Response to review

Respect Dr. Pokrovsky:

We want to begin by thanking Dr. Pokrovsky for writing that “Conclusions nicely reflect the main findings, and even if some of them are speculative (L 675-678), they can be stated as they are.” We greatly appreciated the constructive comments and suggestions to improve the original manuscript. Based on the comments, we have made carefully revision. We addressed all the points raised, as summarized below.

Comment 1. L74-87: This is too detailed literature review, not directly linked to the subject of this study. It is probably not necessary.

Response: Thanks for the advices. We have deleted those sentences.

Comment 2. Physio-geographical parameters of rivers should be listed in a table (% of coverage by peat, degree of affection by palm plantations, runoff, slope etc).

Response: Thanks for the great advice. We have added those parameters in Table R1 (i.e. Table 1 in the manuscript). However, the slope of the rivers was not available.

Table R1 The physio-geographical parameters of sampled rivers. (n.a. stands for not available.)

River Names	Total Basin [a]	Runoff (km ^{3} yr^{-1})	Coverage rate by peat (%) [a]	Degree of affection by palm plantations (%)[a]
Rajang	50000	114 [b]	7.7	9.1
Maludam	197	0.14 [c]	87	8.1
Sebuyau	538	n.a.	54	4.5
Simunjan	788	n.a.	44	30
Samusam	163	n.a.	10	0
Sematan	287	n.a.	0	0

[a] Modified from Bange et al., 2019

[b] Cited from Staub et al., 2000

[c] Cited Müller et al., 2016

Comment 3. Three type of Se behavior are well identified and summarized in L349-358. However, the presentation of each individual river in Figs 2-5 takes too much space. Either consider presenting just an example of each group or the average of all rivers in each group

Response: Thanks for the advice. We have moved Fig. 2 and Fig. 4 to the supplement, and Fig. 3 and Fig. 5 were merged to one figure. The three typical groups of relationships between Se species and salinity for Rajang, Maludam and Sematan estuaries were selected to present in Fig. R1 (i.e. Fig. 3 in the manuscript), and those for Sebuyau and Samunsam were moved to supplement.

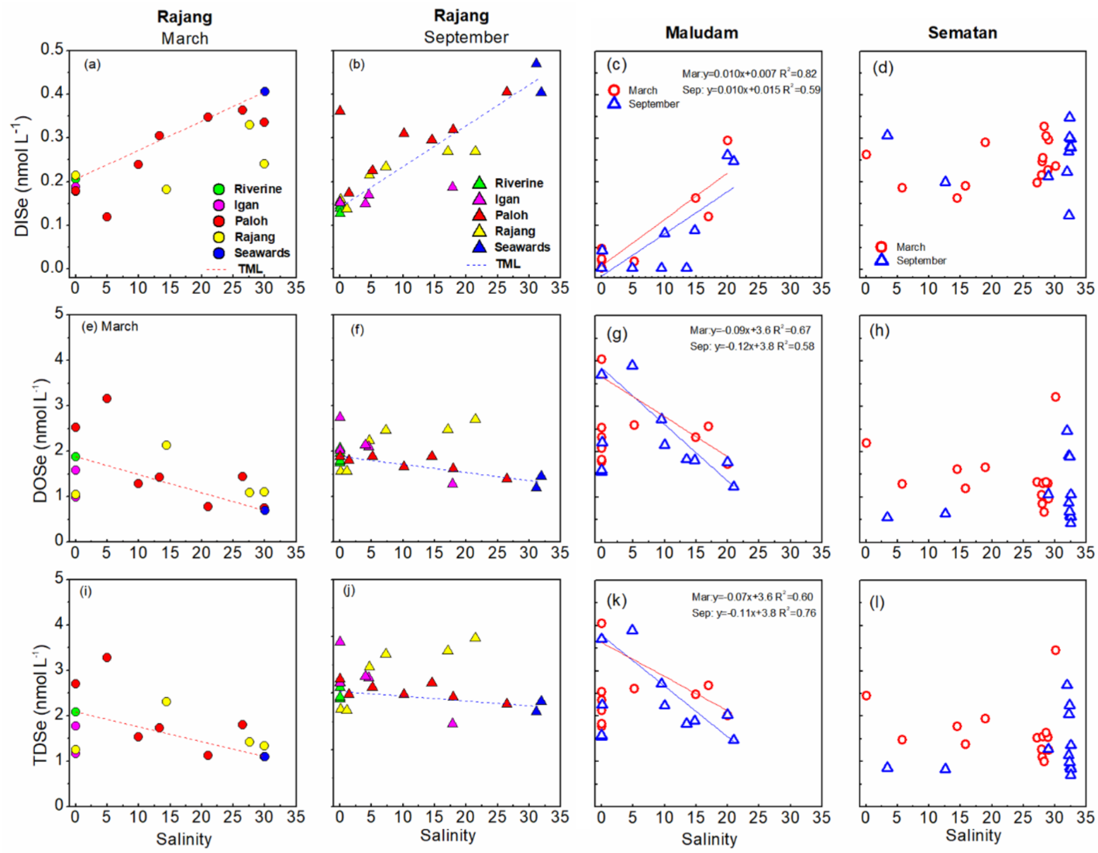

Fig. R1. Relationships between DISe (a - d), DOSe (e - h), and TDSe (i - l) concentrations with salinity in the Rajang and three Rajang tributaries (Igan, Lassa, and Rajang), and in the Maludam and Sematan estuaries in March and September 2017. TML refers to the theoretical mixing line, which was defined using two endmembers: freshwater in the riverine system and seawater.

Comment 4: L407-412: Please explain, what is the mechanism of Se(IV)/Se(VI) increase with DO increase. Oxidation is more pronounced at high DO, yet the observations are reverse to that.

Response: As shown in Fig. R2, Se(IV)/Se(VI) ratios in the freshwater of the sampled rivers increased as DO concentrations increased statistically. However, the Se(IV)/Se(VI) ratios were calculated only if Se(IV) and Se(VI) concentrations were both above the detection limits, meaning that the data are limited. The limited Se(IV)/Se(VI) ratios roughly fell into two groups, one group was low Se(IV)/Se(VI) ratios with low DO concentration and the other was high Se(IV)/Se(VI) ratios with high DO concentration. The liner relationship between Se(IV)/Se(VI) ratios and DO concentration probably was false appearance with the limited data. We have deleted this figure in the manuscript.

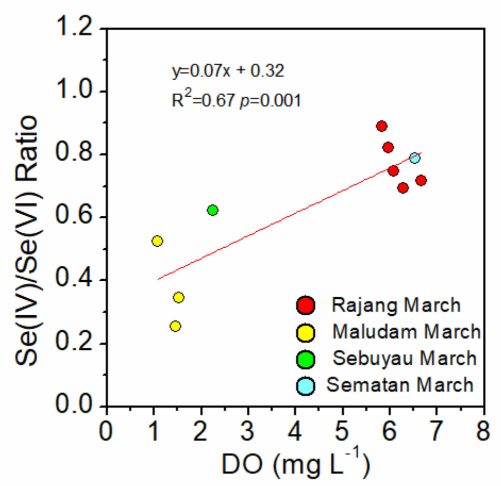

Fig. R2 Relationships between Se(IV)/Se(VI) ratios and DO values n freshwater (Salinity < 1) for the Rajang, Sematan, Maludam, Sebuyau, Samunsam, and Simunjan rivers in March and September. Se(IV)/Se(VI) ratios were calculated only if Se(IV) and Se(VI) concentrations were both above the detection limits, meaning that the data are limited.

Comment 5: L434-439: This information should be in the site description table of river watershed parameters

Response: We have deleted those sentences, changed to “As shown in table 1”.

Comment 6: L468-477: There are certainly some structural data (e.g., XAS) on molecular status of Se bound to organic matter.

Response: Thanks for the advices, we have deleted those sentences, including “However, the mechanisms behind the interactions between Se and dissolved organic ligands are still poorly understood.”

Comment 7: L505-507: The analogy with NO3 is not straightforward: nitrate is a nutrient but Se is not always a nutrient

Response: We have deleted the “Nitrate behaves in a similar way in the Dumai River estuary (Sumatra, Indonesia), another tropical blackwater river

(Alkhaitb and Jennerjahn, 2007)."

Comment 8： Fig 8a-c present the data from other papers and as such not necessary. Citations of main results from these papers would be enough.

Response: Consider that the DOSe were not related to the CDOM in the Rajang, we have deleted the Figure 8a-e, kept Figure 8f-h as Figure 5 in the revised manuscripts.

Comment 9: L580-643: Basically, the same comment. Too many specific details from other papers; the whole section can be greatly shortened, and only main findings are presented.

Response: We have deleted those specific literature details and shorted this section from 63 lines to 41 lines, as followed:

"Moreover the peat-draining rivers demonstrated a liner relationship between DOSe concentrations and HIX and humic-like CDOM components (Fig. 4d, 4e) indicating that DOSe may be associated with dissolved humic substances. In addition, DOSe correlated with $S_{275\text{-}295}$ and $SUVA_{254}$ (Fig. 5a, 5c) suggesting that DOSe was associated closely with high-molecular-weight and highly aromatic DOM. Also, the positive correlations between DOSe and the humic-like C3 component (Fig. 5b) which derived corresponded to aromatic and black carbon compounds with high molecular weight, also indicates that DOSe fractions are associated with high-molecular-weight aromatic DOM (Fig. 6). Pokrovsky et al. (2018) also found that Se were transport in the form of high molecular weights organic aromatic-rich complexes from peat to the rivers and lakes in the Arctic. Bruggeman et al. (2007) and Kamei-Ishikawa et al. (2008) both found that 50% to 70% of Se(IV)–humic substances associates had high molecular weights (>10 kDa), that consistent with our findings.

During the estuarine mixing, the negatively correlation between DOSe/DOC and DOSe/DISe ratios with C2/C1 ratios which is enhanced by photodegradation (Wang et al., 2019; Fig. 5d, 5e), indicating that compared to bulk DOM, the DOSe fractions were more susceptible to photodegradation, and that DOSe was probably photodegraded to DISe. As suggested by Martin et al (2018) that most photochemical transformations of DOM in Sarawak likely take place after DOM reaches the sea. Thus, photodegradation plays an important role in DOSe processing once transported to offshore, and DOSe might contain a significant photoreactive fraction that facilitates photodegradation of DOSe into lower mean molecular weights or gaseous Se or photomineralization to DISe (Fig. 6). Considerable amounts of Se may be volatilized when methylselenide compounds form (Lidman et al., 2011). A field study found that volatile species of Se were naturally emitted from peatland at concentrations of around 33 nmol L^{-1} (Vriens et al., 2015). As a result of the method used in the present study, volatile methylselenide compounds in the DOSe fractions may not have been detected, so DOSe may have been underestimated. In future work, particular attention should be given to methylselenide. Studies have shown that photodegradation of DOM results in a range of bioavailable products (Miller and Moran, 1997). Peatland-derived DOSe might be degraded to a lower molecular weight or DISe in the coastal areas, both of which are bioavailable for phytoplankton and may stimulate their growth, and thereby impact the marine animals via food chain. The photoreactive DOSe fractions are probably transported across the marginal sea and circulated globally. Given that the bioavailability and biogeochemical cycling of the peatland-derived DOSe fractions may differ from those of peptides produced *in situ* by phytoplankton in the ocean, the impact on coastal and open ocean ecosystems should be evaluated in the future.”

Comment 10: L621-625. This conclusion is true, however it is based on very indirect observations (many parameters are from already published works). Note that the main source of Se in peatland waters as from highly aromatic DOM of peat horizons has been recently evidenced in Siberian lakes (Pokrovsky et al., 2018 Env Sci Technol)

Response: Thanks for the great advice. We have learned a lot from the literature (Pokrovsky et al., 2018 Env Sci Technol), and cited in the manuscripts, as following:

"In the high-latitude peatland-draining rivers, dissolved Se concentrations are spatial variable, with concentrations of up to 13 nmol L^{-1} being observed in northern Minnesota, US (Clausen and Brooks, 1983), from 0.38 to 5 nmol L^{-1} in the Krycklan catchment, Sweden (Lidman et al., 2011) and from 0.25 to 1.25 nmol L^{-1} in the Siberian (Pokrovsky et al., 2018)."

"the positive correlations between DOSe and the humic-like C3 component (Fig. 5b) which derived corresponded to aromatic and black carbon compounds with high molecular weight, also indicates that DOSe fractions are associated with high-molecular-weight aromatic DOM (Fig. 6). Pokrovsky et al. (2018) also found that Se were transport in the form of high molecular weights organic aromatic-rich complexes from peat to the rivers and lakes in the Arctic."

Comment 11: Fig 2: How representative is Rajang to other rivers, why it is shown?

Response: The Se distribution in the peatland draining estuaries is largely unknown. Compared with other rivers, Rajang is the longest river in Malaysia, and the delta plain is mainly composed by organic matter enriched sediments which was identified as peat deposits with a maximum depth of 15 m (Staub et al., 2000). Considering the space of the manuscript, Fig. 2 were moved to

Supplement.

Comment 12: Fig 3 is fine Fig 4 might not be needed - may be in Supplement? Previous Fig 3 is way more informative. Fig.4 should be shortened, at least.

Response: Fig. 4 were moved to supplement, Fig 3 were kept.

Comment 13: Fig.6: what is the difference with fig 3? (hard to apprehend) Fig 6: The size of panels is too small, please enlarge

Response: Fig.6 is the laboratory mixing experiments that simulated estuarine mixing processes. Fig. 3 is the results of field observations. Considering the incompleteness of the mixing experiments, Fig.6 was deleted.

Comment 14: Fig. 8: The plots showing no relationships between variables are not needed to be shown; it is enough just to state that there is no link between variables.

Response: We have deleted the Fig.8 a – e but kept Fig 8. f – k as Fig.5 in the revised manuscripts.

General comment: The authors could present the fluxes of Se to the ocean, in different forms. The yield from watersheds of different rivers (i.e., in kg/km2/y) could be compared with that of other large and small rivers of the world, if the data are available. How important are small rivers of Borneo on a global scale of DISe and Dose delivery to the ocean? Are the yields disproportionally high? Conclusions nicely reflect the main findings, and even if some of them are speculative (L 675-678), they can be stated as they are.

Response: We have added the “4.3 TDSe flux” section in the manuscripts with estimation of the riverine TDSe flux in Table R2 (i.e. Table 2 in the manuscript).

The TDSe flux was estimated to be 16 × 10^3 and 0.044 × 10^3 kg yr^{-1} for the Rajang and Maludam, respectively (Table R2). On a global scale, the TDSe delivered from Rajang were less than those large rivers including Changjiang, Amozon, Zhujiang, Orinoco and St.Lawrence River, but exceeded other small rivers reported so far (Table R2). The TDSe delivered by Rajang and Maludam contributed nearly 1% of the total riverine TDSe input to the ocean with only 0.3% of freshwater discharge (Nriagu, 1989; Milliman and Farnsworth, 2013). The TDSe yields for the Rajang and Maludam were just below large river Changjiang and the polluted Scheldt River, but were exceed the other rivers (Table R2). As for the DOSe yields for the Rajang and Maludam were one or even two orders of magnitude higher than other reported rivers so far (Table R2). This indicates that the numerous small blackwater rivers draining from peatland are very efficient TDSe and DOSe sources for the coastal waters. The roughly estimated TDSe flux from tropical peatland (439,238 km^2, Page et al., 2011) could be roughly around 120× 10^3 kg yr^{-1}, which were nearly 35% of the current total riverine TDSe flux, based on average TDSe yield from Rajang and the Maludam (0.27 kg km^{-2} yr^{-1}). On a global perspective, TDSe export from peat-draining rivers is quantitatively more significant than previously thought. It can be expected that increasing anthropogenic disturbing of peat can release a great amount of Se to rivers, and then transported to the coastal areas, the impact to the ecosystem should receive more attention in future studies.

Table R2 Overview of the TDSe concentrations and DOSe/TDSe ratios in the river and the magnitude of riverine TDSe flux and yield to the ocean.

River Name	TDSe (nmol L^{-1})	DOSe/TDSe Ratio	TDSe flux [a] (10^3 kg yr^{-1})	TDSe yield [a] (kg km^{-2} yr^{-1})	DOSe yield [a] (kg km^{-2} yr^{-1})	Reference
Rajang (Malysia)	1.76	0.90	16	0.32	0.28	This study
Maludam (Malysia)	4.04	0.99	0.044	0.22	0.22	This study
Amazon (Brazil)	0.48	0.85	250	0.041	0.035	Cutter and Cutter, 2001
Changjiang (China)	4.59 [b]	n.a. [c]	652	0.72	n.a. [c]	Chang et al., 2016
Zhujiang (China)	4.87 [b]	n.a. [c]	100	0.20	n.a. [c]	Yao et al., 2006
Orinoco (Venezuela)	0.45	n.a. [c]	39	0.036	n.a. [c]	Yee et al., 1987
St.Lawrence (Canada)	2.12	0.11	57	0.047	0.0051	Takayanagi and Wong, 1985
Rhone (France)	2.18	0.14	9.3	0.10	0.013	Guan and Martin, 1991
James river (America)	2.08	0.40	1.4	0.020	0.008	Takayanagi and Wong, 1983; 1984
Sacramento (America)	0.91	0.38	1.2	0.023	0.009	Cutter and Cutter, 2004
San Joaquin (America)	15.8	0.23	5.0	0.060	0.014	Cutter and Cutter, 2004
Jiulongjiang (China)	2.44	0.21	1.6	0.11	0.022	Hu et al., 1995
Kaoping (China)	1.19	0.47	0.26	0.081	0.038	Hung and Shy, 1995;
Erhjen (China)	1.11	0.47	0.044	0.13	0.059	Hung and Shy, 1995;
Shinano (Japan)	0.50	<0.1	0.55	0.046	0.006	Suzuki et al., 1981
Scheldt (Belgium)	29.2 [b]	n.a. [c]	13.83	0.63	n.a. [c]	Van der Sloot et al., 1985

[a] The calculation used river basin areas and discharge rate were cited from Milliman and Farnsworth, 2013

[b] The data were DISe species.

[c] The DOSe were not measured

Reference

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