| 2  | Controls on the spatial and temporal variability of                                       |  |  |  |  |  |  |  |  |
|----|---|--|--|--|--|--|--|--|--|
| 3  | <sup>222</sup> Rn in riparian groundwater in a lowland Chalk                              |  |  |  |  |  |  |  |  |
| 4  | catchment   |  |  |  |  |  |  |  |  |
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## 1 **Abstract**

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Radon is a powerful tracer of stream-aquifer interactions. However, it is important to consider the source and behaviour of radon in groundwater when interpreting observations of river radon in relation to groundwater discharge. Here we characterise the variability in groundwater radon concentrations in the riparian zone of a Chalk catchment. Groundwater <sup>222</sup>Rn (radon) concentrations were determined in riparian zone boreholes at two sites in the Lambourn catchment, Berkshire, UK, over a two year period. In addition, borehole core material was analysed for <sup>226</sup>Ra (radium) and to determine radon emanation. Radon and radium concentrations and radon emanation were found to change with depth and temporal variations in groundwater radon concentrations were found at different scales. The abundance of radium and emanation of radon increased nearer the surface leading to greater groundwater radon concentrations. It is shown that seasonal changes in water table elevation can to lead to variable radon concentrations in groundwater as zones of radon production become hydraulically active. Groundwater radon concentrations in shallow piezometers were found to respond to both seasonal changes in the water table and individual rainfall events. Riparian sources of radon can be variable and are therefore potentially influential in the radon signals observed in rivers and should be properly characterised when interpreting river radon inputs.

## 1. Introduction

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Groundwater-river water interactions are of great importance in groundwater dominated catchments such as those found in Chalk areas. The conditions under which groundwater will maintain river flow and the interactions between these bodies of water must be understood to allow effective catchment management. Groundwater makes up one third of public water supplies in England and Wales, with approximately half of this coming from Chalk aquifers (Downing, 1993). The importance of Chalk catchments for water supply, as well as their ecological and amenity value, means that competing pressures on groundwater abstraction and river flows must be balanced. Tracers are an important tool in the study of interactions between ground and river waters and enable the understanding of transport and mixing processes between water bodies. Tracers allow the identification and quantification of flow paths and estimation of the mixing ratios of water bodies, if sufficiently different chemical signatures can be identified for each end member. Radon is a radioactive noble gas produced by the decay of radium that has been widely used as a tracer in groundwater studies. There are three naturally occurring isotopes of radon: <sup>219</sup>Rn, <sup>220</sup>Rn and <sup>222</sup>Rn. Here we are concerned only with the <sup>222</sup>Rn isotope, as the others have short half lives ( $t_{1/2}$  < 1 minute) which preclude them from the methods of analysis used here.  $^{222}$ Rn ( $t_{1/2} = 3.82$  days) is the daughter of  $^{226}$ Ra and both are members of the <sup>238</sup>U decay series. From here on the term 'radon' refers solely to the <sup>222</sup>Rn isotope. The terms 'parent' and 'daughter' are used to describe the decaying and the resulting nuclides respectively. Uranium is present in different mineral phases of all rock types at varying concentrations (Ball et al., 1991). Differences in the abundance and distribution of

minerals, and therefore uranium and radium, lead to differences in radon production and emanation between geological units. Emanation describes amount of radon produced by decay of radium released from a material, in this case that which escapes the rock matrix into solution in the saturated zone of an aquifer. Emanation may be referred to as a rate if measured as the activity of radon released from a material per unit time or as a coefficient, which reflects the proportion of radon produced by radium that is released from a material. Radon emanation and the processes involved are discussed comprehensively in Osmond and Ivanovich (1992) and Porcelli and Swarzenski (2003). Radon is chemically and biologically inert and so in the saturated zone of an aquifer there are no sources other than the decay of <sup>226</sup>Ra and no sinks other than its own radioactive decay. The chemical and physical characteristics of radon make it a potentially powerful tracer in the study of groundwater-river water interactions. Radon has several advantages over other natural and anthropogenic tracers. As a gas, radon is readily lost from river water to the atmosphere, where concentrations are negligible by comparison. The enrichment of radon in groundwater with respect to surface waters means that it is easily detectable in zones of groundwater discharge and subsequent degassing to the atmosphere allows the detection of successive groundwater inputs to a river. Sample collection and analysis requires relatively simple field and laboratory techniques and analysis by liquid scintillation spectrometry can be automated allowing for high sample throughput (Pates and Mullinger, 2007). In a closed system, <sup>222</sup>Rn will reach radioactive equilibrium with its parent <sup>226</sup>Ra. Radioactive equilibrium occurs when the activity of the daughter nuclide is equal to that of its parent, which is reached after approximately five daughter half-lives. The

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3.82 day half life of radon means that equilibrium with a radium source will be achieved in approximately 20 days. Previous studies of radon in groundwaters have shown that its presence in solution is usually in excess of any dissolved radium (summarised in Osmond and Cowart, 1992). Therefore, the majority of dissolved radon must have been produced by decay of solid-phase radium and then released into groundwater via emanation processes. Due to radon's short half-life and the tendency for concentrations to evolve towards equilibrium with its parent, we can assume that radon in groundwater is mostly derived from the recent flow path and not solution phase radium from historic flow paths. Radon is, therefore, indicative of the immediate geological environment unlike other solutes which may be transported over greater distances. Radon has been applied to the tracing of groundwater inputs into rivers and oceans (e.g. Burnett et al., 2006; Cook et al., 2003; Genereux et al., 1993; Hamada, 1999; Lee and Kim, 2006; Swarzenski et al., 2007) and in investigations of groundwater-river water interactions (e.g. Bertin and Bourg, 1994; Hoehn and von Gunten, 1989; Macheleidt et al., 2002; Schubert et al., 2004). These studies have usually sampled a number of groundwater monitoring wells in a catchment or aquifer and utilised either the mean or most reasonable single values as representative of catchment waters, i.e. a constant source term to the river is assumed (Cook et al., 2003; Ellins et al., 1990; Genereux and Hemond, 1990). Cook et al. (2006) took account of spatial variability of radon production within a river reach due to changes in geology while Genereux and Hemond (1990) distinguished between vadose zone water and saturated zone water from the soil and bedrock horizons. Recent work (Mullinger et al., 2007) has shown that temporal as well as spatial variation of radon in groundwater inputs can occur. Mullinger et al. used radon to

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develop an understanding of some of the controls on groundwater inputs to the Pang and Lambourn Chalk catchments in South-East England. In the Pang catchment temporally variable radon concentrations in spring discharges exerted a strong influence over river radon concentrations. In the Lambourn, the modelled radon concentration in groundwater entering the river varied over time and appeared to reflect local flow accretion. Grapes et al. (2006) observed that river stage in the Lambourn was closely related to local groundwater levels and the Lambourn's alluvial aquifer. The water table immediately adjacent to the river was generally 0.2-0.4 m higher than the river stage, with no flow in the river when the water table falls below the level of the river bed (Grapes et al., 2005). Changes in radon inputs are thought to be indicative of connectivity with the alluvial aquifer located in the river valley. Therefore, it is hypothesised that changes in the level of the local water table have an impact on the source of radon being supplied to the river in the Lambourn. This hypothesis is the focus of the present study. Groundwater studies of radon in the Chalk of the UK have previously focused on deep groundwaters. Sampling has typically been from public supply boreholes for the purpose of investigating bulk aquifer properties (e.g. Low, 1996; Younger and Elliot, 1995). Observations by Low at public supply boreholes in the Chalk of East Anglia, UK showed temporal variations in groundwater radon concentrations of up to 3 Bg 1<sup>-1</sup> in individual boreholes and a range of 3.5 to 9 Bq l<sup>-1</sup> across the different sites. A large recharge event was shown to significantly lower groundwater radon concentrations across several boreholes. Low suggests that lower radon concentrations occur during times of recharge. Another example of temporal changes in groundwater radon concentrations is found in a study of spring water derived from the Mendip Hills near Bristol (Andrews

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1 and Wood, 1972). Andrews and Wood (1972) observed generally higher radon

concentrations during periods of higher rainfall in water from two springs discharging

3 from the Carboniferous Limestone of the Mendips. The studies by Low (1996) and

Andrews and Wood (1972) show that, despite differences in the systems being

observed, temporal changes in groundwater radon can exist under different

6 hydrological conditions.

The source and behaviour of radon in groundwater are important factors in interpreting observations of river radon in relation to stream-aquifer interactions. In this paper we concentrate on the spatial and temporal variations that exist in the groundwater system only. The aims of this work are to characterise spatial variability in the sources of radon relevant to groundwater-river interactions, i.e. the groundwater and aquifer immediately adjacent to the river, to understand temporal changes in groundwater radon in this zone and to add to the relatively sparse data that exits on

## 2. Site description

radon in Chalk catchments.

The Lambourn is a groundwater dominated Cretaceous Chalk catchment (Bradford, 2002) located in West Berkshire, UK (Fig. 1). The outcropping solid geology is almost entirely Chalk with minor Palaeogene deposits overlying the lower catchment and significant drift deposits in the river valley (Allen et al., 1997).

Groundwater dominated catchments are often, but not exclusively, characterised by stable flow conditions with a high base flow index as well as stable temperature and chemistry signals (Sear et al., 1999). The base flow index in the Lambourn is 0.96 (Grapes et al., 2005) and Neal et al. (2004) observed stable Ca, Mg and Sr profiles along the length of the perennial river channel. Stable physical and chemical

1 parameters in catchments of this type mean that many natural tracers are of limited 2 use due to the homogeneity of water in the aquifer and river. As well as its importance 3 as a water source, the River Lambourn is of high ecological value as recognised by its 4 designation as a Site of Special Scientific Interest. The Lambourn catchment itself has a topographical catchment area of 234 km<sup>2</sup> (Griffiths et al., 2006) and is a tributary of 5 6 the River Kennet, which in turn flows into the River Thames. 7 This paper focuses on the results of investigations carried out at two borehole 8 installations in the Lambourn valley at Maidencourt Farm (MCT) and East Shefford 9 (ESF) (Fig. 1). The study sites are located approximately halfway down the length of 10 the catchment, where boreholes with nested piezometer arrays have been drilled in the 11 riparian zone. Drilling took place in June 2004 and at each site five boreholes were 12 installed, each with two piezometers screened at different depths. Inset diagrams in 13 Fig. 1 show the layout of each site with respect to the river channel. The nomenclature 14 of the boreholes is described by the site initials (MCT or ESF) followed by a number 15 (1-5), which relates to the individual boreholes shown in Fig. 1 and Fig. 2, and a 16 second number (1 or 2), which defines the piezometer, piezometer 1 being the deeper 17 and piezometer 2 the shallower. E.g. MCT4-2 describes the shallow piezometer in 18 borehole 4 at Maidencourt Farm. The piezometers at these sites range in depth from 19 1.6 to 12.5 m bgl, with screened sections 0.8 m in length. At each site, four of the 20 boreholes are located in the alluvial plain close to the river and an additional deeper 21 borehole at each site (MCT1 and ESF1) is located further from the river on an 22 adjacent hill slope with both piezometers in these boreholes being located in the 23 Chalk rather than the alluvium (Fig. 2). A common lithology is found at these 24 borehole sites. From 0 to 0.5 m there is a layer of stony / gravely soil below which

alluvial material is found mixed with marl and flint gravels to approximately 4 m.

1 This alluvial layer is very heterogeneous between boreholes, with some sandy and

2 clayey layers found amongst chalky soils, and tends to be 0.5-1 m thicker at East

3 Shefford. From 4 to 10 m is predominantly weathered Chalk and below 10 m the

4 Chalk becomes more consolidated. The hill slope boreholes (MCT1 and ESF1) have a

5 soil layer (~ 0.5 m) overlying weathered Chalk.

#### 3. Methods

8 Routine borehole sampling was carried out between January 2005 and May 2006.

This involved collecting triplicate samples from each piezometer on an approximately

monthly basis. Additional sampling was also carried out at the end of 2006 to monitor

the response of groundwater radon concentrations to rainfall events.

Groundwater samples were taken from piezometers using a submersible pump fitted with a suitable length of hose and the pump inlet positioned in the middle of the screened section. First the piezometer was purged of three times the volume of the screened section to remove any water that may have been residing there for some time. Then 5 litres water were carefully pumped to a bucket from which a standard 600 ml bottle was filled and sealed under the water surface to exclude any air. A further three screened section volumes were removed between samples, with triplicate sampling used to ensure that the radon concentrations measured were representative and had stabilised after purging.

Dissolved radon concentrations were determined by liquid scintillation spectrometry (LSS) according to Pates and Mullinger (2007). Samples were extracted using 20 ml of toluene, with a 10 ml aliquot being combined with the scintillation cocktail Ultima Gold LLT (PerkinElmer) in a low-potassium glass vial before counting for 60 minutes on a Packard Tri-Carb 3170 liquid scintillation counter.

1 Borehole radon concentrations are means of triplicate samples and errors are standard 2 deviations of the triplicate results. This method is hereafter referred to as the "Toluene" method. 3 4 To verify the results of the Toluene analyses an additional method (ASTM, 1998) 5 was used. Ten millilitre samples of borehole water were taken from an overflowing 6 funnel attached to the pump outlet using a disposable syringe. These samples were 7 injected into pre-weighed glass scintillation vials containing the water-immiscible 8 scintillation cocktail Ultima Gold F (PerkinElmer), which were then counted (as 9 above) with no further processing. This method of analysis was the only one 10 employed in event sampling due to the logistics of collecting and analysing samples 11 over the observation period. Results for these analyses are for single samples; the 12 errors quoted are the result of propagated counting and analytical errors. In all cases 13 the relative errors of radon analysis by both methods are less than 10 %. Field trials of 14 these radon analysis methods found highly reproducible results between replicate 15 sample analyses. Relative standard deviations of 5.2 % (n = 25) and 2.3 % (n = 19) 16 were obtained for the ASTM and Toluene methods respectively (see Pates and 17 Mullinger (2007) for a detailed discussion on the analytical errors and reproducibility 18 of both radon analysis methods used). 19 Cores retained from the borehole drilling were analysed for radium content. Sub-20 samples of 200-500 g were taken from the borehole core material at approximately 21 one metre intervals from two boreholes at each site to a depth of 5 m. MCT2, MCT5, 22 ESF3 and ESF5 were selected as providing the most continuous profiles of recovered 23 core material. Samples were dried in an oven at 60 °C to constant mass, ground and 24 homogenised using a tungsten carbide Tema mill and then passed through a 250 µm 25 mesh sieve. Radium was analysed by gamma spectrometry; 20 grams of sample was

- 1 pressed into a pellet and sealed in polycarbonate petri dishes using an epoxy resin.
- These were then left for at least 30 days to allow <sup>214</sup>Pb to reach secular equilibrium 2
- with <sup>226</sup>Ra before counting by gamma spectrometry. Radium concentrations (Bq kg<sup>-1</sup> 3
- of dry sample) were calculated from the <sup>214</sup>Pb 352 keV photopeak with errors derived 4
- 5 from counting statistics (Currie, 1968) and errors in sample size.
- 6 Estimates of radon emanation from borehole core material were made by slurrying approximately 60 g of sample material, prepared as for radium analysis, in a 60 ml 7 glass bottle with deionised water. The bottle was then filled to the brim with deionised 8 9 water and sealed to exclude any air by a plastic screw cap with a PTFE faced sealing 10 disc. The mass of rock sample and volume of water added was measured by weighing 11 the sample bottle at each step. At least 30 days were allowed for radon to reach 12 equilibrium concentrations in the water. Ten millilitres of water was then extracted 13 from the bottle and analysed for radon content via the ASTM (1998) method giving results as Bq 1<sup>-1</sup>. This concentration was then used to calculate the radon emanation 14  $(E_{Rn})$  from each rock sample in Bq kg<sup>-1</sup> using equation (1).

$$E_{Rn} = \frac{[Rn] \cdot V_{w}}{M},\tag{1}$$

where [Rn] is the concentration of dissolved radon in the sample bottle (Bq  $1^{-1}$ ),  $V_w$  is 16 17 the volume of water (litres) and M is the mass of the rock sample (kg). Results of emanation measurements are presented as Bq kg-1 of dry sample material and are 18 means of triplicate analyses, errors are standard deviation of triplicate analyses. 19

Emanation coefficients were then calculated using equation (2).

$$E_{coeff} = \frac{E_{Rn}}{[Ra]},\tag{2}$$

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- where  $E_{coeff}$  is the emanation coefficient and [Ra] the concentration of radium in the
- 2 rock sample (Bq kg<sup>-1</sup>).
- To estimate the density and porosity at these borehole sites, retained complete
- 4 core samples (10 cm in diameter) from each piezometer depth were sectioned into 5
- 5 cm sub-samples. These sections were then hydrated at 20 °C under pressure (max. 4
- 6 bar) to ensure maximum saturation before weighing. The cores were then oven dried
- 7 at 60 °C until constant mass was reached. The porosity was calculated by mass
- 8 difference from the volume of water lost after drying and the density was calculated
- 9 from the dry sample mass and sample volume. Errors in porosity and density
- 10 estimates are calculated by the propagation of errors in the measurement of sample
- 11 size and mass.
- From the results of radon emanation, porosity and density measurements the
- 13 following equation was used to calculate the theoretical radon concentration of
- groundwater ( $[Rn]_{gw}$  in Bq  $l^{-1}$ ) that would be in equilibrium with the local rock.

$$[Rn]_{\rm ew} = E_{\rm Rn} \rho \phi \,, \tag{3}$$

- where  $E_{Rn}$  is the emanation of radon from a sample (Bq kg<sup>-1</sup>),  $\rho$  is density (kg dm<sup>-3</sup>)
- 16 and  $\phi$  is porosity.
- 17 Slug tests were carried out to measure the hydraulic conductivity of each
- piezometer and a number of the deeper piezometers at each site were equipped with
- 19 pressure sensors (PS2100, Greenspan; HOBO U-20-001-01 series, Onset Computer
- 20 Corp. and miniTROLL, In-Situ Inc.). These sensors were set to log at 15 minute
- 21 intervals in order to monitor the changes in local hydraulic head. At least three repeat
- 22 measurements were made of hydraulic conductivity and results presented are the
- 23 mean and standard deviation of these measurements.

River flow at each borehole site was measured using a handheld acoustic Doppler flow meter (SonTek FlowTracker). Surveys were carried out following the guidance

2 flow meter (SonTek FlowTracker). Surveys were carried out following the guidance

of the relevant British Standard (ISO748:2000, 2000) and Environment Agency R&D

Technical Report W4 (Ramsbottom et al., 1997) to provide discharge data with an

accuracy of  $\pm$  10 %. Rainfall was measured by means of a tipping bucket rain gauge

6 located at East Shefford.

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## 4. Results

The results of groundwater radon sampling at Maidencourt Farm and East Shefford in Table 1 and Table 2 respectively. Mean radon concentrations and the maximum and minimum observed concentrations are given for each piezometer. The results of hydraulic conductivity measurements, as well as porosity and density estimates made from borehole core material recovered from each screened section, are also given. Table 3 shows the results of radium and radon emanation measurements for borehole core material.

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## 4.1 Spatial variability

18 The water table is usually deeper at Maidencourt Farm than at East Shefford

during the study period (Fig. 3a). There is also greater variation observed in the water

table at Maidencourt Farm compared to East Shefford. Over the monitoring period the

water table varied by 0.8 m at Maidencourt Farm and by 0.2 m at East Shefford.

Maidencourt Farm is the perennial head of the River Lambourn during late summer,

low flow conditions (Grapes et al., 2006).

24 As a result of the deeper water table and its variation during the sampling period

samples were not obtained from MCT2-2. It was also not possible to obtain samples

1 from MCT4-2 due to the low hydraulic conductivity (Table 1) at this piezometer 2 preventing purging and sampling on a reasonable time scale. The results from each 3 sampled borehole show consistently low radon concentrations in water samples from 4 the deeper piezometers, located in weathered Chalk, with means of between 1.1 and 1.8 Bq l<sup>-1</sup> and a maximum range of 0.6 Bq l<sup>-1</sup> in MCT4-1 (Table 1). 5 6 There are differences between the observed radon concentrations in the shallower piezometers MCT3-2 and 5-2. MCT3-2 has elevated radon concentrations relative to 7 8 all other sampled piezometers at Maidencourt Farm including MCT5-2, which is at a 9 similar depth and located on the opposite river bank. Radon in MCT3-2 switches between two relatively stable concentrations of approximately 5 and 7 Bq l<sup>-1</sup> (Fig. 4a) 10 11 during the sampling period while MCT5-2 remains stable at between 1.2 and 1.8 Bq 1<sup>-1</sup> (Table 1). Geological logs indicate that the cores from these piezometers consisted 12 13 of sandy gravel material, which at MCT5-2 is mixed with some Chalk. 14 At East Shefford a similar vertical distribution of groundwater radon 15 concentrations (Table 2) is observed as at Maidencourt Farm. The piezometers deeper 16 than 4 m bgl have low and stable mean groundwater radon concentrations from 1.1 to 2.2 Bq l<sup>-1</sup> with ranges of 0.7 Bq l<sup>-1</sup> in ESF3-1 and 5-1. ESF4-1 is the shallowest of the 17 'deep' piezometers (3.9 m bgl) and has a mean radon concentration of 3.3 Bq 1<sup>-1</sup>. 18 ESF3-2, 4-2 and 5-2 all have relatively high mean groundwater radon concentrations 19 (> 10 Bq l<sup>-1</sup>), comparable to MCT3-2 (Fig. 4). ESF2-2 has low groundwater radon 20 21 concentrations compared with piezometers of similar depths and locations at ESF and 22 may be considered analogous in this respect to MCT5-2. As with MCT5-2 it is the only piezometer in the flood plain on its respective side of the river (Fig. 1). Overall it 23 24 is found that the shallower piezometers at East Shefford have higher groundwater 1 radon concentrations than those found in the shallower piezometers at Maidencourt

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Radium concentrations in core samples retained from drilling at Maidencourt Farm reflect the trend seen in groundwater radon concentrations, with greater values being observed nearer the surface (Fig. 5a). Radium concentrations in borehole core material are generally within the range 4.4-14.1 Bq kg<sup>-1</sup> with a notable exception in the upper most sample from MCT2 of 39.4  $\pm$  1.3 Bq kg<sup>-1</sup>. Core material from MCT2-2 has a very high radium concentration, which is associated with a clayey region identified in the geological log of this borehole. There is a general increase in radium concentration with decreasing depth of sample. There is clear heterogeneity in radium across both sites in the top few meters of each alluvial aquifer borehole. The mixture of clay, gravel, flint material mixed to varying degrees with Chalk leads to differences in radium concentration between the borehole core samples. Emanation coefficients from borehole core material ranges from 0.05-0.23 with a single high value at 0.37 corresponding to the upper most sample from MCT2, where a high radium concentration was also found (Fig. 5b). There is a general increase in emanation with decreasing depth, similar to that observed in the core radium and groundwater radon data. These emanation results broadly reflect the range found in other emanation studies. The emanation from a wide range of rock and soil types has been investigated by Baretto et al. (1972) with other studies looking at limited rock and soil types (Andrews and Wood, 1972; Bonotto and Andrews, 1997; Misdaq and Amghar, 2005; Przylibski, 2000; Rama, 1991). No studies have been found in the literature for Chalk and soils of the type investigated here. Soils and heavily weathered material have high and variable emanation coefficients, typically between 1 0.1 and 0.6. Consolidated rocks usually have much lower emanation, between 0.01

2 and 0.2, with higher values for less consolidated materials.

The mean porosity and density measured in recovered core sections from the two borehole sites are 39 % (range 24-47 %, standard deviation 8 %) and 1.7 g cm<sup>-3</sup> (range 1.5-2.2 g cm<sup>-3</sup>, standard deviation 0.2) respectively. From these data and the radon emanation results, estimates of equilibrium groundwater radon concentrations were made as described in equation (3) (Fig. 5c, errors calculated from propagation of errors in emanation, porosity and density measurements). For comparison the mean observed radon concentrations in each piezometer to a depth of 6 m bgl are shown in Fig. 5d (error bars show standard deviation of observations from each piezometer). These results are comparable with the observed data, with the exception of the high radon value obtained for the upper most sample of MCT2 associated with the high radium and emanation values. No groundwater radon samples were available from MCT2-2 for comparison with the high estimated value, due to the low water table at this site during the study period (Fig. 2a). Hydraulic conductivities of the measured piezometers at both sites range from 0.4 to 156.2 m d<sup>-1</sup>. The lowest hydraulic conductivity occurs at MCT4-2 where its low conductivity and close proximity to the water table led to slow recovery from

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# 4.2 Temporal variability

Deep piezometers (below 4 m bgl) at both borehole sites exhibit relatively low radon concentrations, which are stable over time (Table 1 and Table 2). There is some

pumping and prevented radon sampling. There is no observed relationship between

the depth and the hydraulic conductivity at each of the piezometers, although there are

a limited number of samples to compare below seven metres.

1 temporal variation in radon concentrations in the piezometers located in the alluvial 2 plain. The consistency in replicate samples and the common patterns of variation in 3 boreholes ESF3 to ESF5 indicates that this variation is not simply a result of sample scatter (Fig. 4). MCT3-2 switches between approximately 5 and 7 Bg 1<sup>-1</sup>, increasing 4 5 between 21 July 2005 and 29 September 2005 and decreasing between 16 March 6 2006 and 4 May 2006. During both periods when switching occurs the riparian zone 7 water table is falling or rising between the limits of 0.8 and 1.1 m bgl, c.f. study 8 period range of 0.6-1.4 m bgl (Fig. 3a). The fall in the water table also results in the 9 source of the Lambourn migrating downstream of Maidencourt Farm, which is why 10 no river flow is recorded at this site between August 2005 and June 2006 (Fig. 3b). The shaded areas in Fig. 3b and Fig. 4a show that the change in radon concentration 12 in MCT3-2 coincides with the no-flow period in the river at Maidencourt Farm. 13 At East Shefford, piezometers ESF3-2, 4-2 and 5-2 show changes in radon 14 concentrations (Fig. 4b-d), but without the distinct periods of stability observed in 15 MCT3-2. During this period there is little movement in the depth of the riparian zone 16 water table at East Shefford, which varies between 0.6 and 0.8 m bgl, and the changes 17 in radon concentrations do not indicate a relationship with seasonal groundwater 18 movements. Additional higher resolution sampling was undertaken to help understand 19 the observed temporal variations in groundwater radon at East Shefford. 20 From higher resolution sampling of groundwater radon during rainfall events it can be seen that short term responses occur at East Shefford. Two sampling periods 22 took place in October and November 2006, during which rainfall events were 23 captured (Fig. 6). The October event is early in the recharge period and the water 24 table at ESF3 is 0.78 m bgl, while in November groundwater has recovered to 0.68 m bgl. 25

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A total of 17.2 mm of rain fell on 11 October (Fig. 6a) contributing to a 7 cm head 1 2 response in ESF3-1 between 07:00 and 18:00 (Fig. 6b), followed by a steady 3 recession period. There is a subtle response in groundwater radon concentrations to this input of water with decreases of 1.5 Bq l<sup>-1</sup> in ESF3-2 and 2 Bq l<sup>-1</sup> in ESF4-2 (Fig. 4 6c). The radon concentration in the shallow piezometers then increases during the 5 6 recession in groundwater level. The magnitude of variation in deeper piezometers 7 makes the responses to rainfall more difficult to discern. 8 On the morning of 28 November 16 mm of rain fell (Fig. 6d) and this event 9 resulted in a 10 cm head response in ESF3-1 (Fig. 6e). Here the radon concentrations, monitored only in ESF3 on this occasion, show a response of 2 Bq 1<sup>-1</sup>. Less of the 10 11 recession curve was captured by post event sampling and so it is not clear whether 12 radon concentrations in ESF3-2 recovered fully after this event. 13 Although only small differences in radon concentrations are found in the first 14 event the simultaneous response of piezometers ESF3-2 and ESF4-2 indicates that the 15 observations are a response to rainfall. This hypothesis is then further supported by 16 the observations during the second event, which show significant differences in pre-17 and post-event radon concentrations. The greater radon response in the November 18 event is likely to be due to the shallower depth of the water table providing more rapid

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#### 5. Discussion

23 5.1 Spatial Variability

From the analysis of groundwater and borehole core samples it can be seen that groundwater radon concentrations are related to the abundance of radium and

connection of the infiltrating water to the aquifer and also to the greater intensity of

rainfall, which can be seen in the hydraulic head response.

differences in emanation with depth. The increased abundance of radium and emanation of radon near the surface results in a distinctive vertical groundwater profile in the riparian zone. It is also shown that measurements of radium concentration and radon emanation can give good indicative estimates of the distribution of groundwater radon concentrations that may be found at a site. The results reflect the types of deposit found in the vertical profile and the degree of weathering in the near surface. The alluvial gravels and sediments in the river valley are materials remaining after the erosion of the overlying Palaeogene deposits and the insoluble fraction of sediments co-deposited when the Chalk was formed. These insoluble sediments, containing clays and other fine material, tend to be higher in radium than the carbonate matrix of the Chalk due to their mineral composition and also their adsorption properties, which allows for accumulation of parent nuclides on surfaces (Bonotto and Andrews, 1993; Osmond and Cowart, 1992).

The higher radon concentrations in groundwaters from the shallow riparian piezometers at East Shefford compared with Maidencourt Farm may be due to the thicker alluvial deposits found at East Shefford, providing a larger radon source, and the elevated water table allowing dissolution of radon from nearer the surface.

The heterogeneity of groundwater radon concentrations found in the shallow piezometers of riparian zone boreholes indicates that spatial variation can occur in near-surface groundwater inputs to surface waters. Therefore, contributions to stream radon from this type of system may vary along the course of a river. In river reaches where there is significant interaction of groundwater with alluvial deposits in the river corridor, such as in the Lambourn Valley, the variation in groundwater radon could be used to identify contributions to stream flow from the near-surface and deeper groundwaters depending upon the concentration of radon in the input water. It is also

1 shown that measurements of radium concentration and radon emanation can give

good indicative estimates of the distribution of groundwater radon concentrations that

3 may be found at a site.

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## 5.2 Temporal Variability

At Maidencourt Farm the temporal trend of radon in MCT3-2 is to switch between two relatively stable concentrations (Fig. 4a), suggesting that the changes occurring at this site are due to the seasonal changes in the water table (Fig. 3). When flow in the river at Maidencourt Farm stops, the radon concentration in groundwater of MCT3-2 increases. This shows that there is a dynamic equilibrium between the radium source and groundwater radon in the vicinity of MCT3-2. The controls on this relationship are not known but may come from changes in water table elevation activating different radon sources. There is relatively little variation in the water table at East Shefford compared to Maidencourt Farm (Fig. 3a) and no seasonal changes in groundwater radon are observed in the shallower piezometers at East Shefford. However, shorter time scale changes in groundwater radon concentrations of the order of weeks were observed at East Shefford during the routine sampling period (January 2005 to May 2006, Fig. 4bd). From greater temporal resolution event sampling (October and November 2006) it can be seen that rainfall events have a rapid impact on the concentration of radon in the shallowest groundwaters (Fig. 6). The precise time of response is not captured in the samples taken and so estimates of the groundwater radon concentrations between samples are indicated by the interpreted response lines. The effect of infiltration of low radon event water is observed as deep as 3 metres below ground level and approximately 2.5 m below the water table, demonstrating relatively rapid subsurface

2 mixing. The effects of greater rainfall intensity and the shallower water table at East

Shefford in November lead to a greater event response in the groundwater radon

4 concentration than in October.

Following the October event, reductions in radon concentrations of 18 and 12 % are observed in ESF3-2 and 4-2, respectively. In November, radon concentrations decrease by 19 % in ESF3-2 following rain. Assuming that infiltrating water has a zero radon concentration, mixing of between 12 and 19 % event water with existing groundwater is implied at these sites for the events observed. These values could be considered a minimum degree of mixing as it assumes that the event water contains no radon when it reaches the saturated zone. In reality it is likely to have accumulated some dissolved radon as a result of dissolution of radon in soil gas during transit through the unsaturated zone. The accumulation of radon by recharge water will depend partially upon the mechanism of recharge and water movement through the unsaturated zone at these sites (e.g. direct recharge by new rain water via fracture flow vs. displacement of existing unsaturated zone water via a piston flow mechanism, see Ireson et al., 2006).

Estimates of the time it would take for groundwater radon to recover to pre-event concentrations were made. The recovery time was estimated as the time taken to reach 95 % of the pre-event radon concentration, assuming a quasi-steady state system, where the pre-event groundwater is in equilibrium with its source and the accumulation of radon post event is governed by radioactive ingrowth (Andrews and Wood, 1972; Hoehn et al., 1992). Ingrowth of radon is calculated by the equation,

$$A_{i} = A_{0}(1 - e^{-\lambda t}), (4)$$

where  $A_t$  is the groundwater radon concentration (Bq  $l^{-1}$ ) at time t,  $A_0$  is the pre-event 1 groundwater radon concentration (Bq  $I^{-1}$ ) and  $\lambda$  is the radioactive decay constant ( $t^{-1}$ ). 2 3 Recovery of radon concentrations from these rainfall events is calculated to take 4 between 5 and 7 days, depending on the piezometer and event. This is a theoretical maximum time for recovery to occur. Rapid recovery of pre-event radon 5 6 concentration is observed in the October event where groundwater radon regains preevent concentrations less than 30 hours from the time of first rainfall. If radon 7 8 recovery continues at the rate seen during recession of the second rainfall event then 9 the groundwater will return to pre-event radon concentration within 48 hours. 10 Groundwater radon concentrations recover more quickly than is predicted by radioactive ingrowth, emphasising that this is not a closed system. Speculation on the 12 driving mechanism for this rapid recovery is not possible from this data set. 13 Unlike at East Shefford short time scale variations in groundwater radon 14 concentrations are not seen in the routine monitoring data from Maidencourt Farm. It 15 may be that the greater depth of unsaturated zone at Maidencourt protects the 16 groundwater from such rapid and significant responses to rainfall events. 17 Mullinger et al. (2007) proposed that the variations in modelled radon inputs to the 18 Lambourn could be caused by changes in the radon concentration of groundwater 19 feeding the river. The data presented here shows that consistently higher radon 20 concentrations are found in groundwater nearer the surface of the alluvial aquifer. It is hypothesised that the fluctuations in the water table provide a changing source of 22 radon in groundwater that may be discharged to a river. To examine the impact of 23 changing water table elevation on radon concentrations in groundwater discharges, a 24 trendline was fitted to the observed groundwater radon data using a linear regression 25 method to provide a relationship with depth (equation (5)).

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 $[Rn] = 14.191d^{-1.234}, (5)$ 

where [Rn] is groundwater radon concentration (Bq  $1^{-1}$ ) and d is depth (m). Fig. 5d 1 2 shows the groundwater radon concentrations estimated by equation (5) against mean observed radon concentrations ( $R^2 = 0.65$ ). By integrating over a given depth range 3 4 the net radon concentration of groundwater discharges can be estimated. This has 5 been done for three example depth ranges, assuming an aquifer base of 3, 5 and 10 m 6 bgl and an equal contribution of groundwater from each depth. One metre has been 7 used as the upper limit of integration in all cases, as extrapolation of the relationship beyond this to shallower depths could not be justified given the rate of increase in 8 9 radon concentration increase that would result from the derived relationship. Results 10 of this exercise are shown in Fig. 7a, which shows the estimated radon concentration 11 of discharging groundwater for a given water table depth. 12 By combining the above approach with observed water table data from Fig. 3a, the 13 variation in groundwater radon concentration over time has been estimated for an aquifer depth of 5m (Fig. 7b). Result ranges are 4-6 Bq I<sup>-1</sup> for MCT3 and 5.3-6.2 Bq 14 l<sup>-1</sup> for ESF3, the greater variation in water table at Maidencourt Farm being reflected 15 16 in the estimated radon concentration. These results could be viewed as a lower limit 17 for variation in discharge radon concentrations due to the assumption of equal 18 contribution from each depth. Exponential mixing of groundwaters in shallow 19 unconfined systems leads to shorter residence times of shallower groundwaters (Cook 20 and Böhlke, 2000). Therefore, groundwater discharges may be made up by a greater 21 proportion of shallower groundwater, which will increase the mean concentration of 22 discharge water. The conceptual model in Fig. 7c illustrates how movement of the 23 water table could result in changes in radon concentrations of groundwater entering a 24 river. As the water table rises it interacts more with the near-surface zone where radon

- 1 production is highest, leading to a higher mean groundwater radon concentration. This
- 2 may also result in greater contributions of shallower groundwater to the river at times
- 3 when the water table is elevated, which will be of higher radon concentration.

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#### 6. Conclusions

Consistently higher groundwater radon and borehole core radium concentrations are found near the surface (0-4 m bgl) at both East Shefford and Maidencourt Farm borehole sites, with low and stable radon concentrations at greater depths in the weathered Chalk. The groundwater radon concentrations are consistent with

observations at other comparable sites in Pang and Lambourn catchments (Mullinger

11 et al., 2007).

A combination of radium abundance and radon emanation control the concentration of radon in groundwater. The enrichment of radium and greater rates of emanation in the near-surface alluvial materials influence the spatial variation of radon at these sites. High spatial variability in the hydraulic conductivities found at these sites mean that local controls will influence groundwater movement and radon concentrations in the riparian zone.

Variations in radon concentrations in near-surface groundwater at East Shefford can be affected by short time scale hydrological events, whereas those at Maidencourt Farm can be influenced by seasonal variations in water table elevation. Perturbations to groundwater radon concentrations following rainfall events are relatively short-lived and of the order of a few days at most in areas where the water table is very close to the surface.

Observations of groundwater radon concentrations over time and the distribution of radium at the Lambourn borehole sites show that there are variable sources of

1 radon for groundwater that is discharged to the river. Seasonal changes in the water

table can lead to a greater source of river water from near the surface of the alluvial

aquifer, which is higher in radon. This may account for the observations of changing

radon inputs to the river in Mullinger et al. (2007) and is illustrated by the conceptual

5 model in Fig. 7c.

These results show that there can be significant spatial and temporal variability in

7 groundwater radon in the riparian zone. The riparian zone, due to the proximity of the

river channel, has a potentially important influence on groundwater radon

concentrations before discharge occurs to rivers. Therefore, it is essential that this part

of the groundwater system is studied in order to properly characterise the sources of

radon being discharged to rivers.

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## Figure captions

Fig. 1 Location of the Lambourn catchment. Cross-sections locations (Fig. 2) and local topography are indicated. Insets show layout of borehole sampling sites.

Fig. 2 Cross sections of (a) Maidencourt Farm and (b) East Shefford borehole sites, showing ground level, relative distance of boreholes from river, piezometer depths and water table minima and maxima during the sampling period. Insets show the full length of cross sections in Fig. 1 and indicate the extent of the enlarged sections.

Fig. 3 (a) Water table in riparian boreholes in MCT3-1 and ESF3-1 (the gap in data from ESF3 is due to logger failure). (b) River flow at East Shefford and Maidencourt Farm. Shaded area indicates period of no flow at Maidencourt Farm.

Fig. 4 Temporal variability of groundwater radon in the most variable boreholes (error bars show standard deviation of triplicate samples). Shaded area indicates period of no flow at Maidencourt Farm.

Fig. 5 (a) Radium concentration of borehole core material (errors derived from counting statistics and errors in sample size). (b) Emanation coefficients from borehole core material (error bars show standard deviation of triplicate samples). (c) Estimated groundwater radon concentrations as calculated by equation (3) (errors calculated from propagation of errors in emanation, porosity and density measurements). (d) Observed mean groundwater radon concentrations in piezometers above 6 m bgl and trendline (equation (5)) fitted by linear regression (error bars show standard deviation of all observations from each piezometer). Hollow symbols in (a-c) represent samples associated with piezometer screened sections. Hollow symbols in (d) represent piezometer screened sections from which core material was analysed for radium and radon emanation.

Fig. 6 Borehole responses to rainfall events in October and November 2006. (a) & (d) Hourly precipitation. (b) & (e) ESF3-1 water table. (c) & (f) Groundwater radon concentrations (ASTM method, errors based on counting statistics and errors in sample size).

Fig. 7 (a) Estimated discharge groundwater radon concentration for a given water table depth assuming equal contributions from each depth, calculated using equation (5) with examples of contributions assuming different aquifer depths. (b) Estimated impact of water table fluctuation on groundwater radon concentration, calculated by the integration of radon over the saturated depth of aquifer down to 5 m bgl using equation (5). (c) Conceptual model of variations in radon input to streams from an alluvial aquifer. A rise in the water table leads to groundwater occupying the zone of higher radon production in the near-surface of the riparian zone. This provides a greater source of high radon water feeding the river, resulting in seasonal changes in the input of radon to rivers.

# **Tables**

 Table 1 Parameters observed for Maidencourt Farm boreholes. All boreholes are screened for 0.8 m, with the pump depth located at the centre of screened section. Errors in porosity and density estimates are calculated by the propagation of errors in the measurement of sample size and mass. Hydraulic conductivity results are mean and standard deviation of repeat measurements.

| Piezometer | Pump depth (m bgl) | <sup>222</sup> Rn (Bq l <sup>-1</sup> ) |           |            | Porosity                 | Density       | Hydraulic conductivity |
|------------|--------------------|---|-----------|------------|--------------------------|---------------|------------------------|
|            |                    | Mean                                    | Min       | Max        | (%)                      | $(g cm^{-3})$ | $(m d^{-1})$           |
| MCT1-2     | 5.6                | 1.7                                     | 1.5       | 1.8        | $45 \pm 2$               | $1.7 \pm 0.1$ | $7.7 \pm 0.5$          |
| MCT1-1     | 9.6                | 1.3                                     | 1.1       | 1.4        | $47 \pm 2$               | $1.6 \pm 0.1$ | $32.3 \pm 2.6$         |
| MCT2-2     | 1.6                | N                                       | ot sample | ed         | $34 \pm 2$ $1.8 \pm 0.1$ |               | $9.9 \pm 0.6$          |
| MCT2-1     | 4.6                | 1.4                                     | 1.2       | 1.5        | $45 \pm 2$               | $1.5 \pm 0.1$ | $49.0 \pm 3.3$         |
| MCT3-2     | 3.0                | 6.1                                     | 4.7       | 7.2        | No sample                |               | $83.2 \pm 20.2$        |
| MCT3-1     | 5.6                | 1.2                                     | 1.0       | 1.4        | $43 \pm 2$               | $1.6 \pm 0.1$ | $12.6 \pm 1.4$         |
| MCT4-2     | 2.4                | Not sampled                             |           | $29 \pm 2$ | $2.0 \pm 0.1$            | $0.4 \pm 0.1$ |                        |
| MCT4-1     | 4.6                | 1.8                                     | 1.5       | 2.1        | $43 \pm 2$               | $1.6 \pm 0.1$ | $46.3 \pm 3.8$         |
| MCT5-2     | 3.5                | 1.5                                     | 1.2       | 1.8        | $42 \pm 2$               | $1.6 \pm 0.1$ | $5.5 \pm 2.1$          |
| MCT5-1     | 6.4                | 1.1                                     | 0.9       | 1.3        | $29 \pm 2$               | $1.9 \pm 0.1$ | $68.0 \pm 4.0$         |

Table 2 Parameters observed for East Shefford boreholes. All boreholes are screened for 0.8 m, with the pump depth located at the centre of screened section. Errors in porosity and density estimates are calculated by the propagation of errors in the measurement of sample size and mass. Hydraulic conductivity results are mean and standard deviation of repeat measurements.

| Piezometer | Pump depth (m bgl) | <sup>222</sup> Rn (Bq l <sup>-1</sup> ) |     |      | Porosity   | Density               | Hydraulic conductivity |
|------------|--------------------|---|-----|------|------------|-----------------------|------------------------|
|            |                    | Mean                                    | Min | Max  | (%)        | (g cm <sup>-3</sup> ) | $(m d^{-1})$           |
| ESF1-2     | 9.6                | 1.6                                     | 1.4 | 2.1  | $45 \pm 2$ | $1.6 \pm 0.1$         | No measurement         |
| ESF1-1     | 12.5               | 1.2                                     | 1.1 | 1.2  | $43 \pm 2$ | $1.6 \pm 0.1$         | No measurement         |
| ESF2-2     | 2.6                | 3.1                                     | 2.3 | 3.6  | No sample  |                       | $156.2 \pm 46.5$       |
| ESF2-1     | 5.5                | 1.1                                     | 0.9 | 1.2  | $42 \pm 2$ | $1.6 \pm 0.1$         | No measurement         |
| ESF3-2     | 2.1                | 10.6                                    | 9.1 | 11.7 | $26 \pm 2$ | $1.8 \pm 0.1$         | $16.8 \pm 1.9$         |
| ESF3-1     | 4.6                | 2.2                                     | 1.7 | 2.4  | $44 \pm 2$ | $1.6 \pm 0.1$         | $13.2 \pm 6.2$         |
| ESF4-2     | 1.6                | 11.0                                    | 8.5 | 14.2 | $24 \pm 2$ | $2.2 \pm 0.1$         | $15.4 \pm 0.4$         |
| ESF4-1     | 3.9                | 3.3                                     | 2.6 | 4.6  | $43 \pm 2$ | $1.6 \pm 0.1$         | $5.3 \pm 0.8$          |
| ESF5-2     | 2.6                | 11.1                                    | 9.2 | 11.8 | No sample  |                       | $1.5 \pm 0.4$          |
| ESF5-1     | 4.5                | 1.5                                     | 1.2 | 1.9  | $39 \pm 2$ | $1.7 \pm 0.1$         | $32.1 \pm 3.6$         |

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|----------|-------|----------------|----------------|-----------------|
|          |       |                | Rn             |                 |
|          | Depth | Ra activity    | emanation      | Emanation       |
| Borehole | (m)   | $(Bq kg^{-1})$ | $(Bq kg^{-1})$ | coefficient     |
| ESF3     | 1.0   | $13.4 \pm 0.5$ | $2.8 \pm 0.2$  | $0.21 \pm 0.02$ |
| ESF3     | 2.3   | $7.9 \pm 0.4$  | $1.2 \pm 0.2$  | $0.15 \pm 0.02$ |
| ESF3     | 3.1   | $12.0 \pm 0.6$ | $1.2 \pm 0.2$  | $0.10 \pm 0.01$ |
| ESF3     | 4.4*  | $4.9 \pm 0.5$  | $0.4 \pm 0.2$  | $0.08 \pm 0.01$ |
| ESF3     | 5.0   | $5.8 \pm 0.3$  | $0.4 \pm 0.2$  | $0.06 \pm 0.01$ |
| ESF5     | 1.1   | $13.1 \pm 0.5$ | $3.0 \pm 0.2$  | $0.23 \pm 0.02$ |
| ESF5     | 2.4*  | $13.0 \pm 0.5$ | $1.6 \pm 0.1$  | $0.13 \pm 0.01$ |
| ESF5     | 3.0   | $10.4 \pm 0.5$ | $1.2 \pm 0.1$  | $0.12 \pm 0.01$ |
| ESF5     | 4.3*  | $6.4 \pm 0.5$  | $0.3 \pm 0.1$  | $0.05 \pm 0.02$ |
| ESF5     | 4.8   | $7.5 \pm 0.4$  | $0.5 \pm 0.1$  | $0.07 \pm 0.01$ |
| MCT2     | 1.2*  | $39.4 \pm 1.3$ | $14.5 \pm 1.2$ | $0.37 \pm 0.03$ |
| MCT2     | 2.1   | $14.1 \pm 0.7$ | $3.3 \pm 0.1$  | $0.23 \pm 0.01$ |
| MCT2     | 2.8   | $6.1 \pm 0.4$  | $0.6 \pm 0.1$  | $0.10 \pm 0.02$ |
| MCT2     | 4.2   | $4.9 \pm 0.5$  | $0.4 \pm 0.1$  | $0.07 \pm 0.01$ |
| MCT2     | 4.9*  | $4.4 \pm 0.4$  | $0.6 \pm 0.1$  | $0.14 \pm 0.01$ |
| MCT5     | 1.1   | $8.4 \pm 0.3$  | $1.0 \pm 0.1$  | $0.12 \pm 0.01$ |
| MCT5     | 1.9   | $10.8 \pm 0.6$ | $1.3 \pm 0.1$  | $0.12 \pm 0.01$ |
| MCT5     | 3.2*  | $11.2 \pm 0.4$ | $1.2 \pm 0.1$  | $0.11 \pm 0.01$ |
| MCT5     | 4.0   | $6.4 \pm 0.4$  | $1.0 \pm 0.1$  | $0.16 \pm 0.02$ |
| MCT5     | 5.4   | $7.4 \pm 0.5$  | $0.4 \pm 0.1$  | $0.05 \pm 0.01$ |

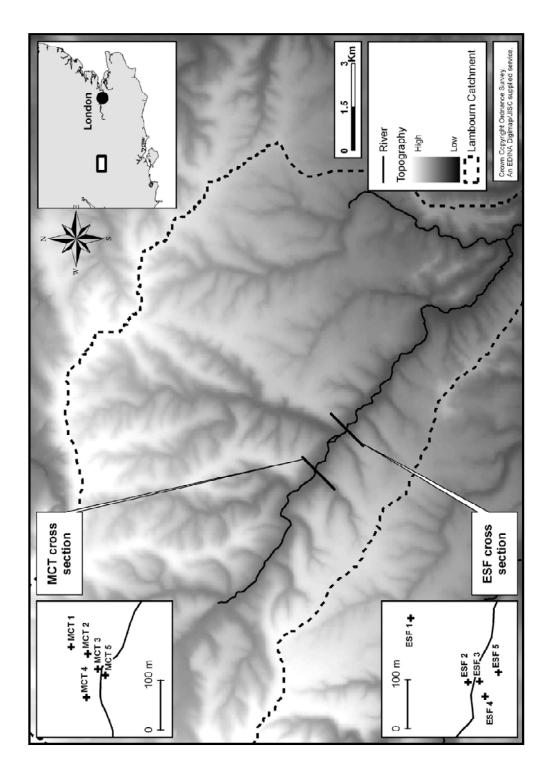


Fig. 1 Location of the Lambourn catchment. Cross-sections locations (Fig. 2) and local topography are indicated. Insets show layout of borehole sampling sites.



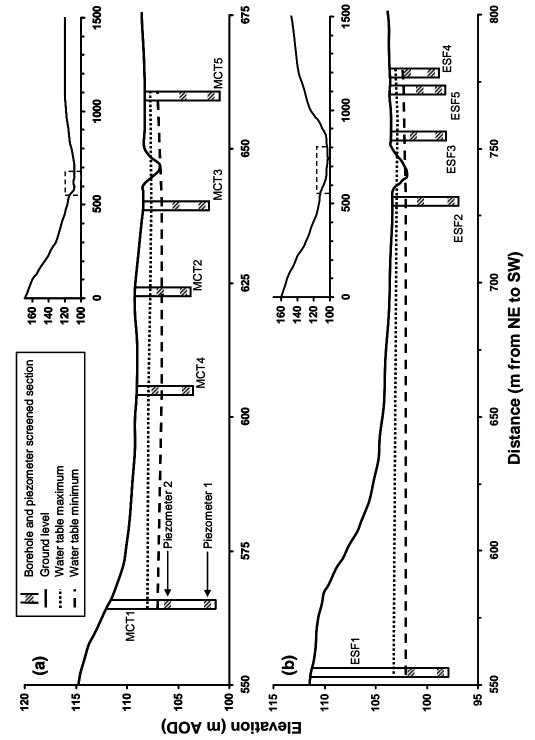


Fig. 2 Cross sections of (a) Maidencourt Farm and (b) East Shefford borehole sites, showing ground level, relative distance of boreholes from river, piezometer depths and water table minima and maxima during the sampling period. Insets show the full length of cross sections in Fig. 1 and indicate the extent of the enlarged sections.

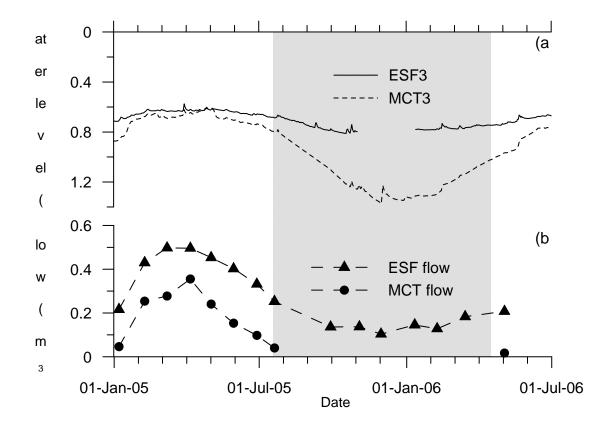


Fig. 3 (a) Water table in riparian boreholes in MCT3-1 and ESF3-1 (the gap in data from ESF3 is due to logger failure). (b) River flow at East Shefford and Maidencourt Farm. Shaded area indicates period of no flow at Maidencourt Farm.

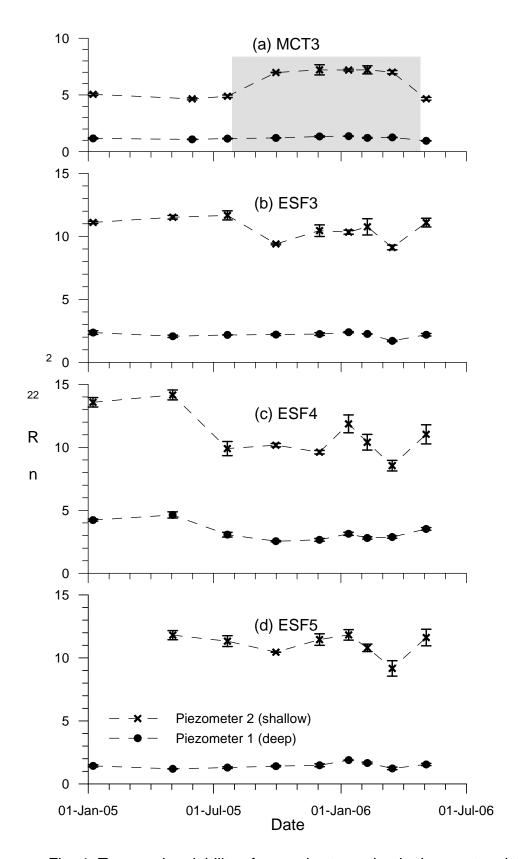


Fig. 4 Temporal variability of groundwater radon in the most variable boreholes (error bars show standard deviation of triplicate samples). Shaded area indicates period of no flow at Maidencourt Farm.

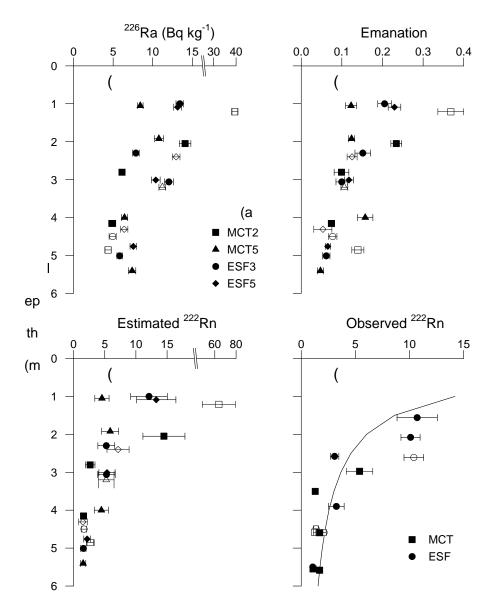
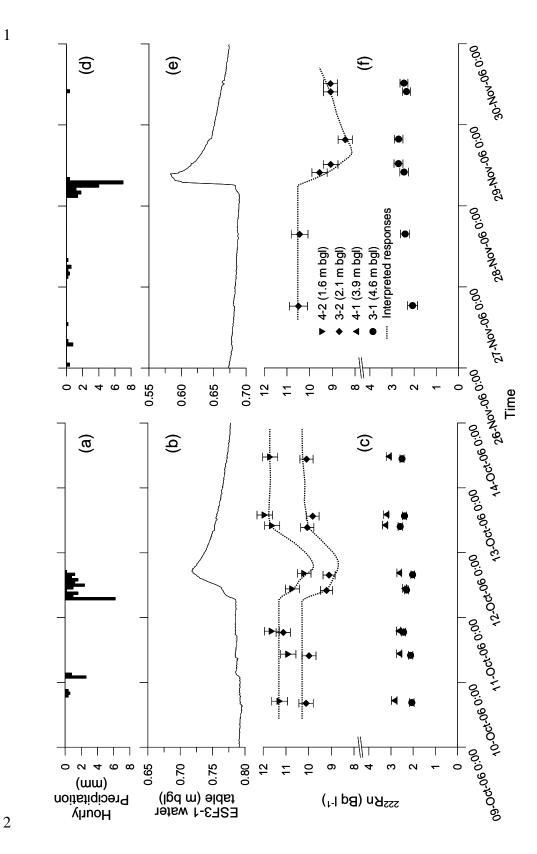


Fig. 5 (a) Radium concentration of borehole core material (errors derived from counting statistics and errors in sample size). (b) Emanation coefficients from borehole core material (error bars show standard deviation of triplicate samples). (c) Estimated groundwater radon concentrations as calculated by equation (3) (errors calculated from propagation of errors in emanation, porosity and density measurements). (d) Observed mean groundwater radon concentrations in piezometers above 6 m bgl and trendline (equation (5)) fitted by linear regression (error bars show standard deviation of all observations from each piezometer). Hollow symbols in (a-c) represent samples associated with piezometer screened sections. Hollow symbols in (d) represent piezometer screened sections from which core material was analysed for radium and radon emanation.



precipitation. (b) & (e) ESF3-1 water table. (c) & (f) Groundwater radon concentrations (ASTM method, errors based on counting statistics and errors in sample size). Fig. 6 Borehole responses to rainfall events in October and November 2006. (a) & (d) Hourly

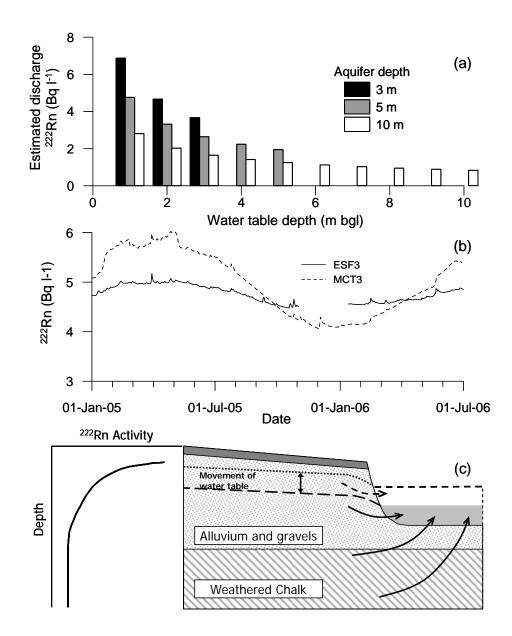


Fig. 7 (a) Estimated discharge groundwater radon concentration for a given water table depth assuming equal contributions from each depth, calculated using equation (5) with examples of contributions assuming different aquifer depths. (b) Estimated impact of water table fluctuation on groundwater radon concentration, calculated by the integration of radon over the saturated depth of aquifer down to 5 m bgl using equation (5). (c) Conceptual model of variations in radon input to streams from an alluvial aquifer. A rise in the water table leads to groundwater occupying the zone of higher radon production in the near-surface of the riparian zone. This provides a greater source of high radon water feeding the river, resulting in seasonal changes in the input of radon to rivers.