



Spatial–temporal distributions, probable health risks, and source identification of organic pollutants in surface waters of an extremely hypoxic river basin in Türkiye

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Abstract This study was carried out to determine the spatiotemporal distributions of organic pollution parameters in the Meriç–Ergene River Basin subjected to intensive agricultural and industrial pressure. A total of 5 basin components, including Anadere (A), Çorlu (Ç), Tunca (T), Meriç (M), and Ergene (E) rivers, and 9 stations (A1, Ç1, T1, M1–M2, and E1–E4) were identified in the watershed, and surface water samples were collected in the dry (end of summer) and wet (end of winter) seasons of 2021–2022. The Water Quality Index (WQI) and Nutrient Pollution Index (NPI) were applied to the data to evaluate the overall water quality characteristics. The Chronic Daily Index (CDI), Hazard Quotient (HQ), and Hazard Index (HI) were applied to the data to reveal the probable noncarcinogenic health risks of organic contaminants. Cluster Analysis (CA) and Principal Component Analysis (PCA) were applied to the data to classify the sampling sites and identify the source apportionment of organic pollution parameters. The recorded spatiotemporal averages of the investigated parameters in the basin are as follows: 6.26 mg/L for DO, 9 for pH, 1626 μ S/cm for EC, 985 mg/L for

TDS, 1 ‰ for salinity, 6.88 mg/L for nitrate, 0.1 mg/L for nitrite, 1.8 mg/L for phosphate, 81 mg/L for sulfate, and 473 mg/L for chloride. The results indicate that the most contaminated components of the basin are Çorlu Stream and Ergene River, and in addition to their quite high salt and nutrient content, they have extreme hypoxic conditions to the extent that it is impossible for many aquatic organisms to live. The contamination degrees of the investigated basin components were determined using the organic pollution risk assessment indices as follows: Çorlu Stream > Ergene River > Anadere Stream > Tunca River > Meriç River.

Keywords Meriç–Ergene River Basin · Organic pollution · Health risk assessment

Introduction

Water contamination by nutrients because of agrogenic domestic discharges and salinization of freshwater habitats because of industrial discharges are among the most significant environmental problems on the globe. Freshwater pollution may occur for natural and anthropogenic reasons (Kumar et al., 2021). But anthropogenic processes and inputs, including domestic and agricultural runoff and industrial wastewater, are among the main factors determining the quality of aquatic habitats and a major source of organic pollutants (İslam et al., 2022; Köse et al., 2020; Men et al., 2018; Ustaoglu et al., 2022; Yüksel et al., 2022).

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Ecological risk assessment indices are useful tools in determining the multiple effects of organic and inorganic contaminants in water ecosystems, and they may provide the determined risk assessment information in a form that regulatory agencies may use to make necessary decisions. Health risk assessment indices are also useful tools for estimating noncarcinogenic hazards, which may occur when people are exposed to certain toxicants (Ahamad et al., 2020; İslam et al., 2021; Men et al., 2020; Nong et al., 2020; Ustaoğlu et al., 2020).

Water quality assessment issues generally require the analysis of many parameters simultaneously. Principle Component Analysis (PCA) reduces the dimensionality of a multivariate data set, while Cluster Analysis (CA) classifies the objects characterized by the values of variables into groups. Therefore, PCA and CA have been frequently used all over the world while dealing with water pollution risk assessment data (Men et al., 2022; Mutlu, et al., 2021; Nasiruddin et al., 2022; Saleem et al., 2019; Ustaoğlu et al., 2020; Varol, 2020).

The Thrace Region is a significant part of the Marmara Region, and it is located in the northwest part of Türkiye. It covers the provinces of Tekirdağ, Edirne, and Kırklareli, and there are 83 municipalities, 26 districts, and 678 villages in the region with a population of about 2 million. The region has significant industrial potential, which is centered on the D-100 Highway (which runs from Ordu and Ercezköy to Muratlı and Lüleburgaz). The total number of industrial facilities in the region is around 2000, with approximately 80% of these industrial facilities located in the Tekirdağ province (in the Ergene River's Çorlu Stream watershed subbasin). The region also has very significant agricultural production potential due to its rich soils and many freshwater resources. For the Thrace Region, paddy and sunflower are the main crops, and approximately 25% of paddy and 80% of sunflower production in Türkiye are supplied from this region (<http://www.cerkezkoy.gov.tr>; <http://www.corlu.gov.tr>; Anonymous, 2010; Çingiroğlu, 2018; Tokatlı & Ustaoğlu, 2020).

The Ergene River, which is the main potamic habitat in the Thrace Region, originates from the springs in Güneşkaya locality around 312 m (Taşpnar Hill) of the Yıldız (Istranca) Mountains in the north of Tekirdağ-Saray District with the name of Ergene Creek and flows in a northeast-southwest direction. Then it joins

the Çorlu Stream near Muratlı-İnanlı Villages and turns to the northwest under the name of the Ergene River. The river joins with the Meriç River near Meriç-Adasarhanlı Villages, 40 km southwest of Uzunköprü District. The length of the Ergene Stream is 91 km from the source in the Yıldız Mountains to the junction with the Çorlu Stream and 194 km to the point where it joins the Meriç River after taking the name Ergene River (a total of 285 km). The river's watershed, with its number of tributaries, covers about 70% of the total area of the Thrace Region (Anonymous, 2010; Emadian et al., 2021; Hasan et al., 2021; Orak et al., 2020; Ordu, 2005; Tokatlı, 2021).

To date, a number of studies have been carried out in many different areas of the Meriç–Ergene River Basin, and the pollution levels of the basin components have been evaluated from different perspectives. In a study carried out by Kaçar (2011), fecal coliform and fecal streptococcal contents were examined in water samples taken seasonally from the Meriç River. In another study carried out by Tokatlı (2015), the water quality of the Meriç River was investigated by determining a total of 19 physical, chemical, and biological pollution parameters. In two studies performed in 2016 and 2019, heavy metal accumulations in the sediments of the Meriç–Ergene River Basin were investigated, and the results were evaluated by ecotoxicological risk assessment indices (Tokatlı & Başatlı, 2016; Tokatlı, 2019a). In 2019, heavy metal accumulations in the groundwater, surface water, and sediments of the Meriç–Ergene River Basin were investigated, and the results were evaluated by health risk assessment indices (Tokatlı, 2019b, c). In three different investigations performed in 2020, pesticide residues in water and sediment samples from fluvial and lacustrine freshwater habitats of the Meriç–Ergene River Basin were investigated (Tokatlı et al., 2020a; Tokatlı, 2020a, b; Köse et al., 2018). In two similar studies carried out by Tokatlı et al., (2020b, c), organic pollution levels and trophic statuses of the Meriç, Tunca, and Ergene Rivers were evaluated using diatom indices. In two separate studies conducted in 2020 and 2021, ecotoxicological and health risk assessments of toxic metals were evaluated in surface and groundwaters of the Meriç–Ergene Basin and Meriç River Delta (Tokatlı & Ustaoğlu, 2020; Tokatlı, 2021). In another study performed in the Ergene River, the micropollutant contents of water samples were investigated (Emadian

et al., 2021). In two similar investigations performed in 2021 and 2022, the effects of the restrictions applied due to the COVID-19 pandemic on the quality of water and sediments in the Meriç–Ergene River Basin in terms of heavy metal accumulations were evaluated (Tokatlı, 2022; Tokatlı & Varol, 2021a). Celen et al. (2022) investigated the water quality of orlu Stream and the Ergene River's source region in a study conducted upstream of the Ergene River. The water qualities of stagnant water bodies in the Meriç–Ergene River Basin (Tokatlı & Varol, 2021b) and orlu Stream (Varol & Tokatlı, 2023) were evaluated in terms of toxic metals using ecotoxicological and health risk assessment indices in two separate current investigations conducted in 2021 and 2023.

Although many researchers have previously carried out investigations on a wide variety of topics in the basin, such as surface and groundwater qualities, ecotoxicological risk assessment of sediments, health risk assessment of heavy metals, use of biotic indices to evaluate pollution statuses, and pesticide accumulations in the basin components, no earlier study has been performed in the field of health risk assessment of organic pollutants in the surface waters of the Meriç–Ergene River Basin by covering the entire watershed. Therefore, this significant watershed has been chosen as a pilot survey on this significant topic, and in this study, water quality and organic contamination levels of Meriç–Ergene River Basin components were evaluated using multivariate statistical methods (PCA and CA), ecological risk assessment indices (WQI and NPI), and noncarcinogenic human health risk assessment techniques (CDI, HQ, and HI).

Materials and methods

Sample collection

In the Meriç–Ergene River Basin, 9 stations were chosen based on pollution resources (Anadere Stream—A1, orlu Stream—1, Tunca River—T1, Meriç River—M1-M2, and Ergene River—E1-E4). The topographic map of the Meriç–Ergene River Basin and sampling locations are given in Fig. 1.

Surface water samples were taken from 0.5 m below the water surface in 1 L precleaned glass bottles with a telescopic water sampling device approximately

3 m from the shore into the precleaned polyethylene bottles at the end of summer–dry season and at the end of winter–wet season of 2021–2022 (APHA, 2005). After measuring some physicochemical parameters in the field, surface water samples were placed in a cooler at 4 °C and transported as immediately as possible to the laboratory for further analysis.

Physicochemical analysis and quality check

Dissolved oxygen (DO), electrical conductivity (EC), total dissolved solids (TDS), salinity (Sal), and pH were determined with a portable multiparameter device (Hach Lange, HQ40D) during the field studies (APHA, 1999). In the laboratory, the water samples were filtered on the day of sampling. In the filtered water samples, nitrate (NO₃) and nitrite (NO₂) levels were measured with a colorimeter device (Hach Lange, DR890), while sulfate (SO₄), phosphate (PO₄), and chloride (Cl) levels were measured with a spectrophotometer device (Hach Lange, DR3900) (APHA, 1999). All the parameter values were recorded as the average of three measurements. Detailed information on the analysis methods of these parameters, including their units, methods, and instruments, is summarized in Table 1.

Ecological risk assessment indices

The Water Quality Index (WQI) is an effective mathematical instrument that provides valuable data to predict the status of surface water quality by combining multiple organic–inorganic water quality variables and by converting them into a single number (Pesce & Wunderlin, 2000; Tokatlı, 2021; Varol, 2020; Zotou et al., 2020).

Nitrate and phosphate are two of the most significant elements that play a vital role in the quality of freshwater. However, nitrate–phosphate contamination in freshwater is a serious problem today all over the world (Arauzo, 2017; Chen et al., 2020; Qian et al., 2020). The Nutrient Pollution Index is a widely used risk assessment tool to evaluate the level of nitrate–phosphate pollution in water ecosystems (Isiuku & Enyoh, 2020; Varol & Tokatlı, 2023).

All the calculation methods of applied ecological risk assessment indices, including Water Quality Index (WQI) and Nutrient Pollution Index (NPI) with their formulas, explanations, evaluation scales, and references, are given in Table 2.

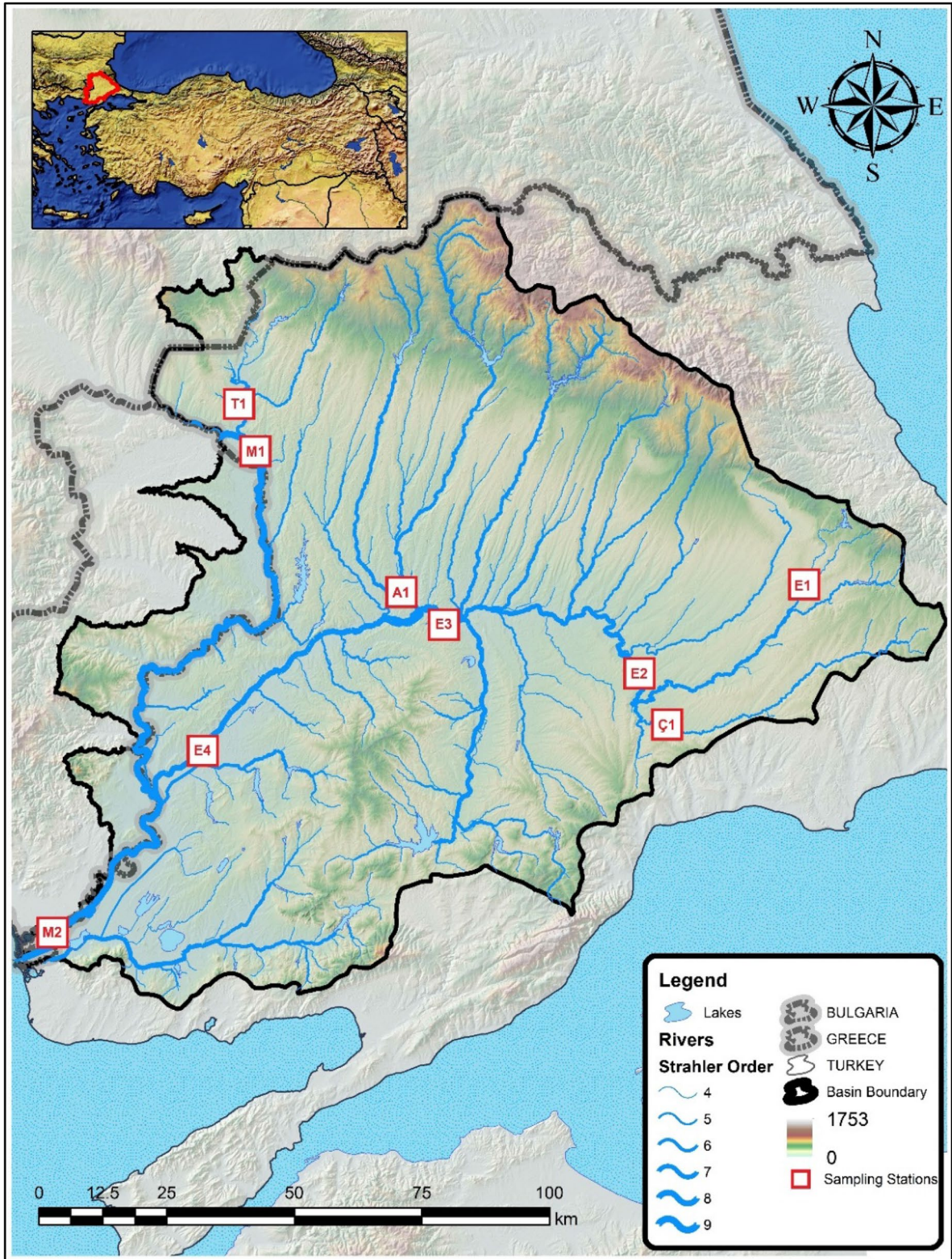


Fig. 1 Meriç-Ergene River Basin and selected stations

Table 1 Water quality parameters, abbreviations, units, analytical methods, and instruments

Parameter	Abbreviation	Unit	Analytical method	Instrument
Dissolved oxygen	DO	mg/L	Portable multiparameter	Hach HQ40D
pH	pH	–	Portable multiparameter	Hach HQ40D
Electrical conductivity	EC	μS/cm	Portable multiparameter	Hach HQ40D
Total dissolved solids	TDS	mg/L	Portable multiparameter	Hach HQ40D
Salinity	Sal	‰	Portable multiparameter	Hach HQ40D
Nitrate	NO ₃	mg/L	Colorimetric	Hach DR890
Nitrite	NO ₂	mg/L	Colorimetric	Hach DR890
Phosphate	PO ₄	mg/L	Spectrophotometric	Hach DR3900
Sulfate	SO ₄	mg/L	Spectrophotometric	Hach DR3900
Chloride	Cl	mg/L	Spectrophotometric	Hach DR3900

Health risk assessment

The calculation methods of applied health risk assessment indices including Chronic Daily Intake (CDI), Hazard Quotient (HQ), and Hazard Index with their formula explanations and references are given in Table 3.

Statistical analysis

Pearson Correlation Index (PCI) and Principal Component Analysis (PCA) were performed to reveal relationships among the physicochemical variables by using SPSS 22. Cluster Analysis (CA) was performed to classify the investigated basin components in terms of their similar water quality characteristics by using PAST 4.07.

Results

Water quality assessment

The results of measuring 10 water quality parameters in Anadere (A1), Çorlu (Ç1), Tunca (T1), Meriç (M1–M2), and Ergene (E1–E4) rivers in dry and wet seasons are given in Fig. 2.

The detected average values of the investigated limnologic variables in surface water of the Meriç–Ergene River Basin are as follows: 5.19 mg/L for DO (0.17 mg/L-E2 to 8.90 mg/L-M1), 8.66 for pH (7.95-E3 to 9.45-E1), 2202 μS/cm for EC (288 μS/cm-E1 to 6510 μS/cm-Ç1), 1237 mg/L for TDS (151 mg/L-E1 to 3790 mg/L-Ç1), 1.26 ‰ for salinity (0.15 ‰-E1 to 3.90 ‰-Ç1), 6.15 mg/L for nitrate (1.10 mg/L-E4 to 18.2 mg/L-E2),

Table 2 Methods of applied ecological risk assessment indices

Index	Calculation	Formula explanations	Rating of water quality	The standards used	Reference
WQI	$WQI = \sum_{i=1}^n Wix(\frac{Ci}{Si}) \times 100$ $Wi = \frac{wi}{\sum_{i=1}^n wi}$	<p>WQI: the relative weight</p> <p>W_i: the values assigned to i (1–5)</p> <p>C_i: the values of measured parameters</p> <p>S_i: the standard values</p>	<p>< 50: excellent water quality</p> <p>50–100: good water quality</p> <p>100–200: poor water quality</p> <p>200–300: very poor water quality</p> <p>> 300: unsuitable</p>	TSWQR, 2021	(Tokatlı, 2022)
NPI	$NPI = (C_N/MAC_N) + (C_P/MAC_P)$	<p>C_N: the levels of NO₃-N in water sample</p> <p>C_P: the levels of PO₄-P in water sample</p> <p>MAC_N: the maximum limit of NO₃-N</p> <p>MAC_P: the maximum limit of PO₄-P</p>	<p>< 1: not polluted</p> <p>1–3: moderately polluted</p> <p>3–6: considerably polluted</p> <p>> 6: very highly polluted</p>	TSWQR, 2021	(Tokatlı, 2022)

Table 3 Methods of applied human health risk assessment indices

Index	Calculation	Formula explanations	Rating of health risk	Reference
CDI	$CDI = C_{water} \times \frac{(IR \times EF \times ED)}{(BW \times AT)}$	C_{water} : level of the NO ₃ and NO ₂ in water (ppb) IR: ingestion rate (L/day) (adult (ad): 2, child (ch): 0.64) EF: exposure frequency (365 day/year) ED: exposure duration (years) (ad: 70, ch: 6) BW: average body weight (kg) (ad: 70, ch: 20) AT: averaging time (days) (ad: 25,550, ch: 2,190)	–	(EPA, 2004)
HQ	$HQ = \frac{CDI}{RfD}$	CDI: chronic daily intake RfD: reference doses of NO ₃ (1.6 ppm) and NO ₂ (0.1 ppm)	–	(EPA, 2004)
HI	$HI = HQ_{NO3} + HQ_{NO2}$	HQ _{NO₃} : Hazard Quotient for NO ₃ HQ _{NO₂} : Hazard Quotient for NO ₂	HI < 0.1: negligible 0.1 < HI < 1: low risk 1 < HI < 4: medium risk 4 < HI: high risk	(EPA, 2004)

0.077 mg/L for nitrite (0.025 mg/l-M1 to 0.127 mg/L-Ç1), 2.36 mg/L for phosphate (0.42 mg/L-M1 to 7.21 mg/L-Ç1), 103 mg/L for sulfate (15 mg/L-E1 to 184 mg/L-Ç1), and 522 mg/L for chloride (14 mg/L-E1 to 1703 mg/L-E2) in the dry season; 7.33 mg/L for DO (0.27 mg/L-Ç1 to 11.13 mg/L-M2), 9.37 for pH (8.68-Ç1 to 9.98-E1), 1050 µS/cm for EC (203 µS/cm-E1 to 3930 µS/cm-Ç1), 723 mg/L for TDS (124 mg/L-E1 to 2810 mg/L-Ç1), 2.89 ‰ for salinity (0.12 ‰-E1 to 0.74 ‰-Ç1), 7.60 mg/L for nitrate (0.50 mg/l-A1 to 15.8 mg/L-Ç1), 0.133 mg/L for nitrite (0.021 mg/l-E1 to 0.227 mg/L-A1), 1.32 mg/L for phosphate (0.25 mg/L-E1 to 2.84 mg/L-E2), 59 mg/L for sulfate (32 mg/L-E1 to 108 mg/L-M2), and 422 mg/L for chloride (10 mg/L-M1 to 1266 mg/L-Ç1) in the wet season.

The water quality characteristics of basin components showed significant changes between the investigated seasons. The recorded percentage changes of average values in the investigated limnologic parameters from dry to wet seasons were as follows: + 101% for DO, + 8.50% for pH, 51.68% for EC, – 39.94% for TDS, 40.39% for salinity, + 126% for NO₃, + 82.19% for NO₂, – 37.66% for PO₄, – 23.79% for SO₄, and – 25.74% for Cl.

Although dissolved oxygen (DO) levels increase and salination (EC, TDS, and salinity) levels decrease in water from all components of the watershed during the rainy season, extremely hypoxic and salinized conditions were recorded in water from Corlu Stream (1) and the Ergene River (E2) during both the dry and wet seasons.

Anadere Stream (A1), Tunca River (T1), Meriç River (M1–M2), and upstream of Ergene River (E1) have the 1st class (unpolluted) (> 8 mg/L) water quality, and Çorlu Stream (Ç1) and mid-down-stream of Ergene River (E2–E4) have the 3rd class (polluted) (< 6 mg/L) water quality in terms of DO in the dry season, while the stations of A1, T1, M1, M2, and E1 have the 1st class water quality, E3 and E4 have the 2nd class (slightly polluted) (6–8 mg/L) water quality, and Ç1 and E2 have the 3rd class water quality in terms of DO in the wet season (TSWQR, 2021).

Upstream of Meriç (M1) and Ergene (E1) Rivers has the 1st class (< 400 µS/cm) water quality, Anadere Stream (A1) and Tunca River (T1) have the 2nd class (400–1000 µS/cm) water quality and Çorlu Stream (Ç1), and downstream of Meriç River (M2) and mid-down-stream of Ergene River (E2–E4) have the 3rd class (> 1000 µS/cm) water quality in terms of EC in the dry season, while the stations of A1, T1, M1, and E1 have the 1st class water quality, M2, E3, and E4 have the 2nd class water quality, and Ç1 and E2 have the 3rd class water quality in terms of EC in the wet season (TSWQR, 2021).

The nitrogen compounds (NO₃ and NO₂) in the water of almost all the watersheds increased significantly in the wet season, while the phosphorus compounds (PO₄) decreased significantly. In the dry season, the Ergene River (E1, E3-E4) has the 1st class (3 mg/L) water quality, while the Anadere (A1), orlu (1), Tunca (T1), and Meriç (M1-M2) rivers have the 2nd class (3–10 mg/L) water quality, the stations of

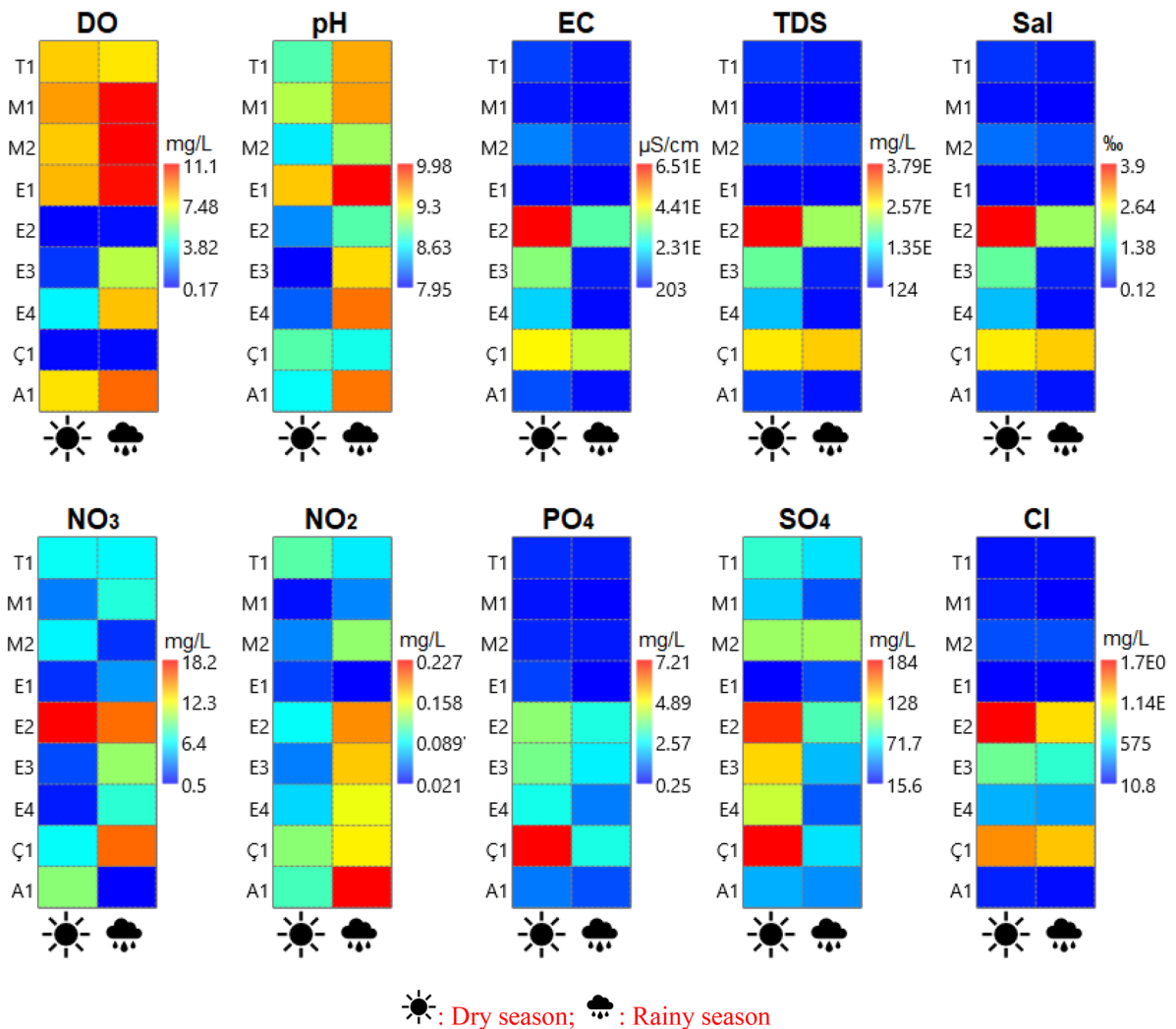


Fig. 2 Results of measured water quality parameters

A1 and M2 have the 1st class water quality, T1, M1, and E1 has the 2nd class water quality in terms of NO₃ (TSWQR, 2021).

The order of the basin components in terms of phosphate accumulations in water is as follows: Çorlu Stream (Ç1) > midstream of Ergene River (E2) > downstream of Ergene River (E3–E4) > Anadere Stream (A1) > Tunca River (T1) > Meriç River (M1–M2) > upstream of Ergene River (E1). Although the phosphate levels in the waters of Çorlu Stream and mid-down-stream of the Ergene River were recorded as extremely high levels and much higher than the other basin components, the PO₄ values in the waters of all basin components were quite high and the entire basin waters have the 3rd class

(>0.16 mg/L) water quality in terms of this parameter in both seasons (TSWQR, 2021).

Ecological risk assessment

The results of the Water Quality Index (WQI) and Nutrient Pollution Index (NPI) applied for a synergistic evaluation of the organic pollution status of the Meriç–Ergene River Basin in dry and wet seasons with the rating information are given in Fig. 3.

According to the results of WQI and NPI, in which the pressure of salinization–organic pollution variables (EC, NO₃, and PO₄ for WQI; NO₃ and PO₄ for NPI) on the system were evaluated, the recorded

scores of applied indices in the Çorlu Stream and mid-down-stream of the Ergene River were much higher than those detected in the Anadere Stream, Tunca and Meriç Rivers and upstream of the Ergene River.

The values of the applied ecological risk assessment indices detected in the water of the basin components showed significant changes and improvements between the investigated seasons. The recorded percentage changes of average values of applied ecological risk assessment indices from dry to wet seasons were as follows: -41.14% for WQI and -36.02% for NPI.

As a result of applied WQI, Tunca River (T1), Meriç River (M1–M2), and upstream of Ergene River (E1) were recorded as “Good quality,” Anadere Stream (A1) was recorded as “poor quality,” and Çorlu Stream (Ç1) and mid-down-stream of Ergene River (E2–E4) were recorded as “unsuitable” in the dry season, while the stations of E1 and M1 were recorded as “excellent quality,” T1, M2, and A1 were recorded as “good quality,” E3 and E4 were recorded as “poor quality,” and Ç1 and E2 were recorded as “unsuitable” in the wet season.

As a result of applied NPI, Anadere Stream (A1), Tunca River (T1), Meriç River (M1–M2), and upstream of Ergene River (E1) were recorded as “moderately polluted” and Çorlu Stream (Ç1) and mid-down-stream of Ergene River (E2–E4) were recorded as “very highly polluted” in the dry season, while the stations of E1 and M1 were recorded as “not polluted,” T1, M2 and A1 were recorded as “moderately polluted,” E3 and E4 were recorded as “considerably polluted,” and Ç1 and E2 were recorded as “very highly polluted” in the wet season.

It has been determined that the contamination degrees of applied salinization–organic pollution risk assessment indices generally increased in the dry season and decreased in the rainy season. The pollution statuses of the investigated basin components were as follows: Çorlu Stream > Ergene River > Anadere Stream > Tunca River > Meriç River in general in terms of both WQI and NPI.

Noncarcinogenic health risk assessment

The results of applied noncarcinogenic health risk assessment of nutrient-based pollutants (NO₃ and NO₂) for the water of Meriç–Ergene River Basin in dry and wet seasons are given in Table 4.

According to the results of applied noncarcinogenic health risk assessment, the upstream of Meriç River (M1) and up-down-stream of Ergene River (E1 and E4) were determined as “negligible (HI < 0.1),” while the Anadere Stream (A1) and Çorlu Streams (Ç1), Tunca River (T1), downstream of Meriç River (M2), and midstream of Ergene River (E2 and E3) were determined as “low risk (0.1 < HI < 1)” in terms of HI values during the dry season.

The values of the applied toxicological risk assessment indices detected in water of the basin components showed significant changes between the investigated seasons. Significant increases were determined in the HQ_NO₃, HQ_NO₂, and HI values detected in all the basin components during the rainy season and all the investigated locations in the watershed during this season were classified as “low risk” in terms of HI values. The recorded percentage changes of average values of applied toxicological risk assessment indices from dry to wet seasons were as follows: +126% for HQ_NO₃, +82.19% for HQ_NO₂, and +92.04% for HI.

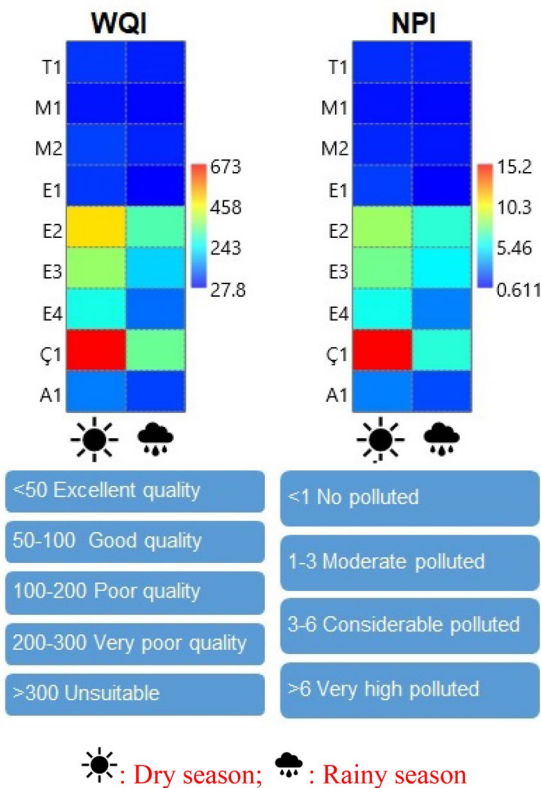


Fig. 3 Results of applied ecological risk assessment indices

Table 4 Probable noncarcinogenic health risks of nutrients in water of Meriç–Ergene River Basin

Indicator	Group	Basin components–stations											
		T1	M1	M2	E1	E2	E3	E4	Ç1	A1			
Dry season	HQ-NO ₃	Infants	3.300E-02	1.700E-02	3.100E-02	8.000E-03	9.100E-02	1.100E-02	5.500E-02	3.250E-03	3.250E-02	4.800E-02	
		Children	2.338E-01	1.204E-01	2.196E-01	5.667E-02	6.446E-01	7.792E-02	3.896E-02	2.302E-01	2.302E-01	3.400E-01	
		Teenagers	1.650E-01	8.500E-02	1.550E-01	4.000E-02	4.550E-01	5.500E-02	2.750E-02	1.625E-01	1.625E-01	2.400E-01	
	HQ-NO ₂	Adults	1.322E-01	6.811E-02	1.242E-01	3.205E-02	3.646E-01	4.407E-02	2.204E-02	1.302E-01	1.302E-01	1.923E-01	
		Infants	9.040E-03	2.000E-03	4.560E-03	3.040E-03	7.280E-03	4.400E-03	6.320E-03	1.016E-02	1.016E-02	8.640E-03	
		Children	6.403E-02	1.417E-02	3.230E-02	2.153E-02	5.157E-02	3.117E-02	4.477E-02	7.197E-02	7.197E-02	6.120E-02	
	HI*	Teenagers	4.520E-02	1.000E-02	2.280E-02	1.520E-02	3.640E-02	2.200E-02	3.160E-02	5.080E-02	5.080E-02	4.320E-02	
		Adults	3.622E-02	8.013E-03	1.827E-02	1.218E-02	2.917E-02	1.763E-02	2.532E-02	4.071E-02	4.071E-02	3.462E-02	
		Infants	4.204E-02	1.900E-02	3.556E-02	1.104E-02	9.828E-02	1.540E-02	1.182E-02	4.266E-02	4.266E-02	5.664E-02	
	Wet season	HQ-NO ₃	Children	2.978E-01	1.346E-01	2.519E-01	7.820E-02	6.962E-01	1.091E-01	8.373E-02	3.022E-01	4.012E-01	2.832E-01
			Teenagers	2.102E-01	9.500E-02	1.778E-01	5.520E-02	4.914E-01	7.700E-02	5.910E-02	2.133E-01	2.133E-01	2.269E-01
			Adults	1.684E-01	7.612E-02	1.425E-01	4.423E-02	3.938E-01	6.170E-02	4.736E-02	1.709E-01	1.709E-01	2.269E-01
HQ-NO ₂		Infants	3.150E-02	3.600E-02	8.000E-03	2.000E-02	7.850E-02	4.950E-02	3.700E-02	7.900E-02	7.900E-02	2.500E-03	
		Children	2.231E-01	2.550E-01	5.667E-02	1.417E-01	5.560E-01	3.506E-01	2.621E-01	5.596E-01	5.596E-01	1.771E-02	
		Teenagers	1.575E-01	1.800E-01	4.000E-02	1.000E-01	3.925E-01	2.475E-01	1.850E-01	3.950E-01	3.950E-01	1.250E-02	
HQ-NO ₂		Adults	1.262E-01	1.442E-01	3.205E-02	8.013E-02	3.145E-01	1.983E-01	1.482E-01	3.165E-01	3.165E-01	1.002E-02	
		Infants	6.800E-03	4.560E-03	1.032E-02	1.680E-03	1.512E-02	1.384E-02	1.232E-02	1.296E-02	1.296E-02	1.816E-02	
		Children	4.817E-02	3.230E-02	7.310E-02	1.190E-02	1.071E-01	9.803E-02	8.727E-02	9.180E-02	9.180E-02	1.286E-01	
HI*		Teenagers	3.400E-02	2.280E-02	5.160E-02	8.400E-03	7.560E-02	6.920E-02	6.160E-02	6.480E-02	6.480E-02	9.080E-02	
		Adults	2.724E-02	1.827E-02	4.135E-02	6.731E-03	6.058E-02	5.545E-02	4.936E-02	5.192E-02	5.192E-02	7.276E-02	
		Infants	3.830E-02	4.056E-02	1.832E-02	2.168E-02	9.362E-02	6.334E-02	4.932E-02	9.196E-02	9.196E-02	2.066E-02	
HI*	Children	2.713E-01	2.873E-01	1.298E-01	1.536E-01	6.631E-01	4.487E-01	3.494E-01	6.514E-01	6.514E-01	1.463E-01		
	Teenagers	1.915E-01	2.028E-01	9.160E-02	1.084E-01	4.681E-01	3.167E-01	2.466E-01	4.598E-01	4.598E-01	1.033E-01		
	Adults	1.534E-01	1.625E-01	7.340E-02	8.686E-02	3.751E-01	2.538E-01	1.976E-01	3.684E-01	3.684E-01	8.277E-02		

*HI coefficients with a probable low risk of human health are given in bold (0.1 < HI)

The locational nutrient pollutant based noncarcinogenic health risk statuses of the Meriç–Ergene River Basin were as follows: $E2 > A1 > \text{Ç1} > T1 > M2 > M1 > E3 > E4 > E1$ during the dry season and $E2 > \text{Ç1} > E3 > E4 > M1 > T1 > E1 > A1 > M2$. Also, the CDI, HQ, and HI risk rankings among the investigated different age groups for all the basin components in both seasons were found as follows: children > teenagers > adults > infants.

Statistical evaluation

Results of the applied Pearson Correlation Index (PCI) with the correlation coefficients are given in Fig. 4 ($n=18$). As a result of PCI, statistically significant negative correlations were detected among the values of dissolved oxygen (DO) with the salinization (EC, TDS, salinity, and Cl) and organic pollution parameters (nitrate, phosphate, and sulfate) ($p < 0.01$), while significant positive correlations were detected among the variables of EC, TDS, salinity, and Cl, NO_3 , PO_4 , and SO_4 ($p < 0.01$).

In this investigation, Principal Component Analysis (PCA) was applied to detected chemical data to determine the effective varifactors on surface water quality of the Meriç–Ergene River Basin ($n=18$ and eigenvalues > 1). The Kaiser–Meyer–Olkin (KMO) test was performed before implementing PCA, and the result of the KMO test that reflects the sampling sufficiency was recorded as 0.813, which means it was quite enough (> 0.5). As a result of applied PCA, 2 factors explained 83.453% of the total variance. Component

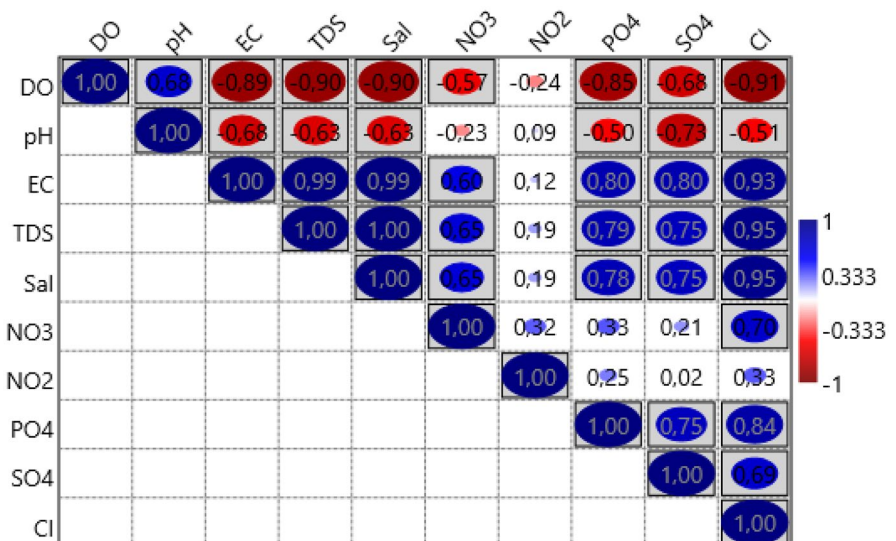
1 (F1—first factor) was named “industrial factor” and explained 63.105% of the total variance. It was related to the variables of EC, TDS, salinity, sulfate, DO, chloride, pH, and phosphate. Component 2 (F2—second factor) was named “agricultural factor” and explained 20.348% of total variance. It was related to the variables of nitrite and nitrate (Fig. 5).

In the current research, Cluster Analysis (CA) was applied to detected chemical data to classify the investigated locations in the Meriç–Ergene River Basin in terms of their similar water quality characteristics. As a result of applied CA, 4 clusters were formed, which were named “Uncontaminated Zone—C1,” “Less Contaminated Zone—C2,” “Contaminated Zone—C3,” and “Highly Contaminated Zone—C4” (Fig. 6). C1 was formed by the Meriç River (M1) and the Ergene River (E1); C2 was formed by the Anadere Stream (A1) and the Tunca River (T1); C3 was formed by the Meriç River (M2) and the Ergene River (E4); and C4 was formed by the orlu Stream (1) and the midstream of the Ergene River (E2 and E3).

Discussion

In this research, spatial–temporal variations of water quality of the Meriç–Ergene River Basin were investigated. As a result of this research, significant spatial differences in water qualities were recorded indicating the influence of anthropogenic applications and

Fig. 4 Results of applied PCI and correlation coefficients



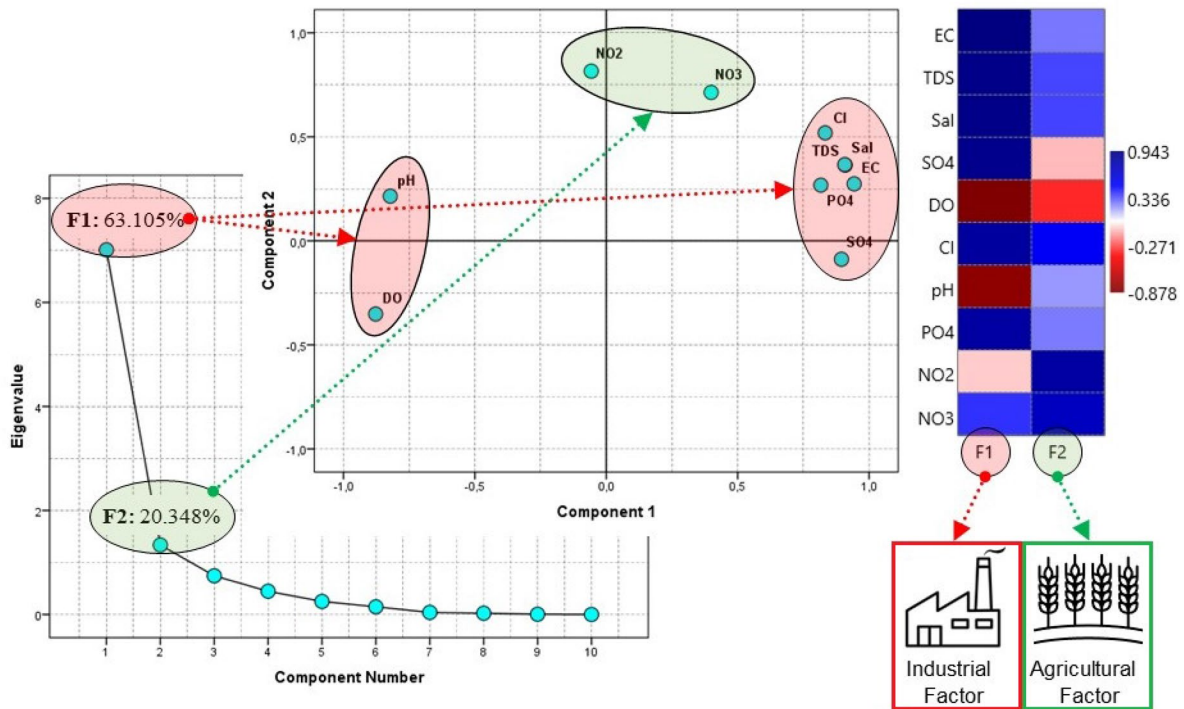


Fig. 5 Component plots with the factor loads of rotated component matrix

also significant temporal differences in water qualities indicating the significant effects of the dry and rainy seasons (Hamid et al., 2020; Varol & Tokatlı, 2022).

Çorlu Stream is an extremely polluted fluvial habitat probably due to domestic and industrial wastewater discharges from Çorlu and Çerkezköy districts, the 2 most populated and industrialized settlements of the Thrace Region and when you go near to the stream it is quite easy to estimate the water quality and pollution from its color and smell. It is known that although the Ergene River is being exposed to an intensive organic–inorganic pollution pressures just right after its source region, the water quality of Ergene River decreases much more after flowing of Çorlu Stream into the Ergene River (Aydın & Elipek, 2022; Celen et al., 2022; Tokatlı, 2021; Tokatlı & Varol, 2021a; Varol & Tokatlı, 2023). According to the applied CA, Çorlu Stream (Ç1) and midstream of Ergene River (E2 and E3) formed a separate cluster named as “Highly Contaminated Zone” (Fig. 6).

Meriç River, which is known as not affected by point-source pollution of industrial–domestic origin as much as Çorlu Stream, is known as affected

by non-point-source pollution of agricultural origin (Tokatlı & Ustaoglu, 2020; Tokatlı & Varol, 2021a; Celen et al., 2022). Due to significant increases in the flow rates of rivers, significant desalination rates were detected in almost all the basin waters, especially in Çorlu Stream and Ergene River, while significant increases were detected in the nutrient levels in almost all the basin waters due to increases in surface runoff in this season, especially in the Meriç River. As a result of PCA, 2 statistically effective factors were detected. The variables of industrial factors (EC, TDS, salinity, sulfate, DO, chloride, pH, and phosphate) were decreased significantly in the rainy season, while the variables of agricultural factors (nitrite and nitrate) were increased significantly in this season (Fig. 5).

Dissolved oxygen is known as a key component of freshwater, and it plays a vital role in everything from the livelihood of microorganisms, plants, and animals to the quality of water (Kannel et al., 2007). When oxygen levels are less than 2 ppm, the water is defined as hypoxic, which may lead to the death of many aquatic organisms (CENR, 2000). Extreme hypoxic conditions were recorded in the Çorlu Stream (Ç1) and midstream

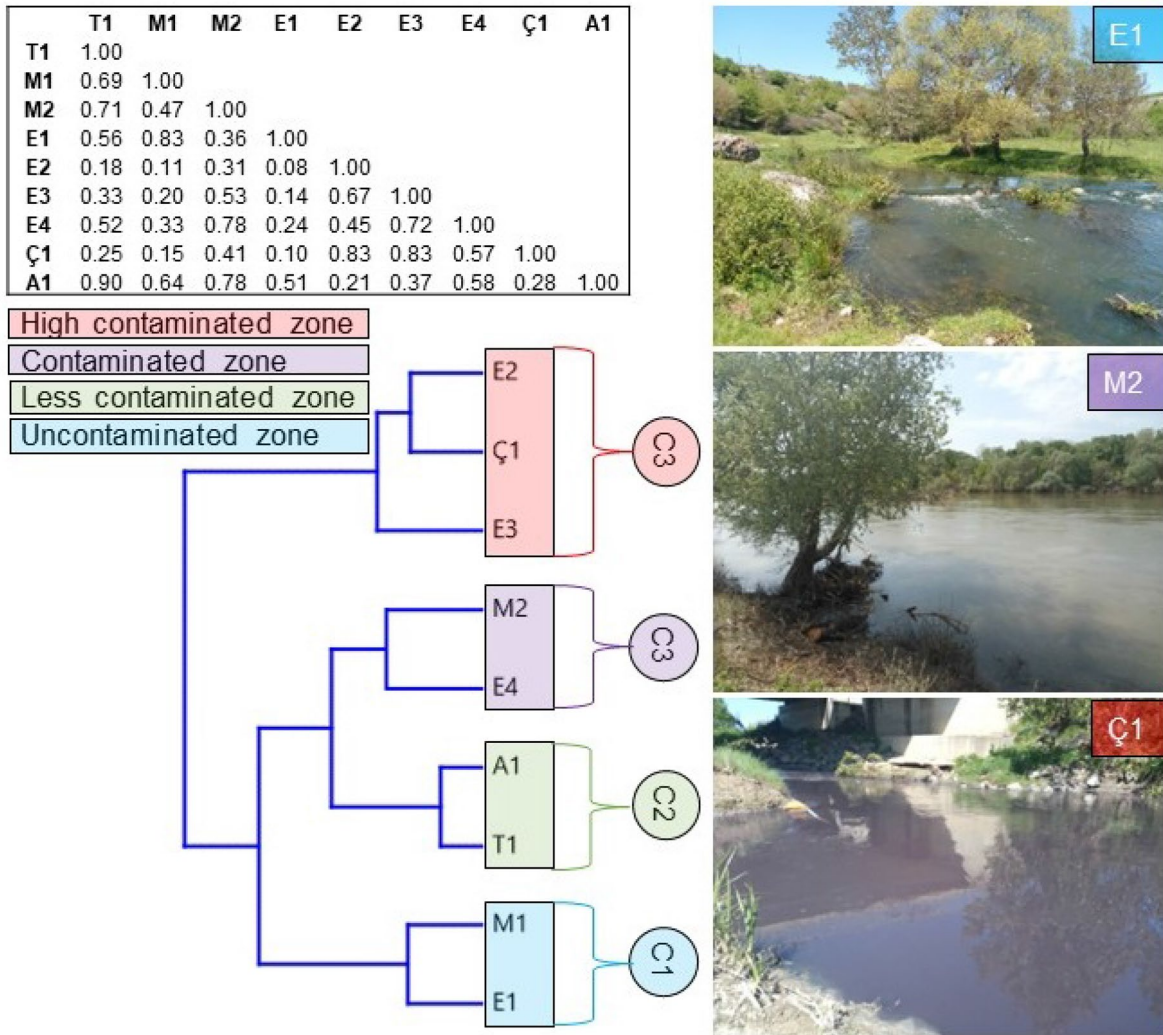


Fig. 6 Hierarchical dendrogram of CA

of the Ergene River (E2). Although DO levels in basin waters increased by an average of 100% during the wet season, no significant increases were observed in the Corlu Stream or the Ergene River’s midstream. DO levels were recorded as almost zero in the water of Çorlu Stream in both seasons (0.26 mg/L in the wet season and 0.27 mg/L in the dry season). In addition, DO levels detected at the E2 station, which was selected right after the point where Çorlu Stream flows into the Ergene River, are also extremely low and recorded as 0.17 mg/L and 0.34 mg/L, respectively, in the dry and rainy seasons.

High organic matter content and decay of organic matter result in the consumption of dissolved oxygen

by microorganisms living in the water (Nong et al., 2020). In this research, the highest levels of indication parameters of organic pollution such as nitrite, phosphate, and sulfate were measured at the stations of Ç1 and E2. Also, as a result of selected ecotoxicological risk assessment indices as organic pollution indicators in terms of ecosystem (WQI and NPI) and human (CDI, HQ, and HI) health, Çorlu Stream was found as extremely contaminated by nutrients and posed a probable non-carcinogenic health risk of nutrients (low risk: $0.1 < HI < 1$). The quite high levels of organic pollutants in Çorlu Stream and Ergene River are thought to be the main reasons for the extreme

hypoxic conditions. Also, the oxygen-depleted waters in these potamic habitats have been associated with the nutrient-rich discharges from settlements and food production–processing facilities located in their watersheds.

Today, most inland waters are faced with the threat of salinization, which has become an increasing risk factor for freshwater habitats and ecosystem functions, due to anthropogenic activities (Canedo-Arguelles et al., 2016). In this research, the highest EC, TDS, and salinity parameters were recorded in Çorlu Stream (an average of 4195 $\mu\text{S}/\text{cm}$ for EC, 2740 mg/L for TDS, and 2.81 ‰ for salinity), followed by the mid-down-stream of the Ergene River (E2–E4). It was also determined that these parameters decreased significantly from midstream to downstream in the water of the Ergene River due to the dilution of salinization parameters in the waters by the flowing of many tributaries with low salinity into the Ergene River from the midstream to downstream.

It was reported that approximately 190 textile industries, where dyeing and bleaching processes are intensively carried out, were in the watershed of Çorlu Stream (Güneş et al., 2008). It was also reported that Çorlu Leather Organized Industrial District contains about 120 leather plants of different sizes and categories, which produce about 40% of total leather production in Türkiye (Töre et al., 2011). Washing, tanning, greasing, and dyeing processes conducted in the leather plants can be described as the main sources which contribute to the nitrogenous–phosphorus compounds and EC contamination in the close freshwater habitats (Celen et al., 2022). In this research, the highest levels of salinization and organic pollution variables were recorded in the Çorlu Stream, representing the point source pollution zone of the Textile and Leather Industries Organized Zone and wastewater discharges from Çorlu and Çerkezköy districts, the most populated settlements of the Thrace Region into the Çorlu Stream.

Corlu Stream, which local governments had ignored for a long time and had tolerated this extensive contamination, was recently protected by the Ministry of Environment and Forestry, along with the entire Ergene River Basin. Although there are a number of industrial facilities in the watershed of Çorlu Stream, it has been prevented by the Ministry of Environment and Forestry that the factories discharge their waste into the

stream without treatment, thanks to the hard measures and inspections taken, especially in recent years. In addition, it was reported in much research performed in the Meriç–Ergene River Basin that the water quality is improving day by day with the “Action Plan of the Ergene Basin Protection” carried out by the Ministry of Environment and Forestry (Aydin & Elipek, 2022; Celen et al., 2022; Çingiroğlu, 2018; Emadian et al., 2021; Orak et al., 2020; Tokatlı, 2015; Tokatlı & Başatlı, 2016; Tokatlı & Ustaoglu, 2020; Tokatlı & Varol, 2021a, b; Varol & Tokatlı, 2023).

Given that the Meriç–Ergene River Basin is heavily used for irrigation of fields to grow Turkey’s main stocks of rice, wheat, and sunflower, which are also exported to Europe, monitoring the water quality of the entire watershed components is critical not only for ecosystem health but also for public health. As a result of our study, the organic pollution pressure on the Meriç–Ergene River Basin components was evaluated by ecotoxicological and statistical indicators, and the riskiest fluvial components of the watershed were determined. Thus, it has been clearly revealed which lotic habitats on the basin should receive more attention while making the basin’s sustainability and conservation plans.

Conclusions

In this research, some multistatistical water quality assessment methods and organic pollution-based ecotoxicological risk assessment indices were used to evaluate the surface water quality of the Meriç–Ergene River Basin. With their relatively high salt and nutrient contents and extreme hypoxic conditions, Corlu Stream and the Ergene River were identified as the most contaminated basin components in this study.

The recorded WQI and NPI scores in the Corlu Stream and mid-downstream of the Ergene River were much higher as a result of applied ecotoxicological risk assessment indices than those detected in the Anadere Stream, Tunca, and Meriç Rivers and upstream of the Ergene River; and the locational nutrient pollutant based noncarcinogenic health risk statuses of the Meriç–Ergene River Basin were as follows: $E2 > A1 > \text{Ç}1 > T1 > M2 > M1 > E3 > E4 > E1$ during the dry season and $E2 > \text{Ç}1 > E3 > E4 > M1 > T1 > E1 > A1 > M2$ during the wet season.

As a result of multistatistical risk assessment indicators, 2 statistically effective factors were determined, which were named as “Industrial Factor” and “Agricultural Factor” according to the results of applied PCA; and 4 clusters were formed, which were named as “Uncontaminated Zone”, “Less Contaminated Zone”, “Contaminated Zone,” and “Highly Contaminated Zone” according to the results of applied CA.

In line with the data obtained in this research, it can be stated that, Çorlu Stream and Ergene River cannot support most aquatic life, except a few extremophiles and eurioc species, because of high organic contamination and extreme hypoxic conditions. In order to improve the quality and provide the sustainability of aquatic life in the Meriç–Ergene River Basin, the use of unconscious pesticides and fertilizers must be avoided in the region, discharges of industrial wastewater without any treatment must be prevented, and discharges of municipal sewage without any treatment must also be prevented.

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Author contribution All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by Cem Tokatlı and Abu Reza Md. Towfiqul Islam. The first draft of the manuscript was written by Cem Tokatlı, and both authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Data availability Data are available based on reasonable request on corresponding author.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Conflict of interest The authors declare no competing interests.

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