

Response to comment #C6634 (reviewer 3)

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 other referees (we note that there are points in common), this has benefitted us greatly. The reviewer makes three general comments, and a number of other suggestions. 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. The response will address these individually.

General Comments:

G1. "This is an important paper. As the authors recognize, problem of selecting end-member concentrations has plagued SGD studies since their inception both in transforming Rn (and Ra) data into water fluxes and in calculating contaminant deliver via SGD. The stable isotope strategy presented here is an innovative approach. I would emphasize the method rather than "the overarching aims of the study to identify the sources of SGD..." (p. 12438 1, line 28). As a demonstration of the method, one might have hoped for a simpler study site, but perhaps, the complications at the Ria Formosa Lagoon serve to demonstrate the utility of the approach."

R: It is very generous of the reviewer to give us this mark of approval on our work. Sincere thanks. Having said that, the suggestion appended is indeed an important and valued one. Our perspective is that if the approach works in such a complex system, and actually highlights aspects of the system that took decades of research to uncover by traditional hydrodynamic and modeling analysis, then it certainly would be of value to the community. Considering also comments along the same vein by the other reviewers, we attempted to rewrite the last two paragraphs of the introduction to hopefully highlight the study and its consequential findings a bit more.

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.'

G2. "Given the sophisticated geochemistry, I found the treatment of the tidal hydraulic overly simplistic. Although I'm willing to concede that the residual tidal exchange is unimportant in the Rn budget, I had little confidence in the results. The tides deserve a more complete treatment in the description of the study site. The tide may indeed be a "traveling wave" (p. 12445 line 7) but I would not be surprised that, in such a tortuous lagoon, it is not; and why use 12 hours as the semi-diurnal period when it's no more calculation effort to use the actual semi-diurnal period (p. 12445 l 21). The issue of the exchange of water among the three inlets (p 12461 l 6-15) is important and should be described earlier in the description of the study area."

R: Andre Pacheco and colleagues (see Pacheco et al, 2010, cited in P12440 L6 and P12449 L3, for example) conducted a comprehensive analysis of inlet hydrodynamics in the Ria Formosa in the context of multi-inlet system morphological stability. The residual tidal prism is very small in spring tides (see Figure 4 and Table 4 of their paper). There is an internal residual flow eastward within the lagoon, which they showcase based on careful analysis of system hydrodynamics, and we independently uncover with a basis on the stable isotope hydrology data (in section 5.2, specifically P12460 L29, P12461 L1-15), the radon distribution between low and high tide (P12461 L16-28), and confirm its coherence with insights provided by their work in P12461 L16-25.

With regards to the exchange fluxes of radon, 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 therefore 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. Given that the Rn budget is controlled essentially by internal lagoon processes, and not by the exchange of its water with the Atlantic, we then treat the entire system as our domain neglecting spatial variability for the purpose of budgeting.

Using 12 hours as the semi-tidal period is a mathematical simplification, for ease of use by others (formula 4c becomes easy to use and recall, under the assumptions of negligible exchange with the outer domain over the lifetime of radon). Either way, it doesn't affect the result at all, since the exchange flux is orders of magnitude lower than the other fluxes as depicted in Table 1 and discussed in section 4.1.2. We also show the lack of distinction between the two SGD estimates, obtained respectively by treating the whole domain as one and using all the data, and using the travelling wave approach and tidal stage inventories in separate in section 4.1.3.

G3. “The “freshwater lens” (p. 12455 l 10) is not described. Is it possible that differences in tidal phase across the outer barrier is driving water between the ocean and lagoon under the barrier? (I believe this has been shown to happen in some sites in Florida and Venice). And what about the extensive marshlands in the lagoon (grey areas in Figure 1)? How does drainage on, off and through the marshland figure into the budget?”

R: This is part of the M12 aquifer (Section 4.2.2, P12451 L19-24). The difference in tidal phase driving water between the ocean and the lagoon is always a possibility. Indeed, there is a seminal paper on this very issue (Bokuniewicz & Pavlik, 1990: Groundwater seepage along a barrier island, Biogeochemistry 10: 257-276) that illustrates the debate perfectly. The question is whether this is saltwater (i.e., re-circulated lagoon water) or freshwater. If it is freshwater (it certainly isn't the case for 2009-2010 as we show), we had previously (see Rocha et al 2009) conducted an analysis of potential hydraulic gradient created by this tidal difference and compared that with that created by the piezometric levels on the continental side of the lagoon, within the same aquifer. We found the hydraulic gradient from land to be the more important force driving the flow. However, it is almost certain from a rational perspective that this oscillation of tidal phase contributes to porewater (beach groundwater) mixing within the beach aquifer. Which brings us to the second point: If it is re-circulated lagoon water that overlays the freshwater beneath, and therefore potentially the dominant fraction of SGD (in the absence of a terrestrial hydraulic gradient driving fresh groundwater flow from the M12 aquifer through the seepage face), this tidal phase gradient-driven mixing and driving force would only contribute to the SGD mode we describe.

With regards to the marshland contribution to the radon budget: at the request of reviewer 1, we added some detail on the way samples were obtained for the measurement of diffusive fluxes of radon (P12444 L7 and subsequent lines) to employ in the budgeting analysis. The section now reads: ‘Sediment-water diffusive fluxes of radon were measured as described in Corbett et al. (1998) in samples ($n=16$) collected throughout the lagoon and directly analysed in the laboratory upon collection. To obtain these samples, undisturbed sediment cores (35 cm length) were collected using polycarbonate core liners (\varnothing 5.5 cm) in both sub-tidal ($n=8$) and intertidal environments ($n=8$), with each environment sub-sampled for sandy and muddy sediments in equal proportions. Resulting fluxes from all analyzed cores were then averaged and the latter value, with its associated uncertainty, used in subsequent mass balance calculations.’ Most of the intertidal areas mentioned are covered in a muddy-silty layer as typical of intertidal marshland, and these were sampled. Hence part (necessarily, as we cannot assume under this budget framework that we are covering all bases completely and have to accept some uncertainty) of the contribution of R_n to the budget is already accounted for, since the diffusive fluxes comprise an average of those measured in sandy and muddy-silty areas, with the associated uncertainty of the mean is also propagated into the calculation. The area employed to upscale this data to basin scale is the Mean Tidal Area, which would cover most of the marshland sections of the map. Since for impermeable sediments we accept that no significant convective or advective water inputs occur (given the poor hydraulic conductivity of the muddy coverage), we accept that most of this contribution will be accounted for in our budget.

Responses

Minor suggestions:

1. I found confusing the use of two names interchangeable for each inlet (p. 12440 l2) especially when only one name is given in Figure 1. Is this necessary?

R: This is a fair comment, and has been put forward by the other reviewers as well. We addressed the issue in the following manner:

a) Figure 1 was redrafted, and now incorporates two panels, with one covering the entire extent of the system, where all the inlets are depicted.

b) There is now a list of all the inlet denominations, with their anglicized and their Portuguese names associated with the new panel in Figure 1.

With regards to inter-changeability of names, we assume this is more of an effort to first make reading and memorization for non-Latin speakers that might find the accentuation of the Portuguese words difficult to accommodate (hence Barra-Nova rather than Faro-Olhão, for example). This might be entirely pedantic, but would also bring to the fore the local denominations, rather than those publicized in the international literature.

2. Page 12448 line 8. Because the ocean waters are Rn-poor why is the mean Rn activity higher on the flood? Here's where a more careful explanation of the tidal hydraulics might have helped.

R: Firstly, we do not treat the system as completely isolated from the adjoining coastal water mass (i.e., by employing the tidal prism method, where all water leaving the system never returns, etc). See Section 5.2, specifically P12460 L3-7, where we quite clearly state that the adjacent coastal water is affected by the mixture of lagoon water, affected by terrestrial sources and surface evaporation, and offshore seawater. This is clear from the isotope data presented in figure 4a.

Secondly, we provide further discussion on P12460-12461 in this regard. In lines 20-25 (P 12460), we infer a hydraulic connection between the Ramalhete Channel, the Ancão inlet and water masses in the eastern sector of the lagoon, as ebb progresses into flood, from the isotopic data (Fig 6a and 6b). This implies, as we then state in L29 (P12460) and then L1-6 (P12461) part of the water flowing out at ebb through the Ramalhete Channel and into the Ancão basin (where stations 3B, 4B and 5B are) is kept in the system, and hence its radon is added to that produced by beach discharge during ebb within the lagoon – this can be through reinsertion of the water during flood through the Barra Nova inlet toward the east, after this water mass leaves through the Ancão inlet (given the eastward along shore currents), or via a loop within the western sector (Ancão basin into the Faro Channel again and from there eastward). Because the Barra Nova inlet is flood dominated, while the others are ebb dominated, the circulation through the lagoon progresses with a westward loop (Barra Nova – Ancão inlet) and a similar loop eastward (Barra Nova – Armona Inlet) taking place and connecting hydraulic flow through these three main inlets and residual flow moving slowly eastward, which is consistent with hydrodynamic studies (Pacheco et al 2010), as we refer. See also first paragraph of the response to general comment 2 (G2, above).

3. Figure 3. It might be more instructive to plot Rn-flux versus the water depth rather than to plot both against time. The figure suggests a more complicated tidal modulation than the simplified flood-ebb analysis used earlier.

R: Indeed, but it would not add in our view any more of substance to the paper. It does suggest more complicated tidal modulation over the whole system, given that this is data from the main inlet, which is flood dominated – this is an aspect that we discuss further in P12460-61, and in the response to comment 2 above.

4. P 12451 | 25. “LEL” is not on these figures (until you get to Figure 6).

R: Here we just mention that the groundwater samples from both aquifer M10 and 12 plot along Local Evaporation Lines (defined in L25) with slopes as indicated. This

intends to show that there is interaction with the atmosphere (hence evaporation) even in the groundwater flow paths. The LELs depicted in Figure 6 are surface water evaporation lines.

5. P. 12455 lines 3 and 7. Are “end-member source” and “source functions” synonyms or the authors mean some (subtle) difference between the two phrases.

R: No, there are subtle differences and we would seek to maintain these as much as possible. The subtlety arises from the perceived mode of variability of the source composition (or absence of variability) when reading its description – if it is described tout-court as a “source” one would more easily associate this, strictly speaking, to a continuous, constant composition; However, the source-function terminology carries the association with a variable end-member composition, either in time or space, that could be described by a mathematical function, for instance. The latter is more realistic.

6. Table 1 Tidal Flux. Does this make sense? More water cannot be moving in than is moving out. If the ocean water is Rn-poor how can the import of Rn be higher than the export?

R: yet, it does make sense – in a multi-inlet system. The Barra Nova (Faro-Olhão) inlet is flood dominated, while the other inlets are ebb dominated. See response to comment 2 above, and relevant discussion in P12460-61.