Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

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

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1. Bayesian Networks

Bayesian Networks (BNs) belong to the family of probabilistic graphical models that are useful for representing knowledge and reasoning while considering uncertainty. BNs development involves the construction of a directed acyclic graph (DAG) of available information (parameters) treated as nodes in the network. The influence/conditional dependence relations among these nodes are represented 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, release rates and concentrations), the BNs were utilized by constructing network structure and learning 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 (e.g., windspeed can be discretized into the states of distinct ranges). In the particular case of BN for exposure modeling, an arc from a node (such as windspeed (m/s)) to a child node (such as air concentration (ng/m^3)) represents the conditional dependence of concentration on windspeed and windspeed is called the parent node of concentration. Thus the arrow indicates that an assigned windspeed directly influences air concentration (manuscript **Fig. 3**). The complexity of BN is dictated by the dimensionality of the conditional probability tables (constructed based on causal links among parameters)[1–4] as the size of a CPT of a node in BN is determined by the number of incoming links (parent nodes), the number of states of each parent node, and the number of states of the node itself.

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 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 the fundamental fate and transport model[5]. The resulting 18 parameter-set (including the initial pool) was used in sensitivity analysis using Alexander's sensitivity indicator[6] in order to guide the selection of final parameter-set for model development. Using Alexander's indicator, each parameter was varied 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 differences was from 0 – 1 indicating 1 as the maximum possible sensitivity or effect on resulting concentrations, where the cutoff threshold was set to 0.97 (any parameter with sensitivity indicator ≥ 0.03) for the selection of parameters in final pool. The indicator's measure of sensitivity of the concentrations to changes in model parameters is given by

$$S = \frac{\sum_{i=1}^{N} \frac{(O_{ik} - O_{ij})^2}{O_{ij}}}{\sum_{\substack{i=1 \\ max}}^{N} \frac{(O_{ik} - O_{ij})^2}{O_{ij}}} \tag{1}$$

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 concentrations at regular time intervals over a one-year simulation period. As an illustration, the predicted compartmental concentrations of TiO_2 in Bern (Switzerland) (with size distribution in air given as (μ = 179nm)) are shown in **Fig. S1**.

Table S1: Parameter sensitivity analysis using Alexander's sensitivity indicator

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

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.

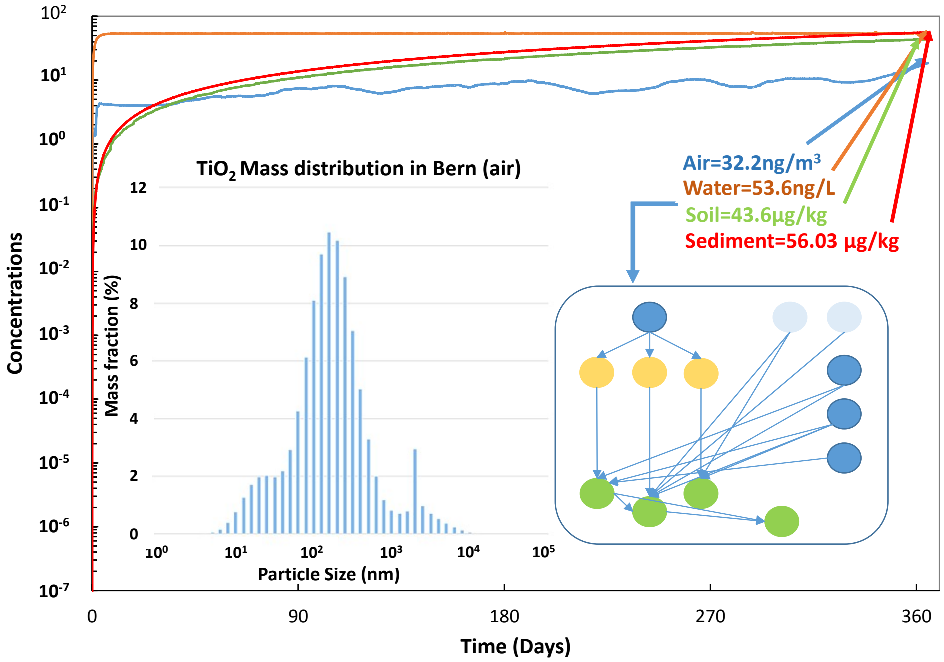

Figure S1: Temporal profile of TiO_2 environmental concentrations in Bern (Switzerland) starting with clean environment. Following a well-established concept of annual cumulative distribution, the concentrations for one-year simulation time period were utilized to construct the BN conditional probability tables (CPTs).

The environmental distributions of ENMs for low releases (in air, water, soil and sediment) were estimated using MendNano for both the training and test sets (Experimental Section). The resulting concentrations from the training set served to construct the BN-nanoExpo model. The adequacy of the resulting BN was then assessed via correlation analysis of predicted MendNano estimated concentrations and the predicted concentrations by BN-nanoExpo for test set. The $\log_{10}$ transformed 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)**) were for nano Cu (1) in air in Houston (-2.63 (2.4×10^{-3} ng/m^3)), and (2) in water in New York (-3.46 (3.5×10^{-4} ng/L)). Exposure concentrations for TiO_2 and ZnO in air and water were relatively higher.

The highest exposure concentrations in air (atmosphere) was for TiO_2 in Berlin (1.81 (~65 ng/m^3)) and the highest exposure concentration in water was for TiO_2 in Bern (1.97 (~94 ng/L)).

	Al_2O_3	CeO_2	Cu	SiO_2	TiO_2	ZnO
Los Angeles	0.63	0.07	-2.20	1.10	1.48	0.93
New York	0.85	0.29	-1.98	1.31	1.69	1.13
Houston	0.22	-0.33	-2.63	0.69	1.08	0.52
Wayne	-1.50	-2.09	-1.48	-1.03	-0.64	-1.22
Maricopa	0.04	-0.52	-0.85	0.51	0.90	0.34
Bern	0.68	0.16	-2.24	1.16	1.53	1.02
Berlin	0.94	0.37	-1.88	1.39	1.81	1.26
London	0.87	0.27	-1.98	1.29	1.75	1.18

1.81 / -2.63

(a)

Al_2O_3	CeO_2	Cu	SiO_2	TiO_2	ZnO
0.11	-1.11	-3.16	-0.25	1.26	0.64
-0.23	-1.38	-3.46	-0.50	0.91	0.29
-0.06	-1.27	-3.32	-0.42	1.09	0.47
-0.40	-1.60	-0.83	-0.78	0.75	0.13
-2.56	-3.17	-2.90	-2.22	-1.84	-2.39
0.83	-0.36	-2.44	0.51	1.97	1.36
0.49	-0.74	-2.78	0.10	1.64	1.02
0.75	-0.50	-2.53	0.33	1.90	1.28

1.97 / -3.46

(b)

2.11

Los Angeles	0.83	0.27	-1.93	1.32	1.64	1.03
New York	1.30	0.75	-1.44	1.80	2.11	1.48
Houston	0.48	-0.07	-2.27	0.98	1.30	0.69
Wayne	-1.24	-1.80	-1.15	-0.74	-0.42	-1.03
Maricopa	0.10	-0.46	-0.73	0.59	0.91	0.30
Bern	0.82	0.29	-1.98	1.32	1.64	1.07
Berlin	1.22	0.66	-1.55	1.71	2.04	1.43
London	1.07	0.50	-1.70	1.55	1.90	1.29

-2.27

(c)

1.99

0.13	-1.05	-3.12	-0.18	1.28	0.66
-0.17	-1.24	-3.37	-0.32	0.95	0.34
-0.03	-1.23	-3.29	-0.37	1.11	0.49
-0.38	-1.61	-0.81	-0.76	0.77	0.15
-2.17	-2.74	-2.98	-1.71	-1.28	-1.81
0.85	-0.33	-2.43	0.54	1.99	1.37
0.52	-0.69	-2.74	0.16	1.67	1.05
0.77	-0.47	-2.51	0.37	1.92	1.30

-3.37

(d)

Figure. S2: ENM Concentrations ($\log_{10}$ transformed) in **(a)** air (*ng/m^3*), **(b)** water (*ng/L*), **(c)** soil (*µg/kg*), and **(d)** sediment (*µg/kg*) estimated by MendNano for 8 selected cities.

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 both soil (**Fig. S2 (c)**) and sediment (**Fig. S2 (d)**) were of nano Cu which were; (1) in soil in Houston (-2.27 (5.3×10^{-3} µg/kg)), and (2) in sediment in New York (-3.37 (4.3×10^{-4} µg/kg)). Exposure concentrations for TiO_2 and ZnO in soil and sediment were higher among the six ENMs due in part to the higher ENM release rates. The highest exposure concentration of TiO_2 in sediment in Bern of ~98 µg/kg (1.99), was likely due to higher release rates as well as higher exposure concentration in water in the above region (as there is a causal relationship of concentration in water with the concentration in 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, and Berlin (in decreasing order) and the lowest overall ENMs releases were in Wayne and Maricopa.

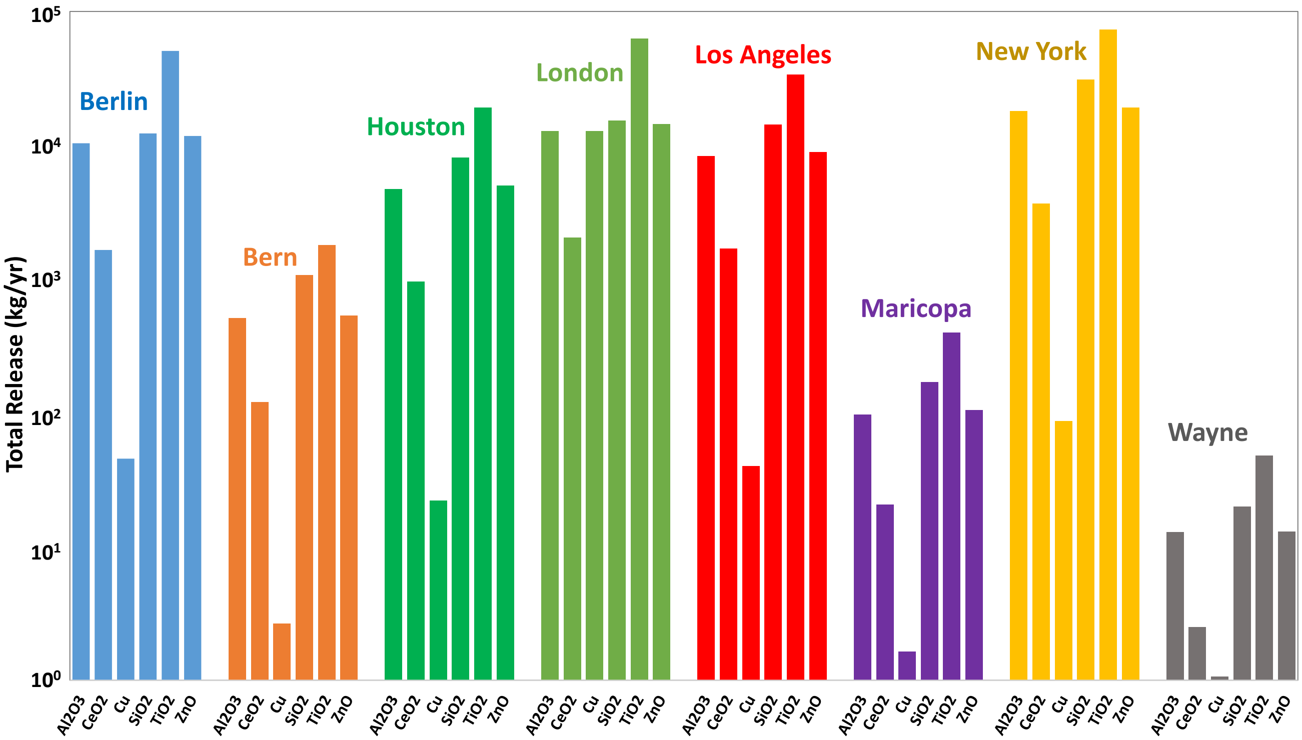

Figure. S3 (a): Low end estimates of the ENMs total releases (kg/yr) for the indicated cities

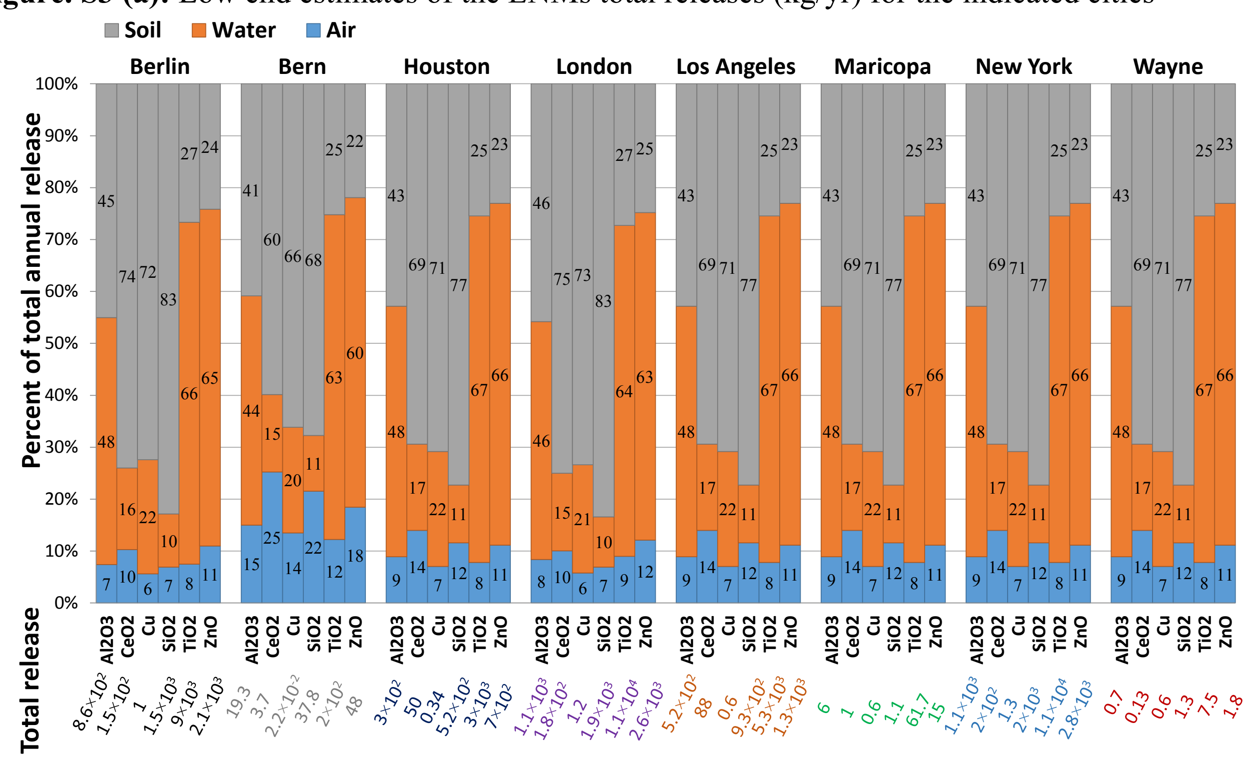

Figure S3 (b): Stacked bar of low estimates of ENM releases to air, water and soil as percentage of the total ENM release for the indicated cities. The total releases (kg/year) for each ENM are reported below the bar charts. Note that the estimated release rates of SiO_2, TiO_2 and ZnO are higher in 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.

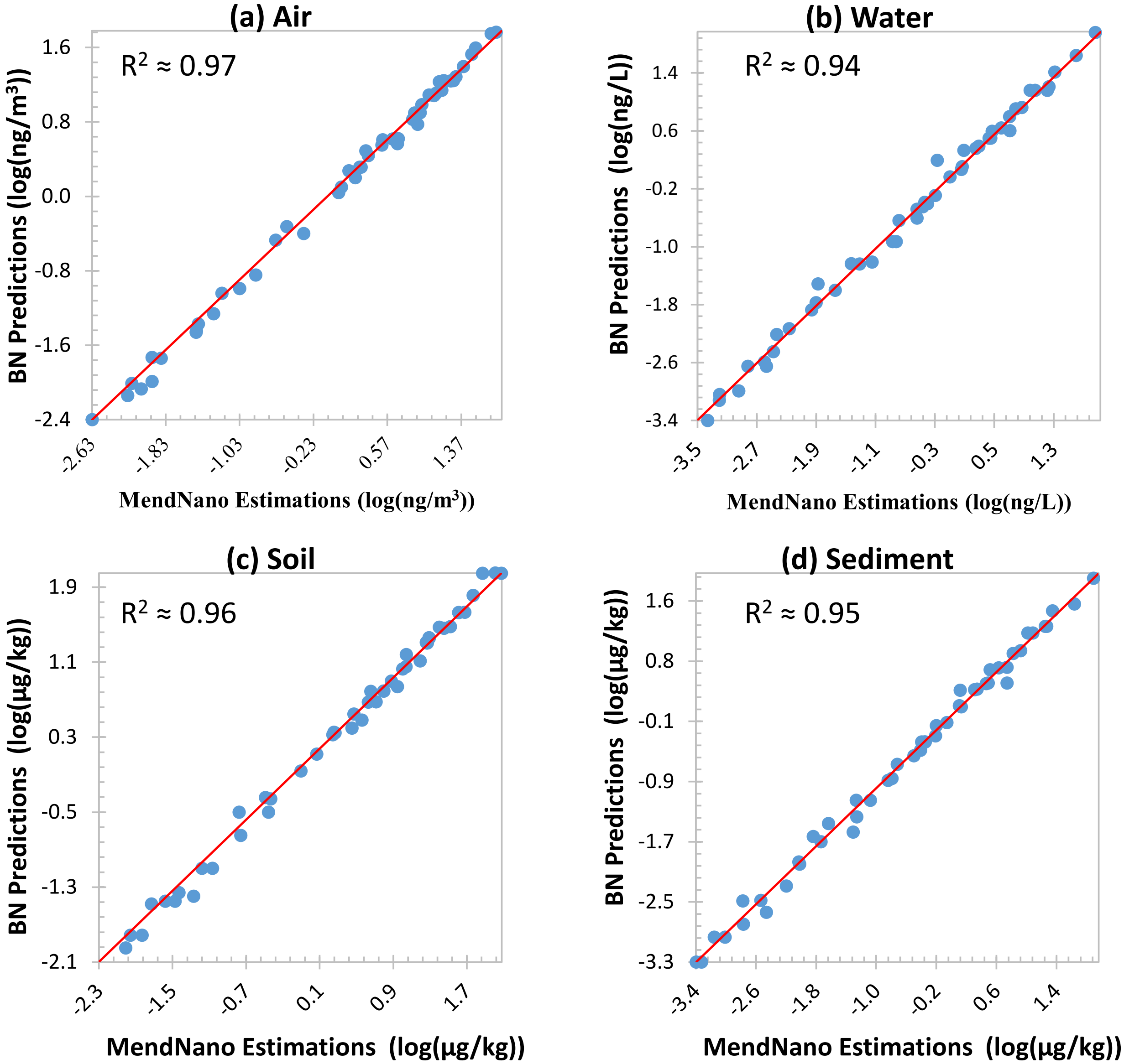

Fig. S4: Observed MendNano estimations vs BN predictions of environmental media concentrations along with R^2 for all four compartments **(a)** air, **(b)** water, **(c)** soil, and **(d)** sediment.

3. Conditional dependence of ENMs concentrations on input parameters

In data driven modeling approaches, the complexity and dimensionality of information poses a 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 outcomes of interest. In this regard, BNs are especially advantageous since they enable the visualization 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, the BN model rapid assessment of the impact of uncertainty in multiple parameters either individually or simultaneously expressed as normal distributions about the mode, on the resulting compartmental concentrations. As an illustration of the above a number of test cases are shown in **Figs. S5 – S10** for Al_2O_3, CeO_2, Cu, SiO_2, TiO_2 and ZnO whereby the impact of uncertainties in multiple parameters (expressed in terms of normal distributions) was evaluated with respect to the resulting distributions of the ENMs concentrations in the various environmental media.

Al_2O_3 parameter values

Parameter	Mean ± Stdev
Release air (kg)	60 ± 30
Release water (kg)	$3.6\times10^2\pm100$
Release soil (kg)	$2.7\times10^2\pm70$
Rain (mm/month)	30 ± 10
Temperature (C)	10 ± 5
Windspeed (m/s)	4 ± 1
Land area (km^2)	$1.2\times10^3\pm100$
Water area (km^2)	30 ± 5
Water current speed	65 ± 10 hrs

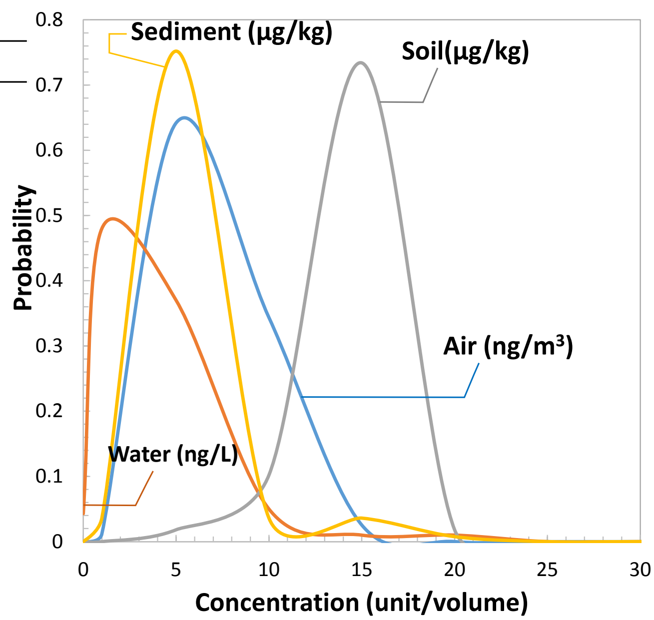

Figure S5: Predicted compartmental concentrations of Al_2O_3 for the range of parameter values. The conditional dependence relationships demonstrate that lower releases of Al_2O_3 (air = 60kg, water = 360kg, soil = 270kg) in land area = 1220km^2 and water area = 30 km^2 resulted in lower compartmental concentrations than those of TiO_2, SiO_2, and ZnO (**Fig. S6-S9**).

TiO_2 parameter values

Parameter	Mean ± Stdev
Release air (kg)	$6.5\times10^2 \pm 100$
Release water (kg)	$4.8\times10^3 \pm 300$
Release soil (kg)	$1.1\times10^3 \pm 200$
Rain (mm/month)	30 ± 10
Temperature (C)	17 ± 5
Windspeed (m/s)	2.85 ± 0.5
Land area (km²)	$1.2\times10^3 \pm 100$
Water area (km²)	75 ± 10
Water current speed	35 ± 5 hrs

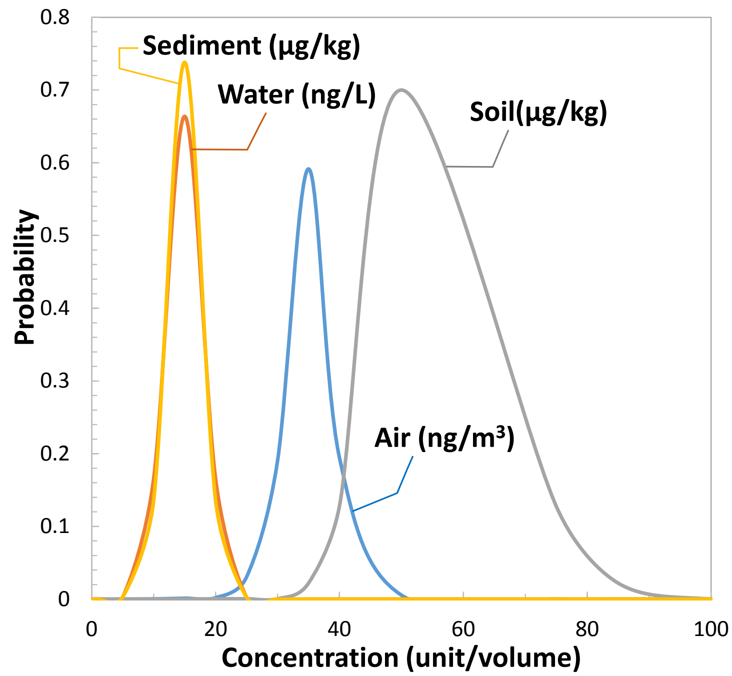

Figure S6: Distribution of predicted compartmental concentrations of TiO_2 for the range of parameter values (i.e., parameter uncertainty).

TiO_2 parameter values

Parameter	Mean ± Stdev
Release air (kg)	$9.8\times10^2 \pm 150$
Release water (kg)	$4.8\times10^3 \pm 300$
Release soil (kg)	$2.3\times10^3 \pm 70$
Rain (mm/month)	30 ± 10
Temperature (C)	10 ± 5
Windspeed (m/s)	2.85 ± 0.5
Land area (km²)	$1.5\times10^3 \pm 200$
Water area (km²)	0.2 ± 3
Water current speed	21 ± 3 days

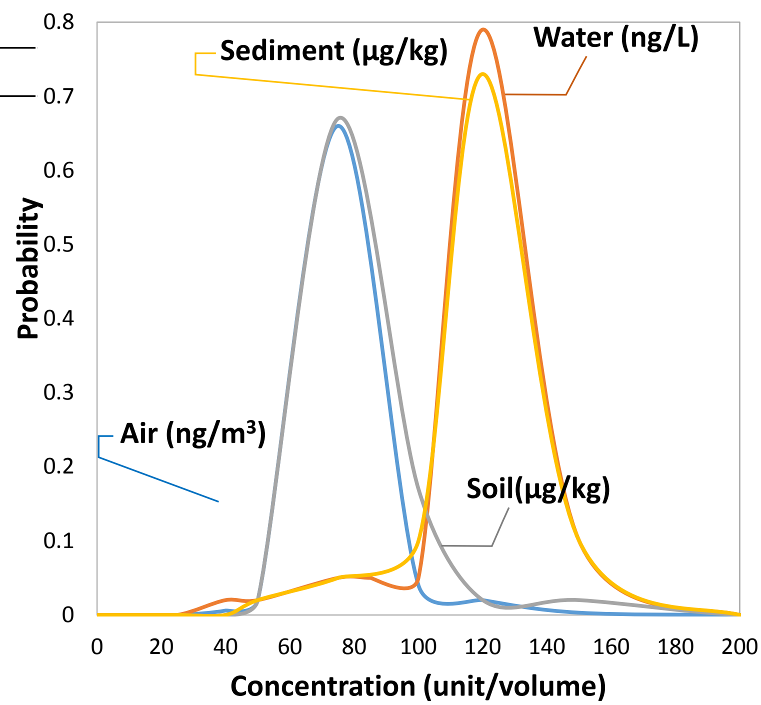

Figure S7: Distribution of predicted compartmental concentrations of TiO_2 for the range of parameter values (i.e., uncertainties). The effect of higher releases of TiO_2 on compartmental concentrations is shown as their probability distributions. Compartmental concentrations of TiO_2 (air ≈ 75 ng/m³, water ≈ 85 ng/L, soil ≈ 120 µg/kg, sediment ≈ 75 µg/kg) resulted higher (compared to **Fig. S6**) due to higher release rates (air = 980kg, water = 2.3×10^3, soil=2.3×10^3) and updated geographical parameters (land area = 1500km², water area = 0.2 km²).

ZnO parameter values

Parameter	Mean ± Stdev
Release air (kg)	$2.3\times10^2 \pm 50$
Release water (kg)	$1.3\times10^3 \pm 150$
Release soil (kg)	$5.5\times10^2 \pm 100$
Rain (mm/month)	45 ± 15
Temperature (C)	10 ± 5
Windspeed (m/s)	2.85 ± 0.5
Land area (km^2)	$8.5\times10^2 \pm 100$
Water area (km^2)	50 ± 10
Water current speed	35 ± 5 hrs

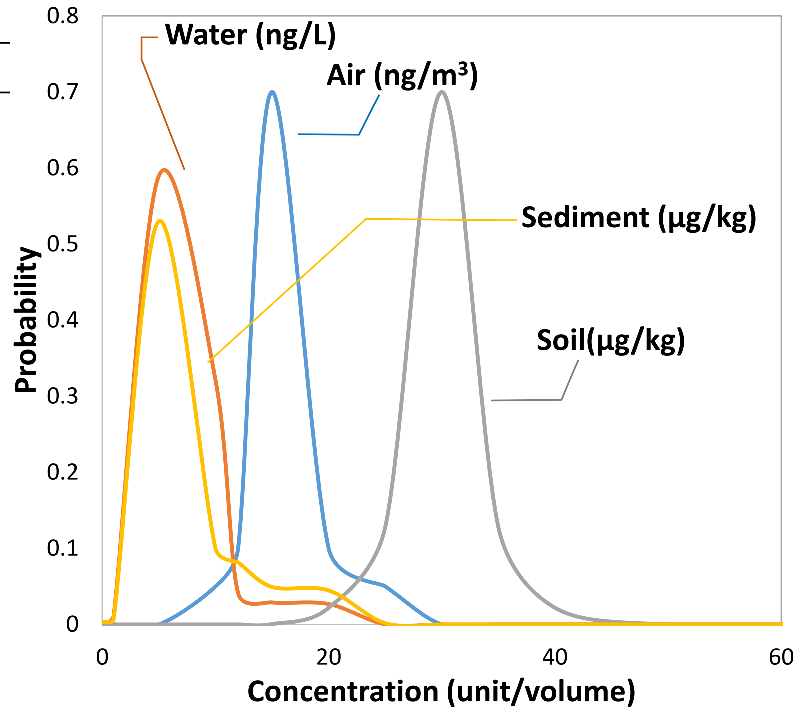

Figure S8: Distribution of predicted compartmental concentrations of ZnO for the range of parameter values (uncertainties).

SiO_2 parameter values

Parameter	Mean ± Stdev
Release air (kg)	$1.1\times10^2 \pm 25$
Release water (kg)	$1.6\times10^2 \pm 30$
Release soil (kg)	$5.5\times10^2 \pm 100$
Rain (mm/month)	50 ± 15
Temperature (C)	11 ± 5
Windspeed (m/s)	2.85 ± 0.5
Land area (km^2)	$9.5\times10^2 \pm 100$
Water area (km^2)	80 ± 15
Water current speed	65 ± 10 hrs

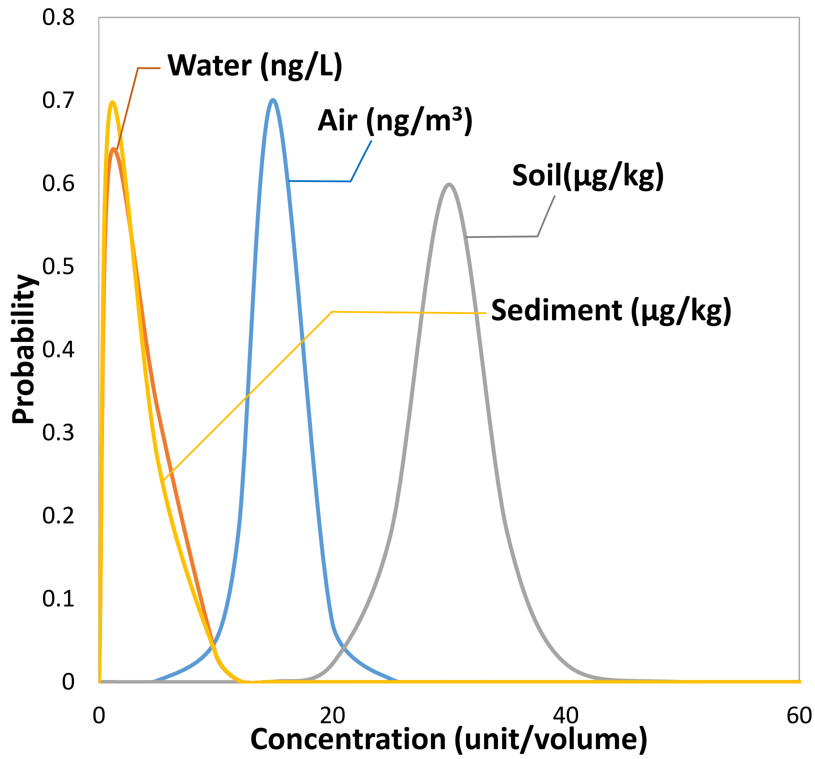

Figure S9: Distribution of predicted compartmental concentrations of SiO_2 for the range of parameter values (uncertainties).

Cu parameter values

Parameter	Mean ± Stdev
Release air (kg)	$7\times10^{-2} \pm 100$
Release water (kg)	0.2 ± 5
Release soil (kg)	0.5 ± 5
Rain (mm/month)	50 ± 15
Temperature (C)	20 ± 5
Windspeed (m/s)	3.5 ± 0.5
Land area (km^2)	$10^3 \pm 100$
Water area (km^2)	90 ± 15
Water current speed	15 ± 3 days

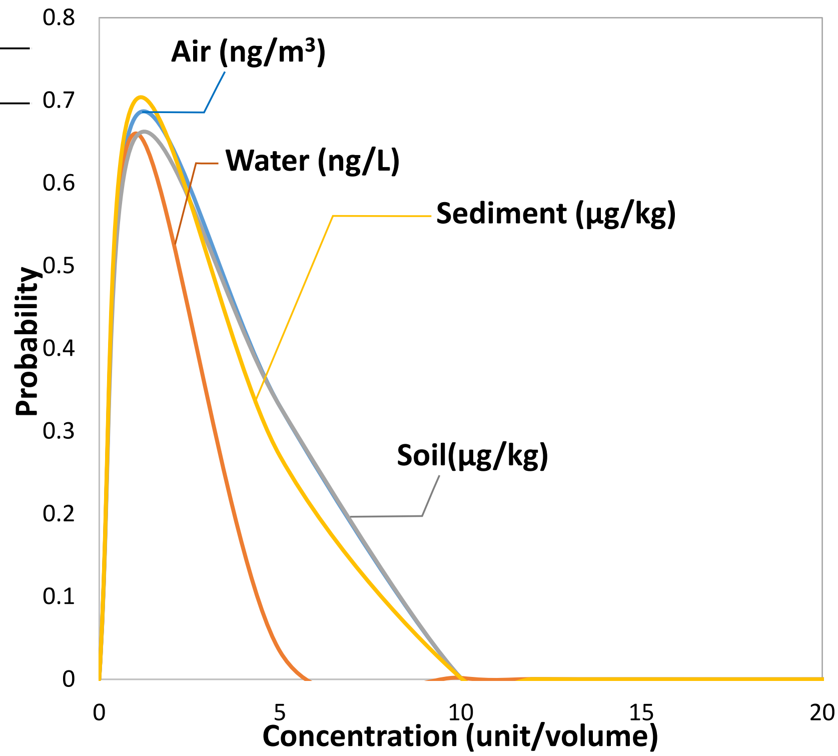

Figure S10: Distribution of predicted compartmental concentrations of nano-Cu for the range of parameter values (uncertainties).

It is important to recognize that ambient particles are present (in both air and water) at significantly higher number concentrations relative to those which may be expected based solely on potential releases of ENMs[7]. Therefore, ENMs are likely to be associated with ambient particulates (due to various surface–surface interactions[8]) given the high available surface area of ambient particles and tendency of most ENMs to agglomerate[9–11]. The particle size of ambient aerosols typically ranges from 0.001 to 2 µm with particle size distribution (PSD) typically described by a trimodal log-normal size distribution[11]. Suspended solids in natural water bodies are typically in the size range of 0.01–1 µm for lakes[12], 1–100 µm for oceans[13], and 30–150 µm for rivers[14], and log-normal size distributions have been often reported[7], with a concentration range that can vary significantly (30 µg L–1–200 mg L–1)[15,16] depending on the specific water body. Here we note that previous work has shown that under most conditions essentially all ENMs would be attached to ambient particles[7]. The extent of heteroaggregation, however, can be quantified via an attachment efficiency or an attachment factor[5], 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) TiO_2 and (b) CeO_2 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.

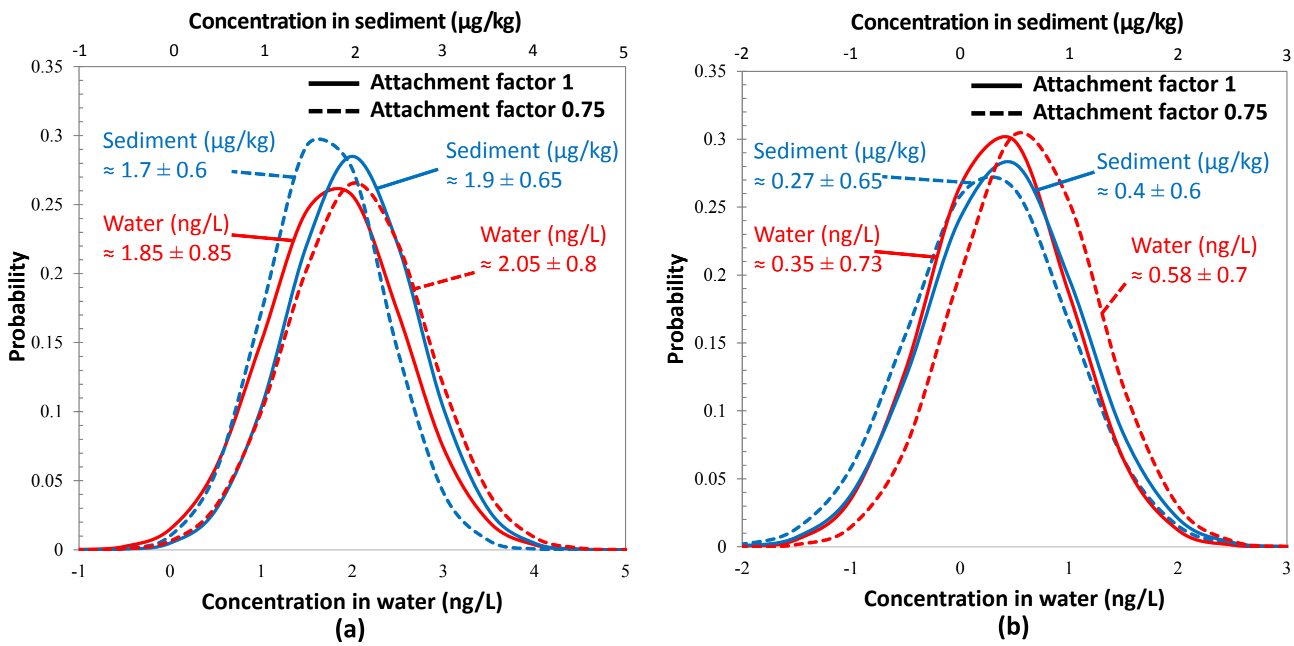

Figure S11. Impact of (a) TiO_2 and (b) CeO_2 ENM attachment to suspended solids (in water) on ENM concentrations in water and sediment in Houston.

4. Quantification of Parameter Significance in Predicting ENM Environmental Distribution

The BN-nanoExpo model allows rapid assessment of the distribution of compartmental concentrations as impacted by input parameter uncertainty. BN sensitivity analysis allows one to determine the influence of input parameters on the resulting ENM environmental concentrations. Here, 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

$$Vr = V(Q) - V(Q|f) \quad (2)$$

where

$$V(Q) = \sum_q P(q)[Xq - E(Q)]^2 \quad (3)$$

$$V(Q|f) = \sum_q P(q|f)[Xq - E(Q|f)]^2 \quad (4)$$

and

$$E(Q) = \sum_q P(q)Xq \quad (5)$$

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.

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	C
Release rate to air [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[64, 15, 0.05, 105, 665, 236]	kg/year
Release rate to water [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[410.5,23, 0.21, 156, 5.8×10^3, 1.4×10^3]	kg/year
Release rate to soil [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[389, 108.5, 0.7, 1.3×10^3, 2.5×10^3, 518]	kg/year

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

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	C
Release rate to air [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[3, 0.9, 3×10^{-3}, 8.1, 23.3, 9]	kg/year
Release rate to water [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[8.5, 0.5, 4.5×10^{-3}, 4, 119, 28.6]	kg/year
Release rate to soil [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[8, 2.2, 1.4×10^{-2}, 26, 48, 10.5]	kg/year

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	C
Release rate to air [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[26, 7, 2.3×10^{-2}, 61, 232, 80.5]	kg/year
Release rate to water [Al_2O_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_2O_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	C
Release rate to air [Al_2O_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_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[489, 27.1, 0.25, 184, 6.9×10^{3}, 1.6×10^{3}]	kg/year
Release rate to soil [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[489, 136.3, 0.9, 1.6×10^{3}, 2.9×10^{3}, 651.3]	kg/year

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

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	C
Release rate to air [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[46, 12.4, 4.2×10^{-2}, 107.5, 410.5, 142]	kg/year
Release rate to water [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[249, 14.8, 0.13, 103, 3.5×10^{3}, 841.4]	kg/year
Release rate to soil [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[220.7, 61.6, 0.4, 717, 1.3×10^{3}, 294]	kg/year

Table S7: Parameters for simulation of environmental 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	C
Release rate to air [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[0.5, 0.14, 4.1×10^{-2}, 1.3, 4.8, 1.7]	kg/year
Release rate to water [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[2.9, 0.2, 0.13, 1.2, 41, 9.85]	kg/year
Release rate to soil [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[2.6, 0.72, 0.42, 8.4, 15.7, 3.4]	kg/year

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

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	C
Release rate to air [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[99, 27, 9×10^{-2}, 233, 888, 308]	kg/year
Release rate to water [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[539, 32, 0.3, 223, 7.6×10^{3}, 1.8×10^{3}]	kg/year
Release rate to soil [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[477, 133, 0.9, 1.5×10^{3}, 2.9×10^{3}, 637]	kg/year

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

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	C
Release rate to air [Al_2O_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_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[0.35, 2×10^{-2}, 0.13, 0.14, 5, 1.2]	kg/year
Release rate to soil [Al_2O_3, CeO_2, Cu, SiO_2, TiO_2, ZnO]	[0.3, 8×10^{-2}, 0.4, 1, 2, 0.42]	kg/year

Supplementary Information References

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

City	Compartment	ENMs Release (kg/yr) (low – high)					
		Al_2O_3	CeO_2	Cu	SiO_2	TiO_2	ZnO
Berlin	Air	64 – 290	15 – 66	0.05 – 1.4	105 – 458	665 – 1,210	236 – 520
	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
Bern	Air	3 – 192	0.9 – 55	0.003 – 1.1	8 – 514	23 – 494	9 – 193
	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
Houston	Air	26 – 1,076	7 – 301	0.02 – 6	61 – 2,814	232 – 2,857	81 – 1,110
	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
London	Air	90 – 455	18 – 96	0.07 – 455	132 – 692	980 – 1,954	319 – 725
	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
Los Angeles	Air	46 – 1,903	12 – 533	0.04 – 11	108 – 4,978	410 – 5,054	142 – 1,963
	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
Maricopa	Air	1 – 22	0.1 – 7	0.04 – 0.13	1.3 – 60	4.8 – 60	1.7 – 23
	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
New York	Air	99 – 4,118	27 – 1,153	0.1 – 23	233 – 10,773	888 – 10,936	308 – 4,249
	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
Wayne	Air	0.1 – 3	0.02 – 0.8	0.04 – 0.15	0.2 – 7	0.6 – 7	0.2 – 3
	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 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)	^oC	Average monthly temperature (^oC) 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, rainfall}
ENM Concentration (sediment)	μg/kg	Estimated ENM concentration in sediment. Affected by {Concentration (water)}
Atmospheric Convective residence time	hour	The average time for a unit volume of air to reside in the simulated region.
Water convective residence time (Water current)	hour	The average time for a unit volume of water to reside in the simulated region.
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))