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Environmental Impact Assessment of Trace Metal Deposition Around the Petrol Filling Stations

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Abstract. The wide use of petroleum products causes contamination of air, water, soil and plants. The present study was conducted to monitor the trace metal deposition in road side soil around the petrol filling stations along the busy roads of Karachi, Pakistan. Total 21 road side soil samples were collected from selected locations of busy roads. The soil samples were digested using acid digestion method and atomic absorption spectrophotometer (AAS) was used for the elemental analysis. Results of the study showed that concentration of lead was highest in the soil samples ranging from 41.3 to 361 mg/kg, then copper from 23.0 to 101 mg/kg, manganese from 36.2 to 125.0 mg/kg and zinc from 27.5 to 213.0 mg/kg, respectively. The correlation-coefficient (r) was also calculated between the metals in soil samples. The correlation matrix showed that all the pollution is coming from the same source. The gravitational sedimentation and impact on vegetation of coarse fraction is responsible for the high lead contamination of vegetation and soils. Collected data showed that, almost all the pollution being generated by automobile exhaust in urban areas of Karachi. The soil acts as an important sink for pollutants released through different activities.

Keywords: petrol filling stations, trace metals, soil, vehicular traffic, acid digestion

Introduction

Air pollution has become a serious environmental issue, mainly due to the presence of toxic trace metals in the atmosphere as a consequence of rapid industrialisation and increased transportation during recent years. Pollution of soils by trace metals is a serious environmental issue. Determination of the trace metal contents of various environmental materials such as soil, natural water, plants, dust etc., have been performed by various researchers (Malakotian *et al.,* 2009; Vinodhini and Narayanan, 2009; Kalantari and Ghaffari, 2008; Karabassi *et al.,* 2008; Itoh *et al.,* 2006; Krolak, 2000; Soylak *et al.,* 2000).

Trace metals have been added into urban soils through urban waste, chemical industries (Chaoyang *et al.,* 2009) and most importantly through the vehicular emission (Xia *et al.,* 2011). The urban road side soil has been recognised as an important repository of trace metals.

Environmental measurements revealed the higher concentrations of metals in road side soil near the petrol filling stations, and most of the signs could be attributed due to petrol fumes (Das *et al.,* 1991). Lead, copper, manganese and zinc are the major metal pollutants of the roadside environment and are released during different operations of the road transport such as combustion, component wear, fluid leakage and corrosion of the metals. Lead is one of the most important trace metals in Pakistani environment because of the extensive use of lead tetraethyl as gasoline additive, since early years of the 20th century until 2005, when this compound was prohibited and the unleaded petroleum introduced. According to Fernandez and Ramirez (2002), the concentration of lead in solid particles from roads of the city of Caracase ranged between 5.500 to 13.000 µg/g, respectively. Carrasquero (2006) reported the levels of lead from 2.000 to 4.000 µg/g in urban soils near the most transited streets and avenues.

Majority of the trace metals are toxic to the living organisms, and can impair important biochemical processes posing a threat to human health, plant growth and animal life (Silva *et al.,* 2005; Jarup, 2003; Michlake, 2003).

Zinc is a trace element that is essential for human health. When people absorb too little quantity of zinc they can experience a loss of appetite, decreased sense of taste and smell, slow wound healing and skin sores. Zinc-shortage can even cause birth defects. Although humans can handle proportionally large concentrations of zinc, too much zinc can still cause eminent health problems, such as stomach cramps, skin irritations, vomiting, nausea and anemia. Very high levels of zinc can damage the pancreas and disturb the protein metabolism, and cause arteriosclerosis. Extensive exposure to zinc can cause respiratory disorders.

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In the work place environment, zinc contagion can lead to a flu-like condition known as metal fever. Zinc can be a danger to unborn and newborn children (Hussein *et al.,* 2012). Similarly, higher concentration of Pb, Cu and Mn can cause serious health issues in human beings.

Long-term exposure to copper can cause irritation of the nose, mouth and eyes and it causes headaches, stomachaches, dizziness, vomiting and diarrhoea. Intentionally high uptakes of copper may cause liver and kidney damage and even death. Industrial exposure of copper fumes, dusts, or mists may result in metal fume fever with atrophic changes in nasal mucous membranes. Chronic copper poisoning results in Wilson's disease (Araya *et al.,* 2008).

Manganese effects occur mainly in the respiratory tract and in the brain. Symptoms of manganese poisoning are hallucinations, forgetfulness and nerve damage. Manganese can also cause Parkinson, lung embolism and bronchitis. When men are exposed to manganese for a longer period of time they may become impotent. A syndrome that is caused by manganese has symptoms such as schizophrenia, dullness, weak muscles, headaches and insomnia. Chronic manganese poisoning may result from prolonged inhalation of dust and fume. The central nervous system is the chief site of damage from the disease, which may result in permanent disability. Symptoms include languor, sleepiness, weakness, emotional disturbances, spastic gait, recurring leg cramps, and paralysis (Nagatomo *et al.,* 1999).

Studies have shown that, such pollutants can be harmful to the road side vegetation, wildlife and the neighbouring human settlements (Turer and Mynard, 2003; Caselles, 1998; Ferretti *et al.,* 1995; Iqbal *et al.,* 1994; Ndiokwere, 1984; Khan and Frankland 1983). It is evident that the urban soil containing the trace metals poses a serious threat to the safety of the human life by ingestion and inhaling (Wei and Yang, 2010) and through the direct contact with the soil on the road sides contaminated by trace metal (Yang *et al.,* 2011; 2010). It links to effects on the skin and penetration into the body tissues also causing genetic modification and affected intelligence of young children (Sims, 1990).

Transportation sources contribute 60-70% of the total aerial burden in Karachi. Over the last 20 years, the number of motor vehicles has risen from 0.8 million to nearly 5 million; an average growth rate in excess of 14%. The highest rise was in two-stroke vehicles (1,751%), while diesel vehicle numbers were three times higher in 2005 than in 1980 (Colbeck *et al.,* 2010). The main source of lead pollution is the emissions from automobile exhausts. Airborne lead is typically present in vehicular exhaust emissions in the form of particles, which have diameter of less than 1 μm and therefore, may be transported to large distance in the atmosphere between one and 4 weeks lifetime depending on climatic factors. The current emission of lead from automobile exhausts is around 7000 tonnes annually. 10% of this level is fallen out as dust on road side and remaining in airborne until it is washed out of the air by rain or it can be removed by contact with vegetation or soil.

The world's petrol consumption is about 84.2 million barrels per day (in 2010). Only United States (North America) consumes 27 million barrels per day (27.3% of the world total), Asia consumes 29 million barrels per day whereas, Pakistan is the 36th biggest consumer of petrol (CIA, 2013).

Very little work has been carriedout in Karachi to assess the contribution of emission sources of trace metals to the total aerial burden and their accumulation in road side soils due to petrol filling stations of the city.

The aim of this study is to determine the concentrations of airborne trace metals lead (Pb), copper (Cu), manganese (Mn) and zinc (Zn) near the petrol filling stations as indicators of the impact produced by the emission of vehicles. This information will be useful to evaluate the potential risk on the habitants of this city.

Materials and Methods

Sampling. For collection of soil samples, 21 locations were selected on the main roads, round abouts and open places near petrol filling stations along the busy roads of Karachi. Table 1 indicating the sampling points along with sample code, Global positioning system (GPS location) and local locations. Soil samples were collected from a depth between 5-10 cm (Marjanovic *et al.,* 2009; Gadkari and Pervez, 2008). The samples were stored in polyethylene bags then treated and analysed separately. The samples were prepared and analysed by AAS for trace elements Pb, Cu, Mn and Zn, respectively.

Reagents. All the experiments were performed with analytical reagent grade chemicals. Distilled and de-ionised water was used for dilution and preparation of reagents and standards. Reference standards were prepared from BDH spectrosol AA standard (1000 mg/L).

Sample preparation. Samples were dried at 120 °C and homogenised, passed through 0.5 mm size sieve. One gram portion of prepared soil samples was gently refluxed

Table 1. Soil sample codes and their locations

Sample code	Local locations	GPS locations	
		Lat	Long
S-1	Shell Petrol Pump	24.9359	67.0727
S-2	PSO Petrol Pump	24.9368	67.0744
S-3	PSO Petrol Pump	24.9255	67.0656
S-4	PSO Petrol Pump	24.9018	67.0455
S-5	Shell Petrol Pump	24.9157	67.0626
S-6	PSO Petrol Pump	24.8923	67.0437
S-7	Shell Petrol Pump	24.8801	67.0389
S-8	PSO Petrol Pump	24.8786	67.0473
S-9	Shell Petrol Pump	24.8806	67.0436
S-10	Caltex Petrol Pump	24.8895	67.0364
S-11	Shell Petrol Pump	24.9179	67.0971
S-12	Caltex Petrol Pump	24.9034	67.0548
S-13	Shell Petrol Pump	24.8831	67.0500
S-14	PSO Petrol Pump	24.9109	67.0307
S-15	PSO Petrol Pump	24.9029	67.0024
S-16	Shell Petrol Pump	24.8918	66.9887
S-17	PSO Petrol Pump	24.8873	67.0330
S-18	Caltex Petrol Pump	24.8556	67.0385
S-19	Caltex Petrol Pump	24.8503	67.0303
CS-20	PSO Petrol Pump	24.9431	67.0952
CS-21	Shell Petrol Pump	24.9467	67.1392

with 2 M nitric acid for 30 min. After cooling the contents were filtered through Whatman 42 into a graduated 50 mL flask and diluted up to the mark. All the glasswares were extensively soaked in diluted HNO_3 and rinsed with double distilled water.

Apparatus. Analysis was performed on atomic absorption spectrophotometer (Hitachi Z-8000), with Zeeman effect background correction. The spectrophotometer was equipped with a graphite furnace, a microprocessor and a built in printer.

Metal analysis. Determination of Cu, Mn and Zn were carried out by flame atomic absorption spectrophotometer (FAAS) whereas Pb was carried out by electro-thermal atomic absorption spectrophotometer (ETAAS), employing the standard addition technique. Measurement was made by using the hollow cathode lamp for the metals Pb, Cu, Mn and Zn, respectively.

Triplicate readings were taken on each sample by AAS, and the mean values of each sample readings were used to calculate results. The working conditions and detection limits for the detected elements by atomic absorption spectrophotometer are presented in Table 2. Accuracy was also monitored by spike with Cu, Mn and Zn in the level of 1.0 mg/L each, whereas Pb in the level of 20 μg/L, respectively. Five soil samples were spiked with Pb, Cu, Mn and Zn to determine the recovery. The average recovery for each metal i. e., Pb, Cu, Mn, and Zn are also given in Table 2.

Results and Discussion

Statistical parameters pertaining to the soil samples for trace metals in the form of range, mean, median and standard deviation are presented in Table 3. Correlation coefficient matrix are also presented in Table 4. The average concentration of lead, copper, manganese and zinc for each of 21 locations is presented in Fig. 1. The level of all investigated ions in soil samples were found at mg/kg.

Figure 1 shows the mean values of total average concentration of lead, copper, manganese and zinc at selected locations in Karachi. The highest average concentration of lead was found to be 361 mg/kg at location S-17; whereas, the lowest concentration was recorded as 70 mg/kg near location S-19, respectively. The mean values

Table 2. Working condition for detected elements and the recovery of metals added to the soil samples by atomic absorption spectrophotometer

Parameters	Pb	Cu	Mn	Zn
Slit (nm)	1.3	1.3	1.3	1.3
Wave length (nm)	283.3	324.8	279.3	213.9
Drying temperature (°C)	80-120	-	-	-
Ashing temperature (°C)	400	-	-	-
Atomisation temperature (°C)	2000	2300	2300	2300
% Recovery of metal added	95.5%	99.6%	98.0%	97.2%

Table 3. Average metal concentration (mg/kg) in soil samples collected from road side near petrol filling stations in Karachi

Metal	Range (mg/kg)	Mean	Median
Pb	41.3 - 361.0	126.5	109.0
Cu	23.0 - 101.0	75.8	76.0
Mn	36.2 - 125.0	73.7	69.0
Zn	27.5 - 213.0	97.6	91.0

Table 4. Correlation coefficient in soil samples collected from petrol filling stations in Karachi

Metals	Pb	Cu	Mn	Zn
Pb	1.00	-	-	-
Cu	0.613	1.00	-	-
Mn	0.703	0.627	1.00	-
Zn	0.850	0.671	0.822	1.00

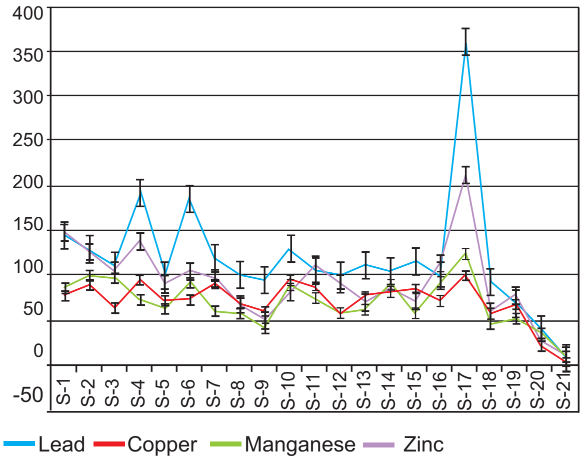

Fig. 1. Concentration of trace metals in soil samples near petrol filling stations.

of total average concentration of lead (Fig. 1) shows that, relatively high concentration of lead was found at locations S-4 (201 mg/kg), S-6 (187 mg/kg) and S-17 (361 mg/kg). These locations of road side filling stations are surrounded by high traffic density and there are several automobile workshops and large parking places for hundred of rickshaws near these filling stations. Moreover, during the period of survey, civil works for bridges was also under progress at these locations.

The lowest lead level at S-19 was found to be 70 mg/kg (Fig. 1). The low values obtained here may be because the location is relatively open place and situated at the intersection of very wide road having large round about. Lead has been found to be one of the major toxic elements generated through the motor vehicle exhaust using leaded gasoline. It is non degraded pollutant and it not only accumulates in the body but also modifies itself as it moves through biological cycles and food chain.

In the last decades, much attention has been directed towards lead in the road side environment as a result of its widespread use as an anti-knocking agent in gasoline (Turer and Mynard, 2003; Hafen and Brinkmann, 1996; Weheeler and Rolfe, 1979; Davis and Holmes, 1972). In the recent years, however, the lead content in gasoline markedly decreased after the introduction of the regulations requiring the reduction in the lead content from 0.64 g/L in 1966 to 0.14 g/L in 1986. This decrease has reduced the addition of lead to the environment by motor vehicles. However, the previously deposited lead remains a major contaminant of the road side environments. Although the lead content in gasoline is minimised these days, the increased traffic has caused an increase in the lead emission in the road side environment (Jones *et al.,* 1991).

Relatively high concentration of zinc was found at location S-1(149 mg/kg), S-4 (159 mg/kg) and S-17 (213 mg/kg), respectively. The maximum concentration of zinc in the soil was found to be 213 mg/kg, at location S-17 and lowest 51.0 mg/kg at location S-8 (Fig. 1). The highest concentration of zinc in soil at location S-17 may be due to the fact that this area is mostly surrounded by automobile workshops and has high traffic density. Most of the vehicles (60%) plying on the roads are using old and out dated imported tyres. These tyres are prone to tear off quickly as compared to new tyres and add more zinc in the soil as zinc is the component of tyres (Howard *et al.,* 1984). The average concentration of lead, copper, manganese and zinc are of the same order of magnitude. These metals are released during different operations of the road transport such as combustion, component wear, fluid leakage and corrosion of batteries and metallic parts such as radiators etc. (Dolan *et al.,* 2006).

Many studies on trace metal concentration in urban soil have been undertaken in various metropolitan cities (Table 5). Table 3 shows that, in the present studies, average concentration of Pb was recorded in the range of 41.3 to 361.0 mg/kg with mean value of

Table 5. Average metal concentration (mg/kg) in urban soils from different cities across the world

City	Pb	Cu (mg/kg)	Mn	Zn	Reference
Galway	58.0	27.0	539.0	85.0	Zhang, 2006
Hong Kong	88.1	16.2	-	103.0	Celine *et al.,* 2006
Madrid	22.0	14.0	249.0	50.0	De-Miguel *et al.,* 2007
Hangzhou	46.2	36.6	415.3	116.1	Hasan *et al.,* 2008
Belgrade	298.6	46.3	417.6	174.2	Mirjanvi *et al.,* 2009
Yorkshire	25.0	15.5	-	56.7	Khalid *et al.,* 2006
Karachi	120.9	72.4	70.7	93.5	Present study

126.5 mg/kg which is higher than that reported from Galway 58.0 mg/kg (Zhang, 2006), Hong Kong 88.1 mg/kg (Celine, 2006), Madrid 22.0 mg/kg (De-Miguel *et al.,* 2007) and Hangzhou city 46.2 mg/kg (Hasan, 2008) and lower from Belgrade 298.6 mg/kg (Marjanovic *et al.,* 2009).

Concentration of Cu in different urban soil from various cities had been reported to be in the range 10-50 mg/kg. In the urban soil samples of Galway the concentration of Cu was 27 mg/kg, in Hong Kong 16.2 mg/kg, in Madrid 14 mg/kg, in Hangzhau 36.6 mg/kg and in Belgrade 46.3 mg/kg.

Average concentration of Cu in the soil samples recorded in the present study was 23.0 mg/kg (Table 3), which is higher than that reported from Hong Kong 16.2 mg/kg, Madrid 14 mg/kg and lower from the soil samples of other cities compared with whereas, the concentration of Mn in different urban soil samples is in the range of 250-550 mg/kg (Table 5). In the present study, the mean concentration of Mn is 73.7 mg/kg (Table 3), which is lower from all other cities compared in this study (Table 5) whereas, the concentration of Zn in this study was recorded in the range of 27.5-213.0 mg/kg with the mean value of 97.6 mg/kg Table 3 which is higher from Galway 85.0 mg/kg and Madrid 50.0 mg/kg and lower from Hangzhou 116.1 mg/kg, Hong Kong 103.0 mg/kg and Belgrade 174.2 mg/kg compared in this study (Table 5).

Location number SC-21 is located about 20 km away from the city centre in the down wind direction. This location was used as control site in this investigation. The highest concentration of lead found at selected locations was more than 50 times greater than the lead found at control site (Locations No: SC-21). The high increase in the concentration of lead at selected locations is due to very high number of vehicles plying at the intersections of filling stations than the control site filling station. The average concentration of lead at control location was found to be 9 mg/kg, Cu 5 mg/kg, Mn 14 mg/kg and Zn 12 mg/kg, respectively. The analysis for soil sample from other locations when compared with the control location (SC-21) shows that almost all the pollution (>90%) near petrol filling stations is being generated due to higher density of vehicular exhaust and leakage of petrol fumes during filling of vehicles and scratching of vehicles tyres on roads. The study conducted to compare road side soils of northern England also shows ten time greater concentration of lead in the soil samples collected along busy roads of petrol filling stations then the control location concentration (Akbar *et al.,* 2006).

The climate of Karachi is tropical and dominated by the monsoon region. The average rain fall in Karachi amounts to about 20 mm and wind direction is SW in summer, and during winter it is usually in the NE. Wind velocity is 10 m/sec during the June-July and 3.5 m/sec from January to March. Because of the dry climate in Karachi lead contamination in the soil is not easily eliminated nor fixed.

Table 4 presents the matrix correlation for all the variables. The correlation coefficient is statistically significant when the values are above + 0.5. The Table 4 clearly shows that the values are positive and approximately all the values are above + 0.5.

Lead in the form of tetraethyl lead acetate is used as anti-knocking agent in petrol. It shows good relation with Cu, Mn and Zn. As the samples of soil were collected from petrol filling stations therefore, the source of trace metals in soil may be vehicular traffic. Lead is anti knocking agent in petrol and copper is the component of engine; hence both are directly related in automobiles and have good agreement in correlation coefficient matrix. Manganese shows the lowest correlation coefficient with all the metals studied. Although it is the component of tyres, but the values show that the correlation of manganese with other metals is not statistically significant. Zinc shows highest correlation with copper. The sources of these metals may also be the motor vehicles. Lead, cadmium, copper, and zinc are the major metal pollutants of the roadside environments and are released from fuel burning, wear out of tyres, leakage of oils, and corrosion of batteries and metallic parts such as radiators etc. (Dolan *et al.,* 2006).

A hospital survey was carried out to assess the impact of pollution on human health (Table 6). Seven hospitals were visited to collect the data about patient suffering from various diseases due to inhalation, during the study period 2009-2010.

Survey of hospitals shows that, the number of patents suffering from air pollution related diseases to that of allergic diseases is about 3:1. The number of male cases as compared to females regarding air pollution allergies, mainly related to chest infections is in the ratio o 2.4:1. This may be due to an extensive exposure of male to the polluted air and professional hazards as compared to females who are mostly house wives and remain indoor.

Table 6. Number of patients suffering from atmospheric pollution related diseases in hospitals of study area

Hospitals	Diseases	Number of cases			Male female ratio
		Male	Female	Total	
Kiran Hospital	Chest infection	12,218	5,479	17,697	2.2 :1
	Cancer	137,232	75,488	212,720	1.8 :1
	Heart ailment	6,487	3,743	10,230	1.7 :1
	Allergy	5,631	2,054	7,685	2.7 :1
	T.B	6,231	3,125	9,356	2.0 :1
	Pulmonary diseases	20,432	9,288	29,720	2.2 :1
Darul Sehat Hospital Gulistan-e-Jauhar	Chest infection	47,738	21,862	69,600	2.2 :1
	Cancer	16,572	7,828	24,400	2.1 :1
	Heart ailment	19,381	9,419	28,800	2.1 :1
	Allergy	7,123	4,189	11,312	1.7 :1
	T.B	9,421	4,127	13,548	2.3 :1
	Pulmonary diseases	85,525	38,875	124,400	2.2 :1
Ashfaq Memorial Hospital Gulshan-e-Iqbal	Chest infection	98,288	54,132	152,420	1.8 :1
	Cancer	69,375	36,145	105,520	1.9 :1
	Heart ailment	328,320	172,800	501,120	1.9 :1
	Allergy	8,421	3,715	12,136	2.3 :1
	T.B	10,220	6,012	16,232	1.7 :1
	Pulmonary diseases	111,822	48,618	160,440	2.3 :1
Abbasi Shaheed Hospital Nazimabad	Chest infection	86,196	49,644	135,840	1.7 :1
	Cancer	7,008	2,672	9,680	2.6 :1
	Heart ailment	117,961	54,423	172,384	2.2 :1
	Allergy	4,614	1,987	6,601	2.3 :1
	T.B	11,503	5,712	17,215	2.0 :1
	Pulmonary diseases	97,960	50,480	148,440	1.9 :1
Imam Clinic North Nazimabad	Chest infection	135,021	61,425	196,446	2.2 :1
	Cancer	3,212	1,704	4,916	1.9 :1
	Heart ailment	123,760	81,345	205,105	1.5 :1
	Allergy	3,614	1,287	4,901	2.8 :1
	T.B	15,503	8,712	24,215	1.8 :1
	Pulmonary diseases	99,543	39,021	138,564	2.6 :1
Shahrukh Hospital North Nazimabad	Chest infection	26,201	12,103	38,304	2.2 :1
	Cancer	712	324	1,036	2.2 :1
	Heart ailment	2,427	987	3,414	2.5 :1
	Allergy	1,614	680	2,294	2.4 :1
	T.B	7,503	2,712	10,215	2.8 :1
	Pulmonary diseases	17,960	7,480	25,440	2.4 :1
Taj Medical Complex Sadar	Chest infection	115,021	47,425	162,446	2.4 :1
	Cancer	9,503	3,712	13,215	2.6 :1
	Heart ailment	143,660	61,245	204,905	2.3 :1
	Allergy	2,427	787	3,214	3.1 :1
	T.B	1,614	580	2,194	2.8 :1
	Pulmonary diseases	91,543	40,021	131,564	2.3 :1
Total		2,126,517	1,043,367	3,169,884	2.0 :1

Few decades ago only industrial emissions was considered as an important risk factor for lung cancer but nowadays polluted air due to vehicular exhaust is the most important factor for lung cancer and other types of chest infections and pulmonary diseases. People in developing countries are commonly exposed to very high levels of pollution for 3-10 h daily over many years. The number of lung cancer cases by air pollution, are also on the increase and mostly male cases due to outdoor exposure in air. The worst affected age group for this disease was between 50 – 60 years, now reducing to 35-55 years. This is also mainly due to increasing air pollution level but some other factors are also involved like personal hygiene, social activity, socio economic conditions, mental worries and smoking etc.

Conclusion

Trace metal contamination in the soil samples collected from main roads petrol filling stations along the busy roads of Karachi shows that growing number of vehicles, leaded gasoline consumption and poor conditions of the roads are major cause of high concentration of lead and other trace metals level in Karachi. Soil acts as an important sink for pollutants released through industrial discharge and other human activities. It is suggested that in the polluted areas such species of the plants should be grown which not only clean the atmosphere but also grow well in the polluted atmosphere as a permanent source of cleaning the environment around the filling stations.

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