

ELECTRONIC SUPPLEMENTARY INFORMATION

Kinetic and thermodynamic determinants of trace metal partitioning at biointerphases: the role of intracellular speciation dynamics

Jérôme F.L. Duval,* Romain M. Présent, Elise Rotureau

CNRS, Laboratoire Interdisciplinaire des Environnements Continentaux (LIEC),
UMR 7360, Vandoeuvre-lès-Nancy F-54501, France.
Université de Lorraine, LIEC, UMR 7360, Vandoeuvre-lès-Nancy, F-54501, France.

*Corresponding author: Jérôme F.L. Duval. E-mail address: jerome.duval@univ-lorraine.fr (J.F.L. Duval)
Tel: + 33 3 83 59 62 63. Fax: + 33 3 83 59 62 55.

N.B. If not explicitly indicated in this document, meanings of symbols are those defined in the glossary reported at the end of the main text. Here-listed references are given at the end of the document.

I. 1. Detailed derivations of eqns (19), (27).

Derivation of eqn (19). After differentiation of eqn (10) with respect to time and combination with eqns (10)-(11), we obtain

$$\frac{dJ_u(t)}{dt} - \frac{dJ_M(t)}{dt} = k_e \left[(1 + \lambda) J_M(t) - \lambda J_u(t) + k_d^* \phi_u^T(t) \right], \quad (\text{S1})$$

, where we used the definition $\phi_u^T(t) = \phi_u^M(t) + \phi_u^C(t)$ for the total concentration of intracellular metal species (free and complexed) at t . Substituting the expression of $J_M(t)$ given by eqn (8) and further introducing the mass transfer resistant $R_T = 1 / (D_{M,\text{out}} \bar{f}_{el} a^{-1})$, eqn (S1) leads to

$$\frac{dJ_u(t)}{dt} + k_e \left(\lambda J_u(t) - k_d^* \phi_u^T(t) \right) - R_T^{-1} \left[\frac{dc_M^*(t)}{dt} + k_e (1 + \lambda) c_M^*(t) - \beta_a^{-1} \left(\frac{dc_M^a(t)}{dt} + k_e (1 + \lambda) c_M^a(t) \right) \right] = 0. \quad (\text{S2})$$

In addition, after combining eqn (13) with eqn (11), it comes

$$\int_a^{\xi_c} \xi^2 [c_M(\xi, t) - c_M(\xi, 0)] d\xi = -a^2 \left[\frac{\lambda}{1 + \lambda} \int_0^t J_u(\nu) d\nu + \frac{1}{1 + \lambda} (\phi_u^M(t) - \phi_u^{M,0}) - \frac{k_d^*}{1 + \lambda} \int_0^t \phi_u^T(\nu) d\nu \right]. \quad (\text{S3})$$

After substitution of eqn (17) into eqn (S3) and subsequent derivation with respect to time, we further obtain

$$\int_a^{\xi_c} \xi^2 \left\{ \frac{dc_M(\xi, t)}{dt} + k_e(1+\lambda)[c_M(\xi, t) - c_M(\xi, 0)] \right\} d\xi = a^2 \left\{ k_e \phi_u^{M,0} - J_u(t) - k_e \int_0^t [\lambda J_u(\nu) - k_d^* \phi_u^T(\nu)] d\nu \right\} \quad (S4)$$

Using eqn (16), eqn (S4) provides

$$\frac{J_u(t)}{J_u^*} - \Omega_1 \left(\frac{dc_M^*(t)}{dt} + k_e(1+\lambda)c_M^*(t) \right) - 2\beta_a^{-1}\Omega_2 \left(\frac{dc_M^a(t)}{dt} + k_e(1+\lambda)c_M^a(t) \right) + \frac{k_e}{J_u^*} \int_0^t [\lambda J_u(\nu) - k_d^* \phi_u^T(\nu)] d\nu + k_e \tau_o = 0 \quad (S5)$$

, where τ_o depends on the initial conditions as specified by eqn (20).

Eliminating the term $\frac{dc_M^*(t)}{dt} + k_e(1+\lambda)c_M^*(t)$ between eqn (S2) and eqn (S5), we finally obtain

$$\frac{\Delta\tau}{J_u^*} \frac{dJ_u(t)}{dt} + \frac{J_u(t)}{J_u^*} (k_e \lambda \Delta\tau - 1) - \frac{k_e k_d^*}{J_u^*} \Delta\tau \phi_u^T(t) - \frac{\tau_L}{K_M} \left(\frac{dc_M^a(t)}{dt} + k_e(1+\lambda)c_M^a(t) \right) - \frac{k_e}{J_u^*} \int_0^t [\lambda J_u(\nu) - k_d^* \phi_u^T(\nu)] d\nu - k_e \tau_o = 0 \quad (S6)$$

, where we used the relationship $\tau_L = -K_M \beta_a^{-1} (\Omega_1 + 2\Omega_2)^{-1}$ and $\Delta\tau = \tau_L - \tau_E = J_u^* \Omega_1 R_T$ derived from

eqn (22) in the main text. Introducing the dimensionless variable $\Sigma(t) = \int_0^t (J_u(\nu) / J_u^*) d\nu$ and

$\Phi^T(t) = \int_0^t \phi_u^T(\nu) d\nu$ defined in the main text, eqn (S6) can be rewritten in the form

$$\Delta\tau \Sigma_{tt} + \Sigma_t (k_e \lambda \Delta\tau - 1) - k_d^* \Delta\tau \Phi_t^T - \tau_L \frac{\Sigma_{tt}}{(1-\Sigma_t)^2} - k_e \tau_L (1+\lambda) \frac{\Sigma_t}{1-\Sigma_t} - k_e \lambda \Sigma(t) + k_d^* \Phi^T(t) - k_e \tau_o = 0 \quad (S7)$$

, where we used the relationships $J_u(t) / J_u^* = \Sigma_t$, $c_M^a(t) = K_M \Sigma_t / (1-\Sigma_t)$ and $\frac{dc_M^a(t)}{dt} = \frac{K_M \Sigma_{tt}}{(1-\Sigma_t)^2}$.

After rearrangements, eqn (S7) finally becomes

$$\Sigma_{tt} = \frac{(1-\Sigma_t)^2 \left[k_e (\tau_o + \lambda \Sigma(t)) - k_d^* [\Phi^T(t) - \Phi_t^T \Delta\tau] \right] + \Sigma_t (1-\Sigma_t) \left[(1-\Sigma_t) (1 - \lambda k_e \Delta\tau) + k_e \tau_L (1+\lambda) \right]}{\Delta\tau (1-\Sigma_t)^2 - \tau_L} \quad (S8)$$

, which is eqn (19) in the main text.

Derivation of eqn (27). Rewriting eqn (S2) in terms of $\Sigma(t) = \int_0^t (J_u(v)/J_u^*)dv$ and $\Phi^T(t) = \int_0^t \bar{\phi}_u^T(v)dv$, we obtain after arrangements

$$\frac{d\bar{c}_M^*(t)}{dt} = -k_e(1+\lambda)\bar{c}_M^*(t) + \left\{ \Sigma_{tt} \left(Bn^{-1} + \frac{1}{(1-\Sigma_t)^2} \right) + k_e \Sigma_t \left(\lambda Bn^{-1} + \frac{1+\lambda}{1-\Sigma_t} \right) - Bn^{-1} k_d^* \Phi_t^T \right\} / x_o \quad (S9)$$

, where we used the relationship $R_T J_u^* = \beta_a^{-1} Bn^{-1} K_M$ recalling that $Bn^{-1} = R_T / R_S$, $J_u^* = K_H k_{int} K_M$ and $R_S = 1 / (k_{int} K_H \beta_a)$. Equation (S9) identifies with eqn (27) in the main text.

I. 2. Demonstration of eqns (23), (24).

Demonstration of eqn (23). Under the electrostatic conditions detailed in the main text ($\psi(a \leq r \leq r_o) \equiv \langle \psi \rangle$ and $\psi(r_o \leq r \leq r_c) = 0$), the time constant τ_L defined by eqn (21) reduces to^{1,2}

$$\tau_L = \frac{1}{S_a K_H k_{int}} \left(V_{soft} + \frac{V_p}{\beta_a \varphi} \right). \quad (S10)$$

Further introducing the critical volume fraction $\varphi^* = k_e V_p / (S_a K_H k_{int} \beta_a)$ in eqn (S10), we obtain the dimensionless form of τ_L provided by

$$k_e \tau_L = \varphi^* (\bar{V} + \varphi^{-1}) \quad (S11)$$

, which is eqn (23) in the main text with $\bar{V} = \beta_a V_{soft} / V_p$.

Demonstration of eqn (24). The time constant τ_E is provided by the expression¹

$$\tau_E = \left\{ V_{soft} + V_p / (\varphi \beta_a) + k_{int} K_H a V_p / (\varepsilon \varphi D_{M,out}) \left[\varepsilon \alpha (1 - \gamma_o) + (1 - \alpha) / \beta_a - \gamma_a \varepsilon / 2 \right] \right\} / (S_a k_{int} K_H) \quad (S12)$$

with $\alpha = a / r_o$, $\gamma_o = r_o / r_c$ and $\gamma_a = a / r_c$. Equations (S11) and (S12) hold at sufficiently low volume fractions φ and for constant electrostatic potential inside the soft surface layer of the microorganism and zero potential at the electrolyte side of the interphase. Using eqn (S10), eqn (S12) can be rewritten

$$\tau_E = \tau_L \left[1 + Bn^{-1} \frac{\bar{f}_{el}}{(1 + \bar{V} \varphi)} \left(\alpha (1 - \gamma_o) + (\varepsilon \beta_a)^{-1} (1 - \alpha) - \gamma_a / 2 \right) \right]. \quad (S13)$$

Realizing that $r_c = r_o \varphi^{-1/3}$ and $\gamma_a = \alpha \varphi^{1/3}$, it comes

$$\tau_E = \tau_L \left\{ 1 + Bn^{-1} \frac{\bar{f}_{el}}{(1 + \bar{V} \varphi)} \left[(\varepsilon \beta_a)^{-1} \left(1 - \frac{a}{r_o} \right) + \frac{a}{r_o} \left(1 - \frac{3}{2} \varphi^{1/3} \right) \right] \right\}. \quad (S14)$$

In addition, with help of eqn (9) it can be shown that under the electrostatic conditions of interest in this

work \bar{f}_{el} reduces to $\bar{f}_{el} = \left[(\varepsilon\beta_a)^{-1} \left(1 - \frac{a}{r_o} \right) + \frac{a}{r_o} (1 - \varphi^{1/3}) \right]^{-1}$. Substituting into eqn (S14) and expanding

the result for $\varphi \ll 1$ that holds for dilute suspensions of microorganisms (case treated in the main text),

we obtain eqn (24) with $\zeta \approx 1 - \frac{\varphi^{1/3}/2}{1 + (\varepsilon\beta_a)^{-1}(r_o/a - 1)}$. The value of ζ is close to unity as the term

$1 + (\varepsilon\beta_a)^{-1}(r_o/a - 1)$ satisfies $1 + (\varepsilon\beta_a)^{-1}(r_o/a - 1) \geq 1$. Accordingly, for the sake of mathematical

simplification the φ -dependent term $\frac{\varphi^{1/3}/2}{1 + (\varepsilon\beta_a)^{-1}(r_o/a - 1)}$ involved in ζ was discarded for the

derivation of eqns (40) and (41) in the main text.

II. 1. Demonstration of eqns (29)-(31) and detailed expressions of the ratio

$$c_M^{*,\infty}(\bar{K}^*) / c_M^{*,\infty}(\bar{K}^* = 0).$$

Demonstration of eqns (29), (30). In the equilibrium regime reached at $t \rightarrow \infty$, eqn (25) provides

$$\Sigma_t(\infty) - \Phi_t^T(\infty) + \bar{\phi}_u^c(\infty) = 0 \quad \text{while eqn (26) yields } \Phi_t^T(\infty) = \left(1 + \bar{K}^{*-1} \right) \bar{\phi}_u^c(\infty).$$

Combination of these two equations further leads to

$$\Phi_t^T(\infty) = (1 + \bar{K}) \Sigma_t(\infty). \quad (\text{S15})$$

Substituting eqn (S15) into eqn (27) in the limit $t \rightarrow \infty$ where $\Sigma_{tt}(\infty) = 0$ and $d\bar{c}_M^*(\infty)/dt = 0$, it comes after rearrangements

$$c_M^{a,\infty} = \beta_a c_M^{*,\infty} \quad (\text{S16})$$

, where we used the relationships $\lambda k_e = k_a^* \rho_S^{V_i} \left(1 + \bar{K}^{*-1} \right) = k_d^* \left(1 + \bar{K}^* \right)$ derived from eqn (18) and from

$\bar{K}^* = k_a^* \rho_S^{V_i} / k_d^*$, and the equality $c_M^{a,\infty} = K_M \Sigma_t(\infty) / (1 - \Sigma_t(\infty))$. As argued in the text, eqn (S16)

legitimizes the systematic applicability of the (thermodynamic) BLM formalism at $t \rightarrow \infty$. Further

substitution of eqn (S15) into eqn (19) taken at $t \rightarrow \infty$ yields after simplification

$$\left[1 - \Sigma_t(\infty) \right] \left[k_e \tau_o + k_e \lambda \Sigma(\infty) - k_d^* \Phi^T(\infty) \right] + \Sigma_t(\infty) \left[1 - \Sigma_t(\infty) + k_e \tau_L (1 + \lambda) \right] = 0. \quad (\text{S17})$$

In the following, we express $\Sigma(\infty)$ and $\Phi^T(\infty)$ as a function of $c_M^{*,\infty}$, recalling that surface and bulk M concentrations are interrelated *via* eqn (S16) in the here-examined equilibrium regime. The integration of eqn (11) between $t = 0$ and $t \rightarrow \infty$ leads to

$$\phi_u^M(\infty) - \phi_u^{M,0} = J_u^* \Sigma(\infty) - (k_e + k_a^* \rho_S^{V_i}) x_M^\infty + k_d^* x_c^\infty \quad (\text{S18})$$

, where we introduced $x_M^\infty = \int_0^\infty \phi_u^M(t) dt$ and $x_c^\infty = \int_0^\infty \phi_u^c(t) dt$. Similarly, the integration of eqn (12) between $t = 0$ and $t \rightarrow \infty$ provides

$$\phi_u^c(\infty) - \phi_u^{c,0} = k_a^* \rho_S^{V_i} x_M^\infty - k_d^* x_c^\infty. \quad (\text{S19})$$

Solving eqn (S18)-(S19) in $x_{M,c}^\infty$, we obtain

$$\begin{cases} x_c^\infty = - \frac{\phi_u^M(\infty) - \phi_u^{M,0} - J_u^* \Sigma(\infty) + \left(1 + \frac{1 + \bar{K}^{*-1}}{\lambda}\right) (\phi_u^c(\infty) - \phi_u^{c,0})}{k_e \bar{K}^{*-1}} \\ x_M^\infty = - \frac{\phi_u^M(\infty) - \phi_u^{M,0} + \phi_u^c(\infty) - \phi_u^{c,0} - J_u^* \Sigma(\infty)}{k_e} \end{cases} \quad (\text{S20a,b})$$

Using eqn (S20), it is straightforward to show that $\Phi^T(\infty) = k_e (x_M^\infty + x_c^\infty) / J_u^*$ is defined after reduction by

$$\Phi^T(\infty) = -(1 + \bar{K}^*) \left[-\Sigma(\infty) + \frac{\phi_u^M(\infty) - \phi_u^{M,0} + (\phi_u^c(\infty) - \phi_u^{c,0})(1 + 1/\lambda)}{J_u^*} \right]. \quad (\text{S21})$$

Equation (12) at equilibrium further yields $\bar{\phi}_u^c(\infty) / \bar{\phi}_u^M(\infty) = \bar{K}^*$ while eqn (10) in the equilibrium limit leads to $\phi_u^M(\infty) = J_u(\infty) / k_e = J_u^* \Sigma_t(\infty) / k_e$. Substitution of these expressions into eqn (S21) gives

$$\Phi^T(\infty) = (1 + \bar{K}^*) \alpha_0 + (1 + \bar{K}^*) \left\{ \Sigma(\infty) - k_e^{-1} \Sigma_t(\infty) \left[1 + \bar{K}^* (1 + 1/\lambda) \right] \right\} \quad (\text{S22})$$

, where we introduced the constant $\alpha_0 = \left[\phi_u^{M,0} + \phi_u^{c,0} (1 + 1/\lambda) \right] / J_u^*$. From eqn (S22), we infer

$$(1 + \bar{K}^*) \Sigma(\infty) - \Phi^T(\infty) = -(1 + \bar{K}^*) \alpha_0 + (1 + \bar{K}^*) k_e^{-1} \Sigma_t(\infty) \left[1 + \bar{K}^* (1 + 1/\lambda) \right]. \quad (\text{S23})$$

Realizing that the term $k_e \lambda \Sigma(\infty) - k_d^* \Phi^T(\infty)$ in eqn (S17) can be rewritten in the form $k_d^* \left[(1 + \bar{K}^*) \Sigma(\infty) - \Phi^T(\infty) \right]$, it comes after combining eqn (S23) and eqn (S17)

$$A \Sigma_{t,\infty}^2 + B \Sigma_{t,\infty} + C = 0 \quad (\text{S24})$$

, where we wrote $\Sigma_{t,\infty} = \Sigma_t(\infty)$ for shortening notations, and the coefficients A , B and C are here defined by $A = -\left[1 + \lambda + \bar{K}^*(1 + \lambda)\right]$, $B = 1 + \lambda + \bar{K}^*(1 + \lambda) + k_e\tau_L(1 + \lambda) - k_e(\tau_o - \lambda\alpha_o)$ and $C = k_e(\tau_o - \lambda\alpha_o)$. The second-order polynomial equation (S24) in $\Sigma_{t,\infty}$ can be transformed into a second-order polynomial expression in terms of the variable $c_M^{a,\infty}/K_M$ with proper use of the equivalence $c_M^{a,\infty}/K_M = \Sigma_{t,\infty}/(1 - \Sigma_{t,\infty})$. After algebraic arrangements, the final result reads as

$$\left(c_M^{a,\infty}/K_M\right)^2 + \left[1 + \frac{1 + \bar{K}^*}{k_e\tau_L} + \frac{\tau_o - \lambda\alpha_o}{\tau_L(1 + \lambda)}\right]\left(c_M^{a,\infty}/K_M\right) + \frac{\tau_o - \lambda\alpha_o}{\tau_L(1 + \lambda)} = 0. \quad (\text{S25})$$

As $\tau_o - \lambda\alpha_o$ is always strictly negative, the discriminant of the quadratic eqn (S25) is positive. After calculation, it is demonstrated that the physically-relevant solution $c_M^{a,\infty}$ of eqn (S25) is then expressed by

$$c_M^{a,\infty}/K_M = \frac{1}{2} \left\{ -\left(1 + \frac{\mu_o}{\tau_L} + \frac{1 + \bar{K}^*}{k_e\tau_L}\right) + \frac{1 + \bar{K}^*}{k_e\tau_L} \left\{ 1 + \frac{2k_e\tau_L}{1 + \bar{K}^*} \left(1 + \frac{\mu_o}{\tau_L}\right) + \left[\frac{k_e\tau_L}{1 + \bar{K}^*} \left(1 - \frac{\mu_o}{\tau_L}\right)\right]^2 \right\}^{1/2} \right\} \quad (\text{S26})$$

, with $\mu_o = (\tau_o - \lambda\alpha_o)/(1 + \lambda)$ (< 0) and τ_o defined by eqn (20). Equation (29) in the main text directly follows from eqn (S26) using the equilibrium expression (S16). The time constant $-\mu_o$ in eqn (S26) simplifies after some developments into

$$k_e\mu_o = k_e \left(\Omega_1 c_M^{*,0} + 2\beta_a^{-1} \Omega_2 c_M^{a,0} \right) - \bar{\phi}_u^{\text{T},0}. \quad (\text{S27})$$

Using the relationship $\tau_L = -K_M \beta_a^{-1} (\Omega_1 + 2\Omega_2)$,¹ it comes

$$k_e\mu_o = \frac{k_e \Delta \tau}{J_u^* R_T} c_M^{*,0} \Delta \bar{c}_o - \frac{k_e \tau_L}{K_M} c_M^{a,0} - \bar{\phi}_u^{\text{T},0} \quad (\text{S28})$$

Further using the equality $R_T J_u^* = \beta_a^{-1} B n^{-1} K_M$ and the definition $x_o = \left[K_M / (\beta_a c_M^{*,0}) \right]^{-1}$, we finally obtain

$$k_e\mu_o = k_e \tau_L \left[(1 - \tau_E / \tau_L) \Delta \bar{c}_o x_o B n - c_M^{a,0} / K_M \right] - \bar{\phi}_u^{\text{T},0} \quad (\text{S29})$$

, which is eqn (30) in the main text.

Demonstration of eqn (31). Starting from eqn (29), the ratio $c_M^{*,\infty}(\bar{K}^*) / c_M^{*,\infty}(\bar{K}^* = 0)$ can be formulated according to

$$\frac{c_M^{*,\infty}}{c_M^{*,\infty}(\bar{K}^* = 0)} = \frac{-\left(1 + \frac{\mu_0}{\tau_L} + \frac{1 + \bar{K}^*}{k_e \tau_L}\right) + \frac{1 + \bar{K}^*}{k_e \tau_L} \left\{ 1 + \frac{2k_e \tau_L}{1 + \bar{K}^*} \left(1 + \frac{\mu_0}{\tau_L}\right) + \left[\frac{k_e \tau_L}{1 + \bar{K}^*} \left(1 - \frac{\mu_0}{\tau_L}\right) \right]^2 \right\}^{1/2}}{-\left(1 + \frac{\mu_0}{\tau_L} + \frac{1}{k_e \tau_L}\right) + \frac{1}{k_e \tau_L} \left\{ 1 + 2k_e \tau_L \left(1 + \frac{\mu_0}{\tau_L}\right) + \left[k_e \tau_L \left(1 - \frac{\mu_0}{\tau_L}\right) \right]^2 \right\}^{1/2}} \quad (\text{S30})$$

Under the peculiar condition $1 + k_e \mu_0 + k_e \tau_L = 0$, eqn (S30) simplifies into

$$\frac{c_M^{*,\infty}}{c_M^{*,\infty}(\bar{K}^* = 0)} = \frac{-\bar{K}^* + (1 + \bar{K}^*) \left[1 - \frac{2}{1 + \bar{K}^*} + \left(\frac{1 + 2k_e \mu_0}{1 + \bar{K}^*} \right)^2 \right]^{1/2}}{2[k_e \mu_0 (1 + k_e \mu_0)]^{1/2}} \quad (\text{S31})$$

, while for $1 + k_e \mu_0 + k_e \tau_L \neq 0$, eqn (S30) can be rewritten in the form

$$\frac{c_M^{*,\infty}}{c_M^{*,\infty}(\bar{K}^* = 0)} = \frac{-[1 + k_e \tau_L (1 + \mu_0 / \tau_L)] - \bar{K}^* + (1 + \bar{K}^*) \left[1 + \frac{2k_e \tau_L (1 + \mu_0 / \tau_L)}{1 + \bar{K}^*} + \left(\frac{k_e \tau_L (1 - \mu_0 / \tau_L)}{1 + \bar{K}^*} \right)^2 \right]^{1/2}}{-[1 + k_e \tau_L (1 + \mu_0 / \tau_L)] \left\{ 1 + \sigma \left[1 - \frac{4k_e^2 \mu_0 \tau_L}{[1 + k_e \tau_L (1 + \mu_0 / \tau_L)]^2} \right]^{1/2} \right\}} \quad (\text{S32})$$

, where we introduced $\sigma = \mp 1$ for $1 + k_e \tau_L (1 + \mu_0 / \tau_L) \gtrless 0$. The complex stability constant $\bar{K}_{1/2}^*$ introduced in §II.5 corresponds to the value taken by \bar{K}^* such that the equation $c_M^{*,\infty}(\bar{K}^* = \bar{K}_{1/2}^*) = c_M^{*,\infty}(\bar{K}^* = 0) / 2$ is verified. For situations where $1 + k_e \mu_0 + k_e \tau_L = 0$, this equation can be solved with use of eqn (S31) and, after developments, we obtain the following solution

$$\bar{K}_{1/2}^* = 3\sqrt{k_e \tau_L (1 + k_e \tau_L)} / 2 \quad (\text{S33})$$

, which is eqn (31a). For situations where $1 + k_e \mu_0 + k_e \tau_L \neq 0$, using eqn (S32) the equality

$c_M^{*,\infty}(\bar{K}^* = \bar{K}_{1/2}^*) = c_M^{*,\infty}(\bar{K}^* = 0) / 2$ leads after lengthy algebra and simplifications to the result

$$\bar{K}_{1/2}^* = \frac{3k_e^2 \tau_L \mu_0}{[1 + k_e \tau_L (1 + \mu_0 / \tau_L)] \left\{ 1 + \sigma \sqrt{1 - \frac{4k_e^2 \tau_L \mu_0}{[1 + k_e \tau_L (1 + \mu_0 / \tau_L)]^2}} \right\}} - \frac{1 + k_e \tau_L (1 + \mu_0 / \tau_L)}{2} \quad (\text{S34})$$

, which identifies with eqn (31b).

II. 2. Physical interpretation of the term $(1 + \bar{K}^*) / (k_e \tau_L)$.

As indicated in the main text (§II.5), eqns (11)-(12) considered at $t \rightarrow \infty$ provide $\bar{\phi}_u^c(\infty) / \bar{\phi}_u^M(\infty) = \bar{K}^*$ and $\bar{\phi}_u^T(\infty) = (1 + \bar{K}^*) \beta_a c_M^{*,\infty} / (K_M + \beta_a c_M^{*,\infty})$. The latter expression can be rearranged in the form $\bar{\phi}_u^T(\infty) = k_e^{-1} J_u^* (1 + \bar{K}^*) \beta_a c_M^{*,\infty} K_M^{-1} / (1 + \beta_a c_M^{*,\infty} / K_M)$. In addition, eqn (23) is equivalent to $k_e \tau_L = k_e \left[V_{\text{soft}} + (\beta_a \varphi)^{-1} V_p \right] / (K_H k_{\text{int}} S_a)$ so that $K_H k_{\text{int}}$ can be expressed as a function of τ_L according to $K_H k_{\text{int}} = \left[V_{\text{soft}} + (\beta_a \varphi)^{-1} V_p \right] / (S_a \tau_L)$. In turn, substituting the latter expression in the above equation defining $\bar{\phi}_u^T(\infty)$, we obtain

$$\frac{1 + \bar{K}^*}{k_e \tau_L} = \frac{S_a c_p \bar{\phi}_u^T(\infty)}{c_M^{*,\infty}} \times \frac{1 + \beta_a c_M^{*,\infty} / K_M}{1 + \bar{V} \varphi} \quad (\text{S35})$$

, where we further used the relationships $c_p = \varphi / V_p$ and $J_u^* = k_{\text{int}} K_H K_M$. The quantity $S_a c_p \bar{\phi}_u^T(\infty)$ in eqn (S35) corresponds to the amount of (free and complexed) metal forms accumulated at $t \rightarrow \infty$ in the overall (intracellular) volume occupied by the microorganisms present at a cell number density c_p . The term $(1 + \bar{V} \varphi)^{-1} \approx 1 - \bar{V} \varphi$ corrects c_p determined on the basis of the volume V_p of a microorganism for the presence of a soft peripheral corona where metals are not accumulated after internalisation. The term $1 + \beta_a c_M^{*,\infty} / K_M$ accounts for the finite number of internalisation sites at the membrane surface with the limits $\beta_a c_M^{*,\infty} / K_M \ll 1$ and $\beta_a c_M^{*,\infty} / K_M \gg 1$ corresponding to the Henry and saturation adsorption regimes, respectively. In turn, the quantity $(1 + \bar{K}^*) / (k_e \tau_L)$ is analogous to a dimensionless thermodynamic constant of an equilibrium reaction ($t \rightarrow \infty$) pertaining to the conversion of bulk free metal ions into internalized free and complexed metal forms in the overall microorganism suspension. In the (unrealistic) situation where $\bar{V} \rightarrow \infty$, which applies for microorganisms with very thick surface layer compared to the typical dimension of their cytoplasm, we have $(1 + \bar{K}^*) / (k_e \tau_L) \rightarrow 0$ meaning that the accumulation of metals is thermodynamically unfavorable, which is physically consistent. In addition, increasing \bar{K}^* at fixed $k_e \tau_L$ displaces the reaction toward the formation of ML_S complexes. Conversely, increasing $k_e \tau_L$ at fixed \bar{K}^* favors the excretion of internalized free metal forms and thus leads to a

decrease in $\phi_u^T(\infty)$. In the saturation regime $\beta_a c_M^{*,\infty} / K_M \gg 1$, we have $\frac{1 + \bar{K}^*}{k_e \tau_L} = \frac{\beta_a S_a c_p \phi_u^T(\infty)}{K_M (1 + \bar{V} \varphi)}$ whereas

the result $\frac{1 + \bar{K}^*}{k_e \tau_L} = \frac{S_a c_p \phi_u^T(\infty)}{c_M^{*,\infty} (1 + \bar{V} \varphi)}$ applies in the Henry regime $\beta_a c_M^{*,\infty} / K_M \ll 1$.

II. 3. Formal demonstration of the relationship $c_M^{*,\infty}(\bar{K}^* \gg 1) / c_M^{*,\infty}(\bar{K}^* = 0) \rightarrow 0$.

After simplification of eqn (S2) taken in the limit $t \rightarrow \infty$ under conditions allowing for a neglect of the intracellular ML_s complex dissociation step (condition satisfied for $\bar{K}^* \gg 1$), we obtain

$$\frac{\lambda}{1 + \lambda} R_T J_u^* \frac{c_M^{a,\infty}}{K_M + c_M^{a,\infty}} - (c_M^{*,\infty} - \beta_a^{-1} c_M^{a,\infty}) = 0 \quad (\text{S36})$$

, where we used eqn (7). After some algebra, eqn (S36) can be transformed in the polynomial equation

$$\left(x^{a,\infty}\right)^2 + x^{a,\infty} \left(1 - x^{*,\infty} + \frac{\lambda}{1 + \lambda} B n^{-1}\right) - x^{*,\infty} = 0 \quad (\text{S37})$$

, where we defined the dimensionless $x^{a,\infty} = c_M^{a,\infty} / K_M$ and $x^{*,\infty} = \beta_a c_M^{*,\infty} / K_M$. The equilibrium eqn (S37) (valid for $\bar{K}^* \gg 1$) must be satisfied irrespective of the value taken by $B n^{-1}$ and λ and it must further be consistent with the thermodynamic Boltzmann relationship $x^{a,\infty} = x^{*,\infty}$ (or $c_M^{a,\infty} = \beta_a c_M^{*,\infty}$) that necessarily applies at equilibrium (see main text, §5). The only solution satisfying these conditions is $x^{a,\infty} = x^{*,\infty} = 0$, which demonstrates that bulk metal concentration at $t \rightarrow \infty$ and at $\bar{K}^* \gg 1$ is 0, *i.e.* $c_M^{*,\infty}(\bar{K}^* \gg 1) / c_M^{*,\infty}(\bar{K}^* = 0) \rightarrow 0$.

II. 4. Detailed asymptotic behavior of eqn (29) for $\bar{K}^* \ll 1$ and $\bar{K}^* \gg 1$, and details on the behavior of eqn (31) at $k_e \tau_L \ll 1$ and $k_e \tau_L \gg 1$ (Table S1).

The Taylor series expansions of eqns (S31)-(S32) with respect to \bar{K}^* and $1/\bar{K}^*$ are useful for apprehending the behavior of $c_M^{*,\infty}(\bar{K}^*) / c_M^{*,\infty}(\bar{K}^* = 0)$ at $\bar{K}^* \ll 1$ and $\bar{K}^* \gg 1$, respectively. After calculations with help of Mathcad software (version 15, PTC), we obtain the results collected in **Table S1** and written in reduced forms. The expressions of $c_M^{*,\infty}(\bar{K}^*) / c_M^{*,0}$ (defined by eqn (29)) in the limits $\bar{K}^* \ll 1$ and $\bar{K}^* \gg 1$ are simply obtained from the results given in **Table S1** using the relation $c_M^{*,\infty}(\bar{K}^*) / c_M^{*,0} = c_M^{*,\infty}(\bar{K}^*) / c_M^{*,\infty}(\bar{K}^* = 0) \times c_M^{*,\infty}(\bar{K}^* = 0) / c_M^{*,0}$ where $c_M^{*,\infty}(\bar{K}^* = 0) / c_M^{*,0}$ is provided by

$$c_M^{*\infty}(\bar{K}^* = 0) / c_M^{*0} = \frac{1}{2x_0} \left\{ - \left(1 + \frac{\mu_0}{\tau_L} + \frac{1}{k_e \tau_L} \right) + \frac{1}{k_e \tau_L} \left\{ 1 + 2k_e \tau_L \left(1 + \frac{\mu_0}{\tau_L} \right) + \left[k_e \tau_L \left(1 - \frac{\mu_0}{\tau_L} \right) \right]^2 \right\}^{1/2} \right\} \quad (\text{S38})$$

, which is derived from eqn (29) taken at $\bar{K}^* = 0$.

Similarly, the limiting behavior of $\bar{K}_{1/2}^*$ at $k_e \tau_L \ll 1$ and $k_e \tau_L \gg 1$ is described by the Taylor series expansions provided in **Table S1** where we discriminate between cases $k_e \mu_0 > -1$, $k_e \mu_0 = -1$ and $k_e \mu_0 < -1$. The limiting expressions given in **Table S1** corresponds to the curves denoted as (a) and (b) in **Figures 2A-2B**.

	$c_M^{*\infty}(\bar{K}^* = 0)$
$\bar{K}^* \ll 1$:	$\left\{ \begin{array}{l} \text{for } 1 + k_e \mu_0 + k_e \tau_L = 0: \frac{c_M^{*\infty}}{c_M^{*\infty}(\bar{K}^* = 0)} \approx 1 - \frac{\bar{K}^*}{2\sqrt{k_e \mu_0(1 + k_e \mu_0)}} \\ \text{for } 1 + k_e \mu_0 + k_e \tau_L \neq 0: \frac{c_M^{*\infty}}{c_M^{*\infty}(\bar{K}^* = 0)} \approx 1 + \frac{\sigma \bar{K}^*}{[1 + k_e \tau_L(1 + \mu_0/\tau_L)] \left\{ 1 - \frac{4k_e^2 \mu_0 \tau_L}{[1 + k_e \tau_L(1 + \mu_0/\tau_L)]^2} \right\}^{1/2}} \end{array} \right.$
$\bar{K}^* \gg 1$:	$\left\{ \begin{array}{l} \text{for } 1 + k_e \mu_0 + k_e \tau_L = 0: \frac{c_M^{*\infty}}{c_M^{*\infty}(\bar{K}^* = 0)} \approx \frac{\sqrt{k_e \mu_0(1 + k_e \mu_0)}}{\bar{K}^*} \\ \text{for } 1 + k_e \mu_0 + k_e \tau_L \neq 0: \frac{c_M^{*\infty}}{c_M^{*\infty}(\bar{K}^* = 0)} \approx \frac{2k_e^2 \mu_0 \tau_L}{\bar{K}^* [1 + k_e \tau_L(1 + \mu_0/\tau_L)] \left\{ 1 + \sigma \left[1 - \frac{4k_e^2 \mu_0 \tau_L}{[1 + k_e \tau_L(1 + \mu_0/\tau_L)]^2} \right]^{1/2} \right\}} \end{array} \right.$
$k_e \tau_L \ll 1$:	$\left\{ \begin{array}{l} \text{for } k_e \mu_0 > -1: \bar{K}_{1/2}^* = 1 + k_e \mu_0 + \frac{1 - k_e \mu_0 / 2}{1 + k_e \mu_0} k_e \tau_L \\ \text{for } k_e \mu_0 = -1: \bar{K}_{1/2}^* = \frac{k_e \tau_L}{4} + \frac{3\sqrt{k_e \tau_L}}{2} \\ \text{for } k_e \mu_0 < -1: \bar{K}_{1/2}^* = -\frac{1 + k_e \mu_0}{2} - \frac{1/2 - k_e \mu_0}{1 + k_e \mu_0} k_e \tau_L \end{array} \right. \quad k_e \tau_L \gg 1: \bar{K}_{1/2}^* = 1 + k_e \tau_L - \frac{k_e \mu_0}{2}$

Table S1. Taylor series expansions for the ratio $c_M^{*\infty}(\bar{K}^*) / c_M^{*\infty}(\bar{K}^* = 0)$ at $\bar{K}^* \ll 1$ and $\bar{K}^* \gg 1$ (indicated) and for $\bar{K}_{1/2}^*$ in the limits $k_e \tau_L \ll 1$ and $k_e \tau_L \gg 1$ (indicated). Results are valid up to first order terms in \bar{K}^* or $1/\bar{K}^*$, $k_e \tau_L$ or $1/(k_e \tau_L)$.

II. 5. Evolution of the ratio $c_M^{*\infty}(\bar{K}^*) / c_M^{*\infty}(\bar{K}^* = 0)$ with \bar{K}^* at different $k_e \tau_L$ (Figure S1).

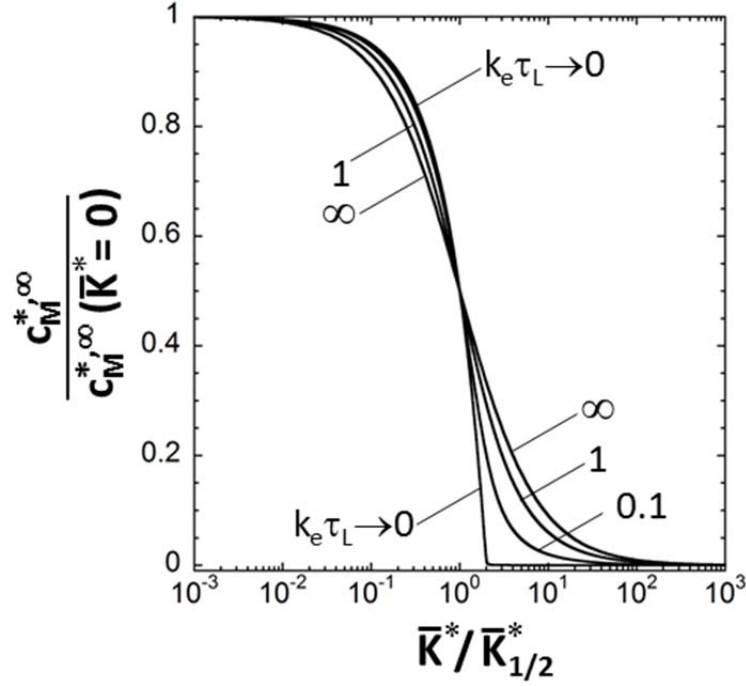


Figure S1. Dependence of the ratio $c_M^{*,\infty} / c_M^{*,\infty}(\bar{K}^* = 0)$ on $\bar{K}^* / \bar{K}_{1/2}^*$ at various values of $k_e \tau_L$ (indicated). Simulations are given for $k_e \mu_0 = -5/2$.

III. 1. Derivation of eqn (32) valid for strong ML_s complexes ($\bar{K}^* \gg 1$).

For strong intracellular ML_s complexes, eqn (27) becomes after rearrangements

$$\frac{dc_M^*(t)}{dt} = -k_e(1+\lambda)c_M^*(t) + \beta_a^{-1} \left\{ \frac{dc_M^a(t)}{dt} \left[1 + \frac{Bn^{-1}}{(1+c_M^a(t)/K_M)^2} \right] + k_e(1+\lambda)c_M^a(t) \left(1 + \frac{\frac{\lambda}{1+\lambda} Bn^{-1}}{1+c_M^a(t)/K_M} \right) \right\} \quad (\text{S39})$$

, where we used $\Sigma_t = c_M^a(t) / (K_M + c_M^a(t))$. The general solution of eqn (S39) can be written in the form

$$c_M^*(t) = C_1 e^{-k_e(1+\lambda)t} + C_2(t) e^{-k_e(1+\lambda)t} \quad (\text{S40})$$

, with C_1 a scalar independent of time and $C_2(t)$ is a function of time defined by

$$\frac{dC_2(t)}{dt} = K_M \beta_a^{-1} e^{k_e(1+\lambda)t} \left\{ \frac{dy(t)}{dt} \left[1 + \frac{Bn^{-1}}{(1+y(t))^2} \right] + k_e(1+\lambda)y(t) \left(1 + \frac{\frac{\lambda}{1+\lambda} Bn^{-1}}{1+y(t)} \right) \right\} \quad (\text{S41})$$

, where we introduced the normalized surface metal concentration $y(t) = c_M^a(t) / K_M$. Integration of eqn (S41) provides after calculation

$$C_2(t) = K_M \beta_a^{-1} e^{k_e(1+\lambda)t} \left\{ y(t) - Bn^{-1} \left[\frac{1}{1+y(t)} - k_e(1+\lambda) e^{-k_e(1+\lambda)t} \int_0^t \frac{e^{k_e(1+\lambda)\nu}}{1+y(\nu)} \left(1 + \frac{\lambda}{1+\lambda} y(\nu) \right) d\nu \right] \right\} \quad (S42)$$

The constant C_1 is obtained from the initial boundary condition $C_1 = c_M^{*,0} - C_2(0)$ with $C_2(0)$ defined from eqn (S42) according to $C_2(0) = K_M \beta_a^{-1} \left\{ y(0) - Bn^{-1} \left[\frac{1}{1+y(0)} \right] \right\}$. The steady state transport condition given by eqn (10) and considered at $t = 0$ further provides

$$c_M^{*,0} - K_M \beta_a^{-1} y(0) = J_u^* R_T \frac{y(0)}{1+y(0)} - k_e R_T \phi_u^{M,0} \quad (S43)$$

, where we used eqns (7)-(8) at $t = 0$. In turn, it comes $C_1 = K_M \beta_a^{-1} Bn^{-1} - k_e R_T \phi_u^{M,0}$ after realizing that the product $J_u^* R_T$ can be rewritten $K_M \beta_a^{-1} Bn^{-1}$. After rearrangements, C_1 reduces to

$$C_1 / c_M^{*,0} = x_o^{-1} Bn^{-1} (1 - \bar{\phi}_u^{M,0}) \text{ where we recall that } x_o = \left[K_M / (\beta_a c_M^{*,0}) \right]^{-1}. \text{ Finally, we obtain}$$

$$c_M^*(t) / c_M^{*,0} = x_o^{-1} Bn^{-1} (1 - \bar{\phi}_u^{M,0}) e^{-k_e(1+\lambda)t} +$$

$$x_o^{-1} \left\{ \frac{c_M^a(t)}{K_M} - Bn^{-1} \left[\frac{1}{1 + \frac{c_M^a(t)}{K_M}} - k_e(1+\lambda) e^{-k_e(1+\lambda)t} \int_0^t \frac{e^{k_e(1+\lambda)\nu}}{1 + \frac{c_M^a(\nu)}{K_M}} \left(1 + \frac{\lambda}{1+\lambda} \frac{c_M^a(\nu)}{K_M} \right) d\nu \right] \right\} \quad (S44)$$

, which is eqn (32) in the main text.

III. 2. Derivation of the expressions for the surface and bulk metal concentrations for $\bar{K}^* \gg 1$ in the regime of strong and weak affinity of M for the internalisation sites (eqns (33)-(34) and eqns (35)-(37), respectively). Simplifications of eqns (37)-(38) for situations where $k_o = k_+ = k_-$ and $k_- = k_e(1+\lambda)$.

Regime of strong M affinity for the internalisation sites in the limit $\bar{K}^ \gg 1$ (eqns (33)-(34)).*

In this regime, we have $K_M \ll c_M^a(t)$ at any time t so that $J_u(t) = J_u^*$. Simplifying eqn (S6) accordingly and discarding the ML_s dissociation terms not relevant in the $\bar{K}^* \gg 1$ limit, it comes

$$\frac{dc_M^a(t)}{dt} + k_e(1+\lambda)c_M^a(t) = -\frac{K_M}{\tau_L} [1 + k_e \lambda t + k_e(\tau_o - \lambda \Delta \tau)] \quad (S45)$$

After simple algebra, the integration of eqn (S45) leads to

$$c_M^a(t) / K_M = \left\{ \frac{c_M^{a,0}}{K_M} + \frac{[1 + k_e(1 + \lambda)(\tau_o - \lambda\Delta\tau)]}{k_e\tau_L(1 + \lambda)^2} \right\} e^{-k_e(1 + \lambda)t} - \frac{1 + k_e(1 + \lambda)[\tau_o + \lambda(t - \Delta\tau)]}{k_e\tau_L(1 + \lambda)^2} \quad (S46)$$

, which is eqn (33) in the main text. To derive $c_M^*(t)$, we now use eqn (S44) (or eqn (32)) in the limit

$J_u(t) = J_u^*$ or equivalently $y(t)/(1 + y(t)) \approx 1$ with $y(t) = c_M^a(t) / K_M$. It comes after simplification

$$c_M^*(t) = (K_M\beta_a^{-1}Bn^{-1} - k_eR_T\phi_u^{M,0})e^{-k_e(1 + \lambda)t} + \left[\beta_a^{-1}c_M^a(t) + \frac{\lambda}{1 + \lambda}K_M\beta_a^{-1}Bn^{-1}(1 - e^{-k_e(1 + \lambda)t}) \right] \quad (S47)$$

In addition, from eqns (8) and (28) we easily derive that the initial M transport flux $J_M(0)$ is given by

$$J_M(0) / J_u^* = x_o Bn \left[1 + \frac{1 + Bn^{-1}(1 - \bar{\phi}_u^{M,0})}{x_o} \right] \left\{ 1 - \left[1 - \frac{4x_o^{-1}Bn^{-1}[1 - \bar{\phi}_u^{M,0}(1 + x_o^{-1})]}{\{1 + x_o^{-1}[1 + Bn^{-1}(1 - \bar{\phi}_u^{M,0})]\}^2} \right]^{1/2} \right\} / 2. \quad (S48)$$

This expression identifies with that derived in Ref [1] in the limit $\bar{\phi}_u^{M,0} = 0$ where the dimensionless metal surface affinity parameter and bioconversion capacity of the microorganism noted A and B in Ref [1], respectively, are defined here by $A = x_o^{-1}$ and $B = x_o^{-1}Bn^{-1}$. At $t = 0$, the condition $J_u(0) = J_u^*$ applies and eqn (10) then simplifies into $J_M(0) / J_u^* = 1 - \bar{\phi}_u^{M,0}$. The latter expression is consistent with eqn (S48) provided that $x_o^{-1} \ll 1$ and $x_o^{-1}Bn^{-1}(1 - \bar{\phi}_u^{M,0}) \ll 1$. Simplifying eqn (28) in these limits, we obtain $c_M^{a,0} / (\beta_a c_M^{*,0}) = 1$ or, equivalently, $K_M\beta_a^{-1}Bn^{-1} - k_eR_T\phi_u^{M,0} = R_TJ_M(0) = 0$. In turn, eqn (S47) becomes

$$\beta_a c_M^*(t) / K_M = \left[c_M^a(t) / K_M + \frac{\lambda}{1 + \lambda} Bn^{-1} (1 - e^{-k_e(1 + \lambda)t}) \right] \quad (S49)$$

, which is eqn (34) in the main text.

Regime of weak M affinity for the internalisation sites in the limit $\bar{K}^* \gg 1$ (eqns (35)-(37)).

In this regime where the condition $K_M \gg c_M^a(t)$ applies, the uptake flux (eqn (7)) can be linearized according to $J_u(t) / J_u^* = c_M^a(t) / K_M = y(t)$. Equation (S6) taken in the extreme $\bar{K}^* \gg 1$ then reduces to

$$\frac{dy(t)}{dt} + \frac{1 + k_e\tau_L(1 + \lambda) - k_e\lambda\Delta\tau}{\tau_E} y(t) + \frac{k_e\tau_o}{\tau_E} + \frac{k_e\lambda}{\tau_E} \int_0^t y(\nu) d\nu = 0. \quad (S50)$$

Equation (S50) leads to the second-order differential equation in $\Sigma(t) = \int_0^t (J_u(v) / J_u^*) dv = \int_0^t y(v) dv$

$$\Sigma_{tt} + \frac{1 + k_e \tau_L + \lambda k_e \tau_E}{\tau_E} \Sigma_t + \frac{k_e \lambda}{\tau_E} \Sigma(t) = -\frac{k_e \tau_o}{\tau_E}. \quad (\text{S51})$$

The characteristic polynomial equation associated with eqn (S51) admits the discriminant Δ that reads after rearrangements

$$\Delta \tau_E^2 = (1 + k_e \tau_L)^2 + 2k_e \tau_E (k_e \tau_L - 1) \lambda + \lambda^2 (k_e \tau_E)^2 \quad (\text{S52})$$

The quantity $\Delta \tau_E^2$ is always ≥ 0 irrespective of the value of λ , which can be shown after evaluation of the discriminant of the second order polynomial eqn (S52) in λ . Let us first examined cases where $\Delta \tau_E^2 > 0$. Then, the general solution of eqn (S51) can be written in the form

$$\Sigma(t) = \alpha_+ e^{-k_+ t} + \alpha_- e^{-k_- t} - \tau_o / \lambda \quad (\text{S53})$$

, where α_{\pm} are independent of time and $k_{\pm} (\geq 0)$ (with $k_- \neq k_+$) are the two kinetic constants defined by

$$k_{\pm} \tau_E = -\left[1 + k_e (\tau_L + \lambda \tau_E)\right] \left\{ -1 \pm \sqrt{1 - \frac{4\lambda k_e \tau_E}{[1 + k_e (\tau_L + \lambda \tau_E)]^2}} \right\} / 2. \quad (\text{S54})$$

α_{\pm} are determined from the boundaries $\Sigma(0) = 0$ and $\Sigma_t(0) = y(0)$, which provides after evaluation

$$\alpha_{\pm} = \mp \left[\frac{c_M^{a,0} / K_M + k_{\mp} \tau_o / \lambda}{k_+ - k_-} \right]. \text{ Using the relationship } y(t) = \Sigma_t, \text{ we finally obtain}$$

$$c_M^a(t) / K_M = \sum_{j=+,-} A_j e^{-k_j t} \quad (\text{S55})$$

, with $A_{\pm} = -\alpha_{\pm} k_{\pm} = \pm k_{\pm} \left[\frac{c_M^{a,0} / K_M + k_{\mp} \tau_o / \lambda}{k_+ - k_-} \right]$, which corresponds to eqn (35) in the main text. The

expression for $c_M^*(t)$ in the weak M affinity regime is obtained from eqn (32) after substitution therein of

the linearized expressions $(1 + y(t))^{-1} \approx 1 - y(t)$ and $y(t) \left(1 + \frac{\lambda}{1 + \lambda} y(t)\right)^{-1} \approx 1 - y(t) / (1 + \lambda)$ with

$y(t) (= c_M^a(t) / K_M) \ll 1$ defined by eqn (S55). After lengthy calculation and algebraic arrangements, we

obtain the reduced expression

$$\frac{\beta_a c_M^*(t)}{K_M} = \frac{Bn^{-1}}{1+\lambda} \left\{ \sum_{j=+,-} \left[\frac{A_j}{1 - \frac{k_j}{k_e(1+\lambda)}} - (1+\lambda) \bar{\phi}_u^{M,0} \right] e^{-k_e(1+\lambda)t} + \sum_{j=+,-} \left[1 + \frac{\frac{\lambda Bn^{-1} \left(1 - \frac{k_j}{k_e \lambda}\right)}{1+\lambda}}{1 - \frac{k_j}{k_e(1+\lambda)}} \right] A_j e^{-k_j t} \right\} \quad (S56)$$

, where we used the relationship $J_u^* R_T = K_M \beta_a^{-1} Bn^{-1}$. Equation (S56) is identical to eqn (37) given in the main text. Finally, the value of $c_M^{a,0}$ is obtained from application of eqn (10) at $t=0$ with $J_u(0)/J_u^* = c_M^{a,0}/K_M$, *i.e.* $R_T^{-1} \left(c_M^{*,0} - \beta_a^{-1} c_M^{a,0} \right) = J_u^* c_M^{a,0}/K_M - k_e \bar{\phi}_u^{M,0}$ which leads to $c_M^{a,0}/K_M = \left(1 + Bn^{-1}\right)^{-1} \left(x_o + \bar{\phi}_u^{M,0} Bn^{-1}\right)$ as indicated in the main text.

Analysis of the case $k_+ = k_- = k_o$.

Let us now address the degeneracy case where $\Delta \tau_E^2 = 0$. This situation is achieved for $k_e \tau_L = 0$ and $\lambda k_e \tau_E = 1$ with the result $k_+ = k_- = k_o = \tau_E^{-1}$, which is inferred from eqn (36) in agreement with **Figure 4**. Then, the pendant of eqn (35) (or eqn (S55)) is $c_M^a(t)/K_M = (A_o + B_o t) e^{-k_o t}$ with $A_o = c_M^{a,0}/K_M$ and $B_o = -k_o \left(c_M^{a,0}/K_M + k_o \tau_o / \lambda \right)$. In addition, it can be shown after some algebra that the equivalent of eqn (37) in the limit $k_+ = k_- = k_o = \tau_E^{-1}$ is provided by

$$\frac{\beta_a c_M^*(t)}{K_M} = \frac{Bn^{-1}}{1+\lambda} \left[\frac{A_o - \frac{B_o}{k_e(1+\lambda) - k_o}}{1 - \frac{k_o}{k_e(1+\lambda)}} - (1+\lambda) \bar{\phi}_u^{M,0} \right] e^{-k_e(1+\lambda)t} + \left\{ \left[1 + \frac{\frac{\lambda Bn^{-1} \left(1 - \frac{k_o}{k_e \lambda}\right)}{1+\lambda}}{1 - \frac{k_o}{k_e(1+\lambda)}} \right] A_o + B_o \left[\left(1 + Bn^{-1}\right) t + \frac{k_e Bn^{-1}}{\left(k_e(1+\lambda) - k_o\right)^2} \left(1 - [k_e(1+\lambda) - k_o] t\right) \right] \right\} e^{-k_o t} \quad (S57)$$

In turn the expression of the transport flux $J_M(t)$ for $k_+ = k_- = k_o = \tau_E^{-1}$ is obtained from eqns (8) and (S57) together with $c_M^a(t)/K_M = (A_o + B_o t) e^{-k_o t}$, which yields

$$J_M(t)/J_u^* = \frac{1}{1+\lambda} \left[\frac{A_o - \frac{B_o}{k_e(1+\lambda) - k_o}}{1 - \frac{k_o}{k_e(1+\lambda)}} - (1+\lambda)\bar{\phi}_u^{M,0} \right] e^{-k_e(1+\lambda)t} + \left\{ \frac{\frac{\lambda A_o}{1+\lambda} \left(1 - \frac{k_o}{k_e\lambda}\right)}{1 - \frac{k_o}{k_e(1+\lambda)}} + B_o \left[t + \frac{k_e(1 - [k_e(1+\lambda) - k_o]t)}{(k_e(1+\lambda) - k_o)^2} \right] \right\} e^{-k_o t} \quad (S58)$$

Analysis of the case $k_- = k_e(1+\lambda)$.

We now examine cases where $k_- = k_e(1+\lambda)$, and the conditions leading to such a situation are detailed below (in particular it is shown that k_+ can not take the value $k_e(1+\lambda)$). Then, realizing that

$A_- \left(e^{[k_e(1+\lambda) - k_-]t} - 1 \right) / [k_e(1+\lambda) - k_-] \approx A_- t$, substitution of eqn (35) into eqn (32) now provides

$$\frac{\beta_a c_M^*(t)}{K_M} = \frac{Bn^{-1}}{1+\lambda} \left\{ \frac{A_+}{1 - \frac{k_+}{k_e(1+\lambda)}} - (1+\lambda)\bar{\phi}_u^{M,0} + \left(\frac{1+Bn^{-1}}{Bn^{-1}} - k_e t \right) (1+\lambda)A_- \right\} e^{-k_e(1+\lambda)t} + \left[1 + \frac{\frac{\lambda Bn^{-1}}{1+\lambda} \left(1 - \frac{k_+}{k_e\lambda}\right)}{1 - \frac{k_+}{k_e(1+\lambda)}} \right] A_+ e^{-k_+ t} \quad (S59)$$

, and the metal transport flux expression now reads as

$$J_M(t)/J_u^* = \frac{1}{1+\lambda} \left\{ \left[\frac{A_+}{1 - \frac{k_+}{k_e(1+\lambda)}} + (1+\lambda) \left[A_- (1 - k_e t) - \bar{\phi}_u^{M,0} \right] \right] e^{-k_e(1+\lambda)t} + \frac{1 - \frac{k_+}{k_e\lambda}}{1 - \frac{k_+}{k_e(1+\lambda)}} \lambda A_+ e^{-k_+ t} \right\} \quad (S60)$$

Evaluation of the conditions leading to $k_- = k_e(1+\lambda)$.

Let us first show that the condition $k_+ = k_e(1+\lambda)$ is never met. Finding the critical value Λ_c taken by $\Lambda = \lambda k_e \tau_E$ such that $k_+ = k_e(1+\lambda)$ comes to solve the equation $k_+ \tau_E = k_e \tau_E + \Lambda$ in Λ where $k_+ \tau_E$ is defined by eqn (36). After calculation, we obtain $\Lambda_c = k_e \tau_E (k_e \tau_E - k_e \tau_L)^{-1} (1 + k_e \tau_L - k_e \tau_E)$. We exclude here the case $\tau_E = \tau_L$ (*i.e.* $Bn^{-1} = 0$) for which the kinetic constant $k_e(1+\lambda)$ is not operational in the practical limit $\bar{\phi}_u^{M,0} = 0$ (ESI, part III.6). As $k_+ \tau_E \leq 1$ over the whole range of Λ (see **Figure 3A**), Λ_c must satisfy the condition $k_e \tau_E + \Lambda_c \leq 1$. In addition, the condition $\Lambda_c > 0$ further imposes that

$(k_e\tau_L <)k_e\tau_E < 1 + k_e\tau_L$. Substituting the above expression defining Λ_c into the inequality $k_e\tau_E + \Lambda_c \leq 1$, we get $1 \leq 1 - (k_e\tau_E)^{-1}$. This inequality is never verified because $k_e\tau_E \geq 0$, so that there are no conditions leading to $k_+ = k_e(1 + \lambda)$. We can now attempt finding the value of Λ_c such that $k_- = k_e(1 + \lambda)$. Adopting the same procedure as above, we again obtain the expression $\Lambda_c = k_e\tau_E(k_e\tau_E - k_e\tau_L)^{-1}(1 + k_e\tau_L - k_e\tau_E)$. However, Λ_c must now satisfy the condition $k_e\tau_E + \Lambda_c \geq 1 + k_e\tau_L$ as $k_-\tau_E \geq 1 + k_e\tau_L$ over the whole range of $\Lambda = \lambda k_e\tau_E$ (see **Figure 3B**). Substituting this latter inequality into the expression of Λ_c , we obtain $(k_e\tau_L <)k_e\tau_E < 1 + k_e\tau_L$, which is also consistent with the necessity to have positive values of Λ_c . In turn, the relationship $k_- = k_e(1 + \lambda)$ is verified at the value $\lambda = \lambda_c$ (specified below) provided that the $(k_e\tau_L, k_e\tau_E)$ couple is in line with the condition $(k_e\tau_L <)k_e\tau_E < 1 + k_e\tau_L : k_-\tau_E$ then reduces to $k_-\tau_E = (1 - \tau_L / \tau_E)^{-1}$ and the critical value of $\lambda_c = \Lambda_c / (k_e\tau_E)$ where $k_- = k_e(1 + \lambda_c)$ is given by $\lambda_c = [k_e(\tau_E - \tau_L)]^{-1} - 1$.

III. 3. Limits of eqns (35), (37) at $\lambda \rightarrow 0$.

It can be shown that A_+ involved in eqn (35) and (37) satisfies the limit $A_+|_{\lambda \rightarrow 0} \approx -k_e\tau_o / (1 + k_e\tau_L)$ while $A_-|_{\lambda \rightarrow 0} \approx c_M^{a,0} / K_M + k_e\tau_o / (1 + k_e\tau_L)$, where we used the limits $k_+|_{\lambda \rightarrow 0} \approx 0$, $k_-|_{\lambda \rightarrow 0} \approx (1 + k_e\tau_L) / \tau_E$. Replacing these limits into eqn (35) we obtain

$$\frac{c_M^a(t)}{K_M} = \left(\frac{c_M^a(0)}{K_M} + \frac{k_e\tau_o}{1 + k_e\tau_L} \right) e^{-t/\tau_d} - \frac{k_e\tau_o}{1 + k_e\tau_L} \quad (S61)$$

, where $\tau_d = \tau_E / (1 + k_e\tau_L)$. Equation (S61) correctly corresponds to the result derived in Ref [2] for microorganisms in the absence of intracellular ML_S formation and in the weak affinity limit. The following limits can be further shown

$$\frac{\lambda}{1 + \lambda} \frac{1 - \frac{k_-}{k_e\lambda}}{1 - \frac{k_-}{k_e(1 + \lambda)}} \Bigg|_{\lambda \rightarrow 0} \approx \frac{1 + k_e\tau_L}{1 + k_e(\tau_L - \tau_E)}, \frac{1}{k_e(1 + \lambda)} \sum_{j=+,-} \left(\frac{A_j}{1 - \frac{k_j}{k_e(1 + \lambda)}} \right) \Bigg|_{\lambda \rightarrow 0} \approx -\frac{\tau_o + \tau_E c_M^a(0) / K_M}{1 + k_e(\tau_L - \tau_E)}$$

, so that substitution into eqn (37) leads after some arrangements to

$$c_M^*(t) = -k_e e^{-k_e t} \left\{ \frac{K_M}{\beta_a} Bn^{-1} \left(\frac{\tau_o + \tau_E c_M^a(0) / K_M}{1 + k_e(\tau_L - \tau_E)} \right) + R_T \phi_u^{M,0} \right\} +$$

$$\frac{K_M}{\beta_a} e^{-t/\tau_d} \left(c_M^a(0) / K_M + \frac{k_e \tau_o}{1 + k_e \tau_L} \right) \left[1 + \frac{Bn^{-1}(1 + k_e \tau_L)}{1 + k_e(\tau_L - \tau_E)} \right] - \frac{K_M}{\beta_a} \frac{k_e \tau_o}{1 + k_e \tau_L}. \quad (S62)$$

This expression correctly compares with the result obtained in Ref [2] for cases where intracellular ML_S formation is not operational.

III. 4. Evolution of $k_+ \tau_E$ with $\lambda k_e \tau_E$ at different $k_e \tau_L$ (Figure S2).

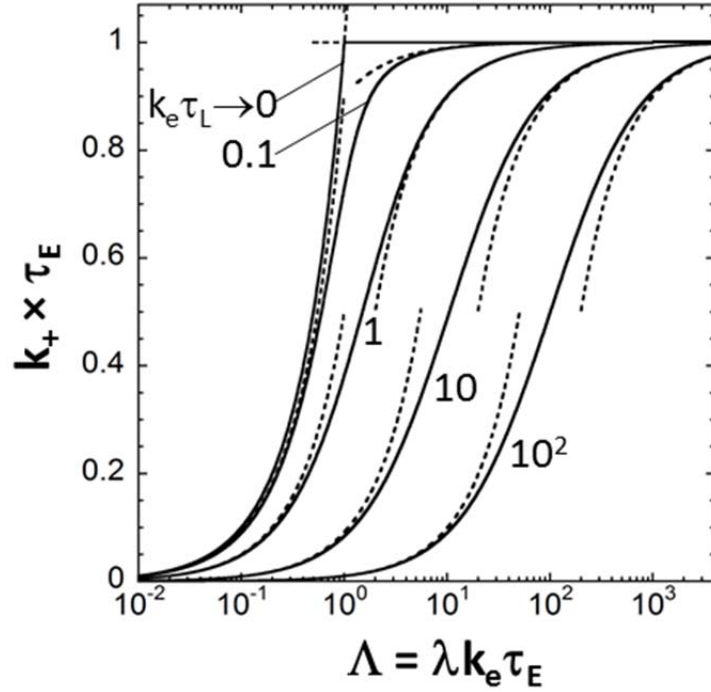


Figure S2. Dependence of the (dimensionless) kinetic constants $k_+ \tau_E$ on $\lambda k_e \tau_E = \Lambda$ at various values of $k_e \tau_L$ (indicated). Dotted lines represent the evolution of $k_+ \tau_E$ at low and large $\lambda k_e \tau_E = \Lambda$ and the corresponding analytical Taylor-series expressions are indicated in Figure 3A.

III. 5. Physical interpretation of the timescale $\tau_E / (k_a^* \rho_S^V \tau_E + k_e \tau_L)$, and comments on Table 1.

Using eqns (21)-(22), τ_E can be rewritten in the form

$$\tau_E = R_S \left(4\pi \int_a^{r_c} \xi^2 \beta_\xi d\xi / S_a - J_u^* \Omega_1 Bn^{-1} \right). \quad (S63)$$

In the following we define the Warburg-like element Z_W with value¹

$$Z_W = 4\pi \int_a^{r_c} \xi^2 \beta_\xi d\xi / S_a - J_u^* \Omega_1 Bn^{-1}. \quad (S64)$$

After algebraic rearrangements, Z_W can be expressed according to

$$Z_W = (\tau_E / \tau_L)(1 + \bar{V}\varphi)V_p / (S_a\varphi) \quad (\text{S65})$$

, where we used eqns (21)-(22). Equation (S65) corresponds to that provided in the main text (see caption

Table 1). In addition, the dimensionless product $k_e\tau_L = 4\pi k_e R_S \int_a^{r_c} \xi^2 \beta_\xi d\xi / S_a$ (eqn (21)) can be viewed

as the ratio between the M membrane transfer resistance R_S and the excretion resistance

$$R_{e,\varphi} = S_a \left(4\pi k_e \int_a^{r_c} \xi^2 \beta_\xi d\xi \right)^{-1}.$$

Under the electrostatic conditions specified in the main text (*i.e.* $1/\kappa \ll \delta$), $R_{e,\varphi}$ can be rewritten as $R_{e,\varphi} = R_e\varphi / (1 + \bar{V}\varphi)$ with $R_e = S_a / (k_e V_p)$ and $\bar{V} = \beta_a V_{\text{soft}} / V_p$. The

timescale $\tilde{\tau}_d = \tau_E / (k_a^* \rho_S^{V_i} \tau_E + k_e \tau_L)$ corresponds to $(k_-)^{-1}$ in the limit $k_a^* \rho_S^{V_i} \gg (1 + k_e \tau_L) / \tau_E$ (see main text). It can be written in the following form

$$\tilde{\tau}_d = R_S Z_W / \left(k_a^* \rho_S^{V_i} \tau_E + \frac{R_S}{R_{e,\varphi}} \right) \quad (\text{S66})$$

, or, equivalently,

$$\tilde{\tau}_d = \frac{\frac{R_S}{k_a^* \rho_S^{V_i} \tau_E} R_{e,\varphi}}{\left(R_{e,\varphi} + \frac{R_S}{k_a^* \rho_S^{V_i} \tau_E} \right)} Z_W. \quad (\text{S67})$$

Equation (S67) is analogous to that defining the characteristic time for discharging a (Warburg-like) element with value Z_W across an interface with charge transfer resistance $R_{ct} = \hat{R}_S R_{e,\varphi} / (R_{e,\varphi} + \hat{R}_S)$

with $\hat{R}_S = R_S / k_a^* \rho_S^{V_i} \tau_E$. R_{ct} is nothing else than the equivalent resistance for the resistances \hat{R}_S and $R_{e,\varphi}$ set in parallel, in agreement with the electrochemical circuit schemed in **Table 1** (case $(k_-)^{-1}$ for

the limit $k_a^* \rho_S^{V_i} \gg (1 + k_e \tau_L) / \tau_E$). Considering the inequality $k_a^* \rho_S^{V_i} \tau_E \gg (1 + k_e \tau_L) (> 1)$ defining the range of applicability of the limit $(k_-)^{-1} \approx \tilde{\tau}_d$, it is easy to verify that $\hat{R}_S / R_S \ll 1$ and $R_{e,\varphi} \gg \hat{R}_S$ (see

Table 1), the latter inequality stemming from $k_a^* \rho_S^{V_i} \tau_E \gg (1 + R_S / R_{e,\varphi}) > R_S / R_{e,\varphi}$. Stated differently,

fast ML_S formation kinetics leads to a decrease of the membrane transfer resistance R_S and microorganisms display a stronger propensity to accumulate M than excrete internalized metal ions. In

turn, fast ML_S formation facilitates the intracellular sequestration of metal ions in the form of ML_S complexes.

Following the above methodology, it is possible to evaluate expressions of charge transfer resistances associated to $1/k_+$. In the limit $k_a^* \rho_S^{V_i} \ll (1/2 + k_e \tau_L) / \tau_E$ (see **Table 1**), **Figure 3A** shows that $(k_+)^{-1} \approx \tau_E (1 + k_e \tau_L) / \Lambda$ with $\Lambda = \lambda k_e \tau_E$, or, equivalently, $(k_+)^{-1} \approx (k_a^* \rho_S^{V_i})^{-1} (1 + k_e \tau_L)$. As the inequality $k_a^* \rho_S^{V_i} \ll (1/2 + k_e \tau_L) / \tau_E (< (1 + k_e \tau_L) / \tau_E)$ applies, we infer $(k_a^* \rho_S^{V_i})^{-1} \gg \tau_E (1 + k_e \tau_L)^{-1}$, *i.e.* $(k_+)^{-1} \gg \tau_E$ (see **Table 1**). In addition, $(k_+)^{-1}$ can be written in the form $(k_+)^{-1} \approx \tau_E (k_a^* \rho_S^{V_i} \tau_E)^{-1} (1 + k_e \tau_L)$, or

$$(k_+)^{-1} = R_S Z_W \frac{1 + k_e \tau_L}{k_a^* \rho_S^{V_i} \tau_E} \quad (S68)$$

, which corresponds to the time for discharging the Warburg-like element Z_W across a charge transfer resistance defined here by $R_{ct} = R_S (1 + k_e \tau_L) / (k_a^* \rho_S^{V_i} \tau_E) \gg R_S$ (see **Table 1**). This means that a sluggish ML_S formation ($k_a^* \rho_S^{V_i} \ll (1/2 + k_e \tau_L) / \tau_E$) effectively leads to increasing the M membrane transfer resistance (blocking effect). In the extreme of fast intracellular ML_S formation ($k_a^* \rho_S^{V_i} \gg (1/2 + k_e \tau_L) / \tau_E (> (1 + k_e \tau_L) / \tau_E)$), we have $1/k_+ \approx \tau_E (1 - k_e \tau_L / \Lambda)^{-1}$ (see **Figure 3A**) or $1/k_+ \approx R_S Z_W \left[1 - (k_a^* \rho_S^{V_i} \tau_E)^{-1} R_S / R_{e,\varphi} \right]^{-1}$. Using $k_a^* \rho_S^{V_i} \tau_E \gg 1 + k_e \tau_L > k_e \tau_L (= R_S / R_{e,\varphi})$ that holds in the limit examined, it comes $1/k_+ \approx R_S Z_W \left[1 + (k_a^* \rho_S^{V_i} \tau_E)^{-1} k_e \tau_L \right]$, which corresponds to the time for discharging the Warburg-like element into a charge transfer resistance that identifies with $R_S \left[1 + (k_a^* \rho_S^{V_i} \tau_E)^{-1} k_e \tau_L \right] \approx R_S$, in line with the result reported in **Table 1**. The absence of any excretion contribution in this charge transfer resistance agrees with the limit $c_M^{*,\infty} / c_M^{*,\infty} (\bar{K}^* = 0) \rightarrow 0$ discussed in §II.5 for $\bar{K}^* \gg 1$ (**Table 1** applies for such strong ML_S complexes), recalling that excretion is required to obtain a finite non-zero M bulk concentration at equilibrium.²

As stated in the main text, it is possible to evaluate from **Table 1** which of the timescale $(k_+)^{-1}$ or $(k_-)^{-1}$ is operational in the regime of slow and fast intracellular ML_S formation (left and right column of **Table 1**, respectively). In the regime of slow ML_S formation, the timescale $(k_+)^{-1}$ is much larger than the free M transfer time τ_E between bulk and intracellular volume (see **Table 1** and **Figure 4**). Accordingly, it is the only $(k_-)^{-1}$ contribution that will be operational in defining kinetics of M bulk depletion and overall uptake. On the opposite, the membrane transfer resistance associated with $(k_-)^{-1}$ in the limit of fast ML_S formation significantly facilitates M uptake-depletion compared to that relevant for the $(k_+)^{-1}$ case. In line with this, the processes subsumed in the $(k_+)^{-1}$ component thus now play a key role in governing/limiting M accumulation and bulk depletion kinetics.

III. 6. Simplification of eqns (37)-(38) for $\bar{\phi}_u^{M,0} = 0$.

Obviously, in the limit of fast intracellular ML_S formation reached at $\lambda \rightarrow \infty$ we have $e^{-k_e(1+\lambda)t} \rightarrow 0$ in eqn (37) and the time constant $(1+\lambda)^{-1}/k_e$ is then not operational in determining M uptake/depletion kinetics. For $\lambda \rightarrow 0$, we showed in §III.3 that eqn (37) reduces to

$$c_M^*(t) = -k_e e^{-k_e t} \left\{ \frac{K_M}{\beta_a} Bn^{-1} \left(\frac{\tau_o + \tau_E c_M^a(0)/K_M}{1 + k_e(\tau_L - \tau_E)} \right) + R_T \bar{\phi}_u^{M,0} \right\} + \frac{K_M}{\beta_a} e^{-t/\tau_d} \left(c_M^a(0)/K_M + \frac{k_e \tau_o}{1 + k_e \tau_L} \right) \left[1 + \frac{Bn^{-1}(1 + k_e \tau_L)}{1 + k_e(\tau_L - \tau_E)} \right] - \frac{K_M}{\beta_a} \frac{k_e \tau_o}{1 + k_e \tau_L}. \quad (S69)$$

In addition, under the condition $K_M \gg c_M^a(t)$ marking the validity of eqn (S69), the time constant τ_o defined by eqn (20) may be written for $\lambda \rightarrow 0$ in the form $\tau_o = -c_M^{a,0} \tau_E / K_M - \bar{\phi}_u^{M,0} (1/J_u^* + k_e \Omega_1 R_T)$.

This equation is derived after combining the expression $\tau_E = -K_M \beta_a^{-1} [\Omega_1 (1 + Bn^{-1}) + 2\Omega_2]$ given in the Supporting Information of our previous work¹ with the relation $c_M^{a,0}/K_M = (1 + Bn^{-1})^{-1} (x_o + \bar{\phi}_u^{M,0} Bn^{-1})$ valid for $K_M \gg c_M^a(t)$ (see **ESI**, part III.2). In turn, for

$\bar{\phi}_u^{M,0} = 0$, we obtain $\tau_o = -c_M^{a,0} \tau_E / K_M$ and $\frac{K_M}{\beta_a} Bn^{-1} \left(\frac{\tau_o + \tau_E c_M^a(0)/K_M}{1 + k_e(\tau_L - \tau_E)} \right) + R_T \bar{\phi}_u^{M,0} = 0$. The

prefactor of the $e^{-k_e t}$ term in eqn (S69) becomes zero so that the time constant $1/k_e$ is not operational, as stated in the main text.

IV. 1. Time-dependence of M transport flux at the membrane surface under conditions of Figure 5 (Figure S3) and time-dependent ratio $c_M^* / c_M^{*,0}$ under conditions of Figure 7 (Figure S4).

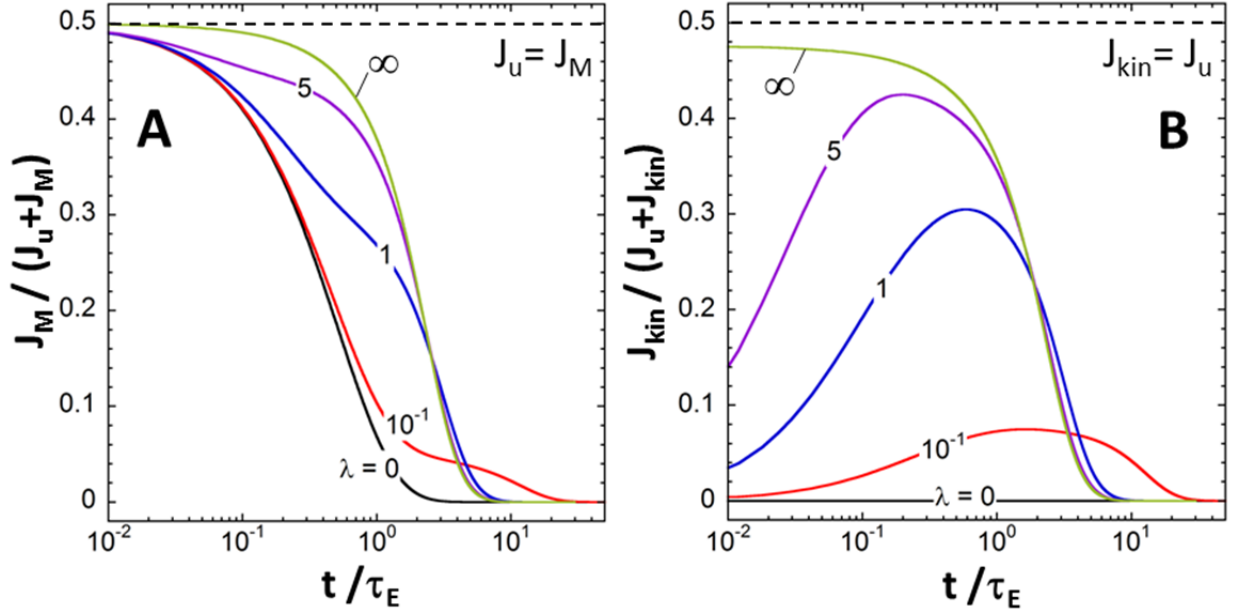


Figure S3. Evolution of the dimensionless ratio $J_M(t)/(J_u(t) + J_M(t))$ (A) and $J_{kin}(t)/(J_{kin}(t) + J_u(t))$ (B) with time t normalized by the M transfer timescale τ_E , at various values of λ (indicated). J_M and J_u are defined in the main text and $J_{kin}(t) = d\phi_u^c(t)/dt = k_a^* \rho_S^{V_i} \phi_u^M(t) - k_d^* \phi_u^c(t)$ corresponds to the net kinetic flux pertaining to intracellular ML_s complex formation. Model parameters: as in **Figure 5** of the main text. The dotted lines in (A) and (B) represent the limits $J_u = J_M$ and $J_{kin} = J_u$, respectively.

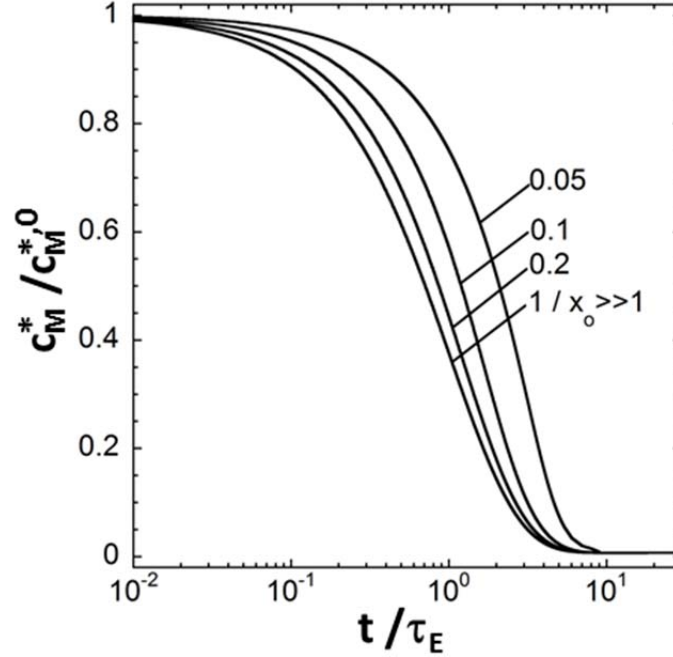


Figure S4. Evolution of the ratio $c_M^*/c_M^{*,0}$ versus t/τ_E for various values of the dimensionless M reciprocal affinity $1/x_0 = K_M/(\beta_a c_M^{*,0})$ (indicated). Model parameters: as in **Figure 7** of the main text.

IV. 2. Expressions of $c_M^{*,\infty}/c_M^{*,0}$ and k_+/k_- as a function of φ/φ^* and details on the corresponding asymptotic behavior at $\varphi/\varphi^* \ll 1$ and $\varphi/\varphi^* \gg 1$ (Table S2). Derivation of eqns (39)-(41).

Equation (29) defines the ratio $c_M^{*,\infty}/c_M^{*,0}$ according to

$$c_M^{*,\infty}/c_M^{*,0} = \frac{1}{2x_0} \left\{ - \left(1 + \frac{\mu_0}{\tau_L} + \frac{1 + \bar{K}^*}{k_e \tau_L} \right) + \frac{1 + \bar{K}^*}{k_e \tau_L} \left\{ 1 + \frac{2k_e \tau_L}{1 + \bar{K}^*} \left(1 + \frac{\mu_0}{\tau_L} \right) + \left[\frac{k_e \tau_L}{1 + \bar{K}^*} \left(1 - \frac{\mu_0}{\tau_L} \right) \right]^2 \right\}^{1/2} \right\} \quad (S70)$$

, with $k_e \mu_0 = k_e \tau_L \left[(1 - \tau_E / \tau_L) \Delta \bar{c}_0 x_0 B n - c_M^{a,0} / K_M \right] - \bar{\phi}_u^{T,0}$ (eqn (30)) which we rewrite in the form $k_e \mu_0 = -k_e \tau_L x_0 \left\{ 1 - \Delta \bar{c}_0 \left[1 - \zeta / (1 + \bar{V} \varphi) \right] \right\} - \bar{\phi}_u^{T,0}$ where we used eqn (24) valid for $\varphi \ll 1$. In turn we obtain $\mu_0 / \tau_L = -x_0 \left\{ 1 - \Delta \bar{c}_0 \left[1 - \zeta / (1 + \bar{V} \varphi) \right] \right\} - \bar{\phi}_u^{T,0} / (k_e \tau_L)$. Further using eqn (23) that defines $k_e \tau_L$ as a function of φ , we finally derive $\mu_0 / \tau_L = -x_0 \left\{ 1 - \Delta \bar{c}_0 \left[1 - \zeta / (1 + \bar{V} \varphi) \right] \right\} - \bar{\phi}_u^{T,0} / \left[\varphi^* (\varphi^{-1} + \bar{V}) \right]$. Substitution of that latter expression and of $k_e \tau_L = \varphi^* (\varphi^{-1} + \bar{V})$ (eqn (23)) into eqn (S70) provides the general relationship that determines the evolution of $c_M^{*,\infty}/c_M^{*,0}$ as a function of the microorganism volume

fraction φ in the suspension. Then, after solving the equation $c_M^{*,\infty}(\varphi_{1/2})/c_M^{*,0} = 1/2$, we obtain eqn (39) that defines the cell volume fraction $\varphi_{1/2}$ needed to achieve a two-fold reduction of the bulk M concentration at $t \rightarrow \infty$ under the condition $\Delta\bar{c}_0 = 0$. Using Mathcad software (version 15, PTC) and after simplifications, we further obtain the Taylor series expansions reported in **Table S2** for $c_M^{*,\infty}/c_M^{*,0}$ at $\varphi/\varphi^* \ll 1$ and $\varphi/\varphi^* \gg 1$. For the sake of simplicity, we derived these expressions in the limit where the term $\bar{V}\varphi$ is so small compared to unity (which is generally the case in practice) that it can be discarded in the mathematical developments.

As detailed in the main text, the kinetic constants $k_{\pm} (\geq 0)$ given by eqn (36) are fully defined upon upon the only specification of $k_e\tau_L$ and $\lambda k_e\tau_E (= k_a^*\rho_S^V\tau_E)$ and the same holds for the ratio k_+/k_- . The expressions of k_{\pm} and k_+/k_- as a function of cell volume fraction φ are simply obtained after substituting into eqn (36) the relationships defining the dependence of $k_e\tau_L$ and $\lambda k_e\tau_E$ on φ , *i.e.* $k_e\tau_L = \varphi^*(\varphi^{-1} + \bar{V})$ (which is eqn (23)) and $\lambda k_e\tau_E = \lambda\varphi^*(\varphi^{-1} + \bar{V})\left[1 + \zeta Bn^{-1}/(1 + \bar{V}\varphi)\right]$. Using Mathcad software (version 15, PTC), the first derivative of the ratio k_+/k_- with respect to φ can be easily computed. Then, the position and the value of the maximum in k_+/k_- can be determined from the value in φ where this derivative is zero. After lengthy simplifications and neglect of the φ -dependent component of ζ (see justification in **ESI**, Part I.2), the expression defining $(\varphi/\varphi^*)_{\max}$ where k_+/k_- is maximum can be written in the form given by eqn (40) and the value $(k_+/k_-)_{\max}$ can be arranged according to eqn (41). Finally, we derived the Taylor-series expansion of the ratio k_+/k_- at $\varphi/\varphi^* \ll 1$ and $\varphi/\varphi^* \gg 1$ and after arrangements we obtain the results in reduced form collected in **Table S2**.

$$c_M^{*,\infty} / c_M^{*,0}$$

$$\varphi / \varphi^* \ll 1:$$

$$\frac{c_M^{*,\infty}}{c_M^{*,0}} \approx 1 - \Delta\bar{c}_o(1-\zeta) + \frac{\varphi}{\varphi^*} \frac{1+\bar{K}^*}{x_o} \left\{ \frac{\bar{\varphi}_u^{T,0}}{1+\bar{K}^*} - \frac{x_o[1-\Delta\bar{c}_o(1-\zeta)]}{1+x_o[1-\Delta\bar{c}_o(1-\zeta)]} \right\}$$

$$\varphi / \varphi^* \gg 1:$$

$$\frac{c_M^{*,\infty}}{c_M^{*,0}} \approx \frac{1}{x_o(1+\bar{K}^*) \left[1 - (1+\bar{K}^*)^{-1} \bar{\varphi}_u^{T,0} \right]} \left\{ \bar{\varphi}_u^{T,0} + \frac{\varphi^*}{\varphi} \left[\frac{x_o \left[1 - (1+\bar{K}^*)^{-1} \bar{\varphi}_u^{T,0} \right] \left[1 - \Delta\bar{c}_o(1-\zeta) \right] - (1+\bar{K}^*)^{-1} \bar{\varphi}_u^{T,0}}{\left[1 - (1+\bar{K}^*)^{-1} \bar{\varphi}_u^{T,0} \right]^2} \right] \right\}$$

$$k_+ / k_-$$

$$\varphi / \varphi^* \ll 1: \quad \frac{k_+}{k_-} \approx \frac{\lambda(1+\zeta Bn^{-1})}{\left[1 + \lambda(1+\zeta Bn^{-1}) \right]^2} \frac{\varphi}{\varphi^*}$$

$$\varphi / \varphi^* \gg 1: \quad \frac{k_+}{k_-} \approx \frac{1-(1-\chi)^{1/2}}{1+(1-\chi)^{1/2}} \left\{ 1 + \frac{4\lambda \left[1 + \zeta Bn^{-1} - \frac{2\bar{V}\varphi^* \left[1 + \lambda(1+\zeta Bn^{-1}) \right]}{1+\bar{V}\varphi^*(1+\lambda)} \right]}{\chi \left[1 + \bar{V}\varphi^*(1+\lambda) \right]^2 (1-\chi)^{1/2}} \right\} \frac{\varphi^*}{\varphi} \quad \text{with } \chi = \frac{4\lambda\bar{V}\varphi^*}{\left[1 + \bar{V}\varphi^*(1+\lambda) \right]^2}$$

Table S2. Taylor series expansions for the ratio $c_M^{*,\infty} / c_M^{*,0}$ at $\varphi / \varphi^* \ll 1$ and $\varphi / \varphi^* \gg 1$ (indicated, valid with discarding term in $\bar{V}\varphi$) and for k_+ / k_- in the limits $\varphi / \varphi^* \ll 1$ and $\varphi / \varphi^* \gg 1$ (indicated). Results are valid up to first order terms in φ / φ^* or φ^* / φ .

IV. 3. Dependence of $c_M^{*,\infty} / c_M^{*,0}$ on φ / φ^* and \bar{K}^* at various values of $\Delta\bar{c}_o$ and $1/x_o$ (Figure S5).

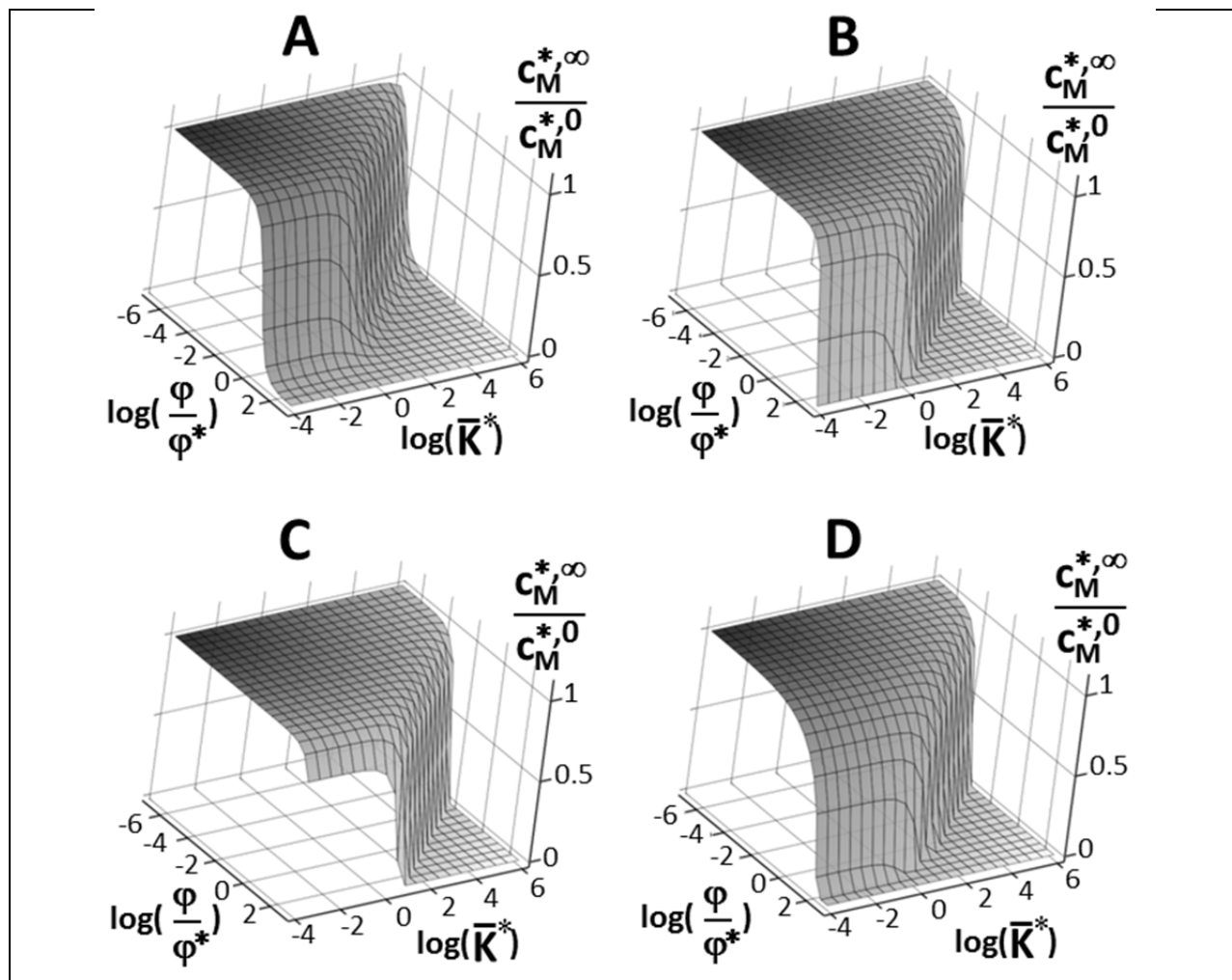


Figure S5. Evolution of the ratio $c_M^{*,\infty} / c_M^{*,0}$ with microorganism volume fraction (normalized by $\varphi^* = R_S / R_e$ with $R_e = S_a / (k_e V_p)$ and $R_S = 1 / (k_{\text{int}} K_H \beta_a)$) and with intracellular dimensionless ML_S complex stability constant (\bar{K}^*). Model parameters: $\varphi^* = 10^{-3}$, $\bar{V} = \bar{\phi}_u^{T,0} = 0$, $a / r_o = \varepsilon = \beta_a = 1$ with (A): $\Delta \bar{c}_o = 0$, $x_o = 1$, (B): $\Delta \bar{c}_o = 0$, $x_o = 5 \times 10^2$, (C): $\Delta \bar{c}_o = 0$, $x_o = 5 \times 10^3$, (D): $\Delta \bar{c}_o = 1$, $x_o = 5 \times 10^2$.

References.

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