Soil quality under different agricultural land uses as evaluated by chemical, geochemical and ecological indicators in mountains with high rainfall (Darjeeling Himalayas, India)

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Abstract

Purpose Agricultural land use associated with intensification in plant growing affects the physicochemical parameters of soils and thus soil quality. The aim of this study was to identify the quality of soils in the Peshok catchment in the Eastern Himalayas under different types of land use (tea, horticulture, rice), under high monsoon rainfall conditions.

Methods Physical and chemical analysis were used, e.g. bulk density, pH, TOC, major and trace element content (such as Ca, Fe, K, Mg, Mn, Na, P, Ag, As, Ba, Cd, Co, Cr, Cu, Ga, Li, Mo, Ni, Pb, Sr and Zn) based on AntonPaar Multiwave 3000 microwave system and nitrogen and sulphur content using CNS Elementar Vario MAX cube analyser. In addition, ecotoxicological analyses were performed using the Microtox test. Soil quality was assessed on the basis of chemical indicators related to the mobility of trace elements (risk assessment code, individual contamination factor and global contamination factor); geochemical indicators (geoaccumulation index, enrichment factor, contamination factor, contamination degree, modified contamination degree, pollution load index and improved Nemerow pollution index); and ecological indicators (potential ecological risk coefficient and potential ecological risk index). A statistical package of Statistica v13 was used for statistical analysis.

Results Soils in the analysed catchment are characterised by low contents of macro- and microelements. It was shown that natural factors, such as high precipitation and steep slopes, favour strong leaching of elements from the soil, and farming systems based on natural and chemical fertilisation, terracing and irrigation are not able to balance them fully. The contents of TOC and N as well as pH were determined mainly by the land use. Results of statistical analyses and geochemical indicators revealed the predominantly natural origins of elements. The higher Ga content indicated a relationship with the local geology and the higher Cu content with the use of fertilisers. Indicators showed a low ecological risk related to the presence of trace elements, and soil ecotoxicity to A. fischeri was generally determined by the acidic pH of the tested soils. Low mobility was observed for most elements, and regardless of the land use, the residual fraction predominated.

Conclusion The study showed that use of many indicators can more fully describe soil quality in relation to land use, especially in the case of a low content of trace elements. Moreover, this approach helps to better understand the changes taking place in soil quality under different land uses in mountains with high rainfall.

Keywords Agricultural soil · Macro- and microelements · Geochemical indicators · Heavy rainfall · Darjeeling Himalayas, India

1 Introduction

Soil quality is defined as the capacity of soil to perform ecological functions and provide ecosystem services to maintain biological productivity and environmental quality and improve plant and animal health (Bunemann et al. 2018; Maurya et al. 2020). Soil quality assessment is generally
based on a combination of the environmental quality, productivity and sustainability of the soil (Liu et al. 2016). Soil quality can be assessed on the basis of characteristics related to soil fertility, which are typical parameters used for crops, such as pH, organic matter content, macronutrients (with the most commonly discussed N, P and K) or grain size (Klimkowicz-Pawlas et al. 2019). Soil quality can also be evaluated based on ecological risks or geochemical indicators. These indicators are primarily aimed at assessing the anthropogenic impact on soil quality and can reveal the potential impacts of trace elements on soil ecology, including the possibility of contamination (Wieczorek et al. 2018; Kowalska et al. 2018; Mekky et al. 2019; Ciarkowska and Gambus 2020; Klimkowicz-Pawlas and Debeane 2020; Fazekašová and Fazekaš 2020). For several years, ecotoxicological tests have also been used in soil quality assessments (Baran et al. 2014; Baran and Wieczorek 2015; Alves and Cardoso 2016; El-Alam et al. 2018; Abou El-Anwar et al. 2021; Salman et al. 2021). All the above methods play an important role in monitoring soil quality to ensure the sustainability of agroecosystems.

Trace elements are crucial to consider in the assessment of soil quality. They are essential for proper plant growth and the maintenance of high levels of food production (He et al. 2005; Tóth et al. 2016). However, many studies have shown that the excessive accumulation of trace elements is a major factor in reducing soil fertility and thus inhibiting soil functions (Wuana and Okieimen 2011; Wieczorek et al. 2018; Ciarkowska and Gambus 2020; Gruszecka-Kosowska et al. 2020; Fazekašová and Fazekaš 2020). Elements are released into the soil environment from both natural and anthropogenic sources. Some of these elements are macro- or micronutrients used by plants; therefore, land use, as well as the system and type of cultivation, affects their content in the soil (Lal 2015; Kopitke et al. 2019; Salman et al. 2021). Deforestation is a common feature of the onset of agricultural development in monsoon climates (Ramakrishnan 1992; Lal 2000). In addition, as agricultural production intensifies, cultivation systems acquire individual characteristics through, for example, the terracing of slopes, irrigation, the introduction of monoculture crops, and artificial and natural fertilisation. All of these affect the physicochemical parameters of soils and thus the soil quality (Singh 2008).

In India, most studies on the impact of land use on soil quality have focused on lowland urbanised, industrialised and intensive agriculture areas (e.g. Gowd et al. 2010; Bhatia et al. 2015; Sakram et al. 2015; Bhatti et al. 2017; Ratnakar 2018; Suhr et al. 2021). Studies evaluating the influence of different land use types on soil quality in cultivated hilly and mountainous areas are limited (Chandrasekaran et al. 2015; Prokop et al. 2018; Kumar et al. 2020; Mishra and Rodrigo-Comino 2021). In particular, such studies are lacking for the Eastern Himalayas, including the Darjeeling Himalayas. This region receives among the highest annual rainfalls (up to 4000 mm) and most frequent heavy rains (up to 500 mm day⁻¹) along the entire southern Himalayan margin (Starkel and Basu 2000; Prokop and Walanus 2017). Deforestation for agriculture under such high rainfall and on such steep slopes may lead to a reduction in soil quality, depending on cultivation (land use). We hypothesised that overall soil quality is reduced in areas with high rainfall and certain agricultural land use systems, and that soil quality differences increase in mountainous environments.

The aim of this study was to evaluate soil quality in the Darjeeling Himalayas in relation to different types of agricultural land use (with particular reference to the role of deforestation and high monsoon rainfall in shaping soil quality) based on chemical, geochemical and ecological indicators. More specifically, the study aimed to (i) investigate the variability of topsoil properties under different land use and compare this soil with natural forest soil; (ii) evaluate soil quality based on the mobility of trace elements; and (iii) determine which soil quality indicators are most important for explaining soil quality.

## 2 Materials and methods

### 2.1 Study area

The research was conducted in the Peshok catchment, an area of 10 km² located in the Darjeeling Himalayas of India’s West Bengal state. The Peshok stream is the right bank tributary of the Teesta River, which joins the Brahmaputra. The elevation of the catchment is 220–1930 m a.s.l. (Fig. 1). This mountain landscape is characterised by narrow, deep valleys dominated by steep slopes between 20° and 40° (Starkel and Basu 2000). According to Köppen (1931), the catchment climate is classified as subtropical monsoonal (Cwb); furthermore, the local climatic conditions do not differ from the regional pattern (Prokop and Walanus 2017). The average annual temperature varies between 13.6 °C at the highest ridges and 23 °C at the Peshok outlet. The annual rainfall increases with elevation, ranging from 1900 to 2700 mm. The summer monsoon between June and September provides 80% of this annual rainfall. The catchment is underlain by gneisses with intercalations of quartzites and mica schists (Acharyya 1980). Metamorphic rocks are the substrate for brown soils classified as Inceptisols (Dystrudepts) (Nayak 2001; Soil Survey Staff 2010) or Cambisols (FAO 2015); they are moderately shallow and well drained.

The Peshok catchment is representative of land use history and structure for the Darjeeling Himalayas (Prokop et al. 2020). In the nineteenth century, the land use structure was determined by the establishment of tea plantations that...
induced workforce migration, as well as by the simultaneous reservation of the forest for silviculture (Taylor 1910). Analysis of topographic maps from 1910 and 1966, tea plantation reports and remote sensing data indicated that the natural deciduous forest, tea gardens and cultivated land have retained the same land uses for at least 100 years.

In 2018, land use and land cover (LULC) of the study catchment, assessed by the visual interpretation of Google Earth satellite images and verified in the field (Fig. 1), consisted of forest (48%), horticulture (37%), tea garden (8%), built-up area (4%) and rice cultivation (3%). The natural vegetation in the catchment is restricted to tropical moist deciduous forest, dominated by *Shorea robusta* and an admixture of evergreen and semi-evergreen species such as *Michelia champaca*, *Schima wallichii*, *Terminalia myriocarpa* and *Ailanthus grandis* (Das 2004; Forest Survey of India 2011). Relatively dense vegetation canopy forms the tea bushes (*Camellia sinensis*) of the Peshok Tea Garden located in the middle part of the catchment. Forests and tea plantations are surrounded by terraced cultivated fields of typical width 2–5 m. Depending on water availability, terraces are either level or sloping. Terraces with slopes up to 1° are used for rainfed horticulture with a dominant crop of perennial large cardamom (*Amomum subulatum* Roxburg), sometimes with an admixture of vegetables such as cabbage (*Brassica oleracea*), cucumber (*Cucumis sativus*) and orange trees (*Citrus reticulata* Blanco). The level terraces are used for rice cultivation (*Oryza sativa*). They are irrigated through a network...
of small canals fed by local springs. Features of land use in the Peshok catchment are presented in Table 1.

Flattened ridges and gentle slopes are occupied mainly by settlements and roads. The catchment is inhabited by about 6000 people (Census of India 2011). Most depend on tea plantation work and agriculture for their livelihoods. Approximately 57% of households keep livestock.

2.2 Soil sampling

The soil sampling scheme assumed that long-term natural soil-forming factors such as climate, parent material, topography and time were relatively uniform over the study catchment. Using this study design, we expected that the differentiation of soil properties could be attributed to four land use types: natural deciduous forest, tea plantations, rice cultivation and horticulture. In November 2018, soil samples were collected from these four LULC types in the centre of the Peshok catchment, lying between 300 and 700 m a.s.l. Within each land use type, five soil samples were collected using an 8 cm diameter corer, providing 20 soil samples in total. Sampling was confined to the upper 10 cm of brown soils.

2.3 Sample analyses

2.3.1 Physical analyses

Soil bulk density was calculated by drying the undisturbed soil samples at 105 °C before weighing them. Soil samples were then passed through a 2 mm mesh sieve. The grain size composition of each sample was determined using a Malvern laser diffraction particle sizer after pre-treatment with H2O. Particle size measurements ranged between 0 and 2 mm. Each soil sample was analysed in triplicate and the results of the analysis of these replicates did not differ by more than ±5%.

2.3.2 Chemical analyses

Basic chemical properties were determined from air-dried soil samples, including pH in 1 mol KCl/dm³ by the potentiometric method, total organic carbon (TOC), and nitrogen and sulphur content using a CNS Elementar Vario MAX cube analyser. The total elemental content (Ca, Fe, K, Mg, Mn, Na, P, Ag, As, Ba, Cd, Co, Cr, Cu, Ga, Li, Mo, Ni, Pb, Sr and Zn) was assessed after digesting samples in a mixture of HNO3 (65%) and HCl (30%; 3:1 v/v; suprapur, MERCK) using an AntonPaar Multiwave 3000 microwave system (Baran et al. 2018; Wieczorek et al. 2018). The fractionation analysis of selected trace elements (Cd, Cr, Ni, Zn, Pb and Cu) in soils was performed by sequential chemical extraction procedure proposed by the Community Bureau of Reference (BCR method). Four fractions of trace elements were distinguished: exchangeable (F1), which is easily soluble in an acidic medium; reducible (F2), which is bound to Fe and Mn oxides; oxidisable (F3), or bound to organic matter; and residual (F4), for elements permanently bound to minerals (Gruszecka-Kosowska et al. 2019). Elemental content was determined using an inductively coupled plasma optical emission spectrophotometer (Perkin Elmer ICP-OES Optima 7300 DV). Two replicates of each soil sample were chemically analysed; if the results for these replicates differed by more than ±5%, two further analyses of that sample were performed. The quality of measurement was verified based on the results of elemental determinations obtained for certified materials: CRM 16–050 (total element content), BCR-701 (trace element fractionation) and Soil Standard Loamy no. 133505 (TOC and nitrogen).

| Table 1 | Features of land use in the Peshok catchment |
|---------|---------------------------------------------|
| Features of land use | Forest | Tea | Horticulture-cardamom | Rice |
| Slope (°) | 0–40 | 0–20 | 1 | 0 |
| Infiltration in topsoil | Very high up to 115 mm/min | High up to 70–90 mm/min | Moderate up to 50 mm/min | Moderate up to 50 mm/min |
| Runoff | - | Only during daily rainfall reaching 25–50 mm | 2.2% of rainfall (on the basis of 6 rainfall events) | - |
| Agrotechnical treatments | - | Fertilising: urea, farmyard manure, phosphate, NPK: 120, 45, 120 kg/ha, pruning, harvesting of tea leaves, planting shade tree species | Fertilising: cattle manure or compost and oil cakes: 1 kg/pit/year, tillage, harvesting of cardamom capsules, planting shade tree species | Fertilising: urea, NPK: 120, 26, 50 kg/ha/year, tillage, irrigation from local springs, harvesting of grains and straw |
| Yield | - | 500 kg/ha/year | 230 kg/ha/year | 2800 kg/ha/year |

Source: Froehlich et al. (1992); Froehlich and Starkel (1995); Sharma et al. (2000); Chaudhury et al. (2005); Bisen and Singh (2012); Gudade et al. (2013)
2.3.3 Ecotoxicological analyses

Soil ecotoxicity was assessed using the Microtox test, which uses as test organisms the luminescent bacteria *Alcaligenes fischeri*, whose bioluminescence is a natural result of metabolic processes. Exposure of the bacterial suspension to toxic substances disrupts the metabolic process, thus reducing the light produced. The toxicity of soil samples was tested by conducting a screening test. Luminescence was measured before and after a 15-min incubation of the bacterial suspension with the test sample using an M500 Analyzer (Microbics Corporation 1992; Baran and Wieczorek 2015). In this study, the toxicity classification developed by Persoone et al. (2003) was used to assess soil toxicity. In this classification, a sample is considered non-toxic when the percentage toxic effect PE is below 20%, and low toxic when 20% ≤ PE < 50%. Samples are considered toxic to an organism if the percentage toxic effect lies in the range 50% ≤ PE < 100%, and very toxic when PE = 100% (Persoone et al. 2003; Baran et al. 2014; Baran and Wieczorek 2015).

2.4 Geochemical and ecological indicators

Soil quality was assessed on the basis of chemical indicators related to the mobility of trace elements (risk assessment code (RAC), individual contamination factor (ICF) and global contamination factor (GCF)); geochemical indicators (geoaccumulation index (I\textsubscript{geo}), enrichment factor (EF), contamination factor (CF), contamination degree (Cdeg), modified contamination degree (mCdeg), pollution load index (PLI) and improved Nemerow pollution index (PIN)); and ecological indicators (potential ecological risk coefficient (Er) and potential ecological risk index (RI)), as shown in Table 2. Both single- and multi-element indicators were used in the study. The multi-element indicators are often considered more valuable as they allow a comprehensive assessment of soil quality (Gong et al. 2008; Kowalska et al. 2018; Kulbat and Sokolowska 2019; Gruszecka-Kosowska et al. 2019, 2020; Kumar et al. 2019a). Calculations were made assuming the soil C horizon geochemical background values (GSI 2021).

2.5 Statistical and graphical assessment

Data were compiled by calculating the mean and standard deviation for each study parameter. Differences between means were determined using one-way ANOVA and t\textsuperscript{2}-Tukey tests at the significance level of 0.05. Relationships between data were analysed using Pearson correlation matrices and principal component analysis. Microsoft Excel 2010 spreadsheets and the Statistica 13 package were used for the statistical analysis and graphical presentation of the results.

3 Results and discussion

3.1 Basic properties of soils

The effects of land use on the basic physicochemical parameters of soils are presented in Table 3. In the literature, characteristics such as grain size, pH, organic carbon content and total nitrogen content are considered important indicators of soil quality and fertility (Zhou et al. 2019). The investigated soils were characterised by their different granulometric compositions, with a clay fraction dominating in forest soils and in soils from tea cultivation, and a sand fraction dominating in soil samples collected from vegetable and rice crops. This is associated with the incorporation of coarser grained subsurface horizons into the topsoil during tillage. Soil pH is important for the activation and the immobilisation of macronutrients and trace elements. In the Peshok catchment, the soil pH was categorised as very acidic and acidic, ranging from 4.05 (tea) to 5.24 (rice). These values are typical of monsoonal regions with high rainfall and steep slopes, where soils are commonly characterised by low pH, below 5.5 (Nayak 2001). The significantly higher pH under rice cultivation is due to irrigation with water of pH = 8.1 from local springs.

Soil organic matter is a basic indicator of soil quality that determines its physical and chemical properties as well as biological processes which, in turn, regulate many transformations, referred to as biological activity (Lal 2020). Significantly, the highest organic matter content was found in forest soils (10.7%), while the organic matter content was nearly two times lower in soils from vegetable cultivation and three times lower in soils from rice cultivation. Forest soils and soils from tea cultivation were characterised by similar TOC and N contents, both containing significantly higher content than soils from vegetable and rice cultivation areas, in which values of both parameters were nearly two times lower.

It is generally believed that intensively cultivated soils have lower carbon and nitrogen contents than forest soils, and this was confirmed in our study. The small decline in organic matter (OM) and TOC content in soil from tea cultivation probably reflects the reduction in organic matter being returned to the soil in the form of fallen leaves and plant debris (Natesan and Rangathanathan 1990). Nitrogen is the most important nutrient element in tea cultivation, accounting for approximately 5% of the dry weight of the harvested shoots. Assuming tea yields of approximately 500 kg ha\textsuperscript{-1} year\textsuperscript{-1} in the Darjeeling Himalayas (Bisen and Singh 2012), about 25 kg ha\textsuperscript{-1} year\textsuperscript{-1} of N is removed with the tea leaves. This loss is supplemented by fertilisation with farmyard manure.
Table 2 Chemical indicators related to the mobility of trace elements, geochemical indicators and ecological indicators

| Index* | Formula | Explanation | Limit value | Classification |
|--------|---------|-------------|-------------|----------------|
| Geoaccumulation index | \( I_{geo} = \log_2(C_i/1.5B) \) | \( C_i \) — content of single element in soil; \( B_i \) — background value; \( 1.5 \) — constant | \( \begin{array}{c} I_{geo} \leq 0 \\ 0 \leq I_{geo} < 1 \\ 1 \leq I_{geo} < 2 \\ 2 \leq I_{geo} < 3 \\ 3 \leq I_{geo} < 4 \\ 4 \leq I_{geo} < 5 \\ 5 \leq I_{geo} \end{array} \) | \( \begin{array}{c} \text{Class 0 — practically uncontaminated} \\ \text{Class I — uncontaminated to moderately contaminated} \\ \text{Class II — moderately contaminated} \\ \text{Class III — moderately to heavily contaminated} \\ \text{Class IV — heavily contaminated} \\ \text{Class V — heavily to extremely contaminated} \\ \text{Class VI — extremely contaminated} \end{array} \) |
| Enrichment factor | \( EF = (C_i/C_{ref})(B_i/B_{ref}) \) | \( C_i \) — content element in soil; \( C_{ref} \) — content of Fe in sample; \( B_i \) — reference content of element; \( B_{ref} \) — reference content of Fe | \( \begin{array}{c} EF \leq 1 \\ 1 < EF \leq 3 \\ 3 < EF \leq 5 \\ 5 < EF \leq 10 \\ 10 < EF \leq 25 \\ 25 < EF \leq 50 \\ EF > 50 \end{array} \) | \( \begin{array}{c} \text{No enrichment} \\ \text{Minor enrichment} \\ \text{Moderate enrichment} \\ \text{Moderately severe enrichment} \\ \text{Severe enrichment} \\ \text{Very severe enrichment} \\ \text{Extremely severe enrichment} \end{array} \) |
| Contamination factor | \( CF = C_m/C_{ref} \) | \( C_m \) — element concentration; \( C_{ref} \) — reference value of element (background value) | \( \begin{array}{c} CF < 1 \\ 1 \leq CF < 3 \\ 3 \leq CF < 6 \\ 6 \leq CF \end{array} \) | \( \begin{array}{c} \text{Low contamination factor} \\ \text{Moderate contamination factor} \\ \text{Considerable contamination factor} \\ \text{Very high contamination factor} \end{array} \) |
| Contamination degree | \( C_{deg} = \sum_{i=1}^{n} CF \) | \( CF \) — contamination factor for element; \( n \) — number of elements | \( \begin{array}{c} C_{deg} < 8 \\ 8 \leq C_{deg} < 16 \\ 16 \leq C_{deg} < 32 \\ 32 \leq C_{deg} \end{array} \) | \( \begin{array}{c} \text{Low degree of contamination} \\ \text{Moderate degree of contamination} \\ \text{Considerable degree of contamination} \\ \text{Very high degree of contamination} \end{array} \) |
| Modified contamination degree | \( mC_{deg} = \frac{\sum_{i=1}^{n} CF}{n} \) | \( CF \) — contamination factor for element; \( n \) — number of elements | \( \begin{array}{c} mC_{deg} < 1.5 \\ 1.5 \leq mC_{deg} < 2 \\ 2 \leq mC_{deg} < 4 \\ 4 \leq mC_{deg} < 8 \\ 8 \leq mC_{deg} < 16 \\ 16 \leq mC_{deg} < 32 \\ 32 \leq mC_{deg} \end{array} \) | \( \begin{array}{c} \text{Nil to very low degree of contamination} \\ \text{Low degree of contamination} \\ \text{Moderate degree of contamination} \\ \text{High degree of contamination} \\ \text{Very high degree of contamination} \\ \text{Extremely high degree of contamination} \\ \text{Ultra high degree of contamination} \end{array} \) |
| Pollution load index | \( PLI = \sqrt{\prod_{1}^{n} PI} \) | \( PI \) — pollution index for element; \( n \) — number of elements | \( \begin{array}{c} PLI < 1 \\ PLI = 1 \\ PLI > 1 \end{array} \) | \( \begin{array}{c} \text{Unpolluted} \\ \text{Baseline level of pollution} \\ \text{Polluted} \end{array} \) |
| Improved Nemerow pollution index | \( PI_{N} = \sqrt{\left( \frac{\sum_{i=1}^{n} PI_{i}^2 + PI_{max}^2}{n} \right)} \) | \( PI \) — pollution index for element; \( max PI \) of all elements; \( n \) — number of elements | \( \begin{array}{c} PI_{N} < 0.7 \\ 0.7 \leq PI_{N} < 1.0 \\ 1.0 \leq PI_{N} < 2.0 \\ 2.0 \leq PI_{N} < 3.0 \\ PI_{N} > 3.0 \end{array} \) | \( \begin{array}{c} \text{Grade 1 — safety domain} \\ \text{Grade 2 — precaution domain} \\ \text{Grade 3 — slightly polluted domain} \\ \text{Grade 4 — moderately polluted domain} \\ \text{Grade 5 — seriously polluted domain} \end{array} \) |
Table 2 (continued)

| Index* | Formula | Explanation | Limit value | Classification |
|--------|---------|-------------|-------------|----------------|
| Potential ecological risk coefficient | $E_r = T_r \times C_{F_i}$ | $T_r$ — toxicity response coefficient of element; $C_{F_i}$ — contamination factor of single PHE | $E_r < 40$ | Low potential ecological risk |
| | | | $40 \leq E_r < 80$ | Moderate potential ecological risk |
| | | | $80 \leq E_r < 160$ | Considerable potential ecological risk |
| | | | $160 \leq E_r < 320$ | High potential ecological risk |
| | | | $320 \leq E_r$ | Very high potential ecological risk |
| Potential ecological risk index | $\sum_{i=1}^{n} E_{ri}$ | Potential ecological risk coefficient for single element; $n$ — number of analysed elements | $\text{PERI} < 150$ | Low potential ecological risk |
| PERI | $\text{RI} > 150$ | Moderate potential ecological risk |
| | $300 \leq \text{PERI} < 600$ | Considerable ecological risk |
| | $\text{RI} > 600$ | Very high ecological risk |
| Risk assessment code | Content of F1 | F1 — content of single elements in F1 | $%F1 < 1$ | No risk |
| RAC | | | $1 < %F1 < 10$ | Low risk |
| | | | $11 < %F1 < 30$ | Medium risk |
| | | | $31 < %F1 < 50$ | High risk |
| | | | $%F1 > 50$ | Very high risk |
| Individual contamination factor | $\text{ICF} = \frac{\text{PMF} \sum_{i=1}^{n} E_{ri}}{F1}$ | PMF — content in potential mobile fractions according to the BCR procedure | No limit values | The higher value the higher ecological risk |
| ICF | | | | | |
| Global contamination factor | $\text{GCF} = \sum \text{ICF}$ | ICF — individual contamination factor of single element | No limit values | The higher value the higher ecological risk |

*Sources: Gong et al. (2008); Kowalska et al. (2018); Kulbat and Sokolowska (2019); Gruszecka-Kosowska et al. (2019, 2020); Kumar et al. (2019a).

It is believed that horticulture with high cardamom cultivation is well adapted to the local soil conditions as a result of terraced fields that reduce erosion, the use of only organic fertilisers, and the presence of shade trees of the $N_2$-fixing Himalayan alder species (*Alnus nepalensis* D. Don), which often accompanies cardamom cultivation (Sharma et al. 2000; Singh et al. 2018). Data based on six rainfall events in the adjacent Sikkim Himalayas revealed only 1.86 and 0.41 kg ha$^{-1}$ year$^{-1}$ losses of TOC and N, respectively, under cardamom cultivation (Sharma et al. 2000). However, data indicate that at a yield of 230 kg ha$^{-1}$ year$^{-1}$ in the Darjeeling Himalayas (Gudade et al. 2013), natural fertilisation is unable to make up for organic carbon and nitrogen losses in this cropping system.

Soils used for rice cultivation had the lowest OM, TOC and N contents of all crops. The decline in soil OM and TOC is the combined effect of a reduction in organic matter being returned to the soil and tillage operations (Chaudhury et al. 2005). The rice yield of 2800 kg ha$^{-1}$ year$^{-1}$ in the Darjeeling Himalayas (Adhikari et al. 2011) contains 1.6% N of this mass in grains and straw, amounting to 45 kg ha$^{-1}$ year$^{-1}$ (Choudhury and Kennedy 2005). In addition, nitrogen depletion through processes of ammonia volatilisation and denitrification under submergence conditions (Choudhury et al. 1997), combined with low fertilisation, results in a significant reduction in the soil nitrogen content.

Organic carbon influences the uptake of nitrogen, as well as the content and rate of organic matter decomposition. The

Table 3  Physicochemical parameters of soils under different LULC

| Physical and chemical properties of soil | Forest | Tea | Horticulture | Rice |
|----------------------------------------|--------|-----|--------------|------|
| Sand % | 19 a | 24 a | 50 b | 47 b |
| Silt | 31 a | 25 a | 27 a | 29 a |
| Clay | 50 b | 50 a | 24 a | 24 a |
| OM | $10.7 \pm 0.47$ | $7.73 \pm 3.21$ | $5.38 \pm 1.76$ | $3.63 \pm 1.81$ |
| $pH_{KCl}$ | $4.73 \pm 0.37$ | $4.05 \pm 0.29$ | $4.41 \pm 0.48$ | $5.24 \pm 0.69$ |
| TOC g/kg | $44.5 \pm 27$ | $42.8 \pm 9.5$ | $20.8 \pm 9.3$ | $17.2 \pm 8.9$ |
| N | $3.33 \pm 1.70$ | $3.39 \pm 0.95$ | $1.91 \pm 0.75$ | $1.57 \pm 0.84$ |
| C:N | 12.8 b | 12.7 b | 10.6 a | 11.2 ab |

*Means followed by the different letters indicate significant differences at $\alpha \leq 0.05$ according to the t-Tukey test.
lower the C/N ratio, the more nitrogen higher plants can utilise. On the other hand, a high C/N ratio (>30) favours soil nitrogen protonisation and may result in nitrogen deficiency (Talgre et al. 2017). The optimum carbon-to-nitrogen ratio is from 8:1 to 10:1 in soils, 20:1 in manure and 15–20:1 in compost. In the tested soil samples, the C/N ratio ranged from 10.6 to 12.8. Significantly, the highest C/N ratio value was found in forest soils and in soils taken from tea cultivation, and the lowest in soils from under vegetables. In a study by Prokop and Plsokonka (2014), the C/N ratios of soils in the piedmont zone of the Darjeeling Himalayas were at a similar level; however, in cultivated soils (rice, tea), they were higher than in forest soils. The reason for higher values in cultivated soils may be lower land shaping and higher fertilisation. In the case of soils under rice, such as those in the Darjeeling Himalayas, the plots are terraced, but irrigated by rivers flowing from the Himalayas. These deposit sediment and nutrients with water, partially compensating for TOC and N losses due to cultivation.

### 3.2 Content of elements

The elemental content of soils depends on a number of both natural and anthropogenic factors. Since some of the elements are nutrients (macro- and micronutrients) for plants, the LULC and the type of cultivation affect their content in soils. The mode of agricultural use also influenced the content of elements in soils of the Peshok catchment (Table 4). Among the macronutrients, significantly, the highest Ca and Mg contents were found in soils from under horticulture and P in forest and tea soils. The higher Ca and Mg values in soils under horticulture may be related to lower runoff, soil erosion and leaching on terraces, as well as relatively low transport of macronutrients with the crop. In contrast, higher P values in tea and forest soils may be due to the fixation of phosphorus in these soils from bedrock and additionally to the use of phosphate in tea cultivation (Bisen and Singh 2012). On the other hand, the reduced P content may result from its higher availability in irrigated rice soils due to increased soil pH as well as relatively low phosphorus fertiliser rates (Choudhury et al. 2007). In the case of horticulture, the low P content of soils may be related to its intensive uptake by crops, as well as high removal with harvested cardamom capsules (Murugan et al. 2012).

The type of agricultural land use did not significantly affect the content of other analysed macronutrients (S, K and Na). Magnesium dominated in all types of land use in terms of quantity, and the content of macronutrients formed the following series: Mg > K > Ca > P > S > Na (forest); Mg > K >Ca > P > Na > S (tea, horticulture, rice). The small variation of S content in soils (not measured in C soil horizon by GSI 2021) is likely related to the occurrence of the copper sulphide deposits within the mica schists underlying the Peshok catchment (Bhattacharya et al. 1985). In the study conducted in the Uttarakhand Himalayas (Western Himalayas), the average macronutrient content in forest soil was at a higher level (except for Mg) and ranged from 1.764 to 3.00 g Na, 5.600 to 9.780 g Mg, 0.790 to 8.912 g P, 0.337 to 0.667 g S, 19.467 to 26.974 g K, and 5.703 to 11.051 g Ca/kg (Kumar et al. 2020). The lower macronutrient content in the Peshok catchment may be due to its much higher rainfall compared to the Uttarakhand Himalayas, where it averages only 1550 mm per year.

The trace element content significantly affects the quality of soils. Although some trace elements have important physiological functions, several (e.g. Cd, Pb, As) may pose a great threat to living organisms. Additionally, most trace elements in the literature are referred to as potentially toxic because they migrate easily in the soil–plant-human system (Kumar et al. 2019a; Ciarkowska and Gambus 2020).

The type of agricultural land use significantly affected the content of Mn, Ag, Ba, Cd, Co, Cu, Ga and Sr (Table 4). Comparison of micronutrient contents between forest soil and individual crops indicated that there were no statistically significant differences for tea. However, they were noted in the soils between the forest and terraced crops. Soils under horticulture had higher contents of Ag, Ba, Cd and Ga. In the case of rice cultivation, the differences were slightly more complex. Cultivation increased Cu and Ga contents and decreased Mn and Sr contents relative to the forest soil.

The mean content of most trace elements in the Peshok catchment was lower than for India natural soil background and India agricultural soil (Kumar et al. 2019b) (Table 4). Similarly, the mean contents of trace elements in forest soils near in the studied catchment were lower than their contents in forest soils in the Uttarakhand Himalayas (Kumar et al. 2020). Irrespective of use, trace element concentrations in soils of the Peshok catchment were below the maximum permissible level as stipulated by the Indian Prevention of Food Adulteration Act (Awasthi 2000), the United States Environmental Protection Agency (US EPA 2002) and the European Union Standard (EU 2002). This indicates the absence of trace element contamination in the studied soils.

The contents of most trace elements in the soils of the Peshok catchment were also lower than the local geochemical background value (GSI 2021). Only the contents of Ag and Cu in the soils (horticulture, rice), Cd (horticulture) and Ga (all land use types) exceeded the geochemical background values. The higher Ag content in soils from terraced crops is probably the result of the relatively low mobility of this element that tends to be retained in topsoils with pH values greater than 4, especially in soils with a high concentration of organic matter (Jacobson et al. 2005; Langdon et al. 2015). Significant sources of Cu and Cd in the soils are chemical fertilisers (such as superphosphates) and natural fertilisers (farmyard manure) as well as plant protection.
Table 4  Content of elements in soils (n=20)

| Elements | Forest (F) | Tea (T) | Horticulture (H) | Rice (R) | Background C soil horizon (GSI 2021) | India natural soil background (Kumar et al. 2019b) | India agricultural soil (Kumar et al. 2019b) | Uttarkhand Himalayas (Kumar et al. 2020) | Indian limits for soil (Awasthi 2000) | US EPA (2002) | European Union Standard (EU 2002) |
|----------|------------|--------|------------------|---------|------------------------------------|-----------------------------------------------|-----------------------------------------------|-----------------------------------------|--------------------------------------|--------------|-----------------------------|
| S        | 0.22±0.05 a** | 0.27±0.04 a  | 0.21±0.03 a      | 0.22±0.06 a  | 0.29±0.67                           | 5.70–11.05                                    | 67.35–54.23                                 | 5.60–9.78                              | 0.3–200                | 0.48 3.0                  |
| Ca       | 2.23±0.90 ab  | 1.11±0.43 a  | 2.79±1.19 b      | 2.19±0.30 ab  | 3.65                                | 5.70–11.05                                    | 19.47–26.97                                 | 3.0                                     | 75                      | 0.3–200                  |
| Fe       | 20.59±0.74 a  | 16.28±3.27 a | 16.04±1.94 a     | 20.12±4.05 a  | 31.30                               | 32.02                                        | 10.34                                        | 67.35–54.23                            | 0.3–200                | 0.48 3.0                  |
| K        | 3.88±0.78 a   | 4.09±1.23 a  | 5.57±0.81 a      | 5.59±1.64 a   | 23.30                                | 19.47–26.97                                 | 19.47–26.97                                 | 0.3–200                                | 1150                    | 0.3–200                  |
| Mg       | 12.97±3.52 ab | 7.88±0.83 a  | 17.60±7.33 b     | 15.51±5.40 ab | 9.98                                 | 5.60–9.78                                    | 19.47–26.97                                 | 171–201                               | 135–270 4300 140       |
| Mn       | 0.71±0.18 b   | 0.52±0.22 ab | 0.69±0.16 b      | 0.33±0.08 a   | 0.84                                 | 0.21                                         | 0.17                                         | 0.92–2.07                             | 1.75–3.40               | 0.88–8.91                |
| Na       | 0.20±0.02 a   | 0.29±0.09 a  | 0.24±0.09 a      | 0.27±0.06 a   | 5.08                                 | 1.75–3.40                                    | 1.75–3.40                                   | 0.88–8.91                             | 0.88–8.91               | 0.88–8.91                |
| P        | 1.12±0.73 b   | 1.14±0.17 b  | 0.51±0.38 ab     | 0.29±0.12 a   | 0.72                                 | 0.88                                         | 0.92                                         | 0.88–8.91                             | 0.88–8.91               | 0.88–8.91                |
| Ag       | 0.04±0.03 a   | 0.11±0.09 a  | 0.07             | 0.11±0.09 a   | 0.07                                 | 0.75                                         | 0.75                                        | 0.88–8.91                             | 0.88–8.91               | 0.88–8.91                |
| As       | 2.51±2.01 a   | 1.89±0.60 a  | 1.61±0.40 a      | 0.83±0.27 a   | 4.14                                 | 14.80                                        | 75                                          | 75                      | 0.3–200                  |
| Ba       | 79.15±21.58 a | 77.58±30.44 a| 167.5±47.1 b     | 134.4±37.5 a  | 601.71                               | 133–179                                      | 133–179                                     | 171–201                               | 135–270 4300 140       |
| Cd       | 0.03±0.01 a   | 0.04±0.02 ab | 0.03±0.03 a      | 0.03±0.01 a   | 0.07                                 | 1.56                                         | 120                                         | 135–270 4300 140       |
| Co       | 12.93±2.29 ab | 10.00±2.84 a | 10.47±1.62 ab    | 15.86±5.03 b  | 20.29                                | 15.20                                        | 8.53                                        | 0.3–200                               | 0.3–200                  |
| Cr       | 21.35±3.81 a  | 21.31±7.49 a | 23.84±2.93 a     | 28.62±6.77 a  | 87.71                                | 114.00                                       | 87.24                                       | 171–201                               | 135–270 4300 140       |
| Cu       | 13.93±3.62 a  | 14.97±4.86 a | 36.86±22.91 b    | 41.55±10.99 b | 27.57                                | 56.50                                        | 48.70                                       | 171–201                               | 135–270 4300 140       |
| Ga       | 102.06±15.05 a| 126.53±53.27 ab| 172.66±32.51 b | 185.37±35.80 b| 22.71                                | 133–179                                      | 133–179                                     | 171–201                               | 135–270 4300 140       |
| Li       | 13.42±2.38 a  | 14.69±3.89 a | 18.58±2.99 a     | 16.96±4.55 a  | 33.71                                | 133–179                                      | 133–179                                     | 171–201                               | 135–270 4300 140       |
| Mo       | Bd a         | 0.07±0.04 a  | 0.08±0.10 a      | 0.03±0.04 a   | 2.09                                 | 2.09                                         | 2.09                                        | 0.88–8.91                             | 0.88–8.91               | 0.88–8.91                |
| Ni       | 21.78±4.90 a  | 19.14±6.43 a | 17.20±4.56 a     | 25.10±8.19 a  | 32.14                                | 27.70                                        | 92.94                                       | 75–150                                | 75–150                   |
| Pb       | 10.50±2.65 a  | 9.23±2.85 a  | 11.08±3.65 a     | 8.50±3.65 a   | 18.86                                | 13.10                                        | 15.71                                       | 150                                   | 150                     |
| Sr       | 10.79±4.57 b  | 7.91±2.28 ab | 7.65±2.87 b      | 4.82±1.23 a   | 56.71                                | 215–353                                      | 143–197                                     | 143–197                               | 143–197                 |
| Zn       | 74.16±10.37 a | 56.26±15.39 a| 45.62±17.46 a    | 63.24±18.93 a | 87.14                                | 22.10                                        | 76.24                                       | 300–600                               | 300–600                 |
| Series   | Fe > Mn > Ga > Ba > Zn > Ni > Co > Cr > Cu > Li > As > Cd > Mo > Ag | Fe > Mn > Ga > Ba > Zn > Cu > Cr > Li > Pb > Pb > Sr > As > Ag > Mo > Ag > Cd | Fe > Mn > Ga > Ba > Zn > Cu | Fe > Mn > Ga > Ba > Zn > Cu | Fe > Mn > Ga > Ba > Zn > Cu | Fe > Mn > Ga > Ba > Zn > Cu | Fe > Mn > Ga > Ba > Zn > Cu | Fe > Mn > Ga > Ba > Zn > Cu | Fe > Mn > Ga > Ba > Zn > Cu | Fe > Mn > Ga > Ba > Zn > Cu |

*Below the detection limit; ** means followed by the different letters indicate significant differences at $\alpha \leq 0.05$ according to the $i$-Tukey test.
products (Acosta et al. 2011; Kumar et al. 2019b; Suhr et al. 2021). In the Peshok catchment, most farmers use cow dung compost, cattle manure and urea for horticulture and rice cultivation. Therefore, it can be assumed that cultivation, unbalanced fertilisation and intensive waste (leaching) in the studied catchment of the Darjeeling Himalayas significantly affect the soil nutrient depletion.

The gallium content in the soils of the Peshok catchment may be of some concern. Gallium is a component of all soils; however, it is relatively rarely analysed. The average gallium content in soils is 5–300 mg/kg (most commonly 5–70 mg kg⁻¹) (Buhl and Poedniok 2000). In the natural environment, gallium accompanies Cu and Al ores and is also associated with feldspars, amphiboles and micas. Elevated gallium concentrations in soils are identified in mining sites, used electronic equipment and wastewater (Sharma et al. 2017; Liu et al. 2021). There are no industrial sources of gallium in or around the Peshok catchment, but its relatively high values are found in soils from the natural forest. This is probably associated with the sphalerite (Al ore) identified in this catchment, as well as micas common in gneisses and schists (Bhattacharya et al. 1985). Significantly higher concentrations of gallium in crop soils compared to forest soil in this catchment may be related to the tendency of gallium to accumulate in acidic soils (Syu et al. 2020), and also to the low mobility of Ga in the soil–plant system compared to other common trace element contaminants, such as Cd, Cu and Zn (Jensen et al. 2018).

In order to present the relationships between the studied physicochemical parameters of soils, a principal component analysis was performed (Table 5). Four principal components were identified in the analysis, explaining 73.7% of the total variance of the data set. The first factor (PC1) explained 29.4% of the total variance; its main components were the clay fraction, organic matter content, and TOC, N, S, P and Mn contents (positive charges > 0.50) as well as the sand fraction and Cu, Ga, Sr and Ga contents (negative charges > 0.50). The second factor (PC2) accounted for 21.7% of the total variance and was dominated by the dust fraction and Fe, K, Na, Co, Cr, Li and Zn contents. The third factor (PC3) explained 13.8% of the total variance and was related to the contents of Ca, Mg, Mn, Ag, Ba, Cd, Cu and Li. The fourth factor (PC4) explained only 8.8% and was related to pH and the contents of Cd, Li, and Mo (Table 5).

In the study, PC1, PC2 and PC3 represent natural factors (related to soil properties) determining the content and sources of elements. PC1 indicated that the clay fraction and the contents of organic matter and TOC had a significant effect on the content of biogenic (fertiliser) elements, i.e. N, S and P; micronutrients, i.e. Mn, Zn and Cu; and the trace elements Sr and Ga. A positive relation was also demonstrated (PC2, Table 5) between the content of silt and Fe fractions, and the content of K, Na and most trace elements (significant for Co, Cr, Li, Ni, Zn), which may suggest that they were of natural origin. The study also showed that the presence of correlations between iron content and other trace elements might indicate their natural sources (Baran and Wieczorek 2015; Gruszecka-Kosowska et al. 2020). PC results (PC1, PC2, PC3) also indicated the presence of significantly positive correlations between elements (Table 5). This is linked to their similar sources and distribution routes, which also indicate their natural origin. Only in the case of Ga and Cu contents can it be assumed that there are additional factors or sources, such as local geology or fertilisation, influencing them (Bhattacharya et al. 1985; Kumar et al. 2019b). Finally, PC4 revealed that pH affected the content of the analysed parameters. Most of the studied soils were acidic or very acidic, which may be caused by both

Table 5 Component matrix for variables (n = 20)

| Variables | PC1   | PC2   | PC3   | PC4   |
|-----------|-------|-------|-------|-------|
| Sand      | −0.807| −0.315| 0.251 | 0.043 |
| Silt      | −0.077| 0.630 | −0.012| −0.273|
| Clay      | 0.884 | 0.135 | −0.264| 0.042 |
| OM        | 0.766 | −0.050| −0.020| −0.203|
| pH        | −0.233| 0.150 | −0.120| −0.527|
| TOC       | 0.925 | 0.015 | −0.055| 0.030 |
| N         | 0.936 | 0.098 | 0.028 | 0.110 |
| S         | 0.627 | 0.391 | 0.097 | 0.287 |
| Ca        | 0.052 | 0.005 | 0.759 | 0.022 |
| Fe        | 0.194 | 0.893 | −0.087| −0.312|
| K         | −0.402| 0.619 | 0.435 | 0.142 |
| Mg        | −0.263| 0.146 | 0.746 | −0.042|
| Mn        | 0.551 | −0.234| 0.558 | 0.324 |
| Na        | 0.071 | 0.603 | 0.000 | 0.439 |
| P         | 0.920 | −0.078| −0.136| 0.123 |
| Ag        | −0.442| −0.386| 0.747 | 0.429 |
| As        | 0.193 | 0.058 | −0.165| 0.236 |
| Ba        | −0.390| 0.231 | 0.834 | 0.290 |
| Cd        | 0.218 | −0.110| 0.582 | 0.788 |
| Co        | −0.116| 0.963 | −0.039| −0.263|
| Cr        | −0.066| 0.796 | 0.376 | 0.195 |
| Cu        | −0.616| 0.302 | 0.548 | 0.052 |
| Ga        | −0.543| 0.455 | 0.483 | 0.136 |
| Li        | −0.024| 0.558 | 0.692 | 0.569 |
| Mo        | 0.038 | −0.273| 0.257 | 0.676 |
| Ni        | 0.141 | 0.958 | −0.055| −0.038|
| Pb        | 0.059 | −0.025| 0.368 | 0.362 |
| Sr        | 0.821 | −0.054| 0.224 | 0.123 |
| Zn        | 0.501 | 0.643 | −0.317| −0.148|
| Total variance % | 29.4 | 21.7 | 13.8 | 8.8 |
| Cumulative variance % | 29.4 | 51.1 | 64.9 | 73.2 |

Loadings > 0.5 are shown in bold
natural and anthropogenic factors. Soil pH affects the mobility of trace elements in soil. Under pH conditions below 5.5, trace element availability increases in acidic and very acidic soils, and excessive aluminium activity can occur, leading to a reduction in soil fertility (Kim et al. 2015; Neina 2019). In the investigated soils, pH may particularly affect the mobility of Cd, Li and Mo (PC4).

### 3.3 Soil quality indicators

#### 3.3.1 Geochemical indicators

Geochemical indicators are used to quantify the degree of anthropogenic impact on the contamination of soil by trace elements (Kowalska et al. 2018). The geoaccumulation index (Igeo) results indicated a contamination class of 0 (practically uncontaminated) for most of the analysed elements. Only Ag, for horticulture, and Ga, for all LULC types, were categorised as contamination class I (uncontaminated to moderately contaminated), as shown in Table 6.

Depending on the element and the type of land use, the enrichment factor (EF) values varied from 0 to 15 (Table 6). However, for most trace elements (except As and Cd), lower EF values were obtained for forest soils, and higher values for cultivated soils. The forest soils scored EF ≤ 1, which indicated their lack of enrichment with trace elements and their natural origin. The only exception was Ga, for which the EF value in forest soils was 7, indicating moderately severe enrichment. In soils from tea cultivation, the findings of EF ≤ 1 for most elements were similar to those of natural forest soils. Minor enrichment was observed only for Cd and Ni and severe enrichment only for Ga. Contrastingly, more elements were enriched in soils under terraced crops. The horticulture soils showed minor enrichment with Cd, Cu, Li and Pb, and severe enrichment with Ag and Ga. Soils from rice cultivation showed minor enrichment with Ag, Co, Cu and Ni and severe enrichment with Ga (Table 6).

The CF values ranged from 0.1 to 8.2. As above, the highest indicator value, irrespective of land use, was found for Ga; the results identified considerable contamination (forest and tea) and very high contamination (horticulture and rice) with this element. Values in the range 1 ≤ CF < 3 were recorded for Ag (forest, horticulture and rice), Cu (horticulture and rice) and Zn (forest), indicating moderate contamination with these metals. For the remaining elements, the CF values were below 1, indicating that their contents in the soil were caused by natural factors (Table 6).

The values of multi-element indicators (Igeo, EF, CF, Cdeg, mCdeg, PLI, PLN) were low, suggesting no, low or medium degrees of soil contamination with trace elements (Table 6). It is worth mentioning that statistically significant correlations were found between the calculated multi-element indicators (0.85 ≤ r ≤ 0.89, p < 0.05). Furthermore, higher values of these indicators were found for soils taken from horticulture and rice crops than for forest soils and tea soils. Based on various geochemical indicators, Kumar et al. (2020) found that forest soils in the Uttarakhand Himalayas of India were moderately to highly contaminated with Rb, Zn, Cr, Cu and Ni, and obtained the Cdeg value of 36.66. Those authors suggested that such contamination was associated with high anthropogenic activity in the vicinity of the study area.

It is worth mentioning that in the Peshok catchment, the Cdeg value ranged from 7.4 (tea) to 14 (horticulture), as

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**Table 6** Single and total complex geochemical contamination indicators of soils

| Elements | Igeo | EF | CF | Cdeg | mCdeg | PLI | PLN |
|----------|------|----|----|------|-------|-----|-----|
| Ag       | 0    | 1  | 0  | 9    | 1     | 0.6 | 0.4 |
| As       | 0    | 0.8| 0  | 0.8  | 0.8   | 0.5 | 0.3 |
| Ba       | 0    | 0.1| 0  | 0.8  | 0.8   | 0.6 | 0.4 |
| Cd       | 0    | 0.7| 0  | 0.7  | 0.7   | 0.6 | 0.4 |
| Co       | 0    | 1  | 0  | 1    | 1     | 0.5 | 0.3 |
| Cr       | 0    | 0.4| 0  | 0.4  | 0.4   | 0.6 | 0.4 |
| Cu       | 0    | 0.8| 0  | 0.8  | 0.8   | 0.6 | 0.4 |
| Ga       | 1    | 7  | 0  | 7    | 7     | 0.5 | 0.3 |
| Li       | 0    | 0.6| 0  | 0.6  | 0.6   | 0.6 | 0.4 |
| Mo       | 0    | 0  | 0  | 0    | 0     | 0.5 | 0.3 |
| Ni       | 0    | 0.1| 0  | 0.1  | 0.1   | 0.5 | 0.3 |
| Pb       | 0    | 0  | 0  | 0    | 0     | 0.5 | 0.3 |
| Sr       | 0    | 0  | 0  | 0    | 0     | 0.5 | 0.3 |
| Zn       | 0    | 0  | 0  | 0    | 0     | 0.5 | 0.3 |

*a, t, h, r: forest, tea, horticulture, rice; Igeo: geoaccumulation index; EF: enrichment factor; CF: contamination factor; Cdeg: contamination degree; mCdeg: modified contamination degree; PLI: pollution load index; PLN: improved Nemerow pollution index
shown in Table 6. Geochemical indicators are used to quantify the anthropogenic impact on the degree of soil contamination with trace elements (Kowalska et al. 2018). The Igeo values indicated pollution class 0 — virtually unpolluted for most of the analysed elements. Only Ag for horticulture and Ga under all LULC types had contamination class I (uncontaminated to moderately contaminated) (Table 6).

The Ef, depending on the element and the type of use, took the value from 0 to 15 (Table 6). However, lower values of this index for most trace elements (except As and Cd) were obtained for forest soils, and higher values for soils under cultivation. The forest soils were characterized by the value of EF ≤ 1, which indicates the lack of enrichment with trace elements and their natural origin. Only for Ga, the value of the EF in these soils was 7, which indicates moderately severe enrichment. In the soils from tea cultivation, for most of the elements, the value of EF ≤ 1 was similar to the soils under the natural forest. Minor enrichment was only accepted for Cd and Ni and severe enrichment for Ga. More elements were enriched in the soils under the controlled crops. Minor enrichment Cd, Cu, Li and Pb and severe enrichment Ag and Ga were found in the soils under horticulture cultivation. Minor enrichment Ag, Co, Cu and Ni and severe enrichment Ga were found in soils under rice cultivation (Table 6).

### 3.3.2 Ecological indicators

The Er values (Er < 40) for As, Cd, Cr, Cu, Ni and Zn indicated low potential ecological risk associated with the presence of individual elements in the studied soils (Table 7). In general, the mean PERI value for trace elements in the studied soils was between 28.3 and 48.4 and showed low risk (PERI < 150). The highest PERI value was determined for soils taken from horticulture crops, and the lowest for forest soils and soils taken from rice crops. The potential ecological risk coefficients (mean ER values) were compared for individual elements, and the Er values formed a series: Cd > Cu > As > N and > Pb > Zn > Cr. In a study by Kumar et al. (2020), the ER value of each element (Cr, Mn, Ni, Cu, Zn) was below 40 and PERI was 75.56, indicating, as in our study, the low ecological risk associated with trace elements in forest soils of the Uttarakhand Himalayas.

### 3.3.3 Ecotoxicity of soils

Depending on the type of soil use, the inhibition of *A. fischeri* luminescence ranged from 71 to 89% (Table 7). Significantly higher bacteria luminescence inhibition was found in forest soils and soils collected from tea crops compared to soils from horticulture and rice crops. However, regardless of the type of land use, the tested soils were toxic (50% ≤ PE < 100%) to the test organism. In the study by Baran and Wieczorek (2015), as in the present study, by far the highest toxicity to the bacteria was found in soils collected from forested areas, with *A. fischeri* luminescence inhibition ranging from −33 to 59% (cultivated soils), −48 to 78% (grassland), and 0 to 88% (forest soils). Using the Microtox test to assess the toxicity of soils contaminated by mining, Loureiro et al. (2005) found that *A. fischeri* was more sensitive to extracts prepared from soils with low trace element contents than extracts from soil with high trace element content. In the Peshok catchment, correlation analysis showed a significantly

| LULC n=20 | Er | PERI | Alivibrio fischeri |
|-----------|----|------|-------------------|
|           |    |      |                   |
| Forest    |    |      |                   |
| Tea       |    |      |                   |
| Horticulture |    |      |                   |
| Rice      |    |      |                   |

### Table 7 Indices of chemical and ecological risk of soils

In the table, the LULC (Land Use Land Cover) column indicates the type of land use, and the Er column shows the potential ecological risk coefficient. The PERI column indicates the potential ecological risk index, and the Alivibrio fischeri column indicates the inhibition of luminescence of *A. fischeri*. The ICF column represents the individual contamination factor, and the GCF column represents the global contamination factor. The RAC column indicates the risk assessment code. Means followed by different letters indicate significant differences at α ≤ 0.05 according to the t-Tukey test; risk: no, low, moderate, high, very high, respectively.

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positive relationship between TOC content (r = 0.58, p < 0.05), clay fraction (r = 0.71, p < 0.05), a significantly negative relationship with pH (r = −0.41, p < 0.05) and most of the analysed trace elements (−0.63 ≤ r ≤ −0.08, p < 0.05), (Table 8). Significant correlations were found for the following: r = −0.57 for Ba, r = −0.52 for Ag, r = −0.63 for Cu, r = −0.56 for Ga, and the A. fischeri response. Therefore, trace element content did not affect the ecotoxicity of soils to bacteria. It is worth mentioning that bioassays indicate the general, or total, toxicity of samples, but they do not identify the cause of the observed toxic responses (Heise et al. 2020). Hence, lack, weak or non-significant correlations between elemental content in soils and the response of organisms suggest that there are other factors influencing toxicity. Of the factors analysed, TOC content, clay fraction and pH had a significant effect. It is likely that the acidic pH played a large part in the toxicity of soils to bacteria. A negative correlation was observed between A. fischeri luminescence inhibition and pH, indicating that soil pH toxicity increased with decreasing pH value. In contrast, TOC and clay fraction have a large sorption surface area and can accumulate other contaminants of agricultural origin (e.g. pesticide residues).

### 3.3.4 Mobility of trace elements

Total trace element contents in soils do not always provide a basis for assessing the ecological risks associated with the presence of trace elements in the environment. It is believed that the identification of their forms gives more reliable information on the potential effects on living organisms, while information on the mobile fraction of trace elements is particularly valuable (Khadhar et al. 2020). The share of selected elements in individual fractions is shown in Fig. 2. In the case of Cr, Cu, Ni and Zn, irrespective of the type of agricultural land use, the residual fraction (F4) was dominant, accounting for 78% (tea) to 85% Cr (rice); 87% (tea) to 95% Cu (horticulture); 86% (tea) to 92% Ni (horticulture); and 73% (rice) to 76% Zn (horticulture), respectively. Cadmium was generally bound to the exchangeable fraction (F1) and the reducible fraction (F2), while lead was bound to the reducible fraction (F2) and the oxidisable fraction (F3). The percentages of Cd in the F1 and F2 fractions according to the type of use were 73% and 18% (rice), 34% and 51% (horticulture), 29% and 33% (tea), and 27% and 45% (forest). The percentages of Pb in the F2 and F3 fractions were 48% and 25% (rice), 68% and 28% (horticulture), 48% and

**Table 8** Pearson correlations between the analysed parameters

| Parameter | Ca | Mg | Al | Sr | Ni | Zn | Cd | Cr | Cu | Mn | Fe | K | Mg | Fe | Mg | Fe |
|-----------|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|
| Ca        | 1.00 | 0.30 | 0.33 | 0.22 | 0.21 | 0.19 | 0.21 | 0.21 | 0.19 | 0.21 | 0.21 | 0.19 | 0.21 | 0.21 | 0.21 |
| Mg        | 0.30 | 1.00 | 0.30 | 0.20 | 0.17 | 0.16 | 0.17 | 0.15 | 0.15 | 0.15 | 0.15 | 0.13 | 0.15 | 0.15 | 0.15 |
| Al        | 0.33 | 0.30 | 1.00 | 0.20 | 0.17 | 0.16 | 0.17 | 0.15 | 0.15 | 0.15 | 0.15 | 0.13 | 0.15 | 0.15 | 0.15 |
| Sr        | 0.22 | 0.20 | 0.20 | 1.00 | 0.30 | 0.20 | 0.20 | 0.20 | 0.20 | 0.20 | 0.20 | 0.20 | 0.20 | 0.20 | 0.20 |
| Ni        | 0.21 | 0.17 | 0.17 | 0.30 | 1.00 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 |
| Zn        | 0.19 | 0.15 | 0.15 | 0.15 | 0.15 | 1.00 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 |
| Cd        | 0.21 | 0.19 | 0.19 | 0.19 | 0.19 | 0.30 | 1.00 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 |
| Cr        | 0.21 | 0.19 | 0.19 | 0.19 | 0.19 | 0.30 | 0.30 | 1.00 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 |
| Cu        | 0.19 | 0.15 | 0.15 | 0.15 | 0.15 | 0.30 | 0.30 | 0.30 | 1.00 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 |
| Mn        | 0.19 | 0.15 | 0.15 | 0.15 | 0.15 | 0.30 | 0.30 | 0.30 | 0.30 | 1.00 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 |
| Fe        | 0.19 | 0.15 | 0.15 | 0.15 | 0.15 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 1.00 | 0.30 | 0.30 | 0.30 | 0.30 |
| K         | 0.19 | 0.15 | 0.15 | 0.15 | 0.15 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 1.00 | 0.30 | 0.30 | 0.30 |
| Mg        | 0.19 | 0.15 | 0.15 | 0.15 | 0.15 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 1.00 | 0.30 | 0.30 |
| Fe        | 0.19 | 0.15 | 0.15 | 0.15 | 0.15 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 1.00 | 0.30 |
| Mg        | 0.19 | 0.15 | 0.15 | 0.15 | 0.15 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 1.00 |
| Fe        | 0.19 | 0.15 | 0.15 | 0.15 | 0.15 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 0.30 | 1.00 |

**Bold indicates significant differences at p < 0.05**
49% (tea), and 40% and 47%, respectively. In summary, the metal contents in the different fractions were as follows. Cd: F1 > F2 > F3 > F4; Cr, Cu, Ni and Zn: F4 > F3 > F2 > F1; and Pb: F2 > F3 > F4 > F1. In the literature, trace elements in the F1 fraction are considered more mobile and more easily bioavailable, thus posing a high ecological risk (Gruszecka-Kosowska et al. 2019).

The RAC classification, which takes into account the presence of metal in the F1 fraction, is presented in Table 7. In our study, a significant risk was found only for Cd, which posed a medium risk in forest soils, and high and very high risks in horticulture and rice soils, respectively. The study also used the ICF and GCF indicators, which account for the content of elements in all analysed fractions. The calculated ICF values were highest for Pb and lowest for Cd and Cu (Table 7). The ICF values describing the risk associated with the mobility of the fraction of potentially mobile elements (∑F1–3) can be ranked in descending order: Pb > Zn > Cd > Cr > Ni > Cu for forest soils; Pb > Cr > Zn > Ni > Cu > Cd for tea soils; Pb > Zn > Cr > Ni > Cd > Cu for horticulture soils; and Pb > Zn > Cr > Cu > Ni > Cd for rice soils. The GCF values ranged from 4.99 to 9.10, with the highest ecological risk associated with mobile forms of trace elements found in horticulture soils, and the lowest in rice soils (horticulture > forest > tea > rice) (Table 7). Risk analysis is important because

Fig. 2 BCR binding forms of trace elements in soils
of the easy movement of metals in trophic chains (soil–plant-human) (Gruszeczk-Kosowska et al. 2019; Ciarkowska and Gambus 2020). This is of particular importance due to the low pH values of soils in the Peshok catchment. It is well known that the mobility and potential bioavailability of some elements increase at low pH values. However, it is important to bear in mind that despite the significant effect of pH on the mobility of trace elements, the total content of trace elements generally has the greatest impact on their availability to organisms (Baran and Wieczorek 2015). Our study showed rather low trace element concentrations, and the contents of most elements in the soils were at or below the geochemical background value in the Darjeeling Himalayas.

4 Conclusions

Soils in the catchment located in the Darjeeling Himalayas (Eastern Himalayas) are characterised by low macro- and micronutrient contents, both in natural forests and under various forms of agricultural use. These contents are generally lower compared to India background soils, averages for agricultural soils in India, forest soils in the Western Himalayas and the local geochemical background. This may support the hypothesis that natural factors, such as the catchment’s high precipitation (the highest in the southern slope of the Himalayas) and steep slopes, favour the strong leaching of elements from the soil, and farming systems based on natural and artificial fertilisation, terracing and irrigation are not able to balance them fully. Cultivation systems clearly differentiate the elemental content of topsoil with increasing environmental interference (deforestation, terracing, irrigation). There is a lack of statistically significant differences in elemental content in soils under tea cultivation compared to natural forest soils. The relatively dense cover of tea bushes and shade trees provide the best protection for soil against erosion and leaching of elements.

Both the indicators used and the results of statistical analyses suggested a rather natural origin of elements. The higher Ga content indicated a relationship with the local geology and the higher Cu content with the use of fertilisers. Indicators showed a low ecological risk related to the presence of trace elements and P, and soil ecotoxicity to A. fischeri was generally determined by the acidic pH of the tested soils. Low mobility was observed for most elements, and regardless of the land use, the residual fraction predominated. Only for Cd were medium (forest), high (horticulture) and very high (rice) risks related to mobility and bioavailability shown. However, the low total Cd content in soils in the Darjeeling Himalayas region suggested a low risk associated with the mobility of this element.

Among the proposed indicators for assessing the degree of soil contamination with trace elements, 13 were used in this study, including 7 geochemical indicators, 2 indicators assessing potential ecological risk, 3 indicators assessing the mobility of elements and 1 indicator assessing soil ecotoxicity. The obtained results made it difficult to clearly indicate which of the indices used were more sensitive. Therefore, especially in the case of a low content of trace elements, the use of many indicators can more fully describe soil quality in relation to land use.

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Declarations

Conflict of interest The authors declare no competing interests, and the manuscript is approved by all the authors for publication. The authors would like to declare that the work described was original research that has not been published previously, and not under consideration for publication elsewhere, in whole or in part.

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