UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
WASHINGTON, D.C. 20460

April 29, 1993

OFFICE OF THE ADMINISTRATOR
SCIENCE ADVISORY BOARD

EPA-SAB-RAC-93-010

Honorable Carol M. Browner
Administrator
U.S. Environmental Protection Agency
401 M Street, S.W.
Washington, D.C. 20460

Subject: Science Advisory Board Review of the Release of Carbon-14 in Gaseous Form from High-Level Waste Disposal

Dear Ms. Browner:

At the request of the Office of Radiation Programs, the High-Level Waste/Carbon-14 Subcommittee of the Science Advisory Board's Radiation Advisory Committee reviewed the issues document, *Issues Associated with Gaseous Releases of Radionuclides for a Repository in the Unsaturated Zone*. Although the framework for the model used in the issues document to calculate carbon-14 release is conceptually valid, the technical basis for a number of the critical assumptions, parameters and parameter ranges adopted in the application of the model is not clear and leads to results that are biased in the direction of higher releases to the environment as well as to underestimates of the uncertainties. The Subcommittee considers the following to be its most significant findings and recommendations.

a) Releases of carbon-14 from a repository may produce an appreciable global population dose over 10,000 years, but the average individual dose would be very low. For a reasonable upper bound release of half the carbon-14 initially contained in a repository, the global population dose over 10,000 years is estimated to be 14 million person rem, and the corresponding average individual lifetime dose would be about 0.01 mrem. Based on the EPA's preliminary risk factor for carbon-14, these doses correspond to calculated lifetime individual risks of 3 x 10^{-9}, and population risks of less than one fatality every two years on average, or 4,000 cancer fatalities world wide over 10,000 years. Whether or not these doses constitute a public health concern is a fundamental issue of principle. The Subcommittee did not try to resolve this issue, but EPA must address it when considering carbon-14 releases. Consistent with the report Reducing Risk, the Subcommittee recommends the predicted individual and population doses be considered in comparison with doses from other sources, with dose limits in other standards, and with other environmental and radiation risks.

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b) The uncertainty analysis performed in the issues document is in a preliminary state and can be improved substantially. When the broader uncertainty bands for various parameters are considered, the overall uncertainty regarding the potential magnitude of carbon-14 releases is quite broad. Consequently it is not possible on the basis of presently available information to predict with reasonable confidence whether releases from an unsaturated repository would be less than or greater than the Table 1 (40 CFR 191) release limits. (The Table 1 release limit is one-tenth of the inventory.)

c) The issues document does not accurately characterize the potential for gaseous carbon-14 releases from the repository to the environment, although the Subcommittee notes it may not be possible to do so based on currently available information. The EPA document's assumptions about release mechanisms and rates of release from the wastes and containers, and about transport mechanisms and rates, do not appear to be supported by sound technical justifications.

d) The description of the effectiveness of engineered barriers designed to reduce or impede releases is not adequate because there has been little research and development of engineered barriers specifically designed to contain carbon-14 in an unsaturated repository. The issues document assumed that such devices contributed little to the containment of carbon-14. Delaying the release of carbon-14 from the waste containers or containment area would allow time for radioactive decay, which would reduce the ultimate release to the environment. (The significance of the reduction in the release would depend on the containment time relative to the 5,730 year half-life of carbon-14.) Therefore, the Subcommittee encourages investigation of the use of multiple barriers to retard the migration of carbon-14 to the accessible environment. In a presentation to the Subcommittee by its contractor, DOE indicated that it is possible that a multilayer waste container could contain the carbon-14 long enough to meet the EPA limit, but that it would not be possible to verify that the container would be gas tight for 10,000 years. The Subcommittee agrees with the issues document that the potential costs and benefits of an upgraded waste container or additional engineered barriers have not been firmly established, and the Subcommittee was unable to agree on the technical feasibility and effectiveness of improved barriers.

e) EPA needs to revise the description in the document of the physical and chemical retardation and transport of carbon-14 from the waste repository to the surface, because the hypothesis that the principal transport mechanism in flat terrain would be diffusion is incorrect. This hypothesis leads to the erroneous conclusion that carbon-14 transport could be greatly reduced by locating an unsaturated

repository in flat terrain. Temperature effects from a heated repository likely would cause the advective component to be dominant under almost any reasonable scenario.

f) Although the document's estimate of the amount of carbon-14 in unreprocessed spent nuclear fuel of 1 curie/metric tonne of heavy metal (Ci/MTHM) appears reasonable, the estimate of the total inventory of carbon-14 in the repository (100,000 curies) should be changed to 70,000 curies because the Congress has limited the first repository's capacity to 70,000 MTHM. Such a change, however, would have no effect on the calculation of the ratio of carbon-14 releases to the release limit in 40 CFR 191.

In responding to the broader issue of risk reduction, the Subcommittee notes that optimizing site selection on the basis of a single criterion may cause a change in optimal conditions for other criteria. For example, carbon-14 releases to the accessible environment would probably be less from a saturated site than from an unsaturated site, but risks from other radionuclides may be greater or smaller depending on a number of factors.

We appreciate having been given the opportunity to conduct this particular review. We request that you provide us with a formal response to our advice, particularly with respect to the issues of engineered barriers, quantitative uncertainty analysis and risk reduction we have raised.

Sincerely,

Raymond C. Loehr

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A-101
Washington, DC

EPA-SAB-RAC-93-010
April 1993

EPA

AN SAB REPORT: REVIEW OF GASEOUS RELEASE OF CARBON-14

REVIEW, BY THE RADIATION ADVISORY COMMITTEE, OF THE RELEASE OF CARBON-14 IN GASEOUS FORM FROM HIGH-LEVEL WASTE DISPOSAL

U.S. ENVIRONMENTAL PROTECTION AGENCY

NOTICE

This report has been written as a part of the activities of the Science Advisory Board, a public advisory group providing extramural scientific information and advice to the Administrator and other officials of the Environmental Protection Agency. The Board is structured to provide balanced, expert assessment of scientific matters related to problems facing the Agency. This report has not been reviewed for approval by the Agency and, hence, the contents of this report do not necessarily represent the views and policies of the Environmental Protection Agency, nor of other agencies in the Executive Branch of the Federal government, nor does mention of trade names or commercial products constitute a recommendation for use.

ABSTRACT

At the request of EPA's Office of Radiation Programs, the High-Level Waste/Carbon-14 Subcommittee of the Science Advisory Board's Radiation Advisory Committee met June 16-17, August 3-4, and September 9-10, 1992 to review, "Issues Associated with Gaseous Releases of Radionuclides for a Repository in the Unsaturated Zone".

The Subcommittee's findings and recommendations address the inventory of carbon-14, the characterization of the mechanisms and release rates for gaseous carbon-14 from the wastes and waste containers, the description of the effectiveness of engineered barriers designed to reduce or impede releases, the description of the physical and chemical retardation and transport of carbon-14 from the waste repository to the surface, quantitative uncertainty analysis, the dichotomy of small individual doses and large population doses, and the need to consider the release of all radionuclides when seeking to optimize site selection

Key Words: High-Level Radioactive Waste, Carbon-14, Uncertainty Analysis, Radionuclides, Risk Reduction, Standards, Release Limits, Containment, 40 CFR 191.

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1. EXECUTIVE SUMMARY

At the request of the Office of Radiation Programs, the High-Level Waste/Carbon-14 Subcommittee of the Science Advisory Board's Radiation Advisory Committee reviewed the issues document, "Issues Associated with Gaseous Releases of Radionuclides for a Repository in the Unsaturated Zone". The EPA also provided two related documents--"Position on the Potential for Gaseous Release from a High-Level Waste Repository" and "Summary of EPA/Office of Radiation Programs Carbon-14 Dosimetry as used in the Analysis for High Level and Transuranic Waste". The Subcommittee's findings and conclusions are summarized below. The number at the end of each finding refers to the section of the Subcommittee report in which a more detailed discussion of the finding is found.

Question 1: Carbon-14 is present in the waste; how and how fast carbon-14 is converted to carbon-14 dioxide gas (or other gases) may influence its release to the atmosphere. Does the EPA document accurately summarize the total inventory of carbon-14 present and the fraction that could potentially be released from the repository via the air pathway?

The estimate of the amount of carbon-14 in unreprocessed spent nuclear fuel of 1 curie/metric tonne of heavy metal appears reasonable. (3.1.1)

The estimate in the issues document of the total inventory of 100,000 curies of carbon-14 in the repository should be changed to 70,000 curies, because Congress has limited the capacity of the first repository to 70,000 MTHM unless a second repository is approved. (3.1.2)

The fraction of carbon-14 that potentially could be released from the waste containers via the air pathway is uncertain. The assumption that 100% of the carbon-14 in the waste containers is available to be oxidized to become gaseous carbon-14 dioxide does not appear to be based on any documented evidence. (3.1.3)

Removal or reduction of carbon-14 from the inventory does not appear to be a promising approach. (3.1.4)

Question 2: Does the Agency's document accurately characterize the mechanisms and release rates for gaseous carbon-14 from the wastes and canisters?

No. The issues document contains a set of assumptions about release mechanisms and rates that appear to be biased toward the high side. However, it is not clear that these mechanisms and release rates can be more accurately characterized based on current information, measurements and other data and still be applicable for a generic repository. (3.2.1)

The assumed long-term release rate of 10^{-4} per year does not have a solid scientific foundation. The total carbon-14 released to the environment is highly

dependent upon the assumed rate of long-term release from the repository system. (3.2.2)

Predictions of the near-field chemical and physical environment and its effects on the integrity of the waste container are uncertain. (3.2.3)

The assumption that 5% of the carbon-14 inventory would be released when the waste container fails appears somewhat on the high side of measured values. (3.2.4)

Question 3. Does the Agency's document accurately describe the effectiveness of engineering barriers designed to reduce or impede releases?

The description of the effectiveness of engineered barriers designed to reduce or impede releases is not adequate because there has been little research and development of engineered barriers specifically designed to contain carbon-14 in an unsaturated repository. (3.3.1)

The possibilities for engineered barrier designs should be further explored. (3.3.2)

The Subcommittee was unable to agree on the technical feasibility and effectiveness of improved barriers to help impede or retard the migration of carbon-14 dioxide to meet the EPA release limits. (3.3.3)

Question 4: Does the Agency's document adequately describe the physical and chemical retardation and transport of carbon-14 from the waste repository to the surface?

The model used in the issues document to calculate carbon-14 transport is conceptually valid. However, the technical basis for a number of the critical assumptions, parameters and parameter ranges adopted in the application of the transport model is not clear and leads to results that are biased in the direction of higher release to the environment as well as underestimated uncertainties. (3.4)

The hypothesis that the principal transport mechanism in flat terrain would be diffusion is incorrect. This hypothesis leads to the erroneous conclusion that carbon-14 transport could be greatly reduced by locating an unsaturated repository in flat terrain. However, temperature effects from a heated repository likely would cause the advective component to be dominant under almost any reasonable scenario. (3.4.1)

Transport of carbon-14 from a repository in the saturated zone to and across the water table is feasible and should be addressed in the document (3.4.2).

Average gas travel times for the case of elevated terrain in the issues document have probably been under-estimated. (3.4.3)

For an evaluation of generic repository sites, information on rock type and topography (flat vs. elevated terrains) as presented in the issues document are not directly relevant site factors. The parameters which should be varied in the sensitivity and uncertainty analyses are intrinsic permeability, air porosity, retardation factor, and effects of geological heterogeneities. (3.4.4)

A retardation factor of 1 for iodine-129 transport as used in the issues document is probably reasonable. However, the possibility that no gaseous iodine may migrate should be acknowledged. The gaseous release of technetium was not considered in the issues document. The Subcommittee agrees that it is not likely that gaseous technetium oxide (Tc_2O_7) could migrate in the presence of water. (3.4.5)

The range for retardation factors for carbon-14 used in the sensitivity analyses should be expanded to take into account the role of metal oxides. If this is done, then it is likely that the retardation factors will be identified by the sensitivity analyses as a parameter to which the release results are sensitive. This is counter to the conclusion in the issues document. (3.4.6)

The issues document needs to recognize the uncertainties associated with the effects of thermal loading of a repository. (3.4.7)

Not enough is known to support or rule out the conclusion that an unsaturated-zone repository site could be found where geological barriers could be shown to be capable of holding carbon-14 releases below the regulatory limits. (3.4.8)

Question 5: Is the Agency's assessment of the magnitude of the release resulting from the factors identified in questions #2, #3, and #4 complete, correct, and clear?

The estimate of carbon-14 releases calculated in the issues document is consistent with the assumptions made and the parameter values adopted. (3.5.1)

The base case analysis of the issues document is based on assumptions that appear to lead to a high estimate of the amount of carbon-14 that would be released. (3.5.2)

The measures used to describe the magnitude of potential releases of carbon-14 are incomplete and insufficiently informative. (3.5.3)

It is likely that EPA has overestimated the maximum individual exposures by a factor of 10 or more. (3.5.4)

Question 6: Does the Agency's document adequately describe the uncertainties associated with the assessment of the magnitude of the release?

The uncertainty analysis performed in the issues document is in a preliminary state and can be improved substantially. (3.6.1)

The objective of the uncertainty analysis should be clearly stated. (3.6.2)

Key uncertainties are underestimated, and some are not discussed. (3.6.3)

Selection of a limited range of parameter values predetermines the outcome of the sensitivity analyses. (3.6.4)

The sensitivity analysis performed in the issues document is a one-at-a-time perturbation of parameter values. Such sensitivity analyses may give misleading results when the model represents a non-linear system. (3.6.5)

When the broader uncertainty bands recommended in this report for various parameters are considered, the overall uncertainty regarding the potential magnitude of carbon-14 releases is quite broad, and spans ratios of the estimated to allowable release ranging from zero to as high as ten. (3.6.6)

The release ratio calculation is for releases within 10,000 years. The result of this analysis is not sensitive to this 10,000 year cutoff. (3.6.7)

The issues document should distinguish between predicting the occurrence of rare events and performing an uncertainty analysis about a "best estimate". (3.6.8)

The Subcommittee notes that the uncertainties in the estimates of risk from carbon-14 releases presented by EPA staff are of comparable magnitude to the uncertainties associated with the estimation of the release of carbon-14 over 10,000 years. (3.6.9)

Within the context of its charge, the Subcommittee offered the following comments relative to the Science Advisory Board's document, Reducing Risk: Setting Priorities and Strategies for Environmental Protection. (3.7)

It is inappropriate to consider the relative performance of different generic repository settings on the basis of carbon-14 alone. (3.7.1)

For a proper interpretation of the effectiveness of risk management decisions, EPA should compare the health and environmental risk posed by a repository, including releases of carbon-14, with the risks posed by other environmental problems. (3.7.2)

From a risk management perspective, a key point to understand about doses and risks from repository releases of carbon-14 is that the perceived significance of the issue hinges upon whether one views the risks from the perspective of an individual or a population. (3.7.3)

2. INTRODUCTION

2.1 Background for the EPA's Analysis

The EPA promulgated standards for the disposal of high-level waste, spent fuel, and transuranic waste in 1985. A Federal Court remanded portions of these standards (40 CFR Part 191) in 1987. To satisfy the Court's ruling, EPA has been revising and updating the standard. The earlier analysis did not consider the potential for gaseous releases from high-level waste disposal (except as a consequence of volcanic eruptions), but it is not certain whether unsaturated sites can comply with the standard with respect to gaseous releases of carbon-14.

2.2 Procedural Aspects of the Science Advisory Board's Review

At public meetings held on June 16-17, August 3-4, and September 9-10, 1992, the High-Level Waste/Carbon-14 Subcommittee of the Science Advisory Board's Radiation Advisory Committee reviewed the issues document, "Issues Associated with Gaseous Releases of Radionuclides for a Repository in the Unsaturated Zone". The Office of Radiation Programs also provided "Position on the Potential for Gaseous Release from a High-Level Waste Repository" and "Summary of EPA/Office of Radiation Programs Carbon-14 Dosimetry as used in the Analysis for High Level and Transuranic Waste". The Subcommittee's review was guided by a charge developed with EPA's Office of Radiation Programs consisting of six questions. The Radiation Advisory Committee approved this Subcommittee review, with minor changes, at a public meeting held on October 29-30, 1992 and the Executive Committee approved the report

2.3 Perspective on the Review

The following critique is made in light of the perceived purpose of the EPA document(s). The Subcommittee assumes a) the purpose is to evaluate the expected performance of a geologic repository with respect to predicted releases of gaseous carbon-14 to the land surface; and b) this evaluation is to be conducted over the full range of conditions likely to be encountered at possible unsaturated zone sites. The Subcommittee understands that these results will be used by the EPA to evaluate the extent to which any site may or may not comply with the Table 1 limit for carbon-14 releases. Consequently, these comments address those aspects of the EPA document(s) which most directly determine the validity and results of the evaluation. The Subcommittee notes that it is difficult to assess the technical and policy aspects of the issues document without being influenced by the information gained from the Yucca Mountain site characterization activities.

3. FINDINGS AND DETAILED DISCUSSION RESPONDING TO THE QUESTIONS IN THE CHARGE

This section is organized according to the questions in the charge.

3.1 Question 1: Carbon-14 is present in the waste; how and how fast carbon-14 is converted to carbon-14 dioxide gas (or other gases) may influence its release to the atmosphere. Does the EPA document accurately summarize the total inventory of carbon-14 present and the fraction that could potentially be released from the repository via the air pathway?

3.1.1 The estimate of the amount of carbon-14 in unreprocessed spent nuclear fuel of 1 curie/metric tonne of heavy metal (Ci/MTHM) (section 2.1.2 of the issues document) appears reasonable.

Estimates of the concentration of carbon-14 in unreprocessed spent fuel are cited in the issues document as 1.0 Ci/MTHM (Van Konynenburg, 1989) and 1.5 Ci/MTHM (DOE, 1988). Both are based on a burnup of 33 000 MWd/MTHM. The former value is supported by measured data, which differ from the calculated result by ± 25%.

3.1.2 The estimate in the issues document of the total inventory of 100,000 curies of carbon-14 in the repository (issues document section 2.1.2 and 3.4.1) should be changed to 70,000 curies, because Congress has limited the capacity of the first repository to 70,000 MTHM unless a second repository is approved.

This change will not affect the ratio of calculated releases to the release limit for a repository, because the release limit is expressed on the basis of curies/MTHM.

The assumed inventory includes 7,000 MTHM equivalent of defense HLW which has negligible carbon-14 because reprocessing of this waste has already oxidized the carbon and released it to the atmosphere as carbon-14 dioxide. (A maximum residue of 10% carbon-14 in the reprocessed defense waste would only increase the inventory by 1%, because the defense waste is only 10% of the total.) However, a reduction of 7,000 MTHM in the inventory due to the defense HLW may be offset by an expected increase in the carbon-14 content of future spent fuel due to the trend towards longer burnup time.

Since 2/3 of the HLW slated for the repository has yet to be generated, changes in future fuel design and reactor design and operation could change the amounts of carbon-14 to be considered. Although increased average burnup

estimates have been factored into the inventory calculations, actual burnup times may differ from the estimates.

3.1.3 The fraction of carbon-14 that potentially could be released from the waste containers via the air pathway (presented in issues document section 2.1.2) is uncertain. The assumption that 100% of the carbon-14 in the waste containers is available to be oxidized to become gaseous carbon-14 dioxide does not appear to be based on any documented evidence.

The chemical form of carbon-14 in the fuel is unknown; it may exist as elemental carbon, a carbide or an oxycarbide. It may be present in a chemical form that would resist oxidation and remain immobile. The initial form is important to establish because most of the carbon-14 is believed to lie within the fuel matrix. It would be helpful to know the chemical form of carbon in the fuel matrix, and it may be experimentally feasible to obtain that information.

3.1.4 Removal or reduction of carbon-14 from the inventory does not appear to be a promising approach.

Three approaches to reduce the inventory of carbon-14 in the repository have been considered: a) reduction of nitrogen-14 in the fresh fuel going into reactors, b) heating spent fuel to drive off the surface fraction of carbon-14, and c) reprocessing. Reduction of nitrogen in the fresh fuel below present levels does not appear to be technically practical, given that these levels are low and that ammonia-based processing of uranium will leave a certain residual that will be difficult to remove. The Subcommittee agrees with the issues document that heating to remove carbon-14 is not an attractive option. Heating could potentially remove perhaps 2% of the carbon-14 from the exterior of the spent fuel, but at some probable detriment to the long-term effectiveness of the cladding and fuel as a barrier to the release of radioactivity. Unless the surface carbon-14 were captured when removed, heating would result in more rapid release of carbon-14 to the environment and result in greater exposure to the public. Consequently, the Subcommittee does not recommend heating the fuel to remove carbon-14. Reprocessing has not been considered by the Subcommittee because of the many other issues involved. In the Subcommittee's view, decisions regarding reprocessing of spent fuel are not likely to be made on the basis of carbon-14.

In addition to consideration of carbon-14, the Subcommittee concluded that the value cited for iodine-129 of 3.13×10^{-2} curies/MTHM (issues document section 2.1.2) appears reasonable. This value is based on results of calculations with the ORIGEN2 computer code for 10 year old fuel for a PWR with a fuel burnup of 33,000 Mwd/MTHM. The Subcommittee sees no reason to question the value. The assumption that all the iodine will be present as a diatomic gas leads to a high release estimate, but not enough is known to reduce the value.

3.2 Question 2: Does the Agency's document accurately characterize the mechanisms and release rates for gaseous carbon-14 from the wastes and canisters?

3.2.1 No. The issues document contains a set of assumptions about release mechanisms and rates that appear to be biased toward the high side. However, it is not clear that these mechanisms and release rates can be accurately characterized based on current information, measurements and other data, and still be applicable for a generic repository.

The Subcommittee bases this conclusion on the following findings:

a) The waste containers have not yet been designed, nor have their construction materials been selected.

b) The waste container environment, which influences corrosion and oxidation of the container's materials, has not been well defined for an unsaturated site.

c) The limiting container corrosion modes and other degradation modes over these time periods of thousands of years have not been established.

3.2.2 The assumed long-term release rate of 10^{-4} per year does not have a solid scientific foundation. The total carbon-14 released to the environment is highly dependent upon the assumed rate of long-term release from the repository system.

The issues document used 10^{-4} per year as a long-term release rate. An upper bound is based on an observed rate of 10^{-3} per year for spent fuel rods in contact with water and, on the assumption that the release rate of carbon-14 will be lower when the waste is not in contact with water. The lower bound was based on the 10^{-5} per year release limit established by the Nuclear Regulatory Commission in 10 CFR 60.113 as a limit to aqueous releases from vitrified wastes in a saturated repository. No justification is provided for selecting 10^{-4} per year as the long term release rate. Because of the importance of release modes, rates and time period, EPA should choose and defend such assumptions on scientific merit and not use an NRC regulatory value, even as a bound. Having said this, the Subcommittee acknowledges that it knows of no solid justification for any selected estimate or range for the long term release rate.

3.2.3 Predictions of the near-field chemical and physical environment and its effects on the integrity of the waste container (as presented in issues section 2.1.3) are uncertain.

Predicting the rates of corrosion of the waste containers and cladding as a function of time and subsequent oxidation of the carbon-14 in the fuel matrix without detailed knowledge of the environment (composition, humidity) is a very difficult venture. Because the near field environment is highly uncertain, the Subcommittee thinks that the waste container life is also highly uncertain. The assumption that waste containerss fail at either 300 or 1,000 years does not reflect the broad uncertainty in this critical performance parameter.

The thermal environment in the repository can be controlled by the areal power density. By maintaining this environment above the boiling point of water for 10,000 or more years, liquid water corrosion could be eliminated (Buscheck and Nitao, 1992). This hypothesis has not been experimentally demonstrated and more study is needed.

3.2.4 The assumption that 5% of the carbon-14 inventory would be released when the waste container fails appears somewhat on the high side of measured values.

The issues document cites measured values in the range from 1 to 10% as that part of the inventory immediately released when the waste container fails. The average of 5% appears without justification. An evaluation of the data contained in the literature indicates that a value of 2% would be more appropriate for unsaturated conditions. (Van Konynenburg, 1989)

3.3 Question 3. Does the Agency's document accurately describe the effectiveness of engineering barriers designed to reduce or impede releases?

3.3.1 The description of the effectiveness of engineered barriers designed to reduce or impede releases is not adequate because there has been little research and development of engineered barriers specifically designed to contain carbon-14 in an unsaturated repository.

The issues document assumed that such barriers contributed little to the containment of carbon-14. Delaying the release of carbon-14 from the waste containers or containment area would allow time for radioactive decay, which would reduce the ultimate release to the environment. Therefore, the Subcommittee encourages investigation of the use of multiple barriers to retard the migration of carbon-14 to the accessible environment.

3.3.2 The possibilities for engineered barrier designs should be further explored.

Upgrading the integrity of the waste containers with respect to carbon-14 releases has primarily been limited to the use of alternative materials to provide greater isolation and reduce degradation rates. Not addressed or considered are options by which the chemical environment of the inside of the waste container could be controlled for a period of time on the order of the half-life of carbon-14 or greater. Alternatively, chemical barriers in the form of carbon dioxide getters might be incorporated into backfill materials. The use of multiple engineered barriers to retard the migration of carbon-14 to the accessible environment should be investigated more aggressively. The importance of multiple barriers to help prevent the release of disposed HLW was emphasized in the 1985 standards due to the inherent uncertainty associated with any one barrier (natural or man-made) over the long time periods involved. The Subcommittee agrees with the issues document that the potential costs and benefits of an upgraded waste container or additional engineered barriers have not been firmly established.

3.3.3 The Subcommittee was unable to agree on the technical feasibility and effectiveness of improved barriers to help impede or retard the migration of carbon-14 dioxide to meet the EPA release limits.

This was the most controversial issue considered by the Subcommittee. "Getters" have the potential to help retard or prevent the migration of carbon-14 dioxide. Similarly they can take up water and oxygen, both of which are agents of corrosion. The issues document recognized the potential for getters to reduce releases, but offered no guidance for utilizing them. Concerns with the use of getters are their possible effects on other aspects of the radionuclide containment system and the present inability to predict behavior for 10,000 years, especially in an elevated temperature environment.

3.4 Question 4: Does the Agency's document adequately describe the physical and chemical retardation and transport of carbon-14 from the waste repository to the surface?

The model used in the issues document to calculate carbon-14 transport is conceptually valid. However, the technical basis for a number of the critical assumptions, parameters and their ranges adopted in the application of the transport model is not clear and leads to results that are biased in the direction of higher releases to the environment as well as underestimates of the uncertainties.

The document did not provide an adequate review of the underlying scientific knowledge about Yucca Mountain or explore the range of conditions that might be found at other unsaturated sites. Proper treatment of these issues could conceivably change the results of the release-time calculations and the overall conclusions made or implied in the document.

The technical and policy aspects of the issues document are admittedly difficult to assess without being influenced by the information gained from the Yucca Mountain site characterization activities. Nonetheless, with respect to carbon-14 transport, it is inappropriate to compare the performance of Yucca Mountain or any other actual site against that of generic or composite sites, as was done in the document, because uncertainties in the conceptual process models are greater for a specific site than for a generic or composite site as typically modeled. As one learns more about a site through site characterization activities, conceptual process models generally become more complex in order to deal with the geological heterogeneities that are discovered. Consequently, in practice, conceptual process models are typically less complex for generic sites than for actual sites. The opposite should be the case; generic models should have greater uncertainties in their conceptual process models to encompass the range of conditions expected at actual sites.

Subsections 3.4.1 through 3.4.8 discuss the Subcommittee's major concerns regarding the assumptions and parameter values used in the transport modeling, and the approach taken in the issues document to develop estimates of the range of carbon-14 release rates to be expected from generic repository sites in the unsaturated zone.

3.4.1 The hypothesis (found in the issues document section 2.2.4) that the principal transport mechanism in flat terrain would be diffusion is incorrect. This hypothesis leads to the erroneous conclusion that carbon-14 transport could be greatly reduced by locating an unsaturated repository in flat terrain. However, temperature effects from a heated repository likely would cause the advective component to be dominant under almost any reasonable scenario.

The main driving force for gas flowing upward from a nuclear-waste repository will be the decay heat of the waste. Calculations for the topography of Yucca Mountain show that heating the repository by 15°C (and assuming that rock temperatures around the repository have reached steady state) reduces travel times by somewhat more than a factor of 2 [Ross et al., 1992]. Thus 15°C of heating provides a stronger driving force for gas flow than the topographic effect observed at Yucca Mountain under present-day conditions (see Subcommittee report section 3.4.4(d)). The heating of a repository is likely to lead to a temperature increase of at least 15° for the entire 10,000 year period; Buscheck and Nitao [1992] found such an increase to hold true for a wide range of thermal loadings. Therefore the gas travel time for a repository in flat terrain would be

shorter, over the entire 10,000 year period, than the travel time for gas driven by topographic effects such as those observed at Yucca Mountain today.

In principle, one could hypothesize a diffusion-dominated system as one with very low permeability, but a permeability sufficiently low for advection to be negligible would probably be inconsistent with the existence of a deep unsaturated zone, because infiltrating rain water would not be able to drain.

3.4.2 Transport of carbon-14 from a repository in the saturated zone to and across the water table is feasible and should be addressed in the document (issues document section 2.2.2).

Transport of carbon-14 from a saturated repository to and across the water table is not treated quantitatively in the issues document. The treatment in the document of saturated repositories relies upon the assumption that significant amounts of gaseous components cannot be rapidly transported upward to the water table. However, it is reasonable to assume that the thermal gradient arising from the heated repository will cause convective transport of water (and dissolved carbon dioxide) above the saturated repository. The authors of the issues document assume that any upward movement of gaseous components below the water table will be diffusive, not advective. Because the rate of diffusive transport in liquid is about four orders of magnitude slower than diffusion in the gas phase, the issues document discounts the possible excursion of gaseous contaminants to and above the water table. Although it is not by any means certain that advective upwelling of radionuclides would be significant, this possibility should not be rejected without additional analysis. Flux upward across the water table is also feasible; an entire industry in the U.S. has been established to delineate the extent of underlying aqueous pollutant plumes based on analyses of aspirated soil gas samples.

3.4.3 Average gas travel times for the case of elevated terrain in the issues document sections 2.2.3 and 3.1.1 have probably been under-estimated.

The document's estimates of gas travel times in the elevated terrain case rely heavily on the results of Ross et al. [1992] which are based on Yucca Mountain. Calculations for Yucca Mountain do not embrace the full range of conditions possible at generic repository sites. Furthermore, these results need to be used carefully, with an appreciation of their limitations. The following points should be borne in mind:

a) Values are reported in Ross et al. [1992] as a range, corresponding to different locations within Yucca Mountain. The issues document uses only the fastest of these travel times, which are for locations at the western edge of the repository where gas can escape quickly to a nearby canyon. However, this escape route is only available to

carbon-14 from failed waste containers located in a particular section of the repository which comprises a small part of the total. Therefore, it is inappropriate to apply this travel time to the entire repository.

b) The reported travel times assume a rock permeability of $1x10^{-11}$ m^2. This value is the upper end of the range of permeability values measured on a large scale at Yucca Mountain, (roughly, $1x10^{-11}$ to $1x10^{-12}$ m^2 in Montazer et al., 1985, and Barnard et al., 1992). Travel time is inversely proportional to permeability when thermal convection is ignored (as is done by Ross et al. [1992]), so the uncertainty in travel time due to the uncertainty in permeability can easily be estimated. Use of permeability values less than the high end of the range will thus give rise to proportionally longer gas travel times.

c) The temperature distributions throughout the mountain in Ross et al. [1992] were obtained by solving the steady-state heat conduction equation, assuming a given repository temperature. When the calculation is done in this way, a large volume of rock is found to be heated. In reality, quite some time will be required for such a large volume to be heated and the time will depend on the design thermal load. Complicating the calculation is the decay of the heat source over time. Calculations by Buscheck and Nitao [1992] and others suggest that it will require on the order of 1000 years before the size of the heated area approaches a dynamic equilibrium with the heat source, a result that is largely independent of the thermal loading and design. The principal force driving gas flow is the difference in weight between air columns in heated and unheated rock. This difference is roughly proportional to the temperature difference multiplied by the height of the column of heated rock. For the first several hundred years, heat enters the rock from the waste but probably does not leave the top of the mountain to any significant extent, and so this product increases with time. During this period, gas migration will be much slower than a naive comparison of repository temperatures with the results of Ross et al. [1992] would suggest.

All of these considerations suggest that mean gas travel times at Yucca Mountain will be greater than indicated in the issues document. Means and ranges of travel times at other sites may differ but, as concluded in Section 3.4.8 of the Subcommittee's report, not enough is known to support the contention that alternative sites could be found with significantly different travel times than Yucca Mountain. (See also Section 3.4.6 of this report, retardation factors).

3.4.4 For an evaluation of generic repository sites, information on rock type and topography (flat vs. elevated terrains), as presented in the issues document sections 2.2.3, 2.2.4, and 2.3, are not directly relevant site factors. The parameters which should be varied in the sensitivity and uncertainty analyses are intrinsic permeability, air porosity, retardation factor, and effects of geological heterogeneities.

a) Principal factors controlling gas travel times. The travel times given in the issues document for elevated terrain are all based on calculations for Yucca Mountain because these are the only calculations available. However, using the topography of Yucca Mountain as a basis for comparative calculations should cause little loss of generality. Physical reasoning indicates that the principal factors affecting gas travel times in a homogeneous system are permeability, drained porosity, and repository depth. Provided that the temperature field is specified (meaning that convective heat transfer is ignored), these are the only site-specific parameters entering the governing equations for gas flow. Both permeability (again ignoring convective heat transfer) and drained porosity have a simple relationship to travel time. Given that repository heat is the main driving force, it would appear intuitively that the principal geometrical consideration influencing travel time is the depth of the repository, and this parameter is fixed by practical considerations within a rather narrow range.

b) Limitations on the ranges of parameter values. The only other unsaturated site that has been suggested for consideration [Winograd, 1981] is the alluvium in the region of the Nevada Test Site. Tuffaceous alluvium would have similar geochemical properties to tuff although with possibly higher gas permeability, which would be unfavorable with respect to retarding gaseous transport. The range of permeabilities that can exist at a site with a deep unsaturated zone is limited by the need for drainage to maintain unsaturated conditions. Shallow depths are ruled out by considerations of erosion and human intrusion.

c) Repository depth relative to valley floor. The observation that "the vertical extent of this flow pattern has not been established" because the repository will be below the adjacent valley floor [issues document section 2.2.3, page 2-21] is misleading. Whether the repository is within the mountain or below the floors of adjacent valleys is, at most, a minor factor in determining whether there is gas flow through it. The main factor is likely to be the permeability of the repository horizon and the rocks above it. Removing the repository from the area of flow requires a low-permeability barrier to be

present. This would be true even for an unheated repository; repository heat provides an additional reason why there will be flow.

d) Implications of seasonal flow. Another statement in the issues document that needs to be clarified is:

"The seasonal variations result in a long-term net upward flow of air inside the mountain. This seasonal advective flow pattern enhances the transport of gases at the site." [issues document section 2.2.3, page 2-22]

The net upward gaseous flux in Yucca Mountain is caused not by seasonal variations, but by the difference between average subsurface temperatures and average surface temperatures, by the greater humidity in the subsurface than in the atmosphere, and by interaction of the prevailing wind with the shape of Yucca Mountain. Although it is indeed probable that seasonal flows will enhance the transport of contaminant species in the rock gas, over long time periods this enhancement is a mixing-type effect rather than an additional contribution to net upward flow. The implications of seasonal flow for transport are mentioned in some of the literature on this subject, but they have not been analyzed quantitatively. It is also inappropriate to reach conclusions on the seasonal effects of gaseous flux over the range of possible repository sites based solely on Yucca Mountain data.

e) Effect of geological heterogeneities. The authors of the issues document assume that advection will be the dominant transport mechanism for gaseous radionuclides throughout all parts of the vadose zone in an elevated terrain. Although pressure differentials in the vadose zone may be created by thermal variability, geothermal gradients, barometric pressure fluctuations, and other factors, it is possible that at many potential repository sites the geology will not be homogeneous and a diffusion-dominated layer which restricts gas flow may be present. The assumption of homogeneity and isotropy in geologic media has been used in the issues document's modeling efforts but is unrealistic in its applicability to actual field sites and may give rise to travel times that are either underestimated or overestimated. In any case, the consequence of modeling the effect of heterogeneities will be to increase the range of uncertainties in the travel-time calculations.

3.4.5 A retardation factor of 1 for iodine-129 transport (as used in the issues document sections 2.1.1 and 2.2.1) is probably reasonable. However, the possibility that no gaseous iodine may migrate should be acknowledged. The gaseous release of technetium was not considered in the issues document. The Subcommittee agrees that it is not likely that gaseous technetium oxide (Tc_2O_7) could migrate in the presence of water.

Iodine-129 was assigned a retardation factor of 1 in the issues document. The thermodynamics of iodine would indicate that, at iodine concentrations of $\geq 10^{-3}$ Moles per liter and $\leq$ pH 7 under a normal atmosphere, diatomic iodine should be the stable species, which would favor gaseous transport. The chemistry of iodine at trace levels is known to be different [Kahn and Kleinberg, 1977] and favors the hydrolysis of iodine to HIO. Losses of volatile trace iodine have been reported for conditions which should have maintained the iodide. It is difficult to ascertain whether the volatile species is elemental iodine or whether it may be organic compounds formed by reaction of organics with HIO. Whether or not substantial amounts of iodine would be in a volatile chemical form under geologic conditions and over very long time periods is not known. Given this uncertainty, using a retardation factor of 1 is probably prudent. However, the possibility that no gaseous iodine may migrate should be acknowledged.

The gaseous release of technetium was not considered in the issues document. Gaseous species to be considered are Tc_2O_7 and $HTcO_4$. Tc_2O_7 is the acid anhydride for pertechnetic acid, a strong acid. Gaseous Tc_2O_7 is not likely to migrate in the presence of water. Technetium volatility has not been reported for any aqueous system other than acid solutions [Anders, 1960].

3.4.6 The range for retardation factors for carbon-14 used in the sensitivity analyses should be expanded to take into account the role of metal oxides. If this is done, then it is likely that the retardation factors will be identified by the sensitivity analyses as a parameter to which the release results are sensitive. This is counter to the conclusion in the issues document.

Processes which could retard carbon-14 transport are as follows:

a) exchange of gaseous carbon-14 dioxide with aqueous dissolved carbon dioxide;

b) exchange of gaseous carbon-14 dioxide with carbonate/bicarbonate adsorbed onto metal oxides; and

c) precipitation of the carbon-14 carbonate ion ($^{14}CO_3^{2-}$) as calcite;

d) exchange and solid-state diffusion into precipitated carbonates (does not require presence of liquid phase).

Process (a), exchange of gaseous carbon-14 dioxide with aqueous dissolved carbon dioxide, was the only one considered in the development of a range of values in the retardation factor (RF) for transport modeling. The methodology used to calculate RF values is essentially that described by Ross et al. [1992] (it is commonly used by many others in the modeling of carbon geochemistry) and assumes the dissolved carbon concentration is established as a function of calcium concentration, assuming equilibrium of the liquid with calcite and with the prevailing partial pressure of carbon dioxide. RF values of 30 to 88 for process (a) were calculated for various tuff units at Yucca Mountain; the calculations are defensible. However, adopting these uncritically for other sites is inappropriate, because they are calculated for specific conditions of temperatures, calcium concentrations and carbon-14 dioxide partial pressures. The averages and ranges for such conditions are likely to be different at other sites, and the issues document should evaluate the range to be expected.

Process (b), exchange of gaseous carbon-14 dioxide with carbonate absorbed on metal oxides, was noted in the issues document, but its effect on RF was not quantified because of lack of data. The Subcommittee believes that sufficient field and laboratory evidence exists for this phenomenon to warrant quantitative treatment in the estimate of a range of RF values. Striegl and Armstrong [1990] measured significant adsorption of carbonate in soils containing iron. Their observation is supported from a mechanistic standpoint by Rundberg and Albinsson [1992], and by similar observations by Reardon et al. [1986]. Striegl and Armstrong also note that the lack of change in isotopic composition indicates a capacity for carbon dioxide beyond that contained in the pore water alone.

The effect of iron oxides on increasing the RF value for carbon-14 dioxide in Yucca Mountain can be estimated from the observed sorption of carbonate onto goethite [Rundberg and Albinsson, 1992]. The sorption isotherm for carbonate at pH 7 and for an aqueous bicarbonate concentration of 0.0015 Moles per liter gives a solid phase concentration of 7.9×10^{-7} moles of carbonate per m^2 of goethite surface area. Yucca Mountain tuffs typically contain 0.1 - 1 wt% iron oxide, almost uniformly distributed through the mountain [Caporuscio et al., 1982; Schuraytz et al., 1986]. The grain size is micron to submicron [Schlinger et al., 1988]. The estimated available surface area of iron oxide in a typical tuff sample is therefore 0.0029 to 0.29 m^2 per cm^3 rock. Thus, the additional capacity due to iron oxide could range from 2.2×10^{-7} to 2.2×10^{-9} moles per cm^3 rock. The combined effects of processes (a) and (b) give a range of RF values for Yucca Mountain from 80 to 250. A similar approach applied to modeling adsorption of neptunium onto Yucca Mountain tuff indicated that results were most consistent for the assumption of 0.029 m^2 of iron oxide surface area per cm^3 rock. This would correspond to an RF for CO_2 of 94. Regardless of the RF for Yucca Mountain, adoption of these values as a representative range for all possible sites is not appropriate.

Process (c), retardation due to precipitation as calcite, has not been included in the value used for transport calculations because its effect on travel times is

probably negligible for the following reasons. First, calcite precipitation due to drying by the repository heat extends only 50 meters into the host rock, a relatively short distance compared to the overall distance to the surface, 450 meters (Codell and Murphy, 1992). Second, the total amount of carbon-14 which could be immobilized as calcite constitutes only 5-15% of the amount released (Codell and Murphy, 1992), which would only slightly reduce the release rate. Finally, calculations of the effectiveness of this process as a retardation mechanism are highly dependent upon the assumptions made about repository heating.

Process (d), exchange solid-state diffusion into precipitated carbonates, is probably negligible in any reasonable scenario. Laboratory data indicate that little diffusion of carbon-14 dioxide into the calcite solid phase occurs, because the crystal morphology of calcite is not conducive to high surface areas and thus limits its capacity for surface adsorption. Furthermore, little calcite has been found in drillcores from Yucca Mountain except in caliche layers near the surface, and at depths below the repository horizon.

3.4.7 The document (issues document section 3.1) needs to recognize the uncertainties associated with the effects of thermal loading of a repository.

Section 3.1 of the issues document does not reflect the findings of recent research into repository heat transfer [e.g., Ross et al., which indicate that future repository temperatures and their effects on moisture conditions are much less predictable than was thought at the time of writing the Site Characterization Plan (SCP) [DOE, 1988]. The uncertainty arises from several sources:

a) Gas-phase convection or natural heat-pipe phenomena may contribute significantly to heat transfer and cause temperatures near a repository to be lower than has been predicted using conduction-only models. Convective heat transfer may be further affected by heat-induced permeability changes caused by fracturing, silica precipitation, clay mineral alteration, etc.

b) Waste containers will arrive at the repository with a range of burnups and ages. This will likely cause considerable variation in temperatures at different locations within the disposal area.

c) Dry conditions may persist after temperatures fall below the boiling point, while water slowly flows back toward the waste [Buscheck and Nitao, 1992].

d) The thermal loading of the repository, originally set by DOE at 57 kw/acre, may be adjusted to accommodate changes in anticipated waste age and to seek advantages from higher or lower waste temperatures [Ramspott, 1991].

The thermal perturbation caused by an unsaturated-zone repository will essentially extend from the repository to the ground surface. The extent of subsurface temperature change needs to be consistently taken into account when evaluating gas travel time distributions. Rapid, thermally induced gaseous flux to ground surface through a "chimney" or heat piping phenomenon should be considered when the full range of possible repository sites is considered.

3.4.8 Not enough is known to support or rule out the conclusion that an unsaturated-zone repository site could be found where geological barriers could be shown to be capable of holding carbon-14 releases below the regulatory limits.

If one uses reasonable ranges of parameter values to describe sites with deep unsaturated zones, carbon-14 travel times from a heated repository are likely less than the 10,000-year regulatory period. One can speculate that a suitable unsaturated site exists where geological conditions would effectively contain carbon-14, but such a site has not been identified or investigated as yet.

3.5 Question 5: Is the Agency's assessment of the magnitude of the release resulting from the factors identified in questions #2, #3, and #4 complete, correct, and clear?

3.5.1 The estimate of carbon-14 release calculated in the issues document is consistent with the assumptions made and the parameter values adopted.

The result in the issues document of greatest interest to the Subcommittee is the estimate of the quantity of carbon-14 released from an unsaturated repository. As noted in the discussion of Question 4, the flat, non-advective case was not considered by the Subcommittee to represent a physically common situation; the more interesting case is when advection occurs, referred to in the report as an elevated repository.

The releases within 10,000 years are calculated in the issues document to be 4.9 times the release limit of 100 curies of carbon-14 per 1,000 MTHM (See Table 1 of 40 CFR 191): This estimate is based on an inventory of 1 curie of carbon-14 per MTHM, on a quick release fraction of 5%, on the assumption that 10% of the waste containers fail at 300 years and the remaining 90% fail at 1,000 years, on a long-term release rate of 10^{-4} per year, a retardation factor of 30 and an unretarded travel time of 10 years (the retardation factor times the unretarded travel time is the effective travel time). The calculation takes radioactive decay of carbon-14 into account.

3.5.2 The base case analysis of the issues document is based on assumptions that appear to lead to a high estimate of the amount of carbon-14 that would be released.

The issues document's base case estimate is that the normalized release ratio (the ratio of calculated release to the allowable release) is 4.9. For comparison, the maximum release ratio possible for a repository with 1 Curie carbon-14 per MTHM is 10; this would be the case for instantaneous release of all the carbon-14. In this analysis, the only factor that acts to restrict carbon-14 releases is the release rate of 10^{-4} per year. The assumed time to waste container failure (300 or 1,000 years) and retarded travel time (300 years) are quite short in comparison to the 5 730 year half-life of carbon-14, so these parameters have little effect on the calculated release. In the base case formulation, the calculated effect of the repository on carbon-14 releases, taking into account both the isolation provided by the geologic setting and engineered features such as waste containers, is almost inconsequential. Ignoring the repository and waste containers entirely, but including retention of carbon 14 in the fuel matrix, would produce a calculated release ratio of 6.0 versus the 4.9 when they are included.

3.5.3 The measures used to describe the magnitude of potential releases of carbon-14 are incomplete and insufficiently informative.

EPA should base its consideration of carbon-14 on a number of alternative assessment endpoints in addition to the normalized release fraction. The normalized release fraction is one relevant release endpoint, but multiple measures of risks are needed to provide an understanding of what is known about the likely nature of risks for potential releases of carbon-14 from a repository.

To provide the necessary multiple perspectives and measures for risks, the following endpoints are suggested for consideration:

a) annual release of carbon-14 as a function of time,

b) individual dose and dose rate for a representative off-site resident as a function of time post closure of the repository,

c) collective dose integrated for 10,000 years as a function of time post-closure of the repository, and

d) individual excess lifetime risk of cancer incidence for a representative off-site resident as a function of time post-closure of the repository.

In all cases, the endpoint used in the assessment should be accompanied by an analysis of uncertainty and an identification of the most sensitive parameters.

3.5.4 It is likely that EPA has overestimated the maximum individual exposures by a factor of 10 or more.

The Subcommittee received a briefing from Dr. Chris Nelson of EPA's Office of Radiation Programs and an accompanying document on individual exposures to carbon-14 (EPA 1992).

This analysis is a worst-case, bounding analysis (Subcommittee description) of exposure to carbon-14 to individuals living and growing their food on Yucca Mountain. The analysis assumes that 10% of the inventory (the upper bound for the quick release fraction) is released and exits the surface within a 100 year time interval. For this case, the maximum individual dose rate calculated was 4.7 mrem/year. Because it is likely that the actual distribution of times to waste container failure and for transport to the surface (given heterogeneities in the site) would spread the release out over thousands of years or more, it is likely that EPA has overestimated the maximum individual exposures by a factor of 10 or more. It also appears to be unrealistic to assume that all food is grown in the near vicinity of Yucca Mountain.

Average individual doses can be shown to be very low. For a release of 35 000 curies (half of that contained in a 70,000 MTHM repository) and a dose of 400 person-rem per curie (EPA 1992; the estimate from UNSCEAR 1988 is 248 person-rem per curie, about 40% lower), the population dose over 10,000 years is estimated to be 14 million person-rem, or an average of 1 400 person-rem per year over the 10,000 year period. The Agency computed the worldwide dose from carbon-14 using a multi-resevoir world model containing the atmosphere, slow turnover terrestrial biosphere, rapid turnover terrestrial biosphere, the surface waters of the ocean (mixed ocean), the thermocline, and the deep ocean. Most carbon-14 dioxide released to the atmosphere moves into the ocean, but a fraction remains in the atmosphere and the terrestrial biosphere, and it increases the specific activity of carbon-14 in those compartments and in people. For an averaged global population of 12 billion during this period, this corresponds to an average individual dose rate of 1.2×10^{-4} mrem/year, or an average individual lifetime dose of about 0.01 mrem. Based on a risk factor of 3.1×10^{-4} fatal cancers per person-rem (EPA 1992), this corresponds to an average lifetime individual risk of 3.1×10^{-9}. Based on this same factor, 14 million person-rem corresponds to about 4 000 cancer fatalities over the 10,000 year period.

3.6 Question 6: Does the Agency's document adequately describe the uncertainties associated with the assessment of the magnitude of the release?

3.6.1 The uncertainty analysis performed in the issues document is in a preliminary state and can be improved substantially.

The issues document is weak in the following aspects:

a) A critical evaluation is needed to determine whether the data cited in the report are appropriate for representing the limits of uncertainty for the model parameters. (See previous sections of this report for specific suggestions.)

b) The uncertainty analysis does not consider possible differences in the conceptual and/or computational model. Nor does it consider differences in assumptions about the uncertainty in the most sensitive model parameters, and the presence or absence of correlations among them. A critical component of the uncertainty analyses will be investigator judgment, given that directly relevant data sets are often lacking, requiring the use of judgment to quantify the limits of parameter uncertainty. Thus the results of an uncertainty analysis will differ depending on investigators assigned to the project.

Uncertainty analyses should be iterative to guide efforts to improve the knowledge base about the most sensitive parameters that control the initial estimates of uncertainty in the prediction. Initially, the most sensitive parameters are identified through a preliminary uncertainty analysis, and then these estimates are improved by investing in additional research and/or soliciting judgment from experts. This process is repeated until the overall uncertainty is, for practical purposes, either irreducible or negligible. Before a negligible level of uncertainty can be identified, a risk management decision is required in terms of a negligible level of either carbon-14 release, exposure, or risk.

The Subcommittee recommends that efforts at modeling release take natural analogs, laboratory data, theoretical analysis, and field data into account to reduce uncertainty and use available scientific knowledge.

3.6.2 The objective of the uncertainty analysis should be clearly stated.

If the analyses aim to represent a range of different repository sites, as seems most appropriate if generic regulations are being developed, then the ranges of parameter values used in the analyses should be reviewed to ensure that they reflect the relevant spectrum of characteristics possible for repository sites. At present, the issues document limits the ranges of values it uses to those measured at a very few, specific sites. In some cases, values measured for Yucca Mountain are applied inappropriately or without documented justification to other generic sites.

The question that is to be answered by an uncertainty analysis must be clearly stated; otherwise inputs will not be consistently defined and the results will

have little value. Such an analysis of carbon-14 transport could be designed to answer any one of several questions, including these, and possibly other, questions:

a) What is the range of possible carbon-14 releases if a repository were to be built at a randomly selected location with a sufficiently thick unsaturated zone?

b) What is the range of carbon-14 releases that could occur if a repository were to be built at some unsaturated-zone site with generally favorable geological characteristics?

c) How much carbon-14 might be released by any HLW repository at Yucca Mountain?

d) What is the range of possible carbon-14 releases from a particular repository (with a particular inventory, thermal loading, and design) that might be built at Yucca Mountain, given present knowledge?

e) How much uncertainty will there be in carbon-14 releases from a Yucca Mountain repository when site characterization and repository design have been completed?

For each assessment question, there will be different ranges of parameter values, and also different lists of parameters. If the uncertainty analysis aims to represent a range of different sites (as seems most appropriate if generic regulations are being developed), then it must address the relevant spectrum of geologic settings. For each of these settings, ranges of parameter values consistent with the existence of a deep unsaturated zone must be determined.

Probabilities of the parameter values in each rock type must be assessed. For each type of setting, frequency distributions of the relevant parameter values should be defined on some scientific or other justifiable basis; if insufficient information exists to define the distribution, then at a minimum a reasonable range of parameter values should be estimated. The earlier discussion (Subcommittee report section 3.4.4a) suggested that the topography of Yucca Mountain could be used for generic calculations because topography should have little influence on gas travel time under thermally loaded conditions. However, values of non-geometrical parameters (permeability, drained porosity, retardation factors) cannot in a generic calculation be limited to ranges observed at Yucca Mountain; generic ranges appropriate for each particular setting must be used.

3.6.3 Key uncertainties are underestimated, and some are not discussed.

As noted in the Subcommittee's report (sections 3.2 and 3.3), the issues document makes assumptions that rule out the possibility of a significant

reduction in carbon-14 releases from engineering measures such as long-lived waste containers that remain intact beyond 10,000 years. In fact, the presumption that all waste containers have failed at 1,000 years seems quite pessimistic for an unsaturated site. The Subcommittee's point is not that early waste container failures are not possible under the analytical framework of a generic repository that lacks a reference repository design. It is instead that the upper end of the distribution for waste container life should extend significantly higher than 1,000 years. In a presentation to the Subcommittee by its contractor, DOE indicated that it is possible that a multilayer waste container could contain the carbon-14 long enough to meet the EPA limit, but that it would not be possible to verify that the container would be gas tight for 10,000 years. Similarly, in its study of waste management by DOE, the Office of Technology Assessment notes that DOE's Defense Waste Processing Facility at Savannah River has adopted a canister design lifetime of 8 000 years for stainless steel canisters in an interim storage building (OTA, Long-lived Legacy, p.25, 1991). The canister degradation mode in the storage building is assumed to be atmospheric corrosion, and other corrosion modes must also be considered for canisters placed in an underground repository.

Similarly, as noted in the Subcommittee's discussion of transport modeling, the uncertainties in assumed geologic features such as permeability and geochemical features that affect travel time and retardation are based too narrowly on a homogeneous setting with some data from Yucca Mountain, whereas a generic repository must have greater variability.

In addition to relying on assumptions that give little credit for retention or retardation of carbon-14 within the waste containers, in the repository, or along the pathways through which the gas would migrate, the analysis assumes that all of the carbon-14 is in a chemical form that will convert to carbon-14 dioxide. While the Subcommittee knows of no analyses of the chemical form of carbon-14 in the waste, it is likely that some is in a stable solid form.

Although not applicable for releases that are reported only in terms of a normalized release ratio, there are additional uncertainties that arise in terms of the doses and risks that would result from carbon-14 release. Section 3.5.3 of this report discusses alternative measures that could be used to characterize these potential releases. One such factor is the assumed global population over the next 10,000 years. The release ratio was derived on the basis of total population dose, and these doses will vary in proportion to the population. Individual doses will be affected by whether the repository contains 70,000 or 100,000 MTHM (See Section 3.1.2 of this report), but the normalized release ratio will not.

In addition to the Subcommittee's view that parameter values have been assigned ranges that are too narrow to reflect their actual uncertainties, the uncertainty analysis is very sensitive to the shapes of the distributions specified for each parameter and to the assumptions made about the presence or absence of correlations among parameters. The use of truncated distributions, such as the uniform, triangular, or log uniform, implies that values beyond the specified

minima and maxima assumed by the issues document are absolutely impossible. Adequate justification for the ranges assumed for the uncertain parameters has not been provided by the issues document.

Finally, the Committee recognizes that there is large uncertainty associated with the assumption that the number of health effects in a population is directly proportional to population dose. Carbon-14 releases from a high-level waste repository will produce exposures to individuals that are a small fraction of, but a finite incremental increase above, the exposure to natural background radioactivity. The uncertainty associated with this small incremental exposure is such that the estimate of health risk may be zero if alternatives to the linear no-threshold dose response model are considered. On the other hand, the upper bound of this uncertainty range is several times higher than the central risk estimate.

3.6.4 Selection of a limited range of parameter values predetermines the outcome of the sensitivity analyses.

Careful and scientifically-justifiable definition of the uncertainty associated with the model parameters is important to identify controlling variables and to recommend lines of research which would be most cost-effective in reducing the dominant uncertainties in the risk analysis or in reducing the risks themselves.

The Subcommittee is concerned that overly narrow ranges have been selected for certain key performance parameters. Notably, the assumption that waste containers fail at either 300 or 1,000 years leads to the result that repository releases of carbon-14 are insensitive to waste container life. The Subcommittee believes that this result is incorrect, and that waste container life may be an important factor in controlling carbon-14 releases. It is important to recognize the sensitivity of the calculated release to assumptions about the performance of the waste container and other engineering barriers. If barriers contain the carbon-14 for 10,000 years, the normalized release ratio will be zero. Other uncertainties regarding potential releases of carbon-14, for example, in inventory or travel time, will not affect this result. Similarly, the assumption that the retardation factor falls between 1 and 60 is likely to lead to an underestimate of the potential importance of retardation.

3.6.5 The sensitivity analysis performed in the issues document is a one-at-a-time perturbation of parameter values. Such sensitivity analyses may give misleading results when the model represents a non-linear system.

In a non-linear system, the results of a one-at-a-time sensitivity analysis are highly dependent on the choice of nominal values for those parameters held constant. An appropriate approach for identification of the most sensitive parameters would be to use the Monte Carlo results directly. The most sensitive parameters would be those that dominate the overall uncertainty. The

identification of the most sensitive parameters can be obtained through regression analyses of the model prediction and each uncertain parameter (IAEA 1989).

Useful quantitative indices of parameter importance are Spearman's Rank Correlation Coefficient or, for a system with strong correlations among its parameters, a Standardized Partial Rank Regression Coefficient. For the identification of parameters that determine values in the extremes of the CCDF, visual inspection of scatter plots of the prediction of carbon-14 releases as a function of the value of each uncertain parameter is recommended.

When the uncertainty analysis is conducted based on the range of input parameter values in the issues document, the mean value estimate of the normalized release ratio is 3, and the median estimate is 2. These are lower than the deterministic or base case estimate of 4.9.

3.6.6 When the broader uncertainty bands recommended in this report for various parameters are considered, the overall uncertainty regarding the potential magnitude of carbon-14 releases is quite broad, and spans ratios of the estimated to allowable release ranging from zero to as high as ten.

On the basis of presently available information, it is not possible to predict with reasonable confidence whether a generic unsaturated repository would or would not meet the Table 1 release limit. Because the characteristics of a generic repository must be assumed to be based on those of representative geologic settings, there is substantial variability in the parameters most important to performance regarding release or retention of carbon-14. In the worst case, a repository could have a rapid travel time to the surface and a short waste container life as was assumed in the base case. In addition, there could be more carbon-14 in the repository than for the base case; the uncertainty on carbon-14 content in the issues document ranges up to 1.5 times the base case. The release rate from the fuel could perhaps be ten times greater than was assumed, on the order of 10^{-3} per year. For this combination of parameters, almost all the carbon-14 in the repository (more than was assumed in the base case) is released and transported to the surface before any significant radioactive decay has occurred.

Conversely, if the site has a retarded travel time approaching or exceeding 10,000 years, or if the waste containers and other engineered barriers prevent releases for longer than this time, no carbon-14 is released within 10,000 years. This wide range of possible outcomes reflects the lack of solid data on key performance parameters and the high variability inherent in describing geologic parameters for a generic site.

3.6.7 The release ratio calculation is for releases within 10,000 years. The result of this analysis is not sensitive to this 10,000 year cutoff.

The main reason for this limited sensitivity is that the carbon-14 half-life is 5 730 years. This means that almost three-fourths of the carbon-14 inventory will have decayed in 10,000 years. The issues document calculates normalized release ratios for releases within 10,000 years. If this time limit is not invoked, the deterministic or base case estimate of the release ratio increases from 4.9 to 5.2.

3.6.8 EPA should distinguish between predicting the occurrence of rare events and performing an uncertainty analysis about a "best estimate".

The end result of an uncertainty analysis is a subjective confidence interval about a "best estimate" prediction, which is specific for the target endpoint of the assessment. The "best estimate" in this case is represented by the median or 50th percentile of the Monte Carlo result. The subjective confidence interval represents a region wherein the true but unknown result is to be found with a sufficiently high degree of confidence (usually a 90 to 95% subjective probability interval).

When the endpoint of concern is a 90 or 95% subjective confidence interval about a best estimate prediction, differences in the assumed shape of a parameter distribution will often be of only minor importance, provided that the mean and variance remain unchanged from one assumed distribution shape to another. The 90 to 95% subjective confidence intervals derived from the Monte Carlo results are most sensitive to the estimates of central tendency (median and mean) and relative uncertainty (coefficient of variation) specified for each uncertain parameter. They are virtually insensitive to the occurrence of extreme values (provided that the mean and variance remain constant among different alternative choices of distribution shapes).

Assumptions about the presence or absence of strong correlations between parameters are important for only the most sensitive parameters in the model. Weak correlations have only a minor influence if they occur among sensitive parameters and practically no influence if they occur among insensitive parameters.

In an uncertainty analysis for a "best estimate" prediction, judgment is used to quantify the "degree of belief" that the true but unknown value for a model parameter will not be exceeded by any given value. Therefore, the term "subjective confidence" is employed. The use of judgment is necessary because in most (if not virtually all) cases, data sets are insufficient for quantifying the range of uncertainty associated with the application of a parameter to a given assessment question. The Subcommittee has not tried to determine whether the subjective confidence that it used as a standard of judgment is equivalent to the reasonable expectation standard in proposed EPA regulations.

3.6.9 The Subcommittee notes that the uncertainties in the estimates of risk from carbon-14 releases presented by EPA staff are of comparable magnitude to the uncertainties associated with the estimation of the release of carbon-14 over 10,000 years.

The August 1992 presentation by C.B. Nelson of the Office of Radiation Programs estimated uncertainties in the prediction of health consequences over 10,000 years resulting from a unit release of carbon-14 to vary within a factor of five about its best estimate (excluding the possibiliity that the lower bound of risk may be zero). This uncertainty is similar to the magnitude of uncertainty estimated in the issues document.

The uncertainties associated with the calculation of health risk per individual or health consequence to the global population are unlikely to be resolved through further research (at least in the near term) and therefore constitute a source of uncertainty that is for all practical purposes irreducible.

3.7 Within the context of its charge, the Subcommittee offered the following comments relative to the Science Advisory Board's document, Reducing Risk: Setting Priorities and Strategies for Environmental Protection.

3.7.1 It is inappropriate to consider the relative performance of different generic repository settings on the basis of carbon-14 alone.

The issues document evaluates several different types of generic sites in terms of their likely carbon-14 releases, including repositories that are saturated, unsaturated, elevated, and flat. As noted earlier, the assessment of potential carbon-14 releases from an unsaturated site inappropriately assigns great differences to flat versus elevated sites. The actual differences, if any, are likely to be much smaller.

One should not get the impression from this evaluation that it is possible, on the basis of considering carbon-14 alone, to determine that one type of site is preferable to another.

Optimizing site selection on the basis of a single criterion (gaseous releases) may or may not cause loss of optimal conditions for other criteria. For example, the issues document estimated that carbon-14 releases from a saturated repository would be well below the release limits of the EPA standard, but risks from other radionuclides may be greater or smaller, depending on a number of factors, such as salinity of the water at the horizon (National Research Council 1983). Other performance characteristics such as the difficulty and complexity of retrievability and cost are also important to an overall evaluation. As noted above, the Subcommittee has encouraged investigation of the use of multiple barriers to

retard the migration of carbon-14 to the surface. In the consideration of such barriers, benefits other than those associated with carbon-14 retardation should also be evaluated.

3.7.2 For a proper interpretation of the effectiveness of risk management decisions, EPA should compare the health and environmental risks posed by a repository, including releases of carbon-14, with the risks posed by other environmental problems.

Such a comparison would provide useful technical input to the complex decision of properly managing health and environmental risk faced by current and future populations. This evaluation should be made using multiple endpoints, as recommended in section 3.5.3 above.

3.7.3 From a risk management perspective, a key point to understand about doses and risks from repository releases of carbon-14 is that the perceived significance of the issue hinges upon whether one views the risks from the perspective of an individual or a population.

From the population perspective (that associated with the use of a normalized release ratio), releases of carbon-14 dioxide from a repository may produce an appreciable global population dose possibly corresponding to more than 1,000 deaths in 10,000 years. Conversely, the estimated individual lifetime risks are much lower than those regulated for radionuclides or carcinogenic chemicals in other contexts. What is at issue here is the need for a consistent approach to the regulation of potentially long-lived, ubiquitously dispersed carcinogens.

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