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RESEARCH ARTICLE

Decadal trends in soil and grain microelement concentrations indicate mainly favourable development in Finland

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Abstract

Background: Observations on declining nutrient contents in crops have raised concerns about soil fertility and food quality. Long-term monitoring data are valuable in exposing trends and indicating the need for interventions.

Aims: This study aimed to assess the soil microelement status and grain nutritional value on a national scale in Finland over the last decades.

Methods: Using nationwide sets of samples and time-series datasets, temporal trends in readily available zinc, copper, boron, iron, manganese, cadmium, molybdenum, nickel, aluminium, cobalt, and lead in cultivated soils of Finland during 1974–2018 and the corresponding total element concentrations in grains of oats and barley during 1988–2019 were determined.

Results: In soil, initially increasing trends of element concentrations tended to prevalently level off towards the end of the study period. A decreasing trend was observed only for cadmium and zinc in coarse soils. In grains of barley and oat, aluminium and lead concentrations decreased between 1988 and 2019. Barley grains exhibited a decreasing trend in manganese and cobalt, whereas an increasing trend was detected for copper and iron in oats.

Conclusions: No alarming decreases in micronutrient contents in agricultural soils or cereal grains were detected over the study period. In grains, the concentrations of potentially toxic elements decreased. Although the nationwide micronutrient status in Finland is on average satisfactory or good, local micronutrient deficiencies may occur.

KEYWORDS
harmful metals, micronutrients, nutrition, soil quality, soil testing

1 | INTRODUCTION

Chemical analyses of the soil labile element pool have been in routine use for decades in assessing the farm-level fertility status and metal pollution of agricultural soils (McLaughlin et al., 2000; Peck & Soltanpour, 1990). Distinguishing deficient, sufficient, and excessive availability of soil nutrients and harmful elements in relation to crop response serves in ensuring efficient fertilizer use and sustainable maintenance of soil productivity (J. B. Jones, 1998; Sims & Johnson, 1991). Agronomic soil testing tends to focus on macronutrients (nitrogen [N], phosphorus [P], potassium [K]), and pH. Micronutrients, for example, boron (B), cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), molybdenum (Mo), nickel (Ni) and zinc (Zn), and harmful elements, for example, aluminium (Al), cadmium (Cd), and lead (Pb), are less often addressed, even though their impact on yield quantity and quality is likewise decisive (Gupta et al., 2008). In micronutrient-depleted soil, substantial increases in yield can be achieved via micronutrient fertilization (e.g., Kihara et al., 2017; Sahrawat et al., 2010).

Micronutrients are essential for plant growth through their protein-related catalytic, activating, and structural functions (Hänsch & Mendel, 2009). For humans and livestock, plants form a major source of essential micronutrients (Welch & Graham, 2004), but plants may also pass harmful microelements into the food chain (Clemens, 2006). In plants, the acquisition and distribution of microelements is tightly controlled to ensure adequate levels while preventing accumulation in toxic amounts (Giehl et al., 2009; Grusak et al., 1999). This homeostasis comprises up-regulation of uptake under deficiency and compartmentalization in storage in the case of excess nonspecific absorption (Puig & Peñarrubia, 2009). However, plant species vary in their microelement densities, and increasing crop microelement concentrations via agricultural measures is feasible to some extent (Lindström et al., 2013; Rengel et al., 1999). The tendency of decreasing grain micronutrient contents with increasing yield and harvest index makes this nutritionally challenging in modern high-yielding cereal cultivars (Fan et al., 2008; Garvin et al., 2006).

In soil, trends in microelement concentrations reflect the outputs and inputs of a given element. The main inputs arise from mineral fertilizers, animal manures, other soil amendments, and atmospheric deposition, whereas the outputs comprise elements removed within harvested crops and by leaching and erosion. In the case of the labile soil element pool, variations in the soil environment, namely, pH, redox potential, organic matter content, microbial activity, temperature, and moisture, have an impact, as microelements may occur in various species and forms differing in mobility (Fageria et al., 2002; Kabata-Pendias, 2000).

The follow-up of plant-available soil microelement status aims to prevent problems related to the quantity and quality of foods and feeds (Sillanpää, 1982). The reported declines in mineral nutrient contents in cereal grains (e.g., Garvin et al., 2006 [USA], Fan et al., 2008 [UK]) and in vegetables and fruits (Ekholm et al., 2007 [Finland]) have raised concerns about the development of the nutritional value of food. The aims of this study were (1) to determine the mean temporal trends in soil acid ammonium acetate—ethylenediaminetetraacetic acid (AAAc-EDTA) extractable Zn, Cu, B, Fe, Mn, Cd, Mo, Ni, Al, Co, Pb, and pH (H_2O) between 1974 and 2018 in the 0–15 cm layer of cultivated soils of Finland, (2) to present the current status of soil labile microelement concentrations, (3) to explore the temporal trends of the same element concentrations (except B) in oat and barley grains between 1988 and 2019, and finally (4) to assess the conformity between the microelement trends in grains and soil.

2 | MATERIALS AND METHODS

2.1 | Soil monitoring network

The soil monitoring network covering the whole agricultural area of Finland was established in 1974 ($n = 2042$) and resampled to a gradually reducing extent (due to ceased farming, unreliability of sampling site location, and resource limitations) in 1987 ($n = 1362$), 1998 ($n = 720$), and 2009 ($n = 611$; Heikkinen et al., 2013; Keskinen et al., 2016). In the most recent campaign conducted in 2018, the number of sites was 630, consisting of the remaining 480 original sampling sites complemented by 150 new sites, randomized accounting for the existing network. Throughout the five sampling rounds, regional coverage was maintained. The coordinates of the sampling sites were determined with the Global Positioning System from 2009 on until the sites were located using printed maps, written descriptions, and map drawings.

According to Heikkinen et al. (2013), the network represents Finnish croplands relatively well with respect to soil texture and management practices. The soils of Finland are young, as they formed after the last glaciation during the previous 12,000 years. In coastal regions, subaquatically formed clays are common, whereas soils of the inland are generally coarser in texture. Organic soils occur dominantly in the west and north. Based on the World Reference Base classification, the soils of Finland fall mostly under the classes Stagnosols, Gleysols, Regosols, and Histosols (Lilja et al., 2006). The climate is humid-continental with cold winters of snow cover and ground frost and cool summers. Although agricultural production extends to the Arctic Circle, it is centred strongly on the most favourable areas in the south and west. The most common annual crops are spring sown barley, oats, wheat, and turnip rape, whereas silage and dry hay are the prevailing perennial crops usually renewed every 3–4 years and rotated with annual crops.

2.1.1 | Soil sampling, classification, and laboratory analysis

At each sampling site, a composite soil sample was formed by bulking a minimum of four subsamples collected from the soil surface layer (0–15 cm) over an area of 10 m × 10 m. The samples were air-dried and ground to pass through a 2-mm sieve. Visible plant material and roots were removed during sampling and grinding.

The textural composition of the samples was assessed visually and by feel between 1974 and 1998. In 2009, all mineral soil samples ($n = 611$) and in 2018, mineral soil samples from the 150 new sites, were analyzed for particle size distribution by the pipette method of Elonen (1971). Total carbon (C) was determined by wet oxidation in 1974 and by dry combustion thereafter, and the obtained concentrations were converted to organic matter contents with the Van Bemmelen factor (1.724). The soils were then classified into four types (mull, peat, clay, and coarse) according to their organic matter and clay (<0.002 mm) contents. Soils containing 20%–40% organic matter were specified as mull, and those containing >40% organic matter were specified as peat. Mineral soils (<20% organic matter) were separated into clays (>30% clay) and coarse soils (<30% clay).

Microelements (excluding B) were extracted with 0.5 M ammonium acetate-acetic acid–0.02 M ethylenediaminetetraacetic acid (AAAc-EDTA) solution adjusted to pH 4.65 (Lakanen & Erviö, 1971). Samples of 25 mL were shaken in 250 mL of the AAAc-EDTA solution for 1 h in an end-over-end shaker at 27 rpm, and thereafter, each suspension was passed through a paper filter (pore size 8–10 μm). In 1974, the element concentrations were analyzed with an atomic absorption spectrophotometer (AAS), except for Mo, which was determined colorimetrically by a zinc-dithiol method (Stanton & Hardwick, 1967). Since 1987, Mo has been analyzed with graphite furnace (GF)-AAS, and other elements have been analyzed with inductively coupled plasma emission optic spectrometry (ICP-OES) with the exception that in 1987, Cd (included in the study at that time) and Pb were still determined with AAS.

For B, hot water extraction was used (Berger & Truog, 1944). In the method, 10 mL of soil mixed with 20 mL of ultrapure water was boiled under reflux for 5 min, after which five drops of 1 M $CaCl_2$ were added, and the suspension was filtered. In 1974, the B concentration was analyzed by the carmine method (Hatcher & Wilcox, 1950) and thereafter with ICP-OES. Soil pH was measured in a 1:2.5 soil–water suspension.

2.1.2 | Grain sampling and laboratory analyses

The quality and safety of Finnish grain harvest are being monitored annually at the farm level by the Finnish Food Authority's Plant Analysis laboratory. Grain samples are collected from 1500 farms selected randomly each year from a register of agricultural and horticultural farms. Thereafter, a subset of c. 100 grain samples per year and per cereal species is randomly selected by the Natural Resources Institute Finland (Luke) for use in national monitoring of Se status. In the sample selection, geographical representativeness is accounted for. In this study, thirty samples per year of oats and barley were randomly chosen from the Luke's archived subset of grains, including 1988, 1993, 1998, 2009, 2018, and 2019. The years 1998, 2009, and 2018 were the same as in the soil monitoring. Grain samples from 1974 and 1987 included in the soil monitoring were not available, so the closest year 1988 was selected. In three out of four of the selected grain sample years (1988, 1998, and 2018), the yields (national mean) were lower than the average yields during the study period (Official Statistics of Finland). Therefore, two additional years, 1993 and 2019, with higher yields (national mean) for oats and barley were included in the grain sample analysis. Approximately 0.25 g of ground grain samples were digested in a Mars 6 iWave microwave digestor in concentrated HNO_3 with the temperature program: ramp 20 min 180°C, hold 15 min. The microelements were measured by inductively coupled plasma mass spectrometry except Fe which was measured by inductively coupled plasma optical emission (ICP-OES) spectrometry.

2.2 | Data analysis

Two different statistical analyses were conducted: temporal changes in the soil microelement concentrations in different soil classes and temporal changes in microelement concentrations in grains of oats and barley.

Because the distributions of all soil microelement concentrations were positively skewed, a log transformation was applied to all microelements. A general linear mixed model was fitted using a normal distribution (identity link) (Gbur et al., 2012). Additionally, a gamma distribution (log-link) was examined, but due to some convergence problems, we used a log-normal distribution for all soil microelement concentrations. In those cases where the models converged with both distributions, the results were practically the same.

The models were fitted using pseudolikelihood estimation (Gbur et al., 2012). The statistical significance of the fixed effects was determined through Wald F-tests, and a conservative approach for estimating degrees of freedom was used by forcing them to the number of subjects (lowest number of sampling sites in any year).

Statistical estimation and hypothesis testing were performed on log-transformed data and the estimated means, and the endpoints of the confidence intervals were converted to the original scale using the exponential function. *p*-Values of 0.01 or less were regarded as statistically significant.

The model fit was checked from the shape of Pearson residuals and observed versus predicted plots. Residual distributions were symmetrical and obs. versus pred. plots were evenly distributed along the diagonal. The effect of outliers was checked by reanalyzing datasets in which observations with Pearson residual > |2.5| were excluded. The results were not changed; therefore, the original dataset was used for analysis. The modelling was performed by the GLIMMIX procedure of the SAS/STAT software (version 9.4, SAS Institute, 2018).

The statistical model for temporal changes in soil microelement concentrations (SMC) was as follows:

$$\begin{aligned}\log(\mathrm{SMC}) &= \text{intercept} + \text{year}\,(F) + \text{soil class}\,(F) + \text{year}\,(F)\\ &\quad \times \text{soilclass}\,(F) + \text{municipality effect}\,(G) + \text{year}\,(R), \quad (1)\end{aligned}$$

where *F* stands for fixed effect, *G* for G-side random effect (random intercept) that gathers the spatial effect of sampling sites clustered within a municipality, and *R* for R-side repeated effect over the years with unstructured (UN) variance-covariance matrix structure and sampling site within the municipality as a subject (Stroup, 2012).

The general shapes of temporal trends in the outcome variables within the soil class between 1974 and 2018 were tested through orthogonal polynomial trend contrasts by confining to first (a linear curve with no bend) and second (a quadratic curve with one bend) order trends (Gbur et al., 2012). Since the years were unevenly spaced, the contrast coefficients were calculated with the ORPOL function of SAS/IML software (version 9.4, SAS Institute, 2018).

Due to multiple statistical tests, *p*-values and confidence intervals in each test-set were adjusted by using a simulation-based correction in a step-down fashion (Westfall, 1997; SAS 9.4 documentation: PROC GLIMMIX, ESTIMATE-statement, ADJDFE = row, ADJUST = sim, STEPDOWN(ORDER = *p*-value)). Conclusions from statistical comparisons are based on adjusted *p*-values and confidence intervals when applicable.

The statistical model for temporal changes in grain microelement concentrations (GMCs) was as follows:

$$\log(\mathrm{GMC}) = \mathrm{intercept} + \mathrm{year}\,(\mathrm{species})\,(F), \qquad (2)$$

where F is a fixed variable.

The general shapes of temporal trends in the outcome variables within cereal species between 1988 and 2019 and the difference between trends of cereal species were tested through orthogonal polynomial trend contrasts as above for the soil data. Multiplicity adjustment was also conducted as above.

3 | RESULTS

3.1 | Temporal trends in soil and grain microelement concentrations

The model estimates for soil AAAc-EDTA extractable microelement concentrations between 1974 and 2018 are presented in Figure 1, and an overview of the general shapes of the obtained trends (first and second order) is given in Table 1. The model estimates for total microelement concentrations in oat and barley grains between 1988 and 2019 are presented in Figure 2, and an overview of the obtained linear and quadratic trends in the grain element concentrations is given in Table 1. Descriptive statistical output covering numbers of samples, annual means, medians, minimum and maximum values, and standard deviations by soil types and cereal species are given in Tables S1 and S2, respectively.

The estimated soil Al concentration remained at a constant level in coarse and mull soils throughout the study period of 1974–2018 (Figure 1). Clay soils exhibited a slightly decreasing trend of Al, but the linear trend was not statistically significant ($p = 0.03$). The most prominent trend in soil Al was the linear increase in peats, slowing down towards the end of the study period. In both barley and oat grains, Al showed a decreasing trend between 1988 and 2019 (Figure 2).

For B, the mean estimates ranged between 0.3 and 0.8 mg L^{-1}, exhibiting a quadratic trend in all soil types, although in mineral soils, the increase in the estimated concentrations seemed to have levelled off after 1987 (Figure 1). The concentrations of B in grains were not determined.

Throughout the years, the model estimate for Cd was consistently 0.08 mg L^{-1} in clay soils and 0.10 mg L^{-1} in mull soils (Figure 1). The coarse soils exhibited a mild decrease at a slightly lower level in comparison to the clays. In the peat soils, the trend of Cd fluctuated, but an overall increase of ca. 30% was observed. In oat and barley grains, Cd concentrations first decreased, reaching the lowest values in 1998 and 2008, and thereafter increased, although not up to 1988 values (Figure 2).

For Co, a quadratic trend was observed in coarse and mull soils, but the change was small (Figure 1). In clay and peat soils, the model estimates followed an increasing linear trend. In cereal grains, Co exhibited a clear linear decrease in barley, whereas in oats, no discernible trend could be seen (Figure 2). The concentrations of Co were consistently higher in oat grains than in barley grains.

In soil Cu, an increasing linear trend was present in all soils with an indication of levelling off after 1998 apart from mulls (Figure 1). The relative change in soil Cu from 1974 to 2018 was highest in peat (115%), followed by mull (65%), coarse (60%), and clay (30%) soils. In the cereal grains, the Cu concentration followed an upward quadratic trend that showed an overall increase in oats but not in barley (Figure 2).

Soil Fe showed an increasing linear trend in coarse, clay, and peat soils, although in the coarse soils, an indication of a cease in the increase was observed, and in the clay soils, some fluctuation in the trend was observed (Figure 1). The overall increase was approximately 20%, 35%, and 65% from the initial level in the coarse, clay, and peat soils, respectively. Mull soils showed no significant trend in Fe. In oat grains, the increasing trend in Fe was statistically significant, although the variation between the years was large. In barley, no trend in Fe was discerned (Figure 2).

The estimates for Mn in the mineral soils were at a level of 40 mg L^{-1} throughout the study period (Figure 1). In mull soils, Mn remained at approximately 30 mg $\mathrm{L}^{-1,}$ and in peats, a similar level was reached due to a linear increase from the 1974 level. In barley grains, a linear decrease in Mn was observed (Figure 2). In oats, the concentrations of Mn were over twice as high as those in barley, and no consistent temporal trend occurred.

In organic soils, the model estimates for soil Mo fluctuated approximately 0.05–0.06 mg L^{-1} and in coarse soils approximately 0.03–0.04 mg L^{-1} over the years (Figure 1). In clay soils, a minor linear increase from 0.03 to 0.04 mg L^{-1} was recorded. The grain Mo concentrations ranged between 400 and 830 μg kg^{-1} dm in oats and 275 and 440 μg kg^{-1} dm in barley, with no discernible temporal trend (Figure 2).

Increasing trends in soil Ni were observed in all soil types except clays, in which the estimates remained at a 1.2 mg L^{-1} level throughout (Figure 1). In coarse mineral soils, the change was marginal (<10%), but in the organic soils, the increases were more prominent: ca. 50% in mull and ca. 70% in peat. The concentrations of Ni were constantly approximately 10-fold higher in oats than in barley, but no temporal trend was detected in either of the cereals (Figure 2).

The trends of the soil Pb concentration in all soil types were characterized by notable temporal fluctuations. However, apart from mull soils, in which the estimates varied by approximately 2.5 mg L^{-1}, the trends showed an overall increase with time (Figure 1). In coarse and clay soils, the increase was only slightly above 10%, but in peat soils, the estimate for Pb increased by ca. 25%. In contrast, in both oat and barley grains, Pb showed a clear decreasing trend over the study years, as the concentrations dropped from above 20 μg kg^{-1} dm in 1988 to above 4 μg kg^{-1} dm in 2019 (Figure 2).

The temporal trends in soil Zn showed an increase in the organic soils although in mull the increase (ca. 40%) seemed to occur mainly between 1987 and 1998, whereas in peats, the increase (ca. 125%) continued quite steadily from 1987 to 2018 (Figure 1). In contrast, in

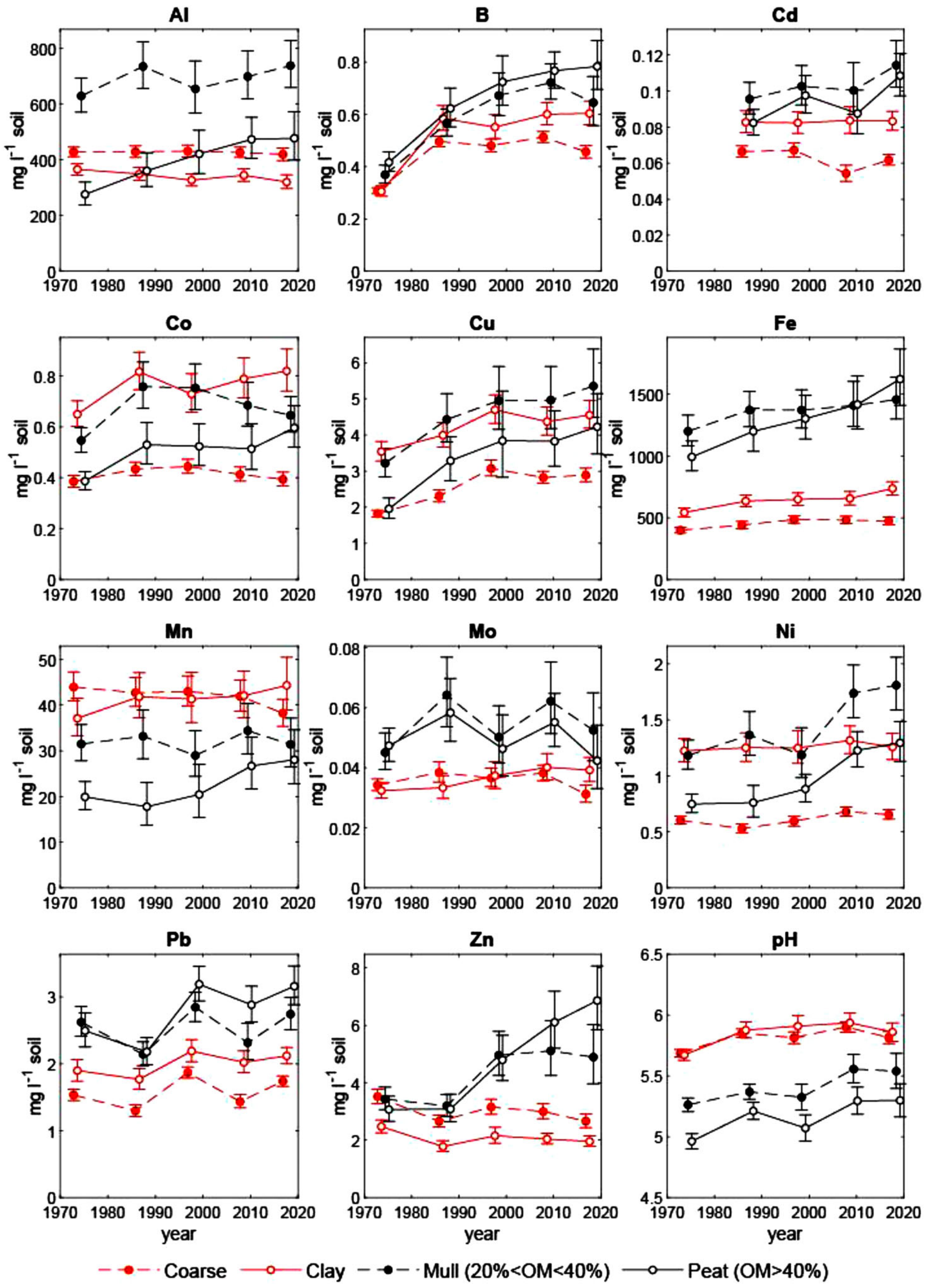

FIGURE 1 Temporal trends in acid ammonium acetate—ethylenediaminetetraacetic acid (AAAc-EDTA) extractable aluminium, boron, cadmium, cobalt, copper, iron, manganese, molybdenum, nickel, lead, and zinc concentrations (mg L^{-1}) and pH (H_2O) between 1974 and 2018 in the 0–15 cm surface layer of cultivated coarse, clay, mull, and peat soils in Finland. The values are mean estimates ±95% confidence intervals.

mineral soils, the trends in Zn were decreasing (20%–25%), although in clays, the linear trend did not turn statistically significant ($p = 0.02$). In the Zn concentrations of barley grains, an upward quadratic shape was discerned with a negligible overall change (Figure 2). Oat grains showed no temporal trend in Zn concentrations, which fluctuated in a similar range of 30–40 mg kg^{-1} dm as in barley.

In mineral soils, pH exhibited a quadratic trend starting from 5.7 and ending to 5.8 in the coarse and 5.9 in the clay soils (Figure 1). In organic soils, the pH increased from 5.3 to 5.5 in the mulls and from 5.0 to 5.3 in the peats, with a small drop in 1998.

3.2 | Current soil microelement status

The most recent (year 2018) mean, median, minimum, and maximum soil AAAc-EDTA extractable element concentrations are shown in Table 2. The values are presented on a volume basis due to major differences in bulk densities between the soil types. The bulk densities are also given to allow conversion to mass-based units. For the nutrient elements B, Cu, Mo, and Zn, soil test reference values describing seven fertility classes (poor, rather poor, fair, satisfactory, good, high, and excessive) are also included (Eurofins Agro, 2020). The current

TABLE 1 An overview of linear and quadratic trend shapes and their statistical significance (adjusted *p*-values) for microelement concentrations in soil between 1974 and 2018 and in barley and oat grains between 1988 and 2019.

		Soil test results				Grain sample results		
Element	**Trend**	**Coarse Adj. *p***	**Clay Adj. *p***	**Mull Adj. *p***	**Peat Adj. *p***	**Oats Adj. *p***	**Barley Adj. *p***	**diff. Adj. *p***
Al	Linear	0.789	0.025	0.312	↗ < **0.001**	↘ <**0.001**	↘ <**0.001**	0.432
	Quadratic	0.709	0.560	0.832	0.027	0.687	0.857	0.949
B	Linear	↗ < **0.001**	↗ < **0.001**	↗ < **0.001**	↗ < **0.001**			
	Quadratic	↗→ < **0.001**	↗→ < **0.001**	↗ < **0.001**	↗ < **0.001**			
Cd	Linear	↘ < **0.001**	0.977	0.039	↗ < **0.001**	0.011	0.695	0.042
	Quadratic	0.046	0.977	0.546	0.667	0.016	↘↗ <**0.001**	0.028
Co	Linear	0.838	↗ **0.003**	0.258	↗ < **0.001**	0.751	↘ **0.004**	0.298
	Quadratic	↗↘ < **0.001**	0.102	↗↘ < **0.001**	0.141	0.393	0.949	0.733
Cu	Linear	↗ < **0.001**	↗ < **0.001**	↗ < **0.001**	↗ < **0.001**	↗ <**0.001**	0.123	<**0.001**
	Quadratic	↗→ < **0.001**	↗→ **0.001**	0.053	↗ **0.002**	↘↗ <**0.001**	↘↗ **0.010**	0.743
Fe	Linear	↗ < **0.001**	↗ < **0.001**	0.095	↗ < **0.001**	↗ <**0.001**	0.214	<**0.001**
	Quadratic	↗→ < **0.001**	0.582	0.579	0.950	0.255	0.214	0.049
Mn	Linear	0.022	0.185	0.996	↗ < **0.001**	0.228	↘ **0.001**	0.529
	Quadratic	0.181	0.733	0.996	0.103	0.925	0.900	0.940
Mo	Linear	0.180	↗ < **0.001**	0.449	0.597	0.053	0.130	0.991
	Quadratic	↗↘ < **0.001**	0.822	0.083	0.091	0.441	0.806	0.643
Ni	Linear	↗ < **0.001**	0.754	↗ < **0.001**	↗ < **0.001**	0.536	0.175	0.073
	Quadratic	↘↗ < **0.001**	0.754	0.111	0.056	0.561	0.115	0.465
Pb	Linear	↗ < **0.001**	↗ < **0.001**	0.328	↗ < **0.001**	↘ <**0.001**	↘ <**0.001**	0.558
	Quadratic	0.073	0.928	0.078	0.521	0.994	0.905	0.872
Zn	Linear	↘ < **0.001**	0.023	↗ **0.001**	↗ < **0.001**	0.022	1.000	0.214
	Quadratic	0.079	0.023	0.446	0.200	0.041	↘↗ **0.002**	0.751
pH	Linear	↗ < **0.001**	↗ < **0.001**	↗ **0.001**	↗ < **0.001**			
	Quadratic	↗↘ < **0.001**	↗↘ < **0.001**	0.821	0.502			

Note: Significant ($p \leq 0.01$) trends are marked with ↗ or ↘ depending on the general direction of the trend. It is noteworthy that these nationwide soil and plant datasets are independent of each other, and the grain samples were not collected from the soil sampling sites.

mean and median values for these elements fall into the range between fair and good, while the minimum values mostly indicate poor, and the maximum values indicate high soil nutrient status. The organic soils tended to exhibit higher fertility classes in comparison to the mineral soils.

4 | DISCUSSION

For most of the studied elements, a significant increasing temporal trend was identified in the readily available soil concentrations over the study period from 1974 to 2018. The increase was most prominent in peat soils in which progressive humification with age increases the ash content and decreases the C content (Hyväluoma et al., 2020). A similar, although gentler, concentrating effect could be seen in the mull soils. For several of the studied microelements, the temporal trends exhibited an increase over the first decades assessed and then levelled off or even seemed to turn to a slight decrease toward the end of the study period. Previous balance calculations have shown atmospheric deposition and fertilizer products to be the major sources of microelements in Finnish arable soils, whereas harvested crops, erosion, and leaching constitute the main outflow routes (Mäkelä-Kurtto et al., 2007; Salo et al., 2018). The introduction and application of micronutrient fertilizers in Finland dates to the 1970s (B and Cu) and 1980s (Zn), which explains the increases in soil concentrations at the beginning of the study (Erviö et al., 1990). However, micronutrient sales decreased in the mid-1980s (Official Statistics of Finland, 1995), and similarly, during 1980s–1990s, the atmospheric deposition of metals (Cd, Cu, Fe, Ni, Pb, Zn) decreased due to tightened environmental regulations and technological improvements (Poikolainen et al., 2004). Both these developments agree with the observed levelling of the trends in soil concentrations. Furthermore, wet deposition data from the 2000s indicate plateauing decreasing trends in metal deposition, except for Pb, and even a turn to an increase for Cu and Fe (Kyllönen et al., 2009). In

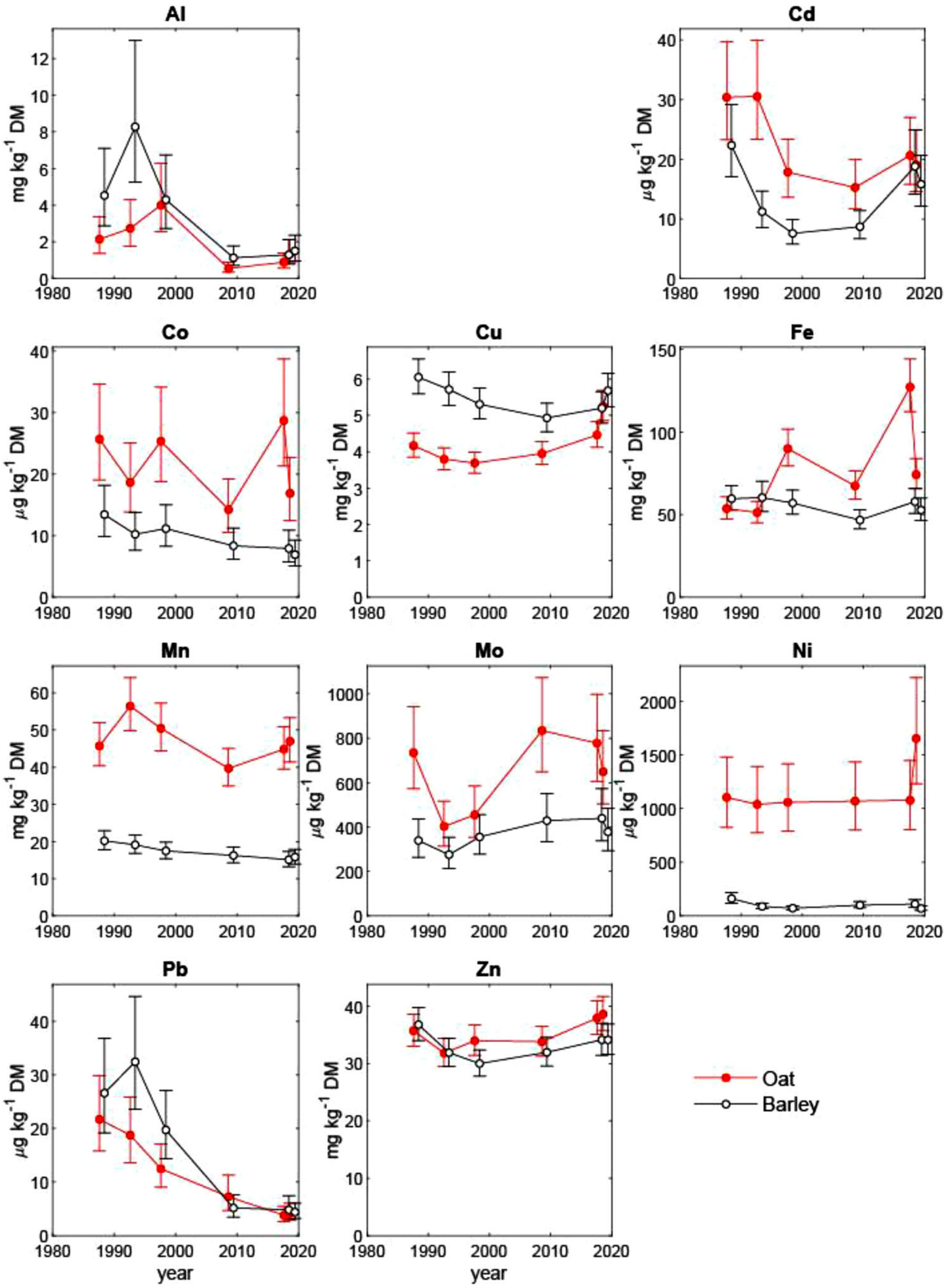

FIGURE 2 Temporal trends in aluminium, cadmium, cobalt, copper, iron, manganese, molybdenum, nickel, lead, and zinc concentrations in oat and barley grains between 1988 and 2019. The values are mean estimates ±95% confidence intervals.

Finland, the majority of trace element deposition are due to long-range atmospheric transport; therefore, meteorological factors contribute to changes in it. Due to the estimated increase in rainfall amounts in Finland within the changing climate, wet deposition is expected to slightly increase (Kyllönen et al., 2009).

The decrease in micronutrient concentrations has been suggested to originate from the depletion of soil because of intensified agriculture, but this view has been questioned due to the lack of direct evidence (Marles, 2017). In the current study, a clear indication of a decreasing soil concentration was observed only for Zn in the coarse-textured soils, and this trend was not reflected in the Zn concentrations of cereal grains. Overall, the trends in micronutrients in oat and barley grains from 1988 to 2019 did not reveal a consistent decrease in the essential micronutrient contents. Only barley grains exhibited a decreasing trend in Mn and Co. Increasing trends were detected for Cu and Fe in oats, which agrees with the general trends in the current soil data.

In the current study, the mean Mn content was higher in oats than in barley, which agrees with previously reported results (e.g., Redshaw et al., 1978; Rubene & Kuka, 2006). Lombnæs and Singh (2003) showed

TABLE 2 Bulk densities (g cm^{-3}) for clay, coarse, mull, and peat soils and concentrations of AAAc-EDTA extractable elements (mg L^{-1}) in clay, coarse, mull, and peat soils in 2018 sampling.

	Soil type			
	Clay	Coarse	Mull	Peat
Element	*n* = 143	*n* = 387	*n* = 37	*n* = 45
Bulk density	0.9 ± 0.1	1.1 ± 0.1	0.7 ± 0.1	0.5 ± 0.1
	(0.5; 1.0; 1.1)	(0.7; 1.1; 1.5)	(0.4; 0.7; 0.9)	(0.3; 0.5; 0.6)
Al	347 ± 195	466 ± 243	840 ± 336	564 ± 368
	(98; 312; 1528)	(81; 425; 1437)	(317; 840; 1939)	(81; 560; 1553)
B	0.7 ± 0.3 (F)	0.5 ± 0.3 (F)	0.7 ± 0.4 (S)	1.0 ± 0.5 (S)
	(0.2; 0.7; 1.9)	(0.1; 0.5; 1.7)	(0.1; 0.6; 1.5)	(0.4; 0.9; 2.3)
Cd	0.10 ± 0.04	0.07 ± 0.03	0.12 ± 0.06	0.10 ± 0.04
	(0.03; 0.09; 0.25)	(0.01; 0.06; 0.32)	(0.05; 0.12; 0.40)	(0.03; 0.09; 0.22)
Co	1.1 ± 0.6	0.5 ± 0.4	0.8 ± 0.4	0.7 ± 0.3
	(0.2; 0.9; 3.3)	(0.1; 0.4; 2.7)	(0.2; 0.7; 2.0)	(0.1; 0.6; 1.5)
Cr	0.4 ± 0.3	0.3 ± 0.3	0.4 ± 0.2	0.2 ± 0.2
	(0.1; 0.3; 1.9)	(0.1; 0.3; 3.1)	(0.1; 0.3; 1.1)	(0.0; 0.1; 0.7)
Cu	5.4 ± 2.4 (G)	3.7 ± 3.1 (S)	6.7 ± 4.2 (G)	6.4 ± 4.0 (G)
	(0.9; 4.7; 12)	(0.3; 2.8; 34)	(1.0; 5.9; 19)	(0.8; 5.6; 17)
Fe	801 ± 425	563 ± 390	1832 ± 719	2023 ± 1238
	(233; 739; 4009)	(109; 466; 2440)	(427; 1781;3535)	(466; 1755; 6265)
Mn	63 ± 63	53 ± 45	40 ± 29	45 ± 40
	(5.9; 51; 545)	(5.1; 38; 318)	(12; 30; 118)	(3.1; 35; 207)
Mo	0.05 ± 0.04 (S)	0.05 ± 0.05 (S)	0.08 ± 0.08 (G)	0.09 ± 0.13 (G)
	(0.01; 0.04; 0.24)	(0.00; 0.03; 0.52)	(0.01; 0.04; 0.42)	(0.00; 0.05; 0.50)
Ni	1.7 ± 1.0	0.8 ± 0.8	2.1 ± 1.4	1.5 ± 0.9
	(0.4; 1.5; 4.9)	(0.2; 0.6; 13)	(0.6; 1.7; 7.1)	(0.3; 1.3; 4.4)
Pb	2.6 ± 1.0	1.7 ± 0.7	2.8 ± 0.8	2.8 ± 1.1
	(1.0; 2.5; 5.5)	(0.4; 1.6; 5.5)	(1.3; 2.7; 4.8)	(0.8; 2.9; 5.9)
Zn	2.2 ± 1.7 (S)	3.7 ± 3.8 (S)	6.8 ± 6.4 (G)	8.7 ± 5.4 (G)
	(0.4; 1.6; 9.7)	(0.3; 2.6; 33)	(1.3; 5.0; 32)	(2.7; 7.2; 29)

Note: Values are means ± standard deviations; minimum, median, and maximum values are given in parentheses. Bulk density values are for ground and sieved soils and applicable for unit conversion to mass basis. Capital letter in parenthesis shows the fertility class (based on the mean value) when available for a given element. Fertility classes according to Eurofins Agro (2020): poor (P), rather poor (RP), fair (F), satisfactory (S), good (G), high (H), and excessive (E); no pH corrections have been applied.

that similar Mn availability resulted in lower Mn in barley, suggesting passive and unrestricted uptake of Mn in oats. According to Sillanpää (1982), the amounts of microelements absorbed by plants indicate reliably the available micronutrient fractions in soil. Thus, the constantly higher Mn content in oat than in barley grains between 1989 and 2019 indicates differing Mn uptake mechanisms in oats and barley rather than deficiency of Mn in soil. Furthermore, there was no increase or decrease in soil plant-available Mn between 1974 and 2018. This suggests that the detected decreasing trend in Mn contents in barley may result from the variation in nutrient uptake between barley varieties and of their yearly prevalence rather than changes in soil nutrient content. Fan et al. (2008) found that at Rothamsted, England, the changes in cultivars and especially the introduction of short-straw cultivars were the main reasons for the decrease in the mineral content of wheat between 1845 and 2005, instead of the depletion of plant-available nutrients in soil or of increasing yields. The decreasing trend in Co contents in barley and at the same time, the over 50% higher Co contents in oats than in barley grains points towards differences in nutrient uptake between the species rather than towards risk of soil born deficiency. In addition, Fe and Ni contents were higher in oats than in barley. For Fe, the finding is parallel with the results of, for example, Jordan-Meille et al. (2021), and for Ni, with the results of Hamnér et al. (2013) and Jākobsone et al. (2015); all results indicate differences in the uptake of these micronutrients between barley and oats.

A positive observation regarding food safety was the decreasing trend of potentially toxic Al and Pb in both oat and barley grains. Lower

Al contents were measured in grains after 2000, a finding in agreement with the results of Ekholm et al. (2007) showing decreased Al contents in Finnish cereal products between 1967 and 2003. The trend may be related to the observed increase in soil pH, which is known to render Al less available to plants. For elements with pronounced pH-dependent mobility, a pH correction to results obtained with buffered soil extractants is often applied to improve the congruence between soil and plant analyses. Although available Pb in soil samples showed an increasing trend between 1974 and 2018 (except in mull), the Pb content in oat and barley grains was decreasing. This finding reflects the reduction in Pb emissions (Kyllönen et al., 2009), as the Pb content in aboveground plant parts is known to be associated with atmospheric deposition rather than uptake from the soil (Zhao et al., 2004). In contrast to Pb, Zhao et al. (2004) found plant root uptake to be the main pathway contributing to the Cd content in grains. According to Andersson and Siman (1991), Cd in P fertilizers may contribute to grain Cd contents. In the present data, the highest Cd grain contents were measured in 1989 and the lowest in 1998 and 2009 in barley and oats, respectively, reflecting the 1986 transformation to Cd-free raw phosphate as the source of P fertilizers in Finland (Erviö et al., 1990). At the same time, the P fertilization applied per hectare decreased significantly (Uusitalo et al., 2007). In general, the Cd contents in Finnish grain samples were at the lower end of the variation reported for barley and oat grains in the United Kingdom (Jordan-Meille et al., 2021) and in Latvia (Jākobsone et al., 2015). In the 2000s, the mean contents of the potentially toxic elements Al, Pb and Cd in oats and barley were all markedly lower than the contents measured for oats and barley grown in Italy (Brizio et al., 2016).

The risk of insufficient replacement of nutrients removed in—macronutrient fertilization-induced—enlarged yields is greatest in the regions of highly weathered and inherently nutrient poor soils, but the risk should not be overlooked in the younger soils of Northern Europe (Jones et al., 2013). A previous survey by Sillanpää (1982) showed that in Finnish soils, Mn and Mo were mainly within, Cu and B were somewhat below, and Zn and Fe were clearly above the global range. Now, approximately 40 years later, the mean and median values of micronutrients (Table 2) generally show a satisfactory or good status. However, the lower range of values indicates local deficiencies in B, Cu, Mn, Mo, and Zn. Since rather high values are also found, a soil analysis can be recommended for assessing fertilization needs. For the harmful elements, the concentrations in the soils of Finland are overall rather low (Reimann et al., 2014), which is reflected in the relatively low grain contents in our material.

5 | CONCLUSIONS

The results of this research revealed no alarming decreases in micronutrient contents in agricultural soils or barley and oat grains. In soils, the microelement trends between 1974 and 2018 were stagnant or increasing except for Zn, which showed a decrease in coarse mineral soils. In cereal grains, only the potentially toxic elements Al and Pb decreased between 1989 and 2019. Increasing trends consistent with the soil data were detected for Cu and Fe in oat grains, in which the micronutrient contents were higher than those in barley. The current soil micronutrient status in Finland is satisfactory or good, but deficiencies in B, Cu, Mn, Mo, and Zn may occur locally.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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