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Levels and sources of potassium, calcium, sulfur, iron and manganese in major paddy soils of the Philippines

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Abstract—Nutrient levels in rice areas can be translated to the soils' capacity to supply the essential elements for rice, while information about nutrient sources can be used to formulate management options to areas deficient or toxic in nutrients. This study was then conducted to assess the potassium (K), calcium (Ca), sulfur (S), iron (Fe) and manganese (Mn) levels in the Philippines' major rice areas. The study also determined the contributions of soil properties, land uses, irrigation water, and farm practices to the nutrient levels. Strategic collection of soil and plant samples, laboratory analyses of samples, and farmer interviews were done. Potassium concentrations of rice plants in La Paz (Tarlac) and Sta. Rosa City (Laguna) exceeded the toxic concentration of 3%. These K concentrations can be due to the increase of the nutrient's availability owing to the neutral soil pH levels in the two sites. The highest total Ca levels were found in the Polangui (Albay), La Paz, and Villasis (Pangasinan) soils. These Ca levels can be ascribed to the occurrence of Ca in sand and silt-sized soil fractions. Total Fe and Mn concentrations were very high in the Sta. Cruz (Zambales) soil due to the input of mine wastes. The San Leonardo (Nueva Ecija) soil's high total Fe concentration is probably due to the occurrence of Fe in clay-sized soil fractions. Iron and Mn levels of rice in most Central Luzon sites exceeded the toxic concentrations of 1000 mg kg^{-1} for Fe and 300 mg kg^{-1} for Mn. These concentrations can be due to the high total soil Fe and Mn levels, and their increased plant uptake due to periodic soil submergence and increased soil acidity from continuous cropping. The study implies that K, Ca, Fe and Mn are enriched in rice areas due to soil properties and farm practices.

Keywords—potassium, calcium, sulfur, iron, manganese, paddy soils

INTRODUCTION

Rice is mostly grown in flooded lowland conditions in bunded fields or paddy soils. Paddy soils are characterized by a plow plan and specific morphological features resulting from tilling the wet soil (puddling) and periodic flooding and drainage regimes.

Philippine paddy soils have higher Fe oxide (7.73%), Mn oxide (0.23%) and Ca oxide (2.83%) contents, but have lower potash (K_2O) (0.99%) contents than most paddy soils in tropical Asia. Philippine paddy soils also have relatively high levels of exchangeable macronutrients such as Ca (14.8 cmol(+)kg^{-1}) and K (0.5 cmol(+)kg^{-1}) (Kawaguchi and Kyuma 1974). Such total and exchangeable levels of soil nutrients may become available to rice depending on the nutrients' chemical nature, on soil properties, and on crop management and environmental factors.

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Soil retention of nutrients is controlled by soil properties (Zeng et al. 2011) and by the intensity of puddling operations employed in rice fields (Sharma and De Datta 1984). Use of fertilizers increases the concentrations of some nutrients in soils and in plants, while the use of pesticides that contain S can result in elevated concentration of this nutrient (Lu 2011). Irrigation water affects the soil nutrient levels depending on the water's nutrient concentration and irrigation intensity. Intensive cropping reduces the soil nutrient levels if the amounts of removed nutrients exceed those returned to the soil. Based on nutrient bioavailability, the amount of nutrients retained in the soil determines the amount absorbed by rice.

Some agricultural soils are near the perceived heavy metal sources like urban areas, highways, and mining areas. Higher quantities of metals are dissolved in the water or suspended in the air of such areas, where soils and plants can be enriched with metals.

Such scenarios highlight the need for assessing the levels and sources of nutrients in the major rice-producing areas of the Philippines. Nutrient levels in paddy soils and in rice can be translated to the soils' capacity to supply the essential elements for rice. Nutrient sources, on the other hand, can be used to formulate management options in areas that are deficient or toxic in nutrients.

The study assessed the K, Ca, S, Fe and Mn levels in soils and plants of the Philippines' major rice areas. It also determined the sources of these nutrients relative to soil properties, land uses, irrigation water, and farm input levels. The study sites were selected based on a detailed morphological, mineralogical, chemical and physical characterization of lowland soils in the Philippines (Miura et al. 1995). These sites are the major soil series devoted to lowland rice production across different agroclimatic conditions of the country's rice-growing areas. Nutrient values established by the present study can be a good reference for future studies that will assess the nutrient levels of rice and paddy soils in the country.

MATERIALS AND METHODS

Sampling Sites

Thirty-two sites were studied: three in Cagayan Valley, 14 in Central Luzon, four in Laguna, five in Bicol, and three each in the Visayas and Southern Mindanao (Figure 1, Table 1). Thirty sites are rice areas, while two control sites—Jaen and Sta. Rosa City a—are mango orchard and uncultivated grassland, respectively.

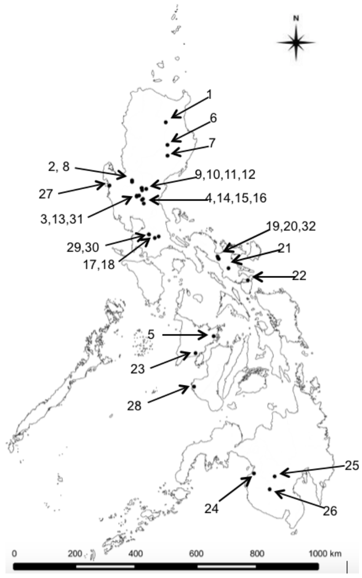

Figure 1. Map of the Philippines showing the study sites (labels in Table 1)

TABLE 1. Classification of sites based on land use or contamination source.

Classification	Site
Deep-well and rainfed sites without solid wastes in irrigation water	1) Solana, Cagayan; 2) Villasis, Pangasinan; 3) La Paz, Tarlac a; 4) Jaen, Nueva Ecija (N.E.); 5) Sara, Iloilo
Dam and river-irrigated sites with solid wastes in irrigation canals	6) Cabatuan, Isabela*; 7) Echague, Isabela*;
	8) San Manuel, Tarlac*; 9,10) Muñoz City, N.E. a*, b*; 11) Llanera, N.E. *; 12) Talavera, N.E. *;
	13) Zaragoza, N.E.; 14,15) San Leonardo, N.E. a*, b*;
	16) San Miguel, Bulacan*; 17) Sta. Cruz, Laguna;
	18) Bay, Laguna; 19) Canaman, Camarines Sur;
	20) Minalabac, Camarines Sur*; 21) Polangui, Albay*;
	22) Casiguran, Sorsogon*; 23) San Miguel, Iloilo*; 24) Cotabato City, Maguindanao;
	25) Kabacan, North Cotabato*; 26) Tacurong City, Sultan Kudarat*
Mine area-adjacent sites with deposits of mine tailings	27) Sta. Cruz, Zambales; 28) Sipalay City, Negros Occidental*
Urban area-adjacent sites with urban wastes	29, 30) Sta. Rosa City, Laguna a, b
Highway-adjacent sites with vehicular wastes	31) La Paz, Tarlac b; 32) Milaor, Camarines Sur

*sites subjected to comparison of nutrient levels at varying distances from irrigation entrance

Sample Collection Protocol

Plant and soil samples were collected during the period between maximum tillering and harvesting of rice. Rice plants were uprooted. Rhizophere soils and the whole rice-aboveground biomass (tops) were placed in clean separate plastic bags. Rhizosphere soils were collected to determine the relationship between soil properties and plant nutrient levels. Two or three soil and plant samples were collected per site depending on the lengths of the farm. Samples were collected from May 2012 to December 2013.

Levels of Nutrients

Soil samples were air-dried, pulverized, and passed through 2-mm (US#10) and 0.422-mm (US #40) mesh sieves. Whole aboveground biomass was washed, oven-dried for 3 days at 60-70^{0}C, and ground using a stainless steel grinder. Non-destructive analyses of total nutrient concentrations of soil and biomass samples were performed in the National Institute of Molecular Biology and Biotechnology, University of the Philippines Los Baños, Laguna. The equipment used was the handheld NITON® XL3T x-ray fluorescence analyzer manufactured by Thermo Fisher Scientific Inc.

Total nutrient levels in soils were compared to the mean of total nutrient levels in Philippine paddy soils (Miura et al. 1995, Domingo and Kyuma 1983). Nutrient levels in plants were compared to the values reported as normal, toxic and deficient in mature leaf tissues for various species and for rice tops. Levels reported by Kabata-Pendias and Pendias (2001), Marschner (2012), Kitagishi and Yamane (1981), and Dobermann and Fairhurst (2000) were used.

Soil Properties and Land Uses that affect Nutrient Levels

Soil samples were analyzed for pH, available phosphorus (P), and organic matter (OM) following standard procedures (PCAARRD 1980). Correlation analyses of data on soil properties and total nutrient levels in soils and in plant tissues were done. Nutrient levels in different land uses (deep-well and rainfed areas, dam and river-irrigated areas, mine area-adjacent sites, urban area-adjacent sites, and highway-adjacent sites) (Table 1) were compared. Kruskal-Wallis and Mann-Whitney U tests were used.

Irrigation Water that affects Nutrient Levels

Soil and plant samples were collected from dam-irrigated sites to determine the effect of irrigation water on nutrient levels in paddy fields (Table 1). Collections were done at three varying distances from the irrigation entrance: close (2–5 m), middle (42–45 m), and far (82–85 m).

Analyses of total Fe and Mn concentrations of irrigation water samples from mine area-adjacent sites (Table 1) were done in the Regional Standards Testing Laboratory, Department of Science and Technology Regional Office III, San Fernando City, Pampanga. Water samples were digested using nitric and hydrochloric acids, and their total Fe and Mn concentrations were measured in a flame atomic absorption spectrophotometer. Total Fe and Mn concentrations in irrigation water were compared with the standards of the Philippines and other countries (Department of Agriculture 2007, Ayers and Westcot 1994, USEPA and USAID 1992, ANZECC and ARMCANZ 2000).

Farm Inputs and Activities that affect Nutrient Levels

A questionnaire was used to survey the levels of puddling operations and the amounts of fertilizers and pesticides applied by farmers. Pearson correlation analysis determined the association of farm input levels with the nutrient levels.

Identifying the Source of Nutrients

Source of nutrients was determined only in sites with toxic or with the highest level of a particular nutrient among the sites. Sources of nutrients were determined based on the results of analysis of soil properties, land uses, irrigation water, and farm inputs that affect the nutrient levels. The analytical framework for determining the sources is shown in Figure 2.

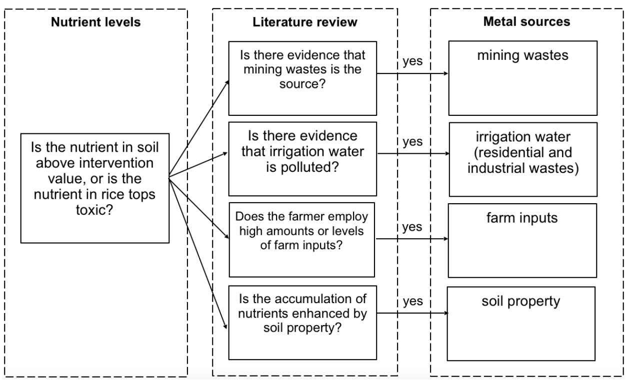

Figure 2. Analytical framework for determining the sources of nutrients in study sites.

RESULTS AND DISCUSSION

Total concentrations of K, Ca, S, Fe and Mn were included in this report. Total concentrations of other elements, such as chromium, nickel, copper, cobalt, zinc, arsenic, molybdenum, cadmium, mercury, lead, strontium, rubidium, titanium and zirconium, were also analyzed. For convenience of interpretation, these total element concentrations were not included in this report.

Total Nutrient Levels in Soils

Total soil nutrient levels were compared to the mean values for Philippine paddy soils (Table 2). The highest total K level among the paddy soils studied was found in Kabacan. The second highest level was observed in San Miguel (Iloilo), while the third highest was found in Cabatuan.

The Polangui soil had the highest total Ca concentration. The La Paz a and Villasis soils had the second and third highest concentrations, respectively.

The highest total Fe level was found in Zambales. The second highest level was observed in Sta. Rosa City a, while the third highest was found in San Leonardo a.

The highest total Mn level was found in Sta. Rosa City a. The second highest level was observed in Jaen, while the third highest was found in Zambales.

Nutrient Levels in Rice Tops

Nutrient levels in rice were also compared to values considered as normal, toxic, and deficient in mature leaf tissues for various species and for rice tops (Table 3). Rice in La Paz a had the highest K level. Rice in Sara and Sta. Rosa City b had the second and third highest levels, respectively. Moreover, rice in Tacurong City, Sipalay City, Muñoz City a, and Milaor also had K concentrations above the toxic level.

For all sites, Fe levels exceeded the toxic level for rice tops (Dobermann and Fairhurst 2000) and for various species (Marschner 2011). The highest Fe concentrations were found in rice tops collected from Muñoz City a, San Miguel (Bulacan), and San Leonardo a.

Manganese levels in Zaragoza, San Manuel, and San Miguel (Bulacan) sites exceeded the toxic level for rice tops (Dobermann and Fairhurst 2000).

Based on nutrient concentrations, no toxic levels of Ca, or deficient levels of K, Ca, Fe and Mn in rice were found.

Soil Properties that affect Nutrient Levels

Significantly positive correlations were found for soil pH vs. plant K (Tables 4 and 5). A significantly negative correlation was observed for soil pH vs. plant Mn.

Significantly negative correlations were found for sand vs. plant Ca, clay vs. plant K, sand vs. total soil Fe, and clay vs. total soil Ca. Significantly positive correlations were observed for clay vs. total soil Fe, and sand and silt vs. total soil Ca.

Significantly negative correlations were found for OM vs. plant Mn. Significantly positive correlations were observed for available P vs. total soil Ca, plant K, and total soil Mn.

Influence of Soil pH on Nutrient Concentration

A significantly positive correlation for soil pH vs. plant K was observed because slightly acidic paddy soils allow less amounts of K for leaching and more for plant uptake. Chen and Barber (1990) showed that nutrient-uptake model accurately predicted the effect of soil pH on K uptake ($Y = 67 + 0.94X$, $R^2 = 0.99$). Also, strongly acidic paddy soils, with more solution concentration of H^+, probably decrease the function of root plasma membrane, and promote loss of K or inhibition of the nutrient's uptake (Alam et al. 1999).

A significantly negative correlation for soil pH vs. plant Mn was found because slightly acidic paddy soils adsorb and precipitate high amounts of Mn, limiting the nutrient's availability for uptake. Mitsios et al. (2005) also found a significantly negative correlation between soil pH and DTPA-extractable Mn.

Influence of Soil Texture on Nutrient Concentration. A significantly negative correlation for sand vs. plant Ca was found because paddy soils with high sand content (loam) probably allow leaching of Ca, while those with low sand content (clay) adsorb and accumulate this nutrient. This result agrees with the findings of Kahlon et al. (2013).

A significantly negative correlation for clay vs. plant K was observed because paddy soils with high clay contents adsorb high amounts of K, resulting in the reduced uptake of the nutrient.

A significantly positive correlation for clay vs. total soil Fe, and a significantly negative correlation for sand vs. total soil Fe were found because the stable forms of soil Fe occur as clay-sized particles. Soil Fe occur as free oxides (Zhang et al. 2012) or as coatings of clay minerals (Rengasamy and Oades 1977) in the clay-sized soil fractions. Furthermore, Fe in the soils studied possibly occur as structural component of clay minerals. Clay minerals in the soils studied are primarily smectites and vermiculite (Miura et al. 1995).

A significantly positive correlation for sand and silt vs. total soil Ca, and a significantly negative correlation for clay vs. total soil Ca were observed because the stable forms of soil Ca occur in sand and silt-sized particles. Calcium is a structural component of minerals in the sand and silt-sized soil fractions. Pyroxene, amphibole and plagioclase are minerals in the fine sand fractions of the soils

TABLE 2. Total nutrient levels in soils as compared to background values and guidelines of countries.

Nutrient Guidelines and Values/Site	K	Ca	Fe	Mn
	cmol(+)kg^{-1}	cmol(+)kg^{-1}	%	%
Mean for Philippine paddy soils (N=34) (Miura et al., 1995)	0.76% K_2O =8.07 K	2.58% CaO =92.01 Ca	9.40 Fe_2O_3 =3.29 Fe	0.17 MnO_2 =0.11 Mn
Mean for Philippine paddy soils (N=54) (Domingo and Kyuma, 1983)	0.99% K_2O =10.51 K	2.83% CaO =100.93 Ca	7.73 Fe_2O_3 =2.70 Fe	0.23 MnO_2 =0.15 Mn
Cagayan Valley				
Solana, Cagayan	15.10	44.16	4.10	0.09
Cabatuan, Isabela	21.71	67.36	4.49	0.07
Echague, Isabela	2.88	7.37	1.30	0.01
Central Luzon				
Muñoz City, Nueva Ecija (N.E.) a	8.15	64.56	3.09	0.05
Muñoz City, N.E. b	5.33	50.99	3.71	0.06
Talavera, N.E.	7.34	59.84	4.66	0.06
Llanera, N.E.	1.59	21.43	1.95	0.05
Zaragoza, N.E.	8.79	63.65	2.31	0.04
San Leonardo, N.E. a	7.70	38.49	5.22	0.07
San Leonardo, N.E. b	7.95	44.72	4.97	0.08
Jaen, N.E.	10.77	56.73	4.34	0.11
San Miguel, Bulacan	11.09	29.67	4.28	0.07
San Manuel, Tarlac	10.47	75.11	3.00	0.09
La Paz, Tarlac a	7.88	89.55	2.14	0.05
La Paz, Tarlac b	9.27	55.99	3.76	0.07
Villasis, Pangasinan	17.77	85.30	4.04	0.10
Sta. Cruz, Zambales	1.24	23.44	7.34	0.11
Laguna				
Sta. Rosa City a	8.84	30.91	5.29	0.13
Sta. Rosa City b	9.89	41.74	5.12	0.09
Bay	6.55	37.69	4.01	0.06
Sta. Cruz	11.33	61.02	3.02	0.04
Bicol				
Milaor, Camarines Sur	8.13	38.07	4.54	0.08
Canaman, Camarines Sur	5.66	53.48	2.67	0.03
Minalabac, Camarines Sur	8.18	36.24	3.60	0.03
Polangui, Albay	15.62	133.72	3.38	0.06
Casiguran, Sorsogon	13.81	18.71	1.31	0.01
Visayas				
Sara, Iloilo	20.19	58.51	3.28	0.07
San Miguel, Iloilo	26.96	45.02	3.21	0.03
Sipalay City, Negros Occidental	21.23	40.64	2.08	0.04
Mindanao				
Cotabato City, Maguindanao	16.71	54.12	4.25	0.05
Kabacan, North Cotabato	38.34	34.98	1.91	0.02
Tacurong City, Sultan Kudarat	15.89	84.68	1.75	0.05

TABLE 3. Nutrient levels in plant tissues as compared to values considered as normal, toxic and deficient in mature leaf tissues for various species and for rice tops.

Nutrient Guidelines and Values/Site	K	Ca	Fe	Mn
	%	%	mg kg^{-1}	mg kg^{-1}
Normal level for various species[a]	-	-	-	30-300
Toxic level for various species[a]	-	-	-	400-1000
Toxic levels for various species[b]	-	-	>500	>200
Toxic levels for rice tops[c]	-	-	500-1000	300-1000
Toxic levels for rice tops[d]	>3.00	>0.70	>300	>800
Deficient levels for various species[b]	-	-	50-150	10-20
Deficient levels for rice tops[d]	<1.20	<0.15	<70	<20
Cagayan Valley				
Solana, Cagayan	2.21	0.37	1656.49	581.63
Cabatuan, Isabela	2.65	0.57	1805.95	746.54
Echague, Isabela	2.01	0.37	972.97	295.37
Central Luzon				
Muñoz City, Nueva Ecija (N.E.) a	3.17	0.48	4109.12	558.33
Muñoz City, N.E. b	2.53	0.55	1665.24	603.22
Talavera, N.E.	2.34	0.42	1263.22	460.59
Llanera, N.E.	2.06	0.43	819.81	681.39
Zaragoza, N.E.	2.12	0.47	927.01	1095.79
San Leonardo, N.E. a	1.83	0.64	1885.62	628.42
San Leonardo, N.E. b	1.79	0.61	1672.57	561.10
San Miguel, Bulacan	1.68	0.55	3381.24	803.43
San Manuel, Tarlac	2.71	0.50	1063.64	1002.84
Villasis, Pangasinan	8.51	0.69	1694.53	<55
La Paz, Tarlac a	2.45	0.35	482.55	512.22
La Paz, Tarlac b	4.33	0.40	1171.64	600.89
Sta. Cruz, Zambales	1.88	0.39	765.65	138.91
Laguna				
Sta. Rosa City b	3.86	0.47	680.88	312.25
Bay	2.98	0.40	796.37	280.98
Sta. Cruz	2.51	0.36	711.82	161.85
Bicol				
Milaor, Camarines Sur	3.12	0.42	1648.76	231.20
Canaman, Camarines Sur	2.81	0.41	964.71	217.45
Minalabac, Camarines Sur	2.34	0.54	887.90	789.87
Polangui, Albay	2.42	0.43	1703.13	195.70
Casiguran, Sorsogon	1.62	0.50	1358.57	202.83
Visayas				
Sara, Iloilo	4.12	0.64	1630.47	729.92
Sipalay City, Negros Occidental	3.22	0.54	484.42	519.63
Mindanao				
Cotabato City, Maguindanao	2.73	0.45	594.25	240.42
Kabacan, North Cotabato	2.34	0.39	581.96	210.12
Tacurong City, Sultan Kudarat	3.25	0.42	488.03	378.49

[a]Kabata-Pendias and Pendias (2001), [b]Marschner (2012), [c]Kitagishi and Yamane (1981), [d]Dobermann and Fairhurst (2000)

TABLE 4. Pearson correlation coefficients of soil properties vs. soil nutrient levels.

Soil Properties	N	K	Ca	Fe	Mn
soil pH	85	0.04	0.20	0.57**	0.58**
sand, %	85	0.10	0.43**	-0.44**	-0.10
silt, %	85	0.06	0.25*	-0.05	0.02
clay, %	85	-0.11	-0.47**	0.36**	0.06
organic matter, %	85	-0.11	-0.43**	0.15	-0.19
available P, mg kg^{-1}	85	0.18	0.37**	0.20	0.31**

TABLE 5. Pearson correlation coefficients of soil properties vs. plant nutrient levels.

Soil Properties	N	K	Ca	S	Fe	Mn
soil pH	80	0.28*	-0.12	0.08	-0.17	-0.30**
sand, %	80	0.12	-0.22*	-0.10	-0.17	0.11
silt, %	80	0.21	0.02	-0.08	0.03	-0.06
clay, %	80	-0.22*	0.154	0.13	0.11	-0.05
organic matter, %	80	-0.22	-0.03	0.18	-0.14	-0.30**
available P, mg kg^{-1}	80	0.46**	0.05	0.19	-0.07	-0.41**

*correlated at 5% significance level; **correlated at 1% significance level

TABLE 6. Comparison of nutrient levels in soils of different land uses[1].

Land Uses	N	K	Ca	Fe	Mn
Deep-well and rainfed areas	9	a	a	b	a
Dam and river-irrigated areas	60	a	b	b	b
Mine area-adjacent sites	6	a	c	ab	ab
Urban area-adjacent sites	4	a	bc	a	a
Highway-adjacent sites	6	a	bc	b	a

TABLE 7. Comparison of nutrient levels in plant tissues of different land uses[1].

Land Uses	N	K	Ca	S	Fe	Mn
Deep-well and rainfed areas	7	ab	a	ab	a	a
Dam and river-irrigated areas	57	c	a	b	a	a
Mine area-adjacent sites	6	bc	a	ab	a	a
Urban area-adjacent sites	4	a	a	ab	a	a
Highway-adjacent sites	6	ab	a	a	a	a

[1]Kruskal-Wallis and Mann-Whitney U tests; same letters are not different at 5% significance level; different letters are different at 5% significance level

TABLE 8. Comparison of nutrient levels in soils collected close, middle and far from irrigation entrance (IE)[1].

Distance from IE	N	K	Ca	Fe	Mn
close (0-2 m)	17	ab	a	a	a
middle (40-42 m)	17	b	a	a	a
far (80-82 m)	17	a	a	a	a

TABLE 9. Comparison of nutrient levels in plant tissues collected close, middle and far from irrigation entrance (IE)[1].

Distance from IE	N	K	Ca	Fe	Mn
close (0-2 m)	16	a	a	b	a
middle (40-42 m)	16	a	a	a	a
far (80-82 m)	16	a	a	ab	a

[1]pairwise comparison using Sign test; same letters are not different at 5% significance level; different letters are different at 5% significance level

studied (Miura et al. 1995). Pyroxenes are present in soils as diopside or hedenbergite. Amphiboles occur as tremolite or hornblende, while plagioclase can be present as anorthite (van der Weijden 2007).

Influence of Organic Matter on Nutrient Concentration. A significantly negative correlation for OM vs. plant Mn was observed because paddy soils with high OM contents chelate high amounts of Mn and enhance the nutrient's leaching. Furthermore, as OM-bound Mn reaches the rhizosphere, the oxidized soil condition possibly mineralizes the OM, and transforms Mn into less available forms (oxides). The combined leaching and adsorption or precipitation of Mn reduced its plant uptake.

A significantly negative correlation for OM vs. total soil Ca was found because of the strong affinity between the OM and clay fractions of the paddy soils. There was a highly significant correlation (0.73**) between OM and clay contents of the 85 soil samples analyzed in this study. Soils with low OM contents have high levels of nutrients that are concentrated in sand-sized particles.

Influence of Available P on Nutrient Concentration. A significantly positive correlation for available P vs. total soil Ca was found because Ca and P concentrations in the solutions of paddy soils possibly increase during submergence (Ponnamperuma 1972), and enhance the precipitation of Ca phosphates. If cation concentrations are too high to be adsorbed on the soil, the available P or phosphate can precipitate with cations to form phosphate compounds. Available Ca and P are required in the formation of Ca phosphate in soils (Tunesi et al. 1999). Patrick and Mahapatra (1968) also reported that an increase in pH due to submergence increases the amount of Ca phosphates. Calcium phosphates are sparingly soluble, and this insolubility possibly prevents the leaching of Ca from the paddy soils.

A significantly positive correlation for available P vs. plant K was found because high P availability during submergence of paddy soils is probably coupled with high amounts of K available for plant uptake. Nutrients, such as P and K, become more available in paddy soils upon submergence (De Datta 1981) due to their release from adsorption sites (Ponnamperuma 1972) or their presence in irrigation water. Husnain et al. (2010) reported that irrigation water contains as much as 7.2 mg L^{-1} K and contributed 93% of the total K input.

A significantly negative correlation for available P vs. plant Mn was found. An increase in nutrient availability during submergence of paddy soils possibly enhances the formation of Fe and Mn plaques on rice roots. These plaques limit the uptake of Mn. Tavakkoli et al. (2011) also observed that silicon application can decrease Mn contents of rice. Application of P (Haldar and Mandal 1981), lime, nitrogen (N)-PK + lime, NPK, K (Alam et al. 2003), Ca or Mg (Ramani and Kannan 1974) can also decrease the Mn concentration of rice.

Land Uses that affect Nutrient Levels

Urban area-adjacent sites had higher total soil Fe and Mn concentrations, and higher plant K concentrations than the other sites (Tables 6 and 7). Deep-well and rainfed sites were enriched with soil Ca and Mn. Highway-adjacent sites had higher total soil Mn and plant S concentrations than the other sites.

Urban area-adjacent soils had higher total Fe and Mn concentrations because these soils are probably being deposited with these nutrients from urban sources. Iron and Mn are possibly carried by the contaminated run-off water, atmospheric particles, and solid wastes that are mixed with the soils. Edori and Edori (2012) suggested that the increased Fe concentrations in soils of machine shops can be due to dumping of Fe scraps, unused body parts of vehicles, tin can, solvents, hydraulic fluid and spent lubricants. Lytle et al. (1995) found that soil Mn concentrations in high traffic areas were up to 100 fold higher than that recorded for lead.

The Sta. Rosa City b site was also irrigated with water containing urban wastes. Total Fe level was higher in the soil collected near than that collected far from the irrigation entrance (52.0% vs. 50.3 %). This higher level indicate that the polluted irrigation water enriched the soil with Fe. Aydinalp et al. (2010) also found that agricultural soils in a highly populated and industrialized area were enriched with Fe due to long-term irrigation of water from a polluted river.

Urban area-adjacent sites had higher plant K concentrations because the soils in these sites have neutral pH levels and clay textures. These soil properties contribute to low amounts of K leached, and increased concentration of the nutrient in plants.

Deep-well and rainfed paddy soils had higher total Ca levels because such soils have high sand and silt contents. Significantly positive correlations for soil Ca levels vs. sand and silt contents were observed (Table 4). Also, Ca leaching is limited in these soils due to less irrigation intensity compared to other paddy soils.

Highway-adjacent sites had higher total soil Mn levels because they were probably deposited with Mn oxides from vehicle exhausts. Lytle et al. (1995) also found that soil Mn levels in high traffic areas were up to 100 fold higher than those recorded for lead.

Highway-adjacent sites had higher plant S concentrations because rice in these sites probably absorb sulfur dioxide from vehicle emissions. Sulfur was detected only in rice collected at 5 and 45 m from the nearby highway in the Milaor site. Huseyinova et al. (2009) also noted that plants near highways (1417 – 3387 mg kg^{-1}) were higher in S concentrations than plants far from highways (71 – 1111 mg kg^{-1}).

Deep-well and rainfed sites had higher total soil Mn levels because these sites are probably being deposited with Mn oxides from exhausts of farm tractors and threshers. These sites are employed with 9, 10, 14 and 112 mechanized farm operations per year.

Irrigation Water as Source of Nutrients

Nutrient Levels for Area Close vs. Middle vs. Far from Irrigation Entrance. Generally, soil samples far from the irrigation entrance had higher total K concentrations than those in the middle (Table 8). Rice plants collected in the middle had higher Fe contents than those collected close to the irrigation entrance (Table 9).

Silt particles were more abundant in the soils near the irrigation canal than those in the middle (Table 10). This abundance suggests that irrigation water in such sites carries upstream particles and deposits them in areas close to the irrigation entrance. Nguyen et al. (2006) also noted that soil texture in a paddy field was altered due to runoff and sediments from the uplands.

Spatial variability of total K concentrations in paddy soils and Fe concentrations in rice is probably controlled by soil texture. Silt particles were less abundant in soils in the middle than those near the irrigation entrance (Table 10). Hence, K leaching is probably higher in the middle than near the irrigation canal. Also, silt particles possibly allow the leaching of Fe, limiting the nutrient's availability for uptake. Fullen and Brandsma (1995) found that mean silt content decreased from 27.8% to 19.6% after soil erosion, and such change in soil texture was accompanied by a decrease in K and Fe.

TABLE 10. Comparison of soil properties in soils collected close, middle and far from irrigation entrance (IE)[1].

Distance from IE	N	Soil pH	OM %	Sand %	Silt %	Clay %	Avail P mg kg^{-1}
close (0-2 m)	17	a	a	a	a	a	a
middle (40-42 m)	17	a	a	a	b	a	a
far (80-82 m)	17	a	a	a	ab	a	a

[1]pairwise comparison using Sign test; same letters are not different at 5% significance level; different letters are different at 5% significance level

Iron and Mn Levels of Irrigation Water from Two Mine-Adjacent Areas. Iron concentration in irrigation water of the Zambales site was higher than the Philippine, and Australia and New Zealand standards. Iron concentration in irrigation water of the Sipalay City site was ten times higher than the Philippine standard, and two times higher than the FAO and Saudi Arabia standards (Table 11). Manganese concentrations in the irrigation water of the two sites did not exceed the standards.

TABLE 11. Comparison of mine-contaminated irrigation water (mg L^{-1}) from study sites with irrigation water standards.

Country/Organization	Fe	Mn
Philippines (Department of Agriculture, 2007)	1.00	0.200
FAO (Ayers and Westcot, 1994)	5.00	0.200
Saudi Arabia (USEPA and USAID, 1992)	5.00	0.200
Australia and New Zealand[1]	0.20	0.200
Sta. Cruz, Zambales	1.14	0.024
Sipalay City, Negros Occidental	10.18	0.075

[1]Australian and New Zealand Environment and Conservation Council, and Agriculture and Resource Management Council of Australia and New Zealand (2000)

The Sipalay City site is adjacent to a Cu mining area. The elevated Fe concentration in irrigation water of the site can be due to the seepage from the mine tailing pond or old open pit. The river, as source of irrigation water, flows along the length of the tailing pond. Moreover, the Zambales site is adjacent to a Ni mining area. The elevated Fe concentration in irrigation water of the site can also be due to seepage from the waste rock piles. Rainwater in these two sites probably collects nutrients within the basin and carry them into the river. Copper porphyry is present in the Sipalay City's Maricalum Mining Corporation (Vigar et al. 2011). Furthermore, Jopony and Tongkul (2009) found that seepage water in the pit area of an abandoned Cu mine had elevated Fe concentration. Johnson et al. (2000) also reported that the oxidation of sulfide minerals in an abandoned Ni–Cu mine released up to 9.8 g L^{-1} of Fe.

High Fe concentration in irrigation water of the Zambales and Sipalay City sites can result in elevated concentration of this nutrient in soils and rice. Abbas et al. (2007) found that irrigating paddy soils with industrial effluents carrying micronutrients will increase the concentrations of the same micronutrients.

Farm Management Practices as Source of Nutrients

Annual one-hectare input levels in study sites are presented in Table 12. Results of correlations for the amounts or levels of farm inputs vs. total soil nutrient levels are presented in Table 13. Amounts of farm inputs were not significantly correlated with total soil nutrient levels.

TABLE 12. Annual one-hectare farm input levels in study sites.

Site	Puddling operations	Granular fertilizers	Foliar fertilizers	Nitrogen in granular fertilizers
	no.	bag	load	kg
Cagayan Valley				
Solana, Cagayan	8	11.1	0.0	198.4
Cabatuan, Isabela	12	12.5	0.0	242.5
Echague, Isabela	8	10.8	12.3	102.4
Central Luzon				
Muñoz City, Nueva Ecija (N.E.) a	8	24.2	0.0	236.3
Muñoz City, N.E. b	9	17.0	0.0	165.5
Talavera, N.E.	12	27.0	0.0	345.0
Llanera, N.E.	8	15.0	24.0	151.5
Zaragoza, N.E.	8	12.3	2.0	174.8
San Leonardo, N.E. a	13	38.0	44.0	253.0
San Leonardo, N.E. b	12	13.8	0.0	264.0
Jaen, N.E.	0	1,081.7	300.0	31.9
San Miguel, Bulacan	8	15.2	20.0	145.5
San Manuel, Tarlac	8	6.0	20.0	73.0
La Paz, Tarlac a	9	12.5	0.0	215.4
La Paz, Tarlac b	8	22.0	0.0	385.0
Villasis, Pangasinan	111	26.0	5.0	399.0
Sta. Cruz, Zambales	12	17.1	6.4	228.8
Laguna				
Sta. Rosa City a	0	0.0	0.0	0.0
Sta. Rosa City b	9	21.4	0.0	482.0
Bay	8	17.1	0.0	297.0
Sta. Cruz	6	7.8	0.0	147.0
Bicol				
Milaor, Camarines Sur	14	1.7	5.2	37.4
Canaman, Camarines Sur	13	2.0	0.0	29.5
Minalabac, Camarines Sur	10	10.0	16.9	134.4
Polangui, Albay	14	13.3	0.0	258.4
Casiguran, Sorsogon	13	4.3	0.0	52.0
Visayas				
Sara, Iloilo	10	12.0	0.0	177.0
San Miguel, Iloilo	12	92.0	80	146.0
Sipalay City, Negros Occidental	15	22.7	72	387.5
Mindanao				
Cotabato City, Maguindanao	8	1.5	9.2	34.7
Kabacan, North Cotabato	14	13.3	16.7	184.3
Tacurong City, Sultan Kudarat	10	16.0	48.0	180.0

TABLE 12. *continued*

Site	Phosphate in granular fertilizers	Potash in granular fertilizers	Foliar pesticides	Solid pesticides
	kg	kg	load	kg
Cagayan Valley				
Solana, Cagayan	23.4	23.4	50.0	6.3
Cabatuan, Isabela	17.5	17.5	40.6	0.0
Echague, Isabela	73.9	43.1	33.8	1.5
Central Luzon				
Muñoz City, Nueva Ecija (N.E.) a	133.4	116.7	10.0	4.0
Muñoz City, N.E. b	98.0	98.0	45.0	2.0
Talavera, N.E.	136.0	86.0	27.0	2.0
Llanera, N.E.	84.0	84.0	81.0	2.0
Zaragoza, N.E.	66.01	9.3	50.0	3.3
San Leonardo, N.E. a	79.0	49.0	84.0	6.0
San Leonardo, N.E. b	21.0	21.0	52.2	2.5
Jaen, N.E.	2.7	2.7	300.0	0.0
San Miguel, Bulacan	106.0	56.0	98.3	0.0
San Manuel, Tarlac	28.0	28.0	72.0	0.0
La Paz, Tarlac a	11.7	11.7	0.0	0.0
La Paz, Tarlac b	28.0	28.0	137.5	34.0
Villasis, Pangasinan	84.0	84.0	77.0	51.0
Sta. Cruz, Zambales	40.0	40.0	15.0	0.0
Laguna				
Sta. Rosa City a	0.0	0.0	0.0	0.0
Sta. Rosa City b	0.0	0.0	128.6	81.4
Bay	14.0	14.0	114.3	0.0
Sta. Cruz	13.2	13.2	23.8	1.2
Bicol				
Milaor, Camarines Sur	0.0	0.0	197.3	3.7
Canaman, Camarines Sur	7.0	7.0	40.0	21.0
Minalabac, Camarines Sur	21.6	47.7	66.2	1.5
Polangui, Albay	18.6	18.6	53.3	0.0
Casiguran, Sorsogon	20.0	20.0	30.0	1.4
Visayas				
Sara, Iloilo	42.0	42.0	80.8	0.0
San Miguel, Iloilo	56.0	56.0	285.9	0.4
Sipalay City, Negros Occidental	70.4	70.4	157.4	1.7
Mindanao				
Cotabato City, Maguindanao	0.0	0.0	61.5	0.0
Kabacan, North Cotabato	0.0	37.7	241.7	2.2
Tacurong City, Sultan Kudarat	0.0	0.0	147.9	0.0

TABLE 13. Pearson correlation coefficients of nutrient levels in soils vs. amount or levels of farm inputs.

Farm Inputs[1]	N	K	Ca	Fe	Mn
puddling operations, no.	32	0.19	0.26	0.04	0.15
granular fertilizers, bag	32	-0.00	0.04	0.11	0.31
foliar fertilizers, sprayer load	32	0.11	-0.02	0.01	0.20
granular fertilizers, bag	32	0.08	0.28	0.14	0.09
phosphate in granular fertilizers, kg.	32	-0.24	-0.10	-0.04	-0.15
potash in granular fertilizers, kg.	32	-0.07	-0.10	-0.09	-0.18
foliar pesticides, sprayer load	32	0.26	0.01	-0.02	0.16
solid pesticides, kg.[1]	32	-0.04	0.14	0.11	0.21

Results of correlation for the amounts of farm inputs vs. plant nutrient levels are presented in Table 14. Significantly positive correlations were found for the amounts of phosphate and potash applied vs. plant Fe levels. Loads of foliar fertilizers applied had a significantly negative correlation with plant Fe levels. Loads of foliar pesticides applied had a significantly positive correlation with plant S levels, and a significantly negative correlation with plant Fe levels. Amounts of solid pesticides applied had a significantly positive correlation with plant K levels.

TABLE 14. Pearson correlation coefficients of nutrient levels in plant tissues vs. amount or levels of farm inputs.

Farm Inputs[1]	N	K	Ca	S	Fe	Mn
puddling operations, no.	30	0.21	-0.13	0.01	-0.04	0.05
granular fertilizers, bag	30	0.17	0.14	-0.20	0.16	-0.03
foliar fertilizers, sprayer load	30	-0.01	0.13	-0.03	-0.27	0.00
granular fertilizers, bag	30	0.31	0.06	-0.25	-0.08	0.00
phosphate in granular fertilizers, kg.	30	-0.00	0.21	-0.19	0.50**	0.32
potash in granular fertilizers, kg.	30	0.12	0.23	-0.21	0.41*	0.19
foliar pesticides, sprayer load	30	0.12	-0.14	0.40*	-0.41*	-0.29
solid pesticides, kg.[1]	30	0.51**	0.08	-0.04	-0.13	-0.26

*correlated at 5% significance level; ** correlated at 1% significance level; [1]solid pesticides=pellet, powder or granular pesticides

Fertilizers. The significantly positive correlation for the amounts of phosphate and potash applied vs. plant Fe levels can be due to the enhanced soil availability and plant uptake of Fe owing to the strong soil acidity from continuous high-yield cropping. The Central Luzon sites are the ones applied with the highest amounts of P and K fertilizers (Table 12). These sites also produce the highest rice yields, and their rice plants had the highest Fe concentrations (Table 3). Significantly negative correlations were found for soil pH vs. amounts of phosphate and potash applied. These indicate that higher amounts of phosphate and potash applied are associated to strongly acidic soil pH. Through their long-term field experiment, Belay et al. (2002) also found that basic cation contents and soil pH levels declined more in NPK treatments with significantly higher yield than single fertilizer treatments. Rice roots release H^+ to balance cation-anion intake or maintain electrical neutrality across the root-soil interface. Modern irrigated rice varieties remove 3.0 kg K, 4.0 kg Ca, and 3.5 kg magnesium in a ton of grain yield (Dobermann and Fairhurst 2000).

Pesticides. A significantly positive correlation for the amounts of foliar pesticides vs. plant S levels was found. Sulfur, a component of foliar pesticides' active ingredients (AI) used in the sites studied, can be absorbed by the leaves of rice. Sulfur-containing AIs were also found in plants and plant products applied with foliar pesticides containing the same AIs. Such AIs are chlorpyrifos (Lu 2011), thiamethoxam (Liu et al. 2009) and mancozeb (Patsakos et al. 1992).

A significantly negative correlation for the amounts of foliar pesticides vs. plant Fe levels was observed because pesticide applications possibly stress the rice plants, and cause the production of phenols and decrease the Fe concentration in rice. Saladin and Clement (2005), and Siddiqui and Ahmed (2006) showed the depressing effects (reduced growth and dry matter accumulation) of foliar pesticides by the increased production of phenolic compounds in plants. Frossard et al. (2000) mentioned that phenolic compounds inhibit Fe absorption. Phenols also reduce and release ferritin Fe, a multi-subunit protein in higher plants (Boyer et al. 1990).

A significantly positive correlation for the amounts of solid pesticides vs. plant K concentrations was found. Solid pesticides, as they effectively control pests, can increase the growth and biomass accumulation of rice. Carbofuran and metaldehyde are the most common solid pesticides used in the study sites. Accordingly, an increase in growth and biomass of rice in the sites studied can be due to the effective control of insect pests by carbofuran granules and the effective control of golden apple snails by metaldehyde pellets. The increase in growth and biomass probably leads to the efficient uptake of K. Carbofuran can effectively control certain rice insect pests (Ukwungwu 1987), and can increase rice growth and yield (Xiao et al. 1995). Furthermore, snail infestation damages young rice seedlings, and causes poor crop stand and reduced yield (Cagauan and Joshi 2002). Commercial molluscicide formulations, including metaldehyde pellets, resulted in up to 69% golden apple snail mortality during the dry season and 90% during the wet season (Dela Cruz and Joshi 2001).

CONCLUSION

Potassium concentrations of rice plants in La Paz (Tarlac) and Sta. Rosa City (Laguna) exceed the toxic concentration of 3%. These K concentrations can be due to the increase of the nutrient's availability owing to the neutral soil pH levels in the two sites. The Polangui (Albay), La Paz, and Villasis (Pangasinan) soils contain the highest total Ca levels. These total Ca levels can be ascribed to the occurrence of Ca in sand and silt-sized soil fractions.

Total Fe and Mn concentrations are very high in the Sta. Cruz (Zambales) soil due to the input of mine wastes. The San Leonardo (Nueva Ecija) soil's high total Fe concentration is probably due to the occurrence of Fe in clay-sized soil fractions. Iron and Mn levels of rice in most Central Luzon sites exceed the toxic concentrations of 1000 mg kg^{-1} for Fe and 300 mg kg^{-1} for Mn. These concentrations can be due to the high total soil Fe and Mn levels, and their increased plant uptake due to periodic soil submergence and increased soil acidity from continuous cropping. The study implies that K, Ca, Fe and Mn are enriched in rice areas due to soil properties and farm practices.

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CONFLICT OF INTEREST

There are no conflicts of interest arising from this study.

CONTRIBUTIONS OF INDIVIDUAL AUTHORS

Jehru C. Magahud, Rodrigo B. Badayos, Pearl B. Sanchez, and Pompe C. Sta. Cruz conceptualized and designed the study, analyzed and interpreted the data, and revised the manuscript for important intellectual content. Jehru C. Magahud collected the data and drafted the manuscript.

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