Supplement of

New model of reactive transport in a single-well push–pull test with aquitard effect and wellbore storage

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Supplementary Materials

S1. Derivation of analytical solutions for the SWPP test

To reduce the complexity in analyzing the influence of input parameters on the output, the

dimensionless parameters are introduced as follows: $C_{mD} = \frac{C_m}{C_0}$, $C_{imD} = \frac{C_{im}}{C_0}$, $C_{inj,mD} = \frac{C_{inj,m}}{C_0}$,

$C_{inj,imD} = \frac{C_{inj,im}}{C_0}$, $C_{cha,mD} = \frac{C_{cha,m}}{C_0}$, $C_{cha,imD} = \frac{C_{cha,im}}{C_0}$, $C_{res,mD} = \frac{C_{res,m}}{C_0}$, $C_{res,imD} = \frac{C_{res,im}}{C_0}$,

$C_{ext,imD} = \frac{C_{ext,im}}{C_0}$, $C_{ext,imD} = \frac{C_{ext,im}}{C_0}$, $C_{umD} = \frac{C_{um}}{C_0}$, $C_{ulimD} = \frac{C_{ulim}}{C_0}$, $C_{limD} = \frac{C_{lim}}{C_0}$, $C_{limD} = \frac{C_{lim}}{C_0}$, $t_D = \frac{|A|}{a_r^2 R_m} t$, $\tau_D = \frac{r}{a_r}$, $\tau_{WD} = \frac{r_w}{a_r}$, $Z_D = \frac{z}{B}$, $\mu_{mD} = \frac{\alpha_D^2 R_m l_m}{A}$, $\mu_{lmD} = \frac{\alpha_D^2 R_m l_m}{A}$, $\mu_{lmD} = \frac{\alpha_D^2 R_m l_m}{A}$, where the subscript “D” represents the

dimensionless parameter hereinafter, $A = \frac{Q}{4\pi B \theta_m}$. By substituting these dimensionless parameters

into the governing equations, one could obtain the dimensionless model of the SWPP test:

\[
\begin{align}
\frac{\partial C_{mD}}{\partial t_D} &= \frac{1}{r_D} \frac{\partial^2 C_{mD}}{\partial r_D^2} - \frac{1}{r_D} \frac{\partial C_{mD}}{\partial r_D} - \epsilon_m (C_{mD} - C_{imD}) - \mu_{mD} C_{mD} - \left( \frac{\theta_{um} \alpha_D^2 v_{um}}{2A \theta_m B} C_{umD} - \right. \\
\frac{\theta_{um} \alpha_D^2 D_u}{2A \theta_m B^2} \frac{\partial C_{umD}}{\partial z_D} \bigg|_{z_D=1} + \left( \frac{\theta_{im} \alpha_D^2 v_{im}}{2AB \theta_m} C_{imD} - \frac{\theta_{im} \alpha_D^2 D_i}{2AB^2 \theta_m} \frac{\partial C_{imD}}{\partial z_D} \bigg|_{z_D=-1} \right), & \tau_D \geq \tau_{WD}, \\
\frac{\partial C_{imD}}{\partial t_D} &= \epsilon_{im} (C_{mD} - C_{imD}) - \mu_{imD} C_{imD}, & \tau_D \geq \tau_{WD}, \\
\frac{\partial C_{umD}}{\partial t_D} &= \frac{R_m \alpha_D^2 D_u}{AB^2 R_m} \frac{\partial^2 C_{umD}}{\partial z_D^2} - \frac{R_m v_{um} \alpha_D^2}{AB R_m} \frac{\partial C_{umD}}{\partial z_D} - \epsilon_{um} (C_{umD} - C_{ulimD}) - \mu_{umD} C_{umD}, \\
Z_D &\geq 1, \\
\frac{\partial C_{ulimD}}{\partial t_D} &= \epsilon_{ulim} (C_{umD} - C_{ulimD}) - \mu_{ulimD} C_{ulimD}, & Z_D \geq 1,
\end{align}
\]

\[
\begin{align}
\frac{\partial C_{imD}}{\partial t_D} &= \frac{R_m \alpha_D^2 D_i}{AB^2 R_{\text{lim}}} \frac{\partial^2 C_{imD}}{\partial z_D^2} + \frac{R_m v_{im} \alpha_D^2}{AB R_{\text{lim}}} \frac{\partial C_{imD}}{\partial z_D} - \epsilon_{im} (C_{imD} - C_{limD}) - \mu_{imD} C_{imD}, \\
Z_D &\leq -1.
\end{align}
\]
\[ \frac{\partial C_{\text{lim}}}{\partial t_D} = \varepsilon_{\text{lim}} (C_{\text{imD}} - C_{\text{limD}}) - \mu_{\text{limD}} C_{\text{limD}}, \quad z_D \leq -1, \quad (\text{S3b}) \]

where \( \varepsilon_m = \frac{\omega_a \alpha_i^2}{A \theta_m}, \) \( \varepsilon_{\text{im}} = \frac{\omega_a \alpha_i^2 R_m}{A \theta_{\text{im}} R_{\text{im}}}, \) \( \varepsilon_{\text{um}} = \frac{\omega_a \alpha_i^2 R_m}{A \theta_{\text{um}} R_{\text{um}}}, \) \( \varepsilon_{\text{lim}} = \frac{\omega_a \alpha_i^2 R_m}{A \theta_{\text{lim}} R_{\text{lim}}}. \)

The analytical solution will be derived using the Laplace transform method and the Green’s functions method, and the detailed information could be seen in the following sections.

\section*{S1.1 Solutions in the injection phase: Eqs. (25a) and (25f)}

Substituting the dimensionless parameters into Eqs. (5) - (6), one could obtain the dimensionless boundary conditions and dimensionless initial conditions for the injection phase:

\[ C_mD(r_D, t_D)|_{t_D=0} = C_{\text{imD}}(r_D, t_D)|_{t_D=0} = C_{\text{umD}}(r_D, z_D, t_D)|_{t_D=0} = C_{\text{limD}}(r_D, z_D, t_D)|_{t_D=0} = 0, \quad (\text{S4}) \]

\[ C_{\text{limD}}(r_D, z_D, t_D)|_{t_D=0} = C_{\text{limD}}(r_D, z_D, t_D)|_{t_D=0} = 0. \]

Conducting Laplace transform to Eqs. (S2a) - (S2b), one has:

\[ s \tilde{C}_{\text{umD}} = \frac{R_m \alpha_i^2 P_u}{A B^2 R_{\text{um}}} \frac{\partial^2 C_{\text{umD}}}{\partial z_D^2} + \frac{R_m \alpha_i^2 \alpha_j^2}{A B R_{\text{um}}} \frac{\partial C_{\text{umD}}}{\partial z_D} + (\varepsilon_{\text{um}} + \mu_{\text{umD}}) \tilde{C}_{\text{umD}} + \varepsilon_{\text{um}} \tilde{C}_{\text{umD}}, \quad z_D \geq 1. \quad (\text{S7a}) \]

\[ s \tilde{C}_{\text{umD}} = \varepsilon_{\text{um}} (\tilde{C}_{\text{umD}} - \tilde{C}_{\text{umD}}) - \mu_{\text{umD}} \tilde{C}_{\text{umD}}, \quad z_D \geq 1, \quad (\text{S7b}) \]

Substituting Eq. (S7b) into Eq. (S7a) will lead to:

\[ s \tilde{C}_{\text{umD}} = \frac{R_m \alpha_i^2 P_u}{A B^2 R_{\text{um}}} \frac{\partial^2 C_{\text{umD}}}{\partial z_D^2} + \frac{R_m \alpha_i^2 \alpha_j^2}{A B R_{\text{um}}} \frac{\partial C_{\text{umD}}}{\partial z_D} - (\varepsilon_{\text{um}} + \mu_{\text{umD}} + \frac{\varepsilon_{\text{um}} \varepsilon_{\text{um}}}{s + \mu_{\text{umD}}}) \tilde{C}_{\text{umD}}, \]
Similarly, Eqs. (S3a) - (S3b) become:

\[ s\tilde{C}_{\text{limD}} = \frac{R_m\alpha_r^2 D_l}{AB^2 R_{\text{lim}}} \frac{\partial^2 C_{\text{limD}}}{\partial z_D^2} + \frac{R_m\nu_m\alpha_r^2}{ABR_{\text{lim}}} \frac{\partial C_{\text{limD}}}{\partial z_D} - (\epsilon_{\text{limD}} + \mu_{\text{limD}})\tilde{C}_{\text{limD}} + \epsilon_{\text{limD}} \tilde{C}_{\text{limD}}, \]

where overbar represents the variables in Laplace domain hereinafter; \( s \) is the Laplace transform parameter in respect to dimensionless time.

Eqs. (S5), (S6a)-(S6b) and (S8) compose a model of the second-order ordinary differential equation (ODE) with boundary conditions, the general solution of Eq. (S8) is:

\[ \tilde{C}_{\text{umD}} = A_1 e^{a_1 z_D} + B_1 e^{a_2 z_D}. \] (S11a)

Similarly, the general solution of Eq. (S10) is:

\[ \tilde{C}_{\text{lmD}} = A_2 e^{b_1 z_D} + B_2 e^{b_2 z_D}. \] (S11b)

where

\[ a_1 = \frac{\frac{R_m\nu_m\alpha_r^2}{ABR_{\text{um}}} \sqrt{\left(\frac{R_m\nu_m\alpha_r^2}{ABR_{\text{um}}}\right)^2 + 4\frac{R_m\alpha_r^2 D_l}{AB^2 R_{\text{um}}} (s + \epsilon_{\text{umD}} + \mu_{\text{umD}} - \frac{\epsilon_{\text{umD}}}{s + \mu_{\text{umD}} + \epsilon_{\text{umD}}})}}{2R_m\alpha_r^2 D_l/AB^2 R_{\text{um}}}, \]

\[ a_2 = \frac{\frac{R_m\nu_m\alpha_r^2}{ABR_{\text{um}}} \sqrt{\left(\frac{R_m\nu_m\alpha_r^2}{ABR_{\text{um}}}\right)^2 + 4\frac{R_m\alpha_r^2 D_l}{AB^2 R_{\text{um}}} (s + \epsilon_{\text{umD}} + \mu_{\text{umD}} - \frac{\epsilon_{\text{umD}}}{s + \mu_{\text{umD}} + \epsilon_{\text{umD}}})}}{2R_m\alpha_r^2 D_l/AB^2 R_{\text{um}}}, \]

\[ b_1 = \frac{-\frac{R_m\nu_m\alpha_r^2}{ABR_{\text{lm}}} \sqrt{\left(\frac{R_m\nu_m\alpha_r^2}{ABR_{\text{lm}}}\right)^2 + 4\frac{R_m\alpha_r^2 D_l}{AB^2 R_{\text{lm}}} (s + \epsilon_{\text{lmD}} + \mu_{\text{lmD}} - \frac{\epsilon_{\text{lmD}}}{s + \mu_{\text{lmD}} + \epsilon_{\text{lmD}}})}}{2R_m\alpha_r^2 D_l/AB^2 R_{\text{lm}}}, \] and
\[
b_2 = \frac{R_m v_{im} a_{Dl}^2}{AB R_{im}} \sqrt{\frac{(R_m v_{im} a_{Dl}^2)^2}{AB R_{im}} + \frac{\gamma_{Dl}^2 (s + \varepsilon_{im} + \mu_{imD})}{s + \mu_{imD} + \varepsilon_{imD}}}.\]

Substituting Eqs. (S11a) - (S11b) into Eqs. (S5)-(S6) leads to:

\[
\tilde{c}_{umD} = B_1 e^{a_2 z_D}. \tag{S12a}
\]

\[
\tilde{c}_{lmD} = A_2 e^{b_1 z_D}. \tag{S12b}
\]

where \(B_1 = \tilde{c}_{mD} \exp(-a_2), B_2 = 0, A_1 = 0\) and \(A_2 = \tilde{c}_{mD} \exp(b_1)\).

Thus, we could obtain the solutions for the aquitards as:

\[
\tilde{c}_{umD} = \tilde{c}_{mD} \exp(a_2 z_D - a_2). \tag{S13a}
\]

\[
\tilde{c}_{umD} = \frac{e_{uim}}{s + \varepsilon_{um} + \mu_{umD}} \tilde{c}_{umD}, \tag{S13b}
\]

\[
\tilde{c}_{lmD} = \tilde{c}_{mD} \exp(b_1 z_D + b_1). \tag{S14a}
\]

\[
\tilde{c}_{lmD} = \frac{e_{lim}}{s + \varepsilon_{lim} + \mu_{limD}} \tilde{c}_{lmD}. \tag{S14b}
\]

In the injection phase, the dimensional boundary conditions Eq. (8) and Eqs. (12a)-(12b) are transformed into their dimensionless forms:

\[
\left[ C_{mD} - \frac{\partial C_{mD}(r, D)}{\partial r} \right]_{r = r_{inj}} = C_{inj, mD}(t_D), 0 < t_D \leq t_{inj,D} \tag{S15}
\]

\[
\beta_{inj} \frac{d C_{inj, mD}(t_D)}{d t_D} = 1 - C_{inj, mD}(t_D), 0 < t_D \leq t_{inj,D}, \tag{S16a}
\]

\[
C_{inj, mD}(t_D = 0) = 0. \tag{S16b}
\]

where \(\beta_{inj} = \frac{v_{w, inj} r_{injD}}{\xi R_m a_r}\).

Conducting Laplace transform to Eqs. (S1a) - (S1b), one has:

\[
s \tilde{C}_{mD} = \frac{1}{r_D} \frac{\partial^2 \tilde{C}_{mD}}{\partial r_D^2} - \frac{1}{r_D} \frac{\partial \tilde{C}_{mD}}{\partial r_D} - (\varepsilon_{m} + \mu_{mD}) \tilde{C}_{mD} + \varepsilon_{m} \tilde{C}_{lmD} - \frac{(\theta_{um} a_{Dl}^2 v_{um})}{2A \theta_{mB}} \tilde{C}_{umD} - \frac{(\theta_{um} a_{Dl}^2 v_{um})}{2A \theta_{mB}^2} \tilde{C}_{umD} \varepsilon_{Dl} \frac{\partial \tilde{C}_{umD}}{\partial z_D} \varepsilon_{Dl} = 1, \tag{S17a}
\]

\[
\left( \frac{\theta_{lm} a_{Dl}^2 v_{lm}}{2A \theta_{mB}} \tilde{C}_{lmD} - \frac{\theta_{lm} a_{Dl}^2 v_{lm}}{2A \theta_{mB}^2} \tilde{C}_{lmD} \varepsilon_{Dl} \frac{\partial \tilde{C}_{lmD}}{\partial z_D} \varepsilon_{Dl} = 1, \right) \tag{S17b}
\]
where

\[ E = s + \varepsilon_m + \mu_m - \frac{\varepsilon_m e_{im}}{s + \mu_{im} + \varepsilon_{im}} + \frac{\theta_1 a_0^2 v_{um}}{2A\theta_m B} - \frac{\theta_1 a_0^2 v_{im}}{2AB\theta_m} - \frac{a_2 \theta_0 a_5 D_u}{2A\theta_m B^2} + \frac{b_2 \theta_0 a_5 D_1}{2AB^2\theta_m}. \]

The boundary conditions of the wellbore and infinity in the Laplace domain are:

\[ \left[ \tilde{C}_{mD} - \frac{\partial \tilde{C}_{mD}(r_D, s)}{\partial r_D} \right]_{r = r_{WD}} = \tilde{C}_{inj,mD}(s), \quad (S19a) \]

\[ \tilde{C}_{mD}(r_D, s) \bigg|_{r_D \to \infty} = 0. \quad (S19b) \]

Conducting Laplace transform on Eqs. (S16a)- (S16b), one has:

\[ \tilde{C}_{inj,mD}(r_w, s) = \frac{1}{s(s \beta_{inj} + 1)}. \quad (S20) \]

Eqs. (S18), (S19a)-(S19b), and (S20) compose a model of the second-order ordinary differential equation (ODE) with boundary conditions. The general solution of Eq. (S18) is:

\[ \tilde{C}_{mD}(r_D, s) = \phi_1 \exp \left( \frac{y_{inj}}{2} \right) A_1 \left( E^{1/3} y_{inj} \right) + \phi_2 \exp \left( \frac{y_{inj}}{2} \right) B_1 \left( E^{1/3} y_{inj} \right). \quad (S21) \]

where \( y_{inj} = r_D + \frac{1}{4E}, y_{inj,w} = r_{WD} + \frac{1}{4E} \); \( \phi_1 \) and \( \phi_2 \) are constants which could be determined by the boundary conditions; \( A_1(\cdot) \) and \( B_1(\cdot) \) are the Airy functions of the first kind and second kind, respectively. As \( B_1(r_D) \) diverges when \( r_D \to \infty \), \( \phi_2 \) has to be zero.

Substituting Eqs. (S21), (S20) and \( \phi_2 = 0 \) into Eq. (S19a), the value of \( \phi_1 \) is:

\[ \phi_1 = \frac{1}{s(s \beta_{inj} + 1) \exp \left( \frac{y_{inj,w}}{2} \right)} \frac{1}{A_1 \left( E^{1/3} y_{inj,w} \right) - E^{1/3} A_1 (E^{1/3} y_{inj})} \quad (S22) \]

where \( A'_1(\cdot) \) is the derivative of the Airy function.
Substituting Eq. (S22) and \( \phi = 0 \) into Eqs. (S21) and (S17b), one could obtain the Laplace-domain analytical solution of solute transport in the injection phase of the SWPP test.

### 1.2 Solutions in the chaser phase: Eqs. (26a) - (26g)

For the chaser phase, conducting Laplace transform on Eqs. (S2a)-(S2b), one has:

\[
\frac{R_m \alpha^2 B_u}{AB^2 R_{um}} \frac{\partial^2 C_{umD}}{\partial z^2} - \frac{R_m \nu_{um} \alpha^2}{ABR_{um}} \frac{\partial C_{umD}}{\partial z} - \left( s + \nu_{um} + \mu_{umD} \right) C_{umD} + \nu_{um} \bar{C}_{umD} + \\
\frac{C_{umD}(r_D, z_D, t_{inj,D}) = 0, \ z_D \geq 1, \ (S23a)}{}
\]

Similarly, Eqs. (S3a) - (S3b) become:

\[
\frac{R_m \alpha^2 B_l}{AB^2 R_{lm}} \frac{\partial^2 C_{lmD}}{\partial z^2} + \frac{R_m \nu_{lm} \alpha^2}{ABR_{lm}} \frac{\partial C_{lmD}}{\partial z} - \left( s + \nu_{lm} + \mu_{lmD} \right) C_{lmD} + \nu_{lm} \bar{C}_{lmD} + \\
\frac{C_{lmD}(r_D, z_D, t_{inj,D}) = 0, \ z_D \leq -1, \ (S25a)}{}
\]

For the chaser phase, conducting Laplace transform on Eqs. (S2a)-(S2b), one has:

\[
\frac{C_{umD}(r_D, z_D, t_{inj,D}) + \nu_{um} C_{umD}(r_D, z_D, t_{inj,D})}{s + \nu_{um} + \mu_{umD}} = 0, \ z_D \geq 1, \ (S24)
\]

Similarly, Eqs. (S3a) - (S3b) become:

\[
\frac{C_{lmD}(r_D, z_D, t_{inj,D}) + \nu_{lm} C_{lmD}(r_D, z_D, t_{inj,D})}{s + \nu_{lm} + \mu_{lmD}} = 0, \ z_D \leq -1, \ (S25a)
\]
\[
\frac{K_m u^2 D_1}{A B^2 R_{lm}} \frac{d^2 c_{lmD}}{d z_D^2} + \frac{K_m u^2 D_1}{A B R_{lm}} \frac{d c_{lmD}}{d z_D} = \left( s + \epsilon_{lm} + \mu_{lmD} - \frac{\epsilon_{lm} \epsilon_{lmD}}{s + \epsilon_{lm} + \mu_{lmD}} \right) \tilde{c}_{lmD} + \\
C_{lmD}(r_D, z_D, t_{inj,D}) + \frac{\epsilon_{lm} c_{lmD}(r_D, z_D, t_{inj,D})}{s + \epsilon_{lm} + \mu_{lmD}} = 0. \quad z_D \leq -1, \quad (S26)
\]

where \( C_{umD}(r_D, z_D, t_{inj,D}) \) and \( C_{uimD}(r_D, z_D, t_{inj,D}) \) are respectively the mobile and immobile concentrations [ML\(^{-3}\)] of the upper aquitard at the end of the injection phase, \( C_{lmD}(r_D, z_D, t_{inj,D}) \) and \( C_{limD}(r_D, z_D, t_{inj,D}) \) are respectively the mobile and immobile concentrations [ML\(^{-3}\)] of the lower aquitard at the end of the injection phase. In this study, we use the Green’s function method to derive the analytical solution of Eqs. (S24) and (S26).

Notice that the boundary condition of Eq. (S6a) is inhomogeneous, thus we need to homogenize it first. Letting \( \tilde{c}_{umD} = \tilde{k}(z_D) + \delta_1 + \delta_2 z_D \), and substituting them into Eqs. (S5) and (S6a) yields:

\[
[k(\tilde{k}(z_D))]_{z_D=\infty} = 0, \quad (S27a)
\]
\[
[k(\tilde{k}(z_D))]_{z_D=1} = 0, \quad (S27b)
\]

where \( \delta_1 = -\delta_2 z_{eD} \) and \( \delta_2 = \frac{c_{lmD}(r_D, s)}{1 - z_{eD}} \).

Defining the spatial operator: \( L_u = \left[ \frac{K_m u^2 D_1}{A B^2 R_{um}} \frac{d^2}{d z_D^2} - \frac{K_m u^2 D_1}{A B R_{um}} \frac{d}{d z_D} - E_u \right] \), one has:

\[
L_u \tilde{c}_{umD} = L_u [\tilde{k}(z_D) + \delta_1] = F_u(z_D), \quad (S28)
\]

Let \( f_u(z_D) = F_u(z_D) - L_u[\delta_1 + \delta_2 z_D] \), one has:

\[
\frac{K_m u^2 D_1}{A B^2 R_{um}} \frac{d^2 \tilde{k}}{d z_D^2} - \frac{K_m u^2 D_1}{A B R_{um}} \frac{d \tilde{k}}{d z_D} - E_u \tilde{k} = -f_u(z_D), \quad (S29)
\]

where \( E_u = s + \epsilon_{um} + \mu_{umD} - \frac{\epsilon_{um} \epsilon_{uimD}}{s + \epsilon_{um} + \mu_{uimD}} \), \( F_u(z_D) = C_{umD}(r_D, z_D, t_{inj,D}) + \frac{\epsilon_{um} C_{uimD}(r_D, z_D, t_{inj,D})}{s + \epsilon_{um} + \mu_{uimD}} \) and \( f_u(z_D) = C_{umD}(r_D, z_D, t_{inj,D}) + \frac{\epsilon_{um} C_{uimD}(r_D, z_D, t_{inj,D})}{s + \epsilon_{um} + \mu_{uimD}} - \frac{K_m u^2 D_1}{A B R_{um}} \delta_2 - E_u(\delta_1 + \delta_2 z_D) \).
The general solution of Eq. (S24) is:
\[
\tilde{c}_{umD} = \int_1^{\infty} g_u(z_D, E_u; \eta_u) f_u(\eta_u) d\eta_u + \frac{x_D - x_{ed}}{1 - x_{ed}} \tilde{c}_{mD}(r_D, s), \quad z_D \geq 1. \tag{S30}
\]
where \( f_u(\eta_u) = C_{umD}(r_D, \eta_u, t_{inj,D}) + \frac{\epsilon_{um}C_{umD}(r_D, \eta_u, t_{inj,D})}{s + \epsilon_{um} + \mu_{umD}} - \frac{R_m v_{umD}^2}{A_B R_m \eta_u} s^2 - E_u(s_1 + s_2 \eta_u), \eta_u \)
is a positive value varying between 1 and \( \infty \) (e.g. \( 1 \leq \eta_u \leq \infty \)); \( g_u(z_D, E_u; \eta_u) \) is the Green's function, and could be expressed as:
\[
g_u(z_D, E_u; \eta_u) = \begin{cases} g_{u1}(z_D, E_u; \eta_u) = N_1 \exp(a_1 z_D) + N_2 \exp(a_2 z_D) & \text{if } 1 \leq z_D < \eta_u \\ g_{u2}(z_D, E_u; \eta_u) = N_3 \exp(a_1 z_D) + N_4 \exp(a_2 z_D) & \text{if } \eta_u \leq z_D < \infty \end{cases} \tag{S31}
\]
where \( N_1, N_2, N_3 \) and \( N_4 \) are coefficients to be determined using the following conditions

a) \( g_u(z_D, E_u; \eta_u) \) satisfying the model of Eqs. (S29) and (S27a)-(S27b);
b) \( g_{u1}(z_D, E_u; \eta_u) = g_{u2}(z_D, E_u; \eta_u); \)
c) \( \left. \frac{dg_{u2}}{dz_D} \right|_{z_D = \eta_u} - \left. \frac{dg_{u1}}{dz_D} \right|_{z_D = \eta_u} = - \frac{AB^2 R_m}{R_m \alpha^2 D_u}, \)

Substituting Eq. (S31) into Eq. (S27a), one has:
\[
N_3 = 0. \tag{S32}
\]
Substituting Eq. (S31) into Eq. (S27b), one has:
\[
N_1 \exp(a_1) + N_2 \exp(a_2) = 0, \tag{S33a}
\]
According to Eq. (S33a), one has:
\[
N_1 = -N_2 \exp(a_2 - a_1). \tag{S33b}
\]
According to above condition of b), one has:
\[
N_1 \exp(a_1 \eta_u) + N_2 \exp(a_2 \eta_u) = N_4 \exp(a_2 \eta_u). \tag{S34}
\]
According to above condition of c), one has:
\[
N_4 a_2 \exp(a_2 \eta_u) - [N_1 a_1 \exp(a_1 \eta_u) + N_2 a_2 \exp(a_2 \eta_u)] = - \frac{AB^2 R_m}{R_m \alpha^2 D_u}. \tag{S35}
\]
In the chaser phase, the values of \( N_1, N_2, N_3 \) and \( N_4 \) could be determined by Eqs. (S33a)-(S35), namely:

\[
N_1 = -N_2 \exp(a_2 - a_1), \quad N_2 = \frac{-AB^2 R_{um}}{R_m a_r^2 D_u [(a_1 - a_2) \exp(a_2 - a_1) \exp(a_1 \eta_a)]} \quad \text{and} \quad N_3 = 0
\]

\[
N_4 = N_2 - N_2 \exp(a_2 - a_1) \exp(a_1 \eta_a - a_2 \eta_u).
\]

As for the analytical solution of the lower aquitard, one could use a similar approach as that used for deriving the analytical solution of the upper aquitard to obtain, and the general solution of Eq. (S26) could be described as:

\[
\bar{C}_{lD} = \int_{-1}^{\infty} g_l(z_D, E_l; \eta_l) f_l(\eta_l) d\eta_l + \frac{z_{D e} + z_D}{z_{D e} - 1} \bar{c}_{mD}(r_D, z_D, s), \quad z_D \leq -1. \quad \text{(S36a)}
\]

\[
g_l(z_D, E_l; \eta_l) = \begin{cases} 
    g_{l1}(z_D, E_l; \eta_l) = M_1 \exp(b_1 z_D) + M_2 \exp(b_2 z_D) - 1 \leq z_D < \eta_l, \\
    g_{l2}(z_D, E_l; \eta_l) = M_3 \exp(b_1 z_D) + M_4 \exp(b_2 z_D) \quad \eta_l \leq z_D < -\infty,
\end{cases} \quad \text{(S36b)}
\]

\[
f_l(\eta_l) = C_{lD}(r_D, \eta_l, t_{in,j,D}) + \frac{\varepsilon_{lim} C_{lim}(r_D, \eta_l t_{in,j,D})}{s + \varepsilon_{lim} + \mu_{limD}} + \frac{R_{lim} \varepsilon_{lim}^{2}}{z_{D e} - 1} - \bar{c}_{mD} E_l \frac{z_{D e} + \eta_l}{z_{D e} - 1}, \quad \text{(S36c)}
\]

where \( \eta_l \) is a negative value varying between \(-1 \) and \(-\infty \) (e.g. \(-1 \leq \eta_l \leq -\infty \) ); \( g_l(z_D, E_l; \eta_l) \) is the Green's function, \( E_l = s + \varepsilon_{lim} + \mu_{limD} \) and \( \varepsilon_{lim} = \frac{\varepsilon_{lim D}}{s + \varepsilon_{lim} \mu_{limD}} \), and the values of \( M_1, M_2, M_3 \) and \( M_4 \) could be described as:

\[
M_1 = -M_2 \exp(b_1 - b_2), \quad M_2 = \frac{-AB^2 R_{lim}}{R_m a_r^2 D_u \exp(b_2 \eta_l - b_1 \eta_l) - b_2 \exp(b_2 \eta_l)}, \quad M_3 = M_2 \exp(b_2 \eta_l - b_1 \eta_l) - M_2 \exp(b_1 - b_2), \quad \text{and} \quad M_4 = 0.
\]

In the chaser phase, the dimensional boundary conditions Eqs. (15a)-(15b) are transformed into dimensionless forms as:

\[
\beta_{cha,D} \left. \frac{\partial c_{mD}(r_D, t_D)}{\partial t_D} \right|_{r_D=r_{WD}} = C_{mD}(r_D, t_D), \quad t_{in,j,D} < t_D \leq t_{cha,D}, \quad \text{(S37a)}
\]

\[
c_{cha,mD}(r_D, t_D) \bigg|_{t_D=t_{in,j,D}} = c_{inj,mD}(r_D, t_D) \bigg|_{t_D=t_{in,j,D}} \cdot t_{in,j,D} < t_D \leq t_{cha,D}. \quad \text{(S37b)}
\]

where \( \beta_{cha,D} = \frac{v_{w,cha} r_{WD}}{\xi R_m a_r} \).
Conducting Laplace transform on Eqs. (S1a)-(S1b) in the chaser phase, one has:

\[ s\bar{C}_{mD} - C_{mD}(r_D, t_{inj,D}) = \frac{1}{r_D} \frac{\partial^2 \bar{C}_{mD}}{\partial r_D^2} - \frac{1}{r_D} \frac{\partial \bar{C}_{mD}}{\partial r_D} - (\varepsilon_m + \mu_m) \bar{C}_{mD} + \varepsilon_m \bar{C}_{imD} - \\
(\theta_{um} \frac{\alpha^2}{2 \theta_m B^2} \bar{C}_{umD} - \frac{\theta_{um} \alpha^2 u_m}{2 \theta_m B^2} \frac{\partial \bar{C}_{umD}}{\partial z_D}) \bigg|_{z_D=1} + (\theta_{im} \frac{\alpha^2}{2 \theta_m B^2} \frac{\partial \bar{C}_{imD}}{\partial z_D}) \bigg|_{z_D=-1}. \]

\[ r_D \geq r_{WD}. \quad (S38a) \]

\[ \bar{C}_{imD} = \frac{\varepsilon_{im}}{(s + \mu_{imD} + \varepsilon_{im})} \bar{C}_{mD} + \frac{c_{imD}(r_D, t_{inj,D})}{(s + \mu_{imD} + \varepsilon_{im})}, r_D \geq r_{WD}, \quad (S38b) \]

where \( C_{mD}(r_D, t_{inj,D}) \) and \( C_{imD}(r_D, t_{inj,D}) \) are respectively the mobile and immobile concentrations [ML\(^{-3}\)] of the aquifer at the end of the injection phase, which could be calculated by Eqs. (S21) and (S17b).

After substituting Eqs. (S30), (S36a)-(S36c) and (S38b) into Eq. (S38a), one has:

\[ \frac{1}{r_D} \frac{\partial^2 \bar{C}_{mD}}{\partial r_D^2} - \frac{1}{r_D} \frac{\partial \bar{C}_{mD}}{\partial r_D} - E_a \bar{C}_{mD} + F = 0, r_D \geq r_{WD}, \quad (S39) \]

where \( E_a = s + \varepsilon_m + \mu_mD - \frac{\varepsilon_m \varepsilon_{im}}{s + \mu_{imD} + \varepsilon_{im}} + \frac{\theta_{um} \alpha^2 u_m}{2 \theta_m B} - \frac{\theta_{im} \alpha^2 v_{im}}{2 \theta_m B^2} - \frac{1}{1 - z_{eD}} \frac{\theta_{um} \alpha^2 u_m}{2 \theta_m B^2} + \frac{1}{z_{eD} - 1} \frac{\theta_{im} \alpha^2}{2 \theta_m B^2} \]

and \( F = C_{mD}(r_D, t_{inj,D}) + \frac{\varepsilon_m c_{imD}(r_D, t_{inj})}{s + \mu_{imD} + \varepsilon_{im}}. \)

The boundary conditions of Eqs. (S37a)-(S37b) in Laplace domain becomes:

\[ \bar{C}_{cha,mD}(r_{WD}, s) = \frac{\beta_{cha,D}}{s \beta_{cha,D} + 1} C_{inj,mD}(r_D, t_D) \bigg|_{t_D=t_{inj,D}}. \quad (S40) \]

The boundary conditions of the wellbore and infinity in Laplace domain are:

\[ \left[ \bar{C}_{mD} \frac{\partial \bar{C}_{mD}(r_D, s)}{\partial r_D} \right]_{r=r_{WD}} = \frac{\beta_{cha,D}}{s \beta_{cha,D} + 1} C_{inj,mD}(r_D, t_D) \bigg|_{t_D=t_{inj,D}}, \quad (S41a) \]

\[ \bar{C}_{cha,mD}(r_{WD}, s) \bigg|_{r_{WD} \rightarrow \infty} = 0, \quad (S41b) \]

Similar to the model of the SWPP test in the injection phase, Eqs. (S39) and (S40)-(S41b) compose a model of the second-order ordinary differential equation (ODE) with boundary
conditions, however, the governing equation is an inhomogeneous differential equation. In this study, we use the Green’s function method to derive the analytical solution of Eq. (39).

Notice that the boundary condition of Eq. (41a) is inhomogeneous, and we need to homogenize it first. Assigning $\tilde{C}_{mD} = \Psi(r_D) + \delta_1 + \delta_2 r_D$, and substituting it into Eqs. (41a) and (41b) yields:

$$[\Psi(r_D, s) - \frac{\partial \Psi(r_D, s)}{\partial r_D}]_{r=r_{WD}} = 0,$$

(S42a)

$$\Psi(r_D, s)|_{r_D \to \infty} = 0,$$

(S42b)

where $\delta_1 = -\frac{\beta_{cha,D}}{s \beta_{cha,D} + 1} \frac{r_D|_{r_D \to \infty}}{r_{WD} - r_D|_{r_D \to \infty} - 1} C_{inj,mD}(r_D, t_D)|_{t_D = t_{inj}}$ and

$$\delta_2 = \frac{\beta_{cha,D}}{s \beta_{cha,D} + 1} \frac{1}{r_{WD} - r_D|_{r_D \to \infty} - 1} C_{inj,mD}(r_D, t_D)|_{t_D = t_{inj}}.$$

Defining a spatial operator: $L = -\left[\frac{d^2}{dr_D^2} - \frac{d}{dr_D} - r_D E_a\right]$, one has:

$$L \tilde{C}_{mD} = L[\Psi(r_D) + \delta_1 + \delta_2 r_D] = Fr_D.$$

(S43)

Let $\varphi(r_D) = Fr_D - L(\delta_1 + \delta_2 r_D)$, one has:

$$\frac{\partial^2 \varphi}{\partial r_D^2} - \frac{\partial \varphi}{\partial r_D} - r_D E_a \varphi = -\varphi(r_D).$$

(S44)

where $\varphi(r_D) = Fr_D - [\delta_2 + r_D E_a (\delta_1 + \delta_2 r_D)]$.

The general solution of Eqs. (42a) - (44) is:

$$\Psi(r_D, E_a; \eta) = \int_{r_{WD}}^{\infty} g(r_D, E_a; \eta) \varphi(\eta) d\eta.$$  

(S45)

where $\eta$ is a positive value varying between $r_{WD}$ and $\infty$ (e.g. $r_{WD} \leq \eta \leq \infty$); $g(r_D, E_a; \eta)$ is the Green's function, and could be expressed as:

$$g(r_D, E_a; \eta) = \begin{cases} g_1(r_D, E_a; \eta) = T_1 \exp \left( \frac{\chi_{cha}}{2} \right) A_1 \left( \frac{1}{r_{cha}} \right) B_1 \left( \frac{1}{r_{cha}} \right) r_{WB} \leq y_{cha} \leq \eta \\ g_2(r_D, E_a; \eta) = T_2 \exp \left( \frac{\chi_{cha}}{2} \right) A_1 \left( \frac{1}{r_{cha}} \right) B_1 \left( \frac{1}{r_{cha}} \right) \eta \leq y_{cha} \leq \infty. \end{cases}$$

(S46)
where \( \varphi(\eta) = F\eta - [\delta_2 + \eta E_a(\delta_1 + \delta_2\eta)] \), \( y_{cha} = r_D + \frac{1}{4E_a} \). As \( B_1(r_D) \) diverges when \( r_D \to \infty \), \( T_2 \) has to be zero. Substituting Eq. (S45) into Eq. (S42a), one has:

\[
\left[ g_1 - \frac{\partial g_1}{\partial r_D} \right]_{r_D=r_D} = 0, \tag{S47}
\]

According to Eq. (S47), one has:

\[
T_1 = -T_2X, \tag{S48}
\]

where \( X = \frac{1}{2B_1(E_a^{1/3}y_{cha, w}) - E_a^{1/3}B_1'(E_a^{1/3}y_{cha, w})}{2A_i(E_a^{1/3}y_{cha, w}) - E_a^{1/3}A_i'(E_a^{1/3}y_{cha, w})} \) and \( y_{cha, w} = r_{wD} + \frac{1}{4E_a} \).

According to above condition of b), one has:

\[
T_1A_i \left( E_a^{\frac{1}{3}}y_{cha} \bigg|_{r_D=\eta^+} \right) + T_2B_i \left( E_a^{\frac{1}{3}}y_{cha} \bigg|_{r_D=\eta^+} \right) = T_3A_i \left( E_a^{1/3}y_{cha} \bigg|_{r_D=\eta^+} \right). \tag{S49}
\]

According to above condition of c), one has:

\[
\left[ \frac{1}{2}T_3 \exp \left( \frac{y_{cha}}{2} \right) A_i \left( E_a^{\frac{1}{3}}y_{cha} \right) + E_a^{\frac{1}{3}}T_3 \exp \left( \frac{y_{cha}}{2} \right) A_i' \left( E_a^{\frac{1}{3}}y_{cha} \right) \right]_{r_D=\eta^-} - \]

\[
\left[ 0.5T_1 \exp \left( \frac{y_{cha}}{2} \right) A_i \left( E_a^{\frac{1}{3}}y_{cha} \right) + E_a^{\frac{1}{3}}T_1 \exp \left( \frac{y_{cha}}{2} \right) A_i' \left( E_a^{\frac{1}{3}}y_{cha} \right) \right]_{r_D=\eta^+} - \]

\[
\left[ \frac{1}{2}T_2 \exp \left( \frac{y_{cha}}{2} \right) B_i \left( E_a^{\frac{1}{3}}y_{cha} \right) + E_a^{\frac{1}{3}}T_2 \exp \left( \frac{y_{cha}}{2} \right) B_i' \left( E_a^{\frac{1}{3}}y_{cha} \right) \right]_{r_D=\eta^+} = -1. \tag{S50}
\]

For solution in the chaser phase, the values of \( T_1, T_2, T_3 \) and \( T_4 \) could be determined by Eqs. (S48) - (S50), namely:

\[
T_1 = -\frac{\pi A_i(y_{ext} \bigg|_{r_D=\eta^+})}{E_a^{1/3}} X, \ T_2 = \frac{\pi A_i(y_{ext} \bigg|_{r_D=\eta^+})}{E_a^{1/3}}, \ T_3 = \frac{\pi A_i(y_{ext} \bigg|_{r_D=\eta^+})}{E_a^{1/3}} \left[ B_i(y_{ext} \bigg|_{r_D=\eta^+}) - X \right] \quad \text{and} \quad T_4 = 0.
\]

\[\text{S1.3 Solutions in the rest phase: Eqs. (27a) - (27f)}\]

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In the rest phase, the flow velocity become zero, and the advection and dispersion terms drop out of the governing equations. After conducting Laplace transform on Eqs. (S2a)-(S2b), the following equations would be obtained:

\[(s + \varepsilon_{um} + \mu_{umD})\tilde{c}_{umD} - \varepsilon_{um}\tilde{c}_{umD} - C_{umD}(r_D, z_D, t_{cha,D}) = 0. \quad z_D \geq 1. \quad \text{(S51a)}\]

\[\tilde{c}_{umD} = \frac{\varepsilon_{um}}{s + \varepsilon_{um} + \mu_{umD}}\tilde{c}_{umD} + \frac{C_{umD}(r_D, z_D, t_{cha,D})}{s + \varepsilon_{um} + \mu_{umD}}, \quad z_D \geq 1, \quad \text{(S51b)}\]

Substituting Eq. (S51b) into Eq. (S51a), one has:

\[\left(s + \varepsilon_{um} + \mu_{umD} - \frac{\varepsilon_{um}\varepsilon_{um}}{s + \varepsilon_{um} + \mu_{umD}}\right)\tilde{c}_{umD} - C_{umD}(r_D, z_D, t_{cha,D}) - \frac{\varepsilon_{um}C_{umD}(r_D, z_D, t_{cha,D})}{s + \varepsilon_{um} + \mu_{umD}} = 0. \quad z_D \geq 1. \quad \text{(S52)}\]

Similarly, Eqs. (S3a) - (S3b) become:

\[(s + \varepsilon_{lm} + \mu_{lmD})\tilde{c}_{lmD} - \varepsilon_{lm}\tilde{c}_{lmD} - C_{lmD}(r_D, z_D, t_{cha,D}) = 0. \quad z_D \leq -1. \quad \text{(S53a)}\]

\[\tilde{c}_{lmD} = \frac{\varepsilon_{lm}}{s + \varepsilon_{lm} + \mu_{lmD}}\tilde{c}_{lmD} + \frac{C_{lmD}(r_D, z_D, t_{cha,D})}{s + \varepsilon_{lm} + \mu_{lmD}}, \quad z_D \leq -1, \quad \text{(S53b)}\]

Substituting Eq. (S45b) into Eq. (S45a), one has:

\[\left(s + \varepsilon_{lm} + \mu_{lmD} - \frac{\varepsilon_{lm}\varepsilon_{lm}}{s + \varepsilon_{lm} + \mu_{lmD}}\right)\tilde{c}_{lmD} - C_{lmD}(r_D, z_D, t_{cha,D}) - \frac{\varepsilon_{lm}C_{lmD}(r_D, z_D, t_{cha,D})}{s + \varepsilon_{lm} + \mu_{lmD}} = 0. \quad z_D \leq -1. \quad \text{(S54)}\]

According to Eqs. (S52) and (S54), one has:

\[\tilde{c}_{umD} = C_{umD}(r_D, z_D, t_{cha,D}) + \frac{\varepsilon_{um}C_{umD}(r_D, z_D, t_{cha,D})}{s + \varepsilon_{um} + \mu_{umD}}, \quad z_D \geq 1, \quad \text{(S55a)}\]

\[\tilde{c}_{lmD} = C_{lmD}(r_D, z_D, t_{cha,D}) + \frac{\varepsilon_{lm}C_{lmD}(r_D, z_D, t_{cha,D})}{s + \varepsilon_{lm} + \mu_{lmD}}, \quad z_D \leq -1, \quad \text{(S55b)}\]

where \(C_{umD}(r_D, z_D, t_{cha,D})\) and \(C_{umD}(r_D, z_D, t_{cha,D})\) are respectively the mobile and immobile concentrations \([\text{ML}^{-3}]\) of the upper aquitard at the end of the chaser phase, \(C_{lmD}(r_D, z_D, t_{cha,D})\)
and $C_{imD}(r_D, z_D, t_{cha,D})$ are respectively the mobile and immobile concentrations [ML$^{-3}$] of the lower aquitard at the end of the chaser phase.

Similarly, the dimensionless governing equation of the mobile zone during the rest phase is:

$$\frac{\partial c_{mD}}{\partial t_D} = -\varepsilon_m (C_{mD} - C_{imD}) - \mu_{mD} C_{mD}, r_D \geq r_{WD}. \quad (S56a)$$

$$\frac{\partial c_{imD}}{\partial t_D} = \varepsilon_{im} (C_{mD} - C_{imD}) - \mu_{imD} C_{imD}, r_D \geq r_{WD}. \quad (S56b)$$

Conducting Laplace transform to Eqs. (S56a) and (S56b) for the rest phase, one has:

$$s\bar{C}_{mD} - C_{mD}(r_D, t_{cha,D}) = -\varepsilon_m (\bar{C}_{mD} - \bar{C}_{imD}) - \mu_{mD} \bar{C}_{mD}, r_D \geq r_{WD}. \quad (S57a)$$

$$s\bar{C}_{imD} - C_{imD}(r_D, t_{cha,D}) = \varepsilon_{im} (\bar{C}_{mD} - \bar{C}_{imD}) - \mu_{imD} \bar{C}_{imD}, r_D \geq r_{WD}. \quad (S57b)$$

According to Eqs. (S57a)-(S57b), one has:

$$\bar{C}_{mD} = \frac{C_{mD}(r_D, t_{cha,D}) + \varepsilon_m C_{imD}(r_D, t_{cha,D})}{(s+\varepsilon_m+\mu_{mD}+\varepsilon_{im}).} \quad (S58a)$$

$$\bar{C}_{imD} = \frac{C_{imD}(r_D, t_{cha,D}) + \varepsilon_{im} \bar{C}_{mD}}{(s+\mu_{imD}+\varepsilon_{im})}. \quad (S58b)$$

1.4 Solutions in the extraction phase: Eqs. (28a) - (28g)

Contrary to the injection and chaser phases, the direction of advective flux is reversed in the extraction stage, Eqs. (S2a) and (S3a) are modified as:

$$\frac{\partial c_{umD}}{\partial t_D} = \frac{R_m a^2 D_u}{A B^2 R_{um}} \frac{\partial^2 c_{umD}}{\partial z_D^2} + \frac{R_m v_{um} a^2}{A B R_{um}} \frac{\partial c_{umD}}{\partial z_D} - \varepsilon_{um} (C_{umD} - C_{umD}), r_{umD} C_{umD}, \quad (S59a)$$

$$z_D \geq 1,$$

$$\frac{\partial c_{imD}}{\partial t_D} = \frac{R_m a^2 D_l}{A B^2 R_{im}} \frac{\partial^2 c_{imD}}{\partial z_D^2} - \frac{R_m v_{im} a^2}{A B R_{im}} \frac{\partial c_{imD}}{\partial z_D} - \varepsilon_{im} (C_{imD} - C_{imD}), r_{imD} C_{imD}, \quad (S59b)$$

$$z_D \leq -1,$$

Conducting Laplace transform on Eqs. (S2b) and (S59a), one has:
\[ s\bar{C}_{umD} - C_{umD}(r_D, z_D, t_{res,D}) = \frac{R_m a_D^2 D_u}{AB^2 R_{um}} \frac{\partial^2 C_{umD}}{\partial z_D^2} + \frac{R_m v_{um} a_T^2}{ABR_{um}} \frac{\partial C_{umD}}{\partial z_D} - \varepsilon_{um}(\bar{C}_{umD} - \bar{C}_{imD}) - \]
\[ \mu_{umD}\bar{C}_{umD}, \quad z_D \geq 1, \quad (S60a) \]
\[ \bar{C}_{imD} = \frac{\varepsilon_{uim}\bar{C}_{umD}}{s + \varepsilon_{uim} + \mu_{uimD}} + \frac{C_{umD}(r_D, z_D, t_{res,D})}{s + \varepsilon_{uim} + \mu_{uimD}}, \quad z_D \geq 1, \quad (S60b) \]

Substituting Eqs. (S60b) into Eq. (S60a), one can has:
\[ \frac{R_m a_D^2 D_u}{AB^2 R_{um}} \frac{\partial^2 C_{umD}}{\partial z_D^2} + \frac{R_m v_{um} a_T^2}{ABR_{um}} \frac{\partial C_{umD}}{\partial z_D} - (s + \varepsilon_{um} + \mu_{umD} - \frac{\varepsilon_{um}\varepsilon_{uim}}{s + \varepsilon_{uim} + \mu_{uimD}})\bar{C}_{umD} + \]
\[ C_{umD}(r_D, z_D, t_{res,D}) + \frac{\varepsilon_{uim}\bar{C}_{umD}(r_D, z_D, t_{res,D})}{s + \varepsilon_{uim} + \mu_{uimD}} = 0, \quad z_D \geq 1, \quad (S61) \]

Similarly, conducting Laplace transform on Eqs. (S3b) and (S59b), one has:
\[ s\bar{C}_{imD} - C_{imD}(r_D, z_D, t_{res,D}) = \frac{R_m a_D^2 D_l}{AB^2 R_{im}} \frac{\partial^2 \bar{C}_{imD}}{\partial z_D^2} - \frac{R_m v_{im} a_T^2}{ABR_{im}} \frac{\partial \bar{C}_{imD}}{\partial z_D} - \varepsilon_{im}(\bar{C}_{imD} - \bar{C}_{imD}) - \]
\[ \mu_{imD}\bar{C}_{imD}, \quad z_D \leq -1, \quad (S62a) \]
\[ \bar{C}_{imD} = \frac{\varepsilon_{lim}\bar{C}_{imD}}{s + \varepsilon_{lim} + \mu_{limD}} + \frac{C_{imD}(r_D, z_D, t_{res,D})}{s + \varepsilon_{lim} + \mu_{limD}}, \quad z_D \leq -1, \quad (S62b) \]

Substituting Eqs. (S62b) into Eq. (S62a), one has:
\[ \frac{R_m a_D^2 D_l}{AB^2 R_{im}} \frac{\partial^2 \bar{C}_{imD}}{\partial z_D^2} - \frac{R_m v_{im} a_T^2}{ABR_{im}} \frac{\partial \bar{C}_{imD}}{\partial z_D} - (s + \varepsilon_{im} + \mu_{imD} - \frac{\varepsilon_{im}\varepsilon_{lim}}{s + \varepsilon_{lim} + \mu_{limD}})\bar{C}_{imD} + \]
\[ C_{imD}(r_D, z_D, t_{res,D}) + \frac{\varepsilon_{lim}\bar{C}_{imD}(r_D, z_D, t_{res,D})}{s + \varepsilon_{lim} + \mu_{limD}} = 0, \quad z_D \leq -1, \quad (S63) \]

where \( C_{umD}(r_D, z_D, t_{res,D}) \) and \( C_{umD}(r_D, z_D, t_{res,D}) \) are respectively the mobile and immobile concentrations [ML\(^{-3}\)] of the upper aquitard at the end of the rest phase, \( C_{imD}(r_D, z_D, t_{res,D}) \) and \( C_{imD}(r_D, z_D, t_{res,D}) \) are respectively the mobile and immobile concentrations [ML\(^{-3}\)] of the lower aquitard at the end of the rest phase.

One could use a similar approach of obtaining the analytical solution of aquitards in the chaser phase to derive the solution of aquitards in the extraction phase. The general solution of (S61) is:
\[
\bar{C}_{u_mD} = \int_1^\infty g_u(z_D, E_u; \beta_u) f_u(\beta_u) d\beta_u + \frac{z_D - z_{ED}}{z_{ED}} \bar{C}_{mD}(r_D, s), \quad z_D \geq 1,
\]

(S64a)

\[
g_u(z_D, E_u; \beta_u) = \begin{cases} 
H_1 \exp(m_1 z_D) + H_2 \exp(m_2 z_D) & 1 \leq z_D < \beta_u \\
H_3 \exp(m_1 z_D) + H_4 \exp(m_2 z_D) & \beta_u \leq z_D < \infty
\end{cases}
\]

(S64b)

\[
f_u(\beta_u) = C_{uM}(r_D, \beta_u, t_{res,D}) + \frac{\epsilon_{um} C_{u_{imD}}(r_D, \beta_u, t_{res,D})}{s + \epsilon_{um} + \mu_{umD}} + \frac{\kappa_{mum}^2 \bar{C}_{mD}(r_D, s)}{1 - z_{ED}} - \]

(S64c)

\[
\frac{\beta_u - z_{ED}}{z_{ED} - 1} E_u \bar{C}_{mD}(r_D, s),
\]

(S65c)

where \(\beta_u\) is a positive value varying between 1 and \(\infty\); \(\beta_l\) is a negative value varying between \(-1\) and \(-\infty\); \(g_u(z_D, E_u; \beta_u)\) and \(g_l(z_D, E_l; \beta_l)\) are the Green's functions, \(H_1 \sim H_4\) and \(l_1 \sim l_4\) are constants which could be determined by the boundary conditions and conditions of a)–c), the values of \(H_1 \sim H_4\) and \(l_1 \sim l_4\) are as follows:

\[
H_2 = \frac{-AB^2}{R_m a_r D_{u} |m_1 - m_2| \exp(m_2 - m_1) \exp(m_1 \beta_u)}.
\]

(S66)

\[
H_3 = 0, \quad H_4 = H_2 - H_2 \exp(m_2 - m_1) \exp(m_1 \beta_u - m_2 \beta_u).
\]

(S67)

\[
l_1 = -l_2 \exp(n_1 - n_2), \quad l_2 = \frac{-AB^2 R_{im}}{R_m a_r D_{u} |\exp(n_2 \beta_l - n_1 \beta_l) - n_2 \exp(n_2 \beta_l)|}.
\]

(S68)

\[
l_3 = l_2 \exp(n_2 \beta_l - n_1 \beta_l) - l_2 \exp(n_1 - n_2), \quad l_4 = 0.
\]

(S69)

\[
m_1 = \frac{-R_{mum}^2}{ABR_{um}} \left( \sqrt{\frac{R_{mum} a_r D_{u}}{ABR_{um}}} \right)^2 + \frac{4R_{mum}^2 D_{u}}{AB^2 R_{um}} (s + \epsilon_{um} + \mu_{umD} - \epsilon_{um} \epsilon_{umD}) \frac{\epsilon_{um} \epsilon_{umD}}{s + \epsilon_{umD} + \epsilon_{umD}}\right),
\]

(S70)
Thus, contrary to the injection and chaser phases, the direction of advective flux is

reversed in the extraction stage, and Eq. (S1a) is modified as:

\[
\frac{\partial C_{mD}}{\partial t_D} = \frac{1}{r_D} \frac{\partial^2 C_{mD}}{\partial r_D^2} + \frac{1}{r_D} \frac{\partial C_{mD}}{\partial r_D} - \epsilon_m (C_{mD} - C_{imD}) - \mu_{mD} C_{mD} - \left( -\frac{\theta_{um} \alpha^2 \nu_{um}}{2A \theta_m B} C_{umD} - \frac{\theta_{um} \alpha^2 \nu_{um}}{2AB^2 \theta_m} \frac{\partial C_{mD}}{\partial z_D} \right) \bigg|_{z_D = 1} + \left( -\frac{\theta_{im} \alpha^2 \nu_{im}}{2AB^2 \theta_m} \frac{\partial C_{imD}}{\partial z_D} \right) \bigg|_{z_D = -1}, \quad r_D \geq r_{WD}. \quad (S66)
\]

In the extraction phase, the dimensional boundary conditions Eqs. (14a)-(14b) are transformed to the dimensionless format:

\[
\beta_{extD} \frac{\partial C_{mD}(r_D,t_D)}{\partial t_D} \bigg|_{r_D = r_{WD}} = \frac{\partial C_{mD}(r_D,t_D)}{\partial r_D} \bigg|_{r_D = r_{WD}}, \quad t_{res,D} < t_D \leq t_{ext,D} \quad (S67a)
\]

\[
C_{mD}(r_D, t_D) \big|_{t_D = t_{res,D}} = C_{res,mD}(r_D, t_D) \big|_{t_D = t_{res,D}}. \quad (S67b)
\]

where \( \beta_{extD} = -\frac{V_{w,ext} r_{WD}}{\xi_{Rm} \alpha_r} \).

Conducting Laplace transform on Eqs. (S58) and (S1b) in the extraction phase, one has:

\[
s \tilde{C}_{mD} - \tilde{C}_{mD}(r_D, t_{res}) = \frac{1}{r_D} \frac{\partial^2 \tilde{C}_{mD}}{\partial r_D^2} + \frac{1}{r_D} \frac{\partial \tilde{C}_{mD}}{\partial r_D} - (\epsilon_m + \mu_{mD}) \tilde{C}_{mD} + \epsilon_m \tilde{C}_{imD} - \left( -\frac{\theta_{um} \alpha^2 \nu_{um} \bar{C}_{umD}}{2A \theta_m b} - \frac{\theta_{um} \alpha^2 \nu_{um} \bar{D}_m \frac{\partial \tilde{C}_{umD}}{\partial z_D}}{2A \theta_m b} \right) \bigg|_{z_D = 1} - \left( \frac{\theta_{im} \alpha^2 \nu_{im} \bar{C}_{imD}}{2AB^2 \theta_m} + \frac{\theta_{im} \alpha^2 \nu_{im} \bar{D}_m \frac{\partial \tilde{C}_{imD}}{\partial z_D}}{2AB^2 \theta_m} \right) \bigg|_{z_D = -1},
\]

\[
r_D \geq r_{WD}. \quad (S68a)
\]
\[
\tilde{C}_{imD} = \frac{\epsilon_{im}}{s+\mu_{imD}+\epsilon_{im}} \tilde{C}_{mD} + \frac{c_{imD}(r_D, t_{res})}{s+\mu_{imD}+\epsilon_{im}}, r_D \geq r_{WD}, \tag{S68b}
\]

After substituting Eqs. (S64a)–(S65c) and Eq. (S68b) into Eq. (S68a), one has

\[
\frac{\partial^2 \tilde{c}_{mD}}{\partial r_D^2} + \frac{\partial \tilde{c}_{mD}}{\partial r_D} - r_D \tilde{\zeta} \tilde{c}_{mD} + r_D \Lambda = 0. \tag{S69}
\]

where \( \zeta = s + \epsilon_m + \mu_{mD} - \frac{\epsilon_{im}\epsilon_m}{s+\mu_{imD}+\epsilon_{im}} - \frac{\theta_{um}\alpha_r^2v_{um}}{2A\theta_mB} + \frac{\theta_{im}\alpha_r^2v_{im}}{2AB^2\theta_m} - \frac{1}{1-ze_D} \frac{\theta_{um}\alpha_r^2D_u}{2A\theta_mB} + \frac{1}{ze_D-1} \frac{\theta_{im}\alpha_r^2D_l}{2AB^2\theta_m} \).

\( \Lambda = C_{mD}(r_D, t_{res}) + \frac{\epsilon_m c_{imD}(r_D, t_{res})}{s+\mu_{imD}+\epsilon_{im}}, C_{imD}(r_D, t_{res}) \) and \( C_{mD}(r_D, t_{res}) \) represent the initial concentrations in the immobile and mobile domains of the SWPP test in the rest phase.

The boundary condition of Eqs. (S67a)-(S67b) in Laplace domain becomes:

\[
s\beta_{ext,D} \tilde{c}_{mD}(r_D, s)|_{r_D=r_{WD}} - \beta_{ext,D} C_{res,m}(r_D, t_D)|_{t_D=t_{res,D}} = \frac{\partial \tilde{c}_{mD}(r_D,s)}{\partial r_D} |_{r_D=r_{WD}}. \tag{S70}
\]

Similar to the model of the SWPP test in the injection phase, Eqs. (S5), (S61) and (S70) compose a model of the second-order ordinary differential equation (ODE) with boundary conditions. However, the governing equation is an inhomogeneous differential equation. In this study, we use the Green’s function method to derive the analytical solution of Eq. (S69).

Similar to Chen and Woodside [1988], Eq. (S69) could be transferred into a self-adjoint form:

\[
\frac{\partial^2 G}{\partial r_D^2} - \left( r_D \zeta + \frac{1}{4} \right) G = -\ell(r_D). \tag{S71}
\]

where \( G = \exp(r_D/2)\tilde{C}_{mD} \) and \( \ell(r_D) = \exp(r_D/2)r_D\Lambda. \)

The boundary conditions of Eqs. (S5) and (S70) could be rewritten as:

\[
G(r_D, s)|_{r_D=\infty} = 0, \tag{S72a}
\]

\[
\left[ (s\beta_{ext,D} + \frac{1}{2}) G - \frac{\partial G}{\partial r_D} \right] |_{r_D=r_{WD}} = \beta_{ext,D} \exp(r_{WD}/2)C_{mD}(r_{WD}, t_{res,D}). \tag{S72b}
\]
One could find that the boundary condition of Eq. (S72b) is inhomogeneous, and we need to homogenize it first. Assigning \( G = U(r_D) + V(r_D) \) and \( V(r_D) = \sigma_1 + \sigma_2 r_D \), and substituting them into Eqs. (S72a) and (S72b) yields:

\[
U(r_D, s)|_{r_D = \infty} = 0, \quad (S73a)
\]

\[
\left( s \beta_{\text{ext}, D} + \frac{1}{2} \right) U - \frac{\partial U}{\partial r_D} \bigg|_{r_D = r_w D} = 0, \quad (S73b)
\]

where \( \sigma_1 = - \frac{\beta_{\text{ext}, D} \exp(r_{w D}/2) c_{m, D}(r_{w D}, t_{\text{res}, D})}{(s \beta_{\text{ext}, D} + \frac{1}{2}) r_{w D}^{1 - (s \beta_{\text{ext}, D} + \frac{1}{2}) r_D}|_{r_D = \infty}} \)

\[
\sigma_2 = \frac{\beta_{\text{ext}, D} \exp(r_{w D}/2) c_{m, D}(r_{w D}, t_{\text{res}, D})}{(s \beta_{\text{ext}, D} + \frac{1}{2}) r_{w D}^{1 - (s \beta_{\text{ext}, D} + \frac{1}{2}) r_D}|_{r_D = \infty}}.
\]

After defining a spatial operator: \( L = - \frac{d^2}{dr_D^2} + \left( r_D \zeta + \frac{1}{4} \right) \), one has:

\[
LG = LU(r_D) + LV(r_D) = \ell(r_D), \quad (S74)
\]

and

\[
LU(r_D) = \ell(r_D) - LV(r_D). \quad (S75)
\]

Let \( f(r_D) = \ell(r_D) - LV(r_D) \), one has:

\[
\frac{\partial^2 U}{\partial r_D^2} - \left( r_D \zeta + \frac{1}{4} \right) U = -f(r_D). \quad (S76)
\]

where \( f(r_D) = \exp(r_{D}/2) r_D \Lambda - \left( r_D \zeta + \frac{1}{4} \right) \left( \sigma_1 + \sigma_2 r_D \right) \).

Right now, the model with an inhomogeneous boundary condition becomes a regular Sturm-Louisville problem. The general solution of Eqs. (S73a) - (S73b) and (S76) is:

\[
U(r_D, \zeta; \varepsilon) = \int_{r_w D}^{\infty} g(r_D, \zeta; \varepsilon) f(\varepsilon) d\varepsilon. \quad (S77)
\]

where \( \varepsilon \) is a positive value varying between \( r_{w D} \) and \( \infty \) (e.g. \( r_{w D} \leq \varepsilon \leq \infty \)); \( g(r_D, \zeta; \varepsilon) \) is the Green's function, and could be expressed as:

\[
g(r_D, \zeta; \varepsilon) = \begin{cases} 
 g_1(r_D, \zeta; \varepsilon) = P_1 A_i(y_{\text{ext}}) + P_2 B_i(y_{\text{ext}}) & r_{w D} \leq y_{\text{ext}} \leq \varepsilon \\
 g_2(r_D, \zeta; \varepsilon) = P_3 A_i(y_{\text{ext}}) + P_4 B_i(y_{\text{ext}}) & \varepsilon \leq y_{\text{ext}} \leq \infty \end{cases} \quad (S78)
\]
where \( f(\varepsilon) = \exp(\varepsilon/2)\varepsilon A - \left( \varepsilon^2 + \frac{1}{4} \right) (\sigma_1 + \sigma_2\varepsilon) \), \( y_{\text{ext}} = \zeta^{1/3} \left( r_D + \frac{1}{4\zeta} \right) \), \( P_1 \), \( P_2 \), \( P_3 \) and \( P_4 \) are coefficients to be determined. As \( B_i(r_D) \) diverges when \( r_D \to \infty \), \( P_4 \) has to be zero.

Substituting Eq. (S78) into Eq. (S73b), one has:

\[
\left[ (s\beta_{\text{ext},D} + \frac{1}{2}) g_1 - \frac{\partial g_1}{\partial r_D} \right]_{r_D = r_{wD}} = 0, \tag{S79}
\]

which leads to

\[
P_1 = -P_2 W. \tag{S80}
\]

where \( W = \left( s\beta_{\text{ext},D} + \frac{1}{2} \right) B_i(y_{\text{ext},w}) - \zeta^{1/3} B_i'(y_{\text{ext},w}) \left( s\beta_{\text{ext},D} + \frac{1}{2} \right) A_i(y_{\text{ext},w}) - \zeta^{1/3} A_i'(y_{\text{ext},w}) \), \( y_{\text{ext},w} = \zeta^{1/3} \left( r_{wD} + \frac{1}{4\zeta} \right) \).

According to the properties of Green’s function, one has:

\[
P_1 A_i(y_{\text{ext}}|r_D = \varepsilon^+) + P_2 B_i(y_{\text{ext}}|r_D = \varepsilon^+) = P_3 A_i(y_{\text{ext}}|r_D = \varepsilon^-). \tag{S81}
\]

\[
\left[ P_3 \zeta^{1/3} A_i'(y_{\text{ext}}) \right]_{r_D = \varepsilon^-} - \left[ P_1 \zeta^{1/3} A_i'(y_{\text{ext}}) + P_2 \zeta^{1/3} B_i'(y_{\text{ext}}) \right]_{r_D = \varepsilon^+} = -1. \tag{S82}
\]

The values of \( P_1 \), \( P_2 \) and \( P_3 \) could be determined by Eqs. (S69) - (S71), namely:

\[
P_1 = -\frac{\pi A_i(y_{\text{ext}}|r_D = \varepsilon^+)}{\zeta^{1/3}}, \quad P_2 = \frac{\pi A_i(y_{\text{ext}}|r_D = \varepsilon^+)}{\zeta^{1/3}}, \quad P_3 = \frac{\pi A_i(y_{\text{ext}}|r_D = \varepsilon^+)}{\zeta^{1/3}} \left[ B_i(y_{\text{ext}}|r_D = \varepsilon^+) - W \right].
\]

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S2. Numerical simulations

To test the assumptions used in the analytical solution of this study, a 3D finite-element method with the help of COMSOL Multiphysics will be used to solve the three-dimensional model. The grid mesh of the aquifer-aquitard system in the numerical modeling could be seen in
Figure S1. The initial drawdown and the initial concentration are 0 for aquifer and aquitards. The hydraulic parameters are: $K_a=0.1$ m/day, $S_a=S_u=S_l=10^{-4}$ m$^{-1}$, and the other parameters are $R_m = R_{im} = R_{um} = R_{uim} = R_{lim} = R_{lum} = R_{lum}=1$, $\theta_{um} = \theta_{lim} = 0.1$, $\alpha_r = 2.5$ m, $\alpha_u = \alpha_l = 0.5$ m, $\mu_m = \mu_{im} = \mu_{um} = \mu_{uim} = \mu_{lim}=10^{-7}$ s$^{-1}$, $r_w = 0.5$ m, $Q_{inj} = Q_{cha} = 50$ m$^3$/d, $Q_{res} = 0$ m$^3$/d, $Q_{ext} = -50$ m$^3$/d, $t_{inj} = 250$ day, $t_{cha} = 50$ day, $t_{res} = 50$ day, $B = 10$ m, $\theta_m = 0.25$, $\theta_{im} = 0.05$, and $\omega = 0.01$ d$^{-1}$. In this modeling, the finite thickness of the aquitard is used to approximate the infinite thickness of the aquitard, and the finite radial length of the aquifer is used to approximate the infinite radial length of the aquifer. Such treatment works well when the tracer has not approach the boundary.

Figure S1. The grid mesh of the aquifer-aquitard system used in the Galerkin finite element program using COMSOL Multiphysics.
Figure S2. Spatial distribution of the flow velocity for different time. The parameters are the same with ones in Figures 2 and 3.

S3. References for Table 4

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S4. Parameter range used in sensitivity analysis

Table S1: parameter range used in sensitivity analysis

| Parameter | Unit | Range       |
|-----------|------|-------------|
| \(\alpha_u\) | m    | 0.05-0.50   |
| \(\alpha_r\) | m    | 0.50-1.00   |
| \(v_{um}\) | m/d  | 0-0.01      |
| \(\theta_{um}\) | -    | 0-0.2       |
| \(\omega\) | 1/s  | 0.0001-0.001|
| \(\theta_m\) | -    | 0.20-0.40   |
| \(V_w\) | m\(^3\) | 0.10-500    |

“-” represents dimensionless unit.