

## New insight into advection of organic contaminate plume at drain outlet areas

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### Abstract:

To provide insight to optimize organic contaminants treatment at the drain outlet areas due to the discharge from the industrial plants, the fate and transport of variety kinds of organic contaminants were investigated. By taking the chemical properties (molecular weight (MW), Henry's law constant (H), vapor pressure ( $P_v$ ), organic carbon normalized soil-water partition coefficient for organic compounds ( $K_{oc}$ ), water solubility (S), air diffusivity ( $D_a$ ), and water diffusivity ( $D_w$ )) of 56 kinds of organic chemical into account, we studied the general transport equation (GTE) regarding the mass balance for the steady-state analysis of the accumulation of a chemical over time by the plume advection. We found that  $K_{oc}$  has significant relationship with S and MW of the chemicals. Furthermore, higher organic carbon-water partitioning coefficient of the chemical will make it easier to stay at the drain outlet area, which gives us hints for next research of nanoscale particles for in situ remediation.

**Keywords:** Environment | Water pollution | Organic contaminant | Transport | Advection | Drain outlet

### Article:

#### 1. Introduction

As a result of industrial practices, organic contaminants have been detected in many drain outlet areas, resulting in contaminant in crop growth, water ecological environment, clean water resource and human health, which is lack of enough concern in some developing countries (Ávila et al., 2015, Honkonen and Rantalainen, 2012, Huang et al., 2015). The need to understand the fate and transport of the organic contaminants has led to numerous studies that used various techniques including dye- and chemical tracer studies (Barns et al., 2015, Subedi et al., 2015), and simple to more complex digital or numerical transport models (Georgi et al., 2015, Simon et al., 2013). However, all of them still did not solve the movement of particular organic contaminant problems with the determination of what happens to them (e.g. when does it end up? What does it transform to? How long does it persist? etc.) by the processes of definition, model, monitor and quantifying.

Obviously, the non-uniform/non-steady-state has more problems of interest. Most of the fate and transport phenomenon of the contaminants in natural or man-made systems are non-uniform and non-steady-state (Ramšak et al., 2013). However, it is difficult to define them and make models to quantify due to their un-directional concentration distribution. Moreover, the real and complex system includes solids and voids and there are lack of detailed data on its configuration within the void space (Yao et al., 2013). Thus, under some conditions, researchers just make it simple and assume it steady-state to do qualitative analysis. Luckily, researchers still could validate the model at a point within the void space by both recognizing the porous medium domain as a whole visualized as a continuum, and each of the phases and components as a continuum (Chabauty et al., 2015, Cheng and Saiers, 2015, Essaid et al., 2015). Sincerely, what more important is to win the public's concern like (Robles-Morua et al., 2012) did before. They showed that the regions in the river are in noncompliance with fresh water pathogen norms. It was shown that these risks are highly sensitive to spatiotemporal variability in river discharges and uncertainty in pathogen removal rates. Thus, the local government and people could be conscious of this series problem.

In this study, we derived the general transport equation (GTE) with the mass balance equation for the 56 kinds of organic chemicals. After solving the problem about GTE, we could get the accumulation of a substance over time within the defined control volume with the independent variables of the flux that we should assume. Specific information about chemical parameters that is needed includes molecular weight (MW), Henry's law constant (H), vapor pressure ( $P_v$ ), organic carbon normalized soil-water partition coefficient for organic compounds ( $K_{oc}$ ), water solubility (S), air diffusivity (Da), and water diffusivity ( $D_w$ ).

## 2. Methodology

In order to investigate the consistency between the tabulated values in Table S1, we firstly calculated the estimated Henry's law constant  $H(\text{cal})$  using the following equation (Lesser et al., 2008, Luo, 2009):

$$H(\text{cal}) = \frac{P_v M}{RTS} \quad (1)$$

where R is the gas constant of  $0.082 \text{ L-vapor atm mol}^{-1} \text{ K}^{-1}$ , T is the absolute temperature (K),  $P_v$  is the vapor pressure (atm), S is the solubility (g/L-water) and M is the molecular weight of the chemical (g/mol).

Furthermore,  $K_{oc}$  has been correlated with the aqueous solubility and the solubility gives a good first approximation of adsorption when  $\log K_{oc}$  is plotted against  $\log S$  for a group of organic compounds as the following equation (Shea, 1989):

$$\log K_{oc} = 3.94 - 0.50 \log S \quad (2)$$

The expressions for fraction of total mass in dissolved phase, fraction of total mass in sorbed phase, and fraction of total mass in vapor phase are shown as follows and the derivative details are included in SI:

$$\text{fraction – dissolved} = \frac{\phi_m C_m V}{\rho C_T V} = \frac{\phi_m}{H\phi_v + \phi_m + \rho K_s} \quad (3)$$

$$\text{fraction – sorbed} = \frac{\rho C_s V}{\rho C_T V} = \frac{\rho K_s}{H\phi_v + \phi_m + \rho K_s} \quad (4)$$

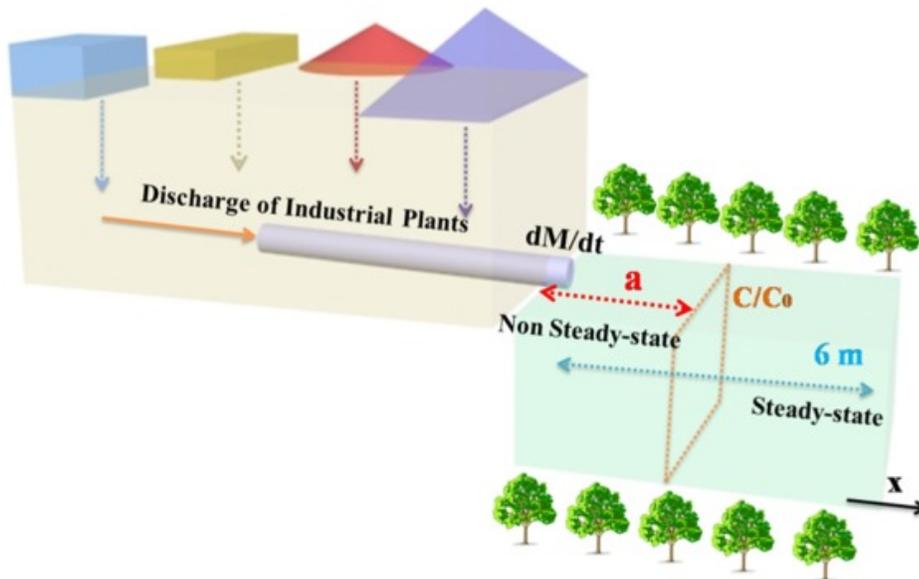
$$\text{fraction – vapor} = \frac{\phi_v C_v V}{\rho C_T V} = \frac{H\phi_v}{H\phi_v + \phi_m + \rho K_s} \quad (5)$$

where, in our study,  $C_m$ ,  $C_s$ ,  $C_v$ , and  $C_T$  are dissolved concentration ( $\text{g/L}^3$ -water), sorbed concentration ( $\text{g/L}^3$ -soil), vapor concentration ( $\text{g/L}^3$ -vapor), and total concentration ( $\text{g/L}^3$ ), respectively, soil bulk density  $\rho=1.5 \text{ kg-soil/cm}^3$ -soil,  $\Phi_v = 0.2 \text{ L-vapor/cm}^3$ -total voids,  $\Phi_m = 0.2 \text{ cm}^3$ -water/ $\text{cm}^3$ -total voids,  $K_s = f_{oc} * K_{oc}$  with fraction of organic carbon  $f_{oc} = 0.01 \text{ g-OC/g-soil}$ .

Using the advection model schematically illustrated in Fig. 1, the following mass balance equation could be obtained to derive the transient concentration curve (Cavanagh et al., 2014, Lundegard and Johnson, 2004):

$$\frac{dM}{dt} = C_0 q_m A - C_{600} q_m A + Re \quad (6)$$

where  $M$  is the mass of the chemical ( $\text{g}$ ),  $t$  is the time ( $\text{day}$ ),  $C_0$  is the source concentration ( $\text{g/L}^3$ ),  $q_m$  is the specific discharge ( $\text{L}^3/\text{day/m}^2$ ),  $C_{600}$  is the concentration at  $600 \text{ cm}$  ( $\text{g/L}^3$ ),  $A$  is the cross area ( $\text{m}^2$ ), and  $Re$  is the reaction term ( $\text{g/day}$ ).



**Fig. 1.** Scheme of the advection model for organic chemicals at the drain outlet areas.

With the assumption of steady-state and no reaction, we can obtain the following equations:

$$\frac{dM}{dt} = 0 \text{ and } C_{600} = C_0 \quad (7)$$

With another control volume cutting at  $x$ , another mass balance equation could be obtained:

$$\frac{dM}{dt} = C_0 q_m A - C_x q_m A + Re \quad (8)$$

So at steady-state  $C = C_0$  over  $x$ .

In this case, we divided the control volume into two control volumes when the plume reaches a with the moving of the plume, then

$$\frac{dM}{dt} = C_0 q_m A - C_a q_m A \quad (9)$$

So we can get  $C = C_0$  when  $x < a$  or  $x = a$ , and  $C = 0$  when  $x > a$ .

For problems, where diffusion, dispersion, and reduction are negligible in comparison with advection, there are stationary vapor and solid phases with the moving bulk water along the  $x$  direction and the geometry is such that the problem can be treated as one-dimensional (Lundegard and Johnson, 2004), the GTE can be reduced from

$$\frac{\partial}{\partial t} (\phi_v C_v + \phi_m C_m + \rho C_s) = - \frac{\partial}{\partial x} (q_{m,x} C_m) + \frac{\partial}{\partial x} \left( D_m^{\text{eff}} \frac{\partial C_m}{\partial x} + \phi_m D_m^{\text{diap}} \frac{\partial C_m}{\partial x} \right) \quad (10)$$

to:

$$\frac{\partial}{\partial t} \{ (\phi_v H + \phi_m + \rho K_s) C_m \} = - \frac{\partial}{\partial x} (q_{m,x} C_m) \quad (11)$$

where  $D^{\text{eff}}$  and  $D^{\text{disp}}$  are diffusion and dispersion coefficient, respectively.

We will look into the solution for the case where chemical is not present initially at the model domain boundaries, the concentration suddenly is increased and remains relatively constant at one boundary ( $x = 0$ ). In this case, the boundary and initial conditions are:

$C = C_0$  at  $x = 0$  and all  $t$ , and  $C = 0$  at  $t = 0$  and all  $x$ .

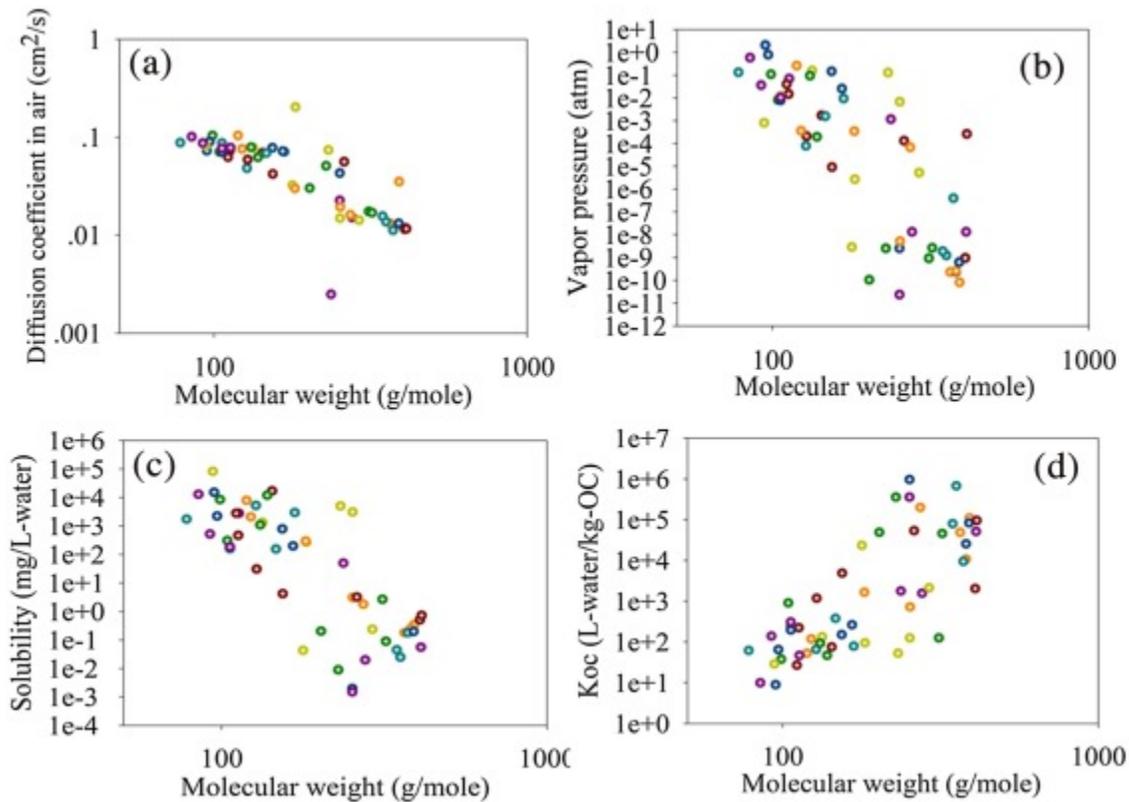
When  $C(x, t) = 0$ , the GTE solution is:

$$x > \frac{q_{m,x}t}{H\phi_v + \phi_m + \rho_b K_{i,s}} \quad (12)$$

When  $C(x, t) = C_0$ , the GTE solution is:

$$x \leq \frac{q_{m,x}t}{H\phi_v + \phi_m + \rho_b K_{i,s}} \quad (13)$$

where, in our model, bulk density  $\rho_b = 1.7$  kg-soil/L-soil; total porosity =  $0.35$  cm<sup>3</sup>-pores/cm<sup>3</sup>-media, moisture content =  $0.32$  cm<sup>3</sup>-H<sub>2</sub>O/cm<sup>3</sup>-total volume;  $\Phi_m = 0.2$  L-water/L-total voids;  $f_{oc} = 0.02$  g-OC/g-solids, and a specific discharge rate of  $10$  cm/min.



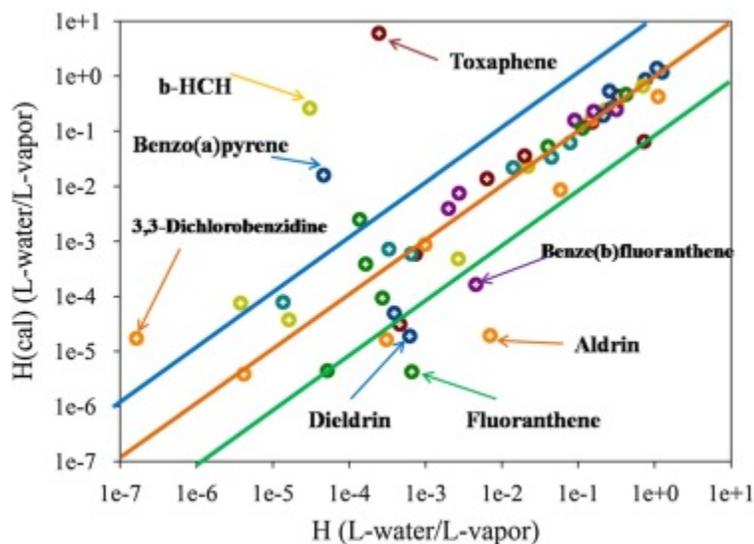
**Fig. 2.** Chemical properties of 56 kinds of organic chemicals (see Table S1). (a). Diffusion coefficient in air vs. molecular weight. (b) Vapor pressure vs. molecular weight. (c) Solubility vs molecular weight. (d) Organic carbon normalized soil-water partition coefficient for organic compounds vs molecular weight.

### 3. Results and discussion

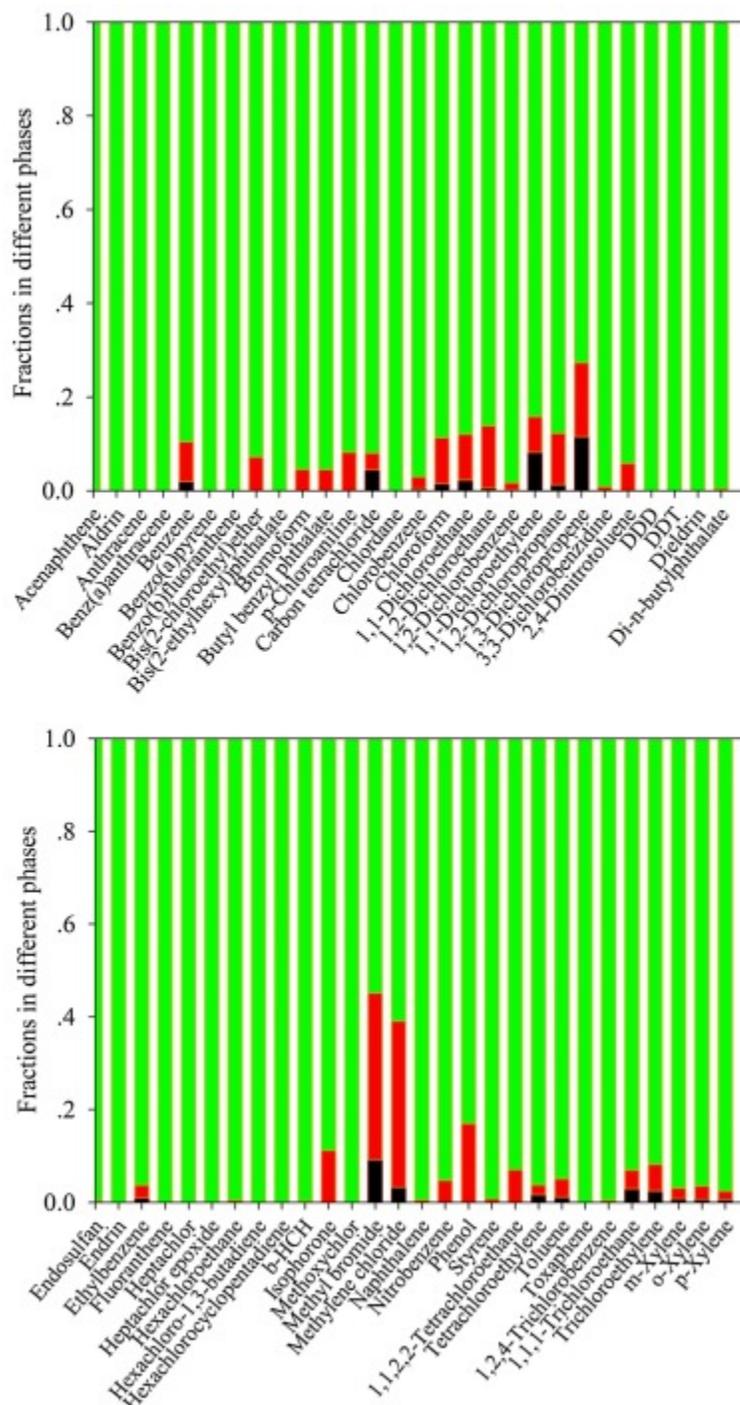
Regarding the chemical properties of the 56 kinds of chemicals from Fig. 2a–d (diffusion coefficient in water vs. molecular weight see Fig. S1), as molecular weights of the chemicals increase, the solubility, vapor pressure, and diffusion coefficient in air or water decrease while organic carbon-water partitioning coefficient increases. Using the molecular weights of 56 kinds of organic chemicals as the reference variables, among them, Fig. 2a, d and Fig. S1 show that

there are only several kinds of chemicals that deviate from the trend by more than one order of magnitude from the average, note that we define one order of magnitude as the level of correlation, which suggests that there is a strong correlation between molecular weight and the organic carbon-water partitioning coefficient/diffusion coefficient in water/air. However, a lot of chemicals have vapor pressures and solubilities that are above or below one order of magnitude from the average, which would suggest that there is no significant correlation between molecular weight and solubility/vapor pressure.

Fig. 3 shows the pairs of estimated Henry's law constant  $H(\text{cal})$  vs. Henry's law constant  $H$  of all 56 chemicals and one order below or above boundary lines. There are a total of 9 organic chemicals that fall outside of the region established by the two boundary lines. The pairs of values of chemicals differ by more than an order of magnitude. These chemicals include 3,3-dichlorobenzidine, benzo(a)pyrene, *b*-HCH, toxaphene, benzo(b)fluoranthene, aldrin, fluoranthene, and dieldrin. Recall the vapor pressures and solubility of these chemical that fall outside of the bands from Table S1, most of them have vapor pressures that are positioned at the bottom 20% of all chemicals in the database. Moreover, Most of them have solubilities that are located within the bottom 20% of all chemicals in the database. It can be concluded that smaller solubility or vapor pressure makes the estimation less reliable. For example, for 3,3-dichlorobenzidine, the estimated  $H$  using equation 1 has more than one order of magnitude greater than that of  $H$  value from table S1. Since 3,3-dichlorobenzidine is a sparingly soluble chemical, some of this chemical staying in the liquid phase might not be dissolved in water, which resulted in the overestimation of  $H$  using the equation 1. Similarly, for benzo(b)fluoranthene, the estimated  $H$  has more than one order of magnitude smaller than that of  $H$  value from table S1. Since benzo(b)fluoranthene is a low vapor pressure chemical, some of this chemical staying in the gas phase might not vaporized, which resulted in the underestimation of  $H$  using the equation 1. However, unlike Henry's law constant, organic carbon-water partitioning coefficient  $K_{oc}$  has good agreement with that of the tabled values as shown in Fig. S2, indicating that  $K_{oc}$  is well correlated with water solubility  $S$  (Shiflett et al., 2006).



**Fig. 3.** Estimated Henry's law constant ( $H(\text{cal})$ ) vs. Henry's law constant ( $H$ ) for the 56 kinds of organic chemicals and the three lines including the 1:1 slope line, a line (green) for which  $H = 10 H(\text{cal})$ , and a line (blue) for which  $H = 0.1 H(\text{cal})$ .



**Fig. 4.** Fractions in different phases for the 56 kinds of organic chemicals with green representing fraction-sorbed, red representing fraction-dissolved, and black representing fraction-vapor.

Fractions in different phases for the 56 kinds of organic chemicals have been plotted in Fig. 4. It indicates that fraction-sorbed is the largest while fraction-vapor is the smallest for all of the chemicals. Especially, the fraction-sorbed of some chemicals is close to 1. From the Eqs (3)–(5),

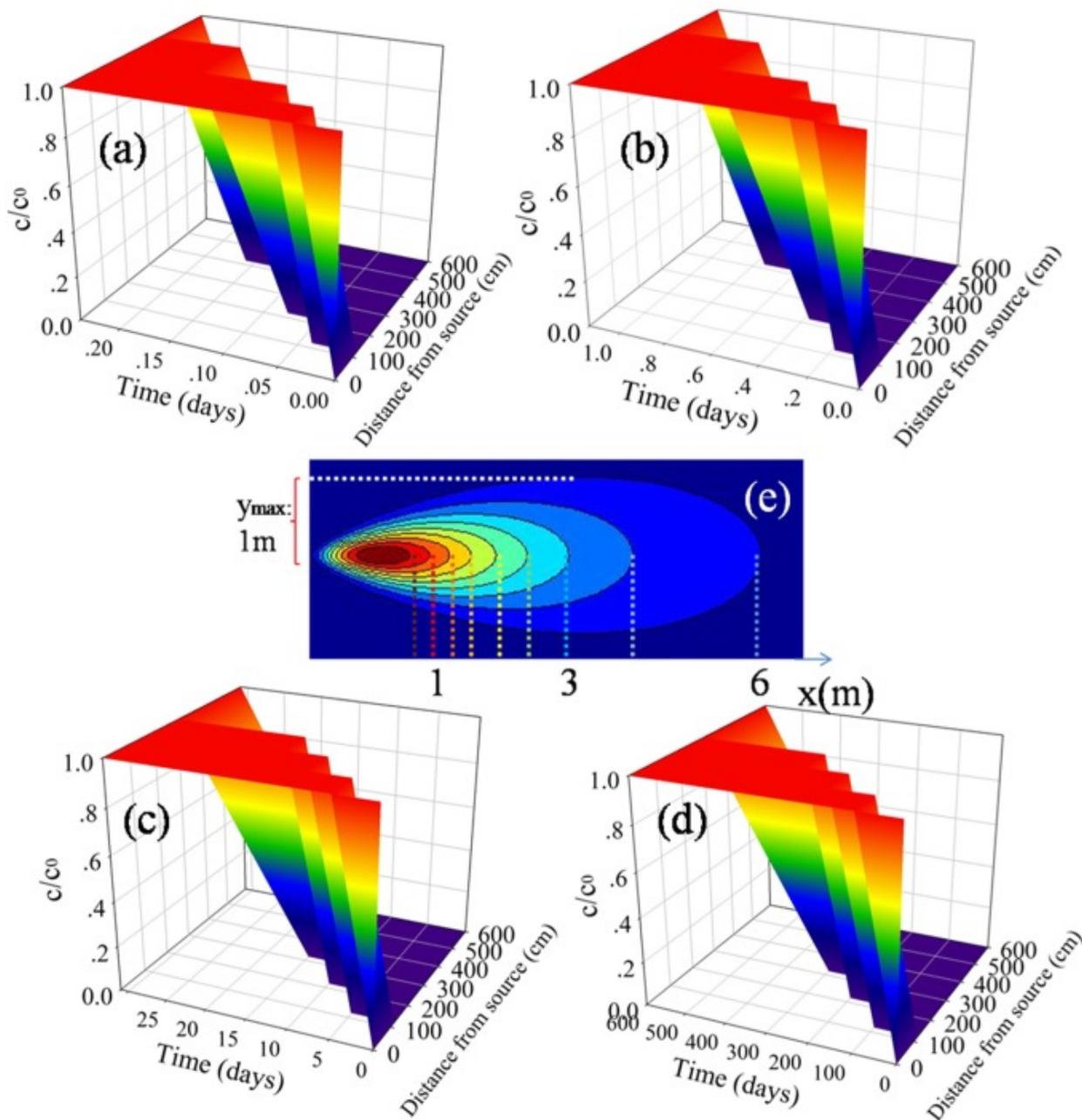
the magnitude of fraction-sorbed, fraction-dissolved and fraction-vapor have relationship with  $\rho$ ,  $K_s$ ,  $H$ ,  $\Phi_v$  and  $\Phi_m$ , respectively. Among them, the  $H$  and  $K_{oc}$  of chemicals basically determine the fraction distribution in different phases (Liu et al., 2013). Generally, higher  $H$  gives rise to relatively higher fraction in vapor, while lower  $K_{oc}$  causes higher fraction in water and lower fraction sorbed in soil. Based on this observation and trend, a rough estimated fractions in different phases can be initially figured out by observing these chemical properties, e.g. higher  $K_{oc}$  indicates more fraction sorbed in soil and lower fraction dissolved in water; and higher  $H$  indicates bigger fraction in vapor than those with lower  $H$ , which could help to determine what kind of phases of the chemicals will present in the drain outlet areas. Since for organic contaminants with higher  $K_{oc}$ , nanoscale particles for in situ remediation could be applied in solid phase resulted from effective subsurface dispersion.

**Table 1.** Steady-state time for the 56 kinds of organic chemicals according to the plume moving from source with water flow.

Group	Chemical Name	Time [days]	Chemical Name	Time [days]
A	Benzene	0.054	Ethylbenzene	0.144
	Bis(2-chloroethyl)ether	0.066	Isophorone	0.048
	Bromoform	0.096	Methyl bromide	0.024
	Butyl benzyl phthalate	0.096	Methylene chloride	0.024
	p-Chloroaniline	0.06	Naphthalene	0.78
	Carbon tetrachloride	0.114	Nitrobenzene	0.09
	Chlorobenzene	0.156	Phenol	0.036
	Chloroform	0.048	Styrene	0.588
	1,1-Dichloroethane	0.048	1,1,2,2-Tetrachloroethane	0.066
	1,2-Dichloroethane	0.042	Tetrachloroethylene	0.186
	1,2-Dichlorobenzene	0.252	Toluene	0.102
	1,1-Dichloroethylene	0.06	1,1,1-Trichloroethane	0.102
	1,2-Dichloropropane	0.048	Trichloroethylene	0.078
	1,3-Dichloropropene	0.036	<i>m</i> -Xylene	0.168
	3,3-Dichlorobenzidine	0.468	<i>o</i> -Xylene	0.138
2,4-Dinitrotoluene	0.078	<i>p</i> -Xylene	0.21	
B	Di- <i>n</i> -butylphthalate	1.02	Hexachloroethane	1.14
	Endosulfan	1.26	<i>b</i> -HCH	1.38
	Endrin	6.6	1,2,4-Trichlorobenzene	1.08
	Heptachlor	6	Acenaphthene	3.06
C	Bis(2-ethylhexyl)phthalate	75	Hexachloro-1,3-butadiene	33
	Chlordane	32.4	Methoxychlor	46.8
	DDD	29.4	Toxaphene	57.6
	Dieldrin	16.8	Aldrin	31.2
	Fluoranthene	31	Anthracene	15
	Heptachlor epoxide	52.8		
D	Benz(a)anthracene	228	DDT	420
	Benzo(a)pyrene	600	Hexachlorocyclopentadiene	122
	Benzo(b)fluoranthene	228		

To study the accumulation of a chemical over time, the steady-state here describes the state of the system when a variable, e.g. concentration, remains constant with time changing. Note that equilibrium can exist where it is not necessary to be steady-state while steady-state can also be reached where equilibrium might not be reached (Paces, 1973). Table 1 shows 4 groups of the steady-state time reaching 6 m for the 56 kinds of organic chemicals according to the plume

moving from the source with water flow. The four groups of chemical are plotted and shown in Fig. S3–S6 based on the steady-state time of 1 day (group A), 1–10 days (group B), 10–100 days (group C) and more than 100 days (group D), respectively.



**Fig. 5.** Steady-state time for the 4 kinds of organic chemicals according to the plume moving from source with water flow using equations (6)–(13): (a) Methyl bromide; (b) Hexachloroethane; (c) Fluoranthene; and (d) Benzo(a)pyrene. (e) The relationship between  $x$  and  $y$  directions for plume advection.

We selected one representative chemical from each of the four groups for a further comparison and discussion, such as Methyl bromide (Fig. 5a), Hexachloroethane (Fig. 5b), Fluoranthene

(Fig. 5c), and Benzo(a)pyrene (Fig. 5d). Fig. 5a–d show the concentration change from 0 to  $C_0$  from  $x = 0$  to 6 m with the plume moving and Fig. 5e shows the plume can reach 1 m in  $y$  direction within 6 m for the 56 kinds of chemicals. All of the four chemicals show the similar shape of the steady-state concentration profiles. Since lower  $H$  and  $K_{oc}$  could make the chemical more hydrophilic and stay with the bulk liquid, lower  $H$  and  $k_{oc}$  give rise to shorter time to reach steady-state as Methyl bromide did. However, it will need much more time for Benzo(a)pyrene to reach steady-state within 6 m. Basically, the largest factor affecting advective transport is  $K_{oc}$ , since  $K_{oc}$  increases, advection decreases, while the change of  $H$  does not significantly affect advection due to that the flow moves as a wall from the beginning to the end of the region of interest if without reaction or significant dispersion/diffusion terms. Therefore, the concentration is best represented by a step function, where concentration is either fully encountered or not yet encountered. And the chemical with high  $K_{oc}$ , e.g. Benzo(a)pyrene, will be easy to stay at the drain outlet area. Moreover, for in situ remediation, the reactions between nanoscale particles and organic contaminants involving sorption, desorption, aggregation will help to study the long-term fate of nanoscale particle, which has been rarely reported, according to the fate and transport of organic contaminants.

#### 4. Conclusion

For the 56 kinds of chemicals we researched, organic carbon-water partitioning coefficient  $K_{oc}$  has significant relationship with water solubility and molecular weight. Our calculation indicates that higher organic carbon-water partitioning coefficient results in more fraction sorbed in soil and lower fraction dissolved in water. More importantly, chemicals with high  $K_{oc}$  will be easier for them to stay at the drain outlet area. It suggests that more effort should be taken to focus on this kind of chemicals when applying the nanotechnology removal for the drain outlet area.

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#### Appendix A. Supplementary data

Supplementary data associated with this article can be found at <http://dx.doi.org/10.1016/j.enmm.2016.08.001>.

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