Extreme storms and changes in particulate and dissolved organic carbon in runoff: Entering uncharted waters?

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[1] We determined the runoff exports of particulate (POC) and dissolved organic carbon (DOC) from a 12 ha forested catchment which received more than 14 storm events in a 16-month period and three extreme events associated with hurricanes. While POC and DOC exports for the small events were comparable, POC exports for the hurricane-associated events were six to eight times the DOC values. Hurricane Irene alone contributed to 56% (21.2 kg C ha^{-1}) and 19% (3.3 kg C ha^{-1}) of the 2011 exports of POC and DOC, respectively. A precipitation threshold beyond which POC fluxes rapidly outpaced the DOC values was also identified. Our study suggests that large, high-intensity storm events that are predicted to increase under future climate-change scenarios will dramatically alter the runoff C regime by enhancing the POC inputs to aquatic ecosystems. Such shift in C forms could have important consequences for aquatic biota, atmospheric C cycling, and ecosystem and human health. **Citation:** Dhillon, G. S., and S. Inamdar (2013), Extreme storms and changes in particulate and dissolved organic carbon in runoff: Entering uncharted waters?, *Geophys. Res. Lett.*, *40*, 1322–1327, doi:10.1002/grl.50306.

1. Introduction

[2] The export of organic carbon (C) in runoff has important environmental and ecological consequences for terrestrial and aquatic ecosystems. Organic carbon plays a key role in the complexation and transport of toxic metals and organic contaminants [*Bolan et al.*, 2010], and its presence in drinking water supplies can lead to the formation of carcinogenic disinfection by-products (DBPs) during disinfection [*Karanfil et al.*, 2008]. In aquatic ecosystems, C provides critical protection against harmful UV radiation [*Williamson and Zagarese*, 1994], is a starting point for the aquatic food chain [*Findlay*, 2010], and regulates key ecosystem processes [*Tank et al.*, 2010]. Lateral flux of C via inland waters also plays a significant role in global carbon cycling and the regional budgets of organic carbon entering the oceans [*Butman and Raymond*, 2011; *Cole et al.*, 2007]. These phenomena, however, are impacted by not only the total amount of organic C in catchment runoff but also the relative proportions of dissolved (DOC) and particulate (POC) forms of organic C (operationally differentiated based on size; DOC typically $<0.45\,\mu m$). Thus, understanding how the relative proportions of C forms vary in runoff and the environmental drivers responsible is critical for characterizing impacts of C on ecosystem processes and services.

[3] It is widely recognized that exports of both POC and DOC are highest during storm events [*Hood et al.*, 2006; *Inamdar et al.*, 2006; 2011; *Jeong et al.*, 2012]. Storm events that constituted only 10%–20% of the total study period contributed to >80% of POC and >70% of DOC exports [*Oeurng et al.*, 2011]. Much less is however known about how the relative fluxes of POC and DOC change for events of varying magnitude, especially the most extreme events. Studies for large catchments suggest that POC constitutes a minor component of the total fluvial C flux [*Alvarez-Cobelas et al.*, 2012]. A review of 40 catchments worldwide by *Hope et al.* [1994] indicated that the ratio of DOC to POC varied between 0.1 and 70 and POC comprised a small percentage (<10%) of the total C export. In contrast, recent studies in small, headwater catchments [*Jeong et al.*, 2012; *Oeurng et al.*, 2011] reveal an opposite result with POC concentrations and exports exceeding the DOC values. POC results from these headwater studies have however been limited to a handful of extreme storms. Much more work is needed in characterizing how the relative proportions of POC and DOC flux evolve across a broad range of storm events, including the largest events.

[4] The influence of magnitude and intensity of storm events for C exports is especially important considering that recent studies have already confirmed that the magnitude and intensity of storms have increased, especially for the Northeast USA [*Groisman et al.*, 2005]. Furthermore, future climate-change predictions suggest that the intensity and magnitude of storm events is likely to increase with a greater potential for stronger hurricanes and tropical storms [*Karl et al.*, 2009]. Thus, addressing the shifts in exports of POC and DOC for the largest and the most extreme storms is not only critical for assessing impacts on water quality and ecosystem services but also for investigating how ecosystem conditions may evolve under weather conditions representative of future climate. Large impacts associated with extreme events coupled with their infrequent occurrence and sampling difficulties make these events especially valuable. Here, we present results from an intensive, high-frequency sampling campaign from a 12-ha forested, mid-Atlantic Piedmont catchment in the United States that was subjected to more than 14 storms within a 16-month period. Two of the events were associated with hurricanes Nicole (2010) and Irene (2011) with precipitation amounts exceeding 150 mm and which yielded very high C exports. In addition, we include data from another recent large storm event—Hurricane Sandy that occurred in October 2012. The key questions we ask: *How did the extreme storm events impact the relative exports of POC and DOC and were these responses different from those for the moderate and smaller events*? *Is there a specific*

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event threshold beyond which the relative exports of POC and DOC change dramatically?

2. Materials and Methods

[5] The study catchment (12 ha, Figure 1) is a part of an ongoing study on C [*Inamdar et al*., 2011; 2012] and is located within the Fair Hill Natural Resources Management Area (39°42' N, 75°50' W) in Cecil County, Maryland (Figure 1). The maximum daily mean temperature is 24.6°C (July), and the daily minimum is –0.6°C (January), with a mean annual temperature of 12.2°C (Maryland State Climatologist Office Data Page). Mean total annual precipitation in this region is 1231 mm with about 350 mm occurring as snowfall in winter (Maryland State Climatologist Office Data Page). The site falls within the Piedmont Plateau region and is underlain by the Mt. Cuba Wissahickon formation which includes pelitic gneiss and pelitic schist. The catchment is covered predominantly (61%) with deciduous forest with pasture along the catchment edges. Dominant tree species are Fagus grandifolia (American beech), Liriodendron tulipifera (yellow poplar), and Acer rubrum (red maple).

[6] Stream flow discharge was monitored at the outlet of the 12 ha catchment using a 6-inch Parshall flume with flow depths recorded every 15 minutes using a Global Water (Inc.) logger and pressure transducer. Precipitation and air temperature data were available at 5-minute frequency from a Delaware Earth Observation System (DEOS) weather station located in the Fairhill NRMA, about 1000 m from the outlet of the 12-ha catchment. POC and DOC data were available for 14 storm events collected over a 16-month period extending from September 2010 to December 2011. POC and DOC data for the remnants of Hurricane Sandy (October 2012) has also been included to add to our sample of extreme events. Baseflow sampling was performed once a month for C; however, since POC concentrations were below detection for the initial set of samples, baseflow sampling was limited to DOC only. Storm event sampling for stream water was performed using automated ISCO samplers which were programmed in a "nonuniform time" mode with a sampling frequency that ranged from as low as 15 minutes on the hydrograph rising limb to 3 h on the recession limb.

[7] All stream water samples were collected in HDPE bottles and filtered through a 0.45-µm filter paper (Millipore, Inc.) within 24 h of collection. The filtered water samples were stored at 4 °C. The filter papers with the retained material was dried to a constant weight by heating in an oven for 103°C–105°C for 1 h. The weight of sediment on the filter was divided with the sample volume to obtain the concentration of suspended sediments in mgL^{-1}. The suspended sediment in this study represented a size range of 0.45 µm to about 1 mm.

[8] Dissolved organic carbon was analyzed using a Tekmar-Dohrmann Phoenix 8000 TOC analyzer at the Biogeochemistry Laboratory at SUNY-ESF, NY. The sediment collected on the filters was analyzed by the University of Delaware (UD) soil testing laboratory. Total organic carbon (%C of sediments) was determined using the Elemental TC analyzer (Elementar VarioMax) by following Dumas method [specific details in *Dhillon*, 2012]. The %C content was multiplied with the concentration of suspended sediments (mg L^{-1}) to determine the concentration of POC in runoff (mg L^{-1}).

[9] The start of a storm event was defined when 10% increase in discharge was observed immediately following precipitation input. The end of the event occurred when streamflow discharge returned to within 10% of the pre-event value or when no perceptible decrease in discharge was observed over a period of 2 h, whichever occurred earlier. To compute the mass flux of POC and DOC for the 14 sampled storm events for 2011, the concentrations of POC and DOC were linearly interpolated for 15-minute time steps to match the frequency of the discharge values measured at 15-minute intervals during the storm events. These concentrations were then multiplied by the corresponding discharge values to arrive at the mass flux for that time step. The total mass flux for the storm events was determined by summation of the mass fluxes for individual time steps. Regression relationships between POC and discharge and DOC and discharge were also developed for the sampled events. These relationships were then extended to compute mass fluxes for 2011 events that were not sampled. Similarly, the mass flux for DOC for baseflow periods in 2011 was determined by interpolating DOC concentrations between measured values and multiplying them with the corresponding baseflow discharge values followed by the summation of the product for the period of interest. Since POC values during baseflow were below detection, baseflow POC flux was neglected. In addition, prior to September 2010, DOC data were also available for both storm events and baseflow for the period of 2008–2010 [*Inamdar et al*., 2011]. Storm event and baseflow mass fluxes for DOC for the period 2008–2010 were also computed (no POC data were available for this period). This allowed us to compare the magnitude of DOC exports during 2011 against

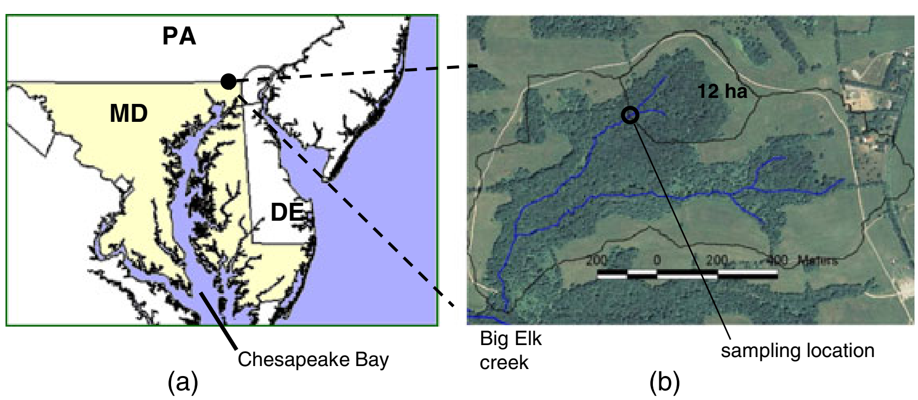

Figure 1. (a) Location of the study site in Maryland (MD) and within the drainage basin of the Chesapeake Bay. (b) The 12 ha forested (61% of area) catchment with the sampling location and its drainage into the Big Elk Creek.

those for the previous years and thus get some estimate on how similar or dissimilar the DOC flux for 2011 was compared to the previous years.

3. Results and Discussion

[10] A total of 14 storm events with a wide range in magnitude and intensity were sampled over 16 months from September 2010 to December 2011 (Figure 2). The total precipitation for the 16-month study period was 1842 mm, of which, 1462 mm occurred during year 2011. The annual precipitation for 2011 was greater than the previous years—2008 (1052 mm), 2009 (1238 mm), and 2010 (972 mm). Total stream discharge measured at the 12-ha outlet for the 16-month study period was 497 mm, resulting in an annual runoff ratio of 0.27. The largest amount of precipitation (155 mm) was recorded for the event of 27 August 2011 (event 10, Table 1), which was associated with remnants of hurricane Irene [*National Oceanic and Atmospheric Administration, NOAA*, 2012] and had a precipitation return period of 25 years [Appendix C, Ward and Trimble, 2004]. This event also produced the highest peak discharge (5.0 mm/h) and highest total amount of streamflow discharge (32.7 mm) for the study period. Similarly, the event of 30 September 2010 (event 1) was associated with remnants of hurricane Nicole [*National Oceanic and Atmospheric Administration, NOAA*, 2012] and yielded a rainfall amount of 151 mm and had a precipitation return period of 25 years [Appendix C, Ward and Trimble, 2004]. Remnants of Hurricane Sandy that lashed the mid-Atlantic region in October 2012, delivered a total of 139 mm with 119 mm associated with our sampled storm event. The total streamflow discharge for this event amounted to 18.5 mm.

[11] Particulate organic carbon and DOC concentrations increased rapidly during storm events with peak POC concentrations being 2–20 times the DOC values. DOC concentrations for storm events ranged from 0.7 to 18.3 mgL^{-1} while the POC concentrations ranged between 0.05 and 252 mgL^{-1} (Figure 2). Storm event exports of POC and DOC (Table 1) varied considerably across the 14 storm events but with a much larger variation in POC (0.04–21 kg C ha^{-1}) versus DOC (0.01–3.3 kg C ha^{-1}). The relative contribution of POC to the event C export ranged from 52% to 92% (Table 1). POC:DOC mass ratios for the individual events varied from 1.1 to a high of 12, with the largest events of 2011 (Events 1, 8, 9, and 10), maintaining a ratio in excess of 6. The sum of the total C flux for the 14 storm events was 53 kg C ha^{-1}, of which, POC contributed 43 kg

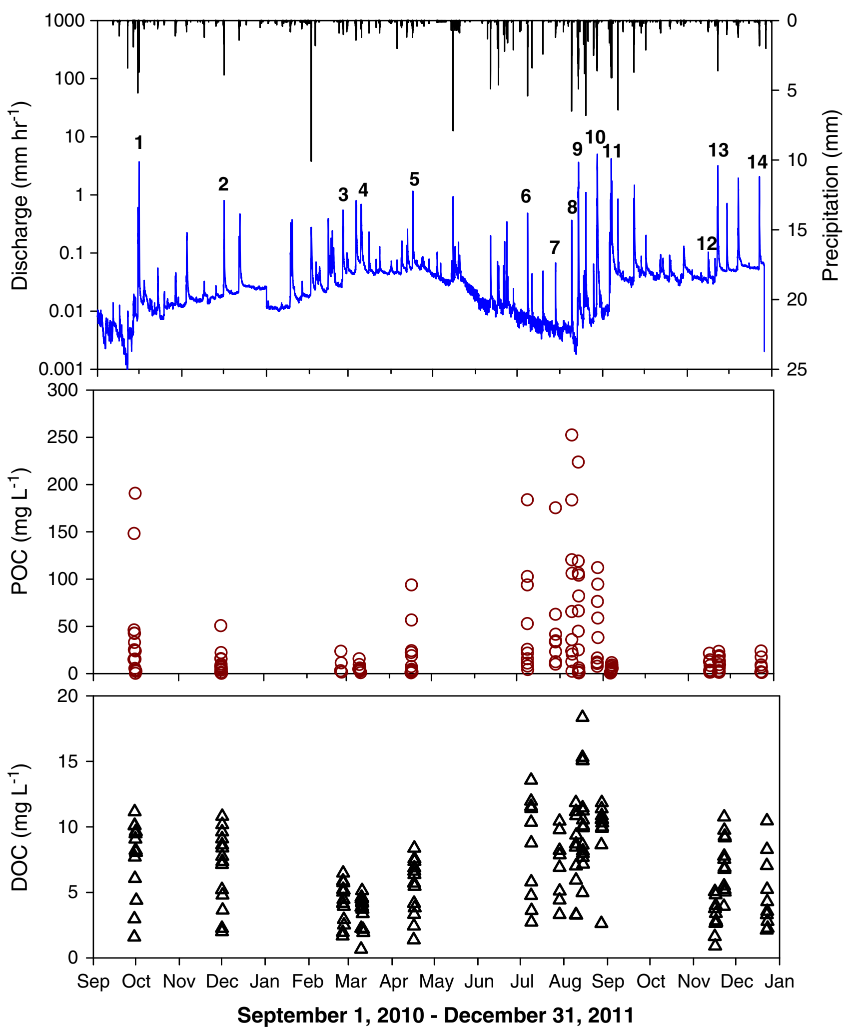

Figure 2. Precipitation (mm), streamflow discharge (mm h^{-1}), and POC and DOC concentrations (mg C L^{-1}) for the 14 sampled storm events over the 16-month study period extending from 1 September 2010 to 30 December 2011. Events 1 and 10 were associated with hurricanes Nicole (2010) and Irene (2011), respectively.

Table 1. Precipitation (mm), streamflow discharge (mm), and mass exports (kg C ha^{-1}) of POC, DOC, and TOC for the 14 sampled events and the relative contribution of POC and DOC toward the total carbon export from the 12-ha catchment

Event	Date	Precipitation (mm)	Streamflow (mm)	POC (kg C ha^{-1})	DOC (kg C ha^{-1})	TOC (kg C ha^{-1})	POC %	DOC %	POC:DOC
1	**30 September 2010[a]**	**151**	**13.5**	**9**	**1.1**	**10.1**	**89**	**11**	**8.2**
2	1 December 2010	34.7	3.8	0.28	0.22	0.49	57	43	1.3
3	25 February 2011	21.2	6.3	0.56	0.24	0.8	70	30	2.3
4	10 March 2011	45.9	11.1	0.64	0.38	1	63	37	1.7
5	16 April 2011	37.7	7.1	1.1	0.37	1.5	75	25	3
6	8 July 2011	23.4	0.8	0.38	0.07	0.45	84	16	5.4
7	28 July 2011	11	0.1	0.04	0.01	0.05	81	19	4
8	9 August 2011	21	0.5	0.48	0.04	0.52	92	9	12
9	14 August 2011	104	9.3	5.9	0.9	6.8	87	13	6.6
10	**27 August 2011[b]**	**155**	**32.7**	**21.2**	**3.3**	**24.5**	**87**	**13**	**6.4**
11	6 September 2011[c]	102	16.5	1.2	1.5[d]	2.7	44	56	0.8
12	16 November 2011	17.1	2	0.11	0.07	0.18	61	39	1.6
13	22 November 2011	52.8	16.1	1.3	1.2	2.5	52	49	1.1
14	23 December 2011	35	9.5	0.8	0.48	1.3	62	38	1.7
Total for 14 events (September 2010–2011)				43.0	9.9	52.9	81	19	4.3
Total 2011 baseflow export				neglected	4.2	4.2	neglected	100	NA
Total 2011 stormflow export (sampled + unsampled events[e])				33.8 + 3.9	8.6 + 4.7	42.3 + 8.6	74	26	2.8
Total 2011 C export				37.7	17.5	55.2	68	32	2.2

The hurricane-associated events (no. 1 Nicole in 2010 and no. 10 Irene in 2011) are indicated in bold.
[a]Storm associated with remnants of hurricane Nicole.
[b]storm associated with remnants of hurricane Irene.
[c]storm associated with remnants of Tropical depression Lee.
[d]DOC was not measured, but mass flux was estimated using regression equation between discharge and DOC; and
[e]values estimated from regression analysis.

C ha^{-1} (81% of the total C). The highest exports of POC and DOC were associated with Hurricane Irene on 27 August 2011, which yielded 21.2 and 3.3 kg C ha^{-1}, respectively (Table 1). This event alone contributed to 56% (21.2 of 37.7 kg C ha^{-1}) and 19% (3.3 of 17.5 kg C ha^{-1}) of the 2011 exports of POC and DOC, respectively. The three largest events in terms of total precipitation for 2011—event 1 (Hurricane Nicole, 30 September 2010), event 9 (14 August 2011), and event 10 (27 August 2011) accounted for 84% of the total POC export and 63% of the total DOC export during the storm events. POC and DOC exports for the storm associated with Hurricane Sandy (October 2012) yielded 7.72 and 1.97 kg C ha^{-1}, respectively, which is comparable to the large hurricane-associated events of 2011 and 2010. This highlights the importance of large precipitation events in the export of total carbon, especially POC. The importance of high-discharge events was further underscored by flow-duration analysis (data not included because of limited space), which revealed that more than 88% of POC and 46% of DOC was exported during high stream flows (more than 0.5 mm/h) which occurred less than 1% of the time during the sampling period. Our observations of the importance of POC to C flux are contrary to some of the early studies for large catchments [*Hope et al*., 1994] but more in line with recent studies involving high-frequency sampling for large events in headwater catchments [*Jeong et al*., 2012; *Oeurng et al*., 2011; *Pawson et al*., 2012].

[12] When our total DOC export for 2011 was compared against those for the previous years (2008–2010, Table 2), the DOC export for 2011 was, not surprisingly, greater. This was primarily because of the elevated stormflow exports of DOC in 2011 (13.26 kg ha^{-1} yr^{-1}), resulting from higher annual precipitation for 2011. The DOC export during baseflow was fairly similar across all years (2008–2011) and ranged between 4.1 and 4.3 kg ha^{-1} yr^{-1}. Considering that POC exports increased more rapidly than DOC for large storm events, we speculate that the POC contribution to the total C flux for 2011 was also likely on the higher side and that POC exports for the more "normal precipitation" years such as 2008–2010 would be on the lower side and comparable to DOC mass exports.

[13] When POC and DOC fluxes for individual events are plotted against corresponding precipitation amounts (Figure 3a), very interesting results are revealed: (a) POC exports were similar or only slightly greater than the DOC values for the smallest events (Figure 3a inset, event precipitation <40 mm); however, the differences between the two were dramatically different for the largest hurricane-associated events (Nicole, Irene, and Sandy) with POC exports six to eight times the DOC values; (b) the rate of increase of POC flux for the large events was much greater than that for DOC; DOC increase was linear for the full range while POC was linear for the small/moderate events (inset) and exponential for the large events; (c) there appears to be a precipitation threshold or transition region beyond which the POC exports increased much more rapidly than DOC—for our study site, this precipitation threshold or transition region appeared to be in the vicinity of 75 mm; (d)

Table 2. Annual Precipitation (mm) and DOC Exports (kg C ha^{-1}) from the 12 ha Catchment for the Years 2008–2011

Year	Annual Precipitation (mm)	Baseflow Contribution (kg C ha^{-1})	%	Stormflow Contribution (kg C ha^{-1})	%	Total DOC Export (kg C ha^{-1})
2008	1052	4.35	47	4.98	53	9.34
2009	1238	4.10	40	6.25	60	10.35
2010	972	4.19	43	5.49	57	9.67
2011	1462	4.24	24	13.26	76	17.5

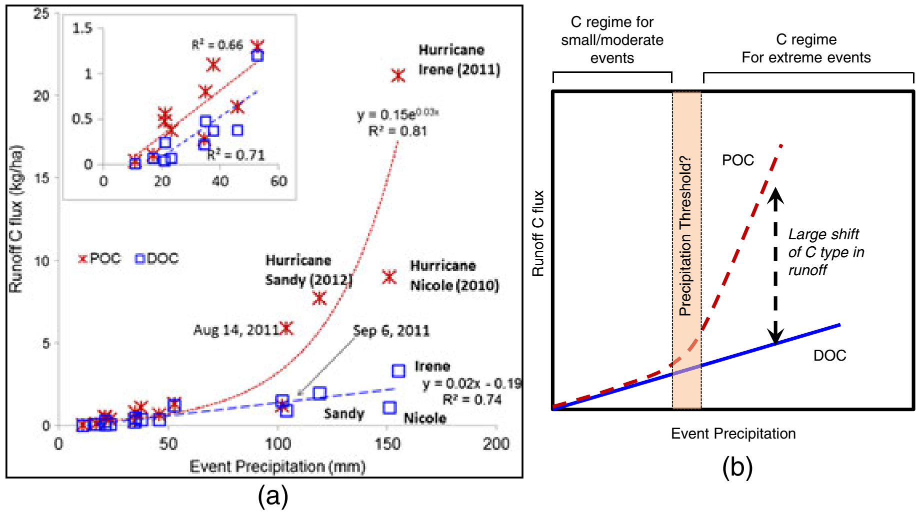

Figure 3. (a) Runoff flux of POC and DOC (kg C ha^{-1}) versus precipitation totals (mm) for the sampled storm events, including three events associated with hurricanes. Inset—a magnified view of the linear relationship between mass exports and precipitation for the small events. (b) Conceptual model based on Figure 3a illustrating the flux trajectories of POC and DOC as a function of event size and the precipitation threshold or transition region beyond which POC exports increase dramatically.

the event of 6 September 2011 which occurred after five consecutive events within a month yielded a very low POC export—this suggests that POC exports will not increase indefinitely and that POC stores in catchment could get exhausted after a sequence of large storm events.

[14] These observations clearly show that large disparities in C forms could be expected when catchments are subjected to extremely large storm events. Building on our observations in Figure 3a, we present a broader conceptual model that characterizes the relative exports of POC and DOC with event magnitude (Figure 3b). POC and DOC follow different trajectories (Figure 3b)—reflecting different supply rates in the catchment. Extreme events with increasing precipitation energy/input tap into newer POC sources while DOC supply appears to be limited. Indeed, our within-event observations for DOC [*Inamdar et al*., 2011, *Dhillon*, 2012, and *Dhillon et al*., In Preparation] reveal dilution in DOC concentrations at peak streamflow discharges associated with the largest events (no such dilution was observed for POC). Primary sources for DOC have been identified as throughfall, litter layer, and surficial soils [*Inamdar et al*., 2011, 2012], whereas POC appears to originate from near and within stream sediments for small/moderate events and more distal upland sediments for the more erosive extreme storm events [*Dhillon*, 2012; *Dhillon et al*., In preparation]. Following this conceptual model, we postulate that as catchments are subjected to larger and more extreme storm events, which are predicted under future climate change scenarios for the northeast United States [*Karl et al*., 2009], the runoff C regime will shift toward a greater dominance of POC over DOC (from left to right in Figure 3b). The shift to POC dominance will likely occur when the precipitation and subsequent runoff energy exceeds a specific threshold, as alluded to in Figure 3b. This threshold for precipitation will likely vary for individual catchments and would be dictated by the catchment hydrologic and geomorphic features, landuse, and the vegetation type.

[15] A recent study by *Yoon and Raymond* [2012] on Hurricane Irene in upstate New York revealed that the event exported 43% of the average annual DOC flux. Simultaneously, they also reported an increase in the aromatic fractions of DOC. Our previous work in this catchment has also shown significant increases in aromatic and humic fractions of dissolved organic matter during storm events which originated from surficial C sources such as throughfall, forest floor, and soils [*Inamdar et al*., 2011]. Findings of this study, however, suggest that it would be POC, rather than DOC, that would shape catchment C flux and its potential consequences during extreme storm events.

[16] Instream metabolism [*Tank et al*., 2010] and the structure of aquatic communities [*Findlay*, 2010] are strongly coupled with the amount and type of allochthonous C inputs that occur to aquatic ecosystems. While we still think that majority of stream baseflow and thus the runoff period will be composed of dissolved C forms, we argue that the large inputs of POC from extreme storms will have significant consequences for instream metabolism and the downstream aquatic community. For example, for our catchment, POC fluxes for the three hurricane associated events (7.72–21.2 kg ha^{-1} yr^{-1}) were itself two to five times the average annual C export in baseflow (2008–2011: 4.22 kg C ha^{-1} yr^{-1}). Such impacts could especially be pronounced for downstream lentic ecosystems as highlighted by *Klug et al*. [2012] who found that lake thermal stability declined and respiration became the primary driver of ecosystem metabolism following large allochthonous inputs from Hurricane Irene. Similarly, *Sadro and Melack* [2012] reported that allochthonous DOC and POC inputs associated with one single extreme storm event (150–200 mm of rainfall in 24 h) were enough to change the metabolism of a 2.7-ha oligotrophic, high-elevation lake in the Sierra Nevada, California, from slightly autotrophic to strongly heterotrophic condition.

[17] Elevated inputs of POC and DOC during extreme storms to municipal drinking water sources are also bound to create a new set of challenges for water treatment utilities. Elevated concentrations of POC and DOC will likely mean additional costs associated with coagulation and filtration processes to remove particulates as well as an increased potential for formation of DBPs during the chlorination phase. Studies such as ours will thus provide important data

and guidance to drinking water utilities that are required to develop adaptive response plans to address climate-change impacts [*Environmental Protection Agency, EPA*, 2012].

[18] Carbon inputs to fluvial networks directly influence CO_2 degassing and thus have important implications for atmospheric and global C cycling [*Battin et al*., 2008; *Butman and Raymond*, 2011; *Cole et al*., 2007]. Headwater or lower-order streams are especially significant sources of CO_2 evasion because of elevated dissolved CO_2 concentrations and high proportion of the total stream surface area that they constitute [*Butman and Raymond*, 2011]. Our results suggest that the large exports of POC following extreme storms could have a significant impact on CO_2 evasion. We speculate that a significant proportion of POC could be deposited in headwater fluvial zones and that such C deposits could form important hotspots or loci for C transformation and CO_2 evasion in the drainage network. *Pawson et al*. [2012] also reached a similar conclusion following their observations of substantial deposition of POC in riparian wetlands and floodplains along the fluvial drainage network.

4. Conclusions

[19] Using multiple storm events covering a wide range in precipitation magnitude, including three hurricane-associated events, this study showed that particulate forms of organic C, rather than dissolved, make up most of the large runoff C exports associated with extreme storm events. These results underscore the need to pay greater attention to how the particulate forms of organic C are generated and transported in catchments, what proportion of these fractions enters downstream aquatic ecosystems, and the bioavailabity of these C forms. Our results also suggest that the release/export rates of POC and DOC from catchment are dramatically different with DOC displaying only a gradual increase with storm magnitude. This study also revealed a potential precipitation threshold or transition region beyond which the erosive energy associated with storms resulted in much greater exports of POC versus DOC. These results provide important insights into how extreme storm events, which are expected to increase in frequency under future climate change scenarios, may impact aquatic ecosystem conditions and ecosystem services. The significant impact of these extreme storms coupled with their infrequent occurrence and the difficulties associated with sampling make such data and results especially valuable and critical.

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