♻EPA Ground Water Issue

Basic Concepts of Contaminant Sorption at Hazardous Waste Sites

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Introduction

The Regional Superfund Ground Water Forum is a group of ground-water scientists, representing EPA's Regional Superfund Offices, organized to exchange up-to-date information related to ground-water remediation of Superfund sites. One of the major issues of concern to the Forum is the transport and fate of contaminants in soil and ground water as related to subsurface remediation. Processes which influence the behavior of contaminants in the subsurface must be considered both in evaluating the potential for movement as well as in designing remediation activities at hazardous waste sites. Such factors not only tend to regulate the mobility of contaminants, but also their form and stability. Sorption is often the paramount process controlling the behavior of contaminants in the subsurface. This paper summarizes the basic concepts of sorption in soil and ground water with emphasis on nonpolar organic contaminants.

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The Concept of Sorption

Sorption can be defined as the interaction of a contaminant with a solid. More specifically, the term can be further divided into adsorption and absorption. The former refers to an excess contaminant concentration at the surface of a solid while the latter implies a more or less uniform penetration of the solid by a contaminant. In most environmental settings this distinction serves little purpose as there is seldom information concerning the specific nature of the interaction. The term sorption is used in a generic way to encompass both phenomena.

There are a number of factors which control the interaction of a contaminant and the surface of soil or aquifer materials. These include chemical and physical characteristics of the contaminant, composition of the surface of the solid, and the fluid media encompassing both. By gaining an understanding of these factors, logical conclusions can often be drawn about the impact of sorption on the movement and distribution of contaminants in the subsurface. The failure to take sorption into account can result in a significant underestimation of the amount of a contaminant at a site as well as the time required for it to move from one point to another.

In introducing sorption theory it is necessary to define the terms sorbate and sorbent. The sorbate is the contaminant that adheres to the sorbent, or sorbing material. In this discussion the sorbate will usually be an organic molecule and the sorbent will be the soil or aquifer matrix.

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Factors Influencing Sorption

The properties of a contaminant have a profound impact on its sorption behavior. Some of these include:

- Water Solubility
- Polar/Ionic Character
- Octanol/Water Partition Coefficient
- Acid/Base Chemistry
- Oxidation/Reduction Chemistry

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Contaminant Characteristics

In discussing sorption it is useful to divide chemicals into three groups. Although there are many ways to divide chemicals into subgroups, for this purpose three categories are presented which transcend normal boundaries between inorganic and organic species. These are: (1) ionic or charged species; (2) uncharged polar species; and, (3) uncharged nonpolar species.

Most inorganic chemicals in aqueous solution will occur as ionic or charged species. This applies to metals and metalloids, and to other molecules such as cyanide and ammonia. However, in contaminated water, metals and other inorganic constituents can exist as polar or nonpolar neutral species. In any event, the chemical form of a contaminant will have a profound effect on its sorption and, therefore, its environmental mobility.

Organic contaminants have representatives in all three of the sorption categories. Many of the more common organic ground-water contaminants are of the nonpolar species, including trichloroethene (TCE), tetrachloroethene (PCE), the chlorinated benzenes, and the more soluble components of hydrocarbon fuels such as benzene, toluene and xylene. Other important organic contaminants including many of the pesticides, phenols and dyes exist in solution as either charged or polar molecules.

Still other, larger organics, such as surfactants, can have both polar and nonpolar ends within the same molecule. The environmental mobility of contaminants with these distinctive properties has been less thoroughly studied than nonpolar organics; therefore, site-specific investigations may provide the most reliable information for their transport characteristics.

Soil Characteristics

If one avoids the difference between positive and negative charges, a simple rule of sorption might be: for charged species, "opposites attract" and for uncharged species, "likes interact with likes." Likes refers to the three categories of contaminants and to the properties of the soil matrix. Some of the most important characteristics of soil affecting the sorptive behavior of subsurface materials include:

- Mineralogy
- Permeability/Porosity
- Texture
- Homogeneity
- Organic Carbon Content
- Surface Charge
- Surface Area

Soil, in its natural state, is primarily composed of sand, silt, clay, water, and a highly variable amount of natural organic carbon. The latter profoundly complicates a soil's sorptive properties. The combination of these characteristics describes the surfaces offered as sorptive sites to contaminants in water passing through the subsurface matrix. For example, silts and clays have much higher surface areas than sand, usually carry a negative charge, and almost invariably associate with natural organic matter.

It can be deduced that sandy materials offer little in the way of sorptive surfaces to passing contaminants while silts and clays, particularly those having substantial amounts of organic matter, provide a rich sorptive environment for all three categories of contaminants. Even the most porous and highly productive aquifers, composed of sands and gravels, usually have some fine grained material, and a few percent of silts and clays can result in a substantial increase in the sorptive behavior of the aquifer material.

Fluid Media Characteristics

Under most contamination situations the primary transporting fluid is water. One of the most important properties of this solvent phase is pH for it dictates the chemical form and, therefore, the mobility, of all contaminants susceptible to the gain or loss of a proton. As an example, pentachlorophenol will primarily be an uncharged polar molecule in an aqueous solution whose pH is below about 4.7 and an anion when the pH is above that value, increasing its solubility from 14 to 90 mg/l.

Other characteristics of water that can influence the behavior of contaminants include the salt content and the dissolved organic carbon content. Chlorides, for example, which are not usually of much concern when dealing with organic contaminants, can have an important effect on the mobility of various metals. Dissolved organic matter, at relatively high concentrations found in many leachates, has a significant effect on the mobility of most nonpolar organics.

Implications of These Characteristics

Although somewhat simplified, it can be assumed for purposes of this discussion, that charged and polar species tend to interact with charged and polar surfaces, and nonpolar compounds interact with nonpolar components of soil, usually the natural organic carbon. In order to make a first estimate of the significance of sorption at a site, it is necessary to determine the polar and nonpolar nature of the material with which the contaminant will come into contact. This is usually done by measuring the cation exchange capacity and the natural organic carbon content, respectively.
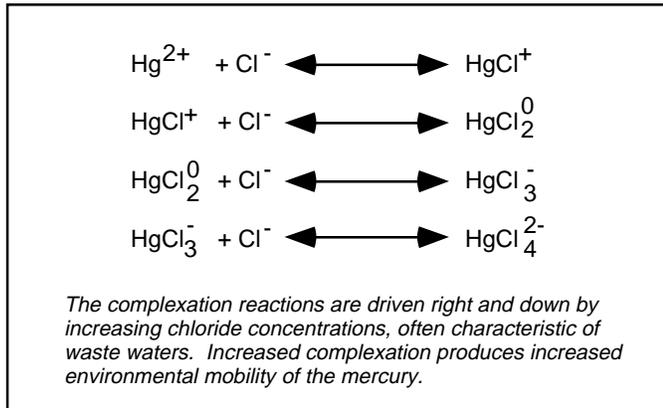
The cation exchange capacity (CEC) provides an estimate of the total negatively-charged sites on the surface of the soil. It is determined by measuring the mass of a standard cation, usually ammonia, that displaces another cation held by the soil. Under normal field conditions these sites will be occupied by cations common to the flowing or percolating water, such as Na^+, K^+, Ca^{2+}, and Mg^{2+}. Larger organic cations and highly-charged metal ions like Hg^{2+} or Cr^{3+} will be preferentially retained at these sites by "exchanging" with their normal occupants. Thus large organic cations and heavy metals would not normally be expected to move far through soils with a measurable cation exchange capacity.

At contaminated sites, however, conditions may not be "normal" and Hg^{2+} may be codisposed with high levels of chloride salts. In the complexation chemistry shown in Figure 1, Hg^{2+} may be replaced by the neutral complex $HgCl_2$ or the negative ion $HgCl_3$, both of which move through the soil more quickly than the cationic form.

Sorption of Nonpolar Organics

As mentioned above the chemicals at many contaminated sites are nonpolar organics. It was representatives of these types of compounds (DDT and other chlorinated hydrocarbon pesticides)

that first focused attention on the potential hazards of chemicals in the environment because of their widespread use, potential human toxicity, and recalcitrance.

$$Hg^{2+} + Cl^- \longleftrightarrow HgCl^+$$

$$HgCl^+ + Cl^- \longleftrightarrow HgCl_2^0$$

$$HgCl_2^0 + Cl^- \longleftrightarrow HgCl_3^-$$

$$HgCl_3^- + Cl^- \longleftrightarrow HgCl_4^{2-}$$

The complexation reactions are driven right and down by increasing chloride concentrations, often characteristic of waste waters. Increased complexation produces increased environmental mobility of the mercury.

Figure 1. Mercury Ion Complexation in Chloride-Rich Water

Transport and fate characteristics of these compounds have been well studied, first by the agricultural community and later by environmental scientists. As a result, an understanding of the sorptive behavior of these compounds has evolved which can be used to assess the environmental consequences posed at a waste disposal site.

Many organics of environmental concern have a limited solubility in water because of their nonpolarity and molecular size: that is, the solubility of an organic contaminant decreases with decreasing polarity and increasing molecular size. But even with limited solubilities, many hazardous chemicals at equilibrium are at measurable, and sometimes toxic concentrations in water. Polar molecules, such as ethanol, are compatible with water. Their combination results in a homogeneous solution regardless of the proportions that are mixed.

Nonpolar organic compounds interact with soil organic matter through a process known as "hydrophobic sorption" which can be explained as the affinity of organic compounds for phases other than water. For example, water being a polar molecule is not compatible with other nonpolar molecules, such as DDT, which is immiscible with water.

Octanol-Water Partitioning

Organic molecules of increasing size, decreasing polarity and therefore water solubility, are said to exhibit increasing "hydrophobicity" which can be quantified by their octanol-water partition coefficient. It is a measure of the distribution of the chemical between a water and an organic (octanol) phase with which it is in contact. The more hydrophobic the contaminant, the more likely it is to partition into the octanol phase. The partition coefficient provides a fairly accurate understanding of the sorptive process occurring between water and the soil, more specifically, the soil organic matter.

The octanol-water partition coefficient, expressed as K_{ow} in Figure 2, is determined by measuring the concentration of a particular compound in the water and the octanol phases after a period of mixing. It is important to note that the more hydrophobic the compound the less accurate the test, and the results should

Octanol-Water Partition Coefficient:

$$K_{ow} = \frac{Concentration_{Octanol}}{Concentration_{Water}}$$

Almost always presented as Log_{10} because the numbers are so large for hydrophobic compounds.

Sorption Coefficient:

$$K_p = \frac{Concentration_{Solid\ Phase}}{Concentration_{Solution}}$$

Units are $\dfrac{mg/kg}{mg/L}$ *, which is L/kg.*

Carbon Normalized Sorption Coefficient:

$$K_{oc} = \frac{Sorption\ Coefficient,\ K_p}{Fraction\ Organic\ Carbon}$$

Figure 2. Relationships Pertinent to Nonpolar Organic Contaminant Transport

be viewed accordingly. It is often sufficient to know that an extremely high coefficient means that the compound is very hydrophobic. Since measured K_{ow} values can be in the millions for important environmental contaminants (PCB's, chlorinated pesticides, dioxins and furans), they are often expressed as the base10 logarithm, Log K_{ow}.

The K_{ow} has two attributes that make it especially useful in environmental assessments. First, it varies in a predictable way within classes of organic compounds. For example, as shown in Figures 3 and 4, if K_{ow} is known for one member of a class of compounds it can be used reasonably well to estimate a value for other members of the same family. In the examples shown, the K_{ow} can be correlated to the number of chlorine atoms or the number of rings in the molecular structure of classes of contaminants.

The second attribute results from the work of a number of agricultural and environmental researchers who correlated sorption on the organic matter of soils with the K_{ow} of the compounds involved. By using these attributes of the K_{ow}, it is possible to estimate the potential sorption of organic contaminants based on the structure of the compounds and the organic carbon content of the soil or aquifer material.

Sorption to Soils

Thus far it has been suggested that nonpolar organic compounds are sorbed by soils as a function of their hydrophobicity (K_{ow}) and the organic carbon content of the soil.   There has been considerable research which suggests that the slow kinetics of the sorption process may be significant in swiftly moving ground water. Sorption studies using flow-through columns produce results sensitive to the flow rate, and batch tests indicate that
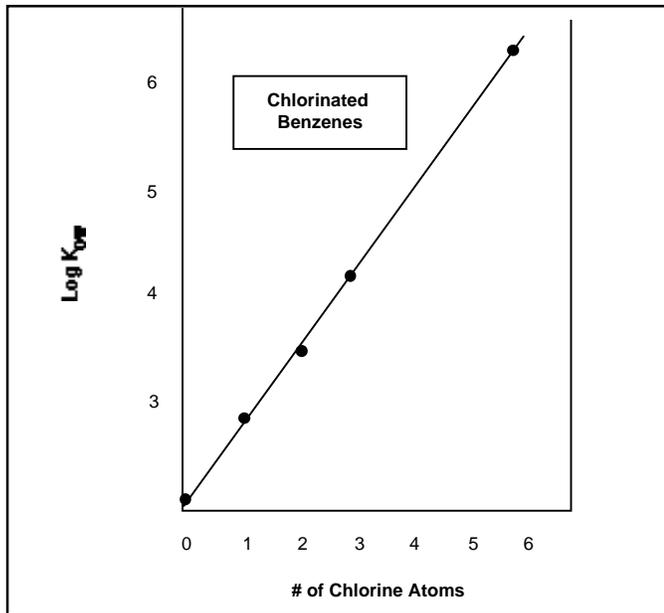
Figure 3. Relationship of Molecular Structure to Hydrophobic Character

contaminants associated with the solid phase to that in solution, and is, therefore, conceptually similar to K_{ow}. The usefulness of K_{ow} in estimating sorption stems from the fact that the soil organic matter serves the same function as octanol in the octanol-water test. As a result, there have been many empirical relationships developed for estimating sorption from the K_{ow} and the soil organic carbon content. One expression, developed in the laboratory by Piwoni and Banerjee, 1989, for the sorption of common environmental contaminants with a low aquifer organic carbon, is:

$$Log\ K_{oc} = 0.69\ Log\ K_{ow} + 0.22$$

When applying such a relationship, it is important to select a study in which the compounds used are similar to those of interest at the site under investigation. However, as shown in Figure 5, even when applying the empirical relationship to a structurally dissimilar compound such as anthracene, if it is a nonpolar organic, the error of estimate should be less than a factor of five.

These estimates of sorption are based, in large measure, on a good evaluation of the soil organic carbon content at a site which is obtained from the degradation of naturally occurring organic matter. In this regard it is important to realize that soils and aquifer materials are very heterogeneous and the organic carbon content can vary considerably both in the vertical and horizontal dimension. Fortunately, this variability tends to be the greatest in the vertical soil profile while most site investigations are concerned with contaminant movement in the ground water away from the source. While the soil organic carbon content in the horizontal plane usually differs by a factor of ten or less, it can vary by a factor of 10 to 100 in the vertical dimension.
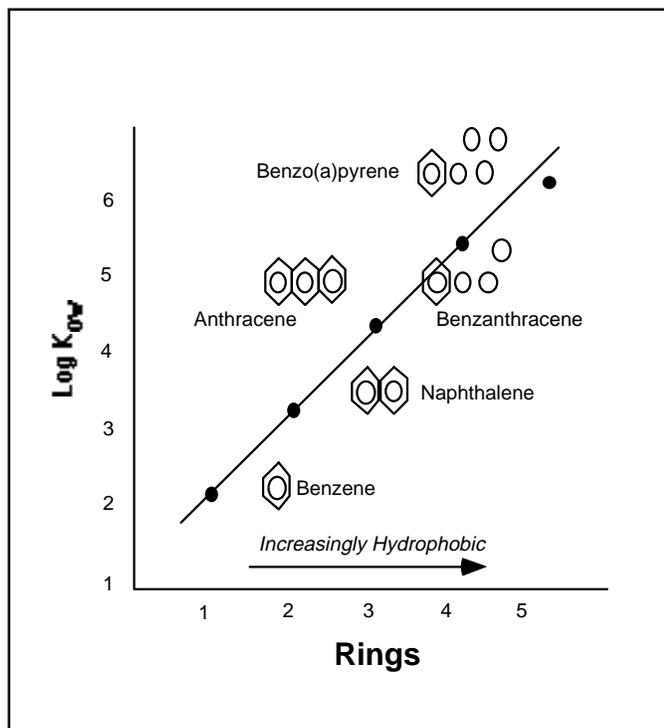
increased sorption occurs with longer exposure times. The practical implication of these findings may be that sorption is overestimated in aquifer systems with relatively high flow rates.

Sorption is expressed in terms of a partition coefficient K_p, which is defined in Figure 2 as the ratio of the concentration of

Figure 4. Relationship of Molecular Structure to Octanol Water Partition Coefficient
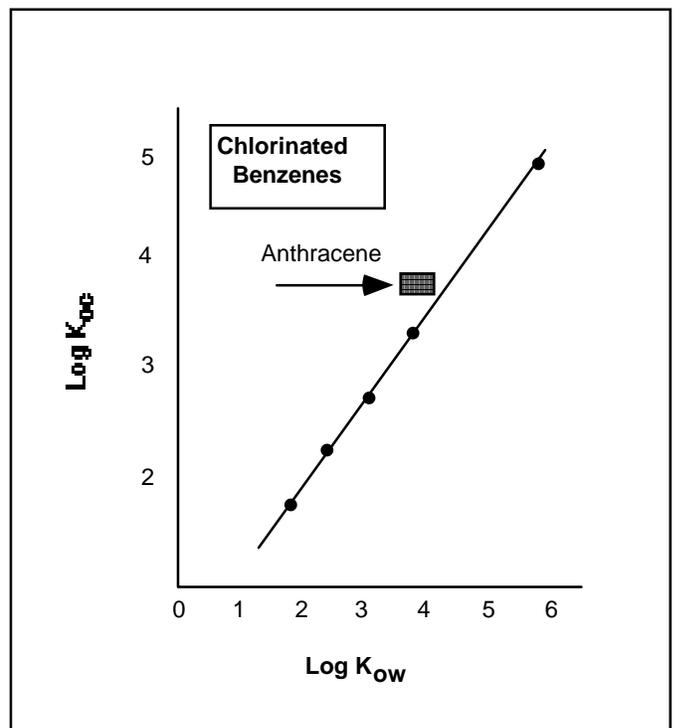

Figure 5. Partitioning on Soil Organic Carbon as Function of Octanol-Water Partition Coefficient

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In order to determine the soil organic carbon content at a site, samples are usually obtained using split spoon sampler or other standard soil sampling devices. Representatives portions of the soil are then burned in an O_2 atmosphere and the produced CO_2 is measured by IR spectrophotometry. Before burning soil samples must be acidized to remove inorganic carbon. The accuracy of measuring organic carbon content can also be questionable, particularly at low levels and in carbonate soils. Existing analytical methods for measuring soil organic carbon were developed for the higher concentrations found near the surface. Therefore, at the low levels found in deeper soils and ground water, the same quality assurance procedures used in determining contaminant levels in water should be followed in determining the subsurface organic carbon content.

The processes driving hydrophobic sorption are nonspecific and depend upon small amounts of energy gained by moving contaminants out of the aqueous phase. The extent to which the process proceeds is dependent upon how receptive the soil matrix is to the organic molecule, which is a function of the organic content. But even when the organic carbon content is very low, some sorption of the most hydrophobic molecules continues because of the soil's mineral surfaces.

Sorption Estimation

In order to use the information provided above in estimating the amount of a contaminant associated with the aqueous and solid phases of an aquifer, it is necessary to develop a contamination scenario. To that end it is assumed that the contaminant at an industrial landfill is 1,4-dichlorobenzene, and there is sufficient data to indicate that: (1) most of the contamination is below the water table; (2) the contaminant concentration in ground water averages 1 mg/l; (3) the measured soil organic carbon is 0.2 percent; and (4) the pore water occupies 50 percent of the aquifer volume. Steps leading to an estimate of the contaminant's distribution between the aqueous and solid phases are:

Field Measurements:

Average contaminant concentration in monitoring wells = 1.0 mg/l

Soil organic carbon = 0.2 percent, therefore f_{oc} = 0.002

Pore water occupies 50 percent of the aquifer's volume.

From The Literature:

Log K_{ow} (1,4-dichlorobenzene) = 3.6

Piwoni and Banerjee Regression, Log K_{oc} = 0.69 K_{ow} + 0.22

Calculated:

Log K_{oc} = 0.69(3.6) + 0.22 = 2.70

therefore: K_{oc} = 506

$K_p = K_{oc}(f_{oc}) = 506 (0.002) = 1.0 = \dfrac{\text{Sorbed C}}{\text{Solution C}}$

Conclusion;

The contaminant, equally distributed between each phase, is expressed as mg/kg (soil) and mg/l (water). Since soil is about 2.5 times more dense than water, 2 liters of aquifer would contain 1 liter of water and 2.5 kg of soil. Therefore, 1.0 mg/l of the contaminant would be associated with the water and 2.5 mg (70 percent) would be sorbed to the aquifer's solid phase.

As can be seen from this example, sorption tends to complicate remediation techniques that require pumping water to the surface for treatment. The desorption process has kinetic constraints that can render a pump-and-treat system ineffective. Slow desorption kinetics result in progressively lower contaminant concentrations at the surface, and less cost-effective contaminant removal. It is not uncommon to pump a system until the contaminant concentration in the pumped water meets a mandated restoration level, while the aquifer's solid phase still contains a substantial mass of contaminant. If the pumps are turned off, concentrations in the ground water will soon return to their equilibrium level.

Measuring Sorption

It is preferable to obtain the best information possible on which to base an estimate of sorption. Therefore, tests should be made with the contaminants of concern, as well as soils and aquifer material from a specific site. The goal is to obtain a partition coefficient, K_p, for use in the prediction of contaminant movement.

There are essentially two methods for measuring the partition coefficient, those being batch and dynamic techniques. Batch techniques are quicker and easier to perform and, therefore, more amenable to replication and quality control. Dynamic or flow through techniques offer the advantage of more closely representing processes occurring in the field.

The standard approach to determine the partition coefficient is to generate a sorption isotherm, a graphical representation of the amount of material sorbed at a variety of solute concentrations. The Freundlich isotherm, $S = K_p C^{1/n}$, is the representation most often used for the sorption of nonpolar organics to soils and aquifer materials. In this equation, S is the mass sorbed per mass of sorbent (mg/kg), C is the solute concentration at equilibrium (mg/l), K_p is the Freundlich partition coefficient, and 1/n is a fitting factor. The equation can be expressed in a linear form for convenience:

$$\text{Log } S = \text{Log } K_p + 1/n \text{ Log } C$$

As shown in Figure 6, Log K_p can be estimated by determining the intercept of the regression of a Log-Log plot of S and C.

Summary

This has been a discussion of the concepts involved in estimating contaminant sorption, particularly nonpolar organics, at hazardous waste sites. After determining the types of contaminants present at a site, it is possible to estimate K_p using K_{ow} values from the literature, an appropriate sorption coefficient/K_{ow} regression equation, and some organic carbon values.

If sorption determinations are within the scope of the project, site representative soil samples and contaminants should be selected
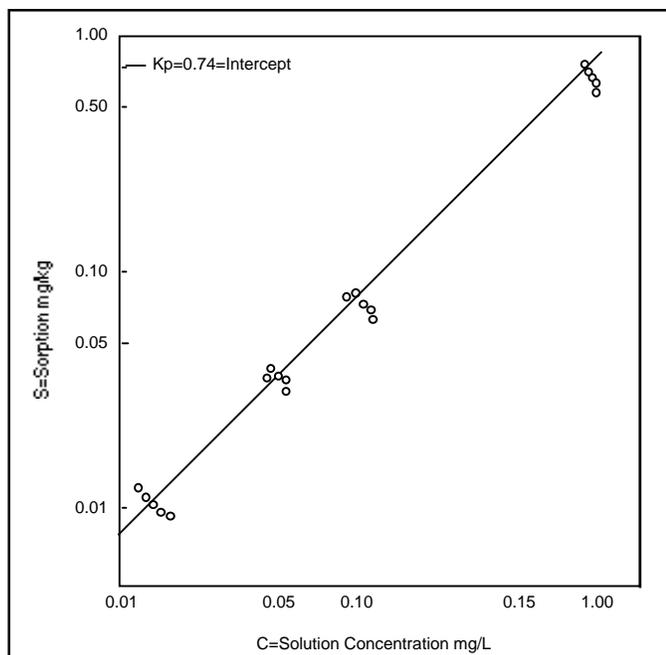
Figure 6. Sorption of 1,4-Dichlorobenzene

from the tests. The measured sorption information is best used to evaluate the validity of preliminary estimates. If the measured partition coefficients differ from the estimates by more than a factor of 2 or 3, it may be useful to select other contaminants from the site and determine K_p values the same soil samples. A plot of K_p values versus K_{ow} values will provide a useful guide for predicting the sorption characteristics of other contaminants at the site.

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