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Mineralogy and Concentration of Potentially Toxic Elements in Soils of the São Francisco Sedimentary Basin

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ABSTRACT: Lithological and geomorphological variations determine formation of soils with different mineralogical constitutions and can influence the distribution of potentially toxic elements (PTE). The aim of this study was to determine the contents of PTE and mineralogical assemblages along soil profiles of different classes and parent materials and to investigate the association among these factors in the São Francisco sedimentary basin in the northern part of the state of Minas Gerais (Brazil). Eight soil pedons of different classes that developed from different parent materials (limestone, siltstone, phyllite, alluvial-colluvial sediments, detrital cover, and rhythmite) were studied in two lithotoposequences. Soil morphological, particle-size, and chemical analyses were carried out for soil characterization and classification. A mineralogical investigation was carried out by X-ray diffraction on soil sand, silt, and clay fractions. Potentially toxic elements were extracted by microwave-assisted acid digestion (EPA 3015A method) and determined in an ICP-OES. Soil mineralogy showed relevant interactions between pedogenesis and morphogenesis. Quartz is the main mineral in the sand and silt fractions, with varying amounts of weatherable primary minerals, whereas kaolinite is the main mineral in the clay fraction, followed by 2:1 clay minerals and oxides. The main soil properties associated with PTE variability were pH, redox environment, mineral and organic reactive surfaces, and clay content. The unique patterns of PTE distribution per pedon and differences in PTE contents in soils from the same parent material, but with different pedogenic evolution, showed that pedogenic processes influence PTE distribution. Mainly Mn, Ni, Pb, and Ba were influenced by parent material, especially limestone, siltstone, and detrital cover, but Cd, As, and Cu were also influenced. To a lesser extent, Mo was influenced by phyllite and Cr by detrital cover and phyllite. Rhodic Ferralsol (*Latossolo Vermelho Distrófico típico*) was the only taxonomic class to show class association with PTE. The association of Ferralsols (*Latossolos*) with Cd, Mn, Ni, Pb, Ba, As, and Cu and the low contents of these elements in soils of this class suggest that pedogenic processes common to Ferralsol (*Latossolo*) evolution are responsible for decreasing levels of these PTE.

Keywords: parent material, pedogenesis, soil minerals, trace elements.

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INTRODUCTION

The contents of potentially toxic elements (PTE) in soils vary with pedogenic and anthropogenic sources, among other factors (Kabata-Pendias, 2010). In soils with little anthropic influence, parent material is the main source of PTE (Alloway, 1995; Kabata-Pendias, 2010) since these elements are constituents of primary minerals (Oliveira et al., 1999; Biondi et al., 2011). Weathering and pedogenic processes contribute to reallocation of PTE from parent material throughout the soil profile and landscape (Burak et al., 2010), thus contributing to their three-dimensional variability.

Soils from sedimentary rocks tend to have low contents of PTE (Alloway, 1995; Kabata-Pendias, 2010; Hugen et al., 2013). However, such contents may vary with soil particle-size distribution and are directly associated with soil clay contents (Alloway, 1995) and clay mineralogy (Biondi et al., 2011).

The microregion of Montes Claros, in the north of the state of Minas Gerais (Brazil), is in the São Francisco Craton. This geology is part of the almost flat, sub-horizontal cover, little deformed on a regional scale, that characterizes the São Francisco River Valley. In this region, Neoproterozoic sedimentary rocks of the Bambuí Group are widely distributed, mainly as siltstones, rhythmites, phyllites, shales, and carbonate rocks. The exposure of these rocks was affected by Cretaceous and Tertiary tectonic reactivation that modeled the regional drainage system, with subsequent formation of eluvial-colluvial detrital cover and fluvial plain sediments (CPRM, 2004; Martinéz, 2007). These factors determine the present soil diversity and distribution. In this region, soil formation and association with potentially toxic elements are relevant due to the large area they represent and to the unique interactions of these elements to rocks and pedogenic processes.

Assuming that there is a relationship of soil parent material and degree of soil development with PTE distribution, the aim of this study was to determine PTE content and mineralogical assemblages in pedons of different soil classes and parent materials and investigate the association among these factors in a sedimentary basin in the north of Minas Gerais, Brazil.

MATERIALS AND METHODS

The studied area is located in the microregion of Montes Claros and encompasses part of the municipalities of Montes Claros, Capitão Enéas, Francisco Sá, and São João da Ponte in the north of Minas Gerais, Brazil (Figure 1). The local climate is classified as Aw in the Köppen system, tropical savanna with dry winter, annual average rainfall and temperature of 1,060 mm and 24.2 °C, respectively.

Local native vegetation is typical of the *Cerrado* (Brazilian tropical savanna) - *Caatinga* biome transition, with predominance of *Carrasco* (Deciduous, high, closed, and unstratified shrubland) and Deciduous Seasonal Forest (Tropical Dry Forest). In the north of the area, a flat to gently rolling relief predominates, whereas to the south, gently to strongly rolling landforms are most common. The predominant lithology of the study area is sedimentary and metasedimentary rocks of the Bambuí Group, together with Cenozoic cover (alluvial, colluvial, and elluvial) (CPRM, 2004). Geomorphologically, the area is within the São Francisco Sedimentary Basin, where planation landscape surfaces and residual plateaus predominate (Leite and Brito, 2012).

In an area of very little anthropic influence, eight soil pedons, typical of the regional lithology, were collected under native vegetation, except for two pedons (T2P3 and T2P4), which were collected under pasture. The pedons were at different relief positions, characterizing two lithotoposequences. Lithotoposequence 1 (T1) comprised three pedons from elevations of 509 to 615 m (Figure 2a). Pedons T1P1 and T1P2 derived from carbonate rocks, whereas T1P3 came from colluvial-alluvial sediments. Lithotoposequence 2 (T2) comprised five pedons from elevations of 559 to 648 m (Figure 2b), where parent materials are siltstone (T2P1), alluvial sediments (T2P2), phyllite (T2P3), detrital cover (T2P4), and rhythmite (T2P5).

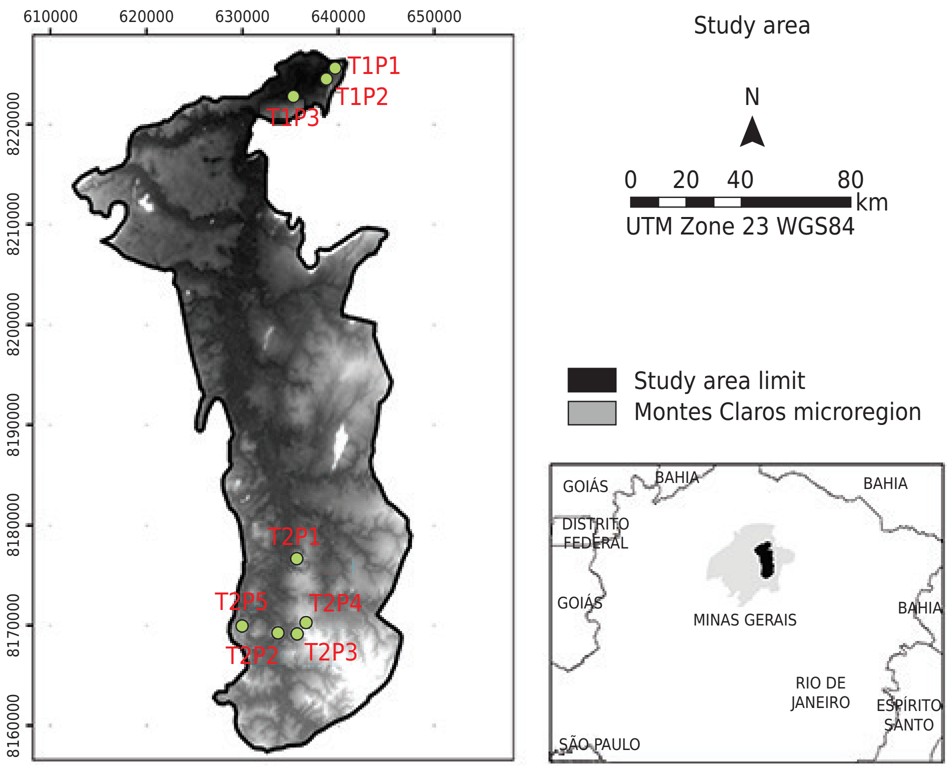

Figure 1. Location of the study area in the Montes Claros microregion (MG, Brazil).

Morphological description and sampling of the profiles followed the approach proposed by Santos et al. (2005), and soils were classified according to the Brazilian Soil Classification System - SiBCS (Santos et al., 2013) and to the World Reference Base for Soil Resources (WRB, 2014). Soil samples were air dried, crushed, and passed through a 2-mm mesh sieve. Then, particle-size analysis was carried out by the pipette method, with chemical dispersion and mechanical stirring (Claessen, 1997). All chemical characterization was performed according to Claessen (1997). Soil pH was determined in water. The exchangeable cations Ca, Mg, Na, K, Al, and H were determined, which served as the basis for calculating the sum of bases (SB = Ca + Mg + Na + K) and cation exchange capacity at pH 7 (CEC = SB + Al^{3+} + H^+). Total organic carbon was also determined.

The samples were subjected to sulfuric acid attack, and the Ki and Kr indexes were calculated for soil classification (Santos et al., 2013). Mineralogical analyses were made by X-ray diffraction (XRD) for the sand, silt, and clay fractions. The sand and silt fractions were irradiated as powder, and clay as a slurry, before and after iron extraction with sodium dithionite-citrate-bicarbonate (DCB) (Mehra and Jackson, 1958). The clay fraction was subjected to the following treatments: saturation with potassium with irradiation at 25 °C and after heating at 350 and 550 °C; and irradiation after saturation with magnesium with and without ethylene glycol saturation. The XRD patterns were determined on a Rigaku diffractometer (Miniflex II model) with Cu tubes as a Kα radiation source (30 kV and 15 mA), and a graphite monochromator. The readings were made at a scanning speed of 0.050° s^{-1}, from 3 to 50° 2θ.

Arsenic, Ba, Cd, Cr, Cu, Mn, Mo, Ni, and Pb were extracted and determined according to the EPA 3051A method (Usepa, 2007), with samples in duplicate. Quality control analysis was performed using a standard reference material NIST 2711 (Montana II soil), which was introduced for each set of 24 analyzed samples, and recovery rates were calculated. Extract samples were read in an inductively coupled plasma-atomic emission spectrometer (ICP-AES), after calibration with multi-element standard curves.

Principal component analysis (PCA) of the PTE was conducted with XL-STAT software.

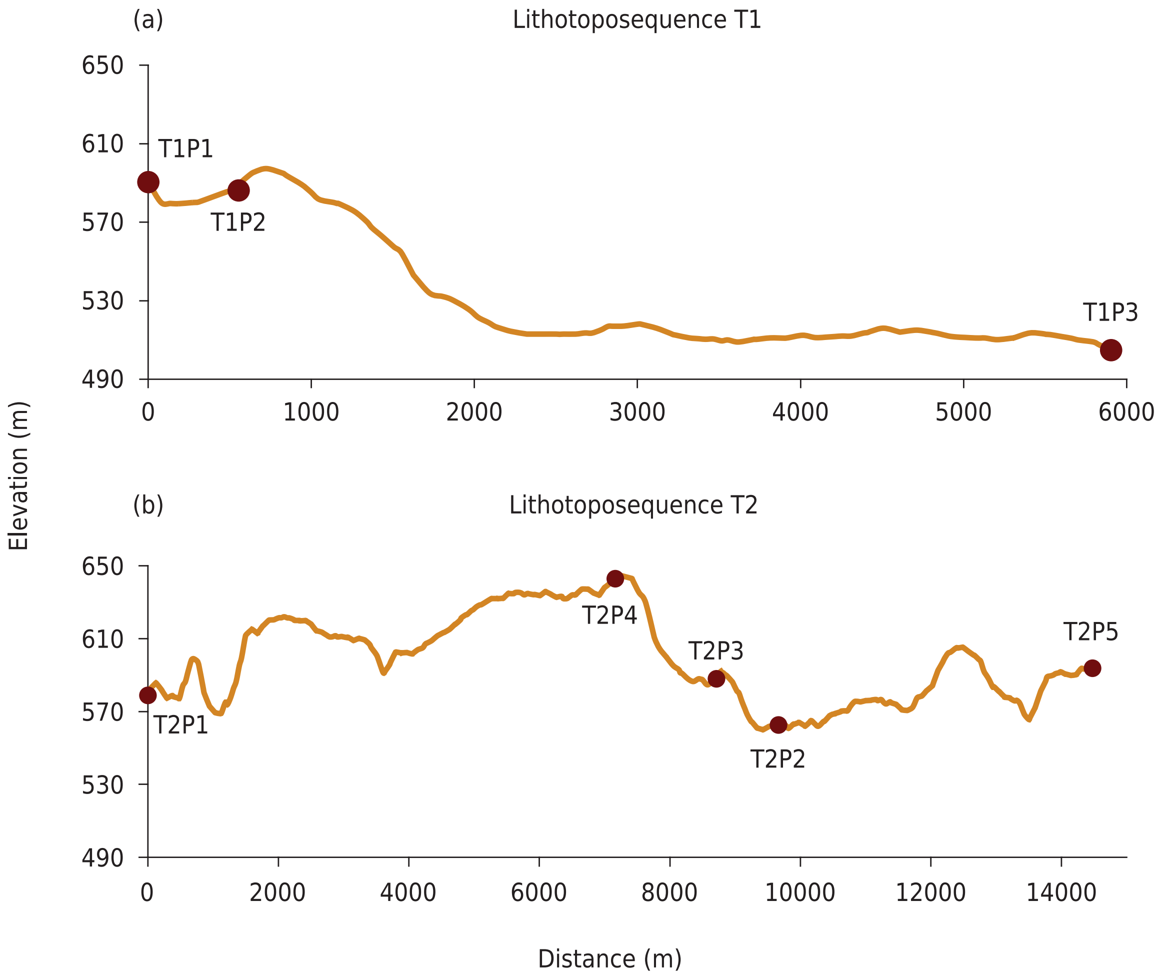

Figure 2. Topographic profile of the lithotoposequences T1 (a) and T2 (b) of the study area at the Montes Claros (MG) microregion.

RESULTS AND DISCUSSION

Soils developed from limestones

Soils derived from limestone are located in the shoulder (Leptosol/*Neossolo Litólico* - T1P1) and backslope (Nitisol/*Nitossolo* - T1P2) positions (Figure 2a). These carbonate materials influence soil chemical properties, reflected by the predominance of Ca as the exchangeable cation, thus increasing base saturation (BS) values, absence of exchangeable-Al, and pH above 7. These profiles were the only two pedons showing detectable amounts of $CaCO_3$ equivalent, which was expected considering their parent rocks (Table 1).

The sand fraction has quartz, mica, feldspar, and carbonates, most likely calcite and aragonite. The silt fraction has a similar mineral composition, but exclusively aragonite as carbonate. More intense reflections of aragonite and feldspars in the surface horizon of T1P2 than in the underlying horizons (Table 2) suggest a colluvial contribution of soils from upslope. Moreover, lower moisture content and higher temperatures can contribute to aragonite remaining in the surface horizons (Jones and Renaut, 2010).

In the T1P1 and T1P2 profiles, the major minerals in the clay fraction are kaolinite and illite (Table 2), which result from mica alteration from limestones (Vasconcelos et al., 2013). The mineralogical species and alteration pathways identified in T1P1 and T1P2 agree with several studies on soils formed from carbonate rocks (Oliveira et al., 1998; Shinzato, 1998; Mota et al., 2007; Pereira et al., 2013).

Soils derived from alluvial-colluvial sediments

The pedons T1P3 (Gleysol/*Gleissolo*) and T2P2 (Cambisol/*Cambissolo*) show little to moderate pedogenic development, mildly acidic to nearly neutral pH values, and high base saturation with high contents of Ca^{2+} and Mg^{2+} (Table 1).

The sand and silt fractions of T1P3 are essentially composed of quartz, while those of T2P2 have also minor amounts of mica and feldspar. Both T1P3 and T2P2 are derived from floodplain alluvial-colluvial sediments. While the T1P3 floodplain is bordered by soils developed from calcilutites and calcarenites of the Lagoa do Jacaré formation, the T2P2 floodplain is bordered by soils developed from metasiltstones and slaty shales of the Serra de Santa Helena formation (Chaves and Andrade, 2011). Therefore, there is not only a contribution of alluvial and colluvial sediments to the parent material of these two pedons, but also the contribution of the restricted drainage that inhibits some mineral alterations, which explains the presence of mica and feldspar, mainly in T2P2.

Table 1. Chemical properties of soil profiles from two lithotoposequences located in the Montes Claros (MG, Brazil) microregion

Horizon	Depth	pH(H_2O)	Ca^{2+}	Mg^{2+}	K^+	Na^+	SB	Al^{3+}	H^+	T	BS	TOC	$CaCO_3$ Eq
	m		$cmol_c$ kg^{-1}								%	g kg^{-1}	
T1P1 - Eutric Leptosol (*Neossolo Litólico Eutrófico típico*)													
A1	0.00-0.50	7.5	16.6	2.6	1.32	0.06	20.6	0	2.7	23.3	88	60.8	20
A2	0.50-0.17	7.4	13.1	0.7	0.43	0.05	14.2	0	1.2	15.4	92	30.5	21
T1P2 - Eutric Nitisol (*Nitossolo Háplico Eutrófico típico*)													
A	0.00-0.15	7.2	11.1	0.5	0.32	0.03	11.9	0	1.7	13.6	87	24.5	18
Bt1	0.15-0.35	7.1	9.2	0.3	0.19	0.02	9.7	0	1.3	11.0	88	14.1	13
Bt2	0.35-0.50	7.1	9.3	0.3	0.11	0.03	9.7	0	1.2	10.9	89	11.9	11
BC	$0.50\text{-}0.60^+$	7.4	9.9	0.3	0.13	0.03	10.4	0	1.2	11.6	90	8.9	15
T1P3 - Eutric Gleysol (*Gleissolo Háplico Ta Eutrófico vertissólico*)													
A	0.00-0.25	5.4	10.2	1.2	0.56	0.14	12.0	0.0	8.0	20.1	60	50.5	0
Bg1	0.25-0.30	5.6	8.7	1.3	0.16	0.15	10.3	0.1	7.9	18.3	56	39.0	0
Bg2	0.30-0.60	7.1	13.3	1.9	0.07	0.19	15.5	0.0	1.1	16.6	93	9.6	0
Cg	$0.60\text{-}1.00^+$	7.7	8.9	1.3	0.04	0.18	10.4	0.0	0.5	10.8	96	3.7	0
T2P1 - Leptic Dystric Regosol (*Neossolo Litólico Distrófico típico*)													
A	0.00-0.15	5.2	0.6	0.3	0.28	0.00	1.2	1.2	4.0	6.4	18	15.0	0
Bi	0.15-0.20	5.1	0.3	0.2	0.19	0.05	0.8	1.9	3.9	6.6	12	10.5	0
Cr	0.20-0.40	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
T2P2 - Vertic Eutric Cambisol (*Cambissolo Háplico Ta Eutrófico vertissólico*)													
A	0.00-0.25	6.4	11.8	1.6	0.21	0.09	13.7	0.0	2.5	16.2	84	32.2	0
AB	0.25-0.50	6.2	9.4	1.0	0.05	0.06	10.6	0.0	2.3	12.8	83	12.0	0
Bi	0.50-1.00	6.5	8.2	1.1	0.03	0.09	9.4	0.0	1.7	11.2	85	9.0	0
T2P3 - Eutric Nitisol (*Nitossolo Háplico Eutrófico típico*)													
A1	0.00-0.15	6.5	5.2	1.8	0.35	0.02	7.4	0.0	2.4	9.8	76	25.5	0
A2	0.15-0.30	6.3	4.7	1.7	0.23	0.02	6.6	0.0	2.4	9.0	74	21.0	0
BA	0.30-0.48	6.3	4.5	1.8	0.29	0.01	6.6	0.0	1.9	8.5	77	16.5	0
Bt1	0.48-0.68	6.5	3.0	1.7	0.30	0.01	5.0	0.0	1.2	6.3	81	8.2	0
Bt2	0.68-1.10	6.8	2.8	2.4	0.28	0.01	5.4	0.0	1.1	6.5	83	3.8	0
T2P4 - Rhodic Ferralsol (*Latossolo Vermelho Distrófico típico*)													
A	0.00-0.15	4.6	0.2	0.2	0.14	0.01	0.6	1.8	7.2	7.8	7	30.0	0
BA	0.15-0.39	4.6	0.0	0.1	0.04	0.00	0.1	1.9	5.4	5.5	2	11.3	0
Bw1	0.39-0.60	4.4	0.0	0.0	0.01	0.00	0.1	1.8	4.3	4.3	1	8.2	0
Bw2	0.60-1.00	4.7	0.0	0.0	0.01	0.00	0.1	1.8	3.9	3.9	1	5.2	0
T2P5 - Rhodic Ferralsol (*Latossolo Vermelho Distrófico típico*)													
A	0.00-0.15	5.0	1.0	0.5	0.22	0.00	1.7	0.4	3.5	5.7	31	10.5	0
BA	0.15-0.35	4.7	0.8	0.4	0.10	0.00	1.3	1.1	3.4	5.8	22	9.0	0
Bw1	0.35-0.70	4.5	0.9	0.5	0.10	0.00	1.5	1.4	3.7	6.6	22	8.2	0
Bw2	0.70-1.10	4.3	0.5	0.3	0.09	0.01	0.9	1.3	3.5	4.4	22	6.0	0
Bw3	$1.10\text{-}1.60^+$	4.8	0.4	0.2	0.09	0.00	0.7	2.1	2.5	5.2	12	3.0	0

SB = sum of bases (Ca^{2+} + Mg^{2+} + K^+ + Na^+); T = CEC at pH 7.0 (CEC = SB + H^++ Al^{3+}); BS = base saturation (BS = 100 × SB/T); TOC = total organic carbon; $CaCO_3$ eq = $CaCO_3$ equivalent; nd = not determined. Chemical analyses were performed according to Claessen (1997).

The main mineral in the clay fraction of these profiles is kaolinite (Table 2). This mineral was likely formed from primary minerals of the surrounding landscapes, such as feldspars and micas. Goethite (Fe oxyhydroxide) is also a common mineral in these two soils from floodplains; this mineral is expected to be more stable than hematite (Fe oxide) in soils developed from alluvial sediments (Vodyanitskii et al., 2007). Smectite is also a common product in the subsurface horizons of both profiles. A floodplain relief with impeded drainage and poor removal of bases and silica favors the formation and stabilization of 2:1 clay minerals. The occurrence of these minerals might also be due to partial alteration of primary micas, forming illite and smectite (Fanning et al., 1989).

Table 2. Particle-size distribution (PSD) and mineralogical constituents of soil profiles collected at two lithotoposequences located in the Montes Claros (MG, Brazil) microregion

Horizon	**PSD**			**Mineralogy**		
	Sand	**Silt**	**Clay**	**Sand**	**Silt**	**Clay**
		g kg^{-1}				
			T1P1 - Eutric Leptosol (*Neossolo Litólico Eutrófico típico*)			
A1	80	480	460	Mi, Fd, Qz, Ar, Ca	Mi, Fd, Qz, Ar, Ca	Kt, Il, Rt, Hm, Gt
A2	40	480	480	Mi, Fd, Qz, Ar, Ca	Mi, Fd, Qz, Ar, Ca	Kt, Il, Rt, Hm, Gt
			T1P2 - Eutric Nitisol (*Nitossolo Háplico Eutrófico típico*)			
A	140	400	460	Qz, Mi, Fd, Ar	Mi, Fd, Qz, Ar	Kt, Il, Rt, Hm, Gt
Bt1	120	370	510	Qz, Mi, Fd	Mi, Fd, Qz, Ar	Kt, Il, Rt, Hm, Gt
Bt2	130	400	470	Qz, Mi, Fd	Mi, Fd, Qz, Ar	Kt, Il, Rt, Hm, Gt
BC	160	350	490	Qz, Mi, Fd, Ar, Ca	Mi, Fd, Qz, Ar	Kt, Il, Rt, Hm, Gt
R	-	-	-	-	-	-
			T1P3 - Eutric Gleysol (*Gleissolo Háplico Ta Eutrófico vertissólico*)			
A	300	170	530	Qz	Qz	Kt, Il, Rt, Hm, Gt
Bgv1	230	160	610	Qz	Qz	Kt, Il, Rt, Gt
Bgv2	240	200	560	Qz	Qz	Kt, Il, Sm, Rt, Gt
Cg	400	170	430	Qz	Qz	Kt, Il, Sm, Rt, Gt
			T2P1 - Leptic Dystric Regosol (*Neossolo Litólico Distrófico típico*)			
A	190	370	440	Qz	Qz, Mi	Kt, Il, Rt, Hm, Gt
Bi	80	420	500	Qz, Mi, Fd	Qz, Mi	Kt, Il, Rt, Hm, Gt
			T2P2 - Vertic Eutric Cambisol (*Cambissolo Háplico Ta Eutrófico vertissólico*)			
A	20	440	540	Qz, Mi, Fd	Qz, Mi, Fd	Kt, Il, Rt, Hm, Gt
AB	60	540	400	Qz, Mi, Fd	Qz, Mi, Fd	Kt, Il, Sm, Rt, Hm, Gt
Biv	80	540	380	Qz, Mi, Fd	Qz, Mi, Fd	Kt, Il, Sm, Rt, Hm, Gt
			T2P3 - Eutric Nitisol (*Nitossolo Háplico Eutrófico típico*)			
A1	160	420	420	Qz, Fd	Qz, Mi, Fd	Kt, Il, Rt, Hm, Gt
A2	160	360	480	Qz, Fd	Qz, Mi, Fd	Kt, Il, Rt, Hm, Gt
BA	130	360	510	Qz, Mi, Fd	Qz, Mi, Fd	Kt, Il, Rt, Hm, Gt
Bt1	90	310	600	Qz, Mi, Fd	Qz, Mi, Fd	Kt, Il, Rt, Hm, Gt
Bt2	70	290	640	Qz, Mi, Fd	Qz, Mi, Fd	Kt, Il, Rt, Hm, Gt
			T2P4 - Rhodic Ferralsol (*Latossolo Vermelho Distrófico típico*)			
A	310	120	570	Qz, Fd, Rt	Qz, Fd, Rt	Kt, Gb, Il, Rt, An, Hm, Gt
BA	290	110	600	Qz, Fd, Rt	Qz, Fd, Rt	Kt, Gb, Il, Rt, An, Hm, Gt
Bw1	260	120	620	Qz, Fd, Rt	Qz, Fd, Rt	Kt, Gb, Il, Rt, An, Hm, Gt
Bw2	190	120	650	Qz, Fd, Rt	Qz, Fd, Rt	Kt, Gb, Il, Rt, An, Hm, Gt
			T2P5 - Rhodic Ferralsol (*Latossolo Vermelho Distrófico típico*)			
A	630	140	230	Qz, Fd, Rt	Qz, Fd, Rt	Kt, Gb, Il, Rt, An, Hm, Gt
BA	600	120	280	Qz, Fd, Rt	Qz, Fd, Rt	Kt, Gb, Il, Rt, An, Hm, Gt
Bw1	490	150	360	Qz, Fd, Rt	Qz, Fd, Rt	Kt, Gb, Il, Rt, An, Hm, Gt
Bw2	480	160	360	Qz, Fd, Rt	Qz, Fd, Rt	Kt, Gb, Il, Rt, An, Hm, Gt
Bw3	520	100	380	Qz, Fd, Rt	Qz, Fd, Rt	Kt, Gb, Il, Rt, An, Hm, Gt

Qz = quartz; Mi = mica; Fd = feldspar; Ar = aragonite; Ca = calcite; Il = illite; Kt = kaolinite; Sm = smectite; Rt = rutile; An = anatase; Hm = hematite; Pl = plagioclase; Gt = goethite; Gb = gibbsite. Particle-size analysis was determined according to Claessen (1997). - : not determined.

Soils derived from siltstone

Pedon T2P1 (Regosol/*Neossolo Litólico*) has pH below 5.2 (strongly acid) and fewer exchangeable bases than exchangeable Al, so it has a low BS (Table 1). Such features are inherent to the parent material itself, as the top of the Santa Helena formation has siltstones constituted by mica, pyrite, and quartz (Iglesias and Uhlein, 2009). In fact, quartz predominates in the sand and silt fractions, with smaller amounts of mica and feldspar in the Bi horizon, which suggests an association of these minerals with the parent material and lithogenic inheritance (Kämpf et al., 1997; Teramoto et al., 2001).

Soils derived from phyllite

The Nitisol (*Nitossolo*) (T2P3) developed from phyllite exhibits a slightly acidic to nearly neutral pH and high base saturation throughout the profile. The sand fraction is mostly composed of quartz. Feldspar is found exclusively in the sand and silt fractions of the surface A horizon, but it is presumed to be allochthonous. This is supported by the more intense feldspar reflexes in the sand fraction of the A1 horizon (0.00 to 0.15 m) compared to the underlying horizons. Even though there is evidence of allochthonous material in the surface horizon, the underlying lithology seems to have a strong influence on soil properties. This is highlighted by the abundance of micaceous minerals throughout the entire profile depth, also observed in the Nitisol (*Nitossolo*) developed from limestones (T1P2). The clay fraction of the Nitisol (*Nitossolo*) horizons contains kaolinite and illite, agreeing with results reported by Araujo et al. (2014) and by Garzón et al. (2016).

Soil derived from detrital cover

These sediments give rise to pedogenically well-developed soil. Pedon T2P4 (Ferralsol/*Latossolo*) is clayey to very clayey (570 to 650 g kg^{-1} of clay), strongly to extremely acidic, with the lowest sum of bases and base saturation among all the evaluated profiles.

The sand and silt fractions are mostly composed of quartz and rutile, which are weathering-resistant minerals. Kaolinite predominates in the clay fraction, but Fe oxides (goethite and hematite) are also found, as well as a small proportion of illite inherited from pre-weathered parent material (Oliveira et al., 1998; Ibraimo et al., 2004; Schaefer et al., 2008; Barbosa et al., 2009; Pereira et al., 2010). Gibbsite is found in small amounts, which signals the high degree of pedogenic development of this soil (Barbosa et al., 2009; Pereira et al., 2010).

Soil derived from rhythmite

The soil developed from rhythmite (T2P5) is classified as a Ferralsol (*Latossolo*) due to its morphological and chemical properties. However, unlike the Ferralsol (*Latossolo*) from detrital cover (T2P4), this soil has a loamy to coarse clayey texture (230 to 380 g kg^{-1} of clay) and greater sum of bases due to its higher amounts of Ca^{2+}, Mg^{2+}, and K^{+}, as well as higher base saturation (Table 1).

The mineralogical assemblage identified in the sand, silt, and clay fractions of T2P5 is essentially the same as that of T2P4. Quartz predominates in the sand and silt fractions, though feldspar and rutile also occur. The amount of rutile in the clay fraction of the A and Bw horizons of T2P5 is greater than the amount in T2P4, confirmed by a larger and more intense X-ray diffraction reflex in the basal spacing concerning this titanium oxide (data not shown). Since these two minerals are very stable, the higher amounts of rutile in the clay and of quartz in the sand fractions of T2P5 (Table 2) show the differences in terms of parent material between profiles P4 and P5. Despite that, the similarity in the morphological, chemical, and mineralogical properties between both Ferralsols (*Latossolos*) (P4 and P5) and the fact that both are situated on apparently stable relief positions (P4 flat and P5 gently rolling) suggest that both have an equivalent degree of pedogenic development.

Distribution of potentially toxic elements in the soil profiles

The standard quality sample (NIST) had recovery rates from 73.93 to 108.76 %, with the lowest rates for Al, Ba, and Cr.

Arsenic

Arsenic (As) contents varied with depth in all soil profiles (Table 3). The decrease in As content towards the subsurface gley horizons in T1P3 is due to the reducing environment, since its mobility in the soil is inversely related to redox potential (Kabata-Pendias, 2010), at least in initial stages (McBride, 1994). Reduction in As contents with depth was greater in T2P2 (15.0 to 0.8 mg kg^{-1}), which is located in a fluvial terrace.

For soils of the orders Leptosol (*Neossolo*), Regosol (*Neossolo*), and Nitisol (*Nitossolo*), As contents not only varied with depth but also between profiles (Table 3). The highest contents are found in the Regosol (*Neossolo*) from siltstone (22.7-29.0 mg kg^{-1}), while the lowest content is in the soil from phyllite (1.9-4.4 mg kg^{-1}). The largest ranges within profiles were found in the Leptosol (*Neossolo*) from limestone (T1P1) and in the Gleysol (*Gleissolo*) (T1P3). This variability can be assigned to the various chemical forms of As in soils, in addition to its varied solubility in accordance with pH, redox potential, and interaction with the surface of oxides, of silicate clay minerals, and of poorly crystalline minerals (McBride, 1994).

Arsenic contents in the Ferralsols (*Latossolos*) were low (2.18-3.95 mg kg^{-1}), with little variation throughout the profile, which confirms As as being adsorbed by oxides at low pH (McBride, 1994), properties identified in the Ferralsols (*Latossolos*) studied here.

Barium

Barium (Ba) contents in T1P1 and T1P2 tend to stabilize with depth, even under increased influence from parent material (limestone) (Kabata-Pendias and Mukherjee, 2007) (Table 3). After weathering, this element has low mobility in soils, which is due to its fast and stable adsorption to reactive soil surfaces, such as those of carbonates, 2:1 clay minerals, or organic matter (McBride, 1994). Conversely, in T1P3 (Gleysol/*Gleissolo*), contents changed up to 21 % (75.8-59.7 mg kg^{-1}) at depths below 0.30 m (Bg2 and Cg horizons), which might be related to the reducing environment (Lee et al., 2007).

Contents of Ba decreased with depth in T2 for Regosol (*Neossolo*) (T2P1), Cambisol (*Cambissolo*) (T2P2), and Nitisol (*Nitossolo*) (T2P3), but remained stable for both Ferralsols (*Latossolos*) (T2P4 and T2P5) (Table 3). In the Regosol (*Neossolo*) (T2P1) and Nitisol (*Nitossolo*) (T2P3), the presence of mica and feldspars might have caused these high values in the surface horizons, since Ba substitutes for K in the structure of such minerals (Who, 1990; McBride, 1994; Vázquez and Anta, 2009). Additionally, surface transport processes may increase Ba contents in surface horizons (Kabata-Pendias and Mukherjee, 2007; Biondi et al., 2011). These results are consistent with the position of profiles along the lithotoposequence T2 (P1-shoulder, P2-colluvial/alluvial plain, and P3-backslope), which increases susceptibility to surface sediment transport compared to P4 and P5.

In addition to little variation with depth, T2P4 and T2P5 have low Ba contents (Shacklette and Boerngen, 1984), ranging from 5.45 to 14.15 mg kg^{-1}, nearly 10 % of the other profiles. Except for the presence of gibbsite (Table 2), lower exchangeable base contents (Table 1), and lower pH(H_2O), no other consistent differences are found between Ferralsols (*Latossolos*) and the other soils to explain such decrease in Ba contents. However, major losses of bases due to pedogenesis are expected in Ferralsols (*Latossolos*), which might have caused a decline in Ba content, as already reported by Marques et al. (2004) in soils of the *Cerrado* biome in an advanced degree of weathering.

Table 3. Potentially toxic element contents in soil profiles from the lithotoposequences located in the Montes Claros (MG, Brazil) microregion

Horizon	Layer	As	Ba	Cd	Cr	Cu	Mn	Mo	Ni	Pb
	m	mg kg^{-1}								
T1P1 - Eutric Leptosol (*Neossolo Litólico Eutrófico típico*) (limestone)										
A1	0.00-0.05	2.37	42.96	1.81	19.99	7.69	541.30	< ld	10.29	17.91
A2	0.05-0.17	11.74	46.27	2.12	25.84	7.99	508.96	< ld	15.79	26.03
T1P2 - Eutric Nitisol (*Nitossolo Háplico Eutrófico típico*) (limestone)										
A	0.00-0.15	12.48	60.56	1.91	26.97	7.57	1268.78	0.14	14.36	31.44
Bt1	0.15-0.35	17.94	52.48	3.01	35.91	12.76	1154.85	< ld	19.78	30.51
Bt2	0.35-0.50	16.52	70.20	2.95	36.43	11.32	1500.21	< ld	25.03	38.91
BC	0.50-0.60$^+$	19.79	68.78	2.92	35.83	12.35	1117.96	< ld	19.24	30.15
T1P3 - Eutric Gleysol (*Gleissolo Háplico Ta Eutrófico vertissólico*) (fluvial sediments)										
A	0.00-0.25	7.10	63.88	0.39	20.06	13.60	32.41	0.44	7.16	12.20
Bg1	0.25-0.30	23.48	65.32	0.22	16.68	14.24	27.98	0.10	4.03	12.19
Bg2	0.30-0.60	8.37	75.77	1.03	16.17	8.93	30.23	2.93	6.96	11.32
Cg	0.60-1.00$^+$	2.16	59.69	1.13	13.96	1.71	22.96	0.04	6.30	9.19
T2P1 - Leptic Dystric Regosol (*Neossolo Litólico Distrófico típico*) (siltstone)										
A	0.00-0.15	29.00	177.57	2.07	20.32	16.19	878.35	0.46	18.08	98.68
Bi	0.15-0.20	22.73	60.32	1.82	23.26	19.35	155.86	0.30	20.17	42.12
T2P2 - Vertic Eutric Cambisol (*Cambissolo Háplico Ta Eutrófico vertissólico*) (fluvial sediments)										
A	0.00-0.25	15.02	61.40	1.76	19.49	8.68	463.80	< ld	11.64	12.81
AB	0.25-0.50	1.95	59.33	1.23	18.17	5.08	381.31	< ld	8.45	11.31
Bi	0.50-1.00	0.84	54.93	1.49	18.88	4.96	326.34	< ld	8.80	13.20
T2P3 - Eutric Nitisol (*Nitossolo Háplico Eutrófico típico*) (phyllite)										
A1	0.00-0.15	1.90	172.13	2.22	68.49	2.67	989.37	< ld	12.78	24.86
A2	0.15-0.30	4.44	87.46	2.03	51.48	2.18	627.42	< ld	12.13	25.03
BA	0.30-0.48	3.31	54.44	1.95	48.41	3.87	319.50	< ld	14.65	16.30
Bt1	0.48-0.68	2.60	53.18	2.38	47.27	4.22	301.57	< ld	12.02	14.53
Bt2	0.68-1.10	3.07	47.91	2.16	52.37	6.08	201.20	< ld	16.51	12.34
T2P4 - Rhodic Ferralsol (*Latossolo Vermelho Distrófico típico*) (detrital cover)										
A	0.00-0.15	3.28	5.61	2.07	130.64	< ld	56.97	0.03	2.32	17.62
BA	0.15-0.39	3.95	5.45	2.36	64.21	< ld	37.17	0.81	1.19	14.90
Bw1	0.39-0.60	2.67	6.34	2.08	53.37	< ld	35.92	0.20	1.45	19.81
Bw2	0.60-1.00	2.18	5.64	2.16	45.90	< ld	34.55	0.00	1.83	17.35
T2P5 - Rhodic Ferralsol (*Latossolo Vermelho Distrófico típico*) (rhythmite)										
A	0.00-0.15	3.20	8.59	0.43	47.86	< ld	71.44	0.00	1.27	7.68
BA	0.15-0.35	3.04	7.09	0.86	40.93	< ld	63.90	0.65	0.85	8.44
Bw1	0.35-0.70	3.73	14.15	0.74	18.55	< ld	40.36	0.20	1.18	9.09
Bw2	0.70-1.10	3.41	6.55	0.87	19.37	< ld	38.04	0.42	0.21	8.58
Bw3	1.10-1.60$^+$	2.56	6.68	0.75	20.17	< ld	32.63	0.15	9.91	8.83

< ld = below detection limit. Potentially toxic elements analyzed by the EPA 3051A method (Usepa, 2007).

Cadmium

Except for a few isolated horizons, and for the entire T2P5 profile (Ferralsol/*Latossolo* from rhythmite), cadmium (Cd) contents were relatively high (McBride, 1994; Cetesb, 2014). This element has medium to high mobility in acidic, well-drained soils (Matos et al., 1994; Pierangeli et al., 2005), mobility that tends to reduce in alkaline soils (pH >7) and reducing environments (McBride, 1994). Indeed, Cd contents were slightly higher in T1P1 and T1P2, where pH values were above 7.0. The levels were also high in T1P3 (Gleysol/*Gleissolo*), from 0.31 mg kg^{-1} in surface horizons (0.00-0.30 m), with pH(H_2O) 5.5, to 1.08 mg kg^{-1} in horizons below 0.30 m, with pH(H_2O) 7.4. In T2P5 (Ferralsol/*Latossolo*) with pH(H_2O) below 5 and loamy to coarse clayey texture, the Cd contents are also low (0.43-0.87 mg kg^{-1}).

Nevertheless, variation in Cd content in the studied soils is not only related to pH or drainage conditions, but also to factors such as parent material and pedogenic processes. In addition, Cd may form several types of complex ions and organic chelates that are easily mobilized and transported in soils during weathering processes (Kabata-Pendias, 2010).

Chrome

Chrome (Cr) contents range from 14 to 36 mg kg^{-1} in soils of T1, and in P1 and P2 of T2 (Table 3). However, in P3 (Nitisol/*Nitossolo*, phyllite), P4 (Ferralsol/*Latossolo*, detrital cover), and P5 (Ferralsol/*Latossolo*, rhythmite) of T2, these contents increase, and are higher in the surface horizons. The A horizon (0.00-0.15 m) of T2P4 has 130.64 mg kg^{-1} Cr, which is more than twice the content of the B horizon (Table 3). Although high, total Cr contents of this magnitude are commonly found in Ferralsol (*Latossolo*) (Marques et al., 2004; Caires, 2009). In this study, however, Cr contents in the A horizon were not associated with pedogenic or lithogenic factors. Additionally, local soil cover with native vegetation (Carrasco) reduces the probability of anthropogenic influences. In general, Cr ion complexation by soil organic matter may explain its surface contents. Some investigations have shown that Cr has low mobility along the soil profile, thus accumulating in surface layers (Fontes et al., 1993; Matos et al., 1994). Nevertheless, Cr contents in the surface horizons of T1P1 (Leptosol/*Neossolo*) and T1P2 (Nitisol/*Nitossolo*) were lower than in the subsurface horizons (Table 3). In the Gleysol (*Gleissolo*) (T1P3), there is a decrease in Cr contents with depth, in accordance with the lower levels of Fe oxides, potential adsorbers of Cr^{3+} (McBride, 1994), and Cr increased solubility in reducing environments, which facilitates leaching (Kabata-Pendias, 2010).

Copper

The copper (Cu) contents are as expected for soils from sedimentary rocks (Marques et al., 2004), with small differences among profiles, parent materials, and depth. None of the evaluated soils showed Cu accumulation in surface layers (Table 3), in contrast with the expected trend of Cu accumulation in surface layers (Ross, 1994) due to bonding to the soil organic fraction. In T1P2 (Nitisol/*Nitossolo*), Cu content was lower in surface layers than in the subsurface. Bini et al. (2011) also observed lower Cu contents in the surface of soils developed from limestone. Since Cu has low mobility in pH near neutrality, as in soils of T1, close-to-neutrality soil subsurface contents are presumed to reflect parent material Cu contents. Copper complexation with carbonates causes the low mobility of Cu in soils in this pH range (Kabata-Pendias, 2010).

The highest Cu contents were found in T2P1 (Regosol/*Neossolo*), with a low degree of weathering, and could be attributed to the contribution of parent rock (siltstone). In contrast, Cu contents were below the detection limit in the Ferralsol (*Latossolo*) profiles (T2P4 and T2P5). According to Baize and Sterckeman (2001), Cu can be mobilized under acidic pH conditions. Thus, the reaction of acidic soil associated with small colloidal surfaces (Table 1), potential sites for Cu adsorption, can explain contents below the detection limit in the studied Ferralsols (*Latossolos*).

In T1P3 (Gleysol/*Gleissolo*) and T2P2 (Cambisol/*Cambissolo*), Cu contents decreased markedly with depth in accordance with changes in organic matter (OM) content, which is consistent with the expected adsorption of Cu by OM (Hugen et al., 2013; Souza et al., 2015). Valadares (1975) also observed a reduction in Cu contents with depth in an Acrisol (*Argissolo*), but little variation along Ferralsol (*Latossolo*) profiles.

Manganese

Manganese (Mn) contents along profiles of both T1 and T2 were variable, with greater differences among profiles than within profiles (Table 3). Contents of Mn ranged from 23 to 1,500 mg kg^{-1}, a 65-fold difference. The high sensitivity of Mn to variations in redox potential and pH makes its contents in soils very dynamic and spatially variable, with high contents associated with Mn precipitates of variable degrees of crystallinity (McKenzie, 1989). Nevertheless, surface horizons tend to have higher Mn contents (Table 3), which contrasts with the expectation of Mn accumulating in horizons enriched with Fe oxides (McKenzie, 1989; Kabata-Pendias, 2010).

In T1P2, developed from limestone, the largest content of Mn is found in the Bt2 horizon (1,500 mg kg^{-1}), where the largest content of Mn oxides was also detected through sulfuric acid attack (data not shown). Soils derived from limestone are believed to have high amounts of Mn (Kabata-Pendias, 2010).

Contents of Mn are lower in T1P3 (Gleysol/*Gleissolo*) and in T2P4 and T2P5 (Ferralsol/*Latossolos*) (Table 3). In Gleysols (*Gleissolos*), this occurs due to increased mobility of Mn in these reducing environments (Fontes et al., 1993; Matos et al., 1994; Kabata-Pendias, 2010). However, in Ferralsols (*Latossolos*), soil acidity (Table 1) and the intense weathering and reworking expected during the evolution of these profiles contribute to this sharp decline in Mn contents in its different chemical forms.

Molybdenum

In the studied soils, Molybdenum (Mo) contents follow the same trend described by Kabata-Pendias (2010) and McBride (1994), i.e., always in very low contents and with higher mobility in neutral and alkaline soils than in acidic soils. Therefore, profiles T1P1, T1P2, T2P2, and T2P3, which have nearly neutral to moderately alkaline soil reaction (Table 1), show Mo contents below the limit of detection, except for the A horizon of T1P2 (Table 3). In T1P3, Mo contents are also low. In profiles T2P1, T2P4, and T2P5, where soil pH is strongly acidic (pH 4.3-5.2), these contents were below 0.81 mg kg^{-1}, but within detectable limits. This shows Mo soil distribution closely associated with pH.

Nickel

Differences in nickel (Ni) content within profiles are smaller than among profiles. In limestone-derived soils (T1), where Ni mobility is expected to be low due to high pH (McBride, 1994), the highest levels of Ni are observed in T1P1 (Leptosol/*Neossolo*) and T1P2 (Nitisol/*Nitossolo*). Although Ni contents in Gleysol (*Gleissolo*) (T1P3) are lower (Table 3), they may be influenced by adsorption on organic matter from the soil surface to a depth of 0.25 m, and on smectite (Table 2), at neutral pH, below the depth of 0.30 m. Lower content in the sub-horizon Bg1 (0.25-0.30 m) of T1P3 may be due to reduced influence of those adsorption surfaces (organic matter or 2:1 clay minerals), which are less abundant, and to a more acidic pH. Conversely, given its greater mobility in acid soils, the lowest Ni contents are in Ferralsols (*Latossolos*) (T2P4 and T2P5), with pH values of 5 or lower.

Lead

Except for T2P1 and T2P3, lead (Pb) contents are relatively homogeneous along the profile, as expected due to its low mobility, especially in soils with high pH or reducing conditions (McBride, 1994). Again, this might be due to specific adsorption reactions on oxides and clay minerals, complexation by organic matter, or precipitation with carbonates, hydroxides, and phosphates (Kabata-Pendias, 2010). The largest reductions in Pb with depth are registered for T2P1 (57 %) and T2P3 (35 %); these profiles also show a decrease in organic carbon contents of about 5 g kg^{-1} with depth.

The highest Pb contents were found in the siltstone-derived Regosol (*Neossolo*), and the lowest contents were in Ferralsols (*Latossolos*) (T2P4 and T2P5) and fluvial plain soils under a reducing environment (T1P3 and T2P2). This suggests that the degree of pedogenic development, expressed by soil taxonomic class, and parent material contribute to variation in Pb content among profiles.

Association of potentially toxic elements (PTE) to soil class and parent material

Principal component analysis (PCA) of the PTE showed that these elements contents were relatively homogeneous within profiles and also allowed distinction of one profile from another (Figure 3). This agrees with the theory that pedogenic processes are differentiated by pedon, defended by Dokuchaiev (Glinka, 1927) and Simonson (1959), which in turn influences PTE contents in soils. Soil profiles from the same parent material, such as T1P1 (Leptosol/*Neossolo*) and T1P2 (Nitisol/*Nitossolo*) are associated with the same group of PTE (Mn, Ni, Cd, and Pb), but in those horizons and pedons that are pedogenically more developed, such as those of T1P2, these associations are more well expressed, showing that soil evolution, and therefore soil class, also affects PTE distribution. Nevertheless, the contribution of parent material to these high PTE contents should not be overlooked, mainly in limestone-derived soils, where increased contents of these elements were found, just as reported in the literature (Kabata-Pendias, 2010).

Moreover, an inverse relationship with PTE is observed in the Ferralsols (*Latossolos*) from rhythmite (T2P5) and from detrital cover (T2P4). Pérez (1997) also observed low contents of Fe, Mn, Cu, Zn, Co, Cr, and Pb in soils developed from sand-clayey sediments. From these observations, it is fair to say that mainly Mn, Ni, Pb, and Ba, but also Cd, As, and Cu, are influenced by parent material, especially limestone, siltstone, and tertiary-cretaceous detrital cover. In the rhythmite-derived soil (T2P4, Ferralsol/*Latossolo*), low contents for all analyzed PTE likely result from loss by pedogenic processes. In soils derived from alluvial sediments and phyllite, there is no clear association of parent material with PTE. In soils derived from alluvial sediments, this is probably due to the reducing environment, given the water table fluctuation throughout the year that favors greater mobility and depletion of PTE (Kabata-Pendias, 2010; Souza et al., 2015). Molybdenum shows direct association with T1P3 (Gleysol/*Gleissolo*, fluvial sediments) and inverse association, of a lower magnitude, with T2P3 (Nitisol/*Nitossolo*, phyllite). Vodyanitskii et al. (2007) reported accumulation of some trace elements in alluvial soils due to transport by water

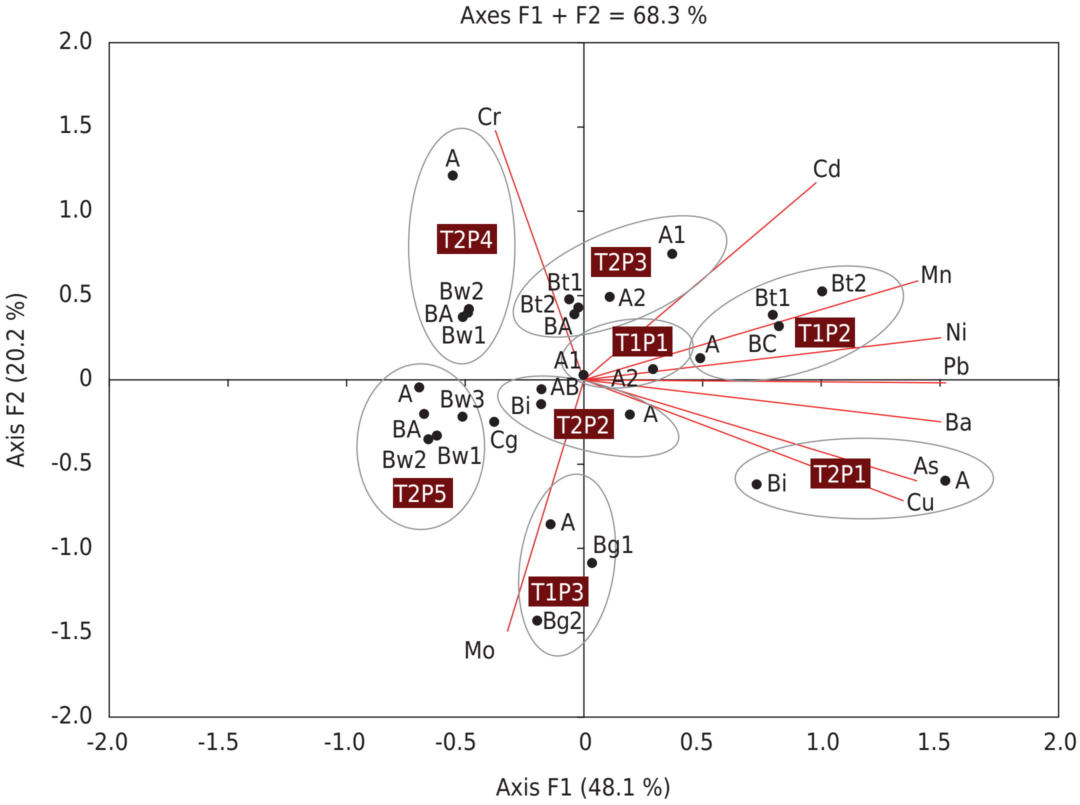

Figure 3. Principal component analysis (PCA) of the potentially toxic elements (PTE) of soil horizons and classes of profiles from the Montes Claros microregion, MG, Brazil.

and to the distribution of finer particle sizes. The strong adsorption of MoO_4^{2-} to the surface of less crystalline oxides and silicate clay minerals (McBride, 1994) could allow its transport by waters and accumulation in the Gleysol (*Gleissolo*) (T1P3). Variation in Mo contents in T1P3 from 0.04 to 2.93 mg kg^{-1} (Table 3) probably results from differentiation of the deposited layers. Associations of Cr are of lower magnitude, and occur at T2P4 (Ferralsol/*Latossolo*, detrital cover) and T2P3 (Nitisol/*Nitossolo*, phyllite).

Though pedogenic development and, thus, soil class are supposed to relate to PTE distribution, when looking at those soil classes of the Brazilian System of Soil Classification (SiBCS) which have more than one profile in the present study (*Neossolo*/Leptosol and Regosol, *Nitossolo*/Nitisol, and *Latossolo*/Ferralsol), one cannot observe a direct association of a soil class with a specific PTE, especially for *Nitossolos* (Nitisol) and *Neossolos* (Leptosol and Regosol). For *Neossolos* (Leptosol and Regosol), only the soil developed from siltstone (Regosol, T2P1) correlates with PTE. Likewise, for the studied *Nitossolos* (Nitisol), the one from limestone (T1P2) correlates with Mn, Ni, Pb, and Cd, whereas the subsurface horizons in the profile from siltstone (T2P3) correlate with Cr. Association of a soil taxonomic class with PTE is only valid for *Latossolos* (Ferralsols) (T2P4 and T2P5), which shows an inverse relationship with most PTE, with the lowest contents of the studied soils. As discussed earlier, this is due to the intense pedogenesis these soils went through, resulting in leached soils with low values of pH and CEC.

Therefore, even though parent materials have large influence on soil PTE contents (Fadigas et al., 2002; Oliveira and Costa, 2004; Campos et al., 2012), the effect of this factor depends on pedogenic development that, through many reactions and processes, can enhance or attenuate the original contents of rocks and sediments (Kampf and Curi, 2012; Souza et al., 2015).

CONCLUSIONS

Mineralogical constitution showing the contribution of parent material, *in situ* pedogenic processes, and *ex situ* (allochthonous) materials emphasizes interaction between pedogenesis and morphogenesis for the development of the studied soils. Quartz predominates in the sand and silt fractions, with variable amounts of easily weatherable primary minerals, whereas kaolinite prevails in clay fractions, followed by 2:1 clay minerals and oxides.

Patterns of PTE distribution unique to each pedon and differences in PTE contents in soils from the same parent material but with distinct pedogenic evolution show that pedogenic processes influence PTE distribution.

The PTE Mn, Ni, Pb, and Ba are most influenced by soil parent materials, but Cd, As, and Cu are also influenced. The most influential parent materials are limestone, siltstone, and detrital cover. To a lesser extent, Mo contents were influenced by phyllite, and Cr contents by detrital cover and phyllite.

Rhodic Ferralsol (*Latossolo Vermelho Distrófico típico*) was the only taxonomic class to show association, as a class, with PTE. The association of Ferralsols (*Latossolos*) with Cd, Mn, Ni, Pb, Ba, As, and Cu, and the consistently low contents of these elements found in soils of this class suggest that pedogenic processes common to the evolution of Ferralsols (*Latossolos*) are responsible for decreasing levels of these PTE.

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