CAUSES OF RADIOLOGICAL FALSE POSITIVES IN ENVIRONMENTAL SOIL AND GROUNDWATER DATA FROM COMMERCIAL LABORATORIES

Walter Kubilius, Brian Crandall, and Terrence Killeen; Westinghouse Savannah River Company, Aiken, SC 29808; and

James Westmoreland; Environmental Physics, Inc., Charleston, SC 29417

The Savannah River Site (SRS) procures about 200,000 radiological analyses of soil and groundwater each year for CERCLA environmental investigations. In response to needs of data users for lower detection limits, SRS since 1994 has required contract laboratories to achieve MDAs of <0.06 pCi/g for most nuclides in soils by alpha and gamma spectroscopy, and <1 pCi/g for most LSC and GFPC nuclides. Many soil samples analyzed under these requirements were reported with low-level detects of short-lived and unsupported radionuclides; suggesting the possibility of false positives.

In order to investigate whether systematic false positives occur in SRS soil and groundwater data produced by contract labs, we interviewed laboratory personnel, and examined instrument printouts, calibration records, and procedure manuals from three radiochemistry laboratories under contract to SRS.

As might be expected after detection limits are reduced, a significant percentage of hits for certain radionuclides were determined to be false positives. They were found to be produced at all three laboratories; most occurred when reported concentrations were <0.5 pCi/g. Affected methods include gamma spectroscopy, alpha spectroscopy, liquid scintillation, and gas flow proportional counting. For the following nuclides, more than half of all hits reported to SRS by contract labs in 1995-6 were found to be spurious: Mn-54, Zr-95, I-129, Pm-147, Eu-155, Np-239. Additional nuclides subject to false positives included: Tc-99, Ra-228, Th-230, Np-237, Am-241, Am-243, Cm-245/246. On the other hand, well-behaved nuclides with few or no recognized false positives in SRS samples included: H-3, K-40, Co-60, Sr-90, Cs-137, Tl-208, Pb-212, Ra-226, Ac-228, Th-228, Th-232, U-234, U-235, U-238.

Root causes of false positives are attributed to both customer (SRS) and laboratory errors. A summary of some of the main causes follows.

CUSTOMER CAUSES

1. Customer and regulatory pressure for very low MDAs increased likelihood for false positives. SRS contracts specify very low MDAs. In alpha spectroscopy, some reported hits of isotopes of Pu, Np, Cm below 0.15 pCi/g are attributed to interference from natural thorium, natural uranium, or from cerium nitrate reagent. In gamma

spectroscopy, reported hits below 0.10 pCi/g for unsupported nuclides such as Na-22, Co-57, Co-58, Zn-65, Ru-106, Pm-146 may be due to spectral anomalies associated with signal processing, or software peak search and identification criteria.

2. SRS personnel provided unclear direction to laboratories. Customer instructions were incompletely documented, and provided by several people with varying degrees of radiological training. Laboratory confusion about customer directions played a role in reporting of gamma spectroscopy false positives at two labs.

3. SRS did not carefully read laboratory Standard Operating Procedures (SOPs) when negotiating contracts with laboratories. One lab's SOP for Ra-228 by GFPC clearly stated that the method was subject to interference from Sr-90. However, SRS sent Sr-90-bearing samples for Ra-228 analysis by this method over an extended period of time.

LABORATORY CAUSES

1. Low abundance gamma peaks of the natural nuclides Tl-208, Pb-212, and Ac-228 were routinely reported as Mn-54, Zr-95, Eu-155, and Np-239. This took place at all three labs, and resulted from two root causes. First, laboratories failed to include low abundance (<1-2%) peaks of Tl-208, Pb-212, and Ac-228 in the libraries used by instrument software for interference calculations. For most environmental samples with moderate MDAs, this would not matter. However, at commonly found concentrations of natural nuclides in soils, interference from these low abundance peaks may generate apparent detections of Mn-54, Zr-95, Eu-155, and Np-239, usually below 0.2 pCi/g, but occasionally up to 0.8 pCi/g. Second, even when instrument software rejected the identifications, the incorrectly identified nuclides were still reported, because the labs' data management systems are not able to distinguish between software-accepted nuclides and software-rejected nuclides. Manual review was not always performed, partly because of confusion due to poorly communicated customer acceptance requirements. Nearly every hit of Mn-54, Zr-95, Eu-155, and Np-239 reported to SRS in 1995-6 was attributable to natural isotopes.

2. I-129 was reported from instruments which were unsuitable for counting I-129 X-rays. One laboratory counted I-129 for SRS using three gamma spectroscopy detectors, one of which was equipped with an aluminum cap which was nearly opaque to the 29 to 39 kev photons characteristic of I-129. This resulted in about 300 unusable analyses being reported to SRS in 1994-6, including both false negatives and spurious false positives.

3. Failure of separation chemistry caused false positives of Pm-147 and Tc-99. Nearly every Pm-147 hit reported to SRS from one lab during 1995-6 was unusable due to interference from unknown (presumed actinide) alpha and beta emitters, or from chemical/thermal luminescence. The laboratory did not review spectra carefully before releasing data. Reported concentrations ranged from 0.1 to 4.8 pCi/g in soils. Many

Tc-99 hits reported to SRS from two labs were unusable because of interference from H-3 or chemical/thermal luminescence. These false positives, ranging up to 2 pCi/g, could have been avoided by selecting the counting windows to exclude low energy counts, or by technical review of spectra.

4. In alpha spectroscopy, peak tailing of high-count nuclides into adjacent Regions of Interest causes false positives of Th-230 and Cm-245/246. The Th-229 tracer isotope contributes spurious counts to the Th-230 window. This phenomenon occurs in groundwater samples, resulting in false positives up to 1 pCi/L. Significant activity of Am-241 (often encountered at SRS) may result in counts attributed to Cm-245/246. Peak tailing can be easily recognized during inspection of spectra, especially for Th-230, but false positives at all three labs indicated that sufficient interpretation of spectra was not always performed.

SRS's requirement of very low MDAs in routine environmental samples has resulted in the reporting of significant numbers of low level false positives in environmental soil and groundwater. These false positives arise from a variety of sources, including limitations in separation chemistry, incomplete accounting for interferences, insufficient technical review of spectra, and poor customer/vendor communication.

Many of the problems are correctable (e.g. interference library in gamma spectroscopy, and review of LSC spectra for chemical/thermal luminescence contributions), and have been adequately investigated and addressed during a series of cooperative explorations involving both SRS and laboratory personnel.