Characterization of the metal leakage from Finnish agricultural acid sulphate soils in the light of the European Water Framework Directive

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On the coastal plains of Finland, widespread acid sulphate soils have developed as a result of agricultural drainage activities and constitute a massive supplier of metals and acidic compounds to the aquatic environment. As a consequence, the aquatic life (fish, etc.) is seriously damaged in many water courses. The EU Water Framework Directive (WFD) which came into force in December 2000, sets new goals for the condition of Europe's waters. For surface waters the overall objective is 'good chemical and ecological status', which shall be achieved by December 2015. Furthermore, there is a list of 'priority substances' in the WFD, for which discharges, emissions and losses have to be reduced or phased out. The existing environmental data concerning the Finnish AS soils show that the current management of these soils results in aquatic impacts which clearly conflicts with the provisions of the WFD.

Introduction

Since the Middle Ages, sulphide-rich fine-grained sediments distributed on the coastal plains of Finland have been reclaimed for agriculture. In the 20th century, however, the drainage (ditching operations) of these sediments accelerated considerably, mainly as a consequence of the introduction of agricultural machines and subsurface drainpipes after the 2nd World War. A negative side effect of the drainage practices is that the penetrating air oxidises metal sulphides in the sediments resulting in production of sulphuric acid. The soil that develops thus becomes acidic and is called an acid sulphate (AS) soil (e.g. Öborn 1989, Dent and Pons 1995, Yli-Halla 1997, Cook *et al.* 2000, Ritsema *et al.* 2000).

The total area underlain by AS soils is not precisely known and estimates are dependent on the criteria used. Using U.S. Soil Taxonomy criteria, the total area in Finland of cultivated AS soils (Typic Sulfaquepts and Sulfic Cryaquept) is approximately 1000 km² (Yli-Halla *et al.* 1999, 2000). There is however another 2000 km² of cultivated soils (Dystric Cryaquept) probably generating acid solutes and metals to stream waters, which do not meet the Soil Taxonomy criteria of an AS soil. In these soils sulphidic materials/sulphuric horizons are deeper than 150 cm from the soil surface, which makes them non-

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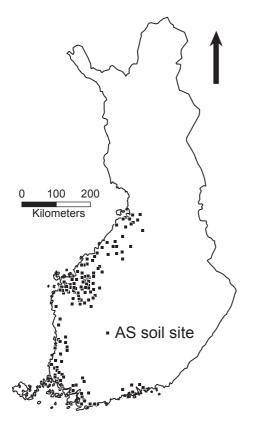


Fig. 1. The total distribution of acid sulphate soils (> 3000 km²) developed in Holocene fine-grained sediments in the coastal areas of Finland. The mapping survey covered a total of 740 random field sites along the coast, of which 20% was identified as an AS soil site. Each square represents ca. 20 km² AS soil. Modifield from Palko (1994).

diagnostic criteria in the soil taxonomy system. In artificially drained soils also these horizons are oxidized periodically and can thus produce a substantial amount of acidity that is leached into the drainage waters (Joukainen and Yli-Halla 2003). Therefore the total area of acidity producing farmland soils that can be considered as AS soils is estimated to be at least 3000 km² (Fig. 1). Analysis of topographical and landscape features and recent geochemical and hydrochemical data, clearly shows that only a minimal part of the AS soils would have developed under natural (unditched) conditions (Österholm 2005).

In order to make crop growth possible and more efficient on these soils, the pH of the topsoil (0-30 cm) is raised by extensive liming. In the subsoil, however, acidity and several metals are released and during subsequent wet periods transported, via subsurface pipes and/or open ditches, into streams (Weppling 1993, Åström and Åström 1997, Edén et al. 1999, Åström and Spiro 2000). This causes deterioration of spawning grounds and occasional sudden fish kills in streams and estuaries (e.g. Kjellman et al. 1994, Hudd and Kjellman, 2002). The first known mass fish kill occurred in 1834, the latest one was in 1996. Apart from Finland, AS soils are relatively widespread (1400 km²) in Sweden (Öborn 1989), while elsewhere in Europe they are scarce. Worldwide, potential and actual AS soils cover an area of some 240 000 km² and are widespread on coastal plains of Australia and south-eastern Asia (Ritsema et al. 2000). In general, the metal leakage from AS soils worldwide is poorly characterised and thus largely unrecognised as a potential massive source of mobile toxic metals.

The Water Framework Directive (WFD) (CEC 2000) is probably one of the most significant legislative instruments in the water field to be introduced on an international basis. For surface waters the overriding objective is to achieve 'good ecological and chemical status' (by 2015), which only allows a slight departure from the chemical, biological and hydrological characteristics which would be expected in conditions of no anthropogenic impact. In addition, there is a list of 33 'priority substances' in the directive (listed in Annex X of the WFD), including the metals cadmium (Cd) and nickel (Ni), for which discharges, emissions and losses have to be reduced or phased out. Within the list of 'priority substances' the Commission has identified 11 'priority hazardous substances' (including Cd), which are of particular concern for the aquatic environment. Member States are forced to implement necessary measures in order to cease or phase out discharges, emissions and losses of the 'priority hazardous substances' within 20 years of agreement on the list. The first list of 'priority substances' was adopted in November 2001 by the Council (CEC 2001). In addition to leaching from the Finnish AS soils of acidity and metals such as aluminium (Al), beryllium (Be), cobalt (Co), manganese (Mn) and zinc (Zn), several recent studies have indicated the leaching also of Cd and Ni in large amounts (e.g. Sundström *et al.* 2002, Åström and Deng 2003, Österholm and Åström 2004), i.e., metals found among the priority substances in the WFD. In this study we have (1) summarized existing information of Cd and Ni behaviour in Finnish AS soils, (2) scrutinized the measures aimed to mitigate the chemical load (acidity) from Finnish AS soils and (3) assessed how the WFD may affect the management and agricultural use of the AS soils, with consideration to the extensive leaching of Cd and Ni in particular. While the chemical leakage and subsequent dispersion of Cd and Ni only are presented, the principles are similar for many other metals abundantly leached from these soils.

Cadmium and nickel in the AS soil environment

The median concentrations of the naturally occurring Cd and Ni in potential AS soils (as yet unoxidised sulphidic/sulphuric horizons) on the coastal plains of Finland are 0.1 ppm (50 samples; unpublished data of the authors) and 31 ppm (317 samples; Åström and Björklund 1997) respectively. These are not high concentrations, as they are only marginally higher than those in the fine fraction (< 0.06 mm) of till in the country (Åström and Björklund 1997). In the potential AS soils, however, there exists a highly mobile metal pool. When the AS soil development starts after artificial drainage of these sediments, it is most likely that Cd and Ni held in easily oxidisable sulphides are gradually released, and as the acidification process proceeds, these metals are released from more resistant sulphides and possibly from additional phases including aluminosilicates and organic compounds (Åström 1998). The released fractions are moved downward in the soil profile and then either directly drained away resulting in extensive export of Cd and Ni and thus concentrations up to $25 \ \mu g l^{-1}$ and 1 mg l⁻¹, respectively, in low-order streams, or reimmobilised between the actual and potential AS soil, i.e. where pH sharply rises (the transition zone) (Åström and Deng 2003). This results in vertical patterns like the ones shown in Fig. 2.

In autumn 1990, the Geological Survey of Finland sampled and analysed over one thou-

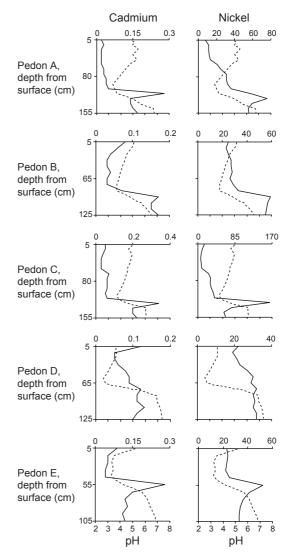


Fig. 2. Vertical variation in pH (dashed line) and in the concentrations of Cd and Ni in five pedons (all from central western Finland) consisting of acid sulphate soil (acidic horizon), transition zone (region of strong pH increase) and parent sediments (circumneutral zone). Modified from Åström and Deng (2003).

sand headwater streams (average drainage area: 30 km²) distributed throughout the country. The results, presented and discussed in detail by Lahermo *et al.* (1996) and illustrated for Cd and Ni in Fig. 3, show that many streams in the central parts of the western coast are clearly enriched in these two metals, and that several streams in other coastal areas are enriched in nickel (Cd mostly below the detection limit).

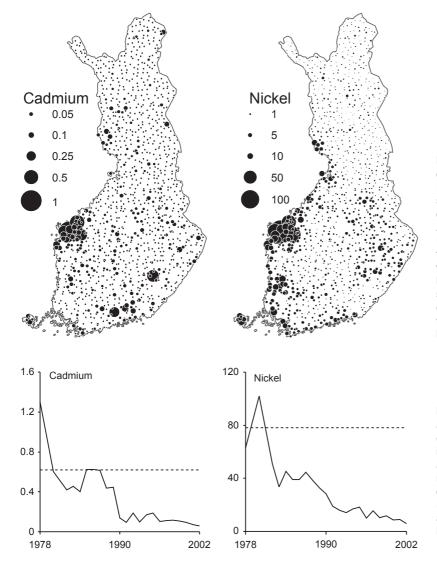


Fig. 3. Cadmium and nickel concentrations (μ g |⁻¹) in Finnish stream waters sampled throughout the country during base flow in August–September 1990. For cadmium, ca. 80% of the samples (937/1154) was below the detection limit (0.02 μ g |⁻¹). Data source: Geological Survey of Finland, Geochemistry Department, regional hydrogeochemical data.

Fig. 4. The cadmium and nickel discharges in effluent from the Finnish industry (continuous line) and the estimated annual metal leakage from agricultural AS soils in Finland (dashed line), expressed as t a^{-1} from 1978 to 2002. Modified from Sundström *et al.* (2002).

These hydrochemical features and the distribution of AS soils in the country overlap (Fig. 1). Subsequent studies have shown that this relationship is causal, i.e. the metals originate from the AS soils by the mechanism described above (Åström 2001).

In order to assess the amount of metals annually leached from the Finnish AS soils, a comparison has been made with the amount discharged in effluent from the Finnish industry (Sundström *et al.* 2002). The results for Cd and Ni are illustrated in Fig. 4. The trends, which are discussed in detail by Sundström *et al.* (2002), are very clear and reflect the fact that the metal leakage from the AS soils has remained unchanged, whereas the industry-related discharge over the last two decades, due to legal regulations and improved technology, has strongly decreased. As a consequence, at present the AS soils constitute a massive relative supplier of Cd and Ni (and other metals) to the aquatic environment.

In principle, the open ditches and drainpipes through which the AS waters exit to natural streams could be regarded as point sources. However, since they are numerous (hundreds of thousands) and mostly spatially unlocated (due to poor knowledge of AS distribution), the AS soil load of Cd and Ni and other metals has a diffuse character. In one respect, the AS soil drainage is similar to acid mine drainage occurring throughout Europe (e.g. Germany, Spain, Portugal, England and Poland), i.e. both are acidic and rich in metals, and the source of the chemical contaminants is oxidised sulphides in geological materials. There are, however, substantial differences in the environmental control of these two waters. The most important is the fact that the AS drainage has a diffuse character while the location and distribution of acid mine drainage is quite well known and thus easier to manage.

Attempts to reduce the chemical loads from AS soils

After extensive fish kills in several strongly acidified Finnish stream waters in 1969-1971, the AS soils were for the first time regarded as a potential source of environmental contamination. A working group, established by the National Board of Waters to investigate the background of the fish kills (National Board of Waters 1973), showed that these were linked with large drainage projects (installation of subsurface drainpipes) in actual/potential AS soils. These activities had increased the acidity and concentrations of Al, Fe and Mn in the exiting waters to a level toxic to the fish. In addition, based on more recent data, there is no question that the concentrations also of several toxic trace metals, incl. Cd and Ni, were strongly elevated in the waters. The working group investigating the fish kills made the following conclusions: (1) the total area of AS soils will, due to the need for more farmland, become more widespread in the future, (2) as long as methods of acidity neutralisation are not developed, massive acid discharges and concomitant unnatural fish kills will occur, and (3) a ceasing of the draining activities may be the only, but a controversial, strategy of preventing future fish kills. Nevertheless, the draining activities have continued unchanged, which have resulted in more production of acidity and metal release and, consequently, in more severe and widespread AS soils releasing toxic waters to the life-supporting aquatic bodies.

As a consequence of the gradually growing environmental concern of the AS soils, different liming strategies for mitigation purposes were applied. The most extensive liming-project 'Liming as a method to mitigate AS soil acidity in Finnish watercourses' was launched in 1985 (ending in 1995) to elucidate the possibilities to restore AS waters by different liming efforts (Weppling 1997). This project included: (1) intensified topsoil liming involving calcitic $(CaCO_2)$ and dolomitic $(CaMg(CO_2)_2)$ limestone powder (< 2 mm) application in large quantities on fields, (2) in-stream liming which means that powdered limestone or CaO is applied to running waters, and (3) lime filter drainage which means that a mixture of soil material and CaO is applied around and above the drainpipe. The results from these experiments showed that liming was not efficient due to the very high amounts of acidity stored in the AS soils and waters (Weppling 1997).

Despite the fact that the results from the liming efforts had been doubtful, intensified topsoil liming and lime filter drainage was included in the Finnish Agri-Environmental Support Scheme (FAESS), implemented in connection with Finland's EU membership in 1995. Thus, the FAESS has given farmers opportunities to take environmental measures which had, by scientists, been shown not to work very well. On intensified topsoil liming, which immediately gained popularity, 6 million euros was spent, part provided by the EU, during the period 1995–1999. However, the high economical costs and the small, if any, effects on the runoff water quality have eventually resulted in distrust to the liming applications which consequently have decreased over the last few years.

Another remedial method also included in FAESS is controlled drainage which has become more and more popular and is presently implemented at a rapid rate into farmlands. The mechanism of this method is to maintain a high water table via controllable wells. It thus has the potential not only to prevent aeration and consequently oxidation of the sulphides, but also to reduce the export of nutrients, increase the harvest and decrease the irrigation costs. The conception of this method is thus very good, and has also been shown to work on some 'less severe AS soils', where most of the acidity already has been transported to watercourses and where the sulphidic material exists at greater depths (Bärlund et al. 2005). However its effects on the runoff water quality have as yet been inadequately tested, and it is alarming that during long periods of drought in summer, the water table drawdown will, where this method is applied, be extensive and reach deep into the sulphidic layers. This means that with present management, sulphide oxidation and thus the release of acidity and associated metals (Cd, Ni and others) will persist, possibly however at a somewhat lower rate in some fields. Hence, the efforts to combat AS soil related aquatic acidification have so far overall been inefficient and unsuccessful.

Another drawback of FAESS is that it does not consider the metal leakage. One might assume that several metals should, at least to some extent, be immobilised if the pH level of the subsoil and drainage waters could be increased by liming or controlled drainage. However, in addition to the mostly unsuccessful attempts to increase the pH, there are many unanswered questions. To what extent will metals such as Cd and Ni remain in solution after a successful artificial increase in pH? Is a chemical time bomb produced if an abundance of potentially toxic metals, including Cd and Ni, are precipitated in small temporary circumneutral zones in an overall acidic environment?

In view of the fact that the first reports on extensive aquatic and biological damage caused by the Finnish AS soils were written already in the early 1970s, and that over the decades that have followed there has been an enormous increase in the general awareness of acidification and heavy metal pollution, it is surprising that so little has been done and achieved with respect to the AS soils.

The Water Framework Directive and future recommendations

The Finnish water (environmental) legislation has so far focused on control mechanisms for point sources without consideration of the status of the water quality itself. Whereas there are no specific criteria, thresholds or cut-off values for polluting substances entering the aquatic environment in the WFD, the 'combined approach' means that the member states have to set emission limit values for point sources as well as quality standards (EQSs) for pollutants, e.g. metals, identified as being discharged in significant quantities into bodies of surface waters. Compliance with the EQSs is required for the achievement of the objective of 'good chemical status'. In this context, the WFD offers a great opportunity for addressing the extensive aquatic pollution caused by the AS soils, which has more of a diffuse than point-source character. However, the challenge is big, especially considering the provision of cessation and a progressive reduction of discharges, emission and losses of priority hazardous substances (Cd) and priority substances (Ni) from which, in contrast to the aim of 'good status', there is no derogation. The Commission will, however, submit proposals of controls (emission controls for point sources) and environmental quality standards for the priority substances.

In order to improve the quality of the AS soil affected surface waters we suggest that a national strategy plan is drawn up including: (1) an elucidation of how the 'most severe AS soils', soils with high capacity of acid and metal generation can be restored by letting the groundwater table rise to a natural level close to the surface and subsequently utilized under the new hydrological conditions, (2) development of more environmental-friendly ditching practices (more shallow ditches) and improvement of the controlleddrainage technique (for 'less severe AS soils') and (3) juridical guidelines for future management of the AS soils under cultivation and for potential AS soils continuously brought above the sea level by the isostatic land uplift. Nevertheless, also after these measures, a leakage of acidity, Cd, Ni and other metals from the AS soils will continue, however at a smaller rate than at present. A kind of minimum demand in the WFD is that member states implement necessary measures to prevent further deterioration of the status of surface waters. This means that, at least until the recommended management plan has been made, all ditching operations carried out in (potential) AS soils are immediately stopped.

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