

Response to review

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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. We have deleted those sentences in the revised manuscripts.

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 1. However, the slope of the rivers was not available.

Revised manuscripts: “The physio-geographical parameters of sampled river basins are summarized in Table 1” were added in *page 6 line 134-135*. Table 1 were added in *page 25 line 711-725*, as followed.

29 Table 1 The physio-geographical parameters of sampled rivers. (n.a. stands for not
30 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

31 ^a Modified from Bange et al., 2019

32 ^b Cited from Staub et al., 2000

33 ^c Cited Müller et al., 2016

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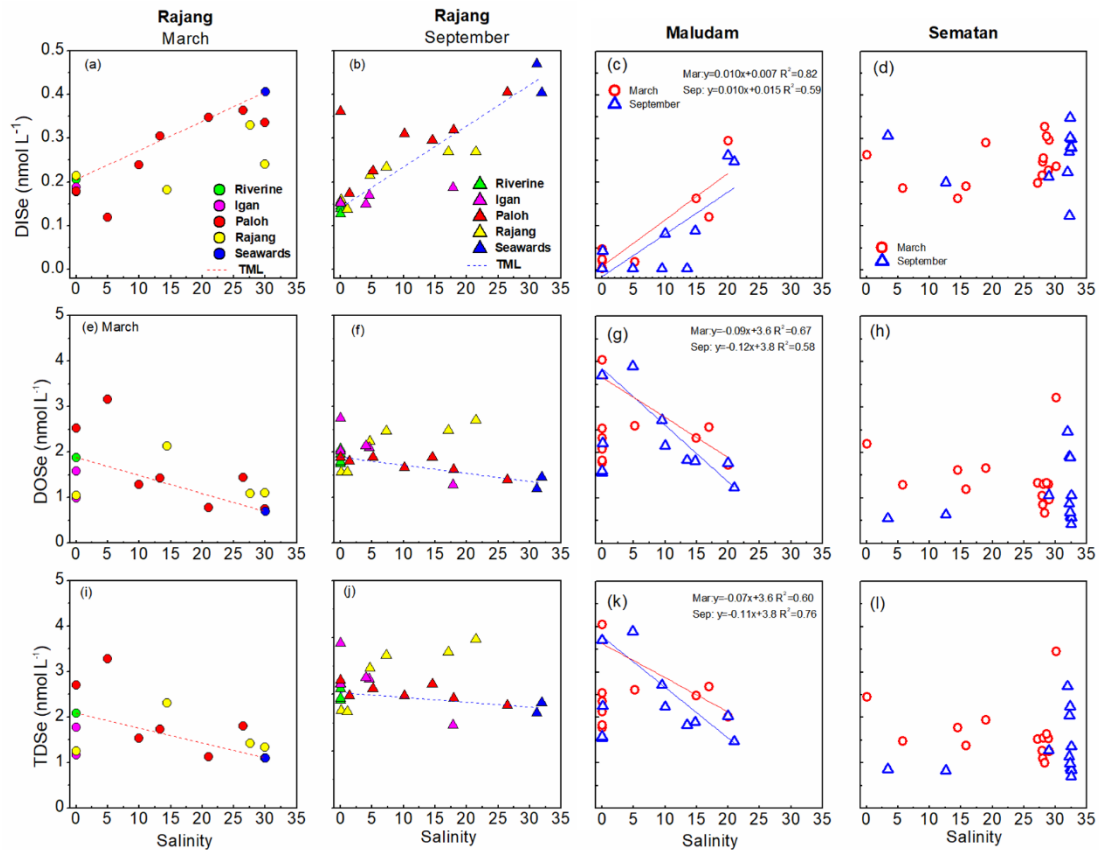
35 **Comment 3.** Three type of Se behavior are well identified and summarized
36 in L349-358. However, the presentation of each individual river in Figs 2-5
37 takes too much space. Either consider presenting just an example of each
38 group or the average of all rivers in each group

39 **Response:** Thanks for the great advice. We have moved Fig. 2 and Fig.
40 4 to the supplement, and Fig. 3 and Fig. 5 were merged to one figure (Fig. 3
41 in the revised manuscript). Three typical groups of relationships between Se
42 species and salinity for Rajang, Maludam and Sematan estuaries were
43 selected to present in Fig. 3 in the revised manuscript, and those for Sebuyau
44 and Samunsam were moved to supplement.

45 **Revised manuscripts:** Figure 3 were present in *page 29 line 738-743*,
46 as followed.

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50 Fig. 3. Relationships between DISe (a - d), DOSe (e - h), and TDSe (i - l) concentrations
 51 with salinity in the Rajang and three Rajang tributaries (Igan, Lassa, and Rajang), and in
 52 the Maludam and Sematan estuaries in March and September 2017. TML refers to the
 53 theoretical mixing line, which was defined using two endmembers: freshwater in the
 54 riverine system and seawater.

55

56 **Comment 4:** L407-412: Please explain, what is the mechanism of

57 Se(IV)/Se(VI) increase with DO increase. Oxidation is more pronounced at
 58 high DO, yet the observations are reverse to that.

59 **Response:** As shown in Fig. R1, Se(IV)/Se(VI) ratios in the freshwater of
 60 the sampled rivers increased as DO concentrations increased statistically.

61 However, the Se(IV)/Se(VI) ratios were calculated only if Se(IV) and Se(VI)

62 concentrations were both above the detection limits, meaning that the data

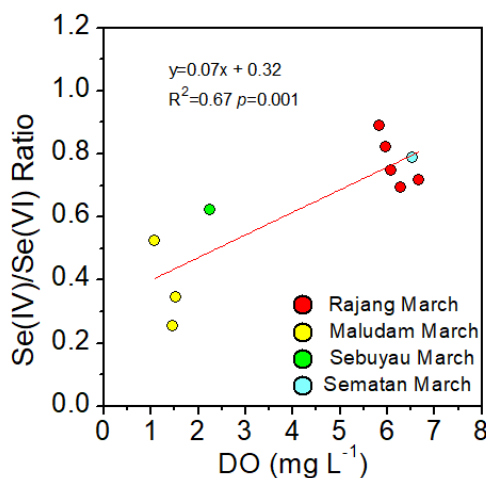
63 are limited. The limited Se(IV)/Se(VI) ratios roughly fell into two groups, one

64 group was low Se(IV)/Se(VI) ratios with low DO concentration and the other

65 was high Se(IV)/Se(VI) ratios with high DO concentration. The liner

66 relationship between Se(IV)/Se(VI) ratios and DO concentration probably was

67 false appearance with the limited data. We have deleted this figure in the
68 manuscript.



69

70 Fig. R1 Relationships between Se(IV)/Se(VI) ratios and DO values n freshwater (Salinity
71 < 1) for the Rajang, Sematan, Maludam, Sebuyau, Samunsam, and Simunjan rivers in
72 March and September. Se(IV)/Se(VI) ratios were calculated only if Se(IV) and Se(VI)
73 concentrations were both above the detection limits, meaning that the data are limited.

74

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

77 **Response:** We have deleted those sentences, table 1 with river
78 watershed parameters were presented as shown in response 1.

79

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

82 **Response:** Thanks for the advices, we have deleted those sentences,
83 including “However, the mechanisms behind the interactions between Se and
84 dissolved organic ligands are still poorly understood. Three hypotheses have
85 been proposed to explain organic-matter-mediated retention of Se, as follows:
86 1) direct complexation of organic matter with Se, 2) indirect complexation via
87 Se-cation–organic-matter complexes, or 3) microbial reduction and
88 incorporation into amino acids, proteins, and natural organic matter (Winkel et
89 al., 2015). Depending on the type of binding, Se may be easily mobilized (e.g.,

90 through adjusting pH) or immobilized (e.g., by covalent incorporation to organic
 91 matter) (Winkel et al., 2015). However, there is ambiguity about the molecular
 92 structure and species of Se that bind to organic matter, and further work is
 93 needed to identify the mechanisms by which Se is bound to, and released from,
 94 organic matter.” in the revised manuscript.

95

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

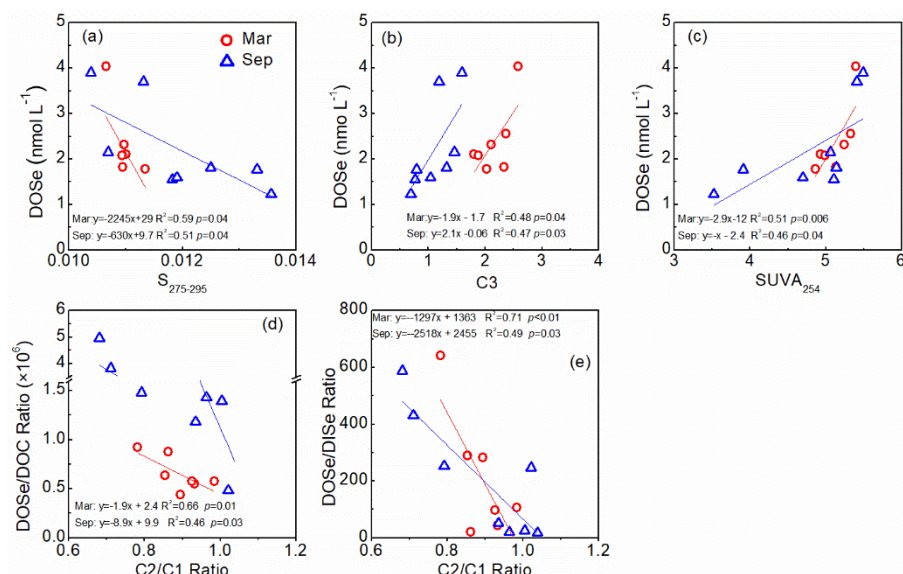
98 **Response:** We have deleted the “Nitrate behaves in a similar way in the
 99 Dumai River estuary (Sumatra, Indonesia), another tropical blackwater river
 100 (Alkhaitb and Jennerjahn, 2007).”

101

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

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

107 **Revised manuscripts:** Figure 5 were present in *page 31 line 757-762*,
 108 as followed.



109

110 Figure 5. Relationships between DOSe concentrations and S275-295, C3
111 components and SUVA₂₅₄, DOSe/DOC ratio and C2/C1 component ratios,
112 and DOSe/DISe ratios and C2/C1 component ratios in the Rajang and
113 Maludam estuaries. The S275-295, SUVA₂₅₄, C1, C2, and C3 components
114 are from Martin et al. (2018) and Zhou et al. (2019) from the same cruises.

115

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

119 **Response:** We have deleted those specific literature details and shorted
120 this section from 63 lines to 41 lines.

121 **Revised manuscripts:** In page 15-16 line 373-414, as followed:

122 “Moreover the peat-draining rivers demonstrated a liner relationship between
123 DOSe concentrations and HIX and humic-like CDOM components (Fig. 4d, 4e)
124 indicating that DOSe may be associated with dissolved humic substances. In
125 addition, DOSe correlated with S₂₇₅₋₂₉₅ and SUVA₂₅₄ (Fig. 5a, 5c) suggesting
126 that DOSe was associated closely with high-molecular-weight and highly
127 aromatic DOM. Also, the positive correlations between DOSe and the humic-
128 like C3 component (Fig. 5b) which derived corresponded to aromatic and black
129 carbon compounds with high molecular weight, also indicates that DOSe
130 fractions are associated with high-molecular-weight aromatic DOM (Fig. 6).
131 Pokrovsky et al. (2018) also found that Se were transport in the form of high
132 molecular weights organic aromatic-rich complexes from peat to the rivers and
133 lakes in the Arctic. Bruggeman et al. (2007) and Kamei-Ishikawa et al. (2008)
134 both found that 50% to 70% of Se(IV)–humic substances associates had high
135 molecular weights (>10 kDa), that consistent with our findings.

136 During the estuarine mixing, the negatively correlation between
137 DOSe/DOC and DOSe/DISe ratios with C2/C1 ratios which is enhanced by
138 photodegradation (Wang et al., 2019; Fig. 5d, 5e), indicating that compared to
139 bulk DOM, the DOSe fractions were more susceptible to photodegradation, and

140 that DOSe was probably photodegraded to DISe. As suggested by Martin et al
141 (2018) that most photochemical transformations of DOM in Sarawak likely take
142 place after DOM reaches the sea. Thus, photodegradation plays an important
143 role in DOSe processing once transported to offshore, and DOSe might contain
144 a significant photoreactive fraction that facilitates photodegradation of DOSe
145 into lower mean molecular weights or gaseous Se or photomineralization to
146 DISe (Fig. 6). Considerable amounts of Se may be volatilized when
147 methylselenide compounds form (Lidman et al., 2011). A field study found that
148 volatile species of Se were naturally emitted from peatland at concentrations of
149 around 33 nmol L⁻¹ (Vriens et al., 2015). As a result of the method used in the
150 present study, volatile methylselenide compounds in the DOSe fractions may
151 not have been detected, so DOSe may have been underestimated. In future
152 work, particular attention should be given to methylselenide. Studies have
153 shown that photodegradation of DOM results in a range of bioavailable products
154 (Miller and Moran, 1997). Peatland-derived DOSe might be degraded to a lower
155 molecular weight or DISe in the coastal areas, both of which are bioavailable
156 for phytoplankton and may stimulate their growth, and thereby impact the
157 marine animals via food chain. The photoreactive DOSe fractions are probably
158 transported across the marginal sea and circulated globally. Given that the
159 bioavailability and biogeochemical cycling of the peatland-derived DOSe
160 fractions may differ from those of peptides produced *in situ* by phytoplankton in
161 the ocean, the impact on coastal and open ocean ecosystems should be
162 evaluated in the future.”

163

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

168 (Pokrovsky et al., 2018 Env Sci Technol)

169 **Response:** Thanks for the great advice. We have learned a lot from the
170 literature (Pokrovsky et al., 2018 Env Sci Technol), and also cited in the
171 manuscripts. The investigation of biogeochemical process in the peat-draining
172 rivers and estuaries in Borneo were international cooperation, the CDOM
173 investigation (Martin et al., 2018; Zhou et al., 2019) cited in our manuscript
174 were conducted by our cooperative partners.

175 **Revised manuscripts:** In *page 4 line 84-88*, and *page 15 line 378-387*
176 as followed:

177 “In the high-latitude peatland-draining rivers, dissolved Se concentrations
178 are spatial variable, with concentrations of up to 13 nmol L⁻¹ being observed
179 in northern Minnesota, US (Clausen and Brooks, 1983), from 0.38 to 5 nmol
180 L⁻¹ in the Krycklan catchment, Sweden (Lidman et al., 2011) and from 0.25 to
181 1.25 nmol L⁻¹ in the Siberian (Pokrovsky et al., 2018).”

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

189

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

192 **Response:** The Se distribution in the peatland draining estuaries is
193 largely unknown. Compared with other rivers, Rajang is the longest river in
194 Malaysia, and the delta plain is mainly composed by organic matter enriched
195 sediments which was identified as peat deposits with a maximum depth of 15

196 m (Staub et al., 2000). Considering the space of the manuscript, Fig. 2 were
197 moved to Supplement.

198

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

202 **Response:** Fig. 4 were moved to supplement; Fig 3 were kept.

203

204 **Comment 13:** Fig.6: what is the difference with fig 3? (hard to apprehend) Fig
205 6: The size of panels is too small, please enlarge

206 **Response:** Fig.6 is the laboratory mixing experiments that simulated
207 estuarine mixing processes. Fig. 3 is the results of field observations.

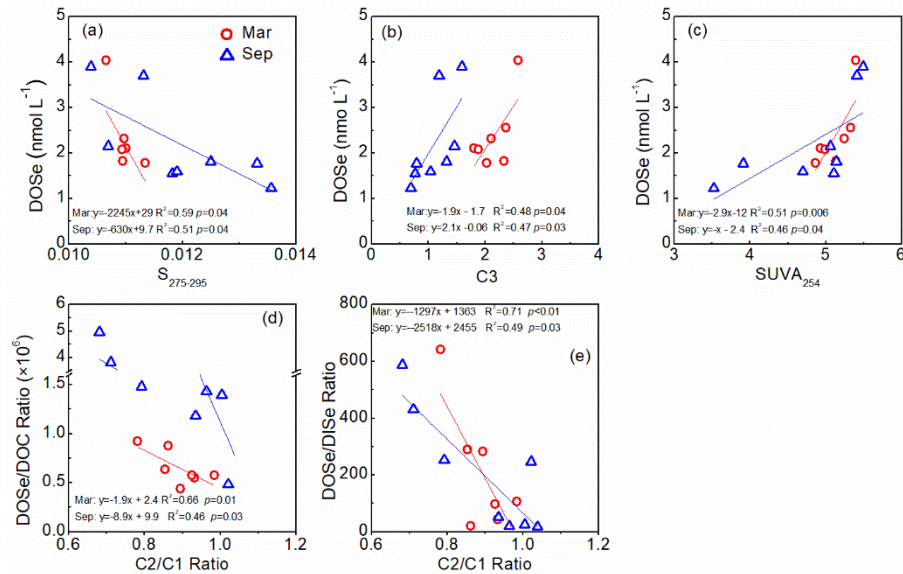
208 Considering the incompleteness of the mixing experiments, Fig.6 was
209 deleted.

210

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

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

216 **Revised manuscripts:** In page 31 line 751, as followed:



217

218 Figure 5. Relationships between DOSe concentrations and S₂₇₅₋₂₉₅, C3 components and
 219 SUVA₂₅₄, DOSe/DOC ratio and C2/C1 component ratios, and DOSe/DISe ratios and
 220 C2/C1 component ratios in the Rajang and Maludam estuaries. The S₂₇₅₋₂₉₅, SUVA₂₅₄,
 221 C1, C2, and C3 components are from Martin et al. (2018) and Zhou et al. (2019) from the
 222 same cruises.

223

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

231

Response: We have added the “4.3 TDSe flux” section in the
 232 manuscripts with estimation of the riverine TDSe flux in Table 2 in the
 233 manuscript.

234

The TDSe flux was estimated to be 16×10^3 and 0.044×10^3 kg yr⁻¹ for the
 235 Rajang and Maludam, respectively (Table 2). On a global scale, the TDSe
 236 delivered from Rajang were less than those large rivers including Changjiang,
 237 Amazon, Zhujiang, Orinoco and St. Lawrence River, but exceeded other small
 238 rivers reported so far (Table 2). The TDSe delivered by Rajang and Maludam

239 contributed nearly 1% of the total riverine TDS_e input to the ocean with only
240 0.3% of freshwater discharge (Nriagu, 1989; Milliman and Farnsworth, 2013).
241 The TDS_e yields for the Rajang and Maludam were just below large river
242 Changjiang and the polluted Scheldt River, but were exceeded the other rivers
243 (Table R2). As for the DOSe yields for the Rajang and Maludam were one or
244 even two orders of magnitude higher than other reported rivers so far (Table
245 R2). This indicates that the numerous small blackwater rivers draining from
246 peatland are very efficient TDS_e and DOSe sources for the coastal waters. The
247 roughly estimated TDS_e flux from tropical peatland (439,238 km², Page et al.,
248 2011) could be roughly around 120 × 10³ kg yr⁻¹, which were nearly 35% of the
249 current total riverine TDS_e flux, based on average TDS_e yield from Rajang and
250 the Maludam (0.27 kg km⁻² yr⁻¹). On a global perspective, TDS_e export from
251 peat-draining rivers is quantitatively more significant than previously thought. It
252 can be expected that increasing anthropogenic disturbing of peat can release
253 a great amount of Se to rivers, and then transported to the coastal areas, the
254 impact to the ecosystem should receive more attention in future studies.

255 **Revised manuscripts:** In page 16-17 line 416-438, page 26 line 716-
256 720, as followed:

257 4.3 TDS_e flux

258 Information on the biogeochemistry of peat-draining rivers is scarce, and so
259 their possible quantitative significance for the oceanic TDS_e budget is
260 unexplored as yet. The TDS_e flux was estimated to be 16 × 10³ and 0.044 ×
261 10³ kg yr⁻¹ for Rajang and Maludam, respectively (Table 2). On a global scale,
262 the TDS_e delivered from Rajang were less than those large rivers including
263 Changjiang, Amazon, Zhujiang, Orinoco and St. Lawrence River, but exceeded
264 other small rivers reported so far (Table 2). The TDS_e delivered by Rajang and
265 Maludam contributed nearly 1% of the total riverine TDS_e input to the ocean
266 with only 0.3% of freshwater discharge (Nriagu, 1989; Milliman and Farnsworth,

267 2013). The TDSe yields for Rajang and Maludam were just below the second
268 largest river Changjiang and the polluted Scheldt River, but were exceeded the
269 other rivers (Table 2). As for the DOSe yields for Rajang and Maludam were
270 one or even two orders of magnitude higher than other reported rivers so far
271 (Table 2). This indicates that the numerous small blackwater rivers draining
272 from peatland are very efficient TDSe and DOSe sources for the coastal waters.
273 The roughly estimated TDSe flux from tropical peatland (439,238 km², Page et
274 al., 2011) could be roughly around $120 \times 10^3 \text{ kg yr}^{-1}$, which were nearly 35% of
275 the current total riverine TDSe flux, based on average TDSe yield from Rajang
276 and the Maludam ($0.27 \text{ kg km}^{-2} \text{ yr}^{-1}$). On a global perspective, TDSe export
277 from peat-draining rivers is quantitatively more significant than previously
278 thought. It can be expected that increasing anthropogenic disturbing of peat
279 can release a great amount of Se to rivers, and then transported to the coastal
280 areas, the impact to the ecosystem should receive more attention in future
281 studies.

282

283

284 Table 2 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 (Malaysia)	1.76	0.90	16	0.32	0.28	This study
Maludam (Malaysia)	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

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

286 ^b The data were DISe species.

287 ^c The DOSe were not measured

Reference

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302 the Gulf of Lions, *Marine Chemistry*, 36, 303-316, 1991.
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304 Erhjen Rivers and Estuaries, southwestern Taiwan, *Estuaries*, 18, 234-240,
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334 estuaries and the Southern Bight (North Sea), *Estuarine Coastal & Shelf*
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340 Orinoco in Venezuela, *Nature*, 326, 686, 1987.
- 341

Response to review

342

343

344 **Respect Anonymous Referee #2**

345 We want to begin by thanking Referee #2 for writing that “I think that the
346 paper adds novel information to our knowledge of the Se cycle.” We are
347 extremely grateful for his/her insightful advice and elaborate revisions of the
348 manuscript. We addressed all the points raised by the referee, as summarized
349 below.

350

351 **General Comments:**

352 1. Overall, I think that the paper adds novel information to our knowledge of
353 the Se cycle. However, it has largely a descriptive character, which could be
354 changed by coming up with a number of hypotheses that can be tested with
355 the data.

356 **Response:** Thanks for the great advice. We have carefully revised the
357 manuscripts based on the comments.

358 We have come up three hypotheses, 1) the DOSe is the major species in
359 those peatland- draining rivers; 2) the source of DOSe probably is peat soils
360 and 3) large amounts of TDSe from peatland-draining rivers were delivered to
361 the coastal water. Those hypotheses were tested with data, as shown in
362 results 3.2 and discussion, DOSe/TDSe ratios ranged from 0.56 to 0.99,
363 indicating that DOSe was the major species of Se in the peat-draining rivers
364 and estuaries (Fig. 2). The relationship between DOSe and HIX, humic-like
365 CDOM components, $S_{275-295}$ and $SUVA_{254}$ (Fig. 4, Fig 5) indicating that peat
366 soils is inferred to be the major source of DOSe in our sampled rivers and
367 DOSe may be associated with with high-molecular-weight and highly humic
368 substances. These was demonstrated in results 3.4 and discussion 4.2. The
369 TDSe flux was estimated in the discussion 4.3. The results showed that TDSe

370 delivered from Rajang were less than those large rivers, but exceeded other
371 small rivers reported so far (Table 2). As for DOSe yields for Rajang and
372 Maludam were one or even two orders of magnitude higher than other
373 reported rivers so far (Table 2). This indicates that the numerous small
374 blackwater rivers draining from peatland are very efficient TDSe and DOSe
375 sources for the coastal waters.

376 **Revised manuscripts:** In page 5 line 108-111, as followed:

377 “We hypothesize that the DOSe is the major species in those peatland-
378 draining rivers which mainly from peat soils and sizable Se from peatland is
379 delivered to the coastal areas.

380

381 2. Moreover, the original results are shown in too much detail, which
382 obscures the general findings. I suggest to come up with figures that combine
383 the results from several or all studied locations and move the original data to
384 the supplementary information.

385 **Response:** We have deleted some details of the results and simplified
386 the manuscripts. The result 3.1 to 3.3 were down sized from 145 to 84 lines
387 for now, and the figures were downsized from nine to six by moving two
388 original figures (Fig. 2 and 4) to supplementary, and we also have combed
389 two original figures (Fig. 3 and 5) to one figure by presenting the three typical
390 salinity-concentrations relationships (detailed response was shown in
391 comment 17).

392

393 3. As already indicated in my preliminary review, the paper suffers from a
394 number (of minor) technical problems. It should be strictly structured
395 according to the objectives, which is not the case in the introduction where the
396 state-of-the-art concerning Objective 3 is not introduced. It is also not the case
397 in the discussion and conclusion sections.

398 **Response:** We have revised the objectives and reorganized the
399 discussions according to the objectives. The three objectives were 1) evaluate
400 the fate of Se species during estuarine mixing in peatland-draining estuaries;
401 2) characterize the DOSe fractions; and 3) estimate the magnitude of Se
402 fluxes delivered from peatland-draining rivers to coastal ocean. The objective
403 2 (i.e. the original objective 3) is about the character of DOSe, which were
404 added in the introduction (Lines 94 to 104, detailed response was shown in
405 comment 11). The discussion sections were structured to three parts
406 according to the objectives, as following: 4.1) Fate of Se species during
407 estuarine mixing; 4.2) Character of the DOSe fractions; and 4.3) TDOSe flux.
408 The conclusion also reorganized to fit to the three objectives and an outlook,
409 details were shown in response 31.

410 **Revised manuscripts:** In *page 5 line 111-114*, as followed:

411 The main objectives of the study were to 1) evaluate the fate of dissolved
412 Se species in peatland-draining estuaries; 2) characterize the DOSe fractions;
413 and 3) estimate the magnitude of Se fluxes delivered from to coastal ocean.

414

415 4. The discussion sections includes results referring to Figs. 7 and 8, which
416 need to be moved to the results section.

417 **Response:** We have moved Figs. 7 and 8 to the results in section “3.4
418 Correlation between Se species with DO, pH and DOM”, detail was shown in
419 response 19.

420

421 5. Finally, the manuscript should be shortened, e.g., by moving all content
422 that is mainly of local interest to the supplementary information.

423 **Response:** The content that is mainly of local interest was moved to the
424 supplementary information and the original detailed results was deleted. We
425 have shorted the manuscripts from 1025 lines to 768 lines.

426 **Comment 1.** l. 25 and l. 52: Please be clear about which organisms really
427 need Se. I know that mammals and humans need it. At the same time plants
428 do not need Se. I am not familiar with marine organisms. Please specify,
429 which marine organisms need Se. I would not have expected that Se is
430 essential for phytoplankton (because it is not for plants). This question is
431 important and should be clearly addressed.

432 **Comment** l. 54-67: About which organisms are you talking? This cannot be
433 generalized!

434 **Comment** l.57-59: Please explain this hypothesis. Its understanding is related
435 with my above criticism, that there is no detailed explanation for which
436 organisms Se is necessary and for which not.

437 **Comment** l. 68: Does phytoplankton really need Se?

438 **Response:** Similar comments were responded together.

439 Se is required for biosynthesis of selenocysteine, the twenty first naturally
440 occurring amino acid in protein (Lobanov et al., 2009). As reviewed by
441 Lobanov et al (2009), selenoproteins show a mosaic occurrence, with many
442 organisms, such as vertebrates and algae, having dozens of these proteins,
443 while other organisms, such as higher plants and fungi, having lost all
444 selenoproteins during evolution.

445 Selenium is an essential microelement for all aquatic organisms-
446 microorganisms, algae, higher aquatic plants and animals (Bodnar et al,
447 2014). In photosynthetic microorganisms, the essential requirement for
448 selenium has been reported in 33 species belonging to six phyla (Table R1,
449 Araie and Shiraiw, 2016). Price and Harrison (1988) found selenoproteins
450 compounds (GSH-Px) in *Thalassiosira pseudonana* and confirmed obligate
451 requirement for Se in marine diatom. When Se was added to the culture
452 medium, growth was stimulated in the diatom *Thalassiosira pseudonana*,
453 *Chysochromulina breviturrita* in Haptophyceae, the dinoflagellates

454 *Gymnodinium catenatum* and *Alexandrium minutum*, and other algae (Table
 455 R1, Araie and Shiraiw, 2016). Studies showed that diatom (*Thalassiosira*
 456 *pseudonana*, *Chaetoceros*) cultures deprived of Se(IV) in seawater for more
 457 than 5 days did not recover even when Se(IV) was added afterwards
 458 (Harrison et al., 1988). The study concluded that it was more difficult for these
 459 Se-dependent microorganisms to recover after exposure to Se depletion than
 460 from exposure to nitrogen or phosphorus limitation. Similar results are found
 461 in Doblin et al. (1999) where three marine phytoplankton species
 462 (*Gymnodinium catenatum*, *Alexandrium minutum*, *Chaetoceros cf.*
 463 *tenuissimus*) showed rapid decline in Se deficient seawater, resulting in
 464 cessation of cell division after eight weeks of Se(IV) depletion. However,
 465 studies on the effect of selenite in the unicellular green alga *Chlamydomonas*
 466 *reinhardtii*, showed only a little simulative effect on growth (Novoselov et al;
 467 2002). Marine phytoplankton show a stronger trend to a preference for Se
 468 than freshwater phytoplankton do (Araie and Shiraiw, 2016).

469 Geochemical analyses of trace elements in Phanerozoic marine pyrite
 470 that sustained periods of severe Se depletion in the past oceans correlate
 471 closely with three major mass extinction events, at the end of the Ordovician,
 472 Devonian and Triassic periods (Long et al., 2016). Considering the essential
 473 of Se for marine phytoplankton, the authors assumed that Se depletion may
 474 have been one of several factors in these complex extinction scenarios (Long
 475 et al., 2016).

476 **Table R1.** Phytoplankton species that were demonstrated to require selenium for their
 477 growth. ^a

Phylum	Species
Diatoms	<i>Amphiprora hyalina</i>
	<i>Chaetoceros debilis</i>
	<i>Chaetoceros pelagicus</i>
	<i>Chaetoceros vixvisibilis</i>
	<i>Coscinodiscus asteromphalus</i>
	<i>Corethron criophilum</i>

	<i>Ditylum brightwellii</i>
	<i>Skeletonema costatum</i> (strain 18c NEPCC)
	<i>Skeletonema costatum</i> (strain 611 NEPCC)
	<i>Skeletonema costatum</i> (strain 616 NEPCC)
	<i>Stephanopyxis palmeriana</i>
	<i>Thalassiosira pseudonana</i>
	<i>Thalassiosira oceanica</i>
	<i>Thalassiosira rotula</i>
	<i>Thalassiosira aestivalis</i>
Dinoflagellates	<i>Alexandrium minutum</i> ^b
	<i>Gymnodinium catenatum</i> ^b
	<i>Gymnodinium nagasakiense</i> ^b
	<i>Peridinium cinctum</i> fa. <i>Westii</i>
	<i>Pyrodinium bahamense</i> ^b
Prymnesiophytes	<i>Chrysochromulina breviturrita</i>
	<i>Chrysochromulina kappa</i>
	<i>Chrysochromulina brevefilum</i>
	<i>Chrysochromulina strobilus</i>
	<i>Chrysochromulina polylepis</i> ^b
	<i>Helladosphaera</i> sp
	<i>Emiliana huxleyi</i>
	<i>Gephyrocapsa oceanica</i>
Raphidophytes	<i>Chattonella verruculosa</i> ^b
Chlorophytes	<i>Platymonas subcordiformis</i>
Chrysophytes	<i>Aureococcus anophagefferens</i> ^b

^a Modified from Araie and Shiraiw, 2016

^b Harmful algae.

478 **Revised manuscripts:**

479 In page 2 line 25, “Selenium (Se) is an essential micronutrient for aquatic
480 organisms”.

481 In page 3 line 52, “Se is an essential trace element for aquatic organisms
482 (Bodnar et al, 2014).”

483 Line 54-67 in the original manuscript “The range of beneficial effects of
484 Se is among the narrowest of all the elements and varies between dietary
485 deficiency (<40 µg d⁻¹) and toxicity (>400 µg d⁻¹) (Fernández-Martínez and

486 Charlet 2009; Schiavon et al., 2017).” were deleted.

487 In line 63, “marine” have been added before phytoplankton in the revised
488 manuscript, and changed to “A number of field and laboratory studies have
489 found that selenite [Se(IV)] and selenate [Se(VI)] can be assimilated by
490 marine phytoplankton with Se(IV) being the preferred species”

491

492 **Comment 2** l. 37: What do you mean by “extremely”? Add numbers.

493 **Response and revised manuscripts:** We have changed to “the
494 concentrations of DISE were extremely low (near or below the detection limits,
495 i.e. 0.0063 nmol L⁻¹)” in page 2 line 37-38.

496

497 **Comment 3** l. 47: I am not sure if the introduction of Se can generally
498 promote productivity. This would only be possible, if Se was an essential
499 nutrient for the considered organism. Furthermore, growth would only be
500 promoted if Se was the limiting element, but other limitations are more likely
501 (e.g., by P or Fe).

502 **Response:** The essential requirement for selenium has been reported in
503 33 species belonging to six phyla (Table R1, Araie and Shiraiw, 2016). The
504 dominant phytoplankton species was diatom in Sarawak coasts (Saifullah et
505 al., 2014). When Se was added to the culture medium, growth was stimulated
506 for diatom, and when it cultures deprived of Se(IV) in seawater for more than
507 5 days did not recover even when Se(IV) was added afterwards (Harrison et
508 al., 1988). Se-limiting for diatom growth were not found in the filed study,
509 although a study in Huon Estuary found that low level Se could be limiting for
510 growth and production of dinoflagellate (Doblin et al., 1999).

511 We are not sure whether Se could be a limiting element in the Malaysia
512 coastal waters, thus we have deleted those sentences.

513

514 **Comment 4** l. 48-49: I don't understand this conclusion. I would prefer a
515 conclusions, which is derived from your results.

516 **Response and revised manuscripts:** Thanks. We have deleted the "The
517 results of this study suggest that the impacts of Se discharges on coastal
518 ecosystems should be evaluated in the future", and have changed to "The TDSe
519 flux delivered by the peat-draining rivers exceeded other small rivers, and it is
520 quantitatively more significant than previously thought" in page 2 line 48-50.

521

522

523 **Comment 5.** l. 64: Why is "organic selenide" mentioned separately? It is
524 included in the oxidation state -II.

525 **Response and revised manuscripts:** We have deleted the "organic
526 selenide" in line 61.

527

528 **Comment 6** l. 88: What do you mean by "various", the previously cited
529 studies? Perhaps better cite them again.

530 **Response:** We have cited the previously mentioned studies again.

531 **Revised manuscripts:** In page 4 line 88-91:

532 "Although these various studies did not report different species of Se
533 (Clausen and Brooks, 1983; Lidman et al., 2011; Pokrovsky et al., 2018), the
534 DOSe probably the dominated species in peatland-draining river"

535

536 **Comment 7** l. 94: Do you mean that "Se speciation" was controlled?

537 **Response and revised manuscripts:** We have changed to "Chang et
538 al. (2016) found that Se speciation was controlled by biological, physical, and
539 redox processes in the estuaries" in page 3 line 74-75.

540

541 **Comment 8** l. 97: "formation" or "generation" instead of "regeneration

542 **Response and revised manuscripts:** We have changed to “generation
543 of particulate organic selenide in the water.” in page 3 line 77.

544

545 **Comment 9** l. 106: How does organic matter influence the bioavailability and
546 fate of Se?

547 **Response:** We have revised the introduction greatly, added the the
548 character of DOSe in the introduction (see response 11), and “It is also known
549 that organic matter plays an important role in the bioavailability and fate of Se
550 in the environment” in the original manuscripts was deleted.

551

552 **Comment 10** l. 110: What do you mean by “behaviour”?

553 **Response and revised manuscripts:** We have deleted “the behavior”,
554 changed to “More works of Se in fluvial systems in Southeast Asia are therefore
555 needed to provide an improved understanding of the biogeochemical
556 processing of Se and the associations with organic matter.” in page 4 line 103-
557 105.

558

559 **Comment 11** l. 122: The third objective “falls from heaven”.

560 **Response:** Thanks for the great advices. We have revised the
561 introduction, and added the DOSe research (i.e. objective 2 in the revised
562 manuscript) status.

563 **Revised manuscripts:** In page 4 line 84-105:

564 “In the high-latitude peatland-draining rivers, dissolved Se concentrations
565 are spatial variable, with concentrations of up to 13 nmol L⁻¹ being observed in
566 northern Minnesota, US (Clausen and Brooks, 1983), from 0.38 to 5 nmol L⁻¹
567 in the Krycklan catchment, Sweden (Lidman et al., 2011) and from 0.25 to 1.25
568 in the Siberian (Pokrovsky et al., 2018). Although these various studies did not
569 report different species of Se (Clausen and Brooks, 1983; Lidman et al., 2011;

570 Pokrovsky et al., 2018), the DOSe probably the dominated species in peatland-
571 draining river. In the open ocean, DOSe was assumed mainly associate with
572 soluble peptides with low molecule weight in surface waters and were relatively
573 refractory (Cutter and Cutter, 1995; 2004). Substantial amounts of dissolved Se
574 also are known to be associated humic substances, Gustafsson and Johnsson
575 (1994) assumed that Se was preferentially incorporated into low molecular
576 weight humic substances fractions by means of microbial reductive
577 incorporation, while Kamei-Ishikawa et al. (2008) found that Se associated with
578 high molecular weights humic acid fractions. The current paucity of information
579 on DOSe characteristics and its export by rivers from tropical peat-draining
580 rivers remains a major gap in our understanding of Se biogeochemical cycling.
581 Highest concentrations of dissolved organic carbon (DOC) globally were
582 reported in tropical peat-draining rivers in Borneo (Moore et al., 2013; Wit et al.,
583 2015). More works of Se in the fluvial systems of this region are therefore
584 needed to provide an improved understanding of the biogeochemical
585 processing of Se and the associations with organic matter.”

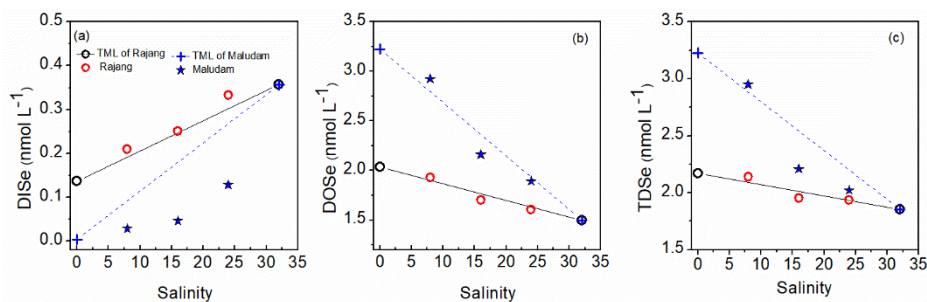
586

587 **Comment 12** l. 176: Why did you remove the colloids from the seawater
588 samples? Doesn't this result in a rather artificial experiment in which some
589 chemical transformations that can occur in the environment are ruled out?
590 Please explain. Furthermore, I suggest to come up with a hypothesis, e.g.,
591 pure mixing vs. chemical transformations (which?).

592 **Response:** Thanks for the advices. The research aim of this experiment
593 is to evaluate the impact of particle-free (i.e. dissolved phase) seawater and
594 river water mixing process on Se species, whether transformation of DISE
595 between DOSe occurs along salinity gradient. Here, the filter (0.45 micrometer
596 pore size) could retain a significant portion of colloids while only remove
597 particles.

598 The results were shown in Fig. R1. The measured DISe, DOSe and
 599 TDOSe concentrations were comparable with theoretical values, indicating pure
 600 mixing in Rajang estuary. However, in Maludam estuary, the measured DISe
 601 and TDOSe concentrations were lower than the theoretical values, while the
 602 measured DOSe concentrations were comparable with the theoretical value.
 603 The losses of DISe were not balanced by increasing in DOSe, indicating that
 604 chemical transformations between DISe and DOSe did not occur in Maludam.
 605 Other studies have reported removal of the humic fractions of DOM, colloidal
 606 iron, and phosphorus by flocculation in the river–sea mixing zones (Eckert and
 607 Sholkovitz, 1976; Forsgren et al., 1996; Asmala et al., 2014). Some of the
 608 DISe may exist in colloidal form in natural water (Takayanagi and Wong,
 609 1984), and DISe may be removed by flocculation. The removal of DISe were
 610 probably be flocculated to Se particulate form.

611 This mixing experiment indeed ruled out the impact of particle and part of
 612 colloids on the Se transformations. In the future, we would add another set of
 613 mixing experiment without filtration, particle-free and particle-included would
 614 be designed to the determine the influence of riverine particles and colloids on
 615 Se chemical cycling. Thus, considering the incompleteness, this section was
 616 deleted here.



617
 618 Fig. R1. Results of laboratory mixing experiments showing variation in DISe, DOSe, and
 619 TDOSe concentrations as a function of salinity using filtered riverine water from the Rajang
 620 and Maludam rivers and filtered coastal seawater. TML refers to theoretical mixing line.

621
 622

623 **Comment 13.** I. 210: Did you check for normal distribution and transform the

624 data if necessary?

625 **Response:** We have checked the normal distribution, and if the data
626 doesn't comply with the normal distribution, Mann Whitney U test were used
627 instead of t-test.

628 **Revised manuscripts:** In page 8 line 185-187:

629 "The Statistical Package for Social Sciences (SPSS) version 23.0 was
630 used to perform Student's t-tests, Mann Whitney U test and linear regression
631 analyses".

632

633 **Comment 14** l.214-235: I am a bit lost here. Perhaps, this can be
634 concentrated to the information in aggregated form that is really necessary to
635 understand the results, thereby shortening it.

636 **Response:** We have shorted the 3.1 section with main findings from 24
637 lines to 7 lines.

638 **Revised manuscripts:** In page 8 line 190-197:

639 "The water chemistry in the freshwater reach of the Maludam, Simunjan,
640 and Sebuyau rivers are typical of blackwater rivers draining from peatland with
641 acidic pH and low DO concentrations, and the mixing with coastal water
642 increased the pH and DO (Table S1, Fig. S1). Values of pH and DO
643 concentrations in the Sematan and Lundu, which drain mostly mineral soils,
644 were higher than those in the blackwater rivers (Fig. S1). In the Rajang estuary,
645 values of pH and DO were lower in the riverine side, especially in the
646 distributaries where covered by the peat (Fig. S2)".

647

648 **Comment 15** l 223 and 231: Shouldn't the numbers of the supplementary
649 figures be switched (according to the sequence of their reference in the
650 manuscript)?

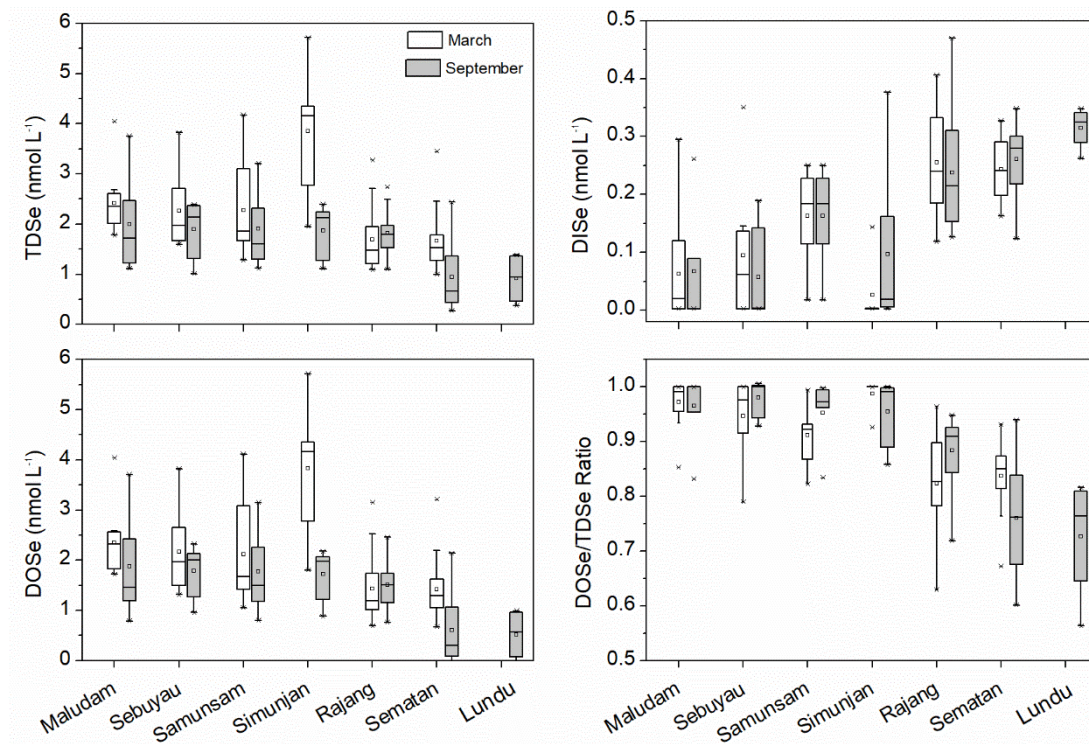
651 **Response:** We have switched the numbers of the supplementary figures

652 as their sequence in the context.

653

654 **Comment 16** 250-268: I suggest to show the results as bar diagram with error
655 and indication of statistically significant difference instead of the current Fig. 2,
656 which I suggest to move to the supplementary information.

657 **Response and revised manuscripts:** We have moved the current Fig. 2
658 to the supplementary information, and draw box plot of TDSe, DISe and
659 DOSe concentration and DOSe/TDSe ratio in the sampled rivers and
660 estuaries (Fig. 2 in the manuscript).



661

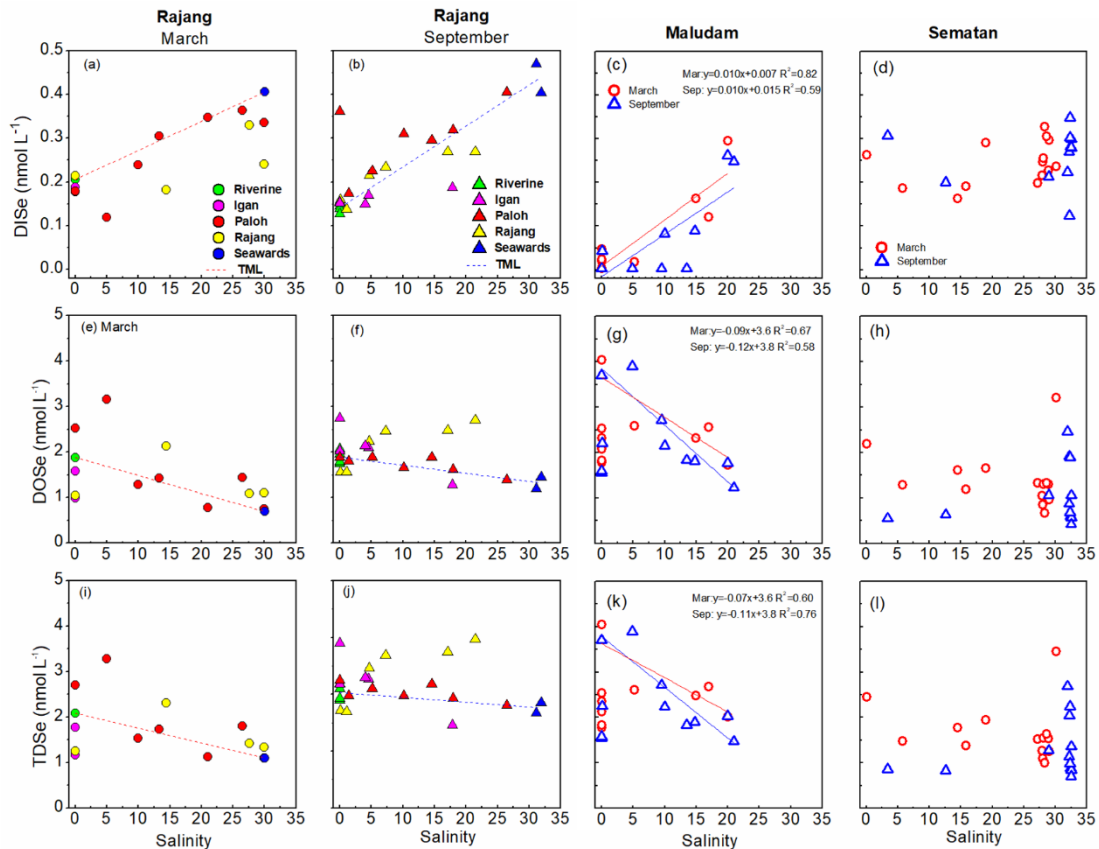
662 Figure 2 The box plot of TDSe, DISe and DOSe concentration and DOSe/TDSe ratio in
663 the sampled rivers and estuaries in Malaysia in March and September 2017, respectively.

664 In the plot of the upper panel, the ends of the box and the ends of the whiskers, and the
665 line across each box represent the 25th and 75th percentiles, the fifth and 95th
666 percentiles, and the median, respectively; the open square indicates the mean value.

667

668 **Comment 17** I. 349-358: I suggest to combine the results of each of your
669 three groups into a figure for the group instead of showing all individual
670 results.

671 **Response and revised manuscripts:** We have merged Fig. 3 and Fig. 5
 672 to one figure. Three typical groups of relationships between Se species and
 673 salinity were selected to present in Fig 3 in the manuscript, including Rajang,
 674 Maludam and Sematan estuaries, and those for Sebuyau and Samunsam
 675 were moved to supplement (Fig. S5 in the supplementary).



676
 677 Fig. 3 Relationships between DISe (a - d), DOSe (e - h), and TDSe (i - l) concentrations
 678 with salinity in the Rajang and three Rajang tributaries (Igan, Lassa, and Rajang), and in
 679 the Maludam and Sematan estuaries in March and September 2017. TML refers to the
 680 theoretical mixing line, which was defined using two endmembers: freshwater in the
 681 riverine system and seawater.

682
 683 **Comment 18.** I. 384ff: The Discussion section should be structured according
 684 to the three objectives into three parts. The objectives should be discussed as
 685 concisely as possible, i.e. the current discussion should be shortened.

686 **Response:** We have revised the objectives and reorganized the
 687 discussions according to the objectives. The discussion session was shorted
 688 from 280 lines to 152 lines.

689 The three objectives were 1) evaluate the fate of Se species during
690 estuarine mixing in peatland-draining estuaries; 2) characterize the DOSe
691 fractions; and 3) estimate the magnitude of Se fluxes delivered from peatland-
692 draining rivers to coastal ocean. The discussion and conclusion sections were
693 structured to three parts according to the objectives, as following: 4.1) Fate of
694 Se species during estuarine mixing; 4.2) Character of the DOSe fractions; and
695 4.3) TDS_{Se} flux.

696

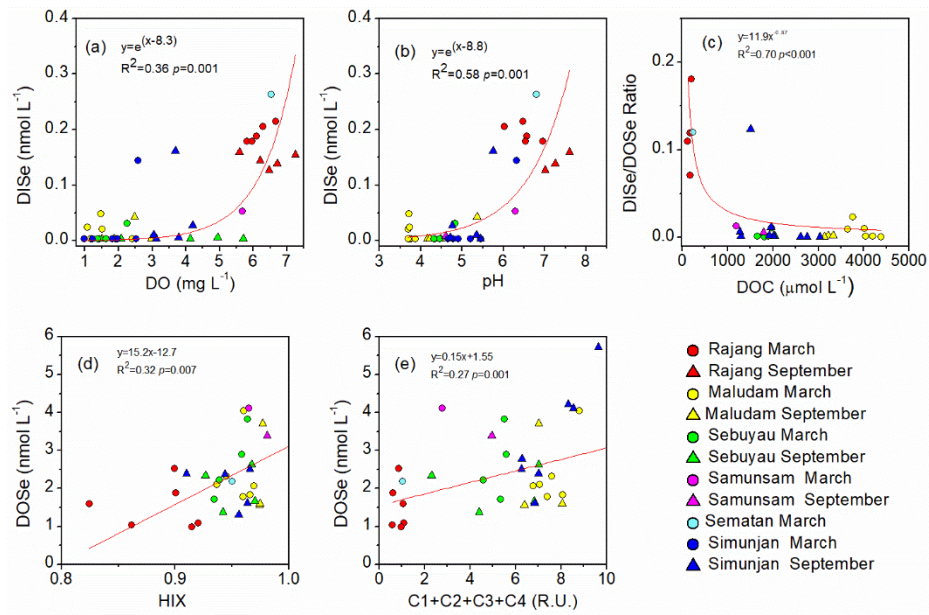
697 **Comment 19** l. 407-412: This belongs to the results.

698 **Response:** We have moved those to the results as section. "3.4
699 Correlation between Se species with DO, pH and DOM".

700 **Revised manuscripts:** In page 11 line 274-287:

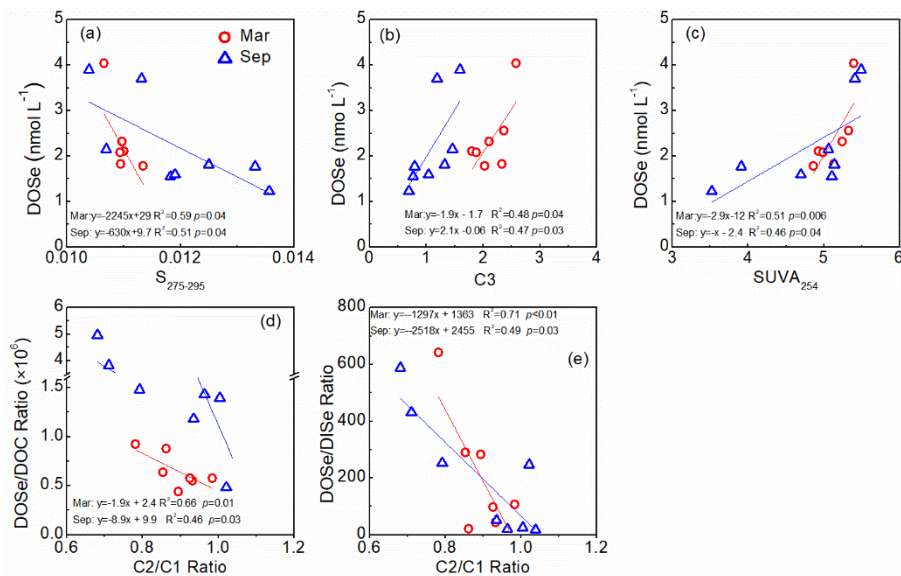
701 "For the freshwaters ($S < 1$) of the studied rivers, DISe concentrations were
702 positively correlated with the DO concentrations and pH values, and the
703 DISe/DOSe ratio was negatively related to DOC concentration (data from
704 Martin et al., 2018) (Fig 4a, 4b); DOSe concentrations correlated positively with
705 the humification index (HIX) and the sum of the humic-like chromophoric
706 dissolved organic matter (CDOM components, C1, C2, C3, and C4) ($p < 0.05$)
707 (data from Zhou et al., 2019) (Fig 4c, 4d).

708 In the Maludam Estuary, DOSe concentrations were negatively correlated
709 with the CDOM spectral slope from 275 to 295 nm ($S_{275-295}$) and were positively
710 correlated with the humic-like C3 component and specific UV absorbance at
711 254 nm ($SUVA_{254}$) during estuarine mixing in both seasons (data from Martin et
712 al., 2018; Zhou et al., 2019) (Fig 5a-c). In addition, DOSe/DOC and DOSe/DISe
713 ratios were negatively correlated with C2/C1 components ratios (Fig 5d; 5e)."



714

715 Figure 4. Relationships between (a, b) DISE concentrations and DO and pH values, (c)
 716 DISE/DISE ratios and DOC concentration values, and (d–e) DOSe concentrations with
 717 the humification index (HIX) and the sum of humic-like CDOM components (C1, C2, C3,
 718 and C4) in freshwater (Salinity < 1) for the Rajang, Sematan, Maludam, Sebuyau,
 719 Samunsam, and Simunjan rivers in March and September. The HIX and C1, C2, C3, and
 720 C4 components are from Zhou et al. (2019) from the same cruises. DO concentrations and
 721 pH values were not available for the Sematan River for September, and the HIX and CDOM
 722 components were not available for the Rajang River for September.



723

724 Figure 5. Relationships between DOSe concentrations and $S_{275-295}$, C3 components and
 725 $SUVA_{254}$, DOSe/DOC ratio and C2/C1 component ratios, and DOSe/DISE ratios and C2/C1
 726 component ratios in the Rajang and Maludam estuaries. The $S_{275-295}$, $SUVA_{254}$, C1, C2,
 727 and C3 components are from Martin et al. (2018) and Zhou et al. (2019) from the same
 728 cruises.

729

730 **Comment 20** l. 432-433: Why is this information important? I suggest to delete
731 it.

732 **Response:** We have deleted the “Se sorption kinetics on humic acids
733 can be expressed by a pseudo-second-order equation (Kamei-Ishikawa et al.,
734 2007).”

735

736 **Comment 21** l. 434-441: This is a repetition. Delete and focus on the important
737 statement in l. 441-442.

738 **Response:** We have deleted the “The Maludam, Sebuyau, and Simunjan
739 catchments are mainly peat, whereas the Samunsam River drains an
740 extensive area of peatland in its upper reaches (Müller et al., 2016; Martin et
741 al., 2018). The Rajang catchment is dominated by mineral soils, with peatland
742 being found only in the delta surrounding the distributaries (Staub et al., 1994,
743 2000)”

744

745 **Comment 22** l. 469: You can omit “as follows” and directly start with the
746 numbered list.

747 **Response:** We have deleted the “as follows”

748

749 **Comment 23** l. 479: Combine 4.1 and 4.2 as joint contribution to Objective 1.

750 **Response:** Thanks for the great advice. We have combined the 4.1 and
751 4.2 to “4.1 Fate of Se species during estuarine mixing”, as joint of contribution
752 to Objective 1.

753 **Revised manuscripts:** In page 12-14, line 289-364:

754 “4.1 Fate of Se species during estuarine mixing

755 On a global perspective, TDS_{Se} concentrations in the sampled rivers were
756 comparable with those in other reported rivers (between 0.2 and 30 nmol L⁻¹);
757 however, in contrast to our findings, DISe generally dominates in other rivers
758 (Table 2, Cutter, 1989b; Conde and Sanz Alaejos, 1997; Pilarczyk et al., 2019).
759 DOSe concentrations in rivers worldwide range from <0.02 to 0.82 nmol L⁻¹
760 (Takayanagi and Wong, 1984; Huang and Shy, 1995; Cutter and Cutter, 2001,
761 2004). In the blackwaters of the Orinoco in South America, TDS_{Se}
762 concentrations were found to range from 0.07 to 0.25 nmol L⁻¹ (Yee et al., 1987).
763 Although they did not analyse DOSe fractions directly, Yee et al. (1987)
764 assumed that DOSe was likely to constitute about 10%–15% of the total Se, a
765 much lower value than the DOSe proportions observed in peat-draining rivers
766 in Sarawak.

767 Species of Se are very sensitive to redox conditions and pH values
768 (Sharma et al., 2015). Se(IV) and the Se(VI) are soluble in water which exists
769 under mild and strong oxidizing conditions (Torres et al., 2010), thus DISe
770 concentrations be expected to increase with DO values (Fig. 4a). Sorption to
771 solid surfaces is a pH-dependent process, with substantial sorption of Se(IV)
772 and Se(VI) occurring at pH values of 4 to 6 and negligible sorption under more
773 alkaline conditions (pH > 8) (BarYosef and Meek, 1987; Balistrieri and Chao,
774 1987; Papelis et al., 1995; Sharma et al., 2015). Adsorption of Se(IV) and Se(VI)
775 by solid surfaces when pH is between 4 and 6 may help to explain the low DISe
776 concentrations in the sampled freshwater, and DISe concentrations be
777 expected to increase as pH increases (Fig. 4b). In addition almost 15% of Se(IV)
778 is removed by adsorption to peat (Kharkar et al., 1968). Se(IV) and Se(VI)
779 associated with humic and fulvic substances appear to be responsible for the
780 immobilization of inorganic Se (Kang et al., 1991; Zhang and Moore, 1996;

781 Wang and Gao, 2001). The DISe/DOS_e ratios negatively related with DOC
782 concentrations (Fig. 4c). DO, pH, and DOC concentrations of the water
783 probably contributed to the observed variations in Se species, and the acidic,
784 low-oxygen, and organic-rich blackwater rivers were not a suitable environment
785 for DISe.

786 During estuarine mixing, reversed DISe concentration–salinity
787 relationships were observed in the Rajang, Maludam, Sebuyau, and
788 Samunsam estuaries (Fig. 3, Fig S5), which were contrast with those reported
789 for other estuaries (Measures and Burton, 1978; Takayanagi and Wong, 1984;
790 Van der Sloot et al., 1985; Cutter, 1989a; Guan and Martin, 1991; Hung and
791 Shy, 1995; Abdel-Moati, 1998; Yao et al., 2006; Chang et al., 2016). The marine
792 endmember of the DISe concentrations in the sampled estuaries (salinity > 31)
793 was 0.30 nmol L⁻¹ (range: 0.12 to 0.47 nmol L⁻¹), encompassing or close to the
794 values reported for surface water in the South China Sea (around 0.38 nmol
795 L⁻¹, Nakaguchi et al., 2004) and the Pacific (mean of 0.24 nmol L⁻¹, range: 0.02
796 to 0.69 nmol L⁻¹) (Cutter and Bruland, 1984; Sherrard et al., 2004; Mason et al.,
797 2018). The salinity-related increases in DISe in a seaward direction indicate
798 that the patterns of distribution of DISe in those peat-draining estuaries are
799 controlled mainly by conservative mixing of ocean-derived DISe. In addition,
800 DISe was removed in March but was added in September in the Rajang estuary.
801 Laboratory studies have shown that Se(IV) can be adsorbed by peat and that
802 60% of the adsorbed Se(IV) can be desorbed upon exposure of the solid phase
803 to seawater (Kharkar et al., 1968). DISe may have been added to the Rajang
804 estuary in September via release of Se(IV) from peat in brackish waters. Other
805 studies have reported removal of the humic fractions of DOM, colloidal iron, and
806 phosphorus by flocculation in the river–sea mixing zones (Eckert and Sholkovitz,
807 1976; Forsgren et al., 1996; Asmala et al., 2014). Some of the DISe may exist
808 in colloidal form in natural water (Takayanagi and Wong, 1984), and DISe may

809 be removed by flocculation. In peat-draining estuaries, ocean-derived DISe
810 may be adsorbed to peat and may be associated with DOM, which is then
811 converted to DOSe and/or flocculated to particulate Se.

812 In contrast to DISe, DOSe concentrations were high in the rivers and
813 decreased in a seaward direction as salinity increased (Fig. 3, Fig S5). DOSe
814 has been shown to behave non-conservatively in other estuaries, with
815 concentrations decreasing along salinity gradients or with mid-estuarine input
816 (Cutter, 1989a; Guan and Martin, 1991; Hung and Shy, 1995; Abdel-Moati,
817 1998). DOSe concentrations in the estuaries studied in Sarawak were higher
818 than those reported in other estuaries (0.1 to 2.5 nmol L⁻¹) (Cutter, 1989a; Guan
819 and Martin, 1991; Hung and Shy, 1995; Abdel-Moati, 1998). The marine
820 endmember of the DOSe concentrations in the sampled estuaries (salinity >31)
821 ranged from 0.42 to 2.91 nmol L⁻¹ (mean: 1.32 nmol L⁻¹) and exceeded those
822 in surface water of the South China Sea (mean: 0.20 nmol L⁻¹, range: 0.33 to
823 0.14 nmol L⁻¹, Nakaguchi et al., 2004) and the Pacific (mean: 0.36 nmol L⁻¹,
824 range: 0.01 to 0.67 nmol L⁻¹ (Cutter and Bruland, 1984; Sherrard et al., 2004;
825 Mason et al., 2018). The high DOSe concentrations in coastal waters in
826 Sarawak (S > 30) suggest a significant contribution from terrigenous DOSe. In
827 the distributary channels of Rajang, there are large inputs of organic matter
828 from peat, thus higher DOSe concentrations than the TML values be expected
829 in most of the brackish waters (Fig. 3).

830

831 **Comment 24** l. 541/545: Isn't the Rhone forming a delta? Or don't you talk
832 about the French Rhone?

833 **Response:** Thanks, it's the French Rhone delta.

834

835 **Comment 25** l. 549: Skip this heading to avoid overstructuring.

836 **Response:** We have reorganized the discussion, skip the heading that

837 mentioned.

838

839 **Comment 26** l. 562: I am confused by the simultaneous use of delta and
840 estuary, because I think that these are two contrasting geomorphological
841 forms, mainly driven by the strength of the tide.

842 **Response:** We have unified the expression of estuary instead of delta in
843 the manuscripts to avoid confusion.

844

845 **Comment 27** l. 563-573: I would move large parts of this and the associated
846 figure to the results section.

847 **Response:** We have moved those to the results as section. “3.4
848 Correlation between Se species with DO, pH and DOM”, detailed response
849 was shown in comment 19.

850

851 **Comment 28** l. 588-589: The numbers should be subscripts.

852 **Response:** In the FDOM area, the C1 represents Component 1 that
853 decomposed from the excitation-emission matrix. In the previous publications,
854 C1 and C2 were widely used. For example, Table 1 in Osburn et al. (2012),
855 Table 1 and Figure 2 in Dainard et al. (2015), there is no need to be
856 subscripts.

857

858 **Comment 29** l. 593-601: I would again move large parts of this and the
859 associated figure to the results section.

860 **Response:** We have moved those to the results as section. “3.4
861 Correlation between Se species with DO, pH and DOM”, detailed response
862 was shown in comment 19.

863

864 **Comment 30.** l. 622: Will photodegradation really be important in the dark

865 DOM-rich waters? Possibly, it is restricted to the uppermost surface-near few
866 mm.

867 **Response:** Martin et al (2018) found that DOM from the Rajang and
868 Samunsam rivers was photolabile, with DOC and CDOM decreasing after
869 sunlight exposure. In addition, the peatland-derived DOM probably has too
870 short a residence time in rivers for significant photodegradation to occur in the
871 rivers before it reaches the sea, thus the authors suggested that most
872 photochemical transformations of tDOC in Sarawak likely take place after
873 tDOC reaches the sea rather than inside the rivers and estuaries.

874 **Revised manuscripts:** In page 15, line 392-398:

875 “As suggested by Martin et al (2018) that most photochemical transformations
876 of DOM in Sarawak likely take place after DOM reaches the sea. Thus, once
877 transported to offshore, photodegradation plays an important role in DOSe
878 processing, and DOSe might contain a significant photoreactive fraction that
879 facilitates photodegradation of DOSe into lower mean molecular weights or
880 gaseous Se or photomineralization to DISe.”

881

882 **Comment 31** l. 665-678: The conclusions should fit to the objectives, i.e.
883 there should be three main conclusions and perhaps a kind of outlook.

884 **Response:** Thanks for the great advices. We have revised the three objectives,
885 were 1) evaluate the fate of Se species during estuarine mixing in peatland-
886 draining estuaries; 2) characterize the DOSe fractions; and 3) estimate the
887 magnitude of Se fluxes delivered from peatland-draining rivers to coastal ocean.
888 The conclusions were reorganized to fit the objectives and have an outlook.

889 **Revised manuscripts:** In page 17, line 440-455:

890 “To the best of our knowledge, this is the first study of seasonal variations
891 in Se speciation in peat-draining rivers and estuaries in Southeast Asia.
892 Contrary to the results from studies elsewhere, DOSe, not DISe, was the major

893 species in the peat-draining rivers and estuaries of Sarawak, Malaysia.
894 Contrary to our expectations, reversed DISE concentration–salinity
895 relationships were observed in those estuaries, indicating a marine origin, while
896 DOSe concentrations decreased with salinity, indicating terrestrial sources. The
897 DOSe fractions may be associated with high-molecular-weight peatland-
898 derived aromatic and black carbon compounds and may photodegrade to more
899 bioavailable forms once transported to oligotrophic coastal waters, where they
900 may stimulate the growth of phytoplankton. The DOSe yields in the peatland-
901 draining rivers were one or even two orders of magnitude higher than other
902 reported rivers. The TDSe flux delivered by the exceeded other small rivers,
903 and it is quantitatively more significant than previously thought. The impact of
904 the sizable Se from increasing anthropogenic disturbing of peat to the
905 ecosystem should be evaluated in the future”

906

907 **Comment 32.** I 678: See my previous comments to the role of Se for
908 biological productivity.

909 **Response:** The detailed response was shown in comment 3. We have
910 changed to “may stimulate the growth of phytoplankton” in line 450.

911

912 **Comment 33** Figs. 3-5 show all individual results. I suggest to aggregate
913 these data in a way that clearly illustrates your main points.

914 **Response:** We have moved Fig. 2 and Fig. 4 to the supplement, and Fig.
915 3 and Fig. 5 were merged to one figure. Three groups of the relationships
916 between Se species and salinity were selected to present in Fig. 3, including
917 Rajang, Maludam and Sematan estuaries, and those for Sebuyau and
918 Samunsam were moved to supplement.

919 **Revised manuscripts:** Figure 3 were present in *page 29 line 738-743.*

920

921 **Comment 34** Figs. 7 and 8 should be included in the Results section.

922 **Response:** We have moved those figures to the results in section. "3.4
923 Correlation between Se species with DO, pH and DOM". The detailed
924 response was shown in comment 19, and figures were show as Fig. 4 and Fig
925 5.

926 **Revised manuscripts:** Figure 3 were present in *page 29 line 738-743*.

927

928

929

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