

Can the Number of Correlation Coefficient (R Value) as a Statistical Indicator of Metal-Polluted Ecosystem? A Comparison between Polluted and Unpolluted Ecosystems

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Article Info

Article history:

Received May 8, 2013

Revised Aug 22, 2013

Accepted Sep 4, 2013

Keyword:

Correlation coefficient

Polluted ecosystem

Significant correlation

Statistical indicator

ABSTRACT

The present paper is the first in the literature to present evidence that correlation coefficient (r value) can be used as a potential indicator of metal-polluted sediments collected from polluted and unpolluted ecosystems which were also supported by a few reported studies. Based on the correlation analysis on the heavy metals in the surface sediments, collected from two contrasting ecosystems, polluted and unpolluted areas, it was evidently found that the polluted ecosystems had higher numbers of significant correlation coefficients ($P < 0.05$) than the unpolluted ones. This could be due to the fact that the polluted ecosystems had wider ranges of values of heavy metal concentrations due to some elevated levels of metals recorded. Therefore, the correlation coefficients (r values) can be used as potential indicators for the polluted ecosystems. However, cautions should be exercised in which the monitoring studies should be able to truly reflect the overall environmental quality of the ecosystem, ranging from effluent-receiving point sources to the clean sites far away from the sources. However, further investigations are still necessary before number of significant r values can be confidently employed as indicator of metal-polluted ecosystem.

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1. INTRODUCTION

In environmental pollution studies, provision of data and evidence for a polluted environment is a major task for most ecotoxicologists. This is due to the fact that, based on field observation on the polluted sampling sites, high pollutant levels and wide ranges of pollutant concentrations for the organisms and abiotic components (especially sediments and water quality), is expected from high anthropogenic sites [1] although sometimes this might be not necessarily reflective of observable human activities [2]. However, if such assumptions are based solely on employment of the high metal concentrations and wide ranges of pollutant levels, the interpretation of data could be just another environmental quality report, even though it is still relevant from ecotoxicological point of view. Hence, more evidence based on statistical approach is much needed in order to prove that the ecosystem is being polluted.

Correlation may be described as the degree of association between two variables [3]. The correlation coefficient (r) can take on virtually any value between -1 and +1, so care must be taken when interpreting the r value [4]. In the case where a very polluted site can produce a very high level of metals, thus, statistically as outliers. Since we understand the reason of these outlier due to anthropogenic sources, this outlying points should not be removed [3].

Since such information is lacking in the literature, the objective of this paper was to compare the correlation coefficients of metal concentrations in the surface sediments, collected from polluted and unpolluted ecosystems and then the potentials of coefficient (r value) as an indicator of pollution was discussed.

2. MATERIALS AND METHODS

The data for the sediment data at the two same lakes Kelana Jaya were cited from those reported by Ismail et al. (2004) [5] while those of the four geochemical fractions of Cd, Cu, Pb and Zn in the surface sediments of the unpolluted offshore and polluted intertidal area of the west coast of Peninsular Malaysia were obtained from those reported by Yap *et al.* (2002, 2003, 2005a, 2005b) [6]-[9]. The four fractions considered were the 1) 'easily, freely, leacheable or exchangeable (EFLE)', 2) 'acid-reducible', 3) 'oxidisable-organic' and 4) 'resistant' fractions [10].

Prior to all the statistical analyses, an additive logarithmic transformation [$\log_{10}(\text{mean} + 1)$] was performed on all the data to remove the effects of orders of magnitude difference between variables to avoid negative numbers [11]. All data were statistically analysed by using Spearman's correlation analysis, conducted by using STATISTICA for Windows [12].

3. RESULTS AND DISCUSSION

Tables 1 and 2 show the r values based on the concentrations of Cd, Cu and Zn and their four geochemical fractions in the surface sediments for polluted and unpolluted lakes at Kelana Jaya, respectively. There are more number of significant ($P < 0.05$) r values (50 out of 105) found for polluted lake at Kelana Jaya (Table 1) while only 32 for unpolluted lake (Table 2). Tables 3 and 4 exhibit the r values based on the concentrations of Cd, Cu, Pb and Zn and their four geochemical fractions, separately so as to see the difference clearly, in the surface sediments for polluted intertidal west coast of Peninsular Malaysia and unpolluted offshore in the Straits of Malacca, respectively. For the correlations based on metals and their four geochemical fractions in the polluted intertidal sediment (Table 3), it is found that 10 (out of 10 pairwises) for Cu, 7 for Cd, 7 for Pb and 9 for Zn, comparing to lower numbers of r values in the unpolluted offshore sediments: 8 (out of 10 pairwises) for Cu, 3 for Cd, 6 for Pb and 1 for Zn (Table 4). In general, more r values are found in the polluted intertidal sediments than the unpolluted offshore sediments.

Table 1. Spearman's correlation coefficients on the heavy metal concentrations in the total metals and their four geochemical fractions in the surface sediments of polluted lake at Kelana Jaya (Lake 1) (N= 9). Values in bold are significantly correlated at $p > 0.05$

	CdTOT	CuTOT	ZnTOT	CdF1	CuF1	ZnF1	CdF2	CuF2	ZnF2	CdF3	CuF3	ZnF3	CdF4	CuF4	ZnF4
CdTOT	1.00	-0.93	-0.92	0.87	-0.72	-0.19	0.88	-0.88	-0.82	0.96	-0.95	-0.80	0.95	-0.86	-0.88
CuTOT		1.00	0.99	-0.76	0.90	-0.12	-0.73	0.91	0.96	-0.89	0.99	0.95	-0.97	0.98	0.97
ZnTOT			1.00	-0.74	0.88	-0.17	-0.74	0.85	0.97	-0.91	0.99	0.96	-0.96	0.98	0.98
CdF1				1.00	-0.54	-0.45	0.88	-0.77	-0.59	0.81	-0.81	-0.59	0.80	-0.67	-0.65
CuF1					1.00	-0.44	-0.38	0.83	0.95	-0.62	0.86	0.96	-0.87	0.95	0.90
ZnF1						1.00	-0.50	0.08	-0.38	-0.16	-0.04	-0.38	0.05	-0.27	-0.25
CdF2							1.00	-0.65	-0.56	0.92	-0.79	-0.53	0.73	-0.61	-0.66
CuF2								1.00	0.82	-0.72	0.89	0.81	-0.92	0.87	0.86
ZnF2									1.00	-0.79	0.93	0.98	-0.92	0.99	0.98
CdF3										1.00	-0.91	-0.77	0.87	-0.82	-0.85
CuF3											1.00	0.93	-0.96	0.97	0.95
ZnF3												1.00	-0.90	0.99	0.95
CdF4													1.00	-0.94	-0.94
CuF4														1.00	0.97
ZnF4															1.00

Note: TOT= total metal concentrations; F1= easily, freely, leacheable or exchangeable; F2= acid-reducible; F3= oxidisable-organic; F4= resistant fractions.

All the data were cited from Ismail et al. (2004) for the analysis of correlation coefficients in the present study [5].

Table 2. Correlation coefficients on the heavy metal concentrations in the total metals and their four geochemical fractions in the surface sediments of unpolluted lake at Kelana Jaya (Lake 3) (N= 9). Values in bold are significantly correlated at $p > 0.05$

	CdTOT	CuTOT	ZnTOT	CdF1	CuF1	ZnF1	CdF2	CuF2	ZnF2	CdF3	CuF3	ZnF3	CdF4	CuF4	ZnF4
CdTOT	1.00	0.77	-0.01	-0.34	-0.73	0.01	-0.12	-0.81	0.81	-0.23	0.45	0.27	0.89	0.40	-0.51
CuTOT		1.00	0.48	0.13	-0.47	0.51	-0.45	-0.93	0.94	0.31	0.78	0.70	0.71	0.32	0.00
ZnTOT			1.00	0.74	0.44	0.99	-0.77	-0.41	0.53	0.95	0.64	0.95	-0.09	-0.31	0.83
CdF1				1.00	0.64	0.77	-0.23	0.04	0.17	0.83	0.56	0.58	-0.28	-0.27	0.65
CuF1					1.00	0.39	-0.12	0.40	-0.35	0.60	0.09	0.20	-0.74	-0.53	0.72
ZnF1						1.00	-0.75	-0.41	0.55	0.95	0.66	0.95	-0.07	-0.26	0.79
CdF2							1.00	0.45	-0.49	-0.63	-0.30	-0.78	0.06	0.38	-0.63
CuF2								1.00	-0.94	-0.25	-0.74	-0.65	-0.69	-0.35	0.02
ZnF2									1.00	0.35	0.80	0.73	0.72	0.21	0.00
CdF3										1.00	0.57	0.85	-0.26	-0.23	0.88
CuF3											1.00	0.73	0.39	0.13	0.24
ZnF3												1.00	0.15	-0.09	0.67
CdF4													1.00	0.37	-0.56
CuF4														1.00	-0.44
ZnF4															1.00

Note: TOT= total metal concentrations; F1= easily, freely, leacheable or exchangeable; F2= acid-reducible; F3= oxidisable-organic; F4= resistant fractions.

All the data were cited from Ismail et al. (2004) for the analysis of correlation coefficients in the present study [5].

Table 3. Correlation coefficients on the heavy metal concentrations in the total metals and their four geochemical fractions in the surface sediments of polluted intertidal sediments of the west coast of Peninsular Malaysia. Values in bold are significant at $P < 0.05$. N= 26

	F1Cu	F2Cu	F3Cu	F4Cu	Total Cu
F1Cu	1.00	0.51	0.98	0.98	0.99
F2Cu		1.00	0.49	0.45	0.48
F3Cu			1.00	0.97	0.99
F4Cu				1.00	0.99
Total Cu					1.00
	F1Cd	F2Cd	F3Cd	F4Cd	Total Cd
F1Cd	1.00	0.35	0.41	0.66	0.78
F2Cd		1.00	0.76	-0.02	0.70
F3Cd			1.00	0.00	0.72
F4Cd				1.00	0.64
Total Cd					1.00
	F1Pb	F2Pb	F3Pb	F4Pb	Total Pb
F1Pb	1.00	0.48	0.42	0.65	0.68
F2Pb		1.00	0.06	0.20	0.24
F3Pb			1.00	0.54	0.81
F4Pb				1.00	0.92
Total Pb					1.00
	F1Zn	F2Zn	F3Zn	F4Zn	Total Zn
F1Zn	1.00	0.84	0.50	0.61	0.75
F2Zn		1.00	0.71	0.72	0.87
F3Zn			1.00	0.77	0.89
F4Zn				1.00	0.94
Total Zn					1.00

Note: F1= EFLE, F2= acid-reducible, F3= oxidisable-organic and F4= resistant.

All the data were cited from Yap et al. (2002) for Cu and Pb, Yap et al. (2003) for Pb and Yap et al. (2005) for Zn, for the analysis of correlation coefficients in the present study [6]-[9].

Table 4. Correlation coefficients on the heavy metal concentrations in the total metals and their four geochemical fractions in the surface sediments of unpolluted offshore sediments of the Straits of Malacca. Values in bold are significant at $P < 0.05$. $N = 31$

	F1Cu	F2Cu	F3Cu	F4Cu	Total Cu
F1Cu	1.00	0.18	0.38	0.59	0.64
F2Cu		1.00	0.29	0.38	0.41
F3Cu			1.00	0.43	0.62
F4Cu				1.00	0.98
Total Cu					1.00
	F1Cd	F2Cd	F3Cd	F4Cd	Total Cd
F1Cd	1.00	-0.16	0.30	0.01	0.23
F2Cd		1.00	0.05	0.15	0.39
F3Cd			1.00	0.13	0.74
F4Cd				1.00	0.69
Total Cd					1.00
	F1Pb	F2Pb	F3Pb	F4Pb	Total Pb
F1Pb	1.00	0.32	0.41	0.21	0.47
F2Pb		1.00	0.07	0.38	0.39
F3Pb			1.00	0.25	0.79
F4Pb				1.00	0.78
Total Pb					1.00
	F1Zn	F2Zn	F3Zn	F4Zn	Total Zn
F1Zn	1.00	-0.10	0.30	0.18	0.30
F2Zn		1.00	-0.31	-0.20	-0.31
F3Zn			1.00	-0.18	0.21
F4Zn				1.00	0.92
Total Zn					1.00

Note: F1= EFLE, F2= acid-reducible, F3= oxidisable-organic and F4= resistant.

All the data were cited from Yap et al. (2002) for Cu and Pb, Yap et al. (2003) for Pb and Yap et al. (2005) for Zn, for the analysis of correlation coefficients in the present study [6]-[9].

Table 5: Comparisons of numbers of significant correlation coefficients (r values) between polluted and unpolluted sediments reported in the literature

No.	Areas	Metals ranges (all are presented in $\mu\text{g/g}$ dry weight except indicated in %).	Status	Number of significant correlation coefficients	References
1.	Dumai coast, Indonesia	Cd: 0.46–1.89; Cu: 1.61–13.84; Pb: 14.63–84.90; Zn: 31.49–87.11; Ni: 7.26–19.97.	Relatively unpolluted.	N= 69. 10 out of 10 at $P < 0.01$.	Amin et al. (2009)
2.	Xiamen Bay	Cu: 19-97; Pb: 45-60; Zn: 65-223; Cd: 0.11-1.01; Cr: 37-134; Ni: 25-65; Fe: 3.08-4.81%.	Relatively unpolluted.	N= 9. 3 out of 21 at $P < 0.05$.	Zhang et al. (2007)
3.	Kaohsiung Harbor (Taiwan)	Hg: 0.1-8.5; Pb: 9.5-470; Cd: 0.1-6.8; Cr: 0.2-900; Cu: 5-946; Zn: 52-1369.	Polluted	N= 48-90. 15 out of 15 at $P < 0.01$.	Chen et al. (2007)
4.	Adyar estuary, India	Zn: 60-168; Cr: 225-318; Pb: BDL-11; Ni: 379-782; Co: 2-11.	Relatively unpolluted.	N= 12. 2 out of 10 at $P < 0.05$.	Achyuthan and Richardmohan (2002)
5.	Muttukadu, India	Zn: 50-125; Cr: 16-73; Pb: 1-3; Ni: 25-94; Co: 7-10.	Relatively unpolluted.	N= 9. 2 out of 10. at $P < 0.05$.	Achyuthan and Richardmohan (2002)
6.	Marakkanam, India.	Zn: BDL-94; Cr: BDL-61; Pb: BDL-15; Ni: BDL-23; Co: BDL-9.	Relatively unpolluted.	N= 12. 8 out of 10 at $P < 0.05$.	Achyuthan and Richardmohan (2002)
7.	Mamallapuram, India	Zn: 52-123; Cr: 62-108; Ni: 49-122; Pb: 9-15; Co: 7-12.	Relatively unpolluted.	N= 12. 0 out of 10.	Achyuthan and Richardmohan (2002)
8.	Lan-yang River, Taiwan	Cd: 0.04-0.42; Cr: 32-140; Pb: 15.3-53.7; Cu: 14-48.4; Ni: 21-67.5; Zn: 61.0-204.	Relatively unpolluted.	N= 20. 12 out of 15 at $P < 0.05$.	Chang et al. (2007)
9.	Erh-jen River, Taiwan	Cd: BDL-0.33; Cr: 29.9-687; Pb: 16.2-84.3; Cu: BDL-237; Ni: 14.1-231; Zn: 81.4-1230.	Relatively polluted.	N= 22. 6 out of 15. at $P < 0.05$.	Chang et al. (2007)
11.	Paddy soils, China	Cd: BDL-0.90; Co: 0.74-15.7; Cr: 19.1-90.6; Cu: 3.02-43.9; Ni: 4.54-32.7; Pb: 15.9-49.9; Zn: 18.5-107.	Unpolluted.	N= 16. 16 out of 21 at $P < 0.05$.	Wong et al (2002)
13.	Lagoon of Venice (Italy)	Cd: 0.2-2.3; Cu: 7.5-58.8; Ni: 6.7-22.2; Pb: 7.3-53.6; Zn: 35-463.	Polluted	N= 13. 9 out of 10. at $P < 0.05$.	Coccioni et al. (2009)
	Lake Shinji, southwestern Japan.	As: 3-14; Pb: 11-39; Zn: 16-201; Cu: 3-43; Ni: 3-40.	Semi-polluted	N= 51. 6 out of 10 at $P < 0.05$.	Ahmed et al. (2010)
	River Ohashi, southwestern Japan.	As: 5-8; Pb: 16-23; Zn: 42-71; Cu: 8-28; Ni: 8-13.	Unpolluted	N= 4. 7 out of 10 at $P < 0.05$.	Ahmed et al. (2010)
14.	Sediments at Kelana Jaya Lakes (Cd, Cu and Zn)	Cd: 0.39-2.94; Cu: 6.84-76.2; Zn: 102.2-532.2.	Polluted	N= 9. 50 out of 105 at $P < 0.05$.	Ismail et al. (2004)
15.	Sediments at Kelana Jaya Lakes (Cd, Cu and Zn)	Cd: 0.71-1.98; Cu: 7.78-13.1; Zn: 51.3-166.5.	Unpolluted	N= 9. 32 out of 105 at $P < 0.05$.	Ismail et al. (2004)
16.	Marine sediments on four geochemical fractions	Cu: 0.40- 314.8; Cd: 0.03-1.98; Pb: 0.96- 69.8; Zn: 3.12-306.2.	Polluted	N= 26. $P < 0.05$. Cu: 10 out of 10. Cd: 7 out of 10. Pb: 7 out of 10. Zn: 9 out of 10.	Yap et al. (2002, 2003; 2005).
17.	Marine sediments on four geochemical fractions	Cu: 0.25- 13.82; Cd: 0.10-1.42; Pb: 3.59- 25.36; Zn: 4.00-79.05.	Unpolluted	N= 31. $P < 0.05$. Cu: 8 out of 10. Cd: 3 out of 10. Pb: 6 out of 10. Zn: 1 out of 10.	Yap et al. (2002, 2003; 2005).

Note: BDL= below detection limit

Discussion

To support the present proposal on the use of r -value as a potential indicator of polluted sediments, data and number of significant r values from eight publications were reviewed and compared, as presented in Table 5. First, Chen et al. (2007) finding's on the polluted Kaohsiung Harbor (Taiwan) also generated all (15) significant ($P < 0.01$) based on Spearman's correlation coefficients for concentrations of Hg, Pb, Cd, Cr, Cu and Zn in the sediments [13]. Their results also showed that these metals were strongly interrelated ($p < 0.01$) with correlation coefficients ranging from 0.37 to 0.83 at the 99% confidence level. Besides, Coccioni et al. (2009) reported the 5 metal concentrations in the polluted Lagoon of Venice (Italy) and found 9 out of 10 significant pairwises [14].

Zhang et al. (2007) performed Pearson's correlation coefficient matrix among Cu, Pb, Zn, Cd, Ni, Cr, Fe and TOC concentrations in sediments of western Xiamen Bay [15]. They found that out of 21 correlation coefficients, only three were significantly ($P < 0.05$) correlated. The metal ranges ($\mu\text{g/g}$ dry weight, except for Fe) for western Xiamen Bay and adjacent Maluan Bay and Yuandang Lagoon varied from 19-97 for Cu, 45-60 for Pb, 65-223 for Zn, 0.11-1.01 for Cd, 37-134 for Cr, 25-65 for Ni and 3.08-4.81% for Fe. All the metal concentrations in sediments met Chinese National Standard Criteria for Marine Sediment Quality (Zhang et al., 2007) and therefore were considered as relatively unpolluted sediments [15]. However, Zhang et al. (2007) found that, although not statistically significant, metal contaminants (Cd, Cr and Ni) show negative correlation with Fe, suggesting these heavy metal concentrations are not strongly controlled by natural weathering processes [15]. Zhang et al. (2007) also found significant correlation between Ni and Cr ($r = 0.749$) indicate Ni and Cr source contamination and significant correlation between Zn and Cu could suggest Zn and Cu source input [15]. This indicated that the strong r value between any two metals investigated could be explained by their similar source input as well as contamination. However, unpolluted sediments from all the above literature were not included in their study. Thus, making the comparison between polluted and unpolluted from similar areas is not possible.

On the other hand, the use of r value as indicator of polluted ecosystem could sometimes be masked by the wide metal ranges found in the sediments. For example, the r values, reported by Achyuthan and Richardmohan (2002), based on Zn, Cd, Pb, Ni and Co in the sediments collected from four relatively unpolluted coasts: Adyar estuary, Muttukadu coast, Marakkanam coast and Mamallapuram coast [16]. However, out of 10 pairwises, the significant r values for the four coasts varied from 2 for Adyar, 2 for Muttukadu, 8 for Marakkanam and 0 for Mamallapuram. These r values cannot be confidently used to indicate the pollution status nor the high metal concentrations at Marakkanam. This was due to all the 5 metals ranged from BDL (below detection limit). For the statistical purpose, the authors put the detection limits for all the 5 metals which were the lowest values. This could potentially raise the difference between the minimum and maximum values and influenced the r values finally generated using the correlation analysis. Another reverse trend was observed in the unpolluted Lan-yang River and polluted Erh-jen River (Taiwan) as reported by Chang et al. (2007) [17]. The maximum values for Cr (687), Pb (84.3), Cu (237), Ni (231) and Zn (1230) at Erh-jen River were evidently polluted sediments but the correlation analysis based on 6 metals exhibited only 6 out of 15 pairwises being significantly ($P < 0.05$) correlated, when compared to the relatively unpolluted Lan-yang River in which there were 12 out of 15 pairwises being significantly ($P < 0.05$) correlated. However, this time, the influence by the wide metal ranges are not strongly evidenced.

Wong et al (2002) reported 7 metal concentrations in the unpolluted paddy soils and found 16 out of 21 pairwises as being significant ($P < 0.05$) [18]. However, it was difficult to assess whether number of r values can be used as indicator of polluted soils since they also studied on the unpolluted crop soils and natural soils in which both type of soils also showed almost similar number of r values (15 out of 21).

The study by Amin et al. (2009) on the relatively unpolluted sediments based on Cd, Cu, Pb, Ni and Zn also showed little evidence since 10 out of 10 pairwises were significantly ($P < 0.01$) correlated [19]. Ahmed et al. (2010) also reported the metal concentrations in Lake Shinji and River Ohashi, in which the former was more polluted than the latter [20]. However, based on correlation coefficients in the selected 5 metals (As, Pb, Zn, Cu and Ni), the number of significant number of pairwises were 6 and 7 out of 10 for Lake Shinji and River Ohashi, respectively. This indicated that the use of the number of r value in evaluating the pollution degree of an ecosystem should be exercised with cautions.

Statistical calculations that neglect correlations often result in incorrect results and erroneous conclusions [21]. However, Asuero et al. (2006) argued that a measure of statistical relationship, such as a correlation coefficient should never be used to deduce a causal connection while the ideas on causation must come from outside statistics [3].

It should be noted that, besides the sampling is being unbiasedly selected, caution should be exercised that the sampling of the environmental parameters should be properly designed and should cover the whole of ecosystem, ranging from effluent-receiving point sources to the clean sites far away from the sources. Hence, this can truly reflect the overall environmental quality of the ecosystem. Perhaps, with all the above

criteria being considered, the use of r values can be potentially used as indicator of environmental quality. Another point to be cautious of is the spurious correlations in which the observed correlation between two variables might be due to the action of a third, unobserved variable which is not included in the study. Hence, the low number of r values in the metal-polluted ecosystem could be due to other unrecorded and unstudied biotic and abiotic factors.

4. CONCLUSION

The present review does points to the potentials of the number of r values in comparison between polluted and unpolluted ecosystem, as indicator of the pollution. Although some reported studies did support the present idea, some reported studies did not support the use of number of r values and this indicated the presence of third factor which was not included in the study. Therefore, further investigations are still necessary before number of significant r values can be confidently employed as indicator of metal-polluted ecosystem.

ACKNOWLEDGEMENTS

This study was partly supported by the Research University Grant Scheme (RUGS), [Pusat Kos: 91986], provided by Universiti Putra Malaysia.

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