

Response to comment #C6187 (reviewer 2)

Preamble

Firstly, we would like to thank the reviewer for the clear, detailed commentary arising necessarily from an in-depth review of the manuscript. As with the first reviewer (we note that there are points in common), this has benefitted us greatly. The reviewer makes a substantial number of comments, divided into “concerns” and “specific” according to their nature. In the interest of clarity and to facilitate cross-referencing, we have transcribed them into the response and have numbered them according to the original order and type: #1-#2 for the “concerns” and S#1-S#23 for the specific ones. The response will address these individually.

We should also state beforehand that an entirely new additional table, providing an overview of the sampling campaigns and associated precipitation data (Table 2) has been added to the original manuscript, that winter radon mass balance data (2009) illustrating the similarity we had mentioned existed with the summer radon mass balance (2010) and addressed in S#16 has been added to Table 1; and two of the original figures (Fig 1 and 4) were updated to help clarify the queries associated with concern #1 and comments S#9, S#12-S#16 and S#22. As such, the redrawn Figure 1 now incorporates two panels, in order to provide a more complete geographical context. It now includes a) the location of all the six inlets mentioned in the text as well as the river Gilão (panel a), and b) additional information pertinent for clarifying pore-water sampling locations for the periods 2007 and 2010-2011, other suggested location references as well as a clear cut definition of the western and eastern sectors of the lagoon area focused upon in this study.

The redrawn Figure 4 on the other hand now includes a fourth panel illustrating the daily precipitation record over the region (2006-2013), taken from public databases, on which we have superimposed the periods of sampling that are relevant to the study to provide a temporal context on the precipitation regime over the region. Hopefully, this will make it easier to follow the discussion and help clarify issues raised in specific comments #1, and S#9 and S#13.

Responses

Main concerns:

#1. “My major concern with the study is the seemingly ad hoc sampling design. The authors state for example that “Samples for stable isotope analysis of water were collected in triplicate from all possible water sources to the lagoon on various occasions between 2007 and 2013.” (P12446 L1); and “Quasi-synoptic distributions of 18O and 2H in water at different tidal stages were obtained for the lagoon in the winter of 2009.”(P12446 L15). With so much temporal variation in all of the tracers, drawing conclusions from multiple sampling campaigns under differing conditions can be problematic to say the least. This is of particular concern with the natural tracers used as concentrations and fluxes of the tracers would very much be affected by rainfall, tide heights etc. While comparing the results of different campaigns can be done, to do so, it would be necessary to demonstrate that the system was operating under similar hydrological conditions during each of the campaigns. To do this, reporting differences in rainfall (both long and short term), temperature, groundwater water levels, groundwater concentrations/signatures and tide heights would be necessary.”

R: This is a fair comment, and raised again in specific comments S13 and S16. A new additional table, providing an overview of the sampling campaigns and associated precipitation data (Table 2) has been added to the original manuscript. Furthermore, winter radon mass balance data (2009) illustrating the similarity we had mentioned existed with the summer radon mass balance (2010) and addressed in S#16 has been added to Table 1; and two of the original figures (Fig 1 and 4) were updated to help clarify this concern and queries related to comments S#9, S#12-S#16 and S#22.

The new section on inter-comparability and the additional data provided might contribute to clarify the issue. This section now reads:

‘Inter-annual comparability of isotopic data

Sampling campaigns were carried out strategically following a field-adaptive protocol. Of primary concern was to capture the extent of temporal end-member variability in isotopic signature under maximum freshwater flow (hi-flow) conditions, in order to a) guarantee coherence of source compositions to feed into mixing models when necessary while assessing the hydrology of the lagoon over wider temporal scales and b) minimizing logistics and costs while guaranteeing inter-comparability. For this purpose, winter season was chosen given that ~61% of the mean annual precipitation falls on the region between November and February (34% in the months of December and January). Stable isotope sampling in winter had the added advantage of minimizing kinetic effects over stable isotope signatures given the lower evaporation potential. Sampling in winter 2007 was exploratory, with two main objectives: firstly, to characterize isotopic signature of M12 groundwater and surface lagoon waters in the western sector, particularly in the area that could be potentially influenced by both SGD and the WWTP outflow under maximum dilution potential (hence high tide), and secondly, conduct an exploratory survey of potential seepage areas along the Ancão peninsula, keeping in mind that the location of at least one of the important SGD seepage sites was known (Leote et al, 2008). Detection of the isotopic signature of groundwater in porewaters at the seepage face at stations Pw_e and Pw_f (Table S1) led to the installation at their location of a nested piezometer transect array in January 2010. This was subsequently used to obtain porewater samples in the 2010/11 winter season (December 2010 and January 2011).

To capture inter-annual variability, the M12 aquifer was sampled twice (winters of 2007 and 2009), with the provision of one common location (Ramalhete) for cross-referencing. Following the same reasoning, the M10 aquifer was sampled in December 2010 while simultaneously sampling Rio Seco (belonging to M12, Table S1). This ensured inter-comparability between groundwater isotopic signatures in 2009 and 2010. Campaigns were planned in advance considering the precipitation over the region to ensure similarity in the hydrological regime and ultimately guaranteeing inter-comparability of results. The sampling itself took place in dry conditions as much as possible, and never after intensive rain that could have promoted flooding (Table 2, Fig 4d). For example, while January 2007 was a dry month (8.8 mm) compared to the historical average (138 mm), the accumulated precipitation during the previous 3 months was 369.7 mm, consistent with the historical average (Table 2). By contrast, both December 2009 and 2010 were relatively wet months (392.2 and 269.6 mm), but followed relatively dry 3-month periods (Table 2). So porewater samples were also taken in January 2011, hence complementing winter 2010/2011. January 2011 followed a wet three-month period (414.7 mm) and was hence comparable with January 2007, also relatively dry but on the back of three wet months (369.7 mm cumulative). The combined dataset therefore contains results from repeated measurements for end-member isotopic composition under hi-flow conditions, across different years. These are

in addition compared to historical data (table S1, Figure 4), leading to a temporally coherent quantitative overview of stable isotopic hydrology over the catchment.'

#2

Another concern is with the selection of an endmember for seawater recirculation. The authors concluded that most of the SGD is comprised of recirculated seawater, but does the beach groundwater endmember represent fully equilibrated recirculated seawater or new seawater with a very low residence time that has yet to fully equilibrate. The authors assume it is fully equilibrated, but it needs to be discussed why they believe this is so.

R: See also response to S#23 below reading the recirculation mechanism and its impact on radon budgeting.

If we assume that the water (from the lagoon, only partly open seawater) infiltrates the unsaturated zone of the beach during flood and is flushed through the permeable zone of the beach driven by tidal pumping, then as part of this endmember, we get radon produced from the sediments during the residence time of the water in the beach plus the radon which was already in the water before recycling through the beach sediments. This recirculation pump is constant, i.e. the residence time of the water in the beach is constant over time, so the added fraction of radon from sediments may be seen as constant. Therefore, we don't require or assume that it is fully equilibrated.

Under the assumptions above, and those pertinent to the calculation of the contribution of re-circulated water to the SGD in the lagoon (lifetime of radon), we always get the same amount of radon from this production in the sediments as long as the recirculation time scales are seen to be constant. So we have an endmember which is probably partly equilibrated (because the residence time within the beach is probably too short for full equilibration to occur) but which is constant; and this endmember was measured, several times (Table 1).

Specific comments

S#1: P12435 L1. I find the first and second sentences contain too many distinct points and both can be written more clearly with shorter sentences.

R: We split the sentences that were too long in order to hopefully make the section clearer. This now reads:

'Natural radioactive tracer-based assessments of basin-scale Submarine Groundwater Discharge (SGD) are well developed. However, SGD takes place in different modes and the flow and discharge mechanisms involved occur over a wide range of spatial and temporal scales. Quantifying SGD while discriminating its source functions therefore remains a major challenge. Yet, correctly identifying both the fluid source and composition is critical. When multiple sources of the tracer of interest are present, failure to adequately discriminate between them leads to inaccurate attribution and the resulting uncertainties will affect the reliability of SGD solute loading estimates.'

S#2: P12435 L19. This is confusing, I suggest something like "SGD can be separated into fresh groundwater inputs and recirculate lagoon waters" to make it a bit clearer.

R: We rewrote the section as suggested to try and avoid confusion, but kept the net water input/no net water transfer designations in, as they are important because of the putative association with autochthonous and allochthonous nutrient inputs if a nutrient mass balance is desired. The section is hopefully made clearer, and now reads:

'Using this approach, SGD in the Ria Formosa could be separated into two modes, a net meteoric water input and another involving no net water transfer, i.e., originating in lagoon water re-circulated through permeable sediments.'

S#3: P12435 L21. I believe "permanent" is the wrong word here as it implies long, multi temporal sampling. Perhaps "dominant" is a better word.

R: Agreed. We have changed accordingly.

S#4: P12435 L26 Remove "so more difficult to predict".

R: Done. The latter part of the sentence now reads: '(...) while the latter is an occasional allochthonous source capable of driving new production in the system'

S#5: P12436 L9. Suggest including a more recent estimate such as: Kwon, E. Y., G. Kim, F. Primeau, W. S. Moore, H. M. Cho, T. DeVries, J. L. Sarmiento, M. A. Charette, and Y. K. Cho (2014), Global estimate of submarine groundwater discharge based on an observationally constrained radium isotope model, Geophysical Research Letters, 41(23), 8438-8444.

R: This is an important point. We modified the paragraph and it now reads: 'Indeed, on a global scale, an estimated 6 % of the freshwater input into the sea, carrying an anticipated 52% of the total dissolved salts crossing the land-ocean interface, was estimated to occur via SGD-Submarine Groundwater Discharge by Zektser and Loaiciga (1993). This early estimate has since been updated by Kwon et al (2014), who show that global SGD is 3-4 times greater than the freshwater flow into the oceans by rivers. This revision means that SGD is by far the largest contributor of terrestrial solutes to the global ocean, hence implying that some global biogeochemical budgets of major elements need revision. Yet, mass flows defining the contribution of SGD to coastal biogeochemical budgets are difficult to quantify in a systematic way (Burnett et al. 2001a).'

S#6: P12437 Radium is normally absent in "fresh" groundwater.

R: Good point. Sentence was amended.

S#7: P12438 L9 Sentence is unclear.

R: Yes, we can see this – it has been a point where we have become somewhat stuck for a while. We have attempted to re-write it in order to make it clearer. The section now reads:

'Linking these two datasets requires care and is underpinned by our ability to correctly identify and quantify the different SGD pathways into any one system. This is because the final SGD solute-load estimate not only depends on how accurate our recognition of the SGD source functions is, but also on the ability to track their path within the system, since this is required to evaluate the biogeochemical history of the source components prior to their mixture into receiving waters.'

S#8: P12438 L13 Remove "so far to progress beyond our ability".

R: Done.

S#9: P12438 L13. I believe this can be expanded upon. The endmember is usually the greatest uncertainty in any tracer mass balance. With most studies using a range of endmembers across the catchment/aquifer/study site, determining the endmember concentration in the area of the likely source of groundwater would very much lead to much less uncertainty in SGD estimates.

R: This is a well-made point and a very useful synopsis of the logical thread, and we followed by incorporating the suggestion. The section now reads: 'Not fulfilling this requisite therefore constitutes the major obstacle to prognosticate upper boundary or 'potential' SGD-related impact, and more importantly, confidently attribute causality. Indeed, the endmember is usually the greatest source of uncertainty in any tracer or solute mass balance. It follows that determining the endmember concentration in the area(s) most likely to be the source(s) of groundwater would decrease uncertainty in SGD estimates, on the one hand, and in biogeochemical budgets derived from those estimates on the other. The current panorama of SGD research at the system scale therefore begs the question of which end-member to use when selecting a source solute concentration in attempts to quantify pollutant fluxes associated with SGD.'

S#10: P12438 L19. I believe a separate paragraph (of which some of the information occurs in the last paragraph of the introduction) on how O18 and 2H can be used and where they have been used to quantify SGD sources.

Considering also comment S#11 below, and the common points made by reviewer 1, we opted to re-write the last two paragraphs of the introduction in order to accommodate what we felt were very valuable contributions.

These now read: 'We contribute an answer to this conundrum with a study conducted in a seasonally hypersaline lagoon in southern Portugal where we combine two datasets: radon surveys are used to determine total SGD in the system while stable isotopes in water (^2H , ^{18}O) are used to specifically identify SGD sources and characterize active hydrological pathways. We show that, in combination with radon budgeting, stable isotope hydrology is a reliable tool to identify different SGD sources in a very complex coastal system, even though it hasn't been used to this end before. This underuse of the methodology has two main reasons. The first is a disciplinary divide: the technique has been the domain of freshwater hydrologists; correlations between $\delta^{18}\text{O}$ and $\delta^2\text{H}$ are central to research into the effect of evaporation and mixing on surface waters (Gat et al. 1994, Gibson and Edwards 2002) and contribute to the disentanglement of different water sources affecting catchments (Rodgers et al. 2005). The other is the paucity of paired $\delta^{18}\text{O}$ – $\delta^2\text{H}$ data on coastal seawater (e.g., Rohling 2007), even if stable isotope datasets might help constrain the origins of freshwater inputs into the ocean when coupled with salinity data (Munksgaard et al. 2012, Schubert et al 2015), or as part of a methodological arsenal in SGD studies combining physical and chemical measurements with radioactive and stable isotope tracers (e.g., Povinec et al 2008). Hence we also bridge the disciplinary gap between marine chemists and hydrogeologists currently extant in SGD studies by using a combined approach merging techniques from both disciplines.

The occurrence of SGD comprising significant freshwater contributions was first detected in the Ria Formosa in 2006–2007 and subsequently described as a prominent source of nutrients, in particular nitrogen derived from fertilizers, to the lagoon (Leote et al. 2008; Rocha et al. 2009; Ibánhez et al. 2011, 2013). However, the unpredictable nature of freshwater availability in the region, coupled with a mixed-source (i.e., a variable mix of groundwater abstraction and surface water collected in reservoirs)

management of public water supply to meet demand (Monteiro and Costa Manuel 2004; Stigter and Monteiro 2008), made it unclear whether meteoric groundwater would be a persistent feature of SGD in the system. This made it difficult to clarify the contribution of SGD to the nitrogen budget of the Ria Formosa, with obvious consequences to environmental management strategies. The overarching aims of the study were therefore to identify the sources of SGD, distinguish its component parts and elucidate the mechanisms of their dispersion throughout the Ria Formosa. The outcomes are then employed to distinguish and quantify nitrogen loads carried into the lagoon by different SGD modes.'

S#11: P12438 L15. There quantification of N inputs into the lagoon has not been set up in the introduction but is mentioned in the abstract, methods etc.

R: See response to comment S#10.

S#12: P12444 L22. Unclear why this input is not included. Is it are large potential source, small one, what is the discharge? Rivers of course can be large sources of tracer and nutrient inputs particularly in times of flood. This should be acknowledged, shown on figure 1 & 2 and addressed as a limitation if no data is available.

R: This is an issue raised by reviewer 1 as well. We should clarify that we are not neglecting surface water inputs as our statement (P12444 L19-23) indicates: 'Usually, an additional term accounting for the radon influx via river flow is added if the water and particulate flux associated with river discharge is significant. However, the only perennial river in the Ria Formosa is the Gilão, located in the eastern limit of the lagoon'. We then explain why we don't think the input is significant.

Firstly, we clearly stated (Section 2.2. Hydrogeological setting, P 12440) that the average salinity found throughout the year within the lagoon (35) was high – due to low effective precipitation on the catchment, a statement supported in the literature which the reviewer 1 requested be removed (#T3). We nevertheless measured surface salinity (Table S1) during our isotope sampling campaign - it was very high during both tidal stages over the whole lagoon, with the exception of the areas influenced by discharge of the WWTP, where it was slightly above 33 (table S1). Hence surface freshwater inputs, other than the WWTP where generally negligible, something that is consistent with previous studies reported in the literature. In addition, we note that we also compared the annual effective precipitation over the whole catchment with the tidal exchange flux – it makes it clear that the mean volumetric tidal flux is 8 times higher than the annual average effective precipitation – thus compounding the argument above in that surface water inputs are negligible in this lagoon.

Secondly, Newton and Mudge (2003), cited in Mudge et al 2008, find that any freshwater influences caused by the Gilão river (in winter, where the potential to do so would be maximized) are localized to the vicinity of its estuary. Even so, we measured salinity (Table S1) at the Gilão estuary mouth in December 2010 (same month, same tidal conditions as in Dec 2009, same meteorological conditions when the isotope data was collected, see Figure 4 panel d), just to make sure - and it was very high (>29) – this is very common occurrence – the saline influence extends far inland into the river. Freshwater discharge into the sea is negligible except under flooding, which did not occur at any time during sampling or beforehand.

To reiterate the importance of the distance factor, we also clearly state that the location of the estuary is important as is its intermittency of discharge (P12440, L16-18) – it is more than 20 km to the east of the eastern border of the area of study represented in the original Fig 1, as the redrafted figure (top panel) now shows clearly. Combined with a

perennial eastward alongshore drift on this coast, the lifetime of radon in surface waters subject to degassing, and the overwhelming contribution of seawater (low Rn) to the discharging mixture at the estuary mouth, the facts are strongly in favour of our contention that the Rn inputs eventually brought into the area of interest by the discharge of the river Gilão are not significant, and certainly, just in terms of freshwater contribution, not even comparable to the WWTP if we go as far in detail as we can and look at our salinity data for the isotopic samples, so we simplify the equation to remove the contribution.

We also verified whether there was any intense precipitation prior to the sampling campaigns that could have led to flooding – see additions to section 3.2, the new Fig 4, panel d, Table 2

S#13: P12446 L1 As discussed above in the general comments, you need to provide specific information on when the sample collection took place and how comparable the different campaigns are. To do this, a minimum of reporting differences in rainfall (both long and short term), groundwater water levels, temperature, groundwater concentrations/signatures and tide heights would be necessary.

R: This is a fair comment and has now been addressed by adding the information requested. We clarify that we had thought carefully about this issue, albeit tensioned against space constraints since the length of the paper was an important consideration. We originally opted to save some space by providing Table S1 (Supplementary materials) as a way in which the reader could have access to the sampling dates and all the data plotted. In addition to the sampling dates in table S1, the sampling periods for groundwater source functions were described in Section 4.2.1 (P 12450 L5-7), where we also drew attention to the temporal similarity in stable isotope signatures of the groundwater end-member (L 7-11).

While revising, we also found some typos – one location (Rio Seco, 08/12/2010, table S1) was mistakenly attributed to the Eastern sector and 2013 is an error.

We have corrected these, tightened up the designations, and provided a discussion of inter-comparability of all the campaigns as suggested in an update to section 3.2, which as a result was comprehensively revised. We complemented this with a new table (Table 2), where we provide a summary of the precipitation during all the sampling campaigns compared to the historical record average, as well as a new panel, added to Figure 4, comprising the daily precipitation record for 2006-2013 in order to provide a wider temporal context to the stable isotope data plotted there and in subsequent figures.

S#14: P12447 L19. A better explanation of the exclusion of winter data should be given. If higher evasion rates were likely during winter than why were Rn concentrations and distribution similar. This points to very different drivers of SGD temporally and as the comments above suggest, that comparing tracer concentrations over multiple campaigns is problematic.

R: This is also an issue raised by reviewer 1. We respectfully disagree here, since we specifically mention uncertainty associated with the evasion term, and do not mention whether it would be higher. We opted originally to exclude the data and just mention it in the current context for two main reasons:

Firstly, the relative uncertainty associated with the advective radon input to the lagoon derived from the winter data is $\sim 120\%$ of the estimated discharge ($7.97 \pm 9.62 \text{ Bq m}^{-2} \text{ day}^{-1}$). Given the variable extremes observed in wind conditions during the survey (see additions to Table 1) and resulting choppy seas (we call attention to the precipitation

data on table 2 and panel d in Figure 4, where it is very clear that stormy conditions where fast developing and we were actually very fortunate to have carried out the work in the first place), we accepted the fact that both the uncertainty associated with the evasion term and that linked to in-way radon activity measurements (see additions to Table 1) were indeed too great and not representative of usual conditions in the region – the resulting SGD estimate, while similar to that obtained in June 2010, would then be severely affected as we point out, and now make explicit in the additions to Table 1, for completeness. We then took the option of repeating the radon survey the following summer. Even so, as reviewer 1 points out in comment #11, ‘the representativeness of the given SGD mean value is rather low and associated with a lot of uncertainty. (...)’. This is of course a well-known fact in SGD radon tracer studies and is well documented in the literature – it is associated with the assumptions needed to close whole basin mass balances of radon, and within these, in particular to the limitations associated with fluxes estimated with a parameterisation of gas exchange (k) with the atmosphere, as shown by Gilfedder et al (2015). If data that we present and discuss, obtained under the best possible circumstances and attention to detail in order to minimize uncertainty give rise to this commentary, discussing the extra data in addition would increase the space used (it is already a rather long paper) and probably give rise to many more comments of the same nature, while failing to add anything of note, as:

Secondly, we had actually stated that the data was similar as to activity range and distribution, and explained why we chose not to showcase the extra data – it would be redundant as the derived SGD discharge magnitude and the Rn activity range and distribution was similar (this is now made obvious in Table 1). It didn't and still doesn't add anything to our point in the context of the paper. Nevertheless, our calculations, as presented before for the summer (2010), and now on their entirety with the additions made to Table 1, are reinforced by a complete error propagation analysis (hence the high associated uncertainty, since it is accumulated) so that the reader can judge on the merits of our reasoning. We would deem this sufficient, and maintain our discussion focused on the summer radon data – we further note that this approach, that we took, is in fact sadly lacking from the vast majority of published SGD studies involving radon mass-balances, particularly in large systems.

S#15: P12448 L26. Detailing the water balance in the lagoon would be helpful ie. The amount of water coming in and the amount of water going out. If the water balance is not equal over the particular tidal cycle where the Rn was measured, this can have significant impacts on the mass balance and should be accounted for.

R: This has been comprehensively done by Andre Pacheco and colleagues (see Pacheco et al, 2010, cited in P12440 L6 and P12449 L3, for example). The residual tidal prism is very small in spring tides (see Figure 4 and Table 4 of their paper). Both radon surveys were conducted in Spring-tide conditions (both tidal amplitudes are now specified in the new additions to Table 1, for comparison). The two mass balances are directly comparable. This information has been incorporated into the mass balances we did, as explained in P12449 L3-6. As a result, the net exchange of radon between the lagoon and the Atlantic (see Table 1) is negligible and doesn't affect the mass balance, as we discuss in P12449 L6-9.

S#16: P12450 L1. Throughout the Stable Isotope Hydrology section, it needs to be clear which samples were collected during the ^{222}Rn surveys and timeseries and if the collection times were different how applicable is it to compare signatures at the different times and how the signatures compare to the ^{222}Rn concentrations/export/import/mass balance.

R: This is a fair comment and has now been addressed, by inclusion of a summary of the relevant sampling information on the new Table 2, and a new section (under 3.2) on the inter-comparability of results. The new section on inter-comparability and the additional data provided might contribute to clarify the issue. This section now reads:

'Inter-annual comparability of isotopic data

Sampling campaigns were carried out strategically following a field-adaptive protocol. Of primary concern was to capture the extent of temporal end-member variability in isotopic signature under maximum freshwater flow (hi-flow) conditions, in order to a) guarantee coherence of source compositions to feed into mixing models when necessary while assessing the hydrology of the lagoon over wider temporal scales and b) minimizing logistics and costs while guaranteeing inter-comparability. For this purpose, winter season was chosen given that ~61% of the mean annual precipitation falls on the region between November and February (34% in the months of December and January). Stable isotope sampling in winter had the added advantage of minimizing kinetic effects over stable isotope signatures given the lower evaporation potential. Sampling in winter 2007 was exploratory, with two main objectives: firstly, to characterize isotopic signature of M12 groundwater and surface lagoon waters in the western sector, particularly in the area that could be potentially influenced by both SGD and the WWTP outflow under maximum dilution potential (hence high tide), and secondly, conduct an exploratory survey of potential seepage areas along the Ancão peninsula, keeping in mind that the location of at least one of the important SGD seepage sites was known (Leote et al, 2008). Detection of the isotopic signature of groundwater in porewaters at the seepage face at stations Pw_e and Pw_f (Table S1) led to the installation at their location of a nested piezometer transect array in January 2010. This was subsequently used to obtain porewater samples in the 2010/11 winter season (December 2010 and January 2011).

To capture inter-annual variability, the M12 aquifer was sampled twice (winters of 2007 and 2009), with the provision of one common location (Ramalhete) for cross-referencing. Following the same reasoning, the M10 aquifer was sampled in December 2010 while simultaneously sampling Rio Seco (belonging to M12, Table S1). This ensured inter-comparability between groundwater isotopic signatures in 2009 and 2010. Campaigns were planned in advance considering the precipitation over the region to ensure similarity in the hydrological regime and ultimately guaranteeing inter-comparability of results. The sampling itself took place in dry conditions as much as possible, and never after intensive rain that could have promoted flooding (Table 2, Fig 4d). For example, while January 2007 was a dry month (8.8 mm) compared to the historical average (138 mm), the accumulated precipitation during the previous 3 months was 369.7 mm, consistent with the historical average (Table 2). By contrast, both December 2009 and 2010 were relatively wet months (392.2 and 269.6 mm), but followed relatively dry 3-month periods (Table 2). So porewater samples were also taken in January 2011, hence complementing winter 2010/2011. January 2011 followed a wet three-month period (414.7 mm) and was hence comparable with January 2007, also relatively dry but on the back of three wet months (369.7 mm cumulative). The combined dataset therefore contains results from repeated measurements for end-member isotopic composition under hi-flow conditions, across different years. These are in addition compared to historical data (table S1, Figure 4), leading to a temporally coherent quantitative overview of stable isotopic hydrology over the catchment.'

S#17: P124550 L14 Please define the acronyms used in figure 4 and 5 in the caption ie.WMMWL

R: Done.

S#18: P12455 L5. Change “discriminate between potential source functions of SGD.” To “discriminate between potential sources of SGD.”

R: Done.

S#19: P12455 L9. Change “potential source functions” to “potential sources”

R: Done.

S#20: P12455 L9. As per the general comments, clarification on the recirculation endmember needs to be addressed. Does the beach groundwater endmember represent fully equilibrated re-circulated seawater or new seawater with a very low residence time that has yet to fully equilibrate. The authors assume it is fully equilibrated, but it needs to be discussed why they believe this is so.

R: See also in the introductory paragraphs. If we assume that the water (from the lagoon, only partly open seawater) infiltrates the unsaturated zone of the beach during flood and is flushed through the permeable zone of the beach driven by tidal pumping, then as part of this endmember, we get radon produced from the sediments during the residence time of the water in the beach plus the radon which was already in the water before recycling through the beach sediments. This recirculation pump is constant, i.e. the residence time of the water in the beach is constant over time, so the added fraction of radon from sediments may be seen as constant. Therefore, we don't require or assume that it is fully equilibrated.

Under the assumptions above, and those pertinent to the calculation of the contribution of re-circulated water to the SGD in the lagoon (lifetime of radon), we always get the same amount of radon from this production in the sediments as long as the recirculation time scales are seen to be constant. So we have an endmember which is probably partly equilibrated (because the residence time within the beach is probably too short for full equilibration to occur) but which is constant; and this endmember was measured, several times (Table 1)

S#21: P12455 L15. Add in “The corresponding volumetric discharges, if each of these potential sources is considered in turn to be the only source of SGD to the lagoon are..”

R: Done.

S#22: P12455 L25. Again this highlights the temporally dynamic nature of the lagoon and comparison of parameters across different campaigns must be discussed.

R: This has now been done. See response to comment S#16, detailing the new section on inter-comparability of campaigns, and additions to Table 1 as well as the new Table 2.

S#23: P12456 L2. Please describe this mechanism (with references) in more detail as I find this explanation highly unlikely. As ^{222}Rn is essentially sourced from sediments and porewater ^{222}Rn is regularly many magnitudes higher in porewater than surface water, surface water contributing ^{222}Rn to the porewater is not feasible. At a guess I would say that wind and current evasion is likely underestimated. Providing more detailed explanation of the terms used in evasion calculations and uncertainties around those estimates may help determine if this is the case.

R: This comment is not entirely clear to us. We assume that the reviewer is requesting further explanation of beach hydrology and beach groundwater dynamics, as this explains the loss of radon from the system found at high tide (explained in P12456, L1-19).

Since the late 1940's, there has been a multitude of studies focusing on hydraulic behavior of beaches, including modeling studies describing flow dynamics above and below the beach water table. Given the abundance of materials, these cannot be cited entirely here and this issue is not the main focus of the paper – it is well-established knowledge, at least on the issue mentioned by the reviewer. However, a comprehensive review of beach groundwater dynamics is given by Diane Horn (Horn, D.P., 2002. Beach groundwater dynamics. *Geomorphology* 48: 121-146), and more recent work with focus on SGD can be consulted for example in Heiss et al (2014): Heiss, Ullman, Michael: Swash zone moisture dynamics and unsaturated infiltration in two sandy beach aquifers, *Estuarine Coastal and Shelf Science* 143: 20-31, or Ibanhez & Rocha, 2014 and 2015 (below).

Briefly, the water table on beaches divides them into two main areas, depending on prevalent pore pressure: the area below the water table, which is permanently saturated and where pore pressures are positive, and the area above the water table (sub-atmospheric), where sediment is unsaturated and pore pressure is negative. The size of these areas across a vertical beach section changes in cyclic fashion according to tide level – during flood, water infiltrates into the unsaturated portion of the beach (recharge), usually at the upper levels of the beach slope, and during ebb, this water flows seaward mainly through the pores driven by the hydraulic gradient established between the beach water table and the sea level at any one point in time during discharge (discharge). Infiltration of water into the unsaturated zone of the beach during flood tide is an advective process, i.e., water physically infiltrates the unsaturated portion of the beach, taking up empty pore space left behind by the previous discharge cycle. This water will eventually discharge through the lower portion of the beach at low tide. This cycle is referred as tidal pumping and is the reason for seawater recirculation through beaches. Hence, when water infiltrates the unsaturated portion of the beach, it injects into the beach any accompanying solutes, and even some particulates that are critical in determining the biogeochemical role of tidal pumping (see for instance Ibanhez and Rocha, 2014 and 2016). This process also affects stable isotope signatures of porewater, as explained for example in P12458, L2-9).

Hence during flood, lagoon water with dissolved radon infiltrates the unsaturated zone of the beaches within the lagoon, removing this radon from the surface lagoon pool, as we explain, with calculations based on our data, in P12456, L1-19. This is not a mixing process between surface lagoon water and saturated beach porewater as the reviewer seems to have understood – it is a removal of surface water and its radon from the surface water pool and into the area below the sediment surface. Mixing occurs within the beach pore space after recharge and the water discharged at the lower portion of the beach slope during low tide incorporates both water infiltrated during the previous high tide and water that had remained in the beach for a longer period of time. We therefore are at loss as to how to make the mechanics of this process more clear.

Ibanhez JSP & Rocha C (2014). Effects of recirculation of seawater enriched in inorganic nitrogen on dissolved organic carbon processing in sandy seepage face sediments. *Marine Chemistry* 166: 48-58

Ibanhez JSP & Rocha C (2016). Oxygen transport and reactivity within a sandy seepage face in a mesotidal lagoon (Ria Formosa, Southwestern Iberia). *Limnology and Oceanography*, 61: 61-77

S#24: P12456 L22 Would “is” be a better choice of words here than “could”?

R: We are not sure – but we wished to keep with the narrative type discussion, where we progress from the unknown into finally taking a supported conclusion as to how conflating the two SGD modes because of the lack of distinction of sources may affect the N balance estimates for the system. So we would maintain the ‘could’.

S#25: P12456 L20 Again the “two different periods” are not clearly defined previously in the manuscript.

R: We are not sure about this comment. The page and line mentioned do not contain the sentence the reviewer alluded to. However, we hope that following the changes to the manuscript and highlighted in our responses to the other comments, the issue of comparability is resolved.

Appendixes: redrafted figures and tables.

Figure 1. Map showing location of the sampling sites within the Ria Formosa and its geographical context. The top panel shows the full geographical extent of the system, with the operational separation of the region of interest into western and eastern lagoon and the names of all the inlets; The lower panel shows an amplified map of the region of interest, including major channels, locations of sampling and tidal stations,

as well as boundaries of the aquifers bordering the lagoon (M10, M11, M12).

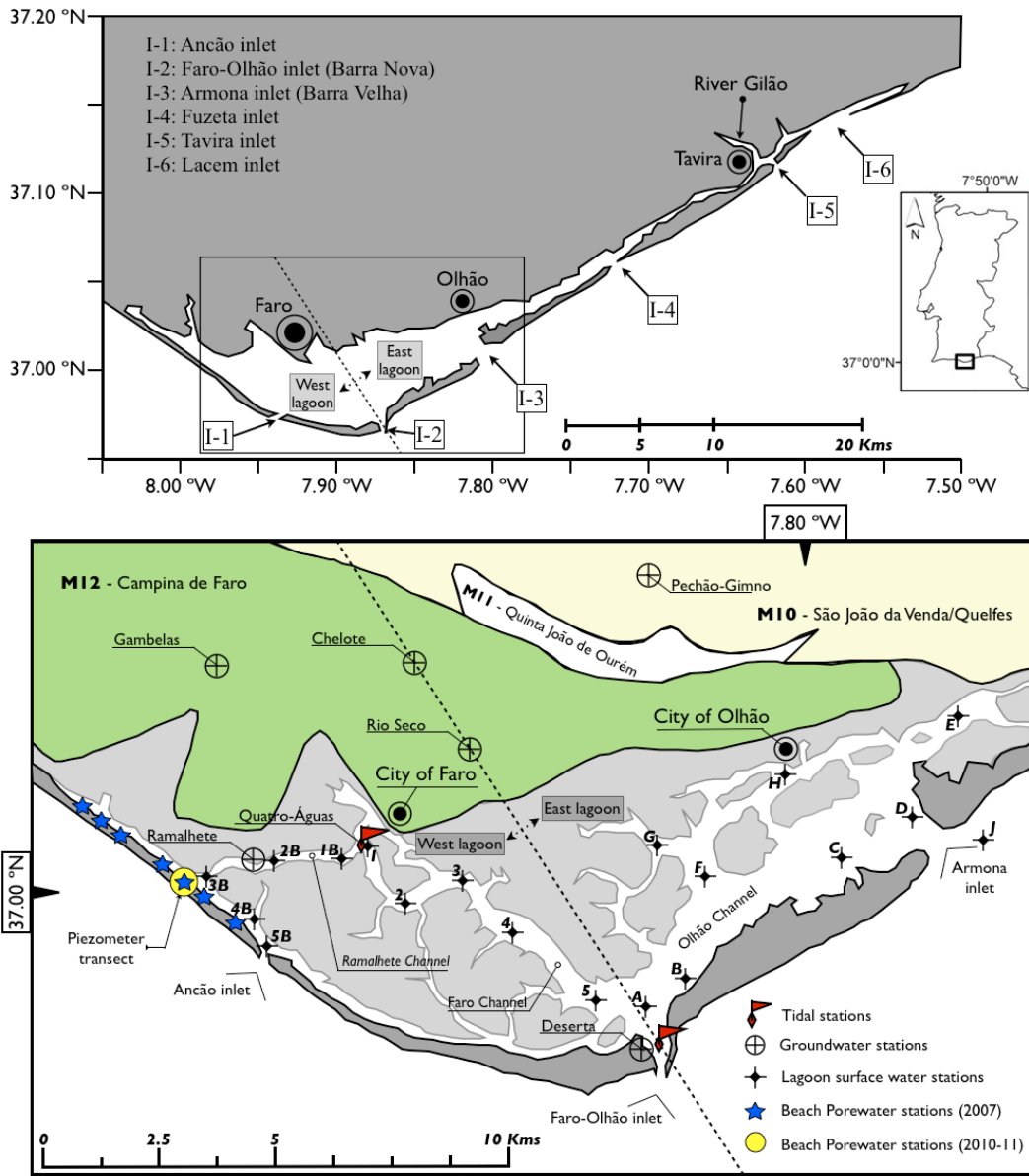


Figure 4. Catchment isotope hydrology. Anticlockwise, from top left: panel a shows the main meteoric water lines framing the isotopic composition of precipitation within the catchment, including the precipitation-seawater mixing line (PP-SW Mix, section 4.2.1.). Panel b plots the isotopic compositional range of water samples taken during 2007, while Panel c plots the isotopic compositional range of water samples taken during the period 2009–2011; the lagoon surface water samples (inset) are shown in more detail on Fig. 6. Panel d provides the complete record of daily precipitation over the region for the period 2006–2013 for contextual support (see also Table 2 for summarized data). EMMWL: Eastern Mediterranean Meteoric Water Line (Gat and Carmi 1970); WMMWL: Western Mediterranean Meteoric Water Line (Celle-Jeanton et al 2001); GMWL: Global Meteoric Water Line (Clark and Fritz, 1997); LMWL: Local Meteoric Water Line (Carreira et al 2005)

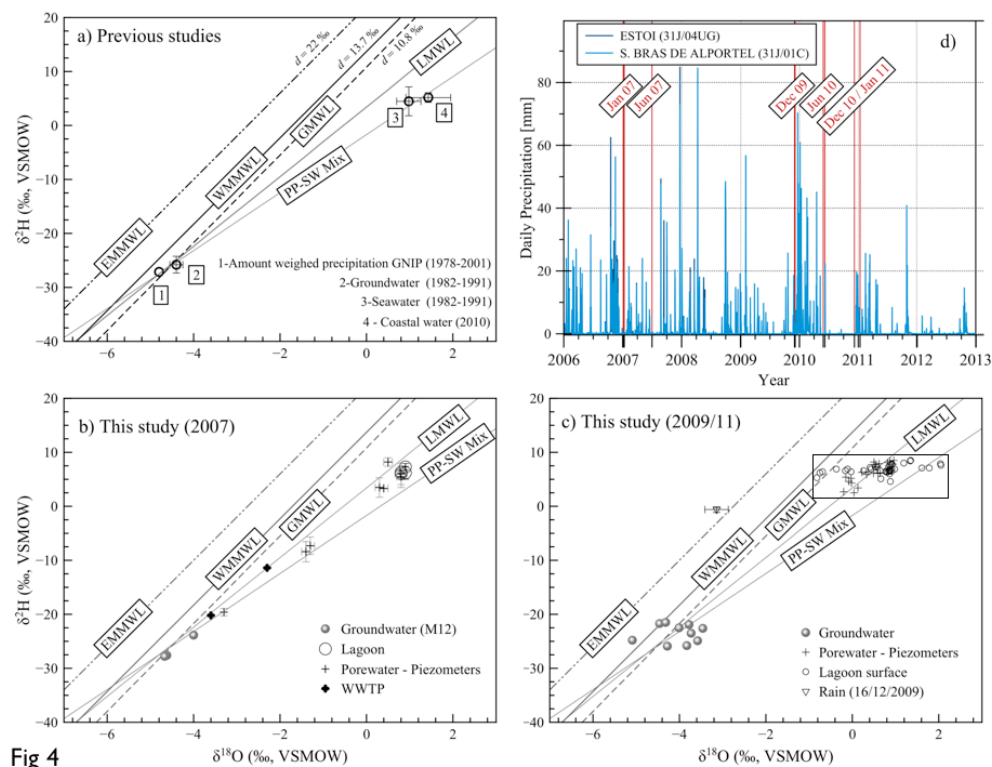


Fig 4

Table 1. Excess ^{222}Rn inventories and relevant fluxes supporting the radon mass balance for the Ria Formosa in winter 2009 and summer 2010 (see Sections 4.1 and 5.1). Notes: ^aCalculated with formulas 4a and 4b, Section 3.1.4.2; ^bCalculated with Formula 3, Section 3.1.4.1; *Referenced to lagoon surface area at MSL, calculated using the residual exchange measured at Faro-Olhão adjusted to the residual tidal prisms for all the inlets reported in Pacheco et al. (2010) and cross-section area for all the inlets. Minus sign signifies net export (seaward). **Per unit cross-sectional channel area

	Winter 2009	Summer 2010
Tidal Amplitude [m]	2.73	2.51
Wind speed [ms^{-1}]	8.4 \pm 8.0	6.3 \pm 1.2
Inventories	^{222}Rn inventory \pm MAD [Bq m^{-2}]	
Ebb stage ^a	55.6 \pm 30.9	54.2 \pm 17.8
Flood stage ^a	73.8 \pm 31.5	74.0 \pm 17.6
All data ^b	66.1 \pm 34.7	65.9 \pm 19.6
Fluxes	^{222}Rn flux \pm σ [$\text{Bq m}^{-2} \text{day}^{-1}$]	
Diffusion	5.7 \pm 1.9	5.9 \pm 1.7
Degassing	1.7 \pm 1.8	1.1 \pm 0.7
Decay	12 \pm 6.3	11.9 \pm 1.6
Residual Exchange*	-5.26(\pm 1.03) $\times 10^{-4}$	-4.74(\pm 0.79) $\times 10^{-4}$
Tidal Flux**	^{222}Rn flux \pm σ [$\text{Bq m}^{-2} \text{day}^{-1}$]	
<i>Quatro-Águas</i>		
Export	-	85.4 \pm 11.1
Import	-	98.6 \pm 16.1
Residual	-	13.2 \pm 2.8
<i>Barra-Nova</i>		
Export	57.0 \pm 6.4	49.8 \pm 1.1
Import	65.5 \pm 4.2	65.0 \pm 4.2
Residual	8.5 \pm 1.1	15.2 \pm 1.0
Potential Rn sources	Salinity	Activity \pm σ [Bq m^{-3}]
Deserta (Well)	0.95	93.8 \pm 59.5
Beach porewater	40.6	304 \pm 182
Ramalhete (borehole)	5.06	6625 \pm 996

Table 2. Precipitation records over the region during the sampling campaigns described by this study, as measured at the São Brás de Alportel meteorological station (www.snirh.pt, Ref 31J/C). Monthly precipitation is contrasted with rainfall during the sampling campaigns and compared with historical monthly averages in order to evaluate the relative wetness of the periods in the wider temporal context. Accumulated precipitation during the 3 months prior to the month fieldwork took place is also shown and similarly compared to the historical record average. For a more detailed contextual assessment, the chronological record of daily precipitation for the period 2006-2013 is shown in Fig 4, panel d, with the sampling periods overlain for easy reference when evaluating the stable isotope hydrology of the catchment defined by this study and previous research. Under ‘Sampling’, and ‘Type’, the type of endmember collected for stable isotope analysis is shown, except when radon survey campaigns were executed in parallel – in this case ‘Radon survey’ is added to the column. More details on the individual samples are shown in Table S1.

Date	Sampling	Precipitation [mm]					
		Survey		Month		Previous 3 months	
mm/yy	Period	Type	Total	Survey month	Historical average	Total	Historical average
Jan 07	3 rd -6 th	<u>Groundwater</u> • M12 aquifer • Beach porewater	0.1	8.8	138	369.7	369
July 07	1 st -3 rd	<u>Groundwater</u> • Beach drainage <u>Surface water</u> • WWTP • Lagoon West	0.0	0.5	3	83.7	125
Dec 09	1 st -8 th	<u>Groundwater</u> • M10 aquifer • M12 aquifer <u>Surface water</u> • Lagoon East • Lagoon West • Seawater <u>Other</u> • Precipitation	10.3	392.2	160	93.6	232
May/June 10	28 th -7 th	<i>Radon survey</i>	0.0	24.1	16	88.6	207
Dec 10	8 th -16 th	<u>Groundwater</u> • Beach porewater <u>Surface water</u> • River Gilão	0.5	269.6	160	147	232
Jan 11	3 rd -12 th	<u>Groundwater</u> • Beach porewater	18.7	48.5	138	414.7	369

