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1 **Fluorescence analysis of centralized water supply systems: indications for**
2 **rapid cross-connection detection and water quality safety guarantee**

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22 **Abstract**

23 Better insight into non-potable reclaimed water and drinking water can produce
24 more reliable risk assessment and help to achieve sustainable water reuse in the
25 long-term. This study illustrates the effectiveness of fluorescence excitation-emission
26 matrix (EEM) for rapid cross-connection detection. Based on samples collected from
27 three different sites of China, it is identified that the overall fluorescence intensity was
28 6-31 times of reclaimed to drinking water in water samples at all FRI regions. This is
29 shown to be highly sensitive over conventional water quality parameters. Furthermore,
30 based on parallel factor analysis (PARAFAC), humic acid and tryptophan are
31 considered as the main components contributing to fluorescence both in secondary
32 and tertiary effluents. Total fluorescence intensities as well as fluorescence peaks of
33 EEM pairs were investigated. Under different mixing scenarios, it is found that the
34 signal is distinguishable as low as 20% of reclaimed water. This study also offers
35 possibility of exploring portable devices with identified fluorescence peaks in EEM
36 regions for risk prevention and water quality monitoring at end user sites.

37

38 **Keywords:** reclaimed water; fluorescence excitation-emission matrix; PARAFAC
39 analysis; cross-connection; portable devices

40

41 **1. Introduction**

42 Nowadays, reclaimed water is being increasingly considered as an important

43 alternative water resource to alleviate water shortage and contamination situations in
44 many places of the world (WateReuse, 2019; Lefebvre, 2018; Yamashita, 2018; Wang
45 et al., 2017). It has been widely applied for agricultural, industrial and urban uses
46 (Ban, 2015; Cooper, 2003; GWI, 2017; Xu et al., 2020). There are examples of
47 large-scale water reuse schemes in office buildings, shopping centers, sporting venues
48 and residential areas worldwide (Lazarova et al., 2013; Takeuchi and Tanaka, 2020).
49 For instance, water reuse quantity in urban areas of China has already reached to 8.6
50 billion m³ in 2018 and is projected to increase further in the upcoming years
51 (MOHURC, 2019). The proportion of centralized systems accounts for 80% of the
52 overall water reuse systems across the country (Z. Chen et al., 2017; Qu et al., 2019).
53 Given the huge amount of available treated effluent, reclaimed water for municipal
54 and residential applications is likely to be further exploited in the near future,
55 especially in water deficient regions and densely populated urban areas (Salgot &
56 Folch, 2018).

57 Considering the economic feasibility, the concept of “fit-for-purpose” water
58 reuse normally applies in real practices which describes the treatment of reclaimed
59 water to a quality that meets the needs of the intended use. However, reclaimed water
60 (i.e. secondary and tertiary effluent) usually contains higher amount of organic
61 matters, nutrients and microorganisms than that of drinking water (Chen et al., 2017;
62 Cui et al., 2019; Rebelo et al., 2020). Hence, cross-connection issues of drinking and
63 reclaimed water pipelines need to be addressed appropriately.

64 Notably, there have been many reported accidents of cross-connection in the past
65 years, and some events have caused residents to inevitably consume or even drink
66 reclaimed water for a certain period of time. For instance, in May 2016, the anomaly
67 in tap water quality was detected in one residential area of Beijing, China, affecting
68 tens of thousands of residents in seven communities. This was caused by
69 cross-connection events between drinking and reclaimed water pipelines (Xu et al.,
70 2019). In Australia, more than 50 cases of cross-connection were discovered at Rouse
71 Hill in 2001, and a case in 2007 led to illnesses of staff in a treatment plant in
72 Melbourne (Hambly et al., 2012). Similar cases were reported in the U.S., Netherland,
73 Israel and Finland (Friedler et al., 2015; Laine et al., 2011; Xu et al., 2019).

74 Mena et al. (2008) conducted quantitative microbial risk assessment on
75 cross-connection simulation and concluded that drinking water that is affected by a
76 cross-connection event can result in a greater contaminated supply than that was due
77 to inadequate treatment. They calculated that risks of *Salmonella* infection range from
78 0.1 after a 1-day exposure to 0.99 for 30 and 90-day exposure of cross-connection
79 durations. As a result, cross-connections issues can lead to potential health risks and
80 public misgivings, which might hinder the utilization and expansion of reclaimed
81 water to a large extent (Son et al., 2012; Thayanukul et al., 2013; Weber et al., 2006;
82 Yi et al., 2011).

83 Consequently, it is vital to identify cross-connection events via effective
84 detection methods and guarantee reclaimed water safety and reliability in distribution

85 systems. At present, conventional parameters such as pH, temperature, turbidity,
86 conductivity, oxidation reduction potential (ORP) and residual chlorine are commonly
87 applied for water quality monitoring. These methods can be employed at reasonable
88 cost, but the reliability is compromised due to limited sensitivity and high fluctuation
89 (Friedler et al., 2015; Jayaratne, 2015; Xu et al., 2019). Some studies reported that
90 DOC and conductivity can be surrogate parameters to assess the removal efficiency of
91 trace organic chemicals during wastewater treatment and reclamation processes
92 (Dickenson et al., 2009; J. Drewes et al., 2012). However, as for cross-connection
93 detection, Hambly et al. (2010) observed that conventional parameters (e.g. UV₂₅₄,
94 DOC, conductivity and pH) normally involve with high detection limits. For instance,
95 a minimum of 70% contamination of reclaimed water in drinking water is required
96 when using conductivity for differentiation.

97 Another low-cost approach is to add non-toxic anti-ingestants, dyes and/or
98 colorants as tracers in water distribution systems (Storey et al., 2007). Nevertheless,
99 there are limited research and due to the complexity in distribution networks, it is
100 difficult to determine the proper dosing points and frequency. In addition, some
101 studies demonstrate that total organic carbon (TOC), total organic halides (TOX) and
102 ultraviolet (UV) light absorbance can be selected as predictors of the contaminants of
103 concern. Hajj-Mohamad et al. (2019) identified caffeine, theophylline and
104 acetaminophen can be suitable markers for sewer cross-connections, especially in
105 residential sectors. Although these methods can provide more insight into the origin of

106 organic matters, they are often costly and require a high degree of expertise for proper
107 assessment and maintenance (Dickenson et al., 2011; J. E. Drewes et al., 2003). There
108 are also some emerging biological sensors and rapid microbiological detection
109 approaches (e.g. flow cytometry and adenosine triphosphate bioluminescence), yet
110 these methods are relatively costly and require a level of expertise, which might be
111 impractical for rapid and wide investigation of cross-connection events (Sorensen et
112 al., 2018; Storey et al., 2011).

113 Optical technologies such as ultraviolet and fluorescence spectroscopy are
114 considered to be sensitive and of high resolution for characterizing organic matters in
115 aquatic systems in recent years (Henderson et al., 2009; Hudson et al., 2008)
116 Particularly, fluorescence spectroscopy requires little time, no addition of chemical,
117 small amount of sample, and can be handled easily. A fluorescence
118 excitation-emission matrix (EEM) measures the fluorescence intensity that is
119 generated at selected excitation wavelengths and detected at a certain emission
120 wavelength range (A. Hambly et al., 2010). It has already been employed to
121 characterize the raw water quality, monitor the treatment processes and serve as an
122 early warning of drinking water pollution (A. Baker et al., 2004; Carstea et al., 2016;
123 Hudson et al., 2008; Heibati et al., 2017). Some literature findings also demonstrate
124 the potentials of using fluorescence EEM as a rapid indication of microbial water
125 quality based on intrinsic fluorophores of microorganisms (Andy Baker et al., 2015;
126 Nowicki et al., 2018; Sherchan et al., 2018; Sorensen et al., 2018).

127 Several studies have already applied fluorescence-based method for the detection
128 and monitoring of cross-connection events in Australia (A. C. Hambly et al., 2010a;
129 Henderson et al., 2009). However, there are limited applications in other places, and
130 the applicability and characteristic wavelengths adaptive to various reclaimed water
131 samples and different mixture ratios still needs further research. Consequently, this
132 study aims to explore the effectiveness of fluorescence-based approaches in several
133 centralized water reuse systems in different places of China, and identify the
134 characteristic fluorescence intensity peaks as well as detection limits of reclaimed and
135 drinking water samples. Afterwards, there is a deep discussion on the potential
136 application of portable devices to provide quick judgement and early warning of
137 cross-connection risks.

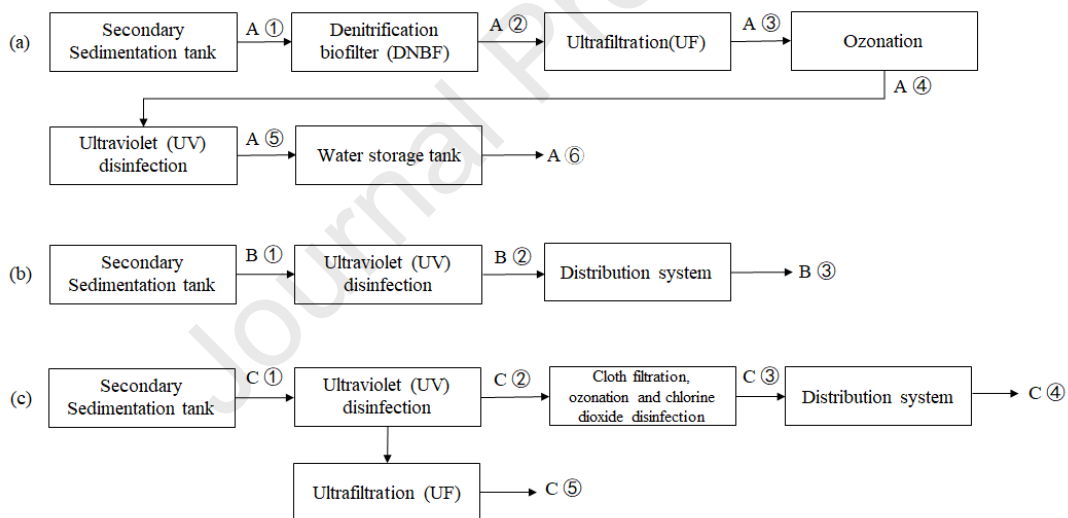
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139 **2. Material and methods**

140 **2.1 Sample locations**

141 Due to collection difficulties of water samples from distribution pipelines in
142 operation and end user sites, this study collected reclaimed water samples from three
143 large-scale centralized water reuse systems in different locations of China. One
144 system is in Beijing, northern China (plant A), and another two systems are in
145 Shenzhen, southern China (plant B and C). The daily water treatment capacity of
146 plant A, B and C is 1 million, 0.5 million and 0.35 million m³ respectively. The
147 process flow charts of each system are shown in Fig 1. To address varied reclaimed

148 water quality under different treatment scenarios, the reclaimed water sampling points
 149 include influent, treatment units (e.g. filtration, oxidation and disinfection), storage
 150 tanks and distribution pipelines. Meanwhile, drinking water samples are sourced from
 151 tap water outlets. Table 1 illustrates average water quality of reclaimed and drinking
 152 water samples. Differences in bulk water quality parameters including turbidity,
 153 conductivity, DOC can be observed. The samples were collected monthly over a
 154 7-month period. After collection, water samples were transported in cold storage (4 °C)
 155 to the laboratory and then filtered with 0.45 μm polyamide membrane filter
 156 (Whatman, England) for measurement.



157

158 Fig 1. Process flow chart and sampling points of the centralized water reuse systems

159 (a) Plant A, (b) Plant B and (c) Plant C

160

161 Table 1. Conventional water quality parameters of reclaimed and drinking water

162 samples

Water quality	Reclaimed	Reclaimed	Drinking	Drinking
---------------	-----------	-----------	----------	----------

parameters	water (A)	water (S)	water (A)	water (S)
pH	6.86–7.34	6.42–7.84	7.24–7.74	7.16–7.84
Turbidity (NTU)	2.3–4.3	0.4–4.7	0.11–0.36	0.12–0.28
Conductivity ($\mu\text{S}/\text{cm}$)	617–1033	497–729	613–816	107–142
DOC (mg/L)	8.0–11.1	4.89–11.27	2.2–4	1.32–2.37
UV ₂₅₄ (cm^{-1})	0.062–0.118	0.062–0.130	0.013–0.023	0.010–0.023

163 Notes: A-Plant A in northern China; S-Plant B and C in southern China; number of
 164 reclaimed water samples (n=21), number of drinking water samples (n=9).

165

166 2.2 Water quality analysis

167 Apart from collected reclaimed and drinking water samples, mixtures of different
 168 proportions of water samples were also conducted, in order to identify the sensitivity
 169 and effectiveness of detection methods for cross-connection detection under low
 170 mixing scenarios. The mixing ratio are set to be 40%, 30%, 20%, 10%, 5% (the
 171 volume proportion of reclaimed water) respectively.

172 Fluorescence EEMs were determined by F-7000 fluorescence spectrophotometer
 173 (Hitachi, Japan) using quartz cuvette with 1 cm optical path. Before sample analyzing,
 174 quinine sulfate solution (1 $\mu\text{g}\text{-QS}/\text{L}$) was applied to evaluate the stability of xenon
 175 lamp of fluorescence spectrophotometer. Afterwards, water samples were analyzed
 176 and ultrapure water was used as negative control. Fluorescence EEMs of water
 177 samples were depicted for excitation wavelength of 220-450 nm at 5 nm increments
 178 and emission wavelength of 240-600 nm at 1 nm increments. In EEM spectra,
 179 Rayleigh scattering region and Raman scattering region were excluded. Notably, some

180 studies also highlight the necessity for correction of the inner filtration effect (IFE)
181 with the presence of high contents of dissolved humic material (Ohno, 2002; Carstea
182 et al., 2016). In this case, correction methods such as dilution or empirical correction
183 based on the absorbance profile of the sample can be applied (Henderson et al., 2009).
184 In this study, no IFE correction was conducted to the dataset and raw fluorescence
185 values since it is indicated that IFE can be neglected when the UV_{254} absorbance
186 value is $<0.3 \text{ cm}^{-1}$ (Ohno, 2002) or TOC of $< 25 \text{ mg/L}$ in treated wastewaters (Hudson
187 et al., 2007; Henderson et al., 2009).

188 Other conventional water quality parameters were also measured for comparison.
189 Ultraviolet absorbance (UV_{254}) spectra were obtained by UV-2600 2300II ultraviolet
190 spectrophotometer (Shimadzu, Japan). Conductivity was measured by HACH-3400
191 2100Q portable meter (HACH, USA) and pH was measured by PB-10 pH meter
192 (Sartorius Design, Germany). Dissolved organic carbon (DOC) was detected by
193 TOC-VCPH analyzer (Shimadzu, Japan).

194 **2.3 Data processing and interpretation**

195 The EEMs were processed by using the software of MATLAB R2010b, and the
196 contour maps were drawn by the software of Origin 9.1. The Rayleigh scattering
197 region and Raman scattering region are omitted from the spectrum. The fluorescence
198 spectra of water samples are divided into five zones by using the Fluorescence region
199 integration (FRI) method described by Chen et al. (2003) and Wang and Zhang (2010).
200 In addition, parallel factor analysis (PARAFAC) was employed to further interpret the

201 possible fluorescence components of water samples. PARAFAC is a generalization of
 202 Principle Components Analysis (PCA) to higher order arrays, which is a useful
 203 method decomposing the fluorescence signal into potential individual fluorescent
 204 phenomena signal to characterize, model and visualize multivariate data of EEM and
 205 quantify the changes of DOM (Bro, 1997; Stedmon and Bro, 2008). In the analysis, it
 206 models the data of EEM using the following equation by minimizing the residual sum
 207 of squares (Sedmon and Bro, 2008).

$$208 \quad x_{ijk} = \sum_{t=1}^F \alpha_{if} b_{jf} c_{kf} + \varepsilon_{ijk}, i = 1, \dots, I; j = 1, \dots, J; k = 1, \dots, K; \quad (1)$$

209 where:

210 x_{ijk} : the fluorescence intensity of sample i measured under the excitation
 211 wavelength of k and emission wavelength of j ;

212 $\alpha_{if}, b_{jf}, c_{kf}$: the parameters of the model;

213 ε_{ijk} : the residual sum of squares, representing the unexplained signal (including
 214 noise and other variation).

215 The PARAFAC simulation was conducted by MATLAB R2010b and the
 216 DOMFluor toolbox developed by Stedmon and Bro (2008) and Murphy et al. (2011).
 217 Specifically, leverage was conducted to remove outliers. Residual and split-half
 218 analysis were then used to identify the optimal components of the PARAFAC model.

219

220 **3. Results and discussion**

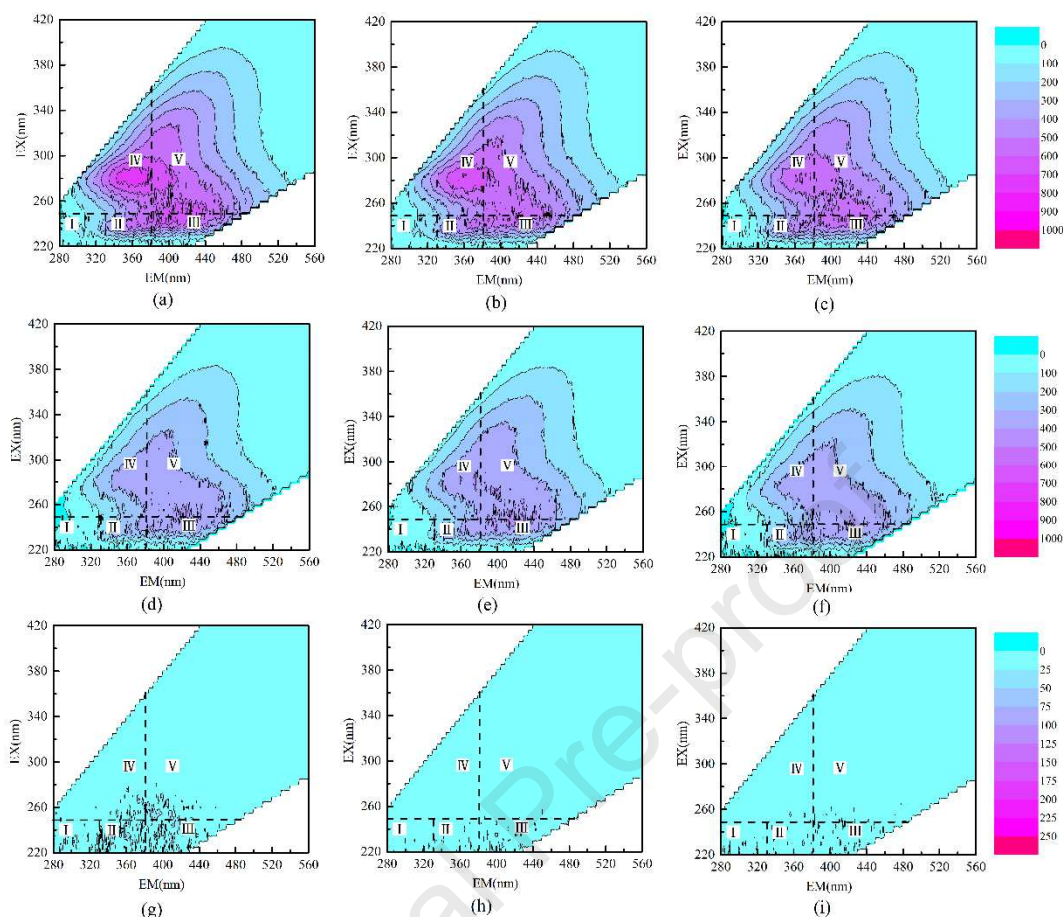
221 **3.1 Conventional water quality parameters**

222 A wide range of conventional parameters are used to assess water quality of
223 reclaimed and drinking water samples in three WRPs of two different cities in China.
224 As shown in Table 1, bulk parameters such as turbidity, conductivity, DOC and UV_{254}
225 enable differentiation between reclaimed and drinking water. These parameters can be
226 equipped and implemented at low cost and are widely used for online monitoring.

227

228 **3.2 Florescence EEMs of reclaimed water and drinking water**

229 Typical fluorescence EEMs obtained over the study period at each sample
230 location are illustrated in Fig. 2, Fig. S1 and Fig. S2 respectively. The spectrograms
231 are analyzed by FRI approach as depicted by Chen et al. (2003) and Wang and Zhang
232 (2010). Each spectrogram includes fluorescence peaks that are mostly related to
233 humic-like, tryptophan-like, and/or fulvic-like organic compounds (Hambly et al.,
234 2012).



235

236 Fig 2. Typical EEMs of effluents of Plant A: (a) secondary sedimentation tank; (b)

237 DNBF; (c) ultrafiltration; (d) ozonation; (e) ultraviolet disinfection; (f) water storage

238 tank; (g)-(i) drinking water.

239 Notes: Region I: Aromatic Protein I and tyrosine. Region II: Aromatic Protein II and

240 tryptophan. Region III: Fulvic acid-like. Region IV: Soluble microbial by-product-like.

241 Region V: Humic acid-like. Regions are divided according to Chen et al. (2003).

242

243 Overall, the fluorescence intensity of reclaimed water samples decreased along

244 through the treatment trains, both in secondary effluent (i.e. secondary sedimentation

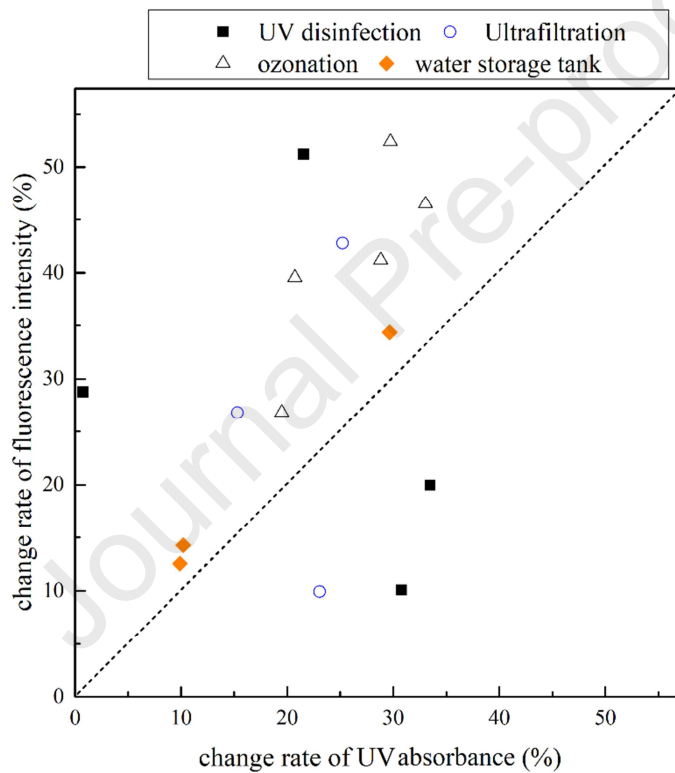
245 and DNBF) and tertiary effluent (i.e. ultrafiltration, ozonation and ultraviolet

246 disinfection). Relatively more significant fluorescence intensity decline was observed
247 in UF (Fig. 2(c)) and ozonation (Fig. 2(d) and Fig. S2 (e)) stages. Comparatively, Vera
248 et al. (2017) studied drinking water samples and found that UF stage hardly removed
249 fluorescent organic matters due to little effect on the rejection of medium and low
250 molecular weight compounds. Since tryptophan, nicotinamide adenine
251 dinucleotide-hydrogen (NADH) and riboflavin in bacteria and metabolites can also
252 exhibit fluorescence emissions, the removal of microorganisms (e.g. bacterial
253 indicators) might contribute to the decrease of fluorescence intensity in UF stage to
254 some extent as well. Nevertheless, the presence of organic matters in reclaimed water
255 makes it difficult to quantify fluorescence signals of microorganisms solely (Sherchan
256 et al., 2018).

257 After ozonation, the fluorescence intensity has decreased considerably since
258 ozone may oxidize and destroy the conjugated structure of fluorescent molecules and
259 affect the intersystem crossing efficiency (Cory and McKnight, 2005). Despite that
260 UV disinfection processes can remove bacterial indicators to some extent, there are
261 limited impacts on overall fluorescence signals. With respect to geographical disparity,
262 since the DOC concentrations of both reclaimed and drinking water samples in Plant
263 A of Beijing are slightly higher than that of Plant B and C in south regions, more
264 intense fluorescence signals are generally observed in secondary effluent from Plant
265 A.

266 To better distinguish reclaimed water from drinking water, statistical analyses on

267 fluorescence EEMs of water samples are further conducted. The total fluorescence
 268 intensity of reclaimed water is found to be 13 times higher than that of drinking water.
 269 UV_{254} absorbance and total fluorescence (TF) are both correlated with concentrations
 270 of organic matters (Gerrity et al., 2012), and the sensitivity of these two parameters
 271 are further compared during advanced treatment processes (i.e. secondary to tertiary
 272 effluent).



273

274 Fig 3. Differential UV_{254} absorbance (ΔUV) and total fluorescence (ΔTF) during
 275 secondary to tertiary treatment (n=15)

276

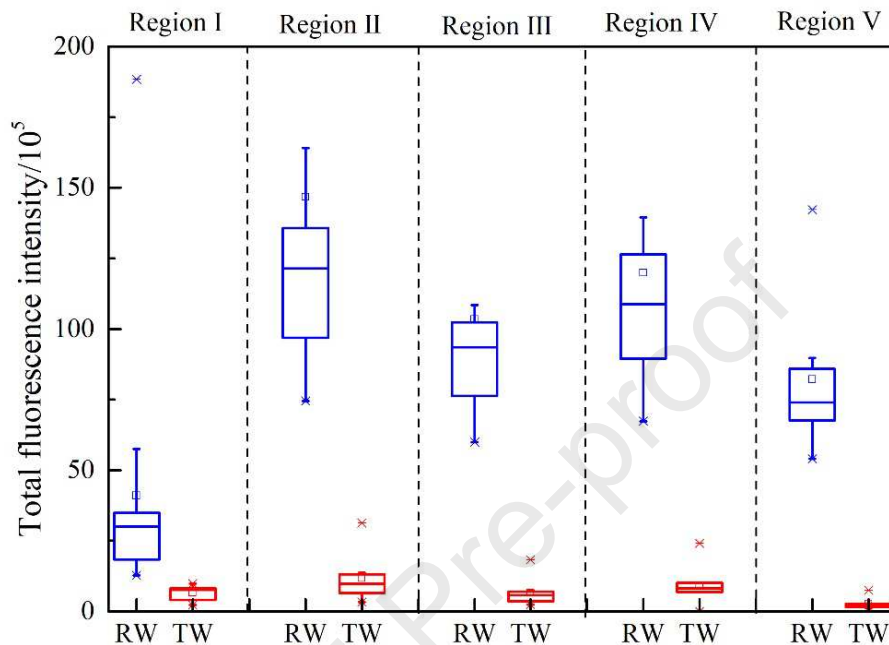
277 According to Fig. 3, both UV_{254} absorbance and total fluorescence declined
 278 along through the water reclamation processes. For a single fluorophore, the ΔTF and

279 ΔUV_{254} show the same relative change when the concentration varies. However, in
280 actual reclaimed and drinking water samples, some fluorescent substances may
281 exhibit low absorbance at 254 nm. As shown in Fig. 3, during advanced treatment
282 processes, larger variations of ΔTF are observed in most reclaimed water samples,
283 indicating that ΔTF is a more sensitive indicator than UV_{254} absorbance. Similar
284 phenomena are also noticed in other previous studies (A. C. Hambly et al., 2010b;
285 Saadi et al., 2006).

286 Meanwhile, comparative analyses are conducted on the total fluorescence
287 intensity of all reclaimed and drinking water samples in each FRI region (W. Chen et
288 al., 2003). This research did not further separate reclaimed water samples according to
289 WRPs and treatment processes in order to represent a wider range of different WRP
290 effluents.

291 According to Fig. 4, the fluorescence intensities of reclaimed water samples were
292 6, 13, 16, 13, and 31 times higher than those of drinking water samples in Region □ –
293 □ separately, and 13 times higher when the intensities of the five regions were
294 summed up. Particularly, distinct differences can be observed in Regions □, □, □ and
295 □. In general, fluorescence peaks at these regions are related to aromatic proteins,
296 fluvic acids, soluble microbial byproducts, humic acids and microbial NADH (W.
297 Chen et al., 2003; Fujioka et al., 2018). Similar results have been observed in the
298 study of Hambly et al. (2010b), in which the differentiation of fluorescence intensity
299 in reclaimed and drinking water samples of Australia is about 5–12 times. These

300 results demonstrate that the fluorescence EEM can be considered as a promising and
 301 straightforward tool for water quality monitoring and cross connection detection.



302

303 Fig 4. Fluorescence intensity of reclaimed and drinking water samples in each FRI
 304 region. 'RW'=reclaimed water (n=21), 'TW'=drinking water (n=9).

305

306 In addition to real-time monitoring via FRI approach, the PARAFAC technique is
 307 further applied to identify the key fluorescent components in reclaimed water samples.
 308 Particularly, it can decompose the fluorescence spectra in a set of unique components
 309 to further analyze the potential underlying EEMs by a least square algorithm (Hidayah
 310 & Cahyonugroho, 2019; Oloibiri et al., 2017; Pan et al., 2017; Vera et al., 2017). This
 311 approach can also shed light on the fingerprint of reclaimed water treatment in
 312 different degrees (Murphy et al., 2011).

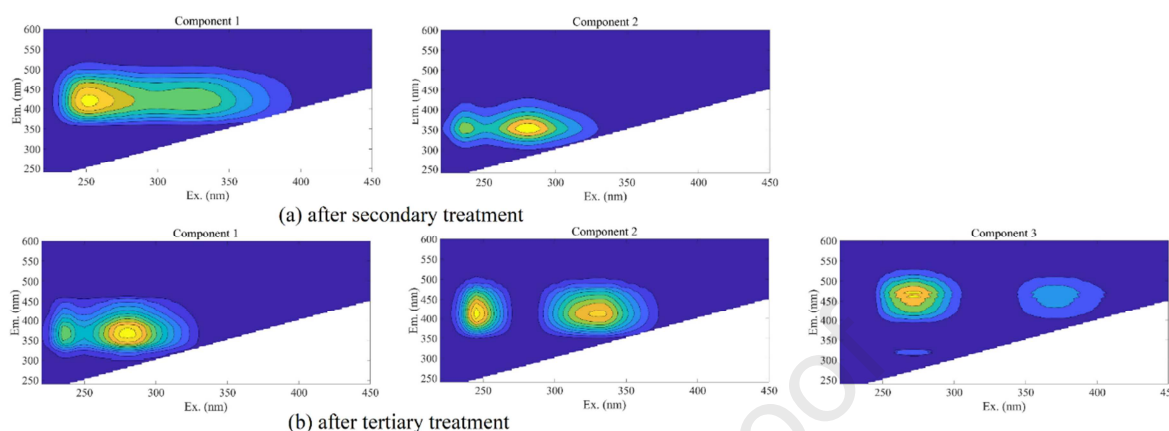
313 More specifically, in the PARAFAC simulation process, model fit was evaluated

314 by checking the residuals. Comparison of the loadings and spectral sum of squared
315 error of 3, 4 and 5 component models showed that the increase of number of
316 components hardly improved model fit, which suggested that 3 or 2 component
317 models were suitable for the data. Split-half analysis was then used to further check
318 the validity of 3 and 2 component models by running the models on two randomly
319 split halves of the data.

320 As can be seen from Fig. 5, for secondary effluent, 2-component PARAFAC
321 model is validated. The identified components exhibit a single peak at λ_{ex}/em of
322 260/425 nm and 280/355 nm respectively. Comparatively, for tertiary effluent, both 2
323 and 3 component models were validated. The plots of residuals in 2 and 3 component
324 models indicated that the latter fit tertiary effluent better. The component C1 exhibit a
325 single fluorescence peak whereas components C2-C3 both contains two peak signals.
326 The inferred contributing substances in water samples are presented in Table 2. In
327 general, humic acid-like, tryptophan and tyrosine-like substances are major
328 fluorescence components identified in all reclaimed water samples despite of
329 treatment levels. The spectral shift during secondary to tertiary treatment is probably
330 attributed to oxidation processes as well as other effects (e.g. pH, matrix and
331 interactions) that affect the fluorophores (Murphy et al., 2011). Nevertheless, most of
332 current studies that combine EEM with PARAFAC or principal component analysis
333 (PCA) are restricted to tracking organic matters. Future research should also address
334 fluorescence signals that emitted by microbial species and related activities especially

335 during secondary treatment.

336



337

338 Fig 5. Fluorescence EEM contour plots and spectral loadings of the major

339 components in secondary and tertiary effluents identified by EEM-PARAFAC

340 analysis

341

342 Table 2. Characteristics of components identified by EEM-PARAFAC analysis

Component	Peak location $\lambda_{ex/em}$ (nm)	Substances	References
(a) Component 1	260/425	Terrestrial humic-like	Yang et al., 2017
(a) Component 2	280/355	Tryptophan & protein-like	W. Chen et al., 2003
(b) Component 1	280/375	Tyrosine-like	Yang et al., 2017
(b) Component 2	330/420; 230/420	Humic acid-like & fulvic acid-like	W. Chen et al., 2003; Vera et al., 2017
(b) Component 3	275/475; 385/475	Humic acid-like	Vera et al., 2017; Wu et al., 2018

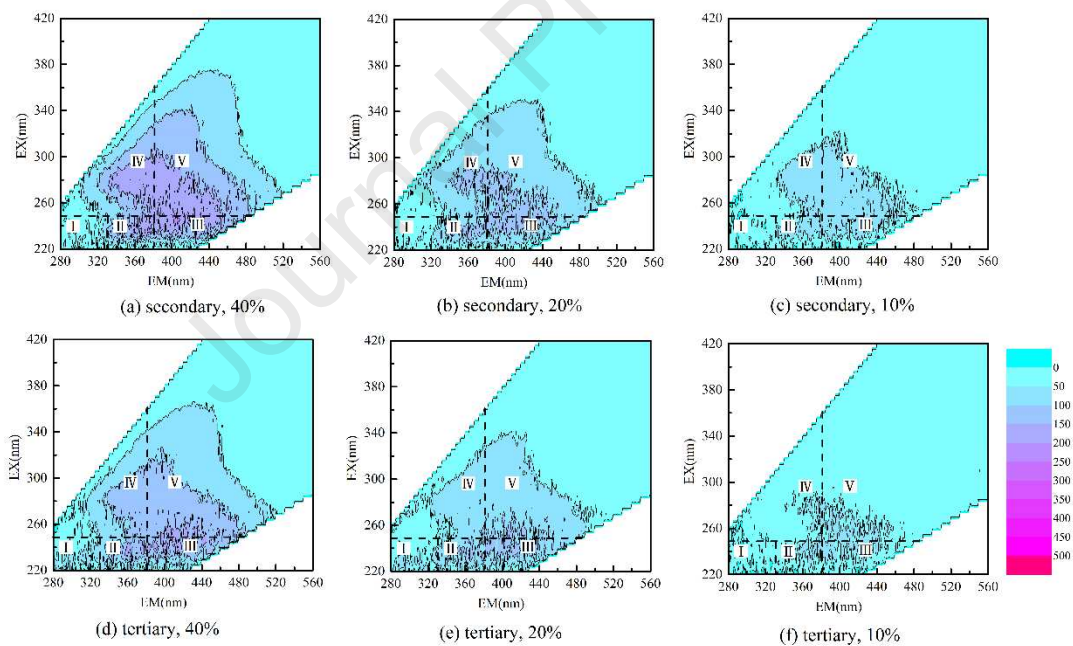
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344 3.3 Detection limits of fluorescence EEM with different mixing ratios

345 Remarkably, cross connection issues not always lead to a complete displacement

346 of drinking water with reclaimed water (A. C. Hambly et al., 2010b; Xu et al., 2019).

347 If the cross connection happens in branches of water distribution networks or at end
 348 user sites, it is likely that only a proportion of reclaimed water is blended with
 349 drinking water. In these situations, it is vital to determine the fluorescence responses
 350 under different water mixing ratios and figure out the detection limits. Fig.6 shows the
 351 typical fluorescence EEMs under different mixing proportions of reclaimed and
 352 drinking water samples. Notably, reclaimed water samples of different quality (i.e.
 353 secondary and tertiary effluent) are included for investigation. With the decreased
 354 proportion of reclaimed water interfusion, fluorescence signals diminish
 355 proportionally.



356

357 Fig 6. Typical fluorescence EEMs of water samples with different mixing ratios

358

359 The calculated total fluorescence intensities with respect to different reclaimed

360 water quality and varied mixing ratios as well as drinking water are listed in Table 3.

361 The detection limit of cross connection is generally defined as the value at which the
 362 fluorescence peak of mixed water sample can consistently be at least three times than
 363 that of uncontaminated drinking water (A. C. Hambly et al., 2010b). Differences
 364 between EEMs of mixed water samples (Fig. 6) and drinking water samples (Fig. 2)
 365 could be observed in even the 10% samples, however, based on the results in Table 3,
 366 20% was a more reliable detection limit.

367

368 Table 3. Calculated average fluorescence intensities of mixed water samples (n=12)

Total fluorescence intensity (10^5)	Different ratios of reclaimed water interfusion			Drinking water (n=9)
	40%	20%	10%	
Blend of reclaimed water (secondary effluent) and drinking water	187	139	93	39
Blend of reclaimed water (tertiary effluent) and drinking water	169	135	80	

369

370 Apart from total fluorescence intensity, peak signals in different FRI regions are
 371 further investigated. The identified fluorescence peaks (wavelength pairs $\lambda_{ex/em}$) in
 372 each region which can distinguish reclaimed water from drinking water largely offer
 373 possibility of developing portable fluorescence devices for onsite cross connection
 374 detection (Coble, 1996; A. C. Hambly et al., 2010b; Lee & Ahn, 2004). Compared to
 375 full excitation wavelength scanning, peak picking is more convenient and affordable
 376 for portable devices (Henderson et al., 2009; Vera et al., 2017). The identified
 377 fluorescence peaks in each FRI region of different water samples are illustrated in
 378 Table 4. The recognized EEM pairs are comparable to previous studies in other places

379 (Carstea et al., 2016; A. C. Hambly et al., 2010b; Lee & Ahn, 2004). The reclaimed
 380 water samples from Plant B and C in southern China displayed apparent fluorescence
 381 signals with related to humic acid-like substances in Region V, while samples from
 382 Plant A generated fluorescence signals as a result of multiple compounds and possible
 383 interactions.

384

385 Table 4. Fluorescence peak of EEM pairs identified in different water samples
 386 (Excitation-emission pairs, $\lambda_{ex/em}$ (nm))

Region	Reclaimed Water (A)	Drinking water (A)	Reclaimed Water (S)	Drinking water (S)
I	250/320	250/320	230/290	230/290
II	245/378	245/378	250/340	250/340
III	245/402	240/402	240/450	240/450
IV	285/362	270/370	280/350	275/320
V	280/380	265/380	335/430	335/430

387 Notes: A-Plant A in northern China; S-Plant B and C in southern China.

388

389 Furthermore, at each FRI region with identified EEM pairs, the sensitivities
 390 under different scenarios of reclaimed water interfusion are researched. As
 391 summarized in Table 5, in all FRI regions, a sufficient detection of cross connection
 392 by fluorescence approach required a minimum of 20-30% reclaimed water in drinking
 393 water. As shown in Table S2, there are minor fluctuations in conventional parameters
 394 under different scenarios of reclaimed water interfusion, suggesting that EEM is more
 395 sensitive than conventional parameters. The above-mentioned results suggest that
 396 when cross-connection incidents happen, fluorescence intensity at certain EEM pairs

397 can also respond sensitively. Therefore, it is promising to use portable devices with
 398 selected EEM pairs for cross connection detection. Nevertheless, the response
 399 threshold value (i.e. signal-to-noise ratio) that is currently set to be roughly 3 still
 400 needs further investigation (A. C. Hambly et al., 2010b; Lee & Ahn, 2004).

401

402 Table 5. Ratio of fluorescence signals of mixed water sample to drinking water at FRI
 403 peak positions (n=12)

Different scenarios of reclaimed water interfusion	Region I	Region II	Region III	Region IV	Region V
10%	2.86±1.12	2.32±0.73	2.53±0.56	2.65±0.68	2.50±0.61
20%	4.86±2.05	3.65±0.83	6.03±1.24	4.87±0.84	4.08±0.62
30%	5.20±1.89	4.49±0.58	7.84±2.24	6.71±1.23	5.87±0.83
40%	6.27±1.78	5.51±0.96	9.06±2.19	8.83±1.50	7.51±1.29

404

405 3.4 Discussion

406 For the actual cross-connection issues of dual reticulation water supply systems,
 407 the mixing ratio of reclaimed and drinking water may vary with time depending on
 408 water supply pressures, pipe junction locations, end use situations, etc. Hence, high
 409 sensitivity of detection tools is of great importance. This study indicates that full
 410 wavelength fluorescence spectroscopy can serve as a sensitive and reliable tool with
 411 considerably low detection limit of chemical and microbial compounds in water. This
 412 provides application potentials to detect failures and risks promptly in WRPs and
 413 water treatment plants. As such, water quality safety can be guaranteed in a more
 414 reliable and systematic way, and engineered storage facilities within plants would be

415 likely reduced due to faster response time, resulting in significant cost savings.

416 Meanwhile, since cross connection events could also happen at the ends of pipe
417 networks and/or end use sites, portable detection devices with certain EEM
418 wavelength pairs can be considered which are handy, less costly and easy to operate.
419 They are expected to have broad application prospects for water practitioners as well
420 as the community. The findings from this study suggests the design of portable
421 devices with several characteristic excitation/emission pairs (e.g. λ_{ex}/em of 240/450
422 nm and 280/350 nm at FRI region III and IV respectively) for real applications. In
423 addition, characteristic wavelength pairs can be used in portable monitoring devices
424 with composite probes (e.g. fluorescence spectrum coupled with conventional
425 parameters) to further ensure detection accuracy. Researchers have proposed the idea
426 of composite monitoring devices. For example, Lu et al. (2012) have reported a
427 portable device with both UV/vis absorbance and fluorescence detectors for food and
428 agricultural safety tests, which is convenient and economical for field analyses.

429 Although this research analyzed reclaimed and drinking water samples from
430 three different places in two cities of China, additional field information is suggested
431 to be collected in future studies so as to give an overview towards a broader area. At
432 present, there are several commercially available online fluorescence sensors yet the
433 optional wavelengths of the light sources are still limited. With the enhanced
434 awareness of water safety among the public, as well as the promotion of reclaimed
435 water reuse for multiple purposes, there are great opportunities and wide prospect for

436 online monitoring. Much effort can be devoted to improve water quality monitoring
437 devices adaptive in water and wastewater treatment processes, as well as water
438 distribution and end use sites.

439

440 **4. Conclusion**

441 This study conducted water quality analyses of reclaimed and drinking water
442 samples in several locations of China with an emphasis of fluorescence techniques.
443 Notably, the fluorescence EEM is found to be effective in terms of high sensitivity (i.e.
444 6-31 times of reclaimed to drinking water in analyzed samples at all FRI regions) and
445 low detection limit under a small proportion of 20% reclaimed water blending. This
446 offers possible indications for cross-connection contamination of reclaimed water (i.e.
447 secondary and tertiary effluent) with drinking water. Moreover, the EEM-PARAFAC
448 approach further recognized humic acid and tryptophan as main fluorophores despite
449 of different treatment stages. In addition, the identified characteristic EEM
450 wavelength pairs such as $\lambda_{ex}/\lambda_{em}$ at 240/450 nm and/or 280/350 nm offer the
451 possibility of designing portable detection devices for real applications at distribution
452 and end use sites.

453

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458

459 **6. Appendix A. Supplementary data**

460 Supplementary data associated with this article can be found, in the online
461 version.

462

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