



Ion leaching and soil solution acidification in a vadose zone under soil treated with sewage sludge for agriculture



Ricardo Perobelli Borba^{a, *}, Victor Sanches Ribeirinho^d, Otávio Antonio de Camargo^b, Cristiano Alberto de Andrade^d, Carmen Silvia Kira^c, Aline René Coscione^b

^a Geosciences Institute, UNICAMP, Rua Carlos Gomes, 250, 13083-855, Campinas, SP, Brazil

^b Soil and Environmental Resources Center, Agronomic Institute of Campinas-SP, Av. Barão de Itapura, 1481, 13012-970, Campinas, SP, Brazil

^c Food Center, Adolfo Lutz Institute, Av. Dr. Arnaldo, 355, CEP 01246-902, São Paulo, SP, Brazil

^d EMBRAPA Environment, Brazil

HIGHLIGHTS

- Long-term monitoring of the soil solution of tropical soil that received sewage sludge for agricultural purposes was done.
- The physical-chemical parameters and chemical composition of soil solution (SS) were evaluated up to 5 m depth.
- The SS acidification was provoked by both nitrite oxidation and ion leaching.
- The ionic strength of the SS has controlled the ion leaching.

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ABSTRACT

In this study, we performed monitoring of the soil solution (SS) over 10 years on a loamy/clayey-textured Dark Red Dystroferic Oxisol that received sewage sludge for agricultural purposes. The SS was obtained by lysimeters installed along the walls of a well at 1 m, 2 m, 3 m, 4 m and 5 m in depth. The major ions found in the SS were NO_3^- , SO_4^{2-} , Cl^- , Ca^{2+} , Mg^{2+} , Al^{3+} , Pb^{2+} , Cd^{2+} and Zn^{2+} , and the pH level ranged from 4 to 6.5 along the profile. Throughout the first three years of monitoring, the pH at a 3-m depth became more acidic, and in the last year, this trend reached 5 m. At the 5-m depth, the pH decreased from 6.5 to 4.5 from the first to the last monitoring. The SS acidification was provoked by both nitrite oxidation and ion leaching. The leaching of H^+ or the possible ion exchange/desorption of H^+ due to the leached cations (Ca^{2+} and Mg^{2+}) at the 4-m and 5-m depth caused the pH decrease. The ionic strength (IS) of the solution controlled the ion leaching. The sludge application increased the IS to 3 m, increasing the density of the soil charges and its ability to absorb ions. After the sludge application was completed, there was a decrease in IS of the SS as well as a decrease in ion absorption and retention abilities, which promoted leaching to greater depths. During the entire monitoring process, NO_3^- , Cd and Pb remained above the potability limit.

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1. Introduction

The agricultural use of sewage sludge has been the primary means of disposition of these solid residues of basic municipal sanitation in countries such as England, Australia, the United States of America, and the European Union (Sharma et al., 2017). In Brazil, it is estimated that only 10% of the sludge produced is currently

used for agricultural purposes, and the remainder is disposed of in sanitary landfills. However, the expectation is that there will be substantial development regarding sludge application (Bettli and Camargo, 2006). The main reasons for the agricultural use of sludge are related to its ability to provide nutrients for crops and organic carbon for soils, especially tropical soils poor in organic matter (Haynes et al., 2009; Bettli and Ghini, 2011).

When applied to the soil, the sewage sludge is mineralized, releasing both inorganic ions and organic matter into the SS. The ions and the organic matter may be leached at the vadose zone—or unsaturated zone—up to crop root systems, even reaching the

* Corresponding author.

E-mail address: borba@ige.unicamp.br (R.P. Borba).

groundwater. During leaching, the interaction between the SS and soil at the vadose zone may lead to ionic exchange, adsorption and desorption, and solubilization and precipitation of amorphous solid phases and minerals, altering the chemical composition and the physical-chemical parameters of the soil (Borba et al., 2015).

Major research on the agricultural use of sludge has focused on the following aspects: efficiency in providing nutrients; crop productivity and diseases; transfer of pathogens present in the sludge to the crops and soils; translocation of toxic elements to the biota; and accumulation of these nutrients and potential toxic elements and hormones in the soil (Lake et al., 1984; Sloan et al., 1997; Silveira et al., 2003; Bettiol and Ghini, 2011; Clarke and Smith, 2011; Marguí et al., 2016; Shargil et al., 2016; Mossa et al., 2017). The availability and transfer of ions (nutrients and potential toxic elements), persistent organic pollutants, hormones and pathogens from the sludge to the SS have been examined in short-term laboratory studies using both leaching columns and incubation (Suciú et al., 2015; Toribio and Romanyà, 2006), lysimeters (Vogeler et al., 2006; Agopsowicz et al., 2008) and in the field (Wang, and Jones, 1994; Joshua et al., 1998; Speir et al., 2003; Wolejko et al., 2013). In general, these works are related to the consequences of sludge application to SSs, whether during its application or in a short-term period after its application, and at shallow depths. The long-term and greater depth consequences of sludge application to the SS are practically unknown, especially in tropical soils.

In this study, after the sludge application was finished, there was a decade-long evaluation of both the variations of physical-chemical parameters and the chemical composition of the SS to a 5-m depth, in the vadose zone, within an area that has received sludge for agricultural purposes.

2. Materials and methods

2.1. Experimental section

2.1.1. Area of study

The study was conducted at the experimental field of Embrapa Environment (Brazilian Agricultural Research Corporation), in Jaguariúna, São Paulo, Brazil, from 2003 to 2013. Since 1999, a project on the agricultural use of sewage sludge has occurred in the field, and a “loamy/clayey-textured Dark Red Dystroferic Oxisol” was applied. Before the experiment started, the area was maintained for nine years without cultivation or treatment except for *Brachiaria* sp. (Vieira et al., 2005). There were six sewage sludge applications: April (64 ton ha⁻¹) and December 1999 (32 ton ha⁻¹), October 2000 (42 ton ha⁻¹), November 2001 (42 ton ha⁻¹), November 2002 (42 ton ha⁻¹), and December 2003 (31 ton ha⁻¹) (Bettiol and Ghini, 2011), and corn cultivation has occurred after every sludge application to the soil.

2.1.2. Sludge applied

Throughout five years, from 1999 to 2003, 238 ton/ha of sludge

(dry base) produced at a sewage treatment station that received household and industrial effluents was applied. The sludge dose applied was eight times more than the quantity recommended, and it was aimed at evaluating its effects on soil and crops (Bettiol and Ghini, 2011). In total, the following was added to the soil (g ha⁻¹): K → 2.97E+05, Na → 1.36E+05, organic C → 7.57E+07, Total Kjeldahl N → 8.56E+06, NH₄⁺-N → 5.72E+05, NO₃⁻-N → 1.79E+07, S → 3.20E+06, Mg → 8.45E+05, Al → 5.00E+06, Ca → 7.14E+06, and Zn → 7.07E+05. Since the first sludge application in 1999 until 2013, there has been annual corn cultivation at the plot where the well was installed. Due to the presence of industrial sewage in the sludge, there has also been the addition of potential toxic elements to the soil, such as (g/ha) Cd → 3.02E+03, Cr → 2.07E+05, Ni → 1.04E+05, and Pb → 5.95E+04.

2.2. Soil sampling

In October 2003, four years after the initial sludge application, a well was constructed with 5 m depth and 1.4 m diameter for the collection of soil and SS samples. During the well construction, there was a continuous collection of soil deformed and undeformed samples throughout the profile (0–5 m depth). After the collection, the well was lined with concrete rings over its entire depth.

2.3. Soil solution sampling

Soil solution extractors were installed along the walls of the well, from which the SSs were collected through the application of a vacuum to the extractors (Fig. SM1b – Supplementary Material). Three extractors (A, B and C) were installed at each depth level (1 m, 2 m, 3 m, 4 m and 5 m), and three samples were collected at each depth. These samples (e.g., 1A, 1B and 1C) were mixed in the laboratory in the same container due to the little volume available for the performance of the chemical analyses and physical-chemical determinations, resulting in a single sample for every depth in each collection (e.g., 1A+1B+1C = 1; 2A+2B+2C = 2; ... 5A+5B+5C = 5). The SS collections occurred in (number of collections in parentheses): 2003–2004 (5); 2004–2005 (14); 2005–2006 (9); 2006–2007 (3); and 2013 (6). The annual average rainfall was 1277 mm, with approximately 65% of rain concentrated in summer, between November and March.

2.4. Analytical methods

2.4.1. Soil

Approximately 10 kg of soil of each deformed sample collected along the soil profile in the well were homogenized and quartered in order to perform the soil determinations described below.

The chemical and physical attributes of the soil (Tables 1 and 2) were determined in samples with granulometry less than 2 mm according to the methods described in Camargo et al. (1986) and Raj et al. (2001), respectively. The Fe oxides and Al amorphous and

Table 1
Soil electrochemical properties and physical-chemical characteristics, and total trace elements of the soil profile.

Depth. (m)	CEC	AEC	Soil pH _{H2O}	PZSE	clay	silt	sand	θ	Al _a	Fe _a	Al _c	Fe _c	Cr	Cu	Ni	Pb	Zn
0–0.2	16.50	0.03	5.5	3.2	46.5	8.9	44.6	–	2.1	1	27	70	96	78	30	27	135
0.2–0.4	3.53	0.03	5.1	3.3	50.1	8.2	41.8	59	–	–	–	–	–	–	–	–	–
0.9–1.0	2.73	0.21	4.4	3.6	46.8	7.4	45.9	53	1.5	0.8	28	71	81	51	22	24	51
1.9–2.1	2.73	0.22	4.2	4.6	50.3	7.2	42.5	57	2.1	0.5	28	71	77	52	22	24	46
2.9–3.1	2.65	0.62	4.9	5.2	54.4	5.5	40.2	55	1.9	0.2	26	72	75	46	21	24	48
3.9–4.1	2.39	0.83	5.5	6	56.5	7.4	36.2	53	1.6	0.2	18	73	75	47	23	26	46
4.9–5.1	2.44	0.89	5.5	6.3	46.5	8.9	44.6	52	1.1	0.2	17	76	75	47	24	25	46

CEC–cation exchange capacity (cmolc kg⁻¹); AEC–anion exchange capacity (cmolc kg⁻¹); PZSE–point of zero salt effect; θ – soil porosity (%); Cr, Cu, Ni, Pb and Zn (mg kg⁻¹).

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