Multi-criteria Assessment of Heavy Metals contaminations in waters and ranking the sites by using PROMETHEE/GAIA method

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Abstract

Purpose In this study, Multi-criteria decision making (MCDM) approach was applied to assess and evaluate the heavy metals (Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Pb and Hg) pollution in Dilovası region (one of the largest industrial area in Turkey).

Methods The heavy metal content of 10 different locations have been evaluated and these locations are ranked according to their metal contents by using PROMETHEEGAIA method, which is one of the pairwise comparisons MCDM methods. PROMETHEE (Preference Ranking Organization Method for Enrichment Evaluation) method was used to rank the locations according to their heavy metal content and GAIA (Geometrical Analysis for Interactive Aid) method was used to analyse and show the relations between alternatives (locations) and criteria (heavy metal).

Results Analysis indicated that location d4 (small harbor of Hereke) is the most polluted especially by Pb, Cd, Cr and As. The location d8 is the least polluted area which is the farthest places from the harbors and industrialized zones.

Conclusions The ranking results clearly showed that the most contaminated locations are wastewater discharge points and small ports. The study also showed that PROMETHEE/GAIA method is very helpful to analyse environmental problems.

Keywords Heavy metals · Contamination · Pollution · MCDM · PROMETHEE / GAIA

Introduction

Translocation, deposition, remobilization and accumulation of heavy metals in seawater have been important issues for the seawater environment due to its toxicity, abundance and persistence in the environment. Recognitions regarding contamination and relative magnitudes from different sources have been widely discussed and reported [1–6].

One reason for heavy metals to be of concern is their deleterious effects on the environment and public health, especially if present at levels above a toxicity threshold [7, 8]. For example, high levels of Cu and Zn in water reduce cell division rates and damage liver, kidneys and nervous systems of marine life [9]. Many other heavy

metals, including chromium (Cr), arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb), also cause various acute or chronic toxicities; the bioaccumulation potential of these heavy metals enhances their environmental problems. Heavy metal residues in water may enter into the human food chain and results in public health problems [3]. Heavy metals in aquatic environments are increasingly recognized as important intermediate sources for subsequent pollution in seawater. Considerable efforts have been expended to assess their presence in harbors and bay. After being released from natural background or anthropogenic sources near the land surface, e.g., rivers carrying significant metal loadings, soluble heavy metal species are immobilized and deposited onto the sediment surfaces through various mechanisms. These immobilization mechanisms consists of adsorption onto soil/sediments by ion exchange, coagulation with dissolved or suspended species in water (e.g., organic matter), incorporation into the lattice structure of minerals, and precipitation by forming insoluble species of heavy metals [10, 11]. The high salinity of seawater enhances the aggregation of suspended particles,

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resulting in more rapid sedimentation of heavy metals [11]. As a consequence, the water and soil/sediments in harbors serve as a pool for heavy metals to be adsorbed, accumulated, and released to nearby and overlying areas. While many marine organisms, particularly those bound to marine sediments, may uptake heavy metals and play important roles in the food chain, the adverse effects of heavy metals in aquatic environments on the ecosystem and public health may be further enhanced [10, 12].

The earlier researchers have employed multivariate statistical analysis to investigate the distribution and contamination of heavy metals in seawater. Multivariate analysis with PCA (Principal component analysis) has been applied to classify contaminated areas in İzmit bay and Dil creek [13]. Multivariate analysis have been used to investigate and characterize contaminated area in the Kaoghsuung Harbor [3]. The pollution characteristics and distribution of heavy metals in Jinzhou Bay have been studied [6]. Most of the researchers have studied mainly assessment of metal contamination rather than ranking the locations [14–19]. In other words, all of the above studies are about the measurement of the heavy metal contamination of related locations.

The effective management of contaminated sites has become very important issue worldwide since the identified number of contaminated sites are increasing. The budget limitations and remediation costs make the management of these sites very difficult. Since the all sites can not be remediated at the same time, the necessity of prioritization or ranking of these sites by a scientifically defensible method is obvious. Multi Criteria Decision Making (MCDM) methods are scientific methods, and there are different methods such as ANP, TOPSIS, ELECTRE, PROMETHEE, etc. In this study, PROMETHEE/GAIA method is preferred because it is an outranking method, incorporates GAIA method and has a visual software support. Therefore PROMETHEE/GAIA is applied to asses and rank the contaminated sites for heavy metal contaminations. PROMETHEE method provides the ranking of all sampled sites and GAIA method acts as graphical decision aid to decision maker. GAIA helps the decision maker to interactively investigate and structure the problem, hence the results provided by the PROMETHEE ranking methods can be better understood.

The main objective of this paper is to rank and evaluate heavy metal contaminated sites by using PROMETHEE/GAIA method which is one of the Multi Criteria Decision Making (MCDM) methods. Based on this ranking and evaluation, decision makers for remediation of contaminated sites may manage their budget more properly. Although, the results given in this work for specific location, the method used in this study might be used for similar problem.

The rest of paper is organized as follows: In “**Materials and methods**” section, material and methods used for this study is presented. In “**Results and discussion**” section, results and discussions are provided. Finally, conclusions and further research given in the last section.

Material and methods

PROMETHEE-GAIA method

PROMETHEE methods belong to the family of outranking methods and developed in early 1980s by Brans *et al* [20]. They have suggested two methodological families: namely, PROMETHEE I for partial ranking and PROMETHEE II for complete ranking. Several years later, other versions of the PROMETHEE methods were developed to tackle more complicated decision-making problems. These versions include PROMETHEE III for ranking based on intervals, PROMETHEE IV for continuous cases, PROMETHEE V for problems with segmentation constraints, PROMETHEE VI for human brain representations, PROMETHEE GDSS for group decision-making, PROMETHEE TRI for sorting problems, and PROMETHEE CLUSTER for nominal classification [21]. The success of the PROMETHEE methodology in various applications is attributed to its flexibility and ease of use. Behzadian *et al* [21] indicated that the application of PROMETHEE can be grouped into nine areas: environmental management, hydrology and water management, business and financial management, chemistry, logistics and transportation, manufacturing and assembly, energy management, social and other topics (medicine, agriculture, education, design, and sports).

The PROMETHEE methods are based on pairwise comparison of alternatives $A = \{a_1, a_2, \dots, a_n\}$ for each criterion $F = \{f_1, f_2, \dots, f_m\}$. Therefore before starting the PROMETHEE methods, an evaluation matrix is needed to be constructed as following:

$$DM = \begin{bmatrix} f_1(a_1) & f_1(a_2) & f_1(a_3) & \dots & f_1(a_n) \\ f_2(a_1) & f_2(a_2) & f_2(a_3) & \dots & f_2(a_n) \\ \dots & \dots & \dots & \dots & \dots \\ f_m(a_1) & f_m(a_2) & f_m(a_3) & \dots & f_m(a_n) \end{bmatrix} \quad (1)$$

where

$f_i(a_j)$ shows the performance of i^{th} alternative on j^{th} criterion,

m is the number of alternatives, and

n is the number of criteria.

In order to apply PROMETHEE method, two additional information are needed. The first one is the

relative importance or weights/priorities of the considered criteria. The second one is the preference function to compare the contribution of alternatives with respect to every criterion. The preference structure of PROMETHEE method is based on pair-wise comparisons of alternatives. The intensity of preference of an alternative a_i over alternative a_j is defined by $p_k(d_k = f_k(a_i) - f_k(a_j))$. If the difference, $p_k(d_k)$ is small, then the decision maker would allocate a small preference to the one of the alternative or no preference. The larger differences indicates the larger preference. The value of the preference scales range between 0 and 1. “0” means there is no preference of alternative a_i over alternative a_j . “1” means there is strict preference of “ a_i ” over “ a_j ”. p_k is the preference function and six different types of preference functions is defined [20], namely usual criterion, quasi-criterion, linear criterion, level criterion, linear with indifference criterion, and Gaussian criterion. Some of the preference functions require threshold parameters (p : preference threshold, q : indifference threshold or s : Gaussian threshold). The value of threshold parameters should be defined by the decision maker. Unlike other popular multi-criteria methods, active participation of decision-makers or qualified specialists is compulsory as they recommend types of preference functions for every criterion [22, 23].

The weighted average of the preference function for alternatives a_i and a_j are calculated by using following equation:

$$\Pi(a_i, a_j) = \sum_{j=1}^n P_j(a_i a_j) w_j \quad (2)$$

where

w_j are weights associated with criteria j .

$\Pi(a_i, a_j)$ represents the strength of decision maker preference of alternative a_i over alternative a_j . As a measure of outranking character of alternative a_i , the leaving flow of a_i is calculated as follows:

$$\phi^+(a_i) = \frac{1}{n-1} \sum_{j=1, j \neq i}^n \Pi(a_i, a_j) \quad (3)$$

As a measure of outranked character of alternative a_i , the entering flow of a_i is calculated as follows:

$$\phi^-(a_i) = \frac{1}{n-1} \sum_{j=1, j \neq i}^n \Pi(a_j, a_i) \quad (4)$$

The positive outranking flow indicates how an alternative “ a ” discards the other alternatives. The higher the value of $\phi^+(a)$, the better is the alternative. For

PROMETHEE II, differences between the entering flow and the leaving flow for each alternative are calculated as net flow and as given below:

$$\phi^{net}(a_i) = \phi^+(a_i) - \phi^-(a_i) \quad (5)$$

The higher the value of $\phi^{net}(a)$ indicates that the alternative is better. That means an alternative with the highest $\phi^{net}(a)$ value is the best alternative. Then the alternatives are ranked from the best one to the worst one.

In order to better understand the results provided by PROMETHEE II, a geometrical tool known as GAIA plane have been proposed [24]. The basic idea of GAIA approach is to perform a PCA (principal component analysis) on the unicriterion net flows calculated for each alternative [25, 26]. The GAIA plane is defined by corresponding unit eigenvectors u and v , resulting from a unicriterion net flows covariance matrix, obtained using principal components analysis (PCA). Using PCA, it is possible to define a plane having the minimal amount of information lost by projection. In the GAIA plane, alternatives are represented by points, and the criteria are indicated by axes. The net flow of an alternative is the vector of its single criterion net flows on weight w . The orientation of axis indicates which criteria are compatible and which ones are in conflict. The length of axis will point out the discriminant criteria. The projection of the weight vector is referred as the decision axis (π). The direction of decision axis gives the direction of the best alternatives. A long decision axis signifies a strong power to select alternatives along the that direction. Criteria expressing similar preferences on the alternatives are oriented on the same portion of the GAIA plane, and the conflicting criteria on the alternatives are located on the opposite part of the GAIA plane [27].

Study area

The data used in this work has been obtained from [13]. In their study, water samples were taken from 10 locations along the Dil Creek and Izmit bay around the Dilovası region (Fig. 1). Dilovası district of the Izmit Bay is one of the most industrialized area in Turkey. They are 185 big companies serving in 45 sectors. These sectors are mainly metal (iron-steel, aluminum), chemistry (e.g., paint) and energy (coal fired electric plants). The area is located at the center of a bowl-like topographic structure, which causes serious environmental problems in the region. Sewer system of Dilovası region is directly connected to Dil Creek (along the locations d1, d2, d3 and d6) without any

treatment. Large industrial plants around this creek also discharge their solid and liquid waste into Dil Creek after limited treatment. As seen from Fig. 1, the Dil creek flows into the Izmit bay, which is in the eastern part of the Marmara sea. The heavy industrialization in the area have polluted the Dil creek and as a result the Izmit bay [13].

Data used for the study

The parameters used for ranking the sites are the heavy metals. The heavy metals are all naturally occurring substances which are often present in the environment at the low levels. However due to human activities, the amount of heavy metals increase in the environment and became dangerous for inhabitants because of their toxicity, persistence and non-degradability in the environment [28–31]. These heavy metals have been discharged into the rivers as a result of industrialization. For this study the most common of ten heavy metals (Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Pb and Hg) contamination of Izmit bay have been considered.

The heavy metal contamination of Izmit bay has been measured using ICP-MS by Bingol et al [13]. They only evaluated the metal contamination by using Chemometric evaluation methods. By using their data, the heavy metal contamination of sampled sites have been evaluated and contaminated sites are ranked by applying PROMETHEE-GAIA method. The PROMETHEE method was used to rank the locations from which the water samples were taken in accordance

with the heavy metals contents, while GAIA plane was used for graphical interpretation of the PROMETHEE results.

Results and discussion

The evaluation matrix of Table 1 is taken from [13]. In this table each heavy metal is assumed to be a criteria, and sites are alternatives. The Visual PROMETHEE (VP) software is used for the study [32]. The input values to the PROMETHEE-GAIA software is given in Table 1. In software, the user can define the alternatives, number of criteria, unit for each criterion, type of the criterion (beneficial or non-beneficial) and type of the preference function. For the sake of this study linear preference function is selected. The corresponding threshold (indifference and difference) values for all the criteria have been assessed by using different water quality standards and assistant menu of the VP program.

The weight of each heavy metals is determined by taking into account their environmental effects such as risk to human health, toxicity, carcinogenic and accumulation in water. According to literature, the heavy metals do not have the same significance, for example Cd, Hg, Pb and As is much more risky to human health than Mn, Cr, Co, Ni, Cu, and Zn. The toxicity order for Cd, Cu and Zn is $Cd > Cu > Zn$ [33–40]. The influence of each metal on environment and health is given in Table 2. The following four parameters have taken into account to estimate the weights; risk to human, toxicity,

Fig. 1 Dilovası Region in Turkey

Table 1 Evaluation matrix [13]

Location	Metal Concentration (mg/l)									
	Cr	Mn	Co	Ni	Cu	Zn	As	Cd	Pb	Hg
d1	0,03	770,66	30,21	2,49	13,25	5,34	1,43	0,15	8,77	2,78
d2	0,02	364,75	554,15	10,42	12,92	6,26	10,51	0,09	2,84	0,07
d3	0,02	331,67	575,61	7,41	13,41	6,35	13,41	0,13	3,12	0,07
d4	0,26	160,13	211,67	8,17	1,66	5,34	8,13	0,35	18,15	0,02
d5	0,07	139,58	123,35	6,77	1,14	2,36	4,05	0,03	1,32	0,02
d6	0,08	311,76	113,67	5,27	2,77	3,96	4,63	0,86	2,08	0,01
d7	0,06	207,16	89,53	7,20	2,17	3,62	8,46	0,23	6,36	0,02
d8	0,03	159,02	21,13	5,08	4,74	2,87	4,34	0,05	0,85	0,02
d9	0,01	303,53	38,49	16,76	3,16	23,91	4,06	0,03	0,02	0,02
d10	0,02	303,00	17,84	16,25	2,99	21,86	1,04	0,20	0,60	0,01
Min	0,010	139,58	17,84	2,49	1,14	2,36	1,04	0,02	0,019	0,008
Max	0,263	770,66	575,61	16,76	13,41	23,91	13,41	0,864	18,15	2,78
Preferences	Cr	Mn	Co	Ni	Cu	Zn	As	Cd	Pb	Hg
Min/Max	min	min	min	min	min	min	min	min	min	min
Weight	0,09	0,09	0,05	0,09	0,04	0,09	0,1	0,16	0,14	0,16
Pre. function	Linear	Linear	Linear	Linear	Linear	Linear	Linear	Linear	Linear	Linear
Thresholds										
q	0,01	181,00	204,00	3,98	0,61	1,08	0,62	0,04	0,91	0,14
p	0,06	365,00	425,00	9,25	3,68	6,46	3,71	0,25	5,44	0,83

carcinogen and accumulation in water. Especially accumulation character of heavy metal is more important. Then, every heavy metal is graded according to their effect as given in Table 2 and the weight to each metal

was estimated by grading parameters. The weight values are positive and normalized.

Then values in Table 1 and Table 2 are entered the Visual PROMETHEE program. The estimated leaving, entering and

Table 2 Given weights for each criterion

Heavy Metal	W (%)	Influence on environment and health (http://www.lenntech.com/periodic/elements)
Pb	14	retained in the human organism and is carcinogenic; Lead accumulates in the bodies of water organisms and soil organisms. These will experience health effects from lead poisoning
Cd	16	accumulate in mussels, oysters, shrimps, lobsters and fish; retained in the human organism and is carcinogenic
Hg	16	Poisonous, not carcinogenic; toxic to the nervous systems; bioaccumulates in fish and shellfish
As	10	one of the most toxic elements; fish absorb arsenic; Toxic to plants
Cr	9	Damage to the nervous system, fatigue, irritability; Chromium(VI) is mainly toxic to organisms. It can alter genetic materials and cause cancer.
Mn	9	Influence on the nervous system Manganese can cause both toxicity and deficiency symptoms in plants, one out of three toxic essential trace elements
Co	5	accumulation in plants and animals may occur, possibly carcinogenic to humans(2B-IARC)
Ni	9	No accumulation in plants or animals. Evidence for carcinogenicity in humans (Group 1 and 2B by IARC)
Cu	4	it can accumulate in plants and animals. Harmful to animals and plants
Zn	9	relatively harmless. Only exposure to high doses has toxic effects, Zinc may also increase the acidity of waters., fish can accumulate zinc
Total	100	

the net flow of alternatives are given in Table 3. Since the preference is minimization, the first rank indicates the least contaminated location. Location d8 is the least, location d4 is the most contaminated location.

In Fig. 2, the PROMETHEE II complete ranking of the considered locations is provided graphically, based on the net outranking flow values. In Fig. 2, the top half of the scale (in green) corresponds to positive net outranking scores and the bottom half (in red) corresponds to negative scores. It is clearly observed that location d8 (seawater) tops the ranking list, followed by location d5 (sea water), location d9 (Dil Creek-Sea eastern part) and location d10 (Dil creek-Sea western part). It signifies that these four locations are the least contaminated locations with respect to ten heavy metal concentration (evaluation criteria). Those are the farthest locations from the heavy metal sources. The heavy metal concentration of location d4 (small harbor) is the worst. It is also obvious that the locations d1, d6, d7 (small harbor) and d3 are heavily contaminated. These places are very close to the industrialized waste water discharge points as seen in Fig. 1.

In order to further analyze the results, the GAIA plane obtained from the software is given in Fig. 3. The positions of the criteria, alternative locations and decision stick (π) axis are exhibited. The direction of the decision axis indicates the superiority of locations d8 over others followed by locations d9 and d10. The locations d9 and d10 are characterized by Ni and Zn metals while the locations d1, d2, and d3 are mainly characterized by Cu and Co. In other words, the Zn contamination in location d9 and d10 is very high. Hg and Mn metals are found to be in low level for all locations except the location d1. The locations d4, d6 and d7 are characterized by Co, Cr, and Pb metals. The location d1 is characterized by Cu, Co As, Hg and Mn. Location d5, d9 and d10 are less contaminated by heavy metals of Co and As, but highly contaminated by Ni, Zn, Mn and Hg. The values of criteria Cr, Cd and Pb are unfavorable for locations d4, d6 and d7.

The quality level of GAIA plane is 83,1%, which indicates that the analysis are reliable [31]. Then the locations can be

Table 3 PROMETHEE flow table

Locations	Phi	Phi+	Phi-
d8	0,27	0,30	0,03
d5	0,23	0,30	0,07
d9	0,10	0,27	0,17
d10	0,07	0,26	0,19
d2	-0,02	0,18	0,20
d3	-0,04	0,18	0,22
d6	-0,05	0,20	0,25
d7	-0,07	0,18	0,24
d1	-0,20	0,22	0,42
d4	-0,29	0,13	0,42

Fig. 2 PROMETHEE II ranking of 10 locations

classified into five groups based on the contaminated heavy metal and their locations on the GAIA plane:

- Group 1: d4, d7 and d6 (contaminated by Cr, Cd and Pb)
- Group 2: d2, d3 (contaminated by Cu, Co, As)
- Group 3: d1 (contaminated by Hg, Mn and Cu)
- Group 4: d9, d10 (contaminated by Zn, Ni)
- Group 5: d5, d8 (non important)

Figure 4 shows the effects of heavy metals on each locations. In this figure, each bar for each locations is composed of different slices, and the height of each slice is proportional to the contribution of one heavy metal on the final ranking of locations. Positive (upward) slices correspond to good features while negative (downward) slices represent the weaknesses. The 10 locations are positioned from left to right according to the PROMETHEE II ranking. From this figure, it is clear that location d8 is better in performance with respect to

To analyze the ranking of the sites under different weight of parameters and equal weight of parameters a sensitivity analysis is carried out by using the walking weight option of VP program. The results are provided in Table 4 and Fig. 5. The idea of the sensitivity analysis is to increase each parameter's weight 100% while reducing the weights of others in equal rate. Six

Table 4 Sensitivity Analysis Results

Conditions	Criteria Weight										Alternative Rankings									
	Cr	Mn	Co	Ni	Cu	Zn	As	Cd	Pb	Hg	1	2	3	4	5	6	7	8	9	10
Main	0,09	0,09	0,05	0,09	0,04	0,09	0,10	0,16	0,14	0,16	d8	d5	d9	d10	d2	d3	d6	d7	d1	d4
As(100%)	0,08	0,08	0,04	0,08	0,04	0,08	0,20	0,14	0,12	0,14	d8	d5	d10	d9	d6	d1	d2	d7	d3	d4
Cd(100%)	0,07	0,07	0,04	0,07	0,03	0,07	0,08	0,32	0,11	0,13	d8	d5	d9	d2	d10	d3	d7	d1	d6	d4
Pb(100%)	0,07	0,07	0,04	0,07	0,03	0,07	0,08	0,13	0,28	0,13	d8	d5	d9	d2	d10	d3	d7	d1	d6	d4
Hg(100%)	0,07	0,07	0,04	0,07	0,03	0,07	0,08	0,13	0,11	0,32	d8	d5	d9	d10	d2	d3	d6	d7	d4	d1
Cr(100%)	0,18	0,08	0,04	0,08	0,04	0,08	0,09	0,14	0,13	0,14	d8	d5	d9	d10	d2	d3	d7	d6	d1	d4
Equal Weights	0,10	0,10	0,10	0,10	0,10	0,10	0,10	0,10	0,10	0,10	d8	d5	d9	d10	d6	d7	d7	d3	d1	d4

combinations are investigated. It can be seen from the Table 4 and Fig. 5 that the ranking of sites are not changing dramatically. The best and worst locations almost remains the same.

Conclusions

The objective of the most of the studies about metal contamination by using multivariate analysis is to classify and group contaminated areas on the basis of heavy metals contamination. The PROMETHEE/GAIA method is used to rank the contaminated areas according to the environmental effects of contained heavy metals. Remediation of contaminated areas has become very important issue for sustainable environment. The effective utilization of limited budgets forces decision makers to determine priority of contaminated sites for remediation. The PROMETHEE/GAIA of MCDA methods is very useful tool for decision makers.

In this paper, an attempt is made to integrate PROMETHEE and GAIA methods to assess and rank the sampled 10 locations in terms of heavy metal contamination in Dilovası area, Turkey based on ten heavy metals. The locations are ranked from the most environmentally risky area to the least one. It is observed from the analysis that the location d4 (small harbor of Hereke) is the most polluted especially by Pb, Cd, Cr and As. The location d8 is the least polluted area which is the farthest places

from the harbors and industrialized zones (Fig. 1). As it can be seen from Fig. 1, the locations d8, d7, d9 and d10 are the farthest locations from the heavy metal sources. The locations d4 and d7 are small ports, and main contaminant is Pb. This indicates that the potential sources for heavy metal pollutions are transport services and industrial activities. When this result was compared to other metal-polluted areas from different regions of the world given in [3], the concentration of heavy metals in this area was relatively low. In addition to that the other areas are polluted mainly by Cr, Pb, Zn and Cu. The study area is polluted by Pb, Mn, Zn and Cu. The Mn concentration does not given in other locations, but it exist in the study area. The concentration of Cr is also very low compared to other ports of world.

This paper also shows the usefulness of PROMETHEE-GAIA method-based graphical tool in guiding the decision makers analyzing and arriving at the best prioritization or ranking of contaminated sites. The method can be also applied similar problems. Among different MCDM methods, PROMETHEE is better because of its simple design, ease of computation and suitable results. However, the preference functions may result in some hesitations for decision makers. The limitations of using PROMETHEE are the definitions of preference function and weights of criteria. These have to be defined by the user or decision makers. However, this

Fig. 5 Sensitivity analysis under different criteria

limitation is overcome by applying sensitivity analysis which has not existed in other MCDM methods.

As a future work, this study can be extended to rank contaminated sites according to environmental risk and health risk grouping heavy metals and other pollutants.

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Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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