Response to Referee #1

We thank Referee #1 for the helpful comments. We will address all changes in the revised manuscript as detailed in our responses below. The referee comments are in black and their line numbers refer to the revised manuscript. Our responses are in blue text.

- Specific comments: Methods 2.2 Radionuclide sampling. . . (lines 108-119): Were the samples precipitated at sea?

Yes, water samples were precipitated at sea. We will clarify it on Lines 107-110.

"For water samples, Po and Pb isotopes (including the added chemical yield tracers of ^{209}Po and stable lead) were co-precipitated with cobalt-ammonium pyrrolidine dithiocarbamate (Co-APDC) (Fleer and Bacon, 1984) at sea, but digested using concentrated HCl and HNO_3 back at the home laboratories."

- 2.4 Quantification of vertical advection (lines 172-201): This section should be clarified by pointing out that depth z in the layer for A1Po (0-z) refers to the different depths to which the 210Po was integrated for application of eqn 1 (i.e. the MLD, Z1%, PPZ and ThEq), as defined in the preceding section (2.3).

Thank you for your suggestion. We will clarify it on Lines 185-188 as the following:
"The activity gradient of ^{210}Po below the depth z (i.e., the MLD, z1%, PPZ, and ThEq) at each station was calculated from the depth z (using the average activity in the layer of 0-z m) as starting point ..."

- Results General comment- It would be very useful to present and plot the 210Po and 210Pb profile data.

The ^{210}Po and ^{210}Pb activity plotted as vertical profile at each station has been published in the companion paper (Tang et al., 2018). We will revise the sentence on Lines 269-272 as the following to clarify:

“The vertical profiles of total ^{210}Po and ^{210}Pb activity at each station have been described in the companion paper (Tang et al., 2018). Here we show the section view of the water column ^{210}Po deficit, which was calculated as total ^{210}Pb activity minus total ^{210}Po activity (Fig. 3).”

- 3.5 POC/210Po ratios in particles (lines 281-292); Values of POC/210Po in the large size fraction were not always “higher” than those in the small size fraction. Many were comparable and a few were lower. (Fig. 4a).

Thank you for pointing this out. We will revise this on Lines 298-302 as:
“Most of the ratios of POC concentration to ^{210}Po activity (μmol dpm^{-1}) in the large size fraction (POC/^{210}Po_LSF, > 53 μm) were comparable to or higher than those in the small size fraction (POC/^{210}Po_SSF, 1-53 μm), whereas a few samples had values of POC/^{210}Po_LSF lower than those of POC/^{210}Po_SSF at stations 13, 26, 44, 64, and 77 (Table 3, Fig. 4a).”

- Discussion 4.1 Physical Advection effects. . . (lines 317-322): It is certainly the case that horizontal physical transport is neglected in most 210Po studies because of lack of spatial resolution, but this is likely not justified- even in the “open ocean”. Many of the stations sampled could be called “open ocean”. Where exactly are advective effects negligible for Po?

The negligible horizontal advective effects for ^{210}Po is often justified in the open ocean settings where the horizontal velocities and/or horizontal gradients in ^{210}Po activities are small.

We will revise the sentence of Lines 336 to 340 as the following:

“This assumption of negligible horizontal physical transport has been made in most ^{210}Po studies because of a similar lack of spatial resolution (e.g., Kim and Church, 2001; Stewart et al., 2010; Rigaud et al., 2015), and may be justified in some open ocean settings where horizontal gradients in ^{210}Po activities are small (e.g., Wei et al., 2011).”

- 4.3 POC flux calculated from 210Po flux (lines 392-420): The authors use the total particulate POC/210Po ratio to calculate the POC flux from the 210Po flux, and as noted, this is equivalent to the POC/Po in the small size fraction. They justify this by arguing that the flux is dominated by the small size fraction. However, I recommend also using the POC/210Po on the large size fraction. Lemaitre et al. (2018) calculate POC fluxes from 234Th deficits using both size fractions, and it would be interesting to compare the Po-derived POC fluxes from both size fractions with the comparable Th-derived POC fluxes (section 4.5). The POC/Po (or POC/Th) ratio that truly applies is that on sinking particles, and that was not determined in either the present study or that of Lemaitre et al. (2018).

Thank you for your suggestion. We will add the POC flux estimation using the POC/^{210}Po ratios on the large size fraction and compare the ^{210}Po-derived POC fluxes from both total particulate fraction (TPF) and large size fraction (LSF) with the corresponding ^{234}Th-derived POC fluxes in section 4.5. The data will be presented as Figure 10:

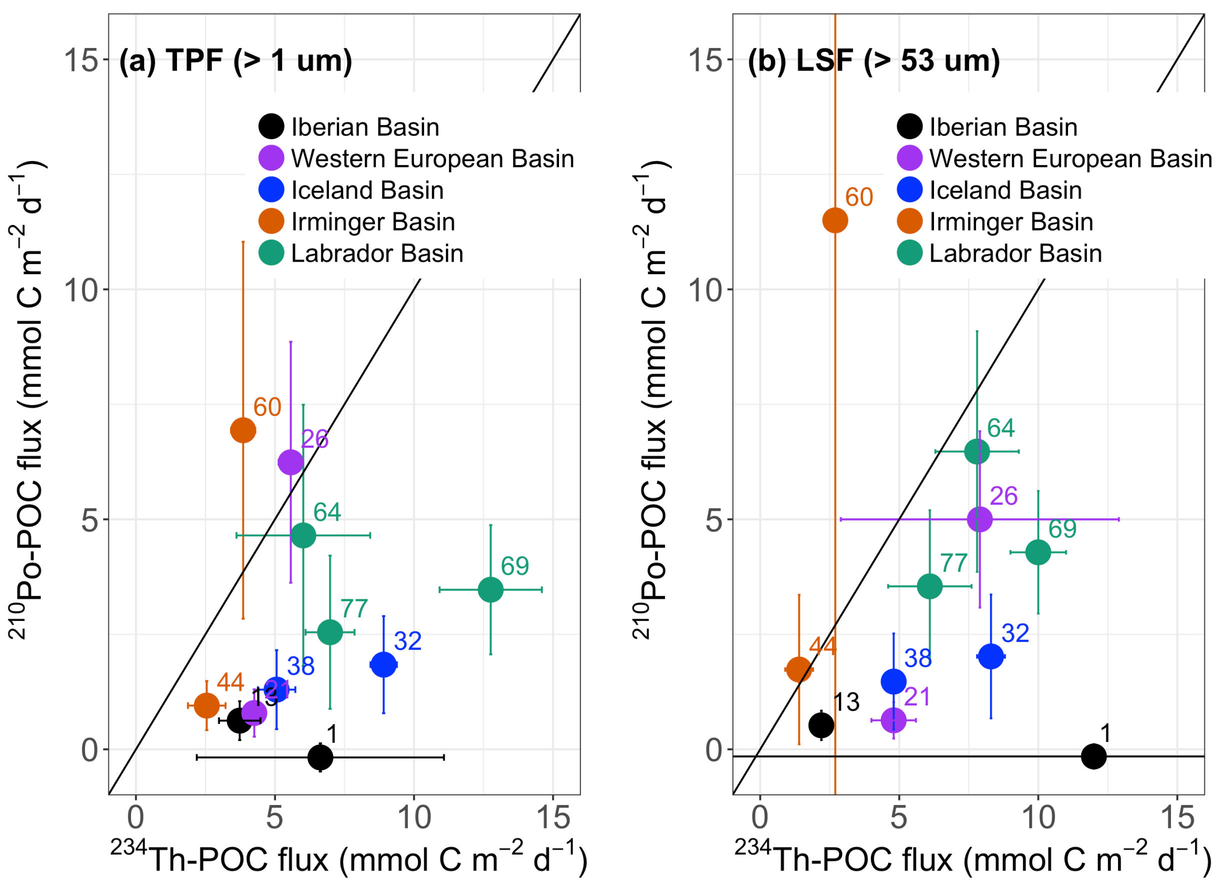

Fig. 10. Plot of the POC flux derived from ^{210}Po (^{210}Po-POC) versus the POC flux derived from ^{234}Th (^{234}Th-POC) at 11 stations along the GA01 transect. Both the fluxes of ^{210}Po and ^{234}Th were calculated from the deficit term alone assuming steady state and negligible physical transport.

The POC/radionuclide ratios on (a) total particulate fraction (TPF, > 1 µm) and (b) large size fraction (LSF, > 53 µm) were used to calculate the POC flux. The fluxes were integrated down to the depth where ^{234}Th activity returned to equilibrium with ^{238}U activity (ThEq). The numbers in the plot are station numbers. The color codes of the stations correspond to the basins.

We agree with the comment that in situ pump filtered particles, either operationally defined small (1-53 µm) or large (> 53 µm) size fractions, may both represent a mixture of sinking and non-sinking particles. Therefore, in the present study and that of Lemaitre et al., (2018), we cannot obtain the POC/^{210}Po or POC/^{234}Th ratios on the "true" sinking particles.

We will change the manuscript by adding:

Section 4.5.2:

Lines 565-569: "In situ pump filtered particles, either operationally defined as small (1-53 µm, SSF), large (> 53 µm, LSF), or total (> 1 µm, TPF) particles, may all represent a combination of sinking and non-sinking particles. In the present study, the particulate POC/radionuclide ratio on the TPF and LSF were examined and used to calculate POC export flux."

Lines 574-578: "In comparison, the LSF POC/^{210}Po ratios were not correlated with LSF POC/^{234}Th ratios (n = 11, R^2 = 0.3, p-value =0.07). The correlation of values within the TPF but not the LSF suggests that the composition of large particles was different from that of the total particles, and that the difference in particle association between POC and ^{210}Po and ^{234}Th was greater in large than total particles".

Section 4.5.3:

Lines 585-589: "When the radionuclide fluxes were multiplied by the POC/radionuclide values, the range of the calculated POC fluxes were negligible to 7 mmol C m^{-2} d^{-1} and negligible to 12 mmol C m^{-2} d^{-1} via the ^{210}Po method using the TPF and LSF POC/^{210}Po ratios, respectively; and from 2.5 to 13 mmol C m^{-2} d^{-1} and from 1 to 12 mmol C m^{-2} d^{-1} via the ^{234}Th method using the TPF and LSF POC/^{234}Th ratios, respectively (Fig. 10)."

Lines 600-607: "When using the large particle POC/radionuclide ratios, only in the Irminger Basin was there higher ^{210}Po-derived POC flux than ^{234}Th-derived POC flux (> 0.2 to 3-fold). In the Iberian Basin, the greatest difference between the methods was found at station 1 where the ^{234}Th-POC flux was greater than the ^{210}Po-POC estimate by 4-fold. In the Western European, Iceland, and Labrador Basins, the ^{234}Th-POC fluxes were larger than ^{210}Po-POC estimates by 5, 4, and 2-fold, respectively."

Lines 611-617: "Since the ratios of POC to radionuclides on total particles had very similar spatial trends along the transect, the discrepancy between TPF ^{234}Th-POC and TPF ^{210}Po-POC flux estimates must be driven primarily by the discrepancy between the SS estimates of ^{234}Th and ^{210}Po fluxes, discussed in section 4.5.1. In contrast, the discrepancy between the POC to isotope ratios in the large particle may have led to some degree of discrepancy between the LSF ^{234}Th-POC and LSF ^{210}Po-POC flux estimates."

- 4.5.1 210Po flux vs. 234Th flux (lines 473-483): Comparing these fluxes in relation to the stage of bloom at different stations is extremely interesting and bears on the use of these two radionuclides as POC flux proxies. Given the large difference in half-lives, one might expect the 210Po deficit resulting from a bloom to persist longer than that of 234Th. If the eastern section was sampled weeks to months after the bloom (Fig. 7), I would expect Po deficits and fluxes to be higher (proportionately) than those of 234Th. More comparable fluxes (i.e. both high) would be expected at stations sampled right after the bloom (i.e. the western section). Fig. 9 shows the latter pattern for the western section stations, but it shows much higher 234Th fluxes sampled in the eastern section stations. This is counterintuitive if the bloom happened weeks to months before. What conclusions can be drawn from this? Does it reflect advective effects on profiles of the two radionuclides? But if so, why do the effects seem higher for 243Th?

First of all, the ^{210}Po deficit integrates the flux over a longer period of time, tending to smooth out episodic export events and resulting in a lower mean. Specifically, the ^{210}Po deficit integrates the flux over months that actually include a period of lower flux prior to the bloom along the GA01 transect, whereas ^{234}Th deficit integrates the flux over weeks that generally represents the bloom itself (Fig. 6). Thus, the POC export fluxes derived from ^{234}Th were generally found higher than those derived from ^{210}Po (Verdeny et al., 2009, and references therein) due to the inherent difference in half-lives between ^{234}Th (24.1 d) and ^{210}Po (138.4 d).

Second, the reason why ^{234}Th export flux was higher in the eastern than western section has not been directly discussed in Lemaitre et al., (2018); the authors focused more on the POC export fluxes that were derived from ^{234}Th along the transect. In the present study, we attempted to explain the trend of ^{234}Th export flux between western and eastern sections mainly in relation to the stage of the bloom. In the Iberian Basin of the eastern section, stations 1 and 13 were sampled weeks to months after the bloom development. Such time periods were generally longer than the mean life of ^{234}Th (35 days). The moderate to relatively high ^{234}Th fluxes observed in this basin are thus surprising. Lemaitre et al. (2018) argue that the greater fluxes there might be related to the proximity of the Iberian margin, where particle dynamics are intense and lithogenic particles are numerous (Gourain et al., 2018). A temporal decoupling between production and export could be an alternative possibility. In the Western European and Iceland Basins, the sampling was carried out during the bloom development with the NPP peaks occurring just before sampling. The high export of ^{234}Th flux there may be associated with the fast sinking particles that have formed during the bloom development. Similar high export associated with fast sinking particles has been reported in the Western European Basin, at the PAP site, during the spring bloom in May 2009 (Villa-Alfageme et al., 2016). In the western section, low export flux of ^{234}Th was suggested to be due to particles' retention in the surface water rather than export in the Irminger Basin (Lemaitre et al., 2018). In the Labrador basin, the relatively low export flux at the decline of the bloom could be also due to the temporal decoupling between production and export.

Third, unlike the higher ^{234}Th export flux in the eastern than western section, no significant difference in ^{210}Po export flux between the two sections was observed. This in fact supports the previous argument that ^{210}Po deficit tends to smooth out episodic events due to integrating over a longer time scale including a period of lower flux, whereas ^{234}Th deficit represents more recent changes in the water column.

Finally, lower ^{234}Th export flux in the western section was unlikely due to the neglecting of advection, as physical processes were suggested to have very limited impact on the measured ^{234}Th export fluxes along the transect (Lemaitre et al., 2018).

Overall, we believe the different trends between ^{234}Th and ^{210}Po export flux along the transect were mostly due to the large difference in the half-lives of the two radioisotopes.

We will rewrite Lines 527-554 as the following:

Lines 527-554:

"These relationships may be related to both the stage of the bloom and different half-lives of the two isotopes. Indeed, ^{234}Th fluxes integrate the conditions that occurred days to weeks prior to the sampling date while the ^{210}Po method integrates the flux over the past few months.

Within the Iberian Basin, stations 1 and 13 were sampled weeks to months after the bloom development (Fig. 6). The moderate to relatively high ^{234}Th fluxes are thus surprising. Lemaitre et al. (2018) argue that the greater fluxes there might be related to the proximity of the Iberian margin, where particle dynamics were intense and lithogenic particles were numerous (Gourain et al., in review). A temporal decoupling between production and export could be an alternative possibility. The Western European and Icelandic Basins were sampled during the bloom development and the NPP peaks occurring just before sampling may have promoted the high fluxes. In fact, these basins have been characterized by the presence of fast-sinking particles during the bloom (Villa-Alfageme et al., 2016), likely also explaining the high exports. In contrast, the lower exports observed in the western section may be related to the fact that the sampling occurred during the decline of the bloom probably with a decoupling between production and export in the Labrador Basin or during particle retention event in the Irminger Basin.

Unlike the observation of higher ^{234}Th export flux in the eastern than western sections, there was no significant difference in ^{210}Po export flux between the two sections. This observation supports the argument that the ^{210}Po deficit tends to smooth out episodic events due to integration over longer time periods. The ^{210}Po deficit records seasonal changes in export fluxes, whereas the ^{234}Th deficit represents more recent changes in the water column (Verdeny et al., 2009; Hayes et al., accepted). Indeed, the ^{210}Po deficit integrates the flux over months that include a period of lower flux prior to the bloom along the GA01 transect, whereas the ^{234}Th deficit integrates the flux only over weeks that include the bloom itself at most of the stations (Fig. 6). Therefore, the specific stages of the bloom shortly prior to the sampling date appears to have a smaller influence on the ^{210}Po-derived than the ^{234}Th-derived export flux along the transect."

- 4.5.3 210Po-derived POC flux vs 234Th-derived POC flux (lines 494-513): The authors have estimated the effects of vertical advection on the Po deficits (and Po-derived POC fluxes). Are these advective effects comparable for 234Th fluxes? One could argue that they would be lower, and, it might be preferable to compare the NSS Po-derived POC fluxes with the SS Th-derived POC fluxes. This seems to improve some of the station comparisons in Fig. 10, but not all. In the end, the difference is attributed to the discrepancy between the Po and Th flux estimates (as shown in Fig. 9), but that discrepancy remains unexplained in my mind (see comment above).

The influence of vertical advection on ^{234}Th export flux along the transect was suggested to be negligible (Lemaitre et al., 2018). Indeed, when the same method described in section 2.3 in the present study was used to calculate the impact of vertical advection on ^{234}Th export flux at the ThEq, we found that the vertical advective flux of ^{234}Th was below or close to the uncertainty of the deficit flux at 10 out of 11 stations. The vertical advection at station 77, in contrast, increased the calculated flux by 40%. Overall, the influence of vertical advection on ^{234}Th export flux was indeed lower than that on ^{210}Po flux and therefore can be ignored. As the influence of vertical advection on ^{234}Th export flux has been thoroughly discussed in Lemaitre et al., (2018) we will not discuss this further in the present study but add the following sentence on Lines 510-512: "The authors discussed the influence of vertical advection on ^{234}Th export flux and conclude it can be neglected."

For Figure 10, both ^{210}Po-derived POC flux and ^{234}Th-derived POC flux were estimated using the stead state (SS) model by neglecting physical processes. Lemaitre et al., (2018) quantified the non-steady state (NSS) ^{234}Th fluxes using the model developed by Savoye et al., (2006). In the present study, we did not attempt to quantify the NSS ^{210}Po fluxes using the same technique for the reasons that have been described on Lines 427-431 as "We did not attempt to apply the same technique to estimate NSS ^{210}Po in this study because the assumption of equilibrium between ^{210}Po activity and ^{210}Pb activity at the starting date of the bloom may be inappropriate. One confounding factor is the timescale of events; the ^{210}Po deficit integrates over a longer time period (months) than a typical bloom event (days/weeks).". We however, discussed the NSS effect on the ^{210}Po fluxes in a more "descriptive" manner.

The discrepancy between the ^{210}Po and ^{234}Th flux estimates will be explained in section 4.5.1 as the following:

"These relationships may be related to both the stage of the bloom and different half-lives of the two isotopes. Indeed, ^{234}Th fluxes integrate the conditions that occurred days to weeks prior to the sampling date while the ^{210}Po method integrates the flux over the past few months.

Within the Iberian Basin, stations 1 and 13 were sampled weeks to months after the bloom development (Fig. 6). The moderate to relatively high ^{234}Th fluxes are thus surprising. Lemaitre et al. (2018) argue that the greater fluxes there might be related to the proximity of the Iberian margin, where particle dynamics are intense and lithogenic particles are numerous (Gourain et al., in review). A temporal decoupling between production and export could be an alternative possibility. The Western European and Icelandic Basins were sampled during the bloom development and the NPP peaks occurring just before sampling may promote high fluxes. Moreover, these basins have been characterized by the presence of fast-sinking particles during the bloom (Villa-Alfageme et al., 2016), likely also explaining the high exports. In contrast, the lower exports observed in the western section may related to the fact that the sampling occurred during the decline of the bloom probably with a decoupling between production and export in the Labrador Basin or during particle retention event in the Irminger Basin.

Unlike the observation of higher ^{234}Th export flux in the eastern than western sections, there was no significant difference in ^{210}Po export flux between the two sections. This observation supports the argument that the ^{210}Po deficit tends to smooth out episodic events due to integration over longer time periods. The ^{210}Po deficit records seasonal changes in export fluxes, whereas the ^{234}Th deficit represents more recent changes in the water column (Verdeny et al., 2009; Hayes et al., accepted). Indeed, the ^{210}Po deficit integrates the flux over months that include a period of lower flux prior to the bloom along the GA01 transect, whereas the ^{234}Th deficit integrates the flux only over weeks that include the bloom itself at most of the stations (Fig. 6). Therefore, the specific stages of the bloom shortly prior to the sampling date appears to have a smaller influence on the ^{210}Po-derived than the ^{234}Th-derived export flux along the transect."

References:

Gourain, A., Planquette, H., Cheize, M., Menzel-Barraqueta, J. L., Boutorh, J., Shelley, R. U., Pereira-Contreira, L., Lemaitre, N., Lacan, F., Lherminier, P. and Sarthou, G.: Particualte trace metals along the GEOVIDE section, Biogeosciences, 2018.

Hayes, C. T., Black, E. E., Andersen, R. A., Baskaran, M., Buesseler, K. O., Charette, M. A., Cheng, H., Cochran, J. K., Edwards, R. L., Fitzgerald, P., Lam, P. J., Lu, Y., Morris, S. O., Ohnemus, D. C., Pavia, F. J., Stewart, G. and Tang, Y.: Flux of particulate elements in the North Atlantic Ocean constrained by multiple radionuclides, Global Biogeochemcal Cycles, https://doi.org/10.1029/2018GB005994

Kim, G. and Church, T. M.: Seasonal biogeochemical fluxes of 234Th and 210Po in the Upper Sargasso Sea: Influence from atmospheric iron deposition, Global Biogeochemcal Cycles, 15, 651-661, 10.1029/2000GB001313, 2001.

Lemaitre, N., Planchon, F., Planquette, H., Dehairs, F., Batista, D. F., Roukaerts, A., Deman, F., Tang, Y., Mariez, C. and Sarthou, G.: High variability of export fluxes along the North Atlantic GEOTRACES section GA01: Particulate organic carbon export deduced from the 234Th method, Biogeosciences, 15, 6417-6437, https://doi.org/10.5194/bg-15-6417-2018, 2018.

Rigaud, S., Stewart, G., Baskaran, M., Marsan, D. and Church, T.: 210Po and 210Pb distribution, dissolved-particulate exchange rates, and particulate export along the North Atlantic US GEOTRACES GA03 section, Deep Sea Research Part II, 116, 60-78, 10.1016/j.dsr2.2014.11.003, 2015.

Savoye, N., Benitez-Nelson, C., Burd, A. B., Cochran, J. K., Charette, M., Buesseler, K. O., Jackson, G. A., Roy-Barman, M., Schmidt, S. and Elskens, M.: 234Th sorption and export models in the water column: A review, Marine Chemistry, 100, 234-249, 2006.

Stewart, G. M., Bradley Moran, S. and Lomas, M. W.: Seasonal POC fluxes at BATS estimated from 210Po deficits, Deep Sea Research Part I: Oceanographic Research Papers, 57, 113-124, 10.1016/j.dsr.2009.09.007, 2010.

Tang, Y., Castrillejo, M., Roca-Martí, M., Masqué, P., Lemaitre, N. and Stewart, G.: Distributions of total and size-fractionated particulate 210Po and 210Pb activities along the North Atlantic GEOTRACES GA01 transect: GEOVIDE cruise, Biogeosciences, 15, 5437-5453, https://doi.org/10.5194/bg-15-5437-2018, 2018.

Verdeny, E., Masqué, P., Garcia-Orellana, J., Hanfland, C., Kirk Cochran, J. and Stewart, G. M.: POC export from ocean surface waters by means of 234Th/238U and 210Po/210Pb disequilibria: A review of the use of two radiotracer pairs, Deep Sea Research Part II, 56, 1502-1518, 10.1016/j.dsr2.2008.12.018, 2009.

Villa-Alfageme, M., Soto, F. C., Ceballos, E., Giering, S. L. C., Le Moigne, F. A. C., Henson, S., Mas, J. L. and Sanders, R. J.: Geographical, seasonal, and depth variation in sinking particle speeds in the North Atlantic, Geophysical Research Letters, 43, 8609-8616, doi:10.1002/2016GL069233, 2016.

Wei, C. L., Lin, S. Y., Sheu, D. D. D., Chou, W. C., Yi, M. C., Santschi, P. H. and Wen, L. S.: Particle-reactive radionuclides 234Th, 210Pb, 210Po as tracers for the estimation of export production in the South China Sea, Biogeosciences, 8, 3793-3808, 2011.