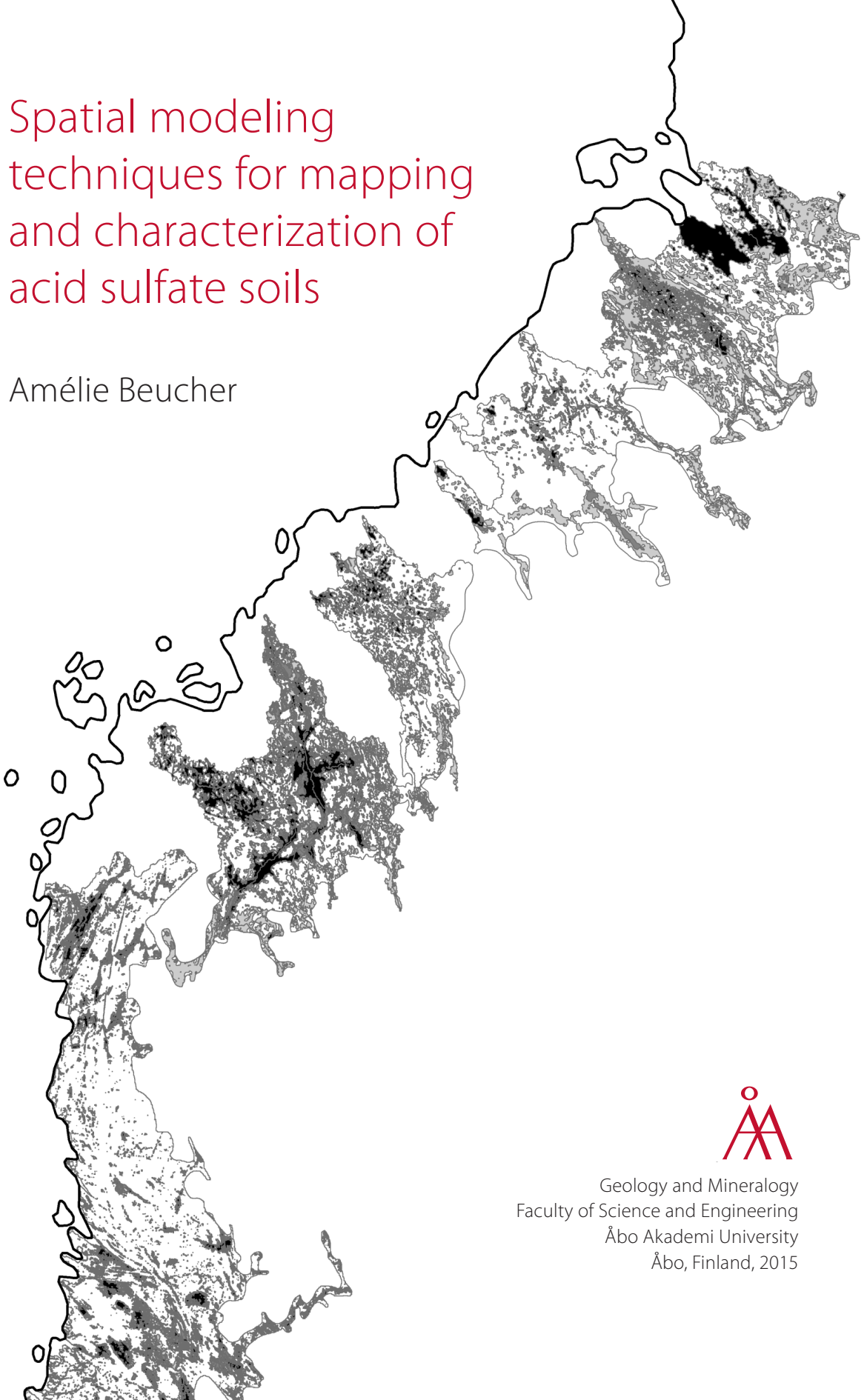


Spatial modeling techniques for mapping and characterization of acid sulfate soils

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Geology and Mineralogy
Faculty of Science and Engineering
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Abstract

Acid sulfate (a.s.) soils constitute a major environmental issue. Severe ecological damage results from the considerable amounts of acidity and metals leached by these soils in the recipient watercourses. As even small hot spots may affect large areas of coastal waters, mapping represents a fundamental step in the management and mitigation of a.s. soil environmental risks (i.e. to target strategic areas). Traditional mapping in the field is time-consuming and therefore expensive. Additional more cost-effective techniques have, thus, to be developed in order to narrow down and define in detail the areas of interest. The primary aim of this thesis was to assess different spatial modeling techniques for a.s. soil mapping, and the characterization of soil properties relevant for a.s. soil environmental risk management, using all available data: soil and water samples, as well as datalayers (e.g. geological and geophysical).

Different spatial modeling techniques were applied at catchment or regional scale. Two artificial neural networks were assessed on the Sirppujoki River catchment (c. 440 km²) located in southwestern Finland, while fuzzy logic was assessed on several areas along the Finnish coast. Quaternary geology, aerogeophysics and slope data (derived from a digital elevation model) were utilized as evidential datalayers. The methods also required the use of point datasets (i.e. soil profiles corresponding to known a.s. or non-a.s. soil occurrences) for training and/or validation within the modeling processes. Applying these methods, various maps were generated: probability maps for a.s. soil occurrence, as well as predictive maps for different soil properties (sulfur content, organic matter content and critical sulfide depth). The two assessed artificial neural networks (ANNs) demonstrated good classification abilities for a.s. soil probability mapping at catchment scale. Slightly better results were achieved using a Radial Basis Function (RBF) -based ANN than a Radial Basis Functional Link Net (RBFLN) method, narrowing down more accurately the most probable areas for a.s. soil occurrence and defining more properly the least probable areas. The RBF-based ANN also demonstrated promising results for the characterization of different soil properties in the most probable a.s. soil areas at catchment scale. Since a.s. soil areas constitute highly productive lands for agricultural purpose, the combination of a probability map with more specific soil property predictive maps offers a valuable toolset to more precisely target strategic areas for subsequent environmental risk management. Notably, the use of laser scanning (i.e. Light Detection And Ranging, LiDAR) data enabled a more precise definition of a.s. soil probability areas, as well as the soil property modeling classes for sulfur content and the critical sulfide depth. Given suitable training/validation points, ANNs can be trained to yield a more precise modeling of the occurrence of a.s. soils and their properties. By contrast, fuzzy logic

represents a simple, fast and objective alternative to carry out preliminary surveys, at catchment or regional scale, in areas offering a limited amount of data. This method enables delimiting and prioritizing the most probable areas for a.s soil occurrence, which can be particularly useful in the field. Being easily transferable from area to area, fuzzy logic modeling can be carried out at regional scale. Mapping at this scale would be extremely time-consuming through manual assessment. The use of spatial modeling techniques enables the creation of valid and comparable maps, which represents an important development within the a.s. soil mapping process.

The a.s. soil mapping was also assessed using water chemistry data for 24 different catchments along the Finnish coast (in all, covering c. 21,300 km²) which were mapped with different methods (i.e. conventional mapping, fuzzy logic and an artificial neural network). Two a.s. soil related indicators measured in the river water (sulfate content and sulfate/chloride ratio) were compared to the extent of the most probable areas for a.s. soils in the surveyed catchments. High sulfate contents and sulfate/chloride ratios measured in most of the rivers demonstrated the presence of a.s. soils in the corresponding catchments. The calculated extent of the most probable a.s. soil areas is supported by independent data on water chemistry, suggesting that the a.s. soil probability maps created with different methods are reliable and comparable.

Sammanfattning

Sura sulfatjordar (SSJ) utgör ett stort miljöproblem eftersom surt vatten med stora mängder metaller kan frigöras och belasta vattendrag med allvarliga ekologiska skador som följd. Eftersom även små förekomster av SSJ kan påverka stora delar av kustvattnen så är det viktigt att kartera de mest kritiska områdena (s.k. ”hot spots”) för att kunna tillämpa olika miljömetoder där de gör störst nytta. Traditionell kartering kräver omfattande och tidskrävande fältarbete och är därför mycket dyrt. För att avgränsa och identifiera de viktigaste områdena måste kompletterande och mer kostnadseffektiva metoder utvecklas. Den primära målsättningen med denna avhandling är att utvärdera olika rumsliga modelleringsmetoder för kartering och karakterisering av markegenskaper som är relevanta för miljöriskhantering av SSJ, med hjälp av mark- och vattendata samt befintligt data över geologi, geofysik och höjdförhållanden.

Olika metoder för rumslig modellering tillämpades på enskilda avrinningsområden eller på större regioner längs med den finländska kusten. Vid utvärderingen tillämpades två olika metoder baserade på s.k. Artificiella Neurala Nätverk (ANN), Radial Basis Function (RBF) -baserade ANN och Radial Basis Functional Link Net (RBFLN), på Sirppu å:s avrinningsområde (c. 440 km²) i sydvästra Finland, medan en metod baserad på s.k. Fuzzy logic tillämpades på bl.a. den södra delen av Finlands kustområden (c. 17,300 km²). Modellerna baserades på data över kvartärgeologi, aerogeofysik och markens lutning (härlett ur en digital höjdmodell). Utöver dessa krävdes också en uppsättning punktdata (dvs. markprofiler motsvarande kända SSJ eller ej kända-SSJ förekomster) för inlärning och/eller validering av modelleringsprocesserna. Genom att tillämpa dessa metoder genererades kartor över sannolikheten för förekomst av SSJ och samtidigt gjordes försök att modellera olika markegenskaper (svavelhalt, organiskt material och kritiskt sulfiddjup). Båda ANN metoderna fungerade bra för kartering av enskilda avrinningsområden. Avgränsningen av områden med mest och minst sannolika sulfatjordsområden var något noggrannare med RBF-baserade ANN än med RBFLN. Lovande resultat för karakteriseringen av olika markegenskaper i de mest sannolika SSJ områden inom enskilda avrinningsområden erhöles också med den RBF-baserade ANN-metoden. Det är också värt att notera att resultaten blev bättre när modernt material med höjddata från flygburen laserskanning (LiDAR) utnyttjades. Kartor över sulfatjordars utbredning samt deras egenskaper utgör ett viktigt verktyg för att kunna anpassa och rikta olika miljömetoder till strategiskt viktiga områden, bl. a. inom jordbruket eftersom de mest produktiva jordbruksmarkerna ofta förekommer på SSJ. Modellering med ANN-metoder kräver lämpliga datapunkter över förekomsten av sulfatjordar och icke- sulfatjordar för

inlärning och validering, och precisionen på kartor och modellerade markegenskaper blir bättre ju mer punktdata som finns till förfogande. Är den befintliga punktdatamängden däremot mer begränsad så är metoder baserade på fuzzy logic ett enklare och snabbare sätt att objektivt genomföra preliminära undersökningar på enskilda avrinningsområden eller i större regional skala. I och med att man då snabbt kan begränsa och prioritera de mest sannolika SSJ förekomsterna fungerar då modelleringen som ett viktigt verktyg vid upplägg av fältstudier. Tack vare att metoderna enkelt kan utvidgas att omfatta nya områden så kan större regioner modelleras och mycket tid kan sparas genom att minimera en tidskrävande manuell analys. Utnyttjande av olika tekniker för rumslig modellering har gett goda resultat och utgör en vidareutveckling av karteringen av SSJ.

Kvaliteten på SSJ på kartor producerade med olika metoder (ANN och fuzzy logic) samt med konventionell kartering för sammanlagt 24 avrinningsområden längs med den finländska kusten (c. 21,300 km²) utvärderades också genom jämförelse med vattenkemiskt data insamlat från respektive avrinningsområde. Två vattenkemiska indikatorer som under ideala förhållanden är direkt relaterade till förekomsten av SSJ (sulfathalt och sulfat/klorid-förhållande) i åvatten jämfördes med den modellerade omfattningen av SSJ för de undersökta avrinningsområdena. Omfattningen av SSJ stöds av de vattenkemiska indikatorerna data, vilket indikerar att kartorna är tillförlitliga och jämförbara sinsemellan. Användningen av kemiska indikatorer i vatten utgör således ett kompletterande och oberoende verktyg för att granska pålitligheten av SSJ sannolikhetskartor.

List of publications

This thesis is composed of four studies, of which three have been published in scientific journals and one has been accepted. The scientific papers, listed below, are referred to in the text by their corresponding Roman numerals **I-IV**. Amélie Beucher was chiefly responsible for the data handling, modeling and interpretation for papers **I** to **IV**. All papers are reproduced by permission of the journals concerned.

I. Beucher, A., Österholm, P., Martinkauppi, A., Edén, P. & Fröjdö, S., 2013. Artificial neural network for acid sulfate soil mapping: Application to the Sirppujoki River catchment area, southwestern Finland. *Journal of Geochemical Exploration* 125, 46-55.

II. Beucher, A., Fröjdö, S., Österholm, P., Martinkauppi, A. & Edén, P., 2014. Fuzzy logic for acid sulfate soil mapping: application to the southern part of the Finnish coastal areas. *Geoderma* 226-227, 21-30.

III. Beucher, A., Siemssen, R., Fröjdö, S., Österholm, P., Martinkauppi, A. & Edén, P., 2015. Artificial neural network for mapping and characterization of acid sulfate soil: Application to Sirppujoki River catchment, southwestern Finland. *Geoderma* 247-248, 38-50.

IV. Beucher, A., Fröjdö, S., Österholm, P., Auri, J., Martinkauppi, A. & Edén, P., 2015. Assessment of acid sulfate soil mapping utilizing chemical indicators in recipient waters. *Bulletin of the Geological Society of Finland*. Accepted and published online.

List of abbreviations

AMD	Acid Mine Drainage
ANN	Artificial Neural Network
a.s.	Acid sulfate
ASTER	Advanced Spaceborne Thermal Emission and Reflection Radiometer
CHNS	Carbon Hydrogen Nitrogen Sulfur
DEM	Digital Elevation Model
DSM	Digital Soil Mapping
GIS	Geographic Information System
GTK	Geologian Tutkimuskeskus (Geological Survey of Finland)
ICP-OES	Inductively Coupled Plasma-Optical Emission Spectrometry
LiDAR	Light Detection And Ranging
LOI	Loss On Ignition
MLP	Multilayer Perceptron
MML	Maanmittauslaitos (National Land Survey)
RBF	Radial Basis Function
RBFLN	Radial Basis Functional Link Net
RSNNS	R Stuttgart Neural Network Simulator
SDM	Spatial Data modeler
SSJ	Sura sulfatjordar (acid sulfate soils)

Table of Contents

1 Introduction	1
2 Study areas	5
3 Material	7
3.1 Soil samples	7
3.2 Water samples.....	9
3.3 Raster datasets.....	10
4 Methods	13
4.1 Conventional mapping	13
4.2 Spatial modeling techniques	13
4.2.1 <i>Fuzzy logic</i>	15
4.2.2 <i>Artificial neural networks</i>	17
5 Results and discussion	21
5.1 Fuzzy logic for preliminary surveys at regional or catchment scale	21
5.2 Artificial neural networks for mapping at catchment scale.....	23
5.3 Artificial neural networks for soil characterization at catchment scale	25
5.4 Impact and limitations of the different input evidential datalayers..	29
5.5 Validation as a critical modeling step.....	30
5.6 Assessment of acid sulfate soil mapping utilizing chemical indicators in recipient waters	31
5.7 Estimation of the extent of the Finnish acid sulfate soils	35
6 Conclusions	37
Acknowledgements	39
References	41
Original publications	53

1 Introduction

Since the Litorina-Sea stage (starting c. 8,000 years BP, during Holocene), large amounts of sulfide-bearing fine-grained sediments have been formed under anoxic conditions at the bottom of the Baltic Sea. Because of the strong isostatic land uplift (today up to 8 mm/year; Donner, 1995), these sediments have been raised up to 90 m above current sea level (Erviö, 1975; Palko, 1994; Fig. 1). The soils originating from these sulfide-bearing sediments constitute the most productive farmland areas in Western Finland. As they tend to be located in low-relief river valley plains, they are heavily drained, mostly for agricultural purposes. These soils can also be drained to enable forestry, peat mining or building infrastructure. Therefore, their upper layer (1 to 2 m) has generally been oxidized into acid sulfate (a.s.) soils ($\text{pH} < 4$), the sulfides reacting with the atmospheric oxygen to produce sulfuric acid subsequently lowering the soil pH. These soils can thus release a toxic combination of acidity and metals (e.g. Al, Cd, Co, Ni, Zn and rare earth elements) to the recipient streams and estuaries (Österholm and Åström, 2004), causing severe ecological damage such as wide-spread fish kills (Hildén and Rapport, 1993; Hudd and Leskelä, 1998; Åström et al., 2005; Saarinen et al., 2010; Toivonen et al., 2013).

Finland has the largest a.s. soil occurrence in Europe (in the order of 1,000 km² using Soil Taxonomy criteria; Yli-Halla et al., 1999). These drained soils bring notably more metals to the water courses than the whole Finnish industry combined (Sundström et al., 2002). The environmental problems related to a.s. soils might also affect more than one third of the Finnish coastal waters (Roos and Åström, 2005) as notably small hotspot areas can impact on large water areas. Mapping these hotspot areas constitutes a critical step for effective mitigation. Traditional mapping in the field is time-consuming and therefore expensive so, to cover large areas, additional more cost-effective techniques have to be developed in order to narrow down and define in detail the areas of interest. After massive fish kills in 2006 and 2007, the Geological Survey of Finland (GTK) initiated a cooperation network in order to create a nationwide a.s. soil map and mitigate the environmental problems due to these soils. This cooperation work also comes within the scope of the EU Water Framework Directive (2000) which aims at restoring a good status for ground and surface waters in Europe. According to Andriess and Meensvoort (2006), a.s. soils presumably cover about 200,000 km² of coastal areas worldwide, in many cases heavily populated with consequent high demand on clean water. Many of these potential a.s. soil areas are poorly known and mapping arises as crucial within this global context, whether at regional scale or at catchment scale. Furthermore, a refined characterization of a.s. soil in the areas of interest defined through mapping can be carried out

through the predictive modeling of important soil properties: sulfur content, organic matter content and critical sulfide depth (i.e. the level where the sulfide reservoir can be reached and, thus, how deep drainage can be carried out without mobilizing acidity from it). This characterization also constitutes an essential step for effective mitigation, providing very useful information for land use, management and rehabilitation.

In Australia, where researchers continuously pioneer a.s. soil mapping, traditional soil survey, air photo interpretation and numerical classification approaches (e.g. Tulau, 1999; Bierwirth and Brodie, 2005) have been used. Recently, the Digital Soil Mapping (DSM) approach was also tested to map a small coastal a.s. soil area, using fuzzy *k*-means algorithm for clustering analysis (Huang et al., 2014). Studies generally rely on various datasets: proximal sensing electromagnetic data, airborne gamma-radiometric data, Digital Elevation Model (DEM), satellite Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER) data, geological maps, as well as marine habitat, tidal, estuarine, bathymetric, vegetation and remotely sensed data, among others. In Finland, a.s. soil mapping has been carried out using conventional (i.e. field-based or in-situ) mapping, soil sampling and subsequent pH-measurements (Purokoski, 1959; Erviö, 1975; Palko, 1994; Puustinen et al., 1994; Triipponen, 1997). A similar procedure was utilized in Denmark (Madsen et al., 1985). Because of previous studies, the localization of a.s. soils is known in broad outline (Palko, 1994; Puustinen et al., 1994; Yli-Halla et al., 1999). They are generally found below the highest shoreline of the former Litorina Sea (Fig. 1), ranging from up to 100 m above current sea level for the northernmost coastal areas to 20 m for the southernmost (Eronen, 1974). Nevertheless, the Finnish a.s. soil extent was imprecisely estimated in previous studies as the criteria used for defining these soils were inappropriate and/or the available data was insufficient.

In this study, different spatial modeling techniques based on existing geostatistics were utilized to map the large potential areas along the entire Finnish coast. These techniques can be applied within various environments, for instance within R (for implementation) and a Geographical Information System (GIS; for visualization), or solely within a GIS. Among the diverse techniques available for spatial modeling, fuzzy logic and two artificial neural networks (ANN) were selected and assessed. The selection of a technique mainly depends on the data available for a specific study. As described by Bonham-Carter (1994), fuzzy logic is a conceptual, knowledge-driven approach, while ANNs are empirical, data-driven. Fuzzy logic adequately represents the imprecise and/or incomplete information contained in different datalayers. ANNs constitute good pattern recognition and classification tools with the ability to generalize from imprecise input data (Porwal et al., 2003).

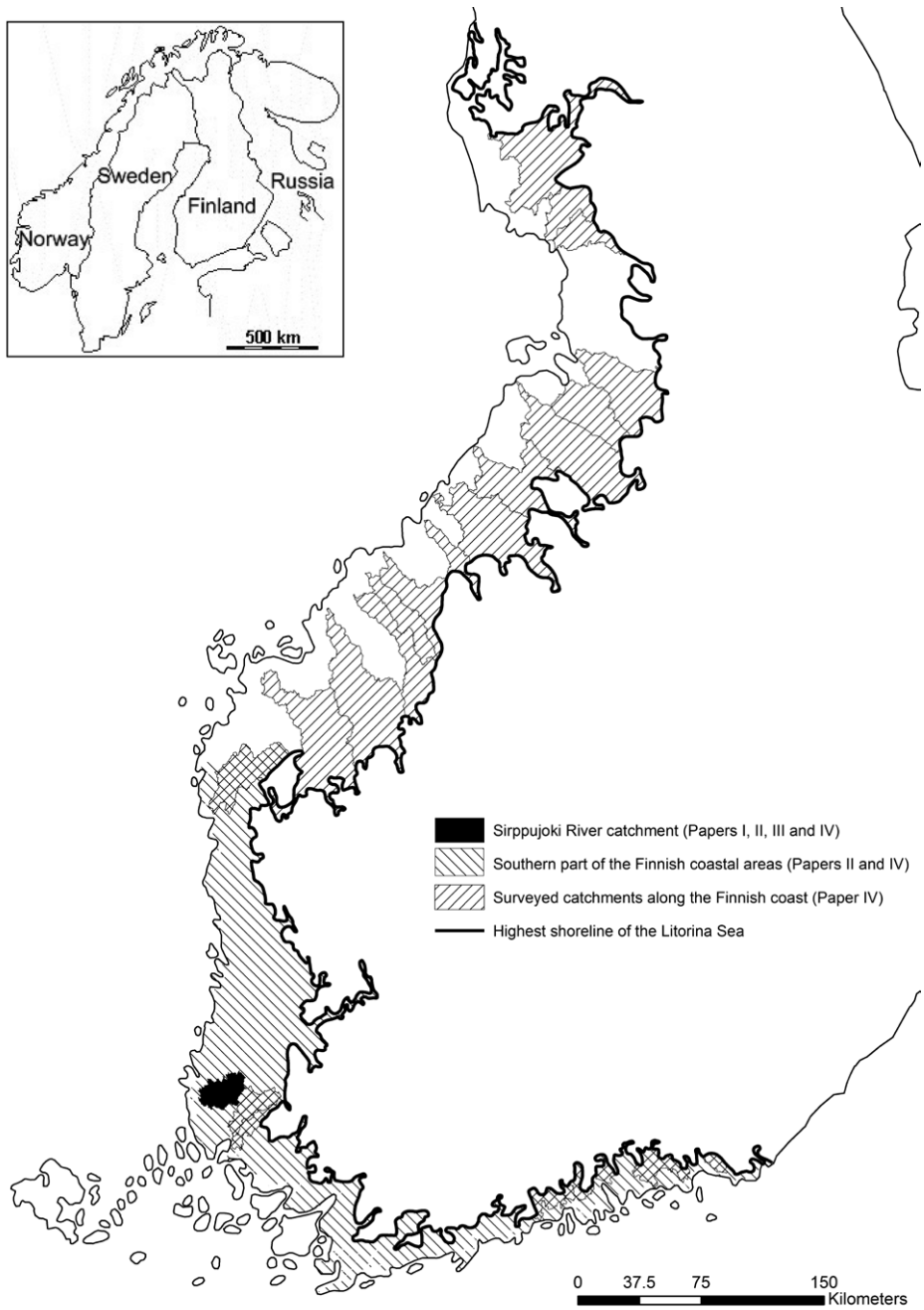


Figure 1. Location of the study areas and diachronous maximum extension of the former Litorina Sea (7,000 - 9,000 BP; Ojala et al., 2013).

These techniques both require the use of various evidential datalayers (also called environmental data or ancillary data within the DSM approach; i.e. DEM, Quaternary and aerogeophysical data). Fuzzy logic is particularly relevant to predict soil properties in areas with an insufficient number or even no known occurrences and thus enables the carrying out of preliminary surveys displaying an overview of potential areas for a.s. soil occurrence at reconnaissance scale (Papers **II** and **IV**). As a supervised, data-driven method, ANNs conversely necessitate known occurrences for network training (i.e. a.s. and non-a.s. soil sites). Moreover, ANNs enable not only the carrying out of a.s. soil probability mapping at catchment scale (Papers **I** and **III**), but also the characterization of different properties for these soils (i.e. the sulfur content, the organic matter content and the critical sulfide depth; Paper **III**). In addition to the papers included in this doctoral thesis, this study resulted in several presentations and peer-reviewed abstracts for scientific congresses and colloquia (Beucher et al., 2012, 2014a, 2014b, 2014c).

The overarching aim of this thesis is to apply and assess a selection of modeling methods for (1) a.s. soil mapping at various scales (catchment scale for Papers **I**, **III** and **IV**, regional scale for Papers **II** and **IV**), and (2) a.s. soil characterization (Paper **III**), using all available data: datalayers, soil and water samples. The presented methods are here central to mapping and localization of a.s. soils, but the versatility of the ANNs allows the study to venture into characterizing the soils as precisely as possible. Furthermore, the use of water chemistry data constitutes a novel and important way to assess the a.s. soil mapping results, comparing the calculated extent of a.s. soils in a catchment to their chemical indicators in the corresponding river (Paper **IV**). To our knowledge, spatial modeling techniques such as fuzzy logic and ANNs have not yet been applied for a.s. soil mapping and characterization. This whole study constitutes, thus, pioneering research work in this specific domain.

2 Study areas

This thesis consists of four papers based on different study areas along the Finnish coast (Fig. 1). Papers **I** and **III** are focused on a small catchment area located in southwestern Finland, the Sirppujoki River catchment (c. 440 km²). Paper **II** covers the southern part of the coastal areas (c. 17,300 km²). Paper **IV** discusses 24 different catchments located along the Finnish coastline (in total, c. 21,300 km², including the Sirppujoki River catchment and some other small catchments located in the southern part of the coastal areas, among others).

The coastal areas located below the highest shoreline of the former Litorina Sea (c. 50,100 km²) are considered the potential areas for a.s. soil occurrence in Finland (Fig. 1). The northern part of the coastal areas extend from the coast to 90-100 m above the current sea level. The southwestern areas reach 40-90 m above current sea level while the southernmost areas reach 20-40 m above current sea level.

Considering the bedrock geology, the coastal areas can be divided into two zones: the southwestern and northern zones. The southwestern zone is underlain by Svecofennian bedrock (c. 1.93-1.89 Ga), comprising volcanic and sedimentary rocks which were folded, metamorphosed and intruded by granitoids and mafic rocks (Taipale and Saarnisto, 1990). The northern zone (approximately from Oulu) is underlain by Archean bedrock (around 3.2-2.5 Ga) and younger volcanic and sedimentary rocks (around 2.5-1.89 Ga). Carbonaceous rocks are notably scarce and largely occur as marble. The ancient, crystalline bedrock is mostly covered by glacial and post-glacial Quaternary deposits, such as till, glaciofluvial sediments (i.e. eskers, sand and gravel), littoral sediments (i.e. sand and gravel), marine and lacustrine clays, silt and peat (Donner, 1995). These Quaternary sediments were mainly formed and deposited during the late Weichselian glaciation and the following deglaciation phases (about 28-10 ka), the Ancylus-Lake (about 10.7-9.8 ka) and Litorina-Sea stages (starting at about 9-8 ka) which evolved to the present Baltic Sea (Wohlfarth et al., 2008).

More detailed information about the characteristics of the different study areas can be found in Papers **I** to **IV**.

3 Material

An overview of the material utilized in this study is presented in Table 1.

3.1 Soil samples

Soil profiles and probing points were collected by conventional soil sampling carried out during the summer from 2009 to 2013. In comparison with previous studies, a novel classification system, more adapted for Finnish a.s. soils (Edén et al., 2012a), was applied. Finnish a.s. soils usually are fine-grained (clay or silt), contain gyttja and display an elevated sulfur content. They generally consist of an oxidised acidic horizon (i.e. actual a.s. soil) and/or a reduced sulfide-bearing horizon (i.e. potential a.s. soil). For actual a.s. soils, field pH is lower than 4.0 because of the sulfide oxidation. If field pH is between 4.0 and 4.4 and no underlying sulfides were observed, further investigations are required (i.e. incubation and sulfur analysis; Edén et al., 2012b). For potential a.s. soils, sulfides are present, field pH is generally higher than 6.0, the total sulfur content is higher or equal to 0.2% and incubation pH is lower or equal to 4.0 with a drop of more than 0.5 units.

Using a portable gouge auger (diameter of 3 cm; Pansu et al., 2001), the profiles were sampled at vertical depth intervals of 20 cm down to 3 m depth. For the probing points, a probe (diameter of 1.5 cm) was utilized and the soil/parent material was generally sampled at one to three different depths depending on the sediment lithology. The sampling sites were chosen in order to cover the areas of interest previously defined (cf. the conventional mapping sub-section of the methods; Paper IV; Österholm et al., 2010; Edén et al., 2012b). Within 24 h from sampling, pH was measured for each sample (deionized water was added to allow proper contact between the electrode and the soil material). A selection of samples was also analyzed for sulfur with Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) after digestion in aqua regia. For samples with a field pH higher than 4.0, pH was measured again after 9 to 16 weeks of incubation at room temperature. Soil profiles were considered as actual a.s. soils when they comprised an oxidised acidic layer (pH lower than 4.0) with an underlying sulfidic horizon within 3 m depth. They were considered as potential a.s. soils when pH decreased at least by 0.5 units to a value lower or equal to 4.0 after incubation (Soil Survey Staff, 1999; Edén et al., 2012b). In the case of the Sirppujoki River catchment, a small number of soil profiles ($n = 13$) previously studied by Triipponen (1997) were included (Papers I and III). For the soil property predictive modeling applied to the Sirppujoki River catchment (Paper III), some soil samples were further investigated. The organic matter content (%) was estimated as

Table 1. Overview of material and methods in Papers I to IV.

	Paper I	Paper II	Paper III	Paper IV
<i>Method(s)</i>	RBLFN	Fuzzy logic	RBF-based ANN	Conventional mapping Fuzzy logic RBFLN
<i>Study area(s)</i>	Sirppujoki River catchment (c. 400 km ²)	Southern half of the Finnish coastal areas (c. 17,300 km ²)	Sirppujoki River catchment (c. 440 km ²)	24 catchments along the Finnish coast (c. 21,500 km ²)
<i>Datalayers</i>	1:20,000 Quaternary geology 2 aerogeophysical datalayers*: • imaginary component • real component DEM**	1:200,000 Quaternary geology 1 aerogeophysical datalayer*: • imaginary component • real component DEM**	1:20,000 Quaternary geology 2 aerogeophysical datalayers*: • imaginary component • real component DEM & LIDAR-based DEM***	1:20,000 Quaternary geology 1:200,000 Quaternary geology 2 aerogeophysical datalayers*: • imaginary component • real component DEM & LiDAR-based DEM*** Bedrock geology Peatland
<i>Soil samples</i>	For training and validation: 62 profiles	For validation: 72 profiles	For training and validation: 62 profiles	For conventional mapping: 3271 profiles and probing points For training and/or validation: 827 profiles and probing points
<i>Water samples</i>				227 + 8 water samples

NB: *High resolution low altitude airborne geophysics, flight altitude 30-40 m and line spacing 200 m

**Original cell size 25 x 25 m and elevation data accuracy 2 m

***Original cell size 2 x 2 m and elevation data accuracy 0.3 m

loss on ignition (LOI) by combusting about 1.0 g of sample (first dried at 105 °C) for 4 hours in an oven at 500 °C (Radojević and Bashkin, 1999). LOI is proportional to the amount of organic carbon and constitute an accurate method for estimating the organic matter content. Nordmyr et al. (2008) found that LOI and organic carbon were strongly correlated ($r = 1.0$ with both Spearman and Pearson correlations). The sulfur content was determined with a Carbon Hydrogen Nitrogen Sulfur (CHNS) Elemental Analyzer (i.e. flash combustion followed by gas chromatography). For each profile, the organic matter content value used within the modeling was measured at the same depth (60-80 cm), below the plough layer and the potential peat layers, in order to represent the underlying potentially oxidised sediments. For each profile, the sulfur content value used within the modeling was measured within the anoxic parent sediments (i.e. below 100 cm depth). The critical sulfide depth was here defined as the upper limit of the sulfidic soil material which had a field pH larger than 4.5 due to limited oxidation, but reached a pH smaller than 4.0 upon 16 weeks of incubation. More detailed description can be found in Papers I to IV. Soil profiles and probing points were used for conventional mapping (Paper IV), but also for the different spatial modeling techniques as training and/or validation points (Papers I–IV; Table 1). Within the a.s. soil probability mapping, the a.s. and non-a.s. soil occurrences are considered as positive and negative points, respectively.

3.2 Water samples

Water data (sulfate and chloride contents, as well as pH and electric conductivity) was extracted from the HERTTA database (OIVA-environment and geographic information service, Finnish Environment Institute) for 21 catchments, and from Nyberg et al. (2011) for the 3 remaining catchments (Paper IV). In all, 235 water samples were utilized in this study (Table 1 and detailed distribution in Table 5). The collected water data was sampled as close as possible from the mouth of the rivers during spring 2002-2012. High flow conditions are best to detect a.s. soil related indicators and comparable hydrological conditions are required to get representative results. Water samples collected from April to May were, thus, utilized in this study, as high flow conditions occur at different times depending on the location along the coast. Electric conductivity and pH were measured in the field. The samples were filtered (0.45 μm) before being analyzed for sulfate and chloride content by ion chromatography for samples from Hertta database and by ICP-OES for samples from Nyberg et al. (2011) (Table 5).

3.3 Raster datasets

The spatial modeling techniques required the use of various datalayers (Table 1). Two digital Quaternary geology maps were used to target fine-grained sediments in which a.s. soils are most likely to occur: one map at scale 1:20,000 for Papers **I**, **III**, and **IV**, and one at scale 1:200,000 for Paper **II**. Since a.s. soils are generally located in low-relief areas (i.e. plains, marshes and river valleys), simple slope models were created from DEMs in order to distinguish these soils from till formations and bedrock outcrops (using a 3 x 3 cell neighbourhood computation method; Burrough and McDonell, 1998). A basic low-resolution DEM with an original cell size of 25 m x 25 m and elevation data accuracy of 2 m was used for Papers **I**, **II** and **IV**. Airborne laser scanning (i.e. Light Detection And Ranging, LiDAR) data was available for Paper **III** and enabled the creation of a DEM with a cell size of 2 m x 2 m and an elevation data accuracy of 0.3 m.

The low electromagnetic frequency (3 kHz) imaginary and real components were derived from high resolution low altitude airborne geophysics (flight altitude from 30 to 40 m and line spacing mainly 200 m; Airo et al., 2014). The imaginary component enables the detection of shallow anomalies which are mainly related to variations in top-soil thickness and/or electric conductivity. The real component indicates anomalies originating deep in the bedrock (down to several tens of meters), for instance from black schists. Sulfide-bearing sediments are expected to yield strong electromagnetic anomalies due to their high contents of soluble salts (Vanhala et al., 2004; Suppala et al., 2005). They appear as diffuse and round-shaped high electric conductivity anomalies. Black schists are often associated with sulfide deposits and might also cause high metal contents in soil or ground water (Airo and Loukola-Ruskeeniemi, 2004). They appear as thin and elongated anomalies. When black schists occurred in one of our study areas, they were taken into account within the interpretation of the modeling results (Papers **II** and **IV**). Using both components is useful as sulfidic materials can occur relatively deep and be covered by coarse, not sulfidic sediments (Österholm et al., 2010).

The Quaternary geology and aerogeophysical datalayers were produced by GTK, and the DEMs by the National Land Survey (MML). Figure 2 displays the different original datalayers for the Sirppujoki River catchment.

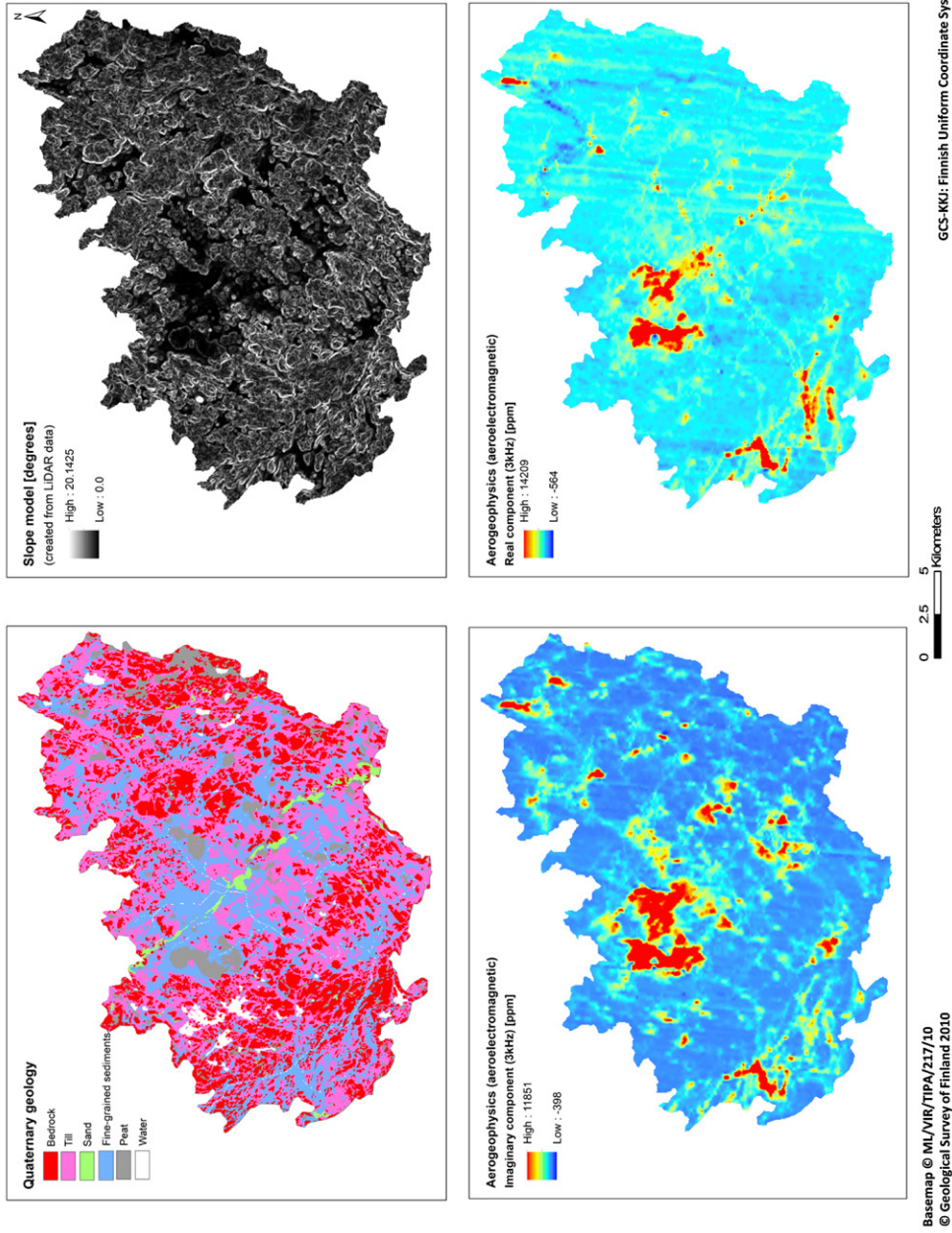


Figure 2. The original evidential datalayers for the Sirppujoki River catchment: Quaternary geology, imaginary component, real component and slope derived from a LiDAR-based DEM.

4 Methods

Most of the coastal areas were assessed at reconnaissance scale using fuzzy logic: the southern part of the coastal areas (c. 17,300 km²; Paper II) and 13 catchments located along the Finnish coast (c. 9,500 km², in all; Paper IV). 10 catchments have been conventionally mapped (c. 11,400 km², in all; Paper IV). Based on ease of access and sufficiency of data coverage, ANN techniques were applied to the Sirppujoki River catchment (c. 440 km², located in southwestern Finland; Papers I, III and IV). An overview of the methods utilized in this study is presented in Table 1. The final probability and predictive maps have a 50 m x 50 m cell size.

4.1 Conventional mapping

The conventional mapping process can be divided into three steps. Prior to the soil sampling in the field, various datalayers (aerogeophysical data, bedrock geology, Quaternary geology, peatland and elevation data) are utilized to prioritize areas of interest (Papers IV; Österholm et al., 2010; Edén et al., 2012b). A soil sampling plan covering all potential areas is established. Secondly, soil sampling is carried out, pH is measured in the field and after 9 to 16 weeks of incubation, and selected samples are subsequently analyzed for sulfur (cf. the soil samples sub-section of the material). Finally, the a.s. soil maps are drawn at 1:125,000 scale based on the combined interpretation of the following data (Paper IV): field observations, measurements and further analyses, topographic maps (i.e. DEMs provided by MML), Quaternary geology, peatland and aerogeophysical data (produced by GTK).

4.2 Spatial modeling techniques

The use of GIS-based spatial modeling techniques has rapidly increased in geosciences over the last two decades. These techniques are based on geostatistics which provide a set of mathematical tools to characterize spatial patterns (Goovaerts, 1999), generate maps from point observations and model the associated uncertainty (Hengl, 2009). Basically, the methods can be divided into two main categories, data- and knowledge-driven approaches (Bonham-Carter, 1994). The knowledge-driven methods (e.g. fuzzy logic, Dempster-Shafer belief theory or decision-tree approaches) are particularly suitable to predict soil properties in areas with an insufficient number or even no known occurrences, the model parameters being estimated by expert knowledge. The data-driven methods (e.g. weights-of-evidence, ANNs or logistic regression approaches) conversely require a sufficient amount of known occurrences which are used

for model training. Among the variety of spatial modeling methods (McBratney et al., 2003), our selection was made depending on the available data. For this study, fuzzy logic and weights-of-evidence were first applied to areas with few or many known occurrences, respectively. The weights-of-evidence technique utilizes only known a.s. soil occurrences and requires an assumption of conditional independence between the different input evidential datalayers (Bonham-Carter, 1994), which constitutes a rather limiting factor when few datalayers are available. This technique was abandoned in favor of ANNs, alternative data-driven techniques which constitute good pattern recognition and classification tools with the ability to generalize from imprecise input data (Porwal et al., 2003). ANNs also utilize both known a.s. and non-a.s. soil occurrences. Fuzzy logic and ANNs have never been used, to our knowledge, for a.s. soil mapping and characterization. Only one recent study assessed the application of a fuzzy k -means clustering method for a.s. soil mapping (Huang et al., 2014). Fuzzy logic and ANNs have been utilized in various fields of geosciences: mineral prospectivity mapping, groundwater mapping, landslide hazard mapping, acid mine drainage (AMD) processes, geological mapping, as well as soil mapping and soil assessment (references in Table 2).

Table 2. Selection of papers utilizing fuzzy logic or artificial neural networks (ANNs) in various domains of geosciences.

	Fuzzy logic	ANNs
<i>Mineral prospectivity</i>	An et al., 1991	Porwal et al., 2003
	D'Ercole et al., 2000	Behnia, 2007
	Nykänen & Salmirinne, 2007	Nykänen, 2008
	Nykänen et al., 2008	
<i>Groundwater mapping</i>	Nobre et al., 2007	Corsini et al., 2009
<i>Landslide hazard mapping</i>	Kayastha, 2012	Fernández-Steeger et al., 2002
	Tien Bui et al., 2012	Ermini et al., 2004
<i>AMD processes</i>	Aroba et al., 2007	
	Maria Valente & Leal Gomes, 2009	
<i>Geological mapping</i>		Barnett and Williams, 2009
<i>Soil mapping</i>	Zhu et al., 2001	Behrens et al., 2005
	Qi et al., 2006	Cavazzi et al., 2013
	Zhu et al., 2010	Taborda Silveira et al., 2013
<i>Soil assessment</i>	Saboya Jr. et al., 2006	Chang & Islam, 2000
	Kaufmann et al., 2009	Lentzsch et al., 2005
	De Gruijter et al., 2011	Lamorsky et al., 2008 Singh et al., 2012

4.2.1 Fuzzy logic

Fuzzy logic constitutes a conceptual approach for spatial modeling, based on expert knowledge (Bonham-Carter, 1994). This method is generally selected when an insufficient number or even no known occurrences are available to carry out a statistical analysis (i.e. the study area has not been mapped before and a limited amount or even no samples are available for validation). Fuzzy logic differs from ANNs as it does not learn from known data. This method enables dealing with uncertainties inherent in complex natural phenomena (McBratney and Odeh, 1997). This method originates from the fuzzy-set theory, introduced by Zadeh (1965), which defines a degree of membership, a value between 0 and 1 without crisp boundary, for a set. A fuzzy set A in X is composed of ordered pairs $[x, \mu_A(x)]$ and is mathematically defined by the following equation in which $\mu_A(x)$ is the membership value of element x in A (Kandel, 1986):

$$A = \{x; \mu_A(x)\}, x \in X$$

These fuzzy membership values are defined on a continuous scale from 0 to 1, where 0 indicates the full non-membership of x in the fuzzy set A (“not indicator”) and 1 the full membership of x (“indicator”). Different evidential datalayers (in our case, geophysical, geology and slope) are used as input data. Their fuzzification constitutes the first step within the fuzzy logic modeling (Fig. 3). Basically, various fuzzy membership functions are applied to each input datalayer (Bonham-Carter, 1994). The resulting fuzzy sub sets (i.e. the “fuzzified” evidential datalayers) are then combined using fuzzy operators (AND, OR, Algebraic Sum, Algebraic Product and Gamma; An et al., 1991). This overlay process constitutes the integration of the input data into a unique fuzzy set, a probability map (or favorability map) answering the main hypothesis (in our case, is there an occurrence of a.s. soil; Fig. 3). The Fuzzy AND (i.e. minimum operator) results in the logical intersection between two or more sets. The Fuzzy OR (i.e. maximum operator) results in the logical union of two or more sets. The Fuzzy Algebraic Sum represents an increasive operator, its output being always larger than, or equal to, the largest contributing membership value. In contrast, the Fuzzy Algebraic Product is a decreaseive operator, its output being always smaller than, or equal to, the smallest contributing membership value. The Fuzzy Gamma corresponds to the combination of the Algebraic Sum and Algebraic Product operators. The degree of the combination is controlled by the Gamma parameter γ , ranging from 0 to 1 and, thus, fluctuating between increasive and decreaseive effects. For Paper II, the whole fuzzy logic modeling was carried out using ArcGIS 10™ (ESRI software) and a public-domain extension for multivariate analysis called Spatial Data Modeller (SDM) (Sawatzky et al., 2009). The fuzzification

and integration sections of the fuzzy logic modeling process are developed in details in Paper II.

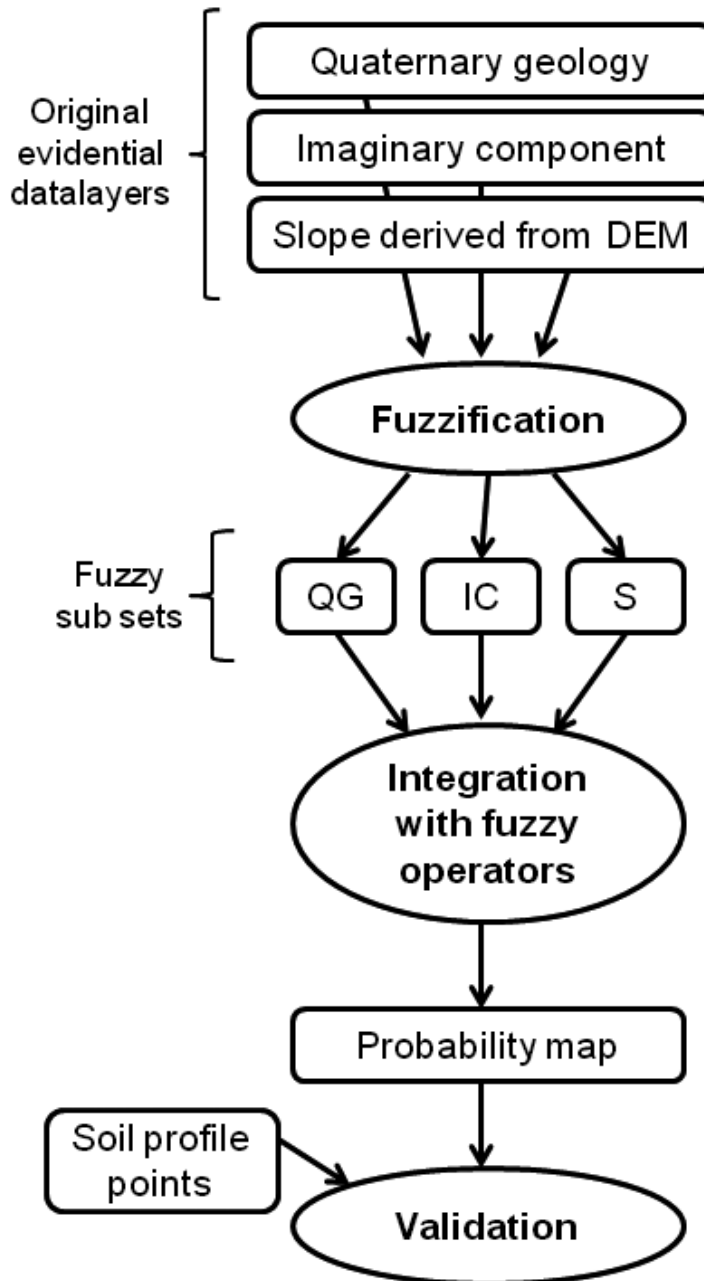


Figure 3. The fuzzy logic modeling steps followed in Paper II.

4.2.2 Artificial neural networks

Artificial neural networks (ANNs) constitute standard machine-learning techniques providing powerful pattern recognition and classification tools. ANNs are basically designed to learn how to classify new, unknown data using known examples for training. They are, thus, considered as data-driven techniques based on a supervised learning algorithm (Bonham-Carter, 1994; Zell et al., 1998; Gershenfeld, 1999). Different evidential datalayers are also used as input data within the modeling (i.e. Quaternary geology, slope, real and imaginary component). The ANN concept and architecture are inspired by biological nervous systems. The artificial neurons are connected and organized in layers. As shown in Figure 4, an artificial neuron basically receives the input signals or variables x (x_1, x_2, \dots, x_n) through synapses which associate the inputs to synaptic weights ($w_{0j}, w_{1j}, w_{2j}, \dots, w_{nj}$). The neuron itself can be schematically characterized by two parts (Tsoukalas and Uhrig, 1997): (1) the first part sums up the weighted inputs, resulting in a value I ; (2) the second part is a non-linear activation function φ applied to the sum of weighted inputs I , resulting in a single output y . The most common activation functions are the sigmoid and the Gaussian functions, which output a number between 0 (for low input values) and 1 (for high input values). Being able to approximate non-linear functions of their inputs, ANNs enable the predictive modeling of complex natural situations involving intricate chemical and physical processes which are not directly observable. They also deal with uncertainty related to input data.

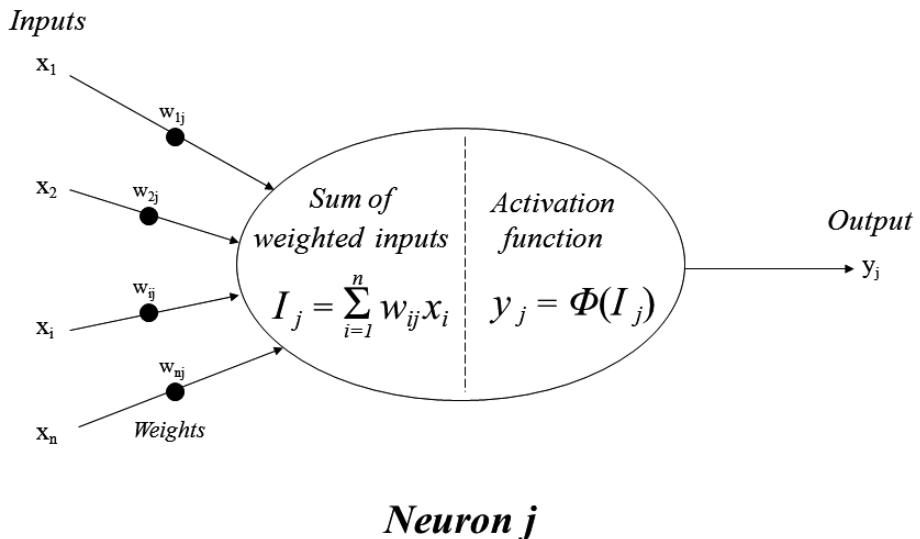


Figure 4. General structure of an artificial neuron according to Tsoukalas and Uhrig (1997).

In this study, two ANNs, both using Radial Basis Functions (RBFs), were assessed on the Sirppujoki River catchment (Papers I and III). The networks based on RBFs present simple training algorithms and architecture, they also train more quickly than multiple layered perceptron (MLP) networks (i.e. a common sort of ANNs; Looney, 2002). In Paper I, a Radial Basis Functional Link Net (RBFLN) was applied. The RBFLN modeling was solely performed using ArcGIS 10 (ESRI software) and a public-domain extension for multivariate analysis called Spatial Data Modeler (SDM; Sawatzky et al., 2009). In Paper III, a simple RBF-based ANN was implemented within R environment, using a package called RSNNS (R Stuttgart Neural Network Simulator; Bergmeir and Benítez, 2012) and in-house routines specifically developed for automatization, data handling and record keeping purposes in this study. The predictive results were visualized within a GIS. This technique was selected, not only as it is comparable to the RBFLN applied to the same study area (Paper I), but also, as it enabled us to access more network modeling parameters (i.e. initialization and learning function parameters), and therefore, automatically and systematically test more models than previously.

Both ANNs are feed-forward networks composed of three layers (Fig. 5; Looney and Yu, 2001): (1) an input layer of N nodes, each node transmitting one input variable (in our case, $N = 4$); (2) a hidden layer of M artificial neurons, each representing a RBF; (3) an output layer of J artificial neurons, receiving a combination of all the single outputs from the hidden neurons in a weighted average (in our case, $J = j = 1$). Before the modeling, the four evidential datalayers are generalized into fewer classes (i.e. the reclassification step for ordered and continuous data). Each input variable corresponds to a class value for one of the four evidential datalayers (i.e. Quaternary geology, slope, real and imaginary component). Combined together, these input variables (x_1, x_2, x_3, x_4) constitute a sole input feature vector x . The input feature vectors are transmitted from the input layer to the hidden layer. Each hidden neuron sums up the weighted inputs and applies the activation function on the sum generating a single output y (Fig. 4). The RBFs are based on a Gaussian density function defined by a center v and a spread parameter σ . The initial centers can indicate the training feature vectors or be randomly selected by the network. The output from the m^{th} neuron of the hidden layer ($m = 1, \dots, M$) for the q^{th} input feature vector $x^{(q)}$ (the combination of the evidential datalayers yield Q feature vectors and $q = 1, \dots, Q$) is (Looney, 2002):

$$y_m^{(q)} = \exp \left[-\frac{\|x^{(q)} - v^{(m)}\|^2}{2\sigma_m^2} \right] \quad (1)$$

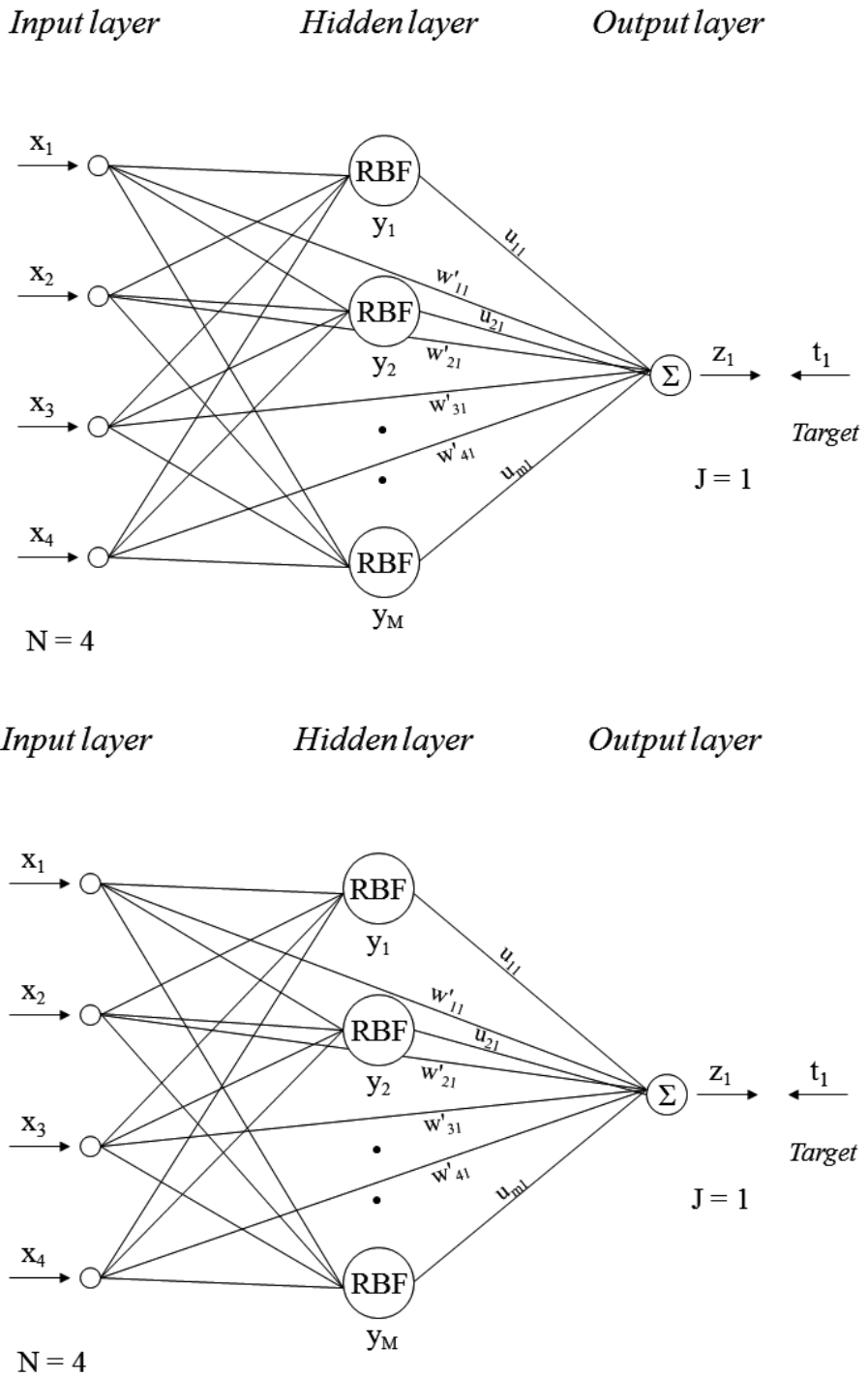


Figure 5. General architecture of a Radial Basis Function Link Net (RBFLN; top) and a Radial Basis Function (RBF)-based artificial neural network (bottom) (Looney, 2002).

The outputs y (y_1, y_2, \dots, y_m) are then transmitted to the output layer neuron, each value of y being multiplied by synaptic weights u_{mj} which can be seen as learned gains from the hidden layer to the output layer (Looney, 2002). In the RBF-based ANN, the output returned by the output neuron is (Looney, 2002):

$$z_j^{(q)} = (1/M) [\sum_{(m=1,M)} u_{mj} y_m^{(q)} + b_j] \quad \leftarrow t_j^{(q)} \quad (2)$$

In equation (2), j is 1 and b_j is the bias included at each output neuron corresponding to the final difference between the output z_j and the target t_j (Looney, 2002). In an RBFLN, the output neuron is also directly connected to the input nodes (Fig. 5). Thus, it receives the inputs x (x_1, x_2, x_3, x_4) multiplied by another set of synaptic weights w'_{nj} . These additional connections constitute the main difference, architecture-wise, between the RBFLN and the RBF-based ANN (Fig. 5). Therefore, in an RBFLN, the output returned by the output neuron is (Porwal et al., 2003):

$$z_j^{(q)} = [1/(M+N)] \{ \sum_{(m=1,M)} u_{mj} y_m^{(q)} + \sum_{(n=1,N)} w'_{nj} x_n^{(q)} + b_j \} \quad \leftarrow t_j^{(q)} \quad (3)$$

Basically, the application of an ANN consists of two stages: training and classification. During the training stage, the network receives the combinations of input variables corresponding to known examples (i.e. the training input feature vectors): a.s. and non-a.s. soil occurrences are called positive and negative training points, respectively. Each training input feature vector is associated to a target value, which can be seen as a probability value: 1 for a positive point and 0 for a negative point. At each iteration, the learning algorithm automatically adjusts the synaptic weights so that the network assigns an output z as close as possible to the known target t for each training input feature vector x . During the classification stage, the network receives all the combinations of input variables existing for the whole study area and classifies them using the learned, calibrated weights. Finally, each input feature vector is associated to a predicted output, a probability value which is calculated by progressive iterations within a self-organizing structure. Looney (2002) states the advantages of an RBFLN: (1) it includes both non-linear functions (through the Gaussian activation function) and linear connections between the input and output layers, and (2) a smaller number of hidden neurons can be used. As summarized by Looney (2002), the RBFLN contains the RBF-based ANN as its non-linear part. Nevertheless, only two parameters, the number of hidden neurons and the number of iterations, could be adjusted within the RBFLN modeling. Within the RBF-based ANN modeling, parameters for the initialization and learning functions could also be tuned. The pre-processing, integration and post-processing sections of the ANN modeling processes are developed in details in Papers **I** and **III**.

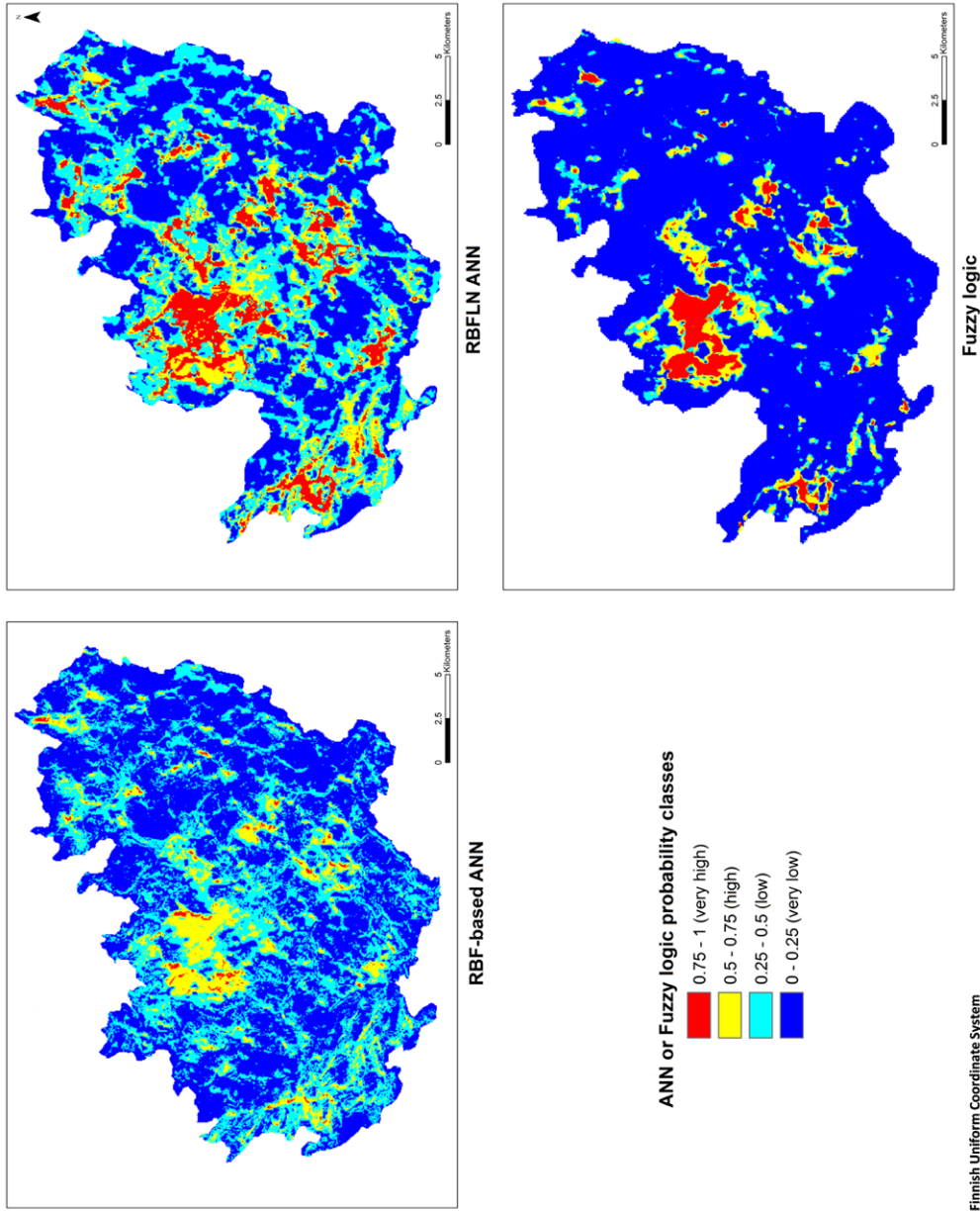
5 Results and discussion

5.1 Fuzzy logic for preliminary surveys at regional or catchment scale

Fuzzy logic was first applied to the southern part of the Finnish coastal areas (c. 17,300 km²; Paper II). This method was particularly suitable to this large study area for which different evidential datalayers and relatively few point data were available ($n = 72$, after a random selection of 1 point per 2 km²). The most accurate model allocated 74% of the known a.s. soil occurrences in the most probable areas (covering c. 12% of the whole study area). These results indicated that fuzzy logic, when used for a preliminary survey, can provide the basis for objective prioritization of the most probable areas for a.s. soil occurrence, in our case, at regional scale. Still, fuzzy logic tends to overestimate the very low probability areas (Paper II), as is particularly evident when comparing the probability maps created with fuzzy logic or ANNs for the Sirppujoki River catchment (Fig. 6; Paper III).

In Paper IV, several catchments along the Finnish coast (in all, c. 9,500 km²) were modeled with fuzzy logic, before conventional mapping was carried out (no available soil profiles to use for validation) or before the completion of the conventional mapping process (soil profiles subsequently available for validation). For Simojoki, Lestijoki and Kalajoki River catchments (belonging to the latter category), validation points corresponding to a.s. soil occurrences were relatively poorly assigned (56, 59 and 47%; Paper IV), indicating an underestimation of the most probable a.s. soil areas. In contrast to conventional a.s. soils (i.e. fine-grained sediments, sometimes located under peat deposits), it was found that, in these catchments, a.s. soils often occur in relatively coarse-grained sediments (Boman et al., 2014). Since the models were adapted to conventional fine-grained a.s. soils, these unusual soil types were allocated low fuzzy membership values and, thus, could not appear as high probability areas. This emphasizes not only the importance of expert knowledge within fuzzy logic, but also the fact that preliminary surveys should lead to more refined models.

Fuzzy logic constitutes a rather simple, fast and objective alternative to delimit and prioritize the most probable areas for a.s. soil occurrence at reconnaissance scale, on its own or prior to conventional mapping. This method offers preliminary synthesized information which can be particularly useful in the field for further soil investigation. Being easily transferable from area to area, it can also be carried out on a large scale, which would be extremely time-consuming through manual assessment.



GCS-KKI: Finnish Uniform Coordinate System

Figure 6. The acid sulfate soil probability maps created for the Sirppujoki River catchment with different approaches.

5.2 Artificial neural networks for mapping at catchment scale

In contrast to fuzzy logic, ANNs offer a more precise modeling of the occurrence of a.s. soils at catchment scale, but require a sufficient amount of point datasets for model training and validation. Two different ANNs were applied to the Sirppujoki River catchment for a.s. soil probability mapping: an RBFLN (Paper I) and an RBF-based ANN (Paper III; Fig. 6). As empirical, data-driven methods, ANNs were notably adapted to this study area for which different evidential datalayers and point datasets (both known a.s and non-a.s. soil occurrences) were available. In Paper I, the RBFLN was applied to a slightly smaller study area (400 km² vs. 440 km² in Paper III) and low-resolution elevation data was utilized as LiDAR data was not yet available when the study was conducted. Similar validation point sets were utilized for both ANNs. The most accurate RBFLN model classified 94% of the known a.s. soil occurrences used for validation in the most probable areas (covering c. 93 km², i.e. 23% of the study area; Table 3), while the most accurate RBF-based ANN model classified all the known a.s. soil occurrences in the most probable areas (covering c. 42 km², i.e. 10% of the study area; Table 3). For both models, the least probable areas (i.e. the low and very low probability areas) contained all the non-a.s. soil occurrences used for validation (Table 3). These results showed that the RBLN model most likely overestimated the most probable areas in comparison to the RBF-based ANN model (Fig. 6; Paper III).

The two ANN properly classified the available unknown a.s. or non-a.s. soil occurrences, but the RBF-based ANN achieved slightly better results validation-wise and excluded more uninteresting areas in terms of total area (c. 90% vs. 77%; Table 3). Furthermore, these results are more consistent with previous studies carried out in the Sirppujoki River catchment. Soil studies (Palko et al., 1985; Triipponen, 1997) recorded the occurrence of a.s. soils in the study area, but used limited data (soil profiles investigated down to 200 cm depth only) and, by today's standards, inappropriate criteria for classification (subsoil pH < 5.0). Nevertheless, all the a.s. soil areas defined by these studies concurred with some of our most probable a.s. soil areas (even if not covering them all). Water studies confirmed the occurrence of a.s. soils, discovering corresponding spatial chemical patterns (high acidity and concentrations of sulfate and dissolved metals) in the recipient waters (Nyberg et al., 2011; Nystrand et al., 2012). Therefore, the RBF-based ANN method appears to narrow down more accurately the most probable areas for a.s. soil occurrence, but also to define more properly the least probable areas than the RBFLN method.

Table 3. Probability class extent and validation point distribution for different acid sulfate soil probability models applied to the Sirppujoki River catchment (from Papers I, II and III).

Model	Probability area	Area		Validation point distribution (%)	
		(km ²)	(%)	positive	negative
RBF-based ANNs					
with LiDAR data	Very high [0.75-1]	2.7	0.6	50	0
	High [0.5-0.75]	39.0	8.9	50	0
	Low [0.25-0.5]	157.0	35.9	0	50
	Very low [0-0.25]	239.0	54.6	0	50
with low-resolution elevation data	Very high [0.75-1]	5	1.1	57	0
	High [0.5-0.75]	48	11.0	43	6
	Low [0.25-0.5]	144	32.9	0	19
	Very low [0-0.25]	240.7	55.0	0	75
RBFLN ANN					
	Very high [0.75-1]	35.9	8.9	47	0
	High [0.5-0.75]	57.5	14.2	47	0
	Low [0.25-0.5]	133.6	33.1	6	33
	Very low [0-0.25]	176.5	43.7	0	67
Fuzzy logic					
	Very high [0.75-1]	16.4	4.1	16	14
	High [0.5-0.75]	31.6	7.8	16	14
	Low [0.25-0.5]	28.7	7.1	36	43
	Very low [0-0.25]	326.9	81.0	32	29

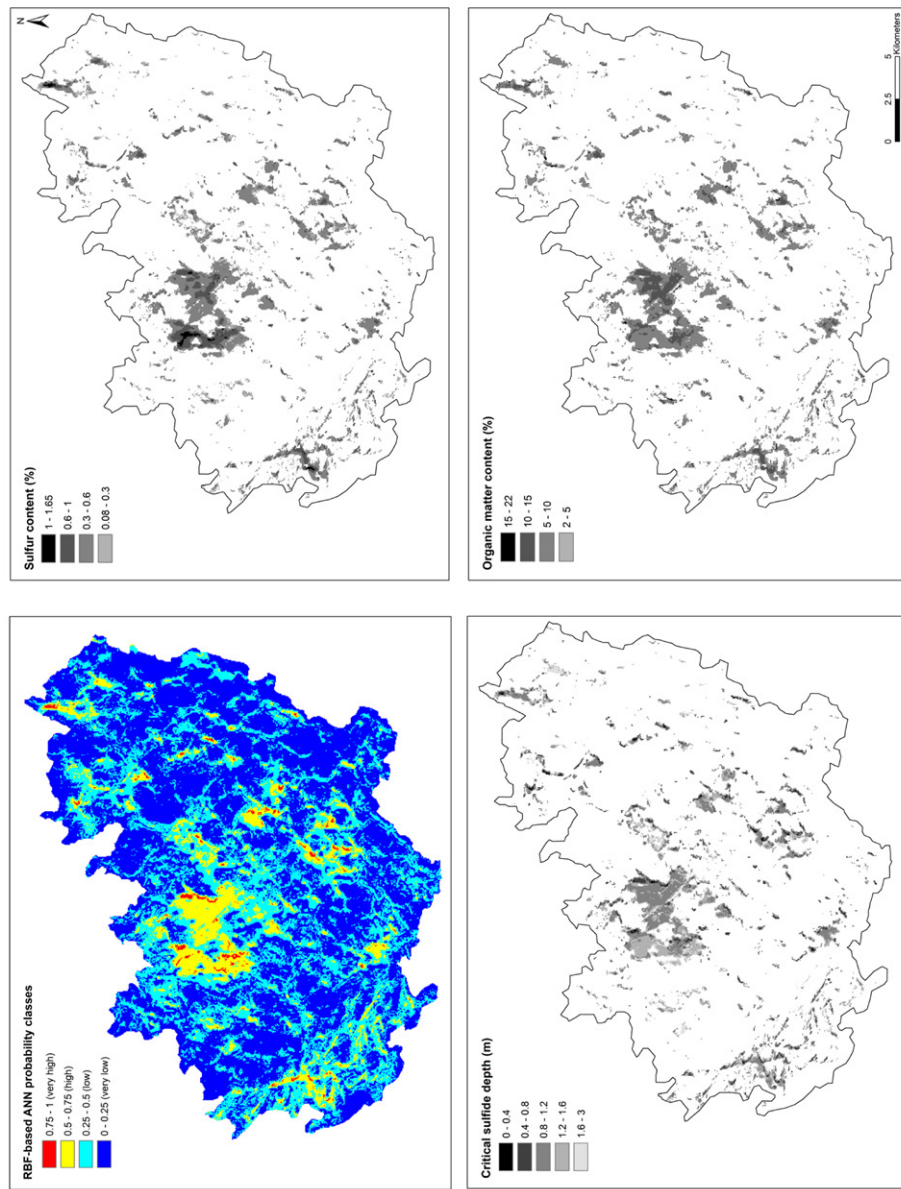
NB: One RBF-based ANN model was created using LiDAR data while the three other models were generated using low-resolution elevation data. The three ANN models were applied at catchment scale (c. 440 km² for the RBF-based ANNs and c. 400 km² for the RBFLN ANN) and using the same training and validation point sets, while the fuzzy logic model was carried out on a much larger scale (17,300 km²) and a different validation point set.

5.3 Artificial neural networks for soil characterization at catchment scale

The localization of a.s. soils constitutes a very first step in their management process, enabling us to prioritize areas of interest which need to be further characterized. In Paper III, the RBF-based ANN method was assessed for characterizing the properties of the a.s. soils in the previously defined most probable areas for a.s. soil occurrence (Fig. 7). Soil property modeling was carried out with ANNs before (Chang and Islam, 2000; Lamorsky et al., 2008), but not for a.s. soils to the author's knowledge. Three different soil properties which are relevant for the mitigation of a.s. soils were modeled: the sulfur content, the organic matter content and the critical sulfide depth. These soil properties were modeled for the whole study area (Table 4), but since the focus was on the areas of interest from a land-use perspective, the resulting predictive maps only displayed the areas concurring with the most probable areas for a.s. soil occurrence (the high and very high probability areas; Fig. 7).

The sulfur content constitutes a crucial factor directly related to acidity load and strongly affecting the metal leaching (Österholm and Åström, 2004; Österholm, 2005). The sulfur content modeled in this study is representative of the sulfides stored in the anoxic parent sediment layers. In 82% of the most probable areas (0.3-1%; Table 4), it was consistent with the typical total sulfur content in the Finnish sulfidic soil horizons (0.2-1.0% and median: 0.54%, for fine-grained sediments in western Finland; Åström and Björklund, 1997). It is notable that all the areas corresponding to the 1 to 1.65% sulfur content class are concurring with the most probable areas for a.s. soil occurrence, indicating that they represent a significant sulfide reservoir with the highest potential to release acidity and metals.

Organic matter represents a relevant factor for various reasons. First, sulfur accumulation during sedimentation is most intensive in organic-rich environments (Toivonen and Österholm, 2011). Second, organic material may hamper the oxidation and subsequent development of acidity in the soil. Third, high contents of organic matter may indicate that sulfur occurs mainly as organic sulfur which does not generate acidity (Österholm and Åström, 2002). The modeled organic matter content value is representative of the potentially oxidised sediments located below the plough layer and the potential peat layers. In 98% of the most probable areas, the modeled organic matter content (LOI) ranged between 5 and 15% (Table 4). These results were also consistent with the modeled sulfur content, indicating that significant sulfur contents (higher than 0.3%) are often associated with notable organic matter contents (higher than 5%; Fig. 7).



GCS-HKI: Finnish Uniform Coordinate System

Figure 7. Predictive soil property maps for organic matter (LOI), sulfur and critical sulfide depth (displaying the most probable areas for a.s. soil occurrence).

The critical sulfide depth constitutes the most pertinent factor from the point of view of management by different drainage techniques. It indicates the level the sulfide reservoir and, thus, how deep drainage can be carried out without mobilizing acidity from it. Taking into account the critical sulfide depth in addition to the sulfur content in the most probable areas for a.s. soil occurrence may enable better mitigation (e.g. for field drainage or construction of roads and buildings). Furthermore, a.s. soil areas generally contain high contents of organic matter, making them prone to compaction. Drainage accelerating compaction which in turn leads to the need for deeper drainage, the importance of critical sulfide depth for drainage management is particularly emphasized. In this study, the modeled critical sulfide depth basically represents the depth to the sulfidic soil material (defined by a field pH higher than 4.5 and an incubation pH lower than 4.0). The critical sulfide depth is comprised between 0.8 and 1.6 m in about 77% of the most probable areas, and is larger than 1.6 m in only 8%. As seen in Figure 7, shallow critical sulfide depths (smaller than 0.8 m, covering 15% of the most probable areas) are relatively common in very high a.s. soil probability areas. The very high probability areas where these shallow critical sulfide depths concur with high sulfur contents (c. 15% of the Sirppujoki River catchment) should be considered as the most critical areas in which any land-use project should be planned carefully (Table 4).

The modeling of soil properties (i.e. their relative spatial distribution in the areas of interest) represents a novel and promising development for a.s. soil mapping and characterization. Since a.s. soil areas constitute highly productive lands for agricultural purpose, the association of an a.s. soil probability map with more specific soil property predictive maps represents a greatly valuable toolset to target strategic areas for subsequent implementation of environmentally sustainable, “low impact” management practices (e.g. controlling drainage and minimizing drainage depth; Österholm and Åström, 2008).

Table 4. Predictive class extent and validation point distribution for the most accurate soil property models in the Sirppujoki River catchment.

Soil property	Predictive classes	Study area		Validation point distribution (n)		Areas concurring with high and very high a.s. soil probability areas	
		(km ²)	(%)	predicted	actual	(km ²)	(%)
Sulfur content (%)	[1 - 1.65]	0.8	0.2	1	1	0.8	1.9
	[0.6 - 1]	49.2	11.3	1	2	9	21.6
	[0.3 - 0.6]	194.3	44.5	9	0	25	60.0
	[0 - 0.3]	192.3	44.0	4	12	6.9	16.5
Organic matter content (%)	[15 - 22]	19.2	4.4	0	1	0.4	1.0
	[10 - 15]	73.6	16.9	0	0	6.1	14.6
	[5 - 10]	250.1	57.3	13	5	34.9	83.7
	[0 - 5]	93.6	21.4	2	9	0.3	0.7
Critical sulfide depth (m)	[0 - 0.4]	-	-	-	-	0.4	1
	[0.4 - 0.8]	-	-	-	-	6	14.4
	[0.8 - 1.2]	-	-	-	-	20	48
	[1.2 - 1.6]	-	-	-	-	12	28.8
	[1.6 - 3]	-	-	-	-	3.3	7.9

NB: The different soil property models were built for the whole study area (using LiDAR data), but are only relevant for the areas concurring with the high and very high a.s. soil probability zones. Furthermore, the critical sulfide depth is only pertinent for these most probable areas and the results are displayed accordingly.

5.4 Impact and limitations of the different input evidential datalayers

The availability of different input evidential datalayers is crucial and may constitute a limiting factor within the modeling (whether for fuzzy logic or ANNs). The availability of LiDAR data for the Sirppujoki River catchment enabled us to compare models created using it to models generated with conventional low-resolution elevation data. LiDAR data allowed a more precise definition of the slope. The slope layer derived from a DEM created using LiDAR data, thus, distinguished in more details the flat/low-relief areas from the uneven/high-relief areas. Furthermore, the models created using LiDAR data delimited more precisely the most and least probable areas for a.s. soil occurrence in comparison with previous studies using low-resolution elevation data (Table 3; Paper III).

Considering the different input datalayers, several issues might be encountered. Continuous data such as the aerogeophysical datalayer represented a combination of sectors surveyed during different airborne campaigns with various years of production and furthermore diverse characteristics. Consequently, normalization (i.e. homogenization) of these different sectors constituted a critical aspect in large-scale studies. Considering categorical datalayers, the resolution or level of information set the limit. In glaciated terrains, detailed Quaternary geology datalayers, such as the one available for Finland, provided a valuable tool for excluding areas dominated by moraine and coarse-grained glaciofluvial material in the upper soil horizons. It did not, however, distinguish glacial from Holocene clays, or identify locally occurring Holocene clays underlain by re-deposited glaciofluvial material (Österholm et al., 2010). High and very high probability areas could be found close to the highest shoreline of the former Litorina Sea, showing that sulfides can form at very shallow water depths in estuaries, with a supply of organic matter and a high sedimentation rate. Significant sulfide formation in estuarine shallow waters of the present Baltic Sea has also been found by Nordmyr et al. (2007), while Fonselius (1970) demonstrated the formation of hydrogen sulfide in the bottom water of deep basins. Moreover, assessing the areas located above the highest shoreline of the former Litorina Sea would constitute an interesting development for a.s. soil mapping. Another possible limitation came from the resolution of the DEM from which the slope layer was derived. The increased availability of LiDAR data will, however, allow this limitation to be overcome worldwide.

The modeling, whether fuzzy logic or ANN, would greatly benefit from the use of additional evidential datalayers (e.g. land use data or additional terrain attributes derived from a DEM). The final a.s. soil probability maps represent

both actual and potential a.s. soils since the evidential datalayers used within the modeling can indicate both. Moreover, the a.s. soil areas modeled with ANN reflect the occurrence of actual and potential a.s. soils as a whole since both were used as training and/or validation points (within the fuzzy logic modeling, they were used only for validation). Since potential a.s. soils may occur below peat layers in Finland, these deposits were ranked just below fined-grained sediments (i.e. gyttja, clay, very fine and fine silts) within the reclassified Quaternary geology datalayers (Table 2 in Paper I and Table 1 in Paper III). Therefore, the resulting models take into account the possible occurrence of a.s. soils below peat deposits. Nevertheless, comparing the modeled a.s. soil areas with land use data may confirm the location of some potential a.s. soil areas under peat layers. As potential a.s. soils can also be located in arable land areas under layers of actual a.s. soils, the use of land use data cannot enable a complete distinction between actual and potential a.s. soils. Additional terrain attributes derived from a DEM, such as the topographic wetness index, might also be utilized for a further mitigation-oriented characterization of a.s. soils.

5.5 Validation as a critical modeling step

Within the modeling process, the validation constitutes a crucial step, whatever the method utilized. The use of validation points represents a simple and effective approach for evaluating the spatial distribution of a.s. soils. The validation of a fuzzy logic model can be particularly challenging as conceptual models are typically used when no or very few known sites are available to be used as validation points (Nykänen et al., 2008). Nevertheless, validation points corresponding to known occurrences (i.e. positive points) are commonly used (D'Ercole et al., 2000; Nykänen et al., 2008; Nykänen and Salmirinne, 2007; Porwal et al., 2003; Zhu et al., 2001). In Paper II, a certain number of known non-a.s. soil sites (i.e. negative points) were available in addition to the known a.s. soil occurrences (i.e. positive points). As the soil profiles constituting the validation points were mainly taken from farmland areas, the negative points were not representative enough of typical non-a.s. soil types (e.g. till). Moreover, the Quaternary geology map located some of the positive points in bedrock or till areas (6 and 8%, respectively), plainly demonstrating the limitation of this map. Since most of the negative points were located in clay areas (75%), properly assigning them to low and very low probability classes constituted a challenging task for the fuzzy logic model. Therefore, considering the study area as a whole, the validation results can be seen as underestimating the accuracy for the low and very low probability classes (Paper II).

Considering the validation results for the most accurate soil property models, the modeling precision appeared as fairly satisfactory with 73% of properly-classified validation points for both the sulfur and organic matter contents, and 67% for the critical sulfide depth (Table 4). Nevertheless, considering the validation point distribution, the predicted values, especially for the sulfur and organic matter contents, still seemed to be overestimated in comparison with the actual values. For the sulfur content, most of the actual values for the validation points were in the 0 to 0.3% class, suggesting that the areas corresponding to this class should have a larger extent (Table 4). Likewise, for the organic matter content, the actual values for the validation points suggested a larger extent for the areas corresponding to the 0 to 5% class (Table 4). Thus, in further studies using the sulfur and organic matter content predictive maps, the predicted values could be adjusted (i.e. multiplied with a correction factor), so that their distribution would be more in line with the distribution of the actual values for validation points. Nevertheless, the principal goal of this modeling remains to define the relative spatial distribution of the soil properties in the areas of interest (i.e. the most probable areas for a.s. soil occurrence).

5.6 Assessment of acid sulfate soil mapping utilizing chemical indicators in recipient waters

While validation points enable a simple and effective evaluation of a.s. soil spatial distribution, the use of water chemistry data constitutes a more suitable approach for assessing probability maps in regard to the extent of a.s. soils. Moreover, as the environmental issue due to a.s. soils was first discovered through the ecological damage affecting the recipient waters, using water chemistry data also represents a logical way to validate a.s. soil probability maps. In Paper IV, we assessed a.s. soil probability maps, created with different methods (i.e. conventional mapping, fuzzy logic and ANN), by comparing the extents of a.s. soils calculated for various catchments along the Finnish coast to water chemistry data in the corresponding rivers. Among the available water chemistry data, we focused on two of the most important variables constituting a.s. soil related chemical indicators in water: the sulfate content and the sulfate/chloride ratio. The sulfate content in recipient waters constitutes the main indicator of a.s. soils as it is directly related to sulfide oxidation. Ideally, the amount of sulfur released by a.s. soils to the recipient streams is proportional to their extent in the catchment. The sulfate/chloride ratio enables distinguishing sulfate anomalies related to sulfide oxidation from those related to evapotranspiration, dilution, evaporites, sea spray or sea water intrusion. Regardless of the latter processes, the ratio is expected to increase due to sulfide oxidation and the presence of a.s. soils in a catchment. In Paper

IV, we notably followed the denomination used for conventional mapping by GTK, the very high and high probability classes (as used in Papers I, II and III) being referred as high and moderate probability areas, respectively.

The sulfate content of the studied rivers correlates with the proportion of areas below the highest shoreline of the former Litorina Sea in the corresponding catchments ($r_s = 0.76$; Paper IV), confirming that these areas constitute the potential areas for a.s. soil occurrence in Finland. Nonetheless, as several surveyed catchments extend far above the former Litorina-Sea limit, the measured sulfate content is representative of the total drainage area and a potential dilution effect has to be considered. The extents of the most probable areas for a.s. soils were, thus, calculated taking into account the total drainage area (Table 5). It is still notable that a relatively small proportion of a.s. soils can affect streams with substantial fluxes of sulfate and metals even though the concentrations of these elements are low in the water because of dilution.

The high sulfate contents measured in most of the coastal rivers, together with the lack of a coherent trend with increasing sulfate contents from north to south, demonstrate the importance of the presence of a.s. soils in the corresponding catchments (Table 5). The sulfate/chloride ratio is mostly coherent with the sulfate content in the different rivers (i.e. the ratio is generally increasing with an increasing sulfate content), indicating that sulfide oxidation is the main cause of elevated sulfate concentrations. The calculated extents of a.s. soils correlate well with the sulfate content and the sulfate/chloride ratio in the different rivers (Table 5 and fig. 8), suggesting that the a.s. soil probability maps created with different methods are reliable and comparable. The sulfate content correlates best with the extent of the combined high and moderate probability areas ($r_s = 0.86$; Fig. 8), indicating that the moderate probability areas are important and should be taken into account. Furthermore, the correlation between the sulfate content or the sulfate/chloride ratio to the extents of a.s. soils in catchments modeled with fuzzy logic or created with conventional mapping techniques do not differ markedly (Fig. 9).

Nevertheless, some discrepancies could be observed for some catchments and between methods, leading to over- or underestimation of the extent. This may be explained by different reasons: variations in the sulfur content in different a.s. soils (i.e. inherent sulfate content or rate of depletion), variations in the activity/drainage of the a.s. soils (i.e. rate of oxidation), as well as the occurrence of black schists in the catchment (i.e. causing high proportion of sulfate unrelated to the former Litorina Sea). It is notable that some areas mapped in this study not only include actual a.s. soils, but also potential a.s. soils, and thus could release much larger amounts of sulfate. On the other

Table 5. Extent of the acid sulfate soil probability areas in surveyed catchments along the Finnish coast and water chemistry data in the corresponding rivers (Paper IV).

Area	Catchment	Method	Total drainage area (km ²)	Extent of a.s. soil probability areas			Water data					SO ₄ /Cl Average	Samples (n)	
				High (km ²)	High and moderate (km ²)	(%)	Min.	10 th perc.	Med.	90 th perc.	Max.			
NW	Kemijoki	FL	51130	370.0	480.0	0.7	0.9	1.4	1.5	1.8	2.2	3.0	2.7	10
	Simojoki	FL	3160	3.0	5.5	0.1	0.2	1.1	1.2	1.4	1.9	2.7	1.9	10
	Kuivajoki	FL	1360	22.0	24.0	1.6	1.8	1.4	1.6	1.9	3.0	4.3	2.1	10
W	Oulujoki	FL	22840	157.0	187.0	0.7	0.8	3.0	3.0	4.1	5.9	8.0	3.2	10
	Temmesjoki	FL	1180	550.0	557.0	46.6	47.2	23.0	-	42.0	-	92.0	4.3	3
	Siikajoki	Conv	4320	96.0	246.0	2.2	5.7	6.4	7.0	9.4	13.2	15.0	4.1	8
	Pyhäjoki	Conv	3710	24.0	63.0	0.6	1.7	12.0	12.9	21.0	32.2	34.0	5.1	10
	Kalajoki	FL	4250	17.0	116.0	0.4	2.7	8.1	8.6	14.0	18.0	19.0	3.2	11
	Lestijoki	FL	1370	29.0	48.0	2.1	3.5	4.1	4.7	6.0	11.0	11.0	2.6	9
	Perhonjoki	FL	2520	77.0	122.0	3.1	4.8	5.2	6.4	10.0	15.0	16.0	4.0	11
	Kruunupyynjoki	Conv	790	67.0	112.0	8.5	14.2	20.0	-	37.5	-	41.0	8.7	4
	Ähtävänjoki	Conv	2050	96.0	168.0	4.7	8.2	12.0	13.4	22.5	33.1	38.0	4.6	6
	Lapuanjoki	Conv	4120	283.0	636.0	6.9	15.4	25.0	32.2	37.0	48.5	53.0	5.4	10
SW	Kyrönjoki	Conv	4920	580.0	784.0	11.8	15.9	30.0	33.0	39.0	49.0	53.0	5.4	32
	Närpiönjoki	Conv	990	113.0	272.0	11.4	27.4	32.0	-	33.0	-	36.0	5.2	3
	Sirppujoki	RBFLN	400	35.9	93.4	9.0	23.4	89.8	-	94.3	-	98.8	7.9	2
S	Laajoki	Conv	390	1.4	27.0	0.4	6.9	26.9	-	27.0	-	32.9	2.5	3
	Mynäjoki	Conv	290	4.0	20.0	1.4	6.9	18.0	-	21.0	-	27.0	2.5	3
	Sipoonjoki	FL	220	8.6	25.6	3.9	11.6	16.6	-	21.4	-	26.2	2.2	2
S	Mustijoki	FL	780	6.2	19.2	0.8	2.5	10.3	12.0	13.8	16.4	21.6	1.8	11
	Porvoonjoki	Conv	1580	12.0	59.0	0.8	3.7	9.4	11.5	13.7	16.5	17.0	1.4	10
	Koskenkyläjoki	FL	900	29.1	47.5	3.3	5.3	8.8	10.2	11.7	13.5	23.5	2.0	11
	Kymijoki	FL	37160	8.7	46.1	0.0	0.1	8.9	9.2	11.0	15.0	17.0	2.0	42
	Vehkajoki	FL	380	0.8	3.1	0.2	0.8	7.3	-	8.9	-	19.0	2.8	4

NB: S: southern; SW: southwestern; W: western; NW: northwestern; Conv: conventional mapping; FL: fuzzy logic; RBFLN: Radial Basis Functional Link Net.

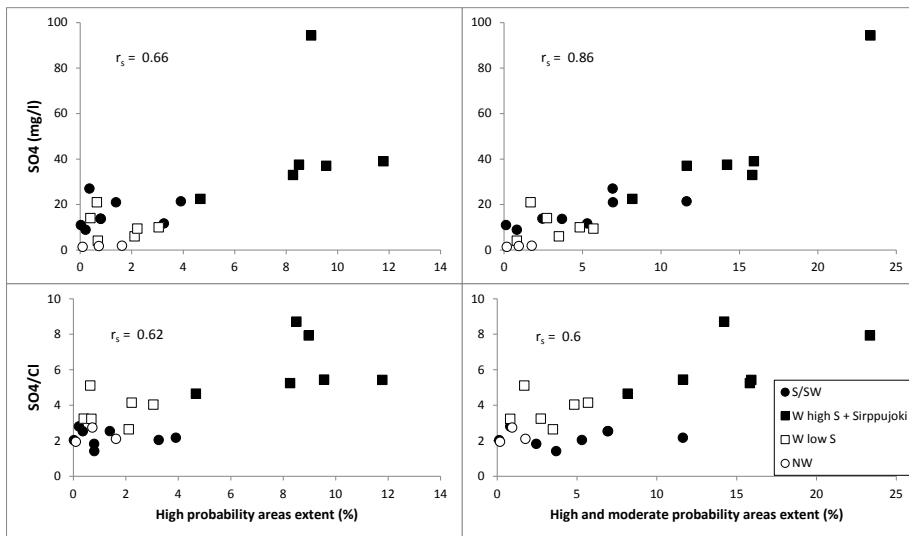


Figure 8. Extent of acid sulfate soils in a catchment (%) versus median sulfate content (mg/l) and sulfate/chloride ratio in corresponding river (S: southern; SW: southwestern; W: western; NW: northwestern; Temmesjoki River catchment is out of range for all graphs).

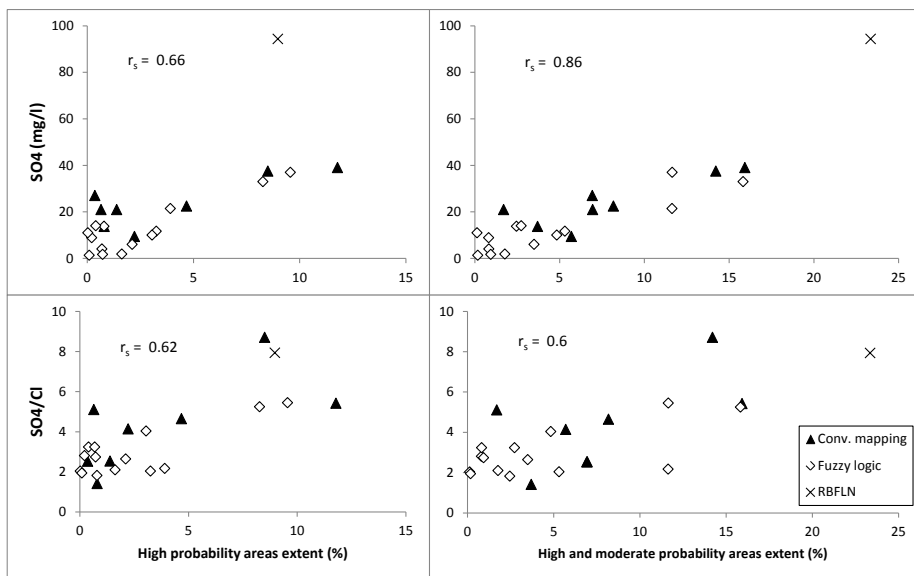


Figure 9. Extent of a.s. soils in a catchment (%) versus median sulfate content (mg/l) and sulfate/chloride ratio in the corresponding rivers, grouped according to the mapping method (S: southern; SW: southwestern; W: western; NW: northwestern; Temmesjoki River catchment is out of range for all graphs).

hand, potential a.s. soils are expected to be relatively scarce, especially in the western and southern parts of Finland, most of the a.s. soils being already drained as they are considered as the most valuable farmlands. This could also explain the case when a catchment displays a large extent value but a relatively low sulfate or sulfate/chloride ratio. Even with a less than perfect estimation of the a.s. soil extent in a catchment, we can still carry out a first assessment of the mapping project.

Considering both the water chemistry data and the calculated extents of a.s. soils, the surveyed catchments could be grouped as follows: (1) the western catchments (i.e. Närpiönjoki, Kyrönjoki, Lapuanjoki, Ähtävänjoki, Kruunupyynjoki, Pyhäjoki and Temmesjoki River catchments), as well as the Sirppujoki River catchment in the southwestern region, with high sulfate contents, high sulfate/chloride ratios and large a.s. soil extents, (2) the remaining western catchments (i.e Perhönjoki, Lestijoki, Siikajoki, Kalajoki and Oulujoki River catchments) with low sulfate contents, high sulfate/chloride ratios and small a.s. soil extents, (3) the southern and southwestern catchments (excluding Sirppujoki River catchment) displaying low and intermediate sulfate contents, respectively, low sulfate/chloride ratios and small a.s. soil extents, and (4) the northwestern catchments displaying very low sulfate contents, low sulfate/chloride ratios and very small extents (Table 5 and Fig. 8).

Therefore, the use of a.s. soil related chemical indicators in water constitutes a complementary independent tool to assess probability maps in regard to the extent of a.s. soils. This assessment also helps in getting an overview of the mapping results, determining regional features, as well as the limits of our maps. Collating soil and water data enables estimating the presence of a.s. soils in different catchments, clearly indicating outlying catchments which would require some corrections or additional soil sampling. The progress of a.s. soils mapping in Finland can, thus, be properly assessed using all available data.

5.7 Estimation of the extent of the Finnish acid sulfate soils

The previous estimations of the extent of the Finnish a.s. soils (c. 3,000 km² according to Puustinen et al., 1994; 600-1,300 km² according to Yli-Halla et al., 1999) were calculated using limited data (soil profiles investigated down to 200 cm depth only, in Puustinen et al., 1994 and Yli-Halla et al., 1999) and, by today's standards, inappropriate criteria for classification (subsoil pH < 5.0 and no sulfur content criterion in Puustinen et al., 1994). For this study, a novel classification system and sampling scheme, more suited for boreal a.s. soils (Edén et al., 2012a), were utilized. Samples were taken down to 300 cm,

not only from cultivated areas, but also from forest and peat-covered areas. They were also incubated, enabling us to identify and take into account potential a.s. soils (Paper I). In Paper II, an estimate of the a.s. soil extent for the southern part of the Finnish coastal areas was assessed using the very high probability class (i.e. with fuzzy membership from 0.75 to 1) from the most accurate fuzzy logic model. The calculated extent was in the order of 800 km² (c. 5% of a total surface of 17,300 km², below the highest shoreline of the former Litorina Sea). In Paper IV, an extent of the a.s. soils for the 24 surveyed catchments was in the order of 2,600 km² (c. 12% of a total surface of 21,300 km², below the highest shoreline of the former Litorina Sea, taking into account the corresponding very high probability areas). This value constituted a rather conservative estimate as it only accounts for the very high probability areas. This extent was much larger than the value calculated for the southern part of the Finnish coastal areas in Paper II. Furthermore, the surveyed areas in this study covering only about 43% of the whole coastal areas, excluding many catchments where a.s. soils can also be expected, the extent of a.s. soils for the whole coastal areas (covering c. 50,100 km², below the highest shoreline of the former Litorina Sea) would obviously be much larger than all the previous estimates.

6 Conclusions

The use of spatial modeling techniques constitute an important development within the acid sulfate (a.s.) soil mapping process as it enables the creation of valid and comparable maps. A spatial modeling technique based on fuzzy logic can efficiently complement conventional mapping as a reconnaissance study to delimit and prioritize hot spot areas, whether at catchment or regional scale. Being both easily transferable and particularly adapted to areas with a limited amount of data available, this method may enable carrying out large scale preliminary surveys in previously poorly defined a.s. soil areas worldwide (in the order of 200,000 km²; Andriessse and Van Mensvoort, 2006). Artificial neural networks (ANNs), like a Radial Basis Functional Link Net (RBFLN) or a Radial Basis Function (RBF) -based ANN, display good predictive classification abilities for a.s. soil probability mapping at catchment scale. The RBF-based ANN method yielded slightly better results than the RBFLN method, narrowing down more accurately the most probable areas for a.s. soil occurrence and defining more properly the least probable areas. The RBF-based ANN also demonstrated promising results for characterization of different soil properties in the most probable a.s. soil areas at catchment scale. The a.s. soil probability map together with more specific soil property predictive maps offers a valuable toolset to target strategic areas for subsequent environmental risk management. The use of laser scanning (LiDAR) data, available in an increasing part of the world, enables a more precise definition of the different a.s. soil probability areas, as well as the soil property modeling classes (for sulfur content and critical sulfide depth).

While a.s. soil mapping represents a crucial step in the management of a.s. soil environmental risks, assessing the reliability of mapping results also constitutes a critical stage, whether using conventional mapping or a spatial modeling method. Validation points enable a simple and effective evaluation of a.s. soil spatial distribution, whereas it was demonstrated that the use of water chemistry data constitutes a complementary, independent and straightforward approach to assess probability maps in regard to the extent of a.s. soils. In this study, the extent of a.s. soils in the surveyed catchments correlated with the two chemical indicators measured in the corresponding rivers, indicating that the probability maps created with different methods were reliable and comparable. In the future, water chemistry data could also be utilized for assessment of a.s. soil mapping at “sub-catchment” scale, which would require sampling water along the main river and/or secondary streams in a catchment. Lastly, the total extent of a.s. soils for the 24 surveyed catchments (covering c. 21,300 km²) was estimated to c. 2,600 km² (i.e. 12% of the area), suggesting that the extent for the whole coastal areas (c. 50,100 km²) would be significantly larger than the previous estimates.

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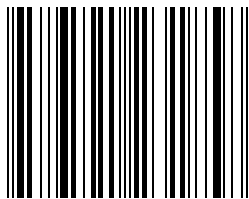
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