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Yu, Lin; Belyazid, Salim; Akselsson, Cecilia; van der Heijden, Gregory; Zanchi, Giuliana

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1 Storm Disturbances in a Swedish Forest—a case study Comparing

2 Monitoring and Modelling

- 3 Lin Yu^{a,*}, Salim Belyazid^a, Cecilia Akselsson^b, Gregory van der Heijden^c and Giuliana Zanchi^b
- ^a Centre for Environmental and Climate Research, Lund University, Sölvegatan 37, SE-223 62 Lund, Sweden
- 5 b Department of Physical Geography and Ecosystem Science, Lund University, Sölvegatan 12, SE-223 62 Lund,
- 6 Sweden

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7 ° INRA – UR 1138 Biogéochimie des Ecosystèmes Forestiers, route d'amance, 54280 Champenoux, France

8 Abstract

A Norway spruce (Picea abies Karst) forest site in southwest Sweden was chosen to study the effects of storm disturbances over the period 1997–2009, during which two storms, 'Lothar' (Dec. 1999) and 'Gudrun' (Jan. 2005), affected the area. Monitored deposition data, soil water chemistry data and forest inventory data were compared with the predictions of an integrated ecosystem model, ForSAFE, in an effort to reveal and understand the effects of storms on acidification/recovery in forest soils. Both storms caused windthrow loss leading to increased nitrate and sulphate concentrations in soil water as a result of stimulated mineralization. Lothar led to increased concentrations of Na⁺, Mg²⁺, and Cl⁻ in soil water due to sea-salt episode. No general sea-salt episode was seen following Gudrun, but small sea-salt episodes were observed in 2007 and 2008. Each sea-salt episode caused a temporary decrease of pH, and a subsequent recovery, but overall, the soil water pH decreased from 4.54 to 3.86 after Lothar. Modelling suggested that the site was recovering from acidification from 1990s, and would continue to recover in future. Both modelled and monitored data showed that storm caused disturbances in the recovery; monitored data even suggested that soil acidification happened due to storm disturbances. Sea-salt episode does not increase soil acidity in the long term, and will probably decrease the soil acidity by replenishing the base saturation. The modelled data also suggested that storms with only windthrow would not have effects on soil acidification recovery in the long term, but they may influence the soil fertility by losses of base cations.

^{*} Corresponding author. Tel.: +46 46 222 86 34.

E-mail addresses: lin.yu@cec.lu.se (L. Yu), salim@belyazid.com (S. Belyazid), cecilia.akselsson@nateko.lu.se (C. Akselsson), gregory.vanderheijden@nancy.inra.fr (G. van der Heijden), giuliana.zanchi@nateko.lu.se (G. Zanchi).

27 Keywords:

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Forest soil, Dynamic model, Storm disturbance, Sea-salt episode, Acidification recovery

1. Introduction

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Swedish forests generally grow on nutrient-poor soils, mainly tills from nutrient-poor bedrocks such as 31 32 granites and gneisses (Wastenson et al. 1990). The fertility of these soils has been further affected by acid deposition (Reuss & Johnson 1986). Furthermore, whole-tree harvesting has become more common in 33 recent decades, leading to further removal of base cations (Iwald et al. 2013; Akselsson et al. 2007; 34 35 Brandtberg & Olsson 2012). One main effect of acid deposition (S and N) on forest soils is the increased leaching of sulphate and nitrate coupled with nutrient cations such as Mg²⁺, Ca²⁺ and K⁺ causing base 36 cations (BC: Ca²⁺, Mg²⁺, K⁺) depletion from the soil (Reuss & Johnson 1986). Base cations depletion in 37 forest soils due to high acid deposition has been simulated in several model approaches in regional scale 38 39 (Sverdrup & Rosen 1998; Akselsson et al. 2007) and in catchment scale (Navrátil et al. 2007; HruŠka et al. 40 2012; Oulehle et al. 2007). However, the N deposition in most of Sweden is generally low (Simpson et al. 2011), and northern forest ecosystems are normally nitrogen (N) limited (Tamm 1991; Jonard et al. 2015). 41 42 Thus, leaching of inorganic nitrogen is generally very low in Swedish forests, except for the 43 southwesternmost part with the highest present and historical N deposition, where the nitrate concentration 44 in soil water is often elevated (Akselsson et al. 2010). 45 S deposition to forest ecosystems has strongly decreased since the 1990s throughout Sweden, but recovery from acidification has been reported to be very slow (Pihl Karlsson et al. 2011; Akselsson et al. 2013). 46 47 This has also been demonstrated through dynamic modelling (Sverdrup et al. 2005), and can be explained 48 by the slow replacement, through weathering and deposition, of the base cations that were lost during 49 acidification. Although N deposition has decreased in some areas of Sweden, it has generally remained 50 constant (Pihl Karlsson et al. 2011). N accumulation in forest soils due to high loads of N deposition is 51 also preoccupying for soil acidification recovery. In forest soils with high N content, intense N leaching 52 episodes may occur following ecosystem disturbances such as clear-cutting (Gundersen et al. 2006; 53 Zanchi et al. 2014), or windthrow from storms (Legout et al. 2009). Intense N leaching episodes, for 54 which no causal explanation was found, have also been reported (van der Heijden et al. 2011). The 55 consequences of such episodes are increased acidity and increased aluminum concentrations in soil 56 solution, along with increased leaching of base cations and aluminum (e.g. Lundell et al. 2001). In the 57 long term, N leaching although episodic may strongly impact the recovery from soil acidification. 58 Natural ecosystem disturbances, such as storms, droughts and fire, may also have considerable effects on 59 the physical, chemical and biological properties of the soil, and thus the soil fertility. Storms are one of the 60 most common types of ecosystem disturbances in Swedish forests (Schlyter et al. 2006). Storms are high-61 wind episodes, sometimes accompanied by heavy rain, which may lead to a reduction in canopy closure 62 due to windthrow and stem breakage, thereby also increasing solar radiation levels in stands (Vodde et al. 63 2011). Storms may also lead to an increase in the availability of nutrients and water (Vygodskaya et al.

2002; Legout et al. 2009), as a result of the reduced uptake and the increase in woody debris which will decompose and release nutrients (Vodde et al. 2011). In fact, the stimulation of mineralization of organic matter, particularly the rate of nitrification, has been reported to be an important change resulting from ecosystem disturbances (Attiwill & Adams 1993; Dahlgren & Driscoll 1994; Legout et al. 2009). This is often accompanied by the leaching of nitrate and BC (Dahlgren & Driscoll 1994).

Apart from the effect of wind on trees, storms with high precipitation often cause sea-salt episodes. Sea-salt episodes affect the cation exchange in soil; and is known as the 'sea-salt effect' (Hindar et al. 1995). The cations transported by sea-salt episodes, primarily Na⁺ and Mg²⁺, cause the displacement of absorbed acid ions: hydrogen (H⁺), labile aluminum (Al³⁺) (Wright et al. 1988; Evans et al. 2001), and other BC: Ca²⁺ and K⁺ (Hindar et al. 1994; Hindar et al. 1995). The relationship between soil acidity and the leaching of H⁺, Al³⁺ and base cations is complex, but it is often observed that the pH falls and Al³⁺ increases and sea salt ions (Na⁺, Mg²⁺, and Cl⁻) increase shortly after sea-salt episodes (Hindar et al. 1995; Pedersen & Bille-Hansen 1995). Disturbances in acidification recovery as a result of sea-salt episodes have been observed in European and Swedish forests (Wright 2008; Akselsson et al. 2013; Laudon 2008). Wright observed that sea-salt episodes were responsible for more than one-third of the low ANC (ANC < -50 μeq l⁻¹) episodes, which were considered as strong indicator of acid episodes, at Birkenes in Norway during the period 1975–2004. Akselsson et al. (2013) and Laudon (2008) pointed out that recovery from acidification was slow at some coastal forest sites in Sweden, as these sites are susceptible to sea-salt episodes.

The aim of this study was to investigate the effects of storms on the acidification/recovery processes in forest soils. We hypothesized that: the high deposition of sea salt and increased rate of nutrient mineralization caused by storms slow down the recovery trend of forest soils from past acidification. For this purpose, a site in southwest Sweden that suffered the effects of the storms 'Lothar' and 'Gudrun' was studied. ForSAFE, a numerical model simulating biogeochemical cycling in forest ecosystems, was applied with monitoring data from the site. Both monitoring data and modelled data were used to study the acidification/recovery trends and the influence of the storms 'Lothar' and 'Gudrun' on these trends. Finally, ForSAFE was used to simulate the influence of frequently occurring storms in the future on soil acidification recovery.

2. Materials & Methods

2.1. Site description

The study site, was a 30 × 30 m square plot located in a forest called Klintaskogen (55° 62'N, 13° 44'E) in the region of Scania (southwest Sweden), and is part of the Swedish Throughfall Monitoring Network (SWETHRO) (Pihl Karlsson et al. 2011). It is also one of 223 intensive monitoring plots managed by the

- 97 Swedish Forest Agency. The average annual precipitation at Klintaskogen is 780mm and the average annual temperature is 7.2 °C (average over 1961–2010). The site is about 25 km from the west coast and about 30 km from the south coast of Sweden. The elevation of Klintaskogen is about 105 m, and it is on the southwestern slope of the horst Romeleåsen.
- Klintaskogen is a managed Norway spruce (*Picea abies* Karst) forest that was planted on a juniperous grass-land in the 19th century. The latest final felling occurred in 1957, and the site was replanted with Norway spruce. Klintaskogen suffered from the severe wind storm *Lothar* in Dec. 1999, which caused windthrow of about 15% of the trees in the study plot. Another wind storm, *Gudrun*, also caused damage at the study site in January 2005, but tree loss (about 5% of the trees) was less severe.
- The forest floor has a thin organic layer (3.5 cm) with 48% organic matter content (Loss on Ignition, LOI) (Table 1). The soil type is dystric regosol. The top 50 cm of the mineral soil profile is sandy and acidic with very low base saturation, and the exchange sites are mainly occupied by Al³⁺ (Table 1). The organic matter content decreases downwards from 10.8% to 2.3%, and the pH_{H2O} increases downwards from 4.2 to 5.0, within the top 50 cm of soil (Table 1).

2.2. Monitoring data

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- 112 The following parameters are monitored at the Klintaskogen site:
 - a) Deposition: Thoughfall was collected monthly between 1997 and 2009 with a total of ten throughfall collectors along two sides of the study plot. Bulk rainfall was collected monthly in an open-field collector between 1997 and 2001.
 - b) **Soil solution chemistry:** Soil solution was collected at 50 cm depth using tension cup lysimeters (5 replicates), three times per year from 1997 to 2009. The 5 sample obtained were combined into a composite sample on each sampling occasion for the analysis of: sulphate (SO₄²⁺), Cl⁻, nitrate (NO₃⁻), ammonium (NH₄⁺), Ca²⁺, Mg²⁺, Na⁺, K⁺, Mn²⁺, Fe³⁺, inorganic Al (iAl), organic Al (oAl) and dissolved organic carbon (DOC). Composite samples with volumes <50 ml were not analyzed. The ANC was computed as follows:
- 122 $ANC = [Ca^{2+}] + [Mg^{2+}] + [K^+] + [Na^+] + [NH_4^+] [SO_4^{2-}] [Cl^-] [NO_3^-]$ (1) 123 where the concentrations are in the units eq l^{-1} .
 - c) Forest inventories: Inventories were carried out on three occasions (1996, 1999 and 2004) within the intensive monitoring program led by the Swedish Forest Agency, and these inventories were complemented with an inventory in 2010, using the same methodology. On each occasion, the diameter at breast height of each tree was measured, and the tree and crown heights were measured in a selected aliquot representing 20% of the trees in the plot.

d) **Soil characteristics:** In 2010, soil samples were collected and C, N, pH, exchangeable cations, grain size distribution, bulk density and total element contents were determined. Total element analysis was accomplished by ALS Scandinavia AB in Agrilab Uppsala in 2010.

The properties of the soil at the study site are given in Table 1, and a detailed description of the SWETHRO network monitoring equipment and analysis protocols has been given in Pihl Karlsson et al.

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135 Table 1

136 Soil properties at Klintaskogen

Horizon	Layer thickness	Stoniness	s ^a	рН			Exch	angeabl	e ions, p	red/g			CEC ^b	BS	Tot- C	Tot- N	(Grain siz	ze ^c
	m	Fraction	H ₂ O	KCI	Fe	Mn	Al	Н	Na	K	Mg	Ca	10 ⁻⁵	%	g/Kg	g/Kg	Clay	Silt	Sand
													kmolc/kg		DS	DS			
0	0.035	0	4.1	3.2	3.5	12.1	27.6	47.9	<0,1	4.6	11.7	71.1	17.9	49.0	312.9	15.0	-	-	_
А	0.06	0.15	4.2	3.3	3.3	0.7	55.4	24.6	<0,1	1.4	1.8	7.5	9.47	11.2	61.5	3.8	4	24	72
AB	0.08	0.15	4.5	3.8	0.8	0.3	41.5	11.0	<0,1	0.7	0.6	1.8	5.67	5.0	32.4	2.2	4	24	72
В	0.16	0.15	4.8	4.2	0.2	0.1	21.1	3.9	<0,1	0.4	0.1	0.9	2.66	5.4	13.8	1.1	8	20	72
С	0.2	0.15	5.0	4.2	0.1	0.1	15.4	3.2	<0,1	0.5	0.4	2.0	2.17	13.4	10.4	0.9	6	18	76

^a The fraction of stones and boulders was assessed visually.

140 2.3. Calculations

- 141 *2.3.1. Tree biomass*
- The biomass of stems, branches, needles, and fine roots was estimated for each tree using Marklund functions (Marklund 1988). The biomass of each tree compartment (wood, needles, and fine roots) of each tree within the study plot were calculated and summed to provide an estimate of the standing biomass per square meter, where wood referred to the sum of stems and branches.
- 146 2.3.2. Atmospheric deposition in study plot
- The total atmospheric deposition (TD) was calculated as the sum of wet (bulk precipitation deposition) and dry deposition. For sulphate, nitrate, Na⁺ and Cl⁻, canopy exchange is assumed to be zero. Dry deposition is thus the difference between throughfall deposition and bulk precipitation deposition. The dry deposition of BC was computed using the Na dry deposition factor (Staelens et al. 2008):

$$151 DD_{\chi} = \frac{(TFD - PD)_{Na}}{PD_{Na}} PD_{\chi} (2)$$

where DD_x is the dry deposition of element x, TFD is the throughfall deposition, and PD is the open-field precipitation deposition.

^b CEC: cation exchange capacity; BS: base saturation.

^c Grain size is expressed in percent.

- Annual deposition was computed, except for years in which values of monthly throughfall and/or bulk
- precipitation data were missing. The total annual deposition of BC and Na⁺ was only available for 1998,
- 156 1999 and 2001; the total annual deposition of ammonium was only available from 1996 to 2001. Missing
- values were estimated as follows:
- 158 1) The main source of Mg²⁺ and Na⁺ deposition was assumed to be sea salt. Therefore, the average
- values of $\frac{TD_{Mg}}{TD_{Cl}}$ and $\frac{TD_{Na}}{TD_{Cl}}$ in 1998, 1999 and 2001 were used to estimate missing TD of Mg²⁺ and
- Na⁺ from measured TD of Cl⁻.
- 2) Ca^{2+} and K^{+} deposition was assumed to be constant over the period. The average TD for the years
- 162 1998, 1999 and 2001 were used to estimate missing values.
- 3) No PD measurements of BC or Na⁺ were available from 2002 to 2009, so it was not possible to
- calculate the TD of ammonium by canopy exchange model. Therefore, the downscaled data for
- ammonium from the European Monitoring and Evaluation Programme (EMEP) were used.
- 166 2.3.3. Statistical tools
- All statistical tests were carried out with the RStudio software (R Core Team 2013). Temporal trends in
- 168 atmospheric deposition and soil solution concentration were tested with the Mann-Kendall test of the
- Kendall R package (McLeod 2011). Correlations between major element concentrations in soil solution
- were tested with the Pearson test. For both tests, p<0.05 indicates statistical significance, and p<0.001
- indicates strong statistical significance.
- 172 2.4. The ForSAFE model
- 173 ForSAFE is a mechanistic model designed to simulate the dynamic responses of forest ecosystems to
- environmental changes (Figure 1). A full description of the model can be found in the literature (Belyazid
- 175 et al. 2006; Wallman et al. 2005).
- 176 Simulations with the ForSAFE model were run over the period 1800 to 2100, and the model was
- 177 calibrated over the period 1997–2009 using the monitoring data collected from the Klintaskogen site
- described above. The ForSAFE inputs (soil properties, climate, deposition and silviculture scenario) are
- described below, and the values used are given in Table 2 and Table 3.

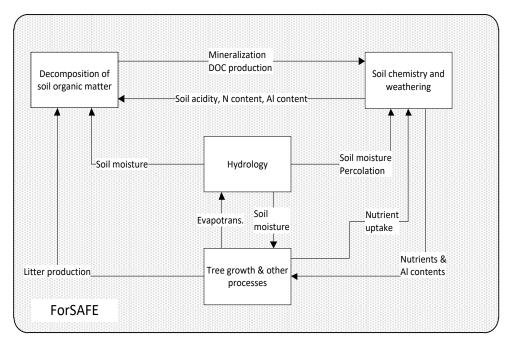


Figure 1. Layout of the ForSAFE model, showing the connections/interactions between the four sub-modules: decomposition, soil chemistry, hydrology and tree.

2.4.1. Soil inputs

The 50 cm soil profile was divided into 5 layers in accordance with the soil measurements (Table 2). Exposed surface areas of minerals were calculated using bulk density and grain size (Warfvinge & Sverdrup 1995; Sverdrup & Warfvinge 1993). The partial pressure of CO₂ (pCO₂) and the aluminium solubility (K_{gibb}) were derived from Warfvinge & Sverdrup (1993) and Sverdrup & Warfvinge (1995). Root fractions were derived for each layer based on the empirical data by Rosengren & Stjernquist (2004). Mineralogical regions from Warfvinge & Sverdrup (1995) were used to assess the potential mineral occurrence in the geographical area. Based on this, and the total element analysis, soil mineral composition was determined using A2M model (Posch & Kurz 2007) (Table 3).

The soil field saturation points for each soil layer were calculated as follows:

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$$FS = 1 - \frac{BD \cdot (1 - HML) \cdot LOI}{\rho_{organic}} - \frac{BD \times (1 - LOI)}{\rho_{particle}}$$
(3)

where FS is the field saturation ($m^3_{water} m^{-3}_{soil}$), BD is the bulk density (kg m⁻³), HML is the heat mass loss from 40 °C to 105 °C (fraction of weight), LOI is the loss on ignition (fraction of weight), $\rho_{organic}$ is the density of organic matter, assumed to be 600 kg m⁻³, and $\rho_{particle}$ is the density of sand particles, assumed to be 2670 kg m⁻³ (Milsom 2003). The field capacity (FC), wilting point (WP) were calculated using the

method described by Balland et al. (2008). The cation exchange capacity and base saturation were calculated from measured soil properties in Table 1.

200 2.4.2. Climate inputs

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The climate input data included precipitation, maximum, minimum & average temperature, atmospheric CO₂ concentration and radiation for the period 1800–2100. A weather station belonging to the Swedish Meteorological and Hydrological Institute (SMHI) situated about 12km southwest of the Klintaskogen site measured daily precipitation and temperature for the period 1961 to 2010 period. Precipitation and temperature data for the period 2010–2100 were derived from the global climate model HADLEY (Johns et al. 2006) follows the SRESA2 emission story-line of the Intergovernmental Panel on Climate Change (Nakicenovic et al. 2000). For the period 1900–1960, data were estimated by repeating the measured values for 1960–1990 twice. For the period 1800–1900, data were estimated by repeating the measured climate data for the year 1960. The CO₂ concentrations from 1970 to 2100 were derived from the SRESA2 scenario. The CO₂ concentration in 1800 was obtained by extrapolation, assuming that it remained constant before 1970. Solar radiation data for 1960–2100 were derived from a study performed by the Regional Climate Group at the University of Gothenburg (David Rayner, personal communication). Solar radiation data were extrapolated back to 1800 using the same scheme for precipitation and temperature data. To avoid the need to calibrate the modeled interception of precipitation, the precipitation data were further scaled to give the throughfall precipitation using the monitored throughfall precipitation factor:

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$$TFprec_{\chi} = \frac{TFprec_{1997-2009}}{Prec_{1997-2009}} Prec_{\chi}$$
 (4)

- where TFprec_x is the throughfall precipitation for month x, $Prec_x$ is the precipitation during month x,
- 218 TFprec₁₉₉₇₋₂₀₀₉ is the total throughfall precipitation monitored by the SWETHRO network from 1997 to
- 2009, and $Prec_{1997-2009}$ is the total precipitation obtained from the SMHI station from 1997 to 2009.

220 2.4.3. Deposition inputs

- The deposition input data included the atmospheric deposition of sulphate, nitrate, and ammonium, Ca²⁺,
- 222 Mg²⁺, K⁺, Cl⁻ and Na⁺ in intervals of one year. The Current Legislation (CLE) scenario and the
- 223 Gothenburg protocol for the UN Convention on Long-Range Transboundary Air Pollution provided by the
- 224 EMEP on a 50 × 50 km grid was used to estimate atmospheric deposition data during the period 1800–
- 2100. The EMEP data include deposition of sulphate, nitrate and ammonium from 1900 to 2100. EMEP
- data were downscaled by the average ratio of the measured deposition to the EMEP deposition (1997–
- 227 2009) for sulphate, nitrate and ammonium. Atmospheric deposition was assumed to be constant and equal
- 228 to the deposition in 1900 during the period 1800–1900. The deposition of Ca²⁺, Mg²⁺, K⁺, Cl⁻ and Na⁺ was
- assumed to be constant over the simulation period (1800–2100) and equal to the average measured

- deposition for each element. The yearly deposition input was converted to monthly amounts in ForSAFE
- assuming that the monthly deposition is proportional to the monthly amount of precipitation.
- 2.4.4. Storm disturbance and the future storm disturbances scenario
- Windthrow caused by the storms was implemented in ForSAFE by simulating thinning at the end of 1999
- to model the effects of *Lothar*, reducing the standing biomass by 15%, and thinning in 2005 to account for
- 235 Gudrun, reducing the standing biomass by 5%. The stimulation of organic matter mineralization in soils
- was implemented in the model by reducing the nutrient immobilization in microbial biomass after each
- storm. It was assumed that the reduction was greatest after storm, and the initial immobilization level was
- recovered after three years.

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- A scenario involving future storm disturbances (FSS) was included in the study for comparison with the
- baseline scenario (BLS). In the FSS scenario, it was assumed that a storm would occur every 20 years
- from 2005 onwards, causing 10% windthrow loss and stimulating organic matter mineralization (as
- described above) at each storm occurrence. All the storms are assumed not to be accompanied with sea-
- salt episodes. The difference in soil nutrient input-output budgets between the BLS and the FSS was
- determined using the model. No silvicultural thinning or harvesting was included in either scenario after
- the clear-cut in 1957 in order that only effects of storm are identified.

2.5. Nutrient input-output budget calculation

- Nitrogen and base cations input-output budgets were calculated over the period 1970–2070 for the two
- scenarios using the methodology applied in many other studies (Sverdrup et al. 2006; Akselsson et al.
- 249 2007; van der Heijden et al. 2013). Nutrient budgets were calculated as the difference between soil inputs
- and outputs. At the forest plot scale, nutrient budgets can be adapted from Ranger & Turpault (1999):

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$$B(N) = AD(N) + F(N) - U(N) - L(N)$$
 (5)

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$$B(BC) = AD(BC) + W(BC) - U(BC) - L(BC)$$
 (6)

- where B(x) is the nutrient budget of x, AD(x) is total annual atmospheric deposition of x, F(N) is the
- 254 fixation of N in the soil, assumed to be 0.12 kmolc ha⁻¹ y⁻¹ (DeLuca et al. 2002), and W(BC) is the
- weathering of BC. U(x) is the net accumulation of x in wood biomass, and L(x) is the leaching flux of x at
- 256 50 cm depth. All fluxes and budgets are expressed in kmolc ha⁻¹ y⁻¹. The plant uptake of N and BC,
- leaching of N and BC and weathering of BC were computed with ForSAFE.

Table 2ForSAFE inputs of soil properties for the plot at Klintaskogen

Layer	z ^{a, b} (m)	Density (kg m ⁻³)	Area (10 ⁶ m ⁻²)	pCO ₂	K_{gibb}	FC (m³water m ⁻³ soil)	WP (m³water m ⁻³ soil)	LP (m³water m ⁻³ soil)	FS (m³water m ⁻³ soil)	Roots fraction
1	0.035	195	0	10	6.5	0.410	0.150	0.258	0.687	0.2
2	0.051	709	754052	10	6.5	0.299	0.099	0.212	0.565	0.2
3	0.068	957	1017944	20	7.6	0.338	0.093	0.203	0.542	0.28
4	0.136	1393	1805718	20	8.6	0.332	0.089	0.180	0.481	0.22
5	0.17	1258	1389149	20	9.2	0.306	0.076	0.160	0.428	0.1

^a Data are based on the results of lab analysis of the soil samples listed in Table 1 (see text for detailed explanation).

Table 3
ForSAFE inputs of soil minerals for the Klintaskogen plot

		Plag	ioclase												
Layer	Quartz ^a	Albite	Anorthite	Hornblende	Illite	Epidote	Vermiculite1	Vermiculite2	Chlorite1	Chlorite2	Muscovite	Apatite	K-Feldspar	Hematite	Rutile
1	16.59 ^b	5.15	1.02	1.01	0.63	0.86	1.17	0.31	0.53	0.34	2.45	0.49	3.44	1.03	0.32
2	55.07	9.34	1.84	1.04	1.72	1.63	1.52	0.59	0.9	0.61	3.3	0.26	6.87	2.34	0.72
3	55.95	10.07	2.12	1.93	1.29	1.79	2.32	0.64	1.09	0.69	4.7	0.24	6.37	2.05	0.73
4	58.11	11.77	3.09	2.67	2.64	1.32	0.5	0.39	2.02	1.7	2.86	0.26	9.42	2.05	0.65
5	50.19	14.25	1.92	1.35	2.09	2.35	4.48	0.74	1.27	0.79	4.11	0.28	10.81	1.03	0.5

^a Quartz: SiO_2 , Albite: $NaAlSi_3O_8$, Anorthite: $CaAl_2Si_2O_8$, Hornblende: $K_{18}Na_{54}Ca_{166}Mg_{210}Fe_{180}Ti_{11}Al_{216}Si_{606}O_{2146}(OH)_{188}$,

Illite: $K_{0.6}Al_2(Al_{0.6}Si_{3.4}O_{10})(OH)_2$, Epidote: $Ca_{80}Fe_{30}Al_{96}Si_{124}O_{495}(OH)_{44}$, Vermiculite1: $Ca_{20}Mg_{103}Fe_{182}Al_{162}Si_{293}O_{832}(OH)_{804}$, Vermiculite2: $Ca_{10}Mg_{103}Fe_{22}Al_{68}Si_{123}O_{249}(OH)_{490}$, Chlorite1: $Na_2Ca_3Mg_{107}Fe_{124}TiAl_{124}Si_{138}O_{540}(OH)_{442}$,

Chlorite2: $Mg_{103}Fe_{58}TiAl_{100}Si_{87}O_{365}(OH)_{302}$, Muscovite: $K_{44}Na_2Mg_8Fe_{12}Ti_2Al_{96}Si_{120}O_{390}(OH)_{94}$, Apatite: $Ca_{10}(PO_4)_6(OH)_2$, K-Feldspar: KAlSi₃O₈, Hematite: Fe_2O_3 , Rutile: TiO_2 . (Warfvinge & Sverdrup 1995)

^b z is the corrected layer depth; Density is bulk density; Area is exposed surface area of minerals; The other abbreviations have been defined in the text.

^b the values given are the percent of the total weight in the soil layer. Water content and organic material content are not presented in the table.

3. Results

3.1. Monitoring data from the study plot

3.1.1. Atmospheric deposition 1997–2009

The total deposition of Cl $^-$, sulphate, nitrate and ammonium is shown in Figure 2. Chloride was the most abundant anion in atmospheric deposition, with an average deposition rate of 1.33 kmolc ha $^{-1}$ y $^{-1}$. Sulphate was the second most abundant anion with an average deposition rate of 0.35 kmolc ha $^{-1}$ y $^{-1}$. The total N deposition was 1.39 kmolc ha $^{-1}$ y $^{-1}$ (nitrate: 0.66 kmolc ha $^{-1}$ y $^{-1}$; ammonium: 0.73 kmolc ha $^{-1}$ y $^{-1}$). Cl $^{-1}$ deposition showed a peak in 2000 (3.76 kmolc ha $^{-1}$ y $^{-1}$), and a small increase after 2006. Sulphate deposition decreased significantly (τ =-0.684, p=0.001) from about 0.5 kmolc ha $^{-1}$ y $^{-1}$ in 1997 to about 0.25 kmolc ha $^{-1}$ y $^{-1}$ in 2009. Ammonium deposition also decreased significantly, from 0.88 kmolc ha $^{-1}$ y $^{-1}$ to 0.63 kmolc ha $^{-1}$ y $^{-1}$, over the same period (τ =-0.615, p=0.004). However, nitrate deposition and total N deposition were relatively stable over the period.

Total atmospheric deposition

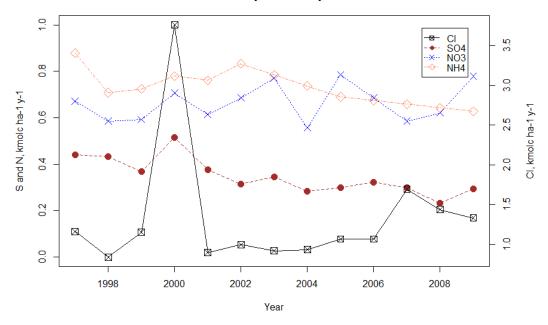


Figure 2. Total atmospheric deposition of Cl⁻, sulphate, nitrate and ammonium over the period 1997–2009 at the study plot in Klintaskogen.

Total atmospheric deposition of Na⁺, Ca²⁺, Mg²⁺ and K⁺ is given in Table 4. The average Na⁺ deposition for the years 1998, 1999 and 2001 was 0.84 kmolc ha⁻¹ y⁻¹, while the average deposition of Ca²⁺, Mg²⁺ and K⁺ over the same period were 0.095, 0.10 and 0.057 kmolc ha⁻¹ y⁻¹, respectively. The monthly deposition (data not shown) of K⁺ showed a decreasing tendency over the period 1998–2001 (τ =–0.306,

p=0.007), and the monthly deposition of Ca^{2+} also seemed to decrease however this visual trend was not statistically significant (τ =-0.215, p=0.06). No significant trends were seen in the monthly Na^{+} and Mg^{2+} deposition over the same period.

Table 4
 Total atmospheric deposition of Na, Ca, Mg and K (kmolc ha⁻¹ y⁻¹)

Year	Na	Ca	Mg	K
1998	0.72	0.092	0.092	0.065
1999	1.07	0.10	0.14	0.067
2001	0.73	0.094	0.080	0.039
Average	0.84	0.095	0.10	0.057

3.1.2. Soil solution chemistry 1997–2009

The soil water at the study site over the period 1997–2009 was acidic. The pH ranged from 3.86 to 4.54, and the ANC was negative, ranging from about -600 to about $-100 \,\mu\text{eq}\,\text{I}^{-1}$, as can be seen in Figure 3. The dominant anion was Cl⁻, and the dominant cation was Na⁺. The concentrations of Na⁺ and Cl⁻ increased significantly over the whole period monitored (Cl, τ =0.448, p<0.001; Na, τ =0.458, p<0.001), while Ca²⁺ and ANC decreased (Ca, τ =-0.292, p=0.02; ANC, τ =-0.3, p=0.02). However, the trend analysis before *Lothar* and after *Lothar* gave different results. Before *Lothar*, the nitrate concentration decreased (τ =-0.491, p=0.04). After *Lothar*, the pH and Ca²⁺ concentration decreased significantly (pH, τ =-0.602, p<0.001; Ca, τ =-0.568, p<0.001), while Cl⁻ and DOC increased significantly (Cl, τ =0.368, p=0.03; DOC, τ =0.459, p=0.02). High concentrations of Cl⁻, nitrate, sulphate, Na⁺, Ca²⁺, Mg²⁺ and oAl were noted in 2000.

Over the period monitored, the Cl⁻ concentration was significantly positively correlated with the concentrations of Na⁺ and Mg²⁺ (Na: R²=0.746, p<0.001; Mg: R²=0.606, p<0.001), but not the Ca²⁺ and K⁺ concentrations. Interestingly, Cl⁻ showed the same positive correlations with Na⁺ and Mg²⁺ after *Lothar* (Na: R²=0.615, p=0.004; Mg: R²=0.522, p=0.02), but it was not correlated with any cations before *Lothar*. The nitrate concentration was positively correlated with Na⁺, Mg²⁺ and Ca²⁺ (Na: R²=0.491, p=0.006; Mg: R²=0.800, p<0.001; Ca: R²=0.526, p=0.003) over the period 1997–2009. Both Mg²⁺ and Ca²⁺ showed positive correlations with nitrate before and after *Lothar*, but no other correlations with nitrate were found before or after *Lothar* for the other elements. The pH was significantly negatively correlated with DOC concentration (DOC: R²=0.651, p<0.001) during the period 1997–2009. The pH was also negatively correlated with DOC both before and after *Lothar*.

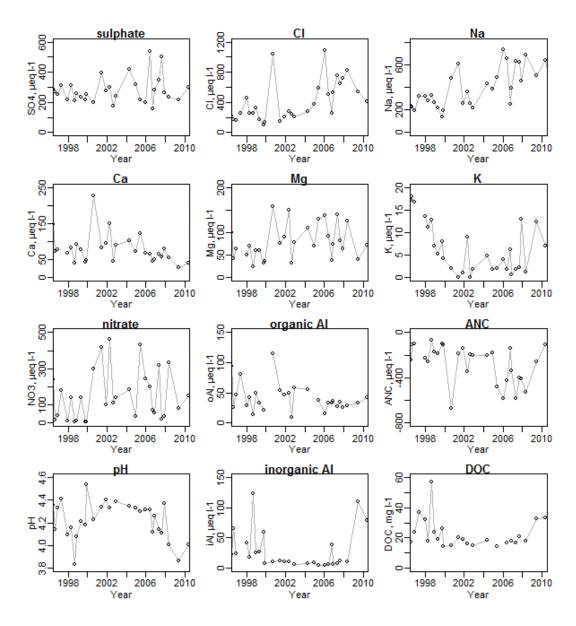


Figure 3. Soil solution concentrations of major chemical species monitored (sulphate, Cl^- , nitrate, DOC, Na^+ , oAl, iAl, Ca^{2+} , Mg^{2+} , K^+), ANC and pH at a depth of 50 cm in the Klintaskogen site plot, from 1997–2009.

3.2. Model validation and scenarios

3.2.1. Model validation

Model validation showed good agreement with the monitored data. The model wood biomass before *Lothar*, and the trend of wood biomass change after *Lothar* agreed well with inventory data (Figure 4). For the simulation of soil water chemistry over the period 1997–2009, the measured concentrations of all chemicals were in the range of modeled concentrations, and ForSAFE also captured the temporal dynamics of concentrations for most of chemicals (Figure 5). The model captured two high concentration episodes of N after *Lothar* and after *Gudrun* respectively, and the overall modeled N concentration over

the period 1997–2009 almost equaled the measured value. The model predicted a decrease in sulphate concentration, but a few measurements outranged the modeled concentration after *Gudrun*. The model simulated two high concentration episodes of BC, one after *Lothar* and one after *Gudrun*, which agreed well with monitored data. But the overall modeled BC concentration was about one-third higher than the measured value. The model reproduced the temporal dynamics of Cl⁻ concentration, and the overall modeled Cl⁻ was only 5% more than that of monitored. The modeled Na⁺ concentration was generally higher than the monitored over the period 1997–2009.

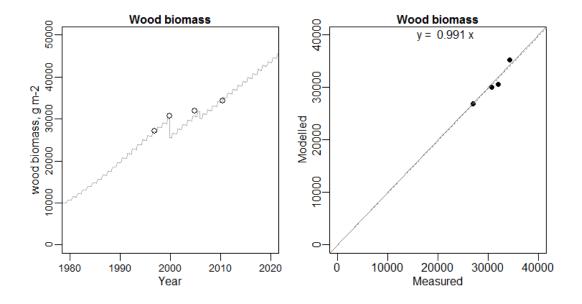


Figure 4. Measured vs. modelled wood biomass. In the left sub-plot, monthly modelled results (grey line) are compared to measured data (points) over the years; in the right sub-plot, the modelled results and measured data are plotted against each other. The dotted line and its slope reported in the equation (y = ax) are an indication of the discrepancy between modelled and measured data.

However, some discrepancies were observed in the modeling of acidity in soil water. The model overestimated the pH over the period 1997–2009, and the discrepancies were greater before 2000 and after 2008 (Figure 5). The prediction of ANC, an important indicator of acidity, well presented the temporal dynamics of measured ANC, but the overall modeled ANC was about 20% higher than the measured value. The DOC concentration, which has a significant effect on pH, was lower in the model than in the measured data. Moreover, the shape of the measured DOC concentration curve was significantly different from the modeled curve.

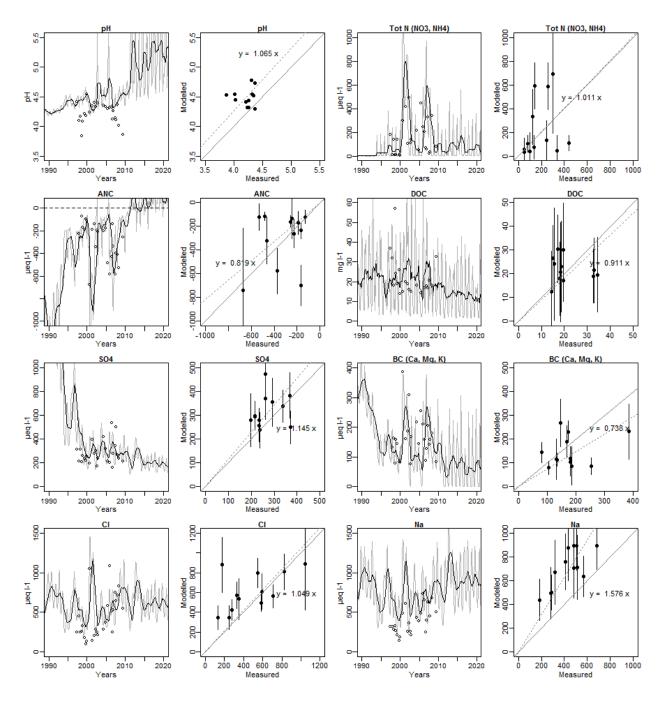


Figure 5. Comparison of modeled and measured soil water chemistry data. The grey curves show the modelled monthly values, and the black lines the moving averages (12-month periods) of these values. ○ − measurements, and ● −yearly mean values. The dotted line and its slope reported in the equation (y = ax) are an indication of the discrepancy between modelled and measured data. The soil water was collected at a depth of 50 cm.

3.2.2. Model scenarios and soil nutrient budget

Clear recovery from the acidification of the 1990s was seen in both the scenarios modelled (Figure 6). This was indicated by a substantial increase in soil solution ANC from $-1000 \,\mu\text{eq} \, l^{-1}$ to $+100 \,\mu\text{eq} \, l^{-1}$, and

by an substantial increase in soil matrix exchangeable base cations from 5 keq ha⁻¹ to 35 keq ha⁻¹. A long-term decreasing trend was observed in the modeled BC concentration. The decrease in BC started in the 1980s, the concentration decreased from about 1500 μ eq I⁻¹ to about 100 μ eq I⁻¹ in the 2000s, by which the modeled BC concentration had reached baseline level.

Significant differences were predicted in pH, soil solution N concentration and BC concentration between the BLS scenario and the FSS scenario. Although a long-term increase in pH was predicted in the FSS scenario, strong acidic episodes were seen as the result of storms. Episodes of high concentrations of soil solution N and BC were predicted simultaneously in the FSS scenario in response to storms. Higher soil solution DOC and lower exchangeable BC in soil matrix were also identified in the FSS scenario due to storms. The storm-induced changes in ANC were less profound when comparing the FSS scenario with the BLS scenario.

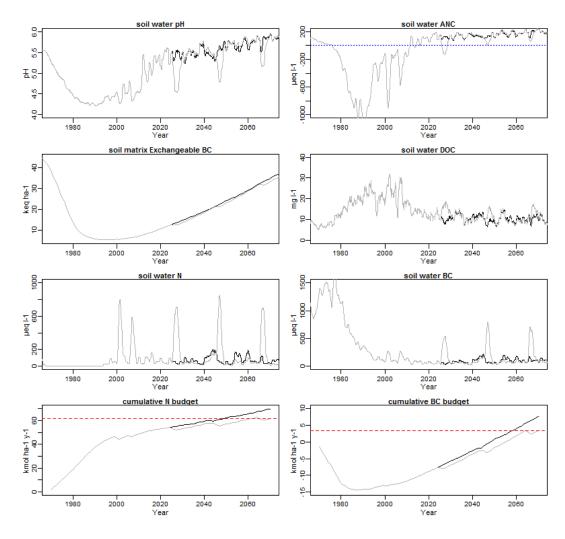


Figure 6. Effects of future storm disturbances on soil (BLS: black line; FSS: gray line). Soil water pH, ANC, BC and N concentrations are at 50 cm depth. Exchangeable BC of soil matrix is at layer 5 (Table 2). Cumulative N and BC budget is based on section 2.5, the red dotted lines go through the end of grey lines horizontally.

N accumulated in the soil in both scenarios over the simulation period 1970–2070 (Figure 6). The rates of accumulation (the slopes of the curve) in most years were positive, but after each storm disturbance, there were a few years with negative values. The rates started to decrease from 1990s and reached a relative stable value after 2010s. Base cations were lost from the soil from 1970, and they started to accumulate from the 1990s, i.e. the rates of accumulation became positive. The rates stabilized after 2010s, and they also shortly became negative after each storm as N did. Differences of cumulative N and BC input-output budget between FSS and BLS could be seen after the first future storm (2025), and they increased after each storm. At 2070, cumulative N and BC budgets were 69.82 and 7.54 kmolc ha⁻¹ for BLS, and for FSS they were 61.64 and 3.41 kmolc ha⁻¹. The BLS reached FSS's 2070 level in 2050 and 2058 separately, in other words, the accumulation of N and BC in soil were delayed by storms for about 20 years and 12 years.

4. Discussion

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- 4.1. Recovery from acidification at the Klintaskogen site before Lothar?
- 378 Klintaskogen is one of the few sites in Sweden which is saturated with both S (Eriksson et al. 1992) and N
- 379 (Akselsson et al. 2010; Pihl Karlsson et al. 2011) due to high historical acid deposition. We observed a 50%
- decrease of S deposition from 1997 to 2009, but there was no significant change in N deposition over the
- period (Figure 2). The change of acid deposition at Klintaskogen over the period 1997–2009 is in line with
- the findings in many other sites in Sweden (Pihl Karlsson et al. 2011; Akselsson et al. 2013). One
- consequence of the decreasing sulphate deposition is a reduction in the sulphate concentration in soil water.
- However, it doesn't necessarily lead to an immediate recovery from the acidification in forest soils (Pihl
- 385 Karlsson et al. 2011; Akselsson et al. 2013), probably due to the problem of sulphate adsorption
- reversibility (Reuss & Johnson 1986) and the complexity of acidification and alkalization in soils (Van
- 387 Breemen et al. 1983). We did not observe significant trends of acidification recovery in soil water before
- 388 *Lothar* in monitored pH, ANC or iAl (Figure 3).
- However, the model simulation clearly showed an increase of pH and a decrease of ANC from 1990s
- 390 (Figure 5 and Figure 6). The main driver for the recovery was the decreased sulphate concentration in the
- soil water, but another important factor was that no disturbances were simulated between 1990 and 2000.
- 392 Therefore we can argue that Klintaskogen should recover from acidification from 1990s if no external
- disturbances happen.
- 4.2. The influence of Lothar and Gudrun storms on recovery from acidification during 1997–2009
- 395 *4.2.1. Influence of storms on soil chemistry*
- We observed frequent high-concentration episodes of nitrates, Cl⁻, Na⁺ and Mg²⁺ after *Lothar* and *Gudrun*,
- 397 which could be due to a combination of the sea-salt effect and stimulated mineralization. The temporary

- 398 changes in pH, Al3+, Na+, Cl- and Mg2+ measured at the study site after Lothar agree well with the
- characteristics of the sea-salt effect, which is also supported by the high peak of Cl⁻ deposition in 1999.
- 400 The continuous high nitrate concentration could probably be explained by an increase in the
- 401 mineralization rate due to canopy opening, radiation and temperature change (Vodde et al. 2011).
- 402 However, soil water chemistry changed slightly after Gudrun. This was a storm with high winds but
- 403 without heavy precipitation, as could be observed from the monitored throughfall precipitation (data not
- shown) and total deposition. Therefore, no evidence of sea-salt effect was found in 2005, whereas
- episodes of remarkably high Cl⁻, Na⁺ and Mg²⁺ concentrations in 2007 and 2008 indicated two new sea-
- salt episodes. However, the Ca²⁺ concentration did not increase in the two new sea-salt episodes, and we
- speculate that this may be due to the greater difficulty for Na⁺ to replace Ca²⁺ in more acidified soil
- 408 (Hindar et al. 1995; Hindar et al. 1994). But the high-nitrate concentration episode had already started in
- 409 2005, showing that the increase in mineralization started directly after *Gudrun*.
- The sulphate seems to decrease before Lothar (τ =-0.424, p=0.06). Visual inspection of the data after
- 411 Lothar gives the impression that the amount of sulphate has decreased, but high-concentration episodes
- occurred more frequently after Lothar, disturbing this trend. The reason for the high-concentration
- 413 episodes could be increased sulphur mineralization from organic matter after storms. This suggests that the
- long-term decrease in sulphate concentration will be disturbed by future storms.
- 4.2.2. *The influence of storms on soil and water acidity*
- 416 Higher nitrate leaching was observed after both storms, which are probably due to an increase in organic
- 417 matter mineralization and an increase in nitrification. It is known that nitrification, in particular when
- 418 excess production of nitrate (not consumed by plants) occurs, has a very strong acidifying effect (Reuss &
- Johnson 1986). Meanwhile, nitrate leaching causes base cations leaching.
- 420 Although high-concentration episodes of sulphate occurred after *Lothar*, sulphate was not correlated with
- any cation or ANC, indicating that sulphate is not the main anion regulating ANC or leaching base cations
- 422 at Klintaskogen.
- Sea-salt episodes were observed after *Lothar* and in 2007 and 2008 at Klintaskogen. They are believed to
- be of little long-term significance since there is often a recovery in soil water after the sea-salt episode
- 425 temporarily acidifies it (Hindar et al. 1995). It is because that the increased H⁺ and Al³⁺ in soil water are
- actually exchanged from soil matrix, i.e. the total acidity of the whole soil does not increase in a sea-salt
- 427 episode. Moreover, the recovery from soil acidification may be accelerated due to the additional base
- cations from the sea-salt episodes (Beier et al. 2003). We observed high correlation between Cl⁻ and Mg²⁺,

indicating that sea-salt episodes also brought a lot of Mg²⁺. Thus sea-salt episode will probably decrease the soil acidity of Klintaskogen on long term since it replenishes the base saturation.

The changes in soil water acidity between 1997 and 2009 reflect the multiple effects of storms, as well as other factors, such as DOC. Before *Lothar*, the variation in pH was driven by the DOC concentration, and indicated by the iAl concentration (Figure 3). The sudden decrease of pH directly after *Lothar* and the subsequent recovery from acidity up till 2002 was explained by the sea-salt effect (Hindar et al. 1995; Evans et al. 2001). Similar temporary acidification and recovery were also identified in 2007 and 2008, when smaller sea-salt episodes were reported also verified by the monitored deposition. The changes in acidity between 2002 and 2007 were generally related to the changes in ANC, with the exception of 2006, when an increment in ANC caused a decrease in pH. This was also characterized by very high Cl⁻, Na⁺ and Mg²⁺ concentrations, indicating a sea-salt episode. However, there was no evidence of a storm in terms of high precipitation or reports by meteorological institutions, and the reason for this exception is still unclear. After 2009, the concentration of DOC increased again, causing another acidification episode.

Overall, there was no significant acidification recovery over the period 1997–2009. In contrary, the measured pH decreased significantly due to storms, from 4.54 in Nov. 1999 to 3.86 in May 2009. Given the fact that sea-salt episode has little long-term influence on acidification, the decreased pH is probably driven by nitrification and leaching of nitrate. Similar decreasing trends of pH were also observed in two other southern Swedish Norway spruce forest sites after 2000, which were both very close to the west coast of Sweden (Akselsson et al. 2013). It indicates that storm disturbances, at sites in close proximity to sea, would cause delay in the acidification recovery. Meanwhile, the considerable effects of DOC concentrations on pH suggest that DOC is also a very important factor and should be included in long-term acidification studies. Visual analysis of the data in this study and the data in Akselsson's study suggests that soil water that contains DOC>20 mg l⁻¹ would have much higher pH sensitivity to DOC.

4.3. Long-term effects of frequent storms on future acidification recovery and fertility loss

4.3.1. Reliability of ForSAFE prediction

Acidity: The pH curve obtained with the ForSAFE model exhibited a different shape from the measurements over the monitored period. The model captured the relationship between pH and ANC, but did not predict the strong impact of DOC on pH very well. Neither was the concentration of DOC well predicted. In addition, the pH change caused by sea-salt effect is not readily included in the model yet. Therefore, the difference between the modelled and the measured pH in some cases was larger than half a unit over the period 1997–2009. However, the modeled ANC was well validated by the measured data over the period.

- In the long term, the effects of sea-salt episodes on acidity is little, and the fact that storms will decrease the ANC in the soil water, regardless the presence of sea-salt episodes, is confirmed by both monitored data and modeled results. Thus the only uncertainty in the long-term modeled pH is DOC. By assuming that no significant change would happen in the DOC concentration on long term in southern Sweden, it is suggested that the modeled long-term trend of acidity change at Klintaskogen is reliable.
- 466 Nutrient concentrations: Both Lothar and Gudrun caused increases in concentrations of N and BC 467 (Figure 2 and Figure 3), thus increases in leaching and loss of N and BC from the soil (Figure 6). The increased leaching of N after both storms was most probably due to the stimulated mineralization (Legout 468 et al. 2009). The increased leaching of Mg²⁺ after both storms was most probably due to the sea-salt effect, 469 but after Gudrun, it was caused by two sea-salt episodes in 2007 and 2008, rather than the storm Gudrun. 470 The increased leaching of Ca²⁺ after *Lothar* was due to the sea-salt effect, but after *Gudrun* the leaching of 471 Ca²⁺ decreased to the level before *Lothar*, and we speculate that this may be due to the greater difficulty 472 for Na⁺ to replace Ca²⁺ in more acidified soil (Hindar et al. 1995; Hindar et al. 1994). The leaching of K⁺ 473 was very small, compared with those of Mg²⁺ and Ca²⁺. 474
 - ForSAFE well simulated the concentration and temporal dynamics of soil solution N, but some discrepancies were seen in soil solution BC. One important reason is that sea-salt effect was not simulated by the ForSAFE model: Firstly, the base cations are lumped as an equivalent base and are assumed to react as a divalent component BC²⁺ in the soil chemistry module of the model. But sea-salt episodes bring a lot of Mg²⁺ but not Ca²⁺ and the vertical reactive transport of these two elements in the soil profile are very different most especially when the soil contains a high organic matter content (van der Heijden et al. 2014).; And secondly, Na⁺ is not currently included in the ion-exchange process in the model. Lumped BC²⁺ is satisfactory when studying soil acidification. However finer detail is required to better simulate the nutrient budget, but we believe the model is sufficient for the purposes of this study to obtain an overview of the long-term trend.
- 485 4.3.2. Long-term effects of frequent storms on acidification/recovery & fertility

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ForSAFE predicts that the Klintaskogen site will slowly recover from acidification in the future. Predicted recovery was slower than prior to 1990s. This recovery is driven by the reduction of S deposition. However, many studies have reported decreasing base cations deposition like Ca²⁺ in north America and Europe over the past decades (Hedin et al. 1994; Watmough et al. 2005; Alewell et al. 2000). This was not simulated here, because no clear evidence of Ca²⁺ deposition reductions was available (Ca²⁺ trend not significant but p=0.06). If Ca²⁺ deposition also decreases in the future, this may further slowdown or invert the recovery trend.

The simulation of frequent storms has effects on the soil solution chemistry and N and BC leaching fluxes during the few years after each storm. This caused increased losses of N and BC compared to the BLS scenario (Figure 6). The leaching of nitrate would have strong acidifying effect on soil water and delayed N accumulation in soil, but this would not be an issue for the N fertility of the soil because big amounts of N would accumulated in soils in both scenarios. The leaching of BC would decrease the exchangeable BC of soil, more problematically, lead to severe loss of BC fertility. The future storms (FSS) lead to loss of 12 years' accumulation of BC fertility comparing to BLS, which is almost 50% of the total accumulation over the period 1970–2070. However, the recovery indicators (pH, ANC, BC concentration, soil exchangeable BC) in the FSS scenario are not influenced in the long term compared to the BLS scenario, although a temporary acidification episode would be seen after each storm. Furthermore, the comparison between the two scenarios showed that these recovery indicators need about five years or longer to recover from storm disturbances, which was much longer than the observed time to recover from a sea-salt episode. It indicates that the influences of mineralization on acidification recovery are stronger than that of sea-salt episode.

Overall, model simulation suggests that storms without sea-salt episodes don't seem to disturb acidification recovery at Klintaskogen on long term. It is in agreement with the finding of Thiffault et al. (2007), that storm disturbances appear not to be the long-term driver of soil chemistry. However, storms raise risks in depleting the BC in the soil. At sites with lower exchangeable pools of Mg²⁺, Ca²⁺ and K⁺, the impact may be more visible.

5. Conclusions

- The monitoring and the modeled data support the hypothesis that ecosystem disturbances such as storms may strongly influence acidification recovery processes.
- There was no monitored evidence proving Klintaskogen was recovering from acidification before 2000, but model simulation implied a trend of recovery. The two storms *Lothar* and *Gudrun* strongly disturbed the recovery trend, in fact over the period 1997–2009 some indicators (pH, ANC and Ca²⁺ concentration) suggested that the soil acidification occurred during that period.
 - Storms occurring with both strong winds (windthrow) and high sea-salt inputs (sea-salt episodes) have an ambiguous effect on acidification recovery. Windthrow and the opening up of the canopy led to increased mineralization/nitrification, thus nitrate and base cations leaching, causing acidification and decreased base saturation; the sea-salt episode led to increased soil water acidity and aluminum concentrations but also possibly replenished the soil cation exchange pool with Mg²⁺, thus on long term increasing the soil pH buffer capacity.

• No evidence that frequently (20 years) occurring storms with windthrow hamper acidification recovery but storms may influence soil fertility by losses of base cations. This may be a concern for the most base cation poor forest soils.

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