



## Effect of Sub-Bituminous Coal on Negative Charge Activity on Secondary Forest and Horticultural Land Contaminated with Pesticides in Sungai Pua, Agam

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### ABSTRACT

Pesticides are increasingly used to deal with crop-disrupting organisms. However, only 1% are on target, and the rest threaten agricultural ecology. This research aims to study the effect of Sub-bituminous coal (SC) on the change of negative charge activity (NCA) on two types of land, namely secondary forest (SF) and pesticide-contaminated horticultural land (HL-P). Two studies respectively used a completely randomized design (CRD) with three replications on two land types and five doses, namely: A = control or 0 t/ha [0 g SC/500 g soil]; B = 10 t/ha [2.5 g SC/500 g soil]; C = 20 t/ha [5.0 g SC/500 g soil]; D = 30 t/ha [7.5 g SC/500 g soil]; and E = 40 t/ha [10 g SC/500 g soil]. The results show that the effect of 40 t/ha SC can increase NCA on the surface of soil colloids ( $\Delta pH$ ) by 43% in SF and 23% in HL-P. The effect of 40 t/ha SC on the two types of land has a significant effect on increasing pH H<sub>2</sub>O, EC, CEC, and OM composition, respectively, by 0.70; 0.04 dS/m; 44.30 cmol(+)/kg and 7.60% in SF and 0.33; 0.01 dS/m; 26.89 cmol(+)/kg and 3.00% in HL-P, compared to the control.

### INTRODUCTION

Pesticides are an important aspect of agriculture to protect against plant pest organisms. Their use is the most effective and widely accepted method of control (Bhandari et al., 2020). However, only 1% of the pesticides used reach the plants; the rest end up in groundwater and enter the food chain (Feld et al., 2015). Soil is the most important natural resource to act as a natural sink for organic and inorganic contaminants, such as pesticides. Pesticide contamination of soil impacts agricultural ecosystems because it can persist at relatively high concentrations in the soil, especially in horticultural land areas that have been heavily impacted by various pesticide applications (Sun et al., 2018). On the other hand, changes in land use that occur in meeting food needs make new land clearing into horticultural land, which directly causes

contamination with pesticides (Fig. 1). Many types of pesticides are used. Still, pesticides with the active ingredient glyphosate are the most widely used in horticultural fields, especially in the horticultural center of Sungai Pua, Agam District.

Various solutions have been made for pesticide-contaminated soil (Morillo & Villaverde, 2018). However, what must be considered in the selection of remediation methods for soil contaminated by pesticides is whether pesticide contamination is isolated or widespread on agricultural land, especially horticultural land. The best remediation development strategy to adopt is through the development of soil amelioration technology. Amelioration technology is a technology that can improve soil fertility and, at the same time, absorb organic or inorganic contaminants (Pant et al., 2020). Sub-bituminous coal can be used as a

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soil amendment, fertilizer, and adsorbent using amelioration technology.

Various studies have shown the optimal potential of sub-bituminous coal (SC) as an ameliorant soil, starting from its high humic substance (HS) content to its powder form and activation process, which can increase pH, CEC, and soil organic matter (SOM) on marginal lands (Herviyanti et al., 2021; Prasetyo et al., 2023; Maulana et al., 2023). Increased soil pH and CEC are important indicators in studying the phenomenon of modification of negative charge activity in soil (Hermawan et al., 2014). The PZC is the pH of the soil when the positive and negative charges on the soil colloids are equal or zero (Karna et al., 2018). The colloidal surface is positively charged if the  $\text{pH PZC} \geq \text{H}_2\text{O}$  value and negatively charged if the  $\text{pH PZC} \leq \text{H}_2\text{O}$  value. Thus, pH PZC can be used as a new system in soil management of nutrient availability and uptake of pollutants in the soil. It is also strongly influenced by the clay, pH, EC, CEC, minerals, and OM composition in the soil. The purpose of this research is to study the effect of SC on negative charge activity (NCA) on secondary forest and horticultural land contaminated with pesticides in Sungai Pua, Agam, West Sumatra.

## MATERIALS AND METHODS

The research was conducted at the Soil Fertility Chemistry Laboratory, Department of Soil and

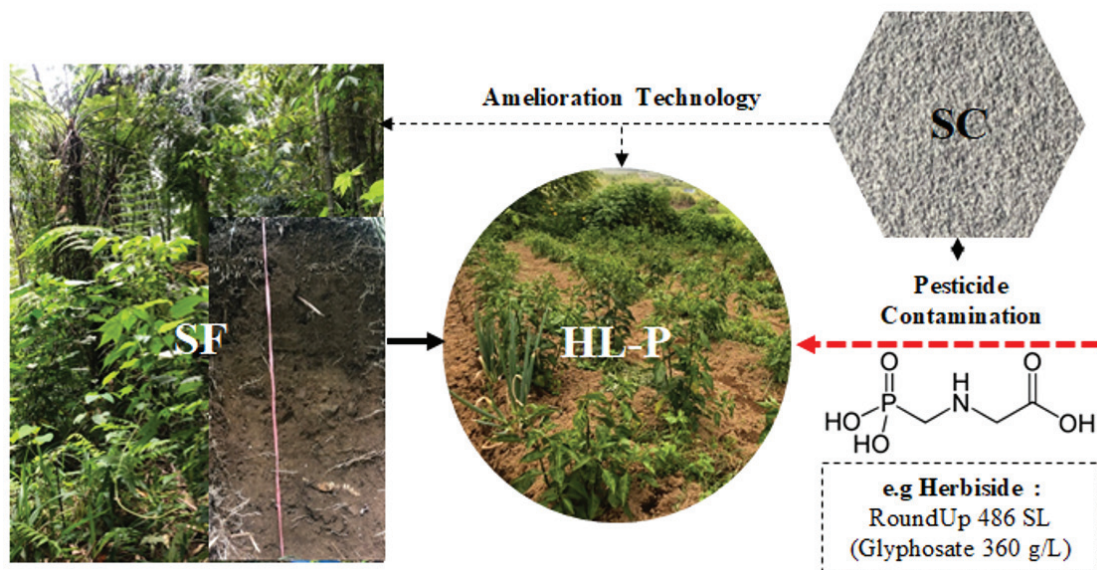
Land Resources of Agriculture, Andalas University, Padang, Indonesia, from May to September 2021.

## Experimental Design

This study used a completely randomized design [CRD] with three replications on two land types, namely: secondary forest [SF] and horticultural land contaminated with pesticides [HL-P], and five doses, namely: A = control or 0 t/ha [0 g SC/500 g soil]; B = 10 t/ha [2.5 g SC/500 g soil]; C = 20 t/ha [5.0 g SC/500 g soil]; D = 30 t/ha [7.5 g SC/500 g soil]; and E = 40 t/ha [10 g SC/500 g soil].

## Sub-Bituminous Coal and Soil Sampling

SC was obtained from Nagari Bonjol Pasaman, West Sumatra, at a soil depth of 1-2 m. SC was cleaned and pulverized with a Disc Mill model FFC 23 and sieved for 10 minutes with an Electromagnetic Sieve Shaker EMS-8 sieve measuring 500  $\mu\text{m}$ . The SC powder was weighed based on the application dose used in this study. Soil samples were taken compositely from Nagari Sariak, Sungai Pua, Agam, West Sumatra, with GPS coordinates -0021'56" LS and 100024'0" East, at a depth between 0-20 cm, with two types of soil. The calculated SC based on the application dose was put into experimental pots and incubated in 500g of absolute equivalent dry soil for two weeks. Soil samples were collected and analyzed to study changes in soil surface negative charge activity [pH, electrical conductivity (EC), mineral, and OM composition and CEC].



**Fig. 1.** Changes in land use from the secondary forest to horticultural land contaminated with pesticides and the effect of sub-bituminous coal on two types of land using amelioration technology.

### Soil and Statistical Analysis

Soil analyses conducted were pH ( $H_2O$ , KCl, PZC, and  $\Delta pH$ ) and EC by electrode method, mineral and OM composition by dry ashing method, and CEC by Leaching of  $NH_4OAC$  1N pH 7 method (Eviati & Sulaeman, 2012; Singh et al., 2017). The results of the soil analysis were then subjected to statistical analysis [Software: Microsoft Excel 2016 and Statistix 8®]. Data were analyzed by analysis of variance and if  $F \text{ Test} \geq F \text{ Table}$  (a significant effect at the 5% level [\*] and 1% level [\*\*]) using the Duncan Test.

### RESULTS AND DISCUSSION

The effect of SC on secondary forest and horticultural land contaminated with pesticides in the Sungai, Agam, has a significant effect on increasing the pH  $H_2O$ , EC, and composition of OM and CEC (Table 1). However, in this case, the soil surface

charge activity is based on soil colloids, which can be seen from the percentage of clay. The percentage of clay in the two types of land used looks different, where the clay content in the secondary forest is 5.90% less than in horticultural land contaminated with pesticides (30.64%). The high clay content in horticultural land contaminated with pesticides is influenced by tillage by farmers. This causes a difference in the NCA of the soil. The three main sources of negative charge on clay minerals are (1) excess negative charge on crystal fractures (Si-tetrahedral and Al-Octahendral); (2) dissociation of  $H^+$  from the OH groups present in the colloidal wall, where this dissociation occurs when the soil pH increases, and (3) isomorphic substitution, which occurs when (a) a low-valent cation substitutes a highly valent cation; (b) the atomic radii are relatively the same; and (c) frequently occurs in 2:1-type minerals.

**Table 1.** Effect of sub-bituminous coal on negative charge activity on secondary forest and horticultural land contaminated with pesticides

Effect of SC (t/ha)	Clay	pH			ΔpH	EC	Composition		CEC
		H <sub>2</sub> O	KCl	PZC			Mineral	SOM	
	%	unit			dS/m	%	cmol(+)/kg		
A. Secondary forest									
0	5.90	4.97 c	4.73 c	4.50	-0.24	0.08 e	70.80 a	29.20 c	30.25 d
10		5.40 b	4.90 b	4.40	-0.50	0.09 d	70.60 a	29.40 c	50.62 c
20		5.53 ab	4.93 ab	4.33	-0.60	0.10 c	65.20 b	34.80 b	53.97 c
30		5.53 ab	4.93 ab	4.33	-0.60	0.11 b	63.20 c	36.80 a	62.65 b
40		5.67 a	5.00 a	4.33	-0.67	0.12 a	63.20 c	36.80 a	74.55 a
CV (%)	-	1.91	0.91	2.50	-	2.56	0.40	0.79	4.96
Duncan's Test	-	**	**	ns	-	*	**	**	**
B. Horticultural land contaminated with pesticides									
0	30.64	4.97 c	4.70 c	4.43	-0.27	0.07 b	77.00 a	23.00 b	57.64 e
10		5.03 b	4.70 c	4.37	-0.33	0.07 b	74.60 b	25.40 b	74.68 d
20		5.10 b	4.73 b	4.37	-0.37	0.07 b	74.40 bc	25.60 ab	77.63 c
30		5.20 ab	4.77 ab	4.33	-0.43	0.08 a	74.00 c	26.00 a	82.12 b
40		5.30 a	4.80 a	4.30	-0.50	0.08 a	74.00 c	26.00 a	84.53 a
CV (%)	-	1.89	0.77	2.37	-	3.46	0.35	1.05	1.58
Duncan's Test	-	*	*	ns	-	**	**	**	**

Remarks: SC = Sub-bituminous coal; EC = Electrical conductivity; CEC = Cation exchange capacity; CV = Coefficient of variation; \*\* = Significant at the 0.01 level; \* = Significant at the 0.05 level and ns = non-significant; n = 15.

The basis of surface charge occurs during the formation of clay minerals, due to which isomorphic substitutional crystal arrangements are created. Cations that are not optimally integrated into the clay crystal lattice, such as  $\text{Al}^{3+}$  cations, can be replaced by  $\text{Si}^{4+}$  cations. The resulting crystal structure has a net negative charge. The electric double layer (EDL) balances the net negative charge of the clay crystal surface. The aft layer is composed of water and a stationary layer of ions on the clay surface. The aft layer is thought to be made up of counterions in a dense layer near the clay surface. The EDL is a diffuse layer of gouy cells that expand into solution as the concentration decreases rapidly. According to Preocanin et al. (2016), Gouy-Chapman, combining measurements of the EDL size Debye-length ( $\lambda$ ) and inverse Debye-length ( $K^{-1}$ ) can be used to measure the EDL value quantitatively. This study has not studied these measurements in detail. However, it is only seen from the percentage level of clay in each type of soil used.

The negative charge can increase due to the dissociation of  $\text{H}^+$  from the OH groups present in the colloidal wall, and this dissociation occurs when the soil pH increases, such as when adding organic matter. The effect of 40 t/ha SC on both soil types has a significant on increasing the pH of  $\text{H}_2\text{O}$  (Table 1). The increase in pH  $\text{H}_2\text{O}$  in SF and HL-P is 0.70 and 0.33, respectively, compared to the control. In Table 1A, it can be seen that the increase in pH in the secondary forest is 0.70 units at the effect of 40 t/ha SC. The effect of 30 and 20 t/ha SC looks the same as the administration of 40 t/ha SC, but it looks different from the effect of 10 t/ha SC, with an increase of 0.36 and 0.43, respectively, compared to the control. This also occurred in horticultural land contaminated with pesticides, where the pH increases by 0.33 and 0.23 at 40 and 30 t/ha SC applications, respectively.

Meanwhile, the effect of 20 and 10 t/ha SC is 0.13 and 0.06, respectively, compared to the control (Table 1B). This increase in pH occurs. The effect of SC through amelioration technology can increase the pH, where the pH in the secondary forest is higher than the pH in horticultural land contaminated with pesticides. According to Herviyanti et al. (2021) and Prasetyo et al. (2021), SC can increase the pH value of soil because it has phenol and carboxyl functional groups.

The EC content in secondary forests is higher than the EC content in horticultural land

contaminated with pesticides. In Table 1A, it can be seen that the application of SC to the secondary forest has a significant on increasing EC, where the highest EC value is 0.12 dS/m at 40 t/ha SC, and as the SC dose increased, the EC content also increased by 0.03, 0.02, and 0.01 dS/m, compared to the control. In horticultural land contaminated with pesticides, the highest EC at 40 and 30 t/ha SC is 0.08 dS/m but appears to be the same at 0 to 20 t/ha SC (Table 1). According to Aydin et al. (2012), SC can increase EC in the soil. The dissociated carboxyl group on SC will generate a negative charge, forming an external electric field in the soil solution. The mobility of singly charged ions is controlled by a simple force balance of the electric field and the viscosity of the solution.

The mineral composition is calculated from the ash content and also the OM composition through the dry ashing method at 550°C for the two types of land use. Table 1 shows that the two types of land use have a higher mineral composition than OM. It explains that the land is classified as mineral soil, namely Inceptisols. The secondary forest has a mineral composition of 70.80% and 29.20% OM, while on horticultural land contaminated with pesticides, the mineral composition is 77.00% and 23.00% OM. This confirms that the mineral composition of horticultural land contaminated with pesticides is higher than that of secondary forest. Still, the OM composition of secondary forest is higher compared to horticultural land contaminated with pesticides. The high mineral composition is closely related to the clay content found in horticultural land contaminated with pesticides.

In contrast, the high OM composition is affected by the input of organic matter, such as litter in secondary forests (Table 1B). Table 1A shows that the effect of SC had a significant on decreasing the mineral composition and increasing the OM composition. The decrease in mineral composition in the secondary forest is highest at doses of 40 and 30 t/ha SC of 7.60%, while 20 t/ha SC was 5.60%, but it looks the same at 10 t/ha SC (70.60%) with the control (70.80%). The increase in OM composition in secondary forests occurred due to the application of SC, which directly adds organic matter composition to the soil. The effect of 30 and 40 t/ha of SC can increase organic C by 4.40%, where 20 t/ha SC is 3.24%, but it looks the same at 10 t/ha SC (17.05%) with the control (16.94%). The high mineral composition of horticultural land



contaminated with pesticides causes a decrease in the mineral composition after the SC effect of 3.00% and 2.60% at a dose of 40 to 20 t/ha SC, but the decrease is different at 10 t/ha SC (2.40%), compared to the control. The effect of SC on increasing the composition of OM in horticultural land contaminated with pesticides According to Gill et al. (2015), SC has an ash content of 34.90% to 46.30% and a C content of 33.68% to 44.28%. SC polymers have a six-carbon aromatic ring connected by various groups. SC is also a high molecular weight cyclic organic compound with long chains and active carboxyl and phenolic groups as importers and binders of cations and anions at certain pH (Khaled & Fawy, 2011). The carboxylates of some carboxyl groups are released at pH 6, leaving a negative charge on the functional groups (Turan et al., 2011). Dissociation of  $H^+$  from amides (NH) can also increase the negative charge. Although there are positively charged protonated groups, overall, the humus is negatively charged.

The CEC is an important indicator for explaining the NCA in the soil. The SC-based amelioration technology for CEC on the two types of land looks different, where the CEC on horticultural land contaminated with pesticides is higher than in secondary forests. The high CEC, one of which is influenced by the percentage of clay in the two types of land used, SC effect has a significant on increasing CEC in both types of land. In the secondary forest, the CEC is 30.25 cmol(+)/kg (control), and it increases with increasing SC doses. The effect of 40 t/ha SC increases CEC by 44.30 cmol(+)/kg, while the effect of 10, 20, and 30 t/ha SC increases CEC by 20.37, 23.72, and 32.40 cmol(+)/kg compared to the control (Table 1A). The effect of SC also increases CEC on horticultural land contaminated with pesticides, with an increase of 26.89 cmol(+)/kg at a dose of 40 t/ha SC. In comparison, the application of 30 t/ha SC increases CEC by 24.48 cmol(+)/kg, but the increase in CEC is the same at doses of 10 and 20 t/ha SC, at 19.99 and 17.04 cmol(+)/kg, respectively, compared to the control (Table 1B). The increase in CEC in both fields from the administration of SC was due to the contribution of negative ions from the phenol and carboxyl groups in SC. Cation adsorption by SC follows a lipotropic sequence. Cations are in solution or adsorbed by SC exchange with clay. Cations are rapidly absorbed by roots, enhancing

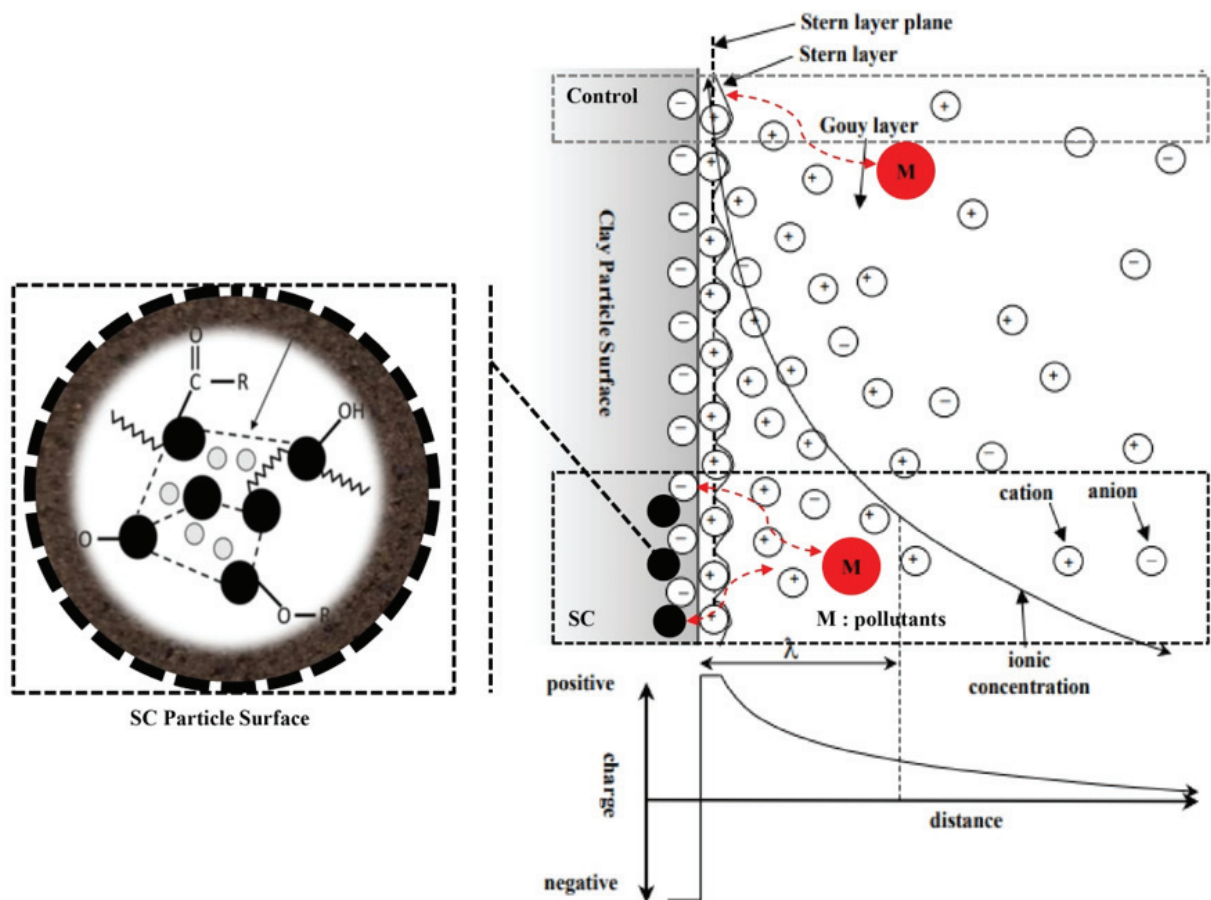
micronutrient transport to the soil system and plants. SC can adsorb cations or metals through (a) direct adsorption, (b) complexation or outer sphere interaction to hydrate, (c) serving as a cation bridge through direct or indirect chelation, and (d) contact with aggregates or amine groups (Sharma & Kappler, 2011). SC, or clays, have a high affinity for phenolic and carboxyl groups. Simple cation exchange with carboxyl groups can retain base cations (Zhang et al., 2013). Changes in CEC impact the electrochemical properties of clays, which are directly related to the amount of complexes present, their chemical composition, and charge properties. Various charges have been found in partially neutralized solutions used to coat clays. The number of layers in the CEC of clay affects the PZC (Preocanin et al., 2016).

Net proton surface charge density ( $\delta H$ ) and zero charge point (pH PZC) are the differences between moles of protons and moles of hydroxide ions complexed by surface functional groups. The importance of pH stems from the wide and dynamic range of proton concentrations in soil solutions, as well as the fact that complexation on the surface of soil particles has a great impact on the adsorption of other cations and anions. According to Mindari et al. (2014), Changes in the chemical characteristics of the molecular surface can be related to changes in the ionic strength of the absorption. The net negative charge on the molecule provided by the SC grows as ionic strength increases. As a result, for anions that can bind via simple coulomb forces, an increase in ionic strength reduces absorption above the zero charge threshold. SC can provide negative charges on the surface and provide more binding sites. According to Preocanin et al. (2016), The pH value of PZC represents the zero charge point, which can affect the concentration levels of other surface complexing ions and directly affect the P value. The three primary zero charge points are generally named (1) zero charge point (PZC), (2) net zero proton charge point (PZNPC), and (3) net zero charge point (PZNC). The total net charge of ions bound to the inner-sphere surface coordination determines the surface charge density of the complex ( $\delta IS$ ). The formation of one or more bonds directly between pollutant molecules and SC surface functional groups without the interposition of water molecules is referred to as inner-sphere complexation. The total net charge of

ions bound to the coordination of the outer-sphere surface determines the surface charge density of the complex ( $\delta OS$ ). The outer sphere complex is formed by water interposed between the pollutant and the SC surface.

Increases in pH, EC, CEC, and OM will increase soil NCA indirectly. This alteration is significant in an endeavor to improve nutrient availability and pesticide absorption, particularly on horticultural land (Fig. 2). Amelioration technology on two types of land with the influence of SC shows an increase of NCA on the surface of soil colloids

( $\Delta pH$ ) of 43% (SF) and 23% (HL-P) at a dose of 40 t/ha SC, where the pH of PZC is lower than pH  $H_2O$ , for both types of land used for each increase in the application of SC dose. Table 1 also shows that the pH, EC, OM, and CEC in secondary forests are higher compared to horticultural land contaminated with pesticides. Secondary forests have a greater range of negative charges on the colloidal soil surface compared to pesticide-contaminated horticultural land. The NCA is high and is expected to buffer pesticide pollution on agricultural land through the clay-SC complex.



**Fig. 2.** Mechanism modification on electric double layer (EDL) in colloidal clay with sub-bituminous coal application

## CONCLUSION AND SUGGESTION

The effect of 40 t/ha SC on two types of land through amelioration technology significantly affects the increase in pH H<sub>2</sub>O, EC, CEC, and OM composition. The increase is 0.70; 0.04 dS/m; 44.30 cmol(+)/kg and 7.60% in SF and 0.33; 0.01 dS/m; 26.89 cmol(+)/kg and 3.00% in HL-P, respectively, compared to control. The effect of 40 t/ha SC can increase NCA on the soil colloid surface ( $\Delta$ pH) by 43% on SF and 23% on HL-P based on pH PZC below pH H<sub>2</sub>O. Thus, the effect of 40 t/ha SC can simultaneously improve nutrient composition and the absorption of pesticides.

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