

Alternative Igneous Source Term Model for the Yucca Mountain Repository

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Abstract – *The Nuclear Regulatory Commission uses the ASHPLUME model in its evaluation of the basaltic volcanism scenario at the possible Yucca Mountain repository. The mixing of magma with the spent-fuel waste form is tied to a reasonable but unverified model that predicts that no dense ash/fuel particles would form. This paper describes an alternative model using a mixing rule that allows the formation of dense ash/fuel particles that would be transported in the volcanic plume differently. The alternative model shows significant sensitivity to the spent-fuel particle size distribution. However, differences in results between the two models are on average less than a factor of two.*

I. INTRODUCTION

Basaltic volcanism at the possible high-level waste repository at Yucca Mountain has the potential for damaging waste packages, releasing radioactive waste, and transporting it to the environment. In preparation for the review of a potential license application for this repository, the Nuclear Regulatory Commission (NRC) is investigating the mechanisms and consequences of volcanism at this site. The NRC is focusing on two volcanic scenarios; (1) intrusive volcanism, for which magma comes into contact with the waste packages, but does not reach the surface. Radionuclides released from the damaged waste packages would reach the accessible environment only through the groundwater pathway; and (2) extrusive volcanism, for which magma entrains radioactive waste, releasing it at the earth's surface in an ash plume. Of the two volcanism scenarios, NRC's results indicate that risk is greater for extrusive volcanism. The present paper deals only with extrusive volcanism.

Although the probability of extrusive volcanism at the possible repository is very small (current NRC estimate is 10^{-7} per year¹), consequences to the potentially exposed group may be larger (up to several hundred mSv per event) than from other, more likely pathways. The NRC currently uses the ASHPLUME model² to evaluate this low-probability event. This model assumes that radioactive waste released from affected waste packages is incorporated into magma, and upon eruption, volcanic ash and radioactive waste is transported in a plume downwind. Once on the ground, the deposited ash and spent fuel would become a source of radioactive contamination, entering the human pathway primarily through inhalation of resuspended dust, and to a lesser extent, through groundwater and ingestion pathways.

Models for the volcanism scenarios are part of a risk-assessment computer code³, which must be run for a large number of probabilistic realizations. This requirement limits the complexity of useful models to generally analytical solutions or those abstracted from more-complicated models run off line.

The volcanic scenario has three main components: (1) interaction of intruding magma with the engineered materials and structures; (2) transport of the radionuclides in the contaminated ash to the biosphere; and (3) exposure of the affected individual or group from the contaminated ash. The first component, interaction of magma with engineered materials and structures, is not well understood, and there are few if any direct examples that are useful for formulating a model. Some useful information can be drawn from real and simulated nuclear reactor accidents, and from experiments with the spent fuel waste form, but these are far from ideal analogies to the volcanic case.

The second component, transport of contaminated ash, is perhaps the best-understood part of the scenario because volcanic plumes and ash deposition can be studied from actual volcanic eruptions. Nevertheless, the models of ash transport used in NRC's analysis are highly simplified for the sake of the probabilistic risk codes, and do not consider phenomena such as changes in wind direction and speed with time and position, nor processes such as Brownian motion that prevent the finest ash particles from settling quickly. Furthermore, the possible transport through processes of wind and water erosion of contaminated ash deposited on the ground is not directly modeled.

Although the NRC uses dose assessment models for a variety of radionuclide release situations in both the high-level waste and decommissioning areas, there are few direct analogies to pathways of human exposure from radionuclides deposited by contaminated volcanic plumes.

The lack of good analogies for the volcanism models has led NRC to develop models that generally err on the pessimistic side. The current paper focuses on the front end of the scenario; *i.e.*, the interaction of the magma with buried waste and its incorporation into ash. The current NRC model of this phenomenon assumes that all waste packages contacted by magma are destroyed, waste mixes with the magma, and becomes part of the ash plume released at the surface of the earth. The current mixing model is based on a reasonable, but unverified relationship between ash particles size and fuel particle size. The analysis presented here attempts to refine the bases for estimating how waste and magma interact to determine if an alternative model of this interaction materially affect the computed consequences.

II. BACKGROUND INFORMATION OF PROPERTIES OF INVOLVED MATERIALS

In developing an alternative model for waste incorporation, we first examined some of the underlying behavior of the involved materials. We confined our work to the dominant waste form, spent UO₂ fuel from commercial reactors:

II.A. Direct Physical Effects of Magma Interacting with the Waste Packages and Spent Fuel Waste Underground

Temperature of the magma may be as high as 1200 °C. Although magma temperature is lower than the melting points of steel and nickel alloy, it may weaken these materials to the point that shear forces from the viscous magma moving with sufficient velocity could break the waste packages apart. Although there is no direct experimental evidence on the survivability of waste packages in such an environment, we conservatively assume that all waste packages contacted by magma will be destroyed.

Spent fuel within the waste packages will be protected by zirconium alloy cladding that has a high melting point (>2500 °C), but is thin and therefore not physically strong. The long, thin fuel rods and supporting structures in the fuel assemblies would also be subject to shear forces from the magma, and are assumed to fail upon waste package failure.

Spent fuel within the fuel rods is a hard ceramic material with a high melting point (>2800 °C), that is unlikely to be affected directly by the magma temperature.

The size distribution of spent fuel may be important to the model. Originally, the model assumed a distribution determined from finely crushed fuel samples. A considerably coarser spent fuel size distribution could be based on other information. For example, experiments in which fuel was subjected to forces equivalent to a 120 mile per hour truck accident showed relatively minor breakup⁴, with more than 90% of mass in diameters greater than 1000 microns. Fuel particles suspended in moving magma may not undergo enough shear to crush particles to finer sizes, although other forces such as rapid degassing might have an effect. Large thermal shocks to fuel from rapid quenching in water (800 °C/second) can cause fuel to break up into smaller fragments, but not a fine powder⁵. The rate of temperature change in the volcanism scenario would be moderated by the large mass of the waste packages. Temperature change rates are likely to be comparable or less than those to which fuel would be exposed during reactor power-up.

II.B. Oxidation of Fuel Underground

UO₂ can oxidize rapidly in air at high temperature, becoming a fine powder⁶. However, basaltic magma is highly reducing.

Data from simulated and actual reactor accidents⁷ indicate that the rate of oxidation under reducing conditions is slow relative to the time that the fuel would be initially suspended in the magma and released to the atmosphere (minutes to hours).

II.C. Dissolution of UO₂ in Magma

Uranium dioxide is soluble in molten basalt up to about 20% at magma temperatures⁸. However UO₂ moving with magma will have little relative velocity; *i.e.*, the only mechanism for fuel to dissolve in magma is by diffusion, which is slow on the time scale of interest. Evidence of slow diffusion in magma is the presence of xenoliths, some of which move tens of kilometers in magma and remain intact, even though they are nearly melted. UO₂ particles would not melt at these temperatures. Fuel particles at the small end of the size range might be affected however, since diffusion is relatively more important for small particles than it is for large ones. We did not specifically account for removal of small particles by dissolution, but the likely effect of this dissolution will be discussed in the conclusions.

II.D. Chemical reaction between zirconium and spent fuel

Zirconium alloy cladding and UO₂ fuel can react chemically at temperatures above 1000 °C, although most data from real and simulated reactor accidents are at temperatures much hotter than magma⁹. Reactions require close contact between the cladding and fuel by imposition of high pressure. Products formed are uranium metal and solid solutions of oxygen in zirconium. Maximum amounts of reaction are about 9% of fuel mass, based mostly on the availability of zirconium. Uranium and UO₂ form eutectics with zirconium and ZrO₂ that could melt at temperatures approaching those of magma, but these are likely only for high-temperature reactions. A source term model could account for a fraction of the fuel that has undergone chemical reaction leading to release of uranium metal and radionuclides to the magma. It is unclear, however, if this would be any worse than the direct incorporation of the fuel into the ash.

II.E. Alteration of fuel above ground

Once ejected at the surface, UO₂ particles would start to oxidize when they come into contact with air. The rate of oxidation in the plume would be controlled by temperature and the availability of oxygen. Products would be oxides like

U₄O₉, U₃O₈ and UO₃. It is likely that only a small fraction of the spent fuel would oxidize in the plume because of the short time it would be aloft, and the rapidly decreasing temperature with distance from the vent. Furthermore, the ash itself might protect the UO₂ from oxygen, depending on its porosity. Once deposited on the ground UO₂ will degrade on a longer time scale at a rate controlled by temperature, oxygen availability and constituents such as silica, water and CO₂. Although there have been measurements on the resuspension of ash itself¹⁰, it is unclear without further experimental evidence, how much of the spent fuel and radionuclides in the contaminated ash would become airborne. The current model assumes simply that airborne dust is created by mechanical forces from ash of any size deposited on the surface, and at the concentration in the ash.

III. NRC'S CURRENT SOURCE TERM MODEL

The current source term model for extrusive volcanism relies on a reasonable, but unverified, relationship for the mixing of waste and magma. The current model assumes that spent UO₂ fuel and ash mix according to an "incorporation ratio" ρ_c , which stipulates that an ash particle of diameter D_a can incorporate fuel particles with a diameter $10^{-\rho_c} D_a$ or smaller. One of the desirable features of the current incorporation model is that it allows the treatment of the ash/fuel mixing as a direct analytical function that is easily incorporated into the Suzuki model¹¹ for ash transport. The Suzuki model depicts the dispersion of ash particles from a volcanic plume into a steady wind field:

$$X(x, y) = \int_{\rho_{\min}^a}^{\rho_{\max}^a} \int_{z=0}^H \frac{5QP(z)f(\rho^a)}{8\pi C(t+t_s)^{5/2}} \cdot \exp\left[\frac{5\{x-ut\}^2 + y^2}{8c(t+t_s)^{5/2}}\right] d\rho dz \quad (1)$$

where $X(x,y)$ = mass of ash per unit area accumulated at location x, y , $\rho^a = \log_{10}$ of ash particle diameter, $\rho_{\min}^a, \rho_{\max}^a$ = range of \log_{10} particle size, z = vertical distance from ground surface, H = height of eruption column above vent, Q = total quantity of erupted material, $P(z)$ = distribution function for particle diffusion out of column, $f(\rho^a)$ = distribution density function of ash particles, c = constant related to eddy diffusivity and fall time of particles, t = particle fall time, t_s = particle diffusion

time in eruption column, and u = wind speed. The distribution function $P(z)$ and particle fall time t are derived in Suzuki¹¹, and presented in Jarzempa², and will not be repeated here. The important point is that they are functions of the terminal velocities of the particles, which in turn depends on the specific gravity of the particles.

Jarzempa modified the Suzuki model to predict the concentration of fuel mass to ash mass, $FF(\rho_a)$:

$$FF(\rho^a) = \frac{U}{Q} \int_{\rho=-\infty}^{\rho=\rho^a} \frac{m(\rho - \rho_c)}{1 - F(\rho)} d\rho \quad (2)$$

where U = total mass of fuel ejected, m = probability density function of fuel mass, and $F(\rho)$ = cumulative distribution of the ash probability density function $f(\rho)$. Development of Eq.(2) required a relationship between ash and fuel that was single-valued; *i.e.*, there was a direct relationship between ash particle size and fuel concentration.

IV. ALTERNATIVE CONCEPTUAL SOURCE TERM MODEL

While simplifying the calculation of the contaminated ash plume significantly, the incorporation ratio model did not allow for the possibility of a range of fuel concentrations for a given ash particle size. It is likely that mixing between ash and fuel would be highly heterogeneous, leading to a wide range of fuel to ash ratios for a given ash particle size. Since fuel is so dense compared to the ash, it is also likely that a fraction of the ash particles would be of high density, and have settling rates higher than lighter ash particles of the same diameter.

IV.A. Alternative Fuel Incorporation Model

NRC has developed an alternative fuel incorporation model that examines the possibility of heterogeneous mixing between the fuel and ash in the volcanic scenario. The new model is based on “parsimony”; *i.e.*, since the actual process of fuel incorporation is unknown, we make the minimum number of assumptions. For this model, the main assumption is that the fraction of mass of fuel incorporated into the ash is proportional to the mass of the ash. Although the assumption is simply stated, implementing it into a model proved to be challenging. The main difficulty lies in expressing the large range of

possible outcomes for mixed ash/fuel particles. For the problem at hand, the total mass of ash and the number of ash particles greatly exceed the fuel mass and number of fuel particles. Also, the size range of ash particles will be much greater than the corresponding range for fuel particles. The two distributions for fuel particle diameters, and a typical value for ash particle diameter is shown in Fig. 1. This did not prove to be a problem with the original incorporation model, which relied directly on smooth distributions of ash and fuel particles.

The alternative model worked with a direct simulation approach which used a relatively small number of “representative” fuel and ash particles in discrete size ranges. It used the Suzuki model¹¹ for ash transport, Eq. (1), but replacing the outer integral by a summation of discrete particle sizes:

$$X(x, y) = \sum_{L=1}^{NI} \Delta m_L \int_{z=0}^H \frac{5QP(z)f(\rho^a)}{8\pi C(t+t_s)^{5/2}} \cdot \exp\left[\frac{5\{x-ut\}^2 + y^2}{8c(t+t_s)^{5/2}}\right] dz \quad (3)$$

where ρ^a_i = the \log_{10} diameter of the i^{th} ash particle bin, Δm_L = the mass of ash/fuel particles associated with the L^{th} “indicator particle” (defined later), and NI is the number of indicator particles.

The algorithm for mixing the fuel with the ash was not so straightforward, mainly because there was often more than 5 order of magnitude difference between the total number of ash and fuel particles. In order to treat all possible combinations of fuel and ash directly by simulation, there would have to be more than 10^5 representative particles. Dealing with such a large number of particles would be computationally demanding, both in computer storage and run time. This was especially true since the overall model is a small part of a much-larger Monte Carlo model for performance of the repository³.

A solution to the problem of excessive number of representative particles was to consider three classes of ash/fuel combinations, in which the discriminant was the ratio of number of fuel particles to an ash particle. First, the distributions of the fuel mass and ash mass were discretized into 100 bins, each bin representing a range of fuel or ash sizes. The bins were determined by dividing on a logarithmic scale the log of the diameter into 100 equal divisions. The mass and true number of particles in each bin were then calculated.

Next, there were a large number (approximately 20,000) “indicator” particles defined to represent the ash particles. The number of these indicator particles was apportioned to each of the 100 ash bins according to the relative mass in each ash bin.

Each of the indicator particles represents initially an approximately equal mass of ash. The indicator particles also represent the mass of an ash particle, its fuel content after mixing, and the number of actual ash particles.

The probability of a fuel particle of size j being incorporated into an ash particle of size i is equal to the fraction of the overall ash mass contained in ash bin i . This leads to a Poisson distribution for the number of fuel particles per ash particle.

Since each indicator particle represents an approximately equal quantity (mass) of ash, there are a varying number of indicator particles in each ash bin. For each ash bin, we cycle through all of the fuel bins to determine the ratio μ (which is also the parameter of the Poisson distribution) = number of fuel particles of size j to number of ash particles of size i . If μ is large (*i.e.*, $\mu > 20$), then there would be a nearly uniform number of fuel particles per ash particle. In this case, all of the indicator particles in the ash bin get an equal number of fuel particles of size j added to them.

For smaller μ (*i.e.*, $20 < \mu < 0.1$), there would be a variable number of fuel particles per ash particle, determined from a Poisson distribution with parameter μ . The indicator particles in the i^{th} ash bin are sampled to determine how many fuel particles of size j would be added, some getting few or none, and some getting multiple additions of fuel. The algorithm is as follows:

1. For each ash bin i , calculate the probability \mathbf{Pr} from the Poisson distribution:

$$\mathbf{Pr} = \frac{e^{-\mu} \mu^p}{p!} \quad (4)$$

where p = number of ash particles of size j to be added, ranging from 1 to 50.

2. The number of fuel particles of size j added to ash bin i , $Np_{i,j}$, would be the number of indicator particles in that ash bin times the probability;
3. Randomly sample each indicator particle in the ash bin without replacement to determine whether it gets p fuel particles until all $Np_{i,j}$ fuel particles are gone.
4. Repeat for p up to 50 fuel particles per ash particle.

For small μ (*i.e.*, $\mu < 0.1$), there would be no more than one fuel particle per ash particle. In this case, there would be a

second category of indicator particles (“small μ indicator particles”), which stand apart from the regular indicator particles. These contain exactly a single fuel particle attached to each ash particle. The “small μ indicator particles” represent a quantity of ash particles equal to the number of fuel particles of size j , apportioned from the total fuel by the mass fraction of ash of size i . An equal number of ash particles are subtracted from the other indicator ash particles in each bin to maintain an overall mass balance of ash.

The algorithm is repeated for all combinations of ash and fuel particle sizes. The particle sizes, densities and fuel content, represented by all indicator particles, are then used with Eq. 3 to predict the ash-layer thickness and fuel concentration at the down-wind location.

The mixing of fuel with ash would be determined by the mechanical, thermal and chemical processes discussed above. However, these relationships have been inferred from incomplete observations of analogous data, and there is little, if any direct experimental support. For this reason, we performed conservative (*i.e.*, pessimistic) calculations assuming that all fuel in the affected waste packages would be incorporated into magma, and for two different size ranges of the spent fuel particles differing by a factor of 10, with the expectation that uncertainties in the mixing rules and other factors would be encompassed by the two sets of results.

V. RESULTS OF ALTERNATIVE MODEL

The alternative model was compared to the current model for a set of 100 random vectors chosen by Latin Hypercube Sampling from distributions typical of NRC’s recent performance assessments³. A constant 100 waste packages are assumed to be involved in the event, which is toward the high end of the expected range. Table 1 shows the input parameters to the TPA³ code relevant to the volcanism source term model. The comparison consisted of a presentation of the peak dose, vector-by-vector, for the two models assuming the volcanic event occurred at 1000 years after repository closure. It should be stressed that the actual calculation of risk from the NRC performance assessment model considers the low probability of volcanic events, averaging dose over the realizations, and a convolution of events over time to generate a mean dose curve³. The figures presented here are for peak doses from a single event time, unweighted by event probability, and do not portray risk correctly. Neither of these factors is important to the comparisons, however, which only intend to show the relative doses for the two models.

Figure 2 shows the particle density versus ash particle diameter for the median dose vector and the coarse fuel distribution,

predicted for both the current and alternative models. As expected, the alternative model predicts a wide range of particle densities, whereas the current model predicts only a single particle density for a given ash particle size. This figure shows the range of particle densities, but not their quantity, and histograms of particle mass for the two models would be nearly indistinguishable because of the overwhelming mass of ash compared to fuel. Nevertheless, it is interesting to note in Table 2 that in many cases, the alternative model predicted a significant portion of the fuel would exist in “dense” particles, defined here arbitrarily as particles with densities greater than 2.0 grams/cm³. The current model does not predict any “dense” particles.

Figure 3 shows the first model comparison for the 100 peak doses using the default, “fine” fuel particle-size distribution. The alternative model consistently predicts a smaller peak dose. The average of the peak doses was 74% higher for the current NRC model.

Figure 4 shows the model comparison for the “coarse” fuel-size distribution, which increased the particle sizes by a factor of 10. Interesting, some of the alternative runs predicted higher peak doses than the current model, although the average peak doses for the current

Table 1 – Volcanic Parameters and Probability Distributions Relevant to Source Term Models

Number of waste packages impacted = 100
Volcanic event duration = uniform ($1.8 \times 10^5 - 1.3 \times 10^6$) seconds
Volcanic event power = uniform ($3.5 \times 10^9 - 5.3 \times 10^{11}$) watts
Maximum raw ash density = 1.6 gm/cm^3
Minimum raw ash density = 0.8 gm/cm^3
Mean ash particle size = logtriangular (0.01, 0.1, 1.0) cm
Ash particle size \log_{10} standard deviation = 1.0
Fuel particle size, fine = logtriangular (0.0001, 0.001, 0.01) cm
Fuel particle size, coarse = logtriangular (0.001, 0.01, 0.1) cm
Wind speed, cm/sec = exponential ($\mu = 0.0083$)
Wind direction – directly toward exposed population

Table 2 – Fraction of Fuel in “Dense” Ash Particles from Alternative Model

	Fine Fuel Distribution	Coarse Fuel Distribution
Minimum	0.013	0.09
Mean	0.088	0.29
Maximum	0.27	0.6
St. Dev.	0.059	0.13

model were 18% higher than the alternative model. Also, the mean dose for the alternative model *increases* for the coarser fuel distribution, whereas the mean dose *decreases* for the current model. Although these results were somewhat surprising, the fact that a particle is denser and therefore falls faster does not automatically lead to a result of a smaller peak dose. Under certain conditions, particles that might have passed over the location because they didn't settle fast enough now land in the target area, contributing to a higher peak dose.

VII. CONCLUSIONS

Preliminary results for the alternative conceptual model for fuel incorporation into ash predict that a significant fraction of spent fuel would be incorporated into dense particles, but that most of the fuel would be incorporated into the bulk of the ash and has essentially the same density as virgin ash. The fraction of spent fuel in dense particles is sensitive to the size distribution of the spent fuel. Most of the dense ash/fuel particles are at the small end of the particle-size range.

No direct experimental evidence exists to determine how magma and the waste form would interact, but analogous data exist for volcanism in general and nuclear reactor research and accidents. Mixing of ash and spent fuel would be sensitive to mechanical, chemical and thermal conditions involving the interaction of magma and the engineered waste form. These factors influence the way in which the fuel would be incorporated in the ash and its degree of heterogeneity. The present NRC model is not very sensitive to considerations of mixing of fuel and ash, but the alternative model is more sensitive. The alternative model shows a significant difference in results when the fuel-size distribution was changed by a factor of 10. Nevertheless, differences between the present NRC model and the alternative model are not large (on average, less than a factor of 2) compared to the many other uncertainties in the modeling of volcanism, lending credibility to the original fuel incorporation model.

The dissolution of spent fuel into magma has not been taken into account specifically, but it is possible that particles at the small end of the size spectrum would be more affected than larger particles. Fuel dissolved directly into the magma would end up somewhat uniformly dispersed into the ash, probably independent of the ash particle size. If this were the outcome, dissolution would likely drive the results closer to the original incorporation ratio model, which tends to give a more uniform concentration of fuel in ash than the alternative model. Therefore, our conclusion that the differences between the original and alternative models are not large is still likely to hold.

VIII. DISCLAIMER

The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of a license application for a geologic repository at Yucca Mountain.

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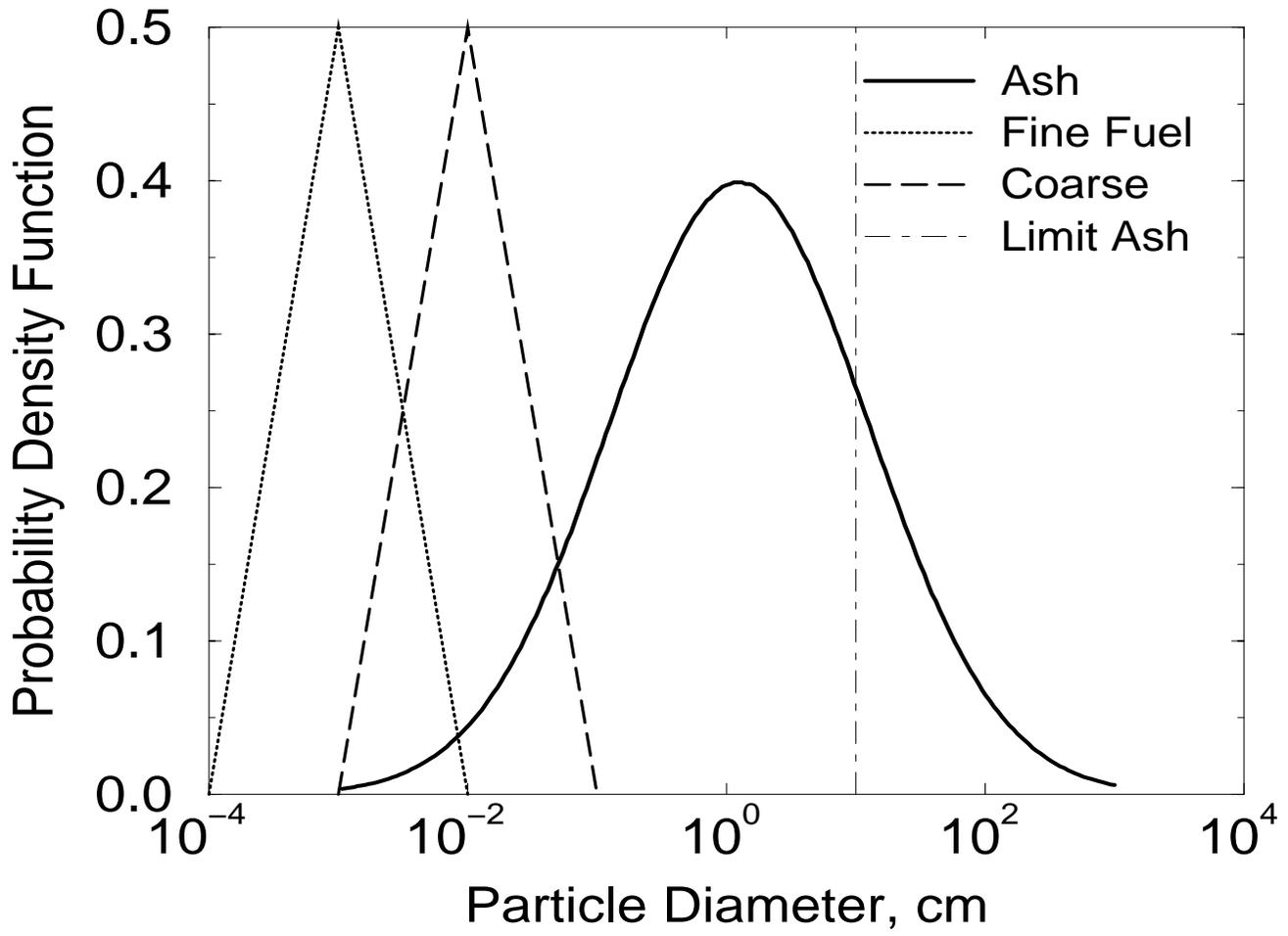


Figure 1 – Ash and Fuel Particle Size Distributions

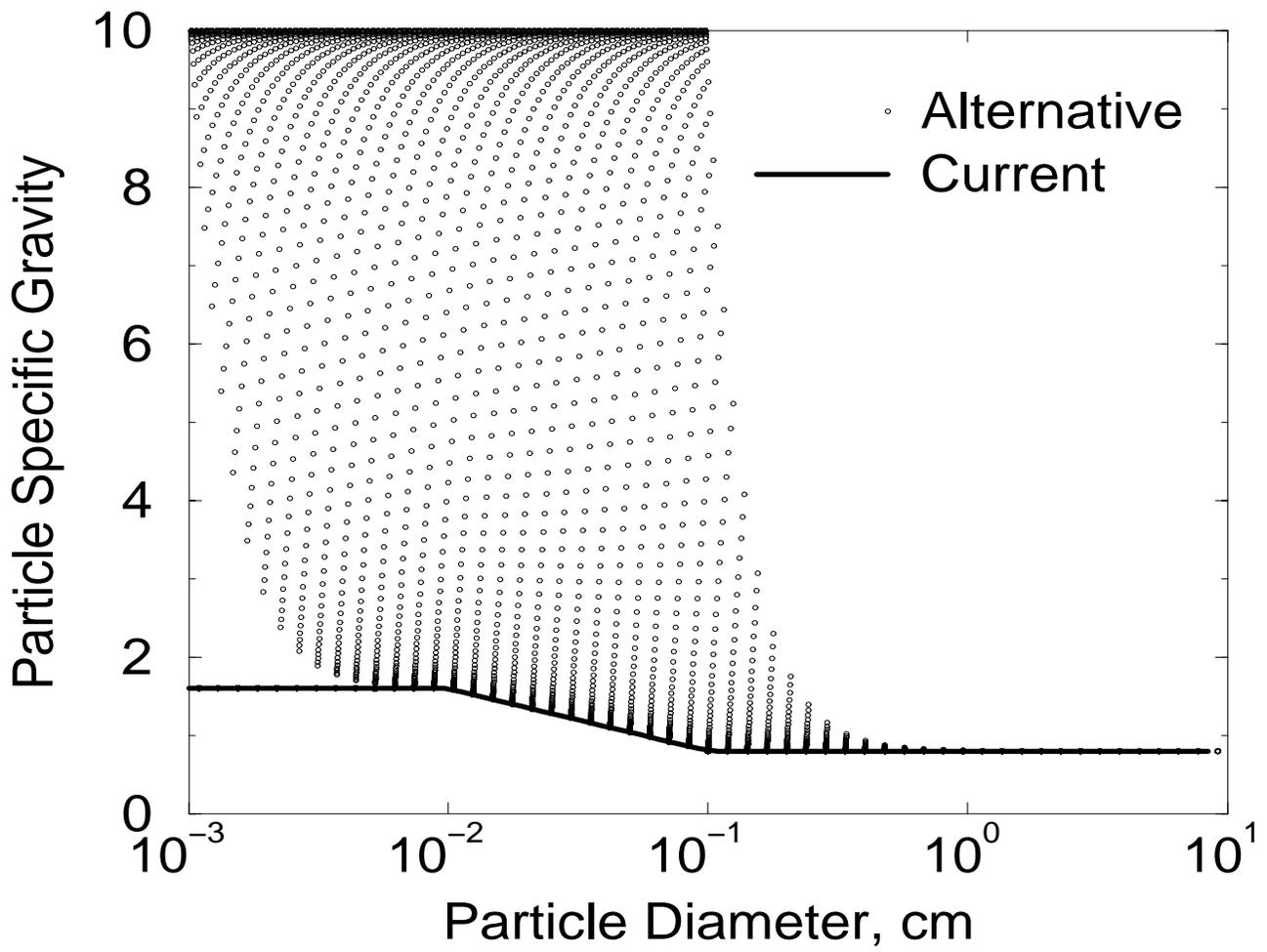


Figure 2 – Specific Gravity for Ash/Fuel
 Particles in Alternative and Current Model – Coarse Fuel, Median Dose Vector

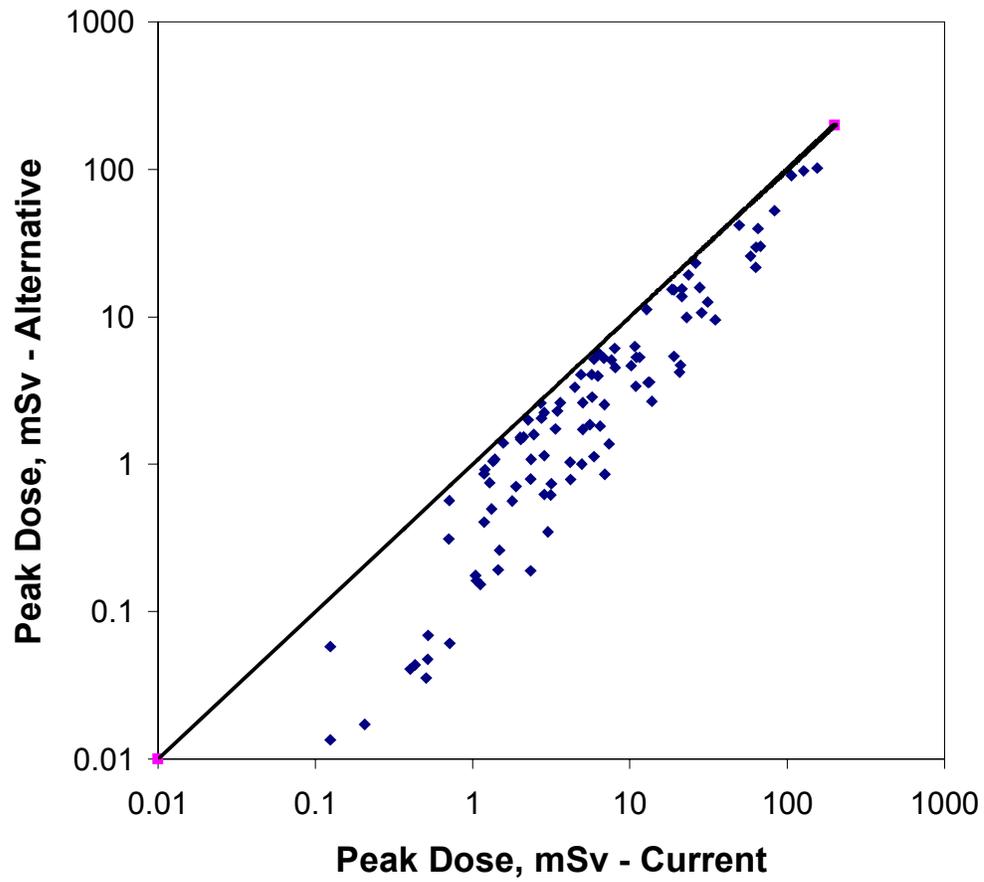


Figure 3 – Peak Dose, Alternative vs. Current Model, Fine Fuel

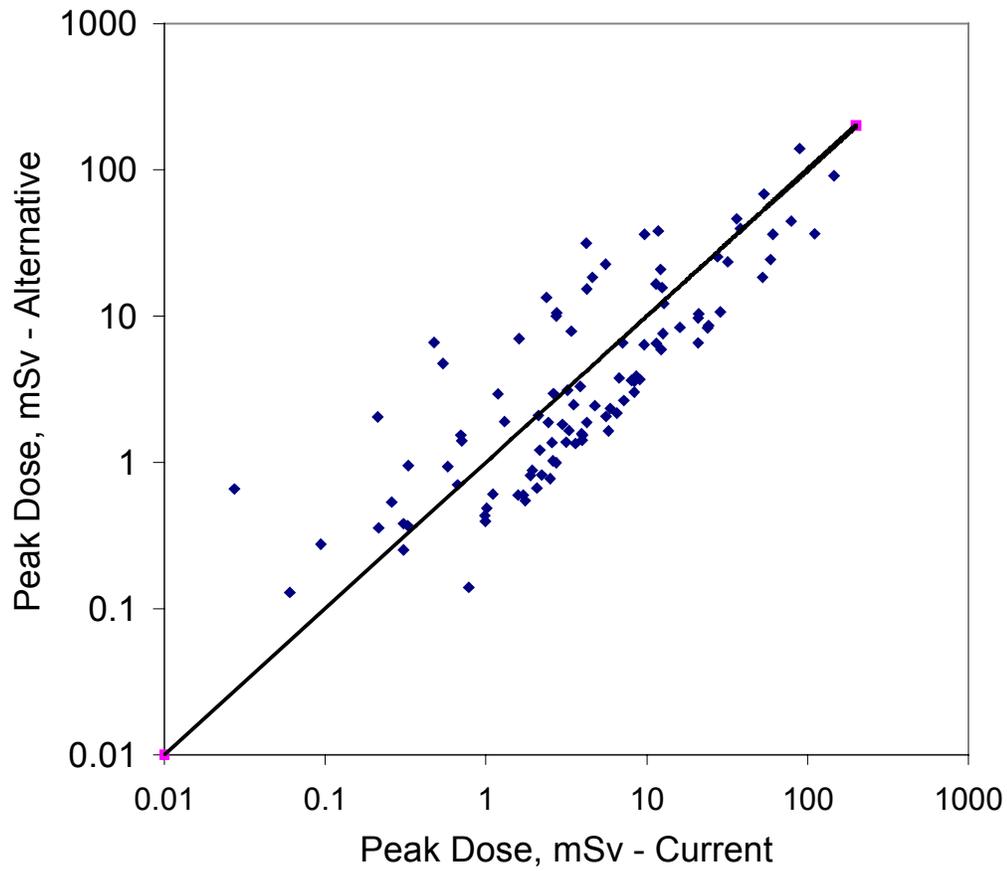


Figure 4 – Peak Dose, Alternative vs. Current Model, Coarse Fuel