

TECHNICAL BASIS DOCUMENT

**DERIVATION AND APPLICATION OF THE
"SUM-OF-FRACTIONS" CALCULATION**

**MOLYCORP'S YORK, PA SITE
SOILS REMEDIATION PROJECT**

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Molycorp, Inc. Soils Remediation Project
York, PA

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1.0 INTRODUCTION

1.1 BACKGROUND

On June 6, 2000, The U.S. Nuclear Regulatory Commission (NRC) issued License amendment No. 9 to Molycorp's York, PA Facility License SMB-1408 (NRC 2000a) approving the decommissioning of the site in accordance with the site decommissioning plan (DP), revision 1, dated June 1999 (Molycorp 1999). The approval was granted in consideration of information and commitments provided as supplements to the DP by Molycorp (Molycorp 1999b) in response to the NRC's requests for additional information (RAI) (NRC 1999).

Molycorp's responses to the NRC's requests for additional information indicated their understanding and acceptance that the residual radioactivity in soil guideline levels cited in the DP did not apply independently, but instead should be considered codependent limits for which the sum-of-fractions (SOF) must be considered (Molycorp 1999b, RSI 2001, RSI 2002).

This technical basis document establishes the rationale and method used by Molycorp in evaluating residual radioactivity concentrations in soil samples relative to the approved soil concentration guidelines for the York, PA site. Specifically, it derives the algebraic expression used to evaluate the SOF for multiple radionuclides potentially present in soil when analyzed with high resolution gamma spectroscopy methods. The expression compares the concentrations of each radionuclide to its specific limit concentration limit such that compliance is indicated when the SOF is less than or equal to unity (NRC 1992, NRC 2000b). As the algebraic expression used to calculate the SOF is dependent upon the analytical method used, previously presented technical basis documents deriving the SOF equations used at the York, PA site for low resolution gamma spectral measurements and without allowance for radon daughter ingrowth are not appropriate for the analytical method employed by MACTEC in the supplemental soil characterization work being performed (Molycorp 2003).

1.2 GENERAL SITE DESCRIPTION

The Molycorp, Inc. York, PA Facility is situated on the outskirts of the City of York, PA at 350 North Sherman St., in Spring Garden Township, PA. The active site consists of approximately six fenced acres bounded by Olive St. to the north, Hudson St. to the west, N. Sherman St. to the east, and by the Norfolk and Southern Railroad track to the south.

1.3 SITE HISTORY

The site was used by Molycorp, Inc. to produce a broad line of inorganic rare earth chemicals used to make catalysts for the chemical industry and for various other industrial purposes. The rare earth processing plant was part of the facility that had raw material containing naturally occurring uranium and thorium. In 1981, Molycorp acquired a radioactive source materials license, SMB-1408, in compliance with applicable federal regulation and based upon the quantity and concentrations of thorium in rare earth materials being processed at the site (NRC 1981)

1.4 CURRENT SITE CONDITIONS

All of the buildings and structures that were at one time in place at the site have since been radiologically surveyed and removed. Approximately two-thirds of the site (~4 acres) have already been released from radiological controls by the NRC based upon extensive excavation of subsurface materials. This revision of the SOF calculation methodology is necessitated by: 1) a change in analysis method from NaI(Tl) gamma spectroscopy to high-resolution gamma spectroscopy, and 2) a departure from the prior practice of immediately counting samples collected in the field instead allowing for sufficient time to elapse to achieve equilibrium conditions. The high-resolution method is able to directly quantify a number of isotopes of interest, which was not possible with the NaI(Tl) spectroscopy method, thus avoiding the need to infer the activity of certain isotopes. The previously developed SOF calculation contained factors used to infer the activities of Ra-226, U-238 + U234, and excess thorium. This technical basis document makes the necessary adjustments to the previously used SOF calculation to account for the better quantification achieved with the high-resolution gamma spectroscopy analytical method now being employed.

The remaining portion of the site for which compliance with the release criteria have not been demonstrated (~2 acres) are to be resampled in accordance with the requirements of the approved DP in order to further characterize the isotopic composition and concentration of residual radioactivity in subsurface soils relative to unrestricted use criteria (NRC 2000a), including the requirement for subsurface soils to be surveyed and averaged using the approved AAR method developed in NRC supplemental guidance (NRC 1997).¹

¹ Molycorp has previously submitted under separate cover a technical basis document designed to demonstrate the applicability of the AAR method to the York, PA site and to derive the components of the AAR method that require site-specific information and calculations (MACTEC 2003)

2.0 DERIVATION OF THE SUM-OF-FRACTIONS CALCULATION

2.1 SITE-SPECIFIC CONCENTRATION GUIDELINE VALUES

The DP lists unrestricted use concentration criteria (or DCGLs) for total thorium (Th-232 + Th-228), natural uranium (U-238 + U-234), and Ra-226 in soil as follows:

- 10 pCi/g average Total Thorium (Th-232 + Th-228)
- 10 pCi/g average Natural Uranium (U-238 + U-234, assuming all daughters in equilibrium and includes 5 pCi/g Ra-226)
- 5 pCi/g average Ra-226²

These DCGLs are derived from the NRC's SDMP Action Plan (NRC 1992b) and are understood to be co-dependent concentration limits for which a SOF calculation is required in order to demonstrate compliance with the intent and basis of their derivation. These criteria deal with two naturally occurring radionuclide series (U-238 and Th-232) found commonly in ore bodies. The concentration limits for both natural uranium and natural thorium assume that all daughters (progeny) are present and in equilibrium. From a health risk perspective, the equilibrium condition is the worst case condition as it assumes the greatest amount of radioactivity that might be present within the decay series. Nonetheless, Molycorp committed in its response to the NRC's RAIs (Molycorp 1999b) that additional, separate criteria would be applied to uranium (U-238 + U-234) and Th-230 if, at some time during the York decommissioning project, significant disequilibrium conditions were identified.

Citing the NRC's Branch Technical Position (BTP) on *Disposal or Onsite Storage of Thorium or Uranium Wastes from Past Operations* (NRC 1981b), Molycorp proposed (Molycorp 1999b) and the NRC accepted (NRC 2000a) that the following additional concentration criteria should be applied in the event that a disequilibrium condition was found to exist:

- 30 pCi/g excess total uranium³ (U-238 + U-234), applicable to uranium activity present in concentrations in excess of Ra-226 activity in the same soil, and
- 14.3 pCi/g excess Th-230 (14.3 pCi/g Th-230 allowed to decay for a period of 1000 years will produce 5 pCi/g of Ra-226).

2.2 EQUILIBRIUM CONDITIONS

2 Since the specified natural uranium concentration guideline (10 pCi/g total U) accounts for 5 pCi/g of Ra-226 assuming that the entire decay series is in secular equilibrium, it is logical to conclude that the 5 pCi/g Ra-226 limit identified in the DP is intended to address the concentration of Ra-226 that may be present in excess of that associated with secular equilibrium with the U-238 decay series. This does not mean that more than 5 pCi/g of Ra-226 is tolerable within the context of the approved DP. Rather, it addresses the apportionment of Ra-226 in the event that a soil sample contains a higher concentration of Ra-226 than U-238 (or U-234). Such a condition represents a non-conservative departure from the assumption basis for the specified uranium limit. Ra-226 activity in excess of U-238 (U-234) is accounted for in the independent Ra-226 limit and is termed "excess Ra-226" in this report as it is in excess of that associated with secular equilibrium with its parent U-238.

3 The most conservative SDMP Action Plan criterion for uranium considered independently of its long-lived progeny.

2.2.1 Natural Thorium Decay Series

The natural thorium (thorium ore) decay series is comprised of isotopes with relatively short and, in general, progressively shorter half lives (Figure 1). This fact results in the rapid return to the secular equilibrium condition even if processes employed at the York facility might have contributed to preferential chemical separation of the various elemental forms found in the decay series. As a result, it is to be expected that concentrations of isotopes in the natural thorium decay series would be found to exist in secular equilibrium. As a result, Molycorp will continue to consider that Th-232 and Th-228 are in equilibrium.

2.2.2 Natural Uranium Decay Series

Because the natural uranium decay series has three relatively long-lived progeny isotopes (U-234, Th-230, and Ra-226) in its decay chain, it is reasonable to consider that these might not be in equilibrium with the parent U-238 isotope.

Figure 2 presents the first portion of the natural uranium decay chain down to the first long-lived progeny isotope, U-234. It is important to note that uranium enrichment, a process that has never been part of the operational history at the York, PA site, is necessary to create a significant disequilibrium condition between U-238 and U-234. No evidence to date suggests that there is any reason to consider that U-238 and U-234 might be in disequilibrium. As a result, Molycorp will continue to consider that they are in equilibrium. Furthermore, the 30 pCi/g excess total uranium concentration limit (which would apply in the event that a disequilibrium condition is found to exist) is protective in the case in which disequilibrium between U-234 and U-238 exists.

The radioactive decay of U-234 produces the next long-lived isotope in the decay series, Th-230 (Figure 3). Th-230 is an alpha emitter without a significant gamma signature. As a result, the only effective means of radiolytic measurement of Th-230 is with alpha spectroscopy. The cost, analytical turn around time, and the potential for variability in analytical results (due to a lack of complete homogeneity in the soil sample itself) associated with alpha spectroscopy analysis makes the actual measurement of Th-230 a poor choice when considering a large number of samples

On the other hand, it has previously been shown through alpha spectroscopy analysis of selected soil samples that Th-230 exhibits a strong and consistent correspondence with U-238 indicating that Th-230 is in equilibrium with its parents, U-238 and U-234 (RSI 2001, RSI 2002a). Consequently, there is no need to measure Th-230 directly. In assessing the concentrations of residual radioactivity in soil at the York, PA site, Molycorp will continue to consider that Th-230 is in equilibrium with both U-238 and U-234.

Ra-226 has an appreciably shorter half life than its parent, Th-230, but is still 1600 years. As a result, many thousands of years would be needed to restore equilibrium conditions between Th-230 and Ra-226 if disequilibrium exists. Previous analyses performed at the York, PA site have focused on establishing a central tendency estimate of the degree of disequilibrium between Ra-226 and the long-lived radionuclides in its parent chain, U-238, U-234, and Th-230, across the entire site (RSI 2001, RSI 2002a, RSI 2002b, S&A 2002). This was necessary because the previously employed analytical method used to assess compliance with the unrestricted use

concentration criteria for residual radioactivity in soil (low-resolution gamma spectroscopy using a sodium iodide detector) could not effectively resolve the lower energy gamma photons emitted by U-238 and Th-234.

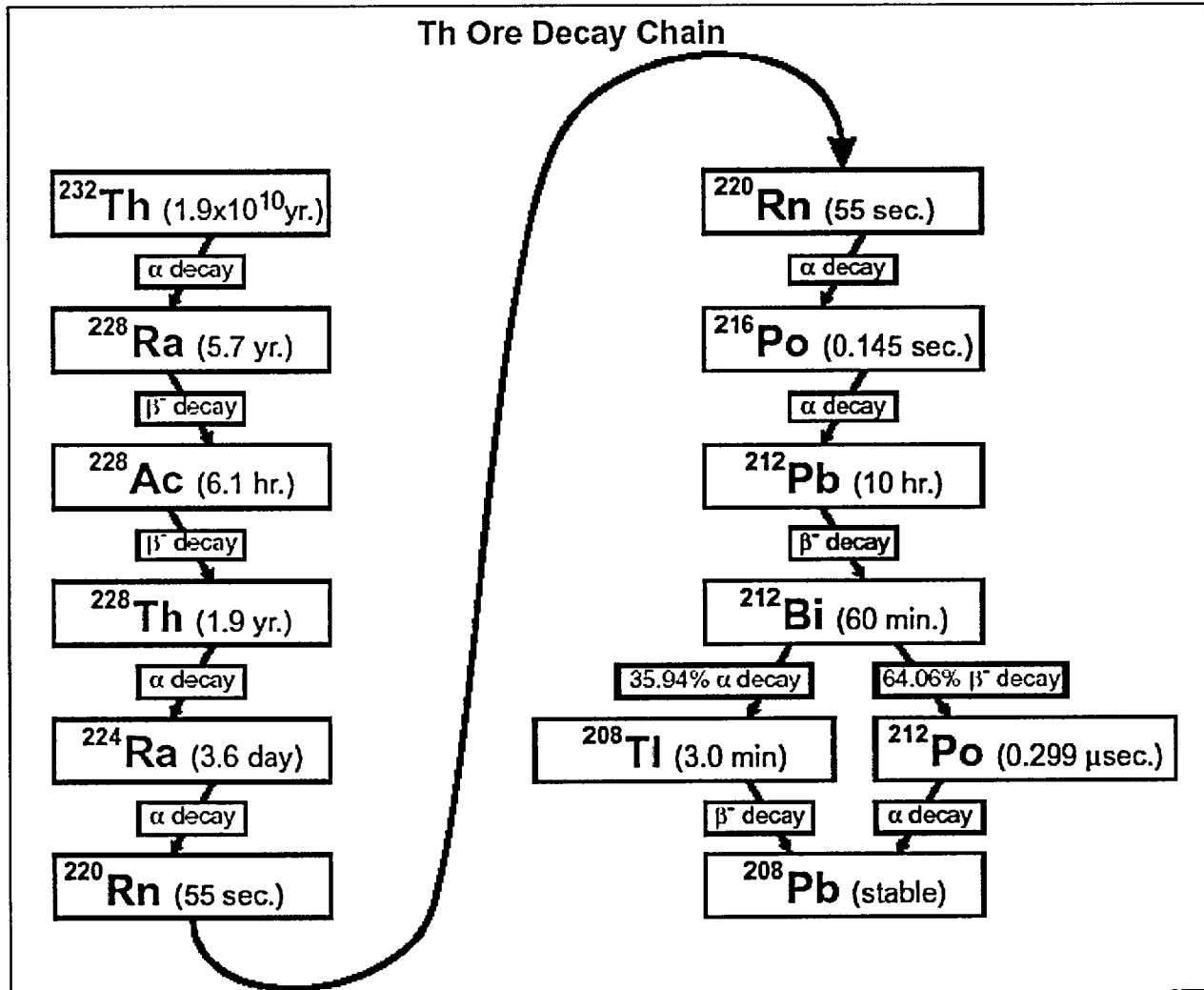


Figure 1 Thorium 232 Decay Chain

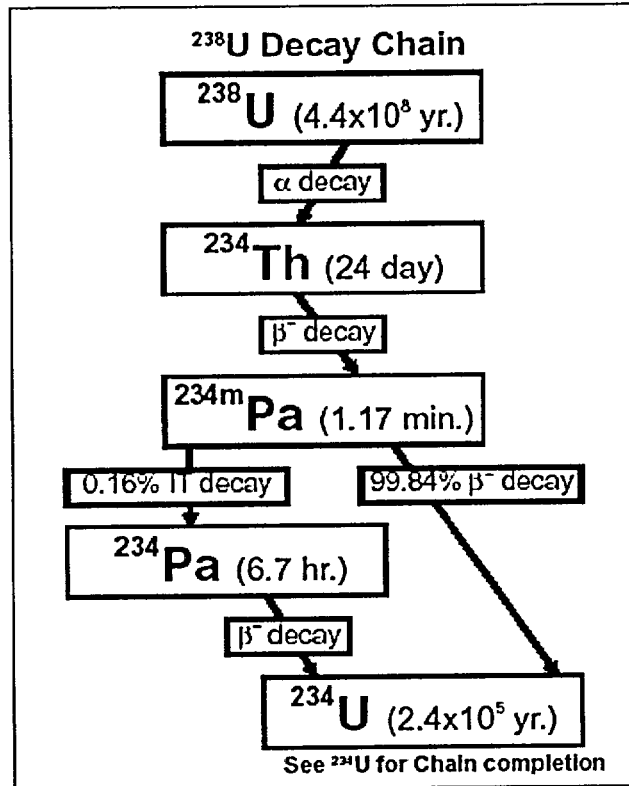


Figure 2 Uranium 238 Decay Chain

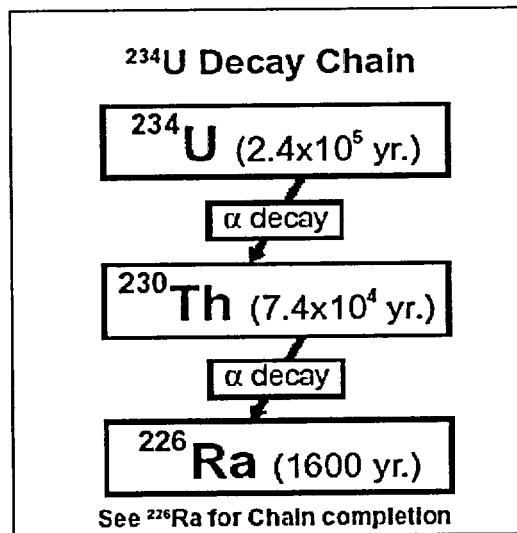


Figure 3 Uranium 234 Decay Chain

To avoid the need for inferring the concentration relationship between Ra-226 and its parents, Molycorp will employ a high-resolution gamma spectroscopy system that can resolve U-238 gamma emissions and measure it directly. Likewise, the measurement method will resolve and measure Ra-226 (and its significant gamma emitting progeny) directly.

The Ra-226 decay chain (Figure 4) involves very short-lived progeny, many of which have significant gamma signatures that can be used to indicate equilibrium within the chain and to infer the concentration of the parent Ra-226 isotope. A potential problem occurs with equilibrium within the Ra-226 decay chain arising from the fact that the first daughter of Ra-226 is an isotope of radon (Rn-222), a noble gas. As a gas, Rn-222 is potentially more mobile than Ra-226. Radon evolution from the soil itself may contribute to a disequilibrium condition between Ra-226 and the rest of the decay chain. Disequilibrium between Ra-226 and Rn-222 is not typically significant in situ, but can be exacerbated by the process of sampling and sample preparation.

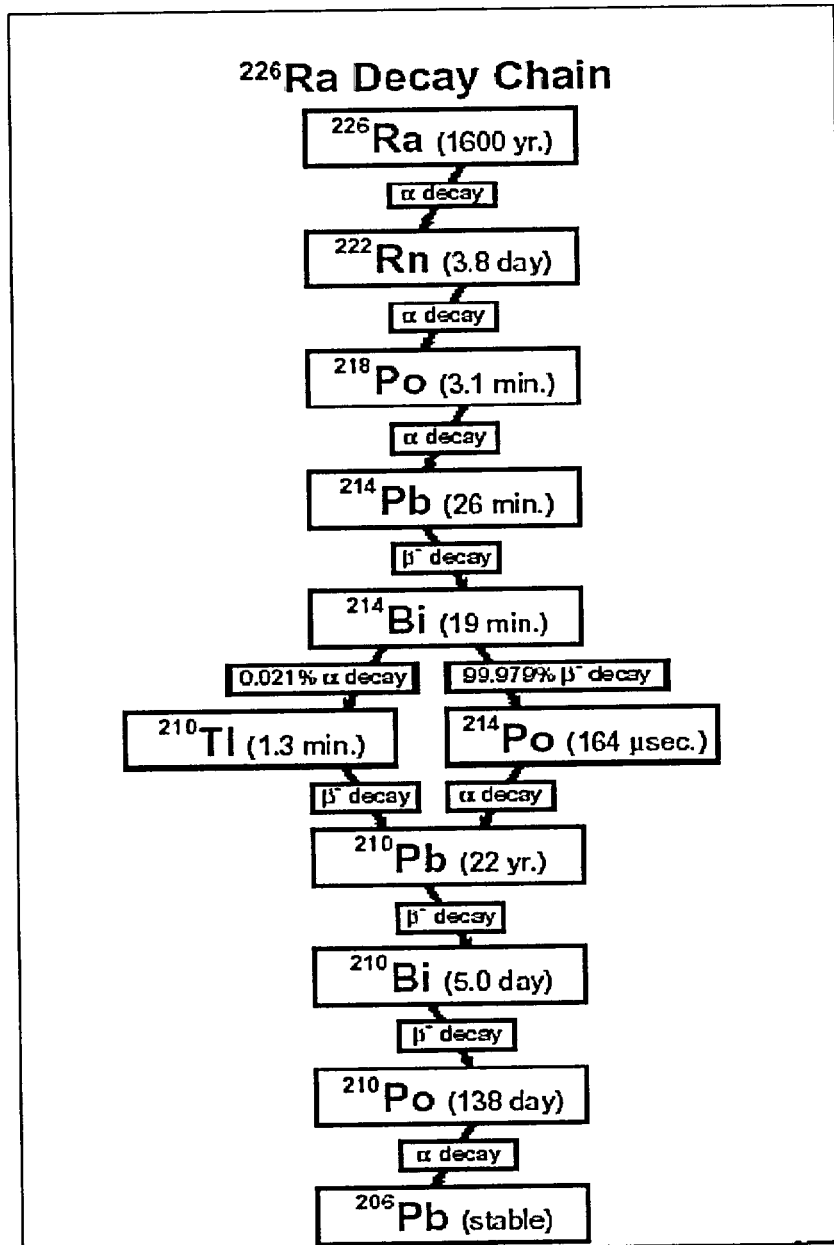


Figure 4 Radium 226 Decay Chain

Again, previously employed analytical methods dictated that a correction factor be applied to account for the potential disequilibrium condition between radon daughters (which were actually measured) and Ra-226 which was inferred. Molycorp has elected to depart from the analytical technique previously used and seal soil samples in an airtight container for a period of not less than 10 days to permit radon daughter ingrowth to reach equilibrium conditions with Ra-226.

2.3 DERIVATION OF THE EXPRESSION USED TO CALCULATE THE SUM-OF-FRACTIONS

The basic formulation of the SOF expression (NRC 1992, NRC 2000b) is shown as Equation 1.

$$\frac{C_1}{DCGL_1} + \frac{C_2}{DCGL_2} + \frac{C_n}{DCGL_n} \leq 1$$

Where :

C = concentration

$DCGL$ = guideline value for each individual radionuclide (1, 2, ..., n)

Equation 1

There are a total of five concentration-based DCGLs for residual radioactivity in soils at the York, PA site (NRC 2000a). The soil DCGLs are tabulated in Table 1.

Table 1 Residual Radioactivity DCGLs for the York, PA Site

Radionuclides	DCGL
Total thorium (Th-232 + Th-228)	10 pCi/g
Natural uranium (U-238 + U-234)—assuming all daughters in equilibrium and includes 5 pCi/g Ra-226	10 pCi/g
Excess Ra-226—applicable to Ra-226 activity present in excess of that associated with U-238 (assuming equilibrium with all of its daughters) in the same soil	5 pCi/g
Excess uranium (U-238 + U-234)—applicable to U-234 + U-238 activity present in concentrations in excess of Ra-226 activity in the same soil	30 pCi/g
Excess Th-230—applicable to Th-230 activity present in concentrations in excess of Ra-226 activity in the same soil	14.3 pCi/g

Equation 1 can be rewritten in terms of the site-specific soil DCGLs (Equation 2).

$$\frac{(Th - 232 + Th - 228)}{10} + \frac{(U - 238 + U - 234)}{10} + \frac{(Excess Ra - 226)}{5} + \frac{(Excess U - 238 + U - 234)}{30} + \frac{(Excess Th - 230)}{14.3} \leq 1$$

Equation 2

Since Th-232 and Th-228 are considered to be in equilibrium with each other, and U-238, U-234, and Th-230 are considered to be in equilibrium with one another, Equation 2 can be reduced and simplified (Equation 3, Equation 4)

$$\frac{(Th - 232)}{5} + \frac{(U - 238)}{5} + \frac{(Excess Ra - 226)}{5} + \frac{(Excess U - 238)}{15} + \frac{(Excess U - 238)}{14.3} \leq 1$$

Equation 3

$$\frac{(Th - 232)}{5} + \frac{(U - 238)}{5} + \frac{(Excess Ra - 226)}{5} + \frac{(Excess U - 238)}{7.32} \leq 1$$

Equation 4

For soils in which the Ra-226 activity is greater than the U-238 activity, excess Ra-226 will be assessed to be present in a concentration equal to the difference in activity (pCi/g) between Ra-226 and U-238. When excess Ra-226 is determined to be present, there can be no excess U-238 and so the excess U-238 term will reduce to zero. On the other hand, in soils where the U-238 activity is greater than the Ra-226 activity, excess U-238 will be assessed to be present in a concentration equal to the difference in activity (pCi/g) between U-238 and Ra-226. In this case, there can be no excess Ra-226 and so the excess Ra-226 term will reduce to zero.

3.0 ADJUSTMENTS TO ACCOUNT FOR BACKGROUND ACTIVITY CONTRIBUTIONS

As the prescribed soil DCGLs are expressed as concentrations in excess of background radioactivity, it is appropriate to adjust the measured activity in soil samples to account for background radioactivity.

In 1985, Oak Ridge Associated Universities (ORAU), under contract to the NRC, conducted a radiological assessment at the York, PA site. As part of that assessment, ORAU collected and analyzed a series of soil samples from unaffected off site areas in order to determine the concentrations of naturally occurring radionuclides (background) occurring soils in the vicinity of the site. The ORAU report (ORAU 1985) concluded that the mean background concentrations of Th-232 and Ra-226 in off site soils were 1.51 and 0.89 pCi/g, respectively. A set of additional off site soil samples designed to supplement the data set acquired by ORAU was collected and analyzed in September of 2001 (RSI 2001b). It was determined that the radioactivity in the supplemental background sample set was not statistically different from that

in the background data set previously acquired by ORAU. Based upon these results, Molycorp decided to adopt the background concentration values determined by ORAU. As these Th-232 and Ra-226 values have been in use throughout the final status survey process to date, it is Molycorp's intent to continue to use these to adjust measured concentrations for background contribution.

It is necessary, however, to make one additional assumption concerning the activity of U-238 in background, given that Molycorp intends to assay soil samples with the use of high-resolution gamma spectroscopy thus avoiding the assumption that there is a fixed linear correlation between U-238 and Ra-226 in impacted soils on site. It is reasonable to assume that the U-238 decay series is in secular equilibrium in non-impacted off site soils. In this case, it is reasonable to conclude that concentrations of U-238 and Ra-226 will be equivalent (on average) in background. Thus, when high-resolution gamma spectroscopy techniques are used to yield a measure of U-238 activity directly (rather than inferring U-238 activity from Ra-226 activity as was previously done), Molycorp will adjust total U-238 activity values by 0.89 pCi/g to account for U-238 activity in background.

4.0 SUMMARY

This technical basis document derives the SOF calculation that is appropriate and applicable to the radiological constituents found in soils at Molycorp's York, PA site and using high-resolution gamma spectroscopy with time allowed for radon and radon daughter products to reach secular equilibrium with Ra-226. It accounts for the three specifically stated concentration limits identified in the decommissioning plan (Molycorp 1999a) as well as the two additional limits agreed to in Molycorp's response to RAIs (Molycorp 1999b).

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