Review of Radiological Monitoring at LBNL

Final Report

Bernd Franke and Anthony Greenhouse

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ifeu-Institut für Energie- und Umweltforschung Heidelberg GmbH
Institute for Energy and Environmental Research, Heidelberg
Wilckensstr. 3, 69120 Heidelberg, Germany

Phone: +49-6221-47670, Fax: +49-6221-476719, Internet: http://www.ifeu.de

A personal foreword

Each project has unique challenges. The work on this project was particularly challenging because (a) IFEU made the scope of the review uniquely broad despite the limited budget available, (b) expectations were high from stakeholders, and (c) an unusually large base of data which was made available for review. In addition, many issues raised by members of the local community are complex and fascinating with respect to the level of detail.

Reading thousands of pages of documents, analyzing raw data, performing sample calculations and writing, this amounted to more hours of work than anticipated. I also tried to keep an open ear to all stakeholders, especially to the concerned citizens of Berkeley who were instrumental in calling for an independent review of radiological monitoring around NTLF. I enjoyed the possibility to attend the public meetings which allowed me to participate in spirited debate despite the fact that I live nine time zones away.

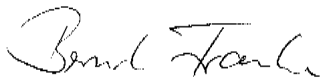
After submission of the preliminary technical report in June 2000, I received many important comments. It was not possible to give proper credit to each and to reply to all comments specifically. To do so would have exhausted resources which were reserved to review those areas of concern which were not addressed previously. All answers to the questions identified are given as to best of the author's knowledge. The report was presented at the CEAC workshop on April 2, 2001 during and after which I received additional questions and comments. I attempted to incorporate my responses in this final version of the report.

I appreciate the assistance of my colleague Anthony Greenhouse who focused on neutron dosimetry issues. Although he provided review on the other sections as well, I take the responsibility for any errors and omissions which remain.

I am well aware that it is difficult to do justice to everybody under these circumstances. There is rarely an answer to a good scientific question that does not raise more questions in the aftermath.

If IFEU's work helped in sort out between the major and minor issues in this debate with regards to radiological impacts from LBNL operations, then it was worth the extra effort.

Heidelberg, August of 2001



Bernd Franke

Executive Summary

This report describes work performed by the Institute for Energy and Environmental Research based in Heidelberg, Germany (IFEU) under contract with the City of Berkeley, CA. The work addresses concerns over past and present radiation exposures resulting from operations at the E.O. Lawrence Berkeley National Laboratory (LBNL). IFEU has reviewed data on present and past emissions and environmental monitoring by LBNL of soils, air, subsurface and ground waters, and plants (including raw data) in order to arrive at conclusions about both the quality of the data collected and the analyses of that data. Seventeen areas of concerns were identified and were divided into four groups: (a) exposures from current operations (1998 onwards), (b) legacy contamination from past operations/Superfund issues, (c) historical exposures (1997 and earlier), and (d) risk related questions. The results can be summarized as follows:

A Exposures from Current Operations (1998 onwards)

A.1 Is the tritium inventory at NTLF adequately determined?
The current inventory of tritium at NTLF is reported to be around 13,000 Ci. The potential error of that estimate, however, is greater than 20% and thus exceeds the reported airborne tritium releases. The inventory data thus does not allow verification of data on releases into the environment. However, for a variety of reasons, it is desirable to improve the accuracy of tritium inventory. With due consideration to limitations expressed above, the inventory data, in connection with other information such as the number of experiments in a given time period, and the amount of tritium in waste streams, will allow us to evaluate of those types of operation at the NTLF which can be regarded as typical. In order to verify the data provided by the new high precision thermoelectric calorimeter, an independent audit of the data is recommended.

A.2 Are the releases of airborne tritium adequately monitored?
The review indicates some uncertainties in NTLF stack data. Emissions of HTO are estimated based on silica gel data. It is possible that the silica gel samples may not catch all the water in NTLF stack effluents as data from the Los Alamos facility shows. While the potential error is likely to be small, an independent evaluation of the efficiency of NTLF stack sampling with the silica gel system would permit better resolution of these questions. It is recommended that silica gel sampling efficiency be verified by installing one or more of these alternatives (a) a series of silica gel samplers, (b) a series of ethylene glycol bubblers, or (c) molecular sieves. In addition, measures should be taken to increase the efficiency of the real-time system by upgrading the shielding and counting system.

For 1998, the silica gel data for the sum of HTO&HT was unreliable because of oxidizer malfunction and the real-time data measured with the Overhoff system was used instead. Due to large uncertainties present in the Overhoff data, this estimate is likely to be unreliable. HT releases from NTLF are only partially reported because room 107 is only monitored for HTO releases. The data regarding the accidental release of 35 Ci of HTO on July 24, 1998 remains inconclusive. It is possible that an additional 15 Ci of HT was released as well suggesting that monitoring for HT for room 107 releases would be a beneficial addition of NTLF emission monitoring. However, HT emissions are of lesser importance due to the low radiotoxicity of HT and a very slow oxidation of HT to HTO in the atmosphere.

The most significant conclusion is that real-time data shows that tritium is often released from NTLF in short events. An ongoing systematic analysis of the time function of the releases is recommended. Future atmospheric dispersion modeling of NTLF releases should take the short-term nature of the releases properly into account (e.g. by using CALPUFF along with CAP88).

A.3 Is tritium in air measured at the right locations?
The current number of sampling locations is below the de facto standard established at other DOE facilities. It is recommended that the number of sites that are monitored for tritium in ambient air be increased to cover at least all 16 wind direction sectors. This will ensure that accidental and diffuse releases that may bypass stack monitors would be detected. As of January 2001, LBNL has proposed to increase the number of ambient air monitoring stations to 14. This is a significant improvement over the current situation. It is noted that not all potentially impacted sectors are covered. It would be useful if LBNL would provide a rationale for the sampling locations.

The selection of precise sampling locations should be based on a detailed evaluation of expected tritium concentrations in air using a dispersion model capable of accounting for the complex terrain and the short-term nature of tritium releases. It is obvious that in a given wind direction sector, the monitored location will not always reflect the largest offsite concentration. There is, however, an upper limit to the ratio of (maximum offsite air concentration)/(maximum monitored air concentration). This ratio can be calculated using appropriate dispersion models. It is suggested that this information be included in the annual environmental monitoring reports.

We understand that considerations are being made to remove the present tritium stack to a new location at building 75. This will likely decrease the impact on off-site locations. In addition, a contract is being arranged with U. C. Davis to perform wind tunnel modeling of the LBNL site which theoretically would provide scientific grounds for the establishment of environmental monitoring stations. We support both of these goals.

A.4 Is the sampling and analysis of tritium in air at a given location sufficiently accurate?
Based on the data reviewed thus far, the analytical data for HTO in ambient air samples is verifiable and is subject to reasonable uncertainties. With regard to analysis of samples collected at the Lawrence Hall of Science, the annual average concentration of tritium appears to be subject to a combined uncertainty of less than 20%. It is important to ensure that the amount of water extracted from silica gel is not diluted with water loaded onto the gel prior to environmental exposure. It is recommended that information regarding the uncertainty of sampling and analytical data be incorporated in the annual environmental reports. The height of air intake has little impact on measured concentrations, the request to install real-time ambient air monitoring has little merit because it lacks the suitability to detect low ambient levels.

A.5 Are radiation exposures to individuals (including sensitive subgroups) from NTLF operations below 10 mrem/yr?
There is considerable uncertainty in the CAP88PC model which is used by LBNL to determine compliance with regulations. Measurements of ambient air are the preferred method to ensure compliance with the 10 mrem/yr dose limit. It is concluded from this review that there is no evidence to suggest that any individual received a radiation exposure resulting from NTLF emissions which exceeded 10 mrem/yr in the years 1998 and 1999. However, some uncertainty

remains because the evidence is limited. In order to reduce these uncertainties, the following activities are suggested:

(a) LBNL should use CALPUFF in addition to CAP88PC to determine compliance with the Clean Air Act, 40CFR61, Subpart H, accounting for discontinuous nature of the releases.
(b) In addition, the assessment should acknowledge the presence of transient receptors. This may best be done by using the property fence as compliance location and conservatively assuming an annual presence there of offsite personnel.
(c) Supplemental to (a) and (b), LBNL should demonstrate by dispersion calculations that the annual average air concentrations at any other offsite location are lower than those at the property fence if discontinuous releases are taken into account.
(d) Given that dose to people is the primary concern, once the stack is moved to B-75, LBNL should conduct routine bioassay for one year on staff not associated with the NTLF operation. The data should be published in the open literature.

A.6 How relevant is the presence of organically bound tritium?

Only a small fraction of the total airborne emissions was captured in trees around NTLF. The inventory in trees in the 200 meter radius around NTLF is estimated to be less than 1 Ci; the tritium inventory in groundwater is estimated to be less than 1 Ci as well. Even if the entire tritium inventory in trees and groundwater were to be released into the air via leaf transpiration, the source term would be equivalent to the amount of tritium emitted from NTLF during a few average days of NTLF operation. It is recommended to continue sampling and analysis of organically bound tritium (OBT) as well as tissue free water tritium (TFWT) in plant tissues. Tree ring analysis can provide valuable information about past exposures.

A.7 How can the levels of tritium in rainwater be explained?

The measured concentration of tritium in rainwater in 1988 of 775,000 pCi/L is in reasonable agreement with expected concentrations. While the concentration measured in 1988 was far in excess of EPA's drinking water standard of 20,000 pCi/L, it should be noted that concentrations in recent years was clearly below that level. There is no correlation between tritium concentrations measured in rainwater and the reported emissions of tritium from NTLF in days prior to rainwater sampling for the year 1994. This illustrates the absence of adequate quality control of data for that year and confirms that tritium in rainwater is not as reliable an indicator as is tritium in ambient air.

A.8 Are measurements of discharges of radionuclides other than tritium into air and water from LBNL and the resulting radiation exposures sufficiently accurate?

In 1999, there were 119 potential locations at LBNL where radioactive materials are present and could be released into the environment. Of these 23 are sampled for airborne releases. Ninety-six (96) potential sources are not monitored because potential doses are estimated by LBNL to be below 0.001 mrem/yr. 40CFR61 Subpart H requires that the documentation be verifiable with regard to compliance with the standard. Due to the limitations of the contract, it was not possible to conduct the required review to determine whether LBNL's sampling program was in compliance with regulatory requirements.

A.9 Are measurements of gamma and neutron radiation from LBNL sufficiently accurate?

Current doses from gamma and neutron radiation resulting from LBNL operations are rather small. Uncertainties are considerably less than those described for tritium.

B Legacy Contamination from Past Operations / Superfund Issues

B.1 Is LBNL's Draft Tritium Sampling and Analysis Plan sufficient to determine the extent and nature of contamination at NTLF?
The Draft Tritium Sampling and Analysis Plan sampling and analysis program should be supplemented. The ambient air monitoring should be expanded to cover all 16 wind direction sectors (of 22.5° each). The selection of precise locations should be based on a detailed evaluation of expected tritium concentrations in air using a dispersion model capable to account for the complex terrain and the short-term nature of tritium releases. The HASL-300 core method for soil sampling should be used; samples to be analyzed for additional depth increments. The issue of sampling of groundwater should be resolved in coordination with the State of California Regional Water Quality Control Board.

B.2 Which other factors need to be addressed in EPA's evaluation of the Superfund status for the NTLF site?
The EPA will score the NTLF site according to the procedures set in the Hazard Ranking System (HRS). Because of concerns in the community that operations at NTLF during the sampling time may not be representative of typical operations, an array of information should be used in determining the type of NTLF operations. This may be done by a review of product shipments and the number of tritiations performed. It is also recommended that EPA should provide information as to how the hazard ranking score would change if Lawrence Hall of Science would be regarded as a school, accounting for student population.

B.3 What is the concern regarding contamination by radionuclides other than tritium?
There is non-tritium radioactive contamination from past operations at LBNL. It is widely believed that cleanup of such areas has either been conducted or were deemed to be unnecessary due to the lack of hazard. The limited resources available in this project did not allow an independent verification of these claims.

C Historical Exposures (1997 and earlier)

C.1 What exposures resulted from neutron and gamma radiation resulted from LBNL operations?
Neutron and gamma doses at various locations at the LBNL site boundary were substantially larger than today. Based on available data, maximum exposures have exceeded 500 mrem/yr using the historical conversion factors. Using current conversion factors for neutron doses, cumulative dose rates at the Olympus Gate station were greater than 2,000 mrem. It is recommended to estimate doses to the nearest residents including the contribution of all LBNL sources and pathways while taking uncertainties in monitoring data, conversion factors and other parameters into account. A recent paper (Heimers, 1999) presents cytogenetic data that suggests that neutron radiation may have a higher relative biological effectiveness (RBE) than is reflected in currently used radiation weighting factors. This paper and other data on the RBE of neutrons should be reviewed further.

C.2 What exposures resulted from past releases of tritium?
Reported concentrations of HTO in ambient air peaked in 1978 (2,200 pCi/m^3). This value is more than a factor of 100 greater than the concentrations measured at LHS in 1999 and would

exceed the current NESHAP compliance standard of 1,500 pCi/m^3, though it did not exceed the then-prevailing limit. Reported concentrations at LHS and Olympus Gate were often equal to those reported for LHS while one would expect lower concentrations due to atmospheric dispersion. The reliability of historical data is limited. It depends on uncertainties in sampling and analysis and should be evaluated further. It is recommended to estimate doses to the nearest residents including the contribution of all LBNL sources and pathways while taking uncertainties in monitoring data, conversion factors and other parameters into account.

C.3 What exposures resulted from past releases of radionuclides other than tritium?
Reported concentrations of alpha and beta activity in air from 1963 onwards does not indicate significant impacts from releases at LBNL. Data for discharges into the sewer indicate that peak releases occurred in the 1970s. The limited review of the data indicated internal inconstancies. In a reconstruction of historical exposures, emphasis should be placed on a review of raw data. It is recommended to estimate doses to the nearest residents including the contribution of all LBNL sources and pathways while taking uncertainties in monitoring data, conversion factors and other parameters into account.

D Risk Related Questions

D.1 What is the potential health risk from past exposures?
Radiation doses from past operations at LBNL were comparable to those at locations where considerable efforts were undertaken to reconstruct exposures to members of the public. In light of uncertainties regarding the magnitude and relative biological effectiveness of neutron exposures and the contribution from other radionuclides and non-radioactive pollutants, an in-depth review is recommended. A prerequisite for the risk assessment process involves dose reconstructions for past LBNL operations.

D.2 What is the potential health risk from current exposures to tritium?
Not available at this time. IFEU will produce a report on the results of the sampling plan, including a qualitative review of the LBNL's revised health risk assessment documents incorporating the results from additional sampling.

D.3 What is the risk in case of accidents, such as fire?
The adequate determination of the consequences of potential accidents at the NTLF is of particular importance to ensure that the facility is in compliance with DOE Standard 1027-92. The Safety Analysis Document concludes on page 3: "The analysis shows that full release of the tritium inventory could not cause 'significant localized consequences'", which are defined as accidental doses at 30 m exceeding 10 rem (which equals 10,000 mrem).

The preliminary review indicates that this claim may be incorrect. Parameters in the Safety Analysis Document were selected without assessing that the resulting doses are realistic for the whole array of potential scenarios. This is evidenced by the comparison of doses calculated in the Safety Analysis Document for the worst accident (a fire at NTLF releasing 15,000 Ci of HTO) with results from alternative calculations. While the Safety Analysis Document concludes that the maximum off-site exposure is 4.8 mrem at a distance of 1,100 meters, doses would be be between 2,900 to 18,000 mrem using the "jogger scenario" from the SENES Inc. report. This assumes that the tritium is released from the stack with no plume rise from the fire; conditions which could prevail if HTO is released at the onset of a fire.

An independent evaluation of the assumptions underlying the scenarios, the calculation model and its parameters is lacking. It is therefore recommended that an independent reassessment of consequences from accidents at NTLF be performed.

Suggested follow-up

The review indicates that the data regarding monitoring of emissions and assessment of the environmental impacts of LBNL operations could only be partially verified. Further investigations are seen to be necessary to evaluate those areas which could not addressed in this review.

First, a full audit of LBNL's radiological monitoring should be conducted to provide in-depth analysis of outstanding issues. The goal of the audit should be (a) to ensure compliance of current operations with requirements in 40CFR61 Subpart H, DOE Order 5400.5 and other pertinent regulations, and (b) to identify any regulatory, technical and scientific deficiencies if they exist.

Second, an independent reassessment of consequences from potential accidents at NTLF appears necessary in light of the shortcomings identified in the Safety Analysis Report.

Third, a reconstruction of doses to members of the public from historical LBNL operations exposures would be beneficial to identify the magnitude of exposures and the associated health risk. Particular emphasis should focus on the uncertainty associated with historical monitoring data.

All activities should be carried out by parties independent of LBNL. Public participation in the process should be ensured.

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1 Introduction

The E.O. Lawrence Berkeley National Laboratory (LBNL) is a multipurpose research facility located in the Berkeley/Oakland hills in Alameda County, California. It is operated by the University of California (UC) under contract with the U.S. Department of Energy (DOE). The site map of the 200 acre facility is shown in Figure 1.

In response to community concerns regarding radiation exposures from LBNL, the City of Berkeley contracted with the Institute for Energy and Environmental Research (IFEU)[1], based in Heidelberg, Germany to review radiological monitoring at Lawrence Berkeley National Laboratory. The concerns cover a wide range of issues, including past and present operations of the laboratory. The scope of work spelled out in the contract is attached in Appendix A.

The review process started with interviews of LBNL scientists and technicians, subcontractors, regulators, city officials, and representatives of the community to generate a list of concerns for evaluation. This took place during the week of February 28, 2000.

The authors also participated in several meetings of the Environmental Sampling Project Task Force meetings, either in person or via teleconferencing. There were many communications via e-mail, telephone and mail between IFEU and the City of Berkeley, community representatives, LBNL personnel and contractors, as well as with staff of EPA's San Francisco Office. The authors wish to acknowledge the substantial documented input received from the Committee to Minimize Toxic Waste (CMTW) which helped focus the review. We also appreciate the willingness of LBNL to respond to many information requests through its contractor Dr. Owen Hoffman (SENES Oak Ridge, Inc.).

In the process, the authors obtained about 20,000 pages of documents for review. In addition, raw data relating to real-time monitoring of tritium releases from the NTLF stack was received. It is in the nature of a review process that additional questions always arise. Consequently, more material is expected for consideration over the next few months. The present report is a progress report. It presents a partial analysis of data evaluated thus far as a basis for further discussion. The work can by no means to be considered complete. Thus it is hoped that it may assist in clarifying some issues, and to prioritize areas of concern. The starting point is a list of concerns, which is presented in question format. The report attempts to consider all questions and outlines where work still needs to be done. It is in the nature of endeavors like this, that each question answered could give rise to a new one.

The reader should be aware of the fact that although the review attempts to cover a large array of questions, the finite resources of IFEU's consulting contract do not allow addressing every question in appropriate depth.

[1] The official legal German name of the institute is "ifeu-Institut für Energie- und Umweltforschung Heidelberg GmbH". It is not part of the Institute for Energy and Environmental Research (IEER) in Takoma Park, MD where the principal author of this report was Executive Director from 1987 to 1998. Founded in 1978, IFEU is a non-profit research organization and has currently a staff of 30 scientists. More details about research areas and activities can be found on the web site (www.ifeu.de).

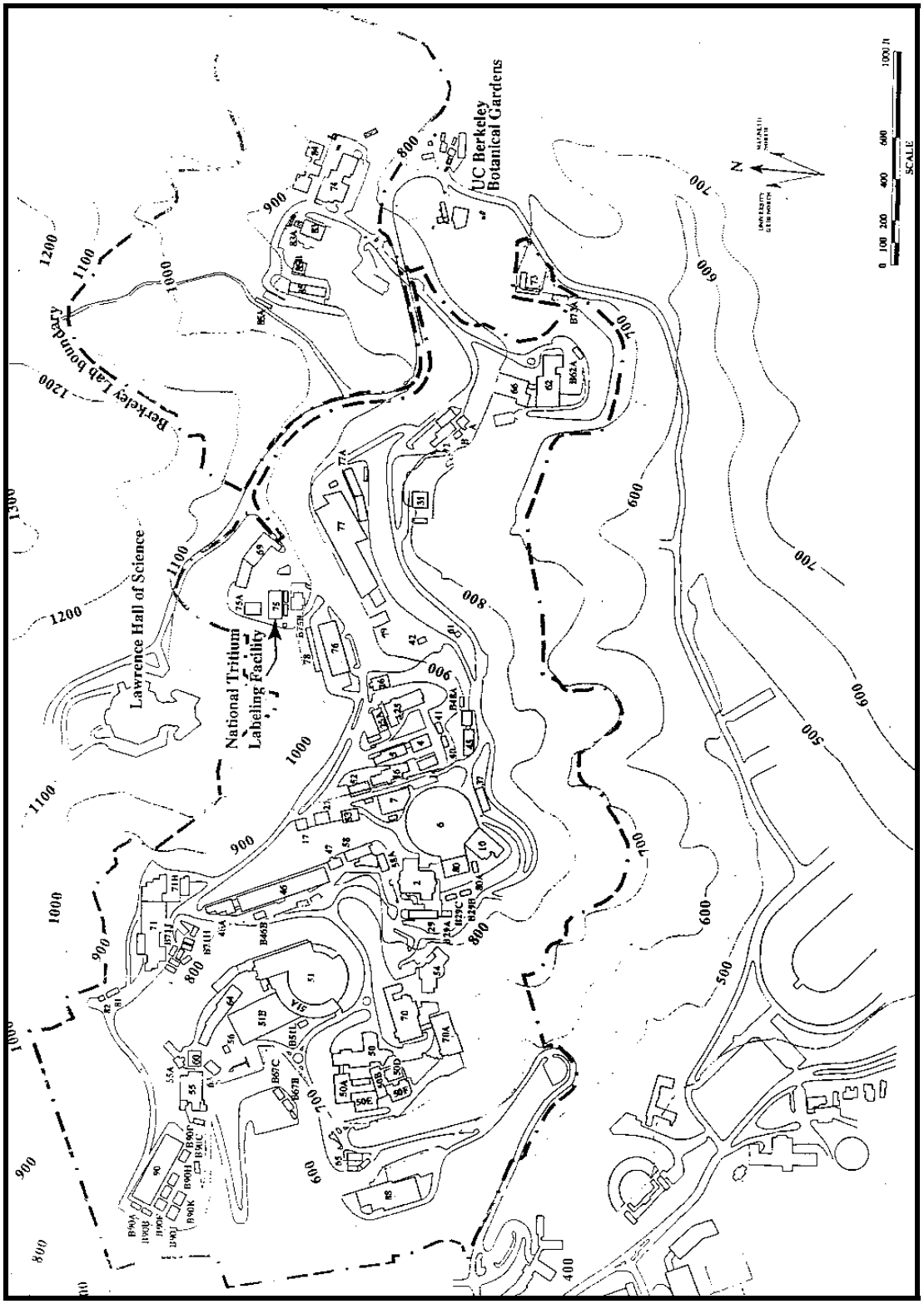

Figure 1. Site map of Lawrence Berkeley National Laboratory (from LBNL/DOE, 1999)

2 Identification of areas of concern

The major concerns of citizens regarding radiation exposures from LBNL operations which were broadly divided into four main categories for the purposes of this review:

A Concerns about current operations
B Concerns about legacy contamination from past operations
C Concerns about historical exposures
D Risk-related concerns

The authors attempted to approach the concerns as completely as possible. The presentation in question format is thought to allow the non-technical reader following the review process. Each chapter is organized in the following format:

- Concern
- Approach
- Findings
- Conclusions and recommendations

Table 1 and 2 contains a list of concerns that were identified and reviewed. While some deal with rather specific technical issues, others cover larger problem areas.

Many questions currently focus on the activities at the National Tritium Labeling Facility (NTLF), in particular in connection with the Superfund evaluation by the US Environmental Protection Agency. In response to these activities, this report contains a section dealing with the LBNL/DOE Tritium Sampling and Analysis Plan, dated May 1999 (concern B.1 and B.2).

The evaluation of current operations is based on data for releases and environmental concentrations in the years 1998 and 1999. Data for earlier years are considered historical due to major changes in sampling and analytical techniques, quality control and documentation.

Table 1. Preliminary list of concerns relative to radiation exposures from current operations at LBNL

Concern	Approach	Status
A. Exposures from Current Operations (1998 onwards)		
A.1 Is the tritium inventory at NTLF adequately determined?	Review inventory data, its accuracy, and relevance to determine the environmental impacts of NTLF	Completed
A.2 Are the releases of airborne tritium adequately monitored?	Review data on stack releases; evaluate internal consistency and uncertainties	Completed
A.3 Is tritium in air measured at the right locations?	Compare potentially affected locations with locations actually sampled	Completed
A.4 Is the sampling and analysis of tritium in air at a given location sufficiently accurate?	Review observed versus expected water collected in silica gel samples Review results of split sampling program Review of contract laboratory performance	Completed
A.5 Are radiation exposures to individuals (including sensitive subgroups) from NTLF operations below 10 mrem/yr?	Review NESHAP compliance assessment; determine exposure scenarios that are not covered	Completed
A.6 How relevant is the presence of organically bound tritium?	Review of data for organically bound tritium	Completed
A.7 How can the levels of tritium in rainwater be explained?	Review of data for tritium in rainwater	Completed
A.8 Are measurements of discharges of radionuclides other than tritium into air and water from LBNL and the resulting radiation exposures sufficiently accurate?	Review of monitoring data regarding radionuclides other than tritium	Completed
A.9 Are measurements of gamma and neutron radiation from LBNL sufficiently accurate?	Review environmental monitoring of gamma and neutron radiation	Completed

Table 2. Preliminary list of concerns relative to legacy contamination from past operations / Superfund Issues, historical exposures and risk related questions

Concern	Approach	Status
B. Legacy Contamination from Past Operations / Superfund Issues		
B.1 Is LBNL's Draft Tritium Sampling and Analysis Plan sufficient to determine the extent and nature of contamination at NTLF?	Review of sampling plan regarding sampling media, sampling locations, analytical techniques, and QA/QC issues	Completed
B.2 Which other factors need to be addressed in EPA's evaluation of the Superfund status for the NTLF site?	Review whether NTLF operations will be typical during sampling period; review of non-radiological data (e.g. number of affected residents)	Completed
B.3 What is the concer regarding contamination by radionuclides other than tritium?	Review of non-tritium radioactive contamination at LBNL	Completed
C. Historical Exposures (1997 and earlier)		
C.1 What exposures to neutron and gamma radiation resulted from LBNL operations?	Review of historical data on neutron and gamma exposures	Completed
C.2 What exposures resulted from past releases of tritium?	Review of historical data on tritium emission and environmental concentrations	Completed
C.3 What exposures resulted from past releases of radionuclides other than tritium?	Review of historical data on emissions and environmental concentrations	Completed
D. Risk Related Questions		
D.1 What is the potential health risk from past exposures?	Comparison of historical doses with doses at other sites	Completed
D.2 What is the potential health risk from current exposures to tritium?	Review of updated health Risk Assessment (expected for late 2001)	Document not available
D.3 What is the risk in case of accidents, such as fire?	Review of LBNL Safety Analysis Document for NTLF	Completed

A Exposures from current operations (1998 onwards)

A.1 Is the tritium inventory at NTLF adequately determined?

Approach
Review inventory data, its accuracy, and relevance to determine the environmental impacts of NTLF.

Findings
LBNL is conducting a tritium inventory using DOE's Nuclear Materials Monitoring System (NMMSS). The purpose of this system is to provide accountability of LBNL's use of tritium. The accuracy of the data, however, is very limited for a variety of reasons:

- All numbers are rounded to 0.01 g of tritium, or 96 Ci
- The reporting threshold is 0.005 g of tritium, or 48 Ci
- LBNL's estimate of tritium inventory at NTLF is associated with an uncertainty of greater than 20%

Figure 2 shows the NMMSS inventory data along with reported airborne emissions of tritium from LBNL. To allow visual comparison, the reported releases were plotted at midpoint of the respective year. The airborne releases are two to three orders of magnitude lower than the reported NMMSS inventory. There is no clear correlation between the two numbers. During the 1990s, the reported releases of tritium decreased even though the reported inventory did not change much.

It is evident that the NMSS data is not a reliable source to estimate releases of tritium into the air. It is not designed or required to determine compliance of NTLF operations with Rad-NESHAP. Direct measurements at the point of releases is the preferred method to determine the quantity of releases.

What then is the use of the inventory data? It is the stated purpose of DOE's tracking system NMMSS that it is an important tool that monitors accounting and reporting under the Atomic Energy Act of 1954 and the safeguards agreement between the U.S. and the International Atomic Energy Agency (IAEA). While the amount of tritium contained in nuclear warheads is still classified, estimates of the US tritium stockpile are in the order of 75 kg. The reporting threshold of 0.005 g reflects that NMSS is set to track the significant amounts of tritium only. Simply put, airborne tritium emissions from NTLF are just too small to be of major importance for the purpose of NMMSS.

There are reasons besides NMMSS to improve the precision of the tritium inventory at NTLF: providing a reliable baseline for risk management, control over potentially hazardous conditions, maintaining control over costs and increasing the credibility with other stakeholders. It is for these reasons that LBNL has purchased an high precision thermoelectric calorimeter which allows to measure 10,000 Ci of tritium with an uncertainty of 1%. It is hoped that the data provided by this instrument will give a better estimate of the current inventory and help resolve lingering questions about claims made by LBNL about the fraction of the inventory shipped as waste, shipped as product and remaining at the site. This information, along with data on the

number of tritiations in a given time period, will allow a determination of whether the type of operation at NTLF can be regarded as typical. It is evident that the releases and also the risk of accidents will increase with the number of tritiations of a similar kind, all other conditions remaining the same. IFEU has neither received nor requested information about the number of planned tritiations for the years to come. It is possible that releases of tritium into the air will increase in the future. The magnitude of such releases cannot be reliably predicted but has to be measured.

Conclusions and recommendations

The current inventory of tritium at NTLF is reported to be around 13,000 Ci. The potential error of that estimate, however, is greater than 20% and thus exceeds the reported airborne tritium releases. The inventory data thus does not allow verification of data on releases into the environment. However, for a variety of reasons, it is desirable to improve the accuracy of tritium inventory. With due consideration to limitations expressed above, the inventory data, in connection with other information such as the number of experiments in a given time period, and the amount of tritium in waste streams, will allow us to evaluate of those types of operation at the NTLF which can be regarded as typical. In order to verify the data provided by the new high precision thermoelectric calorimeter, an independent audit of the data is recommended.

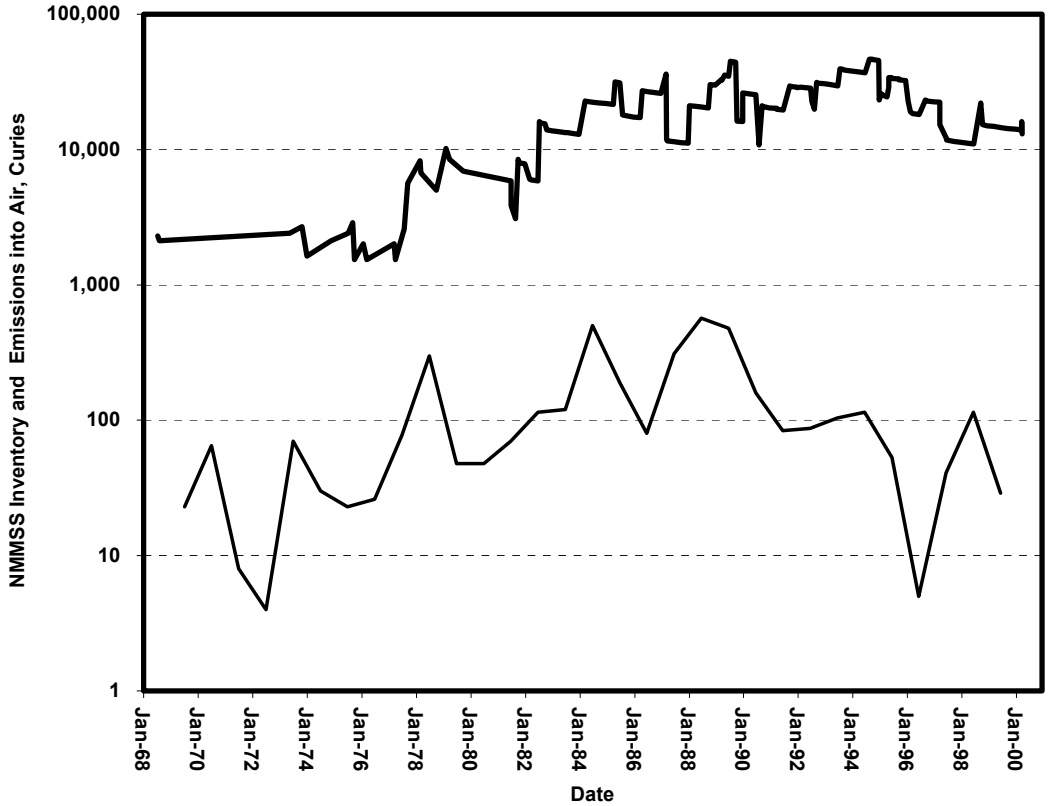

Figure 2. Reported tritium inventory at NTLF (top line) in relation to reported airborne releases of tritium into the air (bottom line)

A.2 Are the releases of airborne tritium adequately monitored?

Approach
Review data on stack releases; evaluate internal consistency and uncertainties

Findings
At NTLF, two independent stack-monitoring systems are in operation. Cumulative weekly releases of HTO and HTO&HT are sampled with a silica gel system. In addition, a real-time system (Overhoff) provides emission data integrated over 100 seconds.

The **internal consistency of silica gel sampling data** can be evaluated by the comparison of data for HTO and HTO&HT since concentrations of HTO should be smaller then those of HTO&HT. Since the end of February, 1998, two separate collection systems exist for NTLF stack emissions. LBNL indicates the uncertainty due to sampling and analysis errors to be ~20%, thus the ratio of (HTO&HT)/HTO should be below 1.2. Figure 3 shows the results of the comparison.

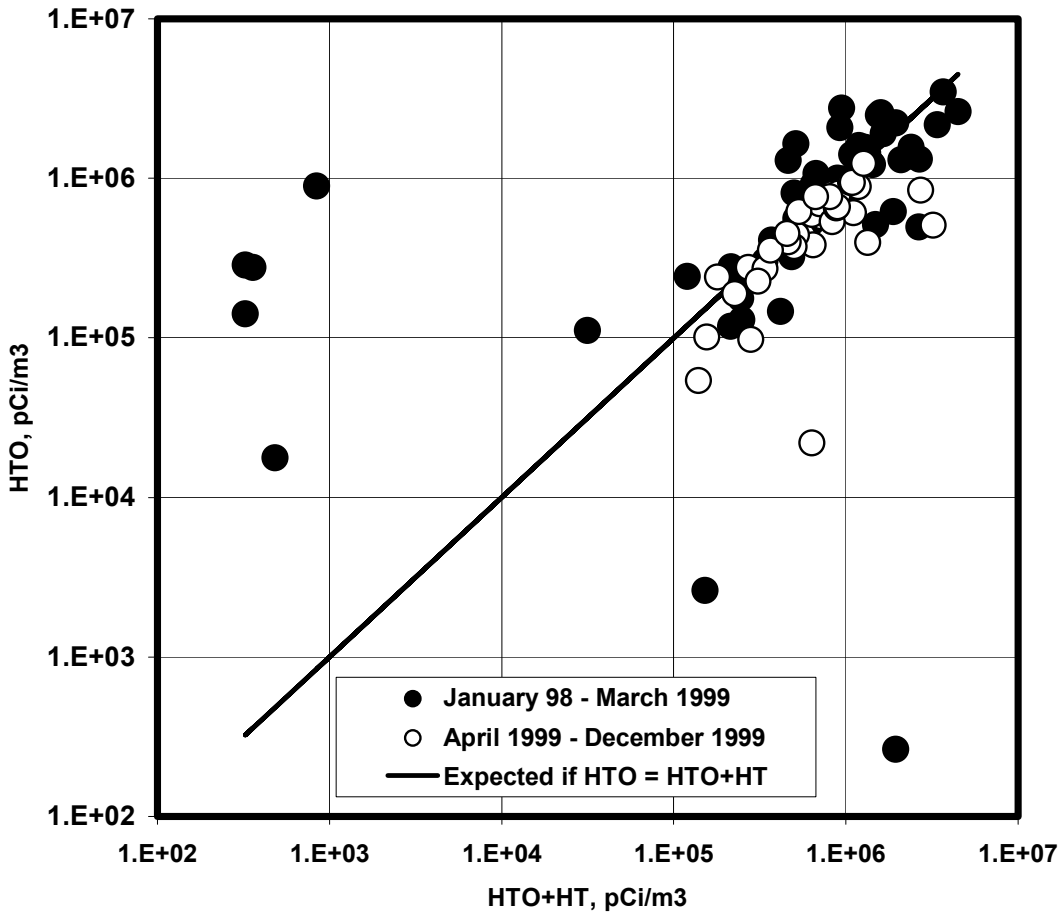

Figure 3. Comparison of concentrations of HTO and HTO&HT determined from weekly silica gel sampling of the NTLF stack

A new oxidizer was installed in March 1998; therefore data before and after this date should be reviewed separately. It is evident that the old oxidizer operating at room temperature was unreliable because for 18 out of 57 samples (32% of the total) the ratio of (HTO&HT)/HTO of greater than 1.2. After April 1999, this number dropped to 1 out of 32 samples (3% of the total). When questioned about this, LBNL gave the following explanation: "The oxidizer was initially operated at ambient temperature based on manufacturer's specifications. Some of the air moisture (HTO) was captured (either from condensation or from surface retention) by the oxidizer, which gave a lower tritium concentration value than the HTO column during certain weeks. Since this is a closed system, the captured HTO moisture was eventually being re-evaporated back to the system, which then contributed to a relatively higher reading on subsequent weeks. In fact, we did observe a small amount of moisture captured by the oxidizer cylinder during some of our maintenance activities. Thus, the oxidizer initially behaved like a "delay circuit" which temporally shifted the weekly results." This explanation appears to be reasonable.

Despite of the above, the question remains about **reliability of reported concentrations measured by silica gel sampling system for airborne releases**. Sample media for tritium (silica gel, ethylene glycol, molecular sieves) may not collect all the water in the air passing though; hence there is a chance that not all the tritium may be collected. This can be illustrated with data from Los Alamos National Laboratory (LANL) facility TA-33. At LANL, stacks are sampled for tritium using a bubbler system with three consecutive vials which contain ethylene gylocol. HTO which is not collected in the first vial is collected in the second vial, HTO passing through the second vial is collected in the third vial. HT emissions are measured at LANL by passing the air from vial #3 through an oxidizer, collecting the converted HTO in another set of three vials. Out of 102 samples at TA-33 in 1999, 88% of the sampled HTO activity was contained in vial #1 (90% confidence interval: 78% to 96%); the remainder was contained in vials #2 and #3. This suggests either a carryover between vials or the fact that not all humidity is captured in the first vial, hence the need for three consecutive vials. For the estimate of total HTO releases, the total activity of all three vials is counted.

From the data provided by LBNL, the efficiency of the silica gel sampling system installed at NTLF could not be determined. Such determination would be easy by installing another set of silica gel samplers after the first one. Alternatively, accuracy of emission data could be verified using ethylene glycol bubblers such as the ones used at LANL or molecular sieves.

This matter has limited impact on calculated doses. Room 107 emissions are sampled for HTO only but emissions from the NTLF main stack are assumed to be HTO even though they actually represent a mixture of HT and HTO.

The question **whether effluent air concentrations were accurately calculated** was verified by a cross check of randomly selected data for several weeks in 1998. The results are shown in Table 3. The calculated concentrations are in good agreement with the values entered in a spreadsheet received from LBNL, differences are due to rounding.

Table 3. Cross check of calculated concentrations of tritium in effluent air for randomly selected set of samples in 1998

Sampling week ending	Sampling location	Sample volume, cm^3	Sample ID	Sample activity, pCi	Calculated pCi/m^3	LBL spread-sheet, pCi/m^3
Feb 18, 1998	75-107H	1.011.685	23588	301.000	297.523	298.000
	75-NTLF-HT	1.009.780	23589	158.000	156.470	156.000
	75-NTLF-HTO	1.009.780	23590	1.390.000	1.376.537	1.3800.000
March 4, 1998	75-107H	1.146.228	23698	365.000	318.436	318.000
	75-NTLF-Total T	864.000	23699	1.370.000	1.585.648	1.590.000
	75-NTLF-HTO	864.000	23700	2.230.000	2.581.019	2.580.000
July 1, 1998	75-107H	1.007.461	24724	768.000	762.312	762.000
	75-NTLF-Total T	993.149	24728	3.640.000	3.665.110	3.670.000
	75-NTLF-HTO	1.003.940	24729	3.490.000	3.476.303	3.480.000
July 8, 1998	75-107H	1.009.138	24813	548.000	543.038	543.000
	75-NTLF-Total T	989.271	24817	1.460.000	1.475.834	1.480.000
	75-NTLF-HTO	1.003.165	24818	515.000	513.375	513.000

Because the 1998 data for HTO&HT was unreliable, the tritium source term for 1998 in LBNL's NESHAP report was based on silica gel data only with regard to HTO; releases of HT were estimated based on **results from the real-time monitoring system**. The NTLF real time monitoring system for the Hillside Stack was purchased from the Overhoff Technology Corporation. The model number was 93-WR-T-HTO. The original system has been modified to obtain improved performance and reliability. It consists of two parallel sets of ionization chamber detectors, one for total tritium (HT+HTO), and the other one measuring HT only after the HTO in air is removed by a desiccant. The emissions of HTO are calculated by subtracting the concentration of HT from HTO+HT.

There are two systematic problems with the comparison of data from silica gel sampling and real-time data for. First, the Overhoff system did not continuously operate for the entire time period in the years of 1998 and 1999 due to system malfunctions; the data thus represents only partial information of releases. Second, the real-time data is subject to a high instrument background and electrical spikes, which could be falsely interpreted as signals of releases. The instrument background reported by LBNL is 0.22 Mq/m^3, equivalent to a release of about 4 Ci per week if no background were to be subtracted. Inspection of the real-time data suggests significant fluctuation of the instrument background due to changes in temperature and other factors (see Figure 4). For March 1998, instrument background appears to have been significantly lower than 0.22 Mq/m^3, emissions calculated on the default background are underestimated.

Bearing these limitations in mind, the 1998 and 1999 data obtained from LBNL was on a weekly basis and plotted in Figure 5. The apparent discrepancy between releases based on silica gel data and integrated releases based on real-time data after nominal instrument background subtraction does not allow an independent verification of silica gel data with real-time data. Even though it may appear that for a number of weeks real-time data suggests larger HTO releases than silica gel data, this finding may well represent an artifact.

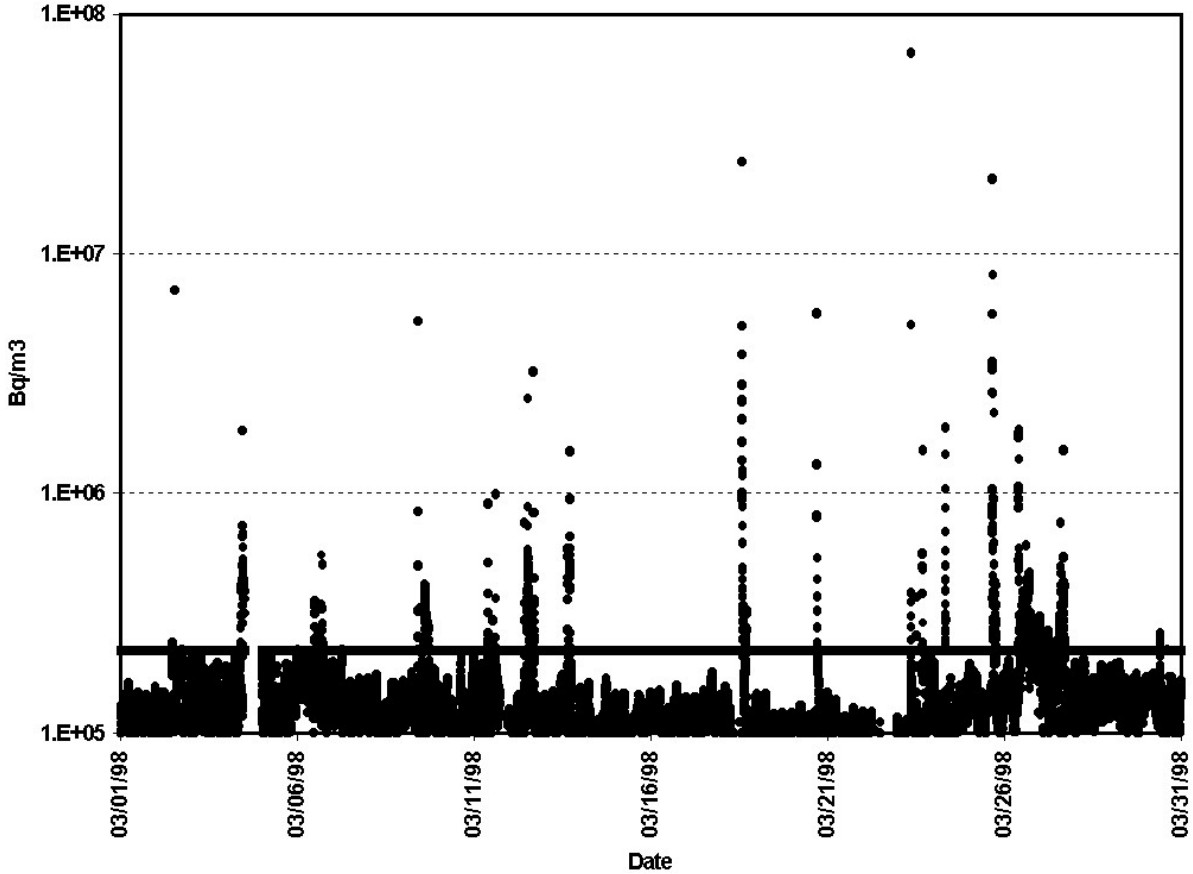

Figure 4. Real-time Overhoff monitoring of HTO releases from NTLF stack, March 1998. The reported instrument background of 220,000 Bq/m^3 is indicated by a horizontal line. Visual inspection suggests that actual background may have been lower.

Fluctuations of the average weekly instrument background could be an explanation of the discrepancies evident in Figure 5. The comparison suggests that real-time data is not a reliable indicator of the quantitative releases from NTLF stacks. Thus, the 1998 releases of HT from NTLF stack, which was estimated on the basis of Overhoff data to be 21.4 Ci is subject to significant uncertainties because of dubious quality of the underlying data. The authors were unable to verify this estimate based on the data provided; it is possible that actual releases were smaller or larger than 21.4 Ci. One should bear in mind, however, that for dose assessments, all the tritium sampled, regardless of chemical form, is assumed to be HTO. This assumption is conservative, hence the exact amount of HT released is of minor importance.

It is obvious that the Overhoff real-time is of limited value when it comes to verify data based on the more sensitive discontinuous silica gel sampling. One should bear in mind that reported emissions are based on the latter; the purpose of the former is mainly to provide rapid information which allows the laboratory to access timely information on episodic process emissions.

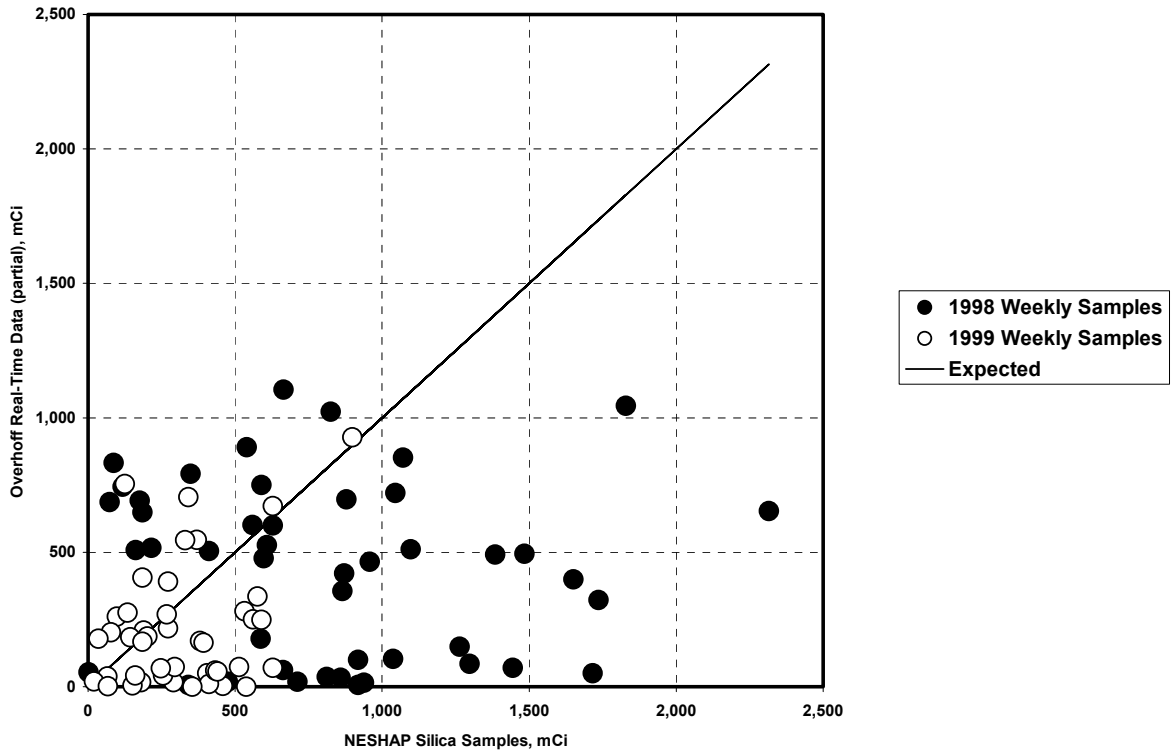

Figure 5. Comparison of weekly releases of HTO determined from weekly silica gel samples and Overhoff real-time measurements at the NTLF stack

With these considerations in mind, two areas where such information is relevant were investigated: (a) the frequency of discontinuous releases in general, and (b) the July 24, 1998 unplanned release in room 107 in particular.[2]

A major part of **tritium at NTLF is released over relatively short time periods** of less than an hour. This is shown in Figure 4 for the month of March 1998, in Figure 6 for the week February 22 to March 1, 1999 and in Figure 7 for March 25, 1998. On March 25, 1998, a total of 0.29 Ci of HTO was released based on Overhoff data. Of this, 0.2 Ci or 69% was released over a short time period of 1000 seconds (about 17 minutes) with a peak at 3:45 pm. Inspection of real-time data for other days in the years 1997-1999 suggests that similar patterns are not uncommon for other time periods as well.

The real-time data can also be used to shed a light on **the accidental release of July 24, 1998** where a silica gel sample containing tritium was being heated in a kiln in room 107. The sample activity was erroneously assumed to be low; heating released the activity. The room 107 fume hood is vented by a small stack on the roof which is sampled for HTO only using silica gel. Based on silica gel samples for the data in question, the release was estimated to be 35 Ci of HTO. While the majority of the activity was likely to be releases via the building 107 roof vent, a

[2] This evaluation was specifically introduced based on comments submitted by Dr. Roger Byrne on December 3, 2000.

smaller part of that activity appears to have been vented via the hill stack as indicated in the data from the Overhoff system shown in Figure 8.

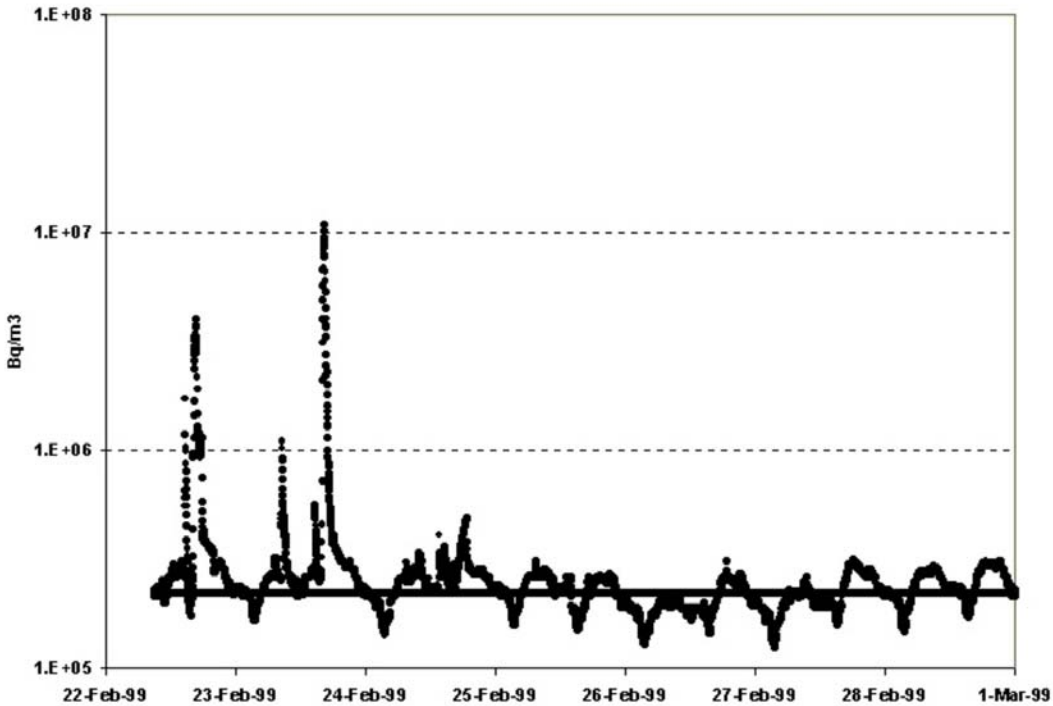

Figure 6. Real-time Overhoff monitoring of HTO releases from NTLF stack, February 22 to March 1, 1999. The reported instrument background of 220,000 Bq/m^3 is indicated by a horizontal line.

The timing of the release (start shortly before noon, major release over about 3 hours) suggests that the reported concentrations from the hillside stack are indeed the result of the accident. It is interesting to note that concentrations of HT+HTO were larger than those for HTO alone, whereas from the accident description, one would expect that all activity would be in the form HTO. The cumulative HTO release in the time from 11:30 am to 4:00 pm was 0.14 Ci, the HT+HTO release was 0.2 Ci; background was subtracted in both cases). The difference is in large part due to the sharp drop of HTO concentrations down to zero around 2:14 pm, rendering the data difficult to interpret. The difference between the expected HTO/(HTO+HT) ratio of 1 and the observed ratio of 0.7 may be (a) an artifact due to malfunctioning of the HTO detector, (b) the result of HT releases in the accident, (c) the result of emissions from the NTLF main facility, or a combination of these factors.

Assuming that (b) is the correct explanation, one would estimate the total tritium release from the accident to be 50 Ci, of which 70% would be in the form of HTO. Even if an additional 15 Ci of HT were emitted during the July 24, 1998 accident, the resulting doses would be of little consequence due to the far lower toxicity of HT compared to HTO. The example illustrates, however, that only part of NTLF emissions are reported; HT emissions from building 107 will not be recorded by the current silica gel system.

One question which was raised in this context is the appropriateness that HT is oxidized to HTO which more radiotoxic. This is done to facilitate to capture HTO in the facility. Emission data for 1999 indicates that not all HT is oxidized: about one third of the total tritium activity released from the hillside stack was HT. Due to much lower radiotoxicity of HT and a very slow oxidation of HT to HTO in the atmosphere, the HT source term is of minor importance. With regard to stack releases, oxidizing HT in the samples air (less than one part in 100,000) of the effluent stream would result in negligible releases even if all oxidized HT would be released back into the air.

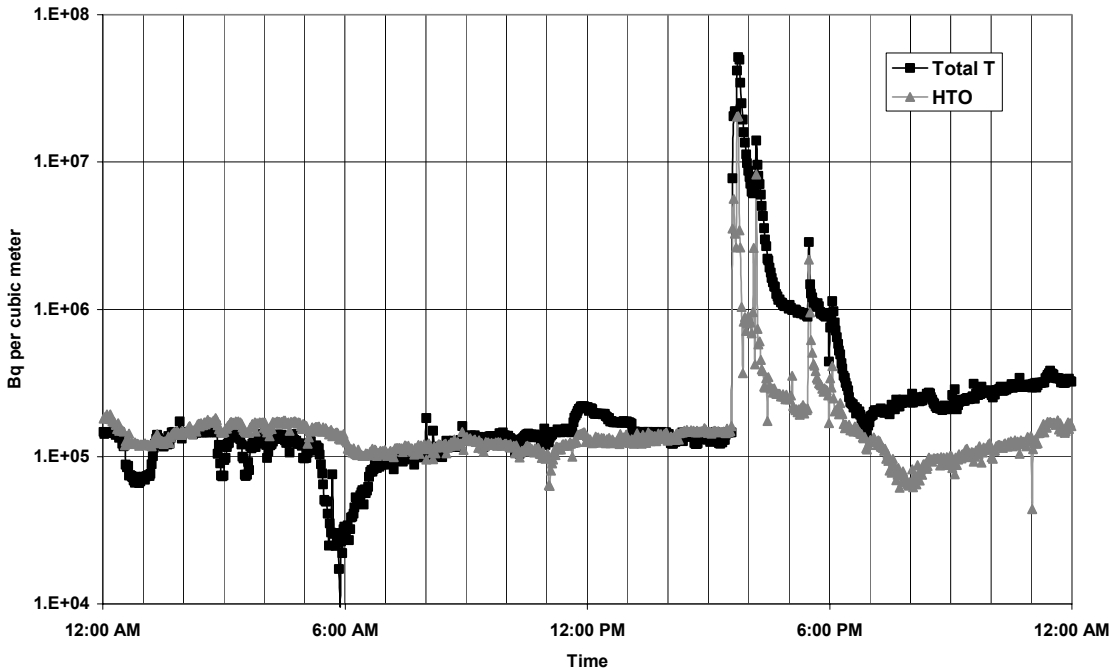

Figure 7. Real-time concentrations of HTO and total tritium in NTLF stack releases on March 25, 1998 (instrument background not subtracted)

Conclusions and recommendations

The review indicates some uncertainties in NTLF stack data. Emissions of HTO are estimated based on silica gel data. It is possible that the silica gel samples may not catch all the water in NTLF stack effluents as data from the Los Alamos facility shows. While the potential error is likely to be small, an independent evaluation of the efficiency of NTLF stack sampling with the silica gel system would permit better resolution of these questions. It is recommended that silica gel sampling efficiency be verified by installing one or more of these alternatives (a) a series of silica gel samplers, (b) a series of ethylene glycol bubblers, or (c) molecular sieves. In addition, measures should be taken to increase the efficiency of the real-time system by upgrading the shielding and counting system.

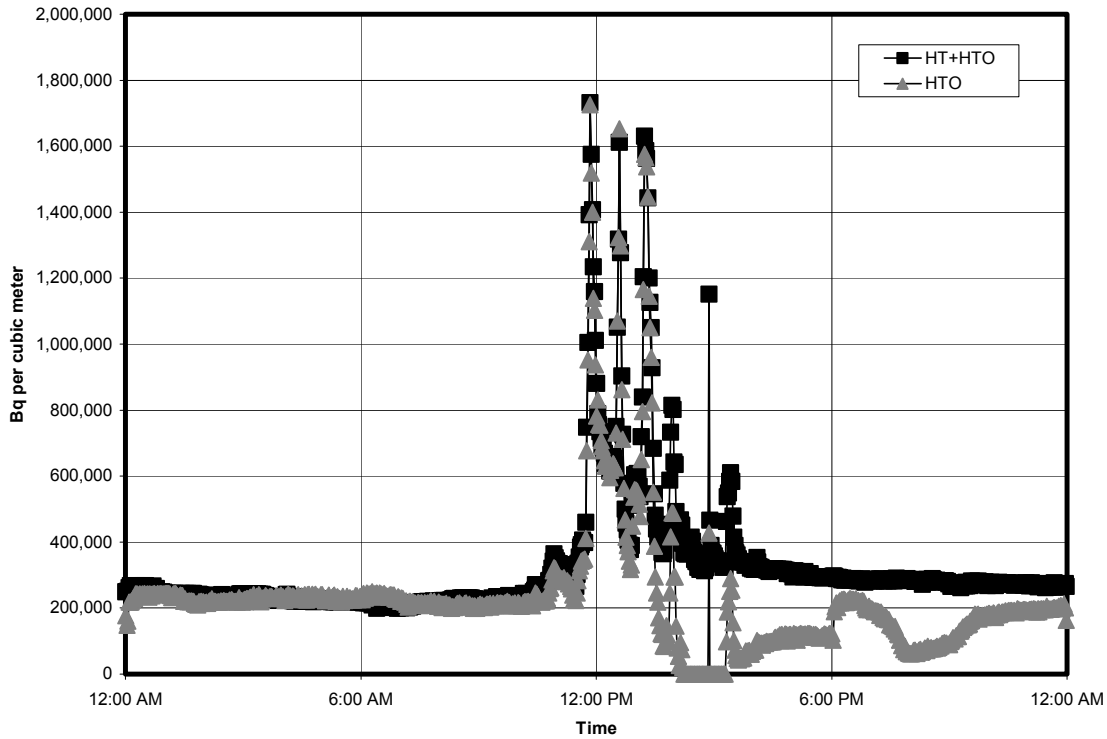

Figure 8. Real-time concentrations of HTO and total tritium in NTLF stack releases on July 24, 1998 (instrument background not subtracted)

For 1998, the silica gel data for the sum of HTO&HT was unreliable because of oxidizer malfunction and the real-time data measured with the Overhoff system was used instead. Due to large uncertainties present in the Overhoff data, this estimate is likely to be unreliable. HT releases from NTLF are only partially reported because room 107 is only monitored for HTO releases. The data regarding the accidental release of 35 Ci of HTO on July 24, 1998 remains inconclusive. It is possible that an additional 15 Ci of HT was released as well suggesting that monitoring for HT for room 107 releases would be a beneficial addition of NTLF emission monitoring. However, HT emissions are of lesser importance due to the low radiotoxicity of HT and a very slow oxidation of HT to HTO in the atmosphere.

The most significant conclusion is that real-time data shows that tritium is often released from NTLF in short events. An ongoing systematic analysis of the time function of the releases is recommended. Future atmospheric dispersion modeling of NTLF releases should take the short-term nature of the releases properly into account (e.g. by using CALPUFF along with CAP88).

A.3 Is tritium in air measured at the right locations?

Approach
Compare potentially affected locations with locations actually sampled

Findings
During 1999, tritium in air was monitored at six monitoring sites. Three of the sites are on Berkeley Lab property, and three are on the adjacent UC property. With the beginning of January 11, 2000, an additional station (ENV-75 EG) is sampled at 21 meters distance from the stack near the fence in the general direction of LHS. In addition, two additional locations will be sampled once the Tritium Sampling Plan becomes effective, the UC Botanical Garden and the Summit Reservoir, the latter one serves as a background station. The rationale for locating air-sampling stations takes into account the predominant wind direction. In addition, the station at LHS allows verifying calculated doses for the current NESHAP compliance location.

One can evaluate the sampling network at LBNL by comparing it to the monitoring systems at other DOE facilities. Table 4 contains the summary data for LBNL as well as for five other facilities for which 1998 tritium emissions were reported to be between 0.054 Ci (Pantex) and 82,700 Ci (Savannah River Site). The number of ambient air monitoring stations for tritium of non-LBNL facilities ranged from 17 to 52. The reported dose to the maximally exposed individual (MEI) which reflects, among other things, the distance to the source of the release, varies greatly between the facilities. Despite the large release at the Savannah River Site, the calculated MEI dose is lower than for LBNL because of the distance between the source and the fence (greater than 10 miles). Based on the data in Table 4, the LBNL site has the smallest number of sampling locations for tritium in ambient air even though the distance from the source of tritium emissions to MEI is nowhere as close as it is at LBNL.

Table 4. Ambient air monitoring for tritium at DOE facilities

Facility	1998 Tritium Release (Ci)[a)]	Number of ambient air stations[b)]	1998 calculated dose to facility MEI from all radionuclides and sources combined
E.O. Lawrence Berkeley National Laboratory	115	6 (+3)[c)]	0.28 mrem
Los Alamos National Laboratoy	818	52	1.1 mrem
Brookhaven National Laboratory	39.5	22	0.21 mrem
Savannah River Site	82,700	17	0.08 mrem
Pantex	0.054	27	0.005 mrem
Lawrence Livermore National Laboratory	110	20	0.055 mrem

a) Based on Site Environmental Reports for the respective facility
b) according to Baumann (2000)
c) as of 2000, the number may total 14 if LBNL's January 2001 plan is realized (see text)

Visual inspection of the sites suggests that the sampling network at most facilities covers all 16 wind direction sectors. It thus appears reasonable to adopt a similar design for LBNL as well.

The suggestion of 16 wind direction sectors is not arbitrary. The Rad-NESHAP compliance program CAP88 relies on data which is averaged over 16 wind directions. Hence, tritium should be monitored in a suitable location in each of these affected sectors. The need to sample in more than the most likely wind direction is also reflected in the Federal Facility Compliance Agreement (FFCA) regarding the Los Alamos National Laboratory.[3]

Why not just sample in the most affected wind direction? Since tritium releases from NTLF occur over comparatively short time periods, these emissions may be dispersed in directions other than what would be expected from the average distribution of wind directions. This makes the precise selection of appropriate stations complicated. If, say, 1 Ci of tritium would be released during a time period of a few minutes, there is a significant chance that the current network of monitoring stations may not properly detect it. If a person happens to be close to the fence during such an event in downwind direction, he or she could receive a dose, which could be larger than the one calculated with CAP88PC for Lawrence Hall of Science.

The main rationale for sampler siting should be to provide an independent verification of the claim that ambient air concentrations at unrestricted areas around NTLF are in compliance with Rad-NESHAP. This is best done by covering all wind directions. As of January 2001, LBNL has proposed to increase the number of ambient air monitoring stations to 14 (see Figure 9). This is a significant improvement compared to the current situation. It is noted that not all potentially impacted sectors are covered. It would therefore be useful if LBNL could provide the rationale for the sampling locations shown in Figure 9. If we focus on the most affected sector (wind blowing toward NW), is the sampling station at the Lawrence Hall of Science LHS itself appropriately located? It has been criticised that the sampler air intake it is 3.5 meters above ground level. It is below the tree level and the sampler is thus sheltered from prevailing winds. On the basis of dispersion modeling accounting for complex terrain and by comparing results of sampling stations LHS and 75-EG (see chapter A.5) there is no basis to suggest significant differences in ambient air concentrations at 3.5 meters and typical breathing level heights.

Conclusions and recommendations

The current number of sampling locations is below the de facto standard established at other DOE facilities. It is recommended that the number of sites that are monitored for tritium in ambient air be increased to cover at least all 16 wind direction sectors. This will ensure that accidental and diffuse releases that may bypass stack monitors would be detected. As of January 2001, LBNL has proposed to increase the number of ambient air monitoring stations to 14. This is a significant improvement over the current situation. It is noted that not all potentially impacted sectors are covered. It would be useful if LBNL would provide a rationale for the sampling locations.

[3] In particular, the May 1996 Compliance Plan (FFCA between LANL and EPA) requires for sampling of non-point sources: ”A standard 16-sector radial array (22.5-degree sector angle) from potential release sites will be used to evaluate potential MEI sectors. As a conservative measure, samplers will also be located in the sectors adjacent to the sectors adjacent to the sectors of maximum off-site impact if any population exists in those sectors”.

The selection of precise sampling locations should be based on a detailed evaluation of expected tritium concentrations in air using a dispersion model capable of accounting for the complex terrain and the short-term nature of tritium releases. It is obvious that in a given wind direction sector, the monitored location will not always reflect the largest offsite concentration. There is, however, an upper limit to the ratio of (maximum offsite air concentration)/(maximum monitored air concentration). This ratio can be calculated using appropriate dispersion models. It is suggested that this information be included in the annual environmental monitoring reports.

We understand that considerations are being made to remove the present tritium stack to a new location at building 75. This will likely decrease the impact on off-site locations. In addition, a contract is being arranged with U. C. Davis to perform wind tunnel modeling of the LBNL site which theoretically would provide scientific grounds for the establishment of environmental monitoring stations. We support both of these goals.

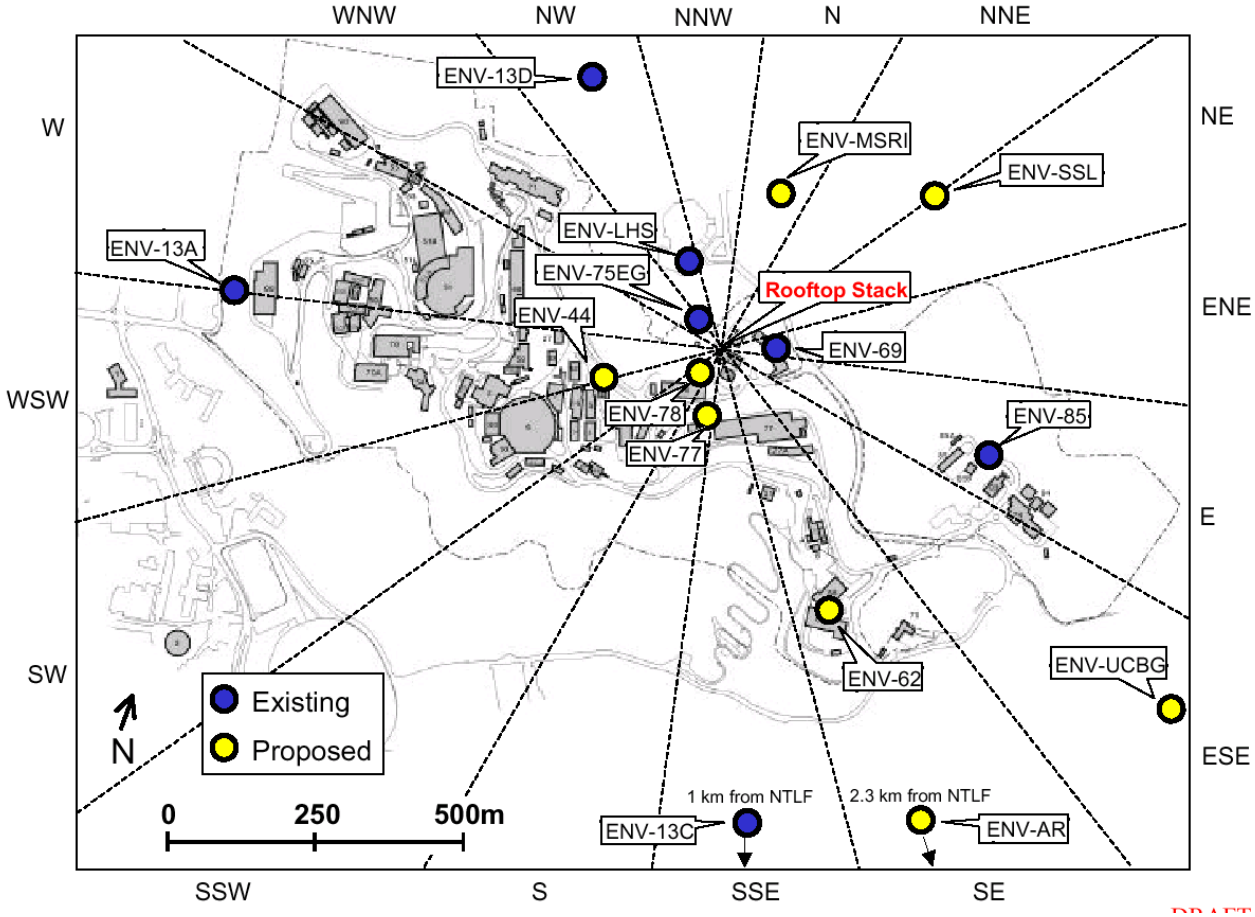

Figure 9. LBNL's current and proposed tritium in ambient air monitoring network (after Thomas et al., 2001)

A.4 Is the sampling and analysis of tritium in air at a given location sufficiently accurate?

Approach
Review observed versus expected water collected in silica gel samples
Review results of split sampling program
Review of contract laboratory performance

Findings
Silica gel sampling of tritium in ambient air will only return adequate information if all water which is in air is actually captured on the gel. Observations of the sampling system at Los Alamos National Laboratory showed that less water was collected than expected from humidity measurements in air. This problem was especially pronounced in dry summer months. The authors asked LBNL to provide a comparison of the amount of water actually collected in the silica gel samples with the amount expected in the volume of air passing through the gel during the same time period. The expected moisture was estimated using data from the onsite meteorological station. The result is shown in Figure 10 using ENV-69 station as an example. There is a reasonable correlation between the two data sets. The ratio (observed/expected) for a monthly sample was in a range of 0.77 to 1.28, for the annual average the range was 0.97 to 1.08. Surely, the problem seen in New Mexico is not observed here.

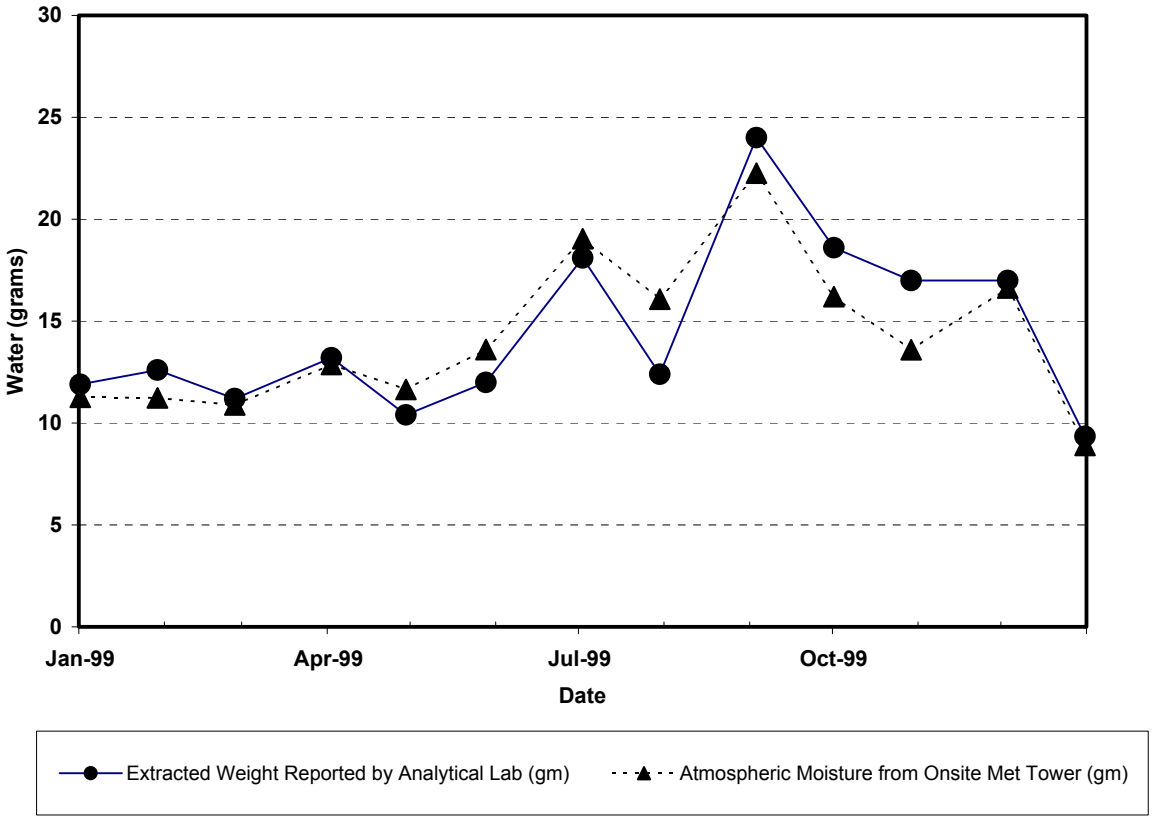

Figure 10. Comparison of observed and expected water collected at ENV-69

Figure 10 indicates on the other hand that in some months, the amount of water extracted from the silica gel exceeded the amount expected from onsite met tower data. This may well be due to the fact that silica gel has an initial water load prior to environmental sampling that is driven out in the distillation process in the laboratory. This additional amount of water can be in the order of several grams. It is therefore suggested that the weight of water collected in silica gel sampling be determined by the difference in silica column weight before and after the sampling period. The weight difference rather than the amount of water extracted should be compared with the amount of water expected from the onsite meteorological data.

Since November of 1997, the water extracted from silica gel samples are split and analyzed both by LBNL and with the EPA laboratory. The results for two stations are shown in Figure 11 and 12. As expected, the correlation is better for station LHS due to higher concentrations of tritium. In the annual average, the values reported by EPA are 20% higher for the station ENV-13D and 3% for ENV-LHS, the uncertainty is larger for individual weekly samples.

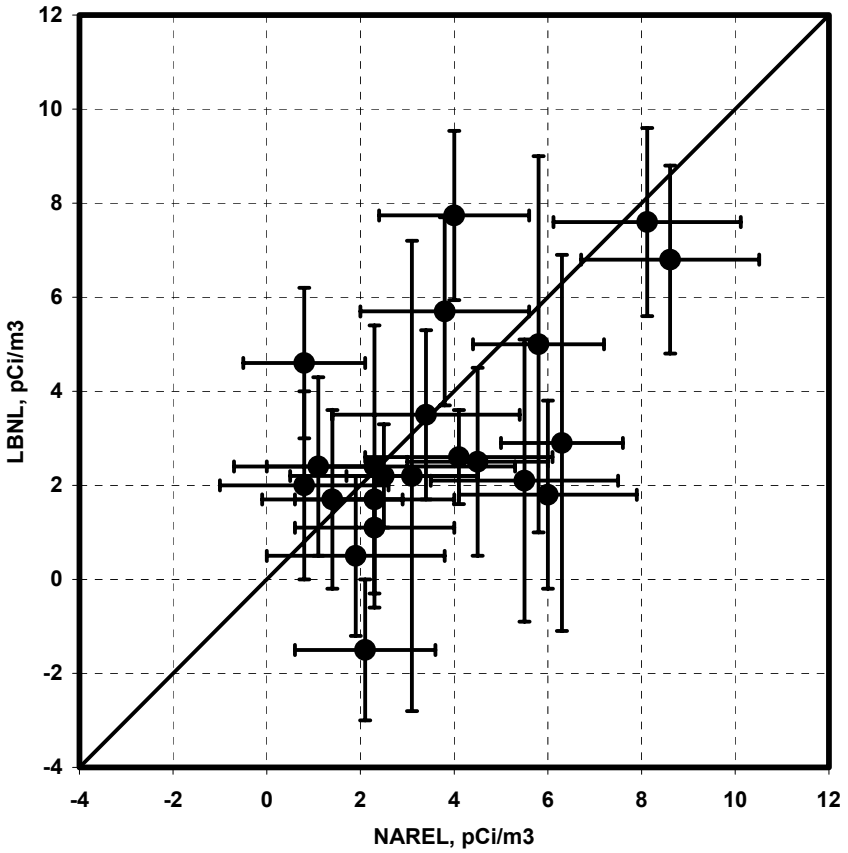

Figure 11. Comparison of tritium split samples EPA's of NAREL and LBNL analytical laboratories for station ENV-13D

Spot checks of the Tritium Monitoring Analytical Laboratory Data for 1998 that was provided by LBNL indicates that the sampling and analysis data are properly documented and that the calculations based on this data are verifiable.

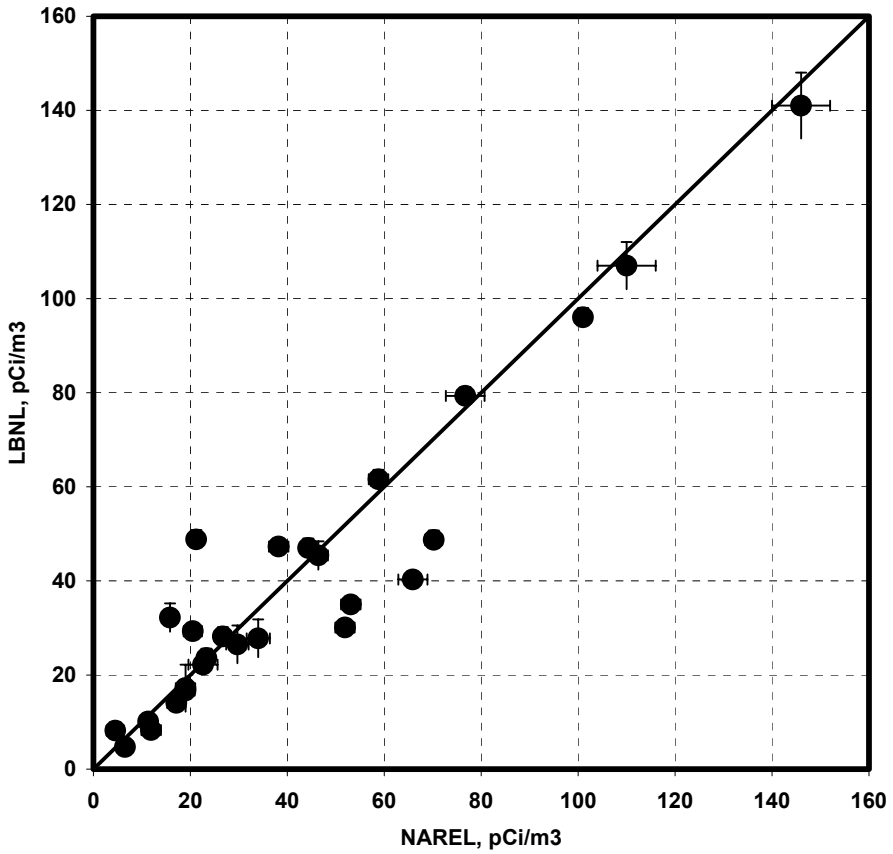

Figure 12. Comparison of tritium split samples of EPA's NAREL and LBNL analytical laboratories for station ENV-LHS

There are two additional concerns which were raised by members of the public: (a) that the intake of air sampler at LHS above the breathing level and that (b) real-time monitoring of ambient air would provide better information than silica gel sampling. Because of atmospheric dispersion, there is little variability of tritium air concentrations between one meter and five meters above ground. Even if the concentration at breathing level would be twice or even ten times as high as at the sampling level, the resulting doses are far below the 10 mrem/yr level. Real-time monitoring is not suitable for ambient air because the detection limit is several orders of magnitude higher than that of silica gel sampling.

Conclusions and recommendations

Based on the data reviewed thus far, the analytical data for HTO in ambient air samples is verifiable and is subject to reasonable uncertainties. With regard to analysis of samples collected at the Lawrence Hall of Science, the annual average concentration of tritium appears to be subject to a combined uncertainty of less than 20%. It is important to ensure that the amount of water extracted from silica gel is not diluted with water loaded onto the gel prior to environmental exposure. It is recommended that information regarding the uncertainty of sampling and analytical data be incorporated in the annual environmental reports. The height of air intake has little impact on measured concentrations, the request to install real-time ambient air monitoring has little merit because it lacks the suitability to detect low ambient levels.

A.5 Are radiation exposures to individuals (including sensitive subgroups) from NTLF operations below 10 mrem/yr?

Approach
Review NESHAP compliance assessment; determine exposure scenarios that are not covered

Findings
The radiation doses to individuals which result from NTLF emissions is a subject of concern and controversy. LBNL is required to determine compliance the operation of NTLF with the dose limits established in Subpart H of 40 CFR 61. LBNL's NESHAPS report for 1998 and 1999 identified the maximum exposed individual as a person located at UC Lawrence Hall of Science (LHS) at 110 meters Northwest of Building 75. For continuous exposure at this location, the doses for this location are reported as follows:

for 1998: 0.28 mrem/year
for 1999: 0.08 mrem/year

The doses were calculated based on the reported emissions from NTLF which are entered as input into the CAP88PC computer program. The program calculates the ambient air concentrations and subsequent radiation exposures based on meteorological data for the year in question. NESHAP requires LBNL to show compliance for locations where there is an residence, office, or school. The compliance location for NTLF is the Lawrence Hall of Science. Compliance with the dose limit is not necessary for other publicly accessible locations. That is a serious shortcoming of the regulation.

Each measurement and model prediction is subject to uncertainties which can be systematically assessed. The limited resources of this project did not allow a full investigation of all input and model parameters to be conducted. Instead, one may simply ask for the year 1998: is it possible that doses for a given individual are greater than 10 mrem/yr or 35 times the LBNL estimate for Lawrence Hall of Science? Which factors could account for a larger dose?

Let us first estimate the dose for the compliance location at the Lawrence Hall of Science. For any location, it is better to use measured concentrations than to rely on the results of a computer model, presuming of course that the measurements are accurate. Calculations with CAP88 and a zero m stack height Byrne (1999) are not in agreement with measured data.

For 1998, the measured concentration of HTO in ambient air at LHS was 46 pCi/m^3. The error associated with sampling and analysis was discussed in chapter A.4 and can be conservatively assumed to be less than 50%; hence the "real" air concentration at the sampling location could have been as high as 69 pCi/m^3. In contrast to this, CAP88PC predicts that all tritium released including HT is immediately converted to HTO and that the annual average HTO concentration at LHS was 86 pCi/m^3. Hence, the predicted concentration for the compliance location is larger than the measured even if uncertainties associated with sampling and analysis are accounted for.

The tritium inhalation dose factor in CAP88PC is 1.3 x 10^{-7} mrem per pCi of inhaled HTO and includes both the contribution from inhalation and skin absorption; the latter is estimated to be one half of intake from inhalation. Based on data provided in the SENES September 2000 report (Thomas and Hoffman, 2000), the upper limit (95% confidence interval) dose factor for the

critical age group (5 yr old) is 2.2 x 10^{-7} mrem per pCi inhaled. Accounting for the additional uptake from skin absorption and conservatively assuming that the relative biological effectiveness RBE of beta radiation from tritium compared to gamma radiation is 5 (rather than 1 as assumed on CAP88), the resulting dose factor would be 1.7 x 10^{-6} mrem per pCi inhaled. If we further assume that the real air concentration at the Lawrence Hall of Science was at the upper limit of 69 pCi/m^3 and that a 5 yr old child was present at the point of air sampling for the entire year in 1998 and breathed 20 m^3 of air per day like an average adult, the resulting dose using the upper limit dose factor would have been 0.86 mrem/yr. This is an upper limit dose estimate for that location; if a person spent less time there, doses would be smaller. For 1999, the upper limit dose calculated with the same set of assumptions was 0.34 mrem/yr.

Hence, the data for 1998 and 1999 does not suggest that any individual present at the Lawrence Hall of Science received a dose in excess of the 10 mrem/yr limit.

Now that we have reviewed the situation at the Lawrence Hall of Science, what can be said about other locations? In the June 30, 1999 preliminary report, IFEU pointed to the possibility that larger doses could occur due to the discontinuous nature of releases at NTLF as discussed in chapter A.2. A partial response to this question is contained in the May 2000 and September 2000 report by SENES Oak Ridge Inc. (Radonjic et al., 2000; Thomas and Hoffman, 2000) which were prepared under contract with LBNL.

Whereas it is well established that short-term releases have occurred, the problem is that rarely do we have measurements at the impacted areas from such releases. It is thus necessary to rely on dispersion models. The compliance model CAP88 is not suitable for such calculations; it does not allow calculations of dispersions for complex terrain or discontinuous releases. These limitations are even mentioned in the user manual. SENES selected the CALPUFF model which is better suited.

Based on model predictions with CALPUFF, the largest dose from a short-term release was calculated for a jogger exercising downwind at a distance of 41 meters from the NTLF while 218 mCi of HTO were released. That was the largest reported release over a 15-minute period for the hillside stack in 1998 and 1999. The resulting dose was calculated to be between 0.042 and 0.26 mrem (95% confidence interval).

In other words: it is possible that a person exercising for 15 minutes at a distance of 41 meters from the stack during a short term release received about 0.26 mrem which is almost a third of the dose to a child who is present each and every day for one year next to the Lawrence Hall of Sciences. While one has to acknowledge that the probability for either scenario is rather small, it is equally prudent to ensure that the doses for **both** scenarios are small as well.

There are two questions one may ask:

(a) What would the dose be when releases larger than 218 mCi in a 15-minute period?
(b) What would the dose be if the exposed person is closer to the stack than 41 meters (the fence is about 21 meters from the stack)?

The answer to (a) is simple. The larger the release, the larger the dose. For the jogger scenario, a 10 mrem limit dose (upper 95% confidence interval) could result if the release was 8.4 Ci. Was there a release this high? The answer is: it may have been. There was one

possibility that a release a 15-minute release exceeded 218 mCi in 1998: the building 107 roof vent during the accident on July 24, 1998. The SENES report (Radonjic et al., 2000) assumed that the reported release of 35 Ci of HTO was evenly distributed over a 6-hour period; this equals 1.5 Ci in a 15-minute period. The report states on page 36:

> *"Evidence does not exist to support a different fractionalization of the total release amount. However, time-variable emissions during this period would lead to significantly different results in predicted concentrations at this location, depending on the wind direction and wind speed during the 6-hour period."*

There is indeed no information available regarding the fractionalization for the total release of 35 Ci because the silica gel sampling does not provide real-time information. However, real-time data from the hillside stack emissions of the same day may suggest a different picture. If one assumes that the release from building 107 roof vent was distributed over time in the same pattern as is indicated by real-time data from the hillside stack shown in Figure 8, about 24% of the total release (8.4 Ci) would have been emitted over a 15-minute time period. It is a shortcoming of the May 2000 SENES report in that it did not evaluate air concentrations and subsequent doses from the accident for locations other those monitored.

Let us assume for a moment, that indeed 8.4 Ci of HTO were released over a 15-minute period. Using the scenarios developed by SENES (unfavorable weather conditions, not the actual weather data for July 24), the maximum dose would be received by a person exercising downwind at a distance of 41 meters from the NTLF stack release (1.6 to 10 mrem). This scenario is entirely theoretical because worst case dispersion conditions did not prevail on July 24 (noontime wind speed of 2 m/s is reported rather than the worst case of less than 1 m/s) and there are no reports that a person was actually present close to the fence during the accident. However, the calculation nevertheless illustrates the possibility that short-term accidental releases similar in magnitude to the 1998 accident could result in doses to people exceeding 10 mrem.

The answer to (b) is more complicated. The very nature of scenarios is that one can always postulate more extreme ones. The closest distance that one can get to the hillside stack is 21 meters, rather than 41 meters as assumed by SENES, although vigorous exercising next to the fence is hardly realistic.

How can one resolve this predicament? The answer may lie in the data provided by the sampling station close to the fence (ENV-75EG) which was sampled since January 2000. For year 2000, average concentrations for the three stations ENV-LHS (Lawrence Hall of Science), ENV-75G (Station close to the fence from the hillside stack) and ENV-69 (on NTLF site) were 18, 56 and 11 pCi/m^3 respectively. The average air concentration at ENV-75G was about 3 times larger than the one at LHS. Hence, even if a person had been present at that location for the entire year, doses would have still be far below the 10 mrem/yr standard. If it is ensured that the dose limit is not exceeded for this sampling location, the possibility that any individual receiving a dose in excess of the 10 mrem/yr limit is greatly reduced.

It is concluded from this review that there is no evidence to suggest that any individual received a radiation exposure resulting from NTLF emissions which exceeded 10 mrem/yr in the years 1998 and 1999. However, care has to be exercised to acknowledge the remaining uncertainties.

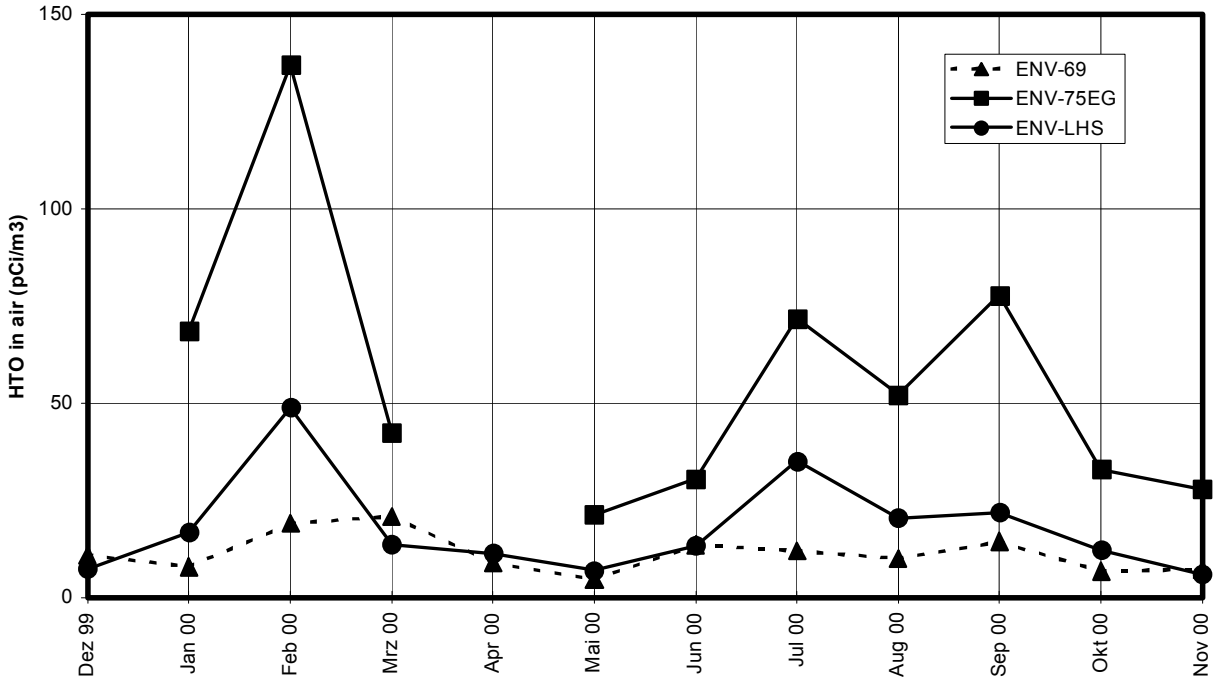

Figure 13. Ambient air concentrations December 1999 to November 2000 at stations ENV-LHS (Lawrence Hall of Science), ENV-75G (close to the fence near the hillside stack) and ENV-69 (onsite)

Conclusions and recommendations

There is considerable uncertainty in the CAP88PC model which is used by LBNL to determine compliance with regulations. Measurements of ambient air are the preferred method to ensure compliance with the 10 mrem/yr dose limit. It is concluded from this review that there is no evidence to suggest that any individual received a radiation exposure resulting from NTLF emissions which exceeded 10 mrem/yr in the years 1998 and 1999. However, some uncertainty remains because the evidence is limited. In order to reduce these uncertainties, the following activities are suggested:

(a) LBNL should use CALPUFF in addition to CAP88PC to determine compliance with the Clean Air Act, 40CFR61, Subpart H, accounting for discontinuous nature of the releases.
(b) In addition, the assessment should acknowledge the presence of transient receptors. This may best be done by using the property fence as compliance location and conservatively assuming an annual presence there of offsite personnel.
(c) Supplemental to (a) and (b), LBNL should demonstrate by dispersion calculations that the annual average air concentrations at any other offsite location are lower than those at the property fence if discontinuous releases are taken into account.
(d) Given that dose to people is the primary concern, once the stack is moved to B-75, LBNL should conduct routine bioassay for one year on staff not associated with the NTLF operation. The data should be published in the open literature.

A.6 How relevant is the presence of organically bound tritium?

Approach
Review of data for organically bound tritium and tissue free water tritium in vegetation

Findings
Tritium is taken up by plants through the roots and leaves. The organically bound tritium (OBT) is incorporated into sugars, carbohydrates and other molecules containing hydrogen. The tritium which can be extracted from plants by drying is called tissue free water tritium (TFWT). While OBT mainly reflects tritium which was incorporated into the plant tissues at the time of their formation, TFWT mainly reflects the current subsurface water uptake well as uptake of airborne tritium through the openings of the leafs. The total inventory of tritium in trees within a 200-meter radius from the NTLF stack was estimated by LBNL to be less than 1 Ci. In support of this estimate, LBNL provided the following information, (a) 500 trees in the 200-meter radius, (b) average tree mass of 6,000 kg, (c) average tritium activity is about 100 pCi/g. Using these values, the resulting inventory is 0.3 Ci. The value of 1 Ci appears to be a reasonably conservative estimate of the tritium activity in trees. It is roughly equivalent to the amount of tritium emitted from NTLF during three average days. Consequently, only a small fraction of the total airborne emissions was captured in trees around NTLF. The tritium inventory in groundwater is estimated to be less than 1 Ci as well (see chapter B.1).

For samples collected in 1996, Menchaca reported an OBT concentration of 524 pCi/g for plant foliage taken from a tree 2 meters from the NTLF stack (Menchaca, 1996). Based on tree samples collected in September 1998 reported in the sampling and Analysis Plan, the largest concentration of 8.7 pCi/g OBT was found in a tree 14 m SE from the stack (sample A1-SE-20-core). Even greater concentrations of 1,280 pCi/g were found in duff 20 m NNW from the stack (A1-NNW-20-duff). Both data sets are in reasonable agreement. Variability of OBT in vegetation is likely to be significant since the measured concentrations depends on the location of the tree, the type of tree and the age of the plant matter actually sampled. If tree wood is sampled for OBT, it is advisable to take samples from tree rings separately since tree ring analysis can provide valuable information about past exposures. OBT sampling can also be used to show the spatial distribution of the impact of past emissions from NTLF. Based on the Menchaca data, the highest concentrations were found in the NW sector. The decrease of activity between tritium in trees from the stack area (524 pCi OBT/g) and in 100 m NW (345 pCi OBT pCi/g) supports that there is a relatively low gradient in annual average airborne activity between the NTLF stack and LHS. The Menchaca data thus supports predictions based on air dispersion modeling.

Conclusions and recommendations
Only a small fraction of the total airborne emissions was captured in trees around NTLF. The inventory in trees in the 200 meter radius around NTLF is estimated to be less than 1 Ci; the tritium inventory in groundwater is estimated to be less than 1 Ci as well. Even if the entire tritium inventory in trees and groundwater were to be released into the air via leaf transpiration, the source term would be equivalent to the amount of tritium emitted from NTLF during a few average days of NTLF operation. It is recommended to continue sampling and analysis of organically bound tritium (OBT) as well as tissue free water tritium (TFWT) in plant tissues. Tree ring analysis can provide valuable information about past exposures.

A.7 How can the levels of tritium in rainwater be explained?

Approach
Review of data for tritium in rainwater.

Findings
There has been considerable debate about the levels of tritium found in rainwater. The discussion focuses on two issues: (a) the magnitude of the observed concentrations and (b) the apparent lack of correlation between tritium in rainwater and reported emissions from NTLF (Byrne, 1999).

The maximum level of tritium in rainwater of 775,000 pCi/L was reported for 1988 (LBNL 1997a), at a sampling location near building 75. How can this correspond with the reported HTO emissions from NTLF of 570 Ci in the same year? A rough calculation can provide an answer. The rainfall in Berkeley in 1988 was about 455 L of rain per square meter (LBNL 1989). Let us consider an average release of 1.6 Ci of HTO in a single day while wind blows into a 22.5 degree sector. We can further conservatively assume that all HTO is essentially washed out within one km. On an average day, there will be a total of about 510,000 liters of rain in the 22.5 degree sector out to distance of one km. If 1.6 Ci of HTO is evenly mixed with the rainwater in the sector, the resulting concentration is about 3,100,000 pCi of HTO per liter of rain water. The actual concentration decreases with distance from the source, and not all HTO is washed out at 1 km. Detailed modeling would require knowledge of the timing of the release and the timing of rainfall. However, the calculation shows that the magnitude of measured values is not surprising and could be expected when HTO emissions are in the order of a few hundred Ci per year (or about one Ci per day). Likewise, the levels of tritium in rainfall measured by Monheit in Albany, 4 miles northwest of NTLF, during fall of 1994 are not unexpected. While the reported HTO release from NTLF during 1994 was 115 Ci, she measured air concentrations up to 400 pCi/L. Whereas the concentration measured in 1988 was far in excess of EPA's drinking water standard of 20,000 pCi/L, concentrations in recent years were below that level.

An additional topic was the lack of correlation between tritium concentrations measured in rainwater and the reported emissions of tritium from NTLF in days prior to rainwater sampling for the year 1994 (Byrne, 1999). The lack of adequate data quality in that year makes this comparison extremely difficult if not impossible. The accuracy of weekly reported tritium emissions cannot be correctly determined. It would also be necessary to determine the precise timing of the releases within that week, the wind direction during the release and the precise start and end of rainfall. This information is not available. The main conclusion of this exercise is that tritium in rainwater is not as reliable an indicator as is tritium in ambient air.

Conclusions and recommendations
The measured concentration of tritium in rainwater in 1988 of 775,000 pCi/L is in reasonable agreement with expected concentrations. While the concentration measured in 1988 was far in excess of EPA's drinking water standard of 20,000 pCi/L, it should be noted that concentrations in recent years was clearly below that level. There is no correlation between tritium concentrations measured in rainwater and the reported emissions of tritium from NTLF in days prior to rainwater sampling for the year 1994. This illustrates the absence of adequate quality control of data for that year and confirms that tritium in rainwater is not as reliable an indicator as is tritium in ambient air.

A.8 Are measurements of discharges of radionuclides other than tritium into air and water from LBNL and the resulting radiation exposures sufficiently accurate?

Approach
Review of monitoring data regarding radionuclides other than tritium

Findings
The issue of monitoring radionuclides other than from NTLF is rather complex. The 1999 LBNL NESHAPS report identified a total of 119 potential NESHAP area sources. Potential sources are Radioactive Material Areas (RMAs) or Radiological Storage Areas (RSAs) at the Berkeley Lab. The threshold for monitoring is the definition of small sources, i.e. only sources with a potential dose exceeding 0.001 mrem/yr are monitored. 12 sources are sampled continuously with monthly analysis; 10 sources are sampled continuously with weekly analysis. Only one source, building 75 (NTLF) is monitored continuously.

The decision to regard an RMA or RSA on the list of "small sources" is a complex process which requires knowledge of the radionuclide inventory in a given building. Based on the inventory data, potential doses are calculated to determine the need for sampling. The requirements outlined in 40CFR61.95 state the following:

> *All facilities must maintain records documenting the source of input parameters including the results of all measurements upon which they are based, the calculations and/or analytical methods used to derive values for input parameters, and the procedure used to determine effective dose equivalent. The documentation should be sufficient to allow an independent auditor to verify the accuracy of the determination made concerning the facility's compliance with the standard. These records must be kept at the site of the facility for at least five years and, upon request, be made available for inspection by the Administrator, or his authorized representative.*

Due to the constraint of resources, it was not possible to conduct the required review to determine whether LBNL's sampling program was in compliance with regulatory requirements. It is noted that reported doses for the maximally exposed individual (MEI) at locations other than NTLF are well below 10 mrem/yr (1998: 0.0054 mrem/yr for building 55/56; 1999: 0.0089 mrem/yr for building 55/56). No documentation was reviewed about the potential emissions from non-sampled sources since establishing the validity of the documented data and the compliance with 40CFR61 Subpart H is a time-consuming process.

Conclusions and recommendations
In 1999, there were 119 potential locations at LBNL where radioactive materials are present and could be released into the environment. Of these 23 are sampled for airborne releases. Ninety-six (96) potential sources are not monitored because potential doses are estimated by LBNL to be below 0.001 mrem/yr. 40CFR61 Subpart H requires that the documentation be verifiable with regard to compliance with the standard. Due to the limitations of the contract, it was not possible to conduct the required review to determine whether LBNL's sampling program was in compliance with regulatory requirements.

A.9 Are measurements of gamma and neutron radiation from LBNL sufficiently accurate?

Approach
Review environmental monitoring of gamma and neutron radiation

Findings
The maximum doses from penetrating gamma and neutron radiation at perimeter locations are reported as follows:

for 1998: 0.4 mrem/yr due to accelerator-produced penetrating radiation (Building 88)
<1 mrem/yr due to irradiator-produced penetrating radiation (Building 74)
for 1999: 0.3 mrem/yr due to accelerator-produced penetrating radiation (Building 88)
<1 mrem/yr due to irradiator-produced penetrating radiation (Building 74)

The doses are well below the limit of 100 mrem/yr for penetrating radiation. Uncertainties associated with these estimates are small.

In addition, noble gasses and gaseous activation products are generated as a result of accelerator operations. All of these products were reviewed with public exposure as the focus. Most of these products have very short half-lives such that they have decayed away before reaching the public environment. The remainder of these emissions was not found to be sufficient to cause significant public exposures.

Conclusions and recommendations
Current doses from gamma and neutron radiation resulting from LBNL operations are rather small. Uncertainties are considerably less than those described for tritium.

B Legacy contamination from past operations / Superfund issues

B.1 Is LBNL's Draft Tritium Sampling and Analysis Plan sufficient to determine the extent and nature of legacy contamination at NTLF?

Approach
Review of sampling plan regarding sampling media, sampling locations, analytical techniques, and QA/QC issues.

Findings
The Draft Tritium Sampling and Analysis Plan currently addresses the following environmental media:

- ambient air
- soil
- sediment and surface water
- vegetation

The EPA hazard ranking for the NTLF site indicates that **ambient air** data will be most decisive for the evaluation. It is therefore necessary to properly address the issues in section A.2 to A.4. It is advisable to expand the network of ambient air sampling stations to cover all 16 wind direction sectors (of 22.5° each). This will ensure that LBNL site is sampled with a network of comparable density to that at other DOE facilities. It is not sufficient to focus on the predominant wind direction sectors because tritium releases from NTLF often occur over very short time periods during which direction of the wind can differ greatly. The placement of the stations should be optimized pending the results of the proposed modeling with a complex terrain and conservative release patterns for NTLF. The suggested increase of monitoring locations to a toatl of 14 (chapter A.3) come close to the suggested improvement. It would be useful if LBNL could provide the rationale for the sampling locations shown in Figure 9.

The proposed sampling locations for **soil** are in all areas that can reasonably expected to have been contaminated. However, given the fact that large tritium releases may have occurred during wind directions with a low frequency, emphasis should be placed on the representative sampling of the area around each site. Rather than the proposed single sample from each site, ten cores should be taken according to the core method in DOE's Environmental Measurements Laboratory Procedures Manual HASL-300, section 2.3.4.1 (EML, 1997). An additional requirement of this procedure is to select undisturbed sites if possible. The sampling plan is further deficient in providing a rationale to limit sampling to one depth increment of 15 to 30 cm. In order to provide a representative results for all soil depths which are potentially contaminated, it would be advisable to collect the following depth increments: 0 to 15 cm, 15 to 30 cm, and 30 to 60 cm, and 60 cm to 150 cm.

It is prudent to adopt the HASL-300 core method for soil sampling because it greatly reduces uncertainties due to the variability in soil activity in a given location. Given that the sampling area is already defined, it will not always be possible to select undisturbed locations as HASL-300 demands. It is, however, possible to document to what extent the sampling site deviates from the requirements for “ideal” sites. In addition, one should differentiate between the proposal to take ten sub-cores at a given location and my proposal to sample for different depth

increments. In any case, it is strongly suggested to take ten sub-cores which are mixed for each depth increment. This is a modest and justified increase in sampling efforts.

LBNL originally proposed to use the HASL 300 Method at any location where a soil sample showed the tritium concentration to be greater than 10 percent of the EPA Preliminary Remediation Goal (PRG) (risk of 10^{-6}) for tritium in residential soil of 11,000 pCi/g. In a December 21, 2000 message from Iraj Javandel to Nabil Al-Hadithy, it is proposed to include all of the 8 sampling locations that are outside of the Berkeley Lab property between the hillside stack and the Lawrence Hall of Science. This matter should be resolved on the basis of good science. As explained above, a technical point of view, the use of the HASL-300 method for all soil samples provides more reliable results. The additional effort to use this methods for all sampling sites is small and justified. It is assumed that EPA is also interested to review soil data which is statistically more reliable even though US EPA Guidance for Performing Site Inspections under CERCLA, of September 1992 allows to take grab samples. Sampling additional depth increments will provide valuable information as to the extent of the contamination. If funding for this is not available at this point, one could take at least samples from these depth increments and store them for later analysis. The cost of taking and bagging additional samples is modest.

The proposed program of **sediment and surface water** and **vegetation** sampling is well designed. No changes appear necessary.

As to the need for **groundwater samples**, the IFEU team feels this issue is best resolved in coordination with the California Regional Water Quality Control Board. The overall inventory in groundwater is likely to be less than 1 Ci. One should note that groundwater is not used for human consumption; hence groundwater monitoring is not required for EPA's hazard ranking. If groundwater is used for other domestic, non-drinking purposes, further evaluation of exposures should be made. While **rain water sampling** is not required by EPA, tritium may be present in rain. The EPA risk assessment should evaluate this exposure pathway.

The NTLF site was not the only one affected by past operations. While Integrated concentrations of tritium in ambient air at **Building 3 (Calvin)** on the UC campus are comparable to those measured at NTLF, this was likely due the fact that air sampling at the Calvin lab was conducted only 2 meters from the roof stack compared to 110 meters for the NTLF hillside stack. Since the original recommendation in IFEU's June 2000 report, soil sampling around building 3 (Calvin) was implemented. According to the results that were received from Dr. Lavely, Office of Radiation Safety at UC Berkeley, only one out of 19 samples on UC property with tritium concentration above the minimum detectable activity. The only tritium concentration above the detection limit was found at the north side of Calvin Hall (+18.8 +/- 4.1 pCi per gram of soil). This concentration is well below to EPA's Cancer Risk Screening Concentration for tritium in soil of 11,000 pCi per gram. Judging from these results, the question about tritium contamination in soil around Calvin has been resolved. There is no Superfund issue at Calvin.

In order to evaluate the **performance of Thermo NUTech** of Richmond, CA, the laboratory selected by LBNL, DOE's Environment Measurement Laboratory (EML) reports on quality assessment the participating the QA program for analysis of uranium in soil were reviewed (EML, 1997-2000). The EML program consists of supplying samples which are spiked with a known amount of radioactive material. EML rates the results as follows: acceptable (A):

between 15th and 85th percentile of cumulative normalized distribution; acceptable with warning (W): between 5th and 15th or 85th and 95th percentile of cumulative normalized distribution; not acceptable (N): less than 5th or greater than 95th percentile of cumulative normalized distribution.

Table 5. EML performance rating for Thermo NUTech of Richmond, CA

EML QAP No.	Date	Matrix	number of radionuclides analyzed	rated A	rated W	rated N
45	Jan 97	Air Filter	10	9	1	
46	Jun 97	Air Filter	16	7	1	8
47	Jan 98	Air Filter	15	14		1
48	Jun 98	Air Filter	16	13	3	
49	Dec 98	Air Filter	13	11	1	1
50	Jun 99	Air Filter	13	8	2	3
51	Dec 99	Air Filter	14	13	1	
52	Jun 00	Air Filter	13	10	2	1
53	Dec 00	Air Filter	13	9	4	
45	Jan 97	Soil	10	10		
46	Jun 97	Soil	9	8	1	
47	Jan 98	Soil	9	8		1
48	Jun 98	Soil	7	4	2	1
49	Dec 98	Soil	11	9		2
50	Jun 99	Soil	11	8	3	
51	Dec 99	Soil	10	8	2	
52	Jun 00	Soil	10	6	4	
53	Dec 00	Soil	11	5	4	2
45	Jan 97	Vegetation	7	6	1	
46	Jun 97	Vegetation	7	4	3	
47	Jan 98	Vegetation	7	7		
48	Jun 98	Vegetation	7	4	3	
49	Dec 98	Vegetation	7	4		3
50	Jun 99	Vegetation	8	7	1	
51	Dec 99	Vegetation	7	4	3	
52	Jun 00	Vegetation	7	4	3	
53	Dec 00	Vegetation	7	5	1	1
45	Jan 97	Water	14	11	3	
46	Jun 97	Water	12	11	1	
47	Jan 98	Water	15	13	1	1
48	Jun 98	Water	14	10	2	2
49	Dec 98	Water	15	11	1	3
50	Jun 99	Water	14	14		
51	Dec 99	Water	14	12	1	1
52	Jun 00	Water	13	12	1	
53	Dec 00	Water	12	10	2	

Table 5 summarizes the results of EML performance rating for the Thermo NUTech, Richmond, CA, for the entire array of radionuclides for which this laboratory reported results. Thermo NUTech's performance rating decreased with regard to radionuclide analysis of soil samples from January 1997 (86% rated "A") to December 2000 (46% rated "A"). The rating for tritium (analyzed in water matrix) was a consistent "A" for the entire time period.

The results indicate that Thermo NUTech has a reliable performance when it comes to analysis of tritium in water. If LBNL relies on the laboratory for other types of analysis as well, in particular for soil samples, the reasons for the decline on performance rating should be evaluated more closely.

Conclusions and recommendations

The Draft Tritium Sampling and Analysis Plan sampling and analysis program should be supplemented. The ambient air monitoring should be expanded to cover all 16 wind direction sectors (of 22.5° each). The selection of precise locations should be based on a detailed evaluation of expected tritium concentrations in air using a dispersion model capable to account for the complex terrain and the short-term nature of tritium releases. The HASL-300 core method for soil sampling should be used; samples to be analyzed for additional depth increments. The issue of sampling of groundwater should be resolved in coordination with the State of California Regional Water Quality Control Board.

B.2 Which other factors need to be addressed in EPA's evaluation of the Superfund status for the NTLF site?

Approach
Review whether NTLF operations will be typical during sampling period; review of non-radiological data (e.g. number of affected residents)

Findings
EPA will assess the score of the LBNL site using the Hazard Ranking System (HRS) scoring process. There are valid concerns in the community that operations at NTLF during the sampling time may not be representative of typical operations. This issue was addressed in section A.1. It was determined that the tritium inventory does not serve as a good indicator of laboratory activity due to the large uncertainty inherent in the data. It is therefore recommended that an array of information be used in the determining whether NTLF is operations are representative including the shipment of products and number of tritiations performed.

EPA concluded in its preliminary assessment that the Lawrence Hall of Science (LHS) is not regarded as a school. While this is a correct finding, one should not neglect the fact that LHS is visited by more than 300,000 people annually, a third of which are students. If LHS would qualify as a school, the number of students would be entered in the HRS scoring system. In order to determine whether this assumption is significant in HRS scoring, it is proposed that EPA provide two separate sets of scoring calculations, one of which assuming LHS as a school, accounting for the full-time equivalent visitor population plus resident staff. Alternatively, the number of repeat visitors should be determined. The exposure to transient receptors (jogger scenario) should be assessed.

Conclusions and recommendations
The EPA will score the NTLF site according to the procedures set in the Hazard Ranking System (HRS). Because of concerns in the community that operations at NTLF during the sampling time may not be representative of typical operations, an array of information should be used in determining the type of NTLF operations. This may be done by a review of product shipments and the number of tritiations performed. It is also recommended that EPA should provide information as to how the hazard ranking score would change if Lawrence Hall of Science would be regarded as a school, accounting for student population.

B.3 What is the concern regarding contamination by radionuclides other than tritium?

Approach
Review of non-tritium radioactive contamination at LBNL

Findings
The limited resources available in this project did not allow more than a cursory review of material in this respect. The authors note that extensive documentation exists regarding potential soil contamination as a consequence of historical operations which was collected as part of RCRA remedial activities. Current and former employees at LBNL who were interviewed indicated that significant contamination of soil no longer exists at the Laboratory. Previously occurring spills of heavy elements have been cleaned up to AEC/DOE prescribed acceptable levels with the exception noted below.

An experiment at the 184 Inch Synchrocyclotron in the 1960s involved irradiation of gram quantities of natural uranium. Some of the target material leaked into the soil at the facility, but analyses indicated that the resulting concentrations were within the range of naturally occurring uranium in ambient soils in the Bay Area. Therefore LBNL management made the "no action" decision regarding remedial action.

Due to resource constraints, the authors did not attempt to independently verify the validity of these claims.

Conclusions and recommendations
There is non-tritium radioactive contamination from past operations at LBNL. It is widely believed that cleanup of such areas has either been conducted or were deemed to be unnecessary due to the lack of hazard. The limited resources available in this project did not allow an independent verification of these claims.

C Historical exposures (1997 and earlier)

C.1 What exposures to neutron and gamma radiation resulted from LBNL operations?

Approach
Review of historical data on neutron and gamma exposures

Findings
At early times neutron exposures of employees (as well as possible offsite exposures) were significant. Professor E. O. Lawrence himself requested that the Physiology Dept. at U. C. Berkeley look into the possible harmful effects of neutrons after finding the at the building which housed one of his cyclotrons had become activated by neutrons. Lawrence et al began construction of the 184 Inch Synchrocyclotron in 1940, the magnet of which was used in his "Calutron" experiments to enrich uranium for use in atomic bomb research. This site was on the "hill", and separate from his earlier cyclotron research on the Berkeley Campus proper. The hill site became the Lawrence Berkeley Laboratory. The accelerators at Lawrence's labs were used primarily for research in high-energy physics, but also for radiation biology, medical research, atomic physics, heavy radionuclide production and research (Heilbron (1981)), and very high intensity photon sources.

The Atomic Energy Commission (AEC), followed by the Energy Research and Development Administration (ERDA), and then the Department of Energy (DOE) maintained regulatory statutes for public protection against radiation-related injury. The AEC protection limits were first established in the 1940s. Later, AEC Manual Chapter 0524 became the reference. The DOE retained Chapter 0524 until 10CFR835 came into effect.

The calculation and/or measurement of radiation doses are called dosimetry. The International Commission on Radiological Protection (ICRP), the National Council on Radiation Protection and Measurements (NCRP), and the International Commission on Radiation Units and Measurements (ICRU) provide guidance on the units and measurement techniques used for protection of personnel.

One of the most recent reports is ICRP Publication Number 60, 1990, Recommendations of the International Commission on Radiological Protection", ICRP (1990). This report recommends that the "equivalent dose", **H**t, equals the absorbed dose, **D**t, times a radiation weighting factor, **w**t. The weighting factor has changed over the years, primarily for neutron radiation for a number of reasons, but primarily because of better knowledge of the effects of that radiation on the health risks that exposure to it engenders. Neutrons of energies between 0.1 and 2 MeV have the highest weighting factor of 20, meaning that neutrons within this energy region are twenty times a damaging per unit absorbed dose as are typical gamma rays or ortho-voltage x-rays. These radiations as well as beta rays from radioactive materials are considered to be the least harmful per unit absorbed dose. The variation in weighting factors over the years, as well as the range of weighting factors can produce factors of at least two fold variations in the allowable dose from broad-spectrum neutron sources such as accelerators. Recommendations in ICRU Report 57 (ICRU (1998)) suggest further that the ISO and ROT conversion coefficients be used for environmental dose estimates since they best satisfy the uncertainty about he

positions and activities of the general public near high-energy accelerators. Thus conditions of exposure as well as the risks attendant to them have become better defined over the years, allowing for more accurate assessments of neutron and other radiation doses.

Operating **accelerators** produce a variety of radiation fields outside of the biological shielding which is intended to protect personnel from radiation exposures. These are primarily neutrons, gamma rays, muons, and other radiations of which neutrons have the highest intensity, and are the most damaging from a health risk point of view.

Personal neutron exposures were monitored as part of the personal dosimetry program at LBNL from ~1959 to the present. During the mid-1980ies it was determined that personal monitoring for neutrons was no longer required for individuals who did not work around the Laboratory's accelerators.

The Laboratory's environmental monitoring reports were reviewed for periods from 1960 to 1976 (LBNL, 1960 to 1977). During this time the Bevatron accelerated protons and other light ions to energies, which reached 6.2 GeV, the maximum endpoint energy for protons. Neutron production was incidental to the acceleration of light ions, and because of their lack of electrical charge, they penetrated the thick concrete shielding to produce exposures in persons both on-site and off-site. In fact, there was no roof shielding during the early period of Bevatron operation. The spectrum of these neutrons was best described by the function 1/E, where E is the neutron energy. Superimposed on this I/E spectrum were the contributions from sky shine and evaporation neutrons from interactions of protons with iron in the magnet structures. One can derive a neutron field through thick shielding for a proton beam at 6.4 GeV such that the neutron spectrum would extend from "thermal" energies (average of 2.5 E–8 MeV) to 6.4 E3 MeV. From the Olympus Gate monitoring station the Bevatron looked like a point source producing a neutron field described by 1/E up to 6.4E3 MeV. "Sky shine" neutrons resulted from high-energy neutrons escaping the shielding in a roughly vertical direction, and interacting with molecules of air, resulting in their being scattered back to the ground at substantially reduced energies (≤10 MeV). The sky shine neutron spectrum declined as roughly 1/r from the Bevatron, whereas the direct neutron spectrum declined as $1/r^2$, where r is the distance from the Bevatron.

An early environmental report (1971) contained data that neutron doses exceeded the allowed annual dose prescribed by the Atomic Energy Commission. This limit was 500 mrem/y, while the dose reported at the Olympus Gate was 800 mrem/y. This dose was verified during a meeting with LBNL representatives last year. However, an LBNL report was published soon after that meeting which raised issues regarding conversion of neutron spectra to dose. That report made a credible case for reducing the earlier reported doses by a factor of at least two (2). A description of the rationale used in the report follows.

Radiation doses calculated for any of a variety of recipient conditions could vary a great deal. No attempt was made to calculate doses to persons beyond the site boundary, but rather to keep the boundary doses within acceptable limits.

The best estimate of the impact of neutrons on the environment is a description of the neutron spectrum as a function of energy (the differential energy spectrum). Issues relating to exposure of persons to that neutron field may have a profound effect on the description of radiation dose and consequent health risk from that exposure. For example, the dose itself can be calculated for persons facing the source (AP), away from the source (PA), laterally from the side of the

body (LAT), rotating with respect to the source (ROT), or exposed to an isotropic source (ISO), ICRP (1997) and ICRU (1998). Additionally, shielding may be provided by a housing structure, for instance. These issues combined with the uncertainty associated with residency times can force a dramatic impact on dose estimates in the public sector. Hence, the decision about minimizing the dose at the site boundary was made.

More recently, and likely in response to inquiries made by these authors, R. H. Thomas, et al, published a document, Thomas (2001), which negated the high neutron doses previously mentioned. This was largely due to reinterpretation of neutron fluence-to-dose information at the site boundary. The neutron fluences as functions of neutron energy previously referred to apparently did not result in doses in excess of those required by the (then) AEC for protection of the general public.

Also, Donahue et al published in draft a report in which the neuron spectra at the Olympus Gate Environmental Monitoring Station (OGEMS) were mathematically generated by Monte Carlo techniques in a computer. The normalized spectra were then multiplied by the measured spectra at the OGEMS, and these results compared with the high neutron dose reported in the 1972 Environmental Monitoring Report. The result, when the newer technique (ICRU, 1998) was used to estimate doses, verified that the earlier reported dose was a factor of 2 too high (Donahue, 2000). The complexity of the Bevatron prevented a completely independent estimate of environmental dose using the Donohue technique.

As noted above, the originally reported exposures were in excess of the dose limit of 500 mrem per year set in AEC Manual Chapter 0524, dated February 1, 1958. It appears that LBNL was subject to AEC Manual Chapter 0524 regulations since the laboratory was an AEC contractor. The Manual chapter 0524-02 paragraph 2 states that existing facilities can apply for a conversion period not to exceed five years if a request is made by an appropriate AEC official. IFEU has asked the LBNL to supply a written copy of the request if it was made at the timely response to this request, LBNL has provided documentation from 1958 and 1959 demonstrating than Berkeley Lab developed shielding plans for the Bevatron to meet the new limits. Whether or not this provides evidence of an application in agreement with the provisions set in manual chapter 0524-02 paragraph 2 states appears to be a legal question that IFEU has been unable to resolve.

In addition to the above, the issue of **relative biological effectiveness (RBE)** of neutron radiation has been evaluated by various researchers in cytogenetic experiments. For example, human lymphocytes were exposed to a mixture of neutron and gamma radiation and the increase of dicentric chromosome aberration in cultured cells was determined (Heimers, 1999). Heimers concludes that “[t]he high RBE values of 96 and 113 respectively found in the present study indicate that the weighting factors for neutrons recommended by ICRP 60 (1990) are probably not conservative. Occupational exposure to neutrons may be more harmful than comparatively low values of physical measurements suggest”. It is recommended that the validity and relevance of this finding should be assessed in the review of neutron exposures from LBNL.

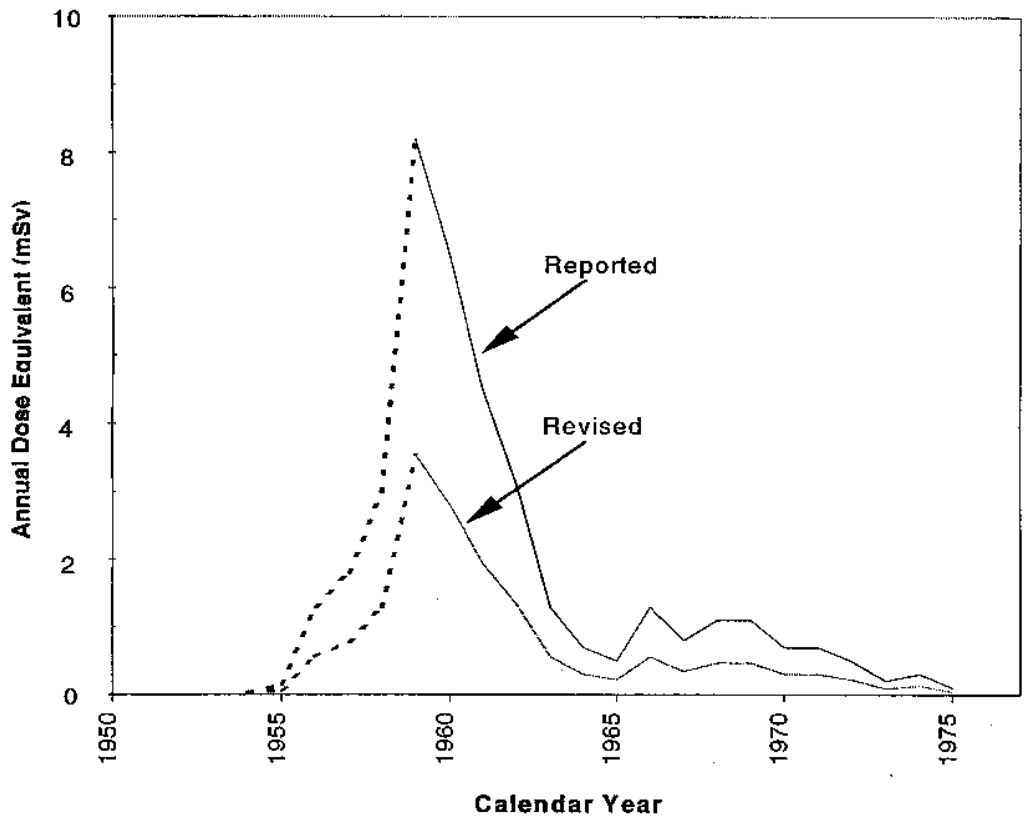

Figure 14. Comparison of reported dose equivalents for 1959-1975 with revised dose equivalents (Thomas et al., 2000)

Conclusions and recommendations

Neutron and gamma doses at various locations at the LBNL site boundary were substantially larger than today. Based on available data, maximum exposures have exceeded 500 mrem/yr using the historical conversion factors. Using current conversion factors for neutron doses, cumulative dose rates at the Olympus Gate station were greater than 2,000 mrem. It is recommended to estimate doses to the nearest residents including the contribution of all LBNL sources and pathways while taking uncertainties in monitoring data, conversion factors and other parameters into account. A recent paper (Heimers, 1999) presents cytogenetic data that suggests that neutron radiation may have a higher relative biological effectiveness (RBE) than is reflected in currently used radiation weighting factors. This paper and other data on the RBE of neutrons should be reviewed further.

C.2 What exposures resulted from past releases of tritium?

Approach
Review of historical data on tritium emission and environmental concentrations

Findings
Measurements of HTO in ambient air for three sampling locations are shown in Figure 15 for the time period 1969 to 1999 along with reported tritium emissions. Starting in 1995, emission data includes HT as well as HTO while before that, only HTO emissions were reported. Three observations can be made on this basis of this data. First, the ratio of reported tritium concentration in ambient air to reported releases is consistently larger for the 1970ies (>10 pCi/m^3 per Ci/yr) compared to the 1980ies and 1990ies (<10 pCi/m^3 per Ci/yr). Second, the concentrations at Building 3 (Calvin) were equal to or larger than levels measured at Lawrence Hall of Science (LHS). The peak concentrations in 1978 of 2,200 pCi/m^3 was more than a factor of 100 greater than the concentrations measured at LHS in 1999 and exceeded the current NESHAP compliance standard of 1,500 pCi/m^3. Third, reported concentrations at LHS and Olympus Gate were often equal to those reported for LHS while one would expect lower concentrations due to atmospheric dispersion.

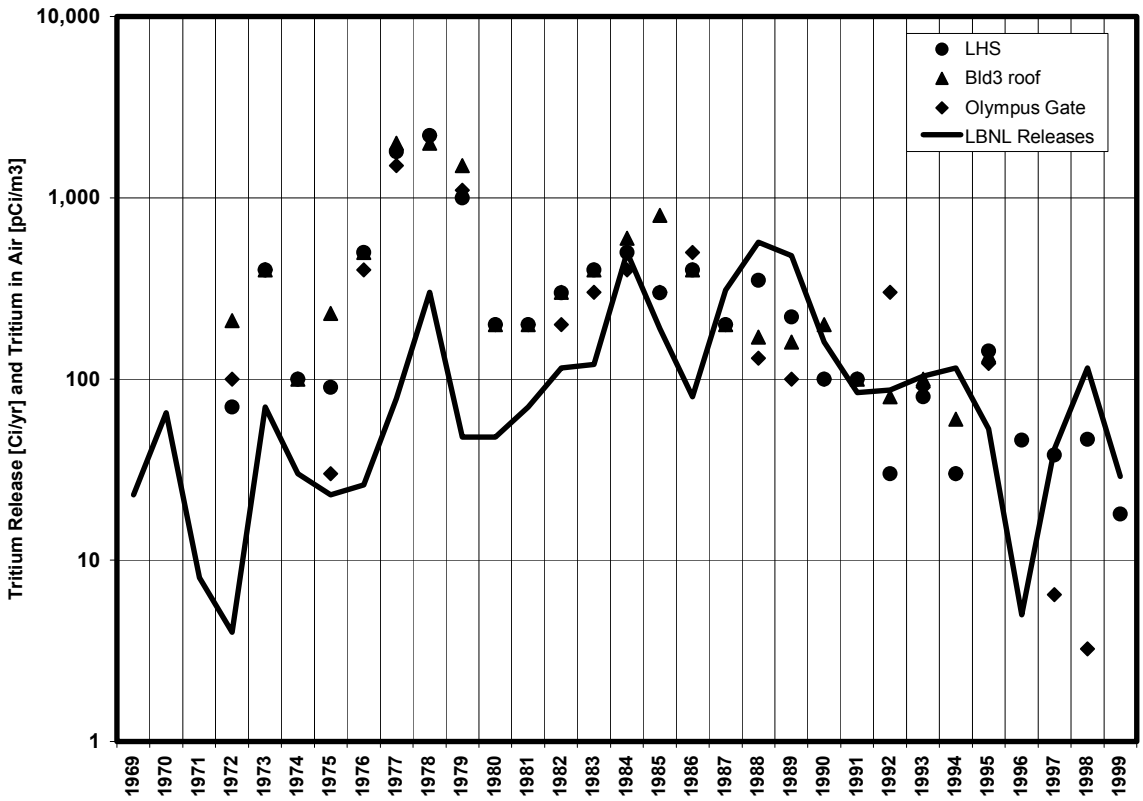

Figure 15. Reported annual tritium releases from NTLF, 1969 to 1999 and annual concentrations of HTO in air at environmental monitoring stations

In response to the data presented in Figure 15, LBNL pointed to the problem of detection limits and provided a series of Figures which suggest that annual ambient air samples at LHS and concentrations exceeded detection limits only for the years 1978, 1979, 1995, 1997, 1998 and 1999. While it is acknowledged that the issue of detection limits is very important, LBNL's presentation it has several errors.

First, the Annual Environmental Monitoring Report of the Lawrence Berkeley Laboratory for 1976 (LBNL, 1977) states that the "special air sampling program for ^{14}C and ^{3}H found detectable concentrations of these nuclides", contradicting LBNL's conclusion made in the year 2000 (LBNL, 2000). The contradiction is simple to resolve: for 1976, the annual average concentration for LHS was 500 pCi/m^3, the maximum weekly sample 3,600 pCi/m^3, while the detection limit was 700 pCi/m^3. Thus, while the calculated annual average is below the detection limit for individual samples, the activity in the maximum sample and maybe for more samples as well, were above the detection limit. Weekly raw data should thus be analyzed in detail before making conclusions about annual averages. Such analysis was apparently not done by LBNL in response to IFEU's June 2000 report.

Second, LBNL's Figure B erroneously reported annual average concentrations for the year 1977 at LHS, Olympus Gate and Building 3 roof to be less than 200 pCi/m^3. The annual monitoring report for 1977 indicates the following values: LHS: 1,800 pCi/m^3, Olympus Gate: 1,500 pCi/m^3, Building 3: 2,000 pCi/m^3. This appears to be an oversight in LBNL's presentation of the data and should be corrected.

Even if only those values are considered that LBNL recently declared to be above the detection limit, reported annual average air concentrations differ by a factor of ~100 for a given tritium release (Figure 16). The variability is likely to be caused by the year-to-year variability in meteorology, analytical errors, the impact of short-term releases and other factors.

Given these uncertainties, historical exposures due to tritium in ambient air should be evaluated in further detail based on raw data.

Conclusions and recommendations

Reported concentrations of HTO in ambient air peaked in 1978 (2,200 pCi/m^3). This value is more than a factor of 100 greater than the concentrations measured at LHS in 1999 and would exceed the current NESHAP compliance standard of 1,500 pCi/m^3, though it did not exceed the then-prevailing limit. Reported concentrations at LHS and Olympus Gate were often equal to those reported for LHS while one would expect lower concentrations due to atmospheric dispersion. The reliability of historical data is limited. It depends on uncertainties in sampling and analysis and should be evaluated further. It is recommended to estimate doses to the nearest residents including the contribution of all LBNL sources and pathways while taking uncertainties in monitoring data, conversion factors and other parameters into account.

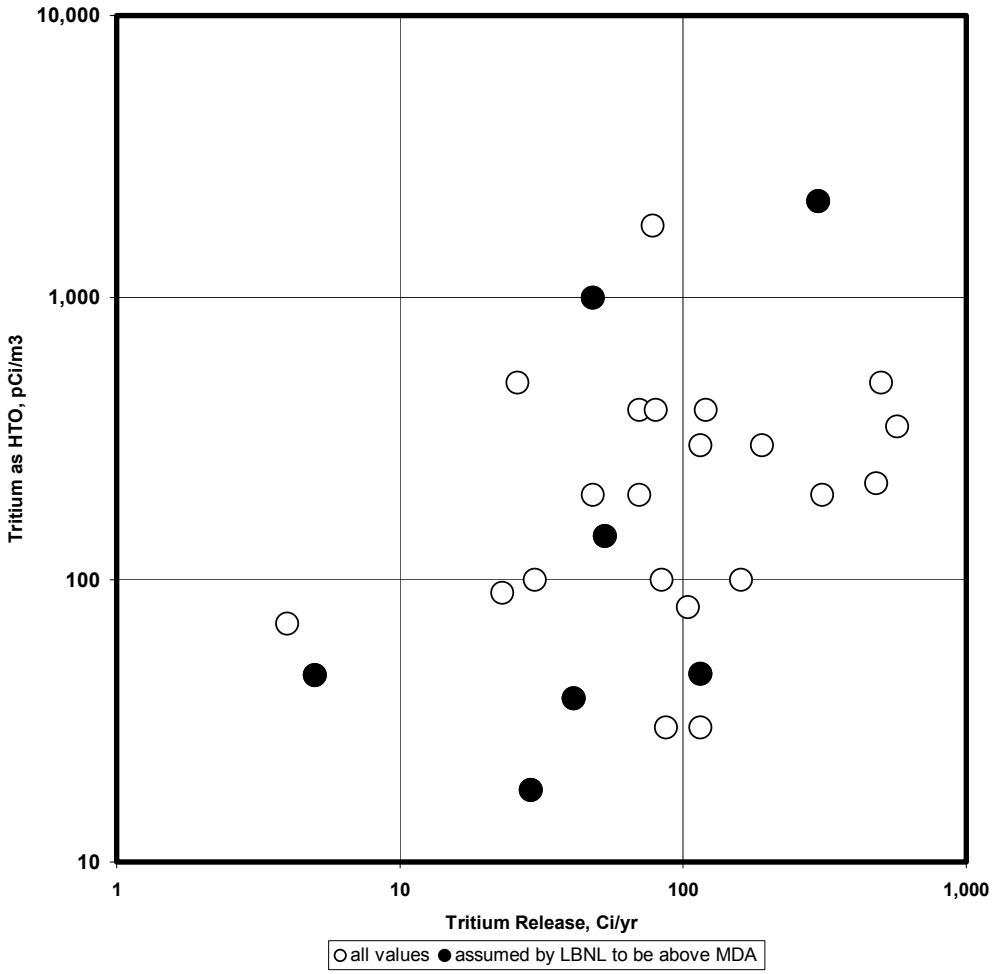

Figure 16. Reported annual tritium releases from NTLF, 1972 to 1999 and annual concentrations of HTO in Air at Lawrence Hall of Science (LHS)

C.3 What exposures resulted from past releases of radionuclides other than tritium?

Approach
Review of historical data on emissions and environmental concentrations

Findings
A variety of radionuclides other than tritium were used over the history of the LBNL laboratories, including alpha emitters such as plutonium. Measurements of alpha and beta activities were first reported in the environmental report for 1963. At that time, a total number of 75 stacks were sampled weekly for alpha and beta activity in particulate form. The numbers for the July-December 1963 (LBNL, 1963) were reported as follows for 2,809 stack samples:

Alpha activity (pCi/m^3):	0.005	average	0.58	maximum
Beta activity (pCi/m^3):	2.9	average	11,730	maximum

These results appear to be unreasonable for the reported average beta activity: if one out of 2,809 samples has a beta activity 11,730 pCi/m^3, the average should be 4.1 pCi/m^3 rather than the reported 2.9 pCi/m^3 even if all other samples have zero activity. However, the average of 0.001 pCi/m^3 for alpha activity and 2.79 pCi/m^3 for beta activity overall concentrations in the air of the local area (onsite sampling stations) was low and was not significantly different from sampling locations in greater distance. The values were also much lower than the historical maximum permissible concentration (MPC) for unknown alpha and beta activity in the annual average of 0.04 and 10 pCi/m^3, respectively.

IFEU performed a preliminary review of data for alpha and beta activity in ambient air for the time period 1963 to 1998. The reported annual averages for the sampling locations do not indicate significant impacts from releases at LBNL. Overall, reported alpha activity in ambient air was less than 0.001 pCi/m^3. Reported beta activity in ambient air was significantly larger in 1963 (5.7 pCi/m^3) and 1964 (1.2 pCi/m^3) than in subsequent years (less than 0.3 pCi/m^3) which is most likely the consequence of aboveground nuclear weapons tests. Additional support for that explanation is the fact that the concentrations for sampling points at local areas were not significantly different from those at perimeter areas.

A summary of reported releases of alpha and beta activity in liquid discharges into the sewer system (Hearst and Strawberry sewer) is shown in Table 6. The largest liquid discharges of alpha activity was reported for the year 1964 and for beta activity in 1974. The historical limits for unknown emitters in sewer discharges were reported as being exceeded only in 1974 where the annual average beta activity (without tritium) was 13,940 pCi/l in the Strawberry sewer, exceeding the applicable standard of 3,000 pCi/l. However, in 1974, when reported discharges of beta activity from LBNL into the sewer were larger, the beta activity was about 1000 times smaller (14 pCi/l). Either the reported discharges were overestimated by a factor of 1,000, or the reported concentration was underestimate by a factor of 1,000, documenting that the reliability of historical records has to be carefully reviewed.

Conclusions and recommendations

Reported concentrations of alpha and beta activity in air from 1963 onwards does not indicate significant impacts from releases at LBNL. Data for discharges into the sewer indicate that peak releases occurred in the 1970s. The limited review of the data indicated internal inconstancies. In a reconstruction of historical exposures, emphasis should be placed on a review of raw data. It is recommended to estimate doses to the nearest residents including the contribution of all LBNL sources and pathways while taking uncertainties in monitoring data, conversion factors and other parameters into account.

Table 6. Reported releases of radioactive materials from LBNL operations into the Hearst and Strawberry sewers

Year	Alpha activity (µCi)	Beta activity[b)] (mCi)	Tritium activity (Ci)
1963	44	0,1	
1964	1.390	12,9	
1965	109	6,8	
1966	229	16,0	
1967	176	6,3	
1968[a)]	14	2,9	
1969	15	8,6	
1970	90	14,0	
1971	197	8,5	
1972	360	7,8	
1973	<63	3,5	
1974	165	5.630,0	
1975	<106	19,0	
1976	255	181,0	
1977	96	98,5	
1978	353	3.260,0	
1979	196	774,0	
1980	149	29,4	
1981	<99	27,3	
1982	94	16,7	
1983	<68	9,8	
1984	11	19,0	
1985	30	20,0	<5,7
1986	<32	42,0	1,4
1987	<29	24,0	0,7
1988	<35	12,4	2
1989	<47	5,0	0,94
1990	<40	18,0	0,29
1991	<7	3,9	0,29
1992	<0.09	3,1	0,11

a) data for July-December 1968 only
b) gross beta without tritium

D Risk related questions

D.1 What is the potential health risk from past exposures?

Approach
Comparison of historical doses with doses at other sites

Findings
The calculated dose for continued residence at the Olympus Gate stations as estimated from data in Thomas et al. (2000) was about 2 rem CEDE. Doses for real individuals were smaller than 2 rem because a 100% residency at the Olympus Gate was hardly realistic. On the other hand, the uncertainty in the underlying raw data and the contribution from other radionuclides should be properly evaluated before accepting the Olympus Gate data by Thomas as an upper limit estimate for all residents near the LBNL site. Compared to other sites, doses in the LBNL vicinity are considered to be significant as the comparison of selected data in Table 7 shows. The comparison is difficult since the methodology of dose calculations and the selected scenarios are not identical. Despite these limitations, the data indicates that considerable attention has focused on the reconstruction of radiation doses at sites where exposures were similar to those at LBNL. While doses at the Hanford and Fernald sites were larger than those at LBNL, doses from releases at Rocky Flats Plant, Colorado were comparable or less.

Table 7. Maximum scenario radiation doses from past activities at selected locations

DOE Facility	Dose estimates (CEDE)	Remarks	Source
Lawrence Berkeley National Laboratory, CA	~ 2 rem	for continuous residence at Olympus Gate	Thomas et al., 2000
Rocky Flats, CO	maximum dose: < 1 rem	resulting from 1957 fire to	RAC (1999)
Nevada Test Site Fallout in Utah	average ~0.8 rem maximum: 4 rem	dose to 1,177 people diagnosed with leukemia	Lloyd et al. (1990)
Feeds Material Production Plant, Fernald OH	28 rem	realistic maximum inhalation exposure[a)]	Till et al. (1998)
Hanford WA, Site	2 to 105 rem[b)] (95% confidence interval)	maximum scenario: females born in 1945 living from Richland on a goat milk diet	Hoffman (1999)

a) Scenario 1, 38 years of exposure, mean dose value
b) weighting factor of 0.05 for conversion of thyroid doses to CEDE

Conclusions and recommendations
Radiation doses from past operations at LBNL were comparable to those at locations where considerable efforts were undertaken to reconstruct exposures to members of the public. In light of uncertainties regarding the magnitude and relative biological effectiveness of neutron exposures and the contribution from other radionuclides and non-radioactive pollutants, an in-depth review is recommended. A prerequisite for the risk assessment process involves dose reconstructions for past LBNL operations.

D.2 What is the potential health risk from current exposures of tritium?

Approach
Review of updated health risk assessment

Findings
IFEU will produce a third report on the results of the sampling plan, including a qualitative review of the LBNL's revised health risk assessment documents incorporating the results from additional sampling.

Conclusions and recommendations
Not available at this time. IFEU will produce a report on the results of the sampling plan, including a qualitative review of the LBNL's revised health risk assessment documents incorporating the results from additional sampling.

D.3 What is the risk in case of accidents, such as fire?

Approach
Review of LBNL Safety Analysis Document for NTLF

Findings
The stated purpose of the Safety Analysis Document (LBNL, 1996) LBNL is "to identify and analyze hazards at the National Tritium Labeling Facility (NTLF) for the purpose of establishing and evaluating the adequacy of safety bases to assure operation at a low risk level for workers and the public".

The accident scenarios involving the release of radionuclides with subsequent exposures to members of the public that were considered in the document are summarized in Table 8. The largest exposure was calculated for the facility fire in which the entire inventory of uranium (15,000 Ci) in the beds is assumed to be released in form of HTO, the chemical form of tritium resulting in the largest dose.

Table 8. Accidents involving radionuclides for which doses to members of the public were calculated in the Safety Analysis Document (LBNL, 1996b)

Parameters	**Valve breaks during loading of Tritium bed**	**Piping to Tritium bed sheared during seismic event**	**Facility fire**
Total Tritium Release (Ci)	2,000	10	15,000
HTO Release (Ci)	20	1	15,000
Release conditions	10 meters remote stack	ground level	38 meters due to plume rise
Maximum dose to member of the public	0.21 mrem at 500 meters	4.4 mrem at 60 meters	4.8 mrem at 1,100 meters

The calculated dose to the maximally exposed member of the general public is 4.8 mrem. The dose is almost identical to the one resulting from the seismic accident in which it is assumed that the piping to the tritium bed sheared, releasing 1 Ci of HTO.

How can the dose be almost identical even though the source term varies by more than a factor 10,000? The reason for the difference stated in the Safety Analysis Document is the plume rise due to the fire. It is assumed that the fire results in the release of $2x10^7$ BTU of heat, or the equivalent of 170 gallons of gasoline. The heat will cause the tritium plume to rise to a height of 38 meters. This assumption is subject to considerable uncertainty. It is certain that a fire will result in the release of heat. The question is whether the heat release is uniform during the entire accident. If the fire starts small and the tritium is oxidized at its beginning, the part of the plume containing tritium will rise to lesser degree than the plume during the rest of the fire. The probability of this scenario may be low, but is it zero?

For sake of discussion, let us assume that the same meteorological conditions prevail as was assumed by SENES for the maximum jogger scenario. SENES calculated that the dose to a female jogger 41 m downwind from the NTLF hillside stack of 0.042 to 0.26 mrem CEDE (90% confidence interval) for a release of 218 mCi. For a source term of 15,000 Ci, all other conditions being identical, the resulting dose would be between 2,900 to 18,000 mrem or 600 to 3,700 times larger than the one calculated in the Safety Analysis Document.

The Safety Analysis Document refers to DOE Standard 1027-92 which lists inventory threshold quantities of radioactive materials that, if released, could cause "significant localized consequences" which are given when the accidental dose at 30 m exceeds 10 rem (which equals 10,000 mrem). The Safety Analysis Document concludes on page 3: "The analysis shows that fiull release of the tritium inventory could not cause "significant localized consequences".

A cursory review of DOE-STD-1027-92 and the underlying model to calculate reporting values for hazardous substances according to 40 VFR 302.4 does not suggest that it is mandatory to use the specific model parameters that were selected in preparation of the Safety Analysis Document. It is therefore likely that a reevaluation of the accident scenario and parameters may significantly change the results and the conclusions.

Conclusions and recommendations

The adequate determination of the consequences of potential accidents at the NTLF is of particular importance to ensure that the facility is in compliance with DOE Standard 1027-92. The Safety Analysis Document concludes on page 3: "The analysis shows that full release of the tritium inventory could not cause "significant localized consequences", which are defined as accidental doses at 30 m exceeding 10 rem (which equals 10,000 mrem).

The preliminary review indicates that this claim may be incorrect. Parameters in the Safety Analysis Document were selected without assessing that the resulting doses are realistic for the whole array of potential scenarios. This is evidenced by the comparison of doses calculated in the Safety Analysis Document for the worst accident (a fire at NTLF releasing 15,000 Ci of HTO) with results from alternative calculations. While the Safety Analysis Document concludes that the maximum off-site exposure is 4.8 mrem at a distance of 1,100 meters, doses would be between 2,900 to 18,000 mrem using the "jogger scenario" from the SENES Inc. report. This assumes that the tritium is released from the stack with no plume rise from the fire; conditions which could prevail if HTO is released at the onset of a fire.

An independent evaluation of the assumptions underlying the scenarios, the calculation model and its parameters is lacking. It is therefore recommended that an independent reassessment of consequences from accidents at NTLF be performed.

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Appendix A: Scope of services

1. Work Plan

1.1. Contractor shall interview LBNL technicians and scientists, subcontractors, regulators, City officials, and representatives of the community to generate a list of concerns for evaluation. Identify the data quality objectives (DQO) from the list of concerns.

1.2. Contractor shall review sufficient representative data on present and past emissions and environmental monitoring by LBNL of soils, air, subsurface and ground waters, and plants (including raw data) in order to arrive at conclusions about both the quality of the data collected and the analyses of that data.

1.3. Contractor shall review and comment on the LBNL/U.S. Department of Energy (DOE) tritium sampling work plans for air, water, soils, plant, and other media.

1.4. Contractor shall review the adequacy and accuracy of LBNL's past monitoring, analyses, and modeling.

1.5. Contractor shall review and comment on the appropriateness of models used for calculations of doses and risks, including confirmatory sampling calculations, as necessary.

1.6. Contractor shall provide recommendations on future data collection and further investigations at LBNL, including dose reconstruction, if necessary.

1.7. Contractor shall review and provide a written report on the revised health risk assessment documents that are expected to be produced by LBNL/DOE at the end of the process.

1.8. Deliverables:

1.8.1. Report #1: Contractor shall produce a **Preliminary Technical Report** on the evaluation of past and present emissions and environmental data. The report shall include a review of the identified DQO and a review of the sampling plans.

1.8.2. Report #2: Contractor shall produce a **Draft Final Scoping Report**, which shall include a review of the appropriateness of models used for the calculations of doses and risks, with recommendations for further investigations, if warranted. The report shall be submitted to the City, which shall be responsible for distribution of the report to stakeholders, as well as for compiling all written comments for transmittal to Contractor.

1.8.3. Evaluation of Comments: Contractor shall review the written comments on the draft reports (both those submitted directly to Contractor and those compiled by the City of Berkeley) and revise the reports as necessary. The result of this task shall be the **Final Scoping Report**. Contractor shall submit this report to the City for distribution to the stakeholders.

1.8.4. Report #3: Contractor shall produce a report on the results of the sampling plan, including a qualitative review of the LBNL's revised health risk assessment documents. The report shall be presented to the City, which shall distribute it to the stakeholders. Written comments received by Contractor or collected by the City will be reviewed and updates made as appropriate.

1.8.5 Presentation. A presentation of the **Final Scoping Report and Report #3** (Deliverables 1.8.3. and 1.8.4.) shall be made before the Berkeley City Council by Contractor and others.