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MEMORANDUM FOR: Daniel J. Fehringer
Repository Projects Branch
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FROM: Harold T. Peterson, Jr.
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and Waste Management, RES

SUBJECT: COMMENTS ON DRAFT ORNL REPORT ON METABOLIC DATA FOR HLW DOSIMETRY (ANNUAL REPORT: FIN B-0289)

As requested by your memorandum of November 17, 1983, I have reviewed the subject report. The report appears to be a comprehensive literature survey of the geochemical behavior (speciation) and mammalian metabolism of the transuranics,(neptunium, plutonium, americium) and strontium. The suggestions for further research appear to be well thought out and are supported by the identified deficiencies in the available information. Detailed comments are enclosed. Most of these are editorial or typographical in nature. Several comments contain optional suggestions for a alternative wording to improve clarity.

If you have any questions, please contact me on 42-74578.

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Enclosure: 1, as stated

REVIEW OF "DETERMINATION OF METABOLIC DATA APPROPRIATE FOR HLW DOSIMETRY (ICRP-30), I.

GENERAL COMMENT: The report is comprehensive and generally well done. The suggestion for additional research are particularly useful. However, it would have been easier to review if page or section numbers had been included.

NOTE: The title page attributes support for the report to the Division of Facility Operations, RES. The ORNL group does do work for that division however, shouldn't it be the Division of Waste Management, NMSS?

MAJOR COMMENTS:

SECTION: "EXPECTED SPECIATION OF THE NUCLIDES"

Strontium: lines 23-24: There are several common organic molecules which form soluble complexes with strontium. The text states: "It is generally believed that stronti has little tendency to form strong water-soluble organic complexes." See detailed comments for details and suggested corrections.

REFERENCES FOR SECTION 2:

The amount of information in the references is variable and suggests that a number of foreign references were abstracts in a abstracting journal such as Chemical Abstracts. If so, these references should be clearly spelled out by noting: (Abstract in _____).

SPECIFIC COMMENTS

SECTION: RELEVANT ENVIRONMENTAL PARAMETERS

Second page, last paragraph, line 4-6. Suggest cite reference(s) for incorrectness of "negative logarithm of hydrogen ion activity" as definition of pH as many texts on thermodynamics or physical chemistry use this definition. "Immeasurability" per se, doesn't make the definition incorrect, but it does limit its usefulness to environmental measurements.

The use of "now" in line 4 and "modern" in line 7 are somewhat confusing and conflicting. Suggest changing line 4 to: "It has been defined as..." which resolves the previous comment as well.

REVIEW OF ANALYTICAL METHODS

First page, second paragraph, line 6. The use of "specie" appears to be restricted to money, "species" is both singular and plural and should be used.

FACTORS WHICH CHANGE SPECIATION DURING TRANSPORT

Second page, second paragraph, second sentence (lines 3-5). This sentence dismisses the problems with filtration of colloids perhaps a bit too quickly. Standard books on analytical chemistry may provide for an expanded description of the general aspects of collodial solutions such as gel formation, etc. Even if they are not specifically applicable to the nuclides of prime interest, they may provide the reader with a better "feel" for these problems.

EXPECTED SPECIATION OF THE NUCLIDES

Strontium, second page, first line should be "exists" not "exist". Second line "...solutions used are generally..."? or should it be: "...solutions used were generally..."?

Strontium, second page, lines 6-8. This sentence is slightly unclear. Suggest:

> "For adsorption of strontium on hydrous oxides, the values of K_d increase sharply with pH[34,35]. At high pH, adsorption on or coprecipitation with hydrous materials such as aluminum or ferric hydroxides may predominate."

Lines 15-18 it might be advisable to link the "low electrophoretic mobility" of radium to the presence of an uncharged, possibly insoluble species, for the benefit of readers who are not chemists and could be incapable of appreciating the significance of low electrophoretic mobility.

Lines 23-24. It is not generally believed that this is true. Most analytical chemists are aware of the affinity of EDTA (Ethylene diamine tetraacetic acid), an organic complexing agent, for calcium and strontium. Another organic complexing agent, DTPA (Diethylene triamine pentaacetic acid) also has a strong affinity for strontium. Soil migration studies show that the strontium compounds of both complexing agents are mobile. Hence delete first sentence and "However," and start with "Studies". Complexing agents such as EDTA are sometimes used in agriculture to enhance plant uptake of essential metals. Such agents could complex radionuclides in soils. These complexing agents, in addition to "humic acid" and other naturally-occurring complexing agents, might increase radionuclide mobility at disposal sites.

Americium: Second sentence from the end of page is unclear. Is it the intent to indicate that strontium carbonate is insolubile and its precipitation would limit the solubility? If so, the meaning would be clearer if it were transposed with the sentence that immediately precedes it. That sentence and the last sentence refer to the formation of soluble complexes.

Neptunium: Second page, third paragraph, lines 5-10. These sentences appear contradictory:

"...it is possible that these particulates might be transported, for considerable distances." versus "There is little in the literature that we have found so far that might indicate the extent to which these particulates could be transported,..."

Either the top phrase should be qualified or the bottom one deleted depending upon the degree of support for the particulates being transported for considerable distances.

Plutonium: Second page, first line should read: "...Pu^{3+}, Pu^{4+}, PuO_2^{+}, PuO_2^{++} ..." not "...Pu_3^{+} , Pu_4^{+}..."

Fifth page, second paragraph, lines 6-9. Is it the "behavior" of polymeric plutonium, the fraction which exists as polymeric plutonium, or both, which is hard to predict at a given site?

Fifth page, last paragraph, line 4: "...Pu^{3+}..." not "...Pu_3^{+}..."

FURTHER RESEARCH NEEDS:

Second page, item 1. It is somewhat unclear what the precise intent of "verification of existing constants." Either a more complete description of which constants are to be verified is needed or the sentence should (if the meaning is appropriate) be combined with the first sentence.

REFERENCES FOR SECTION 2:

Note: NRC usage is to underline journal names and book titles (NUREG-0650). Articles (symposium papers) in a book are set-off in qoutes.

[15] title should read: "...User's Guide..." instead of "...User[s".

[19] The first author is "Åberg" not "Abeng".

[51] Line 4: "ACS (American Chemical Society)" instead of "AES" Symposium.

BIOPHYSICAL AND BIOCHEMICAL PROPERTIES OF Pu, Am, Cm, Np, AND Sr.

GENERAL:

It would be helpful if the sections dealing with each element were set off: "Plutonium", "Americium," "Strontium," etc.

First page, third paragraph, line 7. It might be helpful to emphasize that polymerization occurs on the high pH side of pH 1 by changing the ending to read: "...begin to polymerize at pH values as low as pH 1."

Page 2, second paragraph, line 10. This reviewer is unfamilar with the term "colloidals" as opposed to "colloids." Colloidal is generally used as an adjective, while colloid is the noun.

Page 2, third paragraph, line 5. It would be useful also to give a synonym for "cytosol" as this is a relatively obscure biological term.

Page 3, second paragraph, lines 1-2. Although cumbersome, it would be clearer if the valences were written out: "3+, 4+, 5+, 6+, and 7+" and "4+, 5+, and 6+".

ABSORPTION FROM THE LUNGS TO BLOOD"

Page 1, first paragraph, first sentence, strickly speaking "The most recent version of the model...[does not view anything]. Suggest:

> "In the most recent version of the model of the respiratory system approved by ICRP[33,34] the respiratory system is considered to be divided into four regions:..."

Page 1, first paragraph, second sentence. Suggest slight alteration for style:

> "The fraction of the material entering the body which deposits in each of the NP, TB, and P regions depends on the..."

Page 1, second paragraph. Although dissolution and solubility are the principle mechanism and parameter used in the ICRP model, it might be noted that there are implied biological clearance processes such as mucociliary clearance and phagocytosis by macrophages which are implicitly incorporated into the model through the clearance rate consistants for the three compartments. You could refer to page 5 of this section which discusses these clearance rates.

Page 2, paragraph 4, lines 3-4. This might be alternately written as:

> "...there was more rapid translocation of Pu-238 than Pu-239 from the lung to systemic organs[36,42]."

In order to emphasize the Pu-238 vs. Pu-239 difference.

Page 7, first full paragraph, last three lines. The relationship to [the intrinsic] specific activity is understandable; however, as there is no unique characteristic particle size for these compounds, it is not clear why the Cm-244 should be generally more soluble than Am-241 due to smaller particle size. The problem seems to be in the language used which tends to make the statement sound like generality when statements like: "...The smaller particle size...of Cm-244 oxide" appear to only apply to the conditions of particular study. Suggest:

> "...perhaps because of the greater specific activity of the Cm-244 compared with Am-241; invoking a mechanism similar to that previously described for the Pu-238/Pu-239 difference."

Page 7, second paragraph, first line. The subject (nitrate and citrate forms) is plural, the verb "is" is singular.

EFFECTS OF CHEMICAL FORM ON GASTROINTESTINAL ABSORPTION

Table 3.4 could use ditto marks (") under species for indicating the same animal was used in consectutive studies. If error vlaues are available for these data, it would be useful if they were included.

DISTRIBUTION AND RETENTION OF ACTIVITY REACHING THE BLOODSTREAM

Page 4 (Americium and curium), first paragraph, line 12. It might be instructive to point out that citrate is a metabolizable complexing agent to contrast it with TTHA: (last line): "...are for the chloride, nitrate, and citate forms (the latter is a metabolizable complexing agent)."

SUITABILITY OF ICRP-30 METHODS FOR TREATING METABOLISM OF DIFFERENT CHEMICAL FORMS.

Page 2, move heading on last line to top of following page.

Fig. 3.2. No indication is given in the title (or figure) of which curve is for the soluble and which is for the colloidial Pu. This distinction is also absent from the text although one can deduce that the upper curve is for the soluble form from the numerical values given.

REFERENCES FOR SECTION 3

[33] third line, "Respiratory" not "Respiratry".

[41] The title of the symposium is not underlined as in the other references of this type.

[42] "Inhaled" in the title should begin with a lower-case "i".

[57] "Radiat. Res." should be underlined an a volume and page citation given.

[60] The volume citation is missing and the publisher citation is unnecessary.

[62] It would be helpful to identify this as a U.K. National Radiation Protection Board publication for those who do not readily identify NRPB.

[66] Needs a fuller title citiation for the report.

[69] The IRPA proceeding are generally issued in several volumes. If this is the case for this reference then a volume number as well as the page citation would be helpful.

SUMMARY AND CONCLUSION

Page 2, last paragraph. It is important in the criticism of the ICRP model to distinguish between the model and the associated parameters such as the solubility classes. The discussion of Pu-238 here appears to indicate a

problem in the solubility classification for Pu-238 as a class Y compound when experimental evidence suggests a more mobile form such as class W. The class designations are not hard and fast rules only guides based upon a review of the available literature at the time the tables were prepared.

Page 3, first (incomplete) paragraph. What is meant by "accurate"? Do you mean "biologically correct" in terms of mechanisms and physiological models? Or that which best portrays the retention of plutonium in the body? This distinction is important as one could have a very incorrect model (in terms of modeling actual biophysical processes) which predicts the body burden or individual organ burdens with reasonable accuracy or a very "realistic" model of the biophysical process which requires so many parameters to be specified that it could be useless for practical applications. A slightly expanded discussion of what is meant by "accurate" in this context would clarify this.

Page 4, paragraph 3, last line. This sentence is not totally clear on the basis for stating that the ICRP-30 data seem reasonable (compared to what?) and what is meant by the "most commonly encountered environmental Sr." Presumably, this means:

> "The metabolic data for soluble strontium given in ICRP-30 are applicable to the most commonly encountered form, Sr^{+2}.