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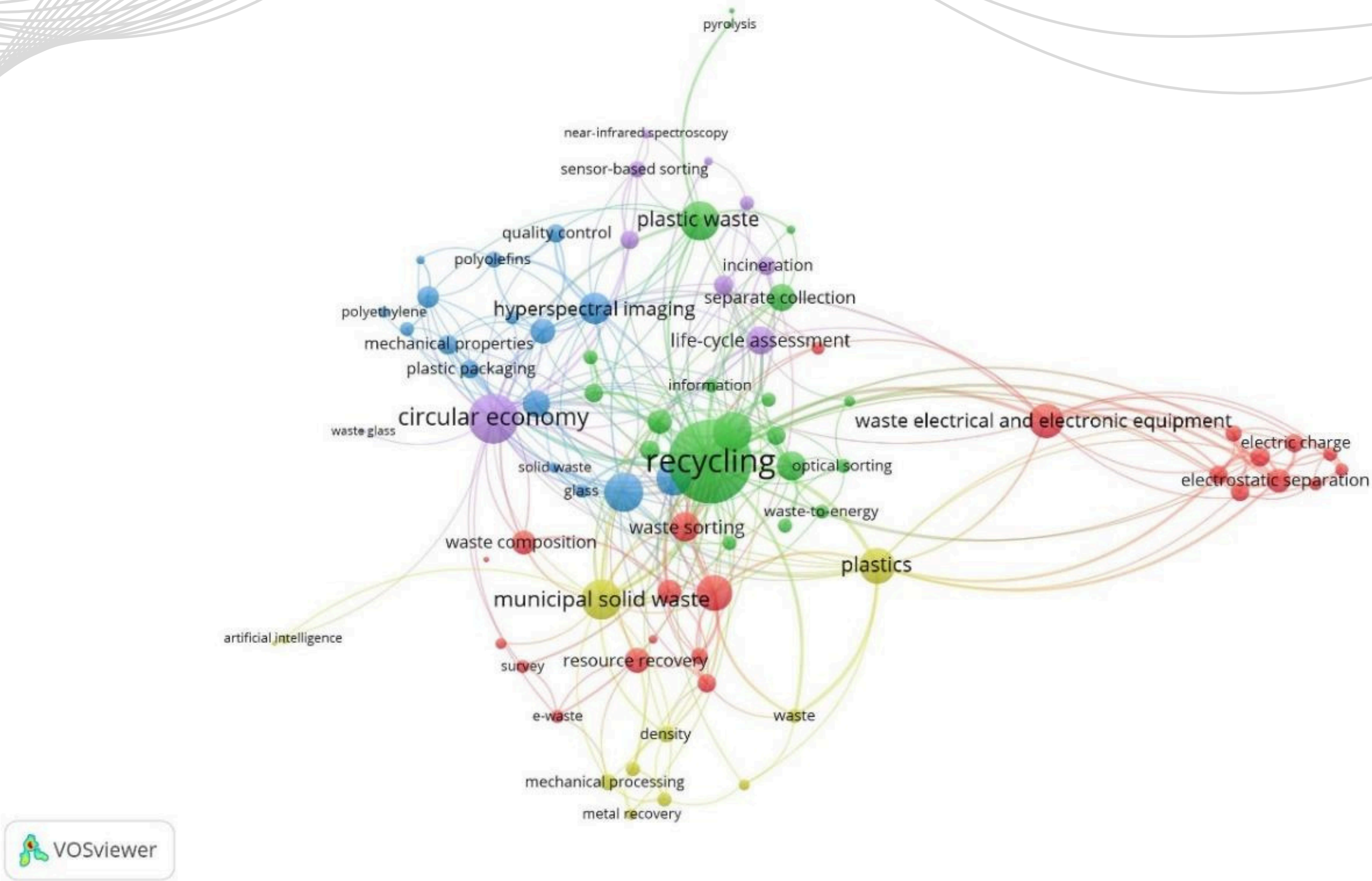

Bibliometric networks of keywords related to sustainable waste management solutions including organic and inorganic waste sorting sorting and recycling technologies, waste management strategies, and integration of AI and economic principles.

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Phone +662 441 5000 ext. 2108
Website: https://ph02.tci-thaijo.org/index.php/ennrj/index
E-mail: ennrjournal@gmail.com

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CONTENT

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Assessment of Microplastic Pollution Load and Ecological Risk Index in Surface Water and Sediment Matrices in the Mangrove Ecosystem along Butuan Bay, Southern Philippines

Ethel P. Keleste[1], Jolina S. Fedelis[1], Wendy Faye T. Opusa[1], Sherley Ann T. Inocente[2,3], Marybeth Hope T. Banda[2,4], Felmer S. Latayada[1], Joycelyn C. Jumawan[5], Temmy P. Vales[1], Romell A. Seronay[6], and Rey Y. Capangpangan[2,7*]

[1]Department of Chemistry, College of Mathematics and Natural Sciences, Caraga State University, Butuan City 8600, Philippines
[2]Research on Environment and Nanotechnology Laboratories, Mindanao State University at Naawan, Naawan, Misamis Oriental 9023, Philippines
[3]Department of Forest Sciences, College of Agriculture, Forestry and Environmental Sciences, Mindanao State University at Naawan, Naawan, Misamis Oriental 9023, Philippines
[4]Science Education Institute, Department of Science and Technology, DOST Compound, Bicutan, Taguig City 1631, Philippines
[5]Department of Biology, College of Mathematics and Natural Sciences, Caraga State University, Butuan City 8600, Philippines
[6]Department of Environmental Science, College of Forestry and Environmental Sciences, Caraga State University, Butuan City 8600, Philippines
[7]Department of Physical Sciences and Mathematics, College of Marine and Allied Sciences, Mindanao State University at Naawan, Poblacion, Naawan, Misamis Oriental 9023, Philippines

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*** Corresponding author:**
E-mail: rey.capangpangan@msunaawan.edu.ph

ABSTRACT

Microplastics have garnered attention for their ubiquitous presence across various ecosystems, yet investigations into their occurrence in mangrove environments, particularly in the Philippines, remain minimal and increasingly necessary. This study seeks to establish a foundational understanding of microplastic distribution in Philippine mangrove forests, with a special focus on Butuan Bay. Surface water and sediment samples were collected from mangrove habitats in Butuan City, Buenavista, and Nasipit along Butuan Bay, in the Southern Philippines. Results indicated that Buenavista (BMS) exhibited the highest microplastic concentrations, with mean abundances of 322.22±103.41 MPs/m^3 in surface water and 88.89±50.33 MPs/kg in sediments. Both blue and transparent microplastics were prevalent, constituting 24.2% and 25.7% of surface water and 22.6% and 25.7% of sediments, respectively. Microplastics predominantly fell within the 101-250 µm range (46% in surface water and 45.7% in sediments), with films and fragments comprising 69% of surface water MPs and 76.1% of sediment MPs. Ten polymer types were identified, with polypropylene (PP) being the most abundant (31.5% in surface water and 51.7% in sediments). The assessment of the pollution load index (PLI) indicated that MP pollution levels were classified as slightly polluted (hazard level I), and ecological risk index (RI) posed varying degrees across the studied mangroves, ranging from minor danger (hazard level I) to extreme danger (hazard level V). Microplastics were observed to pose ecological risks within these mangrove areas, as indicated by the highest levels of risk shown by BMS and BCMS. Future studies should examine other surrounding waters of Mindanao, such as the adjacent Gingoog Bay, including inland freshwater bodies, to comprehensively assess MP pollution and its potential effects on marine wildlife in the region.

1. INTRODUCTION

Plastics, due to versatility and cost-effectiveness, are extensively manufactured worldwide. In 2021, global plastic production surged to 390.7 billion kg from 1.5 billion kg in 1950 (Statista, 2021; Vlasopoulos et al., 2023). Despite utility, plastics pose significant environmental concerns due to slow biodegradation. A 2017 study showed that only a fraction of the 8,300 billion kg of plastics produced between 1950 and 2015 were

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recycled, with the majority ending up in landfills and oceans (Geyer et al., 2017). Over time, discarded plastics break down into microplastics, smaller than 5mm (Arthur et al., 2009), found in various environmental compartments, including sediments, water bodies, and marine organisms (Liu et al., 2021a; Sayed et al., 2021; Zantis et al., 2021), human tissues (Ragusa et al., 2021), and blood (Leslie et al., 2022).

Microplastics (MPs), due to size, pose significant dangers as contaminants, progressing undetected through the food chain and carrying toxic heavy metals (Brennecke et al., 2016) and persistent organic pollutants (Santana-Viera et al., 2021). Urgent action is needed globally to isolate, dispose of, and prevent further environmental contamination from microplastics.

Mangrove ecosystems are among the few places with a strong tolerance to pollutants and purification capacities for heavy metals, petroleum, and other domestic sewage contaminants (Liu et al., 2022b). Mangroves prevent coastal erosion, promote shoreline protection (Meera et al., 2022), and serve as the last barrier preventing river pollutants from reaching the ocean (Liu et al., 2022b). Nonetheless, the boundary between mangrove areas and land has been identified as a microplastic (MP) accumulation hotspot (Chen, 2022).

Research indicates that mangroves act as plastic sinks (Deng et al., 2021; Martin et al., 2020), accumulating toxic metals and pollutants (Bayen, 2012). Mangroves are now considered one of the ecosystems severely threatened by microplastic pollution from marine and terrestrial sources (Prarat et al., 2024). Despite the Philippines' rich mangrove biodiversity (Garcia et al., 2013) and ranking 15th globally (Giri et al., 2011), only two studies have investigated MP presence in Philippine mangroves, particularly in sediments (Bonifacio et al., 2022; Navarro et al., 2022a). Organisms in mangrove ecosystems likely ingest MPs, which is alarming as they are food sources for nearby communities. Plastics adsorb heavy metals and pollutants (Naik et al., 2019), emphasizing the need for baseline data on microplastic pollution in mangroves to guide mitigation efforts.

Butuan Bay hosts diverse habitats, including mangroves, amid industrialized cities and municipalities. Tourism and economic growth in these areas lead to inevitable plastic pollution, endangering bay biodiversity, mainly mangroves. Only a few studies have documented microplastics in Butuan Bay mangroves, focusing on sediments (Navarro et al., 2022a) and the clam *Polymesoda erosa*, a species commonly found in the mangroves of Cabadbaran, Buenavista, and Nasipit in Butuan Bay, Mindanao Philippines (Navarro et al., 2024b). This study aims to document the microplastic presence, abundance, and types in selected mangrove sites along Butuan Bay's western portion, including Butuan City, Buenavista, and Nasipit. These sites were chosen for their extensive mangrove cover along waterways leading to Butuan Bay.

2. METHODOLOGY

2.1 Study area

The study focused on Butuan Bay in northeastern Mindanao, Philippines, an extension of the Bohol Sea, receiving water from various sources, including the Agusan River, the country's third-longest. It supports diverse habitats like mangroves. Sampling occurred at three mangrove sites along Butuan Bay: Barangay Masao (BCMS), Barangay Matabao (BMS), and Barangay Ata-atahon (NMS) chosen for extensive mangrove cover (Figure 1). Sampling in September 2022 collected nine surface water and sediment samples from each site for microplastic analysis. These mangroves provide crucial industrial, economic, and topographic benefits to the local community.

2.2 Sample collection

Transect lines were established with three sampling points spaced roughly 50 meters apart. Following Tien et al. (2020), surface water samples were manually collected using 1-liter glass bottles, capturing 20 liters within the top 50 cm. Samples were sieved through stacked metal sieves (5.0 mm and 0.25 mm) to remove debris larger than 5.0 mm (Egessa et al., 2020). Particles on the 0.25 mm sieve were rinsed, transferred to glass jars, and covered with aluminum foil. A total of 27 surface water samples (nine per site) were collected.

For sediment sampling (Masura et al., 2015), about 300 g of wet sediments were scraped up to 1 cm deep at each sampling point using a metal shovel or grab sampler. Samples were stored in glass containers covered with aluminum foil and kept in the dark until analysis. Nine sediment samples were taken from each mangrove site, totaling 27 samples overall.

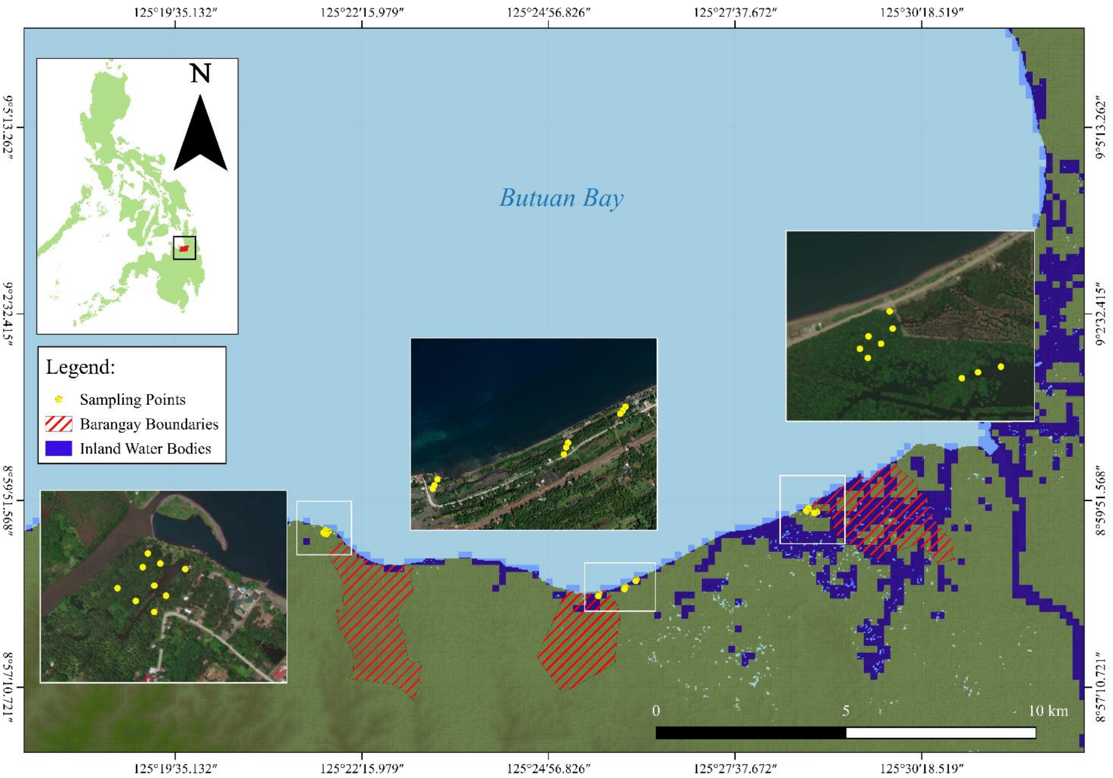

Figure 1. Study area for microplastic assessment in mangrove sites along Butuan Bay, Agusan del Norte, Philippines

2.3 Sample processing

Samples of surface waters and sediments underwent pretreatment to remove impurities and organic matter according to the methods of Inocente et al. (2023). Sediment samples were oven-dried at 90°C for 24 h (Inocente et al., 2023; Masura et al., 2015). Dried sediments (150 g) and surface water samples were digested individually in 10% KOH at 40°C for 40 h (Inocente et al., 2023; Karami et al., 2017) and 60°C for 24 h (Inocente et al., 2023; Dehaut et al., 2016) to dissolve organic material. The resulting supernatants were filtered using a 47 mm Whatman GF/C glass microfiber filter under vacuum. Density separation was conducted on unclear samples with residues by adding a 30% aqueous NaCl solution at a 2:1 ratio and constant agitation for 2 h. The solution was vacuum filtered using Whatman GF/C glass microfiber filters. Filter films were dried overnight in a clean petri dish before isolating suspected microplastics.

2.4 Identification and quantification of micro-plastics

Microfiber filter films were observed with an Amscope® Dissecting Microscope at 20-40X magnification. Suspected microplastics were isolated on glass slides with a clean needle and labeled. They were then counted and classified by color and morphotype following criteria by Hidalgo-Ruz et al. (2012) and Su et al. (2016a). Using an Amscope® camera connected to a Motic microscope and TCapture software (version 5.1.1.0), photographs were taken, and sizes were measured based on (Li et al., 2020a). Universal Attenuated Total Reflectance Fourier-Transform Infrared (UATR-FTIR) Spectroscopy (PerkinElmer Spectrum Two, USA) determined microplastic polymer type. Spectral data were compared to a plastics library (Jung et al., 2018) at a 0.50 correlation score for identification, considering sample size, impurities, and degradation.

2.5 Quality control

To ensure experimental accuracy, non-plastic lab equipment was prioritized, and regular station cleaning minimized sample contamination. Laboratory windows remained closed to prevent interference from dust and foreign materials. The glassware was thoroughly rinsed with distilled water and stored covered with aluminum foil to prevent airborne contamination. The experimental blank controls containing distilled water were processed following the same procedure for the collected samples. As procedural blanks, clean filters were also exposed inside the laboratory alongside filters containing samples. All samples and blanks were run in triplicates. No MPs were detected in all the blanks.

2.6 Data treatment

The experimental data were analyzed using Microsoft Excel 2016 and the PAST Statistical tool v.4.03. To determine the abundance of microplastics in the surface water and the sediments, the following formulas were used:

$$\text{MP Abundance (Surface Water)} = \frac{\text{number of confirmed microplastics}}{\text{volume of the water sample (m}^3\text{)}} \quad (1)$$

$$\text{MP Abundance (Sediments)} = \frac{\text{number of confirmed microplastics}}{\text{dry mass of the sediment sample (kg)}} \quad (2)$$

These formulas were modified and derived from by Liu et al. (2021a) and Egessa et al. (2020). The Shapiro-Wilk test was utilized to check for the normality of the data. One-way ANOVA test (with Tukey's test) or Kruskall-Wallis test (with Dunn's test) was used to test the variance between the site's mean microplastic abundances. The significance level was assigned at 0.05.

2.7 Ecological risk assessment

This study assessed the potential ecological risks associated with MP pollution in these mangroves along Butuan Bay using the pollution load index (PLI_i) and the ecological risk index (RI). The PLI_i (Tomlinson et al., 1980; Yang et al., 2023) was calculated based on Equations (3) to (5):

$$C_f^i = \frac{C_s^i}{C_b^i} \quad (3)$$

$$PLI_i = \sqrt{C_f^i} \quad (4)$$

$$PLI_{zone} = \sqrt[n]{PLI_1 \times PLI_2 \times \ldots \times PLI_n} \quad (5)$$

Where; C_f^i is the ratio of the MP concentration at each sampling site (C_s^i) and the background MP concentration (C_b^i). The background MPs used in this research have the lowest concentration observed in the studied mangrove areas due to the lack of available data for background MP concentration for mangrove waters and sediments in the southern part of the Philippines to be used as reference. The PLI_{zone} represents the MP pollution load index for mangroves along Butuan Bay and is calculated as the square root of the product of PLI_i obtained from all sampling sites. Moreover, by employing the Håkanson method (Hakanson, 1980; Peng et al., 2018) as a guide, the toxicity response factor (Tr^i) was computed, along with the ecological risk factor (Er^i), and the ecological risk index (RI) for the environment via Equations (6) to (8):

$$Tr^i = P_n \times S_n \quad (6)$$

$$E_r^i = Tr^i \times C_f^i \quad (7)$$

$$RI = \sum_{i=1}^{n} Er^i \quad (8)$$

Where; Tr^i represents the toxicity response factor corresponding to each polymer constituent, and as described by the Lithner approach (Lithner et al., 2011), this factor is calculated by multiplying the percentage of each MP polymer type in the total sample at each sampling site (P_n) by the hazard score associated with the polymer type (S_n) (Lithner et al., 2011). S_n is the hazard score for each polymer type of MP. The hazard score values for polypropylene (PP), polyethylene terephthalate (PET), general purpose polystyrene (GPPS), polyvinyl chloride (PVC), poly (methyl methacrylate) (PMMA), ethylene vinyl acetate (EVA), high-density polyethylene (HDPE), low-density polyethylene (LDPE), polyamide 6 (NY6), and polyamide 66 (NY66) were 1, 4, 0, 10 001, 1 021, 9, 11, 11, 50, and 63, respectively (Lithner et al., 2011; Yang et al., 2023; Prarat et al., 2024). The sum of the ecological risk factors for each plastic polymer is the environment's ecological risk index (RI). The criteria for the risk level of MP pollution are presented in Table S2.

3. RESULTS AND DISCUSSION

3.1 Microplastic abundance and distribution in surface water and sediments

Over 96% of the collected surface water and sediment samples in this study contained microplastics, signifying the prevalent distribution of microplastics in three mangrove areas in Butuan Bay. No confirmed microplastics were detected in all experimental blank controls used in the study.

Microplastics were found in 26 of 27 surface water (SW) samples from three mangrove sites in Butuan Bay. Among the 439 suspected microplastics isolated, 124 (28%) were confirmed via UATR-FTIR analysis. BMS had the highest concentration at 322.22±103.41 MPs/m^3, followed by NMS at 250±114.56 MPs/m^3 and BCMS at 116.67±106.07 MPs/m^3 (Table 1). Statistical analysis revealed significant differences in mean microplastic abundances between mangrove sites ($p<0.05$), particularly between Butuan City, Buenavista, and Nasipit.

On the other hand, microplastics were present in 26 of 27 sediment (Sed) samples from selected mangrove sites in Butuan Bay. Among 442 suspected microplastics extracted, about 52% (230 particles) were confirmed. BMS had the highest abundance at 88.89±50.33 MPs/kg, followed by NMS at 55.56±30.91 MPs/kg and BCMS at 25.93±20.93 MPs/kg (Table 1), matching surface water findings. Statistical analysis revealed significant differences in mean abundance between mangrove areas ($p<0.05$), particularly between Butuan City and Buenavista.

Table 1. Microplastics abundance in surface water and sediments of Butuan Bay Mangroves

Sampling area	Number of suspected MPs		Number of confirmed MPs		Abundance of confirmed microplastics (MPs)	
	SW	Sed	SW	Sed	No. of MPs/m^3 surface water	No. of MPs/kg sediment
Butuan City (BCMS)	140	142	21	35	116.67±106.07	25.93±20.93
Buenavista (BMS)	137	176	58	120	322.22±103.41	88.89±50.33
Nasipit (NMS)	162	124	45	75	250.00±114.56	55.56±30.91
Total	439	442	124 (28%)	230 (52%)	229.63±135.35	56.79±43.53

Variations in microplastic abundance among sites suggest environmental and anthropogenic influences (Ronda et al., 2023; Syversen and Lilleng, 2022). In one study (Zhou et al., 2020), microplastics were most prevalent in tourist-heavy mangrove areas, correlating with Buenavista's high microplastic levels. Anthropogenic activities, including waste dumping and proximity to tourist sites and fishing grounds, likely contribute. Larger plastic debris, such as packaging and clothing, was commonly found during sampling. Furthermore, there is a positive correlation between the distance from populated centers and the abundance of plastics found in mangrove areas (Garcés-Ordóñez et al., 2019).

The high microplastic concentration in Buenavista's sediments may result from denser microplastics settling from surface waters (Deng et al., 2021). Floating microplastics may sink due to increased density from organism biofouling (Kowalski et al., 2016). Sediments are significant microplastic sinks (Martin et al., 2020).

Surface water microplastic abundance (229.63±135.35 MPs/m^3) in Butuan Bay mangrove sites was three times higher than in Chabahar Bay, Iran, and Kingston Harbour, Jamaica (Aliabad et al., 2019; Rose and Webber, 2019), but thirty-six times lower than in Beibu Gulf, China (Li et al., 2020b) (Table 2). Sediment microplastic abundance (56.79±43.53 MPs/kg) in Butuan Bay was higher than in Iran and Singapore (Naji et al., 2019; Nor and Obbard, 2014) but lower than in Jinjiang Estuarine and Beibu Gulf, China, and Colombian mangrove habitats (Deng et al., 2020; Garcés-Ordóñez et al., 2019; Zhang et al., 2020). Comparable to 2022 data in Cabadbaran City, Buenavista, and Nasipit, Butuan Bay (Navarro et al., 2022a) (Table 3). Nearby Panguil Bay reported microplastic occurrence (62.72±18.31 MPs/m^2) (Bonifacio et al., 2022). Variances in global mangrove microplastic reports reflect local conditions and anthropogenic impacts.

Table 2. Comparison of microplastic abundance in surface water of mangrove areas

Study Area	MPs/m^3	Reference
Butuan Bay, Philippines	229.63±135.35	This study
Goiana Estuary, Brazil	477	Lima et al. (2016)
Kingston Harbour, Jamaica	0.76	Rose and Webber (2019)
Yunxiao Mangrove Reserve, China	275	Pan et al. (2020)
Beibu Gulf, China	399-5’531	Li et al. (2020b)
Chabahar Bay, Iran	0.14±0.06	Aliabad et al. (2019)

Table 3. Comparison of microplastic abundance in sediments of mangrove areas

Study area	MPs/kg	Reference
Butuan Bay, Philippines (Butuan City, Buenavista, Nasipit)	56.79±43.53	This study
Butuan Bay, Philippines (Cabadbaran, Buenavista, Nasipit)	53.34±21.87	Navarro et al. (2022a)
Panguil Bay, Philippines	62.72±18.31*	Bonifacio et al. (2022)
Five mangrove habitats, Iran	19.5±0.36-34.5±0.71	Naji et al. (2019)
Seven mangrove habitats, Singapore	36.8±23.6	Nor and Obbard (2014)
Six mangrove habitats, Colombia	31-2,863	Garcés-Ordóñez et al. (2019)
Jinjiang Estuarine, China	1,926±351	Deng et al. (2020)
Beibu Gulf, China	273±23-3,520±107	Zhang et al. (2020)

* MPs/m^2

3.2 Morphological characteristics of microplastics

This section details data and analyses of morphological characteristics (color, morphotype, size) and chemical composition (polymer type) of microplastics from three Butuan Bay mangrove sites in the Philippines. Identifying these characteristics and their abundance informs plastic origins and degradation pathways, which are valuable for future mitigation efforts.

3.2.1 Color

Plastics were initially colored for market appeal (Imhof et al., 2016), leading to diverse colors detected in the environment due to limited recycling. This study found 14 colors in microplastics from surface water and sediment samples: white, green, blue, brown, transparent, black, yellow, pink, gray, mixed, tan, orange, violet, and red (Figure 2).

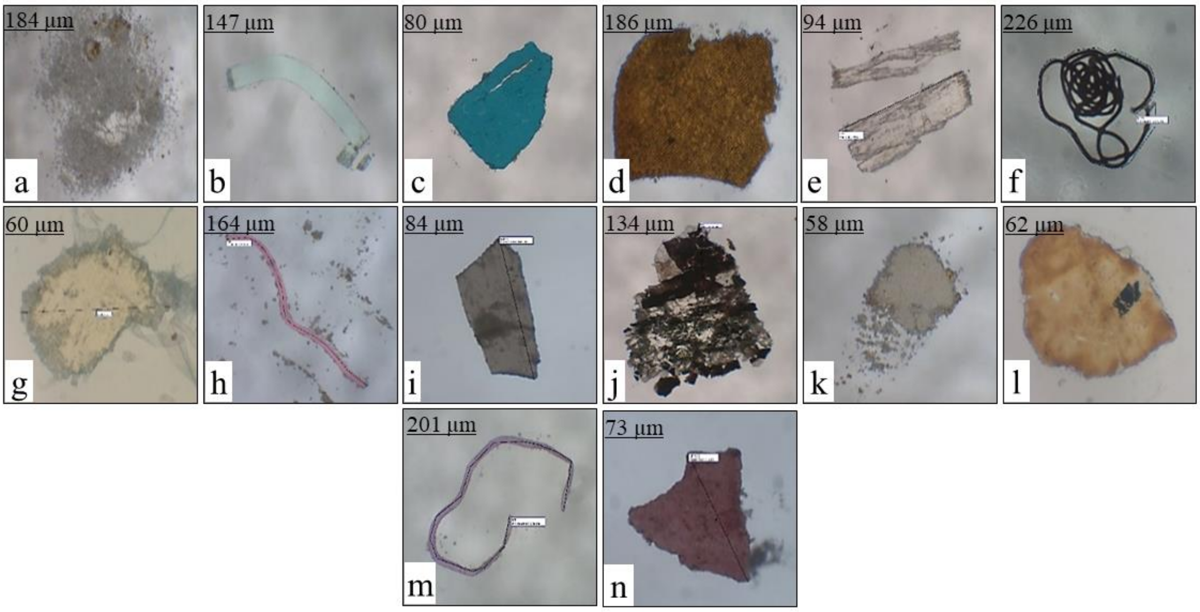

Figure 2. Varying colors, morphology, and sizes of confirmed microplastics from selected mangrove areas

Blue and transparent microplastics prevailed in both water and sediment samples, comprising 22.6% to 25.7% each, respectively (Figure 3). This color distribution aligns with findings from Singaporean coastal mangroves (Nor and Obbard, 2014), Butuan Bay sediments (Navarro et al., 2022a), and Fish Cage Water samples in Butuan and Nasipit (Similatan et al., 2023). However, the results of this study differ from those of Banda et al. (2024), who found that brown microplastics were prevalent in the water samples and white microplastics in the sediment samples of the Taguibo River, which also empties into Butuan Bay. This discrepancy can be attributed to the differing nature of the samples: while our study focuses on coastal samples, Banda et al. (2024) analyzed freshwater samples. These microplastics, mainly PP, LDPE, HDPE, and PET, likely originate from everyday items like bags, containers, and bottles.

Environmental exposure may bleach microplastics and make them transparent (Fan et al., 2019).

Microplastic colors and morphotypes serve as indicators of origin and can be bioindicators of environmental pollution (Lavers et al., 2016). Colors influence ingestion rates in biota, resembling prey colors (Wright et al., 2013). The prevalence of blue and transparent microplastics in mangroves is concerning, as Asian clams predominantly ingest them (Su et al., 2018b). Mud clams, which share similar feeding habits, are likely to consume them too. Given mud clams' status as a staple food in mangrove communities, they pose as potential vector for human microplastic ingestion.

3.2.2 Morphotype

Microplastics were categorized into six morphotypes: fiber, filament, film, foamed, granules, and fragment (Hidalgo-Ruz et al., 2012; Su et al., 2016a) (Figure 2). A fiber is slender and elongated, while a filament has a broader band. Film refers to a thin layer of plastic debris; foamed microplastics are spongy, and granules appear grainy. Fragments are thicker pieces of broken plastic debris used when a microplastic particle does not fit other categories.

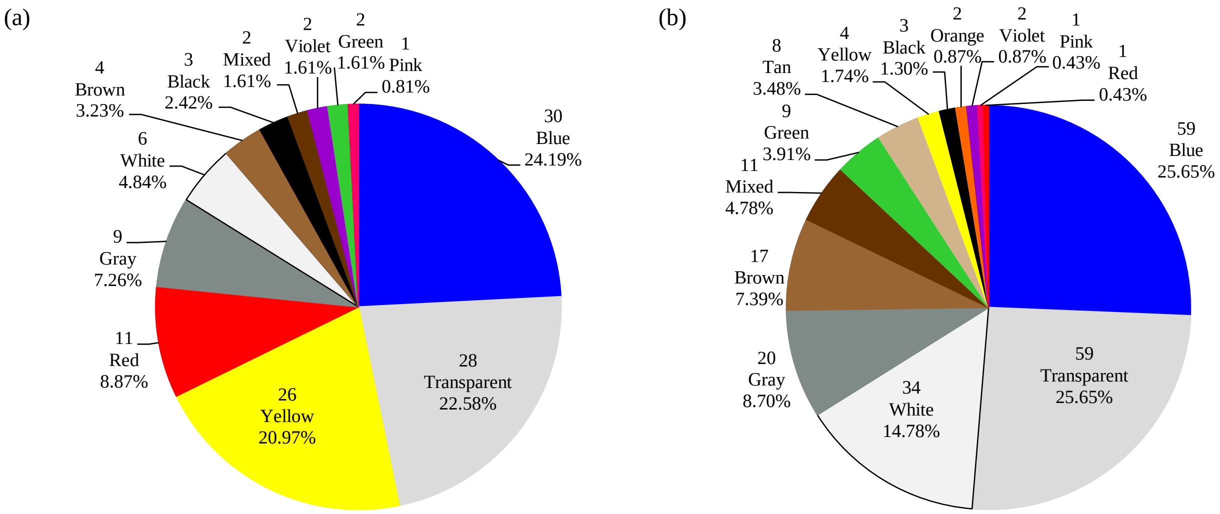

Figure 3. Color distribution of the confirmed microplastics between sample types (a) surface water and (b) sediment

Only four microplastic morphotypes (fiber, film, filament, fragment) were observed in surface water samples. At the same time, all six were found in sediment samples (Figure 4). Fragmented micro-plastics predominated in surface water (41.9%). In contrast, film-dominated sediment samples (39.6%) (Figure 4). Fiber and filament were third and fourth most abundant in both water and sediment samples across mangrove sites. These findings align with those from the Persian Gulf shoreline, South African estuaries, and Manila Bay River mouths (Govender et al., 2020; Nabizadeh et al., 2019; Osorio et al., 2021). Water morphotypes detected in this study differ from Similatan et al. (2023) from Nasipit samples (fiber) and Banda et al. (2024) (fiber) but agree with the Butuan water morphotypes of Similatan et al. (2023). Sediment morphotypes do not align with the results of Navarro et al. (2022a) and Banda et al. (2024). Films mainly derive from single-use plastic degradation, while fragments originate from larger plastic debris (Martin et al., 2020; Wang et al., 2019), such as containers, bottle caps, and food packaging. Proximity to human settlements suggests microplastics originate from household and business waste. The prevalence of films and fragments underscores poor plastic waste management in surrounding communities.

3.2.3 Size

Microplastic sizes were categorized as <50 μm, 50-100 μm, 101-500 μm, and >500 μm (Li et al., 2020a) (Figure 2). Most microplastics fell in the 101-500 μm range, followed by 50-100 μm, with some <50 μm and >500 μm sizes detected (Figure 5). These findings align closely with Govender et al. (2020), who found 47% and 75% of mangrove microplastics in the surface water and sediments, respectively, were <500 μm. Microplastic ingestion is heavily influenced by size and abundance, especially for benthic species (Rodríguez-Seijo and Pereira, 2017). Sizes of 50-500 μm are small enough for indiscriminate ingestion by mangrove organisms, impacting their growth and reproduction (Cole et al., 2015). Since many of these organisms are food sources, microplastics will likely enter the local food web. The prevalence of microplastics within 50-500 µm may suggest in-situ biodegradation by mangrove-endemic microorganisms (Auta et al., 2022; Kannahi and Sudha, 2013).

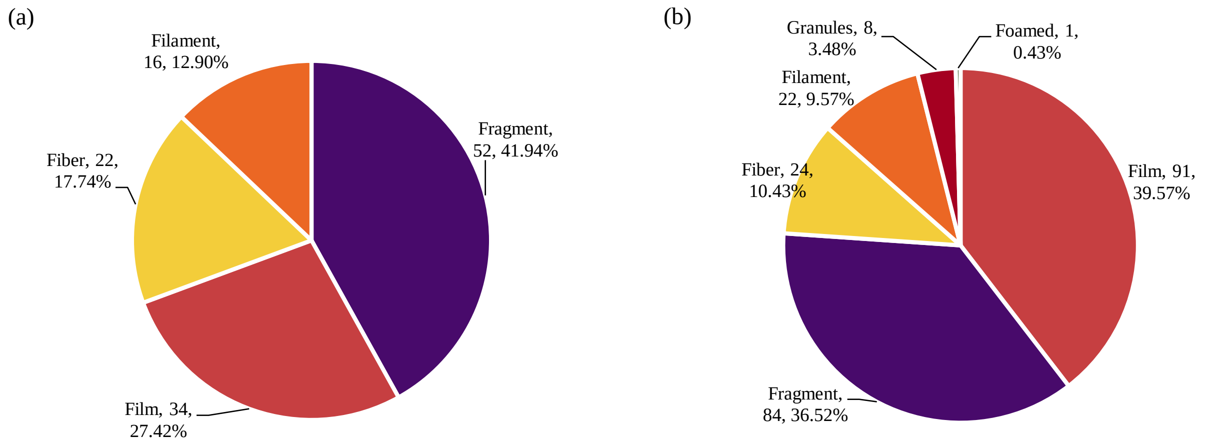

Figure 4. Morphotype distribution of confirmed microplastics between types of samples - (a) surface water and (b) sediment

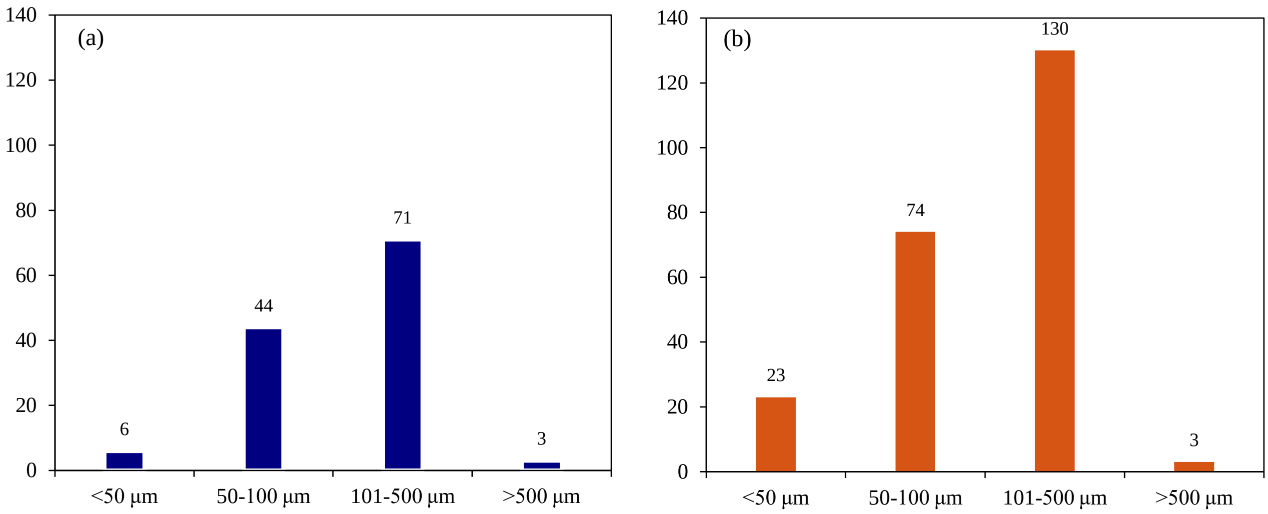

Figure 5. Size distribution and comparison of confirmed microplastics between types of samples - (a) surface water and (b) sediment

3.2.4 Polymer type

Ten polymer types of microplastics were confirmed in surface water and sediment samples via UATR-FTIR: ethylene vinyl acetate (EVA), general purpose polystyrene (GPPS), high-density polyethylene (HDPE), low-density polyethylene (LDPE), polyamide 6 (NY6), polyamide 66 (NY66), polyethylene terephthalate (PET), poly (methyl methacrylate) (PMMA), polypropylene (PP), and polyvinyl chloride (PVC). PP and LDPE were most abundant in both sample types, followed by PET and HDPE (Figure 6). Representative IR spectra of PP and LDPE show characteristic peaks (Figure 7). These findings align with Navarro et al. (2022a) on microplastics in Butuan Bay sediments. Results for water differ from Similatan et al. (2023), in which both sites (Barangay Camagong, Nasipit, and Barangay Ata-atahon, Nasipit) have EVA as the predominant polymer detected. Results also differ from those of Banda et al. (2024), who found polyacetylene and regenerated cellulose fibers to be the predominant polymers in water samples. PP, widely produced globally, likely originates from everyday items like bags, containers, and packaging (Geyer et al., 2017; Grand View Research, 2022; Jones et al., 2020; SpecialChem, 2024). The COVID-19 pandemic likely contributed to PP microplastics, especially from disposable masks (DFMs), known to release and capture microplastics (Chen et al., 2021). LDPE, HDPE, and PET, also predominant, originate from various products like bags, bottles, and packaging (Jones et al., 2020). Other polymers (EVA, GPPS, PVC, PMMA, NY6, NY66) likely come from diverse sources such as foam, electrical equipment, and textiles (Amann and Minge, 2011; Jones et al., 2020; ScienceStruck, n.d.).

It should be noted that the polymer types detected are all common plastic polymers widely used in various applications. The prevalence of these polymer types may reflect the economic and social activities, as well as the consumption preferences of the population around these sampling sites. The Philippines is often referred to as having a "sachet economy", characterized by a heavy reliance on small, single-use packages for standard household products such as shampoos, powdered drinks, and other daily necessities. These sachets are typically composed of polyethylene (PE), low-density polyethylene (LDPE), and polypropylene (PP) (Ahmed et al., 2023; Allahvaisi, 2012). This widespread use of sachets is driven by economic factors (Ang and Sy-Changco, 2007), such as affordability and convenience, which cater to the needs of consumers who prefer or can only afford to purchase products in smaller quantities (Manalo and Manalo, 2022; Gomez et al., 2023). Additionally, the cultural preference for such packaging formats contributes to the high prevalence of these plastic polymers in the environment.

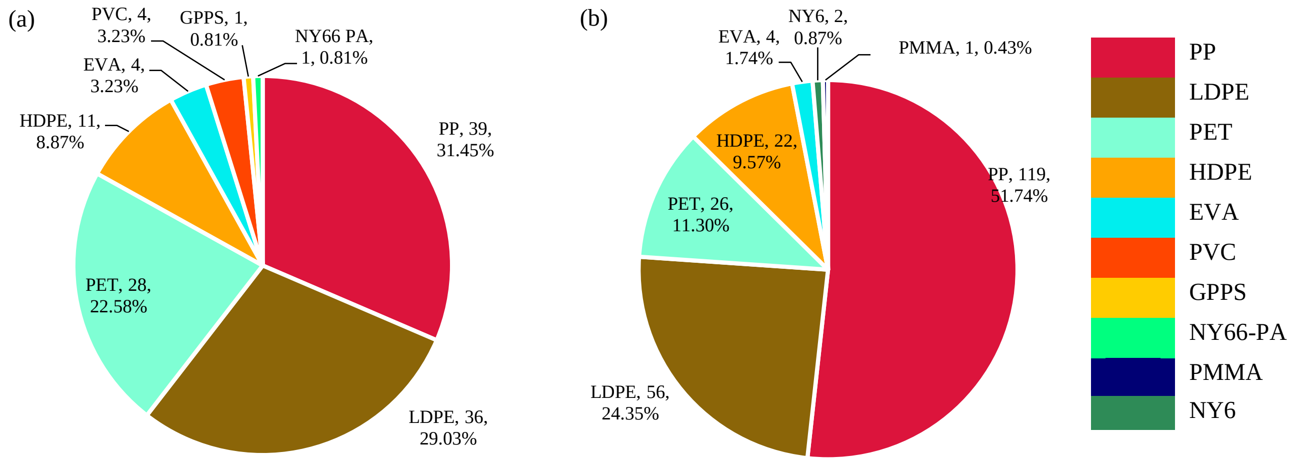

Figure 6. Microplastic polymer type distribution and comparison of the confirmed microplastics between sample types - (a) surface water and (b) sediment

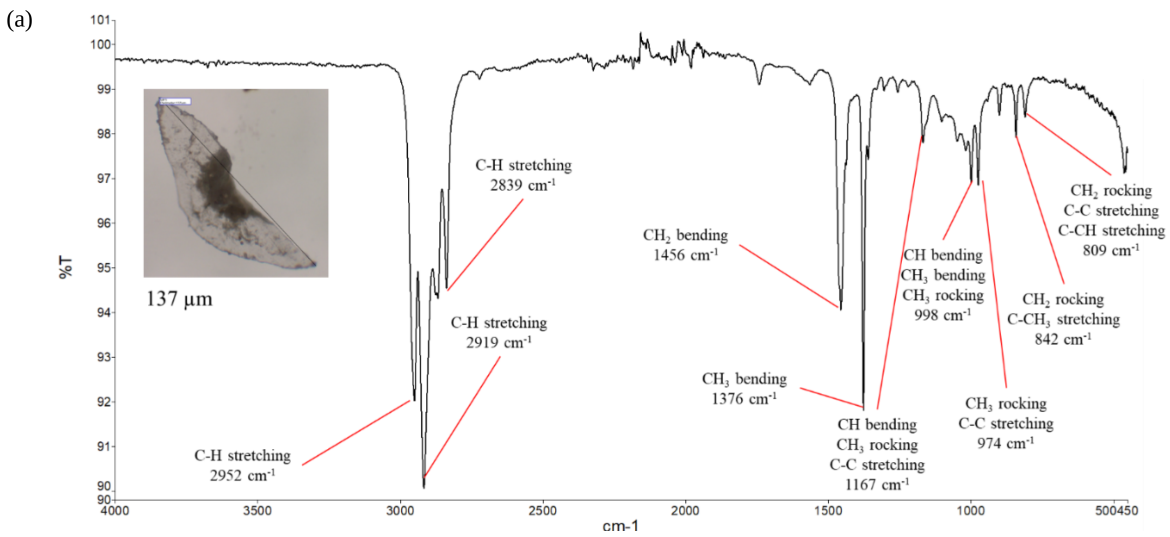

Figure 7. Representative IR Spectra of the two most dominant polymer types in the surface water and sediments of selected mangrove sites. (a) Polypropylene, and (b) Low-Density Polyethylene

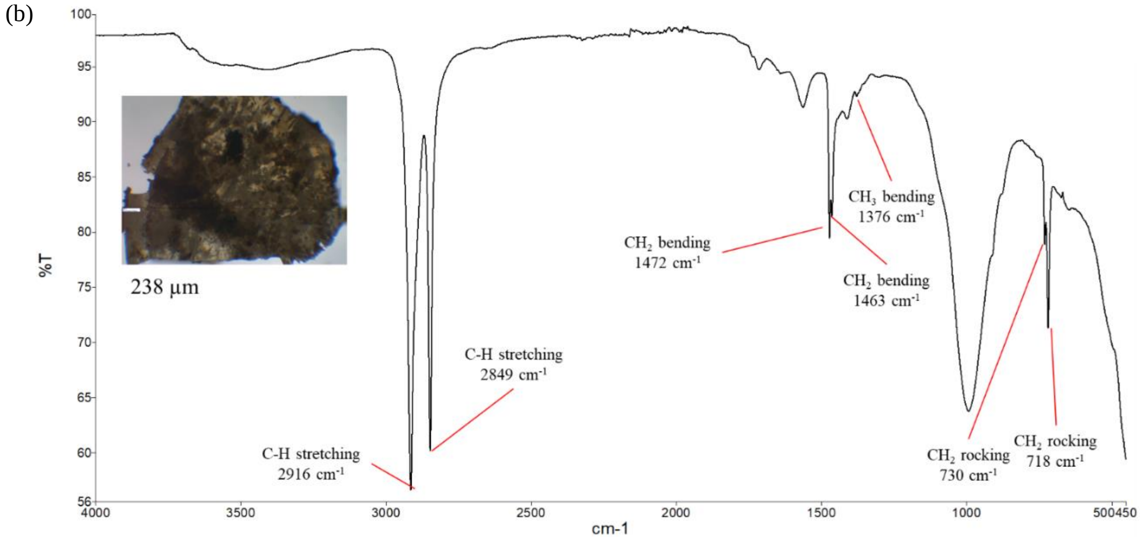

Figure 7. Representative IR Spectra of the two most dominant polymer types in the surface water and sediments of selected mangrove sites. (a) Polypropylene, and (b) Low-Density Polyethylene (cont.)

3.3 Ecological risk posed by MPs in mangrove surface waters and sediments

This study combines the PLI and RI models to assess the ecological risks of MP pollution in mangrove surface waters and sediments along Butuan Bay. As shown in Figure 8 and Table S5 (Supplementary data), the MP pollution levels in both study samples in the mangrove areas were classified as slightly polluted (hazard level I) based on the PLI_{zone} values. NMS mangroves ranked the highest among the surface water samples, followed by BCMS and BMS mangrove areas. On the other hand, BMS ranked the highest, followed by BCMS and then NMS for sediment samples. These findings were found close to the reported observations in other mangrove areas of Southern China, which reported a hazard level of I (<10), except for the Futian mangrove, which had a higher hazard level (II, 10-20) (Li et al., 2020a; Dou et al., 2021), also in both dry and wet seasons data in the southern Hainan Island, China (Yang et al., 2023), and comparable to KB, PT, and SH and lower than the MR and LB sediment sampling stations in the coastal mangroves in Eastern Thailand (Prarat et al., 2024). Although the PLI model estimates low overall microplastic (MP) pollution in this study and other areas, the ecological impact of MPs in mangrove ecosystems should not be underestimated. This is because the PLI model does not account for the potential toxicity of different polymer types.

Moreover, to investigate the ecological risk of MPs, the RI model was used considering both the MP hazard data and MP concentrations. The RI values from the studied mangrove areas for both sample types, surface waters and sediments, ranged from 737.94 to 3548.83 and 36.35 to 213.60 (Figure 8 and Table S5), respectively. Both BMS and BCMS mangroves score the highest ecological risk of MP pollution at level V (extreme danger, >1,200) due to the high abundance of MPs in surface waters, while NMS falls to the danger category at level IV (600≤RI≤1,200) also due to its high MP abundance. It is worth noting that the RI values of the sediment samples from BCMS fall only to the medium danger category level II (150≤RI<300), while BMS and NMS fall to minor danger (level I) with RI values both less than 150. This disparity of the RI values of surface water and sediment samples can be attributed to the differences in the abundance and type of polymers detected. The RI values obtained for surface water samples in this study (risk levels IV-V) were higher than those reported for southern Hainan Island (risk levels II-IV) in China for wet and dry seasons (Yang et al., 2023). However, for sediment samples, this study has low ecological risk levels, which were in the range I-II lower than those found in mangroves of Southern China (risk levels III-V) (Li et al., 2020a) and in Eastern Thailand (risk levels III-V) (Prarat et al., 2024).

Consequently, our risk assessment of MP pollution based on PLI and RI models revealed varying ecological risks to the mangrove ecosystems in Butuan Bay, Southern Philippines, with BMS and BCMS carrying the highest ecological risk. It was evident that the MP pollution in the study mangrove areas was notably widespread, and this presents the likelihood of MP ingestion by the tiny organisms, which could transfer or move up to the food chain in the mangrove food web and could lead to the bioaccumulation of MPs via biomagnification together with their associated pollutants and eventually posing health risks to human being through food consumption of mangrove animals (Parolini et al., 2023; Prarat et al., 2024).

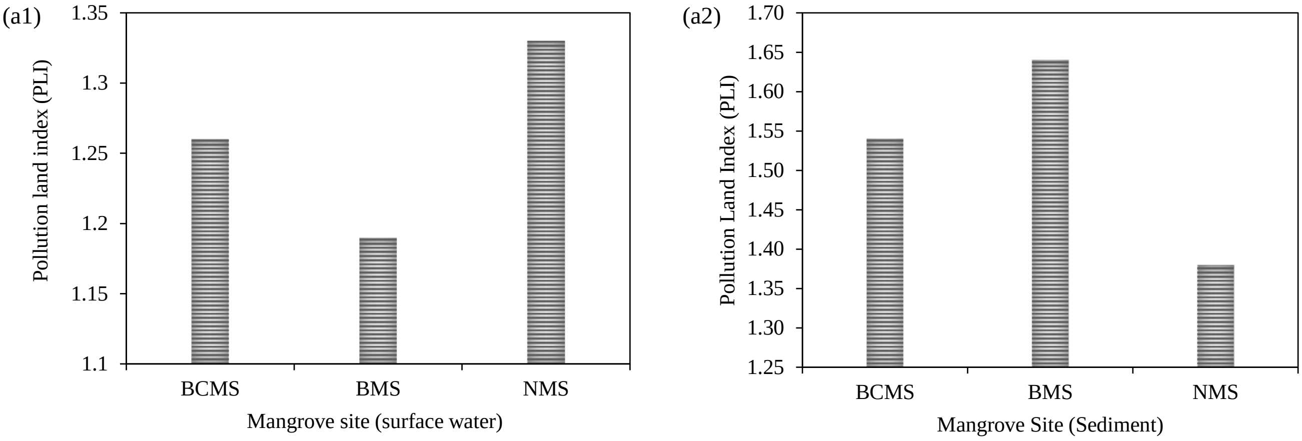

Figure 8. The pollution load index (PLI) (a1 and a2) and ecological risk index (RI) of microplastic pollution (b1 and b2) in mangrove areas located along Butuan Bay, Southern Philippines. Letters I, II, III, IV, and V indicate the risk level categories.

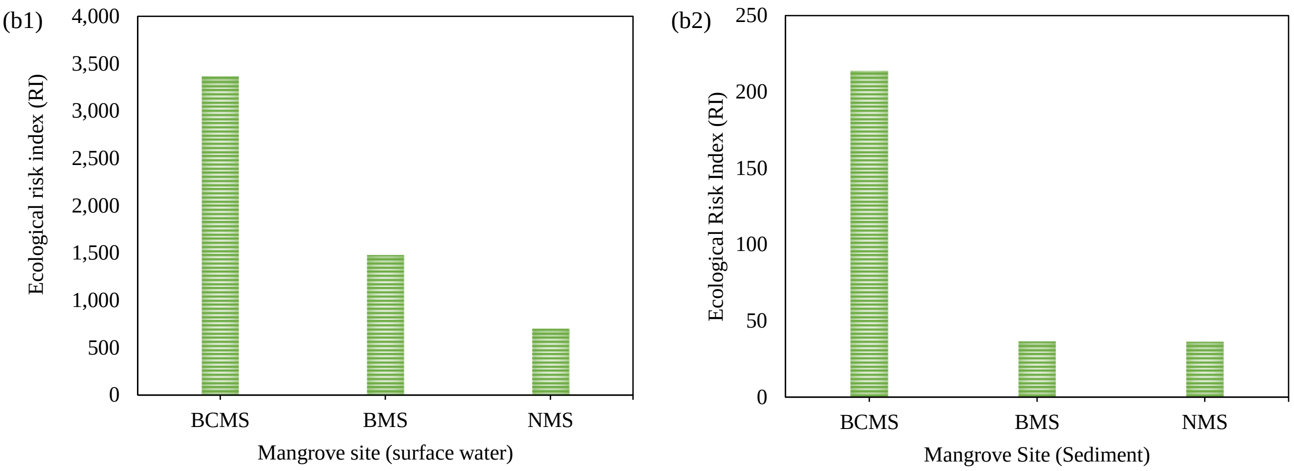

Figure 8. The pollution load index (PLI) (a1 and a2) and ecological risk index (RI) of microplastic pollution (b1 and b2) in mangrove areas located along Butuan Bay, Southern Philippines. Letters I, II, III, IV, and V indicate the risk level categories (cont.).

4. CONCLUSION

The results of this study indicate that microplastics are present in all the selected mangrove sites along Butuan Bay, Southern Philippines. This again supports the notion that mangrove areas retain microplastics from marine and terrestrial domains. The morphology of the extracted microplastics suggests that it is of secondary origin and must have come from household and business establishments around the mangroves. Additionally, the prevalence of polypropylene suggests that these microplastics hail from everyday household and establishment litter, such as pieces of discarded clothing, textiles, plastic food packaging, plastic bags, plastic cups, etc., indicating unrestrained local waste dumping of plastic waste in the area. Moreover, the dominance of blue and transparent colored microplastics is especially alarming. Asian clams, which have feeding behaviors similar to those found in mangroves, have been found to ingest them, preferably. This increases the likelihood of humans consuming microplastic as clams remain a vital food source in communities around the area. In addition, the relatively small microplastics - mostly <500 µm - found in the sites pose significant threats to the biodiversity of the living organisms in the areas. Smaller-sized microplastics mean easy and faster uptake of this type of pollutant by the biota endemic to the mangrove territories, thus affecting their growth and propagation.

Furthermore, the ecological risk assessment, based on PLI and RI models, revealed differing levels of ecological risk linked to MP pollution across studied mangrove areas. The data gathered in this study establishes baseline information for better policy formulation and stricter implementation of the existing measures countering plastic pollution in the city and municipalities involved. These findings also offer crucial baseline data for future research, assisting in point source tracing and pollution monitoring to safeguard the mangrove and coastal ecosystems.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

Conceptualization: Ethel P. Keleste, Jolina S. Fedelis, Wendy Faye T. Opusa, Felmer S. Latayada, and Temmy P. Vales. Data Curation: Ethel P. Keleste, Jolina S. Fedelis, Wendy Faye T. Opusa, Temmy P. Vales, and Felmer S. Latayada. Investigation: Ethel P. Keleste, Jolina S. Fedelis, Wendy Faye T. Opusa, Sherley Ann T. Inocente. Formal Analysis: Ethel P. Keleste, Jolina S. Fedelis, Wendy Faye T. Opusa, Temmy P. Vales, and Felmer S. Latayada. Methodology: Marybeth Hope T. Banda and Sherley Ann T. Inocente. Project Administration: Rey Y. Capangpangan, Marybeth Hope T. Banda, and Sherley Ann T. Inocente. Resources: Rey Y. Capangpangan, Felmer S. Latayada, Joycelyn C. Jumawan, Romell A. Seronay, Temmy P. Vales. Supervision: Felmer S. Latayada, Temmy P. Vales, Sherley Ann T. Inocente. Writing - original draft: Ethel P. Keleste, Felmer S. Latayada, Marybeth Hope T. Banda. Writing - review and editing: Ethel P. Keleste, Felmer S. Latayada, Marybeth Hope T. Banda, Joycelyn C. Jumawan, Romell Seronay.

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Effects of Glucose and Lime Levels on the Growth and Water Quality of *Chlorella ellipsoidea* Cultivation

Rapeepan Yongyod[1], Pichasit Sangmek[2], and Narong Kamolrat[2*]

[1]*Faculty of Public Health, Kasetsart University Chalermphrakiat Sakon Nakhon Province Campus, Sakon Nakhon, Thailand*
[2]*Faculty of Natural Resources and Agro-Industry, Kasetsart University, Chalermphrakiat Sakon Nakhon Province Campus, Sakon Nakhon, Thailand*

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*** Corresponding author:**
E-mail: narong.ka@ku.th

ABSTRACT

This study aimed to investigate the effects of glucose and lime concentrations in the culture medium on growth of *Chlorella ellipsoidea* and water quality during the cultivation. The experiment included seven treatment groups: a control group (0.5 g/L glucose, 0.4 g/L lime) and groups with reduced glucose concentrations (0.4, 0.3, 0.2, 0.1 g/L) and lime concentrations (0.2, 0.1 g/L). Cultivation lasted for 15 days, with algal growth assessed by cell counting, and water quality parameters such as alkalinity, pH, and ammonia levels analyzed daily. The results showed that the G20 (0.1 g/L glucose) group achieved highest cell density of $7.15\pm0.92 \times 10^7$ cells/mL on day 10, which was not significantly different ($p>0.05$) from the G40, G60, and L50 groups, but higher than the control and other treatment groups. Furthermore, alkalinity in the G20 group remained within the range of 130-187 mg/L, which is suitable for cultivation and subsequent use as feed for *Moina* sp. The pH values of all groups remained consistent, ranging from 7.2-8.8 throughout the experiment. Total ammonia levels remained below 1 mg/L during the first 9 days, increasing in the later period, with no significant differences between the treatment groups. In conclusion, reducing the glucose concentration to 0.1 g/L in the culture medium promoted maximum growth of *C. ellipsoidea*, while maintaining water quality at an optimal level. This approach offers potential for developing cost-effective algal culture media and scaling up cultivation for commercial production.

HIGHLIGHTS

1. Study explored nutrient formula for *Chlorella ellipsoidea* and measured water quality.
2. G20 formula (0.1 g/L glucose) gave highest growth, safe water quality for *Moina* sp.
3. Reduced algae culture costs, created suitable environment for *Moina* sp. cultivation.

1. INTRODUCTION

Unicellular microalgae such as *Chlorella* sp. are recognized as valuable sources of nutrients, bioactive compounds, and renewable biomass for various applications (Chew et al., 2017; Singh and Patidar, 2018). *Chlorella* sp. is rich in proteins, essential amino acids, vitamins, pigments like chlorophyll and carotenoids, and polyunsaturated fatty acids beneficial for human and animal nutrition (Barka and Blecker, 2016; Kong et al., 2024; Kamolrat et al., 2024). Its superior nutritional profile, ease of cultivation, and ability to accumulate high biomass make *Chlorella* sp. a promising feedstock for live feed production in the aquaculture industry (Muller-Feuga et al., 2003). *Chlorella* sp. cells possess spherical morphology, exist as single cells without mucilage and setae (Safi et al., 2014; Kim et al., 2018), making them highly suitable as feed for aquatic animal larvae or as a nutritional source for zooplankton, which are commonly used as feed in larval aquaculture and ornamental fish cultivation (Lan et al., 2022; Joshua et al., 2024). Among the important cultivated zooplankton species,

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Moina sp. is particularly prevalent. *Moina* sp. is a crucial live feed organism for aquatic animal larviculture and ornamental fish rearing. As a member of Subphylum Crustacea within the Order Cladocera, it represents an essential zooplankton in natural food web systems. Its significance stems from its appropriate body size matching the mouth dimensions of fish larvae and small ornamental fish species (Gogoi et al., 2016). The organism exhibits high reproductive rates, filter-feeding behavior, and possesses rich nutritional composition, including essential amino acids, highly unsaturated fatty acids, and vitamins (Rottmann et al., 2003; Shidik et al., 2021; Rasdi et al., 2020; Suhaimi et al., 2022; Wang et al., 2022). These characteristics have made it widely adopted in aquaculture operations. Consequently, efficient mass production of *Moina* sp. is vital to meet the increasing demand for quality live feeds in aquaculture hatcheries.

However, the cultivation of Chlorella algae as feed for *Moina* sp. often faces challenges in controlling water quality in the culture system, particularly the increase in alkalinity, pH, and accumulation of ammonia to levels that can significantly impact the survival and growth of *Moina* sp. (Benider et al., 2002; Yang et al., 2012; Shidik et al., 2021). Alkalinity levels above 200 mg/L can significantly reduce the survival rate of *Moina* sp. juveniles (Benider et al., 2002; Rottmann et al., 2003). The optimal pH for *Moina* sp. growth is between 7.0 and 8.0. This pH range supports the best growth and reproduction of *Moina* sp. If the pH is too low or too high, the production rates significantly decrease (Yuslan et al., 2021; Rottmann et al., 2003), and the optimal ammonia level for the growth of *Moina mongolica* should not exceed 2.63 mg (NH_3-N)/L. This level is lower than the harmful threshold for aquatic insect larvae (4.3 mg (NH_3-N)/L) but higher than the optimal level for *Daphnia magna* (0.66 mg (NH_3-N)/L) (He et al., 2001).

The main components in algal culture media contributing to increased alkalinity and pH are carbon sources like molasses or ammonium hydroxide and the alkalizing agent lime ($CaCO_3$) (Ilavarasi et al., 2011). Glucose, used as the primary carbon source, and lime are crucial in maintaining water quality in closed photobioreactor systems or indoor cultivation, enhancing water transparency for light absorption (Thewaratmaneekun et al., 2006). Monitoring ammonia levels in algal culture water is essential before introducing *Moina* sp. into the pond to prevent nutrient accumulation and high concentration (American Public Health Association, 2005). While previous studies have optimized algal culture media for general purposes (American Public Health Association, 2005; Thewaratmaneekun et al., 2006; Ilavarasi et al., 2011), research specifically investigating suitable glucose and lime levels for *Chlorella* sp. cultivation aimed at producing feed for *Moina* sp. is lacking. Maintaining optimal water quality parameters is crucial for high survival and growth rates of *Moina* sp. when using algae as feed. The economic and environmental importance of producing high-quality and high-quantity *Moina* sp. in the aquaculture industry is significant. By improving *Moina* sp. production, reliance on natural live feeds can be reduced, preserving natural resources and enhancing farm productivity. Furthermore, reducing the impact on natural ecosystems from harvesting wild live feeds can have positive environmental outcomes.

This study aims to develop practical *Chlorella ellipsoidea* cultivation processes that can be applied in the industry by determining the appropriate levels of glucose and lime for optimal *C. ellipsoidea* growth while maintaining suitable water quality for subsequent use as feed for *Moina* sp. Additionally, advancements in photobioreactor designs and mixotrophic cultivation strategies could improve Chlorella cultivation for live feed purposes (Acién Fernández et al., 2019; Yu et al., 2023). Enhancing this process will increase the efficiency of live feed production in aquaculture farms, benefiting both the economy and the environment.

2. METHODOLOGY

2.1 Algal strain and culture conditions

C. ellipsoidea strain TISTR 8261 used in this study was isolated from local natural water bodies using plankton net sampling (Figure 1). Unialgal culture was established through single-cell isolations by micropipette washing technique (Stein, 1973). The isolated cells were transferred to illuminated test tubes containing Bold's basal medium for growth. After approximately 4 weeks, the culture was scaled up in Erlenmeyer flasks until sufficient cell density was achieved for experimentation. The algae were maintained under aseptic conditions to prevent contamination (Joseph and Ajithkumar, 2015) in a culture room equipped with fluorescent lighting at 3,000 lux and temperature controlled at 25±2°C.

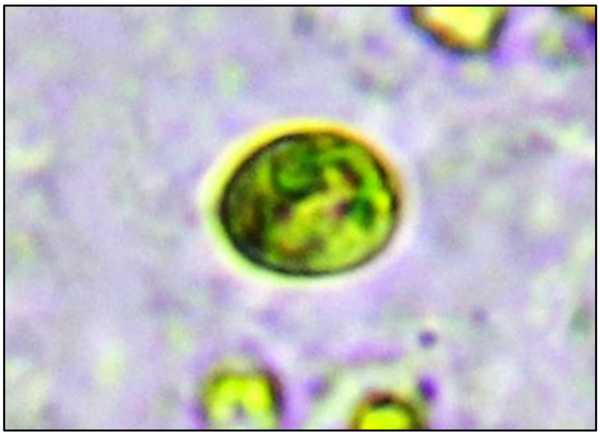

Figure 1. Microscopic view of *C. ellipsoidea* TISTR 8261 cells in culture at 400× magnification

2.2 *C. ellipsoidea* culture medium preparation

The basal culture medium consisted of (g/L): urea (CH_4N_2O) 0.6, phosphorus pentoxide (P_2O_5) 0.2, and potassium oxide (K_2O) 0.2, with carbon sources from glucose ($C_6H_{12}O_6$) and lime ($CaCO_3$). Previous studies on *Chlorella* sp. cultivation have reported glucose concentrations ranging from 0.5-2.5 g/L in culture media. In this study, the culture medium was optimized with initial concentrations of glucose at 0.5 g/L and lime at 0.4 g/L. The experiment was divided into seven treatment groups with varying levels of glucose and lime (Figure 2).

The experiment was designed as a Completely Randomized Design (CRD) with three replications per treatment (n=21). Nutrient solutions were prepared by dissolving each component in 1,000 mL Erlenmeyer flasks and made up to a final volume of 750 mL with distilled water. Each flask was equipped with air inlet and outlet tubes and sealed with cotton plugs. Initial *C. ellipsoidea* inoculum with cell density of 10^6 cells/mL or higher was added at 20 mL/L to each experimental group, resulting in an initial *C. ellipsoidea* concentration of 2.1×10^5 cells/mL. *C. ellipsoidea* cultures were maintained in a controlled environment room at 25±2°C, illuminated by fluorescent lamps providing 3,000 lux intensity with a 24:0 hour light: dark cycle.

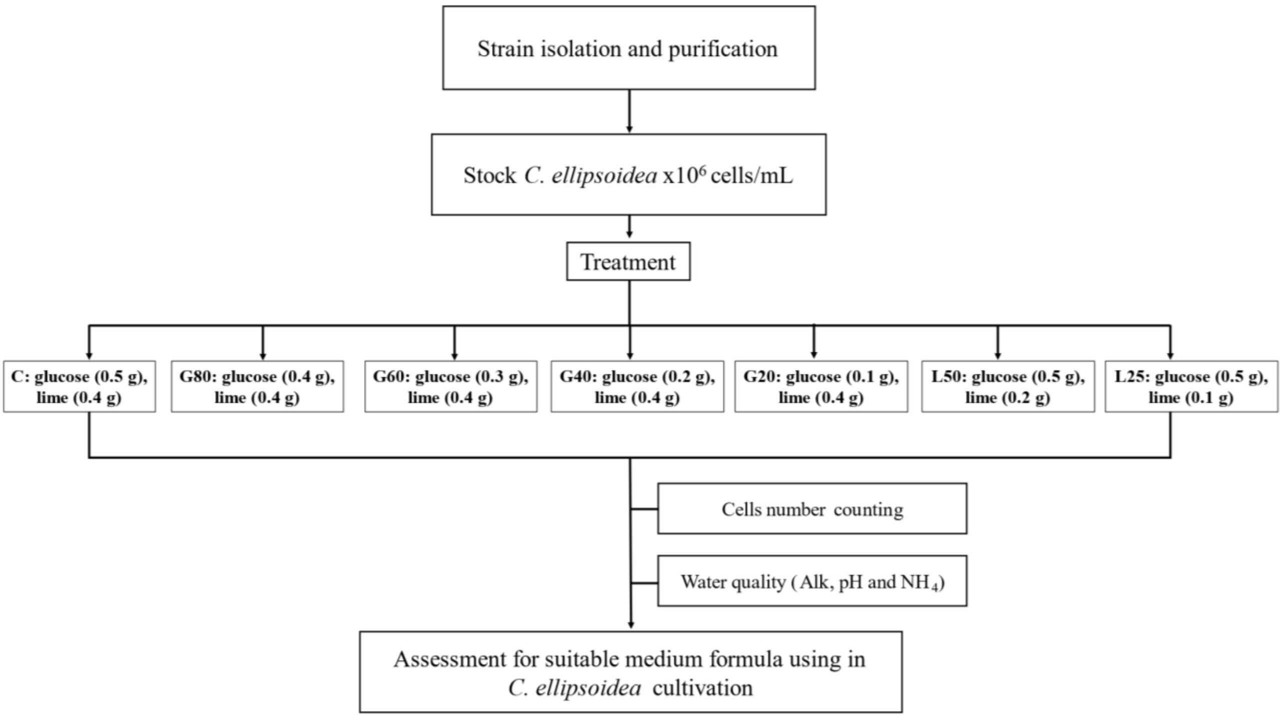

Figure 2. Flowchart of *C. ellipsoidea* culture system in the laboratory

2.3 Sampling and analysis

C. ellipsoidea cells samples (5 mL) were collected daily at 11:00 am to study growth rates through cell counting, from the initial inoculation until *C. ellipsoidea* reached death phase. Cell counting was performed using a hemocytometer (1/10 mm; BOECO; Germany) under a microscope at 400x magnification by randomly counting five small squares as described by Absher (1973). The total algal cells per milliliter were calculated using the following formula:

$$\text{Total cells/mL} = \frac{\text{grid}\,(1+2+3+4+5)}{5} \times 0.25 \times 10^6$$

Water quality parameters were monitored daily at 11:00 am throughout the culturing period, where alkalinity was measured by titration with standardized HCl acid and reported as mg/L $CaCO_3$ equivalent based on standardized $NaHCO_3$ solution, pH was determined using a digital pH meter (resolution 0.01), and Total ammonia nitrogen (TAN) was analyzed using the Phenate method involving sample digestion with alkaline hypochlorite and sodium nitroprusside oxidizing solution, followed by absorbance measurement at 640 nm wavelength against an ammonium chloride standard curve and reported as mg/L of nitrogen.

2.4 Statistical analysis

Algal growth was analyzed for statistical differences using one-way analysis of variance (ANOVA), with mean comparisons between treatment groups performed using Duncan's new multiple range test. The relationship between glucose and lime concentrations and algal growth was evaluated using Pearson's correlation analysis. A 95% confidence level ($p<0.05$) was set for accepting statistical significance. The SPSS software version 22 was used for statistical computations and analyses.

3. RESULTS

3.1 Growth rate of *C. ellipsoidea*

Glucose level in the culture medium significantly affected the growth rate of *C. ellipsoidea*. The group with an 80% reduction in glucose (G20) exhibited the highest growth in both the lag and log phases. During the initial 3-day lag phase and on day 6 of the log phase (Figure 3), the G20 group showed notably higher cell densities compared to other groups. On day 10, when the algae reached maximum cell density, the G20 group attained 7.15×10^7 cells/mL, while the control and other glucose reduction groups ranged from $2.70\text{-}5.47\times10^7$ cells/mL. Statistical analysis revealed no significant difference between the G20 group and the G40, G60, and L50 groups.

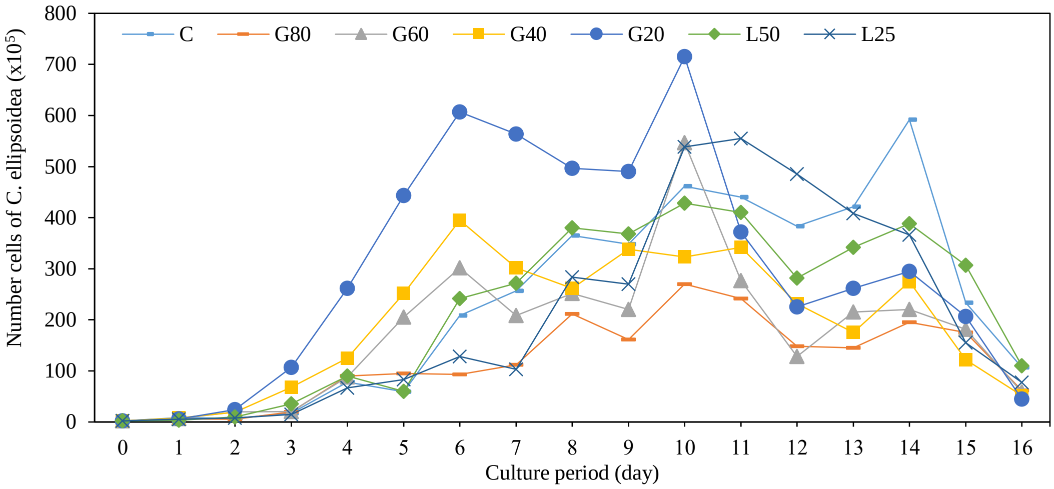

Figure 3. Growth rates of algae under different treatment levels

3.2 Water quality of *C. ellipsoidea* cultivation

Glucose level also influenced water quality during cultivation. The G20 group maintained alkalinity levels between 130-187 mg/L, suitable for *Moina* sp. feed, especially during maximum algal growth from days 6-10. In contrast, other treatment groups exhibited alkalinity levels exceeding 200 mg/L (Table 1). All treatment groups showed pH fluctuations within the range of 7.2-8.8 throughout the 15-day cultivation period (Table 2), which is safe for *Moina* sp. cultivation.

Total ammonia levels gradually increased over time in all groups but remained below 1 mg/L for the first 9 days before rising further, without significant differences between the treatment's groups (Figure 4).

Table 1. Alkalinity (mg/L as $CaCO_3$) during 15-day *C. ellipsoidea* cultivation with different glucose and lime levels

Treatment/day	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
C	153	221	221^{a}	221^{a}	221^{a}	221^{a}	221^{a}	221	221	221	221	221	221	210	221^{a}
G80	142	221	221^{a}	221^{a}	221^{a}	221^{a}	221^{a}	221	221	221	221	221	221	221	221^{a}
G60	164	221	221^{a}	221^{a}	221^{a}	221^{a}	221^{a}	221	221	221	221	221	221	215	221^{a}
G40	130	215	221^{a}	221^{a}	221^{a}	221^{a}	221^{a}	221	221	221	221	221	221	221	221^{a}
G20	102	193	130.33^{b}	147.33^{b}	187^{b}	153^{b}	153^{b}	210	198	198	210	221	221	210	175.67^{ab}
L50	153	221	221^{a}	221^{a}	221^{a}	221^{a}	221^{a}	221	221	221	221	204	210	210	187^{ab}
L25	147	221	221^{a}	221^{a}	221^{a}	221^{a}	221^{a}	221	221	221	221	221	198	176	148^{b}

Means within rows with different letters indicate significant differences ($p<0.05$) between treatment groups.

Table 2. pH values during 15-day *C. ellipsoidea* cultivation with different glucose and lime levels

Treatment/day	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
C	7.3	8.5	8.1	8.5	8.6	8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.3	8.5	8.2
G80	7.7	8.3	7.8	8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.2
G60	7.2	8.7	8.2	8.5	8.6	8.5	8.8	8.8	8.3	8.5	8.5	8.3	8.5	8.5	8.3
G40	7.7	8.3	8.2	8.5	8.6	8.5	8.5	8.5	8.3	8.5	8.5	8.5	8.5	8.5	8.5
G20	8.0	8.2	8.0	8.0	8.5	8.3	8.0	8.2	8.3	8.2	8.3	8.5	8.5	8.3	8.2
L50	7.8	8.7	8.0	8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.3	8.3	8.5	8.2
L25	7.3	8.5	8.5	8.5	8.7	8.5	8.5	8.5	8.3	8.3	8.3	8.0	8.2	8.0	7.8

No significant differences ($p>0.05$) in mean pH values were observed between treatment groups on any given day.

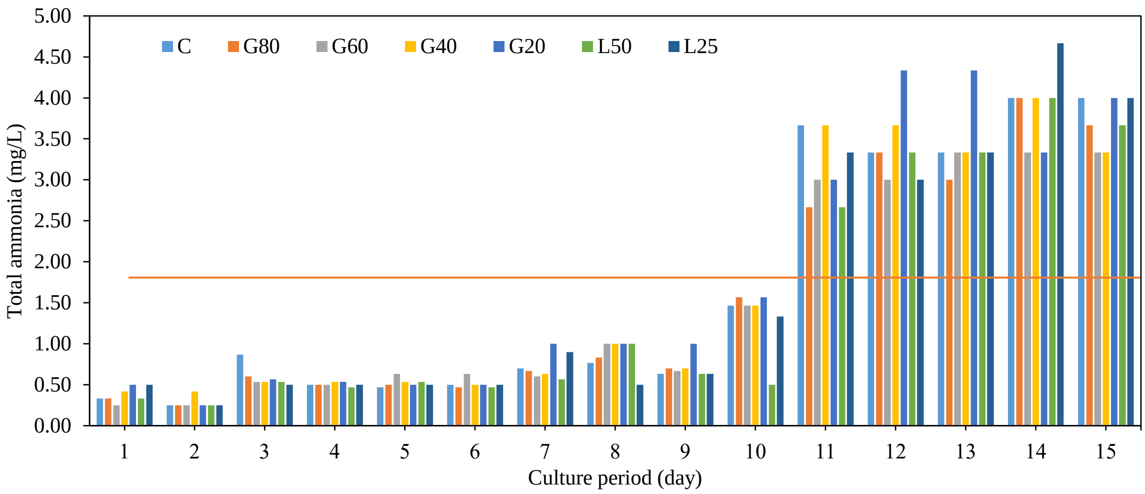

Figure 4. Changes in ammonia nitrogen levels during 15-day *C. ellipsoidea* cultivation under different media formulations

4. DISCUSSION

The study demonstrated that glucose level in the culture medium plays a crucial role in promoting the growth of *C. ellipsoidea*. Reducing the glucose level to 80% (0.1 g/L) led to the highest cell yield during both the lag and log phases of cultivation. According to literature reviews, glucose has been used as a component in Chlorella culture media, with optimal concentrations typically ranging from 0.5-2.5 g/L, depending on species and cultivation objectives (Heredia-Arroyo et al., 2011; Yeh and Chang, 2012; Kong et al., 2013). However, this study demonstrates that glucose concentration can be reduced while maintaining the growth rate of *C. ellipsoidea*. This effect may be attributed to the algae's ability to enhance carbon utilization efficiency through the carbon concentrating mechanism (CCM) under low carbon conditions, as described by Raven et al. (2017). The CCM process increases carbon dioxide uptake efficiency, which is crucial for microalgae to convert CO_2 into biomass and bioproducts (Chew et al., 2017; Kong et al., 2024). Additionally, lowering the glucose level helped maintain suitable water quality, particularly alkalinity, which is critical for controlling pH within an appropriate range for both algal growth and subsequent use as *Moina* sp. feed.

Although this study used a closed culture system, which tends to exacerbate alkalinity issues due to compound accumulation (Wang et al., 2014), the G20 group maintained alkalinity levels between 130-187 mg/L. This range aligns with the optimal alkalinity for aquatic organisms in aquaculture, typically 60-150 mg/L as $CaCO_3$ (Wurts and Durborow, 1992). The findings correspond with Ramaraj et al. (2015), who recommended an alkalinity range of 75-225 mg/L for *Chlorella* sp. cultivation. However, crustaceans may require higher alkalinity levels for body and resting eggs (ephippia) composition formation, molting processes, and osmoregulation (Stevenson, 1985; Shapiera et al., 2011). Nevertheless, excessive alkalinity levels (above 420 mg/L as $CaCO_3$) can adversely affect for *Moina* sp. (Bogart et al., 2016). Higher glucose levels led to a rapid increase in alkalinity, a common issue in mixotrophic cultivation systems using both organic carbon and light (Wang et al., 2014). Maintaining optimal alkalinity is crucial for ensuring high biomass quality, as demonstrated for *Artemia* sp. cultivation in salt ponds (Anh et al., 2009). No significant differences in pH were observed between the treatment groups, with values ranging from 7.2-8.8 throughout the experiment, which are appropriate for both algal cultivation and subsequent use as *Moina* sp. feed (Dhert, 1996). Therefore, pH control may not be a primary concern in developing this algal production system.

Juneja et al. (2013) proposed an optimal pH range of 6.5-9.0 for *C. ellipsoidea* cultivation, indicating that the observed pH levels were acceptable for algal growth. Achieving an optimal pH range depends on various factors such as algal species, carbon and nitrogen sources, light intensity, and temperature. Ammonia levels, another critical factor affecting *Moina* sp. cultivation, remained below 1 mg/L for the first 9 days before increasing later, which resulted from an inverse relationship with *C. ellipsoidea* growth rate. Specifically, during days 1-9, *C. ellipsoidea* utilized ammonia for growth until reaching the stationary phase. When nutrients were depleted, *C. ellipsoidea* entered the death phase. As

C. ellipsoidea mortality increased in the system, ammonia levels consequently rose after day 10 of cultivation. However, no significant differences were found between treatment groups, possibly due to the urea concentration in the culture medium at 0.6 g/L being at an appropriate level for efficient biomass production without affecting ammonia accumulation in *C. ellipsoidea* cultivation. Studies have shown that when *C. ellipsoidea* use urea as a nitrogen source for efficient biomass production, ammonia levels can be effectively controlled (Yu et al., 2023).

Additionally, the balance between nitrogen release and uptake is crucial to prevent toxic ammonia buildup, which is essential for the growth of *Chlorella* sp. (Anyanwu et al., 2022). The observed ammonia levels are considered safe for *Moina* sp. cultivation during the first 9 days, as levels between 5-10 mg/L can adversely affect *Moina* sp. growth and survival (Collos and Harrison, 2014). In large-scale or mass culture outdoor production, agricultural fertilizers such as urea or ammonium sulfate are commonly used (Liao, 1983) to reduce production costs compared to laboratory-scale culture media. While urea is effective for ammonia control, residual ammonia in large-scale systems with high pH (due to dense algal populations, especially during daytime) (Tucker and D'Abramo, 2008) may cause toxicity from unionized ammonia (NH_3) to *Moina* sp. Therefore, large-scale cultivation systems should implement regular water exchange or fresh water addition to reduce ammonia toxicity and organic matter, or utilize aeration systems to convert nitrogen into safer forms. Additionally, aeration can enhance water flea production by improving fecundity and size (Ovie and Ovie, 2004).

The adjustment of lime, serving as a source of calcium and alkalinity, also impacted algal growth. Using 50% of the standard lime level (L50 group) provided comparable cell yields to the G20 group while maintaining better alkalinity control compared to the standard medium formulation. This differs from González-Garcinuño et al. (2014), who reported that calcium levels significantly influence growth rates and lipid accumulation in several Chlorophyta species, including *C. ellipsoidea*. Although a clear relationship between lime levels and algal growth was not observed in this study, calcium remains necessary for maintaining water quality balance in the culture system. Further investigations into the specific roles of calcium, particularly in different algal strains with varying calcium requirements, may be warranted.

In summary, reducing the glucose and lime levels from the standard culture medium formulation for *C. ellipsoidea* cultivation in a closed laboratory system can promote algal growth rates comparable to or better than the standard formulation while maintaining water quality parameters at suitable levels for subsequent use as *Moina* sp. feed. This approach may contribute to developing an efficient culture medium for producing Chlorella algae as a live feed for aquatic animal cultivation, while also reducing the cost of raw materials. Further studies in commercial-scale production systems and with various culture media formulations should be conducted to validate and optimize this approach for practical applications.

5. CONCLUSION

This study demonstrated that reducing the glucose level in the culture medium had a more pronounced effect on promoting the growth of *C. ellipsoidea* compared to reducing the lime level. The G20 group exhibited the highest growth rates and maximum cell density of 7.15×10^7 cells/mL on day 10. The 0.1 g/L glucose level maintained alkalinity within the optimal range of 130-187 mg/L, suitable for *Moina* sp. feed. Lime levels did not significantly affect alkalinity, as both the L50 and L25 groups exhibited alkalinity levels exceeding 200 mg/L. No significant differences in pH and ammonia levels were observed among the groups. Reducing glucose to 80% proved most effective for growth while maintaining suitable water quality. Further investigations in commercial-scale production systems are needed to validate these findings for practical applications.

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DECLARATION OF COMPETING INTEREST

The authors declare no conflict of interest.

AUTHORE CONTRIBUTION

Conceptualization, R.Y. and N.K.; Methodology, N.K. and P.S.; Formal Analysis, R.Y. and N.K.; Investigation, N.K. and R.Y.; Resources, P.S.; Data Curation, N.K.; Writing - Original Draft Preparation, R.Y. and N.K.; Writing - Review and Editing, R.Y., P.S. and N.K.; Visualization, N.K.; Supervision, N.K.; Project Administration, N.K.

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Nutrient and Coliform Levels in the Surface Waters of a Protected Upland Lake in Ormoc City Philippines

Eugie V. Cinco, Cherryhel B. Toralde, Olga G. Corales-Ultra, and Rolly G. Fuentes*

Division of Natural Sciences and Mathematics, University of the Philippines Tacloban College, Tacloban City 6500, Philippines

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*** Corresponding author:**
E-mail: rgfuentes@up.edu.ph

ABSTRACT

Lake Danao in Ormoc City, Philippines, is a legally protected freshwater ecosystem classified as a Class A water body by the government. However, its ecosystem services and biological stability are continuously threatened by untreated runoff from human activities in the upland regions surrounding the lake. A study was thus conducted to evaluate the water quality of Lake Danao using water quality indices and microbiological assessments. Water samples were collected in five sampling stations from April 2021 to June 2022. Results ranged between 0 and 20.53 mg/L for nitrates, 0 and 1.06 mg/L for nitrites, 0.07 and 2.5 mg/L for phosphates, 2.86 and 7.73 mg/L for dissolved oxygen, and 2.83 and 1,600 MPN/100 mL for total coliform. On average, the readings of these parameters were above the Philippines' Department of Environment and Natural Resources maximum allowable limits for Class A waters. In addition, fecal coliform (0-849.67 MPN/100 mL, $\bar{x}$=40.17 MPN/100 mL) consistently exceeded the limit. Mean readings in fecal coliform and phosphate levels were drawing near Class B levels. The results may impose possible threats to human health, as high levels of coliform suggest that the lake is already an unsafe source of potable water. In addition, fluctuations across sampling periods were observed for all parameters measured, except for total and fecal coliforms. Lastly, heavy precipitation resulted in a very low N/P ratio (<1) which suggests that the possible source of the phosphate is anthropogenic, and runoff from the surrounding land has carried significant concentrations of phosphates and coliform into the water body. The nutrient pollution index shows that Lake Danao is "moderately polluted," while its trophic state index states that it is "mesotrophic." It is recommended that strategies in runoff treatment should be advanced, be it by nature-based solutions, such as ensuring thick vegetation cover along the buffer zone, or via man-made interventions like no-till farming. Regular monitoring of the lake water quality should therefore be continued, particularly for nutrients and coliform, to ensure the maintenance, protection, and responsible use of the protected upland lake.

1. INTRODUCTION

Lakes are one of the most important sources of fresh water, though they account for only about 0.3% of the total surface waterbody sources. Lake waters are used by humans for drinking, aquaculture, and recreational activities (Vasistha and Ganguly, 2020). Unfortunately, the conditions of lakes and other water bodies have been in constant deterioration due to increased anthropogenic activities, such as agriculture, industry, and urbanization. Water contamination and scarcity are the utmost concerns because they pose threats to public health and food security, impair ecosystem services, and hinder economic growth (UNESCO, 2021).

Eutrophication happens when excess nutrients like nitrates and phosphates cause too much plant and algae growth, reducing dissolved oxygen (DO) and harming aquatic life (Schindler, 2006; Isiuku and

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Enyoh, 2020). A major source of these nutrients is agricultural runoff from fertilizers and livestock waste (USEPA, 2002). A study of Al-Afify et al. (2023) on Burullus Lake in Egypt showed severe eutrophication and extreme environmental stress due to runoffs from several drains containing agricultural, industrial, and domestic effluents. The lake's nutrient loading behavior was greatly reduced due to the presence of high traces of nitrates in the form of ammonia (1,221 µg/L) exceeding Canadian guidelines and orthophosphates (436 µg/L) that were approximately twice as high as they were during the summer season. The high levels of nutrient contaminants in the lake affected its primary productivity and deteriorated the water quality. To improve water quality, the locals surrounding the lake have shifted to various programs and activities, such as improved farming methods, dredging and deepening the inlet link, and investigating several drainage water management options.

Total coliform (TC) and fecal coliform (FC) counts are indicators of pollution as well as the water supply's sanitary condition. TC includes bacteria that are usually found in water and soil that have been contaminated by human or animal waste. These bacteria are less likely to cause illness, but their presence, especially in large numbers, is an indication that the water supply may be susceptible to contamination by pathogens. On the other hand, FC bacteria are gram-negative, non-spore-forming rods that are found in the intestines and feces of humans and other warm-blooded animals. Thus, the presence of FC in water sources may indicate contamination of the waterbody by human and/or animal feces. Most strains of FC bacteria are pathogenic, with *Escherichia coli* as the most predominant (USEPA, 1995). Niyoyitungiye et al. (2020) assessed the presence of coliform bacteria in Lake Tanganyika in Burundi, as its water is used by nearby residents for cooking, drinking, and washing. Results showed that FC is low, but TC is high; thus, to be considered safe for drinking and bathing purposes, the lake water requires treatment prior to use.

Different physicochemical parameters were used by García-Avila et al. (2023) to assess a high Andean lake's trophic state and water quality. Results from several eutrophication and trophic state indices from nutrient concentrations, dissolved oxygen levels, and biological productivity indicated that the lake had a high level of eutrophication attributed to excessive accumulation of nutrients in the water. The results showed that the lake is in a hypereutrophic state, indicating a high concentration of nitrates, which ranges from 1 to 5 mg/L in the summer season with concentrations between 0.8 and 2.7 mg/L in winter, and phosphates of 2.89 mg/L and 1.84 mg/L in summer and winter, respectively. Anthropogenic causes were identified as the main sources of eutrophication, including tourism and agriculture, suggesting means be considered to mitigate human activity in the area and control pollution in the lake.

Lake Danao in Ormoc City, Leyte, Philippines, is a protected upland area with a total surface area of 139.83 hectares (de la Cruz et al., 2024). The lake was classified as Class A freshwater according to the Philippine Water Quality Guidelines, which designate it as a water source suitable for public water supply, requiring conventional treatment to meet drinking water standards (DENR, 2016). At present, it is primarily used for recreation and tourism, but locals also rely on it for food (fish, mussels, and prawns) and agricultural water (Romero et al., 2023).

This study was conducted to assess the current water quality of Lake Danao, an under-researched upland lake in the region, using water quality indices and microbiological assessment, which focused on addressing the critical intersection of nutrient enrichment and microbial contamination. Its first spatiotemporal assessment of both nutrient concentrations and coliform levels provides a clearer view of the lake's ecological health, particularly in relation to human impacts such as settlements and agriculture. Specifically, nutrients (phosphates, nitrates, and nitrites) and DO were measured *in situ*, while the levels of TC and FC bacteria were estimated using the Multiple-Tube Fermentation Technique. The data was analyzed to check for variations between sampling stations and sampling periods using statistical methods. Then, nutrient pollution and trophic state indices were calculated and used to categorize the status of the lake waters.

2. METHODOLOGY

2.1 Study site

The study was conducted in Lake Danao Natural Park (LDNP) in Ormoc City, Leyte, located in the eastern central region of the Philippines. The lake is classified as Class A, which means that the lake waters can be a source of water supply after completing treatment required by the Philippine National Standards for Drinking Water (PNDSW). During the sampling period, the temperature of the area ranged from 27.3-29.4°C and the precipitation ranged from 150.8-558.6 mm (PAGASA, 2022). The whole area of the lake and sampling stations are shown in Figure 1, the description of each sampling station is listed in Table 1, and the monthly precipitation data of the sampling periods are in Table 2.

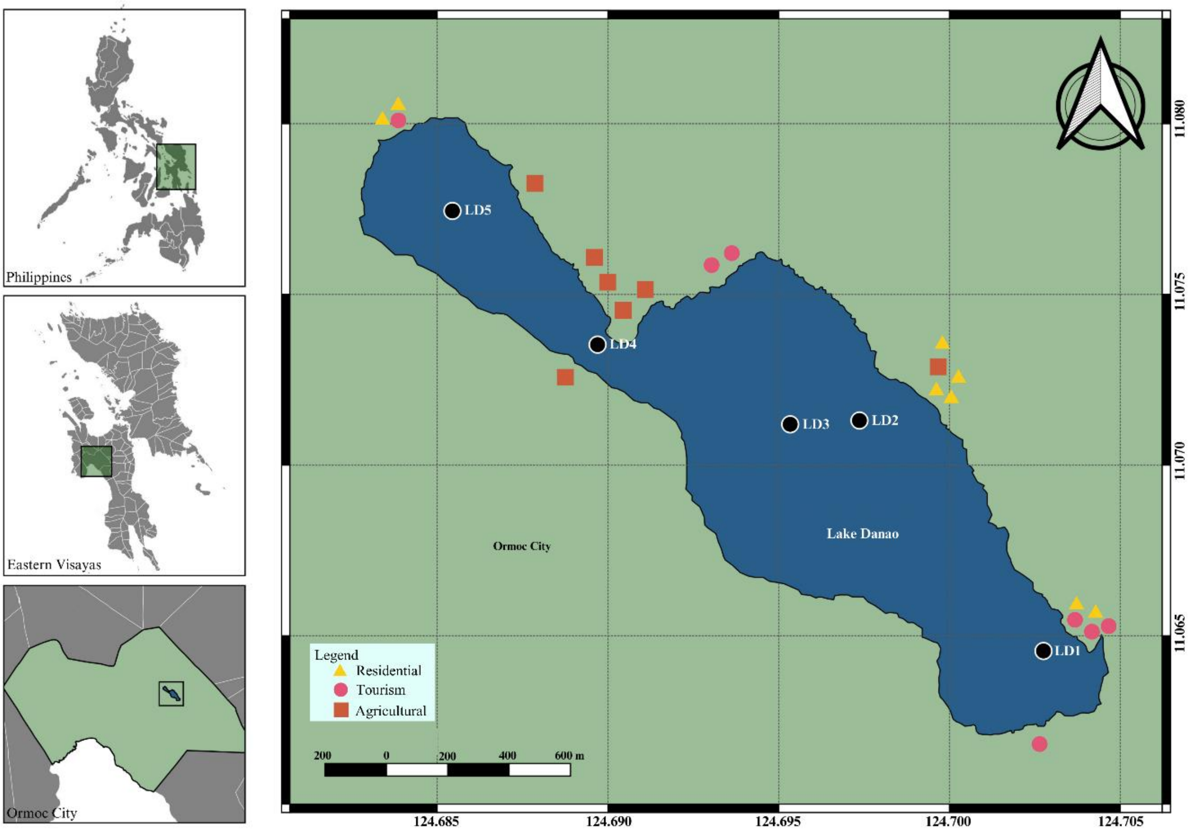

Figure 1. Map of Lake Danao, Ormoc City showing the five sampling stations (LD1-LD5).

Table 1. Site description and geographical coordinates of Lake Danao sampling stations

Stations	Mean depth (m)	Geographical coordinates	Characteristics
LD1	33.35	N 11°03'52.4" E 124°42'09.0"	*Inawasan area* (where water flows out of the lake), recreational establishments and residential areas are concentrated.
LD2	69.47	N 11°04'16.7" E 124°41'50.5"	Below the primary school and community, patches of agricultural land were observed nearby.
LD3	71.57	N 11°04'16.3" E 124°41'43.2"	Least disturbed by humans.
LD4	18.81	N 11°04'24.7" E 124°41'22.9"	Most agricultural areas are located nearby.
LD5	23.20	N 11°04'38.8" E 124°41'07.6"	Newly opened recreational cottages, and near some agricultural land.

Table 2. Average precipitation in Lake Danao from April 2021-June 2022.

Sampling periods	Months	Average precipitation (mm)[a]
SP1	Apr-Jun 2021	204.40
SP2	Jul-Sep 2021	190.27
SP3	Jan-Mar 2022	340.83
SP4	Apr-Jun 2022	330.27

[a]PAGASA (2022)

2.2 Sample collection and in situ physicochemical parameter measurements

The method on the collection of water samples was adapted from the Environmental Management Bureau Ambient Water Quality Monitoring Manual Volume 1 (DENR-EMB, 2008). Bimonthly collection (every 2nd and 4th week of the month) of water samples in triplicate was done from April 2021 to June 2022, in the morning between 9 am and noon. No sampling was conducted in the months of Sep to Dec 2021 due to logistical concerns and Typhoon Odette. Pre-sterilized and acid-washed 150 mL glass bottles were used to collect water samples in the five sampling stations in triplicate, both for nutrient and coliform analyses.

DO levels were measured in situ using a Hanna portable multiparameter (HI7698194). Meanwhile, the nutrient content of the water samples was analyzed using nitrate (Hanna HI977288), nitrite (Hanna HI97707), and phosphate (Hanna HI97713) photometers. The protocol suggested by the manufacturers of the photometers and the multi-parameter was used in the analysis. Distilled water was used as the negative control. Water transparency was measured in terms of Secchi disk depth.

2.3 Total coliform and fecal coliform analysis

The multiple-tube fermentation technique was used to estimate total coliform (TC) and fecal coliform (FC) levels using a '333' tube series with lactose broth. This three-step method gives results as the Most Probable Number (MPN), following the American Public Health Association's guidelines (APHA, 1998). It includes three phases: a presumptive test using lactose tryptose broth, a confirmed test with brilliant green bile broth for TC, and a completed test using *E. coli* broth for FC. Peptone water was used as a diluent to support bacterial growth for easier detection.

2.4 Data analysis

The Kruskal-Wallis nonparametric test followed by the Mann-Whitney test were conducted to test significant differences ($p<0.05$) of some water parameters (nutrients, coliform levels, and DO) at sampling stations during select sampling periods. The nitrate-to-phosphate ratio (N/P) was calculated as an indicator of the extent of algal or phytoplankton production in water bodies. N/P is usually assumed to be equal to the Redfield ratio of 16N:1P; however, deviations to this value have been observed depending on both phytoplankton species and study sites. A higher ratio indicates that P is limiting, and a lower ratio means that N is limiting (Spalinger and Bouwens, 2003; Isiuku and Enyoh, 2020).

Nutrient pollution index (NPI) was also computed using Eq. 1., where MATN/P is the maximum allowable value based on DENR (2016) guidelines. The following are the classifications based on NPI values: < 1-no pollution; 1 ≤ 3-moderately polluted; > 3 ≤ 6-considerably polluted; and > 6-very highly polluted (Isiuku and Enyoh, 2020).

$$\mathrm{NPI} = \frac{\mathrm{TP}}{\mathrm{MATN}} + \frac{\mathrm{TP}}{\mathrm{MATP}} \tag{1}$$

To determine the lake's trophic state classification as the basis of primary productivity, a numerical trophic state index (TSI) model developed by Carlson (1977) was used. Carlson's TSI was calculated using Secchi disk transparency (SDT), total phosphorus (TP), total nitrogen (TN), and existing mathematical model computations (Equation 2-4). Mean values from the three variables were taken to determine the overall trophic state classification.

$$\mathrm{TSI\ (SDT)} = 10\left[6 - \left(\frac{\ln \mathrm{SDT}}{\ln 2}\right)\right] \tag{2}$$

$$\mathrm{TSI\ (TP)} = 10\left[6 - \left(\frac{\ln \frac{80.32}{\mathrm{TP}}}{\ln 2}\right)\right] \tag{3}$$

$$\mathrm{TSI\ (TN)} = 54.45 + 14.43\ (\ln \mathrm{TN}) \tag{4}$$

3. RESULTS

3.1 Spatial variation of water quality parameters

Figure 2 shows that nutrients and DO levels did not vary among the five sampling stations in Lake Danao. However, significant variations were observed between sampling stations in terms of TC and FC. The estimated average values for TC counts ranged from 2.83 to >1,600 MPN/100 mL, with some sampling stations showing values above the Class A standard water limit (1,000 MPN/100 mL) set by the DENR (2016). LD4 and LD5 showed high average TC levels, 431.00 MPN/100 mL and 523.12 MPN/100 mL, respectively, as compared to the other stations. For the FC counts, all measured values in the five sampling stations are above the maximum permissible limit (<1.1 MPN/100 mL) for Class A water. LD3 had the least mean FC estimate of 4.78 MPN/100 mL, significantly lower than the other four sampling stations.

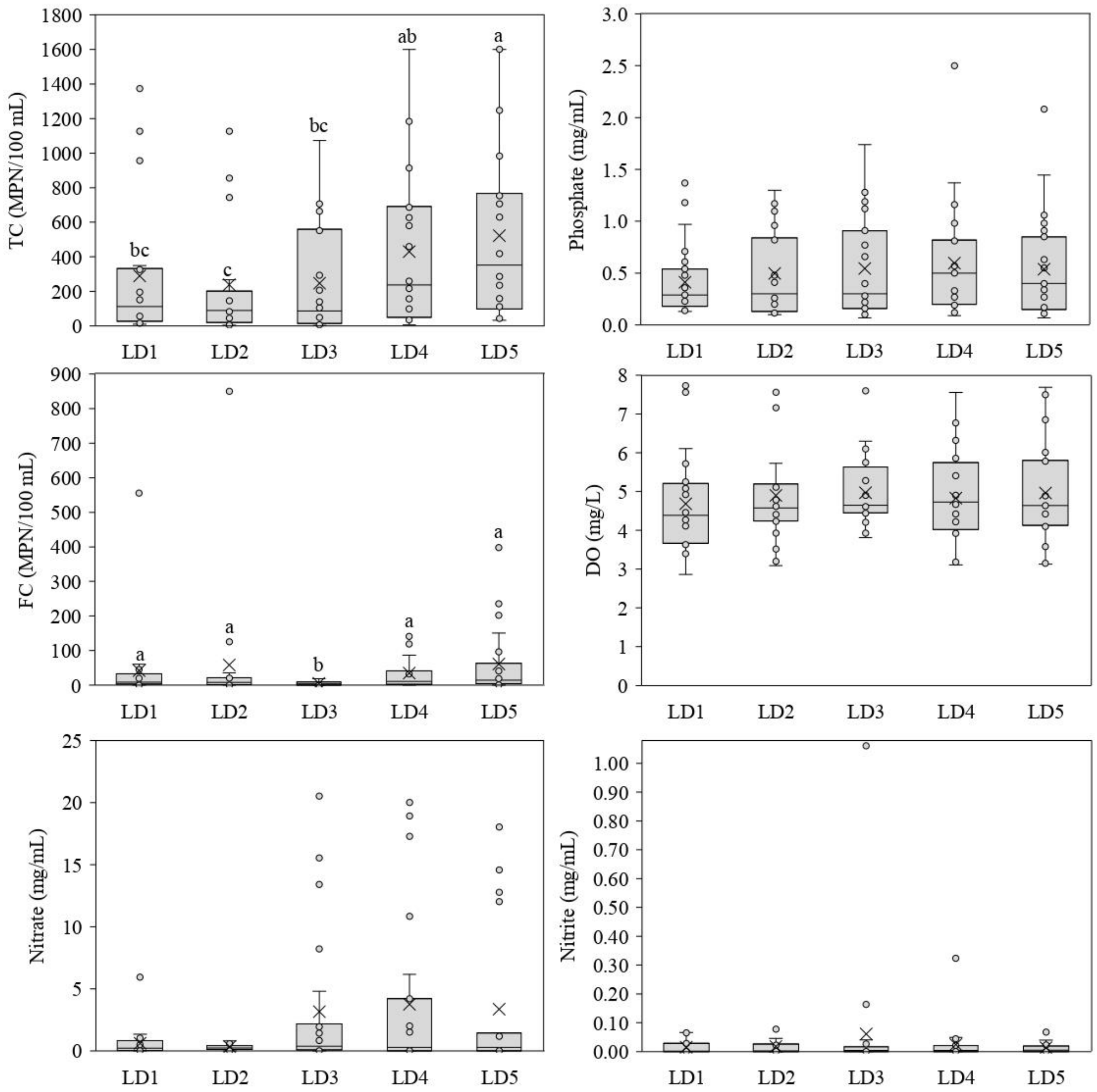

Figure 2. Box plot of coliform levels (TC: total coliform, FC: fecal coliform), nutrients (phosphate, nitrate, and nitrite), and dissolved oxygen (DO) based on sampling stations in Lake Danao, Ormoc City (n=23). Groups with different letters are significantly different ($p<0.05$) by Kruskal-Wallis and Mann-Whitney tests. LD-Sampling stations.

Figure 3(a) shows the calculated N/P values for each sampling station, with the mean values for LD3, LD4, and LD5 close to the Redfield ratio of 16N:1P. In LD1 and LD2, the N/P values were <<16, which means that N is limited in these locations.

3.2. Temporal variation of water quality parameters

There are no pronounced seasons in the Eastern Visayas Region. The sampling periods were divided based on the monthly precipitation data (Table 2). SP3 and SP4 recorded higher amounts of precipitation as compared to SP1 and SP2, a factor that may have significantly contributed to the amount of nutrients, DO, and coliform detected in the lake waters. Significant variations in water quality parameters were observed in terms of the sampling period, as shown in Figure 4.

High TC levels were observed in SP4 (17,632.7 MPN/100 mL), while SP1 and SP2 recorded the lowest levels (4,536.60 MPN/100 mL and 5,924.47 MPN/100 mL, respectively). Based on the guidelines of the DENR (2016), the waters of Lake Danao were contaminated with FC in all sampling periods. The highest FC levels (2,344.8 MPN/100 mL) were recorded in SP4, while SP2 had the lowest (94.4 MPN/100 mL).

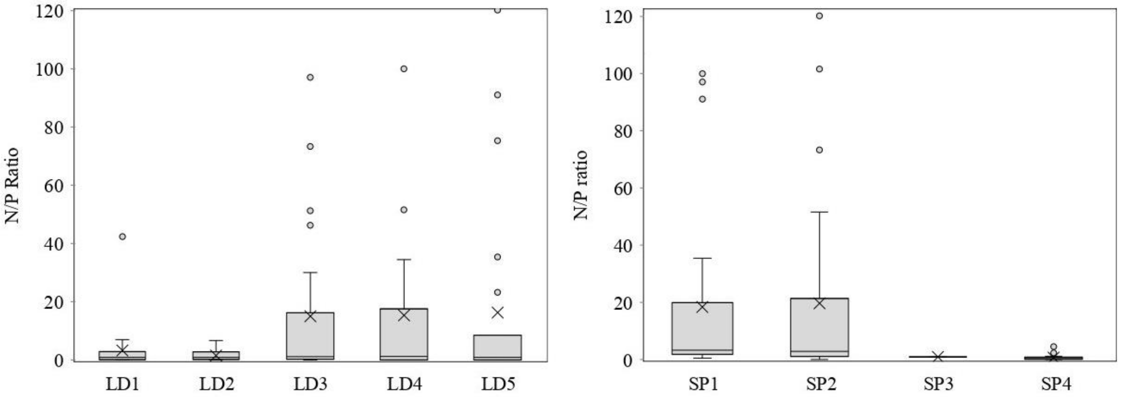

Figure 3. Nitrate to phosphate ratio (mean±stdev) of Lake Danao, Ormoc City based on (left) sampling stations (n=23) and (right) sampling periods (SP1: Apr-Jun 2021, SP2: Jul-Sep 2021, SP3: Jan-Mar 2022, SP4: Apr-Jun 2022) in Lake Danao, Ormoc City (n=30). N/R 0-16: nitrate is limiting; N/R above 16: phosphate is limiting (Spalinger and Bouwens, 2003; Isiuku and Enyoh, 2020). LD-Sampling stations. SP-Sampling periods.

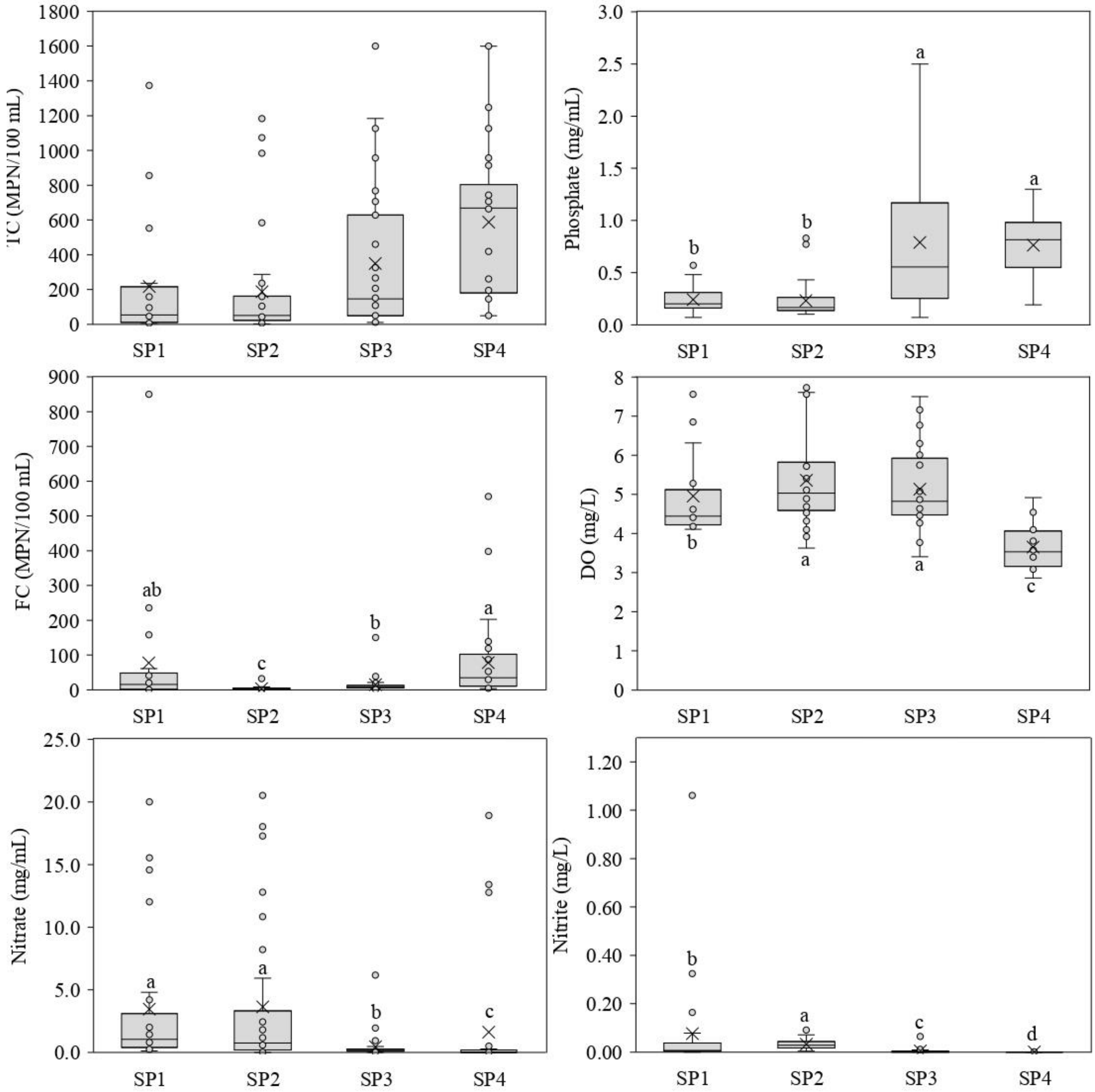

Figure 4. Box plot of coliform levels (TC: total coliform, FC: fecal coliform), nutrients (phosphate, nitrate, and nitrite), and dissolved oxygen (DO) based on sampling periods (SP1: Apr-Jun 2021, SP2: Jul-Sep 2021, SP3: Jan-Mar 2022, SP4: Apr-Jun 2022) in Lake Danao, Ormoc City (n=25). Groups with different letters are significantly different ($p<0.05$) by Kruskal-Wallis and Mann-Whitney tests. SP-Sampling periods.

For nitrates, high levels (3.33 mg/L) were measured in SP1 and SP2, then decreased to 0.44 mg/L and 1.61 mg/L in SP3 and SP4, respectively. Similarly for nitrites, high levels (0.052 mg/L) in SP1 and SP2 were observed, followed by significantly lower levels (0.003 mg/L) in SP3 and SP4. An opposite trend was observed for phosphates. Phosphates were below the maximum allowable limit set by DENR Water Quality Standard for Class A and B waters during SP1 and SP2 (0.24 mg/L) but increased to above the allowable limit in SP3 and SP4 (0.80 mg/L). The maximum allowable limits for Class A waters for nitrate, nitrite, and phosphate are 7 mg/L, 1 mg/L, and 0.5 mg/L, respectively.

The observations on the fluctuation in nutrient levels are consistent with the computed N/P in Figure 4(b). In SP1 and SP2, the N/P values are relatively close to the observed Redfield ratio, but in SP3 and SP4, the N/P values were <1 due to the very high amounts of P available, with limited N. The increasing amount of phosphate also affected the DO levels (Figure 4), with SP4 having significantly lower values (3.6 mg/mL) than the other three earlier sampling periods.

3.3 Nutrient pollution index and trophic state index

NPI considers the effects of nitrate and phosphate on the overall quality of Lake Danao. Figure 5(a) shows the NPI values for the five sampling stations with LD1 classified as "not polluted" and the rest of the other stations as "moderately polluted". Spatial variation in TSI for Lake Danao using three limnological parameters (SDT, TP, and TN) shows significant variation for TN only (Figure 6(a)). Primary productivity in terms of SDT was observed to fluctuate within the oligotrophic range. In terms of TN inputs, TSI was fluctuating from oligotrophic to eutrophic state, while all locations are eutrophic in terms of TP. The average primary productivity for Lake Danao is "mesotrophic" (48.55±1.33 TSI) for LD1 and LD2, which is attributed to moderate nutrient supply as well as dissolved and suspended solid inputs affecting the water clarity. On the other hand, LD3, LD4, and LD5 are "eutrophic" (58.02±1.07 TSI) or have a good supply of nutrients resulting in enhanced production and reduced transparency.

Figure 5(b) shows the calculated NPI values for each sampling period. Lake Danao is "not polluted" when precipitation is low but becomes "moderately polluted" when precipitation is high. TSI calculated for Lake Danao using the same limnological parameters shows monthly variations (Figure 6(b)). Primary productivity in terms of SDT was observed fluctuating within the oligotrophic range, while in terms of TN inputs, TSI fluctuated from ultraoligotrophic to mesotrophic state. Measured high phosphate concentration in the lake water corresponds to increasing trophic state trends and primary productivity. TSI in terms of TP was observed to increase from eutrophic to hypereutrophic. Generally, the lake reservoir's primary productivity can be classified as "mesotrophic" (48.14±2.55 TSI).

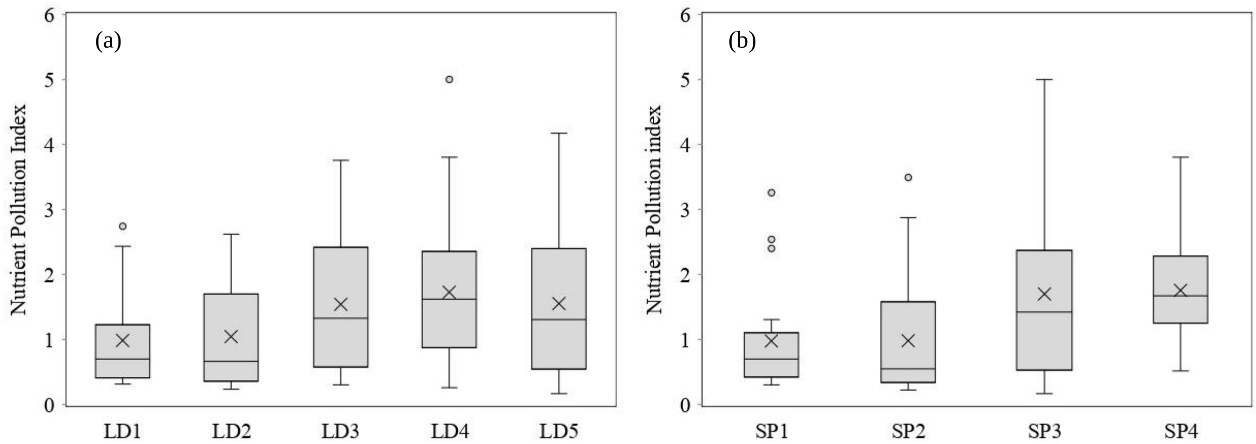

Figure 5. Box plot of nutrient pollution index (mean±stdev) based on (left) sampling stations (n=23) and (right) sampling periods (SP1: Apr-Jun 2021, SP2: Jul-Sep 2021, SP3: Jan-Mar 2022, SP4: Apr-Jun 2022) in Lake Danao, Ormoc City (n=30). NPI values: <1-no pollution; 1≤3-moderately polluted; >3≤6-considerably polluted; and >6-very highly polluted (Isiuku and Enyoh, 2020). LD-Sampling stations. SP-Sampling periods.

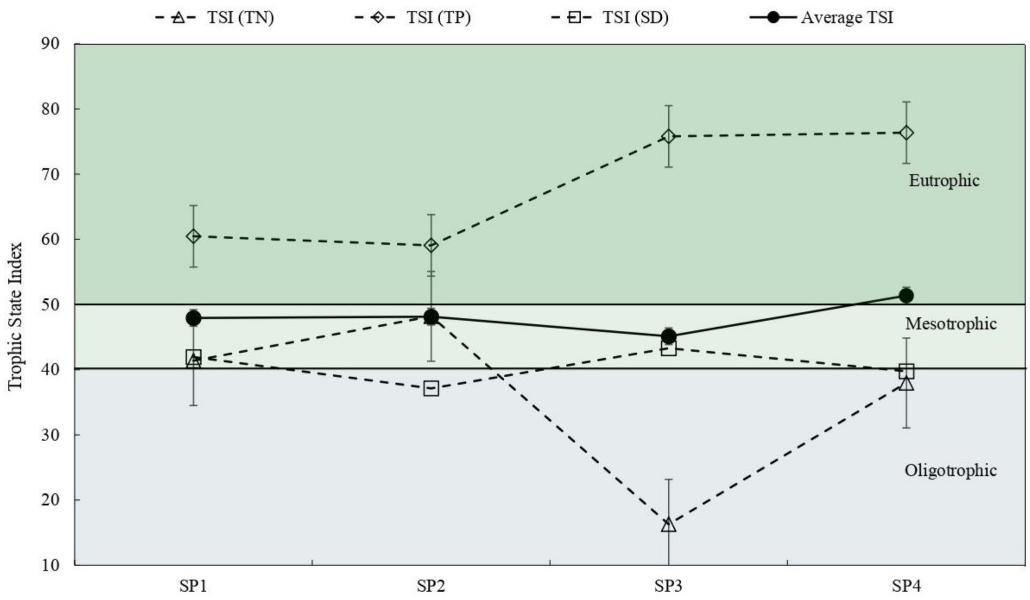

Figure 6. Trophic State Index (mean±stdev) fluctuations using three limnological parameters (TN: Total nitrogen, TP: Total phosphorus, SD: Secchi disk depth) in Lake Danao, Ormoc City (A) sampling stations (n=12) and (B) sampling periods (SP1: Apr-Jun 2021, SP2: Jul-Sep 2021, SP3: Jan-Mar 2022, SP4: Apr-Jun 2022) in Lake Danao, Ormoc City (n=30). SP-Sampling periods.

4. DISCUSSION

This study reports the spatial and temporal variations of nutrients and coliforms in the waters of Lake Danao, Ormoc City (Philippines), as well as an assessment of the lake's water quality and trophic status based on NPI and TSI values.

Data analysis showed that the water quality parameters measured in different areas of Lake Danao were not significantly different from each other, except for TC and FC. The calculated NPI values of the five sampling stations indicated that the waters in LD1 are "not polluted" but the other stations are "moderately polluted." Then based on the TSI, LD1 and LD2 are "mesotrophic" while LD3, LD4, and LD5 are "eutrophic." Even though tourism activities are abundant in LD1 and LD2, these locations are also near the stream outlet or Inawasan area, where nutrients can be flushed out of the lake quickly (Shaw et al., 2004). LD4 and LD5, on the other hand, are near agricultural lands.

On the other hand, the variability in terms of sampling periods is significant for all parameters, indicating that weather conditions, such as the amount of precipitation, are the main factors affecting the water quality of Lake Danao. The high TC and FC values measured in Lake Danao waters relative to Class A minimum standards are already an indication that the water is unsafe for direct consumption as pathogens may be present in the water. According to Bera (2022), TC and FC bacteria in freshwater ecosystems can drastically increase due to high levels of tourist disturbance coupled with the tourism industry, sewage, and household discharge from nearby residential areas, runoff, and local activities. TC estimates were found to be fluctuating and unpredictable across the five sampling sites since most coliforms are ubiquitous in nature and are cosmopolitan in habitat (Olstadt et al., 2007). Data from stations LD4 and LD5, where farming is common, indicates that runoff from these areas is the main cause of high TC levels, sometimes exceeding the DENR's maximum limit. For LD1, LD2, and LD3, these stations are far from the contamination source, and the bacteria are subjected to sedimentation, predation, dilution, and bacterial death before reaching these stations (Dan and Stone, 1991). In LD1 and LD2, higher water flow towards the stream outlet is likely to reduce TC levels, while minimal human activity at LD3 allows sedimentation to lower TC. However, FC levels were above the limit at all stations. The presence of fecal coliform in concentrations above the tolerable threshold in the lake water constitutes the presence of sewage contamination, which serves as an indicator that a potential health risk exists for individuals exposed to this water. Their presence is associated with pathogens that may cause severe illness, including typhoid fever, viral and bacterial gastroenteritis, and hepatitis when ingested. These bacteria have occurred in the ambient lake water because of the overflow of observed domestic waste and other nonpoint sources of human and animal waste (Staley, 2012).

When precipitation is low (SP1 and SP2), levels of nitrates, nitrites, and DO were high, while TC, FC, and phosphates were low. This coincides with the study of Liu et al. (2024), where rainfall increases nutrient input, particularly nitrogen, through stormwater runoff, which carries organic matter and pollutants into the reservoir. This nutrient influx promotes algal blooms, which, in turn, lower dissolved oxygen (DO) levels. Short-term rainfall periods lead to spikes in nutrient concentrations, while long-term rainfall dilutes the nutrients but still causes higher overall nutrient loads due to increased water volume entering the system (Liu et al., 2024). In all sampling periods, both nitrate and nitrite levels in Lake Danao were below the maximum allowable limit set by the DENR and USEPA. Common sources of nitrate that contaminate lakes include septic systems, animal feedlots, fertilizers, manure, industrial wastewater, sanitary landfills, and garbage dumps (Cooper and Monroe, 2021). The NPI value calculated during these sampling periods also indicates that Lake Danao is “not polluted,” thus the high levels of DO. The N/P values are also relatively close to the usually observed Redfield ratio.

On the other hand, during the sampling periods with high amounts of precipitation (SP3 and SP4), TC, FC, and phosphates were high, while levels of nitrates, nitrites, and DO were low, resulting in low values of N/P ratio. Low levels of nitrates and nitrites in SP3 and SP4 may be due to dilution effects, while the high levels of phosphates in the same sampling periods can be attributed to runoff from nearby residential areas, farmlands, and other anthropogenic activities (Sarpong et al., 2023). Based on the calculated NPI values, the lake became “moderately polluted” during these sampling periods. Similar results were reported by Isiuku and Enyoh (2020) when they compared the levels of nutrients in several water bodies in Nigeria during dry and wet seasons. High levels of P decrease DO, conditions that accelerate the loss of N through denitrification (Downing and McCauley, 1992; Zhang et al., 2018), which may explain the low levels of nitrates and nitrites detected.

Interactions between a lake’s physicochemical characteristics and primary productivity give a clear picture of the whole ecosystem’s food cycle (Dillon and Rigler, 1974; Dodson et al., 2000). Productivity can be quantified and described in terms of its nutrient, clarity, and, to some extent, chlorophyll content (Lewis, 2011). The study on Lake Tana by Sotomayor et al. (2024) reveals significant physicochemical characteristics, including varied physicochemical parameters and nutrient levels, which influence the lake’s overall productivity. High levels of chlorophyll-a indicate elevated primary productivity, predominantly driven by phytoplankton dynamics. The findings highlight the complex interplay between nutrient availability and physical conditions, which are critical for sustaining the lake’s ecosystem services. On the other hand, Liu et al. (2010) applied correlation and multiple regression analysis to their dataset and concluded that Secchi disk depths, phosphates, and nitrates were linked to TSI.

Changes in N/P ratios alter the trophic structure of water bodies (Zhang et al., 2018) as reflected by the component TSI for TN and TP. According to Downing and McCauley (1992), the value of the N/P ratio reflects the source of nutrients. Oligotrophic lakes have high N/P values since the nutrient sources are natural, and undisturbed ecosystems give more N than P. Mesotrophic and eutrophic lakes, meanwhile, have a low N/P ratio since the P-rich nutrient sources include runoff from surrounding residential, urban, and agricultural areas. In general, when the different parameters measured were considered in the computation of TSI, Lake Danao is classified as mesotrophic (Carlson, 1977), which suggests that the lake has an intermediate level of productivity and moderate levels of nutrients. It has relatively clear water, sufficient oxygen levels to sustain diverse aquatic life, and manageable algal growth. This trophic status is generally considered favorable for maintaining biodiversity and ecological stability (Bhateria and Jain, 2016). However, the rising phosphorus levels observed in the study could push the lake toward a eutrophic state, characterized by excessive nutrient enrichment. This transition can lead to increased algal blooms, reduced water clarity, oxygen depletion (particularly in deeper layers), and negative impacts on fish populations, water quality, and overall aquatic health (Liu et al., 2021).

Fink et al. (2018) reported that in the world’s 100 great lakes, P-stimulated eutrophication is high in developing countries and use of inorganic fertilizers is a major contributor. Fertilizers are a possible source of P in Lake Danao since several areas surrounding the lake have been converted into farmland. Animal manure, sewage, and detergents from nearby residential communities are other possible contributors. Phosphates are only moderately soluble and, unlike nitrates, are not very mobile in soil and groundwater. Phosphates tend to remain attached to soil particles, especially in lentic water ecosystems like lakes (Wurtsbaugh et al., 2019), which can explain the steady rise of phosphate levels since Feb 2022. The negative impacts of excess phosphate on lakes and eutrophication have been extensively studied. Unlike nitrates, moderate increases in P can trigger a series of undesirable events, such as algal blooms and low DO, resulting in the death of aquatic organisms (Smith, 2003; Schindler et al., 2006; Bhateria and Jain, 2016). This cascade of events can further lead to significant economic consequences such as loss of fisheries, reduced recreational activities, and increased costs for drinking water treatment due to the need for advanced filtration and removal of algal toxins (Dodds, 2006). In Lake Danao, the DO levels measured during the last sampling period were found to be below the permissible range (5-7 mg/L) set by the DENR for Class A waters.

The results of the water quality analysis proved that stations generally closer to agriculture and residential areas (LD4 and LD5) have relatively higher FC counts and eutrophic water quality status. Similar results were obtained by Tumanda et al. (2021) for Lake Mainit in Mindanao. High levels of FC and TC were observed in the sampling stations near the three populated areas around the lake. High levels of nutrients were also measured in sampling sites near agricultural and residential lands. Thus, management and policy interventions to address these concerns should be focused on these areas (Frei et al., 2021). These include promotion of sustainable agricultural practices and enforcement of wastewater management strategies for residential areas in accordance with the DENR guidelines to minimize coliform contamination and nutrient pollution (Withers et al., 2014). Examples include the establishment of riparian buffer zones with native vegetation around the lake to minimize nutrient runoff, as well as educating the residents of the nearby communities about the importance of proper waste disposal and the impacts of coliform contamination on the lake.

Continuous monitoring of physicochemical parameters, coliform, and heavy metals is also necessary to provide more reliable evidence for adaptive management strategies of Lake Danao in the future. The evidence of contaminants, specifically high levels of fecal coliform, highlights the urgent need for community education on waste management and stricter controls on agricultural runoff, providing actionable and relevant recommendations for policymakers to mitigate pollution and safeguard public health. The results of these recommendations can be used for proper tourism zoning, reclassification of water, and ensuring a shared responsibility among stakeholders, community members, and policymakers for the continuous improvement and maintenance of the lake water and ecosystem services.

In summary, variation across sampling periods was observed to be more significant than across sampling stations, except for TC and FC levels. Discharge from water runoff during periods of high precipitation was identified as a major factor for the increase or decrease of the metrics. The conditions and activities of the surrounding areas of the lake, such as tourism and agricultural activities, are possible sources of contamination. The observed high phosphate levels constitute a threat to Lake Danao's water quality. High levels of this nutrient can result in algal blooms leading to a decrease in DO levels, indicators of eutrophication. For further study, we recommend increasing sampling frequency, adding more sampling points (for better spatial and temporal representation), and expanding the parameters to include heavy metals or emerging pollutants. This more comprehensive assessment would provide valuable insights into the lake's water quality. Finally, conducting three years of continuous monitoring is also recommended to reevaluate the lake's classification and better understand long-term trends and seasonal variations.

5. CONCLUSION

Lake Danao is classified by the Philippine government as a Class A water body; however, this study reports that there are some parameters that are beyond the maximum allowable limits set by the DENR. FC was consistently recorded to be beyond the permissible levels, indicating high amounts of pathogens in the water. This study recommends reviewing the reclassification of Lake Danao from Class A to Class B. Data analysis revealed significant variability in terms of sampling periods for all parameters, except for TC and FC, indicating that weather conditions, such as the amount of precipitation, are the main factor affecting the water quality of Lake Danao. Based on the NPI, most sampling stations in Lake Danao are "moderately polluted," especially when the amount of precipitation is high. Moreover, the calculated TSI value indicates that Lake Danao's trophic status is "mesotrophic," but the high levels of phosphates obtained mean that the lake is also at risk of eutrophication. Early signs of this were observed in some locations. Therefore, regular monitoring of Lake Danao water quality is imperative to ensure the maintenance, protection, and responsible use of the protected upland lake.

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AUTHOR CONTRIBUTIONS

All authors were involved in the conception and design of the study and carried out the material preparation, data collection, and analysis. All authors provided feedback on earlier drafts. Each author has reviewed and approved the final version of the manuscript.

DECLARATION OF COMPETING INTERESTS

The authors affirm that there are no competing interests regarding the research presented in this manuscript. Any sources of funding and relevant relationships have been transparently disclosed, and no financial or personal factors have influenced the study.

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Research Trends in Organic and Inorganic Waste Sorting Technology: A Bibliometric Analysis

Mahliza Nasution[1*], Muhammad Irwansyah[2], Hermansyah[1], and Rospita Gultom[1]

[1]*Faculty of Engineering, Universitas Medan Area, Indonesia*
[2]*Faculty of Engineering, Universitas Asahan, Indonesia*

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*** Corresponding author:**
E-mail: mahliza@staff.uma.ac.id

ABSTRACT

This research presents major trends in organic and inorganic waste sorting technology over the past 20 years (2003-2023). To do so, we employed bibliometric analysis and VOSviewer and Biblioshiny software. The data used in this study included 810 documents from the Scopus database. This study also used literature analysis of 11 documents discussing the use of AI in organic and inorganic waste sorting technology. The results showed that leading journals covering the topics 'Waste Management and Resources', 'Conservation', and 'Recycling', had the highest publication volume and total citations. Journals in China also had high volume and citations. The literature analysis of the articles further showed that integrating AI into waste-sorting technology can improve organic and inorganic waste-sorting efficiency and effectiveness, and also contribute to sustainable environmental planning. Future research opportunities include enhancing AI accuracy in waste sorting, developing renewable waste-to-energy technologies, and promoting interdisciplinary collaboration to advance sustainable waste management solutions.

HIGHLIGHTS

1. This study reviews research trends in waste sorting tech from 2003 to 2023.
2. Bibliometric analyses used VOSviewer and Biblioshiny on 810 Scopus documents.
3. AI integration enhances sorting accuracy, driving sustainable waste management solutions.

1. INTRODUCTION

Rapid population growth indirectly leads to an increase in waste production; unfortunately, an ineffective waste management system will accumulate waste at the final processing site. This accumulation of waste can potentially produce gases that are harmful to health and the environment (Harjanti and Anggraini, 2020). Indonesia is one of the countries dealing with increasing plastic waste, which has become a serious concern. According to Indonesian data from the Ministry of Environment and Forestry's National Waste Management Information System (SIPSN) for 2022, with input from 2022 districts/cities throughout Indonesia, national waste generation reached 21.1 million tons. Of the total national waste production, 65.71% (13.9 million tons) can be managed, while the remaining 34.29% (7.2 million tons) needs to be managed properly (Rahmatullah, 2023). Based on its nature, waste can be classified into organic and non-organic (Djawad et al., 2022). Organic waste is easily recyclable waste, does not harm the environment, and can be decomposed with the help of microbes. Nevertheless, organic waste still requires careful handling, if not properly processed, it can become a source of odor that disturbs environmental comfort and contributes to environmental pollution (Febriadi, 2019). On the other hand, inorganic waste is made of inorganic materials whose decomposition process takes a very long time (Dzakiya et al., 2019).

In general, waste sorting is still done manually using human hands. Nurdin et al. (2020) stated that

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waste management can be implemented with the 3R principle: reduce, reuse, and recycle. Reduce is an effort to reduce waste generation in the environment at its source even before waste is generated (Arisona, 2018). Each source can contribute to reducing waste by changing its consumption habits, especially switching from wasteful habits that produce much waste to economical, efficient ones that produce little waste. Meanwhile, reuse refers to the reuse of materials or resources so that they do not end up in the trash. Examples include filling milk cans with refillable milk, using paper back and forth, and reusing used beverage bottles into drinking water containers (Junaidi and Utama, 2023). Recycling is one way to reduce existing waste, namely by making waste something that can be reused (Siagian et al., 2022).

Since the waste-sorting process is still done manually, it requires much time and energy. Therefore, the right solution is needed to speed up and simplify sorting organic and non-organic waste (Wibysono et al., 2022). The volume of waste can be gradually reduced by classifying waste correctly so that waste processing becomes easier (Aulia et al., 2021). The use of AI technology helps sort and recycle waste. Artificial intelligence (AI) in waste management is becoming increasingly important in countries worldwide, including China and Europe. AI technology optimizes waste collection routes in major cities such as Shanghai and Beijing. For example, intelligent waste collection systems use sensors and AI algorithms to monitor the volume of waste in each area and plan the most efficient collection routes (Zhu et al., 2019). AI has also become common in waste management in some European countries, such as Sweden and Norway. AI technology monitors and optimizes waste collection, identifying the most efficient collection routes based on volume and time data (Rekabi et al., 2024).

Considering the importance of using artificial intelligence in improving efficiency and waste management, this study aims to explore the development of research related to waste sorting and the use of artificial intelligence through a bibliometric analysis approach. It showcases the importance of using VOSviewer analysis in waste management research to identify research topics, identify research trends, and provide direction for future efforts. Bibliometric analysis is used to identify key trends, analyze key relationships, and evaluate the impact of AI on the efficiency, effectiveness, and sustainability of waste management while identifying unexplored research areas and potential research areas that can lead to innovation-based AI. The research questions addressed in this study are: 1) How have organic and inorganic waste-sorting trends evolved globally in recent years? 2) What are the challenges and opportunities in developing and implementing AI-based waste-sorting systems? Therefore, the main objective of this study is to find out the trends in research on waste-sorting and the use of AI in the waste sorting process.

2. METHODOLOGY

This research uses a bibliometric analysis approach to literature related to sorting organic and inorganic waste and using artificial intelligence in waste sorting. This bibliometric analysis is a quantitative method that involves the application of statistics to evaluate the relationship and impact of publications, authors, institutions, and countries in a particular field of research (Fu et al., 2023). The database used in this research originated from Scopus and was exported on February 10, 2024. The data retrieval process is shown in Table 1.

Table 1 shows the data collection process in waste management research. The inclusion criteria in the study only selected journal articles related to waste sorting, recycling, and AI applications. In addition, this selection focused only on English publications classified as "articles" in Scopus. Meanwhile, the exclusion criteria filtered out less relevant sources, such as conference papers, book chapters, review articles, and studies that broadly addressed waste management or AI without specifically addressing waste sorting. These selection processes considerably improved the accuracy of the studies in analyzing AI applications in waste sorting research. Data collection began by pulling data from the Scopus database. A data search was conducted using the keyword "waste", which yielded 573,104 documents related to waste in general. Then, the search was narrowed to "waste management", resulting in 69,006 documents. This query was further refined by including the keyword "recycling", resulting in 14,989 documents. After that, the keywords were restricted to "sorting", resulting in only 810 documents being identified.

The data was then processed using the bibliometric analysis tools VOSviewer and Biblioshiny to spot research trends, identify key contributors, and visualize networks within the existing literature, providing a basic understanding of how AI has been applied in waste management research. The final stage of the query is integrating AI into the search with the terms “artificial intelligence” or “AI” alongside the previous keywords. The result was only 11 documents, indicating very little research on using AI for waste management, especially in the sorting process. These 11 articles are then analyzed in more detail in the research results.

Table 1. Data collection overview: Waste management research keywords and results

Keywords	Query	Result	Date	Data process
Waste	(TITLE-ABS-KEY (waste)	573,104 documents	10 February 2024	Bibliometric: Vos-viewer Biblioshiny
Waste management	(TITLE-ABS-KEY (waste) AND TITLE-ABS-KEY (“Waste Management”)) AND (LIMIT-TO (LANGUAGE, “English”)) AND (LIMIT-TO (DOCTYPE”ar”))	69,006 documents		
Recycling	(TITLE-ABS-KEY (waste) AND TITLE-ABS-KEY (“Waste Management”) AND TITLE-ABS-KEY (recycling)) AND (LIMIT-TO (LANGUAGE, “English”)) AND (LIMIT-TO (DOCTYPE, “ar”))	14,989 documents		
Sorting	(TITLE-ABS-KEY (waste) AND TITLE-ABS-KEY (“Waste Management”) AND TITLE-ABS-KEY (recycling) AND TITLE-ABS-KEY (sorting)) AND (LIMIT-TO (LANGUAGE, “English”)) AND (LIMIT-TO (DOCTYPE, “ar”))	810 documents		
AI OR “ARTIFICIAL INTELLIGENCE”	(TITLE-ABS-KEY (waste) AND TITLE-ABS-KEY (“Waste Management”) AND TITLE-ABS-KEY (recycling) AND TITLE-ABS-KEY (sorting) AND TITLE-ABS-KEY (ai OR “ARTIFICIAL INTELLIGENCE”)) AND (LIMIT-TO (LANGUAGE, “English”)) AND (LIMIT-TO (DOCTYPE, “ar”))	11 documents		Literature

3. RESULTS AND DISCUSSION

3.1 The research trend analysis of waste sorting technology based on annual publication and countries

The bibliometric method was used to analyze waste sorting research trends. First, the research trend is shown based on annual data production. The annual data production was processed using different bibliometric and network analysis programs: Microsoft Excel and VoSViewer. VoSViewer software was used to determine the research trends on organic and inorganic waste sorting technology, and Microsoft Excel was used to visualize the data.

Figure 1 shows the annual scientific article production data for the last 20 years (2003-2023). Based on the data shown in Figure 1, the research trend of organic and inorganic waste sorting from 2003 to 2023 shows significant development. At the beginning of the period, article production tended to be low, even reaching the lowest point in 2003 with no articles produced. However, since 2009, there has been a steady and sharp increase in the last years (2017-2023). This is due to the awareness of the importance of efficient waste management, the demand for environmental sustainability, technological advances in waste sorting, and the changing era of the Industrial Revolution. Despite a slight decrease in 2023, the number of articles produced remains high compared to the beginning of the period, indicating that this topic remains relevant and in demand by researchers in recent years.

In order to explore the data on countries conducting research in this field, this study investigated author countries based on multi-country or single-country collaboration. The most prominent countries in organic and inorganic waste sorting technology are shown in the documents published in each country and the number of citations (Figure 2). China is ranked first, with the highest number of publications and total citations. Denmark is the most dominant country in organic and inorganic waste sorting technology, with 110.80 citations per document. Next is Singapore, with 71.50, and the Netherlands, with 65.10 citations per document.

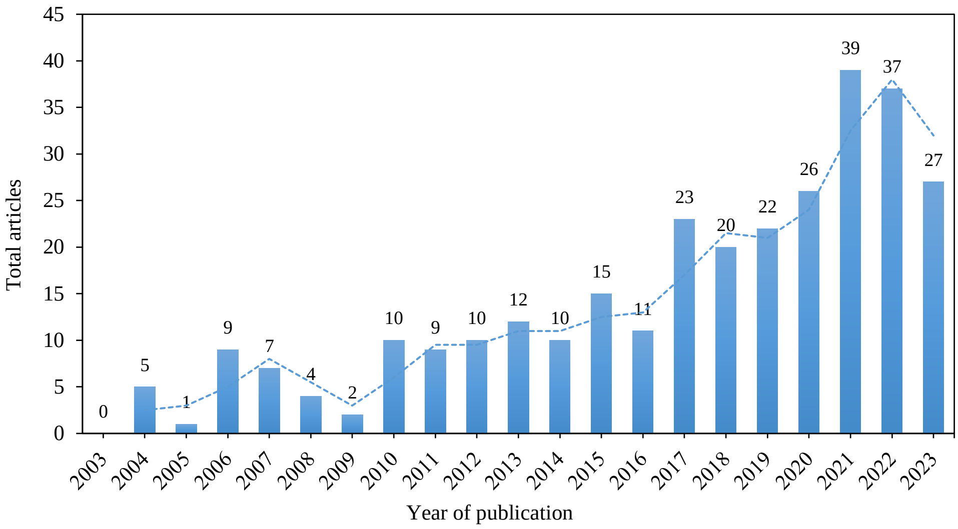

Figure 1. Trends in the number of citations per year 2003-2023

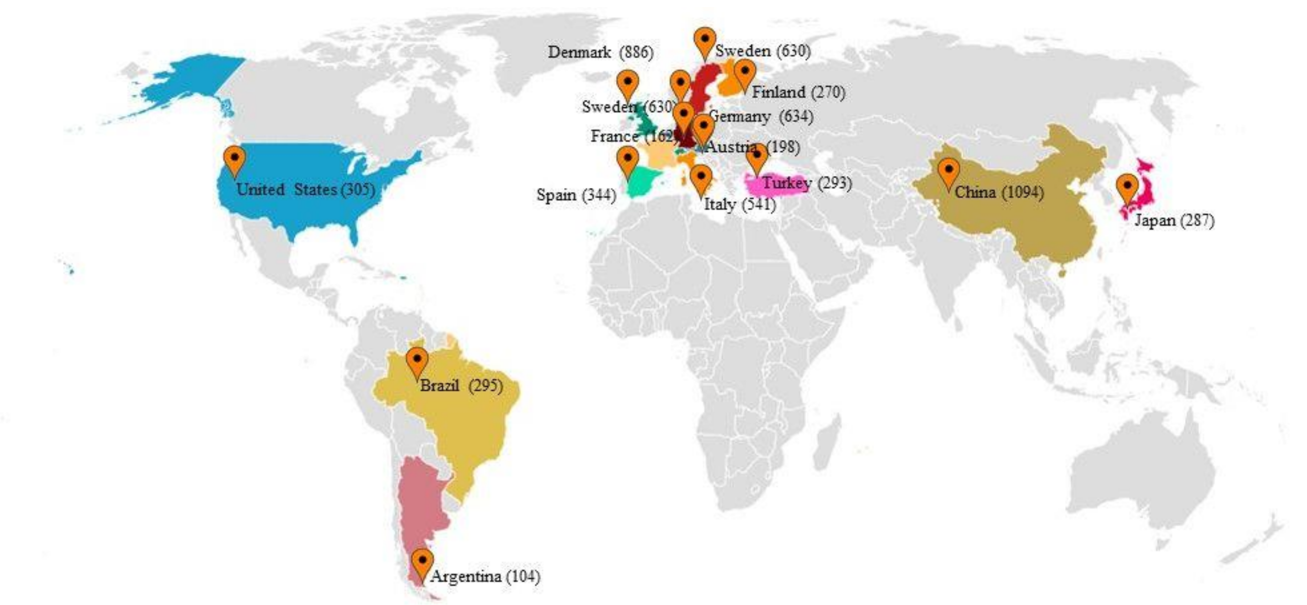

Figure 2. The distribution of countries with highest citation counts in organic and inorganic waste sorting technology research

Figure 2 illustrates the distribution of countries with the highest citations in research on organic and inorganic waste sorting technologies. China leads with 1094 citations, followed by Denmark with 886 citations and Germany with 634 citations. Sweden, the Netherlands, Italy, Spain, the United States, Brazil, and Turkey complete the top 10 list. This data demonstrates the global reach and impact of research efforts in waste sorting technologies, with contributions and recognition coming from countries across continents such as Europe, Asia, and the Americas. However, the detailed country production is shown in Table 2.

Table 2 compares the number of citations and average citations of articles from different countries in waste sorting technology research. China leads with 1,094 total citations but a relatively lower average of 26.70 per article. In contrast, Denmark has fewer citations (886) but a much higher average of 110.80 citations. This indicates the impact and quality of their research output. Other countries include the Netherlands and Singapore, which have relatively high average citations.

3.2 Research trend analysis of leading authors and source of publication

The study found ten of the most influential and prolific authors researching organic and inorganic waste sorting technology. Miezah K's study published in 2015 was the most cited, with 340 citations and an average number of citations per year of 34.0. Similarly, this author published the most recently published article to enter the top 10 most cited articles, (Dahlbo et al., 2017) and (Bernstad, 2014) took second and third place in terms of number of citations, with a total of 222 and 201 citations, respectively, and an average annual number of citations of 31.71 and 18.27, respectively (see Figure 3).

Table 2. Detailed country production information over time

No.	Country	Total citations	Average article citations
1	China	1,094	26.7
2	Denmark	886	110.8
3	Germany	634	25.4
4	Sweden	630	42.0
5	Netherlands	586	65.1
6	Italy	541	33.8
7	Spain	344	49.1
8	Usa	305	27.7
9	Brazil	296	37.0
10	Turkey	293	41.9
11	Japan	287	22.1
12	Finland	270	38.6
13	Belgium	235	21.4
14	Austria	198	14.1
15	France	162	16.2

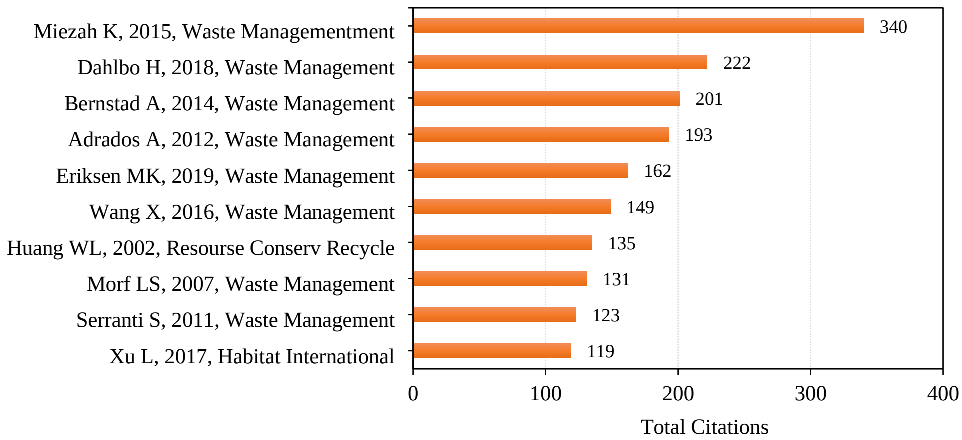

Figure 3. Most global citations based on research on organic and inorganic waste sorting technology

In addition, this research examines the most-published journal data related to research on organic and inorganic waste-sorting technology. It found that the journal with the most published articles was Waste Management (77 articles), followed by Resources, Conservation and Recycling articles and Waste Management and Research articles, with totals of 26 and 22 (see Figure 4).

3.3 Analysis of keywords in research on organic and inorganic waste sorting technology

The most relevant keywords were also analyzed to highlight trending topics and possible future research topics. The research map of organic and inorganic waste-sorting technology articles publishing illustrates the grouping of keywords marked by their color and number of nodes.

The grouping of keywords is categorized by color in Figure 5, and includes sorting and recycling technologies, waste management strategies, and integration of AI and economic principles into sustainable waste management solutions. Cluster 1 (red), consisting of 23 sorting technology items consisting of classification, construction and demolition waste, experimental design, electronic waste, electric charge, electric field, electrostatic process, electrostatic separation, granular materials, image processing, mass balance, particulates, resource recovery, sorting, sorting at source, sorting efficiency, shredding, solid waste management, survey, triboelectricity, waste composition, waste electrical and electronic equipment, and waste sorting.

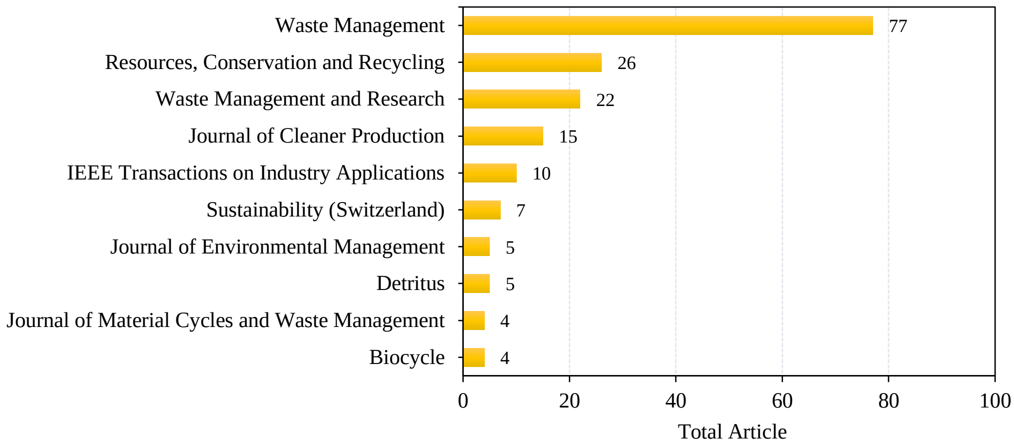

Figure 4. The distribution of published articles in top journals

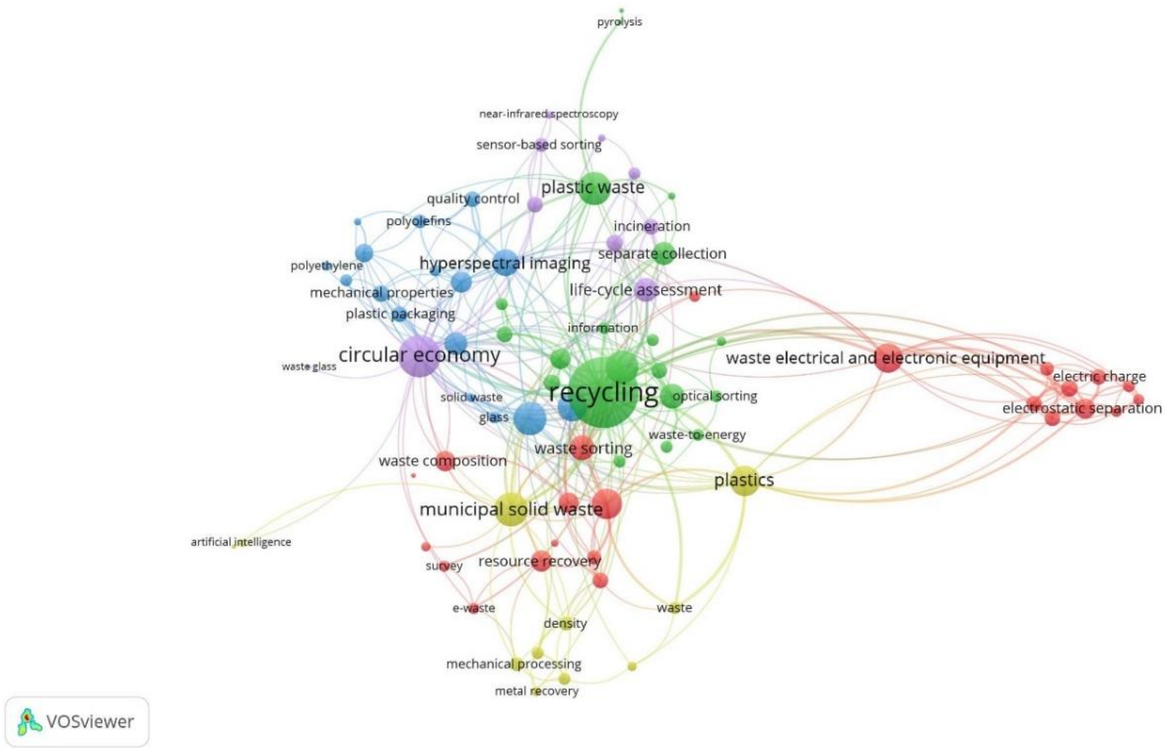

Figure 5. The most relevant keywords related to organic and inorganic waste sorting technology [Source: Data Processing Result-Vosviewer (2024)]

Cluster 2 (green) (green) consists of recycling technologies with 20 items including of China, flotation, food waste, household waste, information, optical sorting, packaging waste, plastic recovery, waste plastics, polymers, pyrolysis, recycling, recycling behavior, separate collection, source separation, sustainability, valorization, waste separation, waste valorization, and waste to energy. Cluster 3 (blue) relates to waste management with 16 items including glass, hyperspectral imaging, material flow analysis, mechanical properties, mechanical recycling, plastic packaging, plastic packaging waste, plastic recycling, polyethylene, polyolefins, post-consumption waste, quality control, recycling, solid waste, waste segregation, and waste management. Cluster 4 (yellow) deals with city solid waste, with 11 items: artificial intelligence, automatic sorting, bottom ash, density, gravity separation, mechanical treatment, metal recovery, metal recycling, municipal solid waste, plastics, and waste. Cluster 5 (purple) is 10 technology-economy items including circular economy, composting, incineration, life cycle assessment, municipal solid waste management, near-infrared spectroscopy, polymer recycling, sensor-based sorting, waste glass, and waste treatment.

As shown in Figure 6, the thematic map was created based on keywords from the authors and mapped into four themes: niche (top left), motor (top right), developing or declining (bottom left), and central (bottom right). In the motor, well-developed research themes are plotted on the top right, including Cluster 1: waste management, waste disposal, municipal solid waste, waste treatment, and solid waste. The basic themes of Cluster 4 include recycling, separation, plastics, plastic recycling, and e-waste. Specific themes include materials such as metals, copper, metal recovery, aluminum, and iron. Emerging or declining themes contain 1 Cluster theme, such as elastomers, electrostatic separators, electronics, electrostatic separation, and polyvinyl chloride.

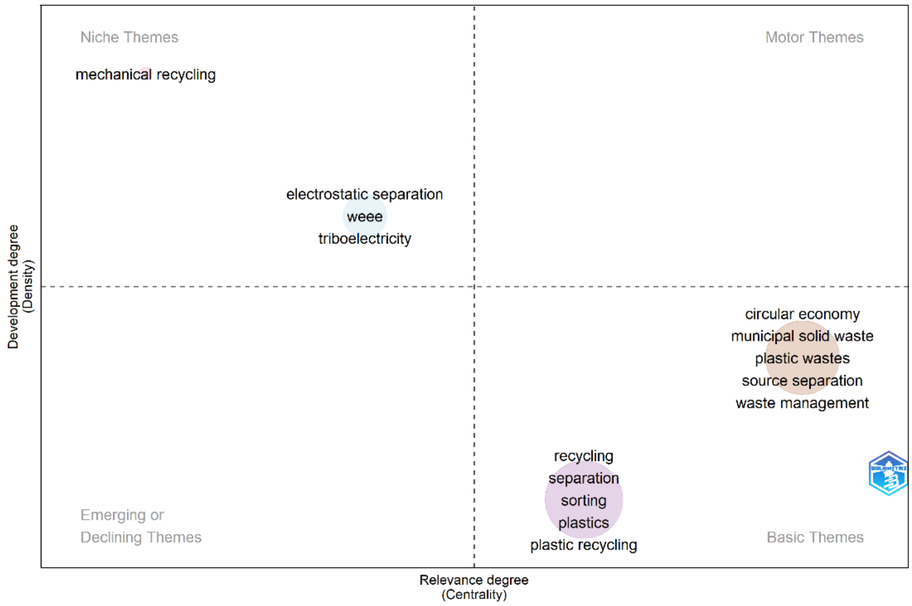

Figure 6. Thematic Map of Authors' Keywords Categorization [Source: Data Processing Result- Biblioshiny (2024)]

Table 3 shows the results of the most relevant literature on integrating artificial intelligence (AI) into waste-sorting technology. The findings show that waste-sorting technology has successfully improved the efficiency and effectiveness of separating organic and inorganic waste. Several research studies have attempted to examine the development and application of advanced AI algorithms in this field. For instance, Liu et al. (2018) have developed an innovative waste sorting system using the SURF-BoW image processing algorithm and Multi-class SVM, which can show improved accuracy in image-based waste sorting. In addition, Chen et al. (2021) and Wang (2022) also tried to use learning techniques to improve the classification and separation of recyclable waste. These findings also show the potential of AI in optimizing the waste management process. The innovations from these studies demonstrate waste segregation and contribute to more sustainable environmental planning and management.

In addition, some studies have also combined AI with other technologies, such as the Internet of Things (IoT), and continue to revolutionize the waste management system. The research of Liao (2022) and Wang et al. (2021) integrated IoT with machine learning and deep learning to create an innovative city waste management system. The system uses real-time data analytics to optimize waste collection routes, monitor waste levels, and improve operational efficiency. In addition, Aroba et al. (2023) and Tandon and Bansal (2023) also examined the application of innovative waste management solutions in smart cities. This demonstrates the transformative impact of AI and IoT in achieving sustainable waste management practices and improving urban living conditions. This literature review shows that AI has also been widely used in waste sorting.

Table 3. The most relevant articles on artificial intelligence integration into waste-sorting technology

No.	Journal title	Author	The role of AI in waste sorting
1	Novel smart waste sorting system based on image processing Algorithms: SURF-BoW and Multi-class SVM	Liu et al. (2018)	Development of an intelligent waste sorting system that uses SURF-BoW and Multi-class SVM image processing algorithms to improve efficiency in the process of image-based waste sorting.
2	Optimization of site selection for construction and demolition waste recycling plant using genetic algorithm	Liu et al. (2019)	Use of AI techniques, in the process of optimizing site selection for construction and demolition waste recycling plants.
3	Artificial intelligence-based e-waste management for environmental planning	Chen et al. (2021)	Application of AI to efficiently manage e-waste, to support more sustainable environmental planning.
4	The adoption of an intelligent waste collection system in a smart city	Aroba et al. (2023)	Implementation of smart waste collection systems in smart cities, which improve the efficiency and effectiveness of waste management through innovative technologies.
5	Intelligent garbage classification system based on deep learning	Liao (2022)	Using deep learning to recognize and separate recyclable waste.
6	IoT-enabled smart city waste management using machine learning analytics	Bakhshi and Ahmed (2018)	Application of Internet of Things (IoT) technology to manage waste in smart cities using machine learning analytics, improving the efficiency of waste collection and processing.
7	A smart IoT system for waste management	Chen et al. (2018)	Implementing a smart waste management system utilizing IoT technology.
8	Research on intelligent garbage classification algorithm based on deep learning	Wang (2022)	Development of deep learning and application of Python in deep learning, and study of intelligent garbage classification algorithms based on Python deep learning.
9	A smart municipal waste management system based on deep-learning and internet of things	Wang et al. (2021)	Combining IoT technology and deep learning to improve waste management efficiency.
10	Waste management system using IoT-based machine learning in university	Khoa et al. (2020)	Optimizing waste collection routes using machine learning techniques.
11	IoT-enabled smart waste management: Leveraging the power of IoT for sustainable solutions	Tandon and Bansal (2023)	IoT-based waste management system can improve the efficiency of waste collection.

The differences between municipal, industrial, and medical waste can affect the accuracy of AI in waste classification and management. For example, AI trained only on municipal waste will have difficulty with accuracy when identifying medical or industrial waste because each type of waste is different. Municipal waste is mixed, industrial waste may contain toxic materials, and medical waste includes syringes and expired medications. In addition, differences in regulations and classification standards can cause AI to classify non-hazardous waste into hazardous waste incorrectly. The technology is also different: computer vision for municipal waste, chemical sensors for industrial waste, and object recognition for medical waste. Therefore, the model must be trained with waste data covering all the above categories for AI to work accurately.

Some future-related research can be developed related to waste management. First, in developing a waste sorting system, AI can improve the accuracy and speed of waste sorting with advanced image recognition and data analysis. In addition, AI can also be used to predict the amount and type of waste generated, which can be used for easier waste management planning and lower operational costs. Secondly, the development of renewable technologies, including the production of bioenergy from organic waste such as biogas and biofuel, and the improvement of the efficiency of of the conversion process should be encouraged. Third, research on the potential size of microorganisms can be developed to help break down various types of organic waste that can be used in waste-conversion technologies. Fourth, multi-disciplinary collaboration on waste management challenges involving scientists, government, industry, and communities to support innovation and investment in waste management technologies.

4. CONCLUSION

The findings of this study show that the trend of research on organic and inorganic waste sorting technology is growing significantly. This indicates that interest in this research is relatively high. This research found that China is the most productive country, based on the number of citations. However, most of the other productive countries are in Europe, making it the continent with the most countries contributing to waste management research. Then, regarding authors, Miezah K's research, published in 2015, was the most cited. Meanwhile, the journal that published the most articles was Waste Management Journal, followed by Resources, Conservation and Recycling, and Waste Management and Research Journal. A literature analysis of the most relevant articles showed that integrating artificial intelligence (AI) into waste-sorting technology can improve the efficiency and effectiveness of sorting organic and inorganic waste and contribute to sustainable environmental planning.

In the future, research and applications related to organic and inorganic waste sorting technology research can be one of the efforts to minimize waste. In addition, research on organic and inorganic waste sorting technology is fundamental to solving environmental problems. This research is the basis for developing new scientific research questions. It is recommended that researchers pursue the development of AI-enabled waste sorting technology, refine the sorting processes, and carry out a thorough impact study. These researchers should also strengthen collaboration with international colleagues and publish in well-known journals so that their output has more impact. Policymakers must put more effort into establishing regulatory standards, funding research, and promoting sorting technologies in industries and cities. Launching educational training and establishing joint efforts with the private sector are also important to hasten the introduction of better and more sustainable waste-sorting technologies.

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DECLARATION OF COMPETING INTEREST

The authors declare no conflict of interest.

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AdjcorT-RBFNN for Air Quality Classification: Mitigating Multicollinearity with Real and Simulated Data

Siti Khadijah Arafin[1], Nor Azura Md Ghani[1], Marshima Mohd Rosli[2,3], and Nurain Ibrahim[1,4*]

[1]School of Mathematical Sciences, College of Computing, Informatics and Mathematics, Universiti Teknologi MARA, Shah Alam, Selangor, Malaysia
[2]School of Computing Sciences, College of Computing, Informatics and Mathematics, Universiti Teknologi MARA, Shah Alam, Selangor, Malaysia
[3]Cardiovascular Advancement and Research Excellence (CARE Institute), Sungai Buloh Campus, 47000 Sungai Buloh, Selangor, Malaysia
[4]Institute for Big Data Analytics and Artificial Intelligence (IBDAAI), Kompleks Al-Khawarizmi, Universiti Teknologi MARA, Shah Alam, Selangor, Malaysia

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*** Corresponding author:**
E-mail:
nurain@tmsk.uitm.edu.my

ABSTRACT

Air pollution levels have remained a significant issue worldwide despite advancements in technology, primarily due to rapid industrialization and urbanization. Among the various pollutants, PM2.5 significantly impacts air quality, posing health risks such as respiratory and cardiovascular diseases. Accurate prediction of PM2.5 levels is essential for effective air quality management. However, multicollinearity in air quality data can hinder model performance. To address this issue, this study introduces the AdjcorT-RBFNN, a two-stage feature selection method, to classify air quality in Klang, Selangor. The AdjcorT-RBFNN model selects the optimal combination of 9 feature combinations from 10 variables and outperforms the RBFNN model, which uses all 10 variables. With 7 hidden nodes and a learning rate of 0.01 for both models, AdjcorT-RBFNN achieves higher accuracy (0.62), sensitivity (0.64), specificity (0.60), precision (0.60), F1 score (0.62), and AUROC (0.62), confirming its effectiveness in classification tasks. The optimal features for predicting air quality in Klang are identified as PM2.5, PM10, relative humidity, SO_2, wind direction, O_3, CO, ambient temperature, and NO_2. Monte Carlo simulations validate the model's effectiveness, showing that AdjcorT-RBFNN consistently outperforms RBFNN, especially with strong negative correlations (ρ=-0.8) and larger sample sizes (N=150 and 200) further enhance classification accuracy. Compared to RBFNN, AdjcorT-RBFNN enhances class discrimination and reduces false positives, improving its reliability in detecting true classifications. These findings highlight the importance of feature selection in improving model performance, particularly in datasets with multicollinearity. Researchers, and health organizations can leverage AdjcorT-RBFNN for more accurate air quality predictions, supporting informed pollution control strategies.

1. INTRODUCTION

Air quality prediction has emerged as a significant issue in recent years, due to the increasing effects of air pollution on human health, climate change, and ecosystems. PM2.5, fine particulate matter with a diameter of 2.5 micrometers or smaller, is a major air pollutant often associated with severe health risks. These particles are small enough to be inhaled deeply into the lungs, and because of their size, they can also enter the bloodstream. PM2.5 is primarily generated from industrial emissions, vehicle exhaust, biomass burning, and other sources of combustion, as well as natural sources such as dust storms and wildfires. Due to its fine nature, PM2.5 can

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carry a variety of toxic compounds, including heavy metals, organic chemicals, and acids, which contribute to its toxicity. Pregnancy complications, lung cancer, cardiovascular and respiratory disorders, and other health problems have all been connected to exposure to PM2.5. Notably, Jalali et al. (2021) found a substantial correlation between elevated PM2.5 levels and higher mortality rates in a country in the Eastern Mediterranean. Furthermore, prolonged exposure to high levels of PM2.5 is known to cause oxidative stress, inflammation, and damage to cells and tissues, which exacerbate these health effects.

As a result, PM2.5 pollution has become a serious public health concern worldwide, including in Malaysia, where the Department of Environment Malaysia (DOE) began measuring PM2.5 in April 2017. Machine learning, particularly neural networks, is increasingly applied in classification and regression tasks. One of the primary advantages of using machine learning for air quality classification is its ability to process complex datasets and identify non-linear relationships among variables. A study by Zhang et al. (2023) on air quality index prediction in six Chinese urban areas found that ensemble approaches enhanced prediction accuracy by including numerous data sources, such as pollution and meteorological data. Additionally, Liu (2024) highlighted the limitations of traditional empirical models, stating that machine learning offers more accurate predictions for air quality management. Radial Basis Function Neural Network (RBFNN) is one of the well-known neural networks that offer fast convergence compared to others, such as Multi-Layer Perceptrons (Zhou et al., 2019a), primarily due to its simpler structure and fewer training parameters. A study by Li et al. (2022) on predicting water quality parameters also found that the RBFNN model demonstrated promising performance with high accuracy in individual indicator predictions, though it still exhibited a significant accuracy gap in some cases.

While neural networks offer advantages in processing complex datasets, effective feature selection is crucial in improving the reliability and performance of machine learning models. Suresh et al. (2022) emphasize that reducing the complexity of the dataset through feature selection can lead to improved model performance. Nazari et al. (2023) also highlight that feature selection helps classification algorithms focus on the most relevant features, reducing computational burden and improving accuracy. Similarly, Ul-Saufie et al. (2022) demonstrate that wrapper feature selection approaches can improve air pollution prediction by selecting important features, with brute-force being particularly effective.

Nevertheless, a major challenge in predictive modelling is addressing multicollinearity among air pollutants and meteorological factors, which can lead to inaccurate predictions. Many air pollutants are strongly correlated, meaning their concentrations tend to change together or also know has multicollinearity exist. Zhou et al. (2019b) found a high correlation between PM2.5 and PM10 levels in their study on air pollution and respiratory diseases. Their findings indicate that PM2.5 often constitutes a significant portion of PM10 concentrations, contributing to shared health impacts on respiratory health. Similarly, gases like carbon monoxide (CO) and nitrogen dioxide (NO_2) also demonstrate correlated increases, primarily as a result of vehicle emissions and combustion processes inherent to urban environments (Wang et al., 2022). When these relationships are not properly addressed, machine learning models may struggle to determine which factors are truly important, thus reducing the reliability of predictions.

This study focuses on Klang, Selangor, due to its severe pollution levels, heavily influenced by industrial emissions and port-related activities. Klang's air quality is largely affected by both local industrial zones and transboundary pollution from shipping activities, making it a crucial case study. Mohtar et al. (2022) highlighted in their study that the Klang station in Klang Valley, near Malaysia's busiest shipping port, Port Klang, often experiences the highest concentrations of particulate matter, making it a significant contributor to the country's air pollution issues. Despite its importance, few studies have examined feature selection techniques focused on multicollinearity issues for air quality classification in Klang.

Furthermore, this study considers ten key input variables influencing PM2.5 levels, including six pollutants (PM10, SO_2, NO_2, O_3, and CO). These pollutants were selected due to their significant impact on air pollution, as supported by previous studies on air quality classification, such as the study by Sapari et al. (2023). Moreover, Liu et al. (2020) found that changes in wind speed and temperature directly influence pollutant concentrations in China. Specifically, their study reported that an increase in wind speed generally improves air quality by dispersing pollutants away from densely populated areas. In addition, Wattimena et al. (2022) highlighted the importance of meteorological factors such as wind speed, cloud volume, air pressure, temperature, relative humidity, and precipitation in forecasting air quality indices and improving prediction accuracy. However, only four meteorological parameters (wind direction, wind speed, relative humidity, and temperature) were included in this study, as these are the key parameters consistently recorded by the DOE.

The AdjcorT two-stage feature selection method has been shown to enhance RBFNN classification by mitigating multicollinearity, as demonstrated in a study by Arafin et al. (2024). In their study, they applied the method to Shah Alam's air quality dataset, improving predictive accuracy. However, their study did not explore its applicability in different urban environments with distinct pollution sources, such as Klang. This research aims to fill this gap by applying the AdjcorT-RBFNN method to Klang's air quality dataset and verifying its performance through Monte Carlo simulations.

By systematically evaluating the effect of multicollinearity and comparing AdjcorT-RBFNN with a RBFNN model, this study seeks to enhance air quality classification for improved environmental decision-making.

2. METHODOLOGY

2.1 Data description

The air quality dataset for the Klang, Selangor (CA21B) area was obtained from the Department of Environment (DOE) for the years 2018 to 2022. The extracted variables include Particulate Matter (PM2.5 and PM10), Sulphur Dioxide (SO_2), Nitrogen Dioxide (NO_2), Ground-Level Ozone (O_3), Carbon Monoxide (CO), wind direction, wind speed, relative humidity and ambient temperature, all in hourly format, comprising a total of 43,824 samples. However, the PM2.5 data for 1st January 2018, is not available for every hour. Therefore, we removed 24 data points from 1st January 2018, reducing the dataset to 43,800 samples. Moreover, the dataset contains missing values as shown in Table 1. According to the table, the percentage of missing values of all variables are below 10%, with NO_2 has the highest missing value (7.7%). Meanwhile, data of ambient temperature has lowest missing value which is 0.4% of dataset. According to Chen and Li (2024), failure of monitoring instruments is one of the most common causes of missing data. The instruments malfunction might happen due to the extreme weather, power outages, or periodic maintenance, resulting in gaps in data collection. (Ghazali et al., 2021). Additionally, missing values in air quality datasets can adversely affect the performance of analytical models, leading to misleading (Ghazali et al., 2021). Thus, this study employed a widely used imputation method to impute the missing values which is linear interpolation. According to Van Rossum et al. (2023), linear interpolation is a simple method yet it has been shown to yield higher imputation accuracy.

Table 1. Percentage of missing values

Variable	N	Missing value
PM2.5	43,572	228 (0.5%)
PM10	43,462	338 (0.8%)
SO_2	40,648	3,152 (7.2%)
NO_2	40,414	3,386 (7.7%)
O_3	41,450	2,350 (5.4%)
CO	41,219	2,581 (5.9%)
WD	43,596	204 (0.5%)
WS	43,594	206 (0.5%)
Humidity	43,511	289 (0.7%)
Temperature	43,606	194 (0.4%)

2.2 Research framework

Figure 1 shows the research framework employed in this study to predict PM2.5 levels in Klang, Malaysia. The process begins with the collection of air quality data from the Department of Environment, Malaysia, for the years 2018 to 2022. Then, the data undergoes pre-processing, involving linear interpolation for imputing missing values, conversion of hourly data to daily averages, binary classification of PM2.5 levels, min-max normalization for feature scaling, and the application of the Synthetic Minority Oversampling Technique (SMOTE) to handle class imbalance.

Next, we use Spearman Correlation to explore the correlation between independent variables within the dataset to understand its extent and impact on model performance. To address this, the AdjcorT feature selection method is employed to rank variables based on their correlation and importance, mitigating the influence of multicollinearity. Subsequently, feature subsets are evaluated using an Artificial Neural Network (ANN) model to identify the best combinations for PM2.5 prediction. Additionally, two models are then developed and compared: a standard RBFNN and a Two-Stage AdjcorT-RBFNN, which integrates AdjcorT feature selection with RBFNN. The models are evaluated based on classification metrics such as accuracy, sensitivity, specificity, and

AUROC to identify the best-performing model. Finally, the robustness of the selected model is validated using simulation data with varying sample sizes and correlations, ensuring its reliability in identifying important predictors under various conditions.

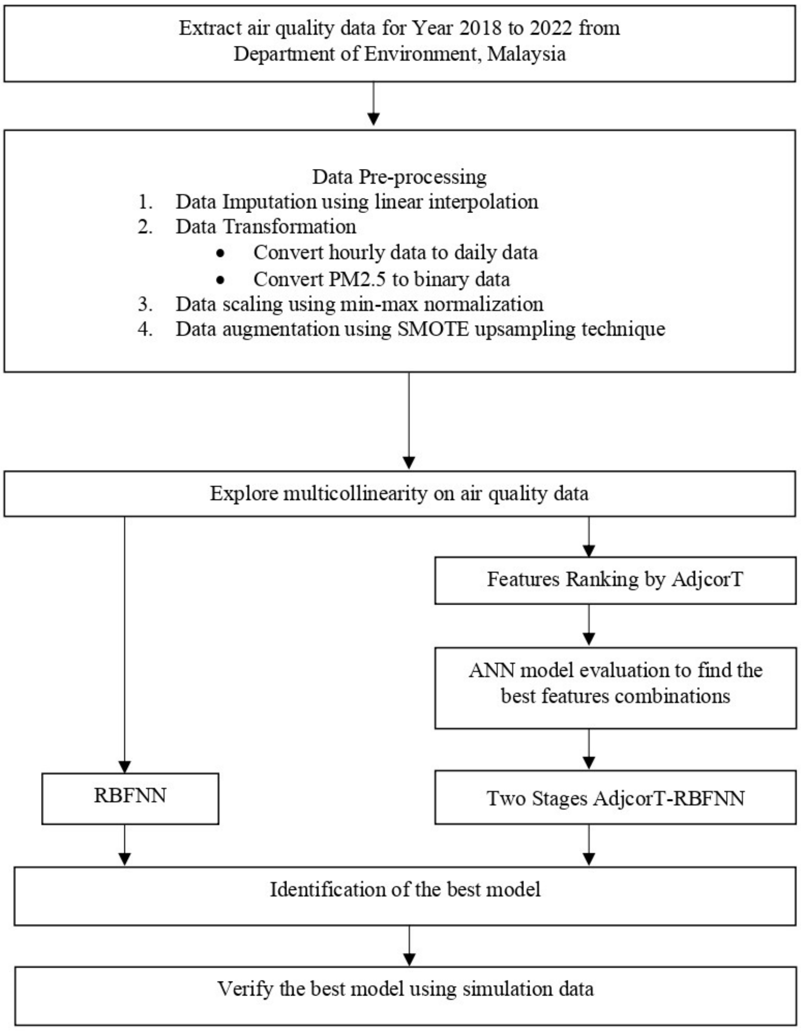

Figure 1. Research framework

2.3 Adjusted correlation sharing t-test

Ibrahim (2020) extended the variable selection method which is correlation sharing t-statistics (corT), by developing an adjusted version namely adjusted correlation sharing t-test (AdjcorT). Both methods rank the importance of features while considering the high correlation between variables. However, the algorithm of AdjcorT allowed both positive and negative correlation between variables, meanwhile corT considers only positive correlations. The standard t-statistics were calculated first using equation shows in (1). S_i is the pooled standard deviation within the group for the i-th variable. $\bar{x}_{ij}$ represents the average of the i-th variable for the j-th class or target variable (where j=0 or j=1). The equation of AdjcorT is displayed in (2):

$$T_i = \frac{\bar{x}_{i1} - \bar{x}_{i0}}{S_i} \quad (1)$$

$$r_i = \text{sign}\left(\frac{\bar{x}_{i1} - \bar{x}_{i0}}{S_i}\right) \times \left[\max_{(0 \le \rho \le 1)} \frac{1}{w} \sum_{j \in C_\rho(i)} \left|T_j\right|\right] \quad (2)$$

A score, or an AdjcorT value, r_i, is assigned to each variable. This value represents the average of all t-statistics for variables that have a correlation (in absolute value) of at least ρ with variable i. Additionally, w denotes the cardinality of $C_\rho(i)$, where $C_\rho(i)$ is the set of indices of variables whose correlation (in absolute value) with variable x_i is greater than or equal to ρ. According to Ibrahim (2020), the optimal value of ρ should be chosen to maximize the average. Moreover, since the t-scores for each variable are calculated to determine the correlation, this method is applicable only to continuous variables.

2.4 Artificial neural network

Artificial Neural Network (ANN) is a machine learning that designed inspired of biological neural networks. An ANN has three layers: the input layer (which consists of input variables), the hidden layer (which applies activation functions), and the output layer. They are designed to identify patterns and address complex problems by learning from data. ANNs consist of interconnected artificial neurons, also known as nodes, that work collaboratively to process information. Each neuron receives input, processes it, and generates output that can be transmitted to successive neurons in the network. This design enables artificial neural networks to perform tasks such as classification, regression, and pattern recognition. In our study, we used R Studio to implement an ANN model using the nnet package. To effectively train an ANN, several key hyperparameters must be set. These include the number of hidden neuron and learning rate, where the number of hidden neurons we set is 7 and 0.01 for learning rate as suggested by Ul-Saufie et al. (2022). The activation function used in the hidden layer is the logistic sigmoid, which maps inputs to a range between 0 and 1.

2.5 Radial basis function neural network (RBFNN)

RBFNN is a subset of ANN that share the same theoretical framework, consisting of three layers which is input, hidden and output layer. The difference is that RBFNNs use a radial basis function, typically Gaussian functions, as the activation function. RBFNN are particularly effective in addressing nonlinear relationships in data, such as air quality measurements. Their ability to approximate complex functions with relatively few parameters makes them highly suitable for tasks like pattern recognition and function approximation. The RSNNS package in RStudio was used to implement the RBFNN.

2.6 Performance metrics

Different performance metrics are essential for evaluating and comparing machine learning classification models, enabling informed decisions about model selection and improvement (Akshay et al., 2022). According to Ibrahim (2020), accuracy, sensitivity, specificity, and Area Under the Receiver Operating Characteristic (AUROC) are common key metrics used in previous study to assess classification model. For instance, Alalwany and Mahgoub (2024) employed accuracy, precision, recall, F1 score, and ROC metrics for evaluating the performance of their model on an ensemble learning-based real-time intrusion detection scheme for in-vehicle networks. Moreover, Chandra et al. (2022) used performance metrics including accuracy, specificity, F1 score, sensitivity, and precision to predict Jakarta's air quality.

In this study, accuracy, sensitivity, specificity, precision, F1 score, and AUROC are used to evaluate model performance of using real-world data and simulated data. Both real-world air quality data and simulated data were used to assess the classification effectiveness of the RBFNN and adjcorT-RBFNN models. The real data evaluation examines model performance based on actual air quality measurements from Klang, while the simulated data evaluation helps analyze the models' behavior under controlled conditions, particularly in handling multicollinearity. The combination of these two evaluations ensures a comprehensive assessment of the models' predictive capabilities. The specific settings for generating the simulated datasets, including sample sizes, correlation structures, and iteration processes, are detailed in Section 2.7.

2.7 Monte carlo simulation

Monte Carlo Simulation is a computational technique used to model the probability of different outcomes in systems influenced by randomness. It relies on repeated random sampling to approximate results, making it useful for solving problems involving uncertainty and complex decision-making (Liu, 2024). This method is widely applied in finance, engineering, healthcare, and machine learning, where it helps optimize strategies by evaluating potential scenarios. Monte Carlo Simulation is particularly valuable when analytical solutions are infeasible due to system complexity. In this study, Monte Carlo Simulation was employed to systematically generate datasets with varying correlation structures and sample sizes. This process allowed for the evaluation of how different levels of correlation affect the classification performance of RBFNN and adjcorT-RBFNN.

To achieve this, we use RStudio to run both the RBFNN and AdjcorT-RBFNN models using simulated data with varying sample sizes (n=50, 100, 150, 200) and correlation values (ρ=-0.8, -0.5, -0.2, 0, 0.2, 0.5, 0.8) to evaluate the impact of correlation and sample size on classification accuracy. Each scenario was simulated over 100 iterations to ensure statistical robustness and reduce variability in performance estimates. During each iteration, the models were trained and evaluated on a newly generated dataset, allowing us to analyse how correlation and sample size influence classification performance. By incorporating multiple iterations, we mitigated the effects of random variations in individual datasets, ensuring more reliable comparisons between the models. This simulation-based validation helps confirm whether AdjcorT effectively addresses multicollinearity and enhances the classification capability of RBFNN, particularly under different correlation strengths.

3. RESULTS AND DISCUSSION

3.1 Data pre-processing

Data transformation was employed in this study, where the hourly dataset was converted to daily data by aggregating the values over 24 hours. Moreover, the dependent variable, $PM2.5_{D+1}$ values was transformed into binary data. Kalajdjieski et al. (2020) suggested separating the air quality category into two groups only, polluted and not polluted. Table 2 shows the PM2.5 breakpoints (24-hour average) according to DOE guidelines.

The descriptive statistics of the dataset after data transformation are shown in Table 3. The total number of samples (N) was reduced to 1,824 because the hourly data was transformed into daily data. Based on the table, the average PM2.5 levels is 26 µg/m^3, while the maximum value is 154 µg/m^3. Moreover, the standard deviation of SO_2 is the lowest (0.001), while the highest standard deviation is 35, which corresponds to wind direction. This wide difference in scale can affect the accuracy of classification. Hence, we employed min-max normalization to standardize these measurements, thereby enhancing the interpretability of the results, following a study by Aarthi et al. (2023). In addition, the distribution of PM2.5 categories is not balanced as shown in Figure 2, where not polluted (86%) category is more than polluted (14%). To address this issue, the applied Synthetic Minority Over-sampling Technique (SMOTE) was applied to the dataset.

Table 2. Binary labels for the respective PM2.5 breakpoint and AQI categories

AQI category	PM2.5 breakpoints	Binary labels
Good	0.0-12.0	Not polluted
Moderate	12.1-35.4	Not polluted
Unhealthy for sensitive groups	35.5-55.4	Polluted
Unhealthy	55.5-150.4	Polluted
Very unhealthy	150.5-250.4	Polluted
Hazardous	250.5 and above	Polluted

Table 3. Descriptive statistics before data pre-processing

Variable	N	Mean	Median	Std. Dev.	Skewness	Min	Max
PM2.5	1,824	26.309	24.206	12.341	3.720	9.134	154.845
PM10	1,824	35.890	33.103	15.529	2.880	10.774	180.227
SO_2	1,824	0.002	0.001	0.001	2.512	0.000	0.009
NO_2	1,824	0.017	0.016	0.005	0.453	0.003	0.040
O_3	1,824	0.015	0.015	0.005	0.662	0.002	0.040
CO	1,824	0.871	0.852	0.264	0.305	0.122	1.835
WD	1,824	169.988	161.410	35.632	1.093	72.039	327.733
WS	1,824	1.375	1.319	0.340	0.933	0.556	3.500
Humidity	1,824	80.518	80.413	5.916	-0.092	58.659	100.000
Temperature	1,824	28.350	28.442	1.157	-0.367	23.204	31.281

The descriptive statistics after data pre-processing were recomputed, as shown in Table 4. This table presents the descriptive statistics of the variables after data normalization and standardization. The total number of samples increased due to the application of the SMOTE up-sampling technique. The dataset now consists of 3,089 samples, with 80% used for training and 20% for testing. According to the table, the minimum and maximum values of all variables are 0 and 1, respectively. This result indicates that all variables has successfully scaled into a standard range between 0 and 1. Furthermore,

Figure 3 shows the distribution of PM2.5 category after employed SMOTE technique are more balanced now with 50.9% of dataset are not polluted category while 49.1% of dataset are polluted category. Hence, the dataset is more suitable to train for classification task because SMOTE method SMOTE can significantly enhance the classification performance of machine learning models, particularly in scenarios where the minority class is critical as highlighted by Ariansyah et al. (2023).

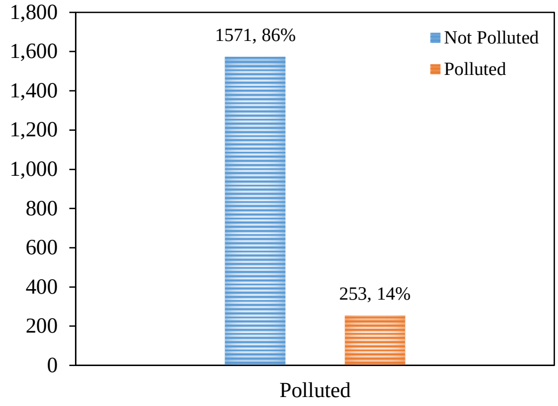

Figure 2. PM2.5$_{D+1}$ distribution (Before SMOTE)

Table 4. Descriptive statistics after data pre-processing

Variable	N	Mean	Median	Std. Dev	Skewness	Min	Max
PM2.5	3,089	0.127	0.108	0.087	2.908	0	1
PM10	3,089	0.158	0.136	0.094	2.259	0	1
SO_2	3,089	0.172	0.147	0.120	2.691	0	1
NO_2	3,089	0.381	0.368	0.133	0.621	0	1
O_3	3,089	0.349	0.329	0.137	0.694	0	1
CO	3,089	0.349	0.329	0.137	0.694	0	1
WD	3,089	0.373	0.337	0.130	1.288	0	1
WS	3,089	0.284	0.266	0.113	0.718	0	1
Humidity	3,089	0.515	0.506	0.140	0.044	0	1
Temperature	3,089	0.644	0.660	0.137	-0.474	0	1

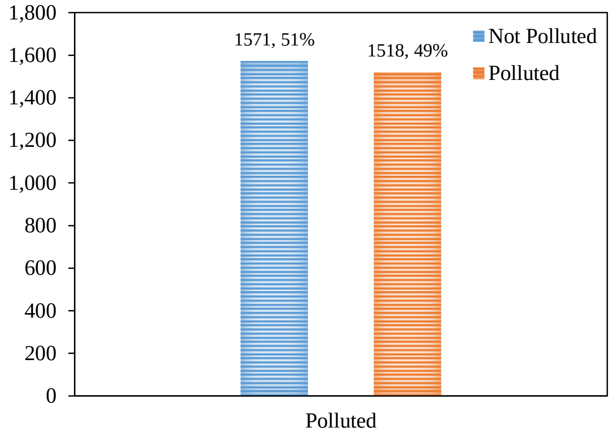

Figure 3. PM2.5$_{D+1}$ distribution (After SMOTE)

3.2 Correlation between features

The high correlation between features or also known as multicollinearity might distort the accuracy of predictive model because the highly correlated variables may share similar characteristics. For instance, a study by Kılıçoğlu and Yerlikaya-Özkurt (2024) highlighted that the high correlation among independent variables may reduce the reliability of regression coefficients, making it difficult to draw meaningful inference from the model. Spearman correlation is less sensitive to outliers than pearson correlation due to its ranking of data rather than raw values, reducing extreme values' influence on the correlation coefficient (Hou et al., 2022). Moreover, the spearman correlation is a non-parametric measure, and hence it does not assume a specific distribution for the data making it more reliable than pearson correlation (Hou et al., 2022).

Therefore, Spearman correlation matrix was computed to examine the correlation between variables in Klang's air quality dataset as shown in Table 5. Spearman correlation values range from -1 to 1, where values closer to 1 indicate a strong positive correlation, and values closer to -1 indicate a strong negative correlation. According to the analysis, the Spearman correlation value between PM2.5 and PM10 is 0.94, suggesting a very strong positive correlation between these variables. Furthermore, relative humidity and ambient temperature show a strong negative correlation, with a value of -0.84. Additionally, both particulate matters (PM10 and PM2.5) have moderate positive correlations with CO, with a value of 0.55 and 0.59 respectively. In addition, NO_2 also have moderate positive correlations with

CO, with a value of 0.57. Notably, wind speed also shows moderate negative correlation with CO with spearman correlation of -0.5. Other variables exhibit correlation values below±0.5.

Although certain variables, such as PM10 and PM2.5, exhibit high correlation, they should not be removed solely based on this criterion. Despite their strong raltionship, each variable may capture unique characteristics that contribute to air quality classification. For instance, PM2.5 and PM10, while strongly correlated, represent different particle size fractions with distinct health and environmental implications. Removing one could result in the loss of valuable information that enhances model performance. While multicollinearity can pose challenges in linear models by inflating variance and reducing interpretability, its impact on non-linear models like RBFNN is less pronounced. RBFNN can effectively learn complex relationships even when inputs are correlated. However, a high degree of multicollinearity may introduce redundancy, which is why the AdjcorT feature selection method was applied in Section 3.4. AdjcorT identifies the most informative features while preserving key variables that contribute to classification accuracy, ensuring that the model benefits from a diverse yet relevant set of inputs.

Table 5. Spearman correlation matrix

Variables	PM2.5	PM10	SO_2	NO_2	O_3	CO	WD	WS	Humidity	Temperature
PM2.5	1.00	0.94	0.09	0.37	0.16	0.59	-0.20	-0.07	-0.33	0.33
PM10	0.94	1.00	0.18	0.39	0.16	0.55	-0.16	-0.05	-0.37	0.33
SO_2	0.09	0.18	1.00	0.03	0.06	0.03	0.09	0.18	-0.20	0.09
NO_2	0.37	0.39	0.03	1.00	-0.05	0.57	-0.08	-0.50	0.14	-0.22
O_3	0.16	0.16	0.06	-0.05	1.00	-0.01	0.02	0.05	-0.39	0.36
CO	0.59	0.55	0.03	0.57	-0.01	1.00	-0.10	-0.23	-0.14	0.07
WD	-0.20	-0.16	0.09	-0.08	0.02	-0.10	1.00	0.03	0.07	-0.09
WS	-0.07	-0.05	0.18	-0.50	0.05	-0.23	0.03	1.00	-0.47	0.42
Humidity	-0.33	-0.37	-0.20	0.14	-0.39	-0.14	0.07	-0.47	1.00	-0.84
Temperature	0.33	0.33	0.09	-0.22	0.36	0.07	-0.09	0.42	-0.84	1.00

3.3 Feature combinations

The first stage involves finding the best feature combinations using the AdjcorT feature selection method. Figure 4 shows the ranking of feature importance, where higher values indicate greater significance of the variable to the target variable ($PM2.5_{D+1}$). According to the table, particulate matter (PM2.5 and PM10) are the most important variables for predicting PM2.5 in Klang, with AdjcorT values of 7.7 and 7.5, respectively, followed by relative humidity, SO_2, wind direction, O_3, CO, ambient temperature and NO_2. Additionally, wind speed is the least important feature for classifying PM2.5 in Klang. The features were then added to the ANN model one by one according to their ranking as shown in Figure 4 to determine the best feature combinations, as suggested by Arafin et al. (2024). The learning rate is set at 0.01 and the number of hidden nodes is determined by summing the number of variables and classes, dividing the result by two, and then adding one (Ul Saufie et al., 2022). Thus, this study use number of hidden nodes is 7. Table 6 presents the ANN model performances with varying numbers of features, based on accuracy, sensitivity, specificity, precision, F1 score, and AUROC. The highest value for each performance metric is typed in bold font. According to the table, the model with nine features achieves the best performance, with higher accuracy (0.67), sensitivity (0.85), F1 score (0.7), and AUROC (0.68). Arafin et al. (2024) concluded that eight features are sufficient to predict next-day PM2.5 concentrations in the urban area of Shah Alam. In contrast, this study found that nine features are needed to predict PM2.5 concentrations. The optimal feature combination for classifying $PM2.5_{D+1}$ in Klang includes PM2.5, PM10, relative humidity, SO_2, wind direction, O_3, CO, ambient temperature, and NO_2, based on AdjcorT value ranking.

3.4 Best model identification

In this section, the performance of the RBFNN model with all 10 variables is compared to the two-stage AdjcorT-RBFNN model, which utilizes the 9 best feature combinations. The RBFNN model with all

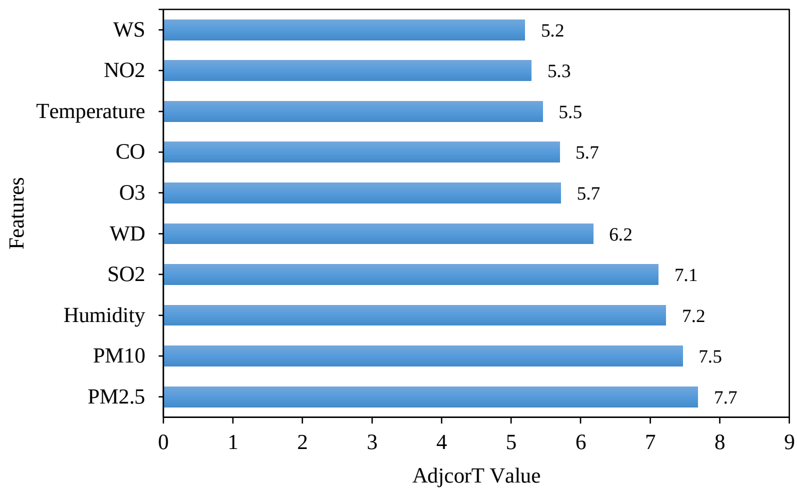

Figure 4. Feature ranking by AdjcorT value

Table 6. Model Performances for different numbers of features combination

No of features	1	2	3	4	5	6	7	8	9	10
Accuracy	0.58	0.55	0.61	0.59	0.63	0.66	0.67	0.65	**0.67**	0.67
Sensitivity	0.60	0.66	0.63	0.60	0.61	0.67	0.72	0.69	**0.85**	0.69
Specificity	0.55	0.51	0.59	0.57	**0.66**	0.65	0.62	0.62	0.60	0.64
Precision	0.55	0.51	0.59	0.57	**0.66**	0.65	0.62	0.62	0.60	0.64
F1 score	0.58	0.58	0.61	0.58	0.63	0.66	0.67	0.65	**0.70**	0.66
AUROC	0.58	0.56	0.61	0.58	0.62	0.66	0.67	0.65	**0.68**	0.67

10 variables represents the standard approach, while the AdjcorT-RBFNN model applies the AdjcorT method to mitigate multicollinearity by selecting the 9 most relevant features. Based on the results in Table 6, wind speed was excluded in the AdjcorT-RBFNN model. The number of hidden nodes is 7 for both models, RBFNN and AdjcorT-RBFNN. In addition, the learning rate for both models are set to 0.01. Table 7 presents a comparison of performance metrics for both models, with the highest values highlighted in bold font. According to the table, the two-stage AdjcorT-RBFNN model outperforms the RBFNN model, achieving higher accuracy (0.62), sensitivity (0.64), specificity (0.60), precision (0.60), F1 score (0.62), and AUROC (0.62). This finding is consistent with the research conducted by Arafin et al. (2024), which demonstrates that the AdjcorT-RBFNN model can enhance the performance of the RBFNN model. However, their study excludes relative humidity and ambient temperature as an important feature for classifying $PM2.5_{D+1}$ in Shah Alam. In contrast, our study found that both meteorological parameter, which is relative humidity and ambient temperature are important factor to predict PM2.5. The differences in the selection of the best features may be due to the different study areas, as our study was conducted in a Klang, while theirs was conducted in Shah Alam.

Table 7. RBFNN and AdjcorT-RBFNN model performances

Model	RBFNN	AdjcorT-RBFNN
Accuracy	0.59	**0.62**
Sensitivity	0.61	**0.64**
Specificity	0.56	**0.60**
Precision	0.56	**0.60**
F1 Score	0.58	**0.62**
AUROC	0.59	**0.62**

3.5 Monte Carlo simulations

A Monte Carlo simulation was applied to verify the best model, AdjcorT-RBFNN, using simulated data. The simulations of both models were run using various scenarios, with different sample sizes (N=50, 100, 150, 200) and correlations (ρ=-0.8, -0.5, -0.2, 0, 0.2, 0.5, 0.8). The line charts of accuracy, sensitivity, specificity, precision, F1 score, and AUROC for both models are shown in Figures 5, 6, 7, 8, 9, and 10, respectively. Based on Figure 5, the accuracy of the AdjcorT-RBFNN model is highest with strong negative correlation (ρ=-0.8) across all sample sizes. The accuracy decreases as correlation weakens, but it gradually improves with positive correlation. However, it doesn't reach the high levels seen with negative correlations. Additionally, larger sample sizes, particularly N=150 and N=200, result in better accuracy. The RBFNN model exhibits less variation across correlation levels, consistently achieving lower accuracy than AdjcorT-RBFNN. Under strong positive correlation (ρ=0.8) and N=200, RBFNN attains 50.9% accuracy, whereas AdjcorT-RBFNN reaches 60.3%, demonstrating its superior ability to select important features. Figure 6 demonstrates that the AdjcorT-RBFNN model's sensitivity is high with strong negative correlation and decreases as correlation weakens, improving with positive correlation but not returning to initial levels. Larger sample sizes improve sensitivity, especially for N=150 and N=200. In contrast, the RBFNN model's sensitivity remains consistent and high across all correlation levels, while AdjcorT-RBFNN's sensitivity fluctuates with changes in correlation and sample size.

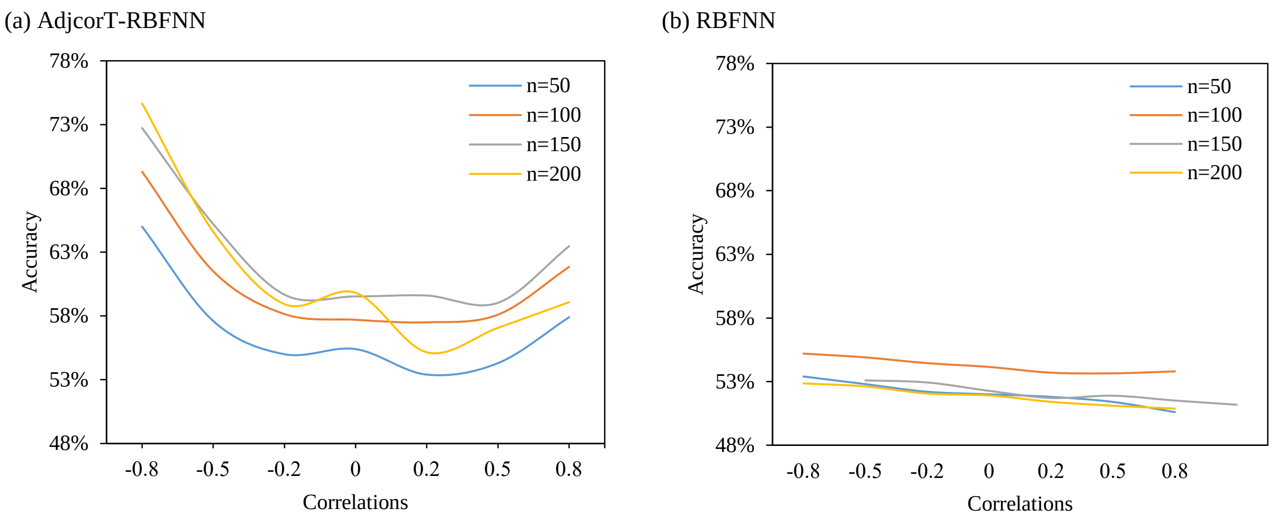

Figure 5. Accuracy of simulation AdjcorT-RBFNN and RBFNN model

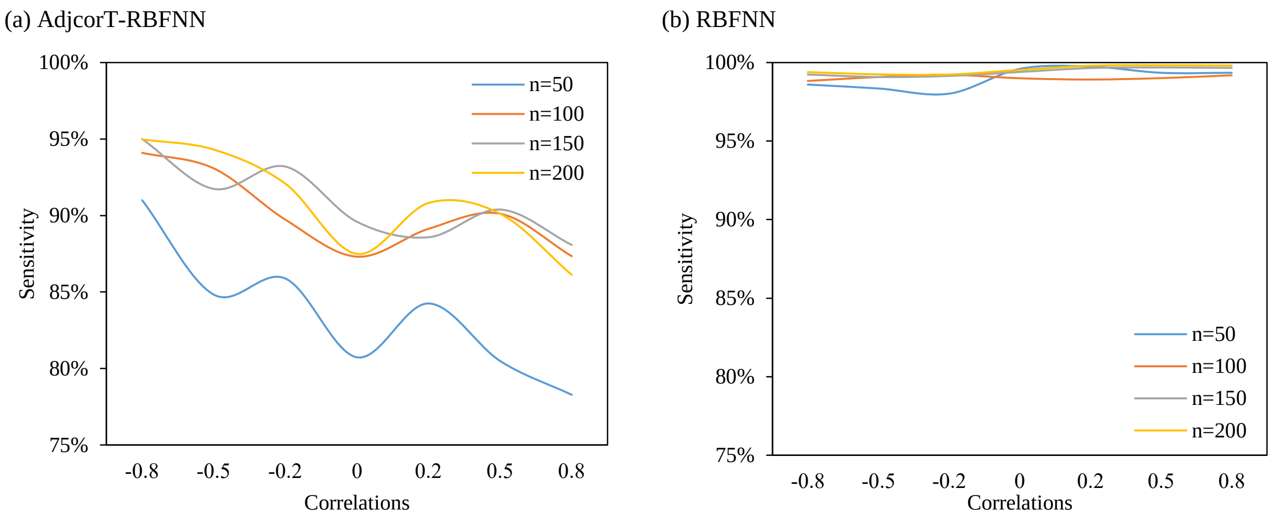

Figure 6. Sensitivity of simulation AdjcorT-RBFNN and RBFNN model

Figure 7 illustrates specificity, showing low values for both models, especially for RBFNN (below 11%) and AdjcorT-RBFNN (21-52%). The AdjcorT-RBFNN model performs better at correctly identifying negative cases, particularly with negative correlations and larger sample sizes. Figure 8 shows the precision of the simulations for both models. According to the line charts, AdjcorT-RBFNN performs better, especially with strong correlations, while RBFNN consistently shows lower precision with minimal variation. The AdjcorT-RBFNN model is particularly effective at reducing false positives among predicted positive cases, especially with stronger correlations and larger sample sizes. Figure 9 compares F1 scores, showing that AdjcorT-RBFNN performs better with larger sample sizes and stronger correlations, especially for negative correlations. In contrast, RBFNN maintain a stable F1 scores between 64% and 70% across all conditions. Lastly, Figure 10 compares AUROC values. The AdjcorT-RBFNN model shows varying AUROC depending on the correlation, with the highest value (74%) for strong negative correlation (ρ=-0.8) and N=200. As correlation weakens, AUROC decreases. RBFNN's AUROC remains between 50% and 55%, suggesting that the model has limited ability to distinguish between classes. To sum up, the AdjcorT-RBFNN model outperforms RBFNN in discriminating between classes, particularly with strong correlations, as demonstrated by simulated data. Similarly, Ibrahim (2020) highlighted that the AdjcorT provides a flexible variable selection approach for classification, particularly in medium to large datasets with negative correlations.

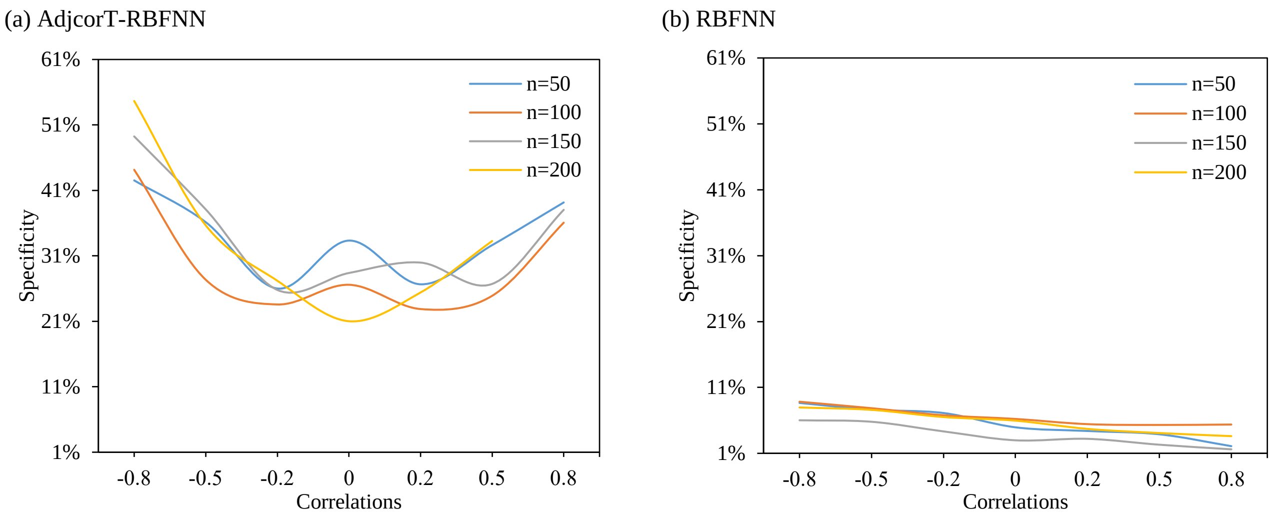

Figure 7. Specificity of simulation AdjcorT-RBFNN and RBFNN model

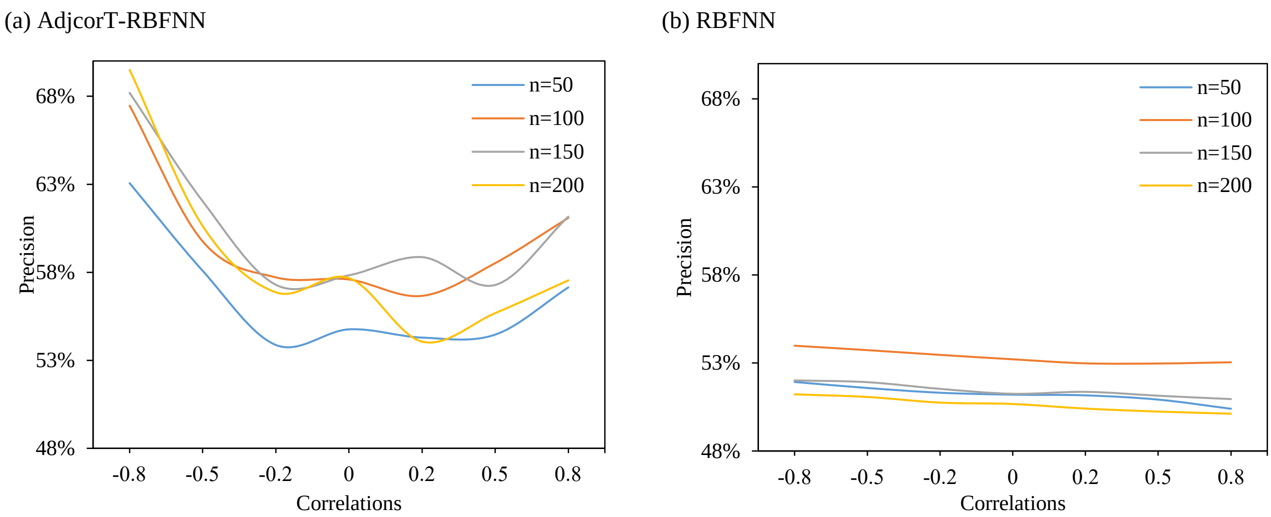

Figure 8. Precision of simulation AdjcorT-RBFNN and RBFNN model

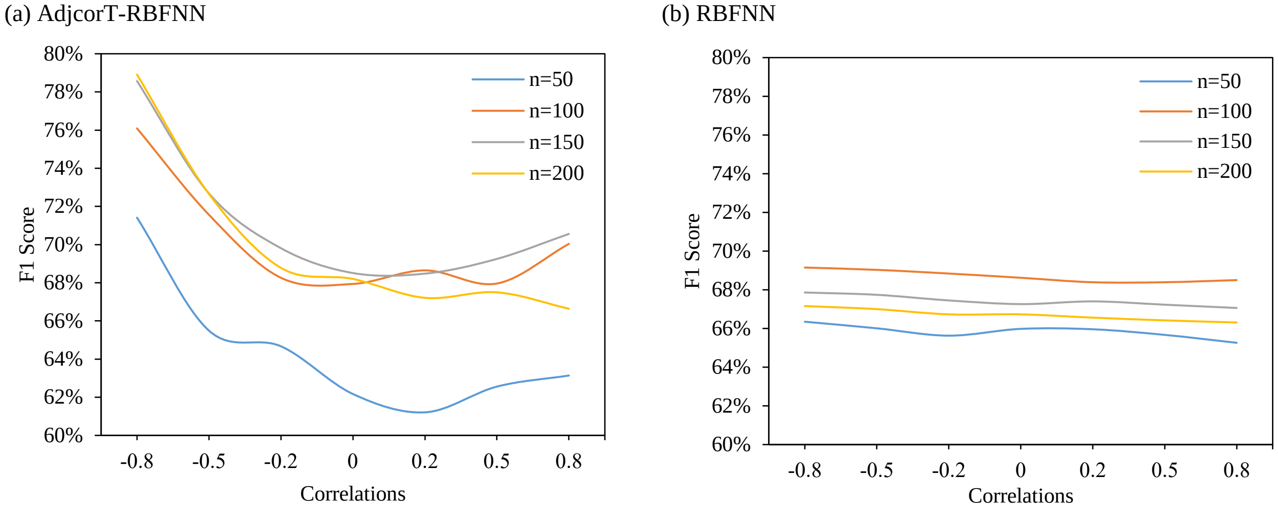

Figure 9. F1 Score of simulation AdjcorT-RBFNN and RBFNN model

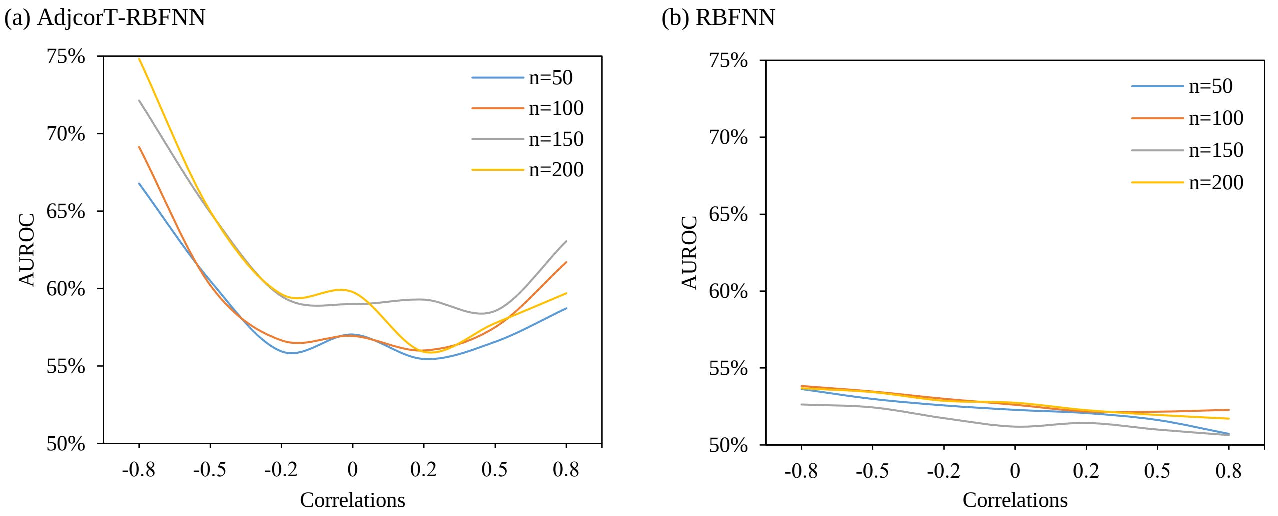

Figure 10. AUROC of Simulation AdjcorT-RBFNN and RBFNN Model

4. CONCLUSION

This study aims to classify air quality in Klang, Selangor while considering the high correlation between features using the two-stages feature selection method, AdjcorT-RBFNN. This study found that the AdjcorT-RBFNN model outperformed the RBFNN model, achieving higher performance metrics, including accuracy, sensitivity, specificity, precision, F1 score, and AUROC. Specifically, the AdjcorT-RBFNN model achieved an accuracy of 0.62, a sensitivity of 0.64, a specificity of 0.60, a precision of 0.60, an F1 score of 0.62, and an AUROC of 0.62, which were consistently higher than those of the standard RBFNN model. Based on the AdjcorT method, 9 features were identified as the best feature combination to predict air quality in Klang, namely PM2.5, PM10, relative humidity, SO_2, wind direction, O_3, CO, ambient temperature and NO_2. These features were selected based on their importance ranking, ensuring that only the most relevant predictors were retained while reducing redundancy caused by multicollinearity.

Moreover, this study verified the AdjcorT-RBFNN model's performance using simulated data. Based on the simulation results, the findings demonstrate that the AdjcorT-RBFNN model consistently outperforms the RBFNN model in distinguishing between classes, particularly when there are strong positive and negative correlations between the variables. When the correlation was strong (ρ=-0.8), the AdjcorT-RBFNN model achieved the highest accuracy, especially for larger sample sizes (N=150 and N=200). In contrast, the RBFNN model exhibited lower accuracy across all correlation levels, with less variation. Specifically, when ρ=0.8 and N=200, AdjcorT-RBFNN achieved an accuracy of

60.3%, outperforming RBFNN, which only reached 50.9%. Furthermore, AUROC values showed that AdjcorT-RBFNN was most effective under strong negative correlations, reaching 74% when ρ=-0.8 and N=200, whereas RBFNN's AUROC remained between 50% and 55% across all conditions. The AdjcorT-RBFNN model's ability to select relevant features based on correlation strength allows it to better handle complex data relationships, resulting in improved performance in terms of accuracy, sensitivity, and other key metrics. In contrast, the RBFNN model shows a more limited ability to differentiate between classes, as it lacks a dedicated feature selection mechanism to address multicollinearity. These results further highlight the importance of an effective feature selection method in improving model performance, especially when the dataset exhibits high multicollinearity.

The two-stage feature selection method, AdjcorT-RBFNN, has been shown to enhance RBFNN classification by considering the high correlation between features, using both real and simulated datasets. However, this study limited to air quality data in Klang an urban area. Therefore, we suggest future researchers apply this method to air quality data in other urban, suburban or rural areas to confirm its effectiveness. Moreover, due to the compromised results of the simulation, we also suggest future researchers apply this two-stage feature selection method in other areas where multicollinearity issues exist.

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AUTHOR CONTRIBUTIONS

Author 1 carried out the experiment and prepared the manuscript content, Author 2, Author 3 and Author 4 verified the manuscript content and Author 4 also supervised the project.

DECLARATION OF COMPETING INTERESTS

No conflict of interest concerning this manuscript.

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Sustainable Management of Chlorine Consumption in an Outdoor Swimming Pool: A Case Study of the Silpakorn University Swimming Pool

Umarat Santisukkasaem*

Department of Environmental Science, Faculty of Science, Silpakorn University, Nakhon Pathom, Thailand

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*** Corresponding author:**
E-mail:
santisukkasaem_u@su.ac.th

ABSTRACT

The general daily maintenance of outdoor swimming pools includes the addition of chlorine for disinfection. Chlorine is a potentially hazardous chemical that is harmful to users, and its excessive addition could lead to health effects in swimmers while insufficient levels may result in inadequate disinfection. This study aimed to optimize chlorine management at the Silpakorn University swimming pool by analyzing the physical and chemical characteristics of outdoor pool water. Initial sampling revealed an unacceptably high residual chlorine concentration of 20 mg/L, exceeding regulatory standards. To address this, a chlorine management strategy was implemented. Chlorine adjustment was conducted by measuring the residual chlorine concentration and calculating the chlorine demand. Post-intervention results indicated that residual chlorine and pH levels were successfully brought within acceptable limits. Further analysis confirmed that parameters such as hardness, ammonia nitrogen, nitrite nitrogen, nitrate nitrogen, suspended and dissolved solids, and total and fecal coliform bacteria met safety standards. Recommendations were given to the pool caretaker, including the use of personal protective equipment (PPE) while handling chlorine, a precise measurement of chlorine (1 kg daily), and regular filtration tank maintenance at least twice a month. Besides improving social and environmental aspects, the optimized chlorine usage resulted in an estimated annual cost saving of $1,213.26 (1 USD ≈ 36.065 THB). This study highlights the importance of sustainable chlorine management in swimming pools, offering a practical approach that can be replicated in similar facilities.

1. INTRODUCTION

Swimming is a sport that people enjoy because it is relaxing and involves using every part of the body. Disinfection is the top priority in controlling the water quality of swimming pools. Chlorine is the most popular disinfectant used to kill germs and bacteria because it is easily available, inexpensive, easy to use, and, most importantly, very effective. However, chlorine, after mixing with pool waters, human excretions, and personal care products, has the potential to form genotoxic and carcinogenic disinfection byproducts (DBPs) (Manasfi et al., 2017). DBPs include trihalomethanes, which can exert health effects on the respiratory system. Shan et al. (2023) studied the toxicity of diethylamino hydroxybenzoyl hexyl benzoate (DHHB), a chemical that poses a major health risk, in swimming pools. They concluded that the source of the detected DHHB was the sunscreens and other personal care products worn by swimmers that were mainly oxidized by the free chlorine in swimming pools. The latest research in Thailand on swimming pools evaluated chloroform and its related health risks. Outdoor pools were found to have the highest average chloroform concentration of 63.89±12.76 µg/L, leading to an unacceptable risk for lifetime cancer in adult male and female groups (Laohapongsomboon et al., 2025).

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Therefore, several studies have focused on chlorine byproducts, specifically, their formation, exposure, and health impacts on pool users. Most of these studies were conducted on indoor swimming pools (Carter and Joll, 2017; Yang et al., 2018; Lempart et al., 2020; Abilleira et al., 2023; Zhang et al., 2023a; Zhang et al., 2023b; Peng et al., 2024). Given that chlorine is the precursor of DBPs, reducing chlorine levels will decrease DBP levels. A future study need is to minimize DBP formation (Chowdhury et al., 2014). Goma et al. (2017) proposed a new method to reduce chlorine oxidant derivatives by (1) replacing hydrochloric acid, the chemical used to reduce pH, with carbon dioxide; (2) using a low-concentration salt electrolysis system to produce hypochlorous acid ($HOCl^-$) to enhance sodium hypochlorite addition; and (3) introducing ultraviolet radiation to degrade chloramines. Studies on the application of peracetic acid (PAA) as an alternative disinfectant in outdoor swimming pools concluded that compared with chlorine, PAA is likely to produce less DBPs but has a higher price (Jia et al., 2023; Lin and Lin, 2024). Existing standards for chlorine concentrations in pool water are inconsistent and depend on the authorities of each country. For example, Dallolio et al. (2013) summarized the physical-chemical standards for swimming pool water in some European countries without considering if a pool was an indoor or outdoor pool. They found that residual chlorine values varied from 0.3 mg/L to 8.2 mg/L, and none of the countries specified the same value. The World Health Organization (WHO) recommended that the free chlorine concentration in public pools should not exceed 3 mg/L (WHO, 2006). The range of 0.6-1.0 mg/L was advised by the public health committee under the Department of Health, Ministry of Public Health of Thailand (2007) and enforced as a criterion of GREEN Health Hotel Standards in Thailand (Department of Health, 2023). Although no comprehensive legislation enforcing the amount of chlorine used exists, the free chlorine range should be set at the local level. Furthermore, considering that chlorine is inexpensive, its consumption has never been strictly controlled. Rosende et al. (2020) analyzed the cost-effectiveness of chlorine-based and unconventional disinfection products and concluded that the most affordable disinfectant agent in the pilot setup was sodium hypochlorite. Apart from the environmental and health impacts of excessive chlorine consumption, the highest expense in pool maintenance under life cycle assessment was found to be human resources (workers) and chlorine purchase (de Moura et al., 2023). Several studies emphasized chlorine consumption and transformation into DBPs, as well as alternative disinfectants, but none had pinpointed the priority of waste management: chlorine consumption reduction. This research gap exists because studies in general focused on the impacts and minimization, but not source reduction, of DBPs. Therefore, the novelty of this present study is the prioritization of the reduction of chlorine consumption to alleviate undesirable effects on sustainability. In contrast to the existing research that deployed reactive measures, which are considered as end-of-pipe treatments in environmental management, this study employed a proactive measure to avoid or prevent generating pollution at the source.

The Silpakorn University swimming pool is an outdoor pool located in the university area of the Sanamchandra Palace Campus, Nakhon Pathom, Thailand. In addition to the amount of chlorine consumed for disinfection, other factors -- such as temperature, rainfall amount, and contaminating particles, i.e., leaves and insects -- must be considered because they contribute to water quality. A preliminary investigation of the pool revealed the absence of a proper chlorine management system. The amount of chlorine added to the pool has never been properly controlled: A maintenance worker simply adds approximately 2 kg of trichloroisocyanuric acid and chlorine powder daily and adjusts the pH to neutral by adding either hydrochloric acid or basic solution. This situation is a typical issue, with researchers observing that in new checklists for swimming pool evaluation, data on chlorine use are not always recorded (Liguori et al., 2014). In the absence of a proper monitoring and recording system for chlorine consumption, the amounts of chlorine added and purchased strictly depend on the worker's experience. However, none of the customers reported health impacts, and the total cost and the amount of chlorine consumed are both negligible relative to those of other materials consumed in the university. The author would like to declare that this research was conducted only to observe the possibility of consumption reduction and identify its benefits. All findings here should not be considered offensive in any way.

The abovementioned issues lead to effects on environmental sustainability. Specifically, the overconsumption of chlorine creates unnecessary toxic pollutants. Their social impact can be seen in health effects experienced by pool users from swimming in water with excessive amounts of residual chlorine and the formation of potential DBPs, as well as those experienced by maintenance workers resulting from the inappropriate handling of chlorine. Finally, economic impact is the additional cost of chlorine purchase. All of the above issues are related. Therefore, in this research, chlorine addition was studied, and the data collected during pre- and postmanagement were considered to provide guidelines for the sustainable and safe handling of chlorine. The ultimate aim of this work is to support Sustainable Development Goal 12: Responsible Consumption and Production because reducing chlorine use can simply help achieve the sustainable management and efficient use of natural resources at the local level (Target 12.2, (UN, n.d.)), as well as promote social and economic benefits.

2. METHODOLOGY

2.1 Sampling plan and locations

The pool was investigated to collect physical and chemical data. A maintenance worker was interviewed regarding chlorine handling and related details, with a disclaimer asserting that the worker will not be held responsible for any issues that may arise from providing information given that the objective of the interview is entirely to gather relevant information. The physical features of the pool surroundings were surveyed. For chemical data, chlorine sampling (Table 1) lasted for five weeks. During chlorine sampling, grab sampling was conducted daily for seven days during the first week and weekly for four weeks (21 samples at all locations per day, yielding a total of 231 samples) and 12 random days in between weeks (1 sample per day at random locations, yielding a total of 12 samples) for a total of 22 times. In the sampling strategy, the samples collected on days 1-7 represented the baseline condition. The baseline data were used in planning the amount of chlorine added. Subsequently, weekly samplings (days 14, 21, 28, and 35) were conducted during the implementation phase to observe the changes in residual chlorine concentration. Lastly, 12 samples were randomly collected to ensure the efficiency of implementation. Other related parameters in pool water quality analysis, namely, hardness (in the form of $CaCO_3$), ammonia nitrogen (NH_3), nitrate nitrogen (NO_3^-), nitrite nitrogen (NO_2^-), total dissolved and suspended solids, and total and fecal coliforms, were analyzed by using the data of the samples collected on days 1 and 35 as the pre- and postimplementation data, respectively. Figure 1 illustrates the 21 sampling points that were identified to represent the water quality of the whole pool. Given that the pool bottom is sloped with varying depths, it was divided into three zones (A-C) along the length of the pool. Each zone was further divided into three levels with respect to the pool depth. The volume of water in the pool was calculated to identify the sample volume.

Table 1. Sampling plan

Day	Sampling frequency	No. of samples
1-7	Daily (7 days in a row)	147 (21 per day)
8-13	Daily (in between week)	3 (1 per random day/location)
14	Weekly (once a week)	21
15-20	Daily (in between week)	4 (1 per random day/location)
21	Weekly (once a week)	21
22-27	Daily (in between week)	2 (1 per random day/location)
28	Weekly (once a week)	21
29-34	Daily (in between week)	3 (1 per random day/location)
35	Weekly (once a week)	21

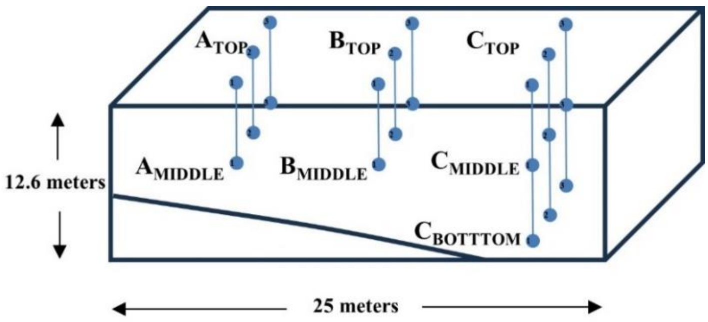

Figure 1. Sampling locations and dimensions of the pool

2.2 Sample analysis

The sample volume was approximately 2 L per sampling point. The samples were mainly analyzed for residual chlorine concentration. Remnants were combined and analyzed for other parameters. Triplicate analysis was conducted on every sample. The results were compared with Thai (Ministry of Public Health of Thailand (Public Health Committee, 2007) and international standards (WHO, 2006). The pH and temperature of the collected samples were measured with a pH meter and thermometer, respectively, and the samples were further analyzed for residual chlorine (iodometric titration) and chlorine demand (calculation). Analysis of variance (ANOVA) (α=0.05) was used to validate the homogeneity of the water throughout the pool. The difference in residual chlorine concentration was tested at three sampling points (number of samples: A=6, B=6, and C=9) on the first day (day 1) and last day (day 35) of the experiments. The additional parameters of hardness (EDTA titration), NH_3 (preliminary distillation step and titration), NO_2^- (diazotization), NO_3^- (diazotization and cadmium reduction), suspended and dissolved solids (gravimetric analysis), and bacteria (total coliform MPN test) were analyzed to ensure that the water quality complied with the standard. Given the limited number of samples (n=3 at each sampling point: A, B, and C), nonnormal distribution was assumed. The Wilcoxon test was therefore performed to verify the difference in the pre- and postcontrol data (α=0.05) for all additional parameters. All water sample analyses were conducted by following standard methods (APHA, 2017).

Chlorine adjustment was conducted simply by measuring the residual chlorine concentration and calculating the chlorine demand, which is the amount of chlorine addition required to provide sufficient residual chlorine to ensure disinfection capacity. Residual chlorine results from the chemical reaction of chlorine and organic materials. When chlorine oxidizes all organic materials, the residual chlorine then acts as a disinfectant. Therefore, the chlorine demand can be calculated by using the differences between the concentrations of the added and residual chlorine. The amount of chlorine adjustment was calculated to supply the residual chlorine concentration in accordance with the standard of the Ministry of Public Health of Thailand of 0.6-1.0 mg/L (Public Health Committee, 2007) and the WHO standard of less than 3.0 mg/L (WHO, 2006), which is the adequate concentration for disinfection.

3. RESULTS AND DISCUSSION

3.1 Preliminary experiments

Theoretically, the amount of added chlorine can be simply calculated from the pool area. However, the outdoor pool investigated in this work may contain interferences, such as rain, sunlight, and leaves, which might affect the amount of residual chlorine. An experiment must then be conducted to reassure that the amount of chlorine added is adequate. Given that the preliminary measurement indicated that the amount of residual chlorine was approximately 20 mg/L, which significantly exceeded the standard of 0.6-3.0 mg/L, experiments were conducted along with daily and weekly sampling to avoid whole-pool water replacement. The pH value reflects the hydrogen ion concentration in water and thus directly affects the structure of the residual free chlorine. A high pH is associated with low hypochlorite (OCl^-) ion levels, implying low disinfectant efficiency given that OCl^- is negatively charged and thus has the tendency to bond with the suspended solids in water and become inactive. However, some OCl^- ions are converted into hypochlorous acid ($HOCl^-$), which has a high disinfectant efficiency. Moreover, even though high temperatures result in high dissolved chlorine levels, an increase in water temperature causes a reduction in $HOCl^-$ and thus, in-efficiency. Temperature is therefore a concern for outdoor pools in Thailand. Nonetheless, it exerts a negligible impact on the pH. As shown in Figure 2, even when the temperature increased, the pH did not decrease. This situation indicates that the controlled amount of added chlorine worked at the typical ambient temperature, causing the pH to meet the standard values of 7.20-7.80 (WHO, 2006), which are not only safe for swimmers but also provide sufficient disinfectant capacity.

3.2 Control of chlorine addition

Given that the residual chlorine concentration exceeded the limit, the amount of chlorine added was controlled and the residual chlorine concentration was measured throughout the experiment. Figure 3 provides the residual chlorine concentration at every sampling point from days 1 to 35. The daily data showed that during the first week, the chlorine concentration gradually decreased from approximately 19 mg/L to 9 mg/L. The weekly data indicated a similar trend, and chlorine concentration remained constant within the range of 3-5 mg/L during the last weeks of the experiment. ANOVA verified that there is no difference among sampling locations A, B, and C. Therefore, the

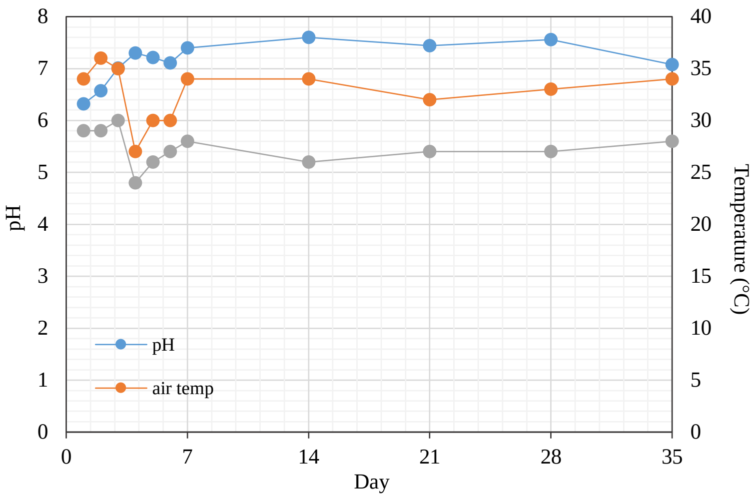

Figure 2. pH and temperature of the pool

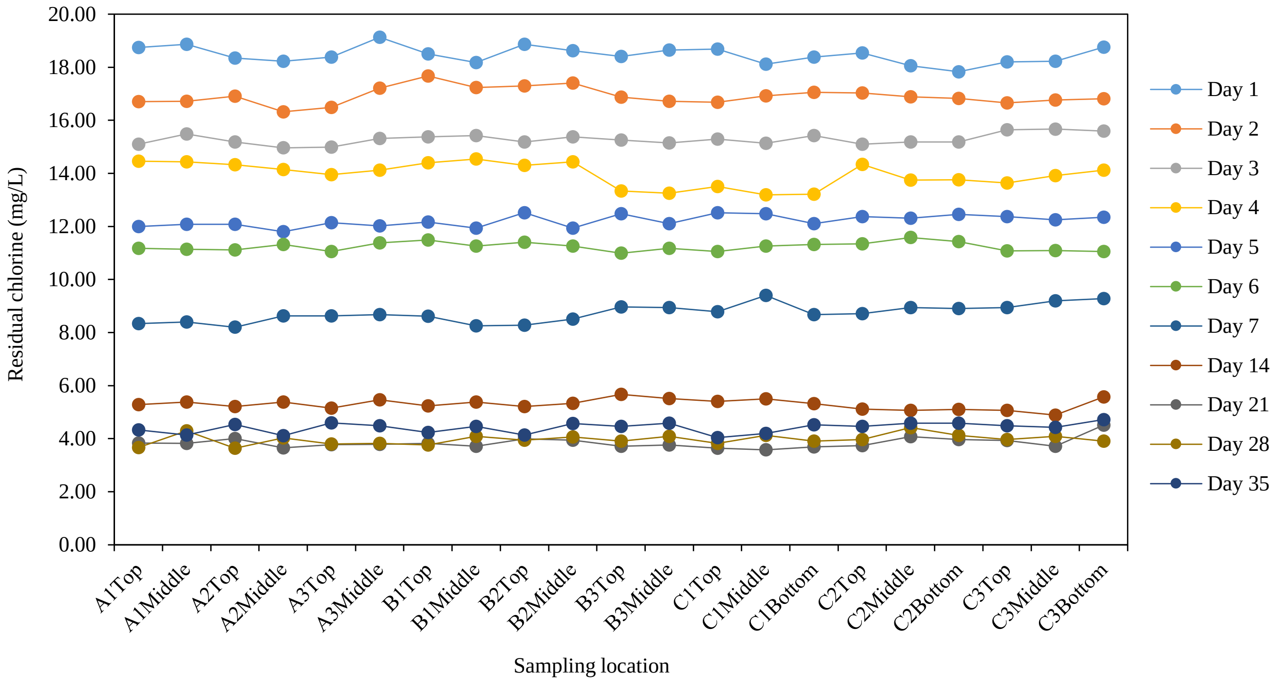

Figure 3. Residual chlorine concentration

pool water is assumed to have mixed homogeneously, enabling the diffusion of the added chlorine throughout the pool. Overall, the control provided a significant reduction in residual chlorine concentration. However, the residual chlorine concentration still exceeded the standard of the Ministry of Public Health of Thailand of 0.6-1.0 mg/L and the WHO standard of less than 3.0 mg/L due to the uncontrolled factors of precipitation; sunlight; temperature; dust particles, debris, and leaves that had fallen into the pool; and the number of pool users. These factors are the main limitations for operating outdoor pools, making pinpointing the exact amount of added chlorine inconvenient. This finding is also confirmed by the fluctuation of the residual chlorine concentration, which ranged from 3.01 mg/L to 7.61 mg/L. The data for the residual chlorine concentration were obtained from random sampling during the experiment. Therefore, the chlorine demand was calculated by using the residual chlorine data from the last four weeks, as shown in Figure 4. The average chlorine demand fell within the range of 1.15±0.39-3.52±0.10 mg/L. In the identification of the amount of chlorine added to provide sufficient disinfection while complying with the standard limit, the data from the day (day 28) that provided the optimum residual chlorine concentration (1.63-3.51 mg/L) that was closest to the standard were selected. The calculation conducted in accordance with the volume of pool water showed that the amount of added chlorine should be 1.10-2.30 kg. The addition of the minimum amount of 1.10 kg of chlorine resulted in the residual chlorine concentration of 1.63 mg/L. Therefore, in theory and practice, the addition of 1 kg of chlorine results in adequate disinfection. In addition, a study conducted on Northeastern Ethiopia outdoor swimming pools, which had similar characteristics as the pool in this study, i.e., similar ranges of temperature, pH, and residual chlorine concentrations, indicated that most pool water samples did not meet the WHO standard limit (Natnael et al., 2024). Therefore, the results of this study could possibly be applied to other swimming pools operated in a similar manner.

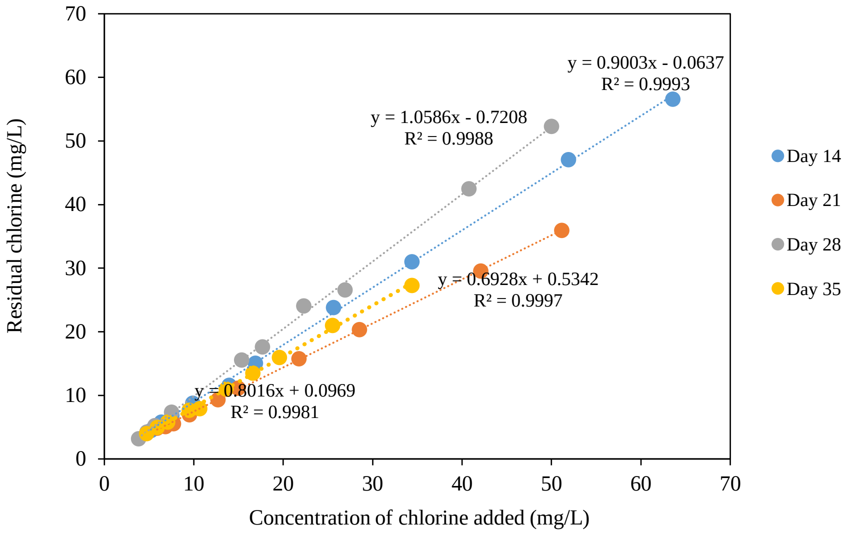

Figure 4. Relationship between residual chlorine and chlorine demand

3.3 Water quality analysis

Other related parameters were also measured to ensure the safety of pool users. All values fell within standard limits. The pre- and postimplementation data were sampled and analyzed (Figure 5) to confirm that reducing the amount of chlorine added would neither interfere with the disinfection efficiency nor create any toxic byproducts. Hardness is the amount of dissolved calcium (in the form of $CaCO_3$) in water. An excessively low hardness level can result in corrosion, causing in the tearing off of sealants, whereas an excessively high hardness level can cause precipitation, thus disturbing system operation. The levels of hardness (in the form of $CaCO_3$) reduced from an average value of 441 mg/L to 415 mg/L before and after the experiment, respectively. The values of hardness (in the form of $CaCO_3$) recommended by the Thai Public Health Committee (2007) are 250-600 mg/L. The average pre- and postexperimental NH_3 levels were 0.34 and 0.07 mg/L, respectively. The standard for NH_3 is less than 20 mg/L. NO_2^- had an average value of 0.0008 mg/L and was undetectable after treatment. The latter effect is typical because NO_2^- does not tend to accumulate in water but is rather converted into nitrate rapidly. The average NO_3^- also declined from 3.55 mg/L to 3.17 mg/L. The standard for NO_3 is less than 50 mg/L. NO_3 values were distributed equally in each zone, indicating that the water had mixed homogeneously throughout the pool. Total suspended solids (TSS) decreased from 2.10 mg/L to nondetectable levels (no standard limit), whereas the total dissolved solids (TDS) nonsignificantly increased (≈18.11%) from 836.28 mg/L to 1,021.28 mg/L without exceeding the standard of 1,000-2,000 mg/L. The last but most important parameters are total coliform bacteria and fecal coliform bacteria, which were confirmed to be both negative. In addition, the Wilcoxon test validated the absence of a statistically significant difference between the pre- and postexperimental concentrations in this work. The data on NO_2^- and TSS were excluded due to the lack of detectable data. Therefore, although the amount of chlorine added was reduced by half, the disinfection efficiency remained the same.

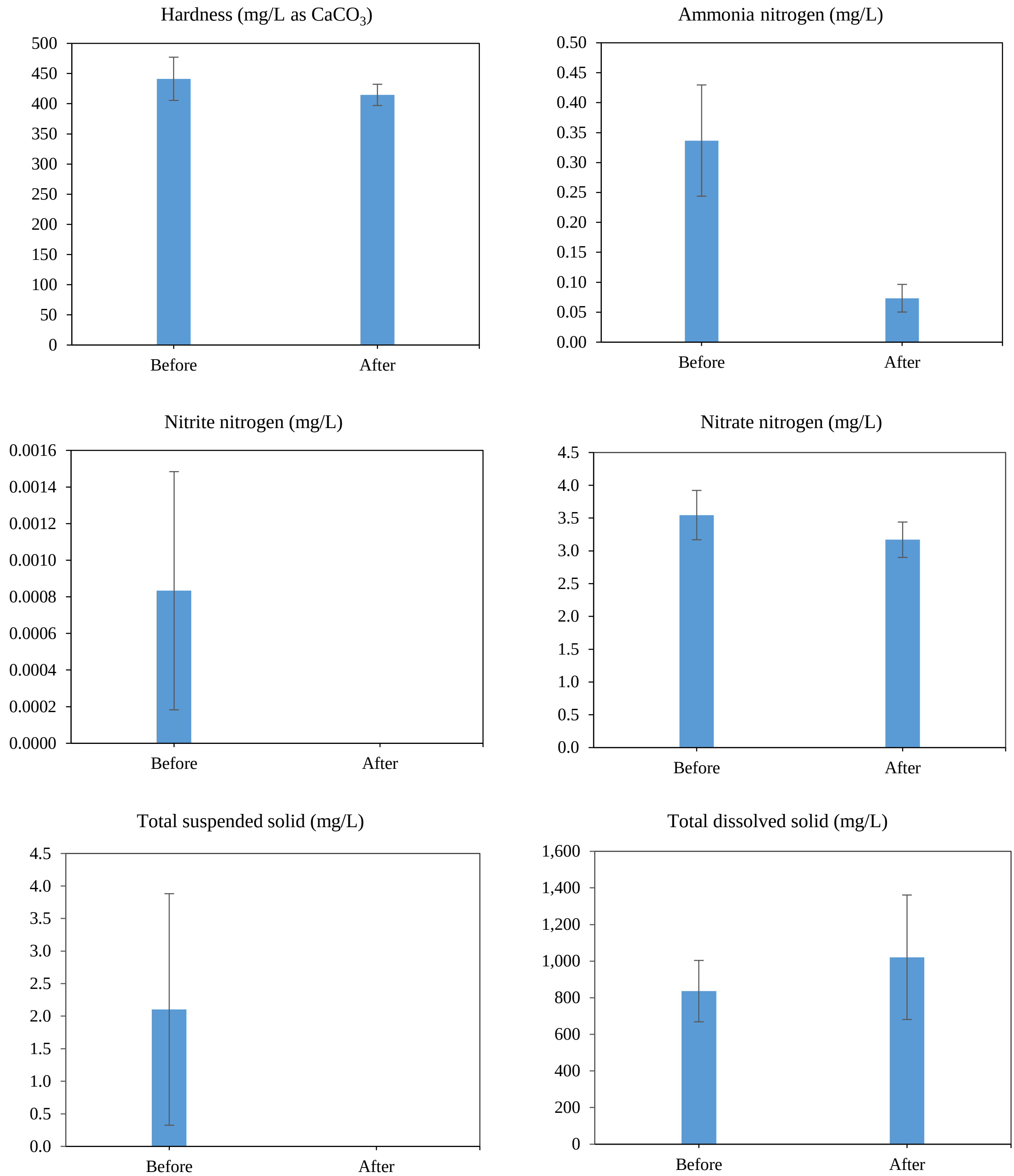

Figure 5. Pre- and postexperimental data on pool water quality

4. CONCLUSION

The experimental results showed that reducing chlorine consumption yielded a similar disinfection efficiency. This research technically supports the practices of SDG12: Sustainable Consumption and Production and partly supports Subtarget 3.9 (Mortality from Environmental Pollution) of SDG3: Good Health and Well-being. The avoidance of excessive chlorine can contribute to sustainability as follows: From an environmental perspective, the values of all related parameters comply with the standard. The underlying point is that the amount of chlorine added is reduced by 50%, i.e., from 2 kg to 1 kg daily. Even though chlorine is not a greenhouse gas, reducing chemical consumption is the first principle in environmental management and circular economy because it can decrease natural resource consumption and prevent pollution throughout the chlorine life cycle. From an economic perspective, reducing chlorine consumption by half has monetary value because using only 1 kg of chlorine daily is equivalent to a reduction of 365 kg of chlorine yearly, resulting in annual cost savings of approximately $1,213.26 (1 USD ≈ 36.065 THB). This figure was calculated by using an average cost of chlorine of $3.33 per kg and a preimplementation consumption rate of 2 kg per day. Therefore, adding 2 kg of chlorine per day for 365 days per year results in the consumption of 730 kg of chlorine or cost of $2,430.90 per year. As mentioned previously, the postimplementation consumption of chlorine reduced by half. The above values may slightly vary due to currency conversion. From a social perspective, excess chlorine can cause damage to the health of pool caretakers and swimmers. Therefore, reducing chlorine additions decreases exposure risk. The results of this research can act as baseline information in the formulation of local policies related to sustainable environmental management and can be further utilized by any corporation with outdoor swimming pools operated in a similar manner. Nevertheless, this work has some limitations, and the following possible future recommendations should be considered: (1) the long-term microbial risks due to the reduced amount of chlorine added should be monitored; (2) biotic and abiotic interferences, such as rain, sunlight, and pool user behavior, should be clearly identified and studied thoroughly; (3) applicability to other pools under different climatic conditions or different pool types should be explored; and (4) potential hidden costs, such as increased maintenance time and worker behavioral changes, should be observed.

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Assessing Spatial-Temporal Patterns of Agricultural Drought Vulnerability and Its Impacts on Economic Crops, Nakhon Ratchasima, Thailand

Suwit Ongsomwang*

School of Mathematics and Geoinformatics, Institute of Science, Suranaree University of Technology, Nakhon Ratchasima 3000, Thailand

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*** Corresponding author:**
E-mail: suwit@sut.ac.th

ABSTRACT

Thailand frequently suffers from rainfall shortages and ensuing droughts and the northeast region is especially vulnerable. The effects of climate change on water resources are further directly related to agricultural drought vulnerability. The objectives of the study were (1) to assess the spatial and temporal patterns of agricultural drought vulnerability based on agricultural drought exposure, agricultural drought sensitivity, and adaptive capacity and (2) to assess the potential impact of agricultural drought vulnerability on economic crops. To do so, this study integrated drought exposure, drought sensitivity and adaptive capacity for assessing spatial and temporal patterns of agricultural drought vulnerability and their potential impacts on economic crops at both the district and subdistrict levels in Thailand's northeastern Nakhon Ratchasima Province. Our results showed that the spatial and temporal patterns of agricultural drought vulnerability in two periods (6m10 and 12m) varied from one region to another. Levels of severity were established, and moderate, high and very high levels were found in 10 districts and 96 subdistricts in the 6m10 period (May to October). They further occurred in 17 districts and 166 subdistricts in the 12m period (January to December). Districts and subdistricts with identical potential impact in both periods included 3 districts and 48 subdistricts. The potential impact of agricultural drought vulnerability on economic crops further was higher in the 12m duration than for 6m10. The highest potential impact was found to be on cassava (2023). In conclusion, the results of the study can be used as basic information for government agencies to monitor and mitigate agricultural drought in Nakhon Ratchasima. The government should further consider implementing a feasibility study for groundwater use in local agriculture, to better mitigate the impact of drought on important vulnerable economic crops.

1. INTRODUCTION

Drought is a complex phenomenon because of its unpredictable start and end, the length of the event, as well as its nonspecific spatial extent (or geographic reach) and both uncertain frequency and intensity. Gordon (1992) stated that drought lacks straightforward entry, duration, and termination points, making it challenging to analyze. According to the United Nations Convention to Combat Desertification (UNCCD) drought has multiple negative impacts, even within a single domain (2023). Drought most severely impacts ecosystems with homogeneous vegetation, which are most susceptible, and especially so under long-term conditions (Ding et al., 2020). The impacts of drought are intensified by their diverse effects across different sectors, including rivers, watercourses, agriculture, electricity production, and industry - leading to significant implications for gross domestic product and international welfare (Begum et al., 2022).

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Thailand frequently suffers from droughts due to rainfall shortages, as well as reduced flow in surface and sub-surface rivers, and poor land management practices. The entire country was affected by severe droughts in 1979, 1994, and 1999 - and the northeastern region, which has the highest poverty rates, remains particularly vulnerable (World Bank Group, 2023).

Conceptually, vulnerability is a relative measure, and it indicates the degree to which a system is susceptible to damage (harm) due to the occurrence of an event (Smit et al., 1999). Most of the definitions of vulnerability originate from two approaches: climate change adaptation (CCA) and disaster reduction risk (DRR) (González Tánago et al., 2016). Many researchers have conducted vulnerability assessments related to the effect of climate change according to the CCA approach (Chandrasekar et al., 2009; Fontaine and Steinemann, 2009; Antwi-Agyei et al., 2012; De Stefano et al., 2015; Murthy et al., 2015; Dabanli, 2018; Hoque et al., 2021; Gao et al., 2023; Mulyanti et al., 2023; Babel et al., 2024; Senapati and Das, 2024).

Therefore, the framework of Fontaine and Steinemann (2009), including drought exposure, drought sensitivity, and adaptive capacity, was here applied to assess agricultural drought vulnerability using geospatial analysis. The specific objectives of the study were (1) to assess the spatial and temporal patterns of agricultural drought vulnerability based on agricultural drought exposure, agricultural drought sensitivity, and adaptive capacity and (2) to assess the potential impact of agricultural drought vulnerability on economic crops.

2. METHODOLOGY

2.1 Study area

The study area is Nakhon Ratchasima Province, which covers 20,729 km^2 (Figure 1). There are 12 Sub-Basins in the study area.

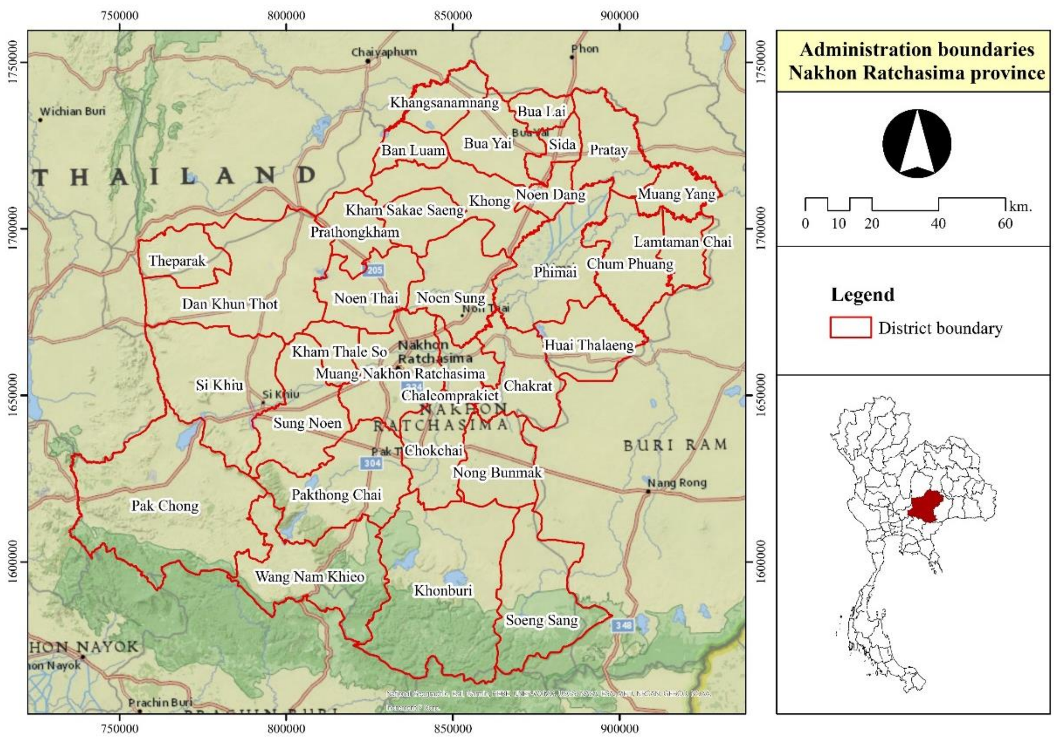

Figure 1. Location map of the study area

2.2 Data

Data collected and prepared for assessing agricultural drought vulnerability included agricultural drought exposure, agricultural drought sensitivity and adaptive capacity components, all summarized below.

- Rainfall data from 1975 to 2022 from 37 stations for SPI calculation
- MOD31A-NDVI product from 2002 to 2022 for VCI extraction
- MOD11B-LST product from 2002 to 2022 for LST extraction

- SRTM DEM for landform and elevation extraction
- Land use data in 2008, 2011, 2015, 2017, 2019, and 2023 for land use extraction
- Soil series for soil drainage extraction
- Agricultural irrigation area
- Waterbody data from 2023 for Euclidean distance calculation
- River network and sub-basin boundary for drainage density calculation
- Average rice harvested area from 2011 to 2023
- Number of rice farmer households in 2023
- Population data in 2023
- Potential groundwater yield

Figure 2 displays the workflow of the research methodology. Brief information is summarized below.

2.3 Assessing agricultural drought exposure (ADE)

ADE was assessed by combining the meteorological drought frequency (MDF) and meteorological drought intensity (MDI) indices of the 2 given periods (6m10 and 12m) using a multiplication operation. For the MDF index, monthly rainfall data (1975-2022) from 37 stations were used to calculate the standardized precipitation index (SPI) in the 2 periods. This was done using Equation 1, with four drought levels: near normal drought (NND), moderate drought (MD), severe drought (SD) and extreme drought (ED), as suggested by McKee et al. (1993) (Table 1).

$$SPI_i = (X_i - X_{mean})/\sigma \quad (1)$$

Where; X_i is standardized rainfall of a given station for period i; X_{mean} and σ are the long-term mean and standard deviation of standardized rainfall for the same period.

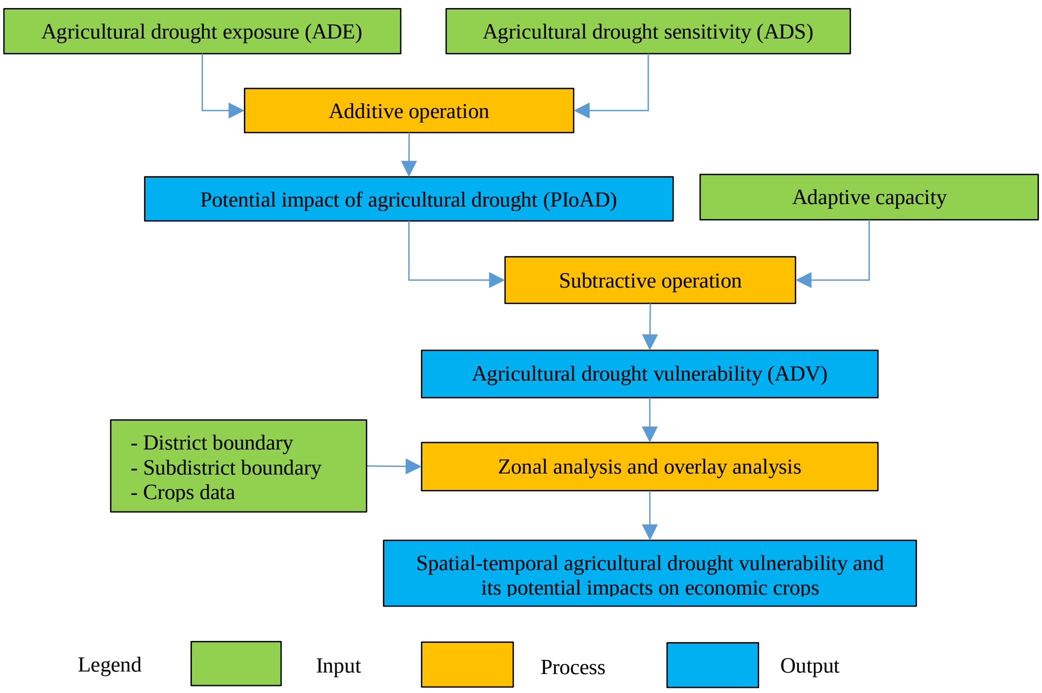

Figure 2. Research methodology workflow

Table 1. Drought classification based on SPI

Level of drought	SPI value	Weights
Near-normal drought (NND)	0 to -0.99	1
Moderate drought (MD)	-1.00 to -1.49	2
Severe drought (SD)	-1.50 to -1.99	3
Extreme drought (ED)	-2.00 and less	4

Thereafter, the probability of drought occurrence (DOc) at each station for each SPI period (6m10 and 12m) and drought level (NND, MD, SD, and ED) was calculated by taking the ratio between counting number in each drought level to the total number of years (48 years) (Sönmez et al., 2005). The

DOc values of 73 stations for each SPI period in each drought level were separately interpolated to create continuous surface data using the Inverse Distance Weighted (IDW) method, as suggested by Tadesse et al. (2010). Later, the interpolated probability of DOc for each period and for each drought level was reclassified into four levels: low, moderate, high, and very high, using the natural break (NB) method, and by assigning ratings for each level (with a value of 1, 2, 3, and 4, respectively). The MDF index of each period, with an accompanying drought severity category, was integrated using Simple Additive Weighting (SAW) (Kaliszewski and Podkopaev, 2016) as:

$$\mathrm{MDFI_{Met}} = (\mathrm{NND_r} \times \mathrm{NND_w}) + (\mathrm{MD_r} \times \mathrm{MD_w}) + (\mathrm{SD_r} \times \mathrm{SD_w}) + (\mathrm{ED_r} \times \mathrm{ED_w}) \quad (2)$$

Where; $MDFI_{Met}$ is the meteorological drought frequency index of a specific time, NND_r is the rating of NND occurrence, NND_w is the weight of NND occurrence, MD_r is the rating of MD occurrence, MD_w is the weight of MD occurrence, SD_r is the rating of SD occurrence, SD_w is the weight of SD occurrence, ED_r is the rating of ED occurrence, ED_w is the weight of ED occurrence.

Meanwhile, the MDI index is considered to be the degree of the precipitation deficiency and the severity of drought measurement (as determined by SPI, when it is less than -1 and equals -1 for the specific period). The MDI index of each period at each station was extracted based on SPI values less than or equal to -1 (Sehgal and Dhakar, 2016) and the extracted MDI values for each period at the 37 stations were separately interpolated to create the MDI index using the IDW method.

Finally, the MDF and MDI indices were combined by multiplication for the MDE index and reclassified into five MDE severity levels (very low, low, moderate, high, and very high) using the NB method. (See the workflow of MDE in Figure 3). Spatial distribution of the MDF, MDI and MDE indices for MDE classification in 2 periods (6m10 and 12m) are displayed in Figure 4.

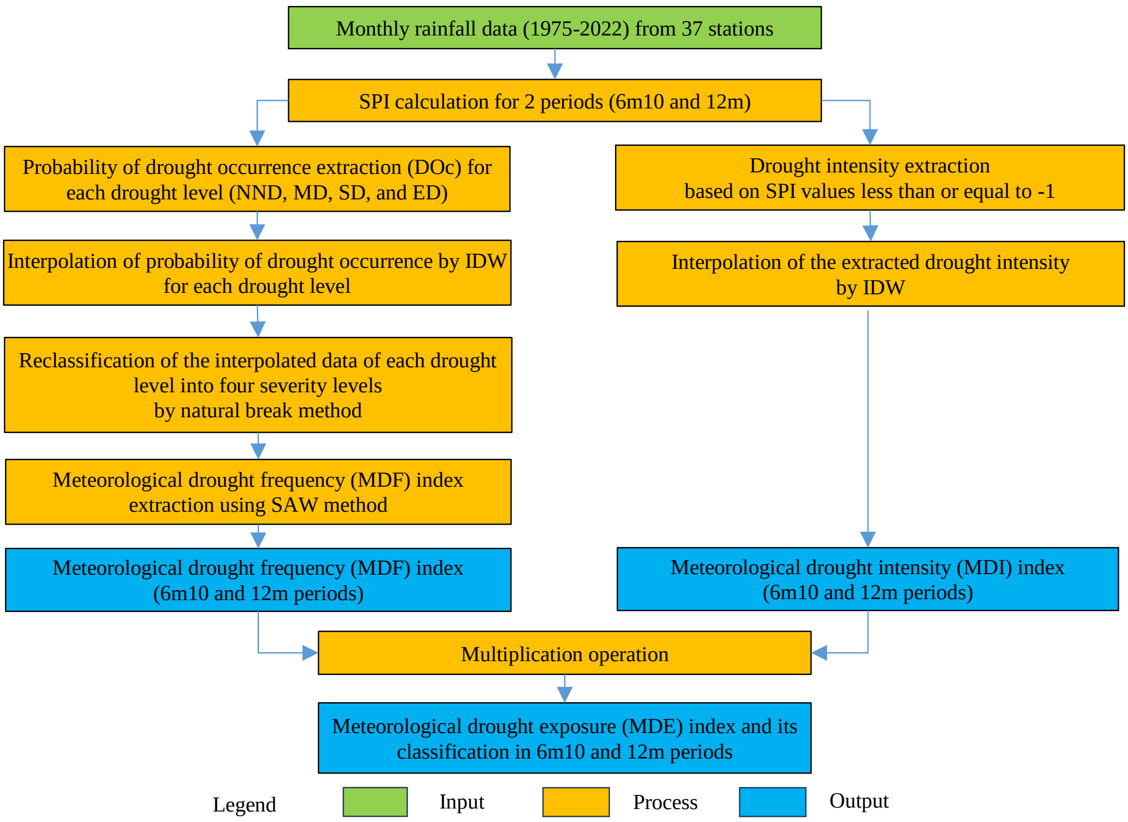

Figure 3. Workflow of agricultural drought exposure assessment

2.4 Assessing agricultural drought sensitivity (ADS)

ADS was assessed using a weighted linear combination (WLC) method with the analytical hierarchical process (AHP) based on selected factors under four conditions: vegetation, climate, physical and socio-economic (Figure 5).

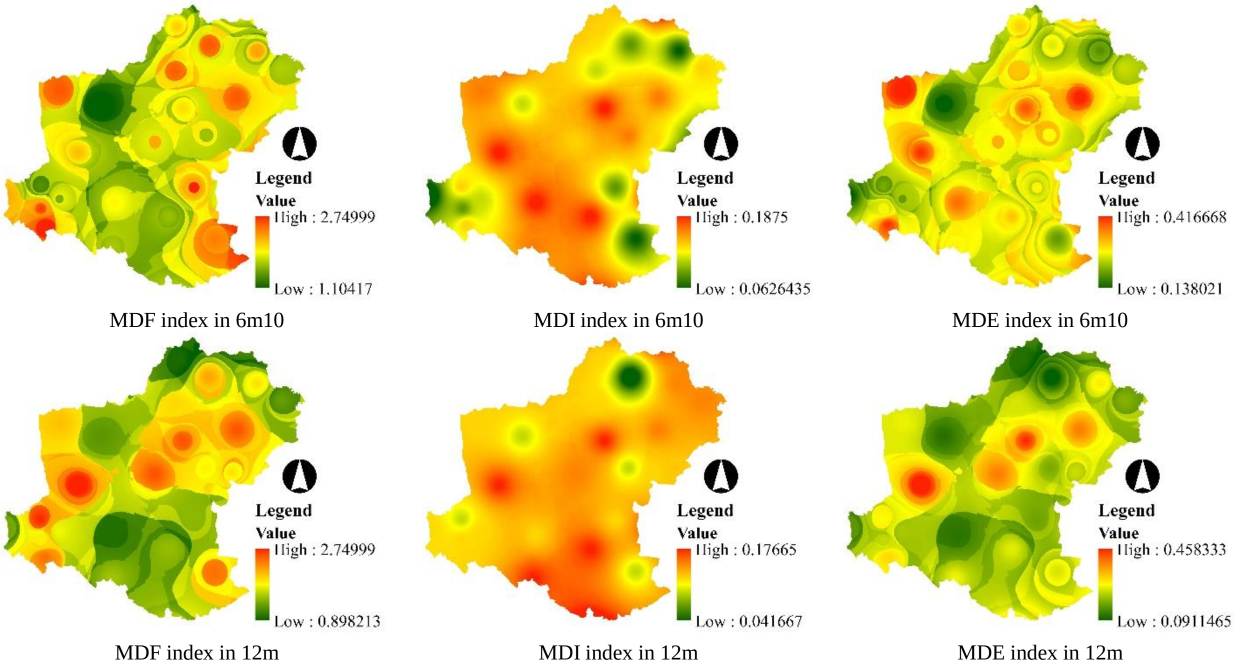

Figure 4. Spatial distribution of the MDF, MDI and MDE indices for MDE classification in 2 periods.

(1) Vegetation condition: Two factors representing vegetation conditions for ADS are agricultural drought frequency (ADF) and agricultural drought intensity (ADI). This study identified ADF based on the vegetation condition index (VCI) computed using the normalized difference vegetation index (NDVI) from MOD31A-NDVI products over a phenology period (May-October) from 2002 to 2022 using Equation 3. If VCI values are 100%, it indicates healthy vegetation conditions. In contrast, VCI values near 0%, identify poor vegetation conditions (Kogan, 1995).

$$VCI=100*\frac{(NDVI_i-NDVI_{min})}{(NDVI_{max}-NDVI_{min})} \tag{3}$$

Where; $NDVI_i$ is the filtered NDVI image in the phenology period, $NDVI_{max}$ is the multi-year maximum NDVI in the phenology period, and $NDVI_{min}$ is the multi-year minimum NDVI in the phenology period.

Since it represents vegetation conditions, any VCI equal to or less than 35% in the phenology period was identified as agricultural drought. All VCI images were reclassified with a threshold value of =<0.35 as 1, while other values were reclassified as 0. Thereafter, all reclassified images (1 and 0) were added together and divided by the total number of images (210 images from 21 years) for ADF. The extracted value was reclassified into five rating scores via the NB method.

Meanwhile, ADI was calculated based on average historical VCI values (0-100%) in the phenology period. Herein, all VCI images were reclassified with a threshold value of =< 0.35, while other values were reclassified as 0. Thereafter, all reclassified images were added and averaged by the number of years for ADI. The extracted value was then reclassified into five rating scores by using the NB method.

(2) Climate condition: Two factors that characterize climate conditions for ADS, as suggested by Hayes et al. (2011) and the World Meteorological Organization (WMO) (2012), are average SPI and SPEI (Standardized Precipitation Evapotranspiration Index). Our SPI was calculated and averaged from rainfall data (2002-2022) drawn from 37 stations in the 2 periods. They were interpolated using the IDW method and reclassified into five rating scores by the NB method.

Meanwhile, SPEI, specifically including characteristics of the region's climate (Vicente-Serrano et al., 2010), was calculated based on monthly rainfall and temperature (2002-2022). Herein, monthly rainfall data were retrieved from 37 stations, while average monthly average temperatures were retrieved from MODIS LST data. Monthly rainfall and temperature data were used to calculate the average SPEI of the 2 given periods using the SPEI calculator. They were interpolated using the IDW method and then reclassified into five rating scores via the NB method.

(3) Physical condition: Seven factors characterize physical conditions for ADS. These include land use, agricultural irrigation areas, distance to waterbodies, drainage density, landform, and elevation.

(3.1) Land use: Land use data in 2008, 2011, 2015, 2017, 2019, and 2023 from the Land Development Department (LDD) were reclassified into five rating scores according to land use type, and then averaged to produce five rating scores.

(3.2) Agricultural irrigation area: ADS, according to agricultural irrigation area, was assigned rating scores for irrigated and rain-fed agricultural areas.

(3.3) Distance to water bodies: Areas closer to waterbodies are less vulnerable to water shortages because of more recharge potential (Jain et al., 2015). Euclidean distance was utilized to calculate the distance to water bodies, and these values were then reclassified into five rating scores by the NB method.

(3.4) Drainage density: Drainage density values were calculated using the total length of stream channels in a drainage basin divided by the surface area of the basin (Pandey et al., 2010). These were then classified into five rating scores by using the NB method.

(3.5) Soil drainage: Soil drainage properties were sorted by soil series obtained from the LDD and then reclassified into five rating scores, as Prathumchai et al. (2001) suggested.

(3.6) Landform: Landform classification was based on the percentage of slope (Land Development Department, 2009) and then reclassified into five rating scores.

(3.7) Elevation: The elevation classification was extracted using DEM, according to LDD standards (2009) and then reclassified into five rating scores.

(4) Socio-economic condition: Socio-economic factors include average areas of harvested rice (2011-2023), farmer households in 2023 and population density in 2023 at the subdistrict level.

(4.1) Average rice harvested area: Average rice harvested areas (2011-2023) were determined at the subdistrict level, then calculated and reclassified into five rating scores by using the NB method.

(4.2) Number of farmer households: The number of farmer households is sensitive to agricultural drought (Pei et al., 2016). Areas are also more vulnerable when the proportion of farmer households increases. The number of farmer households in 2023 was extracted and reclassified into five rating scores by using the NB method.

(4.3) Population density: Shahid and Behrawan (2008) applied population density measurements to assign agricultural drought sensitivity scores. Population densities in 2023 were extracted and reclassified into five rating scores via the NB method.

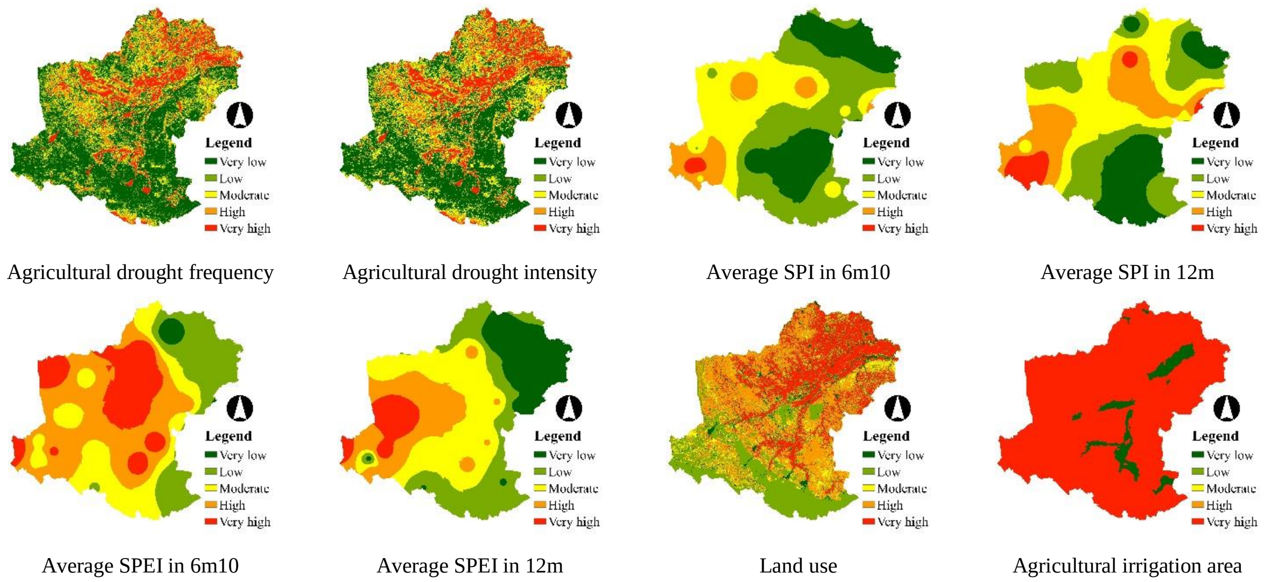

Figure 5. Spatial distribution of ADS assessment factors

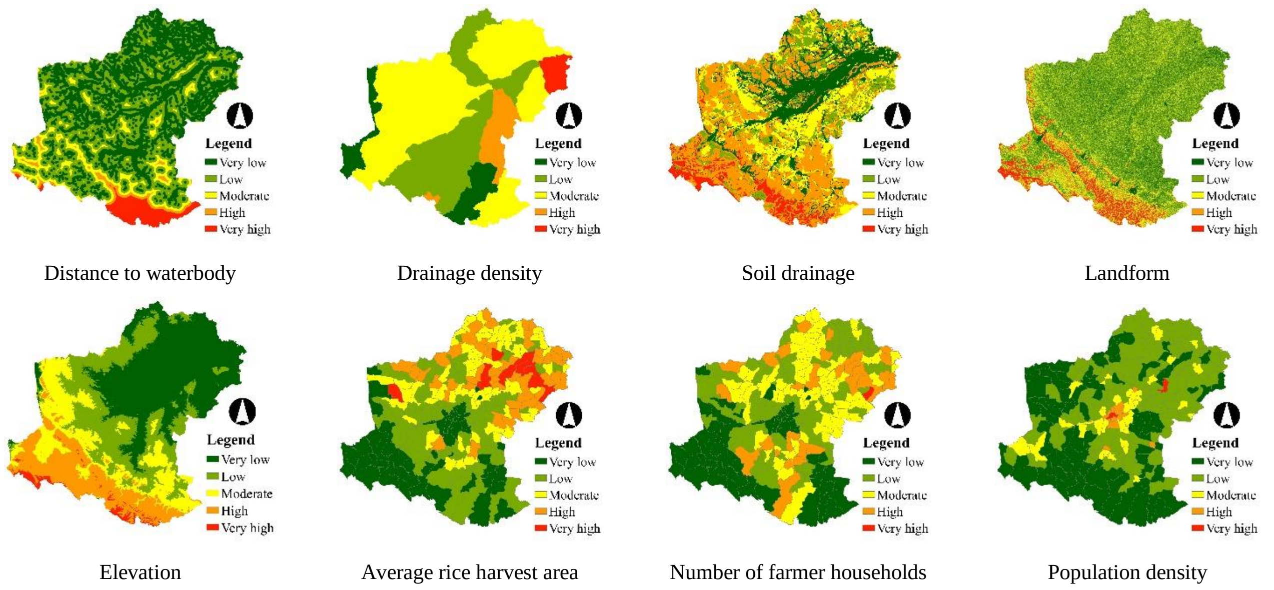

Figure 5. Spatial distribution of ADS assessment factors (cont.)

Since all ADS factors have different units, the rating score for each factor was normalized into a shared standard using a standardized ranking value (Tsangaratos et al., 2013):

$$v = a + (b - a) * \left[\frac{V - A}{B - A}\right] \tag{4}$$

Where; v is the new rating value that is between a and b, V is the original rating value that is between A and B, A is the minimum for original rating values, B is the maximum for original rating values, a is the new minimum of standardized rating values, and b is the new maximum of standardized rating values.

In this study, the minimum original rating value (A) is 1, and the maximum (B) is 5. Meanwhile, the new desired minimum of standardized rating values (a) is 1, and the new desired maximum of standardized rating values is 3.

Thereafter, the weight of each factor on ADS was determined using the AHP based on pairwise comparisons with the standard scale from 1 to 9 (Saaty, 1987). Value 9 indicates that one indicator is extremely important, even more so than the others, while value 1 indicates equal importance. The AHP was implemented by generating a pairwise comparison matrix and calculating its principal eigenvector directly to produce the best-fit set of weights with Weight and MCE modules (Eastman et al., 1995) under IDRISI software.

The normalized rating score and weight of each factor (Table 2) were applied to calculate the ADS index for the 2 given periods using the WLC method (Malczewski, 2000):

$$A_i = \sum_{j=1}^{n} w_j \times a_{ij} \tag{5}$$

Where; A_i is total importance of an alternative when all the criteria are considered simultaneously, and w_j denotes the relative weight of importance of the criterion C_j, and a_{ij} is the performance value of alternative A_i when it is evaluated in terms of criterion C_j.

Finally, the ADS index was reclassified into five severity levels (very low, low, moderate, high, and very high) using the NB method. See the workflow of ADS in Figure 6.

Table 2. Normalized rating and weight for ADS assessment

No	Factor	Normalized rating score					Weight
		Very low	Low	Moderate	High	Very high	
1	Agricultural drought frequency	1	1.5	2	2.5	3	0.1749
2	Agricultural drought intensity	1	1.5	2	2.5	3	0.1749
3	Average SPEI: 6m10 and 12m	1	1.5	2	2.5	3	0.1555
4	Average SPI: 6m10 and 12m	1	1.5	2	2.5	3	0.1510

Table 2. Normalized rating and weight for ADS assessment (cont.)

No	Factor	Normalized rating score					Weight
		Very low	Low	Moderate	High	Very high	
5	Land use	1	1.5	2	2.5	3	0.0820
6	Agricultural irrigation area	1	Not applicable	Not applicable	Not applicable	3	0.0570
7	Distance to waterbody	1	1.5	2	2.5	3	0.0530
8	Drainage density	1	1.5	2	2.5	3	0.0385
9	Soil drainage	1	1.5	2	2.5	3	0.0385
10	Landform	1	1.5	2	2.5	3	0.0239
11	Elevation	1	1.5	2	2.5	3	0.0194
12	Average rice yield	1	1.5	2	2.5	3	0.0115
13	Farmer households	1	1.5	2	2.5	3	0.0110
14	Population density	1	1.5	2	2.5	3	0.0089

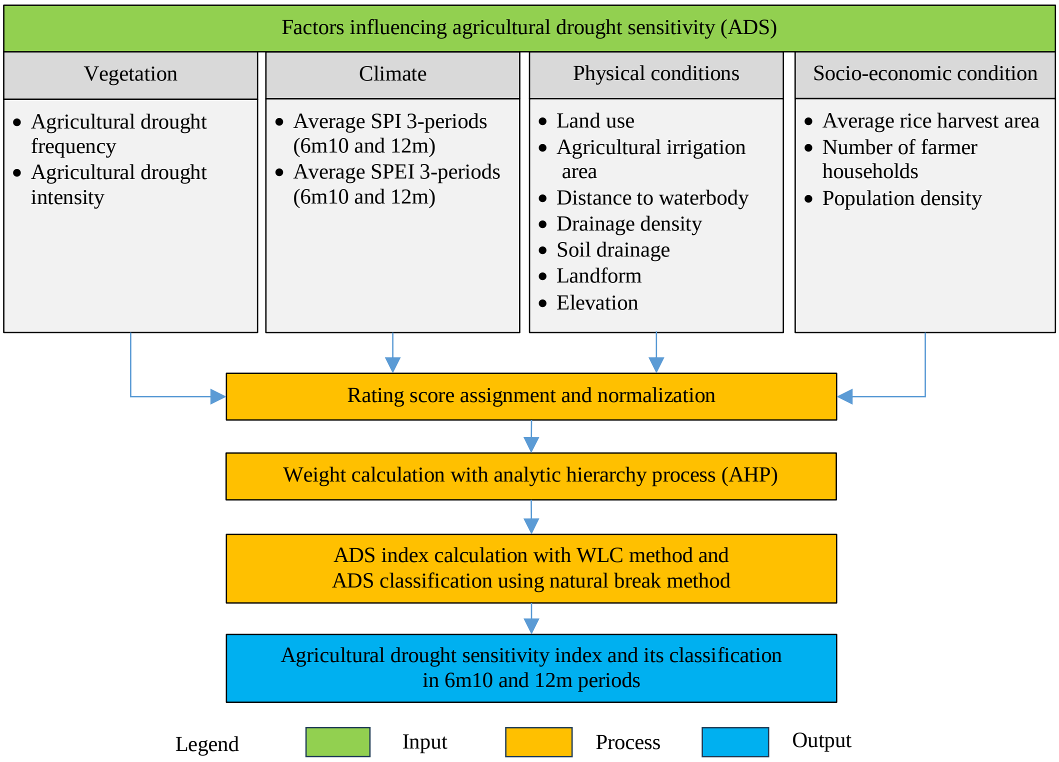

Figure 6. Workflow of agricultural drought sensitivity assessment

2.5 Assessing adaptive capacity

The AC index was assessed using the WLC method (Malczewski, 2000) based on suitability factors for groundwater use in agriculture, including potential groundwater yield, land use in 2023 and landform (Table 3). Finally, the AC index was reclassified into five suitable levels (very low, low, moderate, high, and very high) using the NB method.

2.6 Assessing agricultural drought vulnerability (ADV)

The ADE and ADS were first combined for the Potential Impacts of Agricultural Drought (PIoAD) using additive operation. Then, the derived data was combined with the AC using a subtractive operation for the ADV, and finally reclassified into five severity levels (very low, low, moderate, high, and very high) using the NB method.

Table 3. Rating and weight of suitability factors for groundwater use in agriculture

Suitability	Groundwater yield	Land use types	Landform	Rate
Not suitable	Waterbody	Others land uses	Waterbody	0
Low suitability	Yield < 2 cubic m/hr.	Cassava	Denudational hills and dissected erosion surface	1
Moderate suitability	Yield 2-10 cubic m/hr.	Sugarcane	High terrace	2
Highly suitable	Yield 10-20 cubic m/hr.	Corn	Low and middle terrace	3
Very highly suitable	Yield > 20 cubic m/hr.	Paddy field	Flood plain	4
Weight	3	2	1	

Note: The Waterbody category includes groundwater yield and landform, as surface waters are not considered suitable for groundwater use in agriculture.

2.7 Assessing spatial and temporal patterns of agricultural drought vulnerability and its potential impact on economic crops

The spatial and temporal patterns of ADV at district and subdistrict levels were analyzed according to severity levels using zonal analysis with majority operation. Meanwhile, overlay analysis was applied to identify the potential impacts of ADV on economic crops from LDD's 2023 land use data. Additionally, spatial correlation analysis was applied to characterize the similarity of the patterns.

3. RESULTS AND DISCUSSION

3.1 Potential impacts of agricultural drought (PIoAD)

The spatial and temporal patterns of the PIoAD for the 2 periods were extracted by combining their ADE and ADS (Figures 7(a) to 7(d)). The results are displayed in Figures 7(e) and 7(f). The spatial distribution of high and very high PIoAD in 6m10 occurred in the northwestern portion of the province. Meanwhile, the spatial distribution of high and very high PIoAD in 12m occurred in the northwestern and eastern regions. The spatial patterns of the PIoAD in the 2 periods differed from one region to the next. However, a spatial correlation analysis between the 2 periods showed a strong positive linear relationship, with a value of 0.7172.

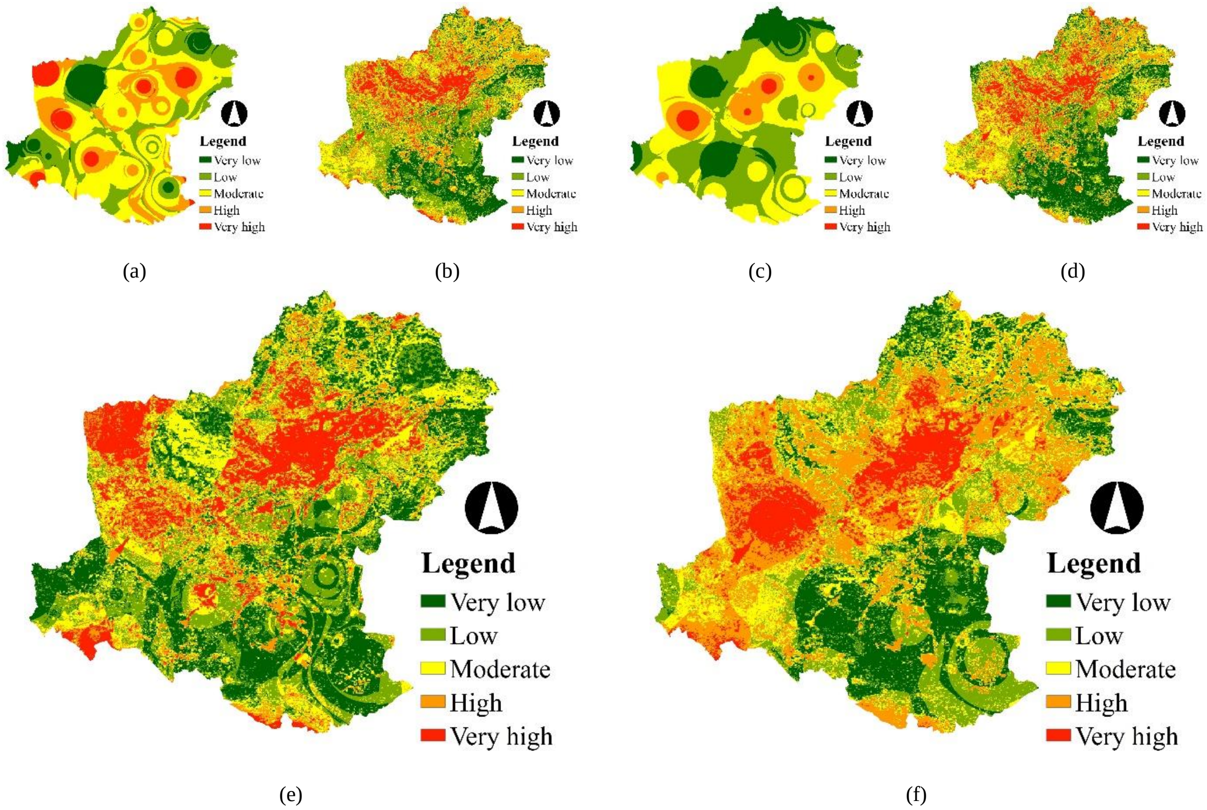

Figure 7. Spatial distribution maps showing (a) ADE in 6m10, (b) ADS in 6m10, (c) ADE in 12m, (d) ADS in 12m, (e) PIoAD classification in 6m10, (f) and PIoAD classification in 12m.

Furthermore, Table 4 reports the number of districts and subdistricts with a majority of severe PIoAD levels in the 2 given periods. In 6m10, combined moderate, high and very high PIoAD severity levels occurred in 18 districts and 146 subdistricts. Meanwhile for 12m, moderate, high and very high levels were found in 21 districts and 204 subdistricts. These results indicated changes in spatial and temporal patterns of PIoAD in both periods. The PIoAD at district and subdistrict levels in 12m were higher than 6m10.

Table 4. Number of districts and subdistricts levels with a majority of severe PIoAD levels in 2 periods

Severity levels of PIoAD	Number of districts and subdistricts			
	6m10 (May-October)		12m (January-December)	
	District	Subdistrict	District	Subdistrict
Very low	8	68	9	51
Low	6	74	2	33
Moderate	12	64	5	39
High	1	21	14	127
Very high	5	61	2	38
Total	32	288	32	288

3.2 Spatial and temporal patterns of agricultural drought vulnerability

The spatial and temporal patterns of ADV, which were extracted by subtraction of the PIoAD in 2 periods (Figures 8(a) and 8(b)) by AC (Figure 8(c)), are displayed in Figures 8(d) and 8(e). The spatial distribution of high and very high ADV in the 6m10 period occurred in the western and southern regions. Meanwhile, the spatial distribution of high and very high ADV in the 12m period occurred in the western region. These findings indicated that spatial and temporal patterns of agricultural drought vulnerability differ from one region to another region over both time periods. A possible reason for these findings is the variation in biophysical and climatic conditions. However, the spatial correlation analysis between the 2 periods showed a strong positive linear relationship, with a value of 0.7568.

Table 5 reports the number of districts and subdistricts accompanied by their ADV severity levels in 2 periods. For 6m10, moderate, high and very high levels of severity occurred in 10 districts and 96 subdistricts. Meanwhile over 12m, moderate, high and very high levels occurred in 17 districts and 166 subdistricts. These results indicated changes in spatial and temporal patterns of ADV in both periods. The potential impact of ADV at moderate, high and very high severity levels in 12m was higher than 6m10. Districts and subdistricts with identical potential ADV impact (moderate, high and very high, in both periods) were found in 3 districts and 48 subdistricts. See details in Table S1 and S2 in Supplementary data. These areas should continue to be monitored for mitigation drought.

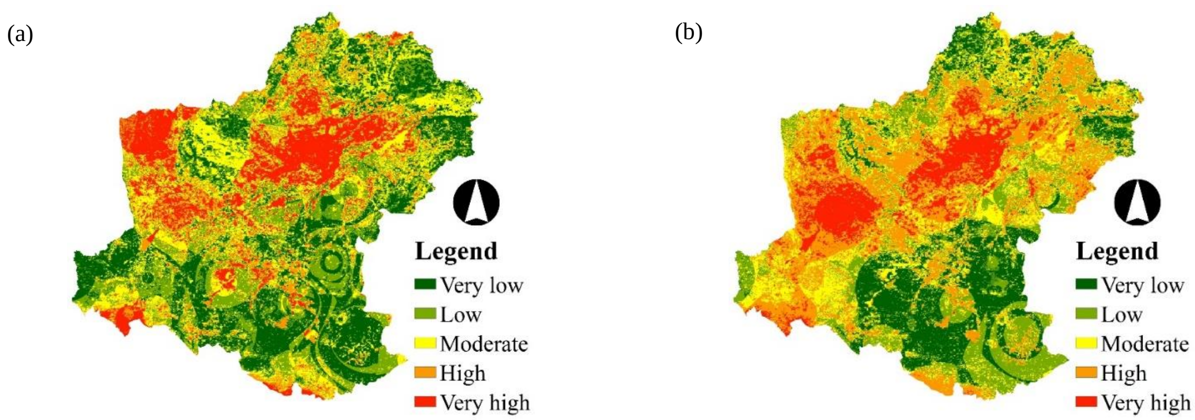

Figure 8. Spatial distribution maps showing (a) potential impact of agricultural drought in 6m10, (b) potential impact of agricultural drought in 12m (c) adaptive capacity (d) agricultural drought vulnerability in 6m10, (e) agricultural drought vulnerability in 12m.

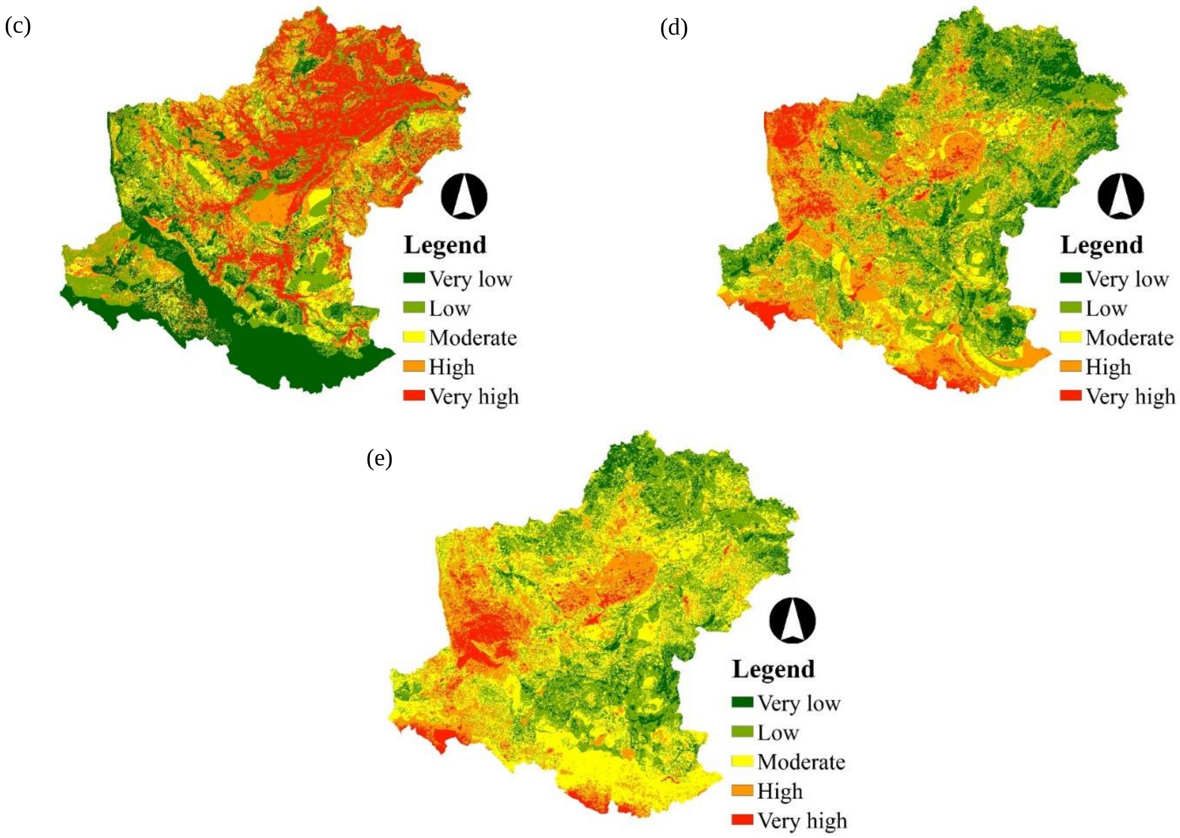

Figure 8. Spatial distribution maps showing (a) potential impact of agricultural drought in 6m10, (b) potential impact of agricultural drought in 12m (c) adaptive capacity (d) agricultural drought vulnerability in 6m10, (e) agricultural drought vulnerability in 12m (cont.).

Table 5. Number of districts and subdistricts with severity levels of ADV in 2 periods

Severity levels of ADV	Number of districts and subdistricts					
	6m10	12m	Duplicate in 2 periods	6m10	12m	Duplicate in 2 periods
	District	District	District	Subdistrict	Subdistrict	Subdistrict
Very low	1	0	0	16	6	1
Low	21	15	13	176	116	98
Moderate	1	15	1	20	120	14
High	8	2	2	68	40	33
Very high	1	0	0	8	5	1
Total	32	32	16	288	288	147

In comparison, the number of districts and subdistricts with moderate, high and very high PIoAD in 6m10 were reduced from 18 to 10 districts and from 146 to 96 subdistricts after applying CA. Likewise, the number of districts and subdistricts with moderate, high and very high PIoAD in 12m was reduced from 21 to 17 districts, and from 204 to 166 subdistricts after applying CA. (See Tables 3 and 4). These findings indicate the significant role of adaptive capacity utilizing groundwater use in agriculture for mitigating agricultural drought. It is recommended that a feasibility study for groundwater use in agriculture be further implemented.

3.3 Potential impacts of agricultural drought vulnerability on economic crops

Table 6 reports the potential impact of ADV in 2 periods on economic crops in 2023. Rice, corn, sugarcane and cassava are all presented according to severity levels. The percentage of the potential impact of ADV with moderate, high, and very high levels in the 6m10 and 12m periods for rice was 28.53% and 41.25% of the total rice area. Meanwhile, the percentage of the potential impact of ADV with moderate, high, and very high levels in 6m10 and 12m periods for corn was 34.97% and 55.74% of the total corn area. The percentage of the potential impact of

ADV with moderate, high, and very high levels in the 6m10 and 12m periods on sugarcane was 32.65% and 41.43% of the total sugarcane area. The percentage of the potential impact of ADV with moderate, high, and very high levels in 6m10 and 12m periods on cassava was 58.92% and 63.77% of the total cassava area. These findings indicate that the potential impact of ADV on economic crops in the 12m period was greater than the 6m10. Besides, the ADV exhibits the highest potential impact on cassava, since the phenological cycle of cassava covers the entire year (2023).

Table 6. Severity level of ADV in 2 periods on economic crops in 2023.

Economic crops	ADV severity level	6m10 (May-October)		12m (January-December)	
		Area (km^2)	Percent	Area (km^2)	Percent
Rice	Very low	1,569.49	25.76	941.94	15.46
	Low	2,785.60	45.72	2,637.54	43.29
	Moderate	973.62	15.98	1,894.23	31.09
	High	756.11	12.41	614.76	10.09
	Very high	7.92	0.13	4.26	0.07
	Total	6,092.73	100.00	6,092.73	100.00
Corn	Very low	182.31	23.28	55.44	7.08
	Low	326.94	41.75	291.16	37.18
	Moderate	122.87	15.69	314.10	40.11
	High	130.23	16.63	118.40	15.12
	Very high	20.75	2.65	3.99	0.51
	Total	783.10	100.00	783.10	100.00
Sugarcane	Very low	394.59	19.26	298.30	14.56
	Low	985.24	48.09	901.65	44.01
	Moderate	348.70	17.02	600.08	29.29
	High	270.64	13.21	220.24	10.75
	Very high	49.58	2.42	28.48	1.39
	Total	2,048.75	100.00	2,048.75	100.00
Cassava	Very low	171.10	4.44	75.53	1.96
	Low	1,411.98	36.64	1,320.26	34.26
	Moderate	902.91	23.43	1,385.77	35.96
	High	979.60	25.42	803.49	20.85
	Very high	388.06	10.07	268.21	6.96
	Total	3,853.65	100.00	3,853.65	100.00

4. CONCLUSION

This study integrated agricultural drought exposure, agricultural drought sensitivity and adaptive capacity for assessing spatial and temporal patterns of agricultural drought vulnerability (ADV) and the potential impact on crops at district and subdistrict levels. The results indicated that the potential impacts of spatial patterns of ADV with moderate, high, and very high severity between January and December (covering 17 districts and 166 subdistricts), were higher than between May and October (with 10 districts and 96 subdistricts). Likewise, the potential impact of agricultural drought vulnerability between January and December on economic crops exhibited more impact than the time between May and October. These results can be used as basic information for government agencies, such as the Department of Agricultural Extension and the Department of Disaster Prevention and Mitigation for monitoring agricultural drought in Nakhon Ratchasima province. The government should further implement a feasibility study for groundwater use in agriculture to mitigate the impact of drought on economic crops.

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AUTHOR CONTRIBUTION

Conceptualization, methodology, software, validation, formal analysis, investigation, resources, data curation, writing - original draft preparation and writing-review and editing, visualization, supervision, project administration and funding acquisition, SO.

DECLARATION OF COMPETING INTEREST

The author declares no conflict of interest.

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Controlling Small Particles for Two-Step Density Sorting of Simulated Microplastics: Overcoming Surface Tension Effects with Surfactants

Md. Ariful Islam[1], Shamim AL Mamun[2], Kei Nakagawa[3], Ken-ichi Shimizu[3], Mitsuharu Yagi[3], Achara Ussawarujikulchai[4], and Hiroshi Asakura[3*]

[1]*Graduate School of Fisheries and Environmental Science, Nagasaki University, 1-14 Bunkyo machi, Nagasaki 852-8521, Japan*
[2]*School of Physical and Chemical Sciences, University of Canterbury, 20 Kirkwood Ave., Ilam, Christchurch 8041, New Zealand*
[3]*Institute of Integrated Science and Technology, Nagasaki University, 1-14 Bunkyo machi, Nagasaki 852-8521, Japan*
[4]*Faculty of Environment and Resource Studies, Mahidol University, 999 Puttamonthon 4 Rd. Salaya, Puttamonthon, Nakornpathom 73170, Thailand*

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*** Corresponding author:**
E-mail:
asakura_hiroshi@yahoo.co.jp

ABSTRACT

Infrared spectrometers are commonly recommended for analyzing microplastics (MPs) in sediment samples. However, these instruments are costly and time consuming, limiting the scope of surveys and our understanding of the distribution and long-term variation of MPs. Although it is challenging to determine MPs by floatation sorting, it is possible to estimate the ratio of MPs that float and sink in seawater. The study employed floatation sorting to confirm whether MPs with densities lower than the liquid float and those with densities higher sink, even for MPs smaller than 1 mm. As expected, large MPs (1 to 4.75 mm in size) with densities higher than that of the liquid sank. Unexpectedly, small MPs (212 µm to 1 mm) with densities higher than the liquid's density also floated. Assuming the unexpected floating was due to surface tension, we added a surfactant to lower it, causing MPs with densities higher than the liquid's to either sink as expected or accelerate sinking. Thus, with the use of a surfactant, even small MPs can be sorted by density if a heavy liquid is used after water.

HIGHLIGHTS

- Large MPs (1 to 4.75 mm) with densities higher than those of the liquid sink as expected.
- Small MPs (212 µm to 1 mm) with densities higher than the liquid may unexpectedly float due to surface tension.
- Adding a surfactant reduces surface tension, accelerates the sinking of small MPs.
- Using a heavy liquid after water enables effective density-based sorting of small MPs.

1. INTRODUCTION

Plastics have become an indispensable material that is widely used in all aspects of our daily lives owing to their low price, durability, versatility, lightness, water repellency, and ductility (Luo et al., 2022; Li et al., 2018). Global production of plastics increased from 245 million tons in 2008 to 390.7 million tons in 2021 (Shukla et al., 2024), and is predicted to reach 600 million tons in 2050 (Yoganandham et al., 2023). However, only 6 to 26% of these plastics are recycled and the remaining 94% become plastic waste that continuously accumulates in landfills or directly enters the environment through various routes (Yang et al., 2022; Huang et al., 2021).

One of the most critical problems brought about by plastic waste is microplastics (MPs). MPs are small plastic particles with sizes less than 5 mm that have entered and polluted the environment (Wang et al., 2022; Wu et al., 2019). MPs are present in all marine ecosystems at varying concentrations and approximately 245 million tons are discharged into the marine environment annually (Alimba and Faggio, 2019). MPs have been widely detected in marine

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sediments, river sediments, soil, air, freshwater, wastewater, food, multiple organisms, and terrestrial ecosystems in recent decades (Chang et al., 2022; Huang et al., 2022; Cutroneo et al., 2021). MPs have a large hydrophobic surface and a rigid organic structure that can adsorb a variety of organic and inorganic pollutants, such as polycyclic aromatic hydrocarbons, polychlorinated biphenyls, heavy metals (including Cu, Zn, Cd, Cr, Pb, Co, Ni, Mn, Fe, Ag, and Hg), pharmaceuticals, and personal care products. Pollutant adsorption by MPs may lead to pollutant enrichment, which may increase local concentration in soil and exert combined effects on plants (Yang et al., 2022; Xiang et al., 2022). Therefore, it is necessary to know the distribution and long-term trends of MPs to prevent environmental pollution.

MPs have been detected in the surface and subsurface waters of the Atlantic Ocean, the Northeastern Pacific Ocean, and Arctic Polar waters, as well as in the surface waters of the North Sea, the Adriatic Sea, the Bohai Sea, and the South China Sea (Akdogan and Guven, 2019). The largest regional releases of MPs are in India and South Asia (18.3%), followed by North America (17.2%), Europe and Central Asia (15.9%), China (15.8%), East Asia and Oceania (15.0%), South America (9.1%), and Africa and the Middle East (8.7%) (Ang et al., 2022). Examples of MP densities in sandy beaches/coastal areas include 45-220 particles/kg (p/kg) in India (Tiwari et al., 2019), 232 p/kg in Bangladesh (Banik et al., 2022), 2.4-2.8 p/kg in the USA (Plee and Pomory, 2020), 338-1,270 p/kg in Norway (Olsen et al., 2020), 60-610 p/kg in South China (Zhang et al., 2019), and 61 p/kg in Mexico (Beckwith and Fuentes, 2018).

The separation methods of MPs in collected beach sediment samples can be classified mainly into physical, chemical, and biological methods (Tirkey and Upadhyay, 2021). The U.S. National Oceanic and Atmospheric Administration's (NOAA) manual for analyzing MPs in beach samples (Masura et al., 2015) comprehensively specifies the separation and analysis of MPs. Many studies have focused on MP analysis in such environmental media as marine, sand, and sediment (Soursou et al., 2023; Nabi et al., 2022) using the density separation (floatation) method (Tiwari et al., 2023; Nabi et al., 2022; Prata et al., 2019). The floatation method is easy and quick to perform and widely used to isolate/extract MPs from sand and sediment samples using a saturated salt solution (Crutchett and Bornt, 2024; Zhang et al., 2021). Salts for density separation include sodium chloride ($NaCl$), sodium iodide (NaI), zinc chloride ($ZnCl_2$), calcium chloride ($CaCl_2$), manganese (II) sulfate ($MnSO_4$), potassium formate ($CHKO_2$), and sodium polytungstate (SPT) (Soursou et al., 2023; Tirkey et al., 2021; Van Cauwenberghe et al., 2015). These heavy liquids are used to float not only MPs with low densities but also MPs with high densities that sink in water.

The NOAA manual recommends the use of an infrared spectrometer for the analysis of separated MP samples. This instrument determines not only whether the collected particles are plastic or not, but also the type of plastic material, thus facilitating the identification of the source of MPs. However, if material determination were required during MP surveys, the surveys would be limited to those conducted by professional researchers. Aside from being costly and time-consuming, the use of an infrared spectrometer limits surveys and prevents us from understanding the distribution and long-term variation of MPs. GESAMP (2019) recommends that surveys be conducted by citizen scientists to gather more information on the environment. Even though a survey by citizen scientists alone cannot determine the materials of MPs, it can provide supporting data on the materials. Asakura (2022) performed floatation sorting of MPs larger than 1 mm in size using water and saturated calcium chloride (SCC) solution and confirmed that MPs with densities lower than the density of the liquid floated and those with densities higher than the density of the liquid sank. This means that MPs can be sorted into two density levels if SCC solution is used after water. Although it is impossible to determine the materials of the MPs, it would be possible to estimate the ratio of MPs that float to those that sink in seawater. Assuming a certain land area, MPs with a specific gravity larger than 1 would originate not from ocean debris but from higher elevations on land. On the other hand, in the case of MPs with a specific gravity smaller than 1, it cannot be distinguished whether they drifted from the sea or land, but the MPs can be evaluated as having the potential to re-drift into the ocean. In this way, knowing the ratio of floating to sinking MPs in beach sediments, in addition to the amount of MPs present, gives us additional information about the current level of contamination and the possibility of contamination in the surrounding area, even without using an infrared spectrometer.

MPs larger than 1 mm in size are relatively large particles. Do small MPs behave as expected, i.e., as reported in Asakura's study? This is because large and small particles have different specific surface areas and are therefore affected differently by surface tension. In this study, we address the following questions. (1) Do MPs measuring less than 1 mm switch between floating and sinking depending on the density of the liquid? (2) If the MPs do not show the expected behavior as shown in (1), is there any way to improve the situation?

2. METHODOLOGY

2.1 Materials

2.1.1 Equipment

Commercially available scissors, nippers, cutters, shear crusher (MF10 Basic, IKA Japan Co., Ltd.), and a small mill (OML-1, Osaka Chemical) were used to shred plastic samples (hereinafter referred to as MPs). Stainless steel sieves (SANPO) with 212 µm, 1 mm, and 4.75 mm mesh sizes were utilized to adjust particle size distribution. For density measurements of MPs (L-size used), several 50 mL pycnometers, a thermometer (TT-508N, TANITA), a precision balance (ATY124, Shimadzu), and a water purifier (RFP841AA, ADVANTEC) were used. For the floatation sorting experiment, 300 mL glass beakers, a stainless-steel spoon, stainless-steel trays, and a dryer (DRD420DA, ADVANTEC) were used. For liquid density measurement, a graduated cylinder and a hydrometer (Ludwig Schneider) were utilized.

2.1.2 Samples

To prepare simulated MP samples, several types (PE, PP, PS, PVC, PET, PC, and PF) of plastic products (Table 1) were crushed and passed through a stainless-steel sieve to obtain L- (1 mm to 4.75 mm) and M- (212 µm to 1 mm) sized MPs.

The density of L-sized MPs was measured (Asakura, 2022). MPs with densities lower and higher than 1 g/cm^3 are called light and heavy MPs, respectively (Table 1). Saturated calcium chloride (SCC, Miyachu Building Materials Division, Inc.) solution was used as a heavy liquid for floatation sorting experiments of MPs because SCC is environmentally friendly and affordable (Debraj and Lavanya, 2023). Commercial kitchen detergent (Soapen Fresh Lime, Kaneyo Soap Co., Ltd.) was used to prepare the diluted detergent solution (hereinafter referred to as surfactant).

Table 1. Details of plastic products

Material	Description	Prepared MP size* for Figure 3						Density (g/cm^3)	Light / Heavy
		L size		M size					
		W	C	W	WS	C	CS		
Polyethylene (PE)	Shopping bag (SB)	x						0.908	L
	Polybottle (PB)	x	x	x	x	x	x	0.934	L
	Rope (RP)	x						0.754	L
	Glove (GV)	x						0.871	L
	Freezer bag (FB)	x		x				0.919	L
Polypropylene (PP)	PET bottle cap (BC)	x		x				0.925	L
	Flat plate (FP)	x		x				0.867	L
	Clothespin (CP)	x	x	x	x	x	x	0.905	L
	Rope (RP)	x						0.486	L
	Oriented PP (OP)	x						0.888	L
Polystyrene (PS)	Expanded polystyrene (EP)	x		x	x			0.018	L
	Flat plate (FP)	x	x	x	x	x	x	1.084	H
	Plastic bottle label (LB)	x	x	x	x			1.031	H
	Compact disk case (MC)	x	x	x	x			1.054	H
	Food tray (FT)	x	x	x		x	x	0.981	L
Polyvinyl chloride (PVC)	Pipe (PI)	x	x	x	x	x	x	1.424	H
	Flat plate (FP)	x	x	x	x	x	x	1.333	H
	Corrugated plate (CP)	x	x	x	x	x	x	1.375	H
	Tablecloth (TC)	x	x					1.305	H
	Non-slip sheet (NS)	x	x	x				1.218	H

Table 1. Details of plastic products (cont.)

Material	Description	Prepared MP size* for Figure 3						Density (g/cm^3)	Light / Heavy
		L size		M size					
		W	C	W	WS	C	CS		
Polyethylene terephthalate (PET)	PET bottle (EB)	x	x	x	x	x	x	1.378	H
	Egg pack (EG)	x	x	x	x			1.315	H
	Lumirror ® film (LF)	x	x	x	x			1.390	H
	Fruit container (FC)	x	x	x	x			1.336	H
Polycarbonate (PC)	Compact disk (CD)	x	x	x	x	x	x	1.163	H
	Safety glasses (SG)	x	x	x	x	x	x	1.166	H
	Flat plate (FP)	x	x	x	x			1.166	H
Phenol-formaldehyde (PF)	Pot knob (PK)	x		x	x			1.469	H

2.2 Methods

2.2.1 Principle of floatation sorting

The present study was conducted with light and heavy plastics to determine plastic behavior in individual floatation experiments. Figure 1 shows an ideal floatation sorting experiment. Weighed (indicated as initial amount, Figure 1) light or heavy plastics are added to individual beakers containing deionized water and stirred with a spoon (Ideal 1 and Ideal 2, Figure 1). After waiting for a while, the light plastics (indicated as floating matter, Figure 1) float on the water surface and are recovered using a spoon (Ideal 1, Figure 1) as the density of light plastics is lower than water density. On the other hand, heavy plastics settle at the beaker's bottom and thus cannot be collected from the water surface (Ideal 2, Figure 1). To collect the heavy plastics, water in the beaker is exchanged with a heavy liquid (i.e., SCC). The heavy plastics float on the liquid surface and are recovered by a spoon (Ideal 3, Figure 1). In this way, light and heavy plastics can be collected separately (two-step density sorting) from mixtures if floatation sorting is performed in water followed by a heavy liquid (Ideal 4, Figure 1).

However, errors occur in actual floatation sorting. Figure 2 shows the failure of the floatation sorting experiment. After mixing light plastics with water, some behave like heavy plastics, sinking to the bottom, whereas those that float on the water's surface are recovered by a spoon (Failure 1, Figure 2). The sinking is believed to result from binding with other particles or the beaker's inner surface. On the other hand, after mixing heavy plastics with water, some behave as light plastics, floating on the water's surface, and these are recovered (Failure 2, Figure 2). This is thought to be due to surface tension or combination with air bubbles. This unexpected behavior of plastics can lead to errors in the estimation of floating and settling fractions. The ratio of the amount recovered to the initial amount is used to determine the recovery rate. In this study, the weight of the particles is measured rather than the number. The unexpected behavior of heavy plastics can be prevented by adding a surfactant to the water to reduce surface tension and promote particle settling at the bottom (Solution for Failure 2, Figure 2).

2.2.2 Procedure for floatation sorting experiment

The floatation sorting experiment of single MPs (L and M size) was conducted to observe light and heavy plastic behavior in several liquids such as deionized tap water, SCC, tap water with surfactant, and SCC with surfactant. The concentration of the surfactant was 1/1,000 of the original solution. First, dry trays were weighed. The beaker (300 mL) was filled with 300 mL of liquid and measured (0.5±0.005 g) MPs of one plastic species (n=6 for L size and 5 for M size) were added to it. Due to experimental constraints, preparing simulated M-sized MPs is more challenging than preparing L-sized MPs, which is why the number of M-sized replicates is reduced. The liquid in the beaker was gently stirred with a spoon for 1 minute to accelerate the sinking or surfacing of MPs and remove air bubbles. After a few minutes, floating MPs were scooped out with a spoon and kept in a tray for drying in a dryer (80°C), and the dry weight was determined.

We verified whether the floating or sinking of MPs could be controlled by changing the density of the liquid. For this floatation sorting experiment, we used light (PC-CD, 1.163 g/cm^3) and heavy (PVC-FP, 1.333 g/cm^3) M-sized MPs (n=5). Water (1.00 g/cm^3) and low (diluted $CaCl_2$ solution, 1.30 g/cm^3), and high (SCC, 1.37 g/cm^3) density solutions with and without surfactants were used as liquids.

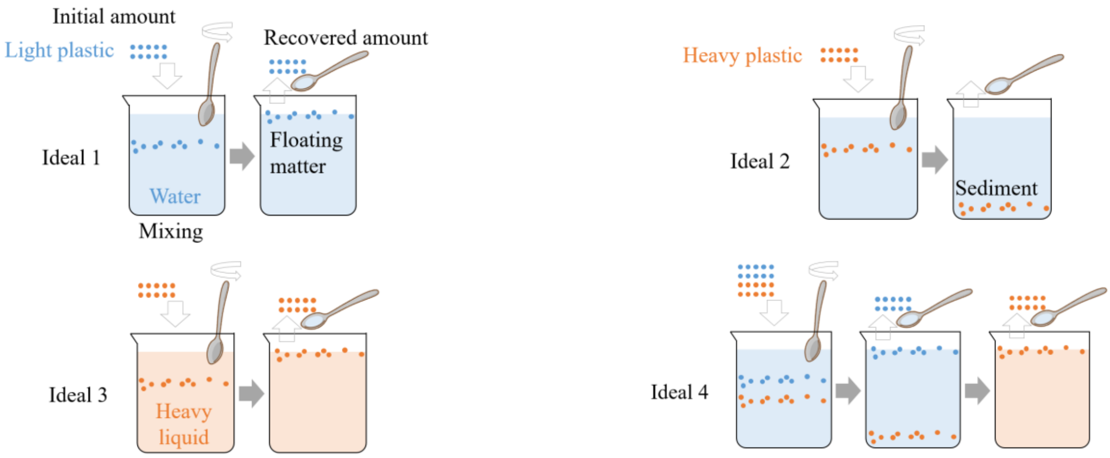

Figure 1. Ideal floatation sorting experiment

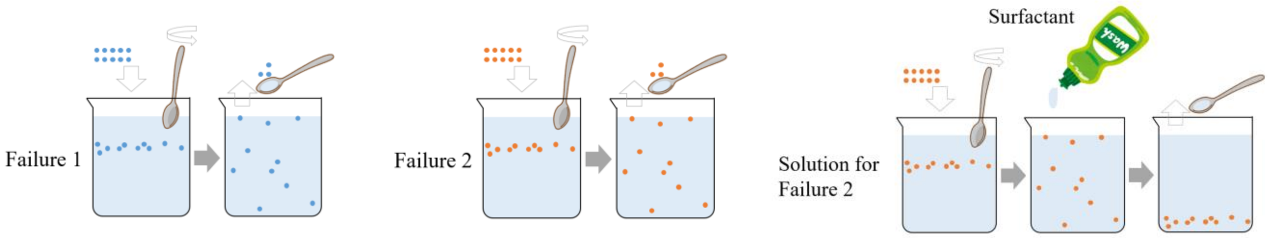

Figure 2. Failure and success in the floatation sorting experiment

3. RESULTS AND DISCUSSION

3.1 Floatation sorting of MPs using water and SCC solution

Figure 3 shows the relationship between the density of MPs and the recovery rate by floatation sorting. The L- and M-sized light MPs floated on the water's (1.00 g/cm^3) surface with recovery rates reaching nearly 100% (Figures 3(a), (b), (c)). In water, most of the L-sized heavy MPs settled at the bottom with a recovery rate of approximately 0% (Figure 3(a)). In contrast, some M-sized heavy MPs tended to float, resulting in a higher recovery rate than the L-sized heavy MPs (Figure 3(b), area shaded in orange). When a surfactant was added to water, the M-sized heavy MPs settled and the recovery rate dropped to nearly 0%, except in the case of PS-FP (Figure 3(c), area shaded in orange).

The L- and M-sized MPs, whose densities are lower than that of SCC (1.37 g/cm^3), floated to the surface, resulting in almost 100% recovery rates (Figures 3(d), (e), (f)). For MPs with densities higher than that of SCC, the L-sized MPs settled at the bottom (Figure 3(d)), while the M-sized MPs floated to the surface, resulting in a 100% recovery rate (Figure 3(e)). However, when a surfactant was added, the sedimentation of the M-sized MPs was accelerated, and the recovery rate decreased to 86% (Figure 3(f)).

In any case, the presence or absence of a surfactant did not influence the approximately 100% recovery rates of MPs whose densities are lower than that of the liquid.

3.2 Floatation sorting of MPs using diluted $CaCl_2$ solution

Figure 4 shows the relationship between the volume mixing ratio (heavy liquid/pure water) and the density of the liquid mixture. The predicted density is calculated using the following equation.

$$\text{Predicted density} = \frac{\text{Water density} + (\text{Mixing ratio} \times \text{Heavy liquid density})}{(1 + \text{Mixing ratio})}$$

The actual density of the liquid mixture is almost equal to the predicted density. This means that liquids with the required density (between water and SCC) can be easily prepared.

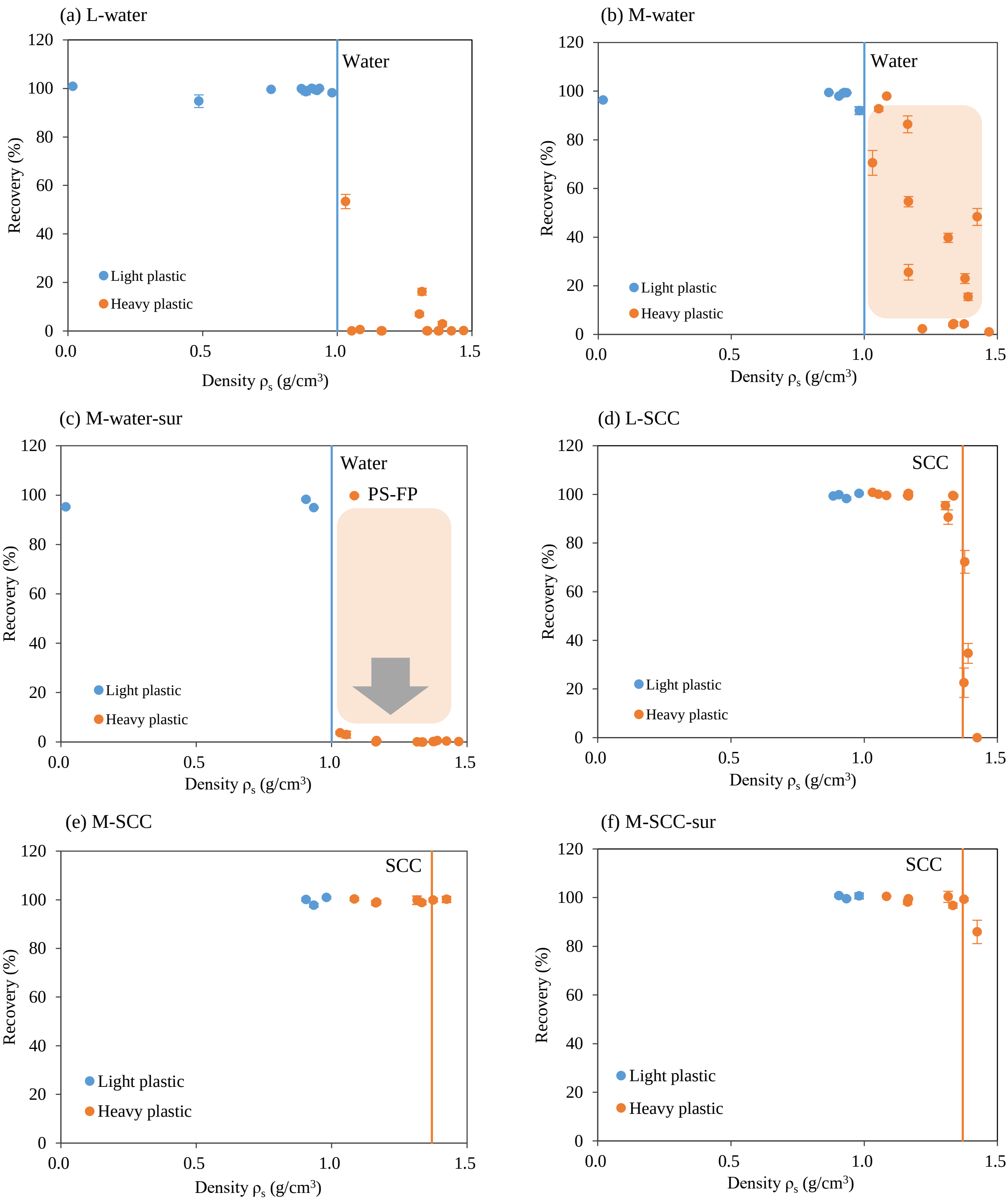

Figure 3. Relationship between the density of MPs and the recovery rate by floatation sorting. n=6 (L: 1 mm to 4.75 mm) or 5 (M: 212 µm to 1 mm). Vertical line: density of liquid (water or SCC). Error bar: standard error (α=0.05). SCC: saturated $CaCl_2$; sur: surfactant.

We also examined the behavior of M-sized MPs by using light (PC-CD) and heavy (PVC-FP) MPs in liquids with various densities. We found that the recovery rate varied with the density of the liquid (Figure 5). When water (1.00 g/cm^3) was used as a liquid, PC-CD (1.163 g/cm^3) floated on the water surface and the recovery rate was 86%. In contrast, when a surfactant was used with water, PC-CD settled at the bottom and the recovery rate was 0.1%. PVC-FP (1.333 g/cm^3) settled when water and water with a surfactant were used as the liquid, and the recovery rate was approximately 4.0% and 0.0%, respectively.

In the diluted $CaCl_2$ solution (1.30 g/cm^3), PC-CD (1.163 g/cm^3) floated, achieving a recovery rate of

98%. In the same solution, some PVC-FP (1.333 g/cm³) also floated, with a recovery rate of nearly 30%. However, after adding a surfactant, most of the PVC-FP settled, reducing the recovery rate to just 2.9%.

In SCC (1.37 g/cm³), both PC-CD and PVC-FP floated regardless of the presence or absence of the surfactant, and the recovery rate exceeded 96%.

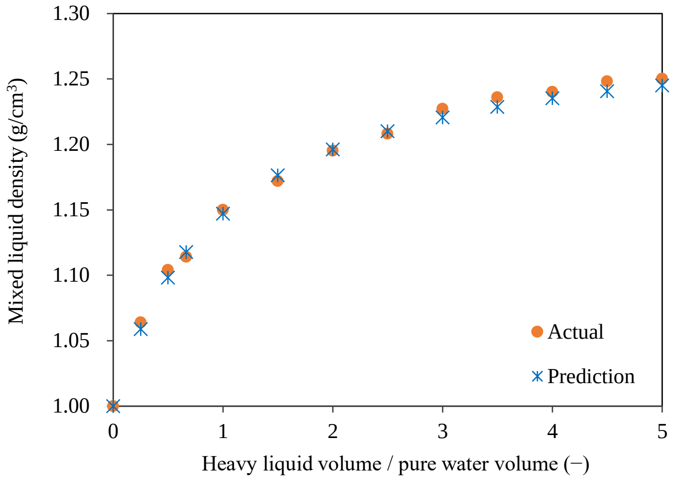

Figure 4. Relationship between volume mixing ratio and density of liquid mixture when heavy liquid (SCC) is mixed with pure water.

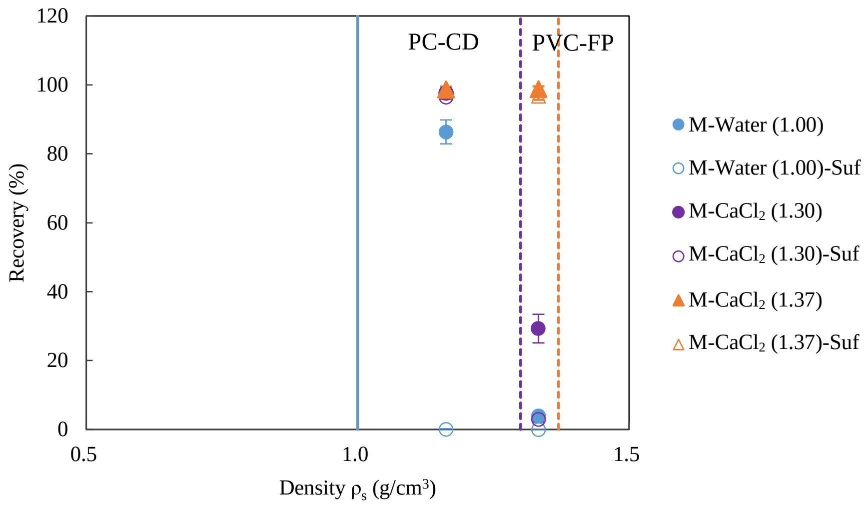

Figure 5. Recovery rates of MPs in liquids with various densities. Values in parentheses in the legend indicate the density of the liquid used (g/cm³); the colors of the symbols match the colors of the lines representing the density of the liquid used. n=5. Size: M (212 µm to 1 mm). Vertical lines: density of liquids. Error bar: standard error (α=0.05).

4. DISCUSSION

4.1 Behavior of small MPs during floatation sorting

In floatation sorting, particles with densities lower than the density of the liquid used should float and particles with densities higher than that of the liquid used should sink (expected floatation behavior). As shown in Figures 3(a) and 3(d), L-sized (1 mm to 4.75 mm) MPs exhibited the expected sinking behavior in water and SCC. However, M-sized (212 µm to 1 mm) MPs with densities higher than that of the liquid floated, contrary to expectations (Figure 3(b) (area shaded in orange) and Figure 3(c)). This means that MPs with low densities are overestimated (and MPs with high densities are underestimated) in the two-step density sorting. Assuming that this unexpected floating was due to surface tension, we added a surfactant to lower the surface tension, and MPs with densities higher than that of the liquid either sank as expected (Figure 3(c), area shaded in orange) or showed accelerated settling (Figure 3(f)), except for a few.

It is easy to dilute SCC to create a liquid with the desired density. The actual densities agreed with the predicted densities obtained from the calculation

(Figure 4). Even in liquids with densities between those of water and SCC, some M-sized (212 μm to 1 mm) MPs with densities higher than that of the liquid floated, contrary to expectations, but sank as expected when a surfactant was added (Figure 5).

MPs with densities lower than that of the liquid floated as expected regardless of the density of the liquid or the presence or absence of surfactant (Figures 3 and 5). This means that MPs with low densities are not underestimated in the two-step density sorting.

Various brine solutions including NaCl, NaBr, NaI, and $ZnBr_2$ were used in density separation experiments to recover different MPs (Nabi et al., 2022; Zhang et al., 2021). Several MPs (500 μm to 3 mm; PE, PP, PVC, PET, PS, EPS, and PUR) exhibited recovery rates of 99%, 96%, 97%, 91%, 92%, 68%, and 96%, respectively, from marine sediments when a saturated NaCl solution (1.2 g/cm³) was used first, followed by a NaI solution (1.8 g/cm³) in density separation experiments (Nuelle et al., 2014). The recovery rates of PVC and PET, which have higher densities than the NaCl solution (Table 1), exceeded 90% due to the use of a high-density NaI solution. Our research results can corroborate this result. Quinn et al. (2017) extracted MPs from marine sediments using tap water and several saturated salt solutions of varying densities including NaCl, NaBr, NaI, and $ZnBr_2$. The recovery rates were higher for smaller (200 to 400 μm) MPs than for larger (800 to 1,000 μm) MPs, increasing as the liquid density increased. We also found that the recovery rates were high for small particles. Other density separation experiments gave similar results—MP size influenced the recovery rate, namely, small MPs have higher recovery rates than large MPs (Nabi et al., 2022; Vermeiren et al., 2020; Coppock et al., 2017). If we simply want to recover small MPs by floating sorting, no countermeasures are necessary, and we will be able to recover more heavy MPs than expected. However, to separate MPs by density, we need a surfactant to control the flotation behavior of small MPs, as indicated by this study's results.

Based on the present study we conclude that the improved density sorting method can be used in environmental monitoring and microplastic research to obtain more reliable data on MP distribution in sediments and water bodies aiding in pollution assessment and mitigation efforts.

4.2 Limitations and further study

Some soft plastics were difficult to shred and did not yield M-sized particles. MPs whose surfaces have been degraded by sunlight or contaminated with microorganisms or oil may behave differently from undamaged MPs.

In this study, we performed a preliminary two-step density sorting experiment to separate light plastics from heavy ones. In the future, we will conduct a two-step density sorting experiment to identify factors that inhibit expected sorting efficiency.

This study serves as a foundational investigation for the density sorting of simulated MPs. However, in the future, we will validate the method using real environmental samples, particularly sediment from sea beaches and seawater, with added MPs of known quantities to assess its applicability.

In this study, we used tap water as a substitute for seawater because the densities of tap water (1.00 g/cm³) and seawater (1.03 g/cm³) are very similar. However, in future work, we plan to focus on using seawater to better align with real-world conditions.

5. CONCLUSION

We examined whether MPs with densities lower than the liquid density float and MPs with densities higher than the liquid density sink by performing floatation sorting experiments. We included MPs less than 1 mm in size. Our main findings are described below.

(1) Large MPs (1 mm to 4.75 mm) with densities higher than that of the liquid sank, as expected. On the other hand, small MPs (212 μm to 1 mm) with densities higher than that of the liquid floated, contrary to expectations. MPs with densities lower than that of the liquid floated, as expected, regardless of the liquid density and the presence or absence of surfactant.

(2) Assuming that the unexpected floating was due to surface tension, we added a surfactant to lower the surface tension, and MPs with densities higher than that of the liquid either sank or showed accelerated sinking, except for some MPs.

Thus, with the aid of a surfactant, even small MPs can be sorted into two groups with different densities if heavy liquid is used after water.

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AUTHOR CONTRIBUTIONS

Conceptualization, Methodology, Visualization, Supervision, Project Administration and Funding Acquisition, H. Asakura; Investigation and Writing - Original Draft Preparation, M.A. Islam; Writing - Review and Editing, H. Asakura, S.A. Mamun, K. Nakagawa, K. Shimizu, M. Yagi, A. Ussawaruji-kulchai.

DECLARATION OF COMPETING INTEREST

The authors declare no conflict of interest.

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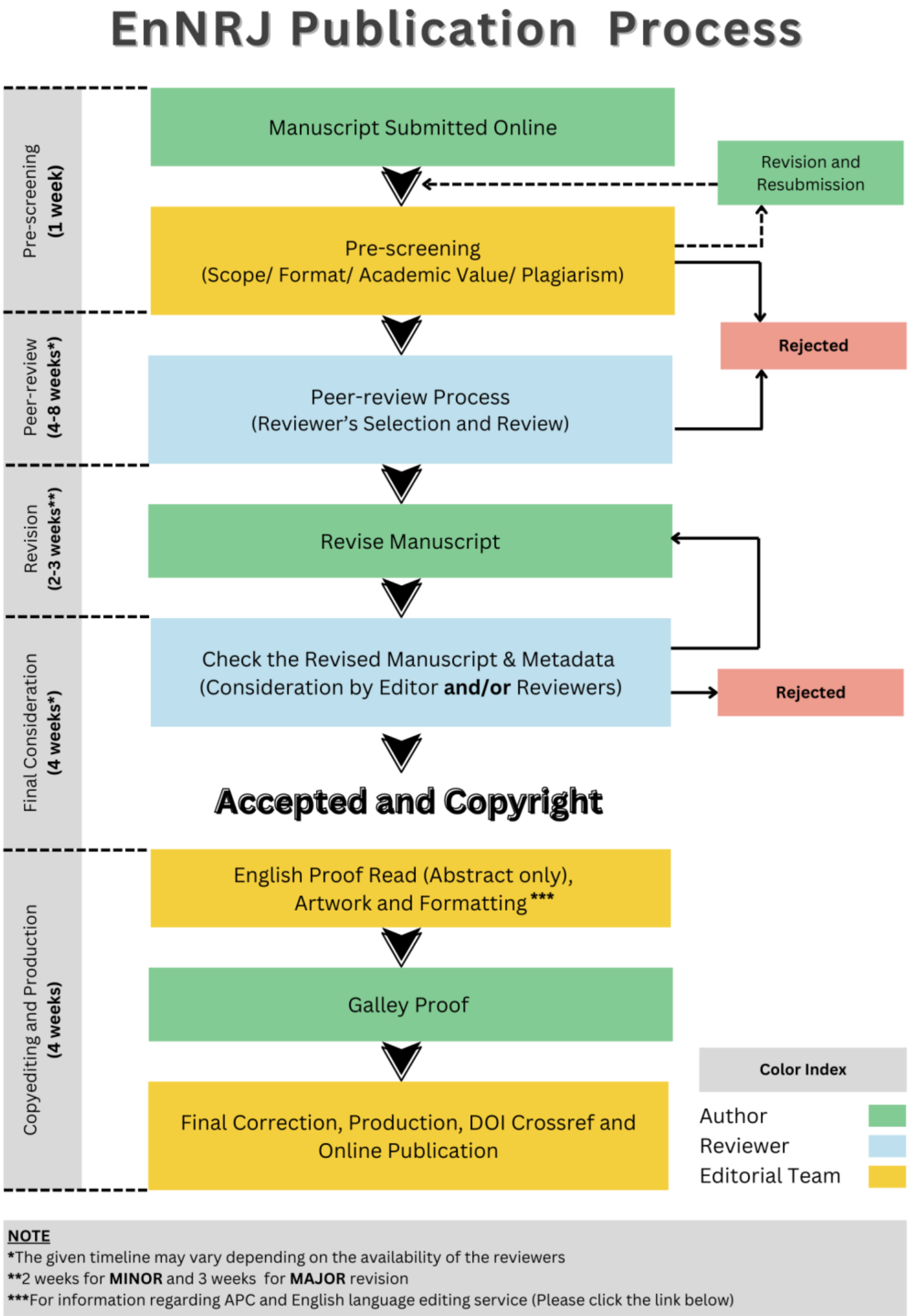

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Papitchaya Chookaew[1], Apiradee Sukmilin[2], and Chalor Jarusutthirak[1*]
[1]Department of Environmental Technology and Management, Faculty of Environment, Kasetsart University, Bangkok, Thailand
[2]Environmental Science and Technology Program, Faculty of Science and Technology, Phranakhon Rajabhat University, Bangkok, Thailand
**Corresponding author: abcxx@xx.ac.th*

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2.1 Sub-heading
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Journal article

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Journal article with Article Number

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Non-English articles

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Article in press

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Website

Orzel C. Wind and temperature: why doesn't windy equal hot? [Internet]. 2010 [cited 2016 Jun 20]. Available from: http://scienceblogs.com/principles/2010/08/17/wind-and-temperature-why-doesn/.

Report organization

Intergovernmental Panel on Climate Change (IPCC). IPCC Guidelines for National Greenhouse Gas Inventories: Volume 1-5. Hayama, Japan: Institute for Global Environmental Strategies; 2006.

Royal Gazette

Royal Gazette. Promotion of Marine and Coastal Resources Management Act 2059. Volume 132, Part 21, Dated 26 Mar B.E. 2558. Bangkok, Thailand: Office of the Council of State; 2015a. (in Thai).

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Faculty of Environment and Resource Studies, Mahidol University, Thailand
999 Phutthamonthon Sai 4 Rd, Salaya, Phutthamonthon District, Nakhon Pathom 73170
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