

# Numerical Modeling of Benzene Transport in Andosol and Sand: Adequacy of Diffusion and Equilibrium Adsorption Equations

Ping Du\*, Masaki Sagehashi, Akihiko Terada, Masaaki Hosomi

**Abstract**—Prediction of benzene transport in soil and volatilization from soil to the atmosphere is important for the preservation of human health and management of contaminated soils. The adequacy of a simple numerical model, assuming two-phase diffusion and equilibrium of liquid/solid adsorption, was investigated by experimental data of benzene concentration in a flux chamber (with headspace) where Andosol and sand were filled. Adsorption experiment for liquid phase was performed to determine an adsorption coefficient. Furthermore, adequacy of vapor phase adsorption was also studied through two runs of experiment using sand with different water content. The results show that the model adequately predicted benzene transport and volatilization from Andosol and sand with water content of 14.0%. In addition, the experiment additionally revealed that vapor phase adsorption should be considered in diffusion model for sand with very low water content.

**Keywords**—Benzene; Transport Model; Adsorption; Soil Contaminant.

## I. INTRODUCTION

THE VOCs transport in soil and volatilization from soil to the atmosphere is important for the preservation of human health and management of contaminated soils. Given the previous reports that VOCs transport is governed mostly by diffusion [1-5], it is of importance to track the behavior of VOC transport in soil. Benzene was chosen as a representative VOC in this study because of its volatility, high toxicity, and high abundance at contaminated sites [6].

To quantify harmful influence of benzene to the environment and to determine a treatment strategy, development of a physically-based numerical model to describe benzene transport in soil and the soil-air interface is one of appropriate approaches [7]. Olesen et al. [8-9] suggest that the effective transport rates of organic chemicals are governed partly by diffusion and/or

adsorption in soil gas and liquid phases. In recent years, it has been reported that liquid phase sorption onto soil organic matter is likely to occur in case of many chemicals and soil types [8, 10, 11-12]. However, the liquid phase sorption isotherm in soil with low organic matter is poorly understood, despite a few studies indicating that linear isotherm is not adequate [13]. Therefore, the adequacy of linear and nonlinear adsorption isotherm for description of benzene transport in two types of soil (i.e. Andosol and sand) was investigated in this study. Moreover, Chiou et al. have proposed that soil as a binary solvent whose components interact with VOCs by different mechanisms: the mineral material and the organic matter. Adsorption of VOCs in mineral is competitively governed by water molecules [3, 14]. To predict benzene transport in soil, the study about the influence of water on adsorption of benzene in mineral or soil with low organic matter (i.e. sand) should be performed.

There have been a few studies observing VOCs adsorption in soil and volatilization simultaneously. These studies have been conducted with a small column experiment or diffusion cell [15-17], however, this cannot necessarily be applicable on site if the design is overly idealized or simplified [18, 19]. A flux chamber used in this study, allowing for simultaneous observation of a benzene concentration profile in soil and the flux from soil surface to the atmosphere, is a promising apparatus to reproduce a real condition on site and, hence, allows one to elucidate the impact of benzene to the environment and humans. Therefore, the objectives of this study were two-fold: to clarify the predictability of a model of benzene transport in soil and volatilization from soil to air; and to investigate the influence of water content on benzene adsorption and diffusion in soil with low organic matter.

## II. MATERIALS AND METHODS

### *Soil Preparations and Properties*

Two kinds of soil were used in the experiment: Andosol (Tachikawa Heiwa Nouen Co. Ltd., Japan) with water content of 15% and sand soil with water content of 0.4% and 15%. After removal of plant residues, the particulates were air-dried until the desired water contents were obtained. Then each particulate was hammered and sieved with a 4 mesh sieve to obtain homogeneous particles. The physical properties are shown in TABLE 1.

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### Chemicals

Benzene with purity of 99.8% was supplied by Kanto

TABLE I  
PHYSICAL PROPERTIES OF THE SOIL USED IN THE STUDY

Property	Andosol	Sand (1)	Sand (2)
Water content, %	16.0 <sup>a</sup> (15.0)	13.2 <sup>a</sup> (15.0)	0.4 <sup>a</sup> (0.0)
Bulk density, $\rho_b$ , g cm <sup>-3</sup>	0.762	1.539	1.550
Air-filled porosity, $\epsilon_a$ , cm <sup>3</sup> cm <sup>-3</sup>	0.533	0.338	0.420
Water-filled porosity, $\epsilon_w$ , cm <sup>3</sup> cm <sup>-3</sup>	0.162	0.190	0.006
Total porosity, $\epsilon_T$ , cm <sup>3</sup> cm <sup>-3</sup>	0.695	0.428	0.426
Particle density, $\rho_p$ , g cm <sup>-3</sup>	2.5	2.7	2.7
Organic carbon content, %	9.5	1.2	1.2
Specific surface area (N <sub>2</sub> , BET), cm <sup>2</sup> g <sup>-1</sup>	26.563 <sup>b</sup>	7.769 <sup>b</sup>	7.769 <sup>b</sup>

<sup>a</sup> Numbers represent the measured value before the experiment.

<sup>b</sup> Determined by Surface Area Analyzer and Pore Size Analyzer (QUADRASORB™ SI, v.1.0, QUANTACHROME INSTRUMENTS, USA).

Chemical Co., INC (Kanagawa, Japan). Carbon disulfide was supplied by Wako Pure Chemical Industries, Ltd (Osaka, Japan).

### Chamber Experiments

In this study, a flux chamber (shown in Fig. 1) was used. The chamber was made of propylene with the size of 300 mm×290 mm×175 mm (volume of 15225 cm<sup>3</sup>) according to the standard of JIS (Japanese Industrial Standards) A 1901 (Objective: Determination of the emission of volatile organic compounds and aldehydes for building products-Small chamber method).

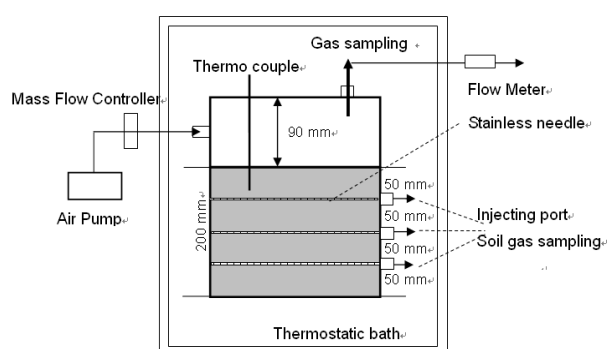


Fig.1 Schematic diagram of the flux chamber

In all experiments, soil was packed into the chamber up to height of 200 mm (volume of 10500 cm<sup>3</sup>). The chamber was equipped with four sampling ports, one at the top was used for headspace gas sampling, whereas the others were on the side

wall at 5 cm intervals for soil interstitial gas sampling, respectively. Each sampling port on the side wall installed a cylindrical Teflon septum holder with an inner diameter of 7 mm and a length of 10 mm. A silicone septum (GL. Science Inc., Tokyo, Japan) was placed into each septum holder. A perforated stainless steel needle (30 cm length, 1 mm i.d. with 10 holes at about 3 cm intervals) was inserted through the septum of each sampling port. The outlet of a needle was plugged with a silicone septum (GL. Science Inc., Tokyo, Japan), allowing the insertion of a gas-syringe for gas sampling.

Benzene was injected into soil with the chamber completely sealed. A syringe (Agilent, German) with a long needle (30 cm) was horizontally inserted for benzene injection through each sampling port on the side wall. Benzene was injected at different positions to provide homogeneous environment in the chamber. During the experiment, benzene vapor in the headspace and soil layers at the three locations was periodically sampled. Dried air from an air pump was supplied from the inlet, located at 45 mm above the soil surface, and flowed through the outlet after benzene vapor was homogeneously distributed in soil. The air flow rate was controlled by a mass flow controller (SEC-B40, HORIBA STEC, Kyoto, Japan) and monitored by a flow meter (Wet Gas Flow, Sinagawa Corp., Tokyo, Japan) placed at the outlet of the chamber. The end of the vent pipe was connected into a ventilating cabinet. A micromanometer (PG-100-103GP, COPAL ELETRONICS, Tokyo, Japan) was used to measure a pressure in the chamber, and no measurable pressure gradient was observed. In all experiments, the chamber was placed in a thermostatic bath to keep constant temperature at 25°C despite a slight fluctuation (TABLE2).

### Sample Analysis

TABLE II  
EXPERIMENTAL CONDITIONS APPLIED IN THIS STUDY

Run	Soil	Initial benzene conc., mg kg <sup>-1</sup>	Soil water content, %	Flow rate, ml/min	Actual temperature (°C)	Experimental period (days)
1	Andoso 1	50	16.0	690	25±2	6
2	Sand	50	13.2	690	25±2	6
2	Sand	50	0.4	600	25±2	6

Gas samples (0.5 ml in volume) were analyzed by a GC-FID (GC-14A, Shimadzu, Japan) with an Agilent HP-1 MS column. The GC was operated at isothermal temperature of 50°C. Injection port and detector temperatures were 150°C and 220°C, respectively.

### Adsorption Experiment

The experiment was performed after the study by Rogers et al. [20]. Five solution concentrations of benzene were used for the adsorption study. These concentrations were 1, 10, 100, 1000, and 10000 µg l<sup>-1</sup> of benzene in 25 ml of liquid. The concentration of benzene in liquid was determined by the Headspace Sampler-GC-MS (HP7694, Agilent, German), and

the amount of benzene adsorbed in soil was determined by the GC-FID. Adsorption-partition constants were determined by use of the following log form of the Freundlich equation:

$$\log(x/m) = (1/n)(\log C) + \log K \quad (1)$$

where  $x$  is the weight of benzene adsorbed ( $\mu\text{g}$ ),  $m$  is the weight of adsorbent ( $\text{kg}$ ),  $C$  is the equilibrium concentration of benzene in solution ( $\mu\text{g l}^{-1}$ ),  $K$  and  $n$  are the Freundlich capacity factor and intensity parameter, respectively.

### III. TWO-PHASE DIFFUSION MODEL

#### Model Theory

The one-dimensional conservation of mass balance equation with no chemical or biological reaction for benzene, without gas and liquid advection, takes the form of the following equation:

$$\frac{\partial C_T}{\partial t} + \frac{\partial J_S}{\partial z} = 0, \text{ and } J_S = -D_s^g \frac{\partial^2 C_g}{\partial z^2} - D_s^l \frac{\partial^2 C_l}{\partial z^2} \quad (2)$$

where  $C_T$  is the total chemical concentration ( $\text{g cm}^{-3}$ );  $J_s$  is the benzene flux ( $\text{g cm}^{-2} \text{s}^{-1}$ );  $t$  is time (s); and  $z$  is the soil depth (cm),  $D_s^g$  is the benzene gas diffusion coefficient in a particulate layer ( $\text{cm}^2 \text{s}^{-1}$ ),  $D_s^l$  is the benzene liquid diffusion coefficient in a particulate layer ( $\text{cm}^2 \text{s}^{-1}$ ),  $C_l$  and  $C_g$  are the benzene concentrations in the liquid and gas phases ( $\text{g cm}^{-3}$ ), respectively.  $C_T$  is given by the following equation:

$$C_T = \rho_b C_s + \varepsilon_w C_l + \varepsilon_a C_g \quad (3)$$

where  $C_s$  is the adsorbed concentration ( $\text{g g}^{-1}$ ) from the liquid phase and it is described by linear or Freundlich equation:

$$C_s = K \cdot C_l, C_s = K \cdot C_l^{1/n} \quad (4)$$

where  $K$  and  $n$  was determined by adsorption experiment in this study. Henry's law was used to state the equilibrium distribution between gas and liquid phases. The value of Henry constant  $K_H$  (-) was taken as 0.224 [21].

Millington-Quirk model [22] was employed to obtain the soil-gas diffusion coefficient and soil-liquid diffusion coefficient:

$$D_s^g = \frac{\varepsilon_a^{10/3}}{\varepsilon_T^2} D^g, D_s^l = \frac{\varepsilon_w^{10/3}}{\varepsilon_T^2} D^l \quad (5)$$

where  $D_s^l$  and  $D_s^g$  are the molecular diffusion coefficient of benzene in bulk water and air, respectively ( $\text{cm}^2 \text{s}^{-1}$ ). The values of  $9.6 \times 10^{-2} \text{ cm}^2 \text{s}^{-1}$  and  $1.02 \times 10^{-5} \text{ cm}^2 \text{s}^{-1}$  were taken from the literature [23].

#### Diffusion Equation in Headspace

Benzene diffusion in the headspace was assumed to merely depend on gas phase diffusion, and the diffusion equation can be written as:

$$\frac{\partial C_g^H}{\partial t} = D^g \frac{\partial^2 C_g^H}{\partial z^2} \quad (6)$$

where  $C_g^H$  is the gas phase concentration in the headspace,

$D^g$  is the benzene gas diffusion coefficient in the headspace, and  $z$  is the height from the topsoil (cm).

#### Model Calculation

Under an ideal condition, the gas phase concentration in the whole chamber reaches a constant value  $C_e$  at the beginning of the experiment, because it is assumed that vapor transportation is governed by Fick's law. Therefore, the initial condition of the second stage can be described as:

$$C_g^H = C_e, L < z \leq H + L \quad (7)$$

$$C_T = C_e, 0 \leq z \leq L \quad (8)$$

where  $H$  is the height of the headspace (cm), and  $L$  is the depth of soil (cm).

The mass balance equations were solved by simultaneous ordinary differential equations. The calculation with assumption of instantaneous adsorption was performed with Visual Basic (Express Version) at a time step of 0.1 s.

## IV. RESULTS AND DISCUSSION

#### Liquid adsorption for two soils

Benzene adsorption on Andosol and sand was determined after 30 h of adsorption. The plots of the Freundlich isotherms are shown in Fig. 2, and Freundlich constants were determined for two kinds of soils. For Andosol, the  $n$  value is very close to 1, indicating that sorption should be linear within the range of increasing benzene concentrations used in this study. On the contrary, the  $n$  value of 1.11 for sand indicates that sorption isotherm of benzene on sand should be nonlinear. The difference should be explained by the organic carbon content and specific surface area of two kinds of soil (TABLE1). It has been reported by many studies that VOCs sorption in soil with high fraction of organic matter should be expressed as linear regression [24]. In contrast, the nonlinear adsorption behavior is much more typical for soils with low contents of organic matter, because of condensation in the soil pores and/or multilayer sorption, resulting in that the number of sorption sites on the surface of soil sorbent is limited [14].

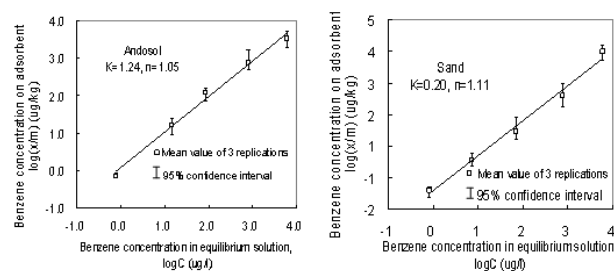


Fig. 2 Freundlich plot for benzene on Andosol (upper figure) and sand (lower figure).

#### Adequacy of the model with linear adsorption for Andosol

Measured and calculated benzene concentrations in the headspace and three soil layers for Andosol are shown in Fig. 3.

The dashed lines in the figure are the calculation curves by the model. Calculations based on linear (the left figure) and nonlinear adsorption isotherms (the right figure) were demonstrated. The sorption constants determined by the adsorption experiment was used for the calculation.

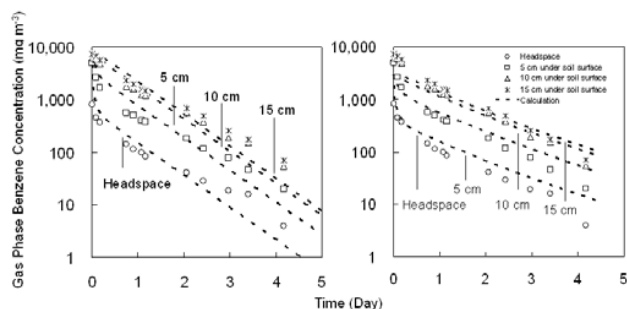


Fig. 3 Measured and calculated benzene concentrations in gas phase (Run 1, filled with Andosol)

Overall the model well predicts the experimental trend in both cases, i.e. linear and nonlinear adsorption isotherm, indicating that a simple two-phase diffusion model with linear adsorption equation was adequate in predicting benzene diffusion in Andosol. Given the empirical equation of  $K = K_{oc} \cdot f_{oc}$  ( $K_{oc}$  is the organic carbon partition coefficient,  $\text{cm}^3 \text{g}^{-1}$ , and the  $f_{oc}$  is the organic carbon fraction of soil),  $K_{oc}$  was calculated as  $13 \text{ cm}^3 \text{g}^{-1}$ . The value is almost 6 times lower than the value ( $K_{oc}=80 \text{ cm}^3 \text{g}^{-1}$ ) reported by Jury et al. [25], but close to the value ( $K_{oc}=23 \text{ cm}^3 \text{g}^{-1}$ ) reported by Voudrias and Li [10]. These differences, however, are within the expected accuracy of the empirical equation.

#### Adequacy of the model with nonlinear equilibrium adsorption for Sand

Time course of measured and calculated gas phase benzene concentrations in Runs 2-3 is shown in Fig. 4.

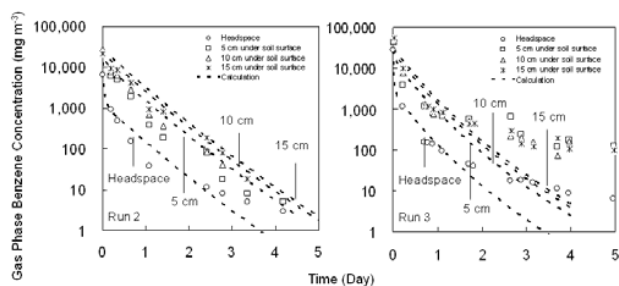


Fig. 4 Measured and calculated benzene concentrations in gas phase (Run 2 (water content of 13.4%) and Run 3 (water content of 0.4%)).

The Freundlich constants determined by the adsorption experiment were used in the calculations. As shown in Fig. 4 (left), the calculation result satisfactorily fit to the wet sand (Run 2) whereas it did not match the result on sand with low water content (Run 3). The good agreement between calculation and experimental results in Run 2 suggests that the diffusion model with nonlinear adsorption equation was

adequate to describe benzene vapor diffusion in wet sand. However, it showed obvious breakthrough between calculated result and measured result after the second day for Run 3.

The poor agreement between the calculation and experiment in Run 3 indicates that the model described above was not enough to predict the transport of benzene in sand with low water content. Depending on the study of Chiou et al. [3], in the absence of water or very low water content, the sorption of vapor proceeds as adsorption on mineral surfaces rather than partitioning in organic matter. It indicated that vapor adsorption should be considered in low water content for sand with low organic matter and high mineral content. For the dry soil area of  $7.769 \text{ cm}^2 \text{g}^{-1}$ , bulk density of  $1.5 \text{ g cm}^{-3}$  (TABLE1), and water surface area of  $11.4 \text{ \AA}^2/\text{molecule}$  [26], a moisture content required for monolayer coverage of the soil of  $0.00316 \text{ ml H}_2\text{O}/\text{cm}^3$  of soil was determined. At  $\epsilon_w=0.006$ , the overall coverage of sand seemed to be less than two layers of water molecules. Therefore, vapor phase adsorption should be considered in the diffusion model. Furthermore, some studies have proposed that organic compounds can be adsorbed easier in low water content than high water content for soil with low organic matter [10]. The same conclusion was obtained comparing the results of Runs 2 and 3, showing that benzene elimination was slower in dry sand than wet sand. This indicated that more benzene was adsorbed in dry sand.

In order to predict the benzene transport in sand with low water content, vapor phase adsorption should be considered. For predicting the vapor adsorption, the adequacy of vapor adsorption equation as reported in previous studies [10, 27] will be performed and related adsorption coefficient should be determined through experiment.

#### V. CONCLUSION

The two-phase diffusion model, accounting for gas and liquid phase diffusion and local equilibrium adsorption, adequately predicted benzene vapor transport in wet Andosol and sand. Liquid phase adsorption for Andosol could be expressed as linear isotherm, while it could be expressed as nonlinear for sand. Furthermore, linear adsorption constants were determined to be  $K = 1.24$  and  $0.20$ ,  $n = 1.05$  and  $1.11$  for Andosol and sand, respectively.

However, it seems that the model is not adequate to predict benzene transport in sand with low water content. More benzene seems to be adsorbed on sand with low water content (0.4%) than high water content (12.3%), indicating that vapor adsorption should be considered in the model in the follow-up study.

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