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The effects of acidity and aluminium leached from acid-sulphate soils on riverine fish assemblages

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Drainage of acid-sulphate soils mainly for agriculture, increases oxidation and leads to extensive leaching of acidity and aluminium (Al) to rivers in western Finland. The lowest average pH (4.55) values and the highest average Al concentrations (7.4 mg l^{-1}) were measured in the Vöyrinjoki, which appeared to be void of fish. Frequency of sites with no fish was clearly the highest (59%) in rivers with average pH < 5. Total fish density and biomass were the highest at the sites with pH > 6. The two fish species that are most tolerant to acidity, pike (*Esox lucius*) and perch (*Perca fluviatilis*), were found in rivers with an average pH as low as 4.6. Acid-sensitive species, especially grayling (*Thymallus thymallus*), brown trout (*Salmo trutta*) and minnow (*Phoxinus phoxinus*), were usually not found in rivers with average water pH < 6. To mitigate acidification of river waters, controlled farmland drainage techniques should be applied.

Introduction

Sulphide-containing soils — usually called acid sulphate (a.s.) soils — cover around 170 000–240 000 km^2 worldwide, with major occurrences in Africa, Australia, Asia and Latin America (Ritsema *et al*. 2000, Andriesse and Mensvoort 2002). In Europe, the largest a.s. soil areas of about 1600–3000 km^2 are found in western Finland (Yli-Halla *et al*. 1999, Fältmarsch *et al*. 2008). The sediments mainly found in the Finnish coastal areas were formed in the Baltic Sea 4000–8000 years ago under anoxic conditions, and later emerged from the sea due to the isostatic land uplift (Palko 1994, Toivonen and Österholm 2011). Today these soils can be found in areas located up to 100 m above the current sea level. Sulfide-bearing sediments are still forming in river estuaries (Toivonen and Österholm 2011).

Oxidation and weathering of these sulphide-containing sediments have taken place as a result of ditching and farmland drainage (Fältmarsch *et al*. 2008). As a result of dissolution and oxidation of metalsulfides, sulfuric acid is produced and metals are released (Åström and Björklund 1996). Acidic pore water rich in metals, e.g. aluminium (Al), is flushed to recipient streams especially during heavy rain periods (Toivonen and Österholm 2011, Nystrand and Österholm 2013). Percentage of a.s. soils in the watershed has been found to correlate with river water acidity in western Finland (Palko and Yli-Halla 1993). Intensified utilization of modern subsur-

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face techniques in farmland drainage has significantly increased acidification (Österholm and Åström 2002, Joukainen and Yli-Halla 2003). In the areas of a.s. soils, river water acidity varies temporally with discharge (Saarinen and Kløve 2012). Accordingly, the lowest pH values are usually recorded during spring floods, and sporadic autumn or winter floods. Considering future climate change scenarios, the river water acidity in this area may increase considerably, especially during winters (Saarinen and Kløve 2012).

In acidic waters, Al is acutely toxic to fish (Exley *et al*. 1991, Poléo 1995). Al is one of the most enriched elements in drainage waters from a.s. soils (Fältmarch *et al*. 2008). In the Finnish a.s. soils, Al occurs as aluminosilicate and probably Al-hydroxide. Acidic conditions (pH 2.5–4.5) promote aluminosilicate weathering and Al-hydroxide dissolution and thus Al solubilisation and transport (Fältmarch *et al*. 2008). The Al concentration in drainage water from a.s. soils correlates strongly with pH, and concentrations up to 260 mg l^{-1} were measured in small ditches in the highly acidified areas of a.s. soils in western Finland (Åström and Björklund 1995, Fältmarch *et al*. 2008). Exposure studies in the laboratory show that even much lower Al concentrations are deadly to fish (e.g. Vuorinen *et al*. 1993, Keinänen *et al*. 2000, Stephens and Ingram 2006).

Low pH and associated high Al contents typically induce damage to gill tissue (Exley *et al*. 1991). Typical symptoms in fish are ionoregulatory disturbance, respiratory dysfunction, osmoregulatory breakdown resulting in a net flux of water into fish, excessive production of mucous and accumulation of Al at the gill epithelium (Youson and Neville 1987, Muniz and Leivestad 1980, Exley *et al*. 1991). Although acidity itself is a serious fish stress factor (Fromm 1980), the combined effect of low pH and high Al level is usually regarded as the main reason behind the physiological disruption, and ultimately fish mortality due to acidification (Muniz and Leivestad 1980, Wood 1989).

The objectives of this study were to (1) evaluate the effect of river water acidity and Al leached from a.s. soils on riverine fish assemblages including species richness (number of fish species), existence of sites void of fish, total fish density and biomass; and (2) compare and rank existing fish species according to their tolerance to acidity.

Material and methods

Electrofishing was performed in August–September 2010–2012 at 200 sampling sites along 35 rivers (latitude at river mouth 62°13.2´–64°51.2´, distance between the farthest river mouths from each other about 350 km, drainage basins 104–4922 km^2) emptying into the Gulf of Bothnia (Baltic Sea, Finland). The sampling sites with the average area of 141 m^2 (range 30–1035 m^2) were fished without using escape nets. Fish were captured with Hans Grassl GmbH 1G 200-2 electrofishing gear using pulsed (50 Hz) DC current with usually 400–600 V voltage adjusted to water conductivity. Each sampling site was fished once by two waders, one using the anode and an assistant collecting the stunned fish with a hand net. All captured fish were counted and identified to species. Total length (TL) of narcotized fishes was measured to the nearest 1 mm and pooled individuals of each species were weighed to the nearest 0.1 g, after which the fish were released.

Results on pH and total Al in the river water in 2005–2010 were retrieved from the Hertta database hosted by the Finnish Environment Institute. Averages of pH for a river or a river section nearest to the electrofishing site were calculated. The number of pH measurements per site varied from 6 to 80, whereas the number of Al concentration values varied from 1 to 79. Al analyses were available for 29 of the 35 rivers. Average pH of the sampled rivers ranged from 4.55 to 6.75 with the overall average equalling 6.05. Average Al concentrations in the sampled rivers ranged from 0.27 to 7.4 mg l^{-1} with the overall average equalling 1.4 mg l^{-1}. Supplementary results of sulphate (SO_4) analyses were also gathered.

A χ^2-test was used to evaluate the differences in frequency of occurrence of sites that are void of fish in the rivers classified to three pH ranges (pH < 5, pH 5–6, pH > 6). Total fish density and biomass in the river classes were compared with

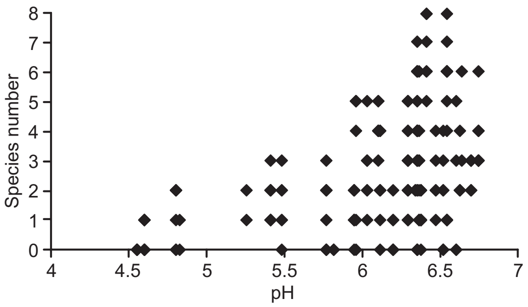

Fig. 1. Fish species numbers depending on river-water pH.

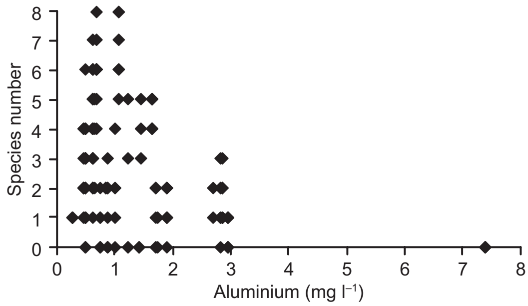

Fig. 2. Fish species numbers depending on river-water total aluminium (Al) concentration.

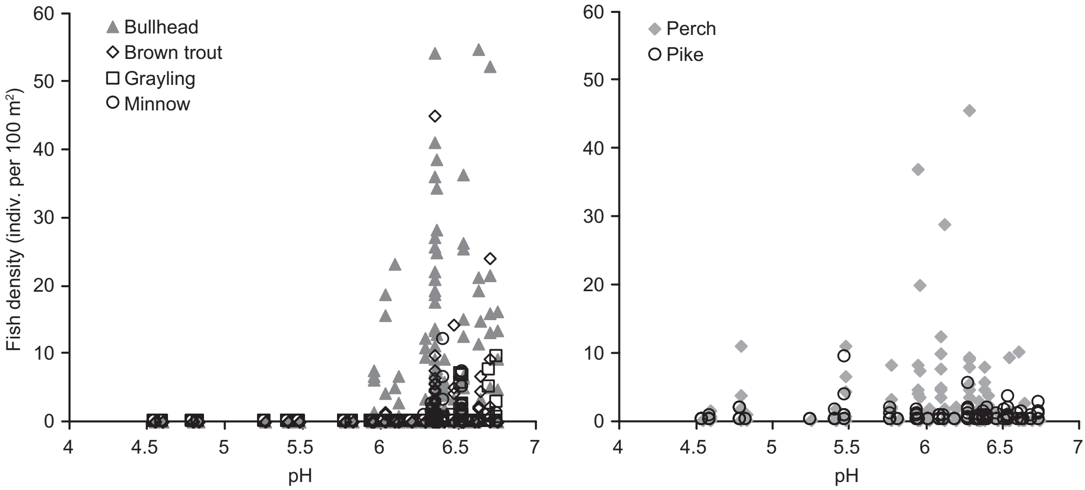

Fig. 3. Density of four fish species considered as acidity sensitive, and two fish species considered as tolerant to river water pH.

the Kruskal-Wallis (KW) test because of the non-normal distributions of these two variables. Statistical tests were performed with SAS® ver. 9.4.

Results

The maximum number of fish species caught by electrofishing decreased gradually from 8 in river water with pH around 6.5 to 2 in river water with pH below 5 (*see* Fig. 1). Respectively, the number of fish species decreased as the average Al concentration increased (*see* Fig. 2). A negative correlation existed between river water pH and Al (Pearson's $r = -0.78$). The lowest average pH (4.55) and the highest average Al concentration (7.4 mg l^{-1}) were measured in the Vöyrinjoki which appeared to be void of fish based on five electrofishing events with no catch in 2010 and 2012 (Figs. 3 and 4). The lowest pH (4.0, $n = 74$) and the highest total Al concentration (19 mg l^{-1}, $n = 67$) were recorded in the Vöyrinjoki. High concentrations of sulphate in this river (mean SO_4 = 161 mg l^{-1}, range 88–260 mg l^{-1}, $n = 69$) confirmed our hypothesis of the sulphate soils being the source of acidity.

Frequency of occurrence of sites void of fish (no fish caught by electrofishing) was clearly the highest in rivers with average pH < 5 (Table 1). The differences in the frequency of sites void of fish among the three pH classes were statistically significant (χ^2-test: $p < 0.001$). Average total fish density and biomass were the lowest at the sites with pH < 5 and the highest at the sites with pH > 6 (*see* Table 1). These differences were statistically significant (KW test: $p < 0.001$).

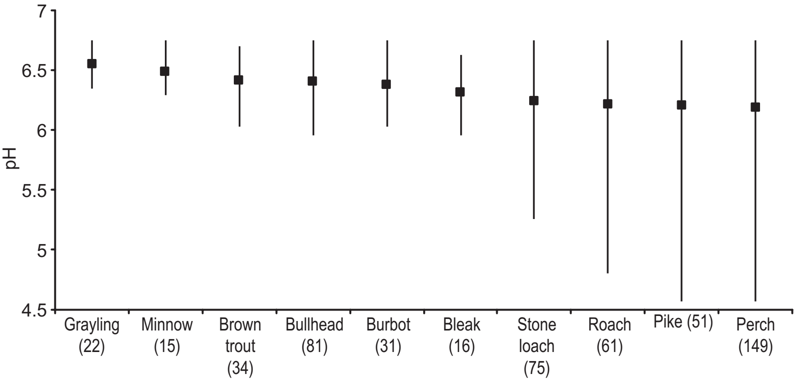

Fig. 4. Average river water pH (squares) and the range (vertical line) at the sampling sites inhabited by the fish species. Numbers of sampling sites inhabited by the fish species is given in parentheses.

The brown trout, grayling, bullhead (*Cottus gobio*) and minnow were the least tolerant of acidification, being usually absent from rivers whose water pH was less than 6 (Fig. 3). In comparison, pike and perch were recorded even at an average pH of less than 5 (Fig. 3).

Our study revealed that the grayling and minnow were the most sensitive and the perch and pike the least sensitive to acidification (*see* Fig. 4).

Discussion

Drainage of a.s. soils in western Finland promotes leaching of acidifying compounds and Al to rivers. Sites void of fish, low species richness and low total fish density were found at sites where river water pH was low and associated Al concentrations high. Relative tolerance of the fish species to acidity could be quantified based on their existence in the sampled rivers differing in water pH.

Average pH used here by us may not give a true picture of acidity, as the temporal variation in pH is often high. The lowest pH values in streams crossing sulphide-rich soil areas are generally recorded during high runoff after heavy rains or during snowmelt (Saarinen *et al*. 2010). Even episodic exposure to acidification has been found to have long-term effects on fish communities (Baker *et al*. 1996, Magee *et al*. 2003, McCormick *et al*. 2009). The problem with applying minimum pH values as the measure of acidification as water samples taken at certain time intervals may not reveal short-term peaks in acidity. Additionally, varying sampling frequency among rivers would be a source of additional bias.

Fish kills and other responses of fish to toxicity have been reported in the rivers and lakes on acid-sulphate soils in western Finland (Kjellman *et al*. 1994, Vuorinen *et al*. 1998, Hudd and Kjellman 2002, Toivonen and Österholm 2011). Also, there are about 70 articles in local newspapers (from the year 1834 onwards) reporting fish kills in the rivers sampled in this study and their estuaries (Sutela *et al*. 2012). In many of those articles, a short-term increase in transparency of the originally humic and thereby brownish waters was reported, which is known to indicate

Table 1. Electrofishing statistics for three river-water pH ranges.

	pH		
	< 5	5–6	> 6
Rivers	4	8	23
Electrofishing samples (*n*)	22	42	136
Frequency of no catch in electrofishing (%)	59.1	14.0	8.9
Average fish density (indiv. per 100 m^2) (range)	1.3 (0–12.7)	10.5 (0–66.7)	18.4 (0–181.1)
Average fish biomass (g) (range)	41.5 (0–487)	148.2 (0–1081)	179.1 (0–3467)

acute acidification. Reported fish kills tend to concentrate in years with high acidity in a.s. soil rivers of western Finland (Sutela *et al*. 2012). In 1969–1971, large drainage projects in this area induced extensive fish kills (Sundström and Åström 2006). These data support the notion of acidity being a crucial factor affecting riverine fish assemblages in the area of acid-sulphate soils in western Finland.

Acidity and high Al levels may also depress fish populations without easily visible fish kills of adult and young fish. Field and laboratory experiments have shown that a common mechanism of population extinction is post-embryonic mortality and subsequent lack of recruitment (Muniz and Leivestad 1980). Acidic water ($pH \leq 5.0$) during spawning and development of perch eggs and larvae resulted in recruitment failure (Linløkken *et al*. 1991). Acidification of the Kyrönjoki estuary in the studied area due to drainage from acidic soils has made part of the estuary unsuitable for fish reproduction (Urho *et al*. 1990). We inferred that acidity accompanied with high Al levels in the rivers of western Finland affect fish populations primarily via reproduction failures.

Al levels measured in the studied acidic rivers were very high as compared with those used in laboratory studies conducted to test the responses of fish to Al and acidity (cf. Brown 1983, Keinänen *et al*. 2000, Stephens and Ingram 2006). Maximum Al concentrations in laboratory exposures are usually < 1 mg l^{-1} (*see* e.g. Gensemer and Playle 1999), and in our study the concentration in the Vöyrinjoki was as high as 7.4 mg l^{-1}. This suggests that fish should have no possibility to reproduce or survive in the Vöyrinjoki. Electrofishing conducted in this river during this study in 2010–2012, and also three earlier electrofishing attempts in the early 2000s yielded no catch (Sutela *et al*. 2010). The fact that, unlike in many other rivers in the study area, no fish kills have been recorded in the Vöyrinjoki since the early 1970s (Sutela *et al*. 2012) may indicate the lack of fish due to the unsuitability of the river for fish reproduction as discussed above.

A comparison of the species-specific laboratory tolerance test results (e.g. Keinänen *et al*. 2000, Poléo *et al*. 1997, Dalziel *et al*. 1995, Gjedrem & Rosseland 2012) with our results suggests that some fish species were quite tolerant to acidity and Al. It should, however, be noted that river waters in western Finland are humic with high dissolved organic matter (DOM) concentrations. DOM protects against the adverse effects of Al by rendering Al in solution less damaging to fish gills (Gensemer and Playle 1999). In addition to Al, also other metals, such as zinc (Zn), cobalt (Co) and cadmium (Cd) are mobilised from a.s. soils in Finland (Fältmarch *et al*. 2008). The effects of these metals on the fish physiology and assemblages in the studied area was considered to be minor as compared with that of Al. Low pH and associated high Al concentration usually form the key combination affecting fish physiology (Muniz and Leivestad 1980, Wood 1989).

The ranking of fish species according to their tolerance to acidity was in line with the previous studies (cf. Urho *et al*. 1990, Bergquist 1991, Poléo *et al*. 1997). Perch and pike have been found to be tolerant also in several other field and laboratory studies (Almer 1974, Muniz 1984, Keinänen *et al*. 2000). However, in contradiction to the results (e.g. Almer 1974, Howells *et al*. 1990, Vuorinen *et al*. 1993), roach was found to tolerate acidity relatively well. One possible explanation for this discrepancy can be seasonal occurrence of roach in boreal river riffles (Sutela *et al*. 2017). Electrofishing in this study was performed in August–September when pH is quite high as compared with the values measured in spring and late autumn–winter lowered by high discharge in this area (Saarinen and Kløve 2012). In contrast to the dominating resident fish species in riffles (e.g. stone loach (*Barbatula barbatula*) and bullhead), roach may exploit a seasonal window of moderate pH levels in August–September. To our knowledge, the tolerance of the bullhead and stone loach to acidity has not been measured in laboratory or riverine field conditions. Our results suggest that the bullhead can be considered sensitive to acidity, whereas the tolerance of the stone loach is intermediate.

Leaching of acidity and Al to rivers can be reduced by modern, controlled farmland drainage techniques. Keeping a reasonably high groundwater level or re-flooding a.s. soils have shown to reduce acidity as well as immobilize Al and other potentially toxic metals (Toivonen and Österholm 2011). Also development of more environmentally friendly ditching practices (more shallow ditches) has been found advantageous (Sundström and Åström 2006). We suggest that all mitigation methods available should be fully applied to reduce the damage to riverine fish and other biota in the areas of acid-sulphate soils.

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