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# A spatial study of the relationships between streamwater acidity and geology, soils and relief (Vosges, northeastern France)

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## Abstract

We have used a geographic information system (GIS) to study the relationships between streamwater pH and alkalinity at base flow with geology, soils and relief in 100 forested catchments located in the sandstone portion of the Vosges mountains, where atmospheric deposition and forest cover can be considered homogeneous. At base flow, streamwater acidity depends primarily on bedrock and soil content of weatherable minerals whose dissolution neutralizes acidity. Catchments are developed on three main stratigraphic levels, consisting of two sandstone layers rich in weatherable minerals, called "rich", at the upper and lower extremity of the stratigraphic sequence, and a quartzitic sandstone, called "poor", in between. Catchments were classified into three major groups in relation to the stratigraphic sequence: (1) only "poor" bedrock, (2) "rich" upstream and "poor" downstream, (3) "poor" upstream and "rich" downstream. Results showed that streamwater pH differed depending on the group. Within each group, the pH was related to bedrock, soil and relief characteristics of the catchments. The relative surface covered by the bedrock located in the lower part of the catchment explained more than 50% of the variability of pH. More than 20% of variability could be explained by soil types in catchments composed of "poor" bedrock only. Soil type apparently did not influence streamwater pH in the other groups. The catchment area, probably related to the increase of soil volume and water residence time, explained up to 20% of pH variability, depending on the catchment type. © 1999 Elsevier Science B.V. All rights reserved.

*Keywords:* Streamwater acidity; Low flow conditions; GIS; Catchment geology

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## 1. Introduction

Since the 1970s, acidification of soil and surface waters has been described in European and North American forest massifs. Remarkable decreases in pH/alkalinity were demonstrated in sensitive areas (Johnson et al., 1991; Battarbee et al., 1990). Catchment sensitivity can be predicted from the relationship between precipitation acidity and streamwater

alkalinity (Almer et al., 1978; Dickson, 1986; Henriksen, 1979). Sensitivity depends primarily on the soil and rock content of weatherable minerals whose dissolution neutralizes acidity and produces alkalinity. Hydrological factors, often linked to the presence of physical obstacles restricting the time and/or the area of contact between minerals and solutions in the soil, can also play an important role (Eilers et al., 1983; Chen et al., 1984), while the retention of sulfur and nitrogen delays the effects of acid deposition.

General relationships between bedrock characteristics and stream acidity have been used to map

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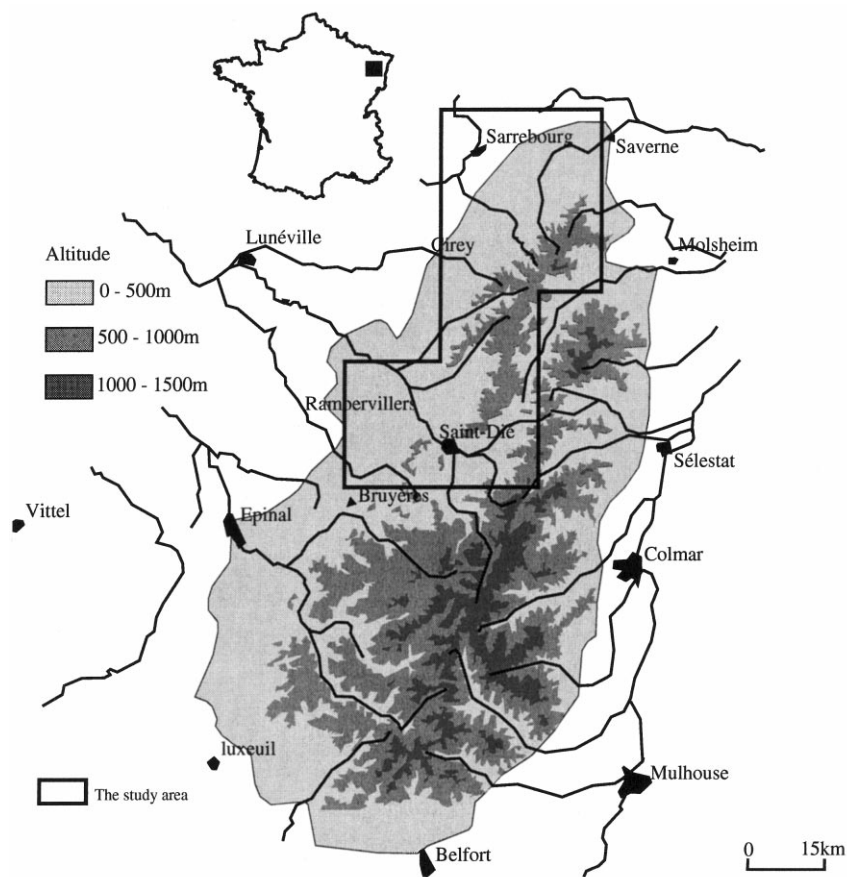


Fig. 1. The Vosges massif and the study area.

catchment sensitivity to acidification (Bricker and Rice, 1989). Based on either expert knowledge (Hornung et al., 1995; Hall et al., 1995) or on statistical analysis (Rochelle et al., 1989; Phillips 1987), several studies have incorporated glacial morphology and relief characteristics in order to improve catchment sensitivity mapping. Integrated studies of environment factors influencing catchment sensitivity, however, remain very rare (Rapp et al., 1987). In the Vosges massif (northeastern France), an inventory of surface water acidity (Probst et al., 1995; Party et al., 1995) has shown that low alkalinity waters were located in two geological areas: acid granites and sandstones. Within these areas, however, considerable diversity of pH values was recorded. The aim of the present study was to describe the variability of water pH on sandstone by using lithologic, pedologic and

topographic parameters. For this purpose, the acidity of 100 streams originating from the sandstone area was measured at low water flow. Information on bedrock, soils and relief were digitized and integrated using a GIS. The crossing of the spatialized information enabled to identify environmental factors most highly related to water acidity.

## 2. The study area

One hundred entirely forested catchments covering a total area of 27 207 hectares (ha) were identified in the western part of the Vosges massif (Fig. 1). The relief is moderate with altitudes between 300 and 900 meters. The climate is oceanic with a continental influence, and cool (average temperature: 9°C at the

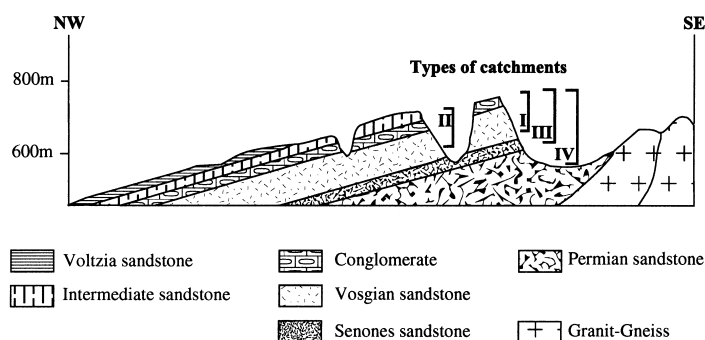


Fig. 2. Geological section through the study area and localization of each type of catchment.

elevation of 400 meters) with an average yearly precipitation of 1000 mm varying with altitude between 800 and 1200 mm. Fir (*Abies alba*) forests predominate throughout the region. Annual acid deposition, calculated from three years (1989–1992) bulk precipitation and throughfall monitoring at three fir forest sites enclosing the studied area varied between  $0.4 \text{ keqH}^+ \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$  at 300 m altitude and  $1 \text{ keqH}^+ \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$  at 900 m altitude. The range of sulfate and nitrate deposition were  $10\text{--}14 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$  and  $8\text{--}14 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$  respectively (Dambrine et al., 1995).

The bedrock is composed of different sandstones and conglomerates dating from the Permian and Trias inferior (Fig. 2). From the lower and older, to the upper and younger level, the stratigraphic sequence is composed of Permian sandstone, Senones sandstone, Vosgian sandstone, Conglomerate, Intermediate sandstone and Voltzia sandstone. Depending on their composition (Perriaux, 1961), it is possible to classify them from the richer to the poorer in weatherable minerals:

- Permian and Voltzia sandstones, at both ends of the stratigraphic sequence, whose percentages of  $\text{SiO}_2$  and  $\text{K}_2\text{O}$  are 74–79% and 7–5%. Although the percentages of Ca and Mg do not differ from the other groups, these stratigraphic levels enclose thin layers of calcite. The Senones sandstone makes the link with the following group, since its mineral composition is similar (81%  $\text{SiO}_2$ , 5%  $\text{K}_2\text{O}$ ) but it is devoid of calcite.
- Intermediate sandstone whose  $\text{SiO}_2$  and  $\text{K}_2\text{O}$  percentages are 88% and 3%. In comparison to

the above, this level is also devoid of calcite layers but poorer in feldspar and mica.

- Vosgian sandstone and conglomerate whose  $\text{SiO}_2$  and  $\text{K}_2\text{O}$  percentages are 93% and 2%, mainly composed of sand and siliceous pebbles.

In the Vosges mountains, soil evolution depends mainly on the composition of bedrock (Bonneau et al., 1978). Soils are sandy, very acid and widely podzolized on Vosgian sandstone and conglomerate (poorest layers), except locally when wind-borne deposits (loess) or colluviums originating from richer sandstone (intermediate or Voltzia layers) occur. On Voltzia, Intermediate, Senones and Permian sandstones, the weathering of feldspars and micas has led to the formation of cambisols of sandy to loamy-sand texture.

### 3. Method

Samples of water were taken at the outlet of all catchments drained by clearwater streams, at low water flow, during three consecutive weeks with almost no rain in the autumn of 1992 (Party et al., 1993). The buffering capacity of soil and rock, in relation to weathering processes, is maximal during that period. The pH of all samples (100) was measured, alkalinity was measured by Gran titration on 43 of them and complete chemical analysis was conducted on 18 samples. From these measurements, the following statistical relationship was deduced

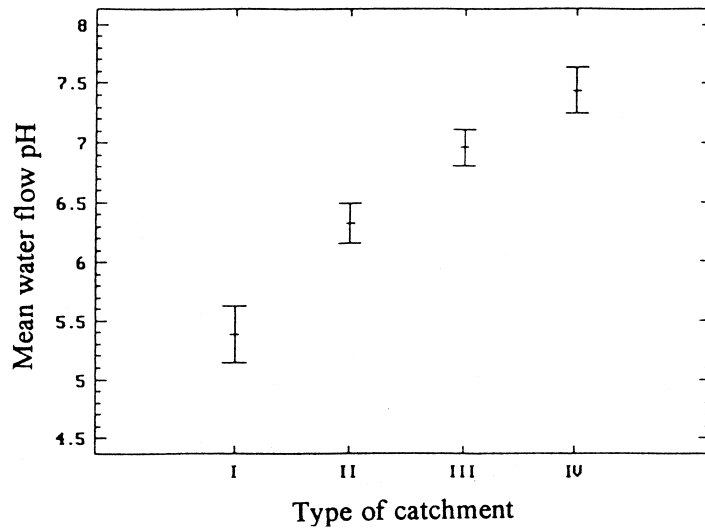


Fig. 3. Multiple comparison of mean pH for each type of catchment (Tukey test).

(Fillion, 1998).

$$\text{Alk}(\text{mmol.l}^{-1}) = [\text{HCO}_3^-] + 2[\text{CO}_3^{2-}] + [\text{OH}^-] - [\text{H}^+]$$

$$\text{Alk}(\text{mmol.l}^{-1}) = 1.3(10^{-7.82+pH} + 2 \times 10^{-18.15+2pH}) + 1000(10^{-14+pH} - 10^{-pH})$$

This relation, indicating a  $\text{CO}_2$  partial pressure higher ( $10^{-2.9}$ ) than atmospheric pressure ( $10^{-3.5}$ ), was applied to calculate alkalinity where measurements were lacking.

Data on bedrocks was digitized over the entire study area from geologic maps at the scale of 1:50,000. The soil map of Saint-Dié at the scale of 1:100 000 (Bonneau et al., 1978) covering 40% of the territory studied was also digitized. Topographical information was provided by a Digital Elevation Model (DEM) at 50 m, extracted from the altimetric database of the National Geographic Institute (IGN). Morphometric parameters of each catchment were derived from that DEM: minimal, maximal and medium altitudes, total area, length of the stream, average catchment slope and slope for each bedrock. A hydrologic algorithm applied to the DEM enabled

the catchments to be automatically delineated from the outlet points (Jenson and Domingue, 1988). Soils were classified into two groups: podzolic soils and cambisols. The relative surface covered by each soil type and bedrock type was calculated in order to describe the soil and bedrock composition of each catchment.

Catchment typology was established on the basis of bedrock composition and stratigraphy. Catchments were grouped in four main types (Fig. 2):

- type I: “poor”. Catchments entirely underlain by Vosgian sandstone and conglomerate.
- type II: “rich over poor”. Catchments whose upper part is underlain by Intermediate sandstone with or without Voltzia sandstone, and whose lower part is based on conglomerate and Vosgian sandstone.
- type III and IV: “poor over rich”. Catchments whose upper part is composed of conglomerate and Vosgian sandstone (with a possible occurrence of intermediate sandstone), and whose lower part is composed of Senones sandstone (type III) or Senones + Permian sandstone (type IV).

The information was compiled in a statistical table in which each line is a catchment and each column a lithologic, pedologic or topographic characteristic of that catchment. A matrix correlation was used to test

Table 1

Correlation matrix between pH and alkalinity and environmental parameters for catchments of type I. In bold type significant linear correlation coefficients at the 5% level; critical value equal to 0.53 for 12 degrees of freedom. Alk = alkalinity; TA = total catchment area; CS = relative surface of cambisols; AS = catchment average slope; AA = catchment average altitude

	pH	TA	CS	AS	AA	Alk	TA	CS	AS	AA	
pH	1.00					Alk	1.00				
TA	<b>0.73</b>	1.00				TA	<b>0.69</b>	1.00			
CS	<b>0.80</b>	-0.38	1.00			CS	<b>0.78</b>	-0.32	1.00		
AS	0.42	<b>-0.72</b>	-0.16	1.00		AS	0.39	<b>-0.71</b>	-0.12	1.00	
AA	-0.32	<b>0.57</b>	-0.08	<b>0.58</b>	1.00	AA	-0.36	<b>0.61</b>	-0.05	<b>0.59</b>	1.00

the influence of each explanatory variable on stream-water acidity. A regression of the most highly correlated variables was then conducted against pH and alkalinity.

#### 4. Results

A multiple comparison of mean pH for each type of catchment (analysis of variance) indicated that the type of bedrock and stratigraphic sequence within the catchment determined great differences in stream-water pH. Fig. 3 shows a plot of the mean for each catchment type and the 95% confidence intervals for the means. Bars that did not overlap in types I, II, III and IV catchments indicate significant differences

between the four mean pH values. The most acidic waters (mean pH = 5.4) corresponded to the group of catchments whose bedrock was entirely poor (type I). For the other types composed of both rich and poor bedrock, the pH appeared to be related to the nature of the substratum located downstream. Acidity was lowest (mean pH = 6.3) when the catchment outlet was located on the poorest bedrock (type II). Mean streamwater pH values were 7 and 7.4 when the lower part of the catchment was composed of rich sandstones (type III and IV respectively).

Considering each group of catchments separately, the distribution of pH and alkalinity was related to the main environmental factors revealed by the correlation matrix.

For catchments entirely located on poor bedrock

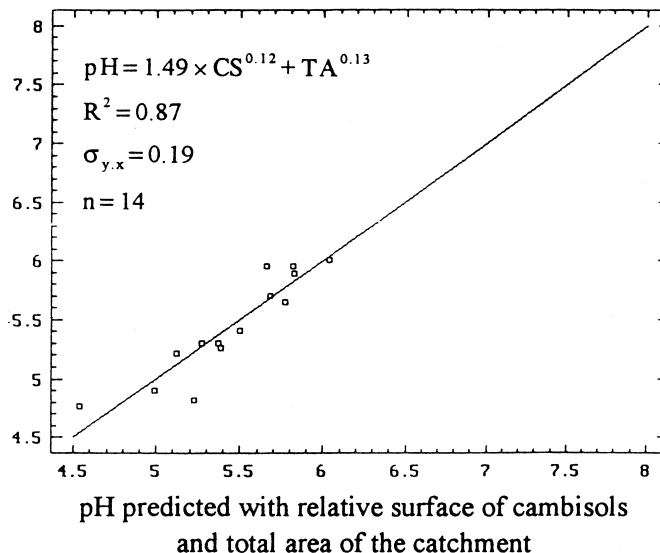


Fig. 4. Relationship between measured and predicted pH for type I-catchments.

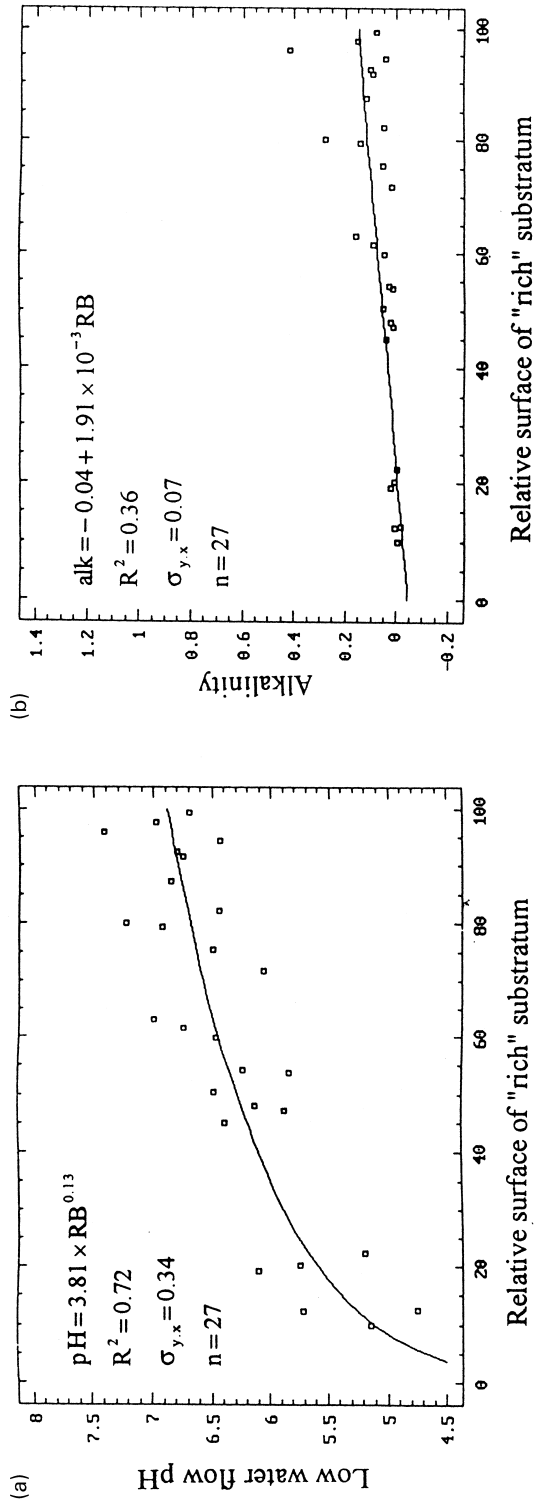


Fig. 5. Relationship between pH (a) and alkalinity (b) and relative surface of "rich" bedrock for type II-catchments ("rich over poor").

Table 2

Correlation matrix between pH and alkalinity and environmental parameters for catchments of type II. In bold type significant linear correlation coefficients at the 5% level; critical value equal to 0.38 for 25 degrees of freedom. Alk = alkalinity; TA = total catchment area; RB = relative surface of rich bedrock (intermediate sandstone); AS = catchment average slope; AA = catchment average altitude

	pH	TA	RB	AS	AA	Alk	TA	RB	AS	AA	
pH	1.00					Alk	1.00				
TA	<b>0.43</b>	1.00				TA	-0.05	1.00			
RB	<b>0.83</b>	<b>-0.63</b>	1.00			RB	<b>0.55</b>	<b>-0.43</b>	1.00		
AS	-0.06	<b>-0.63</b>	-0.19	1.00		AS	-0.24	<b>-0.72</b>	-0.22	1.00	
AA	0.13	<b>0.45</b>	-0.02	<b>0.69</b>	1.00	AA	0.34	<b>0.55</b>	-0.06	<b>0.71</b>	1.00

(type I), the relative surface of cambisols (CS) was the variable most correlated with streamwater pH, explaining 67% of the variance. The total area (ha) of the catchment (TA) explained 20% of the variance (Table 1). The best fit was obtained with a multiplicative model (Fig. 4), the equation being:

$$pH = 1.49 \times CS^{0.12} + TA^{0.13} \quad R^2 = 0.87$$

pH and alkalinity were linearly correlated in this range of pH values.

In type II catchments (“poor over rich”), streamwater pH varied primarily as a function of the relative surface of rich substratum (RB) with 72% of variance being explained (Fig. 5a). The total catchment area explained 5% of the variance (Table 2). The equation produced by a multiple regression procedure was:

$$pH = 1.59 \times RB^{0.23} + TA^{0.08} \quad R^2 = 0.77$$

In type III catchments (“poor over rich”), streamwater pH was related primarily to the percentage of rich sandstone (Senones) located at the outlet of the catchment (61% of the variance explained). Total catchment area was no longer involved (Table 3).

Table 3

Correlation matrix between pH and alkalinity and environmental parameters for catchments of type III. In bold type significant linear correlation coefficients at the 5% level; critical value equal to 0.32 for 35 degrees of freedom. Alk = alkalinity; TA = total catchment area; RB = relative surface of rich bedrock (Sénones sandstone); AS = catchment average slope; AA = catchment average altitude

	pH	TA	RB	AS	AA	Alk	TA	RB	AS	AA	
pH	1.00					Alk	1.00				
TA	0.11	1.00				TA	0.14	1.00			
RB	<b>0.74</b>	-0.12	1.00			RB	<b>0.75</b>	-0.14	1.00		
AS	-0.24	<b>-0.62</b>	0.08	1.00		AS	-0.01	<b>-0.67</b>	-0.08	1.00	
AA	-0.12	<b>0.50</b>	-0.19	<b>0.49</b>	1.00	AA	0.09	<b>0.50</b>	-0.17	<b>0.48</b>	1.00

The equation was (Fig. 6):

$$pH = 6.12 \times RB^{0.04} \quad R^2 = 0.61$$

The same was observed for type IV catchments, with 67% of the variance explained by the relative surface of rich sandstones (Senones + Permian sandstones) (Table 4). The equation was (Fig. 7):

$$pH = 6.64 \times RB^{0.03} \quad R^2 = 0.67$$

In all cases, the standard deviation of estimation ( $\sigma_{y,x}$ ) remained below 0.35 for pH.

## 5. Discussion

Waters drained at low flow in autumn have undergone a long contact time with the soil matrix of the deepest horizons of the soil, especially with the weathered zone located in the lower part of the catchment. On Vosgian sandstone, which is composed almost exclusively of pure quartz sand, variations of weathering potential were expressed by the relative proportion of cambisols. Cambisols occur when sandstone is slightly enriched locally in weatherable fine minerals, or on thick colluviums at the lower part of



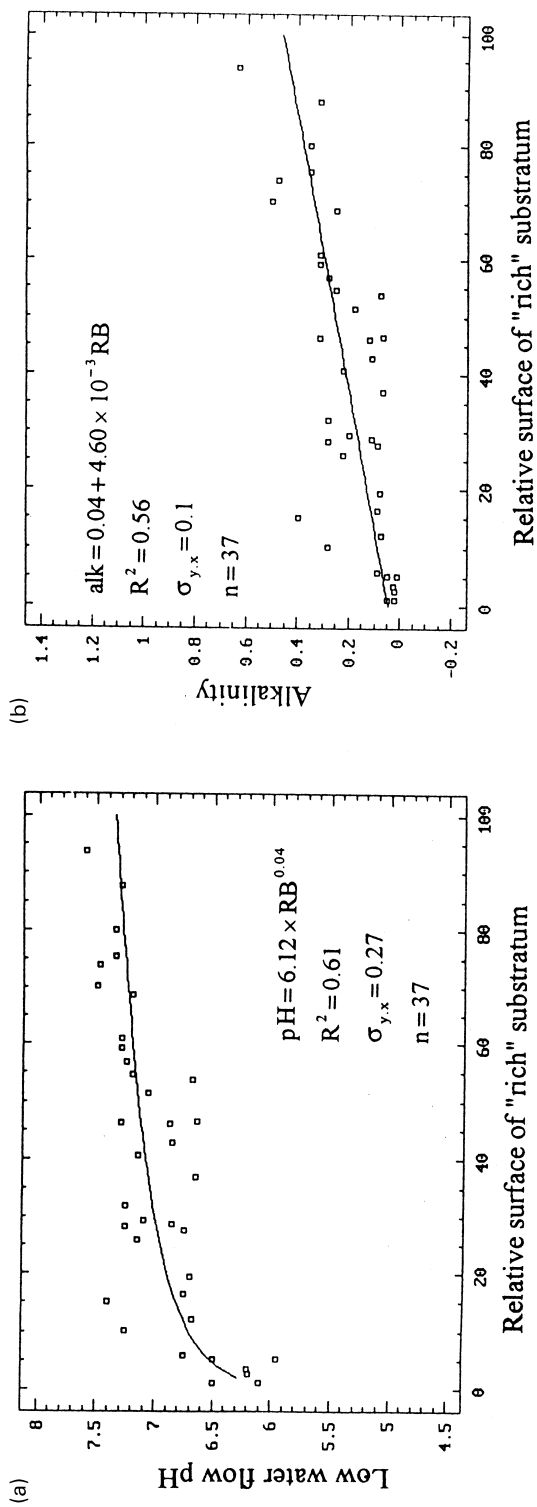


Fig. 6. Relationship between pH (a) and alkalinity (b) and relative surface of "rich" bedrock for type III-catchments ("poor over rich").

Table 4

Correlation matrix between pH and alkalinity and environmental parameters for catchments of type IV. In bold type significant linear correlation coefficients at the 5% level; critical value equal to 0.42 for 20 degrees of freedom. Alk = alkalinity; TA = total catchment area; RB = relative surface of rich bedrock (Permian and Sénonès sandstones); AS = catchment average slope; AA = catchment average altitude

	pH	TA	RB	AS	AA	Alk	TA	RB	AS	AA	
pH	1.00					Alk	1.00				
TA	-0.11	1.00				TA	-0.35	1.00			
RB	<b>0.64</b>	0.22	1.00			RB	<b>0.60</b>	0.35	1.00		
AS	0.26	<b>-0.66</b>	0.13	1.00		AS	0.07	<b>-0.64</b>	.27	1.00	
AA	0.13	<b>0.61</b>	-0.14	<b>0.68</b>	1.00	AA	0.34	<b>0.65</b>	-0.26	<b>0.67</b>	1.00

the slopes. Neutralization also occurred to greater extents in large catchments where the general slope becomes lower and soil thickness increases (Rochelle et al., 1992). The influence of the catchment area cannot be easily explained by other ways: (1) water coming from small catchments could have retained a CO<sub>2</sub> partial pressure higher and closer to that of soils, but then catchment size should not change alkalinity; (2) dissolved organic carbon (DOC) in water draining small catchments could be higher. Such a relationship does not appear in the set of complete water analyses available, since DOC was consistently below 4 mg.l<sup>-1</sup>. Moreover, this should have induced a distortion in the pH/alkalinity relationship at low pH, which was not observed.

When the upper part of the catchment was composed of rocks and soils richer in weatherable minerals, the proportion of poor bedrock located downstream or rich bedrock located upstream explained most of the variation in streamwater acidity. In this case, soil type did not appear to influence pH, probably because soil types provide redundant information highly correlated with bedrock types. Because at low flow stream water mainly originates from the bottom of the catchment, a substrate rich in weatherable minerals at the lower part of the catchment led to a greater increase in pH (and alkalinity) than that produced by an equivalent proportion of the same material located in the upper part of the catchment.

The observed linear relationships between alkalinity and the proportion of richer bedrock in the catchment probably reflect the progressive neutralization of acidity by weathering. This should imply a relation between pH and the logarithm of the relative surface of rich bedrock. The better fit observed with power

relationships might be partly due to the fact that these statistical relationships are strongly dependent on the data measured at high pH, which are often scattered.

Monitoring of atmospheric deposition in the Vosges mountains have shown that deposition increased with altitude, but also from the South to the North of the study area. One may suspect the direct effect of the deposition level on stream pH. This effect is possible but difficult to demonstrate. Within the whole data set, as well as within each catchment type, stream pH was not related to the mean altitude of the catchment. Moreover, on the basis of 150 streams analysed in the same area, Boudot (in Dambrine et al., 1998) has shown that strong acid concentrations were not different in all catchment types.

## 6. Conclusion

Results of this study show that GIS is a powerful tool for describing and analyzing the spatial variability of streamwater pH when geology, soil and relief maps are available. More than 60% of the variability of streamwater pH and alkalinity was in fact explained by the nature of the bedrock in the lower part of the catchment, soil types and total catchment area. Although the results obtained may appear specific for the sandstone part of the Vosges, the importance of catchment location in the stratigraphic sequence for understanding the variability of streamwater pH should be stressed. In general, sensitivity mapping studies consider the dominant bedrock of each geographical unit (Hettelingh et al., 1991). This study shows that the spatial distribution of bedrocks is very important to consider. This idea should not be

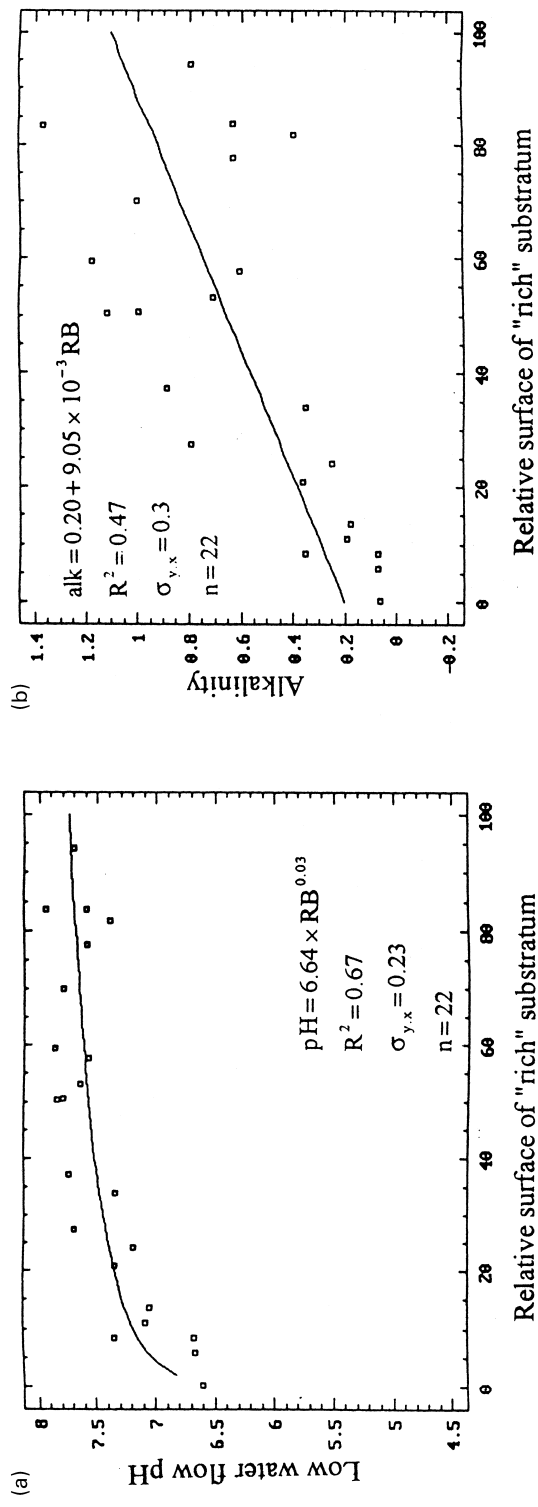


Fig. 7. Relationship between pH (a) and alkalinity (b) and relative surface of "rich" bedrock for type IV catchments ("poor over rich").

restricted to the sedimentary world. The sedimentary environment, poorly influenced by glaciers, has evidently facilitated our analysis but conclusions might also be applied to igneous rocks locally influenced by hydrothermalism (Mansuy, 1992) or glaciers.

A similar study at high flow may change the relative importance of the factors involved to a limited extent because sandstones are porous and therefore water residence time should be rather long even at high flow. The combination of such studies at high and low flows (Philips and Stewart, 1991), especially in glaciated areas, however, may provide an assessment of water pathways in relation to soil structures inherited from the glacial periods.

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