Title: **Environmental tracers to evaluate groundwater residence times and water quality risk in shallow unconfined aquifers in sub Saharan Africa**

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Highlights:

- 150 hand pump boreholes (HPBs) in Ethiopia, Uganda and Malawi investigated
- Mean residence times of shallow groundwater in the order of 20 to 50 years
- Recharge rates are 30 to 330 mm y^{-1} for investigated sites
- Recharge processes are often rapid enabling contamination from ground surface
- Improved HPB construction would safeguard resource

Key words:

- sub-Saharan Africa
- Environmental tracers
- Groundwater recharge
- Chloride mass balance
- Water quality
- Groundwater resilience

Abstract

In sub-Saharan Africa, shallow aquifer systems are relied on as the main safe and secure water resource available to rural communities. Information on the sustainability and vulnerability of groundwater abstraction is becoming increasingly important as groundwater development increases. As part of the UpGro Consortium Project- Hidden Crisis, 150 hand pumped boreholes (HPBs), ranging between 15 to 101 m depth were investigated to examine the resilience of aquifer systems in the Ethiopian Highlands, and the crystalline basement rocks of Uganda and Malawi. Environmental tracers (chlorofluorocarbons (CFCs), SF_6, chloride and the stable isotopes of water), water quality indicators (nitrate and E. coli), and groundwater-level time series data were used to estimate groundwater residence time and recharge at a regional scale (100-10,000 km^2) and investigate the risks to water quality and water supply over different timeframes, and geological and climatic environments. Average estimated recharge rates using three different techniques (CFCs, chloride mass balance, water table fluctuation method) were between 30–330, 27–110 and 30–170 mm y^{-1}, for sites in Ethiopia, Uganda and Malawi, respectively. These estimates of recharge suggests abstraction from dispersed low-yielding HPBs is sustainable. Comparison of stable isotopes in rainfall and groundwater indicates that there is little evaporation prior to recharge, and recharge events are biased to months with greater rainfall and more intense rainfall events There was a weak correlation between nitrate and CFCs within all three countries, and no correlation between E. coli and CFCs within Ethiopia or Malawi. The presence of E. Coli at a large proportion of the sites (Ethiopia = 38 %, Uganda = 65 % and Malawi = 47 %) suggests rapid transit of contaminated surface water into the borehole and its presence in groundwater that has CFC-12 concentrations less than 75 pg kg^{-1} indicates mixing of very young water with water more than 40 years old. The rapid transit pathways

are most likely associated with damaged HPB headworks and poor construction. In several monitored HPBs, daily drawdown due to pumping, drew the groundwater levels close to the base of the HPB, indicating that these HPBs were located in parts of the aquifer with low permeability, or were poorly designed, offering limited capacity for increased demand. Improved HPB siting and construction, coupled with groundwater level monitoring are required to capitalise on the more resilient groundwater within the shallow aquifers and safeguard adequate and good quality water supply for rural communities.

1 INTRODUCTION

In sub-Saharan Africa, shallow aquifer systems are often the only safe and secure drinking water resource available to a rural community, particularly during drought (MacAllister et al., 2020; MacDonald et al., 2019). Hand pumped boreholes (HPBs) are being installed at an increasing rate to access safe drinking water (Fisher et al., 2015; MacDonald and Calow, 2009; Truslove et al., 2019) and currently comprise at least 50 % of rural water supply (UNICEF and WHO, 2019). With increased pressure from population growth and climate change it becomes increasingly important to understand and characterise the sustainability of these sources – a key aspect of United Nations Sustainable Development Goal 6. Central to investigating the resilience of these supplies is characterising recharge mechanisms and how they vary across different geological, land use and climatic environments (Edmunds, 2012; Scanlon et al., 2006; Taylor et al., 2013).

Estimates of groundwater recharge (focused and/or diffuse) can be determined using a range of physical, empirical, chemical, tracer and modelling techniques for different spatial and temporal scales (Healy, 2010). There are advantages and limitations to each method and some methods provide additional information on recharge processes and hydrogeology. The water balance method is often used to estimate groundwater recharge over time scales of days to years but requires a high frequency of data from multiple sources (e.g. vegetation rooting depth, evapotranspiration, soil moisture conditions, etc). The water table fluctuation method is widely used to estimate recharge because data on groundwater levels over time is relatively easy to collect, however, the method does require knowledge on the specific yield of the aquifer (Healy and Cook, 2002). Environmental tracers, including chlorofluorocarbons (CFC-11: trichlorofluoromethane- $CFCl_3$ and CFC-12:

dichlorodifluoromethane- CF_2Cl_2) and sulphur hexafluoride (SF_6), have been widely used to determine groundwater residence times, geochemical processes and apparent ages of young groundwater (Busenberg and Plummer, 2000; Chambers et al., 2018; Cook and Solomon, 1995; Cook and Solomon, 1997; Gooddy et al., 2006; Zoellmann et al., 2001). Environmental tracers that allow estimates of groundwater age can be used to determine whether shallow aquifers are susceptible to anthropogenic contamination (Alikhani et al., 2016; Ekwurzel et al., 1994; MacDonald et al., 2003; Morris et al., 2005). Groundwater age can also be useful for estimating aquifer recharge (Healy, 2010; Zuber et al., 2011). Chloride, another environmental tracer, has been applied in many different environments to determine long-term average groundwater recharge rates (Eriksson and Khunakasem, 1969), including in sedimentary aquifers (e.g. Allison and Hughes, 1983; Foster et al., 1982), fractured rock aquifers systems (Cook, 2003) and wet and dry climatic regions (de Vries and Simmers, 2002; Edmunds and Gaye, 1994). Comparison of recharge rates with groundwater demand can provide information on aquifer resilience and help determine water security (Alley and Alley, 2017; Calow et al., 2010; MacDonald and Calow, 2009). The stable isotopes of the water molecule (oxygen-18 and deuterium) have also proved useful in evaluating groundwater recharge processes and identifying the contribution of various water sources (Kendall et al., 2003).

The aim of this study was to investigate the resilience of the shallow groundwater systems to support community hand pumped water supplies in sub-Saharan Africa. We investigated a range of geological and climatic environments in Ethiopia, Uganda and Malawi. Environmental tracers (CFCs, SF_6, chloride and the stable isotopes of water), water quality indicators (nitrate and E. coli), and groundwater-level time series data from 150 shallow HPBs in the investigated countries were used to determine groundwater recharge at a regional scale. We then use these data to discuss the sustainability of groundwater abstraction from HPBs in rural Africa and investigate the risks to water quality. Furthermore, the capacity of the aquifer and hand pump borehole construction were explored to identify some of the factors that might limit the ability of these sources to meet community water supply demands and World Health Organisation water quality criteria guidelines and standards.

2 STUDY AREA: CLIMATE, GEOLOGY AND HYDROGEOLOGY

The following section provides a general description of the regional climate, geology and hydrogeology of survey areas in the three countries: Ethiopia, Uganda and Malawi (Figure 1). Further details can be found in the Supplementary Material. In each country, groundwater provides the dominant domestic water supply to the rural population via community HPBs (UNICEF and WHO, 2019).

2.1 Ethiopia

The regional geology of Ethiopia is dominated by volcanic rocks but there are also significant areas of sedimentary and metamorphic rocks (Kebede, 2012) (Figure 1 and Figure S1 Supplementary Material). Most people live in the highland areas, which are underlain by the volcanic aquifers, making these the most important hydrogeological domain for rural water supply. These can have moderate to high transmissivity – but porosity is often low, 2 to 10 % (Kebede, 2012; MacDonald et al., 2012). Most of the rainfall in Ethiopia occurs between June and September, however, in some areas there is minor rainfall between March and May. There is little or no rainfall between November and February.

The four main survey areas in Ethiopia are located within the districts of Ejere (9.0°, 38.38°), Abeshege (8.32°, 37.63°), Sodo (8.22°, 38.51°) and Mecha (11.39°, 37.14°) (Figure 1 and Figure S1 Supplementary Material). Five nearby climate stations include Addis Ababa, Bale Robe, Awassa, Bahar Dar and Jimma where the average annual rainfall (1982-2012) is 1165, 941, 1091, 1416 and 1766 mm y^{-1}, respectively. The annual average daily air temperature in the wet season at these stations is 16.5, 15.9, 20, 20.2 18.2 and 19.3 °C, respectively. The average elevation for all five stations is 1991 metres above sea level (m ASL) and ranges from 1652 to 2450 m ASL. The Ejere district is located 45 km to the west of Addis Ababa and the majority of the HPBs are located within the volcanic basalt aquifers, although some HPBs (less than 5 where the geology was known) are within the unconsolidated Quaternary sedimentary aquifers, which despite being shallow, are high yielding. The Abeshege district is located 150 km to the southwest of Addis Ababa and the HPBs are completed in the highly weathered basalt aquifer, which has highly variably transmissivity (2 to 6,000 m^2 d^{-1}). The Sodo district is located 85 km to south-southwest of Addis Ababa and the majority of the HPBs are within a pyroclastic tuff deposit, with moderate transmissivity (15 to 110 m^2 d^{-1}). The Mecha district is located 35 km to the south-west of the township of Bahar Dar in the Lake Tana basin (350 km to the north of Addis Ababa) and the main productive aquifers are within the fractured and strongly weathered Quaternary basalts, which have transmissivity between 100 and 200 m^2 d^{-1} (Kebede et al., 2005).

2.2 Uganda

The regional geology of Uganda is dominated by crystalline basement rocks, which constitute 90 % of the land area, and are covered by a thick layer of weathered saprolite material (Taylor and Howard, 2000; Taylor and Howard, 1998) (Figure 1 and Figure S2 Supplementary Material). The major aquifer systems are within the weathered (saprolite) and fractured Precambrian crystalline basement rocks, which typically have low storage and transmissivity (0.1 to 30 $m^2 d^{-1}$) and low porosity, 6 to 10 % (Cuthbert et al., 2019; Tindimugaya, 2008). These saprolite and fractured bedrock aquifers are the most widely used for shallow HPBs with yields generally between 0.1 and 3 L s^{-1} (Taylor et al., 2003).. Rainfall in Uganda occurs throughout the year, and is characterised by two wet seasons with heavier rainfall from March to May and September to November with less rainfall from December to February and June to August (Figure 1 and Figure S2 Supplementary Material).

The sampled HPBs within Uganda were clustered together in three survey areas: the first survey, 60 km to the north of the capital city, Kampala in the Luwero district (0.83°, 32.51°) of Central Uganda, the second survey in Northern Uganda in the Oyam district (2.44°, 32.52°) situated 45 km to the northwest of the township of Lira, and the third survey area in Eastern Uganda in the districts of Kumi (1.59°, 33.95°) and Budaka (1.14°, 33.95°) (Figure 1 and Figure S2 Supplementary Material). Four nearby climate stations include Kampala, Masindi, Lira and Soroti where the average rainfall (1982-2012) is 1264, 1345, 1218 and 1365 mmy^{-1}, respectively. The annual average daily air temperature for these four stations is 23.2 ^{o}C and ranges from to 21.2 to 25.3 ^{o}C, and the average elevation is 1151 m ASL and ranges from 1056 to 1223 m ASL.

2.3 Malawi

Malawi is positioned at the southern end of the East African Rift system, which strongly influences the topographic, climatic, hydrological and geological features. The country is underlain mainly by crystalline basement rock (Schlueter, 2006), which forms a fractured rock aquifer with relatively low transmissivity values (5 to 35 $m^2 d^{-1}$) and low porosity, 8 to 12 % (Mkandawire, 2004; Smith-Carington and Chilton, 1983). Cretaceous sedimentary rocks occur in the Shire Basin in southern Malawi, and alluvium is associated with Lake Malawi. Malawi has a sub-tropical climate characterised by a wet season from November to March and dry season from April to October (Figure 1 and Figure S3 Supplementary Material).

The sampled HPBs within Malawi were from three survey areas and include the districts of Lilongwe- central region (13.97°S, 33.79°), Balaka and Machinga districts- southern region (14.95°S, 35.25°) and Nkhotakota-central region, near Lake Malawi (13.04°S, 34.08°) (Figure 1 and Figure S3 Supplementary Material). The closest climate stations to the survey areas are Lilongwe, Kasungu and Balaka, where the average annual rainfall (1982-2012) is 860, 822 and 971 mm y^{-1}, respectively. The average daily air temperature for these three stations is 22.4 ^{o}C and ranges from to 16.1 to 26.8 ^{o}C, and the average elevation is 913 m ASL and ranges from 634 to 1056 m ASL.

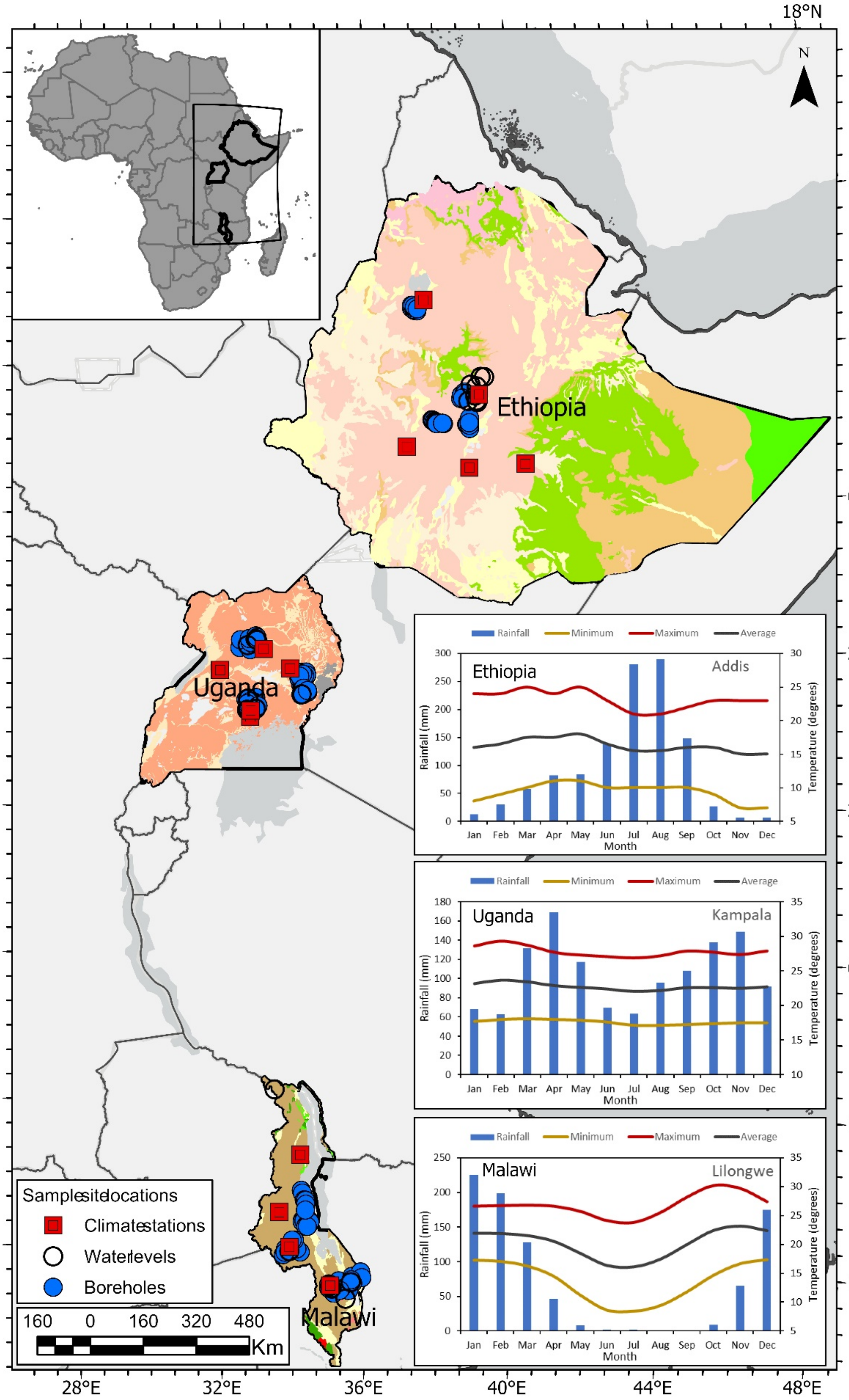

Figure 1. Location map showing the three countries: Ethiopia, Uganda, and Malawi. The regional geology and the sample site locations are shown in greater detail in the Supplementary Material. Inset figures show the variation in the average monthly rainfall and air temperature (minimum, maximum and average) for selected climate stations in each country.

3 APPROACH

3.1 Groundwater Age Indicators- Chlorofluorocarbons and Sulphur Hexafluoride

Chlorofluorocarbons and SF_6 are synthetic organic compounds that are produced for a range of industrial and domestic purposes and are an effective tracer for young-modern groundwaters (Chambers et al., 2018; Gooddy et al., 2006). Comparison of the concentrations of CFC and SF_6 in groundwater with atmospheric concentrations (Figure S4 Supplementary Material) indicates the time at which a groundwater sample was last in contact with the atmosphere, and hence the groundwater residence time (Busenberg and Plummer, 1992; Szabo et al., 1996). Where mixing of different groundwaters occurs, either within the aquifer or at the well-head, the residence time obtained with these tracers will represent a mean residence time (MRT). The use of multiple tracers can be useful to resolve possible discrepancies in the mean residence times between different tracers and provide information on the mixing process (McCallum et al., 2014).

Environmental tracers that provide information on groundwater residence time can be used to estimate aquifer recharge rates, particularly in sedimentary aquifers (Cook and Bohlke, 1999). If sampling takes place close to the water-table, then the recharge rate (*R*) may be estimated by:

$$R = \frac{Z\theta}{t} \qquad \text{(Equation 1)}$$

where *Z* is the depth below the water-table, θ is the porosity of the aquifer and *t* is the groundwater residence time.

In heterogeneous aquifers, such as fractured rocks, profiles of environmental tracers with depth can also provide information on the depth of circulation of groundwater and vertical connectivity (Cook et al., 1996; Manning and Solomon, 2005). Groundwater age can provide information on the susceptibility of groundwater to contamination, with young groundwater much more susceptible to contamination than older groundwater (Manning et al., 2005). Plots of contaminant concentrations versus groundwater age can provide information on the history of contamination of aquifers, particularly for diffuse source contaminants such as nitrate (Böhlke and Denver, 1995). If contaminants occur in groundwater that has an apparent age that pre-dates the use of the particular contaminants, then this implies mixing of young and older water.

3.2 Stable Isotopes of Water

Oxygen-18 ($\delta^{18}O$) and deuterium ($\delta^{2}H$) ratios in water can be used to evaluate the contributing sources and origins of recharge waters and evaluate groundwater recharge mechanisms and processes (Clark and Fritz, 1997; Coplen et al., 1999; Kendall and McDonnell, 2012). Other factors being equal, the isotopic composition will be more depleted during heavy rainstorms than during lighter events. This might cause the mean isotopic composition of groundwater to be more depleted than mean rainfall, if groundwater recharge primarily occurs during large rain events (Jasechko and Taylor, 2015). Also, rainfall at higher elevations will be more isotopically depleted than rainfall at lower elevations, and rainfall during cold climatic periods will be more depleted than rainfall during warmer periods (Ingraham, 1998; Mazor, 2003).

3.3 Chloride Mass Balance

The chloride mass balance (CMB) technique can be used to determine long-term groundwater recharge rates (Eriksson and Khunakasem, 1969).

If steady state conditions can be assumed, then the mass balance between chloride input in rainfall and chloride output in recharge can be used to estimate the recharge rate (*R*):

$$R = \frac{PC_p}{C_{gw}} \qquad \text{(Equation 2)}$$

where *R* is the estimated recharge [mm y^{-1}], *P* is the average annual rainfall [mm y^{-1}], C_p is the chloride concentration of rainfall [mg L^{-1}], and C_{gw} is the chloride concentration of groundwater [mg L^{-1}].

Equation 2 assumes that chloride is not gained or lost from the system, and that all chloride ultimately enters the aquifer as recharge. It also assumes negligible contribution of chloride from weathering or fertilisers. The method is most applied in arid areas, where runoff is negligible. In catchments with steep terrain and higher rates of surface runoff, estimated recharge rates will have a greater uncertainty as chloride will be removed from the catchment via the surface water drainage network (Wood, 1999). In data-poor areas, surface runoff can be estimated from global data bases and hydrological models (e.g. Alcamo et al., 2003; Sutanudjaja et al., 2018), as was the case for this study. Where runoff is significant and not accounted for, the method will provide an upper bound for the recharge rate.

3.4 *Groundwater Level Hydrographs*

In this study, we use daily variations in the water-table as an indication of borehole reliability (water demand relative to aquifer yield), and seasonal variations as an indication of the aquifer recharge rate. Aquifer recharge (*R*) is thus estimated from the seasonal variation in the water-table according to:

$$R = S_y \frac{\Delta h}{\Delta t} \qquad \text{(Equation 3)}$$

where Δh is the change in water level height over a specific time interval Δt, and S_y, is the specific yield. Specific yield typically varies between 0.02 and 0.2 (Healy and Cook, 2002; Johnson, 1967).

Higher specific yield values are typical for unconsolidated sands and gravels, and lower values for igneous and metamorphic rocks and some lithified sedimentary rock. Short-term fluctuations in the water-table reflect the ability of the HPB and surrounding aquifer to supply the daily groundwater needs of the community. The ratio between the magnitude of the daily fluctuations in water levels and the thickness of aquifer penetrated by the borehole is thus a performance measure of the HPB water supply.

3.5 Sustainability of Groundwater Supply

Interpretation of environmental tracer data in combination of hydraulic head data from the HPBs thus allows estimates of the sustainability of groundwater to supply local communities:

(i) In combination with estimates of groundwater use, estimates of aquifer recharge (derived from CFCs, SF_6, chloride, and from seasonal variations in groundwater levels) provide information on the sustainability of water supply aquifers from a quantity perspective.

(ii) Understanding of recharge processes (oxygen-18 and deuterium) can provide information on the likely impact of climate change on groundwater systems and evaluate the contributing sources and origins of the recharge waters.

(iii) The apparent age of groundwater sampled at the HPBs (CFC and SF_6) provides information on the susceptibility of aquifers to contamination. Comparison of groundwater age with other water quality parameters (e.g. NO_3 and E. coli), provides information on aquifer contamination history and contaminant sources.

(iv) Evaluation of watertable fluctuations induced by pumping with the construction specifications of the HPB provides information on the performance of the HPB to meet present and increasing water demands.

4 METHODS

As part of Survey 1 (conducted in 2016) of the UpGro Consortium Project - Hidden Crisis: unravelling current failures for future success in rural groundwater supply, 600 hand pumped boreholes (HPBs) located in rural communities in Ethiopia, Uganda and Malawi were visited via a two-stage stratified sampling process, and sampled for a range of physical hydrological variables (Kebede et al., 2017; Mwathunga et al., 2017; Owor et al., 2017). As part of Survey 2 (conducted in 2017 and 2018), 150 of the 600 HPBs sampled in Survey 1 were selected for more detailed and comprehensive analysis of borehole construction, water quality and groundwater sampling (Kebede et al., 2019; Mwathunga et al., 2019; Owor et al., 2019) (Table S1 Supplementary Material). The HPBs were sampled in Ethiopia between January and December 2017, Uganda between June and November 2017, and Malawi between September and December 2017.

The groundwater parameters analysed in Survey 2 included alkalinity, pH, dissolved oxygen (DO), oxidation-reduction potential (ORP), specific electrical conductivity (SEC), temperature, major ions and trace elements, stable isotopes of water, noble gases, chlorofluorocarbons, sulphur hexafluoride and E. coli. Individual water quality sensors were used to measure pH, specific electrical conductance (SEC), dissolved oxygen (DO), oxidation-reduction potential (ORP) and temperature during purging of the boreholes at the outlet of a portable Grundfos submersible pump, which was deployed in the borehole to conduct an aquifer test after the hand pump infrastructure had been removed. Total alkalinity (as $CaCO_3$ concentration) was measured in the field using a HACH™ titration kit (www.hach.com). E. coli was measured in the field using Aquagenx® bags. Samples are reported as Most Probable Number (MPN 100 mL^{-1}; equivalent to Colony Forming Units-CFU 100 mL^{-1}) of total coliforms, thermotolerant coliforms and E. coli in water, based on the World Health Organisation: Guidelines for Drinking Water Quality (WHO, 2017).

4.1 Environmental Tracers

CFC-11, CFC-12 and SF_6 samples were collected in glass bottles (45, 50 and 46 samples in Ethiopia, Uganda and Malawi, respectively), via the sampling pump outlet hose, which were submerged in an over-flowing bucket to avoid atmospheric contamination, as described by the IAEA (2006). Groundwater samples for major and trace elements were filtered using a 0.45-micron filter, collected in 60 mL HDPE plastic bottles, cation samples were acidified with nitric acid (1 % by volume HNO_3). Further details on the groundwater and rainfall sampling and the laboratory analysis techniques are described in the Supplementary Material.

4.2 Groundwater Level Hydrographs

Approximately 10 HPBs in each country were equipped with pressure transducer dataloggers to record water level time series (hydrographs). These HPBs were used to assess groundwater recharge rates and recharge mechanisms based on the water-table fluctuation method (Healy and Cook, 2002). The HPBs monitored for water levels were not the same as those sampled for hydrochemistry. In most cases, drawdown during the day in response to pumping and recovery of water levels at night, is apparent from the logger data and provides some indication of the borehole performance and aquifer properties (Bonsor et al., 2014; MacDonald et al., 2019). Because of pumping, water level responses to individual rainfall events are difficult to discern, which in-turn makes it difficult to identify individual recharge events. However, annual fluctuations in water level in areas with well-defined wet seasons provide some information on annual recharge.

The installation and setup of pressure transducers was different in each country in order to accommodate the different types of pumps that were installed and the design of the well headworks. In Ethiopia, measurements were recorded from July 2016 to November 2018 with a sample frequency between 15 min and 1.5 hours. In Uganda, measurements were recorded from May 2016 to August 2018 with a sample frequency of 20 minutes. In Malawi, measurements were recorded from August 2013 until August 2018 with a sample frequency of 15 minutes. All measurements were corrected for atmospheric changes using a nearby barometric pressure logger. The longer length of record in Malawi allows some assessment of long-term trends, which is not possible with the data from Ethiopia or Uganda.

Five statistics are calculated from the hydrograph data:

1) Mean static water level (SWL). This is the mean recovered water level i.e., after pumping events are excluded.
2) Average annual head fluctuation. The average annual head fluctuation is calculated from the minimum and maximum water levels, after drawdown associated with individual pumping events is excluded.
3) Seasonal head difference. This is the difference between the minimum and maximum water levels (excluding pumping events).

4) Daily head difference percentiles. This describes the magnitude of daily head fluctuations, mostly attributed to groundwater pumping. Median and 90th percentile values are reported, where the latter is the daily head difference that is exceeded 10 % of the time.
5) Borehole reliability. This is calculated as:

$$1 - \frac{P_{90}}{H} \qquad \text{(Equation 4)}$$

where P_{90} is the 90th percentile of the daily head difference, and H is the depth of the water column within the borehole (total borehole depth minus mean SWL). Where the total borehole depth is greater than 75 m, the depth of water within the borehole is calculated as 75 m minus the mean SWL. This acknowledges that hand pumps can only extract water from water-tables shallower than 45 metres for the India Mark 2 and Afridev and up to approximately 75 m for the India Mark 2 deep pump (although not as widely used), and so water below this depth is unavailable to local communities.

5 RESULTS AND ANALYSIS

5.1 *Chlorofluorocarbons*

CFC-11 concentrations of the HPBs sampled in Ethiopia, Uganda and Malawi ranged from 3 to 831, 5 to 793, 6 to 1110 pg kg^{-1}, respectively and the CFC-12 concentrations ranged from 8 to 319, 10 to 420, 0 to 371 pg kg^{-1}, respectively (Figure 2). The broad range of concentrations represents recent water (< 10 years residence time) to waters greater than 40 years old or a mixture of different ages. Fifteen of the 45 samples from Ethiopia had at least one of the CFC compounds higher than the equivalent modern atmospheric concentrations of CFC-11 and CFC-12. Whereas most of the 50 samples from Uganda plot below the atmospheric CFC concentration line (indicative of some degradation of CFC-11) and only 4 of the 50 samples had at least one of the CFC compounds higher than the equivalent modern atmospheric concentrations of CFC-11 and CFC-12. The majority of the 46 samples from Malawi plot below the atmospheric CFC concentration line and 4 of the 46 samples had at least one of the CFC compounds higher than the equivalent modern atmospheric concentrations of CFC-11 and CFC-12. Concentrations above the equivalent modern atmospheric concentrations were considered contaminated by anthropogenic sources (Busenberg and Plummer, 1992) (Table S1, S2 and S3 Supplementary Material). Samples that fall below the atmospheric CFC concentration line are usually interpreted as microbial degradation of CFC-11, which is common in anaerobic environments (Hinsby et al., 2007; Oster et al., 1996; Sebol et al., 2007). Measured dissolved oxygen concentrations of 30 % of the samples from Uganda were less than 0.5 mgL^{-1}, but there was no clear relationship between dissolved oxygen concentration and location of samples on the CFC-11 versus CFC-12 plot (Table S1, S2 and S3 Supplementary Material).

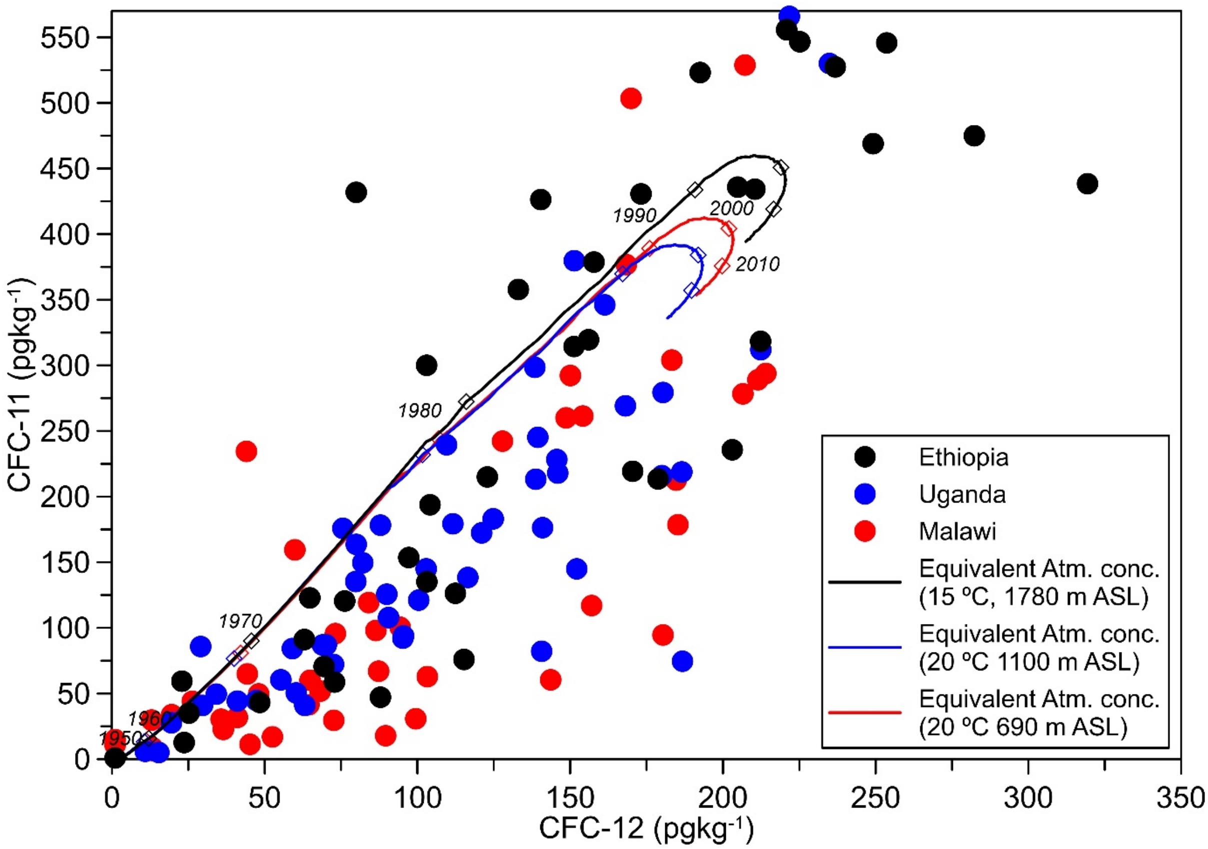

Figure 2. Groundwater CFC-11 versus CFC-12 concentrations ($pgkg^{-1}$) for Ethiopia, Uganda and Malawi. Also plotted is the equivalent CFC concentrations in the atmosphere since CFC production at a recharge temperature of 20 degrees (15 degrees for Ethiopia) at an average elevation of the survey areas (1780 m ASL- Ethiopia, 1100 m ASL- Uganda and 690 m ASL- Malawi). For Ethiopia, 4 of the samples had CFC-11 concentrations above 550 pg kg^{-1}. For Uganda, one sample had a CFC-12 concentration above 420 pg kg^{-1}. For Malawi, 2 samples had CFC-11 concentration above 550 pg kg^{-1}.

5.2 *Tracer depth profiles*

The CFC-12 concentration depth profiles (presented as top of bore screen below the water-table) show modern groundwater up to 40 metres deep (equivalent to 65 m below ground level) in Ethiopia (Figure 3A), 50 metres deep in Uganda (Figure 3D), and 60 metres deep in Malawi (Figure 3G). The presence of CFCs to these depths is indicative of active groundwater circulation and an aquifer system with relatively high vertical connectivity (TABLE S1, S2 and S3 Supplementary Material).

Although there is some scatter, for Ethiopia the CFC-12 concentrations tend to decrease with depth (ρ=-0.63, α<0.01), however, depth trends are not apparent for nitrate (as NO_3^-)

(ρ=0.04, α>0.2) or E. coli (ρ=-0.17, α>0.2) (Figure 3A, B and C). The lack of a trend with depth for nitrate and E. coli is somewhat surprising, as deeper groundwater should be older than shallower groundwater, and hence would be expected to have lower nitrate and E. coli concentrations. The lack of a strong trend in CFC-12 concentration with depth, may reflect large spatial variations in recharge rates. However, it may also indicate mixing of water within the aquifer or within the borehole (see further discussion in sections 5.3 and 5.4).

In the samples from Uganda there is a general trend of increasing MRT (decreasing CFC concentrations) with increasing depth (ρ=-0.40, α<0.02, when only wells with screen lengths less than 20 m are considered). There is also a weak trend in E. coli concentration with depth (ρ=-0.34, α<0.1), but no clear relationship between nitrate concentration and depth (ρ=-0.02, α>0.2; Figure 3D,E,F). As was found in Ethiopia, the sampled HPB's from Malawi showed that there was no correlation between E. coli concentrations and depth (ρ=-0.07, α>0.2) (Figure 3G, H and I).

Groundwater recharge within the last 30 years should have CFC-12 concentrations of between 121 and 153 pg kg^{-1}, whereas CFC-12 concentrations less than 75 pg kg^{-1} indicate groundwater MRT in excess of 40 years. The lifetime of E. coli in groundwater is generally less than 3 to 4 months, depending on the environmental conditions (e.g. microflora, temperature) (Edberg et al., 2000). The presence of E. coli in groundwater that has CFC-12 concentrations less than 75 pg kg^{-1}, therefore indicates mixing of very young water with water more than 40 years old.

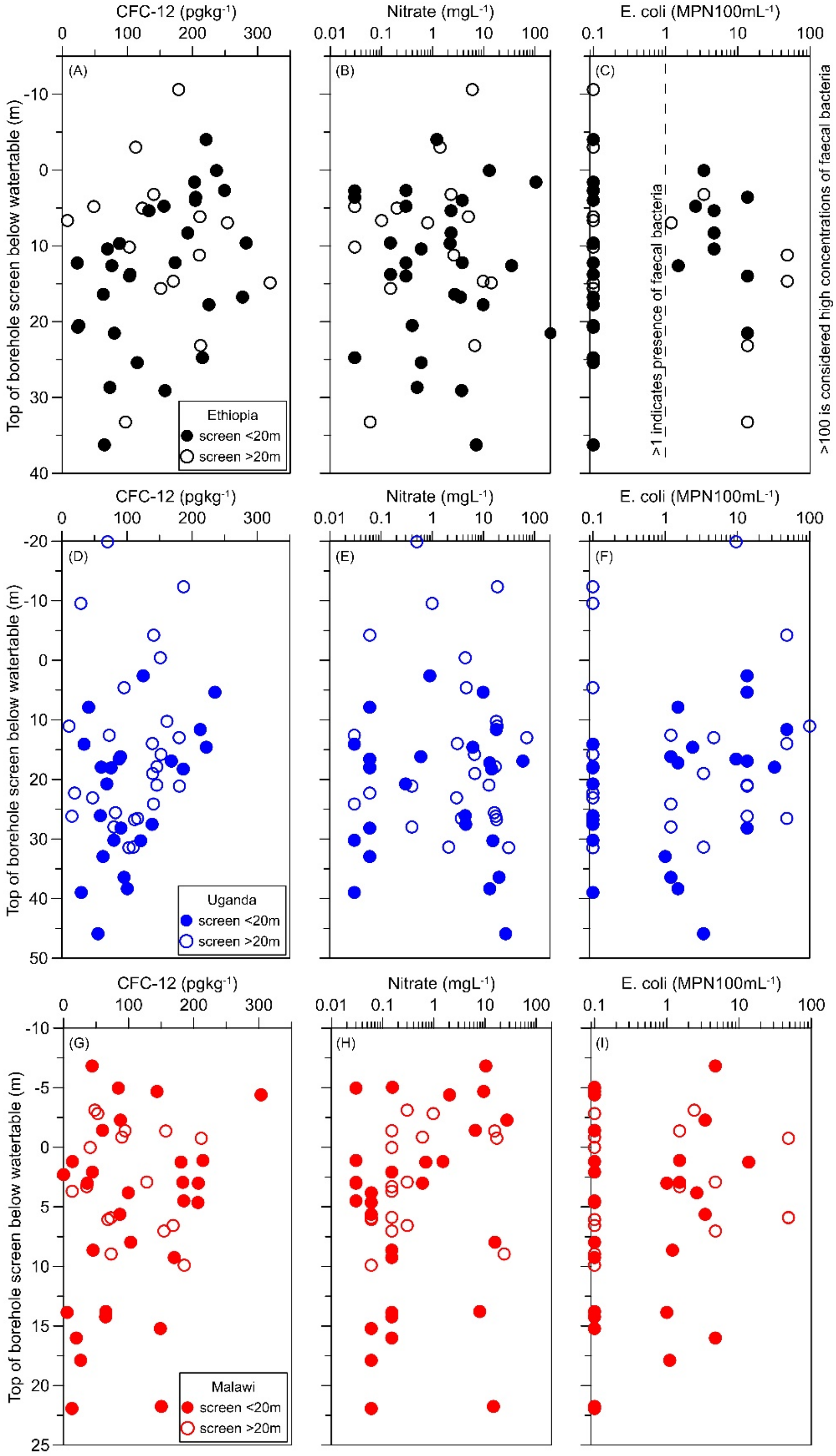

Figure 3. Groundwater CFC-12, nitrate and E. coli concentrations from each site versus top of borehole screen below the water-table for each site for (A, B, C) Ethiopia, (D, E, F) Uganda, and (G, H, I) Malawi. Negative depth values occur when the top of the screen is found to be above the water level in the borehole. Solid symbols are HPBs that have a screen less than 20 m long, whilst hollow symbols are HPBs that have screens longer than 20 m. One sample for E. coli from Uganda had a concentration of 9,435 MPN 100 mL^{-1} and is not shown. The dashed vertical line (>1 MPN 100 mL^{-1}) indicates the presence of faecal bacteria contamination and >100 MPN 100 mL^{-1} indicates high concentrations of faecal contamination.

5.3 Chlorofluorocarbons versus water quality indicators

A summary of the groundwater nitrate, E. coli, and CFC-12 concentration data are shown in box and whisker plots in Figure 4A, B and C to illustrate the differences between datasets of the three countries. Nitrate concentrations range from 0.03 to 201.3, 0.03 to 70.3, and 0.03 to 26.8 mg L^{-1} with median concentrations of 1.3, 4.4, and 0.2 mgL^{-1} for Ethiopia, Uganda and Malawi, respectively. These results are consistent with those of another large study across the three countries (Lapworth et al., 2020). Measured E. coli values were low in Ethiopia and Malawi (generally <5 MPN 100 mL^{-1}) but moderate in Uganda (generally <10 MPN 100 mL^{-1}).

Comparison of nitrate versus CFC-12 concentrations shows that the majority of the sampled HPBs in Ethiopia have nitrate concentrations less than 5 mg L^{-1} (Figure 4D). E. coli is also low, but was measured as present (> 1 MPN 100 mL^{-1}) in 38 % of the sites (similar to that measured by Lapworth et al. (2020) in a larger survey in Ethiopia), with none considered a high risk (>100 MPN 100 mL^{-1}) by the World Health Organisation: Guidelines for Drinking Water Quality (WHO, 2017). There is only a weak correlation between nitrate and CFC-12 concentration (ρ=0.32, α>0.05), and no correlation between E. coli and CFC-12 (ρ=0.00, α>0.2) or between E. coli and nitrate (ρ=0.20, α>0.2). There are some groundwater samples, despite relatively low CFC-12 concentration (<100 pg kg^{-1}), that have nitrate concentrations above 5 mg L^{-1} indicating anthropogenic contamination. Several samples with low CFC-12 concentrations also have elevated E. coli concentrations (Figure 4E).

In comparison, many of the sampled HPBs in Uganda had nitrate concentrations greater than 5 mg L^{-1}, including some with relatively low CFC-12 concentrations (<75 pg kg^{-1}) (Figure 4D). There is a correlation between CFC-12 and nitrate concentration (ρ=0.41, α<0.02), which becomes more significant when only HPBs with screen lengths less than 20 m are considered (ρ=0.54, α<0.01). Samples with high nitrate concentrations predominantly occur in groundwater with high CFC-12 concentrations and short (< 20 m) borehole-screens, all samples with CFC-12 concentrations above 150 pg kg^{-1} (n=6) have nitrate concentrations above 6 mg L^{-1}. This indicates widespread contamination of nitrate in young groundwater, but at concentrations that are low relative to reported values in surface water and urban areas (Nayebare Shedrack et al., 2014; Withers and Lord, 2002). E. coli was measured as present in approximately 65 % of samples, twice the rate measured using TTCs in a recent larger survey in Uganda (Lapworth et al., 2020), but none were classed as high risk. Elevated E. coli values in the samples also indicated anthropogenic contamination, and there is a similar weak negative relationship between E. coli concentration and CFC-12 concentration (ρ=-0.44, α<0.02) (Figure 4E).

For Malawi, only 35 % of the sampled HPBs had nitrate concentrations greater than 5 mg L^{-1}. E. coli is low with a median value of 2.6 MPN 100 mL^{-1} and was present in approximately 47 % of the sites. There is only a very weak relationship between nitrate concentration and CFC-12 concentration (ρ=-0.26, α>0.1), and several samples with relatively low CFC-12 concentrations (<100 pg kg^{-1}, and even < 50 pg kg^{-1}) have high nitrate concentration (Figure 4D). Elevated E. coli concentrations show no correlation with CFC-12 (ρ=0.11, α>0.2) or nitrate concentration (ρ=-0.02, α>0.2) (Figure 4E and Figure 4F). E. coli is present even where CFC-12 concentrations are low. It is unlikely that groundwater with CFC-12 concentrations below 50 pg kg^{-1} (MRT more than 46 years, and hence recharge years prior to 1973) would have high nitrate concentrations. This suggests a mixture of young and old water and a source of anthropogenic contamination at the well-head, as indicated by the presence of E. Coli.

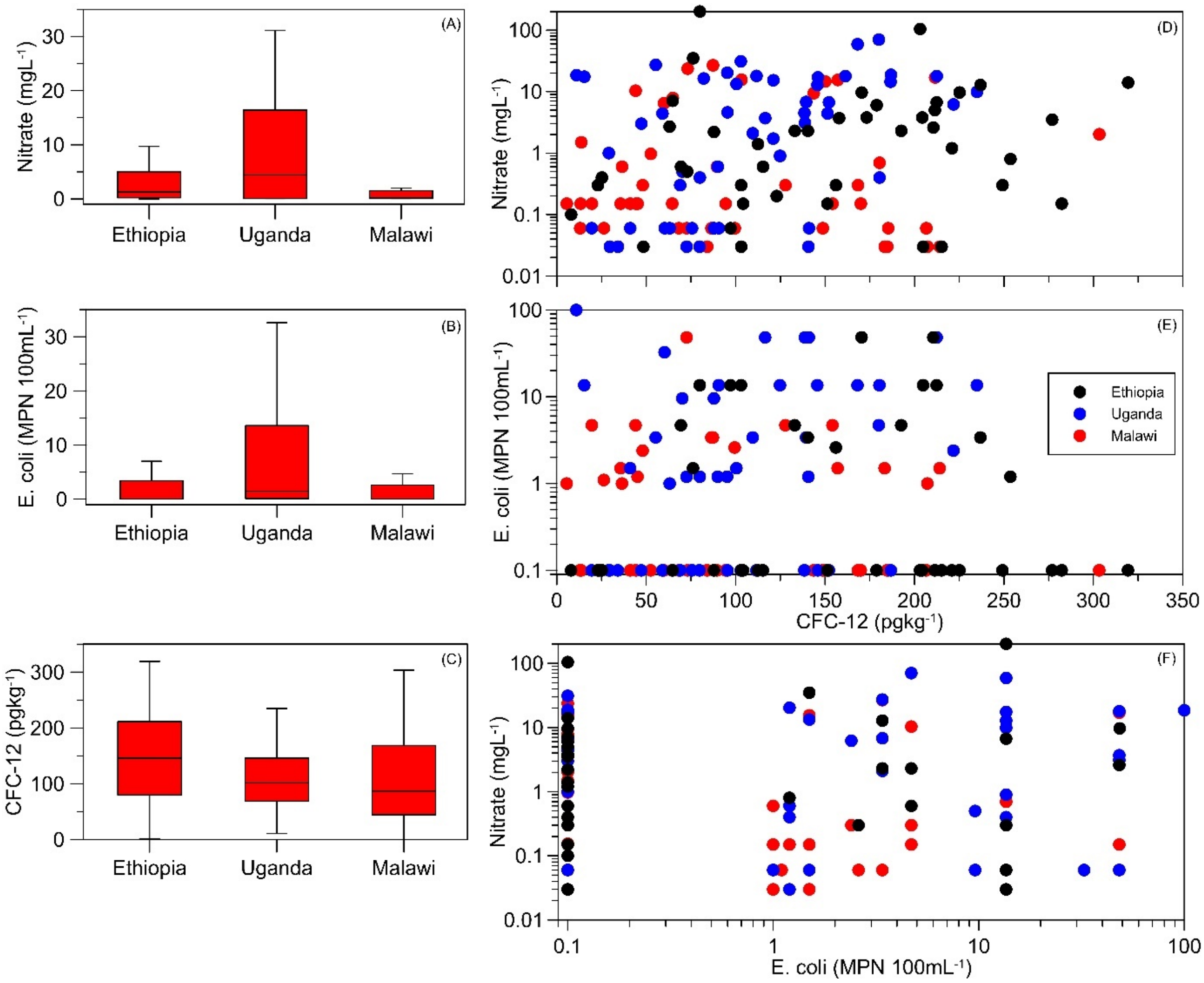

Figure 4. Box and whisker plots of groundwater (A) Nitrate, (B) E. coli and (C) CFC-12 concentrations. Also shown are groundwater nitrate and E. coli versus CFC-12 concentrations (D and E) and nitrate versus E. coli concentrations (F) for Ethiopia (black symbols), Uganda (blue symbols) and Malawi (red symbols).

5.4 Vertical Flow Velocities

The CFC-12 concentrations versus depth below the water-table were compared to several 1D piston flow recharge models to estimate groundwater recharge rates (using a range of plausible porosity values) (Figure 5). CFC-12 concentrations do not show clear trends with depth (as discussed above), which might be due to large spatial variability in recharge rates and mixing of different water residence times within the aquifer or across the borehole screened interval. For the water velocities modelled in Figure 5, a 20 m long borehole-screen would mix water with residence times spanning 20 to 80 years. Due to the non-linearity of the concentration-residence time relationship, particularly for MRT greater than 50 years, this degree of mixing within the borehole-screen causes some uncertainties in MRTs. For this reason, we distinguish between borehole screens greater and less than 20 m in length, although the relationship between measurement concentration and age appears similar for all groups of samples.

Despite the lack of a clear trend with depth, most of the concentrations from Ethiopia are consistent with recharge rates between 20 and 70 mm y^{-1}, considering porosity values between 4 to 8 % (Figure 5A). For Uganda, most of the samples fit within the modelled recharge rate between 40 and 100 mm y^{-1}, using porosity values between 6 and 10 % (Figure 5B), and for Malawi, samples fit within the modelled recharge rates between 30 and 70 mm y^{-1} (porosity = 8 to 12 %) (Figure 5C). HPBs with screens greater than 20 m lie within a similar range of modelled recharge rates. However, since few HPBs were sufficiently deep to sample groundwater older than 1960 (and hence with very low or background CFC concentrations), and due to the evidence of mixing, these recharge values can be considered a lower limit.

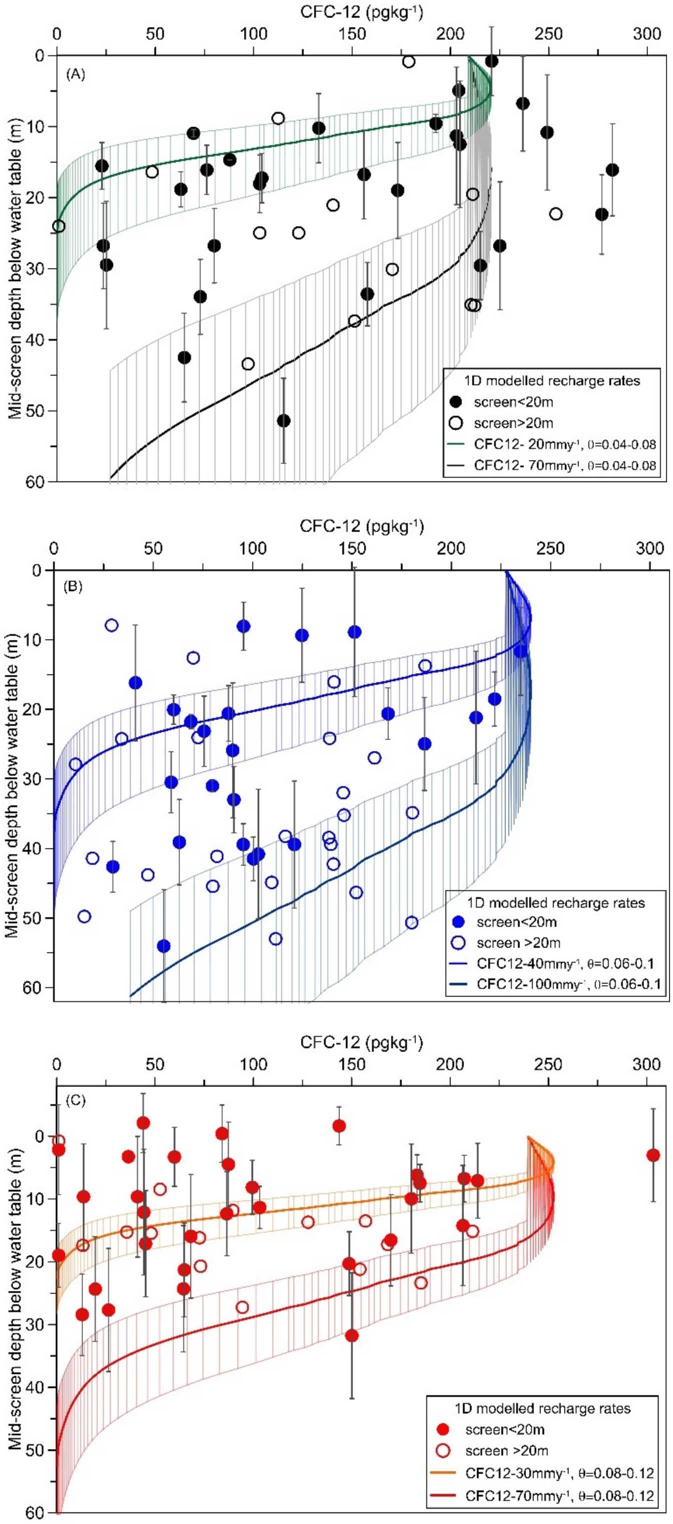

Figure 5. Groundwater CFC-12 concentration versus depth below the water-table for (A) Ethiopia, (B) Uganda and (C) Malawi. Solid symbols are HPBs that have a screen less than 20 m long with screen length shown (error bars), whilst hollow symbols are HPBs that have screens longer than 20 m. Modelled recharge rates between 20 and 100 mm y^{-1} are shown using porosity values between 4 and 12 % (hashed-out area), recharge temperature of 20 degrees (15 degrees for Ethiopia) and an elevation of 1780 m ASL (Ethiopia), 1100 m ASL (Uganda) and 690 m ASL (Malawi). For Ethiopia one sample had a CFC-12 concentration above 300 pg kg^{-1} and is not shown in figure. For Malawi, measured water levels in several HPBs were below the top of the borehole screens and therefore plot above zero metres depth in Figure 5C.

5.5 Groundwater Mixing

Comparing the SF_6 and CFC-12 concentrations can sometimes provide information on mixing between groundwater of different apparent ages and identify anthropogenic contamination (Figure 6). If a sample of rainfall was collected at any time in the past, and concentrations of these two tracers measured, then they should fall along the atmospheric equilibrium line, unless they are affected by other geochemical processes (e.g., sorption, degradation, excess air) (Darling et al., 2012). If a line is drawn connecting any two points on the atmospheric equilibrium line, then sample concentrations along this line are possible by mixing water having these two different ages (e.g. old water- low CFC and SF_6 mixing with modern water- high CFC and SF_6). The shaded mixing envelope is obtained by drawing all possible mixing lines. Concentrations within the mixing envelopes are possible due to mixing of water having different MRTs, whereas concentrations falling outside of these envelopes cannot be ascribed solely to mixing processes (Cook et al., 2017).

Approximately half of the groundwater samples from Ethiopia fall within the mixing envelope, with the other half of the samples either having elevated SF_6 with respect to CFC-12 concentrations or vice versa (Figure 6- black symbols). The high SF_6 concentrations are most likely caused by terrigenic sources of SF_6 (Harnisch et al., 2000) that naturally occur in fluorites, and igneous and metamorphic rock types (rock types that are present in the study area) or modern-air contamination during drilling and borehole development (this is more likely when high-pressure compressed air is used in the drilling method and development of the borehole). A few samples fall just above the mixing envelope, and these might be explained by excess air rather than mixing. Mean residence times for the groundwaters (n= 29; the other 14 samples had elevated CFC concentrations) based on the CFC-12 piston flow model (using a recharge temperature of 15 degrees and an elevation of 1780 m ASL) ranged from 19-53 years, with a median residence time of 36 years.

Most of the samples from Uganda lie within the mixing envelope (Figure 6- blue symbols). Thirteen of the groundwater samples had very high SF_6 concentrations (above 650 fg kg^{-1} and not shown in the figure), well above modern atmospheric concentrations, which most likely can be attributed to terrigenic sources from the fractured granite bedrock aquifer (Harnisch et al., 2000; Lapworth et al., 2013). This was also found in other hydrogeological studies in Uganda that were investigating high-intensity groundwater extractions from the crystalline bedrock aquifers (Maurice et al., 2019). Mean residence times for the groundwaters based on the CFC-12 piston flow model (using a recharge temperature of 20 degrees and an elevation of 1100 m ASL) ranged from 21-53 years, with a median residence time of 40 years. There were three samples that had a residence time greater than 53 years (at or close to detection limit).

For Malawi, SF_6 concentrations are lower than those for Ethiopia and Uganda, with most samples plotting close to the air equilibration line, or within the mixing envelope (Figure 6- red symbols). Mean residence times (CFC-12 piston flow model using a recharge temperature of 20 degrees and an elevation of 690 m ASL) ranged from 28-53 years, with a median residence time of 36 years. There were eight samples that had a residence time greater than 53 years (at or close to detection limit).

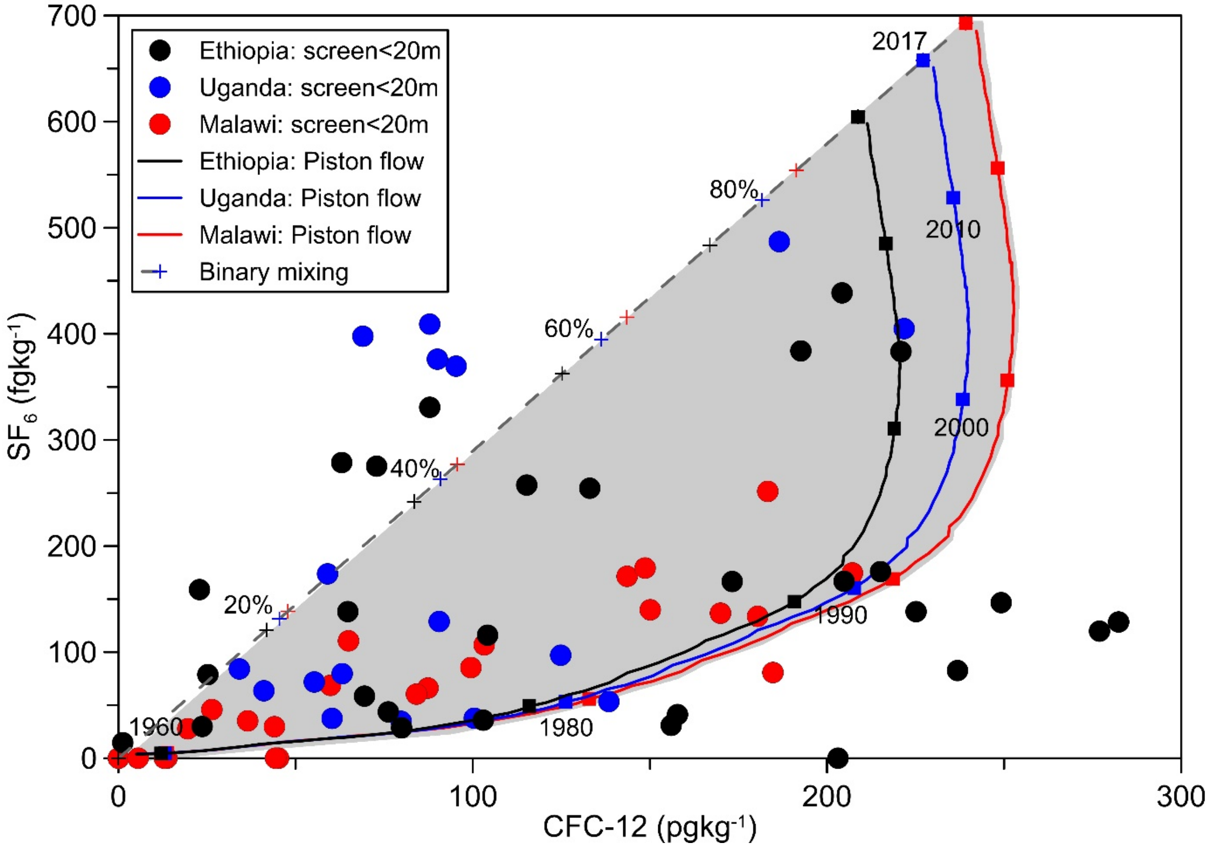

Figure 6. Groundwater SF_6 versus CFC-12 concentrations for each of the three countries. Only HPBs that have a screen less than 20 m long, are shown. Piston flow and binary mixing model curves based on CFC-12 and SF_6 concentration in groundwater at a recharge temperature of 20 degrees (15 degrees for Ethiopia) and an elevation of 1780 m ASL (Ethiopia), 1100 m ASL (Uganda) and 690 m ASL (Malawi). Grey shaded area represents the mixing envelope of different groundwater ages.

5.6 Chloride Mass Balance

The rainfall-weighted average chloride concentration from three rainfall collectors installed in Ethiopia was 0.97 mg L^{-1} (based on rainfall samples collected between 2016 and 2017; Table S4 Supplementary Material). The variation in groundwater chloride from the sampled HPBs (n= 44) ranged from 0.5 to 185 mg L^{-1} (median = 3.7 mg L^{-1}, IQR = 1.4 to 9.2 mg L^{-1}), with 90 % of them having a concentration less than 36 mg L^{-1} (Figure 7). It is worth noting that some of the groundwater chloride values are less than the minimum that was measured in rainfall. This most likely reflects the temporal variability of chloride in rainfall, which has not been fully captured in the 12-month rainfall sampling. Similar findings were found by Edmunds et al. (2002), who used lower chloride values in modelling groundwater recharge than what was measured in rainfall in Nigeria. Nevertheless, these low concentrations of chloride in groundwater indicate locally, very high rates of groundwater recharge. The enrichment of chloride in groundwater compared to chloride in rainfall is likely to be a result of transpiration processes as opposed to an additional chloride source, based on an evaluation of the major ion, and δ^2H and $\delta^{18}O$ data (plots not shown).

From the eight rainfall collectors installed in Uganda, the weighted average chloride concentration in rainfall was 0.6 mg L^{-1} (based on bulk rainfall samples collected between May 2017 and July 2018). The variation in groundwater chloride from the sampled HPBs (n= 51) ranged from 0.2 to 228 mg L^{-1} (median = 6.3 mg L^{-1}, IQR = 2 to 18.2 mg L^{-1}), and 90 % of the HPBs having a concentration less than 44 mg L^{-1} (Figure 7).

The average chloride concentration in rainfall from the five rainfall collectors installed in Malawi was 0.65 mg L^{-1} (based on samples collected between years 2017 and 2018). Measured chloride concentrations in groundwater from the sampled HPBs (n= 46) ranged from 0.7 to 488 mg L^{-1} (median = 5.8 mg L^{-1}, IQR = 3.3 to 15.3 mg L^{-1}), with 90 % having a chloride concentration less than 48 mg L^{-1}.

Despite insufficient reliable hydrological data on surface water runoff, reasonable estimates of surface runoff can be calculated using global hydrology and water resource models and observational data (e.g. Global Runoff Data Centre streamflow records and Global Precipitation Climatology Centre precipitation) (Sutanudjaja et al., 2018). Using the surface runoff data from the grid-based PCR-GLOBWB 2 global hydrology and water resource model by Sutanudjaja et al. (2018), long-term annual average (1980-2014) surface runoff values for the monitored sites in Ethiopia, Uganda and Malawi, ranged from 6 to 170 mm yr^{-1} (median

= 43, IQR = 10 to 116 mm yr^{-1}), 0 to 553 mm yr^{-1} (median = 177, IQR = 63 to 203 mm yr^{-1}) and 0.03 to 94 mm yr^{-1} (median = 11, IQR = 4 to 132 mm yr^{-1}), respectively.

Taking the median and the interquartile range of runoff coefficients into consideration together with the median chloride concentrations in groundwater, the estimated recharge rate for the study areas in Ethiopia, Uganda and Malawi is 327 mm y^{-1} (IQR = 308 to 336 mm y^{-1} or 24 to 26 % of rainfall), 112 mm y^{-1} (IQR = 104 to 117 mm y^{-1} or 8 to 9 % of rainfall), and 86 mm y^{-1} (IQR = 75 to 87 mm y^{-1} or 9 to 10 % of rainfall), respectively.

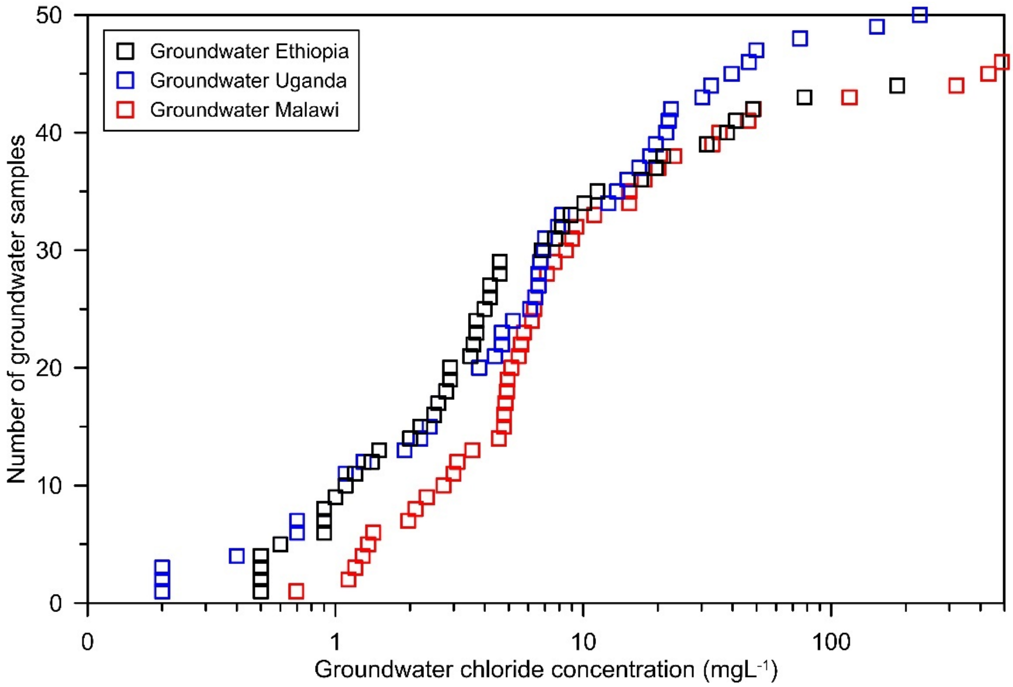

Figure 7. Range of groundwater chloride concentrations from the sampled HPBs in Ethiopia (median concentration was 3.7 mg L^{-1} and IQR = 7.9 mg L^{-1}; black symbols), Uganda (median concentration was 6.3 mg L^{-1} and IQR = 16.2 mg L^{-1}; blue symbols), and Malawi (median concentration was 5.8 mg L^{-1} and IQR = 12 mg L^{-1}; red symbols).

5.7 Stable Isotopes of Water

The stable isotope composition of the groundwater samples from Ethiopia have a large range in values with δ^2H between -33.8 to 9.9 per mil and δ^{18}O of -5.6 to -0.2 per mil (Figure 8A). The groundwater samples plot closely to the local meteoric water line (LMWL) and slightly above the global meteoric water line (GMWL). The data show no noticeable trends of isotopic enrichment or depletion, which suggests relatively rapid infiltration processes. Groundwater samples are mostly depleted in δ^2H and δ^{18}O compared to mean rainfall, which is consistent with recharge dominated by high intensity rainfall events (Jasechko and Taylor, 2015). The isotopically enriched mean rainfall for Addis Ababa in relation to local groundwater isotopic values has been reported as an exception compared to observations in other countries in eastern Africa (Darling and Gizaw, 2002).

The stable isotope composition of the groundwater samples from Uganda plot tightly around the weighted average rainfall value of the LMWL (Figure 8B). The groundwater samples had δ^2H values between -7.2 to 2.2 per mil and δ^{18}O between -2.7 to -1.4 per mil and showed no signs of isotopic enrichment or depletion away from the meteoric water line. Thus, there is very little evidence of rainfall evaporation (deviation to the right of the MWL) as it infiltrates to the aquifer system and that recharge is likely to be occurring all year round. The bulk monthly rainfall samples collected between 2017 and 2018 (Table S4 Supplementary Material), showed a large range in values with δ^2H between -62.6 to 35.1 per mil and δ^{18}O between -9.2 to 4.6 per mil. Months with high rainfall volumes tend to be more depleted in both δ^{18}O and δ^2H, which is likely to be attributed to an amount effect where precipitation is depleted in the heavy isotopes during the wet season (Dansgaard, 1964) (Figure S5 Supplementary Material).

The groundwater samples from Malawi plot closely to the LMWL and GMWL with δ^2H between -41.6 to -28 per mil and δ^{18}O between -6.5 to -4.7 per mil and no clear signs of isotopic enrichment or depletion away from the meteoric water lines (Figure 8C). The groundwater samples are predominantly depleted in δ^2H and δ^{18}O compared to the weighted average rainfall as reported by Banda et al. (2019), indicating that recharge occurs during higher intensity rainfall events.

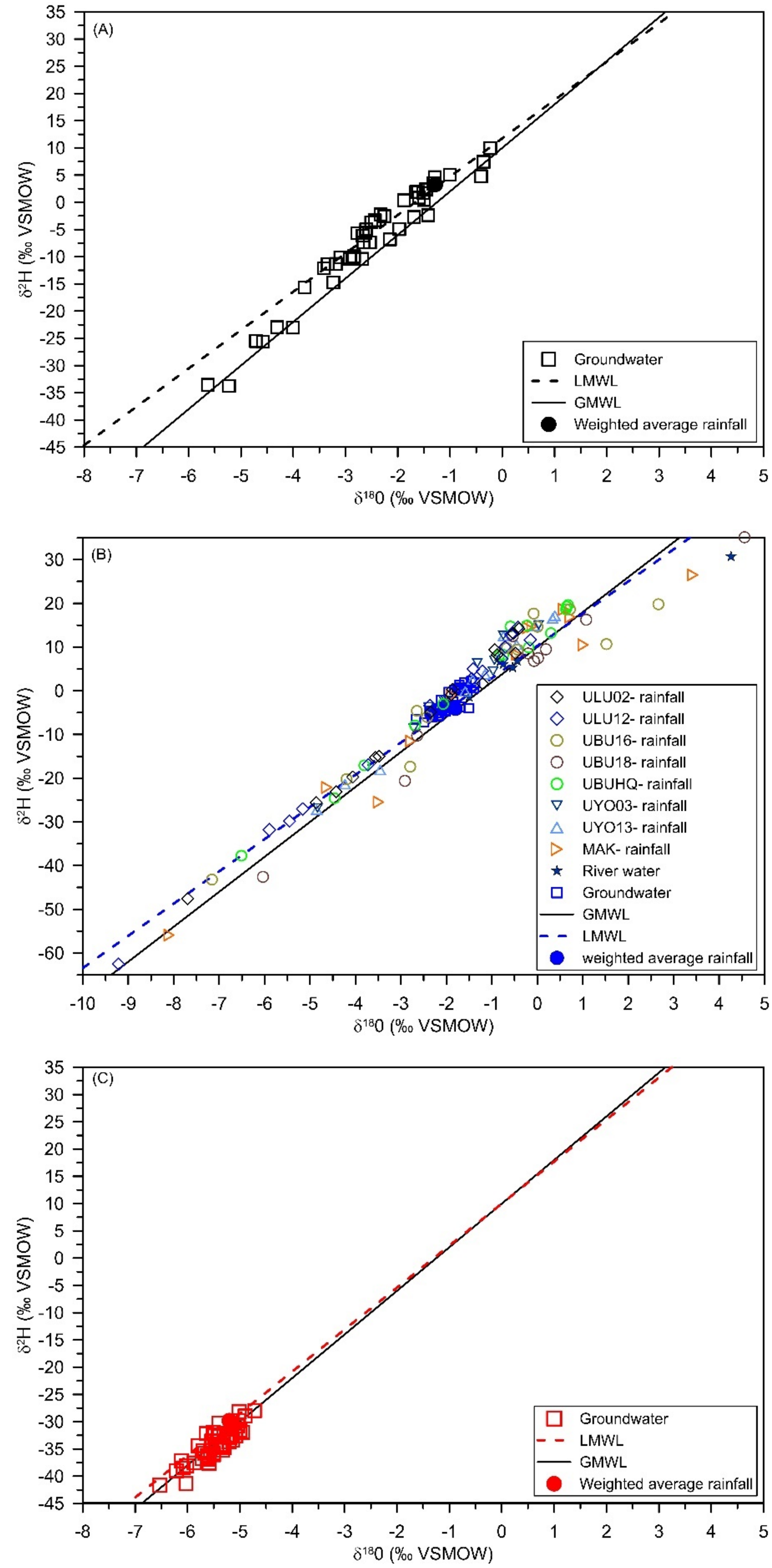

Figure 8. Stable isotopes of water of the groundwater samples collected from the HPBs in (a) Ethiopia, (b) Uganda and (c) Malawi. Also shown is the LMWL and weighted average rainfall from the IAEA GNIP stations at Addis Ababa, Ethiopia and Masaka, Uganda (International Atomic Energy Agency (IAEA), 2018) and a LMWL for Malawi from the study by Banda et al. (2019) compared to the GMWL (Craig, 1961).

5.8 Groundwater Level Hydrographs

Water level time series data was collected from eight groundwater boreholes in Ethiopia (three of which are actively pumped HPBs for water supply), five boreholes in Uganda (all of which are actively pumped), and six boreholes in Malawi (two of which are actively pumped but are missing well depth details and four that are observation wells only) (Table 1). In Ethiopia, HPB depths for the three boreholes that were actively pumped range between 5 and 20 m, whereas the other five monitoring boreholes are deeper– up to 530 m (data not shown in table). Seasonal water levels for the pumped HPBs range between 1.5 and 6 m and water level time series data over a period of two years show that the monitored HPBs that are frequently pumped experience localised drawdown. The magnitude of this drawdown is likely to reflect the borehole efficiency (which is a function of the drilling and borehole construction techniques) as well as the storativity of the aquifer in which the HPBs are completed and the groundwater use (e.g. Figure 9). The median daily drawdown due to pumping ranges from 0.11 m to 1.6 m, and the 90th percentile ranges from 0.52 m to 3 m. The borehole reliability ranges from 0.54 (on 10 % of days, drawdown is more than 46 % of available storage) to 0.94 (on 10 % of days, drawdown is more than 6 % of available storage). However, while this presents a useful indicator of the reliability of individual HPBs to supply the current demand, we have not been able to estimate this statistic for an adequate number of bores to supply adequate statistics at a regional scale.

Groundwater use in these regions is typically less during the wetter months of higher rainfall from July to October compared to the drier months between November to June, which is reflected in the daily water level fluctuations and trends. For example, AABH6, has a daily drawdown that is much greater in March to June than in August to September (Figure 9). The seasonal trend in the water level over a period of one year, fluctuates no more than six metres. Using the magnitude of the seasonal water-table fluctuation and an estimate of the specific yield, we can estimate net aquifer recharge. For the three HPBs shown in Table 1, the seasonal water table fluctuation ranges between approximately 1.5 and 6 m, with a mean value of approximately 3.2 m. Basalt igneous rocks are the dominant rock type in these areas and groundwater lies within fractured bedrock aquifers where the specific yield values are comprised of both the fracture and matrix porosity. Typical values of porosity for basalt rocks range from 3 to 35 % (Fetter, 2001), although values are often at the lower end of the range (Wood and Fernandez, 1988) and hence a specific yield value of between 0.01 to 0.05 may be reasonable (MacDonald et al., 2012), and gives a mean recharge rate between 32 and 160 mm y^{-1}.

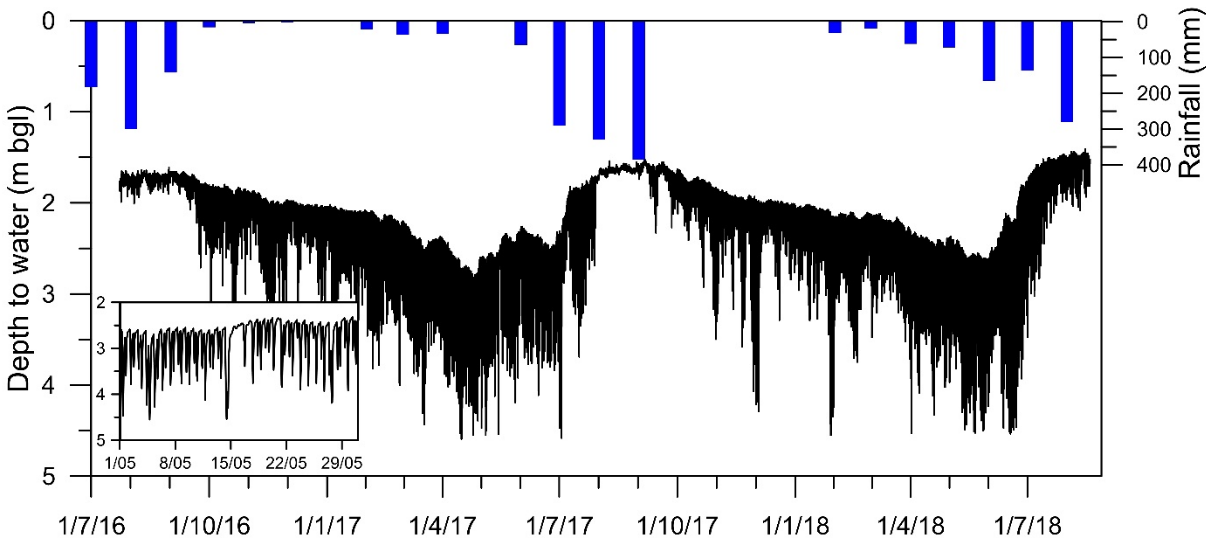

Figure 9. Selected hydrograph showing the seasonal trend in the water-table: HPB AABH6, Becho Plain, and the monthly rainfall. Inset figure shows the drawdown and recovery in the water levels from daily groundwater pumping (HPB AABH6) over a 1-month period in May 2017.

Water level time series data from Uganda is available for five HPBs, all of which are actively pumped for water supply (Table 1). Borehole depths range between 35 and 76 m, and standing water levels range between 1.7 and 32.1 m. Water level time series data over a period of two years confirms that the HPBs that are frequently pumped during the day show signs of rapid localised drawdown and recovery (Figure 10).

In some instances, borehole yields were so low that the water level within the borehole fell below the logger depth when the borehole was actively pumped. Nevertheless, recovery of the water level in the aquifer after a day's pumping is rapid and returns to non-pumped conditions in a matter of hours (Figure 10). The median daily drawdown due to pumping ranges from 3.8 m to more than 13 m, and the 90th percentile ranges from 4.6 m to more than 14 m. Borehole reliability is less than 0.32 on all four HPBs where data is available to calculate this statistic.

For the five HPBs shown in Table 1 from Uganda, the seasonal water table fluctuation ranges between approximately 0.3 and 3.3 m, with a mean value of approximately 1.34 m. Specific yield values range between 0.03 to 0.06 for the fractured crystalline basement rocks in Uganda (Cuthbert et al., 2019; Owor et al., 2009; Taylor et al., 2010; Taylor et al., 2013; Tindimugaya, 2008), which gives a recharge rate between 40 and 80 mm y^{-1}. The specific yield of the aquifers within weathered saprolite overlying the basement rocks would be slightly higher than this value.

Table 1. Summary of the water level data from selected HPBs in Ethiopia, Uganda and Malawi that are actively used for domestic water supply. Also shown are data from unequipped boreholes. Seasonal head difference is the average measured water level difference between the dry and wet seasons. Daily head difference is the median measured water level over the period of a day. Daily head difference and borehole reliability data is shown for pumped HPBs only. HPB depth is unknown for UOY13, MLI08 and MBA19, and so borehole reliability is not calculated.

Country	Site ID	Kebele/ District	Borehole depth (mbgl)	Sample period (years)	Mean SWL (mbTOC)	Average annual head fluctuation range (mbTOC)	Seasonal head difference (m)	Daily Head Diff. (m)		Borehole reliability
								Median	90^{th} Percentile	
Ethiopia	AABH6	Sululta Plain	5	2.00	1.7	1.7-3.2	1.5	0.8	1.5	0.54
Ethiopia	AABH7	Selale Plain	20	2.00	2.2	1-3	2.0	1.6	3.0	0.83
Ethiopia	AABH8	Selale Plain	10	2.00	1.3	0-6	6.0	>0.11[##]	>0.52[##]	<0.94[##]
Uganda	ULU02	Luwero	75.3	1.25	21.3	21-21.8	0.8	>13.8[##]	>14.2[##]	<0.26[##]
Uganda	ULU12	Luwero	55.7	1.25	32.1	31.9-32.2	0.3	5.7	7.3	0.31
Uganda	UOY03	Oyam	46.1	1.25	8.4	7.5-9.2	1.7	10.5	12.1	0.32
Uganda	UOY13	Oyam		1.25	6	5-8.3	3.3	3.8	4.6	n/a
Uganda	UOY16	Oyam	35.9	1.25	1.7	1.4-2	0.6	6.7	6.7	0.20
Malawi	MLI08	Lilongwe		1		2-3	1	>2.1[##]	>3.0[##]	n/a
Malawi	MBA19	Balaka		1	5.6	5-5.5	0.5	0.9	1.16	n/a
Malawi	Chitipa water office	Chitipa	102	3.6	6.5	5.5-7.2	1.7			Not pumped. Observation only.
Malawi	Ntaja water office	Machinga		3.63	15	11.5-18.5	7			Not pumped. Observation only.
Malawi	Balaka water office	Balaka		2.54	4	1-6	5			Not pumped. Observation only.
Malawi	Songani	Zomba	80	3.45	7.5	5-10	5			Not pumped. Observation only.

[##]Daily head difference is under-estimated and borehole reliability is over-estimated because daily drawdown frequency exceeds depth of logger.

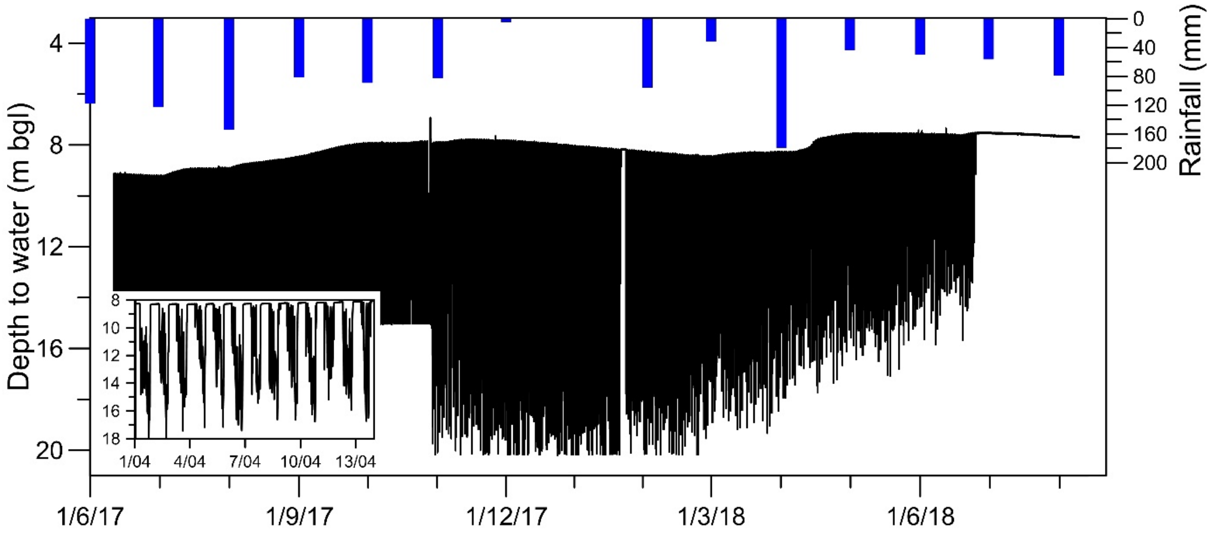

Figure 10. Selected hydrograph from Uganda showing the seasonal trend in the water-table during wetter and drier months of the year- HPB UOY03, Oyam and the monthly rainfall. Inset shows the impacts of groundwater pumping with significant drawdown during daylight hours- HPB UOY03. The logger depth was repositioned deeper in UOY03 mid-November 2017.

Water level time series data from Malawi of dedicated groundwater monitoring boreholes and HPBs that are frequently pumped show the seasonality in the shallow aquifers in response to rainfall and the drawdown and borehole recovery in response to daily pumping (Figure 11). The seasonal trend in the hydrographs shows that it takes approximately three months for the water levels to increase to their peak in response to rainfall and then the recession limb is more gradual, taking approximately eight months to return to the seasonal low (Figure 11A). The seasonal water level fluctuates between 2 and 4 metres between the wet and dry period of the year and based on the available logger data there is no substantive long-term decline in the water levels. Water level data was only available for two actively pumped HPBs, and in one of these, the water level within the borehole fell below the logger depth when the borehole was actively pumped, and so the 90th percentile of daily drawdown cannot be reliably calculated but must be greater than 3.0 m (Figure

11B). For the other, the 90th percentile for daily drawdown is 1.2 m. A summary of the water-table fluctuations for HPBs that were monitored is shown in Table 1.

For the six HPBs shown in Table 1 from Malawi, the seasonal water table fluctuation ranges between approximately 0.5 and 7 m, with a mean value of approximately 3.37 m. Previous investigations in Malawi on the capacity of the fractured rock aquifers (Chilton and Foster, 1995; MacDonald et al., 2012; Smith-Carington and Chilton, 1983) report specific yield values between 0.01 and 0.05, which gives a recharge rate of between 34 and 170 mm y^{-1}.

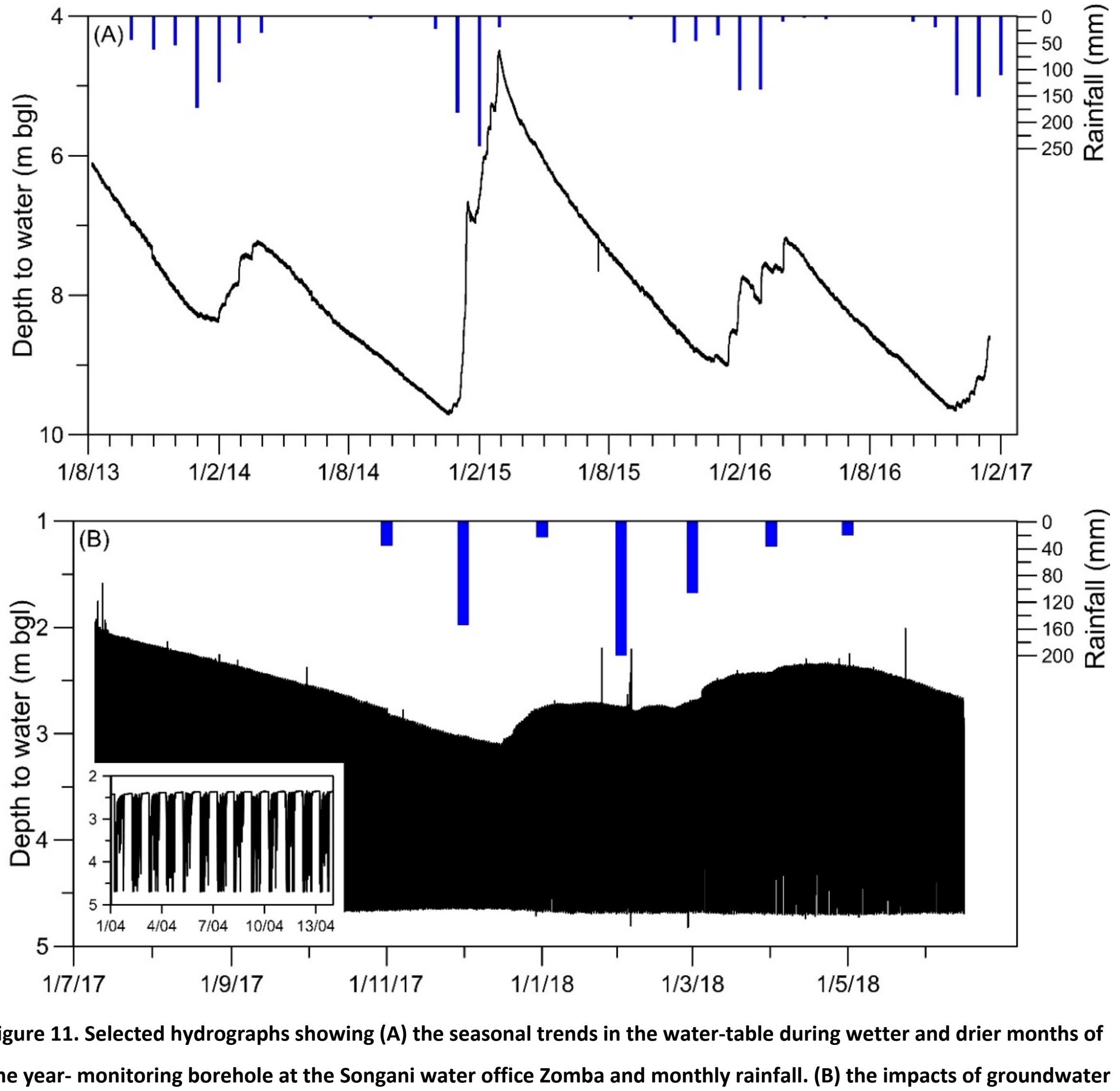

Figure 11. Selected hydrographs showing (A) the seasonal trends in the water-table during wetter and drier months of the year- monitoring borehole at the Songani water office Zomba and monthly rainfall. (B) the impacts of groundwater pumping with significant drawdown and the logger going dry- HPB MLI08, Lilongwe and monthly rainfall. Inset figure shows 1 week of data from MLI08 and the water level drawdown and recovery in response to regular daily pumping.

6 DISCUSSION

Environmental tracer and groundwater level data from three contrasting countries in sub-Sahara Africa, including the mountainous terrain of Ethiopia, the predominantly wet climate and weathered crystalline basement of Uganda and the dry subhumid crystalline basement of Malawi, provide valuable information on the groundwater flow systems that support HPB water supply. CFCs, SF_6, chloride and time series water level data provide estimates of groundwater recharge and sustainability. The stable isotope data provides an indication of recharge pathways and processes, whilst the anthropogenic tracers (e.g. E. coli and nitrate) are helpful water quality indicators that assist with the assessment of aquifer vulnerability and the quality of borehole construction.

Measurable concentrations of CFCs (apparent groundwater age less than ~ 60 years old) were found in the majority of the HPBs in each country. The presence of CFCs indicates active circulation of young groundwater in the major aquifers where the HPBs were installed. This is up to 74 m below ground level in Ethiopia, 68 m in Uganda and 48 m in Malawi (reported as mid-screen depth below ground level). Our data show mean residence times in the order of 20-50 years, which is comparable to the findings of Lapworth et al. (2013) in West Africa. The large volume of young groundwater in both the sedimentary and basement rock aquifers suggests resilience to climate variability, and therefore, mitigating the effects of reduced rainfall and recharge during short-term periods of drought. However, the findings do not imply that future climate change is not a problem.

Several HPBs have CFC or SF_6 concentrations greater than those that can be explained by equilibration with the atmospheric source. Of the 140 samples collected across the three countries, nine samples (6.4 %) have apparent contamination of CFC-12 (values more than

10 % above maximum concentrations that can be explained by equilibration with the atmosphere) sixteen samples (11.4 %) have apparent contamination with CFC-11, and fifteen samples (11.1 %) have apparent contamination of SF_6 (Uganda only). CFC-11 contamination could be a result of disused aerosol cans and CFC-12 contamination could be due to old refrigerators and air conditioner units from vehicles. For SF_6, excess air cannot explain the measured concentrations alone. A typical value for excess air of 3 cm^3 kg^{-1} results in enrichment of only 50 % above atmospheric equilibrium, whilst a value of 10 cm^3 kg^{-1} results in up to 170 % and cannot explain the high concentrations of 13 samples in Uganda. Local sources of SF_6 are less likely, but elevated concentrations can occur due to in situ production, in particular rock types and geology (Harnisch et al., 2000; Koh et al., 2007; von Rohden et al., 2010).

SF_6 contamination can arise from drilling of boreholes or from borehole development, when compressed air is used during this process (Poulsen et al., 2020), which could be a possibility based on drilling practices in Ethiopia. Unlike traditional excess air (which is incorporated in groundwater during natural recharge processes; e.g. Heaton and Vogel (1981)), air contamination during drilling or borehole construction could result in elevated gas concentrations in groundwater, which is otherwise old. Incorporation of 10 cm^3 kg^{-1} of contaminated air can cause groundwater, which is otherwise very old (with zero SF_6 concentrations) to have SF_6 concentrations of more than 500 fg kg^{-1}. Pumping over a long period of time should remove contamination, although we did not find a relationship between the age of the HPB (i.e. when it was installed) and the extent of SF_6 contamination.

E. coli was detected (>1 per MPN 100 mL^{-1}) on average in 50 % of these sites with notable variations between the countries (Ethiopia = 38 %, Uganda = 65 % and Malawi = 47 %).

Nitrate concentrations elevated above expected background (5 mg L^{-1}) were also found in 28.5 % of the sites. Correlation between nitrate concentration and CFC-12 concentration or depth are weak or non-existent in all three countries (a similar result is found for the correlation between E. coli counts and CFC-12 concentration or depth). Significant population growth has occurred across all three countries and there has been an increase in the use of pit latrines (Nakagiri et al., 2016), and so a relationship between groundwater mean residence times, and nitrate concentration might be expected (Ouedraogo and Vanclooster, 2016). Due to a half-life of 3 to 4 months for E. coli within most groundwater systems (Edberg et al., 2000; John and Rose, 2005), E. coli would not be expected to be found in groundwater with residence times greater than a year. A positive correlation between CFC-12 and E. coli count might therefore also be expected but is not seen in our data. There was also little to no correlation between nitrate concentration and E. coli count in all three countries. The relationships between nitrate concentration, E. coli counts, and CFC-12 correlation are best explained by contamination of groundwater at the well-head and the water protection area around the well-head (Ferrer et al., 2020). Contamination at the well-head may also contribute to CFC contamination, which was evident in the lack of a clear trend in the CFC concentration depth profiles. Any detectable count of E. coli or TTCs in water directly intended for drinking is considered a risk to human consumption by the World Health Organisation Water Quality Guidelines (WHO, 2017). There was a higher presence of E. coli (50 %) in the sampled boreholes than the presence of TTC measured from a larger survey across the 3 countries (Lapworth et al., 2020), however, of the sites that had E. coli present, only 18 % were medium (>10 MPN 100 mL^{-1}) and 2 % were high (>100 MPN 100 mL^{-1}) risk (WHO, 1997). It is recognised that in most untreated rural water supplies, especially in developing countries where onsite sanitation is common, faecal contamination is likely, and procedures need to be put in place to mitigate the source or treat the water appropriately. In the global review of faecal contamination of drinking-water in low- and middle-income countries by Bain et al. (2014), faecal indicator bacteria were used to evaluate the proportion of samples with detectable (>1 per 100 mL) and high concentrations (>100 per 100 mL) along with other sanitary risk score indicators to assess water from improved supply sources. Previous studies suggest that during the construction of boreholes and the completion of the sanitary seals (surface casing to prevent surface water entering bores) there is a lack of technical supervision and poor drilling completion practices, which may contribute to subsequent well-head contamination issues (Fisher et al., 2015; Foster, 2013; Harvey, 2004; Kalin et al., 2019). This could help explain why 50 % of the HPBs showed data of anthropogenic contamination (E. coli) at the well-head, which has been identified in studies of urban groundwater sources (Lapworth et al., 2017; Sorensen et al., 2015).

Most of the monitored HPBs that were actively pumped recovered to pre-pumped conditions during the night, or early the following day. Median daily water level drawdown in water levels on HPBs that were regularly pumped for community water supply range from 0.8 m to 10.5 m, and the 90th percentile ranges from 0.52 m to more than 14.2 m. Comparison of the 90th percentile of daily drawdown with the total available drawdown provides an indication of the ability of the borehole to supply the current water demand (here termed *borehole reliability*). Although we were unable to collect reliable continuous long-term water level data from a sufficient number of boreholes to draw any regional conclusions, our small data set show that a number of boreholes have a borehole reliability value greater than 0.8. This would indicate a susceptibility to drought periods when demand increases. If a larger dataset could be obtained, then we believe that this would provide useful information on reliability of the HPBs to supply water demand. Increasing the borehole completion depth does not necessarily increase water supply as there is a general decrease in hydraulic conductivity with depth within the aquifers at least in the crystalline basement aquifers of Uganda and Malawi (MacDonald et al., 2012). In this case, an increase in the number of individual community boreholes or multiple sources would be required to increase the resilience and reliability of supply (McAllister, 2020).

Comparison of the different methods to estimate groundwater recharge (chloride mass balance, groundwater age using CFCs and the water-table fluctuation method) provided recharge estimates that ranged from 30–327, 27–107 and 30–170 mm y^{-1}, for Ethiopia, Uganda and Malawi, respectively. Obtaining accurate estimates of recharge in these environments is difficult. Analysis of borehole hydrographs is a widely used technique (Healy and Cook, 2002), although specific yield is difficult to estimate in hard rock aquifers (Cook, 2003) and pumping of most boreholes means that short-term responses to rainfall events are not clearly discernible. One of the approaches that we have used has involved analysis of groundwater age – depth profiles, this method is also difficult in hard rock areas where groundwater flow is dominated by fractures (Cook et al., 2005). Available data from Ethiopia, Uganda and Malawi did not show clear trends in groundwater age with depth. This is probably related to heterogeneity with the aquifer systems but could also be influenced by problems associated with borehole construction (e.g. silting of borehole screens or casing failure). For these reasons, recharge estimates obtained using the chloride mass balance approach (Ethiopia: 327 mm y^{-1} (IQR = 308 to 336 mm y^{-1}), Uganda: 112 mm y^{-1} (IQR = 104 to 117 mm y^{-1} and Malawi: 86 mm y^{-1} (IQR = 75 to 87 mm y^{-1}), are probably the most reliable, despite uncertainty associated with loss of chloride in surface runoff (Wood, 1999). While our sampling covered a number of different regions within each country, these figures should not be considered representative, and further work would be required to extrapolate the data across larger areas.

A key finding of the study by Lapworth et al. (2013), who used environmental tracers to investigate residence times of shallow groundwater in West Africa, was the resilience of rural groundwater resources to short-term inter-annual variation in rainfall and recharge, which appear to sustain diffuse, low volume abstraction. The resilience of groundwater to drought was also supported by the recent studies by MacDonald et al. (2019) and MacAllister et al. (2020) in the Ethiopian Highlands who identified HPBs were more reliable than hand dug wells and springs.

Our stable isotopes data also indicate rapid infiltration of rainfall during recharge with little evidence of evaporation. The isotope data also indicate that recharge is associated with higher rainfall months and more intense events (Jasechko and Taylor, 2015). Our study suggests that although recharge rates are adequate, meaning that aquifers represent a reliable and resilient water resource, issues associated with borehole construction can present problems for both water quality and water quantity. In particular, evidence of well-head contamination of the shallow HPBs, probably caused by poor borehole construction, raises health concerns (e.g. infant methemoglobinemia- high nitrate or E. coli as an indicator of the potential presence of disease-causing organisms). These issues can be easily remedied by a greater focus on borehole design and construction.

7 CONCLUSIONS

Our study used a combination of different hydrogeological and environmental tracer methods to evaluate groundwater residence times, recharge mechanisms, and borehole susceptibility to anthropogenic contaminants in shallow groundwater aquifer systems in Ethiopia, Uganda and Malawi. Shallow aquifer systems are widely developed for community water supplies using hand pump boreholes. Despite the different rainfall, geology, climatic conditions and type of aquifer systems evaluated in each country investigated, there were some common findings. Environmental tracers; CFCs and SF_6 showed that recharge to aquifers was rapid and that the minimum depth of active groundwater circulation (defined by mean residence time <60 years) was relatively deep (48-74 m). Comparison of the stable isotopes of water in rainfall and groundwater samples reveal that there is little evaporation prior to recharge, and recharge events are biased to months with greater rainfall and more intense and heavier rainfall events.

Long-term recharge estimates based on the chloride mass balance for Ethiopia, Uganda and Malawi was 327 mm y^{-1} (IQR = 308 to 336 mm y^{-1}), 112 mm y^{-1} (IQR = 104 to 117 mm y^{-1} and 86 mm y^{-1} (IQR = 75 to 87 mm y^{-1}), respectively. Whilst the water table fluctuation method provided recharge estimates that ranged between 32–160, 27–80, 34–170 mm y^{-1}, respectively. In comparison, the minimum recharge estimated using CFCs were between 20–70 mm y^{-1} for Ethiopia, 40–100 mm y^{-1} for Uganda, and 30–70 mm y^{-1} for Malawi, for field sites in each of the three countries.

The resilience of the aquifers to ensure sufficient water supply and water of good quality has advantages over surface water sources, however, the results from this study indicated that recharge processes are rapid and there is an inherent risk of contamination from anthropogenic pollution at the ground surface, which is associated with the borehole construction (e.g. well-head and sanitary seal failure). Improved borehole construction would help to mitigate risk of contamination from anthropogenic pollution.

Groundwater monitoring is essential to evaluate the ability of aquifers to meet domestic and agricultural demand and to provide resilience in the face of climate variability. To facilitate monitoring, HPBs need to be designed to accommodate the installation of water level dataloggers. Alternatively, sensor techniques are emerging, which may better facilitate widespread adoption of groundwater monitoring (e.g. Thomson et al., 2012). In many environments monitoring in abandoned boreholes reconstituted as dedicated unpumped monitoring boreholes may be the most feasible option for widespread, and rapid initiation of groundwater monitoring.

Whilst the data suggests that shallow HPBs can support a small number of households, groundwater for other purposes of a greater supply volume such as town water supplies or for irrigation development requires a greater investment in aquifer characterisation, borehole siting and construction techniques.

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Supplementary Material

Further information to the study sites, the collected samples and laboratory analysis techniques is contained in the Supplementary Material.

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