RESULTS AND EVALUATION OF RADIOCHEMICAL SAMPLING AT SIX WASTE MANAGEMENT, INC. CALIFORNIA LANDFILLS

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1.0 INTRODUCTION

1.1 Purpose

This report presents the results of radiochemical testing of leachate and groundwater at six Waste Management, Inc. (WM) landfills, as requested by the California State Water Resources Control Board (SWRCB) and applicable regions of the California Regional Water Quality Control Board (RWQCB). The data from all WM sites included in the SWRCB testing program are compiled and evaluated in this report, as discussed with, and approved by the SWRCB during an August 23, 2002 meeting between WM and SWRCB representatives. The intent of a combined report is to allow for more meaningful data interpretation. Because all of the radiochemical parameters measured as part of the SWRCB program occur naturally, identification of anomalous results is confounded by the inherent variability of radioactivity in the environment. Incorporation of all WM data in a single report is intended to provide more meaningful and proper consideration of naturally-occurring radiochemical sources and associated natural variability of radiochemical background levels.

The radiochemical testing program for the six WM landfills presented herein was a coordinated effort using consistent sampling procedures, analytical methods and data evaluation techniques. The data and information included in this report fulfill the SWRCB/RWQCB radiochemical testing and reporting requirements, summarized in Section 1.2, for the following WM landfills:

- Altamont Landfill and Resource Recovery Facility, Livermore, California
- Anderson Landfill, Anderson, California
- Bradley Landfill and Recycling Center, Sun Valley, California
- El Sobrante Landfill, Corona, California
- Kettleman Hills Facility, Kettleman City, California
- Redwood Landfill, Novato, California

The geographic locations of the above landfills are shown on Figure 1.

1.2 SWRCB/RWQCB Radiochemical Testing and Reporting Requirements

On April 25, 2002, the SWRCB issued a letter to each of California's RWQCBs requesting radiochemical testing of landfill leachate and groundwater for selected landfills in their respective regions. Each RWQCB, in turn, issued a letter directly to affected landfills on the SWRCB list. The list of affected landfills was modified by the SWRCB in a July 18, 2002 letter to Solid Waste Industry Group (SWIG) representatives, on which the RWQCBs were copied.

The purpose of the radiochemical testing program, as stated in the April 25, 2002 SWRCB letter is to "establish basic information on radioactivity in leachate and

groundwater beneath landfills in the State." In their letter, the SWRCB concluded that California landfills could be expected to contain naturally-occurring radioactivity, including radioactivity from naturally-occurring potassium-40 in food products such as bananas and salt substitutes. The SWRCB did not, however, make note of the expected presence of natural radioactivity in groundwater, particularly from uranium-238 and its decay products. Uranium-238 and its decay products are responsible for the greatest contribution of radioactivity observed in natural waters (Hem, 1992), and occur in such significant concentrations in natural waters that they are a major focus of the U.S. Environmental Protection Agency (EPA) under the federal Safe Drinking Water Act (EPA, 2000).

Program requirements outlined by the SWRCB in their April 25, 2002 letter to the RWQCBs were general in nature and described the types of samples to be collected and radiochemical analyses to be performed. Each RWQCB, in turn, communicated the general SWRCB requirements to the affected landfills, setting RWQCB-specific deadlines and, in some cases, prescribing specific sampling locations and reporting requirements. Table 1 summarizes the general SWRCB program requirements, as well as the specific RWQCB program requirements, for the six WM landfills presented in this report. As shown on Table 1, original program deadlines were extended until January 2003 with the earliest deadline of January 15, 2003.

1.3 Technical Concerns

The California Solid Waste Industry Group (SWIG) submitted a letter to the SWRCB on June 20, 2002, detailing several regulatory and technical concerns regarding the SWRCB's proposed radiochemical testing program. The primary concern, detailed in the June 20, 2002 SWIG letter, was the lack of clearly defined project objectives, which precluded development of an effective sampling plan and data evaluation strategy. The SWIG letter noted that if the SWRCB's premise is that the program will identify sites where inadequate load checking procedures enabled acceptance of radioactive wastes, then the program results will likely be confounded by naturally-occurring radionuclides and yield potentially erroneous conclusions. In particular, the SWIG noted that the SWRCB program does not include a procedure for proper data evaluation, including the necessary consideration of naturally-occurring radionuclides. The following major points were communicated:

- Evaluation of radiochemical data is meaningless without the ability to compare the results to natural background levels. The distribution of naturally-occurring radionuclides depends on the distribution of rocks from which they originate and the processes which concentrate them (USGS, 1994). Previous studies by the U.S. Geological Survey (USGS) suggest that characterization of background levels is difficult due to the high variability of radionuclide concentrations as a function of lithology, depth, age of water, etc. Additionally, inorganic parameters such as chloride, calcium, barium, TDS, and dissolved oxygen can affect the mobility of radionuclides in groundwater, further confounding the development of background levels.

- The current program utilizes data evaluation methods and associated trigger mechanisms tied to comparison with drinking-water standards. This approach lacks the pragmatic decision-making process necessary in the evaluation of naturally-occurring constituents in groundwater, and it can falsely generate

erroneous conclusions and unwarranted further testing. These concerns are substantiated by recent USGS studies which have demonstrated that many of the nation's drinking-water aquifers exhibit natural radionuclide concentrations in excess of drinking-water standards (Focazio, et. al, 2001; USGS, 1996).

- Similar to the procedures developed for evaluating naturally-occurring inorganic parameters in groundwater (such as iron or manganese), statistical methods would be required to properly characterize background radionuclide levels before any meaningful data evaluation could be completed. As prescribed under CCR Title 27 and Federal Subtitle D Regulations, development of rigorous statistical procedures is required to assess deviations from background conditions. Since radionuclides are also naturally-occurring, similar types of procedures are necessary for a meaningful evaluation of radionuclide data. However, this issue is further complicated by the physical characteristics of radionuclides in that they will undergo natural transformations not associated with landfilling activities. In addition, it is extremely important to note that spatial variability is a significant concern at most landfill sites, and simple upgradient to downgradient comparisons will have a high probability of yielding erroneous conclusions.

- The current program does not indicate how the data to be collected will be utilized and disseminated to the general public. It is imperative that the data be released in a manner that assures a fair and balanced presentation in the context of natural radioactivity. There should be emphasis placed on understanding and explaining the naturally-occurring sources of radioactivity. Without proper context, this program has the potential to generate misleading and alarming information.

A meeting was held between SWIG representatives and SWRCB staff on August 23, 2002 to discuss the concerns expressed by the SWIG in their June 20, 2002 letter to the SWRCB. Based on the meeting, the SWRCB agreed to (1) allow a combined technical report that would allow a more comprehensive presentation of the data collected and (2) consider review of the data collected by a SWRCB-selected technical review panel, so the data can be evaluated in proper context (i.e., with respect to natural radioactivity) and disseminated to the public in a manner that assures a fair and balanced presentation in the context of natural radioactivity.

2.0 NATURALLY-OCCURRING RADIONUCLIDES

This section presents information on potential natural sources of the radionuclides that are the focus of this testing program. Relevant information on background geochemical issues is also presented. The purpose of this section is to provide a more meaningful context in which the radiochemical data collected for this program can be evaluated.

2.1 Background Issues

As expressed in the June 20, 2002 SWIG letter to the SWRCB, the absence of a meaningful standard against which the collected radiochemical data will be compared is a primary concern for this program. Naturally-occurring radionuclides are ubiquitous trace elements found in rocks and soils (Focazio, et. al., 2001). Because all of the radiochemical parameters measured as part of this program occur naturally, it is difficult to assess whether the reported values are anomalous with respect to natural background levels.

Maximum contaminant levels (MCLs) developed by the EPA can be used to assess risk for drinking water but are not particularly relevant to non-potable groundwater and landfill leachate. Furthermore, exceedance of a water quality standard (without a thorough understanding of natural background conditions) cannot be used to identify potential impacts to the environment resulting from operations at a landfill or any other type of facility. It is important to understand that exceedance of an MCL in leachate would be of no great surprise, nor of any real significance. Landfill leachate typically contains chemicals in excess of water quality standards, and, for that reason, current landfill design standards require construction of a leachate collection and removal system (LCRS) to prevent leachate from entering waters of the State.

Extensive research on background radioactivity in selected aquifer systems has been conducted by the EPA and USGS in support of the final Radionuclides Rule recently developed under the Safe Drinking Water Act (EPA, 2000). As part of USGS's research for the final Radionuclides Rule, a national reconnaissance survey of selected radionuclides in public groundwater supplies was performed (Focazio, et. al., 2001). Ninety-nine samples were collected from 27 states and 8 physiographic provinces, with 94 of those samples collected from wells in use for drinking water. The results showed that 21 of the 99 samples collected exceeded the MCL for combined radium-226 and radium-228 due to natural radioactivity (Focazio, et. al., 2001). Another USGS study for the National Water Quality Assessment Program showed that 80 percent of 267 samples from drinking-water wells in the Lower Susquehanna and Potomac River Basins exceeded the proposed drinking-water standard for radon, due the decay of natural uranium (USGS, 1996). These results further highlight the ubiquitous nature of the radionuclides of concern and the problematic aspects of comparing non-potable groundwater and leachate samples from landfills to drinking-water standards.

2.2 Pertinent Radionuclide Considerations

The natural radionuclides include the primordial elements (e.g., uranium and thorium) that were incorporated into the Earth's crust during its formation, the radioactive decay products of these primordial elements, and radionuclides that are formed in the

atmosphere by cosmic ray interactions, e.g., tritium (EPA, 2000). Most natural radioactivity is generated through the progressive decay of naturally-occurring radionuclides through three decay series, beginning with uranium-235, thorium-232, and uranium-238, and referred to as the "Actinium Series," "Thorium Series," and "Uranium Series," respectively (see Figures 2, 3, and 4).

The Uranium Series is the most predominant contributor to radioactivity in groundwater (Hem, 1992). However, other lighter radioisotopes (e.g., potassium-40 and tritium) can be present in measurable quantities in natural water. For the heavier isotopes (e.g., from the above decay series), each transformation emits an alpha- or beta-particle unit (and, in some cases, gamma radiation), creating daughter products, until stability is achieved with one of three stable isotopes of lead. Figures 2, 3, and 4 show the half-life and type of decay (i.e., alpha or beta) for each of the daughter products.

The occurrence of radionuclides in groundwater depends first on the presence and solubility of the parent products. Each radioactive decay product has its own unique chemical characteristics that differ from the radionuclide parent. Consequently, the occurrence and distribution of a parent radionuclide in solution does not necessarily indicate the presence of a daughter radionuclide in solution (EPA, 2000).

Geochemical information on the behavior of the radiochemical parameters of interest for this study in groundwater is summarized below. The information is brief and focused on issues pertinent to this study. For more detailed information, the reader is referred to general radiochemistry textbooks as well as the research performed by EPA and USGS in support of the final Radionuclide Rule under the Safe Drinking Water Act.

2.2.1 Uranium

Uranium is found in concentrated amounts in granite, metamorphic rocks, lignites, monazite sand and phosphate deposits, as well as in the uranium-rich minerals of Uraninite, Carnotite, and Pitchblende (EPA, 2000). Natural uranium in rock contains three isotopes: uranium-234 (0.006% by weight), uranium-235 (0.72%) and uranium-238 (99.27%); the activity to mass ratio of the sum of the three radioisotopes in rock is 0.68 pCi/ug (EPA, 2000). These crustal abundances of uranium are not duplicated in groundwater. Uranium-234 is enriched in water relative to rock when standardized to uranium-238 in the water. EPA uses a uranium-234/uranium-238 ratio of approximately 1.0 to indicate the presence of natural uranium in groundwater; this natural uranium isotopic ratio may vary regionally, as well as seasonally, with uranium-234/uranium-238 ratios ranging from 0.9 to 1.3 (EPA, 2000).

Uranium concentrations in groundwater are predominantly redox controlled. Uranium forms soluble complexes under oxygen-rich conditions, particularly with carbonates, causing higher concentrations in zones of recharge due to the relatively high solubility of the oxidized uranium (VI) species (EPA, 2000; Clark and Fritz, 1997). Alternatively, uranium precipitates from groundwater under oxygen-poor conditions and can be concentrated in secondary deposits (Cothern and Rebers, 1990). Consequently, uranium concentrations in water can be expected to range widely with different geologic environments. USGS studies show that uranium is preferentially concentrated in wetland environments where uranium-rich rocks and soils occur (USGS, 1994).

2.2.2 Radium

Radium-226 and radium-228 are derived from uranium and thorium, respectively. Radium-226 is the sixth member of the Uranium Series, has a half-life of approximately 1,600 years, and decays by alpha-particle emission (see Figure 4). Radium-228 is the second member of the Thorium-232 series, has a half-life of about 5.7 years, and decays by beta-particle emission (see Figure 3).

Because uranium and thorium are ubiquitous components of rocks and soils, radium radionuclides are also ubiquitous trace elements in rocks and soils. However, the chemical behaviors of the radium isotopes differ significantly from their parent radioisotopes. As noted above, uranium solubility is primarily redox controlled with elevated uranium concentrations in zones of higher oxidation. Alternatively, thorium is extremely insoluble (Cothern and Rebers, 1990) and, thus, is not subject to mobilization in most groundwater environments. Groundwater with high radium-226 levels tends to have low uranium levels and vice-versa, even though uranium-238 is the parent of radium-226 (EPA, 2002).

Radium isotopes tend to be most mobile in reducing groundwater that is chloride-rich with high concentrations of total dissolved solids (Zapecza and Szabo, 1986, Kramer and Reid, 1984). Radium-228 activities tend to be the highest in arkosic sand and sandstone aquifers (Cothern and Rebers, 1990). Radium behaves similarly to other divalent cations such as calcium, strontium, and barium. Therefore, in aquifers with limited sorption sites, radium solubility can be enhanced by the common ion effect in which competing cations are present in abundance and occupy sorption sites keeping the radium in solution (EPA, 2000). For example, relatively high concentrations of radium were found to be associated with groundwater in recharge areas that were geochemically affected by agricultural practices. Such groundwater was strongly enriched with competing ions such as hydrogen, calcium, and magnesium (Szabo and dePaul, 1998). For this reason, elevated concentrations of radium could occur in high-TDS groundwater samples and landfill leachate samples.

2.2.3 Potassium-40

Naturally-occurring potassium is ubiquitous in the environment, a major crustal element, and a major nutrient for living organisms. The radioactive component of natural potassium (i.e., potassium-40) comprises 0.012% by mass of naturally-occurring potassium. Potassium-40 decays directly to stable calcium via predominantly beta emission and has a half-life of 1.27 billion years (Friedlander, et. al., 1964).

Potassium-40 is a strong beta emitter that produces surprisingly high activity levels in rocks, soils, and many types of food. It is well documented that foods such as bananas, artichokes, nuts, etc., emit activity levels on the order of 3,000 pCi/kg or more due to their natural potassium-40 content. Natural potassium chloride (KCl, a common salt substitute) emits activities on the order of 500 pCi/g, or more than 20 times background soil, due to its potassium-40 content. A salt shaker containing 100g of KCl salt substitute would exhibit a total activity of approximately 40,000 pCi. Though such levels can be a significant contributor to both internal and external beta-particle exposure, potassium-40 is generally not of environmental concern because the absorption of potassium by the human body is under strict homeostatic control and, therefore, not

influenced by variations in environmental levels (Canada Department of National Health, 1995).

The mobility of potassium, and hence potassium-40, in the groundwater system is controlled primarily by the dissolution of potassium-bearing silicate minerals and cation exchange reactions involving the adsorption of potassium ions. Although potassium is a major crustal element, it seldom occurs in high concentrations in groundwater due primarily to the general stability of potassium silicates and irreversible cation exchange reactions with clay minerals (Hem, 1992). Alternatively, potassium concentrations in landfill leachate can reach relatively high concentrations (i.e., >100 mg/L) due to the dissolution of highly soluble potassium salts present in organic matter, including food and green waste, as well as fertilizers. The most common potassium salt, KCl, is highly soluble in water and would be expected to be present in most landfill leachates in measurable concentrations and, therefore, act as a potentially significant source of natural radioactivity. Mass-activity calculations indicate that approximately 1 pCi/L of activity can be expected for every 1 milligram of dissolved potassium in water with the vast majority of that activity being beta-particle activity (potassium-40 decay is 89% beta emission). With an extremely long half-life, potassium-40-related beta activity may be expected to persist indefinitely in landfill leachate.

2.2.4 Tritium

Tritium is the radioactive isotope of hydrogen (^{3}H). It occurs both naturally and as a man-made by-product in industrial or medicinal radioactive materials, as wells as fallout from atmospheric nuclear weapon tests from the 1950s to the 1970s. Tritium has a half-life of 12.43 years (Unterweger et al, 1980), is produced naturally by cosmic-ray interactions with the atmosphere, and is a very low-energy beta emitter. Because of its very low energy level, tritium is used for a variety of unregulated industrial and consumer products that contain either gaseous tritium or luminous tritium-bearing phosphor (e.g., watches, compasses, depth gauges, exit signs, instrument dials, and street signs; Eisenbud, 1987).

Input of anthropogenic tritium into the global groundwater system occurred in a series of spikes following periods of atmospheric testing of nuclear weapons that began in 1952 and reached a peak in 1963-1964, when tritium concentrations in precipitation reached levels on the order of 10^{5} pCi/L (Clark and Fritz, 1997). Concentrations of tritium in precipitation have decreased significantly since the mid-1960's bomb-testing peak, except for some relatively small increases from French and Chinese tests in the late 1970s (USGS, 2002). However, even at peak levels, anthropogenic tritium fallout was not regarded as a radiological threat to health (Clark and Fritz, 1997). Canada imposes a tritium drinking-water standard of 7,000 Bq/kg (Clark and Fritz, 1997), which is approximately 190,000 pCi/L. The EPA imposes a more conservative MCL for tritium in drinking water of 20,000 pCi/L.

Elevated tritium levels in landfill leachates are well documented but are generally regarded to be of low radiological significance (Hicks, et. al., 2000). In the United Kingdom, a study by Robinson and Gronow (1996) noted a mean tritium value of 7,714 tritium units (or approximately 25,000 pCi/L) for 30 landfills. That study concluded that "the source of tritium is likely to be general household and/or commercial disposal of tritium-containing items which have fulfilled their useful life, and are contained within

general waste streams being landfilled." Particular focus was paid to gaseous tritium light sources (GLTSs) used to construct gaseous tritium light devices (GLTDs) that are commonly used for fire-safety "exit" signs. One GLTD can contain up to 1 Curie of tritium (Hicks, et. al., 2000), which can be released only if the GLTD is damaged.

Tritium in water is highly mobile as the tritium atom becomes part of the water molecule. As part of the water molecule, tritium movement in the groundwater system is unimpeded by chemical reactions or other physicochemical processes that are known to affect other radionuclides (Hull and Hechanova, 2002). However, tritium does not persist in the environment due to its relatively short half-life; approximately 75% of tritium in groundwater would decay in 25 years.

2.2.5 Gross Alpha/Beta-Particle Activity

The measure of gross alpha/beta-particle activity is (with some analytical limitations, including those described in Section 3.2.1) the sum of activities of alpha-emitting isotopes and beta-emitting isotopes, respectively. In natural groundwater, uranium-238, radium-226, and polonium-210 are the principal alpha emitters in the dissolved phase (EPA, 2000). Radon-222 is an alpha emitter in gaseous phase and is not measured as part of gross alpha-particle activity in water. In natural groundwater, radium-226 and potassium-40 are the principal beta emitters (EPA, 2000; Welch et al., 1995). Tritium and carbon-14 are low-level beta emitters that are not measured (in the case of tritium) or partially measured (in the case of carbon-14) as part of the gross beta-particle activity analysis (STL, personal communication). Drinking-water standards developed by EPA for gross beta exclude any beta contribution by potassium-40. According the final Radionuclide Rule for public water supplies, only when the beta-particle activity minus the naturally-occurring potassium-40 activity in a drinking-water supply sample exceeds 50 pCi/L must the system speciate the beta emitters (EPA, 2001).

3.0 SAMPLING AND ANALYSIS

This section presents the following information: (1) field sampling procedures (Section 3.1); (2) analytical procedures (Section 3.2); and (3) descriptions of sampling locations for each landfill included in the testing program (Section 3.3). Plot maps showing basic landfill features, groundwater flow direction, and radiochemical sampling locations are presented for each landfill in Figures 9 through 14.

3.1 Sampling Procedures

Sampling procedures were developed for this program based on consultations with the analytical laboratory, consultations with the field sampling team, and review of "suggested sampling protocols" provided by the SWRCB. Appendix A presents the detailed sampling procedures implemented for this program. Key aspects of the sampling procedures were:

- Samples were collected unfiltered and unpreserved in the field.
- Sample containers were pre-cleaned by the manufacturers to meet EPA Specifications and Guidelines for Contaminant Free Sample Containers, and come with a certificate of analysis. One-liter high density Polyethylene (HDPE) containers were used for all parameters.
- Samples were filtered through a 0.45-micron filter. Preservation was performed by the analytical laboratory on receipt of the sample and within five days of sample collection.
- One field blank was prepared for each sampling program using tested, reagent-grade water supplied by the analytical laboratory.
- Groundwater samples were collected in accordance with procedures outlined in the approved groundwater sampling plan for each site (i.e., wells were purged, field monitored, and sampled according to established standards).

3.2 Analytical Procedures

The analytical procedures used for this program were consistent for the landfills presented herein and were developed in consultation with analytical laboratories, SWIG, and technical consultants. The analytical protocols incorporated the guidance provided by the SWRCB and RWQCB (i.e., types of analyses and thresholds for gross alpha). All laboratory measurements of radiochemical parameters were performed by Severn Trent Laboratories (STL) in St. Louis, Missouri. STL is a California State Department of Health Services (DHS)-certified laboratory for all the required radiochemical analyses.

Figure 5 presents a flow chart summarizing the analytical protocol used for this program. As shown on Figure 5, samples were analyzed for tritium by EPA Method 906.0 and initially screened for gross alpha/beta-particle activity using EPA Method 900.0. As requested by the SWRCB, if gross alpha-particle activity was detected above 5 pCi/L but less than 15 pCi/L, then the sample was analyzed for radium-226 and radium-228 using

EPA Method 9315 and EPA Method 9320, respectively. If gross alpha-particle activity was detected above 15 pCi/L, then the sample was analyzed for uranium using method DOE RP-725 for speciated uranium (uranium-234, uranium-235, and uranium-238) in addition to radium-226 and radium-228. Method references are provided in the Certified Analytical Reports (Appendix B); detailed laboratory standard operating procedures (SOPs) for the methods are available on request.

In most cases, the detection limits for gross alpha-particle activity by EPA Method 900.0 were above 15 pCi/L, precluding the use of this method for identifying the need for any additional radium and uranium analyses. In order to achieve detection limits below 5pCi/L and support the ability to make a decision on the need for additional radium and uranium testing, gross alpha-particle activity was measured using SMWW Method 7110 (gross alpha by coprecipitation) for those samples where EPA Method 900.0 lacked sensitivity.

Problems with the analytical sensitivity of EPA Method 900.0 are described in further detail in Section 3.2.1. Details on interpreting analytical results are presented in Section 3.2.2.

3.2.1 Limitations on Gross Alpha/Gross Beta-Particle Activity Measurements

High concentrations of dissolved solids in several samples resulted in increased detection limits for those samples. This situation occurred for all landfill leachate samples, as well as many groundwater samples. As discussed previously, many landfills are situated in areas of naturally poor groundwater quality, with elevated concentrations of dissolved solids.

The primary non-radioactive interference for the gross alpha/beta-particle activity analysis, using a gas flow proportional counter (GFPC), is residue mass (derived from dissolved solids). As the sample aliquot is concentrated under acidic conditions, the solid material contained in the sample, as well as any crystallization of salts, is deposited on the stainless steel planchet. This mass effectively blocks a portion of the alpha particles and, to a lesser degree, beta particles. This blocking effect is referred to as “self-absorption of attenuation.” (STL personal communication, Dec. 9, 2002).

During calibration of a GFPC, an attenuation calibration is performed. This measures the effect of increasing mass to the efficiency of the detector. Figures 6 and 7 show attenuation charts for alpha and beta, respectively. The alpha attenuation curve shows a significant reduction in counting efficiency as the mass increases. This is due primarily to the relatively large size of alpha particles and their resulting inability to escape from the residue mass. The beta attenuation curve demonstrates a reduced effect because the relatively higher energy and smaller size of beta particles are less inhibited by the residue mass, relative to alpha particles.

The internal standards for the analysis of samples for gross alpha/beta-particle activity allow a maximum residual mass of 100 mg. At this threshold, the analytical laboratory concludes that the reduction in counting efficiency no longer permits an accurate assessment of sample activity (STL personal communication, Dec. 9, 2002). During the preparation of samples for analysis, a prescreen is performed to determine the aliquot size that would approximate a 70 mg residual mass. In samples with high solids

content, this aliquot can be only a few milliliters. This reduction in aliquot size also increases the uncertainty (2 sigma error) and detection limit. A copy of the equations used for the generation of gross alpha/beta results is provided for verifying the effect of the reduced aliquot to final results (see Figure 8).

3.2.2 Interpretation of Numerical Results

Radiochemical parameter results for this program are reported in terms of "activity" which is defined as the number of nuclear transformations (i.e., decay) of a radioactive substance which occur in a specific time interval (EPA, 1991). Results are expressed in terms of activity because it is not the mass of the radiochemical parameter of interest, but the radioactive emissions. Activity is related to half-life, which is defined as the length of time required for a radionuclide to lose 50% of its activity by decay. A radionuclide with a shorter half-life (e.g., radium-226 at 1,600 years) emits higher activity whereas a radionuclide with a longer half-life (e.g., uranium-238 at 4,500,000,000 years) emits lower activity (EPA, 2000). A radionuclide half-life can range from millionths of a second to billions of years (NRC, 2001). The decay rates of the radionuclides of interest for this study were previously discussed in Section 2.2.

The specific activity unit reported for data presented herein is picocuries per liter (pCi/L). One Curie is equal to a nuclear transformation rate of 3.7×10^{10} decays per second. One picocurie is equal to 10^{-12} Curies, which equates to one decay per 27 seconds or 2.2 decays per minute. Analytically, the laboratory measures the minimum detectable activity (MDA) for a specific analytical procedure. The MDA is the smallest amount of activity that can be measured given the conditions of a specific sample. The MDA is reported at the 95% confidence level, meaning that there is a 5% chance that a false signal was reported as activity, and a 5% chance that true activity went undetected (STL SOPs).

The MDA, which is purely a measure of activity (e.g., picocuries), is converted to activity-concentration units (i.e., picocuries per liter) by calculating the activity (i.e., picocuries) per unit volume (i.e., liters of aqueous sample). The minimum activity concentration is designated the "MDC" (minimum detectable concentration) on the laboratory reports and can be viewed as a traditional analytical reporting limit. All results are reported in activity concentration units (i.e., pCi/L) and expressed with an error term that expresses uncertainty in the form of +/- two standard deviations. The statistical errors intrinsic to the counting of a sample decrease the longer a sample is counted (i.e., the number of counts in the region of interest go up, thus giving a better representation of the actual number). Non-detected results can sometimes be expressed as negative values since the activity measurement incorporates the subtraction of background radioactivity. The reporting of a negative value is not unusual given the uncertainties associated with both the measurement of radioactivity in the actual sample as well as in the background sample.

For purposes of this report, results are presented in a manner typical of environmental analyses. That is, numerical results are reported without an error term and non-detected values are simply expressed as less than the MDC. The CARs (Appendix B) include the numerical error terms and numerical results below the MDC for interested reviewers.

3.3 Sample Locations and Rationale

The following section summarizes the sampling locations at each of the six WM landfills included in the radiochemical testing program. The Central Valley RWQCB prescribed specific sampling locations for Altamont Landfill and Anderson Landfill. For the other sites, sampling locations were chosen based on the criteria provided in RWQCB correspondence.

3.3.1 Altamont Landfill

Samples collected for radiochemical testing at the Altamont Landfill were selected using the criteria set forth in the May 21, 2002 Central Valley RWQCB letter. The following sample locations were selected:

- LS: the Unit 1 leachate sump
- VD: the Unit 1 valley subdrain
- GWIB: the Unit 1 groundwater interceptor barrier
- Well E-05: previously-impacted groundwater monitoring well providing information on landfill-impacted groundwater
- Well E-07: previously-impacted monitoring well providing information on landfill-impacted groundwater
- Well E-20B: VOC-impacted monitoring well providing information on landfill-impacted groundwater

In addition, a groundwater sample from monitoring well MW-4A was collected to provide information on background radiochemistry. Sample locations are shown on Figure 9.

3.3.2 Anderson Landfill

Sampling locations at the Anderson Landfill were selected using the criteria set forth in the May 21, 2002 letter from the Central Valley RWQCB. The following sampling locations were selected:

- MW-1: upgradient groundwater monitoring well providing information on background radiochemistry
- MW-2: non-impacted groundwater monitoring well downgradient of the WMU
- MW-4A: non-impacted groundwater monitoring well downgradient of the WMU
- MW-5: non-impacted groundwater monitoring well downgradient of the WMU
- L-WMU-2B: WMU-2B leachate collection system

Sample locations are shown on Figure 10.

3.3.3 Bradley Landfill

Sampling locations at the Bradley Landfill were selected using the criteria set forth in the June 7, 2002 letter from the Los Angeles RWQCB. The following sampling locations were selected:

- Well 4915A: upgradient groundwater monitoring well providing information on background radiochemistry
- Well 4916H: VOC-impacted groundwater monitoring well providing information on landfill-impacted groundwater
- Leachate Sump A
- Leachate Sump B
- Leachate Sump E

Sample locations are shown on Figure 11.

3.3.4 El Sobrante Landfill

Sampling locations at El Sobrante Landfill in were selected using criteria consistent with other landfills as specific written guidance was not sent by the Santa Ana Region RWQCB until after sampling was performed. Initially, El Sobrante Landfill received verbal direction from the Santa Ana RWQCB to proceed with the program as described in the April 25, 2002 SWRCB letter.

Samples were collected at the following locations:

- Well MW3-1: upgradient well providing information on background conditions
- Well MW-2: VOC-impacted groundwater monitoring well providing information on landfill-impacted groundwater
- LCRS Sump #2
- LCRS Sump #3

LCRS Sump #1 was not sampled because it did not contain liquid at the time of sampling. Sample locations are shown on Figure 12.

3.3.5 Kettleman Hills Facility

Sampling locations at the Kettleman Hills Facility (KHF) in Kettleman City, California, were selected using the criteria set forth in the May 2, 2002 letter from the Central Valley RWQCB and verbal guidance to Mr. Paul Turek from Messrs. Shelton Gray and James Dowdall of the Central Valley RWQCB. Focus was placed on the active WMUs at B-18 and B-19 because there are no known landfill-related impacts to groundwater at the KHF. Landfill B-18 is the only hazardous waste landfill evaluated in this study. Specifically, the following KHF sample locations were selected for radiochemical analysis:

- Well K67: non-impacted well monitoring groundwater beneath landfill B-18
- Well K71: non-impacted well monitoring groundwater beneath landfill B-18
- Well K68: non-impacted well monitoring groundwater upgradient of landfill B-18 and representing background
- Leachate Sample B181AP: primary LCRS in sump 1A of landfill B-18
- Well K69: non-impacted well monitoring groundwater beneath landfill B-19
- Well K70: non-impacted well monitoring groundwater beneath landfill B-19
- Well K41: non-impacted well monitoring groundwater upgradient of landfill B-19 and representing background
- Leachate Sample B191AP: primary LCRS in sump 1A of landfill B-19

Sample locations are shown on Figure 13.

3.3.6 Redwood Landfill

Sampling locations at the Redwood Landfill were selected using the criteria set forth in the June 5, 2002 letter from the San Francisco RWQCB. The following sampling locations were selected:

- Well MWH-8: upgradient well providing information on background conditions
- Well MWH-19: non-impacted well monitoring groundwater downgradient of landfill
- Leachate pump B (LD#B)
- Leachate pump C (LD#C)
- Leachate pump D (LD#D)

Sample locations are shown on Figure 14.

4.0 RESULTS

Table 2 summarizes the radiochemical results for all samples collected at the six WM landfills included in this program. The CARs are presented in Appendix B and a diskette containing the data summarized in Table 2 is included in Appendix C. For each landfill, individual sample points, sample types, dates of collection, analytical methods, and units are represented on Table 2. In addition, a calculated TDS value is presented for each sample, based on the residue mass measured for the gross alpha/beta analysis.

The following sections summarize the landfill-specific results.

4.1 Altamont Landfill

Groundwater Samples (4 samples):

- Gross alpha-particle activity was detected in all samples, ranging from 3.28 pCi/L (E-07) to 11.8 pCi/L (E-20B).
- No radium isotopes were detected in samples from wells E-05, E-20B, and MW-4A, which were analyzed for radium-226 and radium-228 due to gross alpha results exceeding 5pCi/L.
- No gross beta-particle activity was detected.
- No tritium was detected.
- No uranium analyses were necessary or performed.

Leachate Samples (3 samples):

- Gross alpha-particle activity was detected in all samples, ranging from 2.5 pCi/L (VD) to 5.9 pCi/L (GWIB).
- Radium-226 (0.47 pCi/L) was detected in GWIB, which was analyzed for both radium-226 and radium-228 due to the gross alpha result exceeding 5pCi/L. No radium-228 was detected.
- No gross beta-particle activity was detected.
- No tritium was detected.
- No uranium analyses were necessary or performed.

4.2 Anderson Landfill

Groundwater Samples (4 samples):

- No gross alpha-particle activity was detected.
- No gross beta-particle activity was detected.
- No tritium was detected.
- No radium analyses were necessary or performed.
- No uranium analyses were necessary or performed.

Leachate Samples (1 sample):

- No gross alpha-particle activity was detected.

- No gross beta-particle activity was detected.
- No tritium was detected.
- No radium analyses were necessary or performed.
- No uranium analyses were necessary or performed.

4.3 Bradley Landfill

Groundwater Samples (2 samples):

- Gross alpha-particle activity was detected in both samples, ranging from 6.5 pCi/L (4915A) to 10.3 pCi/L (4916H).
- No radium isotopes were detected in either sample (both samples were analyzed for radium-226 and radium-228 due to gross alpha results exceeding 5pCi/L).
- Gross beta-particle activity was detected in both samples at 5.9 pCi/L.
- No tritium was detected.
- No uranium analyses were necessary or performed.

Leachate Samples (3 samples):

- Gross alpha-particle activity was detected in all samples, ranging from 3.9 pCi/L (Sump B) to 4.60 pCi/L (Sump A).
- Gross beta-particle activity was detected in all samples, ranging from 187 pCi/L (Sump B) to 940 pCi/L (Sump E).
- Tritium was detected in all samples, ranging from 3,710 pCi/L (Sump E) to 10,500 pCi/L (Sump B).
- No radium analyses were necessary or performed.
- No uranium analyses were necessary or performed.

4.4 El Sobrante Landfill

Groundwater Samples (2 samples):

- Gross alpha-particle activity was detected in both samples, ranging from 3.61 pCi/L (MW-3-1) to 8.4 pCi/L (MW-2).
- No radium isotopes were detected in sample MW-2, which was analyzed for radium-226 and radium-228 due to gross alpha results exceeding 5pCi/L.
- Gross beta-particle activity was detected in one sample (MW-3-1) at 5.5 pCi/L.
- No tritium was detected.
- No uranium analyses were necessary or performed.

Leachate Samples (2 samples):

- Gross alpha-particle activity was detected in both samples, ranging from 2.63 pCi/L (LCRS-2) to 3.84 pCi/L (LCRS-3).
- Gross beta-particle activity was detected in one sample (LCRS-3) at 26 pCi/L.
- Tritium was detected in both samples, ranging from 3,980 (LCRS-2) to 22,900 pCi/L (LCRS-3).
- No radium analyses were necessary or performed.
- No uranium analyses were necessary or performed.

4.5 Kettleman Hills Facility

Groundwater Samples (6 samples):

- Gross alpha-particle activity was detected in all samples, ranging from 0.78 pCi/L (K70) to 1.77 pCi/L (K67).
- No gross beta-particle activity was detected.
- No tritium was detected.
- No radium analyses were necessary or performed
- No uranium analyses were necessary or performed.

Leachate Samples (2 samples):

- Gross alpha-particle activity was detected in both samples, ranging from 5.1 pCi/L (B191AP) to 15.5 pCi/L (B181AP).
- Radium-226 was detected in sample B181AP at 0.37 pCi/L; this sample was analyzed for radium isotopes due to gross alpha results exceeding 5pCi/L.
- Radium-228 was detected in sample B191AP at 1.27 pCi/L; this sample was analyzed for radium isotopes due to gross alpha results exceeding 5pCi/L.
- Uranium (total) was detected in sample B181AP at 1.75 pCi/L; this sample was analyzed for uranium isotopes due to gross alpha results exceeding 15pCi/L.
- No gross beta-particle activity was detected.
- No tritium was detected.

4.6 Redwood Landfill

Groundwater Samples (2 samples):

- Gross alpha-particle activity was detected in both samples, ranging from 1.43 pCi/L (MWH-8) to 9.92 pCi/L (MWH-19); note these are calculated values based on the radium-226 and isotopic uranium analyses performed on both groundwater samples in lieu of gross alpha by co-precipitation.
- Radium-226 was detected in both samples, ranging from 0.42 pCi/L (MWH-19) to 0.89 pCi/L (MWH-8).
- Radium-228 was detected in both samples, ranging from 1.16 pCi/L (MWH-8) to 2.99 pCi/L (MWH-19).
- Uranium (total) was detected in both samples, ranging from 0.54 pCi/L (MWH-8) to 9.5 pCi/L (MWH-19).
- No gross beta-particle activity was detected.
- No tritium was detected.

Leachate Samples (3 samples):

- Gross alpha-particle activity was detected in one sample (LD#C) at 0.41 pCi/L; note this is a calculated value based on radium-226 and isotopic uranium analyses that were performed on leachate samples in lieu of gross alpha by co-precipitation.
- Radium-228 was detected in all samples, ranging from 2.67 pCi/L (LD#B) to 3.34 pCi/L (LD#D).

- Gross beta-particle activity was detected in all samples, ranging from 222 pCi/L (LD#B) to 325 pCi/L (LD#D).
- Tritium was detected in all samples, ranging from 480 pCi/L (LD#C) to 7,140 pCi/L (LD#D).
- Uranium (total) was detected in one sample (LD#C) at 0.41 pCi/L.
- No radium-226 was detected.

4.7 Field Quality Control Samples

One field blank was prepared for each landfill included in this program. The field blanks were prepared in the field using the same HDPE bottles used for radiochemical sample collection and laboratory-grade water supplied by STL. The field blanks were tested for gross alpha/beta-particle activity. No gross alpha/beta- particle activity was detected in any of the field blanks.

4.8 Data Quality Issues

A review of laboratory quality control sample data accompanying the radiochemical analyses did not reveal any significant analytical problems; additional details are provided in the case narratives of the CARs, presented in Appendix B. However, an additional tritium analysis was performed on leachate sample LS from Altamont Landfill due to an anomalous tritium result. The initial sample, collected on 8/15/02 returned a tritium result of 84,300 pCi/L with a re-analysis result of 85,800 pCi/L. These results were considered suspect because all other Altamont Landfill leachate tritium results (four others in addition to LS) returned values of “not detected” at activity levels of 430 pCi/L or less. All of the Altamont Landfill leachate samples were collected in close proximity to one another and would be expected to return similar results. The LS sample location was resampled on 11/8/02 and retested for tritium, returning a value of “not detected” (i.e., <430 pCi/L). No explanation of the original highly anomalous tritium values was apparent after consultations with the analytical laboratory and field sampling team. All of the tritium results for LS are provided in Appendix B; however, only the final tritium result (<430 pCi/L) is presented on Table 2 and evaluated herein.

5.0 DISCUSSION

The limited data collected for this program preclude a rigorous evaluation of the sources and implications of detected radionuclides in groundwater and leachate samples from the landfills of interest. The factors summarized above in Section 1.3 and 2.0 limit the ability to draw firm conclusions. Simple comparisons of data from upgradient monitoring locations to downgradient monitoring locations are not sufficient to draw any meaningful conclusions on potential environmental impact from the landfills, given the known complexities associated with spatial and temporal variability of monitoring data. For example, in a radiochemical study of radon activity in groundwater, the USGS found that radon concentrations ranged up to 3 orders of magnitude in water from a single geologic unit, and differed significantly from well to well (USGS, 1998). Furthermore, in the absence of sufficient background data, comparative statistics that could account for temporal and spatial variability based on the procedures outlined in CCR Title 27 cannot be performed.

Comparison of the data to MCLs is also of minimal value. Landfill leachate typically contains chemicals in excess of water quality standards. For that reason, engineering controls are designed to contain landfill leachate, thus preventing it from being discharged directly into waters of the U.S. Also, as noted in Section 2.1, MCLs are at levels such that a significant proportion of drinking-water supply samples are found to exceed MCLs. Therefore, a more relevant evaluation may be the comparison of landfill-related groundwater samples to public water supply samples. Comparisons to public water supply samples provides some context regarding the levels of naturally-occurring radionuclides that may be expected, and this comparison may provide an indication of the presence or absence of unusual and/or potential anthropogenic sources of radionuclides in landfill-related groundwater samples. The comparison to public drinking-water samples is considered conservative because of the generally poor water quality at landfills (i.e., high TDS). As discussed in Section 2.0, elevated TDS can, in some cases, account for elevated radionuclide levels in groundwater.

The DHS maintains a database of samples collected from public water sources in California and analyzed for radionuclides during the period 1984-2002. Queries were performed on this database to extract relevant radiochemical data. Table 3 presents a statistical summary for each radiochemical parameter of interest, based on Los Angeles County public water supply samples. The following observations are presented with respect to the data in Table 3 as well as the alpha-particle activity threshold values used for this program:

- Comparison of the landfill groundwater samples collected for this study (Table 2) to the maximum values measured in the Los Angeles County public water supply samples (Table 3) indicates that none of the groundwater samples analyzed for this program exceeded the maximum values exhibited for Los Angeles County public water supply samples.

- 7 out of 20 groundwater samples exceeded the 5 pCi/L threshold for gross alpha-particle activity (4 using method SMWW 7110, 2 using method 900.0, and 1 calculated). Gross alpha-particle activity levels in the 7 samples were generally similar; three samples represent non-impacted groundwater or “background”

samples (MWH-19 at Redwood Landfill, 4915A at Bradley Landfill, and MW-4A at Altamont Landfill) and four were collected from impacted or previously impacted wells (4916H at Bradley Landfill, E-20B and E-05 at Altamont Landfill, and MW-2 at El Sobrante Landfill). This indicates no correlation between gross alpha-particle activity levels and groundwater impact (i.e., a natural alpha source is indicated).

- Of the 7 groundwater samples exceeding the 5 pCi/L threshold for gross alpha-particle activity, only the two samples from Redwood Landfill (MWH-8 and MWH-19) had detectable radium-226 and/or radium-228. These samples also had detectable uranium, suggesting a uranium source for the radium. In addition, a uranium-234/uranium-238 ratio of approximately 1.0 indicates that the uranium is naturally-occurring.

- None of the 20 groundwater samples exceeded the 15 pCi/L threshold for gross alpha-particle activity.

- 3 of 14 leachate samples exceeded the 5 pCi/L threshold for gross alpha-particle activity. One sample is from Altamont Landfill (GWIB) and two are from the Kettleman Hills Facility (B181AP and B191AP). Only sample (B181AP) exhibited detectable radium-226 and only sample (B191AP) exhibited detectable radium-228.

- 1 of 14 leachate samples exceeded the 15 pCi/L threshold for gross alpha-particle activity. This sample (B181AP at the Kettleman Hills Facility) exhibited a uranium-234/uranium-238 ratio of approximately 1.0, indicating that the uranium is naturally-occurring.

Though not anomalous with respect to uranium concentrations reported for public water supply samples in the DHS database, the highest uranium value (9.5 pCi/L, total uranium in groundwater well MWH-19) measured in this study is from Redwood Landfill, a “bayfront” landfill located near San Francisco Bay. This observation is consistent with the information noted previously that organic, marshland environments tend to concentrate natural uranium. The near absence of uranium in landfill leachate from this landfill, as well as the uranium-234/uranium-238 ratio of 1.0 clearly supports a natural uranium source for well MWH-19 groundwater. Similarly, in the other sample (leachate sample B181BP at Kettleman Hills) where both uranium-234 and uranium-238 were quantified (at a total concentration of 1.75 pCi/L), a uranium-234/uranium-238 ratio of approximately 1.0 ratio was measured, indicating a natural uranium source. By inference, the low levels of radium-226 and/or radium-228 in these and other samples at both Redwood Landfill and the Kettleman Hills Facility indicate that the radium isotopes are likely derived from the naturally-occurring uranium.

Gross beta-particle activity levels in some leachate samples appear elevated, although additional information on site-specific background conditions would be necessary to identify truly anomalous levels. It is believed that much of the beta-particle activity detected in the landfill leachate samples is attributable to natural beta radiation emitting from disposed organic material in the landfills, particularly food wastes, with natural potassium-40 as the primary source. A landfill leachate collection and recovery system (LCRS) is an effective containment system but, as such, will concentrate organic-rich

leachate containing high dissolved salts, including potassium salts. As explained in Section 2.2.3, radioactivity emitting from naturally-occurring potassium is so significant that relatively small amounts can produce substantial beta-particle activity. Leachate samples commonly contain hundreds of mg/L of natural dissolved potassium, potentially capable of producing hundreds of pCi/L of beta-particle activity, as described in Section 2.2.3. Figure 15 presents a scatter plot of dissolved potassium versus gross beta-particle activity for the samples collected for this program. As shown on Figure 15, a strong, positive statistical correlation (R^2=0.78) is exhibited, with high potassium concentrations coinciding with high gross beta-particle activity.

Tritium activity in some leachate samples are higher than what can be explained by contributions from natural precipitation, even in sites where waste was disposed during periods of atmospheric weapons testing. This suggests an anthropogenic tritium source (i.e., within the landfill waste stream). As described in Section 2.2.4, the presence of elevated tritium activity in leachate is well documented and has been attributed to general household and commercial waste containing tritium-bearing items that have fulfilled their useful life. The maximum tritium value measured in leachate (22,900 pCi/L at El Sobrante Landfill) is below the mean leachate tritium value reported in the UK study (Hicks, et. al., 2000), indicating that the tritium level is not anomalous with respect to leachate data contained in the literature. Elevated tritium activity measured in leachate samples for this study is, therefore, not regarded as unusual. The containment of water in an LCRS will serve to concentrate any tritium in leachate, from both natural and anthropogenic sources.

6.0 CONCLUSIONS

Landfill groundwater and leachate samples collected for this study do not appear to exhibit radioactivity levels of radiological significance, nor do they indicate the presence of the unauthorized disposal of regulated radioactive materials or waste in any of the six landfills examined. Furthermore, the landfill groundwater samples do not exhibit particularly unusual or anomalous radioactivity levels relative to California public water supply samples. Where uranium and radium isotopes were detected in groundwater and leachate samples, the concentrations were low and natural a uranium source is supported by the data.

Apparently elevated levels of gross beta activity observed in some leachate samples appear to be related to naturally-occurring potassium-40. As explained in this report, the relatively small fraction of radioactive potassium-40 that comprises natural potassium can produce significant levels of gross beta-particle activity in water. Furthermore, the beta-particle activities measured in the leachate samples are lower than the potassium-40-related beta-particle activities of many types of food. It is important to note that the samples exhibiting the apparently elevated beta activity are all LCRS-contained leachate samples; no elevated beta activity was observed in groundwater samples, all of which exhibit relatively low concentrations of dissolved natural potassium.

Apparently elevated levels of tritium activity were observed in some leachate samples. No tritium was detected in any groundwater samples, including from known landfill-impacted wells. The highest tritium value detected in leachate (22,800 pCi/L) slightly exceeds the MCL of 20,000 pCi/L. However, this level of tritium activity is not anomalous with respect to leachate data contained in the literature and, therefore, is not regarded as unusual. Studies on elevated tritium activity in landfill leachate indicate that the source is likely household and/or commercial disposal of tritium-containing items which have fulfilled their useful life, and are contained within general waste streams being landfilled (Robinson and Gronow, 1996).

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