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Analysis of Contaminants in Paddy Irrigation System: Assessment on Water Quality

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Paddy and rice industry has garnered much interest from various aspects of research, due to its interconnection with food safety, food security, and socio-economic factors. The yield is highly influenced by the uses of various agrochemicals in different cultivation stages. Despite its advantages, uncontrolled use of agrochemicals may adversely affect humans, aquatic species, and the surrounding ecosystem. Hence, monitoring and controlling the contaminant, especially in water, is vital to ensure the toxicity will not affect the general population and environment. This study selected the Muda Agricultural Development Authority (MADA), a major rice-producing region (38.8%) in Malaysia as a focal point. Sampling was conducted from February to September, following the typical management schedules for paddy cultivation for the year 2022. Samples were collected and analyzed for various important physico-chemical parameters. Water Quality Index (WQI) was established and analyzed for elements and heavy metals, conductivity, phosphate, sulfate, nitrate, and pesticide residue screening. Considering rainfall is also a common, major water input to paddy fields, the correlation between rainfall data and cultivation stages against water quality and contaminant occurrences was also evaluated. Results show that 5% of data were classified as polluted (WQI <60) while the remaining gave WQI > 60, an indication of clean and slightly polluted status respectively, and in compliance with the minimum requirement for irrigation (Class IV) imposed by the DOE Water Quality Index Classification. However, the distribution of observed data shows that the WQI mostly declined during land preparation and sowing activity (April – May), supported by the decreasing subindex value for total suspended solids. National Water Quality Standards (NWQS) were used to assess the status of other contaminants in the irrigation system. Pesticide screening analysis on 28 active compounds perceived the residue was below the limit of quantitation (LOQ). Hence it demonstrates that the weather patterns and agronomic practices can alter the quality of irrigation water.

Keywords: Water quality; contaminants; paddy cultivation; MADA

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Water is a fundamental element in agriculture. Paddy cultivation, a staple crop in Malaysia, is bound to use large amounts of water. It is a short-term crop with a maturity period of 90 – 140 days post germination and planted twice (cropping season) annually. Generally, the main season (humid weather, Aug-Feb) is based on a non-irrigation system. In contrast, the off-season (dry weather, Mac – July) requires an irrigation system [1] Normal practice in paddy and rice production begins with land preparation (soil plowing, flooding), sprout planting, plant management and finally harvesting. In some of the activities, machinery was used to assist farmers, especially for land preparation and harvesting. Throughout the cultivation stages, paddy requires utmost attention and regular maintenance. Application of various agrochemicals such as soil amendment, pesticides, and fertilizers were used to maintain the yield [2]. Despite the advantage, intensive agricultural management practices in every stage of cultivation may contribute to adverse effects on humans, aquatic species, and the surrounding ecosystem [3]. Water quality problems evolve around inputs of contaminants into paddy systems, where the polluted water is exported via effluent pathways to the surrounding environment. Considering the above factors, there is an urgent need to assess the level of contaminants, especially in water bodies. Continuous monitoring is needed to create public awareness in order to prevent severe pollution in the water system. Therefore, it is necessary to quantify the degree of pollution, to manage the issues systematically. In Malaysia, the two primary standards employed to classify water quality are the Water Quality Index

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(WQI) and the National Water Quality Standards (NWQS). The WQI was developed by the Department of Environment, to evaluate the surface water quality. Six typical physicochemical water quality parameters, namely pH, Dissolved Oxygen (DO), Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Ammoniacal Nitrogen (NH_3-N), and Total Suspended Solid (TSS) were used to derive the sub-index of WQI. Benchmarking index comprised of three classifications, which are clean (81 – 100), slightly polluted (60 – 80) and polluted (0 – 59) [4] [5]. Further investigation on elements and heavy metals, conductivity, phosphate, sulfate, nitrate, and pesticide residue contamination were checked for compliance with NWQS Classification. Muda Agricultural Development Authority (MADA), a major rice-producing region (38.8%) in Malaysia was selected as a focal point for this study [6]. The Muda Irrigation Scheme covers an area of 130,282 ha that encompasses some parts of Perlis and Kedah, where, approximately 100,685 ha is utilized for paddy cultivation. A representative data collected from 34 sampling points scattered within the irrigation system of the granary area were affixed throughout the study.

EXPERIMENTAL

Sampling

Surface water sampling was carried out at 34 sampling points from The Muda Irrigation Scheme during the first season of the paddy cultivation schedule which is from February to September 2022. Water samples were collected in duplicate and stored in amber wide-mouth HDPE (1L) containers. Samples were transported from the sampling site to the main laboratory in Serdang for analysis. Samples were kept and maintained at low temperatures (< 10℃) during transportation.

In-situ Analysis

In-situ measurements for physical analysis namely pH, conductivity, and temperature were carried out using the YSI PRO DSS Multiparameter instrument (YSI Inc., Ohio, USA). Before use, all sensors were calibrated and verified using standard solutions to ensure the accuracy of data.

Ex-situ Analysis

Prior to laboratory analysis, samples were filtered through 0.45 µm pore cellulose membranes. Dissolved oxygen (DO) was determined using the YSI ProODO Dissolved Oxygen instrument (YSI Inc., Ohio, USA). Ammoniacal nitrogen (NH_3-N), phosphate (PO_4) sulphate (SO_4), nitrate (NO_3), and total suspended solids (TSS) were analysed using HACH DR 1900 spectrophotometer. Determination of biological oxygen demand (BOD) and chemical oxygen demand (COD) was carried out following Standard Methods for Examination of Water and Wastewater (APHA 2005) [7]. Analysis of elements (B, Ca, Fe, K, Mg. Mn, P, Na, S, Zn) and heavy metals (Al, As, Pb) were analysed using Inductively Coupled Plasma Emission Spectrophotometer (ICPOES) Perkin Elmer Optima 7300 DV (PerkinElmer Inc., Massachusetts USA).

Pesticide Residue Screening

Samples underwent further filtration using a 22 µm, 25mm PTFE hydrophilic membrane filter. The direct injection method was used to determine the pesticide residue [8]. Pesticides were screened against a mixture of 28 pesticide standards. The multiple reaction monitoring (MRM) detection method was carried out using liquid chromatography coupled to tandem quadrupole mass spectrometry (LC-MS/MS). The combination of equipment consists of an Agilent 1200 High-Performance Liquid Chromatography (HPLC) system coupled to an API 5500 QTrap mass spectrometer and controlled by Analyst Software. Chromatography was performed using Synergy Fusion-RP 100A 2.5µm column, 50 mm x 2 mm i.d (Phenomenax). The method used a gradient system with a flow rate of 400 µL/min. Eluent A was 0.01% formic acid in water, and eluent B was 0.01% formic acid in acetonitrile.

Water Quality Index (WQI) Determination

The WQI is used to indicate the level of pollution and the corresponding suitability in terms of water uses according to the NWQS. The six variables in WQI (DO, BOD, COD, TSS, NH_3-N, and pH) as adopted by the Malaysia Department of Environment (DOE) were calculated using the following equation [4]:

$$WQI = (0.22*SIDO) + (0.19*SIBOD) + (0.16*SICOD) + (0.15*SIAN) + (0.16*SISS) + (0.12*SIpH) \quad (1)$$

where SI stands for the sub-index of DO, BOD, COD, NH_3-N, SS, and pH respectively.

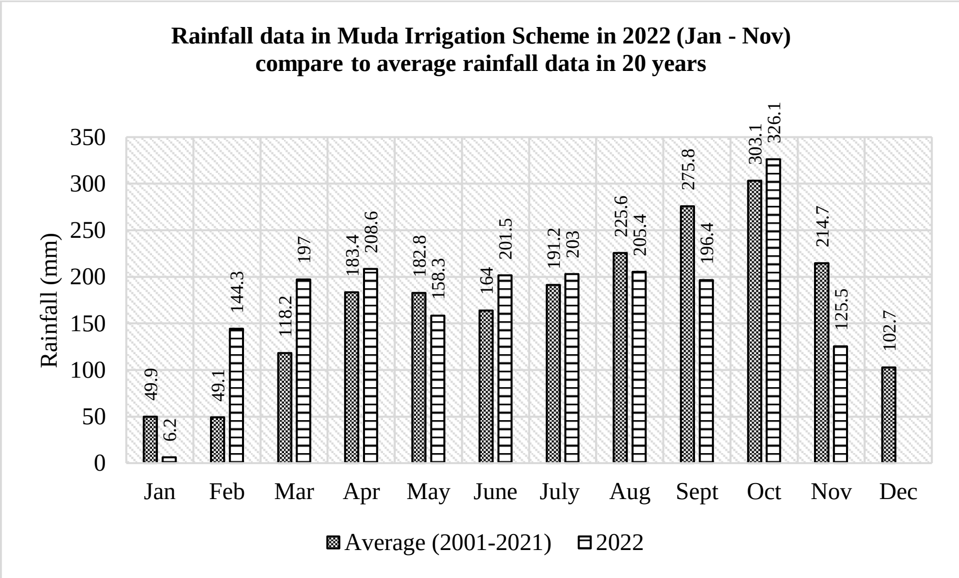

Figure 1. Rainfall distribution pattern at MADA in 2022 and average rainfall 2001-2021.

RESULTS AND DISCUSSION

Cultivation Management Schedule and Rainfall Data in Muda Irrigation Scheme.

Paddy cultivation management schedules proposed by MADA [9] started with land preparation in March/ April. During this event, extensive soil manipulation takes place to ensure favorable conditions for the growth of crops before the next activity, which was sowing seeds in May. After sowing, water input was controlled to accommodate the demand in every stage of plant growth. Agrochemical inputs were applied when the need, in the form of fertilizer to support the nutrient requirement, or pesticide to control insect infestation and diseases. The water supply gradually ceased by July/August to enable the harvesting process to take place. Sufficient water for irrigation is important for paddy cultivation. According to priority, MADA taps from four types of water sources to irrigate paddy fields, which are rainfall (39.6%), dam (33.1%), river flow (17.2%), and reusable drainage (10.1%). The total amount of yearly rainfall recorded in the Muda Irrigation Scheme until November 2022 is 1988.3 mm [9]. The monthly rainfall distribution is displayed in Figure 1.

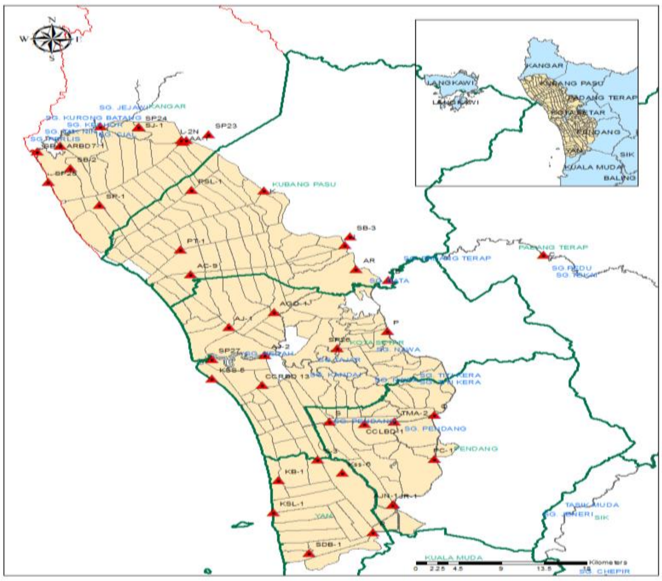

Figure 2. Location of 34 sampling points in MADA granary area.

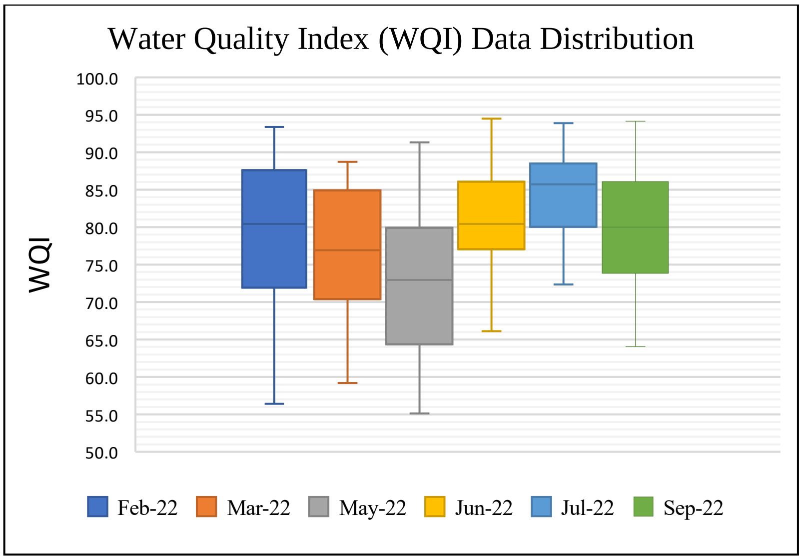

Figure 3. Data set for monthly monitoring of Water Quality Index (WQI)in MADA.

Soil manipulation and rainfall do play a role in the quality of the irrigation water due to water seepage and surface run-off. In corresponding to the first season of the cultivation schedule, six series of water sampling activities were carried out. The first sampling was in February, before the start of the season. The next sampling was in March, followed by May, June, and July and the final sampling was in August/September. The monthly set of data derived from 34 sampling locations is considered sufficient to discuss the occurrences of contaminants for this season. However, further monitoring is required to imply the actual water quality status in MADA. The reference location of the specific site is shown in *Figure 2*.

Assessment of Water Quality Index (WQI) and Physicochemical Data

Water Quality Index derived from six parameters (pH, DO, COD, BOD, NH_3-N, and TSS) were calculated. *Figure 3* shows the chart of WQI distribution for each month during the observation period. It was observed that 5% of data are categorized as polluted, while the rest are clean and slightly polluted. The observed polluted water was sampled in Feb, March, and May. The interquartile range (IQR) shows the distribution of the data set in May was relatively low, indicating the overall poor WQI compared to other months. Further investigation shows that TSS results have contributed to the drop in WQI value. In most relevant cases, the TSS value has exceeded Class IV of the National Water Quality Standard for irrigation which is 300 mg/l. This suggested that water quality tends to deteriorate during an early stage of cultivation due to soil preparation.

Other physicochemical parameters were analyzed and are summarized in *Table 1*. Results are compared with the minimum standard requirements for irrigation, which is Class IV of NWQS. All the samples had neutral pH in the range of 6.60 – 7.83. According to NWQS, it is considered unpolluted water and suitable for paddy cultivation. The daily variations of pH value may be due to the photosynthesis and respiration cycles of algae in the water [10].

Oxygen is needed by fish and other aquatic organisms. The root system also requires oxygen to promote healthy growth. Hence, analysis of DO is important. It is observed that before land preparation (February), the DO value was 6.62 mg/L (Class II), however; it dropped drastically (4.70mg/L and 4.28 mg/L) to slightly polluted Class III during land preparation and sowing stages (Mac – May). Moderate rainfall in April and May does not refrain from the negative physical impact contributed by soil manipulation. In the subsequent cultivation stage, the WQI values went back to Class II until the crop harvesting activity. The increasing trend in rainfall distribution from June to September has influenced the concentration level of contaminants by altering the volume and positively affecting DO.

The BOD value represents the amount of DO consumed by biological organisms when they decompose organic matter in water, whereas the COD is the amount of oxygen consumed when water samples are chemically oxidized [11]. The mean concentration of BOD and COD obtained throughout the monitoring period were slightly polluted (Class II and III), nevertheless is considered suitable for paddy irrigation. Land preparation activities affected the concentration of TSS, however, the values recorded were within the acceptable range for irrigation (< 300 mg/L). The maximum average value detected in May was slightly higher than the threshold level of TSS for supporting aquatic life in a freshwater ecosystem (150 mg/L) [12].

Algae blooms caused by water eutrophication were hastened in the presence of excessive nutrients such as PO_4, N, NH_3-N, or NO_3 in the water system [13]. Ammonium, labeled as NH_3-N in fertilizer is a readily available and water-soluble form of nitrogen. In this study, a comparatively high value of NH_3-N and NO_3 was detected in June and July, explained by the constant use of fertilizer [14], as the height of the darker green paddy plants grows. Irregular data of NH_3-N was observed in February, when the value was higher than the Class IV NQWS classification and, therefore considered polluted. The possible reason for the outlier was due to the accumulation of NH_3-N in thesoil. Ammonium is positively charged, it can be attracted to and held by negatively charged soil particles. In contrast, NO_3 is negatively charged, so it is not attracted to soil particles and therefore leached from the system with water.

Heavy metal accumulation is a concern due to its effect on food safety and crop growth. Phytotoxic trace elements and heavy metals may stunt the growth of plants or render the crop unfit for human consumption or other intended uses [15]. In this study, all the elements observed were within the acceptable range of Class I – Class IV, except for Aluminum (Al). Aluminum detected during this monitoring period was consistently > 0.5 mg/L. Although the values detected were slightly high, the occurrence of Al^{3+} toxicity that may cause stunted root growth was not observed during this study. This is because the phenomenon only exists in acidic soils.

Table 1. Compliance of physico-chemical parameters with NWQS.

PARAMETERS	UNIT	SAMPLING DESCRIPTION						STANDARD REFERENCES	
		Feb	Mar	May	June	July	Sept		
		Ave n=34	*Ave n=34*	*Ave n=34*	*Ave n=34*	*Ave n=34*	*Ave n=34*	*NWQSM (Class IV)	Others
NH_3-N	mg/l	**6.88**	0.17	0.53	1.16	0.40	0.34	2.7	
BOD	mg/l	2.23	1.41	1.27	2.18	1.51	1.87	12	
Conductivity	us/cm	290.00	589.00	218.00	71.73	120.70	180.95	6000	
COD	mg/l	15.61	36.90	17.78	8.70	14.30	10.47	>100	
DO	mg/l	6.62	4.70	4.28	5.91	5.45	6.19	<3	
pH		7.39	7.83	7.11	6.60	6.65	6.69	5 to 9	
PO_4	mg/l	0.44	0.84	0.81	0.64	0.60	0.00	Not available	
SO_4	mg/l	9.76	12.68	11.52	10.54	9.28	2.17		250 (Class II)
NO_3	mg/l	0.04	0.11	0.00	0.09	0.05	0.00	5	
TSS	mg/l	32.92	71.00	201.79	39.00	21.57	118.98	300	
Al	mg/l	**1.02**	**7.02**	**5.76**	**4.39**	**1.72**	**4.88**	0.5	
As	mg/l	0.01	0.00	0.00	0.05	0.02	0.05	0.1	
B	mg/l	0.03	0.08	0.08	0.04	0.03	0.03	0.8	
Ca	mg/l	10.88	14.31	11.87	8.81	6.79	8.52	Not available	
Cd	mg/l	0.00	0.02	0.01	0.00	0.01	0.00	0.01	
Fe	mg/l	0.76	3.41	4.91	1.96	1.33	2.96	1 (leaf) 5 (others)	
K	mg/l	2.60	10.04	5.08	3.76	3.46	6.43	Not available	
Mg	mg/l	6.11	13.53	6.43	4.37	3.30	3.15	Not available	
Mn	mg/l	0.03	0.08	0.15	0.04	0.02	0.12	0.2	
Na	mg/l	35.84	69.52	26.03	13.94	9.47	19.15	3SAR	
P	mg/l	0.10	0.14	0.10	0.08	0.10	0.16		0.1 (Class III)
Pb	mg/l	0.00	0.00	0.00	0.00	0.00	0.00	5	
S	mg/l	4.68	12.56	3.65	3.98	2.28	3.81	Not available	
Zn	mg/l	0.03	0.06	0.09	0.04	0.03	0.00	Not available	

Note:

1. **NWQS National Water Quality Standards of Malaysia*
2. ***Bold** value observed did not comply with NWQS standard*

Table 2. Pesticide residue screening of irrigation.

Compound	**RT (min)**	**Transition (precursor – product) (m/z)**			**LOQ (µg/L)**	**Recovery (%) at fortification level**		**Residue in sample (MADA)**	
		Pre	Quan	Qual		1 µg/L	100 µg/L	Detected (µg/L)	Reported
Florpyrixaufen Benzyl	4.41	439.1	91.1	65.2	10.00	117.87	87.08	*nd*	*nd*
Pymetrozine	4.40	218.2	105.2	78.1	9.00	111.29	97.68	*nd*	*nd*
Buprofezin	3.17	306.1	201.0	116.0	7.55	116.54	92.88	*nd*	*nd*
Isoprothiolane	3.55	292.2	231.9	190.0	10.00	81.08	106.66	*nd*	*nd*
Cartap	0.35	150.1	105.1	61.1	10.00	*na*	107.25	*nd*	*nd*
Azoxystrobin	3.32	404.1	372.1	344.1	8.10	111.23	77.63	*nd*	*nd*
Difenoconazole	4.07	406.1	251.1	253.1	9.51	120.41	100.19	*nd*	*nd*
Chlorantranilliprole	2.79	484.1	453.1	286.1	6.50	105.50	113.51	*nd*	*nd*
Thiamethoxam	0.72	292.1	211.1	132.0	10.00	87.95	71.08	2.61	< LOQ
Carbofuran	1.99	222.0	123.0	165.1	9.50	117.82	91.61	*nd*	*nd*
Cyromazine	0.33	167.1	85.0	68.0	10.00	*na*	101.37	*nd*	*nd*
Diazinon	4.08	305.3	97.0	153.0	9.27	86.7	85.56	*nd*	*nd*
Diflubenzuron	3.53	311.0	158.1	141.1	8.92	104.82	98.55	*nd*	*nd*
Dimethoate	1.00	229.8	199.0	125.0	7.41	97.08	75.64	*nd*	*nd*
Dimethomorph	2.89	388.0	165.0	301.0	8.69	109.86	103.92	*nd*	*nd*
Fenthion	4.08	279.0	247.1	169.1	8.34	119.49	86.33	*nd*	*nd*
Imidacloprid	0.93	256.1	175.0	209.0	7.45	*na*	92.91	2.14	< LOQ
Methomyl	0.69	163.1	88.0	106.0	9.98	104.79	98.84	*nd*	*nd*
Myclobutanil	3.23	289.0	70.2	125.2	9.76	74.77	87.83	*nd*	*nd*
Carbaryl	2.20	202.0	145.0	127.0	8.05	117.15	95.31	*nd*	*nd*
Carbendazim	0.45	192.0	160.0	132.3	9.57	117.16	80.56	*nd*	*nd*
Diuron	2.42	233.0	72.0	46.0	9.79	114.32	94.58	*nd*	*nd*
Abamectin	5.02	890.6	567.4	305.3	10.00	*na*	72.33	*nd*	*nd*
Tolclofos methyl	4.30	301.7	124.8	205.7	9,86	120	103.02	*nd*	*nd*
Prochloraz	2.74	378.4	267.9	70.1	9.90	110.68	95.27	*nd*	*nd*
Malathion	3.59	331.3	127.0	98.9	8.94	112.98	102.53	*nd*	*nd*
Fipronil	4.00	435.0	330.0	250.0	9.78	106.97	90.04	*nd*	*nd*
Tryclopyr	5.61	256.0	198.0	196.0	10.00	*na*	89.7	*nd*	*nd*

Note : *na* (not available) a*nd* (not detected)

Pesticide Residue Screening

Water bodies got contaminated by pesticides via some major routes such as leaching through soil, surface runoff aerial drift, dust, and transport of vapor. Pesticides with high water solubility are more potent to move from agricultural to surface water through runoff or irrigation. Nevertheless, in heavy rainfall seasons, pesticides that are strongly bound to soil particles can also escape into the surface water [16]. The method for screening 28 compounds of pesticides shall be verified before use. Standard addition at two levels of concentration, i.e., 0.001 mg/L and 0.100 mg/L, in five replicates respectively, were prepared in matrix match (ultrapure water) samples. The samples were introduced directly to LCMSMS and quantification was performed against a matrix-matched calibration curve All the pesticides were successfully detected, and the recoveries were compared with the acceptable tolerance of 70% to 120% range (SANCO/825/00 rev.8.1 2021) [17]. At a spiked level of 0.100 mg/L, recoveries for all pesticides fell within the acceptable limit, whereas, at a lower concentration of 0.001 mg/l, six pesticides namely cartap, cyromazine, fenvalerate, imidacloprid, difentrothion and abamectin were unquantifiable. Table 2 shows a summary of LCMSMS information, recovery results, and limit of quantification. Screening results on water samples collected from the MADA irrigation system were also summarized in Table 2. Two active compounds, thiamethoxam, and imidacloprid, were found at detectable levels below the quantification limit <LOQ specified for each compound. Both insecticides (neonicotinoids) are widely used in paddy cultivation to control sucking and chewing insects (rice hoppers, aphids, thrips, whitefly, etc.) [18].

CONCLUSION

The variation in water quality is related to the location of the sampling points and also the paddy planting activities. Based on WQI classification, in general, irrigation canals' water quality was within clean to slightly polluted status during the first season of MADA's paddy cultivation schedule. Nonetheless, 5% of WQI data falls in Class V indicating polluted water. Implicitly, the amount of TSS significantly increases due to earthworks, particularly at the early stage of cultivation, and the relevant deterioration may most likely be seen during high-flow conditions within the basin. Even though other contaminants were also observed, nevertheless the concentration was not exceeding the NWQS permittable limit for irrigation activities, except for NH_3-N and heavy metal Al. Aluminium detected during this monitoring period was slightly high (> 0.5 mg/L). The occurrence of Al^{3+} toxicity that may cause stunted root growth was not observed during this study. Pesticide screening shows two active compounds, thiamethoxam and imidacloprid were found at detectable levels, nevertheless, below the quantification limit <LOQ specified for each compound.

The outcomes of this study do not represent the long-term scenario of water quality in MADA as it is evidence that irrigation water quality is susceptible to changes due to weather conditions and agronomy practices. Hence, continuous monitoring and extensive data collection should be appropriately captured to improve the water quality status to ensure sustainable paddy cultivation without compromising human health and the environment.

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