Supplementary data

Influence of graphene oxide nanoparticles on the transport and cotransport of biocolloids in saturated porous media

by

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Description of microbial cultures, and preparation of suspensions and colony formation.

The microbial cultures were maintained at –80 °C (as frozen stock) in eppendorf safe lock tubes containing growth media supplemented with 50% glycerol. Prior to each experiment, duplicates of all bacteria were cultured in sterile petri dishes, containing non-selective medium (Nutrient Agar), and were incubated in an oven at 37 ° C for 48 hours, till the forming of visible colonies. Fresh bacterial suspensions were prepared by isolating and homogeneously dispersing a sort amount of the sufficiently formed colonies of each microbial culture in sterile tubes containing 20 mL PBS solution ($Na_2HPO_4{}^{-}2H_20/KH_2PO_4$, I_s=2 mM, pH=7). The inoculated PBS tubes were incubated at 37°C for 10 min to adjust the inoculums standard to a 0.5 McFarland, so that 0.1 optical density of a uniform microbial suspension at 600 nm corresponds approximately to a concentration of 10^8 CFU/mL. A UV–visible spectrophotometer (UVmini-1240, Shimadzu) was used to obtain the optical density measurements at 600 nm. The dense bacterial suspensions were diluted with appropriate volume of PBS solution to obtain a cell concentration of ~10^5 CFU/mL, which was used as the initial bacterial concentration for both transport and cotransport experiments. For the cotransport experiments, the three bacterial strains were co-present in one microbial suspension, maintaining its total cell concentration at 10^5 CFU/mL. The concentration of bacteria was determined by conducting serial decimal dilution of the sample with PBS and plating out (in duplicates) the aliquot (300 μL) on the surface of the nutrient media Harlequin (*E.coli*/Coliform Medium, product code HAL008), Slanetz and Bartely Agar (code LAB166) and Mannitol Salt Agar for the growth of *E. coli*, *E. faecalis* and *S. aureus*, respectively. The plates were incubated at 37 °C for 48 h and the total number of colonies was counted. Note that reliable dilutions for quantification were considered to be the ones resulting in formation of 30-300 distinct colonies. Concentration of bacteria in the media was calculated taking into account dilution of the sample and the amount plated out on the solid media and was reported as colony-forming units per milliliter (CFU/mL).

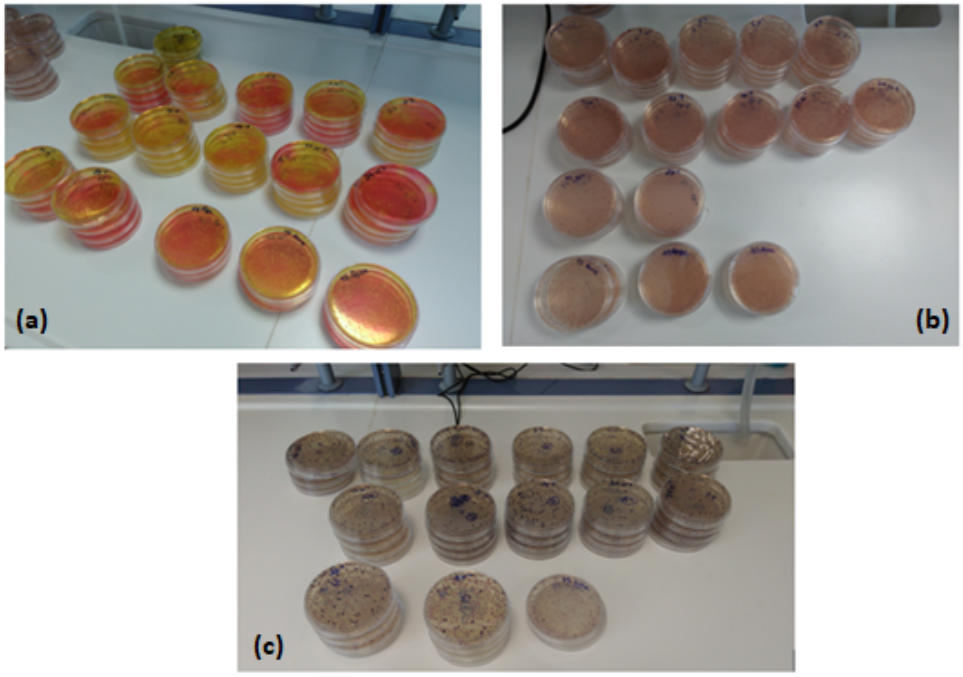

Fig S1: Images of petri dishes containing selective nutrient media after incubation and colony formation of: (a) *S. aureus*, (b) *E. faecalis*, and (c) *E. coli*.

GO Calibration curve

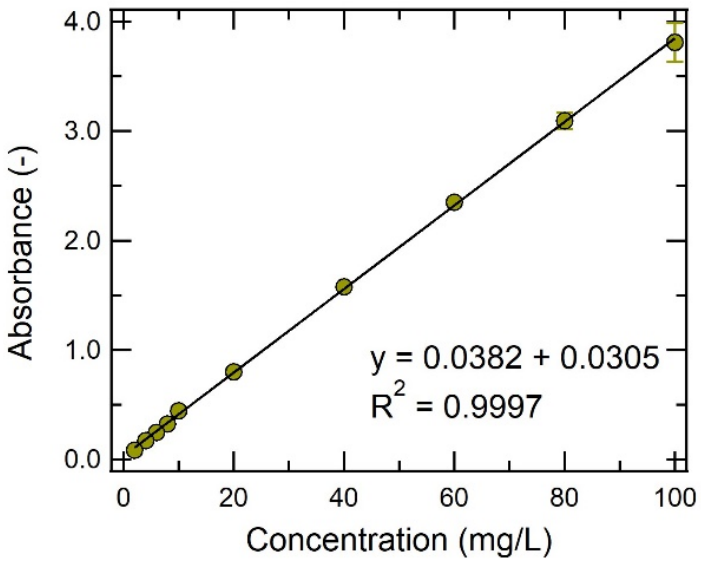

Fig S2: Calibration curve of GO for the experimental conditions (i.e. I_s=2 mM, pH=7).

GO transport and cotransport experimental data

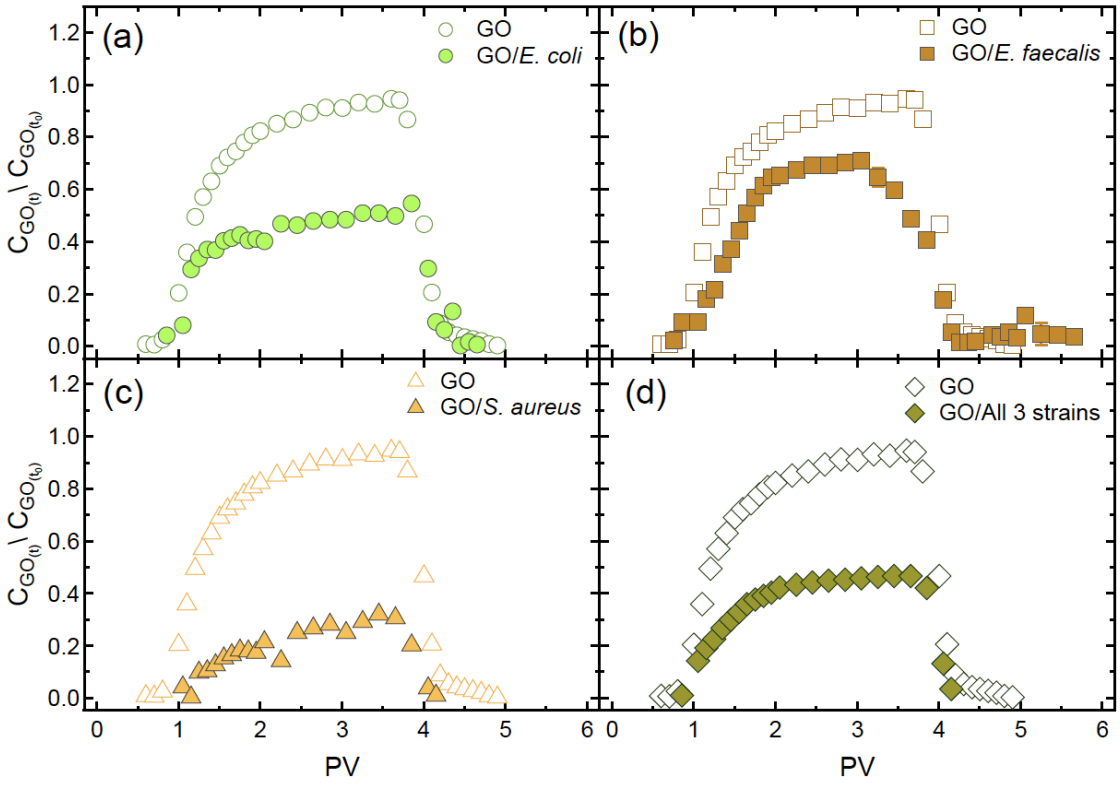

Fig S3: Normalized experimental breakthrough concentrations of GO NPs in presence of: (a) *E. coli*, (b) *S. aureus*, (c) *E. faecalis*, and (d) all three co-existing bacterial strains.

Zeta potential (ζ) and hydrodynamic diameter (d_H) measurements

Table S1. Measured zeta potentials and hydrodynamic diameters.

Particles	Experimental Conditions			Measurements	
	pH	I_S (mM)	T (°C)	ζ-potential (mV)	d_H (nm)
Escherichia coli (DMS 498)	7	2	25	-38.9 ± 5.5	1090.00 ± 62.0
Enterococcus faecalis (ATCC 14506)	7	2	25	-43.1 ± 2.3	1081.9 ± 102.9
Staphylococcus aureus (isolated from poultry sample)	7	2	25	-36.7 ± 1.9	729.9 ± 85.9
GO NPs	7	2	25	-39.6 ± 1.9	546.3 ± 43.4
Coarse Quartz sand (>850 µm, sieve No. 20, coarse sand)	7	2	25	-57.3 ± 2.1	

Biocolloid transport

The individual transport of the suspended biocolloids in one-dimensional, water saturated and homogenous porous media, under uniform flow, accounting for first order kinetics of attachment and inactivation of bacteria suspended in the aqueous phase and attached onto the solid matrix is described by the following partial differential equation, which was first introduced by Sim and Chrysikopoulos [1]:

$$\frac{\partial C_b}{\partial t} + \frac{\rho_s}{\theta}\frac{\partial C_b^*}{\partial t} = D_x \frac{\partial^2 C_b}{\partial x^2} - U_x \frac{\partial C_b}{\partial x} - \lambda_b C_b - \lambda_b^* \frac{\rho_s}{\theta} C_b^* \qquad \text{(S1)}$$

where C_b [CFU/mL] and C_b^* [CFU/mL] are the concentration of biocolloids in suspension and attached on the solid matrix respectively; D_x [L^2/t] is the longitudinal hydrodynamic dispersion coefficient [2], defined as:

$$D_x = \alpha_L U_x + \mathcal{D}_e \qquad \text{(S2)}$$

where α_L [L] is the longitudinal dispersivity; $\mathcal{D}_e$ [L^2/t] is the effective molecular diffusion coefficient which given from relation $\mathcal{D}/\tau^*$, where $\mathcal{D}$ [L^2/t] is the molecular diffusion coefficient of each biocolloids particle and $\tau^* \geq 1$ [-] is the tortuosity; θ [-] is the porosity of the porous medium; U_x [L/t] is the interstitial velocity, defined as q_{Darcy}/θ; ρ_s [M/L^3] is the bulk density of the solid matrix; λ_b [1/t] is the inactivation rate of the biocolloids suspended in solution; λ_b^* [1/t] is the inactivation rate of the attached biocolloids; and t is time.

The second term of equation (SI1) describes the rate of attachment of the suspended biocolloids to the solid matrix (i.e., sand) and is mathematically described by the following first order equation [3,4]:

$$\frac{\rho_s}{\theta}\frac{\partial C_b^*}{\partial t} = r_{b-b^*} C_b - r_{b^*-b} \frac{\rho_s}{\theta} C_b^* \qquad \text{(S3)}$$

where r_{b-b^*} [1 / t] and r_{b^*-b} [1 / t] are the attachment and detachment rate constants of the biocolloid to and from the solid matrix, respectively.

The representative initial and boundary conditions, for a semi-infinite and one-dimensional medium, under the assumption of a continuous feed of biocolloids at the column inlet (broad pulse), are described by the following equations [1,5]:

$$C_b(0,x)=0 \tag{S4}$$

$$-D_x \frac{\partial C_b(t,0)}{\partial x} + U_x C_b(t,0) = \begin{cases} U_x C_{b_0}, & t \le t_p \\ 0, & t > t_p \end{cases} \tag{S5}$$

$$\frac{\partial C_b(t,\infty)}{\partial x} = 0 \tag{S6}$$

where C_{b_0} [M/L^3] is the source concentration of biocolloidal suspension and t_p [t] is the duration of the microbial suspension injection. The initial boundary condition (S4) assumes an absence of biocolloid concentration within the experimental column when injection process begins. The upstream boundary condition (S5) refers to a continuous source at the inlet that provides constant concentration of biocolloids over a predefined pulse period (t_p) (i.e. constant flux at the inlet). While the downstream boundary condition (6) preserves the biocolloidal concentration continuity within the semi-infinite medium [6].Thus, the analytical solution in conjunction with relationship (S3), subject to conditions (S4)–(S6) has been reported by Sim and Chrysikopoulos [1] as follows:

$$C_b(t,x) = \begin{cases} \Omega(t,x), & 0 < t \le t_p \\ \Omega(t,x) - \Omega(t-t_p,x), & t > t_p \end{cases} \tag{S7}$$

where

$$\Omega(t,x)=\frac{U_x C_{b_0}}{D_x^{0.5}}\exp\left[\frac{U_x x}{2D_x}\right]\left\{\int_0^t\int_0^\tau He^{-Ht}J_0\left[2\left(B\xi\left(\tau-\xi\right)\right)^{0.5}\right]\right.$$
$$\cdot\left\{\frac{1}{(\pi\xi)^{0.5}}\exp\left[\frac{-x^2}{4D_x\xi}+\left(H-A-\frac{U_x^{\,2}}{4D_x}\right)\xi\right]\right.$$
$$-\frac{U_x}{2D_x^{\,0.5}}\exp\left[\frac{U_x x}{2D_x}+(H-A)\xi\right]$$
$$\left.\left.\cdot\operatorname{erfc}\left[\frac{x}{2\left(D_x\xi\right)^{0.5}}+\frac{U_x}{2}\left(\frac{\xi}{D_x}\right)^{0.5}\right]\right\}d\xi\,d\tau\right.$$
$$+e^{-Ht}\int_0^t J_0\left[2\left(B\xi\left(\tau-\xi\right)\right)^{0.5}\right]$$
$$\cdot\left\{\frac{1}{(\pi\xi)^{0.5}}\exp\left[\frac{-x^2}{4D_x\xi}+\left(H-A-\frac{U_x^{\,2}}{4D_x}\right)\xi\right]\right.$$
$$-\frac{U_x}{2D_x^{\,0.5}}\exp\left[\frac{U_x x}{2D_x}+(H-A)\xi\right]$$
$$\left.\left.\cdot\operatorname{erfc}\left[\frac{x}{2\left(D_x\xi\right)^{0.5}}+\frac{U_x}{2}\left(\frac{\xi}{D_x}\right)^{0.5}\right]\right\}d\xi\right\} \tag{S8}$$

where J_0 is the Bessel function of the first kind of zeroth order; “exp” and “erfc” are the exponential function and the complementary error function, respectively; ξ and τ are dummy integration variables. The parameters A, B and H are given from the following equations:

$$A=r_{b-b^*}+\lambda_b \tag{S9}$$

$$B=r_{b-b^*}\left(\lambda_b^{\,*}-H\right) \tag{S10}$$

$$H=r_{b^*-b} \tag{S11}$$

The above analytical solution is incorporated in the nonlinear least squares regression software ColloidFit [7] and the various model parameters were estimated by fitting the analytical solution to the experimental data.

Table S2. Fitted model parameter values and 95% confidence intervals, as obtained with the software ColloidFit [7].

Transport experiment	D_X [cm^2/min]	$a_{L,b}$ [cm]	$r_{b\text{-}b^*}$ [1/min]	$r_{b^*\text{-}b}$ [1/min]	λ_b [1/min]	λ_b^* [1/min]
E. coli	0.053 ± 0.026	0.121	4.09×10^{-6}	8.10×10^{-1}	2.58×10^{-4} ± 1.61×10^{-4}	1.29×10^{-4} ± 8.07×10^{-5}
E. faecalis	0.106 ± 0.073	0.251	9.63×10^{-5}	1.00×10^{-4}	5.40×10^{-3} ± 8.07×10^{-5}	2.60×10^{-3} ± 4.04×10^{-5}
S. aureus	0.214 ± 0.109	0.498	8.21×10^{-5}	4.74×10^{-2}	6.90×10^{-3} ± 1.61×10^{-3}	3.45×10^{-3} ± 8.10×10^{-4}

Moments analysis

The breakthrough data of biocolloid concentration collected at the exit of the experimental column (x=L) were analyzed by normalized absolute temporal moments [8]:

$$M_n(x) = \frac{m_n(x)}{m_0(x)} = \frac{\int_0^\infty t^n C_b(x,t)dt}{\int_0^\infty t^0 C_b(x,t)dt} = \frac{\int_0^\infty t^n C_b(x,t)dt}{\int_0^\infty C_b(x,t)dt} \quad \text{(S12)}$$

where the subscript n=0, 1, 2, … indicates the order of the moment, the subscript i indicates *E. coli*, *E. faecalis*, and *S. aureus* and m_n is the governing equation for the absolute temporal moments. The zeroth absolute temporal moment, $m_0\left[(CFU_{(t)}\times m/L)/(CFU_{(t_0)}\times m/L)\right]$, quantifies the total mass of biocolloids passed through the porous media, thus the total biocolloidal mass which enclosed under the concentration breakthrough curve. In the present study, only the first normalized temporal moment, M_1 [t], was determined using the fitting software ColloidFit. Note that M_1 characterizes the center of mass of the concentration breakthrough curve, defining the mean breakthrough time or average velocity. In addition, quantification of the recovered mass, M_r, of the suspended biocolloids at the exit of the column was achieved using the ColloidFit software by applying the following mathematical relationship [9]:

$$M_{r_b}(L) = \frac{m_0(L)}{C_{b_0} t_p} = \frac{\int_0^\infty C_b(L,t)dt}{\int_0^{t_p} C_b(0,t)dt} \quad \text{(S13)}$$

Colloid filtration theory (CFT)

The dimensionless collision efficiency α [-], was calculated from the breakthrough curves, based on the following model proposed by Rajagopalan and Tien [10]:

$$\alpha = -\frac{d_s \ln(RB)}{3(1-\theta)\eta_0 L} \quad \text{(S14)}$$

where d_s is the average grain diameter of quartz sand, which is related to the forward rate constant of attachment, r_{b-b^*}, through the following equation [11,12]:

$$\frac{r_{b-b^*}}{\alpha} = \frac{3(1-\theta)}{2d_s} U_x \eta_0 \quad \text{(S15)}$$

RB [-] is the ratio of the recovered biocolloid concentration when equilibrium has been reached in the porous medium (steady state conditions), C_{bss} [$CFU_{(tss)}$/mL], relative to the initial concentration of the injected microbial suspension, C_{b_0} [$CFU_{(t0)}$/mL]:

$$RB = \frac{C_{b_{ss}}}{C_{b_0}} \quad \text{(S16)}$$

For favorable deposition (i.e. in absence of double layer interaction energy), the parameter $\eta_{0,}$ which symbolizes the dimensionless single-collector removal efficiency, is given from the following correlation [13]:

$$\eta_0 = \eta_D + \eta_I + \eta_G \quad \text{(S17)}$$

where the dimensionless coefficients η_D, η_I, and η_G, depend on mechanisms of diffusion, interception, and sedimentation, respectively:

$$\eta_D = 2.4 A_s^{1/3} N_R^{-0.081} N_{Pe}^{-0.715} N_{vdW}^{0.052} \quad \text{(S18)}$$

$$\eta_I = 0.55 A_s N_R^{1.675} N_A^{0.125} \quad \text{(S19)}$$

$$\eta_G = 0.22 N_R^{-0.24} N_G^{1.11} N_{vdW}^{0.053} \quad \text{(S20)}$$

A_s is the Happel flow parameter; N_R is the relative size number; N_{Pe} is the Peclet number; N_{vdW} is the van der Waals number; N_A is the attraction number; and N_G is the gravity number, which are expressed mathematically as follows:

$$A_s = \frac{2\left(1-\varepsilon_\theta{}^5\right)}{2-3\varepsilon_\theta+3\varepsilon_\theta{}^5-2\varepsilon_\theta{}^6} \quad \text{(S21)}$$

where

$$\varepsilon_\theta = (1-\theta)^{1/3} \quad \text{(S22)}$$

$$N_R = \frac{d_b}{d_s} \quad \text{(S23)}$$

$$N_{Pe} = \frac{d_s q}{\mathcal{D}} \quad \text{(S24)}$$

$$N_{vdW} = \frac{A_{123}}{k_B T} \quad \text{(S25)}$$

$$N_A = \frac{N_{vdW}}{N_R N_{Pe}} \quad \text{(S26)}$$

$$N_G = \frac{d_b{}^2\left(r_b - r_w\right)g}{18\mu_w q} \quad \text{(S27)}$$

Also, the diffusion coefficient is described by the Stokes-Einstein equation:

$$\mathcal{D} = \frac{k_B T}{3\pi\mu_w d_b} \quad \text{(S28)}$$

In the above equations, d_b is the measured diameter of the biocolloid particle (values listed in Table 1); ρ_b is the biocolloid particle density (1091 kg/m^3 for *E. coli* [14,15], 1132 kg/m^3 for *E. faecalis* [16]; 1693 kg/m^3 for *S. aureus* [17,18] and 2200 kg/m^3 for GO NPs [19,20]; ρ_f=999.7 kg/m^3 is the fluid density at the absolute temperature of 298 K; μ_w=8.91×10^{-4} kg/(m·s) is the absolute fluid viscosity; and g=9.81 m/s^2 is the gravitational acceleration.

Extended DLVO theory of colloid stability

The extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) theory treats the total interaction energy, Φ_{DLVO}, between two smooth, homogeneous surfaces or particles with ideal geometries as the sum of an attractive energy due to van der Waals forces, Φ_{vdW}, an electrostatic repulsion energy arising from the overlap of electrical double layers, Φ_{dl}, the Born repulsion energy due to overlapping electron orbitals of the molecules comprising the different surfaces at very close

separation distances, Φ_{Born} [21], as well as the Lewis acid-base interactions, Φ_{AB} [22]:

$$\Phi_{XDLVO}(h) = \Phi_{vdW}(h) + \Phi_{dl}(h) + \Phi_{Born}(h) + \Phi_{AB}(h) \quad \text{(S29)}$$

In the present study, the interactions between bacteria (*Escherichia coli, Enterococcus faecalis* and *Staphylococcus aureus*) and quartz sand, bacteria and bacteria, and GO and bacteria were modeled based on XDLVO theory using the following approximations for sphere-plate (i.e. bacteria-sand) interactions [23-27]:

$$\Phi_{vdW}(h) = -\frac{A_{123} r_p}{6h}\left[1 + \left(\frac{14h}{\lambda}\right)\right]^{-1} \quad \text{(S30)}$$

$$\Phi_{dl}(h) = \pi\varepsilon_r\varepsilon_0 r_p\left[2\Psi_p\Psi_s \ln\left(\frac{1+e^{-\kappa h}}{1-e^{-\kappa h}}\right) + \left(\Psi_p^2 + \Psi_s^2\right)\ln\left(1-e^{-2\kappa h}\right)\right] \quad \text{(S31)}$$

$$\Phi_{Born}(h) = \frac{A_{123}\ \sigma_{Born}^6}{7560}\left[\frac{8r_p + h}{\left(2r_p + h\right)^7} + \frac{6r_p - h}{h^7}\right] \quad \text{(S32)}$$

$$\Phi_{AB}(h) = 2\pi r_p \lambda_{AB} \Phi_{AB(h=h_o)} \exp\left[\frac{h_0 - h}{\lambda_{AB}}\right] \quad \text{(S33)}$$

and the following approximations for sphere-sphere (i.e. bacteria-bacteria and GO-bacteria) interactions [24,28-29]:

$$\Phi_{vdW}(h) = -\frac{A_{123}}{12}\left\{\frac{R_p}{\xi^2 + \xi R_p + \xi} + \frac{R_p}{\xi^2 + \xi R_p + \xi + R_p} + 2\ln\left[\frac{\xi^2 + \xi R_p + \xi}{\xi^2 + \xi R_p + \xi + R_p}\right]\right\} \quad \text{(S34)}$$

where

$$R_p = \frac{r_{p2}}{r_{p1}} \quad \text{(S35)}$$

$$\xi = \frac{h + r_{p1} + r_{p2}}{2r_{p1}} \quad \text{(S36)}$$

$$\Phi_{dl}(h) = \pi\varepsilon_r\varepsilon_0 \frac{r_{p1}r_{p2}}{(r_{p1}+r_{p2})}\left[2\Psi_{p1}\Psi_{p2}\ln\left(\frac{1+e^{-\kappa h}}{1-e^{-\kappa h}}\right)+\left(\Psi_{p1}^2+\Psi_{p2}^2\right)\ln\left(1-e^{-2\kappa h}\right)\right] \quad \text{(S37)}$$

$$\begin{aligned}\Phi_{Born}(h)=\frac{A_{123}}{75600\xi}\left(\frac{\sigma_{Born}}{r_{p1}}\right)^6 \Bigg[& \frac{-4\xi^2-14(R_p-1)\xi-6(R_p^2-7R_p+1)}{(2\xi-1+R_p)^7} \\ & +\frac{-4\xi^2+14(R_p-1)\xi-6(R_p^2-7R_p+1)}{(2\xi+1-R_p)^7} \\ & +\frac{4\xi^2+14(R_p-1)\xi+6(R_p^2+7R_p+1)}{(2\xi+1+R_p)^7} \\ & +\frac{4\xi^2-14(R_p-1)\xi+6(R_p^2+7R_p+1)}{(2\xi-1-R_p)^7}\Bigg]\end{aligned} \quad \text{(S38)}$$

$$\Phi_{AB}(h) = 2\pi\frac{r_{p1}r_{p2}}{r_{p1}+r_{p2}}\lambda_{AB}\Phi_{AB(h=h_o)}\exp\left[\frac{h_0-h}{\lambda_{AB}}\right] \quad \text{(S39)}$$

where h [nm] is the separation distance between the approaching surfaces, A_{123} [J] represents the combined Hamaker constant for substances “1” and “3” in medium “2” [(1-colloid)-(2-water)-(3-collector)], $\lambda\approx10^{-7}$ m is the characteristic wavelength, $\varepsilon_r=\varepsilon/\varepsilon_0$ is the dimensionless relative dielectric constant of the suspending liquid (for the water $\varepsilon_r=75$), ε [$C^2/(J\cdot m)$] is the dielectric constant of the suspending liquid, ε_0 [$C^2/(J\cdot m)$] is the dielectric permittivity of free space ($\varepsilon_0=8.85\times10^{-12}$ $C^2/(J\cdot m)$), r_p [m] is the colloid particle (i.e., biocolloids or GO) radius, Ψ_p [V] is the surface potential of the colloid particle, Ψ_s [V] is the surface potential of the collector surface (plate), and κ [1/m] is the inverse of the diffuse layer thickness, known as the Debye-Hückel parameter [25]:

$$\kappa = \left[\frac{2\,I_s\,A_N 1000e^2}{e_r e_0 k_B T}\right]^{1/2} \quad \text{(S40)}$$

where I_s [mol/L] is the ionic strength, $A_N=6.02\times10^{23}$ [1/mol] is Avogadro's number, $e=1.602\times10^{-19}$ [C] is the elementary charge, $k_B=1.38\times10^{-23}$ [J/K] is the Boltzmann constant, and $T=298$ [K] is the fluid absolute temperature. Note that the commonly used value of the Born collision parameter: $\sigma_{Born}=5$ Å [25], results in an acceptable minimum separation distance, at $h=h_0$ (i.e. at “contact”), equal to $h_0\approx$

2.5 Å = 0.25 nm. For the estimation of Lewis acid-base free energy of interaction between two surfaces at $h=h_0=0.25$ nm, $\Phi_{AB(h=h0)}$ [J/m^2], the Yoon et al. [30] empirical approach, based on the determination of the degree of hydrophobicity using water contact angles, was employed:

$$\Phi_{AB(h=h_o)} = -\frac{K_{123}}{2\pi h_0 \lambda_{AB}} \quad \text{(S41)}$$

where λ_{AB}= 1 nm [26], was used as the decay (Debye) length of water, and the hydrophobic force constant, K_{123} [J], was predicted by the following empirical relationship:

$$\log K_{123} = -7.0\left(\frac{\cos\beta_1+\cos\beta_3}{2}\right)-18.0 \quad \text{(S42)}$$

where β_1 [°] and β_3 [°] are the water contact angles of materials "1" and "3", respectively.

In this study, the combined Hamaker constants for the systems GO-water-quartz sand was set to $A_{123}=1.92\times10^{-21}$ J [31], and for bacteria-water-quartz sand was set to $A_{123}=6.5\times10^{-21}$ J [32]. Moreover, the combined Hamaker constants for GO-water-GO was set to $A_{121}=2.23\times10^{-21}$ J [33], for bacteria-water-bacteria was set to $A_{323}=6.8\times10^{-20}$ J [34] and for GO-water-bacteria the geometric mean combining rule was used [30]:

$$A_{123}=\sqrt{A_{121}\times A_{323}} \quad \text{(S43)}$$

to obtain $A_{123}=1.23\times10^{-20}$ J. Furthermore, the following contact angles were employed: $\beta_{GO}=26.8°$ [35], $\beta_{E.\ coli}=22.2\pm0.6°$ [36], $\beta_{E.\ faecalis}=36\pm2°$ [37], $\beta_{S.\ aureus}=21.9\pm0°$ [38], and $\beta_{sand}=70.8\pm0.5°$ for clean quartz sand [39]. Note that graphene is a neutral material with a water contact angle measured within the range of 87–127° [40-42]. However, GO displays hydrophilic properties with $\beta_{GO}\approx30–60°$ [43,44].

Table S3. Parameter values employed in the theoretical considerations.

Parameter	Symbol	Values	References
Combined Hamaker constants for the system GO-water-quartz sand	A_{123}	1.92×10^{-21} J	Chrysikopoulos et al. (2017) Ref. [31] of Supporting data
Combined Hamaker		6.5×10^{-21} J	Rijnaarts et al. (1995)

constants for the system bacteria-water-quartz sand			Ref. [32] of Supporting data
Combined Hamaker constants for the system GO-water-GO		2.23×10^{-21} J	Mcallister et al. (2007) Ref. [33] of Supporting data
Combined Hamaker constants for the system bacteria-water-bacteria		6.8×10^{-20} J	Rijnaarts et al. (1999) Ref. [34] of Supporting data
Combined Hamaker constants for the system GO-water-bacteria		1.23×10^{-20} J	measured in this study (based on the geometric mean combining rule; see Ref. [30] of the Supporting data)
Characteristic wavelength	λ	10^{-7} m	
Hydrodynamic diameter of *E. coli*	d_p	1090.0±62.0 nm	measured in this study
Hydrodynamic diameter of *E. faecalis*		1081.9±102.9 nm	measured in this study
Hydrodynamic diameter of *S. aureus*		729.9±85.9 nm	measured in this study
Hydrodynamic diameter of GO NPs		546.3±43.4 nm	measured in this study
Dimensionless relative dielectric constant of water	$\varepsilon_r=\varepsilon/\varepsilon_0$	75 [-]	
Dielectric permittivity of free space	ε_0	8.85×10^{-12} $C^2/(J\cdot m)$	
z-potential of *E. coli*	ζ	-38.9±5.5 mV	measured in this study
z-potential of *E. faecalis*		-43.1±2.3 mV	measured in this study
z-potential of *S. aureus*		-36.7±1.9 mV	measured in this study
z-potential of GO NPs		-39.6±1.9 mV	measured in this study
z-potential of coarse quartz sand		-57.3±2.1 mV	measured in this study
Inverse of the diffuse layer thickness (Debye-Hückel parameter)	κ^{-1}	3.06×10^{-8} m	Ruckenstein and Prieve (1976) Ref. [25] of Supporting data
Ionic strength	I_s	2×10^{-3} mol/L	this study
Avogadro's number	N_A	6.02×10^{23} 1/mol	
Elementary charge	e	1.602×10^{-19} C	
Boltzmann constant	k_B	1.38×10^{-23} J/K	
Fluid absolute temperature	T	298 K	this study
Born collision parameter	σ_{Born}	5 Å	Ruckenstein and Prieve (1976) Ref. [25] of Supporting data
Decay (Debye) length of water	λ_{AB}	1 nm	van Oss (1993) Ref. [26] of Supporting data
Water contact angles of materials	β_{GO}	26.8°	Wei et al. (2014) Ref. [35] of Supporting data
	$\beta_{E.\ coli}$	22.2±0.6°	Daffonchio (1995) Ref. [36] of Supporting data
	$\beta_{E.\ faecalis}$	36±2°	Gallardo-Moreno (2002) Ref. [37] of Supporting data
	$\beta_{S.\ aureus}$	21.9±0°	Hamadi and Latrache (2008) Ref. [38] of Supporting data
	β_{sand}	70.8±0.5°	Chen and Zhu (2005) Ref. [39] of Supporting data

Table S4: Calculated Φ_{max1}, Φ_{min1}, and Φ_{min2} values for sphere-plate and sphere-sphere models based on the XDLVO theory.

Conditions pH, I_S (mM)	h (nm)	Φ_{min1} (k_BT)	h (nm)	Φ_{max1} (k_BT)	h (nm)	Φ_{min2} (k_BT)
***E. coli*-Quartz sand (Sphere – plate)**						
7, 2	-	n.d.	19.20	126.30	79.00	-1.239×10^{-1}
***E. faecalis*-Quartz sand (Sphere-plate)**						
7, 2	-	n.d.	11.17	425.60	79.93	-1.205×10^{-1}
***S. aureus*-Quartz sand (Sphere – plate)**						
7, 2	-	n.d.	19.41	77.14	78.47	-8.392×10^{-2}
GO-Quartz sand (Sphere – plate)						
7, 2	-	n.d.	10.93	214.70	90.43	-1.448×10^{-2}
Bacteria-Bacteria (Sphere – sphere)						
E. coli- E. coli						
7, 2	-	n.d.	27.73	12.71	106.07	-7.633×10^{-3}
E. faecalis- E. faecalis						
7, 2	-	n.d.	11.61	153.00	107.43	-7.583×10^{-3}
S. aureus- S. aureus						
7, 2	-	n.d.	28.19	7.093	100.89	-6.625×10^{-3}
E. coli- E. faecalis						
7, 2	-	n.d.	19.63	45.11	106.75	-7.608×10^{-3}
E. coli- S. aureus						
7, 2	-	n.d.	27.95	9.308	104.09	-6.467×10^{-3}
E. faecalis-S. aureus						
7, 2	-	n.d.	19.85	33.08	104.75	-6.465×10^{-3}
GO-GO (Sphere-sphere)						
7, 2	-	n.d.	11.23	68.60	123.55	-1.639×10^{-4}
GO-Bacteria (Sphere-sphere)						
GO- *E. coli*						
7, 2	-	n.d.	19.43	28.55	116.41	-8.670×10^{-4}
GO- *E. faecalis*						
7, 2	-	n.d.	11.41	97.61	117.07	-8.685×10^{-4}
GO- *S. aureus*						
7, 2	-	n.d.	19.65	22.40	112.45	-1.001×10^{-3}

Table S5. Calculated values of $\Phi_{AB(h=h0)}$ (PBS solution, pH=7, I_S=2 mM).

Interacting materials	$\Phi_{AB(h=h0)}$ (mJ/m^2)
E. coli-Quartz sand	-4.231×10^{6}
E. faecalis-Quartz sand	-4.472×10^{3}
S. aureus-Quartz sand	-4.876×10^{6}
GO-Quartz sand	-3.465×10^{3}
GO-GO	-3.007×10^{3}
E. coli-E. coli	-4.485×10^{9}
E. faecalis-E. faecalis	-5.008×10^{3}

S. aureus-S. aureus	-5.954 x 10^9
E. coli-E. faecalis	-4.739 x 10^6
E. coli-S. aureus	-5.168 x 10^9
E. faecalis-S. aureus	-5,460 x 10^6
GO-*E. coli*	-3.672 x 10^6
GO-*E. faecalis*	-3.880 x 10^3
GO-*S. aureus*	-4.231 x 10^6

Estimation of normalized standard deviation (SD) values

The range (N) and standard deviation (SD) are measures of the experimental data spreading, used as descriptive error bars. Range error bars encompass the lowest and highest values. In this work, the normalized SD values were calculated by the following equation for N=3:

$$SD = \left(\frac{1}{\bar{C}_{b0}} \right) \sqrt{\frac{\sum_{i=1}^{N} (C_{b,i(t)} - \bar{C}_{b(t)})^2}{N-1}} \quad \text{(S44)}$$

where $C_{b,i(t)}$ (CFU/mL) is the experimental concentration data at a given time interval, $\bar{C}_{b(t)}$ (CFU/mL) is the average microbial concentration at a given time interval resulting from three experimental concentration measurements at the same time interval, Σ is the sum of N experimental concentrations obtained at the examined time interval, and SD (-) is the difference between the experimental concentration data obtained at a given time interval and the average concentration at the same time interval, normalized by the initial average concentration (CFU/mL) of the examined microbial suspension.

Table S6. Experimental concentrations and associated estimated SD values (for N=3), normalized with the initial average concentration (CFU/mL), for the transport experiments.

Transport						
	E. coli		***E. faecalis***		***S. aureus***	
PV	**$C_{b(t)}$ / C_{b0}**	**SD normalized (N=3)**	**$C_{b(t)}$ / C_{b0}**	**SD normalized (N=3)**	**$C_{b(t)}$ / C_{b0}**	**SD normalized (N=3)**
0.00	0.000	0.000	0.000	0.000	0.000	0.000
0.20	0.000	0.000	0.000	0.000	0.000	0.000
0.40	0.000	0.000	0.000	0.000	0.000	0.000

0.60	0.000	9.959×10^{-5}	0.000	0.000	0.000	0.000
0.70	0.023	2.963×10^{-3}	0.002	6.511×10^{-4}	0.007	1.261×10^{-3}
0.80	0.231	1.593×10^{-2}	0.030	7.893×10^{-3}	0.033	3.760×10^{-3}
1.00	0.849	7.967×10^{-2}	0.445	2.850×10^{-2}	0.126	5.405×10^{-3}
1.10	0.924	3.735×10^{-2}	0.555	4.385×10^{-3}	0.293	9.608×10^{-2}
1.20	1.003	7.085×10^{-2}	0.691	4.385×10^{-3}	0.378	6.005×10^{-2}
1.30	1.025	5.478×10^{-2}	0.678	2.412×10^{-2}	0.766	3.303×10^{-2}
1.40	1.033	6.722×10^{-2}	0.648	4.824×10^{-2}	0.637	6.005×10^{-2}
1.50	0.993	2.158×10^{-1}	0.715	8.113×10^{-2}	0.550	2.102×10^{-2}
1.60	1.019	1.263×10^{-1}	0.715	4.604×10^{-2}	0.726	2.882×10^{-1}
1.70	0.984	1.881×10^{-1}	0.716	5.701×10^{-2}	0.766	9.308×10^{-2}
1.80	0.961	9.025×10^{-2}	0.643	5.481×10^{-2}	0.605	5.705×10^{-2}
1.90	0.956	4.233×10^{-2}	0.650	5.481×10^{-2}	0.607	9.008×10^{-2}
2.00	0.996	5.818×10^{-2}	0.733	5.481×10^{-2}	0.503	3.003×10^{-3}
2.20	0.983	6.908×10^{-2}	0.664	3.947×10^{-2}	0.709	2.342×10^{-1}
2.40	0.992	6.046×10^{-2}	0.662	4.166×10^{-2}	0.546	5.705×10^{-2}
2.60	0.986	1.494×10^{-1}	0.726	8.770×10^{-3}	0.739	1.862×10^{-1}
2.80	1.005	1.182×10^{-1}	0.695	3.508×10^{-2}	0.677	5.705×10^{-2}
3.00	1.002	2.385×10^{-1}	0.642	4.385×10^{-2}	0.586	6.606×10^{-2}
3.20	1.014	5.478×10^{-2}	0.650	4.604×10^{-2}	0.571	6.305×10^{-2}
3.40	0.977	4.731×10^{-2}	0.653	5.043×10^{-2}	0.550	3.003×10^{-3}
3.60	0.956	7.469×10^{-3}	0.614	2.631×10^{-2}	0.527	3.603×10^{-2}
3.80	0.849	4.980×10^{-3}	0.524	4.385×10^{-3}	0.524	9.308×10^{-2}
4.00	0.172	3.935×10^{-3}	0.305	2.193×10^{-3}	0.452	3.903×10^{-2}
4.10	0.060	6.553×10^{-3}	0.113	1.535×10^{-2}	0.242	3.003×10^{-2}
4.20	0.020	5.124×10^{-3}	0.042	2.412×10^{-3}	0.066	3.903×10^{-3}
4.30	0.012	2.115×10^{-3}	0.016	2.850×10^{-3}	0.060	2.072×10^{-2}
4.40	0.007	2.113×10^{-3}	0.012	2.412×10^{-3}	0.045	6.005×10^{-3}
4.50	0.005	4.482×10^{-4}	0.008	6.178×10^{-4}	0.016	1.462×10^{-3}
4.60	0.004	2.490×10^{-5}	0.007	1.507×10^{-3}	0.012	1.100×10^{-3}
4.70	0.003	4.980×10^{-4}	0.007	1.311×10^{-3}	0.009	2.102×10^{-4}
4.80	0.003	1.569×10^{-3}	0.007	8.406×10^{-4}	0.007	2.102×10^{-4}
4.90	0.002	1.494×10^{-4}	0.005	8.963×10^{-4}	0.007	9.308×10^{-4}
5.00	0.002	0.000	0.006	6.358×10^{-4}	0.007	7.506×10^{-4}
5.20	0.002	2.988×10^{-4}	0.005	9.867×10^{-4}	0.005	3.003×10^{-5}
5.40	0.001	1.743×10^{-4}	0.005	1.096×10^{-4}	0.004	4.804×10^{-4}
5.60	0.001	1.245×10^{-4}	0.004	1.973×10^{-4}	0.002	9.008×10^{-5}
5.80	0.001	7.469×10^{-5}	0.003	2.631×10^{-4}	0.002	3.003×10^{-5}
6.00	0.002	6.474×10^{-4}	0.004	1.316×10^{-4}	0.003	1.501×10^{-4}
6.50	0.001	9.959×10^{-5}	0.003	1.316×10^{-4}	0.002	3.303×10^{-4}
7.00	0.001	2.490×10^{-5}	0.003	4.166×10^{-4}	0.001	2.702×10^{-4}
7.75	0.001	7.469×10^{-5}	0.002	1.754×10^{-4}	0.001	6.005×10^{-5}
8.00	0.000	0.000	0.000	0.000	0.000	0.000

Table S7. Experimental concentrations and associated estimated SD values (for N=3), normalized with the initial concentration (CFU/mL), for the cotransport experiment.

Cotransport						
E. coli/E. faecalis/S. aureus						
	E. coli		***E. faecalis***		***S. aureus***	
PV	$\mathbf{C_{b(t)}/C_{b0}}$	**SD normalized (N=3)**	$\mathbf{C_{b(t)}/C_{b0}}$	**SD normalized (N=3)**	$\mathbf{C_{b(t)}/C_{b0}}$	**SD normalized (N=3)**
0.00	0.000	0.000	0.000	0.000	0.000	0.000
0.20	0.000	0.000	0.000	0.000	0.000	0.000

0.40	0.003	1.176×10^{-4}	0.002	9.355×10^{-5}	0.002	4.163×10^{-4}
0.60	0.057	1.960×10^{-3}	0.039	4.678×10^{-4}	0.056	8.598×10^{-3}
0.70	0.118	5.880×10^{-3}	0.076	1.403×10^{-2}	0.072	1.662×10^{-2}
0.80	0.175	3.920×10^{-3}	0.104	2.183×10^{-2}	0.141	1.057×10^{-2}
1.00	0.477	4.704×10^{-2}	0.281	1.715×10^{-2}	0.406	1.493×10^{-1}
1.10	0.912	3.920×10^{-3}	0.479	2.183×10^{-2}	0.602	2.660×10^{-1}
1.20	1.058	1.960×10^{-3}	0.535	7.796×10^{-3}	1.056	7.976×10^{-2}
1.30	0.898	7.840×10^{-3}	0.529	6.237×10^{-3}	0.570	0.000
1.40	1.010	9.604×10^{-2}	0.719	1.871×10^{-2}	0.899	5.883×10^{-2}
1.50	1.123	7.448×10^{-2}	0.648	6.237×10^{-3}	1.099	2.956×10^{-2}
1.60	0.976	3.528×10^{-2}	0.678	5.457×10^{-2}	0.742	1.267×10^{-1}
1.70	0.851	7.840×10^{-2}	0.739	2.183×10^{-2}	0.707	5.883×10^{-2}
1.80	1.063	5.880×10^{-3}	0.657	5.613×10^{-2}	0.964	9.963×10^{-2}
1.90	1.074	4.900×10^{-2}	0.633	8.732×10^{-2}	0.522	3.168×10^{-2}
2.00	0.912	1.176×10^{-1}	0.718	1.559×10^{-3}	1.088	1.221×10^{-1}
2.20	1.028	7.056×10^{-2}	0.669	1.013×10^{-1}	0.858	1.448×10^{-1}
2.40	1.051	2.352×10^{-1}	0.660	7.952×10^{-2}	0.966	1.810×10^{-2}
2.60	0.966	1.274×10^{-1}	0.678	9.823×10^{-2}	0.637	1.584×10^{-1}
2.80	1.094	1.156×10^{-1}	0.678	1.715×10^{-2}	0.826	2.715×10^{-2}
3.00	1.109	4.704×10^{-2}	0.592	6.393×10^{-2}	0.666	3.620×10^{-2}
3.20	1.088	1.960×10^{-3}	0.684	6.237×10^{-3}	0.557	9.051×10^{-3}
3.40	0.933	1.235×10^{-1}	0.533	4.678×10^{-3}	0.531	3.620×10^{-2}
3.60	0.876	1.142×10^{-1}	0.703	7.172×10^{-2}	0.854	4.525×10^{-3}
3.80	0.653	2.097×10^{-1}	0.450	1.871×10^{-2}	0.333	7.241×10^{-2}
4.00	0.696	1.215×10^{-1}	0.460	1.091×10^{-2}	0.486	1.086×10^{-1}
4.10	0.524	3.528×10^{-2}	0.326	3.118×10^{-3}	0.256	7.241×10^{-2}
4.20	0.273	5.880×10^{-3}	0.163	1.559×10^{-2}	0.214	6.150×10^{-2}
4.30	0.116	1.568×10^{-2}	0.067	1.073×10^{-2}	0.103	0.000
4.40	0.073	9.801×10^{-3}	0.026	3.118×10^{-4}	0.049	5.431×10^{-3}
4.50	0.017	1.960×10^{-4}	0.018	3.592×10^{-3}	0.023	6.605×10^{-3}
4.60	0.010	1.372×10^{-3}	0.009	9.355×10^{-4}	0.009	3.168×10^{-4}
4.70	0.010	9.801×10^{-4}	0.011	4.678×10^{-4}	0.008	1.855×10^{-3}
4.80	0.005	1.960×10^{-4}	0.007	1.559×10^{-4}	0.005	3.168×10^{-3}
4.90	0.004	5.880×10^{-4}	0.007	1.247×10^{-3}	0.003	2.715×10^{-4}
5.00	0.003	1.372×10^{-3}	0.006	1.247×10^{-3}	0.010	2.217×10^{-3}
5.20	0.002	5.880×10^{-5}	0.005	1.091×10^{-3}	0.005	6.336×10^{-4}
5.40	0.001	3.724×10^{-4}	0.004	4.990×10^{-4}	0.002	4.525×10^{-4}
5.60	0.001	1.372×10^{-4}	0.003	2.330×10^{-4}	0.003	2.715×10^{-4}
5.80	0.001	5.292×10^{-4}	0.004	2.027×10^{-4}	0.002	4.514×10^{-4}
6.00	0.001	4.737×10^{-4}	0.002	9.602×10^{-4}	0.003	6.400×10^{-5}
6.50	0.001	1.764×10^{-4}	0.002	7.796×10^{-5}	0.002	9.535×10^{-4}
7.00	0.000	6.402×10^{-5}	0.002	6.698×10^{-4}	0.002	1.825×10^{-4}
7.50	0.001	4.704×10^{-4}	0.002	1.655×10^{-4}	0.001	7.093×10^{-4}
8.00	0.000	0.000	0.000	0.000	0.000	0.000