Influence of the number of inhabitants, surface and population density on soil contamination with As, Pb, Hg, Zn, Cd and Cu in urban recreational areas of Romania

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Abstract. The surface of a locality, number of inhabitants and density are socio-demographic descriptive indicators of territorial and administrative divisions frequently related to urban soils contamination in environmental scientific research. The present study aims at assessing the connection between these indicators and pollution with various chemical elements in Romanian urban topsoils. In order to do so data for 19 cities has been analysed in correlation with As, Pb, Hg, Zn, Cd and Cu mean concentrations investigated in recreational areas. The obtained results highlighted that there is limited connection between the number of inhabitants or surface of the cities and the obtained concentrations. Therefore, when discussing cities in Romania one cannot assert that the biggest and most populated ones are implicitly more polluted. However the population density correlates, in a negative statistically significant manner with the analysed concentrations, indicating pollution problems related to historical contamination in large surface formerly industrialised cities.

Key Words: recreational urban areas, soil contamination, inhabitants, surface, population density.

Introduction. The surface of a locality, the number of inhabitants and density are socio-demographic descriptive elements used to characterise a territorial and administrative division. Specialised studies focusing on the assessment of metal contamination of urban soils always refer to these three elements when characterising a studied area (Aelion et al 2009; Hursthouse et al 2004; Chen et al 1997; Tume et al 2007; Zhang et al 2011; Figueirredo et al 2011; Hruba et al 2012). High population density is known to lead to an increasing level of urban environmental pollution (Shi et al 2011). Large, densely populated cities are investigated in order to assess soil contamination and the most common conclusion is that soil quality is deteriorating and contaminants such as metals often exceed the approved limit (Chen et al 2005; Miguel et al 2007; Shi et al 2011).

In Romania, urban settlements are diverse, both in respect of size and development. This study aims to examine the extent to which there is a connection between the number of inhabitants, surface or population density and the metal contamination of recreational areas. This will allow us to formulate documented conclusions in respect to the evolution of the characteristics of localities from a socio-demographic point of view and soil pollution aspects.

After determining the connection between the number of inhabitants, surface and density and pollution with various metals, one can establish the extent to which these elements can be considered pollution predictors.

Material and Method

Description of the studied areas. This study is carried out in 19 cities in Romania (Figure 1). These were selected because they provide heterogeneous samples in respect of the number of inhabitants, surface and density. Using the data taken from the database of the Romanian National Institute of Statistics, the number of inhabitants of the cities included in the study varies considerably, from 9,734 inhabitants in Huedin and up to 1,191,352 inhabitants in the country’s capital city, Bucharest (https://statistici.insse.ro).

The surfaces occupied by the analysed cities, (Figure 1), are also extremely diverse, and have a significant influence on the values calculated for population density.
significantly influence the values calculate for population density.

The data regarding the number of inhabitants, surface and density in correlation with the identified contaminate concentrations identified was processed in the statistical program SPSS 16 for Windows. Density was obtained by dividing the number of a city’s inhabitants to its surface.

**Sample treatment and analytical procedures.** In order to identify As, Cu, Cd, Zn, Pb and Hg concentrations 264 soil samples were collected from 88 different recreational areas and analysed. Samples of about 300 g were collected from the soil surface, within the first 15 cm, dried for 24 hours at ambient temperature and for 1 hour in a drying chamber at 110 degrees. Rough pieces of rock and vegetal metal were removed and the samples were shredded to reduce agglomeration. During this phase the concentration of volatile elements such as Hg might have been significantly diminished. In order to obtain homogeneous and representative split of contaminates (Kulmatiski & Beard 2004), the soil sample were downsized by repeated quartering (Miguel et al 2007) for subsequent analysis. For analytical analysis samples were acid digested at ambient temperature using aqua regia (3 parts HCl: 1 part HNO3) in sterile laboratory vessel. Concentrations for As, Pb, Hg, Zn, Cd and Cu were determinate in solutions using atomic absorption spectrometry. For this type of analysis an accurate Solaar S4 Atomic Absorption Spectrometer was utilized. The mean value of each contaminant analysed/city was calculated.

**Statistical analysis.** Using SPSS 16 program for Windows the data was analysed, in order to establish a possible a connection between the number of inhabitants, surface, density and the mean concentrations calculated for the analysed metals. Data distribution coefficients were calculated to determine the appropriate type of correlation coefficient for this determination. A commonly encountered situation in environmental statistical analysis was established, namely a distribution which differs from the normal one (Pallant 2001). Therefore, a non-parametric Spearman correlation coefficient was chosen for the statistical analysis (Reimann & Fizmoser 2000).
Results and Discussion. The variables considered in this study are diverse and exhibit a number of characteristics. The concentrations of the metals identified, using atomic absorptive spectrometry, in the soil of recreational areas, parks and playgrounds, differ significantly from one city to another. The mean concentrations for each city are listed in Table 1.

Mean concentrations for each city (mg kg⁻¹) and Romanian guidelines for normal values expressed in Nr. 756 Governments Order from 3rd November 1997 for approving regulations for environmental pollution

<table>
<thead>
<tr>
<th>City</th>
<th>As</th>
<th>Cu</th>
<th>Cd</th>
<th>Zn</th>
<th>Pb</th>
<th>Hg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cluj-Napoca</td>
<td>19,74</td>
<td>60,08</td>
<td>0,41</td>
<td>153,33</td>
<td>52,81</td>
<td>0,82</td>
</tr>
<tr>
<td>Turda</td>
<td>28,09</td>
<td>73,93</td>
<td>0,47</td>
<td>281,00</td>
<td>98,23</td>
<td>1,33</td>
</tr>
<tr>
<td>Câmpia - Turzii</td>
<td>21,64</td>
<td>45,05</td>
<td>0,44</td>
<td>172,05</td>
<td>69,05</td>
<td>1,15</td>
</tr>
<tr>
<td>Dej</td>
<td>23,82</td>
<td>28,30</td>
<td>0,50</td>
<td>143,69</td>
<td>59,96</td>
<td>0,29</td>
</tr>
<tr>
<td>Gherla</td>
<td>19,50</td>
<td>23,99</td>
<td>0,43</td>
<td>80,99</td>
<td>28,58</td>
<td>0,39</td>
</tr>
<tr>
<td>Huedin</td>
<td>21,66</td>
<td>26,14</td>
<td>0,47</td>
<td>112,34</td>
<td>44,27</td>
<td>0,34</td>
</tr>
<tr>
<td>Tg. Mureș</td>
<td>11,88</td>
<td>26,36</td>
<td>0,19</td>
<td>83,80</td>
<td>25,80</td>
<td>0,30</td>
</tr>
<tr>
<td>Baia Mare</td>
<td>47,68</td>
<td>96,30</td>
<td>1,35</td>
<td>255,62</td>
<td>287,33</td>
<td>1,63</td>
</tr>
<tr>
<td>Arad</td>
<td>28,83</td>
<td>37,85</td>
<td>0,28</td>
<td>157,19</td>
<td>35,37</td>
<td>0,34</td>
</tr>
<tr>
<td>Timișoara</td>
<td>24,52</td>
<td>44,40</td>
<td>0,24</td>
<td>117,33</td>
<td>41,23</td>
<td>0,52</td>
</tr>
<tr>
<td>Oradea</td>
<td>25,41</td>
<td>37,51</td>
<td>0,23</td>
<td>115,81</td>
<td>34,61</td>
<td>0,41</td>
</tr>
<tr>
<td>Craiova</td>
<td>18,86</td>
<td>15,19</td>
<td>0,17</td>
<td>53,15</td>
<td>46,56</td>
<td>0,33</td>
</tr>
<tr>
<td>Slatina</td>
<td>25,27</td>
<td>37,30</td>
<td>0,31</td>
<td>117,73</td>
<td>41,40</td>
<td>0,49</td>
</tr>
<tr>
<td>Vaslui</td>
<td>32,53</td>
<td>55,83</td>
<td>0,75</td>
<td>170,00</td>
<td>130,24</td>
<td>0,83</td>
</tr>
<tr>
<td>Botoșani</td>
<td>18,27</td>
<td>19,06</td>
<td>0,23</td>
<td>63,85</td>
<td>29,46</td>
<td>0,28</td>
</tr>
<tr>
<td>Bacău</td>
<td>23,53</td>
<td>32,06</td>
<td>0,28</td>
<td>122,44</td>
<td>44,57</td>
<td>0,45</td>
</tr>
<tr>
<td>Brăila</td>
<td>21,75</td>
<td>32,35</td>
<td>0,35</td>
<td>145,55</td>
<td>49,86</td>
<td>0,55</td>
</tr>
<tr>
<td>Buzău</td>
<td>19,36</td>
<td>27,00</td>
<td>0,24</td>
<td>89,55</td>
<td>36,38</td>
<td>0,36</td>
</tr>
<tr>
<td>Bucharest</td>
<td>18,16</td>
<td>28,17</td>
<td>0,58</td>
<td>130,00</td>
<td>57,54</td>
<td>0,66</td>
</tr>
<tr>
<td>Romanian guidelines for normal values of contaminants in soil</td>
<td>5</td>
<td>20</td>
<td>1</td>
<td>100</td>
<td>20</td>
<td>0.1</td>
</tr>
</tbody>
</table>

As indicated in Table 1, the highest mean values for As, Cu, Cd, Pb and Hg concentrations were recorded in Baia Mare and for Zn in Turda. The lowest mean values of contaminant concentrations were recorded in Craiova for Cu, Cd and Zn, in Târgu Mureș for As and Pb and in Botoșani for Hg. Nevertheless, as shown in table 1, with an exception for Cd contaminant concentrations identified frequently exceed values regulated through Romanian legislation.

Surface, number of inhabitants and density are descriptive elements for the analysed cities, which also differ considerably (Table 2).
Table 2

<table>
<thead>
<tr>
<th>City</th>
<th>Surface (km²)</th>
<th>Number of inhabitants</th>
<th>Density (loc/km²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cluj-Napoca</td>
<td>179.5</td>
<td>301,913</td>
<td>1,808</td>
</tr>
<tr>
<td>Turda</td>
<td>91.43</td>
<td>56,701</td>
<td>630</td>
</tr>
<tr>
<td>Cluj-Turzii</td>
<td>23.78</td>
<td>261,94</td>
<td>1,100</td>
</tr>
<tr>
<td>Dej</td>
<td>109.12</td>
<td>38,065</td>
<td>290</td>
</tr>
<tr>
<td>Gherla</td>
<td>36.30</td>
<td>22,080</td>
<td>560</td>
</tr>
<tr>
<td>Huedin</td>
<td>61.00</td>
<td>9,734</td>
<td>150</td>
</tr>
<tr>
<td>Tg. Mureș</td>
<td>49.30</td>
<td>143,221</td>
<td>2,593</td>
</tr>
<tr>
<td>Baia Mare</td>
<td>235.73</td>
<td>137,455</td>
<td>520</td>
</tr>
<tr>
<td>Arad</td>
<td>46.18</td>
<td>164,208</td>
<td>3,500</td>
</tr>
<tr>
<td>Timișoara</td>
<td>130.50</td>
<td>307,551</td>
<td>2,447</td>
</tr>
<tr>
<td>Oradea</td>
<td>156.20</td>
<td>204,358</td>
<td>1,257</td>
</tr>
<tr>
<td>Craiova</td>
<td>81.40</td>
<td>297,510</td>
<td>2,994</td>
</tr>
<tr>
<td>Slătina</td>
<td>65.25</td>
<td>76,726</td>
<td>1,175</td>
</tr>
<tr>
<td>Vâlcea</td>
<td>68.44</td>
<td>66,992</td>
<td>1,027</td>
</tr>
<tr>
<td>Botoșani</td>
<td>40.70</td>
<td>114,799</td>
<td>2,625</td>
</tr>
<tr>
<td>Bacău</td>
<td>41.30</td>
<td>174,192</td>
<td>3,494</td>
</tr>
<tr>
<td>Brăila</td>
<td>77.90</td>
<td>208,464</td>
<td>2,314</td>
</tr>
<tr>
<td>Buzău</td>
<td>81.30</td>
<td>130,320</td>
<td>1,333</td>
</tr>
<tr>
<td>Bucharest</td>
<td>228.00</td>
<td>1,919,352</td>
<td>8,518</td>
</tr>
</tbody>
</table>

As presented in Table 2, the analysed city occupying the largest surface is Baia Mare, followed by the capital city Bucharest and Cluj-Napoca. The cities occupying the smallest surface are Gherla, Botoșani and Bacău. The largest number of inhabitants was recorded in Bucharest, followed by Timișoara and Cluj and the smallest in Huedin, Gherla and Câmpia Turzii. The highest population density was recorded in the capital city Bucharest, followed by Arad and Bacău and the lowest in Dej, Baia Mare and Gherla.

By analysing the resulting correlation coefficients (Figure 2) one can see that there is no connection between the number of inhabitants and the concentrations of the analysed metals. Therefore, in respect of the cities analysed in Romania one cannot conclude that the most populated cities are implicitly the most polluted. Also, the surface of the cities does not correlate with metal concentrations either and thus the size of a city can also not be associated with contamination.

However, the analysis carried out suggests a particularly interesting aspect. Population density, defined as the number of inhabitants reported to km² correlates, not very negatively, but statistically significantly, with all the analysed metals. As, Cu, Cd, Zn, Pb and Hg concentrations are inversely proportional to population density. Thus, with respect to analysed samples, the cities with low population densities seem to be more contaminated than those with higher densities. Such examples from the study carried out are Baia Mare, Turda or Câmpia Turzii. In these cities the mining and industrial activities were intense, but considerably reduced in recent years. As a result, soil contamination with metals is a normal consequence (Allgaier 1997; Bell & Donnelly 2006; Fodor & Baican 2001), but one must bear in mind the fact that these cities are historically contaminated (Levei et al 2009). Furthermore, once the mines or factories were closed, the population started to migrate to other urban settlements where it could find jobs or it returned to the rural areas (Rieniets 2009; Antonescu & Popa 2012; Popescu 2014). Thus, cities occupying a relatively large surface, due to vast industrial areas, were left...
with a relatively low number of inhabitants and considerable pollution-related problems.

<table>
<thead>
<tr>
<th>Correlations</th>
<th>N</th>
<th>DimensLoc</th>
<th>DensitatePop</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spearman’s rho</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Correlation Coefficient</td>
<td>N</td>
<td>.030</td>
<td>-.121</td>
</tr>
<tr>
<td>Sig. (2-tailed)</td>
<td>1.000</td>
<td>.614**</td>
<td>.614**</td>
</tr>
<tr>
<td></td>
<td>.</td>
<td>.</td>
<td>.</td>
</tr>
<tr>
<td>N</td>
<td>98</td>
<td>98</td>
<td>98</td>
</tr>
<tr>
<td>DimensLoc</td>
<td>Correlation Coefficient</td>
<td>N</td>
<td>.030</td>
</tr>
<tr>
<td>Sig. (2-tailed)</td>
<td>1.000</td>
<td>.614**</td>
<td>.614**</td>
</tr>
<tr>
<td></td>
<td>.</td>
<td>.</td>
<td>.</td>
</tr>
<tr>
<td>N</td>
<td>98</td>
<td>98</td>
<td>98</td>
</tr>
<tr>
<td>DensitatePop</td>
<td>Correlation Coefficient</td>
<td>N</td>
<td>.030</td>
</tr>
<tr>
<td>Sig. (2-tailed)</td>
<td>1.000</td>
<td>.614**</td>
<td>.614**</td>
</tr>
<tr>
<td></td>
<td>.</td>
<td>.</td>
<td>.</td>
</tr>
<tr>
<td>N</td>
<td>98</td>
<td>98</td>
<td>98</td>
</tr>
</tbody>
</table>

** Correlation is significant at the 0.01 level (2-tailed).
* Correlation is significant at the 0.05 level (2-tailed).

Figure 2. Spearman correlation coefficients between metal concentrations and number of inhabitants, surface and population density in the analysed cities – results generated by SPSS).

Due to the fact that this study analyses only the situation of recreational areas, the connection between low population density and high contaminant concentrations can also be explained from the point of view of maintenance work carried out in the parks and playgrounds within the cities. Thus, one can assume that in cities with a higher population density, such as Bucharest or Arad, where recreational areas are more frequented, these are also better maintained than the recreational areas in former industrial cities.

**Conclusions.** This study investigated whether the urban soil contamination increases with surface and population density. The analysis was performed by calculating the coefficients of statistical correlation between surface, number of inhabitants and population density in 19 cities in Romania and average concentrations of the contaminants (As, Cu, Cd, Zn, Pb and Hg) present in the soils of certain recreational areas from the analysed cities.

The obtained results highlight two major aspects. First of all, due to the fact that the correlation coefficients between soil contamination with metals and the number of inhabitants and surface of a city were not identified as statistically significant, one may conclude that there is no connection between the number of inhabitants of a city and the analysed metal concentrations. Therefore, when discussing cities in Romania one cannot assert that the most populated ones are implicitly more polluted. Therefore, when discussing cities in Romania one cannot assert that the most populated ones are implicitly more polluted. The surface of the cities does also not correlate with metal concentrations and thus a city’s size can also not be associated with contamination. As a result, in Romania the number of inhabitants or surface of a city cannot be considered a relevant predictor for soil pollution in the urban environment. Since the existence of a cause-effect relationship could not be proven, one cannot assert that the inhabitants of larger cities are more exposed to contact with soil contaminants than the inhabitants of smaller cities. A more careful monitoring of soil pollution in the recreational areas of these cities is necessary only if there is another type...
of substantiated suspicion referring to a possible contamination. Second of all, one can see that population density correlates, in a negative statistically significant manner, but not very strongly with contaminant concentrations. Thus one may assert that the attention of those responsible for the environment should be oriented towards the cities with lower population densities, mainly former industrial cities, characterised by large surfaces and historical contamination.

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