu/^{239}Pu_{(fallout)} = 0.166 \pm 0.004$ and $^{240}Pu/^{239}Pu_{(non\text{-}fallout)} = 0.055$

[*] Reference date: 1 January 1994; - not measured.

$^{137}Cs/^{239+240}Pu$ Inventory Activity Ratios

As stated earlier, in 1994 integrated global fallout should be characterized by a $^{137}Cs/^{239+240}Pu$ activity ratio of 38 ± 2. Figure 9 shows the relationship between the areal inventories of ^{137}Cs and $^{239+240}Pu$ and isolates those coring sites where the greatest deviation from linearity between these isotope inventories occurred. The locations closest to the ICPP (37E, 54E, and 69E; Figure 4), while showing elevated ^{137}Cs inventories *do not* evidence correspondingly high $^{239+240}Pu$ inventories. Cores taken near the SDA (45E, 46E), and downwind to some distance, show elevated $^{239+240}Pu$ inventories, but only modest ^{137}Cs inventories. Only location 70E shows both elevated ^{137}Cs and $^{239+240}Pu$ inventories but the Cs/Pu activity ratio is entirely consistent with that expected for integrated global fallout (37 ± 1; Table 9) which is confirmed by the $^{240}Pu/^{239}Pu$ atom ratio (0.168) measured there (Table C, Appendix 1). The most likely mechanism for producing elevated inventories of both ^{137}Cs and $^{239+240}Pu$ at this site is from the accumulation of fine-grained soils deposited there by wind erosion. In contrast, the soils at location 77E (Figure 4) show modest ^{137}Cs inventories, enhanced $^{239+240}Pu$ inventories (and consequently a low Cs/Pu activity ratio), and an unusually low $^{240}Pu/^{239}Pu$ atom ratio (0.073; Table C, Appendix 1). It is probable, therefore, that the origin of some fraction of the Pu at this site is of SDA origin deposited there by the same wind erosion processes that led to high ^{137}Cs and $^{239+240}Pu$ inventories at site 70E. This conclusion is supported by $^{241}Am/^{239}Pu$ atom ratio measurements discussed below.

The slope of the regression line in Figure 9 is 24 rather than the integrated fallout value of 38 principally because of the smaller and uncorrelated quantities of both ^{137}Cs and $^{239+240}Pu$ of INEEL origin. This is more clearly seen when the Cs/Pu inventory activity ratios are plotted versus the $^{240}Pu/^{239}Pu$ atom ratios at each site. Only a few locations fall within the expected 1σ uncertainties of the Cs/Pu global fallout ratio showing the influence of these releases across the site. These emissions raise the Cs/Pu activity ratio in a substantial number of cores that would otherwise show lower ratios because of "excess" Pu deposited there as evidenced by their lower $^{240}Pu/^{239}Pu$ atom ratios. This conclusion is further substantiated by plotting the estimated annual ^{137}Cs and $^{239+240}Pu$ releases from ICPP between 1953 and 1963 (Figure 10; DOE, 1991). From the slope of the regression line, the Cs/Pu activity ratio over this time period is > 3100, or more than two orders of magnitude higher than that expected from integrated global fallout. It is likely, therefore, that all of the onsite coring locations contain some ^{137}Cs from ICPP emissions.

$^{237}Np/^{239}Pu$ Atom Ratios and $^{237}Np/^{239+240}Pu$ Activity Ratios

The $^{237}Np/^{239}Pu$ atom ratio difference between northern hemisphere integrated global fallout (0.45), NTS fallout (≤ 0.03) and ICPP releases (»10) suggest that such ratios might be sensitive indicators of ICPP contributions of Pu in INEEL soils. That coring to 10 cm at the INEEL captures the entire ^{237}Np inventory can be seen from Figure 11 where the ^{237}Np and $^{239+240}Pu$ activity inventories are plotted for those INEEL cores whose $^{240}Pu/^{239}Pu$ atom ratios exceed 0.166, viz., those with little NTS fallout contribution. The slope of the regression line gives a $^{237}Np/^{239+240}Pu$ activity ratio (0.0031) that is identical to that measured in the Northern Tier, 30-cm deep soil cores, confirming the retention of ^{237}Np in the upper 10 cm of the soil column. Figure 11 illustrates another important point; unlike the ^{137}Cs data, cores collected nearest the ICPP (37E, 54E, 69E) give little indication that "excess" ^{237}Np has been deposited at

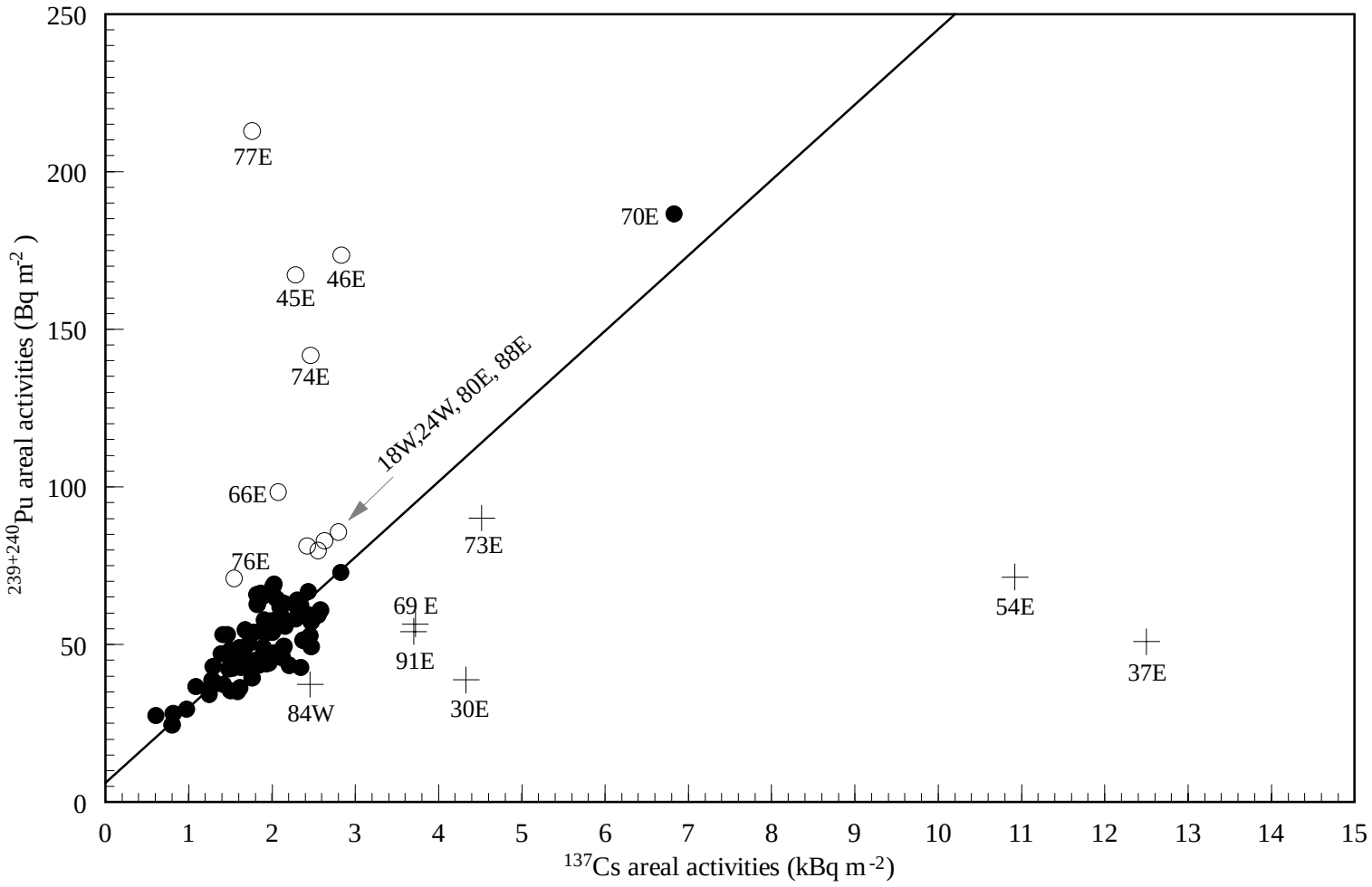

Figure 9. Correlation between $^{239+240}$Pu and ^{137}Cs areal activities for onsite cores at INEEL. Cores above the regression line (○) all have "excess" Pu with "low" ^{240}Pu/^{239}Pu atom ratios. Cores below regression line (+) show influence of ^{137}Cs contributions from ICPP. Regression line through points (●) represents the remainder of the coring sites.

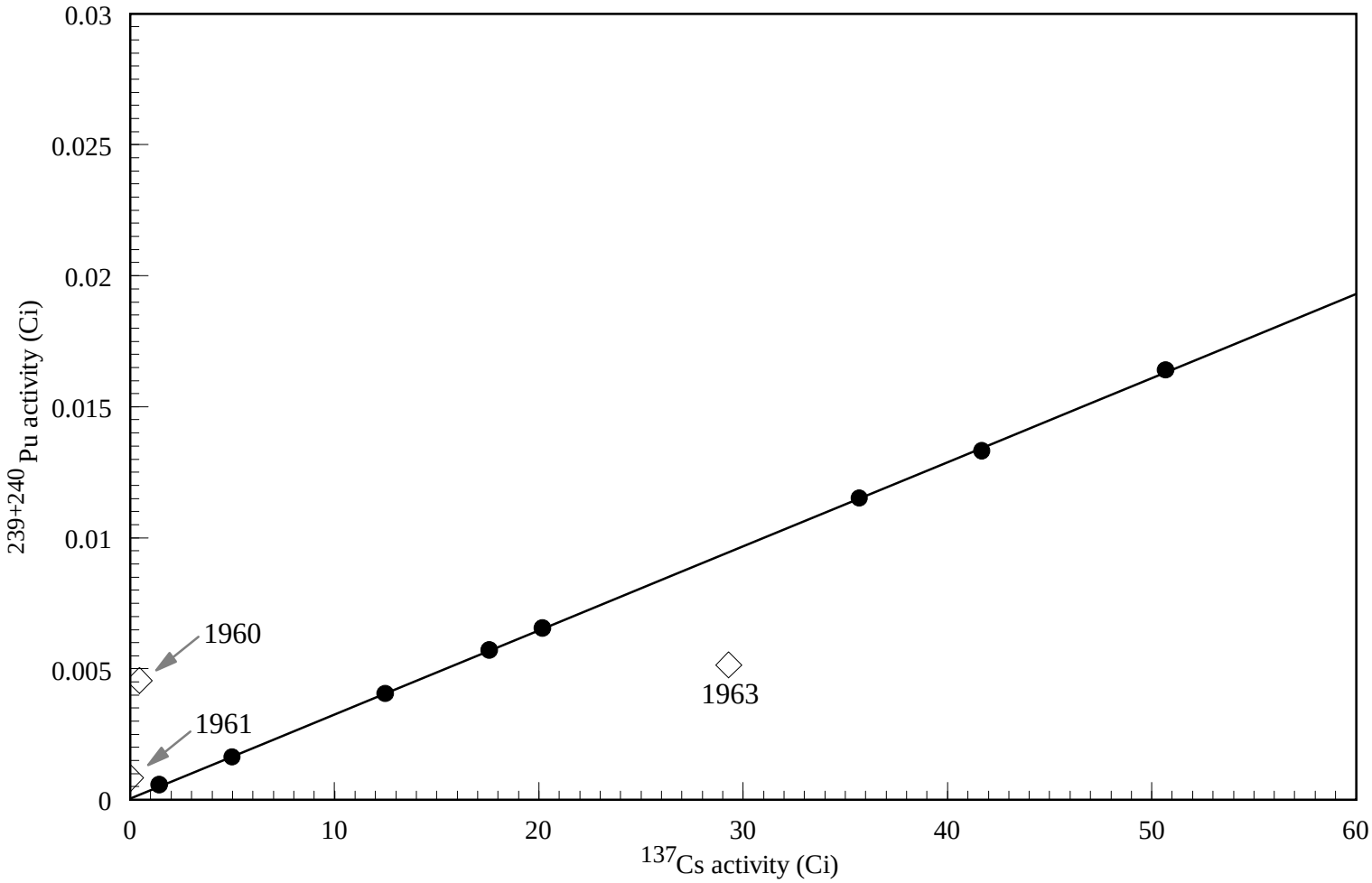

Figure 10. Cesium-137 and $^{239+240}$Pu activities in reconstructed radionuclide emissions from the ICPP for the years 1953 through 1963. The slope of the regression line (●) is 0.000320 giving a mean Cs/Pu activity ratio for those years of 3125 or more than two orders of magnitude larger than the same ratio for integrated global fallout.

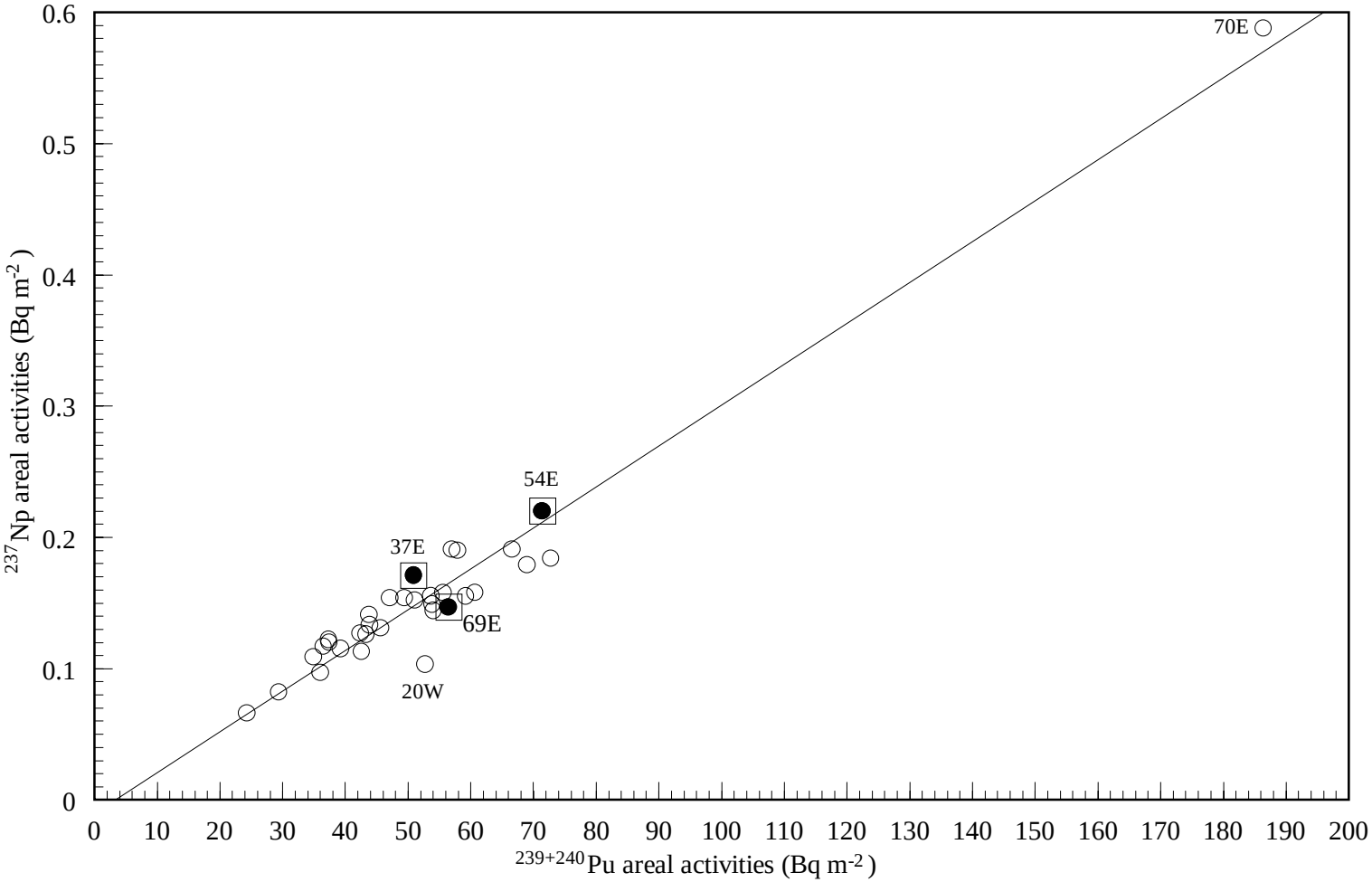

Figure 11. Correlation plot of ^{237}Np and $^{239+240}$Pu areal activities for those INEEL soil cores whose ^{240}Pu/^{239}Pu atom ratios exceed 0.166 suggesting a percent non-fallout Pu of zero. The correlation coefficient (r^2) for all data shown is 0.96 and the slope of the line equals 0.0031. The ^{237}Np/$^{239+240}$Pu activity ratio of integrated fallout in Northern Tier soils is 0.0031. Cores nearest ICPP (⊡) show little evidence of "excess" ^{237}Np .

these locations. The absence of enhanced ^{237}Np deposition across the INEEL can be further demonstrated by plotting the ^{237}Np areal inventories versus $^{239+240}$Pu areal inventories from which the non-fallout Pu contribution (Table 8) has been subtracted. Figure 12 shows such a plot for all onsite cores. When a ^{240}Pu/^{239}Pu atom ratio of 0.055 is used in calculating the percentage of non-fallout Pu present at each coring site (A), the slope of the Pu-corrected regression line is 0.0031, i.e., equal to that of global fallout. A number of cores fall below the regression line, but if it is assumed that the non-fallout ^{240}Pu/^{239}Pu atom ratio at these coring sites is 0.07 (within the range of isotopic material buried at the SDA) rather than 0.055, the corrected regression line (B) has a slope of 0.0030 and only three coring sites give evidence of Pu at these locations whose ^{240}Pu/^{239}Pu atom ratios exceed 0.07. Thus, within the analytical and sampling limitations discussed earlier, ^{237}Np inventories on the site are indistinguishable from those expected for integrated global fallout.

^{238}Pu/$^{239+240}$Pu and ^{241}Am/$^{239+240}$Pu Activity Ratios

Because of the assumed elevated concentrations of ^{238}Pu in ICPP airborne emissions over its operating history (DOE 1991), a selected number of cores were analyzed for this radionuclide and for ^{241}Am as further interrogators of the presence of nonfallout-derived transuranic radionuclides across the INEEL. However, it must be emphasized that the use of ^{238}Pu/$^{239+240}$Pu or ^{241}Am/$^{239+240}$Pu activity ratios to calculate the percentage of Pu or Am in INEEL soils from non-fallout sources suffers from two disadvantages. First, the uncertainties in the ^{238}Pu/$^{239+240}$Pu activity ratios (± ~ 50 percent) for historical ICPP releases introduce commensurately large uncertainties in any calculated ICPP Pu contributions. Second, there are no historical data for airborne ^{241}Am releases from the ICPP.

Table 10 lists the ^{238}Pu/$^{239+240}$Pu and ^{241}Am/$^{239+240}$Pu activity ratios for a number of cores analyzed by alpha spectrometry techniques. Of particular interest are the elevated ^{238}Pu/$^{239+240}$Pu activity ratios at coring sites 18W, 30E, 37E, 54E, and 69E. ^{240}Pu/^{239}Pu atom ratio measurements show non-fallout Pu contributions at 18W and 30E of 32 and 20 percent, respectively (Table 9), while the non-fallout contributions at sites 37E, 54E, and 69E are calculated to be zero. With the exception of core 18W, all other sites have Cs/Pu activity ratios that substantially exceed those expected from integrated global fallout which indicates the influence of airborne emissions from the ICPP. However, the extent of this influence is not large. Assuming that the ^{238}Pu and $^{239+240}$Pu activities at each site are derived only from a mixture of global fallout whose ^{238}Pu/$^{239+240}$Pu activity ratio equals 0.06 ± 0.01 and ICPP airborne emissions with an operationally constant ^{238}Pu/$^{239+240}$Pu activity ratio equal to 6.8 ± 3.5, the calculated percentages of ICPP-derived ^{238} Pu at 18W, 30E, 37E, 54E, and 69E are 1.0 ± 0.5, 2 ± 1, 10 ± 5, 10 ± 5, and 1.6 ± 0.9 percent, respectively. Reducing the assumed ^{238}Pu/$^{239+240}$Pu activity ratio of global fallout to 0.05 ± 0.01 does not change these percentages appreciably. Thus, while the TIMS ^{240}Pu/^{239}Pu atom ratio measurements argue for enhanced inventories of non-fallout Pu at 18W and 30E, the ^{238}Pu/$^{239+240}$Pu activity ratios measured at these sites imply that the ICPP contribution to their Pu inventories is < 3 percent.

ICPP Pu contributions to INEEL soils can also be demonstrated by comparing ^{238}Pu/^{239}Pu atom ratios, computed from the data of Table 10 and Table D, Appendix 1 with those expected from non-ICPP sources. Figure 13 shows a plot of the ^{238}Pu/^{239} versus ^{240}Pu/^{239} atom ratios for the

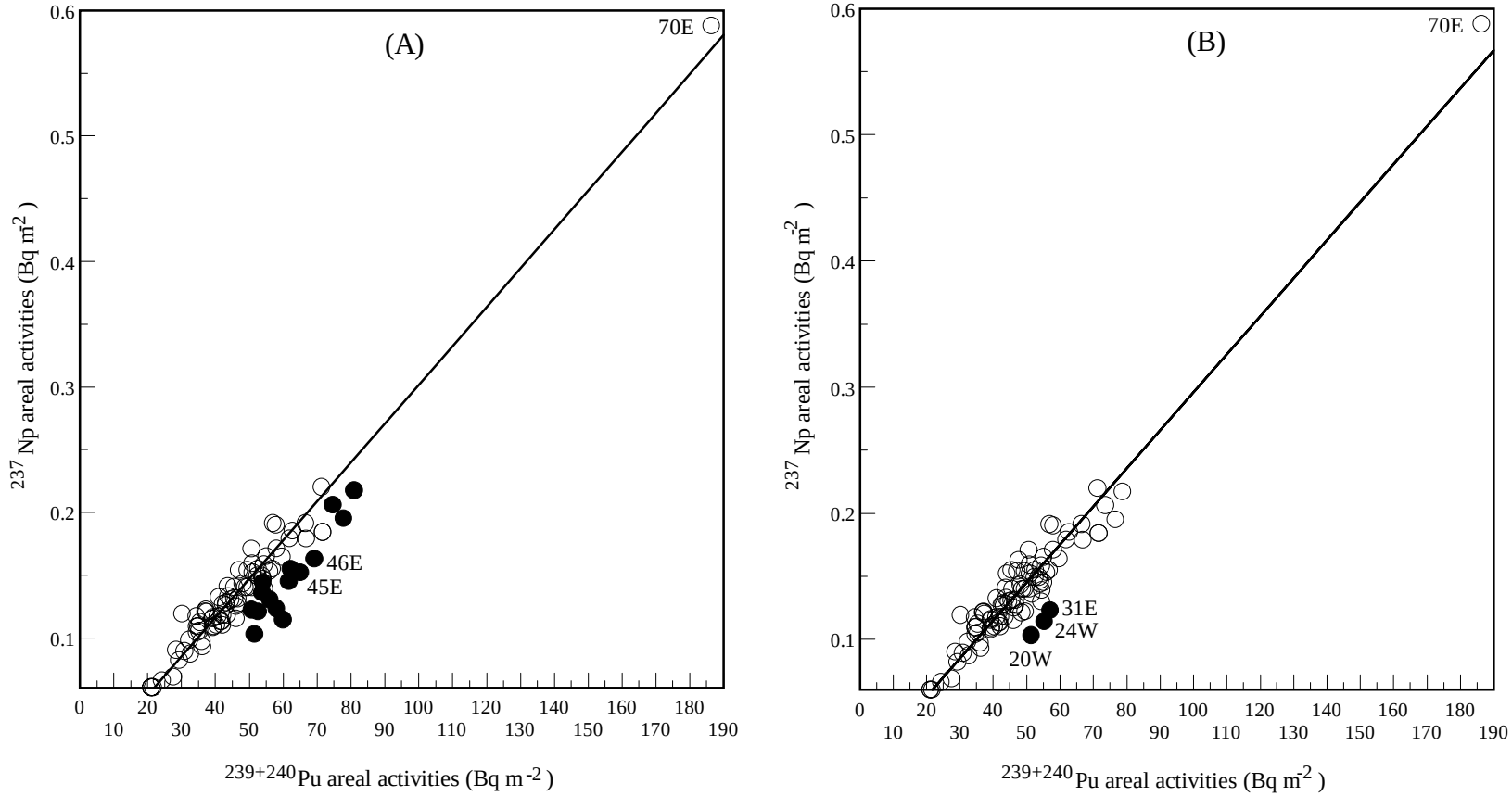

Figure 12. Correlation between ^{237}Np and $^{239+240}$Pu areal inventories when non-fallout Pu from the SDA is subtracted from the measured Pu inventory. Cores in (A) were corrected using a ^{240}Pu/^{239}Pu atom ratio of 0.055 for material from the SDA. Applying a ^{240}Pu/^{239}Pu ratio of 0.07 to those cores falling below the regression line in (A; ●) reduces the dispersion (B) suggesting that material of differing Pu isotopic ratios have been redistributed from the SDA. The majority of cores below the regression line in (A; ●) are down wind (NE direction) from cores 45E and 46E (near the SDA); cores 45E and 46E both show non-fallout contributions in their Pu inventories that exceed 70 percent. The slopes of the regression lines in (A) and (B) are 0.0031 and 0.0030, respectively, with correlation coefficients (r^2) for both plots ≥ 0.95.

TABLE 10

$^{238}Pu/^{239+240}Pu$ and $^{241}Am/^{239+240}Pu$ ACTIVITY RATIOS AT SELECTED INEEL LOCATIONS

Core	$^{238}Pu/^{239+240}Pu$	$^{241}Am/^{239+240}Pu$	Core	$^{238}Pu/^{239+240}Pu$	$^{241}Am/^{239+240}Pu$
18W	0.130 ± 0.009[†]	0.27 ± 0.02	43 E	0.08 ± 0.01	1.64 ± 0.08
19W	0.049 ± 0.005	0.26 ± 0.03	44E	0.062 ± 0.008	0.57 ± 0.04
26W	0.059 ± 0.006	0.91 ± 0.04	45E	0.036 ± 0.004	1.53 ± 0.05
27W	0.083 ± 0.007	0.33 ± 0.03	46E	0.028 ± 0.001	1.43 ± 0.03
55W	0.061 ± 0.005	0.29 ± 0.02	47E	0.062 ± 0.008	0.54 ± 0.04
56W	0.068 ± 0.007	0.35 ± 0.02	48E	0.066 ± 0.008	0.59 ± 0.04
57W	0.036 ± 0.004	0.29 ± 0.02	50E	0.066 ± 0.008	0.94 ± 0.05
58W	0.050 ± 0.005	0.37 ± 0.02	51E	0.06 ± 0.01	0.40 ± 0.03
81W	0.047 ± 0.008	0.41 ± 0.04	52E	0.064 ± 0.007	0.32 ± 0.03
85W	0.041 ± 0.006	0.29 ± 0.03	54E	0.72 ± 0.06	0.42 ± 0.03
92W	0.046 ± 0.007	0.32 ± 0.02	64E	0.032 ± 0.004	0.29 ± 0.02
28E	0.058 ± 0.008	0.37 ± 0.02	65E	0.051 ± 0.005	0.26 ± 0.02
29E	0.08 ± 0.01	0.27 ± 0.03	66E	0.073 ± 0.008	0.28 ± 0.02
30E	0.21 ± 0.02	0.47 ± 0.03	67E	0.12 ± 0.01	0.36 ± 0.05
32E	0.032 ± 0.006	0.26 ± 0.03	68E	0.10 ± 0.01	0.35 ± 0.03
33E	0.084 ± 0.009	0.32 ± 0.02	69E	0.17 ± 0.02	0.32 ± 0.02
34E	0.062 ± 0.008	0.33 ± 0.03	70E	0.050 ± 0.004	0.334 ± 0.013
35E	0.044 ± 0.005	0.38 ± 0.02	73E	0.133 ± 0.009	0.37 ± 0.02
36E	0.11 ± 0.01	0.43 ± 0.03	74E	0.077 ± 0.011	0.28 ± 0.02
37E	0.72 ± 0.07	0.44 ± 0.06	75E	0.040 ± 0.006	0.38 ± 0.03
38E	0.062 ± 0.008	0.26 ± 0.02	76E	0.048 ± 0.006	0.29 ± 0.02
39E	0.051 ± 0.008	0.24 ± 0.03	77E	0.049 ± 0.004	0.334 ± 0.023
40E	0.03 ± 0.01	0.35 ± 0.05	78E	0.088 ± 0.011	0.343 ± 0.025
42E	0.045 ± 0.005	0.21 ± 0.02	91E	0.052 ± 0.006	0.26 ± 0.02

[†]1σ propagated uncertainties from counting rate data only. Reference date: 1 January 1998.

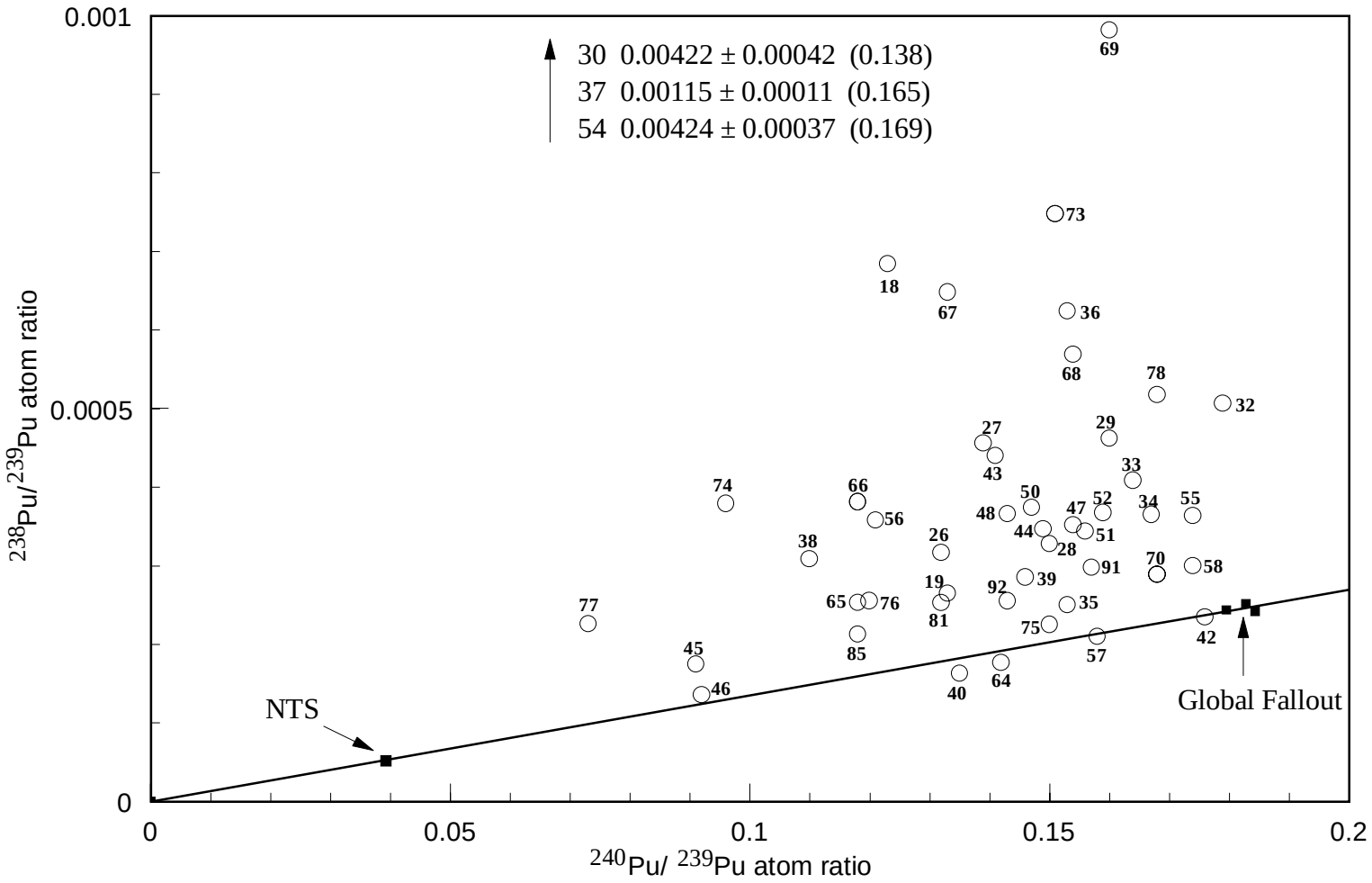

Figure 13. ^{238}Pu/^{239}Pu vs.^{240} Pu/^{239} Pu atom ratios for selected coring sites at the INEEL in 1995. Global fallout values represent those measured in North Eastham, MA, Conoy Township, PA., and West Donegal, PA. (Krey and Beck 1981). Uncertainties in the ^{238}Pu/ ^{239}Pu atom ratios are ≤ 10 percent while those for the ^{240}Pu/ ^{239}Pu atom ratios are ≤ 2 percent.

cores listed in Table 10; the line represents Pu isotopics in mixtures where the end members are global and NTS-derived fallout. As is evident, the great majority of cores lie above the mixing line indicating the presence of small, but discernible, amounts of ICPP-derived ^{238}Pu at the sites sampled. However, because the ICPP-derived ^{238}Pu at 18W is calculated to be only 1.0 ± 0.5 percent, the cores lying below 18W in Figure 13 have proportionately less ^{238}Pu than this amount.

While the $^{238}Pu/^{239+240}Pu$ activity ratios at sites 30E, 37E, 54E, and 69E show higher ICPP contributions to their Pu inventories than for all other cores listed in Table 10, their $^{240}Pu/^{239}Pu$ atom ratios (Table C) argue for a mixture of NTS and global fallout-derived Pu only. The resolution of this apparent discrepancy rests on the value of the $^{240}Pu/^{239}Pu$ atom ratio assigned to the non-fallout component of the Pu inventory. From discussions presented earlier, if the $^{240}Pu/^{239}Pu$ atom ratio of ICPP-derived Pu is assigned a value of 0.045, then small additions of Pu to INEEL soils from this source would be indistinguishable from that attributed to Pu derived from the SDA. However, if the $^{240}Pu/^{239}Pu$ atom ratio from ICPP airborne emissions substantially exceeds that of Pu from the SDA (0.055), then it is entirely possible for ICPP *and* SDA-derived Pu to combine to produce Pu soil inventories where the resultant $^{240}Pu/^{239}Pu$ atom ratio (~ 0.166) is identical to that resulting from global fallout and NTS input only. It is also possible that both low $^{240}Pu/^{239}Pu$ (0.045) and high $^{240}Pu/^{239}Pu$ atom ratio material (~ 0.25) from ICPP and Pu from the SDA could combine to produce the $^{240}Pu/^{239}Pu$ ratios of 0.165 and 0.166. Such an interpretation is consistent with what appears to be a slight increase in ^{237}Np inventories recorded at 37E and 54E over nearby sites (Table 9) as well as the enhanced $^{241}Pu/^{239}Pu$ atom ratios observed at these locations (Table 6). This latter interpretation also explains the finding of *low* $^{240}Pu/^{239}Pu$ atom ratio material in the smaller core samples taken near 54E (Table 5). If correct, future analysis of small aliquots of soil from these single cores should occasionally produce $^{240}Pu/^{239}Pu$ ratios significantly higher than 0.166 indicating the presence of high $^{240}Pu/^{239}Pu$ atom ratio particles of ICPP origin.

The elevated $^{241}Am/^{239+240}Pu$ activity ratios observed in cores 45E and 46E near the SDA (Table 10; Figure 4) suggests that this material is not representative of weapons-grade Pu, even when production of ^{241}Am from ^{241}Pu decay is taken into account. For example, Markham et al. (1978) listed cumulative buried inventories of ^{241}Pu and $^{239+240}Pu$, through 1970, as 6.48 PBq (1.75×10^5 Ci) and 777 TBq (2.1×10^4 Ci), respectively. Assuming a $^{240}Pu/^{239}Pu$ atom ratio of 0.055 as representative of material buried at the SDA, the number of ^{241}Pu and ^{239}Pu atoms represented by these activities is 4.24×10^{24} and 7.09×10^{26} atoms, respectively. It is clear that the $^{241}Am/^{239}Pu$ atom ratio could never rise above 0.006 if the ^{241}Am in the buried waste at the SDA derived exclusively from ^{241}Pu decay; indeed the ratio must be less than this value owing to the decay of ^{241}Am following its production over time. This fact is further evident from Figure 14 which shows the expected $^{241}Am/^{239}Pu$ versus $^{240}Pu/^{239}Pu$ atom relationship (inset) in graphite-moderated reactors fueled with natural uranium (Croff 1980), i.e., like the production reactors at Hanford. The $^{241}Am/^{239}Pu$ atom ratios observed at sites 43E, 45E, 46E, and 50E are more than two orders of magnitude greater than would be expected in weapons-grade Pu ($^{240}Pu/^{239}Pu$ atom ratio ≤ 0.05). This pronounced enrichment in ^{241}Am suggests that its origin is from fuel reprocessing waste where chemical separation has increased its concentration in the process waste stream.

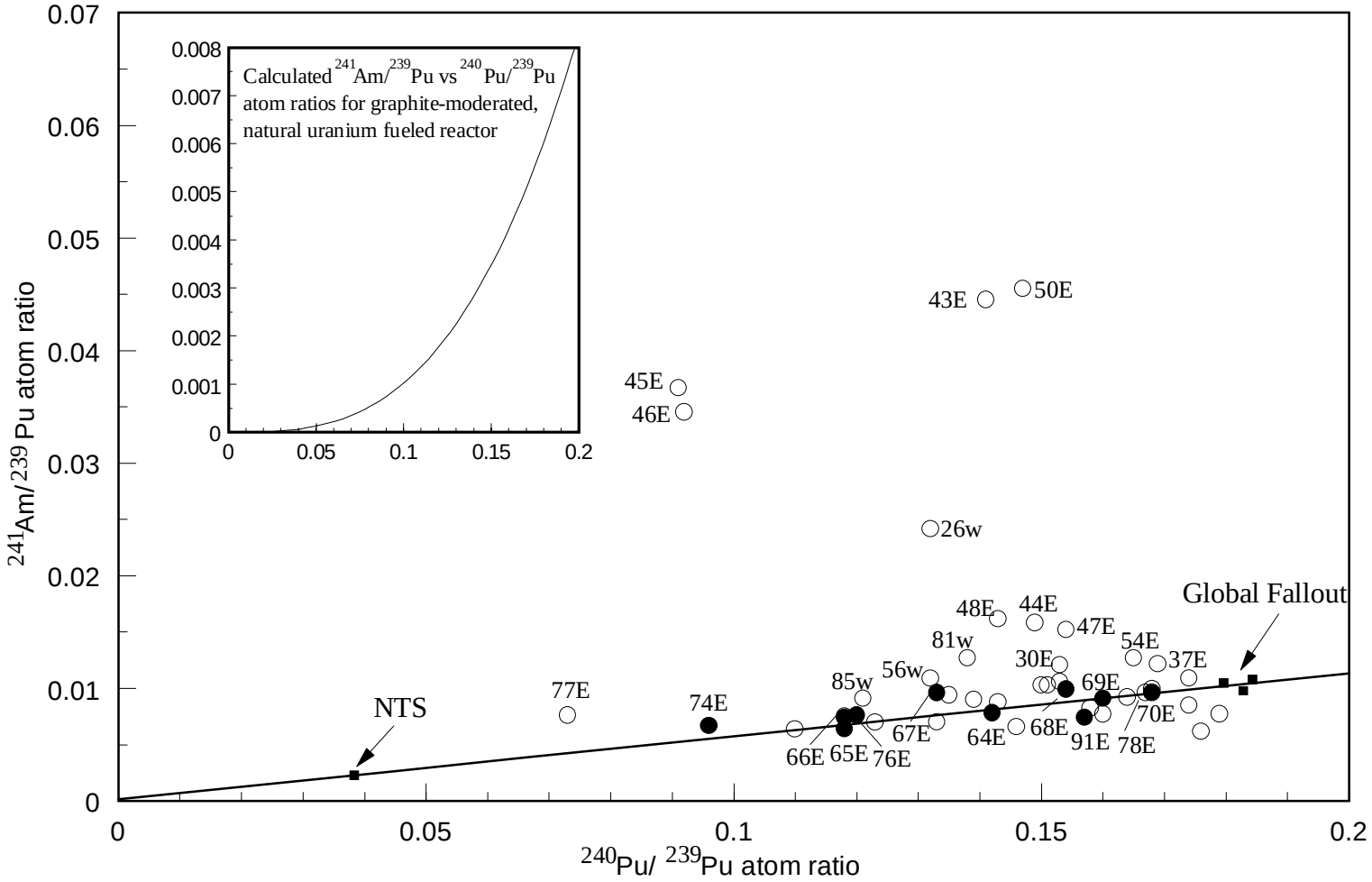

Figure 14. $^{241}Am/^{239}Pu$ vs. $^{240}Pu/^{239}Pu$ atom ratios for selected cores at the INEEL. Global fallout values represent those measured at North Eastham, MA, Conoy Township, PA, and West Donegal, PA (Krey and Beck 1981). Inset shows the expected atom ratios for Pu produced in graphite-moderated reactors using natural uranium as fuel (Croff 1980). Uncertainties in the $^{241}Am/^{239}Pu$ atom ratios are ≤10 percent while those for the $^{240}Pu/^{239}Pu$ atom ratios are ≤ 2 percent.

Figure 14 also shows the two end-member mixing curve for NTS and global-fallout derived $^{241}Am/^{239}Pu$ and $^{240}Pu/^{239}Pu$ atom ratios as of 1995. Those coring sites which lie above the mixing curve contain contributions of the "excess" ^{241}Am confirming transport of this material to sites 77E, 81W and 85W. Coring sites lying on or near the mixing line cannot be unequivocally assigned to fallout sources because of the close similarity in the atom ratios of NTS fallout and Rocky Flats waste.

Prior to the Pu alpha spectrometry measurements at sites 64 through 67E, it was thought that the elevated, non-fallout Pu in these cores might be due to material dispersed there from the waste solvent burning incident at the ICPP in 1959 (DOE 1991). As process material, ^{237}Np in the waste solvent might well have been depleted relative to Pu resulting in the low $^{237}Np/^{239}Pu$ atom ratios measured at these sites. However, the $^{238}Pu/^{239+240}Pu$ activity ratios for *all* cores from 64E through 91E (Table 10) are of such magnitude as to preclude any consequential contribution of ICPP-derived Pu to these soils.

INVENTORY OF NON-FALLOUT Pu AT THE INEEL

From the data of Table 9 it is possible to broadly approximate the non-fallout inventory of Pu over the area sampled in this research. As discussed above, the dynamic nature of the soil redistribution processes occurring at the INEEL do not produce "textbook" gradients away from point sources of above-ground contamination, especially over decadal time scales. Consequently, attempts at detailed isoplething of the non-fallout inventories shown in Table 9 are not useful. It is more economical of time and thought to broadly constrain these inventories into selected numerical ranges and to then judge the most appropriate values to be used for inventory calculations. Figure 15 shows such a plot from the data of Table 9 and indicates that the great majority of cores have non-fallout Pu excesses in the range of 2 to 10 Bq m^{-2}; a smaller number of cores situated to the south and west of the RWMC show slightly higher excess inventories in the range of 10 to 23 Bq m^{-2}. Isolated cores within the bounded area shown in Figure 15 show "hot spots" where excess Pu inventories exceed 25 Bq m^{-2}, but their areal extent is not large. It seems reasonable, therefore, to constrain the non-fallout Pu inventory estimate to the integration of the two areas bounded by the areas delineated in Figure 15.

The entire area bounded by the 2-10 Bq m^{-2} isopleth is ~ 9.8 x 10^8 m^2 while that bounded by the 10-23 Bq m^{-2} isopleth is ~ 5.0 x $10^7 m^2$. Assigning a mean $^{239+240}Pu$ inventory of 5 Bq m^{-2} to the area bounded by the 2-10 Bq m^{-2} isopleth gives a non-fallout Pu inventory of 4.9 GBq or ~ 130 mCi. Assigning a mean $^{239+240}Pu$ inventory of 15 Bq m^{-2} to the area bounded by the 10-23 Bq m^{-2} isopleth adds an additional 0.75 GBq (~ 20 mCi) of non-fallout Pu bringing the total to ~ 5.7 GBq or ~ 150 mCi. This quantity could, and most probably does, represent an over-estimate of the amount of $^{239+240}Pu$ which has migrated from the SDA because an SDA origin is assigned to *all* sites where low $^{240}Pu/^{239}Pu$ atom ratio material was detected.

Excluding cores 37E, 54E, and 70E, the 23 cores showing only fallout Pu input (Table 9) have average inventories of 48 ± 12 Bq m^{-2}. Assuming that this is representative of the entire 9.8 x 10^8 m^2 shown in Figure 15, the fallout Pu in this area is approximately 40 ± 10 Gbq (~ 1.1 ± 0.3 Ci). Thus, non-fallout Pu would have increased the Pu inventory from global and regional fallout by ~ 14 percent.

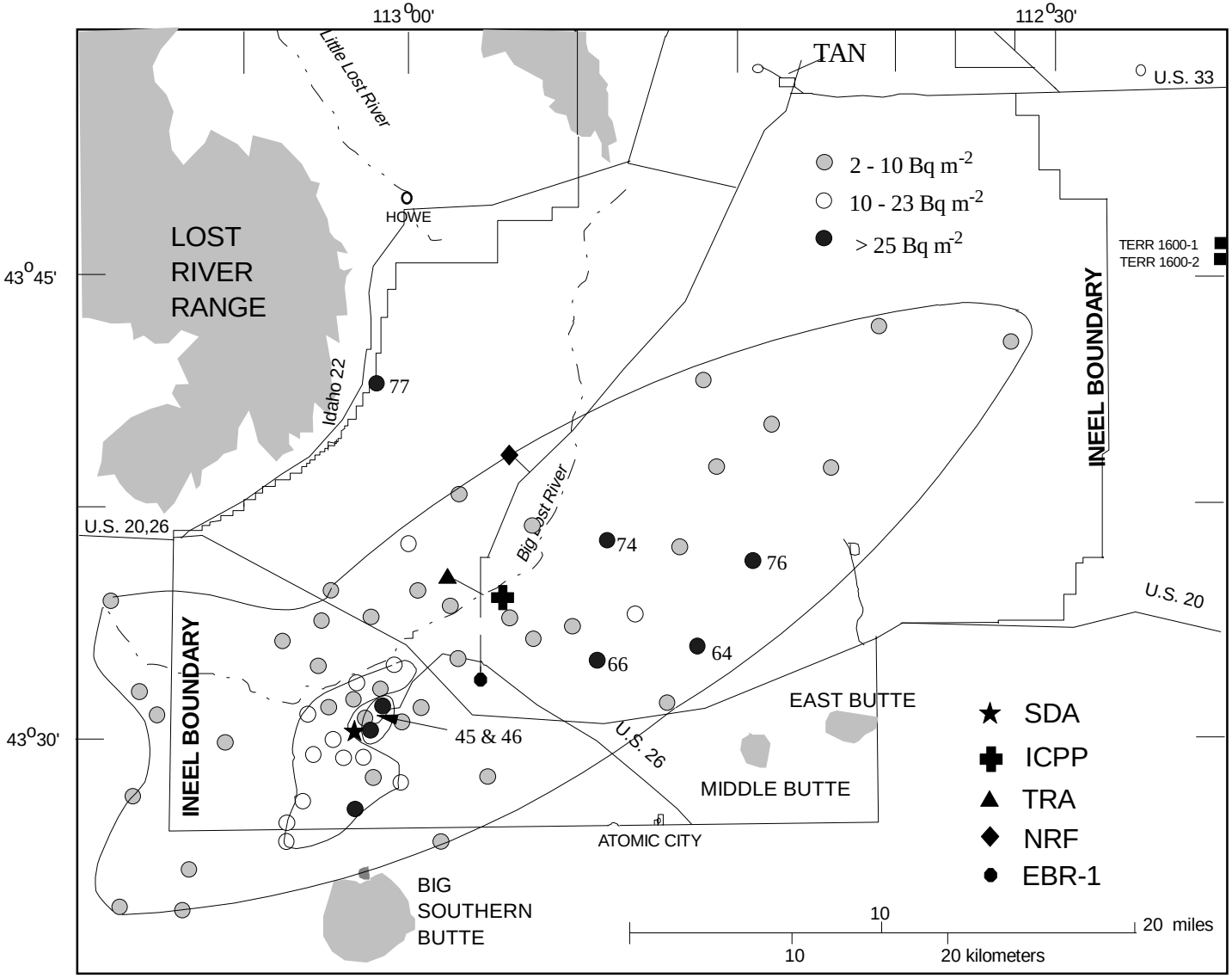

Figure 15. Isopleths of SDA-derived $^{239+240}Pu$ at the INEEL. Where areal activities exceed 25 Bq m^{-2}, the highest inventories were observed at coring sites 45,46,74, and 77.

CONCLUSIONS

The data contained in this report represents the first effort to map the isotopic Pu signatures in onsite soils at, and near, the INEEL. It is clear from a review of past measurements using only alpha spectrometry techniques, that thermal ionization mass spectrometry measurements provide a far higher information content regarding the origin of Pu on the site than has been available in the past. As such, the present data base provides the necessary "background" information against which any future fugitive releases of Pu from the SDA can be assessed.

Using a combination of thermal ionization mass spectrometry and alpha spectrometry techniques, a partial deconvolution of the contributions of non-fallout Pu from the SDA and the ICPP to INEEL soils has been attempted. Because monitoring data for airborne Pu emissions from the ICPP during the 1950s and 1960s do not exist, calculational partitioning of ICPP inputs rests with the assumption of a time-invariant $^{238}Pu/^{239+240}Pu$ activity ratio (6.8 ± 3.5) determined in stack effluents characteristic of plant operations in the mid-to-late 1970s. At 47 coring sites sampled in this study all showed evidence of measurable ^{238}Pu that could be ascribed to releases from the ICPP. It must be emphasized that ascribing virtually all of the non-fallout Pu in INEEL soils to the SDA is subject to revision if future analyses of curatorial samples should show that the early $^{238}Pu/^{239+240}Pu$ activity ratios in stack emissions from the ICPP were substantially lower. In the absence of such analyses, another interrogator of ICCP emissions will be required and that interrogator might well be the presence of ^{236}U in INEEL soils.

In addition to the ^{236}U measured in the monitoring well waters listed in Table 2, material from core 54E (composite) was analyzed for this radionuclide using TIMS to establish its presence where elevated $^{238}Pu/^{239+240}Pu$ activity ratios indicated the existence of ICPP-derived Pu. The measured $^{235}U/^{238}U$ atom ratios in soil aliquots (1.29 and 1.24 g dry) from this composite core were 0.007296 ± 0.000011 and 0.007292 ± 0.000015, indicating the presence of excess ^{235}U in the soils (Table 2). The ^{236}U atom concentration was ~ (8 ± 1) x 10^8 atoms g^{-1} dry. Such measurements require the most sophisticated mass spectrometers in operation today in order to detect and distinguish these low atom concentrations in the presence of the much larger amounts (~ 2 x 10^{16} atoms g^{-1} dry) of ^{238}U in the same sample size. However, recent developments in the measurement of U isotopes using accelerator mass spectrometry techniques (Zhao et al.1997) hold the promise of being able to detect low atom concentrations of ^{236}U without the limitations that accompany its measurement using TIMS. Because ^{236}U has not yet been shown to be a measurable component of global fallout debris, its presence in INEEL soils might well be a unique tracer for ICPP airborne releases and its usefulness in this respect should be actively explored.

Finally, time and resources constrained the total number of soil samples that could be analyzed in the present study and necessarily prevented a more extensive sampling in areas contaminated by the waste solvent burning (1959) and Fuel Element Cutting Facility (1958) releases of radioactivity described elsewhere in this report. The authors recommend that serious consideration be given to studying the areas known to have been contaminated by these events to determine both the inventory and isotopic signatures of the transuranic elements deposited there.

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APPENDIX 1

Table A. Onsite core locations as determined by Global-Positioning System measurements.
Table B. Soil core weights from collections in ID, MT, WY and the INEEL.
Table C. Ratios of ^{237}Np and Pu isotopes in soil cores collected on the INEEL.
Table D. Atom concentrations of ^{237}Np and Pu in soil cores collected on the INEEL.

TABLE A

ONSITE AND NEAR-SITE CORE LOCATIONS AS DETERMINED BY GLOBAL-POSITIONING SYSTEM (GPS) MEASUREMENTS. VALUES ARE ROUNDED TO THE NEAREST SECOND

Core	Latitude	Longitude	Core	Latitude	Longitude
1W	43 24 14	112 59 53	23W	43 30 48	113 04 56
2W	43 24 59	113 01 27	24W	43 30 07	113 04 21
3W	43 25 43	113 04 25	25W	43 29 48	113 03 57
4W	43 26 09	113 06 22	26W	43 29 31	113 03 20
5W	43 25 41	113 09 12	27W	43 28 43	113 02 11
6W	43 25 29	113 12 03	28E	43 32 51	112 54 07
7W	43 34 22	113 13 56	29E	43 31 36	112 55 46
8W	43 31 15	113 12 09	30E	43 32 21	112 57 44
9W	43 29 35	113 08 07	31E	43 34 25	113 03 22
10W	43 27 41	113 04 42	32E	43 34 46	113 04 47
11W	43 26 22	113 01 21	33E	43 32 50	112 59 50
12W	43 26 24	112 58 17	34E	43 35 54	113 03 16
13W	43 26 30	112 56 03	35E	43 34 25	112 59 20
14W	43 31 00	113 06 32	36E	43 33 50	112 57 58
15W	43 29 12	113 04 10	37E	43 33 23	112 56 34
16W	43 29 58	113 05 08	38E	43 35 59	112 59 54
17W	43 28 07	113 03 28	39E	43 33 33	113 01 30
18W	43 27 29	113 02 22	40E	43 32 44	113 05 13
19W	43 28 17	113 00 11	41E	43 31 59	113 03 51
20W	43 28 53	113 00 54	42E	43 31 28	113 04 41
21W	43 28 32	113 01 38	43E	43 31 37	113 02 16
22W	43 28 57	113 01 41	44E	43 31 11	113 03 24

TABLE A (cont'd)

ONSITE AND NEAR-SITE CORE LOCATIONS AS DETERMINED BY GLOBAL-POSITIONING SYSTEM (GPS) MEASUREMENTS. VALUES ARE ROUNDED TO THE NEAREST SECOND

Core	Latitude	Longitude	Core	Latitude	Longitude
45E	43 30 34	113 01 15	67E	43 33 29	112 49 27
46E	43 30 15	113 01 57	68E	43 33 36	112 52 22
47E	43 31 15	113 01 05	69E	43 33 55	112 55 33
48E	43 31 09	113 02 02	70E	43 39 19	113 03 36
49E	43 30 42	113 02 11	71E	43 38 21	113 00 26
50E	43 30 29	113 02 40	72E	43 37 28	112 57 25
51E	43 30 42	112 59 58	73E	43 36 23	112 54 10
52E	45 30 36	112 59 15	74E	43 35 56	112 50 46
53E	43 29 52	112 56 11	75E	43 35 37	112 47 27
54E	43 33 13	112 57 17	76E	43 35 14	112 44 01
55W	43 26 58	113 03 23	77E	43 41 25	113 01 56
56W	43 26 58	113 05 17	78E	43 41 42	112 56 01
57W	43 26 58	113 07 38	79E	43 39 58	112 52 08
58W	43 28 20	113 06 50	80E	43 38 11	112 45 41
59E	43 42 46	112 38 16	81W	43 26 58	113 05 16
60W	43 33 31	113 03 42	82W	43 24 11	113 09 51
61W	43 32 42	113 02 06	83W	43 24 19	113 12 52
62W	43 31 57	113 00 25	84W	43 26 55	113 12 30
63W	43 30 40	112 58 04	85W	43 27 52	113 12 17
64E	43 30 39	112 48 00	86E	43 43 26	112 54 33
65E	43 32 24	112 46 39	87E	43 42 01	112 49 39
66E	43 32 02	112 51 20	88E	43 41 06	112 46 23

TABLE A (cont'd)

ONSITE AND NEAR-SITE CORE LOCATIONS AS DETERMINED BY GROUND-POSITIONING SYSTEM (GPS) MEASUREMENTS. VALUES ARE ROUNDED TO THE NEAREST SECOND

Core	Latitude	Longitude	Core	Latitude	Longitude
89E	43 39 34	112 43 16	93E	43 43 30	112 50 18
90E	43 38 08	112 40 28	94E	43 43 42	112 47 02
91E	43 28 25	112 56 11	95E	43 41 28	112 40 16
92W	43 30 57	113 11 23	96E	43 42 10	112 32 06

TABLE B

PERCENTAGE OF TOTAL INITIAL WEIGHT IN FINAL 0.5 MM FRACTION OF PROCESSED SOILS COLLECTED IN IDAHO, MONTANA, WYOMING, AND INEEL.[b]

Location	Weight (g)	% total weight*	Location	Weight (g)	% total weight*
Carey, ID[a]	1340.0	96.9	1W	1332.3	71.7
Challis, ID[b]	1379.6	96.9	2W	1470.9	71.7
Howe, ID[a]	1246.6	89.4	3W	1935.8	96.0
Island Park, ID[b]	1792.7	95.3	4W	1856.8	94.3
Richfield, ID[a]	1486.2	95.6	5W	1857.3	91.5
Salmon, ID[b]	1851.4	98.1	6W	1919.4	96.1
Terreton, ID (1)[a]	1987.0	89.5	7W	1623.8	89.3
Terreton, ID (1[§])[b]	2231.5	96.0	8W	1753.7	92.4
Terreton, ID (2[¶])[b]	2160.0	93.0	9W	2121.8	98.7
Darby, MT[b]	1238.8	78.8	10W	2000.9	90.9
Ennis, MT[a]	490.8	95.6	11W	1930.6	94.4
Ennis, MT [‡b]	1944.6	87.0	12W	1997.8	95.3
Raynolds Pass, MT[a]	148.7	56.0	13W	1849.1	94.0
Basin, WY[a]	566.6	92.9	14W	2266.3	96.4
Cody, WY[a]	509.2	99.0	15W	1999.4	91.8
Daniel, WY[a]	541.9	95.9	16W	2168.2	96.5
DuBois, WY[a]	541.1	88.1	17W	2172.8	95.8
Ethete, WY[a]	455.0	90.7	18W	1939.9	87.0
Fontanelle, WY[a]	391.7	99.3	19W	2206.3	97.8
Meeteetse, WY[a]	351.5	78.8	20W	2004.3	98.0
Thermopolis, WY[a]	337.5	86.3	21W	2141.0	94.3
Worland, WY[a]	393.1	94.3	22W	1890.1	95.0
Yellowstone, WY[a]	188.0	85.1	23W	2330.6	95.7

TABLE B (cont'd)

PERCENTGE OF TOTAL INITIAL WEIGHT IN FINAL 0.5MM FRACTION OF PROCESSED SOILS COLLECTED IN IDAHO, MONTANA, WYOMING, AND AT INEEL[b]

Location	Weight (g)	% total weight*	Location	Weight (g)	% total weight*
24W	2050.9	96.2	47E	2121.9	95.2
25W	2063.9	96.8	48E	2077.4	92.8
26W	1840.9	93.8	49E	2062.4	96.4
27W	2140.5	95.7	50E	1985.6	96.4
28E	2125.3	94.7	51E	2018.2	96.8
29E	2112.6	95.1	52E	1993.5	95.4
30E	1900.4	84.5	53E	2003.0	97.0
31E	2053.2	97.4	54E (comp)	1864.0	95.1
32E	2031.0	97.1	54E (R-1)[c]	525.9	95.1
33E	1514.4	70.4	54E (R-2)[c]	504.7	95.1
34E	1957.2	96.4	54E (R-3)[c]	366.6	92.7
35E	1485.1	76.0	54E (R-4)[c]	535.3	94.1
36E	1820.2	93.7	55W	2033.2	97.8
37E	1754.1	79.7	56W	2029.1	96.7
38E	2282.5	90.8	57W	1834.9	90.7
39E	2300.6	95.5	58W	1875.0	96.9
40E	2127.7	94.9	59E	2278.2	93.0
41E	2042.1	97.1	60W	1988.6	82.7
42E	2438.5	96.3	61W	2248.0	95.0
43E	2109.7	97.7	62W	1347.6	59.9
44E	2071.6	96.0	63W	2202.4	95.4
45E	1920.5	93.7	64E	1999.2	97.2
46E	1884.7	94.0	65E	1839.2	91.4

TABLE B (cont'd)

PERCENTGE OF TOTAL INITIAL WEIGHT IN FINAL 0.5 MM FRACTION OF PROCESSED SOILS COLLECTED IN IDAHO, MONTANA, WYOMING, AND AT INEEL[b]

Location	Weight (g)	% total weight*	Location	Weight (g)	% total weight*
66E	2394.0	93.1	82W	1993.4	92.1
67E	2293.4	92.5	83W	1764.4	88.5
68E	2167.6	85.6	84W	1789.2	89.5
69E	1618.8	79.4	85W	2008.8	96.2
70E	1946.2	97.6	86E	2217.3	90.4
71E	2350.7	94.7	87E	2240.8	95.9
72E	1820.5	98.4	88E	2173.3	98.8
73E	1938.2	93.0	89E	2447.1	96.3
74E	2397.4	97.1	90E	2095.4	98.7
75E	2067.9	89.8	91E	2230.3	95.7
76E	2104.6	87.4	92W	1856.8	83.3
77E	1894.4	91.4	93E	2219.4	97.6
78E	2191.4	96.7	94E	2163.4	93.6
79E	1950.2	97.7	95E	2385.6	89.9
80E	2378.2	96.0	96E	2304.5	92.7
81W	1902.8	92.2			

Represents percentage of core weight passing < 0.5 mm diameter sieve (see text); [a]cores taken with EML corer with area = 61 cm^2. Cores with weights >500 g represent four cores taken at the corners of an ~ 1 m square, while cores with weights ~ 500g represent single core; [b]cores taken with USGS drive core sampler with combined area = 181.5cm^2. All onsite cores were collected with the USGS drive core sampler. [c]Single cores taken with drive core sampler (area = 45.4 cm^2) at random distances from 54E (comp) which itself represents four cores taken at the corners of an ~ 1 m square; [§]second core at first Terreton coring site; [¶]second coring site at Terreton ~ 1 km from site 1; [‡]second coring site at Ennis, MT ~ 20 km south of first Ennis site.

TABLE C

ATOM RATIOS OF NEPTUNIUM AND PLUTONIUM IN ONSITE AND NEAR-SITE INEEL SOILS

	Atom Ratio ± 1σ Uncertainty				Activity Ratio ± 1σ
Location	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu^{*}$	$^{242}Pu/^{239}Pu$	$^{237}Np/^{239,240}Pu^{\dagger}$
1W	0.366 ± 0.010	0.172 ± 0.001	0.00248 ± 0.00005	0.00347 ± 0.00005	0.00253 ± 0.00008
2W	-	0.168 ± 0.001	0.00236 ± 0.00005	0.00384 ± 0.00006	-
3W	-	0.169 ± 0.001	0.00244 ± 0.00015	0.00378 ± 0.00013	-
4W	-	0.165 ± 0.001	0.00237 ± 0.00027	0.00364 ± 0.00022	-
5W	0.366 ± 0.009	0.152 ± 0.001	0.00218 ±0.00004	0.00325± 0.00005	0.00264 ± 0.00007
6W	0.471 ± 0.028	0.169 ± 0.001	0.00249 ± 0.00007	0.00368 ± 0.00008	0.00327 ± 0.00020
7W	-	0.159 ± 0.001	0.00227 ± 0.00006	0.00354 ± 0.00006	-
8W	-	0.160 ± 0.001	0.00232 ± 0.00007	0.00362 ± 0.00008	-
9W	0.340 ± 0.016	0.139 ± 0.001	0.00200 ± 0.00008	0.00265 ± 0.00008	0.00265 ± 0.00008
10W	-	0.140 ± 0.001	0.00189 ± 0.00008	0.00284 ± 0.00008	-
11W	0.372 ± 0.018	0.166 ± 0.001	0.00239 ± 0.00007	0.00329 ± 0.00007	0.00261 ± 0.00012
12W	0.369 ±0.015	0.160 ± 0.001	0.00232 ± 0.00005	0.00342 ± 0.00006	0.00262 ± 0.00010
13W	0.409 ±0.011	0.163 ± 0.001	0.00247 ± 0.00005	0.00341 ± 0.00006	0.00288 ± 0.00008
14W	0.400 ± 0.016	0.163 ± 0.001	0.00246 ± 0.00006	0.00346 ± 0.00008	0.00282 ± 0.00011
15W	-	0.134 ± 0.001	0.00196 ± 0.00005	0.00297 ± 0.00006	-

TABLE C (cont'd**)**

ATOM RATIOS OF NEPTUNIUM AND PLUTONIUM IN ONSITE AND NEAR-SITE INEEL SOILS

	Atom Ratio ± 1 σ Uncertainty				Activity Ratio ± 1σ
Location	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$	$^{237}Np/^{239,240}Pu^{\dagger}$
16W	0.470 ± 0.019	0.170 ± 0.001	0.00244 ± 0.00010	0.00347 ± 0.00012	0.00326 ± 0.00013
17W	0.432 ± 0.016	0.174 ± 0.001	0.00243 ± 0.00006	0.00358 ± 0.00008	0.00297 ± 0.00011
18W	0.258 ± 0.012	0.123 ± 0.001	0.00163 ± 0.00005	0.00213 ± 0.00005	0.00200 ± 0.00009
19W	0.328 ± 0.013	0 .133 ± 0.001	0.00187 ± 0.00006	0.00240 ± 0.00007	0.00248 ± 0.00010
20W	0.276 ± 0.033	0.163 ± 0.001	0.00243 ± 0.00048	0.00263 ± 0.00047	0.00194 ± 0.00024
21W	0.300 ± 0.018	0.152 ± 0.001	0.00225 ± 0.00006	0.00326 ± 0.00008	0.00217 ± 0.00013
22W	0.267 ± 0.014	0.136 ± 0.001	0.00201 ± 0.00004	0.00259 ± 0.00006	0.00201 ± 0.00011
23W	0.427 ± 0.013	0.163 ± 0.001	0.00230 ± 0.00011	0.00354 ± 0.00011	0.00302 ± 0.00009
24W	0.183 ± 0.042	0.130 ± 0.001	0.00187 ± 0.00016	0.00238 ± 0.00014	0.00140 ± 0.00053
25W	0.418 ± 0.013	0.167 ± 0.001	0.00249 ± 0.00022	0.00353 ± 0.00045	0.00292 ± 0.00008
26W	0.281 ± 0.010	0.132 ± 0.001	0.00221 ±0.00025	0.00228 ± 0.00015	0.00213 ± 0.00008
27W	0.296 ± 0.042	0.139 ± 0.002	0.00201 ± 0.00015	0.00281 ± 0.00016	0.00221 ± 0.00032
55W	0.488 ± 0.014	0.174 ± 0.001	0.00245 ± 0.00005	0.00365 ± 0.00006	0.00336 ± 0.00010
56W	0.269 ± 0.007	0.121 ± 0.001	0.00172 ± 0.00003	0.00223 ± 0.00007	0.00210 ± 0.00006
57W	0.404 ± 0.012	0.158 ± 0.001	0.00224 ± 0.00012	0.00358 ± 0.00010	0.00288 ± 0.00009

TABLE C (cont'd)

ATOM RATIOS OF NEPTUNIUM AND PLUTONIUM IN ONSITE AND NEAR-SITE INEEL SOILS

	Atom Ratio ± 1 σ Uncertainty				Activity Ratio ± 1σ
Location	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$	$^{237}Np/^{239,240}Pu^{\dagger}$
58W	0.466 ± 0.015	0.174 ± 0.001	0.00262 ± 0.00011	0.00368 ± 0.00010	0.00320 ± 0.00010
60W	0.345 ± 0.010	0.149 ± 0.001	0.00198 ± 0.00013	0.00276 ± 0.00012	0.00251 ± 0.00008
61W	0.415 ± 0.012	0.165 ± 0.001	0.00214 ± 0.00014	0.00318 ± 0.00016	0.00292 ± 0.00009
62W	0.322 ± 0.009	0.144 ± 0.001	0.00217 ± 0.00006	0.00290 ± 0.00007	0.00237 ± 0.00007
63W	0.378 ± 0.011	0.163 ± 0.001	0.00250 ± 0.00011	0.00344 ± 0.00013	0.00267 ± 0.00008
81W	0.284 ± 0.007	0.132 ± 0.001	0.00182 ± 0.00030	0.00242 ± 0.00004	0.00215 ± 0.00006
82W	0.348 ± 0.010	0.153 ± 0.001	0.00217 ± 0.00004	0.00307 ± 0.00004	0.00251 ± 0.00007
83W	0.363 ± 0.009	0.155 ± 0.001	0.00228 ± 0.00004	0.00316 ± 0.00004	0.00260 ± 0.00007
84W	0.470 ± 0.012	0.176 ± 0.001	0.00261 ± 0.00004	0.00385 ± 0.00005	0.00322 ± 0.00008
85W	0.205 ± 0.005	0.118 ± 0.001	0.00167 ± 0.00004	0.00188 ± 0.00005	0.00161 ± 0.00004
92W	0.314 ± 0.007	0.143 ± 0.001	0.00207 ± 0.00003	0.00276 ± 0.00003	0.00232 ± 0.00005
28E	0.283 ± 0.035	0.150 ± 0.002	0.00207 ± 0.00031	0.00271 ± 0.00026	0.00206 ± 0.00026
29E	0.398 ± 0.025	0.160 ± 0.001	0.00242 ± 0.00008	0.00328 ± 0.00009	0.00283 ± 0.00018
30E	0.308 ± 0.017	0.138 ± 0.001	0.00208 ± 0.00008	0.00269 ± 0.00009	0.00230 ± 0.00013
31E	0.279 ± 0.023	0.157 ± 0.001	0.00233 ± 0.00008	0.00317 ± 0.00010	0.00200 ± 0.00016

TABLE C (cont'd)

ATOM RATIOS OF NEPTUNIUM AND PLUTONIUM IN ONSITE AND NEAR-SITE INEEL SOILS

	Atom Ratio ± 1 σ Uncertainty				Activity Ratio ± 1σ
Location	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$	$^{237}Np/^{239,240}Pu^{\dagger}$
32E	0.458 ± 0.013	0.179 ± 0.001	0.00250 ± 0.00010	0.00375 ± 0.00010	0.00311 ± 0.00009
33E	0.407 ± 0.017	0.164 ± 0.001	0.00234 ± 0.00006	0.00344 ± 0.00006	0.00288 ± 0.00013
34E	0.445 ± 0.016	0.167 ± 0.001	0.00238 ± 0.00007	0.00340 ± 0.00007	0.00311 ± 0.00012
35E	0.323 ± 0.008	0.153 ± 0.001	0.00222 ± 0.00005	0.00305 ± 0.00005	0.00233 ± 0.00006
36E	0.373 ± 0.030	0.153 ± 0.001	0.00214 ± 0.00008	0.00302 ± 0.00009	0.00260 ± 0.00022
37E	0.477 ± 0.017	0.165 ± 0.001	0.00272 ± 0.00006	0.00370 ± 0.00005	0.00334 ± 0.00012
38E	0.280 ± 0.024	0.110 ± 0.001	0.00145 ± 0.00034	0.00212 ± 0.00019	0.00231 ± 0.00020
39E	0.316 ± 0.011	0.146 ± 0.001	0.00205 ± 0.00003	0.00298 ± 0.00004	0.00232 ± 0.00008
40E	0.325 ± 0.009	0.135 ± 0.001	0.00193 ± 0.00009	0.00234 ± 0.00012	0.00245 ± 0.00007
41E	0.389 ± 0.010	0.156 ± 0.001	0.00236 ± 0.00005	0.00319 ± 0.00005	0.00278 ± 0.00007
42E	0.393 ± 0.011	0.176 ± 0.001	0.00261 ± 0.00010	0.00385 ± 0.00010	0.00269 ± 0.00007
43E	0.279 ± 0.011	0.141 ± 0.001	0.00205 ± 0.00003	0.00270 ± 0.00004	0.00208 ± 0.00007
44E	0.359 ± 0.010	0.149 ± 0.001	0.00227 ± 0.00008	0.00294 ± 0.00007	0.00261 ± 0.00007
45E	0.107 ± 0.004	0.091 ± 0.001	0.00168 ± 0.00002	0.00115 ± 0.00001	0.00091 ± 0.00003
46E	0.112 ± 0.003	0.092 ± 0.001	0.00159 ± 0.00001	0.00119 ± 0.00001	0.00094 ± 0.00003

TABLE C (cont'd)

ATOM RATIOS OF NEPTUNIUM AND PLUTONIUM IN ONSITE AND NEAR-SITE INEEL SOIL

	Atom Ratio ± 1 σ Uncertainty				Activity Ratio ± 1σ
Location	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$	$^{237}Np/^{239,240}Pu^{\dagger}$
47E	0.311 ± 0.011	0.154 ± 0.001	0.00226 ± 0.00005	0.00324 ± 0.00006	0.00224 ± 0.00008
48E	0.359 ± 0.014	0.143 ± 0.001	0.00197 ± 0.00005	0.00293 ± 0.00006	0.00266 ± 0.00011
49E	0.382 ± 0.011	0.165 ± 0.001	0.00231 ± 0.00004	0.00354 ± 0.00005	0.00269 ± 0.00008
50E	0.323 ± 0.009	0.147 ± 0.001	0.00220 ± 0.00003	0.00303 ± 0.00004	0.00236 ± 0.00007
51E	0.351 ± 0.012	0.156 ± 0.001	0.00223 ± 0.00008	0.00305 ± 0.00008	0.00251 ± 0.00009
52E	0.375 ± 0.012	0.159 ± 0.001	0.00209 ± 0.00006	0.00319 ± 0.00006	0.00267 ± 0.00009
53E	0.374 ± 0.011	0.162 ± 0.001	0.00242 ± 0.00004	0.00342 ± 0.00005	0.00265 ± 0.00008
54E	0.443 ± 0.023	0.169 ± 0.001	0.00287 ± 0.00009	0.00382 ± 0.00009	0.00308 ± 0.00016
59E	0.380 ± 0.010	0.156 ± 0.001	0.00260 ± 0.00014	0.00323 ± 0.00072	0.00273 ± 0.00007
64E	0.333 ± 0.011	0.142 ± 0.001	0.00236 ± 0.00014	0.00278 ± 0.00013	0.00246 ± 0.00009
65E	0.227 ± 0.007	0.118 ± 0.001	0.00160 ± 0.00004	0.00219 ± 0.00004	0.00179 ± 0.00006
66E	0.187 ± 0.006	0.118 ± 0.001	0.00162 ± 0.00003	0.00181 ± 0.00003	0.00147 ± 0.00005
67E	0.275 ± 0.016	0.133 ± 0.001	0.00191 ± 0.00005	0.00254 ± 0.00005	0.00208 ± 0.00012
68E	0.335 ± 0.016	0.154 ± 0.001	0.00228 ± 0.00005	0.00328 ± 0.00006	0.00241 ± 0.00011
69E	0.368 ± 0.013	0.160 ± 0.001	0.00259 ± 0.00003	0.00367 ± 0.00004	0.00261 ± 0.00009
70E	0.454 ± 0.015	0.168 ± 0.001	0.00240 ± 0.00003	0.00358 ± 0.00003	0.00316 ± 0.00010

TABLE C (cont'd)

ATOM RATIOS OF NEPTUNIUM AND PLUTONIUM IN ONSITE AND NEAR-SITE INEEL SOILS

	Atom Ratio ± 1 σ Uncertainty				Activity Ratio ± 1σ
Location	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$	$^{237}Np/^{239,240}Pu^{\dagger}$
71E	0.395 ± 0.016	0.166 ± 0.001	0.00229 ± 0.00004	0.00331 ± 0.00005	0.00277 ± 0.00011
72E	0.355 ± 0.011	0.156 ± 0.001	0.00221 ± 0.00004	0.00305 ± 0.00004	0.00254 ± 0.00008
73E	0.332 ± 0.010	0.151 ± 0.001	0.00220 ± 0.00003	0.00324 ± 0.00004	0.00241 ± 0.00007
74E	0.131 ± 0.005	0.096 ± 0.001	0.00126 ± 0.00001	0.00119 ± 0.00002	0.00109 ± 0.00004
75E	0.335 ± 0.010	0.150 ± 0.001	0.00212 ± 0.00004	0.00329 ± 0.00005	0.00244 ± 0.00008
76E	0.208 ± 0.006	0.120 ± 0.001	0.00175 ± 0.00003	0.00206 ± 0.00003	0.00163 ± 0.00005
77E	0.074 ± 0.002	0.073 ± 0.001	0.00094 ± 0.00001	0.00069 ± 0.00001	0.00066 ± 0.00002
78E	0.471 ± 0.018	0.168 ± 0.001	0.00243 ± 0.00004	0.00356 ± 0.00005	0.00328 ± 0.00012
79E	0.424 ± 0.010	0.166 ± 0.001	0.00248 ± 0.00004	0.00355 ± 0.00005	0.00297 ± 0.00007
80E	0.362 ± 0.009	0.157 ± 0.001	0.00231 ± 0.00004	0.00316 ± 0.00004	0.00259 ± 0.00007
86E	0.468 ± 0.013	0.176 ± 0.001	0.00257 ± 0.00006	0.00366 ± 0.00007	0.00320 ± 0.00009
87E	0.416 ± 0.010	0.172 ± 0.001	0.00248 ± 0.00006	0.00371 ± 0.00007	0.00287 ± 0.00007
88E	0.329 ± 0.008	0.157 ± 0.001	0.00219 ± 0.00006	0.00324 ± 0.00007	0.00235 ± 0.00006
89E	0.374 ± 0.009	0.152 ± 0.001	0.00224 ± 0.00007	0.00292 ± 0.00009	0.00271 ± 000007
90E	0.376 ± 0.010	0.154 ± 0.001	0.00212 ± 0.00005	0.00320 ± 0.00006	0.00270 ± 0.00007

TABLE C (cont'd)

ATOM RATIOS OF NEPTUNIUM AND PLUTONIUM IN ONSITE AND NEAR-SITE INEEL SOILS

	Atom Ratio ± 1 σ Uncertainty				Activity Ratio ± 1σ
Location	$^{237}Np/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$	$^{237}Np/^{239,240}Pu^{\dagger}$
91E	0.411 ± 0.013	0.157 ± 0.001	0.00227 ± 0.00006	0.00334 ± 0.00007	0.00294 ± 0.00009
93E	0.383 ± 0.012	0.162 ± 0.001	0.00237 ± 0.00011	0.00320 ± 0.00010	0.00270 ± 0.00009
94E	0.398 ± 0.012	0.164 ± 0.001	0.00234 ± 0.00008	0.00329 ± 0.00008	0.00280 ± 0.00008
95E	0.363 ± 0.009	0.161 ± 0.001	0.00204 ± 0.00007	0.00347 ± 0.00007	0.00257 ± 0.00006
96E	0.364 ± 0.028	0.161 ± 0.001	0.00218 ± 0.00024	0.00302 ± 0.00026	0.00258 ± 0.00020

^{237}Np was not measured in these first samples until acceptable laboratory procedures for its separation were established at EML; - not measured; *decay corrected to 1 January 1995 using a $T_{½}$ of 14.33 ± 0.02 yr; †the $^{237}Np/^{239,240}Pu$ alpha activity ratio was calculated from mass spectrometrically determined atom ratios and known half lives. Values used for the half lives of ^{237}Np, ^{239}Pu, ^{240}Pu, and ^{241}Pu were $(2.14 \pm 0.01) \times 10^{6}$ yr, $(2.4119 \pm 0.0027) \times 10^{4}$ yr, and $(6.564 \pm 0.011) \times 10^{3}$ yr, respectively.

TABLE D

NEPTUNIUM AND PLUTONIUM ATOM CONCENTRATIONS IN ONSITE AND NEAR-SITE INEEL SOILS

Location	Atom Concentrations ± 1σ Uncertainty (atoms g^{-1}dry)				
	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu†	^{242}Pu
1W	2.44 ± 0.07 (10^8)	6.67 ± 0.14 (10^8)	1.15 ± 0.03 (10^8)	1.70 ± 0.04 (10^6)	2.32 ± 0.06 (10^5)
2W	-	*	*	*	*
3W	-	2.79 ± 0.04 (10^8)	4.71 ± 0.07 (10^7)	6.81 ± 0.41 (10^5)	1.05 ± 0.04 (10^6)
4W	-	3.96 ± 0.07 (10^8)	6.51 ± 0.12 (10^7)	9.4 ± 1.1 (10^5)	1.44 ± 0.09 (10^6)
5W	1.36 ± 0.04 (10^8)	3.73 ± 0.06 (10^8)	5.68 ± 0.10 (10^7)	8.13 ± 0.20 (10^5)	1.21 ± 0.03 (10^6)
6W	1.42 ± 0.09 (10^8)	3.02 ± 0.05 (10^8)	5.12 ± 0.09 (10^7)	7.51 ± 0.25 (10^5)	1.10 ± 0.03 (10^6)
7W	-	4.30 ± 0.09 (10^8)	6.82 ± 0.14 (10^7)	0.98 ± 0.03 (10^6)	1.52 ± 0.04 (10^6)
8W	-	4.34 ± 0.07 (10^8)	6.96 ± 0.12 (10^7)	1.01 ± 0.03 (10^6)	1.57 ± 0.04 (10^6)
9W	9.08 ± 0.43 (10^7)	2.67 ± 0.05 (10^8)	3.73 ± 0.07 (10^7)	5.35 ± 0.23 (10^5)	7.08 ± 0.24 (10^5)
10W	-	3.75 ± 0.04 (10^8)	5.26 ± 0.07 (10^7)	7.08 ± 0.31 (10^5)	1.06 ± 0.03 (10^6)
11W	1.45 ± 0.07 (10^8)	3.90 ± 0.08 (10^8)	6.48 ± 0.14 (10^7)	9.33 ± 0.33 (10^5)	1.29 ± 0.04 (10^6)
12W	1.37 ± 0.02 (10^8)	3.72 ± 0.06 (10^8)	5.93 ± 0.10 (10^7)	8.61 ± 0.24 (10^5)	1.27 ± 0.03 (10^6)
13W	1.48 ± 0.04 (10^8)	3.62 ± 0.06 (10^8)	5.98 ± 0.10 (10^7)	8.96 ± 0.24 (10^5)	1.24 ± 0.03 (10^6)
14W	1.23 ± 0.05 (10^8)	3.06 ± 0.05 (10^8)	4.99 ± 0.08 (10^7)	7.53 ± 0.20 (10^5)	1.06 ± 0.03 (10^6)
15W	-	3.36 ± 0.04 (10^8)	4.50 ± 0.05 (10^7)	6.56 ± 0.16 (10^5)	9.96 ± 0.24 (10^5)

TABLE D (cont'd)

NEPTUNIUM AND PLUTONIUM ATOM CONCENTRATIONS IN ONSITE AND NEAR-SITE INEEL SOILS

Location	Atom Concentrations ± 1 σ Uncertainty (atoms g^{-1}dry)				
	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu†	^{242}Pu
16W	9.92 ± 0.41 (10^7)	2.11 ± 0.04 (10^8)	3.60 ± 0.07 (10^7)	5.15 ± 0.23 (10^5)	7.33 ± 0.29 (10^5)
17W	1.24 ± 0.05 (10^8)	2.86 ± 0.05(10^8)	4.97 ± 0.09 (10^7)	6.95 ± 0.22 (10^5)	1.02 ± 0.03 (10^6)
18W	1.56 ± 0.07 (10^8)	6.05 ± 0.10 (10^8)	7.47 ± 0.17 (10^7)	9.88 ± 0.33 (10^5)	1.29 ± 0.04 (10^6)
19W	9.70 ± 0.40 (10^7)	2.96 ± 0.05 (10^8)	3.94 ± 0.07 (10^7)	5.54 ± 0.20 (10^5)	7.10 ± 0.23 (10^5)
20W	9.04 ± 0.89 (10^7)	3.28 ± 0.23 (10^8)	5.34 ± 0.40 (10^7)	8.0 ± 1.7 (10^5)	8.6 ± 1.6 (10^5)
21W	1.01 ± 0.06 (10^8)	3.36 ± 0.06 (10^8)	5.11 ± 0.10 (10^7)	7.53 ± 0.25 (10^5)	1.09 ± 0.03 (10^6)
22W	8.74 ± 0.46 (10^7)	3.27 ± 0.05 (10^8)	4.46 ± 0.08 (10^7)	6.57 ± 0.18 (10^5)	8.47 ± 0.23 (10^5)
23W	1.01 ± 0.03 (10^8)	2.35 ± 0.04 (10^8)	3.83 ± 0.07 (10^7)	5.42 ± 0.28 (10^5)	8.33 ± 0.28 (10^5)
24W	9.8 ± 3.7 (10^7)	5.34 ± 0.18 (10^8)	6.92 ± 0.24 (10^7)	9.99 ± 0.90 (10^5)	1.27 ± 0.08 (10^6)
25W	9.85 ± 0.28 (10^7)	2.35 ± 0.04 (10^8)	3.93 ± 0.06 (10^7)	5.85 ± 0.53 (10^5)	8.3 ± 1.1 (10^5)
26W	1.12 ± 0.04 (10^8)	3.97 ± 0.07 (10^8)	5.25 ± 0.10 (10^7)	8.8 ± 1.0 (10^5)	9.07 ± 0.63 (10^5)
27W	1.05 ± 0.15 (10^8)	3.55 ± 0.11 (10^8)	4.94 ± 0.17 (10^7)	7.14 ± 0.56 (10^5)	9.98 ± 0.64 (10^5)
55W	1.66 ± 0.05 (10^8)	3.41 ± 0.06 (10^8)	5.93 ± 0.11 (10^7)	8.36 ± 0.23 (10^5)	1.24 ± 0.03 (10^6)
56W	1.15 ± 0.03 (10^8)	4.25 ± 0.06 (10^8)	5.13 ± 0.08 (10^7)	7.30 ± 0.18 (10^5)	9.50 ± 0.31 (10^5)
57W	9.43 ± 0.28 (10^7)	2.34 ± 0.04 (10^8)	3.68 ± 0.06 (10^7)	5.22 ± 0.29 (10^5)	8.37 ± 0.27 (10^5)

TABLE D (cont'd)

NEPTUNIUM AND PLUTONIUM ATOM CONCENTRATIONS IN ONSITE AND NEAR-SITE INEEL SOILS

Atom Concentration ± 1 σ Uncertainty (atoms g^{-1}dry)

Location	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu†	^{242}Pu
58W	1.33 ± 0.04 (10^8)	2.84 ± 0.07 (10^8)	4.95 ± 0.13 (10^7)	7.46 ± 0.35 (10^5)	1.05 ± 0.04 (10^6)
60W	1.82 ± 0.05 (10^8)	5.27 ± 0.12 (10^8)	7.86 ± 0.18 (10^7)	1.04 ± 0.07 (10^6)	1.46 ± 0.07(10^6)
61W	9.94 ± 0.28 (10^7)	2.39 ± 0.04 (10^8)	3.94 ± 0.08 (10^7)	5.12 ± 0.35 (10^5)	7.60 ± 0.41 (10^5)
62W	1.95 ± 0.05 (10^8)	6.05 ± 0.11 (10^8)	8.72 ± 0.17 (10^7)	1.31 ± 0.04 (10^6)	1.75 ± 0.06 (10^6)
63W	9.10 ± 0.28 (10^7)	2.41 ± 0.05 (10^8)	3.92 ± 0.08 (10^7)	6.03 ± 0.28 (10^5)	8.29 ± 0.34 (10^5)
81W	1.30 ± 0.03 (10^8)	4.59 ± 0.06 (10^8)	6.07 ± 0.09 (10^7)	8.36 ± 0.19 (10^5)	1.11 ± 0.02 (10^6)
82W	1.24 ± 0.03 (10^8)	3.55 ± 0.05 (10^8)	5.45 ± 0.08 (10^7)	7.72 ± 0.17 (10^5)	1.09 ± 0.02 (10^6)
83W	1.40 ± 0.03 (10^8)	3.87 ± 0.05 (10^8)	6.01 ± 0.08 (10^7)	8.81 ± 0.18 (10^5)	1.22 ± 0.02 (10^6)
84W	1.19 ± 0.03 (10^8)	2.53 ± 0.03 (10^8)	4.44 ± 0.06 (10^7)	6.58 ± 0.13 (10^5)	9.73 ± 0.18 (10^5)
85W	3.87 ± 0.09 (10^7)	1.89 ± 0.03 (10^8)	2.23 ± 0.03 (10^7)	3.14 ± 0.09 (10^5)	3.54 ± 0.10 (10^5)
92W	1.04 ± 0.02 (10^8)	3.31 ± 0.04 (10^8)	4.75 ± 0.06 (10^7)	6.85 ± 0.13 (10^5)	9.12 ± 0.16 (10^5)
28E	1.08 ± 0.13 (10^8)	3.81 ± 0.17 (10^8)	5.72 ± 0.27 (10^7)	7.9 ± 1.2 (10^5)	1.03 ± 0.11 (10^6)
29E	1.05 ± 0.06 (10^8)	2.64 ± 0.05 (10^8)	4.22 ± 0.08 (10^7)	6.38 ± 0.24 (10^5)	8.66 ± 0.24 (10^5)
30E	8.30 ± 0.45 (10^7)	2.70 ± 0.05 (10^8)	3.73 ± 0.07 (10^7)	5.62 ± 0.24 (10^5)	7.25 ± 0.26 (10^5)
31E	1.06 ± 0.08 (10^8)	3.80 ± 0.09 (10^8)	5.95 ± 0.14 (10^7)	8.86 ± 0.37 (10^5)	1.21 ± 0.05 (10^6

NEPTUNIUM AND PLUTONIUM ATOM CONCENTRATIONS IN ONSITE AND NEAR-SITE INEEL SOILS

Location	Atom Concentration ± 1 σ Uncertainty (atoms g^{-1}dry)				
	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu†	^{242}Pu
32E	9.49 ± 0.26 (10^{7})	2.07 ± 0.04 (10^{8})	3.71 ± 0.07 (10^{7})	5.18 ± 0.22 (10^{5})	7.77 ± 0.24 (10^{5})
33E	1.53 ± 0.06 (10^{8})	3.75 ± 0.06 (10^{8})	6.15 ± 0.11 (10^{7})	8.76 ± 0.24 (10^{5})	1.29 ± 0.03 (10^{6})
34E	1.39 ± 0.05 (10^{8})	3.12 ± 0.06 (10^{8})	5.22 ± 0.10 (10^{7})	7.43 ± 0.24 (10^{5})	1.06 ± 0.03 (10^{6})
35E	1.66 ± 0.04 (10^{8})	5.15 ± 0.09 (10^{8})	7.85 ± 0.14 (10^{7})	1.14 ± 0.03 (10^{6})	1.57 ± 0.04 (10^{6})
36E	1.13 ± 0.09 (10^{8})	3.03 ± 0.06 (10^{8})	4.63 ± 0.09 (10^{7})	6.49 ± 0.27 (10^{5})	9.15 ± 0.31 (10^{5})
37E	1.72 ± 0.06 (10^{8})	3.60 ± 0.05 (10^{8})	5.96 ± 0.09 (10^{7})	9.79 ± 0.25 (10^{5})	1.33 ± 0.03 (10^{6})
38E	9.25 ± 0.78 (10^{7})	3.30 ± 0.07 (10^{8})	3.36 ± 0.08 (10^{7})	4.8 ± 1.1 (10^{5})	7.00 ± 0.65 (10^{5})
39E	8.70 ± 0.29 (10^{7})	2.75 ± 0.04 (10^{8})	4.03 ± 0.06 (10^{7})	5.64 ± 0.12 (10^{5})	8.20 ± 0.16 (10^{5})
40E	7.46 ± 0.23 (10^{7})	2.29 ± 0.04 (10^{8})	3.10 ± 0.06 (10^{7})	4.42 ± 0.22 (10^{5})	5.36 ± 0.29 (10^{5})
41E	1.43 ± 0.04 (10^{8})	3.68 ± 0.06 (10^{8})	5.75 ± 0.10 (10^{7})	8.68 ± 0.23 (10^{5})	1.17 ± 0.03 (10^{6})
42E	7.02 ± 0.19 (10^{7})	1.79 ± 0.03 (10^{8})	3.14 ± 0.06 (10^{7})	4.67 ± 0.19 (10^{5})	6.88 ± 0.21 (10^{5})
43E	1.14 ± 0.04 (10^{8})	4.09 ± 0.06 (10^{8})	5.75 ± 0.10 (10^{7})	8.68 ± 0.23 (10^{5})	1.10 ± 0.02 (10^{6})
44E	1.09 ± 0.03 (10^{8})	3.05 ± 0.06 (10^{8})	4.54 ± 0.09 (10^{7})	6.92 ± 0.26 (10^{5})	8.98 ± 0.26 (10^{5})
45E	1.40 ± 0.04 (10^{8})	1.30 ± 0.02 (10^{9})	1.18 ± 0.02 (10^{8})	2.18 ± 0.04 (10^{6})	1.50 ± 0.03 (10^{6})
46E	1.53 ± 0.04 (10^{8})	1.37 ± 0.02 (10^{9})	1.26 ± 0.02 (10^{8})	2.17 ± 0.03 (10^{6})	1.63 ± 0.03 (10^{6})
47E	1.01 ± 0.04 (10^{8})	3.24 ± 0.05 (10^{8})	4.99 ± 0.08 (10^{7})	7.30 ± 0.19 (10^{5})	1.05 ± 0.02 (10^{6})

TABLE D (cont'd)

NEPTUNIUM AND PLUTONIUM ATOM CONCENTRATIONS IN ONSITE AND NEAR-SITE INEEL SOIL

Location	Atom Concentration ± 1 σ Uncertainty (atoms g^{-1}dry)				
	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu†	^{242}Pu
48E	$9.50 \pm 0.39 (10^{7})$	$2.65 \pm 0.04 (10^{8})$	$3.79 \pm 0.07 (10^{7})$	$5.22 \pm 0.15 (10^{5})$	$7.77 \pm 0.21 (10^{5})$
49E	$1.24 \pm 0.03 (10^{8})$	$3.25 \pm 0.05 (10^{8})$	$5.35 \pm 0.08 (10^{7})$	$7.50 \pm 0.16 (10^{5})$	$1.15 \pm 0.02 (10^{6})$
50E	$1.36 \pm 0.04 (10^{8})$	$4.20 \pm 0.06 (10^{8})$	$6.18 \pm 0.09 (10^{7})$	$9.26 \pm 0.18 (10^{5})$	$1.27 \pm 0.02 (10^{6})$
51E	$7.76 \pm 0.25 (10^{7})$	$2.21 \pm 0.03 (10^{8})$	$3.45 \pm 0.06 (10^{7})$	$4.92 \pm 0.19 (10^{5})$	$6.73 \pm 0.20 (10^{5})$
52E	$1.16 \pm 0.03 (10^{8})$	$3.10 \pm 0.05 (10^{8})$	$4.93 \pm 0.08 (10^{7})$	$6.47 \pm 0.20 (10^{5})$	$9.89 \pm 0.23 (10^{5})$
53E	$1.04 \pm 0.03 (10^{8})$	$2.79 \pm 0.04 (10^{8})$	$4.50 \pm 0.07 (10^{7})$	$6.74 \pm 0.14 (10^{5})$	$9.52 \pm 0.19 (10^{5})$
54E	$1.98 \pm 0.11 (10^{8})$	$4.48 \pm 0.08 (10^{8})$	$7.59 \pm 0.15 (10^{7})$	$1.29 \pm 0.05 (10^{6})$	$1.71 \pm 0.05 (10^{6})$
59E	$1.01 \pm 0.28 (10^{8})$	$2.67 \pm 0.05 (10^{8})$	$4.15 \pm 0.07 (10^{7})$	$6.94 \pm 0.40 (10^{5})$	$8.62 \pm 0.35 (10^{5})$
64E	$9.27 \pm 0.30 (10^{7})$	$2.79 \pm 0.06 (10^{8})$	$3.97 \pm 0.09 (10^{7})$	$6.58 \pm 0.42 (10^{5})$	$7.75 \pm 0.39 (10^{5})$
65E	$1.13 \pm 0.03 (10^{8})$	$4.99 \pm 0.08 (10^{8})$	$5.86 \pm 0.10 (10^{7})$	$7.96 \pm 0.23 (10^{5})$	$1.09 \pm 0.03 (10^{6})$
66E	$1.07 \pm 0.03 (10^{8})$	$5.70 \pm 0.08 (10^{8})$	$6.72 \pm 0.10 (10^{7})$	$9.25 \pm 0.21 (10^{5})$	$1.03 \pm 0.02 (10^{6})$
67E	$8.52 \pm 0.49 (10^{7})$	$3.10 \pm 0.05 (10^{8})$	$4.12 \pm 0.07 (10^{7})$	$5.92 \pm 0.18 (10^{5})$	$7.88 \pm 0.21 (10^{5})$
68E	$9.01 \pm 0.41 (10^{7})$	$2.69 \pm 0.04 (10^{8})$	$4.15 \pm 0.06 (10^{7})$	$6.12 \pm 0.16 (10^{5})$	$8.81 \pm 0.20 (10^{5})$
69E	$1.61 \pm 0.05 (10^{8})$	$4.38 \pm 0.06 (10^{8})$	$7.02 \pm 0.10 (10^{7})$	$1.13 \pm 0.02 (10^{6})$	$1.60 \pm 0.03 (10^{6})$
70E	$5.34 \pm 0.17 (10^{8})$	$1.18 \pm 0.02 (10^{9})$	$1.98 \pm 0.03 (10^{8})$	$2.83 \pm 0.05 (10^{6})$	$4.21 \pm 0.07 (10^{6})$

TABLE D (cont'd)

NEPTUNIUM AND PLUTONIUM ATOM CONCENTRATIONS IN ONSITE AND NEAR-SITE INEEL SOILS

Location	Atom Concentration ± 1 σ Uncertainty (atoms g^{-1}dry)				
	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu†	^{242}Pu
71E	1.12 ± 0.04 (10^8)	2.84 ± 0.04 (10^8)	4.71 ± 0.07 (10^7)	6.51 ± 0.14 (10^5)	9.41 ± 0.18 (10^5)
72E	1.59 ± 0.05 (10^8)	4.47 ± 0.07 (10^8)	6.97 ± 0.11 (10^7)	9.86 ± 0.21 (10^5)	1.36 ± 0.03 (10^6)
73E	1.98 ± 0.05 (10^8)	5.96 ± 0.09 (10^8)	8.99 ± 0.13 (10^7)	1.31 ± 0.03 (10^6)	1.93 ± 0.04 (10^6)
74E	1.14 ± 0.04 (10^8)	8.70 ± 0.12 (10^8)	8.39 ± 0.12 (10^7)	1.09 ± 0.02 (10^6)	1.04 ± 0.02 (10^6)
75E	9.23 ± 0.27 (10^7)	2.75 ± 0.04 (10^8)	4.13 ± 0.06 (10^7)	5.84 ± 0.13 (10^5)	9.07 ± 0.18 (10^5)
76E	9.70 ± 0.27 (10^7)	4.66 ± 0.06 (10^8)	5.58 ± 0.08 (10^7)	8.17 ± 0.17 (10^5)	9.61 ± 0.19 (10^5)
77E	1.31 ± 0.03 (10^8)	1.76 ± 0.02 (10^9)	1.29 ± 0.02 (10^8)	1.66 ± 0.03 (10^6)	1.22 ± 0.02 (10^6)
78E	1.53 ± 0.06 (10^8)	3.26 ± 0.05 (10^8)	5.47 ± 0.08 (10^7)	7.91 ± 0.16 (10^5)	1.16 ± 0.02 (10^6)
79E	1.15 ± 0.03 (10^8)	2.70 ± 0.04 (10^8)	4.47 ± 0.06 (10^7)	6.68 ± 0.15 (10^5)	9.58 ± 0.19 (10^5)
80E	1.53 ± 0.04 (10^8)	4.23 ± 0.06 (10^8)	6.66 ± 0.10 (10^7)	9.76 ± 0.20 (10^5)	1.34 ± 0.03 (10^6)
86E	9.29 ± 0.13 (10^7)	1.99 ± 0.03 (10^8)	3.50 ± 0.06 (10^7)	5.11 ± 0.14 (10^5)	7.26 ± 0.18 (10^5)
87E	1.51 ± 0.04 (10^8)	3.63 ± 0.07 (10^8)	6.25 ± 0.11 (10^7)	8.98 ± 0.26 (10^5)	1.35 ± 0.04 (10^6)
88E	1.59 ± 0.04 (10^8)	4.82 ± 0.09 (10^8)	7.57 ± 0.15 (10^7)	1.06 ± 0.04 (10^6)	1.56 ± 0.05 (10^6)
89E	7.51 ± 0.18 (10^7)	2.01 ± 0.04 (10^8)	3.05 ± 0.06 (10^7)	4.49 ± 0.17 (10^5)	5.87 ± 0.20 (10^5)
90E	1.56 ± 0.04 (10^8)	4.15 ± 0.08 (10^8)	6.40 ± 0.12 (10^7)	8.79 ± 0.26 (10^5)	1.33 ± 0.04 (10^6)

TABLE D (cont'd)

NEPTUNIUM AND PLUTONIUM ATOM CONCENTRATIONS IN ONSITE AND NEAR-SITE INEEL SOILS

Location	Atom Concentration ± 1 σ Uncertainty (atoms g^{-1}dry)				
	^{237}Np	^{239}Pu	^{240}Pu	^{241}Pu†	^{242}Pu
91E	1.26 ± 0.05 (10^8)	3.07 ± 0.05 (10^8)	4.83 ± 0.08 (10^7)	6.97 ± 0.20 (10^5)	1.02 ± 0.03 (10^6)
93E	5.24 ± 0.16 (10^7)	1.37 ± 0.02 (10^8)	2.22 ± 0.04 (10^7)	3.24 ± 0.16 (10^5)	4.39 ± 0.15 (10^5)
94E	6.73 ± 0.18 (10^7)	1.69 ± 0.02 (10^8)	2.77 ± 0.04 (10^7)	3.96 ± 0.14 (10^5)	5.56 ± 0.15 (10^5)
95E	1.22 ± 0.03 (10^8)	3.35 ± 0.06 (10^8)	5.40 ± 0.10 (10^7)	6.85 ± 0.26 (10^5)	1.16 ± 0.03 (10^6)
96E	1.37 ± 0.07 (10^8)	3.75 ± 0.22 (10^8)	6.02 ± 0.37 (10^7)	8.2 ± 1.0 (10^5)	1.13 ± 0.12 (10^6)

- ^{237}Np was not measured in these first samples until acceptable laboratory procedures for its separation were established at EML;
* run without ^{244}Pu tracer to determine isotopic ratios only; † decay corrected to 1 January 1995 using a $T_{½}$ of 14.33 ± 0.02 yr.