

Hydrochemical gradients and seasonal compartmentalization of legacy organochlorine pesticides in tropical estuarine water and sediments

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Abstract

Organochlorine pesticides (OCPs) remain mobile in tropical estuaries through runoff, particle transport and sediment remobilization. Paired surface-water and surface-sediment samples were collected for one year at twelve stations in five river–estuary systems on the South Coast of Kenya. The study generated 144 paired station–month observations for each matrix. Fifteen identified OCPs and one unresolved chromatographic signal (HCN*) were retained for mixture analysis; HCN* was excluded from compound-specific interpretation and from the $\Sigma 15\text{OCP}$. Mean concentrations ranged from 0.661 to 0.819 $\mu\text{g L}^{-1}$ in water and 1.614 to 1.731 $\mu\text{g kg}^{-1}$ dry weight in sediment. Centered-log-ratio principal component analysis showed multidimensional mixtures, with the first two axes explaining 33.8% of water and 29.0% of sediment variation. PERMANOVA detected seasonal and estuary-by-season structure in water profiles, whereas sediments showed estuarine and interaction effects. Crossed mixed-effects models estimated approximately 8.18-fold and 13.47-fold higher $\Sigma 15\text{OCP}$ burdens during the long rains than short rains in water and sediment, respectively. Dissolved oxygen was negatively associated with water burden. Partial redundancy analysis attributed 9.1% of residual water-mixture variation to hydrochemistry, with dissolved oxygen and pH contributing significant unique fractions. Paired total burdens were strongly correlated (Spearman $\rho = 0.856$), but complete mixture configurations were moderately concordant. The findings indicate that seasonal hydrological forcing regulates contaminant loading, whereas local estuarine conditions influence mixture composition, environmental partitioning and water–sediment coupling. There is need to integrate monitoring of water and sediment compartments for understanding the transport, retention and ecological exposure potential of legacy OCPs in tropical estuarine ecosystems.

1. Introduction

Organochlorine pesticides (OCPs) remain a prominent class of legacy contaminants because environmental persistence outlasts the regulatory decisions that restricted their manufacture and use. Their resistance to degradation, semivolatility and affinity for organic phases allow residues to circulate among soils, rivers, suspended particles, estuarine sediments and biota for decades. Climate-driven changes in rainfall, runoff and sediment disturbance can further reactivate historical stores, so contemporary detection does not necessarily indicate recent application but does demonstrate continuing exposure potential (Noyes et al. 2025). Recent studies from African and Asian waters continue to report complex OCP mixtures in both water and sediment, including Kenyan river systems, Lake Victoria tributaries and tropical wetlands (Hu et al. 2024; Ogola et al. 2024; Kagoya et al. 2026). Because OCPs remain detectable long after their use has ceased, understanding the environmental controls governing their transport, retention and remobilization has become increasingly important for risk assessment and environmental management.

Estuaries are especially important in the environmental cycling of persistent pollutants because they combine freshwater discharge, tidal exchange, particle flocculation, organic-matter processing and repeated sediment resuspension. Although estuaries are often regarded as terminal sinks for persistent

contaminants, growing evidence demonstrates that they function as dynamic storage and exchange zones capable of both retaining and releasing legacy pollutants. Recent Marine Environmental Research studies illustrate the importance of treating sediments, suspended particles and resident organisms as dynamically linked compartments: pollutant bioavailability changes with sediment chemistry, particle composition, redox state and hydrodynamic setting, and total concentrations alone may not predict biological exposure (Benthofage et al. 2023; Kwasigroch et al. 2023; Liu et al. 2023; Pereira et al. 2023).

The strength and direction of these processes vary seasonally. Rainfall can mobilize residues from cultivated soils, obsolete storage areas, settlements and channel margins, while high discharge increases erosion and particle transport. Conversely, dry and monsoon periods can increase residence time, salinity intrusion and sedimentation, creating conditions in which the water column and bed respond on different time scales. Studies of estuarine environmental variability show that seasonal changes in temperature, salinity, dissolved oxygen and freshwater input reorganize biological and chemical patterns, sometimes more strongly than fixed spatial gradients (Inyang et al. 2023; Bilbao et al. 2023; Sepahvand et al. 2023). A design that samples only once, or that pools seasons into an annual average, may therefore miss the periods when legacy residues become most mobile and ecologically available. Seasonally resolved monitoring is therefore essential for distinguishing transient hydrological effects from persistent contamination patterns and for identifying periods of heightened ecological risk.

Hydrochemical variables provide a mechanistic bridge between catchment forcing and contaminant behaviour within estuarine systems. Salinity and conductivity affect flocculation, ionic strength and the partitioning of hydrophobic compounds; pH influences surface charge and transformation pathways; temperature modifies volatilization and degradation; and dissolved oxygen is associated with redox conditions and organic-matter mineralization. Nutrients can also identify runoff, wastewater and productivity gradients that co-vary with contaminant inputs. The effects are rarely independent, and salinity does not have a universal relationship with pesticide mobility because its influence depends on dissolved organic carbon, suspended solids, compound hydrophobicity and sediment composition (Khouni et al. 2023). Consequently, hydrochemistry should be evaluated within a multivariate repeated-measures framework rather than through isolated bivariate correlations.

A distinction is also required between contaminant burden and mixture composition. The sum of identified OCPs describes the magnitude of exposure, whereas the proportional profile indicates which chemical groups dominate and may reveal differences in source history, transformation and phase partitioning. Consequently, evaluating both contaminant burden and mixture structure provides a more complete understanding of environmental exposure than consideration of total concentrations alone. Two matrices can be strongly correlated in total concentration but weakly concordant in composition if compounds differ in particle affinity, degradation rate or remobilization. Compositional methods are appropriate because concentrations expressed as a profile are constrained relative to one another; centered-log-ratio transformation and ordination avoid the spurious dependence created by analyzing raw proportions (Aitchison 1986; Legendre and Gallagher 2001). Recent coastal assessments likewise

emphasize that mixture structure, not only a single total, is necessary for interpreting ecological risk and source complexity (Wang et al. 2024; Zhang et al. 2023).

The Western Indian Ocean coast of Kenya contains mangrove-fringed river mouths and shallow estuaries that support fisheries, shoreline protection and local livelihoods. Historical work documented OCP residues in the Tana and Sabaki rivers, while more recent sediment surveys confirmed that banned compounds and transformation products remain measurable in Kenyan coastal environments (Lalah et al. 2003; Wanjeri et al. 2022). However, most Kenyan studies have been spatially limited, focused on individual river systems or relied on single-season sampling, restricting understanding of temporal variability and estuarine-scale processes. Comparable tropical estuaries show that legacy organic contaminants are retained in depositional zones but are redistributed by changing hydrology and particle transport (Zanardi-Lamardo et al. 2019; Montuori et al. 2020). Smaller South Coast systems, however, have received less repeated, paired water-sediment monitoring than the larger northern rivers.

Addressing this knowledge gap requires moving beyond annual mean concentrations to examine the interacting effects of seasonality, estuarine characteristics and hydrochemical variability on contaminant dynamics. Specifically, three management-relevant questions remain unresolved: whether contamination is driven primarily by regional seasonal forcing or persistent local sources; whether hydrochemical conditions modify the relative mixture after repeated station effects are controlled; and whether water and sediment provide redundant or complementary information. Recent marine environmental research investigations have shown that sediment-associated contaminants, biological uptake and environmental gradients can yield different spatial signals even within the same coastal system (Camacho-Jiménez et al. 2023; Han et al. 2024; Beyer et al. 2023). Paired observations are therefore needed to distinguish synchronous loading from compound-specific partitioning.

The present study analyzed monthly paired surface-water and surface-sediment samples from twelve stations in the Mapu, Mwena, Mkurumdzi, Ramisi and Uмба river-estuary systems. It tested four linked propositions: (i) that total OCP burden and mixture composition vary among hydrological seasons; (ii) that estuary identity modifies seasonal responses; (iii) that hydrochemistry explains a measurable fraction of water-mixture variation after station and season are conditioned; and (iv) that water-sediment coupling is stronger for total burden than for the complete compositional profile. Crossed mixed-effects models, compositional ordination, constrained permutation tests, partial redundancy analysis and concordance statistics were combined to provide a process-oriented account of legacy OCP dynamics in a tropical estuarine setting. The study represents one of the most comprehensive paired water–sediment assessments of legacy pesticide dynamics conducted in Kenyan estuarine ecosystems.

The study further distinguishes an unidentified chromatographic signal (HCN*) from the fifteen confidently identified compounds. Retaining the signal within exploratory compositional analyses preserves potentially informative analytical information, while excluding it from the primary Σ OCP calculations and compound-specific interpretation avoids biasing source attribution and ecological

inference. This approach provides a transparent framework for managing analytical uncertainty and aligns with best practice in environmental contaminant monitoring.

Tropical estuaries of the Western Indian Ocean remain underrepresented in global contaminant monitoring programmes despite supporting high biodiversity, productive fisheries and rapidly growing coastal populations. Their strong seasonal hydrology, extensive mangrove systems and increasing anthropogenic pressures create conditions that may differ substantially from those reported for temperate estuaries. Understanding contaminant dynamics in these systems is therefore important not only for local environmental management but also for improving global understanding of persistent pollutant behaviour in tropical coastal environments.

2. Materials and Methods

2.1 Study area and sampling design

The study covered the Mapu, Mwena, Mkurumdzi, Ramisi and Uмба river–estuary systems in Kwale County, Kenya. Twelve sampling stations were strategically selected to represent the salinity and hydrological gradient from freshwater-influenced reaches through transitional mixing zones to estuarine environments. Sampling was conducted monthly from September 2018 to August 2019, thereby encompassing the major hydroclimatic seasons of the Kenyan coast, namely the short rains (September–November), northeast monsoon (December–February), long rains (March–May) and southeast monsoon (June–August). At each station, surface-water samples and surficial (0–5 cm) biologically active sediment samples were collected during the same field visit to facilitate direct comparison between aqueous and sediment-associated contaminant dynamics. Three analytical replicates were obtained for each station–month combination and averaged prior to statistical analysis, yielding 144 observations for the water matrix and 144 observations for the sediment matrix. This repeated-measures design enabled assessment of both seasonal variability and estuary-specific differences in OCP occurrence and composition.

2.2 Hydrochemical and pesticide measurements

Hydrochemical measurements comprised temperature, dissolved oxygen, conductivity, total dissolved solids, salinity, pH, phosphate, nitrate and ammonia. Water was collected in pre-cleaned amber glass bottles, while surface sediment was collected from the biologically active upper layer. A 1 mL water aliquot and a 1 g dry-sediment aliquot were used for analytical preparation and concentration normalization. Water concentrations are reported as $\mu\text{g L}^{-1}$ and sediment concentrations as $\mu\text{g kg}^{-1}$ dry weight.

The analytical list comprised HCB, α -HCH, β -HCH, γ -HCH, heptachlor, heptachlor epoxide, cis-chlordane, trans-nonachlor, o,p'-DDE, p,p'-DDE, o,p'-DDD, p,p'-DDD, o,p'-DDT, p,p'-DDT and mirex. One additional chromatographic peak was consistently detected but could not be confidently assigned to any available

analytical standard and was therefore designated as HCN*. To preserve potentially informative environmental variation, HCN* was retained in descriptive visualizations and multivariate compositional analyses. However, it was excluded from calculations of Σ OCP concentrations and from compound-specific source and risk interpretations because its chemical identity could not be verified. Sensitivity analyses conducted with and without HCN* yielded qualitatively identical results, indicating that inclusion of the unidentified signal did not influence the overall conclusions of the study.

Extracts were quantified using gas chromatography with electron-capture detection and confirmatory retention-time and mass-spectral matching. Calibration standards, internal and surrogate standards, procedural blanks and matrix-spike checks accompanied the analytical batches. Concentrations below the limit of detection were not recorded as zero. Identification of target compounds was based on comparison of retention times and detector responses with certified analytical standards, while compounds lacking satisfactory confirmation criteria were classified as unidentified signals.

2.3 Statistical analysis

All analyses used station-month means to prevent analytical triplicates from being treated as independent environmental observations. The primary total burden, Σ OCP, was the sum of the fifteen identified compounds. HCN* was retained in mixture-composition analyses to preserve observed compositional variation but excluded from Σ OCP calculations and substantive compound interpretation because its chemical identity could not be verified.

OCP profiles were treated as compositional data. Values were transformed using the centered log-ratio transformation before principal component analysis (PCA), PERMANOVA and redundancy analysis (Aitchison 1986). Separate PCA ordinations were performed for water and sediment to visualize dominant patterns in mixture composition among estuaries and seasons. PERMANOVA used Euclidean distances on centered-log-ratio profiles and sequentially tested estuary, season and the estuary $\times$ season interaction. Estuary tests permuted complete station blocks, whereas seasonal and interaction tests used permutations constrained within stations (999 permutations). Homogeneity of multivariate dispersion among groups was evaluated using PERMDISP to verify that significant PERMANOVA results reflected differences in centroid location rather than differences in within-group dispersion (Anderson, 2001).

Total identified OCP (Σ OCP) burden was analyzed separately for water and sediment using crossed linear mixed-effects models on the natural-log transformed concentrations. Fixed effects included season, dissolved oxygen, pH, and a mineralization index derived from the first standardized principal component of conductivity, TDS, and salinity. The resulting index explained 83.3% of the combined variance of these highly correlated variables and was used to reduce multicollinearity among hydrochemical predictors. Random intercepts were fitted for station and month. Effect estimates were exponentiated and reported as multiplicative ratios with 95% confidence intervals. Marginal and conditional R^2 values were calculated to quantify the variance explained by fixed effects alone and by the full model including random effects, respectively (Nakagawa & Schielzeth 2013).

A partial redundancy analysis (RDA) was used to evaluate relationships between water-column OCP composition and hydrochemical conditions, including temperature, dissolved oxygen, mineralization, pH, phosphate, nitrate and ammonia, after conditioning on station and season. Overall and marginal significance were assessed by 999 within-station permutations. Adjusted variation partitioning was used to quantify the unique contributions of hydrochemistry, season and station identity, together with, their shared explained component and unexplained variation (Peres-Neto et al. 2006).

Water–sediment coupling was evaluated using compound-specific Spearman rank correlations for paired station–month observations, with Benjamini–Hochberg false-discovery-rate adjustment applied to account for multiple testing. Configuration-level concordance between water and sediment compositional profiles was assessed using Procrustes correlation and a Mantel test based on Euclidean distance matrices of standardized centered-log-ratio profiles. Statistical significance was evaluated using 999 permutations constrained within stations.

2.4 Data basis, reporting units and reproducibility

All inferential analyses used station–month means rather than the three within-event measurements, giving twelve stations by twelve months ($n = 144$ per matrix). Water concentrations are reported in $\mu\text{g L}^{-1}$ and sediment concentrations in $\mu\text{g kg}^{-1}$ dry weight, consistent with companion benchmark analysis. HCN* was retained in ordinations to preserve the observed mixture signal but excluded from $\Sigma 150\text{CP}$ calculations and compound-specific interpretation because its chemical identity could not be confirmed. The station–month dataset, analytical workflows, statistical scripts and laboratory QA/QC documentation were archived to ensure full reproducibility of data processing, transformations, constrained permutation procedures and sensitivity analyses.

3. Results

3.1 Hydrochemical setting

The station network encompassed a broad range of hydrochemical conditions across the five river–estuary systems. Temperature varied over a comparatively narrow tropical range, whereas conductivity, TDS, salinity and nutrients showed much wider variation (Table 1). The lowest conductivity ($190.57 \mu\text{S cm}^{-1}$) and salinity (0.10 ppt) were recorded at Mapu, while the highest conductivity ($5513.30 \mu\text{S cm}^{-1}$) occurred at Mkurumdzi Estuary and the highest salinity (6.99 ppt) at Mwena Estuary. Dissolved oxygen ranged from 4.95 to 8.52 mg L^{-1} and pH from 6.20 to 9.00. Phosphate concentrations ranged from 0.05 to 3.49 mg L^{-1} , nitrate from 0.02 to 1.28 mg L^{-1} and ammonia from 0.04 to 2.83 mg L^{-1} . Minimum and maximum values for individual hydrochemical variables occurred at different stations and sampling months (Table 1).

Table 1

Observed hydrochemical ranges and station–month combinations at the minima and maxima

Parameter	Observed range	Minimum station/month	Maximum station/month
Temperature (°C)	26.50–29.20	Mwena Estuary / Nov 2018	Mwena Estuary / Jul 2019
Dissolved oxygen (mg L ⁻¹)	4.95–8.52	Mapu / Oct 2018	Mwena Manda / Feb 2019
Conductivity (μS cm ⁻¹)	190.57–5513.30	Mapu / May 2019	Mkurumdzi Estuary / May 2019
TDS (mg L ⁻¹)	84.83–3158.27	Mapu / Feb 2019	Mwena Majoreni / Feb 2019
Salinity (ppt)	0.10–6.99	Mapu / Oct 2018	Mwena Estuary / Jun 2019
pH	6.20–9.00	Mkurumdzi Estuary / Jul 2019	Ramisi Mwachande / Sep 2018
Phosphate (mg L ⁻¹)	0.05–3.49	Umba Lunga-Lunga Bridge / Dec 2018	Mkurumdzi Gazi / Sep 2018
Nitrate (mg L ⁻¹)	0.02–1.28	Ramisi Mwachande / Nov 2018	Umba Lunga-Lunga Bridge / Mar 2019
Ammonia (mg L ⁻¹)	0.04–2.83	Mwena Manda / Jul 2019	Mkurumdzi Gazi / Aug 2019

3.2 Occurrence and descriptive concentrations of OCPs

All fifteen identified compounds and HCN* were detected in both matrices. Concentrations of individual compounds were broadly comparable within each matrix, although sediment concentrations consistently exceeded those observed in water (Table 2). Mean water concentrations ranged from 0.661 μg L⁻¹ for p,p'-DDD to 0.819 μg L⁻¹ for HCB, while sediment concentrations ranged from 1.614 μg kg⁻¹ dry weight for β-HCH to 1.731 μg kg⁻¹ dry weight for Mirex. Across compounds, variability was substantial, as reflected by standard deviations that were generally similar in magnitude to the corresponding means. Median concentrations were typically lower than mean concentrations in both matrices, indicating the influence of occasional elevated observations. Interquartile ranges overlapped extensively among compounds, suggesting relatively similar concentration distributions across the monitored OCPs.

The unidentified chromatographic signal, HCN* occurred throughout the dataset and exhibited mean concentrations of 0.723 μg L⁻¹ in water and 1.636 μg kg⁻¹ dry weight in sediment. Consistent with the analytical approach described in Section 2, HCN* was retained for descriptive and multivariate compositional analyses but excluded from ΣOCP calculations and compound-specific interpretation.

Spatial variability was evident for both water and sediment OCP concentrations (Figs. 3 and 4). However, no single compound consistently dominated across all stations, and concentration patterns varied

among estuaries and sampling locations.

Table 2

Descriptive statistics for identified OCPs and the unidentified chromatographic signal (HCN*) in paired surface-water and surface-sediment samples. Values are presented as mean $\pm$ standard deviation (SD) and median (interquartile range, IQR).

Compound	Water mean $\pm$ SD ($\mu\text{g L}^{-1}$)	Water median (IQR)	Sediment mean $\pm$ SD ($\mu\text{g kg}^{-1}$)	Sediment median (IQR)
HCB	0.819 $\pm$ 0.538	0.845 (0.351–1.101)	1.725 $\pm$ 1.452	1.289 (0.309–2.922)
HCN*	0.723 $\pm$ 0.529	0.755 (0.232–1.005)	1.636 $\pm$ 1.347	1.249 (0.329–2.905)
α -HCH	0.728 $\pm$ 0.499	0.775 (0.293–1.009)	1.720 $\pm$ 1.413	1.539 (0.386–2.894)
β -HCH	0.731 $\pm$ 0.468	0.765 (0.363–1.006)	1.614 $\pm$ 1.355	1.040 (0.342–2.906)
γ -HCH	0.695 $\pm$ 0.454	0.728 (0.326–0.996)	1.669 $\pm$ 1.391	1.460 (0.241–2.911)
Heptachlor	0.700 $\pm$ 0.470	0.737 (0.297–1.004)	1.717 $\pm$ 1.431	1.344 (0.450–2.895)
Heptachlor epoxide	0.731 $\pm$ 0.505	0.758 (0.269–1.008)	1.723 $\pm$ 1.395	1.466 (0.310–2.937)
cis-Chlordane	0.686 $\pm$ 0.442	0.687 (0.324–1.003)	1.693 $\pm$ 1.399	1.446 (0.374–2.931)
trans-Nonachlor	0.706 $\pm$ 0.461	0.698 (0.332–1.002)	1.665 $\pm$ 1.375	1.204 (0.428–2.957)
o,p'-DDE	0.710 $\pm$ 0.448	0.693 (0.354–1.005)	1.680 $\pm$ 1.405	1.299 (0.409–2.872)
p,p'-DDE	0.704 $\pm$ 0.471	0.636 (0.328–1.004)	1.681 $\pm$ 1.371	1.352 (0.395–3.002)
o,p'-DDD	0.733 $\pm$ 0.448	0.741 (0.396–1.005)	1.681 $\pm$ 1.412	1.463 (0.215–2.777)
p,p'-DDD	0.661 $\pm$ 0.549	0.549 (0.143–1.003)	1.705 $\pm$ 1.419	1.438 (0.266–2.990)
o,p'-DDT	0.703 $\pm$ 0.462	0.690 (0.324–1.002)	1.726 $\pm$ 1.398	1.523 (0.326–2.979)
p,p'-DDT	0.695 $\pm$ 0.454	0.670 (0.332–1.003)	1.687 $\pm$ 1.413	1.495 (0.258–2.992)
Mirex	0.701 $\pm$ 0.461	0.674 (0.302–1.005)	1.731 $\pm$ 1.397	1.499 (0.382–2.891)

HCN* denotes an unidentified chromatographic OCP signal. It is retained in figures and multivariate composition analyses but excluded from Σ OCP and compound-specific interpretation

3.3 Spatial and seasonal structure of OCP mixtures

Water mixture composition differed significantly among seasons and through an estuary-by-season interaction, but the estuary main effect was not significant under station-block permutation (Table 3). Season explained 8.1% of the variation in water OCP composition, while the estuary $\times$ season interaction accounted for a further 15.5%. Water dispersion also differed among seasons (PERMDISP $p = 0.001$), indicating that part of the seasonal PERMANOVA signal reflected changing within-season heterogeneity as well as centroid displacement.

Sediment composition differed among estuaries and showed a significant estuary-by-season interaction, while the season main effect was not significant. In contrast to water, sediment composition exhibited significant spatial structuring among estuaries. Estuarine dispersion was homogeneous in sediment (PERMDISP $p = 0.872$), supporting interpretation of the estuary effect as a difference in multivariate centroids. Seasonal dispersion was heterogeneous ($p = 0.001$), so the interaction was interpreted cautiously.

Table 3
PERMANOVA and PERMDISP results for centered-log-ratio OCP mixture composition

Matrix	Term	df	Pseudo-F	R ²	Permutation p
Water	Estuary	4, 139	2.460	0.066	0.311
Water	Season	3, 136	4.331	0.081	0.001
Water	Estuary $\times$ season	12, 124	2.299	0.155	0.001
Sed.	Estuary	4, 139	2.925	0.078	0.045
Sed.	Season	3, 136	0.985	0.020	0.422
Sed.	Estuary $\times$ season	12, 124	1.278	0.099	0.031
Water	PERMDISP: estuary	—	1.984	—	0.079
Water	PERMDISP: season	—	66.537	—	0.001
Sed.	PERMDISP: estuary	—	0.196	—	0.872
Sed.	PERMDISP: season	—	41.030	—	0.001

3.4 Mixed-effects models of total identified OCP burden

Total identified OCP burden showed strong seasonal structure in both matrices. Relative to the short rains, water Σ OCP was 5.14 times higher during the northeast monsoon, 8.18 times higher during the

long rains and 3.59 times higher during the southeast monsoon (Table 4). The largest seasonal increase was observed during the long-rains period. A one-standard-deviation increase in dissolved oxygen was associated with a 7.0% reduction in water burden. Mineralization and pH were not significant after station and month random effects were included.

Sediment Σ OCP was 9.78 times higher during the northeast monsoon and 13.47 times higher during the long rains than during the short rains. As observed in water, the largest seasonal increase occurred during the long-rains season. The southeast-monsoon contrast and hydrochemical covariates were not significant. Month-level variance greatly exceeded station variance in sediment, indicating greater temporal than spatial variability in sediment Σ OCP. Fixed effects explained 68.6% of water and 55.6% of sediment variance; conditional R^2 values were 97.5% and 97.7%, respectively. Including HCN* in the total burden produced the same directional and significance conclusions.

Table 4
Crossed mixed-effects models for total identified OCP burden. Ratios are exponentiated fixed-effect coefficients

Matrix	Fixed effect	Multiplicative ratio	95% CI	<i>p</i>
Water	Long rains vs short rains	8.18	3.77–17.76	< 0.001
Water	Northeast monsoon vs short rains	5.14	2.37–11.15	< 0.001
Water	Southeast monsoon vs short rains	3.59	1.65–7.80	0.001
Water	Dissolved oxygen (per SD)	0.93	0.89–0.98	0.003
Water	Mineralization index (per SD)	0.96	0.91–1.02	0.161
Water	pH (per SD)	0.98	0.94–1.01	0.183
Sed.	Long rains vs short rains	13.47	2.78–65.25	0.001
Sed.	Northeast monsoon vs short rains	9.78	2.02–47.39	0.005
Sed.	Southeast monsoon vs short rains	1.56	0.32–7.58	0.579
Sed.	Dissolved oxygen (per SD)	1.03	0.99–1.08	0.184
Sed.	Mineralization index (per SD)	1.04	0.98–1.10	0.168
Sed.	pH (per SD)	0.99	0.94–1.03	0.537

3.5 Principal component analysis of mixture composition

The first two centered-log-ratio principal components explained 33.8% of water and 29.0% of sediment mixture-composition variance (Figs. 5 and 6). No single component accounted for a large proportion of the total compositional variance in either matrix..

In water, PC1 was positively associated with o,p'-DDD, mirex and p,p'-DDT negatively associated with γ-HCH, cis-chlordane and heptachlor epoxide. PC2 contrasted p,p'-DDE and β-HCH with HCB, α-HCH and

p,p'-DDD. In sediment, PC1 was positively associated with trans-nonachlor, mirex, cis-chlordane and negatively associated with o,p'-DDE with γ -HCH, while PC2 contrasted p,p'-DDE and p,p'-DDD negatively associated with heptachlor epoxide, β -HCH and heptachlor. HCN* was included in the ordinations but was not assigned a chemical interpretation.

3.6 Hydrochemical constraints and variation partitioning

After station and season were partialled out, hydrochemistry explained 9.1% of residual water-mixture variation (adjusted $R^2 = 4.4\%$; pseudo- $F(7, 122) = 1.738, p = 0.001$). RDA1 and RDA2 accounted for 40.0% and 21.5% of constrained variation, equivalent to 3.6% and 1.9% of total residual composition variance (Fig. 7). Dissolved oxygen and pH contributed significant unique fractions after all other hydrochemical variables were controlled. Temperature and nitrate were marginal, whereas the mineralization index, phosphate and ammonia were not significant. The mineralization index did not explain a significant unique fraction of compositional variation after accounting for station and seasonal effects.

Table 5 Marginal tests of hydrochemical variables in the partial RDA of water OCP composition			
Variable	Pseudo-F	Unique variance (%)	Permutation <i>p</i>
Temperature	1.641	1.22	0.062
DO	3.357	2.50	0.002
Mineralization	1.415	1.05	0.198
pH	2.089	1.56	0.022
Phosphate	0.579	0.43	0.798
Nitrate	2.006	1.50	0.094
Ammonia	0.950	0.71	0.364

Note. Unique variance (%) represents the adjusted proportion of compositional variance uniquely attributable to each hydrochemical variable after accounting for all other predictors in the model

Table 6
Adjusted variation partitioning of water OCP mixture composition

Variance fraction	Percent
Hydrochemistry, unique	3.2
Season, unique	5.3
Station, unique	10.3
Shared explained component	1.0
Unexplained	80.3
Total adjusted explained variance	19.7
Note: Percentages are adjusted variation fractions derived from variation partitioning and may not sum exactly because of rounding	

3.7 Water–sediment concordance

Paired water and sediment concentrations were positively correlated for every OCP signal. Spearman correlations ranged from 0.742 for HCN* to 0.851 for o,p'-DDE, and all remained significant after false-discovery-rate adjustment. The total identified burden had $\rho = 0.856$ ($q < 0.001$; Fig. 8). Despite strong compound-wise burden correlations, complete mixture configurations were only moderately concordant: the Procrustes correlation was 0.346 ($p = 0.001$) and the Mantel distance correlation was 0.392 ($p = 0.001$). Water and sediment exhibited strong agreement in total OCP burden but more moderate agreement in overall mixture composition.

Table 7
Spearman correlations between paired station–month water and sediment concentrations. q values are Benjamini–Hochberg false-discovery-rate-adjusted significance levels

Compound/metric	Spearman ρ	FDR q
HCB	0.769	< 0.001
HCN*	0.742	< 0.001
α -HCH	0.801	< 0.001
β -HCH	0.826	< 0.001
γ -HCH	0.842	< 0.001
Heptachlor	0.823	< 0.001
Heptachlor epoxide	0.812	< 0.001
cis-Chlordane	0.837	< 0.001
trans-Nonachlor	0.842	< 0.001
o,p'-DDE	0.851	< 0.001
p,p'-DDE	0.833	< 0.001
o,p'-DDD	0.838	< 0.001
p,p'-DDD	0.831	< 0.001
o,p'-DDT	0.815	< 0.001
p,p'-DDT	0.830	< 0.001
Mirex	0.827	< 0.001
Σ OCP (identified)	0.856	< 0.001

4. Discussion

4.1 Seasonal forcing and the magnitude of OCP burden

Season was the clearest determinant of total OCP burden. Relative to the short rains, the long-rains period produced an 8.18-fold increase in water and a 13.47-fold increase in sediment. These increases indicate enhanced contaminant mobilization during periods of elevated rainfall and runoff. Legacy compounds stored in agricultural soils, floodplain deposits, and channel margins can enter rivers when storm runoff reconnects previously isolated surfaces, while erosion supplies contaminated fine material to depositional reaches. Noyes et al. (2025) similarly argue that climate and hydrological extremes can remobilize persistent organic pollutants long after primary emissions decline. The Kenyan pattern

therefore supports a reservoir-remobilization mechanism, although it does not by itself distinguish historical stores from any continuing local use.

The stronger fold-change in sediment than in water suggests that sediments integrate contaminant inputs over longer periods than the water column. Water concentrations respond rapidly to inflow, dilution, and downstream export, whereas the upper sediment layer integrates particle-bound inputs and can preserve a pulse after the water column has changed. Comparable water-sediment contrasts have been reported in the Volturno River and in karst wetlands, where hydrophobic OCPs were redistributed between dissolved and particulate phases rather than remaining fixed in one matrix (Montuori et al. 2020; Hu et al. 2024). Dynamic pollutant transport documented for the Rur River also shows that flow events can alter the apparent relationship between water and bed sediments over short intervals (Schwanen et al. 2024). The observed pattern indicates that sediments function both as repositories of contaminant inputs and as potential sources of subsequent remobilization..

The temporal signal is consistent with broader estuarine evidence. Recent Marine Environmental Research studies demonstrate that seasonal freshwater input, salinity and sediment properties reorganize contaminant availability and benthic exposure, even where annual spatial means appear modest (Kwasigroch et al. 2023; Pereira et al. 2023; Sepahvand et al. 2023). In the present study, elevated northeast-monsoon sediment burden may reflect delayed deposition and retention of material delivered earlier, while elevated water burden outside the long rains may reflect continued exchange with sediments and catchment base flow. These findings demonstrate that single-season surveys may not adequately capture the temporal variability of OCP exposure.

The management implication is that annual averages may underestimate episodic exposure. Sampling during the long rains should be prioritized as a priority window for source tracing, while short-rains observations provide a useful baseline condition. Kenyan monitoring has historically documented OCP occurrence but has rarely resolved seasonal pulses across paired matrices (Lalah et al. 2003; Wanjeri et al. 2022; Ogola et al. 2024). Consequently, strengthening temporal coverage may provide greater management value than expanding analyte lists within single sampling campaigns..

4.2 Spatial structure and mixture composition

While total OCP burden was primarily structured by season, mixture composition exhibited both spatial and temporal organization. Water mixtures were structured by season and the estuary-by-season interaction, whereas sediment mixtures showed clearer estuarine differentiation and an interaction effect. This contrast indicates that the water column was dominated by regionally synchronized hydrological forcing, while sediments retained greater local memory of catchment inputs, depositional setting and sediment properties. Similar spatial persistence in estuarine sediments has been reported for organic contaminants in the Hooghly-Sundarban system and in Kenyan coastal estuaries (Zanardi-Lamardo et al. 2019; Wanjeri et al. 2022). These findings suggest that water and sediment provide complementary information on contaminant dynamics, with water reflecting short-term hydrological conditions and sediments integrating longer-term environmental history.

The modest variance explained by the first two principal components is consistent with a system in which no single source axis controlled all compounds. OCP mixtures combine HCH isomers, heptachlor-related compounds, chlordane-related residues, DDT and its metabolites, HCB and mirex, each with different physicochemical properties and transformation histories. Recent coastal assessments likewise report multidimensional contaminant profiles that cannot be reduced reliably to one diagnostic ratio or one dominant compound (Zhang et al. 2023; Wang et al. 2024). The South Coast ordinations therefore support a mixed-source and mixed-process interpretation rather than a single-pesticide narrative.

The significant estuary-by-season interaction indicates that seasonal change was not identical in every estuary; local geomorphology, freshwater discharge and land use modified the regional rainfall signal. This is consistent with tropical mangrove-estuary studies in which environmental gradients produce distinct biological and chemical assemblages among tributaries despite common climatic forcing (Inyang et al. 2023; Bilbao et al. 2023). Monitoring programmes should therefore retain estuary-specific station identities rather than combine all South Coast sites into a single seasonal mean. Failure to account for local estuarine characteristics may obscure important spatial differences in contaminant sources, transport pathways and ecological exposure.

Dispersion effects must also be considered when interpreting PERMANOVA. A significant location effect can arise partly because groups differ in within-group variability. The accompanying dispersion checks reduce, but do not eliminate, this concern. Greater dispersion during high-flow periods may reflect the convergence of contaminants from multiple runoff pathways and source areas, generating more heterogeneous mixture profiles. Ecologically, greater dispersion during high-flow periods would itself be informative because storms create heterogeneous mixtures from multiple runoff pathways. Future models could use multivariate generalized linear mixed models or Bayesian compositional models to estimate compound-specific random slopes and uncertainty directly.

4.3 Hydrochemical controls after repeated spatial and temporal structure

Dissolved oxygen was negatively related to total water burden, and dissolved oxygen and pH contributed significant unique fractions to residual mixture composition. These associations remained after station and season were accounted for, suggesting that they were not merely proxies for the sampling calendar. Lower oxygen may accompany high organic-matter loading, microbial respiration and fine-particle accumulation, all of which can co-occur with contaminant-rich runoff. It may also favour slower oxidative transformation or release from reducing microzones. Nevertheless, dissolved oxygen should be viewed as an indicator of environmental conditions associated with contaminant occurrence rather than a direct causal driver of OCP accumulation.

The result is compatible with recent Marine Environmental Research evidence that contaminant bioavailability depends on sediment redox conditions and associated iron and organic phases (Kwasigroch et al. 2023). It also aligns with studies showing that water and sediment characteristics influence the distribution of benthic organisms and, by implication, their exposure to particle-bound

contaminants (Sepahvand et al. 2023; Woof et al. 2023). These findings suggest that hydrochemical conditions may influence ecological exposure by modifying contaminant partitioning, persistence and bioavailability.

The unique pH fraction may reflect changes in particle-surface charge, dissolved organic matter and degradation, but the effect probably differs among compounds. Khouni et al. (2023) emphasize that salinity-related pesticide behaviour is system- and compound-specific; the same caution applies to pH. The absence of a dominant unique salinity effect after conditioning suggests that apparent salinity correlations may largely track season and estuarine position. These findings highlight the importance of multivariate approaches when evaluating environmental controls on contaminant behaviour.

Hydrochemistry explained 9.1% of residual water-mixture variation, with an adjusted fraction of 4.4%. Environmental contaminant profiles are governed simultaneously by source timing, flow, suspended solids, organic carbon, degradation and measurement error; hydrochemical variables cannot be expected to explain the majority of variation. Comparable multi-stressor estuarine studies increasingly combine chemistry, sediment properties and biological responses because no single environmental gradient is sufficient (Liu et al. 2023; Benthorage et al. 2023; Han et al. 2024). The observed contribution of hydrochemistry is therefore best interpreted as one component of a broader network of interacting environmental controls.

4.4 Water-sediment coupling: strong burden, moderate compositional concordance

The strong Spearman correlation between paired water and sediment totals ($p = 0.856$) indicates that both compartments responded to a common loading regime. Yet Procrustes and Mantel concordance of complete mixtures was only moderate. These results demonstrate that contaminant burden and contaminant composition provide complementary information on environmental contamination. A station-month with high total burden in both matrices may still contain different dominant compounds because partition coefficients, particle affinity and degradation products differ among analytes.

Recent estuarine studies support this distinction. Liu et al. (2023) found that dissolved, particulate and sediment-associated contaminants displayed related but non-identical distributions in Bohai Bay, and Pereira et al. (2023) showed that sediment chemistry and organism uptake could diverge from bulk concentration. Benthorage et al. (2023) similarly demonstrated that resident bivalves integrated pollutant exposure in ways not fully represented by one environmental matrix. The moderate compositional concordance observed in the present study therefore reflects the influence of compound-specific transport, transformation and partitioning processes acting on a shared seasonal loading signal.

The coupling also has a temporal dimension. Sediment can receive the same wet-season pulse as water but release compounds later, while transformation in the bed changes the proportions of parent compounds and metabolites. Consequently, relationships observed between paired water and sediment samples may not fully capture delayed contaminant exchange processes.

4.5 Ecological and monitoring implications

Although this study on abiotic matrices, the findings have direct implications for biological exposure. The pronounced increase in OCP burden during the long-rains season suggests enhanced contaminant transport and greater potential exposure for benthic and filter-feeding organisms. Reviews of persistent pollutants in marine crustaceans show that environmental mixtures can produce bioaccumulation and sublethal cellular responses even when individual water concentrations are low (Camacho-Jiménez et al. 2023). Marine Environmental Research studies using oysters, ascidians and other sentinels likewise demonstrate the value of linking sediment and water chemistry with organism-level endpoints and biological indicators of exposure and effect (Benthota et al. 2023; Beyer et al. 2023).

A defensible monitoring programme should therefore combine three layers: contaminant measurements in water and sediment, hydrochemical and sediment-property measurements that explain transport, and biological indicators that reveal bioavailability. The strong seasonal signal observed in both matrices suggests that monitoring intensity should be aligned with hydrological conditions rather than distributed uniformly throughout the year. In particular, enhanced sampling during the long-rains season would improve detection of contaminant pulses, while dry-season observations would help determine whether sediments function as continuing reservoirs and secondary sources of contamination.

The results also demonstrate the importance of considering contaminant composition alongside Σ OCP. Total burden is useful for identifying exposure pulses, mixture composition provides additional information on persistence, transformation pathways and potential ecological risk. Management decisions based solely on aggregate concentrations may therefore overlook shifts in the relative importance of individual compounds.

The unidentified HCN* peak illustrates the importance of maintaining a clear distinction between observed analytical signals and confirmed chemical identifications. Retaining the signal in exploratory analyses while excluding it from risk interpretation ensures that analytical uncertainty is acknowledged explicitly and does not influence conclusions regarding contaminant sources or ecological effects..

4.6 Study limitations and priorities for further research

The study's strengths include the paired monthly design, the averaging of analytical triplicates before inference and the use of compositional and mixed-effects methods. Several limitations nevertheless constrain causal interpretation. Suspended solids, sediment grain size, total organic carbon and river discharge were not available as linked station-month covariates, limiting direct evaluation of the mechanisms controlling contaminant transport and retention. In addition, water samples were analyzed as bulk samples, preventing differentiation between dissolved and particle-associated contaminant fractions, while the absence of discharge data precluded estimation of contaminant loads. Laboratory records should also be retained with complete recoveries, detection limits, blank corrections and certified-reference performance to support data verification and reproducibility.

The study covered one annual cycle. A second year with contrasting rainfall would show whether the observed 8.18- and 13.47-fold long-rains effects are stable or were specific to the study year. Climate-related changes in storm intensity make this replication particularly important (Noyes et al. 2025). Future investigations incorporating event-based storm sampling, hydrograph analysis, catchment soil measurements and passive sampling approaches would improve understanding of the relative contributions of surface runoff, sediment remobilization and base-flow transport pathways.

A further limitation was the inability to distinguish dissolved-phase transport from particle-associated transport. During high-flow conditions, elevated concentrations may reflect increased contaminant delivery, greater suspended sediment loads, or a combination of both processes. Because suspended solids and particulate organic matter were not quantified, the relative importance of these pathways could not be determined directly. Future studies should separate filtered and unfiltered water fractions, quantify suspended solids and organic carbon, and estimate flow-weighted loads to better resolve contaminant transport mechanisms and source pathways. Studies should also evaluate temporal lags between contaminant inputs, sediment storage and subsequent remobilization using approaches such as passive sampling, time-series analysis and distributed-lag modelling. Measurements of sediment organic carbon and grain-size composition would further improve understanding of contaminant partitioning and retention processes.

Finally, chemical occurrence should be linked more directly to biological effect. Sediment toxicity tests, benthic-community metrics and tissue residues can determine whether seasonal chemical pulses translate into adverse outcomes. The controlled-exposure work of Woof et al. (2023) illustrates how avoidance and oxygen-consumption responses can reveal ecological consequences not evident from chemistry alone, while Han et al. (2024) demonstrates the importance of coupling accumulation with risk screening. Such weight-of-evidence approaches represent an important next step for understanding the ecological significance of legacy OCP contamination in Kenya's South Coast estuaries.

4.7 Synthesis

The collective evidence supports a conceptual model in which regional seasonal forcing controls the timing and magnitude of OCP inputs to estuarine systems, local estuarine properties modify the composition and retention of contaminants within sediments, and hydrochemical conditions contribute a smaller but significant influence on water composition. Water and sediment are tightly coupled in overall burden but only moderately concordant in compound proportions. These findings demonstrate the complementary value of the two matrices, with water reflecting short-term transport and exposure dynamics and sediment integrating longer-term retention, transformation and remobilization processes.. The study therefore advances beyond a simple occurrence survey a basis for seasonally targeted, multi-compartment monitoring of legacy pesticides along the tropical Western Indian Ocean coast by identifying the processes that structure contaminant distributions.

5. Conclusions

Paired monthly observations across five tropical river–estuary systems showed that OCP burden and mixture composition were controlled by different but interacting processes. Total burden was dominated by season, with pronounced increases during the northeast monsoon and long rains. Water composition changed through season-dependent estuarine responses, while sediment retained stronger spatial differentiation. Hydrochemistry explained a significant but modest independent fraction of compositional variation after spatial and seasonal effects were accounted for, with dissolved oxygen and pH contributing more than the mineralization gradient. Water and sediment burdens were strongly correlated, although complete mixture configurations were only moderately concordant. Together, these findings demonstrate that total contaminant burden and mixture composition provide complementary information and should be interpreted jointly when assessing contaminant dynamics in tropical estuaries. They further support integrated, seasonally targeted monitoring programmes that incorporate both water and sediment compartments. HCN* should remain excluded from compound-specific interpretation until its analytical identity is confirmed.

Declarations

Ethics approval:

Not applicable; the study involved water and sediment sampling only.

Consent to participate:

Not applicable.

Funding:

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Competing interests: The authors declare no competing interests.

Data availability: The datasets analysed in this study are available from the corresponding author on reasonable request.

Author contributions:

Kobingi Nyakeya: conceptualization, investigation, laboratory coordination, data curation, formal analysis and original draft. Getabu Albert, Eric Okuku, Jane Moraa Nyamora, Zipporah Moraa Gichana, James Onchieku, Masese Frank Onderi, Chrispine Nyamweya and Paul Orina: supervision, methodology, interpretation, critical review and editing. All authors approved the manuscript.

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Figures

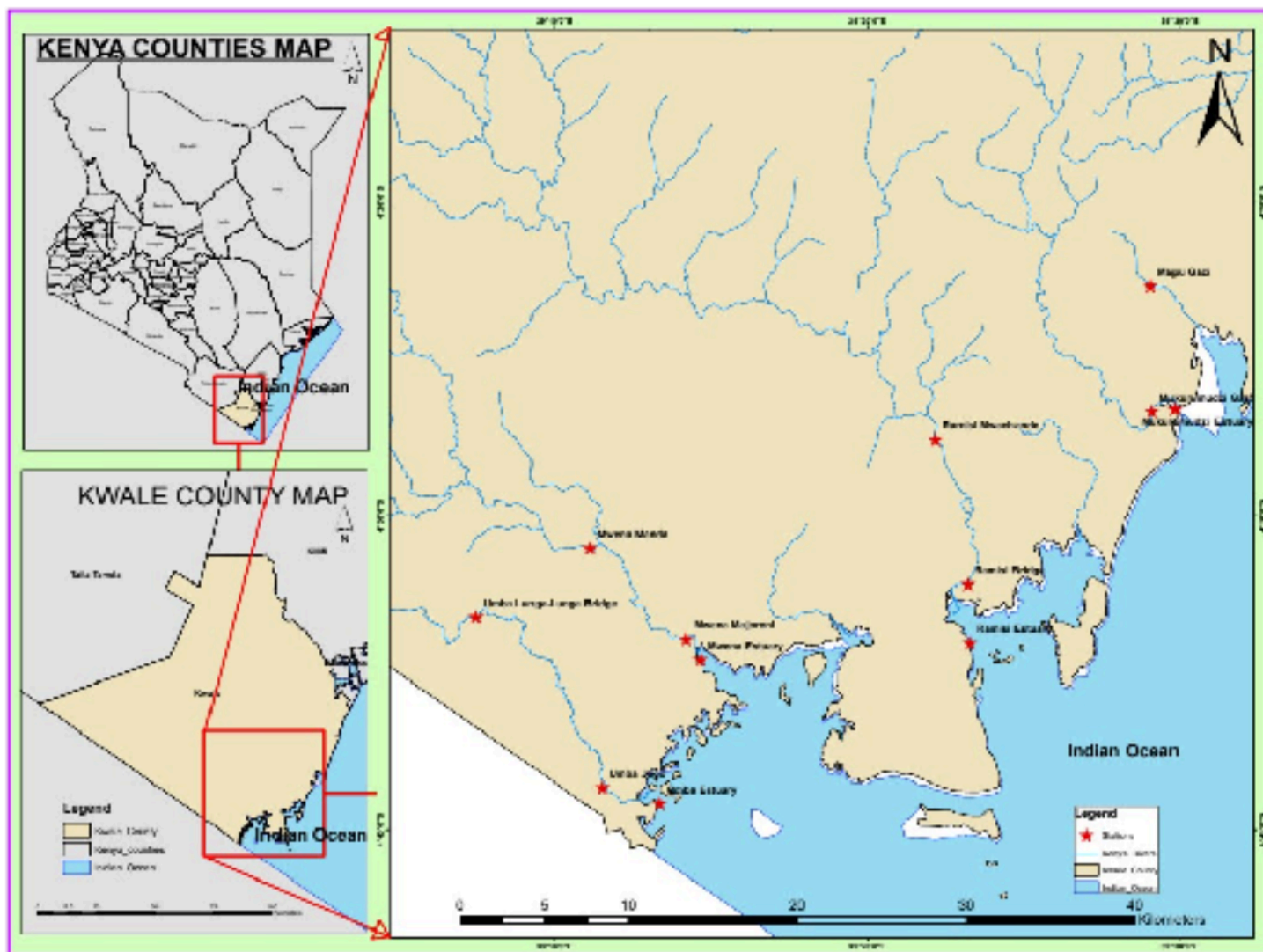


Figure 1

Study area and sampling stations in the Mapu, Mwena, Mkurumdzi, Ramisi and Uмба river–estuary systems on the South Coast of Kenya.

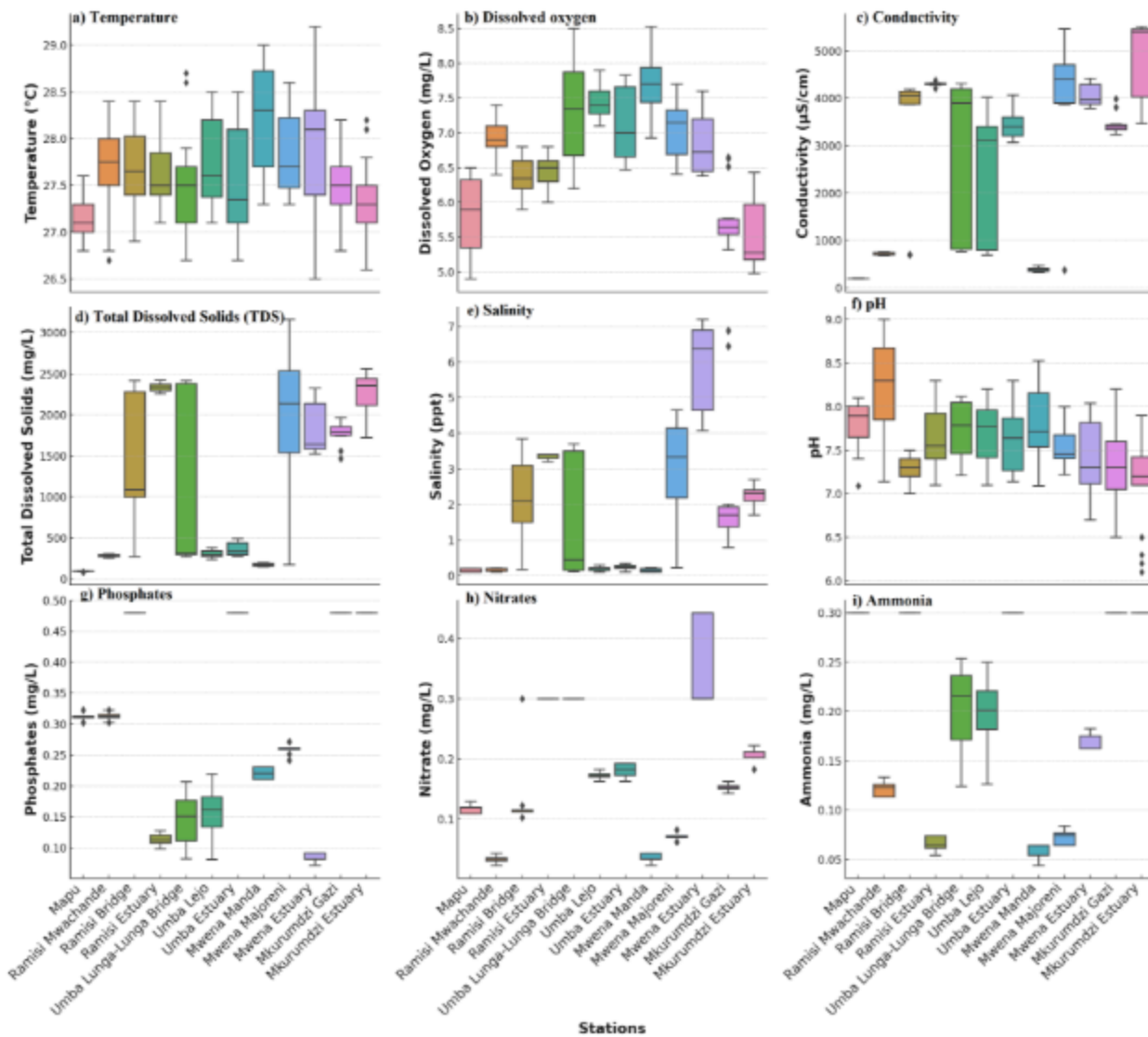


Figure 2

Spatial distributions of temperature, dissolved oxygen, conductivity, total dissolved solids, salinity, pH, phosphate, nitrate and ammonia across the twelve sampling stations. Boxes show medians and interquartile ranges, whiskers represent $1.5 \times \text{IQR}$, and points indicate outliers.

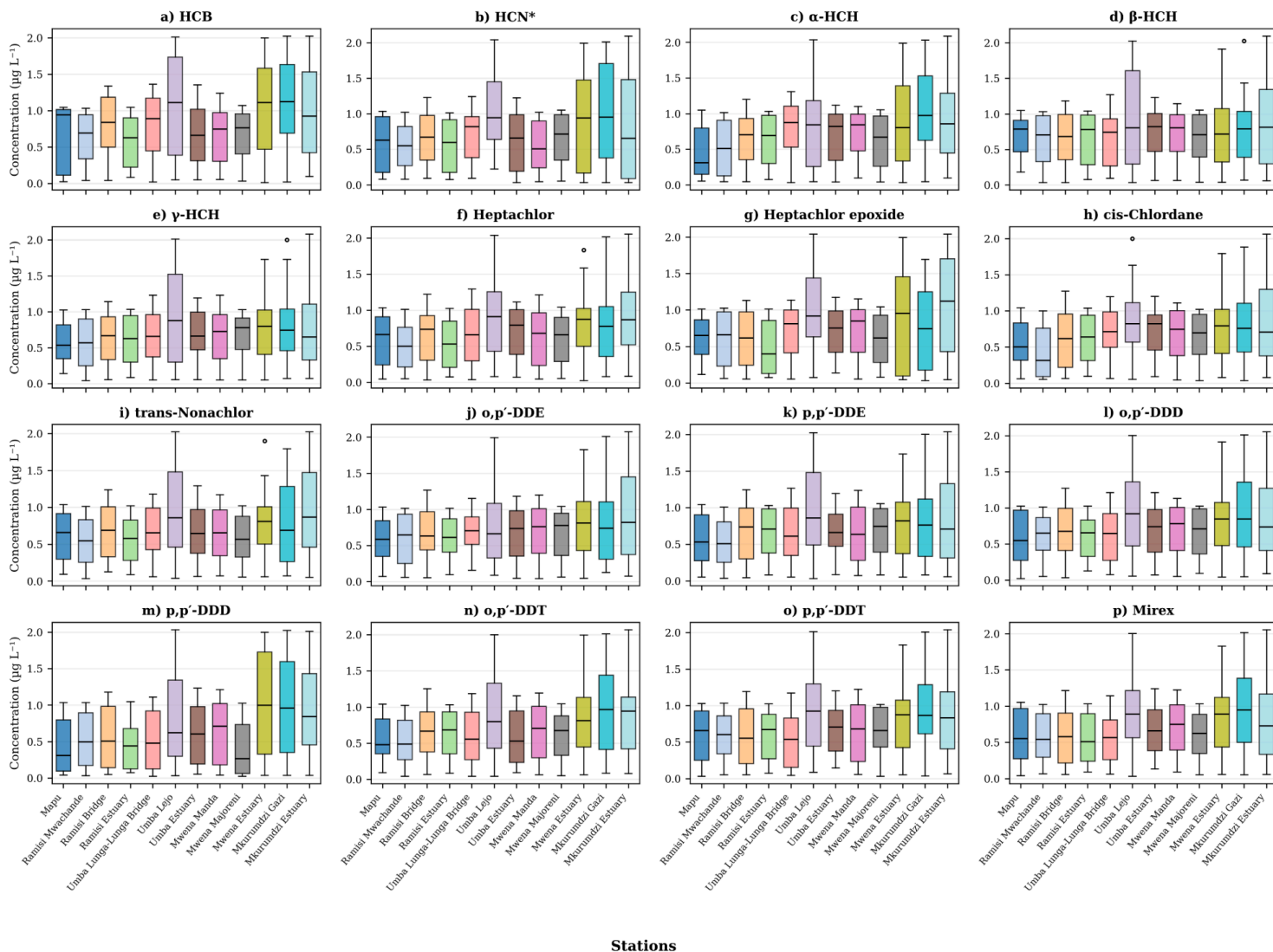


Figure 4

Spatial distributions of the fifteen identified organochlorine pesticides (OCPs) in surface water across the twelve monitoring stations. Boxplots show median values, interquartile ranges, whiskers and outliers for station–month observations collected between September 2018 and August 2019

Water OCP mixture composition (CLR-PCA)

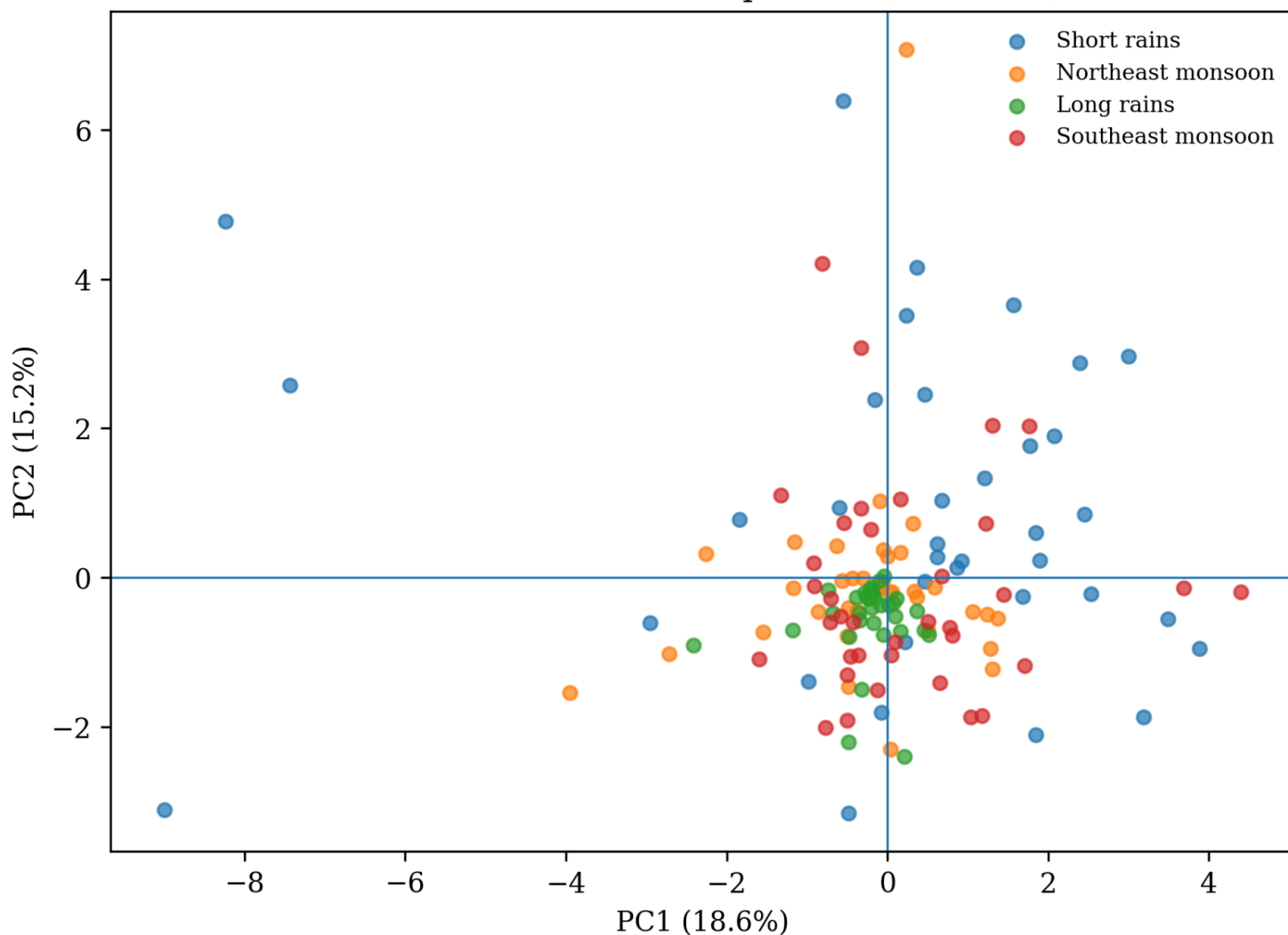
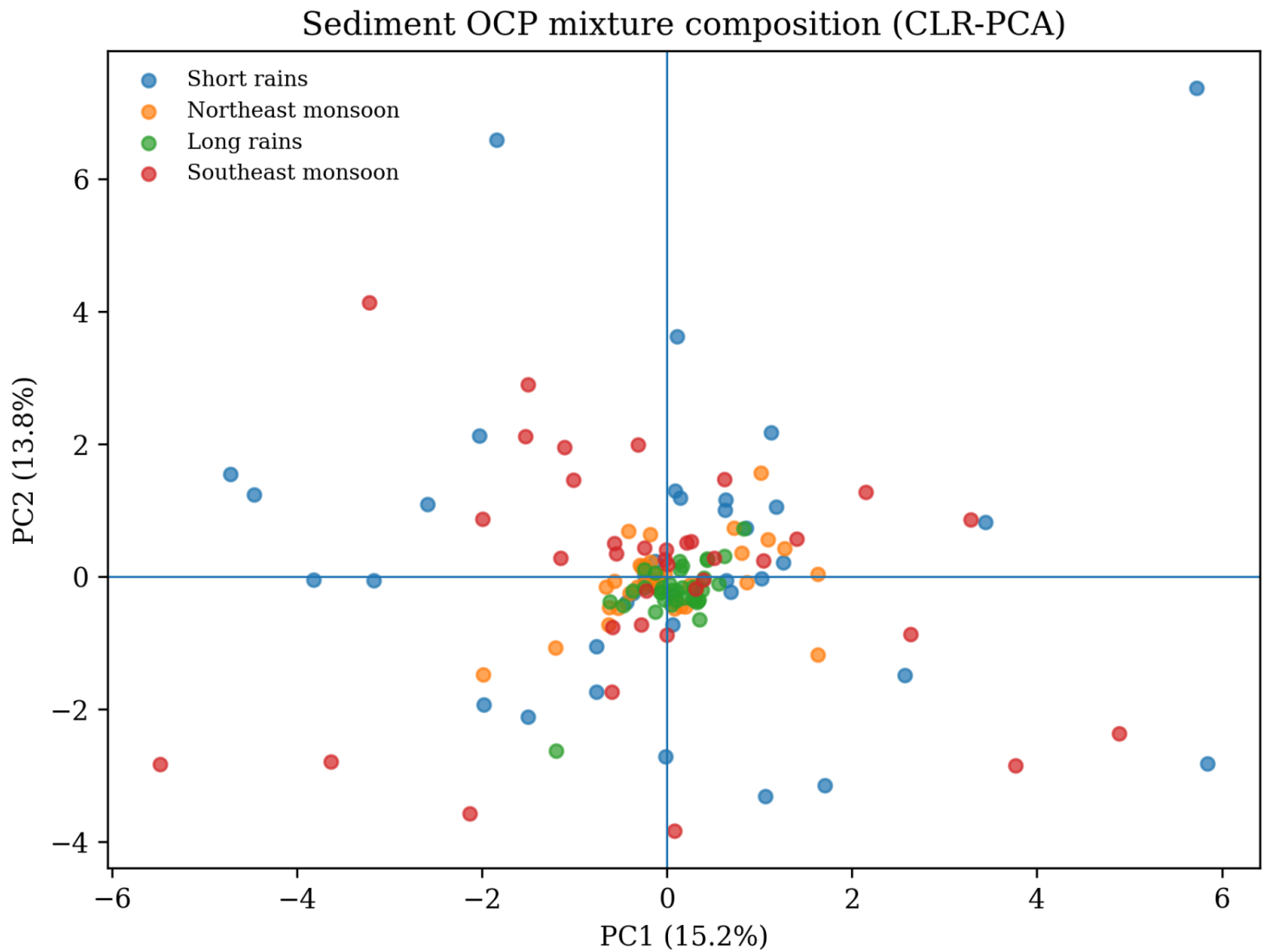
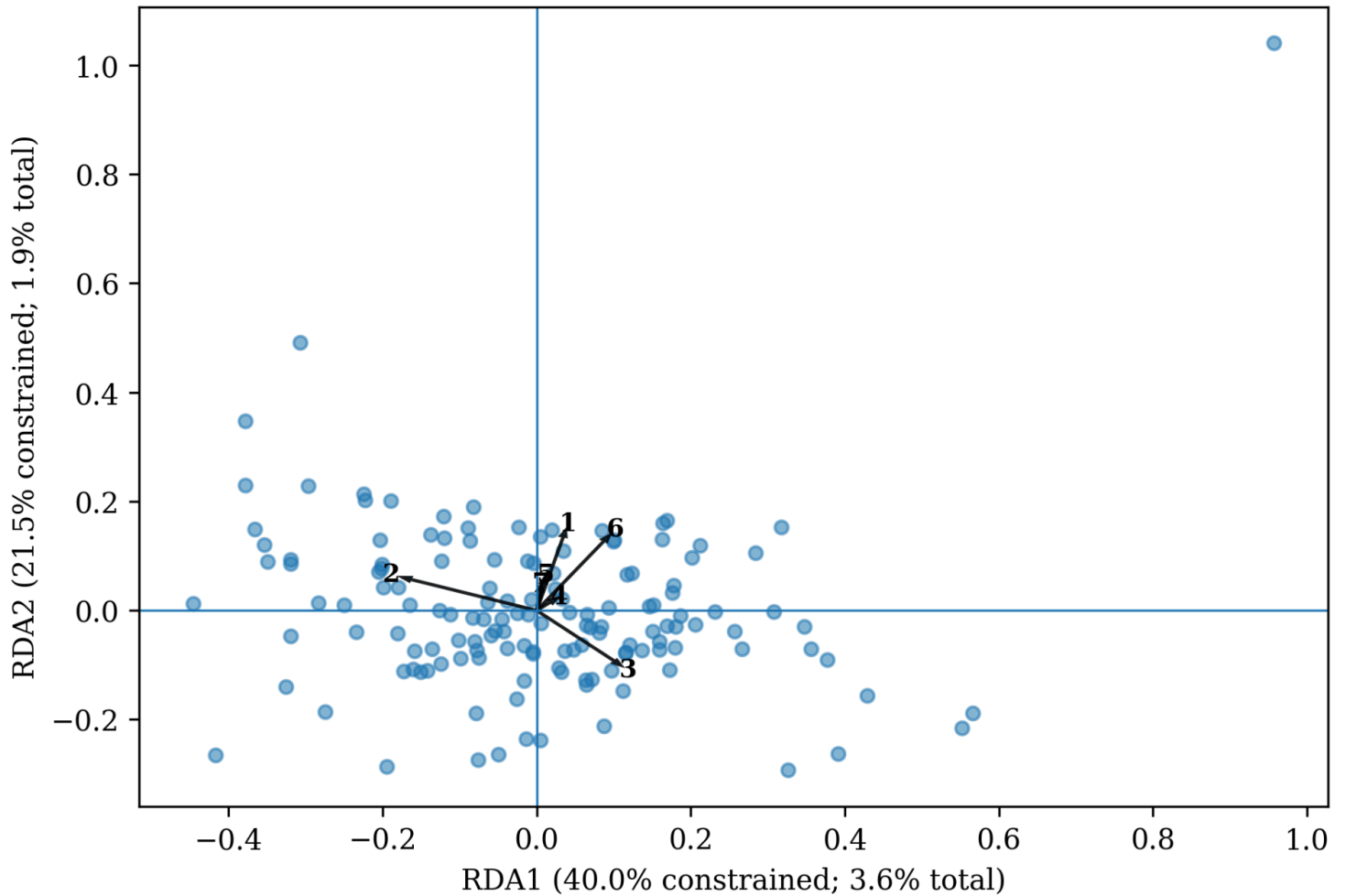


Figure 5

Principal component analysis (PCA) of centered-log-ratio-transformed water OCP composition. PC1 and PC2 explain 18.6% and 15.2% of compositional variance, respectively



Partial RDA of water OCP composition



1 Temperature; 2 DO; 3 Mineralization; 4 pH; 5 Phosphate; 6 Nitrate; 7 Ammonia

Figure 7

Partial redundancy analysis (RDA) of centered-log-ratio-transformed water OCP composition after conditioning on station and season. Environmental vectors represent temperature (1), dissolved oxygen (2), mineralization index (3), pH (4), phosphate (5), nitrate (6) and ammonia (7)

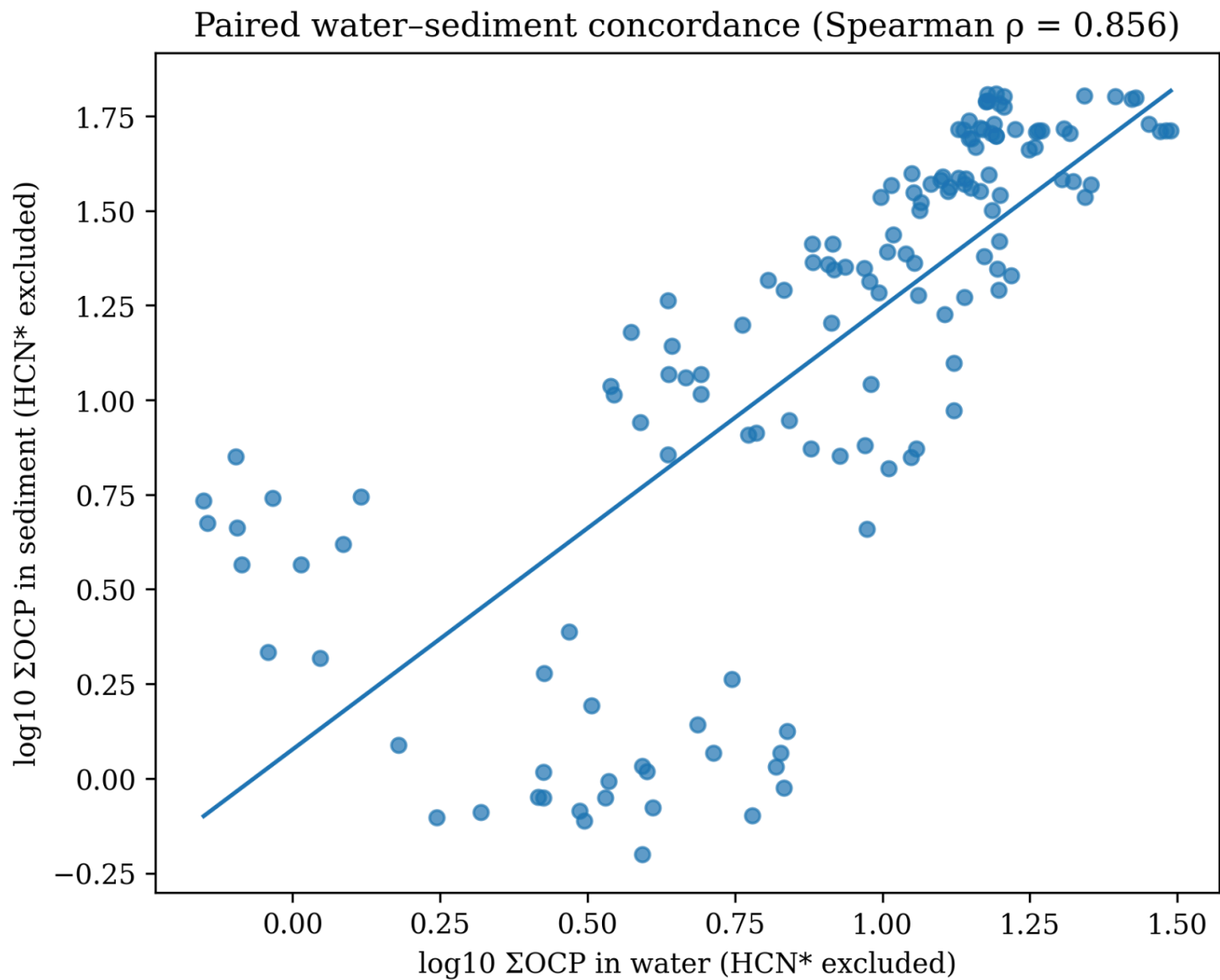


Figure 8

Relationship between total identified OCP burden in paired water and sediment samples ($n = 144$ station-month observations). HCN was excluded from Σ OCP calculations in both matrices