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Long-Term Operation of Brick-Kilns Led Heavy Metal Contamination of Soil-Plant-Animal Continuum in Kashmir Himalayas

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ABSTRACT

Brick kiln operation in Kashmir has One Health implications. The present study focused on the influence of brick kilns on Soil-Plant-Animal system vis-à-vis Pb, Cd and Cr. Lead concentration in soil, paddy straw, rice bran and cow milk was significantly higher ($P<0.05$) in areas closer to brick kilns. However, majority of the sera samples showed Pb in below detection limits. Cadmium concentration in soil and paddy straw representing area upto 500 meter distance from the kiln site was higher than the faraway and control sites. Chromium concentration in all the study samples save rice bran was significantly ($P<0.05$) higher in areas closer to brick kilns. Lead and Cr concentration in milk (from cows residing upto 500 meters) exceeded the maximum permissible levels. The study concluded that continuous operation of brick kilns and long duration exposure of Soil-Plant-Animal system to such ill influence affects the overall health of the continuum.

HIGHLIGHTS

- The influence of brick kilns on Soil-Plant-Animal system was studied in a village having the presence of functional brick kilns.
- Maximum permissible level of Pb and Cr in milk of cows influenced by long term and scientifically ill designed brick kiln functioning poses a significant public health risk.

Keywords: Brick Kilns, Heavy Metals, Soil-Plant-Animal System, Public Health Risk

Environmental pollution is a major global problem posing serious risk to man and animals. Development of modern technology and rapid industrialization are among the foremost factors for environmental pollution. The environmental pollutants are spread through different channels, many of which finally enter into food chain of livestock and man (Kaplan *et al.,* 2010). Heavy metal toxicity is one of the major current environmental health problems and potentially dangerous because of bio-accumulation through food chain (Aycicek *et al.,* 2008). Due to increase in industrial activity, pollution from heavy metals has grown widely throughout the world since the late 19th and early 20th centuries (Alimardan *et al.,* 2016).

Brick production is a very large and traditional industry in many parts of Asia. The brick sector in India, although unorganized, is tremendous in size. India is the second largest brick producer in the world (Maithel *et al.,* 2003). Brick kilns are widespread in Kashmir valley (J&K-India) resulting in environmental degradation due to emissions of significant quantities of particulates and gaseous pollutants (Skindar *et al.,* 2014). Brick kilns emit a variety

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of harmful gases and heavy metals like Pb, Cd, Cr, Hg etc as well as suspended particulate matter during brick production (Asgher and Singh, 2003; Ahmed *et al.,* 2008; Assadi *et al.,* 2011; Ismail *et al.,* 2012). Fly ash is also an important polluting agent containing heavy metals such as Pb, Cd, Hg and Zn (Luo *et al.,* 2009).

Furthermore, release of harmful gases from brick kilns to the environment alters the natural cycles such as nitrogen cycle and other natural processes which results in decreased fertility and nutrients in soil (Krishna and Govil, 2007). Hazardous elements may accumulate in soils around brick kiln area from industrial activities, fuel and coal combustion, wood burning, rubber tyre burning and furnace oil in the brick kiln (Alam *et al.,* 2009; Wei *et al.,* 2010; Wuana *et al.,* 2011). Heavy metals and fly ash both fall down to soil due to gravity after emission. From brick kilns, ambient hazardous elements are being brought forward every year which accumulate in soils causing toxicity (Khan *et al.,* 2006). Transfer of heavy metals from soil to agricultural crops causes food pollution (Lai *et al.,* 2010).

Human beings and other animals suffer damaging effects due to chronic intake of heavy metals (Lai *et al.,* 2010; John *et al.,* 2011). Heavy metals like as Cr, Ni, As, Cd, Hg and Pb have been considered as the most toxic elements in the environment by the US Environment Protection Agency (Lei *et al.,* 2010). The damage associated with Pb, Cd and Cr is of great concern throughout the world because of their toxic and mutagenic effects even at low concentration (Das, 1990). Heavy metals in the body lead to nervous system disorders, renal failure, genetic mutations, various types of cancers, respiratory and cardiovascular disorders, immune system weakening and infertility (Aazami *et al.,* 2017). Many toxic metals exert their bad effects by distressing the enzyme systems of animals by binding to specific enzymes and proteins necessary for cellular function. These also compete with other elements essential for maintenance and function of cells resulting in mineral deficiencies (Davis, 2010). After competing with the minerals, heavy metals render them unavailable to body leading to ill-health (Dodd *et al.,* 2013), for example, ingestion of Pb leads to competitive inhibition of selenium uptake and diminishing its absorption by up to 26% (Neathery *et al.,* 1987). Toxic metals also exert their effects by interfering with the production, release, biological action and metabolism of hormones (Zacharewski, 1998). Tissue damage or even mortality might occur as a result of heavy metal accumulation in the biological tissues and body fluids of animals that are fed on plants grown in soils with industrial pollution (Kaya *et al.,* 2002; Beskaya *et al.,* 2008). It is necessary to monitor and assess the extent of aggregate exposure to heavy metals concerning different environmental media and pathways to understand the relationship between the concentration of trace elements in soil, plants, water and animal system (Sharma *et al.,* 2007; Ahmad *et al.,* 2013). Moreover, heavy metals can be vertically transmitted to offsprings in dairy cattle and have potential toxicity from view point of public health (Simsek *et al.,* 2000). Highest heavy metal content has been found in milk samples collected from industrial region (Simsek *et al.,* 2000). Heavy metals except Zn have been found in toxic limits to an extent of 100% in milk samples of dairy cattle consuming fodder grown in land irrigated by polluted water (Raj *et al.,* 2006), and monitoring their concentration periodically may be useful to increase the productivity of cows (Ekicii *et al.,* 2015). So, from „One Health" point of view it is important to assess and monitor heavy metal contamination of soil-plant-animal continuum and examine various sources of contamination (Saghaei *et al.,* 2012; Ekicii *et al.,* 2015).

In Kashmir valley, livestock productivity and production is not up to their potential, and multiple causal factors may be the reason. Widespread and unorganized operation of brick kilns since long may also be responsible for heavy metal accumulation/pollution. Considering the Kashmir scenario, literature is also scant on influence of brick kilns on soil-plant- animal system with respect to heavy metals. The present study was endeavoured to study the influence of brick kiln operation on soil-plant-animal system in District Kulgam of Kashmr valley (J&K- India) vis-a- vis selected heavy metals. A site housing three decade old brick kilns in close proximity to each other was selected in village Hablishi of District Kulgam, and an agro-geo-climatically similar site in village Tankipora some 1.8 kilometres away from brick kiln was selected to serve as matched control (D0). Agricultural land adjacent to the brick kilns used for cultivating livestock feed and fodder and rice for human consumption was divided into three zones [D1 (0-250 m), D2 (250-500 m) and D3 (500-750 m)] for studying the variation in pollution with respect to distance from brick kilns.

MATERIALS AND METHODS

Study area and outline of the study

To assess the effect of brick kilns on Soil-Plant-Animal system, the study was conducted in Hablish village (33°39′41″N 75°06′26″E) of district Kulgam, Jammu & Kashmir (Fig. 1), with the assumption that the overall health of soil- plant-animal continuum/system is influenced by the ill effects of decade old brick kilns. The area was selected as the brick kilns had a decade long operational history and an agricultural land in close proximity of the brick kilns used for cultivating fodder for livestock. The area is livestock rich with no history of flooding in the last decade. All the parameters studied were compared with a matched control population in village Tankipora, Kulgam (33°39′53.2″N 75°05′26.3″E), a nearby area having similar agricultural and livestock rearing practices. In first phase of study, the two areas (Brick kiln area and control area) were surveyed. Soil, fodder, bran, milk and blood samples were collected, labeled, stored and brought to lab of Research Centre for Residue and Quality Analysis, Division of Entomology, SKUAST-Kashmir. In second phase, the collected samples were processed and analyzed for selected heavy metals.

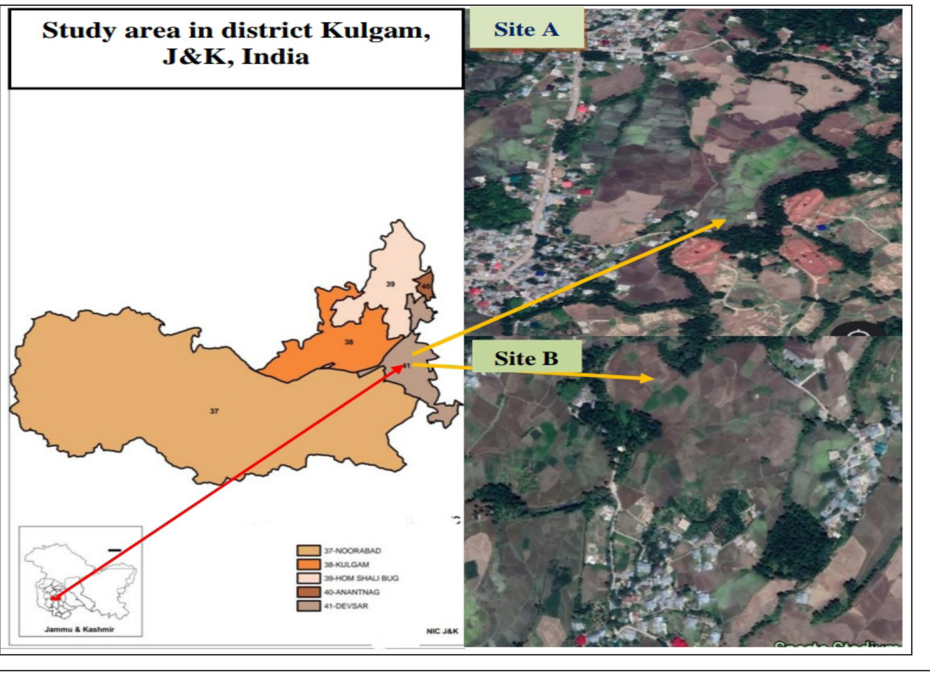
Study design

The agricultural land in the vicinity of brick kilns (source) was divided into 03 categories/zones as mentioned below, and samples (n=6) like soil, paddy straw, rice bran, cattle blood and milk were collected with equal representation to each zone:

i. Up to 250 meters from the source.

ii. 250 to 500 meters from the source.

iii. 500 to 750 meters from the source.



Study Location	Sampling Site	Geographical coordinates	Fodder/Crop/ grown	Number of Brick kilns	Age of Brick kilns
Hablish, Kulgram (Brick kiln area)	A	33°39’38.3’’N 75°06’15.3’’E	Paddy, Oats	3	10-11 years
Tankipora, Kulgram (Control area)	B	33°40’00.0’’N 75°05’23.4’’E	Paddy, Oats	0	0

*Distance between brick kiln control = 1.8 kilometers

Fig. 1: Study area showing brick kiln and control area in district Kulgram, J&K

Dairy cattle in early lactation having had the history of more than one year exposure and fed with paddy straw and rice bran obtained from the brick kiln affected agricultural land were only included in the study. Area having similar agricultural and livestock rearing practices, but no presence of brick kilns within a minimum of 1500 meters radius was selected as control area for comparison purposes.

Laboratory analysis

Soil, fodder, rice bran, serum and milk samples were collected as per standard procedures and digested as described by Maurya *et al.,* 2018; Jones *et. al.*, 1991; AOAC, 2000; Kolmer *et al.,* 1951; Richards, 1968, respectively. Heavy metals (Pb, Cd, Cr) were analyzed by Atomic Absorption Spectrometer (AAS-Agilent Technologies 200 series AA) at Research Centre for Residue and Quality Analysis, Division of Entomology, SKUAST- Kashmir, Shalimar, Srinagar.

Statistical analysis

The data obtained was analyzed by factorial Completely Randomized Design experiment using "R" software. The relationship between different parameters was determined by calculating Pearson's coefficient of correlation.

RESULTS AND DISCUSSION

The average heavy metal concentration in soil, paddy straw, rice bran, cattle serum and cow milk in the present study area (Table 1) revealed higher concentration of these metals in brick kiln areas and showed diverse pattern/ variation with respect to distance from the brick kilns. Average lead concentration in soil, paddy straw, rice bran and cow milk was significantly higher in areas closer to brick kilns than the reference area. Average concentration of lead in soil of brick kiln and reference area was higher than the maximum permissible levels (6.0 mg/kg). In paddy straw (D1 area; 6 .15±0.38 mg/kg) and milk (D2 area; 0.74±0.30 mg/kg and D1 area; 0.57±0.29 mg/kg) the concentration of lead exceeded maximum permissible level (5 mg/kg and 0.2 mg/kg respectively). In majority of serum samples, lead concentration observed was in below detection limits. Lead concentration in soil, paddy straw, rice bran and milk was positively correlated. Average soil cadmium concentration in D1 area (0.56±0.01 mg/ kg) was comparable to D2 area (0.52±0.01 mg/kg) and was significantly higher than D0 (0.44±0.01 mg/kg) and D3 (0.40±0.01 mg/kg) areas. Cadmium concentration in paddy straw (D2 and D1), rice bran (D1) and milk (D2) was comparatively higher than rest of areas. However, the cadmium concentration of all the samples did not exceed the maximum permissible levels. Average soil chromium concentration was significantly higher in D1 (22.50±0.73 mg/kg) and D2 (22.57±1.30 mg/kg) compared to D3 (17.28±1.08 mg/kg) and D0 (15.49±0.21 mg/kg). Similar trend was observed for chromium concentration in paddy straw, cattle serum and cow milk. All the straw samples in D3 and majority in D0 plus majority of the rice bran samples irrespective of the area showed chromium concentration in below detection limits. Chromium concentration in soil of all the areas, in paddy straw of D1, in serum of D1, D2, D3 and in milk of D1 and D2 areas exceeded the maximum permissible levels.

The average Pb, Cd and Cr concentration in soil, paddy straw and rice bran in present study was found comparatively higher in areas closer to brick kilns (D1>D2) than those that are farther from brick kilns (D3) and control area (D0). Variation in concentration of Pb, Cd, and Cr with distance from brick kilns might be attributed to wind direction and velocity dependent fallout of pollutants/ particulates on agricultural land. The higher concentration of Pb, Cd and Cr in brick kiln areas might be due to burning of huge amount of coal and low quality fuels like rubber tyres and other waste in non scientific ways (Wuana and Okieimen, 2011). Heavy metals have been found in solid particles in smoke from tyres and in other emissions (Little and Martin, 1972). The accumulation of heavy metals is soil also depends on factors like physiological properties of soil and atmospheric deposition of heavy metals influenced by environmental conditions such as temperature, moisture, wind velocity (Sharma *et al.,* 2009). The higher concentration of heavy metals in paddy straw and rice bran might also be due to aerial deposition of ash and dust particulates produced during combustion of various fuels in brick kiln areas (Wang *et al.,* 2003). Other possible reasons for presence of heavy metals in plants include application of fertilizer and pesticides in agriculture soil (Herawati *et al.,* 2000). Plants take different metals through roots and also through stomatal opening, get dissolved in cell sap and circulate through entire plant

Table 1: Concentration (mg/kg) of heavy metals (Mean±SE) in Brick Kiln/control area

Metal	Area	Soil	Paddy straw	Rice Bran	Cattle Serum	Cow Milk
Pb	D_0	13.29±0.71 (1.12^{c})	2.33±0.51 (0.42^{h})	2.58±0.28 (0.48^{gh})	0.00±0.00 (0.30^{i})	0.11±0.11 (0.26^{i})
	D_1	26.73±1.29 (1.42^{a})	6.15±0.38 (0.81^{d})	4.52±0.26(0.71^{e})	0.00±0.00 (0.30^{i})	0.57±0.29 (0.23^{d})
	D_2	22.50±1.44 (1.35^{ab})	3.76±0.30 (0.62^{ef})	4.64±0.58(0.70^{e})	0.02±0.02 (0.283^{i})	0.74±0.30 (0.21^{c})
	D_3	18.87±0.35 (1.27^{b})	4.04±0.45 (0.65^{ef})	3.18±0.29(0.56^{fg})	0.00±0.00 (0.30^{i})	0.12±0.12 (0.26^{i})
	MPL	6 (FAO/WHO, 2001)	5 (WHO/FAO, 2007)	1 – 6 (Suttle, 2010)	0.01-0.2 (Puls,1988)	0.2 (FSSAI, 2011)
Cd	D_0	0.44±0.01 (0.22^{e})	0.12±0.07 (0.28^{abcd})	0.07±0.05 (0.28^{ab})	0.00±0.00 (0.29^{a})	0.02±0.01 (0.28^{ab})
	D_1	0.56±0.01 (0.19^{f})	0.22±0.05 (0.26^{cd})	0.15±0.02 (0.27^{abcd})	0.02±0.01 (0.28^{abc})	0.04±0.02 (0.27^{abcd})
	D_2	0.52±0.01 (0.20^{ef})	0.24±0.07 (0.26^{cd})	0.06±0.03 (0.29^{ab})	0.02±0.01 (0.28^{abc})	0.06±0.02 (0.25^{d})
	D_3	0.40±0.00 (0.22^{e})	0.11±0.05 (0.28^{abcd})	0.08±0.04 (0.28^{ab})	0.02±0.01 (0.29^{ab})	0.01±0.01 (0.29^{ab})
	MPL	3 (EU, 2002)	0.2 (WHO/FAO, 2007)	0.2 (WHO/FAO, 2002)	0.006-0.066 (Puls, 1988)	0.1 (FSSAI, 2011)
Cr	D_0	15.49±0.21 (1.19^{c})	0.23±0.23 (0.29^{e})	0.00 (0.30^{e})	0.07±0.04 (0.25^{e})	0.15±0.07 (0.19^{efg})
	D_1	22.50±0.73 (1.36)^{a}	4.46±1.20 (0.66^{d})	0.00 (0.30 ^{e})	0.54±0.05 (0.04^{h})	0.60±0.20 (0.12^{fgh})
	D_2	22.57±1.30 (1.34^{ab})	1.16±0.26 (0.28^{e})	0.00 (0.30^{e})	0.57±0.13 (0.11^{fgh})	0.43±0.11 (0.09^{gh})
	D_3	17.28±1.08 (1.23^{bc})	0.00±0.00 (0.30^{e})	0.09±0.09 (0.25)	0.35±0.11 (0.11^{fgh})	0.14±0.07 (0.21^{ef})
	MPL	1.3 (WHO,19 96a)	1.3 (WHO,19 96a)	3 (Mc Dowell *et al.* 1992)	0.006-0.066 (Puls, 1988)	0.008-0.250 (Puls, 1988)

Values along columns bearing different superscripts differ significantly ($P \leq 0.05$).

Data in Brackets represents transformed data.

D_0 (Control area); D_1 (0-250 m from brick kilns); D_2 (250-500 m from brick kilns); D_3 (500-750 m from brick kilns).

(Meagher, 2000). Heavy metal concentration in plant species depend on factors like the growing environment (pH, temperature, soil aeration), type of plants, size and root system, the availability of heavy metals in soil solution or foliar deposits, leaf morphology, soil moisture (Nagajyoti *et al.,* 2010). Particulate matter bound heavy metals are mobilised by air and deposited in irrigation water, agricultural/grazing soils, and transferred to plants and other parts of ecosystem, entering the trophic chain (Yilmaz *et al.,* 2009; Alloway, 2013). Similar results were observed by Achakzai *et al*. (2015) in relation to presence of heavy metals in soil and plants with respect to distance from brick kilns, however higher concentrations of Pb and Cd were found than the present study. Dey *et al*. (2017) observed higher heavy metal soil concentration nearer to brick kiln chimney. Bisht *et al*. (2015) observed highest concentration of Pb (11.82 mg/kg) and Cr (10.01 mg/kg) in soil at 50 meter distance from brick kiln. Sikder *et al*. (2016) observed comparatively higher concentration of Pb than the present study within 500 meter distance from brick kilns. The results of heavy metals in soil, paddy straw and rice bran in our study are also supported by Ismail *et al*. (2012), who noticed varied concentrations of Cd and Cr with respect to distance from brick kilns. Ishaq *et al.* (2010) observed comparatively lower concentration of Pb and Cd in soils influenced by brick kiln activity. Proshad *et al*. (2017) observed higher average Cr, Cd and lower Pb concentration in rice crop grown around brick kilns.

High concentration (more than MPL) of heavy metals (Pb and Cr) in milk of dairy cows in brick kiln areas (D1 & D2) compared to cows in D3 and control area and might be due to continuous intake of feed and fodder grown on brick kiln influenced agricultural land and endorse upward mobility of heavy metal pollutants through the food chain. More than the maximum permissible levels of Pb and Cr in milk of dairy cows reared in D1 and D2 study areas is quite dangerous from One Health Point of view and could pose a serious health risk to the population consuming such milk. Dzoma *et al.* (2010) concluded that the heavy metal levels in serum and faeces are generally influenced by environmental levels, and the occurrence of trace metals in grass and livestock specimens indicate the upward mobility of heavy metal pollutants through the food chain. Lead concentration in most of the serum samples observed were below detection limits and lower than that of in milk, which could be attributed to the fact that approximately 90% of the absorbed Pb is taken up by red blood cells and the remaining portion is largely bound to albumin (NRC, 2005), and to the metal retention (formation of bioactive lipophilic complex) property of milk (Leeuwen and Pinheiro, 2001). Swarup *et al*. (1993) observed higher levels of Pb and Cd in blood and milk of apparently healthy cows and buffaloes reared around industrial areas, and suggested possible accumulation of toxic metal residues in animal tissues. Ruminants are mostly affected with lead and cadmium ingestion due to the tendency of particulate matter to settle down in reticulum and get converted to soluble acetates in the acid medium (Radostits *et al.,* 1994). The high concentration of heavy metals in milk in the present study are in agreement with the findings of other authors who observed high Pb and Cd in blood and milk of cattle reared in a mining area (Chirinos-Peinado *et al.,* 2020). Similar findings (high Pb, Cd, Cr concentration in serum and milk of dairy cows fed fodder grown on polluted area/sewage farm) have also been observed by Somasundaram *et al*. (2005). In contrast to our findings high concentration of Pb and low concentration of Cd was observed by Ogundiran *et al.* (2012) in serum and milk of dairy cows reared in a polluted (Lead Slag) area of Nigeria.

CONCLUSION

Continuous emission of particulates and fly ash from brick kilns leads to environmental pollution and deterioration of quality of agricultural produce. Soil-Plant-Animal health is influenced by continuous operation of brick kilns due to persistence and mobility of heavy metals like lead and chromium, particularly in areas that are in close proximity (upto 500 meters) to the brick kilns. Long-term operation of brick kilns, and continuous exposure of dairy cows to brick kiln caused pollution leads to more than the maximum permissible levels of lead and chromium in milk, posing a serious public health risk. The study needs to be conducted beyond 750 meter distance away from the brick kilns, and at multiple locations of the valley with inclusion of more number of subjects, heavy metals and related elements for validation of the present study findings and better understanding of brick kiln induced health hazards.

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