SUPPORTING INFORMATION

Dynamics of metal uptake by charged soft biointerphases: impacts of depletion, internalisation, adsorption and excretion.

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References quoted in Supporting Information are reported at the end of this document.

SI1. Derivation of eqn (16) in the main text.

The general solution of eqn (13) may be written in the transcendental integral form

$$c_{\mathrm{M}}^{*}(t)=\left(K_{\mathrm{M}}\beta_a^{-1}Bn^{-1}-k_{\mathrm{e}}\phi_{\mathrm{u}}^{0}R_{\mathrm{T}}\right)\mathrm{e}^{-k_{\mathrm{e}}t}+K_{\mathrm{M}}\beta_a^{-1}\left\{\bar{c}_{\mathrm{M}}^{\mathrm{a}}(t)-Bn^{-1}\left[\frac{1}{1+\bar{c}_{\mathrm{M}}^{\mathrm{a}}(t)}-k_{\mathrm{e}}\mathrm{e}^{-k_{\mathrm{e}}t}\int_0^t\frac{\mathrm{e}^{-k_{\mathrm{e}}\xi}}{1+\bar{c}_{\mathrm{M}}^{\mathrm{a}}(\xi)}\mathrm{d}\xi\right]\right\}. \quad \text{(S1)}$$

Under the conditions $k_{\mathrm{e}}\to 0$ and/or $t=0$, eqn (S1) simplifies into

$$c_{\mathrm{M}}^{*}=\left(K_{\mathrm{M}}\beta_a^{-1}Bn^{-1}-k_{\mathrm{e}}\phi_{\mathrm{u}}^{0}R_{\mathrm{T}}\right)+K_{\mathrm{M}}\beta_a^{-1}\left[\bar{c}_{\mathrm{M}}^{\mathrm{a}}-\frac{Bn^{-1}}{1+\bar{c}_{\mathrm{M}}^{\mathrm{a}}}\right], \quad \text{(S2)}$$

where we have dropped the variable t for simplicity. Equation (S2) may be rewritten in the form of the second order polynomial equation in $x=c_{\mathrm{M}}^{\mathrm{a}}/\left(\beta_a c_{\mathrm{M}}^{*}\right)$ according to

$$A\left(1+B\omega^{0}\right)+x\left[1-A-B\left(1-\omega^{0}\right)\right]-x^{2}=0, \quad \text{(S3)}$$

with $\omega^{0}=k_{\mathrm{e}}\phi_{\mathrm{u}}^{0}/J_{\mathrm{u}}^{*}$, $A=K_{\mathrm{M}}/\left(\beta_a c_{\mathrm{M}}^{*}\right)$ and $B=J_{\mathrm{u}}^{*}R_{\mathrm{T}}/c_{\mathrm{M}}^{*}$. The physical solution of eqn (S3) reads as

$$x=\left\{\left[1-A-B\left(1-\omega^{0}\right)\right]+\left\{\left[A+B\left(1-\omega^{0}\right)-1\right]^{2}+4A\left[1+B\omega^{0}\right]\right\}^{1/2}\right\}/2, \quad \text{(S4)}$$

which is eqn (16) in the main text.

SI2. Derivation of eqn (19) in the main text, Demonstration of $\Omega_{1,2}\le 0$, $-\tau_{\mathrm{o}}>0$. Justification of the physical interpretation of $-\tau_{\mathrm{o}}$.

In the cases where $\Gamma^{\mathrm{S}}(t)\to 0$ and $Q^{\mathrm{V}}(t)\to 0$, eqn (18) becomes

$$\int_a^{r_{\mathrm{c}}} r^{2}\left\{\mathrm{d}c_{\mathrm{M}}(r,t)/\mathrm{d}t+k_{\mathrm{e}}\left[c_{\mathrm{M}}(r,t)-c_{\mathrm{M}}(r,0)\right]\right\}\mathrm{d}r=a^{2}\left(-J_{\mathrm{u}}(t)+k_{\mathrm{e}}\phi_{\mathrm{u}}^{0}\right). \quad \text{(S5)}$$

Substituting eqns (5) and (6) for $c_{\mathrm{M}}(a\le r\le r_{\mathrm{o}},t)$ and $c_{\mathrm{M}}(r_{\mathrm{o}}\le r\le r_{\mathrm{c}},t)$, respectively, and eqn (1) for $J_{\mathrm{u}}(t)$, performing the integration, we obtain

$$c_{\mathrm{M}}^{\mathrm{a}}(t)/\left[K_{\mathrm{M}}+c_{\mathrm{M}}^{\mathrm{a}}(t)\right]-\Omega_{1}\left[\mathrm{d}c_{\mathrm{M}}^{*}(t)/\mathrm{d}t+k_{\mathrm{e}}c_{\mathrm{M}}^{*}(t)\right]-2\Omega_{2}\beta_a^{-1}\left[\mathrm{d}c_{\mathrm{M}}^{\mathrm{a}}(t)/\mathrm{d}t+k_{\mathrm{e}}c_{\mathrm{M}}^{\mathrm{a}}(t)\right]+k_{\mathrm{e}}\tau_{\mathrm{o}}=0, \quad \text{(S6)}$$

where the time constant τ_{o} is given by eqn (24) in the main text. To derive eqn (S6), we have used the following integrals given in our previous work[1]

$$4\pi\int_{r_{\mathrm{o}}}^{r_{\mathrm{c}}} r^{2}c_{\mathrm{M}}(r,t)\mathrm{d}r=4\pi c_{\mathrm{M}}^{*}(t)\left[G_{r_{\mathrm{o}},r_{\mathrm{c}}}+\left(c_{\mathrm{M}}^{\mathrm{a}}(t)\beta_a^{-1}/c_{\mathrm{M}}^{*}(t)-1\right)\left(1+F_{a,r_{\mathrm{o}}}\lambda^{-1}\right)H_{r_{\mathrm{o}},r_{\mathrm{c}}}^{r_{\mathrm{c}}}/F_{r_{\mathrm{o}},r_{\mathrm{c}}}\right], \quad \text{(S7)}$$

$$4\pi\int_a^{r_\text{o}} r^2 c_\text{M}(r,t)\text{d}r = 4\pi c_\text{M}^*(t)\left[\lambda^{-1}H_{a,r_\text{o}}^a + \left(G_{a,r_\text{o}} - \lambda^{-1}H_{a,r_\text{o}}^a\right)c_\text{M}^\text{a}(t)\beta_\text{a}^{-1}/c_\text{M}^*(t)\right], \tag{S8}$$

with $F_{r_1,r_2} = \int_{r_1}^{r_2} r^{-2}\beta_r^{-1}\text{d}r$, $G_{r_1,r_2} = \int_{r_1}^{r_2} r^2\beta_r\text{d}r$ and $H_{r_1,r_2}^{r_3} = \int_{r_1}^{r_2} r^2\beta_r F_{r,r_3}\text{d}r$. The sum of eqns (S7)-(S8) identifies to

$$\int_a^{r_\text{c}} r^2 c_\text{M}(r,t)\text{d}r = -a^2 J_\text{u}^*\left[\Omega_1 c_\text{M}^*(t) + 2\Omega_2\beta_a^{-1}c_\text{M}^\text{a}(t)\right], \tag{S9}$$

where $\Omega_1 = -\left[G_{r_\text{o},r_\text{c}} - \left(1-\varepsilon^{-1}f_\text{el}/f_\text{el,in}\right)H_{r_\text{o},r_\text{c}}^{r_\text{c}}/F_{r_\text{o},r_\text{c}} - H_{a,r_\text{o}}^a a\varepsilon^{-1}f_\text{el}\right]/\left(a^2J_\text{u}^*\right)$ and $\Omega_2 = -\left[\Omega_1 + G_{a,r_\text{c}}/\left(a^2J_\text{u}^*\right)\right]/2$. The reasoning reported in our previous work (Supporting Information therein) led to $\Omega_1 \le 0$. While this inequality is correct, the given demonstration was partially inexact. Indeed, unlike our statement in Ref. [1], the ratio $\varepsilon^{-1}f_\text{el}/f_\text{el,in}$ satisfies $0 \le \varepsilon^{-1}f_\text{el}/f_\text{el,in} \le 1$, which is derived from eqn (7). After writing the term F_{r,r_c} involved in $H_{r_\text{o},r_\text{c}}^{r_\text{c}}$ as the sum $F_{r,r_\text{o}} + F_{r_\text{o},r_\text{c}}$, we obtain $H_{r_\text{o},r_\text{c}}^{r_\text{c}} = F_{r_\text{o},r_\text{c}}G_{r_\text{o},r_\text{c}} + \int_{r_\text{o}}^{r_\text{c}} r^2\beta_r F_{r,r_\text{o}}\text{d}r$. Realizing that $\int_{r_\text{o}}^{r_\text{c}} r^2\beta_r F_{r,r_\text{o}}\text{d}r \le 0$, $F_{r_\text{o},r_\text{c}} \ge 0$ and $G_{r_\text{o},r_\text{c}} \ge 0$, it comes $H_{r_\text{o},r_\text{c}}^{r_\text{c}}/F_{r_\text{o},r_\text{c}} \le G_{r_\text{o},r_\text{c}}$. Combining with $0 \le \varepsilon^{-1}f_\text{el}/f_\text{el,in} \le 1$, we then infer $\left(1-\varepsilon^{-1}f_\text{el}/f_\text{el,in}\right)H_{r_\text{o},r_\text{c}}^{r_\text{c}}/F_{r_\text{o},r_\text{c}} \le \left(1-\varepsilon^{-1}f_\text{el}/f_\text{el,in}\right)G_{r_\text{o},r_\text{c}} \le G_{r_\text{o},r_\text{c}}$. Since $H_{a,r_\text{o}}^a \le 0$, it comes $\Omega_1 \le 0$.

Using the definition of Ω_1, the scalar $\Omega_2 = -\left[\Omega_1 + G_{a,r_\text{c}}/\left(a^2J_\text{u}^*\right)\right]/2$ may be written according to $\Omega_2 = -\left[G_{a,r_\text{o}} + \left(1-\varepsilon^{-1}f_\text{el}/f_\text{el,in}\right)H_{r_\text{o},r_\text{c}}^{r_\text{c}}/F_{r_\text{o},r_\text{c}} + H_{a,r_\text{o}}^a a\varepsilon^{-1}f_\text{el}\right]/\left(2a^2J_\text{u}^*\right)$. Let us now show that $\Omega_2 \le 0$. We have $H_{a,r_\text{o}}^a = -F_{a,r_\text{o}}G_{a,r_\text{o}} + \int_a^{r_\text{o}} r^2\beta_r F_{r,r_\text{o}}\text{d}r$, which is obtained after writing the term $F_{r,a}$ involved in H_{a,r_o}^a according to $F_{r,r_\text{o}} + F_{r_\text{o},a}$. Because $\int_a^{r_\text{o}} r^2\beta_r F_{r,r_\text{o}}\text{d}r \ge 0$, it comes $F_{a,r_\text{o}}G_{a,r_\text{o}} + H_{a,r_\text{o}}^a \ge 0$ or $G_{a,r_\text{o}} + H_{a,r_\text{o}}^a af_\text{el,in} \ge 0$ where we have used $1/F_{a,r_\text{o}} = af_\text{el,in} \ge 0$. Recalling that $H_{a,r_\text{o}}^a \le 0$ and $0 \le \varepsilon^{-1}f_\text{el} \le f_\text{el,in}$, we thus obtain $G_{a,r_\text{o}} + H_{a,r_\text{o}}^a a\varepsilon^{-1}f_\text{el} \ge 0$. Since $\left(1-\varepsilon^{-1}f_\text{el}/f_\text{el,in}\right)H_{r_\text{o},r_\text{c}}^{r_\text{c}}/F_{r_\text{o},r_\text{c}} \ge 0$, we then conclude that $\Omega_2 \le 0$.

Substituting Ω_2 in eqn (24), we obtain for τ_o

$$\tau_\mathrm{o} = \Omega_1 \left[c_\mathrm{M}^*(0) - \beta_a^{-1} c_\mathrm{M}^\mathrm{a}(0) \right] - \left(a^2 J_\mathrm{u}^* \right)^{-1} \left[a^2 \phi_\mathrm{u}^0 + \beta_a^{-1} G_{a,r_\mathrm{c}} c_\mathrm{M}^\mathrm{a}(0) \right]. \tag{S10}$$

Using $\Omega_1 < 0$, $G_{a,r_\mathrm{c}} > 0$ and $c_\mathrm{M}^\mathrm{a}(0) \le \beta_a c_\mathrm{M}^*(0)$ (the steady-state surface metal concentration can not exceed the value predicted by equilibrium Boltzmann law), we demonstrate that $-\tau_\mathrm{o} > 0$. Combining eqn (24) with eqn (S9), one shows that $-\tau_\mathrm{o}$ satisfies the relationship $-J_\mathrm{u}^* \tau_\mathrm{o} = 4\pi \int_a^{r_\mathrm{c}} r^2 c_\mathrm{M}(r,0) \mathrm{d}r / S_\mathrm{a} + \phi_\mathrm{u}^0$, which supports the physical interpretation given in the main text for the time constant $-\tau_\mathrm{o}$.

After elimination of the quantity $\mathrm{d}c_\mathrm{M}^*(t)/\mathrm{d}t + k_\mathrm{e} c_\mathrm{M}^*(t)$ from the combination of eqns (S6) and (13), we obtain

$$\mathrm{d}c_\mathrm{M}^\mathrm{a}(t)/\mathrm{d}t = \frac{c_\mathrm{M}^\mathrm{a}(t)\left\{ \left[K_\mathrm{M} + c_\mathrm{M}^\mathrm{a}(t) \right]^{-1} - k_\mathrm{e} \beta_a^{-1} (\Omega_1 + 2\Omega_2) \right\} + k_\mathrm{e} \tau_\mathrm{o}}{J_\mathrm{u}^* \Omega_1 R_\mathrm{T} K_\mathrm{M} \left[K_\mathrm{M} + c_\mathrm{M}^\mathrm{a}(t) \right]^{-2} + \beta_a^{-1} (\Omega_1 + 2\Omega_2)}, \tag{S11}$$

with $R_\mathrm{T} = 1/\left(D_\mathrm{M,out} f_\mathrm{el} a^{-1} \right)$ the microorganism surface resistance. Introducing $\bar{c}_\mathrm{M}^\mathrm{a}(t) = c_\mathrm{M}^\mathrm{a}(t)/K_\mathrm{M}$ and using the relationships $\tau_\mathrm{L} = 4\pi R_\mathrm{S} G_{a,r_\mathrm{c}} / S_\mathrm{a} = -K_\mathrm{M} \beta_a^{-1} (\Omega_1 + 2\Omega_2)$ and $\tau_\mathrm{E} = \tau_\mathrm{L} - J_\mathrm{u}^* \Omega_1 R_\mathrm{T}$,[1] eqn (S11) may be rewritten in the concise form

$$\mathrm{d}c_\mathrm{M}^\mathrm{a}(t)/\mathrm{d}t = \frac{c_\mathrm{M}^\mathrm{a}(t)\left\{ \left[1 + \bar{c}_\mathrm{M}^\mathrm{a}(t) \right]^{-1} + k_\mathrm{e} \tau_\mathrm{L} \right\} + k_\mathrm{e} K_\mathrm{M} \tau_\mathrm{o}}{(\tau_\mathrm{L} - \tau_\mathrm{E}) \left[1 + \bar{c}_\mathrm{M}^\mathrm{a}(t) \right]^{-2} - \tau_\mathrm{L}}, \text{ or, equivalently,} \tag{S12}$$

$$\left\{ \frac{(\tau_\mathrm{L} - \tau_\mathrm{E}) - \tau_\mathrm{L} \left[1 + \bar{c}_\mathrm{M}^\mathrm{a}(t)^2 \right]}{\left[1 + \bar{c}_\mathrm{M}^\mathrm{a}(t) \right] P\left(\bar{c}_\mathrm{M}^\mathrm{a}(t) \right)} \right\} \mathrm{d}\bar{c}_\mathrm{M}^\mathrm{a}(t) = k_\mathrm{e} \tau_\mathrm{L} \mathrm{d}t, \tag{S13}$$

where we have defined the second order polynomial $P(x)$

$$P(x) = (\tau_\mathrm{o}/\tau_\mathrm{L}) + \left\{ \left[1 + k_\mathrm{e} (\tau_\mathrm{L} + \tau_\mathrm{o}) \right] / (k_\mathrm{e} \tau_\mathrm{L}) \right\} x + x^2. \tag{S14}$$

The discriminant Δ of $P(x)$ is given by

$$\Delta = \left\{ \left[1 + k_\mathrm{e} (\tau_\mathrm{L} + \tau_\mathrm{o}) \right] / (k_\mathrm{e} \tau_\mathrm{L}) \right\}^2 - 4\tau_\mathrm{o}/\tau_\mathrm{L}. \tag{S15}$$

It is straightforward to verify that $\Delta > 0$ since $-\tau_\mathrm{o} > 0$. The roots $\bar{c}_\pm$ of $P(x)$ are

$$\overline{c}_{+} = \left\{-\left[1+k_{\mathrm{e}}\left(\tau_{\mathrm{L}}+\tau_{\mathrm{o}}\right)/\left(k_{\mathrm{e}}\tau_{\mathrm{L}}\right)\right]+\Delta^{1/2}\right\}/2 \tag{S16}$$

and
$$\overline{c}_{-} = \left\{-\left[1+k_{\mathrm{e}}\left(\tau_{\mathrm{L}}+\tau_{\mathrm{o}}\right)/\left(k_{\mathrm{e}}\tau_{\mathrm{L}}\right)\right]-\Delta^{1/2}\right\}/2\,, \tag{S17}$$

with $\overline{c}_{+} > 0$ and $\overline{c}_{-} < 0$. $\overline{c}_{\pm}$ can be rewritten in the form

$$\overline{c}_{\pm} = \left(2k_{\mathrm{e}}\tau_{\mathrm{L}}\right)^{-1}\left\{-\left[1+k_{\mathrm{e}}\left(\tau_{\mathrm{L}}+\tau_{\mathrm{o}}\right)\right]\pm\left[1+2k_{\mathrm{e}}\left(\tau_{\mathrm{L}}+\tau_{\mathrm{o}}\right)+k_{\mathrm{e}}^{2}\left(\tau_{\mathrm{L}}-\tau_{\mathrm{o}}\right)^{2}\right]^{1/2}\right\}, \tag{S18}$$

which is eqn (23) in the main text. Finally, the differential equation governing the time dependence of $c_{\mathrm{M}}^{\mathrm{a}}(t)$ is given by

$$\left\{\frac{\left(\tau_{\mathrm{L}}-\tau_{\mathrm{E}}\right)-\tau_{\mathrm{L}}\left[1+\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t)^{2}\right]}{\left[1+\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t)\right]\left[\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t)-\overline{c}_{+}\right]\left[\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t)-\overline{c}_{-}\right]}\right\}\mathrm{d}\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t) = k_{\mathrm{e}}\tau_{\mathrm{L}}\mathrm{d}t\,. \tag{S19}$$

Equation (S19) identifies with eqn (19).

SI3. Derivation of the expressions given in Table 1, case $\boxed{\mathrm{B}}$ ($K_{\mathrm{M}} << c_{\mathrm{M}}^{\mathrm{a}}(t)$).

This case corresponds to a strong affinity of M for the internalisation sites, which simplifies eqn (1) into $J_{\mathrm{u}}(t) \approx J_{\mathrm{u}}^{*}$. Substitution into eqn (12) provides $\mathrm{d}J_{\mathrm{M}}(t)/\mathrm{d}t = -k_{\mathrm{e}}J_{\mathrm{M}}(t)$ where $J_{\mathrm{M}}(t)$ depends on $c_{\mathrm{M}}^{\mathrm{a}}(t)$ and $c_{\mathrm{M}}^{*}(t)$ according to eqn (8). In turn, we obtain

$$\mathrm{d}c_{\mathrm{M}}^{*}(t)/\mathrm{d}t + k_{\mathrm{e}}c_{\mathrm{M}}^{*}(t) = \beta_{a}^{-1}\left[\mathrm{d}c_{\mathrm{M}}^{\mathrm{a}}(t)/\mathrm{d}t + k_{\mathrm{e}}c_{\mathrm{M}}^{\mathrm{a}}(t)\right], \tag{S20}$$

which implies that the equilibrium Boltzmann law applies for the metal concentration profile in the extracellular volume, *i.e.*

$$c_{\mathrm{M}}^{*}(t) = \beta_{a}^{-1}c_{\mathrm{M}}^{\mathrm{a}}(t)\,. \tag{S21}$$

Substituting eqn (S21) into eqn (S6), it comes

$$\mathrm{d}c_{\mathrm{M}}^{*}(t)/\mathrm{d}t + k_{\mathrm{e}}c_{\mathrm{M}}^{*}(t) = -K_{\mathrm{M}}\beta_{a}^{-1}\left(1+k_{\mathrm{e}}\tau_{\mathrm{o}}\right)/\tau_{\mathrm{L}}\,, \tag{S22}$$

where we have used $\tau_{\mathrm{L}} = 4\pi R_{\mathrm{S}}G_{a,r_{\mathrm{c}}}/S_{\mathrm{a}} = -K_{\mathrm{M}}\beta_{a}^{-1}\left(\Omega_{1}+2\Omega_{2}\right)$.[1] Solving eqn (S22) provides the expression of $c_{\mathrm{M}}^{*}(t)$ given in Table 1, case $\boxed{\mathrm{B}}$. The surface metal concentration $c_{\mathrm{M}}^{\mathrm{a}}(t)$ is then simply obtained from eqn (S21).

SI4. Derivation of the expressions given in Table 1, case $\boxed{\mathrm{C}}$ ($K_{\mathrm{M}} >> c_{\mathrm{M}}^{\mathrm{a}}(t)$) and of eqn (31). Demonstration of $\alpha_{1} = 0$ for $\phi_{\mathrm{u}}^{0} = 0$.

In the situation $K_{\mathrm{M}} >> c_{\mathrm{M}}^{\mathrm{a}}(t)$, we have $\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t) << 1$. Retaining the first order term in the Taylor expansion of eqn (13) with respect to $\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t)$, we obtain

$$\mathrm{d}c_{\mathrm{M}}^{*}(t)/\mathrm{d}t = -k_{\mathrm{e}}c_{\mathrm{M}}^{*}(t) + K_{\mathrm{M}}\beta_{\mathrm{a}}^{-1}\left[k_{\mathrm{e}}\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t) + \left(1+Bn^{-1}\right)\mathrm{d}\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t)/\mathrm{d}t\right]. \tag{S23}$$

The solution of eqn (S23) in $c_{\mathrm{M}}^{*}(t)$ is given by

$$c_{\mathrm{M}}^{*}(t) = \alpha_1 \mathrm{e}^{-k_{\mathrm{e}}t} + K_{\mathrm{M}}\beta_{\mathrm{a}}^{-1}\left\{\left(1+Bn^{-1}\right)\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t) - k_{\mathrm{e}}Bn^{-1}\mathrm{e}^{-k_{\mathrm{e}}t}\int_{0}^{t}\overline{c}_{\mathrm{M}}^{\mathrm{a}}(\xi)\mathrm{e}^{-k_{\mathrm{e}}\xi}\mathrm{d}\xi\right\}, \tag{S24}$$

where α_1 is a constant to be determined from the initial boundary condition $c_{\mathrm{M}}^{*}(t=0) = c_{\mathrm{M}}^{*}(0)$. In addition, eqn (S19), or, equivalently, eqn (S12), becomes to the first order in $\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t)$

$$\mathrm{d}c_{\mathrm{M}}^{\mathrm{a}}(t)/\mathrm{d}t + \left(\frac{1+k_{\mathrm{e}}\tau_{\mathrm{L}}}{\tau_{\mathrm{E}}}\right)c_{\mathrm{M}}^{\mathrm{a}}(t) = -K_{\mathrm{M}}k_{\mathrm{e}}\tau_{\mathrm{o}}/\tau_{\mathrm{E}}\,. \tag{S25}$$

This differential equation admits the general solution

$$\overline{c}_{\mathrm{M}}^{\mathrm{a}}(t) = \left(\overline{c}_{\mathrm{M}}^{\mathrm{a}}(0) + \frac{k_{\mathrm{e}}\tau_{\mathrm{o}}}{1+k_{\mathrm{e}}\tau_{\mathrm{L}}}\right)\mathrm{e}^{-t/\tau_{\mathrm{d}}} - \frac{k_{\mathrm{e}}\tau_{\mathrm{o}}}{1+k_{\mathrm{e}}\tau_{\mathrm{L}}}, \tag{S26}$$

where we have introduced the characteristic timescale of metal depletion $\tau_{\mathrm{d}} = \tau_{\mathrm{E}}/\left(1+k_{\mathrm{e}}\tau_{\mathrm{L}}\right)$. Substituting eqn (S26) into eqn (S24) and performing the integration, we obtain after rearrangements

$$c_{\mathrm{M}}^{*}(t) = \alpha_1\mathrm{e}^{-k_{\mathrm{e}}t} + \frac{K_{\mathrm{M}}}{\beta_a}\mathrm{e}^{-t/\tau_{\mathrm{d}}}\left(\overline{c}_{\mathrm{M}}^{\mathrm{a}}(0) + \frac{k_{\mathrm{e}}\tau_{\mathrm{o}}}{1+k_{\mathrm{e}}\tau_{\mathrm{L}}}\right)\left[1 + \frac{Bn^{-1}\left(1+k_{\mathrm{e}}\tau_{\mathrm{L}}\right)}{1+k_{\mathrm{e}}\left(\tau_{\mathrm{L}}-\tau_{\mathrm{E}}\right)}\right] - \frac{K_{\mathrm{M}}}{\beta_a}\frac{k_{\mathrm{e}}\tau_{\mathrm{o}}}{1+k_{\mathrm{e}}\tau_{\mathrm{L}}}. \tag{S27}$$

The constant α_1 is defined by the expression

$$\alpha_1 = -k_{\mathrm{e}}\phi_{\mathrm{u}}^{\mathrm{o}}R_{\mathrm{T}} - \frac{K_{\mathrm{M}}}{\beta_a Bn}\frac{k_{\mathrm{e}}\left[\tau_{\mathrm{o}} + \overline{c}_{\mathrm{M}}^{\mathrm{a}}(0)\tau_{\mathrm{E}}\right]}{\left[1+k_{\mathrm{e}}\left(\tau_{\mathrm{L}}-\tau_{\mathrm{E}}\right)\right]}, \tag{S28}$$

where we have used eqn (16) in the limit $K_{\mathrm{M}} >> c_{\mathrm{M}}^{\mathrm{a}}(t)$, which provides

$$c_{\mathrm{M}}^{\mathrm{a}}(0) = \beta_a\left[c_{\mathrm{M}}^{*}(0) + k_{\mathrm{e}}\phi_{\mathrm{u}}^{\mathrm{o}}R_{\mathrm{T}}\right]\left(1+Bn^{-1}\right)^{-1}. \tag{S29}$$

Equations (S26)-(S29) match the results given in Table 1, case $\boxed{\mathrm{C}}$. Under the condition where $K_{\mathrm{M}} >> c_{\mathrm{M}}^{\mathrm{a}}(t)$, the time constant τ_{o} may be written in the form $\tau_{\mathrm{o}} = -\overline{c}_{\mathrm{M}}^{\mathrm{a}}(0)\tau_{\mathrm{E}} - \phi_{\mathrm{u}}^{\mathrm{o}}\left(1/J_{\mathrm{u}}^{*} + k_{\mathrm{e}}\Omega_1 R_{\mathrm{T}}\right)$, which is derived after combining eqn (24), eqn (S29) and the expression $\tau_{\mathrm{E}} = -K_{\mathrm{M}}\beta_a^{-1}\left[\Omega_1\left(1+Bn^{-1}\right) + 2\Omega_2\right]$ given in the Supporting Information of our previous work.[1] In turn, for $\phi_{\mathrm{u}}^{0}=0$, we have $\tau_{\mathrm{o}}=-\bar{c}_{\mathrm{M}}^{\mathrm{a}}(0)\tau_{\mathrm{E}}$ and $\alpha_1=0$. This justifies the existence of the only characteristic timescale $\tau_{\mathrm{d}}=\tau_{\mathrm{E}}/(1+k_{\mathrm{e}}\tau_{\mathrm{L}})$ for bulk metal depletion kinetics when $\phi_{\mathrm{u}}^{0}=0$. Considering the limits $R_{\mathrm{T}}\rightarrow 0$ and $Bn^{-1}=R_{\mathrm{T}}/R_{\mathrm{S}}<<1$, eqn (S29) reduces to $c_{\mathrm{M}}^{\mathrm{a}}(0)=\beta_a c_{\mathrm{M}}^{*}(0)$. Using the latter equation and simplifying eqns (S26)-(S28) for $Bn^{-1}=R_{\mathrm{T}}/R_{\mathrm{S}}\rightarrow 0$, we obtain eqn (31) in the main text.

SI5. Derivation of the differential equation that determines $c_{\mathrm{M}}^{\mathrm{a}}(t)$ for cases where 2D and 3D adsorptions of metal ions are relevant.

In the most complex scenario where 2D and 3D adsorption processes take place, the conservation condition for metal ions within the Kuwabara unit cell of radius r_{c} is given by eqn (18)

$$\int_a^{r_{\mathrm{c}}} r^2\left\{\mathrm{d}c_{\mathrm{M}}(r,t)/\mathrm{d}t+k_{\mathrm{e}}\left[c_{\mathrm{M}}(r,t)-c_{\mathrm{M}}(r,0)\right]\right\}\mathrm{d}r=$$

$$a^2\left\{-J_{\mathrm{u}}(t)+k_{\mathrm{e}}\phi_{\mathrm{u}}^{0}-\left[k_{\mathrm{e}}\Gamma^{\mathrm{S}}(t)+\mathrm{d}\Gamma^{\mathrm{S}}(t)/\mathrm{d}t\right]\right\}-(4\pi)^{-1}\left[k_{\mathrm{e}}Q^{\mathrm{V}}(t)+\mathrm{d}Q^{\mathrm{V}}(t)/\mathrm{d}t\right]. \quad \text{(S30)}$$

Using eqns (S5)-(S6), one shows that eqn (S30) is equivalent to

$$c_{\mathrm{M}}^{\mathrm{a}}(t)/\left[K_{\mathrm{M}}+c_{\mathrm{M}}^{\mathrm{a}}(t)\right]-\Omega_1\left[\mathrm{d}c_{\mathrm{M}}^{*}(t)/\mathrm{d}t+k_{\mathrm{e}}c_{\mathrm{M}}^{*}(t)\right]-2\Omega_2\beta_a^{-1}\left[\mathrm{d}c_{\mathrm{M}}^{\mathrm{a}}(t)/\mathrm{d}t+k_{\mathrm{e}}c_{\mathrm{M}}^{\mathrm{a}}(t)\right]+k_{\mathrm{e}}\tau_{\mathrm{o}}+$$

$$J_{\mathrm{u}}^{*-1}\left[k_{\mathrm{e}}\Gamma^{\mathrm{S}}(t)+\mathrm{d}\Gamma^{\mathrm{S}}(t)/\mathrm{d}t\right]+\left(4\pi a^2 J_{\mathrm{u}}^{*}\right)^{-1}\left[k_{\mathrm{e}}Q^{\mathrm{V}}(t)+\mathrm{d}Q^{\mathrm{V}}(t)/\mathrm{d}t\right]=0. \quad \text{(S31)}$$

From eqn (2), we further have

$$\mathrm{d}\Gamma^{\mathrm{S}}(t)/\mathrm{d}t=K_{\mathrm{M}}^{\mathrm{S}}\Gamma_{\mathrm{max}}^{\mathrm{S}}\left[K_{\mathrm{M}}^{\mathrm{S}}+c_{\mathrm{M}}^{\mathrm{a}}(t)\right]^{-2}\mathrm{d}c_{\mathrm{M}}^{\mathrm{a}}(t)/\mathrm{d}t, \quad \text{(S32)}$$

and from eqns (3) and (5), we derive

$$\begin{cases}\mathrm{d}Q^{\mathrm{V}}(t)/\mathrm{d}t=\tilde{q}_{\mathrm{a}}(t)\mathrm{d}c_{\mathrm{M}}^{\mathrm{a}}(t)/\mathrm{d}t+\tilde{q}^{*}(t)\mathrm{d}c_{\mathrm{M}}^{*}(t)/\mathrm{d}t\\ Q^{\mathrm{V}}(t)=q_{\mathrm{a}}(t)c_{\mathrm{M}}^{\mathrm{a}}(t)+q^{*}(t)c_{\mathrm{M}}^{*}(t)\end{cases}, \quad \text{(S33, S34)}$$

where we have defined $\tilde{q}_{\mathrm{a}}$, $\tilde{q}^{*}$, q_{a} and q^{*} as follows

$$\begin{cases}\tilde{q}_{\mathrm{a}}(t)=4\pi K_{\mathrm{M}}^{\mathrm{V}}\rho_{\mathrm{max}}^{\mathrm{V}}\int_a^{r_{\mathrm{o}}}\left\{\omega_r^{a}/\left[K_{\mathrm{M}}^{\mathrm{V}}+c_{\mathrm{M}}(r,t)\right]^2\right\}r^2\mathrm{d}r\\ \tilde{q}^{*}(t)=4\pi K_{\mathrm{M}}^{\mathrm{V}}\rho_{\mathrm{max}}^{\mathrm{V}}\int_a^{r_{\mathrm{o}}}\left\{\omega_r^{*}/\left[K_{\mathrm{M}}^{\mathrm{V}}+c_{\mathrm{M}}(r,t)\right]^2\right\}r^2\mathrm{d}r\end{cases} \quad \text{(S35, S36)}$$

and

$$\begin{cases} q_{\mathrm{a}}(t) = 4\pi\rho_{\max}^{\mathrm{V}} \int_a^{r_{\mathrm{o}}} \left\{ \omega_r^a / \left[K_{\mathrm{M}}^{\mathrm{V}} + c_{\mathrm{M}}(r,t) \right] \right\} r^2 \mathrm{d}r \\ q^*(t) = 4\pi\rho_{\max}^{\mathrm{V}} \int_a^{r_{\mathrm{o}}} \left\{ \omega_r^* / \left[K_{\mathrm{M}}^{\mathrm{V}} + c_{\mathrm{M}}(r,t) \right] \right\} r^2 \mathrm{d}r \end{cases}, \tag{S37, S38}$$

with ω_r^a and ω_r^* two functions of the radial coordinate r given by

$$\omega_r^a = \beta_r \beta_a^{-1} \left(1 + \lambda^{-1} F_{a,r} \right) \tag{S39}$$

and

$$\omega_r^* = -\beta_r \lambda^{-1} F_{a,r}, \tag{S40}$$

respectively, with $\lambda = -\left(\varepsilon F_{r_{\mathrm{o}},r_{\mathrm{c}}} + F_{a,r_{\mathrm{o}}} \right) = -\varepsilon a^{-1} / f_{\mathrm{el}}$. Substituting eqns (S32)-(S34) in eqn (S31) and eliminating the quantity $\mathrm{d}c_{\mathrm{M}}^*(t)/\mathrm{d}t + k_{\mathrm{e}} c_{\mathrm{M}}^*(t)$ with use of eqn (13), we obtain after arrangements the following differential equation

$$\mathrm{d}\bar{c}_{\mathrm{M}}^{\mathrm{a}}(t)/\mathrm{d}t \left\{ \left[1 + \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t) \right]^2 \tau_{\mathrm{L}} - (\tau_{\mathrm{L}} - \tau_{\mathrm{E}}) + \tau_{\mathrm{S}} \left[\frac{1 + \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t)}{1 + \theta \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t)} \right]^2 + \frac{K_{\mathrm{M}}}{S_{\mathrm{a}} J_{\mathrm{u}}^*} \left[\tilde{q}^* \beta_a^{-1} B n^{-1} + \left(\tilde{q}_{\mathrm{a}} + \beta_a^{-1} \tilde{q}^* \right) \left[1 + \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t) \right]^2 \right] \right\} +$$

$$\bar{c}_{\mathrm{M}}^{\mathrm{a}}(t) \left[1 + \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t) \right] \left\{ 1 + \left[1 + \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t) \right] k_{\mathrm{e}} \tau_{\mathrm{L}} + k_{\mathrm{e}} \tau_{\mathrm{S}} \left[\frac{1 + \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t)}{1 + \theta \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t)} \right] + \frac{k_{\mathrm{e}} K_{\mathrm{M}}}{S_{\mathrm{a}} J_{\mathrm{u}}^*} \left(q_{\mathrm{a}} + \beta_a^{-1} \tilde{q}^* \right) \left[1 + \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t) \right] \right\} +$$

$$k_{\mathrm{e}} \tau_{\mathrm{o}} \left[1 + \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t) \right]^2 + k_{\mathrm{e}} \frac{q^* - \tilde{q}^*}{S_{\mathrm{a}} J_{\mathrm{u}}^*} \left[1 + \bar{c}_{\mathrm{M}}^{\mathrm{a}}(t) \right]^2 c_{\mathrm{M}}^*(t) = 0 \tag{S41}$$

where we have introduced $\theta = K_{\mathrm{M}} / K_{\mathrm{M}}^{\mathrm{S}}$ and $\tau_{\mathrm{S}} = \theta \Gamma_{\max}^{\mathrm{S}} / J_{\mathrm{u}}^* = k_{\mathrm{int}}^{-1} K_{\mathrm{H}}^{\mathrm{S}} / K_{\mathrm{H}}$. In the limit where 2D adsorption is most predominant, it comes $\tilde{q}_{\mathrm{a}} = \tilde{q}^* = q_{\mathrm{a}} = q^* = 0$, and eqn (S41) then reduces to eqn (35) in the main text.

SI6. Derivation of eqn (36)-(40) in the main text.

In the situations where the inequalities $K_{\mathrm{M}}, K_{\mathrm{M}}^{\mathrm{S}} << c_{\mathrm{M}}^{\mathrm{a}}(t)$ and $K_{\mathrm{M}}^{\mathrm{V}} << c_{\mathrm{M}}(r,t)$ all apply, we have $c_{\mathrm{M}}^*(t) = \beta_{\mathrm{a}}^{-1} c_{\mathrm{M}}^{\mathrm{a}}(t)$ from eqn (12) (see eqns (S20)-(S21)) and eqn (S31) further simplifies into

$$\mathrm{d}c_{\mathrm{M}}^*(t)/\mathrm{d}t + k_{\mathrm{e}} c_{\mathrm{M}}^*(t) = -K_{\mathrm{M}} \beta_a^{-1} \left\{ 1 + k_{\mathrm{e}} \left[\tau_{\mathrm{o}} + \left(\Gamma_{\max}^{\mathrm{S}} + V_{\mathrm{soft}} \rho_{\max}^{\mathrm{V}} / S_{\mathrm{a}} \right) / J_{\mathrm{u}}^* \right] \right\} / \tau_{\mathrm{L}}. \tag{S42}$$

The solution of this equation is identical to that of eqn (S22) provided that τ_{o} is replaced by the timescale $\tilde{\tau}_{\mathrm{o}}$ defined by

$$\tilde{\tau}_{\mathrm{o}} = \tau_{\mathrm{o}} + \left(\Gamma_{\max}^{\mathrm{S}} + V_{\mathrm{soft}} \rho_{\max}^{\mathrm{V}} / S_{\mathrm{a}} \right) / J_{\mathrm{u}}^*, \tag{S43}$$

which is eqn (36) in the main text.

In the other extremes where $K_{\mathrm{M}}, K_{\mathrm{M}}^{\mathrm{S}} >> c_{\mathrm{M}}^{\mathrm{a}}(t)$ and $K_{\mathrm{M}}^{\mathrm{V}} >> c_{\mathrm{M}}(r,t)$, eqns (1), (2) and (3) may be linearized with respect to $c_{\mathrm{M}}^{\mathrm{a}}(t)/K_{\mathrm{M}}$, $c_{\mathrm{M}}^{\mathrm{a}}(t)/K_{\mathrm{M}}^{\mathrm{S}}$ and $c_{\mathrm{M}}(r,t)/K_{\mathrm{M}}^{\mathrm{V}}$. After elimination of $\mathrm{d}c_{\mathrm{M}}^{*}(t)/\mathrm{d}t + k_{\mathrm{e}}c_{\mathrm{M}}^{*}(t)$ with use of eqn (13) and substitution in eqn (S31), the differential equation governing the time dependence of the metal surface concentration now reads as

$$\mathrm{d}c_{\mathrm{M}}^{\mathrm{a}}(t)/\mathrm{d}t + \left(\frac{1+k_{\mathrm{e}}\tilde{\tau}_{\mathrm{L}}}{\tilde{\tau}_{\mathrm{E}}}\right)c_{\mathrm{M}}^{\mathrm{a}}(t) = -K_{\mathrm{M}}k_{\mathrm{e}}\tau_{\mathrm{o}}/\tilde{\tau}_{\mathrm{E}}, \tag{S44}$$

where $\tilde{\tau}_{\mathrm{E}}$ and $\tilde{\tau}_{\mathrm{L}}$ are defined by eqns (37)-(40) in the main text. The solution $c_{\mathrm{M}}^{\mathrm{a}}(t)$ of eqn (S44) is similar to that of eqn (S25) with replacing τ_{L} and τ_{E} by $\tilde{\tau}_{\mathrm{L}}$ and $\tilde{\tau}_{\mathrm{E}}$, respectively. The quantity x_{a} expressed by eqn (39) and involved in the definition of $\tilde{\tau}_{\mathrm{L}}$ necessarily satisfies $x_{\mathrm{a}} \geq 1$ because $\Omega_2 \leq 0$, $G_{a,r_{\mathrm{o}}} \geq 0$ and $G_{a,r_{\mathrm{o}}} + H_{a,r_{\mathrm{o}}}^{a} a\varepsilon^{-1} f_{\mathrm{el}} \geq 0$, as shown in §SI2. In addition, the inequalities $H_{a,r_{\mathrm{o}}}^{a} \leq 0$ and $\Omega_1 \leq 0$ (§SI2) lead to $x^{*} \geq 1$.

7. Figures S1 and S2.

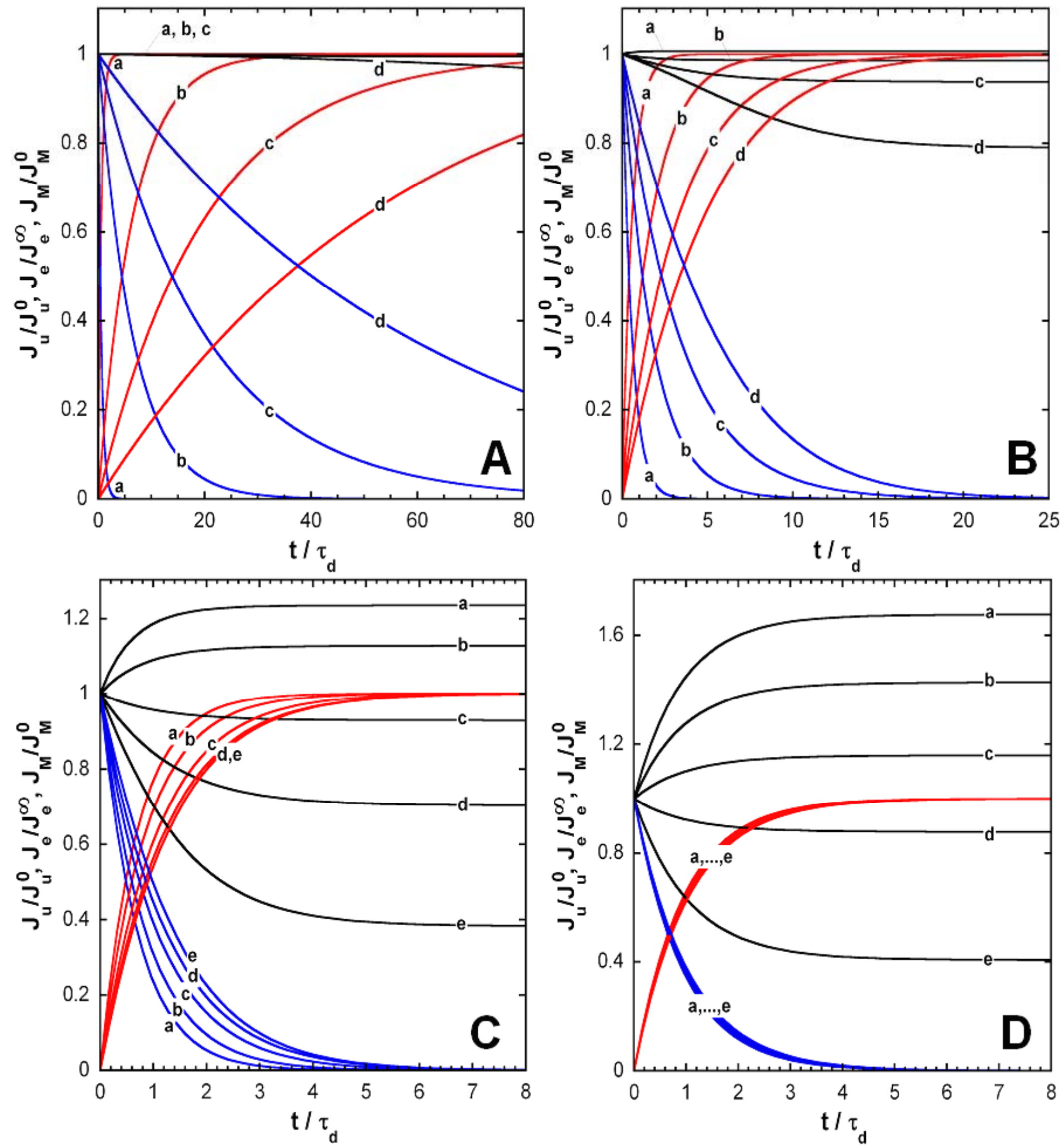

Figure S1. Characteristic dependence of the normalized fluxes $J_{\mathrm{u}}/J_{\mathrm{u}}^{0}$ (black), $J_{\mathrm{e}}/J_{\mathrm{e}}^{\infty}$ (red), $J_{\mathrm{M}}/J_{\mathrm{M}}^{0}$ (blue) on the dimensionless time t/τ_{d} for various values of k_{e} ($J_{\mathrm{u}}^{0} \equiv J_{\mathrm{u}}(t=0)$, $J_{\mathrm{e}}^{\infty} \equiv J_{\mathrm{e}}(t \to \infty)$). (A) $K_{\mathrm{M}} = 10^{-7}$ mM, (B) $K_{\mathrm{M}} = 10^{-6}$ mM, (C) $K_{\mathrm{M}} = 10^{-5}$ mM and (D) $K_{\mathrm{M}} = 10^{-4}$ mM. The letters (a), (b), (c), (d) and (e) in panels (A), (B), (C) and (D) correspond to the values of $k_{\mathrm{e}}\tau_{\mathrm{L}}$ in decreasing order as adopted in Figure 2A,B,C and D, respectively. The other model parameters are identical to those adopted in Figure 2 of the main text.

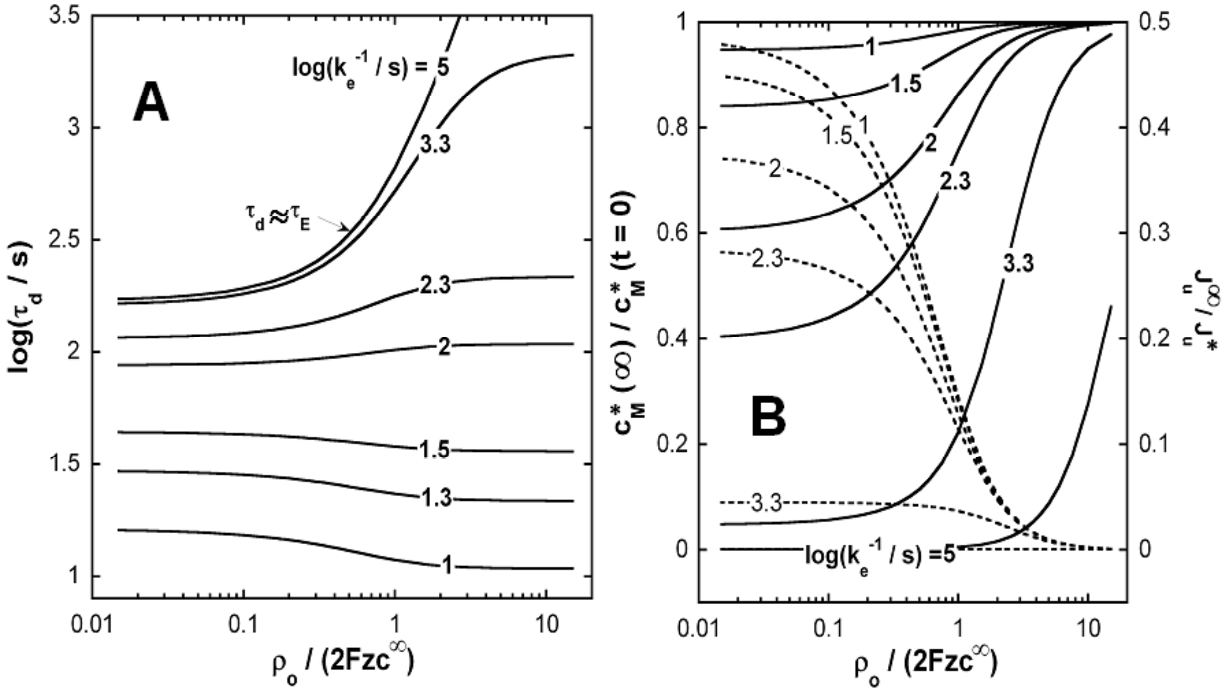

Figure S2. (A) Dependence of the characteristic depletion timescale τ_d on the dimensionless charge density $\rho_o/\left(2zFc^{\infty}\right)$ in the soft surface layer for various values of $1/k_e$ (indicated). Model parameters : $K_M = 10^{-5}$ mM, and $\rho_o/F = +30$ mM. Other parameters: $a = 400$ nm, $d = 50$ nm, $\varphi = 10^{-6}$, $k_{int}K_H = 2\times10^{-3}$ ms^{-1}, $D_{M,out} = 10^{-9}$ m^{2}s^{-1}, $\varepsilon = 1$, $\phi_u^0 = 0$, $c_M^*\left(t=0\right) = 10^{-5}$ mM, $z = 1$, $z_M = 2$, $\Gamma_{max}^s = \rho_{max}^V = 0$ (metal adsorption on the biomembrane and in the soft surface layer is ignored in eqn (18)), $\Im = \varepsilon_s/\varepsilon_r = 1$, $y_a = \bar{\sigma}_a = 0$ (the membrane surface is uncharged). (B) Dependence of the ratios $c_M^*\left(t\to\infty\right)/c_M^*\left(t=0\right)$ (plain lines) and J_u^{∞}/J_u^* (dotted lines) under the conditions of panel (A).

References.

1. J. F. L. Duval, *Phys. Chem. Chem. Phys.*, 2013, **15**, 7873-7888.