

# Quantifying Gas Phase Dispersion in Soil Media

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## ABSTRACT

Hydrocarbon contamination of unsaturated soils has been a serious environmental problem for decades. Two of the most common methods of remediating these sites are soil vapour extraction (SVE) and bioventing. Predictive models have been developed for the SVE and bioventing processes in order to estimate mass transfer coefficients for experimental systems and clean up times for field sites. These techniques are typically modeled using the advection-dispersion system of equations. Unfortunately, many of the required modeling parameters are still poorly understood or unavailable. Modellers of the process either utilize correlations to estimate dispersion coefficients or neglect dispersion altogether. Previous studies have demonstrated that uniform packed bed correlations can seriously underestimate dispersion occurring in non-uniform unsaturated soils under dry and moist conditions.

Currently, very little literature information is available regarding gas phase dispersion in unsaturated soils. Most experimental studies aimed at measuring dispersion have used disturbed soils, manipulated to some degree from field conditions. Therefore, dispersion estimates are not a true representation of actual field sites. Accordingly, research has been conducted on the development of a laboratory technique that can measure dispersion in both disturbed and undisturbed soils under unsaturated conditions. The apparatus has been developed and has been used to measure dispersion in disturbed soils. These results and the application to field sites will be discussed.

## INTRODUCTION

Recent advancements in the understanding of contaminant behaviour in soil media has led to the development of complex mathematical models for predicting subsurface contaminant transport. Numerous studies have focused on the behaviour of aqueous phase contaminants under saturated and unsaturated conditions. Unfortunately, there still exists little information in the literature regarding gas phase transport in unsaturated soils. This information is vital in predicting the behaviour of volatile compounds released from landfills and accidental spills as well as predicting the efficiency of bioventing and soil vapour extraction (SVE) remediation strategies.

Vapour movement through soil is determined by both the chemical properties of the contaminant, including solubility and Henry's Law coefficient; and soil properties,

including texture, porosity, permeability, organic matter and water content. Water content also affects the effective porosity and permeability of a given soil type and therefore becomes an important parameter when examining the effects of physical soil properties on subsurface transport.

Bioventing and SVE remediation induces the transport of contaminant vapours under convective conditions by means of forced air flow through the soil. One dimensional and three dimensional experiments of SVE and bioventing systems have been used to aid in the development of mathematical models for predicting removal of volatile compounds from contaminated soils. These system models are typically based around the advection-dispersion equation.

Studies by Kaleris and Croise (1999), Harper et al. (2002), Karen et al. (1995), Abriola et al. (1997), have all shown the advection-dispersion approach to modeling SVE and bioventing systems. Typically, the velocity term is easily determined, while the dispersion coefficient is more difficult to ascertain. Because of this fact, many studies have opted to neglect the dispersion term altogether (Harper et al., 2003; Nadim et al. 1997) or utilize packed bed dispersion correlations (Karan et al., 1995). Unfortunately, both of these cases can lead to erroneous results when attempting to predict the operational efficiency of SVE and bioventing systems.

The study by Gidda et al. (2004) demonstrated that current uniform packed bed correlations can seriously underestimate gas phase dispersion for non-uniform, air dry soils, tested over a broad range of soil types. Uniform packed bed correlations are typically based around a single mean particle diameter (Levenspiel 1972), which is not representative of a non-uniform packed bed material. This is especially true when the particle size distribution is very broad. Gidda et al. (2004) and Han et al. (1985) observed an increasing trend in solute dispersion with increasing particle size distribution for both the gas and aqueous phase transport. Additionally, Gidda et al. (2004) and Costanza-Robinson and Brusseau (2002) both demonstrated that gas phase dispersion increased with increasing moisture content beyond that measured for air dry conditions. It is apparent from the current literature that both the degree of non-uniformity in packed beds and moisture content has an impact on the degree of gas phase dispersion occurring in unsaturated soil.

Numerous studies have focused on aqueous phase dispersion in both saturated and unsaturated soil, as well as gas phase dispersion in packed beds. Costanza-Robinson and Brusseau (2002) attempted to draw the parallel between aqueous and gas phase porous media transport in order to explain the dispersive processes occurring under gas phase conditions. Clearly, additional research in this area is required to further understand and quantify the effects of gas phase dispersion in unsaturated soils in order to improve the predictive capabilities of SVE and bioventing models.

The proposed research was broken into two phases. Phase I was to improve the experimental technique used by Gidda et al. (2004) in order to quantify the contributions of both particle size distribution and moisture content on the level of gas phase dispersion occurring in unsaturated soils in the disturbed setting. Current literature values on gas phase dispersion have all focused on laboratory prepared (disturbed) soils. Phase II of the research is to use the improved experimental techniques and analyze undisturbed soil samples, in the form of soil cores. This will allow us to further quantify the effects of particle size distribution on dispersion, based on material better representative of field conditions.

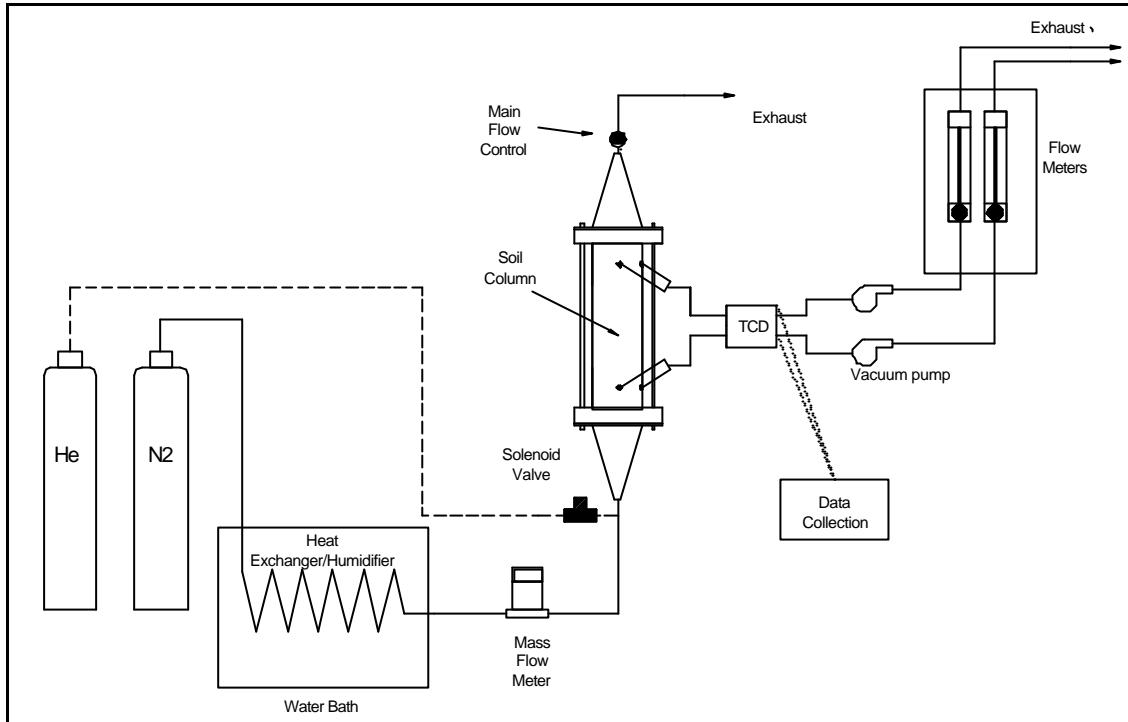
In this paper, emphasis is placed on Phase I of the research. Modifications to the experimental technique will be presented, as well as the dispersion coefficients for a range of disturbed soils under different conditions. The impact of having Phase II capabilities will also be discussed. This information will be valuable for the calibration and use of predictive SVE and bioventing models.

## **MATERIALS and METHODOLOGY**

### **Experimental Apparatus**

The experimental set up for this system is shown in Figure 1. Experiments for Phase I are completed using a 10.16 cm (4") inside dia stainless steel column that was 30 cm long. This column is packed with the soil being tested. The column is compressed between two identical supporting stainless steel plates and gaskets providing an air tight seal. Diffusers are welded on to each plate in order reduce the momentum of the flowing air stream and provide a uniform velocity profile at the base of the soil column. Steel grating (3mm and 0.5mm x 0.5mm openings) is placed at the bottom of the column to ensure an even distribution of flow and hold the weight of the soil. For the Phase II, it is expected that a 6.35 cm (2.5") inside dia column will be used.

Two sampling locations are located 5cm from either end of the column. At each sampling location, two 1/8" NPT fittings are connected to the column. Each fitting is over bored to accept a 1/8" diameter stainless steel sampling probe. The holes are drilled so that the two probes cross each other at 90 degrees inside the column. A series of four 3/64" holes are drilled along the length of each probe. Each probe is sealed on one end and inserted into the column with the holes perpendicular to the flow. Both probes are then connected together through 1/8" dia Teflon tubing and then connected to the inlet of one channel on the thermal conductivity detector (TCD). The flow rate through each of the TCD channels is controlled on the downstream end of the detector by a Neptune DynaPump (10psig), regulated through a Cole Parmer Flowmeter (N042-15) and then exhausted to the fume hood. The temperature of the TCD is maintained at a constant 42°C through an Omega DPi16 thermal process controller.



**Figure 1. Experimental Apparatus**

### **Porous Media**

Four different soils have been selected for use in the dispersion experiments, representing a broad range of soil types and particle size distributions. The four soils include Ottawa Sand (OS), Simcoe Loamy Sand (SLS), Elora Silt Loam (ESL), and Brookston Clay (BC). Both disturbed and undisturbed experiments will be conducted using these soil types. Properties of the four soils are shown in Table 1.

**Table 1. Soil properties**

| Soil Properties    | OS   | SLS  | ESL  | BC   |
|--------------------|------|------|------|------|
| % Sand (by weight) | 98.8 | 69.4 | 34.0 | 21.0 |
| % Silt (by weight) | 1.2  | 24.4 | 50.1 | 35.1 |
| % Clay (by weight) | 0    | 6.2  | 15.9 | 43.9 |
| Porosity           | 0.41 | 0.47 | 0.52 | 0.58 |

For Phase I, all the soils are disturbed with the exception of Ottawa Sand, and are put through a homogenization process. Ottawa Sand is different as it is homogeneous when purchased. The homogenization process involves air-drying the soil for 3-5 days, crushing it in order to remove large aggregates and then passing the material through a No. 10 sieve (2mm openings). Material remaining on the sieve after the first pass is then

crushed and passed through the sieve again. The overall process was repeated three times for each soil type. The material remaining after the third repetition was discarded. All processed material is kept in sealed plastic containers until use.

For Phase II it is expected that undisturbed soil samples will be collected in both the 2.5" dia and 4" dia stainless steel columns to test for wall effects. Special caps have been designed to slip over the ends of the columns so that they may be driven into the ground. The top cap will provide the surface for impact and prevent damage to the top of the soil column, while the bottom cap provides the cutting edge to force the soil up into the column as it is driven into the ground.

### **Tracer and Carrier Gas**

In order to isolate for dispersive effects, a non-reactive tracer was selected for the experiment. This prevents any additional rate limited mass transfer effects between organic matter or soil water which may enhance breakthrough curve spreading (Constanza-Robinson and Brusseau, 2002). Helium was selected as the non-reactive tracer in this study due to its high Henry's Law constant and low aqueous solubility. Helium has also been used as a tracer gas in the study by Westerterp et al. (1996) under similar experimental conditions on dispersion in uniform packed beds. The study by Gidda et al. (2004) used Sulphur Hexafluoride ( $\text{SF}_6$ ) as the non-reactive tracer for dispersion experiments. Due to the environmental impacts as a green house gas,  $\text{SF}_6$  was not considered for use in these experiments. Balloon grade helium was found to work well with the TCD, while helping to reduce some of the experimental costs. Nitrogen (99.8% pure) from a compressed gas cylinder was used to provide flow through the experimental apparatus.

### **Procedure**

The following experimental procedure is identical for both the disturbed and undisturbed soil samples, with the exception of the soil preparation. All disturbed soil experiments will start as pre-processed, air dried soils. Soil is added to each column in a series of lifts. Moisture contents will vary for each soil type ranging from approximately 5% moisture up to field capacity. Moisture content experiments will be prepared using a number of small lifts. The appropriate amount of water will be added to each lift using a repetitive volumetric dispenser and allowed to spread throughout the soil due to passive infiltration, similar to the procedure used by Gidda et al. (2004). For undisturbed soil experiments, soil cores will be collected at field sites and stored in the laboratory until use. Core samples will be sealed at each end in order to prevent drying. Each column will be pre-drilled in order to reduce sample disturbance when attaching sampling probes and fittings.

Nitrogen is used as the carrier gas through the system for the He tracer injections. Nitrogen will be first passed through a heat exchanger/humidifier system placed in a water bath at 18°C to prevent drying of the moist soil columns. Flow through the column is controlled using a combination nitrogen gas pressure adjustment and a ½” Swagelok Bonnett Needle Valve. Flow measurements through the system are measured using an Omega Engineering FMA 1828ST (0-50 L/min) mass flow meter. Flow through each channel of the TCD is maintained at 50 mL/min by the flowmeter on the downstream side of the detector.

A 0.1 second pseudo-dirac pulse of helium is injected into the system using a Burkett 3-way solenoid valve controlled by a relay for precise injection timing. The helium concentration is controlled through a pressure regulator on the compressed gas cylinder and also by the injection timing. This type of injection procedure reduces some difficulties experienced by Gidda et al. (2004) including syringe clogging, septum leaks and injection inconsistency. The TCD response signal and response duration are measured using a National Instruments PCI-6013 data acquisition card and recorded using LabVIEW 7.0 software. Signal conditioning is performed using an NI SCC-AI06 conditioning module to filter out additional 60 Hz noise from the TCD signal. Solenoid valve timing and tracer injection are controlled through a 5V digital output line on the data acquisition card. A series of three tracer injections will be performed over a range of flow rates from 2-25 L/min, depending on the column size being tested. These values correspond to interstitial velocities ranging from approximately 30 cm/min to over 4000 cm/min. These values cover a broad range of velocities which may occur and SVE extraction or bioventing injection wells. Gibson et al. (1993) has observed interstitial velocities around SVE extraction wells over 100 cm/min. For each experimental condition, three tracer injections will be performed at each of the four flow rates tested.

## Data Analysis

Tracer breakthrough curves are evaluated based on residence time distribution analysis. Since two sets of probes are used at either end of the column, two breakthrough curves will be analyzed for each tracer injection. Figure 2 demonstrates a typical double breakthrough curve from experimental results. Mean residence time and variance values for the breakthrough curves will be determined through an analysis of moments. The first moment analysis is used to obtain the mean residence time and is given in discrete form by Levenspiel (1972) as Equation 1:

$$\bar{t} = \frac{\sum t_i S_i \Delta t}{\sum S_i \Delta t} \quad (1)$$

where

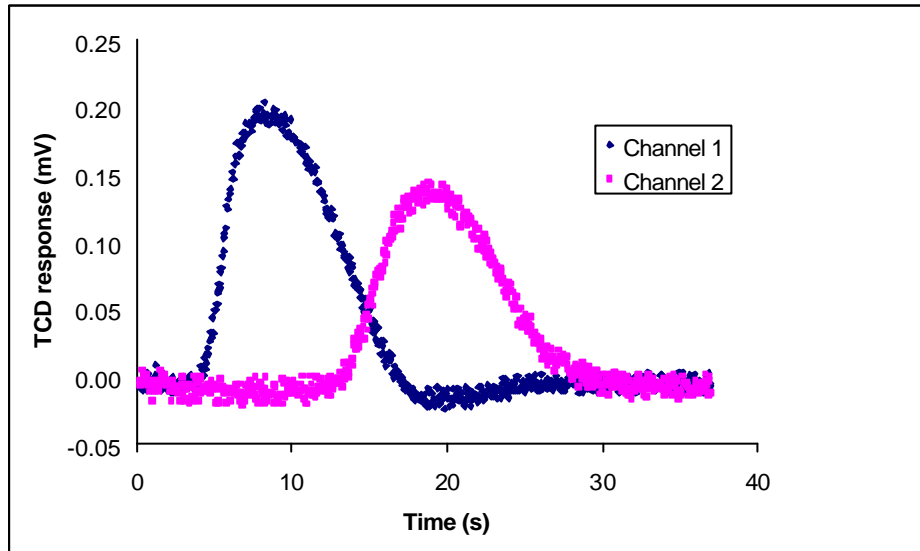
- $\bar{t}$  = mean residence time (s)
- t = time (s)
- S = TCD signal response (mV)

The second moment analysis provides information on the variance of the breakthrough curve and is given in discrete form by Levenspiel (1972) as:

$$s^2 = \frac{\sum t_i^2 S_i \Delta t}{\sum S_i \Delta t} - \bar{t}^2 \quad (2)$$

where

$s^2$  = variance of the break through curve ( $s^2$ )



**Figure 2. Typical double TCD breakthrough curve**

Dispersion coefficient values are obtained through the assumption that the experimental system represents open vessel boundary conditions. For this case, it is assumed that dispersion occurs throughout the media and that flow at the measurement locations is undisturbed. Also Levenspiel (1972) has indicated that if the measurements are taken at least 2 to 3 particle diameters into the bed then the open vessel boundary condition holds closely. Since the sampling probes are located 5 cm from the top and bottom of the soil column, this condition is successfully met.

The additivity property of variances established by Levenspiel (1972) allows for the calculation of dispersion coefficient values by subtracting the difference in variance between measurement points in a single vessel, and creating a modified normalized variance.

The normalized variance may be defined as:

$$\mathbf{s}_q^2 = \frac{\mathbf{s}_{pt2}^2 - \mathbf{s}_{pt1}^2}{(\bar{t}_{pt2} - \bar{t}_{pt1})^2} \quad (3)$$

where

$$\begin{aligned} \mathbf{s}_q^2 &= \text{normalized variance (-)} \\ \mathbf{s}_{pti}^2 &= \text{break through curve variance (s}^2\text{) at points } i = 1, 2 \\ \bar{t}_{pti} &= \text{mean residence time (s) at points } i = 1, 2. \end{aligned}$$

Therefore, based on the assumption of open vessel boundary conditions, the dispersion coefficient may be calculated using Equation 4 (Levenspiel, 1972):

$$\mathbf{s}_q^2 = 2 \frac{D}{vL} + 8 \left( \frac{D}{vL} \right)^2 \quad (4)$$

where

$$\begin{aligned} D &= \text{dispersion coefficient (cm}^2\text{/s)} \\ v &= \text{interstitial velocity (cm/s)} \\ L &= \text{length between measurement points (cm)} \end{aligned}$$

The dispersion coefficient is considered to be comprised of both mechanical mixing and molecular diffusion, represented as:

$$D = av + D_m \quad (5)$$

where

$$\begin{aligned} a &= \text{dispersivity (cm)} \\ D_m &= \text{molecular diffusion coefficient of the pure substance (cm}^2\text{/s)} \end{aligned}$$

The mechanical mixing contribution to the dispersion term is captured by the dispersivity component. Mechanical mixing is considered to be a result of pore scale velocity variations due to pore size distribution, friction along the particle surface and the tortuosity of the paths (Fetter, 1994). The dispersivity value is assumed to remain constant at different flow rates and different lengths. (Beven et al., 1993).

By isolating the mechanical mixing component from the equation the dispersivity value may be obtained as the slope of a best fit line through the calculated dispersion coefficients and associated interstitial velocities.

## **RESULTS and DISCUSSION**

The experimental apparatus and procedure developed as part of Phase I have been designed to further improve the prediction of gas phase dispersion in soils through the use of open vessel boundary conditions. Previous experiments conducted by Gidda et al. (2004) employed a closed boundary condition approach to estimating dispersion values. The experimental apparatus used in the study was limited to only exploring dispersion properties of laboratory prepared (disturbed) soils. Also, it was estimated that more accurate dispersion values may be obtained by switching to an open boundary condition system.

The study by Gidda et al. (2004) was unable to guarantee that all the dispersion occurring in the system was attributed to the soil media. There were some discrepancies as to whether closed vessel boundary conditions accurately predicted the conditions in the experimental column. Glass wool was packed at either end of the column to reduce mixing in the dead space before the tracer entered the soil media. It is expected that some dispersion occurred in the glass wool before and after the tracer left the system. Based on closed boundary condition assumptions, plug flow conditions exist before and after encountering the porous media and therefore the level of dispersion in these areas is approximately zero. Since some dispersion may have occurred in the glass wool, it is not possible to conclude that the dispersion values calculated were solely due to the porous media. Although Gidda et al. (2004) initially attempted to account for the dispersion in the glass wool, it was considered to be of little importance when using soils with greater dispersivity values. The limitations based on the assumed boundary conditions were addressed in the study and it was concluded that an open vessel boundary condition system would be more appropriate for future dispersion experiments. Despite the potential disadvantages of the experimental apparatus, the results presented by Gidda et al. (2004) fell within the range of gas phase dispersion values presented by Constanza-Robinson and Brusseau (2002), Ruiz et al. (1999), and Popovicova and Brusseau (1997).

Currently, the study by Gidda et al. (2004) has been the only investigation into dispersion beyond the simplest of non-uniform porous media. Previous studies have focused primarily on sand (Constanza-Robinson and Brusseau (2002); Ruiz et al. (1999)) or glass bead packing (Popovicova and Brusseau (1997)). Gidda et al. (2004) was also the first to demonstrate that both particle size distribution and moisture content affect the degree of dispersion occurring in the soil.

The experimental results from Gidda et al. (2004) in Table 2 demonstrate a noticeable difference in dispersivity with increasing moisture content, with the exception of the Elora Silt Loam soil at 20% moisture. Constanza-Robinson and Brusseau (2002) also observed an increase in dispersivity with soil moisture content for a sand soil, but only above 15% moisture. Below this value, there appeared to be no change in dispersivity. Therefore it is possible that a non-linear relationship between soil moisture and dispersivity exists for each soil, depending on how the moisture distributes itself within

the pore space as more water is added to the system. It is estimated that small amounts of moisture will have little effect on the available pathways for air flow within the soil. As larger amounts of water is added, larger air filled pore spaces may become blocked or restricted, therefore creating more tortuous pathways within the soil and also increasing the amount of friction experienced by air passing through smaller connected void spaces. The current experimental procedure will investigate this phenomenon by examining the relationship between soil moisture and dispersivity between low moisture and field capacity conditions.

**Table 2. Dispersivity values for air dry and wet soil experiments**

| Soil Type        | Moisture Content | Observed Dispersivity (cm) |
|------------------|------------------|----------------------------|
| Ottawa Sand      | Air Dry          | 0.6                        |
|                  | 3%               | 1.2                        |
| Delhi Loamy Sand | Air Dry          | 1.2                        |
|                  | 20%              | 2.6                        |
| Elora Silt Loam  | Air Dry          | 1.6                        |
|                  | 20%              | 1.1                        |
|                  | 30%              | 1.9                        |
| Brooksten Clay   | Air Dry          | 1.3                        |
|                  | 40%              | 2.4                        |

Phase I of the current research has been limited to only experimental results on air dry Ottawa Sand at this point. Preliminary experiments conducted using the new experimental apparatus developed in this paper has resulted in a calculated dispersivity value of 0.042 cm for air dry Ottawa Sand. This is approximately an order of magnitude lower than observed by Gidda et al. (2004), and estimated to be a result of differences in experimental apparatus. Small changes have been made to the experimental apparatus since these initial test results in order further improve the control on flow rates through the TCD. At this point, additional tests have not yet been conducted to confirm these results.

Difficulties were also encountered with the current experimental apparatus when attempting to measure dispersivity in other air-dried soils. Loose soil particles have tended to plug the sampling probes so that little or no tracer was detected at one or both of the sampling locations upon injection. Therefore, air-dry results appear to be unattainable under the current experimental design, with the exception of Ottawa Sand, without jeopardizing the performance of the detector.

Since laboratory prepared soils no longer retain the structure from which they were collected, very little is known regarding the effects of micro or large scale aggregates on

gas phase dispersion. It is not known whether laboratory scale dispersion results correlate to field conditions. Phase II of the current study is designed to investigate the ability of the gas phase dispersion relationships developed on laboratory prepared soils to predict dispersion in soil core samples. This appears to be the first study of its kind and should provide valuable information for bioventing and SVE modellers attempting to predict the removal efficiency of current or future system designs. Since it has already been shown that current uniform packed bed correlations can seriously under predict dispersion for laboratory prepared soils, it is assumed that these correlations will be even less accurate when applied to field conditions where larger scale non-uniformities exist.

## **CONCLUSIONS**

Research is being completed to fill gaps in the literature regarding gas phase dispersion in unsaturated soils as current SVE and bioventing modelling efforts rely on dispersion correlations for uniform packed bed material or neglect dispersion altogether. This leads to poor predictions as it has been demonstrated that uniform packed bed correlations underestimate gas phase dispersion in non-uniform porous media for both air dry and moist soil conditions. The first phase of the research has been the development of a new experimental apparatus to investigate the effects of particle size distribution and moisture content on a number of soil types. Preliminary experiments have shown comparable results to those in the current literature although further testing of the system is required. The second phase is designed to quantify gas phase dispersion on undisturbed field samples as opposed to controlled laboratory (disturbed) soil conditions. Correlations developed in the first phase of the research will then be compared to results from the second phase in order to further refine the predictive ability of gas phase dispersion. These results will be valuable for those attempting to model field scale SVE and bioventing systems.

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