Migration of VOC Plume in the Subsurface Domain at the Y-12 National Security Site

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During the production of thermonuclear fusion weapons at the Y-12 National Security Complex (Y-12 NSC) in Oak Ridge, Tennessee, between 1950 and 1963, the regional environment was extensively contaminated by volatile organic compounds (VOCs). Old Salvage Yard (OSY) on the western side of the site has been characterized as the major source of VOCs. In order to analyze the long-term fate and transport of chlorinated VOC sources, an integrated surface and subsurface flow and transport model was developed for the Y-12 NSC using the hydrodynamic and transport numerical package MIKE-SHE. The model was developed considering the recent hydrogeological investigations on preferential flow and transport pathways at the site. The model was calibrated using the recorded groundwater flow and water-quality data. The modeling simulated migration of the VOC plume for the next 100 years. Considering a range of hydrogeological and transport parameters, uncertainty of the results is discussed. The modeling predicted that tetrachloroethene, trichloroethene, and 1,2-dichloroethene may exceed human health–related risk levels for the next 10 to 20 years. However, the contamination is unlikely to migrate to surface water under the current hydrogeological conditions and will decay below acceptable risk levels within approximately 20 years. © 2013 Wiley Periodicals, Inc.

INTRODUCTION

The Y-12 National Security Complex (Y-12 NSC) was built in 1943 in Oak Ridge, Tennessee, as part of the classified Manhattan Project for the production of thermonuclear fusion weapons. Old Salvage Yard (OSY) on the western side of the site has been identified as the major source of volatile organic compounds (VOCs) at the site. During the 1950s and early 1960s, the surrounding environment was contaminated with considerable amounts of volatile organic VOCs as well as metals, nitrates, and radionuclides. The main release mechanism was spills and leaks from the process buildings to the foundations, soil, groundwater, and drainage systems. The process buildings were shut down in 1963; however, spills and leaks continued from the surface soil to deeper layers, groundwater, and the drainage system, not only from former process buildings, but also from disposal facilities. Although remedial activities, initiated in the mid-1980s, removed the highly contaminated surface soil layers and displaced most of the waste storage facilities from the site, field surveys show that much of the lost or unaccounted leaks of VOCs are still

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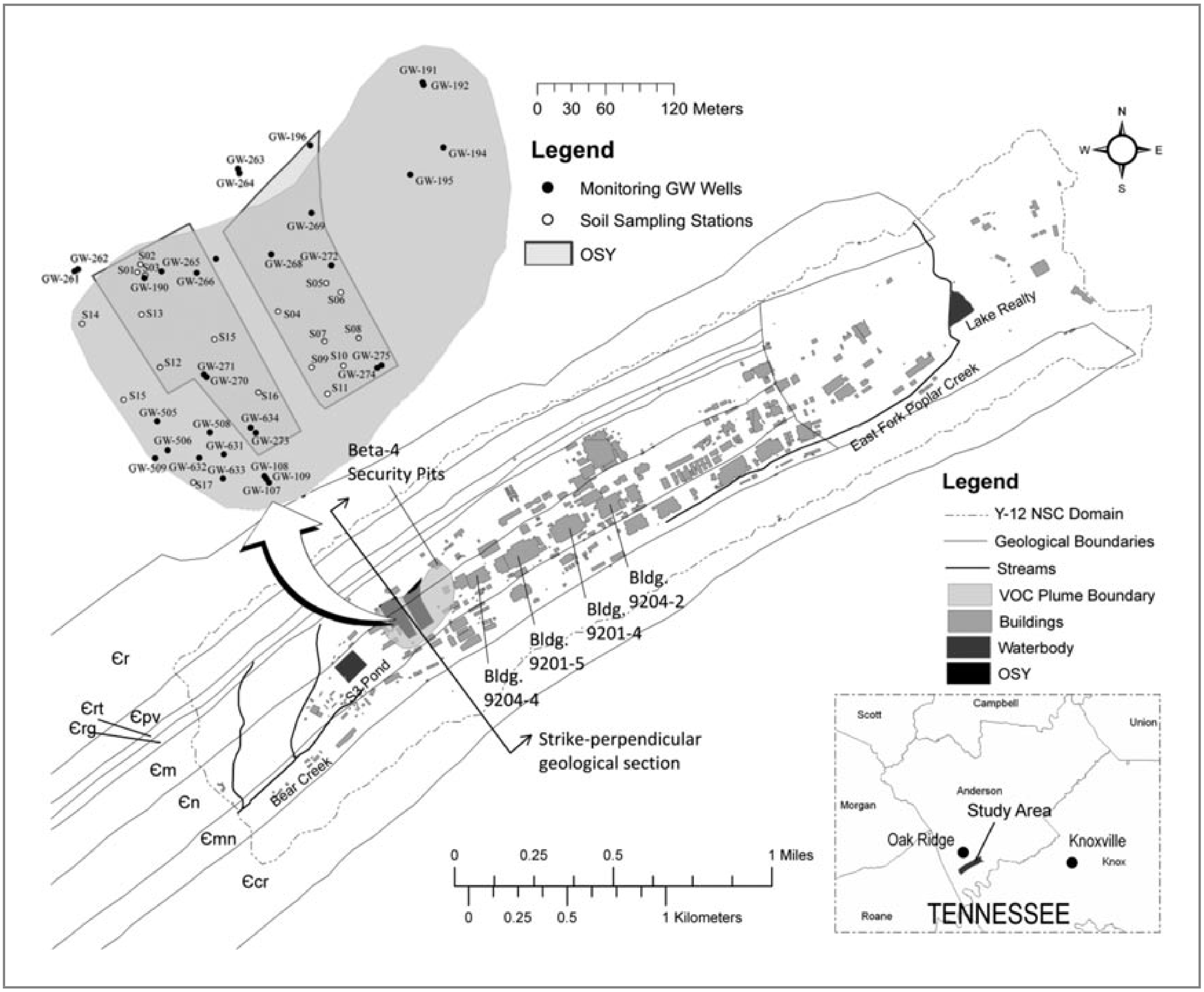

Exhibit 1. Location of the site, VOC plume, and soil sampling stations and groundwater monitoring wells

Єr = Rome Formation, Єpv = Pumpkin Valley Shale, Єrt = Rutledge Limestone, Єrg = Rogersville Shale, Єm = Maryville Limestone, Єn = Nolichucky Shale, Єmn = Maynardville Limestone, and Єcr = Copper Ridge Dolomite.

present in deeper soil layers, bedrock, and groundwater under the former process buildings and disposal areas. More than 80 percent of the chlorinated VOCs, including tetrachloroethene (PCE), trichloroethene (TCE), and 1,2-dichloroethene (1,2-DCE), both *cis* and *trans* isomers, are present in soil and groundwater in the vicinity of OSY due to the historical placement of the waste storage facilities at this area (US Department of Energy [US DOE], 1998). The VOC-affected area inside the Y-12 NSC is shown in Exhibit 1. Matrix diffusion and leaching of VOCs from soil constitute a source to groundwater that continues to spread the contaminants spatially.

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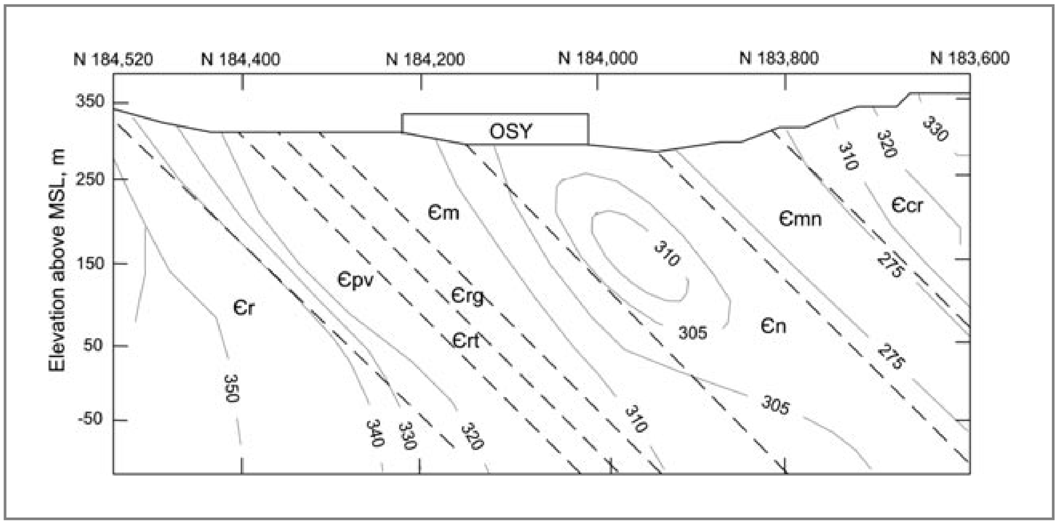

Exhibit 2. Strike-perpendicular hydrogeologic cross-section underneath OSY as shown in Exhibit 1

Thin dark lines are estimated hydraulic head in meters above MSL after Dreier et al. (1993). The coordinate system is North American Datum 1983 State Plain of Tennessee. Geological formations are divided by dashed lines. Geological formations are the same as defined in Exhibit 1.

OSY is immediately underlain by the bedrock of Maryville Limestone to the north and the Nolichucky Shale to the south (Exhibit 1), which is weathered to an overburden clayey fill and residual soil with fractures and macropores immediately below the ground surface (US DOE, 1998). The flow and transport in this region is governed by matrix diffusion and flow through macrospores (Jardine et al., 1999; Reedy, Jardine, Wilson, & Selim, 1996). The groundwater flow and transport of various contaminants (mostly nitrate) at the neighboring site of S3 Pond have been investigated through a number of numerical studies using the equivalent porous media (EPM) model (e.g., McKay, Stafford, & Toran, 1997), a dual porosity model (e.g., Stafford, Toran, & McKay, 1998), multiple porosity models (e.g., Wilson, Jardine, & Gwo, 1992), and immobile and mobile phases in dual porosity concept (e.g., Luo et al., 2005).

Underneath the top layer of weathered clay-rich saprolite and in the Nolichucky Shale formations, the groundwater flow and transport is mainly horizontal along the strike not only immediately below the saprolite layer but also throughout the Nolichucky Shale bedrock to much greater depths. This is confirmed through groundwater pressure tests using core holes during the 1990s (Dreier, Early, & King, 1993), as shown in Exhibit 2.

Data for groundwater samples obtained from OSY and Beta-4 security pits (see Exhibit 1) suggest the migration of VOC groundwater plume eastward along the strike in the Nolichucky Shale through the fractured bedrock in the last 20 years. Currently, the VOC plume in shallow groundwater (less than 20 meters deep) covers the entire OSY area. Previous studies were mainly focused on the subsurface flow and transport of nitrate in neighboring sites. Considering the recent hydrogeological surveys, the main purpose of the present study was to construct a simple numerical model with the minimum degrees of freedom capable of estimating the recent groundwater flow and water-quality observations as well as reasonably predicting the future long-term plume migration.

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The hydrogeological characteristics of the site obtained during the field monitoring surveys over the last several decades were combined to model the migration of the chlorinated VOC plume in both the shallow and intermediate subsurface layers underneath OSY. Chlorinated VOCs, due to relatively high solubility limits and low partitioning coefficients, k_d, dissolved into groundwater in high concentrations and migrated throughout the subsurface domain much faster than other contaminants, such as mercury. Therefore, the relevance and applications of this study extend beyond this particular site.

SITE CHARACTERISTICS

Geographical and Hydroclimatic Conditions

OSY is located on the western side of the Y-12 NSC, shared by Anderson and Roane Counties in Tennessee. The watershed is bound by Pine Ridge on the north and Chestnut Ridge on the south. The majority of the surface water runoff at the site is collected by the process buildings' drainage systems and eventually discharges to East Fork Poplar Creek (EFPC) through Outfall 200. A very small portion of OSY surface water on the west side of the site discharges toward S3 Pond and is intercepted by tributaries to Bear Creek (for comprehensive surface water flow and hydrology of the site, see Malek-Mohammadi, Tachiev, Cabrejo, & Lawrence, 2012). The site is underlain by the Nolichucky Shale formation on the south and the Maryville Limestone formation on the north. Groundwater is mainly recharged by precipitation, which varies annually and seasonally. Approximately 50 percent of the annual precipitation is lost to evapotranspiration, almost 40 percent runs off the ground surface, and only 10 percent recharges the groundwater (Solomon, Moore, Toran, Dreier, & McMaster, 1992). The groundwater table is higher on ridges and decreases toward the valley. Principal groundwater flow is from ridges toward the valley and along the strike toward the east, and a very small portion along the strike toward the west. The Nolichucky Shale exhibits a high hydraulic conductivity zone compared to the Maryville Limestone; therefore, groundwater flow and contaminants predominantly migrate along the strike while the lateral transport is limited.

Hydrogeology and VOC Transport

Based on pumping tests, tracer tests, and geological studies (Cook et al., 1996; Moore, 1997; Solomon et al., 1992; Van der Hoven, Solomon, & Moline, 2005), the subsurface flow system underneath OSY is characterized by three distinguished zones: (1) storm flow zone in the unconsolidated clayey weathered soil material located at the top 3 to 4 meters of the soil profile; (2) the transition zone in the Nolichucky Shale formation, located immediately underneath the saprolite layer and characterized by high fracture density, high hydraulic conductivity along the strike, and high permeability (Jardine et al., 1999); and (3) the saturated zone of fresh bedrock. Only 10 percent of the groundwater flow occurs in the bedrock zone, with about 8 percent in the shallow water table interval and 2 percent in intermediate and deep intervals (Solomon et al., 1992).

The primary release of VOCs to groundwater occurs through leaching from soils. Infiltration and groundwater recharge during heavy rainfall events dissolves the

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contaminants in surface soil layers and, through gravity, drives the contaminated water to deeper soil layers eventually reaching the groundwater table. In weathered saprolite, matrix diffusion is the primary release and transport mechanism. In this clay-rich unconsolidated residual soil layer, VOCs have accumulated throughout the years. Contaminants are diffused from the soil matrix to groundwater through fractures and macrospores. This zone is called the storm flow zone, as contaminants are flushed during storms through this unconsolidated layer. In the transition zone, the major transport mechanism is advection with groundwater flow along the strike through this high-hydraulic-conductivity and high-permeability zone. High VOC concentrations have been detected in samples from this zone (Chen et al., 2006; Luo et al., 2005). The shallow groundwater flow in the saprolite and transition zones will eventually release to buried tributaries, utility lines, and operating building sumps (Buildings 9204-4, 9201-5, and 9201-4 as shown in Exhibit 1, east side of OSY and along the geological strike) or strike-perpendicular fractures, which divert the flow downgradient toward East Fork Poplar Creek in the south.

NUMERICAL MODEL

An integrated overland and subsurface flow and transport model was developed using the MIKE-SHE numerical package (Refsgaard & Storm, 1995) to simulate the fate and transport of VOC plumes and soil point sources of PCE, TCE, and 1,2-DCE in the vicinity of OSY. The input parameters, such as precipitation, surface topography, land use, vegetation, and permeability, were extracted from the US Geological Survey, Tennessee Department of Environment Conservation, Oak Ridge Environmental Information System (OREIS) database, and published reports by the US DOE and Oak Ridge National Laboratory. The development of the overland flow model is explained further in Malek-Mohammadi et al. (2012).

For the subsurface flow modeling, the three zones defined earlier, including the unconsolidated top soil layer (saprolite), transition zone, and bedrock zone, were considered. Saprolite is represented by three layers with a total thickness of 4 m. The transition zone is modeled assuming two layers with a total thickness of 3 m. The bedrock is modeled considering three layers of 10, 30, and 50 m each to a depth of 97.1 m.

Major assumptions of the numerical modeling are: (1) homogeneity of the initial plume throughout the modeled depth due to scarcity of the field data, (2) neglecting the presence of VOCs in the unsaturated zone (storm flow zone), (3) modeling the flow through fractures using macrospores and secondary porosity option, and (4) neglecting the chemical reactions between VOCs and other contaminants present at the site, such as metals.

Parameters

Throughout the years, a number of pumping and tracer tests have been performed at the site to estimate the hydrogeologic parameters. The range of estimated parameters is extremely diverse and, in some cases, irrelevant. US DOE (1998) reported the hydraulic conductivity values range from 0.01 to 1 m/d for saprolite and from 0.1 to 2 m/d for the

Exhibit 3. Chlorinated VOCs' transport parameters

	Unit	PCE	TCE	1,2-DCE
Molecular weight	g/mol	165.8	131.4	96.9
Solubility	Mg/L	150	1,100	800
Biodegradation rate[1]	yr^{-1}	0.15	0.15	0.09
Half-life[2]	yr	4.5	4.5	7.9
k_d [3]	L/kg	0.795	0.300	0.233
K_{oc} [1]	L/kg	265.0	94.0	77.5

[1] US DOE (1998).

[2] Half-lives for VOCs are less than two years, and the values shown in the table are considered as upper bounds used by the US DOE.

[3] K_d calculated based on $K_{oc} \times f_{oc}$, which was extracted from US DOE (1998).

bedrock. Numerical models developed by the US DOE have used different calibrated values from 0.02 to 3 m/d for the preferential pathway along the strike in the transition zone. Recent flowmeter tests show that the conductivity is generally low in the saprolite and high in the transition zone, and low in bedrock (Chen et al., 2006; Luo et al., 2005). Luo et al. (2005) suggested the conductivity of 2 to 4 m/d for the transition zone. In the present model, the calibrated values of 0.3 m/d for the saprolite, 2 m/d for the along-strike flow in transition zone, and 0.4 m/d for the bedrock in the Nolichucky Shale were used.

The porosity of the saprolite layer has been reported to be between 0.2 and 0.6 (Van der Hoven et al., 2005). The average values of 0.5 for the first layer, 0.4 for the second layer, and 0.3 for the lowest layer of saprolite were used in the present model. For the secondary porosity of macrospores, 0.1 was used, as suggested by Wilson et al. (1992). For the bedrock, the effective porosity of 0.05 was used in the calibrated model, which is relatively close to the range of 0.002 to 0.04 estimated by the US DOE through tracer studies. Longitudinal and traverse dispersivity values of 1.0 m were calibrated for the present model, which is in close agreement with the 0.8 m used by McKay et al. (1997). Transport parameters for PCE, TCE, and 1,2-DCE used in the calibrated model are summarized in Exhibit 3.

VOC Plume and Point Sources

Groundwater and soil VOC data at the vicinity of OSY were extracted from the OREIS database (OREIS, 2011). The soil sampling locations and groundwater monitoring wells are shown in Exhibit 1. Groundwater data from 1985 to 1995 were used for source estimations, and recent data (after 1995) were used to validate the numerical simulations. The hypothetical plume was estimated by geospatial analysis on groundwater and soil pollution data using ARC-GIS spatial interpolation utility. The inverse distance method was used to create the continuous surface of the plume. The boundary of the plume was

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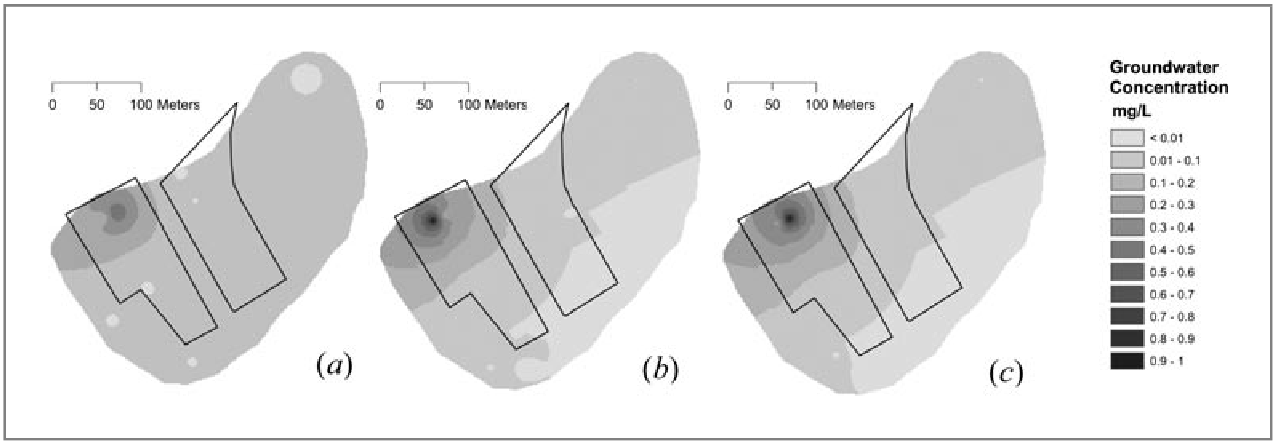

Exhibit 4. VOC source plumes: (a) PCE, (b) 1,2-DCE, and (c) TCE

estimated based on literature discussions and spatial variations of concentrations in groundwater and soil. The VOC plume boundary is shown in Exhibit 1. The spatial distributions of PCE, TCE, and 1,2-DCE are shown in Exhibit 4. The plume present in the groundwater is considered to be bound from the top to the bottom of the saprolite and extends down to the depth of 15 m below ground surface as suggested by groundwater and soil sampling data. Initially, the plume is assumed to have the same spatial contaminant distribution throughout the depth.

RESULTS AND DISCUSSIONS

Flow

Groundwater flow was calibrated using the groundwater levels recorded at the well-monitoring stations. The main calibrating parameters were hydraulic conductivity in saprolite and bedrock layers along the strike, porosity, and dispersivity. Exhibit 5 represents the map of computed hydraulic head at the fifth layer (inside the transition zone, 7 m below ground surface). The primary ground flow direction is shown by arrows.

Monitoring-well stations along the primary groundwater flow path are shown in Exhibit 5. Major groundwater flow directions in the transition zone at the site are from Pine Ridge on the north and Chestnut Ridge on the south toward the valley, and then along the strike in the Nolichucky Shale toward the east. The groundwater velocity in the Nolichucky Shale transition zone is calculated in the order of 2 m/d close to OSY (at GW-271), which increases to 4.5 m/d, 1 kilometer away along the strike (at 56-6A). One kilometer further downstream (not shown in Exhibit 5) it increases to almost 12 m/d. This is consistent with the assumptions of numerical modeling performed by the US DOE using CRAFLASH (US DOE, 1998), which predicted the groundwater velocity along the strike in the transition zone between 5 and 8.5 m/d.

The computed phereatic surface along the primary flow path (along the strike in the Nolichucky Shale toward the east) is compared with the recorded groundwater elevation at the monitoring-well stations in Exhibit 6.

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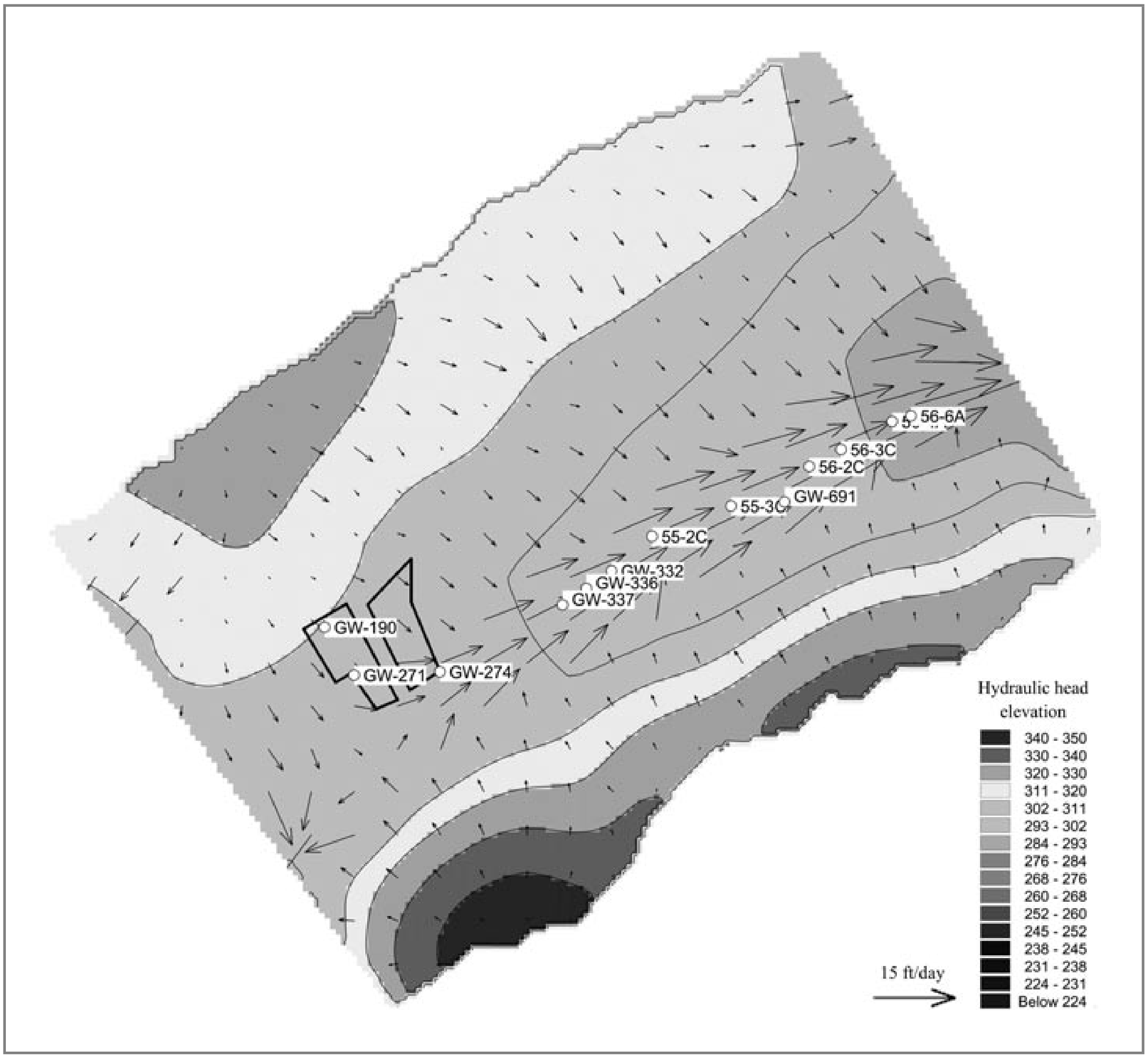

Exhibit 5. Hydraulic head elevation map at the transition zone; white circles are groundwater monitoring wells along the primary flow path, vectors are groundwater flow velocity vectors

Transport

Assuming the initial plumes as shown in Exhibit 4, the migration of the plume is predicted for the next 100 years. Measurements of VOC concentrations at the sampling stations between 1998 and 2010 were used to calibrate the transport computations. The groundwater sampling stations used for the calibration are shown in Exhibit 7.

Considering all the assumptions embedded in creating the present subsurface numerical transport model, it was able to roughly estimate the groundwater concentration of VOCs at the monitoring stations as shown in Figure 6. Based on the calibrated model, the VOC plume migration is simulated for the next 100 years. The plume migration is shown in Exhibit 8 for 10, 50, and 100 years.

Retardation factors of the VOC plume were determined for each contaminant using the numerical model simulations. Transport of a tracer plume with a zero partitioning

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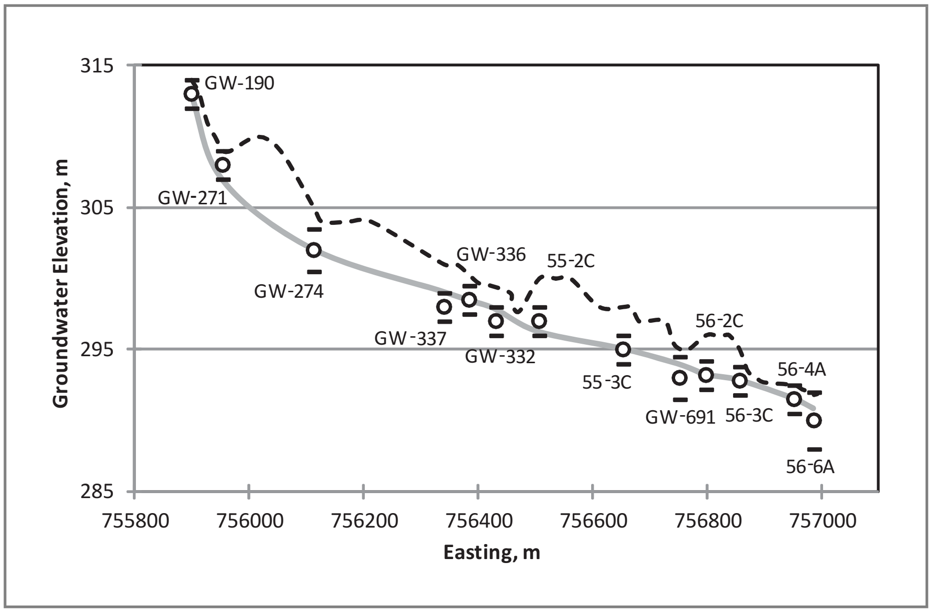

Exhibit 6. Computed phereatic surface along the primary groundwater flow direction (along strike in Nolichucky Shale) against the recorded groundwater elevation in monitoring wells; the light continuous line is computed phereatic surface, circles and bars are recorded groundwater elevation and standard deviations, and the dashed line is ground surface elevation

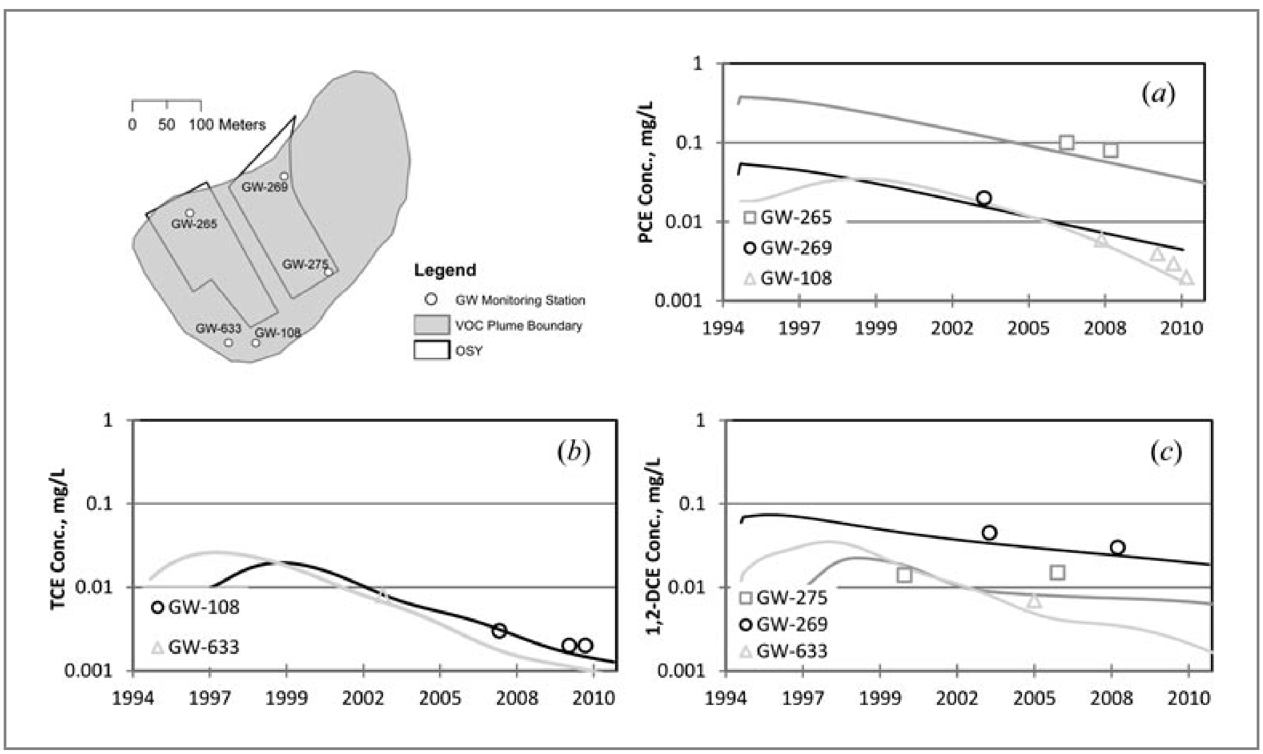

Exhibit 7. Calibrated model predictions against observed data for the transition zone layers (5 to 8 meters below ground surface): (a) PCE, (b) TCE, and (c) 1,2-DCE; continuous lines are model estimations at the location of groundwater monitoring stations shown in the top left corner

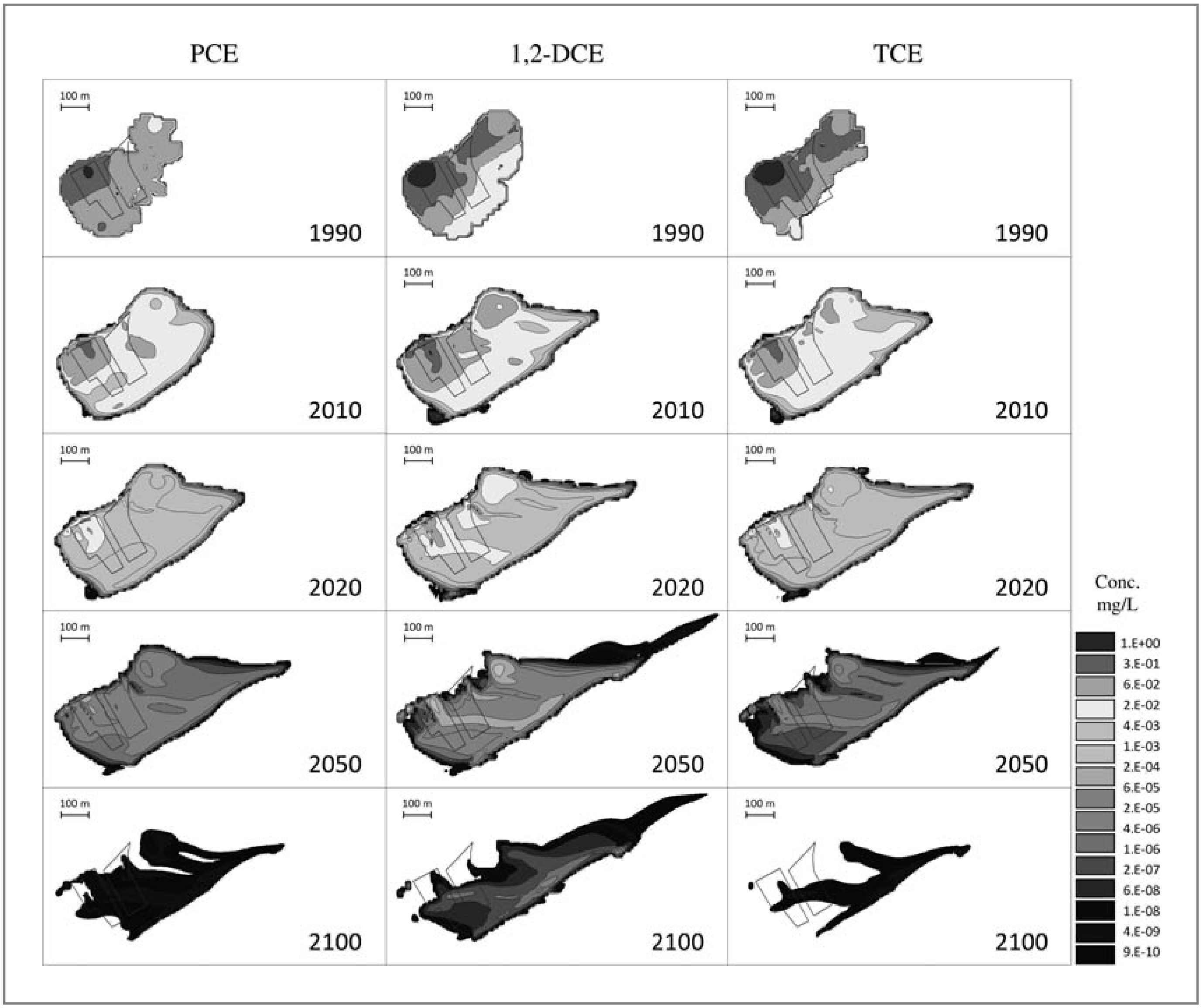

Exhibit 8. VOC plume migration in transition zone for the next 100 years

coefficient ($k_d = 0$) were modeled using the same initial concentration distribution as the VOC plume. The relative distance of the center of mass of the tracer plume and VOC plume were considered as the retardation factor for the corresponding VOC plume. Retardation factors are compared in Exhibit 9.

As expected, the retardation factors of the VOC plume are not constant with time since the Freundlich adsorption isotherms were used in the model. The lower the concentration values, the higher the retardation factor. Rogers (1992) reviewed the retardation factors of PCE obtained during laboratory and field studies to be between 1.1 and 22 for k_d values ranging from 0.3 to 2 L/kg. Benker, Davis, & Barry (1998) reported a similar review on the retardation factor of TCE to vary from 1.2 to 1.7 for K_{oc} values of 33 to 100 L/kg.

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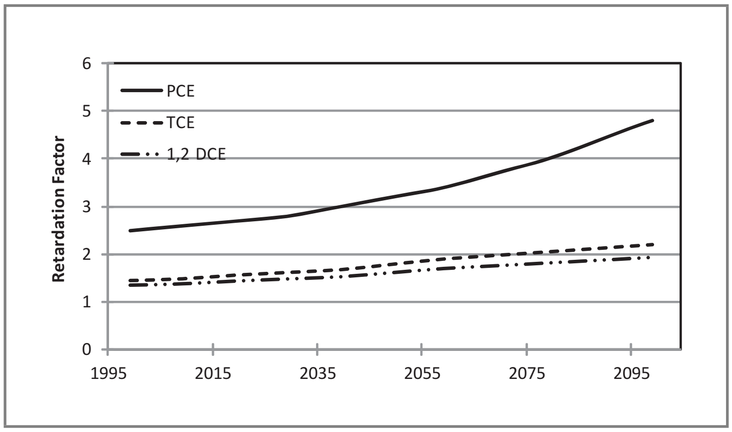

Exhibit 9. Retardation factor of VOCs computed using the numerical simulation of plume migration

The estimated concentration of VOCs in groundwater depends on the site hydrogeologic parameters as well as chemical characteristics of the specific VOCs. Chemical characteristics, as listed in Exhibit 3, have already been verified through laboratory and field studies. However, the hydraulic conductivity of the transition zone along the strike is still debated. A series of simulations has been performed to study the sensitivity of this parameter to the model results.

As shown in Exhibit 10, conductivity has the least effect on the transport of VOCs inside the plume boundary (Point 1); as this region is not entirely inside the Nolichucky Shale and the groundwater velocity is the lowest, as shown in Exhibit 5. For Point 2 on the plume boundary (with initial zero concentration for TCE and 1,2-DCE, and 0.02 mg/L initial PCE concentration), conductivity, and, therefore, groundwater velocity starts to play a significant role in plume transport. Lower-conductivity values remarkably delay the initial transport of the plume by more than 10 years, while the effect of higher-conductivity values is almost negligible. On the other hand, higher conductivity hastened the migration of the plume after the maximum concentration is reached, while the lower conductivity almost has no effect on the plume migration at this time. It seems the plume boundary acts as an inflection point for the effect of conductivity on the plume transport. In other words, inside the plume boundary, k_d values dominate the transport and the effect of conductivity is minimal. Outside the plume boundary (Point 3), the transport is mainly dominated by groundwater velocity and, thus, hydraulic conductivity values. Significant delay in the case of low conductivity and significant increase in the case of high conductivity is estimated at Point 3. Higher conductivity values in this region may cause the VOC plume to reach higher concentration values almost seven years earlier. Groundwater flow in this region will eventually be captured by tributaries and drainage network of the process buildings.

Estimation of the conductivity along the Nolichucky Shale is essential for remediation plans and strategies. Based on the above simulations, the human health risk-based groundwater concentration values for PCE, TCE, and 1,2-DCE (US DOE, 1998) will not be exceeded beyond the OSY boundaries even in the presence of a higher-conductivity transitional zone.

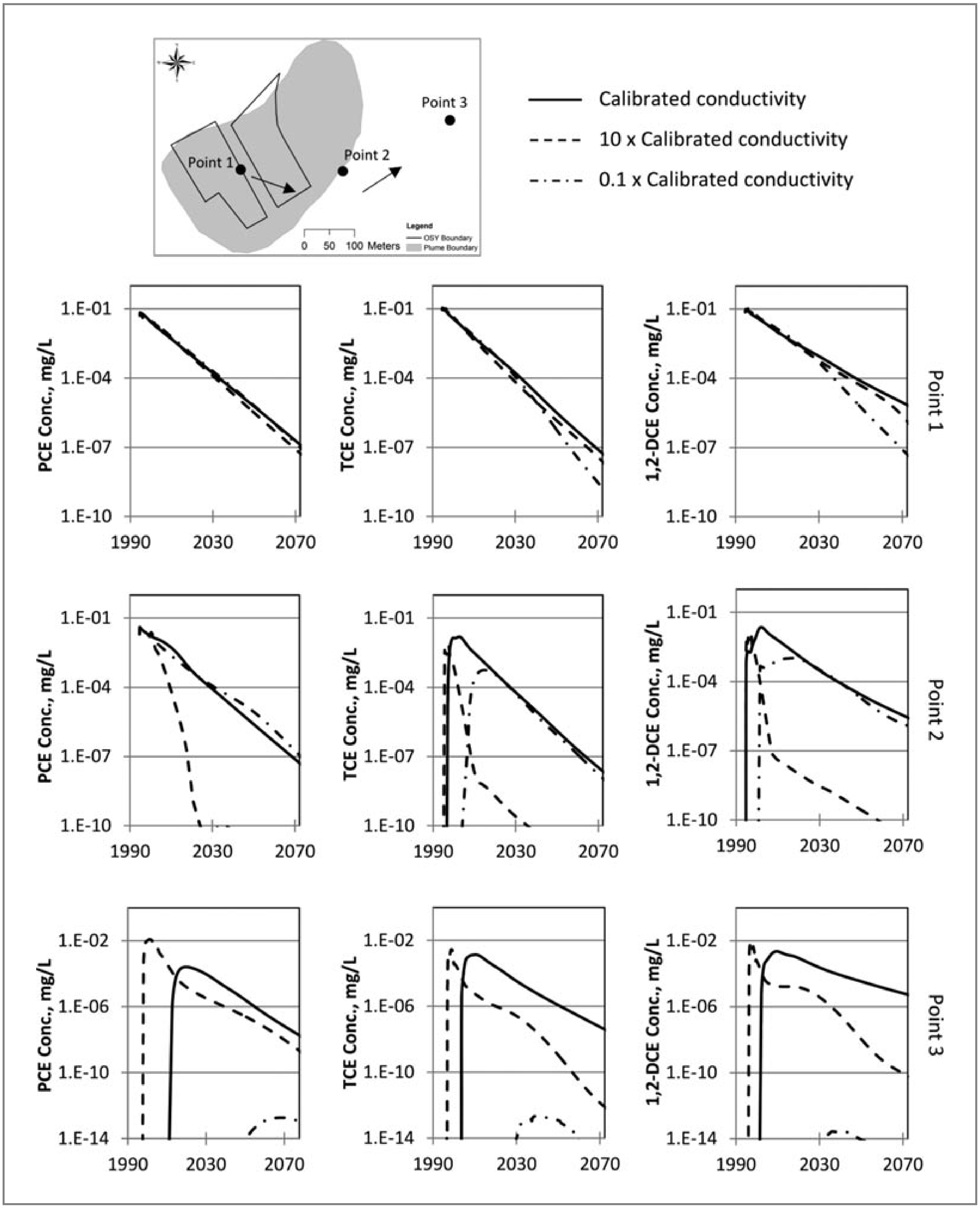

Exhibit 10. Effect of hydraulic conductivity of the transition zone on plume migration; top row: Point 1, middle row: Point 2, and bottom row: Point 3, as shown on the guide map

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CONCLUSION

Considering the recent hydrogeological surveys at the Y-12 NSC site, an integrated surface/subsurface flow and transport model was developed to simulate the long-term fate and transport of the chlorinated VOC plume in groundwater at the vicinity of OSY. The focus of numerical modeling was on the preferential flow pathway identified in the transitional zone inside the Nolichucky Shale bedrock. Despite the major assumptions in generating the model, it shows an acceptable accuracy in predicting the recent migration of the VOC plume. Based on the simulation results for the next 100 years, concentrations of PCE, TCE, and 1,2-DCE will not exceed the human health risk-based levels beyond the boundary of the OSY. After almost 20 years, the concentration of VOCs in groundwater is expected to decay below the risk-based levels even inside the OSY boundary. Transport of VOCs inside the initial plume boundary is mainly governed by matrix diffusion and partitioning coefficient, while outside the plume boundary, hydraulic conductivity and, thus, the groundwater flow dominate the plume migration. Uncertainty analysis shows that for higher-conductivity values in the transitional zone, the plume reaches the process buildings drainage system 7 years earlier and in higher-concentration values. Possible errors in measuring the conductivity of this preferential flow pathway or any other future hydrogeological deformations at the site may result in contamination of surface water by VOCs.

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