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Maximum Hypothetical Accident Analysis for the Natura Resources Molten Salt Research Reactor at Abilene Christian University

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Maximum Hypothetical Accident Analysis for the Natura Resources Molten Salt Research Reactor at Abilene Christian University

by

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Report

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Dedication

To my parents and brother. Altyd ‘n ander ding!

Acknowledgements

I would like to start with thanks to my supervisor and reviewers – Derek, for giving me the opportunity to work on this project and being willing to take a chance on me, and Tracy, for being a font of knowledge and ever patient with a plethora of questions.

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Abstract

Maximum Hypothetical Accident Analysis for the Natura Resources Molten Salt Research Reactor at Abilene Christian University

by

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This work proposes a mechanism for and the consequences of a Maximum Hypothetical Accident at the Natura Resources Molten Salt Research Reactor on the campus of Abilene Christian University. This accident should be constructed as to have consequences that envelop all credible accident consequences. For this work, which is not exactly representative of evaluations performed for licensing, it is proposed to be a full-inventory fuel salt spill due to a catastrophic pipe failure coupled with a simultaneous loss of site power. Nuclide liberation from the salt was predicted from documentation from Oak Ridge National Laboratory's Molten Salt Reactor Experiment and MATLAB, atmospheric transport was evaluated utilizing the methods presented in Regulatory Guide 1.145, which was coupled with dose coefficients from Federal Guidance Reports 11 and 12 and the ICRP dose compendium. After 60 days with no intervention and highly conservative conditions, the dose consequence to individuals directly on the property downwind of the building for the full duration of the accident was 0.8024 mSv, and 0.5680 mSv for those at the property line. These doses are well below the yearly allowable dose to members of the public, and demonstrate that the MSRR operations, including any credible accidents, do not endanger the health and safety of the public or environment.

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Chapter 1: Introduction to Molten Salt Reactors and Regulatory Bases

Molten salt reactors differ from conventional reactors in that their fuel is liquid during normal operations and tend to operate at higher temperatures without the direct use of water as a coolant, instead relying on molten salts such as FLiBe or FLiNaK. This confers several advantages to prospective reactor owners/operators, including the capability to remove neutron poisons, long-lived wastes, or commercially/medically valuable isotopes while the reactor is operating; burn other reactor's spent fuel, and lowered risks of high-pressure steam explosions.

One of the first molten salt reactors was known as the Molten Salt Reactor Experiment (MSRE), a 7.4 MW_{th} reactor using FLiBe salt which was operated by Oak Ridge National Laboratory (ORNL). Natura Resources is currently working to design and construct the Molten Salt Research Reactor (MSRR), a 1 MW_{th} reactor also using FLiBe salt which is to be situated at Abilene Christian University's (ACU) Science Engineering Research Center (SERC). The licensing effort is led by ACU and supported by The University of Texas at Austin, Texas A&M University, the Georgia Institute of Technology, and Natura Resources.

Under the United States Nuclear Regulatory Commission's (NRC) licensing standards, codified in 10 CFR 50 and recently guided by NUREG 1537 for advanced research reactors [1], the defining accident for a research reactor's licensing is known as Maximum Hypothetical Accident (MHA). The MHA differs from the power reactor's design basis event (DBE) in that it is a non-credible, non-mechanistic accident whose consequences bound those of any credible accidents that can occur at the facility. Thus, the analysis of an MHA should include several non-credible conservative assumptions that ensure that realistic doses from any accident would be below those proposed by the MHA analysis. This report seeks to document a viable MHA for the MSRR and analyze the consequences of the MHA to bound any realistic accidents that could occur.

Chapter 2: MSRR Layout and Accident Proposal

The MSRR resembles a pressurized water reactor in that it consists of two loops: a reactor loop which features the core and sits within a stainless-steel enclosure, and a coolant loop which also contains molten salt that serves as the coolant and rejects heat to the environment. This enclosure sits within a covered trench, known as the cell. The salt, reactor, enclosure, and cell represent four layers of barriers intended to retain radionuclides during normal operations.

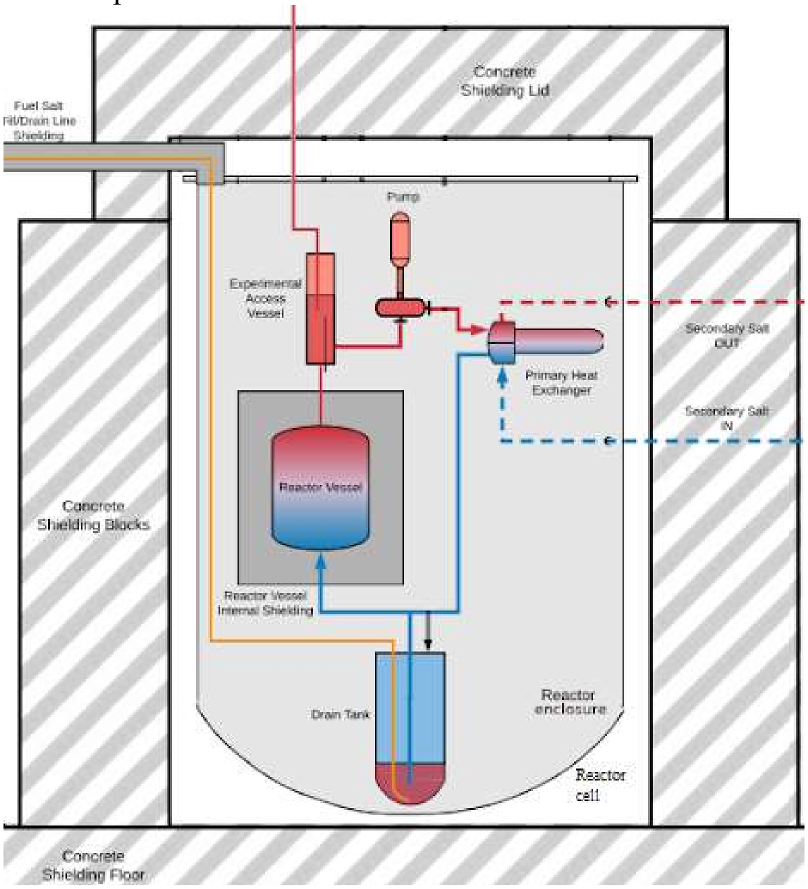

Figure 1: MSRR High-Level Cutaway Showing Cell and Enclosure Boundaries

During the 2019 ORNL “Molten Salt Reactor Initiating Event and Licensing Basis Workshop,” [2] it was proposed for a power-producing molten salt reactor that a breach in the fuel salt primary boundary could be the Design Basis Event (DBE). The DBE, which is the highest-consequence plausible event for a power reactor, is not directly analogous to the MHA due to differing use cases and implementations but can serve as a useful reference when establishing what kinds of events may constitute an MHA for research and test molten salt reactors.

Similarly, the MSRE safety analysis, which was performed under the Atomic Energy Commission's oversight rather than the modern NRC's oversight, identified a "Maximum Credible Accident" that would pose the maximum dose to the public and maximum threat to barrier integrity and designed their barriers to mitigate such a scenario [3]. The MSRE Maximum Credible Accident is initiated by a catastrophic failure resulting in the release of the entire fuel salt inventory from the primary loop, coupled with a secondary failure that results in mixing water and hot fuel salt, resulting in steam generation causing a pressure spike and subsequent fission product dispersion.

Thus, based on the ORNL workshop and the MSRE's safety analysis, the following sequence of events is proposed to be the MHA for the MSRR: A catastrophic failure of the reactor piping results in the relocation of all fuel salt into the bottom of the reactor enclosure and immediately liberates all nuclides considered capable of release as a gas. Simultaneously, the SERC loses offsite power, disabling the reactor's cooling system as well as preventing airflow through the stack which could dilute the atmospheric concentration of radionuclides. Lastly, the reactor bay is rendered inoperable and open to the environment such that any nuclides that enter the bay are emitted directly into the environment. This combination of events, when coupled with the other conservative assumptions discussed in Chapter 4, results in an accident whose consequences will bound those of any accident that can credibly occur.

Chapter 3: Selection of Nuclides at Risk

While the fundamental radionuclides present in a nuclear reactor are mostly invariant of the type of reactor, the radionuclides that can potentially disperse and the fractions of dispersal for such nuclides vary significantly with fuel type. As opposed to traditional research reactors where a meltdown results in a potential dispersal of radionuclides, the MSRR's fuel is always molten during operation. The nuclides considered in the MSRR MHA are thus based on empirical data from the MSRE, which categorized radionuclides into three categories: salt-seekers, noble metals, and noble gases [4]. The salt-seekers, like cesium, rubidium, and strontium, easily dissolve into the salt and remain stable within it, and thus tend to remain co-located with the salt. Noble metals such as molybdenum and ruthenium do not easily dissolve in the salt and are salt-phobic, and thus tend to accumulate at the interfaces with salt and other materials, forming much of the crud that was found in the MSRE piping as well as aggregating into small, entrained particulates [5]. Lastly, the noble gases, such as xenon and krypton also do not readily dissolve into the salt and instead either escape or are entrained as circulating voids and bubbles. Of these three categories, only the noble gases are expected to escape during the MHA and will be assumed to be released in full inventory quantities.

Some nuclides defy such simple categorizations. Tritium is not truly a noble gas, but behaves most similarly to one and is generated in significant quantities due to the $^{6}\text{Li}(n,\alpha)^{3}\text{H}$ reaction in the fuel and coolant salts. All tritium generated in the reactor during the lead-up to the accident is assumed to be retained until the accident initiates, at which point the tritium is released. Tellurium and iodine are also given unique categories in ORNL's analysis: tellurium, which is nominally a noble metal, has an appreciable vapor pressure of 13 torr at the 650C operating temperature of the MSRE, and there was some uncertainty among the MSRE staff as to where the tellurium was migrating [4]. In turn, this unique characteristic affects the distribution of tellurium's daughter product iodine, which is nominally expected to be a salt seeker. This salt-seeking behavior of iodine is supported by the fact that iodine isotopes with short-lived tellurium parents migrate with the salt in near-inventory quantities while isotopes with longer-lived tellurium parents behaved more unpredictably [5]. However, due to the tellurium parent's transport behavior influencing iodine distribution in the MSRE, the ORNL report did not classify iodine as a salt seeker. Thus, this analysis conservatively assumes that full-inventory quantities of iodine and tellurium are released upon accident initiation. Additionally, bromine is also assumed to be released due to its chemical similarity to iodine and its importance in cesium production. Table 1, below, summarizes the nuclides considered for release and their activities upon accident initiation. Nuclides with an initial release of 0 Bq build-in as a result of other nuclides decaying.

It should be noted that not every mass chain was utilized for this analysis: Of the Te-I-Xe-Cs mass chains, 131, 132, 133, 134, 135, and 137 were utilized, which comprise 75.77% of the activity at t = 0 but 99.99% of the activity within 12 hours of accident initiation. Likewise, for the Br-Kr-Rb-Sr mass chains, 85, 88, 89, 90, and 91 were selected. These comprise 67.39% of the activity at t = 0 and 94.74% of the activity after 12 hours.

Table 1: Initial Nuclide Inventories Assumed for the MHA

Nuclide	Initial quantity released to enclosure (Bq)
H-3	3.92E+13
Te-131	1.59E+15
Te-131m	2.62E+14
I-131	1.77E+15
Xe-131m	1.93E+13
Te-132	2.64E+15
I-132	2.66E+15
I-132m	7.44E+12

Te-133	2.06E+15
Te-133m	2.30E+15
I-133	4.10E+15
I-133m	2.91E+14
Xe-133	3.98E+15
Xe-133m	1.18E+14
Te-134	4.24E+15
I-134	4.78E+15
I-134m	2.30E+14
Te-135	2.03E+15
I-135	3.86E+15
Xe-135	3.83E+15
Xe-135m	7.34E+14
Cs-135	0
Te-137	2.50E+14
I-137	1.87E+15
Xe-137	3.75E+15
Cs-137	0
Br-85	7.83E+14
Kr-85	4.70E+13
Kr-85m	7.86E+14
Br-88	1.05E+15
Kr-88	2.13E+15
Rb-88	0
Br-89	6.57E+14
Kr-89	2.71E+15
Rb-89	0
Sr-89	0
Br-90	3.36E+14
Kr-90	2.81E+15
Rb-90	0
Sr-90	0
Y-90	0
Br-91	1.35E+14
Kr-91	2.01E+15
Rb-91	0
Sr-91	0
Y-91	0
Y-91m	0

Chapter 4: Accident Progression Modeling

Based on the above description of accident progression, the MHA was modeled utilizing a three-compartment system comprising of the enclosure, the reactor cell, and the environment, with no credit claimed for the reactor bay building. Nuclides flow from the enclosure, into the cell, and out into the environment with no deposition. It should be noted that based on RELAP5 analysis performed by Dr. Jonathan Scherr [6] on the pressure rise in the enclosure as a result of both the sudden depressurization of the reactor atmosphere into the enclosure and decay heat, the enclosure remains at negative pressure relative to the cell's atmospheric ambient pressure throughout the duration of the accident. Radionuclide movement from the enclosure to the cell is thus driven by partial pressure-driven diffusion against the total pressure gradient. It is assumed that the noble gases, which do not chemically interact with the metal, will diffuse at a rate ten times that of the iodines and solid particulates. Although this proposed magnitude has relatively high uncertainties from a lack of data, evidence exists from ORNL solid fuel failure experiments analyzing the release of cesium and iodine against that of the noble gases in drilled or ruptured fuel elements. In the drilled fuel experiments where diffusion is the dominant transport mechanism, it was demonstrated that noble gases are readily released in inventory quantities while iodine and cesium both showed a strong dependence on time permitted for diffusion through the element [7, Tab. 40] as well as temperature [7, Fig. 36]. Additionally, attempts to fit other experimental data demonstrate that iodine's diffusion rate constant is far more sensitive to defect size than that of the noble gases, with smaller defects resulting in iodine rate constants 20 times smaller than that of noble gases [8]. This difference in diffusion rate is only applied to the enclosure-to-cell pathway, as nuclide migration from the cell to the environment is driven by whole-air movement, and thus all nuclides move at the same rate. Table 2 describes all assumptions utilized for evaluating nuclide transport.

Table 2: Assumptions For Nuclide Transport

Assumption	Values/Notes
Enclosure-to-cell leak rate	0.01% per day for noble gases, 0.001% per day for all others
Cell-to-environment leak rate	1% per day for all radionuclides
Daughter product behavior	All nuclides involved in the accident, either directly or as daughters, do not resuspend into the salt, plate out onto surfaces, or otherwise get removed from the material at risk
Chemical forms for internal dose evaluation	For particulates (Te, Cs, Ba, Br, Rb, Sr, and Y): 1um Activity Mean Aerodynamic Diameter (AMAD) particulates with the most conservative clearance class (D, W, or Y) given in FGR 12 for that particular isotope; for iodine, I_2 vapor; for noble gases, gaseous diatomic noble gases; for H-3, HTO vapor
Plume modeling weather conditions	Constant “calm” conditions (1 mph winds, in the same direction, Pasquill stability class F), highly unlikely but conservative.

Immediately upon accident initiation, the full inventory quantities of nuclides considered at risk for escape (H, Te, I, Xe, Br, and Kr) migrate to the enclosure environment.

$$\frac{dN}{dt} = -N(L+\lambda) + \sum_{i=1}^{i}(Br_i * N_i * \lambda_i) + (N_{source}L_{source}) \qquad (1)$$

Where

- N is the number of atoms of the isotope of interest in the current compartment (atoms)
- L is the leakage rate from the current compartment to the next for the isotope of interest (sec^{-1})
- λ is the decay constant for the nuclide of interest (sec^{-1})
- Br_i is the branching ratio to the nuclide of interest from parent *i*
- N_i is the number of atoms of parent isotope *i* in the current compartment (atoms)
- λ_i is the decay constant for the parent nuclide being analyzed (sec^{-1})
- N_{source} is the number of atoms in the upstream compartment (atoms)
- L_{source} is the leak rate of the upstream compartment (sec^{-1})

And are bounded by the following initial conditions:

- 100% of fuel salt Te, I, Xe, Br, Kr, and H are liberated at T_0 into the enclosure
- $N_0 = 0$ at T_0 for all other compartments or isotopes

DILUTION FACTOR CALCULATION

Dilution factors, also referred to as relative source strengths, were then calculated for two significant distances from the reactor; at the property boundary 100m away and in the direct wake of the building. For the property boundary 100 meters away, the methodology from Regulatory Guide 1.145 [9] was utilized. From this regulatory guide, the relative source strength takes on one of the following forms:

$$\frac{\chi}{\dot{Q}} = \frac{1}{\overline{U}_{10}\left(\pi\sigma_y\sigma_z + \frac{A}{2}\right)} \tag{2}$$

$$\frac{\chi}{\dot{Q}} = \frac{1}{\overline{U}_{10}\left(3\pi\sigma_y\sigma_z\right)} \tag{3}$$

$$\frac{\chi}{\dot{Q}} = \frac{1}{\overline{U}_{10}\pi\Sigma_y} \tag{4}$$

Where

$\frac{\chi}{\dot{Q}}$ is the concentration of a nuclide ($X\frac{Bq}{m^3}$) normalized to the source strength ($\dot{Q}\frac{Bq}{s}$),

$\overline{U}_{10}$ is the windspeed at 10m above ground-level

σ_y is the lateral plume spread as a function of atmospheric stability and distance (f, 100m)

σ_z is the vertical plume spread as a function of atmospheric stability and distance (f, 100m)

$\Sigma_{zj}(X)$ is the vertical plume spread with a volumetric correction for a release within a building wake cavity.

A is the smallest vertical-plane cross sectional area of the SERC (140 m^2)

π is 3.14159

Given the conservative meteorological conditions associated with our analysis (constant direction, 1 m/s windspeed, class F stability conditions), these equations yield values of 98.90, 86.71, and 115.61 for equations (2), (3), and (4) respectively. As instructed in the Regulatory Guide, (2) and (3) were compared, and the higher of the two values (the value from (2)) is taken. Then, that value was compared against (4), and the lower of those two values (again, (2)) was selected. Thus, a relative concentration of 98.90 sec/m^3 was selected for the 100-meter dilution factor.

Conversely, in the immediate vicinity of the building, the relative concentration was identified utilizing the methodology presented in Lamarsh and Baratta [10] for evaluating concentrations in the direct wake of a building leaking from a vent, which takes the following form:

$$\frac{X}{\dot{Q}} = D_b \; where \; D_b = cA\bar{v} \qquad (5)$$

Where

c is a constant which depends on building geometry, usually conservatively taken at 0.5

A is the building cross-section area, which for the SERC is 140 m^2, and

$\bar{v}$ is the average wind speed over the duration of the release.

For 1 m/s windspeed, this means that our dilution factor is 70 sec/m^3 in the direct building wake.

DOSE RATE MODELING

Dose rates were then calculated from these concentrations, using a combination of Federal Guidance Report (FGR) 11 and 12 dose conversion factors and the International Commission on Radiological Protection (ICRP) Report 23 reference man's breathing rates, along with a more conservative dose conversion factor for iodine vapor taken from the ICRP dose compendium.

Submersion dose rates were calculated from FGR 12 [11], with the dose conversion factors converted from their units of $\frac{Sv}{hr} * \frac{1}{Ci/m^3}$ by multiplying by a factor of 1.332E+19, comprising of one factor of 3600 for seconds to hours, one factor of 100,000 to convert sieverts to millirem, and one factor of 3.7E10 to convert becquerels to curies. Note that all values reported here are converted back to SI units from the United States Customary units.

Inhalation dose rates for the inhalatory hazard nuclides were calculated using the dose conversion factors in the ICRP dose compendium [12] (for iodine) and FGR 11 [13] (for all others) and converted from their given units of $\frac{Sv}{Bq}$ to $\frac{mrem}{Ci}$ by multiplying them by 3.7E15. Due to there being no most probable start time, instead of modeling three eight-hour periods per day, an average air volume inhaled per hour was found by averaging the ICRP 23 [14] standard reference man's daily air intake of 23,000 liters/day and converting it into 0.958 $\frac{m^3}{hr}$ by dividing by a factor of 24,000. This factor is made up of one factor of 24 to convert days to hours, and one factor of 1,000 to convert liters to cubic meters.

External dose rates were calculated by the following:

$$\dot{D} = \mathrm{X} * DCF_{Submersion} \qquad (6)$$

Where X is the airborne activity concentration and DCF is the dose conversion factor. Internal dose rates, in turn, were evaluated using the following:

$$\dot{D} = \mathrm{X} * B_{avg} * DCF_{inhalation} \qquad (7)$$

Where

- $\dot{D}$ is the committed effective dose equivalent "rate"
- B_{avg} is the average breathing rate of the ICRP 23 reference man over 24 hours
- $DCF_{inhalation}$ is the inhalatory dose conversion factor from the ICRP dose compendium (for Iodine) or FGR 11

The internal committed effective dose rates and the external dose rates were then summed together to produce a singular combined total effective dose equivalent (TEDE) dose rate.

Chapter 5: Dose Consequence Analysis

Combined TEDE rates are plotted in Figure 2

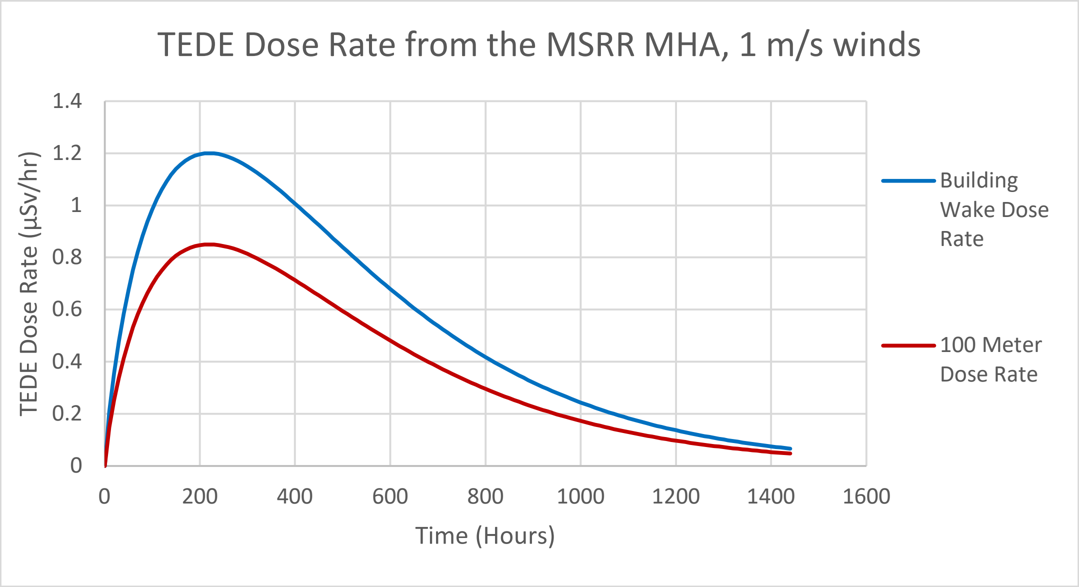

Figure 2: Total Effective Dose Equivalent Rates as a function of time for each distance from the reactor

On a nuclide-by-nuclide basis, 84.49-99.48% of the dose can be attributed to I-131, I-133, and Te-132. I-131 dominates for the entire accident duration, comprising 48.57% of the total dose at the first timestep and 99.46% at the final timestep. I-133 is a significant contributor in the beginning of the accident, providing 16.14% of the dose at the first timestep but is mostly absent after 100 hours, while Te-132 starts at 19% of the total dose and continues to contribute at least 5% to the dose for about 400 hours. Figure 3, below, details the evolution of nuclides most responsible for contributing to the dose over time.

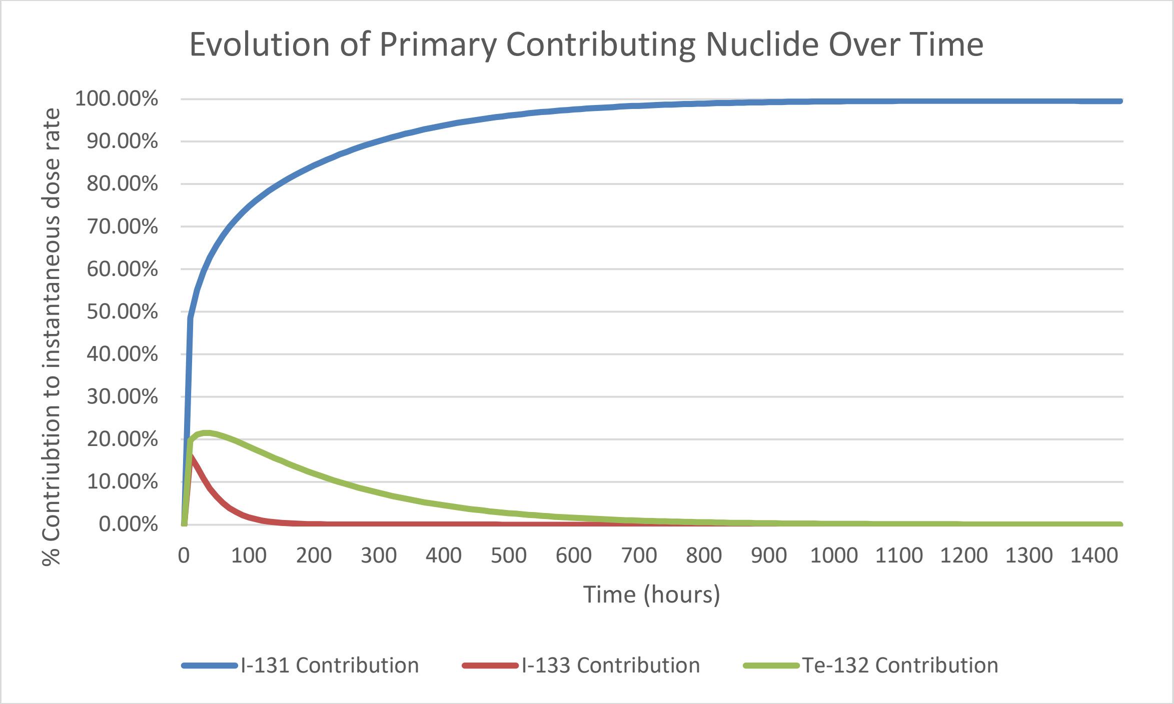

Figure 3: Contribution fraction for I-131, Te-132, and I-133 as a function of time

By generating a 6th-order polynomial best fit for the dose rate curves, integrated doses can be evaluated by directly integrating these lines of best fit from t = 0 hours to t = 1440 hours. This results in a cumulative dose of 0.8024 mSv in the direct lee of the building, and 0.5680 mSv at the property line 100m away. These numbers provide the upper bound for doses, as the actual wind conditions and behavioral assumptions will likely result in lower releases and activity concentrations, bounding an actual accident scenario.

Conclusion

In terms of regulatory significance, it should be noted that there is no formally defined limit for doses due to an MHA. However, the acute dose consequences of the MSRR MHA do not exceed the 1 mSv (100 mrem) yearly dose limit to members of the public during normal operations from 10 CFR 20.1301 [15], nor does it exceed the 0.02 mSv (2 mrem) over any one hour limit from the very same section. These two items in conjunction mean that the MSRR, even under severe accident conditions with all the conservatisms posed here, poses a minimal risk to members of the public. Further research that would support a reduction in source term, particularly for iodine, would have a dramatic impact on lowering of doses resultant from this postulated accident scenario.

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