**Response to Referee #2:**

Dear Referee, we appreciate your insightful and thorough comments and suggestions, according to which we improved our revised MS carefully. Thank you for your time and critical evaluation and enclosed please kindly find our responses (written in blue) as follows.

**General comments:**

The authors applied 3 models, the three-end member mixing model, the mass-balance model, and the time-series model to estimate the SGD flux to the Krka River estuary. In calculating the flushing time, the river discharge is not included, which should be considering the great river discharge of the Krka River during the investigation. In the three end-member mixing model and the mass-balance model, desorption of radium as Ra a source in estuaries is not included or evaluated. In the time-series model, the assumptions are not substantiated. Ra and water depth at the time-series station change with tide/time. These changes are not only due to SGD, but also due to seawater into the estuary. The estuarine Ra background thus changes with time. All these changes are not considered in the time-series model. Only when these models are corrected, can the results and discussion be assessed and evaluated. There is not much discussion on implications for SGD-associated nutrients. It's better for the authors to turn to an English editor to go through all the texts.

**RESPONSE:**

Thank you very much for your comments. We did a major revision of our manuscript according to your suggestions and hope that we achieved a desired scientific quality both in using the most appropriate methods and in presentation of our results. We will explain all the details considering the improvements according to your comments.

Firstly, we have added the river discharge term into flushing time model and used the equation from Sanford et al. (1992) to re-calculate the flushing time (Equation 7).

Secondly, considering the low suspended particle matter concentration in the Krka River Estuary, in the three end-member mixing model and the mass-balance model, we have evaluated desorption of radium from particles and it appeared to be a negligible term. Therefore, we did not include it into the model. We agree that our presentation was unclear in the submitted original manuscript, and therefore we have corrected these in the revised manuscript.

Thirdly, in the time series model, we have chosen the minimum measured Ra activity of the time series observation as the background of the estuarine water. The estimated SGD was obtained by subtraction of background Ra activities from the measured Ra activities. So, we could exclude the other Ra sources variations.

The similar cases can be found in our previous work and other publications (Peterson et al., 2008; Wang et al., 2016).

Fourthly, we have extended the discussion on implications for SGD-associated nutrients in the revised manuscript.

Finally, the whole manuscript is edited by the professor who is a native English speaker.

Sanford, L. P., Boicourt, W. C., and Rives, S. R.: Model for estimating tidal flushing of small embayments. J. Waterw., Port, Coastal, Ocean Eng., 118(6):635-54, doi: 10.1061/(ASCE)0733-950X(1992)118:6(635), 1992.

Peterson, R. N., Burnett, W. C., Taniguchi, M., Chen, J., Santos, I. R., and Ishitobi, T.: Radon and radium isotope assessment of submarine groundwater discharge in the Yellow River delta, China. J. Geophys. Res.: Oceans, 113(C9), doi:10.1029/2008JC004776, 2008.

Wang, X. and Du, J.: Submarine groundwater discharge into typical tropical lagoons: A case study in eastern Hainan Island, China, Geochem. Geophys. Geosyst., 17(11), 4366-4382, doi:10.1002/2016GC006502, 2016.

**Minor comments:**

Page 1 Line 17: ‘in tidal period’ specify the time frame: 24 hours or 12 hours.

**RESPONSE:** It’s 24 hours, and we have added it into the revised manuscript.

Line 21: ‘9.5-38.3% to the total DSi flux’ can’t be taken as ‘a major source’.
**RESPONSE:** We have corrected it according to new calculations.

Line 22: ‘likely’ is not proper to be used here. Quantitative results should allow the authors to determine whether SGD is a major source or not. This is no longer a possibility.

**RESPONSE:** We have corrected it according to new calculations.

Line 26: what is a river-dominated estuary? The sentence “It is the primary pathway: : :” is awkward. Rewrite it.

**RESPONSE:** We have rewritten that as “An estuary is the critical zone connecting the mainland and adjacent sea, and the primary region where continuous exchange of water and chemical components between land and sea/ocean occurs.”

Page 2 Line 23: what is tidal amplitude? Tidal range? Tidal height?

**RESPONSE:** It is the tidal range and we have corrected it in the revised manuscript.

Line 27: “have” to “has”; “transporting” to “transported”. Line 34: “transect” to “transects”. Page 3 Line 5: “24 hours-time” to “24-hour time”

**RESPONSE:** We have corrected them.

Line 9: dissolved oxygen (DO) measured using a multi-parameter probe needs to be calibrated with DO measured using classic Winkler titration. Otherwise, DO data are questionable.

**RESPONSE:** The multi parametric probe has been calibrated using the Winkler titration. We have found a significant linear regression ($R^2$=0.791, n=88, df=86, $p<0.0001$) between DO values from the Winkler method and the multi parametric probe. Winkler method values were higher and considered as accurate, therefore all measurements from the probe were recalculated to the Winkler values via the equation of the regression line: Winkler DO=0.972*Probe DO+0.507 mg $O_2$/l. The conversion factor 1 mL $O_2$/L=1.42903 mg $O_2$/L (Owens and Millard 1985, Garcia and Gordon 1992) was employed. As we did not expect hypoxic samples in our study, i.e. the most accurate measurements by the Winkler method were not a priority, we used calibrated multi-parameter probe.

Garcia, H.E. and Gordon, L.I.: Oxygen solubility in seawater: Better fitting equations, Limnol. Oceanogr., 37(6),1307-1312, doi: 10.4319/lo.1992.37.6.1307, 1992.

Owens, W.B. and Millard Jr, R.C.: A new algorithm for CTD oxygen calibration, J. Phys. Oceanogr., 15(5), 621-631, doi: 10.1175/1520-0485(1985)015<0621:ANAFCO>2.0.CO;2, 1985.

Line 13: the pore size seems too big to do the filtration. Double check the pore size of the cartridge.

**RESPONSE:** We made a mistake. The pore size is 0.5 µm and we have corrected it in the revised manuscript.

Line 17: the sentence “the 228Ra activities: : :” needs to be revised.

**RESPONSE:** We have revised it as follows “while $^{228}Ac$ (338 keV and 911 keV peaks) was used for measuring $^{228}Ra$ activity.”

Line 24: Briefly explain the method used to measure these nutrients. Provide the detection limits of these nutrients.

**RESPONSE:** The concentrations of nitrate ($NO_3^-$), nitrite ($NO_2^-$), $NH_4^+$, reactive orthosilicates ($SiO_4^{4-}$, hereafter termed DSi), orthophosphate ($PO_4^{3-}$, hereafter termed dissolved inorganic phosphorus, i.e. DIP), were determined as described in Strickland and Parsons (1972) and Grasshoff et al. (1983), using a spectrophotometer (PerkinElmer Lambda15) combining 1 cm and 10 cm cuvettes, as needed. The detection limits and reproducibility for nutrients were as follows: 0.05 and 0.025 μmol $L^{-1}$ for $NO_3^-$; 0.01 and 0.01 μmol $L^{-1}$ for $NO_2^-$; 0.1 and 0.098 μmol $L^{-1}$ for $NH_4^+$; 0.1 and 0.06 μmol $L^{-1}$ for $SiO_4^{4-}$ and DIP 0.03 and 0.03 μmol $L^{-1}$ for DIP. We have added these into the revised manuscript.

Grasshoff, K., Kremling, K., and Ehrhardt, M.: Methods of seawater analysis, 2nd ed. Weinheim: Verlag Chemie GmbH, 1983.

Strickland, J. D. and Parsons, T. R.: A practical handbook of seawater analysis, 2nd ed. Ottawa, Bulletin of the Fisheries Research Board of Canada, 1972.

Line 28: “investigated” to “investigation”.

**RESPONSE:** We have Corrected it.

Fig. 5 Line 15: “It was particularly pronounced for 228Ra that had lower effect than in the open Adria Sea”. What effect?

**RESPONSE:** Due to the shorter half-life of $^{228}$Ra, in the open Adriatic Sea the $^{228}$Ra activity is much lower than that in the estuary relative to the $^{226}$Ra activity. Therefore, using $^{228}$Ra to establish the three end-member mixing model is more appropriate due to its lower mixing effect from the open sea.

Line 16-18: how about adsorption of radium from particles as a source? It is not included in your three end-member mixing model and its effect should be evaluated.

**RESPONSE:** As we stated above, we have evaluated desorption of radium from particles, which appeared to be a negligible term, so we did not include it into the model. Our presentation was unclear in the original manuscript, and we have corrected that in the revised manuscript.

Fig. 6 Line 3: give the values of each fraction to support the statement “the fraction of the river water was higher than those of the open seawater and groundwater”.

**RESPONSE:** We have corrected it.

Line 4: “lower changes” is an awkward phrase. Is 28-37% the change in the fraction or the fraction?

**RESPONSE:** It means the smaller variation range. We have corrected it.

Line 10: This physical model is not proposed by Moore et al. (2006). Cite the original paper. Moreover, the model used here is suitable for estuaries with low river discharge, so river discharge is not considered. In your case the river discharge is not negligible and should be included in calculating the flushing time. Refer to the original paper for the proper formula to use here.

**RESPONSE:** We have added the river discharge term into the model and used the equation from Sanford et al. (1992) to re-calculate the flushing time as mentioned above.

Line 27: Every parameter on the right hand side of Eq. (8) is given as an average value with a standard deviation. How is the value on the left hand side of Eq. (8) calculated to be a range of values?

**RESPONSE:** We obtained the value with a standard deviation first, and then presented it as a range. Since we used three methods to estimate the SGD flux, we believe that using a range that covers all the values is accurate, and the values presented in this style were reported only for the SGD flux and its derived nutrients in our manuscript.

Page 7 Line 6: The sentence is broken.

**RESPONSE:** We have corrected it.

Line 12: Again in Eq. (10) desorption of radium from particles is a source of dissolved radium that needs to be included in the mass-balance model.

**RESPONSE:** We have corrected that in the revision as mentioned above.

Line 27: the estuarine background changes with tides. When a minimum is chosen to be the estuarine background, an overestimate of SGD may result.

**RESPONSE:** The Ra background generally includes riverine inputs, desorption from particles and mixing with an open sea, in which the mixing term changes with tides' height variation; and the minimum in the time-series observation also includes the SGD source. The variation range of mixing with open sea is much lower than the SGD source (Figure 8). Therefore, for each measured Ra activity, the minimum is subtracted out and that results in a conservative estimation.

Page 8 Line 1-3: the time-series Ra activity changes with tide. I suspect that the surface layer depth changes also with tide (the observation of water depth at the time-series station will verify this). Then the excess Ra inventory calculated with a constant water depth is not appropriate.

**<u>RESPONSE:</u>** The water depth varied within the time series observation, but the amplitude was small (no more than 0.3 m here), so we neglected the variation and assumed it to be a constant water depth, as also used by Peterson et al. (2008) and Wang et al. (2016).

Peterson, R. N. Burnett, W. C., Taniguchi, M., Chen, J., Santos, I. R., and Ishitobi, T.: Radon and radium isotope assessment of submarine groundwater discharge in the Yellow River delta, China. J. Geophys. Res.: Oceans, 113(C9), doi:10.1029/2008JC004776, 2008.

Wang, X. and Du, J.: Submarine groundwater discharge into typical tropical lagoons: A case study in eastern Hainan Island, China, Geochem. Geophys. Geosyst., 17(11), 4366-4382, doi:10.1002/2016GC006502, 2016.

Figure 1. Groundwater sampling stations are not shown.

**<u>RESPONSE:</u>** We have corrected it.

Figure 7. Only one groundwater point is shown. From Table 1 two groundwater samples were collected for Ra and both should be shown here.

**<u>RESPONSE:</u>** We have corrected it.