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Final Report to DES and NH State Legislature, June 1998 – November 2001 Monitoring Demonstration at a Top-Soil Manufacturing Site in New Hampshire

William H. McDowell & Tamara J. Chestnut University of New Hampshire March 15, 2002

Executive Summary

A gravel pit reclamation and top-soil manufacturing site in Hooksett, NH was studied to determine if current management practices pose a threat to groundwater quality. The site has had repeated applications of biosolids on an annual basis since 1989 with top-soil removal approximately every five years. Each year materials were typically stockpiled on-site up to 9 months prior to application. The site was instrumented with groundwater monitoring wells within the biosolids application area, in an adjacent control field, and both up- and downgradient from the biosolids treatment area. Hydrologic and chemical characteristics of the groundwater were monitored on a bi-weekly basis during the growing season (April – November) and monthly during the winter for several years (1998-2001).

Our results show that unacceptably high levels of nitrate were found in some wells within the biosolids application area, but not in others. Elevated nitrate levels in groundwater tended to be found directly beneath, and immediately downgradient from, the stockpiling areas. Low-nitrate groundwater was found under other portions of the reclamation site. These two observations suggest that repeated application of residuals does not by itself cause significant increases in groundwater nitrate levels, but that stockpiling of residuals prior to application does cause significant groundwater contamination. Further research would be necessary to verify this conclusion. With the data we have collected, we can state unequivocally that DES regulations at the time of this study were not sufficiently protective of groundwater quality to prevent nitrate contamination. We also examined concentrations of metals in groundwater at this site. We found that they were well below NHDES and EPA allowable limits at all times, and in all areas. We can thus state with equal assurance that DES regulations were sufficiently protective to prevent groundwater contamination with trace metals at this demonstration site.

Our results from this site also show that groundwater in control and upgradient areas often has surprisingly high nitrate levels. Although these values were not above NHDES or EPA allowable limits, the data do suggest that some off site contamination may be entering our site or may be present in the surrounding groundwater. This makes it more difficult to determine the effects of biosolids applications at our site, and will make regulation of other reclamation sites more difficult if our findings are typical.

Problem Statement

Beneficial re-use of residuals, such as biosolids and short paper fiber, has become an increasingly important topic in both environmental policy and science over the past decade. These materials, which are a natural by-product of waste management, are becoming a significant disposal problem. Reclamation activities, such as those at abandoned gravel pits, provide a way for these secondary products to be recycled back into the environment. However, the same attributes that make this material valuable as an organic material also may cause deleterious effects to groundwater without proper management and monitoring. Excessively high nitrogen content in biosolids may lead to nitrate concentrations in groundwater that exceed EPA allowable limits and lead to harmful environmental and human health effects.

The State of New Hampshire along with the entire New England region have been actively trying to enact policies dealing with the use of residuals specifically for reclamation activities. However, environmental policy and effective management practices are extremely difficult to develop without the aid of comprehensive scientific studies that examine the impacts of these activities on the ecosystem.

Previous Research

Gravel and sand mining reclamation and restoration have become a major environmental concern for protecting water quality, preventing erosion, and re-establishing vegetation. The use of municipal biosolids and papermill sludge has become a cost effective alternative to performing these restoration activities and disposing of waste products. As with any land application of biosolids, the environmental impacts of this activity must be monitored to ensure ecological and public safety. The use of biosolids in reclamation activity provides a necessary catalyst for rehabilitating severely disturbed lands that would be very slow to recover without intervention. These disturbed areas require both the addition of nutrient fertilizer and the development of topsoil in order to support plant growth. This is an important distinction between reclamation and the application of biosolids to natural or agricultural lands. Therefore, the positive and negative impacts of land spreading biosolids for reclaiming mined lands is of particular interest to policy makers and land managers.

Several studies have examined the effects of utilizing municipal biosolids (semi-solid, solid and composted), paper mill sludge, and a combination of the two materials on revegetating gravel and sand mining operations following soil removal operations. Catricala et al. (1996) performed a microcosm study to examine the effects of using a combination of paper mill sludge and wood ash mixed with sand to reclaim an abandoned gravel pit in Maine. Chemical constituents responded to the sludge application to varying degrees. Chloride, SO_4^2 and Na mobilized quickly in the first year, whereas NO₃ and Ca leached late in each growing season (October – November) as plant uptake and microbial immobilization slowed and N mineralization and nitrification increased. Organic matter decomposition caused the elution of dissolved organic carbon, copper, magnesium and potassium. Based on the results of this study, they suggest that a paper mill sludge with a

C:N ratio of 30:1 is optimal for minimizing water quality hazards and providing adequate plant re-growth.

Another study of reclaimed sand and gravel mines was performed in Virginia (Daniels et al. 1998) where anaerobically digested municipal biosolids mixed with sawdust (C:N ratio > 25) was applied. Corn was planted following biosolids application and increased yields were observed in treated plots. Nitrate leached from plots treated with biosolids once corn was harvested in the fall and declined slowly until spring of the following year. Plots where biosolids were mixed with sawdust had significantly lower $NO₃-N$ leaching. Groundwater was unaffected by biosolids application in terms of $NO₃-N$ contamination at this site. As with the study by Catricala et al. (1996), Daniels et al. (1998) found that increasing the C:N ratio of the biosolids to greater than 25 did not appear to have any impact on crop yields but did significantly reduce the amount of $NO₃-N$ leaching from the plots.

Composted municipal biosolids were used in the New Jersey pinelands in order to reclaim sites where sand mining and other soil removal operations were performed. Jacobsen (1998) examined a composted biosolids application site at the Lakehurst Naval Air Engineering Station in New Jersey for groundwater quality and vegetation growth effects. Groundwater chemistry was monitored for 2 years following composted biosolids application. Concentrations of various chemical contaminants including nitrate, copper, zinc, lead, ammonia, calcium, sulfate, magnesium, sodium, chloride, potassium and dissolved organic carbon were found to increase above ambient levels for the first year following application and subsequently decreased to ambient levels by the end of the second year.

Project Purpose

The purpose of this project was to document any effects of reclamation utilizing residuals regulated by Env-Ws 800 on groundwater quality. Specifically, the project assesses the impact of residual application on nitrogen concentrations (nitrate, ammonium, and dissolved organic N) in groundwater at a reclamation site in New Hampshire. The site uses biosolids and/or short paper fiber (SPF) to reclaim (revegetate) a former gravel pit and manufacture topsoil. The primary goal of the project was to demonstrate whether current management and application practices are sufficient to protect groundwater from contamination with $NO₃-N$ and other forms of dissolved nitrogen, and possibly to identify ways to improve best management practices (BMPs). A secondary goal of the project was to assess levels of trace metals and trace organic compounds in groundwater at the reclamation site.

Project Description

The site was evaluated over a multi-year period for nitrate, ammonium, dissolved organic nitrogen (DON) and dissolved organic carbon (DOC) in groundwater. At the site, a treatment plot was established that utilized residual materials including a manufactured topsoil (biosolids and SPF) and biosolids only.

Both the treatment plot and the control area were identified, and lysimeters and groundwater wells were installed in both areas. Wells were also installed to monitor groundwater quality beneath the treatment plot, and at locations presumed to be hydrologically up- and down-gradient of the treatment plot. Original stainless steel wells were insufficient for hydrologic and chemical characterization and were replaced in 1999 with 2" diameter PVC wells with 2' of slotted well screen. An additional six PVC wells were installed in May 2000 to supplement chemical and hydrologic data.

Site Description

Figure 1: Map of Martin's Ferry in Hooksett, NH. Open squares indicate groundwater wells and solid circles indicate soil lysimeters. Scale = $1" \approx 200$ ft.

The site is Martin's Ferry located in Hooksett, NH and consists of a 5-acre topsoil manufacturing operation, which has had reclamation activity with biosolids for the past ten years and an adjacent control field of approximately the same size with no history of biosolids activity. The site is currently permitted under Env-Ws 800 as a site appropriate for biosolids utilization. This site is being monitored primarily to examine the impact of long-term biosolids applications on groundwater. At the site, biosolids only have been applied approximately annually since 1989 with removal of the organic topsoil approximately every five years. An application of a biosolids and SPF mixture was applied in October of 1999 with a C:N ratio of 27:1 (Table 1). Approximately 1,000 cubic yards of topsoil was removed in 1996, returning soils to their native condition of excessively drained Windsor loamy sand.

Year	Residuals	Biosolids (yds^3)	Total Nitrogen (#/acre)
1996	Biosolids only	166	512
1997	Biosolids only	337	1040
1998	Biosolids only	460	826
1999	Biosolids & SPF	353	1600
2000	None		0
2001	None		

Table 1: Residuals application rates from 1996 to 2000 at the Martin's Ferry Site.

Hydrologic Characterization

Hydrologic characterization was done from 1999 to 2001 at Martin's Ferry in Hooksett to determine the flow path of groundwater through the plot. An additional objective was to determine if water from the Merrimack River was flowing through the subsurface and into our treatment plot, which would distort our assessment of the effects of residual applications on groundwater quality. Nine 2" diameter PVC wells were installed and surveyed over a several-month period. By April 2000, all nine wells were set to a depth sufficient to reach the water table. Six additional 2" diameter PVC wells were installed in May 2000 to aid in hydrologic studies and to characterize upgradient conditions. Our data show that the water table at Martin's Ferry is sloping steeply from the hillslope toward the river, and the path of groundwater is nearly perpendicular to the river. Our data also show that it is highly unlikely that the Merrimack River infiltrates into groundwater at this site (Figure 2).

Figure 2: Hydrologic flownet for Martin's Ferry in Hooksett.

Sample Collection

Samples of soil solution and groundwater were collected every two weeks during the growing season (August 1 - November 1) and were continued on a monthly basis through the winter (December - April). Groundwater wells were evacuated with a Teflon bailer until three volumes of water within the well had been exchanged prior to sample collection, where feasible. If recharge rates were slow, less than three volumes were withdrawn. Samples from lysimeters and groundwater wells were placed in clean HClwashed polyethylene bottles (HDPE) and remained on ice in coolers until delivery to UNH for chemical analysis. Samples were filtered with a 0.7 μ m GF/F ashed filter and frozen until analysis. Samples for trace metals analysis were filtered with a 0.45 µm Metricel[®] membrane filter and treated with 3% nitric acid for refrigerated storage until analysis.

Chemical Analysis

Samples were analyzed for $NO₃$, $NH₄$ ⁺, DON, and DOC at the analytical laboratory (Ecosystems Analysis Laboratory) of Professor William H. McDowell, University of New Hampshire. Nitrate and NH₄⁺ are analyzed using flow injection analysis colorimetry (Lachat) with cadmium reduction for $NO₃$ and the phenol hypochlorite method with sodium nitroprusside enhancement for NH_4^+ . Total dissolved nitrogen (TDN) is measured using high temperature catalytic oxidation (Shimadzu TOC 5000) with chemiluminescent nitrogen detection (Antek 720, Merriam et al. 1996); DON is then calculated as the difference between TDN and $(NO₃⁺ NH₄⁺)$. Dissolved organic carbon

is measured using a total carbon analyzer (Shimadzu TOC 5000). Trace metals are analyzed using an inductively coupled plasma emission spectrometer (ICP) at the Analytical Services Laboratory at the University of New Hampshire.

Results

Prior to 1998, and before involvement of the current research team, only $NO₃-N$ was

Figure 3: $NO₃-N$ concentrations for groundwater using PVC wells (May 1999 – January 2001). (a) Control and upgradient wells; H-10 dark blue, H-14 black, H-15 red, H-16, green, H-20 purple, H-21 light blue, H-22 yellow; (b) Biosolids wells; H-7 dark blue, H-8 & H-18 yellow, H-9 purple, H-11 black, H-12 red, H-17 green, H-19 light blue. 5/1/1999 9/1/1999 1/1/2000 5/1/2000 9/1/2000 1/1/2001 5/1/2001 9/1/2001

analyzed at this site. Starting in June 1998, we extended this data set to include NH4-N, DON and DOC in groundwater and some soil solution. Data reported here only include data collected by this research team using PVC wells, and thus span from May 1999 to January 2001, and focus on groundwater only. For more information on soil solution and data from stainless steel wells installed prior to 1999 refer to Appendix A.

Chemical Characterization

Groundwater concentrations of $NO₃-N$ for this site are significantly higher in the biosolids application area than the control area (Figure 3). Overall, we see that the biosolids treatment wells seem to track each other over time with increases in concentration in the late summer and fall. We observed a major increase in $NO₃-N$ concentrations in the fall of 1999 following the last biosolids application. This increase subsequently declines over time until the summer of 2000 where another large increase in concentration is observed. Concentrations peaked again in the late summer of 2001, but overall concentrations seem to be declining (Figure 3). However, there are significant differences between the different wells within the biosolids treatment area. Concentrations in the control and upgradient wells also show some spatial variability but do not appear to exhibit seasonal patterns. Overall concentrations of $NO₃-N$ in control and upgradient wells do not exceed EPA allowable limits of 10 mg/L and are considerably lower than those of the biosolids treatment area.

Spatial Variability

There is considerable spatial variability in $NO₃-N$ concentrations in the groundwater at this site. Spatial heterogeneity in soils and or vegetation can lead to minor variability in groundwater $NO₃-N$, but at this site there is no evidence that these factors play any role in driving the observed patterns. Similarly, preferential groundwater flowpaths may lead to modest increases in $NO₃-N$ concentrations in certain areas, but data from the well installation boring logs show that soils are extremely homogenous and preferential flowpaths are unlikely.

Spatial variability in $NO₃-N$ concentration is associated with patterns of past stockpiling (Figure 4). This suggests that stockpiling at the site may have led to N saturation of the soils in isolated areas. The highest levels of $NO₃-N$ in groundwater are located either in these areas of past stockpiling or immediately downgradient from them (Figure 4). We also find areas outside the biosolids treatment area both upgradient and in the control area $(H-20$ and H-22) that contain elevated levels of NO₃-N; however, concentrations in these wells did not exceed the EPA allowable limit of 10 mg/L. This suggests that there may be a secondary source of contaminated groundwater entering the site from the surrounding area, and contributing to the elevated groundwater levels found on-site. However, this secondary source is not sufficient to explain the exceedingly high $NO₃-N$ levels in some wells.

Figure 4: Topographic map with color-coded wells for Hooksett site. Scale: 1cm = 20m. Green shaded area = biosolids and SPF treatment area, orange shaded areas = stockpile locations, blue shaded area = Merrimack River. Red = wells > 10 mg/L, Yellow = 3 $mg/L <$ wells $<$ 10 mg/L, Blue = wells $<$ 3 mg/L. Average well concentrations generated over the following time period: H-9 (5/99 – 11/01); H-7 and H-11 (6/99 – 11/01); H-10, H-12 and H-15 (7/99 – 11/01); H-14 and H-16 (9/99 – 11/01); H-17, H-19, H-20 and H-21 $(5/00 - 11/01)$; H-18^{*} includes data from H-8 $(9/99 - 5/00)$ and H-18 $(5/00 - 11/01)$.

Not all biosolids wells had high NO_3-N levels; in fact, some wells were lower in NO_3-N concentration than control wells. This spatial variability also supports the conclusion that stockpiling, rather than the actual biosolids application, may be driving the high levels of $NO₃-N$ seen in groundwater at this site. Based on information from the site manager we have determined that the wells with elevated $NO₃-N$ concentrations were in locations where stockpiling was heaviest during the past ten years of activity at this site (Figure 5).

Figure 5: Mean NO₃-N concentration (mg/L) for individual wells grouped according to stockpile impact. Biosolids wells are H7, H9, H11, H12, H17, H-18; control wells are H14, H15, H16, H22; downgradient well is H19, and upgradient wells are H10, H20, H21.

Chemical Relationships

Relationships among various chemical constituents at this site are consistent with biosolids as a source of the elevated nitrate concentrations that we have observed.

Average values for temperature, conductivity, pH and dissolved oxygen are reported for each individual well (Table 2). Temperature was consistent between wells and was highest in the shallowest wells. Temporal variability for each well was quite low, however spatially variability within the treatment and control areas was quite high particularly for conductivity. This variability was consistent with that found for other chemical constituents, specifically $NO₃-N$. Dissolved oxygen concentrations were consistently high, suggesting an aerobic environment in both biosolids and control areas.

Table 2: Mean values for temperature, conductivity, pH and dissolved oxygen for individual wells by category (95% confidence interval reported in parentheses).

There was a statistically significant relationship between conductivity and $NO₃-N$ in groundwater samples collected at this site (Figure 6a). A similar relationship was found between pH and NO_3-N , which is expected since nitrification, a by-product of organic matter decomposition, is an acidifying reaction (Figure 6b). The acid produced by nitrification both decreases pH and increases conductivity, as H^+ ions are highly conductive.

Other constituents (NH4-N, DOC and DON) at this site between May 1999 and November 2001 exhibit some differences between treatment and control groups for groundwater. Ammonium-N concentrations showed no significant differences between control and treatment wells and were near analytical detection limits for most samples (Figure 7). DON concentrations were significantly different between treatment wells (biosolids and downgradient) and control wells (control and upgradient); however, overall concentrations of DON in groundwater were not particularly high (Figure 7). We also found a positive linear relationship between DON and NO₃-N for groundwater wells (r^2 = 0.503, $p<0.01$). This suggests that the elevated levels of NO₃-N found in groundwater at this site are related to increased organic matter content in the treatment area due to the application of biosolids. However, the relatively low overall concentrations of DON in

groundwater indicate that DON production associated with biosolids application is of relatively minor environmental concern.

Figure 6: (a) Conductivity versus $NO₃-N$ (mg/L), (b) pH versus $NO₃-N$.

Figure 7: Means for $NO₃-N$, $NH₄-N$, DOC and DON in groundwater with 95% confidence intervals shown as bars. Statistical differences between means are noted with lowercase letters (p<0.05), and were determined using One-way Analysis of Variance with 4 levels and Tukey's pairwise comparison.

Figure 8: DOC concentrations for groundwater using PVC wells (May 1999 $-$ January 2001). (a) Control and upgradient wells; H-10 dark blue, H-14 black, H-15 red, H-16 green, H-20 purple, H-21 light blue, H-22 yellow; (b) Biosolids & Downgradient wells; H-7 dark blue, H-8 & H-18 yellow, H-9 purple, H-11 black, H-12 red, H-17 green, H-19 light blue.

Similar to $NO₃-N$, DOC concentrations showed an increase in late summer 2000 in the groundwater wells, suggesting an association with biosolids application. DOC concentrations seemed to increase slightly in autumn 2001. However, overall concentrations of DOC in groundwater were quite low at this site (Figure 8). DOC concentrations in leachate following residuals application at Deadwater Pit in Maine (Catricala et al. 1996) were on average 10 times higher than those reported here. DOC concentrations were significantly higher for the biosolids treatment wells compared to the control and upgradient wells (Figure 7). However, these increases in DOC concentrations in groundwater in the biosolids plot compared to the control and upgradient wells were relatively small (35%) compared to the greater than 900% increase observed for $NO₃-N$ in groundwater.

There was a significant relationship between DOC and $NO₃$ -N for groundwater wells ($r²$) $= 0.514$, p < 0.01) suggesting that the application and stockpiling of organic biosolids material at the site was the common driving factor for increases in both DOC and $NO₃-N$. Although increases in DOC and $NO₃-N$ in groundwater appear to be caused by the application of biosolids, the relatively small increase in DOC compared to $NO₃-N$ suggests that there is a shortage of available carbon for microbial metabolism. Therefore, the microbial community where biosolids have been applied, particularly in stockpile areas, is utilizing the process of nitrification to generate energy for microbial metabolism. The process of nitrification provides the microbial community with the energy it requires through the conversion of NH_4 -N to NO_3 -N. This subsequently creates an excess of NO_3 -N in the system that is leached to the groundwater. As mentioned earlier, pH and conductivity values provide further support the hypothesis that nitrification of mineralized organic nitrogen from stockpile areas is causing the excessively high $NO₃-N$ values in certain groundwater wells.

Metals, Pesticides, Volatile and Semi-volatile Organics

Analysis of metals was conducted at UNH for samples collected prior to the biosolids application in October 1999 through the end of the project in November 2001. Samples for dissolved metals indicate that concentrations of arsenic, cadmium, chromium, copper, nickel, lead, selenium and zinc are all well below the NHDES and EPA safe drinking water limits for groundwater (Figure 9; for graphs of all metals analyzed see Appendix B).

Concentrations for these metals were not significantly different between biosolids and control wells. Upgradient wells showed a significantly lower concentration than other wells for cadmium, nickel and selenium. The downgradient well was also significantly lower in nickel than the biosolids and control wells (Figure 10).

Figure 9: Concentrations for arsenic, cadmium, and lead using PVC wells $(7/99 - 11/01)$. Red circles are biosolids wells; blue squares are control wells; yellow triangles are upgradient wells; and green diamonds are downgradient wells.

Figure 10a: Means for arsenic, cadmium, chromium, and copper in groundwater with 95% confidence intervals shown as bars. Statistical differences between means are noted with lowercase letters (p<0.05), and were determined using One-way Analysis of Variance with 4 levels and Tukey's pairwise comparison.

Figure 10b: Means for nickel, lead, selenium and zinc in groundwater with 95% confidence intervals shown as bars. Statistical differences between means are noted with lowercase letters (p<0.05), and were determined using One-way Analysis of Variance with 4 levels and Tukey's pairwise comparison.

The Massachusetts Water Resources Authority (MWRA) also analyzed a small number of samples for a wide variety of compounds before and after biosolids application. Results for metals are for total rather than dissolved metals, which includes sediments within the water sample. Groundwater samples at this site are quite variable with regard to the

represent the environmentally significant dissolved fraction in groundwater at the site. Their results show that some of the highest concentrations of metals were observed in control or up-gradient wells, where according to hydrologic characterization, as well as $NO₃-N$ data, there has been no impact of biosolids application. There were no pesticides detected in any of the treatment or control wells. The only semi-volatile organic compound detected was bis(2ethylexyl) phthalate or DEHP, which is a typical laboratory and field contaminant. Based on the total amount of DEHP present in the biosolids and SPF which has been applied at the site, loading rates do not exceed the NH DES standard for contaminated soils of 39 mg/kg. This standard is designed to ensure that groundwater concentrations of DEHP do not exceed safe drinking water standards of 6 ug/L in groundwater. As with the metals, DEHP was detected in both control and up-gradient wells in addition to treatment wells (see Appendix B for more information).

Conclusions

The monitoring demonstration at this biosolids application site in Hooksett, NH has produced several key findings. First, there are significant increases in average $NO₃-N$ concentrations in groundwater when wells beneath biosolids treatment areas are compared to control and upgradient wells at the site. However, the concentrations of $NO₃-N$ in groundwater both within and downgradient from the biosolids treatment area show high spatial variability. This suggests that $NO₃-N$ contamination has not resulted from the relatively uniform biosolids applications, but rather has resulted from nonuniform stockpiling at the site. Based on the management history of the site and the location of areas of high NO_3-N concentrations in groundwater we are convinced that this stockpiling activity is the most likely cause of deleterious impacts on groundwater.

We have also found moderately increased levels of $NO₃-N$ in groundwater upgradient from the biosolids treatment area and in the control field suggesting that there may be a secondary source of contaminated water entering the site from the surrounding area. Although this secondary source contributes to the high levels observed beneath some of the biosolids application area, it is not a major contributor and can not explain the very high $NO₃-N$ concentrations observed at several of the wells located in or adjacent to the treatment area.

We believe that gravel pit management practices can lead to nitrogen saturation of soils (Aber et al. 1989), a condition in which soil microbes and plants can no longer utilize available N in a predictable or effective way, resulting in contamination of groundwater with nitrogen. This was most likely the case at the Hooksett site where past stockpiling and application activity may have led to an inability of the soil to utilize the available N provided by the biomix application in October of 1999. Although available nitrogen appears to be high, the available carbon at the site appears to be quite low. This lack of available carbon as an energy source for microbial processing has led to increased levels of nitrification causing an increase in $NO₃-N$ production and a subsequent leaching of $NO₃-N$ to the groundwater.

The stated purpose of this project was to examine the impacts of current management practices associated with biosolids and SPF reclamation activity on groundwater quality. We can say unequivocally that the management practices employed at this site are not adequate to protect water quality, but we cannot identify with certainty which aspect of site management was most responsible for the groundwater contamination. The timing, magnitude, and frequency of biosolids applications, as well as on-site stockpiling, could all have contributed to the elevated levels of nitrate in groundwater. Based on the data we have collected, we believe that stockpiling is the most likely cause of the elevated $NO₃-N$ concentrations. Stockpiling is the only cause of elevated $NO₃-N$ that would show strong spatial variability. If the magnitude (application rate) or frequency of biosolids application were responsible for the elevated $NO₃-N$ levels, we would expect high and relatively uniform concentrations of $NO₃-N$ in the treatment area and in downgradient wells. Likewise, if the timing of the last application were primarily responsible for the elevated concentrations, we would expect a large pulse of $NO₃$ in the spring following a fall application, and significant declines in subsequent months. Instead, we observed greatest concentrations over a year after the last application. Therefore, we believe that past stockpiling activities is the most reasonable explanation for the increases in groundwater $NO₃-N$ observed at this site.

Concentrations for other nitrogenous compounds and dissolved organic carbon were consistently low for both biosolids and control wells. Similarly, dissolved metals concentrations were consistently low and well below the NHDES and EPA safe drinking water standards for groundwater. Based on the data collected, $NO₃-N$ is the only considerable risk to groundwater quality at this site.

Further work with a strong experimental design (including groundwater data before biosolids application and replicated sites) would be necessary to document the impacts of one-time applications of biosolids to reclaim gravel pits. Our data show that repeated stockpiling at a gravel pit carries significant risks of groundwater contamination, but they should not be used to condemn all gravel pit reclamation with biosolids.

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Appendix A – Martin's Ferry, Hooksett – Soil Solution Chemistry & Previous Data

Tension lysimeters were installed at depths of 50-60 cm for collecting soil solution. Vacuum (tension) on lysimeters was checked prior to sample collection to ensure that samplers were still drawing soil solution. Lysimeter samples were evacuated into a vacuum flask. After sample collection, a vacuum was applied (0.5 to 1.0 bars of tension) to each lysimeter and the lysimeter tubes were clamped until the next collection. Soil solution sampling was discontinued in the fall of 1999 due to technical difficulties and a lack of need for more information. Levels of $NO₃-N$ in soil solution were extraordinarily high in the biosolids plot at Hooksett in 1997, but have decreased in recent years. Concentrations were as much as three orders of magnitude greater than the control area (Figure A1).

Figure A1: $NO₃-N$ concentrations for the entire study period (1997-2000) for Martin's Ferry, Hooksett. $NO₃-N$ in soil solution from both Biosolids and Control plots.

Due to various complications in the field including lysimeter damage by animals, dry conditions and sandy soils, we were unable to collect soil solution samples for the summer and fall in 1998 at this site (except for June 26, 1998). Even following repair of lysimeters, sample collection was limited. Statistical analysis of soil solution data was not appropriate, given these problems in sampling frequency and number.

Data for average groundwater concentrations using stainless steel wells beginning in June 1998 suggests similar patterns to that seen in subsequent PVC wells. Although the spatial coverage of the stainless steel wells was poor, we still find that the wells closest or immediately downgradient from the stockpiling areas showed periodic increases in NO₃-N (Figure A2).

Figure A2: Groundwater $NO₃$ -N concentrations for stainless steel wells from June 1998 to December 1999. (a) Control wells; (b) Treatment wells. Lines represent individual wells (3 control wells, 3 treatment wells).

Appendix B – Martin's Ferry, Hooksett – Metals and MWRA Data

Metals Data

Metals data collected and analyzed by UNH found no elevated levels for any constituents above the NH DES or EPA allowable limits for safe drinking water. The remainder of the timeline graphs for metals in groundwater wells is shown below.

Figure B1: Concentrations of trace metals over time (July 1999 through November 2001). Red circles are biosolids wells; blue squares are control wells; yellow triangles are upgradient wells; and green diamonds are downgradient wells.

MWRA Data

Prior to the biomix application in October of 1999, one week after application and approximately one month after application, additional water samples were collected by UNH for analysis of PCBs, total metals, volatile organic compounds and semi-volatile organic compounds by the Massachusetts Water Resources Authority (MWRA). Samples were collected with a Teflon bailer cleaned with nitric acid and methanol and placed into clean sample bottles provided by MWRA. Samples were kept refrigerated and delivered to MWRA within 48 hours of collection. UNH also submitted to MWRA two equipment blanks and a field duplicate as part of QA/QC procedures. No analyses of PCBs, total metals, or VOC's were conducted by UNH with NEBRA funding.

Data on total metals, PCB's, semi-volatile and volatile organic compounds were at or below detection limits for most constituents analyzed. A complete list of all constituents analyzed is included in Table B1. Detection limits for MWRA were quite high for some compounds, particularly metals because samples were not filtered prior to analysis. Data for metals represent total concentrations, rather than the soluble fraction. Interpretation of the metals data is difficult because EPA and NH standards, as well as other research studies, use dissolved metals rather than the total metals reported by MWRA.

Concentrations of many total metals and DEHP (bis(2ethylexyl) phthalate) were actually higher prior to application of the biosolids and SPF mixture than after application, and levels were sometimes elevated within control wells (H-14 and H-15). In fact, some of the highest levels recorded were found in control wells (Table B2). Concentrations of all constituents analyzed were very low in the Merrimack River both upstream and downstream of the application area. The only organic found with some frequency was DEHP, which MWRA characterizes as a common laboratory contaminant (Steve Rhode, pers. comm.). The loading rates of DEHP for the site over the last five years do not exceed the NH DES standard for DEHP of 39 mg/kg, which is designed to protect groundwater concentrations from exceeding the standard of 6 ug/L. Given these results, we do not believe there is any evidence in this data set showing negative effects of residuals on trace organic contaminants or metals. Pesticides were not detected in either treatment or control wells. Additional analysis of dissolved metals will be conducted with separate funding by a UNH Ph.D. candidate during 2000-2001.

Table B1: Complete list of all constituents analyzed by MWRA. All constituents were below detection limits for all samples and for all dates except for those data reported in Table B2. Detection limits are in μ g/L unless otherwise noted.

Table B2: MWRA data for constituents above detection limits. Total metals are unfiltered. All data are in µg/L unless otherwise noted. DEHP represents bis(2ethylexyl) phthalate. Rivu and Rivd represent upstream and downstream samples on the Merrimack

River, respectively. $C =$ control wells, $X =$ experimental wells, $U =$ up-gradient wells. A mixture of biosolids and short paper fiber was applied to the experimental wells on October 25, 1999.

Appendix C – Hooksett Individual Well Data for NO3-N

We present in this Appendix the nitrate concentrations over the length of this experiment for the individual wells (biosolids, control, upgradient, and downgradient).

Appendix D – Ambrose Pit - New Hampton, NH

Project Description

The Ambrose Pit (New Hampton, NH) is a 35-acre sand and gravel operation that has had reclamation activity with residuals for the past five years. Active reclamation was begun in 1998 for a non-reclaimed area. This site was evaluated over a multi-year period for nitrate, ammonium, dissolved organic nitrogen (DON) and dissolved organic carbon (DOC) in soil solution and groundwater. At the site, various treatment plots were established that utilize different residual materials including a manufactured topsoil (biosolids and SPF) plot and a biosolids only plot.

At each plot within the site tension lysimeters were installed at depths of 50-60 cm for collecting soil solution. Wells were also installed to monitor groundwater quality beneath the treatment plot, and at locations presumed to be hydrologically up- and down-gradient of the treatment plots (for more information see NEBRA Progress Report May 1998 – July 1999).

Treatment plot 1 (50 ft by 100 ft) was begun in April 1998 using biosolids only with an application rate of 3830 #N/acre; however in November 1998 the area was scraped of all topsoil and a manufactured topsoil of biosolids and SPF was applied with an application rate of 3770 #N/acre and a C:N ratio of 30:1. A map of the study area is included (Figure C1). Native soils consist of Endoaquents and Udorthents that are sandy and well drained (Order 1 Soil Survey). The site is currently permitted under Env-Ws 800 as a facility and site appropriate for biosolids and SPF utilization.

Sample Collection

Samples of soil solution and groundwater were collected every two weeks during the growing season (August 1 - November 1) and were continued on a monthly basis through the winter (December - April). Vacuum (tension) on lysimeters was checked prior to sample collection to ensure that samplers were still drawing soil solution. Lysimeter samples were evacuated into a vacuum flask. After sample collection, a vacuum was applied (0.5 to 1.0 bars of tension) to each lysimeter and the lysimeter tubes were clamped until the next collection. Groundwater wells were evacuated with a teflon bailer until three volumes of water within the well had been exchanged prior to sample collection, where feasible. If recharge rates were slow, less than three volumes were withdrawn. Samples from lysimeters and groundwater wells were placed in clean HClwashed polyethylene bottles (HDPE) and remained on ice in coolers until delivery to UNH for chemical analysis. Samples are filtered with a 0.7 μ m GF/F ashed filter and frozen until analysis.

Figure C1: Map of Site A, the Ambrose Pit in New Hampton, NH. Open squares indicate groundwater wells and solid circles indicate soil lysimeters. Scale = $1\degree \approx 250$ ft.

Results

Sampling at the Ambrose site was discontinued in the fall of 1999 due to technical and management issues. Biosolids treatment plots were small and difficult to characterize hydrologically. Control wells were located in an area hydrologically and geologically different from the treatment plots. Because these control wells showed high concentrations of $NO₃-N$ with no biosolids addition, it is impossible to say whether biosolids application had an effect on groundwater at this site.

The long-term record for $NO₃-N$ at this site indicates that soil solution concentrations are higher at Plot #1 where biosolids have been applied then at the control plot. Biosolids

Date

Figure C2: $NO₃-N$ concentrations for the entire study period (1997-1999) at Ambrose Pit, New Hampton. (a) $NO₃$ -N concentrations in soil solution and groundwater for all treatment and control plots; (b) $NO₃$ -N concentrations for groundwater collected from biosolids, biomix and control plots. First arrow indicates approximate date of the first biosolids application, second arrow indicates date of biomix application.

were applied to Plot #1 in the spring of 1998 (early April), resulting in the increased concentrations of $NO₃-N$ from August to November and a subsequent increase during the late summer and fall of 1999 (Figure C2). Although concentrations of NO_3-N for soil solution were higher for the biosolids plot than the control plots, we only have one lysimeter within the biosolids treatment area. Therefore, statistical analysis on soil solution was not performed.

Nitrate concentrations in groundwater for the biosolids only treatment were lower than the control, and this difference was statistically significant. At several times following biosolids application in April, groundwater did exceed the EPA allowable level of 10 mg/L. Because this level was exceeded even more frequently in control wells, no conclusions can be drawn regarding the impact of biosolids application on groundwater quality. Due to operator error at the site (destruction of a plot), data after November 1998 represent only one sampling well.

Following biosolids application at this site a conservation mix of grasses was planted as the primary vegetative cover. This type of vegetation grows very rapidly, subsequently utilizing much of the available nitrogen applied. Growing vegetation (particularly conservation mix grasses) with biosolids fertilization in gravel pit reclamation may actually reduce NO₃-N concentrations in groundwater below those found under ambient conditions at this site.

Concentrations of $NO₃-N$ in groundwater at the biomix site were consistently well below the EPA maximum allowable limit for drinking water. This corroborates the findings of Catricala et al. 1996, who determined that a C:N ratio greater than 20:1 significantly reduces the impacts of $NO₃-N$ on groundwater quality. Unlike with the biosolids only application, $NO₃-N$ concentrations did not increase during the late summer and fall when plant growth decreased.

Groundwater concentrations show significant differences between biomix, biosolids and control wells with higher concentrations of DOC and NH4-N in the biomix and biosolids wells than in control wells (Figure C3). However, $NH₄$ -N concentrations were consistently low for treatment and control wells. Concentrations of DOC and NH4-N were not significantly different between biomix and biosolids wells. Concentrations of DON in groundwater were not significantly different (p>0.05) between biomix and biosolids and control data, and overall concentrations were typically low.

Figure C3: Least Squares Means for NO₃-N, NH₄-N, DON and DOC in groundwater using One-way Analysis of Variance with 3 levels (biomix, biosolids and control).

Appendix E – Letarte Pit – Tamworth, NH

Project Description

This site was evaluated over a multi-year period for nitrate, ammonium, dissolved organic nitrogen (DON) and dissolved organic carbon (DOC) in soil solution and groundwater. At the site, treatment plots were established that utilized short paper fiber (SPF) only.

At the site both treatment plots and control areas were identified, and lysimeters and groundwater wells were installed in both areas. At each plot of the three sites tension lysimeters were installed at depths of 50-60 cm for collecting soil solution. Wells were also installed to monitor groundwater quality beneath the treatment plot, and at locations presumed to be hydrologically up- and down-gradient of the treatment plots (for more information see NEBRA Progress Report May 1998 – July 1999).

Figure D1: Map of Site B, Letarte Gravel Pit in Tamworth, NH. Open squares indicate groundwater wells and solid circles indicate soil lysimeters. Scale = $1" \approx 150$ ft.

The Letarte Pit is located in Tamworth, NH and is a 20-acre sand and gravel operation that has had reclamation activity with residuals in the past. Active reclamation was begun in 1997 for a non-reclaimed area. SPF (primary/secondary mix with high N) was applied to a small treatment plot (50 ft by 50 ft) in September 1997 with an application rate of 2598 #N/acre and a C:N ratio of 19:1. A map of the study area is included (Figure D1).

Native soils consist of Udorthents that are sandy and excessively drained (Order 1 Soil Survey). The site is currently permitted under Env-Ws 800 as a site appropriate for utilization of biosolids.

Sample Collection

Samples of soil solution and groundwater were collected every two weeks during the growing season (August 1 - November 1) and were continued on a monthly basis through the winter (December - April). Vacuum (tension) on lysimeters was checked prior to sample collection to ensure that samplers were still drawing soil solution. Lysimeter samples were evacuated into a vacuum flask. After sample collection, a vacuum was applied (0.5 to 1.0 bars of tension) to each lysimeter and the lysimeter tubes were clamped until the next collection. Groundwater wells were evacuated with a teflon bailer until three volumes of water within the well had been exchanged prior to sample collection, where feasible. If recharge rates were slow, less than three volumes were withdrawn. Samples from lysimeters and groundwater wells were placed in clean HClwashed polyethylene bottles (HDPE) and remained on ice in coolers until delivery to UNH for chemical analysis. Samples are filtered with a 0.7 μ m GF/F ashed filter and frozen until analysis.

Results

Data collection at the Letarte Pit ceased in the fall of 1999 due to technical and maintenance issues. The Letarte Pit is an active gravel pit where excavation of sand and gravel was being performed within meters of groundwater monitoring wells for both treatment and control areas, making it impossible to generate accurate and meaningful data. The long-term record for the Letarte Pit indicates $NO₃-N$ concentrations in both groundwater and soil solution for the SPF treatment are significantly higher than those of the control (Figure D2), but the fact that concentrations increased substantially prior to SPF application makes it difficult to interpret this result.

Short paper fiber was applied to the treatment area on September 25, 1997. On the sampling date prior to the SPF application (September 9, 1997), both soil solution and groundwater showed a large increase in nitrate concentrations. This five-fold increase in groundwater $NO₃-N$ concentrations resulted in concentrations that exceeded the EPA maximum allowable limit prior to SPF application. Levels continued to be high for fall samplings after the SPF application (Figure D2).

Groundwater concentrations returned to below EPA allowable levels during the summer of 1998 but again exceeded 10.0 mg/L during the fall of 1998 and were still significantly elevated compared to control wells (Figure D2). Sampling could not be continued long enough to determine whether this pattern continued in the fall of 1999.

Vegetation at the SPF treatment plot is predominatly Lamb's Quarters (*Chenopodium album*), which grows rapidly in the spring and summer. Unlike the conservation mix grasses planted at the Ambrose Site it begins to die back early in the fall (September). The increases in nitrate concentrations we observed coincided with the dieback of Lamb's Quarters, and thus we conclude vegetation at the site may play a very important part in regulating nitrate concentrations of groundwater.

Figure D2: $NO₃-N$ concentrations for the entire study period (1997-1999) for Letarte Pit,

Tamworth. (a) $NO₃-N$ in both soil solution and groundwater for SPF and control plots; (b) $NO₃-N$ in groundwater only for both SPF and control plots.

Concentrations of DON and NH4-N in groundwater samples collected between June 1998 and April 1999 followed a similar pattern to $NO₃-N$ with significantly higher concentrations in the SPF treatment area than in the control area (Figure D3), however overall concentrations of both constituents were quite low. Concentrations of DOC in both groundwater and soil solution were not significantly different.

Figure D3: Least Squares Means for $NO₃-N$, NH₄-N, DON and DOC for groundwater at the Letarte Pit in Tamworth using One-way Analysis of Variance with 2 levels (SPF and control).