Application of Rn-222 isotope for the interaction between surface water and groundwater in the Source Area of the Yellow River

M. J. Zheng, C. W. Wan, M. D. Du, X. D. Zhou, P. Yi, A. Aldahan, H. J. Jin, D. L. Luo, Z. B. Yu and M. Gong

ABSTRACT

A pioneering rapid and direct measurement of dissolved ^{222}Rn in the field has been used here to explore interaction between surface and groundwater in the source area of the Yellow River (SAYR). The results indicate average ^{222}Rn activity of 2,371 Bq/m^3 in surface water and 27,835 Bq/m^3 in groundwater. The high ^{222}Rn activity (up to 9,133 Bq/m^3) found in the southeast part of the SAYR suggests possible influence of permafrost on the exchange between surface water and groundwater. The remarkable contrast among the different samples of a stream in the Shuangchagou basin, a typical basin in the SAYR, clearly indicates groundwater infiltration along the north tributary and occurrence of groundwater end-member in the south tributary. Considering no ^{222}Rn decay and atmospheric evasion, the daily average fraction of groundwater input to the surface water through the end-member in a location (S1) is estimated at 19%. Despite the up to 40% uncertainty, this is the first estimate of a reference value for groundwater input in this basin and which can be improved in the future with more samples and measurements. ^{222}Rn can be a rapid and easily measured tracer of surface water–groundwater interaction for future investigation in the Qinghai-Tibet Plateau.

Key words | ^{222}Rn, groundwater, permafrost, Qinghai-Tibet Plateau, surface water

M. J. Zheng
C. W. Wan
M. D. Du
X. D. Zhou
P. Yi
Z. B. Yu
M. Gong
College of Hydrology and Water Resources,
Hohai University,
Nanjing,
China

X. D. Zhou
P. Yi (corresponding author)
Z. B. Yu
State Key Laboratory of Hydrology-Water Resources and Hydraulic Engineering,
Hohai University,
Nanjing,
China
E-mail: *pengyi1915@163.com*

A. Aldahan
Department of Geology,
United Arab Emirates University,
Al Ain,
UAE

H. J. Jin
D. L. Luo
State Key Laboratory of Frozen Soils Engineering,
Cold and Arid Regions Environmental and Engineering Research Institute, Chinese Academy of Sciences,
Lanzhou, Gansu,
China

INTRODUCTION

It is essential to understand the exchange of surface water and groundwater to ensure sustainable water resource utilization and protection, in terms of water quality and quantity. Under a warming climate and degrading permafrost condition, surface water and groundwater transformation may have changed greatly on the Qinghai-Tibet Plateau, including the source area of the Yellow River (SAYR). The SAYR has received increasing attention in the last decade due to regional socioeconomic development in the region (Jin *et al.* 2009; Luo *et al.* 2012). Several methods have been widely used in assessing surface–groundwater interaction including analytical and field measurements (Krause *et al.* 2007; Anibas *et al.* 2011; O'Connor & Moffett 2015). However, due to the harsh natural conditions in the SAYR, field work faces many difficulties. For example, numerical methods require significant amounts of detailed hydrological information and long time-series monitoring data of groundwater to identified unique interaction fingerprints.

Isotope techniques have been used widely to investigate the exchange between surface water and groundwater in terms of amounts and directions. Isotopes, such as ^{18}O, ^{2}H

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(Kumar et al. 2008; Herczeg & Leaney 2010; Petitta et al. 2010), and also dissolved noble gases and anthropogenic compounds, such as SF_6 or chlorofluorocarbons (CFCs), have been commonly used (Cook et al. 2003, 2006). In addition to these tracers, the naturally occurring radon-222 (^{222}Rn) constitutes a vital tool for quantifying groundwater outflows to surface water because it is greatly enriched in groundwater compared to surface water (by 2–4 orders of magnitude), relatively easy to measure, and chemically conservative (Cook et al. 2008; Chanyotha et al. 2014). Since ^{222}Rn has a short half-life (3.82 d), it has great potential in the study of rapid mixing processes that occur on time scales of hours to several days. The obvious differences of ^{222}Rn activity can be found in surface water if groundwater discharge exists (Dimova et al. 2013). In addition, the developed instrumentation (RAD H_2O, Durridge Inc.) allows for easy and rapid direct measurement of ^{222}Rn in water.

^{222}Rn has been widely used to evaluate the exchange between groundwater and surface water (Wu et al. 2004; Cook et al. 2006; Oyarzún et al. 2014). The ^{222}Rn method also has potential in harsh environments, such as the Qinghai-Tibet Plateau, in order to assess hydrological interactions. For example, Su et al. (2014) present a new mass balance method with the tracer ^{222}Rn estimating gross surface water and groundwater exchange in Nalenggele River, in the southwestern part of the Qaidam basin, where conventional hydrologic data are sparse. The reliability and accuracy of the method have also caused some concerns during the last few decades due to uncertainties regarding variation of ^{222}Rn activity in groundwater, heterogeneous distribution of ^{222}Rn parent nuclide ^{226}Ra and hyporheic exchange (Cook et al. 2006; Stellato et al. 2008).

Presently, obtaining hydrological data in general in the Qinghai-Tibet Plateau are hampered by the harsh environment and difficulties in performing long-lasting field work. Technological advances in the measurement of dissolved ^{222}Rn have opened up the possibility for relatively rapid and direct assessment in the field, thereby making field campaigns profitable in terms of time and effort. Based on this, we have initiated a pioneering ^{222}Rn investigation in water systems of the SAYR in the hope of making the method a potential spatial and temporal monitoring program. The investigation's main objective is to perform an assessment of the suitability of ^{222}Rn as a hydrological tracer in plateau areas, such as the SAYR, exploring details of the exchange between surface water and groundwater for a larger scale investigation in the future.

MATERIALS AND METHODS

Study area

The SAYR (33°56′-35°31′N, 95°55′-98°41′E, elevation 4,100–5,442 m) is located along the northeastern Qinghai-Tibet Plateau with a total catchment area of approximately 2.9×10^4 km^2 (Luo et al. 2014a). Surrounded by northwest–southeast trending mountains, the SAYR includes the Gyaring and Ngöring (Sisters) Lakes and the Yellow River floodplains with flat topography and meandering river channels (Jin et al. 2009). The climate of the area is characterized by a mean annual precipitation range from 282 to 590 mm decreasing gradually from southeast to northwest (more than 80% precipitation occurred from May to October) (Luo et al. 2012; Wen et al. 2015). The mean annual temperature is below −3.2 °C leading to the occurrence of frozen ground and permafrost. The distribution of permafrost in SAYR (Figure 1) is also strongly controlled by elevation and geomorphic features (mainly elevation and soil thickness), resulting in the occurrence of discontinuous and continuous permafrost (Jin et al. 2009).

The selected study area is in the Shuangchagou basin which is located on the south bank of Ngöring Lake with an area of 10 km^2 (Figure 1). This area is characterized by Quaternary clastic aquifers with mosaic distribution of discontinuous and sporadic permafrost (Figure 2). Different types of permafrost-related groundwater including the supra-permafrost, in-permafrost and sub-permafrost water may exist in this area. Ground thawing in the Shuangchagou basin occurs during the middle or end of May and ground freezing starts at the beginning or middle of October (Luo et al. 2014b). The Shuangchagou stream has two tributaries (south tributary and north tributary) and flows through the basin from west to east. The landscape of the Shuangchagou basin is mainly alpine grassland and barren soil which is rich in organic matter at the upper part and in inorganic clay fractions at the middle to

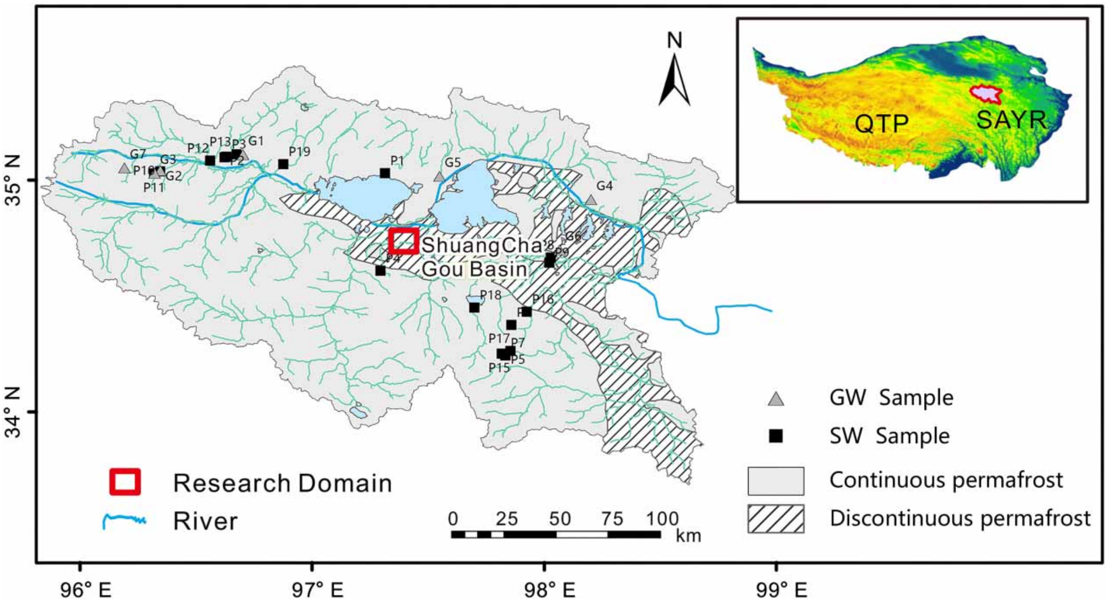

Figure 1 | Map of permafrost distribution in the SAYR (adapted from Jin et al. (2009)).

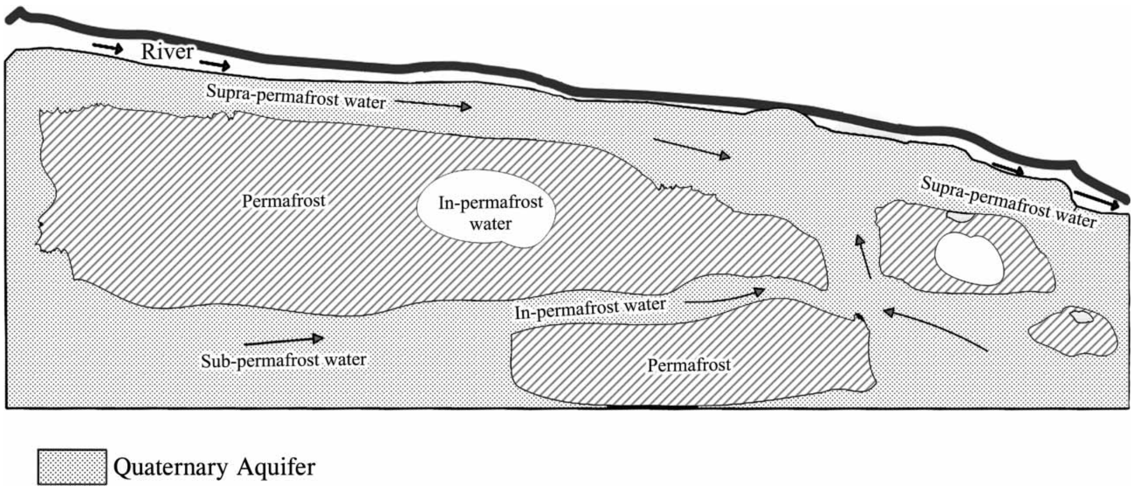

Figure 2 | Schematic diagram of permafrost and groundwater situation in the Shuangchagou basin.

lower parts. There are not obvious differences between these two tributaries as both the aquifers and the cover vegetation are comparable.

Sampling

The information presented here is derived from two sampling campaigns, carried out in the autumn of 2014 (Figure 3), which were free of precipitation. In total, 19 surface water and seven groundwater samples from different locations in the SAYR were measured for ^{222}Rn during the pilot sampling project presented here (25th September–1st October). Two groundwater water samples were taken from two wells that were pumped for at least 1 hour before sampling. The other five groundwater samples were collected directly from springs. In addition, a field trip was conducted on 25th September for the collection of 13 surface water samples along the Shuangchagou stream

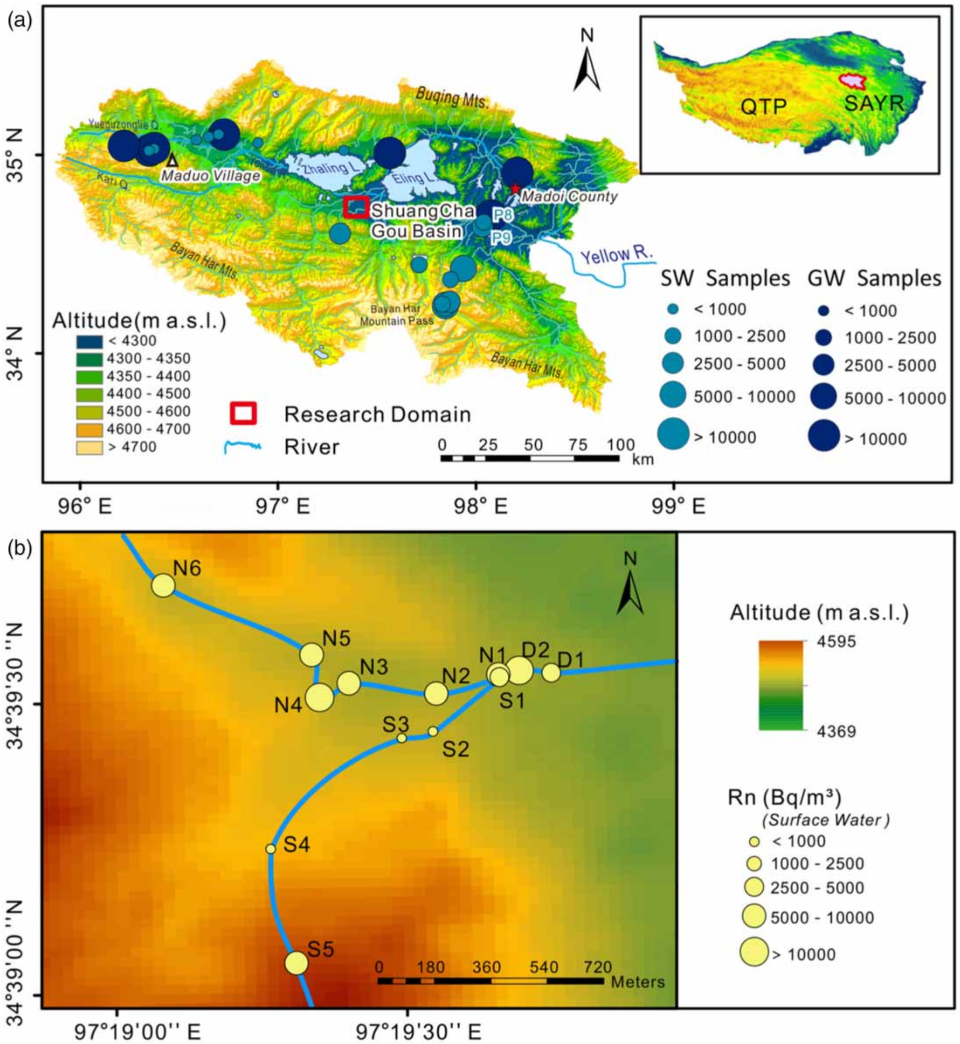

Figure 3 | Location of samples and the range of ^{222}Rn activity in the SAYR (a) and Shuangchagou stream (b). N, S, and D represent the north tributary, south tributary, and the main stream in the Shuangchagou basin, respectively.

(including the trunk and two tributaries). Continuous monitoring of water on the 26th–27th September at a time interval of 2 hours was performed at the two locations (nine samples for location S1 and 10 samples for location D2), where the ^{222}Rn activities were exceptionally higher than other upstream locations. Unfortunately, we could not analyze groundwater in the Shuangchagou basin due to lack of accessible wells or springs.

All water samples were collected in 250 mL glass bottles designed for ^{222}Rn analysis (WAT-250 system; Durridge Company; Figure 4). In order to minimize the degassing of ^{222}Rn during sampling, all the samples were filled up completely underwater (avoiding contact with the atmosphere) and properly sealed. Most of the samples were analyzed by a RAD H_2O within hours immediately after taking samples. The RAD H_2O (RAD H_2O; Durridge Company) is a portable and automatic ^{222}Rn monitor that provides results with a sensitivity that match or exceed those of liquid scintillation methods. ^{222}Rn activity is determined by counting its alpha-emitting, positively charged radioactive daughters (Po-218 and Po-214), which are detected by a silicon detector. Theoretically, RAD H_2O has a detection limit of 370 Bq/m^3 (in 20 minutes count time) and detection limit can be lowered by increasing the

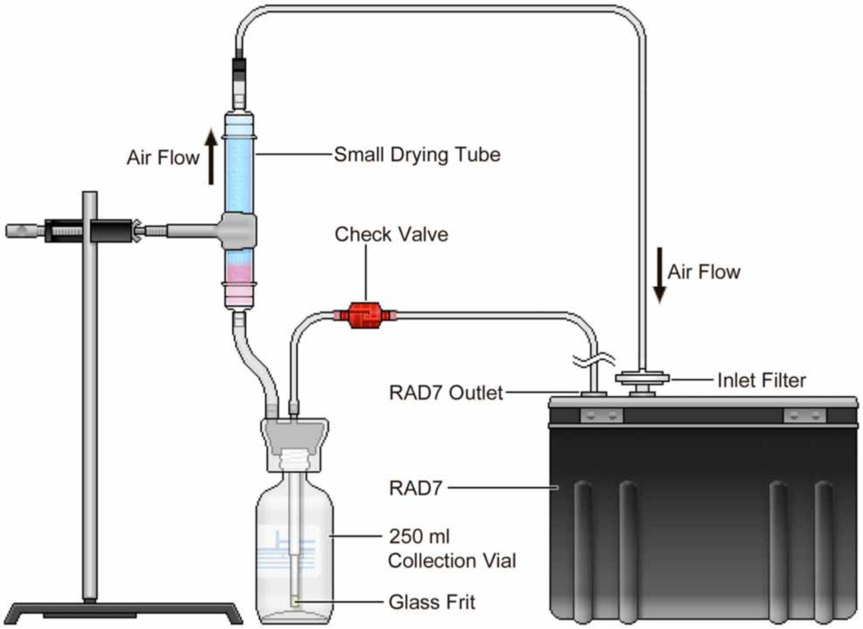

Figure 4 | Illustration of RAD H_2O configuration (after WAT-250 system; Durridge Company, USA).

count time. Water samples with ^{222}Rn activity below the 20 minute detection limit were subjected to counting time of 1 hour or 2 hours in order to achieve acceptable activity values. A few samples were analyzed within 2–3 days, and the necessary correction for the decay was performed following the protocols established by the manufacturer (i.e., the time between the sampling and analysis is considered for the radioactivity decay). The decay correction factor (DCF) is given by the formula $\mathrm{DCF} = \exp(T/132.4)$, where T is the decay time in hours.

A model based on mass conservation theory was used to account for the amount of ^{222}Rn discharged from groundwater to surface water at any location. The mass balance equation can be expressed as follows:

$$C_sQ_s = C_gQ_g + C_b(Q_s - Q_g) \tag{1}$$

where C_s is the concentration of ^{222}Rn in the stream; C_g is the concentration of ^{222}Rn in the groundwater; C_b is the concentration of ^{222}Rn from the upstream, based on estimated loss of ^{222}Rn resulting from the gas exchange and radioactive decay; Q_s is the stream discharge and Q_g is the groundwater discharge.

Therefore, the fraction of groundwater end-member at a possible location can be solved as follows:

$$f = \frac{Q_g}{Q_s} \times 100\% = \frac{C_s - C_b}{C_g - C_b} \times 100\% \tag{2}$$

RESULTS

The result of dissolved ^{222}Rn during the pilot sampling indicates a notable contrast of activity levels (at least by one order of magnitude) between groundwater and surface water in the SAYR (Figure 3(a)). The average ^{222}Rn activity in surface water is 2,371 Bq/m^3, with the minimum and maximum of 103 Bq/m^3 and 9,133 Bq/m^3, respectively. The groundwater contains much higher ^{222}Rn activity than the surface water regarding a range from 10,398 to 41,583 Bq/m^3 with a mean value of 27,835 Bq/m^3.

The ^{222}Rn activity of surface water in the Shuangchagou basin varies from 297 to 19,740 Bq/m^3 with an average value of 6,661 Bq/m^3 (Figure 3(b)). The highest ^{222}Rn activity is observed in the trunk stream (location D2). The south tributary shows a wide range of ^{222}Rn (297–5,880 Bq/m^3) with an average of 2,331 Bq/m^3. Compared with the south tributary, the north tributary has less variation but a much higher average activity of ^{222}Rn (around 8,705 Bq/m^3), spanning from 6,636 to 10,245 Bq/m^3. The continuous monitoring at 2 hourly intervals in locations S1 and D2 is also presented in Figure 5. The ^{222}Rn activity range in S1 is from 3,280 to 9,184 Bq/m^3 while in D2 the range is much wider, from 10,280 to 25,200 Bq/m^3, during the 2 days of the sampling period.

DISCUSSION

^{222}Rn distribution in SAYR

A comparison of the ^{222}Rn activity in the SAYR with some worldwide published data is shown in Table 1. Cartwright *et al.* (2014) used ^{222}Rn as a tracer to quantify groundwater inflow to the King River, South Australia and found up to 10 m^3/m/day. In the arid Limari basin, North Chile, a low ^{222}Rn level (less than 1,000 Bq/m^3) was found in the reservoir waters compared to groundwater (around 20,000 Bq/m^3), indicating that ^{222}Rn is easily lost in surface water (Oyarzún *et al.* 2014). Similarly, a large difference of activity of ^{222}Rn between surface water and groundwater was also observed in China (Wang 2002). Most ^{222}Rn activity measured in surface water of the English Lake District was found to be low, with a few exceptions where samples taken from a limestone area or faulted area indicated direct influence of bedrock and groundwater discharge (Al-Masri & Blackburn 1999). A case study in central Italy, presenting different ^{222}Rn activity in wells, indicates infiltration from the river to the aquifer (Stellato *et al.* 2013). These examples suggest that the difference in activity of ^{222}Rn between surface water and groundwater makes ^{222}Rn a potential tracer of interaction between these systems.

In the SAYR data, at least one order of magnitude difference of ^{222}Rn activity is observed between surface water and groundwater (Figure 3(a)). Although there is no explicit range of typical ^{222}Rn activity in surface water and groundwater, a maximum reference value of about 1,000 Bq/m^3 can be considered for surface water (Bennett 1994; Wang 2002). Green & Stewart (2008) also indicated that ^{222}Rn values greater than 1,000 Bq/m^3 in surface water can be indicative of a recent addition (i.e., groundwater discharge to the surface flow). ^{222}Rn activity in surface water of up to 9,133 Bq/m^3 observed in the SAYR, clearly suggests the influence of groundwater. The SAYR is located in the mosaic transition zones of seasonally frozen ground, and discontinuous and continuous permafrost, which can play a critical role in water exchange process. Distribution and thermal regimes of permafrost and seasonal freeze–thaw processes are closely related to groundwater dynamics.

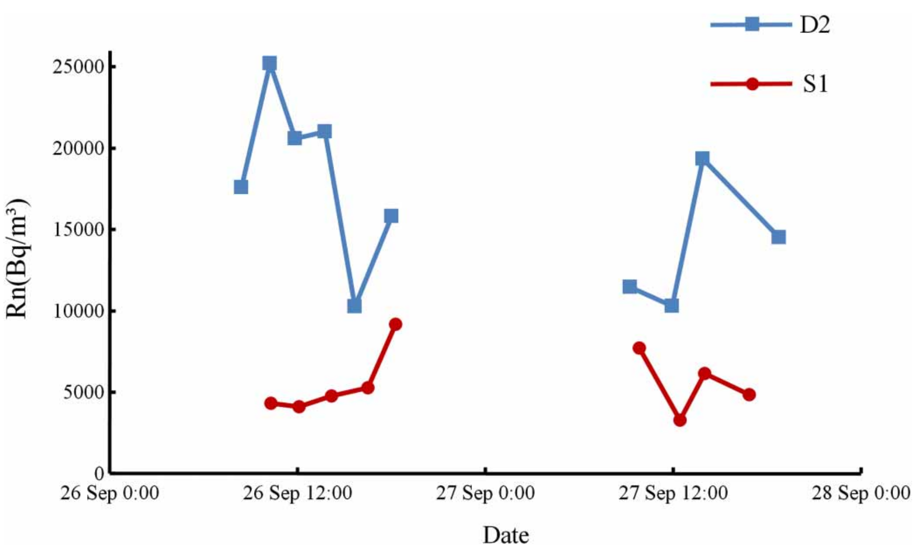

Figure 5 | ^{222}Rn activity measured during 26th and 27th September at an interval of 2 hours, in location S1 and D2.

Table 1 | ^{222}Rn activity (Bq/m^3) in the groundwater and surface water studied here compared with some selected data from the published literature

	Surface water		Groundwater		
Study	Range	Average	Range	Average	Reference
South Australia	122–2,085	735	17,250–76,170	46,629	Cartwright *et al.* (2014)
Central Italy	100–1,300	400	7,000–19,000	14,700	Stellato *et al.* (2013)
North Chile	100–2,500	908	800–21,500	8,494	Oyarzún *et al.* (2014)
England	54–1,288	378	–	–	Al-Masri & Blackburn (1999)
China	10–8,940	1,260	200–3,274,000	81,096	Wang (2002)
SAYR, China	102–9,132	2,562	10,398–41,583	27,809	This case

Most samples in the SAYR are in the continuous permafrost zone, except for samples P8 and P9 (Figure 1). The discontinuous permafrost zone with limited areal extents and thicknesses, is closely linked to groundwater effects from the nearby free of permafrost area. The conditions for groundwater recharge, flow, and discharge are generally good which promote frequent exchanges with the surface water, and the consequent high ^{222}Rn activity in P8 and P9 (Figure 3(a)).

The ^{222}Rn activity in the northwest of SAYR shows values of less than 1,000 Bq/m^3, and thus is consistent with the suggested reference value for surface water. However, ^{222}Rn activity in the southeast of the SAYR is found to be up to around 4,068 Bq/m^3 on average, indicating possible groundwater influence in these places. The continuous permafrost is generally thick and serves as a relatively stable regional aquitard with poor vertical water percolation. As a result, exchange with deep groundwater (e.g., sub-permafrost water) in this zone is meager due to limitations exerted by the permafrost layer. Instead, the shallow groundwater (e.g., supra-permafrost water) is dynamically exchanged with the surface water (Cheng & Jin 2012). The hydrogeology in the continuous permafrost zone, therefore, is characterized by extensive occurrence of aquifer and linear (point or strip) enrichment (accumulation) of groundwater. Another possible explanation of high ^{222}Rn activity in the southeast of the SAYR is the possible degradation of the permafrost. Owing to the warming climate and increasing human activities, the permafrost degradation has been more striking around the Sisters Lakes and in the east of the SAYR, causing the reduction in areal extent from continuous and discontinuous to sporadic and patchy permafrost, thinning of permafrost, and expansion of taliks (Jin *et al.* 2009). The conditions of permafrost may change from aquitard to an aquifer upon thawing or warming to the melting point of water, and talik channels can be formed or enlarged. Accordingly, the high ^{222}Rn activity in the southeast of the SAYR can also be detected as a result of the degradation of permafrost which can also enhance the frequent exchanges of groundwater and surface water.

^{222}Rn in Shuangchagou basin

Contrast in the ^{222}Rn activity is found between the two stream tributaries (Figure 3(b)) in the Shuangchagou basin. Generally, the direct contact of the water with the atmosphere and turbulence effects allow for escape of ^{222}Rn, if present. This situation produces low ^{222}Rn activity in the surface water unless there is a ^{222}Rn source from the sediment decay or groundwater discharge along the stream. However, high ^{222}Rn occurs in S5 (5,880 Bq/m^3) and S1 (4,580 Bq/m^3), while ^{222}Rn activity consistent with the reference value in surface water (less than 1,000 Bq/m^3) is observed in the south tributary (S2, S3, and S4). The sampling area is around 5 km^2, and sample locations are close to each other sharing the same Quaternary clastic aquifer conditions. Possible groundwater end-member (discharging water) would be expected near S1 and S5 in the south tributary. All the samples in the north tributary stream show high ^{222}Rn activity (on average 8,705 Bq/m^3), thus suggesting active groundwater infiltration along the north tributary. From the geomorphological point of view, there may exist a flow path or talik channel that may enhance groundwater flow along the north tributary pathway. The high ^{222}Rn activity in the north tributary is rather in agreement with groundwater discharge along the tributary rather than discharge through the end-member. The highest ^{222}Rn (up to 19,740 Bq/m^3) is observed in D2 which was sampled in the trunk stream near the confluence of two tributaries. ^{222}Rn activity in D2 is much higher than the total contribution from N1 and S1, indicating also possible groundwater discharge near D2.

The variation of ^{222}Rn activity in S1 and D2, observed in Figure 5, may be explained by the variation of daily temperature and the presence of permafrost. The study period (the end of September) is at the transition when ground thawing approaches the end and the freezing period starts. The variation of ground temperature may influence the distribution of the permafrost, which in turn affects porosity and permeability of the aquifer (Luo *et al.* 2014b). The increasing temperature at noon will also cause more ground ice to melt and recharge to the stream while the low night–morning temperature will reduce the recharge. The increasing trend of ^{222}Rn in S1 in the late afternoon suggests incorporation of permafrost partial melting in the groundwater discharge which is most likely due to diurnal higher temperature in the morning and afternoon. Compared to location S1, the larger ^{222}Rn activity variation in D2 is likely due to influences of both the groundwater discharge and input of the two tributaries.

Here we ignore the gas exchange with the atmosphere and decay loss as the low activity in the upstream and at the sampling locations is comparable. Also, ^{222}Rn activity in the groundwater can barely be monitored as no springs or wells exist nearby. Therefore, assumption of the average ^{222}Rn input from the upstream of 297 Bq/m^3 (the measured value in S2), ^{222}Rn activity in groundwater of 27,835 Bq/m^3 (the average ^{222}Rn activity of groundwater measured in the SAYR), and the mean daily ^{222}Rn activity monitored during the 2 days of sampling were considered. The faction of groundwater discharged in the possible groundwater end-member (location S1) can be calculated by using Equation (2). This calculation provides a minimum estimate assuming that ^{222}Rn is not subjected to any atmospheric evasion or decay losses through downstream transit. The average contribution of groundwater to the surface water is 19% on the first day, while a similar value (18.9%) is also estimated for the second day. This is the minimum groundwater input value, as larger inputs would be expected if correction for atmospheric evasion and ^{222}Rn decay is included. The result is also consistent with the data in other places of the Qinghai-Tibetan Plateau (20–48% groundwater input; Yao & Yao 2010) and in other permafrost areas of China (12–15%; Liao *et al.* 2008). Although we could not estimate the groundwater contribution to the whole water budget in Shuangchagou basin because of limited samples, we emphasize that the estimation given here could act as a reference value for groundwater input to surface water.

The ^{222}Rn activity in groundwater end-member is crucial for the application and may be a source of uncertainty (Burnett *et al.* 2007). For example, Santos & Eyre (2011) estimated an uncertainty of 28% in the final discharge rate for the variability in groundwater end-member. The groundwater ^{222}Rn activity calculated here is estimated by the mean value of 27,835 ± 10,869 Bq/m^3 ($n = 7$) in the SAYR. This value when combined with temporal variability, may result in 40% uncertainty of ^{222}Rn activity in the estimates of groundwater discharge. A larger number of groundwater samples both in the Shuangchagou basin and the SAYR should be collected in future work in order to reduce the uncertainties.

The ^{222}Rn activity approach presented here has proved to be an efficient and inexpensive method to assess groundwater input even when detailed hydrogeological information is not available. Especially for the harsh environment of the Qinghai-Tibet Plateau, where hydrological experiments are difficult to perform, the method becomes even more valuable. However, further investigation including seasonal ^{222}Rn measurements coupled with ^{226}Ra are needed to comprehensively explore the interaction between groundwater and surface water in the Shuangchagou basin.

CONCLUSION

Measurement of dissolved ^{222}Rn in the SAYR provided a simple and effective tool for assessing the surface water and groundwater connectivity in the Qinghai-Tibet Plateau area. Distinctive ^{222}Rn activity values are observed in the surface water and groundwater indicating specific characterization of each water type. Applying a simple ^{222}Rn mass balance model, the fraction of groundwater contributes around 19% to the surface water in location S1. Future continuous measurement and expansion of the base line data in the region will facilitate tracing of exchanges between surface and groundwater and thereby provide a monitoring system for changes in the water supply to the Yellow River. The data will also provide an interesting monitor of the permafrost conditions once detailed data of ^{222}Rn activity distribution are acquired.

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