Sandia National Laboratories
Albuquerque, New Mexico 87185

January 18, 1983

Dr. T. J. Walker
U. S. Nuclear Regulatory Commission
Office of Nuclear Regulatory Research
7915 Eastern Avenue
Silver Spring, MD 20912

Dear Tom,

At your request, we have reviewed the document entitled, "Aerosol Release from Sodium-Concrete Reactions", by L. D. Muhlstein and R. P. Colburn[1]. This study concludes that the only materials present in appreciable quantities in the cover gas space above the sodium pool during sodium-concrete interaction tests are sodium vapor, hydrogen, and the inert cover gas. No products of sodium/concrete reactions have been observed as aerosols. Our experimental findings agree with this. The aerosols from our tests have consisted of sodium with a minor amount of NaH. On this basis we agree with the primary conclusion of the Muhlstein and Colburn paper that the aerosol generated by sodium-concrete interactions is primarily sodium.

There are, however, several other aspects of aerosol generation and behavior during accidents beyond design basis at CRBR which deserve attention. Beyond Design Basis accidents involve both sodium and core debris interactions with concrete. Aerosol generation and fission product release are likely to be more intense and from a variety of sources in this situation. Aerosol generation and behavior not fully explored in either the study under review or the TMBDB analyses are:

1) Characterization of aerosols produced from a boiling sodium pool has not been done. Characterization of size, mass loading rate, and behavior parameters is necessry to evaluate the effectiveness of the filterd vent system and engineered safety features in containment.

2) Release of refractory fission products is not treated conservatively. Plutonate chemistry i not considered and aerosolization of sodium-fuel compounds from the pool is not completely treated.

3) Release of volatile fission products is conservatively treated for early times in the accident but may not be conservative at times when venting occurs.

H001

4) The possibility that aerosols may plug flow pathways and vents remains an area of uncertainty.

These areas are discussed more completely below. The data base for dealing with these areas is quite sparse.

Aerosol characterization has not been done for aerosols generated by a boiling sodium pool. The overwhelming majority of the aerosol produced in a reactor accident will be produced from sodium vapor. Knowledge of the size distribution, mass concentration, and other parameters which describe this aerosol are necessary to evaluate the performance of the filtered vent system on CRBR. For example, the ability of an aerosol particle to pass through a filter, "filter penetration", is strongly dependent on the aerosol particle size. Figure 1 is an illustration of filter penetration through a gravel bed as described by Lee.[2] Different particle removal mechanisms are dominant in different particle size regimes. Small aerosol particles are removed by diffusion while large particles are removed by interception, gravitational settling, and impaction. Particles of intermediate size are removed by a combination of these mechanisms and usually are not removed as efficiently as smaller or larger particles. In general, these mechanisms and the sensitivity to particle size are applicable to all filter systems. This example, shows that the particle size distribution of the aerosol can dramatically change filtration efficiency.

Some characterization of sodium fire aerosols has been done at ORNL[3] and HEDL. These aerosols are, however, not necessarily characteristic of an aerosol formed when sodium vapor evolved during sodium boiling and sodium interactions with concrete condenses or reacts in the CRBR containment atmosphere. There are a number of aerosol behavior codes such as MAEROS, CONTAIN, QUICK, AEROSYM, MSPEC, HAARM, and HAA which calculate the evolution of aerosols in containment. The aerosol source rate and parameters describing aerosol size distribution are necessary input to these codes. Successful application of these codes, therefore, requires the correct initial description of the aerosol. There seems to be no significant data base on aerosols formed by sodium condensation or reaction processes expected to occur in a beyond-design basis accident at CRBR. The aerosol source term sensitivity study being conducted for you at Sandia should be most useful in determining which characteristics of the aerosol source most affect aerosol behavior in containment and therefore must be known in most detail.

The above discussion also applies to the filters that protect in-containment sensors from aerosols. High efficiency filters may become clogged with aerosol

effectively isolating the sensor from containment. Low efficiency filters may allow enough aerosol penetration to alter the sensor's calibration.

The sensitivity of filter performance to aerosol particle size means that the time evolution of particle size during an accident must be calculated carefully. The CRBRPO has analyzed the aerosol behavior in containment with the HAA code which assumes the particles are log normally distributed in size. Analyses have been replicated at Sandia with the CONTAIN code which does not place such a rigid constraint on the particle size distribution. The analyses with CONTAIN suggest the results obtained with HAA are probably conservative with respect to the amount of aerosol suspended in containment and consequently the amount of aerosol that must be filtered. On the other hand, HAA predicts the aerosol particles in containment are larger and therefore more easily filtered than does the CONTAIN code.

The release of refractory fission products is not conservatively treated in TMBDB. TMBDB recognizes aerosolization of particulate suspended in the sodium by a sparging mechanism (Jordan Ozawa[4]). It does not account for fission product compounds dissolved in the sodium which may also be released by sparging. The treatment of plutonate chemistry in TMBDB is incomplete and this affects the amounts of both dissolved and suspended fission product in the sodium.

Dissolved and suspended fission product compounds are released as aerosols from the sodium pool by bursting bubbles. When bubbles burst at liquid surface, droplets with dimension of microns are thrown off (Tomaides and Whitby[5]). The droplets contain both dissolved material, which has a concentration the same as that of the bulk liquid, and suspended particulate at a concentration which is not necessarily proportional to that of the bulk liquid (Quinn, et al.[6]). The results of Quinn, et. al. imply that the concentration of suspended particulate can be higher in the droplets than in the bulk liquid. Rising bubbles transport suspended particulate to the liquid surface by flotation. These results also imply that there is a limit to the amount of floated material that is released from at the sodium surface. When this flotation of suspended particulate to the sodium surface occurs, the concentration of particulate in ejected droplets will be independent of concentration in the bulk liquid. The size and number of ejected droplets from the bursting bubbles is very important in determining the mass of liquid aerosolized. The data of the Tomaides and Whitby[5] for an aqueous system, when applied to the estimated conditions in TMBDB, suggest an aerosolized mass between 190 kg (0.55 cm bubbles) to 1.3×10^4 kg (0.14 cm

bubbles). For a concentration of 6.10^{-4}gPu/gNa (15% of the plutonium inventory dissolved or suspended in $5x10^{4}$kg of sodium) and assuming that the droplet concentration is the same as that of the sodium pool, the corresponding plutonium release is between 82g and 7.8 kg. This calculation ignores the potential concentration enhancement in the droplets of suspended particulate from the pool by flotation. This release would continue throughout sodium boiling and can be much larger than that proposed in TMBDB for the sodium boiling period.

TMBDB references the work of Jordan and Ozawa[4] when treating aerosols produced by sparging. The Jordan and Ozawa work underplays the effect of dissolved fission product compounds in the sodium pool. Jordan and Ozawa measured uranium release from a boiling mixture of sodium and UO_2 particulate. They found uranium release to be independent of the concentration in the bulk liquid. They proposed vaporization of Na_3UO_4 as an explanation. The results of Quinn, et al. provide a more likely explanation. In addition, in the work of Jordan and Ozawa, oxygen was not continuously supplied to the pool as it would be in the TMBDB case where Na/concrete reactions are occuring. It is likely that, in the former case, the uranate contributions to aerosolized uranium was small compared to UO_2 particulate contributed whereas, in the latter case, the relative contributions are unknown.

The formation of sodium urano-plutonates is directly affected by the oxygen content of the pool. This, in turn, is determined by the Na/concrete interaction which is a source of oxygen to the pool. Sodium and mixed fuel oxides react according to the following stoichiometry (Adamson, et al.[7]):

$$3y\ Na(l, O\ dissolved) + (2 + y)\ U_{1-x}P_xO_{2-x} \qquad (5)$$

$$(y-z)\ Na_3U_{1-x'}P_{x'}O_4(s) + (2 + z)\ U_{1-x_1}P_{x_1}O_{2-y} \qquad (6)$$

The extent of this reaction (and other similar reactions) as well as the amount of fission product compounds retained in solution is directly affected by the available oxygen supplied by the Na/concrete interaction. The preliminary review of the CRBRP safety case for TMBDB-SMBDB[8] by SNLA indicated that this could cause a large release of plutonium to the pool. Analogous chemically induced release of Eu, Ru, and Sr fission products is suggested by the available literature. (The VANESSA model[9] is currently under

development at SNLA to calculate melt chemistry and vaporization release of various compounds during core melt/concrete interaction. An extended version of the model is planned for treating sodium pools. Sodium urano-plutonate chemistry and subsequent vaporization release will be included). Material released into the pool is subject to release from the pool by bubble bursting.

An additional unknown is the effect of (potentially violent) core debris/concrete interactions on aerosol generation. As explained above, material is transported to the pool surface and released by gas bubbles generated by the interaction at the pool-concrete interface. It has been argued that aerosols produced at the bottom of the pool are trapped as they are carried through the sodium pool. The amount of aerosol trapped by bubbling aerosol ladden gas through a liquid is dependent on particle size and bubble size. A simple bubbling model developed by Fuchs[10] illustrates this. Figure 2 shows the "decontamination factor" (defined as concentration of aerosol entering the pool divided by the concentration emerging from the pool) as a function of particle size for several bubble diameters in water. Solid lines in this figure show decontamination when gas within bubbles circulates. The dotted lines refer to decontamination when gases within the bubbles do not circulate. Circulation of gas within the bubble can be stopped or slowed by particulate material depositing on the gas-liquid interface. (Quinn, et al.[6], Powers, et al.[11]). The figure illustrates that by changing particle size, bubble diameter and/or interfacial contamination, the amount of aerosol trapped in a sodium, pool can change substantially. For adverse conditions, it is possible then that substantial quantities of the aerosols produced by debris interaction with concrete could emerge from the sodium pool and enter containment. Further, material removed from the bubble is released into the sodium pool and subject to release by the mechanisms discussed above.

Assuming 100% release of the volatile fission products early in the accident may not be a conservative approach. The TMBDB assumption of rapid total release of the volatile fission products means the aerosol initially in the CRBR containment contains all of this material. The production of additional aerosols by the boiling sodium pool leads to "wash out" of the initial aerosols by agglormeration and settling processes. This scavenging effect is illustrated by the CONTAIN[12] calculation presented in figure 3. An aerosol initially present in containment at $1 g/m^3$ is depleted over a period of less than 1 hour to a concentration of about $2 \times 10^{-4} g/m^3$ when an intense source of addition aerosol is active.

The rapid depletion of the fission product aerosols means the filtered vent system will not have to contend with the volatile fission products if they are assumed to be quantatively released very early in an accident.

Had a slower release of volatile fission products been assumed, then a higher concentration of volatile fission products would be present in containment at later times when the vent system is operational.

The question of whether or not the vents between the reactor cavity and the containment will plug has not been resolved. A simplified model of tube plugging formulated by Vaughn[13] has been used to assess the problem. The model gives the mass input to plug, M, as a function of the tube diameters, and an empirical constant, K:

$$M = KD^3$$

The following table summarizes the model's predictions.

D (cm)	(kg)
10	1 - 5
30	30 - 140
100	1×10^4 - 5×10^4

In CRBR, there are 5×10^4 kg of sodium and two 30 cm diamter vents between the reactor cavity and containment. The potential for plugging of these vents exists. Plugging of lines up to 10 cm in diamter has been observed in sodium/concrete interaction experiments involving only a few hundred kg of sodium.

The Vaughn model makes several simplifying assumptions about key parameters which are treated by other conventional deposition theories (e.g. Ingham[14] or Wood[15]). These are listed in Table I

Selection of the parameter K from experimental data implicitly assumes the dependencies listed in Table I are averaged for that data set. Application of the model to problems different from the data base is not appropriate. The model can be used to fit other data but adjustments of the parameter K for different conditions without supporting data is conjecture. The model is independent of everything but vent size and mass input to the vent. It is not known

how different conditions increase or decrease K. It is not known if different conditions enhance or retard plugging. To use this model for vent plugging, tests must be done in which vent size, flow regime, temperature, gas properties and aerosol distribution and mass loading are prototype.

Discontinuities in the flow pathway have been found to enhance deposition and plugging. Material remaining from a failed rupture disc (or values in the filtered vent system) may cause plugging earlier than expected.

A more mechanistic treatment of plugging can be done with currently available deposition models which include treatment of the parameters discussed above. Fairly simple qualification tests on large scales are also quite feasible.

We hope the above discussion of aerosols is of use to you. If you have further questions, please contact us.

Sincerely yours,

Erik Randich

John E Brockmann

Erik Randich and John E. Brockmann
Reactor Containment Safety Studies
Division 9422

ER/JEB:9422:jp

Copies to:
9422 D. A. Powers

TABLE I

PARAMETER	VAUGHN	OTHER DEPOSITION MODELS
Flow Rate	Independent	Determines deposition quantity; residence time in tube over which deposition occurs.(14,15)
Flow Regime	Independent	Laminar and turbulent flows have difficult deposition mechanisms active. Turbulent flows can give higher deposition rates for 1 to 10 μm sized particles.(14,15)
Particle Size	Independent	Deposition rates a strong function on particle size in some size ranges and flow regimes. Large particles in turbulent flow: deposition rate particle size, small particles in laminar flow: deposition particle size squared.(14,15)
Pipe Roughness	Independent	Deposition rates can vary over 2-1/2 orders of magnitude for variation of pipe roughness over the range of hydraulically smooth pipes.(15)
Temperature and Gas Properties	Independent	Deposition rates functionally dependent on on these quantities.(14,15)

REFERENCES

1. Muhlstein, L. D. and Colburn, R. P.; "Aerosol Release from Sodium-Concrete Reactions", October, 1982.

2. Lee, K. W.; "Maximum Penetration of Aerosol Particles in Granular Bed Filters", Journal of Aerosol Science, Vol. 12, p. 79, 1981.

3. Kress, T. S. and A. L. Wright, "LMBFR Aerosol Release and Transport Program quarterly Progress Report for July-September, 1978", NUREG/CR-0482.

4. Jordan, S. and Y. Ozawa; "Fuel Particle and Fission Product Release from LMFBR-Core Catcher", in "Fast Reactor Safety and Related Physics, Chicago, October 508, 1976," CONF-761001, 1976, pp. 1924-1929.

5. Tomaides, M. and K. T. Whitby, "Generation of Aerosols by Bursting Single Bubbles", FINE PARTICLES; 13.7.11 Liu, ed. p. 235, Academic Press, Inc.; New York, 1976.

6. Quinn, J. A., R. A. Steinbrook, and J. L. Anderson, "Breaking Bubbles and the Water-to-Air Transport of Particulate Matter", Chemical Engineering Science, Vol. 30, p. 1177 (1975).

7. Adamson, et al., Journal of Nuclear Materials, 97, 203 (1981).

8. "A Preliminary Review on the CRBRT Safety Case for -TMBDB - - SMBDB", Staff, Advanced Reactor Safety Research Program, Sandia National Laboratories, December 1981.

9. VANESSA - reference NUREG 0956, Appendix C.

10. Fuchs, N. A.; "The Mechanics of Aerosols", Pergamon Press, November, p. 240, 1964.

11. Powers, McNiel, Subramanian, and Cole, Journal of American Ceram.Soc., Vol. 65; 6 p. 289, June .1982.

12. Senglaub, M. E.; J. P. Odom; M. J. Clauser; J. E. Kelly and P. S. Pickard, "CONTAIN, A Computer Code for the Analysis of Containment Response to Reactor Accidents - Version 1A", Draft Copy, NUREG/CR-2224, SAND81-1495.

13. Vaughn, R U, "Aerosol Leakage Model", Quarterly Progress Report, LMFBR Safety Program October-December 1977, Atomics International Rep. AI-DOE-13223, 1978.

14. Ingham, D. B., "Gravity Settling of Aerosol Particles in Horizontal Rectangular Tubes with Some Diffusion", Journal of Aerosol Science, Vol. 8, p. 139. 1977.

15. Wood, N. B., "A Simple Method for the Calculationm of Turbulent Deposition to Smooth and Rough Surfaces", Journal of Aerosol Science, Vol. 12, No. 3, p. 255, 1981.

FIGURE 1.

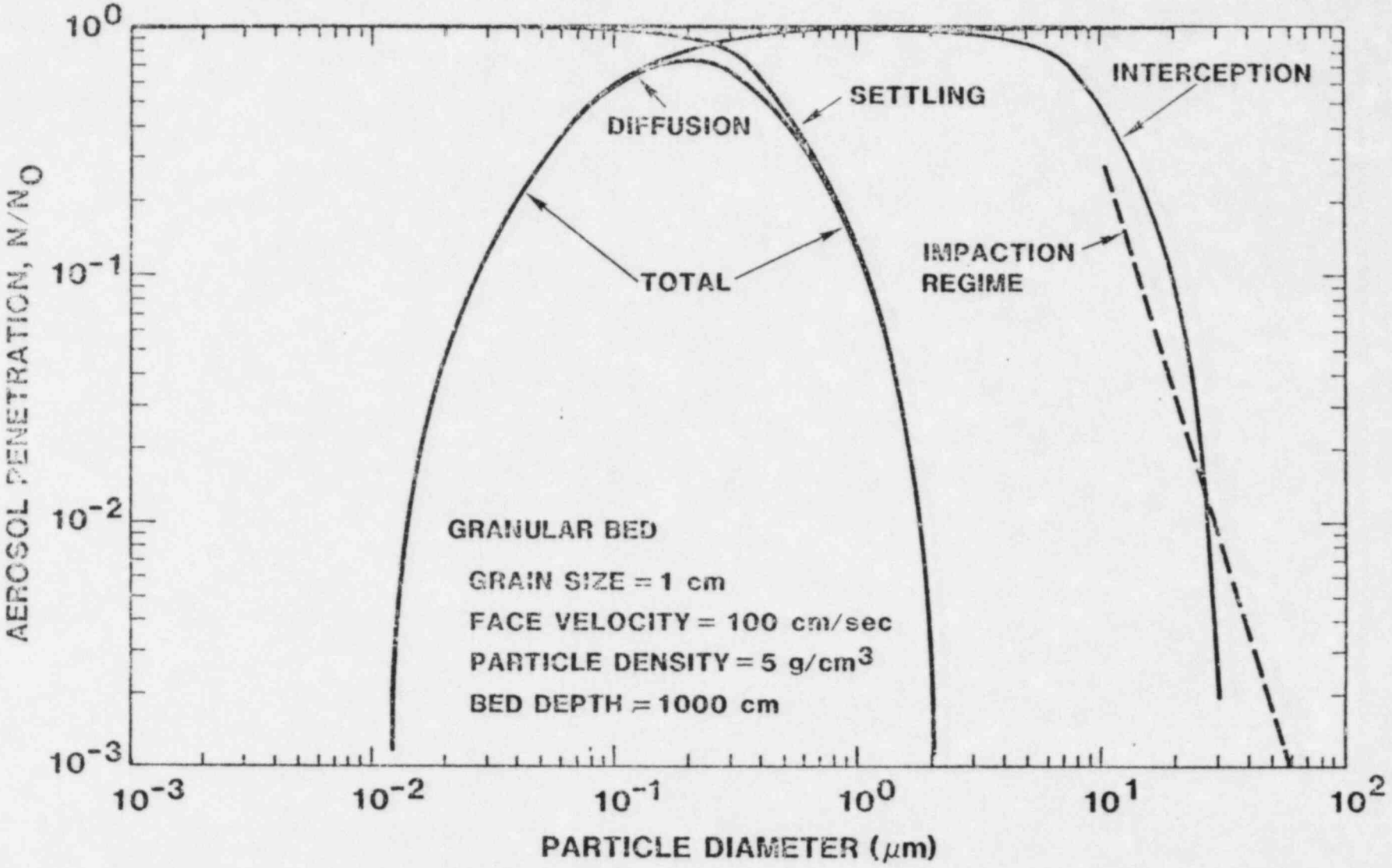
AEROSOL PENETRATION, N/N_O
10^0
10^{-1}
10^{-2}
10^{-3}
DIFFUSION
SETTLING
INTERCEPTION
TOTAL
IMPACTION REGIME
GRANULAR BED
GRAIN SIZE = 1 cm
FACE VELOCITY = 100 cm/sec
PARTICLE DENSITY = 5 g/cm^3
BED DEPTH = 1000 cm
10^{-3}
10^{-2}
10^{-1}
10^0
10^1
10^2
PARTICLE DIAMETER (μm)

Figure 2.

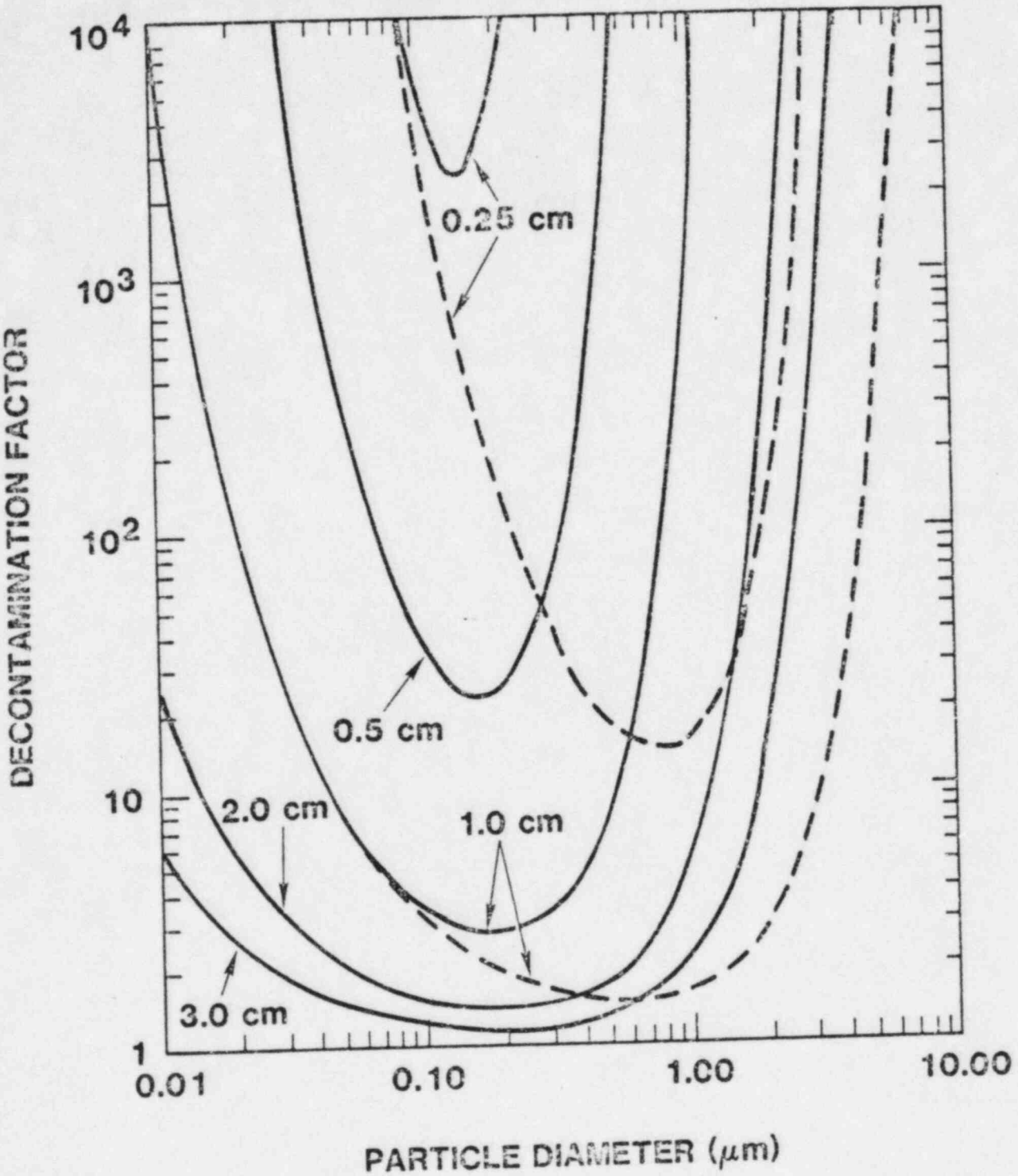
DECONTAMINATION FACTOR
10^4
10^3
10^2
10
1
0.01
0.10
1.00
10.00
PARTICLE DIAMETER (μm)
0.25 cm
0.5 cm
2.0 cm
1.0 cm
3.0 cm

Figure 3.

Washout of Primary Aerosols by Secondary Aerosols