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A Numerical Investigation of Vapor Intrusion — the Dynamic Response of Contaminant Vapors to Rainfall Events

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Abstract

The U.S. government and various agencies have published guidelines for field investigation of vapor intrusion, most of which suggest soil gas sampling as an integral part of the investigation. Contaminant soil gas data are often relatively more stable than indoor air vapor concentration measurements, but meteorological conditions might influence soil gas values. Although a few field and numerical studies have considered some temporal effects on soil gas vapor transport, a full explanation of the contaminant vapor concentration response to rainfall events is not available. This manuscript seeks to demonstrate the effects on soil vapor transport during and after different rainfall events, by applying a coupled numerical model of fluid flow and vapor transport. Both a single rainfall event and seasonal rainfall events were modeled. For the single rainfall event models, the vapor response process could be divided into three steps: namely, infiltration, water redistribution, and establishment of a water lens atop the groundwater source. In the infiltration step, rainfall intensity was found to determine the speed of the wetting front and wash-out effect on the vapor. The passage of the wetting front led to an increase of the vapor concentration in both the infiltration and water redistribution steps and this effect is noted at soil probes located 1 m below the ground surface. When the mixing of groundwater with infiltrated water was not allowed, a clean water lens accumulated above the groundwater source and led to a capping effect which can reduce diffusion rates of contaminant from the source. Seasonal rainfall with short time intervals involved superposition of the individual rainfall events. This modeling results indicated that for relatively deeper soil that the infiltration wetting front could not flood, the effects were damped out in less than a month after rain; while in the long term (years), possible formation of a water lens played a larger role in determining the vapor intrusion risk. In addition, soil organic carbon retarded the transport process, and damped the contaminant concentration fluctuations.

Keywords

Vapor intrusion; rainfall; soil gas sampling; numerical modeling

1. Introduction

For over twenty years, the evaluation of vapor intrusion has been a topic of the U.S. federal and many state guidelines (DTSC, 2011; EPA, 2006; EPA, 2008; EPA, 2012a; EPA, 2004; ITRC, 2007), field investigations (DeVaull, 2007; Fischer *et al.*, 1996; Fitzpatrick and Fitzgerald, 2002; Folkes *et al.*, 2009; Garbesi *et al.*, 1993; McAlary *et al.*, 2009; Moseley and Meyer, 1992; Sanders and Hers, 2006), experimental studies (Fischer and Uchrin, 1996; McHugh *et al.*, 2012), and modeling (Abreu and Johnson, 2005; Bozkurt *et al.*, 2009;

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Johnson and Ettinger, 1991; Tillman and Weaver, 2007; Yao *et al.*, 2011). Soil gas sampling has been considered as an integral part of subsurface characterization practice, and diverse sampling methods are available (ASTM, 1992; Pacific, 2009), and a variety of protocols for soil gas sampling have been used (API, 2005; ASTM, 1992). While higher soil moisture content reduces soil permeability, thus making soil gas sampling more difficult, especially near the groundwater source, soil gas sampling is nonetheless possible under such conditions (McAlary *et al.*, 2009). In fact, interest in soil gas sampling has been in part motivated by the general interest in soil gas and soil water interaction and in part for generally characterizing contaminated groundwater itself (Thomson *et al.*, 1997). For instance, Moseley and Meyer (1992) used shallow soil gas data (4 ft) to determine the presence of subsurface contamination: the soil gas sampling was more rapid and economical than well installation and they suggested it be used to locate the groundwater contaminant plume and to place monitoring wells.

Compared to groundwater sampling, soil gas sampling is less well established, and a lack of knowledge of the influence of environmental conditions, such as rainfall or irrigation infiltration, on soil gas has been pointed out (Pacific, 2009). When comparing soil gas sampling data under various meteorological conditions, it was found for both vapor and radon measurements that there was less than 5 times soil gas contaminant concentration variation (ITRC, 2007). More recently, the U.S. EPA has considered soil gas sampling during or soon after significant rain (> 2.5 cm) in their Superfund vapor intrusion FAQs (EPA, 2012b). EPA has warned that immediately after a significant rain event (> 2.5 cm) the soil gas measurements may provide a “worst case” scenario, as water displaces soil gas and increases vapor intrusion potential in a building. On the other hand, they also cite the possibility of creation of a clean water lens over a groundwater source, and this may trap contaminant. In other words, the advice is qualitative and does not give a clear indication of the direction of effects of their dynamics. Rivett *et al.*, (2011) recently presented a very comprehensive and general review of VOC transport in the unsaturated zone, not only for vapor intrusion. The lack of knowledge and the challenges related to the understanding of transient processes in field and modeling studies were pointed out. Infiltration was a main feature of their transient review, most of which discussed the numerical models describing the transport from leaching sources or NAPL (Jang and Aral, 2007). For example, Parker (2003) simplified vapor advection resulting from both barometric pumping and groundwater level fluctuation into a dispersion term and compared that with molecular diffusion.

Rainfall infiltration effects on vapor intrusion have been suggested in several field investigations of vapor intrusion (McAlary *et al.*, 2009; Parker, 2003). In the field study by McAlary *et al.* (2009), continuous rainfall, producing wet conditions, was thought to result in about a four orders of magnitude decrease of the soil gas sampling results of 30 shallow probes and several deep probes, compared to another sampling event after extended drought. From the most recent and comprehensive modeling study of vapor intrusion (EPA, 2012b), rainfall events were mentioned as an important factor, awaiting future modeling. Tillman and Weaver (2007) studied the influence of temporal soil moisture changes and suggested that soil moisture content measurements taken somewhere other than beneath a building of interest could not be reliably used to calculate the indoor vapor concentration effects. While infiltration was not addressed in most other field and modeling studies, other transient factors, such as groundwater level fluctuations (Picone *et al.*, 2012), and groundwater concentration fluctuation (Folkes *et al.*, 2010) were discussed. Short term fluctuation might have little or no effect, but the definition of “short term” or “long term” might be site specific and will need more research to define (Folkes *et al.*, 2010).

Numerous methods are available to estimate rainwater infiltration rate. The history of modeling infiltration of rain water goes back a century to the well-known Green-Ampt

(1911) model, which assumes a piston-like wetting front and stated that the wetting front depth with time is proportional to the saturated permeability, and inversely proportional to the specific yield. Besides numerous empirical models, other widely used models include solving the highly non-linear Richards equation (Luckner *et al.*, 1989; Miller *et al.*, 1998; Van Dam and Feddes, 2000; Zhan and Ng, 2004), where many mathematical difficulties have been encountered (Miller *et al.*, 1998) particularly when applying van Genuchten (1980) relations for soil retention. While there are few analytical solutions of the Richards equation describing transient infiltration and drainage processes under limited conditions, fortunately numerical methods of solving this equation were available (Vogel and Ippisch, 2008). Further, the U.S. EPA (1998a and 1998b) has provided extensive insights into the infiltration estimation models which can be an integral part of the assessment of contaminant fate and transport.

For vapor intrusion modeling, contaminant partitions between soil water, soil gas, and soil particles, and thus, flows in both soil gas and soil water phase need to be considered. The traditional models simulating water infiltration into the unsaturated zone only consider water phase flow (Celia and Binning, 1992; Touma and Vauclin, 1986). The interaction between infiltration water and soil gas requires consideration in this case, and the potential importance of soil gas phase advection for contaminant transport has been noted (Celia and Binning, 1992). Moreover, from the U.S. EPA database (EPA, 2008), most of the contaminant vapor sources have been characterized to be groundwater sources.

When a shallow groundwater table is present, soil gas would be effectively blocked by the groundwater table which is relatively impermeable to the soil gas, and prevent from flowing downward (Adrian and Franzini, 1966). A comprehensive experimental and numerical study of the problem has been presented by Touma and Vauclin (1986), where soil moisture content profiles were measured and compared to modeling values under different infiltration rates for both open- and bounded-bottom soil columns. Celia and Binning (1992) presented a reliable model of two-phase flow under different infiltration conditions.

While the above studies contributed much to the understanding of the infiltration process, the influence of infiltration on vapor intrusion has not been completely analyzed. In this manuscript, in order to address this process, methods for understanding the infiltration of water in soil were first derived, and then applied to a vapor intrusion model involving contaminant transport. Vapor transfer was modeled over the short term and long term. Primary factors, such as rainfall intensity, duration and pattern, and soil organic content were analyzed for their effects on the soil gas contaminant concentrations at different depths and times. Guidelines for soil gas sampling under rainfall condition were developed, based on the results.

2. Methodology and Validation of the Infiltration Model

Table 1 shows the governing equations, boundary conditions, and initial conditions used in modeling both infiltration and contaminant transport. The Appendix shows definitions of all symbols used in the table. Figure 1 illustrates the modeling scenario for solving the two-phase infiltration flow equations (1) and (2), and the contaminant transport equation (3). Within the scope of this manuscript, a one-dimensional (vertical) direction was considered adequate for describing the physical phenomenon. The reference pressure datum is assumed to be at the groundwater surface ($p_w = p_g = 0$ at $x = 0$). The initial condition for soil water was chosen to be hydrostatic (Zhan and Ng, 2004). Soil elastic storage and soil gas compression were found to be negligible and therefore the two-phase flow equations were simplified to their incompressible forms; i.e. ρ_w and ρ_g were constant. The initial condition assumed for contaminant transport was the steady state concentration distribution that was

calculated given the equilibrium hydrostatic soil water distribution. Constitutive equations that were used in the calculations are shown in Table 1. A detailed estimation method for the hydraulic conductivities for two-phase flow system were presented by Luckner *et al.* (1989) where several water retention curves were compared. In the present study, the primary wetting curve was chosen here, and the constitutive equations are Equations (4) ~ (14). Equations (4) ~ (9) are the van Genuchten equations (van Genuchten, 1980). Equation (10) represents the relationship between the capillary capacity of the water and gas phases. Equation (11) represents Henry's law partitioning of the contaminant vapor between gas and water phases, whereas Equation (12) represents partitioning to soil organic carbon. Equation (13) is merely the representation of gas phase contaminant concentration in a non-dimensional form, as used in Equation (3). Equation (14) is a modified form of the Millington-Quirk (Millington and Quirk, 1961) effective diffusivity for contaminant species in soil, but including a dispersion term (Bear, 1988; Bear and Cheng, 2008).

Tetrachloroethylene (PCE) was selected as a typical chlorinated volatile organic contaminant, and the chemical properties were taken from the U.S. EPA implementation of the Johnson & Ettinger model (EPA, 2004). Local equilibrium of PCE in all phases—soil gas, soil water and soil—was assumed. While some studies (Cho *et al.*, 1993) showed the effect of considering mass transfer rate at interfaces, assumption of local equilibrium are found to be common (Griffoll and Cohen, 1996) and sufficient (Picone *et al.*, 2012), and this is shown in the constitutive Equations (11) and (12). Contaminant diffusion in soil gas and soil water was expressed by Millington-Quirk (Millington and Quirk, 1961) Equation (14), while other methods, such as (Kristensen *et al.*, 2010) give similar modeling results (Shen *et al.*, 2012; Shen *et al.*, 2011). A finite element code COMSOL was used in solving this system of equations.

In order to introduce the effects of water infiltration into vapor intrusion modeling, available infiltration experimental results and modeling methods were examined. A series of experiments of infiltration into a vertical sandy soil column was conducted by Touma and Vauclin (1986). The infiltration rate used here was 8.3 cm/h, which corresponded to the experiments under non-ponding condition. Both soil gas and soil water flows were taken into account (Equations [1] and [2] were solved simultaneously) for the bounded soil column (*i.e.* neither soil gas nor the soil water could flow out of the bottom of the column). All of the parameters, including soil properties ($\alpha^* = 4.4/\text{m}$, $M^* = 0.55$, $\theta_r = 0.0265$, $\theta_t = 0.312$), permeability relations, initial conditions and infiltration rate, were obtained from their description of the experiment. No contaminant transport was considered yet in this simulation. Figure 2 shows the simulated results for the time evolution of the soil moisture content. Soil gas and soil water pressures as a function of position and time are also shown. The soil moisture content results indicate a trend consistent with the measured data (dots). The pressures profile agrees well with the simulated result obtained by Celia and Binning (1992). As the infiltration wetting front propagates, air pressure builds up vertically.

3. Results and Discussion

Since the numerical model is seen to properly capture the main aspects of the infiltration behavior of water in soil, it can now be applied to the modeling of vapor intrusion. Regarding the contaminant vapor response to infiltration, the vapor intrusion modeling scenario is also simplified to be one-dimensional. A hypothetical soil column such as that described in the experiment in Section 2 above (and used in the validation of the infiltration model) was “expanded” from 0.935 m to 3 m deep. This model represents a groundwater contaminant source at 3 m below ground surface (as in Figure 1, $x = 0$ refers to the groundwater surface). Loamy sand is arbitrarily selected, and the soil properties are again taken from the U.S. EPA Johnson & Ettinger model spreadsheet implementation (EPA,

2004) ($\alpha^* = 3.475/\text{m}$, $M^* = 0.427$, $\theta_r = 0.049$, $\theta_t = 0.39$, and $k_s = 1.623 \times 10^{-12} [\text{m}^2]$). Two events are modeled: rainfall intensities at 2.56 mm/h (moderate rain) for 24 h duration, and 20.5 mm/h (extreme rain) for 3 h duration. It is worth noting that the two events involve the same total amount of rain water. The rainfall intensity is classified as in Tokay and Short (1996). For simplicity, rainfall intensities used here are at the non-ponding condition for loamy sand (EPA, 1998) and surface run off is not accounted for. The initial state of soil moisture distribution is chosen to be hydrostatic (Zhan and Ng, 2004), and this is calculated using Equation (4), (5), (6) and (8).

The boundary condition for water flow at the groundwater surface (bottom of the modeling domain) is important because the contaminant source is assumed to be groundwater. Although it is difficult to know the nature of any particular source, this boundary condition could be governed by two scenarios: namely, the bounded bottom scenario, when infiltrated water does not drain and builds up a water lens above the source; and the open bottom scenario, in which the infiltrated water drains and joins the groundwater whose level does not change. If the groundwater surface prevents soil gas and soil water flowing out through the bottom of the domain, assuming incompressibility, the superficial volumetric infiltration water flow equals the superficial volumetric soil gas flow out of the open ground surface. Also the results of modeling the experiment above showed that the downward flow of the water was not impeded by the upward flow of soil gas at the rates of interest. Thus, the two-phase flow equations are simplified to considering only the water flow equation. That means

$$u_w = -u_g \quad (15)$$

This agrees with the assumption made by Crifoll and Cohen (1996) and the results presented by Celia and Binning (1992) for non-ponding conditions. Therefore, when considering the last two terms in Equation (3) for contaminant advection:

$$-\frac{\partial}{\partial x}(u_g c) - \frac{\partial}{\partial x}(u_w \frac{c}{H}) = -\frac{\partial}{\partial x}[c(u_g + \frac{u_w}{H})] = -\frac{\partial}{\partial x}(cu^{eff}) \quad (16)$$

where u^{eff} is the effective advective velocity which represents:

$$u^{eff} = u_w(-1 + \frac{1}{H}) \quad (17)$$

as $H < 1$ for PCE, the effective velocity is in the same direction as water infiltration, therefore the effective advective velocity is downward. The following results are obtained from the solving governing Equation (1) and (3), since (2) does not need to be included given Equation (15).

3.1 Vapor Response to a Single Rainfall Event

Figure 3 presents the evolution of soil moisture and contaminant concentration profiles for the bounded bottom scenario of moderate (left panel) and extreme rain events (right panel). Both the fraction of organic carbon in soil f_{oc} and the evapotranspiration rate are assumed to be 0 in this calculation. Figure 3(b) and (b') show that depending upon the depth, the concentration of soil gas contaminant can change by an order of magnitude, as the process takes place. These changes will be looked at in more detail below. It needs to be recognized that the scenarios represented are intended to be illustrative for certain features, and not necessarily to represent "real-world" conditions. Regarding the evolution of the profiles, three main steps can be identified: infiltration, soil water redistribution and water lens formation. Although there is no distinct time division between these processes, to separate

them into three steps might be helpful in understanding of the processes. The three steps are analyzed in chronological order below.

The initial height of the capillary fringe is about 0.47 m, calculated from the method given by Dexter and Bird (2001). The initial contaminant vapor concentration profile is obtained based on the initial state (I.C. [a]), which assumes hydrostatic distribution of realistic soil moisture (calculated from Equation [4], [5], [6] and [8]), and this profile results in more than one order of magnitude drop in contaminant vapor concentration within the capillary fringe when the above is solved together with [3], [11], [13] and [14] at steady state. This results from the relatively large resistance to contaminant diffusion through the moist soil in the capillary fringe.

The first step can be characterized by water infiltration taking place during the rainfall. In this step, an obviously important factor is infiltration intensity. Comparing the 3 h soil moisture content lines for Figure 3(a) and (a'), it is obvious that the larger the infiltration intensity, the faster the wetting front propagates, and the more moist the soil becomes. To be more specific, Figure 4 shows the short term soil moisture and vapor concentrations at hypothetical shallow soil probes under these two rainfall events, where the infiltration steps (*i.e.* actual rain periods) are shown in the shaded areas. These results are plotted for the top of the ground surface, where greater effect has been found as the wetting front moves through. Figure 4(b) and (b') are for soils without organic carbon, while (c) and (c') are soils with 0.1 % organic carbon. The wetting front travels as a wave. The larger the infiltration intensity, the faster the wetting front propagates, and the faster contaminant concentration changes. The concentration first increases and then decreases, and its fluctuation is within 0.1~1.6 times of the initial value. Although the effective advection is downward (Equation [17]), by virtue of water dissolving progressively more PCE, the contaminant vapor concentration first increases a small amount. This is partly because the wetting front acts as a boundary or capping to the upward transport of vapor, as discussed below. After increasing for a short time, the contaminant is washed out and concentration decreases. Note that at the "probe" position closest to the surface (0.1 m depth) the vapor concentration peak occurs well before the maximum in soil moisture content is achieved. In fact, if the movement of the infiltration front is taken to be characterized by when soil moisture content reaches 0.2, then this front moves at about 0.015 m/h (left) or 0.08 m/h (right), which is also close to the rate at which the peak in vapor concentration moves downward. Note that the velocity of this front is much greater than the value of the infiltration rate, because the open area for flow becomes quite small as pores fill with water (bearing in mind that Figure 4(a) and (a') reflect this filling). The upward gas flow velocity must become higher as pores become water filled, as of course Equation (15) has been forced.

To further understand the initially observed increase in contaminant concentration, two effects need to be considered. One is an upward displacement by water of more contaminated gas from deeper in the soil, and the other is the diffusion upward against the capping by the water layer. The latter contribution can be demonstrated by variation of the effective diffusivity (Equation [14]); reduction of that diffusivity for the 2.56 mm/h, 24 hour rainfall case leads to smaller peaks, such as those that characterize the 20.5 mm/h, 3 h rainfall (shown in Figure 4 [b']). This is because in the latter extreme rainfall case, the movement of the infiltration front, relative to diffusion, is much faster than in the moderate rainfall case, and concentration cannot build up as much beneath that "capping" front. The actual peak in concentration is itself a consequence of the dissolution of PCE vapor into the water phase. As the pores become water filled with passage of the infiltration front, PCE is scrubbed out of the soil gas phase, in maintaining Henry's Law equilibrium. When no water partitioning is allowed (results not shown) the distinct peak behavior of Figure 4(b)

disappears. Note also that for the extreme rainfall in Figure 4(b'), this scrubbing leads to lower ultimate levels of gas phase contaminant, over the time frame represented in this figure (longer times will be examined below). A similar phenomenon of concentration increase and decrease was found in experimental results presented by Illangasekare *et al.* (2011).

Soil that contains organic carbon absorbs PCE and this damps out the concentration change process (see Figure 4 [c] and [c']). The fluctuation of vapor concentration never exceeds one order of magnitude of the initial value for both rainfall events even without organic carbon being present.

Immediately after the infiltration step, for the upper soil, the soil moisture content begins to decrease slowly--see Figure 4(a) and (a'). Meanwhile, the contaminant concentration (Figure 4 [b]) starts to again increase slowly after the infiltration step, as the contaminant is released from the infiltrated water.

Following the initial infiltration step, the next step is termed a water redistribution step. There is no explicit time frame for the end of this step. At a depth of 1 m below the ground surface, for the above two rainfall events, the soil moisture and contaminant vapor concentrations after 1 month are virtually identical and thus only results for 20.5 mm/h rainfall events are shown in Figure 5. As no more water is supplied at the soil surface during this stage, the downward flow rate of water greatly slows down. The wetting front is smoothed and thus, the wave type behavior becomes much less distinct. In deeper soil, the capping effect prevails over the washing effect, and contaminant concentration at 1 m below the ground surface increases slowly, though only by a small amount (about 30 %) compared to the short time effect seen in the upper soil. This agrees with recent soil gas field data and with previous radon literature (ITRC, 2007) which indicated that the soil gas contaminant concentration variation is less than a factor of 2 within a season.

Evapotranspiration is added and included in Figure 5. The evapotranspiration rate is chosen to be the same as used by Tillman and Weaver (2007), 58.42 cm/year. Other more complicated expressions for evapotranspiration rates are available (Griffoll and Cohen 1996). During the infiltration step, the water upward flux by evapotranspiration is small compared to the infiltration rate ($0.0667 \text{ mm/h} \ll 20.5 \text{ mm/h}$). At longer times, during the water redistribution step, the evapotranspiration rate decreases the amount of water available for distribution and accumulation. Considering mass conservation of water, all the infiltrated water can be lost to evapotranspiration after 39 days, at which point, the initial hydrostatic water distribution is recovered. In the presence of evapotranspiration, the influence of rainfall on vapor intrusion is shortened to include only the infiltration and water distribution steps; no water lens would accumulate above the groundwater table.

When evapotranspiration is set to zero, in a single rainfall event, the third and final step involves the formation of a water lens atop the groundwater and ends in a final steady state which is a new steady state, if that lens is not assumed to be absorbed into the original groundwater. The final groundwater and capillary zone height were shown in Figure 3. However, the contaminant vapor concentration is still far from the steady state profile at 1 year. This implies that the contaminant vapor transport is approaching a new steady state over the course of years. Of course, it is unrealistic that there would be no further rain (or evapotranspiration), so this case is clearly hypothetical. But if the infiltrated water reaches the source and forms a water lens in this bounded bottom scenario, such a water lens behaves as a large resistance to contaminant vapor transport, and this would cause the vapor concentration to drop between the first several months and 1 year. This is shown in Figure 6(b) and (b'). The new steady state water distribution again reaches hydrostatic. The only

difference between the initial steady state and the final steady state is that the groundwater level increased. It is important to note that the shape of the final steady state soil water retention curve remains the same. From mass conservation, based on the water infiltrated, the final groundwater height increase Δh can be calculated by:

$$q_{\text{rain}} t_{\text{rain}} = \int_{3-\Delta h}^3 \theta_w(x, t=\infty) dx + \theta_t \Delta h - \int_0^3 \theta_w(x, t=0) dx \quad (18)$$

The total rainfall amounts, or the accumulated infiltration were 0.062 m (2.4 inches) for both of these two rainfall events, while the calculated Δh is 0.22 m which is larger because of the fact that the water must fill in available porosity.

A simplified water lens model is simulated, assuming an initial condition corresponding to the same water height as achieved in the infiltration model at long times. The simplified model solves Equation (3), without solving Equations (1) or (2), as neither the capping effect nor washout effect are considered. It determines the magnitudes of contaminant concentration due to the presence of a clean water lens atop the groundwater source. The result of this simplified model is shown at the right panel of Figure 6, which is almost the same as the solution in the left panel which is that for the actual 20.5 mm/h rainfall events. Even though the infiltrated water (left case) dissolves some contaminant as it percolates towards the groundwater source, the water is still “clean” relative to the groundwater source, which is why the results of the two models are so similar to one another.

In either case while the vapor concentration steadily increases at a probe 2.8 m deep (0.2 m above the source), vapor concentrations at higher probes first decrease and then increase. Diffusion through the water lens in both models is the controlling, or rate limiting step, as PCE diffusivity in pure water is only $8.2 \times 10^{-9} \text{ m}^2/\text{s}$. The sorption to the organic carbon increases the time scale, comparing Figure 6(a) to (b), and (a') to (b'). The final states are the same for both models. In other words, addition of a thin clean water lens atop the original contaminant groundwater will have some effects of decreasing soil gas contaminant vapor concentrations, but the effects would be observed only over a very long time scale as compared to the initial infiltration step. A permanent change to groundwater contaminant concentration profile, as this illustration depicts, can have a dramatic influence on the potential for vapor intrusion. Keep in mind that only a 0.22 m thick lens was added atop a source 3 m beneath the soil surface. While it is of course not realistic to consider only a single rainfall event over the course of many years, the more significant aspect of the present analysis is that concentration profiles, near the top of the water table, can confound analysis of a situation in which the underlying bulk groundwater is not observed to change very much in contaminant concentration. This of course implies that where a monitoring well draws water can make a very large difference in predicted hazard posed by the contaminant in the water.

For an open bottom scenario, in which the groundwater (the source) completely mixes with infiltration water, and water drainage keeps the groundwater level the same, the contaminant concentration profile (results not shown) has the same shape as the bounded bottom scenarios discussed above, but the original steady state is recovered. The dynamics of any long-term process associated with infiltration affected water table levels are slow, and if the original water table height is recovered, will depend on how fast this recovery happens.

Vapor Response to Seasonal Rainfall Events—After discussion of the cases with one simple rainfall event, attention is turned to a “wet site” that is subject to seasonal rainfall

characterized by a long series of small rainfall events. The modeling results are shown in Figure 7. The hypothetical rainfall intensity is assumed to be described by:

$$q_{rain}=0.26[\text{mm/h}] \cdot \left[-\frac{1}{2}\cos\left(\frac{2\pi}{24[h]} \cdot t\right)+\frac{1}{2} \right] \quad (\text{iv})$$

which has a period of 1 day. The maximum rainfall of 0.26 mm/h can be classified as a very light rain. There are assumed to be 30 rainfall events over 30 days, then these stop for the rest of the year. Evapotranspiration is again set to be zero in order to see the maximum influence of rainfall. These events are modeled for the bounded bottom scenario. The accumulated rainfall is 0.094 m for 30 days; and it forms a water lens with a maximum thickness of about 0.33 m above the source at about 1 year after the events. The buildup of this lens takes place at later time than the single rainfall cases because water is added more slowly. From Figure 7, superposition of rainfall events is obvious, as both soil moisture and vapor concentration in the upper soil changes continuously under a continuous rainfall over 30 days, with small fluctuation every day. The small fluctuation in vapor concentration results from the same effects as described before for the single rainfall events, but here reflects the gradual buildup due to cycling of rain. However, in soil deeper than 1 m below the ground surface, this kind of small soil moisture fluctuation is hardly visible (Figure 7 [c]). The wetting front reaches the 1.5 m probe in a month. Therefore, in Figure 7(c), soil moisture at probes 1 and 1.5 m deep show 30 % increase at 1 month. The capping and washing effect on contaminant concentration are also obvious (Figure 7 [d]). It should be noted that the roughly one order of magnitude decrease of concentration after infiltration results not only from the washing effect, but also from the water lens effect as discussed for the single rainfall cases.

Conclusion

From the above modeling results, the dynamic processes of contaminant vapor response to both single rain event and seasonal rain events can be observed to be quite complex. The influence of infiltration on vapor transport is complicated, as it not only includes the change of the advection rate of the vapor, but also the change of soil moisture content and its diffusivity. Regarding the single rainfall events, the effect of infiltration normally can be divided into three steps, and the time scale of the three steps differs, *i.e.* hours for the infiltration step, weeks for the water redistribution step, and years for the final water lens formation step if there is a significant change in groundwater level on this time scale (though this is not necessarily realistic). Different factors dominate different steps. In the infiltration step, the rainfall intensity and duration matter. The depth of the wetting front can be predicted by using a simple one dimensional infiltration model with known soil types and rainfall. During the infiltration step, probes in the upper soil that is flooded by the wetting front show increase and then decrease in vapor concentration. Since these are not commonly sampled locations in soil vapor probing, this result is mainly of academic interest. The rainfall effect on the lower soil vapor concentration is much more modest. The depth of soil gas probes can be optimized by using such a model assuming that common rainfall patterns are known. For example, at a relatively dry site with rare rainfall events, soil gas probes at 1 m below the ground surface experience minimal the change of soil moisture and the change of the vapor concentration right after rainfall is also minimal, provided that there is no flooding.

Regarding a site with a wet season characterized by continuous light rain, the superposition of individual rainfall effects is apparent. As the infiltration rate relies on the initial condition of the soil moisture distribution, knowledge of the initial soil moisture profile is necessary. Since the water distribution step requires the order of weeks to occur, the effects of rainfall

events with intervals of weeks will build on the previous events. With continuous rainfall of a month duration, the wetting front propagates to more than 1 m under the ground surface, and therefore, probes at this depth will show obvious change over month timescales.

When evapotranspiration is considered, infiltrated water from the single rainfall event can be evapotranspire on the timescale of days to tens of days, and then the original hydrostatic water distribution in the soil is recovered. If, however, evapotranspiration is small or otherwise ignored, the infiltrated rain water can accumulate atop the groundwater table and forms a clean water lens. This new water lens can act as a large resistance to vapor diffusion and therefore greatly slow diffusion of vapor from the original source. The time for a new 0.2 m thick water lens to reach its final concentration can be on the order of years. This clean water absorbs and greatly slows diffusion of contaminant, and the concentration at soil probes decreases until this water is finally saturated. The vapor transport rate out of the soil decreases as the source now is under a saturated lens that acts as an extra diffusion barrier. Again, this result is important to bear in mind regardless of how such a clean water layer might come to be. If there is reason to suspect that there might be development of a significant contaminant concentration gradient near the groundwater surface, samples from deeper groundwater monitoring wells might offer a misleading picture of vapor intrusion potential (source strength). A lower source strength, due to existence of a relatively clean water lens would lead to a decrease of the vapor exposure risk on a site. Measurement of groundwater levels, groundwater velocity, and sampling of the soil water/soil gas above the source are crucial. High groundwater velocity enhances mechanical dispersion that helps better mix the groundwater with the infiltrated water and then decreases the water lens capping effect.

Finally, the presence of organic carbon in the soil will retard the process of contaminant transport, and influence the observed dynamics. The effects are not very large, for a carbon content of 0.1% in the soil.

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Appendix. Definition of Symbols

c	contaminant concentration normalized by concentration in the gas phase
c_i	contaminant concentration in the i phase [ug/m ³]
C_{ci}	capillary capacity of fluid i [1/m]
D_i	the molecular diffusivity of contaminant in the water or gas phase [m ² /s].
f_{oc}	fraction of organic carbon in soil (assumed to be 0.001)
g	the acceleration due to gravity [m/s ²] acting in the vertical direction z
H	the Henry's law constant [dimensionless]
H_c	capillary pressure head [m], assumed to be positive
i	denote the phase, g : soil gas; w : soil water; $soil$: soil
k_{oc}	organic carbon partition coefficient [m ³ /kg]
$k_{r,i}$	the relative water or gas permeability compared to the fluid saturated permeability
k_s	the intrinsic saturated permeability [m ²]
l	the thickness of soil from source to the ground surface
p_c	capillary pressure [Pa]
p_i	the fluid pressure [Pa]
q_{rain}	the rainfall infiltration intensity [m/s]
q_{evapo}	the evapotranspiration rate [m/s]
Se_w	effective water saturation
u_i	the superficial velocity of the fluid i [m/s]
u^{eff}	the effective contaminant vapor advective velocity [m/s]
x	vertical axis; $x = 0$ at the source

α^* [1/m] and M^*	van Genuchten parameters
θ_g	volumetric soil gas content
θ_r	residual soil moisture content
θ_t	the total soil porosity which is either water (θ_w) or gas (θ_g) filled
θ_w	volumetric soil moisture content
μ_i	the fluid viscosity [kg/m/s]
ξ	the longitudinal dispersivity or simply dispersivity [m] (selected to be 0.005 m)
ρ_i	the fluid density [kg/m ³]
ρ_b	soil bulk density [kg/m ³]

Figure 1.

The modeling scenario. Left panel: water-soil gas flow; right panel: contaminant transport.

The equations and boundary conditions are in Table 1

Figure 2.

The modeled vertical soil moisture (left Figure), soil water and soil gas pressures profiles (right Figure), all of which are plotted at 6 min intervals. Dots: experimental measurements from Touma and Vauclin (1986). The dashed arrows show the evolution of time. The qualitative shape of the soil column corresponding to the simulation scenario is shown on the left

Figure 3.

Modeling results for soil moisture (a) and (a') and vapor concentrations (b) and (b') at times 0, 3 h, 1 day, 1 week, 1 month, 1 year and final steady state. Left panel assumes rainfall intensity of 2.56 mm/h, duration: 24 h; right panel assumes rainfall intensity of 20.5 mm/h, duration: 3 h. Initial and final heights of groundwater and capillary fringe are shown by horizontal dashed lines. These simulations assume $f_{oc} = 0$ and $q_{evapo} = 0$. Again, these are 1-D simulations for a hypothetical soil column as illustrated above

Figure 4.

Short term soil moisture content (a) and (a'), and contaminant concentration (b), (b'), (c) and (c'), at 0.1, 0.2 and 0.3 m deep. Left panel assumes rainfall intensity: 2.56 mm/h, duration: 24 h; Right panel assumes rainfall intensity: 20.5 mm/h, duration: 3 h. In (b) and (b') $f_{oc} = 0$; in (c) and (c') $f_{oc} = 0.001$. All simulations assume $q_{evapo} = 0$

Figure 5. Soil moisture content (a) and (a') and contaminant concentration (b) and (b'), at probes at 1, 1.5 and 2 m deep at 1 month after rainfall events of intensity: 20.5 mm/h, duration: 3 h, and $f_{oc} = 0$. Results (a) and (b) are at $q_{evapo} = 0$, while (a') and (b') are for non-zero evapotranspiration, $q_{evapo} = 0.0667$ mm/h

Figure 6.

Vapor concentrations at the soil gas probes located 0.2, 0.5, 1, 1.5, 2, 2.5 and 2.8 m deep at long term conditions when $q_{evapo}=0$. Left panel: rainfall 20.5 mm/h for 3 h duration; right panel: simplified water lens model. In (a) and (a') $f_{oc}=0$; in (b) and (b') $f_{oc}=0.001$. The initial groundwater levels are indicated

Figure 7. Soil moisture content (a) and (c), and vapor concentration (b) and (d) under seasonal rainfall conditions; $f_{oc} = 0$. Left panel: results for probes at 0.1, 0.2, and 0.3 m deep. Right panel: results for probes at 1, 1.5 and 2 m deep

Table 1

Equations and parameters

Governing equations

$$\text{Soil water flow} \quad \frac{C_{c,w}}{\rho_w g} \frac{p_c}{t} = -\frac{1}{x} \left[\frac{k_s k_{r,w}}{\mu_w} \left(-\frac{p_w}{x} + \rho_w g \right) \right] \quad (1)$$

$$\text{Soil gas flow} \quad \frac{C_{c,g}}{\rho_w g} \frac{p_c}{t} = -\frac{1}{x} \left[\frac{k_s k_{r,g}}{\mu_g} \left(-\frac{p_g}{x} + \rho_g g \right) \right] \quad (2)$$

$$\text{Contaminant transport} \quad \left(\theta_g + \frac{\theta_w}{H} + \frac{\rho_b k_{oc}^f}{H} \right) \frac{c}{t} + c \left(1 - \frac{1}{H} \right) \frac{\theta_g}{t} = -\frac{1}{x} (D^{eff} \frac{c}{x}) - \frac{1}{x} (u_g c) - \frac{1}{x} (u_w \frac{c}{H}) \quad (3)$$

Boundary conditions (as shown in Figure 1)

$$u_w(x = L, t) = -\frac{k_s k_{r,w}}{\mu_w} \left(-\frac{p_w}{x} + \rho_w g \right) = q_{rain} - q_{evapo} \quad \text{B.C.(a)}$$

$$u_w(x = 0, t) = -\frac{k_s k_{r,w}}{\mu_w} \left(-\frac{p_w}{x} + \rho_w g \right) = 0 \quad \text{B.C.(b)}$$

$$p_g(x = L, t) = -\rho_g g L \quad \text{B.C.(c)}$$

$$u_g(x = 0, t) = -\frac{k_s k_{r,g}}{\mu_g} \left(-\frac{p_g}{x} + \rho_g g \right) = 0 \quad \text{B.C.(d)}$$

$$c(x = L, t) = 0 \quad \text{B.C.(e)}$$

$$c(x = 0, t) = 1 \quad \text{B.C.(f)}$$

Initial conditions

$$p_w(x, t = 0) = -\rho_w g x \quad (\text{hydrostatic}) \quad \text{I.C.(a)}$$

$$p_g(x, t = 0) = -\rho_g g x \quad \text{I.C.(b)}$$

$$c(x, t = 0) = \text{steady state conc. distribution at initial hydrostatic condition} \quad \text{I.C.(c)}$$

Constitutive equations

$$H_c = \frac{p_c}{\rho_w g} = \frac{p_g - p_w}{\rho_w g} \quad (4)$$

$$Se_w = [1 + (a^* H_c)^{1/(1-M^*)}]^{-M^*} \quad (5)$$

$$\theta_w = \theta_{r,w} + Se_w(\theta_t - \theta_{r,w}) \quad (6)$$

$$k_{r,w} = Se_w^{1/2} [1 - (1 - Se_w^{1/M^*})^{M^*}]^2 \quad (7)$$

$$\theta_g = \theta_t - \theta_w \quad (8)$$

$$k_{r,g} = (1 - Se_w)^{1/3} (1 - Se_w^{1/M^*})^{2M^*} \quad (9)$$

$$C_{c,w} - C_{c,g} = \frac{\theta_w}{H_c} \quad (10)$$

$$c_g = H c_w \quad (11)$$

$$c_{soil} = \rho_b^k \rho_{oc}^f \rho_{oc} c_w \quad (12)$$

$$c = c_g / c_{\text{source vapor}} \quad (13)$$

$$D^{eff} = D_g \frac{\theta_g^{10/3}}{\theta_t^2} + \frac{D_w}{H} \frac{\theta_w^{10/3}}{\theta_t^2} + \xi(|u_g| + |u_w|) \quad (14)$$