Beryllium-7 wet deposition variation with storm height, synoptic classification, and tree canopy state in the mid-Atlantic USA

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Abstract
Short-lived fallout isotopes, such as beryllium-7 (^{7}Be), are increasingly used as erosion and sediment tracers in watersheds. Beryllium-7 is produced in the atmosphere and delivered to Earth's surface primarily in precipitation. However, relatively little has been published about the variation in ^{7}Be wet deposition caused by storm type and vegetation cover. Our analysis of precipitation, throughfall, and sediments in two forested, headwater catchments in the mid-Atlantic USA indicates significant variation in isotope deposition with storm type and storm height. Individual summer convective thunderstorms were associated with ^{7}Be activity concentrations up to 5.0 Bq L^{-1} in precipitation and 4.7 Bq L^{-1} in throughfall while single-event wet depositional fluxes reached 168 Bq m^{-2} in precipitation and 103 Bq m^{-2} in throughfall. Storms originating from the continental USA were associated with lower ^{7}Be activity concentrations and single-event wet depositional fluxes for precipitation (0.7 – 1.2 Bq L^{-1} and 15.8 – 65.0 Bq m^{-2}) and throughfall (0.1 – 0.3 Bq L^{-1} and 13.5 – 98.9 Bq m^{-2}). Tropical systems had relatively low activity concentrations, 0.2 – 0.5 Bq L^{-1} in precipitation and 0.2 – 1.0 Bq L^{-1} in throughfall, but relatively high single-event depositional fluxes due to large rainfall volumes, 32.8 – 67.6 Bq m^{-2} in precipitation and 25.7 – 134 Bq m^{-2} in throughfall. The largest sources of ^{7}Be depositional variation were attributed to storm characteristics including precipitation amount and maximum storm height. ^{7}Be activity associated with fluvial suspended sediments also exhibited the highest concentration and variability in summer (175 – 1450 Bq kg^{-1}). We conclude the dominant source of variation on event-level ^{7}Be deposition is storm type. Our results illustrate the complex relationships between ^{7}Be deposition in precipitation and throughfall and demonstrate event-scale relationships between the ^{7}Be in precipitation and on suspended sediment.

1. Introduction

In recent years, beryllium-7 (^{7}Be) has grown in popularity for sediment fingerprinting and erosion tracing studies (Walling, 2012). Attractive due to its natural production and a short half-life (54 d), research applications of ^{7}Be include tracing atmospheric circulation processes (Dibb et al., 2003; Pacini et al., 2011; Usoskin et al., 2009), hillslope erosion (Schuller et al., 2006; Walling et al., 2009), and fluvial sediment budgets and transport from daily to seasonal timescales (Blake et al., 2002; Fisher et al., 2010; Kaste et al., 2014). The processes that control the deposition of ^{7}Be via precipitation to the Earth surface, however, are poorly understood.

Beryllium-7 is produced cosmogenically in the stratosphere and upper troposphere (Lal et al., 1958). Production is greatest at middle and high latitudes and may vary with solar activity (Kaste and Baskaran, 2011). Delivery to the Earth's surface occurs primarily in precipitation rather than dry deposition (Ioannidou and Papastefanou, 2006; Juri Ayub et al., 2009; Kaste et al., 2002). Previous studies have noted seasonal differences in ^{7}Be wet depositional fluxes, usually with higher fluxes in spring (Kim et al., 2000; Olsen and Larsen, 1985), although these seasonal peaks may not be consistent from year to year (Baskaran, 1995). In many cases, particularly when ^{7}Be activities are measured in monthly or semi-monthly bulk precipitation collections, a correlation exists between flux and total rainfall (Caillet et al., 2001; Juri Ayub et al., 2009; Kaste et al., 2002; Salisbury and Cartwright, 2005) to the extent that the flux of ^{7}Be is driven primarily by the amount of precipitation (Baskaran, 1995). Trends in ^{7}Be depositional flux (Bq m^{-2}) are more complicated when examining that flux as the product of ^{7}Be activity concentration (Bq L^{-1}) and total precipitation. Furthermore, variations in activity concentration and precipitation at the event timescale reveal more nuanced relationships between ^{7}Be activity concentration and rainfall. The few studies measuring event-scale variation in ^{7}Be have found activity was greatest at smaller precipitation rates and amounts (Ioannidou and Papastefanou, 2006). These differences may be attributed to sampling interval (Salisbury and Cartwright, 2005), duration of the antecedent dry period (Caillet et al., 2001), and/or rain drop size (Ioannidou and Papastefanou, 2006).

Wet-deposited ^{7}Be readily sorbs to soil mineral grains and organic material (Taylor et al., 2012; You, 1989), leading to the interest in this isotope as a fingerprint or tracer of surface erosion. Differences in ^{7}Be deposition to the soil surface have been attributed to the spatial variability of vegetation interception in grasslands (Kaste et al., 2011) and tree plantations (Ali et al., 2011), but much more research is necessary to determine watershed and landscape-scale relationships. Other fallout isotopes used in erosion tracing, such as ^{137}Cs, have been found to interact with tree canopy such that they initially have lower activity in throughfall than precipitation, which can be released, in part, from the canopy and deposited in subsequent events (Kato et al., 2012).

In order to effectively use ^{7}Be in sediment fingerprinting, the spatial and temporal patterns in its deposition to the ground surface, as well as the connection between the deposited activity, potentially erosive soil, and fluvial sediment must be understood (Walling, 2012). Therefore, the objectives of this study were to (1) quantify event-level ^{7}Be in precipitation and canopy throughfall to determine the effect of canopy vegetation, (2) evaluate the influence of storm type on ^{7}Be with respect to rainfall characteristics and maximum storm heights, and (3) evaluate the event-scale relationship between the catchment input of ^{7}Be in precipitation and output of ^{7}Be associated with suspended sediments. This research was carried out in two md-Atlantic watersheds during a series of precipitation events in 2011-2012, including but not limited to Hurricane Irene (27-28 August 2012) and Hurricane Sandy (28-29 October 2012).

2. Methods

2.1 Study Sites

We studied two forested field sites in the mid-Atlantic piedmont associated with the Christina River Basin Critical Zone Observatory (CRB CZO) (Figure 1). The research site at Fair Hill Natural Resources Management Area (FH), a first-order tributary to the Big Elk Creek (in turn a tributary to the Chesapeake Bay), is located in northeastern Maryland (39°42′N, 75°50′W). The plot is located within a 12 hectare forested catchment with a stand density of 225 trees ha^{-1}, a stand basal area of 36.8 m^2 ha^{-1}, a mean diameter at breast height (dbh) of 40.8 cm, and a mean tree height of 27.8 m. The forest canopy is dominated by *Liriodendron tulipifera* L. (yellow poplar) and *Fagus grandifolia* Ehrh. (American beech), with *Acer rubrum* L. (red maple) and *Quercus alba* L. (white oak) also present. The forest canopy has a leaf area index (LAI) of 5.3 m^2 m^{-2}. The dominant canopy trees are approximately 80-100 years old.

The research site at Boulton Run (BR), a first-order tributary to the East Branch of the White Clay Creek (one of several major catchments within the Christina River Basin), is located in southeastern Pennsylvania (39°51′N, 75°47′W). The 15 hectare BR catchment is 60% forested. Similar to FH, dominant canopy species include *L. tulipifera*, *F. grandifoli*a, *A. rubrum,* and *Q. alba*. The dominant canopy trees are approximately 55-75 years old, with older trees in the riparian area adjacent to the stream. The canopy is approximately 69% closed and has a LAI of 4.3 m^2m^{-2}. The upper 40% of the catchment, above the channel headcut, is agricultural in a mixture of row crops (25%) and permanent pasture (15%).

Mean 30-year total annual precipitation is approximately 1200 mm, with an average of 523 mm winter seasonal snowfall and the rest falling as rainfall with little annual variation (MD State Climatologist Office 2013). The wettest season is autumn (320 mm), followed by summer (314 mm), spring (308 mm), and winter (262 mm). Frontal precipitation patterns are typical for fall, winter, and spring; convective precipitation events dominate the summer. It is necessary to distinguish between storm types because convective storm systems have a larger vertical extent than the typical mid-latitude cyclone and can gain access to levels of the troposphere where ^{7}Be concentrations are naturally greater (Lal et al., 1958).

2.2 Hydrometeorologic Monitoring

For the Fair Hill site, data were obtained from a Delaware Environmental Observing System (DEOS) station located in a nearby clearing 0.5 km southwest of the research site. The DEOS station monitored ambient weather conditions and rainfall characteristics at 5-minute intervals and provided intra-storm information regarding precipitation magnitude and intensity, storm duration, and antecedent dry period. For the Boulton Run site, hourly precipitation data were retrieved from the US Climate Reference Network (USCRN) Avondale, PA station located 0.25 km west of the research site. For both sites, maximum storm height was obtained from Mosaic 3D Radar Reflectivity cross sections (NOAA NMQ, http://nmq.ou.edu/). Storm types were identified from the National Center for Environmental Prediction Daily Surface Weather Maps (NOAA NCEP, http://www.hpc.ncep.noaa.gov/dailywxmap/). Maximum storm height was used to parameterize the atmospheric extent of accessible ^{7}Be, assuming larger vertical cloud profiles preferentially gain access to upper tropospheric source regions of ^{7}Be production.

Throughfall hydrologic flux was measured at Fair Hill with eight TE-525MM tipping-bucket gages randomly located under a beech-poplar canopy (Texas Electronics, Dallas, Texas, USA). The TE-525MM has an accuracy of ±1% at precipitation rates up to 2.54 cm h^{-1}. All data were quality checked through manual determination and throughfall gauges that registered clogs were removed from analysis. Throughfall regression models (eq. 1-2) were constructed from 210 individual rainfall events ranging in magnitude from 3.1 mm to 155.5 mm and represent all phases of canopy foliation. Stand age and species composition were similar at both sites, therefore these data were used to construct regression equations to calculate throughfall at Boulton Run, where hydrologic flux instrumentation was not deployed.

During the growing season when leaves are present in the canopy, throughfall flux in a beech-poplar canopy can be estimated by

$$TF_{leaf} = 0.95\ P - 3.58 \qquad (1)$$

where *P* is precipitation in mm (R^2=0.96). During the dormant season when leaves are absent, throughfall flux can be estimated by

$$TF_{leafless} = 0.85\ P - 0.21 \qquad (2)$$

(R^2=0.98). Regardless of season, the forest canopy inhibits the movement of precipitation to the forest floor and this discrepancy in hydrology flux must be taken into consideration when comparing ^{7}Be across hydrologic compartments.

2.3 ^{7}Be Sample Collection and Analysis

Beryllium-7 samples were collected in rainfall and canopy throughfall during individual events from August 2011 through October 2012 at each of the two sites. Acid-washed polyethylene bottles with 20.3 cm diameter funnels were placed on platforms approximately 1 m above the ground surface. At each study site ^{7}Be was monitored with a randomly located throughfall collector beneath the forest canopy and a separate collector located in a nearby clearing for precipitation.

Samples were retrieved after the conclusion of a storm event (e.g. within 24-48 hours). Upon retrieval, the funnels were rinsed with dilute trace-metal grade hydrochloric acid and the rinses added to the sample. Beryllium-7 was extracted from the precipitation samples using ion-exchange resins (Jungck et al., 2009; Komura et al., 2006). Samples received one of two resin treatments, based on availability, either (1) Amberlite MB-3 (combined cation and anion) or (2) POWDEX–PCH (cation) and –PAO (anion) exchange resins. Treatment 1 consisted of a single product, Amberlite MB-3, with the capacity for both anion and cation exchange. The cation exchange portion within the Amberlite MB-3 is a sulfonated polystyrene type styrene divinylbenzene co-polymer with a vendor-provided exchange capacity of 1.80 meq mL^{-1}. The anion portion within the Amberlite MB-3 is comprised of a benzuldimethyl (2-hydroxyethyl) ammonium functional group with a vendor-provided exchange capacity of 1.25 meq mL^{-1} by wetted bed volume. We determined the exchange capacity of Amberlite MB-3 to be 1.0 – 1.5 meq g^{-1} for measured wetted-bed volumes ranging between 0.80 and 0.84 mL g^{-1}. Treatment 2 consisted of the addition of separate materials for the exchange of cations and anions. The cation exchange –PCH is a sulfonated copolymer of styrene and divinylbenzene in the hydrogen form, while the anion –PAO is a trimethylamine functionalized copolymer of styrene and divinylbenzene in the hydroxinde form. Each POWDEX resin, -PCH and –PAO, contains 4.8 meq g^{-1} of exchange capacity. Approximately 12 g of exchange resins were added for each 1 L of precipitation to ensure ample exchange capacity for removing all ions in the precipitation samples, including ^{7}Be presumed to be in the Be^{2+} form. Upon addition, samples were agitated for a minimum of 10 minutes.

Prior to extraction, a spike of stable beryllium, in the form of $^9BeSO_4$, was added to each sample in order to evaluate the recovery of beryllium on the exchange resins (Canuel et al., 1990). A 15 mL aliquot of supernatant was kept following resin extraction process, which was subsequently analyzed for beryllium via Inductively Coupled Plasma Mass Spectrometery (ICP MS). Beryllium concentrations in the aliquots normalized to initial concentration in the spiked sample prior to resin extraction ranged from 0.00 to 0.49, with an average of 0.32 (+/- 0.05 standard deviation) for the Amberite resin. The POWDEX resin treatment relative recovery varied between 0.00 and 0.25 with an average of 0.01 (+/- 0.05 standard deviation). It was presumed that the balance of the beryllium not measured in the aliquot was bound to the exchange resin. Recovery amounts of stable beryllium spike were used to correct the ^{7}Be activity concentration of precipitation and canopy throughfall, such that the recovery of the ^{9}Be was presumed to be in proportion to the recovery of the ^{7}Be, as the binding activity of the isotopes has been found equivalent (Taylor et al., 2012). Additional quality control was provided by performing a second resin treatment on 10% of the analyzed samples, selected randomly. In this case, following the initial resin treatment, the supernatant was retained and a subsequent resin treatment was performed on the supernatant following the above protocol. In all cases, there was no measurable activity of ^{7}Be on the samples resulting from the second resin treatment.

After each resin treatment, the exchange resins were sealed in 90.5 mL (5.8 cm diameter, 3.2 cm height) aluminum cans for analysis on high-purity germanium gamma ray spectrometers (Canberra Model 6L2020R Low Energy Germanium LEGe Detectors). Each sample, including the second extraction samples for quality control, was counted for 24-48 hours within 108 days of rainfall collection. ^{7}Be peak area, centered at 477 keV, was computed. Activities (Bq L^{-1}) and associated standard deviations were computed for each sample based on the protocol of (Gilmore, 2008). Detector efficiency was determined from counting a mixed isotope standard (Eckert & Ziegler Analytics, Atlanta, GA) and the efficiency at the ^{7}Be peak range was calculated to be 1.4% based on a cubic interpolation of efficiencies measured at reference peaks (Lead-210 at 46.5 keV, Americium-241 at 59.5 keV, Cadmium-109 at 88.0 keV, Cobalt-57 at 122.1 keV and Cesium-137 at 661.7 keV) using Canberra GENIE 2000 software.

2.4 Sediment-associated ^{7}Be flux

Stream water samples were collected from White Clay Creek (Figure 1), the receiving stream of Boulton Run, between August 2011 and October 2012. Suspended sediments were isolated by settling from 210 L bulk stream water samples, oven dried at 60 °C, and sealed in petri dishes (8.9 cm diameter, 1.3 cm height) for analysis on high-purity germanium gamma ray spectrometers (Ortec GEM-50195 Model, 38-P21174B) at the University of Exeter. The detector efficiency of 3.37% for ^{7}Be was determined by interpolation of efficiencies measured at reference peaks for Lead-210 (46.5 keV) and an 8-isotope mixed standard (Barium-133 at 81 keV and 356 keV, Cobalt-57 at 122 keV, Cerium-139 at 166 keV, Strontium-85 at 514 keV, Cesium-137 at 662 keV, Manganese-54 at 835 keV, Yttrium-88 at 898 keV, and Zinc-65 at 1116 keV) (QSH Global GmbH, Braunschweig, Germany) using Ortec Maestro software.

Stream discharge and total suspended solids (TSS) data were obtained for White Clay Creek from the Christina River Basin Critical Zone Observatory (Figure 1) (Aufdenkampe et al., 2012; Stroud Water Research Center, 2012). We used a linear interpolation between TSS measurements to determine TSS for the time of each discharge observation during the events. Event total suspended solid flux was computed as the sum of the product of discharge and total suspended solids for each 5-minute time interval throughout the event. Storm total sediment-associated ^{7}Be flux was calculated as the product of sediment-associated ^{7}Be activity concentration and event total suspended solid flux. In the case that more than one sediment-associated ^{7}Be concentration measurement was taken over the course of the event, the event hydrograph was subdivided temporally and the ^{7}Be sediment-associated load was computed for each portion of the event. The portions were then summed to compute the event total sediment-associated ^{7}Be flux.

2.5 Statistical Methods

The product of ^{7}Be activity concentration (Bq L^{-1}) and total precipitation or canopy throughfall hydrologic flux (mm) were used to calculate the total activity deposited over a reference area during each event, herein referred to as the ^{7}Be wet deposition flux (Bq m^{-2}).

Statistical relationships between both the activity concentration (Bq L^{-1}) and wet deposition flux (Bq m^{-2}) in precipitation and canopy throughfall were examined in both the leaf-on and leaf-off seasons using a one-way Analysis of Variance (ANOVA) with Tukey contrasts to test for significance between the means of groups based on season and canopy state. Leaf-on and leaf-off seasons were defined based on our observations and correspond to samples taken in April through September and October through March, respectively. Four groups resulted from this definition: (1) precipitation in the leaf-on season, (2) precipitation in the leaf-off season, (3) deciduous forest canopy throughfall in the leaf-on season, and (4) deciduous forest canopy throughfall in the leaf-off season. We also evaluated the difference in ^{7}Be activity concentration and wet deposition flux associated with precipitation and canopy throughfall in calendar-based seasons: DJF (December, January, February), MAM (March, April, May), JJA (June, July, August) and SON (September, October, November). We compared precipitation and throughfall to each other across seasons. We also compared precipitation to throughfall within individual seasons. Additional ANOVA models were constructed to examine the difference in mean ^{7}Be activity concentration and wet deposition flux in precipitation across storm types, classified based on National Center for Environmental Prediction Daily Surface Weather Maps. All statistical differences between group means were evaluated at the α = 0.05 level of significance. Patterns were also compared between ^{7}Be deposition and the maximum cloud height of storms. For statistical analysis, one event (rainfall on 10 August 2012) was removed from all evaluation due to insufficient sample size and acidification of the sample collector.

3. Results

From August 2011 through October 2012, 19 precipitation events were sampled – 10 events at Boulton Run and 9 events at Fair Hill. Three of these events were sampled at both sites, while the remaining 16 were sampled at only one of the two sites (Table 1). These events represent storms across all four seasons, with total precipitation ranging from 7.0 mm to 167.9 mm. Three storms were categorized as strong cold fronts, 5 were categorized as convective summertime thunderstorms, 4 were categorized as Low Pressure systems originating from the Midwest, 4 were categorized as Low Pressure systems originating from the Great Lakes, and 2 were tropical cyclones (Hurricane Irene and Hurricane Sandy) (Table 1). Convective thunderstorms were associated with the tallest storm heights resulting from intense summertime surface heating and corresponding atmospheric instability.

Within the 19 event collections, the ^{7}Be activity concentration in open precipitation varied from 0.2 – 5.0 Bq L^{-1} and in canopy throughfall from 0.2 – 4.7 Bq L^{-1} (Table 1). Wet deposition flux from open precipitation ranged from 4.6 Bq m^{-2} to 177.5 Bq m^{-2} while wet deposition flux from canopy throughfall ranged from 11.0 Bq m^{-2} to 134.2 Bq m^{-2} (Table 1). Open precipitation activity concentrations were not significantly different between canopy phases (leaf on: 1.98 Bq L^{-1}, leaf off: 0.88 Bq L^{-1}, $p = 0.063$) nor were throughfall activity concentrations (leaf on: 1.50 Bq L^{-1}, leaf off: 0.85 Bq L^{-1}, $p = 0.134$) (Figure 2a). Open precipitation wet deposition flux was significantly different between canopy phases (leaf on: 90.49 Bq m^{-2}, leaf off: 68.09 Bq m^{-2}, $p = 0.035$) as was throughfall total wet deposition flux (leaf on: 77.58 Bq m^{-2}, leaf off: 26.23 Bq m^{-2}, $p = 0.012$) (Figure 2b). Precipitation and throughfall activity concentration did not differ from each other significantly within a single canopy phase (leaf on: $p = 0.885$, leaf off: $p = 0.433$) (Figure 2a), nor did the wet deposition flux (leaf on: $p=0.559$, leaf off: $p=0.165$) (Figure 2b).

At the seasonal scale, precipitation activity concentration ($p = 0.134$), throughfall activity concentration ($p = 0.477$), and throughfall wet deposition flux ($p = 0.110$) were not significantly different between seasons (Figure 2c-d). A more robust pairwise ANOVA comparison of precipitation wet deposition flux (Holm and Bonferroni adjustments) reveals no significant difference in precipitation wet deposition flux between seasons (JJA-SON: $p=0.160$, JJA-DJF: $p=0.120$, JJA-MAM: $p=0.180$, SON-DJF: $p=1.000$, SON-MAM: $p=1.000$, DJF-MAM: $p=1.000$). Within each season, precipitation and throughfall activity concentration only differed significantly in MAM ($p=0.0388$) (Figure 2c). Activity concentration did not significantly differ between precipitation and throughfall within any other single season (JJA: p=0.398, SON: p= 0.846) (Figure 2c). Wet deposition flux did not significantly differ between precipitation and throughfall in any single season (MAM: p=0.330, JJA: p=0.463, SON: 0.734) (Figure 2d). The DJF season only had a single throughfall collection, so we do not report ANOVA comparisons between precipitation and throughfall within this season.

Events within the JJA (summer) season exhibited the largest range in precipitation activity concentration and wet deposition flux as well as the most diverse group of storm types. The largest open precipitation activity concentration (5.0 Bq L^{-1} on 18 August 2011) and wet deposition flux (177 Bq m^{-2} on 13 August 2011) were observed during summer convective thunderstorms (Table 1, Figure 3). The largest throughfall activity concentration (4.7 Bq L^{-1}) was also measured during the summer convective thunderstorm on 18 August 2011 although the largest throughfall wet deposition flux (159 Bq m^{-2}) was observed during Hurricane Irene (27 August 2011) (Table 1, Figure 3). Conversely, open precipitation activity concentrations (0.69 Bq L^{-1} on 21 November 2011) and wet deposition flux (7.46 Bq m^{-2} on 27 January 2012) were relatively low during low pressure systems of continental origin in non-summer seasons. Activity concentrations were also low during tropical systems (e.g. hurricanes), however the wet deposition flux was high due to the large amount of precipitation during these events (Table 1, Figure 3). Statistically significant differences do exist between open rainfall activity concentrations ($p < 0.001$) and wet deposition flux ($p = 0.003$) during different event types (Figure 3), which can be quantitatively described by maximum storm height in the atmosphere (Table 1, Figure 3). Pairwise comparisons of storm types indicate statistically significant differences in activity concentration between summer convective thunderstorms (JJA-CTS) and continental-origin low pressure systems in DJF, MAM, SON as well as and tropical systems/hurricanes (H) (Table 2). Significant differences in activity concentration were also observed between tropical systems/hurricanes (H) and both MAM and SON low pressure systems, respectively (Table 2). Non-summer continental low pressure systems (*i.e.*, DJF, MAM, SON) did not exhibit statistically significant differences in ^{7}Be activity concentrations. On average, throughfall ^{7}Be activity concentration was largest in JJA-CTS storms (2.18 Bq L^{-1}) while throughfall ^{7}Be wet deposition flux was largest in H (102.09 Bq m^{-2}) and JJA-CTS (71.33 Bq m^{-2}). However, neither activity concentration (p = 0.308) nor wet deposition flux (p = 0.284) in throughfall were significantly different across storm types. Within individual storm types, activity concentration in precipitation and throughfall differ significantly only in MAM (p=0.0388), while the wet deposition flux of ^{7}Be in precipitation and throughfall differs only in convective thunderstorms (JJA-CTS: p=0.0418) (Table 3).

4. Discussion

4.1 Canopy Influence on Deposition

We did not find a statistically significant difference in the activity concentration (Bq L^{-1}) of ^{7}Be between open precipitation and canopy throughfall as a function of canopy leaf phase, although statistically significant differences were observed in wet deposition flux for both precipitation and throughfall (Figure 2). The variability of activity concentration in precipitation increased during the leaf-on canopy phase (Figure 2a), a trend which was localized to events observed in JJA (Figure 2c) and further localized to convective thunderstorms (JJA-CTS) within the JJA season (Figure 3), suggesting the high concentration of ^{7}Be observed during the summer precipitation events is derived from convective storm processes rather than the direct influence of vegetation.

The variability of throughfall activity concentration exhibited no response to canopy leaf phase or season (Figure 2a-c). Conversely, the variability of both precipitation and throughfall ^{7}Be wet deposition flux increased during the leaf-on canopy phase (Figure 2b). At the seasonal scale, both precipitation and throughfall wet deposition fluxes exhibited peak variability during JJA, and smaller yet comparable variability during adjoining seasons (SON and MAM), and the smallest variability during DJF (Figure 2d). The trend of increased variability beginning in MAM and continuing through SON suggests that ^{7}Be deposition flux, especially in throughfall, is controlled by a combination of both seasonality of storm types and canopy leaf phase. Quite possibly, the transition from leafless canopy in early MAM to full canopy in JJA and senescence in SON resulted in hydrologic differences in throughfall quantity (Kramer and Holscher, 2009) and the occurrence of convective and non-convective storms during JJA both led to the observed increase in variability of deposition fluxes.

Examining individual events, throughfall ^{7}Be activity concentration was not consistently higher or lower than the ^{7}Be activity concentration in precipitation (Table 1). This was unexpected, as interception by vegetation canopies has been found to consistently reduce ^{7}Be (Ali et al., 2011). Furthermore, ^{7}Be has been measured on plant material (Kaste et al., 2011) and canopy interception of incident rainfall is the presumed source. However, the presence of a vegetation canopy did not consistently reduce ^{7}Be concentration or flux in throughfall compared to precipitation during all observed events in our study (Table 1). For example, during the September 23, 2011 event sampled at both sites, canopy throughfall had both a higher ^{7}Be activity concentration and wet deposition flux. This could be due to the wash-off of dry ^{7}Be deposition occurring on the surface of the leaves since the previous rainfall event or residual ^{7}Be retained by the canopy from a previous event and subsequently washed from the canopy, as has been observed for other short-lived fallout isotopes (Kato et al., 2012). An alternate hypothesis for increased ^{7}Be throughfall wet deposition flux in SON is related to leaf senescence. Similar to ^{137}Cs, which has been suggested to originate partially from the forest canopy itself (Kato et al., 2012), it is possible that ^{7}Be also may be leached from the plant surfaces. In fact, prior work at this site has shown that Ca^{2+}, Mg^{2+}, and K^{+} increased in throughfall leaching fluxes from senescing leaves in SON while SO_4^{2-} and K^{+} increased in throughfall leaching fluxes from emerging leaves in MAM (Van Stan et al., 2012). Coupled with the frontal-based precipitation that dominates autumnal rainfall events at the study site, leaching from the forest canopy would be most enhanced via saturating, long duration events. More individual event collections of throughfall in the SON season will allow for further testing of these possible mechanisms.

The sequence of the individual events allows for the temporary detention of ^{7}Be in the tree canopy in any leaf-on season, which can be subsequently released in a large and/or intense event. For example, the series of events recorded in Boulton Run in August 2011 has a sequence of three events followed by Hurricane Irene (Table 1). The three preceding events all had higher ^{7}Be activity concentration and wet deposition flux in precipitation than Hurricane Irene (Table 1). The first two of these three events (13 August 2011 and 18 August 2011) had higher concentrations and wet deposition fluxes in precipitation than throughfall (Table 1). During the third, the throughfall sampler was inoperable. Throughfall during Hurricane Irene had both more concentrated ^{7}Be and an overall higher wet deposition flux of ^{7}Be than precipitation. This could have been due to the extreme and intense nature of Hurricane Irene causing the release of any temporarily stored ^{7}Be in the canopy on leaf and branch surfaces. Hurricane Sandy, however, did not exhibit this same trend despite it also being an extreme event. The throughfall ^{7}Be wet deposition flux was less than that of the open precipitation (Table 1). Perhaps this is due to the fact that Hurricane Sandy took place late in the autumn rather than in the summer and brought less total rainfall over a longer duration than Hurricane Irene. We hypothesize the combination of preceding events and rainfall intensity caused the differences in precipitation and throughfall activity concentrations and deposition fluxes between Hurricanes Irene and Sandy. The series of convective thunderstorms leading up to Hurricane Irene brought some of the highest measured precipitation ^{7}Be activity concentrations (Table 1), followed by Hurricane Irene, characterized by rainfall conditions that were capable of saturating the forest canopy and washing out any residual ^{7}Be held in the canopy thereby increasing throughfall activity concentration and wet deposition flux. In contrast, Hurricane Sandy was not preceded by convective thunderstorms. Events typical of the autumn (e.g. SON in Table 1) have significantly lower ^{7}Be activity concentrations and deposition fluxes than the summer convective thunderstorms (JJA-CTS) (Table 2, Figure 3), hence the canopy was not likely to have stored the same levels of ^{7}Be prior to Hurricane Sandy.

4.2 Individual storm radioactivity concentration and flux

Storm type, and its atmospheric vertical extent, is the predominant factor corresponding to differences in wet ^{7}Be deposition observed over the course of our study. This variation is particularly notable during the summer (Figure 3), when precipitation can occur in association with strong convective systems, which have relatively high maximum cloud heights (13.5 – 15.5 km) compared to events during other seasons. Due to the cosmogenic origin, ^{7}Be production is higher in the stratosphere and upper troposphere (Lal et al., 1958), hence storms reaching higher into the atmosphere preferentially gain access a larger source of this isotope.

For ^{7}Be wet deposition flux in precipitation, summer convective thunderstorms were significantly different from all other continental low pressure systems in DJF, MAM, SON and tropical systems (Table 2). However, no significant differences were observed between non-summer storms or tropical systems (Table 2). No significant pairwise comparisons exist between storm types for aerial deposition flux. Maximum storm height can be used to separate summertime convective precipitation and tropical cyclonic activity from events that occur at other times of the year. In mid-latitudes, the troposphere shrinks during the winter and expands in vertical extent during the summer (Hoinka, 1998). Storms that occur in the winter are limited in vertical extent by the lower troposphere, but are also limited by available energy to develop into towering systems. Storms that occur in the summer are able to reach higher vertical extent because of the expanded troposphere, but also because there is more available potential energy in the system from enhanced surface heating and convection).

In the mid-Atlantic region of the United States, rainfall events fall into several different synoptic categories. From the months of September through May, mid-Atlantic precipitation is typically dominated by frontal systems and extratropical cyclones. Extratropical cyclones develop at the interface of converging air masses and are fueled by frontal-based instability and an increase in pressure gradient surrounding the center of low pressure. Frontal lift and storm intensity is dictated by the degree of difference between the intersecting air masses. Conversely, summertime precipitation is typically dominated by localized convective thunderstorms. Summertime thermal instability at the surface creates towering cumulonimbus clouds that reach the upper troposphere. At these heights, summertime thunderstorms have access to atmospheric layers at greater altitude than normal frontal-based storm systems.

While the majority of our summertime (JJA) events were convective thunderstorms, not all were, such as the Low Pressure event originating from the Great Lakes on June 12, 2012 and the tropical cyclone, Hurricane Irene, on August 29, 2012 (Table 1). A seasonal combination of all events in the JJA season shows a large variation in ^{7}Be activity concentration and wet deposition flux (Figure 2c-d). However, this variation narrows and trends emerge when separating the summertime events by storm type. The non-convective JJA event had lower ^{7}Be activity concentration and wet deposition flux than the JJA-CTS events. In fact, the ^{7}Be activity concentration and wet depositional flux of the non-convective JJA event were more similar to that of other low pressure events in other seasons than to other JJA-CTS events (Figure 3). The tropical systems (H) also had lower ^{7}Be activity concentration but similar depositional flux to the convective thunderstorms (Figure 3).

Other researchers have noted seasonal variation in ^{7}Be wet deposition flux and they attribute these seasonal patterns to the narrowing of the troposphere in spring and increased stratospheric-tropospheric exchange (Ali et al., 2011; Lozano et al., 2011). Few have posited this variation could be due to other atmospheric factors that also vary between seasons, such as cloud height (Baskaran, 1995), air mass source or trajectory (Sanders et al., 2011), or amount of precipitation falling as snow (Ishikawa et al., 1995). In addition to seasonal differences, total precipitation (Conaway et al., 2013; Lozano et al., 2011) and time since previous precipitation (Ali et al., 2011; Caillet et al., 2001) have also been proposed as drivers of variation in ^{7}Be deposition. With the exception of very few studies (Caillet et al., 2001; Conaway et al., 2013; Ishikawa et al., 1995), the vast majority of these observations have been made on monthly, not individual event-level, data, hence they were able to note seasonal patterns but not differences within seasons based on other factors. This could be due to either the homogenization over the course of a month and/or other study locations not receiving the same variety of storms as the mid-Atlantic USA.

With the exception of extreme events such as tropical cyclones, our data indicate maximum storm height is the best predictor of event-level ^{7}Be variation, particularly in the JJA season. Based on our data from non-extreme events, we estimate a linear relationship between ^{7}Be activity concentration and storm height in open rainfall (^{7}Be = 0.4867*height – 3.9388, R^2 = 0.66) (Figure 4a). A similar relationship exists between ^{7}Be wet deposition flux and storm height (^{7}Be =19.629*height – 154.99, R^2 = 0.63) (Figure 4b). However, a threshold relationship may exist whereby ^{7}Be activity in precipitation is relatively stable in storm events below 12 km and begins to increase when storm heights exceed 12 km. The nature of the relationship between storm height and ^{7}Be activity concentration and wet deposition flux will only become clearer with more data, but our results suggest that the variation we see in ^{7}Be concentration and deposition flux in non-extreme events corresponds with storm height. Similar but weaker relationships exist for throughfall activity concentration (^{7}Be = 0.2838*height – 1.9664, R^2 = 0.35) (Figure 4c) and throughfall wet deposition flux (^{7}Be = 9.6858*height – 66.466, R^2 = 0.41) (Figure 4d).

This is the first study we know to report ^{7}Be activity concentration and wet deposition flux in relation to storm type. Several other studies (Caillet et al., 2001; Conaway et al., 2013; Ishikawa et al., 1995) have monitored ^{7}Be activity on the same temporal resolution as ours. Our data fall within the range of other studies reporting on a storm-by-storm basis (Figure 5). As a whole, these data show a trend of decreasing ^{7}Be activity concentration with increasing event precipitation. However, the range of variability is 10-fold for low-rainfall events (e.g. < 25 mm) and decreases as rainfall increases. Storm type and corresponding maximum storm height explain the variation in activity concentration in our study. The mid-Atlantic is situated at the convergence of dry continental air masses from the west and moist maritime air masses from the south (Gulf of Mexico) and the east (Atlantic Ocean), which contribute to uniquely variable weather patterns (Brook et al., 1995; Davis et al., 1993; Greene et al., 1999; Yarnal and Frakes, 1997). Additional research that incorporates the effects of storm type on precipitation characteristics and ^{7}Be chemistry will clarify the relationship between activity concentration and rainfall depth. These data will be valuable in other locations in addition to the mid-Atlantic United States.

Hurricanes, such as Irene (29 August 2011) provide a notable exception to our observed correlation between summertime maximum storm height and increased ^{7}Be activity concentration. They are also the only type of storm in which a relatively low activity concentration corresponds to a relatively high deposition flux (Figure 3b and c). While Hurricane Irene had a relatively high cloud height (14.5 km), its precipitation contained very little measureable ^{7}Be. A similar pattern was noted with typhoon remnants on the central

California coast (Conaway et al., 2013). We hypothesize this was due to the long duration and tropical origin of the hurricane.

Despite our individual event activity concentrations falling within the reported range of values (Figure 5), the large between-event variation, particularly in JJA, does not follow patterns noted elsewhere in the literature. For example, we do not see the progressively lower ^{7}Be concentrations in sequential events that have been found elsewhere (Ishikawa et al., 1995). In our study, the series of events in August 2011 does not demonstrate a decreasing activity concentration or aerial deposition with the progression of events, nor does their concentration appear to be a factor of event total rainfall (Table 1). We hypothesize that during this period of sequential convective precipitation events in August 2011, ^{7}Be concentrations were continuously replenished by high altitude storm heights that had access to upper tropospheric source regions of ^{7}Be.

The results of our study offer confirmation, along with several recent studies, that ^{7}Be wet deposition is much more complicated than originally believed. While precipitation magnitude may still influence ^{7}Be wet deposition flux (Baskaran, 1995), additional factors including season, individual storm height, antecedent conditions, and vegetation presence influence ^{7}Be activity concentration and wet deposition flux. Within seasons, variation exists which could be related to the series and types of individual storm events.

4.3 Implications for Sediment Fingerprinting

In recent years, ^{7}Be has increasingly been used for determining the recent source and/or transit time of fluvial sediment (Kaste et al., 2014; Walling, 2012). While the main focus of this study was the variation in ^{7}Be wet deposition, our study is connected to broader critical zone research concerning the source and transport of fluvial sediments. Through these linked interdisciplinary studies, we can evaluate the influence of ^{7}Be wet deposition on the use of this isotope as a fingerprint for sediment source determination and dating.

Within the sediment fingerprinting community, the spatial uniformity of fallout is considered to be one of the open questions regarding the use of ^{7}Be as a tracer (Taylor et al., 2013; Walling, 2012). The spatial distribution in rainfall is cited as a cause for the spatial heterogeneity in ^{7}Be wet deposition (Taylor et al., 2013). Our study affirms that spatial heterogeneity in ^{7}Be depositional flux is due to the spatial heterogeneity in rainfall fluxes, rather than differences in the ^{7}Be activity concentration within the rain when the same events are experienced over the entire study area. Spatial variability of ^{7}Be deposition was observed in this study insomuch that a large degree of spatial variability is associated with summertime convective storm systems. These storms are notoriously patchy, and as illustrated by our data, can produce large isolated quantities of precipitation at locations in close proximity to one another. For example, our two sites did not experience the same JJA-CTS events. In the case of other more spatially uniform events, such as Hurricane Irene or fall and spring low pressure systems, our study indicates no significant difference in activity concentration (precipitation: $p = 0.969$, throughfall: $p=0.809$) or wet deposition flux (precipitation: $p = 0.937$, throughfall: $p=0.633$) of ^{7}Be between our study sites when the same events were measured at both sites (20 km apart). Understanding this variation can inform future ^{7}Be deposition monitoring studies, particularly those associated with sediment fingerprinting, such that monitoring targets the spatial and temporal variation in precipitation as the main driver of the heterogeneity in ^{7}Be wet deposition flux.

The overriding variation in deposition that we saw was between storms of different types, corresponding to different atmospheric cloud heights. The storm-to-storm variation we saw in the precipitation activity concentration is mirrored on the suspended sediment exported from the watershed. In general, storms with higher ^{7}Be activity concentration in the rainfall have higher activity concentration on the stream suspended sediment (Tables 1 and 4, Figure 6a). There is no correlation, however, between ^{7}Be fluxes in suspended sediments vs. in wet deposition (Tables 1 and 4, Figure 6b) and the ratios of the two show that less than 6% of the ^{7}Be wet depositional flux entering a watershed is exported during that same storm. We therefore hypothesize that suspended material exiting the watershed might be newly tagged from direct precipitation onto the channel or the near channel area during individual events. Regardless, our data from a limited number of events indicates a correlation between the ^{7}Be activity concentration entering the watershed in precipitation and that exiting the watershed on suspended sediment during individual events. A correlation between seasonal ^{7}Be deposition in rainfall and on coarse fluvial sediments has been found in New England and attributed to the delivery of ^{7}Be directly to the fluvial system by summer convective thunderstorms (Kaste et al., 2014). These results highlight the need to understand the temporal patterns in watershed input of the isotopic fingerprint relative to the watershed export of suspended sediment at the event timescale.

5. Conclusion

Event-scale variation exists in the activity concentration and deposition of ^{7}Be in precipitation and consequently in canopy throughfall. With the exception of extreme events, higher ^{7}Be deposition corresponds to storms reaching higher in the atmosphere, as determined by maximum cloud height. Even for throughfall, this variation predominates any variation in deposition due to canopy leaf phase. Our results indicate the storm type, particularly in the summer months, is the main source of variation in ^{7}Be activity concentration and wet deposition flux. Sediment fingerprinting studies using ^{7}Be as an indicator of stream sediment source need to be aware of and account for this variation in input to the watershed in order to interpret the catchment export of this radionuclide particularly on the storm-event timescale. Future sediment fingerprinting studies can use this information in planning precipitation monitoring for ^{7}Be and hydrologic flux along with suspended sediment sampling during individual events.

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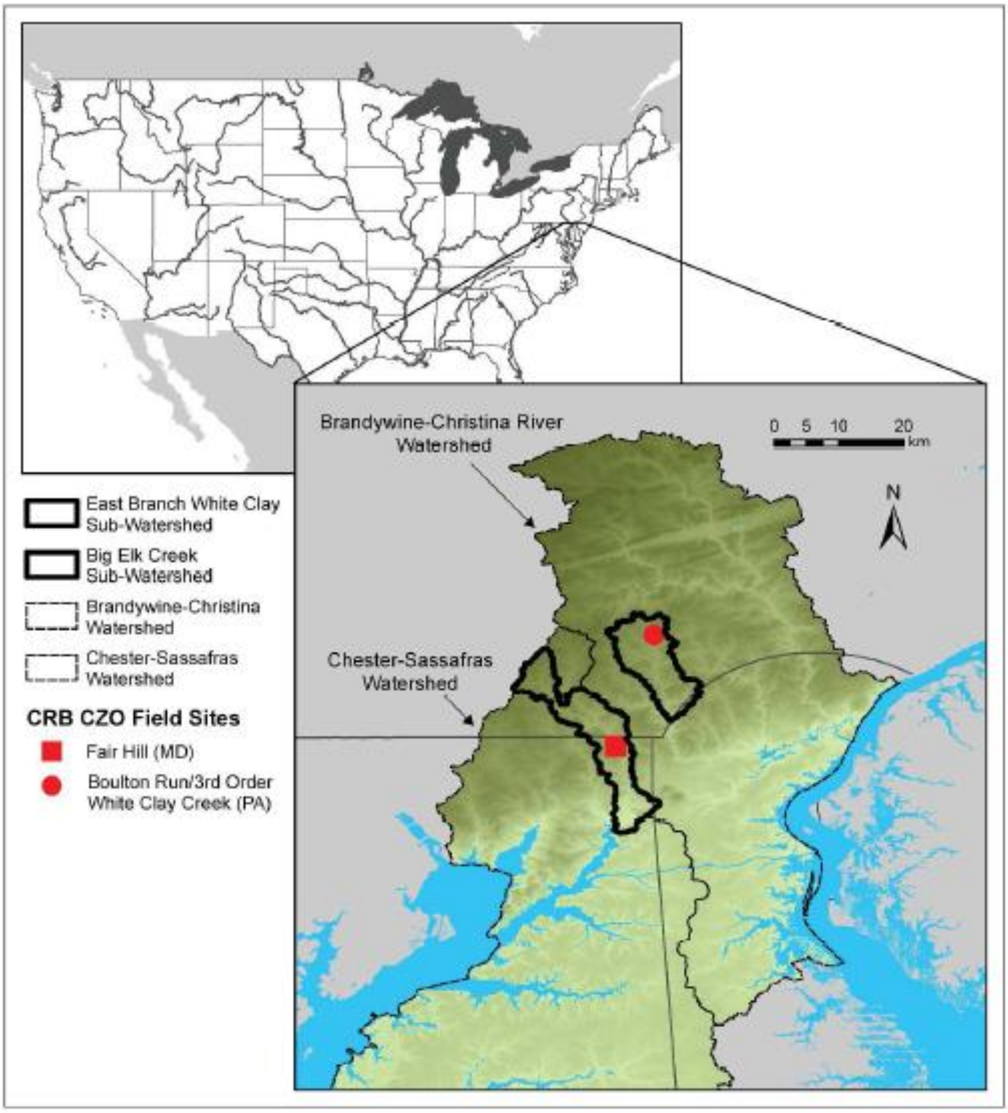

Figure 1 – Study area location

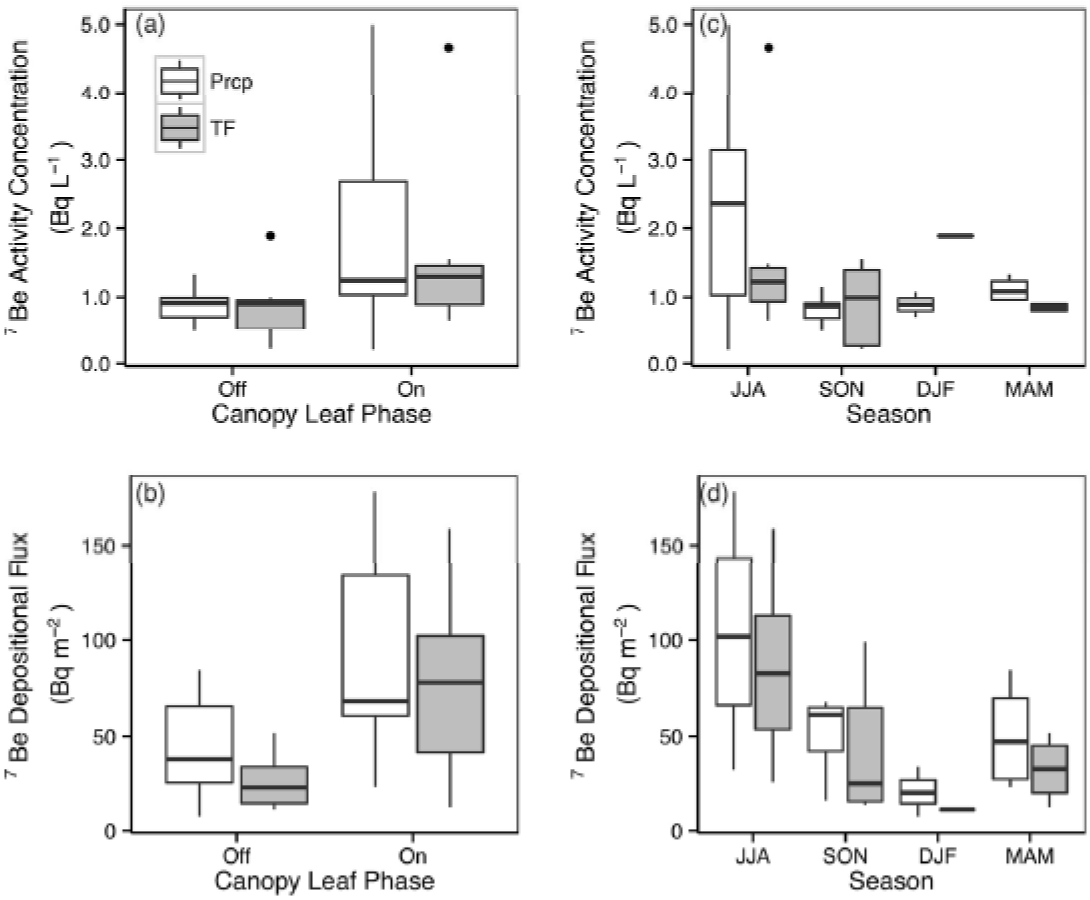

Figure 2 – ^{7}Be and activity concentration (a, c) and wet deposition flux (b,d) in precipitation and throughfall across canopy leaf phases and seasons.

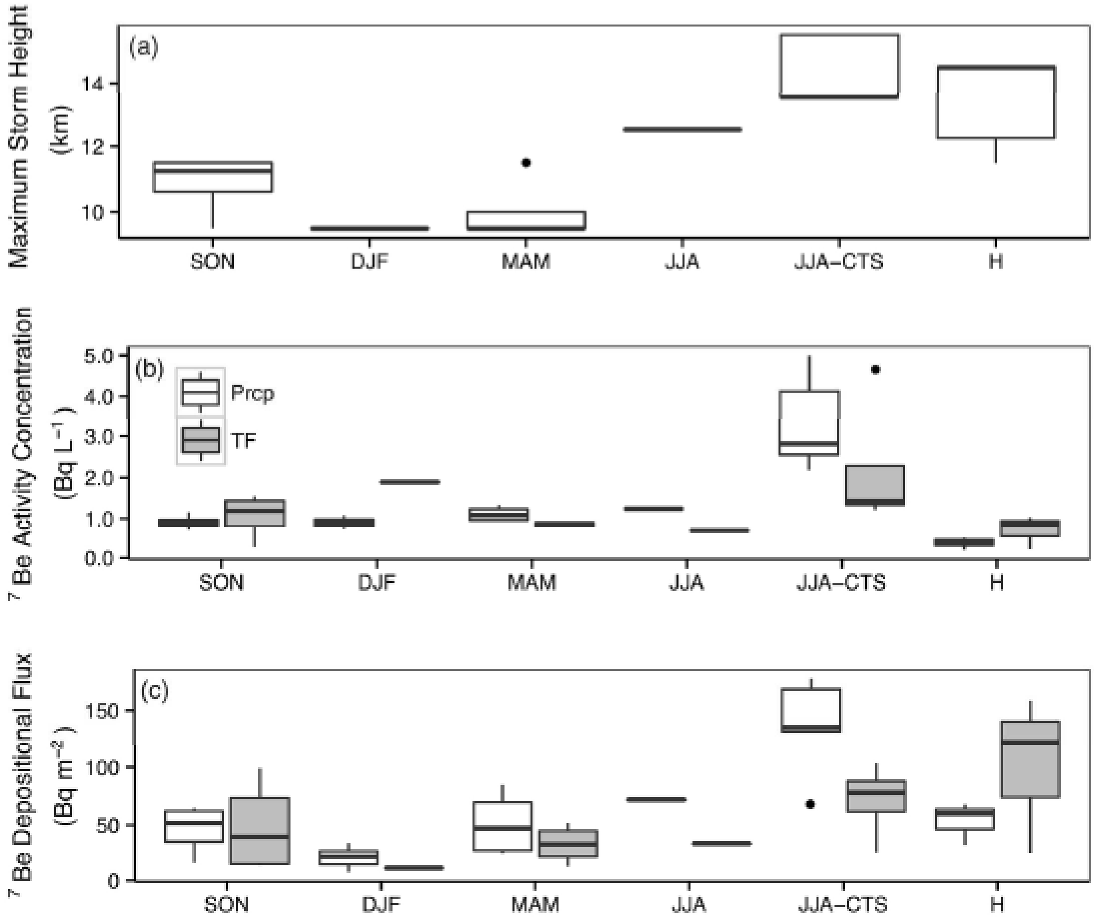

Figure 3 – Maximum storm height (a), ^{7}Be and activity concentration (b) and wet deposition flux (c) across season and storm types. Abbreviations for event groups: SON- Autumn non-tropical cyclones; DJF – winter events; MAM-spring events; JJA- summer low pressure systems; JJA-CTS- Summer Convective Thunderstorm; H- Hurricane or Tropical Cyclones occurring in summer and autumn.

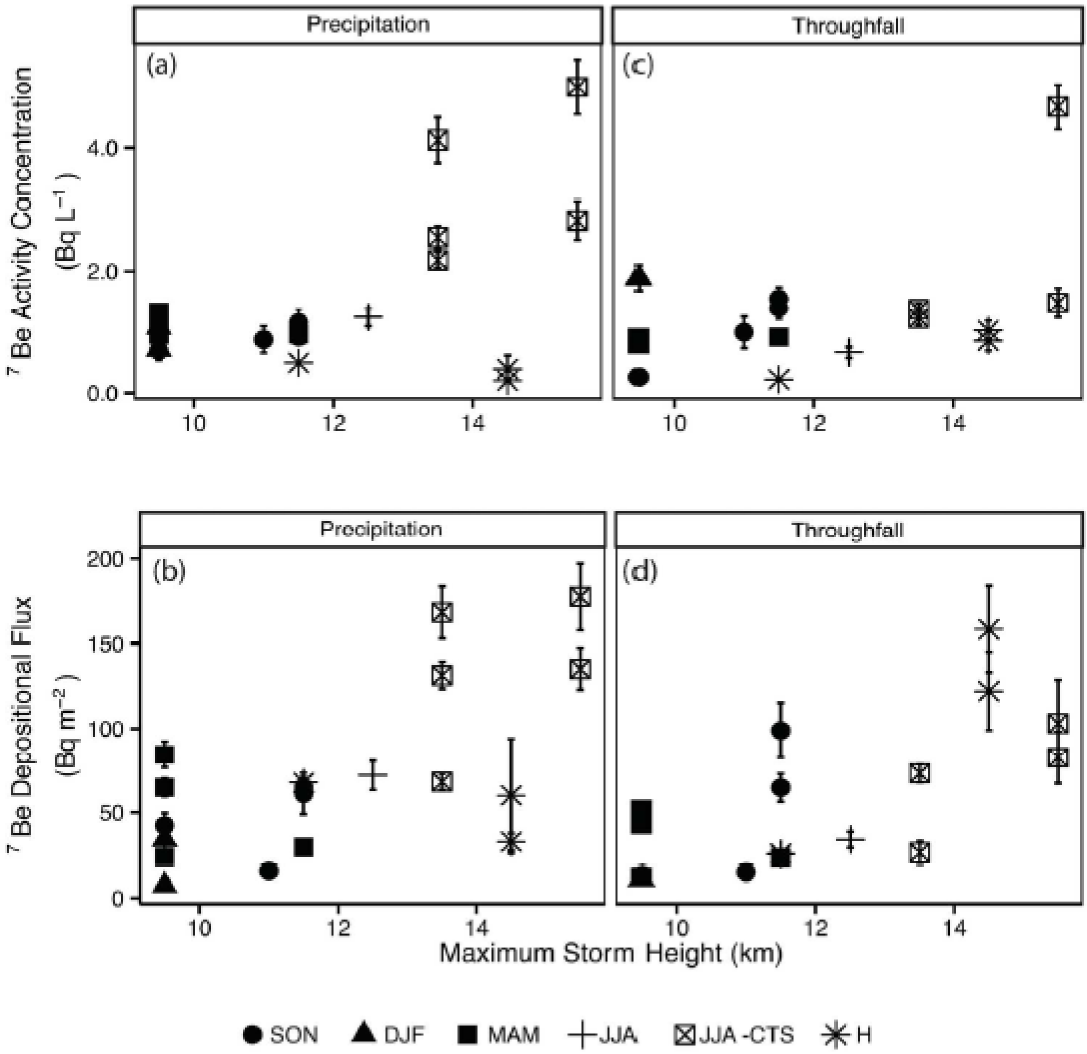

Figure 4 – Relationship between maximum storm height and ^{7}Be activity concentration (a,c) and wet deposition flux (b,d) in precipitation and throughfall. Error bars on activity concentration was calculated as a 1-sigma standard deviation per (Gilmore, 2008). Error bars on fluxes were propagated from the activity standard deviation and the reported error on our hydrologic flux measurements.

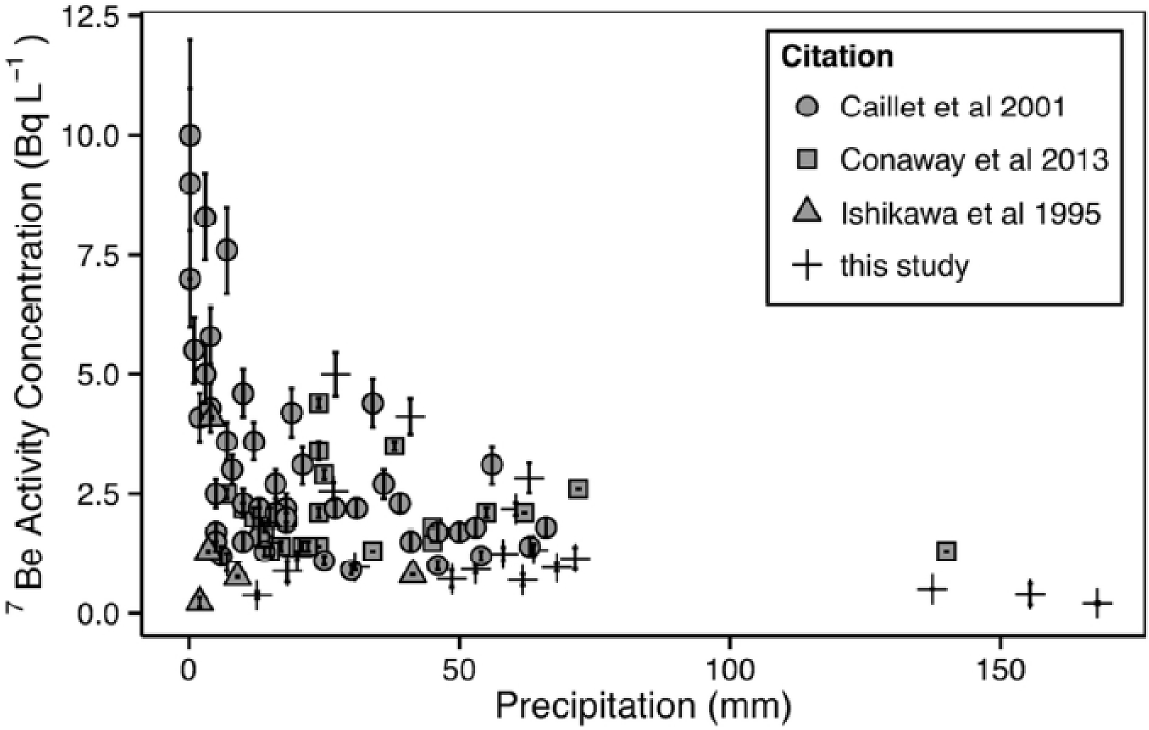

Figure 5 – ^{7}Be activity concentration by storm total precipitation from studies reporting event-based collections: our study, Conaway et al 2013, Caillet et al 2001, and Ishikawa et al 1995. Errors on individual concentration measurements were reported in all of the original publications and computed for this study as a 1-sigma standard deviation per (Gilmore, 2008).

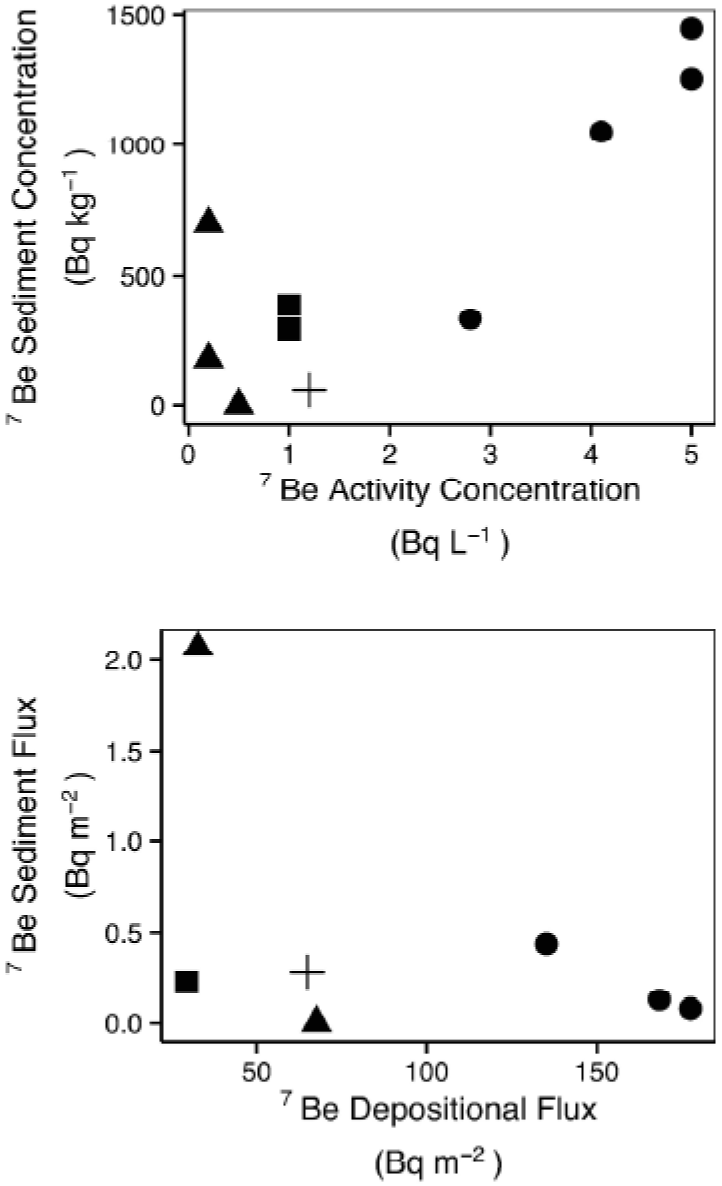

Figure 6 – ^{7}Be activity concentration in the precipitation and on the stream sediment within a single storm event. Different symbols indicate different storm types: CTS – Summer Convective Thunderstorm; H- Hurricane or Tropical Cyclone occurring in summer and autumn; LowGL – Low Pressure system originating from the Great Lakes; LowML – Low Pressure system originating from the Midwest. Standard deviation values for sediment concentrations average 5% of the measured concentration and plot within the symbols.

Table 1 - Summary of sampled events at Boulton Run (BR) and Fair Hill (FH) sites including ^{7}Be activity concentration (Bq L^{-1}) and wet deposition flux (Bq m^{-2}). Standard deviation of the ^{7}Be activity concentration is given in parentheses following concentration. Storm types are classified as CF- cold front, CTS- convective thunderstorms, H- hurricane, Low(GL)- low pressure with Great Lakes origin, and Low(MW)- low pressure with Midwest origin

Event Date	Site	Precipitation (mm)	Throughfall (mm)	Duration (h)	Canopy Phase	Season	Storm Type	Maximum Storm Height (km)	Precipitation ^{7}Be (Bq L^{-1})	Precipitation ^{7}Be Wet Deposition Flux (Bq m^{-2})	Throughfall ^{7}Be (Bq L^{-1})	Throughfall ^{7}Be Deposition (Bq m^{-2})
8/13/11	BR	62.9	56	49	Leafed	JJA	CTS	15.5	2.8 (0.3)	177.5	1.5 (0.2)	82.6
8/18/11	BR	27.1	22.1	21	Leafed	JJA	CTS	15.5	5.0 (0.5)	135.2	4.7 (0.4)	103.0
8/19/11	BR	40.9	35.2	16	Leafed	JJA	CTS	13.5	4.1 (0.4)	168.4	n.s.	n.s.
8/27/11	FH	155.5	143.7	23	Leafed	JJA	H- Irene	14.5	0.4 (0.2)	60.1	0.9 (0.2)	122.1
8/27/11	BR	167.9	155.4	30	Leafed	JJA	H-Irene	14.5	0.2 (0.0)	32.8	1.0 (0.2)	158.5
9/23/11	BR	71.4	64	87	Leafed	SON	Low(MW)	11.5	0.9 (0.1)	64.9	1.6 (0.2)	98.9
9/23/11	FH	53	46.6	15	Leafed	SON	Low(MW)	11.5	1.2 (0.2)	61.1	1.4 (0.2)	64.7
11/16/11	BR	18.2	15.3	22	Leafless	SON	Low(MW)	11	0.9 (0.2)	15.8	1.0 (0.3)	15.1
11/21/11	FH	61.7	52.4	62	Leafless	SON	Low(MW)	9.5	0.7 (0.1)	42.3	0.3 (0.1)	13.5
12/6/11	FH	48.6	41.2	45	Leafless	DJF	CF	9.5	0.7 (0.2)	34.3	n.s.	n.s.
1/27/12	FH	7	5.8	5	Leafless	DJF	Low(GL)	9.5	1.1 (0.2)	7.5	1.9 (0.2)	11.0
2/29/12	BR	30.7	26	14	Leafless	MAM	Low(GL)	11.5	1.0 (0.1)	29.5	0.9 (0.1)	23.6
4/22/12	BR	63.7	54.1	36	Leafless	MAM	CF	9.5	1.3 (0.1)	84.4	0.8 (0.1)	42.9
4/22/12	FH	68	57.8	50	Leafless	MAM	CF	9.5	1.0 (0.1)	64.8	0.9 (0.1)	51.8
5/8/12	FH	20	15.4	36	Leafed	MAM	Low(GL)	9.5	1.2 (0.1)	24.0	0.8 (0.1)	12.3
6/12/12	FH	58.1	51.5	17	Leafed	JJA	Low(GL)	12.5	1.2 (0.2)	72.0	0.7 (0.1)	34.0
7/15/12	FH	60.4	53.6	44	Leafed	JJA	CTS	13.5	2.2 (0.1)	131.4	1.4 (0.1)	73.2
8/1/12	BR	26.7	21.7	15	Leafed	JJA	CTS	13.5	2.6 (0.2)	68.1	1.2 (0.1)	26.5
10/28/12	BR	137.4	113.6	66	Leafless	SON	H- Sandy	11.5	0.5 (0.2)	67.6	0.2 (0.0)	25.7

Table 2 - Pairwise ANOVA p-value results for ^{7}Be activity concentration and wet deposition flux in precipitation by storm type. Statistically significant values (α= 0.05) appear in **bold**. Non-summer storms were all of continental low pressure origin and classified by season: DJF, MAM, and SON. Summer storms were separated into continental low pressure (JJA) and convective thunderstorm (JJA-CTS). Tropical systems occurring in both JJA and SON were classified together: H. A single JJA was observed and therefore omitted from ANOVA analysis due to limited sample size.

	DJF	MAM	JJA	JJA-CTS	SON	H
Precipitation Activity Concentration						
DJF	-	0.285	-	**0.04**	0.937	0.055
MAM	0.285	-	-	**0.008**	0.174	**0.002**
JJA	-	-	-	-	-	-
JJA-CTS	**0.04**	**0.008**	-	-	**0.005**	**0.006**
SON	0.937	0.174	-	**0.005**	-	**0.009**
H	0.055	**0.002**	-	**0.006**	**0.009**	-
Precipitation Wet Deposition Flux						
DJF	-	0.268	-	**0.017**	0.25	0.15
MAM	0.268	-	-	**0.012**	0.808	0.889
JJA	-	-	-	-	-	-
JJA-CTS	**0.017**	**0.012**	-	-	**0.007**	**0.022**
SON	0.25	0.808	-	**0.007**	-	0.66
H	0.15	0.889	-	**0.022**	0.66	-

Table 3 - ANOVA p-value results for the comparison of precipitation and throughfall activity concentration and wet deposition flux within each storm type. Statistically significant values ($\alpha = 0.05$) appear in **bold**. Non-summer storms were all of continental low pressure origin and classified by season: DJF, MAM, and SON. Summer storms were separated into continental low pressure (JJA) and convective thunderstorm (JJA-CTS). Tropical systems occurring in both JJA and SON were classified together: H. A single JJA event and a single DJF throughfall event were observed and therefore omitted from ANOVA analysis due to limited sample size.

	DJF	MAM	JJA	JJA-CTS	H	SON
activity concentration	-	**0.0388**	-	0.262	0.262	0.655
wet deposition flux	-	0.33	-	**0.0418**	0.301	0.935

Table 4 - Suspended sediment ^{7}Be activity and event total sediment-associated ^{7}Be flux for storm events in which both precipitation and sediment samples were obtained in the BR-WCC watershed. One standard deviation of the sediment ^{7}Be concentration, computed based on counting statistics, is provided in parenthesis.

Date Time (EST)	WCC Sediment ^{7}Be (Bq kg^{-1})	WCC ^{7}Be Flux (Bq m^{-2})
8/14/2011 12:50	330.4 (19.0)	0.08
8/18/11 19:03	1447.1 (38.5)	0.44
8/18/11 18:03	1254.9 (75.1)	0.44
8/25/11 11:25	1051.9 (11.3)	0.13
8/28/11 1:09	175.2 (13.6)	2.07
8/28/11 5:55	698.6 (11.9)	2.07
9/23/11 19:24	58.3 (4.7)	0.28
2/29/12 13:35	386.6 (23.1)	0.23
2/29/12 22:30	289.9 (17.4)	0.23
10/28/12 17:01	Below Detect	0.00
10/28/12 20:39	Below Detect	0.00