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Surface fouling as a mechanism for chemotaxis in isotropic catalytic swimmers

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Supporting information

1 Linear fuel gradient

To model a fuel gradient linearly increasing in the $\hat{z}$ direction, we substitute $C_F = D_F r_0 \cos\theta + C_{F,0}$ into equations 11 and 12 in the main paper. Breaking $\hat{r}$ into its Cartesian components, but retaining the spherical angles as integration variables for the sake of symmetry, we get:

$$\begin{aligned}\vec{v}_{tot} = -ak_P K_F r_0^2 \Bigg[&\hat{x} \int_0^{2\pi} \int_0^{\pi} \frac{\sin^2\theta \cos\varphi (D_F r_0 \cos\theta + C_{F,0}) d\theta d\varphi}{1 + K_F (D_F r_0 \cos\theta + C_{F,0})} \\ &+ \hat{y} \int_0^{2\pi} \int_0^{\pi} \frac{\sin^2\theta \sin\varphi (D_F r_0 \cos\theta + C_{F,0}) d\theta d\varphi}{1 + K_F (D_F r_0 \cos\theta + C_{F,0})} \\ &+ \hat{z} \int_0^{2\pi} \int_0^{\pi} \frac{\sin\theta \cos\theta (D_F r_0 \cos\theta + C_{F,0}) d\theta d\varphi}{1 + K_F (D_F r_0 \cos\theta + C_{F,0})} \Bigg]\end{aligned} \tag{1}$$

For the $\hat{x}$ and $\hat{y}$ components, the only azimuthally dependent terms are $\cos\varphi d\varphi$ and $\sin\varphi d\varphi$, both of which disappear when integrated from 0 to 2π. Thus, the total velocity is directed solely along the $\hat{z}$ direction, as expected. Performing the integration gives us the result:

$$\begin{aligned}\vec{v}_{tot} = \hat{z} \frac{2\pi a k_P}{D_F^2 K_F^2} \Bigg[&(C_{F,0} K_F + 1) \ln\left(\frac{C_{F,0} K_F + 1 - D_F K_F r_0}{C_{F,0} K_F + 1 + D_F K_F r_0} \right) \\ &+ 2 D_F K_F r_0 \Bigg]\end{aligned} \tag{2}$$

We first notice that the argument of the logarithm is negative semidefinite where $C_{F,0}K_F + 1 \leq D_F K_F r_0$. There is a singularity where the equality holds. This is because at some point, as the radius r_0 of the particle increases, a portion of the particle's surface will be in a negative concentration region. This occurs because the concentration gradient is linear and therefore the range of concentration is not bounded by zero. Since a negative concentration is unphysical, we ignore situations where $C_{F,0}K_F + 1 \leq D_F K_F r_0$. We next show that

$$(C_{F,0}K_F + 1)\ln\left(\frac{C_{F,0}K_F + 1 - D_F K_F r_0}{C_{F,0}K_F + 1 + D_F K_F r_0}\right) + 2D_F K_F r_0 < 0 \tag{3}$$

for all $C_{F,0}K_F + 1 > D_F K_F r_0$. To do this, we note that $C_{F,0}$, K_F, D_F, and r_0 are all positive constants, so we can write the relation:

$$C_{F,0}K_F + 1 = s(2D_F K_F r_0) \tag{4}$$

where $s > 1/2$. This allows us to rewrite the condition in equation 3 above as

$$\ln\left(\frac{2s+1}{2s-1}\right) > \frac{1}{s} \tag{5}$$

for all $s > 1/2$. We can see that this condition is always true by noting that the logarithm can be rewritten as an inverse hyperbolic cotangent and expanded in a series:

$$\ln\left(\frac{2s+1}{2s-1}\right) = 2\coth^{-1}(2s) = \frac{1}{s} + \frac{2}{3(2s)^3} + \frac{2}{5(2s)^5} + \cdots > \frac{1}{s} \tag{6}$$

for all $s > 1/2$. Thus, the condition in equation 3 above is fulfilled and velocity is constant and negative along the $\hat{z}$ direction.

2 Linear foulant gradient

Letting $C_T = D_T r_0 \cos\theta + C_{T,0}$ and performing the integration in equation 12 in the main paper:

$$\begin{aligned}\vec{v}_{tot} = -ak_P K_F C_F r_0^2 \Big[&\hat{x}\int_0^{2\pi}\int_0^{\pi} \frac{\sin^2\theta\cos\varphi d\theta d\varphi}{1 + K_F C_F + K_T(D_T r_0 \cos\theta + C_{T0})} \\ &+\hat{y}\int_0^{2\pi}\int_0^{\pi} \frac{\sin^2\theta\sin\varphi d\theta d\varphi}{1 + K_F C_F + K_T(D_T r_0 \cos\theta + C_{T0})} \\ &+\hat{z}\int_0^{2\pi}\int_0^{\pi} \frac{\sin\theta\cos\theta d\theta d\varphi}{1 + K_F C_F + K_T(D_T r_0 \cos\theta + C_{T0})}\Big]\end{aligned} \tag{7}$$

Letting $K_0 \equiv 1 + K_F C_F + K_T C_{T0}$, we arrive at:

$$\vec{v}_{tot} = \hat{z}\frac{2\pi ak_P K_F C_F}{K_T^2 D_T^2} K_0[\ln(K_0 + K_T D_T r_0) - \ln(K_0 - K_T D_T r_0)] \tag{8}$$

and this expression is clearly constant and positive along the $\hat{z}$ direction. We note briefly the singularity at $K_0 = K_T D_T r_0$ which arises for the same reason as in the case of linear fuel gradient; that is, a portion of the particle's surface lies in an unphysical "negative concentration" region. To avoid this problem, we only consider the case where $K_0 > K_T D_T r_0$.

Expanding the logarithmic term in r_0 gives:

$$\ln\left(\frac{K_0 + K_T D_T r_0}{K_0 - K_T D_T r_0}\right) = \frac{2K_T D_T r_0}{K_0} + \frac{2K_T^3 D_T^3 r_0^3}{3K_0^3} + \cdots \tag{9}$$

demonstrating that the particle velocity is proportional to its radius.

3 Point source foulant gradient

The relevant integral is:

$$\vec{v}_{tot} = -\hat{z}2\pi r_0^2 a k_P K_F C_F \int_0^\pi \frac{\sin\theta\cos\theta d\theta}{A + \left(\frac{B}{\sqrt{r_T^2 - 2r_T r_0\cos\theta + r_0^2}}\right)} \tag{10}$$

where $A = 1 + K_F C_F$, $B = K_T C_1$, r_0 is the particle radius, and r_T is the distance of the center of the particle from the point source. The solution to this integral has a closed form expression given by:

$$\vec{v}_{tot} = \hat{z}\frac{\pi a k_P K_F C_F}{3A^5 r_T^2}\Bigg[4A^3 B r_0^3 + 6A^2 B^2 r_T r_0 - 6AB^3 r_0 + (3B^4 - 3A^2 B^2(r_T^2 + r_0^2))\ln\left(\frac{B + A(r_T + r_0)}{B + A(r_T - r_0)}\right)\Bigg] \tag{11}$$

It is not at all obvious from this equation that the velocity is nonnegative for all values of r_T. To examine the general behavior of the function, we pull a factor of $1/3A^4 r_T^2$ inside the bracket in equation 11 above to get the following expression:

$$\begin{aligned} &\frac{4r_0}{3}\left(\frac{B}{A}\right)\left(\frac{r_0}{r_T}\right)^2 + 2\left(\frac{B}{A}\right)^2\left(\frac{r_0}{r_T}\right) - \frac{2}{r_0}\left(\frac{B}{A}\right)^3\left(\frac{r_0}{r_T}\right)^2 \\ &\quad + \frac{1}{r_0^2}\left(\frac{B}{A}\right)^4\left(\frac{r_0}{r_T}\right)^2 \ln\left[\frac{\frac{B}{A} + r_T + r_0}{\frac{B}{A} + r_T - r_0}\right] \\ &\quad - \left(\frac{B}{A}\right)^2 \ln\left[\frac{\frac{B}{A} + r_T + r_0}{\frac{B}{A} + r_T - r_0}\right] \\ &\quad - \left(\frac{B}{A}\right)^2\left(\frac{r_0}{r_T}\right)^2 \ln\left[\frac{\frac{B}{A} + r_T + r_0}{\frac{B}{A} + r_T - r_0}\right] \end{aligned} \tag{12}$$

A dimensional analysis of the velocity surface integral shows that B/A has units of distance. This means that B/A and r_0 are related by a unitless number, σ. We evaluate the above equation for $B/A = \sigma r_0$ and we let $r_0/r_T = \rho$, noting that the domain of ρ is $0 < \rho < 1$ and the domain of σ is $0 < \sigma < \infty$. We obtain the following expression after dividing by $2\sigma r_0^2$ and converting the logarithms to inverse hyperbolic cotangents:

$$\begin{aligned} f(\sigma,\rho) =& \frac{2}{3}\rho^2 + \sigma\rho + \sigma^3\rho^2 \coth^{-1}\left(\sigma + \frac{1}{\rho}\right) \\ & - \sigma^2\rho^2 - (1+\rho^2)\sigma \coth^{-1}\left(\sigma + \frac{1}{\rho}\right) \end{aligned} \tag{13}$$

We show that $f(\sigma,\rho) > 0$ everywhere, which is equivalent to showing that velocity is always positive along the $\hat{z}$ axis, in the direction of the source. To do so, we first substitute $x = \sigma + 1/\rho$, noting that the domain of x is $1 < x < \infty$, and we rewrite the inverse hyperbolic cotangents as a series to get:

$$\begin{aligned} f(x,\rho) =& \frac{2}{3}\rho^2 + 3x\rho - 2 + x^2\rho^2 \\ & + (x^3\rho^2 + 2x + \rho - 3x^2\rho - x\rho^2) \sum_{n=0}^{\infty} \frac{1}{(2n+1)x^{2n+1}} \end{aligned} \tag{14}$$

Through straightforward, albeit tedious, algebraic manipulation, we can recast this equation in the following form:

$$\begin{aligned} f(x,\rho) = \sum_{n=0}^{\infty} \Bigg[& \frac{4n + 10 - 2\rho^2}{(2n+3)(2n+5)x^{2n+2}} \\ & - \frac{4n\rho + 4\rho}{(2n+3)(2n+5)x^{2n+3}} \Bigg] \end{aligned} \tag{15}$$

Since $0 < \rho < 1$, both terms in the sum are positive. Since $x > 1$, the series converges absolutely and the difference between the terms (and therefore the total function $f(x,\rho)$) will be positive if:

$$4n + 10 - 2\rho^2 > 4n\rho + 4\rho \tag{16}$$

or equivalently:

$$4n(1-\rho) + 2(5 - 2\rho - \rho^2) > 0 \tag{17}$$

Since $0 < \rho < 1$, this expression is always true. Therefore, the velocity is always positive along the $\hat{z}$ axis in the direction of the foulant source. The final expression for velocity is:

$$\vec{v}_{tot} = \hat{z}\frac{2\pi r_0 a k_P K_F C_F B}{A^2} f(x,\rho) \tag{18}$$

The admittedly exhaustive analysis of the velocity presented above is necessary in light of two facts: a) the point source gradient is likely to be useful in a realistic modeling scenario, and b) the solution to this problem given by equations 11-13 above is extremely numerically unstable for large σ. The instability comes from the fact that a calculation of $f(\sigma, \rho)$ from equation 13 is dominated by the difference:

$$\sigma^3 \rho^2 \coth^{-1}\left(\sigma + \frac{1}{\rho}\right) - \sigma^2 \rho^2 \approx (\sigma + \epsilon)^2 - \sigma^2 \tag{19}$$

From the series expression in equation 15 above, we see that this difference ends up being $O(1/\sigma^2)$, and such a small difference of large numbers leads to a significant amount of rounding error at large values of σ.

Equation 15 above gives us one more important insight: the leading term of the series is proportional to $1/r_T^2$.