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Using stable isotope tracers to identify hydrological flow paths, residence times and landscape controls in a mesoscale catchment

P. Rodgers[1,*], **C. Soulsby**[1], **S. Waldron**[2], **and D. Tetzlaff**[1]

[1]Department of Geography and Environment, University of Aberdeen, Elphinstone Road, Aberdeen, AB24 3UF, United Kingdom
[2]Scottish Universities Environmental Research Centre, Scottish Enterprise Technology Park, East Kilbride, Glasgow, G75 OQF, United Kingdom
[*]now at: SEPA, Rivers House, Irongray Road, Dumfries, DG2 OJE, United Kingdom

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Correspondence to: P. Rodgers (paul.rodgers@sepa.org.uk)

Abstract

$\delta^{18}O$ tracer measurements of precipitation and stream waters were used to investigate hydrological flow paths and residence times at nested spatial scales in the mesoscale (233 km^2) River Feugh catchment in the northeast of Scotland over the 2001–2002 hydrological year. Precipitation $\delta^{18}O$ exhibited strong seasonal variation, which although significantly damped by catchment mixing processes, was reflected in stream water outputs at six sampling sites. This allowed $\delta^{18}O$ variations to be used to infer the relative influence of soil-derived storm flows with a seasonally variable isotopic signature, and groundwater of more constant isotopic composition. Periodic regression analysis was then used to examine the sub-catchment differences in the mixing of these two main hydrological sources processes more quantitatively, using an exponential flow model to provide preliminary estimates of mean stream water residence times, which varied between 0.4–2.9 years. This showed that the effects of increasing scale on estimated mean stream water residence time was minimal beyond the smallest (ca. 1 km^2) headwater catchment scale. Instead, the interaction of catchment soil cover and topography acted as the dominant influence. Responsive hydrological pathways, associated with peat soils in the headwater sub-catchments, produced seasonally variable $\delta^{18}O$ signatures in runoff with short mean residence times (0.4–0.8 years). In contrast, areas dominated by more freely draining soils and larger groundwater storage in shallow aquifers appear to provide effective mixing and damping of variable precipitation inputs implying longer residence times (1.4–2.9 years). These insights from $\delta^{18}O$ measurements extend the hydrological understanding of the Feugh catchment gained from previous geochemical tracer studies, and demonstrate the utility of isotope tracers in investigating the interaction of hydrological processes and catchment characteristics at the mesoscale.

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1. Introduction

Over the past 2 decades, interpretation of changes in the stable oxygen ($^{18}O/^{16}O$) isotopic signatures of catchment waters have provided insights as tracers for identifying hydrological source areas/flow paths under different flow conditions and estimating mean catchment residence times (Sklash, 1990; Genereux and Hooper, 1998; Burns, 2002). To date, most studies have focused on storm event sampling in relatively small (<10 km^2) catchments (Buttle, 1994). However, the use of isotope tracers to upscale flow path understanding in mesoscale (ca. 10^2–10^3 km^2) catchments has been scarce (e.g. Skalsh et al., 1976; Turner and Barnes, 1998; Frederickson and Criss, 1999; Uhlenbrook et al., 2002). Moreover, those few investigations of scale influence on the mean residence time of runoff have generally been restricted to relatively small catchments (Brown et al., 1999; McDonnell et al., 1999; McGlynn et al., 2003). This reflects the logistical difficulties of sampling in larger catchments, the potential loss of isotopic tracer resolution at larger spatial and temporal scales and the expense of isotope analysis (Buttle, 1998).

From a specific UK perspective, there is a general paucity of experience in the use of stable isotopes for investigating catchment hydrology (Darling et al., 2003). This stems from the often complex climatic and catchment-specific factors controlling their composition, such that for many routine monitoring purposes their measurement is deemed to be of little practical use. From a catchment hydrology perspective however, it is this complexity that provides the potential for insights that are unavailable from other methods. Stable isotope tracers therefore have the potential to play an increasingly important role as the hydrological research community faces increasing pressure to provide improved process understanding and quantitative knowledge at the larger scales where water resource decision-making occurs (Healy, 2001; Naiman et al., 2001; Soulsby et al., 2003). These efforts are likely to be most productive in settings where hydrological processes can be examined at nested catchment scales, where process understanding most commonly gained from small, intensively monitored headwater catchments can be more readily interpreted in relation to behaviour at the larger scale (Soulsby et al., in review[1]).

This paper reports the use of $\delta^{18}O$ measurements as a natural tracer to provide information on hydrological flow paths and residence times for nested sub-catchments in the mesoscale (233 km^2) Feugh catchment in the northeast of Scotland. Previous hydrological studies at this site have focused on the use geochemical tracers to provide information on the role of hydrological flow paths over a range of temporal and spatial scales (Soulsby et al., 2003, 2004, in review[1]). The use of $\delta^{18}O$ measurements was anticipated to build on this work by providing complimentary insight on catchment residence times and the mixing of hydrological sources that will further elucidate the influence of catchment scale on hydrological functioning. This parallels ongoing work to upscale hydrological understanding in the Feshie catchment in the Cairngorm Mountains of Scotland as part of the NERC-funded CHASM (Catchment Hydrology And Sustainable Management) initiative (Rodgers et al., 2004; Soulsby et al., 2004[1]). The aims of the paper therefore are to: (i) characterise spatial and temporal variation in $\delta^{18}O$ of precipitation and stream waters in the Feugh catchment; (ii) establish the main hydrological controls on stream water $\delta^{18}O$ using information from other geochemical tracers; (iii) produce preliminary estimates of the mean residence time of runoff in the catchment and its major sub-catchments and (iv) relate these to catchment landscape controls.

2. Study area

The Water of Feugh drains a 233 km^2 area in northeast Scotland (Fig. 1a). The catchment is predominantly upland in character, with an altitude range from 70–776 m. The climate is cool and wet, with an estimated mean annual precipitation of 1130 mm which

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mainly falls as rain, though snow does occur during the winter months and significant snow pack accumulation can occur in cold years (Soulsby et al., 1997). The catchment is mainly (ca. 85%) underlain by granite, though the most northern parts of the catchment as well as the southern boundary in the Water of Dye sub-basin are underlain by metamorphic rocks (mainly pelites and psammites) (Fig. 1b).

The Feugh is formed by three tributaries (the Dye, Aven and Upper Feugh), which are confluent some 4 km upstream of Heugh Head (the gauging station at the catchment outfall) (Fig. 1a). The largest of these sub-catchments (at 90 km^2), the Water of Dye, is the most southerly and drains a granite-dominated area, although there is a significant outcrop of schist in its headwaters (Fig. 1b). The sub-catchment is characterised by extensive plateaux areas on the interfluves above 450 m that are dominated by peats (up to 5 m deep) and peaty podzols (ca 1m deep) (Fig. 1c). Only on the more incised catchment slopes do the most freely draining humus iron podzols (<1 m deep) occur and the main river valleys generally have freely draining alluvial deposits and soils.

The Water of Feugh sub-catchment is the most northerly with granite-dominated headwaters grading to metamorphic rocks in the lower catchment near Powlair. In comparison with the Dye, the catchment has been over widened by glacial erosion and meltwater action, with more restricted plateaux areas, lower peat coverage and larger areas of more freely draining podzols (Fig. 1c). More extensive alluvial deposits of sands and gravels (>10 m deep) occupy the valley floor, especially in the Powlair area. The smallest sub-catchment (30 km^2) is occupied by the Water of Aven, which lies between the Dye and Upper Feugh. The upper sub-catchment drains an extensive peat-covered plateau underlain by granite, but downstream the valley is very steeply incised, mainly due to erosion by meltwaters. In the lowest part of the sub-catchment, extensive alluvial deposits form a fan, where the Aven confluences with the upper Feugh, and further extensive deposits fill the valley floor between this confluence and the gauging station of Heugh Head (Fig. 1b).

Given the topography and soil coverage in the catchment, land use is largely restricted to grouse (*Lagopus lagopus)* and Red deer (*Cervus elaphus*) shooting on heather moorland in the upper reaches of all three sub-basins (Fig. 1d). The moorlands are managed by regular burning to retain the mosaic of habitats required by grouse. The long history of burning may have contributed to peat erosion, as the peat is extensively "hagged" in many places, particularly in the Aven and upper Feugh catchments (Thompson et al., 2001). This dictates that a high density of ephemeral drainage channels covers the peat, connecting it to the perennial stream channel network. In all three sub-basins, agriculture occupies the better floodplain soils, though this mainly comprises livestock grazing (Fig. 1d). The more extensive coverage of freely draining soils in the upper Feugh sub-basin and the lower catchment above and below the tributary confluences is the main area where arable farming occurs (Table 1). Some of the valley hillslopes are forest-covered, most notably in the lower valleys of the Water of Dye and Upper Feugh. In the former case, the forestry is mainly commercial woodlands, whilst in the latter, semi-natural forests of Scots Pine (*Pinus sylvestris*) predominate (Fig. 1d; Table 1).

The mean annual runoff at Heugh Head, the catchment outfall, is 5.55 $m^3 s^{-1}$, with a range between a Q_{95} of 0.9 $m^3 s^{-1}$ and a Q_{10} of 11.4 $m^3 s^{-1}$. Water balance estimates suggest annual evaporation rates of ca. 450 mm. In addition to this site, the Scottish Environment Protection Agency (SEPA) also monitor flows for the 42 km^2 Charr catchment in the Water of Dye (Fig. 1a). Further flow records in the Water of Dye were also collected from Brocky Burn, where a flume and pressure transducer were established by the University of Aberdeen (Dawson, 1999). This gave accurate nested flow records for 233, 42 and 1.3 km^2 for the Feugh, Charr and Brocky Burn, respectively (Soulsby et al., 2003).

3. Methods

Samples of stream water for the 2001–2002 hydrological year were collected at approximately weekly intervals at six sites in the catchment (Fig. 1a). The availability of flow data for the three nested catchments at Brocky Burn (1.3 km^2), Charr (42 km^2) and

Heugh Head (233 km^2), provided a concentration of sampling sites down the Water of Dye sub-basin, which was further supplemented by Bogendreip (90 km^2) (Fig. 1a). A further two sampling sites were located on the other two sub-basins of the Feugh, the Aven and the Upper Feugh, in order to characterise their overall contribution to the isotopic signature of stream water leaving the catchment at Heugh Head (Fig. 1a). Catchment precipitation was sampled at approximately the same weekly intervals as stream water samples from a rain collector located in the Water of Dye catchment at Charr. These were then averaged over longer fortnightly to three-week intervals depending on precise sample timing in order to produce a more consistent, structured seasonal pattern to the data. This is consistent with other studies (e.g. Darling and Talbot, 2003) where high resolution sampling, particularly following minor precipitation events with extreme isotope signatures, disguised seasonal trends in isotopic composition which are more important when investigating annual time scales.

All samples were collected and stored according to standard procedures (cf. Clark and Fritz, 1997) and analysed at the Scottish Universities Environment Research Centre (SUERC) using a gas source isotope ratio mass spectrometer. Ratios of $^{18}O/^{16}O$ are expressed in delta units, $\delta^{18}O$ (‰, parts per mille) defined in relation to V-SMOW (Vienna standard mean ocean water). The analytical precision was ±0.1‰. Stream water samples were also analysed for Gran alkalinity by acidimetric titration to end points of pH 4.5, 4.0 and 3.0 as described by Soulsby et al. (2003).

Seasonal trends in $\delta^{18}O$ in precipitation and stream water were modelled using periodic regression analysis to fit seasonal sine wave curves to annual $\delta^{18}O$ variations in precipitation and stream water (cf. DeWalle et al., 1997), defined as:

$$\delta^{18}O = X + A[\cos(ct - \theta)], \quad (1)$$

where $\delta^{18}O$ is the modelled $\delta^{18}O$, X is the mean annual measured $\delta^{18}O$, A is the measured $\delta^{18}O$ annual amplitude, c is the radial frequency of annual fluctuations (0.017214 rad d^{-1}), t is the time in days after the start of the sampling period (1 October 2001), and θ is the phase lag or time of the annual peak $\delta^{18}O$ in radians.

To estimate mean residence times from these patterns, the commonly used exponential model was applied in which precipitation inputs are assumed to mix rapidly with resident water in the major soil water and groundwater catchment stores and an exponential distribution of residence times results (Maloszewski et al., 1983; Stewart and McDonnell, 1991). Thus, the decrease in amplitude of stream water outputs relative to precipitation inputs can be used as the basis for estimating mean residence time (Unnikrishna et al., 1995). The sine wave models fitted to input and output water δ^{18}O variations were used and and the mean residence time (T) of water leaving the system is calculated as:

$$T = c^{-1}[(Az2/Az1)^{-2} - 1]^{0.5}, \tag{2}$$

where $Az1$ is the amplitude of precipitation, $Az2$ is the amplitude of the stream water outputs and c is the radial frequency of annual fluctuations as defined in model (Eq. 1). Given the relatively simple nature of this model and the size and complexity of the Feugh catchment, the results can only be taken as preliminary estimates of mean residence times. Nonetheless, studies elsewhere have suggested that the model is likely to be useful for such a first approximation (Stewart and McDonnell, 1991; Uhlenbrook et al., 2002). Furthermore, the relatively coarse temporal and spatial sampling procedure precluded reasonable application of more complex residence time models (e.g. Kirchner et al., 2000).

4. Results

4.1. Seasonal variation in precipitation inputs

Precipitation inputs to the catchment show marked seasonal variation, with winter precipitation (November to April: mean −10.60‰) more ^{18}O-depleted than summer rainfall (May to October: mean −7.31‰) (Table 2). This follows the anticipated, approximately sinusoidal, seasonal pattern of precipitation δ^{18}O whereby winter months are dominated by colder northerly and easterly air masses that bring rain and snow which, due to low temperatures, is more ^{18}O-depleted (Fig. 2). By contrast, summer weather systems are mainly south-westerly in origin, resulting in more ^{18}O-enriched precipitation. Despite precipitation inputs to the Feugh generally following this seasonal pattern, it can be seen that the most ^{18}O-depleted period of precipitation occurred at the end of the winter months in February and March (Fig. 2). This was when the influence of colder weather systems was most sustained during the year compared with the general influence of the more variable weather systems earlier in the winter. Surprisingly though, precipitation inputs remained ^{18}O-depleted well into April when rising temperatures would normally be expected to lead to gradually more ^{18}O-enriched precipitation. However, the hydrological year of 2001–2002 was cooler and wetter than normal. Thus, this transition appears to occur very abruptly (Fig. 2), although the relative lack of rainfall/storm activity during April means that this shift is particularly emphasized by the data.

4.2. Stream water outputs

In comparison to precipitation inputs, stream water $\delta^{18}O$ is generally very damped, reflecting the influence of catchment processes in effectively mixing seasonally variable inputs (see Heugh Head response in Fig. 2). However, stream water $\delta^{18}O$ response for different sites exhibits notable differences, which in turn reflect important sub-catchment variation in hydrological behaviour (Table 2).

The most variable site in the catchment is the 1.3 km^2, peat-dominated Brocky Burn sub-catchment ($\delta^{18}O$ range 3.2‰: Table 2). Increasing scale downstream in the Water of Dye leads to a reduction in $\delta^{18}O$ range observed for the 42 km^2 Charr sub-catchment (range 2.3‰), with the downstream site at Bogendreip (90 km^2) displaying a further reduction in range (1.9‰). Beyond this, the overall $\delta^{18}O$ range measured over the year at the catchment outfall at Heugh Head (233 km^2) shows no difference to that measured at Bogendreip (1.9‰: Table 2). This is despite the influence of stream water inputs from the other two sub-basins, the Water of Aven (30 km^2) and the Water of

Feugh at Powlair (61 km^2). These exhibit the lowest overall range and variability in δ^{18}O over the year (ranges of 1.8 and 1.7‰, respectively: Table 2).

In addition to the annual range for each site, there are also notable differences in mean δ^{18}O. Brocky Burn has the most ^{18}O-enriched mean stream water overall (−8.52‰). The second highest mean δ^{18}O was observed at both Charr and Heugh Head, which show the same annual mean (−8.61‰: Table 2). Bogendreip exhibits more of an intermediate mean δ^{18}O (−8.82‰), whilst the Water of Aven and Powlair show the lowest, most ^{18}O-depleted means (−9.06 and −9.15‰: Table 2).

4.3. Hydrological controls on stream water δ^{18}O

Figure 4 shows the stream water δ^{18}O time series for the six sub-catchment sampling sites in the Feugh during the 2001–2002 hydrological year. As with the precipitation inputs, stream waters exhibit seasonal differences, being generally ^{18}O-depleted during the winter months when rainfall and snowmelt generate the highest flows. The effect of more ^{18}O-enriched precipitation is evident in summer stream water δ^{18}O. However, it is also notable that the most ^{18}O-enriched samples occur during the first month of the sampling period in association with the two largest flows sampled for the year (Fig. 2). Thus, in addition to the seasonal precipitation influence determining stream water δ^{18}O patterns on a catchment-wide basis, specific hydrological events (and therefore variability in sub-catchment hydrological behaviour) can also lead to differences in isotopic composition between sites. This can be most readily shown by comparing δ^{18}O with corresponding stream water alkalinity time series (Fig. 4).

Gran alkalinity has proven utility as a tracer, particularly in the UK uplands. It effectively distinguishes between low alkalinity high flows derived mainly from acidic, organic soil horizons which generate rapid overland flow or shallow sub-surface storm flow; and higher alkalinity water from lower soil horizons and/or groundwater which dominates base flows (Hill and Neal, 1997; Wade et al., 1999). As a result, Gran alkalinity can be seen to vary predictably with flow in the Feugh and its sub-catchments

(Fig. 3, but see Soulsby et al., 2003 for a full analysis). The availability of stream water alkalinity measurements sampled at the same time as $\delta^{18}O$ for each of the six sites therefore acts as a surrogate for flow (especially for ungauged sites), and provides insight into the role of different hydrological flow paths that will be affecting the observed $\delta^{18}O$.

It is apparent that there is a considerable amount of event-related variation in $\delta^{18}O$ from sample to sample for the majority of sites (Fig. 4). At the most variable site, Brocky Burn, the two highest $\delta^{18}O$ samples during the first month of sampling are particularly marked (−7.3 and −7.0‰ on 1 October 2001 and 22 October 2001: Fig. 4). These relate to two of the largest sampled flows of the year (as indicated by the lowest stream water alkalinity values), with the corresponding $\delta^{18}O$ displaying notable ^{18}O-enrichment over the intervening samples (Fig. 4). The same effect is observed at the other Feugh monitoring sites. Rather than these peaks relating closely to the timing of maximum precipitation $\delta^{18}O$ as might be expected (Fig. 2), the occurrence of stream water $\delta^{18}O$ maxima at the very start of the sampling year present some uncertainty as antecedent precipitation and stream water $\delta^{18}O$ are unknown. Furthermore, initial sampled precipitation $\delta^{18}O$ for October 2001 are not as enriched as stream water during this period (Fig. 2). It is likely that catchment runoff is dominated by the displacement of ^{18}O-enriched summer precipitation stored in the catchment prior to sampling, especially as these October events followed a six-week period with limited high flows. Tracer experiments during hydrological events (e.g. Sklash and Farvolden, 1979) commonly show such displacement of "old" pre-event by "new" precipitation.

After this initial period of intense variation, stream water $\delta^{18}O$ was generally more predictable. As anticipated, the small, peat-dominated Brocky Burn shows the most marked response to more depleted winter precipitation events with 3 particularly ^{18}O-depleted samples between January and March 2002 (Fig. 4). The first two of these occurred in January following on from notable snowfalls in the latter half of December. However, the most ^{18}O-depleted precipitation samples do not occur until the more prolonged colder weather systems of February and March (Fig. 2), with the lowest stream water $\delta^{18}O$ sample at Brocky Burn subsequently occurring during this period (5 March 2002, −10.2‰: Fig. 4). Stream water $\delta^{18}O$ then exhibits a recovery through the remainder of March and then April. Although precipitation was still relatively ^{18}O-depleted over this period, it was not reflected in stream water $\delta^{18}O$ due to the rainfall totals being low; and increasing evaporation leading to catchment drying, as seen from the gradual increase of stream water alkalinity to near base flow levels at all sites through to around the end of May (Fig. 4). Stream water $\delta^{18}O$ during the summer months at Brocky Burn exhibits a rapid response to more ^{18}O-enriched summer precipitation. In particular, there are two periods of high flow in the summer at the start of June and end of July (where alkalinity is seen to decrease significantly), which result in relatively sustained increases in stream water $\delta^{18}O$ (compared with downstream sites) over more stable base flow conditions for the intervening samples (Fig. 4).

The general seasonal pattern of stream water $\delta^{18}O$ response observed at Brocky Burn is replicated at the increasing downstream scales of Charr, Bogendreip and Heugh Head, albeit in a more damped manner (Fig. 4). The stream water $\delta^{18}O$ time series for the Water of Aven and the Water of Feugh at Powlair, however, as well as being generally more ^{18}O-depleted, are also notably less varied in terms of response to short-term hydrological variation. The damped $\delta^{18}O$ output for the Water of Aven initially appears surprising, given its relatively high peat coverage (56% cf. Charr 66%: Tables 1 and 2). The extensive erosion of the blanket peat in the Aven probably leads to more significant recharge of groundwater and therefore greater mixing of precipitation inputs (Boorman et al., 1995). However, it is also possible that this reflects the effect of the more freely draining mineral soils that cover the steeper slopes of the catchment as well as the significant alluvial deposits at the base of the catchment as it emerges from its incised valley. In contrast, Charr only really displays relatively confined valley bottom alluvial deposits suggesting that there is more mixing of groundwater in the Aven to dampen variation in stream water $\delta^{18}O$. Previous studies using alkalinity-based end member mixing to perform hydrograph separations in the catchment have suggested this to be the case (Soulsby et al., 2003, 2004[1]). A similar influence is observed for the most damped $\delta^{18}O$ time series for the Water of Feugh at Powlair (Fig. 4), given that this is the sub-catchment where the influence of freely draining humus iron podzols and valley bottom alluvial aquifer deposits is most significant (Fig. 1, Table 1).

These $\delta^{18}O$ and alkalinity variations in stream water can be viewed conceptually as the combination of two components: a relatively stable base flow end member and a seasonally variable storm flow end member. This conceptualisation is consistent with the two-component end member mixing previously used to assess the hydrology of the Feugh based on alkalinity data alone (Soulsby et al., 2003, 2004[1]). Figure 5 shows this relationship more clearly, presenting seasonally differentiated $\delta^{18}O$-alkalinity mixing plots. As in Fig. 4, alkalinity measurements are used to provide a more direct indication of hydrological sources affecting measured stream water $\delta^{18}O$. Theoretically, the influence of seasonally variable precipitation inputs should result in an approximately triangular shaped plot of $\delta^{18}O$ and alkalinity measurements comprising a low alkalinity, seasonally variable storm flow end member (with low $\delta^{18}O$ during winter and higher $\delta^{18}O$ during summer), which mixes with higher alkalinity base flow waters with more stable, intermediate $\delta^{18}O$. At most sites this conceptual structure is apparent, although there are significant inter-site differences.

As expected, the most responsive site at Brocky Burn shows the clearest seasonally differentiated $\delta^{18}O$ pattern (Fig. 5). Sites where there is less distinction between summer and winter mixing lines implies a greater mixing of source waters and this is most evident for Powlair and the Aven given their more damped $\delta^{18}O$ variability observed in Fig. 4. The expected downstream increase in the mixing of sources with scale is apparent from Brocky Burn to Charr to Bogendreip and Heugh Head (Fig. 5). Despite this, Heugh Head nonetheless displays considerable scatter due to the contrasting isotopic signature of sub-catchment drainage that it integrates. These mixing plots also illustrate how constrained sub-catchment base flows (highest alkalinities) are in terms of $\delta^{18}O$ variation. The least variable Powlair and Aven sites unsurprisingly display the most constant $\delta^{18}O$ at lower flows, whereas the lowest flow (highest alkalinity) samples for Brocky Burn exhibit quite notable seasonal differences $\delta^{18}O$ as was also suggested

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from the time series in Fig. 4. This can probably be attributed to limited groundwater storage in such a small headwater catchment, which contributes groundwater that is far more seasonally variable than larger sub-catchments where groundwater storage is more extensive and well mixed. Base flow δ^{18}O at Charr appears to be reasonably well defined but variation increases downstream at Bogendreip, and then further at Heugh Head, reflecting the greater mix of isotopic signatures that it receives from the three sub-basins.

4.4. Seasonal analysis of δ^{18}O patterns: preliminary estimate of mean residence times

The seasonal δ^{18}O trends observed in precipitation and stream water were interpreted more quantitatively by use of periodic regression analysis to fit seasonal sine wave models to annual δ^{18}O time series (Fig. 6). The modelled curves, in particular for precipitation, oversimplify the patterns of variation evident in the raw data and this has a subsequent impact on the strength of correlations between observed and modelled δ^{18}O for most sites (i.e. r^2=<0.50). However, all the results are statistically robust (p=<0.02), and the level of agreement is generally comparable with results from similar studies (e.g. DeWalle et al., 1997; McGuire et al., 2002; Soulsby et al., in review[1]). Furthermore, it should be noted that this type of analysis is often based on monthly data, where lower variability leads to better "fit" using a simplistic annual model structure.

Figure 6 shows the fitted models for annual δ^{18}O variation in precipitation and stream water. Precipitation inputs are only relatively crudely described using the seasonal sine wave model (r^2=0.34) and the modelled amplitude of seasonal δ^{18}O variation is significantly reduced as a result. This appears to be a factor of the particularly rapid transition from the most ^{18}O-depleted winter/spring precipitation to more ^{18}O-enriched inputs early in the summer, as well as the generally scattered inputs over the first 4 months of the year (Fig. 6). It was therefore considered appropriate to use an additional optimised precipitation model weighted to the seasonal extremes in precipitation δ^{18}O, providing a larger model amplitude of 3.48‰ (dashed regression curve; Fig. 6). This could then be used to provide a better upper estimate of mean residence time.

In terms of stream water sampling sites, those with the least variable δ^{18}O stream water (the Aven and Powlair) are those that are least well described by the seasonal sine wave model (r^2=<0.3: Fig. 6). In contrast, modelled δ^{18}O for the remaining four sites at Brocky Burn, Charr, Bogendreip and Heugh Head show generally stronger predictions and larger annual δ^{18}O amplitude values. In line with the general annual variability observed from Table 2, the significant downstream increase in modelled δ^{18}O annual amplitude from the headwater scale of Brocky Burn (1.3 km^2) to the sub-catchment scale of Charr (42 km^2) has the most significant impact on modelled amplitudes (0.73‰ to 0.39‰). This is presumably in response to the added influence of valley bottom alluvial deposits that facilitates mixing with older groundwater sources. However interestingly, at the base of the catchment at Heugh Head, despite the much larger catchment size and a more significant alluvial aquifer, only a minimal further decrease in the amplitude of modelled stream water δ^{18}O is produced (0.37‰: Fig. 6). This is despite the further damped, low amplitude influence of flows from the other Water of Aven and Water of Feugh sub-basins.

The model described by Eq. (2) was used to translate the results into estimates of mean stream water residence time (Table 3). These provide a very general, but useful, indication of the degree of mixing of hydrological sources in each sub-catchment and thus offer a valuable integrated assessment of the differences in runoff processes in the Feugh catchment. It is interesting to note that the substantially longer residence times for the Water of Aven and Water of Feugh at Powlair (1.40–2.40 and 1.69–2.91 years) coincide with generally more depleted mean δ^{18}O stream water than for the other sites (Table 3). This probably reflects the greater influence of depleted winter precipitation inputs, implying that the more extensive coverage of freely draining soils in these sub-catchments are responsible for greater recharge to groundwater stores and therefore longer travel times, particularly during the wettest times of the year. This is also implied by the greater groundwater contributions to annual runoff, estimated by chemically based hydrograph separations (Soulsby et al., 2003), which show greater groundwater contributions in these sub-catchments (Table 3).

In the Water of Dye by comparison, the high peat coverage at Brocky Burn and Charr are likely to produce more marked storm runoff response and therefore shorter travel times during these wettest periods of the year, allowing comparatively less recharge to groundwater and lower groundwater contributions to flow (Table 3). Recharge of catchment storage in these peat dominated headwaters of the Feugh may therefore be more likely to show a bias towards higher δ^{18}O summer precipitation, when generally drier conditions lead to comparatively longer travel times for precipitation inputs than during the wetter times of the year. The shortest estimated residence time for Brocky Burn (0.41–0.74 years) therefore highlights this overall effect. This is clearly influential at Charr as well, but the larger scale and increased valley bottom storage in alluvium provides greater potential for older water (longer hydrological pathways) to contribute to runoff through the year (mean residence time 0.81–1.41 years). Estimated residence times are again similar for Bogendreip and Charr owing to their general similarity in annual δ^{18}O variability (Table 3). However, the responsive upstream peat influence from Charr appear to mask the potential influence of older waters at the larger scale of Bogendreip, given the more ^{18}O-depleted mean stream water observed here.

This is also reflected in the estimated residence time for the base of the Feugh at Heugh Head (0.86–1.49 years), which continues to reflect the importance of rapid storm runoff response from headwater peat at this much larger scale. It appears that the influence of the larger groundwater contributions in the lower catchment (54.6% of annual flows) coupled with more mixed, longer residence time waters from Powlair and the Aven does not result in a marked increase in estimated mean residence time.

4.5. Influence of catchment characteristics on hydrology

The relationships between estimated mean residence times and catchment landscape controls were examined in a more formal manner by simple linear regression (Table 4 and Fig. 7). Sub-catchment soil cover had a dominant effect, with percentage cover of responsive peat soils exhibiting a strong negative correlation with mean residence time. Similarly, the percentage coverage of more freely draining podzolic and alluvial soils was positively correlated with mean residence time, a relationship also reflected by a strong positive correlation with mean catchment slope (which strongly influences soil distribution).

Mean catchment residence times were also strongly correlated with percentage groundwater contribution to annual runoff in each sub-catchment (which is tabulated in Table 3) from the earlier work by (Soulsby et al., 2003). Unsurprisingly the percentage groundwater contribution is also strongly correlated with soil cover and hillslope gradient. Simply stated, higher peat coverage on flatter catchment interfluves results in rapid hydrological responses to precipitation, leading to reduced recharge, lower groundwater contributions to baseflows and shorter residence times. Higher coverage of freely draining podzols on steeper hillslopes or alluvium in valley bottom areas increases recharge, produces higher groundwater contributions to annual flow and longer residence times. It appears that landscape organisation and the combination of soil/topographic units in different sub-catchments, rather than scale alone has the strongest influence on the hydrological characteristics of flow path partitioning and mean residence times.

5. Discussion

These results contribute to an improved understanding and conceptualisation of catchment hydrology for the Feugh, previously based on geochemical tracer analysis (Soulsby et al., 2003, 2004[1]). Variation in stream water δ^{18}O is generally consistent with relatively simple two-component mixing, where well-mixed, longer residence time groundwater sustains base flows and more recent, seasonally variable precipitation inputs in soil waters mainly account for storm flow response. Over the course of the hydrological year, this mixing process resulted in a reasonably well defined, seasonally evolving isotopic signature that reflects important differences in sub-catchment hydrological processes, and allows intra-catchment differences in stream water residence times to be estimated.

The results provide interesting insights for current understanding on the scaling and integration of hydrological processes in larger catchments. In particular, the results for the largest scale at Heugh Head indicated that the hydrological responsiveness of headwater peat soils (in the Water of Dye) exert the dominant influence on the overall seasonal patterns and residence times observed at the larger catchment scale, despite significant downstream groundwater inputs and mixing with more constant δ^{18}O signatures in more groundwater dominated sub-basin drainage. This displays parallels with recent findings from the similar sized Feshie catchment in the Cairngorm Mountains of Scotland (Rodgers et al., 2005; Soulsby et al., 2004[1]). However, residence times in the Feshie ranged from 3–6 months in more responsive catchments, to 12–15 months in more groundwater dominated catchments. Mean residence time at the catchment outfall (230 km^2) were 4–7 months. These shorter residence times could reflect the higher precipitation levels in the Feshie and the more mountainous terrain.

The results from the Feugh also bear interesting comparison with the findings of other tracer studies; though different sampling strategies and analytical approaches mean that comparisons are semi-quantitative. The flashy, responsive nature of the Feugh to rainfall and snowmelt indicates the importance of much shorter residence time in the catchment soils in headwaters like Brocky burn. Earlier isotope work in the Allt a' Mharcaidh in the Feshie catchment indicated mean residence times of water in peaty soils could be as little as 2 months (Soulsby et al., 2000). Similarly, others, such as Robson et al. (1992) at Plynlimon in Wales and Nyberg et al. (1999) in Sweden, have used tracer data to imply very short residence times for responsive peaty soils in generating storm runoff. These studies showed that although tracer breakthrough to streams could occur in a matter of minutes or hours, catchment soils still stored significant tracer quantities after a period of a few months.

The importance of groundwater contributions to flow in mountainous environments has increasingly been highlighted in Scotland (e.g. Soulsby et al., 2004[1]) and elsewhere. The results of this study indicate baseflow mean residence times of several years for parts of the Feugh catchment. Similarly, Uhlenbrook et al. (2002) showed that shallow and deep groundwater, respectively accounted for 69% and 20% of annual runoff in 40 km^2 Brugga catchment in the Black Forest of Germany. These shallow and deep groundwater sources were each estimated as having mean residence times in the ranges of 2–3 and 5–10 years. Similar residence times have also been estimated in for baseflows, borehole waters or springs in upland environments as different as Plynlimon, Wales (Haria and Shand, 2004); Maimai in New Zealand (McGlynn et al., 2003); pre-Alpine catchments in Switzerland (Vitvar and Balderer, 1998); the Bavarian Alps, Germany (Maloszewski et al., 1983); and the Catskills, USA (Vitvar et al., 2002). Whilst the mean residence times presented in this study do not give direct groundwater residence times, earlier work by Soulsby et al. (1999, 2000) in the Cairngorms made a preliminary estimate of mean residence times for shallow and deeper groundwater sources of 2 and >5 years, respectively. All these studies strongly suggest the presence of long tails in residence time distributions in such mountainous catchments (Kirchner et al., 2000).

Ultimately, it should be stressed that the residence time estimates presented in this study are mean estimates. In reality, catchment runoff is composed of a much wider and more complex range of internal catchment residence time distributions that are currently unknown (Kirchner et al., 2000, 2001). Future work in the Feugh would therefore benefit from direct assessment of different groundwater and soil water stores that are likely to be highly variable (Frederickson and Criss, 1999; Gonfiantini et al., 1998). These could be assessed indirectly through more intensive sampling of stream base flows or possibly using other tracers such as tritium (cf. McGlynn et al., 2003) or CFCs (cf. Uhlenbrook et al., 2002). Moreover, further insights would be gained for improved spatial and temporal resolution of precipitation and stream water samples, which is a key objective in future work.

The results nonetheless highlight the pragmatic utility of stream water oxygen isotope measurements as an analytical tool in the study of mesoscale catchments given that they effectively integrate the influence of these complex catchment heterogeneities as well as indicating the relative importance of different sources in runoff production. This further suggests that the potential of such an approach to improve current understanding of scaling in catchment hydrological processes remains largely underdeveloped (Brown et al., 1999; Genereux and Hooper, 1998; McDonnell et al., 1999, Uhlenbrook et al., 2002). It is important, therefore, that tracer studies such as these are continued in order to refine our understanding of flow paths and residence times, and to help structure and validate more accurate hydrological models.

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Table 1. Characteristics of the Feugh catchment.

			Geology				Soils				Land Use		
	Area	Mean altitude	Granite	Semi-pelite	Psammite	Alluvial	Peat	Peaty Podzol	Humus Iron Podzol	Alluvial	Woodland	Moorland / Peat	Grassland
	km^2	m	%	%	%	%	%	%	%	%	%	%	%
1. Brocky Burn	1.3	419	100	0	0	0	84.1	15.9	0	0	0	100	0
2. Charr	41.8	420	73.3	21.5	3.2	1	65.9	34.1	0	0	0	99.4	0.6
3. Bogendreip	90.1	357	80.2	12.3	2.8	4.2	48.5	38.0	13.2	0	22.1	72.4	4.2
4. Aven	30.1	427	99.2	0	0	0.8	55.7	30.3	13.2	0.8	5.6	92.7	1.6
5. Powlair	61.1	356	86.5	0	5.0	8.4	19.4	38.8	36.9	4.9	10.1	74.2	9.5
6. Heugh Head	233	329	78.5	4.8	8.5	7.9	32.1	34	26	7.9	18.1	68.2	10.7

Table 2. Arithmetic mean, range and standard deviation of $\delta^{18}O$ (‰) in the Feugh catchment (1 October 2001–30 September 2002).

	Mean	Minimum	Maximum	Standard deviation
Inputs:				
Winter/spring precipitation	−10.60	−13.5	−8.4	2.29
Summer/autumn precipitation	−7.31	−10.3	−5.1	1.80
Stream water:				
1. Brocky burn	−8.52	−10.2	−7.0	0.72
2. Charr	−8.61	−9.5	−7.2	0.46
3. Bogendreip	−8.82	−9.7	−7.8	0.43
4. Aven	−9.06	−9.8	−8.0	0.31
5. Powlair	−9.15	−10.0	−8.3	0.29
6. Heugh head	−8.61	−9.4	−7.5	0.42

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Table 3. Mean and amplitude of modelled $\delta^{18}O$, approximate mean residence times and estimated groundwater flow contributions for sub-catchment sites in the Feugh (2001–2002). Mean residence times range from a minimum estimate derived from the un-weighted precipitation model, to a higher estimate based on more typical seasonal extremes. Groundwater proportions based on annual two-component mixing analysis using Gran alkalinity (see Soulsby et al., 2004).

	Modelled Mean (‰)	Amplitude (‰)	Mean residence time (years)	% Groundwater contribution to annual flow
Inputs:				
Precipitation	−8.86	2.03		
Weighted precipitation	−9.36	3.48		
Stream water:				
1. Brocky Burn	−8.55	0.73	0.41–0.74	26.1
2. Charr	−8.60	0.39	0.81–1.41	38.4
3. Bogendreip	−8.82	0.40	0.79–1.38	36.6
4. Aven	−9.06	0.23	1.40–2.40	42.3
5. Powlair	−9.15	0.19	1.69–2.91	54.7
6. Heugh Head	−8.62	0.37	0.86–1.49	54.6

Table 4. Coefficients of variation r^2 and trends (negative or positive) between controlling catchment variables and both mean water residence times and groundwater contribution.

	Mean residence times		Groundwater contribution	
	r^2	trend	r^2	trend
Area	0.00	–	0.43	pos
Topography				
Mean elevation	0.02	neg	0.42	neg
Min elevation	0.31	neg	0.50	neg
Max elevation	0.24	pos	0.39	pos
Mean slope	0.66	pos	0.54	pos
Max slope	0.44	pos	0.66	pos
Drainage density	0.36	pos	0.70	pos
Soils				
Peat	0.52	neg	0.87	neg
Peaty podzol/humus iron podzol	0.35	pos	0.49	pos
Alluvial/humus iron podzol	0.47	pos	0.83	pos
Responsive soils*				
Freely draining soils**	0.52	pos	0.87	pos
Land use				
Heather/peatland	0.11	neg	0.50	neg
Coniferous woodland	0.01	pos	0.20	pos
Grassland	0.21	pos	0.79	pos
Geology				
Granite	0.01	pos	0.14	neg
Alluvial	0.22	pos	0.73	pos
Groundwater contribution/residence times	0.50	pos		

* Peat

** Humus iron podzol/peaty podzol and alluvial/humus iron podzol

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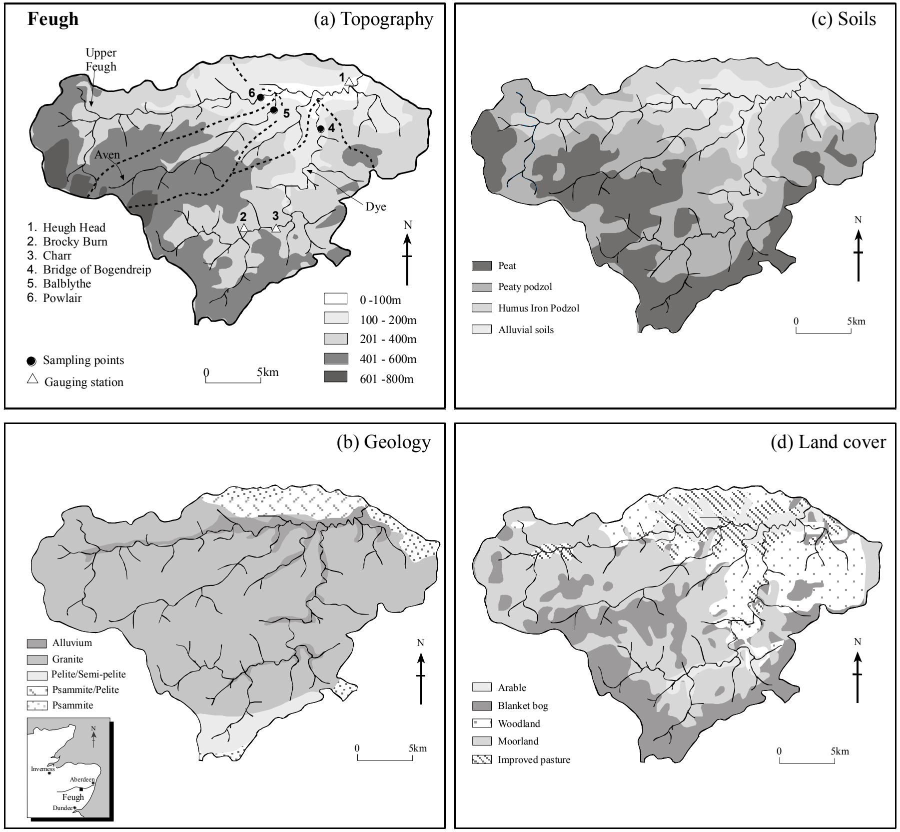

Fig. 1. Catchment maps of the Feugh, showing **(a)** topography and monitoring network, **(b)** geology, **(c)** soil coverage and **(d)** land cover.

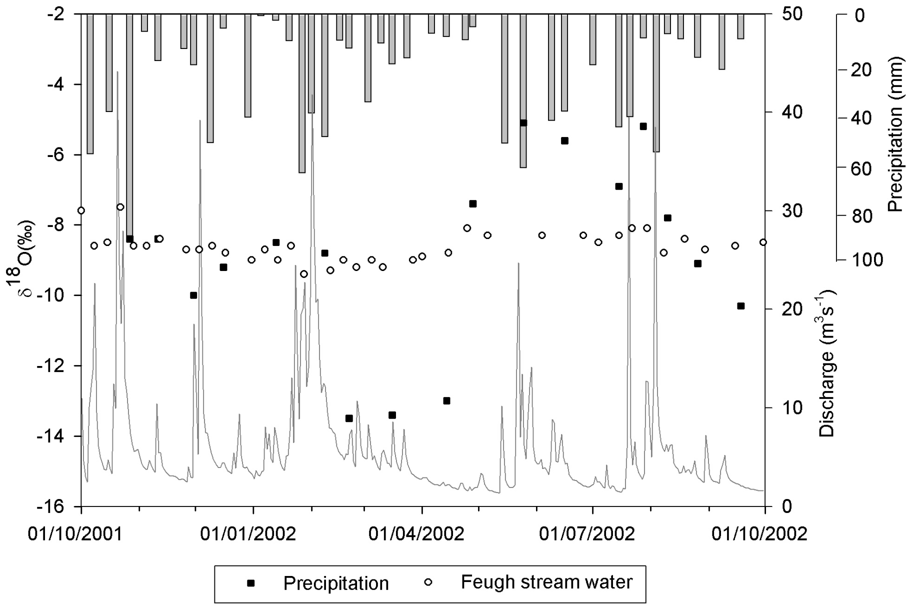

Fig. 2. Temporal variation in precipitation and stream water $\delta^{18}O$, annual run-off and rainfall for the Feugh catchment (1 October 2001–30 September 2002).

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Stable isotope tracers and mesoscale catchment controls

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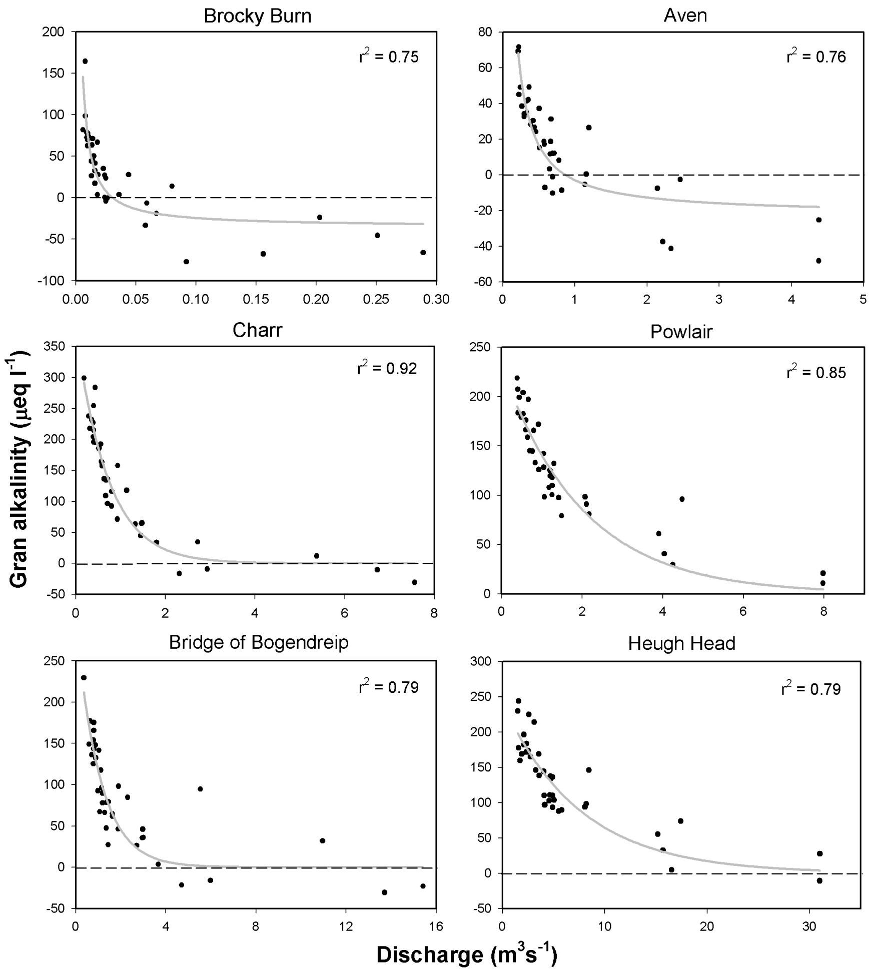

Fig. 3. Alkalinity variation with flow for sub-catchment sampling sites in the Feugh.

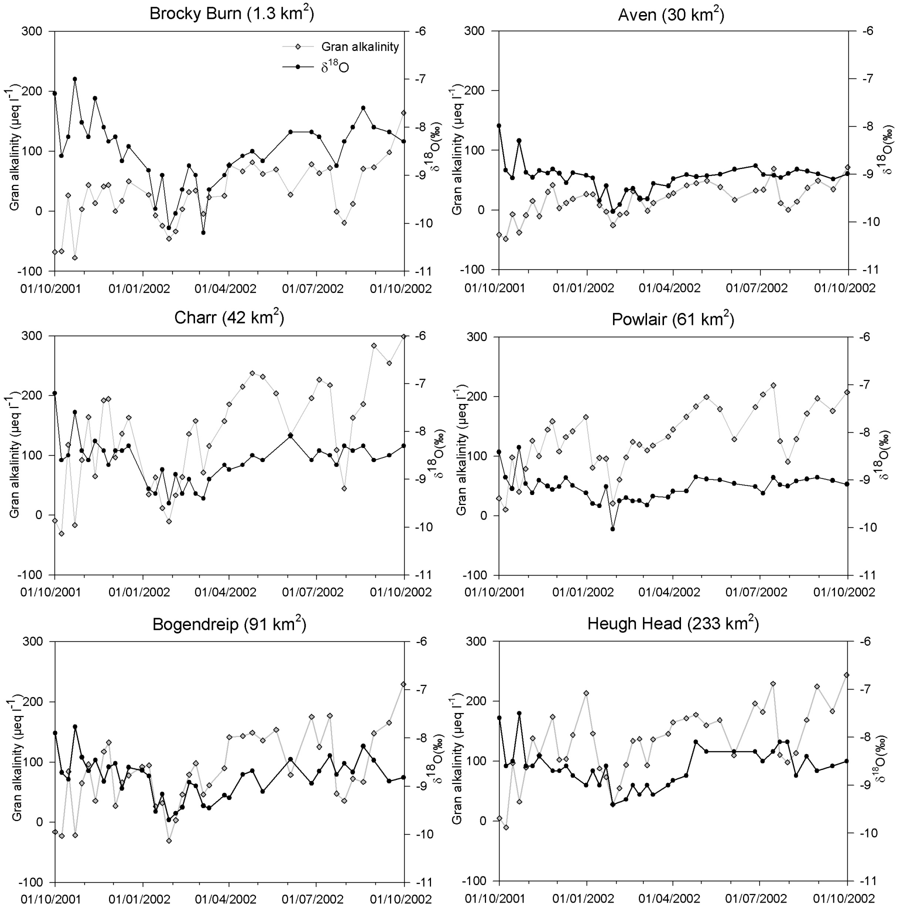

Fig. 4. Temporal co-variation in stream water δ^{18}O and alkalinity for Feugh sub-catchments.

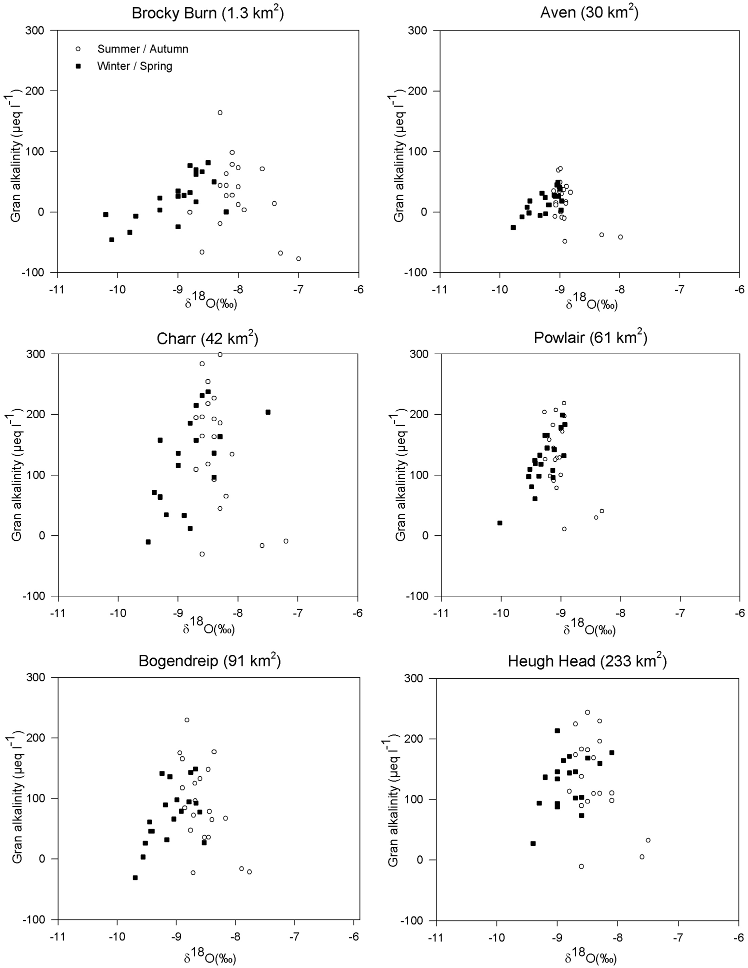

Fig. 5. Mixing plots for stream water δ^{18}O, showing seasonal and flow (alkalinity) related variation.

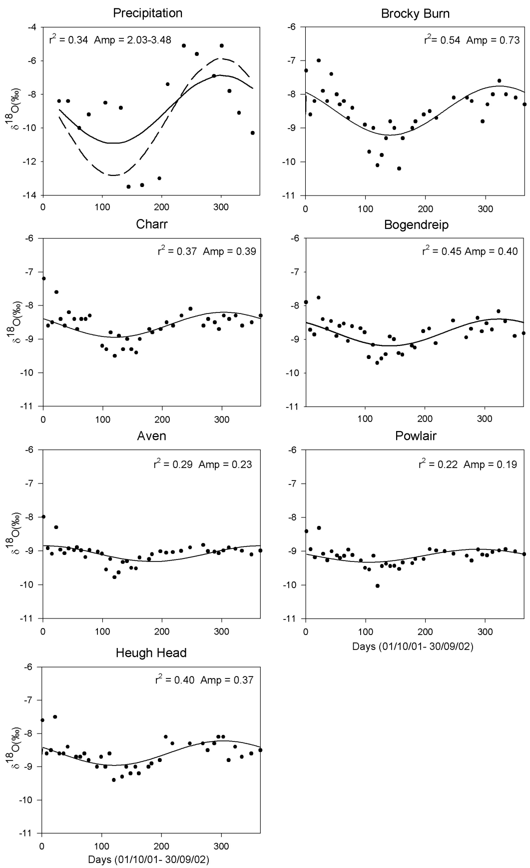

Fig. 6. Fitted annual regression models to δ^{18}O for precipitation and stream water in the Feugh. Precipitation model represented by the solid regression line is based on the raw data series while the dashed regression line is fitted to an optimised data set to represent the full seasonal variability in sampled precipitation.

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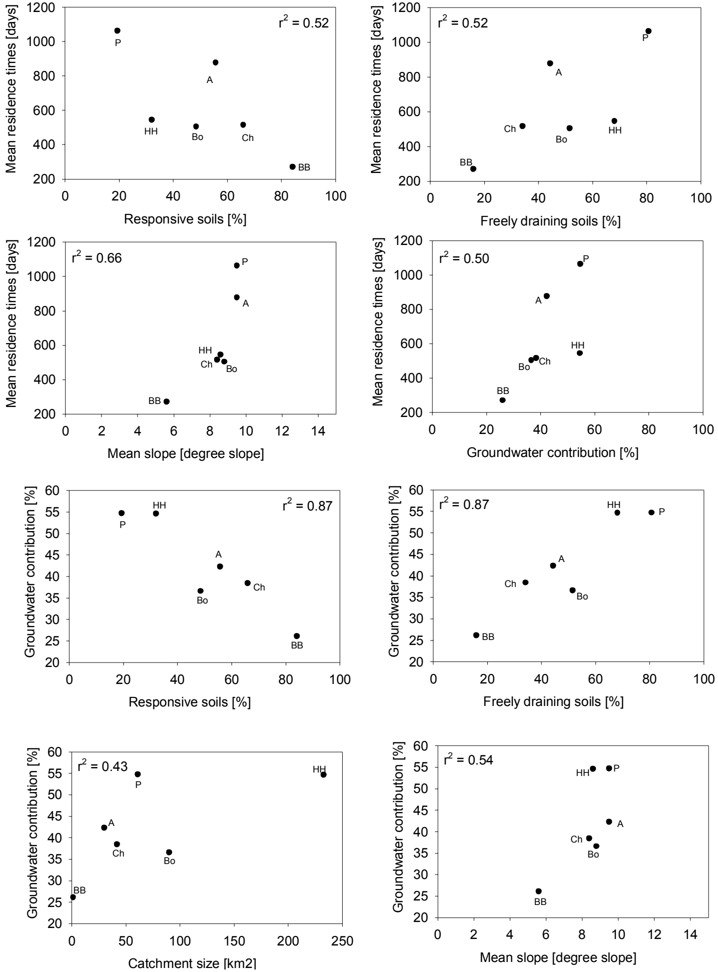

Fig. 7. Selected relationships between catchment characteristics and mean residence time and percentage groundwater contributions to flow.