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Rapid Radiochemical Method for Curium-244 in Air Particulate Filters, Swipes and Soil for Environmental Remediation Following Radiological Incidents

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RAPID RADIOCHEMICAL METHOD FOR CM-244 IN AIR PARTICULATE FILTERS, SWIPES AND SOILS FOR ENVIRONMENTAL REMEDIATION FOLLOWING RADIOLOGICAL INCIDENTS

1. Scope and Application
   - 1.1. This method provides for the rapid determination of curium-244 (^{244}Cm) in air particulate filters, swipes and soil samples.
   - 1.2. The method uses americium-243 (^{243}Am) tracer as the basis for quantification of ^{244}Cm and as a yield monitor.
   - 1.3. A sample test source (STS) is prepared by microprecipitation. The test source is counted by alpha spectrometry for ^{244}Cm.
     - 1.3.1. Curium-243 (^{243}Cm) emits alpha particles that are isoenergetic with ^{244}Cm. Alpha spectrometry measurements that show activity in the region of interest for ^{244}Cm should be reported as $^{244/243}$Cm.
   - 1.4. Measurement Quality Objectives (MQOs):
     - 1.4.1. Air Particulate Filters:
       - 1.4.1.1. This method is capable of achieving a required method uncertainty for ^{244}Cm of 1.4 pCi/filter at an analytical action level (AAL) of 10.5 pCi/filter. To attain this MQO, a sample aliquant of one filter and a count time of at least 4 hours are recommended. Sample count times may vary based on differences in instrument parameters such as detection efficiency and background. The concentration in air (i.e., pCi/m^3) to which this MQO corresponds will vary according to the volume of air sampled on the filter.
       - 1.4.1.2. The method is capable of achieving a required minimum detectable concentration (MDC) for ^{244}Cm of 0.25 pCi/filter. To attain this MQO, a sample aliquant of one filter and a count time of at least 4 hours are recommended. Sample count times may vary based on differences in instrument parameters such as detection efficiency and background. The concentration in air (i.e., pCi/m^3) to which this MQO corresponds will vary according to the volume of air sampled on the filter.
     - 1.4.2. Swipes (or Organic-Polymer-Based Air Particulate Filters):
       - 1.4.2.1. This method is capable of achieving a required method uncertainty for ^{244}Cm of 0.051 pCi/swipe at an AAL of 0.39 pCi/swipe. To attain this MQO, a sample aliquant of one swipe and a count time of at least 4 hours are recommended. Sample count times may vary based on differences in instrument parameters such as detection efficiency and background. The surface concentration (i.e., pCi/cm^2) to which this MQO corresponds will vary according to the area sampled on the swipe.

1.4.2.2. This method is capable of achieving a required MDC for ^{244}Cm of 0.065 pCi/swipe. To attain this MQO, a sample aliquant of one swipe and a count time of at least 4 hours are recommended. Sample count times may vary based on differences in instrument parameters, such as detection efficiency and background. The surface concentration (i.e., pCi/cm^2) to which this MQO corresponds will vary according to the area sampled on the swipe.

1.4.3. Soil:

1.4.3.1. This method is capable of achieving a required method uncertainty for ^{244}Cm of 0.66 pCi/g at an AAL of 5.09 pCi/g. To attain this MQO, a sample weight of 1 gram and a count time of at least 4 hours are recommended. Sample count times may vary based on differences in instrument parameters such as detection efficiency and background.

1.4.3.2. This method is capable of achieving a required MDC for ^{244}Cm of 0.66 pCi/g. To attain this MQO, a sample weight of 1 gram and a count time of at least 4 hours are recommended. Sample count times may vary based on differences in instrument parameters such as detection efficiency and background.

1.5. This ^{244}Cm method was single-laboratory evaluated following the guidance presented for Level E Method Validation: Adapted or Newly Developed Methods, Including Rapid Methods" in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (EPA 2009, Reference 16.1) and Chapter 6 of *Multi-Agency Radiological Laboratory Analytical Protocols Manual* (MARLAP, EPA 2004, Reference 16.2).

1.5.1. Since californium (Cf) and americium (Am) track closely with curium (Cm) through the chemical separation, it may be possible to determine isotopes of Cf, as well as isotopes of Am (e.g., ^{241}Am) that may be present in the sample test source.

1.5.2. The sample turnaround time and throughput may vary based on additional project MQOs, the time for analysis of the sample test source, and initial sample weight/volume.

1.5.3. The method, as implemented at the laboratory, must be validated prior to use following the protocols provided in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (EPA 2009, reference 16.1).

2. Summary of Method

2.1. This method is based on the use of extraction chromatography resins (TEVA® + DGA resins) to isolate and purify Cm by removing interfering radionuclides and matrix components in order to prepare the Cm fraction for counting by alpha spectrometry. The method utilizes vacuum-assisted flow to improve the speed of the separations. An ^{243}Am tracer is equilibrated with the sample as a yield monitor.

2.1.1. Glass-fiber or cellulose-based air particulate filter samples are wet-ashed with repeated additions of nitric acid (HNO_3) and hydrofluoric acid (HF),

and hydrogen peroxide. The residues are treated with nitric-boric acid and dissolved in a load solution containing ~3 molar (M) HNO_3 - 1M $Al(NO_3)_3$ before continuing with chemical separations.

2.1.2. Cotton-twill swipe and organic-polymer-based air particulate filter samples are dry-ashed in a beaker for 30-60 minutes using a ramped program to minimize the risk of flash-ignition. The residue is transferred to a Teflon beaker with HNO_3 and hydrogen peroxide (H_2O_2), digested with HF, and taken to dryness. The residues are then wet-ashed with HNO_3 and hydrogen peroxide and taken to dryness before being treated with nitric-boric acid and dissolved in a load solution containing ~3M HNO_3 - 1M $Al(NO_3)_3$ before continuing with chemical separations.

2.1.3. Soils are finely ground before being fused with sodium hydroxide (NaOH) in zirconium crucibles. The fusion cake is dissolved in water and Cm preconcentrated from the alkaline matrix using an iron/titanium hydroxide precipitation (enhanced with calcium phosphate precipitation) followed by a lanthanum fluoride matrix removal step. The fluoride precipitate is dissolved with nitric-boric acid and diluted in HNO_3 and aluminum nitrate [$Al(NO_3)_3$] to yield a load solution containing ~3M HNO_3-1M $Al(NO_3)_3$ before continuing with chemical separations.

2.1.4. The size of the sample aliquant may need to be decreased for samples containing high alpha activity. This may require delay of addition of the tracer after the sample has been dissolved and split, and would require that the appropriate dilution factor be applied.

2.2. Extraction chromatography resins (TEVA® + DGA resins) are then used to isolate and purify Cm by removing interfering radionuclides and other matrix components. The method utilizes vacuum-assisted flow to improve the speed of the separations. Following chemical separation of Cm and Am, the sample test source (STS) is prepared by microprecipitation with cerium (III) fluoride (CeF_3).

2.3. The alpha emissions from the source are measured using an alpha spectrometer and used to calculate the activity of ^{244}Cm in the sample.

3. Definitions, Abbreviations, and Acronyms

3.1. Analytical Protocol Specifications (APS). The output of a directed planning process that contains the project's analytical data needs and requirements in an organized, concise form.

3.2. Analytical Action Level (AAL). The term "analytical action level" is used to denote the value of a quantity that will cause the decision-maker to choose one of the alternative actions.

3.3. Discrete Radioactive Particles (DRPs or "hot particles"). Particulate matter in a sample of any matrix where a high concentration of radioactive material is contained in a tiny particle (generally micron [µm] range).

3.4. *Multi-Agency Radiological Analytical Laboratory Protocols Manual* (MARLAP) provides guidance for the planning, implementation, and assessment phases of those projects that require the laboratory analysis of radionuclides (EPA 2004, Reference 16.2).

3.5. Measurement Quality Objective (MQO). MQOs are the analytical data requirements of the data quality objectives and are project- or program-specific. They can be quantitative or qualitative. MQOs serve as measurement performance criteria or objectives of the analytical process.

3.6. Required Method Uncertainty (u_{MR}). The required method uncertainty is a target value for the individual measurement uncertainties, and is an estimate of uncertainty (of measurement) before the sample is actually measured. The required method uncertainty is applicable below an AAL.

3.7. Required Relative Method Uncertainty (φ_{MR}). The relative required method uncertainty is the u_{MR} divided by the AAL and is typically expressed as a percentage. It is applicable above the AAL.

3.8. Sample Test Source (STS). This is the final form of the sample that is used for nuclear counting. This form is usually specific for the nuclear counting technique used in the method such as a solid deposited on a filter for alpha spectrometry analysis.

4. Interferences

4.1. Radiological

4.1.1. The alpha emissions from ^{243}Cm fall in the same region as ^{244}Cm and cannot be differentiated from those of ^{244}Cm using alpha spectrometric determinations.

4.1.1.1. Although the ^{244}Cm and ^{243}Cm alpha emissions overlap, monitoring the region of the spectrum between 5.8 and 6.0 MeV for less intense emissions of ^{243}Cm may qualitatively indicate the presence of ^{243}Cm in a sample.

4.1.1.2. Alpha spectrometry measurements that show activity in the region of interest for ^{244}Cm should be reported as $^{244/243}$Cm.

4.1.2. Americium (Am) and Cf are chemical analogs of Cm in the separations scheme used for this analysis. Several isotopes of Cf emit alpha particles within the region of interest for ^{244}Cm. These include ^{249}Cf and ^{251}Cf. If high levels of Cf could be present in samples, alpha spectrometry results should be monitored for other isotopes of Cf.

4.1.3. Americium 243 may be present in certain sources that contain ^{244}Cm. In cases where ^{243}Am is observed or suspected to be present in samples, ^{241}Am may be used in place of ^{243}Am as the yield tracer. Although there is no reason to expect different performance, the approach should be validated by the laboratory prior to implementing.

4.1.4. Radionuclides of other elements (or their short-lived progeny) that emit alpha particles that are isoenergetic with ^{244}Cm (e.g., thorium-227 (^{227}Th) or actinium-225 (^{225}Ac) at 5.81 mega electron volts [MeV]) must be chemically separated to prevent positive interference with the measurement. This method separates these radionuclides effectively. For example, a thorium (Th) removal rinse is performed on DGA® resin in the event that Th passes through TEVA® resin onto DGA resin.

4.1.5. Vacuum box lid and holes must be cleaned frequently to minimize the risk of cross-contamination. This is especially important when processing samples containing elevated levels of radioactivity.

4.1.6. A dilute HNO_3 rinse is performed on DGA® resin for soil samples to remove residual calcium (Ca) and lanthanum (La) ions which could end up in the final alpha test source as fluoride solids. While the method takes this into account, the volume may be increased slightly to achieve better removal of Ca and La ions and possibly improve alpha peak resolution, but this will have to be validated by the laboratory.

4.1.7. Zirconium crucibles used in the furnace ashing and fusion process may be reused to minimize the risk of cross-contamination. This is especially important when processing samples containing elevated levels of radioactivity.

4.1.7.1. Before reuse, the crucibles should be cleaned very well using soap and water, followed by warm HNO_3 (multiple rinses) and then water.

4.1.7.2. It is recommended that crucibles be identified and that a record be made to document which crucible is used for each sample.

4.1.7.3. Blank measurements should be carefully monitored to ensure effective cleaning and control against cross-contamination.

4.1.7.4. Crucibles used for low and high activity samples should be segregated to minimize the risk of cross-contamination while maximizing the efficient use of crucibles.

4.2. Non-Radiological:

4.2.1. Anions that can complex Cm and Am, including fluoride and phosphate, may lead to depressed yields. Boric acid added to the load solution will complex residual fluorides, while aluminum (Al) in the load solution will complex residual phosphate ions that may be present.

4.2.2. High levels of Ca present in soil samples may have an adverse impact on Cm and Am retention on DGA resin. This method is designed to minimize this interference, and enhance Cm and Am affinity by increasing the nitrate concentration in the load and initial rinse solutions. A dilute HNO_3 rinse is performed on DGA resin to minimize the risk that calcium may carry through chemical separations and be deposited on the sample test source as calcium fluoride. For samples containing elevated concentrations of calcium, it may be advisable to increase the volume of this rinse step

slightly to improve alpha peak resolution. This modification must be validated by the laboratory prior to use with samples.

5. Safety
   - 5.1. General
     - 5.1.1. Refer to your safety manual for concerns of contamination control, personal exposure monitoring, and radiation safety manual for radiation dose monitoring.
     - 5.1.2. Refer to your laboratory's chemical hygiene plan (or equivalent) for general safety rules regarding chemicals in the workplace.
   - 5.2. Radiological
     - 5.2.1. Hot particles (DRPs)
       - 5.2.1.1. Hot particles, also termed "discrete radioactive particles" (DRPs), are small particles, generally in the micron range. Typically, DRPs are not evenly distributed in the media and their radiation emissions are anisotropic (i.e., not uniform in all directions).
       - 5.2.1.2. Samples containing measureable activity of ^{244}Cm may have DRPs. If suspended solids are removed by filtration, they should be checked for potential radioactivity.
       - 5.2.1.3. Cm present in DRPs may not be chemically available, and will not be determined, unless it is dissolved prior to chemical separation.
     - 5.2.2. For samples with detectable activity concentrations of these radionuclides, labware should be used only once due to potential for cross contamination.
   - 5.3. Procedure-Specific Non-Radiological Hazards: Particular attention should be paid to the use of hydrofluoric acid (HF). HF is an extremely dangerous chemical used in the preparation of some of the reagents and in the microprecipitation procedure. Appropriate personal protective equipment (PPE) must be used in strict accordance with the laboratory safety program specification.

6. Equipment and Supplies
   - 6.1. Alpha spectrometer calibrated for use over a range that includes 4.5 through 7.0 MeV.
   - 6.2. Analytical balance with minimum 10^{-2} g readability.
   - 6.3. Centrifuge tubes, 225-mL, 50-mL capacity, or equivalent.
   - 6.4. Centrifuge, to accommodate centrifuge tubes.
   - 6.5. Crucibles, 250-mL, zirconium, with lids.
   - 6.6. Heat lamp.
   - 6.7. Hot Plate.
   - 6.8. Laboratory ware of plastic, glass, or Teflon; 150-, 250-, 500- and 1,000-mL capacities, as needed.
   - 6.9. Oven capable of temperatures ranging from 100-600 °C.
   - 6.10. Pipettor, electronic, and appropriate plastic tips, 1-10-mL as needed.
   - 6.11. Pipettors, manual, and appropriate plastic tips, 100-microliter (µL), 200-µL, 500-µL and 1-mL, or equivalent, as needed.

6.12. Sample test source mounts:

6.12.1. Polypropylene filter, 0.1-µm pore size, 25-mm diameter, or equivalent.

6.12.2. Stainless steel planchets, adhesive backed disks (e.g., Environmental Express, Inc. P/N R2200) or equivalent (calibrated for 25-mm filter geometry).

6.13. Tweezers.

6.14. Vacuum box system

6.14.1. Vacuum box/rack (e.g., Eichrom Technologies, Inc., Lisle, IL part number AC-24-BOX), or equivalent.

6.14.2. Cartridge reservoirs, 10- or 20-mL syringe style with locking device, or columns (e.g., empty Luer-lock tip, Image Molding, Denver, CO, part number CC-10 M) plus 12mL reservoirs (e.g., Image Molding, Denver, CO, part number CC-06-M), or equivalent.

6.14.3. Vacuum box tips, white inner, Eichrom Technologies, Inc., Lisle, IL part number AC-1000-TUBE-PE, or PFA 5/32" × ¼" heavy wall tubing connectors, natural, Cole Parmer Instrument Company, LLC, Vernon Hills, IL part number 00070EE, cut to 1 inch, or equivalent.

6.14.4. Vacuum box tips, yellow outer, Eichrom Technologies, Inc., Lisle, IL part number AC-1000-OT, or equivalent.

6.14.5. Laboratory vacuum source.

6.15. Vortex mixer.

7. Reagents and Standards

NOTE: All reagents are American Chemical Society (ACS) reagent grade or equivalent unless otherwise specified.

NOTE: Unless otherwise indicated, all references to water should be understood to mean Type I Reagent water (ASTM D1193, Reference 16.4). All solutions used in microprecipitation should be prepared with water filtered through a 0.45-µm (or better) filter.

NOTE: Low levels of uranium are typically present in $Al(NO_3)_3$.

7.1. Aluminum nitrate solution: 2M: Add 750 g of aluminum nitrate ($Al(NO_3)_3 \cdot 9\ H_2O$) to ~500 mL of water and dilute to 1 liter with water.

7.2. Americium-243 tracer solution: 10-40 dpm of ^{243}Am per aliquant.

7.3. Ammonium hydrogen phosphate, 3.2M: Dissolve 106 g of $(NH_4)_2HPO_4$ in 200 mL of water, heat gently to dissolve and dilute to 250 mL with water.

7.4. Ammonium hydroxide, 15M: Concentrated NH_4OH.

7.5. Ascorbic acid, 1.5M: Dissolve 66 g $C_6H_8O_6$ in 200 mL of water, warming gently to dissolve, and dilute to 250 mL with water. Shelf life is 30 days or less.

7.6. Calcium nitrate, 1.25M: Dissolve 73.8 g of $Ca(NO_3)_2 \cdot 4\ H_2O$ in 100 mL of water and dilute to 250 mL with water.

7.7. Cerium carrier, 0.5 mg Ce/mL: dissolve 0.16 g $Ce(NO_3)_3 \cdot 6\ H_2O$ in 50 mL water and dilute to 100 mL with water.

7.8. Curium-244 standard solution: 10-40 dpm of ^{244}Cm per aliquant.

7.9. DGA resin, normal, 2-mL cartridge, 50- to 100-µm mesh size, Eichrom Technologies, Inc., Lisle, IL part number DN-R50-S, or equivalent.

7.10. Ethanol, 95%: Reagent C_2H_5OH, or mix 95 mL 100% ethanol and 5 mL water.

7.11. Hydrochloric acid, 12M: Concentrated HCl.

7.11.1. Hydrochloric acid, 0.01M: Add 0.8 mL of concentrated HCl to 500 mL of water and dilute with water to 1 L.

7.11.2. Hydrochloric acid, 0.25M: Add 21 mL of concentrated HCl to 500 mL of water and dilute with water to 1 L.

7.11.3. Hydrochloric acid, 1.5M: Add 125 mL of concentrated HCl to 500 mL of water and dilute with water to 1 L.

7.11.4. Hydrochloric acid, 3M: Add 250 mL of concentrated HCl to 500 mL of water and dilute with water to 1 L.

7.11.5. Hydrochloric acid, 4M: Add 333 mL of concentrated HCl to 500 mL of water and dilute with water to 1 L.

7.12. Hydrofluoric acid, 28M: Concentrated HF

7.13. Hydrogen peroxide, 30 weight percent (wt%) (H_2O_2).

7.14. Iron carrier, 50 mg/mL: Dissolve 181 g of ferric nitrate ($Fe(NO_3)_3 \cdot 9\ H_2O$) in 300 mL water and dilute to 500 mL with water.

7.15. Lanthanum carrier, 1.0 mg La^{3+}/mL: Dissolve 1.56 g lanthanum (III) nitrate hexahydrate [$La(NO_3)_3 \cdot 6\ H_2O$] in 300 mL water and dilute to 500 mL with water.

7.16. Nitric acid, 16M: Concentrated HNO_3.

7.16.1. Nitric acid, 0.075M: Add 4.7 mL of concentrated HNO_3 to 700 mL of water and dilute to 1 L with water.

7.16.2. Nitric acid, 0.1M: Add 6.3 mL of concentrated HNO_3 to 700 mL of water and dilute to 1 L with water.

7.16.3. Nitric acid, 1M: Add 63 mL of concentrated HNO_3 to 700 mL of water and dilute to 1 L with water.

7.16.4. Nitric acid, 3M: Add 190 mL of concentrated HNO_3 to 700 mL of water and dilute to 1 L with water.

7.16.5. Nitric acid, 6M: Add 380 mL of concentrated HNO_3 to 500 mL of water and dilute to 1 L with water.

7.16.6. Nitric acid, 7M: Add 443 mL of concentrated HNO_3 to 500 mL of water and dilute to 1 L with water.

7.17. Nitric acid – boric acid, 3M – 0.25M: Add 15.5 g of H_3BO_3 and 190 mL of concentrated HNO_3 to 500 mL of water, heat to dissolve, and dilute to 1 liter with water.

7.18. Nitric acid – HF, 3M – 0.25M: Add 8.9 mL of concentrated HF and 190 mL of concentrated HNO_3 to 700 mL of water. Dilute to 1 liter with water and mix well

7.19. Phenolphthalein indicator solution, 0.5 wt% ($C_{20}H_{14}O_4$): Dissolve 0.5 g in 100 mL ethanol (95%).

7.20. Sodium nitrite solution, 3.5M: Dissolve 6.1 g of $NaNO_2$ in 25 mL of water. Prepare fresh daily.

7.21. Sodium hydroxide pellets.

7.22. Sulfamic acid solution, 1.5M: Dissolve 72.8 g of H_3NSO_3 in 400 mL of water and dilute to 500 mL with water.

7.23. TEVA® resin, 2-mL cartridge, 50- to 100-µm mesh size, Eichrom Technologies, Inc., Lisle, IL part number TE-R50-S and TE-R200-S, or equivalent.

7.24. Titanium (III) chloride ($TiCl_3$) solution, 10 wt% in 20-30 wt% HCl.

Note: If 10 wt% $TiCl_3$ is not available, other concentrations of $TiCl_3$ (e.g., 12-20%) may be used if the amount is adjusted based on the assay of the solution to deliver the same or slightly more titanium. For example, if 17 wt% $TiCl_3$ is used, the volume may be decreased to 3 mL.

8. Sample Collection, Preservation, and Storage

8.1. No sample preservation is needed for air particulate filters, swipes, or soil samples.

9. Quality Control

9.1. Batch QC results shall be evaluated and meet applicable Analytical Protocol Specifications (APS) prior to release of unqualified data. In the absence of project-defined APS or a project specific quality assurance project plan (QAPP), the QC sample acceptance criteria defined in the laboratory quality manual and procedures shall be used to determine acceptable performance for this method.

9.1.1. A Laboratory Control Sample (LCS) shall be run with each batch of samples. The concentration of the LCS should be at or near the action level or level of interest for the project.

9.1.2. One method blank shall be run with each batch of samples. The laboratory blank should consist of demineralized water.

9.1.3. One laboratory duplicate shall be run with each batch of samples. The laboratory duplicate is prepared by removing an aliquant from the original sample container.

9.1.4. A matrix spike sample is not required as a chemical yield tracer is used in each sample.

9.2. The source preparation method should produce a sample test source in which the full width at half maximum (FWHM) for the tracer peak is less than 100 keV.[1]

9.2.1. Each spectrum should be reviewed for evidence of peaks that overlap or evidence of interference with the tracer or analyte peaks.

9.2.2. The sample test source may require reprocessing to remove interfering mass if the FWHM limit cannot be achieved and there are any indications that degraded resolution may have impacted the quantification of ^{244}Cm.

[1] This helps minimize interference from alpha-emitting isotopes with potentially overlapping energies.

10. Calibration and Standardization

10.1. Set up the alpha spectrometry system according to the manufacturer's recommendations consistent with ASTM Standard Practice D7282, Section 9.3, "Alpha Spectrometry Initial Instrument Set-up" (ASTM D7282, Reference 16.3). The energy range of the spectrometry system should at minimum include the region that encompasses 4.5 and 7.0 MeV.

10.2. Establish initial instrument QCs as described in ASTM Standard Practice D7282, Sections 10-15, "Initial Instrument Quality Control Testing" (ASTM D7282, Reference 16.3)

10.3. Calibrate each detector used to count samples according to ASTM Standard Practice D7282, Section 18, "Alpha Spectrometry Instrument Calibrations" (ASTM D7282, Reference 16.3).

10.4. Perform Continuing Instrument Quality Control Testing according to ASTM Standard Practice D7282, Sections 20, 21, and 24, "Continuing Instrument Quality Control Testing" and "Quality Control for Alpha Spectrometry Systems" (ASTM D7282, Reference 16.3).

11. Procedure

11.1. Sample Preparation for Furnace Ashing and Acid Digestion of Swipes and Organic-Polymer-based Air Particulate Filters

Note: The sample and associated QC samples may be split after digestion to provide a back-up fraction.

11.1.1. Aliquant the entire swipe into a 150-mL glass beaker.

11.1.2. Set up an empty 150-mL glass beaker for use as a reagent blank

11.1.3. Set up an LCS by adding a known amount of ^{244}Cm to a 150 mL-glass beaker.

11.1.4. Add 10-40 dpm ^{243}Am tracer to the blank, LCS, and sample beakers following laboratory protocol.

11.1.5. Heat beaker with swipe on hot plate to dryness.

11.1.6. Place beaker in furnace at 200 ºC and ramp to 550 ºC. Hold at 550 ºC for 30 to 60 minutes and allow to cool.

11.1.7. Digest furnace-ashed swipe as follows:

11.1.7.1. Add 5 mL concentrated HNO_3 to the glass beaker and 1 ml of 30 wt% H_2O_2, warm on a hot plate with medium heat to dissolve residue and transfer to 250-mL Teflon beaker.

11.1.7.2. Add 5 mL concentrated HNO_3 to the glass beaker and 1 ml of 30 wt% H_2O_2, warm on hotplate and transfer the rinse to the Teflon beaker.

11.1.7.3. If necessary to remove any sample residue, add 3 mL concentrated HNO_3 and 1 ml 30 wt% H_2O_2 to the glass beaker, warm on hot plate with medium heat and add rinse to the Teflon beaker.

11.1.7.4. Add 2 ml concentrated HF to each beaker. Evaporate to dryness.

11.1.7.5. Add 3 mL concentrated HNO_3 and 2 ml 30 wt% H_2O_2 and evaporate to dryness.

11.1.7.6. Add 3 mL concentrated HNO_3, 2 ml 30 wt% H_2O_2 and 3 mL 3M HNO_3 - 0.25M boric acid and evaporate to dryness.

11.1.8. Dissolve the sample residue by adding 7 mL 6M HNO_3, to each beaker, warming on a hot plate.

11.1.9. Add 7 ml 2M $Al(NO_3)_3$. Swirl to mix well.

11.1.10. Continue with Step 11.4, Rapid Cm Separation using TEVA® and DGA resins.

11.2. Sample Preparation for Air Filter Samples

Note: This method is effective for cellulose-based or glass fiber air filters. The sample and associated QC samples may be split after digestion to provide a back-up fraction.

11.2.1. Aliquant the entire 2" – 4" air filter into a 250-mL Teflon beaker.

11.2.2. Set up an empty 250-mL Teflon beaker for use as a reagent blank sample.

11.2.3. Set up a laboratory control sample by adding a known amount of ^{244}Cm to a 250-mL Teflon beaker.

11.2.4. Add 10-40 dpm ^{243}Am tracer to all samples following laboratory protocol.

11.2.5. Digest air filters as follows:

11.2.5.1. Add 5 mL concentrated HNO_3, 5 ml concentrated HF, and 2 ml of 30 wt% H_2O_2. Evaporate to dryness on a hot plate with medium heat.

11.2.5.2. Add 3 mL concentrated HNO_3 and 2 ml of 30 wt% H_2O_2 and take to dryness.

11.2.5.3. Repeat Step 11.2.5.2 twice.

Note: Step 11.2.5.2 may be repeated as needed to effect complete digestion of the sample matrix.

11.2.5.4. Add 3 mL concentrated HNO_3, 2 mL 30 wt% H_2O_2 and 3 mL 3M HNO_3 - 0.25M boric acid and evaporate to dryness.

11.2.6. Dissolve the sample residue by adding 7 mL 6M HNO_3, to each beaker, warming on a hot plate.

11.2.7. Add 7 ml 2M $Al(NO_3)_3$. Swirl to mix well.

11.2.8. Continue with Step 11.4, Rapid Cm Separation using TEVA® and DGA resins.

11.3. Fusion of soil samples

11.3.1. In accordance with the DQOs and sample processing requirements stated in the project plan documents, remove extraneous materials from the soil sample using a clean forceps or tweezers.

11.3.2. Set up an empty crucible for use as a reagent blank sample.

11.3.3. Set up a laboratory control sample by adding a known amount of ^{244}Cm to an empty crucible.

11.3.4. Weigh out a representative, finely ground 1-g aliquant of dry sample into a crucible.
11.3.5. Add 10-40 dpm ^{243}Am tracer to all samples following laboratory protocol.
11.3.6. Place crucibles on a hot plate and take to dryness at medium heat.

NOTE: Heat on medium heat to dry quickly but not so high as to cause splattering.

11.3.7. Remove crucibles from hot plate and allow to cool.
11.3.8. Add 15 g NaOH of sodium hydroxide to each crucible.
11.3.9. Place the crucibles with lids in the 600 °C furnace using tongs.
11.3.10. Fuse samples in the crucibles for ~15 minutes.

NOTE: Longer times may be needed for larger particles.

11.3.11. Remove hot crucibles from furnace very carefully using tongs, and transfer to hood.
11.3.12. Add ~25-50 mL of water to each crucible ~8-10 minutes (or longer) after removing crucibles from furnace, and heat on hotplate to loosen/dissolve solids.
11.3.13. Transfer each fused sample to a 225-mL centrifuge tube, rinse crucible well with water, and transfer rinses to each tube.
11.3.14. If necessary to obtain complete dissolution, add more water and warm as needed on a hotplate. Transfer the rinse to the 225-mL tube. If needed, repeat this step until all solids have been dissolved and transferred to the centrifuge tube.
11.3.15. Add 10 mL 3M HNO_3 to each crucible and heat crucibles on a hot plate until hot. Transfer the 3M HNO_3 rinse to the 225-mL tube, followed by additional rinses of water.

NOTE: The iron (Fe) and La carriers may be added to the 225-mL centrifuge tube before adding the dissolved sample.

11.3.16. Pipet 2.5 mL of iron carrier (50 mg/mL) and 4 mL 1.0 mg La/mL into the 225-mL centrifuge tube.
11.3.17. Dilute each sample to approximately 180 mL with water.
11.3.18. Cool the 225-mL centrifuge tubes in an ice water bath to approximately room temperature, as needed.
11.3.19. Pipet 2.5 mL 1.25M $Ca(NO_3)_2$ and 5 mL 3.2M $(NH_4)_2HPO_4$ into each tube. Cap tubes and mix well.
11.3.20. Add 5 mL of 10 wt% $TiCl_3$ to each tube. Cap and mix immediately.

Note: If 10 wt% $TiCl_3$ is not available, other concentrations of $TiCl_3$ (e.g., 12-20%) may be used if the amount is adjusted based on the assay of the solution to deliver the same or slightly more titanium. For example, if 17 wt% $TiCl_3$ is used, the volume may be decreased to 3 mL.

11.3.21. Cool the 225-mL centrifuge tubes in an ice water bath for ~5 minutes.
11.3.22. Centrifuge tubes for 6 minutes at 3500 revolutions per minute (rpm).
11.3.23. Pour off the supernate and discard to waste.
11.3.24. Add 1.5M HCl to a total volume of ~80 mL to redissolve each sample.
11.3.25. Cap and shake each tube to dissolve solids as well as possible.

NOTE: There typically will be undissolved solids, which is acceptable.

11.3.26. Dilute each tube to ~170 mL with 0.01M HCl. Cap and mix well.
11.3.27. Pipet 1 mL of 1.0 mg La/mL and 1 mL 1.25M $Ca(NO_3)_2$ to each tube and swirl to mix.
11.3.28. Add 3 mL of 10 wt% $TiCl_3$ into each tube. Cap and mix immediately.

Note: If 10 wt% $TiCl_3$ is not available, other concentrations of $TiCl_3$ (e.g., 12-20%) may be used if the amount is adjusted based on the assay of the solution to deliver the same or slightly more titanium. For example, if 17 wt% $TiCl_3$ is used, the volume may be decreased to 2 mL.

Note: It may be possible to achieve effective precipitation in Step 11.3.29 using as little as 10 mL HF but any such substitution would need to be validated as described in Step 1.5.

11.3.29. Add 25 mL of concentrated HF into each tube. Cap and mix well.
11.3.30. Place tubes in an ice water bath for ~10 minutes to get the tubes very cold.
11.3.31. Remove the tubes from the ice water bath, wait 5 minutes, then centrifuge for ~6 minutes at 3000 rpm or more or as needed.
11.3.32. Pour off supernate, and discard to waste.
11.3.33. Pipet 5 mL of 3M HNO_3 - 0.25M boric acid into each tube.
11.3.34. Cap, mix and transfer contents of the tube into a labeled 50-mL centrifuge tube.
11.3.35. Pipet 6 mL of 7M HNO_3 and 7 mL of 2M $Al(NO_3)_3$ into each tube. Cap and mix (shake or use a vortex stirrer), and transfer rinse to 50-mL centrifuge tube.
11.3.36. Pipet 3 ml of 3M HNO_3 directly into the 50-mL centrifuge tube.
11.3.37. Warm each 50-mL centrifuge tube in a hot water bath for a few minutes, swirling to dissolve.
11.3.38. Remove each 50-mL centrifuge tube from the water bath and allow to cool to room temperature.
11.3.39. Centrifuge the tubes at 3000 rpm for 5 minutes to remove any traces of solids (may not be visible prior to centrifuging).
11.3.40. Transfer solutions to labeled beakers or tubes for further processing. Discard any solids.
11.3.41. Continue with Step 11.4, Rapid Cm Separation using TEVA® and DGA resins.

11.4. Rapid Cm Separation using TEVA® and DGA resins

Note: A smaller volume of the total load solution may be taken and analyzed as needed for very high activity samples, with appropriate dilution factor calculations applied.

11.4.1. Add 0.2 mL of 1.5M sulfamic acid to each sample. Swirl to mix.

NOTE: A smaller volume may be taken instead of the total load solutions, this smaller volume should be diluted to ~15 mL with 3M HNO_3 before proceeding with the valence adjustment.

NOTE: If ^{237}Np is potentially present in the sample, also add 0.5 mL of 4 mg/mL iron carrier to enhance neptunium (Np) reduction to Np^{4+}. The addition of ascorbic acid in the next step will convert Fe^{3+} to Fe^{2+}, which ensures removal of neptunium on TEVA® resin.

11.4.2. Add 1.25 mL of 1.5M ascorbic acid to each sample. Swirl to mix. Wait 3 minutes.

NOTE: Plutonium (Pu), if present, will be adjusted to Pu^{4+} to ensure retention and removal on TEVA® resin. A small amount of brown fumes results from nitrite reaction with sulfamic acid. The solution should clear with swirling. If the solution does not clear (is still dark) an additional small volume of sodium nitrite may be added to clear the solution.

11.4.3. Add 1 mL of 3.5M $NaNO_2$ to each sample. Swirl to mix.

NOTE: The load solution nitrate concentration is increased after valence adjustment to provide greater retention of Cm and Am and more effective removal of calcium ions on DGA resin.

11.4.4. Add 1.5 mL concentrated HNO_3 to each sample and swirl to mix.

NOTE: The steps in this section were optimized for a commercially available filtration system. Other vacuum systems may be substituted here. The cartridges may be set up and conditioned with HNO_3 so that they are ready for column loading just prior to completion of the valence adjustment steps. More than one vacuum box may be used to increase throughput.

11.4.5. Set up TEVA® and DGA cartridges on the vacuum box system.

11.4.5.1. Place the inner centrifuge tube rack (supplied with vacuum box) into the vacuum box with the centrifuge tubes in the rack. Place the lid on the vacuum box system.

11.4.5.2. Place the yellow outer tips into all 24 openings of the lid of the vacuum box. Fit an inner white tip into each yellow tip.

11.4.5.3. For each sample, assemble a TEVA® and a DGA cartridge and lock these onto the inner white tip (DGA cartridge below TEVA®).

11.4.5.4. Place reservoirs on the top end of the TEVA® cartridge.

11.4.5.5. Seal unused openings on the vacuum box by inserting yellow caps included with the vacuum box into unused white tips to achieve good seal during the separation. Alternately, plastic tape can be used to seal the unused lid holes.

11.4.5.6. Turn the vacuum on and ensure proper fitting of the lid.

11.4.5.7. Add 5 mL of 3M HNO_3 to the column reservoir to precondition the TEVA® cartridges.

11.4.5.8. Adjust the vacuum to achieve a flow-rate of ~1 mL/min.

IMPORTANT: Unless the method specifies otherwise, use a flow rate of ~1 mL/min for load and strip solutions and ~2-3 mL/min for rinse solutions.

11.4.6. TEVA® and DGA Resin Separation

11.4.6.1. Transfer the load solution from Step 11.4.4 into the appropriate reservoir. Allow solution to pass through the stacked TEVA® + DGA cartridge at a flow rate of ~1 mL/min.

11.4.6.2. Rinse each tube/beaker with 5 mL of 6M HNO_3 and transfer the solution to the appropriate reservoir (the flow rate can be adjusted to ~2 mL/min).

11.4.6.3. Rinse the columns with 5 mL of 6M HNO_3 (~2 mL/min).

11.4.6.4. Turn off vacuum, discard rinse solutions and remove TEVA® cartridges. Discard TEVA® cartridges and reservoirs and place new reservoirs on the DGA cartridges.

11.4.6.5. Rinse each DGA column with 10 mL of 3M HCl at ~2 mL/min.

11.4.6.6. Rinse each DGA column with 3 mL of 1M HNO_3 at ~2 mL/min.

11.4.6.7. Rinse each DGA column with 20 mL of 0.1M HNO_3 at ~1-2 mL/min.

11.4.6.8. If La was used in alkaline fusion preconcentration steps (i.e., soil matrix), add a 10-mL rinse of 0.075M HNO_3 to remove La from DGA resin at ~1-2 mL/min.

NOTE: The rinses with dilute HNO_3 remove uranium while Cm and Am are retained. Precipitation of uranium during microprecipitation is also inhibited by adding hydrogen peroxide to ensure uranium is present as UO_2^{2+}.

NOTE: If problems with peak resolution are encountered, the volume of the 0.075M HNO_3 rinse may need to be increased to 15 ml to more effectively remove lanthanum prior to eluting Cm.

11.4.6.9. Rinse each column with 15 mL of 3M HNO_3–0.25M HF at ~1-2 mL/min to complex and remove Th from the DGA resin.

11.4.6.10. Place a fresh reservoir onto each column to minimize residual fluoride.

11.4.6.11. Rinse residual fluoride from each DGA column with 5 mL of 4M HCl at ~2 mL/min.

NOTE: Once the HCl has passed through the column, quickly pulse the vacuum two or three times to minimize the amount of residual HCl in the column prior to proceeding.

11.4.6.12. Place clean, labeled plastic tubes in the tube rack under each cartridge. Also place clean connector tips on each column prior to Cm/Am elution.

11.4.6.13. Elute Cm and Am by adding 10 mL of 0.25M HCl solution to each cartridge and reducing the flow rate to ~1 mL/min (or slightly slower).

11.4.6.14. If La was used in alkaline fusion preconcentration steps (i.e., soil matrix), continue eluting Cm and Am by adding 5 ml of 0.25M HCl at a flow rate of ~1 mL/min.

11.4.6.15. Set the Cm fraction in the plastic tube aside for cerium fluoride (CeF_3) microprecipitation, Step 11.5.

11.4.6.16. Discard the DGA cartridge.

11.5. Preparation of the Sample Test Source

NOTE: Instructions below describe preparation of a single sample test source (STS). Several STSs can be prepared simultaneously if a multi-channel vacuum manifold system is available.

11.5.1. Pipet 100 µL (50 µg) of the cerium carrier solution into each tube.

NOTE: Hydrogen peroxide is added to convert tetravalent uranium (U4+) to the hexavalent uranyl ion (UO_2^{2+}) which does not coprecipitate on CeF_3.

11.5.2. Pipet 0.2 mL 30 wt% H_2O_2 into each tube to prevent any residual uranium from precipitating.

11.5.3. Pipet 1 mL of concentrated HF into each tube.

11.5.4. Cap the tube and mix. Allow samples to sit for ~15 minutes before filtering.

11.5.5. Set up a filter apparatus to accommodate a 0.1 micron, 25 mm membrane filter on a microprecipitation filtering apparatus.

Caution: Following deposition of the microprecipitate, there is no visible difference between the two sides of the filter.

11.5.6. If a hydrophobic filter is used, add a few drops of 95% ethanol to wet each filter and apply vacuum. Ensure that there are no leaks along the sides before proceeding.

11.5.7. With vacuum applied, add 2-3 mL of filtered Type I water to each filter and allow the liquid to drain.

11.5.8. Add the sample to the reservoir, rinsing the sample tubes with ~3 mL of water and transfer this rinse to filter apparatus. Allow to drain.

11.5.9. Wash each filter with ~2-3 mL of water and allow to drain.

11.5.10. Wash each filter with ~1-2 mL of 95% ethanol to displace water.

11.5.11. Allow to drain completely before turning the vacuum off.

11.5.12. Mount the filter on a labeled adhesive mounting disk (or equivalent) ensuring that the filter is not wrinkled and is centered on mounting disk.

11.5.13. Place the filter under a heat lamp and dry gently for approximately 5 minutes or longer until it is completely dry.

11.5.14. Count filters for an appropriate period of time by alpha spectrometry.

11.5.15. Discard the filtrate to waste for future disposal. If the filtrate is to be retained, it should be stored in a plastic container since glass will be attacked by HF.

NOTE: Other methods for STS preparation, such as electrode position or microprecipitation with neodymium fluoride, may be used in lieu of the CeF_3 micro-precipitation, but any such substitution must be validated as described in Step 1.5.

12. Data Analysis and Calculations

12.1. Equation for determination of initial screening result, combined standard uncertainty, and radiochemical yield (if required):

12.1.1. The activity concentration of an analyte and its combined standard uncertainty are calculated using the following equations:

$$AC_a = \frac{A_t \times R_a \times D_t \times I_t}{V_a \times R_t \times D_a \times I_a}$$

and

$$u_c(AC_a) = \sqrt{u^2(R_a) \times \frac{A_t^2 \times D_t^2 \times I_t^2}{V_a^2 \times R_t^2 \times D_a^2 \times I_a^2} + AC_a^2 \times \left(\frac{u^2(A_t)}{A_t^2} + \frac{u^2(V_a)}{V_a^2} + \frac{u^2(R_t)}{R_t^2} \right)}$$

where:

AC_a	=	activity concentration of the analyte at time of collection (or other reference time), in picocuries per gram, m^3 or swipe (pCi/g, pCi/m^3, pCi/swipe)
A_t	=	activity of the tracer added to the sample aliquant at the tracer solution reference time (pCi)
R_a	=	net count rate of the analyte in the defined region of interest (ROI), in counts per second (cps)
R_t	=	net count rate of the tracer in the defined ROI (cps)
V_a	=	size of the sample aliquant (g, m^3 or swipe)
D_t	=	correction factor for decay of the tracer from its reference date and time to the midpoint of the counting period
D_a	=	correction factor for decay of the analyte from the time of sample collection (or other reference time) to the midpoint of the counting period
I_t	=	probability of α emission in the defined ROI per decay of the tracer (Table 17.1)
I_a	=	probability of α emission in the defined ROI per decay of the analyte (Table 17.1)
$u_c(AC_a)$	=	combined standard uncertainty of the activity concentration of the analyte (pCi/g, pCi/m^3, pCi/swipe)
$u(A_t)$	=	standard uncertainty of the activity of the tracer added to the sample (pCi)
$u(R_a)$	=	standard uncertainty of the net count rate of the analyte (s^{-1})
$u(R_t)$	=	standard uncertainty of the net count rate of the tracer (s^{-1})
$u(V_a)$	=	standard uncertainty of the size of the sample aliquant volume (g. m^3, swipe)

NOTE: The uncertainties of the decay-correction factors and of the probability of decay factors are assumed to be negligible.

NOTE: The equation for the combined standard uncertainty ($u_c(AC_a)$) calculation is arranged to eliminate the possibility of dividing by zero if $R_a = 0$.

NOTE: The standard uncertainty of the activity of the tracer added to the sample must reflect that associated with the activity of the standard reference material and any other significant sources of uncertainty such as those introduced during the preparation of the tracer solution (e.g., weighing or dilution factors) and during the process of adding the tracer to the sample.

12.1.2. The net count rate of an analyte or tracer and its standard uncertainty are calculated using the following equations:

$$R_x = \frac{C_x}{t_s} - \frac{C_{bx}}{t_b}$$

and

$$u(R_x) = \sqrt{\frac{C_x + 1}{t_s^2} + \frac{C_{bx} + 1}{t_b^2}}$$

where:

R_x	=	net count rate of analyte or tracer (cps)
C_x	=	sample counts in the analyte or the tracer ROI
t_s	=	sample count time (s)
C_{bx}	=	background counts in the same ROI as for x
t_b	=	background count time (s)
$u(R_x)$	=	standard uncertainty of the net count rate of tracer or analyte (cps) [2]

If the radiochemical yield of the tracer is requested, the yield and its combined standard uncertainty can be calculated using the following equations:

$$RY = \frac{R_t}{0.037 \times A_t \times D_t \times I_t \times \varepsilon}$$

and

$$u_c(RY) = RY \times \sqrt{\frac{u^2(R_t)}{R_t^2} + \frac{u^2(A_t)}{A_t^2} + \frac{u^2(\varepsilon)}{\varepsilon^2}}$$

where:

RY	=	radiochemical yield of the tracer, expressed as a fraction
R_t	=	net count rate of the tracer (cps)
A_t	=	activity of the tracer added to the sample (pCi)
D_t	=	correction factor for decay of the tracer from its reference date and time to the midpoint of the counting period

[2] For methods with very low counts, MARLAP Section 19.5.2.2 recommends adding one count each to the gross counts and the background counts when estimating the uncertainty of the respective net counts. This minimizes negative bias in the estimate of uncertainty and protects against calculating zero uncertainty when a total of zero counts are observed for the sample and background.

I_t = probability of α emission in the defined ROI per decay of the tracer (Table 17.1)
ε = detector efficiency, expressed as a fraction
$u_c(RY)$ = combined standard uncertainty of the radiochemical yield
$u(R_t)$ = standard uncertainty of the net count rate of the tracer, (cps)
$u(A_t)$ = standard uncertainty of the activity of the tracer added to the sample (pCi)
$u(\varepsilon)$ = standard uncertainty of the detector efficiency

12.1.3. If the critical level concentration (L_c) or the minimum detectable concentration (MDC) are requested (at an error rate of 5%), they can be calculated using the following equations: [3]

$$L_c = \frac{\left[0.4\times\left(\frac{t_s}{t_b}-1\right)+0.677\times\left(1+\frac{t_s}{t_b}\right)+1.645\times\sqrt{\left(R_{ba}t_b+0.4\right)\times\frac{t_s}{t_b}\times\left(1+\frac{t_s}{t_b}\right)}\right]\times A_t\times D_t\times I_t}{t_s\times V_a\times R_t\times D_a\times I_a}$$

$$\mathrm{MDC}=\frac{\left[2.71\times\left(1+\frac{t_s}{t_b}\right)+3.29\times\sqrt{R_{ba}\,t_s\times\left(1+\frac{t_s}{t_b}\right)}\right]\times A_t\times D_t\times I_t}{t_s\times V_a\times R_t\times D_a\times I_a}$$

where:
R_{ba} = background count rate for the analyte in the defined ROI (cps)

12.2. Results Reporting

12.2.1. The following data should be reported for each result: volume of sample used; yield of tracer and its uncertainty; and FWHM of each peak used in the analysis.

12.2.2. The following conventions should be used for each result:
12.2.2.1. Result in scientific notation ± combined standard uncertainty.

13. Method Performance

13.1. Method validation results are to be reported and documented as required.

13.2. Expected turnaround time per batch of 10-20 samples plus QC:

13.2.1. For swipe or organic-polymer-based air particulate filter samples, ~11 ¼ hours.

[3] The formulations for the critical level and minimum detectable concentration are based on the Stapleton Approximation as recommended in MARLAP Section 20A.2.2, Equations 20.54 and 20A.3.2, and Equation 20.74, respectively. The formulations presented here assume an error rate of $\alpha = 0.05$, $\beta = 0.05$ (with $z_{1-\alpha} = z_{1-\beta} = 1.645$) and $d = 0.4$. For methods with very low numbers of counts, these expressions provide better estimates than do the traditional formulas for the critical level and MDC.

13.2.2. For air particulate filter samples, ~10 hours.

13.2.3. For soil samples, ~9 ¾ hours.

14. Pollution Prevention: The method utilizes small volume (2-mL) extraction chromatographic resin columns. This approach leads to a significant reduction in the volumes of load, rinse and strip solutions, as compared to classical methods using ion exchange resins to separate and purify the Cm fraction.

15. Waste Management

15.1. Types of waste generated per sample analyzed

15.1.1. Approximately 45-65 mL of acidic waste from loading and rinsing the two extraction columns will be generated.

15.1.2. Approximately 25-35 mL of acidic waste from the microprecipitation method for source preparation will be generated. The waste contains 1 mL of HF and ~5 mL of ethanol.

15.1.3. TEVA® cartridge – ready for appropriate disposal.

15.1.4. DGA cartridge – ready for appropriate disposal.

15.1.5. The sample test source consisting of a polypropylene filter disk with ~100 micrograms of CeF_3.

15.1.6. These waste streams may contain low levels of ^{243}Am (added as tracer), ^{244}Cm (added to LCS) and other radionuclides as may be present in samples.

15.2. Evaluate all waste streams according to disposal requirements by applicable regulations.

16. References

Cited References

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Tables, Diagrams, Flow Charts, and Validation Data

17. Tables

Table 17.1 Alpha Particle Energies and Abundances of Importance [1]

Nuclide	Half Life (Years)	Λ (s^{-1})	Abundance	α Emission Energy in kilo electron volts (keV)
^{244}Cm	18.11	1.213×10^{-09}	0.7690	5805
			0.2310	5763
^{243}Cm	29.1	7.55×10^{-10}	0.0150	6066
			0.047	6058
			0.010968	6010
			0.0568	5992
			0.0069797	5876
			0.730	5785
			0.115	5742
			0.015954	5686
			0.0019942	5682
			0.0013959	5639
$^{244/243}Cm$ (combined)	18.11	1.213×10^{-09}	1.000	5805
^{242}Cm	0.4462	4.923×10^{-08}	0.2592	6113
			0.7408	6069
^{245}Cm	8.50×10^{3}	2.58×10^{-12}	0.0058	5529
			0.0083	5489
			0.0045	5370
			0.9320	5361
			0.5000	5304
			0.0032	5234
^{246}Cm	4.76×10^{3}	4.61×10^{-12}	0.822	5387
			0.178	5344
^{247}Cm	1.560×10^{7}	1.408×10^{-15}	0.138	5267
			0.057	5212
			0.0120	5147
			0.0200	4985
			0.0160	4943
			0.710	4870
			0.047	4820
^{243}Am	7.370×10^{3}	2.980×10^{-12}	0.0016	5349
			0.0016	5321
			0.871	5275
			0.112	5233
			0.0136	5181
^{243}Am (combined)	7.370×10^{3}	2.980×10^{-12}	0.9998	5275
^{241}Am	432.6	5.078×10^{-11}	0.0037	5545
			0.00225	5512
			0.848	5486
			0.131	5443
			0.01660	5388

[1] Particle energies with abundances less than 0.1% have been omitted unless they are contiguous with the radionuclide region of interest. Data were queried from the NUDAT database (http://www.nndc.bnl.gov/nudat2/indx_dec.jsp) on 9/19/2014.

Table 17.2 Alpha Emissions Sorted by Decreasing Energy

Nuclide	Half Life (years)	λ (s^{-1})	α Emission Energy (keV)[1]	Abundance
^{242}Cm	0.4462	4.923×10^{-08}	6113	0.7408
^{242}Cm	0.4462	4.923×10^{-08}	6069	0.2592
^{243}Cm	29.1	7.55×10^{-10}	6066	0.0150
^{243}Cm	29.1	7.55×10^{-10}	6058	0.047
^{243}Cm	29.1	7.55×10^{-10}	6010	0.010968
^{243}Cm	29.1	7.55×10^{-10}	5992	0.0568
^{243}Cm	29.1	7.55×10^{-10}	5876	0.0069797
^{244}Cm	18.11	1.213×10^{-09}	5805	0.7690
^{243}Cm	29.1	7.55×10^{-10}	5785	0.730
^{244}Cm	18.11	1.213×10^{-09}	5763	0.2310
^{243}Cm	29.1	7.55×10^{-10}	5742	0.115
^{243}Cm	29.1	7.55×10^{-10}	5686	0.015954
^{243}Cm	29.1	7.55×10^{-10}	5682	0.0019942
^{243}Cm	29.1	7.55×10^{-10}	5639	0.0013959
^{241}Am	4.326×10^{2}	5.078×10^{-11}	5545	0.0037
^{245}Cm	8.50×10^{3}	2.58×10^{-12}	5529	0.0058
^{241}Am	4.326×10^{2}	5.078×10^{-11}	5512	0.00225
^{245}Cm	8.50×10^{3}	2.58×10^{-12}	5489	0.0083
^{241}Am	4.326×10^{2}	5.078×10^{-11}	5486	0.848
^{241}Am	4.326×10^{2}	5.078×10^{-11}	5443	0.131
^{241}Am	4.326×10^{2}	5.078×10^{-11}	5388	0.01660
^{246}Cm	4.76×10^{3}	4.61×10^{-12}	5387	0.822
^{245}Cm	8.50×10^{3}	2.58×10^{-12}	5370	0.0045
^{245}Cm	8.50×10^{3}	2.58×10^{-12}	5361	0.9320
^{243}Am	7.370×10^{3}	2.980×10^{-12}	5349	0.0016
^{246}Cm	4.76×10^{3}	4.61×10^{-12}	5344	0.178
^{243}Am	7.370×10^{3}	2.980×10^{-12}	5321	0.0016
^{245}Cm	8.50×10^{3}	2.58×10^{-12}	5304	0.5000
^{243}Am	7.370×10^{3}	2.980×10^{-12}	5275	0.871
^{247}Cm	1.560×10^{7}	1.408×10^{-15}	5267	0.138
^{245}Cm	8.50×10^{3}	2.58×10^{-12}	5234	0.0032
^{243}Am	7.370×10^{3}	2.980×10^{-12}	5233	0.112
^{247}Cm	1.560×10^{7}	1.408×10^{-15}	5212	0.057
^{247}Cm	1.560×10^{7}	1.408×10^{-15}	5147	0.0120
^{247}Cm	1.560×10^{7}	1.408×10^{-15}	4985	0.0200
^{247}Cm	1.560×10^{7}	1.408×10^{-15}	4943	0.0160
^{247}Cm	1.560×10^{7}	1.408×10^{-15}	4870	0.710
^{247}Cm	1.560×10^{7}	1.408×10^{-15}	4820	0.047

[1]Particle energies with abundances less than 0.1% have been omitted unless they would be contiguous with the radionuclide region of interest. Data were queried from the NUDAT database (http://www.nndc.bnl.gov/nudat2/indx_dec.jsp) on 9/19/2014.

17.1. Ingrowth Curves and Ingrowth Factors

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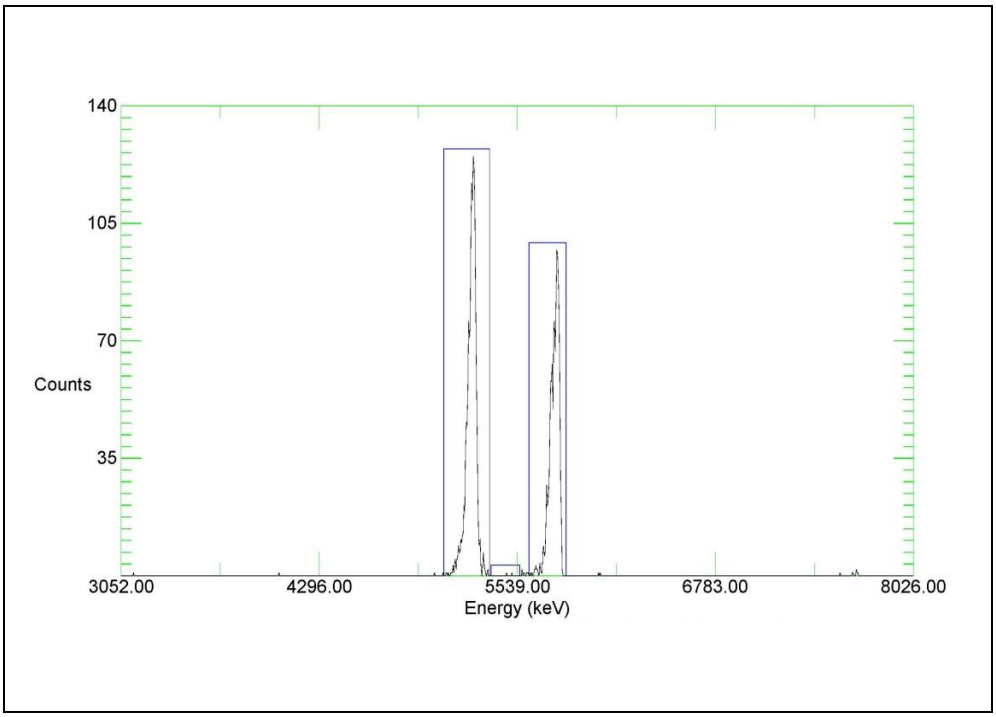
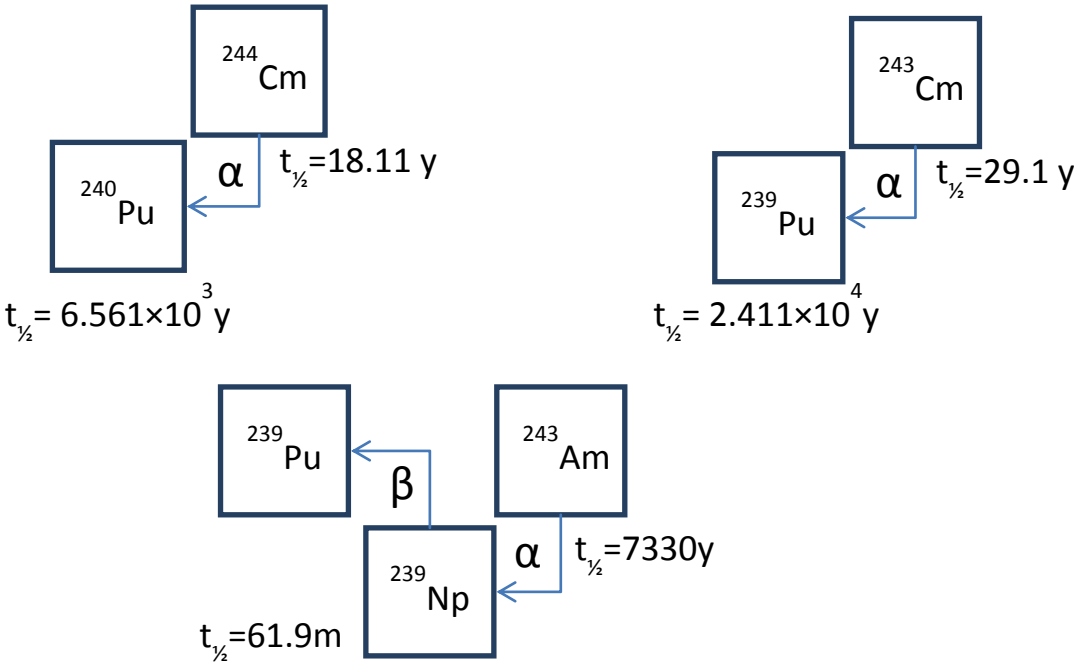
17.2. Spectrum from a Processed Sample



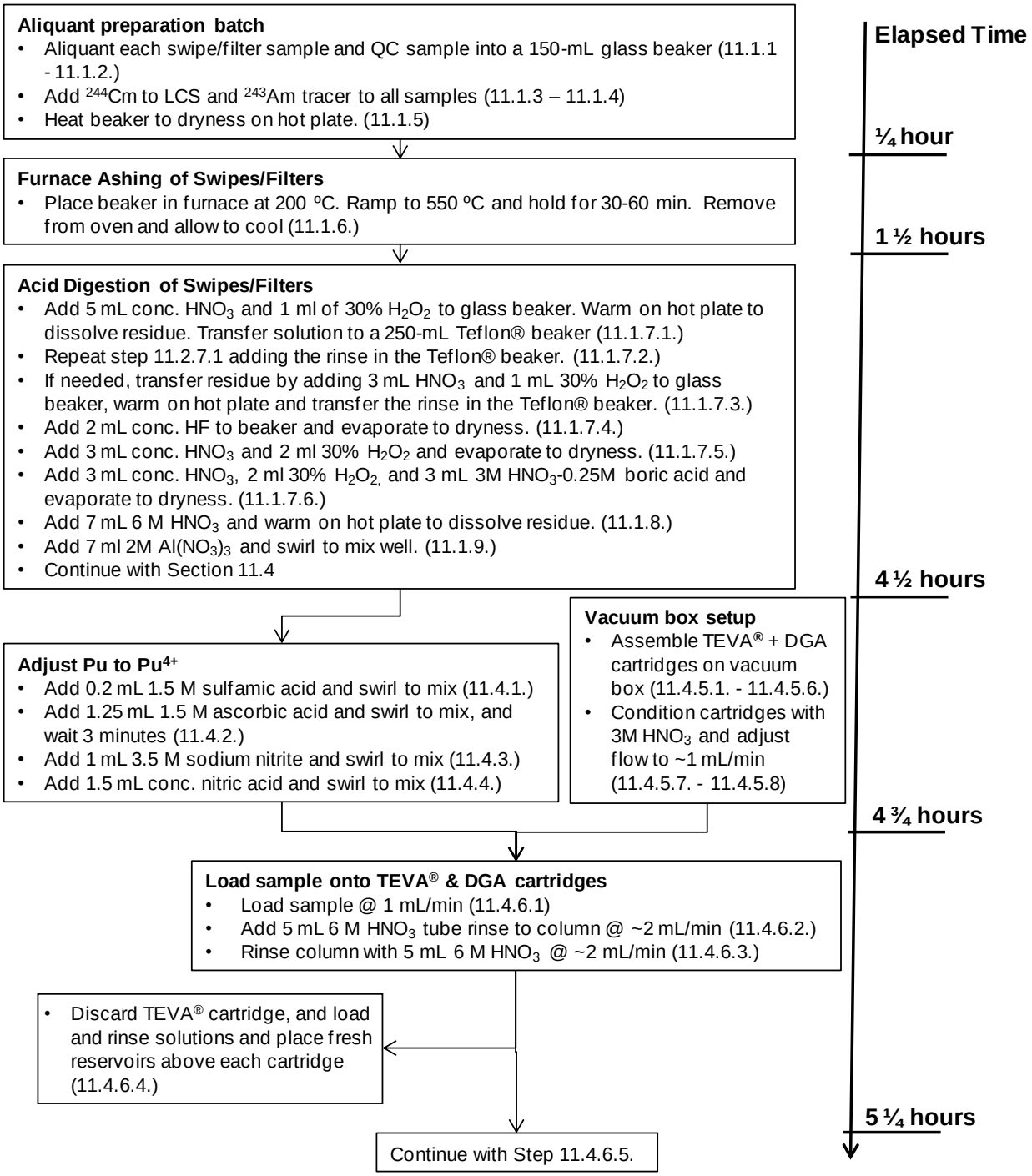
Curium-244 Spectrum

17.3. Decay Scheme

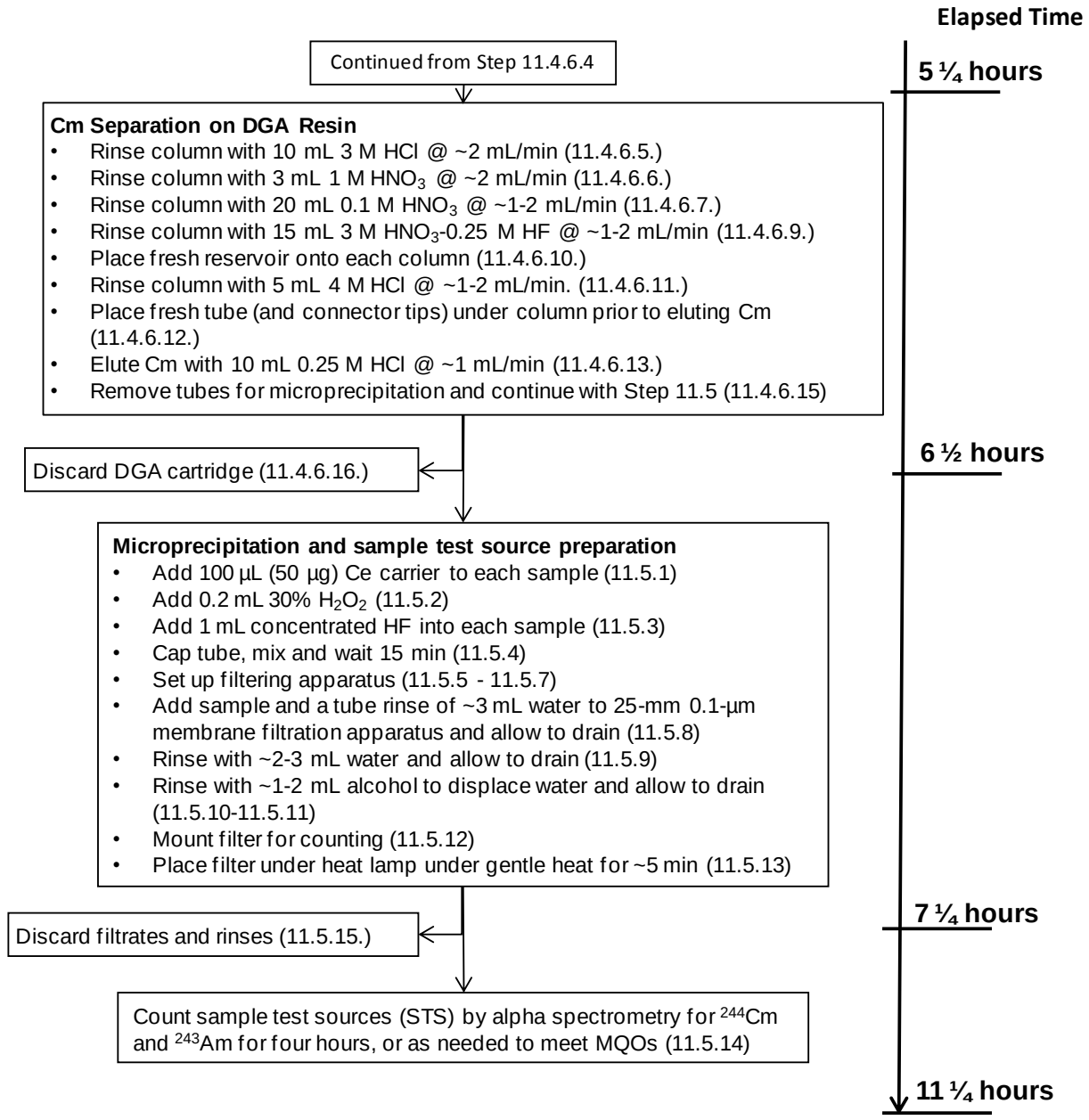


17.4. Flow Chart

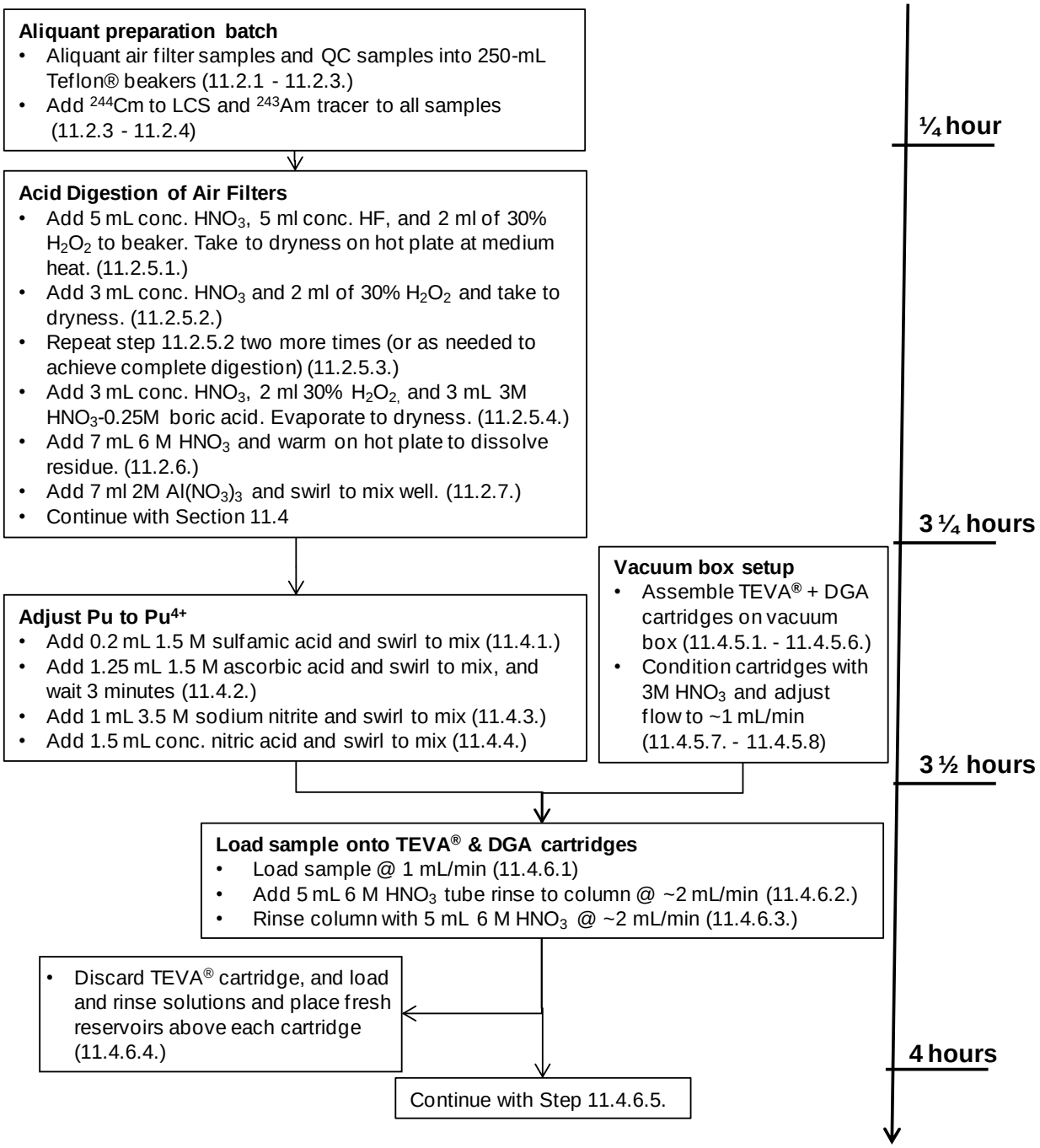
Separation Scheme and Timeline for the Determination of ^{244}Cm in Swipes and Organic-Polymer-Based Air Particulate Filters



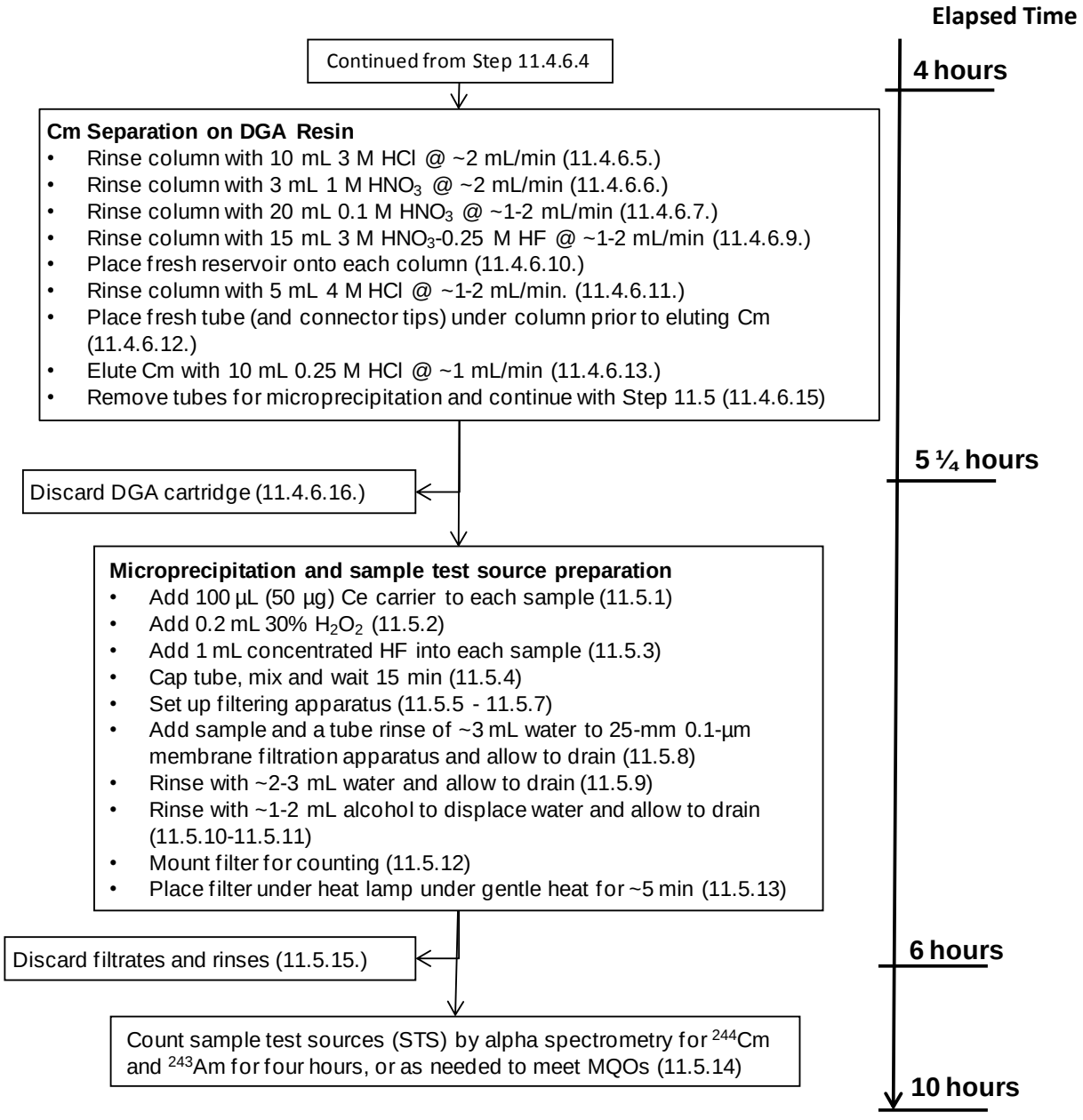
Separation Scheme and Timeline for the Determination of ^{244}Cm in Swipes and Organic-Polymer-Based Air Particulate Filters (cont.)



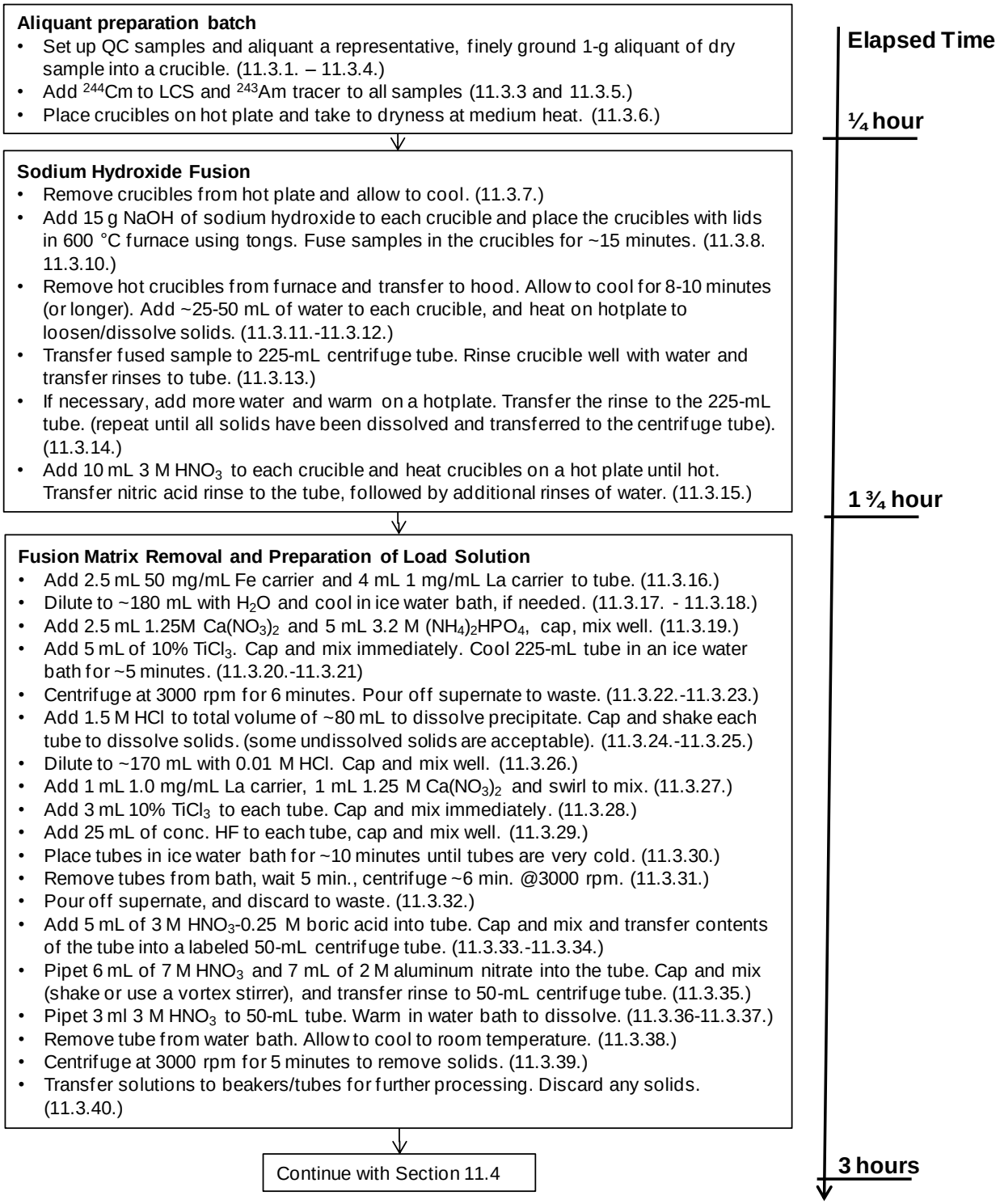
Separation Scheme and Timeline for the Determination of ^{244}Cm in Air Particulate Filter Samples



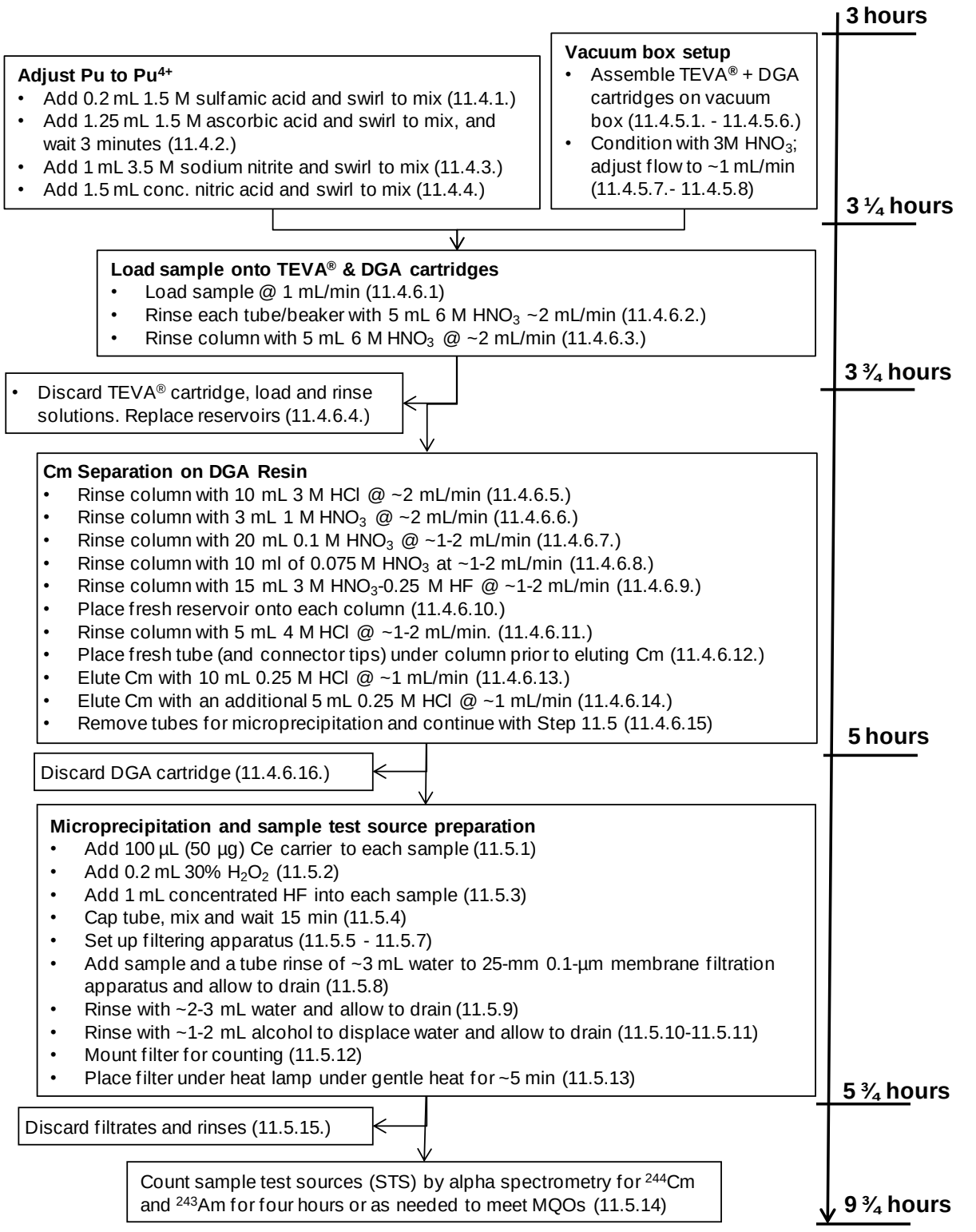
Separation Scheme and Timeline for the Determination of ^{244}Cm in Air Particulate Filter Samples (cont.)



Separation Scheme and Timeline for the Determination of ^{244}Cm in Soil



Separation Scheme and Timeline for the Determination of ^{244}Cm in Soil (cont.)



Appendix A:
Composition of Soil Test Samples Used for Validation

Three soil samples were digested according to the method described section 11.4 of the method. The resulting solution was analyzed by inductively-coupled plasma mass-spectrometry (ICP-MS) to obtain the concentration of 24 metals that would be in solution at the initiation of the chemical separations for Cm. The results of the three replicate determinations were averaged and the results reported below.

Metals by ICP-MS	Concentration (mg/kg) [&]
Be	0.758 (J)
Na	6,200
Mg	4,600
Al	28,400
K	11,170
Ca	33,500
V	35.9
Cr	17.1
Mn	260
Fe	11.990
Co	3.61
Ni	6.72
Cu	5.32
Zn	31.5
As	5.51
Se	1.53
Sr	228.4
Mo	0.92
Ag	0.197 (J)
Cd	0.356 (J)
Sb	0.477(J)
Ba	380
Tl	0.238 (J)
Pb	9.86
Th	4.46
U	1.31
Qualifiers: (J) – Result falls between the Instrument Detection Limit (IDL) and the reporting limit	
[&] Mean of triplicate analyses [#] Mean ± 2 standard deviations of replicate analysis of seven samples	

Appendix B:
National Cooperative Soil Survey

LOCATION OTERO CO+KS NE NM OK WY
Established Series
Rev. LAN/GB/JWB
02/2006

OTERO SERIES

The Otero series consists of very deep, well or somewhat excessively drained soils that formed in alluvium and eolian material. Otero soils are on hills, plains, blowouts, ridges, stream terraces, and fans. Slopes are 0 to 20 percent. The mean annual precipitation is 36 centimeters (14 inches) and the mean annual temperature is 11 degrees C. (51 degrees F.) at the type location.

TAXONOMIC CLASS: Coarse-loamy, mixed, superactive, calcareous, mesic Aridic Ustorthents

TYPICAL PEDON: Otero sandy loam - grassland. (Colors are for dry soil unless otherwise noted.)

A--0 to 15 centimeters (0 to 6 inches); grayish brown (10YR 5/2) sandy loam, dark grayish brown (10YR 4/2) moist; weak very fine granular structure; soft, very friable; strongly effervescent; moderately alkaline (pH 8.0); clear smooth boundary. (8 to 20 centimeters (3 to 8 inches) thick)

AC--15 to 36 centimeters (6 to 14 inches); light brownish gray (10YR 6/2) sandy loam, dark grayish brown (10YR 4/2) moist; very weak medium subangular blocky structure parting to weak coarse granular; soft, very friable; strongly effervescent; moderately alkaline (pH 8.0); gradual smooth boundary. (0 to 23 centimeters (0 to 9 inches) thick)

C--36 to 152 centimeters (14 to 60 inches); very pale brown (10YR 7/3) sandy loam, brown (10YR 5/3) moist; massive; slightly hard, very friable; secondary calcium carbonate occurring discontinuously and at variable depths in the form of soft masses, and in thin seams and streaks; violently effervescent; moderately alkaline (pH 8.2).

TYPE LOCATION: Baca County, Colorado; approximately 563 meters (1,848 feet) west and 61 meters (200 feet) north of the southeast corner of Sec. 6, T. 31 S., R. 50 W.

RANGE IN CHARACTERISTICS:
Soil moisture: moist in some part of the moisture control section for about 40 to 90 cumulative days while the soil temperature is 5 degrees C. (41 degrees) or higher; moist intermittently April though August
Moisture regime: ustic bordering on aridic
Mean annual soil temperature: 8 to 14 degrees C. (47 to 58 degrees F.)
Mean summer soil temperature: 15 to 26 degrees C. (59 to 79 degrees F.)
Depth to secondary calcium carbonate: at the surface, but are noncalcareous for 3 to 25

centimeters (1 to 10 inches) in some pedons.
The weighted average organic carbon content surface 15 inches: 0 to 1 percent, and the organic carbon decreases uniformly with increasing depth
Sand/clay ratios: 3 to 15
Particle size control section:
Clay content: 5 to 18 percent
Silt content: 5 to 35 percent
Sand content: 50 to 82 percent, with 15 to 35 percent being fine or coarser sand
Rock fragment content: typically less than 2 percent, ranges from 0 to 15 percent.

A horizon or AC (if present):
Hue: 7.5YR to 5Y
Value: 4 to 7, and 3 to 6 moist
Chroma: 2 to 4
Where the value is as dark as 5 dry and 3 moist, the horizon is too thin or contains too little organic matter to be a mollic epipedon.
Reaction: neutral to moderately alkaline
Texture: typically sandy loam but includes fine and very fine sandy loams, loam, loamy very fine sand, and loamy fine sand

C horizon:
Hue: 7.5YR to 5YR
Value: 6 or 7, 4 to 6 moist
Chroma: 3 or 4
Calcium carbonate equivalent: 0 to 4 percent
Amount and distribution of visible secondary calcium carbonate is erratic.
Texture: fine sandy loam, sandy loam, and loamy very fine sand.

COMPETING SERIES: These are the Ashollow, Lindrith, and Pitchdraw series.
Ashollow soils formed in residuum from sandstone and do not have visible secondary calcium carbonate in the control section.
Lindrith soils are dry in May and June.
Pitchdraw soils have a paralithic contact at depths of 51 to 102 centimeters (20 to 40 inches).

GEOGRAPHIC SETTING:
Landscape: hills, and alluvial plains
Landform: fans, blowouts, terraces, hills, ridges, and plains (Surfaces are frequently wind-reworked and have a low dune-like relief)
Slopes: 0 to 20 percent.
Parent material: alluvial sediments that have been wind-modified in many places
Mean annual precipitation: 33 to 43 centimeters (13 to 17 inches), with peak periods of precipitation during April through August
Mean annual air temperature: 9 to 12 degrees C. (48 to 54 degrees F.)
Mean summer temperature: 20 to 23 degrees C. (68 to 74 degrees F.)
Frost free period: 120 to 165 days.

GEOGRAPHICALLY ASSOCIATED SOILS: These are the Mitchell, Nelson, Olnest, and Vona soils.
Mitchell soils are coarse-silty.
Nelson soils have an ustic aridic moisture regime.
Olnest and Vona soils have an argillic horizon.

DRAINAGE AND SATURATED HYDRAULIC CONDUCTIVITY:
Drainage well or somewhat excessively drained
Runoff: low to medium
Saturated hydraulic conductivity: high

USE AND VEGETATION: These soils are used for native rangeland or for dry and irrigated cropland.
Native vegetation consists of tall and short grass associations with some yuccas and sand sage.

DISTRIBUTION AND EXTENT: Eastern Colorado, western Nebraska, and southeastern Wyoming. LRR G, MLRA 67; The series is of large extent.

MLRA SOIL SURVEY REGIONAL OFFICE (MO) RESPONSIBLE: Denver, Colorado

SERIES ESTABLISHED: Arkansas Valley Area, Colorado, 1926.

REMARKS: Diagnostic horizons and features recognized in this pedon are:
Ochric epipedon: 0 to 15 centimeters (0 to 6 inches) (A horizon)

The classification is revised from an Ustic Torriorthent to an Aridic Ustorthent. Last updated by the state 3/94.

Changes to the moisture pattern reflect peak periods of precipitation at the type location in Baca County, Colorado.

Modified by Lee Neve in January 2002 to make changes in the format to semitab.
Taxonomic Version: Second Edition, 1999

Modified format by LRM in 1/2006 to include metric conversion and change permeability to saturated hydraulic conductivity.

National Cooperative Soil Survey
U.S.A.

Appendix C:
Composition of Air Particulate Filter Test Samples Used for Validation

Three air particulate filters were digested according to the method described section 11.3 of the method. The resulting solution was analyzed by inductively-coupled plasma mass-spectrometry to obtain the concentration of 24 metals that would be in solution at the initiation of the chemical separations for Cm. The results of the three replicate determinations were averaged and the results reported below.

Metals by ICP-MS	Concentration (ug/filter) [&]
Be	0.041 (J)
Na	38,500
Mg	5,430
Al	5,200
K	1,690
Ca	12,800
V	0.91
Cr	16.1
Mn	4.1
Fe	140
Co	0.056 (J)
Ni	1.23
Cu	1.56
Zn	25.7
As	2.13
Se	0.026 (J)
Sr	8.84
Mo	0.112
Ag	0.307
Cd	0.104
Sb	0.140
Ba	31.7
Tl	0.0085 (J)
Pb	0.564
Th	0.134
U	0.213

Qualifiers:

(U) – Result is less than Instrument Detection Limit (IDL) – <IDL is reported

(J) – Result falls between the IDL and the reporting limit

Footnote: [&] Mean of triplicate analyses

Appenxix D:
Composition of Swipe Samples Used for Validation

Three swipes were digested according to the procedure described section 11.2 of the method. The resulting solution was analyzed by inductively-coupled plasma mass-spectrometry to obtain the concentration of 24 metals that would be in solution at the initiation of the chemical separations for Cm.

Metals by ICP-MS	Concentration (µg/swipe) [&]
Be	0.000490 (J)
Na	186.3
Mg	69.7
Al	4.7 (J)
K	6.88
Ca	166.0
V	<0.00065 (U)
Cr	0.227
Mn	0.80
Fe	11.4
Co	0.0037 (J)
Ni	0.109
Cu	0.50
Zn	2.11
As	<0.00086 (U)
Se	<0.0010 (U)
Sr	1.103
Mo	0.0113 (J)
Ag	0.029 (J)
Cd	0.0185 (J)
Sb	0.070 (J)
Ba	0.439
Tl	0.013 (J)
Pb	0.31
U	0.00083 (J)
Th	0.00083 (J)

Qualifiers:

(U) – Result is less than Instrument Detection Limit (IDL) – <IDL is reported

(J) – Result falls between the IDL and the reporting limit

Footnote: [&] Mean of triplicate analyses