



## Abundant stocks and mobilization of elements in boreal acid sulfate soils



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### ABSTRACT

Large amounts of sulfate and divalent iron (Fe) are released into the pore water of acid sulfate (AS) soils upon oxidation of sulfidic materials. The simultaneously produced acidity dissolves metals from the soil matrix. Reduced horizons of AS soils commonly have a large mineral nitrogen (N) stock in the form of  $\text{NH}_4^+$ -N, which is a potential source of N leaching and gaseous emissions. This study was carried out at three AS soil sites in Finland. Cation composition of pore water was monitored in a monolithic lysimeter experiment. Timothy samples grown in an AS soil were analysed for mineral elements and the soil was investigated for zinc (Zn) distribution in different chemical species at four depths down to 85 cm. The composition of pipestems formed in previous root channels was investigated by SEM, X-ray EDX and XRD. Emissions of  $\text{CO}_2$  and  $\text{N}_2\text{O}$  were measured in an AS field which had a peaty topsoil. Monitoring by closed chambers was carried out at three sites differing in the depth of peat layer (15, 30 and 60 cm). In the sulfuric horizon, large amounts of calcium and magnesium were mobilized. Reflooding stopped the mobilization of those elements but resulted in abundant dissolution of Fe, which became the dominant cation in the pore water. Timothy growing in an AS soil showed only small deviations from the average composition measured in Finland with the exception of Zn that was at the deficiency level. As Zn in the root zone had been dissolved and subsequently leached, it was recovered in easily soluble forms in the subsoil. Pipestem composition indicated accumulation of Fe and formation of a new solid phase probably mostly in the form of schwertmannite even though jarosite was also detected. The annual  $\text{CO}_2$  emissions from the peaty AS soil were about  $6000 \text{ kg C ha}^{-1}$  but the  $\text{N}_2\text{O}$  emissions were relatively small, with a magnitude more typical of mineral rather than organic soils. Thus, large N stock of an AS soil do not necessarily contribute to abundant gaseous N emissions.

### 1. Introduction

Substantial changes in soil chemistry take place upon natural or artificial drainage and subsequent oxidation of sulfidic materials in acid sulfate (AS) soils. Sulfate and divalent Fe are released into the pore water from the oxidizing  $\text{FeS}/\text{FeS}_2$ . The pH is lowered from neutrality to values below 3.5, dissolving heavy metals and Al from the solid matrix. Part of the metals (at least Ni, Mn, Co and Cu) also originates from the oxidizing  $\text{FeS}_2$  (Sohlenius and Öborn, 2004; Claff et al., 2011). Owing to low pH, many metals released from the solid phase remain

dissolved in pore water in elevated concentrations resulting in increased leaching of these substances. Abundance of dissolved  $\text{Al}^{3+}$  displaces base cations from exchange sites and subjects them to leaching (Hartikainen and Yli-Halla, 1986). Gradually the oxidized horizons become saturated with exchangeable Al (Dent, 1986; Yli-Halla, 1997; Yvanes-Giuliani et al., 2014). Leaching of Al itself occurs in abundant amounts only after most exchange sites are occupied with  $\text{Al}^{3+}$  (Hartikainen and Yli-Halla, 1986; Shamshuddin et al., 2004; Virtanen, 2015). The stock of Al in soil is inexhaustible. There is a close relationship between acidity and sulfate concentration as well as Al and

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Fig. 1. The locations of the experimental sites. The shading around the Baltic Sea stands for the area that was under the Litorina Sea and is thus the potential area for the coastal acid sulfate soils.

sulfate concentration in stream and river water (Toivonen and Österholm, 2011), and strong negative relationships between pH and Al concentration in pore water, discharge water and groundwater in AS soils have been found (Virtanen et al., 2014b). Österholm and Åström (2002) estimated that within 2 m of soil surface the stock of S that can be leached from a typical AS soil of Rintala area in Finland was about 40 tons  $\text{ha}^{-1}$ . In this particular AS soil area about 630 kg S  $\text{ha}^{-1}$  was annually leached (Österholm and Åström, 2004) while in non-AS arable soils leaching has amounted to 15 kg  $\text{ha}^{-1}$  (Turtola and Jaakkola, 1986) or 20 kg  $\text{ha}^{-1}$  (Eriksen and Askegaard, 2000) in Finland and Denmark, respectively. Concentrations of metals in Finnish streams draining AS soils are much higher than in other streams (e.g., Åström and Björklund, 1995; Åström and Corin, 2000; Toivonen and Österholm, 2011). The total metal concentration in sulfidic sediments of Sweden and Finland is not higher than in other soils but the metals seem to be mobilized to a larger extent (Sohlenius and Öborn, 2004). Therefore, it has been hypothesized that plants growing in these soils may be high in heavy metals, but little proof has thus far been found (Palko, 1986; Fältmarsch et al., 2010).

Translocation of Fe out of the soil profile is not as marked as that of many other metals (Sohlenius and Öborn, 2004) and Fe concentrations of streams draining AS soils do not differ substantially from streams in non-AS soils (Åström and Corin, 2000; Sundström et al., 2002). Instead, upon oxidation of  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$  new solid phases are precipitated as continuous coatings or pipestems around former root channels, with Fe released from  $\text{FeS}/\text{FeS}_2$  being redistributed within the soil profile. However, in reflooded soils which contain plenty of precipitated Fe oxides, concentration of  $\text{Fe}^{2+}$  in the pore water can increase substantially (e.g. Johnston et al., 2014; Virtanen et al., 2014a) and result in formation of Fe sulfides (e.g. Burton et al., 2011; Johnston et al.,

2011) or leaching of Fe into watercourses (Cook et al., 2000; Virtanen, 2015). At the initial stages of oxidation and leaching, intermediate oxidation products such as jarosite  $\text{KFe}_3(\text{OH})_6(\text{SO}_4)_2$  and schwertmannite  $\text{Fe}_8\text{O}_8(\text{OH})_6(\text{SO}_4)_n\text{H}_2\text{O}$  are commonly identified in AS soils, and upon further leaching, they are gradually transformed into Fe hydroxides. The pipestems have a marked effect on the hydraulic properties of an AS soil. While the reduced subsoil is practically impermeable, the pipestems and permanent desiccation cracks stabilized by Fe hydroxides create effective transport routes for substances in the oxidized horizons (Dent, 1986; Joukainen and Yli-Halla, 2003; Johnston et al., 2009).

Abundant reserves of  $\text{NH}_4^+$ -N have been measured in the reduced subsoil, creating potential for N leaching or gaseous N losses. In western Finland Paasonen-Kivekäs and Yli-Halla (2005) measured as much as 435 kg  $\text{ha}^{-1}$  mineral N in the upper 240 cm of an AS soil while 137 kg  $\text{ha}^{-1}$  was measured in an adjacent non-AS soil. Results of similar magnitude have been obtained also in an AS soil in Helsinki, Finland by Šimek et al. (2011), who suggested that  $\text{NH}_4^+$ -N originates from slow decomposition of organic matter and accumulates in the subsoil because nitrification does not take place in those permanently water-logged conditions. Uusi-Kämpä et al. (2012) measured N losses of as much as 50 kg  $\text{ha}^{-1}$  in drainage water from an AS soil while the average amount leached from soils of Finland is about 16 kg  $\text{ha}^{-1}$  (Tattari et al., 2017). The abundance of plant available N in soil was also demonstrated by the fact that wheat yields contained 140–150 kg N  $\text{ha}^{-1}$  while only 90 kg N  $\text{ha}^{-1}$  had been applied in fertilizer (Uusi-Kämpä et al., 2012).

Often stocks, fluxes and transformations of different substances are investigated one by one in separate studies. In this paper, we try to pull together information of multiple elements from boreal acid sulfate soils

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