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Chemical Detection of Buried Landmines

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Of all the buried landmine identification technologies currently available, sensing the chemical signature from the explosive components found in landmines is the only technique that can classify non-explosive objects from the real threat. In the last two decades, advances in chemical detection methods has brought chemical sensing technology to the foreground as an emerging technological solution. In addition, advances have been made in the understanding of the fundamental transport processes that allow the chemical signature to migrate from the buried source to the ground surface. A systematic evaluation of the transport of the chemical signature from inside the mine into the soil environment, and through the soil to the ground surface is being explored to determine the constraints on the use of chemical sensing technology. This effort reports on the results of simulation modeling using a one-dimensional screening model to evaluate the impacts on the transport of the chemical signature by variation of some of the principal soil transport parameters.

landmines, chemical sensors, soil transport

I. INTRODUCTION

The organic chemicals of the explosives in the buried landmine environment can exist in or on four phases: solid phase of the neat explosive material, vapor phase in the soil air, aqueous phase in the soil water solution, and sorbed onto soil solid phases. The chemical signature begins as a surface coating from production or depot storage and through continuous emission by permeation through the mine case or through leaks in seals and seams. Once the chemicals enter the soil environment, they experience phase transitions, partitioning into the soil air, soil water and sorbing onto soil particles. The impact of temperature and chemical gradients, and precipitation/evaporation will cause movement of the chemical signature. Part of this transport is upward to the soil surface where chemical detection technology is envisioned to be used. Simulation modeling is a technique that can evaluate the impacts of many of the environmental variables that can dampen or accentuate the surface expression of the chemical signature. Model results will be shown that describe the magnitude of the changes that accompany variations due to chemical properties of the explosive and properties of the soil environment.

II. CHEMICALS IN THE SOIL ENVIRONMENT

Soils are a porous media with a number of physico-chemical properties that affect the transport of explosive chemicals. Soil bulk density is a measure of the compaction of the soil and is defined as

$$\rho_b = \frac{M_s}{V_s} \qquad [1]$$

where ρ_b is the soil bulk density (g/cm^3), M_s is the mass of soil particles (g), and V_s is the volume of soil (cm^3). Soils under natural conditions have bulk densities ranging from 1.0 to 1.8 g/cm^3. However, soils that have been excavated and replaced, such as during the emplacement of a landmine, may have bulk densities much less than 1. The soil bulk density is inversely proportional to the soil porosity as follows

$$\phi = 1 - \rho_b / \rho_s \qquad [2]$$

where ρ_s is the soil particle density (ranges from 2.6 to 2.8 g/cm^3 for most soils). The soil porosity, or void volume, is defined as

$$\phi = \frac{V_w + V_a}{V_s} \qquad [3]$$

where ϕ is the soil porosity (cm^3/cm^3), V_w is the volume of soil water (cm^3) and V_a is the volume of soil air (cm^3). Soil porosity values range from 0.3 for sands to 0.6 for clay rich soils. The volumetric moisture content describes how much water is present in the soil and changes greatly during precipitation/drainage events and evaporation conditions. Volumetric water content is defined as

$$\theta = \frac{V_w}{V_s} \qquad [4]$$

where θ is the volumetric water content (cm^3/cm^3). Soil moisture contents have values from near zero up to the soil porosity value. When the soils are not fully saturated, the balance of the soil pore space not filled with water is termed the air filled porosity, and is defined as

$$a = \frac{V_a}{V_s} \qquad [5]$$

where V_a is the volumetric air content (cm^3/cm^3).

It is often more convenient to use soil saturation (S_l) because it is a measure of the relative saturation of a particular soil pore space with water.

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DISCLAIMER

$$S_l = \frac{\theta}{\phi} \qquad [6]$$

Since the explosive chemicals can exist as solutes in the soil water and the movement of soil water can be a significant transport mechanism, water solubility is an important parameter. Water solubility is defined as

$$C_L = \frac{M_{chem}}{V_w} \qquad [7]$$

where C_L is the concentration in aqueous phase (g/cm^3 soil water) and M_{chem} is the mass of chemical (e.g. TNT) (g). Water solubility, however, is not constant and is typically an increasing function with temperature.

Henry's Law constant is a relative measure of the amount of the chemical that exists in the gas phase to that in the aqueous phase at equilibrium, and is defined as

$$K_H = \frac{C_G}{C_L} \qquad [8]$$

where K_H is the Henry's Law constant (unitless) and C_G is the concentration in gas phase (g/cm3 soil gas)

The soil partition coefficient is a relative measure of how much of the chemical is temporarily bound to the soil to that in the soil aqueous phase

$$K_d = \frac{C_S}{C_L} \qquad [9]$$

where K_d is the linear soil-water partition coefficient (cm^3/g) and C_S is the concentration sorbed on the soil solid phase (g/g of soil).

The soil water partition coefficient is often correlated with the fraction of organic carbon found in the soils. In this way, the variability between soils can be reduced. The organic carbon distribution coefficient is defined as

$$K_{oc} = \frac{K_d}{f_{oc}} \qquad [10]$$

where K_{oc} is the organic carbon distribution coefficient and f_{oc} is the fraction of organic carbon.

III. PHYSICAL CHEMICAL PROPERTIES OF EXPLOSIVES

The principal explosive chemicals found in landmines are TNT and RDX (NGIC, 1995). DNT, as a production by-products of TNT, is also considered to be a significant signature chemical for buried landmines. As a group, these chemicals have very low vapor densities and moderately low water solubilities. Table 1 shows these properties and the Henry's Law constant at 20°C (Phelan and Webb, 1997).

Table 1. Vapor Density, Water Solubility and Henry's Law Constant of Explosive Compounds at 20°C

	TNT	DNT	RDX
Vapor Density ($\mu g/m^3$)	43.5	122	0.024
Water Solubility (mg/l)	130	270	50
Henry's Law Constant	3.35E-7	4.51E-7	4.73E-10

Pennington and Patrick (1990) measured the soil water partition coefficient (K_d) of TNT in fourteen soils from locations across North America. The mean value was 3.8 cm^3/g with a standard deviation of 1.34. The highest value was 6.8 cm^3/g and the lowest value was 2.3 cm^3/g. Xue et al. (1995) evaluated two soils and showed mean values for TNT of 2.66 cm^3/g and 3.64 cm^3/g. DNT and RDX have very little data. Xue et al. (1995) showed values for RDX of 1.59 cm^3/g and 1.57 cm^3/g. McGrath (1995) showed a K_{oc} value of 251 for DNT. For the fourteen soils evaluated by Pennington and Patrick (1990), the mean value for the fraction of organic carbon was 0.0173 with a standard deviation of 0.11. Using these values, the K_d for DNT has a mean value of 4.4 ± 2.7 cm^3/g (one std. dev.). In summary, the soil water partitioning coefficients for TNT, DNT and RDX all fall into an approximate range between 1.5 and 7.0 cm^3/g. This is a rather narrow range as common chemicals can have values one to two orders of magnitude lesser and greater than these.

The biochemical half-life of explosives in near surface soils has not been studied well outside of the biotreatment technology area for contaminated soils. However, long-term surface soil degradation tests at Los Alamos National Laboratory (LANL) followed the degradation of soils doped with 1000 mg/kg of various explosives over 20 years (Dubois and Baytos, 1991). Table 2 shows the half-lives estimated from these long-term experiments.

Table 2. Estimates of Half-Lives of Explosives from LANL Long-Term Surface Soil Tests

Explosive	Half-Life (years)
TNT	1
RDX	36
HMX	39
PETN	92

IV. SCREENING MODEL

The environmental fate and transport of organic chemicals including volatilization and leaching losses has been used to explore the distribution of agricultural pesticides in soils (Mayer et al. 1974, Farmer et al. 1980, and Jury et al. 1980). These models were primarily intended to simulate specific circumstances. However, Jury et al. (1983, 1984a, 1984b, 1984c) developed and validated a general screening model (Behavior Assessment Model, BAM) that included volatilization, leaching, and degradation to explore the major loss pathways of agricultural pesticides as a function of specific environmental conditions. The model simulations can be used to assess the behavior of different chemicals under particular environmental conditions, but is not intended to predict a definitive concentration distribution in the field. As such, the predictions from the screening model are only an indication of expected conditions.

This model is valuable in that it can express the total concentration of a chemical in the gas, aqueous and sorbed phases. The total concentration is expressed as

$$C_T = \rho_b C_S + \theta C_L + a C_G \qquad [11]$$

where C_S is the concentration sorbed to the soil, C_L is the solute concentration in the aqueous phase, and C_G is the gas phase concentration. In addition, Jury (1983) shows how equation [11] can be rewritten in terms of one of the variables alone

$$C_T = R_S C_S = R_L C_L = R_G C_G \qquad [12]$$

where

$$R_S = \rho_b + \frac{\theta}{K_d} + a\frac{K_H}{K_d} \qquad [13]$$

$$R_L = \rho_b K_d + \theta + a K_H \text{, and} \qquad [14]$$

$$R_G = \rho_b \frac{K_d}{K_H} + \frac{\theta}{K_H} + a \qquad [15]$$

are the solid, liquid and gas phase partition coefficients, respectively.

An adaptation of the BAM was developed to be applicable to the conditions of contaminated soil buried under a known depth of clean soil - Buried Chemical Model, BCM (Jury et al., 1990). Simulations based on a modification of Jury's BCM are used in this report to simulate the behavior of the chemical signature from buried landmines. The Buried Chemical Model of Jury et al. (1990) is based on the following assumptions. A detailed discussion of these assumptions is given in Jury et al. (1990).

1. The chemical may adsorb on the solid phase, be dissolved in the aqueous phase, or exist in the vapor phase.
2. The chemical flux is the sum of the vapor flux and the dissolved solute flux using Fick's law.
3. The porous medium factors for gas and liquid phase diffusion are given by the Millington and Quirk (1961) model as extended for liquid diffusion by Jury et al. (1983).
4. The chemical will undergo first-order degradation due to biological and chemical effects.
5. Chemical movement is one dimensional.
6. The adsorbed and dissolved phases undergo reversible, linear adsorption.
7. The dissolved and gaseous phase concentrations are related through Henry's law.
8. The soil properties are constant in space and time.
9. Water flux is constant in space and time (relaxed in the present application).
10. Volatilization of the chemical to the atmosphere is by vapor diffusion through an air boundary layer of constant thickness.

In the present implementation of Jury's model, a constant source term has been added to reflect the chemical source from the landmine at a specific location.

Under these assumptions (including the source term) the model formulation becomes

$$\frac{\partial C_T}{\partial t} + \mu C_T = D_E \frac{\partial^2 C_T}{\partial z^2} - V_E \frac{\partial C_T}{\partial z} + \sigma \qquad [16]$$

where C_T is the total chemical concentration, μ is the biochemical decay constant, and σ is the source term. The effective velocity (V_E) is defined as

$$V_E = \frac{J_w}{\rho_b K_d + \theta + a K_H} \qquad [17]$$

where J_w is the precipitation/evaporation flux. The effective diffusion coefficient (D_E) of the chemical is defined as

$$D_E = \frac{a^{10/3} K_H D_g^a + \theta^{10/3} D_l^w}{\phi^2 \left(\rho_b K_d + \theta + a K_H\right)} \qquad [18]$$

where D_g^a is the diffusivity of the gas phase of the chemical in air and D_l^w is the diffusivity of the chemical in aqueous phase . The boundary conditions for the problem are diffusion through a boundary layer at the upper surface, and a zero chemical concentration at infinity at the lower boundary. These boundary conditions can be expressed as

$$-D_E \frac{\partial C_T}{\partial z} + V_E C_T = -H_E C_T \qquad [19]$$

where

$$H_E = \frac{h K_H}{\rho_b K_d + \theta + a K_H} \qquad [20]$$

and

$$h = \frac{D_g^a}{d} \qquad [21]$$

and

$$C_T(\infty, t) = 0.$$

The initial conditions are an initial concentration, C_0, over an interval from L to W, or

$$C_T(z,0) = 0 \qquad L \rangle z \rangle W$$

$$C_T(z,0) = C_0 \qquad L \le z \le W$$

The above model results in a closed form solution as a function of space and time; the results are rather lengthy and will not be presented here but are given by Jury et al. (1983, 1990). In the present simulation, the assumption of constant water flux in time will be relaxed. Therefore, sequences of water fluxes representing desired conditions (rainfall followed by evaporation) can be simulated to determine the effect of water flux variations on the location of TNT in the soil and the surface TNT vapor flux. A numerical solution was developed and verified by comparison to the results given by Spencer et al. (1988) and Jury et al. (1990) (Phelan and Webb, 1997).

Using this solution, simulations were performed using a landmine that has contributed an initial soil concentration (C_o) based on the surface contamination of the landmine. It has been assumed that the entire surface contamination was completely and uniformly transferred to the soil just prior to the beginning of the simulation runs. Surface contamination data (Hogan et al., 1992) showed a median surface contamination of 15 ng/cm^2 from 42 domestic and foreign landmines. Using the dimensions of an anti-tank (AT) mine of 30 cm diameter by 10 cm high, the surface contamination would provide 3.5×10^{-5} g of TNT for initial distribution in the soil. Using the volume of the AT mine that this mass of TNT is distributed into, the initial concentration (C_o) would be $\sim 5 \times 10^{-3}$ µg/cm^3.

The constant source term emanation rate was derived from vapor collection chamber experiments on two mines (Spangler, 1975). Values ranged from 10^{-16} to 10^{-18} g/cm^2-s. The higher rate of 10^{-16} g/cm^2-s (8.6×10^{-6} µg/cm^2-day) was used in these simulations. If the top of the AT mine was buried at a depth of 10 cm, the burial zone of the initial contamination is from 10 to 20 cm, and the constant source term is placed at a depth of 15 cm.

The diffusivity of gas in air (D_g^a) and diffusivity of liquid in water (D_l^w) were selected from Jury et al. (1983). The biochemical half-life value of 365 days was selected from a long term field experiment (Dubois and Bayton, 1991).

The precipitation/evaporation rates and periods followed in all the simulations here were the low desert scenario from Phelan and Webb (1997). This scenario was derived from data found in HELP (Hydrological Evaluation of Landfill Performance) model (Schroeder et al., 1994a and 1994b).. The HELP model showed that the low desert had 1 day of precipitation followed by 7 days of evaporation. For simplicity, total precipitation and total evaporation for each cycle are assumed to be equal and for these simulations the cycles were continued for approximately four to ten years.

Table 2 shows the input parameters used in the simulations.

Table 2. Simulation Parameters

parameter	units	base case	variant cases
θ	cm^3/ cm^3	0.25	0.375
ϕ	cm^3/ cm^3	0.5	*
ρ_b	g/cm^3	1.5	*
K_d	cm^3/g	1.6	3.8 6.0
K_H	--	5.9E-7	4.73E-10
air boundary layer	cm	0.5	*
$t_{1/2}$	days	365	180 60
C_o	µg/cm^3	4.6E-3	0
J_c	µg/cm^2-day	8.6E-6	0
D_l^w	cm^2/day	0.432	*
D_g^a	cm^2/day	4320	*
burial depth, top	cm	10	*
burial depth, bottom	cm	20	*
cycles/yr	--	45	*
precipitation	days	1	*
evaporation	days	7	*
precipitation rate	cm/day	0.44	*
evaporation rate	cm/day	- 0.063	*
total precipitation/ evaporation	cm/year	20	*

* - same as the base case

V. DISCUSSION

For each of the figures shown, there is a distinct oscillation of the surface vapor flux. This feature is a result of the cycling of precipitation and evaporation. To evaluate the effect of the Henry's Law constant, two simulations were performed where all parameters were kept constant with one case using a K_H equal to that of TNT and one for RDX (both at 20°C). Figure 1 shows the results and indicate that the TNT surface vapor flux would be expected to reach a greater steady state value than RDX, approximately proportional to the ratio of Henry's Law constants. For TNT, a temperature increase from 0°C to 40°C will increase the K_H value by a factor of about 100. It appears that seasonal and diurnal soil temperature changes could make a significant effect on the subsurface transport and surface flux of explosive signatures.

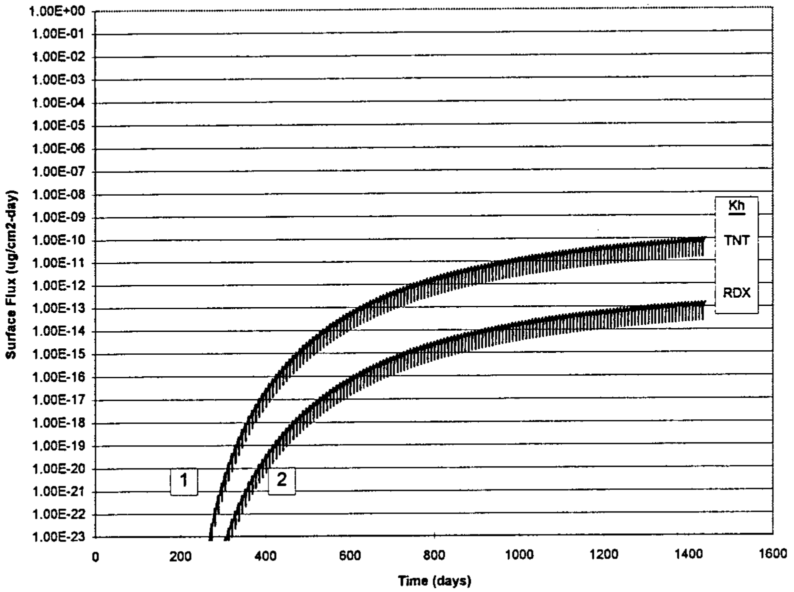

Figure 1. Effect of Henry's Law Constant on Surface Vapor Flux

Depth concentration profiles at the end of the simulation period show that for both cases the concentration of TNT and RDX are essentially equal. This implies that most of the transport upward to the ground surface is within the aqueous phase and that the release of the chemical into the vapor phase above the ground surface is directly proportional to the Henry's Law constant.

Next, simulations were performed to evaluate small changes in the soil water partitioning coefficient. The K_d values for TNT, DNT and RDX have values from about 1.5 to 7. Figure 2 shows the surface flux over time for K_d values of 1.6, 3.8 and 6.0 cm^3/g.

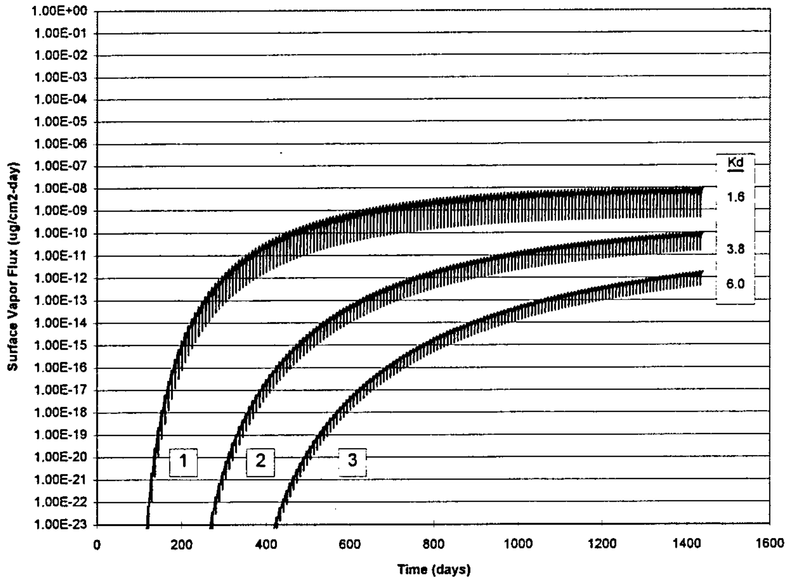

Figure 2. Effect of Soil Water Partitioning Coefficient on Surface Vapor Flux

These simulations show that even though the soil water partitioning coefficient appear to vary only slightly among many soils, there is a significant impact to the transport of the chemical to the ground surface. As the K_d value increases, the lag period becomes much longer and the steady state concentrations stabilize at much lower levels. Also with the lower K_d values, the effect of precipitation/evaporation cycles becomes more pronounced. This is consistent with the lower K_d value, since more of the mass of the chemical is found in the aqueous phase and is affected by the upward and downward flux of water. Figure 3 shows the depth profiles from the simulations varying the soil water partitioning coefficients. These curves show that the simulations with lower K_d values have more significant transport of the chemicals to soils both above and below the source zone than the higher K_d values.

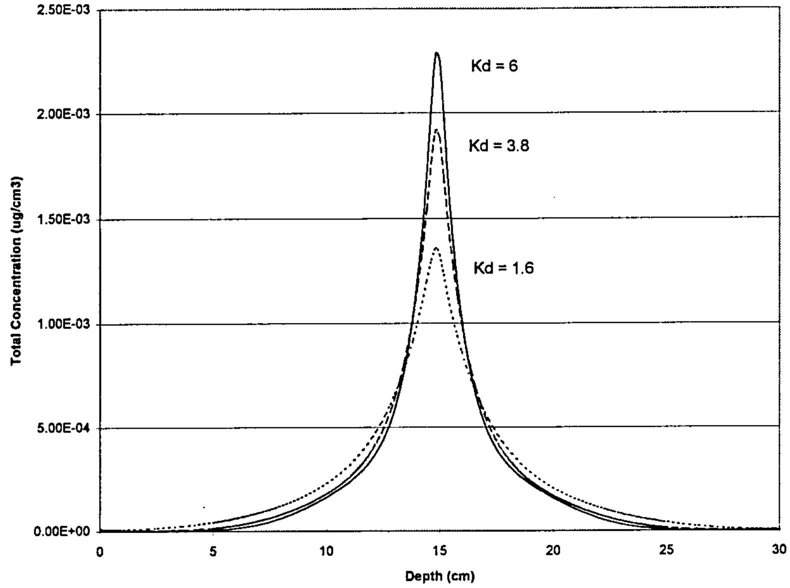

Figure 3. Depth Profile After 4 Years With Variant K_d Values

The next simulations were an evaluation of the source term parameters. Very little data is available on the initial concentration of explosives coating the outer surface of a mine and even less so on the surface emission flux. Figure 4, curve one shows the effect of reducing the surface emission flux to essentially zero (a value of 1E-20 ug/cm2-day was used). This curve is essentially the same as curve one in Figure 1 where the surface emission flux (J_c) was equal to 8.6E-6 ug/cm2-day. This implies that the surface emission flux makes very little contribution to the overall mass transport in the soil. Curve two in Figure 4 shows the effect where the initial concentration on the mine is essentially zero (a value of 1E-20 ug/cm3 was used). This shows a significantly longer lag time and about four orders of magnitude lower steady state surface vapor flux at the end of the simulation period. Figure 4 indicates that the initial concentration is a much more important parameter than the surface flux for the mass transport of chemicals to the ground surface. Figure 5 shows the cases where the initial concentration is increased by a factor of 10 (curve 2) and a factor of 10E+5 (curve 1) over the base case (curve 3). These simulations show that the lag period remains about the same; however, the increase in steady state flux is proportional to the increase in initial concentration (C_0).

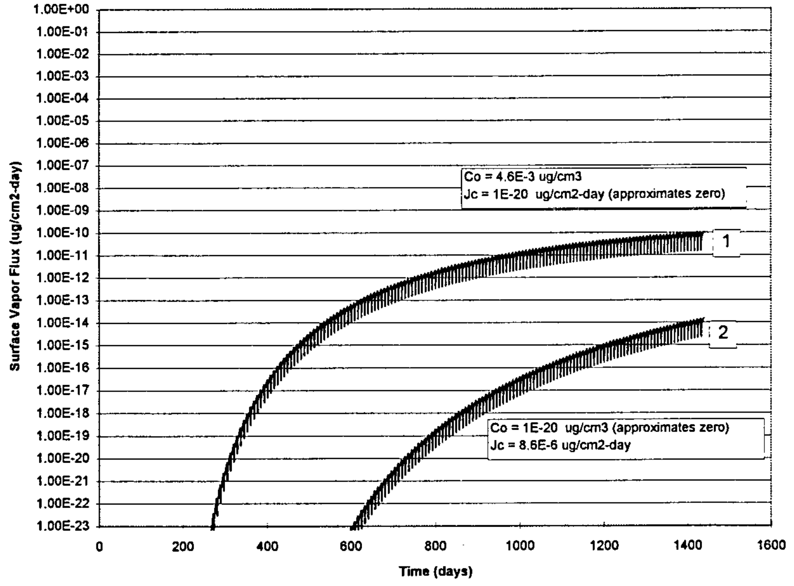

Figure 4. Effect of Source Term Variation on Surface Vapor Flux

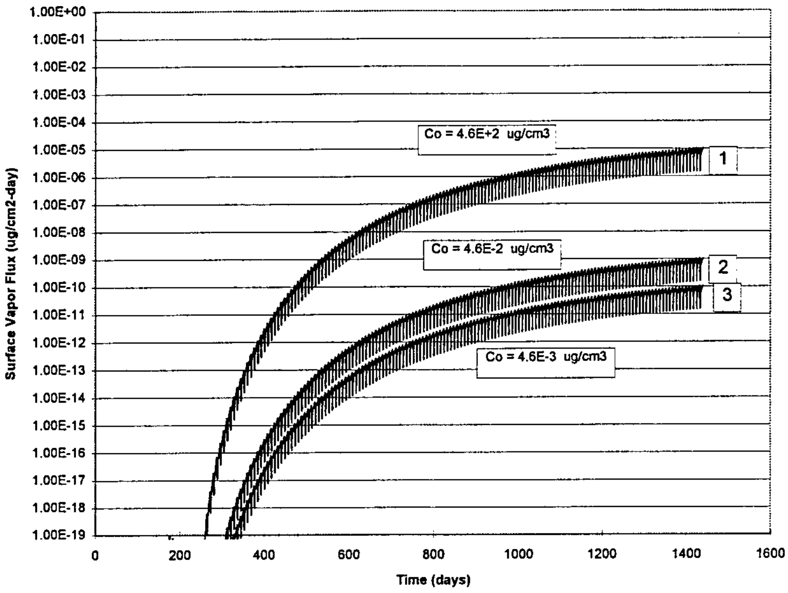

Figure 5. Effect of Source Term Variation on Surface Vapor Flux

Another parameter where there is very little data is the biochemical half life ($t_{1/2}$). There are many influences on the magnitude of this parameter and the variability is expected to be large. Simulations over ten (10) years were completed to assess the impact of decreases in the biochemical half-life from 365 days (curve 1) to 180 days (curve 2). Figure 6 shows that over the long-term, the shorter half-life will significantly decrease the steady state surface flux. Biochemical decay constants that are very large (e.g. RDX values over 30 years) appears to have minimal impact to the short term soil transport phenomena evaluated in these simulations.

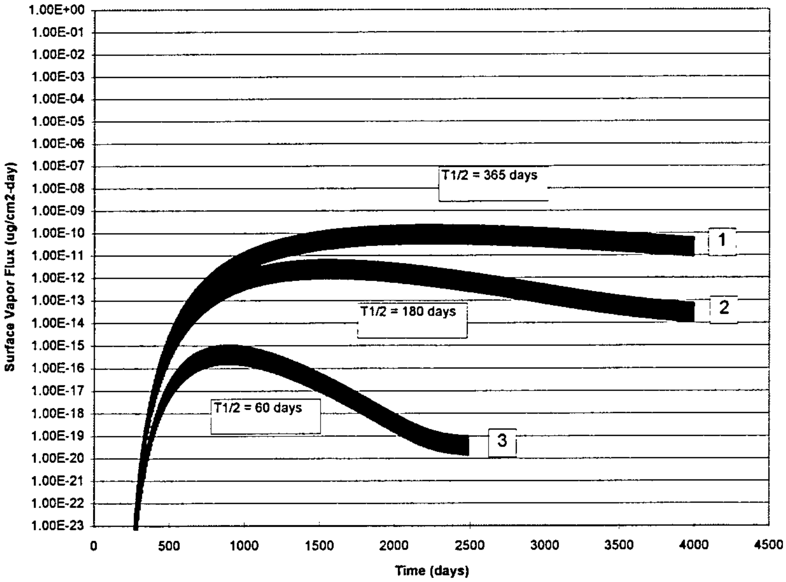

Figure 6. Effect of Half-Life on Surface Flux

Initial simulations using this approach showed that for a particular scenario, the constant moisture content value had a significant impact on the lag period (when the vapor flux reached the ground surface) and the steady state concentrations (Phelan and Webb, 1997). Higher moisture contents showed significantly shorter lag periods and greater steady state concentrations. Figure 7 shows how increasing the soil saturation from 0.5 to 0.75, decreases the lag period substantially and increases the steady state surface flux by two orders of magnitude. The constant moisture content assumption in this screening model allows for a simplified mathematical solution to a complex transport phenomena. One must recognize this assumption and not over generalize the information gleaned from these simulations. Future efforts will include the development of a numerical simulation capability that can explore the transport phenomenon of chemical signatures from buried landmines in more detail and fewer simplifying assumptions.

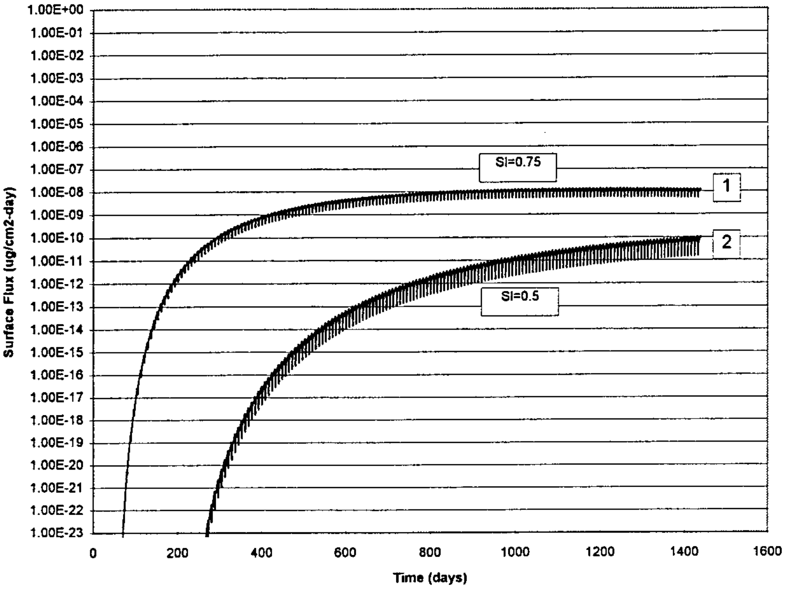

Figure 7. Effect of Soil Moisture Content on Surface Flux

In some parts of the world, minefields are located in areas that experience very distinct wet and dry climatic periods. Figure 8 shows the result of a simulation to assess the effect of a short-term continuos precipitation period followed by a short-term evaporation period. The baseline simulation (Figure 1, curve 1) was run for 1440 days followed by 1 cm/day of precipitation for 30 days which was then followed by -0.5 cm/day of evaporation for 60 days. This shows the immediate drop in the steady-state surface flux after precipitation begins. Once the evaporation period begins, there is a short lag period where the surface vapor flux stays nearly constant before rising to just above the flux before the precipitation began.

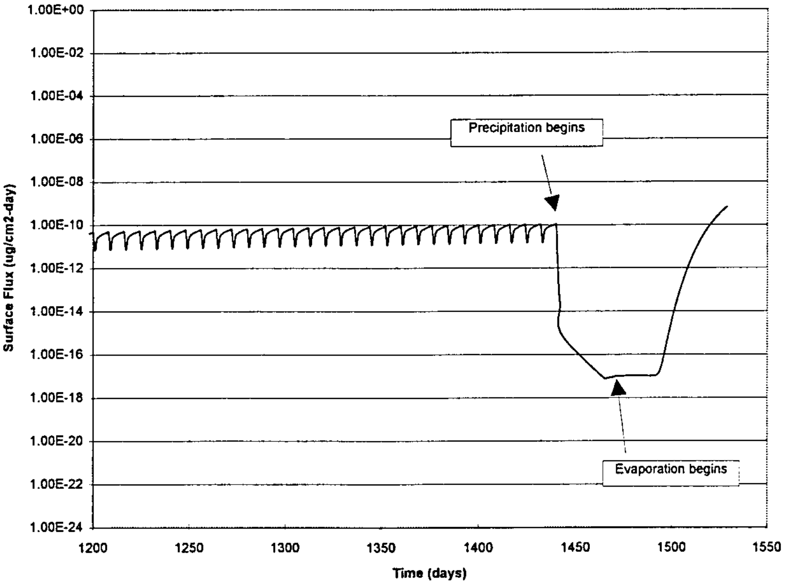

Figure 8. Effect of Continuous Precipitation Followed by Continuous Evaporation

VI. SUMMARY

The detection of buried landmines has become a significant challenge to the technical community. The challenges of locating a buried object in near surface soils have driven the need for technology from traditional geophysical sensor systems to those looking for the chemical signatures of the explosives derived from within the landmine. The transport of the chemicals found in the explosive charge through soils is a complex process involving phase changes, interactions with the soil solid phase, and biochemical reactions.

An investigation with computational simulation was initiated to explore the impacts of several of the various input parameters with a pesticide screening model adapted to the landmine chemical sensing problem. It was found that the variations in the Henry's Law constant are directly proportional to the changes seen in the steady-state surface vapor flux. Small changes in the soil water partitioning coefficient made large changes in the lag period and steady-state surface vapor flux. The initial source term and continuous source flux of chemicals from the landmines is poorly understood and expected to be highly variable. These simulations found that with the baseline case, if the continuous source flux was absent, there was no significant difference in the surface vapor flux or subsurface distribution at the end of the simulation period. This implies that the continuous source flux may be much less important than the initial surface contamination. It appears that the magnitude of the surface vapor flux is directly proportional to the amount of the initial surface contamination. The biochemical half-life is another parameter that is likely to have very different values depending on the location and climatic conditions. Simulations over ten years showed that the steady state surface vapor flux declines steadily when the biochemical half-life becomes smaller than one year. Finally, the importance of heavy precipitation (such as a monsoon season) followed by a dry season was explored. The impact of the heavy precipitation was to lower the surface flux seven orders of magnitude; however, the evaporation period that followed returned the surface flux to approximately the pre-monsoon surface flux.

VII. ACKNOWLEDGEMENT

This work was performed with internal laboratory-directed research and development funds at Sandia National Laboratories to explore applications for miniaturized chemical sensors. Follow-on work to develop a numerical model is just beginning with funds from the Strategic Environmental Research and Development Program (SERDP) for application to unexploded ordnance chemical sensing and from the Defense Advanced Research Projects Agency (DARPA) for application to buried landmines.

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