Electronic Supplementary Material (ESI) for Environmental Science: Nano.

Electronic Supplementary Information (ESI): A Model Sensitivity Analysis To Determine The Most Important Physicochemical Properties Driving Environmental Fate and Exposure of Engineered Nanoparticles

Johannes A.J. Meesters[a,b*1], Willie J.G.M. Peijnenburg[b,c], A. Jan Hendriks[a] and Dik van de Meent[a], Joris T.K. Quik[b]

a:Institute for Water and Wetland Research, Department of Environmental Science, Radboud University Nijmegen, P.O. Box 9010, NL-6500 GL Nijmegen, The Netherlands
b: National Institute of Public Health and the Environment RIVM, P.O. Box 1, 3720 BA, Bilthoven, The Netherlands
c: Institute of Environmental Sciences (CML), Leiden University, PO Box 9518, 2300 RA Leiden, The Netherlands.

*: Corresponding author: Email: joris.meesters@rivm.nl.

A: Sensitivity plots of predicted free, bioavailable and total concentrations in air, water, sediment and soil

Predicted environmental concentrations are displayed as a function of the following physicochemical properties of ENPs: diameter, specific weight, transformation rate constant, attachment efficiency with natural particles, and Hamaker constant with natural particles in water.

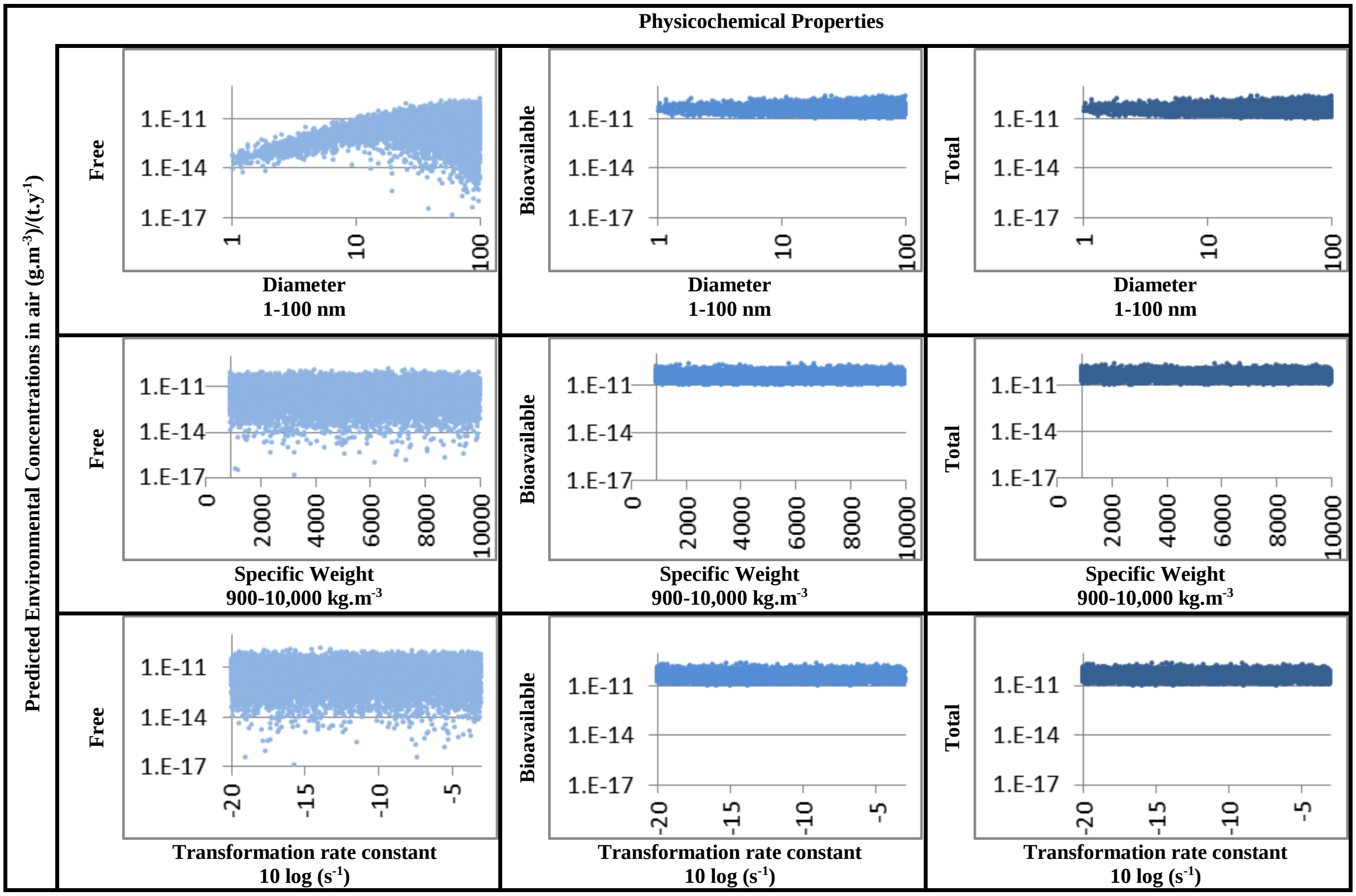


Figure 1. Extracted data sheet plots for SB4N sensitivity of ENP-diameter, specific weight, and transformation rate constant on free, bioavailable and total concentrations in air at 1 $t.y^{-1}$ emission.

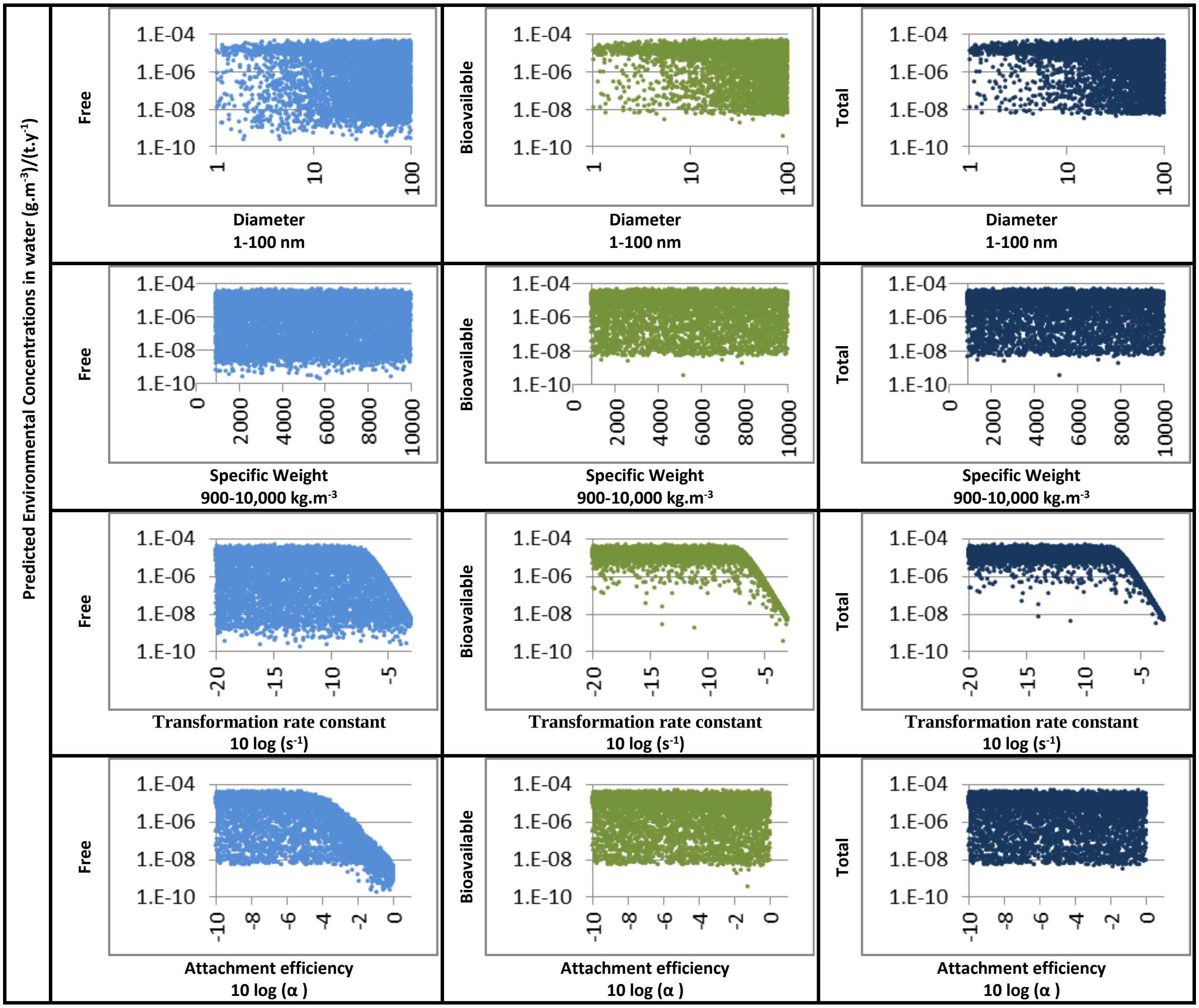


Figure 2. Extracted data sheet plots for SB4N sensitivity of ENP-diameter, specific weight, transformation rate constant, and attachment efficiency on free, bioavailable and total concentrations in water at 1 t.y^{-1} emission.

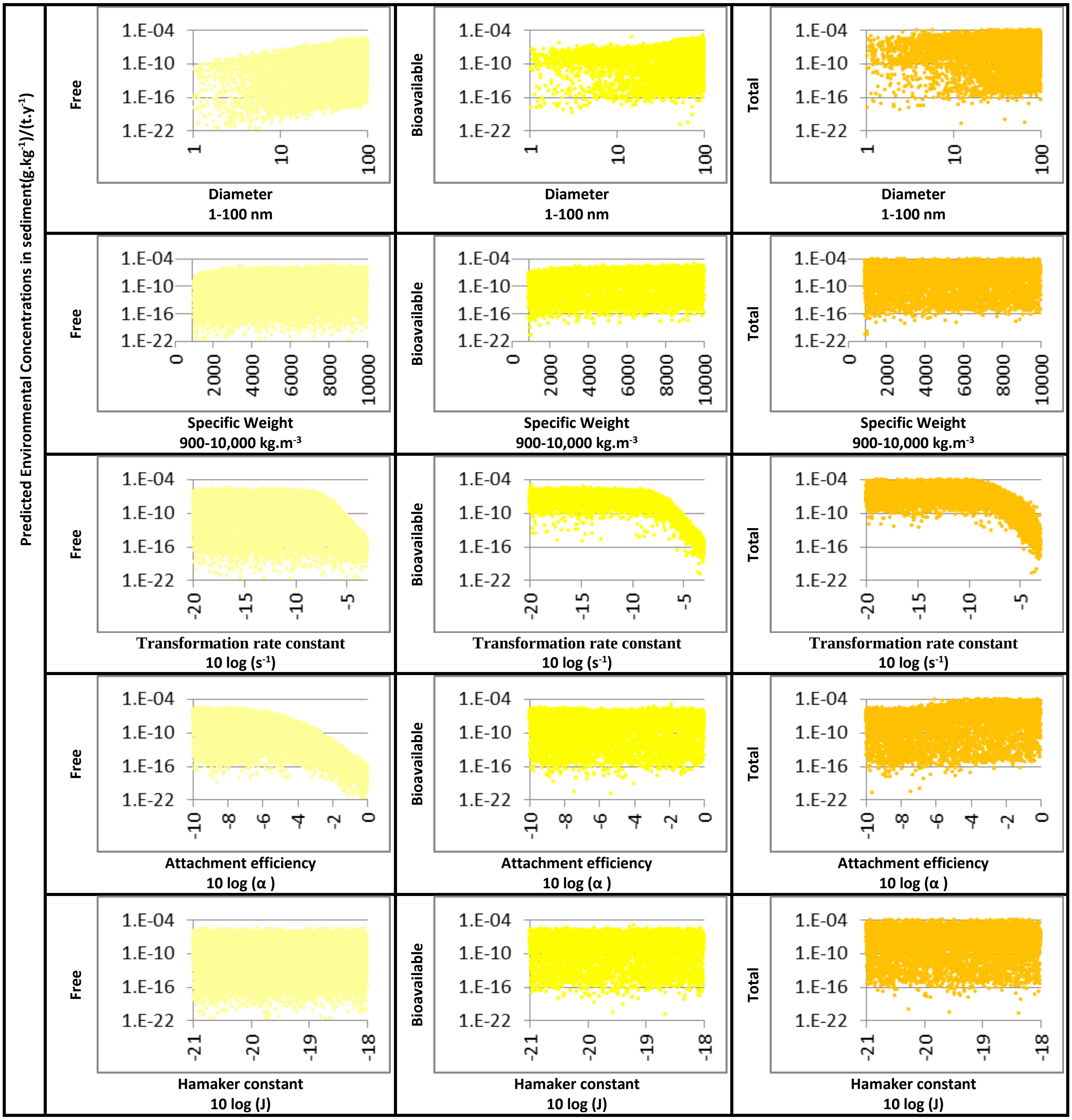


Figure 3. Extracted data sheet plots for SB4N sensitivity of ENP-diameter, specific weight, transformation rate constant, attachment efficiency and Hamaker constant on free, bioavailable and total concentrations in sediment at 1 t.y^{-1} emission.

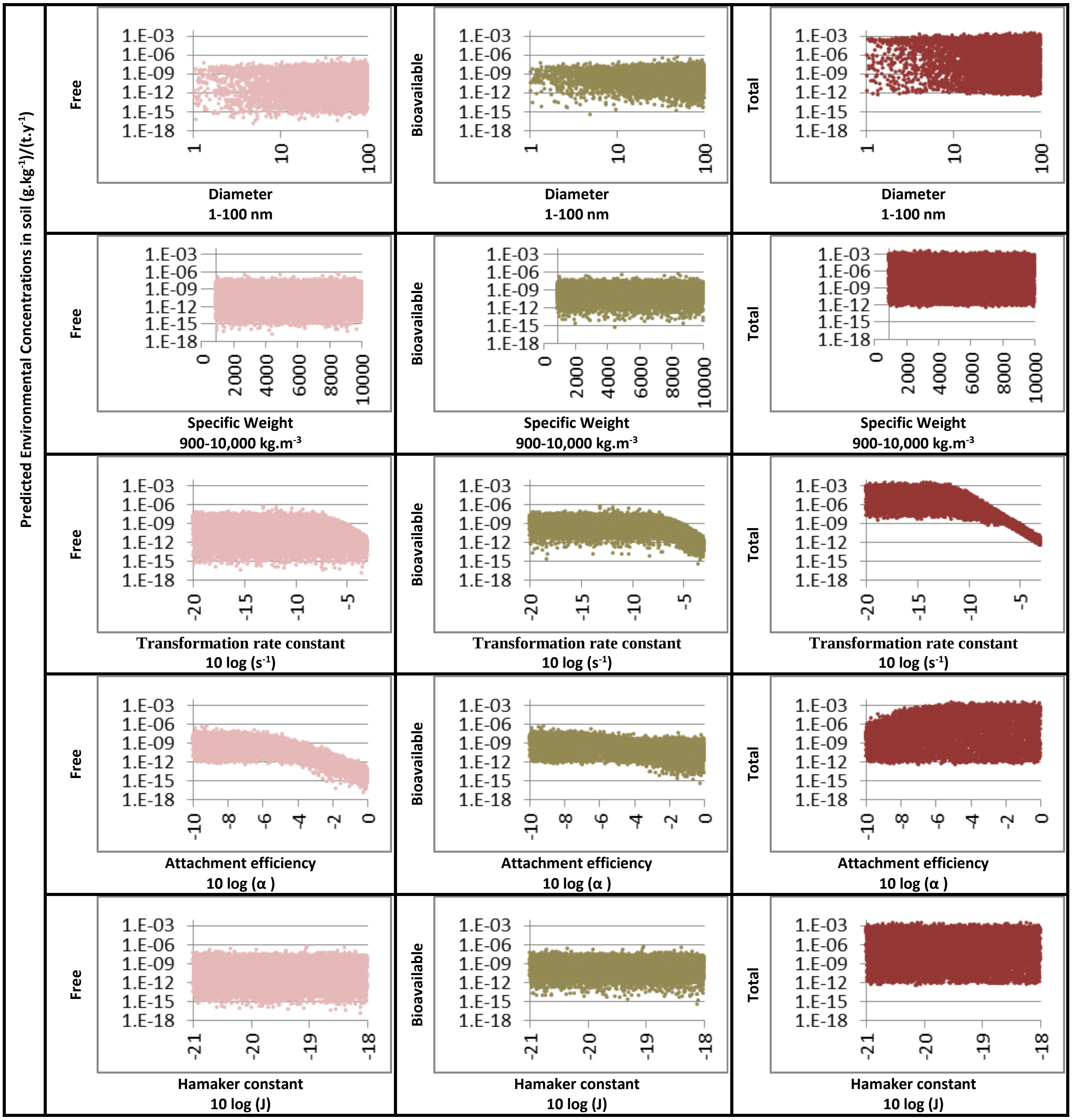


Figure 4. Extracted data sheet plots for SB4N sensitivity of ENP-diameter, specific weight, transformation rate constant, attachment efficiency and Hamaker constant on free, bioavailable and total concentrations in soil at 1 $t.y^{-1}$ emission.

B: Sensitivity plots of simulated environmental fate processes

B1: Air

The extracted data sheet resulting from the Monte Carlo simulation performed for the scenario of 1 t/y emission to air includes 10,000 iterations of rate constants for every atmospheric fate process included in the SimpleBox4nano (SB4N) model. The distribution in simulated values for the rate constants per atmospheric fate process removing free ENPs from air is given below in a box plot (ESI Figure 5).

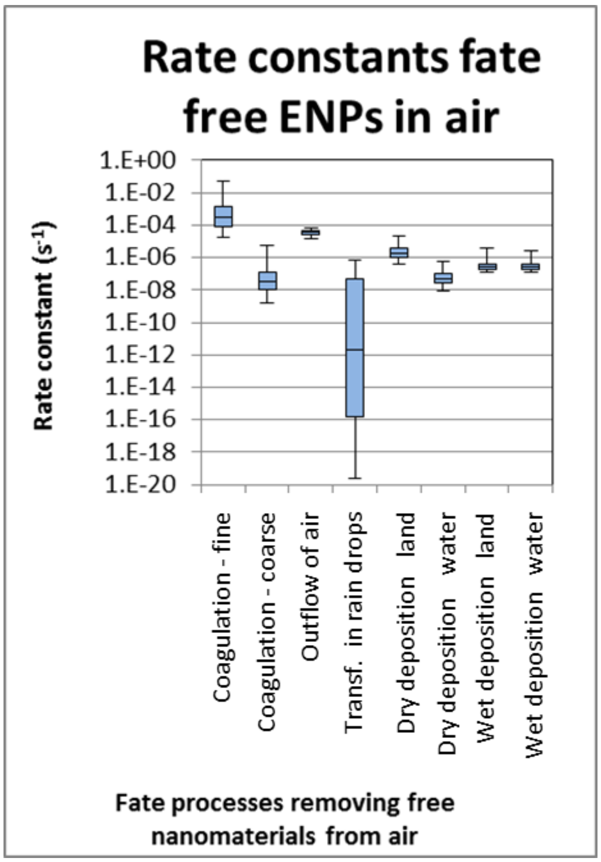


Figure 5. Box plots of simulated rate constants for atmospheric fate processes removing free ENPs from air. The boxes represent 25th , 50th and 75th percentiles, whereas the whiskers indicate the 2.5 and 97.5-percentiles.

Coagulation with fine aerosols and outflow of air are the dominant process removing free ENPs from air. Outflow of air is included in SB4N as an advective process that is independent of physicochemical properties of the ENP, whereas the algorithms to derive coagulation with fine aerosol include a function of ENP size. The rate constant for coagulation with fine aerosols is included in SB4N as the sum of the rate constant for coagulation with nucleation and accumulation mode aerosols. The rate constant for coagulation with accumulation mode aerosols is dominant for small ENPs, but decreases with ENP size. The rate constant for coagulation with nucleation mode aerosols increase with ENP size, but the variation in values for the simulated rate constants is larger (ESI Figure 6). The critical ENP diameter at which coagulation with nucleation mode aerosols becomes larger than that of accumulation mode aerosols is calculated to be 38 nm by median (95% CI = 21-60 nm).

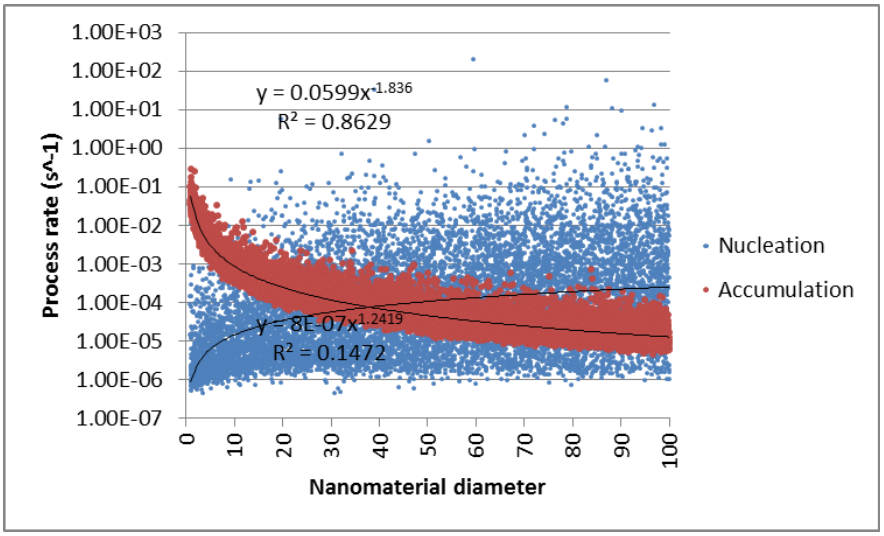
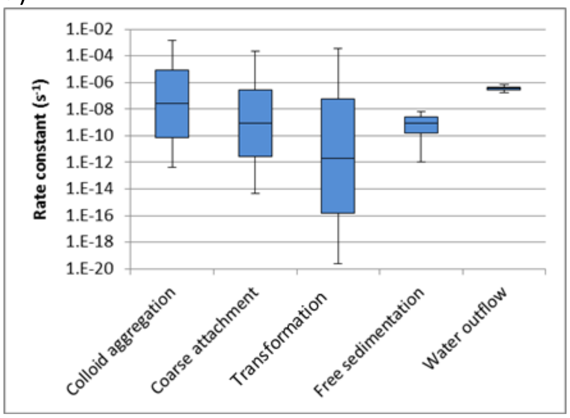


Figure 6. Scatter plot of simulated rate constants for coagulation with nucleation and accumulation mode aerosols as a function of the diameter of the engineered nanoparticle (ENP).

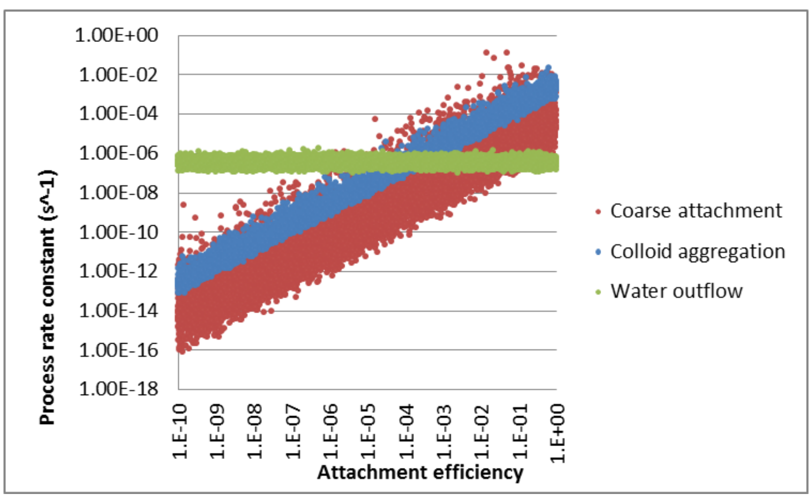
B2: Water

The extracted data sheet resulting from the Monte Carlo simulation performed for the scenario of 1 t/y emission to water includes 10,000 iterations of rate constants for every aquatic fate process included in the SimpleBox4nano (SB4N) model. The distribution in simulated values for the rate constants per aquatic fate process removing free ENPs from the water column is given below in a box plot (ESI Figure 7)



ESI Figure 7. Box plots of simulated rate constants for aquatic fate processes removing free ENPs from water. The boxes represent 25th , 50th and 75th percentiles, whereas whisker the 2.5 and 97.5-percentiles.

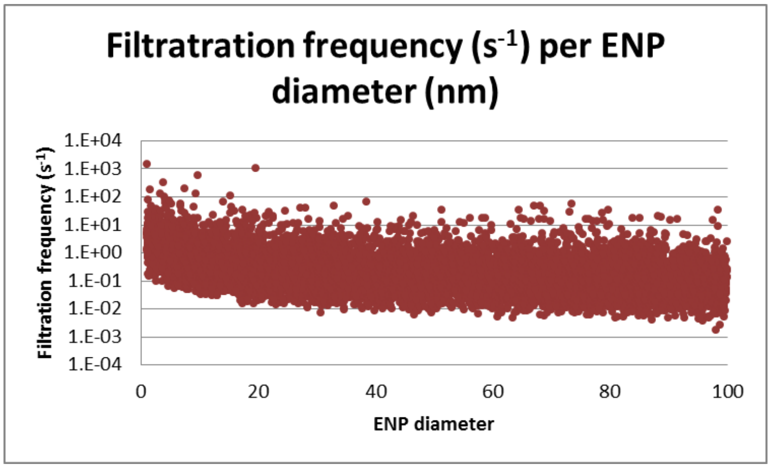
The rate constant for transformation is directly inserted in SB4N as an input parameter, whereas aggregation with natural colloids and attachment to natural coarse particles are included in SB4N as the frequency of collisions with the natural particles multiplied with the attachment efficiency. As such the rate constants for aggregation with natural colloids and attachment to natural coarse particles are linear to attachment efficiency (ESI Figure 8). The critical attachment efficiency at which the sum of the rate constants for aggregation with natural colloids and attachment to natural coarse particle is larger than removal by the outflow of water is calculated to be 1.1 10^{-4} by median (95%CI = 7.5 10^{-5} - 1.6 10^{-4}).



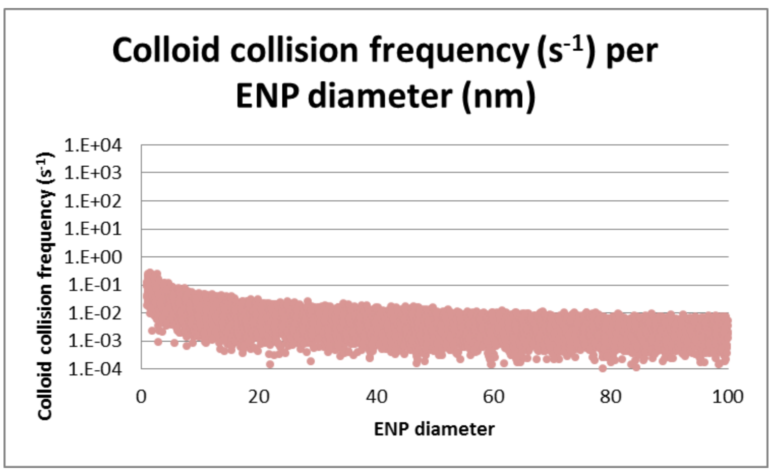
ESI Figure 8. Scatter plot of simulated rate constants for aggregation with natural colloid particles and attachment to natural coarse particles as function of their attachment efficiency with the engineered nanoparticle in comparison with the simulated rate constant for water outflow.

B3: Soil

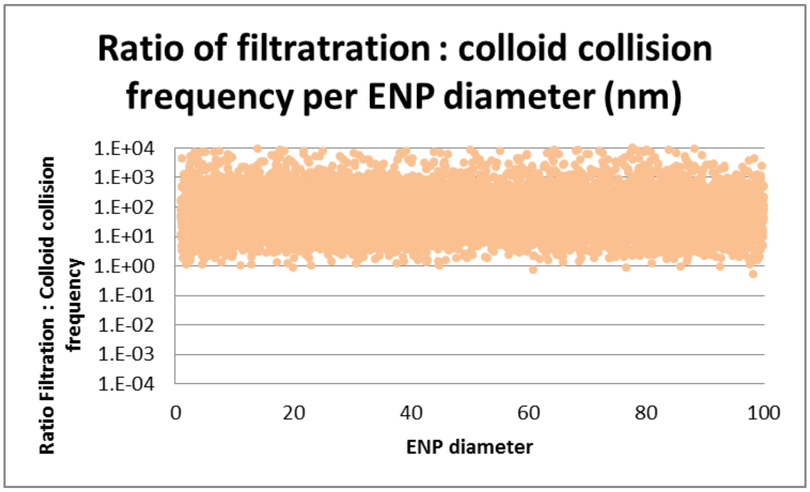
The simulated filtration frequency of ENPs in soil weakly decreases with ENP diameter (ESI Figure 9). The frequency of collision with natural colloids in soil pore water also decrease weakly with ENP diameter (ESI Figure 10), but the ratio of filtration and colloid collision frequency remains greater than 1 independent of ENP diameter (ESI Figure 11). As such, filtration of ENPs by soil grains is dominant over hetero-aggregation with natural colloid particles in the soil pore waters at similar attachment efficiencies.



ESI Figure 9. Filtration frequency of engineered nanoparticles (ENPs) n soil as a function of their diameter



ESI Figure 10. Frequency of collisions between natural colloid particles and engineered nanoparticles (ENPs) in soil as a function of ENP diameter



ESI Figure 11 The ratio of filtration frequency to the frequency of collisions with natural colloid particles as a function of the diameter of the engineered nanoparticle (ENP).

C: Simplification and verification of algorithms explaining model sensitivity

C1: Verification by deriving R^2 values.

The original algorithms of SB4N include mechanistic expressions to calculate the rate constants for environmental fate processes as a function of ENP physicochemical properties and environmental conditions. These original algorithms are consulted to derive simplified equations that mechanistically express the relationship between the PECs and the physicochemical properties determinant for the fate processes that are identified to be dominant. The extent to which these simplified expressions explain the sensitivity plots is assessed by calculating their R^2 against the extract data of the MC simulations:

$$R^2 = 1 - \frac{\sum_i (y_i - f_i)^2}{\sum_i (y_i - \bar{y})^2} = 1 - \frac{SS_{res}}{SS_{tot}} \tag{1}$$

Here y_i represents a data point for a PEC per iteration of the MC simulation, f_i represents the PEC calculated with the newly derived equation per iteration, $\bar{y}$ is the average of the simulated PEC for all 10,000 iterations, SS_{res} is the sum of all squares for the residuals and SS_{tot} the total sum of squares.

C2: Mass balance equations

Mass balance equations used to explain sensitivity and general patterns in predicted concentrations SimpleBox(4nano) model employs mass balance equations to calculate a steady state situation as a Level III Mackay multimedia chemical fate model[1-5]. The emission of ENPs refers to a free and pristine state in the air. The SB4N model does not include transport of ENPs from water or soil to the air or speciation processes that transform free ENPs in the air.[1] Therefore the sensitivity analysis on predicted free ENPs in the air can be expressed as a one-compartment Mackay model with:

$$m = \frac{e}{\sum k} \tag{2}$$

Here *m* refers to the steady state chemical mass, *e* to the emission and *Σk* to the sum of the rate constants for the processes responsible for the removal of the chemical. The chemical concentration (C) is then derived by dividing the steady chemical mass with the volume of the environmental compartment, so that:

$$C = \frac{m}{V} = \frac{e}{V}\frac{1}{\sum k} \tag{3}$$

Hence, the steady state chemical concentration is proportional to the inverse of the rate constant for the fate processes responsible for the removal of the chemical.

$$C \sim \frac{1}{\sum k} \tag{4}$$

C3: Simplified algorithms for air concentrations

Following the mass balance equations (ESI Section C2) the free ENPs in air the concentration is defined as:

$$C_{free\ in\ air} = \frac{e}{V_{air}} \frac{1}{\sum k_{free\ in\ air}} \tag{5}$$

The processes included in SB4N removing free ENPs in air are coagulation with fine aerosols, coagulation with coarse aerosols, outflow of air to a continental scale, wet deposition and dry deposition, so that:

$$\sum k_{free\ in\ air} = k_{coag.fine} + k_{coag.coarse} + k_{out.air} + k_{dep.wet} + k_{dep.dry} \tag{6}$$

The sensitivity plots that refer to the simulated rate constants for these removal processes indicate that coagulation and outflow obscure the other processes (ESI Figure 5). The sum of all rate constants for processes removing free ENPs out of the air can be approached with the sum of the rate constants for coagulation with fine aerosols and outflow of air alone:

$$k_{coag.fine} + k_{out.air} \approx \sum k_{free\ in\ air} \tag{7}$$

The concentration of free ENPs in the air can then be approached as:

$$C_{free\ in\ air} \approx \frac{e}{V_{air}} \frac{1}{k_{coag.fine} + k_{out.air}}, \qquad R^2 > 0.99 \tag{8}$$

C4: Simplified algorithms for water concentrations

The sensitivity analysis was performed on a scenario with emission to the water compartment alone. As such there is now inflow included from the transport of ENPs from compartments air and soil . The predicted concentrations in water by SB4N can therefore be compared with a one-compartment Mackay model in which the steady state chemical concentration is proportional to the inverse of the sum of the rate constants calculated for the fate processes responsible for the removal of the chemical (ESI Section C2):[2]

$$C \sim \frac{1}{\sum k} \tag{11}$$

The fate processes responsible for the removal of ENPs out of the water compartment are the outflow of water to a continental scale ($k_{out.water}$), (*ii*) the gravitational settling free ENPs and ENPs attached to natural particles to sediment ($\Sigma k_{set.total}$) and (*iii)* transformation (k_{trans}) so that the total concentration in water can be approached as:

$$C_{total\ in\ water} \approx \frac{e}{V}\frac{1}{k_{trans}+k_{out.water}+\sum k_{set.total}} \qquad R^2 = 0.89 \tag{12}$$

As such, the total concentration is proportional to the inverse of the rate constants for the removal processes considered:

$$C_{total\ in\ water} \sim \frac{1}{\sum k} = \frac{1}{k_{trans}+k_{out.water}+\sum k_{set.total}} \tag{13}$$

The sum of the rate constants for the settling of free ENPs, ENPs hetero-aggregated with natural colloids and ENPs attached to suspended coarse particles is derived as:

$$\sum k_{set.total} = k_{set.free} + \left(\frac{1}{k_{agg.colloids}} + \frac{1}{k_{set.agg-colloids}}\right)^{-1} + \left(\frac{1}{k_{att.coarse}} + \frac{1}{k_{set.att-coarse}}\right)^{-1} \tag{14}$$

Transformation becomes the dominant removal process when it is greater than the sum of the other removal process of water outflow and the settling of free ENPs and ENPs attached natural particles:

$$k_{trans} > k_{out.water} + \sum k_{set.total} \tag{15}$$

The critical rate at which transformation becomes the dominant removal process is then:

$$k_{trans(crit.total\ in\ water)} = k_{out.water} + \sum k_{set.total} \tag{16}$$

The same approach is followed in the derivation of the critical attachment efficiency and critical transformation rate constant at which predicted free concentrations in water become sensitive to these two physicochemical properties of the ENP. The processes responsible for removal of free ENPs are transformation, water outflow, settling of free ENPs, hetero-aggregation with natural colloids and attachment to coarse particles. The free concentration of ENPs can then be approached as:

$$C_{free\ in\ water} \approx \frac{e}{V_{water}} \frac{1}{k_{agg.colloids}+k_{att.coarse}+k_{trans}+k_{out.water}+k_{set.free}} \quad R^2 = 1$$

(17)

As such, the free concentration is proportional to the inverse of the sum of the rate constants for the removal processes considered:

$$C_{free\ in\ water} \sim \frac{1}{\sum k} = \frac{1}{k_{trans} + k_{agg.colloids} + k_{att.coarse} + k_{out.water} + k_{set.free}}$$

(18)

The attachment efficiency refers to the probability whether the ENPs sticks to a natural particle at a collision event. The attachment rates for hetero-aggregation and attachment to suspended coarse particles are expressed in the SB4N algorithms as:[1]

$$k_{agg.colloids} = \alpha\, f_{ENP-colloids},$$

(19)

, and

$$k_{att.coarse} = \alpha\, f_{ENP-coarse}$$

(20)

The free concentration is then proportional to :

$$C_{free\ in\ water} \sim \frac{1}{\sum k} = \frac{1}{\alpha(f_{ENP-colloids} + f_{ENP-coarse}) + k_{trans} + k_{out.water} + k_{set.free}}$$

(21)

Hetero-aggregation with natural colloids and attachment to natural coarse particles become dominant over the other removal processes if:

$$k_{agg.colloids} + k_{att.coarse} > k_{trans} + k_{out.water} + k_{set.free}$$

(22)

,so that

$$\alpha(f_{ENP-colloids} + f_{ENP-coarse}) > k_{trans} + k_{out.water} + k_{set.free}$$

(23)

The critical attachment efficiency at which hetero-aggregation and attachment to coarse particles become dominant over the other removal processes is then derived as:

$$\alpha_{crit.free\ in\ water} = \frac{k_{trans} + k_{out.water} + k_{set.free}}{(f_{ENP-colloids} + f_{ENP-coarse})}$$

(24)

The critical transformation rate complementary to attachment efficiency is then derived as:

$$k_{trans(crit.free\ in\ water)} = \alpha(f_{ENP-colloids} + f_{ENP-coarse}) + k_{set.free} + k_{out.water}$$

(25)

C5: Simplified algorithms for sediment concentrations

The concentration of ENPs in sediment ($g.kg^{-1}$) is expressed as the steady state mass of ENPs in sediment divided by the volume (m^3) multiplied with the density of the sediment compartment ($kg.m^{-3}$)

$$C = \frac{m}{V\rho} \tag{26}$$

The steady state mass of ENPs in sediment is reached if the ENP mass inflow ($g.s^{-1}$) equals the mass outflow ($g.s^{-1}$).

$$\frac{dm_{free\ in\ sed}}{dt} = F_{in} - F_{out} = 0 \tag{27}$$

Free ENPs enter the sediment via settling from the water column and are removed from sediment via transformation, resuspension, burial, hetero-aggregation with natural colloids and attachment to coarse particles via filtration, so that:

$$F_{in.free\ in\ sed} = m_{free\ in\ water} \cdot k_{set.free}, \tag{28}$$

and

$$F_{out.free\ in\ sed} = m_{free\ in\ sed} \cdot (k_{trans} + k_{burial} + k_{resusp} + k_{agg.colloid\ (sed)} + k_{filtr.sed}) \tag{29}$$

The ENP mass inflow ($g.s^{-1}$) then equals the mass outflow ($g.s^{-1}$) if:

$$m_{free\ in\ water} \cdot k_{settling.free} = m_{free\ in\ sed} \cdot (k_{trans} + k_{burial} + k_{resusp} + k_{agg.colloid(sed)} + k_{filtr.sed}) \tag{30}$$

The steady state mass of free ENPs in water follows a mass balance equation that describes the emission divided by the sum of the rate constant for the removal processes(ESI Section C2) so that:

$$m_{free\ in\ water} = \frac{e}{k_{trans} + k_{out.water} + k_{set.free} + k_{agg.colloids(water)} + k_{att.coarse(water)}} \tag{31}$$

Inserting the equation above in the mass balance for free ENPs in sediment then yields:

$$\frac{e}{k_{trans} + k_{out.water} + k_{set.free} + k_{agg.colloids(water)} + k_{att.coarse(water)}} \cdot k_{set.free} =$$

$$m_{free\ in\ sed} \cdot (k_{trans} + k_{burial} + k_{resusp} + k_{agg.colloid(sed)} + k_{filtr.sed}) \tag{32}$$

The mass of free ENPs in sediment is then derived as a function of emission and rate constants for the environmental fate of free ENPs in the water column and sediment:

$$m_{free\ in\ sed} = e \cdot k_{set.free} \frac{1}{(k_{trans} + k_{burial} + k_{resusp} + k_{agg.colloid(sed)} + k_{filtr.sed})(k_{trans} + k_{out.water} + k_{set.free} + k_{agg.colloids(water)} + k_{att.coarse(water)})}$$

$R^2>0.99$

(33)

C5.1 Free concentrations in sediment linear to Stokes settling velocity

The equation above is verified (ESI section C2) by the derived R^2 that is greater than 0.99. As such, it can be proposed that the concentration of free ENPs in sediment is linear proportional the settling rate of free ENPs:

$$C_{free\ in\ sed} \sim k_{set.free}$$

(34)

The settling rate of free ENPs is calculated by SB4N as the Stokes settling velocity ($m.s^{-1}$) divided by the depth of the water column (m):

$$k_{set.free} = \frac{v_{stokes}}{h_{water}}$$

(35)

As such, the predicted concentration of free ENPs in sediment is also linear proportional to the Stokes settling velocity of ENPs.

C5.2 Critical transformation rate constant for free concentrations in sediment

The critical transformation rate is defined as the rate at which transformation is the dominant process removing free ENPs from sediment. Transformation of ENPs occurs both in the water column prior to settling as well as in the sediment compartment itself. The critical transformation rate constant is therefore interpreted as the transformation rate constant at which the impact of removal by transformation is equal to the removal by other the fate processes, which is here defined as the predicted free mass of transforming ENPs in sediment is equal to half of the mass if the ENPs would be not transform at all:

$$m_{free\ in\ sed, k_{trans}=crit} = \frac{1}{2} m_{free\ in\ sed, k_{trans}=0}$$

(36)

Filling eq 33 for both the left and right term in the equation above then yields:

$$\frac{k_{set.free}}{(k_{trans.crit} + k_{burial} + k_{resusp} + k_{agg.colloid(sed)} + k_{filtr.sed})(k_{trans.crit} + k_{out.water} + k_{set.free} + k_{agg.colloids(water)} + k_{att.coarse(water)})} = \frac{1}{2} \frac{k_{set.free}}{(k_{burial} + k_{resusp} + k_{agg.colloids(sed)} + k_{filt.sed})(k_{out.water} + k_{set.free} + k_{agg.colloid(water)} + k_{att.coarse(water)})}$$

(37)

Then, if '$k_{burial}+k_{resusp}+k_{agg.colloids(sed)+}k_{filtr.sed}=a$', '$k_{out.water}+k_{set.free}+k_{agg.colloid(water)+}k_{att.coarse(water)}=b$' ,'$k_{set.free}$ =c' and '$k_{trans.crit}=x$', the mathematical solution for the equation would be:[6]

$$x = \frac{\sqrt{(b^2 + 6ab + a^2)} - a - b}{2}$$

(38)

Inserting eq (38) in the extract datasheet of the MC simulation gives a median critical transformation rate constant of $k_{trans.crit}$ =1 10^{-7} s^{-1} with a 95% confidence interval of 1 10^{-9} -1 10^{-3} s^{-1}. The broad 95% confidence interval is largely determined by the variation in hetero-aggregation with natural colloids and attachment to natural coarse particles as a consequence of the range inserted for attachment efficiency (main article table 1). For stable ENPs (α =0), there is no removal of free ENPs by hetero-aggregation with natural colloids and attachment to natural coarse particles, so that the eq 37 can be simplified to:

$$\frac{k_{set.free}}{(k_{transs.crit} + k_{burial} + k_{resusp})(k_{trans.crit} + k_{out.water} + k_{set.free})} = \frac{1}{2}\frac{k_{set.free}}{(k_{burial} + k_{resusp})(k_{out.water} + k_{set.free})}$$

(39)

Then, if '$k_{burial}+k_{resusp}=a$', '$k_{out.water}+k_{set.free}=b$' ,'$k_{set.free}$ =c' and '$k_{trans.crit}=x$', the mathematical solution for the equation would be:[6]

$$x = \frac{\sqrt{(b^2 + 6ab + a^2)} - a - b}{2}$$

(40)

Inserting eq. 40 in the extract datasheet of the MC simulation gives a median critical transformation rate constant for stable ENPs of $k_{trans.crit}$ =1 10^{-8} with a 95% confidence interval of 1 10^{-9} -5 10^{-8}

C5.3 Critical attachment efficiency for free concentrations in sediment

The critical attachment efficiency is interpreted as the attachment efficiency at which the impact of removal by hetero-aggregation with natural colloids and attachment to natural coarse via filtration in sediment and collisions in the water column is equal to the removal by other the fate processes. This is the case if the predicted free mass of instable ENPs in sediment is equal to half of the mass if the ENPs would be completely stable (α = 0):

$$m_{free\ in\ sed,\alpha=\alpha_{crit}} = \frac{1}{2} m_{free\ in\ sed_{\alpha=0}},$$

(41)

Filling eq 33 for both the left and right term in the equation above then yields:

$$\frac{k_{set.free}}{(k_{trans} + k_{burial} + k_{resusp} + \alpha(f_{agg.colloid(sed)} + f_{filtr.sed}))(k_{trans} + k_{out.water} + k_{set.free} + \alpha(f_{agg.colloid(water)} + f_{att.coarse(water)}))} = \frac{1}{2}\frac{k_{set.free}}{(k_{dis} + k_{burial} + k_{resusp})(k_{trans} + k_{out.water} + k_{set.free})}$$

(42)

Then, if '$k_{settling.free}$=a', 'k_{transs}=b','$k_{burial}+k_{resuspension}$=c', '$f_{aggcolloid(sed)}+f_{filtr.sed}$=d', '$k_{out.water}+k_{settling.free}$=e', '$f_{agg.colloid(water)}+f_{att.coarse(water)}$=f', and '$\alpha_{crit}$ =x', the mathematical solution for the equation would be:[6]

$$x = \frac{\left(\sqrt{(c^2+2bc+b^2)f^2+[(6c+6b)de+(6bc+6b^2)d]f+d^2e^2+2bd^2e+b^2d^2}\right)+\left((-c)-b\right)f-de-bd)}{2df}$$

(43)

Inserting eq. 41 in the extract datasheet of the MC simulation gives a median critical attachment efficiency of = 1.1 10^{-6} for all ENPs and a median critical attachment efficiency of 6.0 10^{-7} for non-transforming ENPs (k_{trans} =0)

C5.4 Speciation of ENPs in water column and sediment

ENPs enter the sediment compartment via settling from the water column. Hetero-aggregation with natural colloids and attachment to natural coarse particle are found to be the dominant mechanisms for such sedimentation.[7] The mass flows of as settling ENPs free species, ENPs hetero-aggregated with natural colloids and ENPs attached to natural coarse particles calculated are calculated as:

$$F_{settling.free} = m_{free\ in\ water} k_{set.free}$$

(44)

$$F_{settling.agg.colloids} = m_{agg.colloids\ in\ water} k_{set.agg-colloids}$$

(45)

$$F_{settling.att.coarse} = m_{att.coarse\ in\ water} k_{set.att-coarse}$$

(46)

The free ENP species, ENPs hetero-aggregated with natural colloids and ENPs attached to natural coarse particles as fractions of the total mass flow of ENPs settling to the sediment compartment are then calculated as:

$$FR_{free\ in\ setling} = \frac{F_{settling.free}}{F_{settling.free} + F_{settling.agg.colloids} + F_{settling.att.coarse}}$$

(47)

$$FR_{agg.colloids\ in\ setling} = \frac{F_{settling.agg.colloids}}{F_{settling.free} + F_{settling.agg.colloids} + F_{settling.att.coarse}}$$

(48)

$$FR_{att.coarse\ in\ setling} = \frac{F_{settling.att.coarse}}{F_{settling.free} + F_{settling.agg.colloids} + F_{settling.att.coarse}}$$

(49)

The fraction of free ENP species, ENPs hetero-aggregated with natural colloids and ENPs attached to natural coarse particles as fractions in the sediment compartment are calculated as:

$$FR_{free\ in\ sed} = \frac{m_{free\ in\ sed}}{m_{free\ in\ sed} + m_{agg.colloid\ in\ sed} + m_{att.coarse\ in\ sed}}$$

(50)

$$FR_{agg.colloids\ in\ sed} = \frac{m_{agg.colloid\ in\ sed}}{m_{free\ in\ sed} + m_{agg.colloid\ in\ sed} + m_{att.coarse\ in\ sed}}$$

(51)

$$FR_{att.coarse\ in\ sed} = \frac{m_{att.coarse\ in\ sed}}{m_{free\ in\ sed} + m_{agg.colloid\ in\ sed} + m_{att.coarse\ in\ sed}}$$

(52)

The statement that speciation pattern of ENPs in sediment is largely determined in the water column is then verified by comparing the fractions the different species in the total mass flow of settling ENP with the fractions of the species in sediment and deriving the respective R^2's(ESI Section C1):

$$FR_{free\ in\ settling} \approx FR_{free\ in\ sed} \qquad R^2 = 0.77$$

(53)

$$FR_{agg.colloids\ in\ settling} \approx FR_{agg.colloids\ in\ sed} \qquad R^2=0.88$$

(54)

$$FR_{att.coarse\ in\ settling} \approx FR_{att.coarse\ in\ sed} \qquad R^2=0.85$$

(55)

C5.5: Critical transformation rate constant for total concentration in sediment

The concentration of ENPs in sediment ($g.kg^{-1}$) is expressed as the steady state mass of ENPs in sediment divided by the volume (m^3) multiplied with the density of the sediment compartment ($kg.m^{-3}$)

$$C = \frac{m}{V\rho}$$

(56)

The steady mass of ENPs in sediment is reached if the ENP mass inflow ($g.s^{-1}$) equals the mass outflow ($g.s^{-1}$).

$$\frac{dm_{total\ in\ sed}}{dt} = F_{in} - F_{out} = 0$$

(57)

ENPs enter the sediment via settling from the water column as free species, ENPs hetero-aggregated with natural colloids and ENPs attached to natural coarse particles and are removed from sediment via transformation, resuspension, burial, so that:

$$F_{in} = m_{total\ in\ water} \sum k_{set.total}$$

(58)

$$F_{out} = m_{total\ in\ sed}(k_{trans} + k_{burial} + k_{resusp})$$

(59)

Inserting eq. 13 for the total mass of ENPs in the water column in the mass balance for ENPs in sediment then yields:

$$\frac{1}{k_{trans} + k_{out.water} + \sum k_{set.total}} \sum k_{set.total} = m_{total\ in\ sed}(k_{trans} + k_{burial} + k_{resusp})$$

(60)

Hence, the total mass of ENPs in sediment can be derived as:

$$m_{total\ in\ sed} \approx e \sum k_{set.total} \frac{1}{(k_{trans}+k_{out.water}+\sum k_{set.total})(k_{trans}+k_{burial}+k_{resusp})} \quad R^2=0.38$$

(61)

The critical transformation rate constant is interpreted as the transformation rate at which the impact of removal by transformation is equal to the removal by other the fate processes, which is the case if the predicted total mass of transforming ENPs in sediment is equal to half of the mass if the ENPs would be not transform at all:

$$\frac{1}{(k_{trans(crit.\ total\ in\ sed)}+k_{out.water}+\sum k_{set.total})(k_{trans(crit.total\ in\ sed)}+k_{burial}+k_{resusp})} = \frac{1}{2}\frac{1}{(k_{out.water}+\sum k_{set.total})(k_{burial}+k_{resusp})}$$

(62)

Then, if '$k_{out.water}$+$\Sigma k_{set.total}$=a', 'k_{burial}+k_{resusp}=b' and '$k_{trans(crit.\ total\ in\ sed)}$=$x$', the mathematical solution would be:[6]

$$x = \frac{\left(\sqrt{b^2 + 6ab + a^2} - b - a\right)}{2}$$

(63)

C 6: verification soil concentrations

The concentration of ENPs in soil ($g.kg^{-1}$) is expressed as the steady state mass of ENPs in soil divided by product of the volume (m^3) and the density of the soil compartment ($kg.m^{-3}$)

$$C = \frac{m}{V\rho} \quad (64)$$

The steady of mass of ENPs attached to coarse grains is reached if the ENP mass inflow ($g.s^{-1}$) equals the mass outflow ($g.s^{-1}$).

$$\frac{dm_{att.coarse\ in\ soil}}{dt} = F_{in} - F_{out} = 0 \quad (65)$$

The mass inflow of ENPs coarse attached ENPs in soil is equal to the steady state mass of free ENPs in soil multiplied with the first-order rate constant for filtration

$$F_{in} = m_{free\ in\ soil} k_{filtr.soil} \quad (66)$$

The free concentration in soil is equal to the emission volume of ENPs divided by the sum of all rate constants removing free ENPs from soil:

$$m_{free\ in\ soil} = \frac{e}{k_{agg.colloid(soil)} + k_{filtr.soil} + k_{leach} + k_{run-off} + k_{trans}} \qquad R^2=1 \quad (67)$$

The mass outflow of ENPs attached to coarse particles in soil is the equal to the steady state mass of the coarse attached ENPs multiplied with the sum of the rate constants for the processes removing them, which are transformation and erosion:

$$F_{out} = m_{att.coarse\ in\ soil}(k_{trans} + k_{erosion}) \quad (68)$$

Inserting equations 66-68 in equation 65 then yields:

$$\frac{e}{k_{agg.colloid(soil)} + k_{filtr.soil} + k_{leach} + k_{run-off} + k_{dis}} k_{filtr.soil} = m_{att.coarse\ in\ soil}(k_{trans} + k_{erosion}) \quad (69)$$

so, that:

$$m_{att.coarse\ in\ soil} = k_{filtr.soil} \frac{e}{k_{agg.colloid(soil)} + k_{filtr.soil} + k_{leach} + k_{run-off} + k_{trans}} \frac{1}{k_{trans} + k_{erosion}}$$

R^2=1

(70)

The mass inflow and outflow to calculate the steady state mass of ENPs hetero-aggregated with natural colloids in soil are:

$$F_{in} = m_{free\ in\ soil} k_{agg.colloid(soil)} \quad (71)$$

and

$$F_{out} = m_{agg.colloid\ in\ soil}\left(k_{trans} + k_{run-off} + k_{leach}\right)$$

(72)

so, that the steady mass of ENPs hetero-aggregated with natural colloids in soil can be derived as:

$$m_{agg.colloids\ in\ soil} = k_{agg.colloid(soil)} \frac{e}{k_{agg.colloid(soil)} + k_{filtr.soil} + k_{leach} + k_{run-off} + k_{trans}} \frac{1}{k_{trans} + k_{run-off} + k_{leach}}$$

(73)

C6.1 Critical attachment efficiency for coarse attached fraction

The fraction of ENPs attached to soil coarse particles is dominant if:

$$m_{att.coarse\ in\ soil} > m_{free\ in\ soil} + m_{agg.colloids\ in\ soil}$$

(74)

Inserting eq 69 and 71 in equation 73 yields:

$$m_{free\ in\ soil} \frac{k_{filtr.soil}}{k_{erosion} + k_{trans}} > m_{free\ in\ soil} + m_{free\ in\ soil} \frac{k_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}}$$

(75)

which can be simplified to:

$$\frac{k_{filtr.soil}}{k_{erosion} + k_{dis}} > 1 + \frac{k_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}}$$

(76)

The first-order rate constant is derived by multiplying the attachment efficiency with the filtration frequency,

$$k_{filtr.soil} = \alpha f_{filtr.soil}$$

(77)

whereas the first-order rate constant for hetero-aggregation with natural colloids is derived by multiplying the attachment efficiency with the frequency of collisions with natural colloids:

$$k_{agg.colloid(soil)} = \alpha f_{agg.colloid(soil)}$$

(78)

The critical attachment efficiency at which the ENPs attached to coarse particles in soil are dominant can then be derived by inserting equation 76 and 77 in 75 as:

$$\frac{\alpha_{crit.att\ in\ soil} f_{filtr.soil}}{k_{erosion} + k_{dis}} = 1 + \frac{\alpha_{crit.att\ in\ soil} f_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}}$$

(79)

so that,

$$\alpha_{crit.att\ in\ soil} \frac{f_{filtr.soil}}{k_{erosion} + k_{trans}} = 1 + \alpha_{crit.att\ in\ soil} \frac{f_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}}$$

(80)

$$\alpha_{crit.att\ in\ soil} \frac{f_{filtr.soil}}{k_{erosion} + k_{trans}} - \alpha_{crit.att\ in\ soil} \frac{f_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}} = 1$$

(81)

$$\alpha_{crit.att\ in\ soil} \left(\frac{f_{filtr.soil}}{k_{erosion} + k_{trans}} - \frac{f_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}} \right) = 1$$

(82)

$$\alpha_{crit.att\ in\ soil} = \left(\frac{f_{filtr.soil}}{k_{erosion} + k_{trans}} - \frac{f_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}} \right)^{-1}$$

(83)

From the extracted data sheet it is derived that the critical attachment efficiency at which coarse attached ENPs are dominant in the soil compartment is by median 1. 10^{-6}. Once ENPs are attached to coarse particles in soil, transformation and erosion are the only removal processes. Transformation thus already becomes the dominant removal processes for coarse attached ENPs if:

$$k_{trans} > k_{erosion}$$

(84)

Hence, the critical transformation rate is equal to the erosion rate, which is by median 4 10^{-12} s^{-1}.

C 6.2 Predicted free concentration

$$C_{free\ in\ soil} = \frac{m_{free\ in\ soil}}{V_{soil}\rho_{soil}}$$

(85)

so that,

$$C_{free\ in\ soil} \sim m_{free\ in\ soil}$$

(86)

Based on eq 66, 76, 77 and 84 it is derived that the free concentration in soil is proportional to the inverse of the rate constants for the fate processes responsible for the removal of free ENPs in soil:

$$C_{free\ in\ soil} \sim \frac{1}{k_{leach} + k_{run-off} + k_{filtr.soil} + k_{agg.colloid(soil)}} \sim \frac{1}{\alpha\left(f_{filtr.soil} + f_{agg.colloid(soil)}\right) + k_{trans} + k_{leach} + k_{run-off}}$$

(87)

C 6.3 Predicted bioavailable concentration in soil

$$C_{bioavailable\ in\ soil} = \frac{m_{bioavailable\ in\ soil}}{V_{soil}\rho_{soil}} \tag{88}$$

$$m_{bioavailable\ in\ soil} = m_{free\ in\ soil} + m_{agg.colloids\ in\ soil} \tag{89}$$

$$m_{agg.colloids\ in\ soil} = m_{free\ in\ soil}\frac{k_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}} \tag{90}$$

Inserting eq 90 in eq 89 then yields:

$$m_{bioavailable\ in\ soil} = m_{free\ in\ soil} + m_{free\ in\ soil}\frac{k_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}} \tag{91}$$

so that:

$$m_{bioavailable\ in\ soil} = m_{free\ in\ soil}\left(1 + \frac{k_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}}\right)$$

Inserting eg 68, 78 and 79 in eq 89 then yields:

$$m_{bioavailable\ in\ soil} = \frac{e}{\alpha(f_{agg.colloids} + f_{filtration}) + k_{leach} + k_{run-off} + k_{trans}}\left(1 + \frac{\alpha f_{agg.colloids}}{k_{trans} + k_{run-off} + k_{leach}}\right) \tag{92}$$

$$m_{bioavailable\ in\ soil} = e\frac{1}{\alpha(f_{agg.colloid(soil)} + f_{filtr.soil}) + k_{leach} + k_{run-off} + k_{trans}}\left(\frac{k_{trans} + k_{run-off} + k_{leach} + \alpha f_{agg.colloid(soil)}}{k_{trans} + k_{run-off} + k_{leach}}\right) \tag{93}$$

$$m_{bioavailable\ in\ soil} = e\frac{\alpha f_{agg.colloid(soil)} + k_{trans} + k_{run-off} + k_{leach}}{(\alpha(f_{agg.colloid(soil)} + f_{filtr.soil}) + k_{leach} + k_{run-off} + k_{trans})(k_{trans} + k_{run-off} + k_{leach})} \tag{94}$$

Hence, inserting eq 94 in 88 proves that:

$$C_{bioavailable\ in\ soil} \sim \frac{\alpha f_{agg.colloid(soil)} + k_{trans} + k_{run-off} + k_{leach}}{(\alpha(f_{agg.colloid(soil)} + f_{filtr.soil}) + k_{leach} + k_{run-off} + k_{trans})(k_{trans} + k_{run-off} + k_{leach})} \tag{95}$$

C6.4: Non-bioavailable coarse attached concentrations in soil

$$C_{att.coarse\ in\ soil} = \frac{m_{att.coarse\ in\ soil}}{V_{soil}\rho_{soil}} \tag{96}$$

Inserting eq 76 and 77 in eq 68 then yields:

$$m_{att.coarse\ in\ soil} = e\ \frac{\alpha f_{filtr.soil}}{\alpha(f_{agg.colloid(soil)} + f_{filtr.soil}) + k_{leach} + k_{run-off} + k_{trans}} \frac{1}{k_{trans} + k_{erosion}} \tag{97}$$

Hence, inserting eq 96 in 97 proves that:

$$C_{att.coarse\ in\ soil} \sim \frac{\alpha f_{filtr.soil}}{\alpha(f_{agg.colloid(soil)} + f_{filtr.soil}) + k_{leach} + k_{run-off} + k_{trans}} \frac{1}{k_{trans} + k_{erosion}} \tag{98}$$

C7: References

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D: Nomenclature

Table D1. Nomenclature

Symbol	Description	Unit
C	concentration	g.m^{-3} ; g.kg^{-1}
$C_{bioavailable\ in\ air}$	Predicted concentration of bioavailable ENPs in air	g.m^{-3}
$C_{bioavailable\ in\ sed}$	Predicted concentration of bioavailable ENPs in sediment	g.kg^{-1}
$C_{bioavailable\ in\ soil}$	Predicted concentration of bioavailable ENPs in soil	g.kg^{-1}
$C_{bioavailable\ in\ water}$	Predicted concentration of bioavailable ENPs in water	g.m^{-3}
$C_{free\ in\ air}$	Predicted concentration of free ENPs in air	g.m^{-3}
$C_{free\ in\ sed}$	Predicted concentration of free ENPs in sediment	g.kg^{-1}
$C_{free\ in\ soil}$	Predicted concentration of free ENPs in soil	g.kg^{-1}
$C_{free\ in\ water}$	Predicted concentration of free ENPs in water	g.m^{-3}
$C_{total\ in\ air}$	Predicted concentration of total ENPs in air	g.m^{-3}
$C_{total\ in\ sed}$	Predicted concentration of total ENPs in sediment	g.kg^{-1}
$C_{total\ in\ soil}$	Predicted concentration of total ENPs in soil	g.kg^{-1}
$C_{total\ in\ water}$	Predicted concentration of total ENPs in water	g.m^{-3}
$Diff_{ENP}$	Diffusion coefficient of ENP	m^2.s^{-1}
$Diff_{fine.aerosol}$	Diffusion coefficient of fine aerosol	m^2.s^{-1}
e	emission volume	g.s^{-1}
f_i	fit for iteration with newly derived equation	-
$f_{agg.colloids}$	collision frequency between ENP and natural colloids prior to aggregation	s^{-1}
$f_{att.coarse}$	collision frequency between ENP and natural coarse particles prior to attachment	s^{-1}
$f_{filtr.}$	Filtration frequency between ENP and natural coarse particles prior to attachment	s^{-1}
F_{in}	Chemical mass inflow	g.s^{-1}
$F_{in.\ free\ in\ sed}$	Mass inflow of free ENPs in sediment	g.s^{-1}
F_{out}	Chemical mass outflow	g.s^{-1}
$F_{out.\ free\ in\ sed}$	Mass outflow of free ENPs in sediment	g.s^{-1}
$F_{settling.agg.colloids}$	Mass flow of ENPs hetero-aggregated with natural colloids settling to sediment	g.s^{-1}
$F_{settling.att.coarse}$	Mass flow of ENPs attached to natural coarse particles settling to sediment	g.s^{-1}
$F_{settling.free}$	Mass flow of free ENPs settling to sediment	g.s^{-1}
$FR_{agg.colloids\ in\ sed}$	The fraction of ENPs in sediment that are hetero-aggregated with natural colloids	(-)
$FR_{att.coarse\ in\ sed}$	The fraction of ENPs in sediment that are attached to natural coarse particles	(-)
$FR_{free\ in\ sed}$	The fraction of ENPs in sediment that are free	(-)
$FR_{agg.colloids\ in\ settling}$	The fraction of ENPs settling to sediment that are hetero-aggregated with natural colloids	(-)
$FR_{att.coarse\ in\ settling}$	The fraction of ENPs settling to sediment that are attached to natural coarse particles	(-)
$FR_{free\ in\ settling}$	The fraction of ENPs settling to sediment that are free	(-)

k	first-order rate constant for environmental fate process	s^{-1}
h_{water}	Water depth	m
$k_{agg.colloid}$	first-order rate constant for hetero-aggregation with natural colloids	s^{-1}
$k_{att.coarse}$	first-order rate constant for attachment to natural coarse particles	s^{-1}
k_{burial}	first-order rate constant for burial of sediments	s^{-1}
$k_{coag.fine}$	first-order rate constant for coagulation with fine aerosols	s^{-1}
$k_{coag.coarse}$	first-order rate constant for coagulation with coarse aerosols	s^{-1}
k_{trans}	first-order rate constant for transformation	s^{-1}
$k_{trans.crit}$	critical transformation rate constant	s^{-1}
$k_{dry\ depostion}$	first-order rate constant transport via dry deposition	s^{-1}
$k_{erosion}$	first-order rate constant erosion	s^{-1}
$k_{filtration}$	first-order rate constant for filtration	s^{-1}
k_{leach}	first-order rate constant for leaching from soil to deeper groundwater layers	s^{-1}
$k_{outflow\ air}$	first-order rate constant for transport by outflow of air to continental scale	s^{-1}
$k_{outflow\ water}$	first-order rate constant for transport by outflow of water to continental scale	s^{-1}
k_{resusp}	first-order rate constant for resuspension of sediments	s^{-1}
$k_{run\text{-}off}$	first-order rate constant soil run-off	s^{-1}
$k_{set.att\text{-}coarse}$	first-order rate constant for setting of ENPs attached to natural coarse particles	s^{-1}
$k_{set.agg\text{-}colloids}$	first-order rate constant for setting of ENPs hetero-aggregated with natural colloids	s^{-1}
$k_{set.free}$	first-order rate constant for setting of free ENPs	s^{-1}
$k_{wet\ depostion}$	first-order rate constant transport via wet deposition	s^{-1}
m	steady state chemical mass	g
$m_{agg.colloids\ in\ sed}$	Steady state mass of ENPs hetero-aggregated with natural colloids in sediment	g
$m_{agg.colloids\ in\ soil}$	Steady state mass of ENPs hetero-aggregated with natural colloids in soil	g
$m_{agg.colloids\ in\ water}$	Steady state mass of ENPs hetero-aggregated with natural colloids in water	g
$m_{att.coarse\ in\ sed}$	Steady state mass of ENPs attached to natural coarse particles in sediment	g
$m_{att.coarse\ in\ soil}$	Steady state mass of ENPs attached to natural coarse particles in soil	g
$m_{att.coarse\ in\ water}$	Steady state mass of ENPs attached to natural coarse particles in water	g
$m_{bioavailable\ in\ soil}$	Steady state mass of bioavailable ENPs in soil	g
$m_{free\ in\ sed}$	Steady state mass of free ENPs in sediment	g
$m_{free\ in\ sed,\ ktrans=crit}$	Steady state mass of free ENPs in sediment for ENPs with a transformation rate constant equal to the critical transformation rate constant	g
$m_{free\ in\ sed,\ ktrans=0}$	Steady state mass of free ENPs in sediment for non-transformable ENPs	g

$m_{free\ in\ soil}$	Steady state mass of free ENPs in soil	g
$m_{free\ in\ water}$	Steady state mass of free ENPs in water	g
$m_{total\ in\ sed}$	Total steady state mass of ENPs in sediment	g
$m_{total\ in\ water}$	Total steady state mass of ENPs in water	g
$N_{fine.aerosol}$	Number concentration of fine aerosols in air	m^{-3}
R^2	Coefficient of determination	-
r_{ENP}	radius of ENP	nm
$r_{fine.aerosols}$	radius of fine aerosols	nm
SS_{res}	sum of all squares for the residuals	-
SS_{tot}	total sum of all squares	-
v_{stokes}	Stokes settling velocity	$m.s^{-1}$
V	Volume of environmental compartment	m^3
V_{air}	Volume of air compartment	m^3
V_{soil}	Volume of soil compartment	m^3
V_{water}	Volume of water compartment	m^3
y_i	data point per iteration	-
$\bar{y}$	average of iteration data points	-
α	attachment efficiency	-
α_{crit}	critical attachment efficiency	-
ρ	Density	$kg.m^{-3}$
ρ_{soil}	Soil density	$kg.m^{-3}$
$\sum k_{free\ in\ air}$	sum of first-order processes of environmental processes removing free ENPs from air	s^{-1}
$\sum k_{set.total}$	sum of first-order processes for removal of ENPs by settling	s^{-1}

E: Natural variability of environmental system

Table E 1. Model parameter distribution reflecting the natural variability for system dimensions, advective flows, and natural particles in the compartments atmosphere, fresh water, soil, and sediment. The shape of the distribution are lognormal (L), triangular (T), uniform (U), Pareto (P), and Weibull (W).

Parameter name	Distri-bution shape	Default, Mean (L), Mode (T), Value (C), Scale (W)	Min (T,P) Location (L,W)	Max (T,P) Shape (W) St. Dev. (L)	Reference
System temperature ˚C	T	10	-5	30	Bakker et al., 2003
N nucleation aerosols ($\#.cm^{-3}$)	L	$1.4\ 10^{3}$	$-3.2\ 10^{3}$	$4.0\ 10^{2}$	Neususs et al., 2002*[1]
Diameter nucleation aerosols (µm)	L	$2\ 10^{-2}$	(-)	$1.6\ 10^{-1}$	Jaenicke et al., 1993
Density nucleation aerosols ($kg.m^{-3}$)	T	$1.3\ 10^{3}$	$5.0\ 10^{2}$	$1.5\ 10^{3}$	Kannosto et al., 2008
N accumulation aerosols($\#.cm^{-3}$)	L	$3.6\ 10^{3}$	$1.2\ 10^{3}$	$4.0\ 10^{3}$	Neususs et al., 2002
Diameter accumulation aerosols(µm)	L	$1.2\ 10^{-1}$	(-)	$2.2\ 10^{-1}$	Jaenicke et al., 1993
Density accumulation aerosols ($kg.m^{-3}$)	T	$1.5\ 10^{3}$	$1.1\ 10^{3}$	$2.0\ 10^{3}$	Kannosto et al., 2008
N coarse aerosols ($\#.cm^{-3}$)	L	2.1	$-1.3\ 10^{-1}$	3.4	Neususs et al., 2002
Diameter coarse aerosols (µm)	L	1.8	(-)	0.43	Jaenicke et al., 1993
Density coarse aerosols*[1] ($kg.m^{-3}$)	T	$1.6\ 10^{3}$	$1.6\ 10^{3}$	$1.9\ 10^{3}$	Neususs et al., 2002
Friction velocity ($m.s^{-1}$)	T	$2.5\ 10^{-1}$	$1.1\ 10^{-1}$	$4.0\ 10^{-1}$	Nho-Kim et al., 2004
Viscous : Total Drag ratio	U	0.27	¼	1/3	Slinn, 1982
Fraction of interception by large collectors (%)	U	5.5	1	10	Slinn, 1982
Small vegetation hair width (µm)	U	5	0	10	Slinn, 1982
Large vegetation collector radius (mm)	U	0.75	0.5	1	Slinn, 1982

Rain dry air ratio	U	(-)	$1\ 10^{-7}$	$3\ 10^{-7}$	Franco and Trapp, 2010
Wind speed ($m.s^{-1}$)	T	5.0	1.7	14	Bakker et al., 2003
Rainrate (mm.d-1)	T	2.0	$2\ 10^{-2}$	6.6	Bakker et al., 2003
NC number concentration in fresh water ($\#.m^{-3}$)	T	$1.7\ 10^{14}$	$4.8\ 10^{13}$	$4\ 10^{14}$	Gallego-Urrea et al., 2010
NC size in freshwater (nm)	P	(-) Default: 60	50	2000	Rosse and Loizeau, 2003
NC density in fresh water ($kg.m^{-3}$)	T	$1.3\ 10^{3}$	$1.1\ 10^{3}$	$2.5\ 10^{3}$	Velzeboer et al., 2014
SP number concentration in fresh water ($\#.m^{-3}$)	U	$3.6\ 10^{10}$	$9.2\ 10^{9}$	$6.3\ 10^{10}$	Praetorius et al., 2012
SP size in freshwater (µm)	L	5	(-)	0.6	Praetorius et al., 2012
SP density in fresh water ($kg.m^{-3}$)	U	$1.8\ 10^{3}$	$1.1\ 10^{3}$	$2.5\ 10^{3}$	Praetorius et al., 2012
Water depth (m)	W	3	2	15	Bakker et al., 2003
Water Flow ($m^3.s^{-1}$)	L	$2.3\ 10^{3}$		$8.9\ 10^{2}$	Bakker et al., 2003
Fresh water shear stress (s^{-1})	U	5	0	10	Praetorius et al., 2012
NC number concentration in soil pore water ($\#.m^{-3}$)	T	$7\ 10^{13}$	$3\ 10^{12}$	$4\ 10^{14}$	Rani et al., 2011
NC size in soil pore water (nm)	T	$1.2\ 10^{2}$	20	$4\ 10^{2}$	Citeau et al, 2006
NC density in soil pore water ($kg.m^{-3}$)	U	(-)	2000	2700	Citeau et al, 2006
Soil grain radius (µm)	T	$1.3\ 10^{2}$	62	$2.9\ 10^{2}$	Cornelis et al., 2013
Soil pore water filtration velocity ($m.s^{-1}$)	T	$7\ 10^{-6}$	$1\ 10^{-4}$	$2\ 10^{-3}$	Schwartz et al., 2000; Tufenkji andElimelech, 2004
Soil Erosion (mm.y-1)	T	$3\ 10^{-2}$	$7.5\ 10^{-4*2}$	$6\ 10^{-2}$	Bakker et al., 2003
FR rainwater run-off (-)	T	0.25	$6.3\ 10^{-3*2}$	0.5	Bakker et al., 2003
FR rainwater infiltration (-)	T	0.25	$6.3\ 10^{-3*2}$	0.5	Bakker et al., 2003

Soil porosity	T	0.20	$3\ 10^{-3}$	0.67	Bakker et al., 2003
Density of soil solids ($kg.m^{-3}$)	T	$2.5\ 10^{3}$	$2.0\ 10^{3}$	$3.0\ 10^{3}$	Bakker et al., 2003
Sediment grain radius(µm)	T	$1.3\ 10^{2}$	6	$2.0\ 10^{2}$	Velzeboer et al., 2014; Jones and Su, 2012
Sediment water filtration velocity ($m.s^{-1}$)*2	U	(-) Default: $5\ 10^{-7}$	10^{-9}	10^{-6}	Higashino et al., 2009
Sediment depth (m)	T	$3\ 10^{-2}$	$1\ 10^{-2}$	0.1	Bakker et al., 2003
Sediment porosity	T	0.8	0.5	0.99	Bakker et al., 2003
Sediment resuspension ($m.h^{-1}$)	T	$1.1\ 10^{-8}$	0	$2.3\ 10^{-8}$*3	Praetoriues et al., 2012
Sediment burial rate ($m.y^{-1}$)	T	$2.7\ 10^{-3}$	$1\ 10^{-3}$	$5\ 10^{-2}$	Bakker et al., 2003; Koelmans et al., 2009

*1 Calculated from raw data of Neususs et al., 2002

*2 Minimal value assumed to be 2.5% of median

*3 Maximum value assumed to be 2 times larger than median

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