

IMPACT OF CITRUS AGRICULTURE ON THE QUALITY OF WATER RESOURCE IN A SMALL STEEP ISLAND, SETO INLAND SEA, JAPAN

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ABSTRACT: Groundwater is an extremely valuable resource thus its pollution a call for concern. Amongst the major threats, agricultural leachates often contribute significantly to groundwater pollution. This paper reports an investigation on the effects of citrus agriculture on the groundwater quality aspects of Osakishimajima island. Fifty-one groundwater samples were collected and analyzed for major ions and nutrients. The Gibbs diagram revealed that the overall hydrogeochemical environment of the study area is controlled by rock-water interaction. Two main hydrochemical facies (CaMg-HCO₃ and CaMg-SO₄) were identified. 48 groundwater samples showed PO₄³⁻-P content (0.06 - 3.2 mg/L), which exceeds WHO's recommended maximum permissible limit. High contamination of PO₄³⁻-P was noticed at depth zones between 3 to 6 m while the highest concentrations were found at depths of 5 m. NO₃⁻-N contamination of groundwater was also observed to decrease with depth. High levels of PO₄³⁻-P in groundwater may be as a result of heavy fertilizer application by farmers.

Keywords: *Citrus agriculture, Groundwater quality, Contamination, Osakishimajima, Seto Inland Sea*

1. INTRODUCTION

Groundwater is the major source of water for domestic, agricultural, and industrial purposes in many countries. Water and its quality are critical environmental determinants in any economy, and it is difficult to imagine productive human activities like agriculture or livestock without a well-functioning water supply [1]. Nutrients are required for maximum growth and production by all plants in the environment, however, nutrients are fast becoming the leading cause of contamination in groundwater, lakes, springs, rivers, and coastal waters. In Asian countries, exploitation of groundwater sources has increased greatly over the past few decades where it serves multiple functions [2]. Contaminants in groundwater such as nitrate and phosphate sources can be associated with rapid population growth, agricultural run-off, industries, and domestic waste. Findings from [3] show that agricultural run-off is the leading source of water contamination and the most difficult to eliminate due to its spatial characteristics.

Several natural and anthropogenic factors controlling groundwater quality have been documented [4] which include natural (rainfall,

topographic relief, mineral dissolution, mineral solubility, ion exchange, oxidation, reduction, residence time, geological structure, and mineralogy of the water and aquifers) and anthropogenic (poor sanitary conditions, application of fertilizers and pesticides for higher crop yields). Extensive studies on groundwater quality in Japan have been carried out by various scholars [5-7]. However little attention has been paid to groundwater quality in the small islands of the Seto inland sea.

Osakishimajima is a small island in the Seto inland with an aging population [8] and experiencing rapid depopulation. Citrus agriculture is the main livelihood activity of the people practiced throughout the year and heavily relying on groundwater for irrigation. Also, the drinking water source in Osakishimajima is from Kure city which is over 100 km² away. In the advent of a natural disaster such as the devastating flood which occurred in June 2018 and contaminated the pipelines with mud and sediments, the villagers were without access to safe drinking water for over ten days, hence a pressing need to assess the groundwater for quality to provide future alternative reliable and quality water sources.

Quality study was carried out in this area to assess the possible effects of citrus agriculture on groundwater resources to provide relevant baseline information. The assessment of water quality in Osakishimajima is an important tool for sustainable development of the community which will help provide critical information for water resources management.

2. METHODS

2.1 Study Area

Osakishimajima is a small island located along the stretches of the Seto inland sea in Hiroshima prefecture, western Japan (Fig. 1) about 10km off the coast of Kure city. Its surface area is about 43 km² with geographic coordinates 34°10'24"N and 132°50'3"E. It has a dwindling aging population of about 3,000 people [8]. This modest-sized island is large enough to be lined with steep mountains and narrow valleys yet small enough to be tucked away amid a cluster of other islands.

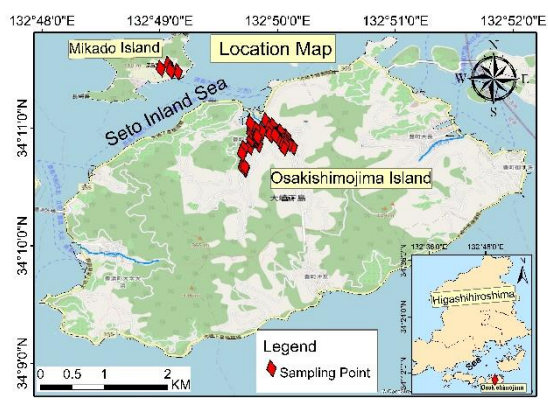


Fig.1 Location of Osakishimajima in Hiroshima Prefecture showing sampling points

2.1.1 Geology

Osakishimajima is one of the Archipelago in the middle of the Seto inland sea. It is made up of late Cretaceous Hiroshima granitic rocks which widely intrude into the remote islands of the Seto inland sea. It's also comprised of late Cretaceous rhyolite-dacitic rocks outcrop and a mélange matrix of Early to Middle Jurassic accretionary complexes associated with Triassic chert block and Carboniferous to Permian limestone which underlay the island [9].

2.1.2 Land-uses

Osakishimajima is dominated by forest and agricultural land (Fig 2) made up of citrus fields. The island has well-drained soil which makes it perfect to grow citrus rendering it famous for its rich cultivation of Mikan

(Japanese mandarin) and lemon.

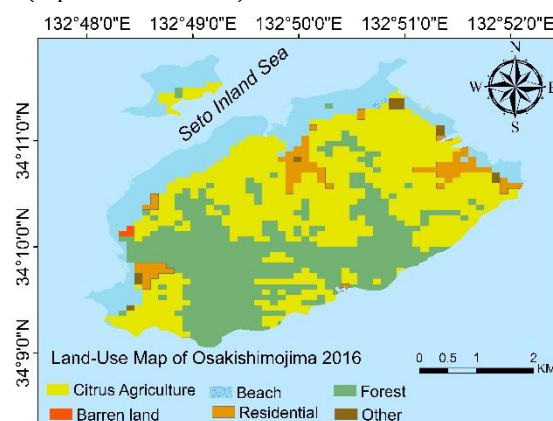


Fig.2 Land-use types of Osakishimajima and sampling locations

Because of its sheltered location and gentle climate, Osakishimajima is Japan's citrus island taking advantage of the calm air, strong sunlight, light rainfall, and good mountain drainage, which is so important for citrus orchards cultivation.

2.2 Sample Collection

Fifty-one water samples (49 wells, 1 stream, and 1 spring) were collected during October 2019 from the villages of Mikado and Kubi in Osakishimajima island (Fig. 1). Samples of each point were collected in acid-washed 250 ml polyethylene bottles after rinsing three times with the sample water and preserved airtight to avoid evaporation. Physicochemical parameters such as pH, electrical conductivity (EC), dissolved oxygen (DO), and water temperature were measured in situ using portable devices. The values of these parameters were recorded after being stabilized. TDS was calculated from EC by an empirical formula $TDS = 0.64 \times EC$ as described by [10]. Samples were filtered through a 0.2 μ m polytetrafluoroethylene filter and preserved at 4°C in a refrigerator before analysis. Alkalinity measurements were carried out by acid titration with 0.02N H₂SO₄ added to each sample to reach its titration endpoint within 24 hours of sample collection.

2.3 Parameter and Chemical Analysis

Concentrations of major ions in water samples were determined in the Biogeochemistry laboratory at Hiroshima University, Japan. Analyses for major ion chemistry included sodium (Na⁺), potassium (K⁺), calcium (Ca²⁺), magnesium (Mg²⁺), chloride (Cl⁻), nitrate (NO₃⁻), bicarbonate (HCO₃⁻), and sulfate (SO₄²⁻) by ion chromatography while the concentrations of Phosphate-phosphorus (PO₄³⁻-P) and Ammonia (NH₄⁺-N) species were determined

with a continuous flow automated nutrient analyzer. Each sample was filtered using a 0.2 μm polytetrafluoroethylene filter and subjected to 10 times and 5 times dilution using distilled water before analyzing for the major ions. Analytical precision for cations and anions was checked using ionic balance error (IBE).

3. RESULTS AND DISCUSSION

3.1 General Groundwater Chemistry and Hydrochemical Facies

3.1.1 General groundwater chemistry

The physico-chemical parameters and statistical summary for all 51 water samples collected from osakishimajima are presented in Table 1. Physical parameters such as pH, DO and EC often gives us the preliminary information that is required in any groundwater study.

Table 1 Statistical analysis of various chemical parameters

Parameters	Min	Max	Mean	STDEV
EC	22.6	543	281.7	115.6
TDS	14.5	347.5	180.3	74
DO	4.2	10.2	8	1.4
pH	6.0	7.4	6.5	0.3
GWT	11.7	20.6	15.6	1.8
Na ⁺	2.55	39.1	14.5	7.9
K ⁺	0.45	13.8	4.4	2.7
Ca ²⁺	5.92	86.3	30.7	14.4
Mg ²⁺	2.03	31.2	10.7	5.7
NH ₄ ⁺ -N	0	3.4	0.5	0.8
HCO ₃ ⁻	0	139.9	53.6	26.9
NO ₃ ⁻	5.4	92.2	34.6	21
NO ₃ ⁻ -N	5.4	20.8	7.8	4.8
PO ₄ ³⁻ -P	0.06	3.2	0.6	0.7
SO ₄ ²⁻	7.6	95.4	41.7	22.1
Cl ⁻	1.94	48.6	14.1	8.9

Note: Ionic concentrations, TDS and DO: mg/L, EC: $\mu\text{S}/\text{cm}$, GWT (groundwater temperature): $^{\circ}\text{C}$

The pH values of the study area ranged from 6.02 - 7.4 with a mean value of 6.5 (Table 1) thus, the groundwater of this area is generally neutral. The maximum depth to the water table (12 m) was relatively low, suggesting the existence of a shallow aquifer system for the observed water sources. The water temperature ranged from 11.7 - 20.6 $^{\circ}\text{C}$ with a mean value of 6.5 $^{\circ}\text{C}$.

In the study area, the lowest pH value was 6.0 and the highest 7.4. Most of the samples were within the range of 6.5 - 7.3 implying that the groundwater is moderately alkaline to acidic. The EC of the groundwater varied between 22.6 to 543 $\mu\text{S}/\text{cm}$ with a mean value of 281.7 $\mu\text{S}/\text{cm}$. The obtained electrical conductivity (EC) values of groundwater samples ranged from 22.6 to 543 $\mu\text{S}/\text{cm}$ (Tables 1). The concentration of total dissolved solids (TDS) in groundwater ranged from 14.5 to 347.5 mg/L (Tables 1). However, [12] categorized water based on TDS concentration into four groups which are represented as fresh (TDS < 1000 mg/L), brackish (> 1000 mg/L), saline (> 10,000 mg/L) and brine (100,000 mg/L). Based on this classification, all the groundwater samples are of the fresh category. Generally, the concentrations of cations and anions vary spatially and were within maximum allowable limits for drinking except a few.

3.1.2 Hydrochemical facies definition

The piper trilinear diagram [13] is used to infer hydro-geochemical facies in water samples. Chemical data of samples from the study area are presented by plotting them on a piper-trilinear diagram (Fig. 3). The plot shows that most of the groundwater samples fall in the CaMg-HCO₃ and CaMg-SO₄ or CaMg-Cl water types field. The majority of the water samples (68 %) belonged to the mixed CaMg-HCO₃ water type. This type indicates mineral dissolution, possibly secondary carbonate and silicate minerals with enough recharge from freshwater. Minor representations of CaMg-SO₄ (32%) may suggest the mixing of freshwater with water from contaminated sources. The order of dominance of ionic species was $\text{Ca}^{2+} > \text{Na}^{+} > \text{Mg}^{2+} > \text{K}^{+}$ and $\text{HCO}_3^{-} > \text{SO}_4^{2-} > \text{NO}_3^{-} > \text{Cl}^{-}$ for cations and anions respectively.

3.1.3 Mechanisms controlling water chemistry

The geochemical process occurring within the groundwater system and reactions with aquifer minerals have a profound effect on water quality. The interaction between rocks and water results in the leaching of ions into the aquifer which subsequently influences the groundwater chemistry. The Gibbs diagram is widely used to establish the relationship of water composition and aquifer lithological characteristics [14]. Three distinctive fields such as precipitation dominance, evaporation dominance, and rock-water interaction dominance areas are shown on the Gibbs diagram (Fig. 4).

The predominant samples fall in the rock-water interaction dominance and a few samples in the precipitation dominance field of the Gibbs diagram. The rock-water interaction dominance field indicates the interaction between rock chemistry

and the chemistry of the percolation waters under the subsurface.

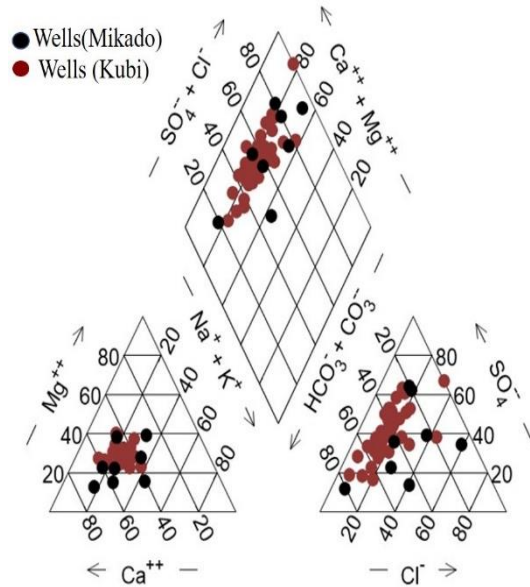


Fig.3 Piper trilinear diagram showing the main Hydrochemical facies of groundwater in Osakishimajima

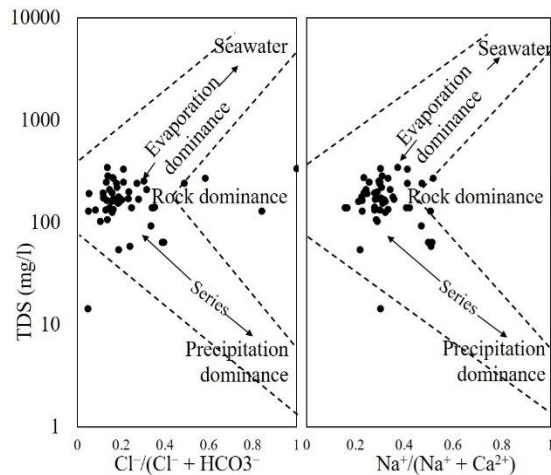


Fig.4 Gibbs plots showing water-rock interaction as the major process regulating the chemistry of waters in Osakishimajima

3.2 Contamination of Groundwater

Phosphates and nitrates are useful nutrients for plant growth but when they become too concentrated in our water system, they become dangerous to both humans and their environment. Leaching of Nitrogen and Phosphorus from arable fields into groundwater usually occurs as a result of high amounts of these nutrients applied in the form of natural and mineral fertilizers needed by plants.

Phosphorus may be moved from point of application by surface run off or moved out of the soil surface layer with percolation [2].

3.2.1 Nitrate-nitrogen (NO_3^- -N) contamination

Nitrate as Nitrogen is the largest applied nutrient in crop production, occurring naturally in the environment and commonly as fertilizer. According to [15], there exist three sources of nitrogen in water; biological fixation, precipitation, and the application of fertilizers however humans significantly affect nitrogen load in the environment.

The warm weather, low rainfall, and volcanic soils of Osakishimajima have promoted the growth of intensive citrus fruit dating back to the 1930s. To maintain and increase crop yield, chemical fertilizers such as Nitrogen, Phosphorous, and Potassium (NPK) fertilizers and pesticides are applied to the fields. Similarly, some organic citrus farmers whose orchards are near the coastline spray their orchard fields with diluted salty seawater to ward off pests and fungus, this certainly contributes to groundwater contamination alongside lithologic effects. NO_3^- -N was obtained by converting NO_3^- to NO_3^- -N using Eq. 1.

$$1 \frac{\text{mg}}{\text{L}} \text{NO}_3^- = 0.226 \frac{\text{mg}}{\text{L}} \text{NO}_3^- - \text{N} \quad (1)$$

NO_3^- -N ranged from 1.2 to 20.8 mg/L with a mean of 7.8 mg/L. Over 70.6% of the samples were within the desirable limit of less than 10mg/l while 29.4% exceeded the maximum contaminant limit of 10 mg/L as recommended by WHO. Plotting NO_3^- -N against well depth presented a homogenous pattern (Fig. 5) ranging from 2 to 13 m with decreasing depth. Such distribution may indicate the superficial origin of the lower percentage of NO_3^- -N confirming the unconfined nature of the groundwater system. Usually, shallow groundwater sources are more vulnerable to NO_3^- -N contamination due to natural N-fixation under aerobic conditions.

According to [3], leaching of Nitrogen and Phosphorus from arable fields to groundwater occurs as a result of high amounts of these nutrients applied in the form of natural and mineral fertilizers needed by plants. Since nitrate is highly leachable and readily moves with water through the soil profile, in case of excessive rainfall or over-irrigation, NO_3^- -N will be leached below the plant's root zone and eventually reach the aquifer system as may be the case in Osakishimajima. However, these results differ from the findings of [16] who observed that groundwater samples from Indramayu exhibited higher NH_4^+ -N concentrations as opposed to NO_3^- -N in arable fields under similar conditions. Table 2 characterizes water samples

based on the adopted maximum contaminant level (MCL) set by WHO [12].

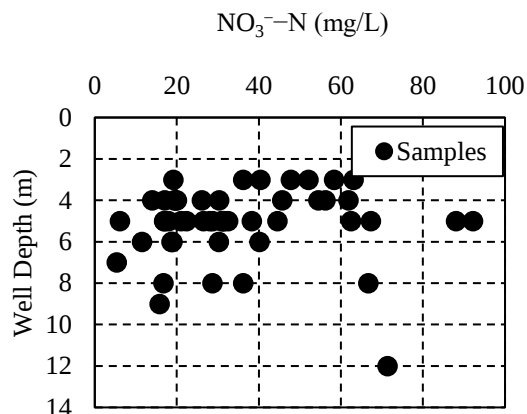


Fig.5 Plot of well depth against NO_3^- -N concentrations in groundwaters

Table 2 Summary of MCL for NO_3^- -N and PO_4^{3-} -P concentrations

[A]	[B]	[C]	[D]	[E]
NO_3^- -N	<10	36	70.6	No
	>10	15	29.4	Yes
PO_4^{3-} -P	<0.03	1	2.0	No
	>0.03	48	98.0	Yes

Note: [A] = parameter (mg/L), [B] = MCL (maximum contaminant limit), [C] = number of wells, [D] = % of wells, [E] = contamination status

3.2.2 Phosphate-phosphorus (PO_4^{3-} -P) contamination

Phosphorus in the natural ecosystem is derived from the erosion of rocks but can be added to the soil through chemical fertilizers, manure, and composted materials which may be moved from point of application by surface runoff or moved out of the soil surface layer by percolation [7]. PO_4^{3-} -P in groundwater of the study area varied slightly with depth (Fig. 6). The PO_4^{3-} -P for water sources ranged from 0 to 3.2 mg/L with a mean of 0.6 mg/L. Thirty samples were within the depths of 4 to 5 m, six samples fell between 6 to 7 m, eight samples within 5, and one sample at 12 m. The concentration of phosphate was found to be more than the WHO permissible limit in ninety-eight (98%) of the water samples (Table 2). It was observed that the input of Phosphorus fertilizers by farmers in Osakishimajima is higher (167 kg P per ha) than the recommended input of (160 kg P per ha) by the government. This would explain the observed high concentrations of PO_4^{3-} -P in the samples and at shallow depths between 3 to 12 m (Fig. 6).

According to [17], phosphorus is retained in the soil by adsorption but once the capacity of the soil to adsorb phosphorus is exceeded, the excess will

dissolve and move more freely with water either directly to a stream or down to an aquifer, this process may be responsible for the high observed values of PO_4^{3-} -P in most of the sample sites.

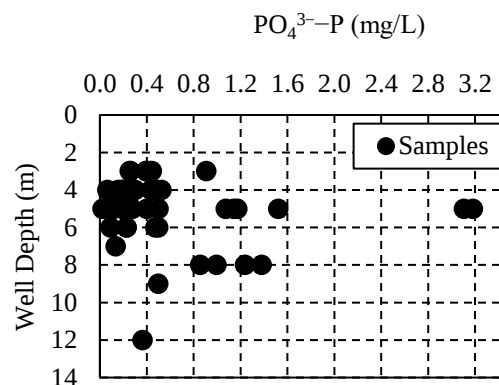


Fig.6 Plot of wells depth against PO_4^{3-} -P concentrations in groundwaters

According to WHO [11], the MCL of phosphate in drinking water is 0.03 mg/L. Consumption of water with phosphate concentration more than this limit causes disease of the kidney and osteoporosis in human beings [18]. The observation of the high concentration of PO_4^{3-} -P and NO_3^- -N in some well samples can be a result of the thin layering of the soil strata as well as the permeability of the soil.

4. CONCLUSION

The types of water that dominated the study area are CaMg-HCO_3 and mixed CaMg-SO_4 water types. The Gibbs diagram reveals that most of the water samples fall in the rock-water interaction dominance field. Groundwater in Osakishimajima is contaminated by both Nitrate-nitrogen and Phosphate-phosphorus. Most of the samples were within the maximum allowable limit except a few samples for NO_3^- -N. On the other hand, PO_4^{3-} -P exceeded the maximum contaminant limit in all but one sample. The application of high amounts of NPK fertilizers to increase crop yields may be the major source of the nutrient; PO_4^{3-} -P and NO_3^- -N in the groundwater. High PO_4^{3-} -P in groundwater poses a future risk of eutrophication in surrounding surface water bodies thus groundwater management strategies are necessary to sustainably protect the groundwater system. Future studies should be focused on using phosphate isotope tracers to determine the point sources of PO_4^{3-} -P in the aquifer system.

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