NITROGEN, PHOSPHORUS AND CARBON DYNAMICS DURING STORMS IN A GLACIATED THIRD-ORDER WATERSHED IN THE US MIDWEST

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Submitted to the faculty of the University Graduate School
in partial fulfillment of the requirements
for the degree
Master of Science
in the Department of Earth Sciences,
Indiana University

December 2011

Accepted by the Faculty of Indiana University in partial fulfillment of the requirements for the degree of Master of Science

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Acknowledgements

First and foremost, I would like to thank my parents and grandparents for instilling in me and my two sisters an appreciation of the importance of environmental preservation. Having parents who have advocated environmental protection for as long as mine, definitely influenced my decision to enter the field of water quality. The support of my friends has also been essential to my success throughout my academic and professional career.

This thesis would not have been completed without the support, suggestions and guidance of Philippe Vidon. As this project evolved, it became necessary to make multiple changes, many of which were suggested by Philippe, Lenore Tedesco and Kathy Licht were also instrumental in this work by providing suggestions, and when the weather didn't cooperate as expected, moral support. Also of note are, Professors Reto Giere, John Graveel, Tim Filley, and Jon Harbor at Purdue University. Working with them helped me discover the fields of hydrology and geochemistry.

Thanks also to the Center for Earth and Environmental Science, especially Bob Hall, this project would never have been successful without his suggestions and assistance in the field. Finally, this project would not have been possible without the funding and laboratory analysis provided by the Central Indiana Water Resources Partnership and Veolia Water, Indianapolis.

ABSTRACT

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The characterization of the nutrients nitrogen, phosphorus and carbon (NPC) export to streams during storms is an integral part of understanding processes affecting water quality. Despite the fact that excessive levels of these nutrients in the Mississippi River basin adversely affects water quality in the Gulf of Mexico, little research has been conducted on NPC dynamics during storms on larger (>20 km^2) agriculturally dominated Midwestern watersheds. This project examined the storm export of nitrate, ammonium, total phosphorus, and dissolved organic carbon (DOC) in the upper Eagle Creek Watershed (UECW) (274 km^2) in Central Indiana, USA. Water samples were collected during five winter and spring storms in 2007 and 2008 on the rising and falling limb of the hydrograph, in order to characterize NPC dynamics during storm events. Stream discharge and precipitation was monitored continuously, and major cations were used to examine changes in source water over the duration of the storm and assist in the determination of potential flowpaths. DOC, total P, and TKN (Total Kjeldahl Nitrogen) tended to peak with discharge, while nitrate usually exhibited a slight lag and peaked on the receding limb. Total phosphorus, NH_3^-, TKN, and DOC appear to be delivered to the stream primarily by overland flow. NO_3^--N appear to be delivered by a combination of tile drain and macropore flow. Overall UECW displayed smoother nutrient export patterns than smaller previously studied watersheds in the area suggesting that scale may influence nutrient export dynamics. Further research is underway on a 3000 km^2

watershed in the area to further examine the role scale may play in nutrient export patterns.

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Introduction

The runoff of the nutrients nitrogen (N), phosphorus (P) and dissolved organic carbon (DOC) has been implicated in the eutrophication of waterways and an overall deterioration of water quality (Dosskey, 2001; Gentry et al., 2007). Nutrient input from agricultural areas in the Midwest has also been linked to the development of the dead zone in the Gulf of Mexico (Goolsby et al., 2001; Royer et al., 2006; Alexander et al., 2009). Between 1961 and 1997, N application by humans doubled, and approximately one third of surface applied N ends up in water reserves, primarily surface water bodies (Poor and McDonnell, 2008). Excessive levels of these nutrients have also been linked to algal blooms in drinking water reservoirs, some of which produce toxins making the water harmful for human consumption (Robertson et al., 2008). Precipitation events often result in large amounts of N, P, and DOC (NPC) export from an area over a brief time period (Dalzell et al., 2005; Hood et al., 2006; Inamdar et al., 2006). Studies in agricultural areas in Illinois have found that up to 56 and 84 percent of the annual export of nitrate (NO_3^--N) and dissolved reactive phosphorus (DRP) occur during periods of high discharge respectively. Particulate P which contributes heavily to total P loading is transported almost exclusively by overland flow during storm events (Royer et al., 2006). Dissolved organic carbon (DOC) exports have been shown to be between 33 (Hook et al., 2005) and 90 (Dalzell et al., 2005) times higher during storm events than during baseflow. Research has indicated that storm export of nutrients has the potential to adversely impact water quality in the Midwest (Royer et al., 2006).

Many of the existing studies performed on nutrient export have focused on small watersheds < 20 km^2 (Hill et al., 1999; Hook et al., 2005; Wagner et al., 2008), and were often performed in mountainous regions (Christopher et al., 2008; Inamdar et al., 2006). Despite the fact that the Midwest contributes heavily to the nutrient loading in the Mississippi River, the area is underrepresented with respect to nutrient export studies (Royer et al., 2006). When properly managed, nutrient losses from watersheds can be minimized (Edwards et al., 1996); however in order to have maximal effectiveness, these best management practices (BMP's) require knowledge of the flowpaths, delivery patterns, and the sources of exported nutrients during high flow events (Wagner et al., 2008).

This study on the Upper Eagle Creek Watershed (UECW) examines NPC exports at a scale that is practical from a policymaking perspective and representative of many agriculturally dominated watersheds in the US Midwest. Larger watersheds are more complex than smaller watersheds in that they receive water from multiple tributaries, and often do not receive uniform precipitation throughout their drainage area due to the heterogeneous precipitation patterns of storms.

The UECW exhibits a hydrological response which is more sluggish than the watershed studied by Wagner et al. in 2006 (< 20 km^2), but more flashy than larger watersheds such as the Upper Whiter River Watershed (3000 km^2). Smaller first-order watersheds which, lack tributaries and receive more homogeneous distributions of precipitation, typically exhibit a flashy hydrograph, with peak discharge occurring rapidly, and then quickly returning to pre-event conditions (Dunne, 1978). With watershed management decisions often made at the county level, it is important to examine watersheds of this scale so larger watersheds is important for policymakers and water quality experts interested in developing best management practices (BMP's) to mitigate the nutrient pollution problem. The acquisition of high temporal resolution data from large watersheds will also assist in the development of accurate, yet less labor intensive lower temporal resolution sampling regimes. Vidon et al. (2008) examined the accuracy of three different sampling strategies and found that approximately 90 percent of the variation in loading estimates can be attributed to the variation in solute concentration occurring over the duration of a storm. The development of less labor intensive and costly sampling regimens will also reduce the cost associated with post construction BMP monitoring and assessment.

This study sampled 5 storms occurring during the late fall, winter, and spring on the UEWC and examined the NPC export patterns. The UECW is an agricultural watershed, and is representative of many agro-ecosystems in the US Midwest. The objectives of the study were to:

1. Determine if the concentration patterns of NPC export were consistent across storms and season, and if they are not what the possible causes of these variations were.
2. Use cation data to examine flowpaths and potential sources of these nutrients over the duration of the event.
3. Compare the nutrient export patterns exhibited by the 3rd order UECW to those observed on smaller order streams and determine what effect if any scale has on nutrient export.

The UECW receives water from multiple tributaries under varying types of land use, therefore, it was hypothesized that this watershed would exhibit a less flashy nutrient signal than smaller first order watersheds. Rather than sharply defined NPC peaks, ECW was expected to have blurred nutrient response with NPC all peaking around the time of maximum discharge.

Background

The export mechanisms and sources of (N) during storms are not well understood. Nitrogen in water exists in two forms; organic and inorganic. Inorganic nitrogen species in water include nitrate (NO_3^--N), nitrite (NO_2^--N), and ammonium (NH_4^+). These species are the most susceptible to biologic uptake (Schlesinger, 1997), and are considered pollutants when present in high concentrations (Royer et al., 2006). Studies in agricultural areas in Illinois have found that up to 56 percent of the annual export of nitrate (NO_3^--N) occurs during periods of extreme discharge (>90th percentile) (Royer et al., 2006). In forested New York watersheds, some studies indicate the main streamwater nitrate source is till groundwater displaced by infiltrating precipitation (Inamdar et al., 2004). Other studies have shown that biogeochemical interactions between the organic soil horizon, and the subsurface water runoff are important regulators of NO_3^- export (Hill et al., 1999). Experiments in mountainous areas in New York have indicated seeps and steep slope gradients as the primary mechanism behind NO_3^--N generation and delivery, with saturated valley bottom riparian areas serving as a secondary nitrate source by delivering intercepted throughfall (water falling through a vegetative canopy) to the stream (Inamdar et al., 2006). In Midwestern watersheds, fertilizers are the main source of NO_3^--N (Royer et al., 2006), and tile drainage is thought to be an important delivery mechanism (Kladivko et al., 2004; Stone and Wilson, 2006).

Total Kjeldahl N (TKN), often referred to as organic nitrogen, is the sum of organic nitrogen and ammonia in a water sample**.** Organic N export does not receive the attention that inorganic N does, however in some ecosystems such as a forested mountainous watershed in New York, it has been shown to contribute heavily to N loading (Inamdar and Mitchell, 2007). In a hilly pasture in New Zealand, Cooke and Cooper (1988) determined that most of the organic N present in surface runoff came from eroded soil material. Inamdar and Mitchell (2007) found leaf litter and throughfall delivered rainwater to be the primary source of DON (Dissolved Organic Nitrate) in streamwater in a forested New York watershed.

Phosphorus is considered a limiting nutrient to plant growth. Phosphorus exists in both organic and inorganic forms, inorganic P, also referred to as SRP (soluble reactive phosphorus) ortho-P, and particulate P. Soluble reactive phosphorus is the species most readily uptaken by plants (Schlesinger, 1997), and is often present in very low quantities. Particulate P is phosphorus which is bound to organics or soil minerals. Decomposition of organic matter can result in particulate P being converted to soluble P (Minnesota Pollution Control Agency Publication, 2007). Total P is the sum of organic and inorganic P. Phosphorus is heavily exported during storms, with studies finding up to 84 percent of SRP export occurring over a one month period, with export being especially high in tiled fields (Royer et al., 2006). Phosphorus export from an area is quite variable, and appears to be dependent upon soil moisture conditions, as it is much more subject to overland flow export than NO_3^- (Royer et al., 2006). Particulate P is especially subject to overland flow export, and has been shown to contribute heavily to Total P loading in agricultural watersheds, since SRP is usually present in very low quantities (Royer et al., 2006, Gentry et al., 2007). A 2008 study by Alexander et al. indicated that P may have a larger effect on the development of the Dead Zone in the Gulf of Mexico than previously thought.

Dissolved organic carbon (DOC) controls ecological and biological processes in soil (Weishaar, 2003), and can influence processes such as denitrification (Schlesinger, 1997). Heavy metals and other contaminants can bind to organic carbon which facilitates their downstream transport (Guggenberger et al., 1994), and is an important regulator of heterotrophic productivity in streams (Dalzell et al., 2005). DOC export is associated with both shallow hydrological flowpaths (Inamdar et al., 2004; 2006), and with overland flow transport (Hood et al., 2006). Calcium and Mg^{2+} in water are generally associated with the weathering of soil, as a result groundwater is generally more enriched in these cations than precipitation and throughfall (Inamdar et al.; 2004; Hood et al., 2006). High levels of Mg^{2+}, Ca^{2+}, Na^{+}, and Cl^{-} in streamwater are often indicative of a shallow groundwater source (Katsuyama, 2001). Potassium is usually associated with throughfall and precipitation, and is delivered to streams primarily by overland flow (Hood et al., 2006).

Traditionally, ^{18}O isotopes have been used to perform hydrograph separations to determine the relative contribution of precipitation and pre-event groundwater to streamflow. This technique assumes that the precipitation and shallow groundwater in the watershed have unique ^{18}O signatures, and that these signatures are consistent throughout the watershed. This assumption has been verified on smaller watersheds ($< 20\ km^2$) (Uhlenbrook and Hoeg, 2003). The large size of this watershed did not allow the cations, Cl^{-}, or O^{18} to be considered conservative tracers. Therefore a hydrograph separation was not performed, however the cations and Cl^{-} were used to qualitatively examine the export patterns of the watershed and assist in flowpath determination.

Watershed factors affecting nutrient export

Many factors can influence the mechanisms by which water and nutrients are exported from a watershed. Topography, soil moisture conditions, and watershed scale are all factors which can influence streamflow and nutrient export. Sidle et al., 1999 examined hydrological flowpaths and streamflow generation processes in four small zero and 1st order basins in a steep forested area in Japan, using tracers and soil moisture measurements. Results indicated that as antecedent soil moisture increased, the ratio of water contributed by overland flow decreased with respect to that contributed by subsurface flow. This was attributed to the enhanced development of the existing macropore network as the soil moisture increased. The effect that increasing catchment size had on the mean residence time (MRT) of streamwater was examined by McGlynn, et al., in 2003 in four watersheds in New Zealand. The study found little correlation between catchment size and MRT, indicating instead that landscape patterns such as topography are the likely control on MRT.

In addition to studies of the purely hydrological variety, many studies on nutrient export dynamics have been conducted on smaller watersheds (Hood, 2006; Inamdar et al., 2004; Christopher et al., 2008). A summer storm examined by Inamdar et al. in 2004, in a mountainous watershed in New York found that NO_3^--N concentrations peaked on the rising limb, and DOC patterns peaked after discharge. A study by Christopher et al., (2008) in the same watershed determined soil moisture to be an important factor in influencing the timing of the NO_3^--N peak. During storms occurring when soil moisture conditions were high, NO_3^--N concentrations peaked before discharge, however under drier soil conditions NO_3^--N concentrations peaked with discharge. A 2006 study by

Wagner et al., in a central Indiana watershed (< 20 km^2), found NO_3^--N export patterns to be quite noisy and lacking a distinctive export pattern. DOC export patterns have also been found to be quite variable. The same study on two smaller watersheds (< 20 km$^{2)}$ in central Indiana have found DOC peaks around the time of maximum discharge (Wagner et al., 2008). However, a study in a New York watershed (< 5 km^2) found that DOC peaks on the receding limb of the hydrograph (Inamdar et al., 2004).

Materials and methods

Site description

The UECW (274 km^2) is part of the larger Eagle Creek Watershed (ECW) (420 km^2) in central Indiana, USA (Figure 1). The surface water from the ECW flows into the Eagle Creek Reservoir, which serves as a major drinking water source for the city of Indianapolis. Central Indiana has a temperate climate with a mean annual temperature of 11.7^{o} C. January is the coldest month with a mean temperature of 3.0^{o} C, and July is the warmest with a mean temperature of 23.7^{o} C. The 1971-2000 average annual precipitation in central Indiana is 105 cm (NOAA, 2005). Highest discharge on ECW typically occurs during March and is the lowest in September (Clark, 1980).

The area is under mixed land use (Table 1), approximately 52 percent agriculture (corn/soybean rotation), 29.9 percent herbaceous, 9.3 percent forested, and 4.3 percent high and low density development (Hack et al., 2008), and is seeing increased levels of development. The upper reaches of the UECW are characterized by extremely flat topography and are dominated by agricultural land use, primarily corn and soybean production with some animal confinement operations present (Tedesco et al., 2005). The sampling site was located in Zionsville, a suburb approximately 20 km north of Indianapolis in Boone County, Indiana (Figure 1).

The primary crops grown in the watershed are corn and soybean, with approximately equal amounts of each planted and rotated seasonally. Corn requires more fertilizer than soybeans, approximately 90 and 76 percent of the nitrogen and phosphorus fertilizers applied in the watershed are applied to corn (Tedesco et al., 2005). There are generally two fertilizer applications each season. The first typically occurs 1-2 weeks prior to planting (Tedesco et al., 2005). The second application is generally applied after the corn reaches approximately 1 foot in height, and the amount applied is generally greater. Some farmers in the watershed elect to apply nitrogen following harvest, this is especially common when winter wheat is going to be grown (Tedesco et al., 2005). Potassium fertilizer is applied primarily in the form of potash. Average annual potash application in the Eagle Creek watershed is approximately 110 and 125 kilograms per hectare per year for soybeans and corn respectively (Tedesco et al., 2005).

This watershed lies within the Tipton till plain, the Tipton till was deposited during the Wisconsin Glaciation, and displays large variations in thickness (West, 1995). The soils contained within the Tipton till plain range from moderately poorly drained to very poorly drained. The tills contain thin loess layers and alluvium deposited since the last glaciation. Till plain soils are excellent for farming, provided that they are artificially drained (Hall, 1999). The headwaters of the UECW are dominated by soils from the Crosby-Treaty-Miami association. This soil is thick, poorly drained, and nearly level. The downstream area is also dominated by the Miami-Crosby-Treaty association. These soils are well to somewhat poorly drained, and exhibit more of a variation in slope ranging from 0-18 percent for the Miami (Tedesco et al., 2005).

Sample collection

For this study five storms were sampled, storm 1, storm 2, and storms 3-A, 3-B, and 3-C. Storms 3-A, 3-B, and 3-C consisted of three separate rain events occurring over a 5 day period during which discharge did not return to pre-event levels. Nine-17 samples were taken for each storm, with a higher sampling frequency occurring on the rising limb of the hydrograph. The sampling interval was determined by the intensity and duration of the storm, and ranged between 5 and 12 hours. The dates and number of samples taken for each storm are summarized in Table 2. Stream level and discharge were measured at 15 minute intervals by USGS gaging station 03353200 located at the sampling site with data available at (www.waterdata.usgs.gov). Hourly and daily precipitation totals were taken from a weather station at the Eagle Creek Airpark (KEYE), approximately 11 km southwest of the sampling site, and obtained from the Indiana State Climate Office at Purdue University with data available at (www.agry.purdue.edu/climate/).

Water samples were taken using an automated water sampler (ISCO 6712). The ISCO was situated to take samples from the middle of the stream, with the sampler intake just above the stream bed. Grab samples were taken prior to each storm to obtain pre-event flow conditions. Samples analyzed for DOC, total P, and TKN were taken in ISCO ProPak bottles with 1 L LDPE liners containing 2 mL of 11 N sulfuric acid. Samples analyzed for magnesium, potassium, sodium, nitrate, nitrite, sulfate, ammonium and chloride were taken in non-acidified 1 L LDPE lined ISCO bottles.

Sample processing

Samples were processed utilizing the methods outlined in Figure 2, within 48 hours of collection. Samples analyzed for DOC, NO_3^--N, NO_2^-, Ortho-P, NH_4^+, and Cl^- were filtered through 0.7 micron glass fiber filters. Samples analyzed for total P, TKN, TOC, sulfate, and the cations were not filtered prior to analysis. All samples were refrigerated between processing and analysis. DOC samples for storms 3-A, 3-B, and 3-C were filtered and frozen within 48 hours of collection, and thawed prior to analysis.

Sample analysis

The analytical methods used for sample analysis and the detection limits of the instrument are summarized in Table 3. NO_3^--N, NO_2^--N, Mg^{2+}, Na^+, NH_4^+, Cl^-, SO_4^-, TOC, and DOC analyses were performed by Veolia Water, Indianapolis. Total P and TKN analyses were performed by ESG Laboratories in Indianapolis, IN.

Data analysis

For each storm, time to peak, runoff ratios, mean stormflow, peak discharge, baseflow discharge, bulk precipitation, and precipitation intensity and 7, 30, and 90 day antecedent discharge and precipitation were calculated. Antecedent precipitation was defined as the amount of precipitation falling during the 7, 30, and 90 days preceding the storm event, and was calculated using daily precipitation data. Precipitation intensity was determined using hourly precipitation data, and was defined as the amount of precipitation falling hourly for the duration of each storm event. Antecedent discharge was calculated by determining the volume of water in cubic meters discharged by the watershed in the 7, 30, and 90 days preceding each event and dividing by the area of the watershed. Time to peak was the time period from which there was a noticeable increase in discharge to the peak in discharge, using 15 minute USGS discharge data. Mean stormflow discharge, peak discharge, and baseflow were calculated using 15 minute discharge data. The runoff ratio was calculated by dividing the volume of water discharged by the watershed over the duration of each flow event, by the volume of water delivered by precipitation to the watershed for each storm event.

Double mass curves were created by plotting the cumulative daily precipitation against cumulated average daily discharge for June 1-May 31 for the 2006-2007 and

2007-2008 time periods. A recurrence interval curve was created using 30-year normal daily discharge data for 1971-2000. This curve allows for the determination of the likelihood that a specific discharge will occur in a thirty year period.

To examine the chemical data, concentration-discharge curves were created for the nutrients DOC, NO_3^-, Total-P, and TKN and the ions K^+, Mg^{2+}, Na^+, SO_4^-, and Cl^-. Discharge concentration curves were created for the nutrients DOC, NO_3^--N, and Total-P and the cations K^+ and Mg^{2+}. These curves were created using the concentration data and 15 minute discharge data obtained from the USGS gauging station 03353200. Box plots were created for the nutrients DOC, NO_3^--N, Total-P, and TKN and the ions K^+, Mg^{2+}, Na^+, SO_4^-, and Cl^-. These plots were created with SigmaPlot Graphical software using the concentrations of each respective solute. Mean and median were also determined using Excel. Loading calculations in $kg.ha^{-1}$ and $kg.ha.hr^{-1}$ were determined for DOC, NO_3^--N, Total-P, and TKN. The export of each nutrient was determined at a 15 minute interval by multiplying the 15 minute discharge in $m^3.s^{-1}$ by the concentration of the nutrient at that specific time. The loadings in $kg.ha^{-1}$ were calculated by using the15 minute discharge data and the concentrations at that time to determine the exports in kg for each storm and dividing by the watershed size in ha (27,400). Loadings in $kg.ha.hr^{-1}$ were determined by averaging the discharge for each minute 15 discharge and dividing by the watershed size.

Results

Hydrological response of UECW to precipitation events

The summer and fall of 2007 was characterized by a dry weather pattern for central Indiana, with no significant discharge events occurring on UECW between May 15 and Dec. 1, 2007 (Figure 3). The precipitation total for May 1-Dec. 1, 2007 was 60 percent of the 30-year normal obtained from NOAA (www.NOAA.gov), and is summarized in Table 4. For the period from May-Sept. 2007, the average monthly flow was 34 percent of the 30 year normal (Table 5). Storms 1 and 2 were single event storms with one discharge peak. Storms 3-A, 3-B, and 3-C were three consecutive storms which resulted in three peaks in discharge occurring before discharge returned to pre-event levels. Rainfall totals for these storms ranged from 25 mm for storm 3-B to 42 mm for storm 2. The 7, 30 and 90-day antecedent precipitation totals, total rainfall and rainfall intensity for each storm are summarized in Table 6. Seven day antecedent precipitation ranged from 4 mm for storm 2 to 66 mm for storm 3-C. Thirty day antecedent totals showed the least variation ranging from 65 mm for storm 3-A to 113 mm for storm 1. Storm 1 had the lowest 90 day antecedent precipitation at 247 mm. Storm 3-C had the highest 90 day antecedent precipitation at 312 mm. Rainfall intensity varied from 0.41-5.73 $mm.hr^{-1}$ for storms 1 and 3-C respectively.

In order to better characterize the hydrology of each storm, pre-event discharge, peak discharge and the time to peak were determined and are summarized in Table 6. Storm 2 had the highest peak discharge (0.96 $mm.hr^{-1}$) and storm 3-A had the lowest (0.092 $mm.hr^{-1}$). Storm 3-C, had the fastest time to peak (8.4 hrs) and precipitation intensity (5.73 mm.hr^{-1}). Storm 2 had the highest runoff ratio (0.73). Storm 3-A had the third longest time to peak (17.25 hrs) and the lowest runoff ratio (0.12).

The 7, 30, and 90-day antecedent discharge was also determined and are summarized in Table 6. This provides a means to characterize pre-event conditions, while eliminating uncertainties associated with the heterogeneous nature of precipitation in a watershed of this size. Storm 1 had the lowest 30 and 90-day discharge at 0.010 and 0.005 mm/hour respectively. Storms 3-C, 2, and 3-A had the highest 7 and 30, and 90-day discharge at 0.110, 0.067, and 0.110 mm/hr respectively. Runoff ratios calculated for each storm are summarized in table 6. Storm 2, which followed several high flow events, had the second highest precipitation intensity (2.23 mm.hr^{-1}) and highest 30-day antecedent discharge (0.067 mm.hr^{-1}. Storm 3-A had the second lowest precipitation intensity (0.83 mm.hr^{-1}).

A recurrence interval curve (Figure 4) was created using 1971-2000 average daily discharge data obtained from USGS gaging station number (03353200). This curve allowed a recurrence interval to be calculated for each storm (Table 7). Discharge for storm 2 was largest with a recurrence interval of approximately 2.7 years while storm 3-A was the smallest with a recurrence interval of less than 1 year, however it was the first rain event producing a perceptible rise in the hydrograph following spring planting and the preceding 7 days had 32 mm of precipitation.

Watershed response to precipitation events was examined using double mass curves (Figure 5). Curves were created for June 1-May 31 for 2006-2007 and 2007-2008. To demonstrate variation in watershed response to precipitation between the years, the starting dates of each sampling event are labeled on each curve. The 2006-2007 double mass curve was almost linear, whereas the 2007-2008 curve displays two distinct slopes with the inflection point occurring shortly after storm 1. For the sampling time period, (June 1, 2007-May 31, 2008), the runoff ratio as determined from the double mass curve was 0.39, for the same time period in 2006-2007, the runoff ratio was 0.68. The runoff ratio for the period of low discharge preceding sampling (June 1-November 30, 2007) was 0.04, however for the period during which samples were collected (December 1, 2007-May 31, 2008) the runoff ratio was 0.71.

Ion concentration dynamics

Box plots were created to illustrate the variation in concentrations of the ions K+, Cl^-, Na^+ SO_4^- and Mg^{2+} (Figure 6); the median values of these ions are summarized in Table 8. Storm 3-A, which occurred during planting season had the highest median Na^+, Mg^{2+}, and Cl^- levels, at 21.7 $mg.L^{-1}$, 21.98, and 37.75 $mg.L^{-1}$, respectively. Storm 2 had the highest median K^+ concentrations at 3.60 $mg.L^{-1}$. Sulfate was most elevated in storm 1 at 40.49 $mg.L^{-1}$, while storms 2 and 3-A had the lowest median level at 22.86 and 21.80 $mg.L^{-1}$.respectively. Storm 1 which occurred during a snow event also had the second highest Cl^- and K^+ and Na^+ levels at 35.71, 2.96 and 18.78 $mg.L^{-1}$, respectively.

Cation-discharge curves were created for Mg^{2+}, K^+, Na^+, SO_4^- and Cl^- for storms 1, 2 and 3-A, 3-B, and 3-C (Figures 7, 8, and 9). Storms 2, 3-A, 3-B, and 3-C had Na^+, Cl^-, SO_4^-, and Mg^{2+} concentrations decreasing as discharge increased, reaching minimum concentrations around peak discharge, and increasing again on the receding limb. During storm 1, Mg^{2+}, Na^+, SO_4^-, and Cl^- decreased with increased discharge and reached minimum concentrations after peak discharge and increased on the second half of the receding limb. For storms 3-A, 3-B, and 3-C, Mg^{2+}, Na^+, Cl^-, and SO_4^- exhibited a drop in concentration following each successive storm peak, and did not return to pre-event levels over the duration of the sampling interval. In storm 1, K^+ peaked around the time of maximum discharge and remained elevated on the receding limb. In storm 2 K^+ peaked with or slightly after maximum discharge. For storms 3-B and 3-C, K^+ concentrations increased with discharge, reached a maximum concentration around peak discharge and dropped rapidly on the receding limb.

Nutrient concentration dynamics

Box plots were also created to display variation in concentrations for NO_3^--N, DOC, Total P and TKN (Figure 10). The median value of each nutrient for each storm is summarized in Table 9. Storm 2 had the highest median concentration of DOC at 6.45 $mg.L^{-1}$. For storms 3-A-3-C, DOC concentration ranged from 3.39 $mg.L^{-1}$ -5.54 $mg.L^{-1}$ for storms 3-A and 3-B respectively. Nitrate levels were more elevated in storms 1, 3-B, and 3-C than in the storm with the highest discharge, storm 2. Storm 3-B had the highest median NO_3^--N levels at 5.79 $mg.L^{-1}$. Storm 2 had the highest median concentrations of Total P and TKN at 0.42 $mg.L^{-1}$ and 1.98 $mg.L^{-1}$ respectively. Storm 3-A had the lowest median concentrations of all nutrients.

Nutrient-discharge curves were created for DOC, NO_3^--N, TKN, and Total P for each storm (Figures 11-13). An NH_3-discharge curve was created only for storm 3, since in storms 1 and 2, NH_4 levels were below the detection limit for the majority of samples. DOC concentration increased with discharge for each sampling event, however storm 2 was unique in that DOC concentrations peaked after discharge, whereas it peaked with discharge for the other storms. For storm 1, NO_3^--N concentration peaked around the time of maximum discharge. For the other storms, NO_3^--N concentrations peaked after discharge. For storms 3-A and 3-B, NO_3^--N exhibited an initial dilution as discharge increased, and peaked in concentration on the receding limb. For storm 3-B, NH_3 concentrations increased rapidly on the rising limb and gradually decreased and increased again on the rising limb of storm 3-C. For all storms, TKN and total P concentrations increased with discharge and peaked slightly prior to or with peak discharge.

Discharge-concentration analysis

Hysterisis analysis was used to examine the flushing patterns exhibited by the cations K^+ and Mg^2 for storms 1 (Figure 14) and 3-A, 3-B, and 3-C (Figure 15), and the nutrients DOC, NO_3^--N, and Total P for storm 1 (Figure 16) and storms 3-A, 3-B, and 3-C (Figures 17 and 18). Storm 2 lacked the sample resolution to perform hysteresis analysis on. The curve for Mg^{2+}, displayed a very similar pattern to Ca^{2+} and Na^+, indicating a similar flushing pattern, therefore only the curve of Mg^{2+} is shown. Magnesium displayed a clockwise rotation for all storms with the exception of storm 3-A which had a counterclockwise rotation and the highest concentrations at baseflow. A clockwise rotation indicates the concentration is lower on the receding limb. Curves for storms 3-B and 3-C were similar indicating similar flushing patterns between the two storms. The curves for NO_3^--N exhibited a counterclockwise rotation for storms 1, 3-A, and 3-B, which indicated higher concentrations on the receding limb. Storm 3-A for Mg^{2+} displayed a counterclockwise rotation indicating storm 3-C lacks the distinct pattern of the other storms, however it does display a counterclockwise rotation, indicating a higher concentration on the receding limb. For all storms with the exception of storm 3-A, DOC displays a counterclockwise rotation, indicating that the concentration is higher on the receding limb. For storms 3-B, and 3-C, DOC and K^+

display a similar pattern, indicating a similar flushing mechanism. Nitrate displays a similar pattern to K^+ for storm 3-B indicating a similar flushing mechanism. In storm 3-C, the concentrations on the rising and falling limbs were very close. For storm 3-B, total P had a counter clockwise rotation, indicating that concentrations were highest on the receding limb. Total P and K^+ display a very similar pattern in storm 1, indicating a similar flushing pattern.

Nutrient export rates

Export rates were calculated for each storm for NO_3^--N, DOC, Total P, and TKN in both $kg.ha^{-1}.storm^{-1}$ and $kg.ha^{-1}.hr^{-1}$ (Tables 10 and 11). This allowed both the net export as well as the export rate to be examined. DOC export rate was highest for storm 2 (0.011 $kg.ha^{-1}.hr^{-1}$, while storm 3-B had the highest rate of NO_3^--N export (0.010 $kg.ha^{-1}.hr^{-1}$). Total P and TKN export was highest in storm 2 at 0.001 and 0.004 $kg.ha^{-1}.hr^{-1}$ respectively. Overall, storm 1 had the highest rate of DOC export, storm 2 had the highest rate of Total P, and TKN export, while storm 3-B had the highest NO_3^--N export.

Discussion

1. Characterization of watershed hydrology and determination of potential nutrient delivery pathways.

Major cations were used to examine changes in potential streamwater sources, and assist in flowpath determination for each storm event. For each storm, the Ca^{2+}, Mg^{2+}, Na^{+}, and Cl^{-} export patterns indicate that shallow groundwater contributes more heavily to stream flow on the rising and receding limbs of the flow event, and contributes least heavily around peak discharge and on the beginning of the receding limb. This is illustrated by the hysteresis analysis performed on K^{+} and Mg^{2+}. The hysteresis for Mg^{2+} indicates a pattern that concentration is decreasing on the rising limb and slowly increasing over the duration of the receding limb. In storms 1 and 2, K^{+} concentrations tended to increase with discharge, and maximum concentrations were found around the time of peak discharge indicating overland flow contribution increased with discharge. Potassium in water has been associated with overland flow contributed water (Hood et al., 2006). The fact that K^{+} concentrations are highest around peak discharge for storms 3-B and 3-C indicates that overland flow contribution was likely highest at peak discharge.

Coupling cation data with nutrient and DOC data provides insight into the delivery pathways of these potential water contaminants (Hood et al., 2005; Inamdar et. al, 2004; Vidon et al., 2008; Wagner et al., 2008). In storm 1, NO_3^{-}-N concentrations peaked with discharge and remained elevated for the remainder of the sampling event. Peak NO_3^{-}-N concentrations coincided with reductions in Mg^{2+}, Cl^{-}, and Na^{+} concentrations, indicating groundwater was likely not the primary source of NO_3^{-}-N.

Storm 1 was the first significant flow event (>80th percentile) following an extended dry period lasting from June 1 to December 10, 2007 (Figure 5). This extended dry period likely caused a drop in the water table. Under aerobic conditions, ammonia can be oxidized to NO_3^--N (Mitsch and Gooselink, 2007). The drop in water table could have increased the size of the aerobic zone in the soil profile, and resulted in nitrification occurring in the near stream area. This could have increased the amount of NO_3^--N available to the first flushing of water through the near-stream soil profile, and could explain why NO_3^--N levels peaked with discharge for this storm, as opposed to after peak discharge for the other four storms. Similar flushing patterns have been observed in other studies. Creed et al. (1996) determined the flushing of accumulated N from soil by snowmelt or autumn storms to be an important mechanism of N delivery to streams. A study by Poor and McDonnell (2008) in an Oregon watershed also found that NO_3^--N levels were elevated in the first fall storm following an extended dry period.

Nitrate peaked after discharge for storms 2, 3-A, 3-B, and 3-C, as Mg^{2+}, Na^+, and Cl^- levels began to return to their pre-event levels, and K^+ was decreasing from its peak at maximum discharge. This indicates that NO_3^--N was likely delivered by a combination of shallow subsurface flow and tile drain flow. Water from the agricultural parts of this watershed likely delivered NO_3^--N applied as fertilizer to tile drains, which then delivered the water to multiple tributaries located around UECW. This resulted in NO_3^--N peaking after discharge for storms 2, 3-A, 3-B, and 3-C. Storm 3-B also had the largest maximum NO_3^- concentration of the sampled events. Storm 3-B was the first high flow event occurring after the planting of crops and application of fertilizer which could account for storm 3-B having the highest maximum nitrate levels of the five storms, despite the fact that storms 2 and 3-C had higher maximum discharges. Storms 2 and 3-C which had the highest discharge of all the sampled storms, had lower NO_3^--N concentrations than storms 1 and 3-B respectively. This may indicate that the available pool of NO_3^--N had been flushed out by the three storms occurring in the one month between storms 1 and 2.

The stepdown pattern observed in NO_3^--N concentrations between storms 3-B and 3-C, is similar to one observed by Rusgan et al., (2008) in a forested watershed in Slovenia, with decreased NO_3^--N concentrations on the third peak of a multiple peak storm. Storms 2 and 3C had the highest and second highest discharges, yet storm 3-B had a higher NO_3^--N export and maximum NO_3^--N concentrations than storm 3B. Storm 3-C occurred less than 3 days after storm 3-B, however it is not possible to definitively say this since there are data points missing on the receding limb of storm 3-C. Storm 2 followed four high flow events in the preceding 29 days which may account for the lower observed NO_3^--N concentrations than for storm 3B. This indicates that perhaps there is a limited pool of available NO_3^--N, which has the potential of being flushed out and depleted by successive storms. Poor and McDonnell (2007) observed a similar pattern in a watershed in Oregon with elevated NO_3^--N levels in fall storms, which decreased over the duration of the rainy season.

Ammonia concentrations peaked on the rising limb of storm 3-A, and with discharge for storms 3-B and 3-C. Storm 3-C had lower NH_3 concentrations than storm 3-B, despite having a higher discharge. Like NO_3^--N, this may indicate that there is also a limited pool of NH_3^--N available for flushing, however with data points missing on the receding limb of storm 3-C, it is not possible to definitively say this. The high levels of ammonia observed in storms 3-A, 3-B, and 3-C with respect to storms 1 and 2, is likely due to the fact that the storm occurred during planting season, and ammonia had been applied to the fields as fertilizer.

Total P and TKN concentrations increase rapidly on the rising limb, reaching maximum concentration shortly prior to or with peak discharge and decreased rapidly on the receding limb. Total P and TKN concentrations seem to almost mirror discharge. Cation data indicates that the primary delivery mechanism for these nutrients is overland flow. Hysteresis analysis show the flushing mechanisms between K^+ and Total P to be similar displaying counterclockwise rotation with concentrations more elevated on the receding limb. Particulate P is subject to transport by overland flow (Gentry et al., 2007; Royer et al., 2006; Clark and Cooper, 1988). Gentry (2007) found that overland flow transported particulate P was the dominant form during storm events, and contributed heavily to total P loading during storm events. TKN is also subject to transport by overland flow. A study by Cooke and Cooper (1987) found that virtually all TKN exported during storm events from a New Zealand watershed was transported to the stream by overland flow. Storm 2 had the highest median levels of Total P and TKN. The high runoff ratio (0.72) of storm 2 indicates saturated soil conditions, which would facilitate overland flow. This may explain why TKN and Total P concentrations were highest for this storm. Additionally, as storm 2 was the storm with the highest discharge, instream sediments may have been mobilized particle associated TKN, resulting in the high observed concentrations. Total Suspended and Turbidity also display a very similar pattern to Total P and TKN for this storm, so clearly sediment was being mobilized by this event at rates similar to Total P and TKN.

Cation data for storms 1, 3-A, 3-B, and 3-C indicate that DOC was delivered primarily by overland flow with some contribution from shallow subsurface flow. The fact that DOC concentration increases with discharge and peaks around the same time as K^+ indicates overland flow is the primary delivery mechanism during periods of high discharge. This is consistent with the findings of a study by Vidon et al. (2008), in a nearby watershed, which found overland flow to be an important delivery mechanism for DOC during periods of high stream discharge. In storm 1, DOC levels remain elevated on the receding limb of the storm indicating that shallow groundwater continued to deliver DOC rich water to the stream following the cessation of overland flow. In storm 2, DOC concentrations peaked after discharge as K^+ concentrations were decreasing, and Mg^{2+}, Ca^{2+}, and Cl^- concentrations were beginning to rebound to their pre-event levels. This indicates that there may have been a heavier groundwater contribution of DOC than during the other storms. Storm 2 had the highest DOC concentrations of any storms, so clearly there was no depletion between storm 1 and storm 3. DOC also did not display the same step down pattern displayed by NO_3^--N or NH_3 between storms 3-B and 3-C, although it is not possible to say this definitively since there are missing data points. This is different than finding by Hood et al. (2006), who noted a reduction in maximum DOC concentration following a three peak storm.

2. Is there seasonal variation in export patterns and loadings?

Several studies have examined seasonal variation in nutrient flux in watersheds under agricultural and mixed land use. Royer et al. (2006) found that the majority of NO_3-N and P export from three agricultural watersheds in Illinois consistently occurred between January and June. Coulter et al. (2004) examined nitrogen and phosphorus export from a mixed land use watershed in Kentucky. In these studies, mean NO_3^--N concentrations were highest in the winter, while total P concentrations were highest in the summer months.

Storms 1 and 3-B had the highest median concentrations of NO_3^--N. Storm 1 also had NO_3^--N levels peaking with discharge, as opposed to after it like the other storms. Storm 1 was the first flushing event following a 6 month period of low flow, and the elevated nitrate levels could be due to the fact that 6 months worth of accumulated NO_3^--N were flushed from the watershed. Storm 3-B was the first high discharge event following the spring planting season NO_3^--N applied as fertilizer was likely flushed from the fields through tile drains. Storm 2 was the fourth high flow event occurring in two months, and it is possible that a lot of accumulated soil nitrate had likely been flushed from the watershed, by these storms, which could explain its lower levels. Storm 3-A had the smallest discharge of any of the storms as well as the lowest median NO_3^--N levels. This may be because the storm was too small to mobilize large amounts of N. Ammonium concentrations for storms 1 and 2 were generally below the detection limit, however ammonium concentrations in storm 3 were much higher and increased with discharge. Storm 3 occurred shortly after planting. Farmers often apply nitrogen in granular form or as ammonia a few weeks before planting (Tedesco et al., 2005), and some of this ammonia was likely flushed from the fields by the storm event. Total P and TKN concentrations were highest in the winter storm. Royer et al. (2006), also found elevated phosphorus concentrations during the winter in an agricultural watershed in Illinois. Storm 2 was also the flashiest, indicating that overland flow was high which would have resulted in increased amounts of particulate P and TKN being delivered to the streams.

The agricultural activity in this watershed likely resulted in NH_4^+ and NO_3^--N levels being the highest in the spring. The winter storm had the lowest mean NO_3^--N levels, and was the only storm where maximum concentration occurred with peak discharge. The winter storm also had the highest median DOC levels, and was the only storm where DOC levels peaked after discharge. Overall, this study did not conclusively indicate that season was a driving force behind the observed variations in nutrient export dynamics. Rather a combination of antecedent conditions, agricultural practices, and storm selection may have played a larger role.

3. Does there appear to be a variation between nutrient export patterns on the UECW and those observed on smaller watersheds?

Work on two smaller watersheds (< 20 km^2) in central Indiana was conducted in summer and fall of 2006 (Wagner et al., 2008). The export patterns on the Eagle Creek in this study were generally smoother, less noisy, and displayed more distinctive patterns than those on the smaller watersheds. In the smaller watersheds, the NO_3^--N export pattern was very noisy and lacked a distinct pattern, whereas in the UECW, NO_3^--N displayed a clear pattern and usually reached maximum concentration after peak discharge. In all three watersheds, DOC tended to peak with discharge (Wagner et al., 2008). DOC export patterns on the smaller watersheds were also quite noisy, concentration tended to increase with discharge, however lacked the smooth pattern of the UECW. Cation and Cl^- patterns were similar on all three watersheds, but were smoother on the UECW. The cation patterns observed in smaller watersheds were similar to those observed in UEWC, Mg^{2+}, Na^+, and Cl^- decreased with increased discharge, and K^+ increasing with discharge, however, similar to the nutrients, the cation patterns on the UEWC were less noisy.

The differences observed between the UECW and the two smaller watersheds validated the hypothesis that larger watersheds would display smoother more homogeneous nutrient export curves. The mixing effect due to multiple tributaries produced a more complex stream network which was more stable and this resulted in smoother more consistent export patterns. This smoothing effect was especially apparent in NO_3^--N export, which was extremely erratic in the smaller watersheds, but displayed a definite pattern in UECW.

Conclusions

This study resulted in an enhanced understanding of nutrient export dynamics at a scale where there is a paucity of knowledge. Total P and TKN appear to be transported to the streams by overland flow and tend to reach maximum concentration around peak discharge. DOC is transported primarily by overland flow, with some contribution from shallow groundwater. Nitrate displays a pattern consistent with tile drain delivery, with some contribution from shallow groundwater or macropore flow. The enhanced knowledge of the transport mechanisms of these contaminants will assist in the development and implementation of BMP's to minimize the problem.

This work also demonstrates that at larger scales, the export patterns of DOC and NO_3^--N are smoother than those observed on smaller nearby watersheds. DOC generally peaks with discharge, whereas, NO_3^--N usually peaks after discharge. These findings provide important insight into the transport mechanisms and export patterns of these contaminants at a scale more practical from a policymaking perspective. An enhanced understanding of the relationship of various solutes, and how their concentrations change with discharge and scale will assist modelers in predicting and evaluating the nutrient loading potential of watersheds. On a more local level, this knowledge will assist in combating the algal blooms occurring each summer in the Indianapolis reservoirs. This is important because some of these blooms produce toxins which are harmful to humans and animals.

Land Use Type	Percent Area	Area (km^2)
Agricultural	52	142.5
Herbaceous	29.9	81.9
Forested	9.3	25.5
High and Low Density Development	4.3	11.8
Other	4.5	12.3

Table 1. Land use summary upstream of sampling site 2007.

Storm	Date	Number of Samples
1	December 10-14, 2007	14
2	January 8-14, 2008	9
3-A	May 7-11, 2008	17
3-B	May 11-14, 2008	13
3-C	May 14-26, 2008	16

Table 2. Date and number of samples taken for each storm.

Sample(s) Analyzed	Instrument Used	Technique Employed	Detection Limit (mg.L^{-1})
NO_3^-	Dionex Ion Chromatograph	EPA Method 300.0	0.1
Mg^{2+}	Dionex Ion Chromatograph	EPA method 300.7	1
K^+	Dionex Ion Chromatograph	EPA method 300.7	0.05
Na^+	Dionex Ion Chromatograph	EPA method 300.7	1
Cl^-	Dionex Ion Chromatograph	EPA method 300.0	8
NH_4^+	Ion Exchanger	SM 4110	8.0
DOC	Dionex Ion Chromatograph	SM 5310 Persulfate oxidation method	0.5
Total P	Outsourced	EPA Method 365.2	0.010
TKN	Outsourced	EPA Method 351.3	0.010

Table 3. Analytical methods and detection limits.

Month	Precipitation (mm)	Percent Normal
May	50.03	0.45
June	56.39	0.54
July	48.26	0.43
August	87.12	0.90
September	40.39	0.55
October	71.12	1.01
November	46.99	0.51

Table 4. Comparison of May-November, 2007 precipitation to 1971-2000 thirty year normal.

Month	2007 Percent 30 Yr. Normal
January	374
Febuary	64
March	273
April	90
May	39
June	17
July	21
August	57
September	40

Table 5. Comparison of January-September 2007 discharge to 1971-2000 thirty year normal.

	Storm 1	Storm 2	Storm 3-A	Storm 3-B	Storm 3-C
Bulk Precipitation (cm)	40.6	42.2	33.3	24.9	40.1
7-day antecedent Precipitation (mm)	48	4	32	40	66
30-day antecedent Precipitation (mm)	113	81	65	90	97
90-day antecedent Precipitation (mm)	247	270	258	287	312
7-day antecedent Discharge (mm/h)	0.022	0.046	0.019	0.033	0.110
30-day antecedent Discharge (mm/h)	0.010	0.067	0.038	0.040	0.046
90-day antecedent Discharge (mm/h)	0.005	0.025	0.110	0.084	0.086
Mean Stormflow (mm/h)	0.113	0.245	0.054	0.204	0.106
Peak Discharge (mm/h)	0.365	0.962	0.092	0.451	0.622
Baseflow (mm/h)	0.007	0.037	0.017	n/a	n/a
Runoff Ratio	0.42	0.73	0.12	0.58	0.56
Time to Peak (hr)	68.4	17.5	17.25	14.0	8.5
Precipitation intensity (mm/hr)	0.41	2.23	0.83	1.55	5.73

Table 6. Total precipitation (mm), 7, 30, and 90 day antecedent discharge, baseflow and peak discharge, runoff ratio, time to peak, and precipitation intensity of storms 1, 2, 3-A, 3-B, and 3-C.

Storm	Recurrence Interval (years)
1	1.08
2	2.7
3-A	< 1
3-B	2.08
3-C	2.27

Table 7. Storm recurrence interval summary (years).

	Storm 1	Storm 2	Storm 3-A	Storm 3-B	Storm 3-C
Mg^{2+}	16.65	13.20	21.98	16.43	20.36
K^{+}	2.96	3.60	2.46	2.88	2.19
Na^{+}	18.78	12.01	21.70	10.45	13.57
Cl^{-}	35.71	22.73	37.75	22.85	26.36
SO_4^{-}	40.49	22.86	33.40	21.80	28.29

Table 8. Median concentration values for major ions ($mg.L^{-1}$).

	Storm 1	Storm 2	Storm 3-A	Storm 3-B	Storm 3-C
DOC	5.02	6.45	3.39	5.54	4.26
NO_3^-	4.14	2.98	1.28	5.79	3.78
TKN	1.50	1.98	0.91	1.53	0.98
Total P	0.28	0.42	0.06	0.24	0.15

Table 9. Median concentration values for nutrients ($mg.L^{-1}$).

	DOC	$N\text{-}NO_3^-$	TKN	Total-P
Storm 1	0.93	0.67	0.17	0.037
Storm 2	2.09	0.93	0.77	0.21
Storm 3A	0.16	0.094	0.049	0.0046
Storm 3B	0.82	0.71	0.28	0.045
Storm 3C	1.18	1.00	0.37	0.081

Table 10. Loading summary in $kg*ha^{-1}$ for DOC, $N\text{-}NO_3^-$, TKN, and Total P.

	DOC	$N\text{-}NO_3^-$	TKN	Total-P
Storm 1	0.0078	0.0056	0.0025	0.00052
Storm 2	0.011	0.0049	0.0040	0.0011
Storm 3A	0.0015	0.00090	0.00044	0.000042
Storm 3B	0.012	0.010	0.0040	0.00063
Storm 3C	0.0040	0.00010	0.0031	0.00080

Table 11. Loading summary in $kg*ha^{-1}*hr^{-1}$ for DOC, $N\text{-}NO_3^-$, TKN, and Total P.

	Storm 1	Storm 2	Storm 3-A	Storm 3-B	Storm 3-C
DOC base	0.026	0.059	0.014	N/A	N/S
DOC peak	0.64	0.51	0.075	0.59	0.80
NO_3^- base	0.0079	0.027	0.0026	N/A	N/A
NO_3^- peak	0.40	0.51	0.015	0.49	0.43
TKN base	0.0050	0.013	0.0029	N/A	N/A
TKN peak	0.28	0.88	0.016	0.34	0.50
Total P base	0.00051	0.0021	0.0026	N/A	N/A
Total P peak	0.071	0.33	0.015	0.49	0.42

Table 12. Export rates for DOC, NO_3^-, Total P, and TKN at base and peak flow in $kg.ha^{-1}.hr^{-1}$ and $kg.ha^{-1}$.

Figure 1. Map of Site.

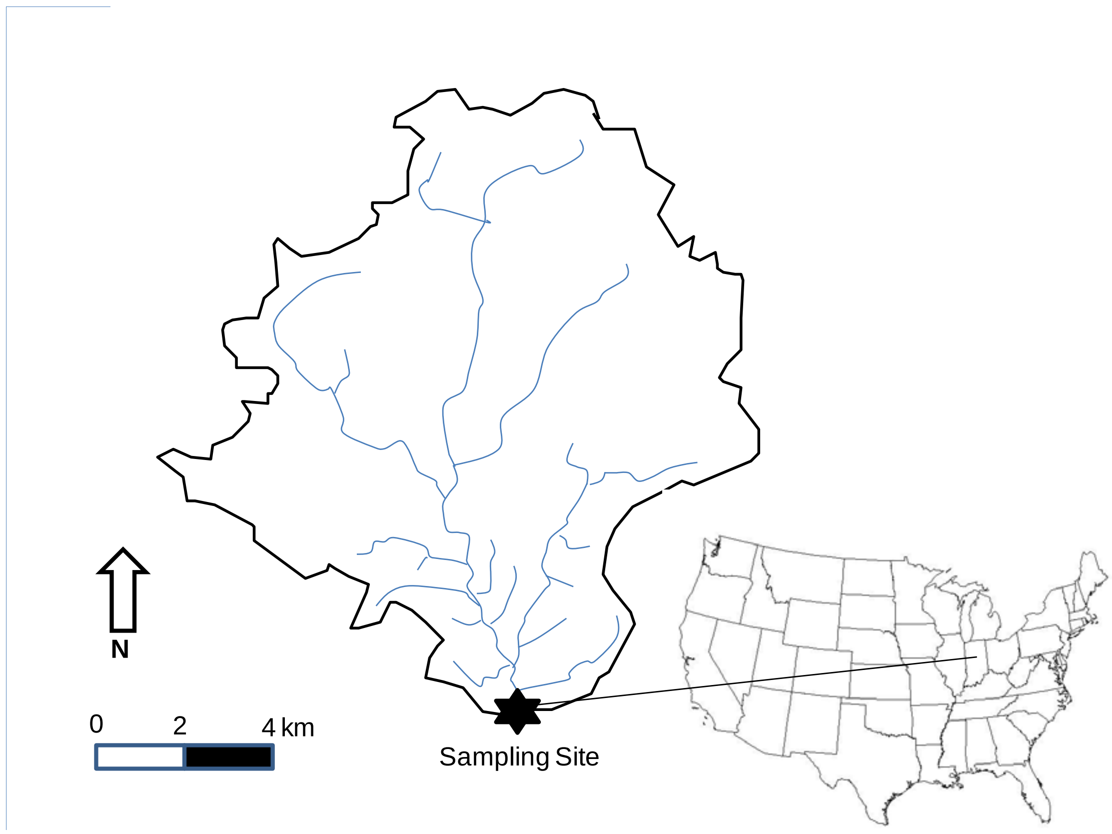


Figure 2. Sample Processing Diagram.

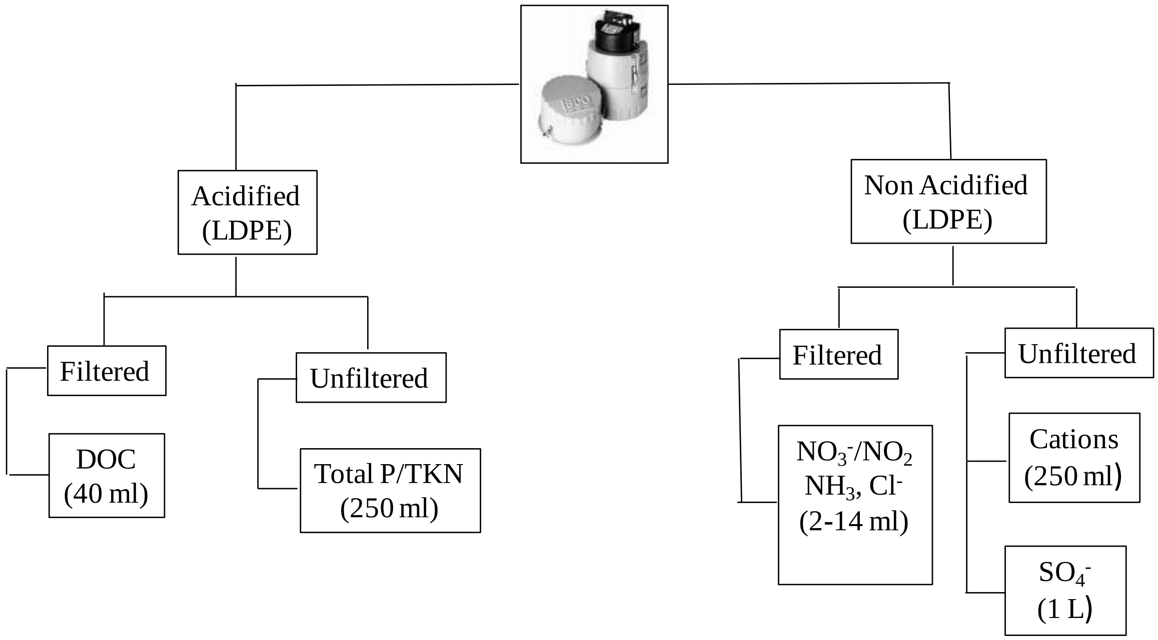


Figure 3. Daily Discharge-Hyetograph showing average daily precipitation in mm and average daily discharge in $m^3.s^{-1}$ for June 1, 2007-June 1, 2008.

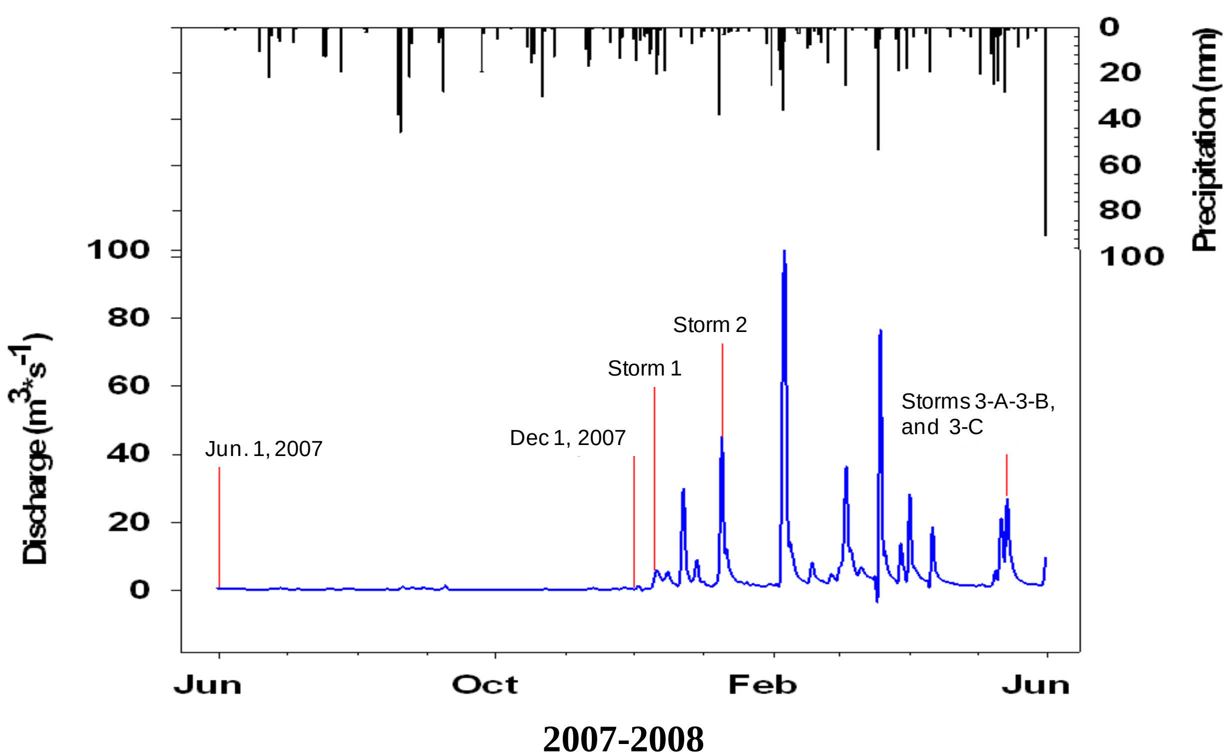


Figure 4. Recurrence interval curve using 30 year daily USGS discharge data from 1971-2000.

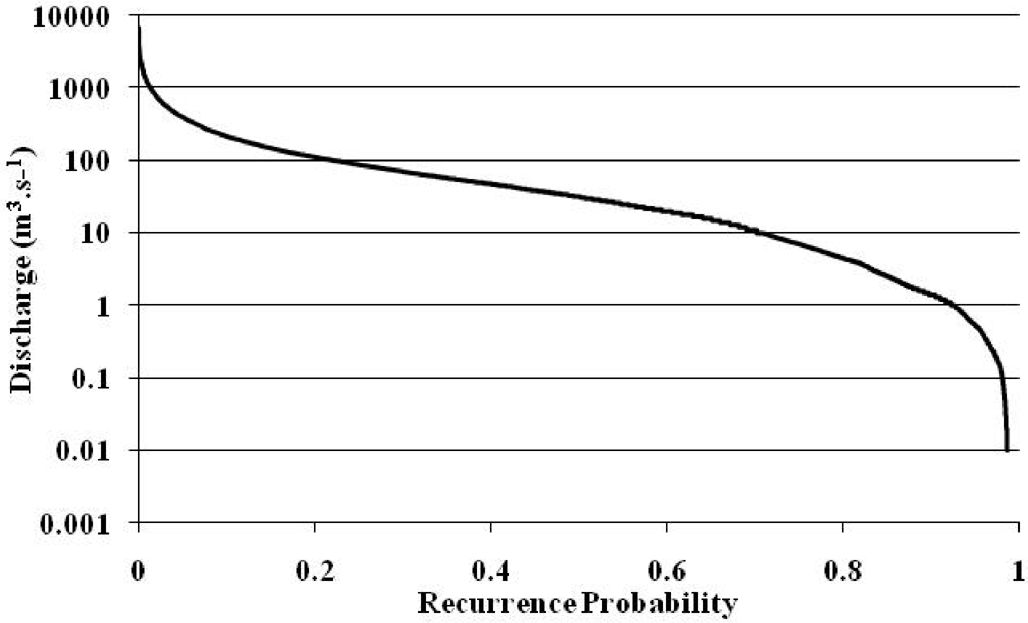


Figure 5. Double mass curves showing commencement of sampling for storms 1 (Dec 10), 2 (Jan 8), and 3-A, 3-B, and 3-C (May 7) for June 1, 2006-May 31, 2007 and June 1, 2007-May 31, 2008.

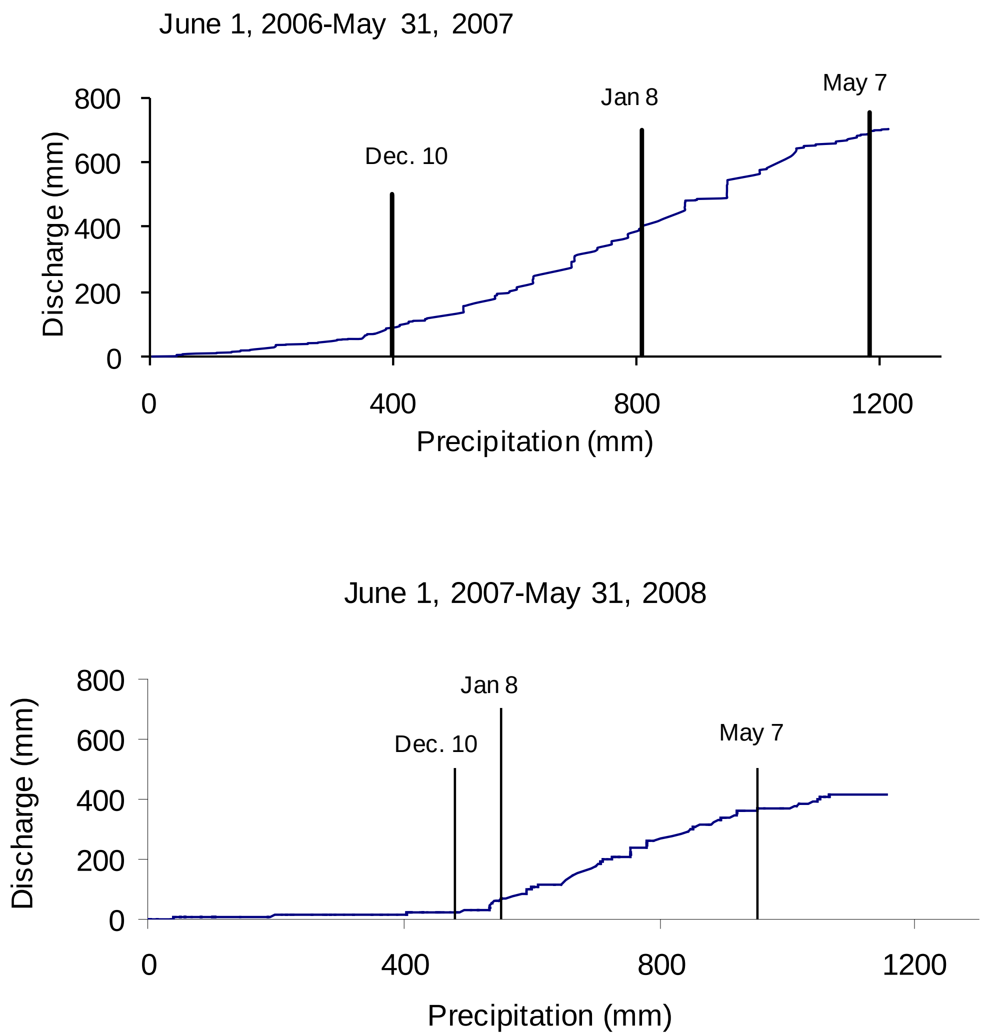


Figure 6. Box plots for Mg^{2+}, K^{+}, Na^{+}, and Cl^{-} and SO_4^{-}. Plots display 5^{th}, 25^{th}, median, 75^{th}, 95^{th} percentiles and outliers for storms 1, 2, 3-A, 3-B, and 3-C. Storms are labeled on the X-Axis.

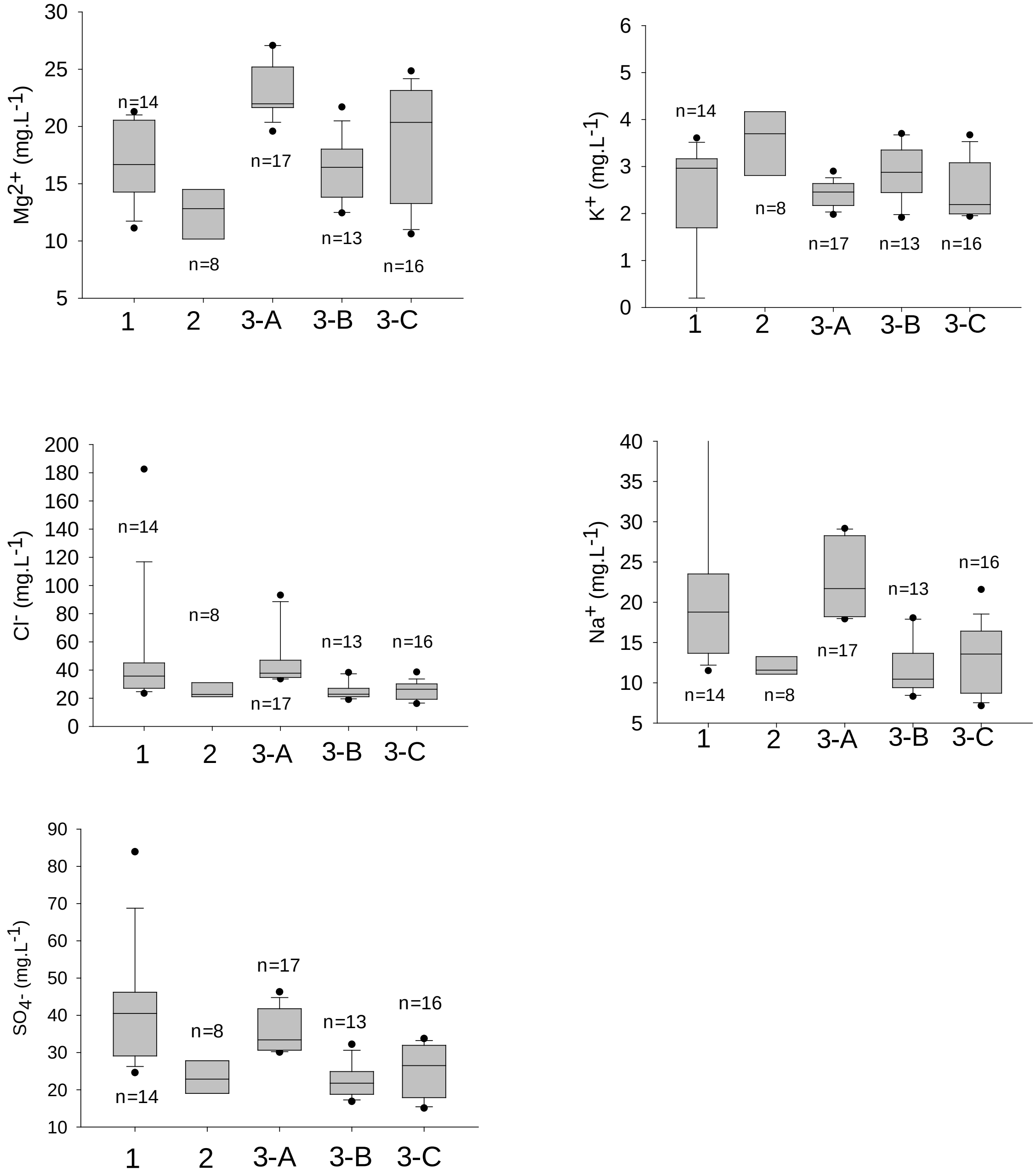


Figure 7. Concentration-Discharge Graphs for storm 1 for cations Mg^{2+}, K^{+}, Na^{+} and anions Cl^{-} and SO_4^{-} in $mg.L^{-1}$.

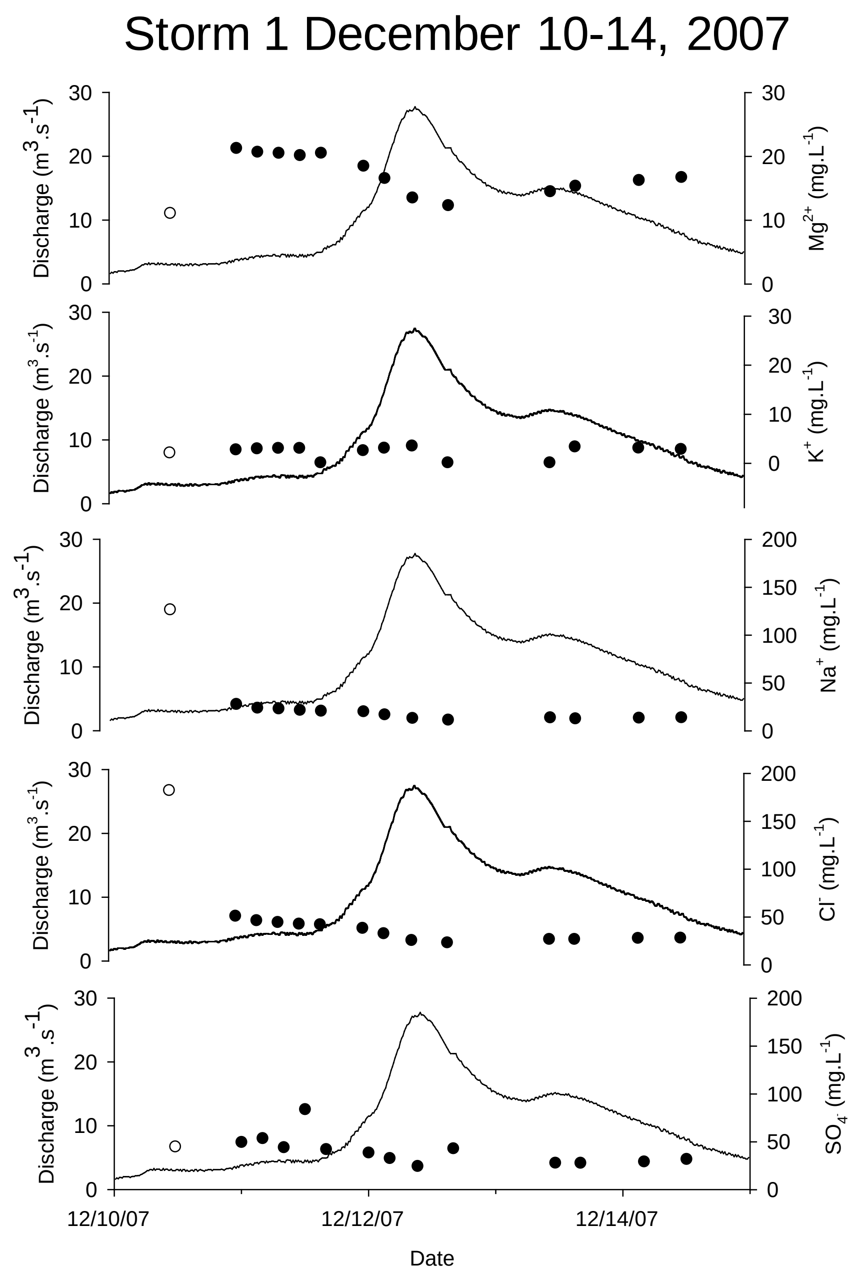


Figure 8. Concentration-Discharge Graphs for storm 2 for cations Mg^{2+}, K^{+}, Na^{+}, anion SO_4^{-}, and Nutrients DOC, NO_3^{-}, Total P, and TKN in $mg.L^{-1}$.

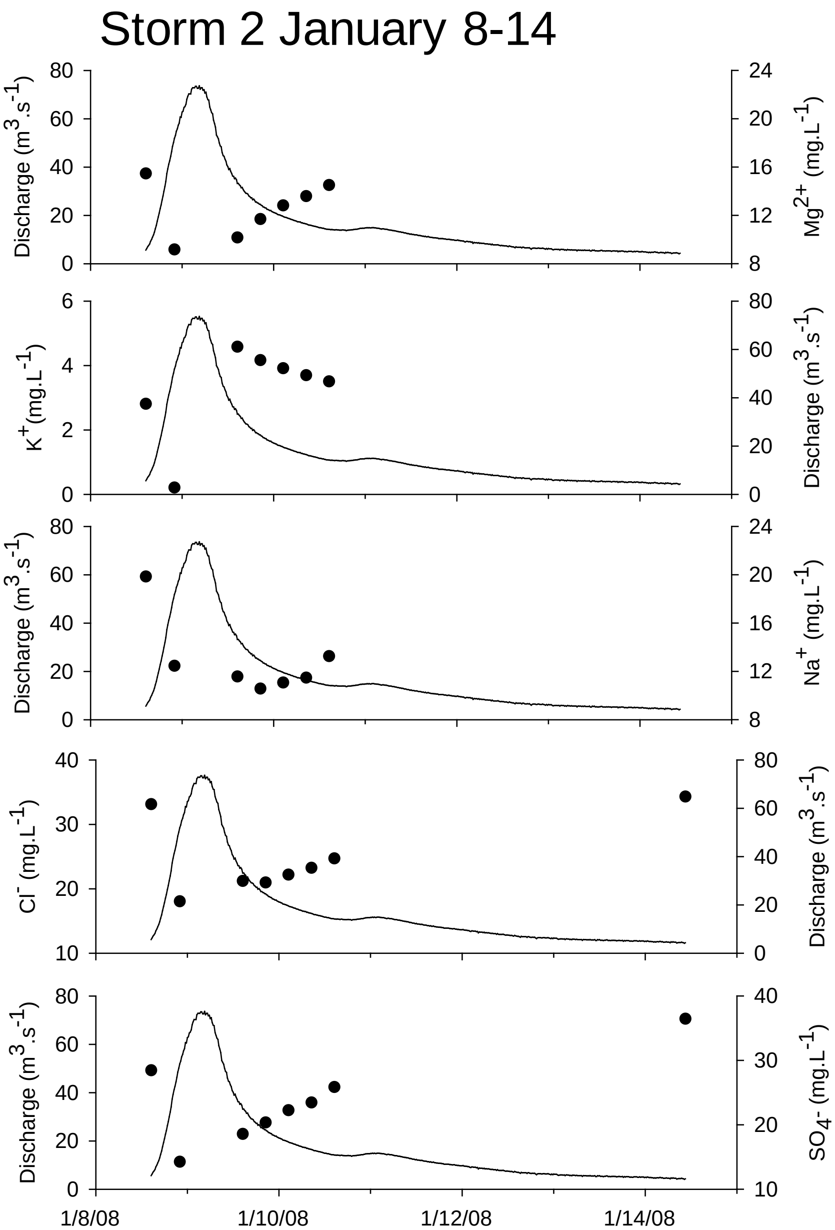


Figure 9. Concentration-Discharge Graphs for storms 3-A, 3-B, and 3-C for cations Mg^{2+}, K^{+}, Na^{+} and anions Cl^{-} and SO_4^{-} in $mg.L^{-1}$.

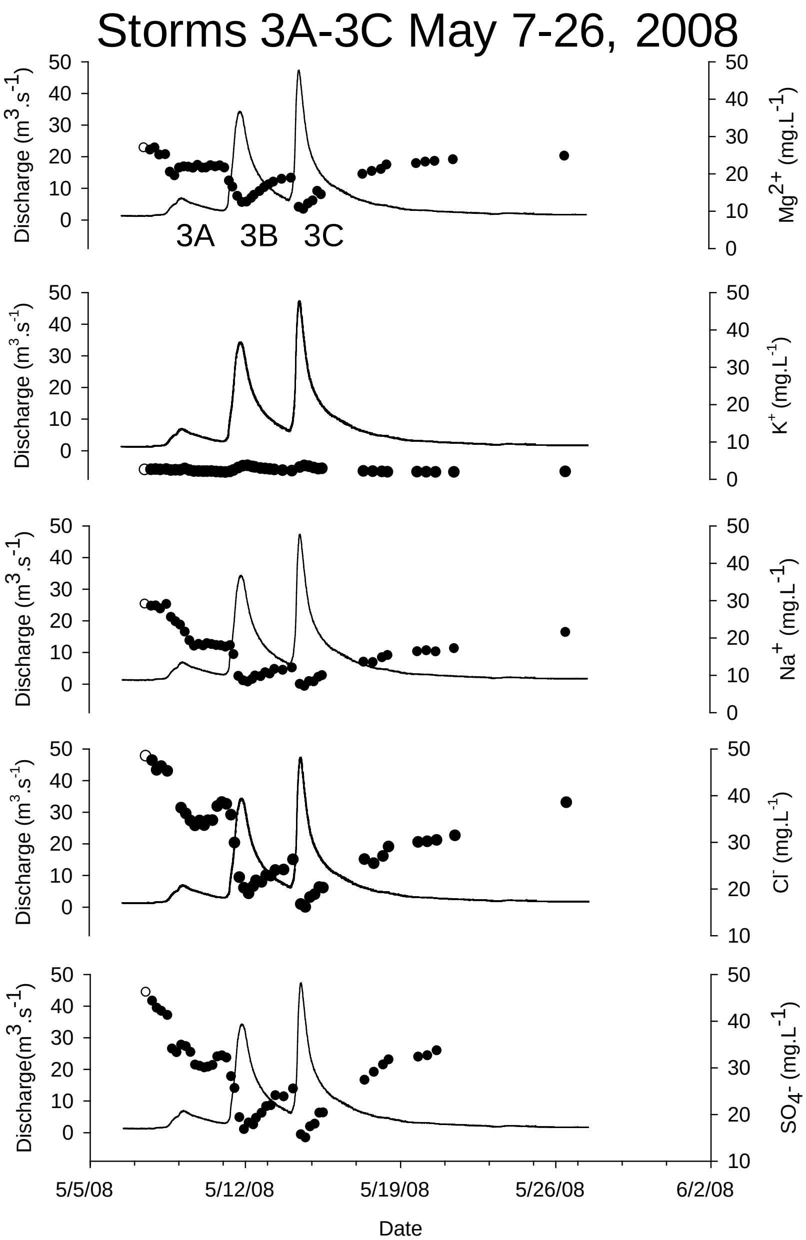


Figure 10. Box Plots for NO_3^-, DOC, TKN, and Total P. Plots display 5th, 25th, median, 75th, 95th percentiles and outliers for storms 1, 2, 3-A, 3-B, and 3-C. Storms are labeled on X-Axis.

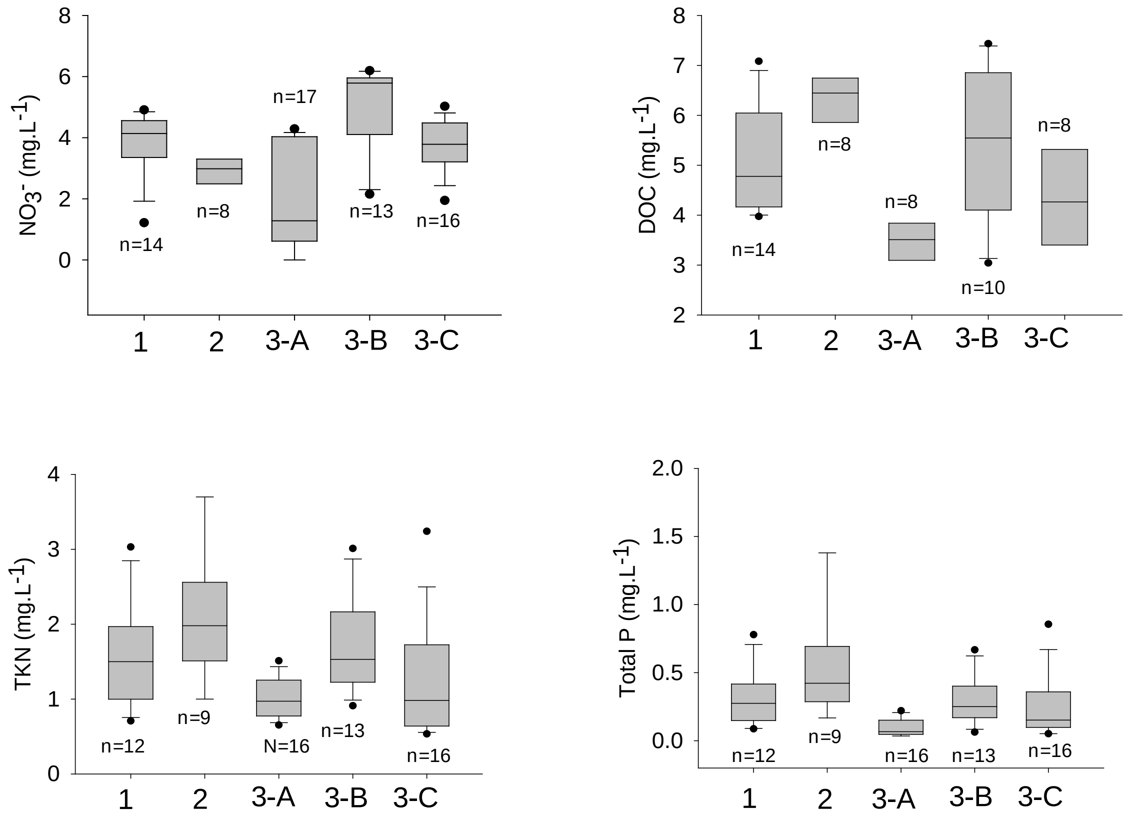


Figure 11. Concentration-Discharge graphs for storm 1 nutrients DOC, NO_3^-, Total P, and TKN in $mg.L^{-1}$.

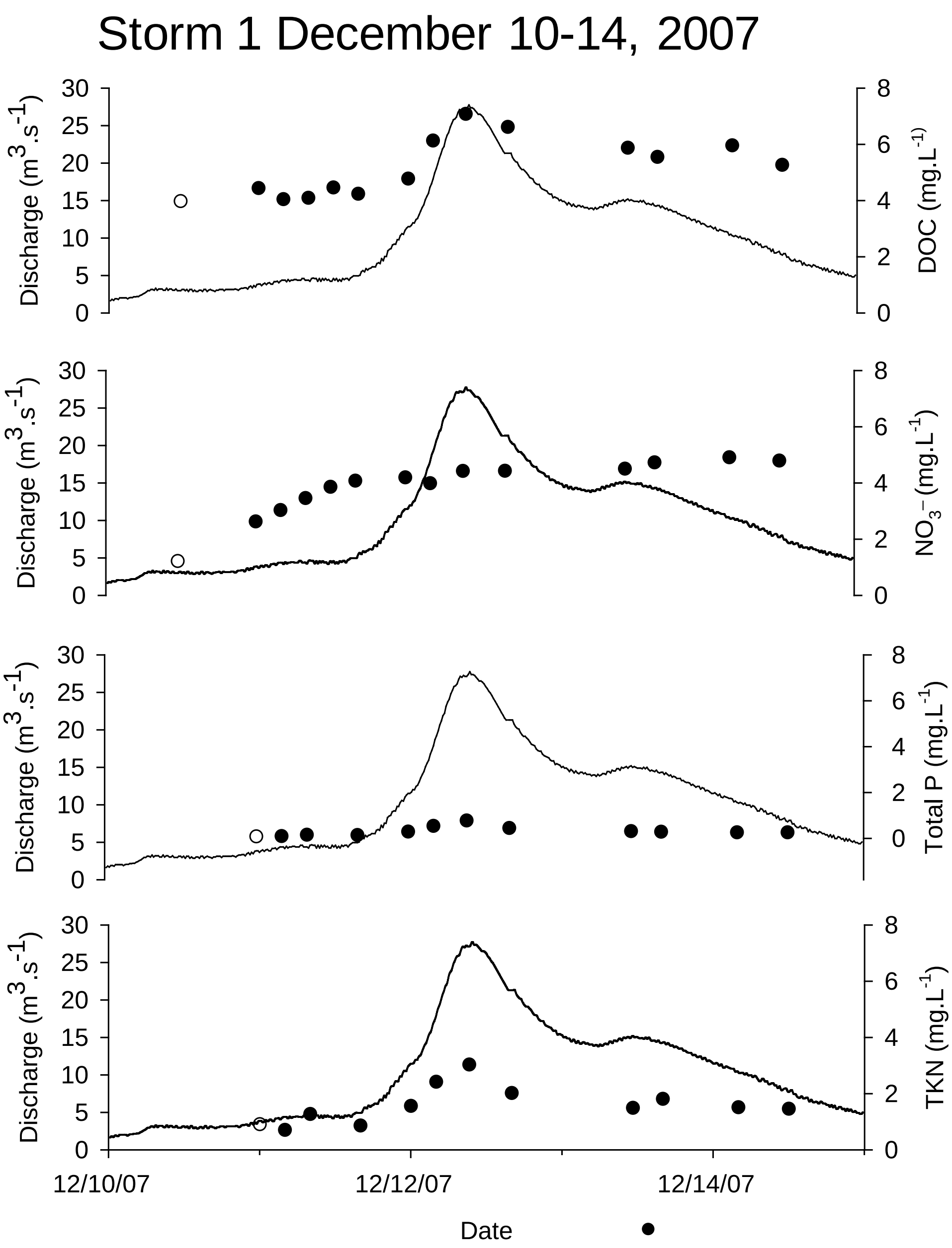


Figure 12. Concentration-Discharge graphs for storm 2 nutrients DOC, NO_3^-, Total P, and TKN in $mg.L^{-1}$.

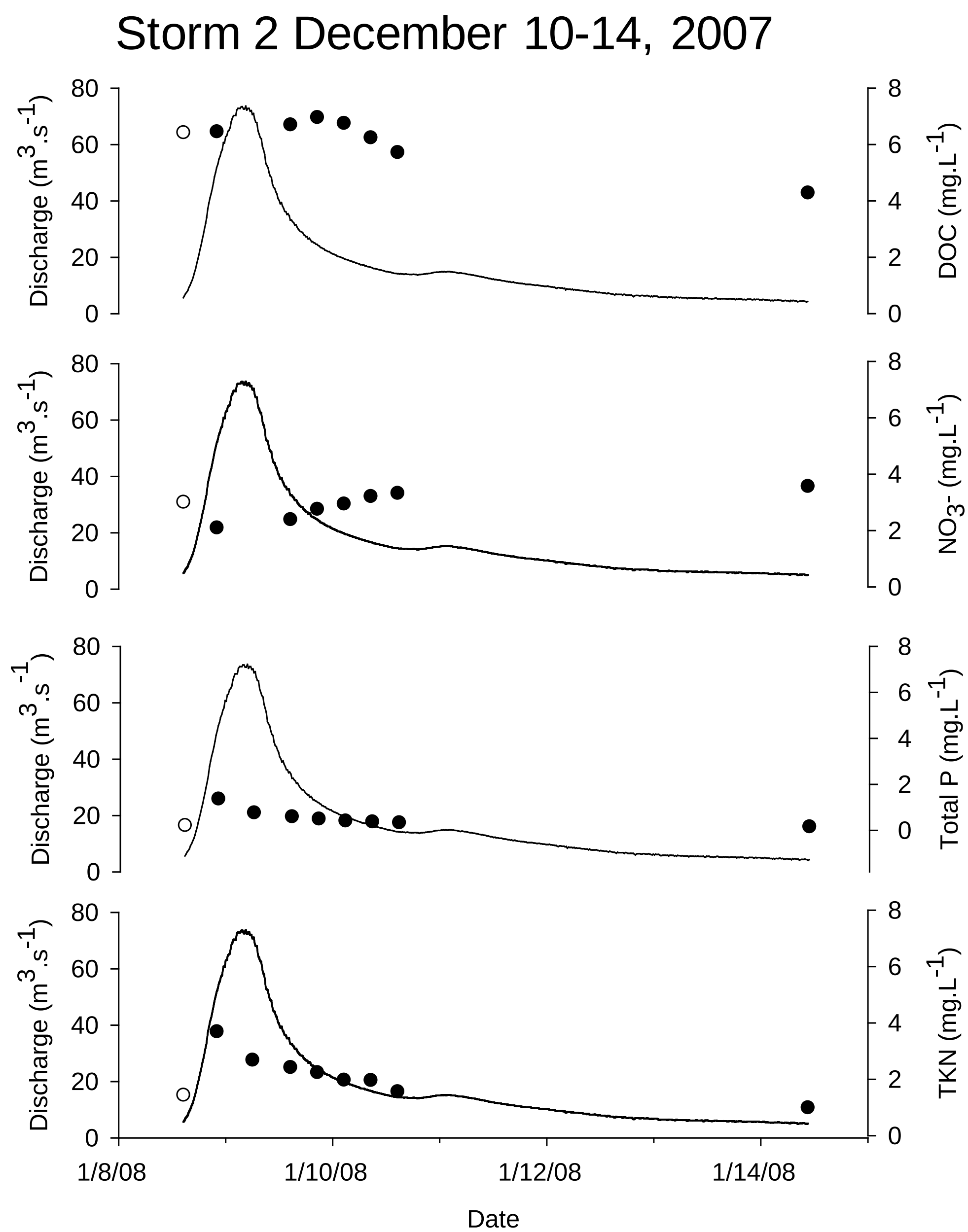


Figure 13. Concentration-Discharge graphs for storms 3-A, 3-B, and 3-C for nutrients DOC, NO_3^-, Total P, NH_4^+ and TKN in $mg.L^{-1}$.

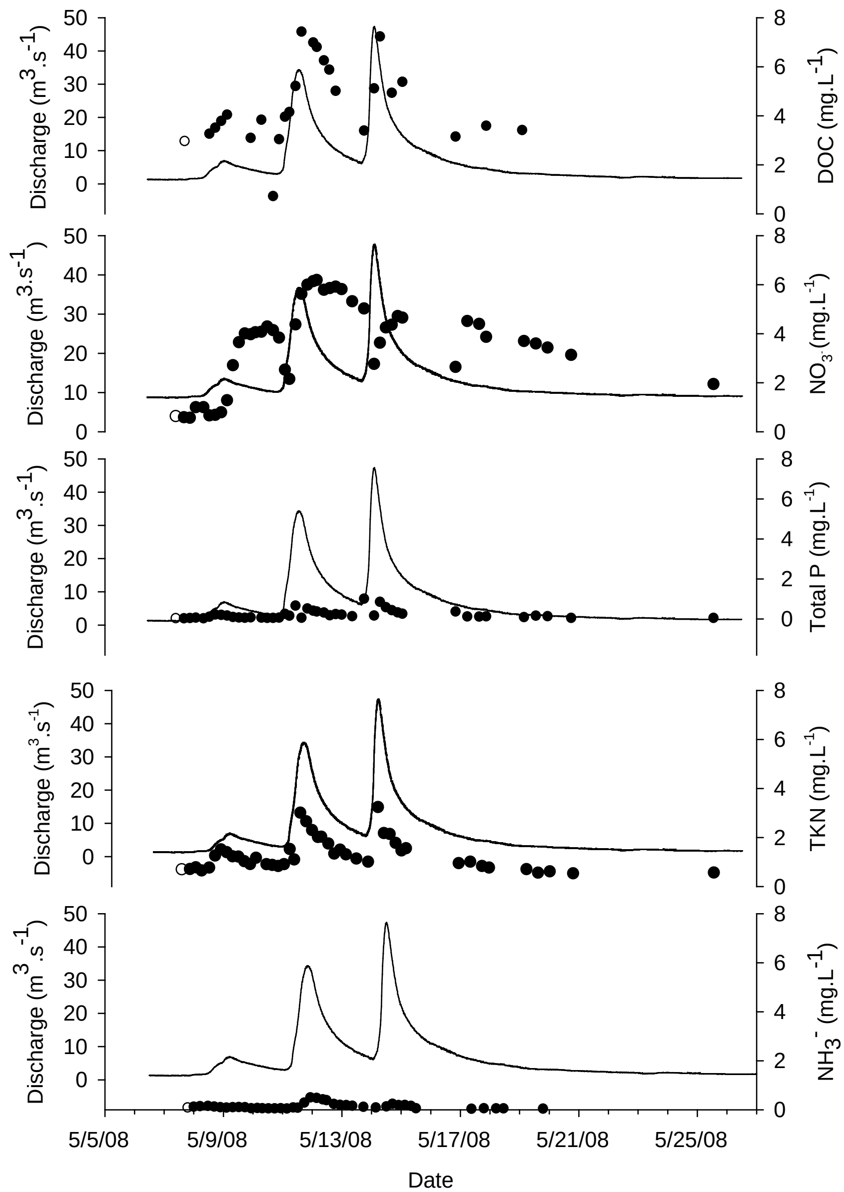


Figure 14. Discharge-Concentration curves of Mg^{2+} and K^{+} for storm 1.

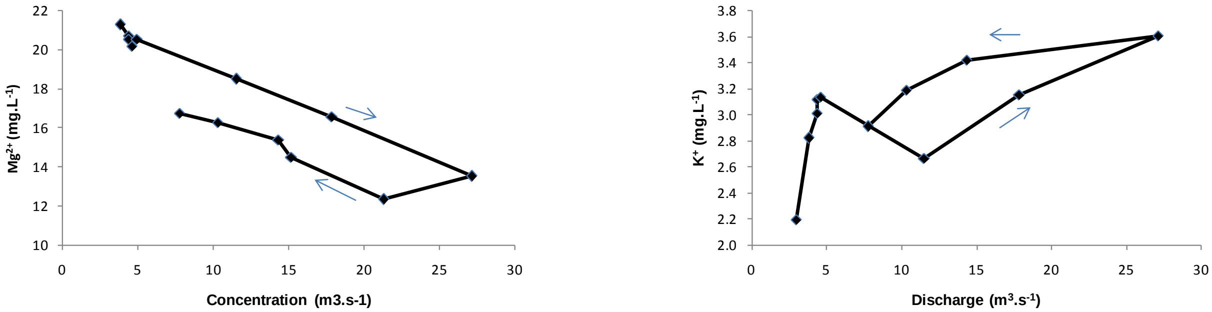


Figure 15. Discharge-Concentration curves of Mg^{2+} and K^{+} for storms 3-A, 3-B, and 3-C.

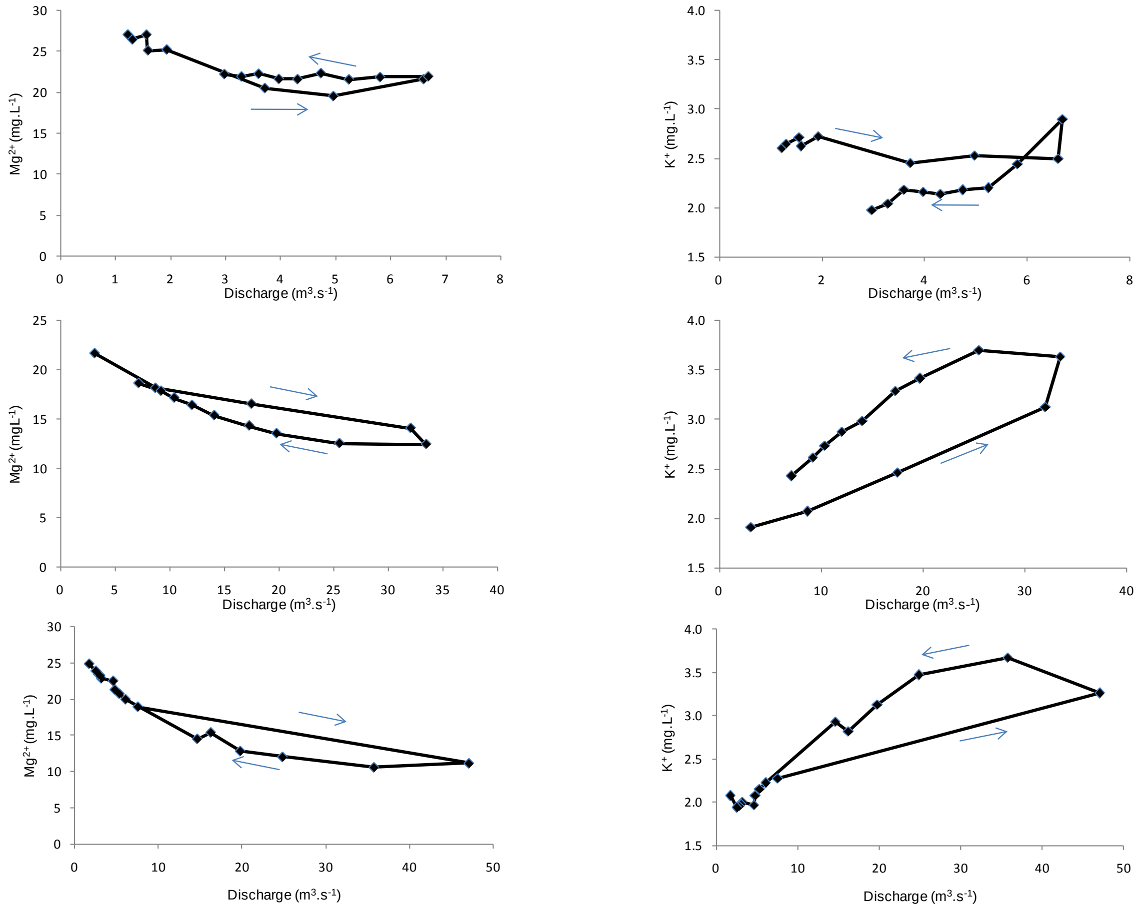


Figure 16. Discharge-Concentration curves of DOC, NO_3^-, and Total P for storm 1.

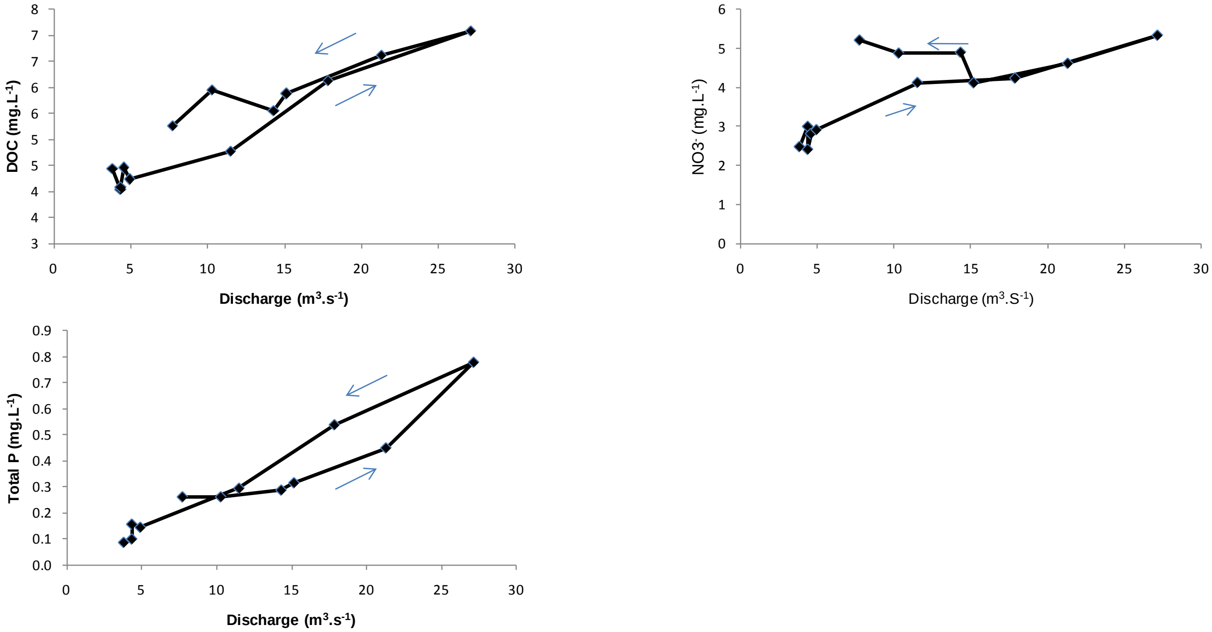


Figure 17. Discharge-Concentration Curves for DOC and NO_3^- for storms 3-A, 3-B, and 3-C.

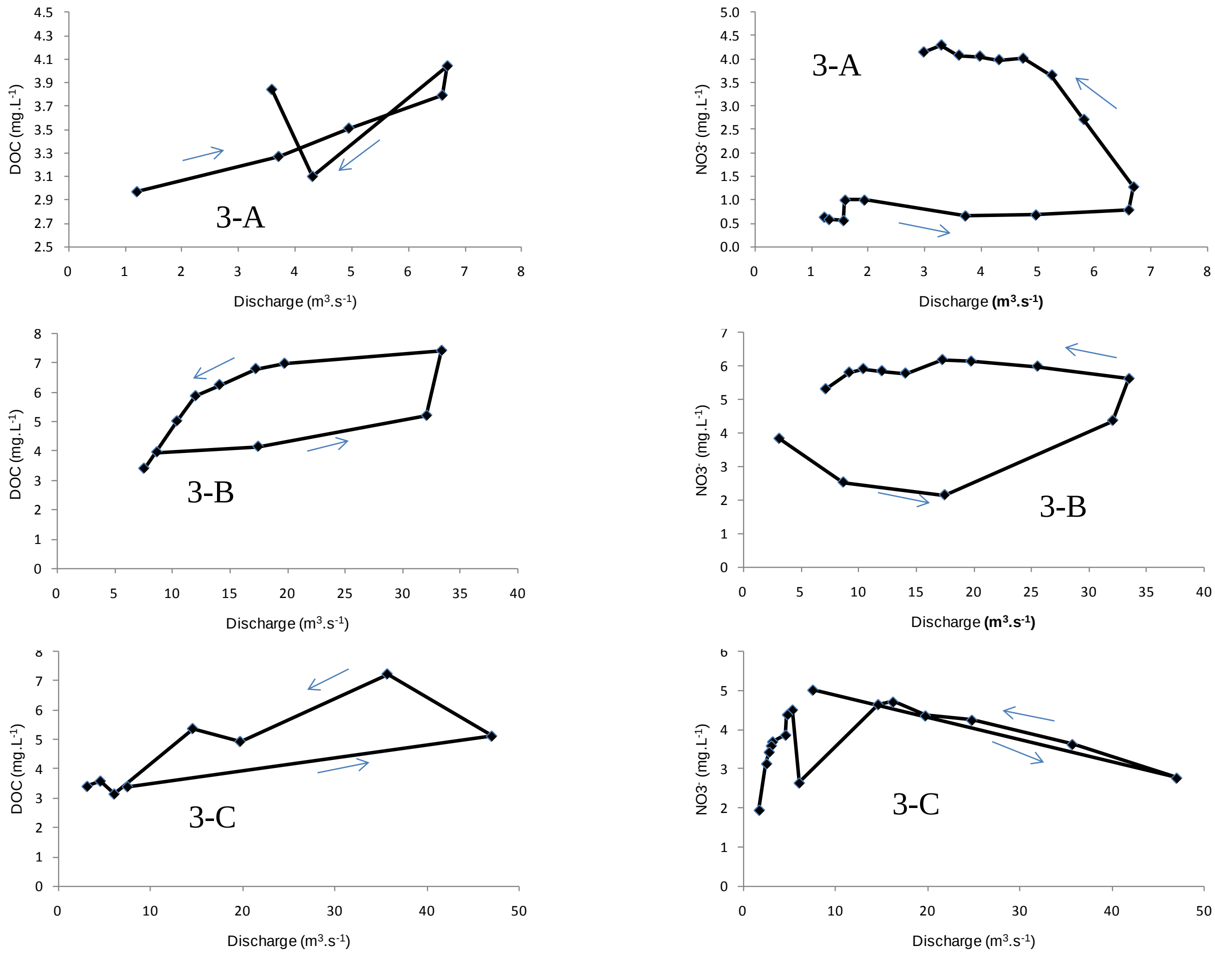
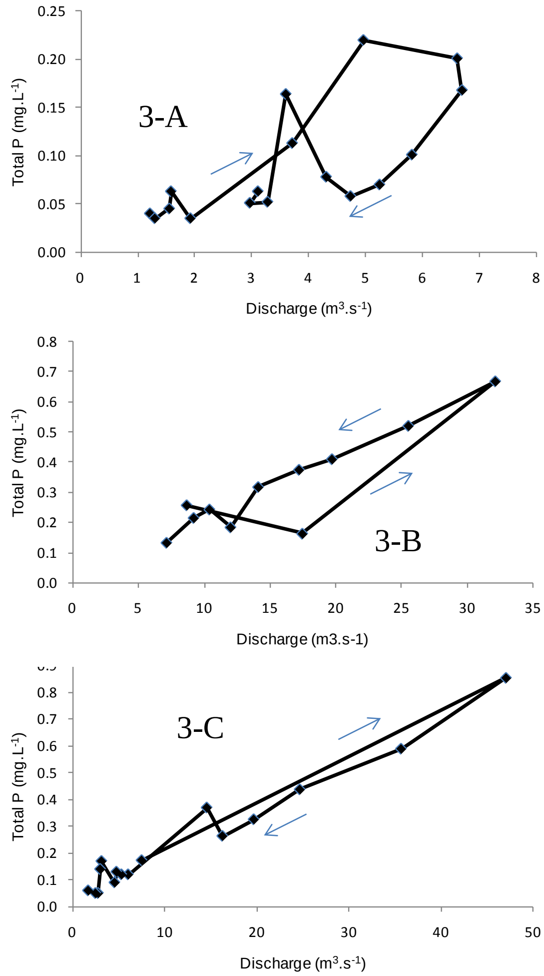


Figure 18. Discharge-Concentration Curves for Total P for storms 3-A, 3-B, and 3-C.



Appendices:

Sample Date	DOC	NO3-N	Total P	TKN	Mg2+	K+	Na+
12/10/07 11:30	3.97398	1.217	NA	NA	11.12	2.193	126.73
12/11/07 0:00	4.44063	2.626	0.0870	0.9070	21.29	2.823	27.87
12/11/07 4:00	4.04346	3.033	0.1000	0.7080	20.72	3.014	24.09
12/11/07 8:00	4.09294	3.46	0.1570	1.2700	20.54	3.119	23.33
12/11/07 12:00	4.45931	3.858	NA	NA	20.18	3.136	21.81
12/11/07 16:00	4.23648	4.078	0.1450	0.8610	20.55	0.2	20.85
12/12/07 0:00	4.77707	4.194	0.2950	1.5600	18.51	2.666	20.37
12/12/07 4:00	6.12968	3.989	0.5390	2.4200	16.59	3.158	17.2
12/12/07 9:15	7.08099	4.425	0.7790	3.0300	13.53	3.607	13.46
12/12/07 16:00	6.6186	4.43	0.4500	2.0200	12.34	0.2	11.51
12/13/07 11:15	5.87585	4.502	0.3160	1.4900	14.52	0.2	13.98
12/13/07 16:00	5.54825	4.726	0.2880	1.8100	15.38	3.422	12.88
12/14/07 4:00	5.96202	4.908	0.2620	1.5100	16.28	3.189	13.73
12/14/07 12:00	5.26446	4.791	0.2620	1.4600	16.76	2.912	14.08

Appendix One. Solute concentration data for storm 1.

Date	DOC	NO3-N	Total P	TKN	Mg2+	K+	NA+
1/8/2008 14:30	6.43	3.02	0.23	1.45	15.46	2.81	19.85
1/8/2008 22:00	6.46	2.10	1.38	3.70	9.17	0.10	12.45
1/9/2008 6:00	NA	NA	0.78	2.69	NA	NA	NA
1/9/2008 14:30	6.70	2.40	0.61	2.43	10.16	4.58	11.57
1/9/2008 20:30	6.97	2.77	0.51	2.25	11.69	4.17	10.57
1/10/08 2:30	6.76	2.95	0.42	1.98	12.82	3.91	11.07
1/10/2008 8:30	6.25	3.22	0.39	1.97	13.58	3.70	11.47
1/10/2008 14:30	5.73	3.33	0.35	1.57	14.50	3.51	13.25
1/14/2008 10:30	4.29	3.58	0.17	1.00	21.75	2.38	16.78

Appendix Two: Solute concentrations for storm 2.

Date	NO3-N	Total P	TKN	Mg2+	K+	NA+
5/7/08 12:00	0.64	0.04	0.70	27.07	2.61	29.17
5/7/08 19:00	0.59	0.04	0.71	26.48	2.65	28.63
5/8/08 0:00	0.57	0.05	0.78	27.06	2.71	28.69
5/8/08 5:00	0.00	0.06	0.65	25.15	2.62	27.90
5/8/08 11:30	0.00	0.04	0.77	25.25	2.73	29.07
5/8/08 16:30	0.67	0.11	1.26	20.56	2.46	25.61
5/8/08 21:30	0.69	0.22	1.51	19.58	2.53	24.46
5/9/08 2:30	0.79	0.20	1.40	21.65	2.50	23.55
5/9/08 7:30	1.28	0.17	1.23	21.98	2.90	21.70
5/9/08 12:30	2.71	0.10	1.22	21.89	2.44	19.34
5/9/08 17:30	3.65	0.07	1.03	21.61	2.20	17.92
5/9/08 22:30	4.01	0.06	0.91	22.37	2.18	18.38
5/10/08 3:30	3.98	0.08	1.17	21.64	2.14	17.99
5/10/08 7:30	4.05	NA	NA	21.67	2.16	18.58
5/10/08 12:30	4.08	0.06	0.91	22.30	2.18	18.34
5/10/08 17:30	4.29	0.05	0.88	21.97	2.05	18.09
5/10/08 22:30	4.14	0.05	0.84	22.26	1.98	18.02

Appendix Three: Solute concentrations for storm 3A.

Date	NO3-N	Total P	TKN	Mg2+	K+	NA+
5/11/08 3:30	3.84	0.06	0.91	21.70	1.92	17.64
5/11/08 8:30	2.53	0.26	1.53	18.17	2.07	18.06
5/11/08 12:15	2.15	0.16	1.10	16.56	2.47	15.66
5/11/08 17:30	4.37	0.67	3.01	14.09	3.13	9.82
5/11/08 22:30	5.62	0.06	2.66	12.45	3.63	8.64
5/12/08 3:30	6.00	0.52	2.30	12.54	3.70	8.31
5/12/08 8:30	6.14	0.41	2.02	13.55	3.41	9.03
5/12/08 11:30	6.19	0.38	2.03	14.34	3.29	9.93
5/12/08 17:30	5.79	0.32	1.75	15.39	2.99	9.76
5/12/08 22:30	5.86	0.19	1.34	16.43	2.88	10.78
5/13/08 3:30	5.92	0.24	1.50	17.18	2.74	10.45
5/13/08 8:30	5.82	0.22	1.31	17.87	2.62	11.67
5/13/08 17:30	5.32	0.13	1.14	18.66	2.43	11.44

Appendix Four: Solute concentrations for storm 3B.

Date	NO3-N	Total P	TKN	Mg2+	K+	NA+
5/14/08 3:30	5.03	0.17	1.01	18.94	2.27	12.10
5/14/08 12:00	2.77	0.86	3.24	11.16	3.26	7.69
5/14/08 17:00	3.63	0.59	2.18	10.61	3.67	7.14
5/14/08 22:00	4.25	0.44	2.14	12.04	3.47	8.42
5/15/08 3:00	4.36	0.33	1.78	12.85	3.13	8.35
5/15/08 8:00	4.72	0.26	1.47	15.41	2.82	9.55
5/15/08 12:00	4.65	0.37	1.56	14.53	2.93	10.02
5/17/08 9:00	2.64	0.12	0.95	19.97	2.22	13.62
5/17/08 19:00	4.52	0.12	1.01	20.74	2.15	13.51
5/18/08 5:00	4.40	0.13	0.83	21.30	2.08	14.83
5/18/08 11:00	3.87	0.09	0.77	22.50	1.97	15.41
5/19/08 19:00	3.70	0.17	0.71	22.85	2.01	16.43
5/20/08 5:00	3.60	0.14	0.56	23.25	1.99	16.69
5/20/08 15:00	3.43	0.05	0.62	23.45	1.96	16.37
5/21/08 11:00	3.14	0.05	0.53	23.88	1.94	17.24
5/26/08 11:30	1.95	0.06	0.57	24.85	2.08	21.58

Appendix Five: Solute concentrations for storm 3C.

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