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ACCIDENT ANALYSIS FOR THE LOW-LEVEL MIXED WASTE "NO-FLAME" OPTION IN THE U.S. DEPARTMENT OF ENERGY WASTE MANAGEMENT PROGRAMMATIC ENVIRONMENTAL IMPACT STATEMENT[1]

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ABSTRACT

This paper outlines the various steps pursued in performing a generic safety assessment of the various technologies considered for the low-level mixed waste (LLMW) "No-Flame" option in the U.S. Department of Energy (DOE) Waste Management Programmatic Environmental Impact Statement (WM PEIS). The treatment technologies for the "No-Flame" option differ from previous LLMW technologies analyzed in the WM PEIS in that the incineration and thermal desorption technologies are replaced by sludge washing, soil washing, debris washing, and organic destruction. A set of dominant waste treatment processes and accident scenarios were selected for analysis by means of a screening process. A subset of results (release source terms) from this analysis is presented.

INTRODUCTION

This paper presents a preliminary assessment of potential accidents for the "No-Flame" option leading to airborne releases at U.S. Department of Energy (DOE) sites. The assessment is being developed in support of the Programmatic Environmental Impact Statement (PEIS) for management of low-level mixed waste by the Environmental Management (EM) Office of DOE. An important consideration in the WM PEIS is the risk to human health of potential radiological releases from facility accidents. An evaluation of facility accidents is a necessary first step in evaluating the risk of accidents to the on-site and off-site populations at each of the sites. This risk evaluation is part of the process of comparing alternative management strategies in the WM PEIS. These strategies include decentralization, regionalization, and centralization of waste treatment activities.

Low-level mixed waste contains both radioactive and Resource Conservation and Recovery Act (RCRA)-controlled substances. LLMW is generated, projected to be generated, or stored, at 37 DOE sites as a result of research, development, and production of nuclear weapons. It is projected that waste management activities will require management of an estimated 226,000 m^3 of LLMW over the next 20 years.

A variety of treatment methods and processes for LLMW were considered in the WM PEIS. For difficult-to-treat LLMW containing organic material, two thermal treatment methods were analyzed: incineration, which EPA considers the best demonstrated available technology for organic waste, and thermal desorption, which bakes the waste at temperatures lower than those used in incineration. A "No-Flame" treatment process is being considered that replaces thermal treatment (incineration and thermal desorption) with sludge washing, soil washing, debris washing, and organic destruction technologies.

The safety documentation that exists for the washing and organic destruction technologies were reviewed to establish which technology may significantly contribute to the overall risk of waste treatment. The technologies were also examined to determine if one or more of the following accident conditions could exist:

1. Conditions which could result in large-scale damage or overpressurization of the various pieces of equipment, tanks, or vessels for each technology;

2. Ignition of flammable gases (including liquids and aerosols) that are always present or ignition after release of retained flammable gas/liquid/aerosol;

3. Process equipment failures which could result in an energetic release of radioactive material;

4. Suspension of radioactive materials by sprays, etc.

Based on the above criterion, it was determined that the accident analysis would focus on Organic Destruction (ORD), due to the potential of overpressurization (point 1), combustibility of the input waste stream (point 2), and energetic releases upon reactor rupture (point 3).

The organic destruction technology is similar to wet-air oxidation except that the organic concentration in the waste feed is significantly higher (greater than 50 percent). Organic destruction is the aqueous-phase oxidation of concentrated organic and inorganic wastes in the presence of oxygen at elevated temperature and pressure. Pressure in the range of 300 to 3,000 psi is used to maintain water in its liquid state, which allows oxidation to progress at lower temperatures than would be required for open-flame combustion. Water serves to moderate the oxidation rate by absorbing excess heat of reaction. Reactor temperatures typically range from 350° to 610 °F (LLNL 1994; Musgrave 1995). The layout of an ORD conceptual facility is presented in Figure I, based on (Musgrave 1995)

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OVERVIEW OF FACILITY ACCIDENT ANALYSIS

The source term associated with an accident is the amount of radioactive material that is released to the immediate environment and is the product of four factors that vary for each radionuclide within the inventory affected by the accident:

Source term	=	material-at-risk (MAR)	(1)
	x	damage fraction (DF) or fraction of MAR available for release	
	x	respirable airborne release fraction (RARF)	
	x	leak path factor (LPF)	

The material-at-risk (MAR) is defined as the inventory of waste impacted by an accident. The damage fraction is defined as the volumetric fraction of the MAR actually susceptible to airborne release. The RARF is the fraction of the total available radioactive material that is released and rendered airborne from primary confinement in a readily dispersible form. The LPF accounts for the reduction of the amount of airborne material due to containment, high-efficiency particulate air (HEPA) filtration, deposition, etc.

Determination of the Material-at-Risk (MAR)

The material-at-risk for the conceptual ORD facility is given by the summation of the major equipment and process piping:

$$MAR_{facility} = MAR_{reactor} + MAR_{feed\ mix\ tanks} + MAR_{process\ lines,\ etc.} + MAR_{separator} \quad (2)$$

The various pieces of process equipment and their operating conditions were reviewed to establish which accident conditions would result in the largest airborne release, based on the present state of knowledge concerning the operation of the technology, potential failure modes, and radionuclide quantity present at the presumed time of failure. The material-at-risk associated with the process lines, heat exchangers, and other pieces of process equipment is neglected in this analysis, due to the presumed low volume of material associated with these items. The same argument is applied to the material-at-risk in the separator, due to the low-temperatures and pressures employed in this unit, as well as low volatility of the treated wastes.

The calculation of material-at-risk for the reactor and feed mix tank takes the following form:

$$MAR_i = TR \text{ x } CONC_i \text{ x } \tau \quad (3)$$

where TR is the treatment throughput rate (m^3/yr) of the ORD facility, $CONC_i$ is the concentration of radionuclide "i" in the feed (Ci/m^3), and τ is the space time (residence time) of the reactor and the feed mix tank. The treatment throughput rate and radionuclide concentration of the feed are obtained from the WASTE MGMT computational model (Avci et al. 1994) and are a function of DOE site, treatment technology, and alternative site configuration.

LLNL (1994) indicates that an organic destruction reactor would require a capacity of 540 gallons for a throughput of 5,177 kg feed per week, resulting in a space time of 33 hours ($\tau_{reactor} \sim 33$ hour). The space time for the feed mix tank is estimated based on equipment size data for a similar wet-air oxidation system (Feizollahi and Shropshire 1994) which indicates that comparable volumes of waste are contained within the reactor and feed mix tank. To assure a continuous flow of waste as feed to the reactor, the space time of the feed mix tank must be similar to that of the reactor ($\tau_{feed\ mix\ tanks} \sim 33$ hour) and therefore the material-at-risk for the feed mix tank is equivalent to that postulated for the reactor. The material-at-risk for the ORD facility is given by:

$$MAR_i = 66 \text{ x } TR \text{ x } CONC_i \ / \ 4{,}032 \quad (4)$$

based on 4,032 hours of operation per year.

DEVELOPMENT OF ACCIDENT SEQUENCES

A spectrum of accidents that occur during treatment were developed based on the waste's physical and radiological characteristics in conjunction with the technology specifications. They range from operational events (i.e., an overpressurization in the reactor chamber) to facility fires to external events (i.e., natural phenomenon events and airplane crashes). The accidents considered are discussed below.

<u>**Rupture of a Single ORD Reactor (accident sequence WAX)**</u>

Due to the similarity in processes between organic destruction and wet-air oxidation (WAO), the limited safety literature for the WAO process was reviewed to determine which accident sequences have been postulated to be risk-dominant. The worst-case *internally*-generated accident generally involved the rupture of the WAO reactor resulting from overpressurization and/or equipment failure (Boomer et al. 1991). The ORD reactor operates at a pressure and temperature of approximately 250 psi and 260°C, respectively (LLNL 1994). If the reactor fails, the solution will flash to steam. The steam will be assumed to condense into particles of less than respirable size (10 micron AED) and be transported out of the ORD facility. The release to the atmosphere will be limited as the release is not energetic enough to breach the facility containment.

A review of the literature for tank and/or connecting pipe failures indicate that the failure rates depend primarily on (1) the design standard or basis when considering specific damage mechanisms, and (2) the inherent conservatism involved. As an example, large high-pressure vessels have a lower failure rate than low-pressure storage tanks. Failure rates between 1×10^{-4} to 1.3×10^{-3} / yr have been reported for various pressure vessels with a median value of 1×10^{-3} / yr. In this analysis, it is assumed that a failure rate of 1×10^{-3} / yr applies to breaches of the ORD reactor that could result in significant releases.

The damage fraction for this sequence is based on the contents of a single ORD reactor. The contents of the three ORD reactors constitute 50% of the facility MAR, so that the contents of a single ORD reactor is about 16.7% of the facility MAR (one-third of 50%).

The respirable airborne release fraction (RARF) is the product of the airborne release fraction (ARF) and the respirable fraction (RF). The RARF for free-fall spill of the superheated aqueous solution in the ORD reactor is determined assuming isentropic expansion. The amount of the ORD reactor contents that will flash to steam upon release was determined from the following:

$$[\text{Mole Fraction of Vapor Flashed}] = \{ H_{L1} - H_{L2} \} / \Delta H_{vap} \tag{5}$$

where H_{L1} is the enthalpy of the feed stream (1,265 Btu/lb at 250 psi and 260°C), H_{L2} is the enthalpy of the liquid (water) after release (180 Btu/lb at 1 atm and 100°C) and ΔH_{vap} is the heat of vaporization at the release temperature and pressure (970 Btu/lb at 1 atm and 100°C). The mole fraction of vapor flashed is estimated from the above equation to be approximately 100%. Thus, all of the solution would flash (evaporate) to steam. The release factor from pressurized releases of superheated aqueous liquid solutions is given by (DOE 1993):

$$\text{RARF} = 0.33 \times [\text{Mole Fraction of Vapor Flashed}]^{0.91} \tag{6}$$

Based on a vapor mole fraction of 100%, the RARF is estimated to be 0.33. It should however be noted that this relatively high release fraction would be mitigated by the presence of double banks of HEPA filtration and moisture-condensing systems such as demisters, condensers, etc.

The characteristics of the WAX accident sequence are given in Table I. The value of the RARF shown in the above table only applies to *nonvolatile* particulate solid radionuclides (such as U-235, Pu-238 and other transuranics, etc.); a release fraction of unity is applied to noble gases and halogens.

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Facility Fire (accident sequence WAF)

The ORD facility is designed to process organic and inorganic semi-solid and adsorbed materials and to destroy soluble materials such as heavy organic oils and emulsions (including chlorosolvents) produced during washing of sludges, soil, and debris. The high organic feed streams are diluted to 5 percent organic or less prior to injection into the reaction vessel. A low-order detonation of the organic waste and oxidant has been postulated to occur; however, the reaction chamber would be designed to avoid detonation (LLNL 1994). In this analysis, it is postulated that a fire occurs outside the ORD feed mix tank following leakage. A fire caused by ignition of combustible solvent would disperse radioactive particulates in the immediate area of the fire and would last for a short period because the amount of combustible material is limited. Due to the high structural integrity of the ORD reactor as well as the dilute aqueous nature of its contents, it is assumed that its contents would be unaffected (i.e., not released in significant quantities) by this accident sequence. (However it may be expected that its continued operation would be impaired.) The accident is presumed to be initiated by failure of the feed mix tank resulting in a large pool of organic liquid, fine particulates, etc. on the ground. This leakage from the feed mix tank is ignited by an electrical short, etc. It is conservatively assumed that all of the tank contents are spilled and burn. The release to the atmosphere will be limited due to the fire protection capabilities of the facility and the assumption that the release would not be energetic enough to breach the facility containment.

A wide range of initiating fire frequencies has been reported in recent NEPA literature, ranging from 7×10^{-4} to 2.0×10^{-2} / yr with a median value of 5×10^{-3} / yr. In this analysis, it is assumed that an initiating frequency of 5×10^{-3} / yr applies to a fire in the ORD feed mix tanks that could result in significant releases.

The damage fraction for this sequence is based on the contents of a single ORD feed mix tank. The contents of the ORD feed mix tank constitute 50% of the facility MAR, leading to a damage fraction of 50%.

The input feed to the ORD reactor is stated to be diluted to 5 percent organic or less. The feed to the ORD facility has been categorized in the WM PEIS as an *Organic Combustible Solution* in an *Aqueous Solution*. Because a large percentage of the input liquid is aqueous in nature, the behavior of the feed mix tank contents upon application of a thermal stress was considered to be consistent with that of a boiling *Aqueous Solution* with droplet formation, for a RARF of 1×10^{-2} (DOE 1993). The characteristics of the WAF accident sequence are given in Table II.

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External Events

External challenges to the ORD facility include airplane impacts and natural phenomenon. The representative natural phenomenon analyzed is a seismic event because of its potential to affect the entire facility. A seismic event is postulated to rupture fittings/connections to the ORD reactors, resulting in aerosol formation. It was however not assumed that this would result in a small fire affecting the facility MAR. The contribution associated with the feed mix tank was neglected as a beyond-design basis seismic event could fracture the concrete footings under the holding tanks, allowing any spilled material to be absorbed by the soil, with negligible atmospheric releases. The accident frequency for seismic events is estimated on the performance goal for a Moderate Hazard facility, as defined in DOE guidelines.

Aircraft impacts were also analyzed as potential man-made external events. Aircraft accident frequencies are site dependent and were obtained from aviation statistics and the locations of DOE sites with respect to major airports and aviation routes.

Functional event trees specific to the organic destruction technology were developed to track the progression of the external accident initiators out to the point of airborne release. Initiating accident frequencies and conditional probabilities of the various event tree branches were determined from applicable safety literature where possible. Further information on development of the external event sequences is available in (Mueller et al. 1994). The assumed characteristics of the various external accident sequences are given in Table III.

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RESULTS AND DISCUSSION

The results of the accident analysis were obtained in the form of a detailed source term and an associated estimated annual frequency. The accidents have been grouped into four categories on the basis of their estimated frequency, with the categories ranging from anticipated (frequency higher than 10^{-2} per year) to extremely unlikely (frequency less than 10^{-6} per year) events.

Table IV provides a sample results with detailed information about the risk-dominant accidents summed over all radionuclides released, including the volume of the material-at-risk (VMAR., in m^3), the material-at-risk (MAR, in Ci), total release fraction (TRF), source term (in Ci), accident frequency, and frequency class. The total release fraction is the product of the leak path factor (LPF), damage fraction (DF), and the respirable airborne release fraction (RARF). Only one WM PEIS alternative, number 36, which involves treatment at 12 sites, is shown because of space restrictions.

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The results in the table suggest that, in general, the risk of releases from the ORD facilities due to accidental causes would be low. Preliminary screening estimates confirmed that the risks to human health involved in LLMW management for the "No-Flame" option would be relatively low. Generally, releases of large amounts of radioactivity are associated with a very low estimated frequency, while more frequent events potentially result in small releases. The relatively low health impacts are the result of a number of factors including less severe operating conditions, absence of a fuel source such as natural gas used in incineration, and dilution with water of the product stream from organic destruction. All these factors may be expected to contribute to a lower health impact in comparison with incineration.

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FIGURE I. Equipment Layout for a LLMW Organic Destruction Treatment Facility

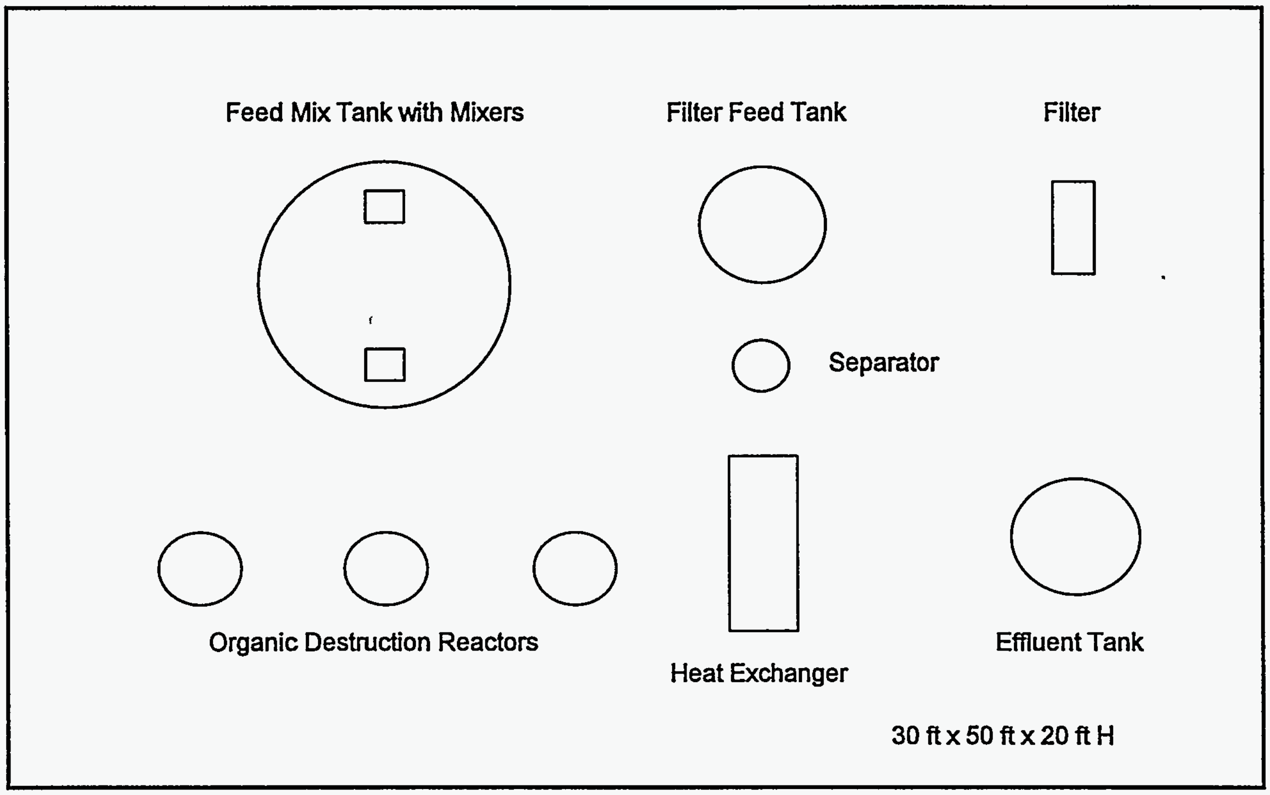

TABLE I. Source Term Parameters and Initiating Frequency for the WAX Accident Sequence

Accident Sequence	Accident Scenario	DF Basis	RARF Basis for Particulate Solids	HEPA Filtration?	Initiating Frequency (1/yr)
WAX	breach of one ORD reactor	1 ORD reactor (16.7% of facility MAR)	overpressurization (RARF=0.33)	unaffected	1E-3

TABLE II. Source Term Parameters and Initiating Frequency for the WAF Accident Sequence

Accident Sequence	Accident Scenario	DF Basis	RARF Basis for Particulate Solids	HEPA Filtration?	Initiating Frequency (1/yr)
WAF	ORD facility fire	1 feed mix tank (50% of facility MAR)	small fire (RARF = 1E-2)	partial loss of filtration	5E-3

TABLE III. Source Term Parameters and Conditional Probabilities for the Airplane Crash and Seismic Events

Accident Initiator	Sequence Number	DF Basis	RARF Basis for Particulate Solids	Filtration Operable?	Conditional Probability
APLL[a]	1	0 (no releases)	none	N/A	0.5
APLL	2	3×10^{-4}	none	yes	0.05
APLL	3	0.5 (ORD reactors)	overpressurization (RARF = 0.33)	none	0.45
		0.5 (ORD feed mix tanks)	small fire		
APLS[b]	1	0 (no releases)	none	N/A	0.9
APLS	2	3×10^{-4}	none	yes	0.09
APLS	3	0.5 (ORD reactors)	overpressurization (RARF = 0.33)	none	0.008
APLS	4	0.5 (ORD reactors)	overpressurization (RARF = 0.33)	none	0.002
		0.025 (ORD reactors)	small fire (RARF = 2×10^{-3})		
EQ	1	0 (no releases)	none	N/A	0.9
EQ	2	3×10^{-4}	none	yes	0.05
EQ	3	0.5 (ORD reactors)	overpressurization (RARF = 0.33)	none	0.04
EQ	4	0.5 (ORD reactors)	overpressurization (RARF = 0.33)	none	0.01
		0.025 (ORD reactors)	small fire (RARF =2×10^{-3})		

(a) refers to large aircraft crash

(b) refers to small aircraft crash

TABLE IV: Frequency and Source Term Parameters for Risk-Dominant Generic LLMW Organic Destruction Facility Accidents

Site	Init	Accident	VMAR (m3)	MAR (Ci)	TRF	Source Term (Ci)	Frequency (1/yr)	Frequency Class
ETEC	EQ3	Earthquake with partially filtered crush	3.1E-03	3.9E-05	1.7E-01	6.8E-06	4.5E-05	III
ETEC	WAF1	Tank fire	3.1E-03	3.9E-05	9.0E-03	3.6E-07	5.0E-03	II
FEMP	EQ3	Earthquake with partially filtered crush	4.4E-02	1.0E-03	1.7E-01	1.8E-04	4.5E-05	III
FEMP	WAF1	Tank fire	4.4E-02	1.0E-03	7.5E-03	7.7E-06	5.0E-03	II
FEMP	WAX1	Tank explosion	4.4E-02	1.0E-03	2.5E-03	2.6E-06	1.0E-03	II
HANF	EQ3	Earthquake with partially filtered crush	1.4E-01	5.4E-01	1.7E-01	9.3E-02	4.5E-05	III
HANF	WAF1	Tank fire	1.4E-01	5.4E-01	1.3E-02	6.8E-03	5.0E-03	II
HANF	WAX1	Tank explosion	1.4E-01	5.4E-01	4.3E-03	2.3E-03	1.0E-03	II
INEL	EQ3	Earthquake with partially filtered crush	7.8E-03	4.7E-02	3.5E-01	1.6E-02	4.5E-05	III
INEL	WAF1	Tank fire	7.8E-03	4.7E-02	2.8E-01	1.3E-02	5.0E-03	II
INEL	WAX1	Tank explosion	7.8E-03	4.7E-02	9.3E-02	4.3E-03	1.0E-03	II
LANL	EQ3	Earthquake with partially filtered crush	6.0E-03	2.1E-02	4.9E-01	1.0E-02	4.5E-05	III
LANL	WAF1	Tank fire	6.0E-03	2.1E-02	4.9E-01	1.0E-02	5.0E-03	II
LANL	WAX1	Tank explosion	6.0E-03	2.1E-02	1.6E-01	3.4E-03	1.0E-03	II
LLNL	EQ3	Earthquake with partially filtered crush	5.2E-02	2.6E+00	5.0E-01	1.3E+00	4.5E-05	III
LLNL	WAF1	Tank fire	5.2E-02	2.6E+00	5.0E-01	1.3E+00	5.0E-03	II
LLNL	WAX1	Tank explosion	5.2E-02	2.6E+00	1.7E-01	4.4E-01	1.0E-03	II
ORNL	EQ3	Earthquake with partially filtered crush	8.8E-01	1.4E-01	1.8E-01	2.4E-02	4.5E-05	III
ORNL	WAF1	Tank fire	8.8E-01	1.4E-01	1.5E-02	2.1E-03	5.0E-03	II
ORNL	WAX1	Tank explosion	8.8E-01	1.4E-01	5.0E-03	7.0E-04	1.0E-03	II
PADUCAH	EQ3	Earthquake with partially filtered crush	1.8E-02	5.1E-03	1.7E-01	8.5E-04	4.5E-05	III
PADUCAH	WAF1	Tank fire	1.8E-02	5.1E-03	2.4E-03	1.2E-05	5.0E-03	II
PADUCAH	WAX1	Tank explosion	1.8E-02	5.1E-03	8.0E-04	4.1E-06	1.0E-03	II
PANT	APLL3	Large aircraft impact	3.5E-02	2.3E-02	9.7E-01	2.2E-02	1.6E-07	IV
PANT	EQ3	Earthquake with partially filtered crush	3.5E-02	2.3E-02	4.9E-01	1.1E-02	4.5E-05	III
PANT	WAF1	Tank fire	3.5E-02	2.3E-02	4.8E-01	1.1E-02	5.0E-03	II
PANT	WAX1	Tank explosion	3.5E-02	2.3E-02	1.6E-01	3.7E-03	1.0E-03	II
PORTS	EQ3	Earthquake with partially filtered crush	1.9E-01	1.1E-02	3.2E-01	3.4E-03	4.5E-05	III
PORTS	WAF1	Tank fire	1.9E-01	1.1E-02	2.3E-01	2.4E-03	5.0E-03	II
PORTS	WAX1	Tank explosion	1.9E-01	1.1E-02	7.7E-02	8.2E-04	1.0E-03	II
SRS	EQ3	Earthquake with partially filtered crush	3.0E-01	9.5E-01	4.5E-01	4.3E-01	4.5E-05	III
SRS	WAF1	Tank fire	3.0E-01	9.5E-01	4.3E-01	4.0E-01	5.0E-03	II
SRS	WAX1	Tank explosion	3.0E-01	9.5E-01	1.4E-01	1.4E-01	1.0E-03	II