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A Multi-Metric Index for Hydrocarbons Source Apportionment

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ABSTRACT: Several studies have been conducted to develop more accurate and precise indices for hydrocarbons source apportionment. The present study, however, develops a new multi-metric index for hydrocarbons source apportionment. It measures Poly Aromatic Hydrocarbons (PAHs) concentration at six stations with well known petrogenic origin, calculating Phe/An, Flu/Py, Chr/BaA, BaA/Chr, An/(An+Ph), Flu/(Flu+Pyr), and IP/(IP+Bghi) indices. All the indices could correctly determine the source of hydrocarbons, except for IP/(IP+Bghi). Subsequently, it uses principle component analysis method to create a combined multi-metric index, based on PAHs, the concentration of which also contributes to the evaluation of new index performance in stations with known origins. Results show that the new multi-metric index can determine the source of hydrocarbons with greater certainty. Then, using this index, the potential source of contamination in the area has been divided into six sections, namely HPY, MPY, LPY, MPE, HPE, and LPE, which indicate origin of high, moderate, and low risk of petrogenic contamination, as well as source of pyrolytic contamination with high, moderate, and low probabilities.

Keywords: Multi-Metric Index, Source Apportionment, Poly Aromatic Hydrocarbon, Petrogenic and Pyrolitic Source.

INTRODUCTION

Due to their acute and chronic effects as well as their potentials to produce genetic changes and destroy ecosystems, petroleum hydrocarbons are one of the most significant environmental pollutants. Today, a large amount of hydrocarbons enters the environment from different sources, e.g. wastewater, runoff, leakage incidents, oil refinery, etc (Commendatore et al., 2000; Cuypers et al., 2002; Bayat et al., 2016).

In many cases, the hydrocarbons travel far away from their sources, or there are several possible sources for an observed hydrocarbon pollution in the environment, especially, in case of non point sources. Also, it is possible that the type and concentration of hydrocarbons change during their travels in the environment. Therefore, it is quite difficult to exactly determine the origin of an observed hydrocarbons pollution in the environment (Hofmann et al., 2007; Pies et al., 2008; Stout et al., 2002; Stout and Wasielewski, 2004; Saber et al., 2006). Numerous indices can provide valuable information about the source and transport processes of organic materials (Hu et al., 2009), several of which have been developed to identify the source of hydrocarbon pollution, based on either physical and chemical properties of hydrocarbons (e.g. reactivity, solubility, and thermodynamic properties) or statistical analysis of the sources data (Zaghden et al., 2006; Medeiros et al., 2005).

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These indices include a number of hydrocarbon compounds, such as Poly Aromatic Hydrocarbons (PAHs), Pristane, Phytane, and N-alkanes with several different carbon atoms, developed and used for specific purposes (Tolosa et al., 2004, 2009; Gavad et al., 2008).

Pyrolitic PAHs are high molecular weight solid or gas compounds that originate from incomplete combustion of fossil fuels (coal, gasoline, and diesel oil). On the contrary, the origin of petrogenic PAHs with a low molecular weight, is crude oil as well as its refinery products, while diagenetic PAHs are produced through decomposition of organic materials. For example, long-term organics' retention and decomposition in sediments could result in the creation of PAH compounds (Budzinski et al., 1997; Boonyatumanond and Wattayakorn, 2006). Table 1 shows the main indices, developed for hydrocarbon source apportionment, using PAH compounds.

Table 1. Hydrocarbon source apportionment, based on PAH compounds

index	Criteriafor petrogenicorigin	Criteria for pyrolitic origin	Explanation	Researcher
Phe/An	>10	<10	Study was conducted at 300°C, 700°C, and 1000°C. As the temperature increased, Anthracene was produced. A rise in retention time resulted in more phenanthrene. The Index's limits were not accurate (Alberty and Rif, 1988; Bauard et al., 1998; Boitsov et al., 2009; Soclo et al., 2000). The ratio was 4 at cars' exhaust, ~ 4-10 for fossil fuels, and 50 for crude oil (Gschwend and Hites 1981; Colombo et al., 1989).	Baumard et al., 1998
Chr/BaA	>1	<1	-	Soclo et al., 1999
Flu/Pyr	<1	>1	For American and European coal. As for Australia, it was 0.3-0.7, being a sign of Pyr dominance (Masclet, 1987). Both Phe/An and Flu/Pyr should be used to obtain a proper apportionment for PAHs source (Budzinski et al., 1997). The ratio was 0.6-0.9 for crude oil, and 1.4 and 1 for burning wood and coal, respectively (Gschwend and Hites 1981).	Baumard et al., 1998
Flu/(Flu+Pyr)	<0.4	>0.5	A ratio of 0.4-0.5 shows the burning of liquid fossil fuels, while the ratio >0.5 is a sign of organic materials burning (e.g. coal) (Zaghden et al. 2007). A ratio of <0.4 is related to coal, except for Australian coal which was >0.4.	Yunker, et al., 2002
BaA/Chr	<0.4	>0.4	Compounds with almost same physical and chemical properties were selected.	Gschwend and Hites, 1981
IP/(IP+Bghi)	<0.2	>0.5	-	Yunker et al., 2002
BaA/(BaA+Chr)	<0.2	>0.35	The relative stability of PAHs were calculated by molecular simulation software. benzo (a) anthracene to benzo (a) anthracene + Chrysene (Molecular Weight =228 g/mol) was used for source apportionment. Ratios 0.2-0.35 refers to both sources.	Yunker et al., 2002
An/(An+Ph)	<0.1	>0.1	The ratio was <0.1 for common petroleum and diesel, and >0.1 for shales.	Yunker et al., 2002
Low Molecular Weight/High Molecular Weight	High	Low	The index was developed empirically, based on kinetic and thermodynamic relations.	Budzinski et al., 1997
BaA/(BaA+Chr)	<0.2	>0.35	The relative stability of PAHs was calculated by molecular simulation software.	Yunker et al., 2002
Naphthalene	-	-	The ratio is expressed in percentage.	Tolosa et al., 2004

The compounds, used for index development, should be selected among those with similar physical and chemical properties (Colombo et al., 1989, 2005; Gschwend and Hites, 1981). Also, the concentration ratios of PAHs with molecular weights of 178, 202, 228, and 276 g/mol got to be known as the best indices for apportionment of natural or human sources of PAHs (Yunker et al., 2002).

Phenanthrene (Phe) and Anthracene (An) are two structural isomers with different physical and chemical properties and behaviour in the environment, leading to various Phe/An relations with the PAHs' source. The former is thermodynamically more stable than the latter, thus Phe/An is higher for petrogenic than pyrolitic sources. High combustion temperature in pyrolytic sources raises the An share (Soclo et al., 2000; Boitsov et al., 2009; Bauard et al., 1998; Alberty and Rif, 1988). Another widely-used index is the ratio of Fluoranthene to Pyrene (Flu/Py) with thermodynamic behaviour similar to Phe/An. The higher the share of petrogenic sources Benzo[α], the lower the Anthracene/Chrysene (BaA/Chr) in sediment (Colombo et al., 1989). Gschwend and Hites (1981) stated BaA/Chr to be almost 0.24 to 0.40 for crude oil.

PAHs with molecular weights of 178 g/mol (e.g. An/(An+Phe)) and 202 g/mol (e.g. Flu/(Flu+Pyr)) are commonly used for source apportionment (Gschwend and Hites, 1981). Flu and Pyr are usually generated during the condensation of low molecular weight PAHs in combustion processes and are known as pyrogenic products (Soclo et al., 2000).

Isomers of BaA and Chr exist in small quantities in petroleum products, while their concentration is higher in heavy refinery cuts, like asphalt and coke (Wu et al., 2011).

Also, the percentage of Perylene to total PAH compounds is an index for hydrocarbon source apportionment, while a ratio greater than 4% is related to natural sources. Usually, lower values are observed in anthropogenic sources (Budzinski et al., 1997).

Generally, petrogenic sources are defined by the predominance of PAHs with low molecular weight, while PAHs with high molecular weight are dominant in pyrogenic pollution (Bundzinski et al., 1997). Also, high concentration of Naphthalene could be a sign of oil pollution, since this combination can be found abundantly in the light oil (Yang et al., 2000; Soclo et al., 2000). Thus, Naphthalene and Low Molecular Weight/High Molecular Weight are introduced as indices for source identification.

Hydrocarbon apportionment indices are extensively and worldwidely used for hydrocarbon source identification; even though, most of them have been developed for specific hydrocarbon sources and climatic conditions with little or no data concerning their worldwide validity. For example, Masclet et al. (1987) and Nelson et al. (1988) showed that Flu/Py was above 1 for American and European coal, while its value ranged between 0.3 and 0.7 for Australian coal. In the present study, in the first step, existing indices got evaluated and in the next step, by means of statistical analyses, a suitable multi-metric indices for the south of Tehran was developed.

MATERIALS AND METHODS

According to Figure 1, in order to determine the distribution of PAH compounds south of Tehran, sampling of sediment, water, and soil was carried out in 33 stations. These samples included 5 sediment samples, 13 water samples, and 15 soil samples. Sampling was done in two steps during January and May 2015.

Water, soil, and sediment samples were transfered to the laboratory immediately after collection, there to be stored below 4°C. They were prepared and analyzed according to MOOPAM test method (ROPME, 1998). PAH compounds were analyzed using GC-MS, with GC Agilent 7890, MS Agilent

5975 (column HP5, 30 m, inner diameter=0.25 mm). The experimental conditions included a temperature of 300°C at injection site with the samples injected with splitless status into the device. The column was kept at 60°C for 2 min and then increased up to 280°C with a rate of 5°C/min, and was kept at this temperature for 20 min. The average recovery rate was between 73 and 95, and then the results were corrected by recovery. Afterwards, the actual concentration of the analyses got calculated. Also, in order to control the method's quality, blank and replicate samples were used in each series of analyses (Clesceri et al., 1989). The discriminant test was used to evaluate the performance of the hydrocarbon sources apportionment indices, while principle component analysis (PCA) was used to develop a multi-metric index. The statistical analysis was performed, using SPSS (v.22).

RESULTS AND DISCUSSION

Table 2 shows the concentration of PAH compounds in six stations, which were a mix of water, soil, and sediment samples.

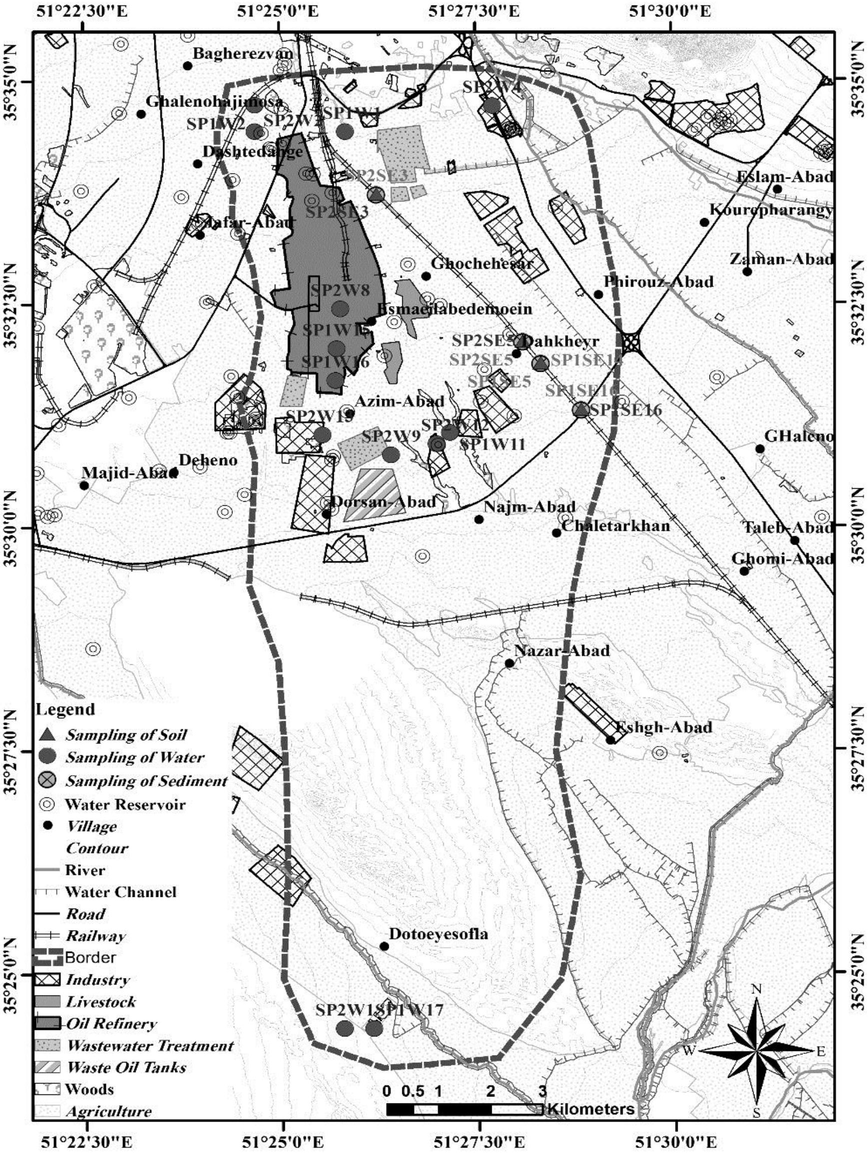

Fig. 1. Sampling points

Table 2. Concentration of aromatic compounds in six control stations (ppb)

PAH$_S$	SP1S8	SP1W15	SP1SE16	SP1W16	SP2W8	SP2W9
Naphtalene	101510.00	70.26	10511.12	5467.97	147127.50	159.53
Acenaphtylen	19556.80	6.78	2177.94	199.16	3534.78	N.D
Acenaphten	6392.80	14.47	399.41	57.15	14664.30	54.86
Florene	25951.40	1.82	5199.14	624.69	2259.80	39.95
Phenanthrene	383383.50	109.13	11992.02	1583.60	20690.37	92.87
Anthracene	9213.10	5.00	5.00	5.00	524.21	5.00
Fluorantene	20668.90	22.38	450.81	17.77	254.74	5.00
Pyrene	446642.00	144.90	3938.40	216.44	3343.83	17.71
Benzo(a)ant	5.00	5.00	5.00	22.81	67.24	N.D
Chrysene	37174.55	80.97	2681.08	273.92	2086.58	N.D
B(b)F	N.D	11.93	24.55	21.14	N.D	N.D
B(k)F	1231.69	N.D	626.90	37.17	389.24	12.78
B(a)P	39072.25	13.53	161.03	44.91	421.81	N.D
Dibenzo(a,h)Anthracene	N.D	28.86	N.D	N.D	39.31	N.D
Benzo(g,h,i)Perylene	2126.24	5.00	5.00	N.D	5.00	N.D
Indeno(1,2,3-cd)Pyrene	2025.00	46.04	39.14	N.D	331.70	N.D

Table 3. Index values in stations with confident pollution sources

Station	Phe/An	Flu/Pyr	Chr/BaA	BaA/Chr	An/ (An+Ph)	Flu/ (Flu+Pyr)	BaA/ (BaA+Chr)	IP/ (IP+Bghi)
Petrogenic	>10	<1	>1	<0.4	<0.1	<0.4	<0.2	<0.2
Pyrogenic	<10	>1	<1	>0.4	> 0.1	>0.5	>0.35	>0.5
SP1S8	41.61	0.05	7434.91	0.0001	0.02	0.04	0.0001	0.49
SP1W15	21.83	0.15	16.19	0.6	0.04	0.13	0.06	0.91
SP1SE16	2398.4	0.11	536.22	0.0019	0.0004	0.11	0.0018	0.9
SP1W16	316.72	0.08	12.01	0.08	0.0031	0.08	0.076	N.D
SP2W8	39.47	0.08	31.03	0.03	0.02	0.07	0.03	1
SP2W9	18.57	0.28	N.D	N.D	0.05	0.22	N.D	N.D
Success, %	100	100	100	100	100	100	100	0

Eight indices were calculated for the six stations, wherein the source of hydrocarbons was definitely petrogenic. Seven indices (except IP/(IP+Bghi)) correctly identified the pollution origin (Table 3). The rejected index was omitted in further analysis for development of multi-metric index.

To develop an index, capable of defining the source of hydrocarbons more accurately, the succeeded indices, namely Phe/An, Flu/Pyr, BaA/Chr, An/(An+Phe), Flu/(Flu+Pyr), BaA/(BaA+Chr), and Chr/BaA, were taken into consideration. Then, the indices were analysed, using PCA procedure by means of SPSS, and factors with wi > 1 were selected (Table 4). The 1st component (C1) with maximum factor loading included Flu/Pyr, Flu/(Flu+Pyr), BaA/Chr, Phe/An, and An/(An+Phe); the 2nd one (C2) contained Chr/BaA; and the 3rd one (C3) had BaA/(BaA+Chr). After that, the coefficient, Ai was calculated (Table 5).

Table 4. Components' specification

Component	v_i	$\sum v_i$	w_i
C_1	0.4305	0.43055	3.014
C_2	0.27226	0.70281	1.906
C_3	0.14295	0.84576	1.001
C_4	0.08706	0.93282	0.609
C_5	0.05031	0.98313	0.352
C_6	0.01335	0.99648	0.093
C_7	0.00352	1.00	0.025

Table 5. Results of PCA and the coefficients

Index	C_1	C_2	C_3	$A_i = \sum_{i=1}^{3}(C_i \times v_i)$
Phe/An	-0.627	0.613	0.431	-0.04
Flu/Py	0.755	0.606	-0.112	0.49
Chr/BaA	-0.591	0.714	0.324	-0.01
BaA/Chr	0.707	-0.133	0.523	0.34
An/(An+Ph)	0.605	0.599	0.122	0.4125
Flu/(Flu+Pyr)	0.708	0.441	0.152	0.4251
BaA/(BaA+chr)	0.577	-0.360	0.621	0.24
Variance	0.435	0.272	0.142	-

Where Ci = component I, vi = variance, ∑vi = cumulative variance, and wi = special variance value.

In the next step, the intended formula was obtained for the study area (Ii=|Ai|×Xi). Rates below 0.3 were not considered in the calculation.

This was done in a way through multiplying the coefficient by the ratio in the worst condition of petrogenetic or pyrolitic pollution and defining the range of the index as well as through multiplying the coefficient (A) by the ratios and obtaining the border of petrogenic and pyrolitic pollution.

In the next step, changes in the pollution from the border of the worst state for petrogenic and pyrolitic pollution were divided into three groups, coded in a way that the first letters represented pollution severity, itself classified into three groups of low, moderate, and high. The next two letters specified the type of pollution, classified into two categories of petrogenic and pyrolitic. For example, HPE was a high petrogenic pollution (Table 6).

Pollution severity of the sampling stations was analysed, using the aforementioned data. All stations showed petrogenetic pollution at different levels.

In previous stages, the pollution for stations was displayed in parts. In this stage, however, pollution varied in different environments (water, soil, and sediment) and was displayed by collecting the indices and compiling an index. As shown in Table 7, the coefficient number (A) was multiplied by the ratios in the worst conditions of petrogenetic and pyrolitic pollution, then to be summed up with the petrogenetic and pyrolitic column numbers. According to Table 8, the numbers ranged from (1.7) to (-1.8), pertaining to the worst condition of petrogenetic and pyrolitic pollution, and were equal to 0.458, i.e., the border between these two pollutions. The range of these numbers was divided into 6 parts.

In Table 9, Ai is multiplied by ratio columns of water, soil, and sediment and the numbers of every column has been summed. The total number is compared with Table 8 data. As shown, the pollution in the three environments was an HPE one.

Table 6. The calculated range of the indices

Index	the highest value for petrogenic sources	the lowest value for pyrolitic sources	coefficient (A)	range	border of Petrogenetic and Pyrolitic sources
Phe/An	≥50	≤4	-0.4	2.07-0.17	0.414
Flu/Py	≤0.6	≥1.4	0.49	0.29-0.69	0.490
Chr/BaA	≥9	≤0.5	-0.01	0.12-0.007	0.014

Table 7. The calculated range of indices for the study area

Index	A_i	the highest value for petrogenic sources	the lowest value for pyrolitic sources	highly petrogenic pollution	highly pyrolitic pollution
Phe/An	-0.04	≥50	≤4	-2.07	-0.17
Flu/Pyr	0.49	≤0.6	≥1.4	0.29	0.69
Chr/BaA	-0.01	≥9	≤0.5	-0.12	-0.007
BaA/Chr	0.34	≤0.24	≥0.9	0.08	0.31
An/(An+Ph)	0.4125	≤0.0004	≥0.9	0.00002	0.37
Flu/(Flu+Py)	0.4251	≤0.0003	≥0.8	0.0013	0.34
BaA/(BaA+Chr)	0.24	≤0.0001	≥0.9	0.00002	0.22

Table 8. Proposed hydrocarbon source apportionment classification

Range of index	Definition	Abbreviation
-1.818 : -1.509	High petrogenic pollution	HPE
-1.509 : -0.3	Medium petrogenic pollution	MPE
-0.3 : 0.458	Low petrogenic pollution	LPE
0.458 : 0.888	Low pyrogenic pollution	LPY
0.888 : 1.3	Medium pyrogenic pollution	MPY
1.3 : 1.7	High pyrogenic pollution	HPY

Table 9. Calculation of the overall ratio for different environments

Ratios	A_i	water ratios	sediment ratios	soil ratios	water	sediment	soil
Phe/An	-0.04	59.64	505	89.38	-2.47	-20.93	-3.7
Flu/Py	0.49	0.23	0.33	0.57	0.11	0.16	0.28
Chr/BaA	-0.01	17.28	131.5	42	-2.4٤	-1.81	-0.58
BaA/Chr	0.34	0.14	0.11	0.19	0.05	0.04	0.07
An/(An+Ph)	0.4125	0.04	0.02	0.11	0.02	0.01	0.05
(Flu/(Flu+Pyr)	0.4251	0.22	0.24	0.28	0.09	0.1	0.12
BaA/(BaA+chr)	0.24	0.1	0.08	0.08	0.02	0.02	0.02
Sum					-2.41	-22.41	-3.75

Table 10. Recommended PAH-based indices

ratio	group 1	group 2	group3
Phe/An	2.916	0.396	0.024
Flu/Py	-59.981	-15.869	-4.626
Chr/BaA	0.62	-0.062	0.146
BaA/Chr	233.441	42.361	9.266
An/(An+Ph)	197.833	41.935	-2.485
Flu/(Flu+Pyr)	145.954	46.133	17.138
BaA/(BaA+chr)	-496.653	-95.425	51.831

To select the suggested ratios, the discriminant test in SPSS software program was used. At first, measurement stations divided the samples into three groups and then the above analysis was carried out. As shown in Table 10, the ratios were divided into three groups, thus the gray parts enjoyed the lion's share. In the next stage, the coefficients of the three groups were multiplied by the numerical value of the ratios and the group with the greatest weight was selected. According to the following relations, the first group enjoyed the lion's share in PAH compounds and the proposed rates were selected from this group, which stood in the following order: BaA/(BaA+Chr), BaAN/Chr, An/(An+Ph), Flu/(Flu+Pyr), Flu/Py, and Phe/An.

Group 1 (2.916* X1) +… (-496.653* X7) = 474

Group 2 (0.396* X1) +…. (-95.425 * X7) = 6.8

Group 3 (0.024* X1) +…. (51.831 * X7) = 8.46

CONCLUSION

In this paper, eight PAH-based hydrocarbon apportionment indices were evaluated and combined, using PCA procedure. It developed a multi-metric index for the source apportionment.

The pollution level in the area was divided into six parts, namely HPY, MPY, LPY, MPE, HPE, and LPE, which stood for high, medium, and low pyrolitic pollution, respectively. According to this index, the pollution in this paper turned out to be HPE. Results from analysis of PAH compounds ratios in the stations with pollution source revealed that seven ratios of PHE/AN, FLU/PY, Chr/BaA, BaA/Chr, An/(An+Ph), Flu/(Flu+Pyr), and BaA/(BaA+chr) were appropriate for evaluating hydrocarbons in southern Tehran.

Discriminant statistical analysis showed that PAH compounds, including PHE/AN, FLU/PY, BaA/Chr, An/(An+Ph), Flu/(Flu+Pyr), and BaA/(BaA+chr) had the best function and Chr/BaA had the worst function in evaluation of hydrocarbons in southern Tehran. The study developed various indices, concerning hydrocarbon source identification using statistical analysis of a good index for hydrocarbon source detection in Southern Tehran. These indices can determine the level of pollution, unlike the ones, presented in previous studies that could only identify the source of pollution. It should also be noted that the index used in the present study was appropriate for this study area.

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