

## Response Reviewer 1:

We thank the reviewer for her/ his positive comments on the manuscript. We took the comments into consideration in the revised manuscript.

*Section 4.1: origin of radium enrichments Prior to this paper, the view was that water to station A3 south of the Polar Front was likely advected from the vicinity of Heard Island and the southern Kerguelen Plateau. The authors used the transit times of two drifters released at station A3 to argue that the circulation in this region is too sluggish to account for the presence of the activities of the short-lived  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  isotopes observed at the stations south of the polar front that are east of Kerguelen. It is interesting to note that a close study of the satellite chlorophyll in figure 2 shows that this recirculation area remains relatively low in chlorophyll through the growing season, suggesting that even if short-lived Radium isotopes are occasionally quickly transferred across the Polar Front, this cross-frontal transport seems to be a less important source of iron compared to the regions experiencing advective transport from Island sources (eg. north of the Polar Front, east of 74\_E from Kerguelen Island; south of the polar front, in the tongue coming up from Heard Island and the Plateau).*

**Section 4.1** It is indeed interesting to compare the Ra distribution in surface waters that highlights cross-frontal input of chemical elements and the chlorophyll distribution that is expected to respond to this input of chemical elements. We agree with the reviewer remark that the cross-frontal transport of chemical elements highlighted by the short-lived Ra isotopes may be a minor source, when considering the chlorophyll images: east of the Kerguelen Islands, indeed, there is an area with relatively low chlorophyll concentrations, which could suggest that the eastward input of chemical elements through the front has a minor impact on the development of phytoplankton. In contrast, large blooms are observed north of the Kerguelen Islands and above the southern Kerguelen Plateau that suggest a major impact of the direct advective transport from the islands (Kerguelen Islands and Heard Island). However, the cross-frontal transport of chemical elements may not only take place east of the Kerguelen Islands but also all along the Polar Front (that is, north and east of the “recirculation area”). This transport could contribute to fuel the large phytoplankton blooms observed in these areas and thus should not be neglected.

*p. 14026 line 8: Should be “One of the largest phytoplankton blooms: : :”*  
corrected p2

*p. 14030 line 7: “: : analyzed \*for\* ca. 120h”*  
for has been added p6

*p. 14033 line 8: delete “towards”: “gradually decrease offshore.”*  
Toward has been deleted p9

*p. 14033 line 22: “: : 200 kilometers offshore \*from\* the Kerguelen Islands”*  
Now “300 kilometers offshore from the Kerguelen Islands” It was a mistake from the reviewer, it’s written 300 km and not 200 km p10

*p. 14034 line 9: “offshore \*from\* the islands”*  
Now “several kilometers offshore from the islands” p10

*P. 14037 line 16: add “: : is the initial ratio \*in source waters\*”*  
Now “...the initial ration in source waters” p14

p. 14037 line 21: should be “see Sect 4.1”

Now “see section 4.1” p15

*Figure 10 caption: Please be specific about what the colorbar is (eg. “Colorbar indicates time elapsed since water body left the 2000 isobath”)*

We added “colorbar indicates time elapsed since water left the 2000 m isobaths” in the Figure 10 caption p22

### **Response to W. Geibert:**

The authors thank W. Geibert for his relevant and constructive remarks.

*Data are well described, only the names of the stations are confusing, but this is probably not the author’s choice.*

The stations have different names but are indeed the names chosen by the project and consequently cannot be changed. UW stations were specific to the Ra study and refer to the surface samples collected “under way” using the ship intake.

### *Specific comments*

#### *Major issues*

*\*It is not clear whether the authors consistently discuss  $^{224}\text{Ra}$  excess and  $^{223}\text{Ra}$  excess, or total  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$ . This is also true for the tables, in which both  $^{224}\text{Ra} - ^{224}\text{Ra}_{\text{ex}}$  and  $^{223}\text{Ra} - ^{223}\text{Ra}_{\text{ex}}$  are used, but not clearly labelled, and possibly  $^{224}\text{Ra}$  designates actually  $^{224}\text{Ra}_{\text{ex}}$  in some cases. Reading the caption for table 2, “The Radium excess, supported by thorium and actinium isotopes, is reported as Raex”, the situation is also not resolved, as Ra excess should be the part \*not\* supported by parent isotopes. Consequently, the authors should clarify in detail in the methods section how they calculate excess activities, clearly differentiate the two, and name and label them consistently throughout the text before publication in Biogeosciences.*

All the  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$  activities used in this study are excess  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$  activities. The  $^{224}\text{Ra}$  activities are corrected for the  $^{224}\text{Ra}$  supported by  $^{228}\text{Th}$  and the  $^{223}\text{Ra}$  activities are corrected for the  $^{223}\text{Ra}$  activities supported by  $^{227}\text{Ac}$ . We clarified this in the section 2.3.2 (Sample analysis section) by adding “The  $^{224}\text{Ra}$  activities are corrected for the  $^{224}\text{Ra}$  supported by  $^{228}\text{Th}$  and the  $^{223}\text{Ra}$  activities are corrected for the  $^{223}\text{Ra}$  supported by  $^{227}\text{Ac}$ . The  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$  activities discussed here thus refer to these excess  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$  activities.” p6 We also replace “ $^{223}\text{Ra}_{\text{ex}}/^{228}\text{Ra}$ ” by “ $^{223}\text{Ra}/^{228}\text{Ra}$ ” and “ $^{224}\text{Ra}_{\text{ex}}/^{228}\text{Ra}$ ” by “ $^{224}\text{Ra}/^{228}\text{Ra}$ ” in table 1 p29 to be uniform as all  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  activities in the manuscript are excess activities

*\*The authors conclude that short-lived Ra isotopes cross the Polar Front from Kerguelen, as they could not migrate northwards from Heard Island on the relevant timescales. There is, however, a third possible source, a plateau west of the Kerguelen Plateau, which could at least in theory supply short-lived Ra isotopes at the timescales in question (Fig. 1). Given the strong eastward current in the Polar front, transport rates of 20 km/day seem easily perceivable. The authors must at least discuss all potential sources and explain why this large topographic feature is no possible source for Ra,*

*if they think so. Please also note how very different water masses and chl-a patterns are in the polar frontal region. Please note that assuming a different source region for excess Ra in some locations could also help to explain a mismatch between model age results and Ra ages in some cases.*

The Leclaire Rise is a topographic feature located west of the Kerguelen Islands (see Figure below; van Beek et al., 2008). Because it is located at ca 350 m depth (Weis and Frey, 2002), an impact of this topographic feature on the surface waters implies i) release of chemical elements from the sediments and ii) vertical mixing that would transport the chemical elements towards surface waters. Internal tides may promote such vertical transport of chemical elements in this area. In addition, because the Leclaire Rise is located north of the Polar Front (Park et al., 2014), the presence of Ra isotopes in the investigated area (east of the Polar Front) also suggests a transport across the front. If vertical mixing transports Ra in surface waters above the Leclaire Rise, it cannot be completely excluded that the Ra enrichments observed south of the Kerguelen Islands along the Polar Front (e.g. UW 23; UW 24; Figure 4) or south of the Polar Front (e.g. UW 15, UW 16 and potentially A3; Figure 4) are partly explained by an input associated with this topographic feature. However, sample UW 14 collected in surface waters lying above this rise (bottom depth at 342 m) does not show significant  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  activities and only low  $^{228}\text{Ra}$  activity, which suggests slight impact of this topographic feature on the Ra activities of surface waters. Station R2, which is located east of the Leclaire Rise - but south of the Polar Front - shows slight enrichments in  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  in surface waters (0.016 and 0.057 dpm/ 100 L, respectively), which could suggest an impact - but relatively minor when considering the Ra activities - in surface waters downstream of this plateau. Fe and other trace metal concentrations (REE, Mn, Al) do not show significant trends in surface waters at R2, which indicates that the Leclaire Rise has a minor impact on surface waters downstream of this plateau (Bowie et al., 2014; van der Merwe et al., 2014; Grenier et al., in prep.). In contrast, these latter studies suggest that the shallow sediments of the Leclaire rise may impact waters in the 200-500 m depth interval downstream of the rise.

This discussion has been reported in the manuscript p13-14 as “South of the Kerguelen Islands (i.e. along the Polar Front at stations UW-23 and UW-24 or south of the Polar Front e.g. at stations UW-15, UW-16, R-2; Figure 4), it cannot be completely excluded that the observed radium enrichments are partly explained by an input of radium associated with the Leclaire Rise located west of the Kerguelen Islands at ca 350 m depth (Weis and Frey, 2002). Station R-2, which is located east of the Leclaire Rise south of the Polar Front, shows significant  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  activities in surface waters. Although these activities are relatively low (0.016 and 0.057 dpm/ 100 L, respectively), they suggest that the waters downstream of the Leclaire Rise may be impacted by this topographic feature. However, sample UW-14 collected in surface waters lying above this rise does not show significant  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  activities and only low  $^{228}\text{Ra}$  activity, which suggest that vertical mixing may not efficiently transport radium released by the shallow sediments towards surface waters above this topographic feature. Note that the influence of the Leclaire Rise on the chemical element concentrations downstream of the rise is also observed in Fe and other trace metal (REE, Mn, Al) concentrations, but only in waters lying in the 200-500 m depth interval (Bowie et al., 2014; van der Merwe et al., 2014; Grenier et al., in prep.). »

Van Beek, P., Bourquin, M., Reyss, J.-L., Souhaut, M., Charette, M. A. and Jeandel, C.: Radium isotopes to investigate the water mass pathways on the Kerguelen Plateau (Southern Ocean), *Deep Sea Res. Part II Top. Stud. Oceanogr.*, 55(5-7), 622–637, doi:10.1016/j.dsr2.2007.12.025, 2008.

Bowie, A. R., van der Merwe, P., Qu  rou  , F., Trull, T., Fourquez, M., Planchon, F., Sarthou, G., Chever, F., Townsend, A. T., Obernosterer, I., Sall  e, J.-B. and Blain, S.: Iron budgets for three distinct biogeochemical sites around the Kerguelen archipelago (Southern Ocean) during the natural fertilisation experiment KEOPS-2, *Biogeosciences Discuss*, 11(12), 17861–17923, doi:10.5194/bgd-11-17861-2014, 2014.

Cai, P., Shi, X., Moore, W. S. and Dai, M.: Measurement of  $^{224}\text{Ra}$ : $^{228}\text{Th}$  disequilibrium in coastal sediments using a delayed coincidence counter, *Mar. Chem.*, 138–139, 1–6, doi:10.1016/j.marchem.2012.05.004, 2012.

Grenier, M., Garcia-Solsona, E., Lemaitre, N., Bouvier, V., Nonnotte, P., Jeandel, C. : Differentiating lithogenic supplies, water mass transport and biological processes on and off the Kerguelen Plateau using the rare earth data, in prep.

Van der Merwe, P., Bowie, A. R., Qu  rou  , F., Armand, L., Blain, S., Chever, F., Davies, D., Dehairs, F., Planchon, F., Sarthou, G., Townsend, A. T. and Trull, T.: Sourcing the iron in the naturally-fertilised bloom around the Kerguelen Plateau: particulate trace metal dynamics, *Biogeosciences Discuss*, 11(9), 13389–13432, doi:10.5194/bgd-11-13389-2014, 2014.

Park, Y.-H., Durand, I., Kestenare, E., Rougier, G., Zhou, M., d' Ovidio, F., Cott  , C. and Lee, J.-H.: Polar Front around the Kerguelen Islands: An up-to-date determination and associated circulation of surface/subsurface waters, *J. Geophys. Res. Oceans*, 119(10), 6575–6592, doi:10.1002/2014JC010061, 2014.

Weis, D. and Frey, F. A.: Submarine Basalts of the Northern Kerguelen Plateau: Interaction Between the Kerguelen Plume and the Southeast Indian Ridge Revealed at ODP Site 1140, *J. Petrol.*, 43(7), 1287–1309, doi:10.1093/petrology/43.7.1287, 2002.

### *Minor issues*

*The figures would benefit from clearly indicating the position of the polar front.*

We represented the PF on Figures 4 and 8 because we think that a representation of the front on the others figures will interfere with the major information they carry.

*The colour scale for the comparison of Lagrangian particle model and Ra ages is very difficult to read (I'm still not sure I understood what the grey colours mean)*

Dark grey represent surface waters with an age between 6 and 1 month. Light grey represent surface waters with an age between 1 and 2 months. The age are the time elapsed since the water body left the 2000 m isobaths, thus, white color is no contact with this isobath or a contact older than 2 months.

### **Response to Reviewer 3:**

We thank the reviewer for her/ his positive comments on the manuscript.

*1/ P. 14030, line 5-7, MnO<sub>2</sub> fibers are analyzed using different protocols and with different detectors. Were there any cross calibrations of the protocols and detectors?*

Cross calibration between the well-type and the semi-planar detectors are frequently done at the underground laboratory to validate the data. We have many data, including KEOPS-2 samples, to testify that. We added p6 "Cross-calibrations between the two detectors were made to avoid any bias in the determination of the Ra activities"

*2/ P. 14030, line 11-12, What are the uncertainties with the gamma detectors. It would be nice to propagate this source of error into the final uncertainties.*

By considering the uncertainty on the detection efficiencies of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , this increases the overall uncertainty by 1-3% only. This source of uncertainty is thus relatively minor in comparison with the uncertainty associated with counting statistics which is the main source of uncertainty.

3/ P. 14037, line 2, : : : are subject to :

Corrected p14

4/ P. 14037, Eq.1, The assumptions inherent to this equation should be made clear.

We added p 15 of the manuscript "The assumptions inherent to this equation can be found in Moore (2000) and are: (1) the  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  activities are constant in the source region (i.e. a constant initial  $^{224}\text{Ra}/^{223}\text{Ra}$  ratio is assumed), (2) the  $^{224}\text{Ra}/^{223}\text{Ra}$  ratio changes are only due to radioactive decay and (3) open ocean waters contain no excess  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$ ".

5/ P.14037, line 22-24, What is the range of the  $^{224}\text{Ra}/^{223}\text{Ra}$  ratios that were used as the initial ratio?

There is a relatively large range of  $^{224}\text{Ra}/^{223}\text{Ra}$  ratios between the samples collected at the beach and waters collected in coastal waters ( $1.50 \pm 0.84$  to  $20.64 \pm 13.83$ ). We did not want to calculate apparent ages using only the ratios of samples collected at the beach as an initial ratio. Therefore, we preferred to average the ratios of samples collected at stations with a bottom depth < 200 m. We used the mean  $^{224}\text{Ra}/^{223}\text{Ra}$  ratio found on the shallow plateau (that reflects the ratio found in shallow waters) as an initial ratio to derive apparent ages.

6/ Conclusions: In the last paragraph, it would be nice to mention that future studies should directly investigate the sources of Ra and chemical components (nutrients, Fe: : : etc) and quantify the fluxes of these species from bottom sediments, say, using the newly developed  $^{224}\text{Ra}/^{228}\text{Th}$  disequilibrium approach (e.g., Cai et al., 2012; 2014). References: a. Cai Pinghe, Xiangming Shi, Willard S. Moore, Minhan Dai (2012), Measurement of  $^{224}\text{Ra}:^{228}\text{Th}$  disequilibrium in coastal sediments using a delayed coincidence counter. *Marine Chemistry* 138-139, 1-6. b. Cai Pinghe, Xiangming Shi, Willard Moore, Shiyun Peng, Guizhi Wang, Minhan Dai (2014),  $^{224}\text{Ra}:^{228}\text{Th}$  disequilibrium in coastal sediments: Implications for solute transfer across the sediment-water interface. *Geochimica et Cosmochimica Acta* 125, 68-84.

A reference to the Cai et al., 2012 paper was added in the section 4.2 p 16 "Future studies in the area could want to track more precisely the sedimentary sources of Ra (and other chemical elements) and to quantify the Ra fluxes out of the sediments using e.g. the method described by Cai et al. (2012)"

Cai, P., Shi, X., Moore, W. S. and Dai, M.: Measurement of  $^{224}\text{Ra}:^{228}\text{Th}$  disequilibrium in coastal sediments using a delayed coincidence counter, *Mar. Chem.*, 138–139, 1–6, doi:10.1016/j.marchem.2012.05.004, 2012.

**Associate Editor Initial Decision: Publish subject to technical corrections** (13 Jan 2015) by Thomas Trull

**The authors thank the Editor for his careful revision of the last version of the manuscript. We now use the term “shelf” to identify the location where iron may be released. We may indeed have used the term “margin” not properly. We hope that the use of the appropriate geographical vocabulary (now “shelf”) contributes to improve the understanding of the discussion and finally the quality of this paper.**

Comments to the Author:

The paper is ready for publication, but needs attention to two final aspects.

1. Careful use of appropriate geographical vocabulary in English.

Specifically, the paper often refers to "the margins" as a location from which iron may be supplied.

But the meaning of this term is rather unclear. For example, in the Abstract there are three sentences that address this issue, yet it is unclear if they all refer to the same thing or are attempts to discriminate among different possible iron sources:

line 4 ...Kerguelen Islands, a bloom which is sustained partly by iron released from the sediments deposited onto the margins.

Line 15 ...chemical elements released from the sediments of the Kerguelen Plateau

Line 17...shallow sediments of the Kerguelen margins.

I think the best solution is probably to eliminate the use of the word “margins” which seems to be a loose translation from the French “marges” and replace it by shelves, or shelf edges, or island edges, or plateau sediments, depending on the specific context.

This issue needs attention primarily in the Abstract but also in the text at lines 30, 53, 58, 73, 264, 270, 284, 394, 450, 454

**In the Abstract, we replaced “margins” by “shelf” in line 5 and “shallow sediments of the Kerguelen margins” by “shelves of the Kerguelen Islands” in line 16.**

**In the text,**

**Lines 31, 54, 69 and line 75, we replaced “margins” by “shelves”**

**Line 59 we replaced “shallow sediments deposited onto the margins” by “shelf sediments”**

**Lines 271, 285, 451 and line 455, we replaced “shallow margins” by “shelves”**

2. Correction of a few remaining typos:

Line 34 change “...offshore the Kerguelen...” to “...offshore of the Kerguelen ...”

Line 44, add comma ‘...Kerguelen Islands, the numerous...’

Line 83: change “...iron fertilization off Kerguelen Islands...” to “...iron fertilization downstream of the Kerguelen islands...”

Line 164: change ‘...given by (Dibarboure et al., 2011).’ to “...given by Dibarboure et al. (2011).”

line 4 ...Ker

**We corrected all the remaining typos following the suggestions of the editor.**