Revision Report

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Title: Dying to find the source – the use of rhodamine WT as a proxy for soluble point source pollutants in closed pipe surface drainage networks

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Dear Editor and Referees,

The authors wish to thank the two referees for their constructive, considered responses. In addition to addressing each specific comment in the table below we would like to also acknowledge their general concerns. We recognise the limitations of this study, particularly with respect to quantifying possible interactions between multiple environmental conditions and in relation to the lack of flow measurements in the field. Whilst examining the potential for interactions between environmental conditions on fluorometric response is indeed important, this study aimed to quantify the fluorometric responses to each examined factor independently of one another. By adopting this approach we can clearly note the effect of altering the temperature on fluorometric response, for instance, and state with confidence that this response is due only to changing temperature. This does not mean that it may not be necessary to quantify confounding effects of multiple environmental factors, particularly when a study site exhibits highly variable water conditions, but such examination was outside of the scope of this particular study. We thank Referee #1 for again bringing this to our attention, and have added a relevant recommendation for future researchers to the conclusions of the manuscript.

We recognise the importance of flow gauging information when conducting precise quantitative dye tracer studies. The collection of flow measurements during this study would have removed the need to make a number of assumptions within the longitudinal dispersion theory analysis, and as Referee #2 correctly states this would have greatly contributed to the quantitative aspect of the study. However, despite the lack of flow gauging and subsequent need to apply assumptions within the analysis, this study demonstrates an appropriate and useful methodology for quantitative RWT release studies that future researchers may build upon through the addition of flow gauging. Similarly, future work may also build upon this methodology through the use of another truly conservative tracer, such as Br as suggested by Referee #2, to tease apart the effects due to RWT behaviour and due to the system under study.

We are providing detailed point-by-point revision report in the table below and would welcome any additional comments or suggestions that the referees and the editor may wish to provide.

Thank you very much for your constructive and helpful comments which have contributed to the improvement of our submission.

Detailed revision report:

Comments	Action Taken
Referee #1 General Comments	
There is no evaluation or discussion of turbidity, which has known and potentially substantial interference with fluorescence.	Explained that effect of turbidity from field site water was briefly examined in the laboratory and deemed insignificant (Introduction, approximately page 4537, line 14).
The authors need to provide the rationale behind the choice of waters used in the experiments and a discussion of the potential influence on results.	Please see response for Specific Scientific Comment #5.
Referee #1 Specific Scientific Comments	
1. (Abstract, page 4536, lines 13-14) This concept is not discussed in the text of the paper.	Removed the sentence from the abstract.
2. (Introduction) The key to this study is the evaluation and field use of an in situ fluorometric sensor specific to Rhodamine WT and may perform differently than standard fluorometric approaches. This point should be clearly emphasized in the introduction because it underscores the fundamental importance of the study.	Additional information added to final paragraph of introduction.
3. (Materials and Methods, Section 2.1 In situ instrumentation) The YSI 6130 has known turbidity interferences (~0.03 ug/L per NTU, but potentially larger under highly turbid conditions, www.ysi.com); however, turbidity was not considered in this study. Given the emphasis on quantitation, the authors need to discuss why turbidity was not included in the study. Understanding turbidity interferences would be particularly important when conducting studies under stormwater runoff/high flow conditions.	Agree that this is a valid concern, and have added additional information in the Introduction to address this (approximately page 4537, line 14).
4. (Materials and Methods, Section 2.3 Laboratory methodology) What were the ambient light conditions during the stability study?	Added this information.
5. (Materials and Methods, Section 2.3 Laboratory methodology) Why were the pH experiments conducted only on the Swan River water and the temperature and salinity experiments conducted only on deionized water? There are likely interactions between environmental conditions and fluorometric response that were potentially overlooked with this approach, particularly for temperature. Background fluorescence may be highly	The intention was to examine the effects of each of the factors independently from one another, which is why deionised water was used for both the temperature and salinity experiments. Due to possible buffering capacity of natural water as opposed to deionised water however, it was decided that a more accurate result may be extracted through using natural waters in the pH experiments. We agree that likely interactions may have

influenced by temperature. In addition responses may be different under turbid conditions.	been overlooked in this approach, and have added to the recommendations for future researchers regarding this concern in the conclusion.
6. (Materials and Methods, Section 2.4 Field methodology) I recognize that flow data are unavailable, but a general description of flow conditions (ie low, moderate, high, runoff) in the piped drainage network during the experiment would be useful to the reader.	Have added this information.
7. (Results and Discussion, Section 3.1 Stability of fluorescence) It is worthwhile to point out that response was generally similar between the DI and Swan River waters, suggesting little fluorescence interference due to natural environmental conditions (at least for the water sample used in the experiment). The only exception is perhaps the 10 ug/L Swan sample (note this concentration is similar to background observed in Whaleback Lake), which as the authors point out is likely explained by the lower signal:noise ratio in this sample.	Agree. Have altered this section accordingly.
8. (Results and Discussion, Section 3.6 Field release, lines 18-24) While the first release was only an order of magnitude greater than the background levels, it was still ~6-fold greater than background. Based on the lack of variability in the ambient data presented and the performance of the 6136 in the laboratory, the authors can state with some confidence that both peaks were the result RWT addition and not background fluctuations.	Agree. Have altered this section accordingly.
9. (Results and Discussion, Section 3.7 Background water quality data) I suggest presenting this information before section 3.6 to provide context for the reader.	Agree. Have moved this information to beginning of discussion section, and changed Figure 9 to become Figure 2.
10 Figures 5-6. The reduced variance around the 0 temperature difference in clearly evident in these graphs; consider discussing this point in the text.	Did not include this point.
Referee #1 Technical Comments	
1. In situ should be italicized throughout the text.	Agree. It is italicised in the Word document but did not appear to carry through to the PDF.
2. (Abstract, page 4536, Lines 15-17) The last sentence of the abstract is somewhat confusing. Suggest the following modification: The field release study succeeded in detecting RWT at concentrations two orders of magnitude greater than background fluorescence. Based on longitudinal dispersion theory, observed RWT peak concentrations were within 10% of predicted peaks.	Agree. Have changed accordingly.
3. (Introduction, page 4536, line 20) Suggested modification "...has become	Agree. Have changed accordingly.

widespread in most developed areas of the world."	
4. (Introduction, page 4536, line 24) Suggest removing "in water bodies"	Agree. Have changed accordingly.
5. (Introduction, page 4537, line 25) Need a comma after "factors"	Agree. Have changed accordingly.
6. (Results and Discussion, page 4545, line 12) Need a comma after "(Shiau et al., 1993)"	Agree. Have changed accordingly.
7. (Results and Discussion, page 4545, line 20) Need commas before and after "however" (: : :pH must, however, be: : :)	Agree. Have changed accordingly.
8. (Results and Discussion, page 4547, line 13) Suggest replacing "significantly" with "substantially" in this context.	Agree. Have changed accordingly.
9. (Results and Discussion, page 4547, line 19) Need a comma after "(equivalent to 0.012 N NaCl)"	Agree. Have changed accordingly.
10. (Results and Discussion, Section 3.5 Salinity effects) The discussion of the Smart and Laidlaw (1977) studies is somewhat confusing; clarify what the differences were in the two studies they conducted that had contradictory results.	Have reworded for clarity.
11. (Results and Discussion, Section 3.6 Field release, line 10) Clarify that the peak concentration given for the 2nd release is for the first peak.	Have reworded to clarify that the second release has the clear double peak.
12. (Results and Discussion, Section 4. Conclusions, page 4550, lines 11-15) This is a long sentence. I suggest modifying as follows: The rapid changes in water quality at the study site, coupled with the potentially significant effects of local water quality conditions on detected RWT concentration highlight the value of in situ fluorometric methods to quantitative release studies; the researcher can assess the measured concentration against real-time water-quality conditions.	Agree. Have changed accordingly.
13. Figure 1. Symbols indicating detection and release points should be more distinctive.	Increased size of these symbols.
14. Figure 4. The reverse order scaling on the x-axis is not intuitive and may be initially confusing to some readers.	Have added note into reference to figure to alert readers.
15. Figure 5. At first glance, this graph appears to show an inverse relation between temperature and fluorometric response. The fact that the graph is showing calibration temperature – sample temperature really needs to be emphasized to ensure readers interpret the graph properly.	Agree, have added note into reference to figure to alert readers.
16. Figure 8. Suggest changing figure description to: Continuously measured RWT concentration at Whaleback Lake: : :	Agree. Have changed accordingly.
17. Figure 9. Suggest removing ORP from the figure because it is not	Agree. Have changed accordingly.

discussed in the text.	
Referee #2 General Comments	
The lack of flow measurements in the field experiment, which is acknowledged by the authors, means that no mass balance is possible. All the calculations from that point onwards are circular and this lack of flow data impacts on the authors' ability to meaningfully discuss the quantitative use of RWT. A flow gauging, if a continuous flow record is not possible, is usually reasonably easy to carry out and should have been part of the study.	We agree that flow gauging would have been beneficial to the study, as demonstrated by our recommendation that future studies involve the concurrent collection of flow measurements. Whilst the lack of flow data does impact upon the degree to which the quantitative application of RWT is performed in this particular study, the methodology has however been successfully demonstrated and is anticipated to be built upon by future researchers.
The multiple peaks observed in the field experiment were stated to be either a result of the tracer properties (2 isomers with different sorption properties) or a result of the drainage network. The authors leave the issue unresolved which I do not think is acceptable for a manuscript claiming to use RWT in a quantitative way. As a tracer, RWT is conservative in some environments and non-conservative in others and in most studies using RWT as a tracer, which I have carried out and are aware of, the first experiment involves comparison of RWT with a truly conservative tracer such as Br or tritiated water. It is very straightforward to use another tracer, such as Br, in conjunction with RWT to determine what is due to tracer behaviour and what results from the system being studied.	We recognise the importance of these comments, and agree that it may be necessary for future studies to include this step in order to meaningfully apply RWT in a truly quantitative manner. This is likely to be a site-specific requirement, given that indeed as you say "RWT is conservative in some environments and non-conservative in others".
I would like to see these concerns addressed before the manuscript is published. This would probably involve some additional experimental work. If this is not possible then the title needs to be modified as it is no longer a "quantitative" use of RWT.	Given the importance of these comments, and the fact that further experimental work is not possible, the title has been modified accordingly.
Referee #1 Specific Scientific Comments	
1. (p 4539, line 6) The excitation and emission wavelengths used to measure the RWT should be given.	Agree. Have changed accordingly.
2. (p 4539, line 25) The detected salinity and specific conductance should be less than a detection, rather than stated to equal to zero.	Agree. Have changed accordingly.
3. (p 4541, line 27) Mention of what measures were taken to ensure mixing of the RWT dye in the pipe and an assessment of how well-mixed the dye was by the time it travelled 160 m or 260 m to the measuring point.	Additional information added.
4. (p4544, line 8-10, also fig 2) Most of the variation from the expected concentration in the 10 µg/L sample in Swan River water would be due to background fluorescence equivalent to 5-6 µg/L. You talk about it for the	Have added this possible interference factor into this discussion.

field experiment but it also affects this lowest concentration for the laboratory study.	
5. (p 4561, fig 8 caption) The last sentence should read “mean background fluorescence value equivalent to a RWT concentration of 6.7 µg/L)”	Agree. Have changed accordingly.