

## HOTSPOTS OF FLUORIDE POLLUTION IN FIELDS AROUND THE COAL-FIRED BRICK KILNS

Rabail Urooj,<sup>a,b</sup> Sheikh Saeed Ahmad,<sup>b</sup> Muhammad Nauman Ahmad<sup>c</sup>

Quetta, Rawalpindi, and Peshawar, Pakistan

**ABSTRACT:** Development sector especially brick baking industry is dependent upon the burning of lignite coal in Pakistan. Due to lack of clean and green technologies, coal burning is ultimately source of pollution emission in atmosphere like NO<sub>x</sub>, SO<sub>x</sub> and fluoride compounds. Whereas, kilns are the main source of releasing fluoride pollution, as fluoride is naturally occurring in coal as well as in brick raw material. The fluoride pollution has phytotoxic effect to nearby growing plants. The objective of this research was to investigate the concentration of fluoride in the soil and crop samples of wheat and maize fields surrounded by kilns in Islamabad and Rawalpindi and to study the spatial distribution pattern of fluoride pollution using geospatial statistical technique, i.e., Getis-Ord. Geospatial statistical analysis was done in ArcGIS 10.2. Two hundred and fifty one samples were collected from different wheat and maize fields. Results of Getis-Ord revealed the significant clustering of high values of fluoride in Jhangi Syedan, Dhok Gujran, Chak jalal-ud-din, and Bara-Giran indicated by hot spots on the basis of lesser p value and high z score. Findings indicated that fields and crop plants nearer to the kilns were affected with the fluoride pollution as compared to the far distant fields. The crop samples in the year 2017 were found more damaged than the samples in the year 2015 and 2016. This temporal variation of fluoride pollution indicating the poor regulation of kiln industry. The result of the study further suggested that fields in vicinity of kilns need serious attention of government to prevent the damages caused by fluoride to crop plants.

Keywords: Fluoride; Foliar injury; Modeling; Probability.

### INTRODUCTION

Pakistan is facing different problems being an economically weak country. One of the common problems is increasing rate of pollution due to the urbanization and its effect on animals, humans, and plant's health.<sup>1</sup> The biggest source of pollution in South Asian countries are industries and automobiles.<sup>2</sup> About 91% of the world's population is breathing in places where the air pollution exceeds the guideline of the World Health Organization.<sup>3</sup> There are many air pollutants that are harmful not only to humans, but also for plants, in term of physical damages and altered physiological processes, include heavy metals, nitrogen and sulfur oxides, ozone and fluoride compounds.<sup>4,5</sup> Brick industry is not formally registered under any governmental institution in Pakistan. The accurate data about the number of operational kilns is also not available at governmental level. There is no proper guideline and policy pertaining to brick kiln. Even brick kiln sector not existing in list of projects which require approval from federal and provincial agencies before operating under IEE and EIA regulation 2000.<sup>6</sup> Demand and production of bricks for infrastructure development increasing day by day is another important triggering factor for the growing number of kilns in peri-urban areas. Ahmad et al. reported that poorly regulated kilns are growing along the peri-urban agricultural area, causing

<sup>a</sup>Environmental Science Department, Sardar Bahadur Khan Womens University, Quetta, Pakistan;

<sup>b</sup>Environmental Science Department, Fatima Jinnah Womens University, Rawalpindi, Pakistan;

<sup>c</sup>Agricultural Chemistry Department, University of Agriculture, Peshawar, Pakistan. \*For correspondence: Rabail Urooj, Environmental Science Department, Sardar Bahadur Khan Womens University, Quetta, Pakistan. Phone: 00923455276713. E-mail: rabailurooj@gmail.com

unrecognized environmental problem, not just in Pakistan but also in all other South Asian countries.<sup>7</sup> It has been reported in the year 2012 that almost 59 billion baked bricks are marketing annually in Pakistan by different kiln designs using different fuels include lignite coal, agrowaste especially rice husk, rubber tires, plastics, and varied industrial waste. Burning of all these fuels, results in the emission of toxic gases.<sup>8</sup> It's also reported that brick industry consume almost 40% of 4 million tons of locally produced coal.<sup>9</sup> The permissible level for particulate matter caused by coal burning in industries has been set by National Environmental Quality Standards at 500mg/cum, but this level has not yet been enforced on brick baking industry.<sup>6</sup>

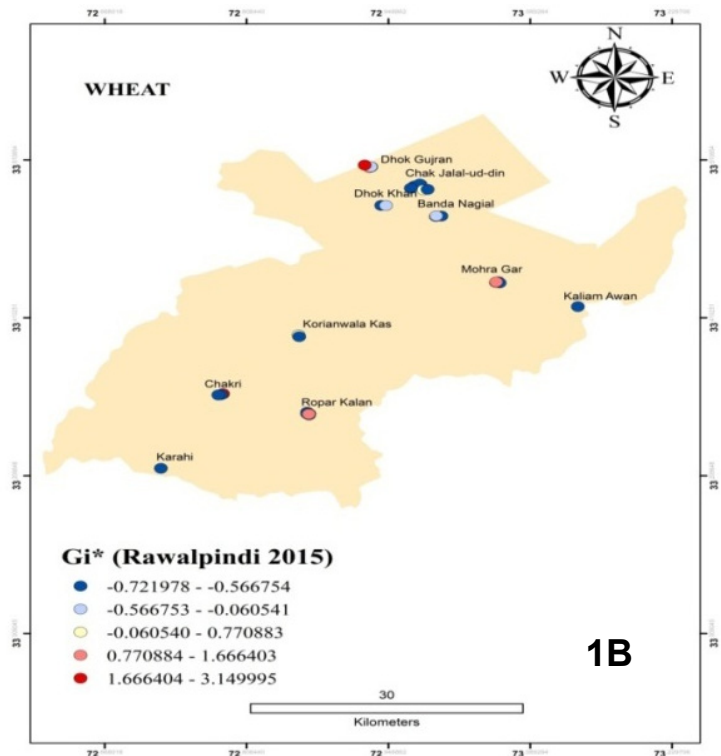
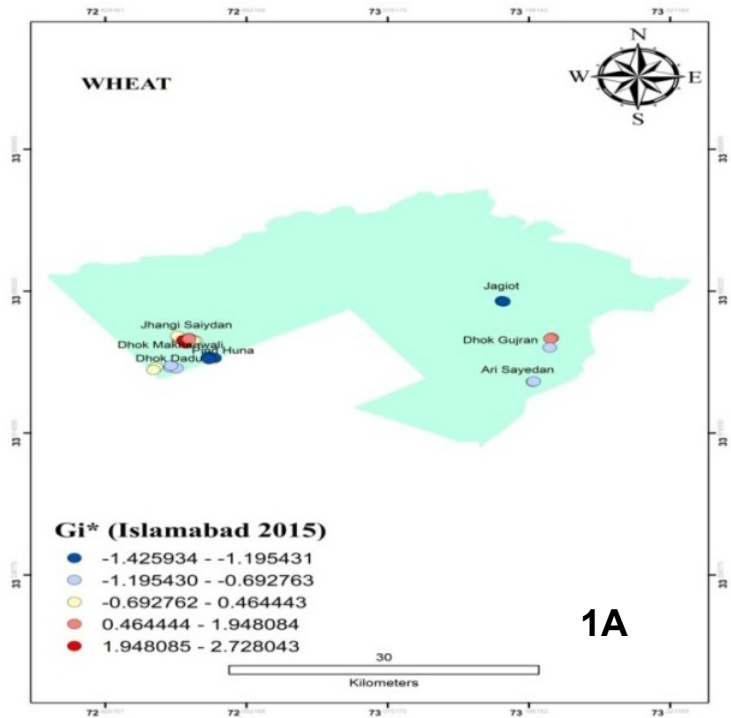
## MATERIALS AND METHODS

The selected study areas were the wheat and maize fields situated in Islamabad and Rawalpindi. Coal-fired kilns surrounded by wheat fields were marked with help of satellite remote sensing for present study. Whereas two hundred and fifty one samples of plants were taken from the wheat and maize fields and chemically analyzed using fluoride-sensitive electrode coupled to an ion-meter (model: crison glp 22+) in laboratory of Fatima Jinnah Women University. Resulting data was statistically analyzed using the ArcGIS build tool Getis-Ord  $G_i^*(d)$  which endorsed hotspots of fluoride in fields at different sites. Hotspot is form of clustering in a spatial distribution. In this study, adjacency is defined using Thiessen polygon continuity weight file which has been constructed based on villages that share common vertices. The output from  $G_i^*(d)$  statistic identifies spatial clusters of high values (hotspots) and spatial clusters of low values (cold spots).<sup>10,11</sup>

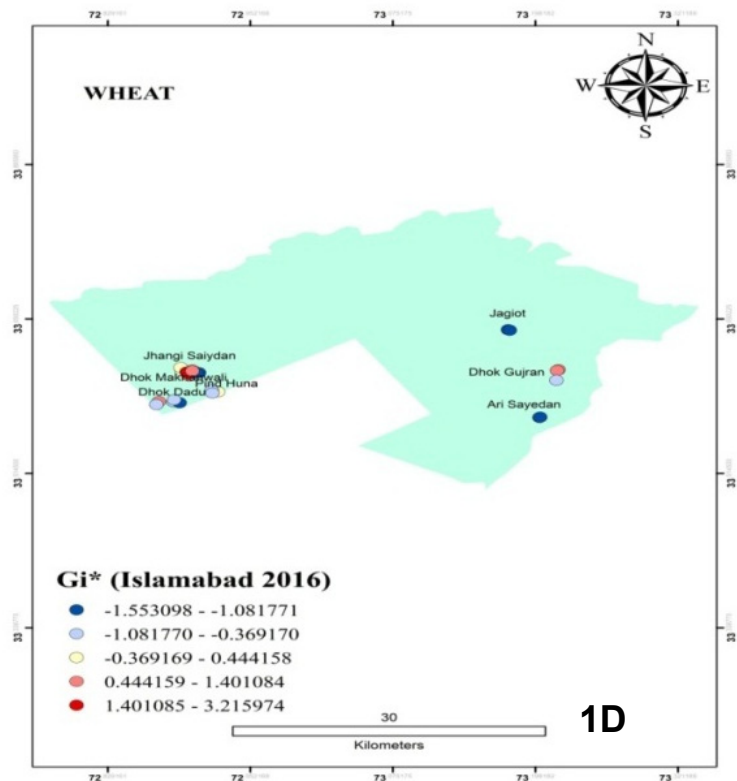
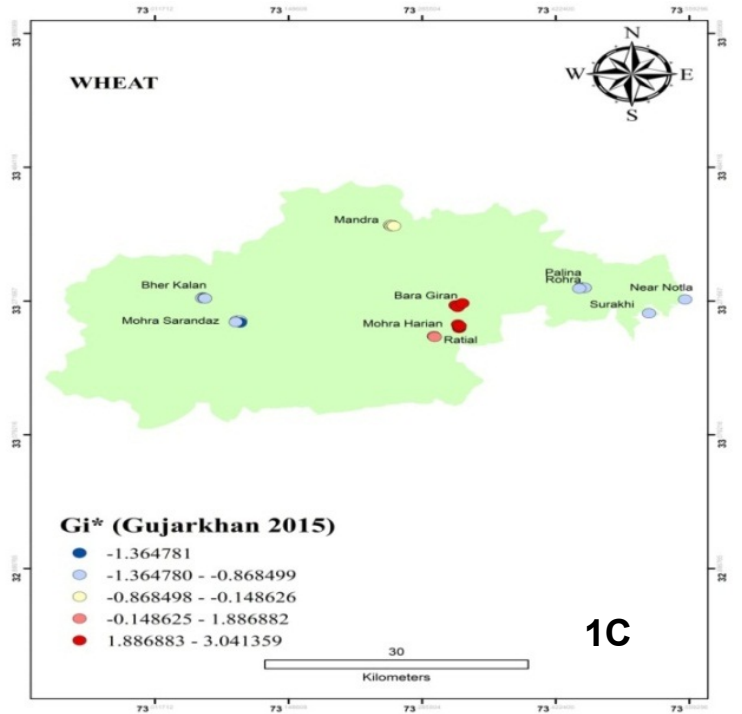
## RESULTS AND DISCUSSION

Total 82 out of 317 kilns were identified for research on the basis of required crop fields by using satellite image. Results of Getis-Ord statistical analysis revealed the hot spots and cold spots of fluoride concentration present in the plant's leaves of wheat and maize fields. Hotspot analysis indicated clustering and non-clustering of high and low values on the basis of p value and z score. Positive high z score and lesser p value indicated the statistically significant clustering of higher values shown by hotspot, while z values near to 0 indicated no clustering. Negative high value of z score presented statistically significant clustering of low values shown by cold spot. The results were quantified by graduated colour symbols presented in maps (Figures 1A–1F, 2A–2C, 3A–3F, and 4A–4C). In all generated maps light colour indicated mild clustering and dark colour presented intensity of clustering. Red colour represented hotspot area and dark blue colour represented cold spots on maps.

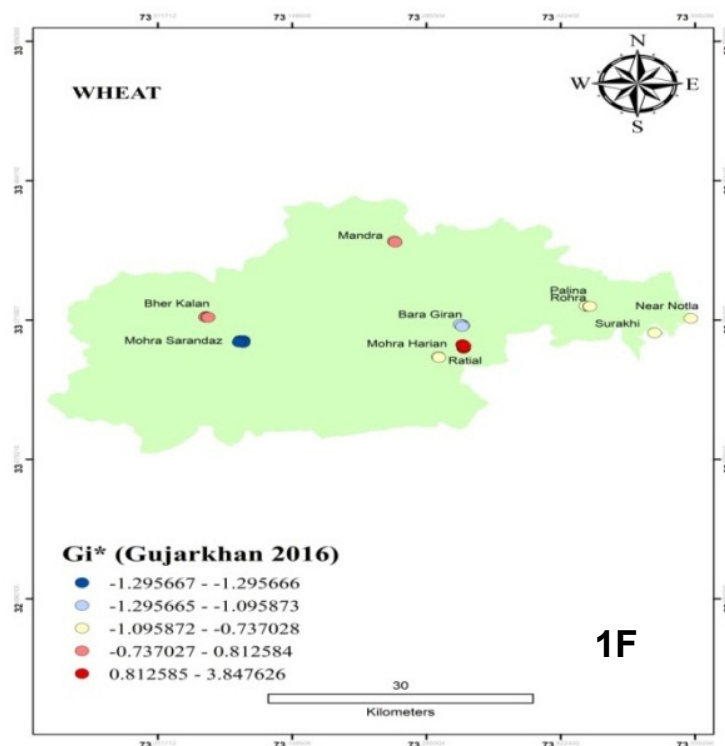
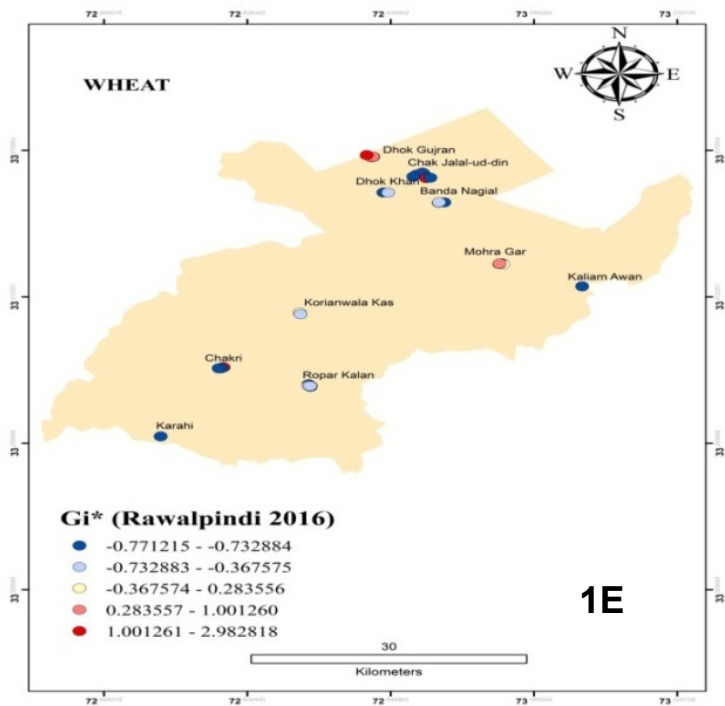
Different fluoride concentrations were found in wheat and maize plants sampled from different areas during the study years (2015–2017). Figures 1A–1F and 2A–2C showed a spatial pattern of fluoride concentration which was analyzed experimentally from wheat plants collected in three study years. These maps provided a more easy and useful representation of fluoride pollution distribution and also made easy for possible detection of fluoride pollution hotspot.



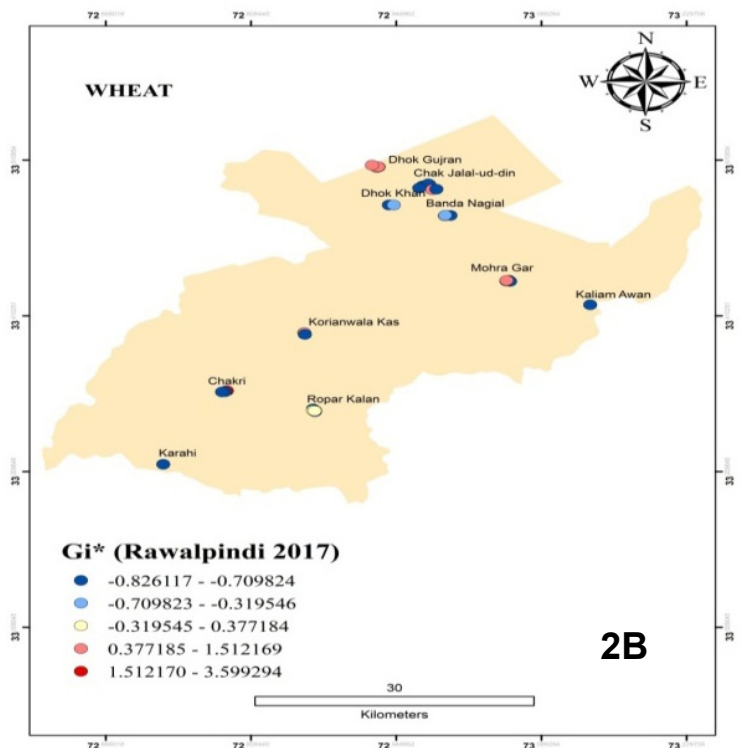
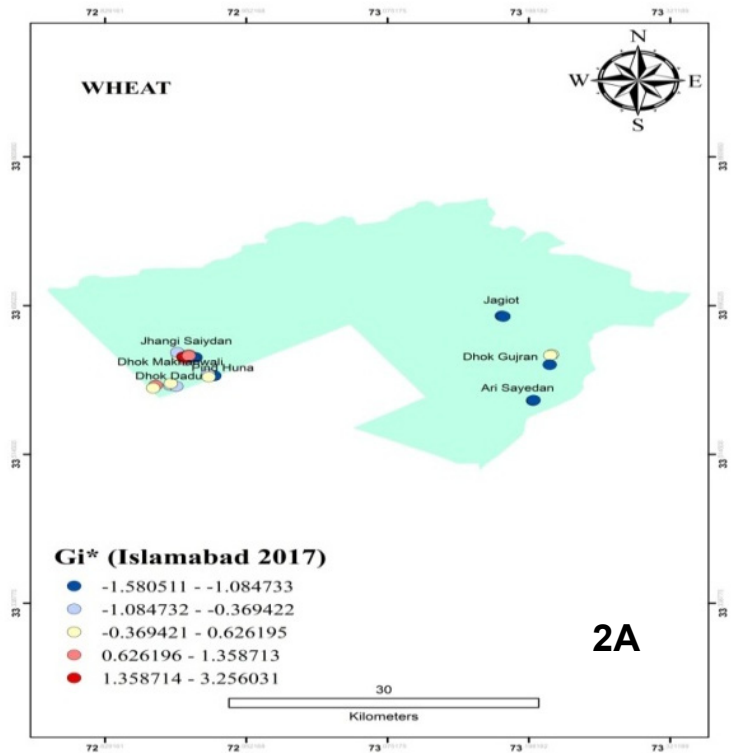
**Figures 1A and 1B.**  
Hotspot map of fluoride concentration in plants of wheat fields in the years 2015–2016. 1A: Islamabad 2015; 1B: Rawalpindi 2015.



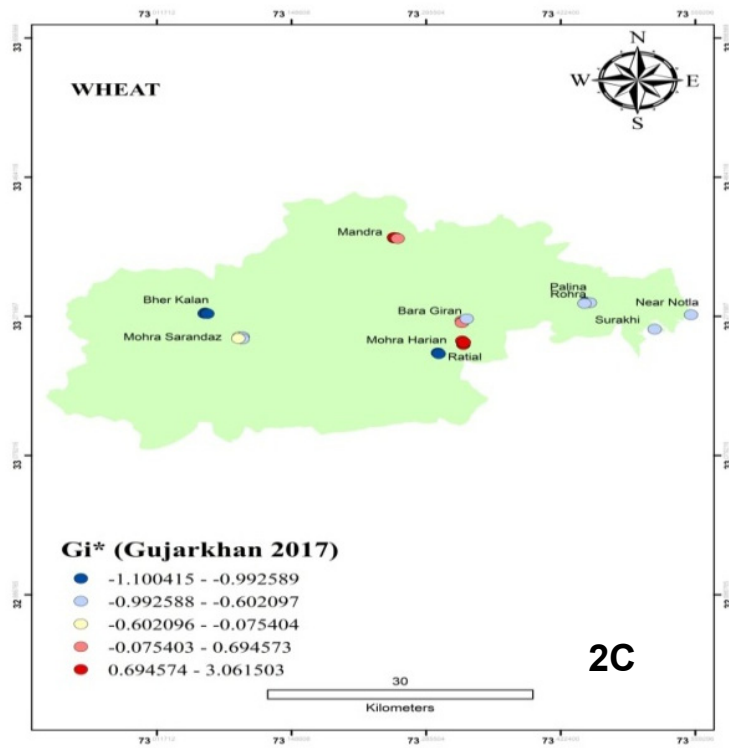
**Figures 1C and 1D.**  
Hotspot map of  
fluoride concentration  
in plants of wheat  
fields in the years  
2015–2016. 1C:  
Gujarkhan 2015; 1B:  
Islamabad 2016.



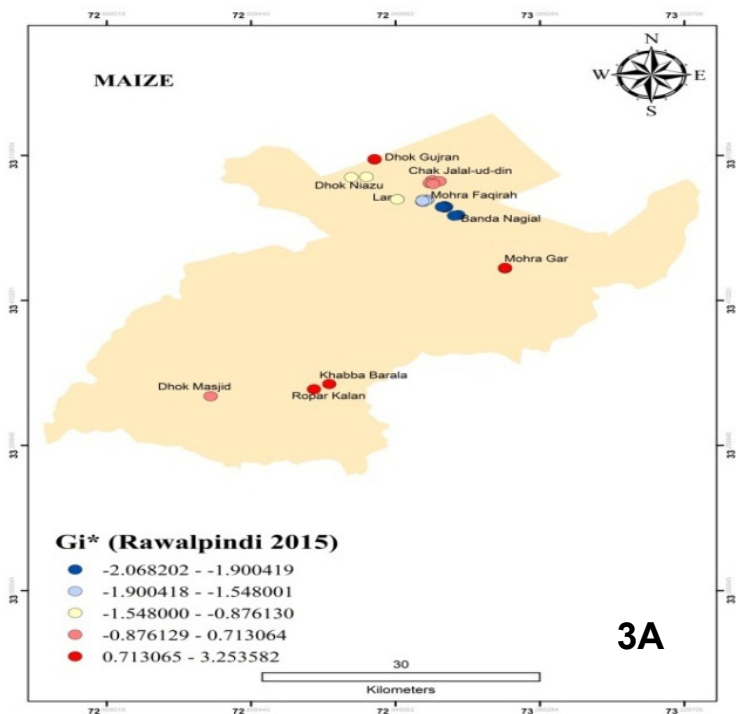
**Figures 1E and 1F.** Hotspot map of fluoride concentration in plants of wheat fields in the years 2015–2016. 1E: Rawalpindi 2016; 1F: Gujarkhan 2016.



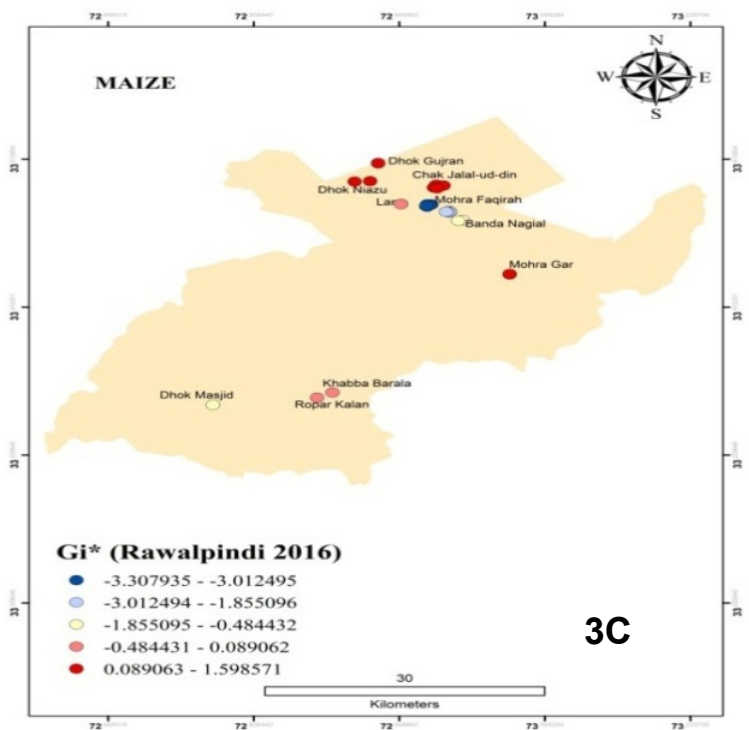
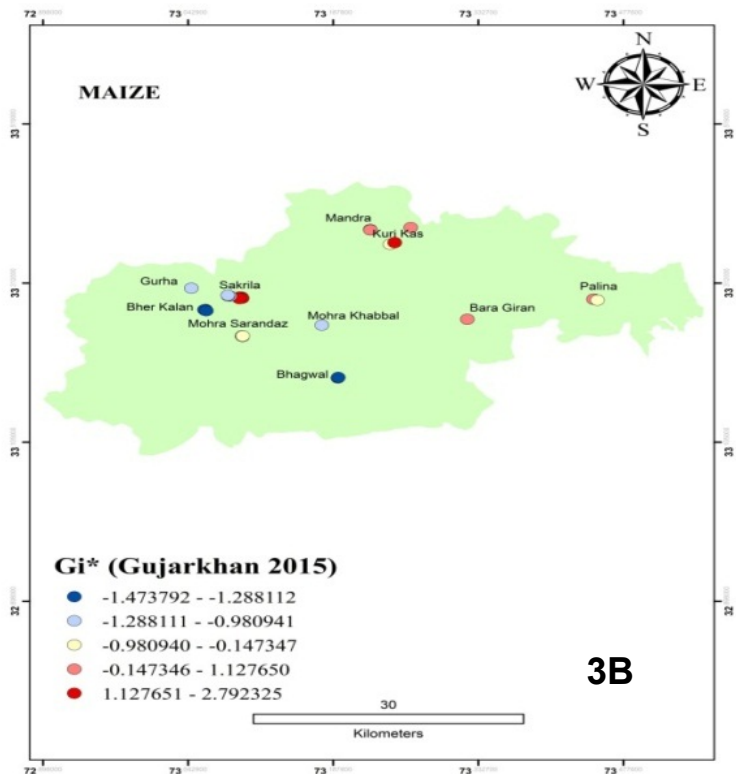
**Figures 2A and 2B.** Hotspot map of fluoride concentration in plants of wheat fields in the year 2017. 2A: Islamabad 2017; 2B: Rawalpindi 2017.



**Figure 2C.** Hotspot map of fluoride concentration in plants of wheat fields in the year 2017. 2C: Gujarkhan 2017.

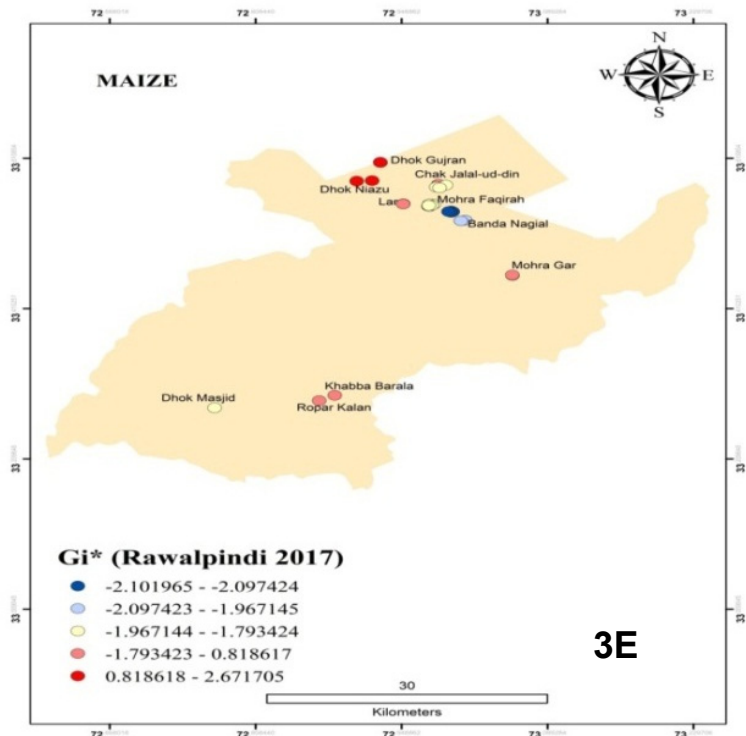
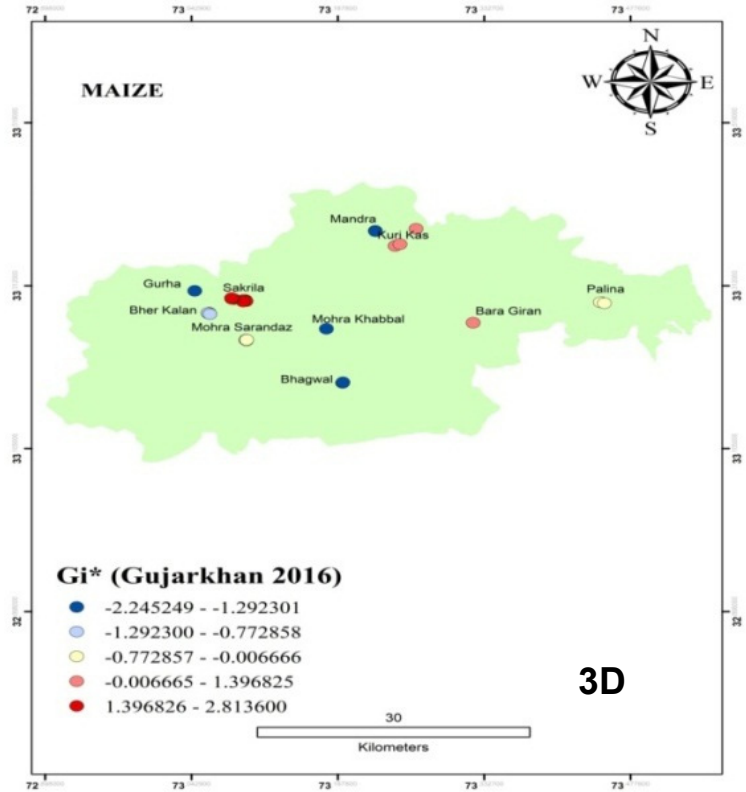


**Figure 3A.** Hotspot map of fluoride concentration in plants of maize fields in the years 2015–2017. 3A: Rawalpindi 2015.

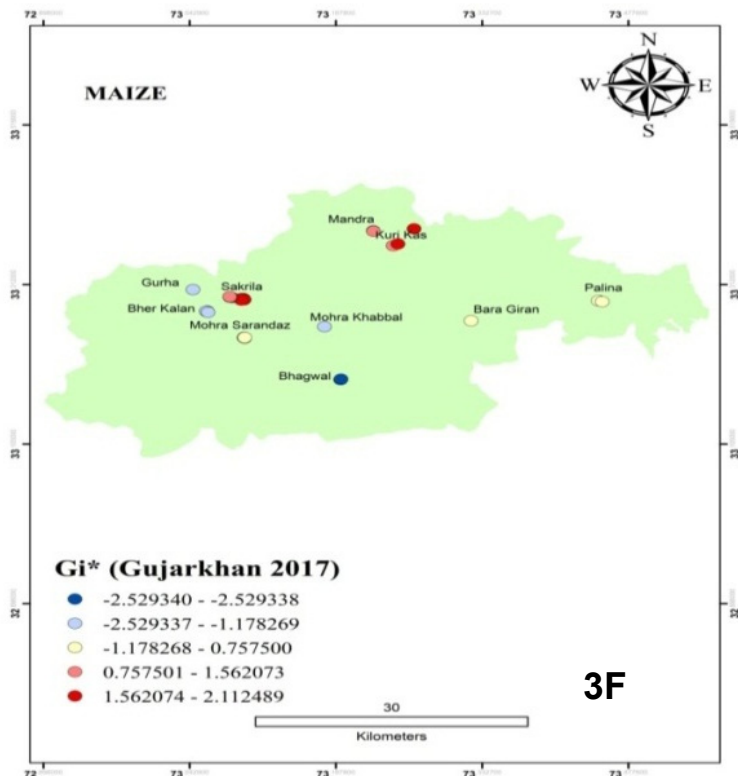


**Figures 3B and 3C.**  
Hotspot map of fluoride concentration in plants of maize fields in the years 2015–2017. 3B: Gujarkhan 2015; 3C: Rawalpindi 2016.

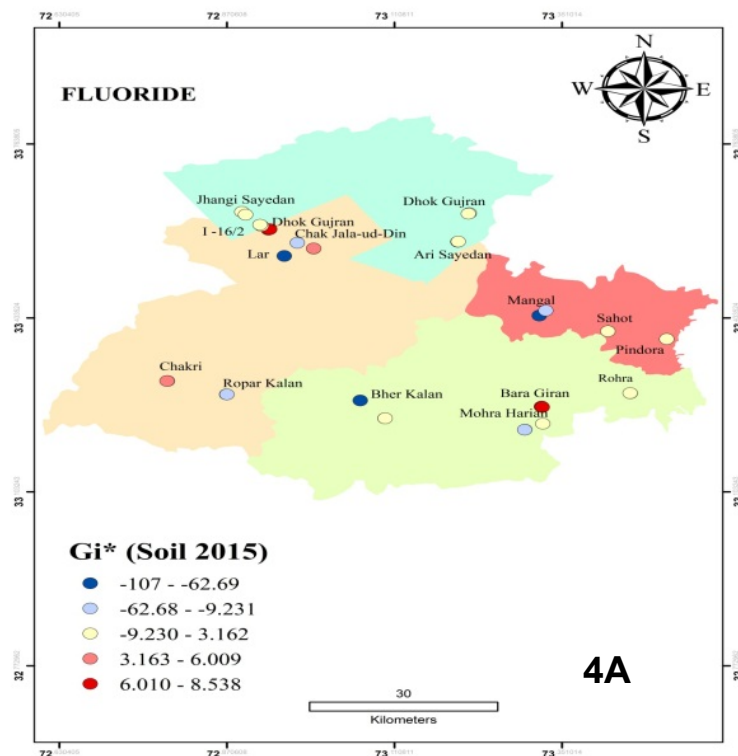




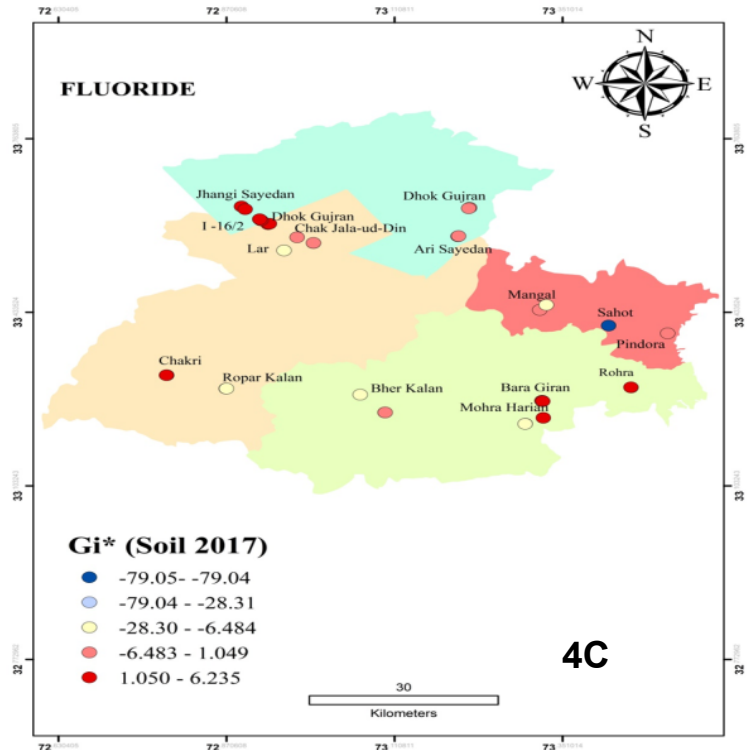
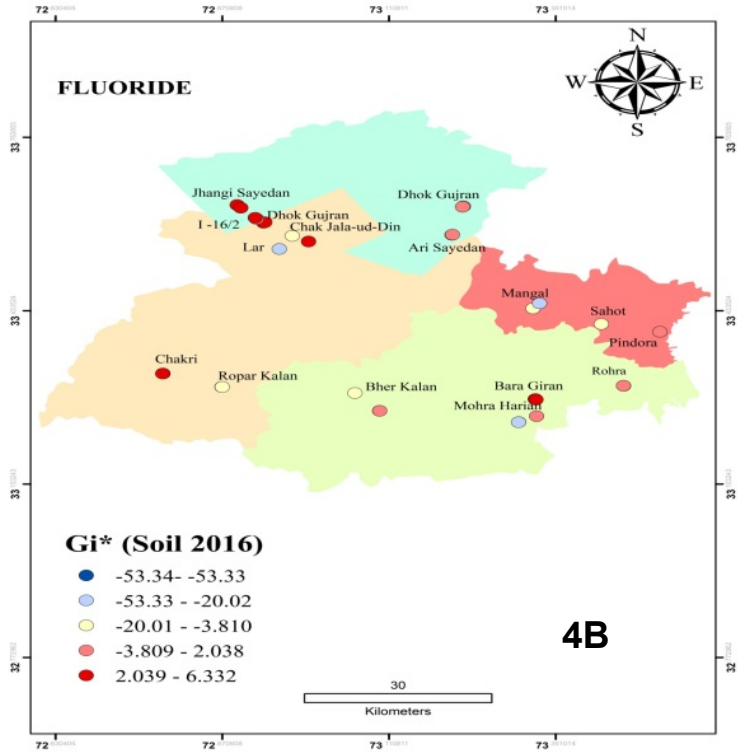
**Figures 3D and 3E.** Hotspot map of fluoride concentration in plants of maize fields in the years 2015–2017. 3D: Gujarkhan 2016; 3E: Rawalpindi 2017.



**Figure 3F.** Hotspot map of fluoride concentration in plants of maize fields in the years 2015–2017. 3F: Gujarkhan 2017.



**Figure 4A.** Hotspot map of fluoride concentration in soil during the years 2015–2017. 4A: Soil 2015.



**Figures 4B and 4C.**  
Hotspot map of  
fluoride concentration  
in soil during the years  
2015–2017. 4B: Soil  
2016; 4C: Soil 2017.

Findings of Hotspot analysis revealed that in the year 2015, fields of Tarnol and G-16/2 located in Islamabad were found as a hot spot with z score ranging from 1.9–2.7 at the threshold distance 700 m (Figure 1A). During the same study year, wheat fields located in the Chakri, Dhok Makhanawali, and Garja road Rawalpindi were found as hotspot at the threshold distance 50 m with z-score value ranging 1.6–3.4 (Figure 1B). Bara-giran and Gujarkhan near the Grand Trunk (GT) road in Tehsil Gujarkhan (Figure 1C) were found as hotspot field areas around the operational kiln. Fluoride concentration found high in wheat leaves. The range of maximum z score was 1.8–3 at 5252 m distance. Wheat fields located far away from the kiln about 300 m were found as a cold spot. Plant samples collected from the field (Zone B) located away from the kiln in G-16/2 had 0.001 ppm fluoride concentration, and calculated z score and p value were –1.45 and 0.15, respectively.

In Islamabad during the study year 2016, fields near to kiln (Zone A) in Tarnol, G16/2 and Jhang sayedan were found significantly as hotspot fields (Figure 1D). Similarly, in Rawalpindi Tehsil, fields nearer to the kiln located in Chakri, Dhok makhanali and Garja road were found significant hotspot area for having most fluoride contaminated wheat plants (Figure 1E). In all places, fields away from the kiln were found a cold spot area except the Dhok Makhanwali where the kiln distant field presented as mild hotspot area. In Tehsil Gujarkhan, fields located in Bara-giran and near GT road were found to be a significant hotspot, Mera and Mohra were found as mild hotspot, and Behr and Guliyana were marked as a cold spot (Figure 1F).

Fields located in Islamabad around the operational kiln at the places of Tarnol and G-16/2 were found as a hotspot for fluoride contamination (Figure 2A) in the year 2017. The range of maximum z score value was 1.35–3.25 at the threshold distance 200 m.

Figure 2B showed the fields located in the Chakri Rawalpindi as hotspot with maximum z score value ranging from 1.5–3.59 at the distance of 30 m during the year 2017. In the same year, Mandra, Mohra, GT road, Bara Giran (Figure 2C) were found as a hotspot area with the z-score ranging 0.6–3.6 at a distance of 395 m. While Jhangi Sayedan, Kangota, Mera and Guliyana were found as a cold spot for fluoride contaminated wheat plants. In the year 2015, maize fields in the northeast of Mohra Gar, in the south east at Khaba Barala, Ropar Kalan, in Northwest at Dhok Makhanwali were found as a hot spot and fields in Mohra Faqira, Banda Nagial and Dokh Karam Baksh were found as a cold spot in Tehsil Rawalpindi (Figure 3A). In Tehsil Gujarkhan fields closer to kilns located in Northwest in Sakrila and in Kuri Kas (North) were found hotspot fields for having more fluoride contamination in maize plants (Figure 3B). While, fields in south at Bhagwal and in west at Bher Kalan were statistically cold spot fields.

In Tehsil Rawalpindi during the year 2016, fields located in Dhok Makhanwali, Dhok Niazu, Mohra Gar, and Chak Jalal-ud-din were the hotspot fields and Mohra Faqirah marked as cold spot (Figure 3C). Whereas in Tehsil Gujarkhan, fields in Gurha and Sakrila were noticeable hotspot and fields in the north and south were cold spot for fluoride contamination (Figure 3D).

In the year 2017, hotspot fields having most fluoride contaminated maize plants were found in Dhok Makanwali and Dhok Niazu in Rawalpindi Tehsil (Figure 3E), and in Sakrila and Kuri Kas in Tehsil Gujarkhan (Figure 3F). While statistically significant clustering of low concentration of fluoride in plants were marked in Dhok Maira in Rawalpindi and Bhagwal in Gujarkhan. Fields in Chak Jalal-ud-din, Malikpur, Mohra Gar and Ropar Kalan were found as mild hotspot fields.

Zone of indifference as the conceptualization of spatial relationship and Euclidean distance as distance method were applied for Gi z-score and p-value calculation. Resulted field values were calculated z score at the threshold distance 200, 250, and 800 for the year 2015, 2016, and 2017, respectively in winter season. Five classes were delineated using the natural break classification method.

Dhok Gujran fields in Rawalpindi Tehsil and Mohra Harian field in Gujarkhan Tehsil were the hotspot fields having a significantly high concentration of fluoride in the soil during the year 2015 (Figure 4A). The maximum z score value indicated by hotspots was ranging between 6 to 8.5 in the study year 2015 at the threshold distance 200 m. Fields located in Mangal (Kallar Syedan) Bher Kalan (Gujar Khan), Ropar Kalan and Lar (Rawalpindi) were the cold spots which indicated significantly less fluoride concentration in soil. The minimum concentration indicated by cold spots was in negative values. Chakri and Chak Jalal-ud-din were found less significant area for fluoride contamination in soil.

In the year 2016, fields located in Islamabad (I-16/2, Jhangi Syedan), Rawalpindi (Dhok Gujran, Chakri and Chak Jala-ud-din), Bara Giran Gujarkhan were the hotspot fields for fluoride contaminated soil (Figure 4B). The range of maximum z score value indicated by hotspot was 2 to 6.3 in the study year 2016 at the threshold distance 250 m. Bher kalan, Rohra in Gujarkhan, Dhok Gujran and Ari-sayedaan in Islamabad, Pindora in Kallar Syedan were the less significant fluoride contaminated hotspot fields whereas no significant cold spot was found, but less significant hotspot fields were identified in Lar (Pindi), Mangal (Kallarsayedaan), and Ratial (Gujarkhan).

Fields located in I-16/2, Jhangi Sayedan, Dhok Gujran, Chakri, Bara Giran, Mohra Harian were the significant hotspot fields for fluoride contamination in soil during year 2017. The range of maximum z score was 1.05–6.235 at the 800 m threshold distance. While fields in Mohra Sarandaz (Gujarkhan), Mangal, Pindora (Kallar Syedan), Chak Jalal-ud-din (Pindi), Dhok Gujran and Ari-Sayedaan were the less significant hot spots fields (Figure 4C). Only Sahot's field in Kallar Syedan was identified as a significant cold spot field.

Geographical spatial patterns are indispensable to monitor and understand the geographical or environmental phenomena, as well as to appropriate decision making. Spatial pattern could be clustered or can be randomly dispersed. But when a cluster pattern of data is observed, it suggests that there is a cause that is behind this cluster, about which one can be interested in it. Results of positive values obtained from Moran I suggested significant clustering of data. This indicated that there is a reason or source for this significant spatial clustering. In the present study, the gradient spatial cluster pattern showed high positive autocorrelation in short threshold distance of samples. Plant leaves samples of Tehsil Rawalpindi showed

high positive Index values at 50 m and 30 m distance in all study years. Chu et al. (2011) specified that the high and positive Moran I value depicted the similar adjacent values and low Moran I value indicated when dissimilar values are surrounded by each other.<sup>12</sup> This proved the clustering pattern of similar values of samples collected from surrounding of kilns. Analyzing spatial gradient clustering is necessary for making predictions by developing spatial models and also requires for understanding the variation in spatial pattern depicted by data set.<sup>13,14</sup>

Once it found that the data of analyzed sample is significantly clustered, hotspot analysis was applied to the data set to discover the locations where the spatial clusters were located. The Moran Index model (MIM) proved an important model to provide deep insight into the spatial clustering of fluoride contaminated plant and soil sample data. But Global Moran Index Model did not present hotspots of pollution, which furthermore was identified by Getis-Ord Geo-spatial technique. The spatial autocorrelation analysis (MIM) of the present study suggested the significant clustering of soil and plant samples throughout the study period (2015, 2016, and 2017) in Tehsil Rawalpindi and Gujar Khan, also in Islamabad just of winter data set. Though no significant clustering was recorded in the data of Kallar Syedan but the results were still effective for the identification of and decision about the polluted areas.<sup>15</sup>

Similar to the study conducted on soil fluoride, results presented significant hotspot and cold spots of fluoride pollution in kiln surrounded fields by indicating the higher and lower z score and lesser p-value.<sup>16</sup> These higher and lower values showed the spatial clustering of recorded values, whereas p-value marked the significance of relationship at a different threshold distance.

## CONCLUSIONS

The hotspot based maps revealed the site of pollution and risk in the fields located around the coal-fired kiln and nearby places. It was found that more significant clustering of fluoride pollution hotspot was found in the summer season and extent of pollution varied according to the climate and study years.

It is recommended that government should formulate policy either to discourage the construction of kilns in nearby cultivating areas or work on or improving the kilns and fuel quality. It is also recommended that governmental agencies should have a regular check and balance upon the emissions from kilns and enforce all devised permissible standards on the kiln industry for the protection, conservation, and improvement of environment by prevention and control of air pollution. These measures are necessary because crop damages by fluoride emission from kilns leads to economic loss.

## CONFLICT OF INTERESTS

The authors declare that they have no conflict of interest.

## REFERENCES

- [1] Wahid A. Influence of atmospheric pollutants on agriculture in developing countries: A case study with three new wheat varieties in Pakistan. *Science of the Total Environment* 2006;37:304-13.
- [2] Emberson LD, Ashmore MR, Murray F, Kuylenstierna JCI, Percy KE, Izuta T, Zheng Y, Shimizu H, Sheu BH, Liu CP, Agrawal M, Wahid A, Latif NM, Van TM, De-Bauer LI, Domingos M. Impacts of air pollutants on vegetation in developing countries. *Water, Air and Soil Pollution* 2001;130:107-18.
- [3] World Health Organization (WHO). Air Pollution.[cited 2018 Apr 2]. Available from: <http://www.who.int/airpollution/en>
- [4] Khan A, Khan S, Khan MA, Qamar Z, Waqas M. The uptake and bioaccumulation of heavy metals by food plants, Their effects on plants nutrients, and associated health risk: A review. *Environmental Science and Pollution Research* 2015;22:13772-9.
- [5] Jochner S, Markevych I, Beck I, Hoffmann C, Heinrich J, Menzel A. The effects of short- and long-term air pollutants on plant phenology and leaf characteristics. *Environmental Pollution* 2015;206:382-9.
- [6] ICIMOD. Brick sector in Pakistan. [cited 2019 Apr 7]. Available from: <https://www.ccacoalition.org/sites/default/files/resources/Fact%20sheet%20brick%20sector%20Pakistan.pdf>.
- [7] Ahmad MN, Berg LJ, Shah HU, Masood T, Buker P, Emberson L, Ashmore M. Hydrogen fluoride damage to vegetation from peri-urban brick kilns in Asia: A growing but unrecognized problem?. *Environmental Pollution* 2012;162:319-24.
- [8] Munawer ME. Human health and environmental impacts of coal combustion and post-combustion wastes. *Journal of Sustainable Mining* 2017;17: 87-96.
- [9] SAARC. Evaluating energy conservation potential of brick kilns in SAARC countries. [cited 2015 Sept 3]. Available from: <http://www.saarcenergy.org/portals/1/Pakistan%20Report%20on%20Brick.pdf>.
- [10] Feser E, Sweeney S, Renski H. A descriptive analysis of discrete U.S. industrial complexes. *Journal of Regional Science* 2005;45:395-419.
- [11] Getis A, Ord JK. The analysis of spatial association by use of distance statistics. *Geographical Analysis* 1992;24:189-206
- [12] Chu HJ, Lin YP, Chang TK. Spatial autocorrelation analysis of soil pollution data in Central Taiwan. *International Conference on Computational Science and Its Applications*. 2011. Santander, Spain: IEEE.
- [13] Jacquez GM. Geographic boundary analysis in spatial and spatio-temporal epidemiology: Perspective and prospects. *Spatial and Spatio-temporal Epidemiology* 2010;1:207-18.
- [14] Fallahzadeh RA, Ghadirian D. Spatial Distribution, Health risk assessment and survey of fluoride pollution source with GIS in drinking water: A case study, Abarkouh, Iran. *Journal of Environmental Health and Sustainable Development* 2018;3:496-503.
- [15] Fan C, Myint S. A comparison of spatial autocorrelation indices and landscape metrics in measuring urban landscape fragmentation. *Landscape and Urban Planning* 2014;121:117-28.
- [16] Xie Z, Li J, Wu W. Application of GIS and geostatistics to characterize spatial variation of soil fluoride on Hang-Jia-Hu Plain, China. *Computer and Computing Technologies in Agriculture* 2007;1:253-66.