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REPORT

Complex interactions between climate change, sanitation, and groundwater quality: a case study from Ramotswa, Botswana

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Abstract
Groundwater quantity and quality may be affected by climate change through intricate direct and indirect mechanisms. At the same time, population growth and rapid urbanization have made groundwater an increasingly important source of water for multiple uses around the world, including southern Africa. The present study investigates the coupled human and natural system (CHANS) linking climate, sanitation, and groundwater quality in Ramotswa, a rapidly growing peri-urban area in the semi-arid southeastern Botswana, which relies on the transboundary Ramotswa aquifer for water supply. Analysis of long-term rainfall records indicated that droughts like the one in 2013–2016 are increasing in likelihood in the area due to climate change. Key informant interviews showed that due to the drought, people increasingly used pit latrines rather than flush toilets. Nitrate, fecal coliforms, and caffeine analyses of Ramotswa groundwater revealed that human waste leaching from pit latrines is the likely source of nitrate pollution. The results in conjunction indicate critical indirect linkages between climate change, sanitation, groundwater quality, and water security in the area. Improved sanitation, groundwater protection and remediation, and local water treatment would enhance reliable access to water, de-couple the community from reliance on surface water and associated water shortage risks, and help prevent transboundary tension over the shared aquifer.

Keywords Climate change · Nitrate · Socioecology · Botswana · Sub-Saharan Africa

Introduction

Groundwater supports about 75% of the Sub-Saharan African (SSA) population as well as industry and some crop irrigation (Calow and MacDonald 2009; Villholth 2013; Niang et al. 2014), and water insecurity across SSA results from a complex interplay of natural and social systems (Howard and Bartram 2010). Groundwater and surface-water quality and quantity are affected by changes in climate, land use, demographics and economic activity (Jiménez Cisneros et al. 2014); therefore, ensuring water security in SSA under a changing climate requires an understanding of these interacting effects on groundwater (Cronin et al. 2007; Taylor et al. 2013; Famiglietti 2014; Niang et al. 2014).

Rapid urbanization in SSA is another driver of change in groundwater and frequently means that infrastructure, including

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sanitation, lags behind the needs of the growing population. Pit latrines are the most common means of human waste disposal, often in areas where communities also rely on groundwater for drinking (Cronin et al. 2007; Lapworth et al. 2017). Unlined pit latrines leach to the groundwater, threatening groundwater quality and human health with pollutants like nitrate (NO_3^-) and pathogens (Cronin et al. 2007; Graham and Polizzotto 2013). In studies of pit latrine impacts on groundwater quality, NO_3^- is the most commonly detected pollutant (Graham and Polizzotto 2013). Though safe levels of NO_3^- for humans in drinking water are still debated (Schullehner et al. 2018), it is clear that this is a critical and widespread pollutant.

Elevated concentrations of NO_3^- have been found in arid environments, including the Kalahari Desert, where it is believed to be the product of the decomposition of ancient vegetation deposits (Heaton et al. 1983; Stadler et al. 2012; Stone and Edmunds 2014). Stadler et al. (2012) differentiated between naturally occurring and anthropogenic NO_3^- in groundwater in the Kalahari using stable isotopes: NO_3^- in the deeper groundwater was older in age and tended to be naturally occurring, while NO_3^- in the shallower, younger groundwater tended to come from human and cattle sources. In a review of studies of NO_3^- pollution in groundwater across southern Africa, the main source of NO_3^- pollution, despite naturally occurring NO_3^-, was anthropogenic activities, including pit latrines and livestock feedlots (Tredoux and Talma 2006). Tillage and associated nitrification of organic matter can be a minor source of NO_3^- leaching. The agricultural soils in inland southern Africa tend to be low in N, and where inorganic N fertilizer is used, it is typically applied at low rates (Tredoux and Talma 2006), though fertilizer application rates may be increasing.

In order to identify health risks associated with groundwater quality and identify remediation options, especially for NO_3^-, in these settings it is important to understand the contamination source. However, discerning between human and livestock waste contamination can be a challenge: humans and livestock have similar NO_3^- stable isotope signatures and fecal indicator bacteria (Ashbolt et al. 2001; Fenech et al. 2012). In recent years, a suite of chemicals called emerging organic contaminants (EOCs), including pharmaceuticals and caffeine (1,3,7-trimethylxanthine), have been used as tracers of human contamination. These substances tend to persist in the environment, and detection limits continue to decrease (Fenech et al. 2012; Lapworth et al. 2012). Caffeine's metabolite paraxanthine (1,7-dimethylxanthine) has also been used, but not as widely. Caffeine and/or paraxanthine have been measured in groundwater and surface water in Europe and the US (Buerge et al. 2003; Glassmeyer et al. 2005; Godfrey et al. 2007; Hillebrand et al. 2012; Reh et al. 2013; Stuart et al. 2014; Hillebrand et al. 2015) and more recently in surface waters, urban groundwater, and roof-harvested rain water in South Africa (Matongo et al. 2015; Sorensen et al. 2015; Waso et al. 2016; Wanda et al. 2017; Rodríguez-Gil et al. 2018).

Denitrification is a naturally occurring microbial process that removes NO_3^- from groundwater. In low oxygen conditions, denitrifying microbes reduce NO_3^- into nitrous oxide (N_2O) and di-nitrogen (N_2) gases that either escape to the atmosphere or, in a closed system like an aquifer, remain in the water in dissolved form (Seitzinger et al. 2006; Robertson and Groffman 2015). Heterotrophic denitrification involves several subreactions (one of which produces N_2O), such that the complete denitrification process can be written as (Schlesinger and Bernhardt 2013):

$$5CH_2O + 4H^+ + 4NO_3^- \rightarrow 2N_2 + 5CO_2 + 7H_2O \quad (1)$$

Given the aforementioned stoichiometry, denitrification requires dissolved organic carbon (DOC), the electron donor, and NO_3^-, the electron acceptor, in a ~1:1 molar ratio. Where naturally occurring concentrations of DOC in the groundwater limit denitrification, bioremediation technologies inject DOC into the aquifer to enhance denitrification (USEPA 2013).

Scientists have warned that direct anthropogenic impacts on groundwater quality could be more consequential than any direct impacts of climate change on groundwater quality (Taylor et al. 2009; Calow et al. 2010; Ferguson and Gleeson 2012), although some of these anthropogenic effects may be the indirect result of climate change, as for example where climate change alters human behavior that in turn has an impact on groundwater. However, few studies have looked at the indirect effects of climate change on sanitation and nitrate pollution of groundwater (Howard et al. 2016). Climate change is happening in southern Africa. Over the last 50–100 years, most of southern Africa's mean, maximum, and minimum temperatures have risen, especially in the last two decades (Niang et al. 2014). In this century, mean temperatures in southern Africa are projected to increase faster than the global average, especially in semi-arid areas like Botswana. Between 1950 and 2000, rainfall in Botswana and neighboring countries has been declining (Niang et al. 2014). Climate models project decreasing annual mean rainfall for Botswana and Namibia, with drier and shorter wet seasons (Niang et al. 2014); thus, there is a need for understanding how climate change affects human behaviors that can affect groundwater quality.

The study system, the Ramotswa aquifer and community, is a coupled human and natural system (CHANS; Liu et al. 2007), meaning there are strong interactions between the biophysical and social systems, where changes in one can drive changes in the other. An interdisciplinary CHANS approach was used to investigate these interactions, i.e., how climate affects water supply infrastructure and sanitation access in Ramotswa, and how those changes impact the quality of the

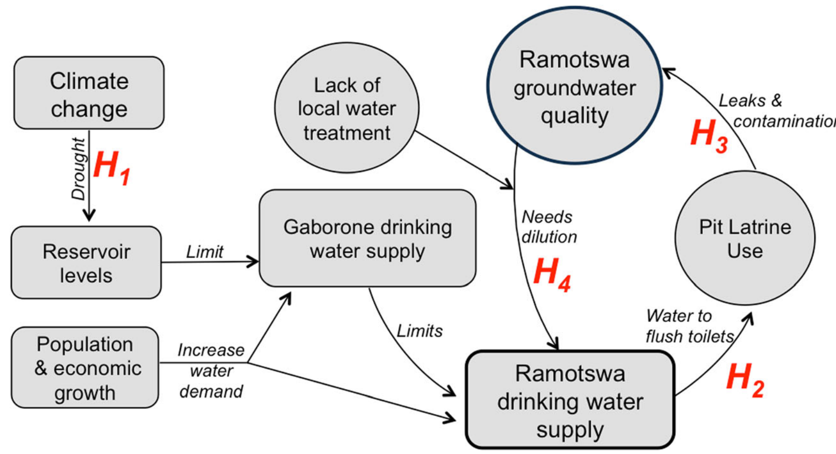

Fig. 1 Conceptual diagram of Ramotswa groundwater coupled human and natural system (CHANS) illustrating how climate change could affect Ramotswa drinking water quantity and groundwater quality. H_1–H_4 in red represent the four hypotheses explained at the end of the 'Introduction'

groundwater (Fig. 1). CHANS methods offer a powerful template for investigating feedbacks between biophysical and social systems like those previously discussed. In fact, Sivapalan et al. (2012) argued that it is impossible to manage or predict water availability without accounting for the feedbacks between water and human systems, because human activity has such pervasive effects on the modern water cycle. The CHANS literature demonstrates that in order to characterize today's most pressing socio-ecological issues, interdisciplinary research approaches are required to understand their complexities and develop realistic solutions (Millenium Ecosystem Assessment 2005; Liu et al. 2007; Collins et al. 2011; Stevenson 2011).

In this paper climate data analyses, sociology, and biogeochemistry are integrated to capture the socio-ecological interactions potentially driving groundwater pollution in Ramotswa. The overarching research question is: "*Does climate change impact groundwater quality in Ramotswa, and if so, how*?" Specifically, the following hypotheses are tested (Fig. 1):

H_1: Climate change-induced droughts are becoming more frequent.
H_2: Water shortages affect sanitation behavior, increasing pit latrine use.
H_3: Nitrate pollution primarily originates from human waste contamination.
H_4: Enhanced denitrification via in situ bioremediation of the Ramotswa aquifer has the potential to reduce nitrate contamination.

This work represents several innovations: the application of a CHANS approach to a groundwater system potentially impacted by climate change, the measurements of both nitrate and caffeine in groundwater in Africa (cf. Sorensen et al. 2015; Rodríguez-Gil et al. 2018), and a preliminary assessment of the potential for in situ bioremediation (ISB) of nitrate pollution in the Ramotswa Aquifer.

Study area

Biophysical characteristics

The peri-urban town of Ramotswa (24.88°S × 25.87°E) is semi-arid and subtropical, about 25 km^2 in size and 20 km south of Botswana's capital, Gaborone (Fig. 2). The town's eastern boundary is the ephemeral Notwane (Ngotwane) River, which flows northward to the Limpopo River, and serves as the boundary with South Africa in this area.

The rainfall is highly seasonal: the wet season (summer) lasts from October to March, and the dry season (winter) is from April to September. Therefore, the water year is from 1 October through 30 September. The mean annual (water year) rainfall in Ramotswa is 454 mm (1980–2016), 89% of which falls in the wet season (data from Botswana Dept. of Meteorological Services). Dry season average rainfall is 47 and 64 mm (1980–2016) in Ramotswa and Gaborone, respectively. Average annual evaporation from two nearby reservoirs in South Africa is 2,120 mm (Altchenko et al. 2016), nearly five times the average annual rainfall.

The geology and hydrogeology of the Ramotswa area are described in Altchenko et al. (2017). Briefly, the outcrop area of the transboundary Ramotswa aquifer is 453 km^2, spanning both Botswana and South Africa—Fig. S1 of the electronic supplementary material (ESM). The aquifer is considered unconfined to semi-confined and consists of the Ramotswa Dolomite formation. In an aerial electromagnetic (AEM) survey, the Ramotswa maximum aquifer thickness in the Ramotswa town area was at least 340 m and on average 150 m thick (Altchenko et al. 2017). The AEM survey confirmed the complexity of the Ramotswa aquifer's hydrogeology due to karstification, faults, and deep vertical dikes (Altchenko et al. 2017). The mean depth to the water table as measured in the present study was 13.7 m below the ground surface (Table S1 of the ESM), while in the larger Ramotswa aquifer area and over a longer time period it was 24 m (Altchenko et al. 2017). The general groundwater flow

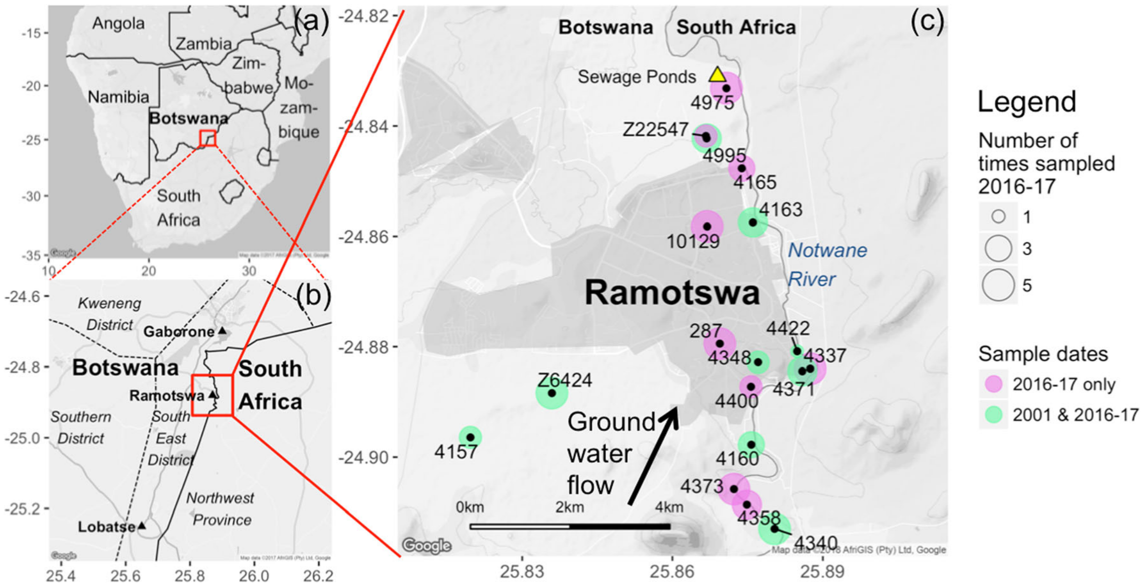

Fig. 2 Maps of **a** Southern Africa, **b** Botswana's South East District, where black triangles represent the three weather stations used in the present study, and **c** Ramotswa borehole locations. Boreholes are labeled and circle size indicates number of times sampled in the present study. Color indicates whether the borehole was sampled in 2001 and this study (green) or only this study (pink). Arrow shows general groundwater flow direction (see also Fig. S1 of the ESM). Yellow triangle is the piped sewage network ponds. Shaded area is the urbanized area

direction is to the northeast (Fig. S1 of the ESM). The Ramotswa aquifer average specific capacity is 2.7 L s^{-1} m^{-1}, average transmissivity is 1,170 m^2 day^{-1}, and the storage coefficient is 5.7×10^{-2} (Staudt 2003; Moehadu 2014). Production borehole yields range from 15 to 150 m^3 h^{-1}, indicating relatively high yields compared to other aquifers in the area. Diffuse recharge is low (<20 mm $year^{-1}$) (Gieske 1992; Post et al. 2012; Altchenko et al. 2017), and active recharge where there is little overburden or soil cover and direct infiltration through dolomite outcrops occurs in the Ramotswa area (Altchenko et al. 2017; Gieske 1992). The latter is critical, as the town of Ramotswa sits on an outcrop of the Ramotswa Dolomite (Fig. S1 of the ESM).

Water supply and treatment infrastructure

The Ramotswa wellfield is about 34 km^2 (Fig. 2) and consists of 14 production boreholes (BHs) managed by the Water Utilities Corporation (WUC), a parastatal organization, and about 45 monitoring BHs, managed by the Botswana Dept. of Water Affairs (DWA). The BHs are not located in a true "wellfield" as they are scattered throughout town and the surrounding area. A series of events explains the current water supply situation in Ramotswa. In the early 1980s production BHs were drilled in Ramotswa to supply drinking water to Gaborone and Ramotswa. When the Gaborone reservoir was completed in 1984, the Ramotswa groundwater was no longer piped to Gaborone and was only used to supply Ramotswa. In 1996, the Ramotswa wellfield was abandoned because NO_3^- concentrations exceeded the national drinking water standard, 50 mg NO_3^- L^{-1} or 11 mg NO_3^-–N L^{-1}(Walmsley and Patel 2011), and because Ramotswa lacked the capacity to treat the water (Moehadu 2014). Water was therefore piped in from Gaborone (Fig. 3). A multi-year drought from 2013 to 2016 severely lowered the Gaborone reservoir level to <5% capacity from Dec 2014 to Mar 2016 (Fig. S2 of the ESM). Below 5% capacity, the reservoir fails to release water. In 2014, the Ramotswa wellfield was reopened as an emergency source of water, and the BHs continued to supply water to Ramotswa and the South East District until Tropical Cyclone Dineo struck Botswana in Feb 2017 and filled the Gaborone reservoir (Fig. S2 of the ESM).

Because NO_3^- concentrations in groundwater from the Ramotswa well field are generally above the drinking water standard (50 mg L^{-1} NO_3^-) and water treatment is unavailable, the drinking water supplied in Ramotswa is a blend of groundwater from the Ramotswa wellfield and surface water supplied by pipes from the Gaborone Dam, supplied by the WUC's Gaborone Water Works (GWW; Moehadu 2014; Fig. 3). The WUC manages all Botswana water supply and sewage. The volumes of dilution water from GWW and produced from the Ramotswa wellfield over time were unavailable. Gaborone relies on supply from the Gaborone reservoir, the North-South carrier, the Molatedi reservoir in South Africa and other sources (Fig. 3). When Gaborone's water supply is low, the water supply to Ramotswa and other towns "downstream" (south) of the GWW is periodically restricted (red "x" in Fig. 3)—defined here as shutting off the water supply for a period of hours or days.

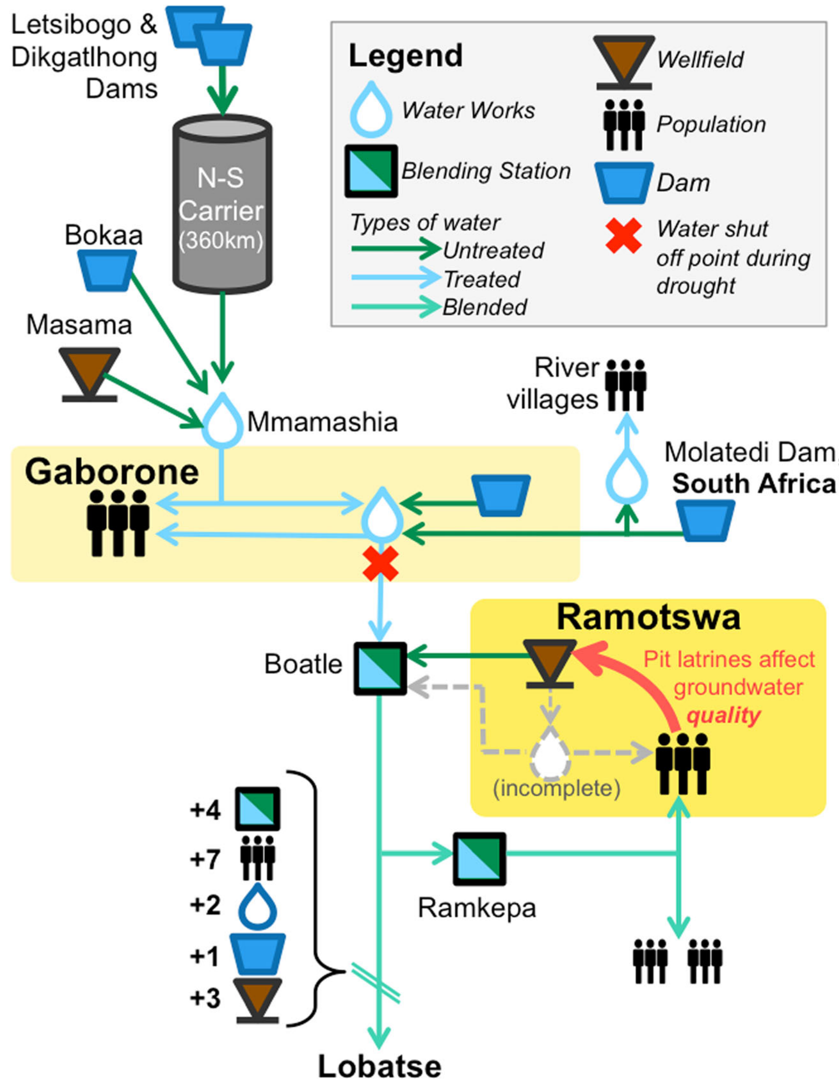

Fig. 3 Water supply scheme for the South East District. The red arrow illustrates the coupling of water quality to water quantity and the grey-dashed arrow indicates how a Ramotswa water treatment facility could de-couple Ramotswa from the Gaborone water supply, which is vulnerable to droughts. Modified from Altchenko et al. (2016). Not to scale.

Human dimensions of water supply and sanitation of Ramotswa town

Botswana's population has rapidly shifted from 9% urban in 1970 to 64% urban in 2011 (Statistics Botswana 2015b). Between 2001 and 2011, the populations of Gaborone and Ramotswa increased by 25 and 50%, respectively (Statistics Botswana 2015c; b), increasing pressure on water supply and sanitation infrastructure (Table 1; Fig. 1). Based on the most recent census in 2011, the population of Ramotswa in 2017 was projected at 37,500 people with a population density of 1,500 persons km^{-2} (Statistics Botswana 2015c). The poverty rate has been declining over time in Botswana, and in 2010 the South East District poverty rate was 13.4% compared to 6.1% in Gaborone (Statistics Botswana 2013).

The annual mortality rate in Botswana for infants is 16 deaths per 1,000 and for children under 5 years it is 27 deaths per 1,000 (Statistics Botswana 2017). Of those deaths, 6.4% of infant deaths and 8% of deaths of children under 5 years are due to "diarrhea and gastroenteritis of presumed infectious origin" (Statistics Botswana 2017), possibly related to contaminated water.

As of 2011, more than one third of Ramotswa households have potable water piped indoors, while about half access potable water using an outdoor pipe (Table 1; Statistics Botswana 2015c). Thirty eight percent of the Ramotswa population had access to a flush toilet as of 2011 (Statistics Botswana 2015c), the majority of which were assumed to be connected to the piped sewage network, while some small fraction used septic tanks (Table 1; Staudt 2003). Fifty six percent of the Ramotswa population relies solely on traditional pit latrines or ventilated improved pit latrines (VIP) (Statistics Botswana 2015c), which translates into an estimated 3,900 pit latrines in the town in 2011. It is unknown how many pit latrines are lined vs. unlined.

Table 1 Selected demographic and sanitation indicators for Botswana and the study area populations for 2011

Parameter		Botswana	Gaborone	Ramotswa	Lobatse
Demography	Population size	2,024,904	*231,592*	30,382	29,689
	Annual pop. growth rate (%) 2001–2011	1.9	2.5	*3.9*	−0.2
	Average household size	NA	3.1	*4.3*	3.1
	Unemployment rate 2011	20	9.2	*26.5*	12.3
Sanitation stats in % of households	Access to potable water piped indoors	NA	*58*	37	42
	Access to potable water piped outdoors	NA	32	*55*	41
	Access to flush toilet	NA	*50*	38	36
	Access to pit latrine	47	3	*56*	10
Source		Statistics Botswana 2015b	Statistics Botswana 2015b	Statistics Botswana 2015a	Statistics Botswana 2015b

Maximum values per row of the three cities/towns are shown in *italics*. *NA* is "not applicable"

Materials and methods

Changes in seasonal rainfall amount and variability

To assess whether the climate in the Ramotswa area has changed over time, daily rainfall data were combined from three weather stations in the broader area (Fig. 2): Ramotswa (24.88°S × 25.87°E, record begins in 1980), Gaborone (20 km north of Ramotswa; 24.70°S × 25.90°E, record begins in 1926), and Lobatse (40 km south of Ramotswa; 25.25°S × 25.65°E, record begins in 1922). These data were provided by the Botswana Dept. of Meteorological Services. Combining the Ramotswa data with the stations in Lobatse and Gaborone allows for analysis of a much longer period (95 versus 37 years) and covers a larger area that includes several reservoirs that supply water to Gaborone (Fig. 3). Due to the strong seasonal pattern in rainfall, wet season (October–March) and dry season (April–September) rainfall totals (referred to hereafter as "seasonal") were analyzed separately.

Multiple linear regression analysis was used to test for trends in five rainfall metrics over time. First, seasonal totals were compared over time to see if annual (water year) volume of rain (mm) is changing. Second, number of days with rain (hereafter "rainy days") were counted per season and compared over time. Third, rainfall intensity per season (mm day^{-1}) was calculated using Eq. (2) from Pryor et al. (2009):

$$\text{Season}_{\text{year}}\ \text{intensity} = \frac{\text{total rain}}{n\ \text{days with rain}} \quad (2)$$

Fourth, climate change can have a greater effect on rainfall extremes and variability than on averages, so variability was compared over time using the coefficient of variation (CV = standard deviation/mean) of monthly rainfall totals per season. Fifth, extreme rainfall seasons were grouped using the upper and lower 10th percentile of seasonal rainfall totals to see if the magnitude of the wettest and driest seasons was changing over time (Pryor et al. 2009; NCEI 2017).

Documenting the impact of climate change on sanitation practices

To document the human behavioral aspects of this study, a small number of key informants were interviewed. The questions were developed to gather general information about sanitation, including undocumented pit latrine details (history, location, types), and how water restrictions affect sanitation practices at the community level. The questions (section S1 of the ESM) were approved prior to the interviews by the Michigan State University Institutional Review Board (IRB# ×16-325e).

The most relevant interview question for testing "H_2: Water shortages affect sanitation behavior, increasing pit latrine use" was somewhat straightforward: "How have water restrictions affected sanitation access in Ramotswa? (access to flush toilets, washing hands, etc.)?" (section S1 of the ESM). Anecdotal belief among researchers and community leaders was that pit latrine use increased during water shortages. Given the time limitations on the research, it was not possible to fully explore the experiences of individual community members using pit latrines. Rather, the focus of this inquiry was to understand the extent of pit latrine use and how community sanitation is impacted by water shortages.

Three informants were chosen because of their access to information, representation, and/or familiarity with a large group of socio-economically diverse people in Ramotswa, and diverse roles in community leadership and resource management in order to expediently capture broad perspectives. Although people in leadership positions can potentially hold an elite bias, it is common practice to use community leaders as key informants for this type of broad, community-level investigation.

The key informants were: (1) a leader in Ramotswa tribal governance, which is independent from state government and prioritizes the well-being of Ramotswa citizens; (2) a WUC representative of the head office in Gaborone, who schedules water shut offs; and (3) a manager at the WUC Ramotswa

Water Works, which is the local office that manages and prioritizes water supply and sewage in Ramotswa. Interviews were conducted in English, audio recorded, transcribed, and coded for thematic analysis.

Groundwater monitoring, water quality sampling, and analyses

Available water quality data from various periods were combined for existing production BHs from 1983 to 2016, a period that has a large gap in measurements between 1999 and 2013, roughly coinciding with the period when the pumps were offline. The only historic water quality data available for monitoring BHs comes from a baseline study conducted in 2001 by Staudt (2003). In addition, data include those collected by Modisha (2017) in August 2016 from a subset of Ramotswa production and monitoring BHs. New data generated in this study were collected from six monitoring and 14 production BHs (Fig. 2) on four occasions in Ramotswa: mid October 2016 (onset of wet season), late November 2016, early January 2017, and early February 2017. The wet season had peaked during January 2017 (Fig. S3 of the ESM). Some data on depth to the groundwater going back to the 1980s are available. However, inconsistency, data gaps, and the effects of production BH pumping prohibit analysis of long-term trends in the depth to water table.

Permission was granted from two farmers to sample their private BHs (BH4157 and BHZ22547), which are equipped with pumps; these data are grouped with the monitoring BHs. At each of the four sampling events sampling from the same 20 BHs was attempted (Fig. 2). Sometimes this was not possible due to inaccessibility or certain production BHs being offline (production BHs are not all running at the same time). Each trip averaged 14–15 BHs. The sampled BHs represent a majority of the BHs available for sampling in Ramotswa; they are also a mix of locations in relation to town (upstream, in town, downstream; Fig. 1) to see if the pit latrines in town had a measureable effect on water quality; and many were also chosen to repeat measurements from 2001 as reported in Staudt (2003). Stratigraphy and casing information from some BH logs are available in Staudt (2003) and summarized in Table S1 of the ESM.

Samples from the monitoring BHs were collected using a Grundfos submersible centrifugal pump with an unregulated flow rate of 0.7–1.5 m^3 h^{-1}, which was the same as used by Modisha (2017) and Staudt (2003) (Grundfos models MP-1 and SQ 1.2-3 N, Bjerringbro, Denmark). Samples from the production BHs were obtained from a valve on the in situ pumps.

The following variables were measured continuously with a calibrated Quanta Water Quality Sensor (Hach, Loveland, Colorado, USA) while sampling: temperature, pH, dissolved oxygen (DO), and oxidation reduction potential (ORP). The outlet of the pump tubing and the sensor were placed at the bottom of a 15-L plastic bucket that was allowed to continuously overflow. This ensured the sensor was measuring water fresh from the pump (presumably equal to in situ water quality) and the water at the top of the bucket acted as a barrier to atmospheric gas exchange. BHs were pumped until physical and chemical variables stabilized (USGS 2006), typically after 30–60 min of pumping, at which time the groundwater samples were collected.

An unfiltered 500-ml sample was collected into a sterilized glass bottle for fecal coliform analysis, a standard indicator of fecal contamination from warm-blooded animals, including humans and livestock. Samples were stored on ice in a cooler until analyzed within 24 h at the WUC Mmamashia Water Treatment Works microbiology laboratory following the ISO standardized membrane filtration method (ISO 9308-1).

A second, 350-ml sample was filtered through a new Supor 0.45-μm membrane filter (Pall Corporation, Ann Arbor, MI, USA) and stored in plastic bottles. These samples were refrigerated and delivered to Waterlab, a private service laboratory in Pretoria, South Africa for the following hydrochemical analyses that were conducted within ten days: NO_3^- ($NO_2^- + NO_3^-$) was measured colorimetrically using hydrazine reduction and mercuric thiocyanate methods, respectively, on an Aquakem 250 spectrophotometer (Thermo Scientific, Waltham, MA, USA) (USEPA 1993); DOC was measured by UV oxidation to CO_2, which was measured using an infrared analyzer (Sievers 900 Analyzer, GE Analytical Instruments, Boulder, CO, USA). Ammonia (NH_3) and ammonium (NH_4^+) were also measured but concentrations were <0.1 and < 0.6 mg L^{-1}, respectively, at all sites and dates. Several additional analytes (section S2 of the ESM) were also measured on the same samples and results are reported in section S3 of the ESM.

Caffeine and paraxanthine were measured in groundwater samples as indicators of human waste contamination. Caffeinated sodas, instant coffee, and black teas are popular beverages in Ramotswa (Fig. S4 of the ESM). The half-lives of caffeine and paraxanthine were estimated in a US estuary to be 3 to >100 days and 11 to >100 days, respectively (Benotti and Brownawell 2009). In a German karst aquifer system, the caffeine half-life was estimated at 89 days (Hillebrand et al. 2015). This high range of values indicates the need for site-specific investigations under the prevailing local conditions. Sample collection was conducted following Matongo et al. (2015). During the November 2016 and January 2017 trips only, 500-ml samples for caffeine and paraxanthine analysis were filtered in the same way as the hydrochemistry samples. Samples were stored in a refrigerator before extraction within one week. Samples were collected and extracted by someone who had not consumed caffeine in the previous 5 days, and stored in coolers and refrigerators not used for food and beverages.

Solid phase extraction for caffeine and paraxanthine followed the method in Matongo et al. (2015). Caffeine standard ($C_8H_{10}N_4O_2$, CAS No. 58-08-2) and paraxanthine standard ($C_7H_8N_4O_2$, CAS 611-59-6) were purchased from

Sigma-Aldrich (St. Louis, MO, USA). A 10-mg L^{-1} stock standard was made by dissolving 10 mg caffeine and 10 mg paraxanthine in 1 L of 50:50 mix of methanol and deionized water. Extracted samples were analyzed 16 weeks after extraction by GC-MS on an Agilent 6890 N gas chromatograph with a 5973 inert mass selective detector (Agilent Technologies, Santa Clara, CA, USA) in select ion monitoring mode utilizing a 15-m Rtx-35 ms capillary column (Restek Corporation, Bellefonte, PA, USA). The oven program was 70 °C for 1 min, a 20 °C min^{-1} ramp to 190 °C, a 15 °C min^{-1} ramp to 210 °C, and a final ramp of 30 °C min^{-1} to 300 °C. The quantifying ions were m/z 194 and 180 for caffeine and paraxanthine, respectively. Three micrometre injections gave a limit of quantitation for caffeine of 0.5 μg L^{-1} and for paraxanthine of 10 μg L^{-1}.

Evidence of denitrification, i.e., the production of N_2O, in the aquifer was sought to determine its potential for ISB (USEPA 2013). Dissolved gas samples were collected from the groundwater for N_2O analysis. Because gas samples were transported to the USA for analysis, they were only collected during the final sampling trip in February 2017 to minimize storage time between collection and analysis. Previous unpublished work has shown that N_2O concentration in gas samples collected this way are stable for 90 days when stored in the dark at room temperature (K. Kahmark, Michigan State University Kellogg Biological Station, personal communication, 2016).

The dissolved gas sample collection method described in Hamilton and Ostrom (2007) was used here with a few modifications. A 30-ml water sample and a 30-ml ambient air sample were drawn into one gas tight syringe, shaken for 5 min to achieve equilibration, and 10 ml of headspace gas was injected to over-pressurize a 5.92-ml glass vial with a rubber septum (Labco Ltd., High Wycombe, UK). To ensure the sampled water was not exposed to the atmosphere, it was drawn through a narrow piece of polypropylene tubing extending from the syringe tip and inserted directly inside the end of the pump tubing or far below the water surface in the bucket. Samples were collected in triplicate at each site along with a fourth sample of ambient air. Gas samples were stored in the dark at room temperature and were analyzed within 30 days on a gas chromatograph (Agilent 7890, Agilent Technologies, Santa Clara, CA, USA). N_2O was analyzed with a ^{63}Ni electron capture detector at 350 °C coupled to a Gerstel MPS2XL automated headspace sampler (Gerstel, Mülheim an der Ruhr, Germany). The system had a two-column back-flush setup using Restek PP-Q 1/8″OD, 2.0-mm ID, 80/100 mesh, 3-m-packed columns (Restek, Bellefonte, PA, USA). The oven was set to 90 °C.

Calculations of N_2O concentrations in groundwater followed those described in Hamilton and Ostrom (2007) and are the result of several steps. Briefly, the concentration of N_2O dissolved in the original liquid sample is back-calculated using the calculated Bunsen solubility coefficient, Henry's Law, and the ideal gas law (Weiss 1974; Weiss and Price 1980). The ambient N_2O concentration of the air drawn into the syringe for headspace equilibration was subtracted from the final headspace concentration. In addition, the amount of N_2O dissolved in the water when it infiltrated the soil was predicted assuming it was in equilibrium with the atmosphere, and this was also subtracted from the measured N_2O concentration.Therefore, reported N_2O concentrations are the amount of N_2O that the water accumulated below ground in excess of what it contained from air equilibration in the unsaturated zone.

Multiple linear regression analyses of rainfall and water quality data were conducted in R 3.3.2 (R Core Team 2017), and coefficients and models are reported here with *adjusted* R^2 values. Plots and maps were created using the R packages ggplot2 (Wickham 2009) and ggmap (Kahle and Wickham 2013).

Results

Changes in seasonal rainfall and variability between 1922 and 2017 in Ramotswa, Gaborone, and Lobatse

Seasonal total rainfall has decreased significantly in both the wet and dry seasons since the 1920s (Gaborone and Lobatse) and 1980 (Ramotswa) (Fig. 4a, model $R^2 = 73\%$, $p < 0.001$, Table 2: model 1). The number of rainy days has decreased significantly in both seasons as well, but more so in the wet season (Fig. 4b, model $R^2 = 81\%$, $p < 0.001$, Table 2: model 2). Daily rainfall intensity (Eq. 2) has increased significantly in both the wet and dry seasons across all stations (Fig. 4c, model $R^2 = 35\%$, $p < 0.001$, Table 2: model 3). Because total rainfall has *decreased*, the increase in intensity is driven by the decline in number of rainy days in both seasons. Rainfall variability (Fig. 4d) measured as the seasonal CV among monthly rainfall totals, has increased significantly in both seasons (model $R^2 = 64\%$, $p < 0.001$, Table 2: model 4). The CV increased more quickly in the dry season.

Rainfall extremes have also risen. Organizing the data by percentiles (Fig. 4e) illustrates how the wettest dry seasons (upper 10th percentile) are getting significantly drier over time (model $R^2 = 90\%$, $p < 0.001$, Table 2: model 5). Also, the wettest wet-seasons have become significantly wetter over time. The driest dry seasons (lower 10th percentile) and the majority of wet and dry seasons (middle 80% of observations) had no significant trends. The wet-season lower 10th percentile showed a drying trend but is not statistically significant.

In summary, rainfall metrics show that the climate in the Ramotswa area has become more extreme, with greater rainfall variability and intensity in both seasons. While both seasons have grown drier over time, the results indicate an increasing probability of both droughts and floods.

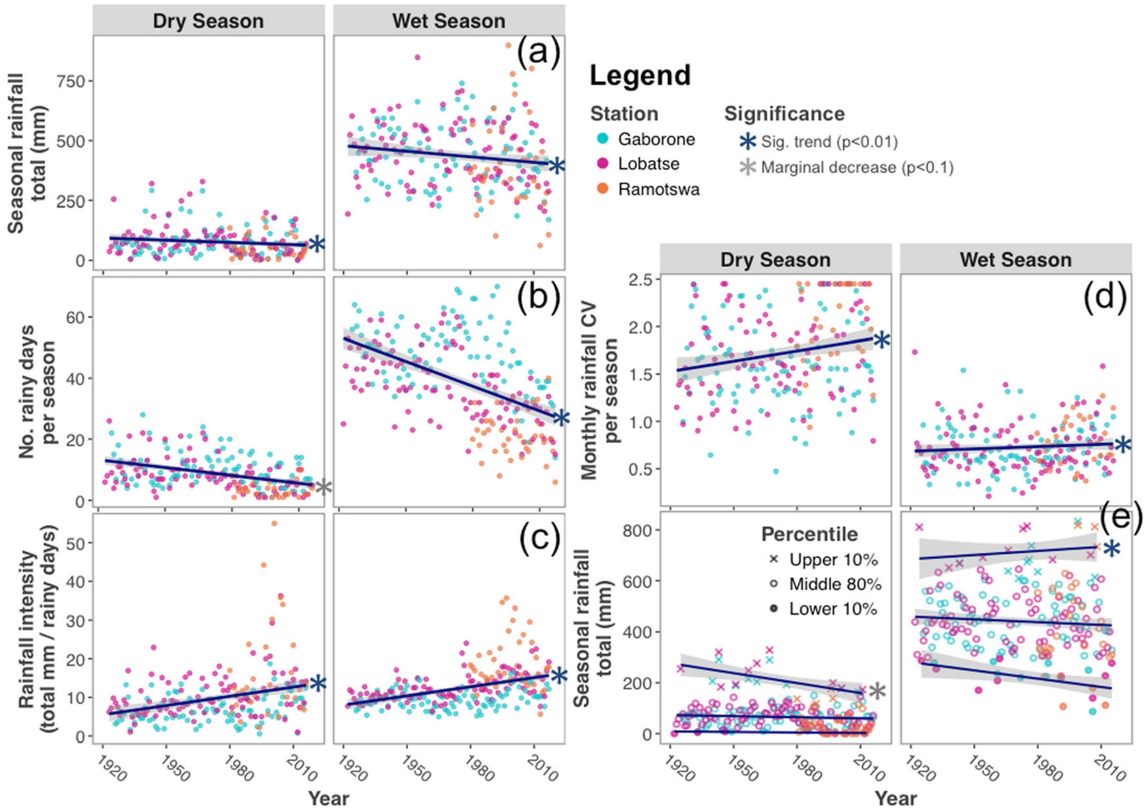

Fig. 4 Dry- and wet-season rainfall patterns over time at Gaborone, Lobatse, and Ramotswa weather stations (color). Significance is indicated with an asterisk. **a** Total rainfall, **b** number of rainy days, **c** rainfall intensity, **d** seasonal coefficient of variation (CV) among monthly rainfall totals, **e** extreme rainfall events including upper and lower 10th percentiles of seasonal rainfall totals, shape). See Table 2 for model results

Interviews on water restrictions and sanitation behavior

The interviews confirmed that pit latrines are commonplace throughout Ramotswa and have been the most common sanitation method in Ramotswa since at least the 1950s. In addition, many of the households with a flush toilet (~38% Ramotswa population, Table 1) also have a new or remnant, functional pit latrine, which they use when the water supply is restricted. Pit latrine depth varies depending on soil depth, but typically they are thought to be about 1–1.5 m deep. Pit latrines closer to the Notwane River are known to flood as the water table rises with heavy rains. The WUC reports that new and existing Ramotswa households continue to install flush toilets and connect into the sewage network to the present day.

Many older homes have more than one pit latrine, some of which are abandoned and/or full. When pit latrines fill, a resident either pays the WUC US$50 to pump the waste out of the pit latrine and truck it to the sewage ponds, or the resident digs a new pit latrine and leaves the old one unused. It was unclear what proportion of households pay to pump out a pit latrine or how frequently this occurs.

Sources explained that during the 2013–2016 drought period, when the reservoir capacity was at <5% (corresponding to failure capacity, Fig. S2 of the ESM), the water supply was shut off for 1–3 days at a time per week without notice. It is common practice during water restrictions for those people who have flush toilets to use pit latrines. This was also confirmed in a 2016 survey of 100 households in Ramotswa, which also found that people did not typically store water for flushing their toilet during water restrictions, usually because they did not have a storage tank and the cost of a tank was prohibitive (T. Matsoga, University of Botswana, personal communication, 2017). Indeed, the Ramotswa WUC representative reported a drastic drop in flow to the sewage system during the 2014–2015 period:

> "*[Water restrictions] do affect people because without those flush toilets … our sewage system is not working. Because of these water restrictions, it's only receiving very little water, which cannot keep it running. It's very visible at our [treatment] ponds…There won't be any flow from one pond to another.*"

Similarly, a source recalled that during the water restrictions, they would "*go for about three days without water. And*

Table 2 Historic rainfall regressions

Model	Estimate	Standard error	p	Adjusted R^2	Model p
1. Seasonal rainfall total = year + season (reference level is dry season)				73%	<0.001
(Intercept)	76.572	7.511	<0.001[b]		
Year	−0.552	0.193	<0.01[b]		
Wet season	359.565	10.469	<0.001[b]		
2. Number rainy days = (year × season) + station (reference levels are dry season and Gaborone station)				81%	<0.001
(Intercept)	13.431	0.720	<0.001[b]		
Year	−0.040	0.021	0.062[b]		
Wet season	30.451	0.779	<0.001[b]		
Lobatse station	−6.580	0.848	<0.001[b]		
Ramotswa station	−13.600	1.221	<0.001[b]		
Year × wet season	−0.180	0.029	<0.001[b]		
3. Intensity[a] = year + season + station (reference levels are dry season and Gaborone station)				35%	<0.001
(Intercept)	2.700	0.061	<0.001[b]		
Year	0.007	0.001	<0.001[b]		
Wet season	0.435	0.066	<0.001[b]		
Lobatse station	0.519	0.072	<0.001[b]		
Ramotswa station	1.023	0.103	<0.001[b]		
4. CV = year × season (reference level is dry season)				64%	<0.001
(Intercept)	1.724	0.260	<0.001[b]		
Year	0.004	0.001	<0.001[b]		
Wet season	−0.995	0.037	<0.001[b]		
Year × wet season	−0.003	0.001	<0.05[b]		
5. Seasonal total mm = year × percentile group × season (reference levels are middle 80% and dry season)				90%	<0.001
(intercept)	65.162	5.214	<0.001[b]		
Year	−0.145	0.194	0.456		
Upper 10th percentile	142.046	15.237	<0.001[b]		
Lower 10th percentile	−60.271	15.136	<0.001[b]		
Wet season	375.420	7.450	<0.001[b]		
Year × upper	−1.160	0.607	0.057[b]		
Year × lower	0.080	0.510	0.875		
Year × middle × wet	−0.218	0.278	0.432		
Wet season, upper 10th perc.	130.839	21.970	<0.001[b]		
Wet season, lower 10th perc.	−157.296	22.076	<0.001[b]		
Year × upper × wet	2.056	0.892	<0.05[b]		
Year × lower × wet	−0.804	0.764	0.294		

For all models, year is centered on the mean year

[a] $(\text{Intensity} + 0.5)^{1/2}$

[b] Indicates statistical significance

definitely then you would need to use the pit latrines." The interviews and household survey cited above suggest that water restrictions increased pit latrine usage in Ramotswa. This view was confirmed informally in conversations with people in other water management positions in Botswana, residents in Ramotswa, and people living in other communities in Botswana affected by water restrictions.

Nitrate concentrations and in situ denitrification

High groundwater NO_3^- concentrations remain a problem in Ramotswa. The highest NO_3^- concentration measured in the present study, 29 mg NO_3^-–N L^{-1}, was recorded at two sites: production BH4422 in town in October 2016 and private BH4157 at the cattle *kraal* (pen) in November 2016 (Fig. S5

of the ESM). BH4422 was unavailable for sampling after October 2016. Monitoring BH4995 ranged from 23 to 27 mg NO_3^-–N L^{-1} for four measurements taken between October 2016 and February 2017. These three BHs (4422, 4157, and 4995) also had three of the highest NO_3^-–N concentrations in Staudt (2003) (Fig. 5). Even though BH4422 and BH4995 showed a decline since 2001 (both around 40 mg NO_3^-–N L^{-1} then and near 30 mg NO_3^-–N L^{-1} in 2017), they are still well above the NO_3^- water quality standard. Given variability in NO_3^- concentrations and the lack of long-term data (Fig. S5 of the ESM), it is unclear if the measurements reported here indicate a long-term declining trend. None of the BHs showed a statistically significant trend in NO_3^- concentrations over time between August 2016 and February 2017.

The molar ratio of NO_3^-–N to DOC tended to be >1 and as high as 44 in the case of BH 4995, suggesting DOC is potentially limiting denitrification at certain BHs (Fig. S6 of the ESM), which indicates the feasibility of ISB. The production BHs that exceeded the NO_3^-–N drinking water standard, BH4422 and BH4400, also showed C-limitation.

N_2O concentrations were orders of magnitude greater than atmospheric equilibrium concentration (most samples had >1 μg N_2O–N L^{-1} compared to the atmospheric concentration of 0.15 μg N_2O–N L^{-1}), indicating high denitrification activity. N_2O concentrations were positively related to nitrate concentration (Fig. 6). A linear regression of log(N_2O concentration + 0.5) by NO_3^- concentration was highly significant ($p < 0.0001$) and explains much of the variability in N_2O ($R^2 = 70\%$, Table S2 of the ESM; Fig. 6). BH4160 had 47.75 μg N_2O–N L^{-1} (the maximum by far). Measurements with the highest NO_3^- and N_2O were all C-limited (Fig. 6), suggesting the potential for ISB.

Nitrate source tracking

Fecal coliforms were not detected in any of the *production* BHs but were detected in several monitoring BHs (Fig. 7). The maximum detection limit of 200 colony forming units (CFU) per 100 ml was observed at least once in BH4371, BH4995, and BH10129, indicating that actual concentrations could be higher. The Botswana drinking water standard is zero CFU per 100 ml (Walmsley and Patel 2011). The presence of fecal coliforms suggests that other fecal pathogens are likely to be present in the aquifer. High NO_3^-–N concentrations did not always coincide with the presence of fecal coliforms, but the presence of fecal coliforms tended to coincide with high NO_3^-–N concentrations (Fig. S7 of the ESM). Regression analysis of samples with fecal coliforms by NO_3^- was significant ($p < 0.05$, Table S2 of the ESM) but only weakly explains the variation ($R^2 = 11\%$).

Caffeine and paraxanthine were present in several BHs at both sampling times (November 2016 and January 2017; Fig. 7b,c). The detection (and nondetection) of paraxanthine at many of the same BHs as caffeine confirms the low likelihood of false positives during sampling and analysis. Both compounds were measured in more wells and at slightly higher concentrations in November compared to January. The few BHs where the compounds were detected in January were BHs where they were also detected in November (Fig. 7b,c). Where detected, caffeine concentrations ranged from 14 to 56 ng L^{-1}, and paraxanthine concentrations ranged from 180 to 770 ng L^{-1}. Caffeine and paraxanthine were found in several monitoring BHs as well as production BHs, including production BH4340, BH4358, and BH4373, which are upstream of town. To test whether caffeine was tracing the NO_3^- pollution, a regression was used to predict caffeine with an interaction between the NO_3^- concentration and C-limitation status

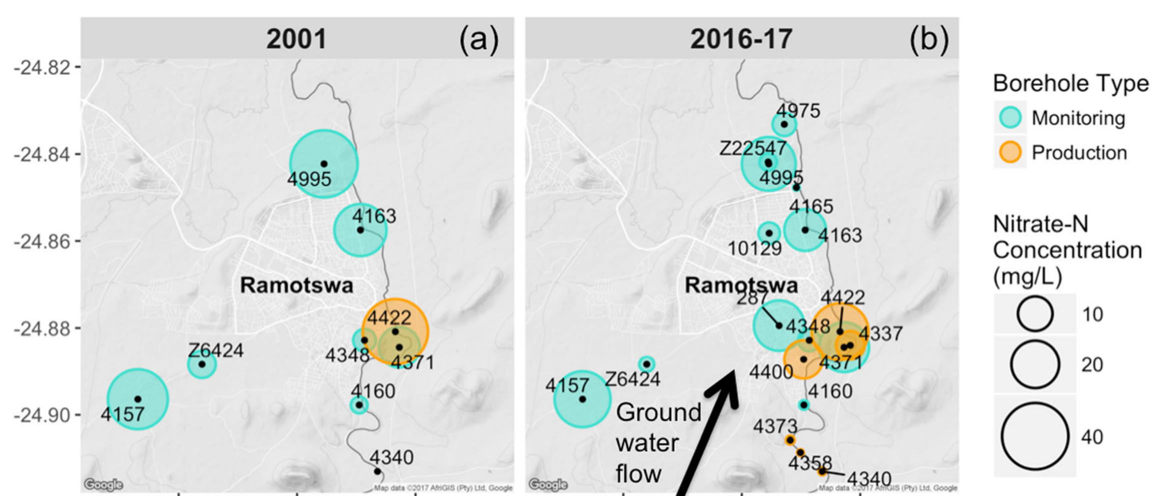

Fig. 5 Changes in NO_3^-–N concentrations in Ramotswa BHs over time and space. Circle size indicates BH NO_3^-–N concentrations. **a** Concentrations in 2001 (Staudt 2003) compared to **b** mean concentrations measured in this study

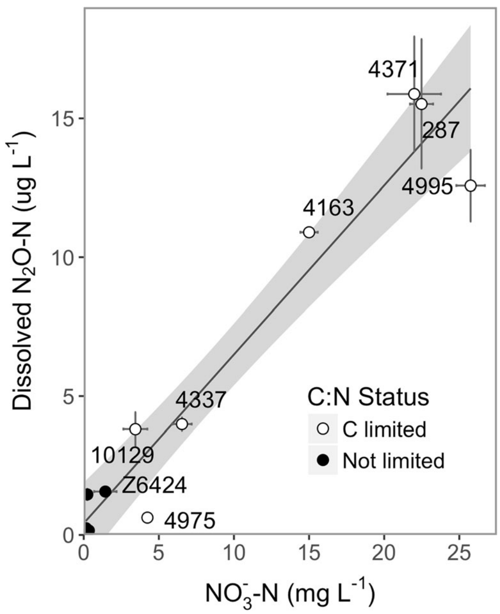

Fig. 6 Dissolved N_2O was significantly and positively related to nitrate concentration. Error bars represent N_2O standard error among three replicate measurements on one sample date. Shaded area is the 95% confidence interval for regression analysis ($p < 0.0001$ and adjusted $R^2 = 0.70$, Table S2 of the ESM). BH 4422 and BH 4400 were inaccessible in February 2017 (only time N_2O samples were collected). Many BHs were C-limited over the four sampling trips (Fig. S6 of the ESM). BH 4160 N_2O concentration was 47.75 μg N_2O L^{-1} (not shown) and was not C-limited

(whether the molar ratio of NO_3^-–N to DOC was >1 or < 1). The regression was significant ($p < 0.01$) and explains 67% of the variability (Table S2 and Fig. S8 both of the ESM).

Discussion

Effect of climate change on water supply in Ramotswa

Declining rainfall and increasing variability

The analyses of the last 90 years of rainfall data strongly suggest that the South East District's climate is changing (Fig. 4). Declines in seasonal total rainfall and number of days with rain, together with increasing variability and extremes, are likely to increase the frequency and intensity of droughts and floods such as the most recent 2013–2016 drought. This supports H_1: Climate change-induced droughts are becoming more frequent (Fig. 1), which is in agreement with other studies of rainfall amounts in southern Africa (Niang et al. 2014; Hodnebrog et al. 2016). The climate models used in the IPCC Fifth Assessment Report project Botswana and Namibia will "very likely" (i.e., >90% probability) continue to see declines in mean annual rainfall, a delay in the onset of the wet season, and by 2100, drier and shorter wet seasons and drier dry seasons (Niang et al. 2014). A delay in the onset of the wet season was not detected in the rainfall data used here.

Changes in the variability and intensity of rainfall may have more significant influences on drought and flood frequency. The South East District's rainfall data showed a decline in the number of days with rain and increases in seasonal rainfall intensity, in agreement with New et al. (2006).

Climate change impacts on Ramotswa drinking water supply

According to the aforementioned climate results and other studies on Botswana's rainfall trends (Kenabatho et al. 2012; Byakatonda et al. 2018), the Gaborone reservoir is likely to reach failure capacity more frequently in the future (Fig. 4). As the temperature is likely to increase 3.4–4.2 °C by 2100 in southern Africa (Niang et al. 2014), evaporation from reservoirs, already >2,000 mm $year^{-1}$ (Altchenko et al. 2016), will likely increase, reducing water security of reservoir storage. For people who rely on surface water for their entire water supply or for diluting a contaminated groundwater supply these changes in the climate threaten their water security.

Effect of drought and water restrictions on sanitation in Ramotswa

The key informant interviews strongly supported H_2: Water shortages affect sanitation behavior, increasing pit latrine use (Fig. 1). Changes in the water system limited many people's sanitation options, and changed their sanitation behavior. With an increasing likelihood of events like the recent 3-year drought and no change in water supply and associated treatment infrastructure, Ramotswa will experience more frequent disruptions in water supply, and as a consequence there will be more use of pit latrines (Fig. 1). Based on the most recent population census, water restrictions potentially increase the number of people using pit latrines by about 14,250 (Statistics Botswana 2015c).

Groundwater quality and in situ denitrification bioremediation potential

The Ramotswa groundwater continues to exhibit NO_3^- concentrations in excess of the Botswana drinking water standard of 50 mg NO_3^- L^{-1}. This includes two of the production BHs, 4422 and 4400, and several monitoring BHs (Fig. 5). The concentrations on the whole do not appear to have increased or decreased significantly since the Staudt (2003) observations, despite Ramotswa's population increasing by 50% between 2001 and 2011 (Statistics Botswana 2015c). There may be reasons why population growth and NO_3^- concentration

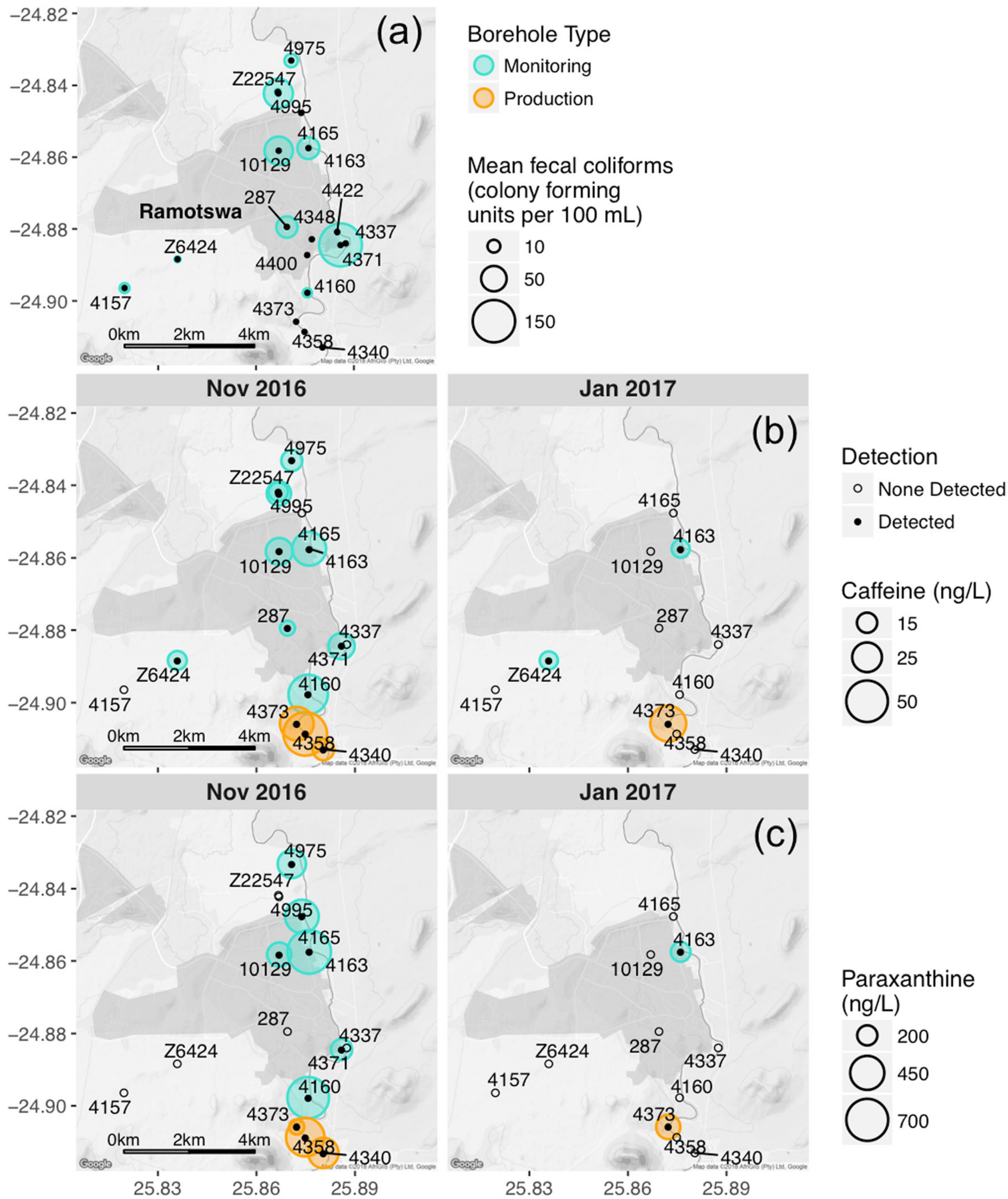

Fig. 7 Maps of fecal contamination indicators. **a** Mean fecal coliform count, an indicator of warm-blooded animal fecal contamination, from a maximum of four measurements between the October 2016 and February 2017. No fecal coliforms were detected at any of the production BHs. **b** Caffeine and **c** paraxanthine concentrations, which serve as indicators of human waste contamination, measured in November 2016 (left panel) and January 2017 (right panel)

are not directly related. Since approximately 2000, the newly expanding peri-urban areas of Ramotswa are connected to the sewage network and most households are believed to be connected (source: interviews), though the recent drought incentivized building pit latrines at new homes. Older households located near the sewage network continue to connect. Also, a greater proportion of new pit latrines, compared to old pit latrines, may be lined, preventing additional nitrate contamination. Given the data available, it seems that the sewage network could be reducing the volume of human waste entering pit latrines and potentially affecting the aquifer. However, with increasing likelihoods of long-term droughts, water shortages, and water restrictions, *people with flush toilets may be forced to use pit latrines*, jeopardizing the preventive effect of the sewage network on groundwater pollution.

High N_2O supersaturation in the groundwater suggests denitrification is occurring in the aquifer (Fig. 6), while low DOC concentrations relative to NO_3^- concentrations indicate that denitrification is C-limited (Fig. S6 of the ESM), supporting H_4: Enhanced denitrification via ISB of the Ramotswa aquifer has the potential to reduce nitrate contamination. Jacks et al. (1999) also demonstrated denitrification occurrence in Ramotswa groundwater by measuring $^{15}N/^{14}N$ ratios in the groundwater and non N-fixing tree leaves.

Sources of nitrate contamination

N fertilizer, organic matter, livestock waste, and human waste are potential sources of NO_3^- contamination; however, N fertilizer is ruled out as a significant contributor to NO_3^- contamination for several reasons. First, two of the production BHs south of town (4358 and 4340) are located in cultivated farm fields, yet their mean concentrations were consistently <1 mg NO_3^-–N L^{-1} and N_2O was also low. Second, samples with fecal coliform contamination also tended to be consistently high in NO_3^- (Fig. S5 of the ESM), suggesting a livestock and/or human source for both. Third, synthetic fertilizer only became available to farmers in the area in the last few years as a subsidy from the government, nevertheless NO_3^- has been high in the Ramotswa aquifer since the 1980s (Staudt 2003).

As for livestock waste, there are no feedlots in Ramotswa where livestock waste is concentrated. Livestock waste on the soil surface is commonplace throughout Ramotswa as animals freely graze around town. Fences protect production BHs but not monitoring BHs. Recharge mechanisms are complex. Recharge can be <1 mm $year^{-1}$ (Post et al. 2012) when it occurs by soil moisture infiltration but it can be rapid, up to 20 mm $year^{-1}$, in localized areas of runoff percolation, ephemeral riverbed infiltration, and preferential flow through karstified outcrops such as the outcrop the town sits on (Gieske 1992). As a consequence, the likelihood of contamination from dispersed livestock waste infiltrating from the surface through soil moisture is low compared to the likelihood of contamination from pit latrines. Livestock concentration in areas like Ramotswa with surface karstification might present more significant livestock contamination risk. With only one BH outside of town used for watering cattle (BH4157), statistical conclusions about the direct impact of livestock are not possible (at this BH, NO_3^- mean concentration was 15 mg NO_3^-–N L^{-1}, fecal coliforms ranged from none detected to 5 FCU, and caffeine and paraxanthine were not detected). Thus, livestock as a source of some of the NO_3^- cannot be completely ruled out.

Several lines of evidence suggest a predominantly human source, supporting H_3**:** Nitrate pollution is primarily originating from human waste contamination. Fecal coliform concentrations were highest in town (but also at BH4157) and consistently low upstream. However, some samples had high NO_3^- and no fecal coliforms indicating that other sources of NO_3^- may exist or that fecal coliforms are not a conservative or reliable tracer of NO_3^- as in Peeler et al. (2006) or human waste as in Glassmeyer et al. (2005). The strongest line of evidence that NO_3^- is from a predominantly human source is that caffeine and paraxanthine were present in many BHs in the town (Fig. 7) and caffeine concentrations correlated significantly with NO_3 concentrations (Fig. S8 of the ESM).

However, this does not identify the exact source of human waste. The thousands of pit latrines in Ramotswa are the most likely sources given the locations and co-occurrence of the aforementioned indicators. A study in Mochudi, a town 55 km north of Ramotswa and also located on the Notwane River, found pit latrines were the source of NO_3^- pollution of the groundwater (Lagerstedt et al. 1994). It is possible that some human waste comes from other sources like improperly maintained or sited septic tanks, ruptures in the sewage network, and/or leaking settling ponds. The sewage settling ponds north (downstream) of town are unlikely sources of contamination, but septic systems and sewage networks have been shown to contaminate groundwater with N and other pollutants (Katz et al. 2011).

The detection of caffeine in the southernmost BHs (4373, 4358, and 4340) is perplexing. These BHs had <0.5 mg NO_3^-–N (Fig. 5) at all sample dates and <1.5 μg N_2O–N L^{-1}, suggesting that there was either no NO_3^- contamination or NO_3^- had been fully denitrified to N_2, or N_2O gas had escaped from the aquifer. In addition, the presence of caffeine at these southern (upstream) locations could mean the caffeine is not coming from the pit latrines. Below are a couple speculative explanations for this peculiarity.

First, these southernmost production BHs would have a large cone of depression when they are pumping, so that their chemistry represents a wider area that potentially includes recharge zones affected by human waste. Along this flowpath, caffeine could remain in the water, but NO_3^- is completely denitrified to N_2 so that neither NO_3^- nor N_2O are present. The caffeine could persist, even along C-limited flow paths (Fig. S8 of the ESM), because denitrifying bacteria might not use caffeine as a source of DOC. Indeed, caffeine is considered toxic to many bacteria (Mazzafera 2004). Also, the conditions that enable microbial caffeine degradation are different from the conditions necessary for microbial NO_3^- removal: denitrification requires low oxygen conditions, whereas microbial degradation of caffeine seems to require aerobic conditions (Mazzafera 2004). Therefore, samples collected downstream from a common source of NO_3^- and caffeine could exhibit caffeine without NO_3^- or vice versa, depending on the microbial community and oxygen availability.

Additionally, caffeine in these southern production BHs could be from the Notwane River, which may carry human and livestock waste contamination when it is

flowing. The river likely loses water to groundwater recharge and, anecdotally, BHs close to the river can be submerged in flood water, potentially affecting the chemistry of these southernmost BHs located in the floodplain. Unfortunately, Notwane River chemistry data at Ramotswa are not available to test this idea. But a joint water quality report by the Departments of Water Affairs (DWA) in Botswana and South Africa (2013) found that NO_3^-–N was 8 mg L^{-1} after the Gaborone waste water discharges into the Notwane River (downstream of Ramotswa and the Gaborone Reservoir), which is low compared to >20 mg L^{-1} in several Ramotswa BHs reported here. Upstream of Ramotswa is the town of Lobatse, which has a similar population size as Ramotswa, confined animal feeding operations, and a slaughterhouse. Its wastewater effluent feeds into the Notwane Dam, 33 km upstream of Ramotswa. There are no other large settlements on the Notwane River between Lobatse and Ramotswa. Perhaps NO_3^- but not caffeine from Lobatse wastewater is removed by the time the river recharges the aquifer in Ramotswa. If the river is the source of caffeine for the southernmost BHs, then it does not explain the presence of caffeine in upland BHs like 287, 10129, or 4995—meaning, pit latrines and not the river likely contribute caffeine to the groundwater at least in these upland BHs.

Greater understanding is needed of the role of the Notwane River in groundwater recharge and quality in Ramotswa to assess its potential as a contamination source. However, the co-occurrence of NO_3^-, fecal coliforms, and caffeine in the sampled BHs (except for the southernmost BHs) strongly suggests human sources of contamination *within* Ramotswa, which are most likely predominantly from the thousands of pit latrines (Table 1).

Indirect impact of climate change on groundwater quality

It was shown that human-induced changes in the climate, sanitation behavior, and groundwater interact to put Ramotswa's water security at risk (Fig. 1). Water insecurity in the district is likely to increase due to changes in environmental and social conditions. Regardless of climate change, social pressures on water supply will likely perpetuate water scarcity in the South East District (Taylor et al. 2009; Calow et al. 2010; Niang et al. 2014), and undoubtedly exacerbate, and be exacerbated by, water quality threats. Ramotswa's dependence on Gaborone for diluting its groundwater means Ramotswa's access to safe drinking water (and water to flush toilets) is paradoxically threatened by the combination of reduced surface-water quantity (via climate change) and poor groundwater quality, i.e., a water *quantity* problem intensifies a water *quality* problem.

Recommendations

A three-pronged approach is recommended: reduce contamination, in situ remediation of the groundwater, and local treatment of Ramotswa groundwater. Individually, none of these approaches provides an immediate or long-term solution, but together they could turn Ramotswa groundwater management into a success story.

Reduce contamination. If Ramotswa's high annual population growth rate from 2001 to 2011 of 3.9% continues (Statistics Botswana 2015c), Ramotswa will see an increase in both water demand and volume of human waste over time. Water and sanitation infrastructure should be optimized to ensure safe and reliable sanitation access, match current and future water availability, and protect groundwater quality. Lining pit latrines should be a priority. When an unlined pit latrine is pumped out, a liner could be installed. Incentives could be designed to increase the number of pit latrines that get pumped rather than abandoning full pit latrines and building new ones. The climate analyses here suggest flush toilets will be less and less usable with increasing frequency of droughts and water restrictions. Captured and stored rain or grey water could be used to flush toilets, but Ramotswa residents need assistance in order to obtain the necessary equipment. Planning for improvements in infrastructure (e.g., expanding the sewage system) requires information about the current system including well production and water use. Useful spatial data that are missing include: up to date maps of the piped sewage network, homes connected, pit latrines and their status (including lined/unlined, in-use/abandoned, etc.), and septic tanks.

Remediate. The presence of N_2O dissolved in the groundwater confirmed that the microbes and conditions in the aquifer are suitable for denitrification. The limited availability of DOC relative to NO_3^- in Ramotswa groundwater demonstrated that ISB with supplemental DOC has the potential to enhance removal of NO_3^- via denitrification (Fig. 6). A vegetable oil amendment could act as a carbon substrate (USEPA 2013), and this has been proposed for ISB in Ramotswa. ISB has the potential to be cost-effective and less energy intensive than traditional water treatment such as reverse osmosis. However, many factors must be taken into consideration to test the feasibility of ISB in Ramotswa, including the cost (relative to the funds needed to build and run a water treatment facility), sustainability, dolomite suitability for ISB (Tompkins et al. 2001), site specific hydraulic characteristics, amendment longevity, performance monitoring using tools such as stable isotopes, and effects on downstream geochemistry (USEPA 2013; Majone et al. 2015). Both contamination prevention and remediation are important approaches to the mitigation of drinking water contamination.

Treat. A water treatment facility for Ramotswa's groundwater using reverse osmosis has been planned in Boatle (grey arrows in Fig. 3) but progress has stalled for over a year. If Ramotswa were able to treat the groundwater and use it for 100% of its supply, then greater extractions will be required, jeopardizing a sustainable pumping rate. Ramotswa's water security is also tied to demand and failures in infrastructure in other parts of the South East District water delivery scheme (Fig. 3). Therefore, it would be helpful to compile past and ongoing water abstraction volumes and conjunctive use of surface and groundwater *across the water supply scheme* in real time in one database to manage both supply and use locally and across the South East District.

To understand how NO_3^- concentrations are responding to the complex factors involved in this CHANS and the efficacy of remediation efforts, systematic water quality and quantity monitoring and associated funding should be a priority. Further, the water quality of private BHs, which provide untreated water to many people during water restrictions, should be monitored.

Conclusions

This work demonstrated the indirect effect of climate change on groundwater quality in Ramotswa, Botswana. Historic rainfall analyses suggest that events like the 2013–2016 drought are likely to become more frequent in Ramotswa and the South East District, increasing pressure on groundwater resources. The drought led to water restrictions, inducing people with flush toilets to use pit latrines. Groundwater NO_3^- concentrations exceeded the drinking water standard. The presence and extent of NO_3^-, fecal coliforms, caffeine, and paraxanthine in groundwater wells suggest that N fertilizer and livestock waste were not likely major sources of NO_3^- pollution; the most reasonable hypothesis is that human waste from the thousands of pit latrines is a major source of the NO_3^-. High dissolved N_2O concentrations in the groundwater suggested in situ denitrification is happening; however, low DOC concentrations relative to NO_3^- concentrations suggest that denitrification is C-limited. Therefore, ISB providing an additional C source in the aquifer could enhance NO_3^- removal by denitrification. The caffeine, DOC, and N_2O analyses provide new information that advances the understanding of the drivers of Ramotswa groundwater quality and potential solutions.

The CHANS approach allowed us to understand water quality in the larger context of interacting social and ecological drivers. This perspective illuminated the interactions between climate (drought), human behavior, and groundwater quality. Without understanding the human behavior aspects of the nitrate contamination, a driver of the problem (pit latrine use) that is likely to increase in frequency and intensity in coming decades would remain obscured. This CHANS study provides a template for socio-hydrological studies in other regions that may lead to improved predictions of indirect impacts of climate change and more sustainable water management.

In a global context, this story is not unique to Ramotswa. Rapid urbanization in developing countries brings rapid increases in the number of pit latrines, often in communities that use shallow groundwater for drinking (Cronin et al. 2007). Rather than thinking about and managing water supply and sanitation separately, communities, resource managers, and policy makers need to plan a water infrastructure system incorporating and addressing the interconnections between climate, water resources, sanitation, and water infrastructure. Such an integrated system that protects groundwater quality and quantity will ultimately reduce the cost of water treatment, reduce water-borne disease transmission, and strengthen the community's water security and resilience in the face of urbanization and climate change.

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