Streamside Management Zone Effectiveness for Protecting Water Quality Following Forestland Application of Biosolids

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Thesis submitted to the Faculty of the Virginia Polytechnic Institute and State University
in partial fulfillment for the requirements for the degree of

Master of Science
in
Forestry

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July 14, 2008
Blacksburg, Virginia

Key Words: loblolly pine (*Pinus taeda* L.) plantation, biosolids, forest fertilization,
Streamside Management Zone (SMZ)

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Abstract

Biosolids, materials resulting from domestic sewage treatment, are surface applied to forest soils to increase nutrient availability. Retaining streamside management zones (SMZs) can limit nutrient pollution of streams. We delineated 15 m SMZs along three intermittent streams in an 18-year-old *Pinus taeda* L. plantation. We applied biosolids outside the SMZ on one side of each of the streams maintaining the other side of the stream as control. We collected water samples from the three treated and six reference streams as well as from the perennial stream both upstream and downstream from the intermittent streams for 12 months following treatment. Along transects perpendicular to the treated streams, we collected overland flow samples, soil solution samples at 60 cm and extracts from ion exchange membranes (IEMs) placed in the surface soil. We found elevated nitrate concentrations outside the SMZ in the treated side soil solution samples, in which concentrations remained below 1.5 mg L^{-1}. Nutrient concentrations outside the SMZ in treated side IEM extracts increased following biosolids application, returning to near control levels after one year. Nutrient concentrations in IEM extracts were not elevated adjacent to the streams. We observed elevated phosphorus concentrations adjacent to the stream in overland flow during one period on the treated side of the stream. Stream nutrient concentrations showed few differences downstream from the treatment with concentrations below 1.5 mg L^{-1}. Our results indicate that a 15 m SMZ protected streams from nutrient pollution for the first year following biosolids application to adjacent forestlands.

Acknowledgements

The Metropolitan Washington Council of Governments, the Sustainable Engineered Materials Institute, and the Forest Nutrition Cooperative provided funding for this project. Synagro Technologies, Inc. assisted in the installation of the study.

I express thanks to committee members Dr. T.R. Fox, Dr. W.M. Aust and Dr. G. Evanylo for their thoughts, encouragement, patience and mentoring. Thank you to Mike Aust for asking tough questions and ensuring that I remembered the graduate student's role. Thank you to Greg Evanylo for keeping a solid perspective on our work. Thank you to Tom Fox for offering the opportunity to come to Tech and for guiding me through my coursework and our research. Your dedication to and work ethic in all aspects of the university is remarkable.

I greatly appreciate David Mitchem, Tal Roberts, Colleen Carlson, and Eduardo Arellano for lending expertise and time to our research project. I also appreciate the administrative support of Kathie Hollandsworth, Sue Snow and Tracey Sherman without which I would have been lost in a sea of impenetrable bureaucracy. Thank you to the Department of Forestry graduate students for all our conversation whether it was over the finer points of denitrification, the future of forestry or how best to spend the weekend. Finally, thanks to my family for setting me on my life's path of which this has been a very pleasant step.

Table of Contents

List of Tables

List of Figures

Chapter I: Introduction

Rationale

Biosolids are the materials resulting from domestic sewage treatment that have been processed in order to be used for land application (Evanylo, 1999b). The 7.1 million dry metric tons of biosolids produced nationally in 2004 were used or disposed of through land application, incineration, or landfilling (Beech et al., 2007). Being rich in macro- and micronutrients, biosolids can be used as an alternative to conventional chemical fertilizers or animal manure on nutrient deficient sites (Evanylo, 1999b).

Most biosolids are applied to agricultural land both nationwide and in Virginia. Of the 3.2 million dry metric tons of biosolids applied to land nationwide in 2004, 74% were put to agricultural use (Beech et al., 2007). Of the 160,000 dry metric tons of biosolids produced in Virginia in 2004, 31% were land applied, nearly all to agricultural land (Beech et al., 2007). In Virginia, 224,000 dry metric tons of biosolids were land applied, which was higher than the amount produced in the commonwealth due to the importation of biosolids from neighboring states and districts such as Washington, D.C. (Beech et al., 2007). All of the 481,000 dry metric tons of biosolids produced in Washington, D.C. in 2004 were land applied, nearly all of it to agricultural land (Beech et al., 2007).

As the agricultural land base decreases, the reliance on agricultural land for biosolids land application could be problematic for wastewater treatment facility managers. Nationwide cropland decreased 3.2 million hectares or 2% from 1997 to 2003, which follows a long pattern during which cropland decreased 21 million hectares or 12% from 1982 to 2003 (USDA Natural Resources Conservation Service, 2007). In Virginia, cropland decreased 22,000 hectares from 1997 to 2003, which follows a long pattern, decreasing 216,000 hectares from 1982 to 2003 (USDA Natural Resources Conservation Service, 2000, 2007). The shrinking amount of available land could be a problem for disposal of biosolids if measures are not taken to account for the decreased land base.

In order to ensure land is available for biosolids application, wastewater treatment facility managers are seeking to diversify the lands applied with biosolids to include more forestland. While the forestland base is experiencing a similar decline as agriculture, it is currently underutilized for biosolids application. Only 1% of nationwide, 8% of Washington D.C., and none of Virginia's biosolids are applied to forestland (Beech et al., 2007).

Forest managers make use of biosolids as a source of macro- and micronutrients on nutrient deficient sites, especially in intensively managed forest plantations. Fertilizing stands improves productivity, reduces rotation times, and increases profits, allowing fiber production on a decreased land base (Fox et al., 2007a, Fox et al., 2007b). However, fertilizer prices are increasing, causing fertilization to be too costly and encouraging forest managers to seek inexpensive alternatives to conventional chemical fertilizers (Albaugh et al., 2007). Biosolids are often available for no cost to the landowner and could fill this need (Evanylo, 1999b).

There are environmental concerns with the land application of biosolids. One concern is that biosolids, characterized by high nitrogen and phosphorus levels, will contribute to non-point source pollution and eutrophication of waterways (Boesch et al., 2001). As the biosolids release these nutrients to the soil, nutrient loads in adjacent streams or groundwater could increase due to runoff and subsurface movement or leaching depending on site characteristics and practices used (Elliott et al., 2002, Grey and Henry, 2002, McLaren et al., 2003).

These impacts can be mitigated through the use of Streamside Management Zones (SMZ). SMZs are protective buffers between streams and nutrient supplemented areas that can, along with other benefits, reduce nutrients moving both on the surface and through the subsurface to streams (Lowrance et al., 1984, Peterjohn and Correll, 1984, Lowrance et al., 1997, Mayer et al., 2007). The width of the SMZ required to achieve the desired nutrient reductions depends on site characteristics and management practices (Castelle et al., 1994).

Two entities in Virginia suggest or require buffers on forestland applied with biosolids. The Virginia Department of Forestry recommends a 15 m SMZ adjacent to all forestry operations (Virginia Department of Forestry, 2002). The Virginia Department of Environmental Quality (DEQ) regulates land application of biosolids in Virginia. DEQ requires stream buffers of variable width dependent upon season and slope and stream type (Evanylo, 1999c). The effectiveness of these buffers has yet to be empirically tested with the recommendations and requirements based on conventional widths.

Objectives and Hypothesis

The purpose of this study was to investigate the effectiveness of SMZs for reducing nutrient movement to streams adjacent to planted loblolly pine (*Pinus taeda* L.) stands on biosolids-amended soils. We collected leachate, subsurface ion availability, and surface runoff samples to assess each of the potential pathways of nitrogen and phosphorus loss to streams and groundwater. We compared water from streams in watersheds applied with biosolids to watersheds without biosolids to evaluate cumulative effects on water quality in streams. The goal of our research was to ensure that current Virginia biosolids land application practices for forestland protect water quality.

The overall objective was to investigate nitrogen and phosphorus movement from biosolids to waterways with a SMZ in place. We hypothesized that biosolids application would not significantly decrease water quality because the SMZ will attenuate the various forms of nitrogen and phosphorus released from the biosolids. This was based on the previously reported success of buffers to protect water quality from nutrients (Lowrance et al., 1984, Peterjohn and Correll, 1984, Lowrance et al., 1997, Mayer et al., 2007).

The sub-objectives of our study are to investigate if nutrients move across the SMZ in subsurface or surface flow or through the soil column and to determine if biosolids application affects water quality on a small watershed scale. Phosphorus and nitrogen in the form of ammonium will both be mostly immobile as these nutrients are bound to organic matter and clay in the soil (Mengel and Kirkby, 2001). Both phosphorus and ammonium may be attached to soil particles carried in runoff, but the runoff deposits these sediments as it crosses the SMZ (Lowrance et al., 1984). Nitrate, as an anion, is more mobile than ammonium or phosphorus, but would be attenuated through denitrification and plant uptake (Verchot et al., 1997a, Tabacchi et al., 2000).

Chapter II: Literature Summary

Forestland Application of Biosolids

Biosolids are "solid, semi-solid, or liquid materials, resulting from treatment of domestic sewage that have been sufficiently processed to be safely land-applied (Evanylo, 1999b)." Other definitions include "sewage sludge that has been treated to meet the regulatory requirements for land application set out in the Code of Federal Regulations (National Research Council, 2002)" and "a sewage sludge that has received an established treatment for required pathogen control and is treated or managed to reduce vector attraction to a satisfactory level and contains acceptable levels of pollutants, such that it is acceptable for use for land application, marketing or distribution in accordance with this chapter (Virginia Code, 1995)." The term is intended to distinguish biosolids from raw sewage sludge or sewage sludge containing large amounts of pollutants.

Typical wastewater treatment processes produce biosolids rich in nutrients such as nitrogen and phosphorus. Anaerobic digestion of biosolids is one process that stabilizes sewage sludge using microbes in oxygen free environments. Though nutrient concentrations vary among wastewater treatment facilities and seasons, a survey of anaerobically digested biosolids, found median concentrations of 4.2 percent total nitrogen and 3.0 percent total phosphorus (Sommers, 1977). Other sources indicated total phosphorus levels closer to half the level of total nitrogen (Smith, 1996). The median concentration of specific inorganic forms of nitrogen were 1600 mg L^{-1} ammonium and 79 mg L^{-1}nitrate (Sommers, 1977).

Biosolids application can increase nutrients in forest soils and improved growth of trees. Biosolids application to a planted radiata pine (*Pinus radiata* D.Don*)* stand significantly increased nitrogen concentration and doubled phosphorus concentration in the forest floor for at least five years following application (McLaren et al., 2007). Planted radiata pine (*Pinus radiata* D. Don) stands in New Zealand applied with biosolids showed volume growth up to 46% over control in year 13 (Wang et al., 2006b). Biosolids applied to a northern hardwood stand at nitrogen rates of 200, 400, and 800 kg per ha increased nitrate concentrations in the soil to 115, 290, and 356 kg per ha, respectively, compared to the control of 33 kg per ha (Aschmann et al., 1992). The addition of macronutrients has contributed to increased foliar weight in red (*Pinus resinosa L.)* and white pine (*Pinus strobus L.)* stands, and therefore increased photosynthetic potential of trees (Brockway, 1983). Biosolids applied sites have shown increased and longer term growth response over conventional chemical fertilizers (Brockway, 1988, Henry et al., 1993). In another study, growth response was similar to conventional chemical fertilizers (Prescott and Blevins, 2005). Some concern exists over the increase of undesirable characteristics in trees such as increased branch width, but the economic benefit of increased volume outweighed these concerns in *Pinus radiata* D. Don (Kimberley et al., 2004).

The amount of biosolids applied to a tract of land is typically calculated according to nitrogen demands of the crop grown (Evanylo, 1999a, Maguire et al., 2000). Typical rates of application to nitrogen deficient loblolly pine forestland in the southeastern United States is between 168 and 224 kg of nitrogen fertilizer ha^{-1} (Fox et al., 2007a). Increasingly, policy makers and scientists are concerned with phosphorus saturation of repeatedly amended soils and have begun to move to a phosphorus-based application rate (Elliott and O'Connor, 2007). Typical rates of application to phosphorus deficient loblolly pine forestland in the southeastern United States is 28 kg of phosphorus ha^{-1} (Fox et al., 2007b).

When applying biosolids based on nitrogen demand, application rates are given in kg of plant available nitrogen (PAN) ha^{-1} (Pierzynski, 1994). PAN accounts for the variable availability of the different forms of nitrogen in the biosolids. One equation for PAN for biosolids equals the initial nitrate content plus the non-volatilizing fraction of ammonium plus the mineralizing fraction of organic nitrogen (Evanylo, 1999a).

$$PAN = Organic\ N\ mineralized - Ammonium\ Volatilized + Nitrate$$

The availability of the fractions of the different forms of nitrogen in biosolids is determined empirically. For anaerobically digested biosolids, volatilization rates of ammonium, which are greatest in the first twenty-four hours, are predicted to be 50% of the ammonium and are negatively influenced by the presence of vegetative cover, low temperatures, and low pH (Donovan and Logan, 1983, Hall and Ryden, 1986). Gilmour et al. (2003) found 37% of organic nitrogen in biosolids mineralized per year in the United States, with rates varying with biosolids decomposability, weather, and nitrogen content. Nitrogen mineralization rates are influenced by soil type, application rate, and type of sludge (Garau et al., 1986). Wang et al. (2003) found nitrogen mineralization rates to be 15 percent of the organic nitrogen per year for anaerobically digested biosolids in New Zealand.

Another factor influencing the PAN is the denitrification rate. Denitrification rates are not included in the PAN formula listed above because nitrate is such a small fraction of the nitrogen in the biosolids used in this study. However, biosolids applied soils have higher denitrification rates than soils without biosolids, decreasing the amount of nitrate available from the biosolids (Chaussod, 1983).

The release of nutrients from biosolids raises environmental concerns as both nitrogen and phosphorus have been shown to leach down the soil profile and runoff in overland flow because the nutrients are supplied in high concentrations. When biosolids application rate is based on nitrogen, the phosphorus supplied typically exceeds crop demand (Evanylo, 1999b, Maguire et al., 2000). The excess phosphorus applied in association with nitrogen-based rates of biosolids application is increasingly an area of environmental concern (Elliott and O'Connor, 2007). Using the median nitrogen and phosphorus percentages in anaerobically digested biosolids listed above, the suggested nitrogen-based rate for biosolids application for forestland of 224 kg of PAN ha^{-1}, would provide 77 kg of plant available phosphorus ha^{-1}, exceeding the suggested phosphorus rates for loblolly pine of 28 kg ha^{-1}.

Phosphorus leaching from biosolids-amended soils depends on the root zone, the soil texture, and the associated metals in the biosolids. Phosphorus leaching from biosolids and effluent treated soil is insignificant below the root zone for many soils (Reikerk and Zasoski, 1979, Urie, 1979, Kofoed and Klausen, 1986). Phosphorus leaching from sandy biosolids-amended soils was less than one percent, while conventional fertilizers were as high as 21 percent with leaching dependent upon iron and aluminum in the biosolids (Elliott et al., 2002). Another study on phosphorus leaching from sandy, iron and aluminum poor, biosolids-amended soils showed significantly increased leaching over control (Alleoni et al., 2008). Another study showed that top soil retained the phosphorus released from the biosolids on sandy biosolids-amended soils with repeated applications (Su et al., 2007). The iron and aluminum released from biosolids plays a role in mitigating phosphorus leaching concerns because these metals increase the phosphorus sorption capacity of amended soils (Maguire et al., 2000, Su et al., 2007). Calcium released from biosolids also plays a role in mitigating phosphorus leaching concerns because the calcium precipitates with phosphorus to form calcium phosphate at soil pH above 6 (Woodward et al., 2007).

Nitrate leaching from biosolids-amended soils depends on application rate, vegetation, and timing. Leaching of nitrate from biosolids, which is comparable to conventional fertilizers, increases with higher application rates (Kofoed and Klausen, 1986, Aschmann et al., 1992). Leachate concentrations decrease if biosolids are applied to areas with significant potential for plant uptake such as established forestland (Reikerk and Zasoski, 1979, Furrer and Stauffer, 1986, McLaren et al., 2007). Nitrate concentrations in leachate following a spring application of biosolids remained at background levels for a year before increasing during the dormant season and returning to background after two years (Brockway, 1983).

Biosolids have complex interactions with overland flow, decreasing the amount of runoff but increasing nutrient concentration in the runoff depending on the number of applications and the process used to create the biosolids. Biosolids-amended soils have lower amounts of runoff than non-amended soils because the material increases infiltration rates and surface roughness (Mostaghimi et al., 1989, Bruggeman and Mostaghimi, 1993). However, nitrogen and phosphorus concentrations in the runoff are increased because biosolids increase nutrient concentrations in the soil (Smith and Evans, 1977, Kladivko and Nelson, 1979, Mostaghimi et al., 1989, Alleoni et al., 2008). Overall nitrogen and phosphorus losses through runoff have been found to be less than one percent of the amount added (Kladivko and Nelson, 1979, Dunigan and Dick, 1980). Repeated application of biosolids can saturate the soil with phosphorus, causing higher phosphorus concentration in runoff and leachate than for soils treated once (Penn and Sims, 2002, Sukkariyah et al., 2007). Phosphorus released from biosolids produced using biological nutrient removal systems is more mobile than biosolids produced using iron only systems (Penn and Sims, 2002).

Streamside Management Zones (SMZs)

Streamside management zones (SMZs) are forested areas adjacent to streams that protect water quality (Virginia Department of Forestry, 2002). SMZs are also referred to as riparian areas or forests buffer strips. Effective SMZs will filter nutrient and sediment moving from upland sites toward aquatic areas in addition to other benefits.

The objectives, nutrient of concern, buffer width, and scale of observation influence the degree of water quality protection an SMZ provides. Objectives for the SMZ, such as sediment and nutrient removal, biodiversity promotion, and water temperature moderation, lead to varying buffer width recommendations (Bren, 1993, Castelle et al., 1994). SMZs provide mechanisms to attenuate both surface and subsurface movement of nitrogen and phosphorus (Muscutt et al., 1993, Vought et al., 1994). Nutrient removal is directly related to buffer width, with wider buffers providing more protection (Dickey and Vanderholm, 1981). Most studies evaluating SMZ effectiveness study the hill slope scale, but watershed scale studies have shown that forested riparian areas maintain water quality at larger scales as well (Dosskey, 2001, Potter et al., 2004).

Suggested buffer widths vary greatly, from 3 to 200 m (Barling and Moore, 1994, Castelle et al., 1994). Castelle et al. (1994) found in a broad review of a variety of buffers in both agricultural and forested settings that 5 to 10 m provide little protection for most objectives under most conditions, while 15 to 30 m buffers provide significant resource protection. If a manager is solely interested in maintenance of chemical integrity of waterways, 15 m is sufficient (Castelle et al., 1994). An intensively managed Radiata pine (*Pinus radiata* D. Don) stand exported less than 1% of the nitrogen and phosphorus applied to the stand to the stream with a 30 m SMZ in place (Hopmans and Bren, 2007). Buffer conditions can affect the suggested width (Dickey and Vanderholm, 1981, Burt et al., 2002).

The Virginia Department of Forestry recommends a minimum 15 m SMZ (Virginia Department of Forestry, 2002). This recommendation applies to streams with a defined channel, typically perennial and intermittent streams. A perennial stream holds water throughout the year, while an intermittent stream goes dry for some period of the year, but has a defined channel with evidence of scouring and an exposed rock or soil streambed. The agency recommends wider SMZs adjacent to trout fisheries and municipal water supplies dependent upon slope.

The Virginia Department of Environmental Quality requires variable buffer widths for streams adjacent to areas applied with biosolids. The buffers are dependent on factors such as season of application, slope, and stream channel classification. On slopes less than 7 percent, perennial streams must have a 15 m buffer, while intermittent streams require a 7.5 m buffer (Virginia Code, 1995). During winter on slopes greater than 7 percent, the distance doubles to 30 m and 15 m, respectively (Virginia Code, 1995).

Attenuation of Nitrogen and Phosphorus in Subsurface Flow in an SMZ

A number of mechanisms within the riparian area of the SMZ exist for attenuating nitrogen and phosphorus in the subsurface including: immobilization, denitrification, and plant uptake. The relative effectiveness and importance of each of these mechanisms depends on the type of nutrient, site characteristics, and season.

Soil Immobilization of Nitrogen and Phosphorus

The two primary forms of inorganic nitrogen behave differently in soil solution, which affects their movements (Mengel and Kirkby, 2001). Ammonium (NH_4^+) is largely immobile because, as a cation, it binds to negatively charged clay minerals. Ammonium can oxidize to nitrate and nitrate can reduce to ammonium (Samson et al., 1990). Nitrate (NO_3^-), as an anion, is mobile in the soil solution and, as an oxidized form of nitrogen, is present in aerated soils. Denitrification is the reduction of nitrate to nitrite (NO_2^-), and further reduction to nitrous oxide (N_2O) or nitrogen gas (N_2). Nitrogen is the first element in the sequence of reduction reactions following imposition of anaerobic conditions with a critical redox potential of 250 to 300 mV (Patrick and Jugsujinda, 1992). Organic matter, donating electrons, facilitates this reduction, translocating nitrogen from the soil to the atmosphere. The translocation avoids nitrogen saturation of the soil and maintains the soil's ability to attenuate nitrogen.

Organic forms of nitrogen can contribute to plant available forms of nitrogen in the soil (Mengel and Kirkby, 2001). Biosolids contain a high percentage of organic nitrogen (Evanylo, 1999b, Wang et al., 2003). In order to make use of this nitrogen, plants rely on microbial mineralization of organic nitrogen to ammonium through the process of ammonification. The carbon supply and aeration of the soil mediates the rate of the relatively slow process of ammonification as well as the subsequent process of nitrification of ammonium to nitrate.

Different factors in the soil environment determine the availability of phosphate in soil solution and its ability to move through the subsurface (Mengel and Kirkby, 2001). For basic soils, calcium cations and pH control calcium phosphate formation and dissolution, which in turn determines the concentration of phosphate in the soil solution (Woodward et al., 2007). For acidic soils, the presence of clay minerals, metal hydroxides, and organic matter controls the specific adsorption of phosphate and phosphate concentrations in the soil solution (Mitsch and Gosselink, 2000). In well-aerated soils, phosphate can be adsorbed to the surface of clays or occluded within a ferric iron-hydroxy skin (Mengel and Kirkby, 2001). In water-saturated soils, the ferric iron can be reduced to ferrous iron, which is water soluble, releasing phosphate to soil solution. Phosphate added to the soil can be fixed in the processes describe above, but, lacking a process similar to denitrification, phosphate attenuation can become limited as the soil becomes saturated.

Though largely immobile in soil solution, organic phosphorus is a potential source of labile phosphate (Mengel and Kirkby, 2001). Some plants have the potential to produce the enzyme phosphotase in their roots, which facilitates hydrolysis of organic phosphorus, adding phosphate to the soil solution. Soil microbes and fungi can also produce phosphotase, increasing phosphate in soil solution.

Denitrification in a SMZ

An important form of inorganic nitrogen in nitrogen-amended soils is nitrate, which could move across a SMZ. In the upland area at the edge of the SMZ, ammonium may be adsorbed to soil colloids or oxidize to nitrate, which is mobile. As the nitrate in the soil solution moves into the SMZ, the riparian soil will promote denitrification of nitrate to nitrogen gas as long as carbon from soil organic matter provides sufficient electrons and the soil saturation is appropriate.

Denitrification is an important mechanism for attenuating nitrate moving from a treated area through a SMZ (Verchot et al., 1997a). Floodplains attenuate nitrate more efficiently than upland zones, which is frequently attributed to denitrification (Simmons et al., 1992). Some riparian areas were observed as being an order of magnitude more efficient at denitrification than upland sites (Meding et al., 2001).

The saturation of the soil determines denitrification (Davidson and Swank, 1986). The concentration of nitrate in soil solution is correlated with dissolved oxygen concentration, indicating the presence of oxygen inhibits denitrification (Hill et al., 2000). Saturated floodplains in the coastal plain of North Carolina were very efficient at removing nitrate from the system through denitrification (Brinson et al., 1984). The poorer draining of two fertilized soils had a higher denitrification rate than the better drained soil (Schnabel and Stout, 1994). However, completely aerobic soils, such as those in upland sites, are an order of magnitude more efficient at denitrification than completely anaerobic soils, such as those that may exist in a continuously flooded soils (Reddy and Patrick, 1975). In continuously flooded soils, denitrification occurs because the soil has a thin aerobic layer in which ammonium oxidizes to nitrate which leaches into an underlying anaerobic layer (Patrick and Tusneem, 1972).

The denitrification potential of transition zones between upland areas and floodplains is dependent on the position of the water table level. The zone of denitrification in soils is typically narrow, associated with a redox front optimum for denitrification (Haycock and Pinay, 1993, Hedin et al., 1998, Hill et al., 2000). The redox front where denitrification rates are highest may be close to the stream or close to the upslope, depending on the riparian width and saturation of the soil across a riparian area (Cooper, 1990, Schnabel et al., 1996).

The denitrification rate is dependent on the fluctuations in the water table. The faster the rate of flux in the water table alternating between aerobic and anaerobic conditions, the faster the rate of denitrification because redox potential rapidly responds to changes in oxygen levels (Reddy and Patrick, 1975). Soil denitrifiers make use of nitrate immediately upon the transition from aerobic to anaerobic conditions (Starr and Parlange, 1975).

If a soil has the necessary water saturation, then nitrate can be a limiting factor for denitrification near the soil surface which is rich in organic matter and carbon (Ambus and Lowrance, 1991). The entire fertilizer nitrogen input was removed from some systems through this process (Lowrance et al., 1984). Several studies have shown that as concentrations of nitrate increase, so too does denitrification (Cooper, 1990, Samson et al., 1990, Schnabel et al., 1996).

If a soil has the necessary water saturation, then carbon can be a limiting factor for denitrification deep in the soil profile. When carbon availability increased, soils from the coastal plain of Georgia increased the concentration of nitrogen gas expelled and decreased the concentration of nitrate in soil solution (Obenhuber and Lowrance, 1991). Decaying roots supply carbon to the root zone, facilitating denitrification (Parkin and Meisinger, 1989, Simmons et al., 1992). However, deeper in the soil, carbon concentrations are lower and nitrate concentration is inversely correlated with carbon concentration at increasing depth (Hill et al., 2000). Denitrification rates have been shown to be lower deeper in the soil than near the surface because of carbon limitations (Lowrance, 1992, Schnabel et al., 1996). Carbon may be limiting in certain ecosystems which have a steady supply of nitrate (Obenhuber and Lowrance, 1991, Schnabel et al., 1996). Repeated nitrogen additions can deplete carbon stores, reducing denitrification rates (Meding et al., 2001). As biosolids are rich in organic carbon, they have the potential to increase denitrification rates if carbon supply limits a site's denitrification activity.

Plant Uptake of Nitrogen and Phosphorus

Plant uptake and storage represents a potential nitrogen sink because nitrogen in the inorganic ammonium and nitrate forms is an essential macronutrient (Mengel and Kirkby, 2001). Capable of taking up both forms of inorganic nitrogen from the soil, plants metabolize ammonium in order to create chlorophyll required for photosynthesis and thus for all energy production in plants. Plants reduce nitrate to ammonium internally, changing the nitrate to a useful form. In addition to chlorophyll, all proteins, amino acids, nucleic acids, and coenzymes contain nitrogen.

Due to their ability to internally recycle nitrogen, the role of plants in attenuating nutrients in the SMZ is less certain than the role of denitrification. Through mass balance calculations, studies have shown denitrification to be the dominant mechanism over plant uptake for removing nitrogen from the soil solution in certain environments (Jacobs and Gilliam, 1985, Verchot et al., 1997a). However, nitrate attenuation rates were higher during the growing season than in the dormant season indicating the importance of vegetation (Lowrance, 1992, Simmons et al., 1992, Pinay et al., 1993). Other studies have found that plants take up as much nitrate as they have access to, reducing nitrate concentrations across a buffer strip (Peterjohn and Correll, 1984, Tabacchi et al., 2000).

Similar to nitrogen, phosphorus is an essential macronutrient for plants (Mengel and Kirkby, 2001). Occurring largely as phosphate, inorganic phosphorus in soil solution is plant accessible. However, most phosphorus in the soil complexes with organic matter and mineral soil, limiting the amount available to plants. Plants and microbes quickly take up any phosphate in soil solution.

Ammonium and Phosphorus in a SMZ

Ammonium can leach if a site has a low cation exchange capacity or the exchange sites of the soil are saturated. If ammonium is the dominant inorganic form, then nitrification can be a limiting factor on nitrate availability and thus denitrification (Wittgren and Tobiason, 1995). Nitrification becomes a limiting factor if cation exchange capacity from colloids is insufficient to immobilize the ammonium and provide surface area for microbial activity required for oxidation to nitrate. Rich in organic matter, biosolids increase the exchange capacity of a soil, mitigating this limiting factor. The proficiency of anaerobic riparian zones for attenuating ammonium is less pronounced, as the ammonium accumulates at cation exchange sites. Ammonium is only oxidized to nitrate during the dry season and could saturate exchange sites if left in a saturated environment, leaving ammonium mobile in the soil solution (Brinson et al., 1984).

Phosphorus can become a concern in subsurface flow if soils are saturated for a broad width around a stream and nitrate concentrations are low. Denitrification is the energetically preferred reduction over the reduction of ferric iron to ferrous iron (Jordan et al., 1993, Mitsch and Gosselink, 2000). However, if both oxygen and nitrate are not available, then soil microbes reduce iron, releasing phosphate sorbed to the iron (Cooper et al., 1995, Carlyle and Hill, 2001). During the dormant season, as ground water levels rise and reduce the level of dissolved oxygen in the soil solution, more iron reduction leads to more phosphorus release (Mulholland, 1992). The lack of plant uptake during the dormant season increases phosphorus mobility. The decomposition of organic matter can also release phosphorus (Osborne and Kovacic, 1993).

Attenuation of Nitrogen and Phosphorus in Surface Flow in an SMZ

Streamside management zones attenuate nitrogen and phosphorus as they flow across the surface of the buffer zone. Many nutrients are bound to sediment, which can be carried in overland flow (Muscutt et al., 1993). SMZs reduce the sediment load through increased infiltration, reduced flow velocity, and vegetative filters (Overcash et al., 1981, Peterjohn and Correll, 1984, Muscutt et al., 1993).

Buffer strips are effective at trapping sediment (Neibling and Alberts, 1979). SMZs are most effective at reducing large particle sediment load (Alberts et al., 1981, Hayes et al., 1984). However, the smaller clay-sized particles that are not attenuated sorb the most nutrients (Alberts et al., 1981, Cooper and Gilliam, 1987). Despite the preferential sorption of phosphorus to smaller particles, SMZs can reduce nutrient loads up to seventy percent (Alberts et al., 1981).

Saturation of the SMZ and elevated water tables can reduce infiltration rates, reducing the effectiveness of the SMZ for reducing runoff (Benston et al., 1988). Repeated simulated rainfalls and models indicate the importance of low water saturation conditions for improving infiltration (Asmussen et al., 1977, Munoz-Carpena et al., 1999). Large rainfall events can produce large amounts of runoff as soils become water saturated, providing high potential for overland flow (Daniels and Gilliam, 1996).

The vegetation in the buffer strip plays an important role in controlling nutrients carried in runoff (Gurnell, 1997, Tabacchi et al., 2000). Vegetation takes up water, reducing saturation of the buffer zone and increasing infiltration (Gurnell, 1997, Verchot et al., 1997b). Vegetation increases SMZ surface roughness, slowing overland flow rates and increasing sedimentation (Smith, 1989, Gurnell, 1997).

Channels through the SMZ can reduce its effectiveness. Shallow uniform flow across a buffer strip efficiently traps sediment, infiltrating overland flow. Concentrated flow paths channel water, reducing infiltration of overland flow (Dillaha et al., 1989). SMZs with channelised flow require longer distances to attenuate the same amount of nutrients as SMZs with shallow uniform flow (Dickey and Vanderholm, 1981).

The effectiveness of SMZs for attenuating phosphorus in runoff can decrease overtime. Repeated rainfall simulations produced more phosphorus in the second simulation as the phosphorus holding capacity of the SMZ declined (Dillaha et al., 1988).

Phosphorus has no mechanism like denitrification to translocate phosphorus to the atmosphere (Brinson et al., 1984). However, input of fresh sediment can mitigate phosphorus saturation, covering the phosphorus saturated surface soil with unsaturated soil (Cooper and Gilliam, 1987).

Other Considerations for Attenuation of Nitrogen and Phosphorus

SMZs are a potential mechanism for holding nitrogen and phosphorus released to the soil from biosolids. Ammonium will be bound to soil particles, taken up by plants and undergo ammonification to nitrate. Plants will take up nitrate, but much will still remain and be mobile. As nitrate moves in the soil solution toward the streams and into saturated soils, it will undergo denitrification and plant uptake. As long as proper anaerobic conditions exist, nitrate should be completely attenuated because carbon, released from the biosolids, will donate the necessary electrons. Phosphorus released from the biosolids will likely be held by the iron and aluminum or calcium in the soil and biosolids. Water-born nutrient-bearing sediments will settle onto the surface of the SMZ due in part to the decreased runoff caused by the increased infiltration resulting from the biosolids material. A wealth of literature exists on both the release of nutrients from biosolids and water quality protections provided by SMZs. Our research will provide additional understanding of the ability of SMZs to protect water quality from nutrients lost from biosolids-amended forest soil.

Chapter III: The Effectiveness of Streamside Management Zones for Protecting Water Quality Following Forestland Application of Biosolids

Abstract

Biosolids, materials resulting from domestic sewage treatment, are surface applied to forest soils to increase nutrient availability. Retaining streamside management zones (SMZs) can limit nutrient pollution of streams. We delineated 15 m SMZs along three intermittent streams in an 18-year-old *Pinus taeda* L. plantation. We applied biosolids outside the SMZ on one side of each of the streams maintaining the other side of the stream as control. We collected water samples from the three treated and six reference streams as well as from the perennial stream both upstream and downstream from the intermittent streams for 12 months following treatment. Along transects perpendicular to the treated streams, we collected overland flow samples, soil solution samples at 60 cm and extracts from ion exchange membranes (IEMs) placed in the surface soil. We found elevated nitrate concentrations outside the SMZ in the treated side soil solution samples, in which concentrations remained below 1.5 mg L^{-1}. Nutrient concentrations outside the SMZ in treated side IEM extracts increased following biosolids application, returning to near control levels after one year. Nutrient concentrations in IEM extracts were not elevated adjacent to the streams. We observed elevated phosphorus concentrations adjacent to the stream in overland flow during one period on the treated side of the stream. Stream nutrient concentrations showed few differences downstream from the treatment with concentrations below 1.5 mg L^{-1}. Our results indicate that a 15 m SMZ protected streams from nutrient pollution for the first year following biosolids application to adjacent forestlands.

Introduction

Biosolids, materials resulting from domestic wastewater treatment that have been processed in order to be used for land application, can be used as a source of macro- and micronutrients (Evanylo, 1999b). The vast majority of the 3.2 million dry metric tons of biosolids applied to land in 2004 were put to agricultural use (Beech et al., 2007). As the nationwide cropland base decreased 3.2 million ha or 2% from 1997 to 2003, sites available for biosolids disposal through surface application could become scarce (USDA

Natural Resources Conservation Service, 2007). This decline is disproportionally larger in areas around major urban areas, requiring greater transportation distances to biosolids disposal sites. In order to ensure that land is available for biosolids application, wastewater treatment facility managers and biosolids disposal contractors are interested in applying biosolids to forestland. The forestland base is currently underutilized for biosolids application with 1% of biosolids applied to forestland (Beech et al., 2007).

Fertilization of intensively managed forest stands is a common silvicultural practice throughout the southern and northwestern United States. In the south, many soils become limited in both nitrogen and phosphorus as stands mature. Fertilizing intermediate age loblolly pine (*Pinus taeda* L.) stands on nitrogen and phosphorus limited soils improves tree growth which reduces rotation times, increases profits, and allows fiber production on a decreased land base (Fox et al., 2007a). However, fertilizer prices are increasing, causing decreased financial returns from conventional fertilization and encouraging forest managers to seek less expensive alternatives (Albaugh et al., 2007). Biosolids are often available for little or no cost and could fill this role (Evanylo, 1999b).

Like managers of cropland, forest managers can use biosolids as a source of macro- and micronutrients on nutrient deficient sites to increase tree growth, especially in intensively managed forest plantations. Stand growth response to biosolids amendment is long lasting and similar to conventional chemical fertilizers. Douglas fir (*Pseudotsuga menziesii* (Mirb.) Franco) stands in the Pacific northwest treated with biosolids showed height growth responses up to 72% over control for 10 years following treatment (Henry et al., 1993). White pine (*Pinus strobus* L.) stands in Michigan treated with biosolids showed significantly increased radial growth response compared to the control (Brockway, 1983). Radiata pine (*Pinus radiata* D. Don) stands in New Zealand treated with biosolids increased volume growth up to 46% over control 13 years following application (Wang et al., 2006b). Western red cedar (*Thuja plicata* Donn ex D. Don), western hemlock (*Tsuga heterophylla* (Raf.) Sarg.), and amabilis fir (*Abies amabilis* (Dougl.) Forbes) stands in the northwest treated with biosolids showed volume growth responses similar to stands treated with chemical fertilizers (Prescott and Blevins, 2005).

Eutrophication of waterways is a major environmental concern. Non-point source pollution of waterways through leaching and runoff of nitrogen and phosphorus has lead to eutrophication in many waterways such as the Chesapeake Bay and its tributaries (Boesch et al., 2001). Land applied biosolids, characterized with high nitrogen and phosphorus concentrations, release these macronutrients to the soil in labile forms (Sommers, 1977). Nitrogen and phosphorus leaching and runoff from biosolids amended-soils showed increases over controls depending on site characteristics and application method (Elliott et al., 2002, Grey and Henry, 2002, McLaren et al., 2003, Alleoni et al., 2008).

As a protective buffer between streams and treated areas, streamside management zones (SMZs) reduce nutrients moving both on the surface and through the subsurface to streams (Lowrance et al., 1984, Peterjohn and Correll, 1984, Lowrance et al., 1997, Mayer et al., 2007). The width of the SMZ required to achieve the desired nutrient reductions depends on site characteristics, management practices, and other factors (Castelle et al., 1994). Castelle et al. (1994) suggests that a 15 m buffer may provide for protection of the chemical integrity of adjacent streams. In Virginia, the Department of Forestry recommends a 15 m SMZ and the Department of Environmental Quality requires a 15 m buffer between intermittent streams and areas applied with biosolids during winter on slopes greater than 7% (Virginia Code, 1995, Virginia Department of Forestry 2002).

The objective of this study was to evaluate the effectiveness of a 15 m SMZ for protecting water quality in streams adjacent to forestland where biosolids were applied. Hillslope scale studies evaluating the effectiveness of buffers for mitigating non-point source pollution should investigate both subsurface and surface movement of nitrogen and phosphorus in order to monitor the different pathways to surface waters. We used ion exchange resins, tension lysimeters, and runoff collection canisters to evaluate inorganic nitrogen and phosphorus concentrations in biosolids-amended soils under planted loblolly pine (*Pinus taeda* L.) across an SMZ adjacent to intermittent streams in the Virginia Piedmont. We hypothesized that nutrient levels would be elevated in biosolids-amended areas outside the SMZs, but decline to control levels adjacent to streams within the SMZs.

Methods and Materials

Site Description

We used three small watersheds (1.0-1.4 ha) located at 37.20873 N, -77.7929 W in the Appomattox River watershed about 50 km southwest of Richmond, VA for this study. The site is located in the Piedmont physiographic province, which has a humid, subtropical climate. In nearby Dinnwiddie County, VA, the mean (1973-1993) annual temperature was 21°C with a mean January temperature of 2°C and a mean July temperature of 25°C. The mean (1973-1993) annual precipitation was 116 cm with July and August being the wettest months. We observed annual precipitation of 91 cm and typical monthly precipitation, except during the months of December 2006, February, March, August, September and November 2007, which were over 3 cm below average (Figure 1). These dry months likely did not alter our findings, as results from atypically dry months were similar to those for average months in the same season. We used a weather station located in Crewe, VA approximately 40 km west of the study site to characterize daily mean temperature and precipitation during the study period (Figure 2).

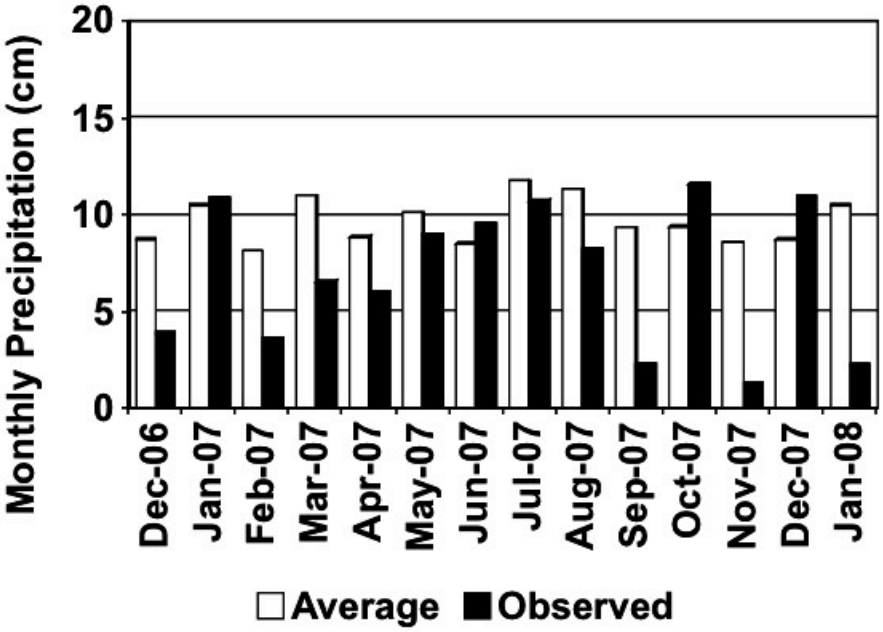

Figure 1: Average (Dinnwiddie County, 1973-1993) and Observed Total Monthly Precipitation

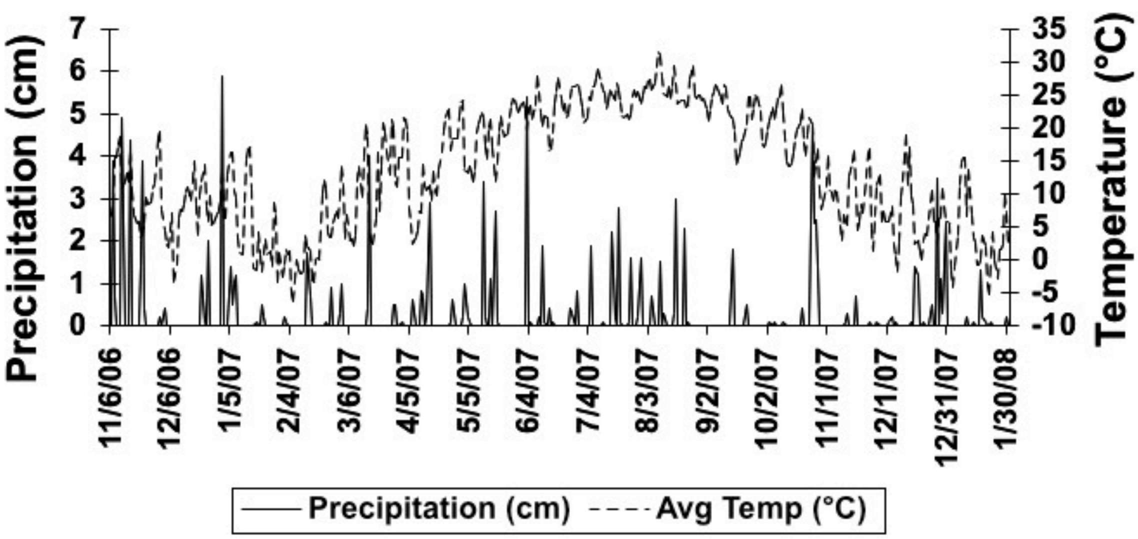

Figure 2: Daily Mean Temperature and Daily Total Precipitation during study period at Crewe, VA

Metamorphic bedrock underlies the watersheds. Intermittent streams drain these watersheds through rolling topography flowing into the perennial Long Branch Creek, which drains to the Appomattox River. Watershed elevation ranges from 76 to 91 m above sea level. The soils were historically used for the intensive cultivation of crops, which has led to the erosion of the A-horizon. The soils tend to be low in inherent fertility. The dominant upland soil is the Appling series, a fine, kaolinitic, thermic Typic Kanhapludult, with the A-horizon heavily eroded due to past land use and an argillic B-horizon. The dominant lowland soil is the Enoree series, a coarse-loamy, mixed, semi-active, nonacid, thermic Aeric Fluvaquent, with an 18 cm A-horizon underlain by a strongly gleyed C-horizon. The current stand of loblolly pines was planted on the site in 1990. The site index for loblolly pine was 24 m at base age 25. The treated stands were recently subject to a mid-rotation fifth row thinning. The basal area was 31.9 m^2 ha^{-1} for non-thinned stands and SMZs and 25.7 m^2 ha^{-1} for thinned stands.

We collected and analyzed soil samples taken from 0-15 cm for pH, phosphorus, iron, carbon, nitrogen, and estimated cation exchange capacity (Table 1). Following air-drying and grinding, we passed samples through a 2 mm sieve. We determined soil pH in a 1:1 soil/water ratio after a 30 minute equilibration. We analyzed phosphorus, iron, and cations of a Mehlich I extraction of the soil using ICAP-AES following the Virginia Cooperative Extension methodology (Mehlich, 1953, Donohue, 1992). We determined total carbon and nitrogen by combustion using a CNS analyzer (Elementar America, Inc, Laurel, NJ). We calculated estimated CEC according to Virginia Cooperative Extension Soil Test Laboratory's methodology (Donohue and Heckendorn, 1994).

Table 1: pH, Mehlich I-extractable P and Fe, total C and N, and Virginia Cooperative Extension Soil Test Laboratory estimated CEC in top 15 cm of untreated soils.

pH	P	Fe	C	N	Est. CEC
	-------------------------mg kg^{-1} --------------------------				$cmol_c$ kg^{-1}
4.77	2.67	51.53	8,600	490	3.43

Study Design and Treatment

We delineated 15 m SMZs on both sides of three intermittent streams adjacent to the loblolly pine stands (Figure 3). Biosolids application occurred outside the SMZ on one side of each of the three streams in early December 2006. We left one side of each stream untreated to serve as a control. Each stream is a block and each streamside is a treatment for a total of three blocks of two treatments. Six other untreated intermittent streams on the site serve as a reference to evaluate treatment effects on stream water.

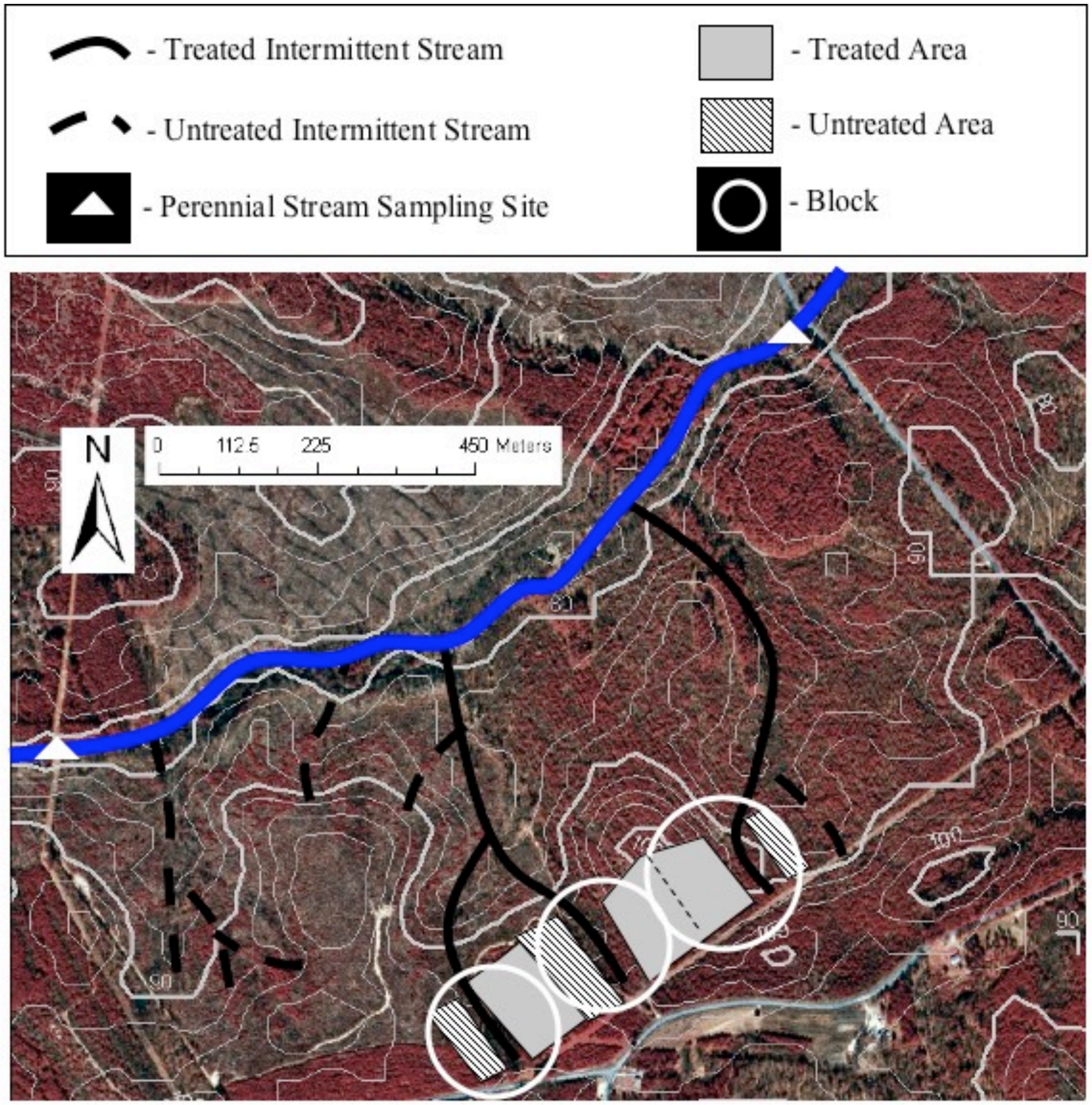

Figure 3: Map of biosolids SMZ study site and study design in Amelia County, VA. Contour intervals are 2.5 m.

A skidder equipped with a side discharge spreader was driven between the rows of planted loblolly pine to apply the biosolids into the stand. We applied biosolids to a total of 3.5 ha of loblolly pine stands at a rate of 1120 and 629 kg ha^{-1} of total nitrogen and total phosphorus, respectively, which we calculated provided 268 and 68 kg ha^{-1} of plant available nitrogen and plant available phosphorus, respectively (Evanylo, 1999a). The target application rate was to provide 224 kg of plant available nitrogen ha^{-1}, which is the standard nitrogen fertilization rate for intensively managed loblolly pine plantations (Fox et al., 2007a). Typical mid-rotation phosphorus fertilization rates for intensively managed loblolly pine plantations is 28 kg ha^{-1} (Fox et al., 2007a). We collected samples on a biweekly to monthly basis for one year following application of biosolids from December 2006 to January 2008.

We used anaerobically digested biosolids obtained from the Alexandria Sanitation Authority, Alexandria, VA (Table 2). A&L Eastern Laboratories, Inc (Richmond, VA) analyzed the biosolids for total and volatile (organic matter) solids using SM2540G and nitrate (NO_3-N) using SM4500-NO3F (APHA, 1998), total Kjeldahl nitrogen (TKN-N) and ammonium (NH_4-N) using USEPA 351.3 and USEPA 350.2, respectively (USEPA, 1983), phosphorus (P), potassium (K), sulfur (S), calcium (Ca), magnesium (Mg), sodium (Na), iron (Fe), manganese (Mn), cadmium (Cd), copper (Cu), lead (Pb), molybdenum (Mo), nickel (Ni), and zinc (Zn) using SW846-6010B (USEPA, 2002), arsenic (As), mercury (Hg), and selenium (Se) using SW846-7061A , SW846-7471A , and SW 846-7741A, respectively (USEPA, 2002), pH using SW 846-9045C (USEPA, 2002), and calcium carbonate equivalent (CCE) using AOAC 955.01 (Kane, 2000).

Table 2: Properties of anaerobically digested biosolids applied to loblolly pine plantation.

Property	Concentration (mg kg^{-1})
Total Solids	270,200
Volatile Solids	592,700
TKN-N	57,100
NH4-N	13,000
Organic N	44,100
NO3-N	6
P	32,200
K	1,100
S	9,100
Ca	27,700
Mg	3,100
Na	500
Fe	46,400
Mn	839
Cd	2
Cu	356
Pb	54
Mo	14
Zn	1,410
Ni	26
As	6.4
Hg	2.8
Se	7.1
pH	8.1
CCE	2,600

Ion Availability Sampling

We used ion exchange membranes (IEMs) (Ionics, Watertown, MA) to determine ammonium, nitrate and phosphate availability in the surface soil. Ion exchange membranes have been used to study inorganic nitrogen and phosphorus availability in the soil (Cooperband and Logan, 1994, Huang and Schoenau, 1996).

We followed the methodology for preparation explained in Cooperband and Logan (1994). We cut the IEMs into 10 by 10 cm pieces. Every two to four weeks, we placed five sets of membranes in a transect perpendicular to the stream, with one set at each of the following positions: adjacent to the stream (0.5 m from stream), center of the SMZ (7.5 m from stream), inside the edge of the SMZ (14 m from stream), outside the edge of the SMZ (16 m from stream), and inside the application area (30 m from stream). Four IEMs composed a set, which included one anion and one cation exchange membrane placed horizontally at the boundary of the O and A horizon and one anion and one cation exchange membrane placed diagonally into the A horizon to a depth of 10 cm. Before being placed in the ground, we rinsed the IEMs with distilled water.

Following retrieval from the field, we brushed the IEMs clean of adhering soil and placed them in individual centrifuge tubes. We added 25 mL of 1 M potassium chloride solution to the tube, which was covered with Parafilm and capped. We mechanically shook the centrifuge tubes for 1 hour and filtered the supernatant into 25 mL scintillation vials through Whatman Grade 2 Qualitative Grade Circles and Sheets (Whatman International Ltd, Kent, United Kingdom). We stored the vials in a freezer for later analysis.

We cleaned and recharged the IEMs for later use. We rinsed and scrubbed IEMs with deionized water and mechanically shook them in a five percent by volume hydrochloric acid bath for five minutes. After a second rinsing with deionized water, we placed the IEMs in 8 L of 1 M sodium chloride solution for at least twenty-four hours before reinstallation.

Soil Solution Sampling

We collected soil solutions samples with porous cup tension lysimeters (Soilmoisture Equipment Corporation, Santa Barbara, CA) placed at the boundary of the B and C horizons at 60 to 80 cm deep. Lysimeters have been used to study both nitrogen and phosphorus leaching (Paramasivam et al., 2001, Woodward et al., 2007).

We attached the porous ceramic cups to 1 m long PVC pipe using epoxy. Before we installed the lysimeters in the soil, we rinsed them with deionized water. We placed five lysimeters in a transect perpendicular to the stream, with one lysimeter at each of the following positions: adjacent to the stream (0.5 m from stream), center of the SMZ (7.5 m from stream), inside the edge of the SMZ (14 m from stream), outside the edge of the SMZ (16 m from stream), and inside the application area (30 m from stream). On a biweekly to monthly basis, we hand pumped the lysimeters to a 50-kPa vacuum in order to draw soil solution through the pores of the cup. We collected samples from the cup the following day and returned them from the field under ice until placed in a freezer.

Overland Flow Sampling

We constructed overland flow collectors from 10 cm diameter, 38 cm long PVC pipe that we capped at the bottom of the pipe using epoxy. We placed a 5.7 cm racquetball inside the tube and glued a 10 by 5 cm flush bushing to the top of the tube in order to stop flow into the collector once it filled with water. We placed a screen over the top of the tube to prevent organic matter and soil material from entering, holding the screen in place with a 10 cm coupling. Installing stovepipe hats over the top of each collector prevented direct interception of rain.

In August 2007, we installed the collectors at 0.5 and 15 m from the intermittent stream channels in areas of ephemeral channelised flow, which are typically tree harvesting equipment trails from previous harvests. We also installed the collectors at 0.5 and 15 m from the intermittent stream in non-channelised areas where overland sheet flow could occur. The collectors were installed in holes in the ground so that the collector sampled water flowing over the A horizon. Overland flow samples were collected following large rainfall events that occurred during the study period. Samples were stored on ice for return to lab and frozen prior to analysis.

Samples from Intermittent and Perennial Streams

We collected grab samples from nine intermittent streams, including three treatment and six reference, by dipping a Nalgene bottle into the upper 15 cm of the water in the streams (Lurry and Kolbe, 2006). In addition, we collected two samples from Long Branch Creek: one upstream from the treatment and one downstream from the treatment. We returned samples from the field under ice before freezing. Samples were collected on a biweekly to monthly basis, placed on ice to return to the lab, and frozen until analysis.

Sample Analysis

We analyzed samples for nitrate and ammonium by colorimetry using a Traacs 2000 analytical console (Bran & Luebbe, Norderstedt, Germany). We analyzed samples for phosphorus by inductively coupled plasma optical emission spectrometry using a Vista-MPX CCD Simultaneous ICP-OES (Varian Analytical Instruments, Varian, Australia). We only analyzed IEM extracts for phosphorus every other month. For samples below detection limits, we replaced the no detection value with a value of half the detection limit (Smith, 1991).

Statistical Analysis

We analyzed all data using mixed models with repeated measures (Littell et al., 2006) including treatment and distance from stream as fixed effects, stream as a random blocking effect, and repeated measures occurring over time. We used the Kenward-Rogers method to calculate degrees of freedom in order to account for the repeated measures. Ammonium availability measured using the IEMs showed a strong relationship between the baseline measures and subsequent responses (p-value <0.05), so we included the baseline corrections in the model. All other models showed no significant relationship between the baseline measures and response, so we did not include baseline corrections in any other models. For the ammonium in overland flow canisters, we dropped the random error effects of the stream from the model in order to meet convergence criteria. We used a slice statement to test for differences between treatment and control at each location on each sampling date. We performed all analyses on SAS V9.1 (SAS Institute, Cary, NC).

Results

Nutrient Availability from Ion Exchange Membranes

The pattern of responses of IEMs placed in the soil and under the forest floor showed few differences. Therefore, we averaged the availability index of nutrients from the IEMs placed in the mineral soil and the forest floor to examine the nutrient availability in the surface soil horizons as a whole.

<u>Ammonium Availability from Ion Exchange Membranes</u>

Following biosolids application, ammonium showed an immediate and significant response outside the SMZ on the treatment side of the streams (Table 3). The significant response persisted at the sample locations 30 m from the stream immediately following the biosolids application through late spring. The significant response persisted at the sample locations 16 m from the stream beginning a month after application through late spring. Sample locations 14 m from the stream showed a significant response at a 0.1 alpha value in the beginning of March. In contrast, sample locations 0 m and 7.5 m from the stream never showed a significant difference between treatment and control sides of the stream. By mid-summer, ammonium response in biosolids applied areas returned to control levels. During sampling intervals in which ammonium availability was significantly elevated outside the SMZ, the availability declined to be the same as control levels within the SMZ (Figure 4). Ammonium availability estimates for the control side remained below 2 mg m^{-2} day^{-1} throughout the study period with one exception. Ammonium availability estimates outside the SMZ on the treated side of the stream during elevated periods ranged from 3.5 to 18.1mg m^{-2} day^{-1}.

Table 3: *p*-values for test of difference between control and biosolids treatment at each sampling location at each date for ammonium in ion exchange membranes. Values significant to alpha 0.05 are bolded.

Date	Distance from stream (m)				
	0	7.5	14	16	30
12/13/06	0.8696	0.9339	0.9788	0.6271	**<.0001**
12/28/06	0.7562	0.8145	0.9456	0.0726	**0.0002**
01/21/07	0.9258	0.9898	0.8656	**0.0210**	**<.0001**
02/03/07	0.8018	0.8577	0.9397	**0.0048**	**0.0051**
02/17/07	0.4722	0.3161	0.3447	**0.0003**	**<.0001**
03/06/07	0.5663	0.8994	0.0979	**0.0020**	**0.0005**
03/31/07	0.6308	0.9489	0.3007	0.1658	**0.0011**
04/28/07	0.5623	0.6553	0.2445	**0.0347**	**<.0001**
05/14/07	0.8076	0.8131	0.3182	0.7336	0.9128
06/01/07	0.8623	0.7386	0.1957	**0.0006**	0.4934
07/02/07	0.9878	0.7560	0.9600	0.7099	0.1162
07/18/07	0.9963	0.8407	0.7929	0.8518	0.7101
08/01/07	0.9564	0.8607	0.8730	0.9532	0.7948
08/14/07	0.9555	0.9956	0.9550	0.8636	0.9967
08/30/07	0.7279	0.8555	0.8289	0.8402	0.6591
09/15/07	0.7720	0.7308	0.9664	0.7577	0.8496
10/06/07	0.8687	0.9988	0.9311	0.9534	0.9686
12/16/07	0.9667	0.7808	0.9797	0.6788	0.9199

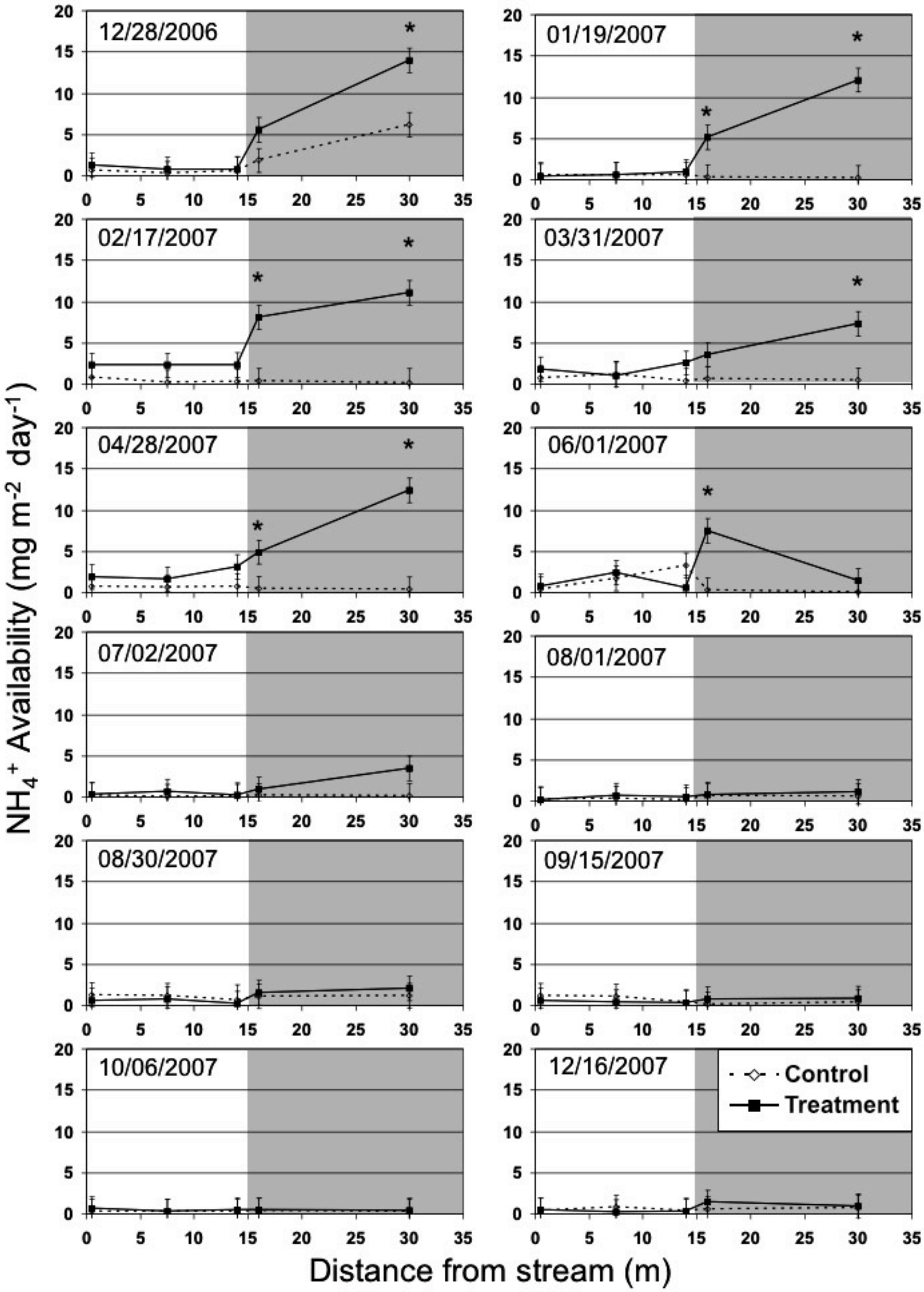

Figure 4: Ammonium availability from ion exchange membranes. The shaded region indicates area outside SMZ. Significant difference ($p < 0.05$) between control and treatment is indicated with *.

Nitrate Availability from Ion Exchange Membranes

Nitrate availability on the treated side of the stream showed a significant difference from control outside the SMZ beginning at the end of April (Table 4). At the sample locations 16 and 30 m from the stream the significant differences were detected in late spring, concurrent with the last sampling period during which ammonium was significantly elevated. The significant difference in nitrate persisted periodically throughout the summer, with non-significant differences corresponding to sampling intervals during periods of very low total rainfall. Results from ion-exchange resin bags have been shown to be sensitive to low field water conditions (Lajtha, 1987).

Nitrate availability on the treated side of the stream was occasionally significantly different from the control within the SMZ (Table 4). Sample locations 14 m from the stream showed a significant difference in late April and late August and nearly significant in the beginning of August (p-value = 0.1060). Sample locations 0 m and 7.5 m from the stream showed no significant difference between treatment and control sides of the stream except for during late April with 0 and 7.5 m significant to an alpha value of 0.1 and 0.0001 respectively. The late April sampling interval showed a significant difference between treatment and control sides of the stream across the entire SMZ to an alpha value of 0.1.

Table 4: *p*-values for test of difference between control and biosolids treatment at each sampling location at each date for nitrate in ion exchange membranes. Values significant to alpha 0.05 are bolded.

Date	Distance from stream (m)				
	0	7.5	14	16	30
12/13/06	0.8249	0.7481	0.6206	0.6106	0.6709
12/28/06	0.9259	0.9929	0.9544	0.9506	0.9805
01/21/07	0.8595	0.7977	0.8587	0.7479	0.9612
02/03/07	0.9347	0.9288	0.8679	0.2877	0.8251
02/17/07	0.5664	0.0971	0.5911	0.5770	0.9427
03/06/07	0.7452	0.9524	0.7088	0.1042	0.9319
03/31/07	0.2142	0.3945	0.2061	0.1407	0.3545
04/28/07	0.0833	**<.0001**	**0.0011**	**<.0001**	**0.0006**
05/14/07	0.7257	0.7034	0.4651	**0.0028**	**0.0085**
06/01/07	0.5668	0.5485	0.6912	**<.0001**	0.0819
07/02/07	0.5839	0.8283	0.2080	**0.0091**	**<.0001**
07/18/07	0.8760	0.7129	0.7749	0.4466	0.6035
08/01/07	0.6124	0.6417	0.1060	**0.0001**	**0.0002**
08/14/07	0.8369	0.9444	0.8187	0.5728	0.7692
08/30/07	0.7835	0.3087	**0.0015**	**0.0002**	**0.0172**
09/15/07	0.9591	0.9397	0.4591	0.5811	0.7122
10/06/07	0.9856	0.9977	0.8523	0.8623	0.8430
12/16/07	0.6544	0.6904	0.2821	**0.0231**	0.1264

During sampling intervals in which nitrate availability was significantly elevated outside the SMZ, the availability typically declined to control levels within 7.5 m of the outside edge of the SMZ (Figure 5). Nitrate extracted from ion exchange membranes increased following biosolids outside the SMZ and remained near control levels closer to the stream. Nitrate in the control remained below 1.8 mg m^{-2} day^{-1}. In contrast, following biosolids application outside the SMZ, nitrate ranged from ranged from 4.2 to 12.5 mg m^{-2} day^{-1}.

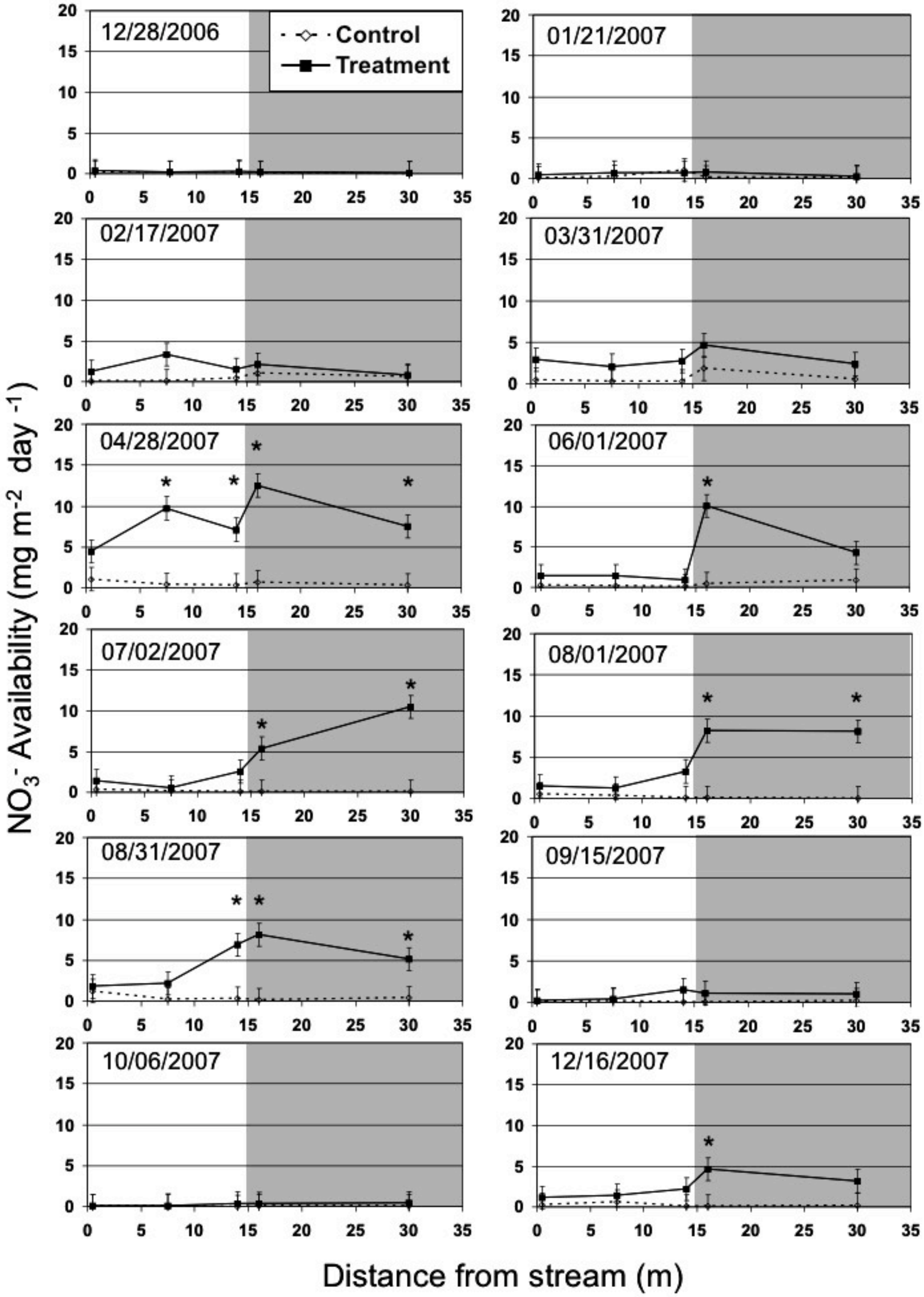

Figure 5: Nitrate availability from ion exchange membranes. The shaded region indicates area outside SMZ. Significant difference ($p < 0.05$) between control and treatment is indicated with *.

Phosphorus Availability from Ion Exchange Membranes

Similar to nitrate, phosphorus availability on the treatment side of the stream was significantly greater outside the SMZ, beginning in mid-winter (Table 5). At the sample locations 16 and 30 m from the stream significant differences were observed in mid-winter and persisted through summer except for 30 m from the stream in early July. Sample locations 14 m from the stream showed a significant response in late April and early August. Sample locations 0 m and 7.5 m from the stream never showed a significant difference between treatment and control sides of the stream. By the fall following application, phosphorus availability in biosolids applied areas returned to control levels.

Table 5: *p*-values for test of difference between control and biosolids treatment at each sampling location at each date for phosphorus in ion exchange membranes. Values significant to alpha 0.05 are bolded.

Date	Distance from stream (m)				
	0	7.5	14	16	30
12/28/06	0.9419	0.8373	0.8054	0.7809	0.9342
02/17/07	0.2481	0.6041	0.9798	0.1665	**0.0388**
04/28/07	0.1874	0.4041	**0.0262**	**0.0050**	**0.0194**
07/02/07	0.4553	0.2033	0.1811	**0.0003**	0.2329
08/01/07	0.4799	0.1353	**0.0003**	**<.0001**	**<.0001**
09/15/07	0.9871	0.8578	0.5031	0.2457	0.2129
12/16/07	0.9541	0.6491	0.4438	0.1998	0.1147

During sampling periods in which phosphorus availability was significantly elevated outside the SMZ, the availability was not different from control levels within 7.5 m of the outside edge of the SMZ (Figure 6). In areas where biosolids were applied phosphorus availability increased outside the SMZ, but were not elevated closer to the stream as shown in 08/01/2007. Phosphorus in the control remained below 0.3 mg m^{-2} day^{-1}, while it ranged from 0.6 to 1.8 mg m^{-2} day^{-1} outside the SMZ following application of biosolids.

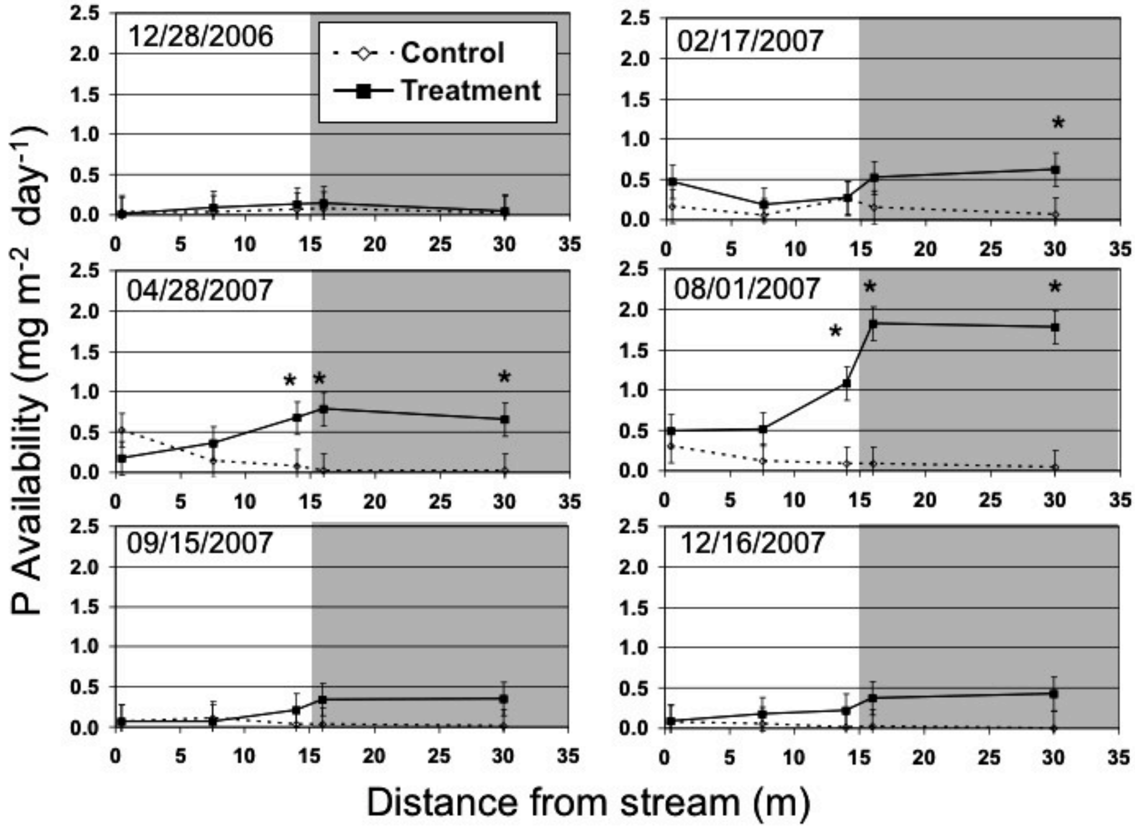

Figure 6: Phosphorus availability from ion exchange membranes. The shaded region indicates area outside SMZ. Significant difference ($p < 0.05$) between control and treatment is indicated with *.

Nutrient Concentration in Soil Solution

The tension lysimeters failed to collect samples during the summer and fall following application because soils were too dry. On dates when samples were collected, over 40% of the lysimeter samples were below the detection limit for all nutrients analyzed.

Ammonium Concentration in Soil Solution

Ammonium showed three significant differences between treatment and control during the sampling period (Table 6). When significant differences were detected, ammonium concentrations in soil solutions remained below 1.1 mg L^{-1} (Figure 7). Some sampling periods, such as early June, had higher concentrations in lysimeters samples on both the treatment and control sides of the streams. For the estimates that were significant, the June and January 2008 samples had one sample missing due to lysimeter failure with one of the remaining measures unusually high. The third significant estimate included the highest sample concentration while the other two samples for that treatment, location, and date were below the detection limit.

Table 6: *p*-values for test of difference between control and biosolids treatment at each sampling location at each date for ammonium in lysimeters. Values significant to alpha 0.05 are bolded.

Date	Distance from stream (m)				
	0	7.5	14	16	30
12/29/06	0.8876	**0.0001**	0.8150	0.9298	0.9277
01/21/07	0.8112	0.0679	0.8651	0.8347	0.6582
02/03/07	1.0000	0.9182	0.9053	0.8914	0.6941
02/17/07	0.9980	0.9537	1.0000	1.0000	0.3738
03/05/07	1.0000	0.8630	1.0000	1.0000	0.7250
03/31/07	1.0000	0.9269	1.0000	1.0000	0.8719
04/28/07	1.0000	0.9414	1.0000	0.9728	0.6398
05/14/07	0.9972	0.6390	0.0814	0.2538	0.3069
06/01/07	0.4566	0.2111	**0.0432**	0.6276	0.2614
07/02/07	N/A	N/A	N/A	N/A	N/A
07/18/07	N/A	N/A	N/A	N/A	N/A
08/01/07	N/A	N/A	N/A	N/A	N/A
08/14/07	N/A	N/A	N/A	N/A	N/A
08/30/07	N/A	N/A	N/A	N/A	N/A
09/15/07	N/A	N/A	N/A	N/A	N/A
10/06/07	N/A	N/A	N/A	N/A	N/A
12/16/07	N/A	N/A	N/A	N/A	N/A
01/08/08	0.9269	0.9465	0.6614	0.8892	**0.0017**

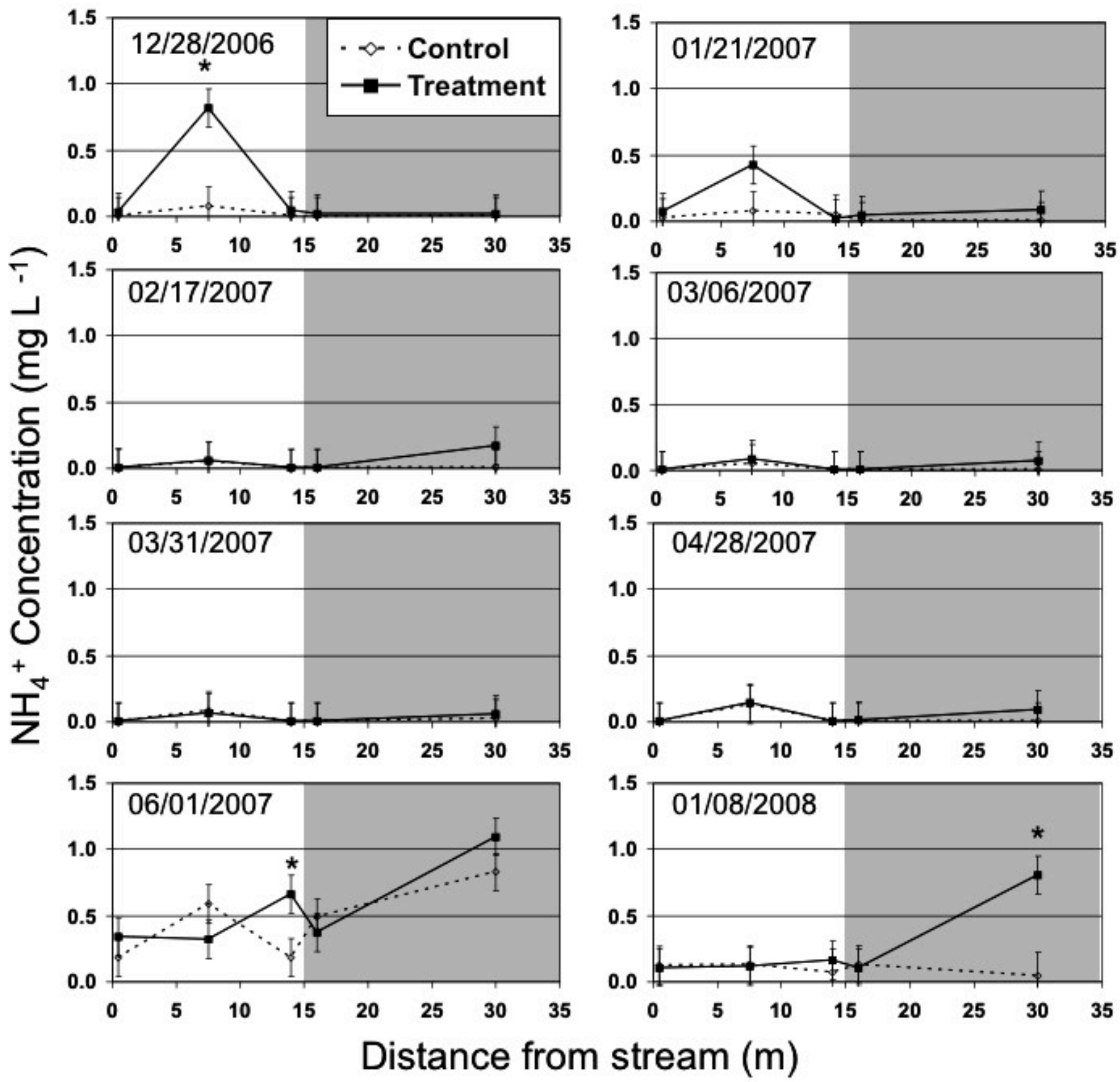

Figure 7: Ammonium concentration in soil solution from lysimeters. The shaded region indicates area outside SMZ. Significant difference ($p < 0.05$) between control and treatment is indicated with *.

<u>Nitrate Concentration in Soil Solution</u>

Nitrate concentrations in soil solution on the treated side of the stream were higher than the control side of the stream at 30 m on three occasions (Table 7). Nitrate concentrations in soil solution remained below 1.5 mg L^{-1} (Figure 8). Elevated nitrate concentrations were first observed on the treatment side of the stream at the same time as elevated nitrate availability in IEMs was first observed. The concentration of the leachate under the biosolids application remains low, despite being elevated compared to leachate on the control side of the stream.

Table 7: *p*-values for test of difference between control and biosolids treatment at each sampling location at each date for nitrate in lysimeters. Values significant to alpha 0.05 are bolded.

Date	Distance from stream (m)				
	0	**7.5**	**14**	**16**	**30**
12/29/06	0.7099	0.9028	0.7651	0.5358	0.9620
01/21/07	0.6086	0.9011	0.4852	0.6434	1.0000
02/03/07	0.5546	0.8414	0.4962	0.6553	0.5773
02/17/07	0.4400	0.9629	0.6456	0.8522	0.6991
03/05/07	0.4249	0.8881	0.6427	0.8664	0.2429
03/31/07	0.4249	0.9108	0.6610	0.9037	0.0906
04/28/07	0.4249	0.9251	0.6348	0.8944	**0.0022**
05/14/07	0.3790	0.7709	0.8564	0.7872	**0.0004**
06/01/07	0.2740	0.4897	0.5414	0.3467	0.2194
07/02/07	N/A	N/A	N/A	N/A	N/A
07/18/07	N/A	N/A	N/A	N/A	N/A
08/01/07	N/A	N/A	N/A	N/A	N/A
08/14/07	N/A	N/A	N/A	N/A	N/A
08/30/07	N/A	N/A	N/A	N/A	N/A
09/15/07	N/A	N/A	N/A	N/A	N/A
10/06/07	N/A	N/A	N/A	N/A	N/A
12/16/07	N/A	N/A	N/A	N/A	N/A
01/08/08	0.4249	0.4759	0.4409	0.0503	**0.0048**

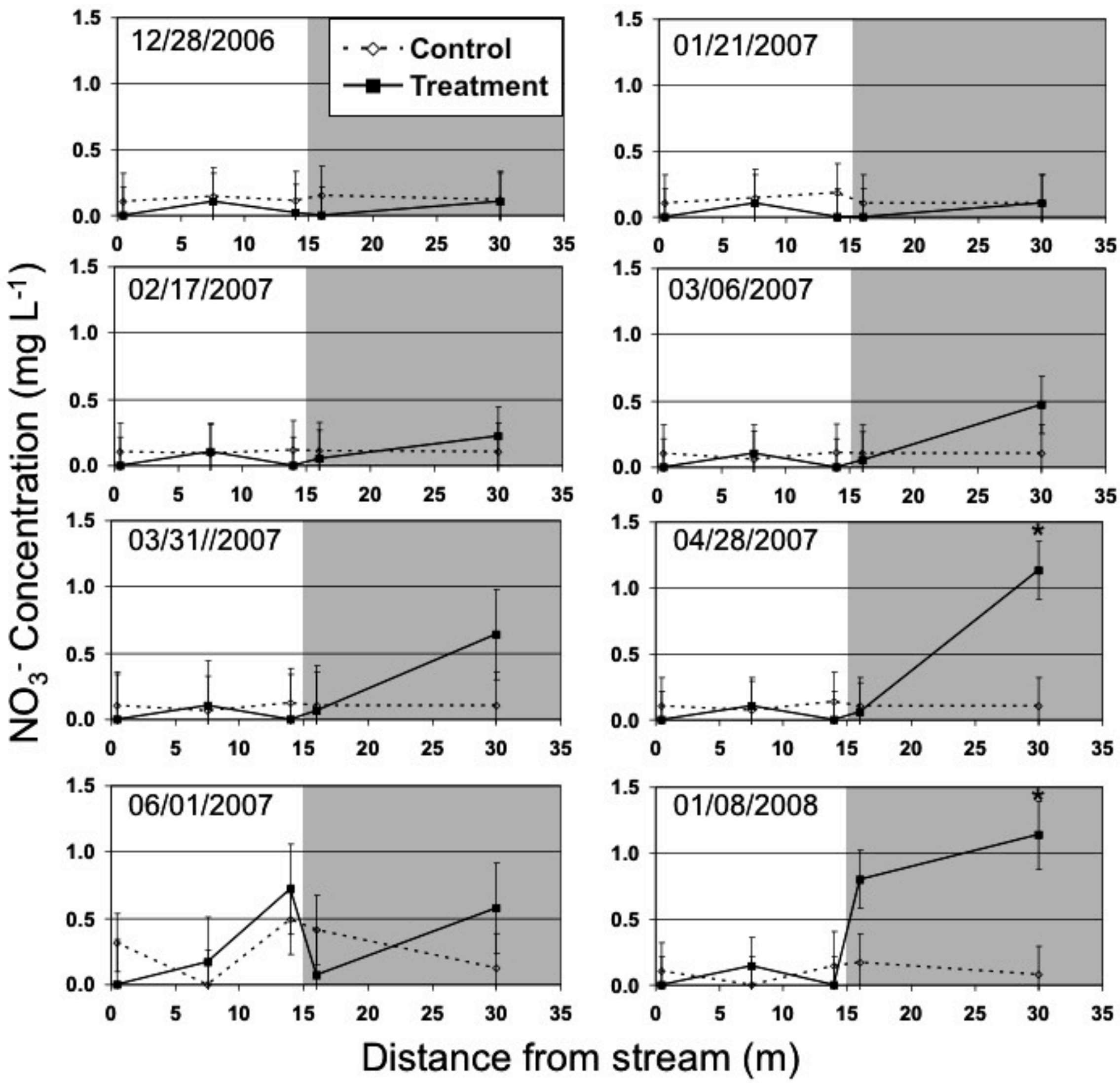

Figure 8: Nitrate concentration in soil solution from lysimeters. The shaded region indicates area outside SMZ. Significant difference ($p < 0.05$) between control and treatment is indicated with *.

Phosphorus Concentration in Soil Solution

Phosphorus concentrations in soil solution were significantly different between treatment and control on three occasions (Table 8). The control estimate for the June sampling date at 14 m was significantly higher than the treatment. When elevated concentrations on the treatment side were detected, concentrations remained below 0.8 mg L^{-1} (Figure 9). Phosphorus concentrations at most sampling periods were very low with most samples below the detection limit.

Table 8: *p*-values for test of difference between control and biosolids treatment at each sampling location at each date for phosphorus in lysimeters. Values significant to alpha 0.05 are bolded.

Date	Distance from stream (m)				
	0	7.5	14	16	30
12/29/06	1.0000	**0.0424**	1.0000	0.7293	0.9886
01/21/07	0.9967	0.8178	1.0000	1.0000	1.0000
02/03/07	1.0000	0.9677	0.9937	0.9783	1.0000
02/17/07	1.0000	0.9580	1.0000	1.0000	0.8181
03/05/07	0.9301	0.8780	1.0000	0.8954	1.0000
03/31/07	0.9708	0.9723	0.9209	1.0000	0.7374
04/28/07	1.0000	0.7998	0.9642	1.0000	1.0000
05/14/07	0.3950	0.4521	0.7235	0.5493	0.5298
06/01/07	0.8354	0.2201	**0.0051**	**<.0001**	0.8942
07/02/07	N/A	N/A	N/A	N/A	N/A
07/18/07	N/A	N/A	N/A	N/A	N/A
08/01/07	N/A	N/A	N/A	N/A	N/A
08/14/07	N/A	N/A	N/A	N/A	N/A
08/30/07	N/A	N/A	N/A	N/A	N/A
09/15/07	N/A	N/A	N/A	N/A	N/A
10/06/07	N/A	N/A	N/A	N/A	N/A
12/16/07	N/A	N/A	N/A	N/A	N/A
01/08/08	0.8729	0.9135	0.6441	0.9826	0.9435

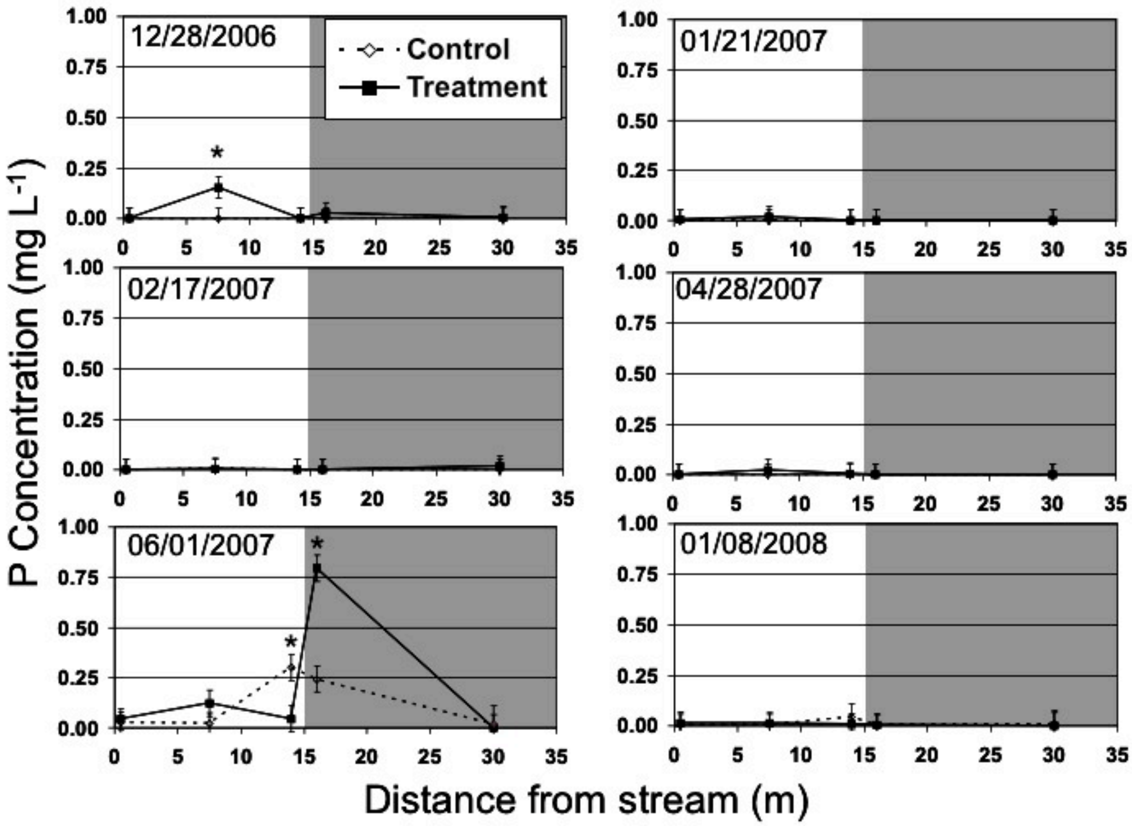

Figure 9: Phosphorus concentration in soil solution from lysimeters. The shaded region indicates area outside SMZ. Significant difference ($p < 0.05$) between control and treatment is indicated with *.

Nutrient Concentration in Overland Flow

We installed the overland flow canisters in summer of 2007, six months after biosolids application. Consequently, we only collected 4 overland flow samples, greatly reducing the number of samples collected. Therefore, we are not able to draw conclusions about SMZs' ability to attenuate nutrients immediately following biosolids application, but can address how they function during the summer, fall and winter following biosolids application. Overland flow samples were collected on four occasions during this period.

<u>Ammonium Concentration in Overland Flow</u>

Ammonium showed no significant differences between treatment and control for all the sampling locations and sampling times (Table 9). Ammonium concentrations in overland flow were below 2.5 mg L^{-1} (Figure 10).

Table 9: *p*-values for test of difference between control and biosolids treatment at each sampling location at each date for ammonium in overland flow. Values significant to alpha 0.05 are bolded.

Date	Distance from stream (m)	
	0	15
08/30/07	0.7814	0.4884
09/15/07	0.4922	0.3005
12/16/07	0.9295	0.7908
01/08/08	0.2690	0.1185

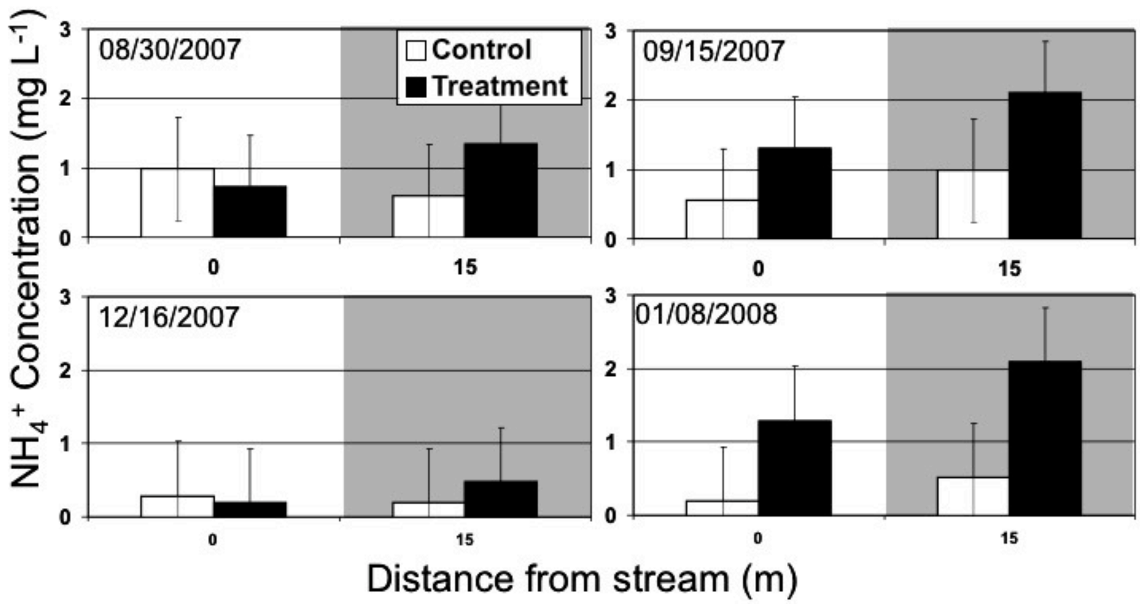

Figure 10: Ammonium concentration in overland flow. The shaded region indicates area outside SMZ. Significant difference (p < 0.05) between control and treatment is indicated with *.

Nitrate Concentration in Overland Flow

Nitrate showed no significant differences between treatment and control for all the sampling locations and sampling times (Table 10). Nitrate concentrations in overland flow were below 2 mg L^{-1} (Figure 11).

Table 10: *p*-values for test of difference between control and biosolids treatment at each sampling location at each date for nitrate in overland flow. Values significant to alpha 0.05 are bolded.

Date	Distance from stream (m)	
	0	15
08/30/07	0.9263	0.4143
09/15/07	0.6615	0.9962
12/16/07	0.6090	0.0592
01/08/08	0.5955	0.6087

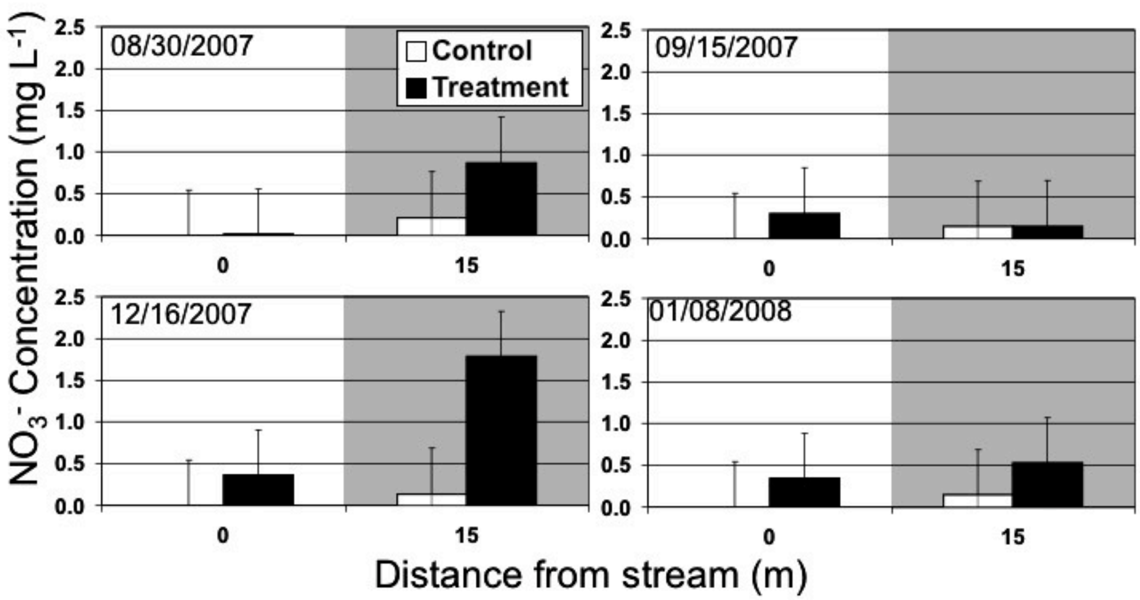

Figure 11: Nitrate concentration in overland flow. The shaded region indicates area outside SMZ. Significant difference ($p < 0.05$) between control and treatment is indicated with *.

Phosphorus Concentration in Overland Flow

Phosphorus concentrations in overland flow were significantly different between treatment and control for the sampling period in mid-December (Table 11). On the date, phosphorus concentrations in overland flow were 4.6 mg L^{-1} (Figure 12). With one exception, phosphorus concentrations were below 1 mg L^{-1} in overland flow samples collected during the other sampling periods.

Table 11: *p*-values for test of difference between control and biosolids treatment at each sampling location at each date for phosphorus in overland flow. Values significant to alpha 0.05 are bolded.

Date	Distance from stream (m)	
	0	15
08/30/07	0.8920	0.9874
09/15/07	0.3368	0.9981
12/16/07	**0.0337**	0.9613
01/08/08	0.8894	0.9507

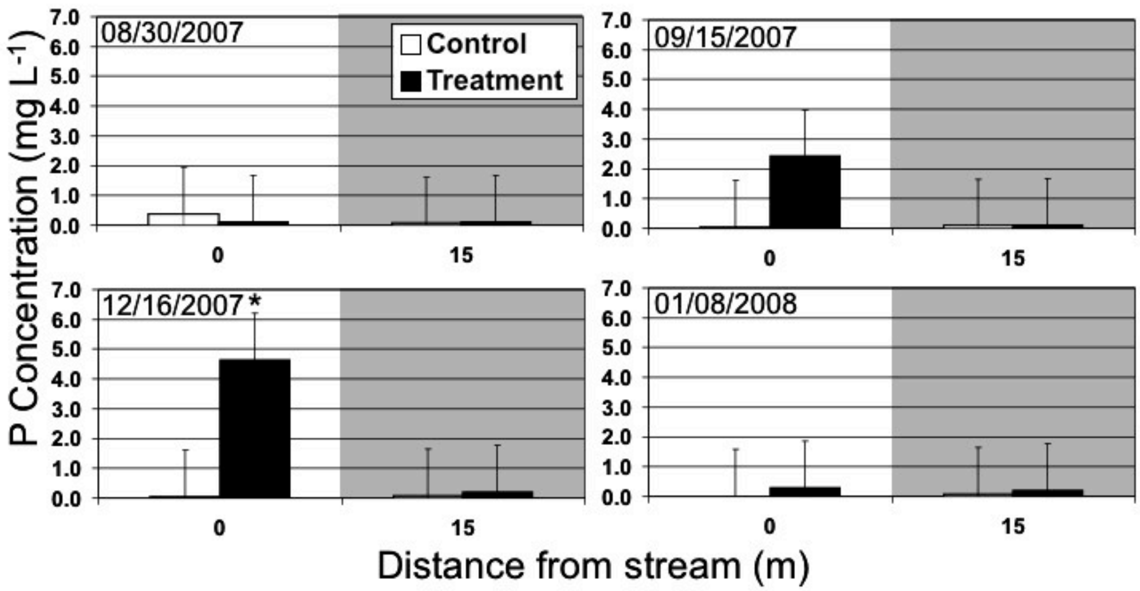

Figure 12: Phosphorus concentration in overland flow. The shaded region indicates area outside SMZ. Significant difference (p < 0.05) between control and treatment is indicated with *.

Nutrient Concentrations in Intermittent and Perennial Streams

Over the course of the study, the intermittent streams showed one significant difference between reference and treated streams for ammonium concentration and two significant differences for nitrate concentrations (Table 12). On the date significantly different ammonium concentrations were observed, concentrations were below 1.5 mg L^{-1} (Figure 12). Ammonium concentrations were below 0.25 mg L^{-1} in stream samples collected during all other sampling periods. Nitrate and phosphorus concentrations were less than 0.15 mg L^{-1} throughout the study period. While intermittent streams showed some elevated nitrogen concentrations, these responses were low and short lived.

Table 12: *p*-values for test of difference between biosolids treatment and reference intermittent streams at each sampling date. Values significant to alpha 0.05 are bolded.

Date	Nutrient		
	NH_4^+	NO_3^-	P
12/28/06	0.4871	0.0808	0.9923
01/20/07	1.0000	0.6426	1.0000
02/03/07	0.9673	0.2069	1.0000
02/17/07	1.0000	**0.0131**	1.0000
03/05/07	1.0000	0.8133	0.9601
03/31/07	0.9980	1.0000	0.9798
04/28/07	1.0000	1.0000	0.9707
05/14/07	0.7295	1.0000	0.0580
06/01/07	**<.0001**	0.2916	0.0819
12/16/07	0.4872	**0.0025**	0.5669
07/02/07	N/A	N/A	N/A
07/18/07	N/A	N/A	N/A
08/01/07	N/A	N/A	N/A
08/14/07	N/A	N/A	N/A
08/30/07	N/A	N/A	N/A
09/15/07	N/A	N/A	N/A
10/06/07	N/A	N/A	N/A
12/16/07	N/A	N/A	N/A
01/08/08	0.8477	1.0000	0.8938

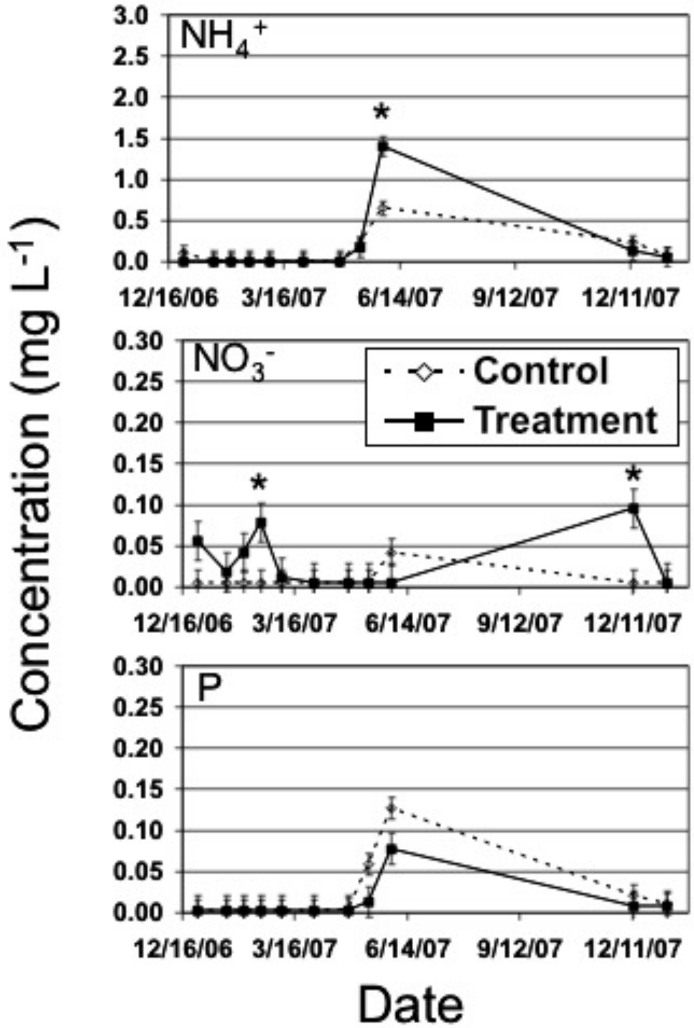

Figure 13: Nutrient concentration in intermittent streams. Significant difference ($p < 0.05$) between control and treatment is indicated with *.

The perennial stream showed higher concentrations of nitrate upstream from the biosolids application compared to downstream concentrations, but no difference for ammonium and phosphorus concentrations were observed. The p-values for paired student's t-tests performed on ammonium, nitrate, and phosphorus were 0.1593, 0.0187, and 0.0753 respectively. The average concentrations for nitrate, ammonium and phosphorus were all below 0.1 mg L^{-1} in the perennial streams (Figure 14).

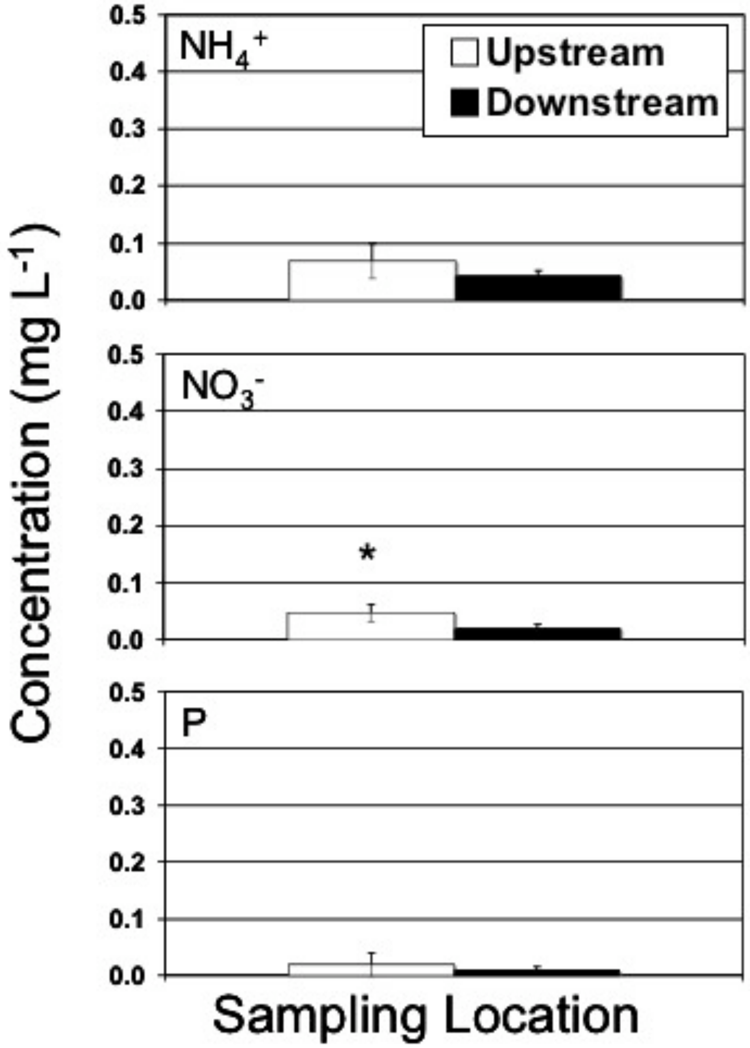

Figure 14: Nutrient concentration in perennial stream. Significant difference ($p < 0.05$) between upstream and downstream is indicated with *.

Discussion

SMZ Effectiveness for Protecting Stream Water Quality

SMZs were effective at protecting stream water from nutrient impairment due to biosolids application to adjacent forests. Intermittent stream water samples had three significant differences between nutrient concentrations in treatment and reference streams. Nitrogen and phosphorus concentrations in the perennial stream were lower downstream from the treatment. When significant differences were present in streams, nitrate, ammonium, and phosphorus concentrations were below 0.095, 1.400 and 0.127 mg L^{-1}, respectively.

Our findings of nutrient concentrations in stream water were similar to other studies in the Piedmont. Lakel et al. (2006) found nitrate and ammonium concentrations of 0.043 and 0.097 mg L^{-1}, respectively, in streams in the Virginia Piedmont with a 15 m SMZ retained adjacent to forest harvest. Our findings have higher concentrations than that study, but are still below regulated levels. With one exception, ammonium concentrations in our study were below 0.653 mg L^{-1}, which is more consistent with Lakel et al. (2006). Franklin et al. (2002) found nitrate, ammonium, and dissolved reactive phosphorus concentrations ranging from 0.07 to 0.11, less than 0.01 to 2.10 and 0.007 to 0.023 mg L^{-1}, respectively. Our findings are similar to those of that study, particularly with regard to the wide range of ammonium concentrations.

Other research supports our findings of the effectiveness of a 15 m SMZ for protecting water quality from nutrient pollution due to nutrient applications to adjacent areas. SMZs provide mechanisms that attenuate both surface and subsurface nutrient movement (Muscutt et al., 1993, Gilliam, 1994, Vought et al., 1994). In a review, Castelle et al. (1994) found in a broad review of buffers that a 15 m SMZ maintains the chemical integrity of streams. Grey and Henry (2002) found a Pacific northwestern forest treated with biosolids outside a 20 m buffer showed that ammonium concentrations in stream water had no differences due to treatment and that elevated levels of nitrate in streams were below drinking water standards. Total phosphorus was elevated above surface water standards, but the authors principally attributed this to stream detritus (Grey and Henry, 2002).

A number of studies have examined the water quality impact of forest fertilization with conventional fertilizers. Hopmans and Bren (2007) found an intensively managed Radiata pine (*Pinus radiata* D. Don) stand exported less than 1 percent of the applied nitrogen and phosphorus to streams with a 30 m SMZ retained. McBroom et al. (2008) observed no change in base flow nutrient concentrations in fertilized forest watersheds with a buffer, though nutrient concentrations in storm flow was elevated for a short period following fertilization. Our sampling scheme does not allow us to evaluate storm flow concentrations. Liechty et al. (2006) found that streams in a fertilized watershed had increased nitrogen concentrations, but found that aerially-applied urea had drifted directly into stream channels.

Subsurface Nitrogen Movement in the SMZ

Given the results for surface soil ion availability, soil solution, and overland flow, we would not expect to observe stream impairment. We observed elevated levels of ammonium, nitrate and phosphorus availability outside the SMZ following biosolids application using ion exchange membranes. Within the SMZ adjacent to the area applied with biosolids the ammonium, nitrate and phosphorus availability consistently declined to control levels at the sampling location 0.5 m from the stream. The decline of nutrient availability across the SMZ suggests that lateral nutrient movement to the stream was limited.

Cation exchange capacity of the biosolids likely moderated the release of ammonium immediately after biosolids application. The high organic matter content, with 59% volatile solids, and an initial pH of 8.1 could have caused a high variable-charge in the biosolids, initially limiting movement of ammonium (Sollins et al., 1988). Biosolids application on all sides of the sampling location 30 m from the stream may have caused immediate elevated ammonium availability, whereas elevated ammonium availability took a month to manifest in the sampling location at the edge of the biosolids applied area where the material was not as concentrated. This spatially dependent lag suggests moderated movement of ammonium from the biosolids across the O horizon into the A horizon.

Once in the soil, ammonium's vertical and lateral movement may have been limited initially by cation exchange capacity of the soils and in spring by plant uptake. Many studies have found that vegetation in forested riparian areas can be a sink for nutrients, especially during the growing season (Lowrance et al., 1984, Peterjohn and Correll, 1984, Simmons et al., 1992, Pinay et al., 1993, Hefting et al., 2005). Plant uptake may have been responsible for the lack of ammonium availability during the summer even in the area applied with biosolids.

Much of the nitrogen in the biosolids applied area in the surface soil was directly released from the biosolids as well as from microbial mineralization of nitrogen in the biosolids. While ammonium availability in the soils outside the SMZ showed an immediate response, nitrate availability showed a later response. Nitrate concentration in the biosolids was 6 mg kg^{-1} compared with 13,000 mg kg^{-1} ammonium and 44,000 mg kg^{-1} organic nitrogen and consequently, initial nitrate availability was low. Average air temperatures below 10°C and cool soil temperatures likely decreased mineralization of organic nitrogen and nitrification of ammonium from December 2006 to February 2007 compared to mineralized during spring and summer (Wang et al., 2003, Wang et al., 2006a, Bagherzadeh et al., 2008).

During March and April, average air temperatures increased to above 10°C, likely increasing microbial mineralization of organic nitrogen in the biosolids, supplying more nitrogen to the soil (Wang et al., 2003, Wang et al., 2006a, Bagherzadeh et al., 2008). The microbial production acts with limited plant uptake to increase nitrogen availability in the soils. April was the only sampling period during which nitrate availability was elevated in ion exchange membranes at the sampling location 7.5 m from the stream. Our initial observation of elevated levels of nitrogen in the soil solution at the Bt and C horizon boundary outside the SMZ occurred in April. As an anion and more mobile in the soil solution than ammonium, nitrate migrated across the SMZ and down through the soil column with high supply from mineralization and limited plant uptake to remove it from the soil solution (Correa et al., 2006).

Plant uptake of nitrate and ammonium could have limited the movement of nitrogen following April. Ammonium availability returned to background levels, while nitrate levels were elevated at the inside edge of the SMZ and outside the SMZ. Some studies have found that denitrification is the dominant mechanism over plant uptake for removing nitrate from soil solution in some riparian environments (Jacobs and Gilliam, 1985, Verchot et al., 1997a). Denitrification on this site may have been limited during the summer due to low amounts of rainfall, leaving soils unsaturated adjacent to the streams and conditions unfavorable for high rates of denitrification (Davidson and Swank, 1986, Schnabel et al., 1996, Hill et al., 2000). Nitrate may have remained elevated outside the SMZ in soil solution at the Bt and C horizon boundary because soil saturation and carbon can limit denitrification at depth (Obenhuber and Lowrance, 1991, Hill et al., 2000). As temperatures declined and nitrogen mineralization slowed, nitrate availability in the ion exchange membranes declined to near control levels outside the SMZ in soils under the biosolids. Though still elevated at 16 m from the stream and nearly significant to the 0.1 p-value at 30 m from the stream, nitrate availability was reduced compared to summer levels.

Nitrate did move vertically, but did not move laterally after it passed through the argillic horizon, as soil solutions collected using tension lysimeters showed no significant differences within the SMZ. McLaren et al. (2003) found nitrate in lysimeters in forest soils treated with biosolids at concentrations higher than control, but concluded that nitrate leaching was unsubstantial. Aschmann et al. (1992) found nitrate in lysimeters in hardwood forests treated with liquid wastewater sludge had concentrations as high as 40 mg L^{-1}. The authors concluded that nitrogen application rates that exceeded uptake capacity of vegetation posed a threat to groundwater pollution (Aschmann et al., 1992). Our results suggest that the application rate used for this study was appropriate for nitrogen uptake in a loblolly pine plantation because we did not observe lysimeter concentrations above 1.5 mg L^{-1}.

Subsurface Phosphorus Movement in the SMZ

We first observed significantly elevated phosphorus availability in the ion exchange membranes outside the SMZ in February. The biosolids released phosphorus to the soil more slowly than ammonium. Iron, which made up 4.6% of the biosolids, could have formed complexes with phosphorus limiting its availability (Maguire et al., 2000, Maguire et al., 2001, Elliott et al., 2002, O'Connor et al., 2004).

Phosphorus availability was lower than nitrate or ammonium availability. While estimates of elevated levels of nitrate and ammonium in the biosolids treated area were regularly above 4 mg m^{-2} day^{-1} and 10 mg m^{-2} day^{-1}, respectively, estimated levels of phosphorus availability were never above 2 mg m^{-2} day^{-1}. Constituents of the biosolids as well as iron and aluminum in the finely textured upland soil where the biosolids were applied likely sorbed much of the phosphorus, limiting its availability and its lateral and vertical movement in the soil (Maguire et al., 2000, Maguire et al., 2001, Elliott et al., 2002, Sukkariyah et al., 2007).

Phosphorus availability was significantly elevated at the inside and outside edge of the SMZ during spring and remained elevated throughout the summer. Phosphorus can be mobile in soil solution in sandy surface soils with low phosphorus sorption capacity, especially during the spring when microbial activity is high and plant uptake is low (Lu and O'Connor, 2001, Alleoni et al., 2008). Despite this imbalance, the mobility of phosphorus is low compared to nitrate due to its formation of complexes with aluminum and iron hydroxides, clay minerals, and organic matter in the soil (Mitsch and Gosselink, 2000, Mengel and Kirkby, 2001). Whereas nitrate availability was elevated across much of the SMZ during April, phosphorus availability was limited to sampling locations within or directly adjacent to the biosolids applied area. During the rest of the spring and summer, phosphorus availability mimics the spatial pattern of nitrate availability: high in the biosolids applied area, lower but still significantly elevated at 14 m from the stream, and returning to non-significantly different levels 7.5 m from the stream. Lowrance et al. (1984), Peterjohn and Correll (1984), and Kelly et al. (2007) observed that plant uptake can be a mechanism for reducing phosphorus across a buffer.

Winter availability of nutrients suggests that there will be some lesser but continued nutrient increases to the plantation outside the SMZ in the second spring and summer following biosolids application. Beginning in fall and continuing through winter, nutrient availability declined to non-significant levels. In December, phosphorus availability mimicked nitrate availability, being nearly significant at the 0.1 p-value at 16 m from the stream. Cogger et al. (2004) found that over 50 percent of the available nitrogen in the soil the second year after biosolids treatment became available during winter. However, given the non-significant nutrient availability in the SMZ, these elevated nutrient levels likely will not pose a water quality concern.

Nutrient Movement in Overland Flow in the SMZ

Overland flow samples collected showed very few significant differences, but non-statistical patterns emerge in graphical depictions of the data. We observed lower estimates of ammonium and nitrate concentrations near the stream than adjacent to the biosolids, suggesting that SMZs may reduce nitrogen concentration in runoff. A number of studies have shown that SMZs can reduce nitrogen content in runoff through infiltration and sediment attenuation (Daniels and Gilliam, 1996, Verchot et al., 1997b, Lowrance and Sheridan, 2005).

Phosphorus shows the reverse pattern with higher concentrations near the stream than in the area adjacent to the biosolids treatment, suggesting that the SMZ is concentrating phosphorus. The significant estimate did include one sample concentration that was over 3 times the next highest sample concentration, which was from a different sampling location and date. The estimates for phosphorus concentration in overland flow adjacent to the biosolids were not significantly elevated. We were unable to determine the source of the phosphorus increase adjacent to the stream, though it seems likely to be biosolids. Other studies have found increased phosphorous concentrations in runoff from soils treated with biosolids (Penn and Sims, 2002, Elliott et al., 2005).We observed no elevated phosphorus concentrations in overland flow samples taken adjacent to the biosolids area, but instead found elevated concentrations adjacent to streams. Unlike Lowrance and Sheridan (2005) who worked in a coastal plain agricultural system using a three zoned buffer, we did not sample runoff in the middle of the buffer so cannot determine as they did that phosphorus decreased in the middle of buffer before increasing adjacent to the stream. We did not measure surface runoff volume across the buffer, so cannot determine if surface runoff increased across the buffer.

If the source of the phosphorus is the biosolids, other studies have found that concentrated flow paths can limit SMZ effectiveness for attenuating nutrients in overland flow. The SMZ had erosional gullies and skidder ruts from a previous harvest that could have served as channels to concentrate runoff. Dickey and Vanderholm (1981) and Dillaha (1989) found that concentrated flow paths through vegetated filter strips reduced the effectiveness of vegetated filter strips for protecting water quality from nutrients by decreasing infiltration and increasing the width required to attenuate sediment. Rivenbark and Jackson (2004) found that 25 percent of the observations during which SMZs failed to attenuate sediment following a harvest where in drainages with either roads or skidder trails and another 50 percent were in drainages with convergent swales, a designation that included erosional gullies.

Conclusion

We monitored 15 m SMZs adjacent to loblolly pine stands treated with anaerobically digested biosolids at an operational rate. We examined how inorganic nitrogen and phosphorus varied across time and space in the SMZ. We sampled from adjacent streams to corroborate the results in the SMZ.

Water quality showed no significant declines over the period sampled when SMZs were installed adjacent to streams in loblolly pine plantations on a typical Piedmont soil where anaerobically digested biosolids were applied. Surface soil nutrient availability measured using ion exchange membranes showed that elevated ammonium, nitrate, and phosphorus levels outside the SMZ were reduced to control levels near streams. The availability index returned to near background levels in the treated areas 1 year after application. In the subsurface soil solution measured using tension lysimeters, over 40% of sample concentrations were below the detection limit. All ammonium and phosphorus concentration estimates were below 1.0 mg L^{-1}. Nitrate concentrations estimates outside the SMZ were below 1.5 mg L^{-1} and showed no significant difference between treatment and control inside the SMZ. Overland flow samples showed no significant difference between control and treatment for ammonium and nitrate with concentrations below 2.5 mg L^{-1} and phosphorus concentrations above that level for one sampling date. Ammonium, nitrate, and phosphorus concentrations were below 0.5 mg L^{-1} in intermittent streams for all but one sampling period and were not significantly elevated in the perennial stream downstream of the site.

Acknowledgements
The Metropolitan Washington Council of Governments, the Sustainable Engineered Materials Institute, and the Forest Nutrition Cooperative provided funding for this project. Synagro Technologies, Inc. provided in kind assistance.

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