

A Study of Coastal Sediments of Sindh Pakistan Using Geochemical Approach for Evaluation of Heavy Metals Pollution

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Abstract: Twenty one locations in the Coastal area of Sindh, Pakistan were chosen with the purpose of study the accessibility of heavy metals and their concentrations in the surficial sediments ($<60\mu\text{m}$). These soil samples were analyzed for Fe (Iron), Mn (Magnese), Cr (Chromium), Pb (Lead), Zn (Zinc), Cu (Copper), Co (Cobalt) and As (Arsenic) to examine metal concentrations in sediments. The concentration of metals Fe, Mn, Cr, Pb, Zn, Cu, Co and As were determined by atomic absorption spectrophotometer. Evaluation of anthropogenic pollution in sediments, Enrichment Factor (EF) and Index of Geoaccumulation (I_{geo}) are calculated. The concentration of cadmium is demonstrating elevated contamination in all the sampling sites. The observations suggested high EF values of the coastal sediments are polluted by Mn and Zn acts as a sink for heavy metals contributed from a multitude of anthropogenic sources in the study area possibly displays the effluent discharge. EF data and Geoaccumulation index values (I_{geo}) indicated that these trace metals predominantly originates from anthropogenic sources. Considering the calculated EF and I_{geo} values, the increased I_{geo} values in south and southeastern part of the study areas are attributed principally to anthropogenic activities.

Key words: Enrichment Factor • Index of Geoaccumulation • Anthropogenic activities

INTRODUCTION

Trace elements found in soils/sediments are immobilized in water and as a result could be concerned in with absorption, co-precipitation and complex formation [1, 2]. Occasionally they are co-adsorbed with further elements as oxides, hydroxides of Fe, Mn, or may occur in particulate form [3,4]. Their concentrations in streams and coastal sediment compartments can be used to reveal the history and concentration of local and regional pollution [5-8]. In the study area, heavy metals penetrate into aquatic ecosystems mostly from anthropogenic sources, such as industrial wastewater discharges, sewage wastewater and fossil fuel combustion [9]. It has been observed worldwide that the impact of anthropogenic perturbation is most powerfully felt by estuarine and coastal environments nearby to the study areas [10].

In view of collection of soil samples for trace metals in the study area has been completed to serve up a basis for the preparation of control strategies to accomplish

enhanced ecological excellence and will as a key for an efficient management of soil quality; comparable widespread investigations of coastal soils have been carried out newly in many countries [11]. These soil samples were analyzed for Fe, Mn, Cr, Pb, Zn, Cu, Co and As to scrutinize metal concentrations in sediment. Therefore, the study will endeavor to evaluate the extent of heavy metal contamination from the surface to the bottom sediments and the degree to which heavy metals are influenced. The interrelationships among elements and Enrichment Factor (EF), geoaccumulation index values (I_{geo}) is calculated to differentiate the origin of metals between anthropogenic and geogenic sources.

MATERIALS AND METHODS

Sediment Sampling and Chemical Analysis: A total twenty one samples of coastal sediments from same number of sites were collected during 2011-2012 (Figure1) using a hand trowel. Watindo stream was chosen to

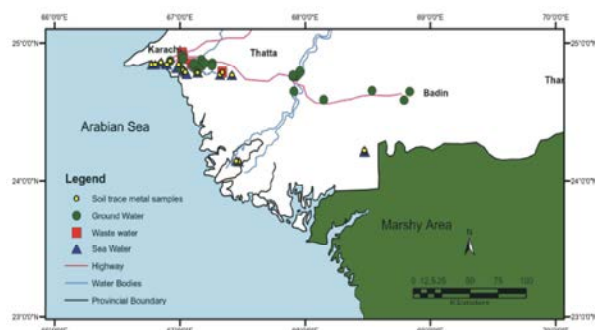


Fig. 1: Map of the study area.

be outside the study area and therefore regarded as unpolluted. The samples were placed in Ziploc bags and transported to the laboratory. The hand trowel was washed with a detergent, rinsed and dried before each use so as to minimize contamination. Sediment samples were dried in an oven at 105 °C overnight, sieved mechanically using a 0.5 mm sieve, homogenized and ground to 0.063 mm fine powder because metals are known to adhere to fine particles. After which, 1.25 g of each sample was digested with 20 ml aqua regia (HCl/HNO₃ 3:1) in a beaker (open-beaker digestion) on a thermostatically controlled hot plate. The digest were heated to near dryness and cooled to ambient temperature. Then 5.0 ml of hydrogen peroxide was added in parts to complete the digestion and the resulting mixture heated again to near dryness in a fume cupboard. The beaker walls were washed with 10 ml of de-ionised water and 5 ml HCl were added, mixed and heated again. The resulting digest was allowed to cool and transferred into a 50 ml standard flask and made up to the mark with de-ionised water. Pb, Co, Cr, Cu, Zn, Mn and Fe heavy metal elements were then analyzed by direct aspiration of the sample solution into a Perkin- Elmer model 2380 flame atomic absorption spectrophotometer (AAS). All metals were analysed using lean-blue air acetylene flame at wavelength 324.8 nm, slit width 0.2 nm and sensitivity check of 5.0 mg/L Cu; wavelength 228.8 nm, slit width 0.7 and sensitivity check of 2.0 mg/l Cd; wavelength 213.9 nm, slit size 0.7 nm and sensitivity check 1.0 mg/L Zn; wavelength 217.0 nm, slit width 0.7 nm and sensitivity check 9.0 Pb and wavelength 279.5 nm, slit width 0.2 nm and sensitivity check 2.5 mg/L Mn. Sediment pH was measured in a suspension of 1:2 sediment to water ratio using a calibrated pH meter (WE-30200). Accuracy of the analytical method was evaluated by comparing the expected metal concentrations in certified reference materials with the measured values. Simultaneous performance of analytical blanks, standard reference

(JG- 3) (12) and calculation of the average recoveries of heavy metals confirmed that the accuracy of method was within acceptable limits.

According to Ergin *et al.*, (1991)[13], the Enrichment factor (EF) is calculated (Table 1) by using this formula:

$$EF = \frac{(Fe/M)_{\text{sample}}}{(Fe/M)_{\text{Background}}}$$

Where Fe is the content of Fe in sample and M is the content of the metal in the sample.

And the geoaccumulation index (I_{geo}) introduced by Muller (1979) and calculated by using this formula:

$$I_{geo} = \log_2 (C_n / 1.5 \times B_n)$$

Where,

- C_n - measured concentration of heavy metal in the sediment,
- B_n - geochemical background value in average shale of element 'n'.
- 1.5 is the background matrix correction in factor due to lithogenic effects

RESULT AND DISCUSSION

The main intention of sediment quality guidelines (SQGs) is to protect aquatic biota from the damaging and poisonous effects related with to sediments bound contaminants and is a functional tool for evaluating potential for contaminants inside sediment to induce biological effects. The source of pollution is determined through the normalization of geoaccumulation values to the reference element. The degree of pollution in sediments can be assessed by determining the enrichment factor and indices such as Geo-accumulation index.

Enrichment Factor: The enrichment factor (EF) is a suitable measure of geochemical trends and is used for making comparisons between areas. Because of the natural basis of metal elements, the gross concentrations of metal elements don't demonstrate the anthropogenic contribution specifically. The evaluation of heavy metals from anthropogenic contribution must be made clear. The natural incidence of heavy metals complicates assessments of potentially contaminated estuarine sediments; measurable quantities of metals do not

Table 1: Enrichment factor values of trace metals for coastal sediments

Sample No.	Name of Locations	Cr	Cu	Fe	Mn	Pb	Co	Zn	As
1	Qazi Muhammad, 1Runn of Kutch	0	0	1	10	1	0	6	1
2	Qazi Muhammad, 2 Runn of Kutch	0	0	1	7	1	0	5	1
3	Main keti Bandar (East)	1	1	1	2	1	1	1	1
4	Keti bandar 1	1	1	1	2	1	1	1	1
5	Keti bandar 2	1	0	1	2	0	1	1	1
6	Port Qasim JT	1	1	1	1	1	1	1	1
7	Steel mill, Bin Qasim	1	1	1	1	1	1	1	1
8	Mazar Russian Beach, Back side of Steel Mill, Bin Qasim	1	1	1	1	1	1	1	1
9	Ibraheem Hyderi Korangi	1	1	1	3	1	1	1	1
10	Sea View, Hyper Star Building, Clifton	1	1	1	20	1	1	3	1
11	Sea View, Floating Ship, Clifton	1	1	1	21	1	1	3	1
12	Main Manora	2	1	1	15	1	1	25	1
13	Manora beach 1	2	1	1	13	1	1	20	1
14	Manora beach 2	1	1	1	13	1	1	22	1
15	Native Jeti Bridge, Left Side, Kemari	1	1	1	14	1	1	22	1
16	Main Sandspit	1	1	1	22	1	1	3	1
17	Main Hawks bay	1	1	1	12	1	1	2	1
18	Hawaks Bay 1	1	1	1	13	1	1	2	1
19	Hawaks Bay 2	1	1	1	13	1	1	2	1
20	Paradise Point 1	1	1	1	10	1	1	2	1
21	Paradise Point 2	1	1	1	11	1	1	2	1

automatically infer anthropogenic enrichment in the estuary. The enrichment factor is a good instrument to discriminate the metal source between anthropogenic and naturally occurring [14]. EF is calculated to determine the level of metals in sediments of the coastal area of the province and its nearby aquatic environment are of anthropogenic origins (e.g., contamination) shown in Table 1.

Differentiating the elements originating from human behavior and those from natural weathering is an essential part of geochemical studies. One such technique largely applied is 'normalization' where metal concentrations were normalized to a textural or compositional characteristic of sediments. To identify anomalous metal concentration, geochemical normalization of the heavy metals data to a conservative element, such as Al, Fe and Si was employed. Several authors have successfully used iron to normalize heavy metals contaminants [15]. In this study, iron is also used as a conservative tracer to differentiate natural from anthropogenic components. EF values were interpreted as recommended by Birch (2003) [16] where EF < 1 indicates no enrichment, < 3 is minor; 3-5 is moderate; 5-10 is moderately severe; 10-25 is severe; 25-50 is very severe; and > 50 is extremely severe. A value of $0.5 \leq EF \leq 1.5$ suggests that traces of metal may be due to crustal materials or natural weathering processes. Samples having EF value greater than 5 are considered to be contaminated with that particular element.

From table no. 1 (EF) represents the EF values of all the trace metals measured in coastal sediments. In the industrial and agricultural waste area all the sampling sites have EF values are ≤ 2 shows minor enrichment except at site 1 and 2 for Mn & Zn. In the same way at industrial and municipal waste fishing harbor sampling site EF values are also ≤ 2 showing minor enrichment at all the sampling sites except for Mn and Zn. Similarly EF values at beach were also ≤ 2 shows minor enrichment except for Mn. EF values < 5 are considered noteworthy. Areas with EF values < 1 should be viewed with caution as they imply preferential release of these metals, making them bioavailable. If the value of EF > 1.5, the trace metal is delivered from others sources suggesting environmental contamination by that particular trace element. It is recommended that high EF values indicate an anthropogenic source of trace metals, mostly from activities such as industrialization, urbanization, deposition of industrial values and others. Since, the bioavailability and toxicity of any trace metals in sediments depend on the chemical form and concentration of the metal, it can be inferred that trace metals in sediment samples with high EF values, along with higher labile fractions in sediments are potential sources for mobility and bioavailability in the aquatic ecosystems. In the study area minimum EF obtained for the elements (Cr, Cu, Fe, Pb, Co, Zn and As) are less than unity in most of the

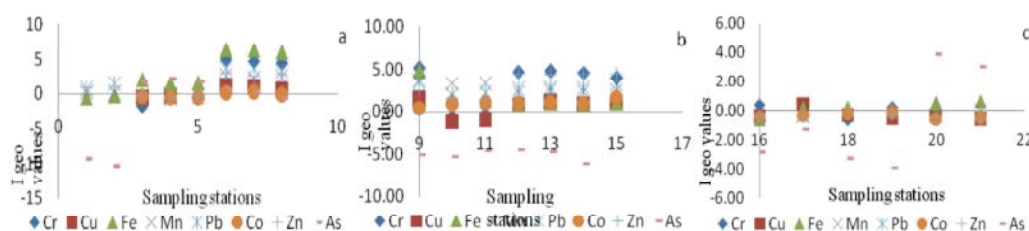


Fig. 1: Igeo values of different sampling sites at coastal belt of Sindh.

a. Industrial and agriculture wastes surface sediments

b. Industrial and municipal wastes (at fishing harbor) surface sediments

c. Beach / creek surface sediments

samples implying that these elements are exhausted in some of the phases relative to crustal profusion in the study area.

Subsequent the understanding of [16] shows no enrichment with respect to Cr, Cu, Fe Pb, Co and As in the study area, although high EF value of Mn and Zn shows severe enrichment. EF (<1) in most of these metals in the sediment indicate its origin predominantly from lithogenous material and suggest the absence of contamination by these metals in the study region. Approximately at all sites, Mn and Zn indicate harsh enrichment particularly in south and south west sampling stations suggesting the anthropogenic input of these metals. The interpretation propose that the coastal sediment is polluted by Mn and Zn acts as a sink for heavy metals contributed from a multitude of anthropogenic sources. High EF values of Mn and Zn in the study area possibly displays the waste matter ejection of nearby chemical industries (fertilizers, heavy metal processing, pesticides and insecticides, petrol refining, chemical and allied industries) and municipal activities through Indus River. In the south and south east estuary industrial effluents, flow pattern of Indus river and synchronous tides lead to high deposition of Mn in sites (9-10) and Zn in points (1, 2, 12-15) whereas agricultural and domestic activities influence major portion in the south eastern part of the estuary makes less pollution compared with the south and south west.

Geoaccumulation Index: The geoaccumulation index (Igeo) introduced by Muller (1979) [17] for metal concentrations in the < 2 μ m fraction and developed for the global standard shale values. This is used here to evaluate metal pollution in sediments of the coastal area of the province.

Different Geoaccumulation index classes along with the associated sediment quality are given in Table 2.

Table 2: Geoaccumulation index classes to assess sediment quality

0-0	< 0	Uncontaminated
0-1	1	Uncontaminated to moderately contaminated
1-2	2	Moderately contaminated
2-3	3	Moderately to highly contaminated
3-4	4	Highly contaminated
4-5	5	Highly to very highly contaminated
5-6	6	Very highly contaminated

The I_{geo} class 0 indicates the absence of contamination while the I_{geo} class 6 represents the upper limit of the contamination. The highest class 6 (very strong contamination) reflects 100-fold enrichment of the metals relative to their background values. I_{geo} calculation is carried out for each functional area independently for the coastal sediments of the study area. The calculated values for coastal sediments and its surrounding marine environment are given in Appendix 1 and their model representations are in Fig. 1 (a, b & c).

The values for Cr and Fe in the coastal sediments acquired from industrial and agricultural waste area exhibited class I_{geo} 4-5 and I_{geo} 6-7 and hence they are very highly contaminated with these metals. From Fig.1 (a) it can be interpreted that sampling stations from the industrial area of the Port Qasim may face a severe trace metal contamination problem with respect to Cr and Fe, while other trace metals such as Pb, Mn, Zn are moderately contaminated and As and Cu has not shown any significant impression of pollution i.e., uncontaminated.

The functional area pertaining to industrial and municipal waste (fishing harbor) (Fig. 1b) are somewhat showing similar picture as of industrial and agricultural waste area. Here Cr and Zn exhibited in I_{geo} class 4-5 highly polluted, Mn and Pb exhibited in I_{geo} class 2-3 moderately.

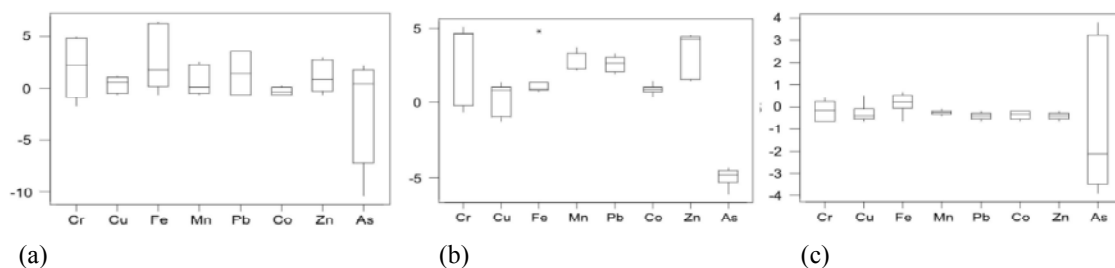


Fig. 2: Box-and-whisker plots of the geoaccumulation index (a) Industrial and agriculture wastes surface sediments. (b) Industrial and municipal wastes (at fishing harbor) surface sediments. (c) Beach / creek surface sediments

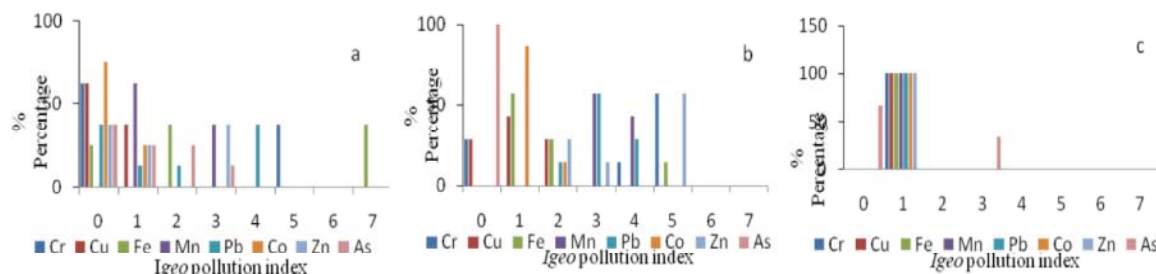


Fig. 3: Percentage of Igeo pollution index of different sampling sites. a. Industrial and agriculture wastes surface sediments. b. Industrial and municipal wastes (at fishing harbor) surface sediments. c. Industrial and municipal wastes (at fishing harbor) surface sediments

Polluted sediments that could be attributed to terrigenous sources, Cu and Co exhibited in I_{geo} class 0-1 unpolluted, while the Arsenic has not demonstrated some marked contamination in this functional area. While the sediments collected from the beach / creek (Fig.1c) does not show any significant impact of contamination with any trace metal studied.

The I_{geo} values for Arsenic fall in class '0' in most of the samples collected, indicating background concentration in all the sites at in different functional areas are not polluted with this metal. Due to the varying quality of sediment and local contamination I_{geo} values of Fe, Cr, Zn, Pb and Mn fall into five classes 6,5,4,3 and 2 indicating moderate to very high contamination which is attributed to the sludge, sewage plant, wastewater discharge and industrial effluent.

Figure 2 shows the range and average values of I_{geo} values for each metal, using local reference material in the form of box and whisker plot. Based on average values of I_{geo} , the ranking of intensity of heavy metal pollution of the functional area of industrial and agriculture wastes surface sediments are as follows: $Fe > Cr > Pb > Zn > Mn > Cu > Co > As$. Similarly the I_{geo} , ranking of intensity of heavy metal pollution of the functional area of industrial and municipal wastes (at fishing harbor) surface

sediments are as under: $Cr > Fe > Zn > Pb > Mn > Co > Cu > As$. In the same way I_{geo} , ranking of intensity of heavy metal pollution of the functional area beach/creek surface sediments are mentioned below: $As > Fe > Cu > Cr > Mn > Co > Pb > Zn$

From Figure 3, functional area belongs to industrial and agriculture waste can be displays sample percentages in Muller classes for Cr, Cu, Fe, Mn, Pb, Co, Zn and As. Co, Cr, Cu Pb, Zn and As concentrations fall mainly in classes '0'. For Cr mainly fall in class '0' (62.5% of total samples) and the remainder 37.5% in class 6. Cu mainly fall in class '0' (62.5%) and the remainder 37.5% in class 1. Fe fall in class '0' (25%) and the remainder 37.5% in class 6 and 7 each. For Mn mainly fall in class 1 (62.5%) and the remainder 37.5% in class 4. Pb samples mainly fall in class.

Likewise functional area of industrial and municipal waste (fishing harbor) displays that As concentrations fall mainly in classes '0'. For Cr mainly fall in class 5 (57.1% of the total sample) and the remainder 28.6% and 14.3% are in class '0' & 4. Cu fall in class '0' (28.6%) and the remainder 42.9% and 28.6 % are in class 1 & 2. Fe mainly fall in class 1 (57.1%) and the remainder 28.6% and 14.3% fall in class 2 & 5. Mn included 57.1% and 42.9% in class 3 and 4. Pb mainly falls in class 3 (57.1%) while the remainder 14.3% and 28.6% fall in class 2 and 4. Co fall

85.7 % of total samples in class 1 and 14.3% are in class 2. For Zn, 28.6% of the samples fall in class 2, 14.3% in class 3 and 57.1% in class 5 and As 100% fall in class '0'.

These results indicate that the surface sediments of this functional area can be categorized as follows: unpolluted with Cr, Cu and As (average $I_{geo} < 0$), unpolluted to moderately polluted with Cu, Fe and Co ($0 < \text{average } I_{geo} < 1$), moderately polluted with Cu, Fe, Pb, Co and Zn ($1 < \text{average } I_{geo} < 2$), moderately to highly polluted with Mn, Pb and Zn ($2 < \text{average } I_{geo} < 3$), strongly polluted with Cr, Mn and Pb ($3 < \text{average } I_{geo} < 4$) and strongly to extremely polluted with Cr, Fe and Zn ($4 < \text{average } I_{geo} < 5$). In the same way surface sediment collected from the functional area of beaches and creeks along the coastal belt of the province demonstrates that 66.6% and 33.3% of the samples fall in class '0' and 3. While the rest of the trace metal (100% of total samples) fall in class 1 i.e. unpolluted to moderately polluted ($0 < \text{average } I_{geo} < 1$).

Fig. 3 reflects that, independent of the background used in three different functional area such as industrial and agricultural waste area, industrial and municipal waste (fishing harbor) and beach / creek area having 40%, 100% and 70% of the samples fall in Class 0 (background concentrations) especially for the metals As, i.e., it leads us to conclude that in the study area this metal has not shown any significant impact. While the contamination status in sediments with respect to trace metal depending on mobility and solubility of these metals that can be displaced from discharge places. Considering the calculated EF and I_{geo} values, the increased I_{geo} values in south and south eastern part of the study area are attributed principally to anthropogenic activities. In general terms, the south and south eastern part of the study areas are most affected by pollution, while south western part are unaffected.

CONCLUSION

Enrichment factor has showed no enrichment with respect to Cr, Cu, Fe Pb, Co and As in the study area, although the high EF value of Mn and Zn indicated severe enrichment. High EF values of Mn and Zn in the study area possibly display the effluent discharge of nearby chemical industries (fertilizers, heavy metal processing, pesticides and insecticides, petrol refining, chemical and allied industries) and urban activities through Indus River. Geoaccumulation index reflects that,

independent of the background used in three different functional areas such as industrial and agricultural waste area, industrial and municipal waste (fishing harbor) and beach / creek area having 40%, 100% and 70% of the samples fall in Class 0 (background concentrations) especially for the metals As, i.e., it leads us to conclude that in the study area this metal has not shown any significant impact. While the contamination status in sediments with respect to trace metal depending on the mobility and solubility of these metals that can be displaced from discharge places. EF data, Geoaccumulation index values (I_{geo}) concluded that these trace metals predominately originate from anthropogenic sources.

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Appendix 1: Geoaccumulation index Igeo values were calculated for different metals as introduced by Muller (1969) is as follows:

S. No.	Cr	Cu	Fe	Mn	Pb	Co	Zn	As
A1	0.0	0.0	-0.6	0.2	0.7	0.0	0.8	-9.4
A2	0.0	0.0	-0.2	0.1	1.4	0.0	0.9	-10.4
A3	-1.8	-0.6	2.0	-0.3	-0.6	-0.5	-0.2	1.8
A4	-0.1	-0.3	1.5	-0.6	-0.6	-0.6	-0.6	2.2
A5	-0.6	0.0	1.6	-0.5	0.00	-0.6	-0.3	1.7
A6	5.0	1.2	6.4	2.6	3.6	0.1	3.0	0.6
A7	4.7	0.9	6.3	2.2	3.6	0.3	2.6	0.3
A8	4.5	0.6	6.0	2.4	3.2	-0.1	2.9	-0.6
B09	5.1	1.4	4.8	3.7	3.3	0.4	2.5	-5.1
B10	-0.6	-1.2	1.4	3.3	1.9	0.8	1.6	-5.3
B11	-0.2	-0.9	1.3	3.3	2.1	1.0	1.5	-4.6
B12	4.6	0.8	0.8	2.3	2.6	1.0	4.4	-4.5
B13	4.7	1.1	1.0	2.3	2.9	1.1	4.3	-4.8
B14	4.6	0.9	0.9	2.2	2.7	1.0	4.4	-6.1
B15	3.9	1.0	1.0	2.3	3.1	1.5	4.5	-4.3
C16	0.4	-0.4	-0.6	-0.2	-0.6	-0.4	-0.4	-2.9
C17	0.0	0.5	0.2	-0.4	-0.4	-0.3	-0.5	-1.3
C18	-0.6	-0.4	0.2	-0.2	-0.3	-0.2	-0.2	-3.3
C19	0.2	-0.5	0.1	-0.3	-0.2	-0.2	-0.3	-3.9
C20	-0.3	-0.3	0.5	-0.3	-0.4	-0.6	-0.4	3.8
C21	-0.6	-0.6	0.7	-0.1	-0.5	-0.5	-0.6	3.0