Supplementary Information

Bayesian Network as Support Tool for Rapid Query of the Environmental Multimedia Distribution of Nanomaterials

Muhammad Bilal¹, Haven Liu¹, Rong Liu¹, Yoram Cohen^{1,*}

Co	ntents	Page
1.	Bayesian Networks	S2
2.	BN Model development for the assessment of ENMs exposure modeling	S 2
3.	Conditional dependence of ENM environmental concentrations on input parameters	S 9
4.	Quantification of Parameter Significance in Predicting ENM Environmental Distribution	S13
Su	oplementary Information References	S18
Fig	ure S1: Bayesian Network construction for ENM environmental distribution	S5
Fig	ure S2: ENMs compartmental concentrations estimated by MendNano	S 6
Fig	ure S3: ENMs release rates in high scenarios estimated by LearNano	S 7
Fig sce	sure S4: Correlation of determination (R^2) for ENMs concentrations in high narios	S 8
Fig reg	ure S5-10: Conditional dependence of ENMs concentrations on parameters for ion	S9-12
Fig	ure S11: Impact of (a) TiO ₂ and (b) CeO ₂ ENM attachment to suspended solids	S13
(in	water) on ENM concentrations in water and sediment.	
Tal	ble S1: Parameter sensitivity analysis using Alexander's sensitivity indicator	S4
Tal	ble S2-S9. Parameters for ENMs simulations of environmental distribution in	S14-17
dif	ferent regions	
Tal	ble S10: ENMs releases to air, water and soil for low and high	S 19
Tal	ble S11: Description of exposure model input parameters	S20

1 1. Bayesian Networks

Bayesian Networks (BNs) belong to the family of probabilistic graphical models that are useful for 2 representing knowledge and reasoning while considering uncertainty. BNs development involves the 3 construction of a directed acyclic graph (DAG) of available information (parameters) treated as nodes 4 in the network. The influence/conditional dependence relations among these nodes are represented 5 6 qualitatively as arcs between nodes and quantitatively by conditional probability tables (CPTs). In the present work for situations where data are available (e.g. geographical, meteorological parameters, 7 release rates and concentrations), the BNs were utilized by constructing network structure and learning 8 9 CPTs using available information for ENMs exposure modeling scenarios. The nodes of continuous/numerical values in BNs are usually discretized into predefined sized ranges called states 10 (e.g., windspeed can be discretized into the states of distinct ranges). In the particular case of BN for 11 exposure modeling, an arc from a node (such as windspeed (m/s)) to a child node (such as air 12 concentration (ng/m³)) represents the conditional dependence of concentration on windspeed and 13 windspeed is called the parent node of concentration. Thus the arrow indicates that an assigned 14 windspeed directly influences air concentration (manuscript Fig. 3). The complexity of BN is dictated 15 by the dimensionality of the conditional probability tables (constructed based on causal links among 16 parameters)¹⁻⁴ as the size of a CPT of a node in BN is determined by the number of incoming links 17 (parent nodes), the number of states of each parent node, and the number of states of the node itself. 18

19

20 2. BN Model development for the assessment of ENMs exposure modeling

In order to develop a data driven predictive model (BN-nanoExpo) for the assessment of ENMs exposure, the selection of parameters for a range of scenarios was accomplished by first designing a structure representing the BN nodes relationships (i.e. parameter-concentration). Given the tradeoff 24 between increased model accuracy and the desire for increased generalization, an initial pool of parameters to be included in BN-nanoExpo model was selected based on the knowledge derived from 25 the fundamental fate and transport model⁵. The resulting 18 parameter-set (including the initial pool) 26 was used in sensitivity analysis using Alexander's sensitivity indicator⁶ in order to guide the selection 27 of final parameter-set for model development. Using Alexander's indicator, each parameter was varied 28 29 sequentially (from minimum to maximum) and the sum of the squared differences for the resulting concentration vectors (air, water, soil and sediment) was calculated (Table S1). The range of the 30 differences was from 0 - 1 indicating 1 as the maximum possible sensitivity or effect on resulting 31 32 concentrations, where the cutoff threshold was set to 0.97 (any parameter with sensitivity indicator \geq 0.03) for the selection of parameters in final pool. The indicator's measure of sensitivity of the 33 34 concentrations to changes in model parameters is given by

35
$$S = \frac{\sum_{i=1}^{N} \frac{(o_{ik} - o_{ij})^2}{o_{ij}}}{\sum_{i=1}^{N} \frac{(o_{ik} - o_{ij})^2}{o_{ij}}}$$
(1)

in which, O_{ij} is the concentration of unit *i* {air, water, soil, sediment} with variable at (previous/minimum) value *j*, O_{ik} is the concentration of unit *i* with variable at (changed/maximum) value *k*, and *N* is the number of units.

The sensitivity analysis suggested the ranking of the BN-nanoExpo parameters listed in (**Table S1**). It is noted that the indicator values are subject to the ranges selected for parameters. The top five parameters (in the order of decreasing significance) were ENM releases (to air, water, soil), rainfall, and windspeed. Temperature, land and water areas were also of significance (sensitivity indicator > 0.03).

The quantification of parameter-concentration relationships in BN-nanoExpo was obtained by learning the CPT of each node using the exposure data training set. The use of a mechanistic model (MendNano) for estimating the environmental distribution of ENMs provided the estimated

- 46 concentrations at regular time intervals over a one-year simulation period. As an illustration, the
- 47 predicted compartmental concentrations of TiO₂ in Bern (Switzerland) (with size distribution in air given

48 as $(\mu = 179$ nm)) are shown in **Fig. S1**.

- Sensitivity Minimum indicator **Parameter** Unit Maximum Atmospheric mixing height 300 2000 10^{-3} meters 10-3 Soil (top layer) depth inches 1 8 5 10-3 Water body depth 1 meters 10^{-3} Soil bulk density g/cm^3 1.1 1.65 1.1×10^{-3} Sediment bulk density g/cm³ 1.1 2.5 1.2×10^{-3} Suspended solids density g/cm³ 1.5 1.65 NP attachment factor (air) 1.8×10^{-3} (%) 0 100 NP attachment factor (water) 0 1.8×10^{-3} (%) 100 3×10⁻³ Atmosphere convective residence time hour 1 50 4×10⁻³ Water convective residence time hour 1 50 km² 7.3×10⁻² Air-soil interfacial area 1 1200 5.5×10⁻² Air-water interfacial area km² 1 100 3×10^{-2} Average monthly air temperature С 40 1 3×10⁻² Average monthly wind speed m/s 1 8 3×10⁻² Average monthly rainfall mm/month 10 1000 Release rate to air 50 0.11 kg 1000 Release rate to water kg 100 1000 0.12 1000 Release rate to soil 50 1 kg
- 49 Table S1: Parameter sensitivity analysis using Alexander's sensitivity indicator

50

Temporal ENM media concentrations from MendNano were provided for a typical period of 365 days (1 sample per hour, **Fig-S1**). In principle, temporal ENMs' media concentrations can be modeled by the variants of BNs called dynamic Bayesian networks (DBNs) which provide a versatile approach to model the temporal system dynamics and prediction of the system state (e.g., ENMs concentrations) at a future time-step. DBNs are a special case of a singly connected BN aimed at time-series data analysis, where identical BN sub-models are duplicated over each time-step and the links between model parameters, as well as time slices that can change according to the system state at each time slice.



Figure S1: Temporal profile of TiO₂ environmental concentrations in Bern (Switzerland) starting with
clean environment. Following a well-established concept of annual cumulative distribution, the
concentrations for one-year simulation time period were utilized to construct the BN conditional
probability tables (CPTs).

63

58

The environmental distributions of ENMs for low releases (in air, water, soil and sediment) were 64 estimated using MendNano for both the training and test sets (Experimental Section). The resulting 65 concentrations from the training set served to construct the BN-nanoExpo model. The adequacy of the 66 resulting BN was then assessed via correlation analysis of predicted MendNano estimated 67 concentrations and the predicted concentrations by BN-nanoExpo for test set. The log10 transformed 68 69 values of the environmental concentrations for the test set are shown in Fig. S2 as heatmaps. The results revealed that the lowest estimated exposure concentrations in air (Fig. S2 (a)) and water (Fig. S2 (b)) 70 were for nano Cu (1) in air in Houston (-2.63 (2.4×10^{-3} ng/m³)), and (2) in water in New York (-3.46 71 $(3.5 \times 10^{-4} \text{ ng/L}))$. Exposure concentrations for TiO₂ and ZnO in air and water were relatively higher. 72 The highest exposure concentrations in air (atmosphere) was for TiO₂ in Berlin (1.81 (~65 ng/m³)) and 73 the highest exposure concentration in water was for TiO₂ in Bern (1.97 (~94 ng/L)). 74

	Al ₂ O ₃	CeO ₂	Cu	SiO ₂	TiO ₂	ZnO	1.81	Al ₂ O ₃	CeO ₂	Cu	SiO2	TiO ₂	ZnO	1.97
Los Angeles	0.63	0.07	-2.20	1.10	1.48	0.93		0.11	-1.11	-3.16	-0.25	1.26	0.64	
New York	0.85	0.29	-1.98	1.31	1.69	1.13		-0.23	-1.38	-3.46	-0.50	0.91	0.29	
Houston	0.22	-0.33	-2.63	0.69	1.08	0.52		-0.06	-1.27	-3.32	-0.42	1.09	0.47	
Wayne	-1.50	-2.09	-1.48	-1.03	-0.64	-1.22		-0.40	-1.60	-0.83	-0.78	0.75	0.13	
Maricopa	0.04	-0.52	-0.85	0.51	0.90	0.34		-2.56	-3.17	-2.90	-2.22	-1.84	-2.39	
Bern	0.68	0.16	-2.24	1.16	1.53	1.02		0.83	-0.36	-2.44	0.51	1.97	1.36	
Berlin	0.94	0.37	-1.88	1.39	1.81	1.26		0.49	-0.74	-2.78	0.10	1.64	1.02	
London	0.87	0.27	-1.98	1.29	1.75	1.18		0.75	-0.50	-2.53	0.33	1.90	1.28	
-2.63						-2.63						-	3.46	
			(a)					(b)						
							2.11							1.99
Los Angeles	0.83	0.27	-1.93	1.32	1.64	1.03		0.13	-1.05	-3.12	-0.18	1.28	0.66	
New York	1.30	0.75	-1.44	1.80	2.11	1.48		-0.17	-1.24	-3.37	-0.32	0.95	0.34	
Houston	0.48	-0.07	-2.27	0.98	1.30	0.69		-0.03	-1.23	-3.29	-0.37	1.11	0.49	
Wayne	-1.24	-1.80	-1.15	-0.74	-0.42	-1.03		-0.38	-1.61	-0.81	-0.76	0.77	0.15	
Maricopa	0.10	-0.46	-0.73	0.59	0.91	0.30		-2.17	-2.74	-2.98	-1.71	-1.28	-1.81	
Bern	0.82	0.29	-1.98	1.32	1.64	1.07		0.85	-0.33	-2.43	0.54	1.99	1.37	
Berlin	1.22	0.66	-1.55	1.71	2.04	1.43		0.52	-0.69	-2.74	0.16	1.67	1.05	
London	1.07	0.50	-1.70	1.55	1.90	1.29		0.77	-0.47	-2.51	0.37	1.92	1.30	
-2.27 (C)							(d	I)		-	3.37			





79 The environmental concentrations of ENMs estimated by MendNano in soil and sediment (Fig. S2 (c, d)) were also reported as heatmaps. The lowest estimated compartmental exposure concentrations in 80 both soil (Fig. S2 (c)) and sediment (Fig. S2 (d)) were of nano Cu which were; (1) in soil in Houston (-81 2.27 (5.3 \times 10⁻³ µg/kg)), and (2) in sediment in New York (-3.37 (4.3 \times 10⁻⁴ µg/kg)). Exposure 82 83 concentrations for TiO₂ and ZnO in soil and sediment were higher among the six ENMs due in part to 84 the higher ENM release rates. The highest exposure concentration of TiO₂ in sediment in Bern of ~98 85 μ g/kg (1.99), was likely due to higher release rates as well as higher exposure concentration in water in 86 the above region (as there is a causal relationship of concentration in water with the concentration in 87 sediment). The low release scenarios, estimated using LearNano (Fig. S3a) with ENMs apportionment air, water and soil (Fig. S3(b)), demonstrated the highest overall ENM releases in New York, London, 88 89 and Berlin (in decreasing order) and the lowest overall ENMs releases were in Wayne and Maricopa.







92

Figure S3 (b): Stacked bar of low estimates of ENM releases to air, water and soil as percentage of the total ENM release for the indicated cities. The total releases (kg/year) for each ENM are reported below the bar charts. Note that the estimated release rates of SiO₂, TiO₂ and ZnO are higher in all cities compared to the release rates of other selected ENMs.

Given the low estimated of release rates of ENMs in the selected cities, BN-nanoExpo predictions demonstrated excellent correlation of R^2 of 0.97, 0.94, 0.96, and 0.95 with MendNano simulations for air, water, soil, and sediment respectively (**Fig. S4**). The correlations between MendNano estimations and BN-nanoExpo predictions indicate that the cause-effect relationships were adequately represented by the BN-nanoExpo model which provide a basis for interrogating the conditional dependence of ENMs multimedia concentrations on model parameters.



Fig. S4: Observed MendNano estimations ($\log(\mu g/\kappa g)$) along with R^2 for all four compartments (a) air, (b) water, (c) soil, and (d) sediment.

3. Conditional dependence of ENMs concentrations on input parameters

In data driven modeling approaches, the complexity and dimensionality of information poses a 107 108 challenge for a model to; (i) visually integrate parameters of different types, (ii) represent the conditional dependence of various parameters, and (iii) investigate the effects of subsets of parameters on the 109 outcomes of interest. In this regard, BNs are especially advantageous since they enable the visualization 110 111 of the conditional parameter-parameter and/or parameter-outcome dependences. Using BNs, one can select a partial line of evidence and assess the impact of selected subset on target outcome. Specifically, 112 the BN model rapid assessment of the impact of uncertainty in multiple parameters either individually 113 or simultaneously expressed as normal distributions about the mode, on the resulting compartmental 114 concentrations. As an illustration of the above a number of test cases are shown in **Figs. S5** – **S10** for 115 Al₂O₃, CeO₂, Cu, SiO₂, TiO₂ and ZnO whereby the impact of uncertainties in multiple parameters 116 (expressed in terms of normal distributions) was evaluated with respect to the resulting distributions of 117 the ENMs concentrations in the various environmental media. 118



Figure S5: Predicted compartmental concentrations of Al_2O_3 for the range of parameter values. The conditional dependence relationships demonstrate that lower releases of Al_2O_3 (air = 60kg, water = 360kg, soil = 270kg) in land area = 1220km² and water area = 30 km² resulted in lower compartmental concentrations than those of TiO₂, SiO₂, and ZnO (Fig. S6-S9).





Figure S6: Distribution of predicted compartmental concentrations of TiO₂ for the range of parameter values (i.e., parameter uncertainty).

126 127



128

Figure S7: Distribution of predicted compartmental concentrations of TiO₂ for the range of parameter values (i.e., uncertainties). The effect of higher releases of TiO₂ on compartmental concentrations is shown as their probability distributions. Compartmental concentrations of TiO₂ (air \approx 75 ng/m³, water \approx 85 ng/L, soil \approx 120 µg/kg, sediment \approx 75 µg/kg) resulted higher (compared to **Fig. S6**) due to higher release rates (air = 980kg, water = 2.3×10³, soil=2.3×10³) and updated geographical parameters (land area = 1500km², water area = 0.2 km²).



Figure S8: Distribution of predicted compartmental concentrations of ZnO for the range of parameter

137 values (uncertainties).



Figure S9: Distribution of predicted compartmental concentrations of SiO₂ for the range of parameter values (uncertainties).



Figure S10: Distribution of predicted compartmental concentrations of nano-Cu for the range of parameter values (uncertainties).

144

141

It is important to recognize that ambient particles are present (in both air and water) at significantly 145 146 higher number concentrations relative to those which may be expected based solely on potential releases of ENMs⁷. Therefore, ENMs are likely to be associated with ambient particulates (due to various 147 surface–surface interactions⁸) given the high available surface area of ambient particles and tendency of 148 most ENMs to agglomerate $^{9-11}$. The particle size of ambient aerosols typically ranges from 0.001 to 2 149 um with particle size distribution (PSD) typically described by a trimodal log-normal size 150 distribution¹¹. Suspended solids in natural water bodies are typically in the size range of $0.01-1 \mu m$ for 151 lakes¹², $1-100 \,\mu\text{m}$ for oceans¹³, and $30-150 \,\mu\text{m}$ for rivers¹⁴, and log-normal size distributions have been 152 often reported⁷, with a concentration range that can vary significantly (30 μ g L-1-200 mg L-153 1)^{15,16} depending on the specific water body. Here we note that previous work has shown that under 154 most conditions essentially all ENMs would be attached to ambient particles⁷. The extent of 155

heteroaggregation, however, can be quantified via an attachment efficiency or an attachment factor⁵, the latter representing the approach followed in the present work. Figure **S11** provides an example of the impact of the attachment factor impact on the concentrations of (a) TiO₂ and (b) CeO₂ in water and sediment (for Houston (United States)) with emission rates estimated by LearNano^{5,18} along with the relevant geographical and meteorological parameters. As the attachment factor increases from 0.75 to 1, the ENMs concentration (as suspended matter) in water decreases, while the concentration in the sediment increases.



163

Figure S11. Impact of (a) TiO₂ and (b) CeO₂ ENM attachment to suspended solids (in water) on ENM
 concentrations in water and sediment in Houston.

167 4. Quantification of Parameter Significance in Predicting ENM Environmental Distribution

168 The BN-nanoExpo model allows rapid assessment of the distribution of compartmental 169 concentrations as impacted by input parameter uncertainty. BN sensitivity analysis allows one to 170 determine the influence of input parameters on the resulting ENM environmental concentrations. Here, 171 sensitivity analysis was conducted via exhaustive search whereby each parameter was sequentially varied to quantify its impact, on the predicted ENM concentrations, in terms of the reduction in variance of predicted outcome. Accordingly, the parameter that reduces the variance of the target outcome to the largest degree is considered most significant. Accordingly, the reduction in variance of ENM environmental concentrations was quantified as the square of the Root Mean Square (RMS) change in ENM concentration given as follows

$$Vr = V(Q) - V(Q|f)$$
⁽²⁾

where

$$V(Q) = \sum_{q} P(q) [Xq - E(Q)]^{2}$$
(3)

$$V(Q|f) = \sum_{q} P(q|f) [Xq - E(Q|f)]^{2}$$
(4)

and
$$E(Q) = \sum_{q} P(q) X q$$
 (5)

where *Q* is the query node (ENM compartmental concentration), *F* is varying node (input parameter), *q* is the state of *Q*, *f* is a state of *F*, *Xq* is the numeric real value corresponding to state *q*, *V*(*Q*) is the variance of the real value of *Q* without any new evidence, *V*(*Q*|*f*) is the variance of the real value of *Q* with evidence *f* at *F*, *E*(*Q*) is the expected real value of *Q* without any evidence and *E*(*Q*|*f*) is the expected real value of *Q* given evidence *f* for node *F*.

 182
 Table S2: Parameters for simulation of environmental distribution of ENMs in Berlin (Germany)

 Parameter
 Parameter value

Air-soil interfacial area	832	km ²
Air-water interfacial area	59.7	km ²
Average rainfall	49.7	mm/month
Average wind speed	4.3	m/s
Average air temperature	9	С
Release rate to air	[64, 15, 0.05, 105, 665, 236]	kg/year
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]		
Release rate to water	$[410.5,23, 0.21, 156, 5.8 \times 10^3, 1.4 \times 10^3]$	kg/year
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]		
Release rate to soil	[389, 108.5, 0.7, 1.3×10 ³ , 2.5×10 ³ , 518]	kg/year
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]		

184

183

Parameter	Parameter value				
Air-soil interfacial area	51	km ²			
Air-water interfacial area	0.6	km ²			
Average rainfall	86	mm/month			
Average wind speed	1.6	m/s			
Average air temperature	9.4	С			
Release rate to air	[3, 0.9, 3×10 ⁻³ , 8.1, 23.3, 9]	kg/year			
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]					
Release rate to water	$[8.5, 0.5, 4.5 \times 10^{-3}, 4, 119, 28.6]$	kg/year			
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]					
Release rate to soil	$[8, 2.2, 1.4 \times 10^{-2}, 26, 48, 10.5]$	kg/year			
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]					

Table S3: Parameters for simulation of environmental distribution of ENMs in Bern (Switzerland)

Table S4: Parameters for simulation of environmental distribution of ENMs in Houston (USA)

Parameter	Parameter value					
Air-soil interfacial area	1553	km ²				
Air-water interfacial area	72.3	km ²				
Average rainfall	114	mm/month				
Average wind speed	3.4	m/s				
Average air temperature	21.3	С				
Release rate to air [Al2O3, CeO2, Cu, SiO2, TiO2, ZnO]	$[26, 7, 2.3 \times 10^{-2}, 61, 232, 80.5]$	kg/year				
Release rate to water [Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]	[140.7, 8.3, 7.4×10 ⁻² , 58.3, 1.9×10 ³ , 476]	kg/year				
Release rate to soil [Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]	[125, 35, 0.24, 405.3, 758, 166.3]	kg/year				

Table S5: Parameters for simulation of environmental distribution of ENMs in London (UK)

Parameter	Parameter value					
Air-soil interfacial area	1572	km ²				
Air-water interfacial area	10-4	km ²				
Average rainfall	48.6	mm/month				
Average wind speed	3.6	m/s				
Average air temperature	11	С				
Release rate to air [Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]	[89.7, 18.3, 7×10 ⁻² , 132, 980, 319]	kg/year				
Release rate to water [Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]	$[489, 27.1, 0.25, 184, 6.9 \times 10^3, 1.6 \times 10^3]$	kg/year				
Release rate to soil [Al2O3, CeO2, Cu, SiO2, TiO2, ZnO]	[489, 136.3, 0.9, 1.6×10 ³ , 2.9×10 ³ , 651.3]	kg/year				

Parameter	Parameter value				
Air-soil interfacial area	1213.9	km ²			
Air-water interfacial area	88.1	km ²			
Average rainfall	32	mm/month			
Average wind speed	2.12	m/s			
Average air temperature	19	С			
Release rate to air	$[46, 12.4, 4.2 \times 10^{-2}, 107.5, 410.5, 142]$	kg/year			
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]					
Release rate to water	$[249, 14.8, 0.13, 103, 3.5 \times 10^3, 841.4]$	kg/year			
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]					
Release rate to soil	$[220.7, 61.6, 0.4, 717, 1.3 \times 10^3, 294]$	kg/year			
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]					

Table S6: Parameters for simulation of environmental distribution of ENMs in Los Angeles (USA) 193

194

195

196

197	Table S7: Parameters for simulation of environmenta	l distribution of ENMs in Maricopa (USA)
	Parameter	Parameter value

Air-soil interfacial area	76.4	km ²
Air-water interfacial area	0.16	km ²
Average rainfall	13.4	mm/month
Average wind speed	2	m/s
Average air temperature	21	С
Release rate to air	$[0.5, 0.14, 4.1 \times 10^{-2}, 1.3, 4.8, 1.7]$	kg/year
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]		
Release rate to water	[2.9, 0.2, 0.13, 1.2, 41, 9.85]	kg/year
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]		
Release rate to soil	[2.6, 0.72, 0.42, 8.4, 15.7, 3.4]	kg/year
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]		

198

Table S8: Parameters for simulation of environmental distribution of ENMs in New York (USA) 199 **Parameter Parameter value**

Air-soil interfacial area	783.8	km ²
Air-water interfacial area	429.5	km ²
Average rainfall	98.5	mm/month
Average wind speed	5.4	m/s
Average air temperature	11.4	С
Release rate to air	[99, 27, 9×10 ⁻² , 233, 888, 308]	kg/year
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]		
Release rate to water	$[539, 32, 0.3, 223, 7.6 \times 10^3, 1.8 \times 10^3]$	kg/year
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]		
Release rate to soil	$[477, 133, 0.9, 1.5 \times 10^3, 2.9 \times 10^3, 637]$	kg/year
[Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]		

200

Parameter	Parameter value				
Air-soil interfacial area	204.5	km ²			
Air-water interfacial area	0.4	km ²			
Average rainfall	70.2	mm/month			
Average wind speed	4.4	m/s			
Average air temperature	9.8	С			
Release rate to air [Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]	[6.5×10 ⁻² , 2×10 ⁻² , 4.2×10 ⁻² , 0.15, 0.6, 0.2]	kg/year			
Release rate to water [Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]	$[0.35, 2 \times 10^{-2}, 0.13, 0.14, 5, 1.2]$	kg/year			
Release rate to soil [Al ₂ O ₃ , CeO ₂ , Cu, SiO ₂ , TiO ₂ , ZnO]	[0.3, 8×10 ⁻² , 0.4, 1, 2, 0.42]	kg/year			

201 Table S9: Parameters for simulation of environmental distribution of ENMs in Wayne (USA)

Supplementary Information References

- 1. Bhanja S, Sarkar S. Probabilistic Modeling of QCA Circuits Using Bayesian Networks. IEEE Transactions on Nanotechnology. 2006;5(6):657–670.
- 2. Campos CP De, Ji Q. Efficient Structure Learning of Bayesian Networks using Constraints. Journal of Machine Learning Research. 2011;12:663–689.
- 3. Vidakovic B. Probability, Conditional Probability and Bayes Formula. 2004:1–13.
- 4. Neapolitan RE. Learning Bayesian Networks. Molecular Biology. 2003;6(2):674.
- 5. Liu HH, Cohen Y. Multimedia environmental distribution of engineered nanomaterials. Environmental Science and Technology. 2014;48(6):3281–3292.
- 6. Alexander ER. Sensitivity Analysis in Complex Decision Models. Journal of the American Planning Association. 1989;55(3):323–333.
- Praetorius A, Scheringer M, Hungerbühler K. Development of environmental fate models for engineered nanoparticles - A case study of TiO2 nanoparticles in the Rhine River. Environmental science & technology. 2012;46:6705–6713.
- 8. Liu HH, Surawanvijit S, Rallo R, Orkoulas G, Cohen Y. Analysis of Nanoparticle Agglomeration in Aqueous Suspensions via Constant-Number Monte Carlo Simulation. Environmental Science & Technology. 2011;45(21):9284–9292.
- Petosa AR, Jaisi DP, Quevedo IR, Elimelech M, Tufenkji N. Aggregation and Deposition of Engineered Nanomaterials in Aquatic Environments: Role of Physicochemical Interactions. Environmental Science & Technology. 2010;44(17):6532–6549.
- Arvidsson R, Molander S, Sandén BA, Hassellöv M. Challenges in Exposure Modeling of Nanoparticles in Aquatic Environments. Human and Ecological Risk Assessment: An International Journal. 2011;17(1):245–262.
- 11. John H. Seinfeld SNP. Atmospheric Chemistry and Physics: From Air Pollution to Climate Change. John Wiley & Sons, Ltd.; 2006.
- Walther C, Büchner S, Filella M, Chanudet V. Probing particle size distributions in natural surface waters from 15 nm to 2 μm by a combination of LIBD and single-particle counting. Journal of Colloid and Interface Science. 2006;301(2):532–537.
- Reynolds RA, Stramski D, Wright VM, Woźniak SB. Measurements and characterization of particle size distributions in coastal waters. Journal of Geophysical Research: Oceans. 2010;115(C8):n/a–n/a.
- 14. Walling DE, Moorehead PW. The particle size characteristics of fluvial suspended sediment: an overview. Hydrobiologia. 1989;176(1):125–149.
- 15. Jacobs MB, Ewing M. Suspended Particulate Matter: Concentration in the Major Oceans. Science. 1969;163(3865):380 LP 383.
- 16. Fitzgerald, B. C., Susfalk, R. B., Knust AM. Characterization of turbidity and total suspended solids in the Upper Carson River, Nevada. 2008.
- 17. Bhanja S, Sarkar S. Probabilistic Modeling of QCA Circuits Using Bayesian Networks. IEEE Transactions on Nanotechnology. 2006;5(6):657–670.
- 18. Keller AA, McFerran S, Lazareva A, Suh S. Global life cycle releases of engineered nanomaterials. Journal of Nanoparticle Research. 2013;15(6).

City	Compartment	ENMs Release (kg/yr) (low – high)					
		Al ₂ O ₃	CeO ₂	Cu	SiO ₂	TiO ₂	ZnO
	Air	64 - 290	15 - 66	0.05 - 1.4	105 - 458	665 - 1,210	236 - 520
Berlin	Water	411 - 5,619	23 - 704	0.2 - 24	156 - 4,287	5,827 - 33,181	1,393 – 7,932
	Soil	389 - 4,429	108 - 876	0.7 - 20	1,263 - 7,487	2,361 - 16,334	518 - 3,272
P	Air	3 - 192	0.9 - 55	0.003 - 1.1	8-514	23 - 494	9 - 193
Bern	Water	9 - 229	0.5 - 47	0.005 - 1.15	4 - 400	119 - 960	29 - 272
	Soil	8-90	2 - 18	0.01 - 0.4	26 - 152	48 - 331	11 - 66
TT (Air	26-1,076	7 - 301	0.02 - 6	61 – 2,814	232 - 2,857	81 - 1,110
Houston	Water	141 - 2,212	8 - 372	0.07 - 10	58 - 2,851	1,985 - 11,047	476 – 2,858
	Soil	125 – 1,421	35 - 281	0.24 - 6	405 - 2,402	758 - 5,241	166 - 1,050
. .	Air	90 - 455	18 - 96	0.07 - 455	132 - 692	980 - 1,954	319 - 725
London	Water	488 - 6,774	27 - 850	0.25 - 6,774	184 - 5,190	6,938 - 39,976	1,656 – 9,554
	Soil	489 - 5,566	136 - 1,101	1 - 5,566	1,587 – 9,408	2,967 - 20,526	651 - 4,112
.	Air	46 - 1,903	12 - 533	0.04 - 11	108 - 4,978	410 - 5,054	142 - 1,963
Los Angeles	Water	249 - 3,912	15 - 659	0.13 - 18	103 - 5,043	3,512 - 19,541	841 - 5,055
	Soil	221 - 2,513	62 - 497	0.42 - 11	717 – 4,249	1,340 – 9,271	294 - 1,857
	Air	1 - 22	0.1 - 7	0.04 - 0.13	1.3 - 60	4.8 - 60	1.7 - 23
Maricopa	Water	3 - 46	0.2 - 8	0.13 - 0.21	1.2 - 60	41 - 229	10 - 60
	Soil	3 – 29	1 - 6	0.42 - 1.3	8 - 50	16 - 109	3.4 - 22
NT N7 1	Air	99 - 4,118	27 - 1,153	0.1 - 23	233 - 10,773	888 - 10,936	308 - 4,249
New York	Water	539 - 8,467	32 - 1,425	0.3 - 40	223 - 10,915	7,599 - 42,288	1,821 - 10,940
	Soil	478 – 5,439	133 – 1,076	1 - 24	1,551 - 9,196	2,900 - 20,062	637 – 4,019
	Air	0.1 – 3	0.02 - 0.8	0.04 - 0.15	0.2 - 7	0.6 - 7	0.2 - 3
Wayne	Water	0.4 - 6	0.02 - 1	0.13 - 0.3	0.1 - 7	5 - 28	1.2 - 7
	Soil	0.3 - 4	0.09 - 0.7	0.42 - 0.62	1 - 6	2 - 13	0.4 - 3

Table S10: ENMs releases to air, water and soil for low and high scenarios (based on Fig. 2 and Fig. S3)

Table S11. Description of ENM Exposure Model Input Parameters

Attributes	Unit	Description
ENM release to air	kg/year	Estimated ENM emission in air per year in a specific region
ENM release to water	kg/year	Estimated ENM emission in water per year in a specific region
ENM release to soil	kg/year	Estimated ENM emission in soil per year in a specific region
Monthly rainfall (average)	mm/month	Average monthly rainfall (in millimeters) in a selected region
Monthly temperature (average)	°C	Average monthly temperature (°C) in a selected region
Monthly windspeed (average)	(m/s	Average monthly windspeed (meters/second) in a selected region
Land area	km ²	air to soil interfacial area of a selected region
Water area	km ²	Air to water interfacial area of a selected region
ENM Concentration (air)	ng/m ³	Estimated ENM concentration in air. Affected by {release (air), temperature, windspeed, rainfall}
ENM Concentration (water)	ng/L	Estimated ENM concentration in water. Affected by {release (water), concentration (air), land area,
		water area, windspeed, rainfall}
ENM Concentration (soil)	µg/kg	Estimated ENM concentration in soil. Affected by {release (soil), concentration (air), land area,
	4	
ENM Concentration (sediment)	µg/kg	Estimated ENM concentration in sediment. Affected by {Concentration (water)}
Atmospheric Convective	hour	The average time for a unit volume of air to reside in the simulated region.
residence time		
Water convective residence time	hour	The average time for a unit volume of water to reside in the simulated region.
(Water current)		
Atmospheric mixing height	meter	The height above the surface throughout which a pollutant/unit volume is dispersed
Soil bulk density	g/cm ³	The weight of soil in a given volume
Sediment bulk density	g/cm ³	
Soil top layer depth	meter	Depth of the top layer of soil
Water body depth	meter	Depth of water body
Suspended solids density	g/cm ³	
Attachment factor (air)	%	Fraction of ENMs attached to ambient particles in air
Attachment factor (water)	%	Fraction of ENMs attached to ambient particles in water
Solubility	ppm	ENM solubility (the ability for the ENM to dissolve in a solvent (water))