RESEARCH ARTICLE



Evaluating toxic element contamination sources in groundwater bodies of two Mediterranean sites

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Abstract

The Driver–Pressure–State–Impact–Response (DPSIR) framework is applied for assessing the pressures and impacts on groundwater bodies of two Mediterranean sites (Megara and Oropos–Kalamos basins). The study areas present joint driving forces (drivers) and pressures. The main driving forces in the areas studied mainly include geology, agricultural activities, and urban development, while the main pressures mainly include the weathering of ultramafic rock masses, application of agrochemicals, and groundwater abstractions for irrigation and drinking uses. Hexavalent chromium (Cr⁺⁶), chromium total (Cr_{total}), manganese (Mn), and nitrate (NO₃⁻) contamination of groundwater bodies are attributed to both anthropogenic and lithological sources. Elevated Cr_{total} (up to 70.3 μ g L⁻¹), Mn (up to 87.7 μ g L⁻¹), and NO₃⁻ (up to 411 mg L⁻¹) contents are recorded for groundwater samples in Megara basin. High Cr_{total} (up to 34.3 μ g L⁻¹), Cr⁶⁺ (up to 27.9 μ g L⁻¹), Mn (up to 132.5 μ g L⁻¹), and NO₃⁻ (up to 30 mg L⁻¹) are also observed for groundwater samples in the Oropos–Kalamos basin. The major response actions needed for the management options of groundwater bodies are discussed. Among the proposed remedial measures, the installation of a continuous groundwater monitoring network and the control in the usage of nitrogen fertilizers seems to be the most effective and tangible for immediate action.

Keywords DPSIR · Chromium · Nitrates · Ultramafic rocks · Groundwater quality · Megara · Oropos-Kalamos

Introduction

European countries should achieve water quality objectives as introduced by the 2000/60/EC Water Framework Directive

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(WFD; EC 2000). Regarding groundwater, the lithology of the aquifer, geochemical processes that take place within the aquifer, climate change, and various land uses are the main factors that influence its quality (Alexakis 2020; Alexakis and Gamvroula 2014; Golfinopoulos et al. 2021; Li et al. 2020; Vaiphei et al. 2020). Its adequacy for a number of uses (agriculture, households, industry) is defined by a number of criteria set in legislation. Applying low-quality water for irrigation results in the reduction of crop productivity, while human consumption of this water can lead to various health problems (Varnavas et al. 2012). Groundwater quality is often degraded by soil salinization (Alexakis et al. 2012), weathering of naturally enriched parent rocks (Alexakis and Gamvroula 2014; Alexakis 2016; Le Luu 2019), decrease of precipitation and sea level rise (Alexakis and Tsakiris 2010), municipal sludge leachates (Emmanouil et al. 2020; Giannakis et al. 2020), agricultural activities (Giakoumakis et al. 2013; Gotsis et al. 2015; Kungolos et al. 2006), and mining activities and metal processing (Alexakis 2011; Chandio et al. 2020). Detailed information on water quality is a crucial issue for the sustainable management of water resources. However, the real challenge for achieving the

sustainable development goals which are related to water quality issues as introduced by Agenda 2030 (UN 2020) is how to promote socioeconomic development that balances both environmental protection and economic growth.

In this context, the present research focuses on developing a DPSIR framework and applies this framework for groundwater contamination in two basins of the Attica region (Greece). The main objectives of this study are to identify and estimate driving forces and pressures on the basins; and to propose response actions for improving the chemical status of the groundwater bodies.

Materials and methods

Study area

Megara basin $(37^{\circ} 57'-38^{\circ} 08' \text{ N}, 23^{\circ} 10'-23^{\circ} 27' \text{ E})$ and Oropos–Kalamos $(38^{\circ} 14'-38^{\circ} 20' \text{ N}, 23^{\circ} 44'-23^{\circ} 55' \text{ E})$ basins are located in western part (approximately 41 km western from Athens) and in the northeastern part of the Attica region (approximately 50 km north of Athens), respectively (Fig. 1).

In the Megara basin, an aquifer with low hydraulic characteristics was developed in the Quaternary alluvial deposits. The crystalline basement rock outcrops in the Megara basin are particularly impermeable rocks. The intensively karstified limestones–dolomites of M.–U.Triassic–L.Jurassic are permeable rocks that constitute the most important aquifer of the Megara basin. The infiltration water of the karstified limestones–dolomites moves laterally to the surrounding Neogene and Quaternary deposits. The Neogene and Quaternary deposits are rich in weathered ophiolite material, while the presence of manganese (Mn) oxides is recorded (Gaitanakis et al. 1984; Gamvroula 2013).

In the Oropos–Kalamos basin, the most important aquifers were developed in the Triassic–Jurassic limestones, Upper Cretaceous limestones, and marly limestones and travertines. The Upper Miocene conglomerates form a poor aquifer. The marly formations with lignite intercalations also form a poor aquifer. The Quaternary alluvial deposits form a phreatic aquifer which possesses low hydraulic characteristics (Voreadis 1952; Gamvroula 2009).

For this study, the updated Köppen–Geiger climate classification is adopted. Based on this classification, the two studied basins belong to the Csa type which is temperate–dry–hot– summer climate, where at least 1 month's average temperature is above 22 °C, the average temperature of the coldest month ranges from 0 to 8 °C, and the temperature of the hottest month is higher than 10 °C (Peel et al. 2007). According to Peel et al. (2007), the precipitation of the driest month in summer is less than the 33% of the rainfall in the wettest



Fig. 1 Map showing the study areas and groundwater monitoring sites

month of the winter and below 40 mm. It should be noted that the hydrological year in the temperate climate type consists of a dry (April through September) and a wet (October to March) period.

The land in Megara and Oropos–Kalamos basins is characterized by intensive agricultural activities, especially vegetables, olive trees, and vineyards. Moreover, extensive lignite mining activities have occurred in the southern part of the Oropos–Kalamos basin from 1870 to 1960. A significant amount of waste which was produced from past mining activities is deposited in piles around the mining areas of the Oropos–Kalamos basin. No other industrial activities are observed either in the Megara or Oropos–Kalamos basins.

Data collection and available data

The monitoring network was designed in the Megara and Oropos-Kalamos basins for representative sampling of groundwater of the aquifers: (a) the shallow alluvial aquifer of Quaternary deposits; (b) the deep aquifer hosted in permeable layers of Neogene deposits; and (c) the deep aquifer hosted in intensively karstified carbonate formations. The sampling procedure is described by Stamatis et al. (2011) and Gamvroula et al. (2013). In brief, water samples were stored into new polypropylene containers, rinsed several times with groundwater sample water prior to sample collection. Each water sample was then divided into two water subsamples. The first set of water subsamples was filtered with a 0.22-µm disposable syringe filter, acidified to pH < 2 with ultrapure HNO₃ and stored in a 100-mL container for trace element analysis. The second set of subsamples was stored in a 1000-mL container and transferred to the laboratory where after filtration through 0.45-µm pore-size membrane filters, major anions and cations were analyzed. The water samples were kept in a cooled plastic box at 4°C. Certified analytical standards (Thermo Scientific Dionex, USA) were used for the calibration of the instruments. More data on the methods used are shown in Table 1. The dataset of groundwater quality parameters was categorized into 9 sub-datasets: (a) Meg– Quat–Wet (N=14); (b) Meg–Quat–Dry (N=14); (c) Meg– Neog–Wet (N=31); (d) Meg–Neog–Dry (N=31); (e) Meg– Karst–Wet (N=5); (f) Meg–Karst–Dry (N=5); (g) OropKal– Quat–Dry (N=5); (h) OropKal–Neog–Dry (N=19); (i) OropKal–Neog–Dry (N=1) (key: Meg: Megara basin; OropKal: Oropos-Kalamos basin; Quat: Quatemary aquifer; Neog: Neogene aquifer; Karst: Karstic aquifer) (Table 2).

The DPSIR model

For investigating environmental issues which are associated with the impacts caused by human activities, the Driver-Pressure-State-Impact-Response (DPSIR) is widely applied in the framework of sustainable water resource management (Alexakis et al. 2013; Bagordo et al. 2016; Kristensen 2004). The relationship between human activity and environmental degradation in the two basins was assessed by applying the DPSIR approach, which was developed by OECD (1993). The DPSIR framework can be summarized as follows (Alexakis et al. 2013; OECD 1993; Kristensen 2004; Pirrone et al. 2005): (a) Drivers or driving forces are anthropogenic activities and natural processes which are able to cause pressures on the water system; i.e., geology, agriculture, and landuse change; (b) Pressures are directly stressed which are derived from the anthropogenic and natural system and affect the ecosystems, i.e., release of pollutants; (c) State describes the environmental conditions of natural systems (water, sediment, and soil quality); (d) Impact is the measure of the effects which are attributed to changes in the state of the environmental system; and (e) Response is the evaluation of actions oriented to propose a remedial strategy.

Pathways in the DPSIR model are presented in Fig. 2. Data (hydrogeological, hydrogeochemical, land use, etc.) of the

 Table 1
 Analytical methods and instrumentation applied in this study

Parameter	Analytical method and instrumentation					
Conductivity (CND), dissolved oxygen (DO), total dissolved solids (TDS), pH	Measured immediately after collection with a YSI Professional Plus portable meter					
Cl ⁻ , NO ₃ ⁻ , PO ₄ ³⁻ , SO ₄ ²⁻ , Ca ²⁺ , K ⁺ , Mg ²⁺ , Na ⁺ , NH ₄ ⁺	Ion chromatography (Dionex ICS-3000 system) on the non-acidified subsamples					
HCO ₃ ⁻	HACH digital titrator on the non-acidified water subsamples					
Cd, Co, Cr _{total} , Cu, Mn, Ni, Pb, Zn	Inductively coupled plasma mass spectrometry (ICP-MS) Agilent model 7700 MassHunter on the acidified subsamples					
Cr ⁶⁺	Cation exchange Cr speciation method, as demonstrated by Ball and Izbicki (2004). Atomic absorption spectroscopy (AAS/Perkin Elmer 1100B) with a graphite furnace (Stamatis et al. (2011) applied for the first time this analytical method in Greece)					
As, U, V	Inductively coupled plasma mass spectrometry (ICP-MS) at ACME Analytical Laboratories Ltd. on the acidified subsamples					

 Table 2
 The arithmetic mean value of parameters in groundwater from study areas (n.d.: not determined; P.V.: parametric value given by the Dir. 98/83/EC (EC 1998); CND: conductivity)

			Megara basin ⁽¹⁾							Oropos-Kalamos basin ⁽²⁾		
			Quaternary aquifer		Neogene aquifer		Karstic aquifer		Quaternary aquifer	Neogene aquifer	Karstic aquifer	
	Units	P.V.	Wet period	Dry period	Wet period	Dry period	Wet period	Dry period	Dry period	Dry period	Dry period	
As	$\mu g L^{-1}$	10	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	14	26	20	
Ca ²⁺	mg L^{-1}	-	140	226	52	53	113	277	105	133	241	
Cd	$\mu g L^{-1}$	5	0.03	n.d.	0.03	0.01	0.08	n.d.	0.03	0.03	0.03	
Cl	mg L^{-1}	250	890	1673	107	105	1868	2241	162	119	2907	
Со	$\mu g L^{-1}$	-	0	1	0	1	0	0	0	0	0	
Cr total	$\mu g L^{-1}$	50	8	15	29	30	13	48	5	5	2	
Cr ⁶⁺	$\mu g L^{-1}$	-	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	4	3	1	
Cu	$\mu g L^{-1}$	2000	1	2	2	3	1	0	3	3	3.1	
HCO ₃ -	mg L^{-1}	-	394	418	385	383	323	389	421	439	457.5	
K ⁺	mg L^{-1}	-	10	12	4	4	35	44	3	2	58.6	
Mg ²⁺	mg L^{-1}	-	139	246	62	58	141	172	52	47	157.8	
Mn	$\mu g L^{-1}$	50	4	3	9	9	0	0	18	11	0	
Na ⁺	mg L^{-1}	200	421	750	61	65	940	1217	62	44	1482	
NH4 ⁺	mg L^{-1}	0,5	0	0	0	0	0	0	1	0	0.03	
Ni	$\mu g L^{-1}$	20	4	8	2	3	1	0	3	12	1	
NO ₃ -	${ m mg}~{ m L}^{-1}$	50	108	133	15	10	22	23	11	12	5.28	
02	mg L^{-1}	-	8	8	10	10	9	9	5	10	n.d.	
Pb	$\mu g L^{-1}$	10	0	0	0	0	0	0	14	1	3.4	
PO4 ³⁻	${ m mg}~{ m L}^{-1}$	-	0	33	0	1	1	0	0	0	0.08	
SO_4^{2-}	${ m mg}~{ m L}^{-1}$	250	187	310	39	48	221	255	39	74	377.5	
Zn	$\mu g L^{-1}$	-	17	9	70	116	6	0	42	110	9.10	
TDS	mg L^{-1}	-	2612	3392	735	729	3533	4333	527	534	450	
pН		6.5–9.5	8	8	8	8	8	7	7	7	7.33	
CND	$\mu S \ cm^{-1}$	2500	3561	4795	1000	997	5738	6033	958	943	882	
U	$\mu g L^{-1}$	-	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2	2	1	
V	$\mu g \; L^{-1}$	-	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	2	2	19	

¹ Gamvroula (2013)

 2 Gamvroula (2009)

two basins (Megara and Oropos–Kalamos) were collected from literature. Physical and chemical analysis data were acquired from previous work (Gamvroula 2009, 2013; Stamatis et al. 2011; Gamvroula et al. 2013).

Results and discussion

Identification and estimation of driving forces and pressures

Identification and estimation of driving forces and pressures in many other research fields have been successfully performed by the usage of the DPSIR approach. Wei et al. (2019) proposed an effective energy efficiency evaluation index system which is based on the DPSIR framework. The DPSIR approach was used by many researchers to identify the critical natural and anthropogenic factors controlling the quantity and quality status of water resources (Alexakis et al. 2013; Pirrone et al. 2005; Bagordo et al. 2016).

In this study, the assessment of pressures and impacts on groundwater resources for the Megara and Oropos–Kalamos basins was performed using the systemic DPSIR model (Fig. 3).

Geology, agricultural activities, livestock, households, and past mining activities are the main driving forces recorded in the basins studied. The population of the Megara and Oropos– Kalamos basins is 35,675 and 19,556 inhabitants, respectively. The leakage of wastewater from household sewerage





systems installed in the study areas contributes to the increase of NO₃⁻, NH₄⁺, NO₂⁻, and Cl⁻. The Megara basin is one of the most productive agricultural regions of Greece. The irrigated area is approximately 2000 and 16,000 ha for the Megara and Oropos-Kalamos basins, respectively. According to Gamvroula et al. (2013) and Stamatis et al. (2011), the over-abstraction of groundwater in the Megara and Oropos-Kalamos basins include decreases in the aquifer level that in turn lead to seawater intrusion. A large part of the Megara and Oropos-Kalamos areas is used for agriculture. The water demand in the Megara basin for irrigation/livestock, drinking, and other uses (industrial, firefighting, irrigation of urban areas etc) is 89%, 7%, and 4%, respectively. Irrigation water is the dominant demand in total water consumption in both areas. Livestock activity represents an important sector in the local economy of the Megara basin. The animal farming in the Megara basin include the following: (a) poultry farming accounting for 3.09% (938,986 poultries) of the national poultry production; (b) sheep farming which accounts for 0.26% (21,737 sheep) of the national sheep production; (c) goat farming accounting for 0.22% (7741 goats) of the national goats production; and (d) bovine farming which accounts for 0.13% (833 animals) of the national bovine production.

Animal breeding activity mainly release pollutants such as organic load, nitrogen and phosphorous. The exerted pressures from livestock are the following (YPEKA 2012): (a) poultry 19, 4, and 0.3 kg/ha/year for organic load, TN, and TP, respectively; (b) sheep/goats 6, 3.8, and 0.2 kg/ha/year for organic load, TN, and TP, respectively; and (c) bovine 3, 1, and 0.05 kg/ha/year for organic load, TN, and TP, respectively.

Nitrate (NO₃⁻) release (Pressures) caused by agricultural activities (Driving forces) in the studied basins reach the groundwater bodies through infiltration (Fig. 3). Nitrate ion emissions from diffuse sources of the study area are related to selected socioeconomic factors such as agricultural

production and livestock. Pirrone et al. (2005) reported that the investigation of variation of nutrient fluxes requires the analysis of socioeconomic situation of the area studied during a long period. The key to understanding the complex geochemistry of Cr in groundwater bodies of the Megara and Oropos-Kalamos areas is the critical role of agricultural activities and the associated release of NO₃⁻ ions which play an important role in controlling Cr speciation. The lack of management practices to reduce pollutant sources and the high nitrogen fertilization intensity has led to the NO₃⁻ contamination of groundwater bodies of the Megara basin (Fig. 3). According to Stamatis et al. (2011), the NO_2^{-}/NO_3^{-} couple controls the Cr speciation in the Oropos-Kalamos basin. Furthermore, Stamatis et al. (2011) initially highlighted the redox transformation of Cr³⁺ into Cr⁶⁺, or vice versa, which takes place in the presence of NO₂^{-/NO₃⁻ derived from nitro-} gen fertilizers applied in cultivated areas. The results showed that the increasing drivers of socioeconomic development (agriculture/livestock) coupled with the presence of ultrabasic rocks have resulted in the degradation of water quality in cultivated areas. Similar findings have been also reported by various researchers (Al-Kalbani et al. 2016; Bagordo et al. 2016; Li et al. 2019) regarding the association of socioeconomic development and degradation of water quantity and quality. The past mining activities recorded in the Oropos-Kalamos basin are associated with the elevated As, U, and V contents in groundwater (Stamatis et al. 2011). According to Pirrone et al. (2005), the DPSIR framework applied into the Po catchment (Italy) provided a clear identification of Driving forces.

Evaluation of the state and analysis of impacts

Figure 4 shows the comparison between the available datasets (two datasets from the Stamatis et al. 2011, Gamvroula et al.



Fig. 3 The DPSIR methodology framework applied to the Megara and Oropos-Kalamos basins (¹only for Oropos-Kalamos basin; ²only for Megara basin).

2013 respectively and the third one from the current campaign) for the physical and chemical parameters studied. The mean values of Cl⁻, Na⁺, NO₃⁻, SO₄²⁻, and Conductivity recorded in groundwater of Megara basin exceed the corresponding parametric value (P.V.) established by the European Community (EC 2000). Higher mean contents of As, Cl, Na, NH₄⁺, Pb, and SO₄²⁻ than the European Community health-based drinking water guidelines (EC 2000) were also observed in groundwater of the Oropos–Kalamos basin.

In the Oropos–Kalamos basin, the release of As, B, Ca, Cl, Cr^{+6} , Cr_{total} , HCO_3^{-} , K, Mg, Mn, Na, Ni, Rb, S, Si, SO_4^{2-} , V, and U in groundwater (Pressures) caused by (Driving forces) leaching from (a) terrestrial salts; (b) volcanic sedimentary

formations; (c) lignite intercalations; and (d) ultrabasic rock masses and chromiferous iron ores) was also observed (Fig. 1).

Several studies have demonstrated an apparent impact of As, Mn, Ni, V, and other trace elements on human health and ecosystems (Alexakis 2016; Alexakis et al. 2019, Golfinopoulos et al. 2021; Kondakis et al. 1989; Varnavas et al. 2012; Varnavas 2014; Varnavas 2016). Extensive research has been carried out by Kondakis et al. (1989) and Varnavas (2016) in groundwater from the Lehaina area, Kourtessi–Varda–Areti area, and Andravida town; all these areas are situated in Eleias Prefecture (western Peloponnese, Greece), highlighting that Mn content in groundwater is well

Fig. 4 Concentrations of Cl, Cr_{total}, Mn, Ni, NO₃⁻, SO₄²⁻, Zn, and TDS in groundwater of the Megara and Oropos-Kalamos basins. The box-and-whisker plots demonstrate the outlier (*,°), maximum value (upper whisker), 75th quartile, median, 25th quartile, and minimum value (lower whisker) (Meg: Megara basin; OropKal: Oropos-Kalamos basin; Quat: Quaternary aquifer; Neog: Neogene aquifer; Karst: Karstic aquifer)



above the parametric value of 50 μ g L⁻¹ established by EC (1998). According to Kondakis et al. (1989), the Mn contents in drinking water were 3.6–14.6 μ g L⁻¹ (Lehaina area), 81.6–252.6 μ g L⁻¹ (Kourtessi-Varda-Areti area), and 1800–2300 μ g L⁻¹ (Andravida town), while the Mn contents in hair (of males and females) were 3.51, 4.49, and 10.99 μ g g⁻¹ dry weight, respectively. The means of Mn in groundwater of the

Megara basin (up to 9 μ g L⁻¹) and Oropos–Kalamos basin (up to 18 μ g L⁻¹) which are comparable with the Mn content in drinking water of Lehaina area, suggest that it is very important to conduct further research studies on the impact of Mn on human health of the inhabitants of the study area. Elevated As, U, and V contents in groundwater of the Oropos–Kalamos basin are associated with of marly formations, lignite

intercalations, marly limestones, and travertines (Gamvroula 2009). According to Alexakis and Gamvroula (2014), high content of As (up to 226.2 mg kg⁻¹), U (up to 8.8 mg kg⁻¹), and V (up to 225 mg kg⁻¹) in the Oropos-Kalamos lignite intercalations and mining waste result in the high concentration of As (up to 246.5 μ g L⁻¹), U (up to 14.73 μ g L⁻¹), and V (up to 19.4 μ g L⁻¹) in the Oropos–Kalamos groundwater. Elevated Cr_{total} and Cr⁺⁶ contents (Pressure) in groundwater of the Oropos-Kalamos basin are derived from the weathering products of ultrabasic rock masses (Driving forces) in the presence of the NO_2^{-}/NO_3^{-} redox couple, which originates from nitrogen fertilizers applied in cultivated areas of the Oropos-Kalamos basin. An additional source of NO₃⁻ in the study area is sludge derived from livestock, because sludge is in most cases directly distributed on cultivated areas. A similar finding is also reported by Pirrone et al. (2005) in the Po River basin which is the largest Italian catchment. Among the subdatasets, Meg-Karst-Dry, Meg-Karst-Wet, OropKal-Karst-Dry, and OropKal-Quat-Dry showed the lowest concentrations for most water quality parameters (Fig. 4).

Besides, values of parameters in groundwater bodies of areas studied were variable with Cr_{tot} being the highest in 6 sub-datasets, Ni being the highest values in 3 sub-datasets, and NO_3^- being the highest in 2 sub-datasets. Since there is the common origin (seawater intrusion) of CI^- , SO_4^{2-} , and TDS in groundwater of the areas studied, the observed value of these water quality parameters showed similar trends.

Proposed responses

After the identification of the pressures and the subsequent impacts, the next step is to propose responses concerning sustainable development practices. The proposed responses mainly include the monitoring of groundwater quality and quantity, measures to reduce non-point sources (agriculture), agricultural policies, and management plans for diffuse pollution loads and point pollution. A remedial strategy should also include the decrease in the applied amounts of nitrogen fertilizers in order to control the redox transformation of Cr^{3+} into Cr^{6+} , or vice versa, which takes place in the cultivated areas.

Conclusions

A DPSIR model was applied to explore the relationships between the socioeconomic system and groundwater resources of two basins in the Mediterranean region. Findings demonstrated that the effects of the drivers to groundwater resources of the two studied basins were increasing while forging ahead with socioeconomic factors (agricultural activities and households). The DPSIR approach proved to be a useful and powerful tool for performing a qualitative assessment of groundwater resource sustainability levels. The synergistic effect of two driving forces in cultivated areas, namely, the application of nitrogen fertilizers and the presence of weathering products of ultrabasic rocks, influences the redox transformation of Cr^{3+} into Cr^{6+} or vice versa and finally controls the concentration of Cr^{6+} in groundwater. Therefore, the restriction of the extensive application of nitrogen fertilizer is expected to decrease the Cr^{6+} content in groundwater and subsequent to reduce the human health risk. Among the proposed remedial measures, the installation of a continuous groundwater monitoring network and the control in the usage of nitrogen fertilizers seem to be the most effective and tangible for immediate action. It is expected that the systematic application of these actions will improve the chemical status of the groundwater bodies in the areas studied.

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Availability of data and materials The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethics approval, consent to participate, and consent for publication Not applicable.

Conflict of interest The authors declare that they have no conflict of interest.

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