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Accumulation of Lead and Cadmium in Soil and Vegetable Crops along Major Highways in Agra (India)

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Abstract: Environmental pollution of heavy metals from automobiles has attained much attention in the recent past. The present research was conducted to study Pb and Cd level concentrations in soil and vegetations along a major highway with high traffic density. Soil and vegetable samples along highway were collected from 10 sites in Agra district (India) and analyzed for two heavy metals (lead and cadmium) using flame atomic absorption spectrophotometer (AAS). The soil physicochemical properties were also determined. The general decrease in concentrations of these metals with distance from the highway indicates their relation to traffic. Higher accumulations of metals have been observed on vegetation and soil samples near to the highway (0-5 m) than on vegetation and soil samples from sites a little farther away (at 5-10 m & 10-15 m). This is attributed mainly to aerial deposition of the metal particulates from motor vehicles. The values of heavy metals were compared with results found by other investigators in various countries worldwide.

Keywords: Environmental pollution, Heavy metals, AAS, Traffic density.

Introduction

The pollution of soils by heavy metals from automobile sources is a serious environmental issue. Results show that roadside soil near motorways is heavily polluted by heavy metals from automobiles[1,2]. These metals are released during different operations of road transport such as combustion, component wear, fluid leakage and corrosion of metals. Lead and cadmium are the major metal pollutants of the roadside environments and are released from fuel burning; wear out of tires, leakage of oils *etc*[3].

The elevated levels of Pb and Cd in urban areas are mainly attributed to automobile exhaust, particularly from leaded gasoline, motor vehicle tires, and lubricant oils[4-6]. Recently it was claimed that Pb in urban areas could be over 1000 ppm, where as tolerable intake of ingested Pb for adults is 3000 µg per week, which is equivalent to an average daily uptake of 430 µg and 130 µg for children[7]. For Cd, critical levels for adults, food source and water are

120, 45 and 13 µg per day respectively. In addition, FAO/WHO recommended maximum tolerable intake of Cd of 400-500 µg per week or equal 70 µg per day. Several studies revealed that 60-80% of heavy metal toxins found in human bodies in urban areas were the results of consuming contaminated foods rather than air pollution[8-11].

A great part of metal pollutants are deposited on adjacent soil where they may be transformed and transported to other parts of the environment for example, to vegetation. In addition to soil, vegetables function as a sink for atmospheric pollutants because of its capacity to act as efficient interceptions of airborne matter. The plants are widely used as passive bio-monitors in urban environments[12,13].There is no doubt that leafy vegetables grown in the neighborhood of major motorways, can contain significant traces of Pb and Cd due to airborne metal particulates derived from motor vehicle emissions. The distribution of these metals (Pb and Cd) in the roadside soils is strongly but inversely correlated with the distances away from the roadside[14,15].

The tremendous increase in the number of motor vehicles in Agra city is leading to increasingly high levels of some heavy metals in the urban as well as highway environment[16]. This work consists of study of the distribution of Pb and Cd in surface soil (0-15 cm) and in leaves of some vegetable crops sampled at distances from the center of traffic flow along a highway.

Experimental

Agra is one of the most famous tourist spots of the country. The city, situated on the west bank of river Yamuna, is known world over as home to a wonder of the world, Taj Mahal. A part of the great Northern Indian plains, Agra has a tropical climate. The climate during summer is hot and dry with temperature ranging from 32 °C to 48 °C. In winter the temperature ranges from 3.5 °C to 30.5 °C. The downward wind is South-South-East *i.e.* SSE 29% and North-East *i.e.* NE 6% in summers and it is West-North-West *i.e.* WNW 9.4% and North-North-West *i.e.* 11.8% in winters. Agra, the city of Taj, (27°10' N 78° 02' E) is located in the north central part of India. It is considered as a semi-arid zone as two third of its peripheral boundaries are surrounded by the Thar Desert of Rajasthan. The atmospheric pollution load is high & because of the down ward wind, pollutant may be transported to the different areas Agra has about 1316177 total populations and population density is about 21148 per sq km with 386635 vehicles registered[16]. Vehicular exhaust accounts for more than 50% of the total pollution from all the sources put together in all big cities of India[17]. In Agra, 60% pollution is due to vehicles[18].

Collection of samples

Samples were collected from the Mathura- Kanpur highway about 15 km from the nearest urban center (Agra city). This highway carrying an average of 10^5 motor vehicles per day. 10 Sites were selected for the study along this highway. At each site, three samples of soils and three samples of vegetables were collected at different distance from the edge of the main road (0-5 m, 5-10 m, 10-15 m). The sites were particularly suitable because: (i) the traffic density is comparatively very high; (ii) there are no urban, large scale urban or industrial activities; (iii) there are no major road intersections which can cause a significant decrease in traffic density to and from Agra city.

Sample preparation and analysis

Soil samples were air-dried, ground in a porcelain mortar to pass through a 2 mm sieve. About 1 g of soil was accurately weighed and transformed to a 100 mL conical flask and 5 mL of conc. HNO_3 (AR 70%) was added and kept it for overnight. The flask was placed on a hot plate inside a fume hood, heated at a temperature of 70 °C for 1 h, and then kept it for cooling for 30 min and 5 mL of aquaregia, a mixture of conc. HNO_3 and $HClO_4$ (AR 70%, Merck) in a ratio of 4:1 was added and again the flask was placed on hot plate, heated at a temperature of 80 ^oC for 2 h. After that it was cooled for 1 h and transferred to 100 mL volumetric flask through filtration (Whatmann 42) and the final volume was made up to the mark with double distilled water, mixed well by shaking, and let settle for at 15 h. The resultant supernatant was analyzed by flame atomic absorption spectrophotometer (FAAS, Perkin -Elmer, ANALYST 100) for total Pb and Cd. The instrument was calibrated using Pb and Cd standard for each element being analyzed. Laboratory blanks were prepared by adding 10% aquaregia to a conical flask containing none of the sample being investigated. This consisted of all components added to the matrix during digestion. All soil and blank samples were analyzed for total trace metal levels.

Leaves of vegetables from different sites have been collected by hand, carefully packed in to polythene bags and brought to the laboratory for the further analysis. These leaves were carefully washed with tap water to eliminate the adhering soil and other contaminates. Plant samples were oven dried at 100 ^oC for 48 h, ground in a porcelain mortar to pass a 2 mm sieve. 1 g of ground-dried sample were taken and placed it in a small beaker.10 mL of concentrated HNO_3 was added and allowed it to stand overnight. Then it was carefully heated on a hot plate until the production of red NO_2 fumes has ceased. It was kept for cooling and a small amount (2-4 mL) of 70% $HClO_4$ was added. Heated again and allowed to operate a small volume. The sample was transferred to a 50 mL flask and made up with double distilled water. Then the quantification of metallic content (Pb & Cd) of digested samples was carried out with the FAAS.

Statistical analysis

One - way analysis of variance (ANOVA) was applied to find out the correlation of metal (total) concentrations in the soils and vegetables with the distances from roadside. In addition, Pearson's correlation was used to evaluate the metal concentration in soil and vegetable versus distance from road with the help of software SIGMA STAT 3.5.

Results and Discussion

The physicochemical properties of soil at roadside agricultural sites in Agra are presented in Table 1. Maximum pH of the roadside soil was found to be 9.20 at site 4(mean value) and minimum pH was observed 7.44 at site 6 (mean value). The maximum electric conductance value for roadside soil was 0.88 ds/m at site 4, which is approximately five times higher than the minimum EC mean value at site 10 (0.16 ds/m). The max mean value of organic carbon (%) was found at site 2 (1.59%) while minimum mean value of organic carbon in roadside soil was 0.38% at site 5.

Table 1. Physicochemical properties of soils in roadside soils. (Mean values)

Site	pH	EC (ds/m)	OC, %
1	8.65	0.65	0.53
2	8.66	0.44	1.53
3	8.55	0.49	0.73
4	9.20	0.88	0.52
5	8.09	0.18	0.24
6	7.44	0.42	0.50
7	8.35	0.18	0.38
8	7.72	0.20	0.47
9	7.98	0.41	0.37
10	7.89	0.16	0.67

In the present study, the distance from the road was served as a treatment. Therefore, heavy metals (Pb & Cd) contents in soil as well as vegetables were measured in triplicates of three regions of each site (0-5 m, 5-10, 10-15 m). Three samples of soils and vegetables each were collected from particular region.

Lead

The amount of lead in roadside soil is strongly but inversely correlated with the increase in the distance from road[19]. Table 2 and Figure 1 show the lead content in roadside soil at different distance. At site 2, the lead concentration is highest (50.10±1.94 mg/kg) but it decreased with the increasing distance from roadside; 24.13±2.57 (5-10 m) & 14.90±3.34 (at 10-15 m). In the present study, the lead content of roadside soil ranged from 0.00 to 50.10 mg/kg (mean values). By considering the general range of the total lead content, it appears that the total lead content in 85% roadside soils was below the critical conc. of 400 mg/kg[20]. In the last decades much attention has been directed towards lead in the roadside environments as a result of its widespread use as an antiknocking agent in gasoline[21-24]. Due to growing concerns about the problems associated with Pb, the use of leaded gasoline has been decreasing globally at an annual rate of about 7%[25]. The maximum level of Pb in leaded gasoline has been set to be less than 0.15 g/L since July 1989[26], but there are still many counties that use of leaded gasoline with Pb content of about 0.4 g/L[25,27]. Although the use leaded gasoline decreased during this time period, but the increasing number of automobile compensated its effect on the vehicles based on lead emission. In addition, wearing down of vehicle tires can also introduce Pb[28] to the roadside soil. Consequently, road transport is still polluting the atmosphere, soil and water near the highway[29-31].

Table 2. Lead (mg/kg) in roadside agricultural soil at selected sites

Site	Distance from road (m)		
	0-5 m	5-10 m	10-15 m
1	12.60± 0.81(11.70-13.30)	10.13±0.25(9.90-10.40)	9.40±0.98(8.30-10.20)
2	50.10± 1.94(48.60-52.30)	24.13±2.57(21.20-26.00)	14.90±3.45(12.40-18.70)
3	11.80±0.98(11.00-12.90)	10.73±0.23(10.60-11.00)	4.63±0.83(4.10-5.60)
4	17.33±0.64(16.60-17.80)	13.90±0.91(13.10-14.90)	12.13±1.44(11.20-13.80)
5	19.50±1.25(18.20-20.70)	3.20±0.43(2.90-3.70)	0.00±0.00(0.00-0.00)
6	5.60±0.36(5.30-6.00)	2.30±0.20(2.10-2.50)	0.00±0.000.00-0.00)
7	4.17±0.32(3.80-4.40)	3.33±0.55(2 .80-390)	2.10±0.17(2.00-2.30)
8	15.13±1.49(13.90-16.80)	12.40±0.78(11.50-12.90)	2.90±0.85(2.00-3.70)
9	6.90±0.55(6.30-7.40)	3.60±0.36(3.20-3.90)	3.10±0.45(2.60-3.50)
10	6.13±0.15(6.00-6.30)	2.73±0.58(2.30-3.40)	4.60±0.60(3.90-5.00)

Mean ± SD (Range)

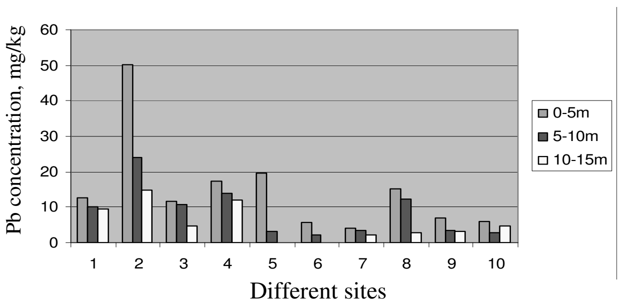

Figure 1. Pb (mg/kg) in roadside agricultural soils

In terms of distance from the main road, there were high correlations between Pb content and distance, these were from r = -0.45, p > 0.05 (at site 10) to r = -0.99, p< 0.01 (at site 6.). Highly significant correlation of Pb content in soil and distance were observed at all sites except of site 10 (r = -0.45, p > 0.05) It means that relationship between distance and average values of lead content was found to be inversely correlated at most of the sites. The mean values of Pb obtained from all the soil sampled at different distances away from the highway were significantly different (p < 0.01). Pb content in soil was still within the acceptable limits as reported by Tsadilas (2000), where the range of Pb accumulated in uncontaminated soils reached 15- 106 ug/g dry weight. In polluted soils, Pb content ranges limit from 100 to 400 ug/g dry weight. Pb content in soil near road was lower than EU upper limit of 300 mg/kg[32] and was at mg/kg[33].

Lead, although not readily soluble in soil, is absorbed mainly by root hairs and is stored in the cell walls. The translocation of Pb from roots to tops is greatly limited[34-36] and as Zimdahl (1976) described[37]; only 3% of the Pb in the root is translocated to the shoot. Lead mobility and bioavailability are controlled by several soil factors such as pH, redox potential, organic matter and the chemical from and species of lead[38]. Airborne Pb, a major source of Pb pollution, is also readily taken up by plants through foliage[39]. The epidermal cells absorb Pb deposited on the leaf surface.

Cauliflower, cabbage, Okra, radish, spinach and brinjal contained high mean concentration of Pb (Table 3 and Figure 2). The concentrations of Pb showed a decreasing trend as the distance increased from the road edge. At site 4, the lead concentration in brinjal beside the roadside was found to be highest (31.20±2.94 mg/kg) but it is decreased with the increasing distances from roadside; 26.30±2.36 (at 5-10 m) & 9.20± 2.98(at 10-15 m). The lowest lead concentration in radish near road was 1.5± 0.1 mg/kg which is decreased to 1.20±0.30 mg/kg (at 5-10 m) followed by 0.00±0.00 mg/kg (at 10-15 m).Simple correlation analysis revealed negative correlations between Pb content and a distance, these were form r = -0.77, p< 0.05 (at site 2) to r = -0.99, p < 0.01 (at site 1).

Table 3. Lead (mg/kg) in vegetable leaves at selected sites

Site	Vegetable	Distance from road (m)		
		0-5 m	5-10 m	10-15 m
1	Cauliflower	10.20±0.95(9.10-10.80)	6.93±0.30(6.60-7.20)	3.40±1.17(2.40-4.70)
2	Cabbage	15.30±1.70(13.60-17.00)	9.70±1.12(9.00-11.00)	10.63±1.32(9.10-11.40
3	Cauliflower	9.70±0.60(9.00-10.10)	7.40±0.45(6.90-7.80)	7.33±0.64(6.60-7.80)
4	Brinjal	31.20±2.94(28.00-33.80)	26.30±2.36(23.80-28.50)	9.20±2.98(6.70-12.50)
5	Brinjal	14.40±0.52(13.80-14.80)	8.70±1.21(7.60-10.00)	8.30±1.25(7.00-9.50)
6	Radish	4.80±0.88(4.10-5.80)	3.00±0.36(2.70-3.40)	0.50±0.26(0.20-0.70)
7	Spinach	2.93±0.75(2.20-3.70)	1.70±0.10(1.60-1.80)	0.00±0.00(0.00-0.00)
8	Spinach	5.90±0.40(5.50-6.30)	5.10±0.81(4.40-6.00)	2.30±0.79(1.70-3.20)
9	Okra	1.70±0.26(1.50-2.00)	1.10±0.30(0.80-1.40)	0.70±0.10(0.60-0.80)
10	Radish	1.50±0.10(1.40-1.60)	1.20±0.30(0.90-1.50)	0.00±0.00(0.00-0.00)

Mean ± SD (Range)

The mean values of Pb obtained from all the vegetables samples at different distances away from the highway were significantly different (p < 0.01). The Pb concentrations in all the leafy vegetable samples (0-5 m) except that of site 9 & of site 10, exceeded the Indian

Prevention of Food Adulteration Act (PFA) permissible limit of 2.5 mg/kg. But if the more stringent CODEX limit of 0.3 mg/kg is used, then 100% of vegetable leaves sample exceeded safe limits. Approximately 50% of the samples exceeded the PFA permissible limit by more than 2 fold (*i.e.,* had concentrations of > 5.0 mg/kg).

Table 4. Cadmium (mg/kg) in roadside agricultural soil at selected sites

Site	Distance from road (m)		
	0-5 m	5-10 m	10-15 m
1	6.96±0.72(6.50-7.80)	6.00±0.20(5.80-6.20)	3.00±0.60(2.60-3.70)
2	21.53±5.51(15.50-26.30)	10.90±6.06(10.35-11.55)	0.00±0.00(0.00-0.00)
3	7.13±0.75(6.40-7.90)	0.00±0.00(0.00-0.00)	3.00±0.30(2.70-3.30)
4	11.00±0.98((9.90-11.60)	9.00±0.98(7.90-9.80)	1.00±0.26(0.88-1.30)
5	10.70±0.70(9.90-11.20)	10.00±0.79(9.10-10.60)	1.60±0.65(0.90-2.20)
6	6.00±0.34(5.80-6.40)	1.80±0.52(1.50-2.40)	0.00±0.00(0.00-0.00)
7	16.50±1.56(15.50-18.30)	15.00±1.83(13.00-16.60)	12.00±1.48(11.00-13.70)
8	13.97±1.33(12.50-15.10)	13.20±1.05(12.40-14.40)	9.00±0.87(8.40-10.00)
9	7.03±0.68(6.50-7.80)	7.00±0.70(6.50-7.80)	3.00±0.36(2.60-3.30)
10	8.90±0.86(8.40-9.90)	7.00±0.36(6.60-7.30)	6.00±0.65(5.30-6.60)

Mean ± SD (Range)

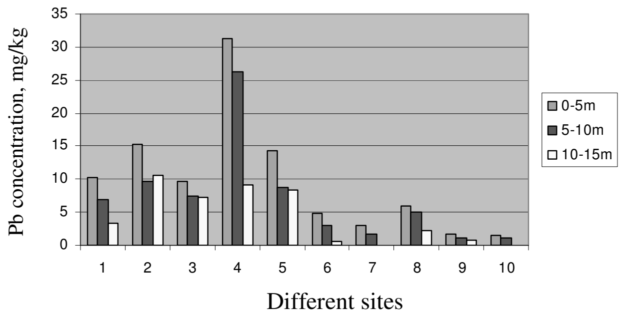

Figure 2. Pb (mg/kg) in roadside vegetable leaves

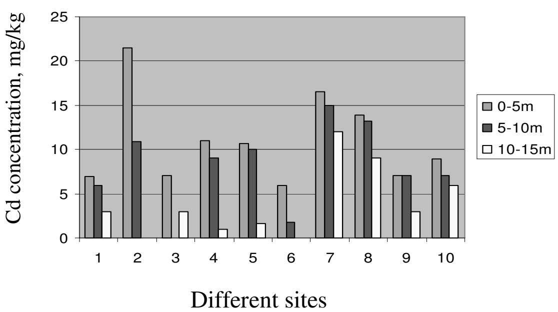

Figure 3. Cd (mg/kg) in roadside agricultural soils

Cadmium

Cd levels in exhaust emissions have been related to the composition of gasoline, motor oil, car tires and roadside deposition of the residues of those materials as well as traffic density[40]. All the sites investigated had Cd above the recommended 1-3 mg/kg limit given by EU.

(Table 4 & Figure 3) Highest concentration of Cd in soil (site 2) at distance 0-5 m from road was 21.53 mg/kg which is decreased to (about 50%) 10.90 mg/kg (5-10 m) & 0.00 mg/kg (10-15 m). The lowest concentration of Cd in soil (site 6) at distance 0-5 m from road was 6.00 mg/kg which is decreased to 1.80 mg/kg (30%) at distance 5-10 m and followed by 0.00 mg/kg at 10-15 m from road. However, the sources of Cd in the urban areas are much less well defined than those of Pb, but metal plating and tire rubber were considered the likely sources of cadmium[41]. In the absence of any major industry in the sampling sites, the levels of cadmium could be due to lubricating oils and/or old tires that are frequently used and the rough surfaces of the roads, which increase the wearing of tires.

Correlation calculations performed on Cd concentrations in soil showed that significant negative correlation are present between Cd content and distance from road at all sites except at site 2 & at site 3. These were from $r = -0.18$, $p > 0.05$ (at site 2) to $r = -0.98$, $p < 0.01$(at site 10). The mean values of Cd of all soil sampled at different distances away from the highway was significantly different ($p < 0.01$). 0-1 mg/kg of Cd in soils indicates non-contamination, 1-3 mg/kg indicates slight contamination and 3-10 mg/kg indicates a contaminated soil[43]. At 0-5 m from road, all the soil samples contained more Cd than 3 mg/kg and could be considered as contaminated.

It is well known that concentrations of Cd in edible vegetables range from 0.05 to 0.9 mg/kg (dry weight) and leafy plants such as cabbage, spinach contains relatively higher Cd than grain or fruit plant such as apple and barley[42]. Table 5 & and Figure 4 shows the samples collected at site 6, which displayed the highest Cd content at distance 0-5 m from road (2.90±0.20 mg/kg).

Table 5. Cadmium (mg/kg) in vegetable leaves of selected sites

Site	Vegetable	Distance from road (m)		
		0-5 m	5-10 m	10-15 m
1	Cauliflower	1.20±0.10(1.10-1.30)	0.60±0.10(0.50-0.70)	0.00±0.00(0.00-0.00)
2	Cabbage	0.30±0.00(0.30-0.30)	0.27±0.05(0.20-0.30)	0.00±0.00(0.00-0.00)
3	Cauliflower	1.20±0.10(1.10-1.30)	0.20±0.10(0.10-0.30)	0.10±0.000.10-0.10)
4	Brinjal	1.00±0.10(0.90-1.10)	0.80±0.00(0.80-0.80)	0.00±0.05 (0.30-0.40)
5	Brinjal	1.30±0.05(1.00-1.10)	0.70±.0.00(0.70-0.70)	0.77±0.11(0.70-0.90)
6	Radish	2.90±0.20(2.70-3.10)	2.30±0.28(1.70-2.20)	1.83±0.20(1.60-2.00)
7	Spinach	0.50±0.00(0.50-0.50)	0.23±0.05(0.20-0.30)	0.00±0.00(0.00-0.00)
8	Spinach	0.90±0.10(0.80-1.00)	0.63±0.05(0.60-0.70)	0.53±0.05(0.50-0.60)
9	Okra	0.53±0.05(0.50-0.60)	0.10±0.00(0.10-0.10)	0.00±0.00(0.00-0.00)
10	Radish	1.63±0.11(1.50-1.70)	0.00±0.00(0.00-0.00)	0.00±0.00(0.00-0.00)

Mean ± SD (Range)

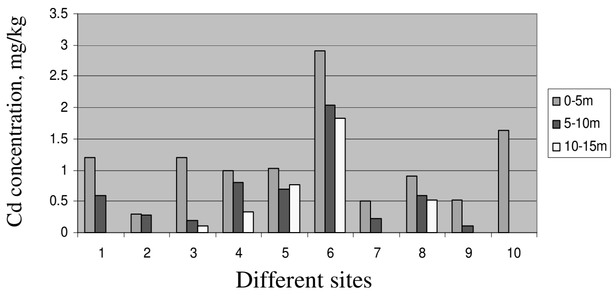

Figure 4. Cd (mg/kg) in roadside vegetable leaves

It is decreased to about 70% (2.03±0.28 mg/kg) at distance 5-10 m & to about 63% (1.83 ± 0.20 mg/kg). The lowest Cd concentration in cabbage leaves at 0-5 m distance from road was found; 0.30 ± 0.00 mg/kg (site2) which is decreased to 0.27 ± 0.58 mg/kg (at 5-10 m) & 0.00±0.00 mg/kg (at 10-15 m). Simple correlation analysis revealed negative correlations between Cd concentration and distance from road, these were $r = -0.75$, $p< 0.05$ (site5) to $r= -0.99$, $p <0.01$ (site7) except that of site 1; $r = -1.00$, p = ERR. The mean values of Cd content from all the vegetables sampled at different distances away from the highway were significantly different ($p<0.01$). The Cd concentration in all the vegetable samples were within the Indian (Prevention of food Adulteration act (PFA) permissible limit of 1.5 mg/kg except the sample of site 6 (2.90±0.20 mg/kg) and of site 10 (1.63 ± 0.11 mg/kg). But all the samples accepted the much more stringent (0.2 mg/kg).

Inter-elemental relationships provide interesting information related to heavy metal sources and pathways. Pearson's correlation coefficients of heavy metal elements in soil and vegetable samples are presented in Table 6 and Table 7 respectively. Table 6 shows that Pb and Cd in soils are significantly positively correlated, suggesting a common origin. As the sampling areas have no industry, we may assume that the heavy metals analyzed derive mostly from motor traffic on the motorway from site 1 to the site 10.

Table 6. Correlation matrix of soil Pb and Cd

Parameter	Pb	Cd
Pb	1	0.83(p<0.01)
Cd		1

Table 7. Correlation matrix of vegetables Pb and Cd

Parameter	Pb	Cd
Pb	1	-0.06(p>0.05)
Cd		1

In case of plant samples, correlation calculations between Pb and Cd gave non significant correlation coefficient. This may indicate that heavy metal pollution in roadside vegetable leaves may be due to other sources in addition to vehicular exhaust.

Conclusion

From the previous discussion, the following conclusions may be drawn:

1. The levels of heavy metals (Pb & Cd) contamination in both soils and vegetables, decreased to background levels with distance on either side of the highway. The decrease of elemental concentrations with distance from highway would indicate aerial deposition of metal particulates in road side environment from extraneous sources and not a function of soil type. In Agra motor vehicles that burn leaded gasoline are mostly responsible for the build up of Pb & Cd in soil and vegetables along the highway through the emissions of particulates and wearing of tires. This conclusion agrees with that of Rodriguez- Flores and Rodriguez – Castellon (1982), who reported that the Cd and Pb levels in soil and vegetables decreased with increasing distance from the road[44].
2. The concentrations of Pb, especially in soil, exhibited a larger variation with distance from the road than those of Cd. This may be explained by the relatively higher background values of Cd in the samples.
3. The roadside environment had a significantly high content of heavy metals (Pb & Cd), especially Pb and the mean values of both these metals in soil and vegetables were significantly different at away from the highway ($p<0.01$).

4. In terms of environment hazards and polluted city environment, it is suggested that the study on heavy metal contamination in soils and in several crops, especially those grown along the main road, should be conducted.

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