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Spatial aggregation of time variant stream water ages in urbanizing catchments

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ABSTRACT

We calibrated an integrated flow-tracer model to simulate spatially distributed isotope time series in stream water in a 7.9 km^2 catchment with an urban area of 13%. The model used flux tracking to estimate the time-varying age of stream water at the outlet and both urbanised (1.7 km^2) and non-urban (4.5 km^2) sub-catchments over a 2.5 year period. This included extended wet and dry spells where precipitation equated to >10 year return periods. Modelling indicated that stream water draining the most urbanised tributary was youngest with a Mean Transit Time (MTT) of 171 days compared with 456 days in the non-urban tributary. For the larger catchment the MTT was 280 days. Here, the response of urban contributing areas dominated smaller and more moderate runoff events, but rural contributions dominated during the wettest periods, giving a bimodal distribution of water ages. Whilst the approach needs refining for sub-daily time steps, it provides a basis for projecting the effects of urbanisation on stream water transit times and their spatial aggregation. This offers a novel approach for understanding the cumulative impacts of urbanisation on stream water quantity and quality which can contribute to more sustainable management.

Keywords: isotopes, transit times, conceptual models, non-stationarity, urban hydrology

1. INTRODUCTION

Advances in quantifying the travel times of water through catchments have shown that this depends on the interaction of flow path partitioning, the size and nature of the soil and groundwater stores and the associated mixing processes that assimilate precipitation into storage (McDonnell et al., 2010). An important insight has been awareness of time variant influences on transit time distributions (TTDs) as highlighted by Rodhe et al. (1996). This time variance reflects the role of short-term individual storm event characteristics and antecedent conditions (Birkel et al., 2012); the effects of seasonal and inter-annual climatic variability (Hrachowitz et al., 2013; Heidbuechel et al., 2013), the mixing assumptions for different stores (Fenicia et al., 2010; van der Velde et al., 2014) and the impact of extreme events (Lyon et al., 2008). Sensitivity of catchment TTDs thus varies geographically depending upon catchment characteristics and the variation in hydroclimate (Tetzlaff et al., 2009). Full understanding is predicated on long-term data sets charactering climatic variability (Hrachowitz et al., 2009), high resolution tracer sampling to capture short-term dynamics (Roa-Garcia and Weiler, 2010), characterisation of older waters (Stewart et al., 2010) and identification of ecohydrologically relevant mixing of waters and solutes (Brooks et al., 2009). Only then can the response of catchments be tracked as it transits through dry periods and times of extreme wetness.

More sophisticated modelling approaches are also needed to capture time variant processes that govern the routing of water and solutes. These should ideally go beyond traditional lumped, time-invariant convolution approaches to fitting TTDs to input-output data. Recent developments include more robust theoretical frameworks for modelling the temporal dynamics of water and solute transport (Botter et al., 2011; Duffy, 2010 Heidbuechel et al., 2012; Rinaldo et al., 2011), use of conceptual rainfall-runoff models to track tracer fluxes and water ages (Dunn et al., 2010; McGuire et al., 2007; Hrachowitz et al., 2013) and the use of particle tracking to simulate evolution of stream water transit time distributions (Davies et al., 2013). Such models should – ideally – have minimum parameterisation to improve identifiability and reduce uncertainty (Beven, 2012). In this regard using both flow and tracers metrics in combined objective functions allows to constrain model structures, improve calibration and aid model evaluation (Birkel et al., 2014).

Such coupled flow-tracer models can provide a step forward for using time variant transit time analysis in applied hydrology. McDonnell and Beven (2014) identified consistent conceptualisation of hydrological response celerity with pore water velocities distributions as a major research frontier in hydrology. Progress is needed to produce models that can adequately characterise the hydrological response dynamics, storage characteristics and mixing relationships in catchments. Only then can robust projections be made about how catchments mediate impacts of land use change on stream water quantity and quality in terms of routing, storage dynamics and transit times (Gall et al., 2013). Monitoring effects of land use change on tracer dynamics~~,~~ thus~~,~~ provides an opportunity for testing such integrated modelling approaches. It also provides an experimental framework for testing hypotheses about how flow partitioning, storage dynamics and water transit times change. Urbanisation provides rapid, direct and large-scale hydrological impacts that affect extensive parts of the world (Grimm et al., 2008). This has policy relevance as more sustainable approaches to urban planning seek to retain catchment storage functions by minimising the extent of impermeable surfaces and creating retention wetlands to mitigate increased flood risk, reduced summer low flows and deterioration of water quality (e.g. Niemczynowicz, 1999; Zipperer et al., 2000). This can be assisted by understanding how the urban environment affects transit times.

Here we use spatially distributed tracer data from an urbanising catchment which transitioned through some hydroclimatic extremes over a 2.5 year period. These are used together with a coupled flow-tracer model to answer the following questions: (1) What are the time-variant characteristics of the age composition of stream water draining urban and non-urban environments? (2) How is the time-variance of stream water age composed in relation to the influence of different land uses at larger scales? (3) Can we use such models to predict the impact of urbanisation on the stream water age? We also highlight research needs for such work to inform the development of green infra-structure to facilitate more sustainable approaches to managing urban water.

2. STUDY AREA AND DATA

The study site is the Burn of Bennie, a 7.9km^2 catchment (Fig. 1 and Table 1) in NE Scotland. Two tributaries with similar soils, geology and topography, form the headwaters; one rural in the north has ~1% urban influence and drains a patch work of forest, wetland and farmland. The southern tributary has been subject to increased urbanisation over the past two decades (now covering 12.4% of its area), with the lower catchment downstream of the main tributary confluence being similarly impacted (13.1% total). Further urban expansion is planned around the southern tributary, increasing the total urbanised area of the catchment to 16.3% (http://aberdeenshire.gov.uk/statistics/area/BanchoryProfile2013.pdf). The climate is temperate; mean annual precipitation is 950mm (varying between ~700-1000mm), mean July and December temperatures are 13^{o}C and 7^{o}C, respectively. Potential evapotranspiration is 500mm and fairly constant. The catchment is underlain by a uniform, low permeability bedrock covered by deep (10-20m) layers of glacial drift, with alluvium in the river valleys. The drifts are coarse textured and podzolic soils, which are mainly freely-draining, dominate.

Five sites were sampled from October 2011 at approximately weekly intervals (Fig. 1 and Table 1). These include the main catchment outfall (ID=outlet), and the lowest points on the rural (ID=rural) and urban tributaries (ID=urban). Also, a site on the upper urban tributary was sampled (ID=urban a) as was the outflow from a retention pond draining a recently urbanised area (ID=urban b). Sampling targeted hydrological events; most significant events were sampled and larger events were sampled daily over the hydrograph. Daily precipitation samples were collected from a small weather station

98 (Fig. 1). Samples were analysed at the University of Aberdeen for ^{18}O and ^{2}H using a Los Gatos DLT-100 (2nd generation) laser spectrometer and are reported in δ units (‰) relative to Vienna Standard Mean Ocean Water (V-SMOW). Analytical precision was 0.2‰ for $\delta^{18}O$ and 0.4‰ for $\delta^{2}H$. Given the greater relative precision we use $\delta^{2}H$ in the modelling that follows.

Precipitation data from 14 surrounding daily gauges were used from the MIDAS data sets at the British Atmospheric Data Centre to derive daily areal totals via spatial interpolation with the gradient inverse distance method (Stahl *et al*., 2006) on a 100m grid. An Automatic Weather Station 10km south of the site was used to derive ET estimates for the catchment using Penman-Monteith. Due to vandalism problems, continuous flow gauging in the urban catchment was precluded. Thus, we used the method of Seibert and Beven (2009) to manually gauge flows (10 gaugings in total) across the flow spectrum as calibration targets for a runoff model to produce flow time series. This was found to give acceptable estimates of daily flows at the rural, urban and outlet sites (see Soulsby et al., 2014).

3. INTEGRATED FLOW-TRACER MODEL

3.1 Model structure

We used the data to develop a simple conceptual, tracer-aided rainfall-runoff model representing the two main landscape units (urban and rural) combined in three sub-catchments represented by the rural, urban and outlet sites (Figure 2). After testing different arrangements of the stores in the model we settled on the most parsimonious semi-distributed structure. This consisted of a single linear reservoir draining urban areas (storage component S_{urban} drained by the rate parameter u) and a two-reservoir model for rural areas. The upper reservoir S_{up} was parameterised (rate parameters k and nonlinearity parameter α) with a nonlinear power-function type equation to account for faster flow components. The lower reservoir S_{low} is filled by a linear recharge flux (parameter *r*) and linearly contributes to streamflow via the rate parameter *b*. Both components add up to total stream flow. For mixed landscape sub-catchments, the rural and urban model units were combined, with the areal fraction of their flow-tracer contribution to the catchment outlet summing to one.

Tracer transport and mixing was directly linked to the hydrological reservoirs using a mixing cell approach similar to Hrachowitz et al. (2013):

$$\frac{d(c\ S\ )}{dt} = \sum_j c_{I,j} I_j - \sum_k c_{O,k} O_k \qquad \text{Eq. 1}$$

With *c* being the isotope signature of storage components (‰) in *j* storage inflows I_j (precipitation and recharge) and *k* outflow O_k components (actual evapotranspiration ET_{act} and upper reservoir outflow Q_{up}), which characterizes the catchment storage *S* dynamics (sum of dynamic and additional storage components available for mixing that do not affect the water fluxes) and associated isotope signature *c*. Additional mixing stores were calibration parameters in the form of fixed mixing volumes (MV_{up} in S_{up} and MV_{low} in S_{low}). These facilitate reproduction of observed tracer attenuation in streams. As urban reservoirs are assumed to directly contribute tracers to the stream no mixing parameters were used. We did not incorporate isotopic fractionation as tests did not show an improvement to simulations. For the different subcatchments, the same parameters were applied to represent the same landscape units and keep the model as parsimonious as possible (7 parameters in total; 5 rainfall-runoff and 2 tracer parameters as indicated in Fig. 2).

3.2 Water age estimates

We used a flux tracking approach (Hrachowitz *et al.*, 2013) to estimate water age. As each new daily input flux can be labelled, the evolving age of water in different stores can be tracked to give a final age label upon discharge to the stream. The stream water age is then calculated as a flux-weighted age from the contributions of the urban and upper and lower reservoirs. Time variant water age distributions for flow generated from the different units and sub-catchments ~~was~~ then calculated:

$$p_{F,Q}\left(t_j - t_i,\ t_j\right) = \sum_{n=1}^{N} p_{F,Q_n}\left(t_j - t_i,\ t_j\right)\frac{Q_n\left(t_j\right)}{Q\left(t_j\right)} \qquad \text{Eq. 2}$$

Where $p_{F,Q}$ is the distribution of water age of contributing fluxes Q_n to total discharge Q with t_j being the time of exit at the catchment outlet and t_i the time of entry with precipitation. Simulations used a 5 year warm-up period to initialise storage filling, signatures and water ages (starting at 1 with the first day of simulations). Isotope storage values were initially set to observed mean discharge signatures for each site. Actual planning proposals for new urban areas (Fig. 1) were used to predict impacts on water age distributions using the calibrated model, assuming a stationary climate.

3.3 Model calibration

We integrated calibration objectives using the manual point gaugings for the streamflow targets and isotope time series measured at the urban, rural, and outlet sub-catchments as the tracer targets. We also used the ratio of measured flows of the rural and urban sub-catchments as an additional stream flow target. Calibration used the modified Kling-Gupta efficiency (KGE) for the two separate (streamflow *KGE_Q* and tracer *KGE_D*) objectives (Kling *et al.*, 2012):

$$KGE = 1 - \sqrt{\left(1-r\right)^2 + \left(1-\gamma\right)^2 + \left(1-\beta\right)^2} \qquad \text{Eq. 3}$$

The *KGE* is a three-dimensional representation (Euclidean distance) of the widely-used Nash-Sutcliffe criterion balancing dynamics (correlation coefficient *r*), bias (bias ratio *ß*) and variability (variability ratio *γ*) with a perfect fit of 1. The combined streamflow and tracer targets were equally weighted and minimized as the unknown Euclidean distance from the optimal model using a multi-objective Non-dominated Sorting Genetic evolution Algorithm (NSGA) (Deb *et al.*, 2002). This searches a relatively wide initial parameter space and sequentially combines and updates 500 parameter sets over 1000000 iterations until no further improvement can be established. The best 500 parameter sets were used for subsequent analysis.

3.4 Parameter identifiability

We used a regional sensitivity analysis (RSA) (Hornberger and Spear, 1981) to assess identifiability of the best 500 parameter sets. The resulting curves or constrained parameter spaces indicate identifiability, while a straight line over the initial parameter space shows non-identifiability.

3.5 Lumped convolution integral model

Transit times from the flow-tracer model were compared to a time invariant convolution integral model (see Hrachowitz et al., 2009) using the two-parameter (shape parameter α and scale parameter β) gamma distribution as a transfer function (Kirchner et al., 2001). The KGE criterion was used to calibrate the gamma model (GM) with a single target (isotope time series measured for each sub-catchment) using a simple Differential Evolution (DE) algorithm for optimization (Mullen *et al.*, 2010). The 2.5 yr record was used for calibration with a warm-up of 5 years. Due to the insensitivity of stable isotopes in detecting older water, the β parameter was set to the upper limit of 5 years.

4. RESULTS

4.1 Hydroclimatic and isotopic variability

During the study the site experienced a range of unusual and contrasting hydroclimatic extremes (Fig. 3). The first year had a dry winter, but the summer of 2012 was the wettest for 20 years in NE

Scotland. In the second year, the autumn was wet and in Dec. 2012 the highest daily precipitation totals and flows were recorded. This was followed by a warm dry summer in 2013 corresponding to a 10 year return period drought which persisted through autumn but was followed by high precipitation in both Jan. and Feb. 2014 (see UK Hydrological Summaries for individual months at http://www.ceh.ac.uk/data/nrfa).

Isotopes in precipitation exhibited general seasonal differences between winter depletion and summer enrichment, but day-to-day variability within season was marked (Fig. 3). Isotope variability in stream flow was most pronounced in the most urbanised sites (Table 2), with urban impacts evident in rapid, direct routing of tracer signals in precipitation both in small and large events. Variations were attenuated in the rural tributary where dynamics in stream water isotope signals ~~were~~ exhibited more gradual changes in the wetter periods in June and Dec. 2012 and Jan/Feb 2013. At sites with both rural and urban influence, the tracer variations reflect both the urban influence of marked isotopic variability in smaller events but a general damping in large events and baseflows.

4.2 Flow-tracer model simulation results

The integrated flow-tracer model captured the measured peak discharge and following recession period in Dec. 2012 (Figure 4a). Additionally, the model was able to match the observed lower flows, despite failing to reproduce the higher observed flows in June 2012. However, the model did capture small and moderate events with dry antecedent conditions where the lower catchment response is dominated by the urban response. The model calibrated to the common flow target performed reasonably well with a best-fit KGE = 0.66 (R^2 = 0.72). Also, most rainfall-runoff model parameters were identifiable over a constrained range (Fig. 5 and Table 3). The least constrained were the nonlinearity parameter α and the recharge rate parameter r.

Stream isotope variations were captured quite well and the posterior variability of the best 500 parameter sets bracket most of the observations (Fig. 4b-d). Additional mixing volumes were identifiable with a constrained upper volume MV_{up} (150 to 280mm) and an identifiable parameter region of MV_{low} at > 800mm (Fig. 5e and f). The best isotope simulations were for the most urbanised catchment; best-fit KGE of 0.86 and R^2 of 0.89 (Fig. 4d). In the rural tributary the challenge of the damping was exacerbated by the effects of evaporative fractionation in the summer of 2014 (Fig. 4c). Thus, invariant isotope values were simulated in outflows from the lower groundwater box which also failed to capture short-term effects of summer events or fractionation. This also explained why isotopic fractionation tests showed little model improvement. Nevertheless, overall simulations were reasonable (best-fit KGE = 0.78 and R^2 = 0.8). The summer problems were also evident to a lesser extent in the simulations for the integrated lower catchment, where the lowest flows are generally dominated volumetrically by the rural catchment (Fig. 4d and Table 3). However, again, overall the model performed well over the whole period (best-fit KGE = 0.83 and R^2 = 0.85).

4.3 Spatial aggregation on stream water age estimates

Stream water ages were estimated by aggregating the age of water derived from flux tracking in the different model landscape units, revealing marked differences for the 3 sites (Fig. 6). In the rural tributary ages show a bimodal distribution (Fig. 6c and c1); with a 50-100 day peak for larger events, and a secondary peak of 350-400 days reflecting the heavy tailing of old water contributions at base flow. The influence of such slower, deeper flow paths is emphasised by <50% of tracer recovery being modelled at the end of a year. In contrast, the outflow of the modelled urban response unit indicated the very short residence times; here, most stream water is a few days old, and about 70% of the flux occurred in <50 days, with >95% tracer recovery within 400 days (Fig. 6a and a1). In mixed land use catchments (urban and outlet), the integration of both sources ~~are~~ evident (Fig. 6b and c). In the urban tributary, the distribution is modal in the 50-100 day age class, with limited tailing (Fig. 6b and b1). However, at the lower site (outlet) the distribution is bi-modal with the urban area giving 50-100 day modal class, and rural inputs causing a secondary peak at 300-400 days (Fig. 6d and d).

The Transit Time Distribution derived using the time invariant Gamma Model fitted to the input-output data had lower performance statistics than the flow-tracer model though still reasonable fits (Table 4). The α parameter in the GM was 0.63 for the rural site, but reduced to 0.2 as the percentage urban area increased. Such a decrease with increasing urban cover was also observed for the MTT. The beta parameter was not identifiable and remained close to the upper boundary of 5 years. The derived best-fit MTTs for the urban, rural and outlet sites from the GM at 366, to 1050 to 456 days respectively, were shorter than for the flux tracking. However, the posterior variability (indicated by the best 500 parameter sets) of the flow-tracer model indicates significant uncertainty (Table 4) which generally brackets the GM estimates.

4.4 Future urban development

The model was also used for projecting future change by increasing the urban area based on actual planning proposals (Fig. 1). Resulting changes to the cumulative frequency distributions of median water ages shows accelerated routing of water and reduced storage at both the urban and outlet sites, with consequent reduced water ages (Fig. 7). The rural tributary remains unchanged. The uncertainties of the projections are large and overlap as indicated in Table 4, but the direction of change is evident in the median age distributions and consistent with impacts so far. Thus, even if relatively small parts of a catchment are affected by urbanization, this is still sufficient for a dramatic effect on TTDs that can propagate to the larger catchment scale. Although the rural tributary remained unchanged, just a 3% increase in urban area showed a clear impact with the future outlet median age CDF approximating the current urban site.

5. DISCUSSION

What are the time-variant characteristics of stream water age of urban and non-urban streams?

The integrated flow-tracer model gave an estimate of the age distribution of streams draining catchments with differential levels of urbanisation. Unsurprisingly, this infers a much higher proportion of newer water, and less tailing of older water, than shown in time-invariant TTD analysis (see Table 4 and Soulsby et al., 2014). It is striking just how much of the precipitation in urban-impacted areas appears to reach the stream within a few days, given the implications for storage and low flows. However, this also reflects the model structure that simply assumed rapid runoff from the urban areas, rather than for example, deeper infiltration in gardens etc. in order to maintain minimal parameterisation (Kirchner, 2006). It is clear that our model does not attempt to accommodate the hydraulic infrastructure of the complex reality in urban systems. However, in the absence of more spatially distributed and higher temporal resolution (sub-daily) flow and tracer data to conceptualize and evaluate a more complex model, such efforts would likely result in increased uncertainty (Beven, 2012). Although we did not present formal uncertainty analysis, the posterior variability of the best 500 parameter sets retained after multi-objective optimization provides an uncertainty proxy; Andrews et al. (2011) showed that the best parameter sets using such optimization methods falls into the range of behavioural parameter sets defined using more formal uncertainty analysis. Also, the model was internally constrained using isotope time series from three sub-catchments resulting in generally good tracer simulations over 2.5 years of marked hydroclimatic variability (Fig. 4) with mostly identifiable parameters (Fig. 5) (Birkel et al., 2014). As a result the isotope simulations for the three sub-catchment time series outperformed flow simulations (Table 3).

Damped tracer signals increase uncertainty of simulations in the rural catchment (MTT = 473 days with a 95th percentile range of 326 - 864 days; Table 4). A bimodal distribution was simulated with relatively young waters being derived from the upper soil reservoir in wet periods, consistent with agricultural drainage effects in the low-lying areas. However, a greater proportion of older water draining by slower flow paths gives bimodality in age distributions at about 1 year and heavier tailing beyond this. Such complex age distributions derived from conceptual models using flux tracking have also been reported by Dunn et al. (2010) and Hrachowitz et al. (2013). Notwithstanding the influence of hydroclimatic variability on stream water age, the bi-modal distributions are more consistent with differences in land use and the resulting variability in flow pathways. Such effects of variable flow pathways on TTDs was also shown by Heidbuechel et al. (2013) albeit in a more markedly seasonal semi-arid catchment in Arizona, USA. Mixing assumptions also influences time-variable TTDs (van der Velde et al., 2014) and the complete mixing used here is overly simplistic. However, the combination of three parallel reservoirs with one nonlinear parameterization creates a more dynamic catchment scale conceptualisation for assessing the time-variant TTDs.

The characterisation of TTDs achieved with the integrated flow-tracer modelling approach is an improvement on using an invariant convolution integral model (Table 4). Although these showed the same ranking of MTTs (e.g. urban site < outlet site < rural site), the resulting MTTs are longer than those produced by the time-variant modelling by a factor of 2.1 for urban, 2.4 for rural and 1.62 for the outlet showing the instability of the time invariant GM for short (<5 year) data time series (Hrachowitz et al., 2009). Nevertheless, for both the urban and outlet sites, GM estimates fall within the 95%ile error bands of the flow-tracer model.

How is time variant streamwater age affected by integration of different land use at larger scales?

Time variant flow-tracer modelling allowed us to develop a hypothesis of catchment function which integrates the urban impact on stream water ages at different scales. This is based on the integration of sub-catchments and their dominant processes in relation to land use, which can be seen as upscaling runoff generation and solute transport processes from the hillslope (e.g. Tetzlaff et al., 2014) to catchment scale. Use of multi-objective calibration targets produced a reasonable representation of the runoff and isotope response in the two main sub-catchments and at the catchment outlet (Fig. 4). The isotope damping at the rural site leads to larger uncertainties~~,~~ in the summer tracer simulations, and this propagates to the outlet site. Despite this, for most of the study period~~,~~ simulations capture the dynamic influence of the two headwater tributaries isotopes at the outlet. Thus, the strong influence of the short-term dynamic of urban drainage is evident in the modal age class of 50-100 days at the outlet site, but the bimodality shows the greater volumetric contributions of older water from the rural catchment in the secondary peak at 300 days (Fig. 6d). It is also notable that despite the similar percentage urban cover at the urban and outlet sites, the effect is much greater at the former, though the urban impact remains cumulative at the outlet.

Although these contrasts mainly reflect the moderating effect of the rural catchment at higher flows, they may also reflect differences in the catchment land use and the nature of the urban developments. The non-urban areas in the sub-catchment of the urban site are predominantly (59%) rough grazing, whilst in the sub-catchment of the rural site it is mainly (64%) forest. This is likely to result in longer residence times in the forested areas due to lower artificial drainage and greater infiltration to depth (e.g. Geris et al., 2014). In addition, urban areas in the upper catchment above the urban site are mainly housing up to 15 years old, which has a higher density of impermeable surfaces and limited sustainable urban drainage systems (soakaways, retention wetlands etc). In contrast, urbanisation in the lower catchment is more recent (mostly <10 years), on-going and comprises an industrial estate and retail outlets, with less dense coverage of impermeable surfaces, more green space, permeable parking areas and some retention ponds (cf. O´Driscoll et al., 2010). Lower density urban development is likely to increase water age due to greater sub-surface storage and mixing (and therefore decrease impacts), but modelling this would require explicit integration of more detailed land use classes accompanied by corresponding data for parameterization. Such improvements to the current model are under way and future work will be directed at detailed investigations about the impacts of smaller scale urbanisation of variable density on stream water ages (cf. Thompson et al., 2013).

Can we project the impact of increased urbanisation on the composition of stream water age?

The flow-tracer modelling approach already shows promise as a tool for predicting change in TTDs resulting from further urbanisation, with the increasing dominance of younger waters at both the urban and outlet sites likely to result from the urban growth already planned in the catchment (Fig. 7). However, as noted, the approach is simplistic and work is needed to reduce the inherent uncertainties and provide knowledge transfer for urban design. A key need is higher resolution data (Kirchner et al., 2004); modelling at a daily time step is only a first approximation in assessing urban effects on TTDs. Although sampling targeted high flow events, in reality, the response time of impermeable urban surfaces is much less than this and will influence stream flows on a sub-hourly basis during more intense precipitation (O´Driscoll et al., 2010). Thus, collecting high resolution data to refine the model for sub-daily scales is needed to characterise the faster tail of the TTD and its overall influence more realistically (Birkel et al., 2012). The current uncertainties are reflected in the 95^{th} percentiles around the MTTs calculated from the best 500 parameter sets (Table 3). At the outlet this results in a MTT of 280 days with 95^{th} percentiles of 208 to 520 days. Although these ranges imply some overlap in the CDFs, the general features such as the observed bi-modality and the direction of projections from rural to more urban remain unaffected.

In addition, dynamics of the longer residence time waters need to be constrained by (a) using tracers that can quantify ages beyond the 5 year limit of stable isotopes (Stewart et al., 2010) and (b) tracking the catchment response over longer periods (Hrachowitz et al., 2009). Despite constraining the older waters in the flux tracking approach by looping the observed time series, it is unlikely that all potential climatic variability is captured over relatively short monitoring periods, though the 2.5 years used here were remarkably variable. This is illustrated by previous analysis of the data from the study site in the first year which covered a relatively dry period: the characterisation of TTDs for the urban and outlet sites in this case showed they were relatively similar (see Soulsby et al., 2014). The new analysis presented here using the 2.5 year data set, including two wetter periods and a more pronounced summer dry period (Figure 2) than were observed in the first year, showed much larger water age differences than previously reported.

Finally, the model calibration resulted in substantial uncertainties stemming from the lack of distributed continuous flow data, particularly from the rural tributaries and the integration of temporally different flow contributions from tributaries to the catchment outlet. Whilst this does not affect the general conclusions derived from daily time steps, it would be inadequate for assessing the sub-daily variations described above. Future work will focus on improving monitoring which in turn can be used to develop a more complex and spatially explicit model. We therefore envision incorporating wetlands and soakaway systems as temporary stores, and a lower reservoir in the urban store that allows infiltration to deeper flow paths, sewer overflow and drainage exports.

The model's capability in integrating intra-catchment differences in daily flow and isotope responses to better understand the heterogeneity of processes that underpins catchment water age distributions is encouraging. There is clear potential use of such insights in urban planning for a sustainable water environment. If the data limitations outlined above can be overcome, an evidence base would emerge that potentially could provide specific guidance on the timescale of water fluxes that could be targets for water retention in new urban developments. This could include, for example, guidance on the design of green infrastructure such as wetlands and detention ponds or permeable surfaces to maintain storage to mitigate flood risk, sustain low flows and improve highly relevant water quality. Currently, such infrastructure is developed from guidelines that often have little specific context for the hydrology and landscape for where the development is occurring. Similarly, as urban development often occurs in a piecemeal *ad hoc* way, the scaling of impacts and their cumulative effects are often ignored. Whilst such issues remain substantial challenges, we present a first step to better understand the cumulative impact of urbanisation on transit times that could be used in future to create more sustainable approaches to urban water management.

6. Conclusion

Flux tracking is a useful tool for characterising complex time-variant water age distributions and their spatial aggregation in catchments impacted by urbanisation. Whereas previous studies have usually looked at input-output relationships for individual catchments (Hrachowitz et al., 2013) or different hydrological response units within a catchment (Birkel et al., 2014), here we have shown its utility in integrating responses from contrasting units in a mixed land cover catchment. The dramatic impact on urbanisation on the quick responding part of the TTD is captured both at the local and larger catchment scale. The flow-tracer model captured bi-modal TTDs with a higher proportion of young water and less tailing of older water than using time-invariant models. Though challenges remain, the approach provides a step towards a framework that could be used in urban design that seeks to restrict the rapid water fluxes traditionally associated with urbanisation and maintain catchment storage to sustain low flows, mitigate flood risk and ameliorate water quality impacts.

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TABLES

Table 1: Physical characteristics of the Burn of Bennie catchment (outlet) and the urban and rural sub-catchments.

ID		Topography			Land use		
	Area (km^2)	Elevation range (m)	Mean Elevation (m)	Mean Slope (°)	Urban (%)	Forest (%)	Agri-culture (%)
Urban a)	0.34	36	90	2.51	63.3	36.7	0
Urban	1.68	55	83	2.26	12.4	28.6	59
Urban b)	0.027	4	64.4	1.08	100	0	0
Rural	4.5	104	90	2.83	1.0	63.8	24.7*
Outlet	7.9	117	86	2.65	13.1	68.5	31.5

*: 10.5% catchment area corresponds to wetland area and landfill site.

Table 2: Isotope summary statistics for the 2.5 years study period (October 2011 – April 2014).

ID		n	Mean	Min	Max	SD
Precipitation	Un-weighted	544	-54.8	-147.3	-8.4	21.7
	Weighted		-59.9			
Urban a)		150	-54.2	-95.0	-26.6	7.6
Urban b)		150	-53.5	-120.0	-14.7	11.6
Urban		157	-53.2	-78.7	-31.6	6.0
Rural		155	-51.6	-61.8	-40.1	3.6
Outlet		162	-52.6	-75.2	-33.5	5.2

Table 3: Best-fit conceptual model simulations and the posterior parameter and performance statistics in terms of the 5th and 95th percentiles around the median.

		Posterior variability			
Parameter	Initial	Best-fit	Median [5th, 95th]	Median KGE_Q [5th, 95th]	Median KGE_D [5th, 95th]
k	[0, 0.2]	0.03	0.03 [0.007, 0.038]	0.62 [0.6, 0.65]	0.76 [0.58, 0.84]
α	[0, 0.9]	0.38	0.39 [0.36, 0.78]		
r	[0, 0.5]	0.1	0.49 [0.05, 0.96]		
b	[0, 0.1]	0.003	0.003 [0.003, 0.08]		
u	[0, 1]	0.31	0.26 [0.008, 0.38]		
MV_{up}	[1, 500]	244	245 [160, 265]		
MV_{low}	[500, 5000]	507	502 [500, 4531]		

Table 4: Best-fit lumped convolution integral model (gamma model GM) is compared to the best-fit conceptual model (CM) for simulations at the rural and urban tributaries and the outlet. Also given are the water age statistics in days (MTT [5^{th}, 95^{th}]) around the median.

		Urban				Rural				Outlet		
Model	Best-fit MTT (days)	MTT [5^{th}, 95^{th}]	Best-fit KGE (-)	Best-fit R^2 (-)	Best-fit MTT (days)	MTT [5^{th}, 95^{th}]	Best-fit KGE (-)	Best-fit R^2 (-)	Best-fit MTT (days)	MTT [5^{th}, 95^{th}]	Best-fit KGE (-)	Best-fit R^2 (-)
GM	366	-	0.74	0.76	1150	-	0.71	0.7	456	-	0.74	0.75
CM	163	171 [87, 391]	0.86	0.89	521	473 [326, 864]	0.78	0.8	316	280 [208, 520]	0.83	0.85

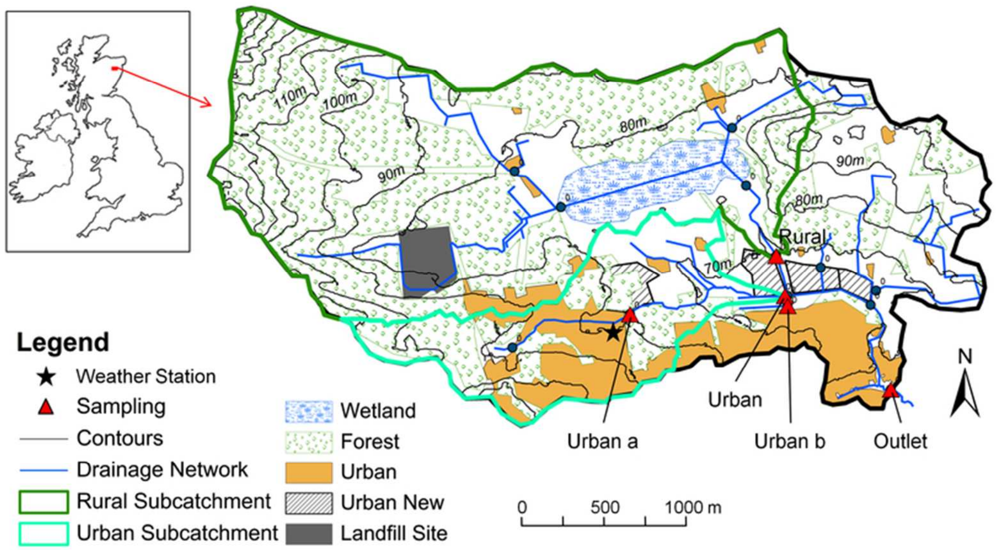

Figure 1: Study catchment location with 5 sampling sites and the main current and proposed extent of land uses (Inset shows location in the UK).
137x78mm (150 x 150 DPI)

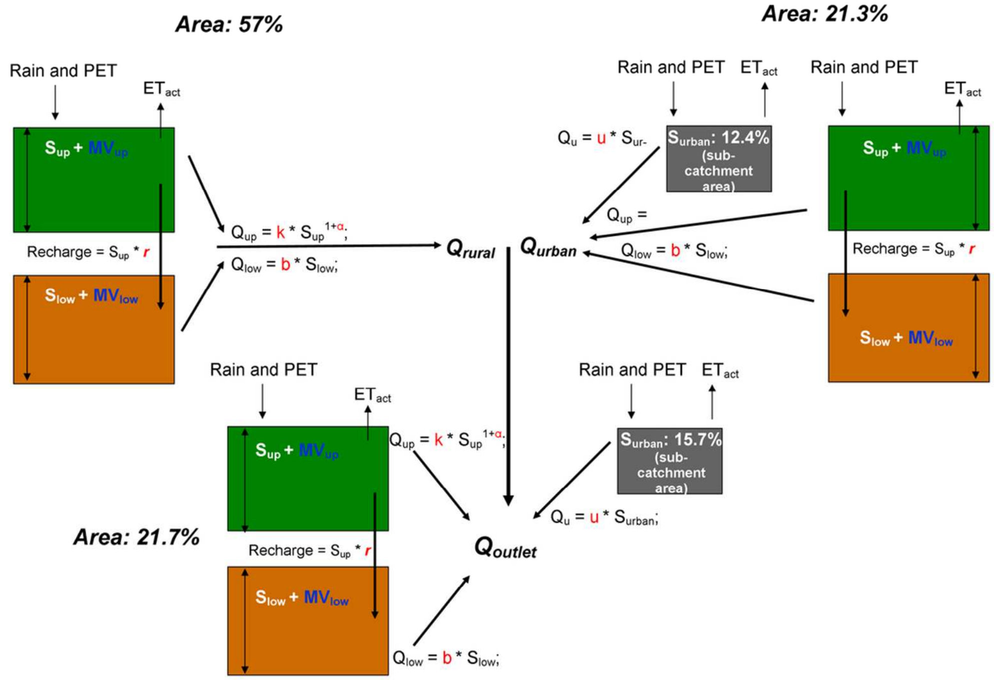

Figure 2: Schematic of model structure and spatial distribution according to catchment configuration. Note that the urban percentages correspond to the respective sub-catchment areas (areas in km^2 given in Table 1), S and MV are dynamic storage and additional mixing volumes, respectively and that the actual ET effectively equals PET. Flux equations are given with hydrologic parameters in red and the tracer transport is indicated with additional mixing volume parameters in blue. However, the same parameters were used for the different sub-catchments to maintain a minimum of calibrated parameters (in total 7).
197x137mm (150 x 150 DPI)

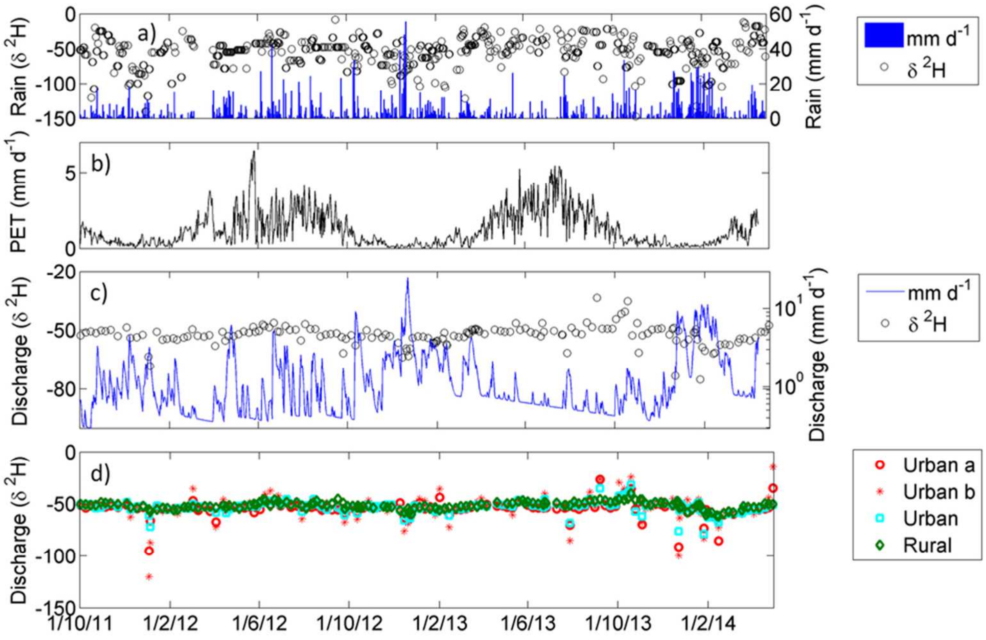

Figure 3: Hydrometric and deuterium isotope daily time series of the study period Oct 2011 to Apr 2014 for a) isotopes and amount of precipitation, b) potential evapotranspiration, c) isotopes and mean simulated discharge at the outlet, and d) discharge isotopes for sites Urban a, Urban b, Urban, and Rural.
162x104mm (150 x 150 DPI)

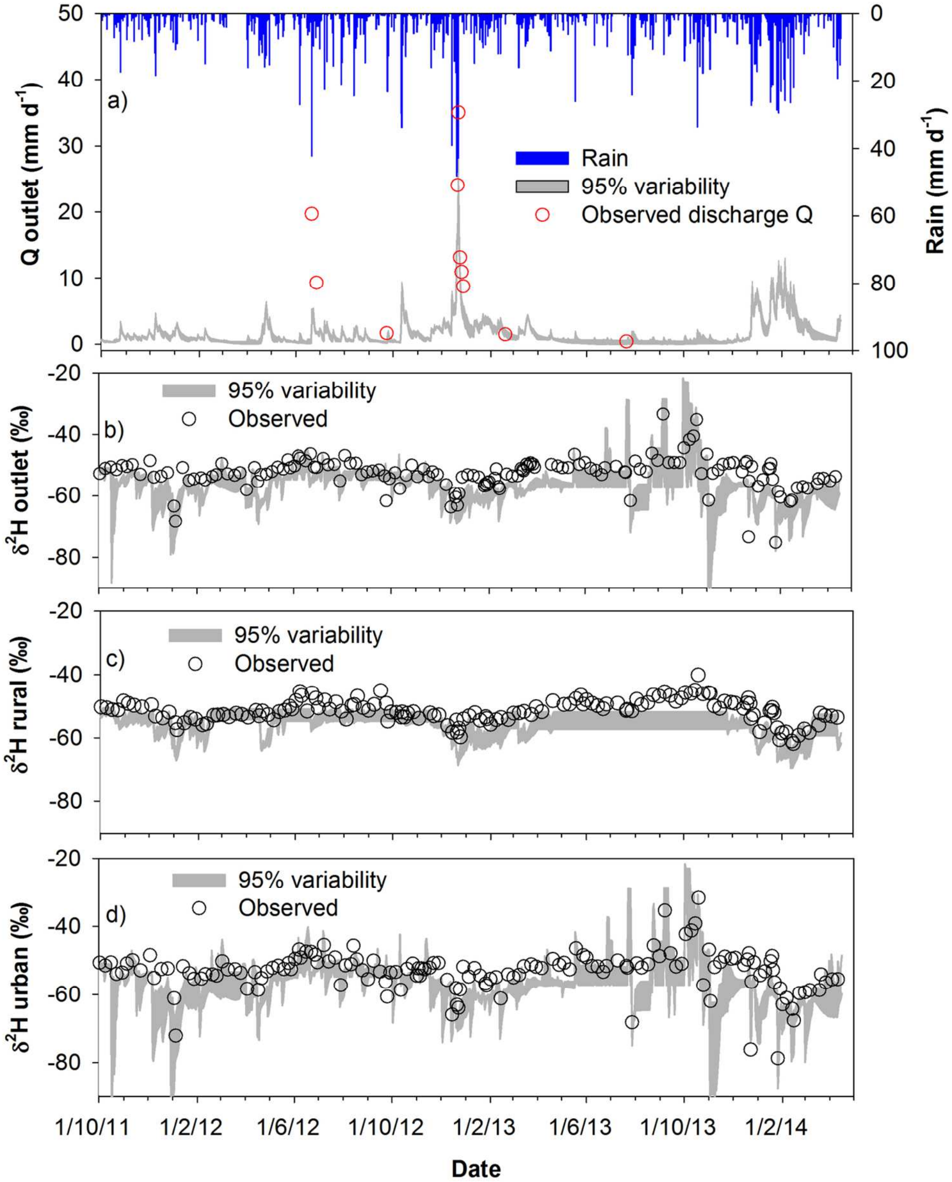

Figure 4: (a) Precipitation and measured and modelled flows at the outlet, together with simulation results showing 95% posterior parameter variability derived from the best 500 parameter sets for the (b) outlet, (c) rural and (d) urban sites.
254x307mm (150 x 150 DPI)

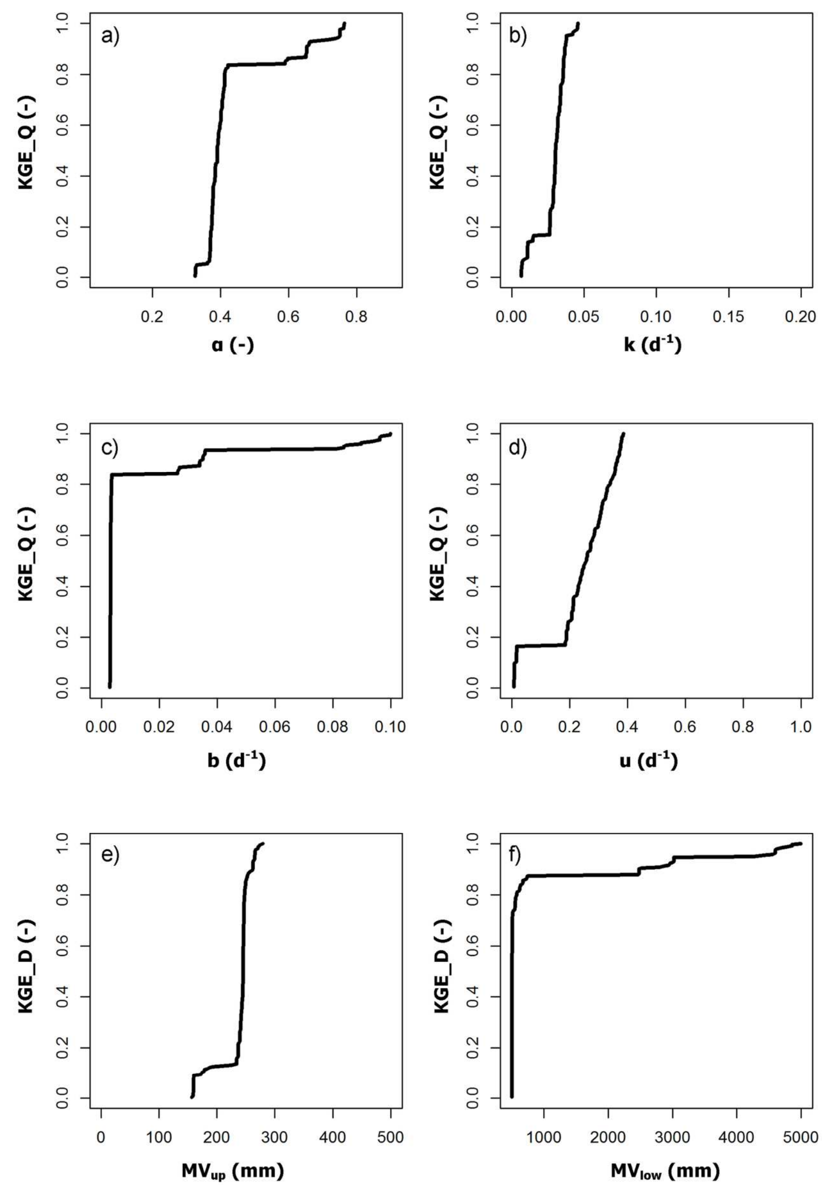

Figure 5: RSA indicates parameter sensitivity for the hydrologic parameters (*k*, *a*, *b*, and *u*) evaluated against the discharge calibration target (KGE_Q) and the tracer parameters (MVup and MVlow) against the isotope calibration target (KGE_D). Increased parameter sensitivity can be derived from stronger curvature compared against a straight line and insensitive parameter. Constrained parameter spaces also indicate parameter sensitivity. Note that the recharge parameter *r* is not shown here.
270x387mm (150 x 150 DPI)

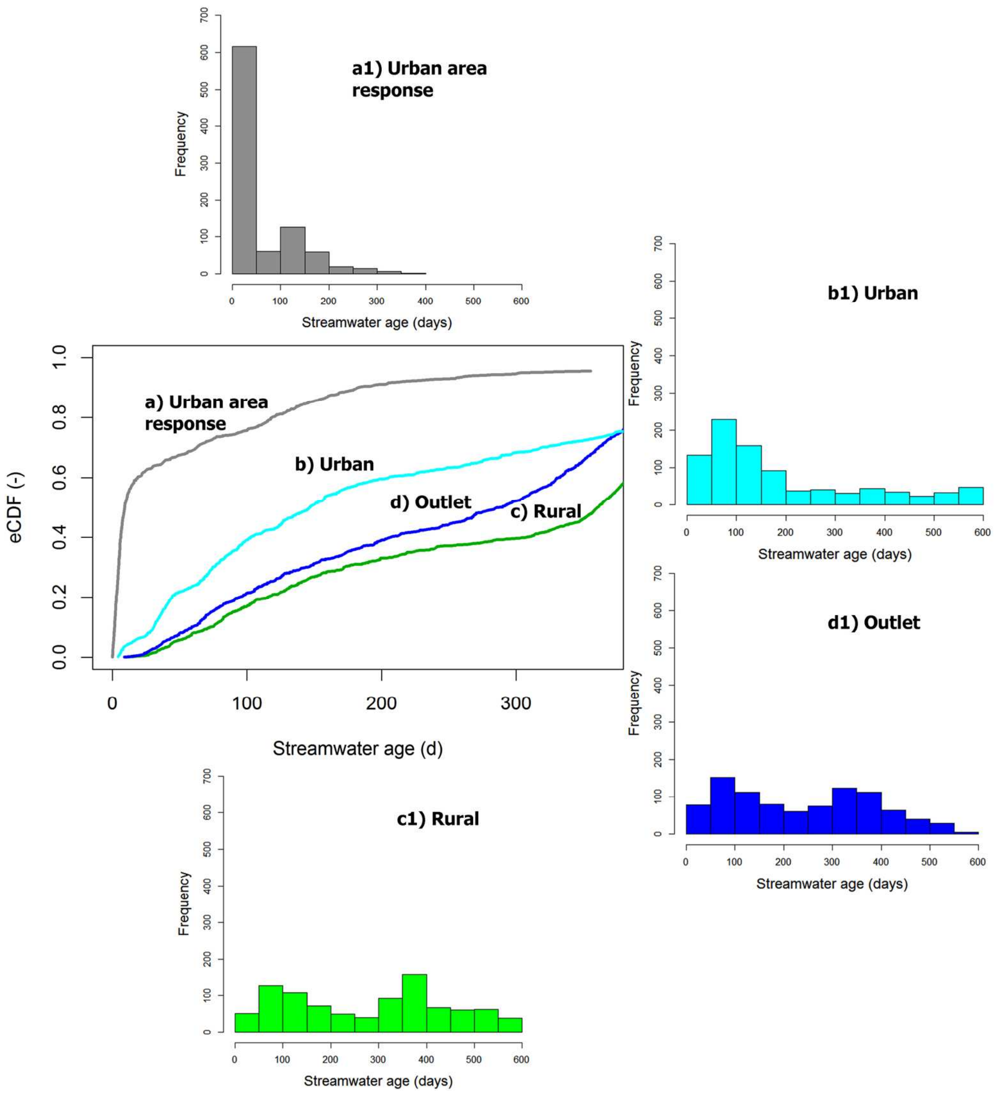

Figure 6: Empirical cumulative median flux water age distribution functions (eCDF) are shown for a) the urban area response (sum of all contributing urban areas), b) the urban and c) rural sub-catchments and d) at the outlet. The streamwater age frequency is visualized as histograms (a1, b1, c1 and d1) using identical axes and classes for comparison.
342x370mm (150 x 150 DPI)

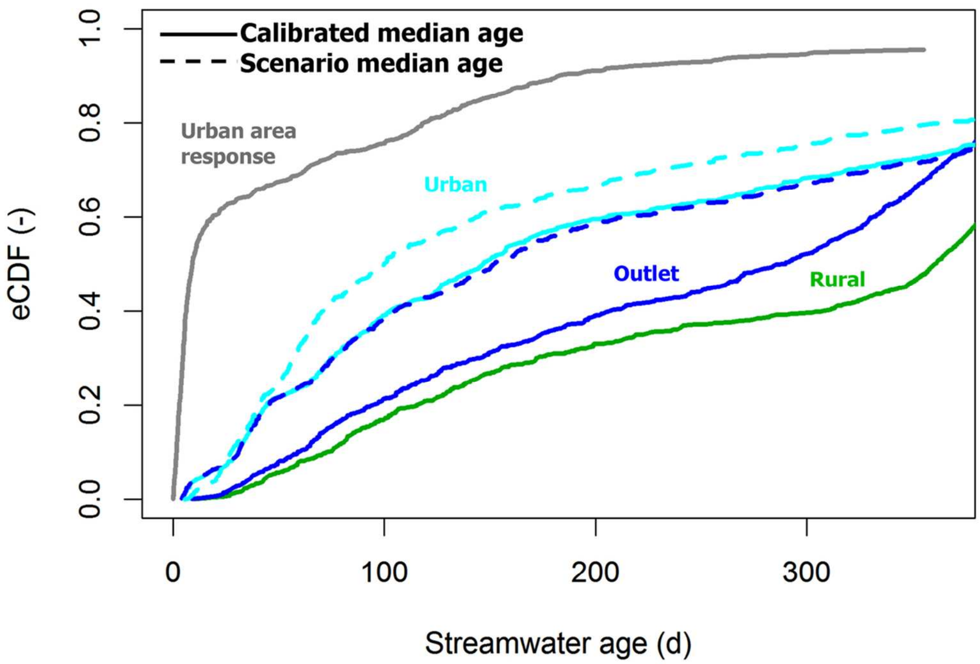

Figure 7: Median water age distributions similar to Figure 6 are compared to scenario water ages (dashed lines) derived from a realistic land use change scenario which will increase the urban cover in the urban sub-catchment and at the outlet by a total of almost 3%. Note that the urban area response refers to the sum of all contributing urban areas.
225x151mm (150 x 150 DPI)