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Experiments and Modeling of High Altitude Chemical Agent Release

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Experiments and Modeling of High Altitude Chemical Agent Release

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Abstract

The evolution of persistent chemical agent as it undergoes high altitude transport is a complex process. Relatively slow deceleration times for liquid masses, as it reenters in an increasing atmospheric density, are significant for assessing post-impact ground effects. This study utilizes a transient supersonic wind tunnel that can be operated in both rarefied and atmospheric pressure regimes. We have, experimentally, explored liquid drop sizes up to one centimeter in diameter and Weber numbers ranging from a few to a thousand. Upper atmospheric transport analysis and modeling incorporate the knowledge gained from experiments to form a consistent picture of agent breakup for different release altitudes. These experiments and analysis address the hydrodynamic breakup and upper atmospheric reentry effects that are relevant for describing agent deposition on the ground.

Problem Definition

The problem of lethal ground effects centers around the fallout of chemical agent, resulting from either a successful ballistic missile intercept or the proper functioning of an incoming chemical warhead. The lethal effect of agent fallout is influenced by two factors, the first is the individual sizes of agent masses which are correlated with lethal dose levels, the second is the areal density of ground deposition produced by fallout. The physical processes that dispersed agent experiences after release are complex, often interconnected, and wrought with attendant uncertainties. The presence of uncertainty in complex problems, which obviates deterministic approaches, requires one to consider the concept of risk and risk assessment. Implementing a good methodical approach to measure risk aids a whole range of decision making issues including avoidance of heavy ground casualties, optimizing defensive deployment and being adequately prepared for mitigation or remedial actions following a hazardous release of agent.

We focus on exo-atmospheric releases of persistent chemical agent occurring at altitudes well in excess of 30 km. The overall process is illustrated in Figure 1 and we will draw attention to the salient features of the exo-atmospheric release problem.

Inertial Breakup

The inertial breakup process is driven by the fast expansion of energized debris resulting from the hypervelocity impact of a missile interceptor with a warhead carrying chemical agent. This process is characterized by fragmentation or splitting of the liquid agent into smaller masses driven by internal velocity gradients within the liquid. The absence of significant air density for intercept altitudes in excess of 60 km inhibits any prompt aerodynamic driven breakup effects. Expansion velocities for the liquid can range from 200 m/s up to 1 km/sec [1] depending upon projectile velocity, type of vessel, and impact location. For low vapor pressure agents, the inertial breakup will produce an initial ensemble of agent masses which then subsequently fall under the pull of gravity and are free to undergo aerodynamic breakup at lower altitudes.

Outgassing of Volatiles

The presence of high vapor pressure impurities or dissolved gases in persistent agent may result in a type of boiling driven breakup of agent as the impurities come out of solution. The presence of this type of phenomena depends highly on the concentration and properties of the impurities which can lead to nucleation and growth of vapor pockets within an agent mass. The propagation speed, or growth rate, of the nucleation is also important because it can influence the rate of liquid expansion which in turns affects the fragmentation process. This mechanism is highly dependent upon the type of agent considered, its production process, and the type and fraction of impurities present during weaponization.

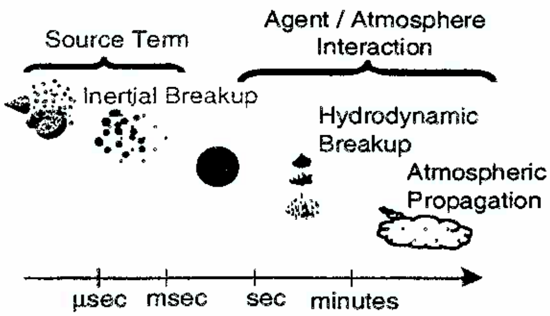


Figure 1. Significant processes that determine agent fallout

It should be noted that both inertial driven breakup and breakup produced by outgassing result only in an initial size distribution of agent masses. The aerodynamic breakup *alone* dominates the production of a final size distribution of agent masses for persistent chemical agent.

Aerodynamic Breakup and Deceleration

The cloud of descending agent masses produced by inertial breakup, and possibly outgassing, reenters through the stratosphere and encounters an ever increasing air density. The variation of increasing air density occurs over altitude ranges measured in tens of kilometers. This characteristic feature allows the reentering agent masses to gradually decelerate in the upper rarefied regions of the atmosphere above 40 km. The relatively "slow" deceleration of the agent masses allows larger masses of agent, with scale sizes measured in millimeters, to remain stable against aerodynamic pressure forces. Agent masses with sizes larger than the aerodynamically stable threshold breakup into smaller, but still significant, sized daughter masses. The defining characteristic of agent reentering at high altitudes is the "slow" deceleration of agent at the high altitudes.

The breakup process can be described as the competition between the aerodynamic pressure and shear forces, which distort and disrupt liquid masses, and the surface tension and viscous forces of the agent which resist the aerodynamic forces. Typically, liquid breakup behavior is characterized empirically with dimensionless parameters such as the Weber number and Ohnesorge number which supposedly correlate these effects to liquid breakup phenomena. The characteristics of liquid breakup occurring in rarefied, supersonic environments represents a previously untouched area of research in dispersed flows. Recent LLNL/UCSB research, detailed in a later section, has contributed new knowledge to this area and generated findings that significantly affect the fallout problem for exo-atmospheric intercept.

The aerodynamic breakup process results in a stable distribution of agent sizes which has decelerated to terminal velocity. The factors that influence the spreading of the stable size distribution include aerodynamic deceleration and atmospheric dispersion processes. Different sized agent masses have different settling velocities and momentum which results in a spatial and temporal spreading of the cloud into a plume shape. The plume descends into the troposphere where atmospheric effects begin to become significant.

Atmospheric Dispersion and Ground Deposition

The plume of now stable agent masses enters into the troposphere where the windfield and turbulent diffusion processes control the spreading of the agent. At this stage, ground deposition concentrations of agent are wholly dependent on the local meteorological conditions present after release. The differing range of agent mass sizes result in further spreading of the plume as millimeter sized masses of agent fallout on time scales of tens of minutes while sub-millimeter drops of agent move with the windfields for hours until ground impaction occurs. Generally, for the agent masses in the lethal millimeter size range, the fallout occurs rapidly giving these pieces of agent a higher areal concentration. For the smaller agent masses the areal concentration is more dependent upon the three-dimensional structure of the flow fields. Deposited agent can produce immediate casualties through inertial impaction or result in the denial of a defended area thereby disrupting useful activity.

The process of agent fallout is clearly very complex in nature. Each aspect contributing to the final outcome can be studied in exhausting detail, requiring a substantial research effort, unless effective shortcuts can be found to narrow the scope of investigation. As described in next section, the exo-atmospheric focus of this research brings about a new, unique demand for basic physical understanding, of the different problem components, to properly assess the level of risk associated with agent fallout. We believe that the development of this physics understanding can be of great value to other problems with similar components such as endo-atmospheric releases. Furthermore, we argue for an approach that examines the conditions that produce defined levels of unacceptable risk and concurrently focuses experimentation at only the necessary pieces of physics needed to understand the agent fallout problem. The additional benefit of this approach is that once it is developed and demonstrated for the exo-atmospheric release problem, it can also be applied to the endo-atmospheric intercept or offensive release scenarios.

Premise of the High Altitude Research

The LLNL/UCSB research effort addressing agent fallout began with the suspicion that no single process in the fallout sequence described previously could render the risk of exo-atmospheric release to be negligible. Available knowledge, at the time, for each one of the components in the fallout sequence was not sufficient to form predictions of the cumulative ground effect. The key elements of reasoning that led us are as follows:

- In order to negate the lethal fallout effects through atmospheric dilution, the agent needs to be broken up into masses with sizes on the order of tens of microns. This outcome can not be guaranteed by any practical means.

- The subject of inertial breakup involves variabilities in the inertial impact of the projectile, mode of structural failure of the warhead, and shock response of the liquid during the release process. Qualitatively, however, we can understand that significant inertial breakup can only occur from differential liquid velocities, and we expect this to be hampered by the absence of a guaranteed mechanism to induce the sharp velocity gradients needed to break the liquid into inconsequential sizes. The physical properties of the agent, surface tension and viscosity, also resist the inertial fragmentation process. There are important scale effects that need to be addressed and these compromise the direct value of terrestial experiments due to limitations of length scales and short experimental observation times.

- The subject of breakup through outgassing is untouched. Volatile constituents are expected to be small, nucleation density highly unpredictable, and growth of the gaseous component controlled by the very "slow" process of liquid-phase diffusion. Furthermore, the resultant liquid configuration produced by outgassing effects would be subject to re-coalescence through the restoring action of surface tension and viscoelasticity.

- Aerodynamic breakup has been studied for many years [2], but the capability remains at the empirical level. Nothing is known about the highly rarefied, supersonic conditions of interest. We expect that these conditions will break similarity to a degree that renders previous empiricism useless. Moreover, previous experience is limited to the breakup of small drops, nothing is known about collective particle effects or successive breakup events and interactions with the atmosphere. Nothing is known about viscoelastic (thickened agent) behavior.

An initial demonstration case was carried out to see whether a significant risk existed for the exo-atmospheric release of agent. This initial scoping of the problem was carried out as follows:

- Experimentally examine the stability of millimeter scale drops in highly rarefied (1e-4 bar), highly supersonic (Mach number M=3) air flows.

- Based upon these stability and drag measurements, determine the fate and dispersion of approximately 200 kg of agent distributed in the mm size range when released in conditions consistent with a 100 km intercept of a chemical warhead.

- Based upon the results of the previous two steps, determine the spreading of agent through atmospheric dispersion processes and track ground deposition over a specific geographical location with real weather conditions.

The results of this hypothetical demonstration illustrate that there is a serious problem in terms of lethal ground effects. Key aspects of the demonstration are summarized below.

Determination of Stable Drop Sizes

The droplet stability experiments were carried out on a transient, supersonic wind tunnel (ALPHA) constructed especially for this purpose. The ALPHA facility is illustrated in Figure 2 and the actual arrangement is shown in Figure 3. ALPHA was designed to test the stability of large liquid masses over long observation time scales (in the hundreds of milliseconds). The flexible design of the ALPHA facility also permitted the generation of "tailored" flow conditions that could be used to explore aerodynamic history effects. ALPHA is capable of reaching operating pressures ranging from 3 bar down to 5.e-4 bar, and with the acquisition of a better vacuum pump we believe that pressures of 1.e-5 bar can be reached. Exchanging flow nozzles will allow ALPHA to achieve flow Mach numbers ranging from $1 < M < 5$ which more than cover the range of supersonic flow conditions of interest. Significant aerodynamic effects experienced by agent begin at approximately the 70 km (7.e-5 bar pressure) level. The ambient air density gradually increases below this level, increasing the amount of aerodynamic drag experienced by the falling agent mass. As shown in Figure 3, the combination of operating pressures and supersonic velocities that ALPHA can achieve more than cover the relevant range of exo and endo-atmospheric conditions needed. Figure 3 contrasts the native capability of ALPHA compared to shock tubes, which are widely used in diagnosing liquid breakup. The difference in capability is obvious. No other liquid breakup facility of this type exists anywhere in the research and development community.

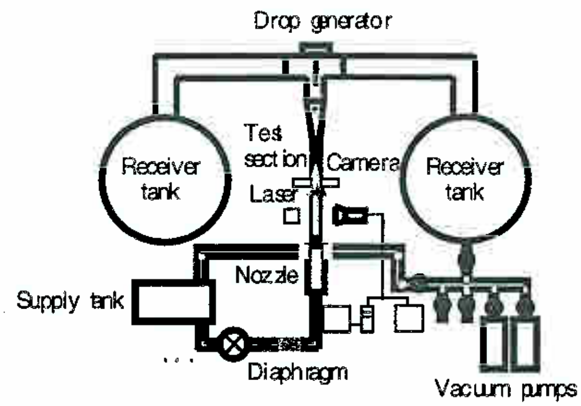


Figure 2. Schematic of the transient, supersonic wind tunnel ALPHA

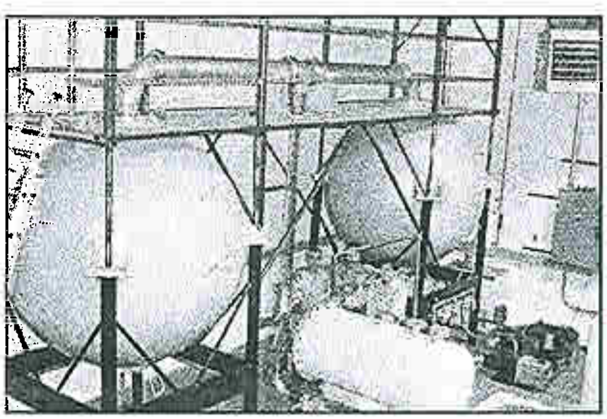


Figure 3. Picture of ALPHA facility

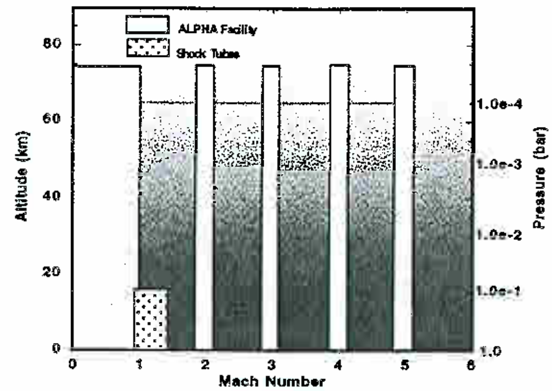


Figure 4. Diagram of the operating range of ALPHA and Shock Tubes

Defining the aerodynamic stability limits for agent drops requires a set of parameters that consider the competing aerodynamic force exerted by the supersonic flow field versus the internal restoring force of the drop. One such dimensionless parameter is the Weber number, shown as:

$$We = \frac{\rho v^2 D}{\sigma} \quad (1)$$

Where ρ is the air density, v is the flow velocity, D is the scale size of interest (droplet diameter in this case), and σ is the surface tension of the drop. The Weber number shows the ratio between the aerodynamic pressure force, which distorts the drop, and the surface tension force, which acts to restore the minimum energy configuration of the drop. The purpose of the initial ALPHA experiments was to determine the critical value of the Weber number at which the an agent simulant drop goes unstable and breaks up. This is done experimentally by systematically testing the behavior of drops at different Weber numbers to define the critical limit. The variable parameters in the experiment are the pressures at the supply and receiving tanks that regulate the velocity and density of the air flow in the test section. Exo and Endo-atmospheric release conditions at supersonic velocities can result in a wide range of Weber numbers from $1 < We < 10^5$. Table 1 shows the range of Weber numbers that can be accessed by ALPHA.

M	Pressure ratio	We Pr =1Pa	We Ps= 1bar	We Ps= 3bar
1	1.9	0.3	10[illegible]	3x10[illegible]
2	7.8	1.0	10[illegible]	3x10[illegible]
3	367	2.0	5x10[illegible]	1.5x10[illegible]
4	151	4.0	2x10[illegible]	6x10[illegible]
5	529	6.0	10[illegible]	3x10[illegible]

Drop diameter = 5mm. Surface tension = 0.027N/ m

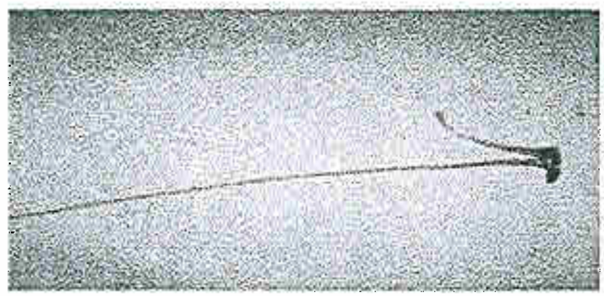
Pr: Pressure in the Receiver tanks
Ps: Pressure in the Supply tank

Table 1. Operating range of ALPHA in terms of Weber number

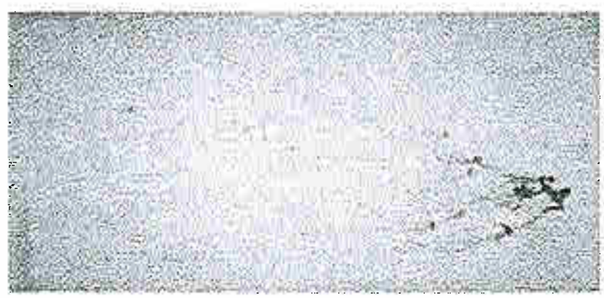
The droplet stability experiments carried out for our scoping case involved M=3 flow conditions at approximately 6.5×10^{-5} bar pressure using unthickened TBP liquid. The Weber number was slowly increased until the threshold for breakup was reached. This set of experiments revealed both the critical Weber number for breakup as well as the stable configuration of drops below the critical limit. The critical Weber number for the unthickened TBP was found to be We ~ 8. This identified threshold gave us a the ability to assess the breakup potential of drops for the demonstration case. We emphasize that this result is indicative only, since the critical limit is expected to change depending upon variations in thickening of the liquid as well as the Mach number. However, it should be noted that the presence of thickener will probably increase the critical Weber number and result in larger stable liquid masses. These changes can also result in a different breakup morphology, depending upon the factors listed above, as shown in Figure5. Observations of stable drops below the critical limit showed a flattening of the drop into a disk shape. This shape was maintained during the entire time scale of measurement. The flattened shape of the drop increases drag significantly and will affect the reentry of stable drops as shown in later sections.

Drop Toxicity and Atmospheric Settling Time

Additional key pieces of knowledge needed for assessing the risk of lethal agent fallout are the definitions of agent size ranges that represent the greatest threat. The size of individual agent drops determines the rough level of toxicity presented as a contact hazard to ground personnel. The size of agent drops also determines the immediacy of the threat in terms of drop fall times. Exo-atmospheric release of persistent agent can be expected to cull out drops with sizes in the ten to few hundred micron range. The settling velocities for these small particles are low enough (sub cm/sec rates) that agent in these sizes will diffuse very broadly and be subjected to hydrolysis, ultraviolet and other atmospheric degradation mechanisms over periods of days to weeks. However larger sized drops, up to millimeters in size, may fall on time scales of tens of minutes and not disperse widely. An important question is posed by asking what is the overlap of agent drop length scales in terms of toxicity and settling time? Figure 6 shows a contrast of the two factors and defines a lethal band of agent drop sizes that are of great concern when assessing agent fallout risk.



Thickened TBP Breakup, We = 30, M = 3



Unthickened TBP Breakup, We = 18, M = 3

Figure 5. Breakup behavior of unthickened and thickened TBP in supersonic, rarefied flow

The upper end of the lethal band of drop sizes, shown in red, is determined by the aerodynamic stability limit for aerobreakup and the lower end of the band is determined by a combination of atmospheric dispersion and toxicity level of the agent. Superimposed on this lethal band are estimates of the single drop lethal doses presented by certain persistent, organophosphorous agents [3].

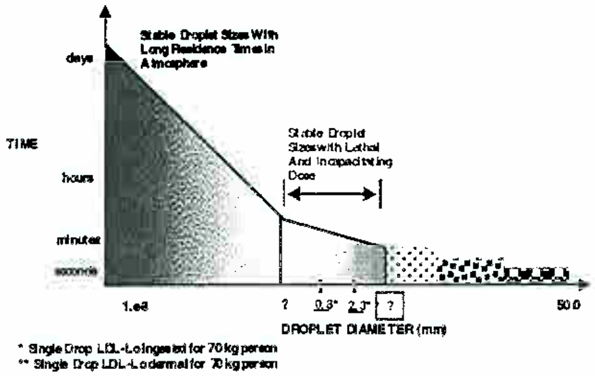


Figure 6. Diagram of the lethal band of agent drop sizes in terms of toxicity and atmospheric settling time

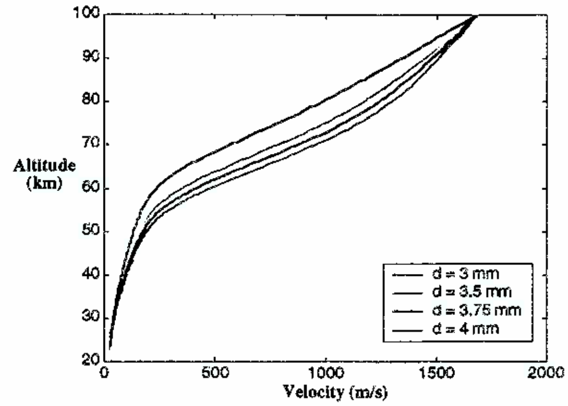


Figure 7. Particle tracker results of deceleration histories of millimeter sized agent drops

High Altitude Transport and Agent Stability

In order to quantify the reentry conditions that stable drops of agent would experience, particle tracking codes were written to model the deceleration and ballistic spreading of an agent cloud produced during the 100 km exo-atmospheric demonstration. The purpose of these calculations was to examine the deceleration characteristics of different sized agent drops and assess what the maximum stable size would be for a representative intercept case. The particle tracking codes calculated the equation of motion for the drops with the proper drag corrections for the stable, flattened drop geometry in supersonic conditions. The thermal content of the drop was also calculated considering free-molecular and transition aerodynamic heating along with radiative cooling.

Figure 7 shows the deceleration history of drops in the 3-4 millimeter range with a release height of 100 km and initial velocity of 1.67 km/s and reentry angle of 30 degrees. The primary deceleration of the agent occurs well above 40 km in the very rarefied regions of the atmosphere.

The Weber number for these droplets was simultaneously calculated and is shown in Figure 8. This calculation revealed that maximum stable agent drop sizes lay in the *3-4 millimeter range*! Furthermore, the deceleration profiles indicate that exo-atmospherically released drops see a gradually increasing Weber number that allows the larger sized drops to maintain a near critical size. These calculations, anchored by the data from ALPHA, indicate that large stable size drops of agent can exist in the single lethal dose range as they enter the lower atmosphere.

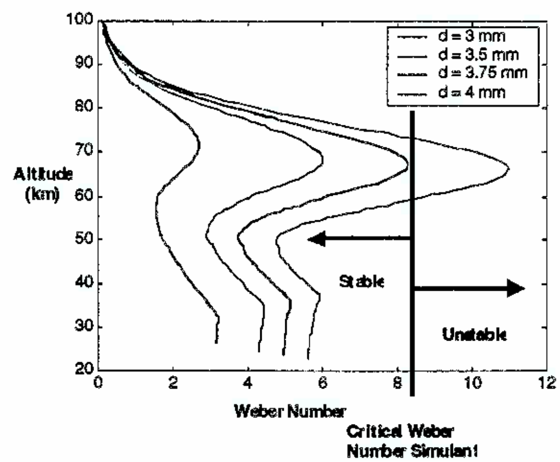


Figure 8. Weber number histories for millimeter sized agent drops

Atmospheric Dispersion and Ground Deposition

The last component of the demonstration case included the modeling of the atmospheric dispersion of the agent cloud using the well developed LLNL National Atmospheric Research Advisory Capability (NARAC). The original distribution of stable sized drops is log-normal with approximately half the initial mass (400 kg initial mass) going into significant size (500 micron to few millimeter) drops. This collection of drops was distributed into 25 size bins and transported using the particle tracker code. At 30 km in altitude, the resulting plume of spatially and temporally distributed droplets was input into the NARAC dispersion codes to calculate ground deposition resulting from fallout over a real geographical location with real wind conditions.

The NARAC capability is composed of a weather and dispersion modeling system. The ADAPT/LODI modeling system is used for both real-time operational applications and detailed assessments of events involving atmospheric releases of hazardous material at the Department of Energy's (DOE) National Atmospheric Release Advisory Center (NARAC) at Lawrence Livermore National Laboratory (http://narac.llnl.gov). The meteorological data assimilation model ADAPT constructs fields of such variables as the mean winds, pressure, precipitation, temperature, and turbulence, using a variety of interpolation methods and atmospheric parameterizations [4]. Non-divergent wind fields are produced by an adjustment procedure based on the variational principle and a finite-element discretization. The dispersion model, LODI, solves the 3-D advection-diffusion equation using a Lagrangian stochastic, Monte Carlo method [5,6]. LODI includes methods for simulating the processes of mean wind advection, turbulent diffusion, radioactive decay and production, bio-agent degradation, first-order chemical reactions, wet deposition, gravitational settling, dry deposition, and buoyant/momentum plume rise. The models are coupled to NARAC databases providing topography, geographical data, chemical-biological-nuclear agent properties and health risk levels, real-time meteorological observational data, and global and mesoscale forecast model predictions. The NARAC modeling system also includes an in-house version of the Naval Research Laboratory's mesoscale weather forecast model COAMPS [7].

The subsequent atmospheric dispersion of the varying agent sizes resulted in wide affected areas, on the order of thousands of square kilometers. However, agent drops in the 2-3 millimeter size range landed in higher concentrations, in approximately an 850 km^2 area, with single lethal dose drops impacting every 18x18 meter square area in the affected zone. Casualty numbers cannot yet be attached to the deposition contours that we have obtained. The relationship of casualties to the ground deposition density and individual drop toxicity is an uncertainty that needs to be addressed in our proposed risk assessment framework.

This demonstration clearly illustrates that it is physically possible for significant amounts of lethal sized agent to descend to earth , over a wide area, following a successful exo-atmospheric ballistic missile defense impact of a chemical warhead. The results of the demonstration confirms our premise for pursuing the path of Risk-Oriented assessment and seeking a higher level of physical understanding of the fallout processes.

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