Long-term land application of biosolids—a case study
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ABSTRACT
Impact of long-term land application of biosolids on groundwater and soil quality of an application site, which had been operated for 8–15 years, was evaluated in this study. During and after the biosolids application, biosolids-amended soil, groundwater, and background soil samples were collected mainly for pathogen, nitrogen, phosphorus, and heavy metal analyses. Soil test data showed that there was no heavy metal accumulation in the biosolids-amended soil even after 10 years of biosolids application. Similar results were also observed from the groundwater samples in which the heavy metal concentrations in all groundwater samples were well below the maximum contamination levels of the drinking water standards. In addition, bacteriological levels of the soil and groundwater samples were close to the background level and below the permissible limits, respectively, thereby showing no pathogen contamination. However, nitrate-nitrogen contamination of the groundwater was occasionally observed probably due to an excess loading of the biosolids in the past. This problem can be alleviated by applying biosolids at agronomic rates so that no excess nitrogen is available for leaching down to the groundwater.

Key words | biosolids, groundwater, heavy metals, land application, nitrate-nitrogen, soil

INTRODUCTION
In recent years, more and more sewage sludge (or biosolids) produced from wastewater treatment plants is beneficially used instead of landfilling, ocean dumping or incineration (Stukenberg et al. 1993). Among various beneficial uses such as daily landfill cover and final cover, land application is the most promising and prevailed option. In 1998, about 41% of the sewage sludge produced was applied to the land (USEPA 1999). It is expected that this proportion will increase to 48% by 2010. By definition, land application involves the spreading of biosolids or the sewage sludge further treated by aerobic/anaerobic digestion, composting or alkaline stabilization on the soil surface or injecting biosolids into the soil (Epstein 2003). Although the biosolids can condition and fertilize the soil, and supplement or even replace commercial fertilizers, excessive application can cause possible surface water and groundwater contaminations by nitrogen, phosphorus and organic materials, accumulation of heavy metals, trace organic chemicals and pathogenic microorganisms in soil, and bio-augmentation of toxic elements in the food-chain (Jacobs 1981).

For protecting public health and the environment, USEPA promulgated regulations “40 CFR Part 503” which establish standards for the final uses and disposal of sewage sludge, thereby preventing any reasonably anticipated
adverse effects of certain pollutants present in the biosolids (USEPA 1993). For instance, heavy metal concentrations in the biosolids and the cumulative loading rate of heavy metals to the site during the land application are regulated.

In this paper, the impact of long-term land application of the biosolids on the groundwater and soil qualities, and other human health and environmental concerns were evaluated. To achieve these goals, groundwater and soil samples were intermittently collected from a successfully operated long-term biosolids application site for evaluating the temporal variation of their physical, chemical and biological characteristics, and for comparing with standards.

**MATERIAL AND METHODS**

A biosolids application site, which covers 55.9 hectares (ha) and had been operated for 8–15 years, was chosen for the long-term impact study (Figure 1). Groundwater, background soil and the biosolids-amended soil samples were collected for pH, conductivity, total solids, fecal coliform (F.C.), fecal streptococci (F.S.), nitrate-nitrogen (NO₃-N), ammonia-nitrogen (NH₄-N), total Kjeldahl nitrogen (TKN), total phosphorous (PO₄³⁻), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) analyses. Biosolids characteristics and application rate were also monitored simultaneously during the land application.

The wastewater treatment plant where biosolids were land applied, consisted of secondary treatment system with two activated sludge aeration basins followed by secondary clarifiers. The primary sludge was thickened in a gravity sludge thickener; while the waste activated sludge was thickened by centrifugation. After mixing the thickened primary and secondary sludge together, the mixtures were introduced into a two-stage anaerobic digester system.

![Map showing the sludge injection site, and location of the wastewater treatment plant, soil (S1, S2, S3 and S4) and groundwater samplings (W1, W2, W3 and W4).](https://iwaponline.com/wst/article-pdf/57/3/345/438576/345.pdf)
containing three primary digesters and one secondary digester. This digestion process aimed to reduce pathogen levels, odor and volatile organic content in the mixed sludge. The digested biosolids were stored in a storage lagoon before injecting to a field nearby and off-site to a privately owned farmland. The on-site injection program was started in 1984 and the average on-site biosolids application rate was 14.9 dry tons/ha yr.

For groundwater monitoring, groundwater samples were collected quarterly from the four observation wells (i.e., W1, W2, W3 and W4) located at the injection site from 1987 to 1993. The biosolids-amended soil samples were

<table>
<thead>
<tr>
<th>Year/Parameters</th>
<th>Application rate (dry tons/ha)</th>
<th>TKN (kg/ha)</th>
<th>NH₃-N (kg/ha)</th>
<th>Org-N (kg/ha)</th>
<th>NO₃⁻ – N (kg/ha)</th>
<th>PAN (kg/ha)</th>
<th>Cd (kg/ha)</th>
<th>Cr (kg/ha)</th>
<th>Cu (kg/ha)</th>
<th>Pb (kg/ha)</th>
<th>Ni (kg/ha)</th>
<th>Zn (kg/ha)</th>
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</thead>
<tbody>
<tr>
<td>1984</td>
<td>12.26</td>
<td>634</td>
<td>323</td>
<td>310</td>
<td>0.83</td>
<td>386</td>
<td>0.100</td>
<td>0.95</td>
<td>13.41</td>
<td>1.80</td>
<td>1.10</td>
<td>14.44</td>
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<td>1985</td>
<td>30.39</td>
<td>1985</td>
<td>937</td>
<td>1048</td>
<td>1.80</td>
<td>1149</td>
<td>0.140</td>
<td>1.63</td>
<td>21.36</td>
<td>3.97</td>
<td>1.62</td>
<td>25.20</td>
</tr>
<tr>
<td>1986</td>
<td>13.29</td>
<td>734</td>
<td>544</td>
<td>190</td>
<td>5.30</td>
<td>588</td>
<td>0.100</td>
<td>0.55</td>
<td>12.66</td>
<td>3.74</td>
<td>0.98</td>
<td>18.09</td>
</tr>
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<td>1987</td>
<td>17.30</td>
<td>1339</td>
<td>836</td>
<td>563</td>
<td>0.97</td>
<td>950</td>
<td>0.086</td>
<td>1.22</td>
<td>15.25</td>
<td>3.28</td>
<td>1.14</td>
<td>19.63</td>
</tr>
<tr>
<td>1988</td>
<td>21.84</td>
<td>1658</td>
<td>560</td>
<td>1098</td>
<td>1.15</td>
<td>781</td>
<td>0.083</td>
<td>0.72</td>
<td>14.50</td>
<td>2.68</td>
<td>0.75</td>
<td>18.58</td>
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<tr>
<td>1989</td>
<td>8.18</td>
<td>389</td>
<td>171</td>
<td>218</td>
<td>0.45</td>
<td>216</td>
<td>0.047</td>
<td>0.56</td>
<td>6.12</td>
<td>1.59</td>
<td>0.46</td>
<td>9.48</td>
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<tr>
<td>1990</td>
<td>17.30</td>
<td>876</td>
<td>367</td>
<td>509</td>
<td>1.37</td>
<td>470</td>
<td>0.105</td>
<td>1.24</td>
<td>12.54</td>
<td>2.89</td>
<td>1.03</td>
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<tr>
<td>1991</td>
<td>11.86</td>
<td>668</td>
<td>295</td>
<td>373</td>
<td>1.01</td>
<td>371</td>
<td>0.063</td>
<td>0.67</td>
<td>6.57</td>
<td>1.74</td>
<td>0.85</td>
<td>12.51</td>
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<tr>
<td>1992</td>
<td>11.37</td>
<td>535</td>
<td>210</td>
<td>325</td>
<td>0.33</td>
<td>276</td>
<td>0.063</td>
<td>0.72</td>
<td>8.09</td>
<td>2.03</td>
<td>0.58</td>
<td>15.04</td>
</tr>
<tr>
<td>1993</td>
<td>5.19</td>
<td>213</td>
<td>93</td>
<td>120</td>
<td>0.13</td>
<td>117</td>
<td>0.025</td>
<td>0.29</td>
<td>3.34</td>
<td>0.69</td>
<td>0.24</td>
<td>5.90</td>
</tr>
<tr>
<td>Total</td>
<td>149.0</td>
<td>9031</td>
<td>4336</td>
<td>4754</td>
<td>13.34</td>
<td>5303</td>
<td>0.812</td>
<td>8.55</td>
<td>113.9</td>
<td>24.4</td>
<td>8.75</td>
<td>159.88</td>
</tr>
</tbody>
</table>

* CPLRs-Cumulative pollutant loading rates from USEPA (1995).
collected in 1992 and 1993 from the locations (i.e., S1, S2, S3 and S4) within the site where the biosolids had been applied recently. For comparison purposes and getting more information, personnel from the University of Missouri (MU) also collected the sludge, biosolids-amended soil samples from the surface layers (0–5 cm) and deep layers (20–25 cm) within the site, and background soil samples at nearby field where no biosolids had been applied. They also collected groundwater samples simultaneously from the monitoring wells. The characteristics of the groundwater, soil and sludge samples were analyzed following the procedures listed in APHA (1998), and USEPA sludge sampling and analysis guidance document (USEPA 1989).

RESULTS AND DISCUSSION

Characteristics of the biosolids

As provided by the wastewater treatment plant, the annual average characteristics of the biosolids are shown in Table 1. The biosolids applied to the site possessed approximately 2.4–5.5% of total solids, 28–440 mg of NO$_3^-$ – N and 9600–23000 mg of PO$_4^{3-}$ – P per kg of the dry biosolids, and various amounts of heavy metals. Except for As, there were only minor variations in the biosolids characteristics (~14–36% of relative standard deviation, RSD) from 1984 to 1993. It is interesting to note that there were noticeable decreasing trends of Cu and Ni concentrations in the biosolids most likely due to improvement in industrial pre-treatment facilities, which reduced the amount of Cu and Ni discharged to the wastewater treatment plant. Although the biosolids contained various types of the heavy metals, their concentrations were below the pollutant concentration limits (PCLs) established in 40 CFR Part 503 except the Pb level in 1986.

Pollutant loading rates

Amount of pollutants in the biosolids applied to a unit area of the site were calculated by multiplying the biosolids application rates by the pollutant concentrations in the biosolids (Table 1). As shown in Table 2, approximately 4300 kg of NH$_3$–N, 4700 kg of organic-nitrogen (Org-N) and 13 kg of NO$_3^-$ – N had been...
applied per hectare of the site from 1984 to 1993, which corresponded to about 5300 kg/ha of the cumulative plant available nitrogen (PAN) loading rate (MDNR 1993). Annual loading rate of PAN continuously decreased from 1990 to 1993 mainly due to the drop in the biosolids application rates and reduction in the PAN concentrations in the biosolids in that period of time. Besides, it is important to note that the cumulative heavy metal loading rates, after 10 years of the biosolids application, were far less than USEPA limits. Thus there seems to be of no immediate concern about heavy metal build-up in the biosolids-amended soils.

Groundwater quality

Variation of the groundwater quality in W1 from 1987 to 1993 is shown in Table 3. It can be readily observed that most the groundwater samples from W1 showed non-detectable levels of heavy metals. Although the samples collected in 1987, 1988, 1989 and 1993 showed detectable levels of Pb, Ni and/or Zn, their concentrations were still less than the maximum contamination levels (MCLs) under the USEPA Safe Drinking Water Act Standards (USEPA 2003). Unlike heavy metals, NO$_3^-$ – N concentration exceeded the MCL 10 mg/L twice in 1991 and 1993. NO$_3^-$ – N level in W2 also exceeded the MCL once in 1993 but those in W3 and W4 were never higher than the MCL (data not shown). It is not clear why NO$_3^-$ – N contamination of the groundwater was only observed in W1 and W2 but was not observed in W3 and W4. Besides, it is believed that the NO$_3^-$ – N contamination in W1 had no direct correlation with the season. In 1991, the contamination occurred in September and November which were relatively low rainfall months. On the other hand, in 1993, it occurred in February and May when the groundwater table was generally high.

To understand the possible reason leading to the unacceptable high NO$_3^-$ – N concentration in W1, the NO$_3^-$ – N concentration was compared with the amount of excess PAN in the biosolids-amended soils (Figure 2). The excess PAN was determined by considering the annual PAN loading rate from the biosolids, the PAN originally existing in the native soil (36 kg/ha yr) (Blanchar et al. 1985), and the PAN annually consumed by growing corn (258 kg/ha) and wheat (100 kg/ha) (USEPA 1983). As seen, the NO$_3^-$ – N concentration profile between 1990 and 1993 was similar to the profile of the excessive PAN between 1984 and 1988 with a lag of 5 to 6 years. The two maxima in both excess
PAN and NO$_3^- - N$ concentration profiles were also separated by a year. The similarity of the profiles indicated that the unacceptable high NO$_3^- - N$ concentration in W1 in 1991 and 1993 was most likely attributed to the slow leaching of the excess PAN in the soils, which resulted from the excess biosolids application from 1984 to 1988.

Quality of the biosolids-amended soils

Characteristics of the biosolids-amended soils in 1992 and 1993 are summarized in Table 4. The TKN and PO$_4^{3-} - P$ concentration in 1993 were relatively lower than those in 1992; whereas the soil heavy metal concentrations in 1993 were slightly higher for some heavy metals. Besides, it is important to observe that the heavy metal concentrations in all the soil samples were far less than the Dutch soil quality standards (The New Dutchlist 2001), thereby indicating no heavy metal contamination of the soil even after 10 years of biosolids application.

Biosolids, groundwater and soil studies

Biosolids, groundwater, and surface and deep soil samples collected by the MU personnel showed the same order of magnitude of the heavy metal concentrations as those reported in Table 1, Tables 3 and 4, respectively, except that the Ni and Zn levels in the soil were higher. In most cases, the deep soil collected in 1993 had lower heavy metal concentrations than the surface soil (Figure 3a). This data was in good agreement with the findings of Dowdy et al. (1991). In fact, many studies indicated that the major portion of heavy metals in treated sludge after land application still remained in the sludge-soil zone and only small portion was taken up by plants and leached into deep soil layer (Boswell 1975; Chang et al. 1982; Dowdy et al. 1991; Yangming & Corey 1993). The heavy metal concentrations in the soil were only somewhat higher than the background concentrations except Cu and Pb levels in 1993 but were still lower than the Dutch soil quality standards. Besides, as illustrated in Figure 3, the NO$_3^- - N$ concentrations in the soil were moderate and only slightly higher than the background levels. The bacteriological soil data also indicated that the levels of F.C. and F.S. were close to background level with no evidence of contamination.
The groundwater quality in W1 and W4 monitored by the MU in February 1994 showed NO$_3^-$ levels in the range of 1.9–5.2 mg/L which was lower than the MCL 10 mg/L. No detectable levels of heavy metals were measured except As which was still below the MCL 10 µg/L. The bacteriological quality of the groundwater showed F.C. and F.S. levels less than 20 most probable number (MPN) per 100 mL, which was an acceptable groundwater quality. On the other hand, the bacteriological quality of the biosolids (F.C. = 48600 colonies/g and F.S. = 41300 colonies/g) placed it in USEPA Class B category so that human and animal contact with the biosolids should be minimized until environmental factors have reduced pathogens to very low levels (USEPA 1999).

**SUMMARY AND CONCLUSIONS**

In case heavy metals were the only pollutants being concerned, there were no contaminations of the groundwater and biosolids-amended soil within the application site even after 10 years of the biosolids application. This is because the heavy metal concentration in all the groundwater, and surface and deep soil samples did not exceed the MCLs and Dutch soil quality standards, respectively. Moreover, the bacteriological levels in the groundwater and soil were well below the permissible limits or close to the background levels, thereby showing no evidence of pathogen contamination. NO$_3^-$ level in the biosolids-amended soil was only slightly higher than the background level. As a result of these observations, land application of the biosolids is believed to be a viable option for recycling/disposing sewage sludge. However, application of the biosolids at agronomic rates is needed to prevent potential groundwater contamination by the excessive nutrients loaded.

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REFERENCES