SUBSURFACE ONE-DIMENSIONAL TRANSPORT OF DIOXIN BENEATH AN UNCONTROLLED DUMP SITE – A CASE STUDY

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ABSTRACT

Measurements of 17 dioxin and furan congener concentrations were made as successive layers of clayey glacial till were excavated at a former municipal and industrial dump site. The measurements demonstrate separation of 1,2,3,4,6,7,8-HpCDD, OCDD and 2,3,7,8-TCDD with increasing depth. The dioxin congener 2,3,7,8-TCDD penetrated the clay to depths exceeding two meters. Application of the advective-dispersive one-dimensional transport equation for a reactive solute resulted in a reasonable fit to subsurface concentration measurements. All boundary and material variables were treated as constants when best-fitting the measured congener concentration distributions. The congener specific retardation factors, R_d's, and initial concentrations, C_o's, were necessarily varied. C_o's for respective congeners were selected to be an area average measured during the initial investigation. R_d's were selected that resulted in the best visual fit of data to the transport equation. A good fit between the measured and calculated depth concentration distribution occurred when the ratio of R_d's was approximately 6:1:4 for 1,2,3,4,6,7,8-HpCDD, 2,3,7,8-TCDD and OCDD respectively. These results differ insignificantly from corresponding ratios of octanol-water partitioning coefficients, approximately 10:1:10, that expectedly would be similar. The depth of 2,3,7,8-TCDD penetration was greater than anticipated. Uncertainties associated with estimating the advective-dispersive transport equation variables explain the difference between expectations and observations.

Dioxin-like compounds other than 1,2,3,4,6,7,8-HpCDD, OCDD and 2,3,7,8-TCDD were not detected at reportable concentrations below the surface. The dioxin-like compounds measured at the dump site were those compounds with nonzero toxicity equivalency factors as identified in Estimating Exposure to Dioxin-Like Compounds , EPA/600/6-88/005B1.

Keywords: composite, soil, sample, dioxin, furan, transport, subsurface, groundwater

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1. INTRODUCTION

1.1 Site Location and History

Dioxin and furan measurements were made at the Krejci Dump Site during both the initial site characterization in 1996 and remediation in 2012 (BOR 1996a, BOR1996b, BOR 2002a, BOR 2002b, NPS 2012). The roughly 200,000 m^2 site is a former municipal and industrial dump and salvage located within the Cuyahoga Valley National Park in Summit County, Ohio, USA (41°16'17.63"N, 81°32'40.33"W). During the years of operation, from approximately 1950 to 1980, large volumes of solid and liquid waste materials were brought to the dump, where significant quantities of hazardous substances were released to the environment as a result of open dumping, spills, leaking containers, and burning. The United States purchased the land in 1980 for management by the Department of the Interior National Park Service (NPS). In 1987, it was determined that the Site constituted a threat to human health and the environment. In response to this determination, the U.S. Environmental Protection Agency (EPA) initiated an emergency removal in June 1987. In November 1988, the NPS completed the removal of wastes that had been staged by the EPA and also removed some unconsolidated wastes and contaminated soil. Large quantities of debris and contaminated soil remained. An extensive remedial investigation was performed during 1994 through 1996. Debris and soils containing unacceptable levels of contaminants were excavated and removed from the site between October 2005 and December 2011. Site restoration was completed in 2012.

1.2 Dioxin Contamination and Remediation

The remedial site investigation discovered unacceptable dioxin-like congener concentrations in surface soil within and adjacent to a 10 m to 20 m deep debris-filled ravine. A fire had purportedly smoldered in this ravine during most of the facilities approximately 30 years of operation. Seventeen discrete locations were investigated to determine dioxin and furan congener concentrations in 1996. The area contaminated with dioxin and furan was estimated to be approximately 44,000 m^2. Many other chemical contaminants, including organic solvents, were also found to be present[5].

Remediation goals were established for dioxin-like compounds and 33 other contaminants. It was determined that soil having a calculated 2,3,7,8 TCDD toxicity equivalency factor (TEQ) greater than 3 pg/g would be excavated and removed from the site (Blancato, 1988, NPS, 2002b) .

The initial remediation effort required removal of: 1) all debris; 2) visibly contaminated or altered soil; and 3) minimally, an additional 15 cm of soil where contaminants had been discovered to exceed remediation goals. This effort was followed by collection of 40-part composite soil samples in each of 11 approximately 4000 m^2 subareas of the site. The 11 subareas are outlined on Figure 1. Contour lines on this figure represent approximately 3 m elevation intervals and generally define the ravine location at the time of the remedial investigation. For each subarea, 40 equal volume specimens of surface soil were collected at approximately 10 m spaced nodes of a rectangular grid and combined to create a composite sample. The concentrations of 17 dioxin and furan congeners were measured for each sample. Following the initial remedial effort, only 2 of the 11 subareas achieved the TEQ goal of 3 pg/g.

Minimally 15 cm of soil was removed from the entire surface of the 9 failing subareas. Forty-part composite samples, one representing each subarea, were collected and analyzed for dioxin-like compounds. At the end of this second remediation effort, 5 of the original 11 subareas had achieved the 3 pg/g TEQ goal. All remaining failing subareas included a part of the ravine.

Again, minimally, 15 cm of soil was removed from the entire surface of the 5 remaining failing subareas. Each remaining subarea was further subdivided into four approximately 1000 m^2 “quarter-areas” to better focus the remaining remedial effort. Quarter-areas had unique alphanumeric identifiers as shown on Figure 1. Forty-part composite samples were created to represent individual quarter-areas by combining equal volume soil increments collected from node points of an approximately 5 m grid.

Subsequent episodes of sampling, analysis and excavation occurred in quarter-areas having calculated TEQ greater than 3 pg/g. The excavation, sampling and analysis sequence was iterated in each quarter-area until the calculated TEQ of the quarter-area composite sample was less than 3 pg/g. Each failure to achieve the desired TEQ resulted in minimally 15 cm of soil being removed from the entire failing quarter-area. Excavation as deep as 1.7 m was required to achieve the TEQ remediation goal following the initial removal effort.

1.3 Stratigraphy and Soil Characteristics

The site is located on a narrow ridge of glacial, glaciofluvial, and lacustrine sediments within the buried pre-glacial valley of the Cuyahoga River (41°16'18.11"N, 81°32'40.64"W). The ridge is approximately 30 m above adjacent valleys. Sediments are at least 60 m deep and consist of four basic glacial till units (BOR 2002a). Only the upper two of these units are pertinent to this discussion and are described in the following two paragraphs. Dioxin contamination was located in and adjacent to a 5 to 20 meter deep ravine that had been filled with municipal and industrial waste mixed with other debris. A fire smoldered in the debris filled ravine for several decades when the dump was in operation. The ravine incised the upper till unit and cut deeply into the second unit. Ravine slopes in the area are typically 2H:1V. Precipitation permeated the waste and debris resulting in locally perched water, both within the debris and adjacent slopes. An unnatural perennial stream in the ravine was maintained by water slowly released from storage within the debris. The water stored in the waste and debris contained dissolved dioxins and furans and infiltrated the adjacent clay tills. This was the primary source of soil dioxin and furan contamination.

The uppermost unit consists of approximately 10 m of weathered, dense, homogenous, relatively stiff, yellow-brown clay with a few scattered pebbles and cobbles. Naturally occurring near surface soil is classified as Lean Clay (CL) with about 10 percent fine sand and about 90 percent medium-plasticity fines (fines are particles less than 0.075 mm in size.) (ASTM, 2011). Petrographic analyses revealed that the near surface clay minerals are composed of about 5 to 25 percent vermiculite, 5 to 25 percent illite/mica, and 5 to 20 percent kaolinite, and contain minor traces of mixed-layer minerals. In-situ dry unit mass is about 1680 kg/m, the measured particle average specific gravity is approximately 2.71, and soil porosity is approximately 0.38. Laboratory measurement of vertical hydraulic conductivities using intact core samples ranged from 2.8 x 10^{-8} cm/s to 8.6 x 10^{-8} cm/s. Significant high angle to vertical fractures were observed in nearby road cuts and later on the excavated ravine slopes. Six approximately 46-cm diameter single-ring surface infiltration tests were performed. Calculated hydraulic conductivities using measurements from these tests ranged between 7 x 10^{-6} cm/s and 6 x 10^{-5} cm/s [5].Twenty-one site-wide measurements of total organic carbon content, representing upper unit soil at a depth of about 0.3 m, ranged between 4.7 and 0.28 percent with an arithmetic mean of about 1 percent.

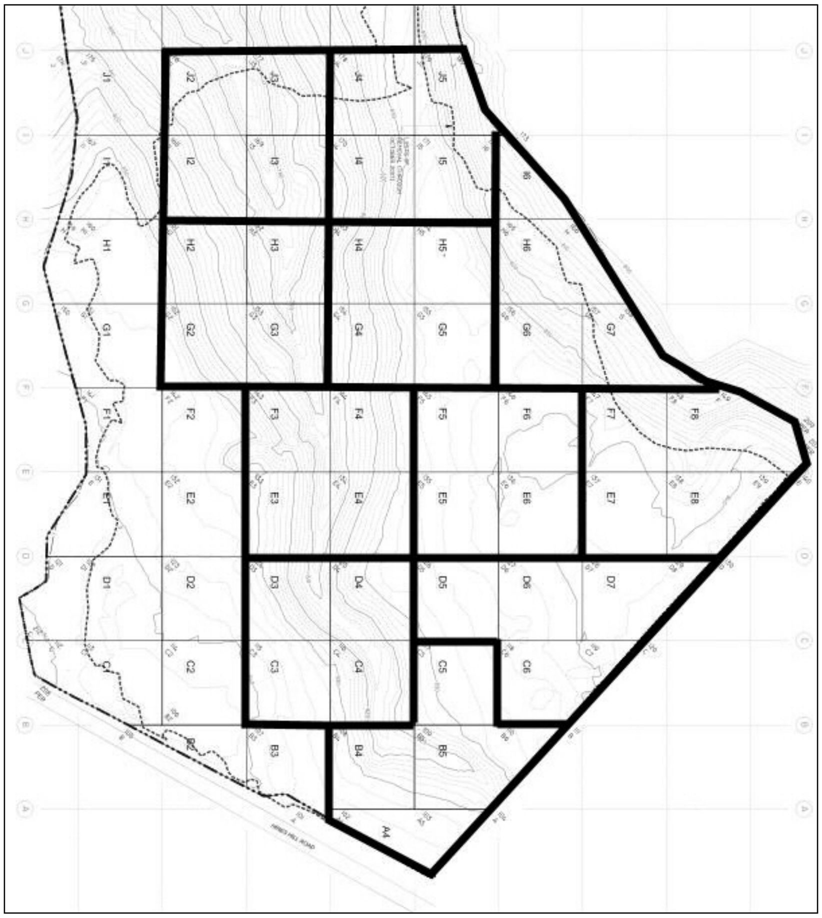

Figure 1. Limits of the Krejci Dump West Site: Eleven Approximately 4000 m^2 Remediation Subareas (bold outline) and Interior Quarter-areas.

The lower second unit is 20 m to 30 m of predominantly unweathered, dense, gray Lean Clay (CL) having approximately 95 percent fines, containing a few pebbles and cobbles, and having infrequent 0.7 m to 3 m thick zones of thinly laminated silts and clays of lacustrine origin. This unit also contains occasional lenses of silt or sand which are generally 0.3 m to 1 m thick. These lenses are expected to be isolated, discontinuous, and may not be oriented horizontally. Laboratory testing on intact core samples measured hydraulic conductivity of the gray clay ranging between 1.5 x 10^{-8} cm/s and 8.7 x 10^{-8} cm/s. This unit is highly erosive and contains infrequent vertical fractures. Vertical fractures were not observed in core samples or laboratory test specimens. Although infrequent, vertical fractures and other minor geologic details are expected to cause the unit to have an average vertical hydraulic conductivity much greater than the average laboratory measured values.

The approximately 5 m to 20 m deep ravine, in which the subject dioxin and furan contamination was found, completely incises the upper unit (yellow clay unit) and extends about 15 m into the lower unit (gray clay unit). Fourteen measurements of cation exchange capacity, representing both units at the site, ranged between 6.9 meq/100g and 15.1 meq/100g.

2. MATERIAL AND METHODS

The dioxin-like compound investigation plan and results are presented and discussed in several project reports (BOR 1996a, BOR1996b, BOR 2002a, BOR 2002b, NPS 2012). Surface dioxin-like compound measurements representing 17 locations were made during the 1996 site characterization. There were no subsurface measurements at that time. Seventeen congeners were evaluated. Dioxin-like compounds moved into the soil adjacent to the debris-filled ravine by mechanisms of advective transport and chemical diffusion. Initially all 17 dioxin and furan congeners were measured at quantifiable concentrations in ravine debris and soil. However, only three of these had reportable concentrations in subsurface soil following the second excavation effort. The remaining quantifiable congeners were 1,2,3,4,6,7,8 HpCDD, OCDD, and 2,3,7,8 TCDD and these are the focus of this paper. Concentrations were observed to decrease with depth for all three of these compounds. The concentration v. depth distribution for these three congeners would expectedly be described by the well-known "advective-dispersive" equation for reactive solutes.

The contaminant transport model for one-dimensional flow of a reactive solute is described by the "advective-dispersive" equation for reactive solutes (Freeze and Cherry, 1979).

$$\frac{\partial c}{\partial t} = \frac{D}{R_d}\frac{\partial^2 c}{x^2} - \frac{v_s}{R_d}\frac{\partial c}{\partial x}$$

Where:

R_d is the " retardation factor"; $R_d = \frac{v_c}{v_s}$;
v_s = average interstitial seepage velocity;
v_c = average velocity of the retarded solute;
t = time elapsed;
x = distance;
c = concentration of organic solute in water;
D = hydrodynamic dispersion coefficient.

From this equation it is evident that the concentration variation with time and distance is completely described by the two ratios D/R_d and v_s/R_d. Herein, values for these two ratios were determined that best-fit the equation to the concentration v. depth distribution for each of the three congeners.

D/R_d and v_s/R_d that best-fit the field data are derived for each of the three congeners. These estimates are then compared to similar ratios calculated using estimates of v_s, R_d and D derived from commonly used predictive mathematical expressions and information acquired during the remedial investigation such as soil density, specific gravity, hydraulic conductivity, and organic carbon content.

The retardation factor, R_d, is expected to be different for each congener. R_d is subsequently identified using the terms $R_{d(HpCDD)}$, $R_{d(TCDD)}$, and $R_{d(OCDD)}$ when referring to 1,2,3,4,6,7,8 HpCDD; 2,3,7,8 TCDD and OCDD respectively.

An analytical solution to the advective-dispersive equation has been developed by Ogata and Banks (Ogata and Banks, 1961) and it is this solution that is best-fit to the field measurements in the next section.

$$\frac{c}{c_o} = \frac{1}{2}\{erfc(Z_1) + erfc(Z_2)erfc(Z_3)\}$$

Where:

$$Z_1 = \frac{x - v_{Rs}t}{2\sqrt{D_R t}}$$

$$Z_2 = \frac{v_{Rs}x}{D_R}$$

$$Z_3 = \frac{x + v_{Rs}t}{2\sqrt{D_R t}}$$

and

$$v_{Rs} = \frac{v_s}{R_d} \quad ; \quad D_R = \frac{D}{R_d}$$

The value of c is the concentration at depth x and time t. The variable t is the time that has elapsed since the introduction of the contaminant at the surface. This solution to the advective-dispersive equation inherently assumes a constant contaminant source concentration c_o and one-dimensional flow into a homogeneous, isotropic half-space of clean soil. The elapsed time (t) used for all calculations is 30 years. Arguably, t could be 40 or even 50 years since the burning of chemicals that could create dioxins may have been ongoing before the 1960's and most remediation measurements were acquired in 2010. However, it is difficult to establish when the source of dioxin was removed. Herein it is presumed that the source was removed in the late 1980's when much of the bulk waste was removed from the valley.

The ratios D/R_d and v_s/R_d, determined by the method just described, will be compared in the next section to ratios D/R_d and v_s/R_d calculated using a typical predictive approach. A hydraulic gradient of one is assumed for both calculations. The assumption of a hydraulic gradient of one is justifiable because the thick, clayey, glacial till is underlain by a sand layer. The sand layer acts to drain water from beneath the clayey till with water exiting the sand layer where it intersects adjacent valley side slopes. Also, the depth of perched water in the debris is small relative to the thickness of the clayey till layer. Under these conditions, the change in total hydraulic head is approximately equal to the change in elevation head: consequently, a gradient equal to approximately one is reasonably expected.

3. RESULTS

3.1 Dioxin Congener Concentration v. Depth

Dioxin-like compounds moved into soils adjacent to the ravine by mechanisms of advective transport and diffusion. Of the 17 dioxin and furan congeners investigated, only three had reportable concentrations in subsurface soil following the second excavation effort. These were 1,2,3,4,6,7,8 HpCDD; 2,3,7,8 TCDD and OCDD. Concentrations were observed to decrease with depth. Ratios of measured concentrations to the measured surface concentrations following the second excavation effort were calculated and are presented on Figure 2. Individual quarter-areas are identified in the legends of these figures using alphanumeric quarter-area identifiers. The depth of the initial soil excavation was not measured but observations suggest it varied between 0.5 m and 2 m. A depth of 1 m is used to represent the initial excavation on Figure 2 and the included graphs are shaded in this depth range to remind the reader that this depth is uncertain. It is also noteworthy that physical and chemical characteristics of the soil and debris removed by initial excavation were sometimes significantly different than unaltered native soil. Furthermore, the depositional characteristics of glacial tills cause them to be inhomogeneous and anisotropic. Regardless, all soil is treated as homogeneous and isotropic to simplify this analysis.

The solution to the advective-dispersive equation for one-dimensional flow of reactive solutes through soil, subject to reversible sorption reactions, was visually best-fit to each of the distributions. The average concentrations measured at the time of the remedial investigation were used to represent C_o for each congener. These concentrations were 1015 pg/g, 11.7 pg/g and 12850 pg/g, for 1,2,3,4,6,7,8-HpCDD; 2,3,7,8-TCDD and OCDD respectively The best-fit relationships are shown on Figures 1, 2 and 3. All material variables in the equations were held constant for all calculations except that different R_d and C_o were selected to represent each congener. A good fit between the measured and calculated concentration distribution with depth occurred when the ratio of R_d's was 6:1:4 for 1,2,3,4,6,7,8-HpCDD; 2,3,7,8-TCDD and OCDD respectively. A ratio of 10:1:10 would be expected if R_d had varied in approximate proportion with octanol-water partitioning coefficients (K_{ow}). The reason for this expectation is discussed in the next section.

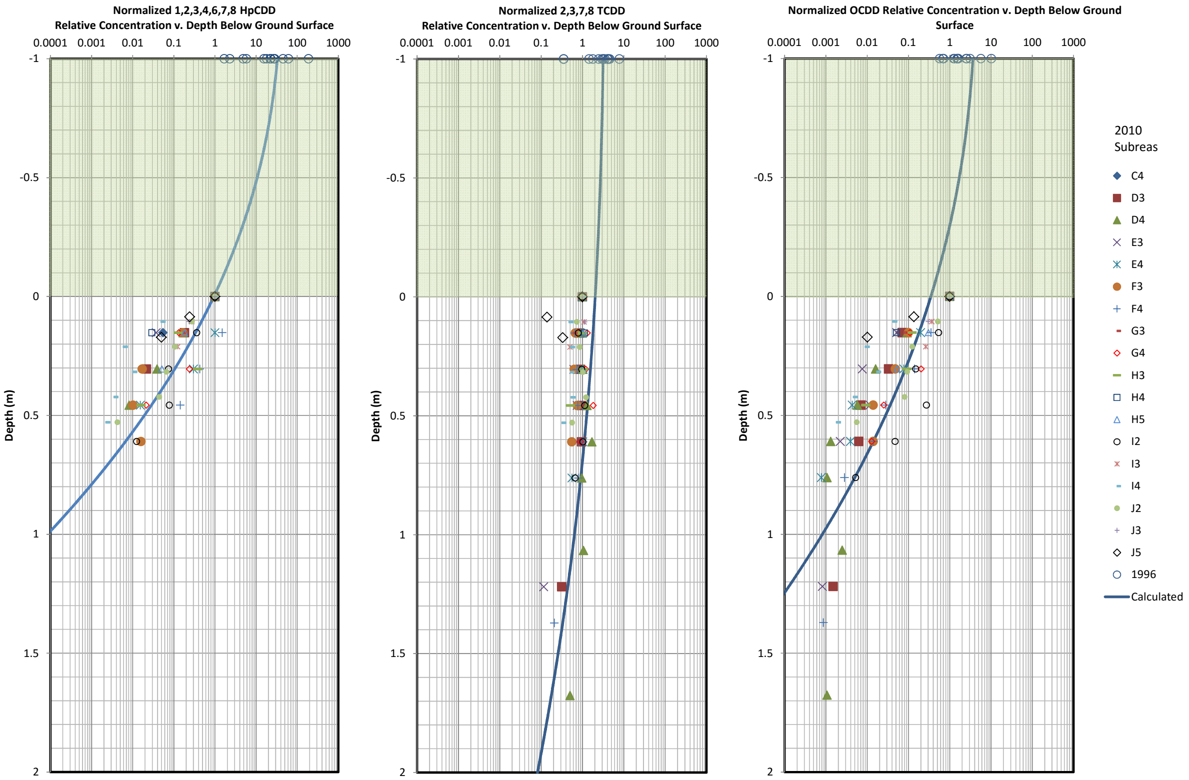

Figure 2. Relative Concentration Measurements v. Depth with Best-Fit Curve.

The values used to visually best-fit the advective-dispersive equation to each congener, including the best fit ratios of D/R_d and v_s/R_d, are presented in Table 1.

Table 1. Values Used to Visually Best-fit the Advective-Dispersive Equation.

Variable	1,2,3,4,6,7,8-HpCDD	2,3,7,8 TCDD	OCDD
$R_d/R_{d(TCDD)}$	6	1	4
D/R_d (m^2/s)	8.17E-11	4.90E-10	1.23E-10
t (sec)	9.46E+08	9.46E+08	9.46E+08
v_s/R_d (m/sec)	1.67E-10	1.00E-09	2.50E-10
C_o (g/g)	1.02E-09	1.17E-11	1.29E-08

3.2 Derivation of D/R_d and v_s/R_d Ratios Using a Common Predictive Approach

Values for variables used in the advective-dispersive equation are estimated and compared to best-fit values presented and discussed in the previous section.

R_d can be estimated for each congener for the condition of saturated flow using soil porosity (n), bulk density (ρ_d), and the compound partition coefficient (K_d).

$$R_d = [1 + K_d \frac{\rho_d}{n}]$$

$\rho_d = 1.650$ g/cm^3 and $n = 0.38$ were previously presented as results of the remedial investigation.

K_d is commonly calculated using the relationships:

$$K_d = K_{oc}(f)$$

$$logK_{oc} = A + B\ logK_{ow}$$

Where:

A and *B* are constants derived by previous study for sets of organic compounds and found in literature (Dragun, 1988). For this analysis, A= .088 and B=.909 (Hassett et al., 1983),

K_{ow} is the octanol-water partitioning coefficient: use $log\ K_{ow} = 7$ for TCDD and $log\ K_{ow} = 8$ for OCDD and HpCDD; these values reflect one-significant-digit approximations of values commonly presented in literature (these values may be +/- 1) (USEPA, 1994),

f is the fraction of soil mass that is organic carbon: the lowest value from remedial investigation report, $f = 0.0028$, is thought to best represent the subsurface soil and used for this analysis.

The calculation results are:

$R_{d(TCDD)} = 75{,}000$;

$R_{d(OCDD)} = R_{d(HpCDD)} = 750{,}000$.

The average interstitial velocity, v_s, is the same for all three congeners since all three are subject to the same water flow rate. v_s may be estimated by the equation.

$$v_s = \frac{ki}{n}$$

Where:

k = hydraulic conductivity; measured during the remedial investigation to be in the range between 10^{-8} cm/sec and 10^{-4} cm/sec; the value 10^{-5}, nearer the higher end of this range is selected since both field and laboratory measurements generally underestimate the effects of minor geologic details resulting in a bias toward low k values.

i = hydraulic gradient; a unit gradient is estimated for the site conditions that existed during dump operations and until debris removal was nearly complete;

n = porosity; $n = 0.38$ is estimated using the results of the remedial investigation.

The calculated value is: $v_s = 2.6 \times 10^{-5}$ cm/sec

To simplify subsequent analysis, *D* will be assumed equal for each congener. This is a reasonable assumption since only an order of magnitude estimate of the value can be made under the best experimental conditions. Furthermore, all congeners are subject to the same flow velocity and congener molecules are all approximately the same size. The magnitude of *D* depends on both molecular diffusion and mechanical dispersion. Herein, a typical value for nonreactive organic compounds, $D = 10^{-6}$ cm^2/sec, will be used.

The ratios D/R_d and v_s/R_d are calculated and presented in Table 2 with other variables used in the advective-dispersive equation to predict congener transport.

Table 2. Calculated ratios D/R_d and v_s/R_d and Other Predictive Parameters.

Variable	1,2,3,4,6,7,8-HpCDD	2,3,7,8 TCDD	OCDD
$R_d/R_{d(TCDD)}$	10	1	10
D/R_d (m^2/s)	1.3E-12	1.3E-11	1.3E-12
t (sec)	9.46E+08	9.46E+08	9.46E+08
v_s/R_d (m/sec)	3.5E-11	3.5E-10	3.5E-11
C_o (g/g)	1.02E-09	1.17E-11	1.29E-08

The predicted curves for t = 30 years are presented as a dashed line on Figure 3 with the previously discussed normalized field data. It is evident that the predicted depth of dioxin movement is considerably less than measured at the site. Such error can be explained as resulting from large uncertainties in estimated values for problem variables. For examples, the estimates of permeability coefficient, octanol water partitioning coefficient, and hydrodynamic dispersion are, at best, accurate only to plus or minus one order of magnitude.

4. DISCUSSION/CONCLUSION

Measurements of 17 dioxin and furan congener concentrations were made as successive layers of clay soil were excavated at the Krejci Dump Site, a former municipal and industrial dump site located in the Cuyahoga Valley National Park, Ohio. The measurements demonstrate separation of 1,2,3,4,6,7,8-HpCDD, OCDD, and 2,3,7,8-TCDD with increasing depth. The dioxin congener 2,3,7,8-TCDD penetrated the clay to depths exceeding two meters. Application of the advective-dispersive one-dimensional transport equation for a reactive solute (transport equation) resulted in a reasonably good fit to subsurface concentration measurements. All boundary and material variables were treated as constants when best-fitting the measured congener concentration distributions. Only the congener specific retardation factors, R_d's, and initial concentrations, C_o's, were varied. C_o's for respective congeners were selected to be the area average measured during the initial investigation. A good fit between the measured and calculated depth concentration distribution occurred when the ratio of R_d's was approximately 6:1:4 for 1,2,3,4,6,7,8-HpCDD; 2,3,7,8-TCDD and OCDD respectively. These results are reasonably different than corresponding ratios of octanol-water partitioning coefficients, approximately 10:1:10, that expectedly would be similar. The depth of penetration of the three congeners was also greater than calculated using common estimates of problem variables.

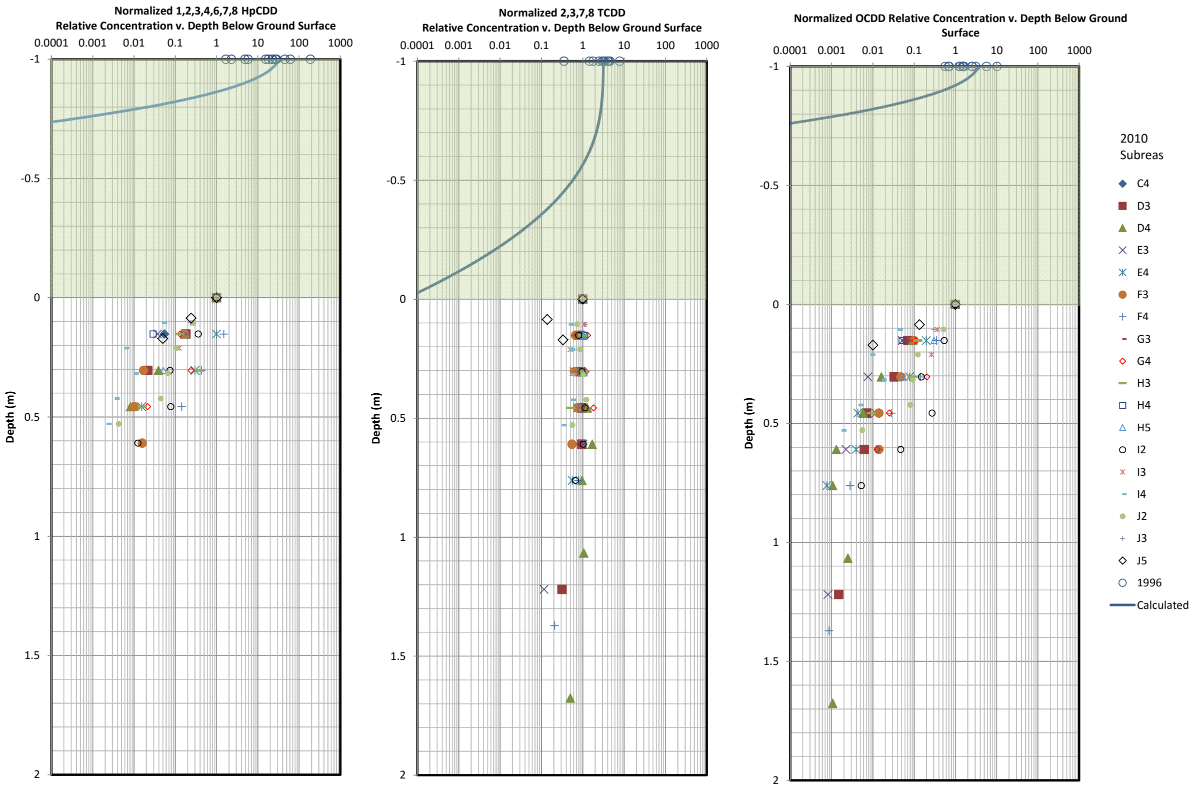

Figure 3. Relative Concentration Measurements v. Depth with Predicted Curve.

The difference between observations and expectations are explained by considering the large uncertainties associated with estimating the advective-dispersive transport equation variables. The large difference between observed and predicted depth of dioxin transport serves as a reminder that the problem variables of the advective-dispersive equation must be carefully defined and the subsurface characteristics measured by laboratory tests and in field studies in order to have a reasonable chance of useful predictions. Even with such care, prediction uncertainty will still remain significant due to the general inability to accurately describe the minor geologic details and associated hydraulic conductivity that control subsurface water movement and contaminant transport.

Mark C. Gemperline, Subsurface One-Dimensional Transport of Dioxin Beneath an Uncontrolled Dump Site– A Case Study, 26th Annual International Conference on Soil, Water, Energy, & Air, March 21-24, 2016 at the Mission Valley Marriott in San Diego, CA, AEHS Proceedings, 2016.

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