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A COMPREHENSIVE ANALYSIS OF CHLORINE TRANSPORT AND FATE FOLLOWING A LARGE ENVIRONMENTAL RELEASE

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ABSTRACT

A train derailment occurred in Graniteville, South Carolina during the early morning of January 6, 2005, and resulted in the release of a large amount of cryogenic pressurized liquid chlorine to the environment in a short time period. A comprehensive evaluation of the transport and fate of the released chlorine was performed, accounting for dilution, diffusion, transport and deposition into the local environment. This involved the characterization of a three-phased chlorine release, a detailed determination of local atmospheric mechanisms acting on the released chlorine, the establishment of atmospheric-hydrological physical exchange mechanisms, and aquatic dilution and mixing.

This presentation will provide an overview of the models used in determining the total air-to-water mass transfer estimated to have occurred as a result of the roughly 60 tons of chlorine released into the atmosphere from the train derailment. The assumptions used in the modeling effort will be addressed, along with a comparison with available observational data to validate the model results. Overall, model-estimated chlorine concentrations in the airborne plume compare well with human and animal exposure data collected in the days after the derailment.

Key Words: atmospheric modeling, chlorine release, air-water mass transfer

1 INTRODUCTION

Two trains collided on a rail spur in the town of Graniteville, South Carolina in the early morning of January 6, 2005. This collision caused a breach and subsequent depressurization of one rail car, resulting in the rapid release of approximately 60 tons of chlorine to the environment[1]. Following the release, a dense cloud of chlorine vapor settled toward the west and southwest into a shallow valley bisected by Horse Creek, which bounds Graniteville to the west. Over the next three hours, the dense chlorine cloud began mixing with the ambient atmosphere and was subsequently transported out of the area by the prevailing south-southwesterly wind. The resulting environmental damage included 9 fatalities, 72 hospitalizations, animal and fish kills, and a severe bleaching of vegetation within a radius of approximately 1km.

Graniteville is located in southwestern South Carolina in a region of gently sloping terrain, referred to as the piedmont. The derailment occurred in the Horse Creek valley, oriented from north to south (Figure 1). The prevailing south-southwest wind at the time of the collision was the result of clockwise flow around a surface high pressure system centered off the southeast

United States coast. Observations in the region show that the wind near the surface was relatively light, ambient temperatures were mild, and measured atmospheric turbulence intensities were indicative of conditions supporting moderate to weak dispersion.

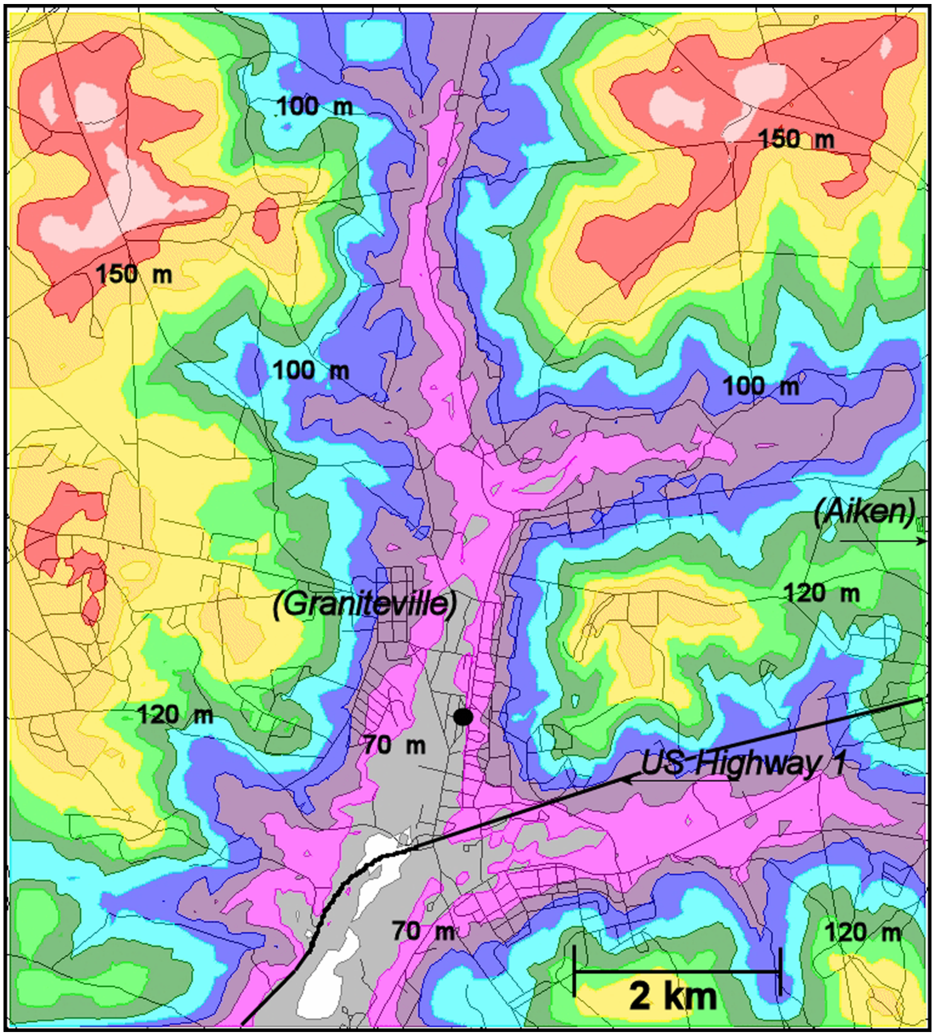

Figure 1: Terrain in the area with contour intervals of 10 meters. Gray and purple shading denotes the lowest elevations, while red and pink shading indicates the highest elevations. Gray shading indicates topographic elevation exceeding 50 m but less than 60 m AGL while light pink shading indicates topographic elevation exceeding 160 m AGL. The large black circle denotes the derailment site, while lighter lines indicate local roads.

There are several water bodies in the vicinity of the derailment that were impacted by the chlorine release (Figure 2). Of these , Horse Creek, oriented from north to south with an average width of 10 m and an average depth of 0.8 m, received the most chlorine from the accident. Horse Creek is located approximately 200 m west of the derailment site at its closest proximity.

The purpose of this paper is to (1) characterize the rapid evolution of the cryogenic pressurized liquid chorine released immediately to the environment from the breached railcar (i.e, the source term); (2) provide a detailed quantitative description of the transport and diffusion of the resulting airborne cloud of chlorine vapor throughout the Graniteville area; and, (3) characterize the interaction of the vapor cloud with the surrounding terrestrial environment, including an estimate of the amount of chlorine deposited into and transported by the adjacent surface waters.

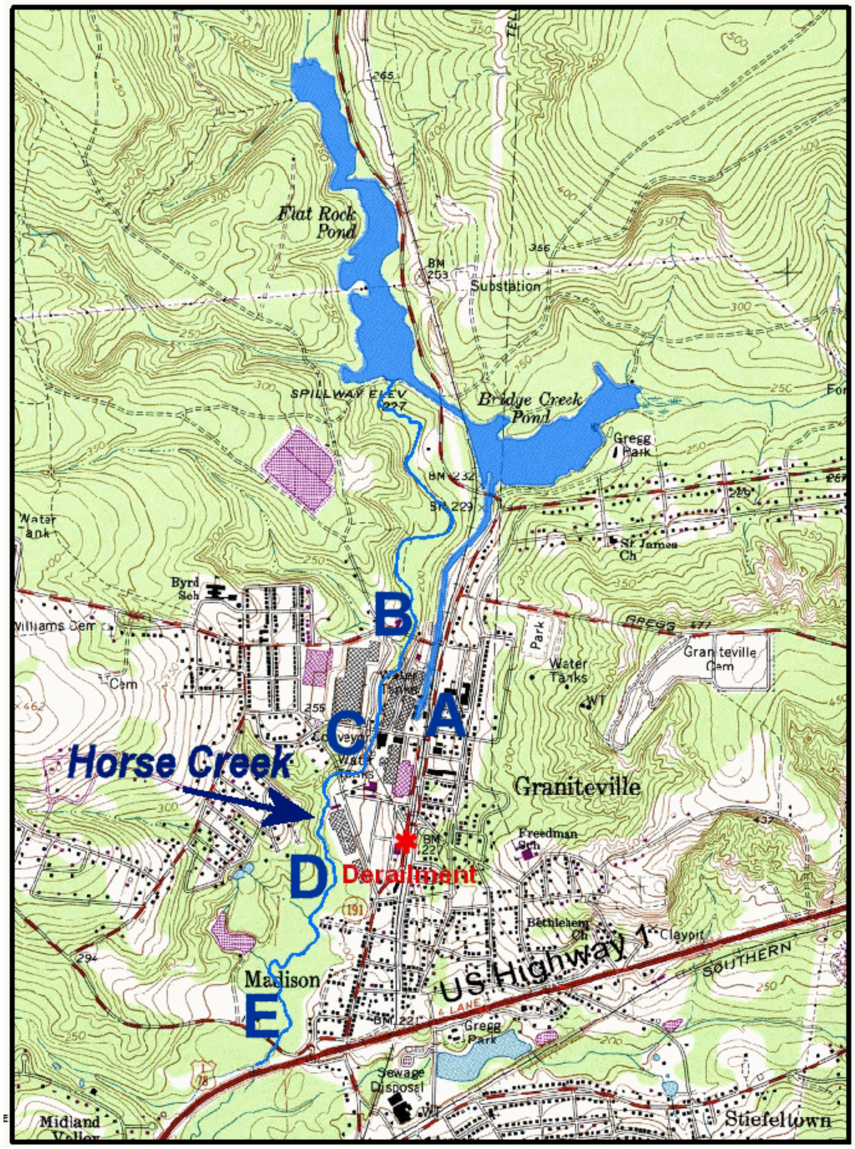

Figure 2: Water bodies considered for this study. Horse Creek runs from north to south and is located just west of the derailment site, while a canal stops just to the north of the site. Letters denote measurement locations discussed later.

All calculations were based on a rapid discharge of 60 tons of chlorine from the railcar, as reported by the National Transportation Safety Board[1]. The meteorology was simulated using the Regional Atmospheric Modeling System (RAMS[2]). Particular attention was placed on accurately characterizing near-surface conditions that affected the diffusion and transport of the initial large, dense discharge from the railcar over and out of the study area. The calculations were run for three hours after the railcar was breached, a period that bounds the eventual dissipation and transport of this large initial release out of the Graniteville area. Transport and dispersion calculations were based on the Second-Order Closure Integrated Puff (SCIPUFF[3]) module of the Hazard Prediction Assessment Capability (HPAC[4]) code, using detailed local meteorological conditions generated with RAMS. Since depletion of the chlorine vapor cloud is important[5] and poorly understood, SCIPUFF was run with a range of deposition velocities and the results compared to published data on human health effects, animal and fish mortality, and bleaching of vegetation.

Ambient chlorine concentrations near the surface, calculated by SCIPUFF, were subsequently used to calculate the amount of chlorine absorbed by the surface waters of interest using a hydrodynamic code developed at SRNL (ALGE[6]). Using an appropriate set of mass transfer coefficients that quantify the chlorine deposition (i.e., absorption) into, or re-emission from the water, ALGE was used to determine the total amount of chlorine absorbed, and the instantaneous time-dependent chlorine concentration in the water due to dilution, transport, and mixing.

2 ENVIRONMENTAL MODELING TECHNIQUES

2.1 Detailed Simulation of Local Meteorological Conditions

RAMS is commonly used by the meteorology community for conducting refined, local scale simulations. The RAMS simulations of local meteorology were conducted using five nested grids with horizontal grid spacings of 20.48 km, 5.12 km, 1.28 km, 320 m, and 80 m. The lateral boundary conditions were provided by output data from the National Center for Environmental Prediction (NCEP) Rapid Update Cycle (RUC) model analysis. A terrain-following coordinate system was used in the vertical dimension using a variable spacing that increased from 15 m in the lowest layer to 1000 m at the highest level. For topography, a high resolution LIDAR dataset (10m) was used to more accurately simulate the interaction between the local hills and valleys and the nightime airflow (Figure 1).

The simulation for this study was initiated at 7:00 pm EST January 5, 2005 (0000 hours UTC January 6, 2005) with appropriate RUC data for initial and boundary conditions. Detailed meteorological conditions were simulated until 6:00 am EST (1100 UTC), January 6, 2005 and the meteorological data from the innermost (5^{th}) grid was then used as input for the transport and diffusion calculations using HPAC.

2.2 Source Term Characterization

Once the railcar was breached, the sudden depressurization resulted in a rapid vaporization (i.e., flashing) of the cryogenic liquid, and an immediate energetic discharge of chlorine to the environment in the form of vapor, aerosols, and liquid. A determination of the rate and composition of the initial release was performed with NOAA's Areal Locations of Hazardous Atmospheres (ALOHA) code[7] and the Industrial Transportation (ITRANS) module of HPAC[4].

The ALOHA calculation resulted in an estimated total chlorine release of 59,000 kg (65 tons) discharged as a two-phased mixture of gas and aerosol due to immediate flashing of the liquefied material from rapid depressurization. ITRANS was used to determine the fraction of gas, aerosol, and liquid. Most of the discharge occurred in the first minute with the total release duration of 5 minutes. This short duration release is corroborated by recent research[8,9], in which detailed calculations of chlorine releases from irregular breaches of the size for the Graniteville accident (i.e., approximately 30 in long by 3 in wide) were estimated. The release of vapor and aerosol was assumed to occur over a period of three (3) minutes, as suggested by the ALOHA calculation and the recent work of Britter et al.[9] while the pooled liquid was assumed to be released instantaneously and then evaporate over time. The starting time of the release was assumed to be 2:40 am EST (the time of the train collision).

2.3 Dense Gas Dispersion

HPAC was developed and is currently maintained by the U.S. Army Defense Threat Reduction Agency[4]. This code was chosen for its ability to simulate dense gas releases. HPAC comprises a suite of programs to read in the gridded meteorological data generated by RAMS and interpolate it to a second user-defined grid defined within HPAC, characterize in detail the source term (chlorine), and run a transport and diffusion code to generate output.

Atmospheric transport, diffusion, and deposition processes acting on the chlorine plume was modeled using the SCIPUFF module within HPAC Version 5.0[3]. SCIPUFF describes diffusion processes using second-order turbulence closure by relating the dispersion rate to turbulent fluctuations in wind direction and wind speed.

Most models used for emergency response tend to ignore removal (deposition) processes for chemicals. However, this can lead to overestimation of the areal extent of the hazard. Thus, deposition was included in this model through the specification of a single dry deposition velocity, v_d. In general, values for v_d for reactive gases such as chlorine will be highly variable in space and time within a given modeling domain, depending on the nature of the surface, the areal distribution and density of structures and vegetation, time of day (i.e. physiological response of vegetation, or for chlorine, photolysis by sunlight), and time of year (i.e. the presence of leaves on deciduous trees). Values of v_d between 1 cm/s and 5 cm/s have been cited in the literature[4]. For large releases such as occurred at Graniteville, surfaces near the release location in contact with extremely high concentrations could quickly become saturated with chlorine, leading to a substantial reduction in the deposition velocity, and thus a concomitant reduction in the magnitude of chlorine plume deposition. Hill and Chamberlain[5] describe experimental data that show the adsorption of chlorine into vegetation becomes very small above an air concentration of 1 ppm, due to damage to the plant's stomatal structure.

Transport and dispersion calculations were performed over a three-hour period starting at 02:40 EST. After 05:40 EST, a substantial portion of the chlorine cloud from the initial release had been transported out of the area.

2.4 Aquatic Dilution and Mixing

When air containing vapors of chlorine or chlorine compounds such as hydrochloric acid (HCl) flows over water some of the vapor will transfer to the underlying water by molecular diffusion. Chlorine dissolved at the water surface will be further transferred to the deeper part of the water body by both molecular diffusion and turbulent mixing within the water body. In general, the magnitude of the turbulent mixing is much larger than that of molecular diffusion, as is the case in the atmosphere. The distribution of the chlorine concentrations between the gaseous form in the air and the dissolved form in the water is often described by Henry's Law, which states that at the interface between the atmosphere and a water surface, the concentrations of chlorine in the atmosphere and in the water are in equilibrium, C_{film}.

The concentration in a thin layer near the surface of the water body can be determined as a function of air concentration, absolute temperature, and Henry's constant. This concentration is then used in conjunction with the interface area between water and air, the depth of the water body, the time of the exposure, and the mass transfer coefficient to determine an overall mass transfer from air to water.

Specifically, the flux of gaseous chlorine into the water bodies is described by Lyman's Two-Film Theory[10] in which the movement of chlorine through the media is considered in two parts: (1) gas-phase; and, (2) liquid-phase. The rate of absorption is estimated as the ratio between the driving forces of mass transfer (the concentration gradient of the target compound) and the resistance to mass transfer (the overall mass transfer coefficient, K_W). The K_W for the liquid phase was established by combining the individual phase mass transfer coefficients for water and air, k_W and k_A, respectively, with a constant that determines their relative interaction (i.e., Henry's constant). Conservative estimates for the chlorine mass transfer coefficients for water of 1.6 cm/hr for the ponds and canal, and 3.8 cm/hr for Horse Creek were used in the calculations.

Reference data were collected for the measured equilibrium concentrations at varying partial pressures, and these values were compared to the concentration predicted by Henry's Law[11]. The highest partial pressure of chlorine above the water surface predicted by HPAC-SCIPUFF for this evaluation was approximately 99 Pa. Since there is no reference data available at this low level of partial pressure, a ratio of actual concentration to that predicted by Henry's law (8.8) was used to predict C_{film}, as this represents the lowest pressure for which there is reference data for comparison. It should be emphasized that this is likely a conservative estimate, as the actual ratio is expected to be higher. Note also that with a ratio of measured concentration to predicted concentration as high as 8.8, the hypothesis that Henry's law alone does not describe the solubility of chlorine in water is validated, particularly at low partial pressures.

The ALGE model was developed by the Savannah River National Laboratory to simulate the discharge of effluent into surface water systems such as tidally-driven estuaries, rivers and cooling lakes. In this application, ALGE used the chlorine concentrations generated by SCIPUFF. It should be noted that surveys of water body depths were made to allow for more realistic simulations.

2.5 Flow Determination

The flow velocity for ALGE was set equal to zero for the larger, stagnant water bodies, (Figure 2). Note that the zero flow assumption potentially underestimates chlorine deposition, as it results in lower turbulent mixing within these water bodies, hence less transfer of chlorine across the air-water interface. For Horse Creek, the flow rate was conservatively estimated to be 1 m^3/s using gauge data from a nearby creek.

3 RESULTS

3.1 Simulated Meteorological Conditions

At the time of the Graniteville derailment, a moderate surface pressure gradient present over the region resulted in the pronounced south-southwesterly wind across the Graniteville and Horse Creek Valley area. Near the surface, winds were simulated to be generally south-southwesterly to southwesterly at speeds ranging from 2 m/s to 6 m/s on well-exposed hill tops to 1 m/s or less in the Horse Creek valley (Figure 3). A few isolated areas of near stagnant flow are present (e.g., just to the southwest of the derailment site). In addition, some terrain-induced channeling is also evident. A strong temperature inversion was present at the derailment site that extended from just above the surface upward to 200 m AGL (not shown). Weak turbulence was believed to be present in a shallow, slightly stable surface layer, which enabled the transfer of momentum from the strong southwesterly flow in the inversion layer toward the ground, negating the possible formation of pronounced gravity-driven drainage flows down the Horse Creek valley toward the southwest. Model results were compared to available observations of wind direction and wind speed in the vicinity of Graniteville. In general, RAMS produced a reasonable representation of local meteorological conditions that occurred on the night of the derailment, providing confidence in the subsequent transport results using SCIPUFF.

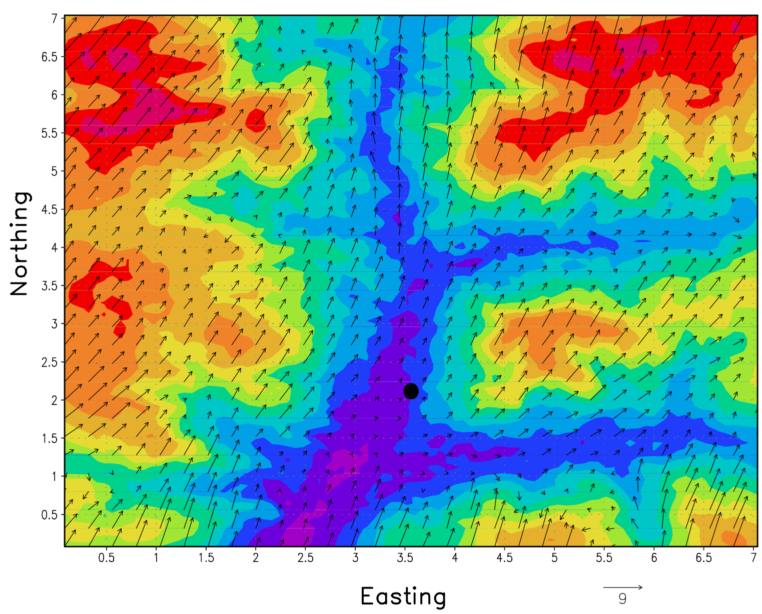

Figure 3: Near surface wind vectors (m/sec) at 2:40 a.m. (0740 UTC), January 6, 2005. Topography is indicated by shading, grid units are in kilometers, and the large black circle indicates the derailment site.

3.2 Dense Gas Transport and Dispersion

The evolution of the chlorine plume from HPAC/SCIPUFF over the 3 hours of the simulation period is illustrated by the contours of integrated surface dose in Figure 4. Plots of the maximum

1-hour integrated ground-level concentration (i.e., chlorine dose) that occurred over the 3-hour period of simulation are shown for 3 different deposition velocities. Contour levels correspond to integrated surface concentration (i.e., dose) values of 2, 5, 20, 400, and 2000 parts per million (ppm) chlorine.

(a) (b) (c)

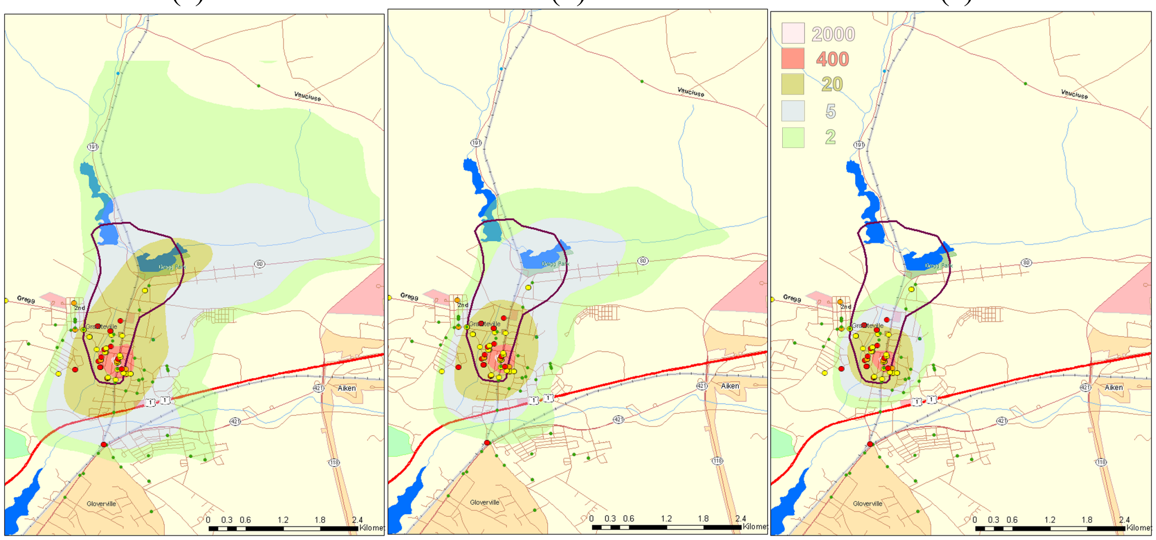

Figure 4: Plots of integrated dose (60-min) for various dry deposition velocities (v_d). The dose values (ppm) are indicated in the legend. The bold purple line indicates approximate vegetation damage, while the markers denote health effects as categorized by SCDHEC[14]. The severity of the cases are ranked by color, where red is the most severe, followed by orange, yellow, green and blue, with blue being the least severe. (a) v_d = 0 cm/s, (b) v_d = 1 cm/s, and (c) v_d = 3 cm/s.

The 2 and 20 ppm levels correspond to the Environmental Protection Agency[12] Acute Exposure Guideline Levels (AEGLs) -2 and -3 for a 60-minute exposure period, respectively. The AEGL-2 level represents the airborne concentration above which the general population, including susceptible individuals, could experience irreversible or other serious, long-lasting, adverse health effects or an impaired ability to escape. The AEGL-3 level represents the airborne concentration above which the general population, including susceptible individuals, could experience life-threatening health effects or death. It is clear from Figure 4 that increased values for deposition velocity leads to significant reductions in the chlorine footprint. The 20 ppm contour approximates reasonably well the visual evidence from the NTSB report[1] for the cases with $v_d > 0$.

Footprints of instantaneous chlorine concentration (Figure 5) depict a time sequence of results for a domain-averaged v_d equal to 1 cm/sec. The plume is seen to spread generally with the terrain both initially at the 10-minute time step and at later times as shown for the 20- to 60-minute time steps. As expected with gravity-driven dense gas releases, higher concentrations tend to occur at the lower elevations. The general orientation of the plumes depicted in Figure 5 and the dose footprints shown in Figure 4 would indicate the competing influences of both the initial effects of gravity on the dense gas as it spreads laterally into the Horse Creek valley, and the prevailing south-southwesterly winds which transports the plume northward through the Graniteville region[13]. Later in the simulation, the concentrations decrease as the dense cloud spreads over a broader area and, as the cloud progressively loses the characteristics of a dense gas, begins to disperse through mixing in the ambient atmosphere and is transported northeastward.

The chlorine footprints were subsequently compared to available data on human health outcomes, reports of animal mortality, and observations of vegetation damage[13]. The human health data, obtained from SCDHEC[14], consists of the locations of occurrence for each of five categories of exposure outcome ranging from 1 for no exposure to 5 for extreme exposure[15]. A comparison of the modeling results with a plot of the exposure outcome data and vegetation damage suggests that model calculations performed with a value for an average value for v_d of 1.0 cm/s generally provided the best agreement with the exposure data. Instances of moderate to extreme exposure are represented by the yellow, orange and red dots in the two figures and are strongly correlated with a moderate to severe medical outcome (i.e., hospitalization or death). These are bounded appropriately by the AEGL-2 and AEGL-3 threshold levels, in virtually all cases. Locations of the nine reported deaths were all within the 500 ppm contour (Fig. 5a) where brief exposures to concentration approaching 1000 ppm could be expected. This is judged to be a reasonable result given the nature of the surface characteristics for the area over which the plume is transported, and the published range of estimated dry deposition velocities for gaseous chlorine. These results are also consistent with an assessment of animal mortality[16].

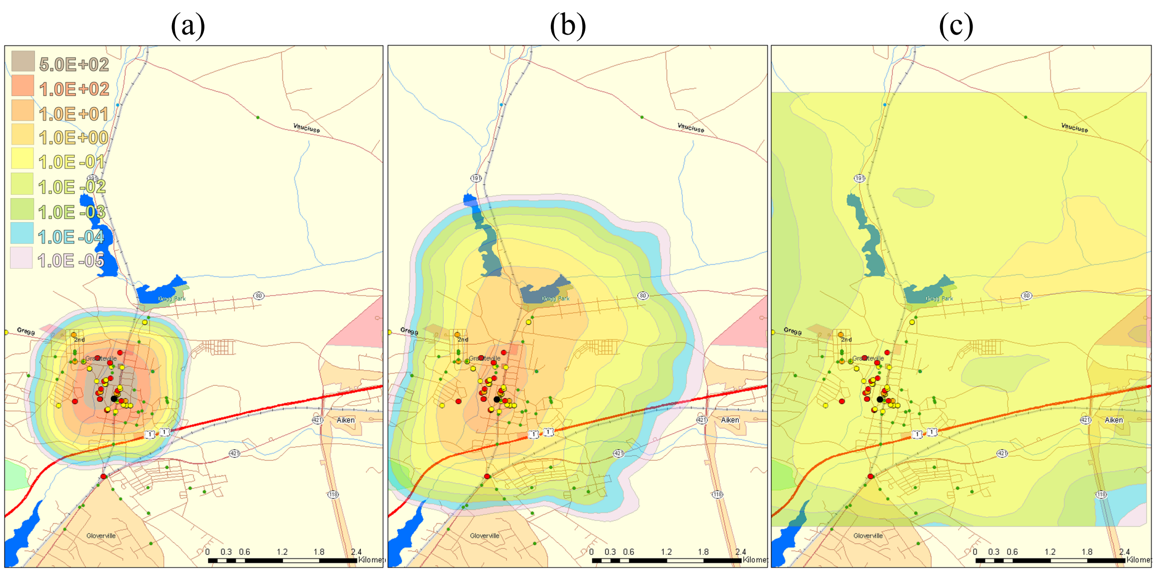

Figure 5: Instantaneous surface concentration (ppm) for v_d = 1.0 cm/s at (a) 10 min, (b) 20 min, and (c) 60 min, after the start of the release with values as noted in the legend. The location of the derailment is indicated by the black circle. Markers denote health effects as in Figure 4.

3.3 Deposition, Dilution and Mixing of Chlorine into Affected Waters

Plots of time-dependent instantaneous concentrations calculated by ALGE are shown in Figure 6, while the locations for each of these plots are shown in Figure 2. Figure 6a illustrates that the chlorine concentrations in the stagnant water bodies increase rapidly as the cloud progresses northward. Although airborne chlorine continues to be transported over these waters from dissipation of both the large initial release and the ongoing residual release from the railcar and surrounding contaminated areas, the air concentrations are sufficiently low to allow diffusion of chlorine from the water surface back into the atmosphere above.

Figure 6b illustrates that water concentrations increased rapidly to significant levels at locations in Horse Creek west and southwest of the derailment site, as the core of the dense chlorine cloud sinks into the shallow valley. (Note that the concentration scales in each figure differ). Chlorine concentrations in the creek peak as high as 0.85 g/m^3 at location D from a combination of chlorine deposition from the air and the transport of chlorine from deposition occurring upstream. Furthermore, concentrations exceed 0.5 g/m^3 for nearly one hour at these locations. The displacement of concentration plots with time for locations C, D, and E clearly reflect the ongoing transport of the slug of deposited chlorine downstream. Predicted concentrations decrease rapidly after the dense cloud breaks up and is transported northeastward and out of the area as a neutrally buoyant gas, and chlorine in the water begins to diffuse back into the relatively clean air above.

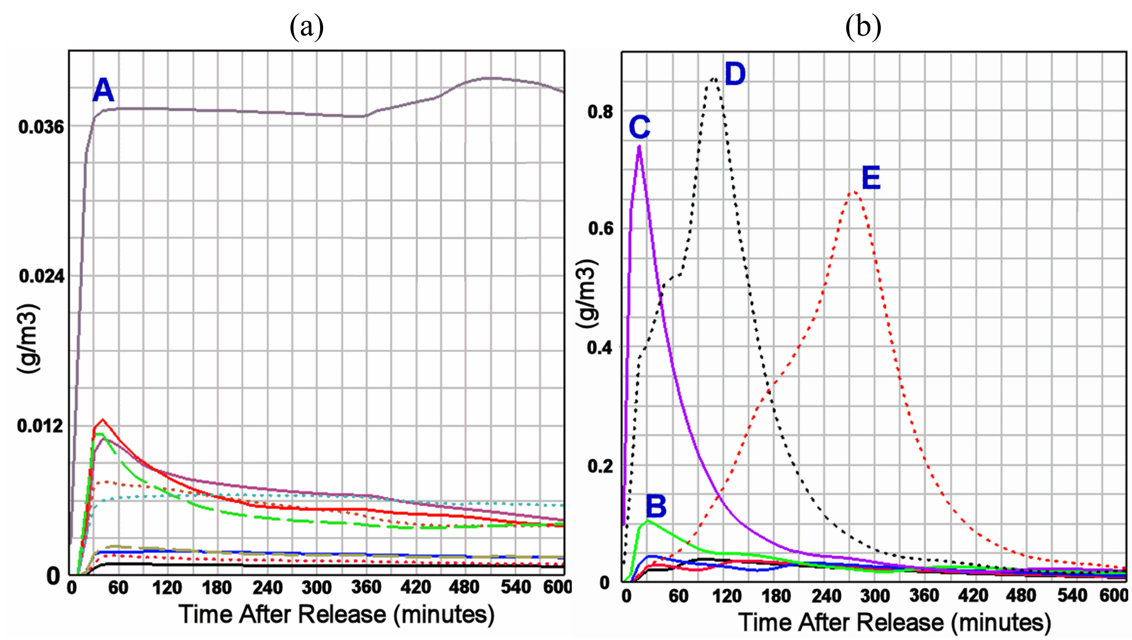

Figure 6: Calculated chlorine concentrations (g/m3) as a function of time at selected locations in (a) the stagnant water bodies, and (b) Horse Creek. The locations A to E are indicated in Figure 2.

Fish mortality data collected after the derailment[17] were assessed using the chlorine concentrations in Horse Creek calculated by ALGE[16]. Results from this analysis confirmed the death of fish species with LC50 concentration (the lethal concentration for 50% of exposed fish) values above 0.5 g/m^3. Furthermore, fish deaths were reported in species with LC50 values well over 1 g/m^3, suggesting the calculations for chlorine deposition into Horse Creek are conservative.

4 CONCLUSIONS

This evaluation used several state-of-the-art benchmarked computer codes to determine the total amount of chlorine vapor that was deposited from the atmosphere into nearby surface waters in association with the train derailment that occurred in Graniteville, South Carolina during the early morning of January 6, 2005. The derailment resulted in the emission of a large amount of cryogenic pressurized liquid chlorine to the environment in a relatively short time period.

Results for total chlorine deposited into applicable surface waters were determined for a range of conditions to account for varying chlorine deposition rates, mass transfer coefficients for chlorine absorption, and other variables. The amount of uncertainty was significantly reduced since the model results were carefully compared to available human mortality and morbidity data, fish mortality data, and observations of tree damage, all resulting from exposure to various concentrations of airborne chlorine from the derailed tank car. These comparisons corroborated the choices in model variables and confirmed the models' ability to estimate the actual physical processes that took place at the time of and subsequent to the derailment with reasonable reliability.

This evaluation concluded that of the approximately 60 tons of chlorine released from the breached railcar, ~11 kg of chlorine was deposited into the adjacent surface waters. Model calculation performed with RAMS, HPAC, and ALGE were compared to available data (i.e, medical outcome, fish toxicity data) and in each case were found to provide reliable results.

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