Hydrological Processes

Isotope hydrology and water sources in a heavily urbanised stream

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Abstract

Complex networks of both natural and engineered flow paths control the hydrology of streams in major cities through spatio-temporal variations in connection and disconnection of water sources. We used spatially extensive and temporally intensive sampling of water stable isotopes to disentangle the hydrological sources of the heavily urbanized Panke catchment (≈ 220 km²) in the north of Berlin, Germany. The isotopic data enabled us to partition stream water sources across the catchment using a Bayesian mixing analysis. The upper part of the catchment streamflow here is dominated by groundwater from gravel aquifers underlying surrounding agricultural land. In dry summer periods, streamflow becomes intermittent; possibly as a result of local groundwater abstractions. Urban storm drainage is also an important part of runoff generation, dominating the responses to precipitation events. Although this dramatically changes the isotopic composition of the stream, it only accounts for 10-15% of annual streamflow. Moving downstream, subtle changes in sources and isotope signatures occur as catchment characteristic vary and the stream is affected by different tributary inflows. However, effluent from a wastewater treatment plant (WWTP) serving 700,000 people dominates the stream in the lower catchment where urbanisation effects are more dramatic. The associated increase in sealed surfaces downstream also reduces the relative contribution of groundwater to streamflow. The volume and isotopic composition of storm runoff is again dominated by urban drainage. As a result, only about 10% of annual runoff in the lower catchment comes from urban storm drains. The study shows the potential of stable water isotopes as inexpensive tracers in

urban catchments that can provide a more integrated understanding of the complex hydrology of major cities. This offers an important evidence base for guiding the plans to develop and re-develop urban catchments to protect, restore and enhance the ecological and amenity value of these important resources.

Keywords: Isotopes, urban hydrology, ecohydrology, wastewater, end member mixing analysis

1. INTRODUCTION

. Introduction

With over 50% of the world’s and 70% of Europe's population now living in cities, many key global challenges revolve about the sustainable management of urban water (United Nations et al., 2019). This is likely to lead to different priorities for urban water management; with various stakeholders, such as water supply and sewage disposal agencies, industrial users and local citizens having competing demands that local governance agencies have to mediate to maintain the quantity and quality of urban water bodies (Brears, 2016). However, quantitative understanding of the complex sources of water and flow paths that sustain urban water bodies is often lacking compared to other environments. Urban streams and other water bodies are variously used as sources of water supply and a means of drainage and disposal of effluents (Gücker et al., 2006; House et al., 1993; Paul & Meyer, 2001); as well as being perceived as a potential hazard in terms of flood risk and pollution from effluents (Kundzewicz et al., 2014). Consequently, urban water systems are usually heavily managed with a range of complex infrastructure to control abstractions, stormwater drainage and effluent disposal. In older cities, urban water has often been subject to an evolutionary history over centuries of ever-changing management decisions as societal needs and priorities have varied (Hassan, 2011; Winiwarter et al., 2016).

The inevitable decrease in catchment permeability as build-up areas expand leads to higher surface runoff in urban streams, mostly routed via stormwater drains and combined sewers, reducing net-infiltration and therefore groundwater recharge (Arnold & Gibbons, 1996), mobilizing pollutants on roads and other urban surfaces (Brinkmann, 1985). This often increases connectivity with untreated wastewater in combined sewers that leads to episodic pollution from organic waste and pharmaceuticals (Klein et al., 2015; Komínková et al., 2016; Launay et al., 2016). However, the exact sources of pollutants, from either combined sewers or wastewater treatment plants (WWTPs) can be difficult to identify (Lee et al., 2010). In addition, installation of general urban infrastructure also includes other less obvious zones of subsurface connectivity via trenches carrying utility cables and pipelines, giving analogies to natural dual-flow hydrological systems and use of the term “urban karst” (Bonneau et al., 2017). Urban catchments can thus be conceptualised as a complex “spiders web” of highly connected water sources, but also more disconnected areas in often extensive areas of urban green space (e.g. parks, gardens, urban forests, urban wetlands etc.). Understanding the integrated interaction between different components of the technical management system and natural flow paths in urban green space is fundamental to understanding urban hydrology in an holistic way (Gessner et al., 2014).

Improving hydrological process understanding in urban areas requires integrating tools that provide insight into both large- and small-scale spatio-temporal variability in catchment function. In this regard, stable isotopes offer outstanding potential as natural tracers in urban hydrology (Ehleringer et al., 2016). The use of stable isotopes ratios of $^{2}H/^{1}H$ and $^{18}O/^{16}O$ within the water molecule has been applied in many investigations to trace precipitation through different types of hydrological systems and at different scales to understand flow paths and the mixing dynamics of precipitation with water already stored in the catchment (Birkel et al., 2011; C. Soulsby, Birkel, Geris, Dick, et al., 2015; C. Soulsby et al., 2011). Although urban studies are notably underrepresented in the isotope hydrology literature, this is rapidly changing. Recent studies have used isotopes to assess how urbanisation affects the age distribution and travel times of runoff (Dimitrova−Petrova et al., 2019; Grande et al., 2020; Morales & Oswald, 2020; C. Soulsby, Birkel, Geris, & Tetzlaff, 2015; Chris Soulsby et al., 2014) and the dynamic influence of different water sources on the urban hydrograph (Jefferson et al., 2015; Pellerin et al., 2008). Additionally, Jefferson et al., (2015) used stable isotopes to investigate stormwater control measures and to assess their effects on event contributions.

The composition of tracers in streams and potential source waters can be used to separate the hydrograph into relative contributions from different sources with contrasting tracer characteristics (e.g. recent rainfall, groundwater and others). This was formalised in end member mixing analysis (EMMA) (Christophersen & Hooper, 1992) which, alongside other means of hydrograph separation, have proved useful tools in isotope hydrology that have been widely used ((He et al., 2020; Klaus & McDonnell, 2013). Preliminary studies have shown potential for source apportionment in urban areas: with tracers variously being used to disentangle tap water sources on a national scale (Bowen et al., 2007; West et al., 2014), or locally within a state or city (Jameel et al., 2016; Sánchez-Murillo et al., 2020; Tipple et al., 2017), and provided a viable method for waterworks to understand their distribution system (Jameel et al., 2016). Furthermore, Houhou et al., (2010) and Kracht (2007) used distinct stable isotope signatures to identify sources within wastewater sewers, while Grimmeisen et al. (2017) used isotopes to understand groundwater contamination due to leaking sewers. Still, how the wider urban hydrological cycle is affected by integration of natural runoff sources, urban drainage and treated effluents is rarely investigated quantitatively through tracers (Follstad Shah et al., 2019; Kuhlemann et al., 2021a; Torres-Martínez et al., 2020).

Similarly, estimating metrics of water ages, such as mean transit times, has proven insightful in isotope hydrology as a tool for assessing flow paths and mixing interactions in catchments. This is based on using the damping and lagging of the precipitation isotope time series in the rainfall-runoff transformation with lumped convolution integral models (McGuire & McDonnell, 2006; Tetzlaff et al., 2018), ensemble hydrograph separation (James W. Kirchner, 2019) or more sophisticated tracer-aided hydrological models that track water and solute fluxes and their associated ages (Birkel and Soulsby, 2015; Douinot et al., 2019; Kuppel et al., 2018). Urban streams can integrate very young waters (<1 day old) as rainfall is routed via storm drains in rainfall events (Soulsby et al., 2015), together with much older water (>decades) that recharges groundwater through urban green spaces (Gillefalk et al., 2021;Kuhlemann et al., 2021b; Nouri et al., 2019). However, in urban areas where significant volumes of effluents are introduced into streams, there are conceptual difficulties in defining water ages, especially if wastewaters are derived from local sources and have similar isotopic signatures (Kuhlemann et al., 2021a). In such cases, assessing the influence of recent rainfall in streamflow is possible by estimating the contribution of the young water fraction (YWF) to runoff (J. W. Kirchner, 2016b, 2016a). This is a simple method for quantifying the quick flow response of catchments based on the YWF, which is the contribution of water less than ≈2 months old to the stream hydrograph. The method provides only a relatively coarse metric of complex age distributions, though it gives insight into the dynamics of catchment runoff responses and provides an index for inter-comparison studies (von Freyberg et al., 2018).

The motivation for this paper is to apply isotopic methods in a complex, heavily urbanized catchment to understand the spatio-temporal dynamics of water sources contributing to streamflow. For this, we focus on the Panke catchment in Berlin, the capital city of Germany. The catchment has a long and ongoing history of urban development and a highly manipulated water management system. However, the way in which this interacts with undeveloped areas in the catchment is poorly understood. Also, although the catchment is well-monitored hydrometrically and most effluent discharges are known, complex groundwater-surface water interactions affect the catchment water balance in a spatially variable way. Thus, tracers offer a means to disentangle effects of natural discharge, storm sewers and effluents. To do this, we collected daily precipitation and stream samples over 15 months, in conjunction with seasonal, spatially distributed synoptic sample surveys. This provided the data to achieve the following specific aims:

1. To characterise the short-term hydrological dynamics of outflow from the Panke catchment and its isotopic composition in relation to time-variant sources of streamflow.
2. To characterise the spatial variation in the isotopic composition of the stream network in relation to the dominant sources of streamflow.
3. To assess how the temporal controls on runoff generation vary spatially at the catchment scale.

The study and results also highlights more general insights into the opportunities and challenges for using isotopes in urban hydrological studies. In addition, understanding the origins of urban streamflow regimes has important implications for restoration management of such heavily modified urban systems which we discuss.

2. Study site

The Panke catchment (220 km^2; Figure 1) is located in the State of Brandenburg and Berlin in northeast Germany and forms the dominant surface water drainage of the northern part of the city of Berlin. Much of the catchment is urban (Table 1). The Panke drains a fairly flat area that naturally ranges from 35 - 90 m a.s.l. with an average slope of 1.8%. Climatic conditions reflect both maritime and continental influences: the average annual precipitation is ~ 590 mm and the mean annual temperature is 9.5°C (1981 - 2010) (DWD & (Deutscher Wetterdienst), 2020). Rainfall is fairly evenly distributed between winter and summer, though the winter is dominated by longer low-intensity frontal rain, whilst summer experiences more high intensity, convectional storms. The region is drought-sensitive and in 2018, Berlin only received ≈ 420 mm of rainfall. During 2019 and 2020, annual precipitation was 589 mm and 513 mm, respectively (DWD, 2021).

The Panke is an effluent-impacted tributary of the River Spree, which flows into the River Havel downstream of the city (Kuhlemann et al., 2020). The river morphology class according to the German Water Framework Directive is between 5 and 7 (where 1 is least impacts and 7 is most heavily impacted) (Senate Department for Urban Development and the Environment, 2012). The Panke originates in the north and flows ≈ 30km in a south-westerly direction to the Spree. The catchment's headwaters are located on the northern edge of the Warsaw-Berlin glacial spillway which drained from Poland to the River Elbe (Figure 1b). The geology consists of >100m of Quaternary deposits (Limberg & Thierbach, 2002). These form a series of aquifers in Berlin and the surrounding area; the aquifer terminology used is the same as Limberg and Thierbach (2002). For the Panke catchment, two main aquifer systems form the shallow aquifer (AQ1) (Figure 2). AQ1.1 is the sub-aquifer in the Barnim plateau (in the East) which is partially confined by an overlying ground moraine. The main shallow "Panke aquifer" (AQ1.2), dominates the main river valley and is unconfined and characterised by sands and gravels above an aquitard of glacial till (Figure 2). The main aquifer beneath Berlin is AQ2, which is confined below the aquitard in the Panke (Limberg et al., 2007). The general direction of groundwater flow is south-west along the slope of the Barnim plateau, the main recharge area. Once the main glacial valley is reached, the groundwater flow is oriented to the South (Senate Department for Urban Development and Housing, 2019a). Berlin and Brandenburg's aquifers have been investigated using long-residence time tracers such as tritium and helium, showing decadal to centuries old water dominating the upper storage of unconfined aquifers, whilst deeper waters could be millennial (Bednorz & Brose, 2017; Gudrun Massmann et al., 2009).

The north of the catchment at has around 30% urban cover (Table 1) but is unaffected by large effluent discharges. Typical for such lowland areas in northern Germany, streamflow generation is primarily groundwater dominated (Smith et al., 2021) with seasonally varying inflows from headwater tributaries with non-urban (forested and agricultural) land use (Figure 1a). During our investigation, the stream was observed to emerge from a managed urban-wetland and lake. Despite this, flows can be intermittent in the upper reaches of the stream network during the summer which might reflect seasonal variation in storage and effects of local groundwater abstractions for irrigation (Jasechko et al., 2021; Kleine et al., 2021).

Within the lower catchment, the more densely urbanized area is characterised by increasing densities of roads and stormwater drains that discharge during rainfall events (Figure 1a and Table 1). Around 26.5 km^2 of the Panke catchment is connected to Berlin's rainwater drainage system; this includes 13.6 km^2 of sealed surface (Senate Department for Urban Development and Housing, 2018). The stormwater overflows (SWOs) of Berlin result in estimated ≈ 3.1 Million m^3/y rainwater discharge as direct runoff into the Panke (Senate Department for Urban Development and Housing, 2019b). Some is routed by combined sewer systems (17.8%), with the remainder mostly having standard separations between wastewater and stormwater (Senate Department for Urban Development and Housing, 2018). In the South of the Panke, combined stormwater overflow dominates the drainage infrastructure (Möller & Burgschweiger, 2008). Sewer runoff is partially influenced by reverse gradients which are controlled by a discharge threshold and only activate in larger storms. Mixed, untreated wastewater with storm runoff can also be discharged into the Panke from a pumping station close to the WWTP (Figure 1).

In the downstream half of the Panke, the stream is increasingly regulated, and flow control structures can divert water into and out of the catchment (Figure 1). WWTP effluents can be either discharged directly into the Panke, or transported out of the catchment via the Nordgraben. The WWTP serves a population of ≈ 700,000 with a dry weather discharge capacity of 105,000 m^3/d (Möller & Burgschweiger, 2008). About 86,400 m^3/d (mean 1 m^3/s, from 0.83 m^3/s up to a maximum of 2.7 m^3/s (Kade, 2020)) of the treated wastewater are directly discharged into the Panke, the rest is drained into the Nordgraben and is usually transferred to the neighbouring Tegeler catchment (Figure 1). A proportion of peak flows can also be diverted out of the catchment via the Nordgraben to reduce flood risk. The weirs that regulate flows are not automated but are manually controlled, most notably in advance to heavy rainfall events, depending on the forecast of a flood risk model (Kade, 2020). Other weir operations were observed during the study period to alter the input of treated effluents to enhance baseflows in the Panke. A small proportion of treated effluents are also discharged for maintaining a former sewage-irrigation farm which is now a wetland and forested area on the north-west side of the catchment, which is drained by forested stream just upstream of the WWTP (Figure 2c) (INKA BB, 2014; Kade, 2020; Lischeid et al., 2015).

The Panke stream is morphologically altered along its length, though in some places limited restoration has been proposed and undertaken (Lange et al., 2015; Wasser- und Bodenverband „Finowfließ“, 2011). The last three kilometres of stream length are heavily canalised, with steel piling and almost no visible hyporheic zone (Senate Department for the Environment, Transport and Climate Protection & (SenUVK), 2019). The latter might limit groundwater – surface water exchange processes as described in (Lewandowski et al., 2019).

3. Data and Methods

The German Weather Service (DWD) climate station in the catchment was used for precipitation and temperature data (Figure 1). Daily precipitation samples for isotope analyses were collected at the Urban Ecohydrological Observatory at Steglitz ≈ 10km south of the catchment where continuous precipitation isotope samples have been collected since the beginning of 2019 (Kuhlemann et al., 2021a). Sample collected from Buch in the Panke catchment in summer 2020 were very similar to those from Steglitz which we use here for the longer time series. Samples were protected against evaporation with a ≈ 3mm Paraffin layer (IAEA/GNIP, 2014).

Stream discharge and water level data for sites on the Panke (Figure 1b) were provided by the *Senate Department for the Environment, Transport and Climate Protection* (Senate Department for the Environment, Transport and Climate Protection & (SenUVK), 2021b) in 15min intervals (Senate Department for the Environment, Transport and Climate Protection & (SenUVK), 2021a). Daily WWTP volumes draining into either the Nordgraben or Panke were provided by the Berlin Water Works (*Berliner Wasserbetriebe*, BWB) and their subcontractor (*Umweltvorhaben-Berlin Brandenburg,* U-BB) (BWB, 2021; Kade, 2020). Daily stable isotope samples were collected from the catchment outlet (OL) near the most downstream gauging station (Figure 2a). Gaps occurred the end of December 2019 and due to a reduced sampling frequency (2-3 times weekly) during COVID19 lockdowns (Mid of March – End of April 2020).

At six locations along the Panke (Figure 2a), grab samples of stream water for stable isotope analysis were taken from October 2019 to December 2020. Initially sampler were collected monthly and for a few months and thereafter every two weeks. Three sites (UP1, UP2, UP3) were upstream of the WWTP inflow, one was at the WWTP discharge point (WWTP), one site downstream (DS) and one at the catchment outlet (OL) (Figure 2a-c). At all locations, the fortnightly sampling captured a diverse range of hydroclimatic conditions and discharge levels. In addition, four seasonal synoptic surveys (October and December 2019, April and July 2020) were undertaken along the Panke, including its major tributaries, encompassing 30 sampling locations, to investigate the isotopic transformation and seasonality within the stream and their tributaries.

Groundwater was sampled for isotope analysis on a monthly basis from January – October 2020 (except for COVID19 gaps in April) from seven wells across the Panke catchment capturing different shallow aquifer systems within AQ1 and AQ2 (Figure 2). We purged the wells through pumping for

30 - 90 min, to ensure that at least double the exchange volume was removed and water quality determinants such as pH, electric conductivity and oxygen concentration were measured until they stabilized using a WTW Multi probe 3630.

All isotope samples were decanted and filtered (0.2 μm cellulose acetate) into 1.5mL vials in the field and refrigerated until laboratory analysis. They were analysed for water stable isotopes ($\delta^{18}O$ and $\delta^{2}H$) by Cavity Ring-Down Spectroscopy with a L2130-I Isotopic Water Analyser (precision: ± 0.025$\delta^{18}O$ and ± 0.1 $\delta^{2}H$, (Picarro, Inc., Santa Clara, USA, 2020)). Isotope values are described in delta-notation using four standards and reference to Vienna Standard Mean Ocean Water (VSMOW) from the International Atomic Energy Agency (IAEA) for calibration. Data correction was performed by the "Chem Correct" software from Picarro to identify potential organic contaminants (Picarro, Inc.,Santa Clara, USA, 2018).

For data processing and analyses, R (Version R version 4.0.3 "Bunny-Wunnies Freak Out" (2020-10-10)) was used. All isotope samples were referenced to the deviation of the Local Meteorological Water Line (LMWL) for Berlin (Kuhlemann et al., 2021a) as line-conditioned excess (lc-excess) as described by (Landwehr & Coplen., 2006):

$$LMWL: \delta^{2} H = 7.76\, \delta^{18} O + 5.66$$

$$lc-excess = \delta^{2} H - 7.76\, \delta^{18} O - 5.66$$

To identify different streamflow sources, we applied the Bayesian EMMA using MixSIAR (version 3.1.12) for the different stream sites along the Panke. MixSIAR is an open-source Bayesian model for R, using a Gibbs sampler, allowing the usage of prior distributions. For calculation, a Markov Chain Monte Carlo (MCMC) method was used for estimation of probability density functions (B. C. Stock et al., 2018; B. Stock & Semmens, 2016). As tracers, we used $\delta^{2}H$, $\delta^{18}O$ as well as lc-excess to characterise different potential streamflow components. Although the lc-excess is dependent on both stable water isotopes, a particularly marked and useful negative lc-excess signal was introduced by the

WWTP as a source, while groundwater generally only had positive or close to zero values. We separated the data set for each site into seasonal data (Winter: December – February; Spring: March – May; Summer: June – August; Autumn: September – November, Northern Hemisphere) categories for the analysis. We assumed that open water fractionation within the stream was negligible and that the tracers behaved conservatively along the channel. For the outlet, the complete dataset (biweekly and daily data) was used as end members. The end member mixing analysis provides two internal statistics to evaluate model performance. The Gelman-Rubin-test must be > 1 for calculating the chain, below 1.1 is acceptable and ≈ 1 is for a convergent model (Gelman et al., 2014; B. Stock & Semmens, 2016). The Geweke test is a two-sided z-test, high z-scores give a basis for model rejection (B. C. Stock et al., 2018; B. Stock & Semmens, 2016) (Details in Appendix).

In the mixing analysis, the stream was considered a potential mix of groundwater, recent precipitation (routed by storm drains), wastewater effluent (where present) and any streamwater inflow from upstream. This means that the regularly sampled stream sites (UP1to 3), and DS (except WWTP) were also used as end members in MixSIAR for sites downstream. For groundwater, AQ1.1 (Barnim aquifer) and AQ1.2 (Panke aquifer) were kept separate due to the potential higher intra-annual variability of the unconfined AQ1.1. The WWTP was only applied as a source for the DS and OL sites. Standard deviations for the different sources were calculated to assess the variability of each source for the given endmember.

We also estimated the young water fraction (YWF) contribution to stream flow at all sites to assess the influence of urban storm runoff. YWF is a simple but useful measure to estimate the contribution of water younger than two months to streamflow (J. W. Kirchner, 2016b). As the seasonal cycle of precipitation is damped due to storage and mixing processes, it gives insights into overall catchment function in terms of young water contributions to streamflow (J. W. Kirchner, 2016b; von Freyberg et al., 2018). A robust estimation was derived from the ratio between the sinusoidal regressions of seasonal variations in precipitation and stream isotopes via an iterative re-weighted least squares (IRLS) R script which was used to minimize the outliers. The script was provided by von Freyberg et al. (2018). We used a discharge weighting for the YWF from the OL site. As goodness-of-fit measures between the regression and observed stable isotopes we used the coefficient of determination (R^2) and the hypothesis significance testing (p-value, (Fischer, 1925)).

4. Results

4.1 Rainfall-runoff characteristics of the Panke catchment

Overall, the sampling period was characterised by relatively dry conditions. After initial rainfall in October, the winter of 2019/20 exhibited frequent, but usually low amounts (<5mm) and intensities of daily rainfall inputs, with February being the wettest month (Figure 3a). March and April were then relatively dry, but early summer was characterised by wetter conditions, particularly with some regular heavy convectional rainstorms. Late summer was again dry, though late September saw the highest daily rainfall of the year with a relatively dry early winter 2020/21 following.

Flows at the Panke outlet (OL) showed a flashy discharge response, typical of an urban catchment, to all substantial precipitation events (Figure 3b). Such abrupt, transient increases in flow were followed by rapid recessions once rainfall stopped. Similarly abrupt, but more persistent changes in discharge were related to the weir operations, causing increases and decreases in baseflows which could range from 0.3 to 1.4m³/s. From higher flows in early October 2019 until the beginning of February 2020, discharge generally decreased from baseflows of ≈ 1 m³/s to ≈ 0.5 m³/s. After a wet February 2020, flows recovered to 0.8 – 1 m³/s and declined again during a very dry April. The dry weather sub-daily flow variation evident during these conditions showed the effect of diurnal changes in WWTP effluents. This was followed by a step change in flows where the volume of wastewater effluent flowing into the river was increased during the drier summer via weir operations to enhance baseflows. Conversely, during wetter periods in October 2019 and 2020 flows were diverted out of the catchment into the Nordgraben to reduce flood risk.

Flows in the upper catchment were measured at UP2 and were unaffected by WWTP discharge, but showed the characteristic seasonality of a groundwater-dominated stream with higher winter baseflows (Figure 3c). However, the stream was responsive to storm events even after prolonged antecedent baseflow condition in summer 2020. Between UP2 and UP3, a water level gauge (not shown) followed the general dynamics of UP2, though comparison of long-term flow data between the two sites suggest losing conditions during summer (Zeilfelder (Berlin SUVK), 2021, pers.

communication). Flows from the WWTP enter between the UP3 and DS sampling points (Figures 2c and 3f) and flows at DS were strongly influenced by weir operations (Figure 3d). The WWTP effluents also showed diurnal variations and other changes which were evident at OL (Figure 3b). While runoff peaks generated by urban storm drains in the lower catchment were also evident in comparing OL and DS (Figure 3b and d), as proportion of runoff peaks was transferred out of the catchment between these two points via weirs at the Nordgraben (Figure 3f).

In Figure 3e, daily groundwater levels are shown for selected wells in the Panke catchment that were also sampled for isotopes. In the partly confined AQ(1.1) aquifer, the water table is ≈4-5m below the ground surface, and around 2-3m in the unconfined AQ(1.2). Artesian conditions prevail in AQ(2) below the confining later. After the dry periods of 2018 and 2019, groundwater levels increased by around 0.1m until March 2020 for AQ(1.1), and by ≈0.25-1m in AQ(1.2). Only small differences in the synchronicity of seasonal water level variations were observed, though the AQ(2) aquifer responded later to recharge. During the summer period after the dry April, water levels fell until September, where they stabilized, except in AQ1.1, which had lower levels by about 0.1m compared to the year before (Senate Department for Urban Development and Housing, 2010).

4.2 Isotope dynamics in the Panke catchment

Stable isotopes in precipitation showed a high variability; as expected, there was pronounced seasonality with samples enriched in heavy isotopes during summer and depleted during winter months (Table 2). However, day-to-day variability could be high in both seasons (Figure 3a). The daily stream samples at the catchment outlet showed that the seasonal variation of the inputs was greatly damped, but the rainfall signal was translated to the stream during storm events, with the effect more pronounced in the larger events (Figure 3b). Consequently, the seasonal variations in rainfall were also evident in the stream, with more enriched anomalies in the summer and depleted anomalies in winter. Usually, the rainfall signal only remained apparent in the stream for a day, but in the case of larger events, the effect could persist over several following days.

Isotope sampling at UP1 and UP2 was not always possible when the stream had dried out completely; e.g. the 26/08/2020 sample was the first possible sampling following a rain event after a prolonged dry period (Figure 3c). During this late August period, UP1 and UP2 showed the most enriched isotope values of with -57.0‰ and -50.65‰ for $\delta^{2}H$ respectively, whilst winter values reached around -64.0‰ for $\delta^{2}H$ at both sites. The sample sites UP3, WWTP and DS all showed broadly similar isotopic dynamics but UP3 was generally more depleted, WWTP was more enriched and DS showed tendencies to be intermediate between both at high flows, but was more strongly influenced by the WWTP (Figure 3d). The WWTP introduced a variable isotope signal during events probably from the mixed stormwater received in the WWTP and discharged within hours or days.

Isotopic signatures in groundwater showed some seasonal variability in AQ1.1 and AQ1.2, though this was very damped compared to precipitation or stream signatures. The isotopic composition of water in the deeper and confined AQ2 showed little change and was the most depleted. Thus, AQ1 showed a higher variability compared to AQ2, suggesting the greater influence of near-surface flow pathways, and mixing. Groundwater from the main unconfined Panke valley (AQ1.2) was quite homogeneous, except for a wetland-influenced groundwater well (the most enriched GW-well in Figure 3e and located upstream of UP3 adjacent to the outflow of a wetland/forested tributary see Figure 3f) which showed some inter-annual variability. The highest variability in isotopic signatures was measured for the Barnim aquifer (AQ 1.1), with the most enriched groundwater isotopes in March, and most depleted in July. A comparison which UP2, which is close to this particular AQ1.1 well (Figure 3f), often showed an overlapping isotope signature possibly implying surface water connectivity. Table 2 provides a summary of the stable water isotopes, as discussed above

4.3 Spatial variability in isotopes

The isotopic signatures of the different sampling sites and potential source waters showed some clear differences in ranges and deviations from the LMWL when plotted in dual-isotope space (Figure 4). Precipitation had the highest variability, and less than half of this variability was observed in the stream during events, while during baseflow conditions almost no variation occurred (Figure 4a). The relative stability of most groundwater samples was evident, plotting mostly along the LMWL, except for the previously mentioned well in AQ1.2 which received wetland drainage and plotted distinctly below with a more enriched and fractionated signal (see location in Figure 2c). Importantly, the groundwater signal was very similar to the stream signature throughout the year at UP1 and UP2 in the north. The stream became progressively more enriched between UP2 and UP3, probably caused by enriched inflows from the north bank tributary (see Figure 3f) draining forested and wetland areas (Figure 4b) where surface evaporation is likely (Kuhlemann et al., 2020; Sprenger et al., 2017). However, below the WWTP, similar isotopic signatures in the lower stream system showed a strong influence of effluent waters. The samples from downstream of the WWTP plotted parallel to the LMWL were very similar to OL (Figure 4b).

Urban inflows were defined as those streams flowing through urbanized areas, while peri-urban pass more agriculture-dominated sub-catchments (Figure 4b). Samples from the urban and peri-urban streams showed higher isotopic variation than the forested stream or the peatland inflows. As noted, the main forested stream has its confluence with the Panke about 2 km upstream of UP3, while urban headwater tributaries can be found along the whole catchment (Figure 2f). The peatland inflow is between UP2 and UP3, south of the forested stream, close to the wetland-lakes in Figure 2. The peatland inflow showed the most enriched signatures of any tributary (mean -6.7 to -51.3 for $\delta^{18}O$ and $\delta^{2}H$, respectively).

The results of the seasonal synoptic surveys are shown in Figure 5, where the regular sample sites and tributaries are highlighted. The North of the catchment (i.e. UP1 – UP3) was characterised by streamflow with more depleted isotopic signals being mostly similar to those of groundwater (see deuterium ($\delta^{2}H$) in Figure 5a). However, the most upstream synoptic sampling site flowed from a lake which became enriched during the summer. The October samples still showed evidence of such enriched water sources following the summer, but during December 2019 and April 2020, the entire northern part of the catchment exhibited depleted isotopic signatures. The July 2020 sampling showed more enriched isotopic signatures again, especially at the most upstream lake site. Further downstream, below the WWTP seasonal variability became larger, with December 2019 most depleted and October 2019 most enriched.

The spatial influence of inputs from the WWTP on the DS and OL sites are clearer for lc excess , though with some seasonal variation (Figure 5b). However, the seasonal patterns for lc-excess in the lower catchment were more complex than for δ^2H, with slightly lower values being estimated in winter, showing greater fractionation effects in the effluent waters. The lc-excess varied between ~ -4 to -4.5‰ during November-March and ~ -2.5 to -3‰ from April – October.

The spatial surveys in December, April and July revealed some of the more subtle influences of tributary streams of the main Panke, especially in the central part of the catchment where the WWTP discharge enters (Figure 5 insets). For example, throughout the year, and even in winter, the outflow of the wetland and forest stream upstream of the WWTP inflow was enriched and contributing to flows which resulted in enriched signatures and low lc-excess values compared to the mainstream. Spatially, the WWTP inflow provided an enriched signal. Overall, downstream of the WWTP signatures remained similar along the stream for all sampling occasions with more enriched and fractionated values than upstream the WWTP.

4.4 Temporal variability in streamflow sources

The end member mixing analysis helped to constrain the sources of flow in the Panke and quantify their contributions to the mainstream (Figure 6; Tables 3 and 5, see Appendix). Results show that UP1 was dominated by groundwater, with contributions from AQ1.2 and AQ1.1 accounting for around 70% of the flow for most of the year (Figure 6a). However, the overall contribution of precipitation from urban storm drains was the highest of all sample sites, being greatest in autumn and winter, though still only accounting for around 20-30% of runoff. At UP2, the groundwater contribution again had similar inflows from AQ1.1 and AQ1.2, though overall contributions from upstream (UP1) accounted for around 50% of flows (Figure 6b). These were lower in summer when the streamflow became intermittent. This leakage from the upper catchment may also explain why the modelled contribution from rainfall was also lower, as summer inflows from storm drains leak into the aquifer before reaching UP2. At UP3 (Figure 6c), a broadly similar picture was evident, though the contributions from upstream were only around 25% of flows, and AQ1.2 appeared to become the dominant source of groundwater, presumably reflecting inputs from the north bank forested tributary shown in Figure 3f. The proportions from upstream (UP2) were relatively low, especially during summer suggesting losing conditions occurred. Precipitation generally contributed 10%, though this increased in summer, in response to greater storm influence and inflows from drains.

There was an abrupt change in contributions downstream of the WWTP. The sampling point DS had a relatively constant, very high contribution of WWTP effluents accounting for around 90% of discharge (Figure 6d). Here, the seasonal variability of contributions was also low compared to the other sites, indicating the overall dominance of WWTP. The slightly higher WWTP contribution during summer compensated low flow conditions in UP3. Groundwater and precipitation each contributed around 5% and 3% to annual runoff, respectively. A similar distribution of sources was evident at the OL sampling point, with over 90% of contribution originating from DS, and with low variability (Figure 6e). The highest DS contribution of about 96% and 89% was during summer and autumn, respectively. In summer, most urban tributaries went dry and the peri-urban tributaries had low water levels, while in autumn with the longest rainfall events during the sampling periods, precipitation had the highest contribution of ~ 7%, which is related to prolonged rain over several days driving variability in the hydrograph (Figure 3b). However, in general, precipitation and groundwater both made small contributions to the stream. Although low, the groundwater contribution was also at its minimum during the summer months at OL, consistent with the seasonally minimum groundwater storage. Table 5 (see Appendix) provides the quality criteria for the EMMA provided by MixSIAR.

The YWF at each site was used as an additional indicator of water sources, by estimating the “younger water” (<2 months) contribution to streamflow (Table 4), which primarily reflects the contributions of urban storm drainage. The dynamics between $\delta^{2}H$ and $\delta^{18}O$ were similar, however, they were slightly different regarding absolute values. At almost all sites, statistically significant fits ($p<0.001$) were obtained. Despite being a heavily urbanised catchment, the YWFs were low at all sites (Table 4). Between both upstream sites UP1 and UP2, the YWF in $\delta^{2}H$ decreased from about 13% to 7%, respectively. At UP3, this increased to ~ 11%. The estimated YWF of the WWTP effluents was 7%, and ~ 10% for DS and OL, with a similar range of uncertainty. These results are broadly consistent with the rainfall estimates from the EMMA of each site.

5. Discussion

5.1 Runoff sources in the Panke catchment

Our study demonstrated that we could successfully use isotopes to identify the spatial and temporal dynamics of runoff processes in a complex, highly managed urban catchment in a major city. The lowland headwaters of the Panke river, where urbanization affects around 34% of the catchment area, still reflect groundwater dominance in streamflow generation, albeit strongly affected by urban storm runoff. This resulted in the highest relative contributions of rainfall and young water to streamflow in the catchment. These spatial changes are shown proportionately and conceptually in Figure 7 and Figure 8 respectively. Whilst tributaries from forested and wetland catchments supplement flows in the catchment headwater, limited recharge from sealed surfaces (Roy et al., 2015; Wenger et al., 2009) and unregulated local groundwater abstractions (Benejam et al., 2010; Jasechko et al., 2021; Vörösmarty & Sahagian, 2000) may result in the stream being "losing" in the summer and leaking into the underlying aquifer. Brooks, 2009 pointed out, that intermittent streams are particularly vulnerable to anthropogenic alterations.

In the lower catchment, the dominant source of runoff becomes effluent from the WWTP, and the stream can be classified as "effluent-dominated" (i.e. where more than 50% of streamflow is comprised of effluent) (Hamdhani et al., 2020) (Figure 7). The isotopic composition of wastewater carries a fractionation signal that allows its contribution to streamflow to be estimated via end member mixing. These WWTP contributions to the Panke are managed and reduced for flood risk mitigation or increased for base flow enhancement, with weirs controlling the volume and timing of transfers. Overall, however, this dominant influence of WWTP effluents dictates that even in the lower catchment, where the urbanization accounts for ~40% of land cover, storm drains are limited to providing only <10% of annual runoff and low (<10%) young water fractions, at least part of which seems also to be water routed through the WWTP. However, this lower contribution also partly reflects drainage of some peak-flows directly from the WWTP into the Nordgraben rather than the Panke.

5.2 Using isotopes in urban hydrology

Urban isotope hydrology in Berlin, or any city operating a "largely closed" water management system, is challenging due to the lack of isotopic differentiation between withdrawals and wastewater returns (G. Massmann et al., 2007). Fortunately, in Berlin, wastewater carries a strong fractionation signal (Kleine et al., 2021; Kuhlemann et al., 2021a), so it can be differentiated from local groundwater and rainfall as an end member in hydrograph separation and for estimations of the young water fractions. The isotopes provided a basis for tracking water source contributions to complement hydrometric measurements of streamflow and effluent releases as reversals in local groundwater – surface water interactions and dictate that during summer, parts of the river become losing reaches and leak into the underlying groundwater as observed in neighbouring catchments (Kleine et al., 2021; Kuhlemann et al., 2021a). This, together with weather-related transfers of water into and out of the catchment confound source attribution from hydrometric measurements alone. Undoubtedly, other geochemical or anthropogenically-introduced tracers can help further constrain urban end member assessment and identify particular sources, if needed. The wide range of emerging pollutants from pharmaceutical metabolites is particularly promising in this regard (Bradley et al., 2020).

Despite these issues, using water ages is conceptually challenging in complex urban systems compared to other catchments where the hydro-demographics of different water sources (e.g. soils, groundwater etc.) can be well-constrained (Sprenger et al., 2019). Whilst identifying young water fractions from recent rainfall or older groundwater from depleted isotope signatures or other dating tracers (CFCs, tritium and others) is possible, effluents are more problematic, especially when they are withdrawn locally. Effluents combine a range of water ages (in mixing groundwater and surface water) and then are recent as a particular type of "young water" on release.

Similarities to the discussion by Hoekstra (2019) about how to address irrigation water in terms of "blue" and "green" water fluxes are apparent. Although, being technically a "blue water" flux (Hoekstra, 2019), it is not necessary sourced within the catchment. There is also a need to address how to include water sources that are imported from beyond the catchment boundaries (Hoekstra, 2003) which have already a characteristic water age. These can include storm overflows, imports under pressured pipes from outside of a catchment, groundwater from a deeper aquifer or tap water sources from a different region. “Imported” water can also provide additional green and blue water fluxes due to irrigation and leakage of pipes and add into the water budged of a catchment. Moreover, this can also include wastewater imported water from outwith a catchment. These waters might be considered as “young” in terms of their effluent release, although its original age on abstraction can be much older, and therefore interfere with common methods to describe water ages.

5.3 Wider implications

The general characteristics of the Panke, a natural groundwater-dominated stream, but also strongly influenced by effluent discharge and urban storm drains, are typical of many other cities built in lowland areas. Future water management plans for such cities are tending towards more sustainable approaches and viewing urban “blue” water as a resource and amenity that should be protected and enhanced in terms of environmental quality. Treated wastewater, will often be a part of this resource as well, either as continuous effluent discharge in runoff, potential groundwater recharge (INKA BB, 2014) or irrigation water for urban green spaces (Nouri et al., 2019). To manage these different water sources sustainably will require new approaches to understanding the complex spatio-temporal dynamics of urban hydrology and how engineered controls of storm runoff and effluent disposal link to the hydrology and connectivity of urban green spaces. In this regard, isotope-based studies can provide an evidence base that can inform policy and decision making, in particular, through integrating management of storm runoff and groundwater-surface interactions to enhance the connectivity of green spaces and low impact developments for natural baseflow generation.

Highly managed systems like the Panke have environmental targets for minimum flows and water quality standards. However, enhancing the ecological status of such degraded streams is challenging where wastewaters provide such a high proportion of flows. In this regard, the Panke is not unlike many other streams in older cities which have a complex hydrology that has evolved over many decades (Paul & Meyer, 2001) and suffering from the “urban stream syndrome” of cumulative degradation (Walsh et al., 2005). As such, challenges in these older urban systems can offer guidance to a rapid expansion of new cities, in terms of avoiding old mistakes; as well as providing insights that can help new developments and re-development to minimise adverse environmental impacts (Fletcher et al., 2013; Miles & Band, 2015). Treated wastewater, might be able to maintain ecological and hydrologic functions not only in catchments in drier climate (Luthy et al., 2015), but also in regions which can be considered as drought sensitive and therefore vulnerable to short and long term climatic changes. To better understand urban catchment interactions such as the significance of the maintained wetlands, ecohydrological studies considering blue and green water fluxes in an integrated way might help enhancing therefore our understanding (Miles & Band, 2015).

Obvious future goals could usefully focus on enhancing permeability through the creative use of urban green space and low impact developments that add to the green infrastructure. Already in Berlin, new developments and re-developments have requirements for permeable green space as a proportion of the (re-)developed area as well as a disconnection from urban storm drains and use of soakways, which is plausible given the permeable subsurface of Berlin to follow the concept of sponge-cities (Nguyen et al., 2019). Such green infrastructure will help return more natural elements of the flow regime of a river like the Panke. However, effluent from the WWTP will continue to dominate flows. On the positive side, this can provide a nutrient subsidy, though eutrophication risk, in terms of ecological energy flows (Aristi et al., 2015; Hamdhani et al., 2020) and sustain dry weather flows conditions in the context of decoupled groundwater-surface water interactions. However, simultaneous improvements in water quality conditions are contingent on 4th level treatment being implemented at the WWTP (pre-COVID-19 planned for Sept. 2021, (Gnirß et al., 2017)). In the meantime, other management tools such as in-stream structures to improve aeration and re-engineering of canalised reaches for habitat diversity are ongoing (Lange et al., 2015; Wasser- und Bodenverband „Finowfließ“, 2011). This is aimed at improving the amenity and ecological value of an important riverine green corridor in the city that is widely used by people and wildlife (Senate Department for the Environment, Transport and Climate Protection & (SenUVK), 2019). However, such steps are merely a start in creating new visions for urban streams that will require the engagement of stakeholders and balance sometimes competing demands (Luyet et al., 2012). This will be a long and complex process but will ultimately require a fundamental re-appraisal of urban hydrology, which isotope studies can contribute.

6. Conclusion

We combined temporally intensive and spatially extensive sampling to monitor stable isotopes in rainfall, streamflow, groundwater, treated wastewater and urban storm runoff in the 220km2 highly urbanised Panke catchment in Berlin, Germany. The monitoring was aimed at assessing the temporal dynamics and spatial patterns of the sources of streamflow. This was achieved by using isotope data in Bayesian approaches to end member mixing to assess contributions by contrasting sources of stream flow. The Panke has a lowland catchment that is naturally groundwater dominated; however, urban surfaces cover ≈ 35% of the catchment and urban storm drains have an important influence on runoff generation. In the upper catchment, groundwater and urban storm drainage accounted for around 75% and 25% of annual runoff, respectively. In the lower catchment, however, effluent from a WWTP accounted for 80% of streamflow, with groundwater and urban storm runoff each accounting for around 10%. Regulation of sources in the Panke by artificial weirs increased WWTP contributions to augment summer baseflows, and reduced contributions from urban storm drains during high flows as a flood alleviation scheme diverted a portion of high flows into a neighbouring catchment. We also estimated the contribution of the young water fraction (i.e. water that is less than around 2 months old) of streamflow, which was low throughout the catchment, varying between around 10-15%. However, age dating urban streams is challenging due to the undefined age of wastewaters. The study showed how isotopes can provide novel and quantitative insights into how managed urban water systems integrated with the more natural hydrological processes in non-urban areas and urban green spaces. Such understanding is vital to a comprehensive understanding of urban hydrology needed to provide an evidence base for more sustainable management of urban waters.

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DATA AVAILABILITY

Stream data and groundwater level based in rating curves, presented in Figure 3, c,d,e are only available upon request from the Berlin Senate, the same applies for the wastewater effluent data which is available from the Berliner Wasserbetriebe (BWB). Precipitation data is available publicly from the Deutsche Wetterdienst (DWD), station Buch. Precipitation and stream isotopes are available upon reasonable request.

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