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Application of chemometric analysis and self Organizing Map-Artificial Neural Network as source receptor modeling for metal speciation in river sediment

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ABSTRACT

Present study deals with the river Ganga water quality and its impact on metal speciation in its sediments. Concentration of physico-chemical parameters was highest in summer season followed by winter and lowest in rainy season. Metal speciation study in river sediments revealed that exchangeable, reducible and oxidizable fractions were dominant in all the studied metals (Cr, Ni, Cu, Zn, Cd, Pb) except Mn and Fe. High pollution load index (1.64–3.89) recommends urgent need of mitigation measures. Self-organizing Map-Artificial Neural Network (SOM-ANN) was applied to the data set for the prediction of major point sources of pollution in the river Ganga.

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1. Introduction

Anthropogenic sources (rapid urbanization, industrialization, mining etc.) have become principal sources of high metal concentration in water and sediments (Suthar et al., 2009). Metals entering into the water body transported from one place to another and join sediments as the ultimate sink. Metals degrade naturally at a very slow rate and ultimately get accumulated in flora and fauna through food chain (Christensen, 1995; Jarup, 2003; Mathew et al., 2003; Goyer, 2004; Jain, 2004; Kelepertzis et al., 2012; Kumar and Chopra, 2013; Rossi et al., 2013). Under suitable conditions, metals may get transferred to liquid matrix (water) from solid matrix (sediment). Therefore, sediments in aquatic ecosystem may behave both as source and sink of metal pollution (Medici et al., 2011). Metals are almost immobile as they are bound to silicates and minerals in unpolluted ecosystem. Under anthropogenic interference, metals may be found in oxides, carbonates, hydroxides, sulfides and other labile forms (Passos et al., 2010). Study of metal concentration in surface water helps in the prediction of metal load over a short period whereas metal speciation study in river sediments helps in long term assessment of metal pollution in the river ecosystem.

River Ganga (flow length 2525 km), with runoff 525 $km^3 year^{-1}$ and annual mean flow 5.9×10^{11} m^3 $year^{-1}$, creates a vast Gangatic plain (861,404 km^2). Due to huge sediment load 1600×10^{12} g $year^{-1}$, Ganga-Brahmaputra Delta (GBD) is formed on the confluence of river Ganga with Bay of Bengal (Purushothaman and Chakrapani, 2007; Subramanian, 2007; Shukla et al., 2012). Increasing population, unplanned urbanization and human interference are the main factors responsible for pollution in river Ganga. Kanpur, Allahabad, Varanasi, Patna are some of the most important cities imposing great anthropogenic pressure to the river. In the present study, one year regular analysis of the river Ganga water and sediment was done to assess the source and extent of pollution level in the river. The present work is different from the earlier work done by us (Pandey et al., 2014) as the earlier work dealt with metal characterization with special reference to metal speciation. Impact of pH on metal species in river sediments was also important aspect of that study.

Earlier, workers have applied ANN as decision making tool for

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prediction of toxic metal mobility in biotic and abiotic components (Hattab and Motelica-Heino, 2014; Hattab and Hambli, 2014; Hattab et al., 2013 (a); Hattab et al., 2013(b)). Similarly, ANN has also been applied to predict the physical and chemical phenomenon in suspended and river bed sediments (Rajaee, 2011; Alvarez-Guerra et al., 2010; Subida et al., 2013). The present work deals with the long term monitoring of river Ganga water vis-à-vis river sediments with the application of chemometric and artificial intelligence as source apportionment model.

2. Material and methods

2.1. Study region

Varanasi (25°16′55″N 82°57′23″E, 76 m amsl), the cultural capital of India, is situated on the bank of Ganga. High Population density (2399 persons km^{-2}), temples, narrow lanes, congested residential colonies and cluster of industries (over 7000 registered various scale industries) are the identities of Varanasi city. Around 350 million liter per day (MLD) untreated wastewater carrying heavy metals discharged into the city sewage which ultimately pollutes the river (DIP, 2014; Tripathi and Tripathi, 2011).

2.2. Sampling and analysis

Water samples were collected weekly in acid washed PTFE bottles (Tarsons, India) from drain-river confluence at fifteen sampling stations from S1 to S15 for 12 months (Fig. 1) using standard methods (APHA AWWA, 2005). Filtered (Whatman, 0.45 μm) and acidified (HNO_3) samples were used for heavy metal analysis by flame Atomic Absorption Spectrophotometer (AAnalyst 800-Perkin Elmer). Sediments sampling was done from the same 15 sites with the help of sediment sampler (1 m cylindrical plastic pipe covered by iron pipe) at the depth of nearly 10–30 cm and an interval of 15 days for 12 months. Due to high sedimentation rate in river Ganga and anthropogenic interference, 15 days interval was decided for the sampling (Singh et al., 2007). Samples were transported to Lab in plastic packets at 4 °C. EC and pH of the sediments samples were observed by standard method (Pansu and Gautheyrou, 2006). Samples were air dried, crushed (agate pestle mortar) and sieved (75 μm) for analysis. Moisture content was obtained by ASTM D2216 (ASTM, 2010). Malvern Mastersizer 2000 was used for the grain size analysis while acid digestion (USEPA, 1996) was done with the help of Teflon vessels. Metal speciation analysis was carried out by Sequential Extraction Procedure (SEP) (Tessier et al., 1979; Rauret et al., 2001; Sutherland, 2010). Sampling station coordinates were recorded by GPS (Garmin etrex Vista). Graphical representations were done with the help of Sigma Plot 12.0.

2.3. Multivariate analysis and Self-Organizing Map-Artificial Neural Network (SOM-ANN)

Multivariate analysis was carried out with the help of cluster analysis (CA) and Principal Components Analysis (PCA). PCA and CA were operated on z-score transformed data. Z-score transformation converts the experimental data into zero (mean) and unity (variance). This neutralizes the effect of multidimensionality and different units of the parameters. The transformation enhances the weight of variable having low variance and vice-versa (Singh et al., 2005b, 2005c; Wunderlin et al., 2001). These methods were applied as source apportionment technique to categorize the entire river stretch on the basis of source and degree of pollution.

Self-Organizing Map (SOM), developed by T. Kohonen, is a nonlinear projection mapping and based on unsupervised competition learning method which is used in pattern analysis and classification. A grid is formed by the SOM where similar models are closely associated while unrelated models are placed far (Astel et al., 2007; Kohonen, 2013). SOM-ANN was applied on the river water data set, having 3780 elements (21 variables, 180 samples). The model was validated using real time data obtained by physico-chemical and metal analysis of river Ganga water collected from different sampling sites. The maximum iteration was 200 having learning rate .5 and training function *trainbu*, based on weight and bias learning rules. Weights were adjusted by back-propagation according to training and learning rule. Six significant clusters were obtained by applying grid size of 3 × 2 on 3780 elements grouping the sampling stations and month depending upon the similarities among the elements. Topology used for neighborhood and distance measure between nodes is hexagonal. Input (variables) is presented to all nodes and response (output or cluster) is calculated. Highest response neuron is winning neuron. Weights are adjusted for winning and neighboring neurons. SOM-ANN has become an important tool to assess the sediments quality (Arias et al., 2008; Subida et al., 2013).

The sole purpose of the application of multivariate and SOM-ANN analysis was the source apportionment of the pollutants along the entire stretch of river. All the statistical operations were done with the help of MS Excel 2010, SPSS 16.0, MATLAB 2012 and R statistical package.

3. Results and discussion

3.1. River water

The physico-chemical results of the river water showed that electrical conductivity (EC) of alkaline river water (pH 8.4) varied between 404.1 μScm^{-1} to 452.2 μScm^{-1}. Low DO (2.2–5.65 mgl^{-1}) with high BOD (35.3–59.76 mgl^{-1}) and COD (53–83.55 mgl^{-1}) indicated significant pollution in the river (Table 1). Seasonal variation of physico-chemical properties of river water is given in Table 2. Iron (Fe) was present in highest mean concentration (134.6 ± 3.6 μgl^{-1}) followed by Cr (44.6 ± 12.4 μgl^{-1}) and Mn (44.3 ± 5.8 μgl^{-1}) in summer season (April to June) (Fig. 2). Cd was present in lower concentration (8.9 ± 1.9 μgl^{-1}) along the entire river stretch. Seasonal variation study revealed highest concentration of metals in summer season followed by winter and least in rainy season. Significantly, higher concentration of metals in summer seasons may be due to reduced volume of water in the river. The principal anthropogenic sources of metals in the region are effluent from metal-alloy industries and textile-dye industries, which get mixed with the city sewage and ultimately join the river. Round the year dead body cremation at S7 and S9 also contribute pollution to the river up to a great extent. Middle to last stretch was found to be the most polluted section of the river. Lack of efficient metal removing technologies in the region is also an important factor for the metal pollution in the river. Alkaline nature of river water (pH 8.4 ± .1) may convert metals into their respective oxide, hydroxide and carbonate forms (Singh et al., 2005a).

3.2. River sediments

Grain size analysis revealed that sand (53%–80%) was the chief component of the river sediments followed by silt (14.82%–40.17%) and clay (2.06%–6.77%) (Fig. 3). Water content varied from 33.2% (S9) to 49.65% (S15). EC and pH of river sediments ranged between 67.3 and 151.55 μS cm^{-1} and 7.71–8.72 respectively. The pollution load index (PLI) continuously increased from upstream to downstream of the river (Fig. 3).

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