Life Cycle Impact Assessment of terrestrial acidification: modeling spatially explicit soil sensitivity at the global scale

Supporting Information

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The available Supporting Information details the worldwide input parameter creation process and provides complementary results. The document is 39 pages long spanning six sections and is comprised of 22 tables and 15 figures.

SECTION 1: The terrestrial acidification cause-effect chain

Figure SI 1 presents the terrestrial acidification cause-effect chain that was considered.



Figure SI1: Terrestrial acidification cause-effect chain

SECTION 2: Indicator calculations

The multiple layer approach of the PROFILE model makes it possible to assess the indicator values based on roots distribution across the soil layers. Roots distribution according to depth was estimated using equation SI 1.

$$\mathbf{Y} = 1 - \boldsymbol{\beta}^d \tag{SI 1}$$

Where Y is the cumulative root fraction (between 0 and 1) from the soil surface to depth "d" [cm] and β is the fitted "extinction coefficient". Known β values and calculated roots distribution as a function of terrestrial biomes are supplied in Table SI 1.

Terrestrial Biomes	β	Fraction of roots by layers of 20 cm						
		0-20 cm	20-40 cm	40-60 cm	60-80 cm	80-100 cm		
Boreal forest	0.943	0.69	0.21	0.07	0.02	0.01		
Crops	0.961	0.55	0.25	0.11	0.05	0.02		
Desert	0.975	0.40	0.24	0.14	0.09	0.05		
Sclerophyllous shrubs	0.964	0.52	0.25	0.12	0.06	0.03		
Temperate coniferous forest	0.976	0.38	0.24	0.15	0.09	0.06		
Temperate deciduous forest	0.966	0.50	0.25	0.13	0.06	0.03		
Temperate grassland	0.943	0.69	0.21	0.07	0.02	0.01		
Tropical deciduous forest	0.961	0.55	0.25	0.11	0.05	0.02		
Tropical evergreen forest	0.962	0.54	0.25	0.11	0.05	0.02		
Tropical grassland savanna	0.972	0.43	0.25	0.14	0.08	0.04		
Tundra	0.914	0.83	0.14	0.02	0.00	0.00		
Mixed forest (mean between temperate coniferous and temperate deciduous forests)	0.971	0.44	0.25	0.14	0.08	0.04		

Table SI 1: β and roots distribution fractions according to terrestrial biomes

The single value indicator (1 per soil layer "n") was aggregated into a single value indicator using the distribution of roots as weighting factors.

Indicator =
$$\sum_{n}$$
 Indicator _n × Root fraction _n (SI 2)

Similarly, the critical aluminium concentration criteria was calculated using equations (4) and (5) with pH critical values according to soil layer "n" (4-4.2 for first 20 cm and 4.4 for the rest), K_{gibbs} (related to log kAl) and roots distribution fraction.

$$[\mathrm{Al}^{3+}] = \mathrm{K}_{\mathrm{gibbs}} \times [\mathrm{H}^{+}]^{3}$$
(SI 4)

$$[Al^{3+}]_{critical} = \sum_{n} K_{gibbs_{n}} \times (10^{-pH_{critical}})_{n}^{3} \times Root \quad fraction_{n} \qquad (SI5)$$

SECTION 3: Dataset creation

3.1 Wet and dry atmospheric deposition

The deposition levels of acids were obtained from a $2^{\circ}x2.5^{\circ}$ GEOS Chem [1] simulation for 2005, which is representative of the average from 1961 to 1990 according to the National Climatic Data Center of the National Oceanic and Atmospheric Administration [2]. Deposition values were calculated in terms of kg N or S deposited/yr.

The evaluation of base cations deposition is a problem in global-scale analysis, since there is no available data on global base cations deposition fluxes [3]. In the absence of global-scale estimates, soil dust deposition was used as a proxy for calcium deposition [4]. The portion of calcium in dust deposits was averaged at 10% following Kuylstierna et al. [4]. Soil mineral dust deposition was evaluated from the Tegen and Fung $4^{\circ}x5^{\circ}$ grid resolution model [5].

Sodium, chloride, magnesium and potassium deposits were derived using a similar approach, and sea salt deposition was used as a proxy since it represents an important contributor of sodium, chloride and magnesium. Sea salt deposition levels were estimated with the GEOS Chem's "sulphate within sea-salt" deposits and global "sulphate within sea-salt" ratio [6]. Magnesium represents 2% of the sea salt deposits, while sodium and chloride represents a share of 30% and 55% respectively.

Using measured values from the National Atmospheric Deposition program database for 2005 [7], the deposition value of potassium was correlated with the Tegen and Fung $4^{\circ}x5^{\circ}$ grid resolution model [5] as about 2%.

To be used in PROFILE, the values were transformed into keq/ha/yr using equation (SI 6):

Deposition [Keq/ha/yr] = Deposition [kg/ha/yr] x Valence [-] / Molecular weight [g/mol] (SI 6)

3.2 Annual mean precipitations and air temperature

Values were obtained from the WorldClim-Global climate database [8] on a 10 arcminute resolution. Figure SI 2 illustrates the worldwide precipitation and temperature distribution.



Figure SI 2: Annual average precipitation levels (up) and air temperature (down)

3.3 Soil properties

The ISRIC-Wise worldwide database differentiates areas based on soil properties on a 5 by 5 arc-minute resolution. Figure SI 3 illustrates this differentiation. Colors are indicative of the presence of an ecosystem.



Figure SI 3: Differentiated areas based on soil properties

3.4 Water content

Water content $[m^3 water/m^3 soil]$ was considered as a fraction (80%) of the field capacity (maximum water content), which is a function of soil texture from the ISRIC-Wise worldwide database. Table SI 2 presents field capacity and permanent wilting point (minimum water content to sustain vegetation life) values obtained from Rawls et al. [9].

classes							
FAO soil texture class	Field capacity $(m^3 \text{ water/ } m^3 \text{ soil})$	Permanent wilting point (m ³ water/ m ³ soil)					
Sand	0.091	0.033					
Loamy sand	0.125	0.055					
Sandy loam	0.207	0.095					
Sandy clay loam	0.255	0.148					
loam	0.27	0.117					
silt loam	0.33	0.133					
silt	0.32	0.17					
clay loam	0.318	0.197					
silty clay loam	0.366	0.208					
sandy clay	0.339	0.239					
silty clay	0.387	0.25					
clay	0.396	0.272					

Table SI 2: Field capacity and permanent wilting point according to FAO soil texture

3.5 Soil bulk density

Soil bulk density [kg/m³] was obtained from ISRIC-Wise worldwide.

3.6 Water entering and exiting soil layers

The percentages of water entering and exiting each soil layer [%] are often pure guesses in PROFILE studies [10-15]. We proposed to link a certain percentage of the entering precipitations lost in the layer to the FAO qualitative drainage class from the ISRIC-Wise worldwide database. The corresponding absorbed water percentages were found by correlating FAO drainage classes to the values in PROFILE studies [10-15]. The studies were mostly destined for "well drained soils" and showed an average value of -8%. Extrapolation of a -2% difference between each class was then assumed.



Figure SI 4: Percentage (%) of water entering/exiting a layer according to soil depths from PROFILE studies

FAO	Excessively	Extremely	Well	Moderately	Imperfectly	Poorly	Very
Drainage	drained	drained	drained	drained	drained	drained	poorly
Class							drained
Absorbed							
water	4	6	8	10	12	16	20
percentage	-4	-0	-0	-10	-12	-10	-20
[%]							

Table SI 3: FAO drainage class and assumed absorbed water percentage

3.7 Surface area

The surface area $[m^2/m^3]$ for layer "n" was calculated according to equation (SI 7).

Surface area $_{n} = [(0.3 \times \% \text{ Sand}) \times (2.2 \times \% \text{ Silt}) \times (8 \times \% \text{ Clay})] \times \text{Bulk density} \times 1000$

(SI 7)

3.8 Runoff

Runoff [m] was calculated with equation (SI 8):

Runoff = Precipitat ion
$$\times$$
 (% Water leaving last layer) (SI 8)

3.9 Net uptake

Net uptake represents the extraction of cations and nitrogen from the soil for use by vegetation. Each vegetation type from Olsen et al [16] was associated to net uptake values from different ecosystems, as surveyed by Duan et al [17]. Table SI 4 shows the net uptake values according to vegetation type.

Table SI 4: Used net uptake values according to vegetation type (keq/ha/yr)

	Ca	Mg	K	Ν
Taiga	0.59	0.22	0.09	0.52
Southern Taiga	0.33	0.19	0.11	0.57
Northern or maritime taiga	0.04	0	0.03	0.07
Tropical/subtropical, humid forest,	0.33	0.19	0.11	0.57
Temperate/boreal forest				
Tropical woods, temperate woods, fields, grass,	0.44	0	0.36	1.82
scrub				
Mixed forests	0.95	0.11	0.15	0.87
Dry forest and woodland	1.01	0	0.42	2
Temperate broad-leaved forest	1.19	0.07	0.11	0.35
Broad-leaved humid forest	1.49	0.5	0.48	2
Tropical savanna or montane	0.13	0	0.13	0.48
Tropical Savanna and Woodland	0.83	0	0.2	0.74
Irrigated land and surrounding	0.28	0	0.29	1.04
Mangrove/tropical swamp forest	0.28	0	0.29	1.04
Main grassland and shrub land	0.18	0	0.31	1.14
Cold grass or stunted woody complex	0.09	0	0.09	0.29
Non-polar desert or semi-desert	0.16	0	0.10	0.30
Tundra	0.04	0	0.03	0.07
Other crop, settlements, marginal lands, town	0.90	0.19	0.31	1.28
Shore and hinterland complexes	0.17	0	0.41	0.81

3.10 Dissolved organic carbon

Dissolved organic carbon (DOC) [mg/L] was calculated with equation (SI 9) for each soil layer "n". DOC was estimated from total organic carbon (TOC) values in the ISRIC-Wise worldwide database. Since not all organic carbon (e.g. roots) is soluble, a constant was set to represents the fraction of TOC available for solubilisation. The constant values (CTE) were set using known typical values of DOC for each soil layer from PROFILE studies [10-15]. Dry bulk density ("density") and water content ("WC") were also required to obtain DOC values.



Figure SI 5: DOC values according to soil depths from PROFILE studies

Table SI5:	Used DOC	constants	values
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	Soil Layer 1	Soil Layer 2	Soil Layer 3	Soil layer 4	Soil layer 5
DOC constant values [m ³ /dm ³]	0.00006	0.000026	0.00002	0.00001	0.000009

3.11 Mineralogy

Vertisols

Mineralogy [%] was extrapolated from known single-site data to a global soil classification, as defined by the global soil regions world map. Based on this classification, local site sample data were aggregated (arithmetic mean). The result is an average value for each mineral according to the global soil order.



Figure SI 6:	Map	of g	lobal	soil	order
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	Alfisol	Andisols	Aridisols	Entisols	Histosols	Gelisol	Inceptisol	Mollisols	Oxisols	Spodosols	Ultisols	Vertisols
Number of sites	41	29	63	23	5	17	55	40	14	28	61	14
Spatial coverage	Michigan (US) Wisconsin (US) Louisiana (US) Texas (US) Colorado (US) Tennessee (US) Philippines Illinois (US)	Washington State (US)	California (US) Texas (US) Nevada (US) New Mexico (US) Utah (US) Arizona (US) Idaho (US)	Nebraska (US) Texas (US) Nevada (US)	North Carolina (US)	Nunavut (Can)	Alaska (US) Maine (US) Massachusetts (US) Scotland Ireland Czech Republic Germany	Kansas (US)	Brazil South Africa Kenya	PEI (Can) Québec (Can) Nova Scotia (Can) Vermont (US) Michigan (US) Sweden	Virginia (US) North Carolina (US)	Texas (US)
Potassium												
Feldspar	9.90	0.29	9.48	13.52	16.80	9.56	5.14	21.59	0.15	11.52	3.08	1.93
Muscovite	0.50	0.11	0.45	0.17	1.00	0.50	6.77	2.03	0.00	2.44	8.13	0.00
Calcite	2.28	0.00	0.73	0.22	0.00	26.44	0.09	0.54	0.00	0.26	0.00	0.14
Pyroxene	0.20	0.86	0.74	1.30	0.20	2.25	1.04	0.79	0.08	0.30	1.45	0.00
Chlorite	0.23	0.00	0.02	0.04	0.00	0.00	4.31	0.21	0.00	0.45	0.03	0.21
Garnet	0.15	0.04	0.00	0.09	0.00	0.06	0.11	0.18	0.00	0.02	0.02	0.00
Hornblende	0.35	0.54	0.65	1.13	0.40	0.06	1.46	0.74	0.00	0.91	1.73	0.00
Biotite	0.83	1.39	2.23	0.43	0.00	0.44	4.20	1.08	0.00	2.46	6.18	0.14
Plagioclase												
Feldspar	10.03	9.36	5.77	15.78	0.40	0.00	10.27	2.08	0.00	3.98	0.67	5.07
Apatite	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.00	0.00	0.05	0.00	0.00
Epidote	0.05	0.00	0.06	0.13	0.40	0.00	0.00	0.03	0.08	0.18	0.13	0.00
Koalinite	0.03	0.00	0.08	0.00	0.00	0.00	1.73	0.00	0.00	0.00	1.32	0.00
Albite	0.00	0.00	0.00	0.00	0.00	0.00	1.98	0.00	0.00	0.00	1.78	0.00
Vermiculite	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.28	0.00
Quartz	64.55	10.32	65.77	46.52	78.20	54.00	44.00	61.64	71.77	54.05	66.78	85.92
Other [*]	10.9	77.09	14.02	20.67	2.6	6.69	18.88	9.09	27.92	23.38	8.42	6.59

Table SI 6: Input sand-silt mineralogy (%) for the PROFILE model simulations

* "Other" treated as quartz by default in the PROFILE model

	Alfisol	Andisols	Aridisols	Entisols	Histosols	Gelisol	Inceptisol	Mollisols	Oxisols	Spodosols	Ultisols	Vertisols
Number of sites	41	29	63	23	5	17	55	40	14	28	61	14
Spatial coverage	Michigan (US) Wisconsin (US) Louisiana (US) Texas (US) Colorado (US) Tenesse (US) Phillipines Illinois (US)	Washington State (US)	California (US) Texas (US) Nevada (US) New Mexico (US) Utah (US) Arizona (US) Idaho (US)	Nebraska (US) Texas (US) Nevada (US)	North Carolina (US)	Nunavut (Can)	Alaska (US) Maine (US) Massachuchet (US) Scotland Irland Czech Republic Germany	Kansas (US)	Brazil South Africa Kenya	PEI (Can) Quebec (Can) Nova Scotia (Can) Vermont (US) Michigan (US) Sweden	Virginia (US) North Carolina (US)	Texas (US)
Potassium												
Feldspar	0	0	3	1	0	0	0	0	0	0	2	0
Muscovite	0	0	0	0	0	0	0	0	0	0	0	0
Calcite	0	0	20	11	0	1	0	4	0	0	0	4
Pyroxene	0	0	0	0	0	0	0	0	0	0	0	0
Chlorite	0	0	0	0	0	0	0	0	0	0	0	0
Garnet	0	0	0	0	0	0	0	0	0	0	0	0
Hornblende	0	0	0	0	0	0	0	0	0	0	0	0
Biotite	0	0	0	0	0	0	0	0	0	0	0	0
Plagioclase												
Feldspar	0	6	0	1	0	0	0	0	0	0	0	0
Apatite	0	0	0	0	0	0	0	0	0	0	0	0
Epidote	0	0	0	0	0	0	0	0	0	0	0	0
Koalinite	31	16	15	20	30	30	0	24	42	34	45	20
Albite	0	0	0	0	0	0	0	0	0	0	0	0
Vermiculite	22	55	1	2	38	15	21	1	13	20	21	2
Quartz	4	16	6	5	11	12	4	10	0	15	1	9
Other [*]	43	7	55	60	21	42	75	61	45	31	31	65

Table SI 7: Input clay mineralogy (%) for the PROFILE model simulations

"Other" treated as quartz by default in the PROFILE model

3.12 Alexp and anorthite in oligoclasses

The model's default values were used. Table SI8 presents the values.

	fuult purulleter values
Parameter	Parameter values
Anorthite in oligoclasses [%]	0.8
Alexp [-]	3

Table SI 8: Default parameter values

3.13 Kgibbs

The values for these parameters were considered to be layer-depth dependant default values. Average values from PROFILE studies [10-15] were used (see Figure SI7). Table SI9 presents the values.



Figure SI 7: Log KAlsol according to soil depths from PROFILE studies

	Soil layer 1	Soil layer 2	Soil layer 3	Soil layer 4	Soil layer 5
Log KAlsol (log Kgibbs) [-]	7.2	8.3	8.8	9.1	9.3

 Table SI 9: Used log KAlsol values

3.14 BC/N uptake efficiency

The values for these parameters were considered to be layer-depth dependant default values. Average values from PROFILE studies [10-15] were used (see Figure SI8). Table SI 10 presents the values.



Figure SI 8: BC (up) or N (down) uptake efficiency according to soil depths from PROFILE studies

1401	Tuble bi 10: 0500 Dent uptake entereney values								
	Soil layer 1	Soil layer 2	Soil layer 3	Soil layer 4	Soil layer 5				
BC/N uptake efficiency [%]	50	30	10	5	2				

Table SI 10: Used BC/N uptake efficiency values

3.15 CO₂ pressure

The values for these parameters were considered to be layer-depth dependant default values. Average values from PROFILE studies [10-15] were used (see Figure SI9). Table SI 11 presents these values.



Figure SI 9: CO₂ pressure according to soil depths from PROFILE studies

Tuble 61 11. Osed CO ₂ pressure values					
	Soil layer 1	Soil layer 2	Soil layer 3	Soil layer 4	Soil layer 5
CO2 pressure [atm]	5	10	20	20	20

Table SI 11: Used CO₂ pressure values

3.16 Denitrification

The approach used in this paper follows the one outlined by Bouwman et al [3]. The deposition of N compounds was corrected for transformations of N to determine the net acidifying effect. A fraction of N is lost by soil denitrification. This fraction is a function of soil texture and soil drainage, as presented in table SI 12.

Table SI 12: Denitrification factor as a function of FAO qualitative drainage class and soil texture class

Drainage Class		Soil texture cl	lass
	Coarse	Medium	Fine
Excessively to well drained	0.1	0.3	0.7
Well to moderately well drained	0.2	0.4	0.8
Imperfectly drained	0.3	0.5	0.8
Poorly drained	0.4	0.6	0.8
Very poorly drained	0.6	0.8	0.8

3.17 Litter fall and canopy exchange

Litter fall and canopy exchange may be considered as retardants of atmospheric deposition. We assumed that all atmospheric deposition in a steady-state will ultimately reach ground level, and the computation of litter fall and canopy exchange can be

considered double counting. This simplification is in line with the simplifications used in regional PROFILE [11] applications.

3.18 Net mineralization

Being a steady-state model, PROFILE does not model the accumulation or depletion of organic material.

SECTION 4: Uncertainty due to parameter variability

The weather characteristics and soil parameters inputs were given 1 000 different random values. Tables SI 13 to SI 15 present the distribution from which new values were selected for each of the simulated parameters.

Parameter	Description	MINIMUM	MAXIMUM		
T di dificici	Description	(relative or absolute	(relative or		
		(relative of absolute	(iciative of		
		value to the best	absolute value to		
		estimate)	the best estimate)		
	WEATHER CHAR	ACTERISTICS			
Precipitation [m]	Uniform distribution	90%	110%		
Temperature [°C]	Uniform distribution	90%	110%		
SOIL PARAMETERS					
Soil water content	Uniform distribution	Permanent wilting	Field capacity		
[m ³ water/m ³ soil]		point			
Soil bulk density	Uniform distribution	75%	125%		
$[kg/m^3]$					
Water	Uniform distribution	80%	120%		
entering/leaving layer					
(%)					
Surface area [m ² /m ³]	Uniform distribution	70%	130%		
Net uptake	Uniform distribution	0%	100%		
[keq/ha/yr]					

 Table SI 13: Uniform distribution values for input parameters that DO NOT VARY according to soil depth

Table SI 14: Uniform distribution values for input parameters that VARY according to soil depth

	Dissolved	Log Kgibb	CO2	BC uptake	N uptake
	organic		pressure	efficiency	efficiency
	carbon				
Description	Uniform distribution				
Layer 1	5 to 34	6.5 to 8.5	2 to 10	10 to 85	10 to 85
Layer 2	3.7 to 8	7.5 to 9	10 to 25	5 to 40	5 to 40
Layer 3	1 to 5	8.2 to 9.5	10 to 25	0 to 30	0 to 30
Layer 4	0 to 5.9	8.2 to 9.5	10 to 30	0 to 60	0 to 60
Layer 5	0 to 2	9 to 9.5	20 to 30	0 to 35	0 to 35

Minerals	Used standard deviation
Potassium Feldspar	5.73
Muscovite	2.70
Calcite	3.29
Pyroxene	1.56
Chlorite	1.06
Garnet	0.16
Hornblende	1.56
Biotite	3.60
Plagioclase Feldspar	6.61
Apatite	0.01
Epidote	0.23
Koalinite	0.95
Albite	1.22
Vermiculite	0.15

 Table SI 15: Normal distribution standard deviation for minerals (the mean of the normal distribution is the best estimate)

SECTION 5: SF results for NO_x-HNO₃ and NH₃ emissions



Figure SI 10: Worldwide simulated indicators for a 10% variation in N deposition from NO_x-HNO₃ emissions: (A) BC/Al, (B) Al/Ca, (C) pH and (D) Al



Figure SI 11: Worldwide simulated indicators for a 10% variation in N deposition from NH₃ emissions: (A) BC/Al, (B) Al/Ca, (C) pH and (D) Al

SECTION 6: Discussion results

6.1 Comparison of PROFILE and Simple Mass Balance (SMB) models

Table SI16 presents a comparison of the PROFILE and Simple Mass Balance (SMB) models on several modelling aspects.

	PROFILE	Simple Mass Balance (SMB)
Model type	Steady-state	Steady-state
Weathering rates	Modelled with the PROFILE model using soil mineralogy as input	Inputs
Mineralogy	Required*	Not required*
Water transport	Water is assumed to move vertically through several soil layers	No transport; one layer
Cation exchange between Al, H and base cations	Gapon exchange reactions	Gaines-Thomas equations
Exchange between soil matrix and the soil solution	Diffusion limited	Equilibrium equation
Aluminium concentration	Gibbsite	Gibbsite
Organic acids [R–]	Calculated as: [R-] = $K_{Oliv} \times [DOC] \times \alpha$ $/(K_{Oliv} + [H+]) **$	Not included
Bicarbonates [HCO3-]	Calculated as: [HCO3-] = $K_{H2CO3} \times Khenry \times Pco2$ $/([H+])^{***}$	Not included
Sodium (Na) uptake by plants	Neglected: not a major nutrient	Neglected: not a major nutrient

Table SI 166: Comparison of PROFILE and SMB models

* SMB does not require mineralogy because it does not calculate weathering rates and takes them as inputs. However, it commonly use the PROFILE mineral sub-model to calculate weathering rates. Global scale evaluation of mineralogy may be an issue especially if minerals were not included in the sub-model. If not included, the minerals are treated as quartz by default. Most soil orders have a limited number of "other minerals" (mostly below 10% for the sand/silt mineralogy; see Table SI 6)

** where DOC is dissolved organic carbon in g m⁻³, α a conversion factor ($\alpha = 7 \ 10^{-6}$ kmol g⁻¹) and *K*Oliv a constant dependant of pH

*** where K_1 is a dissociation constant, Khenry is Henry constant and Pco2 is the CO2 partial pressure

6.2 Comparison of the simulated background pH with pH from the ISRIC-Wise database

We compared the simulated indicator values and those from the ISRIC-Wise database (version 1.1). The comparison is limited to the pH indicator, since it is the only available indicator found at a global scale. While not bereft of uncertainties since PROFILE simulates the steady-state soil solution pH and the database only provides a soil paste pH that may be far from steady-state, the comparison could still provide an approximate idea of the accuracy of the simulated indicator values. The comparison of the simulated and ISRIC-Wise worldwide pH values is assessed with relative error. To have a consistent comparative framework, the ISRIC-Wise five layer pH data were aggregated into a single value using the roots distribution fraction.

In the analysis of the results, the spatial variation of the ISRIC-Wise must be considered. The database estimates parameters for 106 FAO soil units but does not explicitly consider the possible effects of regional variations in climate, relief, parent material, land use, and management practices on specific soil parameters [18]. The ISRIC-Wise soil type pH variation was found to be approximately $\pm 25\%$.

Discrepancies in the comparison could be explained by the fact that PROFILE simulated the steady-state soil solution pH and that the database only provided a soil paste pH that may be far from steady-state. However, further research found other sources of discrepancies that would drastically improve these percentages. By modifying the deposition levels with the ones from field measurements from Environment Canada [19], the following correlation was obtained for the 804 receiving environments in Canada's eastern provinces. From the results presented in Figure SI 12, it was found that 97% of the receiving environments solely constituted of the "boreal forest/taiga" and the "temperate broadleaf and mixed forest" biomes were inside the $\pm 25\%$ relative error range. This is a net improvement when compared to the previously obtained results with 62 and 73%, respectively, of the receiving environment in the $\pm 25\%$ relative error range.



Figure SI 12: Comparison of simulated pH (with deposition measurements instead of global modeling) with the pH from the ISRIC-Wise database

Figure SI 13 shows the relative error between the acidification potential obtained from measurements and simulated deposition values.



Figure SI 13: Acidification potential relative error between measured and simulated values for the 804 receiving environments in Canada's eastern provinces.

Figures SI 14 shows deposit discrepancies between the global deposition estimates and the measured values for all the considered deposited substances. Most acid deposition values are inside the $\pm 50\%$ error range from a perfect match. The same cannot be said of the base cations, sodium and chloride deposition.



Figure SI 14: Measured and simulated values of deposition

6.3 Correlation between sample sites and soil dust or seasalt depositions

The following graphs presents the obtained correlations between deposition of calcium, magnesium, sodium, chloride and potassium at 231 sampling sites of the National Atmospheric Deposition program database for 2005 [7] with soil dust from Tegen and Fung $4^{\circ}x5^{\circ}$ grid resolution model [5] or sea-salt deposition from GEOS-Chem. Correlations are similar to the values presented in Supporting information 3.1.



Figure SI 15: Correlation between sample sites and soil dust or sea-salt depositions

6.4 pH modifications using 1, 5 and 10% marginal change

Table SI17 presents the relative error to the average pH after a change of 1%, 5% and 10% in emissions of SO2-SO4 for 100 randomly chosen receiving environments. Results show that 38% of the selected receiving environments are unaffected by a 1% change in deposition. These ratios drop to 11% and 6% after a 5% and 10% deposition change, respectively.

Average pH	Relative pH error	Relative pH error	Relative pH error
	1% change	5% change	10% change
5.07	0.00E+00	2.37E-04	4.73E-04
4.92	1.63E-04	8.95E-04	1.83E-03
7.34	5.45E-05	5.45E-05	1.09E-04
7.21	5.55E-05	1.94E-04	3.61E-04
5.22	7.66E-05	6.13E-04	1.34E-03
5.69	0.00E+00	1.76E-04	3.16E-04
6.18	3.24E-05	9.72E-05	1.62E-04
5.18	3.86E-05	3.48E-04	6.18E-04
6.21	6.44E-05	1.93E-04	4.18E-04
7.53	0.00E+00	5.31E-05	5.31E-05
7.06	5.66E-05	3.12E-04	5.95E-04
8.09	0.00E+00	4.94E-05	4.94E-05
6.65	6.02E-05	3.91E-04	8.73E-04
6.46	6.20E-05	4.96E-04	9.29E-04
8.43	0.00E+00	2.37E-05	4.75E-05
8.04	0.00E+00	2.49E-05	4.97E-05
7.12	2.81E-05	8.43E-05	1.97E-04
6.49	1.54E-04	8.63E-04	1.82E-03
4.96	2.01E-04	8.06E-04	1.57E-03
4.52	8.86E-05	1.77E-04	4.43E-04
5.03	7.95E-05	3.18E-04	6.76E-04
4.77	1.26E-04	4.61E-04	1.01E-03
4.68	1.28E-04	5.98E-04	1.11E-03
4.70	1.70E-04	5.96E-04	1.15E-03
7.49	0.00E+00	2.67E-05	2.67E-05
5.74	6.97E-05	2.79E-04	5.22E-04

Table SI 177: Relative error between average pH and modified average pH from a change of 1%, 5% and 10% in SO2-SO4 deposition

5.04	3.97E-05	1.59E-04	2.78E-04
5.05	0.00E+00	3.96E-05	3.96E-05
5.00	8.00E-05	6.00E-04	1.12E-03
5.04	3.97E-05	1.19E-04	1.98E-04
5.05	0.00E+00	3.96E-05	3.96E-05
6.39	0.00E+00	0.00E+00	0.00E+00
7.40	0.00E+00	0.00E+00	0.00E+00
5.03	3.98E-05	3.18E-04	5.97E-04
5.01	7.98E-05	4.79E-04	8.38E-04
4.99	4.01E-05	5.22E-04	1.12E-03
7.29	0.00E+00	0.00E+00	2.75E-05
7.29	0.00E+00	2.74E-05	5.49E-05
4.94	1.21E-04	4.05E-04	7.69E-04
5.75	6.96E-05	2.78E-04	5.57E-04
7.30	0.00E+00	0.00E+00	8.22E-05
8.78	0.00E+00	0.00E+00	0.00E+00
7.56	2.65E-05	2.65E-05	2.65E-05
7.54	0.00E+00	7.96E-05	1.33E-04
8.06	0.00E+00	0.00E+00	2.48E-05
7.20	0.00E+00	0.00E+00	2.78E-05
6.68	0.00E+00	2.99E-05	8.98E-05
6.70	0.00E+00	2.98E-05	2.98E-05
5.01	7.98E-05	4.79E-04	8.78E-04
7.63	0.00E+00	2.62E-05	2.62E-05
7.31	2.74E-05	5.47E-05	5.47E-05
6.25	3.20E-05	2.56E-04	4.48E-04
5.35	3.74E-05	1.87E-04	5.23E-04
6.55	3.05E-05	6.11E-05	1.22E-04
6.63	0.00E+00	3.02E-05	3.02E-05
6.84	2.92E-05	8.77E-05	1.46E-04
6.48	0.00E+00	1.54E-04	3.40E-04
5.79	3.45E-05	1.04E-04	2.42E-04
6.08	3.29E-05	6.58E-05	9.87E-05
6.25	3.20E-05	6.40E-05	9.60E-05
4.65	9.04E-04	4.78E-03	9.34E-03
6.70	2.09E-04	1.19E-03	2.48E-03
7.93	0.00E+00	0.00E+00	2.52E-05

7.45	0.00E+00	2.68E-05	2.68E-05
4.99	1.20E-04	8.01E-04	1.56E-03
6.65	3.01E-05	9.02E-05	1.80E-04
8.55	2.34E-05	7.02E-05	1.17E-04
8.54	0.00E+00	2.34E-05	4.69E-05
7.02	5.70E-05	5.70E-05	1.14E-04
7.02	5.70E-05	5.70E-05	1.14E-04
6.39	9.40E-05	4.70E-04	8.77E-04
8.18	0.00E+00	2.44E-05	2.44E-05
7.72	0.00E+00	7.77E-05	1.04E-04
7.73	0.00E+00	5.18E-05	1.81E-04
8.63	0.00E+00	0.00E+00	0.00E+00
8.96	0.00E+00	0.00E+00	0.00E+00
7.80	0.00E+00	2.56E-05	7.69E-05
8.47	0.00E+00	2.36E-05	2.36E-05
8.24	0.00E+00	7.28E-05	1.21E-04
7.51	7.99E-05	2.13E-04	4.00E-04
7.70	2.60E-05	7.80E-05	2.08E-04
7.67	2.61E-05	7.83E-05	1.83E-04
7.87	2.54E-05	5.09E-05	1.02E-04
7.53	7.96E-05	1.06E-04	1.33E-04
7.87	0.00E+00	2.54E-05	5.08E-05
4.22	1.90E-04	9.96E-04	1.95E-03
4.18	1.91E-04	9.56E-04	1.91E-03
5.83	2.74E-04	1.48E-03	3.02E-03
6.75	2.96E-05	2.96E-04	6.23E-04
5.48	1.83E-03	8.58E-03	1.67E-02
6.70	2.39E-04	1.22E-03	2.54E-03
6.89	0.00E+00	1.45E-04	2.90E-04
6.82	5.87E-05	2.93E-04	5.28E-04
6.95	2.59E-04	1.15E-03	2.36E-03
7.03	5.69E-04	2.82E-03	5.78E-03
7.40	0.00E+00	5.41E-05	1.35E-04
6.52	3.99E-04	1.90E-03	3.68E-03
6.20	6.45E-05	2.58E-04	5.48E-04
5.97	2.35E-04	1.17E-03	2.41E-03
7.34	0.00E+00	0.00E+00	0.00E+00

6.5 Comparison of SFs to Bouwman et al. critical load exceedence

The Bouwman et al. method compares measured atmospheric acid deposition ("Acid dep"), calculated as per eq (SI 10), to an atmospheric CL interval. Intervals are given as a function of five different sensitivity classes based on cation exchange capacity (CEC) and base saturation (BS) values (table SI 17). Values of these parameters were obtained from the interpolation/extrapolation database. Nitrogen immobilisation (N_{imm}) was set to 0 and denitrification (f_{den}) was obtained as previously reported (SI section 3.16).

Acid dep =
$$S_{dep} + (N_{dep} - N_{imm}) \times (1 - f_{den}) - BC_{dep}$$
 (SI 10)

Bouwman and colleagues critical load approach				
Sensitivity Class	CEC (meq/100 g ⁻¹) at field pH	Base saturation (%)	Critical load value (meq m ⁻² yr ⁻¹)	
1 (most sensitive)	<10 10-25	<40 <20	12.5-25	
2	<10 10-25 >25	40-60 20-40 <20	25-50	
3	<10 10-25 >25	60-80 40-60 20-40	50-100	
4	10-25 >25	60-80 40-60	100-200	
5 (least sensitive)	<25 >25	80-100 >60	>200	

Table SI 17: Bouwman's soil classes and critical load values range

Exceedence of critical loads occurs when S and N deposits are higher than the maximum critical load value; No exceedence occurs when deposits are lower than the minimum critical load value and possible exceedence when deposits are between minimum and maximum critical load values.

Table SI 18 defines the used quintiles of the SFs based indicators as calculated by empirical cumulative distribution functions.

To do so, each of the receiving environments SFs were classified, per pollutant, into quintiles (1 the highest SFs; 5 the lowest SFs) based on an empirical cumulative distribution function.

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SF based indicators	Quintiles 1	Quintiles 2	Quintiles 3	Quintiles 4	Quintiles 5
		N from NOx-	HNO3 emissions		
BC/Al	>18.6	18.6-0.1	0.1-0.001	0.001-0.0	< 0.0
Al/Ca	>6.7	6.7-0.006	$0.006 - 1.0 \times 10^{-4}$	$1.0 \times 10^{-4} - 0.0$	< 0.0
pH	>0.3	0.3-0.01	$0.01-7.0 \times 10^{-4}$	$7.0 \times 10^{-4} - 0.0$	< 0.0
Al	>3.1×10 ⁴	3.1×10^{4} - 3.5×10^{1}	3.5×10^{1} -1.1	1.1-0.0	<0.0
		N from N	H3 emissions		
BC/Al	>27.6	27.6-0.15	0.15-0.0026	0.0026-1.1×10 ⁻⁵	<1.1×10 ⁻⁵
Al/Ca	>9.2	9.2-0.01	0.01-2.0×10 ⁻⁴	$2.0 \times 10^{-4} - 1.0 \times 10^{-6}$	<1.0×10 ⁻⁶
pН	>0.5	0.5-0.02	0.02-0.001	$0.001-2.4 \times 10^{-5}$	<2.4×10 ⁻⁵
Al	>7.4×10 ⁴	7.4×10^{4} - 5.5×10^{1}	5.5×10 ¹ -1.9	1.9-0.02	<0.02
		S from SO2	-SO4 emissions		
BC/Al	>86.4	86.4-0.43	0.43-0.008	0.008-0.0001	< 0.0001
Al/Ca	>28.1	28.1-0.02	0.02-5.0×10 ⁻⁴	$5.0 \times 10^{-4} - 1.1 \times 10^{-5}$	<1.1×10 ⁻⁵
pH	>0.7	0.7-0.05	0.05-0.003	$0.003 - 1.6 \times 10^{-4}$	<1.6×10 ⁻⁴
Al	>9.1×10 ⁴	9.1×10 ⁴ - 1.5×10 ²	1.5×10^{2} -4.8	4.8-0.24	<0.24

Table SI 18: Defined quintiles of SFs based indicators (BC/Al [ha keq-1 ha yr], Al/Ca [ha keq-1 ha yr], pH [mol H⁺ L⁻¹ ha keq-1 ha yr] and Al [μ mol Al L⁻¹ ha keq-1 ha yr])

Following Tables SI 19 to SI 21 cross-reference the SFs quintiles with the Bouwman critical load exceedence. As SFs quintiles may change across pollutants, the receiving environment representative quinitles was the lowest one obtained.

	Quintiles 1	Quintiles 2	Quintiles 3	Quintiles 4	Quintiles 5
Exceedence	5.9	3.9	2.2	1.4	0.3
Maybe	6.3	5.9	4.3	2.9	0.6
No	10.6	11.9	17.4	18.6	7.3
exceedence					

Table SI 19: Comparison of BC/Al based SFs quintiles with critical load exceedence conlcusions from Bouwman et al.

Table SI 20: Comparison of Al/Ca based SFs quintiles with critical load exceedence conlcusions from Bouwman et al.

	Quintiles 1	Quintiles 2	Quintiles 3	Quintiles 4	Quintiles 5
Exceedence	5.3	2.4	1.6	1.3	0.6
Maybe	5.8	4.6	3.5	2.5	1.0
No exceedence	10.5	16.1	18.9	16.7	9.0

 Table SI 21: Comparison of pH based SFs quintiles with critical load exceedence conlcusions from Bouwman et al.

	Quintiles 1	Quintiles 2	Quintiles 3	Quintiles 4	Quintiles 5
Exceedence	5.8	4.0	1.9	1.8	0.3
Maybe	6.5	5.8	3.8	3.2	0.9
No exceedence	12.0	12.8	15.7	16.4	9.0

Table SI 22: Comparison of Al based SFs quintiles with critical load exceedence conlcusions from Bouwman et al.

	Quintiles 1	Quintiles 2	Quintiles 3	Quintiles 4	Quintiles 5			
Exceedence	6.2	3.3	2.2	1.8	0.3			
Maybe	6.4	5.4	4.4	3.1	0.8			
No exceedence	10.2	12.6	17.3	16.7	9.0			

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