Supporting Information: Multimedia Modeling of Engineered Nanoparticles with SimpleBox4nano:

Model Definition and Evaluation

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Keywords: Multimedia Modeling Environmental Concentrations Engineered Nanoparticles

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**Table S1. Glossary of terms**

| Symbol                  | Description                                                                 | Value / Unit | Eq. Nr. | Ref. |
|-------------------------|-----------------------------------------------------------------------------|--------------|---------|------|
| \( A \)                | First-order removal and transport rate constant matrix                       | s\(^{-1}\)   | 1       | 1    |
| \( A_{ENP} \)           | ENP surface                                                                  | m\(^2\)      | -       | (-)  |
| \( A_{Hamaker(123)} \)  | Overall Hamaker constant for ENP (1), water (2), and grain collector (3)     | (-)          | -       | 2    |
| \( A_{Arrhenius} \)     | Arrhenius constant for Arrhenius equations                                  | (-)          | -       | 3    |
| \( A_s \)               | Analytical Smoluchowski-Levich solution                                      | (-)          | 35      | 4    |
| \( \hat{a}_{veg} \)     | Vegetation hair width                                                        | 1.0 \( \times \) 10\(^{-5}\) m | -       | 5    |
| \( \hat{A}_{veg} \)     | Vegetation large collection radii                                            | 5.0 \( \times \) 10\(^{-4}\) m | -       | 5    |
| \( c_{env,cond} \)      | Influence on dissolution rate by environmental conditions                    | (-)          | -       | 3    |
| \( C_0 \)               | Experiment’s initial concentration                                           | N.m\(^{-3}\) | -       | 1    |
| \( C_{c_i} \)           | Cunningham coefficient for atmospheric particle \( i \)                      | (-)          | 13      | 6    |
| \( \bar{v}_{thermal(i)} \) | Thermal velocity of particle \( i \) in air                               | m.s\(^{-1}\) | 15      | 7    |
| \( C(t) \)              | Experiment’s concentration at time \( t \)                                 | N.m\(^{-3}\) | -       | 1    |
| \( C_{v}/C_d \)         | Viscous versus total drag ratio                                              | 0.3          | -       | 5    |
| \( d_{Grain} \)         | Grain collector diameter                                                     | 0.256 mm     | -       | 8    |
| Symbol     | Description                                                                 | Unit     | Reference  |
|------------|-----------------------------------------------------------------------------|----------|------------|
| $D_{air(i)}$ | Diffusivity of particle $i$ in air                                           | m$^2$.s$^{-1}$ | 12, 9      |
| $D_i$      | Diffusivity of particle $i$ in water                                         | m$^2$.s$^{-1}$ | 41, 10     |
| $D_{SP}$   | Diffusivity of solution product                                              | m$^2$.s$^{-1}$ | - (-)      |
| $d_{ENP}$  | Diameter of the engineered nanoparticle                                      | m         | - (-)      |
| $d_{rain}$ | Representative diameter of a rain droplet                                   | m         | MA7$^*$ 11,12 |
| $e$        | Emission of ENPs to the environment                                          | g.s$^{-1}$ | 1, 1       |
| $e_{electron}$ | Elemental charge                                                              | 1.6 $10^{-19}$ V | - (-) |
| $E_{Ai}$   | Total raindrop collection efficiency for atmospheric particle $i$            | (-)      | 52, 13     |
| $E_{ABrown}$ | Raindrop collection efficiency for Brownian motion                          | (-)      | 53, 13     |
| $E_{Aintercept}$ | Raindrop collection efficiency for interception                            | (-)      | 54, 13     |
| $E_{Agrav}$ | Raindrop collection efficiency for gravitational impaction                   | (-)      | 55, 13     |
| $E_{dBrown(soil)}$ | Collection efficiency of Brownian motion to soil surfaces                   | (-)      | 67, 5      |
| $E_{dBrown(water)}$ | Collection efficiency of Brownian motion to surface water                  | (-)      | 64, 5      |
| $E_{d int.cept(soil)}$ | Collection efficiency of interception to soil                              | (-)      | 68, 5      |
| $E_{d int.cept(water)}$ | Collection efficiency of interception to surface water                    | (-)      | 65, 5      |
| $E_{dgrav(soil)}$ | Collection efficiency of inertial impaction to soil                        | (-)      | 69, 5      |
| $E_{dgrav(water)}$ | Collection efficiency of inertial impaction to surface water              | (-)      | 66, 5      |
| Symbol     | Description                                                                 | Unit/Value  |
|------------|-----------------------------------------------------------------------------|-------------|
| $E_{\text{activation}}$ | Required activation energy for dissolution                                  | J           |
| $f$         | Porous medium porosity                                                      | (-) 0.456   |
| $f_{\text{coag}(i,j)}$ | Polydisperse coagulation coefficient between atmospheric particle $i$ and $j$. | s$^{-1}$ 10  |
| $f_{\text{col}(i,j)}$   | Collision rate between ENPs and natural particles in aqueous media        | s$^{-1}$ 20  |
| $f_{\text{Brown}}$     | Collision frequency due to Brownian motion                                 | s$^{-1}$ 16  |
| $f_{\text{Intercept}}$ | Collision frequency due to interception                                    | s$^{-1}$ 17  |
| $f_{\text{grav}}$      | Collision frequency due to gravitational settling differences             | s$^{-1}$ 18  |
| $f_{\text{Brenner}(0,0,r/r+h)}$ | Brenner function for deposition to surfaces                                | (-) 45      |
| $g_1(H)$   | Universal hydrodynamic function                                             | (-) 45      |
| $G_{\text{shear}}$     | Surface water shear rate                                                    | 10 s$^{-1}$  |
| $h$        | Separation distance between particle $i$ and $j$                           | m           |
| $I$        | Ionic strength of the aqueous medium                                        | 1-10 $\cdot 10^{-3}$ M |
| $k_b$      | Boltzmann constant                                                          | 1.38 $\cdot 10^{-23}$ \text{J.K}^{-1} |
| $k_{\text{AARagg}}$   | Rain drop collection rate for aggregated ENP species                        | s$^{-1}$ 51  |
| $k_{\text{AARatt}}$   | Rain drop collection rate for attached ENP species                          | s$^{-1}$ 51  |
| Parameter | Description | Unit | Value | Notes |
|-----------|-------------|------|-------|-------|
| $k_{AA\ell r\text{free}}$ | Rain drop collection rate for free ENP species | s$^{-1}$ | 51 | (-) |
| $k_{aggA}$ | Aggregation rate for ENPs with aerosol particles in dry air (A) | s$^{-1}$ | 2 | (-) |
| $k_{aggS}$ | Aggregation rate for ENPs with natural colloids in soil (S) pore water | s$^{-1}$ | 6 | (-) |
| $k_{aggSE}$ | Aggregation rate for ENPs with natural colloids in sediment (SE) pore water | s$^{-1}$ | 8 | (-) |
| $k_{aggW}$ | Aggregation rate for ENPs with natural colloids in surface water (W) | s$^{-1}$ | 4 | (-) |
| $k_{attA}$ | Attachment rate for ENPs with coarse particles in dry air (A) | s$^{-1}$ | 3 | (-) |
| $k_{attS}$ | Attachment rate for ENPs with solid grains in soil (S) | s$^{-1}$ | 7 | (-) |
| $k_{attSE}$ | Attachment rate for ENPs with solid grains in sediment (SE) | s$^{-1}$ | 9 | (-) |
| $k_{attW}$ | Attachment rate for ENPs with suspended particles in surface water (W) | s$^{-1}$ | 5 | (-) |
| $k_{bur}$ | Burial of ENPs to deeper sediments | s$^{-1}$ | 76 | 14-16 |
| $k_{depA\text{Sagg}}$ | First-order rate constant for dry deposition from dry air (A) to soil (S) for aggregated ENP species | s$^{-1}$ | MA 6*1 | (-) |
| $k_{depA\text{Satt}}$ | First-order rate constant for dry deposition from dry air (A) to soil (S) for attached ENP species | s$^{-1}$ | MA 6*1 | (-) |
| $k_{depA\text{Sfree}}$ | First-order rate constant for dry deposition from dry air (A) to soil (S) for free ENP species | s$^{-1}$ | MA 6*1 | (-) |
| $k_{depAW\text{agg}}$ | First-order rate constant for dry deposition from dry air (A) to water (W) for aggregated ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depAW\text{att}}$ | First-order rate constant for dry deposition from dry air (A) to soil (W) for attached ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depAW\text{free}}$ | First-order rate constant for dry deposition from dry air (A) to soil (W) for free ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depRS\text{agg}}$ | First-order rate constant for wet deposition from rain (R) to soil (S) for aggregated ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depRS\text{att}}$ | First-order rate constant for wet deposition from rain (R) to soil (S) for attached ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depRS\text{free}}$ | First-order rate constant for wet deposition from rain (R) to soil (S) for free ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depRW\text{agg}}$ | First-order rate constant for wet deposition from rain (R) to water (W) for aggregated ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depRW\text{att}}$ | First-order rate constant for wet deposition from rain (R) to soil (W) for attached ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depRW\text{free}}$ | First-order rate constant for wet deposition from rain (R) to soil (W) for free ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depWSE\text{agg}}$ | First-order rate constant for deposition from water (W) to sediments (SE) for aggregated ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{depWSE\text{att}}$ | First-order rate constant for deposition from water (W) to sediments (SE) for attached ENP species | s$^{-1}$ | MA 6* | (-) |
| Parameter | Description                                                                 | Unit | Value 1 | Value 2 |
|-----------|-----------------------------------------------------------------------------|------|---------|---------|
| $k_{depWSE\text{free}}$ | First-order rate constant for deposition from water (W) to sediments (SE) for free ENP species | s$^{-1}$ | MA 6* | (-) |
| $k_{erosionSW\text{att}}$ | First-order rate constant for run-off from soil (S) to water (W) for attached ENP species | s$^{-1}$ | 74 | (-) |
| $k_{rsSE\text{Wagg}}$ | First-order rate constant for resuspension from sediments (SE) to water (W) for aggregated ENP species | s$^{-1}$ | 69 | 14-16 |
| $k_{rsSE\text{Watt}}$ | First-order rate constant for resuspension from sediments (SE) to water (W) for attached ENP species | s$^{-1}$ | 70 | 14-16 |
| $k_{rsSE\text{Wfree}}$ | First-order rate constant for resuspension from sediments (SE) to water (W) for free ENP species | s$^{-1}$ | 69 | 14-16 |
| $k_{run\text{Wagg}}$ | First-order rate constant for run-off from soil (S) to water (W) for aggregated ENP species | s$^{-1}$ | 73 | (-) |
| $k_{run\text{Wfree}}$ | First-order rate constant for run-off from soil (S) to water (W) for free ENP species | s$^{-1}$ | 73 | (-) |
| $Kn_i$ | Knudsen number of the atmospheric particle $i$ | (-) | 14 | 6 |
| $K_F$ | Pseudo-first-order rate constant for attachment efficiency | s$^{-1}$ | 44 | 2 |
| $m$ | Steady state ENP mass per compartment and species | g | 1 | 1 |
| $m_{as}$ | Mass of altered species particle | g | 47 | (-) |
| $m_{CP}$ | Mass of counter particle | g | - | (-) |
| $m_i$ | Individual mass particle $i$ | g | - | (-) |
| $m_{\text{ENP}}$ | Mass of the engineered nanoparticle | g | - | (-) |
| Symbol | Description                                                   | Unit          | Dimension |
|--------|---------------------------------------------------------------|---------------|-----------|
| M_{SP} | Molarity of solution product                                 | M             | (-)       |
| N_A    | Avogadro’s number                                            | 6.02 \times 10^{-23} mol^{-1} | (-)       |
| N_{acc}| Number concentration of Aitken accumulation mode aerosol particles | 2.9 \times 10^{9} N.m^{-3} | 20        |
| N_{coarse}| Number concentration of coarse mode aerosol particles | 3 \times 10^{5} N.m^{-3} | 20        |
| N_{NC(S)} | Number concentration of natural colloids in soil pore water | 1 \times 10^{11} N.m^{-3} | 21        |
| N_{NC(SE)} | Number concentration of natural colloids in sediment pore water | 1 \times 10^{11} N.m^{-3} | 21        |
| N_{NC(W)} | Number concentration of natural colloids in surface water | 1 \times 10^{11} N.m^{-3} | 21        |
| N_{nuc} | Number concentration of nucleation mode aerosol particles    | 3.2 \times 10^{9} N.m^{-3} | 20        |
| N_{SP(W)} | Number concentration of suspended particles (>450 nm) in surface water | N.m^{-3} | 21        |
| N_R    | Aspect ratio number                                           | (-)           | 37 4      |
| N_{PE} | Peclet Number                                                | (-)           | 38 4      |
| N_{VDW} | Van der Waals attraction number                              | (-)           | 39 4      |
| N_G    | Gravity number                                               | (-)           | 40 4      |
| p_0    | Precipitation rate                                           | 2.22 m.s^{-1} | 10-8 14-16 |
| r      | Radius                                                       | m             | (-)       |
| Symbol | Description                                      | Unit       | Value         | Comment |
|--------|--------------------------------------------------|------------|---------------|---------|
| $r_{NC}$ | Radius of natural colloids in water           | nm         | 300           | 21      |
| $r_{NLP}$ | Radius of natural larger particles in water   | cm         | 2.8 $10^5$    | 14-16   |
| $r_{as}$  | Radius of individual altered species particle | m          | 50            |         |
| R       | Ideal gas constant                             |            | 8.314 J.K$^{-1}$.mol$^{-1}$ |         |
| $R_A$   | Aerodynamic resistance                         | s.m$^{-1}$ | 33            | 5       |
| $r_{rain}$ | Rain drop radius                              | m          | MA 7           | 11,12   |
| $R_{S(i)}$ | Surface resistance for atmospheric particle $i$ | s.m$^{-1}$ | -             | 5       |
| Re      | Reynolds number                                 | (-)        | 56            | 13      |
| S(β)    | Function of Spielman & Friedlander for surface collisions | (-)        | -             | 22      |
| $Sc_i$  | Schmidt number of atmospheric particle $i$     | (-)        | -             | 13      |
| $[SP_{aq(0-δD)}]$ | Solution product concentration within diffusion boundary layer | mg.l$^{-1}$ | (-)         |         |
| $[SP_{bg}]$ | Background concentration of solution product | mg.l$^{-1}$ | -             | (-)     |
| St*     | Critical Stokes Number                         | (-)        | 61            | 13      |
| $St_i$  | Stokes number of atmospheric particle $i$      | (-)        | 59            | 13      |
| t       | Time                                            | s          | -             | (-)     |
| T       | Temperature                                     | K          | -             | (-)     |
| Symbol | Description | Value |
|--------|-------------|-------|
| $T_{air}$ | Air temperature | 285 K |
| $u_*$ | Friction velocity | 0.19 m.s$^{-1}$ |
| $U_{Darcy}$ | Darcy approach velocity | 9 $\times$ 10$^6$ m.s$^{-1}$ |
| $v_{dep(i)}$ | Dry deposition velocity atmospheric particle $i$ | m.s$^{-1}$ |
| $v_{set(i)}$ | Gravitational settling velocity of suspended particle $i$ | m.s$^{-1}$ |
| $v_{term(i)}$ | Terminal velocity of atmospheric particle $i$ | m.s$^{-1}$ |
| $v_{term(rain)}$ | Representative terminal velocity of a rain droplet | m.s$^{-1}$ |
| $V_{CP}$ | Volume of individual counter particle | m$^3$ |
| $V_{ENP}$ | Volume of individual ENP | m$^3$ |
| $V_{as}$ | Volume of individual altered species particle | m$^3$ |
| $V_{EDL(i,j)}$ | Electric double layer repulsive energy between particle $i$ and $j$ | J |
| $V_{max(i,j)}$ | Maximum interaction energy barrier between particle $i$ and $j$ | J |
| $V_{VDW(i,j)}$ | Van der Waals attractive energy between particle $i$ and $j$ | J |
| $V_T$ | Total energy barrier between particle $i$ an $j$ | J |
| $z$ | Counter ion valence | e.g. 4 |
| $\alpha_{agg}$ | Aggregation efficiency between ENP and natural colloid (<450 nm) | (%) |
| $\alpha_{att}$ | Attachment efficiency between ENP and large particle (>450 nm) | (%) |
| Symbol | Description                                                                 | Value          | Unit | Significance |
|--------|------------------------------------------------------------------------------|----------------|------|--------------|
| $\alpha_{CC}$ | Empirical constant for deriving Cunningham coefficient | 1.142          |      |              |
| $\beta_{CC}$ | Empirical constant for deriving Cunningham coefficient | 0.558          |      |              |
| $\beta_{IFBL}$ | Interaction force boundary layer approximation parameter | (-)            | 43   | 2            |
| $\beta_{i,j}$ | Transitional correction coefficient for coagulation between atmospheric particle $i$ and $j$. | (-)            | 11   | 9            |
| $\gamma_{CC}$ | Empirical constant for deriving Cunningham coefficient | 0.999          |      |              |
| $\gamma_\psi$ | Porosity dependency parameter | (-)            | 36   | 4            |
| $\psi_i$ | Surface potential of particle $i$ | V              |      | (-)          |
| $\psi_{NP}$ | Surface potential of natural particle | 55 mV          |      | 25           |
| $\varepsilon_0$ | Dielectric permittivity in vacuum | $8.854 \times 10^{-12}$ F.m$^{-1}$ |      | (-)          |
| $\varepsilon_r$ | Relative dielectric permittivity of water | 78.5 at 25°C |      | (-)          |
| $\eta_0$ | Total collection efficiency for deposition of ENPs onto solid grains in porous media | (-)            | 32   | 4            |
| $\eta_{Brown}$ | Collection efficiency for deposition onto solid grains by Brownian motion | (-)            | 33   | 4            |
| $\eta_{Intercept}$ | Collection efficiency for deposition onto solid grains by interception | (-)            | 34   | 4            |
| $\eta_{rav}$ | Collection efficiency for deposition onto solid grains by $rav$ | (-)            | 35   | 4            |
| Characteristic | Value                            | Unit   | Remarks |
|---------------|----------------------------------|--------|---------|
| $\eta_{\text{air}}$ | Kinematic viscosity of air | $1.48 \times 10^{-5}$ J.m$^3$.s$^{-1}$ | - | (-) |
| $\kappa_{\text{Debye}}$ | Debye length | m | - | 26 |
| $\lambda_{\text{air}}$ | Mean free path in air | $66 \times 10^{-9}$ m | - | 27 |
| $\lambda_{cw}$ | Characteristic wave length | $\approx 100$ nm | - | 28 |
| $\lambda_{\text{filter}}$ | Filtration in porous media | m$^{-1}$ | 31 | 4 |
| $\lambda_i$ | Scavenging coefficient of atmospheric particle $i$ | s$^{-1}$ | 51 | 13 |
| $\Gamma_i$ | Dimensionless surface potential of particle $i$ | (-) | 27 | 2 |
| $\rho_{as}$ | Density of individual altered species particle | kg.m$^{-3}$ | 49 | (-) |
| $\rho_{\text{aerosol}}$ | Assumed density for nucleation, accumulation and coarse mode aerosols | $1.371 \times 10^5$ g.m$^{-3}$ | - | 29 |
| $\rho_{\text{air}}$ | Density of air | $1.225$ kg.m$^{-3}$ | - | (-) |
| $\rho_{\text{ENP}}$ | Density of ENP | kg.m$^{-3}$ | - | (-) |
| $\rho_l$ | Density of particle $i$ | kg.m$^{-3}$ | - | (-) |
| $\rho_{\text{water}}$ | Density of water | $998$ kg.m$^{-3}$ | - | (-) |
| $\delta_D$ | Diffusion boundary layer thickness | m | 23 | 26 |
| \( \tau_{air(i)} \) | Relaxation of atmospheric particle \( i \) in air | s | 60 | 13 |
|-----------------|------------------------------------------|----|----|----|
| \( \mu_{air} \) | Dynamic viscosity of air | \( 1.81 \times 10^{-5} \) J.m\(^3\).s\(^{-1}\) | - | (-) |
| \( \mu_{water} \) | Dynamic viscosity of water | \( 8.9 \times 10^{-4} \) Pa.s | - | (-) |

*1 MA= Main Article
Mass balance equations expressed as matrix algebra

\[ m = -A^{-1} \cdot e \]  
(Eq. S1)

The mass balance equations of SB4N, with \( m \) is steady state ENP mass (g) per compartment and species, emission \( e \) to the environment (g.s\(^{-1}\)) and transport and removal rates (s\(^{-1}\)) expressed in matrix \( A \).
**Table S2.** First-order rate constants for total removal ($\Sigma k_r$) per compartment and species of ENP

| Species in compartment | Total sum of removal rates per species in compartment |
|------------------------|-------------------------------------------------------|
| Free in dry air:       | $\Sigma k_r^\text{free} = k_{agg} + k_{att} + k_{\text{Attr}} + k_{\text{depASfree}} + k_{\text{depAWfree}}$ |
| Agg. in dry air:       | $\Sigma k_r^\text{agg} = k_{\text{Agg}} + k_{\text{depASagg}} + k_{\text{depAWagg}}$ |
| Att. in dry air:       | $\Sigma k_r^\text{att} = k_{\text{Attr}} + k_{\text{depASatt}} + k_{\text{depAWatt}}$ |
| Free in rain:          | $\Sigma k_r^\text{free} = k_{\text{depRSfree}} + k_{\text{depRWfree}} + k_{\text{dissolveRfree}}$ |
| Agg. in rain:          | $\Sigma k_r^\text{agg} = k_{\text{depRSagg}} + k_{\text{depRWagg}} + k_{\text{dissolveRagg}}$ |
| Att. in rain:          | $\Sigma k_r^\text{att} = k_{\text{depRSatt}} + k_{\text{depRWatt}} + k_{\text{dissolveRatt}}$ |
| Free in soil pore water: | $\Sigma k_r^\text{free} = k_{\text{ags}} + k_{\text{atts}} + k_{\text{leachSfree}} + k_{\text{runSWfree}} + k_{\text{dissolveSfree}}$ |
| Agg. in soil pore water: | $\Sigma k_r^\text{agg} = k_{\text{leachSagg}} + k_{\text{runSWagg}} + k_{\text{dissolveSagg}}$ |
| Att. to soil grains:   | $\Sigma k_r^\text{att} = k_{\text{erosionSWatt}} + k_{\text{dissolveSatt}}$ |
| Free in water:         | $\Sigma k_r^\text{free} = k_{\text{aggW}} + k_{\text{attW}} + k_{\text{depWSEfree}} + k_{\text{dissolveWfree}}$ |
| Agg. in water:         | $\Sigma k_r^\text{agg} = k_{\text{depWSEagg}} + k_{\text{dissolveWagg}}$ |
| Att. in water:         | $\Sigma k_r^\text{att} = k_{\text{depWSEatt}} + k_{\text{dissolveWatt}}$ |
| Free in sediment pore water: | $\Sigma k_r^\text{free} = k_{\text{aggS}} + k_{\text{atts}} + k_{\text{burSEfree}} + k_{\text{rsSEWfree}} + k_{\text{dissolveSEfree}}$ |
| Agg. in sediment pore water: | $\Sigma k_r^\text{agg} = k_{\text{burSEagg}} + k_{\text{rsSEWagg}} + k_{\text{dissolveSEagg}}$ |
| Att. to sediment grains: | $\Sigma k_r^\text{att} = k_{\text{burSEatt}} + k_{\text{rsSEWatt}} + k_{\text{dissolveSEatt}}$ |
Table S3. Derivation of aggregation and attachment rates for compartments atmosphere (A), water (W), soil (S) and sediment (SE)

| Compartments | Equations for attachment and aggregation rate |
|--------------|-----------------------------------------------|
| **Atmosphere** | 2. \( k_{aggA} = f_{coag}(\text{ENP,nuc}) \cdot \alpha_{\text{ENP,nuc}} \cdot N_{\text{nuc}} + f_{coag}(\text{ENP,acc}) \cdot \alpha_{\text{ENP,acc}} \cdot N_{\text{acc}} \)  
|              | 3. \( k_{attA} = f_{coag}(\text{ENP,coarse}) \cdot \alpha_{\text{ENP,coarse}} \cdot N_{\text{coarse}} \) |
| **Surface water** | 4. \( k_{aggW} = f_{col}(\text{ENP,NC}) \cdot \alpha_{\text{agg(ENP,NC)}} \cdot N_{\text{NC(W)}} \)  
|              | 5. \( k_{attW} = f_{col}(\text{ENP,NLP}) \cdot \alpha_{\text{att(ENP,SP)}} \cdot N_{\text{SP(W)}} \) |
| **Soil** | 6. \( k_{aggS} = f_{col}(\text{ENP,NC}) \cdot \alpha_{\text{agg(ENP,NC)}} \cdot N_{\text{NC(S)}} \)  
|              | 7. \( k_{atts} = \lambda_{\text{filter}} \cdot \alpha_{\text{att(ENP,grain)}} \cdot \eta_{0(\text{ENP,grain})} \cdot U_{\text{Darcy}} \) |
| **Sediment** | 8. \( k_{aggSE} = f_{col}(\text{ENP,NC}) \cdot \alpha_{\text{agg(ENP,NC)}} \cdot N_{\text{NC(SE)}} \)  
|              | 9. \( k_{attSE} = \lambda_{\text{filter}} \cdot \alpha_{\text{att(ENP,grain)}} \cdot \eta_{0(\text{ENP,grain})} \cdot U_{\text{Darcy}} \) |
Table S4. Deriving coagulation rates between ENPs and natural aerosol particles\textsuperscript{7,9}

| Parameter                          | Equation                                                                 |
|------------------------------------|--------------------------------------------------------------------------|
| Coagulation coefficient            | $f_{\text{coag}(i,j)} = 4\pi (\eta_i + \eta_j) (D_{\text{air}(i)} + D_{\text{air}(j)})$ |
| Transitional correction coefficient | $\alpha_{(i,j)} = \left( 1 + \frac{4(D_{\text{air}(i)} + D_{\text{air}(j)})}{(\eta_i + \eta_j) \sqrt{\bar{c}_{\text{thermal}(i)}^2 + \bar{c}_{\text{thermal}(j)}^2}} \right)^{-1}$ |
| Particle diffusivity in air        | $D_{\text{air}(i)} = \frac{(k_b T_{\text{air}} C_{Cl})}{(6\pi \eta_{\text{air}} \eta_i)}$ |
| Particle Cunningham coefficient    | $C_{Cl} = 1 + Kn_i (\alpha_{CC} + \beta_{CC} \cdot e^{\frac{\gamma_{CC}}{Kn_i}})$ |
| Particle Knudsen number            | $Kn_i = \lambda_{\text{air}} / r_i$                                     |
| Particle thermal velocity          | $\bar{c}_{\text{thermal}(i)} = \frac{(8k_b T_{\text{air}})}{\sqrt{\pi m_i}}$ |

Table S5. Characterization of background concentrations of natural aerosol particles\textsuperscript{*1,20}

| Mode         | Diameter (nm) | Size standard deviation | Number concentration (N.m\textsuperscript{-3}) |
|--------------|---------------|-------------------------|-----------------------------------------------|
| Nucleation   | 20            | 0.245                   | 3.2 \textsuperscript{10}                     |
| Accumulation | 116           | 0.217                   | 2.9 \textsuperscript{10}                     |
| Coarse       | 1800          | 0.380                   | 3 \textsuperscript{10}                       |

*1. The density of an aerosol particle itself is characterized as 1.37 \textsuperscript{10}\textsuperscript{2} kg.m\textsuperscript{-3} based on the average chemical composition of measured aerosols\textsuperscript{29}
**Table S6.** Collision rates between ENPs with natural colloids (<450 nm) and larger suspended particles in water.\(^3\)

| Collision mechanism       | Equation                                                                 |
|---------------------------|--------------------------------------------------------------------------|
| Brownian motion           | \( f_{\text{Brown}(i,j)} = \frac{2k_bT}{3\mu_{\text{water}}} \frac{(r_i + r_j)^2}{r_i \cdot r_j} \) |
| Interception              | \( f_{\text{Intercept}(i,j)} = \frac{4}{3} G_{\text{shear}} (r_i + r_j)^3 \) |
| Differential settling     | \( f_{\text{grav}}(i,j) = \pi (r_i + r_j)^2 \cdot |v_{\text{set}(i)} - v_{\text{set}(j)}| \) |
| Gravitational settling velocity | \( v_{\text{set}(i)} = \frac{2 (\rho_i - \rho_{\text{water}}) g r_i^2}{9 \mu_{\text{water}}} \) |
| Total collision frequency coefficient | \( f_{\text{col}}(i,j) = f_{\text{Brown}(i,j)} + f_{\text{Intercept}(i,j)} + f_{\text{grav}}(i,j) \) |
| Collision rate            | \( k_{\text{col}(i,j)} = f_{\text{col}}(i,j) \cdot N_j \) |

**Table S7.** Experimentally derived efficiencies for hetero-aggregation (\(\alpha_{\text{agg}}\)) between ENPs and natural colloidal particles in different water types. Original data from Quik, J.T.K., Ph.D. Dissertation, Radboud University Nijmegen, The Netherlands, 2013.\(^3\)

| Sample Site (The Netherlands) | Water type | Nano-C60 | Nano-CeO2 | Nano-SiO2-Ag | Nano-PVP-Ag |
|------------------------------|------------|----------|-----------|--------------|-------------|
| Karregat Pool                | n.a.       | 0.161    | n.a.      | 0.692        |             |
| Brabantse Aa Brook           | 6.75E-03   | n.a.     | 0.222     | n.a.         |             |
| River Rhine River            | n.a.       | 0.996    | 0.444     | 0.292        |             |
| IJsselmeer Lake              | n.a.       | 0.121    | 0.252     | 0.102        |             |
| Nieuwe Waterweg Canal        | 0.231      | 0.854    | 0.603     | 0.678        |             |
| North Sea Sea                | 1          | 1        | 1         | 1            |             |
Figure S1. Aggregation efficiency as a function of electrophoretic mobility (EPM). Original data from Keller et al., 2010. Environmental Science & Technology, 2010, 44, 1962-1967.
Table. S8 Electrophoretic mobility (EPM) in $\mu\text{ms}^{-1}\text{V}^{-1}\text{cm}$ given for different ENPs in different water types. Original data from Keller et al., 2010. Environmental Science & Technology, 2010, 44, 1962-1967. 32

| Water Type       | nano-TiO2 | nano-CeO2 | nano-ZnO |
|------------------|-----------|-----------|----------|
| Natural Seawater | -0.04±0.09 | -0.05±0.24 | -0.04±0.26 |
| Artificial Seawater | -0.05±0.15 | -0.04±0.09 | -0.27±0.52 |
| Lagoon           | -0.82±0.09 | -0.75±0.16 | -1.54±0.17 |
| Groundwater      | -1.11±0.03 | -1.05±0.05 | -1.21±0.03 |
| River            | -1.24±0.13 | -1.13±0.11 | -1.63±0.07 |
| Treated Effluent | -1.39±0.05 | -1.36±0.05 | -1.13±0.04 |
| Mesocosm Effluent | -1.89±0.04 | -1.91±0.05 | -1.75±0.04 |
| Storm Water      | -2.09±0.05 | -2.01±0.05 | -1.69±0.07 |
| Mesocosm freshwater | -2.4±0.05 | -2.39±0.09 | -2.22±0.06 |
Table S9. Theoretical derivation of aggregation efficiencies between ENPs and natural colloids*1 2

| Parameter                          | Equation                                                                 |
|-----------------------------------|--------------------------------------------------------------------------|
| Aggregation efficiency            | $\alpha_{agg} \approx \kappa_{Debye} 2r_i e^{-\frac{V_{\max(i,j)}}{k_b T}}$ |
| Diffusion boundary layer          | $\sigma_D = \kappa_{Debye}^{-1} = \left(\sqrt{\left(\varepsilon_r \varepsilon_0 k_b T \right)/(2N_A e^2 I)}\right)^{-1}$ |
| Interaction energy                | $V_{\max(i,j)} = \max(V_{EDL(h,i,j)} + V_{VDW(h,i,j)})$                   |
| Attractive Van der Waals Energy   | $V_{VDW(h,i,j)} = -A_{Hamaker(i,water,j)} \frac{r_i r_j}{6h(r_i + r_j)(1 + 14h/\lambda_{cw})}$ |
| Repulsive Electric Double Layer Energy | $V_{EDL(h,i,j)} = 64\pi \varepsilon_0 \varepsilon_r \frac{r_i r_j}{r_i + r_j} \left(\frac{k_b T}{z \cdot e_{electron}}\right)^2 \Gamma_i \Gamma_j \exp(-\kappa \cdot h)$ |
| Dimensionless surface potential   | $\Gamma_i = \tanh \left[\frac{z e_{electron} \Psi(i)}{4k_b T} \right]$       |

*1 According to the DLVO theory, the interaction energy between suspended colloidal particles can be evaluated as the sum of the attractive van der Waals ($V_{VDW}$) and the repulsive electrical double-layer ($V_{EDL}$) energies. The resultant interaction energy ($V_T$) determines the work that is necessary for the colloids to stick to each other as they must overcome the repulsive energy between them.35 In case the ENP and natural colloid are of about equal size a simple approximation ($V_T \approx V_{\max}$) for the aggregation efficiency for the two colliding colloids can be applied.34 The aggregation efficiency ($\alpha_{agg}$) is ultimately derived as a function of the ionic strength of the surrounding water ($I$), the radii ($r_{ENP}$, $r_{NC}$), Hamaker constants ($A_{Hamaker(ENP,water,NC)}$), and surface potentials ($\Psi_{ENP}$, $\Psi_{NC}$) of the ENPs and the natural colloids (NC).²
Table S10. Experimentally derived efficiencies for attachment of ENPs to suspended particulates in different water types ($\alpha_{\text{attW}}$). Original data from Quik, J.T.K., Ph.D. Dissertation, Radboud University Nijmegen, The Netherlands, 2013.\textsuperscript{31}

| Sample Site (The Netherlands) | Water type | Nano-CeO\textsubscript{2} | Nano-SiO\textsubscript{2}-Ag | Nano-PVP-Ag |
|-------------------------------|------------|----------------|----------------|--------------|
| River Rhine                   | River      | 0.96           | 0.98           | 0.82         |
| Nieuwe Waterweg Canal         | Canal      | 1              | 1              | 1            |
| North Sea                     | Sea        | 0.85           | 0.93           | 0.88         |

Table S11. Theoretical derivation of attachment efficiencies between ENPs and suspended particles (>450 nm) in surface waters \textsuperscript{*1}.\textsuperscript{35,36}

| Equations                                      |
|-----------------------------------------------|
| Attachment efficiency                         |
| 28 $\alpha_{\text{att(i,j)}} = \left( \int_{\delta_D} V_{\text{VDW(i,j)}} + V_{\text{EDL(i,j)}} \right) d_h / (k_B \cdot T)^{-1}$ |
| Van der Waals energy                          |
| 29 $V_{\text{VDW}} = - \frac{A_{\text{Hamaker(i,water,j)}} \cdot r_i}{12\hbar}$ |
| Electric double layer energy                  |
| 30 $V_{\text{EDL}} = 2\pi \varepsilon_0 \varepsilon_r \Gamma_i \Gamma_j e^{-kh}$ |

\textsuperscript{*1} It is assumed that the interaction between ENPs and larger natural particles can be approached as an interaction between a nanoparticle and a surface, because of their relatively large difference in size (<100 nm versus 450 nm).
Table S12. Application of the particle filtration theory\(^4\)

| Equation |  |
| --- | --- |
| **Filtration** | 31 \(\lambda_{filter} = \frac{3 \ (1 - f)}{2 \ d_{grain} \ f} \) |
| **Total collection efficiency** | 32 \(\eta_{0(i,j)} = \eta_{Brown} + \eta_{Intercept} + \eta_{grav} \) |
| **Brownian collection** | 33 \(\eta_{Brown} = 2.4A_s \frac{1}{3} N_R^{-0.081} N_{Pe}^{-0.715} N_{VDW}^{0.053} \) |
| **Collection by interception** | 34 \(\eta_{Intercept} = 0.55N_R^{-1.55} N_{Pe}^{-0.125} N_{VDW}^{0.125} \) |
| **Gravitational collection** | 35 \(\eta_{grav} = 0.22N_R^{-0.24} N_G^{1.11} N_{VDW}^{0.053} \) |
| **Porosity dependent parameter** | 36 \(A_s = \frac{2(1 - \gamma^5)}{2 - 3\gamma + 3\gamma^5 - 2\gamma^6}, \text{ with } \gamma = (1 - f)^{1/3} \) |
| **Aspect ratio Number** | 37 \(N_R = \frac{r_i}{r_j} \) |
| **Peclet Number** | 38 \(N_{Pe} = \frac{U_{Darcy} \cdot d_j}{D_i} \) |
| **Van der Waals Number** | 39 \(N_{VDW} = \frac{A_{Hamaker(i,water,j)}}{k_b \cdot T} \) |
| **Gravity Number** | 40 \(N_G = \frac{2r_i^2 (\rho_i - \rho_{water}) g}{9\mu_{water}U_{Darcy}} \) |
| **Diffusivity of particle \(i\) in water** | 41 \(D_i = \frac{k_b T}{6\pi \mu_{water} r_i} \) |
Table S13. Experimentally derived attachment efficiencies between engineered Cu\(^0\) nanoparticles and the solid grains of a saturated quartz column. Original data from Jones & Su, Water Research. 2012, 46, 2445–2456.\(^8\)

| Test | Sample Description                | Attachment Rate (\(k_{att}\)) (s\(^{-1}\)) | Attachment Efficiency (\(\alpha_{att}\)) |
|------|-----------------------------------|---------------------------------------------|------------------------------------------|
| 1    | Deionised water + 1 mM ph7 Trizma | 0.00068                                     | 0.1146                                   |
| 2    | Deionised water + 1 mM ph9.1 Trizma | 0.00011                                     | 0.0191                                   |
| 3    | Deionised I water + 1 mM ph9.1 Trizma | 0.00014                                     | 0.0237                                   |
| 4    | Deionised water + 1 mg/L Humic Acid | 0.00059                                     | 0.0996                                   |
| 5    | Deionised water + 1 mg/L Humic Acid | 0.00044                                     | 0.073                                    |
| 6    | Deionised water + 1 mg/L Humic Acid | 0.00048                                     | 0.798                                    |
| 7    | Deionised water + 5 mg/L Humic Acid | 0.00059                                     | 0.0987                                   |
| 8    | Deionised water + 10 mg/L Humic Acid | 0.00036                                     | 0.0601                                   |
| 9    | Deionised water + 1 mg/L Fulvic Acid | 0.00039                                     | 0.0645                                   |
| 10   | Deionised water + 1 mg/L Fulvic Acid | 0.00039                                     | 0.0645                                   |
**Table S14.** Theoretical derivation of attachment efficiencies between ENPs and the solid grains in porous media using the interaction force boundary layer (IFBL) approximation*1 22,37

| Equation | Attachment efficiency |
|----------|-----------------------|
| 42       | $\alpha_{att(i,j)} = \left( \frac{\beta_{IFBL(i,j)}}{1 + \beta_{IFBL(i,j)}} \right) S_{\beta(i,j)}$ |

| IFBL function | 43 |
|---------------|----|
| $\beta_{IFBL(i,j)} = \frac{1}{3} (2)^{1/3} \Gamma \left( \frac{1}{3} \right) A_s^{-1/3} \left( D_l \frac{U_{Darcy} \tau_j}{U_{Darcy} \tau_j} \right)^{1/3} \left( \frac{K\tau(i,j) \tau_j}{D_l} \right)$ |

| Uncorrected pseudo-first rate attachment rate constant | 44 |
|------------------------------------------------------|----|
| $K_F(i,j) = D_l \left\{ \int_0^{\delta_D} [g_1(H) \exp(V_{Total(i,j)}/kBT) - 1] dh \right\}^{-1}$ |

| Brenner function | 45 |
|------------------|----|
| $g_1(H) = f_{Brenner(0,0,\frac{r}{\kappa+\tau})}^{-1}$ |
| $= \left( 1 + \frac{3}{4} \left( \frac{r_l}{h + r_l} \right) + 0 \left( \frac{r_l}{h + r_l} \right)^2 \right)^{-1}$ |

| Total interaction energy | 46 |
|--------------------------|----|
| $V_{Total(h,i,j)} = V_{VDW(h,i,j)} + V_{EDL(h,i,j)}$ |

*1 With the IFBL approximation, attachment efficiencies can be derived from the radius ($r$), density ($\rho$), surface potential ($\Psi$) and Hamaker constant ($A_{Hamaker(ENP,water,grain)}$) of the ENP, and the radius of the grain ($r_{grain}$), porosity ($\iota$), and Darcy velocity ($U_{Darcy}$) of the porous medium, see SI Table 8 [2].

**Table S15.** Characterization of size and mass of altered species of ENPs (aggregated*1 or attached*1)

| Altered species property | Equation |
|--------------------------|----------|
| Mass                     | 47       |
| $m_{as} = m_{ENP} + m_{CP}$ |
| Volume                   | 48       |
| $V_{as} = V_{ENP} + V_{CP}$ |
| Density                  | 49       |
| $\rho_{as} = m_{as}/V_{as}$ |
| Radius                   | 50       |
| $r_{as} = \left( V_{as}/(4/3 \pi) \right)^{1/3}$ |

*1 Further characterization of the Hamaker constant and surface potential of the altered species is not necessary.
Table S16. Scavenging coefficients as function of particle size and density\textsuperscript{13}

| Equation | Scavenging coefficient 51 |
|----------|--------------------------|
| \( k_{Al} = \Lambda_i = \frac{3}{2} \frac{E_{Al}p_0}{d_{rain}} \) |

| Equation | Collection coefficient 52 |
|----------|--------------------------|
| \( E_{Al} = E_{ABrown}^{(i)} + E_{AIntercept}^{(i)} + E_{Agrav}^{(i)} \) |

| Equation | Brownian collection 53 |
|----------|------------------------|
| \( E_{ABrown}^{(i)} = \frac{4}{Re} \left[ 1 + 0.4Re^{1/2}Sc_l^{1/3} + 0.16Re^{1/2}Sc_l^{1/2} \right] \) |

| Equation | Collection by interception 54 |
|----------|-------------------------------|
| \( E_{AIntercept}^{(i)} = \frac{r_i}{r_{rain}} \left[ \frac{\mu_{air}}{\mu_{water}} + \left( 1 + 2Re^{1/2} \frac{d_{ENP}}{d_{rain}} \right) \right] \) |

| Equation | Collection by gravitational impaction 55 |
|----------|------------------------------------------|
| \( E_{Agrav}^{(i)} = \left( \frac{St_i - St^*}{St_i - St^* + 2/3} \right)^{3/2} \) if \( St_i > St^* \) |

| Equation | Reynolds Number 56 |
|----------|-------------------|
| \( Re = \frac{d_{rain} \cdot \nu_{term(rain)} \cdot \rho_{air}}{2 \cdot \mu_{air}} \) |

| Equation | Particle terminal velocity 57 |
|----------|-----------------------------|
| \( \nu_{term(i)} = d_i \left( \frac{\rho_i - \rho_{air}}{g \cdot C c_i} \right) \) |

| Equation | Particle Schmidt Number 58 |
|----------|---------------------------|
| \( Sc_l = \frac{\mu_{air}}{\rho_{air} \cdot D_{air}^{(i)}} \) |

| Equation | Particle Stokes Number 59 |
|----------|---------------------------|
| \( St_i = \frac{2\tau_{air}^{(i)} \left( \nu_{term(rain)} - \nu_{term(i)} \right)}{d_{rain}} \) |

| Equation | Particle relaxation time 60 |
|----------|-----------------------------|
| \( \tau_{air}^{(i)} = \frac{\left( \rho_i - \rho_{air} \right) d_i^{2} C c_i}{18 \mu_{air}} \) |

| Equation | Critical Stokes Number (\( E_{Agrav} = 0 \) if \( St^* > St_i \)) 61 |
|----------|----------------------------|
| \( St^* = \frac{1.2 + \frac{1}{12} \ln(1 + Re)}{1 + \ln(1 + Re)} \) |
Table S17. Dry deposition velocities as a function of particle size and density.\textsuperscript{5,7}

| Equation | Dry deposition velocity of particle $i$ to compartment 2 | Surface resistance | Brownian collection at deposition to water | Interception at deposition to water | Gravitational impaction at deposition to water | Brownian collection at deposition to soil | Interception at deposition to soil | Gravitational impaction at deposition to soil |
|----------|----------------------------------------------------------|--------------------|-------------------------------------------|---------------------------------|---------------------------------------------|------------------------------------------|---------------------------------|---------------------------------------------|
| 62       | $v_{\text{dep}(i,\text{dry air},2)} = \frac{1}{R_A(2) + R_S(i,2)} + v_{\text{terminal}(i)}$ | $R_S(i,2) = \frac{1}{u_* (E_{\text{dBrown(i,2)}} + E_{\text{d Intercept(i,2)}} + E_{\text{dgrav(i,2)}})}$ | $E_{\text{dBrown(i,water)}} = Sc_i^{-1/2}$ | $E_{\text{d Intercept(i,water)}} = 0$ | $E_{\text{dgrav(i,water)}} = 10^{-3St_i}$ | $E_{\text{dBrown(i,soil)}} = Sc_i^{-2/3}$ | $E_{\text{d Intercept(i,soil)}} = \frac{C_0}{C_d} \left[ f_{rv} \left\{ \frac{r_i}{r_i + A_{\text{veg}}} \right\} + (1 - f_{rv}) \left\{ \frac{r_i}{r_i + A_{\text{veg}}} \right\} \right]$ | $E_{\text{dgrav(i,soil)}} = \left( \frac{St_i}{St_i + A_{\text{veg}}} \right)^{\beta_{\text{veg}}}$ |
Table S18. ENP dissolution rates*1 for different mechanisms

| Mechanism                                      | Equation                                                                                                                                 |
|------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------|
| Noyes-Whitney for readily soluble ENPs         | $k_{\text{dis}(NW)} = A_{\text{ENP}} \frac{D_{SP}}{\delta_D} \left( [SP_{aq(0-\delta_D)}] - [SP_{bg}] \right) \cdot M_{SP}$ |
| Practically insoluble                          | $k_{\text{dis}(I)} = 0$                                                                                                               |
| Experimentally determined apparent dissolution rate | $k_{\text{dis}(\text{experiment})} = - \frac{\ln(C_0/C(t))}{t}$                                                                     |
| Dissolution following Arrhenius expression     | $k_{\text{dis}(\text{Arrhenius})} = c_{\text{env, cond}} \cdot A_{\text{Arrhenius}} \cdot e^{-E_{\text{Activation}}/RT}$               |

*1 The dissolution mechanism to be applied in SB4N is selected on the information that is available per case. Dissolution rates can also differ per aqueous medium and ENP species. SB4N easily deals with these possible differences, because its matrix A considers the environmental media as well as the free, aggregated, and attaches species of ENP. Different dissolution rates can be assigned per matrix cell and thus per environmental medium and ENP species.

Table S19. First-order rate constants for advective transports

| Mechanism                                      | Equation                                                                                                                                 |
|------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------|
| Resuspension of free or agg ENPs              | $k_{rsSEWfree,agg} = \frac{v_{rs} \cdot \text{AREA}_{W}}{VOLUME_{SE}} \cdot FR_{porewaterSE}$                                             |
| Resuspension of att. ENPs                     | $k_{rsSEWatt} = v_{rs} \cdot \text{AREA}_{W} \cdot FR_{solidsSE}$                                                                       |
| Sediment burial                               | $k_{bursE(i)} = v_{bur} \cdot \text{AREA}_{SE}/VOLUME_{W}$                                                                           |
| Pore water leaching of free or agg. ENPs      | $k_{\text{leachfree,agg}} = \frac{FR_{\text{infiltration}} \cdot p_0 \cdot \text{AREA}_{S}}{VOLUME_{SOILPOREWATER}}$          |
| Run-off of free or agg. ENPs                  | $k_{\text{runSWfree,agg}} = \frac{FR_{\text{runsoil}} \cdot p_0 \cdot \text{AREA}_{S}}{VOLUME_{SOILPOREWATER}}$            |
| Erosion of soil grains with ENPs attached     | $k_{\text{erosionSWatt}} = \frac{v_{\text{erosion}} \cdot \text{AREA}_{S}}{VOLUME_{SOILSOLIDS}}$                                   |
Table S20. System dimensions for realistic nano-TiO$_2$ emission scenario in Switzerland$^{39}$

| Compartment | Area (m$^2$) | Height / depth (m) | Volume (m$^3$) |
|-------------|-------------|--------------------|----------------|
| Atmosphere  | 4.13 $10^{10}$ | 1000              | 4.13 $10^{13}$ |
| Soil$^1$    | 4.00 $10^{10}$ | 0.2$^2$ and 0.05$^3$ | 2.23 $10^9$   |
| Water       | 1.24 $10^9$    | 3                  | 3.7 $10^9$     |

*1 Density of soil = 1500 kg. m$^{-3}$. Soil is composed of a solid fraction of 0.6, a pore water fraction of 0.2 and an air fraction of 0.2$^{14-16}$

Table S21. Input parameter and their values for the realistic nano-TiO$_2$ emission scenario in Switzerland$^{39}$

| Symbol | Parameter               | Value               |
|--------|-------------------------|---------------------|
| r$_{ENP}$ | ENP radius            | 10 nm              |
| $\rho_{ENP}$ | ENP mass density      | $4.23 \times 10^3$ kg.m$^{-3}$ |
| $\alpha_{agg}$ | Aggregation efficiency | 0.1$^1$          |
| $\alpha_{att}$  | Attachment efficiency  | 0.1$^1$            |
| $k_{dissolve}$  | Dissolution rate constant | 0 s$^{-1}$,$^2$  |

*1 Assumed default value

*2 No dissolution, as nano-TiO2 is practically insoluble.
Table S22. The first-order rate constant values that SimpleBox4nano calculates for environmental transport and removal processes for the Mueller and Nowack nano-TiO₂ emission scenario

| Symbol      | Transport / Removal Process                                                                 | Rate*⁻¹  |
|-------------|---------------------------------------------------------------------------------------------|----------|
| kₘₙₐₜₐ   | Aggregation rate for ENPs with aerosol particles in dry air (A)                              | 1.57E-06 s⁻¹ |
| kₘₙₐₜₐ   | Attachment rate for ENPs with coarse particles in dry air (A)                                | 1.32E-08 s⁻¹ |
| kₐₐₚₐₜₐ₂₈  | Rain drop collection rate for free ENP species                                              | 6.02E-06 s⁻¹ |
| kₐₐₚₐₜₐ₂₈  | Rain drop collection rate for aggregated ENP species                                         | 1.81E-06 s⁻¹ |
| kₐₐₚₐₜₐ₂₈  | Rain drop collection rate for attached ENP species                                          | 4.44E-06 s⁻¹ |
| kₐₘₙₐₜₜₜₛ  | First-order rate constant for dry deposition from dry air (A) to soil (S) for free ENP species | 1.59E-06 s⁻¹ |
| kₐₘₙₐₜₜₜₗ  | First-order rate constant for dry deposition from dry air (A) to soil (S) for aggregated ENP species | 2.51E-07 s⁻¹ |
| kₐₘₙₐₜₜₜₘ  | First-order rate constant for dry deposition from dry air (A) to soil (S) for attached ENP species | 9.41E-07 s⁻¹ |
| kₐₘₙₐₜₜₜₜ  | First-order rate constant for dry deposition from dry air (A) to soil (W) for free ENP species | 2.40E-08 s⁻¹ |
| kₐₘₙₐₜₜₜₜ  | First-order rate constant for dry deposition from dry air (A) to water (W) for aggregated ENP species | 2.80E-08 s⁻¹ |
| kₐₘₙₐₜₜₜₜ  | First-order rate constant for dry deposition from dry air (A) to water (W) for attached ENP species | 3.00E-08 s⁻¹ |
| kₐₘₙₐₜₜₜₜ  | First-order rate constant for wet deposition from rain (R) to soil (S) for free, aggregated or attached ENP species | 2.39E-03 s⁻¹ |
| kₐₘₙₐₜₜₜₜ  | First-order rate constant for wet deposition from rain (R) to water (W) for free, aggregated or attached ENP species | 7.62E-05 s⁻¹ |
| kₐₘₙₐₜₜₜₜ  | Aggregation rate for ENPs with natural colloids in soil (S) pore water                      | 8.42E-07 s⁻¹ |
| kₐₘₙₐₜₜₜₜ  | Attachment rate for ENPs with solid grains in soil (S)                                       | 3.65E-03 s⁻¹ |
| kₐₘₙₐₜₜₜₜ  | First-order rate constant for run-off from soil (S) to water (W) for free or aggregated ENP species | 2.93E-07 s⁻¹ |
| Parameter | Description | Value |
|-----------|-------------|-------|
| $k_{erosionSWatt}$ | First-order rate constant for run-off from soil (S) to water (W) for attached ENP species | 1.68E-11 s$^{-1}$ |
| $k_{leachSagg,free}$ | First-order rate constant for run-off from soil (S) to water (W) for free ENP species | 2.93E-07 s$^{-1}$ |
| $k_{aggW}$ | Aggregation rate for ENPs with natural colloids in surface water (W) | 8.42E-07 s$^{-1}$ |
| $k_{attW}$ | Attachment rate for ENPs with suspended particles in surface water (W) | 6.10E-10 s$^{-1}$ |
| $k_{depWSEfree}$ | First-order rate constant for deposition from water (W) to sediments (SE) for free ENP species | 2.34E-10 s$^{-1}$ |
| $k_{depWSEatt}$ | First-order rate constant for deposition from water (W) to sediments (SE) for attached ENP species | 2.36E-05 s$^{-1}$ |
| $k_{depWSEagg}$ | First-order rate constant for deposition from water (W) to sediments (SE) for aggregated ENP species | 1.64E-08 s$^{-1}$ |
| $k_{rsSEWfree,agg}$ | First-order rate constant for resuspension from sediments (SE) to water (W) for free or aggregated ENP species*2 | 7.60E-07 s$^{-1}$ |
| $k_{rsSEWatt}$ | First-order rate constant for resuspension from sediments (SE) to water (W) for attached ENP species*2 | 3.04E-06 s$^{-1}$ |

*1 Calculated as a function of the system dimensions and default input parameters given in SI Table 16 and 17

*2 SB4N calculates the 1-year-PECs for this scenario as a function of one-directional transport only, since backward processes can usually be neglected. However, it should be considered that the backward resuspension process (an advective process that transports the free, aggregated, and attached ENPs that have settled in the sediment back to the water compartment) may not be negligible. Therefore, the influence of resuspension on both the 1-year-PECs in the water and sediment compartment has been investigated for this scenario. This was done by correcting the settling rates for backward resuspension by expressing resuspension as a resistance to settle:

$$v_{sett(netto,i)} = \frac{1}{1 + \left(\frac{v_{resusp}}{v_{sett(i)}}\right) \cdot v_{sett(i)}}$$

This approach has been verified by comparing the calculated concentrations PECs derived from the formulations for steady state, with the calculated concentrations using the corrected settling rates over infinite time. There were no major differences observed, which indicates that it is an acceptable approach to express resuspension as a resistance to settle for this scenario. However, this approach still neglects a minor mechanism contributing to concentration of ENPs attached to suspended particles in water: free ENPs may settle to the sediment and then attach to the solid grains in the sediment compartments. If these solid grains resuspend, they will transport these ENPs back the water compartment as attached species. Nonetheless, this mechanism actually can be assumed to be negligible as settling of free ENPs is negligible.
compared to the settling behavior of ENPs attached to suspended particles, which is also observed in experiments.  

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