

Modeling Acidification Recovery on Threatened Ecosystems: Application to the Evaluation of the Gothenburg Protocol in France

David Moncoulon · Anne Probst · Liisa Martinson

Abstract To evaluate the acid deposition reduction negotiated for 2010 within the UNECE LRTAP Gothenburg Protocol, sulphur and nitrogen deposition time-series (1880–2100) were compared to critical loads of acidity on five French ecosystems: Massif Central basalt (site 1) and granite (2); Paris Bassin tertiary sands (3); Vosges mountains sandstone (4) and Landes eolian sands (5). The SAFE model was used to estimate the response of soil solution pH and $\frac{[Al]}{[BC]}$ ratio to the deposition scenario. Among the five sites, critical loads were exceeded in the past at sites 3, 4 and 5. Sites 3 and 4 were still expected to exceed in 2010, the Protocol year. Further reduction of atmospheric deposition, mainly nitrogen, would be needed to achieve recovery on these ecosystems. At sites 3, 4 and 5, the delay

between the critical load exceedance and the violation of the critical chemical criterion was estimated to be 10 to 30 years in the top soil and 50 to 90 years in the deeper soil. At site 5, a recovery was expected in the top soil in 2010 with a time lag of 10 years. Unexpectedly, soil pH continued to decrease after 1980 in the deeper soil at sites 2 and 5. This time lag indicated that acidification moved down the soil profile as a consequence of slow base cation depletion by ion exchange. This delayed response of the soil solution was the result of the combination of weathering rates and vegetation uptake but also of the relative ratio between base cation deposition and acid compounds.

Keywords acidification · atmospheric deposition · France · recovery · SAFE model

D. Moncoulon · A. Probst (✉)
Laboratoire des Mécanismes de Transfert en Géologie,
UMR 5563, CNRS-IRD-Université Paul Sabatier,
14 avenue Edouard Belin, 31400 Toulouse, France
e-mail: anne.probst@ensat.fr

L. Martinson
Lund University Centre for Sustainability Studies,
P.O. Box 170, 221 00 Lund, Sweden

Present address:

A. Probst
ECOLAB UMR 5245,
CNRS-INPT-Université Paul Sabatier,
ENSAT Avenue de l'Agrobiopole,
BP 32607 Auzeville, Tolosane,
31326 Castanet, Tolosan, Cedex, France

1 Introduction

Acidification of soil and water, following the significant increase in sulphur and nitrogen deposition on terrestrial and aquatic ecosystems have been evidenced by many authors in North and Central Europe since the 1970s (e.g. Blank, 1985; Wright & Snekvik, 1978). In France, the exposed areas were located in the Center, North and North-East of the country (Landmann & Bonneau, 1995; Probst, Massabuau, Probst, & Fritz, 1990). The most threatened ecosystems were mainly characterized by low soil weathering rate and the important buffering effect of atmospheric base cation

deposition as well as base cation uptake due to forest productivity on French ecosystems has previously been estimated (Moncoulon, Probst, & Party, 2004). Since the 1980s, a series of protocols under the UN/ECE Convention on Long-Range Transboundary Air Pollution (LRTAP) have been negotiated to reduce atmospheric pollution over Europe. Sulphur deposition has reduced by 83% between 1980 and 2005 in metropolitan France. Over the same period, NO₃ decreased by 33% and NH₄ remained constant (CITEPA, 2003). Since 1999, a new protocol for sulphur and nitrogen (the so-called Gothenburg protocol) has been negotiated to reduce these emissions by the year 2010. In this post-acidification context, to determine the ecosystem behaviour, five representative sites of the french territory were chosen. The objectives of this study were (1) to identify among five ecosystems, which have been exposed to a critical acid pollution in the last decades; (2) to estimate the effects of acid pollution decrease on these ecosystems, estimated by calculating the pH and $\frac{[Al]}{[BC]}$ ratio in the soil solution; (3) to evaluate the acid deposition reductions negotiated within the Gothenburg protocol. Critical loads were defined as “the quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to the present knowledge” (Nilsson & Grennfelt, 1988). To achieve our goals, acid atmospheric deposition time series over the 1880–2100 period have been compared to critical loads to determine the exceedance areas. Dynamic modelling has been applied to evaluate the impact of acidification on soil chemical criterions and recovery.

2 Material and Methods

2.1 Site Description

A new ecosystem classification has been set up at the French territory scale to integrate soil, bedrock and vegetation data for critical load calculations. 281 ecosystems have been defined among which 241 concern forests (Moncoulon et al., 2004). In order to take into account the variability of the French ecosystem sensitivity to acidification, five sites were selected, reflecting different combinations of bedrocks, soils and

vegetation species (Table 1). Sites 1 and 2, located in the Massif Central, were both subjected to Mediterranean influence and remoteness from highest pollution sources but differed strongly by their bedrock types (basalt and granite). Site 3, in the Center of France, was close to pollution sources and characterized by inert parent material (sand) and remoteness from Mediterranean influence. Site 4 was closer to high pollution sources from Central Europe, and characterized by sandstoneous bedrock type. Site 5, a sandy soil located near the Atlantic Ocean, receive high seasalt cations and was far from high pollution sources. The vegetation on sites 1, 2, 3 and 4 was planted by deciduous forest whereas on site 5, the forest was coniferous.

2.2 Critical Load Calculation

Critical loads were calculated at the five sites using a mass balance approach in steady state conditions for the soil solution down to one meter soil depth (Hettelingh, Posch, & De Amet, 2001). For each ecosystem, one indicator species was chosen (Table 1). The critical limiting value of pH and $\frac{[Al]}{[BC]}$ were determined from soil solution of French ICP Forest network for non significant health effect on trees: $[H^+]_{crit} = 25 \mu\text{eq l}^{-1}$, which corresponded to pH=4.6 and $\frac{[Al]}{[BC]} = 1.2$ (Moncoulon et al., 2004; Party, 1999). This value, which corresponded to coniferous conditions (*Picea abies* L.) was used for the acid forest soils in general. However a comparison has been also performed with the critical limits for $\frac{[Al]}{[BC]}$, specific of the different plant species, found in the literature (Sverdrup & Warfvinge, 1993). The critical deposition of sulfur was calculated as:

$$CL_{\max}(S) = BC_{dep} + BC_{weath} - BC_{upt} - Ac_{le(crit)} \quad (1)$$

$$Ac_{le(crit)} = -Q \times [H^+]_{crit} - 1.2 \times \left(\frac{[Al]}{[BC]} \right)_{crit} \times (Q \times [BC]) \quad (2)$$

To determine the critical load for nitrogen, sulphur deposition had to be fixed. If sulfur deposition was equal to critical load:

$$Cl_{\min}(N) = N_{imm} + N_{upt} \quad (3)$$

Table 1 Description and location of the five studied sites

Site	1	2	3	4	5
Location	Massif Central	Massif Central	Paris bassin	Vosges mountains	Landes
Bedrock	Basalt	Granite	Tertiary sands	Sandstone	Eolian sands
Soil	Andosol	Dystric cambisol	Podzoluvisol	Podzol	Podzol
Indicator species	Fagus sylvatica L.	Quercus humilis Mil.	Carpinus betulus L.	Fagus sylvatica L.	Pinus pinaster Ait.
Forest acid soil $\frac{[Al]}{[BC]}$ critical ratio	1.2	1.2	1.2	1.2	1.2
$\frac{[Al]}{[BC]}$ critical ratio for the indicator species	1.6	1.6	0.85	1.6	0.85

If there was no sulfur deposition:

$$CL_{\max}(N) = N_{\text{imm}} + N_{\text{upt}} + CL_{\max}(S)/(1 - F_{\text{de}}) \quad (4)$$

CL was the critical load, BC base cation, $_{\text{dep}}$ atmospheric deposition, $_{\text{weath}}$ weathering rate, $_{\text{upt}}$ vegetation uptake, $_{\text{crit}}$ critical limit, Ac_{le} acidity leached, Q flux of water percolating in the soil, $_{\text{imm}}$ immobilisation, F_{de} denitrification factor (0 to 1). All fluxes were eq ha⁻¹ year⁻¹.

2.3 Determination of Exceedance

Exceedance was determined by comparing sulfur and nitrogen deposition to critical loads of sulfur and nitrogen (UBA, 2004) calculated with different critical limits. Deposition pairs (S and N) was plotted in Fig. 1 and compared with the critical load functions, which represented the link between the three critical loads on the (S, N) graph. On each point of the critical load function, the pollutant pair (S and N) represented a critical acid deposition. Exceedance existed if the plot was in region 2 of the graph (over the critical load function) for the chosen indicator species.

2.4 The SAFE Model

SAFE was a dynamic process-oriented multi-layer soil chemistry model (Warfvinge, Falkengren-Grerup, & Sverdrup, 1993), which simulated long-term reaction of the soil solution chemistry to atmospheric deposition. Cation exchange reactions and dissolution rate of 14 specific minerals using kinetic rate laws were considered. Weathering was related to soil solution chemistry. SAFE required time-series of input data regarding nutrient uptake and cycling, which could be derived

using the MAKEDEP model (Alveteg, Walse, & Warfvinge, 1998). Plant uptake was determined in relation to forest management. Several basic assumptions have been done in the model development: sulphate adsorption was not taken into account; gibbsite equilibrium was used to describe the concentrations of aluminium; all base cations were considered divalents; all N was supposed to be present as NO₃⁻, assuming complete and immediate nitrification in the soil. In this study, the SAFE model was used on a 200 year period (1900–2100). The S and N atmospheric deposition followed a site-specific scenario. Base cation deposition was considered as constant over the period since no data on historical variations exist. Soil solution parameters were simulated by the model and calibration was done using the base saturation value in 1995. SAFE was calibrated by varying the initial base saturation until an agreement between measurements and SAFE calculations was achieved for each soil layer.

2.5 Input Data

2.5.1 Atmospheric Deposition

Two datasets of atmospheric deposition were used: (1) the EMEP model deposition data for acid deposition time series and (2) the French ICP Forest RENECOFOR network data to determine present-day base cations and chloride deposition.

EMEP deposition time-series EMEP was a European network, which collected and centralised deposition data from national networks. The EMEP model was an acid deposition Lagrangian model (Iversen, 1993). In France, EMEP used eight stations for measurements of acid compounds in wet and dry deposition. The EMEP

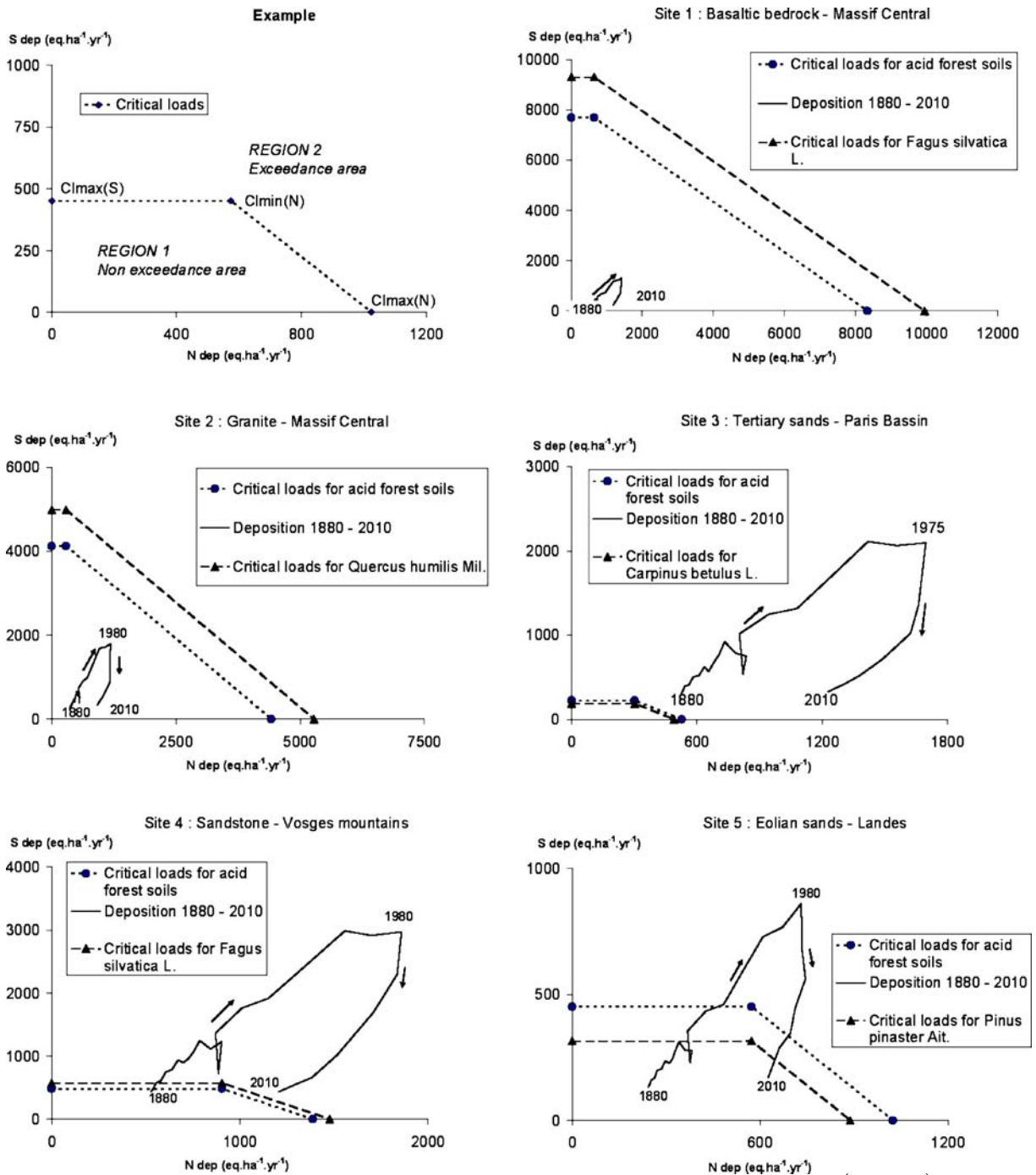


Fig. 1 Comparison between acid deposition (S, N) and two critical load functions for the acid forest soil ($\frac{[Al]}{[BC]} = 1.2$) and for the ecosystem indicator species

model integrated national emissions, stratospheric and atmospheric winds over Europe and meteorological data to determine the atmospheric deposition on a 150×150 km grid. Deposition data for S and N were

available under forest or vegetation cover. For the present study, historical deposition data for S and N over the 1880–2010 period was considered from EMEP deposition time-series (Schöpp, Posch, Mylona,

& Johansson, 2003). The deposition value over the 2010–2100 period was equal to the deposition negotiated in the Gothenburg protocol.

RENECOFOR present-day deposition data BC_{dep} (Ca, Na, K, Mg deposition), N_{dep} (NO_3 and NH_4 deposition), S_{dep} (sulfur deposition) and Cl_{dep} (Chloride deposition) were determined using field measurement data performed by the National Forest Office in charge of the RENECOFOR network for the period 1993–1996 and extrapolated to a 10×10 km grid (Croisé, Ulrich, Duplat, & Jaquet, 2005). Only open field deposition data were available at the national scale. To estimate the non-marine part of the deposition, all data have been sea-salt corrected considering Na deposition as 100% originating from sea-salts. The sea-salt ratio in sea-water was calculated from major ion concentrations in sea-water. To mitigate the lack of specialised

throughfall data at the national scale, a simple coefficient was applied to open field data to derive total deposition. This coefficient has been calculated on the RENECOFOR network sites and extrapolated to the ecosystem classification for critical load approach.

2.5.2 Soil Parameters

The physical and chemical soil data were determined from Brêthes and Ulrich (1997) and described in Table 2. The soils were pooled into three mineral layers. The layer thickness was the same for all the sites: 10 cm (layer 1), 30 cm (layer 2), 60 cm (layer 3). The CO_2 partial pressure in the three layers was set to 5, 10 and 20 times ambient partial pressure, respectively. The mineral surface area was calculated from

Table 2 Soil parameters and SAFE model input data for the five sites (1, 2, 3, 4 and 5) and the three soil layers

Site	1	2	3	4	5	Unit
Soil layers	0–10	0–10	0–10	0–10	0–10	cm
	10–40	10–40	10–40	10–40	10–40	
	40–100	40–100	40–100	40–100	40–100	
Bulk density	1,400	1,400	1,350	1,400	1,350	$kg\ m^{-3}$
	1,500	1,350	1,350	1,350	1,350	
	1,400	1,350	1,350	1,350	1,350	
Volumetric water content	0.1425	0.098	0.1	0.098	0.0675	$m^3\ m^{-3}$
	0.165	0.0675	0.0675	0.0675	0.0675	
	0.135	0.0678	0.04	0.0675	0.0675	
Mineral surf.	4.74	1.67	2.07	0.942	0.666	$10^6\ m^2\ m^{-3}$
	3.855	0.813	1.01	0.972	0.270	
	1.361	0.508	1.01	0.70	0.233	
CEC	12	8.72	1.74	2.88	3.65	$Ceq\ kg^{-1}$
	10	6.15	0.90	2.47	3	
	10	3.94	0.66	1.29	2.93	
Base sat. 1994	42	29	60	13	30	%
	68	14	17	4.9	27	
	94	11	47	6.2	18.3	
DOC	10	20	50	40	50	$mg\ l^{-1}$
	5	10	20	10	20	
	0	1.5	11	10	12	
Quartz	0	46.5	91.09	84.29	93.49	%
Olivine	20.91	0	0	0	0	
Orthose	7.80	22.7	3.87	9.28	3.15	
Plagioclases	59	17	2.80	0.80	2.72	
Apatite	2.36	0.29	0	0	0	
Biotite	0	13.51	1.92	4.23	0.53	
Muscovite	0	0	0	1	0	
Chlorite	9.93	0	0.32	0.40	0.11	

the soil texture and the bulk density of the soil according to Sverdrup and Warfvinge (1995).

2.5.3 Forest Nutrient Uptake and Management

For each ecosystem, the forest planting year has been set to 1800 with regular harvesting. Nutrient uptake values reached an equilibrium in 1980. For every ecosystem, it was assumed that 20% of the nutrient uptake was located in layer 1, 60% in layer 2 and 20% in layer 3. Values for nutrient uptakes have been shown in Table 2.

3 Results

3.1 Critical Loads and Exceedances

The comparison between acid deposition time-series (1880–2010) and critical loads of sulphur and nitrogen at the five sites, for two different critical limits, was depicted in Fig. 1. At site 1, the high critical loads were mainly due to the high weathering rate (2000 eq ha⁻¹ year⁻¹): the andosol was characterised by high content of fast dissolving minerals like olivine and apatite, and a particularly high content of plagioclase. At site 2, located on a granitic bedrock, the low weathering rate (250 eq ha⁻¹ year⁻¹) was due to slowly dissolving minerals and a relative high content of quartz. In the south of France, for both sites 1 and 2, a large part of the buffering capacity was due to high deposition of base cations, 1,011 and 1,507 eq ha⁻¹ year⁻¹, respectively (Moncoulon et al., 2004), mainly due to calcium inputs partly originating saharian dusts (Croisé et al., 2005). Acid deposition had never exceeded critical loads at sites 1 and 2 during the entire studied period. At site 3, both weathering rate (30 eq ha⁻¹ year⁻¹) and base cation deposition (210 eq ha⁻¹ year⁻¹) were low, explaining the low buffering capacity of this ecosystem. At this site, (S + N) deposition had exceeded critical loads during the entire studied period. At site 4, the podzol developed on sandstone presented a very low weathering rate due to high percent of quartz. However, probably originating the high pollution sources, base cation deposition was rather high (815 eq ha⁻¹ year⁻¹). This site was close to the German border, in the area where anthropogenic acid and basic deposition from Central Europe had been shown to be

important (Dambrine et al., 1995). At site 4, acid deposition had exceeded critical load since 1885 ($\frac{[Al]}{[BC]} = 1.2$) or 1890 ($\frac{[Al]}{[BC]} = 1.6$). At site 5, the weathering rate was very low (30 eq ha⁻¹ year⁻¹) on this acid sand podzol, whereas base cation atmospheric deposition (corrected from sea-salt influence) was intermediate (600 eq ha⁻¹ year⁻¹). On this site, the exceedance took place during 45 years ($\frac{[Al]}{[BC]} = 1.2$) or 60 years ($\frac{[Al]}{[BC]} = 0.85$).

3.2 Soil Solution Chemistry Modeling

Figure 2 depicted the pH estimated by SAFE over the 1900–2100 period at the five sites for the three soil layers. Acid deposition increased in the period 1950–1980 and decreased in the period 1980–2010. Thus, SAFE simulations were expected to show that soil solution pH decreased during the acidification period (1950–1980) and increased after 1980. After 2010, acid deposition was supposed to be constant and soil solution pH reached a new steady state.

At site 1, the effect of acid deposition during the 1950–1980 period was only significant in the first layer and pH remained constant after 1980. No significant variation occurred for $\frac{[Al]}{[BC]}$ ratio during the same period. At site 2, contrary to site 1, the pH of the second and third layers decreased during the 1950–1980 period, and the decrease continued until 2100. For these layers, the $\frac{[Al]}{[BC]}$ ratio had decreased between 1930 and 1950 and remained close to zero until nowadays. At site 3, the pH varied as expected in response to the acid deposition scenario for the whole soil profile. The $\frac{[Al]}{[BC]}$ ratio was higher than the critical limit for acid forest soils ($\frac{[Al]}{[BC]} = 1.2$) and for *Carpinus betulus* L. specifically ($\frac{[Al]}{[BC]} = 0.85$) in the three layers, all along the simulation. For the *Quercus* genus ($\frac{[Al]}{[BC]} = 1.6$), the Al/BC ratio was higher than the critical limit since 1900 (layer 1), 1940 (layer 2) and 1950 (layer 3), respectively with a time lag of 10, 50 and 60 years after critical load exceedance. At site 4, the pH varied as expected in response to the acid deposition scenario for the whole soil profile. The $\frac{[Al]}{[BC]}$ ratio reached the critical limit with a time lag of 30 years ($\frac{[Al]}{[BC]} = 1.2$) for the acid forest soil and 60 years ($\frac{[Al]}{[BC]} = 1.6$) for *Fagus sylvatica* L. in the topsoil layer. In the deeper soil, the time lag is 75 years (layer 2) and 80 years (layer 3) for both critical limits. Recovery was only simulated for *F. sylvatica* L. ($\frac{[Al]}{[BC]} = 1.6$) respectively in 2020 (layer 2)

and 2030 (layer 3). At site 5, the solution pH in the third layer kept on decreasing until today. Using the acid forest soil indicator ($\frac{[Al]}{[BC]} = 1.2$), the $\frac{[Al]}{[BC]}$ was higher than critical limit with a time lag of 10 years after exceedance of the critical load. In 2000, acid deposition reached a safe level (below critical load) in the first layer, and $\frac{[Al]}{[BC]}$ recovery occurred with a 10 years time delay.

4 Discussion and Conclusion

Among the five studied sites, only sites 1 and 2, both located in the Massif Central, have never been threatened by acid deposition. This is the result of both high critical loads – due to high weathering rates (site 1) and base cation deposition (site 1 and 2) – and remoteness from pollution sources. The three other

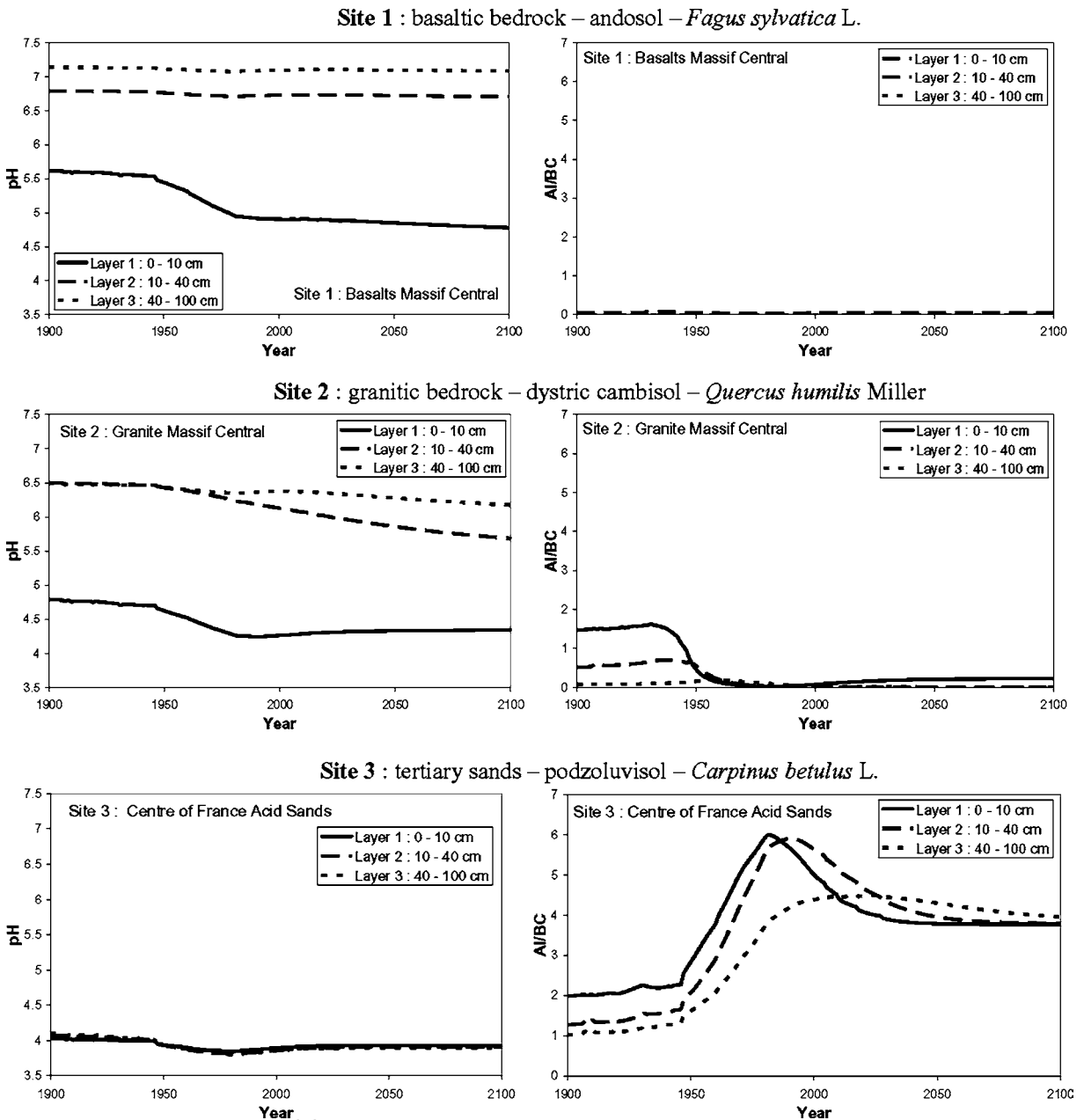
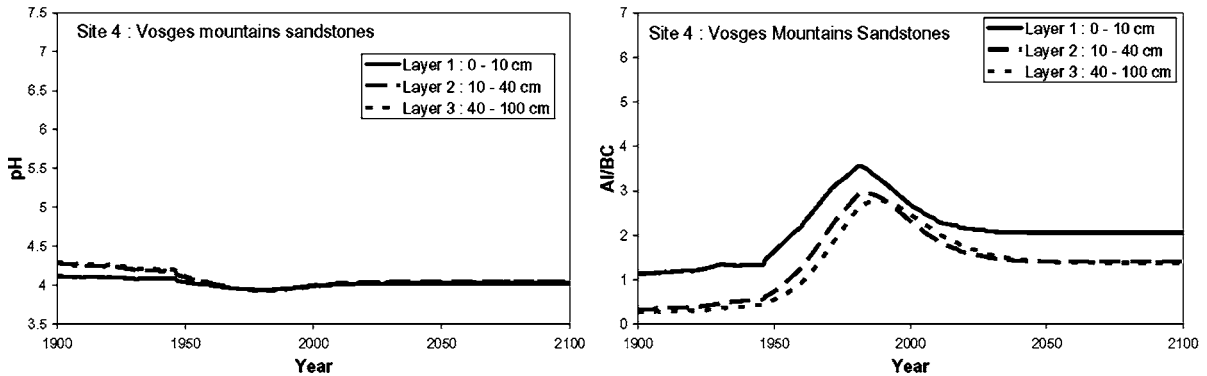


Fig. 2 Temporal trends of pH and $\frac{[Al]}{[BC]}$ ratio estimated by the SAFE model for the five studied sites

Site 4 : Sandstone – Podzol – *Fagus sylvatica* L.



Site 5 : Eolian sands – Podzol – *Pinus pinaster* Aiton

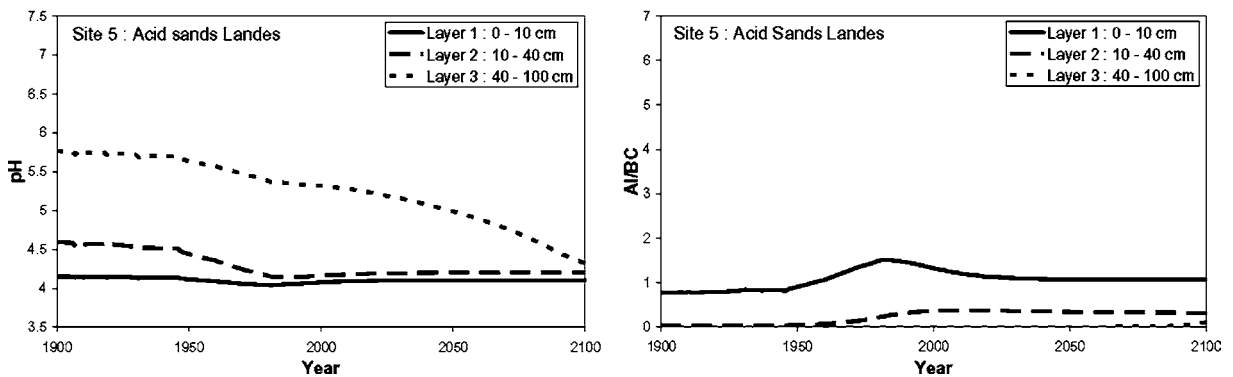


Fig. 2 (Continued)

sites were all submitted to critical load exceedances during different periods. At the Gothenburg protocol year, 2010, only site 5 will reach a non-exceedance state. Site 3 (Center of France) and site 4 (Vosges Mountains) will still be exceeded in 2010, mainly because of slow regressing nitrogen deposition.

Focusing on soil solution pH, the acidification of the top soil layer was significant at the five sites, even if these sites were not concerned by critical load exceedances. The acidification of the deeper layers was specifically detected at the most sensitive sites (3, 4) during the 1950–1980 period. Despite the acid deposition decrease, the high weathering rate at site 1 efficiently buffered the impact of acid inputs in the deeper soil. The acidification process in the deep layers was still running with a time lag at sites 2 and 5. This time delay in the deeper horizons indicated that, in sites 2 and 5, acidification has moved down the soil profile as a consequence of slow base cation depletion from CEC. Indeed, for site 2, base saturation decreased from 20 to 5% (second layer) between 1950 and 2050 and 11 to 6%

(third layer) between 1980 and 2075. For site 5, in the third layer, base saturation decreased from 25 to 1% between 1965 and 2100. This time lag in the deep horizons of sites 2 and 5, which were characterised by low weathering rates, was probably linked to the ratio of low acid inputs over a significant base cation atmospheric inputs. This guaranteed an efficient buffering capacity to the acidification of the soil profile. These behaviours in the deeper soil layers are not so surprising. Indeed, springwater analysis over the 1963–1996 period in the French Vosges mountains sandstones have shown a continuous acidification (Probst et al., 1999) whereas the sulphur deposition showed a 70% decrease by 1980.

The $\frac{[Al]}{[BC]}$ criterion was less sensitive than pH. During the acidification process, base cations were efficiently released from the humic and clay complex by protons exchange and $\frac{[Al]}{[BC]}$ ratio did not change significantly or even decreased in soils with high CEC (sites 1 and 2). In the andosol (site 1) and the dystric cambisol (site 2), the important base cation release from the exchangeable complex – reloaded mainly by weathering (site 1)

or atmospheric deposition (sites 1 and 2) – was the main process, which neutralised acid ions. On the opposite, because of the combination of poor base cation pool in the soil and high acid deposition, the acidification even concerned the deep soil of sites 3 and 4. The site 5, which was also very sensitive to acidification, received less acid deposition than sites 3 and 4, and thus $\frac{[Al]}{[BC]}$ ratio only exceeded the critical limit in the first layer. For site 5, recovery occurred in the first soil layer, 10 years after the end of critical load exceedance, thanks to significant base cation atmospheric deposition. This study presented a first estimation of the time delay between critical load and critical limit exceedance, using $\frac{[Al]}{[BC]}$ ratio: 10 to 30 years in the first soil layer and 50 to 90 years in the deepest soil. Comparison was made between the different values of critical $\frac{[Al]}{[BC]}$ for the different indicator species. The global acid forest soil indicator ($\frac{[Al]}{[BC]} = 1.2$) was an average value used for critical load calculations at the national scale. Regarding more precisely the different species, it appeared that the most acidification sensitive species, *Carpinus betulus*, was threatened, on site 3, 40 to 50 years before *Quercus* in the deepest soil layers. On site 4, *P. abies* was threatened 35 years before *F. sylvatica* in the first soil layer and 5 to 10 years in the deepest soil.

In the present study, constant base cation deposition had been used, due to the lack of available time-series over the period. This was realistic on sites where natural base cation deposition was important (sites 1, 2 and 5) or on site 3 with poor base cation deposition. However, on site 4 (Vosges mountains), an important part of base cation deposition originated industrial emissions and was thus probably not constant over the period (Probst, Fritz, & Viville, 1995). As a consequence, such time-series would be used in the future to improve modelling results.

The sulphur and nitrogen deposition over the period 2010–2100 considered in the present study, corresponded to the Gothenburg protocol values. As we could observe on Fig. 1, the Gothenburg protocol deposition in 2100 will still exceed critical loads for sites 3 and 4. This was confirmed by the simulation of the soil solution response: no recovery for $\frac{[Al]}{[BC]}$ will occur without further deposition reduction. Moreover, acidification process was still detectable in sites 2 and 5. Even though significant sulphur deposition reduction after 1980 had reached its critical load in 4 sites on 5, further reduction of nitrogen will be needed to

reach a safety situation in sensitive ecosystems. Moreover, this study highlighted the importance of taking into account not only weathering rates and vegetation uptake but also the relative ratio between base cation deposition and acid compounds to predict ecosystem response to atmospheric pollution scenario.

Acknowledgements The authors are grateful to Jean-Paul Hettelingh and an anonymous reviewer for their fruitful comments, and thank the ADEME, particularly Laurence Galsomiers, for supporting the French NFC investigations. The authors also thank Erwin Ulrich for providing RENECOFOR data and Jean-Paul Party for his appreciated expertise.

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