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Title: Air Modelling As An Alternative To Sampling For Low-Level Radioactive Airborne Releases

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Air Modelling As An Alternative To Sampling For Low-Level Radioactive Airborne Releases

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INTRODUCTION

The Environmental Restoration Program for Department of Energy (DOE) nuclear weapons complex facilities faces challenges ranging from well-defined cleanups of specific contaminated sites to broader questions of the effects on the environment and human health of forty years of laboratory operations. Efforts in the latter category were accelerated by the promulgation of the National Environmental Policy Act (NEPA) of 1970, the Resource Conservation and Recovery Act (RCRA) of 1976, the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) of 1980, and the Superfund Amendments and Reauthorization Act (SARA) of 1986 which require the DOE nuclear weapon complex facilities to comply with the same standards applied to industry. The DOE responded to the new regulatory requirements by the implementation of the Comprehensive Environmental Assessment and Response Program (CEARP), designed to bring LANL into compliance with RCRA and CERCLA.[1-4] A particularly striking example of the effect of these regulatory requirements is the new scrutiny focused on airborne radioactive releases from DOE laboratories and the cumulative effect of past releases. This paper describes our efforts to assess the effect of these airborne releases at one DOE laboratory using air modelling based on historical data.

Among the facilities affected by these developments is Los Alamos National Laboratory (LANL) in New Mexico. LANL is located on several mesas above the Rio Grande, encompassing 42 square miles of land approximately 30 miles northwest of Santa Fe in north central New Mexico. LANL was established in 1943 for Project Y of the Manhattan Project to develop the world's first nuclear weapon. Currently, LANL is operated by the University of California for the Department of Energy. LANL continues to conduct research in a variety of military and civilian areas.[5]

RCRA, as amended by the Hazardous and Solid Waste Amendments (HSWA) in 1984, requires all facilities which involve the treatment, storage, and disposal of hazardous waste obtain a RCRA/HSWA waste facility permit. LANL complied with CEARP by initiating a process of identifying potential release sites associated with LANL operations prior to filing a RCRA/HSWA permit application. In the process of preparing the RCRA/HSWA waste facility permit application to the U.S. Environmental Protection Agency (EPA), a total of 603 Solid Waste Management Units (SWMUs) were identified as part of the requirements of the HSWA Module VIII permit requirements.[6] The HSWA Module VIII permit requires LANL to determine whether there have been any releases of hazardous waste or hazardous constituents from SWMUs at the facility dating from the 1940's by performing a RCRA Facility Investigation to address known or suspected releases from specified SWMUs to affected media (i.e. soil, groundwater, surface water, and air). Work plans for the RCRA facility investigations must be submitted to the U.S. EPA for evaluation. The permit also requires LANL to take corrective actions for such releases.

Among the most troublesome of the potential releases sites are those associated with airborne radioactive releases. Air quality has been continually monitored to assess compliance with all applicable regulations, however it is conceivable that routine low-level radioactive emissions along with some accidental releases may have contaminated soil in the area. RCRA provides no guidelines for radioactive soil contamination, but investigation of these matters is required by the terms of LANL's RCRA/HSWA permit.

In order to assess health risks associated with radioactive contaminants in a manner consistent with exposure standards currently in place, the DOE and LANL have established Screening Action Levels (SALs) for radioactive soil contamination. The maximum allowable dose was set at 10 mrem/year, which is the same as the maximum effective dose equivalent for members of the public from ambient air established by the National Emission Standards for Hazardous Air

Pollutants (NESHAPs). The SALs for each radionuclide in soil are derived from calculations based on a residential scenario in which individuals are exposed to contaminated soil via inhalation and ingestion as well as external exposure to gamma emitters in the soil. The applicable SALs are shown in table 1.[7]

SITE HISTORY

A group of facilities at Technical Area 3 of LANL (shown in figures 1 and 2), located adjacent to the town of Los Alamos, have emitted very low levels of radioactive airborne contamination throughout their lifetime. Some small unplanned releases have also occurred. Perhaps the most important source is the Chemical and Metallurgical Research Building (CMR Building). This building has housed a variety of research and development and analytical operations handling radioactive material. One wing of the CMR Building contains a facility for examining irradiated nuclear fuels. Operations at the CMR Building involve isotopes of uranium and plutonium, iodine, mixed fission products and tritium. Effluent from hood stacks at the CMR Building has been passed through High Efficiency Particulate Air (HEPA) filters as well as filters using fabric, charcoal, and Aerosolve 95®.

Radioactive emissions from the stacks have been monitored. The available data indicate that almost all of the radioactivity emitted from the CMR Building has been in the form of tritium (11,000 Curies) along with fractions of a Curie of the other isotopes listed above. Data are available from the beginning of operation in 1953 to the present.

Several other buildings housing radiochemical operations are located near the CMR Building. The documentation on airborne radioactive releases from these sites is less complete than that for the CMR Building. Of particular importance are the Cryogenics Building and the Van de Graaff Accelerator Laboratory. The former is known to have released 28,000 Curies of tritium from 1976 to 1985, but data on earlier releases (the facility opened in 1955) are not available. The Van de Graaff Accelerator Laboratory, in operation since 1951, released 14,000 Curies of tritium

from the 1960's through 1992. Several other facilities in the same locale may have been responsible for considerably smaller releases. An important consequence of the uncertainty in the historical data on release rates is that attempts to evaluate the environmental significance of these releases must allow for the effect of different hypothetical release rates to be determined.

APPROACH

As discussed above, neither RCRA, the Clean Air Act nor other regulations provides specific guidelines for investigations of this kind of potential release site. In the absence of specific requirements, our approach was guided by three primary concerns. The first is that, during the period of operation of these facilities, a great deal of construction has occurred around the CMR Building (figures 1 and 2). Sampling is thus impractical both because the area is now covered with buildings and because radiochemical operations in these facilities would make it difficult to conclusively attribute any contamination found to airborne contamination.

The second concern is to establish that no significant radioactive contamination reached the townsite, located about a mile north of the CMR Building. Finally, any study of this site must account in a reasonable way for the uncertainties arising from incomplete release data. All of these considerations provide grounds for the conclusion that air modelling is a more appropriate strategy than soil sampling for evaluating this potential release site.

All of the facilities concerned are located within 800 feet of the CMR Building. Although there have been some small "puff releases" from these facilities, the overwhelming majority of the radioactivity was released at a low steady rate. Thus, the emissions can be modeled as a point source exposed to the year-round average wind distribution.

Neither legal requirements nor DOE regulations specify the method to be used in calculating the geographic distribution of radionuclides for the purpose of determining soil contamination. However, two computer programs, CAP-88 (Clean Air Act Assessment Package-1988) and

AIRDOS-PC, have been approved by the EPA for determining compliance with NESHAPs limits on airborne radioactive exposure (other than radon) at DOE facilities.[8]

CAP-88 consists of modified versions of the programs AIRDOS-EPA and DARTAB.[9] It was selected for our modelling effort because of the high correlation observed between CAP-88 predictions of annual average ground-level concentration and the actual environmental measurements by the U.S. EPA Office of Radiation Programs.[9] In addition, CAP-88 has been used by the LANL's Radionuclide Air Emission Management group to determine the effective dose equivalents for NESHAPs compliance for airborne radionuclide emissions.

CAP-88 uses a modified Gaussian plume equation of Pasquill (an atmospheric dispersion equation)[10,11] in conjunction with the local meteorological and population data to estimate the effective dose equivalent as well as the radionuclide air concentration, dry deposition rate, wet deposition rate, and ground deposition rate in 16 directions at various distances around a point source.[9]

In the CAP-88 calculation, all the stacks were considered as one point source of radioactive air emissions due to their proximity. All radioactive air emissions were assumed to be in the form of particulates. Heavier annual precipitation, slower stack gas exit velocity, a lower mixing height, and a lower stack height were used instead of the actual parameters in the CAP-88 calculation to ensure conservative results. The plume rise is calculated based on the exit gas at ambient temperature. Meteorological data collected at the nearest meteorological station, Technical Area 6, and Los Alamos population data were used for the calculation.

The average release rate for each isotope at each facility was calculated for the years in which data were available. These calculated release rates were then extrapolated for periods of facility

operation for which emissions data are not available. A mixing depth of 1 mm was assumed and decay of the radioisotopes was neglected as a further conservative measure.

RESULTS

The calculated radioactive deposition distribution is shown graphically in figure 3. The distribution is fairly symmetrical with somewhat higher contamination levels east of the source than in other directions. Significantly, the modelling results indicate that contamination levels decrease by two orders of magnitude before reaching the townsite boundary north of the source.

More important than the distribution of contamination, however, are the very low calculated contamination levels. Our results are based on the ground deposition rate, which is the highest rate available from CAP-88. Nonetheless, the level of contamination at the point of maximum deposition is still far less than the SALs, and of no toxicological significance. For example, the maximum tritium contamination level is calculated to be 1.2×10^{3} pCi/g dry soil; the corresponding SAL is 1.5×10^{7} pCi/g dry soil. The calculated maximum uranium-235 contamination level is 2.6×10^{-5} pCi/g dry soil (SAL = 18 pCi/g dry soil). Likewise, the calculation demonstrates that airborne radioactive contamination of the townsite or other areas of the laboratory site has not occurred to a significant extent.

CONCLUSIONS

An important advantage of this approach to historical airborne radioactive contamination problems is that one can allow for errors in the reported values of radioactive release rates. In any reasonable model, calculated ground contamination levels vary linearly with the release rate. Thus, the fact that the calculated contamination levels are several orders of magnitude below the SALs indicates that the actual release rates would have to be several orders of magnitude greater than those measured in order for soil contamination levels to approach the SALs.

In cases of the sort described here— where construction around the release site makes sampling infeasible or where other potential sources of contamination exist— air modeling represents the most practical and responsible means to carry out a RCRA facility investigation for airborne radioactive releases. Moreover, the software are easy to use and have been accepted by EPA, reducing the cost of performing and documenting the investigation. The DOE Environmental Restoration Program activities will likely present many additional opportunities to demonstrate the utility of this modelling approach.

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Table 1. LANL Screening Action Levels for Radionuclides in Soils

Radionuclides	Soil Screening Action Levels (pCi/g dry soil)
Iodine-129	41.0
Plutonium-238	27.0
Plutonium-239	24.0
Strontium-90	8.90
Tritium	1.5×10^7
Uranium-234	86.0
Uranium-235	18.0
Uranium-238	59.0

(from reference 7)

FIGURE CAPTIONS

Figure 1. Aerial view of Technical Area 3 in 1955 looking south. The CMR Building is the multi-wing structure located in the center of the picture. The H-shaped structure is the LANL Administration Building.

Figure 2. Aerial view of Technical Area 3 in 1994 looking east. The LANL Administration Building is at the center of the picture. A portion of the CMR Building can be seen on the right.

Figure 3. Contour plot of airborne uranium-235 soil contamination as calculated by CAP-88. The contours correspond to contamination levels of 3.2×10^{-6}, 1.0×10^{-6}, 3.2×10^{-7}, 1.0×10^{-7}, 3.2×10^{-8} pCi/g from center outwards. The broken line represents the boundary of LANL property.

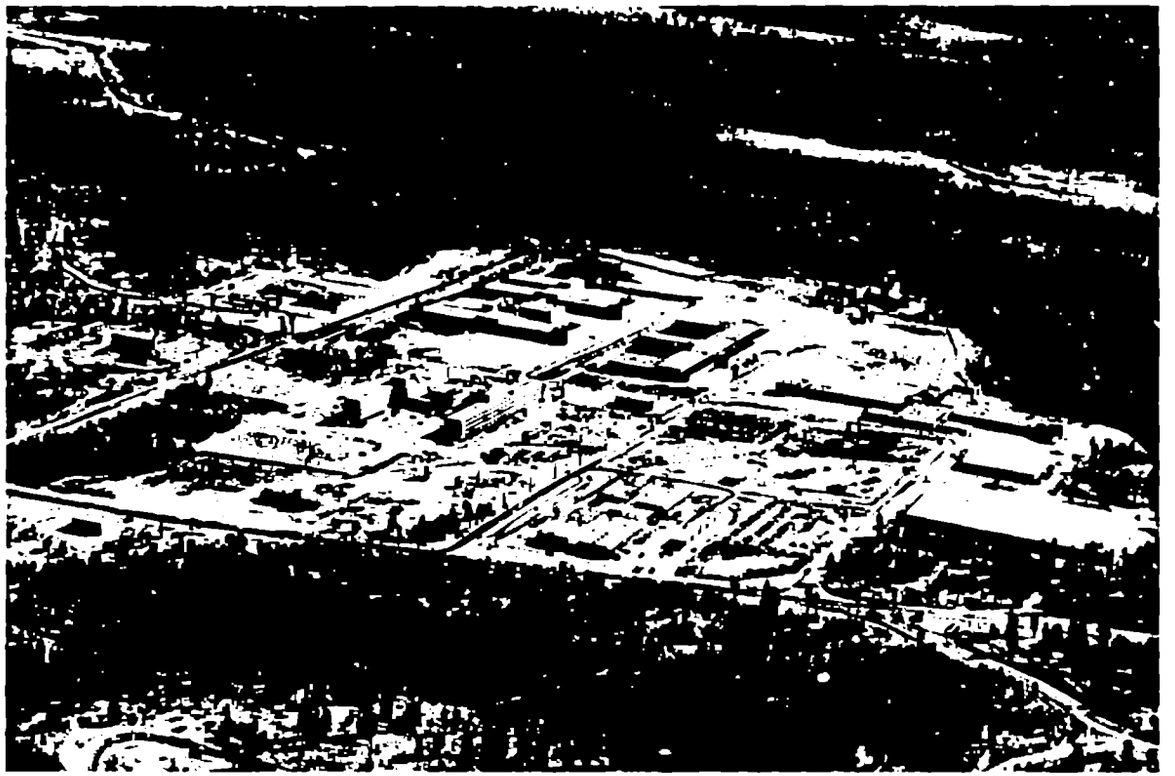

Figure 1

Figure 2

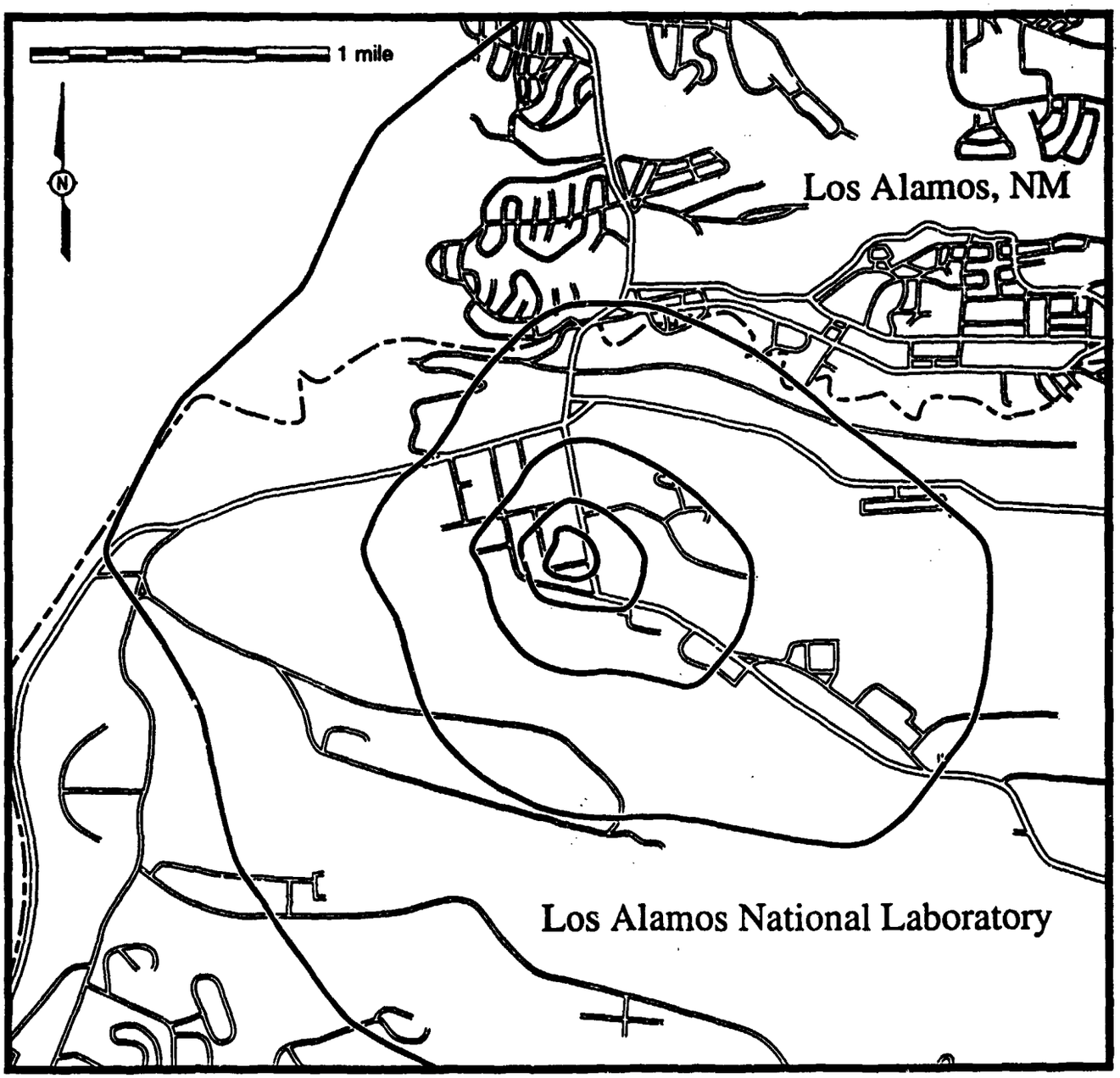

Figure 3