Spatial distribution patterns of phosphorus in top-soils of Greater London Authority area and their natural and anthropogenic factors

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Abstract

Soil phosphorus (P) has a strong impact on soil and water quality. Soils in urban areas tend to be enriched with P, however, they have not been adequately investigated. A total of 6467 top-soil samples collected and analysed by the British Geological Survey provided basic data for studying the top-soil P distribution patterns and their environmental implications. The hotspots and cool spots were identified using the index of local Moran's I, which is a powerful methodology for discerning spatial clusters and spatial outliers. Combined with the results of one-way analysis of variances (ANOVA), a strong geogenic control of P was illustrated with elevated concentrations in areas of alluvium and river terrace deposits. P concentration in the lower Thames Estuary was clearly influenced by the tidal effect which has diluted the P-rich sediments. The high concentrations of SiO_2 and low pH level were linked to the low value clusters of P in Hyde Park and Richmond Park. Besides the natural control, the high value clusters concentrated in the city centre and built-up area, which indicated soil P content was strongly affected by human activities. The results of a *t*-test also showed the significant distinction of mean P concentrations between urban area and non-urbanised area, implying that urbanization and built-up materials accounted mostly for the locations and magnitude of the P pool. To conclude, the spatial patterns of P observed in the study area were controlled by both natural (parent materials and geomorphology) and anthropogenic (urbanization) factors.

Key words: London; phosphorus (P); GIS; local Moran's I; urbanization; the Thames Estuary; parent materials (PMs)

1. Introduction

With the rapid urbanization in the world, urban soils are receiving considerable attention due to their associations with the life quality of human beings (Norra and Stüben, 2003). Phosphorus (P), as a limiting nutrient for organisms, is responsible for water eutrophication (Conley et al., 2009; Halecki and Gąsiorek, 2015). The P-enriched urban soils can affect groundwater quality via leaching (Zhang et al., 2001). Moreover, P derived from urban household waste can decrease the stability of organic carbon (C) in urban soils, resulting in organic C loss (Chen et al., 2014). Therefore, the locations and scale of soil P accumulation provides crucial information for P management and prevention of the leaching of P to water bodies.

Previous researches have revealed the P enrichment in urban soils, including Nanjing (Zhang et al., 2001), Hangzhou (Zhang, 2004), Beijing (Xia et al., 2013), Nanchang (Chen et al., 2014) of China, Bangkok (Faerge et al., 2001) of Thailand, Gälve (Nilsson, 1995) of Sweden and Phoenix (Metson et al., 2012) of the USA. There are a number of factors influencing P accumulation and sequestration in urban areas, including parent materials (PMs), hydrology, biotic processes, and current and historical land use and management (Bennett, 2003; Bennett et al., 2004). These factors can be classified into two categories: the geogenic (internal) factors and the anthropogenic (external) factors. The geogenic source of P comes from PM which plays a role in driving differences in P status in long-timescale (Mage and Porder, 2013). Appleton et al. (2013) reported that some elements in the London region were governed by a mixture of geogenic and anthropogenic factors, for example, PM controls 12-16% of the variance of P. The external sources of P are more complicated, such as, fertilizer application in for agricultural purpose in fields and green spaces, food and human waste, building materials like asphalt, wood and cement (Metson et al., 2012), among which sewage treatment works and septic tanks are two main inputs of P to urban soils (Neal et al., 2005). The phenomenon of P accumulation in surface soils implies that human activities can modify P concentration to some extent. Interestingly, P has long been used to indicate human activities in the field of archaeology during pre-agricultural and agricultural ages (Holliday and Gartner, 2007), demonstrating the recognition of human influences on soil P.

The study area, London, is the capital of the United Kingdom with a two-thousand-year history as a major settlement. The population was 8.2 million based on the 2011 Census with an increasing trend of growth, particularly in suburban areas, where the land use and soil quality could be changed dramatically. The local government continues to make efforts to upgrade sewage (including sludge) treatment capacity and develop the Thames Tideway Sewer Tunnels in order to address the issues of sewer overflows and improve the water quality (Great London Authority, 2016). Consequently, to reduce economic cost and increase the efficiency of controlling P pollution, the the urban soils with elevated P which pose a a potentially high risk should be targeted critically and the input sources should be assessed (Huang et al., 2012).

When considering of a mixture of geogenic and anthropogenic controls, geographical information system (GIS) and spatial analyses, for instance, inverse distance weighted (IDW) interpolation and local Moran's I, are powerful tools to explore P spatial distribution and allow the identification identify of high and low concentration areas (hotspots and cool spots). The local Moran's I, a Local Indicators of Spatial Association (or Autocorrelation) (LISA) method, has been widely used for extraction of spatial patterns (Bone et al., 2013; Voutchkova et al., 2014; Majumdar and Biswas, 2016), while in soil pollution researches, hotspots often represent pollution spots in comparison to the values of their neighbours (Zhang et al., 2008).

In this study, the hotspot analysis is applied to reveal the pools of P accumulation and the objectives were: i) to reveal the spatial patterns of P in top-soils of the Greater London Authority (GLA) area; ii) to identify the locations of P of accumulation and depletion; and iii) to explore the geogenic and anthropogenic factors causing the spatial patterns.

2. Materials and methods

2.1 Soil parent material

A surface PM map (Fig.1) with simplified geological types (Miles and Appleton, 2005) was adopted for the study. Cretaceous and Palaeogene bedrocks cover the most of the GLA area, meanwhile some parts of the area are underlain by substantial Quaternary superficial deposits (British Geological Survey, 2011). The white chalk subgroup, formed in the Cretaceous, is the oldest bedrock within the GLA area, while the Thanet sand formation is the oldest Palaeogene deposit. The Lambeth group, Thames group, and Bracklesham group belong to Palaeogene deposits, cropping out extensively, among which Thames group is the most widespread. The Quaternary deposits include clay-with-flints, till, river terrace deposits, alluvium, and Head. The clay-with-flints formed from the original Palaeogene cover, and earlier Quaternary deposits are heterogeneous in texture, colour, and clast content. Till is deposited by the Anglian ice sheet which comprises mostly of stiff pebbles or boulders and silty clay. River terrace deposits have been formed by the diversion of the River Thames, and the gravel deposition crops out mainly on hilltops. Alluvial deposits that primarily form a flat surface in valleys of the rivers Thames and Lee are most recent deposits within the research area. Generally, the alluvium consists principally of silty clay and fine- to coarse-grained sands.

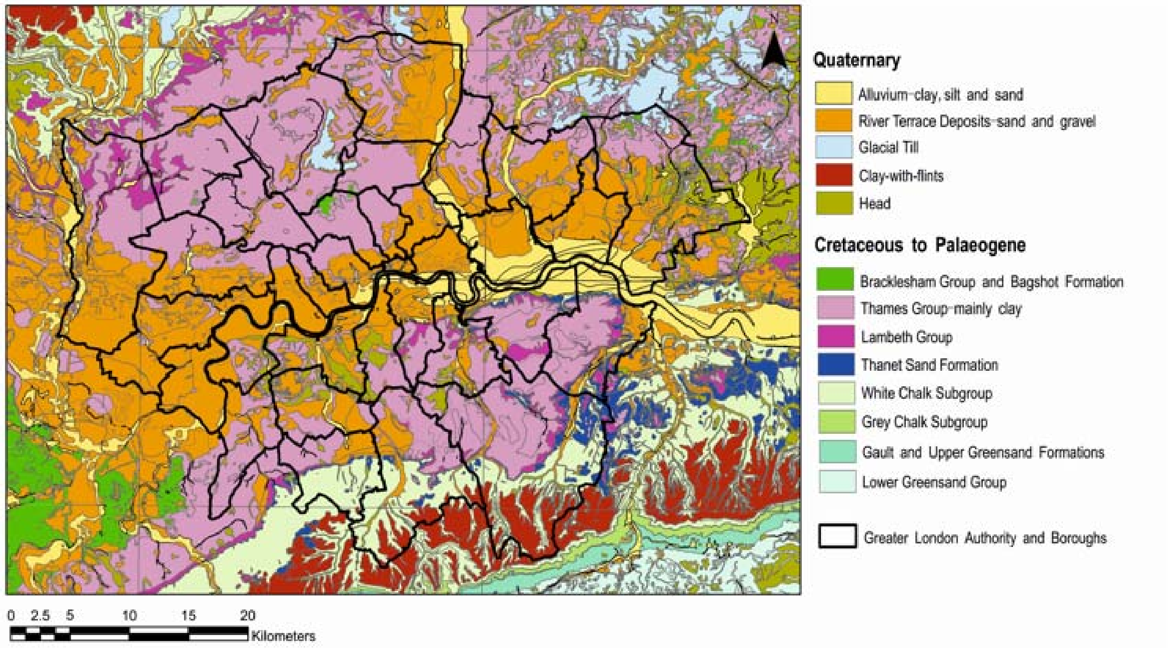


Fig. 1. Superficial and bedrock geology map of the London region.

2.2 Soil chemistry data

In GLA area, a hand-held auger was used to collect 6467 top-soil samples by British Geology Survey. The sampling density was 4 samples per square kilometre and the sampling depth was ca. 5-20 cm. At each site, composite samples based on 5 sub-samples were taken at the centre and four corners of a 20 metre square. Forty-eight trace and major chemical elements were measured by X-ray fluorescence spectrometry (XRFS) after soil samples were dried and sieved to < 2 mm. Besides, loss on ignition (LOI at 450 ℃) and pH were also determined. Details of sample preparation, analytical methods, and quality control procedures were described in Allen et al. (2011) and Johnson (2011).

2.3 Statistical and geostatistical analyses

The one-way analysis of variance (ANOVA) was applied to determine the variation between PM groups. Normal probability plot suggested that the data distribution needed to be normalized prior to ANOVA analysis. Therefore, Box-Cox transformation was performed to improve the normality to meet the statistical assumption. Subsequently, Tukey's multiple comparisons method was employed to further observe differences among distinct PM groups.

Geostatistical analysis and GIS mapping techniques were applied to present spatial distribution maps and to identify the hotspots, in order to explore the probable sources of P. The relatively simple interpolation method of inverse distance weighted (IDW) is sufficient to identify the overall pollution pattern,. IDW is based on the assumption that each existing point datum has a local impact that decreases with distance. There are two main parameters for an IDW. The first one is the power value, which decides how rapidly the influence of the point falls off with distance . The higher the power is, the more influence the nearest neighbour has to the prediction location. The second one is the number of neighbours to be included which is to some extent arbitrary (Zhang et al., 2011).

The P concentration hotspots can be recognized using the local Moran's I index (Anselin, 1995; Getis and Ord, 1996):

$$I_i = \frac{z_i - \overline{z}}{\delta^2} \sum_{j=1, j \neq i}^{n} \left[w_{ij}(z_j - \overline{z}) \right]$$

where z_i is the value of the variable z at location i; $\overline{z}$ is the mean value of z of the n samples; z_j is the value of the variable at all the other locations (where $j \neq i$); σ^2 is the variance of variable z; and w_{ij} is a weight which can be defined in different ways, such as a fixed distance band (Zhang et al., 2008). The weight w_{ij} can also be generated according to the specific spatial relationships among all the features in a dataset.

Spatial clusters, including high-high clusters and low-low clusters, are indicated by high positive local Moran’s I values, which have similarly high or low values as their neighbours. Spatial outliers, containing high-low clusters and low-high clusters, are identified by high negative local Moran’s I values, which behave differently from their neighbours (Lalor and Zhang, 2001). The significance level of Local Moran’s I index can be assessed on an assumption of a normal distribution due to standardization (Anselin, 1995). In this study, the significance level was set at 0.05 and the resulted map was based on 999 permutations to avoid sensitivity on the specific randomization. More detailed explanations of local Moran’s I are available in Anselin (1995) and Zhang et al. (2008).

2.4 Data analyses and computer software

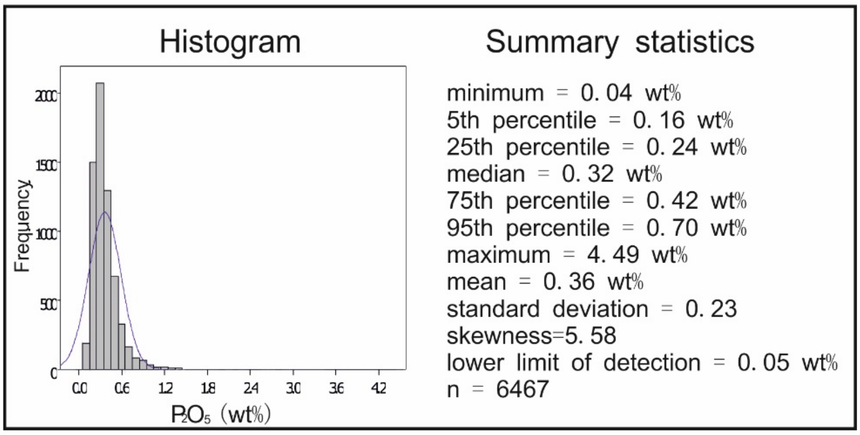
Raw data were stored in a file of MS Excel®. Basic statistical parameters were obtained and the test for normality was carried out using SPSS® ver. 23. The calculation of Local Moran’s I index was performed in the software GeoDa (version 1.6.7 by Luc Anselin, 2015). All maps were produced using ArcGIS (version 10.3) software.

3 Results and discussion

3.1 Geogenic factors

3.1.1 Influences of parent materials

Summary statistics (Fig. 2) demonstrate that P_2O_5 concentrations had a strong positively skewed distribution. The average value of P_2O_5 concentration in the surface soils in GLA area was 0.36 wt%, which was slightly higher than the average value of 0.3 wt% in the earth’s crust (Rawlins et al., 2012).



Summary statistics

minimum = 0. 04 wt%
5th percentile = 0. 16 wt%
25th percentile = 0. 24 wt%
median = 0. 32 wt%
75th percentile = 0. 42 wt%
95th percentile = 0. 70 wt%
maximum = 4. 49 wt%
mean = 0. 36 wt%
standard deviation = 0. 23
skewness=5. 58
lower limit of detection = 0. 05 wt%
n = 6467

Fig. 2. Summary statistics of P for top-soil samples from the London urban area.

The superficial and bedrock geology map overlaid by the hotspots (Fig. 3(a)) shows that the majority of high-high clusters overlap two typical PMs, alluvium and river terrace deposit, from central-west and central-north to city centre. The elevated P concentrations are associated with alluvium and river terrace deposit due to the impact of hydrology, erosion processes and transformations of various forms of P (Hunter and Walton, 2008; Neal et al., 2005). Most alluvium is carried and deposited during floods when erosion is most active and the carrying capacity is at a maximum (Turowski, 2013). Alluvial deposits which are the most recent deposits within the GLA area form an approximately flat surface in valley floors and occur mainly in the valleys of the River Thames and River Lee. On average, the alluvium consists largely of clayey silt and silty clay clayey silt with subsidiary sands.. Within the GLA area, river terrace gravels were deposited throughout the Thames valley, extending north into the Lee valley (British Geological Survey, 2011). Fluvial sediments are recognized as being extremely crucial in controlling P flux in water-sediment system (Haggard et al., 2007). Cooper et al. (1995) demonstrated that riparian soils tended to enrich a great quantity of readily-transportable forms of P. The riparian soils perform transient storage of P between the aquifer and the stream, which makes alluvial floodplains as well as some groundwater mirror stream concentrations by having total P concentrations (Heeren et al., 2011). The close association between hydrology and P was also implied by water area percentages map (Fig. 3(b)) based on land-use data (Gov.uk, 2005). The land-use statistics (Generalised Land Use Database) 2005 (Enhanced Basemap) comprises areas of domestic buildings, non-domestic buildings, gardens, green space, road, rail, paths, and water. Besides, studies have shown that the retention capacity of stream sediments is in direct proportion to the P concentrations in streams (Forelich, 1998; Bridgham et al., 2001). Sediments surrounding areas of water have a controlling function of retention and release of P between adjacent aquatic systems and uplands (Thompson and McFarland, 2010; Xia et al., 2011).

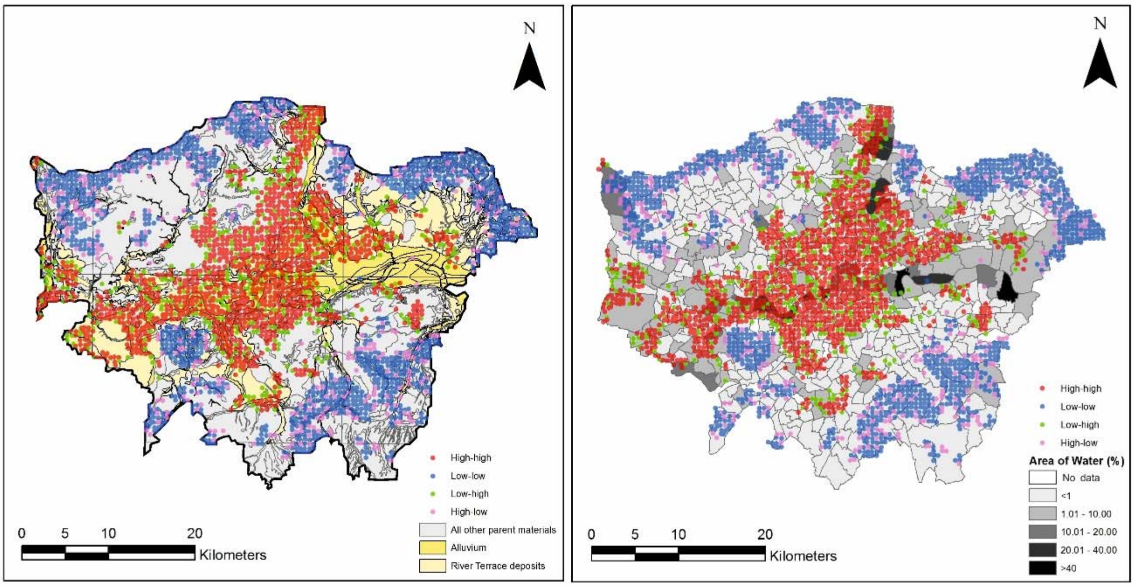


Fig. 3. (a) Spatial distribution map of significant hotspots and cool spots overlaying on geology map; (b) Spatial distribution map of significant hotspots and cool spots overlaying on water area percentages map.

To further explore the association with fluvial sediment, one-way ANOVA was applied to examine whether there were significant differences among PM groups. Box-Cox transformations were carried out after adding 1.00 to the raw data to bring the minimum value above 1.00 with the optimal $\lambda = -3.8$, which produced a more normal distribution than the Box-Cox transformation of the raw data with the optimal $\lambda = -0.25$ directly (Osborne, 2002; Osborne, 2010), though neither transformed data passed the Kolmogorov–Smirnov test (K–S test). Statistical tests are more efficient for relatively small sample sizes, which tends to reject of most statistical hypotheses for the large sample size (>1000) (Zhang et al., 2005). The result of one-way ANOVA showed a significant difference among PMs ($p < 0.001$). Given that Tukey HSD is helpful to determine specifically which PM groups are different from each other, the results reveal that alluvium and river terrace deposit have higher concentrations of P than other types of PM groups (Fig. 4). Based on the results, the variability of P concentrations in top-soils of GLA area is obviously linked to the different PM groups.

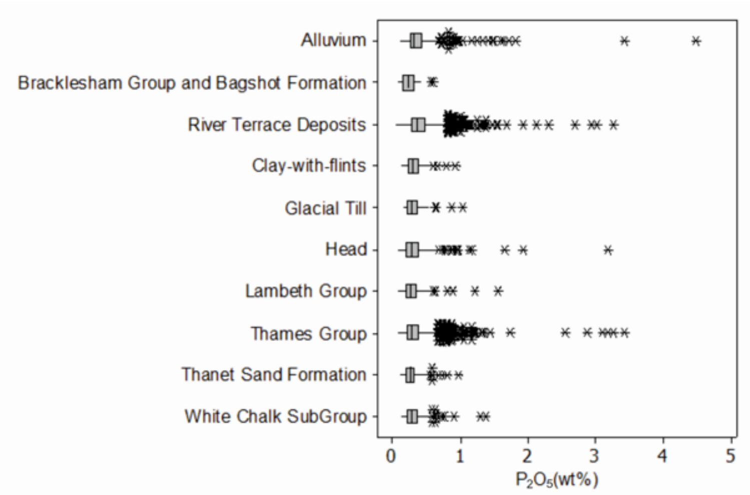


Fig. 4 Boxplot of P in top-soils of different PM groups from London urban area (n = 6467; box = interquartile range).

3.1.2 Influence of the Thames Estuary

Due to the close association between P concentrations with fluvial deposits, hotspots existed along the River Thames, the River Lee and Grand Union Canal, except for downstream below Tower Bridge of the Thames (Fig. 5) where high-high spatial clusters were expected. This “special” pattern could be related to the Thames estuary, a typical macro tidal funnel-shaped estuary, where river flow, tides, and storm surges come into interaction (Uncles and Mitchell, 2011). The River Thames is divided into three zones. Firstly, from Teddington to near Tower Bridge, the suspended particulate matter (SPM) concentrations are low and mainly land-derived. In this area the tidal effect is minimal and the bank sediment is mostly made up of the clay fraction. Downstream from the Tower Bridge tides are more influential and the sediment is much coarser than that of the upstream. Secondly, from Woolwich reach to Gravesend reach, high SPM and a high rate of deposition exists. Lastly, below Woolwich reach and down to Southend, the sedimentation is gradually predominated by a marine-derived bedload (Mikhailov and Mikhailova, 2012). The Tanshui Estuary, situated on the outskirts of the largest city of Taiwan, Taipei, showed a similar deposition pattern of P. The majority of total P derived from the upstream as well as city sewages deposited in the upper estuary and the rest was exported to the coast (Fang, 2000). Together with tides, drying and rewetting events leading P loss, the sediments of lower estuary and bay exhibit lower quantities of organic matter and P (Berbel et al., 2015; Gao et al., 2016).

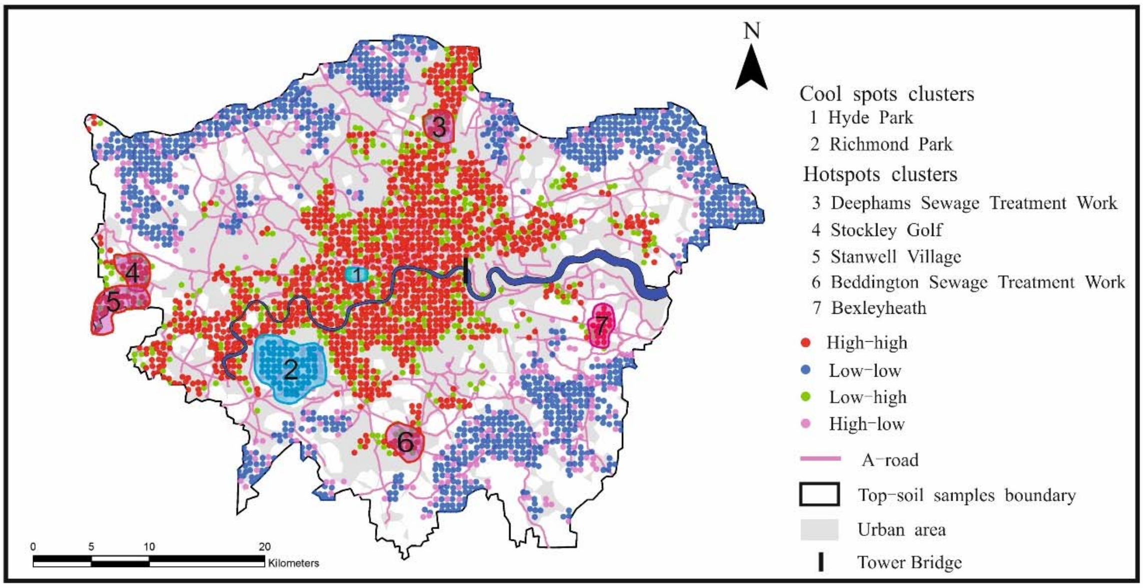


Fig. 5 Map of significant hotspots and cool spots of P overlaid onto a London street map.

3.1.3 Influences of silicon (Si) and pH

IDW is a quick deterministic interpolator without any assumptions which can be easily applied to produce an interpolated map. In this study, the power value was set as 1 in order to reduce the "bulls-eye" effect around data locations; the number of neighbours was set as 16. The distribution maps for Si and pH values were shown in Fig.6. London city centre had a high concentration of P, except for Hyde Park area (Fig. 5). Hyde Park, along with Kensington Gardens, Green Park, and St. James's Park form an almost continuous "green lung" in the heart of London. The most likely reason behind the relatively low concentrations in these parks was the less human activities in this area. Richmond Park was revealed as a significant cool spot cluster in the map which is the largest royal park in London with nationally and internationally importance for wildlife conservation. Richmond Park is an upland where the human disturbance is rare and the concentration of SiO_2 is high in sandy soils (Fig. 6(a)). The Si content is expressed as the percentage of SiO_2 and the SiO_2 content rises with the increase of sandstone in sandy soils. In contrast, the SiO_2 percentage decreases by dilution with organic matter and $CaCO_3$ (Bear, 1964). The SiO_2 distribution map shows that Richmond Park had high Si concentration (Fig. 6(a)) which indirectly meant low organic matter content. Given that nearly half the soil P occurs in combination with organic matter in surface soils, the P of this area was largely mineral or inorganic combination forms. As weathering proceeded in soils, the phosphates became increasingly bonded to aluminium (Al) and iron (Fe) rather than silicate minerals and are released from the sandy soils (Bear, 1964). Consequently, the soil containing the high concentration of Si was characteristic with low concentrations of P and other heavy metal elements. Moreover, the low pH of Richmond Park (Fig. 6(b)) is indicative of low inorganic P which occurs mainly as calcium phosphate in high pH alkaline and calcareous soils. With the development of acidity in soils, calcium phosphate is dissolved and precipitated as Al and Fe phosphate in leaching channels (Halvorson, 2016). In summary, high Si concentration and low level of pH synergistically foster the cool spots of P.

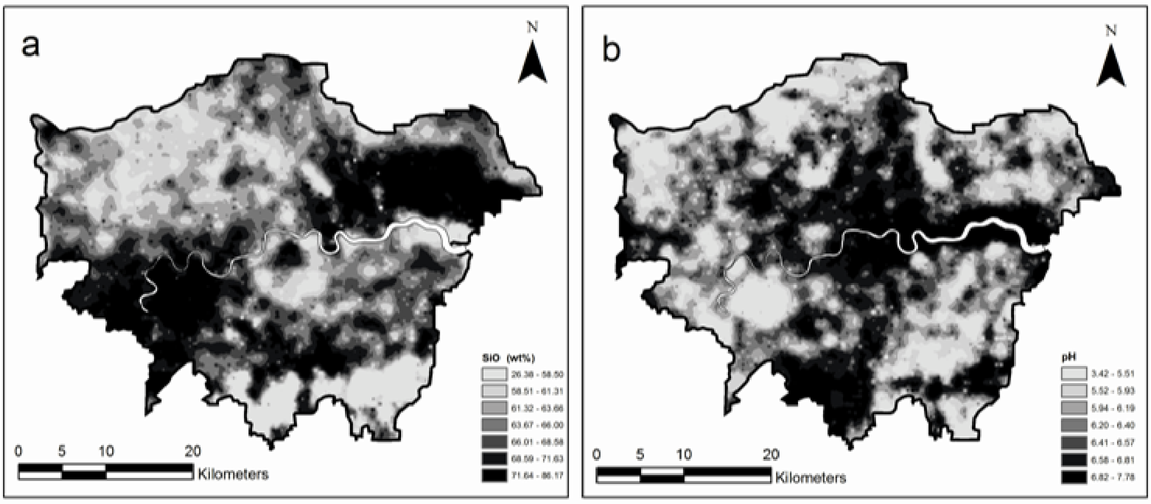


Fig. 6 (a) Spatial distribution map of SiO_2; (b) Spatial distribution map of pH.

3.2 Anthropogenic factors

3.2.1 Influences of urbanization

The P distribution surface of the London urban area was created using IDW with raw data (Fig. 7(a)), displaying a pattern of high P values concentrated in the city centre along with low values in suburban areas (Fig. 5). To find out the significance of urbanization, the data were divided into two sectors: the urban area and non-urbanised area. The urban area map was generated by building a shapefile of the GLA area downloaded from Geofabrik (2016) using the built-up tool in ArcGIS. The sample sizes were 2683 and 2784, with the mean values were 0.41wt% and 0.31 wt% in the urban area and non-urbanised area, respectively. The independent *t*-test was used to compare the difference in P concentrations between urban area and non-urbanised area after Box-Cox transformation and the results are given in Table 1. Results from Levene's test suggested that the Box-Cox transformed data set met with the equal variances assumption ($p > 0.05$). The t-value of 25.258 was large, with a significant difference between urban area and non-urbanised area ($p < 0.001$). The distribution pattern corresponded to the population density of London (Fig. 7(b)) which was produced according to London Census 2011 data (London Datastore, 2011). Moreover, the median P_2O_5 concentration of the London urban area was notably higher than that of rural areas in England and Wales (Rawlins et al., 2012), and slightly higher than that of some relatively small cities, such as Doncaster, Scunthorpe, Mansfield, and Sheffield (Table 2), which suggests that the larger more highly populated city and higher degree of urbanization tended to accumulate higher concentration of P_2O_5.

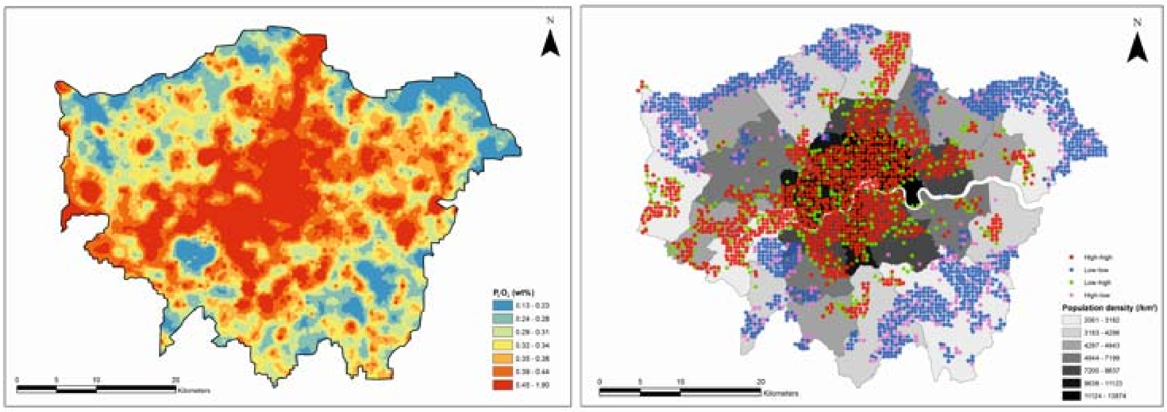


Fig. 7 (a) IDW spatial interpolation map of P_2O_5; (b) Spatial distribution map of significant hotspots and cool spots overlay on the London population density map.

Table 1 Results of Levene's test and *t*-test of P between urban area and non-urbanised area.

	Levene's test for equality of variance		***t*-test for equality of means**	
	F	Significance	*t*	Significance (two-tailed)
Equal variances assumed	2.474	0.116	25.258	0.000
Equal variances not assumed			25.138	0.000

Table 2. Comparison of median P_2O_5 concentrations in London urban top-soils with other cities and rural areas in England and Wale.

	London	Doncaster	Lincoln	Mansfield	Scunthorpe	Sheffield	Rural areas in England and Wale
Median of P_2O_5 (wt%)	0.32	0.26	0.32	0.27	0.26	0.31	0.19
Sample size	6467	279	216	257	196	575	5691
Reference		O' Donnell (2005a)	O' Donnell (2005b)	Freestone et al. (2004a)	O' Donnell (2005c)	Freestone et al. (2004b)	Rawlins et al. (2012)

There are a few aspects related to urbanization affecting the P behaviour in urban soils. As human waste and food count for the major parts of imported P in urban ecosystems, population density shapes the concentration pattern of P (Brett et al., 2005; Steinke et al. 2013). In addition, built-environment materials, such as asphalt, cement, and wood are contributing to the elevated P (e.g. the urbanized-area pattern is visible in Fig.5). Different forms of P released from a variety of sources could be mobilised into streamwater by urban stormwater (Brezonik and Stadelmann, 2002), causing eutrophication of aquatic ecosystems (Yuan et al., 2007). As a result, the most urbanized area, city centre, with highest population density made urban ecosystems P-rich environments.

3.2.2 Influences of sewage treatment works (STWs)

Besides the large high-high value cluster concentrated within the city centre, several small high-high value clusters were scattered in the outskirts, illustrating the spatial heterogeneity of urban soils (Fig. 5). The hotspot analysis showed that areas around two sewage treatment works (STWs), Beddington and Deephams, had high P concentrations. In the UK, the major concern over P pollution is associated with sewage effluent sources from urban and industrial sources as well as farming (Vaze and Chiew, 2004; Neal et al., 2010b). Considerable money has been spent on dealing with P removal from final effluents at sewage works (Neal et al., 2010a). In 2013, the City of London wastewater treatment plants removed 91% of the P, however, flows exceed the capacity of the treatment plants during rain events (City of London Environment and Engineering Services, 2014). Additionally, the majority of removed P becomes sludge. Although 75% of treated sludge is transferred to agricultural land (Shepherd et al., 2016) in England and Wales, the sludge residue remains as secondary pollution source of P which needs to be monitored. Moreover, there is a trunk sewer near Deephams sewage treatment work (Fig. 5) which may be another source of P input. Another finding is Stanwell Village (Fig. 5), located southwest of Heathrow Airport, where the high P concentration is possibly sewage related .

3.2.3 Influences of fertilizer application

Bexleyheath and Stockley Golf Course showed high levels of P (Fig. 5). Bexleyheath Golf Course is situated in the lowland, whilst Stockley Park Golf Course covers a much larger area, extending over 6,625 yards. Fertilizers are intensively added in golf courses (Shuman et al., 2000). P generally improves the growth of ornamental plants and turf grass (McDowell et al., 2001) and is essential to the start-up or green-up phase (King and Balogh, 2011). Two sports fields of Townley Grammar School, close to Bexleyheath Golf Course, might also contribute to the high-high cluster of P in Bexleyheath area. Application of high concentrations of P fertiliser can lead to losses via sub-surface drains such as creeks and ponds in the golf course, thus degrading water quality (Shuman, 2003). Low dose applications, organic formulation, and reduced rate of fertilizer are recommended to improve P fertility management (King et al., 2012).

4 Conclusions

This study has demonstrated that the variation of P in top-soils of an urban area was influenced by a combination of geogenic and anthropogenic controls. Geogenic factors, consisting of PM, the hydrology in the Thames Estuary, the distribution of Si and pH level in top-soils, were clearly associated with the P distribution. In addition, anthropogenic factors, such as population density, STWs and fertilizer application, also had an effect on the P concentration and spatial distribion, causing localised high concentrations. The combination of these effects and their interactions makes up the unique P distribution patterns in the GLA area and the identified P hotspots. The spatial distribution patterns provide important information for soil management.

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