*Reviewer Comments are in blue
**Authors responses are in black *italics*

The authors feel that the comments made below have improved the manuscript substantially and would like to thank the reviewers for their efforts.

Reviewer #1

General comments:
This study explores the possibilities to estimate sediment-water fluxes of oxygen, nutrients and DIC from vertical water column profiles of 224Ra. In areas with a lack of information on sediment properties, this can indeed be an interesting alternative approach to direct (in situ) flux measurements, given the assumptions of the 1-D diffusion model are met. The paper is written very clearly, the model explanation is easy to follow, results including figures and tables are generally very explanatory. The discussion accurately addresses the questions scientific questions raised, although I think some remain to be answered. To my opinion, the paper can be published in BG after minor revision of the following specific comments.

Response: the authors appreciate these comments and will gladly make the revisions suggested.

Specific comments:
Comment:
Abstract
P9202 L2: add "Direct": Direct quantification of such benthic fluxes . . .
Response: The authors have made this change (line 12)

Introduction
P9203 L13-16: redundant, omit
Response:
The text has been omitted from the revised manuscript.

M&M
P9206 L9: add (a reference to) the assumptions of the 1D diffusion model?
Response:
Line 138: the revised manuscript adds a sentence to the end the paragraph:
"These assumptions follow those made by Moore (2000), and are described in more detail below.

P9209 L10: appropriate depths: add depths, this can now only be deduced from Fig 5.
Response:
Line 147: The following sentence has been added to the revised manuscript:
"Sampling depths varied for each daily profile due to varying weather conditions, with bottom sampling depths ranging from 66-70m, ~~chosen using the ships sounder~~, and a vertical spacing of 6-8 m between samples (see Fig. 5). "

Please clarify that the sampling period encompassed only 5-6 weeks (end of Oct to beginning of Dec), so the Ra profiles were taken more or less every week.

Response:

Line 142: This sentence now reads (added words are underlined):

"In total, five vertical profiles of 224Ra were sampled regularly over a six week period between October and December of 2010."

Furthermore, Line 258:

The phase "six-week" was added to the first sentence on the results section, to help clarify this point:

"The vertical 224Ra distributions during the six-week sampling period….."

P9210 L9-10: analysis method of oxygen nitrate, phosphate, chl-a and POC?

Response:

Line 174-177: The authors have added the appropriate sentences and references to the end of the methods section.

Results

To my opinion, this section is already too much integrated with result interpretations that belong to the discussion. E.g. P9210 L17-21 , P9211 L11-20, P9212 L1-14 . . .

*Response: The authors agree and appreciate this comment. The three sections listed appear to be ones requiring attention. The first section has been left in results section because the 2 sentences would not add to the discussion and would appear out of place. The longer 2 sections (*P9211 L11-20, P9212 L1-14), *have been incorporated into section 4.1 of the discussion.*

Discussion

The oxygen consumption here reported is quite high for such low temperatures. Is there any literature information available on direct flux measurements in the area or similar, nearby areas? The study from Hargrave and Taguchi (1978) is also indirect.

Response: To the best of our knowledge, no direct measurements have been made in the Basin. The high oxygen consumption given the low temperature has been discussed in the revised manuscript (Starting on line: 446).

When it comes to the interpretation of the pathways of OM mineralization, some basic information from literature on sediment composition would be at its place: what is the (approx.) sediment granulometry (cohesive mud, permeable sand? My guess is mud. . .), organic content, oxygen penetration depth, etc. Also, faunal composition of the sediment could help: are bioturbation and bio-irrigation taking place that could explain enhanced reoxidation of reduced substances? But in case abundant irrigating fauna is present, then the assumptions of the 1-D diffusion model are violated.

Response:
The authors appreciate this point, and have added sentences to the discussion section 4.3 (Lines: 422-424) outlining information on sediment characteristics. Additional information regarding bioirrigation, oxygen uptake, delayed responses, etc. have been discussed in an additional paragraph (Line 446-460).

In case all this abiotic and biotic information on sediment composition is not available for the study area, I would put more emphasis in the introduction on the fact that this paper is the first one to explore sediment – water fluxes in the area.

Response: We have included information on benthic biomass (Line: 458) and sediment composition (Line: 422 & 447). Also, in the introduction, the authors have put emphasis on the fact that the study represents the first direct benthic flux observations in the Basin (Line 80).

Why does oxygen consumption increase from end of Oct to beginning of December? Given the constant temperature during the sampling period, this is strange. Is there a delayed POC decomposition (e.g. Rudnick et al. 1986)? Is it increased fauna activity because of the POC arrival?

Response:
In response to this suggestion, the authors have added sentences to the end of section 4.3 (Line 455-460) suggesting a slight response lag of the benthic system, and have added the references (Rudnick and Oviatt, 1986) and (Ståhl et al., 2004).

Technical corrections:
Fig. 5b caption: dotted line: you mean "within dashed box"?
Response:
This refers to the dotted line along the x-axis, which is supposed to represent the sediment surface.
The caption has been modified so the caption reads:
"…distance from the sediment surface (x-axis, dotted line). Each…"

Fig. 6e caption: the DIC decrease . . . add s - decreases?

Response:
In response to these suggestions, the authors have chosen to remove this semtence from the revised manuscript as it tends to confuse to reader.
Indeed the flux is not negative, the gradient is negative, resulting in a positive flux. We thank you for pointing this out.

References

Burdige 2012 (in text) should be Burdige 2011 (in reference list)
Response:
The in text corrections have been made, thank you.

Men et al. 2011 not in text
Response:
The reference is present on line 143.

Reviewer #2:

General Comments: Overall, I like this article and think it merits publication. The data presented is a nice set of measurements, and the article addresses an interesting topic, as it evaluates remineralization rates from plankton debris, based on making a mass balance for water column nutrient budgets. This approach avoids some of the complications of direct flux measurements and core incubations, although it has some complications of its own, due to non-steady state behavior. The article is clearly written, well illustrated, and should draw significant interest. I have a few concerns about the details of the approach.

Major ones follow:

1. As the authors demonstrate, the system shows non-steady state behavior. The data presented follows a rapid flushing of the basin. With its short half-life, the 224Ra tracer will return to a steady state distribution in a couple of weeks, and the repeated profiles indicate that it does. However, the time for nutrient and O2 profiles to relax to steady state gradients is considerably longer. Relaxation time can be roughly estimated by the ratio of depth below sill (squared) to the eddy diffusivity (about 35^2/K = 50 days for K=2cm2/s). Thus, the increase in fluxes calculated represent relaxation toward what may be a steady state flux (although flux could be changing with time). Consequently, the calculation they present for carbon oxidation rate from the change in the water column composition, may be closer to the accurate flux than the rate they find as the average of their sampling times. They could easily solve the non-steady state problem by doing a numerical simulation (perhaps a multi-box diffusion model) to see if they can reproduce the time dependence of the observed profiles. In constructing this model, they might also evaluate whether the flux should largely come from the bottom, or if they need a water column source/sink term. I suspect that their interpretation that most of the flux comes from the bottom is correct, but this could be demonstrated more convincingly.

2. The authors have chosen a 1-D model because it is easily applied. However, they need to justify this (on page 9207 perhaps). They might do so through scaling arguments of horizontal eddy diffusivity (see Okubo, DSR, 1971) and basin dimension. Or they could assume that horizontal eddy diffusivity is fast and include a source term for 224Ra in the water column (and they will also have to introduce an area factor in the diffusivity term). Or, they could use the numerical model they should construct as noted above in comment 1, with real basin geometry for each layer. They might consult an interesting paper by Colman and Armstrong (L&O 32, p577, 1987) and references therein.

Response:

The authors thank the reviewer for these remarks. In response to these comments, the authors have created a time-dependant numerical simulation for the Basin using a finite-difference version of the model. This finite difference diffusion model covers the bottom

40m of the basin, and consists of 80 boxes (each 0.5 m thick), which for Ra, are influenced by diffusion from the neighboring boxes, and by decay. For DIC and O_2 the decay term is removed, but a water-column respiration term is added. The details of the model have been described in a separate section of the methods in the revised manuscript (Section 2.5). A section of the discussion has also been devoted to the model results (section 4.4). Also, a figure has been added to the revised manuscript, which illustrates the results of the model runs (Fig. 8).
The authors feel that the addition of these numerical simulations add substantially to the manuscript. The numerical model results are consistent with the assumptions of the 1-D diffusion model, including the assumption regarding steady-state and a dominant benthic source for Ra. The DIC and O_2 results are also consistent with the assumptions regarding a single benthic source/sink of DIC/O_2 and somewhat negligible water-column respiration.

Technical Comments: Other suggestions of lesser concern follow:

1. (p.9292, line26)
While sediment compaction, or macrofaunal irrigation MIGHT play some role in introducing the Ra isotopes to the overlying water, the short scale distance for this isotope (probably <1cm, depending on adsorption coefficient) means that molecular diffusion will very likely dominate input. Thus, pore fluid is not "discharged". In my opinion, I would not refer to this as "SGD" (p.9203, line 23), as seems to be implied (but not actually stated).

Response:
We do not intend to imply this flux falls into the category of SGD, so the authors appreciate this comment. Line 47: The word 'discharged' has been replaced by 'released', and 'submarine groundwater discharge (SGD)' was replaced with 'fluxes from sediment pore waters' (Line 64).

2. (p.9209, lines 1-3) Adding a bit more info about the fitting routine would be helpful. Was this done with a standard software package (Kaleidagraph, Matlab?), and was the equation first transformed into a log format (this can make a significant difference, and use of the general curve fit routine of Kaleidagraph is a great way to use the exponential form directly. Weighting by analytical uncertainty (as they did) is also likely to be an important factor.

Response:
The authors used a general fitting routine in MATLAB, and have added that statement to the manuscript (line 227).

3. In Table 1, there are a few more significant figures than needed. Also, I do not understand why the uncertainty for 3 Nov QRa is so large in comparison to those for K and Ao (also p. 9211, line 28). If the equation A = Ao*exp(-az) was fit to the data, the fractional uncertainty for K is twice that for the parameter a (so a has uncertainty of about 30%). The fractional uncertainty in flux (calculated as a*Ao) should then be about 40% when calculated by error propagation. Also, it might be nice to add a mean +- sig in this table (excluding the outlier).

Response:
The authors agree that too many sigfigs are present in Table 1, and have removed 1 sigfig, making the table less cluttered.
The authors appreciate the comment regarding uncertainty propagation, and have found the small mistake creating these particular uncertainties. The propagation of the Q_{Ra} uncertainty was corrected in the MATLAB code, and the correct uncertainties have been placed in Table 1.
Further analysis of the values is Table 1 resulted in the authors locating one error in the Q_{DIC} column, and two values in the Q_{O2} column. These small errors are mistakes in copying table results into the manuscript, and therefore the overall results were not altered.

4. In looking at the data in Table 1, I do not see a significant time trend for QRa or K. I would suggest just using an average value for K, as 224Ra should have relaxed to near steady state. It would also be nice to know what the buoyancy gradient is for this basin, over the depth range sampled. This would make the data more readily compared to other settings (Sarmiento et al, 1976 for example), rather than the rather wide range noted for off the UK.

Response:
The authors agree that no significant trend for QRa and K are present in the results, and have altered the results section accordingly (lines 260 and 265). Table 1 now also includes a 'Mean' row for K, A_0 and Q_{Ra}. This was also useful as some of these mean values were used in the time-dependant model.
*The authors have calculated the buoyancy frequency (N^2) for each sampling day in the basin, and has attached the figure to this response. The figure shows N^2 over the whole water column (panel a), and over the depth range sampled (panel b). This term is listed as 'buoyancy gradient' by Sarmiento et. al. (1976). The N^2 decreases considerably below 30m depth, and ranges between 1.7 - 4.2 * 10^{-6} s^{-2} within the depth ranges sampled. Using the constant of proportionality listed by Sarmiento et al. (1976), we calculated Kv's between 0.09-0.24 cm^2 s^{-1}, about 1 order of magnitude different than our results. These order of magnitude differences in results were also obtained by Sarmiento et al. (1976).*

The authors agree that more comparisons to the results are useful, and have added the results of the Sarmiento et al. (1976) paper to the discussion (~line 336-339).

5. (p.9210, line 3) I am not familiar with the VINDTA, but it sounds like this technique is for coulometric TCO2. How did you obtain Alkalinity? pH measurement? If so, presumably this was done before HgCl2 step, as HgCl2 affects alkalinity (and thus pH). Some clarification here would be nice.

Response:
The VINDTA technique measures DIC and A_T directly using coulometric and potentiometric detection, following protocol outlined in the Department of Energy (DOE) handbook. This has been clarified in the manuscript (line 167). The effect on the alkalinity from addition of HgCl2 is well below the precision of the VINDTA.

6. (p. 9212, lines 7-11) Not sure the comparison of Ra flux in atoms/m2 sec is quite equivalent to the Rn flux of Berelson et al. While the methodology is somewhat similar, as the authors note, the absolute values will depend on sediment characteristics (these were not described here, although a note about this would be useful)..

Response:
The authors agree with this statement, and appreciate this comment. These statements were removed from the manuscript, as both reviewers as well as the authors felt this comparison was not of particular use in the discussion.
As for descriptions of the sediment characteristics, these have been added to the manuscript in section 4.3 (lines 422-423, 446-460).

7. (p. 9213, line 19) "remineralization" would be a bit less confusing than "return flow".

Response:
The authors appreciate that 'return-flow' is a bit confusing. However, 'remineralization' through the interface seems confusing also. The authors have used the word 'transfer' in the revised manuscript (line 321).

8. A more precise way to get the O2:CO2:N:P stoichiometry would be to plot each of these parameters vs. O2, rather than comparing the slopes of the calculated flux vs. time. That will remove the systematic variations due to uncertainty in eddy diffusivity for each profile set. It might also reveal whether there are changes in the remineralization stoichiometry as the basin becomes less oxygenated, and denitrification in sediments becomes more likely.

Response:
The authors agree and thank the reviewer for this comment.
We have changed the methodology for calculating the stoichiometry, and, in turn, the N and P fluxes. Table 3 has been adjusted with the new ratios and fluxes. Also, the "O2 vs. other parameters" plot has been included in the revised manuscript (as Figure 7).
This method has lowered the uncertainties, while also yielding slightly different results, which the authors believe are more reasonable given the characteristics of the system.

Specifically, the new stoichiometry (-135:150:14:1) suggests denitrification is important, which makes sense given the sediment type (anoxic muds). These new interpretations have been added to the discussion, which also addresses the reviewers point #11 below.

The authors feel that these revisions have improved the results and discussion on the manuscript considerably, and thank the reviewer for this suggestion.

9. It is of interest that the flushing of the basin is due to input of warmer (but more saline) water apparently as an annual event. I gather that it gradually becomes colder and fresher as the winter passes. A few comments about this would be nice to include in the discussion.

Response:
The authors agree these events are of interest, and have added these statements to the end of section 4.2. (line 388)

10. (p. 9215, line 16-18) It is not apparent from Table 1 that the flux of 224Ra changed at all with time, due to input of any sediment that is richer in 228-Th, so I would delete this comment.

Response:
The authors agree with this comment, and have removed the statement from the manuscript.

11. (most of p. 9216). The nutrient stoichiometry will not reveal whether the reactions are taking place in the water column or in the sediments unless the denitrification is sufficiently large to make a big impact. This does not seem to be the case. This discussion needs to be modified a bit. It is also interesting to see the Alkalinity profiles. I do not know if the precision is good enough, but there appears to be a negative flux. In fact, oxic respiration converts particulate ammonium to nitric acid and should cause a slight decrease in alkalinity (about 0.15 moles per mole of TCO2). In most settings, carbonate dissolution masks this, but maybe not here. Other reactions in sediments (oxidation of metals, sulfide) may remove alkalinity, but this is balanced by the alkalinity produced in forming them, so only the nitrogen reactions or carbonate reactions should play a role. On page 9217, a comment is made about possible sulfidic character of the sediments. Is there any info about sediment characteristics here? Any profiles of Corg with accumulation rates that could be used to calculate a DIC flux for comparison?

Response:

The authors agree with the first statement, and have added sentences explaining the assumption of negligible water column respiration (lines 397-403).
Regarding the alkalinity gradients, the authors feel that although the Alkalinity precision is high (1-2 umol/kg), there is not substantial, or only weak, evidence for gradients in either direction, as is stated in the original manuscript. The authors have chosen not to

overemphasize these observations because they are near the uncertainty range of our method. Higher resolution sampling would be needed to properly reveal such patterns. The authors agree that more information about processes within sediments is needed, and this is stated in the original manuscript (line: 408). However, the authors have added information regarding the sediment characteristics (line: 422-423).
Beginning on line 446, the authors mention results from a sedimentation study (Hargrave and Taguchi, 1978), and compare the organic carbon deposition rate (17mmol C m^{-2} d^{-1}) to the DIC fluxes calculated in the basin.

12. (p. 9218, line 9) Some rewording here would make more sense. "modest allochthonous inputs" do not "sustain" the oxic conditions, although they play a role in the O2 balance. Ultimately, the oxic conditions are sustained by the replacement of basin water through episodic inflow and diffusive transport, at rates exceeding the oxygen demand from metabolizable POM through allochthonous and overlying water inputs.

Response:
The authors agree with this statement and have reworded the sentence in the revised manuscript (~line 523-526).

13. Their suggestions about temporal variations in Corg input seem like an excellent way to explain the mismatch between the annual trap flux previously measured and the 3x higher flux they will calculate with a non-steady state model. Or perhaps the trap measurements miss nepholoid transport during flushing.

Response:
The authors agree with this statement, and feel the sentence in the orginal manuscript (line 529) "an inflow of dense surface water, rich with labile POM and resuspended sediments, could import…." adequately describes this comment.