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The impact of onsite wastewater disposal systems on groundwater in areas inundated by Hurricane Sandy in New York and New Jersey

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ABSTRACT

Coastal onsite wastewater disposal systems (OWDS) were inundated by Hurricane Sandy's storm tide. This study compares the shallow groundwater quality (nutrients, pharmaceuticals, and hormones) downgradient of OWDS before and after Hurricane Sandy, where available, and establishes a baseline for wastewater influence on groundwater in coastal communities inundated by Hurricane Sandy. Nutrients and contaminants of emerging concern (CECs) were detected in shallow groundwater downgradient of OWDS in two settings along the New Jersey and New York coastlines: 1) a single, centralized OWDS in a park; and 2) multiple OWDS (cesspools) in low-density residential and mixed-use/medium density residential areas. The most frequently detected pharmaceuticals were lidocaine (40%), carbamazepine (36%), and fexofenadine, bupropion, desvenlafaxine, meprobamate, and tramadol (24–32%). Increases in the number and total concentration of pharmaceuticals after Hurricane Sandy may reflect other factors (seasonality, usage) besides inundation, and demonstrate the importance of analyzing for a wide variety of CECs in regional studies.

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1. Introduction

Much of the New Jersey and New York coastline is vulnerable to sea-level rise and other extreme or unanticipated coastal hazards, such as the storm tide brought by Hurricane Sandy in 2012. Inundation of community infrastructure during coastal flood events can damage onsite wastewater disposal systems (OWDS), which typically provide minimal treatment, and may increase the load of poorly treated sewage into groundwater and adjacent coastal waters. Approximately 40% of the population in the United States lives in counties along the shore (National Oceanic and Atmospheric Administration, 2015). OWDS in coastal environments can act as a significant source of nutrients and organic wastewater-associated compounds (including pharmaceuticals, personal care and domestic use products, and endocrine active compounds referred to here as contaminants of emerging concern (CECs)) to receiving surface waters (Phillips et al., 2015; Del Rosario et al., 2014; Zhao et al., 2011; Kroeger et al., 2006a). Many of the densely populated areas along the East Coast (i.e., New Jersey and New York) are served by OWDS. Approximately 380,000 residents of Suffolk County, New York, rely on simple OWDS such as cesspools and septic systems. The presence of OWDS in areas inundated by the storm tide of Hurricane Sandy in 2012 has raised concerns over whether saltwater inundation affected 1) the presence of wastewater-associated compounds in the near shore shallow groundwater flow system and 2) the ability of the shallow groundwater flow system to attenuate wastewater-associated compounds. The high hydraulic conductivity of the sandy surficial aquifer of the New Jersey–New York coastal region makes these areas particularly vulnerable to organic wastewater contamination.

Nutrients and a variety of CECs have been associated with OWDS. As the population continues to increase within the extended metropolitan New York City area, which includes Long Island, there is growing interest to decrease the amount of nitrogen entering surrounding water bodies (Harrington, 2015). Studies have directly linked nitrogen loading to coastal waters through groundwater discharge with population density (Kasper et al., 2015; Arnold et al., 2014; Zhao et al., 2011; Kroeger et al., 2006b). Nitrogen, particularly from anthropogenic sources, has been blamed for deteriorating the ecological health of estuaries (e.g., algal blooms, hypoxia events, and eel grass die off) (Howarth, 2008). OWDS have been identified in previous research as one of the major anthropogenic nitrogen contributors to coastal groundwater (Zhao et al., 2011). CECs are of particular concern because their sources often are related directly to OWDS and they have been associated with potential adverse impacts on ecosystems. CECs can impair the ability of native bacteria in groundwater to reduce nitrogen concentrations (Underwood et al., 2011). Exposure of certain CECs (i.e., antidepressants and natural and synthetic estrogens, and other endocrine active compounds [EACs]) to fish and other non-target aquatic organisms has been identified as a

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cause for declines in fisheries and shellfish, disruption in reproductive development and function and behavioral patterns (Hinck et al., 2009; Painter et al., 2009; Vajda et al., 2008; Barber et al., 2007; Kidd et al., 2007). Studies in sewage-impacted coastal waters of western Long Island report higher ratios (>1) of female to male fish (Duffy et al., 2009), and declines in fish and shellfish populations (Kraeuter et al., 2005). Even if CECs are detected at low (nanogram per liter) concentrations, the exposure from multiple CECs on aquatic organisms and ecosystems may present potentially additive adverse effects (Kidd et al., 2007). On the basis of these many concerns, it is important that monitoring of groundwater quality in coastal areas proximal to OWDS be conducted to better understand the fate and transport of nutrients and CECs in shallow groundwater systems.

The co-occurrence of some pharmaceuticals and personal care and domestic use products (PCDU) in wastewater has led to the use of these compounds to assess the impacts of OWDS on groundwater contamination and as indicators of contributions from OWDS to loads of nutrients to groundwater. Such compounds include (but are not limited to) carbamazepine (an anticonvulsant), sulfamethoxazole (an antibiotic), caffeine (a stimulant), and 1,7-dimethylxanthine (a caffeine metabolite). Zhao et al. (2011) distinguished nitrogen sources by analyzing for pharmaceuticals and pesticides in groundwater samples collected from sewered and unsewered regions located along the north shore of Long Island. The presence of carbamazepine in the unsewered region samples was an indication that wastewater (from OWDS) was a dominant source of nitrogen.

Variations in the types of pharmaceuticals and PCDU compounds detected in groundwater impacted by OWDS can be attributed in part to product use and habits of the residing population (Conn et al., 2010; Godfrey et al., 2007; Conn et al., 2006). Seasonal effects and differences in land use (residential and nonresidential) have also been identified as factors contributing to variations in observed pharmaceutical and PCDU concentrations (Conn et al., 2010; Conn et al., 2006; Phillips et al., 2015). With the increased concern about loading of nitrogen and CECs, including EACs, to sensitive estuaries, there is a need to understand the regional influence OWDS have on groundwater quality in coastal watersheds. OWDS in coastal settings may contribute a disproportionate amount of nutrients and CECs to adjacent surface-water bodies due to the combination of sandy soils, thin unsaturated zone, and short distances to groundwater discharge zones (Barber et al., 2009; Carrara et al., 2008; Kroeger et al., 2006a, 2006b; Swartz et al., 2006). Yet, the effects of saltwater inundation on OWDS are not well documented (Bowleg and Allen, 2011; Illangasekare et al., 2006). Inundation may limit the ability of OWDS bacteria to degrade wastewater effluent in groundwater prior to its discharge into the coastal watershed, which in turn may increase potential aquatic (and human) health impacts in the environment. Groundwater-quality monitoring of indicator compounds would be useful for assessing biogeochemical conditions that could potentially be affected when inundated by saltwater (from storms like Hurricane Sandy). Furthermore, these data are necessary for coastal resiliency efforts (e.g., considerations when engineering wastewater treatment options for shoreline communities) and for the improvement of health to adjacent estuaries.

1.1. Purpose & scope

This study focused on two different types of OWDS within coastal communities: New Jersey (NJ) locations included a wastewater treatment plant that serves the buildings at a park facility and discharges to groundwater via an onsite leach field, and New York (NY) locations included multiple OWDS (i.e., cesspools) in land-use categories ranging from low-density residential to mixed use (residential and commercial). In each setting, areas where OWDS are in use were inundated with seawater by the storm tide resulting from landfall of Hurricane Sandy in 2012. Most groundwater samples were collected during the fall 2013, except for samples collected from Shinnecock Tribal Lands in Southampton, NY, which were collected in the summer of 2014. Previous research (conducted prior to Hurricane Sandy) on CECs in the shallow groundwater at Fire Island, NY, indicated that the highest pharmaceutical concentrations were present in shoreline wells (Phillips et al., 2015). The same shoreline wells were resampled for our study to investigate potential changes in groundwater quality resulting from Hurricane Sandy. Similar groundwater-quality data documenting conditions prior to Hurricane Sandy were not available for other settings sampled within the region. Another objective of our study was to establish a baseline for wastewater influence on groundwater in other coastal communities inundated by Hurricane Sandy. This baseline provides a means to assess change following future storms and identify factors affecting the presence of nitrogen and CECs in groundwater downgradient of OWDS in coastal settings.

The scope of this study includes relating the observed occurrence of nitrogen and CECs in groundwater downgradient of OWDS to population density. Developing this relation will provide information related to where in the coastal zone OWDS may adversely impact adjacent coastal waters. Data presented in this study can also be used to help describe geochemical processes in the coastal shallow aquifer one year after inundation by the storm tide of Hurricane Sandy. To our knowledge, this is the only NY–NJ regional study assessing the occurrence of nutrients and CECs in coastal communities served by OWDS that were inundated during Hurricane Sandy and the impact these systems have on downgradient groundwater quality.

2. Material and methods

2.1. Well network

The network consists of samples from two settings: 1) an area served by a single large institutional OWDS at a park facility in Sandy Hook, NJ, and 2) multiple coastal locations in NY served by several OWDS in residential and mixed-use land-use settings (Fig. 1). The NY samples included samples collected on Fire Island (a barrier island separating the Atlantic Ocean from the Great South Bay) and samples collected on the mainland of Suffolk County. Previous work on groundwater quality in this region was limited to the barrier island. This distinction allows for comparisons between our study and the previous work (Phillips et al., 2015).

2.1.1. Sandy Hook network

Sandy Hook is part of the Gateway National Recreational Area. The park is primarily a summer tourist destination popular for access to bathing beaches, camping, and wildlife viewing. Wastewater at Sandy Hook is handled through a central wastewater treatment plant, which serves all buildings (park restrooms and concessions, a vocational school, U. S. Coast Guard station, single family housing, and sanitary discharge from the marine research laboratory) on the Sandy Hook peninsula and discharges effluent after primary and secondary treatment to adjacent (onsite) leach fields. The park is visited by more than two million people annually with peak visitation during the summer months. The storm tide brought by Hurricane Sandy inundated the leach fields and disabled the wastewater treatment plant, which was operating at a reduced efficiency for months following the storm.

Groundwater samples from Sandy Hook were collected along a transect from the wastewater treatment plant leach fields west-southwest towards Sandy Hook Bay (Fig. S1). Wells sampled at Sandy Hook range in depth from 1.5 to 6 m below land surface (table S1). Two samples were collected from at different depths in the leach fields: one from a permanent observation well (SH01) screened 1.5–6 m below lands surface, and the other using a drive-point piezometer (SH02) to a depth of 1.8 m. The other three samples were collected using the drive-point piezometer at sites downgradient of the leach fields along the perceived groundwater flow-path (Fig. S1). All samples were collected in November 2013.

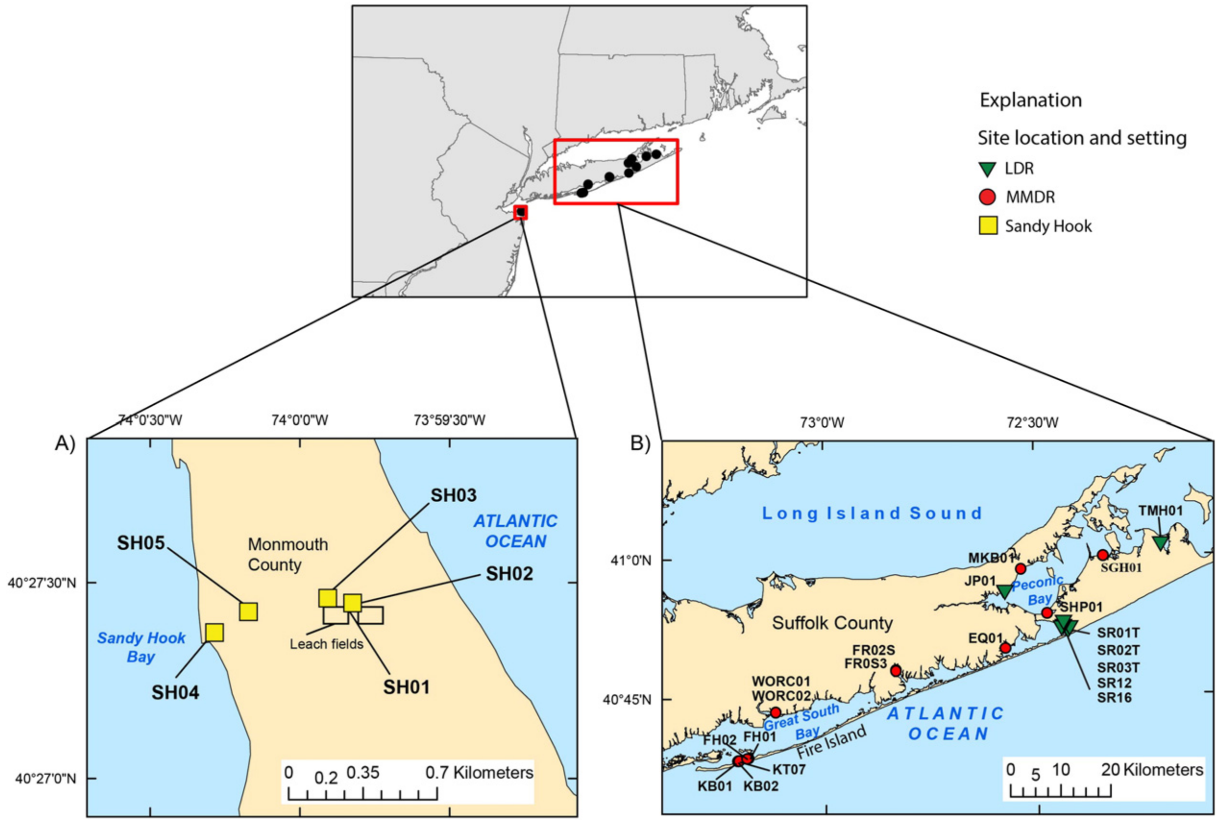

Fig. 1. Locations of sites sampled in 2013 in A) Sandy Hook, NJ, and B) Suffolk County, Long Island, NY. LDR = low density residential, MMDR = mixed-use/medium density residential. Additional site code information is available in Tables S1 and S2.

2.1.2. New York network

Groundwater samples were collected from the shallow groundwater flow system along the shoreline of (1) a barrier island summer community and (2) the mainland of Long Island (Fig. 1). Both locations are distinctive coastal communities in Suffolk County, NY. The coastal communities selected are in areas inundated by the storm tide brought on by Hurricane Sandy. Specific locations were selected in areas along the shore that are within 180 m downgradient from OWDS and just above the reaches of the spring high-tide mark along the shoreline. For our study, beach areas without bulkheads (a retaining wall built for shoreline protection) were targeted due to the need to access areas downgradient of OWDS. Cesspools, which are covered underground pits lined with cement blocks or rings without a sealed bottom, are the primary types of OWDS installed in these communities. Cesspools provide minimal wastewater treatment, typically relying on bacteria to breakdown the solid waste while untreated water percolates into the sandy surficial aquifer.

2.2. Land-use classification

This study includes a land-use classification for an assessment of results between varying land-use in the NY network (Table S2). (Note that a land-use assessment was not assigned for the Sandy Hook study area because all samples were collected from a single park and there was one (centralized) source for wastewater.) The land use 1.1 ha upgradient from each sample location was assessed for potential sources of contamination. New York sites were assigned to two land-use categories primarily based on residential density as described by the land-use assessment of the Suffolk County Department of Planning (2004). Low density residential (LDR) sites include locations with no more than one dwelling per 0.4 ha, while medium density residential locations included between two and four dwellings per 0.4 ha. Medium density residential locations also include some areas with substantial amounts of non-residential land-use, including commercial, institutional, or even vacant properties. Sites assigned to this category are referred to here as mixed use/medium density residential (MMDR). Descriptive information about the land-use classification methods applied at each site can be found in supplemental information.

2.3. Sample collection

All wells were sampled once and most were sampled during the fall 2013. All wells are shallow and ranged in depth from 1 to 6 m below land surface (Table S1). Most samples were collected using a hand-driven piezometer with 6.35 cm screen. Observation wells KT07, FR2S, FR3S, SR12, and SR16 had 1.5 m screens; SH01 a 4.6 m screen (table S1). Groundwater samples were collected using a peristaltic pump equipped with a small piece (less than 30 cm) of Masterflex C-flex tubing. Polytetrafluoroethylene-lined polyethylene tube was fitted to the drive-point piezometer head, through the frame (if needed for temporary well installation), and out to the C-Flex tubing end at lengths required to reach depths just below the water table. Sampling procedures outlined in the U.S. Geological Survey National Field Manual (variously dated) were followed. Guidelines in the field manual were used to determine when each well was adequately purged to collect a groundwater sample representative of each location. Samples were collected and analyzed using laboratory methods for nutrients, pharmaceuticals, and EACs (hormones) at each location. Nutrient samples were filtered through an inline 0.45 μm capsule filter. Pharmaceutical and EACs samples were filtered through an inline Teflon filter assembly with a baked 0.7 μm glass-fiber filter. All samples were placed on ice and shipped overnight to the USGS National Water Quality Laboratory (NWQL) in Denver, Colorado.

2.4. Sample analysis methods

All samples were analyzed for the same suite of nutrients, pharmaceuticals, and hormones (EACs) at the NWQL by methods (with the same reporting levels (RLs)) also used in the 2011 study conducted on Fire Island (Phillips et al., 2015), which allows for direct comparison between samples collected at the same locations. The nutrient method analyses determined ammonium, nitrite, nitrate plus nitrite, and orthophosphate as described by Fishman (1993) and Patton and Kryskalla (2011). Nutrient method reporting levels are shown in table S3. The pharmaceutical method determines 110 compounds (all but 7 of which are classified here as pharmaceuticals or their degradates; see table S4) using direct-aqueous injection (100 μL) high-performance liquid chromatography coupled to a triple-quadrupole mass spectrometer with an electrospray ionization source operated in the positive-ion mode (Furlong et al., 2014). Reporting levels range from 2.24 ng/L to 199 ng/L for this method (table S4). The hormone method uses solid-phase extraction of a 0.5-L sample, and analysis by gas chromatography with tandem mass spectrometry with concentrations quantified using an isotope dilution procedure (Foreman et al., 2012). The hormone method determines 20 analytes including estrogens, androgens, progesterones, and additional EACs, with reporting levels ranging from 0.8 to 8 ng/L for hormones, 100 ng/L for bisphenol A (BPA), and 200 ng/L for 3β-coprostanol (COP) and cholesterol (CHO) (table S5).

Concentrations reported as "estimated" (E) are given where the detected concentration was less than the laboratory reporting limit or less than the lowest calibration standard (Childress et al., 1999). In some cases, concentrations were reported below the method detection limit. These concentrations indicate a detection of the compound, but were not used for subsequent calculations reported here and were reassigned DNQ (detected but not quantified) in Tables S6, S7, S9, and S10.

2.5. Calculations and statistical methods

Nitrogen and pharmaceutical concentrations are presented in this study as total concentrations. Calculating and presenting the data in this manner makes it easier to summarize overall occurrence and differences of nitrogen and pharmaceuticals detected in the samples. Total inorganic nitrogen (TIN) was calculated as the sum of nitrite plus nitrate and ammonium. Ammonium dominant samples are samples where all (100%) of the inorganic nitrogen detected using this method is present in the form of ammonium. Additional nutrient analysis methods used for this study are available in the Supplemental Material. Total pharmaceutical concentration is given as the sum of quantified pharmaceutical concentrations per site.

The Tukey test, a non-parametric test of association, was used to assess differences in concentrations between wells within different land use settings. Non-parametric statistical analysis is commonly used in water-quality studies because data are typically not normally distributed (Helsel and Hirsch, 2002). The SAS software (version 9.4) was used to perform the statistical analyses.

2.6. Quality assurance

Quality-assurance samples included one equipment blank and three replicate samples. All of these samples were analyzed using the nutrient, pharmaceutical, and hormone methods. The equipment blank was prepared using inorganic-free water (for nutrient method) and organic-free water (for pharmaceutical and hormone method) and the same tubing and pump assembly used to collect all environmental samples. None of the compounds in any of the three methods were detected in the equipment blank.

Three replicates for the pharmaceutical method had 27 detections; most (23) of these had a detection in both the environmental and replicate sample (Table S6). Median relative percent differences were less than 30% for compounds in each replicate set. Four compounds had detections in either the environmental or the replicate sample, but not both; these are referred to as unmatched replicates. The concentrations for the detections in the unmatched replicate comparisons were between the method detection limit and the reporting limit, or below the method detection limit.

Three replicate samples were collected and analyzed for the hormone method (Table S7). Three compounds were detected in these three samples: cis-androsterone (CAND), estrone (E1), and dihydrotestosterone (DHT). CAND was detected in both the replicate and the environmental sample at one site, with a difference of 43%. The two other compounds were unmatched replicates having a detection in either the environmental or the replicate sample, but not both. The concentrations of these two analytes in the unmatched replicate were low: E1 concentration was only 0.8 ng/L, which is at the reporting limit; and the detection of DHT was less than the method detection limit.

3. Results and discussion

3.1. Concentrations of nutrients, pharmaceuticals, and endocrine active compounds in the Sandy Hook, New Jersey, network

Concentrations of TIN and the total pharmaceuticals (Figs. 2, S2, and S3), as well as the total number of pharmaceuticals detected (Figs. 3 and S4), were highest in the samples collected at the leach fields and decreased with increasing distance from the leach fields (Fig. S1). Ammonium was the predominant nitrogen species for four of the five Sandy Hook sites (Table S8), with a concentration range of detections from 0.69 to 5.3 mg/L (Fig. 2). The site furthest from the leach fields (wastewater source), SH04, was the only site without a detection of ammonium, nitrate, or nitrite. There is a correlation between the distance from the leach field and decreasing ammonium concentrations that shows the impact of the OWDS on the downgradient groundwater system.

Nineteen of the 103 pharmaceuticals analyzed for were detected in samples collected from Sandy Hook (table S9). Carbamazepine was detected in four of the five wells and the highest detected concentration was 214 ng/L (SH02). Acyclovir, an antiviral, was the highest concentration (284 ng/L, SH01) detected of all the measured pharmaceuticals, and was detected in two of the five (40%) wells sampled. Furlong et al. (2014) note that matrix enhancement can occur for some pharmaceuticals in their method; this effect was observed for acyclovir in fortified (spike) surface water, effluent, and influent samples. Thus, these results may overestimate true concentrations. Other pharmaceuticals detected (frequency shown if more than one well) included bupropion (60%), carisoprodol, desvenlafaxine (40%), diazepam (40%), famotidine, fexofenadine, fluconazole, lidocaine (60%), metaxalone, metoprolol, nevirapine, oseltamivir, propoxyphene, pseudoephedrine + ephedrine, temazepam (40%), tramadol, and warfarin.

Total pharmaceutical concentrations and TIN are positively correlated for the Sandy Hook samples reflecting the co-occurrence of nutrients and CECs along this flowpath (Fig. 2). These results show that a single OWDS serving more than two million people annually can result in elevated nitrogen and CEC concentrations up to 450 m downgradient of a wastewater source (table S1, Figs. S2 and S3). Although these results do not fully capture the impact of the OWDS at Sandy Hook (i.e., groundwater from deeper in the aquifer was not sampled, high-use season was not sampled), these results provide some of the first CEC data and relates these to nutrient concentrations in the shallow groundwater adjacent to Sandy Hook Bay.

Only two of the 20 analytes included in the hormone method were detected at low quantifiable concentrations in samples from Sandy Hook (Table S10). Estrone (E1), an estrogen, was detected at two locations, SH01 and SH05. Cholesterol was detected at one location (SH05) at a concentration of 311 ng/L.

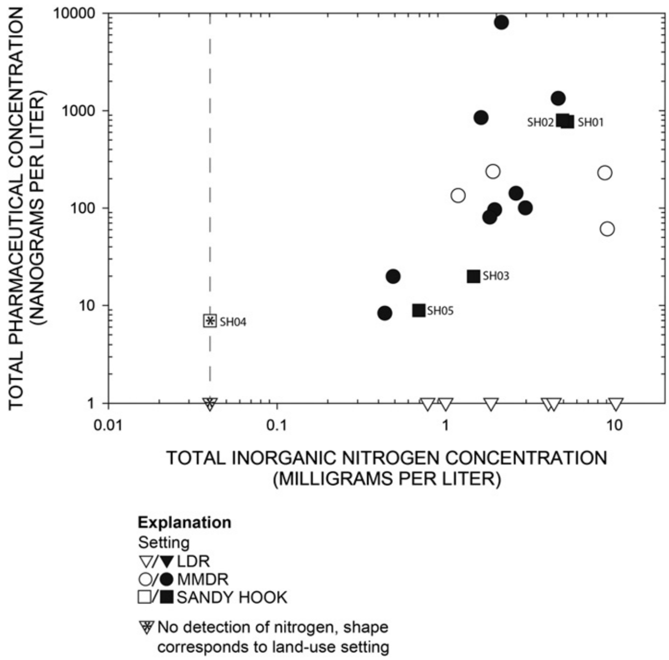

Fig. 2. Total pharmaceutical concentration as a function of total inorganic nitrogen (TIN) and grouped by setting. LDR = low density residential, MMDR = mixed-use/medium density residential. Dashed gray line shows method detection limit (MDL) for nitrogen analysis (0.04 mg/L). Open symbols are samples where nitrate was the predominant nitrogen species; closed symbols are samples where ammonium was the predominant nitrogen species, except for symbols plotted at the MDL. Total pharmaceutical concentrations plotted at 1 ng/L represent no quantifiable pharmaceutical detections.

3.2. Concentrations of nutrients, pharmaceuticals, and endocrine active compounds in the New York network

Concentrations of many wastewater-associated compounds are higher at sites in the MMDR setting compared to the LDR setting, indicating OWDS density as an important factor affecting shallow groundwater quality in near-coastal settings. Twenty-nine of 103 pharmaceuticals measured were detected at least once at sample sites in NY (table S6). Other detected CECs include PCDUs (caffeine, nicotine, and metabolites), methyl-1H-benzotrizole (a corrosion inhibitor), and piperonyl butoxide (a pesticide synergist). Lidocaine, an over-the-counter topical anesthetic, was the most commonly detected pharmaceutical (35% of NY samples). Other commonly detected pharmaceuticals included fexofenadine (an over-the-counter antihistamine detected in 30% of samples), and carbamazepine, desvenlafaxine (an antidepressant), meprobamate, metformin (antidiabetic), and tramadol (an opioid) each detected in 25% of the samples). Just two LDR sites (JP01 and SR03T) had detections of pharmaceuticals, but these were not quantified (table S6). Total pharmaceutical concentrations ranged from 19.8 ng/L to 8060 ng/L for sites classified as MMDR (Figs. 2, and S3; table S11). Meprobamate, an anxiolytic drug and also the primary active metabolite of carisoprodol, was the highest measured concentration (6020 ng/L at site MKB01) for any pharmaceutical and was detected in five samples. Other pharmaceuticals having relatively higher concentrations include carisoprodol (1870 ng/L) and desvenlafaxine (837 ng/L), which were detected at MKB01 and KB02, respectively. The lack of pharmaceuticals detected in LDR settings is likely due to the lower loading of wastewater, but may also be attributed to complex biogeochemical conditions of the shallow surficial aquifer. Microbial degradation of most organic wastewater-associated compounds increases in oxic conditions, with the exception of carbamazepine and sulfamethoxazole, which are both resistant to biodegradation and very persistent in groundwater (Sui et al., 2015; Fram and Belitz, 2011). Both carbamazepine and sulfamethoxazole have been detected in oxic groundwater at Cape Cod, Massachusetts (Schaider et al., 2016; Swartz et al., 2006), but not in oxic/suboxic (<1 mg/L) conditions for our study. However, the few detections of PCDUs (caffeine) suggest groundwater in LDR settings are still influenced by wastewater sources, but to a lesser extent than MMDR settings (Fig. S5).

Although concentrations of TIN did not differ significantly ($p = 0.062$) by land-use category for the NY sites, ammonium concentrations are higher at MMDR sites (Figs. 2 and S2, table S12). The form of nitrogen in most of the samples collected from MMDR settings was predominantly ammonium, while all samples collected in the LDR setting were dominated by nitrate. Samples with ammonium as the predominant nitrogen ion had a range of TIN from 0.44 mg/L to 4.66 mg/L; well KB02 had the highest ammonium concentration. Samples that were predominantly nitrate had a range of TIN from 0.78 mg/L to 10.2 mg/L (Table S12). Only one site, SR16, did not have a measurable concentration of nitrogen in any form. The differences in nitrogen species present in the two land-use settings suggest variations in nitrogen source and/or biogeochemical conditions. Nitrate in the sites classified as LDR suggests the primary source of nitrogen may not be derived entirely from sewage. Other potential sources of nitrogen could be from fertilizer application or atmospheric deposition (Kroeger et al., 2006b; Stinnette, 2014). An additional factor could be the semi-oxic (1 mg/L) conditions (table S1) within the permeable (coastal) sediments in LDR settings may potentially induce biotransformation of ammonium to nitrate.

Total pharmaceutical concentrations generally increase with increasing TIN concentrations for the NY wells classified as MMDR land use (Fig. 2; Fig. S5). This suggests that the most probable source of nitrogen in the groundwater at these settings is a septic source, and indicates that the presence of pharmaceuticals in the near-shore aquifer reflects use upgradient. Two MMDR sites sampled downgradient of a restaurant and a small hotel (<50 occupants) have the largest number of pharmaceuticals detected (>15) (Table S11), reflecting the importance of different demographics, population served, and water use. Sample collection of the shallow groundwater close to these institutions (within 180 m) ensures a representative sample of the most septic-influenced groundwater. Wastewater discharged to OWDS of a hotel or restaurant

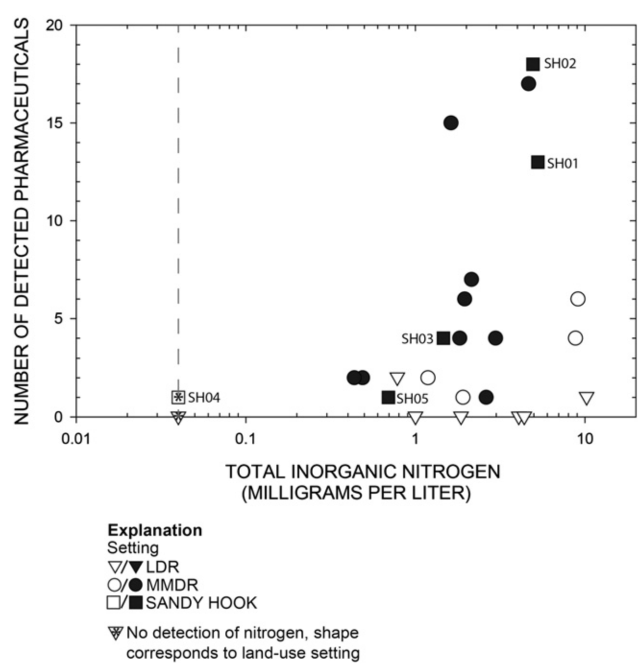

Fig. 3. Number of detected pharmaceuticals (including DNQs) as a function of total inorganic nitrogen (TIN) and grouped by setting (Table S2). LDR = low density residential, MMDR = mixed-use/medium density residential. Dashed gray line shows method detection limit (MDL) for nitrogen analysis (0.04 mg/L). Open symbols are samples where nitrate was the predominant nitrogen species; closed symbols are samples where ammonium was the predominant nitrogen species, except for symbols plotted at the MDL.

typically contains a wider variety, higher concentrations, and a greater load of CECs than residential OWDS because these commercial establishments serve a greater number of individuals. The wider range in the total pharmaceuticals concentration values (8.3 to 8060 ng/L) for the MMDR category likely is representative of the demographic variability and water use in the seasonal tourist population.

Hormones were infrequently detected in samples from the NY sites; when detected, these were generally present in Fire Island wells, or in select MMDR wells (table S7). Two androgens (CAND and DHT) were detected at two locations: CAND and DHT at KT07 and DHT at FH01. CAND was the only androgen detected above the method detection limit. Two estrogens, E1 and estriol (E3), were detected in three samples and one sample, respectively. E1 was detected at concentrations ranging from 0.8 to 6.4 ng/L; these detections occurred in three MMDR wells (KB02, KT07, and WORC01); the highest estrogen concentration (6.4 ng/L) was detected in the sample from WORC01. E3 was detected at well KB02 with a concentration of 2.3 ng/L. Other compounds from the hormone method with only one detection each in the NY network include CHO at a concentration of 208 ng/L at SGH01 and the endocrine-disrupting compound BPA at a concentration of 442 ng/L in KB02. The limited detections of hormones may be a result of biodegradation and (or) sorption (to organic carbon in aquifer sediments) processes during their transport from OWDS to groundwater. Although detections of hormones were relatively rare, these data do indicate that septic systems can act as sources of androgens and estrogens.

Land use is an important factor in the presence of pharmaceuticals and nitrogen in groundwater; however, geochemical conditions (i.e., redox conditions) may be another important factor. The differences in nitrogen species between locations are consistent with anoxic conditions in MMDR settings (median < 1 mg/L DO) and suboxic/oxic conditions in LDR settings (median 1.2 mg/L DO). However, for our study, the lack of carbamazepine indicates that redox conditions do not affect pharmaceutical concentrations in LDR settings.

3.3. Occurrence of pharmaceuticals and mixtures of pharmaceuticals in samples

The occurrence of pharmaceuticals in groundwater downgradient of OWDS depends on many factors, including the extent of loading upgradient, land use along the groundwater flow path, time of transport through the unsaturated zone, potential for biodegradation, and solubility (Li, 2014; Lapworth et al., 2012). Of the 103 pharmaceuticals measured in the study, 43 were detected, with as many as 18 detected within a given sample (SH02). The occurrence of pharmaceuticals in groundwater samples was summarized using two approaches: 1) occurrence of individual pharmaceuticals in samples, and 2) occurrence of select pharmaceuticals in samples with mixtures of four or more pharmaceuticals. These approaches are complementary; although most studies focus on the individual occurrence of pharmaceuticals (the former approach), the latter approach focuses on the occurrence of pharmaceutical combinations of compounds, which offers insights into the prevalent mixtures present in samples.

Seven pharmaceuticals were frequently detected in the 25 samples collected in the study. The most commonly detected compounds present include lidocaine and carbamazepine, which were present in 40% and 36% of the samples, respectively (Fig. 4). Other pharmaceuticals detected in 24–32% of the samples included fexofenadine, bupropion, desvenlafaxine, meprobamate, and tramadol.

A national survey of source water (i.e., groundwater and surface water drawn for public supply) identified carbamazepine, erythromycin-H_2O (erythromycin metabolite), trimethoprim (antibiotic), sulfamethoxazole (an antibiotic), diphenhydramine (antihistamine), dehydronifedipine (antianginal), codeine (analgesic), diltiazem (antihypertensive), and fluoxetine (antidepressant) as commonly detected pharmaceuticals (Focazio et al., 2008). Of this list, only carbamazepine and sulfamethoxazole were detected in our study, and these were not the most frequently detected compounds (erythromycin was included in our study, but not its metabolite). Many studies have identified the antibiotics sulfamethoxazole (Barnes et al., 2008; Standley et al., 2008; Godfrey et al., 2007; Vulliet and Cren-Olive, 2011) and trimethoprim (Schaider et al., 2014; Standley et al., 2008) as commonly present in groundwater downgradient of OWDS. Sulfamethoxazole was detected in just two of the 25 samples collected from the entire region (FR2S and EQ01). For our study, five antibiotics (tables S6 and S9) were included in the analysis; however, they were not frequently detected and thus would not be good indicators of wastewater influence on groundwater quality for this region.

Several studies have identified select pharmaceuticals to use as indicators of wastewater influenced groundwater. One example is carbamazepine (Arnold et al., 2014; Zhao et al., 2011; Katz et al., 2009; Godfrey et al., 2007), which was commonly found in the complex mixture samples in our study, but that was not detected as frequently as lidocaine, fexofenadine, bupropion, desvenlafaxine, and tramadol (Fig. 4). Samples with four or more pharmaceuticals present were classified as complex mixture samples for this study; thus, slightly less than half (44%) of the samples collected were classified as complex mixtures. Eight of the 11 sites having complex pharmaceutical mixtures were collected at MMDR sites, with the remainder collected from Sandy Hook, NJ. None of the LDR sites had a complex mixture sample.

The occurrence and transport of the pharmaceuticals detected in this study vary in groundwater and depends on the load to the OWDS (usage by population served) and their solubility. Typically, greater amounts and higher detections of CECs reflect a consistent load to the groundwater system and proximity to wastewater source (Conn et al., 2006; Swartz et al., 2006). Limited detections of CECs at some sites (WORC01, SH04, and SH05) suggest geochemical processes decrease their transport in the aquifer. Carbamazepine has been found to be very persistent in groundwater and can be readily transported through the aquifer further from the wastewater source (Godfrey et al., 2007).

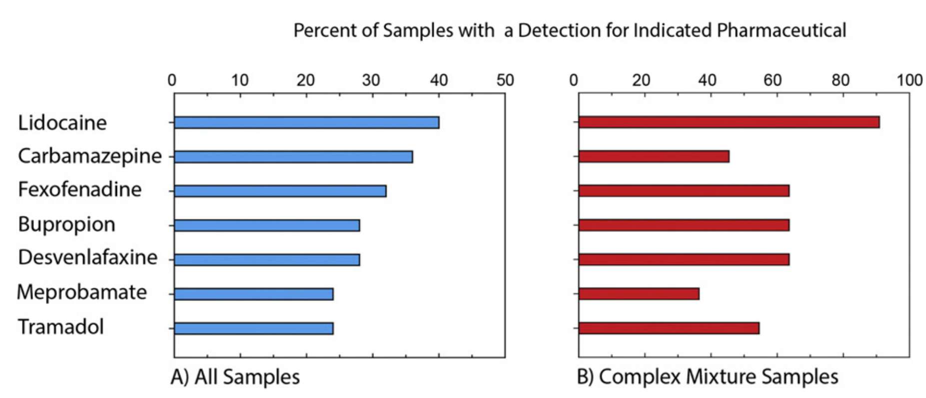

Fig. 4. Percent of A) all samples, and B) complex mixture samples with a detection for indicated pharmaceuticals for 25 groundwater samples collected in New York and New Jersey downgradient of onsite wastewater disposal systems. [Complex mixture samples classified as those with detections of four or more pharmaceuticals].

Five pharmaceuticals detected in our study have not been widely evaluated or documented in the United States: lamivudine (antiretroviral), triamterene (diuretic), alprazolam (antianxiety), penciclovir (antiviral), and thiabendazole (antifungal). Lamivudine was detected in three of our samples with the highest concentration of 22.9 ng/L, and is seldom found in the literature related to environmental fate and transport in groundwater. It is readily removed during wastewater treatment (K'oreje et al., 2016; Prasse et al., 2010); however, it has been widely detected in Kenyan wastewater effluent and river water, and it is attributed to the high pharmaceutical consumption of antiretrovirals to treat widespread diseases among the population (K'oreje et al., 2016).

Given the high variability in type of pharmaceuticals detected for our study in comparison to other studies (Schaider et al., 2014; Conn et al., 2010; Standley et al., 2008), it seems prudent to analyze for a wide variety of CECs introduced by OWDS when conducting a regional study.

3.4. Comparison of nutrient, pharmaceutical and hormone concentrations before and after Hurricane Sandy at Fire Island, New York

Pre-Hurricane Sandy groundwater data (November 2011) exist for three locations at Fire Island, NY (KB01, KB02, and KT07) (Phillips et al., 2015). TIN concentrations in samples collected before (2011) and after (2013) Hurricane Sandy were unchanged at KB01, 75% lower at KB02 (18.4 mg/L in 2011 and 4.7 mg/L in 2013) and nearly 80% higher at KT07 (0.3 mg/L in 2011 and 1.6 mg/L in 2013) (Fig. 5; table S13). Ammonium was the predominant form of nitrogen at all three locations in samples collected before and after Hurricane Sandy.

There is less than a 10% difference between total pharmaceutical concentrations for KB01 and KB02 before and after Hurricane Sandy (Fig. 5). A similar number of pharmaceuticals were also detected at each site for each year (Tables S14–S15). In contrast, the total pharmaceutical concentration increased between 2011 and 2013 by 87% for site KT07 (107 ng/L and 845 ng/L, respectively). The number of pharmaceuticals detected at KT07 increased from four in 2011 to 15 in 2013 (a 375% increase) (Table S15). Considering all three sites, an additional 12 pharmaceuticals (acyclovir, bupropion, carisoprodol, diphenhydramine, lamivudine, methadone, nadalol, oxycodone, phenytoin, propoxyphene, thiabendazole, and warfarin) were detected after the hurricane, while just one (metaxalone) of the 13 pharmaceuticals detected in samples collected before the hurricane was not detected after the storm.

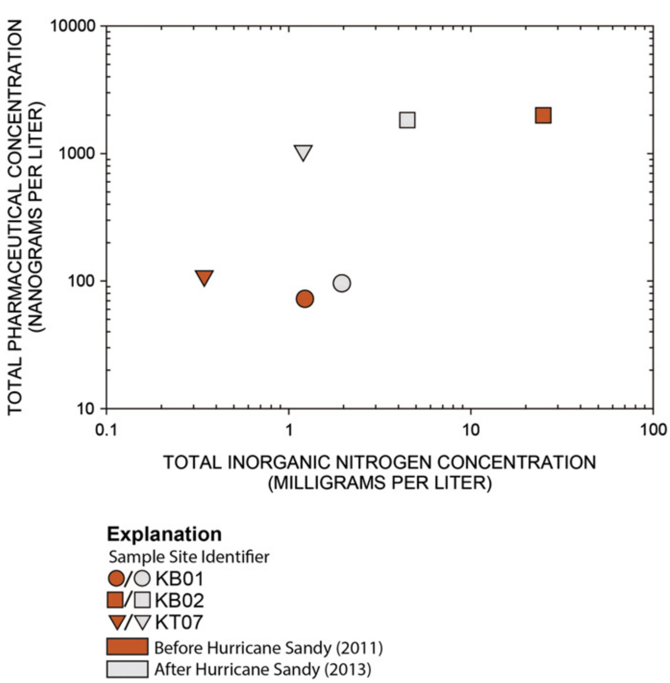

Fig. 5. Total pharmaceutical concentration as a function of total inorganic nitrogen (TIN) before and after Hurricane Sandy at three sites on Fire Island, NY. Orange symbols represent samples collected before Hurricane Sandy (2011) and gray symbols represent samples collected after Hurricane Sandy (2013).

Few EACs were detected in Fire Island samples before and after Hurricane Sandy (Table S16). CAND and E1 were detected at site KT07 in 2013, but not in 2011. Estrogens E1 and E3 were detected in both before and after Hurricane Sandy samples collected at KB02. BPA was detected in 2011 at KB01 and KB02, but only at KB02 in the 2013 sample.

Overall, total pharmaceutical concentrations did not change much in samples collected before and after Hurricane Sandy except at one location. Although many factors could contribute to the difference in the patterns among the sites for the 2011 and 2013 samples, it appears that saltwater inundation at these locations on Fire Island did not decrease concentrations and the observed increase (especially at KT07) may reflect other factors besides Hurricane Sandy. The proximity of KT07 to a seasonal restaurant, combined with the collection of the 2013 samples in September may have resulted in a greater effect of the OWDS associated with the restaurant during this period compared to the 2011 sample, which was collected in December, three months after the summer tourist season. Data from KB01 and KB02 indicate a significant influence of wastewater on the groundwater beneath Fire Island, and that continued monitoring of the Fire Island groundwater is needed to adequately characterize the seasonal and long-term changes in nutrients, pharmaceuticals, hormones, and other CECs from OWDS in this region.

4. Conclusions

Onsite wastewater disposal systems in the coastal regions of New Jersey and New York contribute nutrients, pharmaceuticals, and other CECs to downgradient shallow groundwater in nearshore settings. In our study, the correlations between TIN concentration, number of detected pharmaceuticals, the total pharmaceutical concentration, and the distance between the sample location and wastewater source (leach field in NJ; homes and institutions in NY) were presented to show the impact of the OWDS on the shallow groundwater system. Land-use settings with the greatest influence on downgradient wells were in locations that catered to the greatest number of people, regardless of the type of OWDS (centralized or cesspools/septic systems). In Sandy Hook, samples collected closest to the wastewater treatment plant leach fields had the highest measured TIN concentration, total pharmaceutical concentration, and the greatest number of pharmaceuticals detected. In NY, samples collected from MMDR settings had the highest total pharmaceutical concentrations. Those settings in NY that also had a restaurant or a small hotel within 180 m of the well had the greatest number of pharmaceuticals detected. The variety of pharmaceuticals detected at each setting is therefore directly related to the use by the surrounding population in this study area and appears to vary among sites.

Several studies have identified specific pharmaceuticals or PCDU compounds as indicators of the presence of wastewater in groundwater; however, limiting the analysis to a single commonly-used wastewater-associated compound (e.g., carbamazepine) is not always as useful as screening for a wide variety of pharmaceuticals. Results of our study demonstrate the importance of analyzing for a wide range of wastewater-associated compounds (such as over-the-counter medications, prescription drugs, and human hormones) in regional studies to account for varied demographics (Tables S6 and S7). Demographics and population densities are the controlling factors for the variety and measured concentration of pharmaceuticals, PCDU, and other CECs detected in groundwater influenced by OWDS (as described by Conn et al., 2006).

Hormones were infrequently detected in samples collected during our study; however, these data indicate that OWDS can be a source of androgens, estrogens, and other EACs, particularly in settings with a higher density of OWDS. It is important to document the presence of EACs and pharmaceuticals, even when they are detected at very low concentrations, because of the potential for adverse effects of mixtures on receiving aquatic ecosystems.

Comparisons between existing groundwater data collected before Hurricane Sandy (2011) and after Hurricane Sandy (2013) at three locations on Fire Island, NY, showed an increase in TIN at two locations and a decrease at the third location (Fig. S5). The number and total concentration of detected pharmaceuticals significantly increased at just one of the three locations. Several factors may be contributing to patterns observed for the 2011 and 2013 samples that appear unrelated to the effects of seawater inundation brought by Hurricane Sandy, though additional storm-related sampling would be required to be certain. These factors include the proximity of a restaurant to one sample location and the different sample collection months (December 2011 and September 2013) in relation to the high summer tourist season. Nevertheless, the data clearly show that OWDS have a continuous influence on groundwater quality beneath Fire Island. Sustained monitoring of the groundwater that will ultimately seep into Great South Bay is needed to characterize the seasonal, long-term, and storm-related changes in nutrients and CECs in groundwater originating from OWDS.

This study provides a baseline for wastewater influences on groundwater quality in these coastal communities and might be indicative of the quality in similar settings. Results from studies that focus on shallow, near-shore groundwater quality are important because coastal communities are vulnerable to extreme storms (e.g., hurricanes), flooding events, and sea-level rise; all of which can damage wastewater infrastructure and lead to biogeochemical changes that disrupt the level of onsite treatment and result in increased discharge of contaminants to estuaries through groundwater seepage. Future studies on the effects of inundation on OWDS may benefit by evaluating the microbial community (including antibiotic resistance) in shallow groundwater settings. Furthermore, these data can be used to improve our understanding of the fate and transport of CECs and their co-occurrence with nitrogen in shallow groundwater, which is necessary for defining and predicting the resiliency of OWDS in coastal settings.

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