

# PHYSICOCHEMICAL CHARACTERIZATION OF PADDY SOILS CONTAMINATED WITH INDUSTRIAL WASTEWATERS: IMPLICATIONS FOR BIOREMEDIATION

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## Abstract

Concern has mounted over recent decades regarding the paddy soils located in Rancaekek, Bandung, West Java, Indonesia that have been heavily polluted by industrial effluents for many years. Recent efforts to remediate the heavily polluted sites have failed due to incomplete understanding of the site characteristics. Hence, this study dealt with the physicochemical characterization of these soils to acquire a better understanding of the physicochemical impacts and eventually improve their soil quality so that they would again be suitable for agriculture. The polluted soils contained much higher salt concentrations, higher salinity, and elevated levels of heavy metals (Cr, Mn, Cu and Zn) than unpolluted soils at control sites. However, no significant difference was observed in the mean of pH between all sites. Maximum water holding capacity and cation exchange capacity (Na and K) in polluted soils were significantly greater than those in unpolluted soils. Moreover, the polluted soils had higher contents of N, P, K, S, organic C, and lower C/N ratio than unpolluted soils. These results indicate that the industrial wastewaters-contaminated paddy soils have major changes in their physicochemical characteristics, therefore being informative to the development of bioremediation strategies of these soils.

Keywords: heavy metals, salinity, maximum water holding capacity, cation exchange capacity

## 1. INTRODUCTION

Pollution of agricultural soils by heavy metals and salts from industrial activities is a recent major environmental concern. One of such polluted agricultural soils is the heavily polluted paddy soils in Rancaekek, Bandung, West Java, Indonesia. These areas are irrigated with water from the Cikijing River in which polluted by industrial wastewaters. Throughout the textile industrial wastewaters are the primary pollution source of the Cikijing River. The irrigation with these wastewater effluents has caused unhealthy paddy soils in these sites, thus lowering crop yield. Recent efforts to remediate the heavily polluted sites have failed due to incomplete understanding of the site characteristics. Thus, the detailed site characterization of these sites is prerequisite for successful bioremediation purposes. It is strongly recommended to determine the nature and extent of contamination; or determine the negative

impact to environment, monitor the migration of the contaminants, clean up the contamination, and monitor the effectiveness of the cleanup (King, Long, and Sheldon, 1998; Fiorenza, Oubre, and Ward, 2000; Swyngedouw and Crepin, 2008).

One of the major site characteristics for effectiveness in soil bioremediation is the soil physicochemical properties (e.g., pH, electrical conductivity (EC), soil texture, soil organic contents, nutrient contents, maximum water holding capacity (MWHC), cation exchange capacity (CEC), heavy metal contents) (Dick, 1992). The characteristics of physicochemical soil can affect the toxicity of organic pollutants; the higher the soil cation exchange capacity (high organic matter or clay content), the higher the adsorption of the organic pollutant and the lower its toxic effect on the ecosystem (McBride, 1994). Soil textures can inhibit or enhance soil enzymatic activity (Labud, Garcia, and Hernandez, 2007).

The high electrical conductivity of soils has adverse effects on soil structural stability, bulk density and permeability (Tejada and Gonzalez, 2005), as well as has detrimental effects on soil microbial biomass and activity (Rietz and Haynes, 2003; Wichern, Wichern, and Joergensen, 2006). Since microbially mediated processes in soils are central to their ecological function in conjunction with bioremediation of pollutants in soils, any reduction in soil microbial activity following soil physicochemical changes is of particular concern.

A better understanding of the physicochemical characteristics of the industrial wastewaters-polluted soils might thus be of help in assessing the recovery potential of these soils before choosing the specific remediation approach in order to develop an effective bioremediation design. The aim of this study was to characterize physicochemically the paddy soils in which contaminated with industrial wastewaters. The paddy fields were located in Rancaekek, Bandung, Indonesia in order to provide the detailed information on their physicochemical properties, thus being helpful for the development of bioremediation strategies of these soils.

## 2. METHODS

### Study Sites and Soil Sampling

The study area was located in Rancaekek District, Bandung Regency, West Java Province, Indonesia. There were two areas studied: soils that have received long-term industrial effluents located in Babakan Rancakeong Village, Linggar Subdistrict, Rancaekek District, Bandung Regency (polluted soils) and soils that have not received industrial effluents (as control sites) located in Walini Village, Bojongloa Subdistrict, Rancaekek District, Bandung Regency (unpolluted soils). Top soil layers (approximately 0-20cm in depth) of arable sites were sampled on May 13, 2007. A total

of 24 soil samples (1 kg each) were transferred to the laboratory. The samples were sieved (2 mm) to remove litter, roots, and coarse particles and stored at 4°C until analysis.

### Soil physicochemical properties and statistical analysis

Soil pH and EC (electrical conductivity) were measured by potentiometer in suspension (1:2.5 w/v) according to DIN ISO 10390 (DIN ISO 1997a). Maximum water holding capacity (MWHC) of soils was measured as described by Zhang *et al.* (2006). Total soil carbon content, total soil nitrogen content and total soil sulfur content were analyzed after dry combustion using a LECO CNS 2000 analyzer, according to DIN ISO 10694 and 13878 (DIN ISO 1996, 1998). Organic carbon was calculated after subtracting the amount of carbonate C. The cation exchange capacity (CEC) of the soil samples was determined according to DIN ISO 13536 (DIN ISO 1997b). Extractable contents of elements were analyzed according to DIN ISO 38414-S (DIN ISO 1983). The data are presented as the arithmetic mean  $\pm$  standard error of the mean. The significance of differences between the soil samples of the study sites was estimated by the one-way ANOVA at significance levels of  $P < 0.05$ , using the software SigmaStat version 2.03 (SPPS Inc., Chicago, Illinois).

## 3. RESULTS AND DISCUSSION

### Salt Contents and Salinity of Soils

Salt contents in the study sites are represented by Na, Mg, Al, Si, Cl and Ca (Table 1). Only salt contents of Na, Cl and Ca in polluted site were statistically greater by a factor of 1.5 - 242 than those in unpolluted site ( $P < 0.05$ ). Levels of Na (5371.99 mg kg<sup>-1</sup>) in polluted soils were 11 times higher than that in unpolluted soils (510.49 mg kg<sup>-1</sup>), while Ca level in polluted soils (6664.63 mg kg<sup>-1</sup>) was one and half times higher than that in unpolluted soils (5710.53 mg kg<sup>-1</sup>). Most strikingly, polluted soils (755.22 mg kg<sup>-1</sup>) had remarkably high Cl concentration 242 times

higher than unpolluted soils ( $3.13 \text{ mg kg}^{-1}$ ). These extractable salts determined concentration of ions contained in soils and could as well be measured by EC (electrical conductivity) values that therefore represented salinity levels. Salinity was positively correlated with EC, which increased with increasing salt content (Table 1). Polluted soils had EC level ( $3.58 \text{ dS m}^{-1}$ ) 36 times higher than unpolluted soils ( $0.10 \text{ dS m}^{-1}$ ) (Table 1). The high salinity of polluted soils obviously arose from industrial wastewaters containing high salt and organic concentrations (Lefebvre and Moletta, 2006). The high salt contents (Na and Cl) in these

soils may impair clay dispersion, soil sealing and severe crusting upon drying. Soil crusting, besides limiting the infiltration of water into soil, favours the evaporation of the water standing over the ground (thus increasing soil-surface salinization) and limits the gas exchange between soil and atmosphere (thus inducing poor soil aeration) (Mamedov and Levy, 2001; Menneer, McLay, and Lee, 2001; Sumner and Stewart, 1992). Salinity is one of the most stressing environmental conditions for soil micro-organisms that might affect the effectiveness of bioremediation (Sardinha *et al.*, 2003).

**Table 1.** Salt Contents and Salinity Of Paddy Soils in Study Sites

Salt content & Salinity	Study sites	
	Polluted (P)	Unpolluted (C)
Na ( $\text{mg kg}^{-1}$ )	5371.99 (326.88) a	510.49 (21.46) b
Mg ( $\text{mg kg}^{-1}$ )	1725.16 (82.77) a	1679.04 (14.03) a
Al ( $\text{mg kg}^{-1}$ )	143531.60 (6807.89) a	135847.60 (2347.27) a
Si ( $\text{mg kg}^{-1}$ )	1556.67 (24.63) a	1626.20 (67.94) a
Cl ( $\text{mg kg}^{-1}$ )	755.22 (110.75) a	3.13 (0.54) b
Ca ( $\text{mg kg}^{-1}$ )	6664.63 (238.47) a	5710.53 (89.40) b
EC ( $\text{dS m}^{-1}$ )	3.58 (0.16) a	0.10 (0.01) b

Values were calculated as means of all soil samples ( $n=6\sim 12$ ) with standard error in parentheses. Statistical differences are indicated by different letters ( $P = 0.05$ ). EC: Electrical Conductivity

### Heavy Metals of Soils

The industrial wastewaters also affected heavy metal concentrations in polluted soils. Polluted soils were contaminated with four heavy metals such as Cr ( $65.42 \text{ mg kg}^{-1}$ ), Mn ( $602.81 \text{ mg kg}^{-1}$ ), Cu ( $75.97 \text{ mg kg}^{-1}$ ) and Zn ( $167.39 \text{ mg kg}^{-1}$ ) (Table 2). The concentrations of these four heavy metals in polluted soils were greater than those in unpolluted soils (Cr:  $30.64 \text{ mg kg}^{-1}$ ; Mn:  $407.21 \text{ mg kg}^{-1}$ , Cu:  $57.34 \text{ mg kg}^{-1}$ , and Zn:  $82.70 \text{ mg kg}^{-1}$ ) by a factor of  $\sim 2$ . The concentrations of Mn ranged from 538 to 753  $\text{mg kg}^{-1}$  for polluted soils and ranged from 324 to 552  $\text{mg kg}^{-1}$  for unpolluted soils. Although the concentrations of Cr, Mn, Cu, Zn in polluted soils were greater than those in unpolluted soils, these heavy metal concentrations were below the maximum permissible concentrations of these heavy metals in soils (De Vivo, Belkin, and Lima, 2008). The average concentration of Cr in

soils is  $84 \text{ mg kg}^{-1}$  (Alloway, 1995). Mn concentrations are found in all soil types in the range of 50 – 11500  $\text{mg kg}^{-1}$  (Alloway, 1995). Maximum permissible concentrations of Cu and Zn in soil at neutral pH (pH 6.0-7.0) are  $135 \text{ mg kg}^{-1}$  and  $200 \text{ mg kg}^{-1}$ , respectively (De Vivo, Belkin, and Lima, 2008). Furthermore, Fe was most abundantly contained in soils (Table 2), probably because of its low bioavailability in aerobic environment at neutral pH apart from the typical properties of tropical soil (oxisol) (Brantley *et al.*, 2001). Metals of V, Co, Ag, Cd and Hg in all sites were undetectable with the exception of V and Co in unpolluted sites that were found in very less concentration (V:  $0.35 \text{ mg kg}^{-1}$ , Co:  $0.57 \text{ mg kg}^{-1}$ ). Furthermore, no significant difference was observed in concentrations of Ni ( $13.88 - 16.43 \text{ mg kg}^{-1}$ ), and Pb ( $3.64 - 4.55 \text{ mg kg}^{-1}$ ) between all sites (Table 2). The presence of these metals in these soils may enhance or inhibit bioremediation by

microorganisms; some metals may be toxic, while many are essential trace nutrients for microbial growth in sustaining bioremediation (Chaerun *et al.*, 2004a; Chaerun *et al.*, 2004b). Moreover, four elevated metal concentrations (i.e., Cr, Mn, Cu and Zn) should be taken into

consideration that may not only result in adverse effects on various parameters relating to plant quality and yield, but also cause changes in the size, composition and activity of the microbial activity (Perez-de-Mora *et al.*, 2006; Giller, Witter, and McGrath, 1998).

**Table 2.** Heavy Metal Concentrations of Paddy Soils in Study Sites

Heavy metal concentration (mg kg <sup>-1</sup> )	Study sites	
	Polluted (P)	Unpolluted (C)
V	ND	0.35 (0.22) a
Cr	65.42 (21.39) a	30.64 (0.33) a
Mn	602.81 (31.83) a	407.21 (31.80) b
Fe	26802.43 (194.30) a	26633.98 (137.04) a
Co	ND	0.57 (0.10) a
Ni	16.43 (1.91) a	13.88 (0.10) a
Cu	75.97 (6.19) a	57.34 (0.52) b
Zn	167.39 (62.64) a	82.70 (2.64) a
Ag	ND	ND
Cd	ND	ND
Hg	ND	ND
Pb	3.64 (1.07) a	4.55 (0.66) a

Values were calculated as means of all soil samples (n=2~6) with standard error in parentheses. Statistical differences are indicated by different letters (P = 0.05). ND: not detected

### MWHC, pH and CEC of Soils

The ability of soil components to hold and release positive ions (cations) from soil constituents was expressed in centimoles of cations per kg of soil (cmol (+) kg<sup>-1</sup>) and was known as cation exchange capacity (CEC). There were five kinds of CEC measured: CEC[sum] as the amount of exchangeable cations (Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, and protons, H<sup>+</sup>), CEC[Na] as the content of exchangeable Na<sup>+</sup>, CEC[Mg] as the content of exchangeable Mg<sup>2+</sup>, CEC[K] as the content of exchangeable K<sup>+</sup>, and CEC[Ca] as the content of exchangeable Ca<sup>2+</sup>. Both sites did not differ statistically in CEC[sum] and CEC[Ca], but they statistically differ in CEC[Na], CEC[K] and CEC[Mg] (Table 3). CEC[Na] and

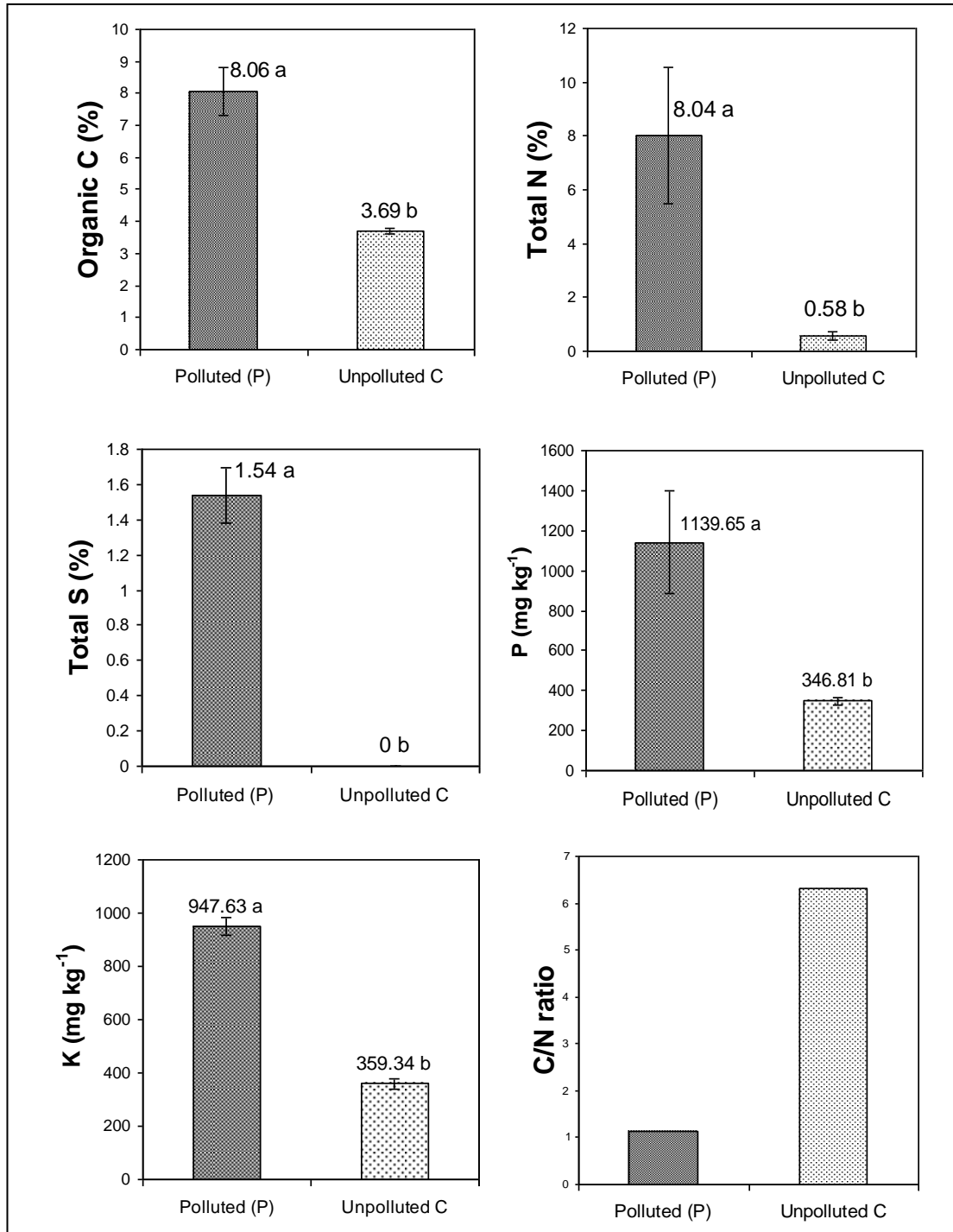
CEC[K] of polluted soils were greater by a factor of 25, 12, respectively, than those of unpolluted soils, whereas CEC[Mg] of polluted soils was lower by a factor of 1.5 than that of unpolluted soils. The influence of industrial wastewaters on CEC[Na] was most pronounced, indicating that polluted soils have high exchange capacity of Na<sup>+</sup> that was positively correlated to their salinity (Table 1). Both soils had circumneutral pH of 6.32 and 6.51, respectively (Table 3), indicating that soil pH was unaffected by industrial wastewaters. In addition, an increase in soil Na and Cl contents in polluted soils caused an increase in maximum water holding capacity greater than unpolluted soils (Table 3).

**Table 3.** MWHC, pH and CEC of Paddy Soils in Study Sites

Properties	Study sites	
	Polluted (P)	Unpolluted (C)
MWHC (g water g <sup>-1</sup> soil)	0.77 (0.04) a	0.66 (0.01) b
pH (H <sub>2</sub> O)	6.32 (0.14) a	6.51 (0.06) a
CEC[sum] (cmol <sub>c</sub> kg <sup>-1</sup> )	21.00 (0.40) a	21.00 (0.40) a
CEC[Na] (cmol <sub>c</sub> kg <sup>-1</sup> )	8.12 (0.55) a	0.33 (0.03) b
CEC[Mg] (cmol <sub>c</sub> kg <sup>-1</sup> )	5.06 (0.42) a	6.85 (0.12) b
CEC[K] (cmol <sub>c</sub> kg <sup>-1</sup> )	1.57 (0.05) a	0.14 (0.03) b
CEC[Ca] (cmol <sub>c</sub> kg <sup>-1</sup> )	15.12 (0.95) a	17.64 (0.64) a

Values were calculated as means of all soil samples (n=6~12) with standard error in parentheses. Statistical differences are indicated by different letters (P = 0.05).

MWHC: Maximum Water Holding Capacity; CEC: Cation Exchange Capacity; CEC[sum]: the amount of exchangeable cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ , and protons,  $\text{H}^+$ ); CEC[Na]: the content of exchangeable  $\text{Na}^+$ ; CEC[Mg]: the content of exchangeable  $\text{Mg}^{2+}$ ; CEC[K]: the content of exchangeable  $\text{K}^+$ ; CEC[Ca]: the content of exchangeable  $\text{Ca}^{2+}$ .



**Fig. 1.** Carbon and nutrient contents of paddy soils in study sites. Values were calculated as means of all soil samples ( $n=4\sim 12$ ). Error bars represent standard error. Statistical differences are indicated by different letters ( $P = 0.05$ ).

### Carbon and Nutrient Contents of Soils

Both sites statistically differed in contents of organic C, total N and total S (Fig. 1). Soils of polluted and unpolluted sites contained organic C (8.06%, 3.69%, respectively), and total N (8.04%, 0.58%, respectively). Polluted soils were composed of 1.54% of sulfur, while unpolluted soils contained no detectable S. Sulfur in these polluted soils may stimulate microbial role in bioremediation process.

Industrial wastewaters may carry chemicals that harbor huge amounts of organic C, thus macronutrients of C, N, S accumulating at polluted site are predominantly industrial wastewaters-derived. Furthermore, the polluted soils also contained higher levels of essential macronutrients for plant growth such as P (1139.65 mg kg<sup>-1</sup>) and K (947.63 mg kg<sup>-1</sup>) than unpolluted soils (P: 346.81 mg kg<sup>-1</sup> and K: 359.34 mg kg<sup>-1</sup>) (Fig. 1). These macronutrient levels in polluted soils were greater than those in unpolluted soils by a factor of ~ 3–4, which presumably arise from nutrient-rich industrial wastewaters due to the use of chemical substances in industrial processes. The discharge of industrial waste-waters has high salinity and high organic content (Lefebvre and Moletta, 2006). C/N ratio of polluted soils (1.13) was extremely lower than that of unpolluted soils (6.31) by a factor of 6, and they were statistically different ( $P < 0.05$ ) (Fig. 1), indicating a high decomposability of organic compounds contained in polluted soils (Guggenberger and Haider, 2002). A high organic C content in polluted soils might be assumed to alleviate the adverse effects of heavy metals and salinity increases (Yadvinder-Singh, Bijay-Singh, Timsina, 2005), while the high contents of N, P, K, S can be essential compounds of importance for microbial nutrition in bioremediation (Chaerun, Asada, and Tazaki, 2007; Chaerun *et al.*, 2004a; Chaerun *et al.*, 2004b).

### 4. CONCLUSIONS

After long-term exposure to industrial wastewaters, the paddy soils in Rancaekek, Bandung (West Java Province, Indonesia) contain high salinity and heavy metals. In addition to these, the industrial wastewaters have rendered not only potentially negative impacts (salt and heavy metal pollution) but also potentially positive impacts; an increase in major macro- and micronutrient contents. Macronutrient content (i.e., organic C, N and S) in polluted soils might act to mitigate the effect of increased salinity and heavy metals. This study provides fruitful information on paddy soils affected by industrial effluent-polluted irrigation waters that might be applicable to the development or application of bioremediation strategies.

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