Environmental impact assessment of selected explosives using field techniques, analytical laboratory and numerical models

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Abstract

Environmental impacts of sites contaminated with explosives have raised considerable concerns in the United States, Canada, and Europe. Assessment of risks and cleanup of these sites are required for protection of human health and the environment. Monitored natural attenuation (MNA) is a potential and cost-effective remedial alternative for explosives-contaminated groundwater at sites where a decline in contaminant mass can be demonstrated to occur at a rate sufficient to ensure the protection of potential receptors. The objective of this study was to provide a technical basis for a systematic measuring strategy for natural attenuation at sites contaminated with explosives. The data collection and processing procedures for evaluation, selection, and implementation of MNA for explosives took a two-year demonstration project followed by long-term monitoring. The sources of contamination (lagoons) had been removed and treated, but groundwater contamination remained. The sampling at the site included groundwater collection from thirty wells quarterly and soil sampling using the direct push technology tools. These data have been used to refine the conceptualization of the site hydrogeology and to collect aquifer material to further support site characterization. The samples have been analyzed for explosives and explosive transformation products and were used in the investigation of microbial biomarker techniques for tracking attenuation processes. A conceptual model of the contaminant plume was developed and a three-dimensional numerical model was used to predict future plume migration and mass. The results are promising and indicate that natural attenuation is a viable remediation option for the site.

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1 Introduction

Monitored natural attenuation (MNA) is a potential remedial alternative for explosives-contaminated groundwater at sites where a decline in contaminant mass can be demonstrated to occur at a rate sufficient to ensure the protection of potential receptors. MNA tasks includes complete characterization of the site hydrogeology and contaminant distribution, long-term monitoring of groundwater, and groundwater modeling to conceptualize the contaminant plume and to predict future migration and attenuation.

The U.S. Environmental Protection Agency has issued a directive concerning the use of MNA at Superfund (inactive), RCRA (active), and underground storage tank sites as corrective action [1]. The guidance requires (1) “Historical groundwater and/or soil chemistry data that demonstrate a clear and meaningful trend of decreasing contaminant mass and/or concentration over time at appropriate monitoring or sampling points, (2) Hydrogeologic and geochemical data that can be used to demonstrate indirectly the type(s) of natural attenuation processes active at the site, and the rate at which such processes will reduce contaminant concentrations to required levels, and (3) Data from field or microcosm studies (conducted in or with actual contaminated site media) which directly demonstrate the occurrence of a particular natural attenuation process at the site and its ability to degrade the contaminants of concern (typically used to demonstrate biological degradation processes only). The groundwater numerical models used to conceptualize and quantify trends in the contaminant plume using real data from the site over time in support of (1) above. The direct push technology can provide sample material in support of (2) and (3) above as well as lithological data for refining site conceptualization.

The data collection and processing procedures for evaluation, selection, and implementation of MNA for explosives took two-year demonstration project followed by a long-term monitoring. The sources of contaminations (lagoons) had been removed and treated, but groundwater contamination remained. The sampling at the site included groundwater collection from thirty wells quarterly and soil sampling using the direct push technology tools. These data have been used to refine the conceptualization of the site hydrogeology and to collect aquifer material to further support site characterization. The samples have been analyzed for explosives and explosives transformation products and were used in the investigation of microbial biomarker techniques for tracking attenuation processes. A conceptual model of the contaminant plume was developed and a three-dimensional numerical model was used to predict future plume migration and mass.

1.1 Locations and hydrogeology characteristics

The site lies within the Western Gulf Coastal Plain physiographic province. Two major landforms, dissected uplands and rolling prairie, are found within the site (Figure 1). Minor landforms include abandoned channels that are typically filled with clays deposited by ancient courses of the ancestral Red River (Figure 2).

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Relief at the site is moderate with elevations varying from about 130 feet (40 m) above mean sea level (MSL).

Previously reported hydrogeologic data at the site indicated that the subsurface geology formation (the Pleistocene section) could be subdivided into Upper and Lower Terrace aquifers [2, 3]. Groundwater in the Upper Terrace aquifer generally exists under water table (unconfined) conditions at depths varying from approximately 5 to 25 feet (1.5 to 7.6 m) below surface ground level (BGL). The Lower Terrace aquifer, while not present in all areas, typically occurs from 25 feet (7.6 m) BGL to the top of the Cane River formation, a confining layer, which is 50 feet (15.20 m) BGL. The Lower Terrace aquifer tends to produce more water than Upper Terrace deposits.

The site selected for this study consisted of 16 unlined lagoons covering approximately 25 acres in the south central part of the installation. Untreated explosive-laden wastewater from munitions loading and packaging operations was discharged into the lagoons at the site. The site also was used as a burning ground for many years. As part of an interim remediation action, the wastewaters at the site were removed and the soil was excavated to a depth of 5 feet (1.5 m). Excavated soil was incinerated and used to backfill the area. The concentration of explosives in the treated soil was below detection. A natural cap of low permeability was placed over the site to inhibit infiltration and further migration of residual explosives below the excavation depth.

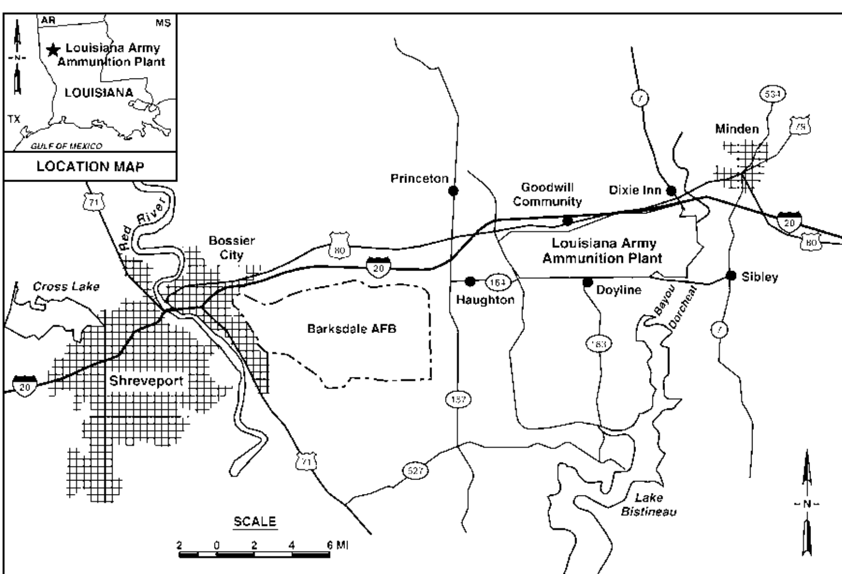

Figure 1: The site plant map and location.

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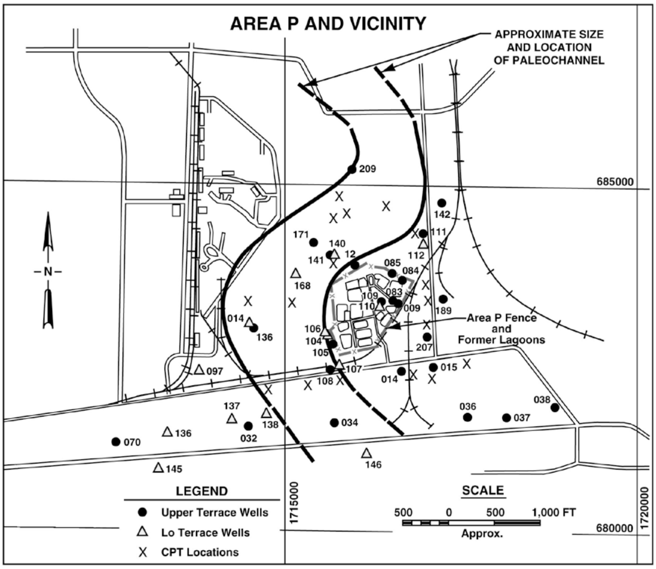

Figure 2: Fluvial channels.

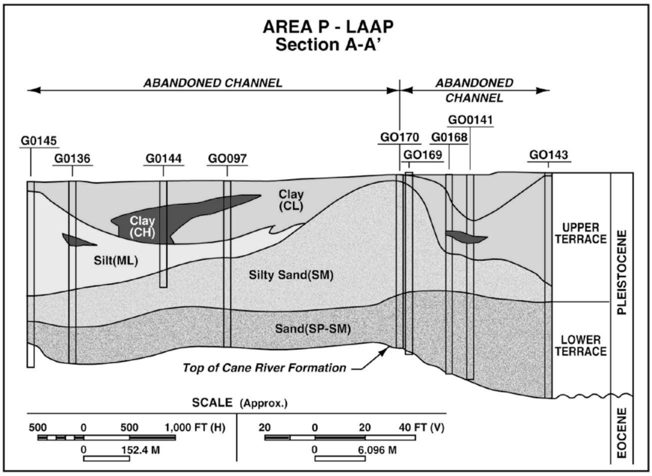

Figure 3: Cross-section A-A'.

1.2 Site geology

Historical geological reports [4, 5] and data collected using the direct push technology (DPT) supported the natural attenuation demonstration project [6] indicated that the shallow aquifers underlying the site consist of Pleistocene age terrace deposits unconformably overlying the Cane River Formation. The subsurface lithology is shown in three different cross-sections (Figures 3, 4,

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and 5). The Terrace deposits in the site (Area P) are subdivided into the Lower Terrace consisting of fine sands and a trace of gravels, and the Upper Terrace consisting of very fine-grained silts, clays and silty clays. An intermediate clay unit is present at some locations, is not uniform over the entire site, and is totally absent at many locations. However, the unit does serve as a limited aquitard as evidenced by the Paleochannel defined by the direct push technology. Collectively, these Pleistocene age units are a fining upwards sequence deposited during a waning glacial episode. The Eocene age, Cane River Formation consists primarily of clay or clay sufficiently indurated to be classified as a claystone. The Cane River Formation is not an aquifer beneath Area P, and is, therefore, considered the confining layer for modeling the site.

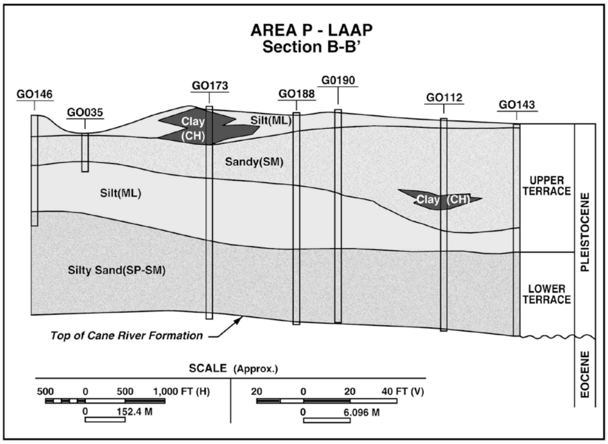

Figure 4: Cross-section B-B'.

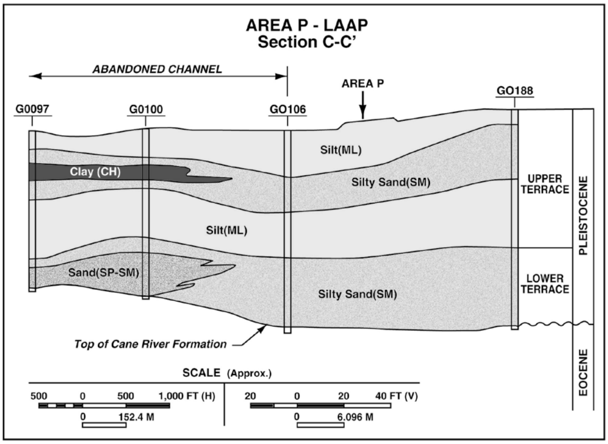

Figure 5: Cross-section C-C'.

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Ground water in the Upper Terrace aquifer generally exists under water-table (unconfined) conditions at depths varying from approximately 5 to 25 feet below surface ground level (BGL). The Lower Terrace aquifer, while not present in all areas, typically occurs from 25 feet BGL to the top of the Cane River, which is about 50 feet BGL. The Lower Terrace aquifer also tends to produce more water than the Upper Terrace deposits. Although none of the Terrace deposits supply water to production wells on the installation, some domestic wells in Haughton, Princeton, Dixie Inn, Minden, Sibley and Doyline are completed in the Terrace deposits. Ground water quality modeling conducted for the site indicated that contaminants (explosives) migration in the Upper Terrace generally traveled downwards with little horizontal spreading [5]. Further, the modeling and water level measurements indicated that the regional groundwater flow in the Upper Terrace aquifer was southwest. Water level data collected for the Natural Attenuation demonstration project [6] indicated that the direction of ground water movement is in different directions in the Upper versus the Lower Terrace aquifers. The Paleochannel located on the western edge of Area P also influences the rate and direction of ground water movement.

1.3 Site climate

The climate of northwest Louisiana is classified as subtropical-humid and continental, with hot summers and cool winters. The average temperature during summer is 81 degree Fahrenheit (F). August is the hottest month with an average temperature of 83 F. During winter, the average temperature is 47F. The average rainfall for the site, as measured for the period 1931-1980 by the National Oceanic and Atmospheric Administration for northwestern Louisiana is 48.80 inches. The months of November through May receive rainfall between 4.16 inches (February) and 4.81 inches (December). August, September, and October are the driest months, receiving an average rainfall of between 2.28 inches and 2.81 inches. The normal average temperature is 65 $^{\circ}$F. The rainfall data measured during 1931 to 1992 were used in a statistical analysis and result is shown in Figure 6.

2 Direct push technology sampling events

The direct push technology (Cone Penetrometer and Hydropunch) sampling event was conducted at the site to support the Natural Attenuation research effort between September 9 and 29, 1996. Soil samples were collected from 24 locations along eight transects. Penetrations were through the entire Upper and Lower Terrace sections. Generally, the penetrations were about 50 feet deep and reached total depth in the Cane River Formation. The locations were selected on the basis of ground water sampling data collected monthly during the previous six months. Additional data collected at the site [2] was utilized to stratify the various lithologies at the site and assist in the location of the sampling sites. TNT and RDX concentrations from previous ground water sampling were contoured to identify “hot spots” and potential source areas. Transects were

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located to assure sampling along a line extending from the zone of highest concentrations to a zone of zero concentration in all four cardinal directions from the source (original lagoons). A thin vertical slice of the soil obtained in the split spoon was removed and analyzed in the field for explosives [7]. The remainder of the sample was retained for laboratory confirmation of explosives and additional research. Vertical profiles of soil were collected at five locations. Depending upon the depth of the CPT hole, samples from varying depths were collected at each location. These soil samples were used to measure vertical variations in permeability for the ground water model and for other research.

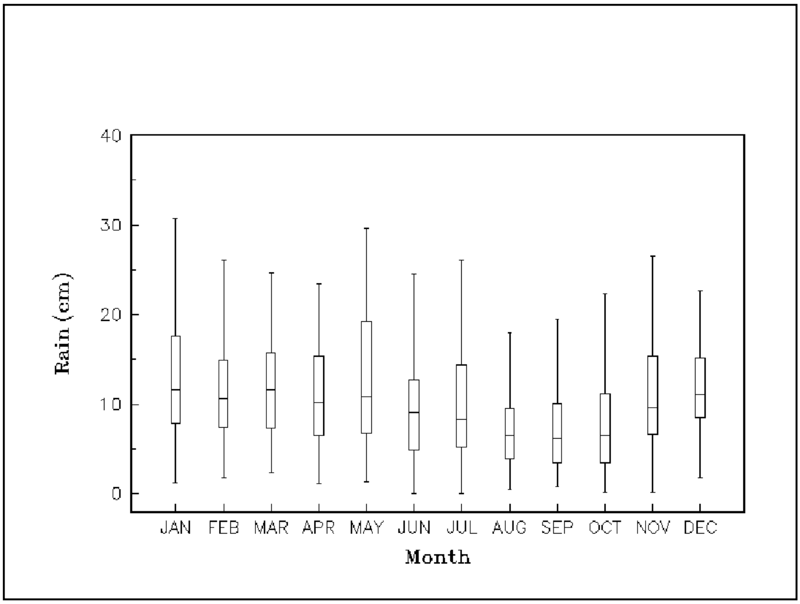

Figure 6: Statistical representation of rainfall data measured at Minden, LA during 1931-1992. The horizontal line in each box represents the median amount of rainfall per month and the limits of the box represent the 95 percent confidence interval for each month over the time period from 1931 to 1992. The vertical bars represent the range of values.

Analysis of existing geological information indicated the existence of a Pleistocene-aged paleochannel on the western edge of the site. This paleochannel was first observed as a meander scar (abandoned channel), visible in some of the earliest aerial photographs of the site. The CPT sampling defined the dimensions of the paleochannel. The paleochannel is at least 200 feet wide (400 feet at some locations), and 25 to 30 feet deep. The hydrogeology effect of this feature is that the low permeability clays and silty clays in the paleochannel act as an aquitard. These clays and silty clays locally separate the Upper and Lower Terrace aquifers and steer the contaminants to the Lower Terrace.

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3 Adsorption coefficients

Results from laboratory batch testing for adsorption of explosives (TNT and RDX) were used to develop input data for modelling purposes. The initial exposure of uncontaminated soil to TNT and RDX could represent the soil response to the contamination front as it migrates through the aquifer. The rates of laboratory-measured sorption of TNT and RDX were about two orders of magnitude faster than the microbial mineralization. Pseudo-equilibrium of TNT and RDX with the site soils was reached within a few days. After equilibrium, the removal rate was dominated by the microbial activity. Adsorption coefficients for TNT and RDX in the site soils are presented in Table 1. The K_d values for TNT ranged from 0.08 to 0.33 L kg^{-1} depending on the type of soil. For RDX, the values ranged from 0.21 to 0.33 L kg^{-1}. An average value of 0.228 L kg^{-1} for TNT and 0.30 L kg^{-1} for RDX were used for the modelling.

Table 1: Explosives adsorption coefficient (K_d, L/Kg) for LAAP aquifer soil and regression coefficient (r^2)[1].

Compound	ML Soil		SP-SM Soil		CL Soil		SM Soil	
	K_d	r^2	K_d	r^2	K_d	r^2	K_d	r^2
TNT	0.33	0.96	0.23	0.99	0.27	0.92	0.08	0.90
RDX	0.21	0.95	0.33	0.97	0.33	0.83	0.33	0.95

[1]Batch tests conducted under aerobic conditions.

3.1 Decay rates

A critical input to the model is an estimate of the rate of contaminant decay or removal that is reflective of the dominant biogeochemical pathways at the site. Direct measurement of in situ rates of microbial degradation currently is not possible. One approach to measure the degradation rates is to sample aquifer sediments and monitor contaminant disappearance as a function of time using batch or column reactors operated under controlled laboratory conditions. Alternatively, the use of radiorespirometric techniques can be applied to measure microbial degradation potential in the laboratory. Radiorespirometry indicates the potential for complete mineralization. The actual rate in the groundwater would differ from laboratory tests due to inherent differences in the biomass, temperature, electron acceptors, and mass transfer limitations.

The decay rates from the laboratory batch studies and radiorespirometric studies on the site soils are given in Tables 2 and 3. The batch tests were conducted using uncontaminated site soils exposed to groundwater contaminated with TNT and RDX (90 g soil/360 g water). The apparent rate constants ranged from 0.01 to 0.03 days^{-1}, with corresponding half-lives on the order of month (Table 2). The heterogeneity of the soil and the complexity of physical and chemical interactions in a multicomponent system render the generation of these values difficult. The radiorespirometric data are based on mineralization of 30 percent slurry of the site soil exposed to either TNT (2 mg L^{-1}) or RDX (21 mg L^{-1}). The apparent rate constants from radiorespirometry are one to two

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orders of magnitude lower than the batch testing results with half-lives ranging from one to ten years. In general, the highest rates of removal were associated with clay soils.

In spite of the limitations of the data, the laboratory tests provided a good approximation of site conditions and should be taken into account by the model. The batch tests would simulate uptake rates at the edge of the plume where uncontaminated soils are initially exposed to TNT and RDX. Because the site has a history of over 40 years of exposure to explosives, microbial degradation is likely to be the dominant factor controlling the removal rate. Therefore, the decay rates from Radiorespirometry (Table 3) were closer to the decay values used in the model.

Table 2: Apparent first order removal rate constants for uptake of TNT and RDX from groundwater on uncontaminated LAAP soil[1].

Soil Type	**TNT**		**RDX**	
	Decay rate constant, 1/day	**Half-life, days**	**Decay rate constant, 1/day**	**Half-life, days**
Sandy silt ML	0.014	48	<0.002	> 350
Sandy silt (SP-SM)	0.014	48	<0.002	> 350
Lean clay (CL)	0.034	20	<0.002	> 350
Silty sand (SM)	0.017	42	<0.002	> 350

[1] Water source was well number MW085u; Results are from batch tests conducted anaerobic conditions; initial concentrations were approximately 8 and 10 mg/L TNT and RDX, respectively.

Table 3: Apparent first order microbial mineralization rate constants for degradation of TNT and RDX from in LAAP soil[1].

Soil Type	**TNT**		**RDX**	
	Radiorespirometry rate constant, 1/day	**half-life, days**	**Radiorespirometry rate constant, 1/day**	**half-life, days**
Sandy silt ML	5.7×10^{-4} to 2.2×10^{-3}	320 to 1220	1.8×10^{-4}	3850
Sandy silt (SP-SM)	4.6×10^{-4}	1510	5×10^{-4}	1390
Lean clay (CL)	$< 1 \times 10^{-4}$	>3900	$< 1 \times 10^{-4}$	> 3900
Silty sand (SM)	$< 1 \times 10^{-4}$	>3900	2×10^{-3}	323

[1]Radiorespirometry tests were run at 23.3± 3.2 degrees C under aerobic conditions. Initial phase concentrations were 2 and 21 mg/L TNT and RDX, respectively.

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The field concentration data represent the change in contaminant concentration at specific locations at the site and incorporate all removal mechanisms and mass transport limitations. The decay rates used in the model were based on the Radiorespirometry results (Table 3) and model calibration using field concentration data. The values used in the model were 10^{-5} day^{-1} (half life of 190 years) for TNT and 8.13 x 10^{-6} day^{-1} (half life of 233 years) for RDX.

4 Numerical modelling

Numerical modelling provides a mean of quantitatively evaluating hydrogeology and multiple natural processes that can be represented as a set of mathematical expressions. Numerical modelling was conducted to compliment the field monitoring and data collection for demonstration and graphic representation of natural attenuation of explosives at the site. The modelling effort focused on conceptualisation of the site hydrogeology and reduction of explosives by processes such as immobilization/degradation, and first order decay.

The Department of Defense Groundwater Modeling System [8] with its subsurface model, FEMWATER [9], was selected for the modeling element of the demonstration. The GMS includes numerical tools to facilitate site conceptualization, mesh and grid generation, geostatistical computations, and visualizations.

4.1 Code description

FEMWATER [9] is a three-dimensional finite element numerical code, which may be used to model flow and mass transport through saturated-unsaturated media. FEMWATER is an enhanced version of two models, 3DFEMWATER (flow; [10]) and 3DLEWASTE (transport; [11]). FEMWATER is integrated into GMS. The flow equations in FEMWATER are based on the continuity and Darcy flow equations. The model application is limited by the assumptions applied to these equations. FEMWATER also can be used for density-dependent problems. FEMWATER simulates the primary processes affecting dissolved-phase contaminant distributions in groundwater including advection, dispersion, sorption, and decay caused by chemical reactions and/or biological transformation. FEMWATER uses the first-order decay as lumped biochemical decay.

4.2 Model limitations

Major assumptions and limitations of FEMWATER include the following: (1) the contaminant is transported as a single constituent, thus inter-solute reactions can not be simulated, (2) abiotic and microbial degradation is treated with a first-order decay model, (3) adsorption coefficient and decay rates can be assigned for different subsurface materials; however, rate constants do not change during simulation time, (4) contaminant sorption is instantaneous and reversible and the adsorbed phase is in local equilibrium.

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Some of the above assumptions may not be applicable to certain field problems. For this application, the above assumptions were applicable by simplifying some site characteristics required for the modeling without deviating much from actual site conditions. For more details, the reader is referred to FEMWATER model theory documentation [9].

4.3 Model construction

FEMWATER requires basic hydrogeologic and chemical data for simulations. These basic data include hydraulic conductivity, porosity, hydraulic gradient, initial and boundary conditions, distribution (partition) coefficients, and decay rates. The distribution coefficient, K_d, relates the sorbate and solute for linear isotherms. Distribution coefficients for explosives were determined on aquifer soil from the site [6].

The modelling domains were defined in three dimensions. In plane view at the site the domain was roughly bounded by D Line, and Pearl Harbor Ave (Fig. 2a) to the west and south, respectively. In the vertical direction the modeling domain included the Upper and Lower Terrace Aquifers (Fig. 2 c, d, e).

The retardation factor is a function of contaminant property, K_d, and soil bulk density, Δ_B, soil porosity, N, and soil water saturation, S_w. The retardation factor provides a general indication of mobility of the contaminant in the soil. Table 1 provides information on TNT and RDX adsorption coefficients and Table 2 shows first-order decay rates for TNT, RDX. The decay rates given in Table 2 were determined from the laboratory batch studies on the site soils [6]. The decay rate used in the model for both sites were 10^{-5} day^{-1} for TNT and 8.13 x 10^{-6} day^{-1} for RDX, which were considerably lower than the batch rates. These decay rates were used based on the model calibration, and on the fact that the field decay rates are normally lower than laboratory batch rates.

4.4 Conceptual model

A conceptual model is a powerful tool for abstracting and simplifying natural phenomena. To develop conceptual models for the site, existing and new hydrogeological data including borehole geologic DPT data, hydraulic conductivity data (measured in DPT samples) and flow boundary conditions were used.

Four stratigraphic units were identified at the site based on lithologic data, pump test data, and laboratory test results. The subsurface was divided into four hydrogeologic zones; an unsaturated (vadose) soil, an upper terrace aquifer, a semi-confined layer, and a lower terrace aquifer. The soil and the upper terrace aquifer form a shallow unconfined aquifer. The terrace deposits are composed of alternating beds of mixed sands and clay. The distribution of hydraulic conductivity for each layer was estimated from the conductivity data at the DPT locations.

The source of flow recharge at the site was assumed to be from rainfall. Precipitation data collected at the town of Minden, LA, were used to estimate average infiltration rate at the site. The yearly averaged rainfall of 1931-1992 data was 50.0 in. (127 cm).

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Figure 7: Initial (February 1996) distribution of TNT concentration.

Figure 8: Predicted distribution of TNT plume in the Upper Aquifer after 20 years.

4.5 Initial distributions of contaminants

Initial conditions of flow and chemical concentration play a major role in the model outcomes. Different numerical techniques available in GMS were compared to establish a realistic initial flow and mass concentration distributions at the sites. The starting (initial) conditions for the simulation were estimated

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based on the first monitoring data. The initial flow and concentration distribution of TNT and RDX were determined using data collected in February 1996 at the site. The GMS was used to interpolate/extrapolate the data for all points of the numerical mesh system.

Figure 9: Predicted distribution of TNT plume in the Lower Aquifer after 20 years.

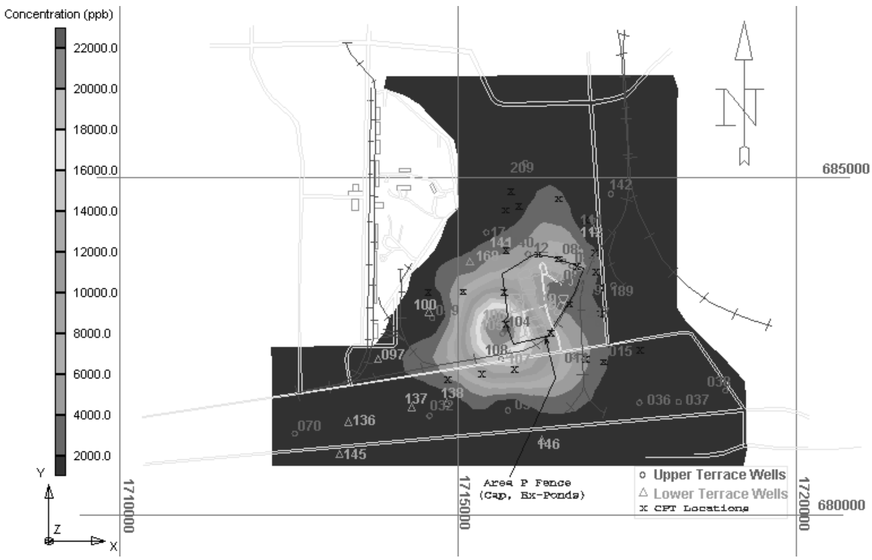

Figure 10: Initial (February 1996) distribution of RDX concentration.

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Figure 11: Predicted distribution of RDX plume in the Upper Aquifer after 20 years.

Figure 12: Predicted distribution of RDX plume in the Lower Aquifer after 20 years.

4.6 Predictive simulations

The long-term prediction of the plume is needed to demonstrate a clear and meaningful trend of decreasing contaminant mass and/or concentration over time. Prediction requires calculating future flow and transport conditioned using available data. The future boundary and other required model conditions are a mathematical statement of certain hypotheses, based on past experiences. The results presented here are based on the following assumptions: 1) no additional source of contamination is added into the site, 2) infiltration rate stays constant throughout the simulations, 3) flow boundary conditions recur every year, 4) no recharge or discharge through pumping occurs during the simulations. The predicted results should be updated and adjusted as new data become available. Jorgensen [12] illustrates an iterative way in which a model prediction may be improved as new information is obtained.

When initial conditions (Figures 7 and 10 for TNT and RDX, respectively) are compared with simulations for 20 year (Figures 8 and 9 (TNT) and Figures 11 and 12 (RDX)), the areal extent of both plumes is diminishing. The highest concentration is reduced from 10,500 to 2134 μgL^{-1} for TNT and 23,200 to 5534 μgL^{-1} for RDX. The highest concentration after 20 yr are in the Lower Aquifer for both TNT and RDX. However, the initial (February 1996) highest concentrations were in the Upper Aquifer. This indicates that the plume has moved from the Upper Aquifer to the Lower Aquifer. The predicated results should be updated, adjusted, and verified as new data become available. An example in which a model prediction can be improved as new information becomes available is described by [12].

5 Conclusions

Sampling, analytical laboratory tests, and numerical modelling refined hydrogeology characterization and contaminant plume extension and fate. By coupling field analyses and rapid laboratory “turn-around” with placement of the CPT, efficiency was optimised while minimizing analysis of uncontaminated samples beyond the plume. Lithology, permeability measurements and contaminant data from CPT samples contributed significantly to development of the site conceptual and numerical models.

The GMS provided efficient numerical tools to integrate and analyse the complex hydrogeologic and chemical field data into useful and easy to interpret data graphic forms. Predictive modelling of fate and transport of contaminant plumes at the site permitted informed decision-making regarding the selection of MNA for the site. Long-term monitoring of chemical data and hydrogoelogic parameters support future fate and transport of the explosives at the site and vicinities.

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