

Environmental monitoring of Micro Prespa Lake basin (Western Macedonia, Greece): hydrogeochemical characteristics of water resources and quality trends

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Abstract The Micro Prespa basin is a trilateral catchment area of significant importance with a unique ecosystem closely related to the homonymous lake. In this frame, a fully operational monitoring project was carried out including continuous real-time measurements in Micro Prespa Lake with the use of a multi-sensor probe, as well as periodical sampling and analyses of all available water systems for an extended set of 85 parameters. Four main interacting water systems were identified, including alluvial and karstic aquifers, Micro Prespa Lake and adjacent drainage network. The results outlined that general environmental conditions are satisfying in respect to the relative legislation and the hydrogeochemical signatures. However, trends of environmental pressures were ascertained as a result of natural (geogenic) factors, embracing seasonal peaks for Ni, Pb, and NH₄ mainly in groundwater systems. Based on chlorophyll *a* records, Micro Prespa is classified as oligotrophic to slightly mesotrophic, subjected to seasonal variations. Heavy metal concentrations are low, except Ni which appears to have elevated values during the dry hydrological period. Finally, the hydrogeochemistry of drainage network is primarily influenced by surface runoff of the surrounding mountainous areas, hence elevated phosphorus values of the Aghios Germanos stream are possibly linked with the leaching of the granitic formations on the east.

Keywords Environmental monitoring · Micro Prespa Lake · Prespa basin · Water quality · Hydrogeochemistry

Introduction

The Prespa area is considered as a region of significant environmental, aesthetical, and cultural value, located in the Balkan Peninsula and shared among three countries (Greece, Albania, and the former Yugoslav Republic of Macedonia (FYROM)); in principal, Prespa includes the two renowned lakes of Micro (Mikri) and Macro (Megali) Prespa that form a transboundary unique natural monument protected by RAMSAR convention as a wetland of international importance (RAMSAR 1974). The two lakes lie at an altitude of around 850 m above sea level, though many of the mountainous surrounding areas exceed 2,000 m. Lake Macro Prespa is shared by three countries, with the largest extent belonging to FYROM while Lake Micro Prespa belongs almost entirely to Greece, except for a small part in the southwest which lies in Albania. The previous decade, a joint Declaration by the Prime Ministers of the three countries demonstrated the importance of the area by creating the Transboundary Prespa Park, which aimed for the protection of the ecological values through the trilateral collaboration, and also to promotion of the economic prosperity of the local communities. The Transboundary Prespa Park is the first transboundary protected area in the Balkans and southeastern Europe (SPP 2013).

The significance of Prespa Lakes is related with their exceptional biodiversity which is reflected to the great

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number of life forms concentrated in such a small geographical area. The region has been recognized as a European and Global Hotspot of Biodiversity (Albrecht et al. 2008; UNDP GEF 2013), not only because of the sheer number of species and habitats present, but also due to their quality, such as rarity and conservation significance. Most of the species are endemic, like the Prespa barbell (*Barbus meridionalis*), bleak (*Alburnus albidus*), and *Chondrostoma nasus* (Cullaj et al. 2005). Water birds migrate through and winter in the region; hence Prespa is characterized as an important habitat (Special Protected Area) by the 79/409/EEC Directive (Catsadorakis 1996). Most important species are the Dalmatian pelican (*Pelecanus crispus*) and pygmy cormorant (*Phalacrocorax pygmeus*). The extent of fauna's biodiversity is demonstrated by the fact that more than half of the species (birds, amphibians, and mammals) which are found in Greece as a whole, inhabit the Prespa area while 9 out of the 23 fish species found are endemic (SPP 2013). Flora is important too, as according to Pavlides (1997), in the wider Prespa Park area more than 1,300 species exist and many of them like *Centaurea prespans* are characterized as endemic and of major importance. In addition, Prespa's region fertile parts are cultivated with the famous Prespa bean, which is a product of Protected Geographical Indication (PGI) and constitutes the main source of income for the local community (Abeliotis et al. 2013).

Micro Prespa's ecosystem is facing various challenges stemming from the existing land use activities and the physiographic characteristics that directly or indirectly affect the entire Prespa basin; the key element to vital ecosystem's functions is water, either as available quantity or as overall quality; water quantity (water level) is being monitored regularly, but water quality is partially monitored, covering in most cases only few physicochemical parameters (e.g., pH, turbidity, total dissolved solids (TDS), etc) or sporadic individual analyses (Abeliotis et al. 2013; Hollis and Stevenson 1997; Mourkidou 2002; Petaloti et al. 2004; Scoullou and Hatzianestis 1989) for a limited range of parameters and within a short and discontinuous timespan of monitoring assessment.

In this frame, the present study embraces the development, demonstration, and assessment of a fully operational and integrated environmental monitoring system for the water quality characteristics of Lake Micro Prespa and its adjacent interacting water systems,

towards the sustainable development of the area. The developed monitoring scheme is in compliance with the requirements and recommendations of the Water Framework Directive (EC 2000) by which "all European surface waters should be monitored in order to be in good quality conditions by the end of 2015". Even though the analyzed parameters were selected to focus on significant environmental pressures (e.g., eutrophication and heavy metals pollution), they may also serve as fundamental data for the generic assessment of the overall hydrogeochemical trends of the water systems within the Micro Prespa basin. Concretely, the main goals are: (a) the in-depth and updated assessment of Micro Prespa's water quality status, as well as of the interacting water systems, (b) coupling different monitoring methods (e.g., continuous real-time records, periodic in situ measurements, laboratory analyses and macroscopic investigations) and diverse environmental parameters (e.g., chlorophyll *a*, DO, major ions, heavy metals and trace elements, etc.) in order to provide a meaningful and robust tool for environmental assessment, (c) Assess the individual values and the relevant fluctuations of key environmental parameters like nutrients (NO₃, NH₄, and P) and chlorophyll *a* within the timespan of a full hydrological period, so as to outline potential trends and environmental threats.

The above assessments may provide an integrated monitoring scheme that will serve as a holistic approach to similar cases where environmental assessments between interacting water systems are required including both surface and groundwaters.

Physical setting

The climate of Prespa area is characterized as a hot and dry Mediterranean type during summer (July 23.6 °C) and mid-European type during winter with long periods of high rainfall, snow, increased cloudiness and low temperatures (January, 0.8 °C) (Hollis and Stevenson 1997). Under an Emberger bioclimatic classification the area is typical of a humid Mediterranean type (Emberger 1963). Previous researchers reported that mean annual rainfall varies from 647 to 742 mm (Bonazoundas 1988; Kassioumis 1991) which is typical for low-lying agricultural lands. Taking into account that mean annual snowfall is about 300 mm (Kassioumis 1991) total precipitation may reach up to 1,000 mm per year, a fact which denotes the significant amount of water recharge

to the entire system. The mean air temperature remains above freezing throughout the year but Lake Micro Prespa is frequently frozen probably due to the descending cold air-streams from the surrounding mountains.

With regard to geology (Fig. 1), the wider area of Prespa Lakes is considered as a major tectonic subduction (graben) which is oriented in a NNW–SSE direction (Amataj et al. 2007). The current structure is believed to be a result of a karstic collapse which consequently created the Prespa Lakes system in Tertiary. Substrate geology is divided into two main parts. The western one is dominated by karstified limestones and dolomites with intercalations of Ni-rich lateritic horizons and a complex tectonometamorphic formation (mélange)

which is consisted of serpentinized Ni-rich ophiolitic blocks and schists. The eastern one is characterized by the presence of granites, gneisses and metamorphic clastic formations (conglomerates, sandstones) of Paleozoic and Lower Triassic age (Kassioumis 1991). The eastern sequence is covered by recent alluvial deposits of variable thickness.

In respect to water resources, the hydrological regime is well documented (e.g., Hollis and Stevenson 1997; Kolaneci 2004; Parisopoulos et al. 2009) in contrast with the hydrogeological setting that appears to be more complicated and insufficiently described. The main water inputs of Micro Prespa Lake are the surface runoff and sub-surface discharge as a result of lateral cross

Fig. 1 Simplified geological map of Micro Prespa Basin



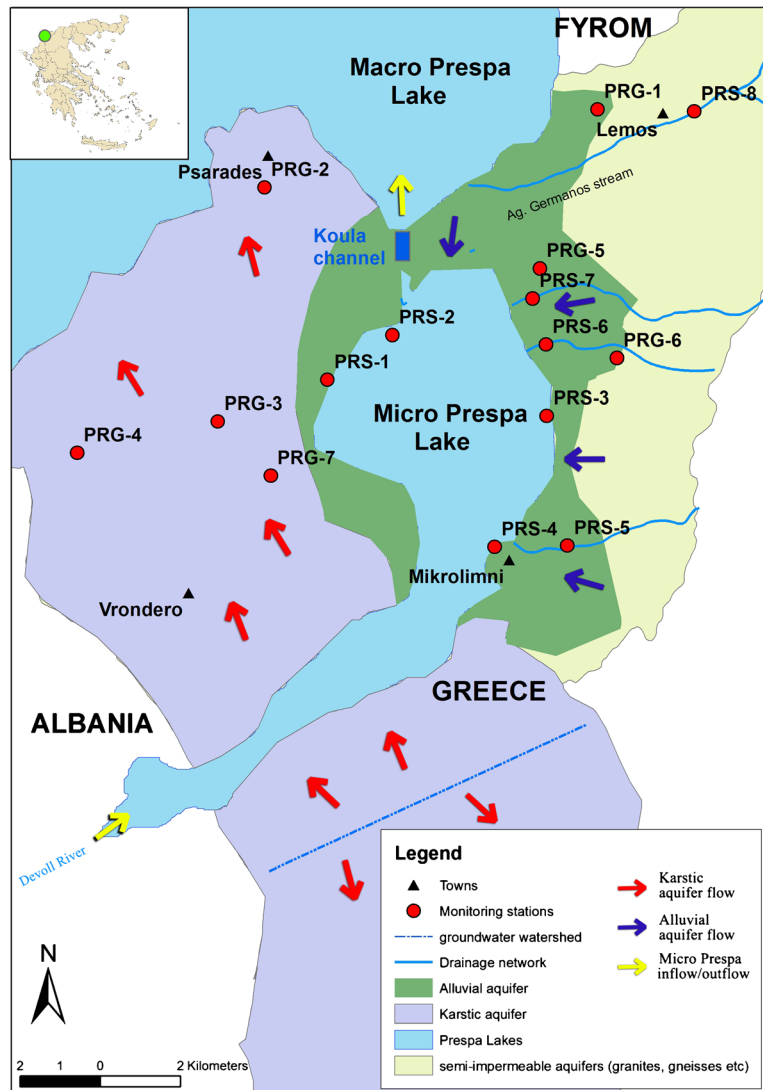
flows from the interacting aquifer systems (Fig. 2). Surface runoff may be summarized to (i) inflows from Devoll River (SW part of Micro Prespa Lake) which derives from Albania, as well as from (ii) temporal creeks mainly on the eastern side of the lake which serve as occasional sources of water inflow, but may only be regarded as of minor importance. On the other hand, main water outputs should be considered (i) the artificial flow through Koula channel (sluice gates) which transfers water to Macro Prespa, and (ii) the irrigation of adjacent crop cultivations through an artificial drainage network, with the aid of a pumping station.

The hydraulic connection of Micro Prespa Lake with the interacting aquifer systems operates both as water inflow and outflow, depending on the local conditions

and the complex hydrogeological regime. Based on the limited hydrogeological surveys that have been conducted so far (Stamos et al. 2011; Stamos and Stefouli 2010) there are two major aquifer systems developed in Micro Prespa basin (Fig. 2). These systems are characterized by variable hydrogeological conditions, with differences in their overall potential and discharge rates. The first system consists of a karstic aquifer body with high permeability and velocities of groundwater flow, while the second one is composed by the recent lacustrine and terrestrial alluvial deposits, with variable permeability and significantly lower potential than the karstic aquifer.

A general overview of the hydrogeological setting is given in Fig. 2, where according to Stamos et al. (2011)

Fig. 2 Simplified hydrogeological map of the study area (modified from Stamos et al. 2011) and monitoring network of Micro Prespa basin



there is a major sub-surface watershed (SW-NE direction) of the karstic aquifer which drives groundwater flow towards Micro Prespa and towards southeast (Kastoria area). Additionally, alluvial aquifer flow is heading to the west. Nevertheless, several other flows are developed within the karstic system, which create temporally semi-individual small aquifers of variable flow directions. The most characteristic one is the aquifer which is developed between Micro and Macro Prespa and changes groundwater direction according to the hydrological period of the year.

Materials and methods

The environmental monitoring system of Micro Prespa Lake basin included real-time records of quality parameters (Micro Prespa Lake) and conventional field sampling (karstic and alluvial aquifers, Micro Prespa Lake and drainage network) within a hydrological year (2012–2013). In respect to real-time records, a multi-sensor probe was installed at a constant depth of 1.5 m below the lake’s surface (total depth in that point was nearly 3 m with minor fluctuations during the year) at the northern side of the lake (PRS2) and values were recorded every 15 min for a total of six quality parameters: pH, electrical conductivity (EC), dissolved oxygen (DO), TDS, chlorophyll *a* (chl *a*) and water temperature (T). The installation point was selected as the most ideal to fulfill some general requirements like representativeness of measurements, safety of equipment and convenience of maintenance/calibration.

In addition to real-time records, four sampling campaigns were conducted to all available water systems during January, April, July, and October (2012–2013) in an effort to simulate the conditions of wet (January, April) and dry (July, October) hydrological periods. Samples were collected from 15 sampling sites which corresponded to four different water systems (Fig. 2):

- (a) Micro Prespa Lake (PRS1–PRS4)
- (b) Drainage network (PRS5–PRS8)
- (c) Karstic aquifer including spring discharge (PRG2, PRG3, PRG4, PRG7)
- (d) Alluvial aquifer (PRG1, PRG5, PRG6)

The unstable field parameters of pH-Eh as well as EC, TDS, and water temperature were measured in situ by means of portable instruments. Water samples were

immediately filtered in the field through conventional 0.45 μm membrane filters and separated into two aliquots of 1 L and 50 ml, respectively. The smaller aliquot were acidified down to pH≈2 with HNO₃ in order to prevent metal precipitation and complexation and stored in new polyethylene bottles for metal analyses. Samples were analyzed for a total of 79 parameters (Table 1) including major ions, trace elements, and physicochemical parameters.

Sampling was accompanied by extended field works which focused on macroscopic investigations and assessment of potential pollution sources, as well as supplementary geological and hydro-lithological mapping. The outcomes showed no obvious potential sources of external (anthropogenic) pollution to the system, apart from (i) the cultivated been plots, mainly on the eastern side of the basin, and (ii) the livestock near Vrondero village (western of the lake upwards of sample PRG7).

The initial raw data (real-time records, in situ measurements and laboratory analyses) were processed with the use of multivariate statistical techniques in order to outline any potential relations, trends or processes between the examined parameters.

Micro-probe’s real-time records for all (seven) parameters were statistically processed with R-mode factor analysis in order to understand the correlation structure of collected data and identify the most important factors contributing to the data structure. In R-mode factor analysis, the relationship among a number of observed quantitative variables is represented in terms of a few underlying, independent variables, called factors, which may not be directly measured or even measurable. The exact number of factors depends on the detail of the

Table 1 Quality parameters of the environmental monitoring system

4 Sampling campaigns (Alluvial and karstic aquifer, Micro Prespa Lake, Drainage network)

Major elements and ions (10): Ca²⁺, Mg²⁺, Na⁺, K⁺, Cl⁻, NO₃⁻, NO₂⁻, NH₄⁺, SO₄²⁻, HCO₃⁻

Trace elements (64): Ag, Al, As, Au, B, Ba, Be, Bi, Br, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Fe, Ga, Gd, Hf, Hg, Ho, In, Ir, La, Li, Lu, Mn, Mo, Nb, Nd, Ni, Os, P, Pb, Pd, Pt, Rb, Re, Rh, S, Sb, Sc, Se, Si, Sm, Sn, Sr, Ta, Tb, Te, Th, Ti, Tl, Tm, U, V, W, Y, Tb, Zn, Zr

Physicochemical parameters (5): Electrical Conductivity (EC), Total Dissolved Solids (TDS), pH, Eh, Water Temperature (T)

Multi-sensor probe (real-time records) (Micro Prespa Lake)

Physicochemical parameters (6): pH, EC, DO, TDS, chl *a*, water temperature (T)

interpretation and at least should explain the 80 % of total variance (communality >80 %); for the present study, three factors were selected which sufficiently explains the 81.3 % of the total variance. Initial data have been standardized in order to eliminate the influence of different units; a correlation matrix was created to account for the degree of mutually shared variability between individual pairs of water quality parameters and consequently were extracted the factor loadings that identify the groups of variables, most highly correlated among them; factor loadings were optimized with Varimax rotation method in order facilitate interpretation by providing simpler factor structure. The higher the factor loading of a parameter the greater is its participation to the examined factor. As high factor loadings were considered those above 0.750, as medium ones those among 0.500 and 0.750, and as weak those among 0.400 and 0.500 (Panda et al. 2006). Results are shown in Table 3.

The analytical data from surface and groundwater samples collected during the four sampling campaigns were treated with cluster analysis (CA) in order to group similar characteristics. The samples classified into a cluster possess high homogeneity within cluster and high heterogeneity between clusters. The levels of the similarity at which observations are merged are used to construct a dendrogram which provides a visual summary of clustering process, presenting a figure of groups and their proximity, with a dramatic reduction in dimensionality of the original data (Chapagain et al. 2009). In this study, hierarchical cluster analysis included the median values of all 21 parameters measured in situ or in laboratory for the entire set of surface and groundwater samples. CA was performed using Ward's method. Squared Euclidean distance was used as measure of distance, which is one of the most commonly adopted measures. Results are shown in Fig. 6.

Results and discussion

Surface water

The installed multi-sensor probe recorded in total 38,624 measurements during June 2012–May 2013. Individual records were collected telemetrically and were statistically processed in order to obtain a median value for each month (Table 2). Outliers (values over the 97.5 % of the statistical population or false values

Table 2 Median monthly values for the recorded quality parameters of multi-sensor probe

Month	Chl <i>a</i> μg/L	EC μS/cm	DO mg/L	TDS mg/L	pH	T °C
June	2.9	268	7.6	175	8.1	24.5
July	2.7	268	7.2	187	8.1	26.7
August	1.8	294	7.4	192	7.9	26.7
September	1.1	286	7.8	200	8.0	21.8
October	2.0	357	7.1	232	8.1	18.9
November	2.3	359	6.9	234	7.8	12.0
December	3.5	267	11.4	192	8.1	5.1
January	3.1	268	11.7	174	8.1	4.2
February	2.8	297	11.7	195	8.1	5.3
March	3.5	347	11.2	227	8.2	7.9
April	2.2	403	9.8	263	7.9	12.2
May	1.3	449	6.7	294	8.1	19.7
median value	2.5	296	7.7	198	8.1	15.6

recorded during maintenance/calibration works) were filtered out and excluded from further data processing. Even though records were taken at a constant depth, they could be considered as representative for the entire column, if we take into account that Micro Prespa is a shallow lake and due to its polymictic character is under a mixing state (“turn over” state) during the winter period (Kagalou and Leonardos 2009; Löffler et al. 1998), with negligible variations between the water surface and the deepest water layer just above the sediment surface.

The parameter of chlorophyll *a* which estimates the biomass of planktonic algae in surface waters is considered as a critical factor for the assessment of a lake's trophic state and its quality classification (Jeppensen et al. 2003; Sondergaard et al. 2005). In that frame, chl *a* values were thoroughly monitored and found to be below or slightly above the mesotrophic threshold of 2.6 μg/L (Carlson and Simpson 1996; OECD 1982) hence Micro Prespa may be regarded as an oligotrophic to mesotrophic lake with seasonal variations. This result denotes a significant improvement in Micro Prespa's trophic status, as according to Tryfon et al. (1997a; b) previous recorded values indicated a clearly eutrophic state; good quality conditions are also confirmed by the fact that according to Poikane et al. (2010), who based on a large data set from European Lakes, reservoirs of similar characteristics (Alpine mid-altitude shallow lakes) with those of Micro Prespa's have a reference

value of around 3.4 µg/L for chl *a* which is higher than the annual median recorded value of 2.5 µg/L for Micro Prespa. As seen on Fig. 3, chl *a* concentrations are declining during the summer period and reach their minimum (1.1 µg/L) in August, while during autumn and early winter they rise and reach their maximum (3.5 µg/L) in December. Consequently, values are dropping again till February (2.8 µg/L) and they acquire their maximum in March (3.5 µg/L) until later, then start declining significantly till May (1.3 µg/L). Results show a clear seasonal variation with the highest values recorded during winter and early spring and the lowest during summer; this kind of fluctuation is typical for many oligotrophic to mesotrophic European Lakes (Constance, Garda, Como, Maggiore, and Geneva) according to Bresciani et al. (2011).

The overall nutrient load as expressed by total phosphorus (TP) and total nitrogen (TN) is a decisive element for chl *a* concentrations (Phillips et al. 2008). Hence, relevant fluctuations of their values may provide valuable information about the characteristics of Micro Prespa’s trophic status. Based on the measured values of the four reference sampling periods, the ratio of N/P ranges from 21 to 181, therefore is clear that the limiting factor is phosphorus. This fact is also documented by Fig. 4, which shows the relevant fluctuations of TP, TN and chl *a* values. It is clear that phosphorus variations follow those of chl *a*; when chl *a* increases, a relative enrichment in TP is also observed and vice versa (simultaneous decrease of TP and chl *a* values in October). Hence, seasonal peaks of chl *a* are related with phosphorus variations as a result of internal and external loadings. External loading should be attributed to

phosphate fertilizers and P-rich runoff from livestock (e.g., near Vrondero village) but its overall impact is expected to be limited compared with the internal loading which is associated with phosphorus release from the lake’s bottom.

Lake sediments have the capacity to absorb and retain P depending on their mineralogical composition and the relevant Fe concentrations (Forsberg 1989; Hupfer and Lewandowski 2008). In well-oxygenated waters, a surface-oxidized sediment micro-layer is formed at the bottom of the lake and can efficiently allow the development of P-binding processes due to changes in redox conditions; hence, Fe (hydro)oxides may therefore act as an “iron trap” for P which is retained in the sediments as long as the water layer is aerobic and the sediment contains more dissolved iron than dissolved P. However, a shift from aerobic to anaerobic conditions may give rise to the release of P from the sediment, as iron is no longer oxidized at the sediment water interface and iron trap will no longer operate. Accordingly, P, which was initially associated with Fe³⁺, is returned in solution and released from the sediment surface (Hupfer and Lewandowski 2008; Smolders and Roelofs 1993), thus explaining the seasonal variations of TP and chl *a* accordingly.

EC has practically constant values during summer period (268–294 µS/cm) and reaches a temporal peak in October–November; consequently, values are declining during December and from then they elevate constantly and reach their highest value in May (468 µS/cm). The annual median value of EC is 269 µS/cm and falls within the threshold (150–500 µS/cm) for the development of the majority of aquatic species (Behar 1997).

Fig. 3 Median monthly values of the real-time multi-sensor’s records (PRS2)

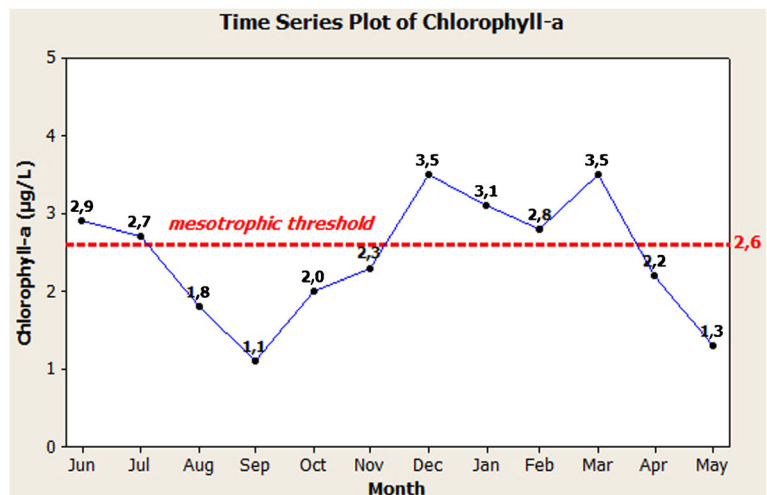
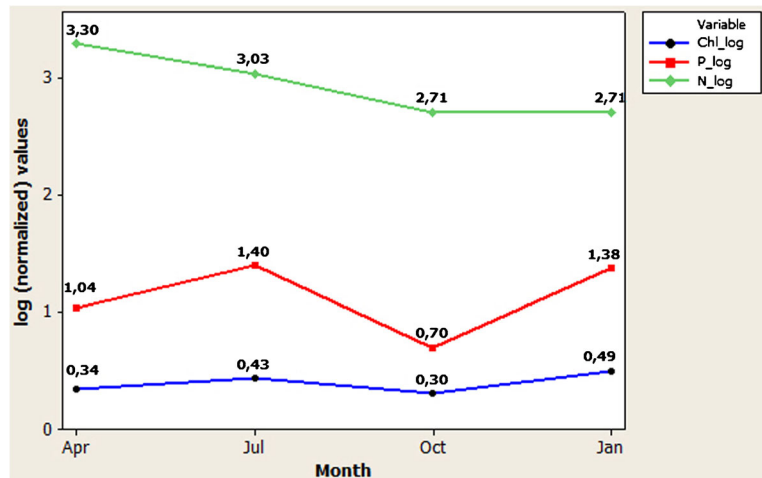


Fig. 4 Relative fluctuations of TN, TP, and chl *a* values for the four reference sampling periods. Normalization through log-transformation was performed for simplification and visual discrimination purposes



DO is also practically constant with minor variations during summer and autumn months (June–November) ranging from 6.9 to 7.8 mg/L; in December, there is a significant increase of DO values which are retained in elevated levels during January and February when they reach their maximum (11.7 mg/L). Consequently, they are starting to drop and reach their minimum levels in May (6.7 mg/L). The annual median value of DO is 7.7 mg/L and is above the reference value of 5 mg/L for the optimal functioning of ecosystems (Behar 1997). TDS are progressively increasing from June to November when they reach a temporal peak of 243 mg/L; from December they decrease again till January when they reach their minimum value of 174 mg/L and consequently they increase again till May when they obtain their maximum (294 mg/L). The variations of TDS are similar with those of EC, a fact which confirms their affinity. Finally, pH values are alkaline and practically constant as they have only minor variance during annual records (7.8–8.2); water temperature follows the seasonal variations (4.2–26.7 °C) that affect air temperature without any significant deviations.

Based on the extended number of probe records for the above parameters, it was attempted to outline any potential relations between them with the aid of R-mode factor analysis. The results (Table 3) showed that the six parameters may be sufficiently interpreted by three factors. The first one (32.3 % of total variance) explains the affinity between DO and Eh which is related with the overall oxygen supply and the consequent aerobic or anaerobic conditions of the lake, while the high negative factor loading of temperature (−0.911) denotes that

water temperature is a limiting factor for the lake's oxygen levels. The second factor confirms the strong relation (high positive loadings) between EC and TDS. The negative factor loading of chl *a* (−0.411) may be explained if we consider that the elevated values of EC and TDS are possibly related with the karstic aquifer; hence during recharge events with fresh water from the karstic system, the physical process of dilution may favor the decrease of chl *a* values in the lake. Finally, the third factor embraces with antithetic loadings the parameters of pH and Eh and explains the significant impact of redox conditions to Micro Prespa's hydrogeochemistry.

Regarding the analyzed parameters of the four sampling campaigns, only 21 (Table 4) out of the 79 total parameters appeared to have significant concentrations above detection limits; the rest were either below them

Table 3 Sorted rotated factor loadings and communalities for the parameters recorded by the multi-sensor probe. Factor loadings below 0.400 are shown as 0 for simplification

Variable	Factor 1	Factor 2	Factor 3	Communality
Water temp (°C)	−0.911	0.000	0.000	0.845
DO (mg/L)	0.865	0.000	0.000	0.818
Eh (mV)	0.700	0.000	0.594	0.854
EC (μS/cm)	0.000	0.982	0.000	0.966
TDS (mg/L)	0.000	0.982	0.000	0.966
Chl <i>a</i> (μg/L)	0.000	−0.464	0.000	0.348
pH	0.000	0.000	−0.907	0.894
Variance	2.262	2.172	1.257	5.690
%Var	32.3	31.0	18.0	81.3

Table 4 Annual median values and ranges (in brackets) for the selected parameters that influence hydrogeochemistry of groundwaters samples

	Alluvial aquifer					Karstic aquifer				
	PRG1	PRG5	PRG6	PRG2	PRG3	PRG4	PRG7			
pH	7.7 (7.6–8)	8 (7.7–8.2)	7.4 (7.3–7.7)	7.2 (7–7.3)	7.2 (7–7.5)	6.9 (6.7–7.3)	7.1 (6.9–7.1)			
Eh	–56.1 (–53.5/–57)	–67.5 (–54.1/–83.9)	–38.5 (–37/–41)	–20 (–12.3/–33.8)	–22.3 (–11.5/–43.7)	–4.4 (–0.3/–32.6)	–11.9 (–3.8/–22.9)			
EC	302 (293–311)	253 (251–260)	163 (159–164)	660 (546–681)	705 (460–731)	645 (619–650)	618 (615–659)			
Na	11.9 (11.3–15.6)	14.4 (13.6–15)	7.9 (7.4–8.2)	2.1 (2–4.3)	3.7 (3.1–4.3)	2.4 (2.2–2.4)	1.2 (1.2–2.4)			
Ca	37.4 (34.4–38.5)	27.6 (24.4–30)	18.3 (16.8–20.2)	141 (111–146)	151.8 (88–164)	146.8 (139–149)	147.2 (128–156)			
Mg	12.8 (12.4–13.8)	10.8 (10.3–11.8)	6 (5.7–6.7)	10.8 (10.5–11.5)	6.5 (6.1–11.7)	4.5 (3.9–4.9)	5.2 (4.9–5.8)			
K	3.4 (2.3–8.2)	1.7 (1.6–2)	1.3 (1.3–1.6)	0.7 (0.4–1.6)	0.6 (0.6–1.4)	1 (0.9–1.3)	0.5 (0.5–0.6)			
Cl	3.3 (2.8–8)	2.7 (2–3.6)	2.6 (2–3–5)	2.9 (2–4.3)	8.7 (4.9–11)	3.8 (3.3–5)	2.1 (2.1–5)			
HCO ₃	170 (166–176)	147 (140–149)	76 (63–85)	438 (421–464)	440 (277–466)	415 (391–432)	437 (418–442)			
NO ₃	3.3 (3–3.7)	3.2 (2.7–3.3)	8.5 (7.1–8.6)	3 (2.3–3.3)	13 (5.7–16.5)	9 (6.4–12.4)	2.2 (1.6–3.1)			
NH ₄	0.2 (0–0.6)	0.1 (0–0.2)	0.5 (0.1–0.7)	0.2 (0–0.5)	0.4 (0–0.7)	0.1 (0–0.4)	0.1 (0–0.2)			
Al	86 (38–534)	38 (30–42)	48 (20–198)	25 (7–97)	40 (9–41)	36 (12–45)	16 (13–396)			
Cu	4 (3–21)	2 (1–5)	3 (2–6)	5 (4–514)	6 (2–7)	2 (2–24)	3 (1–318)			
Fe	68 (63–182)	52 (27–75)	50 (27–75)	52 (5–80)	25 (5–117)	5 (5–56)	5 (5–1309)			
Mn	2 (1–4)	3 (1–16)	4 (1–10)	3 (<1–14)	6 (1–8)	1 (1–3)	3 (<1–35)			
Ni	8 (3–117)	6 (<1–30)	8 (<1–44)	19 (6–47)	27 (<1–73)	5 (<1–49)	7 (<1–12)			
P	22 (13–25)	23 (14–29)	49 (28–53)	19 (12–33)	16 (5–24)	25 (5–29)	19 (3–42)			
Pb	2 (1–2)	3 (1–3)	3 (1–17)	4 (2–108)	4 (<1–8)	4 (2–8)	1 (<1–4)			
Si	12.8 (11.8–13.5)	15.2 (6.2–15.7)	13.9 (13.5–15)	3.9 (1.9–6)	3 (1.8–3.3)	4.8 (4.1–5.1)	2.4 (2.1–2.5)			
U	10 (10–11)	9 (4–9)	3 (2–5)	1 (<1–3)	<1	<1	<1			
Zn	19 (21–74)	14 (12–48)	11 (7–49)	32 (26–1854)	22 (17–80)	28 (7–48)	18 (10–40)			

(<0.5 µg/L for the majority of the parameters) or showed variations of minor importance close to detection limits and/or the analytical error, so they were practically ignored from further data processing as having negligible impact to water systems' hydrogeochemistry. Individual analytical results were checked for quality control through their ionic balance ratio (Appelo and Postma 2005) and were found to be below 10 % for all samples. The outlines of the analytical results of surface and groundwater samples are shown in Tables 4 and 5.

The analytical results of Micro Prespa's samples showed that pH is alkaline (8.2–8.3) with minor variations during the year, while redox conditions remain anoxic ranging from –86.8 to –89.8 mV. The latter is probably the result of oxygen depletion due to the existence of the abundant organic material in lake sediments which is oxidized and favors the creation of reductive conditions. Major ions (Na, K, Ca, Mg, Cl, HCO₃) fall within the range of natural reservoirs (Hem 1985) and are relatively low compared with groundwater values. Nitrate (NO₃) concentrations are almost negligible (0.8 to 1.1 mg/L), a fact that denotes that no pollution occurs due to the use of N-fertilizers. This assessment is rather important if we consider that a critical balance has been established between the sensitive ecosystem of Micro Prespa Lake and the crop cultivations which are fundamental for Prespa's financial prosperity. On the contrary, ammonium (NH₄) concentration values are slightly elevated (0.3 to 0.6 mg/L) reaching up to 1.2 mg/L seasonally. High NH₄ values should be attributed to the relative high organic matter of the lake's sediment that stimulates the decomposition processes and therefore mobilize nutrients, including inorganic nitrogen (Smolders et al. 2006). Even though this process releases both NO₃ and NH₄, the prevailing anoxic conditions favor the domination of ammonium instead of nitrates, as the latter is progressively reduced. Heavy metals concentrations are in general low except from nickel (Ni) which periodically (mainly in April's sampling) has elevated values reaching up to 116 µg/L at PRS1. At this specific sampling period (April 2012), all samples including Micro Prespa's had their highest values for Ni, hence special attention is needed. If we exclude the fact of laboratory systematic contamination and/or a random unknown event that triggered Ni concentrations, we are led to the conclusion that these peaks should be attributed to periodic changes in the hydrogeological regime (e.g., piezometric level, flow

direction) of the area which involves the hydraulic connection of Micro Prespa with the leached Ni-rich lateritic horizons of the substrate.

The drainage network of Prespa basin has nearly neutral pH values and Eh close to zero denoting well-oxygenated waters; major ions concentrations are even lower than those of Micro Prespa's. Nitrates are slightly higher (up to 9 mg/L for PRS5) but they do not justify any potential pollution from external sources (N-fertilizers, manure, or domestic wastes) as their ranges are within the natural background of pristine waters (Hem 1985). Ammonium values are low, since the triggering factor that stimulates NH₄ (rich organic content and anoxic conditions) release is absent. Heavy metals concentrations are generally low except manganese (Mn) (PRS6—25 µg/L, PRS8—32 µg/L) and iron (Fe) which is rather enriched in stream water compared with the rest of the samples, reaching up to 469 µg/L (PRS8). Phosphorus is also enriched in streams ranging from 60 to 167 µg/L; its presence should be attributed to natural origin and not to anthropogenic effects related with phosphate fertilizers, manure, and/or domestic wastes (e.g., sewage or septic tanks), as the above factors cannot be documented. Such hypotheses would require the spatial correlation of cultivated plots with the stream sampling sites, validation by macroscopic field surveys, and the presence of elevated nitrate values either as a result of the fertilization process or as a basic constitute of domestic effluents.

On the contrary, the natural enrichment of phosphorus is a possible scenario which may potentially derive from two different sources related with geology; phosphorus has been reported as content in granitic rocks (London et al. 1990; London 1992) and associated with elevated Fe values as a result of supergene alteration of secondary Fe-rich phosphate minerals by percolating meteoric waters (Llorens and Moro 2012). Taking into account that the eastern side of Micro Prespa basin is formed by similar geological formations and that Fe in stream samples has a strong positive correlation with P (Pearson correlation coefficient +0.89) we may assume the soundness of the above hypothesis for the P enrichment process. Additionally, in nearby Neogene basins of Western Macedonia (Florina, Ptolemaida, Amyndeo) that contained lignitic ores, the presence of iron-phosphates in the form of vivianite has been reported (Stamatakis 2004). Such a hypothesis may additionally justify the phosphorus and iron enrichment in stream water samples, as a result of the leaching of lignitic

Table 5 Annual median values and ranges (in brackets) for the selected parameters that influence hydrogeochemistry of surface water samples

	Micro Prespa Lake					Drainage network				
	PRS1	PRS2	PRS3	PRS4	PRS5	PRS6	PRS7	PRS8	PRS8	
pH	8.3 (8.1–8.4)	8.2 (8.2–8.3)	8.3 (8.1–8.3)	8.3 (8.2–8.5)	7.5 (7.4–7.6)	7.6 (7.4–7.7)	7.4 (7.2–7.6)	7	7	
Elh	-88.3(-61.6/-97.5)	-86.8(-61.6/-93.7)	-89.1(-58.1/-94.7)	-89.8(-86.1/-100.9)	-34.5(-30.2/-38.7)	-38.2(-37.9/-38.5)	-27.2(-25.5/-28.8)	-16.1	-16.1	
EC	262 (248–291)	271 (250–293)	264 (245–290)	258 (238–288)	181(180–181)	93.1 (88–98.1)	72.6 (72–73.2)	58	58	
Na	4.2 (4–6.4)	4.2 (4–4.4)	4.3 (4.1–4.6)	4.4 (4–4.8)	9.5 (9–9.9)	5.6 (5.5–5.7)	3.8 (3.6–3.9)	3.6	3.6	
Ca	42.9 (30.9–53.6)	38.6 (32.7–44.4)	38.5 (32.8–43.7)	39.2 (31.4–44.7)	19.7 (17.2–22.2)	12.2 (9–15.5)	10.8 (7.5–14.1)	7.7	7.7	
Mg	12.9 (12.5–22.2)	12.9 (12.6–15.2)	12.7 (12.2–14.8)	13.1 (12.9–15.2)	5.9 (5.8–5.8)	3 (2.9–3.1)	2.4 (2.3–2.4)	1.7	1.7	
K	1.9 (1.5–3)	1.9 (1.9–2.1)	1.9 (1.6–2.1)	2 (1.9–2.2)	1.3 (1.1–1.5)	1.8 (1.7–1.9)	1.3 (1.1–1.4)	1.6	1.6	
Cl	3.5 (2–4.2)	3.8 (3–4.3)	3.7 (3–4.6)	3.8 (3.6–3.8)	10.9 (9–12)	1.3 (<1–2.6)	1.1 (<1–2.2)	0.5	0.5	
HCO ₃	164 (151–188)	165 (159–195)	161 (150–177)	156 (151–178)	70 (69–72)	57 (44–73)	40 (31–49)	25	25	
NO ₃	1.1 (<1–1.4)	<1(<1–1.5)	1 (<1–2.9)	<1 (<1–1.2)	9.2 (7.3–11.2)	1.4 (1.2–1.6)	3.3 (2.3–4.3)	1.2	1.2	
NH ₄	0.4 (<0.1–1)	0.4 (0.3–1.2)	0.6 (0.4–0.7)	0.3 (0.1–0.3)	0.2 (<0.1–0.3)	0.1 (<0.1–0.1)	0.1 (<0.1–1)	0.1	0.1	
Al	54 (9–134)	19 (13–33)	72 (16–116)	16 (14–20)	33 (14–52)	107 (55–119)	119 (45–193)	268	268	
Cu	3 (<1–14)	2 (<1–3)	2 (1–6)	2 (1–4)	2 (1–2)	2 (2–3)	3 (2–3)	2	2	
Fe	110 (52–150)	67 (37–84)	86 (36–130)	77 (45–77)	41 (34–48)	231 (157–305)	211 (122–299)	469	469	
Mn	19 (11–56)	31 (9–40)	16 (7–49)	25 (20–34)	9 (6–13)	25 (16–34)	11 (10–11)	32	32	
Ni	4 (<1–116)	14 (<1–49)	2 (1–41)	3 (<1–72)	11 (1–20)	2 (1–2)	12 (2–21)	1	1	
P	28 (19–32)	18 (5–24)	28 (17–42)	28 (8–34)	64 (43–84)	76 (72–81)	60 (58–62)	167	167	
Pb	2 (1–10)	2 (<1–6)	1 (<1–4)	2 (1–4)	1 (<1–1)	2 (1–4)	2 (1–3)	1	1	
Si	1.4 (0.3–1.8)	0.5 (0.1–1.3)	1.2 (0.3–6.8)	0.3 (0.1–0.8)	7.5 (4.6–10.4)	10.3 (9.1–11.5)	7.1 (7.4–8.1)	6.6	6.6	
U	1 (<1–1)	1 (<1–1)	1 (<1–1)	1 (<1–1)	2 (2–3)	<1	<1	<1	<1	
Zn	10 (8–34)	9 (7–56)	7 (8–33)	22 (15–45)	16 (16–28)	17 (17–19)	10 (10–34)	17	17	

groundwater systems. Nitrogen compounds (NO_3 and NH_4) have low values, except sample PRG6 which has an annual median concentration for NH_4 that lie on the verge of maximum parametric limit (0.5 mg/L) for potable waters (EC 1998).

Nickel is elevated in PRG3 (annual median value is 27 $\mu\text{g/L}$) but shows seasonal peaks, respectively, for PRG1 (117 $\mu\text{g/L}$), PRG2 (27 and 47 $\mu\text{g/L}$), PRG3 (73 and 48 $\mu\text{g/L}$), PRG4 (49 $\mu\text{g/L}$), PRG5 (30 $\mu\text{g/L}$), and PRG6 (44 $\mu\text{g/L}$). Its existence is related with the Ni-rich geological formations of the substrate that include (i) the lateritic horizons and (ii) the serpentinized blocks of ophiolites along with schists and other metamorphic rocks (tectonometamorphic *mélange*); similar formations in other Greek areas are reported to be rich in Ni, Cr, Fe, and Co (Skarpelis 2005). The occurrence of Ni sources is well documented and described by the mining activities in Greece (Hieropigi area—South from Prespa) and Albania (Devol and Korce areas) according to Apostolikas (2010) and Shallari et al. (1998)

The enrichment process involves the progressively leaching of these formations which may provide spatially distributed (diffuse) Ni sources for groundwater, like similarly reported for other Greek areas (Tziritis 2009; Tziritis et al. 2012). Key factors that control the process are: the overall hydrogeological regime and its relevant temporal changes (flow direction, piezometric level, etc), mineralogy of the strata and homogeneity of the horizons, as well as the recharge conditions. A simplified schematic geological section that shows the general geological-hydrogeological setting of the water bodies beneath Micro Prespa which affect Ni enrichment is shown in Fig. 5.

Seasonal high peaks for aluminum (Al) concentrations (PRG1, 534 $\mu\text{g/L}$) are temporal and related with dissolution of aluminosilicate minerals or external undefined events, like the one recorded for PRG7 during October's sampling, when several outliers were detected including Al (396 $\mu\text{g/L}$), Cu (318 $\mu\text{g/L}$), and Fe (1,309 $\mu\text{g/L}$); nevertheless, the lack of periodicity for these values renders no explicit environmental risk and suggests that these events are likely to be random rather than systematic. Similarly, a high concentration of Pb (108 $\mu\text{g/L}$) was recorded for PRG2 during July's sampling along with elevated values for Cu (514 $\mu\text{g/L}$) and Zn (1854 $\mu\text{g/L}$). The source of Pb enrichment is yet unclear and could not be related directly with natural or anthropogenic factors. Previous researches (Schoullou and Hatzianestis 1989) reported relative enrichment in metals (including Pb) at

the southern part of the lake (surface waters) near the borders with Albania. Nevertheless, no sufficient evidence was provided to justify human impact, but this should not be excluded. Other researchers (Krstic 2012) support that on the Skopjan side of Prespa Lake there are several medium-size enterprises from industrial branches, including food processing, poultry farming, textile, metal processing, wood processing, and chemical industry. Some of the above enterprises may potentially be a source of Pb, especially textile industries through alkalic lead carbonate ($2\text{PbCO}_3 \cdot \text{Pb}(\text{OH})_2$), which was used in the past (prohibited now in EU) from similar industries as white pigment. Krstic (2012) also reported the detection of heavy metals (Pb, Cu, Zn, Mn) in significant quantities in the watershed of Macro Prespa Lake (Rivers Istočka and Golema), but their exact origin is not documented. Finally, phosphorus is slightly enriched in the samples of alluvial aquifer in respect to the karstic ones, probably as a result of the relative enrichment that has been already described to take place in the eastern side of the basin, while uranium (U) concentrations of PRG1 (10 $\mu\text{g/L}$) and PRG5 (9 $\mu\text{g/L}$) are attributed to the geogenic enrichment of granites.

The potential relationships between samples were documented with the use of a hierarchical cluster analysis; samples were categorized into clusters according to their hydrogeochemical signature which stemmed from their annual median values. As seen in Fig. 6, the distinguished individual clusters were similar with the monitored water systems as described initially. From the relative dendrogram, it is clear that the hydrogeochemistry of Macro Prespa's samples is influenced both by the karstic and the alluvial aquifer (equal Euclidean distance). Special attention should be given to the fact that the drainage network (PRS6–PRS8) is not interacting with the alluvial aquifer (apart from PRS5) although it crosses the plain parts where the latter is located; hence, streams are not likely to interact with the alluvial aquifer through vertical crossflows but by the seasonal runoff of the mountainous areas on the east.

Conclusions

Micro Prespa Lake and its relative hydrological basin is a rather sensitive ecosystem with global significance that harbors important endemic floral and faunal species. Despite its importance, the integrated water quality monitoring of Micro Prespa's interacting water systems

has never been performed, hence rendering water resources management efforts incomplete. In that frame, an integrated environmental monitoring scheme was developed and delivered so as to provide an in-depth and updated insight to the overall water quality characteristics by coupling different monitoring methods (real-time records, in situ measurements, laboratory analyses and macroscopic investigations) and a wide range (85) of diverse environmental parameters.

Results identified that Lake Micro Prespa is in good quality condition and classified as oligotrophic to slightly mesotrophic according to its chl *a* values; the higher values are recorded during winter and early spring while the lower ones during summer. Nitrates are practically negligible reaching up to 1.1 mg/L. On the contrary, ammonium values are slightly elevated reaching up to 1.2 mg/L seasonally, and are attributed to the relative high organic matter of the lake's sediment that stimulates the decomposition processes and therefore mobilize nutrients.

Phosphorus is the limiting factor for the overall nutrient load as indicated by N/P ratio. Phosphorus variations follow those of chlorophyll *a* during the hydrological year; the main factor of their co-variance is attributed to the redox changes in lake sediments that trigger P release and causes an increment in chlorophyll *a* accordingly. Apart from the internal sources (lake sediments), phosphorus loads should be attributed to other external sources like phosphate deposits of the substrate.

In respect to relevant legislation for surface and groundwaters (EC 2000), none of the parameters' annual median concentrations exceeded the maximum parametric values imposed. However, seasonal peaks regarding mainly Ni, Pb, and NH₄ were documented and these parameters should be closely monitored in the future. The origin of these peaks are rather natural than anthropogenic. The elevated Ni values should be attributed to leaching of Ni-rich geological formations favored by seasonal variations of the hydrogeological regime. Lead's origin is still unclear, but possibly related with pollution loads deriving from Macro Prespa, while NH₄ is likely to derive as result of the organic rich sediments around the lake. Concretely, the overall quality conditions of interacting water systems in Micro Prespa basin are good. Nevertheless, continuous common inter-scientific and multi-disciplinary efforts are required by stakeholders, as well as public awareness of local communities in order to manage sustainably this dynamic aquatic ecosystem.

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