Response of Andisols to intensive agricultural land use: Implication on changes in P accumulation and colloidal surface charge

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Abstract. The response of Andisols to intensive agricultural practices were not fully explored although intensive cultivation has been practiced for a long period of time. The objective of the review is to bring together all findings, which have been documented with the focus on changes in soil organic matter, P content and retention, and colloidal surface charge. The Andisols contain an abundance of nano-clay consisting mainly of hollow allophane spherules forming globular aggregates of about 100 nm in diameter, indicating the large reactive surface area. A huge quantity of P has been accumulated in the uppermost two soil horizons, deriving from residue of continuous application of P fertilizer. Andisols have a strong ability to retain organic C through ligand exchange of metal hydroxyl functional groups with organic functional groups. The soil colloids bear a positive charge at soil pH less than 5. The colloidal negative charge is continuously increased with increasing soil pH over 5.0. Hence, the Andisols should be managed by maintaining soil pH above pH 5.0 by adding lime or compost to allow soil capability to retain cation and reduce P fixation. Nano-clay has positive impact on carbon stabilization and therefore C sequestration.

1. Introduction

Indonesia is located in the areas called the ring of fire and has 127 active volcanoes out of ~450 total volcanoes of the ring, resulting in widespread of Andisols. Many of these Andisols are intensively used for agriculture owing to their high productivity and Indonesian Andisols have been shown to be the world's highest human-carrying capacity [1, 2].

In Indonesia, the majority of land use/land cover of Andisols is primarily native rainforest, tea plantation, horticultural crops, terraced paddy fields and other food crops [3]. Agricultural practices on Andisols have taken place over long periods of time (>100 years) and it seems that these practices resilience with the properties of Andisols. Hence, it is necessary to evaluate the response of Andisols experiencing a long time of agricultural practices concerning phosphate fertilizer application and changes in soil organic matter and colloidal surface charge.

The unique properties of Andisols are high P fixation, high organic matter content, a clay-size fraction dominated by nanocrystalline materials (allophane, imogolite and ferrihydrite), low bulk density, high porosity, high water retention capacity, high mesopore and colloidal surface charge dominated by pH-dependent charge [1, 3–6]. In addition, Andisols have many unusual and distinctive morphological features, including dark topsoil colour, very friable/crumb and granular structure,
smeary, different inter-layering associated with periodic volcanic material deposition, and high pH in NaF solution (pH >9.5). Chemical properties are characterized by high variable charge of colloidal surfaces, low base status, and high anion exchange capacity [7, 8].

The nanocrystalline materials of Andisols consisted of allophane, imogolite and ferrihydrite. The Allophane is a noncrystalline or ‘short-range order’ aluminosilicate with an Al/Si ratio between 1 and 2. The allophane has a hollow spherule with an outer diameter of 3.5 to 5.0 nm [9, 10]. The spherule wall is composed of an outer Al octahedral (gibbsitic) sheet and an inner Si sheet with 0.7-1.0 nm thick. The perforated wall structure about 0.3 nm diameter was attributed to defecton [11–13]. Chemical structure of allophane is given in figure 1, showing outer and inner sphere diameters of 3 to 5 nm and 0.3 to 0.5 nm, respectively.

The nanocrystalline materials formed globular aggregates of about 100 nm in diameter and mainly built up by hollow allophane spherules [14]. Compared to clay, the nano-clay contains more organic matter (carbon and nitrogen) consisting of mainly polysaccharides and nitrogen, and has a lower isoelectric point [14]. The reactivity of colloidal surfaces is influenced by total surface areas. The surface areas of spherule allophane varied between 700 to 1000 m² g⁻¹, depending on the method used, being high in the unit particle size and density, and low in the retention of ethylene glycol and ethylene glycolmonoethyl ether [15, 16]. The mineralogical properties of allophane and imogolite type materials in Andisols, provide the high reactive surface area and accounted for the C preservation [17–19]. The objective of the review is to bring together all findings, which have been documented with the focus on changes in soil organic matter, P content and retention, and colloidal surface charge.

![Figure 1](image)

Figure 1. Chemical structure of allophane. A: Full structure; B and C: Atomic arrangement in the cross-section at pore region [20]
2. Phosphate fixation in Andisols
Andisols have high P retention (>85%) [21] and can limit agricultural productivity associated with limiting plant availability of P. It is well known that Andisols have a specific P adsorption. The specific adsorption occurs through a ligand exchange reaction by incorporating anions in the coordination shell of Fe or Al atoms [22]. According to Shoji et al. [1], the ligand exchange reaction takes place through the anions react with active Al and Fe compounds to form covalent bonds or inner-sphere complexes; and the hydroxyl functional groups on the surface of active Al or Fe compounds are exchanged by anions. At present, there is little information on how P retention and availability in tropical Andisols change with different land use and agricultural practices.

Table 1. The response of Andisols to phosphate fertilizer under intensive agricultural practices.

| Location/Sample | Depth cm | Texture | P$_2$O$_5$ HCl 25% mg 100 g$^{-1}$ | Land cover/land use | P$_2$O$_5$ ratio$^a$ |
|-----------------|----------|---------|---------------------------------|---------------------|------------------|
| Lower slope of Galunggung volcanic mountain in West Java | VG4/I 0-25 | 52 38 10 | 314 | Horticultural crops | 11 |
| VG4/II 25-60 | 52 35 13 | 330 | | Natural forest | 11 |
| VG2/I 0-20 | 62 31 7 | 29 | | Natural forest | 11 |
| VG2/II 20-40 | 52 35 13 | 31 | | | |
| Lower slope (VP4) and middle slope (VP3) of Gede volcanic mountain, West Java | VP 4/I 0 - 20 | 17 57 26 | 457 | Horticultural crops | 9 |
| VP 4/II 20 - 40 | 26 54 20 | 455 | | Natural forest | 12 |
| VP 3/I 0 - 20 | 47 40 13 | 49 | | Natural forest | 12 |
| VP 3/II 20 - 40 | 56 27 17 | 37 | | | |
| Middle slope of Pangrango volcanic mountain, West Java | VP7/I 0 - 20 | 50 18 32 | 133 | Tea plantation | 2 |
| VP7/II 20 - 40 | 63 20 17 | 107 | | Natural forest | 1 |
| VP8/I 0 - 20 | 48 29 23 | 74 | | Natural forest | 2 |
| VP8/II 20 - 40 | 45 29 26 | 72 | | | |
| Lower slope of Sindoro volcanic mountain in Central Java | WD1/I 0 - 20 | 29 40 31 | 274 | Horticultural crops | 3.6 |
| WD1/II 20 - 40 | 31 46 23 | 248 | | Natural forest | 4.9 |
| Plateau of Dieng volcanic mountain in Central Java | WD 5/I 0 - 20 | 22 56 22 | 1,067 | Horticultural crops | 13.9 |
| WD 5/II 20 - 40 | 18 59 23 | 1,078 | | Natural forest | 21.1 |
| Middle slope of Ijen volcanic mountain in East Java | VBVW11/I 0 - 20 | 31 35 34 | 152 | Horticultural crops | 3 |
| VBVW11/II 20 - 40 | 21 39 40 | 181 | | Natural forest | 3 |
| VBVW5/I 0-20 | 43 36 21 | 50 | | | |
| VBVW5/II 20-40 | 43 35 22 | 58 | | | |
| Volcanic plain of Kerinci mountain in Jambi, Sumatera | VK5/I 0 - 20 | 23 45 32 | 82 | Tea plantation | 1.1 |
| VK5/II 20 - 40 | 23 50 27 | 45 | | Natural forest | 0.9 |
| VK7/I 0 - 20 | 33 48 19 | 394 | | Horticultural crops | 5 |
| VK7/II 20 - 40 | 35 43 22 | 177 | | Natural forest | 3 |
| VK2/I 0 - 20 | 23 56 21 | 77 | | | |
| VK2/II 20 - 40 | 45 36 19 | 51 | | | |

Note:
$^a$P$_2$O$_5$ ratio is the value in the given agricultural land use divided by the corresponding value in natural forest;
$^b$For Sindoro and Dieng in central Java, the P$_2$O$_5$ of natural forest is not available, hence the highest value of the natural forest is used for comparison, i.e. the P$_2$O$_5$ of VK2 site; Source: unpublished data by the senior author
Van Ranst et al. [23] showed P-fixation and/or P-retention are serious problem that needs strategic management when using Andisols for crop production in Java Island. They compared Andisols properties in East, Central and West Java Provinces and showed that the soil’s mineralogical and chemical properties are considerably different. The allophane content is found to increase from East to West Java, corresponding to the increase in active Al and Fe, which in turn increase in P-fixation and/or P-retention. They concluded that the P-fertilizer strategy for the Andisols in Java Island can be divided into three regions: the East, Central and West Java, where phosphorus requirements, being lower in the east as compared to the West Java.

Anda and Dahlgren [3] compared the Andisol P-fixation in rainforest and agriculture, and reported that the high P fixation (>97%) occurred under forest vegetation. However, P fixation decreased (<87%) in the upper 40 cm of the soil profile under intensive horticultural crops owing to the application of horse manure resulting in the P availability of 3.0 to 7.7 mg kg⁻¹. In Andisols, maximum sorption of phosphate mostly occurs in the pH range of 3.0 to 4.5 and decreases with increasing soil pH [24].

The response of Andisols on continuous regular application of phosphate fertilizer in agricultural practices showed a dramatic huge accumulation of P-residue (Table 1). This response was assessed by comparing the phosphate content of Andisols under natural forest and Andisols under intensive horticultural crops. Representative Andisols in East, Central, and West Java Provinces and that in Jambi (Sumatra) showed P₂O₅ content under rainforest varied from 29 to 77 mg 100g⁻¹ that dramatically increased varying between 152 and 1078 mg 100 g⁻¹ (mostly 248 to 457 mg 100 g⁻¹) under horticultural crops. The data indicated the accumulation of P-fertilizer has achieved 3.6 to 13.9-fold greater under intensive horticultural land use as compared to the natural conditions (table 1). For Andisols used for tea plantation, the P-residue fertilizer achieved 82 and 133 mg 100 g⁻¹ (1.1-2.0-fold compared to natural forest), which was much lower than Andisols used for horticultural crops.

The huge increase in P-residue fertilizer under horticultural crops was associated with 3 to 4 growing seasons each year, where the farmers continuously applied P-fertilizer for each growing season. The high cumulative accumulation of P-residue application each year and has been practiced for a long period of time (about 50-100 years) accounted for the high phosphate content in the surface layer of Andisols under horticultural crops. This continuous practice is related to no soil test in the farming of farmers. Hence no information on phosphate status content and its availability in the soils. In the case of Andisols used for tea plantation, the low residue of P-residue application was owing to less frequency and low rate of P-fertilizer application. This is due to the state company is regularly tested the P content status in soils and thereby applied P-fertilizer when it is needed according to tea crop requirement.

### 3. Surface colloidal charge of Andisols

Soil surface charges derive from multi-site surfaces including inorganic functional groups (hydroxyl of Si, Al and Fe) and organic functional groups (enolic, phenolic, and carboxyl) [25, 26]. Type of colloidal surface charges consists of negative and positive charges, which are responsible for retaining cations and anions, respectively. Based on the origin and behaviour of colloidal surface charge, the soil charge was distinguished into variable charge and permanent charge components [27–29]. Soil surface charge may be dominated by either permanent charge or variable charge or their mixture [27], depending on the degree of weathering and type of mineral constituents.

Two parameters are commonly used to assess soil surface charge characteristics, namely point zero charges (PZC) and point of zero net charges (PZN). The PZC is the point on the pH scale where equal number of protonated and deprotonated sites on colloidal surfaces of the soil variable charge component [30]. The point of zero net charges (PZN) is defined as the pH where the number of cation exchange sites equals the number of anion exchange sites, deriving from both variable and permanent charge sites.

In Andisols, the colloidal surface charge is dominated by the variable charge component. The study on charge characteristics of tropical Andisols by Anda and Dahlgren [6] showed the PZC and PZN
values decreased following conversion from rainforest to horticultural crops, suggesting generation of negative charge. PZC for pine forest was 4.5-4.9 and decreased slightly to 4.4-4.7 for soils in horticultural crops (Table 2). The corresponding values for PZN C were 4.4-5.2 in the pine forest and decreased to 3.6-4.2 in intensive horticultural crops. The Andisols under pine forest bear net positive charge ($N_{p,C}$) varied between 0.01 and 4.2 cmol kg$^{-1}$ and consistent with the magnitude order of PZN C > PZC > pH values. In contrast, Andisols under intensive cultivation for horticultural crops bear net negative charge varying between 6.0 and 20 cmol kg$^{-1}$ and consistent with the magnitude order of pH > PZC > PZN C values. The implication of this information is to increase soil pH over PZN C values, i.e., above pH 4.4 and could be raised to over pH 5.0 and even as at pH 5.9 to generate a high amount of negative charge (20 cmol kg$^{-1}$) (table 2).

**Table 2.** Surface charge of Andisols at natural pH conditions under different land use/management systems.

| Horizon            | Depth cm | $^a$Soil pH | $^b$CEC$_B$ | $^c$CEC$_A$ | $^d$CEC$_T$ | $^e$AEC | $^f$N$_{p,C}$ | $^g$PZN C | $^h$PZC |
|--------------------|----------|-------------|-------------|-------------|-------------|--------|--------------|----------|--------|
| PF1 Typic Hapludand under pine forest | A 0-24 | 4.43 | 1.10 | 0.08 | 1.19 | 1.19 | +0.01 | 4.45 | 4.47 |
|                    | Bw1 24-60 | 4.22 | 0.04 | 0.06 | 0.10 | 3.41 | +3.31 | 5.19 | 4.80 |
|                    | Bw2 60-100 | 4.24 | 0.05 | 0.05 | 0.10 | 3.15 | +3.15 | 5.17 | 4.75 |
|                    | Bw3 100-140 | 4.27 | 0.07 | 0.06 | 0.13 | 3.33 | +3.20 | 5.17 | 4.85 |
| TP1 Typic Hapludand under tea plantation | Ap 0-24 | 4.43 | 0.62 | 0.08 | 0.70 | 0.94 | +0.25 | 4.50 | 4.77 |
|                    | Bw1 24-54 | 4.50 | 0.47 | 0.08 | 0.55 | 0.65 | +0.10 | 4.50 | 4.64 |
|                    | Bw2 54-88 | 4.51 | 0.53 | 0.06 | 0.59 | 1.67 | +1.08 | 4.82 | 4.80 |
|                    | Bw3 88-130 | 4.30 | 0.06 | 0.04 | 0.10 | 4.28 | +4.18 | 5.37 | 4.89 |
| HF1 Typic Melanudand under horticultural crops with fallow system | Ap 0-21 | 4.95 | 7.63 | 0.01 | 7.64 | 0.63 | -7.01 | 4.20 | 4.22 |
|                    | Bw1 21-48 | 5.32 | 10.73 | nd | 10.72 | 0.36 | -10.36 | 4.16 | 4.71 |
|                    | Bw2 48-70 | 5.38 | 14.14 | nd | 14.13 | 0.32 | -13.82 | 3.95 | 4.57 |
|                    | Bw3 70-118 | 5.36 | 6.17 | nd | 6.16 | 0.59 | -5.57 | 4.55 | 5.06 |
| IH1 Typic Melanudand under intensive horticultural crops with horse manure | Ap 0-21 | 5.86 | 19.97 | nd | 19.97 | 0.01 | -19.96 | 3.64 | 4.45 |
|                    | Bw1 21-48 | 5.70 | 15.15 | nd | 15.15 | 0.54 | -14.61 | 3.75 | 4.38 |
|                    | Bw2 48-70 | 5.27 | 10.14 | nd | 10.14 | 0.56 | -9.59 | 4.18 | 4.55 |
|                    | Bw3 70-110 | 5.40 | 13.83 | nd | 13.82 | 0.46 | -13.37 | 4.04 | 4.70 |

Notes:

$^a$Soil pH was measured in 1:10 soil:solution ratio (0.002 M CaCl$_2$);

$^b$CEC$_B$, negative charge for base cations (Ca adsorbed);

$^c$CEC$_A$, negative charge for acid cations (Al adsorbed).

$^d$CEC$_T$, total negative charge (Ca + Al adsorbed);

$^e$AEC, positive charge (Cl adsorbed);

$^f$N$_{p,C}$, net charge (AEC - CEC$_T$);

$^g$PZN C = point zero net charge;

$^h$PZC = point zero charge.

The + and – signs of N$_{p,C}$ and PC correspond to net positive and net negative charge, respectively, nd = not detected. Adapted from Anda and Dahlgren [6]

Van Ranst et al. [2] studied physico-chemical properties of Andisols of East-West Java sequence to assess the influence of parent material changes on soil properties. They reported the PZN C and PZC increased from east Java to West Java Provinces, indicating the development of net positive variable charges from east to west Java and led them to argue that East Java is capable of holding more basic
cations than Central and West Java at natural soil pH with the impact that the Andisols in east java are less likely to be deficient in Ca and Mg. The presence of positive charge in Andisols has been reported by measuring the anion exchange capacity (AEC) of allophane-rich soils and reported the value ranged from 0.4 to 12.2 cmol, kg$^{-1}$ [31–33]. The positive charge is accounted for the high retention of anions such as NO$_3$, SO$_4$ and PO$_4$.

The high values of PZC and PZNC in Andisols could be lowered using humic substances [2, 34–36]. This is consistent with PZNC values reported by Anda and Dahlgren [6] in which the Andisols have a lower PZNC in surface horizons having higher organic matter compared to subsoils with low soil organic matter content. Soil organic C concentrations exceeded ~30 g kg$^{-1}$ in soil horizons of tropical Andisols, which led to conclude that humic substances play an important role in regulating the magnitude of soil’s PZNC [6].

Other studies have reported organic matter is responsible for lowering soil PZNC, while Fe (hydroxides) are responsible for increasing soil PZNC [35–37]. This relationship was demonstrated by Anda et al. [38] by removing soil organic matter and found the increased PZC of Oxisols from 3.9-5.7 to 5.3-6.7. In addition, the removal of SOM and iron oxides resulted in the decrease in PZC from 3.9-5.7 to 3.3-4.5, corresponding to the increase in the amount of negative charge.

4. Organic C preservation and management of Andisols

Soil carbon storage is dependent on soil mineral constituents, with volcanic ash soils typically having exceptionally high potential C stocks owing to their high content of active Al and Fe constituents (e.g., allophane, imogolite, ferrihydrite and metal-organ complex). Preservation of soil organic matter (SOM) in Andisols has been reported by many workers with various findings, among them are i) association of the SOM with mineral and/or with metallic (Al/Fe–humus) to form complexes [24, 39], ii) low activity of soil microorganisms owing to acidic soil pH and Al toxicity [40, 41], iii) physical protection of the SOM in stable microaggregates [42, 43], and iv) burial of organic-rich surface horizons by periodic additions of air fall tephra deposition [44].

Matus et al. [45] reviewed soil carbon storage and stabilization in andic soils and concluded that the ligand exchange is the most important mechanism of soil organic matter sorption by short range ordered amorphous minerals. The nanoclay contains more organic matter (carbon and nitrogen) with a larger proportion of polysaccharides and nitrogen-containing compounds, and has a lower isoelectric point than the coarse clay (non-nano clay) [14].

Concerning greenhouse gases, Andisols are notable for having the highest soil carbon storage capacity compared to other mineral soil Orders in temperate and tropical climatic regimes with an average carbon stock of 25.4 kg C m$^{-2}$ [46]. Anda and Dahlgren [3] found soil carbon and nitrogen stocks in the upper 1 m of tropical Andisols were higher in agricultural soils (25-29 kg C m$^{-2}$; 1.7–2.3 kg N m$^{-2}$) than forest soil (17 kg C m$^{-2}$; 1 kg N m$^{-2}$) and they distributed from topsoil to subsoil horizons.

The intensive use of Andisols for horticultural crops with the additions of organic manure increased extractable mineral N (especially NO3), reduced P fixation, increased available P, higher pH, and higher concentrations of exchangeable base cations (Ca$^{2+}$, Mg$^{2+}$ and K$^{+}$) [3]. This indicates the Andisol properties may resilient with agricultural practices if they are managed properly. In Italy, the Andisols at high slope steepness and experienced long-anthropic pressure (70 years) showed low run off and minimal soil erosion owing to good infiltration and conservation practices [47].

Applications of lime and horse manure were effective in raising the pH and base saturation of tropical Andisols from very strongly acidic in the rainforest to slightly acid in the horticultural soils [3]. The increased base saturation is associated with the presence of soluble salts (Ca, Mg, and K) derived from lime, horse manure and K fertilizer application. Therefore, the application of animal manure and lime appears to be an effective nutrient management strategy to enhance P availability in Andisols [3].

It seems that the Andisols under intensive cultivation for horticultural crops have accumulated huge amount of phosphate due to high fertilizer residue and the preference of metal functional groups to
adsorb phosphate. The implication is to take advantage of phosphate residue by omitting P-fertilizer and promoting the P-dissolution or availability from phosphate residues using solubilizing phosphate microorganisms, especially under tropical conditions in Indonesia. According to Stevenson and Cole [48] the application of synthetic phosphorus fertilizers to Andisols was absorbed by plants only between 5 and 25% while the remaining (75–95%) was fixed or retained in insoluble forms by soil colloids, active complexes of Al, Fe, Ca and organic matter. Adesemoye et al. [49] reported that the application of microorganisms, such as phosphate solubilizing bacteria could increase soluble P in soils which is an available P form for crops. The inoculation of Bacillus thuringiensis has been shown to increase the soluble P in soil, resulting in a higher crop yield [50].

5. Conclusions
Active Al and Fe of amorphous materials play the main role in P accumulation in the topsoils. A huge quantity of P-fertilizer residue has accumulated in the topsoils under intensive cultivation for agriculture suggests the potential to reduce or omitted the P-fertilizer application. Attempts should be made using microbial solubilizing P to increase P availability for crops.

Colloidal surfaces of Andisols was dominated by variable charge component with PZNC of 4.4-5.2 in the rainforest and decreased to 3.6-4.2 in intensive horticultural crops, corresponding to the increase in a colloidal negative charge. Organic manure and lime application are effective to decrease PZNC and increase soil negative surface charge. The implication of surface charge determination is the soil pH should be maintained above pH 5.0 in the management of Andisols.

A mineralogical constituent of Andisols was dominated by nano-clay and has high capacity to preserve soil organic carbon and therefore has a positive impact in C sequestration, which contributes to greenhouse gas mitigation.

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