

REMEDIAL ACTION GUIDES
DEPLETED URANIUM AND THORIUM IN SOIL

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ABSTRACT

Aerojet Heavy Metals Company is performing remedial actions for soil and sediments contaminated with depleted uranium and thorium-232 from several areas of its Jonesboro, Tennessee site. This report describes the development of remedial action guides for these activities. The guides are based on existing federal standards and guides and independent environmental radiological pathway analyses, and have been approved by the State of Tennessee which licenses the facility.

The recommended guides are:

Level 1: For unrestricted land use - 35 pCi/g of depleted uranium and 5 pCi/g of thorium-232.

Level 2: For a disposal area on the industrial site, with land use restrictions - 2000 pCi/g of depleted uranium and 50 pCi/g of thorium-232.

These guides are based on the U.S. Nuclear Regulatory Commission guides for onsite disposal of thorium and uranium wastes from past operations, and reliable U.S. Environmental Protection Agency standards and guides.

INTRODUCTION

Aerojet Heavy Metals Company (AHMC) of Jonesboro, Tennessee, is consolidating soils and sediments with radioactive contamination from a storage pile area and from an inactive process waste pond into an on-site disposal area. The disposal area is located adjacent to the developed site area, is inside a fenced area, and is well inside of the AHMC property.

This paper presents guides approved by the Tennessee Department of Health and Environment for conducting the remedial action at the AHMC site. The guides are based on present pertinent radiation guidelines and standards of federal and state agencies and on an independent assessment of potential radiation doses. The guides are specifically developed for the AHMC site, and are the basis for the site design and future land use limitations.

This paper provides a discussion of the Site and Contaminated Material, Existing Guides and Standards, an independent Environmental Pathway Analysis, and guides for the project.

SITE AND CONTAMINATED MATERIAL

The AHMC facility is located several miles south of Jonesboro, in northeastern Tennessee. The site covers about 165 acres, with the industrial facilities covering an area of about 30 acres.

The remedial action is related to a storage pile of soil (about 1000 m²), an inactive process pond (about 4000 m²), and the area around the pond (about 10,000 m²). The material on the storage pile is the result of a prior action when contaminated soil from inside a controlled area was removed. The pond was used in conjunction with former thorium operations and depleted uranium (i.e., UF₆ to UF₄ greensalt) activities. The high mobility and highly contaminated material on the bottom of the pond have been removed and disposed of at a licensed, low-level, radioactive waste disposal site. This action is concerned with contaminated soils beneath the pond and adjoining the pond.

The pond was originally built by excavating material from the moderately sloping hillside and constructing a clay berm on the downslope edge. The excavation for the pond intercepted some bedrock pinnacles. The soils are red, silty clay, tan to brown silty clay to clayey silt. The soils are of low permeability and the groundwater flow is primarily at the soil bedrock interface.

Depleted Uranium and Thorium Contamination

The remedial action is concerned with the consolidation and isolation of depleted uranium and thorium-232 contaminated soil. Uranium and thorium are both naturally occurring radionuclides present throughout nature.

Depleted uranium is a byproduct from uranium enrichment. The material undergoes numerous chemical and physical process steps after being mined as uranium ore before it becomes depleted uranium metal. As a result of these processes, the depleted uranium metal does not contain thorium-230, radium-226 or the decay products of radium-226. Depleted uranium (DU) is "depleted" in uranium-235 (fissile nuclide of natural uranium) compared to natural uranium. The uranium-235 has been removed by the enrichment process.

The thorium-232 releases to the pond were essentially pure thorium, without decay products. However, the decay products of thorium have relatively short half lives (i.e., less than six years) and grow into radioactive equilibrium in a relatively few years. One of the decay products, radon-220, is a noble gas which has many of the radiological, physical, and radiation health risk^{1,2} properties of radon-222. In order to prevent confusion with radon-222, radon-220 will be referred to by its historic name, thoron.

The isotopes of uranium and thorium are primarily alpha emitters. The internal dosimetry for uranium and thorium and their decay products are somewhat similar and is presented in the literature.²⁻⁶ It should be noted that although there is very little external gamma exposure from DU (i.e., radium is not present), because of the relatively short half-lives of the thorium decay products, they grow back and there is considerable external gamma exposure from thorium.

The radiation dosimetry of thoron and its decay products is similar to the dosimetry of radon-222 and its decay products,¹ and the exposure from the inhalation of thoron decay products is also given in the units of working level (WL), the unit often used for radon-222.² However, because of differences in the decay chains, there are basic differences in the working level equivalent to radon daughter concentration ratios for thoron versus radon-222, and also in the relative health significance of the concentrations.^{1,2,7} Whereas the definition of 1 WL is based on a concentration of 100 pCi/l of radon-222 decay products in equilibrium, it only takes the decay products in equilibrium with 7.5 pCi/l of thoron to produce an exposure of 1 WL.⁷ However, because of the decay characteristics of thoron, an exposure of 1 WL from thoron produces only one-third the radiation dose or health significance as 1 WL of radon-222.^{1,2}

EXISTING GUIDES AND STANDARDS

Several federal agencies have issued guides and standards that have bearing on the proposed remedial action.

U.S. Nuclear Regulatory Commission

The U.S. Nuclear Regulatory Commission (NRC) has presented four options for disposal or on-site storage of thorium or DU (there are also other options for non-depleted uranium).⁸ The options, for different materials and types of land use, are based on standards issued by the U.S. Environmental Protection Agency (EPA) and the basic radiation protection standards of the NRC.⁹ The options range from completely unrestricted release of the land to retaining a site under license. The criteria apply to depleted uranium, thorium-232, and the decay products of these isotopes.

Information on the options for DU and thorium-232 is summarized in Table I. The fourth option is storage pending the availability of an appropriate disposal site. There is no requirement for continued licensing of the material or the site for the first three options.

The NRC Option 1 criteria are 35 pCi/g of depleted uranium (total of uranium-234 and -238) and 5 pCi/g of thorium-232. The thorium-232 value is based on the decay products all being present in equilibrium concentrations. These criteria are based on potential radiation doses being equal to or less than those of the EPA proposed Federal Guidance for plutonium,¹⁰ and an external gamma exposure rate of less than 0.01 mrem per hour.¹¹ The EPA trans-uranium guidance is based on a low dose that is appropriate for the general population, and an estimated risk for individuals of less than one chance in a million per year that a person would develop cancer from the exposure.¹⁰

The Option 1 criteria are for completely unrestricted land use, and hence for material on the surface. Although, the area and depth of material is not specified, the criteria specified by the EPA for uranium mill tailings standards (i.e., average over a 100 m² area and 15 cm depth) are considered to be acceptable.

NRC Options 2 and 3 require isolating the material, and in the case of Option 3 limiting the land use. Both of these options are based on applying the dose criteria used in Option 1 for projected exposures. However, the criteria for Options 2 and 3 also recognize the possibility of accidental intrusion into the waste. Option 2, is applicable for unrestricted land use; whereas Option 3 is for burial in areas zoned for industrial use and where the recorded title documents can be amended to state that radioactive material is buried on the land and specify appropriate land use limitations. The criteria in Option 2 are based on a potential dose of 170 mrem per year to an intruder. This is equal to or less than the standards for exposures to an average sample of the population, specified by the NRC in 10 CFR 20 and the National Council on Radiation Protection and

TABLE I
SUMMARY OF REMEDIAL ACTION STANDARDS

Depleted Uranium and Thorium-232 In Soil

Agency and Reference	Intended Use	DU (pCi/g)	Th-232 (pCi/g)
NRC ⁸			
Option 1	Unrestricted land use, surface	35	5
Option 2	Unrestricted land use, buried	300	25
Option 3	Restricted land use, buried	3000	250
EPA ¹²	Unrestricted land use		
	● Surface	--	5
	● Subsurface	--	15
DOE ¹³	Unrestricted land use, surface	150	20

Measurements. 9,14 Option 3, which is limited to applications on industrial sites, is based on a potential dose to an intruder of 500 mrem/yr, which is equal to the general population standards for individuals listed above. The dose criterion for Option 3 is based upon the assumption that intentional intrusion is less likely to occur if there is a warning on land documents not to excavate below specified depths, etc.

The application of either Options 2 or 3 requires consideration of the site characteristics and specifying the burial design. Option 2 requires burial at a minimum depth of about 1.2 m, and Option 3 specifies burial under prescribed conditions. It is necessary to demonstrate that the materials will be stabilized in place and not transported off the site, and that intrusion is unlikely.

The NRC Options are based on assessing the doses from the external gamma, inhalation, and ingestion pathways. However, the inhalation dose pathway for the decay products of thoron, in the thorium-232 chain, does not appear to have been included. The external gamma dose is generally the limiting pathway for thorium-232. 11

U.S. Department of Energy

The U.S. Department of Energy (DOE) has published guides for remedial actions at sites used by the former U.S. Army Corps of Engineers Manhattan Engineering District and the U.S. Atomic Energy Commission primarily to process and/or store uranium ores and their products. 13 The FUSRAP sites include private land and Federal land.

Many possible scenarios for exposure have been considered. The "intruder/agriculture" scenario, where a family, unaware of the presence of the residual material builds a home and raises food crops, is projected to result in the maximum dose to individuals.

The DOE guides are based on a plot of contaminated waste 140 m x 140 m, 1.5 m deep, at the surface; i.e., not covered. The guides are based on maximum potential whole body doses to individuals of less than 500 mrem/year, and individual organ doses of less than 1500 mrem/year. The DOE guide for uranium is 75 pCi/g of uranium-238, assuming uranium-234 is also in equilibrium. Hence, the guide for total uranium (i.e., equivalent to the NRC guides) is 150 pCi/g. This assumes thorium-230 and the subsequent decay products are not present. DOE does not provide a specific guide for thorium-232, but suggests a limiting value of 40 pCi/g based on external gamma exposure if the wastes are on the surface of the ground. This value does not include an assessment of the potential exposure from thoron. DOE is presently reevaluating these guides.

U.S. Environmental Protection Agency

The U.S. Environmental Protection Agency (EPA) has promulgated several radiation standards that relate to defining acceptable concentrations of radionuclides in soil and general radiation exposure. The most applicable EPA standards are Parts D and E of 40 CFR 192 12 which present standards for the control of byproduct materials from uranium ore processing.

The EPA guidance for transuranium elements in soil 10 and the Uranium Fuel Cycle Standards, 40 CFR 190 15 provide useful guidance on acceptable

levels of radiation exposure. The dose guidance in the EPA transuranium guides has been used as the basis for the NRC guidance for burial of thorium and uranium wastes. 8,11 The basic EPA criteria are 1 mrad and 3 mrad per year to the lung and bone, respectively. Assuming a quality factor of 20, the dose equivalents are 20 and 60 mrem per year, respectively, for the lung and bone. Using the organ dose weighting factors of the ICRP, 5 this is similar to a whole-body equivalent dose of about 10 mrem. The fuel cycle standards are based on an annual dose of 25 mrem to the body or any organ other than the thyroid. The NRC has referenced and used this standard in the development of regulations for low-level waste disposal. 16

The pertinent EPA uranium mill tailings standards are given as concentrations of radium-226 and thorium-232 in soil. 12 The standards for radium-226 in soil are primarily based on the risks associated with radium-226 in tailings and the potential of subsequent exposure to radon-222 in buildings built on contaminated soil. 2 Although some pathway and risk assessments for thorium-232 were performed, the EPA standards in 40 CFR 192 Part E, are basically an extension of the radium-226 standards. 2

The pertinent EPA standards in 40 CFR 192 are 5 pCi/g of radium-226 in the surface soil, and 15 pCi/g at depths below 15 cm. It is specified that these concentrations can be averaged over 15 cm (6 in) depths and areas of 100 m². 2 These standards are extended to thorium-232.

Other guides for the concentration of radionuclides in soil have been given by Healy et al. 17 and the Conference of Radiation Control Program Directors.

A summary of the guides is given in Table I.

Past Implementation of Standards for the Cleanup of Sites

A number of sites contaminated with uranium and/or thorium-232 decay chain radionuclides have been cleaned up. These sites include: Middlesex, New Jersey; the Frankford Arsenal in Pennsylvania; the AMAX Site in West Virginia; Vesical Chemical Corp. in Michigan; and Huntington Alloys Inc., in West Virginia. Open literature reports are not available on all of the sites.

The NRC guides for uranium and thorium disposal were used for cleaning up the Frankford Arsenal Site, 18 the AMAX Site, 19 and the Huntington Alloy Site. The Frankford Arsenal site, which contained depleted uranium from the development and testing of depleted uranium armor-piercing munitions was cleaned up using Option 1 (i.e., 35 pCi/g). The AMAX site was cleaned up using the NRC Option 1 criteria for much of the site, and Option 3 for burying material on the site. 18 The burial area is covered with a clay cap and future uses of the site are limited.

The Vesical Chemical Corp. site in St. Louis, Michigan, was cleaned up using criteria of 65 pCi/g for uranium-234 plus -235 and 5 pCi/g for thorium-232. The guides were based on concurrence by EPA and NRC (private communication).

ENVIRONMENTAL PATHWAY ANALYSIS OF RADIONUCLIDES IN SOILS

Environmental pathway analyses provide perspective in applying remedial action criteria for the various radionuclides on a consistent risk basis. The

methodology employed in the analyses is from the RQ/PQ environmental pathways model.²⁰ The pathways considered are:

- Radioactive gas transport
- Leaching from the soil to an adjacent stream
- Direct external gamma radiation
- Uptake by vegetation grown in the soil
- Inhalation of contaminated dust

These pathways pertain to exposures from material on the surface of the ground, transport by groundwater, and the potential exposure of individuals who may intrude into the waste. There is essentially no exposure from buried depleted uranium and thorium-232 for the gas transport, external gamma, and dust inhalation pathways (i.e., the pathways only apply to surface deposits and intrusion events). The leaching and, to a much lesser extent, the vegetation pathways apply to both surface and buried deposits.

The gas transport pathway pertains to the release of thoron, a gaseous decay product of the thorium-232 decay chain. The production and transport of thoron is somewhat similar to that of radon-222; hence, the radium-226/radon-222 pathway is included to provide a comparison between radium and thorium guides and the EPA standards for uranium mill tailings. The half-life of radon-222 (i.e., produced by radium-226) is sufficiently long that even when material is buried by 0.5 m of cover, a large fraction of the radon-222 diffuses to the surface. Since the half life of thoron is about one minute, a soil cover of about 10 cm will reduce emissions to background levels. Hence, a cover of 0.5 m essentially eliminates any thoron release. The pathway calculations for this assessment only assume the presence of a 5 cm layer of stagnant air--analogous to a wood floor and subfloor.

The primary risk associated with the gas transport pathway for radon and thoron is their accumulation in buildings and the subsequent ingrowth of the decay products because of reduced air exchange. For relatively small deposits of thorium in open land areas, even if the material is on the surface, thoron gas is dispersed in the atmosphere before there is any significant ingrowth of decay products.

The second pathway, leaching of material, involves the potential off-site transport of the nuclides and is the pathway that could affect the largest segment of the population. This pathway applies to both surface and subsurface deposits but for subsurface requires that subsurface water interact with the material.

The last three pathways pertain to doses from external gamma radiation, from the inhalation of dust created by disturbing the contaminated soil, and from the consumption of vegetation grown in the contaminated soil. Potential exposure via these pathways primarily pertains to material on the surface or brought to the surface by intrusion scenarios. The vegetation pathway is based on vegetation growing in soil contaminated throughout the total root zone. In order to be conservative the vegetation pathway is considered for both surface and buried waste.

In order to provide consistent impacts from the ingestion and inhalation of radionuclides and to use parameters similar to those used to develop the NRC guides, critical organ dose factors were taken from Reference 4. A quality factor of 20, to correspond to the assessments for the NRC guides and ICRP recommendations was used. Values of other key parameters

quantifying the scenarios and pathways were obtained from References 3, 6, and 20.

The results of the pathway analyses are summarized in Table II. The results are expressed in terms of the exposure rate per 100 pCi/g of nuclide in the soil. The doses from ingestion are for the bone and for inhalation of dust are for the bone or the lung, whichever is highest. A value of 0.02 WL for radon-222 daughters for 5 pCi/g of radium-226 is used to normalize the analysis for radon.

It is hard to directly compare the radon and thoron working level exposures to the other exposures. A comparison can be made by equating the working level exposures to health effects or radiation doses and calculating the equivalent internal radiation dose. Although the radon and thoron exposures can be converted to doses there is considerable uncertainty in the dose conversion parameters.

Comparisons using parameters from NRC⁶ and the NCRP²¹ indicate that an annual exposure to 1 WL is equivalent to a radiation dose of over 100 rem to over 1000 rem. Hence, it is readily apparent that the radon-222 gas pathway is the limiting pathway for radium-226. For thorium-232 the equivalent radiological exposure for external gamma exposure and the exposure to the decay products of thoron are potentially of the same order of magnitude. However, because of the short half life of thoron, and hence its short diffusion path, the potential for exposure for the thoron pathway is less than indicated by the above analysis. Hence for most land use scenarios (e.g., construction of buildings) the limiting exposure pathway for thorium-232 will be the external gamma dose. The results indicate that the reclaimer scenarios yield the largest exposures, with the radioactive gas pathway being limiting for the thorium-230/radium-226 chain, and the external gamma pathway being limiting for the thorium-232 chain.

The analysis indicates that the limiting exposures for uranium are by the soil to vegetation

TABLE II
ENVIRONMENTAL PATHWAY ANALYSIS

Pathway	Dose Equivalent Based on 100 pCi/g (mrem/yr per 100 pCi/g)		
	Radionuclide Being Assessed		
	Th-232	U-238	Ra-226 For Comparison
Groundwater to Stream	0.08	0.04 ^a	1.0
Soil to Vegetation	40	10 ^a	40
Inhalation of Dust	30	0.7 ^a	26
External Gamma	1100	12 ^b	700
WL Exposure for Thoron and Radon-222	0.02 WL		0.4 WL

- a There is normally only about 0.15 pCi of uranium-234 per pCi of uranium-238 in DU. The total dose is the sum from both isotopes.
- b The exposure is from the decay products between uranium-238 and uranium-234. Hence the base concentration is uranium-238.

and external gamma pathways. Since uranium-234 contributes to the dose via the soil to vegetation pathway but does not contribute to the external gamma dose, the soil vegetation pathway is generally limiting. Pathway analyses by NRC,¹¹ DOE,¹³ and Healy et al.¹⁷ have generally indicated that the inhalation of dust was much more significant than the external gamma exposure. However, these analyses have generally used airborne dust loadings of 5000 micrograms/m³. Although it is possible to obtain such elevated airborne dust concentrations, they would not be sustained for long periods of time. The assessment in Table II is based on a more reasonable loading of 100 micrograms/m³, and hence indicates lower doses than projected by the NRC and DOE analyses.^{13,11}

GUIDES FOR REMEDIAL ACTION AT AHMC

The previously discussed standards and guides cover a range of values. The differences are due to the varying degrees of isolation of the material, differences in the dose standards used to derive the guides, and differences in the pathway models used to estimate potential doses associated with the guides. The extent to which material is isolated or excluded from the general environment is one of the most important aspects for specifying different concentration guides. The isolation can be provided by restrictions for land use, by placing the material below the surface of the ground or by burying the material according to special design specifications. The differences in the NRC Options primarily result from different degrees of isolation. These vary from unrestricted land use, to burial and limiting the use of the land.

There is over an order of magnitude difference in the basic radiation standards that were applied to derive the different soil concentration guides. The NRC and EPA guides are derived from projected doses equivalent to a whole body dose of about 25 mrem/yr (excludes radon); whereas, the DOE guides are based on whole body doses of about 500 mrem/year and individual organ doses of 1500 mrem/year.

The recommended guides for the AHMC site are based on limiting projected annual doses to less than about 25 mrem/year and applying the principles of ALARA. Specifically, it is proposed that the criteria of NRC Options 1 and 3 be applied. The independent pathway analyses indicates that the projected radiation doses will be below the NRC dose projections used to determine the guides. The AHMC site is an industrial facility and the property documents will be amended to indicate that material is buried and that the specified area shall not be used for agriculture or to build residential or industrial buildings. Furthermore, the isolation and size of the proposed disposal area makes it generally undesirable for agriculture use or for constructing buildings. The recommended guides are based on burying material under a minimum of about 1.2 m (4 ft) clay cover. The buried material will be above the 100-year flood plain and above the soil-bedrock interface and is confined between the hillside and a massive rock buttress wall. The surface will be contoured and planted to limit erosion. For added assurance, AHMC will continue to monitor the site to ensure that material is not dispersed by groundwater and that the integrity of the disposal area is maintained.

The intent of the AHMC remedial program is not only to clean up the proposed site; but also to minimize the potential for human exposure and minimize

the need for controls. To meet these objectives two levels of guidance are proposed:

Level 1 - Completely unrestricted use. This guide will be used to specify the cleanup of the storage pad and the associated area and the low areas around the pond and generally within or near the 100-year flood plain.

Level 2 - Material to be confined and buried beneath a 1.2 m cover of material. The cover will include at least 0.6 m of compacted clay to prevent infiltration of precipitation. This guide will be applied to material in the proposed burial area.

The Level 1 guides, for total unrestricted use are 35 pCi/g of total uranium as depleted uranium, and 5 pCi/g of thorium-232 with the decay products in radioactive equilibrium. These are the NRC Option 1 guides for total unrestricted use.⁸ These guides correspond to the EPA transuranium guidance,¹⁰ the EPA uranium fuel cycle standards,¹⁵ and EPA standards for control of byproduct materials from ore processing.¹² Uncontrolled use of the land would result in a maximum radiological risk to an individual of about one in a million per year. This is a small fraction of the risks people are exposed to in life.^{22,23} For example, this is less than the risk of smoking several cigarettes.²²

Level 2 guides of 2000 pCi/g of depleted uranium and 50 pCi/g of thorium-232 will be used for material which will be buried in the hillside, above a clay liner, below a 1.2 m clay cover. The AHMC facility is an industrial site and the land title documents will be amended to indicate the presence of the material and restrictions for future land uses. Although monitoring by AHMC has indicated that the uranium is immobile and much of the uranium released was as immobile compounds, a conservative intermediate value based on potential partial solubility is used. The difference in the limits for soluble versus insoluble uranium is based on the potential dose for the agricultural pathway for soluble uranium. This is an industrial site and future agricultural use will be excluded. Hence, an intermediate value between the NRC guides of 3000 pCi/g and 1000 pCi/g for insoluble and soluble depleted uranium, respectively, is used.

The Level 2 guide for thorium-232 is based on the dose criteria for NRC Option 3,⁸ but a reduced concentration guide is recommended. The independent pathway analysis (see Table II) indicates that the external gamma dose for intrusion for the NRC soil guide of 250 pCi/g for thorium-232 may be as high as 2700 mrem/yr, versus the specified dose guide of 500 mrem/yr.^{8,11} Therefore, in order to be conservative, the recommended Level 2 guide for the AHMC remedial action is 50 pCi/g of thorium-232, or one-fifth of the NRC criteria. Assuming there is no inadvertent intrusion, the analysis by NRC and the external analysis indicates that implementation of the recommended guidance, as specified, will result in projected doses similar to or less than the basic radiation standards that have been specified by EPA and the NRC criteria for disposal of thorium and uranium wastes.

The recommended Level 2 guides are based on NRC Option 3 which limits operational (e.g., not intrusion) doses to about 1 mrad to the lung and 3 mrad to

the bone per year (e.g., based on QF of 20, 20 mrem and 60 mrem, respectively). Even in the event of human intrusion into the burial site, the doses are projected to be less than about 500 mrem/yr, which is the basic radiation protection standard for individuals in the general population.⁹ As previously noted, the potential risks due to the yearly projected radiation exposures are less than the risks of smoking several cigarettes.²² Furthermore, even in the unlikely event of individuals intruding into the buried wastes, the yearly risks are similar to those of traveling about three thousand miles by car or smoking about one hundred cigarettes.²²

Extensive surface and core sampling by AHMC has indicated that the depleted uranium or thorium-232 has not extensively reached or contacted the bedrock. Therefore, it is projected that the pond bottom, etc., can be excavated to implement the Level 1 guides. However, if low-level concentrations of depleted uranium have penetrated the bedrock interface in limited areas, intermediate guides between Levels 1 and 2 will be applied to minimize the need to excavate bedrock. The alternate guides are three times the Level 1 criteria. This is within the context of the EPA standards,¹² where the standards for radium-226 concentrations 15 cm beneath the surface are three times the surface (equivalent to the Level 1 criteria) standards. The criteria for hard to excavate material at the soil-bedrock interface are 100 pCi/g for depleted uranium and 15 pCi/g of thorium-232. These alternate criteria are at or below the NRC Option 2 criteria, and the subject material will be greater than 1.2 m below the surface after the area has been backfilled and contoured.

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