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Assessment of harvested rainwater quality and surrogate parameter development for drinking water provision in rural Sri Lanka

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ABSTRACT

In rural dry zones of Sri Lanka, where drinking water safety is severely challenged, rainwater harvesting emerges as a crucial pathway toward safe water provision. However, knowledge gaps persist regarding water quality evolution under tropical environment with intense wet-dry alternation, while conventional monitoring approaches prove unsuitable for resource-limited settings. This study conducted a systematic quality assessment of harvested rainwater revealing that several water quality indicators exceeded drinking water standards, including turbidity, colour, chemical oxygen demand, ammonia and total coliforms. The results of source identification reveal that agricultural activities, infrastructure deterioration, roofing material, road-dust redistribution and atmospheric deposition are the major sources of rainwater contaminants. The comprehensive evaluation demonstrated poor initial rainwater quality (Water Quality Index (WQI) 113–395), while temporal analysis showed significant improvement through 30-day natural purification (WQI reduced by 20–49 %), despite microbial fluctuations under tropical conditions. Crucially, we demonstrated water colour's superior diagnostic capability over turbidity in multi-source pollution monitoring. As an integrative indicator, colour dynamically reflects both particulate and dissolved contaminants during rainfall-runoff processes. The statistically validated colour-microbe correlation during the long-term storage process provides a rapid assessment approach for microbial safety in rural areas, supporting decision-making through simplified colour threshold determination. Based on these findings, an integrated strategy was proposed combining first-flush diversion, natural purification processes, and colour-based simplified monitoring, which provides a practical and feasible management approach for resource-limited regions. This strategy offers an effective solution for highly improving the quality of harvested rainwater, though further treatment remains essential to ensure the drinking water safety.

1. Introduction

Access to safe drinking water remains a critical global challenge, with 2.2 billion people still lacking this basic necessity [1]. Despite the United Nations' Sustainable Development Goal 6 (SDG 6) targeting universal access to safely managed drinking water by 2030, current progress requires six-fold acceleration globally [2]. This challenge is particularly acute in developing nations, where implementation gaps are widening due to infrastructural and resource constraints [3]. In Sri Lanka, where nearly 80 % of the population resides in rural areas and approximately 40 % lack access to safe drinking water [4], 47.5 % of rural communities rely on groundwater that suffers from high levels of fluoride, hardness, and nitrates, posing serious health risks (such as chronic kidney disease and blue baby syndrome) [5,6]. Moreover, the drinking water problem in Sri Lanka's dry zone is particularly severe, affecting over 3 million people who must contend with both water

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scarcity and high groundwater salinity. Consequently, rainwater harvesting (RWH) has emerged as a promising alternative, endorsed by the United Nations for its potential to accelerate SDG 6 progress [7–9], due to its cost-effectiveness and simple maintenance requirements [10,11]. Furthermore, Sri Lanka's favorable natural conditions, such as substantial annual monsoon precipitation (averaging 1800 mm) and rainwater's inherently low mineral content, support the implementation of RWH as a safer substitute for contaminated groundwater [12,13].

However, in Sri Lanka's rural dry zones, where 80 % of annual rainfall is concentrated within 4–5 months, the feasibility of rainwater as a reliable drinking water source faces a unique challenge. The extreme hydrologic regime, characterized by prolonged dry seasons followed by intense monsoons, creates complex water quality dynamics that remain inadequately understood. This wet-dry alternation environment, combined with tropical high temperature and suboptimal sanitary conditions, accelerates water quality deterioration during collection and storage periods. Moreover, the distinct characteristics of these regions, including seasonal precipitation patterns and multiple potential pollution sources, generate complex contamination mechanisms (e.g., interactions between catchment surfaces and rainfall, atmospheric deposition processes, and seasonal microbial dynamics in storage systems) that current research has not fully addressed. While previous studies have examined rainwater quality in various contexts, critical knowledge gaps persist regarding long-term storage dynamics and multi-source pollution patterns under tropical wet-dry alternation conditions. Furthermore, ensuring rainwater safety demands comprehensive monitoring of physical, chemical, and microbiological parameters, typically requiring sophisticated laboratory equipment and technical expertise. In rural areas with limited infrastructure and resources, such extensive monitoring proves neither economically viable nor practically feasible [14,15]. Traditional water quality indicators like turbidity demonstrate significant limitations under tropical conditions, where intense seasonal rainfall and high temperatures create complex particulate matter dynamics that weaken the correlation between turbidity measurements and actual contamination risks [16]. Most water quality assessment frameworks and treatment strategies prove inadequate for addressing the unique challenges of tropical dry zones, where water quality evolution follows distinctly different patterns during extended storage periods under elevated temperatures [17]. Therefore, there is an urgent need for both region-specific water quality assessment frameworks that capture the impacts of seasonal transitions and simplified yet reliable monitoring systems suitable for resource-limited settings. This approach would not only advance our understanding of water quality dynamics in tropical dry zones but also provide practical solutions for ensuring safe rainwater utilization in developing regions.

To address these challenges, this study presents a systematic research framework. Initially, a comprehensive quality assessment of harvested rainwater in Sri Lanka's rural dry zones was conducted, encompassing pollution source apportionment, analysis of water quality dynamic characteristics, and monitoring of long-term storage trends, aimed at scientifically evaluating the feasibility of rainwater as a rural drinking water source. In addition, based on the assessment outcomes, the study presents sustainable rainwater harvesting system implementation strategies tailored to tropical developing countries' conditions. These strategies serve as a model for adaptation in nations facing similar challenges. Finally, addressing the practical needs of resource-constrained regions, this study aims to develop an innovative, simplified water quality monitoring framework, focusing on identifying easily measurable surrogate parameters that maintain reliable correlations with key water quality indicators, thereby overcoming the limitations of traditional monitoring methods in practical applications.

2. Materials and methods

2.1. Study area and rainfall monitoring

Sri Lanka lies at the southern tip of the South Asian subcontinent in the Indian Ocean. The country has a tropical monsoon climate with an average annual temperature of 28°C, characterized by wet and dry seasons. The wet seasons occur during April-June and October-December, while the remaining months constitute the dry season. This study focused on two rural areas in Sri Lanka: Anuradhapura in the north-central region and Puttalam in the northwestern region (Fig. 1). Both sites were selected based on their significant drinking water safety issues. The north-central area of Anuradhapura undergoes prolonged drought periods up to six months annually, showing minimal rainfall during June-August. The northwestern district of Puttalam exhibits similar water scarcity patterns during dry seasons. Both districts demonstrate typical tropical monsoon characteristics, alternating between periods of abundant rainfall and severe water shortages.

Harvested and long-term stored rainwater was investigated to characterize rainwater properties during wet seasons and monitor the temporal variations of stored rainwater throughout dry periods. A total of 54 water samples were collected across both sites during the final month of the wet season for rainwater assessment. For stored rainwater evaluation, 60 samples were collected across both sites over a 150-day period following the rainfall monitoring. The related rainfall information including rainfall depth, duration, intensity, and antecedent dry days (ADD), is listed in Table S1. The rainfall depth was measured using a digital rain gauge, and data for ADD was obtained by on-site monitoring. Rainfall intensity was calculated as millimeters per hour over the entire rainfall duration.

2.2. Sample collection and analytical methods

In this study, four houses were selected as sampling sites in each of the rural areas of Puttalam and Anuradhapura. All sampling sites were equipped with standardized rainwater harvesting systems, consisting of gutters, downpipes, first-flush diverters, and storage tanks. The houses featured uniform roofing material of red clay tiles. During rainfall events, rainwater runoff flows along the surface of the clay tiles into the gutters before being conveyed through the downpipe system to a high-density polyethene storage tank. Discrete samples were taken immediately at the start of runoff discharge from the downpipe, and every five minutes thereafter for the first 30 min, to properly capture and characterize the first flush. Flow rates were measured using flow meters for smaller fully-filled downpipes and volumetric estimation for larger diameter downpipes, enabling accurate estimation of runoff pollutant concentration by accounting for volume effects. During each sampling event, direct rainfall samples were collected in a 10-L container placed in an open area away from the roof runoff to determine the background concentrations (BCs) of pollutants in precipitation. Rainfall characteristics determined the sampling strategy, with sampling frequency adjusted according to rainfall depth, intensity, and duration to capture the dynamic changes in runoff concentration. All the samples were collected throughout the storm hydrograph.

The water quality parameters, including colour, turbidity, pH, ammonia, chemical oxygen demand (COD), chloride (Cl), chromium (Cr), manganese (Mn), iron (Fe), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), lead (Pb), and total coliforms, were mostly measured according to the Standard Methods for the Examination of Water and Wastewater [18], the detailed information for analytical method is summarized in Table S2. A composite sample made of 12–15 discrete samples collected from the entire rainfall sample was used for particle size distribution (PSD) measurement, and its analysis was performed using an AccuSizer 780 A particle analyzer.

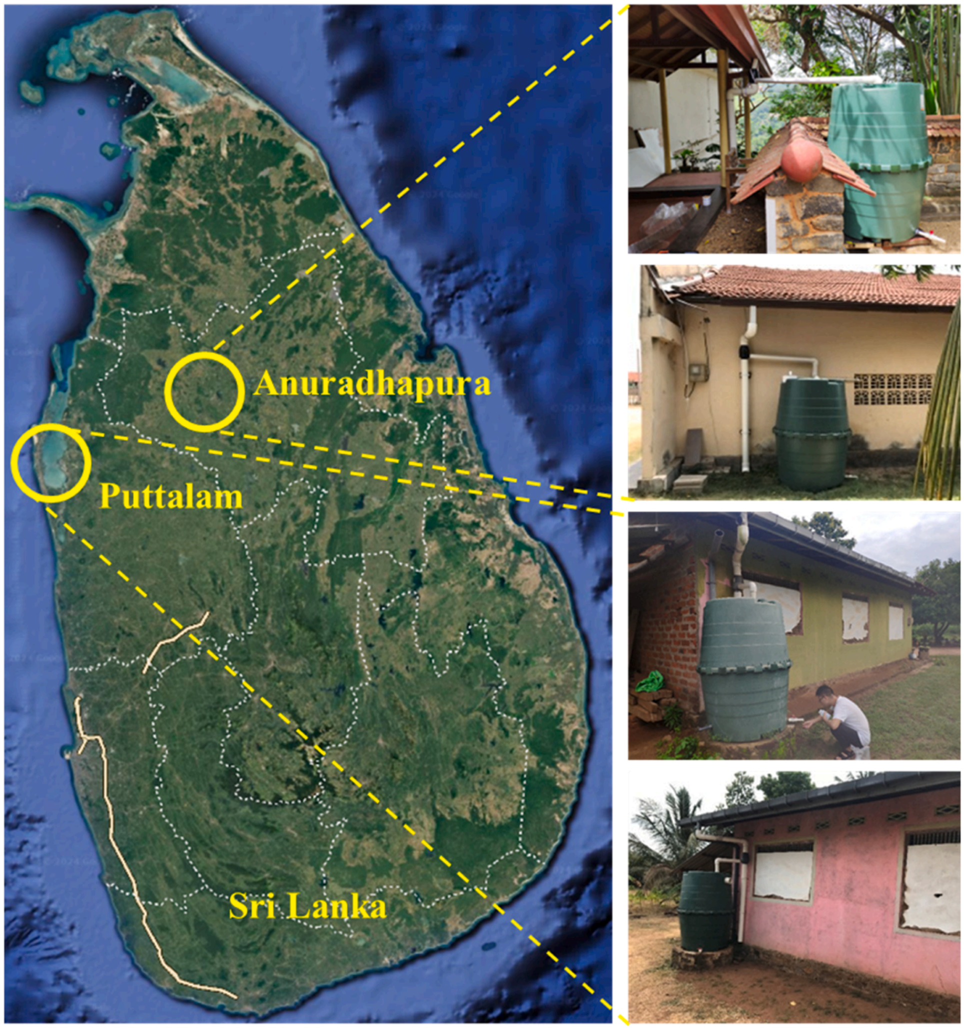

Fig. 1. Geographic location of rainwater quality monitoring sites in the dry zones of Sri Lanka and sampling points in Puttalam and Anuradhapura regions.

2.3. Determination of event mean concentration in rainwater

The pollutant loading patterns were quantified using event mean concentrations (EMCs), calculated as:

$$EMC = \frac{\int C_t Q_t d_t}{\int Q_t d_t} \quad (1)$$

where C_t and Q_t represent the instantaneous concentration of water quality parameter and flow rate at time t, respectively. The units for C_t vary depending on the specific water quality parameter as detailed in

Table 1
Comparison of EMCs and BCs of water quality parameters with Sri Lankan drinking water standards (SLS-614:2013).

Parameters	Units	E1		E2		E3		E4		E5		SLS-614:2013
		EMCs	BCs	EMCs	BCs	EMCs	BCs	EMCs	BCs	EMCs	BCs	
Colour	Pt-Co	82.52	6.00	15.62	2.00	88.67	20.00	118.93	21.00	32.01	6.00	15
Turbidity	NTU	17.01	0.20	6.43	0.14	13.23	0.81	18.87	0.68	2.17	0.15	2
pH	-	6.31	6.17	5.56	4.91	6.17	6.34	6.10	6.41	6.00	6.53	6.5–8.5
Ammonia	mg/L	0.12	0.05	0.16	0.04	1.19	0.57	1.04	0.62	0.21	0.14	0.2
COD	mg/L	9.32	2.32	3.91	1.28	10.11	7.74	12.89	10.13	5.57	3.71	10
Cl	mg/L	2.56	0.54	0.47	0.33	1.42	1.04	1.59	1.20	0.41	0.31	250
Cr	μg/L	0.23	0.14	0.37	0.12	0.43	0.14	0.29	< 0.08	0.13	< 0.08	50
Mn	μg/L	2.68	1.65	5.45	1.15	1.14	0.62	1.89	0.24	1.34	< 0.12	100
Fe	μg/L	19.02	1.62	4.33	2.47	19.22	8.10	13.25	3.14	9.19	1.11	300
Ni	μg/L	0.34	0.20	0.14	0.02	0.39	0.08	0.35	0.10	0.12	< 0.05	20
Cu	μg/L	1.57	0.76	0.75	0.27	2.15	0.88	2.22	0.20	0.89	0.10	1000
Zn	μg/L	16.68	7.09	12.18	3.87	6.46	0.54	6.97	0.61	6.13	0.27	3000
As	μg/L	0.19	< 0.03	0.16	< 0.03	0.24	0.20	0.23	0.21	0.09	0.06	10
Pb	μg/L	0.41	< 0.07	0.14	< 0.07	0.34	0.07	0.31	0.09	0.22	< 0.07	10
Total coliforms*	CFU/100 mL	65	3	48	2	100	46	117	43	36	23	10

* Total coliform bacteria shall not exceed 10 in any 100 mL sample for decentralized water supplies in rural.

Table 1, while Q_t is expressed in L/s. The calculated EMCs were compared against Sri Lankan national drinking water standards (SLS-614:2013) to assess the suitability of harvested rainwater for potable use and evaluate regulatory compliance. The relationships between water quality parameters and rainfall characteristics were examined through multivariate statistical techniques, including factor analysis and Pearson correlation coefficients. All statistical analyses were conducted using IBM SPSS Statistics 29.0.

2.4. Source identification of pollutants in rainwater

A positive matrix factorization (PMF) analysis (carried out by USEPA PMF 5.0) was performed to identify the major pollutant sources in harvested rainwater from the rural dry zones of Sri Lanka. Following the approach described by Paatero and Tapper [19], fifteen water quality parameters were selected as model inputs. The objective of PMF was to minimize the objective function Q, given by:

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\left[\left(\frac{1}{U_{ij}}\right)\left(C_{ij} - \sum_{k=1}^{p} g_{ik}f_{kj}\right)\right] \quad (2)$$

where C_{ij} and U_{ij} are the measured concentration and estimated uncertainty of species j in sample i, g_{ik} is the factor score (source contribution) of factor k to sample i, f_{kj} is factor loading (source profile), and n, m, and p represent the number of samples, species, and sources respectively.

The uncertainties were calculated based on method detection limits and a conservative 10 % measurement error using the following equations:

$$Conc. \leq MDL : Uncertainty = 5/6MDL \quad (3)$$

$$Conc. > MDL : Uncertainty = \left([Error \times Conc.]^2 + [0.5MDL]^2\right)^{0.5} \quad (4)$$

Data quality was assessed using the signal-to-noise (S/N) ratio, classifying each parameter as strong, weak or bad, and adjusting uncertainties accordingly. Any outlier values exceeding four standard deviations from the mean were assigned a substantially larger uncertainty to reduce undue influence on the results. The number of factors (i.e., pollutant sources) was determined through an iterative process, testing solutions with two to eight factors. For each potential solution, we examined (1) whether Q_{robust} was approximately equal to Q_{true}; (2) whether predicted-versus-observed plots showed acceptable correlations with standardized residuals mostly lying between −3 and + 3; (3) whether factors could be feasibly attributed to known emission or deposition processes in the study area [20,21]. We also varied the rotational freedom parameter (F_{peak}) to explore potential alternative rotations but observed no tangible improvement over the base run. Additionally, model stability was further checked using the built-in bootstrap (BS), displacement (DISP), and bootstrap displacement (BS-DISP) methods [22,23].

2.5. Overall assessment of rainwater quality with water quality index method

The comprehensive water quality assessment was conducted using the Water Quality Index (WQI) methodology following the classical Brown WQI approach [24]. The calculation process involved three sequential steps:

First, the quality rating (q_i) for each parameter was calculated by comparing the measured concentration (C_i) with its corresponding drinking water standard value (Si):

$$q_i = (C_i/S_i) \times 100 \quad (5)$$

The relative weight (w_i) of each parameter was then determined based on its standard limit:

$$w_i = 1/S_i \quad (6)$$

Finally, the overall WQI was computed as the weighted average of all parameters:

$$WQI = \sum q_i w_i \Big/ \sum w_i \quad (7)$$

where C_i represents the measured concentration of each analyzed parameter, S_i denotes the corresponding drinking water standard value based on Sri Lanka drinking water standards or WHO guidelines [25], q_i indicates the quality rating scale (0−100), w_i represents the relative weight of each parameter, and $\sum w_i$ signifies the sum of all relative weights. The resulting WQI values were interpreted as follows: excellent (<50), good (50−100), poor (100−200), very poor (200−300), and unsuitable for drinking (>300) [26].

3. Results and discussion

3.1. Characteristics and source identification of harvest rainwater in the rural dry zones of Sri Lanka

To characterize the water quality of roofing rainwater from the rural dry zones of Sri Lanka, a comparison of EMCs and BCs of selected water parameters with the Sri Lanka drinking water standards was carried out, and its result is presented in Table 1. For all the rainfall events, the EMCs and BCs of heavy metals including Cr, Mn, Fe, Ni, Cu, As, and Zn as well as the concentrations of Cl, fully meet the requirements for drinking water, relating to the lower industrialization and less pollutant emission in the rural dry zones. Nevertheless, the problems of rainwater quality are mainly correlated with the organoleptic parameters, microbiological indicators and ammonia. As for both studied sites, exceedances of colour and turbidity commonly occurred in roof runoff, the highest colour and turbidity in Puttalam and Anuradhapura were 82.52 Pt-Co, 17.01 NTU and 118.93 Pt-Co, 18.87 NTU, respectively, nearly 5–9 times over the limited values. Additionally, the BCs of colour in E3 and E4 were also over the standards, relating to the substantial wet deposition flux of colour in Anuradhapura. In terms of pH, the EMCs and BCs in all the events were slightly acidic and lower than the standard neutral value presumptively owing to the dissolution of atmospheric carbon dioxide gas. On the other hand, it was observed that the ammonia concentrations in roof runoff and rainwater collected from Puttalam and Anuradhapura exceeded the SLS-614:2013 permissible limit of ammonia (0.2 mg/L) for drinking purposes, especially the EMCs and BCs of E3 and E4. As to organic matter, the EMCs of COD in E3 and E4 were higher than the standard, contributing to roofing runoff pollution in Anuradhapura.

Compared with the concentration of roof runoff and raw rainwater, significant differences ($p < 0.05$) were observed in most of the selected parameters, signifying that those pollutants mainly originated with dry deposition. Conversely, the minor differences of EMCs and BCs for the ammonia, COD, Cl and As were observed in Anuradhapura, the background concentrations accounted for 58.1 %, 73.9 %, 74.8 % and 80.4 % of EMCs, respectively. It was reported that the atmospheric pollutants generated from agricultural activities, livestock breeding, agricultural wastes and abuse of fertilizers pose a great impact on the chemical composition of rainwater [27], extensive farming in Anuradhapura likely contributes to the elevated pollutant levels in raw rainwater.

The quality of harvested rainwater faces continuous challenges from various pollutants, particularly in regions with limited monitoring resources [28]. To identify major pollution sources and guide the selection of appropriate surrogate indicators, this study employed PMF analysis on fifteen water quality parameters collected from harvested rainwater in Sri Lanka's Puttalam and Anuradhapura dry zones (Fig. 2). Major components of factor 1 for the rainwater from Puttalam include ammonia, pH, and COD, while the dominant species of factor1 in the

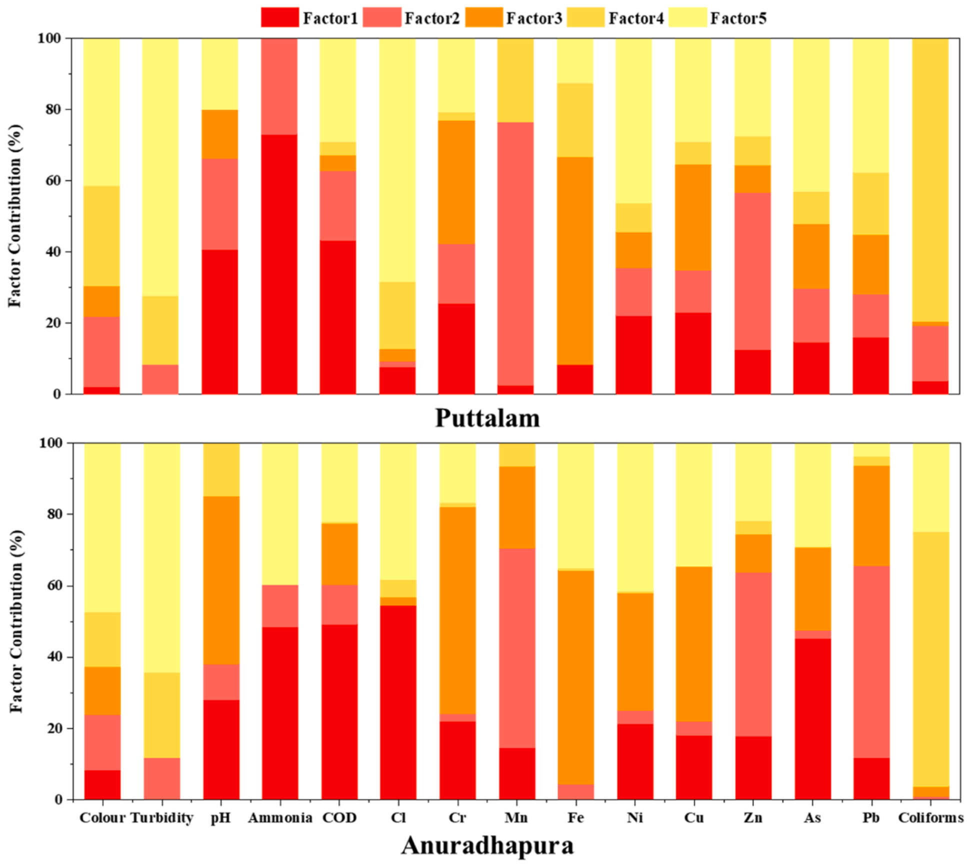

Fig. 2. Source profiles from positive matrix factorization analysis for rainwater quality data.

harvested rainwater from Anuradhapura were ammonia, Cl, As and COD. These components suggest contributions from agricultural sources [29], such as regional farmlands, farming activities, manure deposition and utilization of pesticides and fertilizer. The second factor consisted of Mn (73.9 %) and Zn (44.3 %) in the rainwater from Puttalam, and Mn (55.6 %), Zn (46.1 %), Pb (54.0 %) in that of from Anuradhapura, which had a relationship with the corrosion of plumbing system [30], wherein the galvanized gutter and downpipes could be the important contributor of Mn, Zn and Pd [31]. In terms of factor 3, it represents the pollutants from roofing materials with high contributions of Cr (34.5–58.0 %), Fe (58.5–60.0 %), and Cu (29.9–43.4 %) in the rainwater from both two sites as well as pH (47.4 %) in that of from Anuradhapura [30]. Commonly, those elements are mainly related to intensive industrial activities, whereas the degree of industrialization in the studied sites is relatively low. Therefore, their sources were much more related to the released constituents from the roof because those elements were reported to mainly originate from red clay tiles [32], which has been widely employed in the studied sites. In addition, Factor 4 is mainly loaded by total coliforms in both Puttalam (79.6 %) and Anuradhapura (71.5 %), while moderate contributions of colour and turbidity for factor 4 were also observed, suggesting that the microbes that appeared in roof rainwater were closely associated with resuspension of road dust [33,34]. Previous studies have implied that a large proportion of particulate matter from the roadside can be redistributed under wind force during the intervals between rainfall events, and accumulated on the roof surface, therefore, substantial microbes attached to the particulate matter can be transported into rainwater [35]. By the measurement of particle size distribution, over than 80 % of particles that appeared in the roofing rainwater from the studied site were smaller than 10 μm (Fig. S1), which means those particles can serve as a carrier with a great potential to transport the microorganisms from ambient environment to harvested rainwater. Hence, factor 4 was characterized as the resuspended road dust. As to factor 5, it consists mostly of colour (41.5–47.4 %) and turbidity (64.3–72.4 %) with respect to the rainwater from two studied sites in the dry zones of Sri Lanka, which is mainly attributed to the contribution of atmospheric deposition [36]. In summary, agricultural activities, plumbing systems, roofing materials, resuspended road dust and atmospheric deposition were the main sources of pollutants in the roofing rainwater of rural Puttalam and Anuradhapura.

PMF analysis revealed that over 80 % of the selected pollutants originated from multiple sources, necessitating careful selection of surrogate indicators for water quality assessment. While turbidity is traditionally used due to its correlation with suspended particles and ease of measurement, it primarily reflects only three pollution sources: atmospheric deposition (64.3–72.4 %), resuspended road dust, and plumbing system corrosion. This limited source representation reduces its reliability as a comprehensive water quality indicator. In contrast, colour demonstrated superior potential as a surrogate indicator by significantly correlating with all five major pollution sources: agricultural activities, plumbing system corrosion, roofing materials, road dust, and atmospheric deposition. Given its sensitivity to dissolved organic matter and influence on water's physical-biological properties, colour exhibits greater potential as a comprehensive surrogate indicator for water quality assessment.

3.2. Dynamic evolution of rainwater quality during the long-term storage process

Following the characterization of harvested rainwater quality and pollution sources, the temporal evolution of water quality during long-term storage was further investigated, which is crucial for ensuring sustainable drinking water safety in rural areas and tropical environments. The comprehensive monitoring program focused on five specific pollutants previously identified as primary concerns (turbidity, colour, COD, ammonia nitrogen, and total coliforms) across eight storage tanks in Puttalam and Anuradhapura regions (Fig. 3). Following roof collection and first-flush diversion, the stored rainwater underwent distinct quality transformation patterns, reflecting the complex interactions between various pollutants and natural purification processes.

Notably, conventional physicochemical pollutants including turbidity, colour, COD, and ammonia nitrogen exhibited significant reduction within the initial 28–35 days of storage, followed by stabilization in the subsequent period. This improvement can be attributed to natural ageing processes, including particle sedimentation, organic matter hydrolysis, and microbial biodegradation [37]. These natural processes effectively reduced turbidity, COD, and ammonia nitrogen to levels meeting drinking water standards, while colour levels approached the threshold. However, contrary to the improvement in physicochemical parameters, total coliform concentrations demonstrated a consistent upward trend, frequently exceeding the Sri Lankan drinking water standards in later stages. This divergent behaviour highlights the limitations of natural ageing processes in ensuring microbiological safety, particularly under tropical climatic conditions where elevated temperatures may accelerate bacterial proliferation.

To elucidate the underlying mechanisms governing these water quality dynamics, redundancy analysis (RDA) was employed to investigate the correlations between total coliforms and various physicochemical parameters throughout the storage period (Fig. 4). The RDA results revealed complex interaction patterns that help explain the observed water quality transformations. Specifically, total coliforms exhibited negative correlations with ammonia, colour, COD, Cl and Fe, suggesting potential inhibitory effects of these parameters on microbial growth. In contrast, several heavy metals (Mn, Ni, Zn, As, Pb and Cu) showed positive correlations with total coliforms, indicating synergistic interactions through biofilm formation and metabolic processes [38]. The orthogonal relationships observed between total coliforms and parameters such as pH and turbidity suggested their limited influence on microbial dynamics during storage.

These correlation patterns illuminate the intricate mechanisms controlling water quality evolution during long-term storage. The positive associations between total coliforms and heavy metals can be attributed to specific microbial-metal interactions: bacterial extracellular polymeric substances facilitate metal ion adsorption, while certain metal concentrations promote biofilm formation through enhanced cellular adhesion and matrix production [39]. Additionally, some metal ions serve as essential trace elements supporting bacterial metabolism and enzyme functions. Conversely, the negative correlations with ammonia, colour, COD and Cl suggest multiple growth suppression mechanisms. The organic substances and metal ions contributing to colour can affect bacterial viability through various pathways, including membrane potential disruption, cellular transport interference, and metabolic enzyme inhibition [14,40]. Under the specific pH and redox conditions typical of stored rainwater, these substances may generate reactive oxygen species, leading to oxidative damage of cellular components and subsequent growth inhibition [14].

3.3. Comprehensive quality assessment of roof-harvested rainwater for drinking water safety

To holistically evaluate the safety of harvested rainwater in Sri Lanka's rural dry zones, fifteen water quality parameters were integrated into the WQI framework. Initial analysis of five monitored rainfall events (E1–E5) revealed WQI values exceeding 100 in four events (E1–E4), categorizing the water as "poor" to "very poor" for drinking (Fig. 5(a)). Elevated concentrations of turbidity, colour, ammonia, COD, and total coliforms were identified as the primary drivers of this degradation, collectively contributing 78–92 % to the composite WQI scores (Fig. 5(b)). However, the contributions of specific pollutants on WQI were different, wherein the microbes and particle matters were the main pollutant species of roof runoff in the rural Puttalam, ammonia nitrogen and turbidity were predominant factors affecting WQI values in the rainwater from Anuradhapura, attributable to site-specific conditions [41].

The WQI analysis of stored rainwater further reveals the impact of natural purification processes during the long-term storage process. As depicted in Fig. 5(c) and 5(e), WQI values decreased progressively over storage time, with Puttalam showing a 20 % reduction (from 113 to 90) within 35 days, while Anuradhapura, despite having much poorer initial water quality, demonstrated a more dramatic 49.4 % improvement (from 395 to 200) within just 30 days, with both eventually dropping below 100. This significant improvement is largely attributable to natural processes-sedimentation, adsorption, and organic matter degradation-that reduce physicochemical pollutants. Nevertheless, the relative impact of various parameters evolved; as shown in Fig. 5(d) and (f), the dominant pollution profile shifted from particulate and dissolved contaminants in the early storage phase to biological pollution in later stages.

Our assessment reveals that the quality of freshly harvested rainwater is primarily limited by elevated concentrations of turbidity, colour, microorganisms, ammonia nitrogen, and COD, most of which were associated with particulate matter [42,43]. Notably, suspended particles in rainwater runoff are predominantly fine. According to Stokes' Law, the settling velocity of these fine particles is proportional to the square of particle diameter, resulting in extremely slow settling rates for such microscale particles. Theoretical calculations indicate that complete settling of 10 μm particles under gravity alone would require approximately 24 h for a 1-meter water column, while particles smaller than 5 μm could take several days or longer. Therefore, current local practices that rely solely on 1–2 days of settling or basic sand filtration are fundamentally limited by these physical principles, potentially compromising the safety of rainwater intended for drinking due to incomplete particle removal. Furthermore, while natural maturation during long-term storage results in significant improvements in physicochemical parameters within approximately one month and brings most indicators close to or within drinking water source standards, the elevated temperatures common in tropical climates cause a late-stage rebound in microbial populations. This outcome highlights the limitations of natural ageing processes in controlling microbial contamination under such conditions, indicating that the entire rainwater harvesting system, from collection through storage, presents substantial safety challenges that require comprehensive optimization.

3.4. Evaluating colour as a surrogate for pollutant transport dynamics

Based on source identification and the need for reliable surrogate indicators, we analyzed the temporal variations of colour and turbidity during different rainfall events to select an optimal parameter for tracking pollutant dynamics, while revealing their underlying transport mechanisms. Fig. 6 illustrates the temporal variations in turbidity, colour, and rainfall distribution during five precipitation events across two locations. The rainfall patterns exhibited distinct characteristics between sites: E1, E2, and E5 demonstrated progressive intensification from low to high precipitation rates, while E3 and E4 displayed a declining pattern with initial high intensity followed by gradual reduction. Despite these contrasting precipitation patterns, pollutant dynamics in the runoff showed remarkably consistent behaviour across all events. Both turbidity and colour exhibited rapid initial increases during the first flush, followed by exponential decay toward baseline levels as rainfall continued. This phenomenon can be attributed to the first flush effect, where accumulated pollutants on rooftop surfaces and conveyance systems are rapidly mobilized into the runoff during the initial precipitation phase [43]. Subsequently, the combination of sustained rainfall and dilution effects led to a rapid decrease in pollutant concentrations until reaching steady-state conditions.

The observed high synchronicity between turbidity and colour

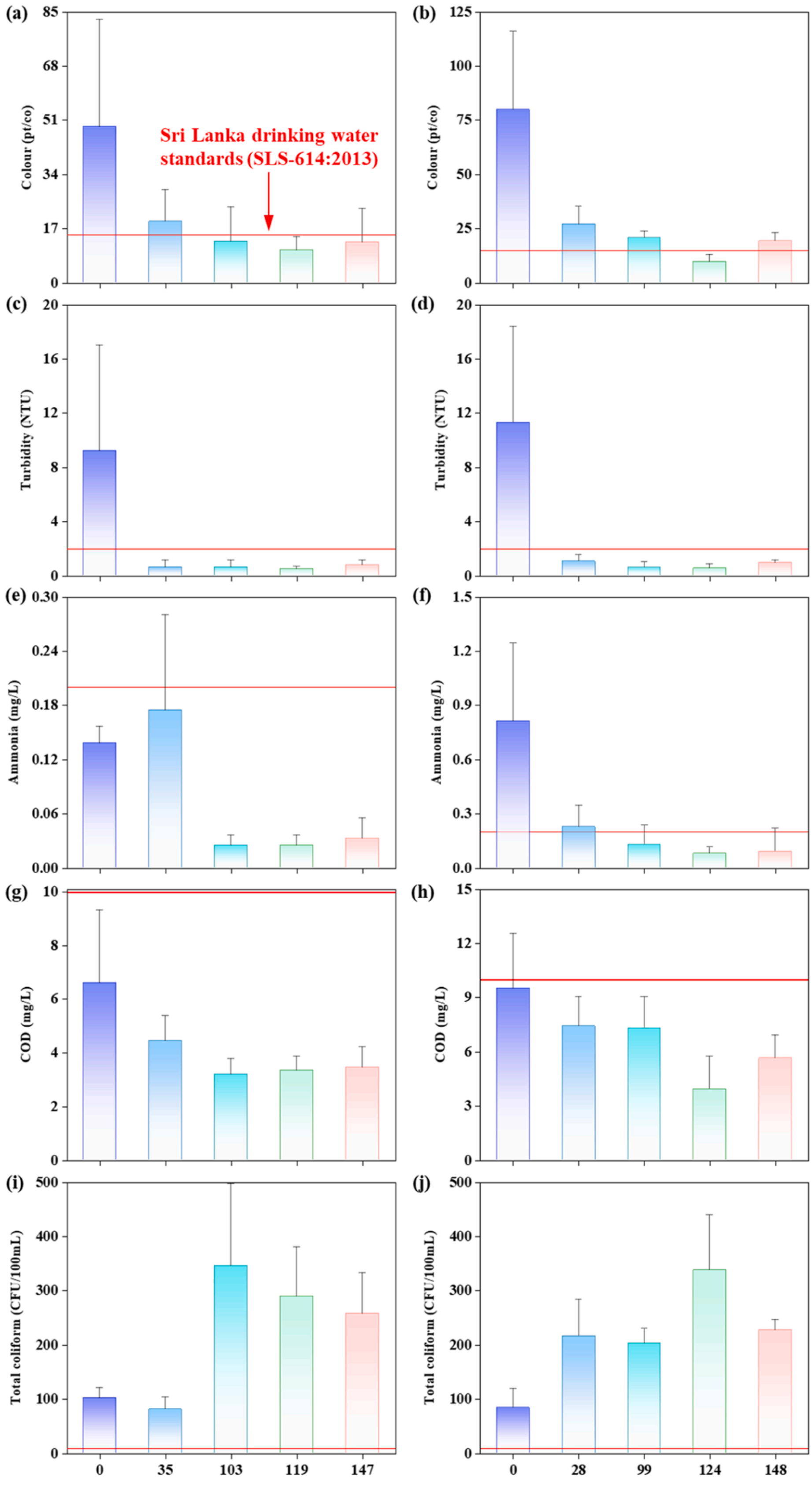

Fig. 3. Temporal dynamics of specific pollutant concentrations in stored rainwater at Puttalam and Anuradhapura: (a,b) colour; (c,d) turbidity; (e,f) COD; (g,h) ammonia nitrogen; (i,j) total coliforms. Left panels (a, c, e, g, i) represent Puttalam and right panels (b, d, f, h, j) represent Anuradhapura. Error bars indicate standard deviation (n = 4).

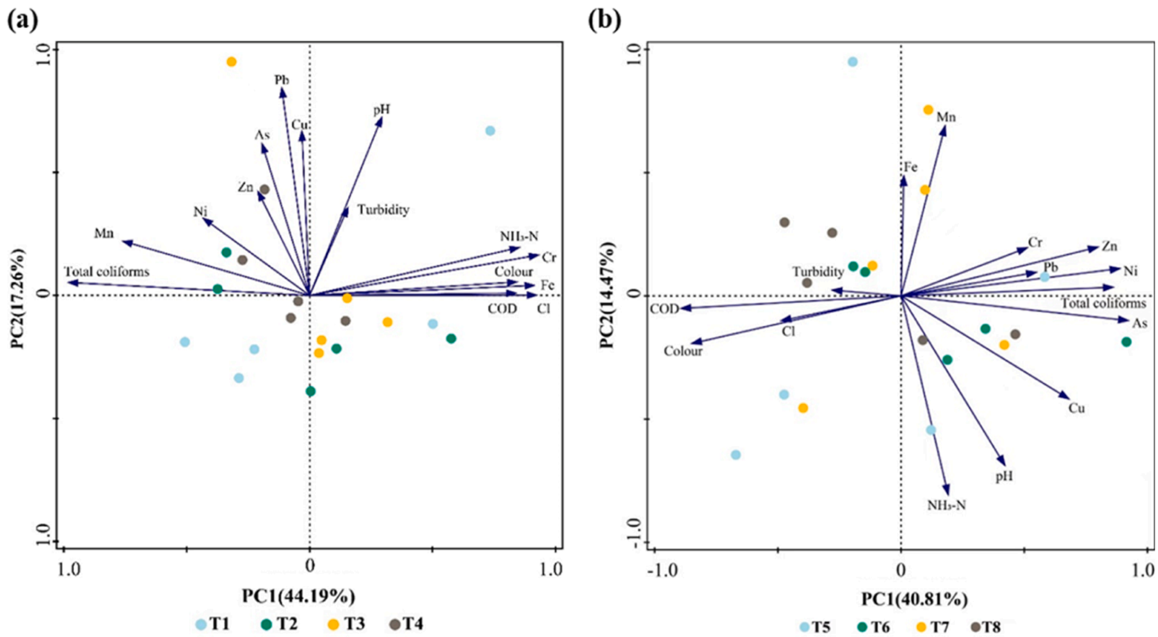

Fig. 4. RDA analysis of water quality parameters in long-term stored rainwater; (a) Correlation patterns between total coliforms and physicochemical parameters in Puttalam;(b) Correlation patterns between total coliforms and physicochemical parameters in Anuradhapura.

variations during rainfall events warrants in-depth investigation. PMF analysis revealed three overlapping sources contributing to both parameters: plumbing system corrosion, resuspended road dust, and atmospheric deposition. These shared sources create a fundamental basis for their synchronized variations, particularly through the behaviour of fine particles. Particle size analysis revealed that over 80 % of particles were smaller than 10 μm, with predominant distributions in the 0.6 μm and 1.1 μm ranges (Fig. S1). This fine particle-dominated distribution produces two significant effects: First, particles approaching or smaller than 0.45 μm can penetrate filter membranes, contributing to "true colour" measurements alongside dissolved substances. Second, these fine particles, with their large specific surface areas, enhance adsorption-desorption interactions with dissolved substances from the same sources, leading to synergistic migration behaviour. For instance, metal ions from plumbing system corrosion can both attach to suspended particles affecting turbidity and form dissolved complexes contributing to colour [14,36]. Similarly, organic matter associated with resuspended dust and atmospheric deposition can exist in both particulate and dissolved forms [44], further strengthening the correlation between these two parameters. This fine particle-mediated "bridging effect" between particulate and dissolved states explains the synchronized variations observed between turbidity and colour during rainfall events, despite their theoretically distinct physical states.

To quantitatively describe this phenomenon and verify its universality, we developed a mathematical model based on the "dry weather accumulation-rainfall wash-off" theory. The model simulates pollutant dynamics by coupling ADD with rainfall characteristics, focusing on particle accumulation and wash-off processes. Specifically, the accumulation process (Eq. 7) employs an exponential function to describe pollutant saturation with increasing ADD, where k_1 represents the accumulation rate and N_{max} denotes maximum accumulation [45]. This description aligns with observed pollutant accumulation patterns: rapid initial accumulation followed by gradual stabilization due to wind transport and biological degradation. The wash-off process (Eq. 8) incorporates rainfall (R) effects on pollutant migration, with k_2 characterizing wash-off efficiency related to rainfall intensity and surface properties [45]. Coupling these processes through Eq. 9 provides a comprehensive description of the accumulation-wash-off cycle while revealing the synergistic effects of antecedent dry conditions and rainfall characteristics on pollutant migration.

$$\text{Build}-\text{up process}: \ N_{initial} = N_{\max} \times \left(1 - e^{-k_1 \bullet t}\right) \tag{8}$$

$$\text{Wash}-\text{off process}: \ N_w = N_{initial} \times e^{-k_2 \bullet R} \tag{9}$$

$$\text{Integrated accumulation}-\text{wash off model}: \ N_w = N_{initial} \times \left(1 - e^{-k_1 \bullet t}\right) \times e^{-k_2 \bullet R} \tag{10}$$

As shown in Fig. S2, model validation in Puttalam and Anuradhapura demonstrated excellent performance (R^2 are 0.9614 and 0.9153; Adjusted R^2 are 0.9596 and 0.9125, respectively). This high-precision simulation not only confirms the model's robustness but also validates colour's effectiveness in characterizing pollutant migration and transformation in rainwater runoff. Notably, the model showed strong adaptability in predicting colour variations under different rainfall patterns ("low-to-high" and "high-to-low"), supporting colour's viability as a surrogate indicator. Correlation analysis (Table S3) revealed significant relationships between colour and other specific pollutants (including COD, ammonia, and microbial indicators), validating colour's comprehensive indicative function and providing reliable support for its practical monitoring applications.

Accordingly, these results demonstrate that compared to turbidity, the traditional surrogate indicator for rainwater runoff pollutants, colour not only encompasses a broader spectrum of pollutant sources but also effectively characterizes the dynamic transport processes of runoff pollutants, making it more suitable for determining first-flush diversion

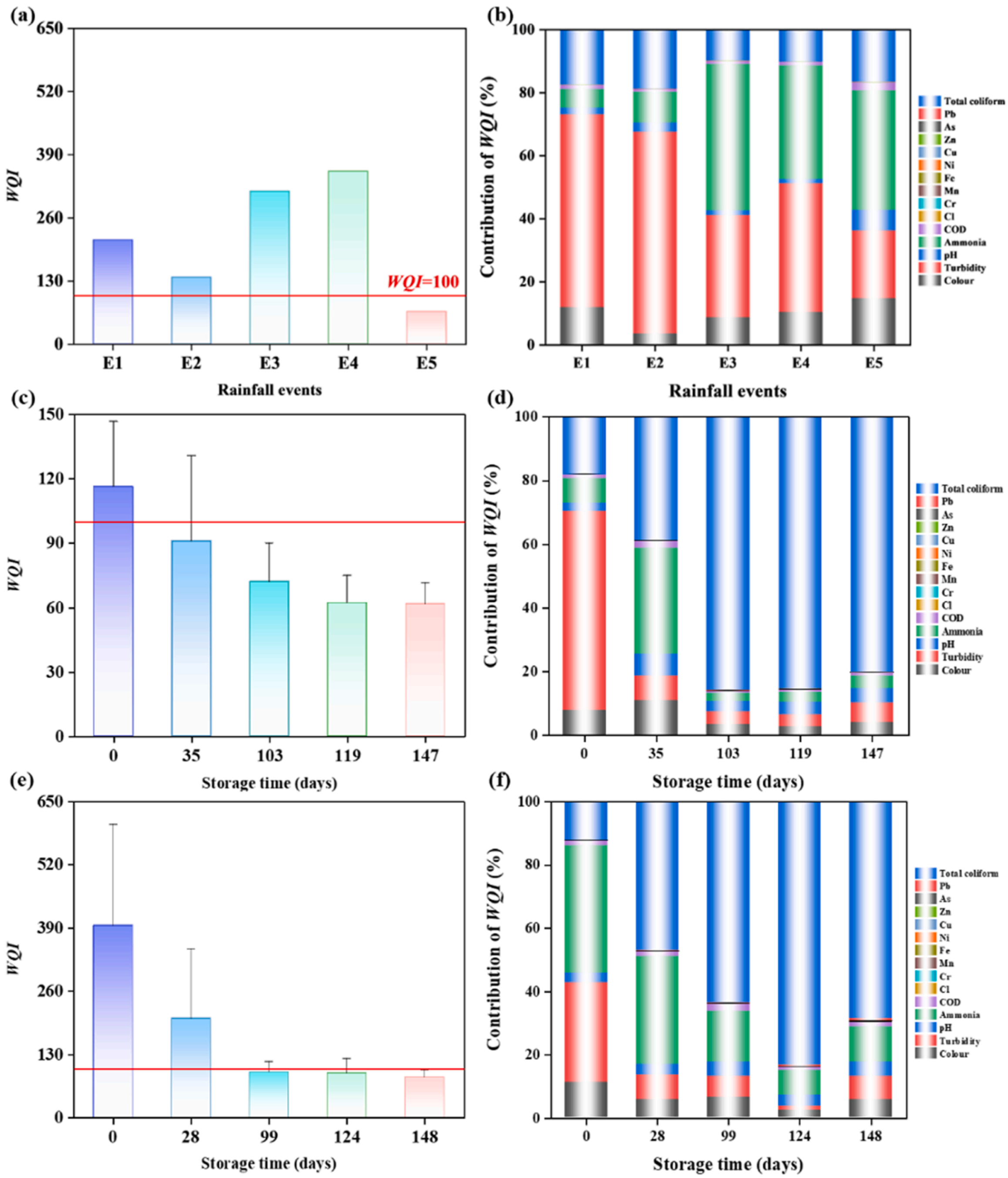

Fig. 5. Water Quality Index (WQI) analysis of harvested rainwater during rainfall events and long-term storage: (a,b) WQI values and relative pollutant contributions during rainfall events in Puttalam and Anuradhapura; (c,e) Temporal variation of WQI during storage in Puttalam and Anuradhapura, respectively; (d,f) Dynamic changes in relative pollutant contributions to overall WQI during storage n Puttalam and Anuradhapura, respectively.

schemes. Previous research indicates that the initial 1–2 mm of runoff rapidly washes and carries deposited particles from roofs or surfaces, creating turbidity peaks; however, soluble pollutants typically dissolve and release gradually throughout rainfall events, forming delayed peaks [46,47]. While conventional approaches recommend using the inflexion point during turbidity peak decline as the diversion criterion, this method may not adequately address soluble pollutant transport patterns, which typically manifest after the turbidity peak due to several mechanisms: initial rainfall primarily facilitates mechanical flushing of accumulated dust and particles, whereas soluble pollutants undergo a relatively slower dissolution process involving sufficient contact with rainwater, the gradual dissolution of roofing materials, and secondary dissolution of particles after being washed off, with continuous rainfall and varying intensities further influencing the release of soluble substances. Colour responds to both soluble and particulate pollutants' temporal migration characteristics, thereby offering crucial guidance for determining first-flush diversion volumes and effectively enhancing collected rainwater quality, which is particularly significant for ensuring the safety of rainwater intended for drinking purposes.

3.5. *Colour-based assessment of microbial-physicochemical changes in long-term stored rainwater*

Building on the established correlation between colour evolution and

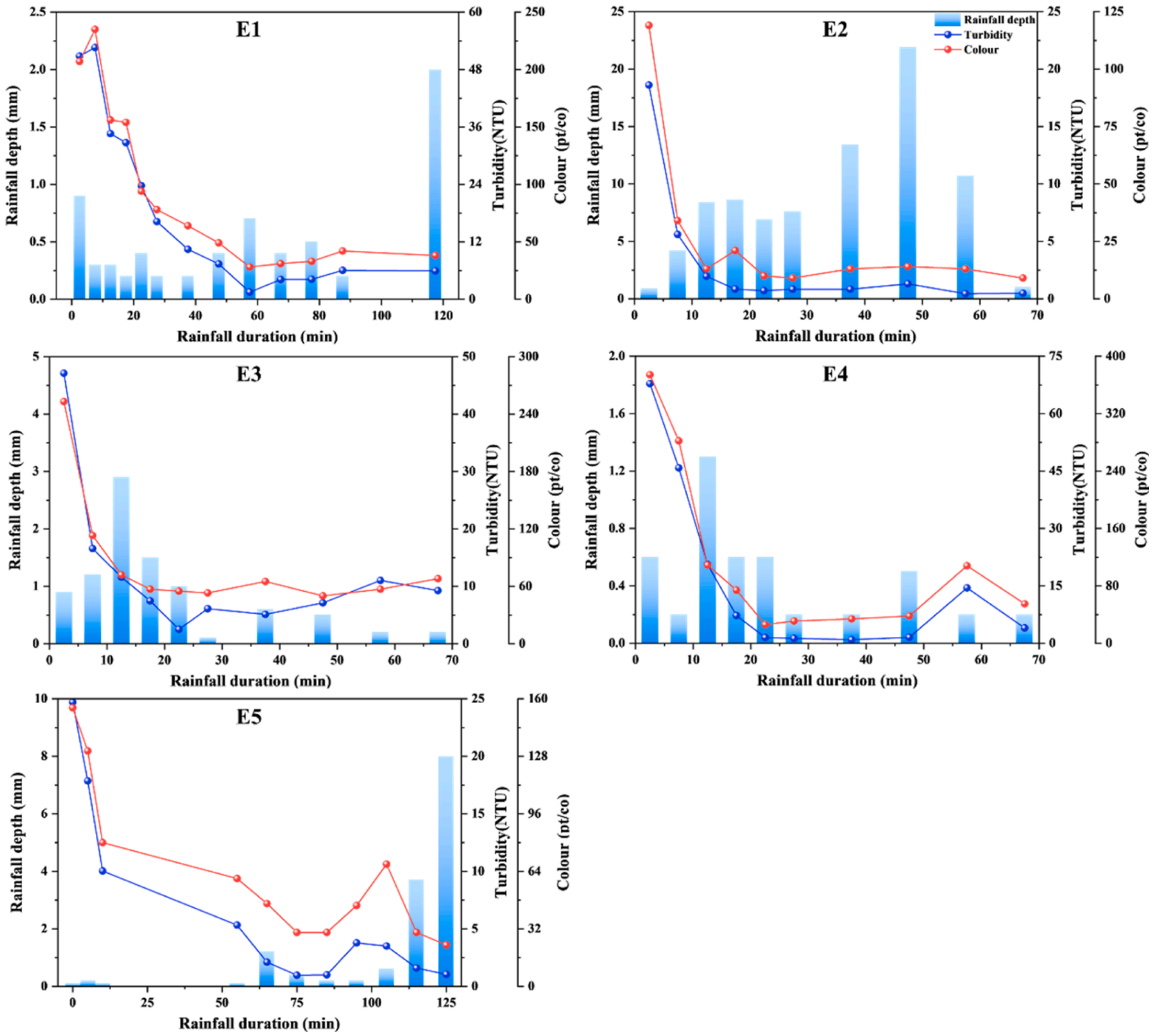

Fig. 6. Temporal variations of turbidity, colour, and rainfall intensity during monitored precipitation events: comparative analysis between Puttalam (E1, E2, E5) and Anuradhapura (E3, E4).

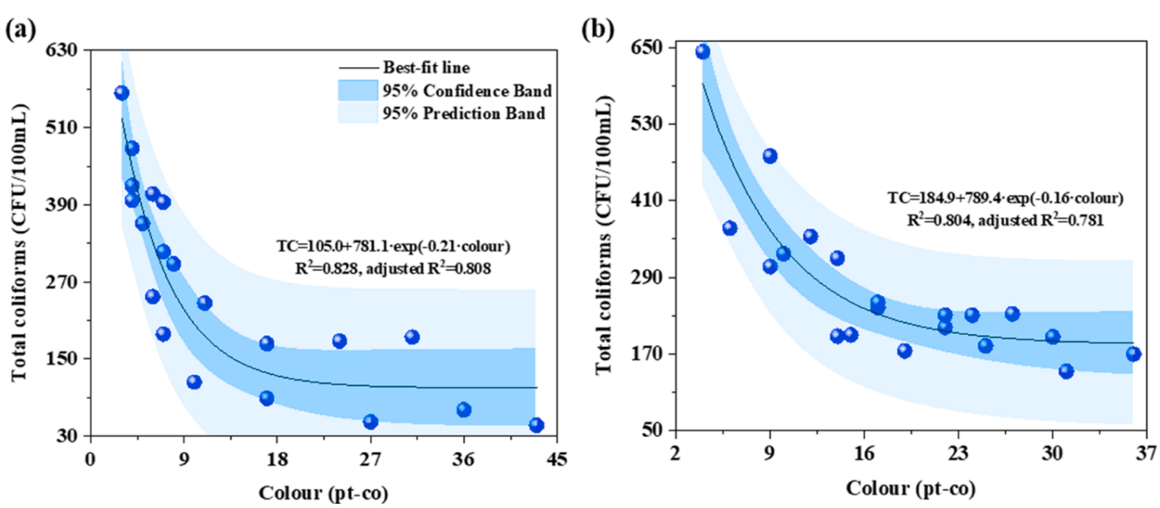

Fig. 7. Exponential decay relationship between colour and total coliform concentrations during the long-term storage process; (a) stored rainwater in Puttalam; (b) stored rainwater in Anuradhapura.

pollutant dynamics in stored rainwater, we further explored its relationship with microbial dynamics during long-term storage. Both study locations exhibited consistent negative correlations between total coliforms and colour,. The relationship between colour and total coliforms (Fig. 7(a) and (b)) demonstrates the cumulative inhibitory effects of colour-related components. As colour intensifies through particle desorption and leaching processes, microbial populations experience sharp declines before stabilizing at lower levels. This phenomenon reflects the complex interactions between dissolved organic matter and microbial communities in stored rainwater systems. The colour-contributing substances, primarily composed of humic and fulvic acids, may influence microbial dynamics in stored rainwater [48]. These substances can form stable complexes with bacterial membrane proteins through their abundant functional groups (e.g., carboxyl and phenolic groups), thereby disrupting membrane integrity and cellular homeostasis [49]. Additionally, the quinone moieties within these organic compounds serve as electron acceptors/donors in redox reactions, potentially generating reactive oxygen species that cause oxidative damage to bacterial cells. The progressive accumulation of these bioactive compounds creates an increasingly hostile microenvironment, evidenced by the observed exponential decline in coliform populations as colour intensity increases.

The RDA results reveal distinct regional patterns: Puttalam's colour correlates with Cr, iron, Cl, COD and ammonia, while Anuradhapura shows strong associations with COD and Cl. These parameters initially demonstrate stronger connections to turbidity in harvested rainwater, suggesting a primary association with particulate matter. During extended storage, progressive dissolution and desorption processes increase colour intensity while altering the chemical environment. The released substances can form various complexes and reactive species, creating multiple barriers to microbial growth through combined chemical and biological mechanisms. The progressive dissolution of particulate matter and subsequent changes in physicochemical properties establish colour as a more effective indicator than turbidity for evaluating microbial safety in long-term stored rainwater systems.

Microbial indicators are critical not only for assessing water source health risks and public hygiene but also for determining the safety of rainwater intended for drinking, a key objective of SDG 6. In many developing countries, particularly across tropical and subtropical regions, microbial proliferation in stored rainwater is a major concern, exacerbated by limited water quality monitoring in resource-constrained settings. In this study, the observed exponential decay between colour intensity and microbial indicators represents a significant breakthrough in rainwater quality assessment. Quantitative correlations between colour levels and total coliform reductions allow for the identification of critical colour thresholds, enabling cost-effective and straightforward colourimetric monitoring in communities that lack advanced testing infrastructure. The high intrinsic value of colour as an indicator lies in its capacity to encapsulate complex physicochemical processes that govern microbial dynamics during storage. However, the interpretation of colour-based data demands careful evaluation of local influences such as atmospheric deposition, container materials, and typical storage durations. Regular validation against standard microbiological methods is essential. Moreover, integrating colour metrics with complementary indicators such as odour, turbidity, and disinfection parameters offers a more robust framework to ensure rainwater meets the stringent quality requirements for potable use. Such an approach not only reinforces the reliability of water quality monitoring but also supports and accelerates progress toward achieving the sustainable development goals related to universal access to safe drinking water.

3.6. Optimized strategy for elevating rainwater safety as a drinking water source

Based on our analysis of rainwater quality and the effectiveness of colour as a surrogate indicator for multi-source pollutants, an integrated strategy is proposed to ensure safe drinking water from rainwater harvesting systems. This approach couples detailed source differentiation and dynamic water quality monitoring with cost-effective, low-maintenance treatment and storage interventions, thereby offering a robust framework particularly suited for resource-limited regions and tropical environments.

At the collection stage, optimizing the first flush diversion is paramount. The analysis supports a dual-threshold approach whereby diversion is initiated when colour readings exceed approximately 50 pt-Co (Fig. 6), and water is considered suitable for storage or subsequent use only once readings decline to around 20 pt-Co (Fig. 7). This surrogate-based monitoring method captures a broad spectrum of pollutants from agricultural runoff, roofing materials, and atmospheric deposition while providing a practical means to reduce the initial pollutant load and control monitoring costs. During the wet season, when rainfall is frequent, the efficiency of this strategy is especially critical for maintaining water quality.

Storage plays a critical role in further enhancing water safety, especially in the context of the Sri Lankan dry zone, where alternating six-month wet and dry seasons necessitate prolonged storage. Findings indicate that an extended storage period of approximately 30 days allows natural processes, including sedimentation, adsorption, and biodegradation, to substantially improve water quality. Drawing on traditional underground storage designs, such systems maintain lower water temperatures and reduce evaporative losses, thereby mitigating microbial proliferation under tropical conditions. Although natural maturation effectively lowers most physicochemical parameters toward acceptable drinking water standards, a modest post-storage treatment (e.g., boiling) remains advisable to address any potential rebound in microbial indicators during the dry season.

In the treatment phase, a two-stage treatment strategy is proposed based on the distinct water quality characteristics during collection and storage. For newly harvested rainwater dominated by fine particulates (>80 % smaller than 10 μm), which are difficult to remove through conventional 1–2 day settling or basic filtration, biological slow sand filtration is recommended as the primary treatment. This technology combines physical straining with biological treatment through the schmutzdecke layer, effectively removing both fine particles and their associated contaminants while maintaining low operational requirements. For stored rainwater where microbial contamination becomes the primary concern, particularly during extended storage periods in tropical conditions, gravity-driven membrane filtration serves as an effective secondary barrier. The dual-treatment approach, when integrated with proper storage management, provides comprehensive protection against both particulate and microbial contamination while remaining technically and economically feasible for resource-limited settings. This strategy significantly improves upon current local practices of brief settling or basic filtration, ensuring consistent water quality throughout both wet and dry seasons.

Finally, the implementation of a comprehensive yet simplified monitoring protocol is essential for sustainable operation. Emphasis is placed on easily measurable parameters such as colour, which has consistently demonstrated a reliable exponential decay relationship with microbial indicators. This Surrogate-Based Monitoring system enables real-time tracking of water quality from collection through storage, simplifies routine assessments in settings with limited technical capabilities, and supports timely operational adjustments to safeguard public health. While color assessment provides valuable benefits for water quality evaluation in extremely impoverished and resource-limited rural areas where comprehensive laboratory testing is unavailable, we acknowledge the inherent limitations of relying on any single parameter for complete microbial safety assessment. Therefore, regardless of color readings, boiling harvested rainwater before consumption is strongly recommended as a mandatory safety measure. Simple and cost-effective disinfection methods such as bringing water to a rolling boil for 1–3 min [25,50], or utilizing abundant solar energy in tropical regions through solar disinfection (exposing clear plastic bottles to direct sunlight for 6 h) [51,52], are particularly suitable for resource-constrained rural communities where conventional disinfection equipment or expensive disinfectants are not readily accessible.

Overall, this integrated strategy leverages rigorous water quality evaluation and surrogate monitoring to offer a practical and scalable pathway toward achieving safe rainwater use. By combining effective source control, adaptive treatment and storage interventions, and streamlined monitoring, the framework provides robust support for achieving SDG 6 and demonstrates a viable pathway tailored for tropical, resource-limited regions with long-period dry seasons where safe drinking water remains a critical concern.

4. Conclusion

This study investigated rainwater quality evolution and improvement strategies in rural dry zones of Sri Lanka. The results revealed that harvested rainwater exhibits significant contamination from turbidity, colour, total coliforms, ammonia nitrogen, and COD, primarily originating from multiple sources including agricultural runoff, infrastructure corrosion, roofing materials, road dust, and atmospheric deposition. Fine particles (<10 μm) were identified as critical pollutant carriers in this system. Quantitative assessment using Water Quality Index revealed initially poor rainwater quality with WQI values ranging from 113 to 395 across different study sites. Natural purification during 30-day storage demonstrated substantial improvement through synergistic sedimentation, adsorption, and biodegradation processes, achieving WQI reductions of 20–49 % and elevating overall water quality from an initial "poor" state to near drinking water standards after 30–100 days of storage. However, microbial indicators suggested a shift from particle-associated to microbiological contamination under tropical conditions. The study identified water colour as a superior surrogate indicator over turbidity for rural communities in dry zones of Sri Lanka, effectively reflecting both particulate and soluble pollutant dynamics in rainwater harvesting process, and enabling quantitative characterization of microbial changes in rainwater during long-term storage. Based on these findings, an integrated strategy combining source control, appropriate storage treatment, and a colour-based simplified monitoring system was developed, providing a practical solution for ensuring the safety and quality of rainwater used as drinking water in resource-limited regions with alternating wet-dry tropical seasons.

CRediT authorship contribution statement

Qingke Yuan: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Guoqing Yao:** Methodology, Investigation, Formal analysis, Data curation. **Wenbin Liu:** Methodology, Investigation, Formal analysis, Data curation. **Sujithra K. Weragoda:** Writing – review & editing, Resources, Methodology, Investigation. **Rohan Weerasooriya:** Writing – review & editing, Resources, Methodology, Investigation. **Meng Ying:** Writing – review & editing, Supervision, Methodology, Formal analysis, Data curation, Conceptualization. **Fubo Luan:** Writing – review & editing, Supervision, Resources, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jece.2025.117632.

Data availability

Data will be made available on request.

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