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## Using <sup>222</sup>Rn to identify and quantify groundwater inflows to the Mundo River (SE Spain)



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#### ABSTRACT

Groundwater discharge to the Mundo River (SE, Spain) has been investigated from 2011 to 2013 by means of <sup>222</sup>Rn activities in river water and groundwater. Starting nearby the river source, some 50 km of river channel have been studied. The Mundo River is located in the water stressed region of the Segura River Basin. Identifying and quantifying groundwater discharge to rivers is essential for the Hydrological Plan of the Segura Basin Authority. Four main areas of groundwater discharge to the river have been identified by means of <sup>222</sup>Rn. Moreover, groundwater fluxes have been quantified using radon activities and, when possible, have been validated with chloride mass balances. The uncertainty range  $(\pm 2\sigma)$  of all water balances has also been assessed. Groundwater discharge  $(Q_{GW})$  values estimated by radon mass balances (RMB) and chloride mass balances (CMB) were similar in the river tracts and/or dates in which surface inputs from tributaries were null or negligible. This adds confidence to the  $Q_{GW}$  values estimated by RMB in the reaches were CMB could not be performed due to the existence of ungauged surface inputs, as is the case of the upper basin of the Mundo River, as well as to the applicability of the method to similar situations. Quantification of groundwater discharge allowed identifying Ayna zone as the main gaining reach of the studied area, with up to 29,553  $\pm$  8667 m<sup>3</sup> day<sup>-1</sup> in year 2011. Overall, the total  $Q_{GW}$ estimated by means of RMB for the studied area was 8-16% of the total river flow. The results are coherent with the meteorological conditions of the study period (average rainfall around 450 mm/y) and also with the undisturbed situation of the aquifers discharging to the Mundo River in the considered area.

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#### 1. Introduction

Groundwater discharge into a river system plays an important role in the hydrological and ecological functions of the river, especially in arid or semiarid regions where perennial streamflow is mainly sustained by aquifer-derived flow. Surface water and groundwater have been often considered two separate entities and investigated separately (Kalbus et al., 2006); however, understanding the processes regulating the complex interactions between rivers and adjacent groundwater systems is critical to water resources management, and they have to be considered components of a unique hydrologic system (Andreu and Solera, 2002; Sahuquillo, 2004; Custodio, 2007; Pulido-Velázquez et al., 2007). In the European Union (EU) water management policies take this issue into account by means of the European Water Framework Directive [2006/60/CE] and the Groundwater Directive [2006/118/EC]. The EU legislation aims for an integrated management of surface water and groundwater.

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Quantifying the relationship between these resources and the locations of groundwater discharge to surface waters is fundamental for developing conceptual models of water systems at catchment scale.

Groundwater contribution to rivers can occur as either discrete flow from springs or diffuse seepage through the river bed. A variety of physical, chemical and numerical methods have been developed to estimate groundwater discharge to a river. However, the heterogeneities and the problems of integrating measurements at different scales are still a challenge to determine and quantify water interactions (Sophocleous, 2002; Kalbus et al., 2006; McCallum et al., 2012).

Natural chemical tracers can be useful tools to identify and quantify groundwater fluxes to rivers as they can provide an integrated view of a large area over a long period of time. The possibility to use chemical tracers depends mainly on the contrast between the concentrations of the tracer in groundwater and surface water. The accurate characterization of tracer's sources and sinks is also essential to identify the groundwater component for adequate mass balance calculations. Furthermore, factors such as the spatial variability and the potential for the tracer to change by evaporation, precipitation, radioactive decay, degassing, or biochemical reactions also play an important role (Unland et al.,

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2013). Major ions (for example Cl<sup>-</sup>), stable isotopes and radioactive isotopes such as <sup>14</sup>C, <sup>3</sup>H, <sup>222</sup>Rn, <sup>226</sup>Ra, <sup>238</sup>U/<sup>234</sup>U, and <sup>87</sup>Sr/<sup>86</sup>Sr are often used to investigate river–groundwater interactions. <sup>222</sup>Rn (hereinafter referred to as radon if otherwise not needed) has proven to be an excellent tracer in river–groundwater interactions and its use has increased in the past decade as the ease of measurement has improved (Ellins et al., 1990; Genereux and Hemond, 1990; Genereux et al., 1993; Cable et al., 1996; Corbett et al., 1997; Cook et al.;, 2003, 2006; Lamontagne et al., 2008; Spizzico, 2005; Dulaiova and Burnett, 2006; Lamontagne and Cook, 2007; Mullinger et al., 2007; Stellato et al., 2013; Burnett et al., 2010; Peterson et al., 2010; Cartwright et al., 2011; Gilfedder et al., 2012; Smerdon et al., 2012; McCallum et al., 2012; Guida et al., 2013; Unland et al., 2013).

Radon is a naturally occurring radioactive noble gas whose concentration is 2–3 orders of magnitude greater in groundwater than in surface waters (Burnett et al., 2001). Natural radon consists of several short-lived radioactive isotopes. Due to their half-lives, the isotopes with some interest for environmental applications would be  $^{210}\rm{Rn}$  [2.4 h],  $^{211}\rm{Rn}$  [14.6 h],  $^{220}\rm{Rn}$  [55.6 s],  $^{222}\rm{Rn}$  [3.8235 day], and  $^{224}\rm{Rn}$  [1.8 h], but due to its half-life  $^{222}\rm{Rn}$  is the most useful and commonly used for groundwater studies.  $^{222}\rm{Rn}$  decays through a long chain of alpha and beta processes involving short-lived radioisotopes into stable  $^{206}\rm{Pb}$ . The  $^{222}\rm{Rn}$  decay is an alpha process producing first  $^{218}\rm{Po}$  (3.10 min half-life), whose accumulation is used to measure  $^{222}\rm{Rn}$  concentration.

The source of <sup>222</sup>Rn in groundwater is <sup>226</sup>Ra, also the longest-lived of radioactive radium isotopes, with a half-life of 1601 years. <sup>226</sup>Ra derives from uranium decay and is ubiquitous in sediments and sedimentary rocks. Thus, the amount of <sup>222</sup>Rn present is primarily determined by the lithology and geochemical composition of the aquifer material (Cecil and Green, 2000; Sakoda et al., 2011) as well as by the time from release into groundwater and the sampling moment. Radon tends to concentrate in areas of tectonic disturbance, and in faulted areas in general, as a result of intense degassing fluxes due to the local high permeability of the bedrock and soil (Ellins et al., 1990; Etiope and Martinelli, 2002; Ioannides et al., 2003). Moreover, as a result of its relative high solubility (Ball et al., 1991), in fractured aquifers radon can be transported long distances from its point of emanation by fast water flow before decaying. Thus, anomalous activities can be present in areas without uranium and radium enriched rocks. Similarly, radon activity can be higher than expected in aguifer areas where thermodynamic conditions induce uranium and/or radium concentration changes by adsorption or precipitation in mineral phases (as secondary or trace components). Hence, the presence of clayey sediments may induce increasing of radon activity at aquifer scale through the accumulation of uranium and radium. These processes have to be considered to use radon in groundwater flow studies, but are less relevant when using it to identify and quantify groundwater discharge to rivers.

The activity of radium in rain water is negligible. Its occurrence in surface waters is derived mostly from groundwater discharge, but due to a relatively short half-life of <sup>222</sup>Rn and the rapid gas exchange to the atmosphere, high radon activities are only present in river locations where there is active groundwater discharge. Although radon surveys are performed on a local approach, the technique provides information at a regional scale, being a good tool for water management (McCallum et al., 2012). Additionally, radon data combined with river discharge and physical dimensions can be used to quantify groundwater discharge by means of radon mass balances. On the other hand, quantifying groundwater discharge rates involves many parameters that are often difficult to measure. Consequently, the associated uncertainty needs to be calculated and made explicit to understand the meaning of the estimated flows.

The aim of this work is to determine and quantify the relationship between groundwater and surface water in the Mundo River using radon activity. The Mundo River is located in the upper Segura River Basin, a water stressed region in SE Spain. Since three decades ago the region receives temporary imported water resources to supply the high water demand areas in the middle and lower parts of the Segura

River Basin. Thus, identifying and quantifying groundwater discharge to rivers, and especially to the Mundo River, which is under natural flow conditions, is essential for the water action plan (Hydrological Plan) of the Segura River Basin. The contribution of this tool and its results would also help in making decisions and adopting policies of sustainable water use for the 2015–2020 Environmental Strategic Plan of the Segura River Water Authority (www.chsegura.es).

To this purpose, three main objectives were established: i) to identify preferential groundwater discharge areas in the Mundo River, ii) to check the performance of radon as a tracer to quantify groundwater discharge to rivers in karstic areas, and iii) to quantify groundwater flows using radon mass balances, among other techniques, as far as using a multi-technique approach to provide more robust information on groundwater–surface water studies.

#### 2. Site description

The Mundo River is a perennial stream with a 2400 km² catchment area located in SE Spain. The river originates from a spring in the mountain plateau Calar del Mundo, forming a 300 m high waterfall known as Los Chorros del río Mundo. The river flows NE for 107 km, being the main tributary of the larger Segura River, which flows from W to E into the Mediterranean Sea. The studied area encompasses 50 km of the Mundo River and extends from the gauging station n° 100 in Fig. 1, 2.09 km downstream the headwater, to station n° 117, 42.76 km from the headwater and 5 km upstream of the Talave Reservoir, where the Mundo River receives water transferred from the Tajo River Basin, in NE Spain, through a 250 km aqueduct.

Following SGOP (1988), the studied river tract is mainly fed by groundwater discharge from two Mesozoic carbonate-rock aquifers, the Pliegues Jurásicos del Mundo and Alcadozo (Fig. 2), and from several tributaries: De la Vega, Salado, Celada, Provencio, Vadillos, Bogarra, Cañadas, and Talave creeks, joining the Mundo River at 3.3, 3.5, 7, 10, 20, 22, 33, and 45 km downstream the river source, respectively (Fig. 1). Only the Bogarra creek is permanent. The De la Vega and Celada creeks are fed mainly by small springs from the Calar del Mundo and Cujón aquifers and by rainfall (Rodríguez-Estrella, 1979; IGME, 2009). The Salado, Provencio, Vadillos, Cañadas, and Talave creeks flow only during and after intense rainfall events.

The basin has a strong topographic gradient from W to E, with heights varying from around 1600 m asl at the western part to around 300 m asl at the eastern sector. The precipitation also displays a regional gradient with average values from around 600 mm/y to around 300 mm/y in the same places (CHS, 2014). Fig. 3 shows the yearly precipitation during the study period at the Bogarra station (AEMET, 2013), located at the central part of the upper basin. The sampling surveys were performed in dry months. Years 2011 and 2012 were quite dry; some significant rainfall took place only in the 2012 winter. There was no rainfall in the area 48 h prior and during all field surveys, except for July 9th 2013, when a small rainfall event of 3.2 mm was measured in the Bogarra station (CHS, 2013).

Stream flow data at the lower part of the Mundo River studied area comes from the only automated gauging station existing in the river (station 116 in Fig. 1). The recorded yearly flow varied during the study period from around 121 \*  $10^6\ m^3\ year^{-1}$  in 2011 to  $96*10^6\ m^3\ year^{-1}$  in 2012, and to 233 \*  $10^6\ m^3\ year^{-1}$  in 2013 (Fig. 3).

The studied area comprises carbonated and detrital rocks of Triassic, Jurassic and Cretaceous ages. The area is characterized by a complex geologic structure and consists of a succession of folds and thrusts with NE–SW direction and verging towards the NE (García-Palomero, 1969). The tectonic complexity facilitates both diffuse and concentrated groundwater discharge to the river and induces the existence of several aquifers in the area. The Pliegues Jurásicos del Mundo and Alcadozo aquifers are mainly formed by Jurassic limestones and dolostones overlying Triassic sandstones and clays containing evaporites, mostly gypsum and anhydrite, with a minor presence of halite (IGME, 1974a, 1974b, 1975a,

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