Response to Review Comments (hess-2021-353)

Response to the Editor

Our responses are in blue and revisions are underlined.

1. I would like to commend the authors for their efforts to address the valuable comments that were received form the two reviewers. Given the extent of the revisions requested, I would appreciate if the authors would submit the revised paper that incorporates the changes they promised.

We have substantially revised the manuscript as promised. Please see below for our point-to-point response to the comments from both reviewers, along with our corresponding revisions.

2. I would also recommend that the STAN model be included in the supplementary material.

We have added the stan model codes to Figures S10-11 in the Supplementary Material.

3. My final recommendation is for all the uncertainty bounds in the figures be defined. I also have concerns about the weak slopes that appear to be influenced by a few high leverage points.

We have revised the figure captions to clarify the uncertainty in all modelled results, as:

- ***Figure 5. Modelled effects of BFI_m and BFI_range on catchment C-Q slopes for each climate zone ($\delta_{BFI\_climate}$) for each water quality parameter. The bars show the 95% credible intervals (the range between 2.5th to 97.5th percentiles of Bayesian posterior distribution) of the modelled effects, and the dots indicate the corresponding median levels. The colours indicate whether an effect is significantly positive (red), significantly negative (blue), or non-significant (grey); a positive effect means that the C-Q slope increases with a higher catchment BFI_m or BFI_range, and vice versa. Black dashed lines show the zero-effect i.e., no effect at all. The plot includes results from models with each of BFI_m and BFI_range as the key predictor, which are differentiated by marker shapes.***

- ***Figure 6. a) Catchment C-Q slope vs. BFI_m and b) Catchment C-Q slope vs. BFI_range, coloured by climate zones. The lines represent the modelled C-Q slope~BFI_m or C-Q slope~BFI_range regression lines for individual climate zones. The bands represent the 95% credible interval (the range between 2.5th to 97.5th percentiles of Bayesian posterior distribution) of the modelled C-Q slopes. The dots represent the 'true' C-Q slopes estimated with C-Q observations at individual catchments. The black dashed lines mark a zero C-Q slope which differentiate mobilisation (C-Q slope>0) from dilution (C-Q slope<0).***

We also added clarification on model uncertainty in text and discussed its implication in Section 3.3 as:

- *L327: "Figure 5 presents the median and the 95% credible intervals of these modelled effects for each water quality variable. The 95% credible interval is the range between the 2.5th and 97.5th percentiles of the posterior distribution of the parameter values, which was derived from the Bayesian posterior estimates of $\delta_{BFI\_climate}$ (Eqn. 3) to quantify the uncertainty in the modelled effects (Gelman et al., 2013). The effects of BFI_m and BFI_range on the C-Q slopes are almost always significant, with the 95% credible intervals not crossing over 0 for most combinations of water quality variables and climate zones."*

Responses to Reviewer #1

Our responses are in blue and revisions are underlined.

Summary:

1. Guo et al. investigate the variability in C-Q relationships in relation to the catchment hydrological conditions (more specifically the BFI) for several water quality parameters across several climate regions in Australia. The authors make use of an impressive data set from an arid region and apply a Bayesian Hierarchical Approach including the BFI, which allows understanding spatial patterns

of export dynamics. This study can thus provide an important contribution for understanding solute transport beyond temperate regions. However, the manuscript still needs more clarity on the research questions and key messages and methodological improvements. I would suggest the manuscript for major revisions as substantial improvements are still necessary.

Thank you for acknowledging the value of the study, and your comprehensive suggestions for further improvement. We provide below a point-to-point response to your comments, along with our corresponding revisions.

General comments:

2. One of my concerns is that given the fact that previous studies in the same region, using the same dataset and similar methods (as I read from the text), are not accessible or provided (under review or in preparation), it is not possible to judge the additional value of this study. The preceding studies (Lintern and Liu) are referenced both when defining the research goals and in the method sections. I definitely see the value of investigating C-Q relationships in various climate zones, but this was also done by these referenced studies. It is hard to judge the additional value without knowing what was shown already.

We believe this comment refers to L65-69 in the Introduction of the original manuscript, which summarized our preceding studies, Lintern et al. (2021, recently published) and Liu et al. (in preparation). Both studies focused on how C-Q relationships vary across the Australian continent. Considering the paper timelines, we decided to keep only Lintern et al. (2021) here and add a summary of this study in the revised manuscript as:

- *L79: "The current knowledge gap in understanding catchment export regimes for regions other than Europe and North America was partially addressed in Lintern et al. (2021), which focused on differences in water quality status and the C-Q relationships across different climate zones in the Australian continent. One remaining question that Lintern et al. (2021) highlighted is our lack of understanding of the substantial variations in C-Q relationships within each climate zone."*

We also added the following discussion on new insights added by this current study to the end of the Introduction, as:

- *L99: "We use a subset of the grand dataset that Lintern et al. (2021) used, which enables us to focus on representative catchments with water quality records captured under a wider range of flow conditions."*

3. The motivation of investigating BFI impact on C-Q relationships was not convincing for me. It needs to be clear 1.) why we need to know that and 2.) what exactly we do not know yet. The first question is not satisfyingly presented: Why do you want to focus on BFI, why is it useful to investigate this relationship? For the second: From my knowledge and in contrast of what you state (see also my comments below), the influence of BFI on the spatial variability of C-Q relationships has been discussed in several previous studies. However, I agree that studies have been biased towards temperate climates. I think the latter should be the main motivation, while generally the literature review on the control of BFI needs to be extended. It is not right, that is has not been investigated. There are studies using BFI as a descriptor for explaining the variability in export behaviour of different solutes, including several studies that you have cited in the introduction but considering other statements. For example, Minaudo et al. 2019 stated, "we found for NO_3^- that high BFI values, low W2, and low erosion differentiated C-Q dilution patterns from non-significant and mobilization types". But also Ebeling et al. 2021, Moatar et al. 2017, Musolff et al. 2015 have used the BFI to explain variability in C-Q relationships among catchments

for several solutes. Moatar et al. 2020 also investigated the impact of discharge flashiness on C-Q slopes and subsequently load flashiness. As BFI and Q flashiness are closely linked, this needs to be mentioned in the introduction. These also need to be discussed in relation to your study in the discussion. Also see further comments below

This study is important because understanding the impact of baseflow contribution on C-Q relationships can help explain both the spatial and temporal variation of C-Q relationships, and to infer key transport pathways.

We agree with the reviewer that previous studies have investigated how variation in baseflow contribution affects C-Q relationships across multiple catchments. The link between C-Q patterns and interannual baseflow index (or similar metrics) were well studied (e.g., Moatar et al. 2017, 2020, Musolff et al. 2015, Ebeling et al. 2021), while Minaudo et al. (2019) explicitly explored the link between instantaneous BFI and C-Q relationships. However, the existing studies on the impact of baseflow contribution are largely focused on the temperate climate. Therefore, we haven't yet well understood the effects of baseflow contribution across a wider range of climate zones, nor been able to quantify this impact.

The key motivation of this study is to understand and quantify how catchment-level metrics of baseflow contribution affect C-Q relationships across catchments in a wide range of climate zones. Through synthesizing continental water quality data, we were able to gain large-scale understanding of how C-Q changes in catchments with different baseflow conditions.

Our specific revisions to clarify these include:

1) Added the following sentences to the end of the Introduction to highlight the importance of the key issue that this study explores:

- *L101: "By analysing the impacts of baseflow contributions on C-Q relationships, this study will i) explain the variations in C-Q relationships within individual climate zones; ii) broaden the existing knowledge of how baseflow contribution impacts C-Q relationships to a wider range of climate conditions, and thus infer key constituent transport pathways in different climate zones."*

2) Revised the literature review and add discussions on the state-of-art research on the impacts of baseflow contribution in the Introduction:

- *L59: "Prior studies have explored the links between C-Q relationships and baseflow index (BFI) and similar hydrological metrics at an interannual scale (e.g., Ebeling et al., 2021; Moatar et al., 2017; Musolff et al., 2015) or at the scale of storm events (e.g., Knapp et al., 2020; Minaudo et al., 2019; Musolff et al., 2021). Across both long and short timescales, a consistent finding is that, within a particular catchment, the C-Q relationship (and thus export behaviour) is dependent on whether streamflow is dominated by baseflow or quickflow, i.e., the baseflow contribution to total flow (Gorski & Zimmer, 2021; Knapp et al., 2020; Minaudo et al., 2019). These studies also identified baseflow contribution as a key driver of the variation in C-Q relationships across catchments (Musolff et al., 2015; Moatar et al., 2017). For example, Knapp et al. (2020) found that for solutes that are partly derived from atmospheric inputs, such as nitrate and chloride, mobilisation behaviours (i.e., positive C-Q slopes) often occur during events with drier antecedent conditions. For nitrate, baseflow contributions can further affect the C-Q relationships via changing the connectivity between surface flow and groundwater (Minaudo et al., 2019). Baseflow variation also affects the capacity of nutrient removal via changing the relative importance of hydrological and biogeochemical processes (Moatar et al., 2017). Further, the variation in the baseflow contribution of a catchment is also a key feature that can be linked to the shift between different* *dominant flow paths during low- and high-flow (e.g., von Freyberg et al., 2018), leading to contrasting sources and mobilisation behaviours for solutes and particulates."*

3) Improved clarification of the knowledge gap on the lack of understanding of how C-Q relationships vary with baseflow contribution across a wide range of climate zones.

- L73: *"Although a substantial body of knowledge has been established on the impact of baseflow contributions on C-Q relationships, the existing studies have largely focused on catchments in temperate climates in Europe and North America (Knapp et al., 2020; Gorski & Zimmer, 2021; Minaudo et al., 2019; Musolff et al., 2015). The narrow range of climate conditions explored so far implies a potential limitation in transferring and systematically comparing new findings to other climate zones and other parts of the world, because climate is proven a key control of the hydrological regime, especially regarding the baseflow contribution and flow paths of individual catchments (Beck et al., 2013; von Freyberg et al., 2018)."*

4.

a) Some of the methods seem inappropriate, especially as there is too few data for some climate-solute combinations to fit robust models/regressions and interpret them (further comments below).
b) Besides, I do not see the value of investigating the BFI impact within each climate zone individually, i.e. separating the climate zones and fitting different models, instead of investigating the BFI impact across the whole climate variability. I think it would be more valuable to know what effect the BFI has across the whole climatic variability, i.e. the continuum of variations. The climate zones, could be represented by their characteristics such as precipitation amount, seasonality, aridity, temperature etc.. Even within the climate zones those variables vary and could potentially explain the deviations not explained by BFI.

a)

In this study, we used a single Bayesian Hierarchical Model (BHM) to fit all data points for each constituent simultaneously, across all climate zones. Although we used different parameters for individual climate zones, this model structure has a great advantage of 'borrowing power', meaning that information is shared across climate zones, and parameter fitting for one climate zone can utilize data from all other climate zones. As illustrated in Figures 5 and 6 (both in the original and revised manuscripts), BHM has been effective in estimating the impact of baseflow contribution (summarized by catchment median BFI and range of daily BFI) on C-Q slopes even for climate zones with limited data, while acknowledging the model uncertainty. This model enables us to estimate the BFI impacts for climate zones with limited data by making use of data from other climate zones; in this way we made the best use of all available data.

To address this comment, we added further discussion to both the Introduction and the Method Section 2.2 (which introduces the modelling approach) on the key advantages of BHM in effectively handling data-limited situations and spatio-temporal data with uneven coverage, and justifications for why BHM is suited to the dataset analysed in this study, as:

- L96: *"We answer our research questions and test our hypotheses with a Bayesian hierarchical model (BHM) (Gelman et al., 2013), which is an integrated framework that enables sharing information across catchments to strengthen the statistical power of explaining variation in individual catchments. The model is a powerful approach to capture water quality variability across catchments of varying conditions and record lengths, which is the case for Australian water quality data (Guo et al., 2019, 2020; Liu et al., 2021)."*

- L168: "The key reason for choosing this model is the high heterogeneity in the national C-Q dataset in both the record period and the representation of individual climate zones, as illustrated in Section 2.1.1. BHM is effective in handling data-limited situations via its 'information sharing' or 'borrowing power' across space (Gelman et al., 2013; Webb & King, 2009), which has been shown to be highly effective in explaining variability in spatial-temporal data under data-limited situations. This has been highlighted in several recent studies in modelling water quality over large regions in Australia (Guo et al., 2019, 2020; Liu et al., 2021). Another advantage of BHM is the ability to account for uncertainty, which is especially important for analysing water quality data, as these data are often associated with high uncertainty due to sparse sampling of the natural variability of chemical species in river flow (Guo et al., 2020; Liu et al., 2021)."

The following references were added to the manuscript support the above arguments:

- Guo, D., Lintern, A., Webb, J. A., Ryu, D., Bende-Michl, U., Liu, S. & Western, A. W. (2020). A data-based predictive model for spatiotemporal variability in stream water quality. Hydrology and Earth System Sciences, 24(2), pp. 827-847.doi:10.5194/hess-24-827-2020
- Guo, D., Lintern, A., Webb, J. A., Ryu, D., Liu, S., Bende-Michl, U., . . . Western, A. W. (2019). Key Factors Affecting Temporal Variability in Stream Water Quality. Water Resources Research, 55(1), 112-129. doi:10.1029/2018wr023370
- Liu, S., Ryu, D., Webb, J. A., Lintern, A., Guo, D., Waters, D., & Western, A. W. (2021). A Bayesian approach to understanding the key factors influencing temporal variability in stream water quality – a case study in the Great Barrier Reef catchments. Hydrol. Earth Syst. Sci., 25(5), 2663-2683. doi:10.5194/hess-25-2663-2021

b)

The reason to investigate BFI effects for individual climate zones separately is to see whether we could identify any statistically significant differences of the effects for different climate zones, and the results suggested that this is the case (from the C-Q slopes in Figure 5 and the simulated C-Q~BFI relationships in Figure 6, in both the original and revised manuscripts). If there is no significant difference of the BFI effects between climate zones, the model is also capable of indicating this – as would be shown with similar, undistinguishable modelled effects of BFI for individual climate zones. Our current model structure enables us to test this hypothesis, since we are essentially developing different models for each climate zone and comparing them within one larger modelling framework. In contrast, a model with only a single parameter for the BFI effects across all climate zones cannot be used to test this hypothesis. We added the following sentences to clarify this in Method Section 2.2, which introduces the modelling approach to exploring the BFI effects:

- L189: "We chose to investigate the effects of baseflow contributions for individual climate zones separately to identify any statistically significant differences of these impacts between climate zones. If there is no significant difference between climate zones, the model is also capable of indicating this – as would be shown with similar, undistinguishable modelled effects of baseflow contribution for individual climate zones. Thus, our BHM incorporates different models for individual climate zones and compares them within one comprehensive modelling framework."

To illustrate the value added by considering climate-specific effects of BFI, we replaced the previous results in Section 3.3.1 (now as part of Section 3.3) with a new comparison of performance between the lumped and climate-specific models. The key results added are included in the revised Table 1 as below:

Table 1. Performance of the BFI-based C-Q models – the columns show four alternative model structures with *BFI_m* or *BFI_range* as the key predictor, and with the impacts of baseflow contribution considered as lumped or specific to individual climate zones. The rows show results for individual water quality parameters. All model performances are summarised by R^2, which quantifies the percentage of variance in C-Q slopes explained by the BFI-based models.

WQ parameter	Median C-Q slope	Current (climate-specific impacts)		Baseline (lumped impact across climate zones)	
		BFI_m	*BFI_range*	*BFI_m*	*BFI_range*
TSS	0.15	0.16	0.11	0	0.04
TP	0.09	0.14	0.17	0	0.08
SRP	0.06	0.02	0	0.03	0.05
TN	0.09	0.18	0.12	0.02	0.03
NOx	0.36	0.22	0.18	0.03	0
EC	-0.07	0	0.01	0	0.01

We added new interpretation of these new results as:

- *L305: "Using catchment-level metrics of baseflow contribution alone (either BFI_m or BFI_range) can explain up to 22% of the variation in catchment C-Q slopes. Although these results represent limited model predictive capacity, the model does cover a large range of catchment conditions such as contrasting land uses and hydro-climate conditions. Therefore, the amount of variation that can be explained by a single BFI metric highlights baseflow contribution as an important factor that influences catchment C-Q relationships. Further, it is also worth highlighting that incorporating climate-specific impacts of baseflow contribution is highly beneficial in explaining these variations. For all six water quality parameters, the baseline model – which uses a lumped effect of catchment baseflow contribution across different climate zones – can barely explain any variation in the C-Q slopes (with all $R^2 < 0.08$, i.e., <8% of the variation explained). In contrast, the climate-specific models generally offer up to 20% increase in the variance explained for C-Q slopes, except for EC and SRP, for which performance is equally low regardless of whether the effects of baseflow contribution are separated for individual climates. The low performances for EC and SRP are likely attributed to the smaller magnitudes of C-Q slopes as highlighted in the lower median C-Q slope in Table 1, making it statistically more difficult to explain variations across catchments for these two water quality variables. These results further emphasise that in general, the impacts of catchment baseflow contribution on C-Q slopes are better defined within individual climate zones, which confirms the validity of our BFI-based C-Q models (Eqn. 3)."*

We expect that the abovementioned revision also removes the previous confusion as highlighted in your Comments #24 and #26 (see details in our specific responses to those comments).

We agree with you that variations in the C-Q slopes can also be explored together with other climate drivers (e.g., temperature, rainfall, aridity), but a key challenge we see in other water quality modelling studies with these potential drivers is the high cross-correlation between these variables, thus we decided to use climate zones as an integrator of many hydroclimatic variables. The focus of this study is BFI and we hypothesise that BFI has a significant effect on C-Q slopes – which is supported by the study results.

To address this comment, we have added further justification to the use of climate zones in Section 2.2.

- *L189: "We chose to investigate the effects of baseflow contributions for individual climate zones separately to identify any statistically significant differences of these impacts between climate zones. If there is no significant difference between climate zones, the model is also capable of* *indicating this – as would be shown with similar, undistinguishable modelled effects of baseflow contribution for individual climate zones. Thus, our BHM incorporates different models for individual climate zones and compares them within one comprehensive modelling framework."*

We also added discussions on the need for further studies on the impact of individual climate drivers on C-Q slopes in the Conclusion.

- *L463: "This study used catchment-level metrics of baseflow contribution as the only predictor of C-Q slopes. The baseflow contribution alone can explain up to 22% variance in the C-Q slopes across the Australian continent. This highlights a substantial role in baseflow contribution in shaping the C-Q relationships, while also suggesting the need of further work to synthesise the impacts of baseflow contribution together with other spatial drivers (e.g., climate, land use, land cover and geology) to include their interactions and establishing their relative importance on influencing C-Q relationships."*

5. The interpretation of BFI_m in terms of variability of flow paths is not convincing to me as you could easily and more directly use the range of BFI to determine the relationships between C-Q slope variability with BFI ranges. I think it would be good to look at this instead of speculating, as you have the data at hand and Figure 3b is not convincing enough for this interpretation, in my opinion. Instead of the range BFI_h-BFI_l you could also consider other metrics of variability.

To address this comment, we have added substantial new analyses to explore the impact of catchment BFI range on the C-Q slopes. Specifically, we explored a new model with the same structure as our original model based on *BFI_m*, which is now based on *BFI_range* (i.e. the difference between the 10th and 90th quantiles of daily BFI). We revised Figures 5 and 6 to include results from both models, which illustrated that the effects of *BFI_range* on C-Q slopes are highly consistent with the modelled effects of *BFI_m* on C-Q slopes. These new results provide more concrete evidence on our previous 'speculation' on how the BFI range in a catchment can impact its C-Q slope.

According to these new results, we have substantially revised the discussions of these results which are presented after Figures 6 and 7 in Section 3.3.

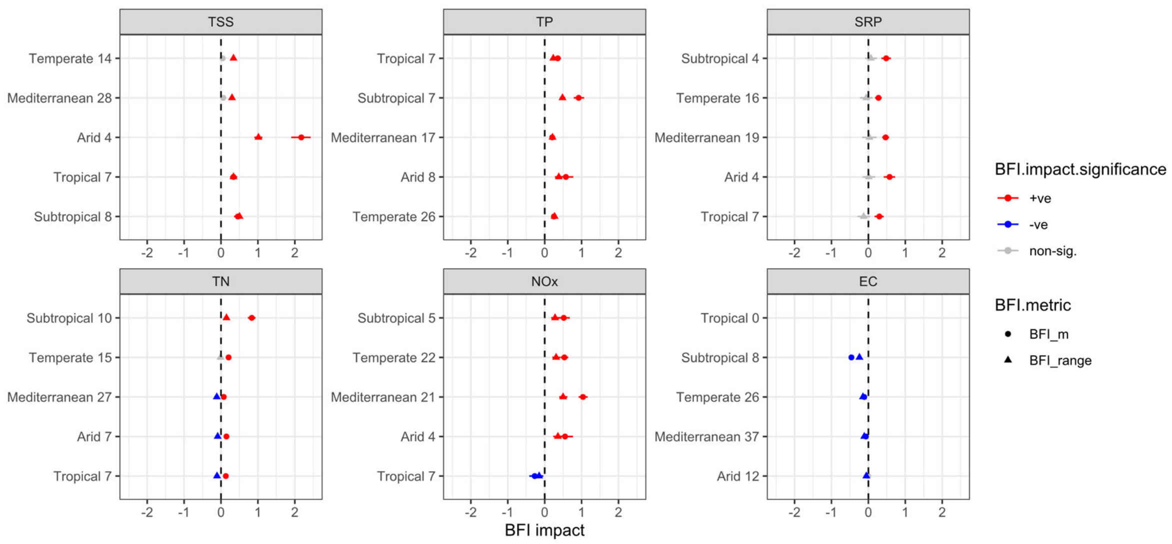


Figure 5. Modelled effects of *BFI_m* and *BFI_range* on catchment C-Q slopes for each climate zone ($\delta_{BFI\_climate}$) for each water quality parameter. The bars show the 95% credible intervals (the range between 2.5th to 97.5th percentiles of Bayesian posterior distribution) of the modelled effects, and the dots indicate the corresponding median levels. The colours indicate whether an effect is significantly positive (red), significantly negative (blue), or non-significant (grey); a positive effect means that the C-Q slope increases with a higher catchment *BFI_m* or *BFI_range*, and vice versa. Black

dashed lines show the zero-effect i.e., no effect at all. The plot includes results from models with each of *BFI_m* and *BFI_range* as the key predictor, which are differentiated by marker shapes.

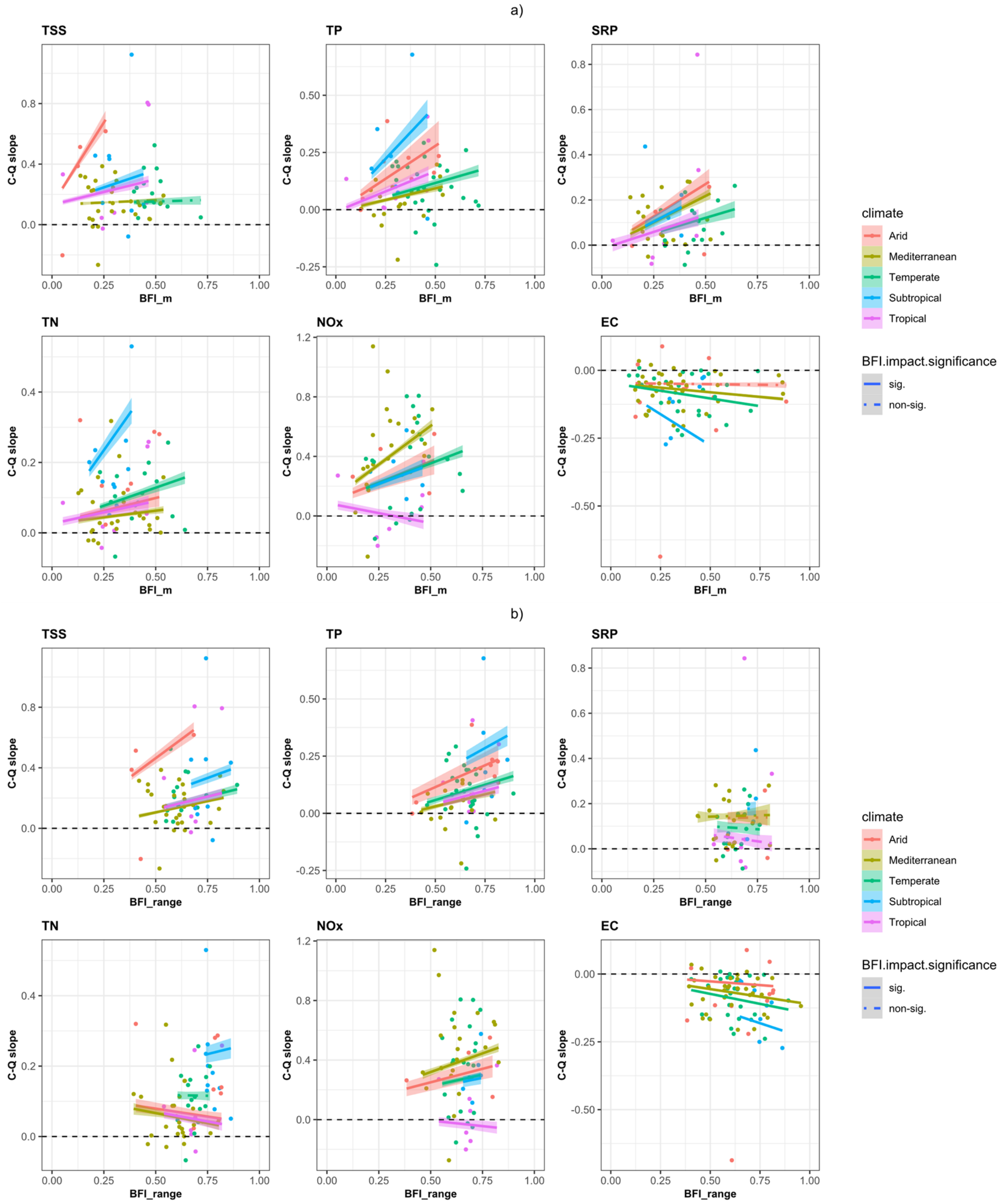


Figure 6. a) Catchment C-Q slope vs. *BFI_m* and b) Catchment C-Q slope vs. *BFI_range*, coloured by climate zones. The lines represent the modelled C-Q slope~*BFI_m* or C-Q slope~*BFI_range* regression lines for individual climate zones. The bands represent the 95% credible interval (the range between 2.5th to 97.5th percentiles of Bayesian posterior distribution) of the modelled C-Q slopes. The dots represent the 'true' C-Q slopes estimated with C-Q observations at individual catchments. The black dashed lines mark a zero C-Q slope which differentiate mobilisation (C-Q slope>0) from dilution (C-Q slope<0).

We have also further investigated the non-linear relationship between *BFI_m* and the *BFI_range* beyond the previous manuscript. We have updated Figure 3b) as below, along with the interpretation in text:

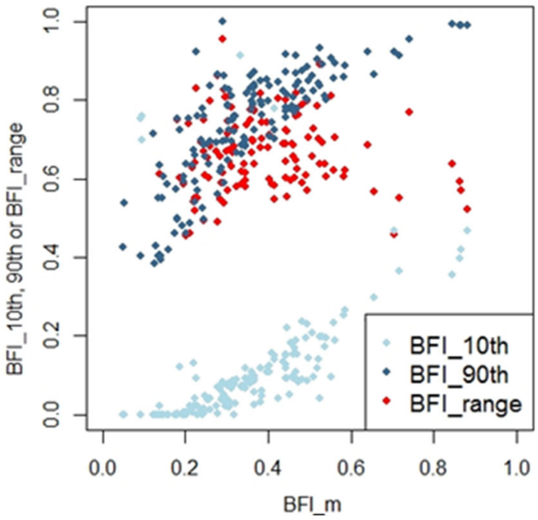


Figure 3 b). the 10th and 90th percentiles of daily BFI (BFI_10th and BFI_90th), and *BFI_range* (BFI_90th – BFI_10th) versus *BFI_m*.

- *L245: "Generally, temperate catchments have the highest BFI_m, while similar BFI_m values are seen across the other four climate zones. BFI_10th and BFI_90th have distributions consistent with BFI_m in all climate zones. As different catchments were analysed for each water quality variable, the same BFI metrics were also generated for each water quality variable, and their distributions are generally consistent across different variables (Figure S4, Supplementary Materials)."*

- *L256: "In general, catchments with high median BFI are likely to have a greater range of variation of daily BFI, as highlighted by the generally increasing BFI_range with higher BFI_m (Figure 3b), Spearman's ρ = 0.33). The link between BFI_m and BFI_range suggests that catchments with higher BFI_m values are more likely driven by highly variable flow pathways. Specifically, a catchment with a low BFI_m tends to be associated with a small range of daily BFI (low BFI_range); thus, the catchment is likely to always have constantly low contributions of baseflow and higher contributions of quickflow, during both dry and wet conditions. In contrast, a catchment with a high BFI_m generally has a large range of daily BFIs (high BFI_range). This means that the catchment is more likely to switch between groundwater contributions in dry conditions (high daily BFI) and surface water contributions during wet conditions (low daily BFI). However, we also note that a small proportion of catchments (9 catchments) with the highest BFI_m (>0.6) actually have smaller BFI_range compared to other catchments with mid-range BFI values (0.4-0.6). This is a result of BFI_10th and BFI_90th both increasing with BFI_m, while the increase in BFI_90th plateaus at high BFI_m. This nonlinearity suggests that the full distribution of catchment baseflow contributions might not be sufficiently represented by either the BFI_m or BFI_range alone, providing further justification for the need to explicitly consider both the overall condition and the variation in catchment baseflow contributions when studying their effects on C-Q relationships."*

6. Linked to the methodology, I have a concern about the conceptualisation in Figure 7 and main conclusions. I think some methodological approaches and results/evidence are not robust and clear enough to generalise the results in the given way.

Figure 7 has been heavily revised (see below) to incorporate our new results with the model that focuses on the impact of *BFI_range* on catchment C-Q slopes (as detailed in our response to your Comment #5). We have also revised the organization of the figure to further improve the clarity and simplicity.

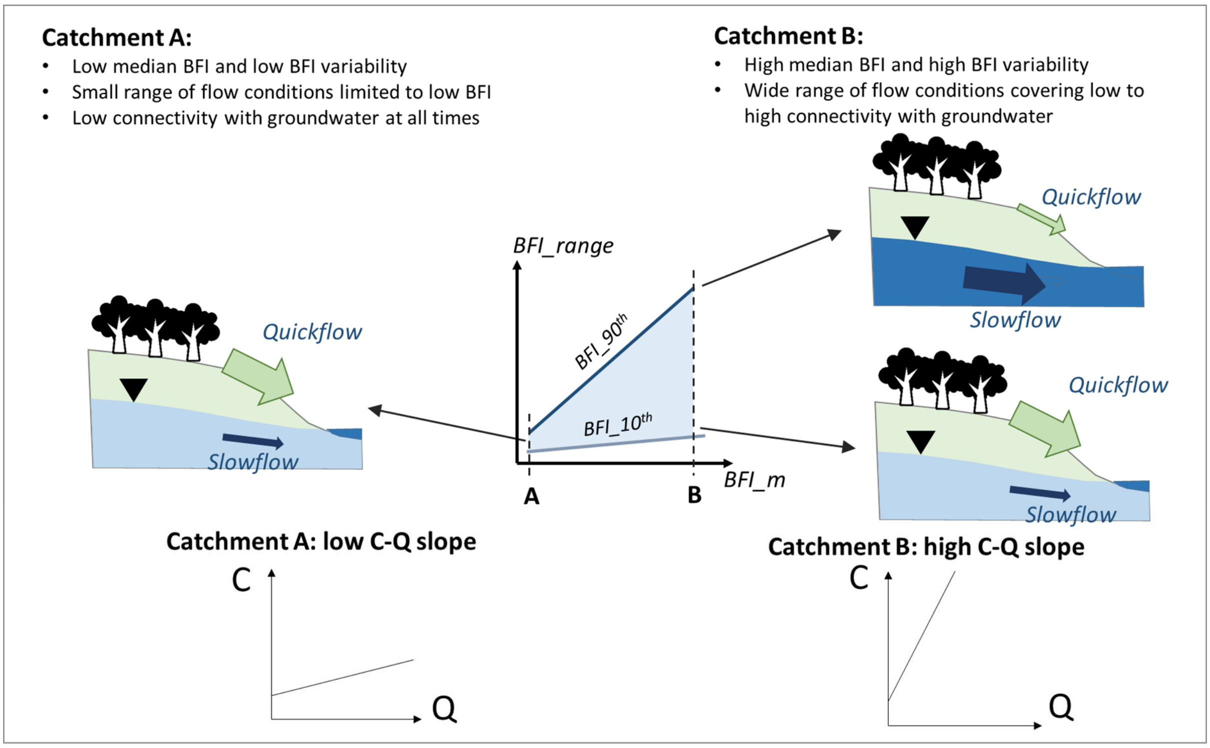


Figure 7. Conceptual diagram of the expected hydrological conditions in catchments with low and high median and variability in baseflow contribution (*BFI_m* and *BFI_range*), as Catchments A and B, respectively. The contrasting hydrological conditions can help explain our modelled results of the impacts of baseflow contributions on C-Q slopes. Note that the C-Q intercepts in the plots are not indicative since we do not investigate the variation in C-Q intercepts in this study.

7. The discussion misses comparison to relevant previous studies. Previous studies investigating hydrological controls (such as the BFI, flashiness etc.) spatial variability of C-Q relationships. The discussions needs extension

To address this comment, we added some in-depth discussion in Section 3.3 on the relevant findings in previous studies, especially the various hydrological controls of C-Q relationships and how they vary across catchments. We highlighted the key findings of these studies, compared our results to the existing studies, and discussed the added value of our work.

- *L379: "The enhanced mobilisation of particulates (TSS and TP) with higher BFI_m is consistent with previous studies in European catchments, which also reported positive effects of BFI on the C-Q slopes of TSS (Moatar et al., 2017; Musolff et al., 2015). However, no physical interpretation of this result was discussed previously. Combining our modelled results of BFI_m together with those of BFI_range, we are able to draw a plausible explanation that links particulate mobilisation with the two highly correlated baseflow metrics (Figure 3b)."*

- *L408: "Catchments with a greater variability in baseflow contribution (Catchment B, Figure 7) are likely to have greater gradients of concentrations for soluble N, between low groundwater-fed concentrations at low flow and high concentrations from surface runoff and/or interflow at high flow (e.g., via leaching), resulting in a stronger mobilisation pattern as illustrated with a higher C-Q slope. Further, a typical temporal pattern of nitrate leaching in Australian catchments is the accumulation of N in soils during periods of low soil water drainage, followed by strong export during high drainage (Drewry et al., 2006), which is also more likely to occur in catchments with greater variation in baseflow contributions."*

- L435: *"…future studies should also consider more broadly the temporal variations in flow regime and baseflow condition and their influences on C-Q relationships. For example, seasonality can play a big role in shaping the C-Q relationships for nutrients, as these relationships over time during the build-up of pollutant sources, and during the flushing of readily available sources at the onset of high flow periods (Bende-Michl et al., 2013). Besides, anthropogenic disturbances and/or management actions in the catchment can cause changes in C-Q relationships over time (Zhang, 2018). Flow flashiness is also shown to influence the C-Q relationships, which differ across particulates and solutes, and across natural and highly regulated catchments (Moatar et al., 2020)."*

Specific comments:

8. L17: Does the baseflow contribution in a catchment impact the concentration itself or the C-Q relationship? For me, it seems like spatial and temporal dimensions are mixed up here.

Here we aim to provide a general statement of the importance of understanding C-Q relationships. For generality and clarity, we removed the 'spatial and temporal variation' and revised the sentence as:

- L15: *'Understanding the concentration-discharge (C-Q) relationships can inform solute and particulate export processes'*

9. L18: This is not true "these patterns have not yet been investigated across large spatial scales", e.g. Minaudo et al. 2019 (see also related comments)

We revised this sentence as:

- L16: *"Previous studies have shown that the extent to which baseflow contributes to streamflow can affect C-Q relationships in some catchments. However, the current understanding on the effects of baseflow contribution in shaping the C-Q patterns is largely derived from temperate catchments. As such, we still lack quantitative understanding of these effects across a wide range of climates (e.g., arid, tropical and subtropical)."*

10. L48: "variable, which" reference is unclear, you mean here the studies? Please revise

We started a new sentence from "which", as:

- L50: *"These studies highlighted a number of critical drivers for these spatial variations, such as land use, land management, lithology and topography (e.g., Ebeling et al., 2021; Minaudo et al., 2019)."*

11. L 57: This sentence needs revision. The concentration variability within one catchment regarding the contribution of baseflow or quickflow to the current discharge is represented by its C-Q relationship, i.e. the variability "within a particular catchment". However, the cited studies also investigate the differences in C-Q relationships among catchments. Therefore, the provided references do not fit to this sentence, in my opinion. This also leads to the next sentence being incorrect. It defines the research gap as the differences among the catchments regarding the hydrological average behaviour not being investigated and understood. E.g. Minaudo et al. 2019 (others, see main comment) considered BFI in the analysis of C-Q relationship variability among catchments.

We agree with the reviewer that previous studies have investigated how variation in baseflow contribution affects C-Q relationships across multiple catchments. As in our response to your Comment #3, we acknowledge the key gap in the existing studies is the lack of understanding of the effects of baseflow contribution on C-Q slopes across a wider range of climate zones, as well as the lack of quantification of the impact of baseflow. The key motivation of this study is thus to understand and quantify how catchment-level metrics of baseflow contribution affects C-Q relationships across catchments in a wide range of climate zones.

To address this comment, our specific revisions include:

1) Added the following sentences to the end of the Introduction to highlight the importance of the key issue that this study explores:

- *L101: "By analysing the impacts of baseflow contributions on C-Q relationships, this study will i) explain the variations in C-Q relationships within individual climate zones; ii) broaden the existing knowledge of how baseflow contribution impacts C-Q relationships to a wider range of climate conditions, and thus infer key constituent transport pathways in different climate zones."*

2) Revised the literature review and add discussions on the state-of-art research on the impacts of baseflow contribution in the Introduction:

- *L59: "Prior studies have explored the links between C-Q relationships and baseflow index (BFI) and similar hydrological metrics at an interannual scale (e.g., Ebeling et al., 2021; Moatar et al., 2017; Musolff et al., 2015) or at the scale of storm events (e.g., Knapp et al., 2020; Minaudo et al., 2019; Musolff et al., 2021). Across both long and short timescales, a consistent finding is that, within a particular catchment, the C-Q relationship (and thus export behaviour) is dependent on whether streamflow is dominated by baseflow or quickflow, i.e., the baseflow contribution to total flow (Gorski & Zimmer, 2021; Knapp et al., 2020; Minaudo et al., 2019). These studies also identified baseflow contribution as a key driver of the variation in C-Q relationships across catchments (Musolff et al., 2015; Moatar et al., 2017). For example, Knapp et al. (2020) found that for solutes that are partly derived from atmospheric inputs, such as nitrate and chloride, mobilisation behaviours (i.e., positive C-Q slopes) often occur during events with drier antecedent conditions. For nitrate, baseflow contributions can further affect the C-Q relationships via changing the connectivity between surface flow and groundwater (Minaudo et al., 2019). Baseflow variation also affects the capacity of nutrient removal via changing the relative importance of hydrological and biogeochemical processes (Moatar et al., 2017). Further, the variation in the baseflow contribution of a catchment is also a key feature that can be linked to the shift between different dominant flow paths during low- and high-flow (e.g., von Freyberg et al., 2018), leading to contrasting sources and mobilisation behaviours for solutes and particulates."*

3) Improved clarification of the knowledge gap on the lack of understanding of how C-Q relationships vary with baseflow contribution across a wide range of climate zones.

- *L73: "Although a substantial body of knowledge has been established on the impact of baseflow contributions on C-Q relationships, the existing studies have largely focused on catchments in temperate climates in Europe and North America (Knapp et al., 2020; Gorski & Zimmer, 2021; Minaudo et al., 2019; Musolff et al., 2015). The narrow range of climate conditions explored so far implies a potential limitation in transferring and systematically comparing new findings to other climate zones and other parts of the world, because climate is proven a key control of the hydrological regime, especially regarding the baseflow contribution and flow paths of individual catchments (Beck et al., 2013; von Freyberg et al., 2018)."*

12. L91: "nitrate-nitrite" I am not sure what you mean with that. Is it the sum of both nitrate and nitrite concentrations? It should be defined once in the manuscript

Thank you for highlighting this, we added this clarification to the revised manuscript as:

- *L121: "... total suspended solids (TSS), total phosphorus (TP), soluble reactive phosphorus (SRP), total nitrogen (TN), the sum of nitrate and nitrite (NO_x) and electrical conductivity (EC)."*

13. L105 "unaffected" this is a strong word, I suggest to say "more robust"

We revised this phrase as:

- *L134: "...this ensures that the C-Q relationships observed are more robust against outliers..."*

14. L106: plural "span"

Thank you. We revised this phrase as:

- *L136: "Having water quality time-series that span at least 3 years..."*

15. L111: "met the above criteria across all the six water quality variables" for me this sounds as if the stations needed to meet the criteria for all the six variables, which was not the case from what I read in the next sentence and the following. I suggest to revise this formulation

We revised this sentence as:

- *L141: "We performed the above catchment selection for each water quality variable, and found a total of 157 sites (catchments)."*

16. L144: Does that mean you fit equation 1 only to baseflow discharge? How can this work, if C is a mix of baseflow and quickflow concentrations? This sentence in unclear, please revise.

No, Eqn 1 was fitted to all available pairs of C-Q data regardless of whether the flow is baseflow dominated or quickflow dominated. To clarify this, we revised the sentence as:

- *L179: "Our model is based on such a single C-Q relationship at each catchment. However, our model enables the slope term (βs) for individual catchments to change according to their baseflow contributions. This model conceptualisation is based on previous literature on the effects of baseflow contribution on C-Q slopes within individual catchments (Gorski & Zimmer, 2021; Minaudo et al., 2019), while aiming to further explore the impact of baseflow contributions on C-Q slopes across multiple catchments."*

17. L146: "the C-Q slopes of all catchments are following a normal distribution with a 'grand mean'" This works if the represented catchments cover the range of variability well. This would not be true if catchment types are overrepresented, would it?

The use of this 'grand mean' for each water quality variable is based on a finding from our preceding study, that the C-Q slopes for each water quality variable do not vary substantially across climate zones (Lintern et al. in review). We used this in our model to represent a baseline level of C-Q slopes for each water quality variable; whereas the C-Q slope of each catchment is the sum of the grand mean and the BFI effect (rather than sampled from a normal distribution), following Eqn 2. To clarify this, we revised this description to clarify the model structure as:

- *L185: "We assume that for each water quality variable, the C-Q slopes of all catchments have a 'grand mean', β_0. Then the variation of C-Q slopes between catchments, away from β_0, is explained by changes in the catchment baseflow contribution."*

As a further comment to the representativeness of catchments that we analysed, we acknowledge the lack of data for some regions due to limited monitoring (i.e., regions of low management prioritization/interest for monitoring, and regions inaccessible for monitoring). Further, climate zones and catchments are unequally distributed across Australia; catchments are more clustered towards the coastline as these areas have higher rainfall and presence of more perennial streams, while the large arid regions in the middle of the continent are dominated by ephemeral/intermittent streams. These mean that we expect the catchment representation to be biased towards regions that have more catchments and are monitored more frequently (e.g., in the coastal regions of Australia). This has already been discussed in Section 2.1.2 (which introduces the dataset), as:

- *L145: "Some sites are biased towards high flows, which is likely due to i) monitoring priority for high flow events to better represent export loads; ii) practical constraints to sample low flows in intermittent rivers and ephemeral streams."*

18. L146f: "Then the variation of C-Q slopes between catchments, away from beta_0, are explained by changes in catchment BFI. " This would only be true, if the BFI is the "only" controlling variable

We intend to use BFI as the only controlling variable because this study focuses on the impacts of baseflow contribution on the variation of C-Q slopes across catchments. We added a sentence within the model introduction (Section 2.2) to clarify this:

- *L187: "Our model conceptualisation also assumes that the catchment baseflow contribution is the only controlling variable of the spatial variation of C-Q slopes, enabling us to understand how well the C-Q slopes can be explained solely by differences in baseflow contributions across catchments."*

19. Fig2: I suggest to add axis labels and ticks for panel a and b

We revised Figure 2 as suggested.

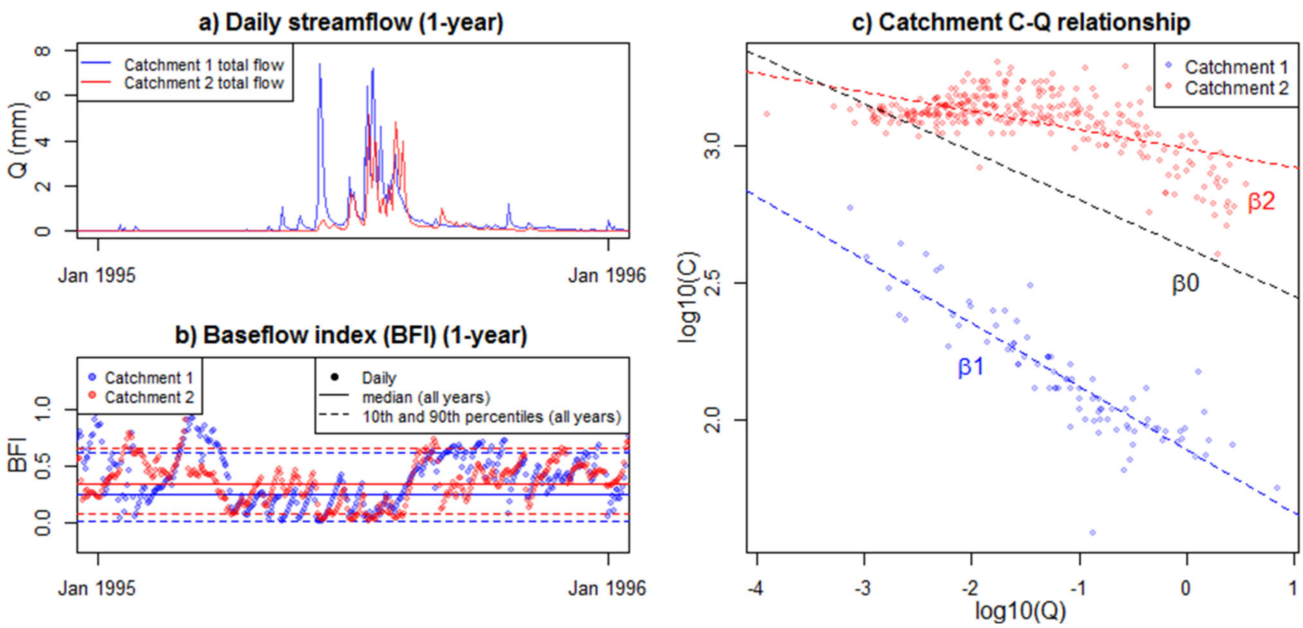


Figure 2. Illustration of conceptualisation of the BFI-based C-Q models (Eqn. 2) with the flow and EC data from two catchments. The catchment median BFI (*BFI_m*) is used as the main predictor of C-Q slope. a) daily flow time-series; b) daily BFI time-series and the corresponding median (*BFI_m*) and the 10th and 90th percentiles. c) C-Q relationships for the two catchments, where the shift in C-Q slope (β_1, β_2) away from the grand mean β_0 is determined by *BFI_m*. Both time-series for the daily flow (a) and BFI (b) are only shown for one year for visualisation.

20. L185: “together with flow” I do not understand this. If c=f(Q) in the C-Q relationship this is already included.

We deleted this phrase as part of the major change in Section 3.3.1 (now as part of Section 3.3 – as detailed in our response to your Comment #4b).

21. L214 “surface flow” is imprecise. There is not just baseflow and surface flow

We replaced this phrase with ‘quickflow’ (now L260).

22. L217: “In contrast, a catchment with a high BFI_m generally has a large range in instantaneous BFIs” I see several high BFI_m with not very high ranges in BFI. Fig3b rather looks like a bell shape with highest ranges for medium values, not like a linear relationship.

We agree with your observation. This nonlinearity highlights the need to explicitly consider the impact of the range of BFI. This is further justified with our additional analyses of the effects of *BFI_range* on C-Q slopes (see out detailed responses to your Comment #5). To highlight this non-linear relationship between *BFI_m* and *BFI_range*, we have updated Figure 3b) as below, and added corresponding interpretation in text:

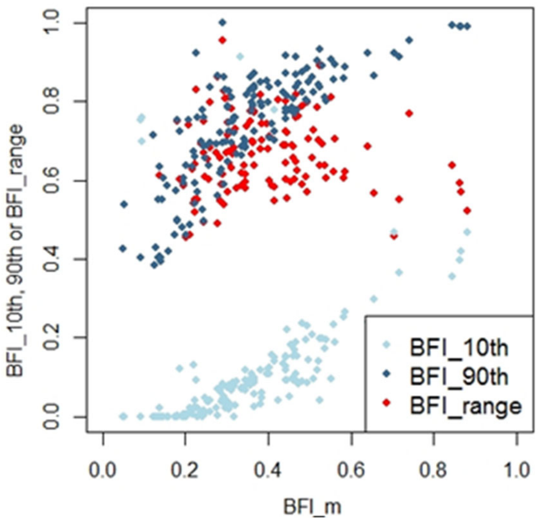


Figure 3 b). the 10th and 90th percentiles of daily BFI (BFI_10th and BFI_90th), and *BFI_range* (BFI_90th – BFI_10th) versus *BFI_m*.

- *L245: “Generally, temperate catchments have the highest BFI_m, while similar BFI_m values are seen across the other four climate zones. BFI_10th and BFI_90th have distributions consistent with BFI_m in all climate zones. As different catchments were analysed for each water quality variable, the same BFI metrics were also generated for each water quality variable, and their distributions are generally consistent across different variables (Figure S4, Supplementary Materials).”*

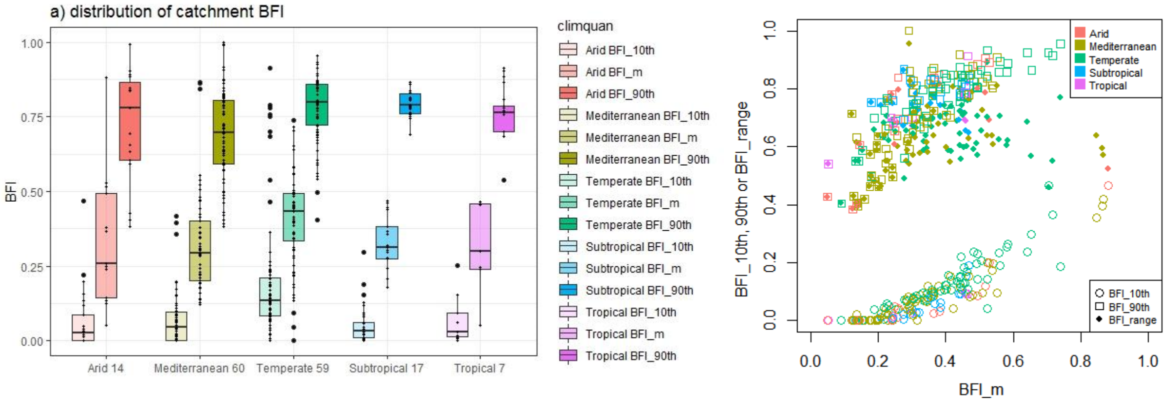
- *L256: “In general, catchments with high median BFI are likely to have a greater range of variation of daily BFI, as highlighted by the generally increasing BFI_range with higher BFI_m (Figure 3b), Spearman’s ρ = 0.33). The link between BFI_m and BFI_range suggests that catchments with higher BFI_m values are more likely driven by highly variable flow pathways. Specifically, a catchment with a low BFI_m tends to be associated with a small range of daily BFI (low BFI_range); thus, the catchment is likely to always have constantly low contributions of baseflow and higher contributions of quickflow, during both dry and wet conditions. In contrast, a catchment with a high BFI_m generally has a large range of daily BFIs (high BFI_range). This means that the catchment is* *more likely to switch between groundwater contributions in dry conditions (high daily BFI) and surface water contributions during wet conditions (low daily BFI). However, we also note that a small proportion of catchments (9 catchments) with the highest BFI_m (>0.6) actually have smaller BFI_range compared to other catchments with mid-range BFI values (0.4-0.6). This is a result of BFI_10th and BFI_90th both increasing with BFI_m, while the increase in BFI_90th plateaus at high BFI_m. This nonlinearity suggests that the full distribution of catchment baseflow contributions might not be sufficiently represented by either the BFI_m or BFI_range alone, providing further justification for the need to explicitly consider both the overall condition and the variation in catchment baseflow contributions when studying their effects on C-Q relationships."*

23. Fig 3: Boxplots can create confounding impressions if the sample size is very different, which I see from Figure 1. I would suggest adapting the boxplot widths according to the sample size and/or writing the sample size number to the plot for each climate zone (probably the numbers in the x axis labels?). For the right panel, I would suggest to colour the dots according to their climate zone as in Figure 1, possibly also for the left panel with different hue or saturation values to distinguish the different BFI quantiles.

Thank you for the suggestions. We already have the sample size for each climate zone labelled on the x-axis, we have revised the figure title to clarify this:

- ***Figure 3. a) Distribution of catchment median, and the 10th and 90th percentiles of daily BFI (BFI_m, BFI_10th, BFI_90th) for each climate zone along with the number of catchments analysed (x-axis)***

We attempted using different colours for individual climate zones in both the left and right subplots as suggested, but found them more confusing (see below) than the alternative versions nor coloured by climate zones. Therefore, we decided to not to colour Figure 3 by climate zones in our revised manuscript.



24. L255: "BFI-based model has only marginally lower performance … BFI-based model, while having the capacity to predict C-Q slope across space, can predict water quality almost as well as using the observed C-Q slope." I do not understand this comparison of individual C-Q slope with a model explaining the variability in C-Q slopes. It sounds like you were expecting worse performance, while actually a more complex model should improve performance to be a valid approach.

As detailed in our response to your Comment #4b), we replaced the previous results in Section 3.3.1 with new results that compare the performance of models with i) climate-specific impact of BFI; and ii) a single impact of BFI across all climate zones. We believe this revision can help clarify our approach and avoid any confusion on our communication of model performance.

25. L269: "confidence intervals"? I do not know credible intervals

"Credible interval" is the term used to describe uncertainty in the posterior samples (including simulations and parameter values) from Bayesian modelling (Gelman et al., 2013). We added a sentence here to clarify this as:

- *L328: "The 95% credible interval is the range between the 2.5th and 97.5th percentiles of the posterior parameter values, which was derived from the Bayesian posterior estimates of $\delta_{BFI\_climate}$ (Eqn. 3) to quantify the uncertainty in the modelled effects (Gelman et al., 2013).*

  Added Reference:

  - Gelman, A., Carlin, J. B., Stern, H. S., Dunson, D. B., Vehtari, A., & Rubin, D. B. (2013). Bayesian Data Analysis, Third Edition: Taylor & Francis.

26. Table 1: Why do you think the NSE of your BFI-base model are lower than the baseline model? Does it actually make sense to fit a more complex model in this case?

As detailed in our response to your Comment #4b), we replaced the current results in Section 3.3.1 with new results that compare the performance of models with i) climate-specific impact of BFI; and ii) a single impact of BFI across all climate zones. We believe this update will clarify our approach and avoid any confusion on our communication of model performance. This comment is not relevant to the revised content anymore.

27. Section 3.2 I do not like this whole section, I think the statements derived from selected examples are not representative for the catchments distribution of TSS export patterns within the corresponding climate zones (Fig6)

We intend to present these cases only as examples to show the potential impacts of BFI. We expect that showing the actual data of C, Q and BFI can help the readers to understand the key model components and relationships we aim to represent. We added a concluding sentence to this section to highlight the limitation of the examples and need to explore a wider range of catchments to generalize the pattern:

- *L299: "Overall, this preliminary analysis on a small subset of catchments suggests that baseflow contribution may indeed drive differences in C-Q relationships between catchments, and that these effects may vary across climate zones. However, it is difficult to conclude on the individual impact of BFI_m and BFI_range on the C-Q slopes, from these individual examples. The separate impacts of the two metrics are evaluated over a wide range of catchment conditions across the Australian continent with the model outputs from our BHM (Section 3.3)."*

28. Figure 5: What is the modelled effect? Is it dBFI_climate from equation 3? What is the NSE above each subplot describing?

Yes, Figure 5 shows the modelled effect is $\delta BFI_{climate}$ in Eqn 3. To clarify this, we added:

- *L327: "Figure 5 presents the median and the 95% credible intervals of these modelled effects for each water quality variable. The 95% credible interval is the range between the 2.5th and 97.5th percentiles of the posterior parameter values, which was derived from the Bayesian posterior estimates of $\delta_{BFI\_climate}$ (Eqn. 3) to quantify the uncertainty in the modelled effects (Gelman et al., 2013).*

Note that the term $\delta BFI_{climate}$ used previously was revised as $\delta_{BFI\_climate}$ throughout the manuscript to avoid the confusion highlighted in Comment #10 of Reviewer 2.

The NSE above each panel summarizes the model performance to predict the concentration of individual water quality variables. We removed these NSE values in the revised Figure 5 for consistency with our new model performance results (Table 1), as detailed in our response to your Comment #4b).

29. L278-285: I do not agree with this approach and subsequent observation. Fitting linear relationships is not appropriate for the given observations and "consistent diverging" behaviour goes beyond what can be interpreted here. Especially for the last two sentences: When looking at the overall point clouds, there is not clearly increasing variance (diverging behaviour) with higher BFI_m. In my opinion, weak relationships are overinterpreted here. These are also transferred to conclusions.

We'd like to clarify that L278-285 (L344-354 in the revised manuscript) do not intend to make inference on the BFI effects here; instead, we clarify that the diverging patterns are a result of the model structure. To clarify, we added a further sentence here:

- *L353: "Since these diverging patterns are a result of the model structure, we do not further interpret any physical representations on the impacts of BFI metrics on C-Q slopes."*

We have not included any summary of these results in the Conclusion section.

We understand the concern about the limitation of the linear model used, but we consider this model structure to be appropriate for our study purpose, which is to synthesize large-scale patterns of BFI impacts across different climate zones. The model structure is suitable for our purpose. To acknowledge this and recommend further improvements, we added some relevant discussions to the Conclusions, as:

- *L466: "Further, this study used a linear model structure to synthesise large-scale patterns of the impacts of baseflow contribution on the C-Q relationships across different climate zones. Although this model structure is limited and likely to be influenced by outliers, we believe it is suitable for the study purpose, as we are able to demonstrate the ability of the model to identify significant effects of catchment baseflow contribution on C-Q slopes, with statistically significant modelled effects for most climates and water quality parameters (Figures 5 and 6). Further studies can build on the learning from the current study to explore alternative model structures, to improve our ability to predict C-Q slopes within individual climate zones."*

30. L283: 'grand mean' I know you have used this term before, but I think it is not well chosen, as it does not tell that it is about the "solute-specific base C-Q slope". Consider changing the term

Thank you for the suggestion, we prefer to keep this term to maintain the link between our model structure and the results discussed. We added the following text to clarify the meaning of the term:

- *L347: "…all catchment C-Q slopes share a common 'grand mean' (Section 2.2) – representing a stable export pattern across Australian climate zones that is specific to the water quality variable (Lintern et al., 2021)."*

31. Figure 6: In my opinion, it is not justifiable to fit linear regressions to all combinations of solutes and climate zones, because several combinations have 1) too little sample sizes, 2) clearly non-linear relationships, and 3) in some cases plus influenced by outliers, e.g. the Tropical NOx fit. The legend titles should be improved.

Firstly, small sample size for some climate zones is not a real limitation of the Bayesian Hierarchical Model due to its ability to use information across data groups, as explained with more details in our response to your Comment #4a). To address this, we added the following text to Method Section 2.2 to discuss the key advantages of BHM in effectively handling data-limited situations and spatio-temporal data with uneven coverage, and justify the choice of BHM as a suitable model for this study, as:

- *L168: "The key reason for choosing this model is the high heterogeneity in the national C-Q dataset in both the record period and the representation of individual climate zones, as illustrated in Section 2.1.1. BHM is effective in handling data-limited situations via its 'information sharing' or 'borrowing power' across space (Gelman et al., 2013; Webb & King, 2009), which has been shown to be highly effective in explaining variability in spatial-temporal data under data-limited situations. This has been highlighted in several recent studies in modelling water quality over large regions in Australia (Guo et al., 2019, 2020; Liu et al., 2021). Another advantage of BHM is the ability to account for uncertainty, which is especially important for analysing water quality data, as these data are often associated with high uncertainty due to sparse sampling of the natural variability of chemical species in river flow (Guo et al., 2020; Liu et al., 2021)."*

Secondly, we consider a linear model suitable for the purpose of this study, which was to synthesize large scale patterns with a simple model structure applied to the whole country. Thus we considered the model structure suitable for our purpose. We added the following discussions to the Conclusion to acknowledge this model limitation and recommend further improvements:

- *L466: "Further, this study used a linear model structure to synthesise large-scale patterns of the impacts of baseflow contribution on the C-Q relationships across different climate zones. Although this model structure is limited and likely to be influenced by outliers, we believe it is suitable for the study purpose, as we are able to demonstrate the ability of the model to identify significant effects of catchment baseflow contribution on C-Q slopes, with statistically significant modelled effects for most climates and water quality parameters (Figures 5 and 6). Further studies can build on the learning from the current study to explore alternative model structures, to improve our ability to predict C-Q slopes within individual climate zones."*

We have clarified the legend titles in the revised Figure 6.

32. How does the modelled effect from Figure 5 relate to the slope of the linear regressions in Figure 6? This seems somewhat redundant to me.

The modelled effects in Figure 5 are the same as the slopes of the linear regression in Figure 6. Although the two figures overlap on the impacts of baseflow contribution on C-Q slopes, they focus on different aspects. Figure 5 focuses on the direction and significance of the baseflow effects themselves, while Figure 6 focuses on illustrating these effects with actual values of C-Q slopes, *BFI_m* and *BFI_range*. Figure 6 is a more intuitive way to show the impacts of baseflow contribution. Therefore, we see it necessary to keep both figures.

33. Figure 7: Firstly: The generalisation shown is questionable (see main and other comments). E.g. Figure 3b shows that highest BFI ranges are for medium BFI_m, suggesting that high BFI might also have more stable flow conditions with generally higher groundwater contributions. Secondly: This Figure takes a lot of time to understand and could benefit from some reworking including/according to other adaptations. The Figure text is unclear without reading the main text, as well as the meaning of a1, a2, b1, b2 only from the second reading. I suggest selecting other identifiers. The spatial organisation, e.g. link between the upper and the bottom panel and left, middle and right column, is not visually clear.

Figure 7 has been heavily revised (see below) to incorporate our new results with the model that focuses on the impact of *BFI_range* on catchment C-Q slopes (as detailed in our response to your Comment #5). We have also revised the organization of the figure to further improve the clarity and simplicity.

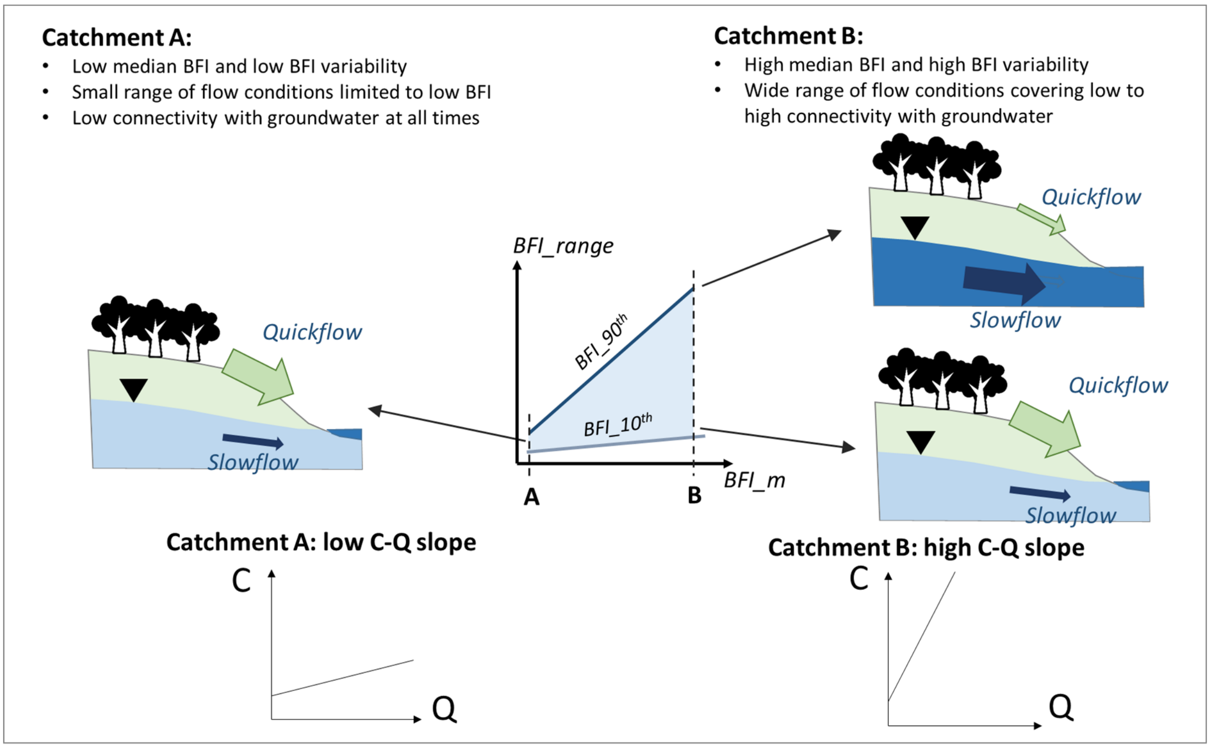


Figure 7. Conceptual diagram of the expected hydrological conditions in catchments with low and high median and variability in baseflow contribution (*BFI_m* and *BFI_range*), as Catchments A and B, respectively. The contrasting hydrological conditions can help explain our modelled results of the impacts of baseflow contributions on C-Q slopes. Note that the C-Q intercepts in the plots are not indicative since we do not investigate the variation in C-Q intercepts in this study.

34. L332-334: This statement could benefit from checking also mean concentrations: are activated sources low or high in more quickflow dominated catchments?

We have checked this and included the results in Figure S11 (now Figure S6 in the Supplementary in the revised manuscript). In general, we see high median concentration at the more quickflow dominated catchments (note: since we work in the log-10 space for both concentration and flow in deriving the C-Q slopes, the median concentration is a more useful indicator than the mean). To address this comment, we added this observation and the corresponding interpretation as below:

- *L375: "Therefore, the transport of TP is likely also enhanced by variations in baseflow conditions in the same manner as for TSS. Note that the overall positive effect of BFI_m in enhancing mobilisation is generally significant despite that catchments with higher BFI_m generally have lower median concentrations of TSS and TP (Figure S6), suggesting relatively limited sources in catchments with high BFI_m."*

35. L398-391: I cannot follow this point unfortunately due to missing information. Moreover, the characteristics land use, geology and climate are all integrated in the base flow index, which is a resulting hydrologic characteristic. This point definitely also needs a discussion part, including further literature and potential controls.

We deleted this sentence in revision as the referenced study (Liu et al., in preparation) is still ongoing. Instead, we added some discussion to highlight the need of further investigating other potential drivers for spatial variation of the C-Q relationships, such as land use, land cover, geology and climate:

- *L463: "This study used catchment-level metrics of baseflow contribution as the only predictor of C-Q slopes. The baseflow contribution alone can explain up to 22% variance in the C-Q slopes across the Australian continent. This highlights a substantial role in baseflow contribution in shaping the C-Q relationships, while also suggesting the need of further work to synthesise the impacts of baseflow contribution together with other spatial drivers (e.g., climate, land use, land cover and geology) to include their interactions and establishing their relative importance on influencing C-Q relationships."*

36. L392: Why is the Bayesian hierarchical model more effective than multiple linear regressions with BFI or other multivariate models? For me, Figure 5 (outcome of the Bayesian model) and Figure 6 (not a direct model output) were somewhat redundant. This is not covered in enough in the discussion section.

Bayesian Hierarchical Model (BHM) and multivariate regressions are not exclusive to each other i.e. BHM is also capable of including multiple predictors. We are not intending to compare BHM with any other model with this statement, instead we highlight the efficiency of BHM in making inferences and synthesizing large-scale patterns with limited and heterogeneous data availability across space. This has been already highlighted in the last paragraph of the Conclusion, and we concluded these advantages of BHM as:

- *L474: "This study also highlights the effectiveness of Bayesian hierarchical models in interpreting water quality data across large spatial scales. Such a model is ideal to analyse water quality data over a large number of catchments, with high heterogeneity in temporal coverage and sampling frequency. This is particularly relevant for Australia, as water quality monitoring is often undertaken under different local/regional programs, and thus limited to certain timeframes and focusing on specific management interests."*

We have further strengthened the justification of applying BHM in the revised Section 2.2 (as detailed in our responses to your Comment #4a) as below. We believe that this revision can also help highlight the effectiveness of BHM.

- *L168: "The key reason for choosing this model is the high heterogeneity in the national C-Q dataset in both the record period and the representation of individual climate zones, as illustrated in Section 2.1.1. BHM is effective in handling data-limited situations via its 'information sharing' or 'borrowing power' across space (Gelman et al., 2013; Webb & King, 2009), which has been shown to be highly effective in explaining variability in spatial-temporal data under data-limited situations. This has been highlighted in several recent studies in modelling water quality over large regions in Australia (Guo et al., 2019, 2020; Liu et al., 2021). Another advantage of BHM is the ability to account for uncertainty, which is especially important for analysing water quality data, as these data are often associated with high uncertainty due to sparse sampling of the natural variability of chemical species in river flow (Guo et al., 2020; Liu et al., 2021)."*

Figures 5 and 6 are intended to show different aspects of the results although they have overlapped contents, we explained this in detail in our response to your Comment #32.

37. SUPPLEMENTS: Figure S3: I do not understand why the BFI_m should be shown per solute, if the BFI depends on discharge and not on concentration.

BFI depends on discharge only, but the catchments analysed for individual water quality variables differ because the catchment selection criteria (Section 2.1.1) considered water quality data availability. This figure shows each water quality variable in as a subplot to incorporate the difference in catchments analysed.

38. REFERENCES: Ehrhardt et al. is already finally published, please change reference to: Ehrhardt, S., Kumar, R., Fleckenstein, J. H., Attinger, S., & Musolff, A. (2019). Trajectories of nitrate input and output in three nested catchments along a land use gradient. Earth Syst. Sci., 23(9), 3503-3524. https://www.hydrol-earth-syst-sci.net/23/3503/2019/

Thank you, we have updated this reference in the revised manuscript.

Responses to Reviewer #2

Our responses are in blue and revisions are underlined.

This manuscript presents a synthesis of baseflow effects on C-Q relationships in watersheds across Australia. The authors have leveraged a Bayesian Hierarchical Model in this research. Overall, I think the research is solid, the manuscript is well written, and it can become an important contribution to the literature on riverine C-Q relationships. I provide below some comments to the author, which I hope can help improve the manuscript.

We thank you for providing valuable feedback on the study for further improvement. We provide below a point-to-point response to your comments, along with our corresponding revisions.

1. The use of Bayesian Hierarchical Model should be more fully justified. I am aware of the research the team has done in the past few years involving Bayesian approaches, but why is it used in this work on C-Q relationships. Please provide your reasoning in the Introduction, probably the last paragraph.

To address this comment, we added further discussion to the end of Introduction and Method Section 2.2 (which introduces the modelling approach) on the key advantages of BHM in effectively handling data-limited situations and spatio-temporal data with uneven coverage, and justifications for why BHM is suited to the dataset analysed in this study, as:

- *L96: "We answer our research questions and test our hypotheses with a Bayesian hierarchical model (BHM) (Gelman et al., 2013), which is an integrated framework that enables sharing information across catchments to strengthen the statistical power of explaining variation in individual catchments. The model is a powerful approach to capture water quality variability across catchments of varying conditions and record lengths, which is the case for Australian water quality data (Guo et al., 2019, 2020; Liu et al., 2021)."*
- *L168: "The key reason for choosing this model is the high heterogeneity in the national C-Q dataset in both the record period and the representation of individual climate zones, as illustrated in Section 2.1.1. BHM is effective in handling data-limited situations via its 'information sharing' or 'borrowing power' across space (Gelman et al., 2013; Webb & King, 2009), which has been shown to be highly effective in explaining variability in spatial-temporal data under data-limited situations. This has been highlighted in several recent studies in modelling water quality over large regions in Australia (Guo et al., 2019, 2020; Liu et al., 2021). Another advantage of BHM is the ability to account for uncertainty, which is especially important for analysing water quality data, as these data are often associated with high uncertainty due to sparse sampling of the natural variability of chemical species in river flow (Guo et al., 2020; Liu et al., 2021)."*

The following references were added to the manuscript support the above arguments:

- Guo, D., Lintern, A., Webb, J. A., Ryu, D., Bende-Michl, U., Liu, S. & Western, A. W. (2020). A data-based predictive model for spatiotemporal variability in stream water quality. Hydrology and Earth System Sciences, 24(2), pp. 827-847.doi:10.5194/hess-24-827-2020
- Guo, D., Lintern, A., Webb, J. A., Ryu, D., Liu, S., Bende-Michl, U., . . . Western, A. W. (2019). Key Factors Affecting Temporal Variability in Stream Water Quality. Water Resources Research, 55(1), 112-129. doi:10.1029/2018wr023370
- Liu, S., Ryu, D., Webb, J. A., Lintern, A., Guo, D., Waters, D., & Western, A. W. (2021). A Bayesian approach to understanding the key factors influencing temporal variability in stream water quality

– a case study in the Great Barrier Reef catchments. Hydrol. Earth Syst. Sci., 25(5), 2663-2683. doi:10.5194/hess-25-2663-2021

2. The authors reported that the Bayesian Hierarchical Model can explain over half of the observed variability in concentration of TSS, EC and P species across all catchments (93% for EC, 63% for TP, 63% for SRP, and 60% for TSS). I feel the intention has switched here from understanding C-Q relationship to predicting water-quality concentrations, which seems to be a distraction to me. Moreover, what's the benefit of adopting the Bayesian Hierarchical Model, given that many statistical models (e.g., WRTDS) have been developed and can probably provide more accurate estimates?

Thank you. In the revised manuscript, we have replaced the current results in Section 3.3.1 with new results to compare the performance of models with i) climate-specific impact of BFI; and ii) a single impact of BFI across all climate zones. We believe this update will help remove the previous confusion on our communication of the model performance, while also illustrating the value of considering climate-specific impacts of baseflow contribution in our study.

The key results added are included in the revised Table 1:

Table 1. Performance of the BFI-based C-Q models – the columns show four alternative model structures with *BFI_m* or *BFI_range* as the key predictor, and with the impacts of baseflow contribution considered as lumped or specific to individual climate zones. The rows show results for individual water quality parameters. All model performances are summarised by R^2, which quantifies the percentage of variance in C-Q slopes explained by the BFI-based models.

WQ parameter	Median C-Q slope	Current (climate-specific impacts)		Baseline (lumped impact across climate zones)	
		BFI_m	*BFI_range*	*BFI_m*	*BFI_range*
TSS	0.15	0.16	0.11	0	0.04
TP	0.09	0.14	0.17	0	0.08
SRP	0.06	0.02	0	0.03	0.05
TN	0.09	0.18	0.12	0.02	0.03
NOx	0.36	0.22	0.18	0.03	0
EC	-0.07	0	0.01	0	0.01

We added new interpretation of these new results as:

- *L305: "Using catchment-level metrics of baseflow contribution alone (either BFI_m or BFI_range) can explain up to 22% of the variation in catchment C-Q slopes. Although these results represent limited model predictive capacity, the model does cover a large range of catchment conditions such as contrasting land uses and hydro-climate conditions. Therefore, the amount of variation that can be explained by a single BFI metric highlights baseflow contribution as an important factor that influences catchment C-Q relationships. Further, it is also worth highlighting that incorporating climate-specific impacts of baseflow contribution is highly beneficial in explaining these variations. For all six water quality parameters, the baseline model – which uses a lumped effect of catchment baseflow contribution across different climate zones – can barely explain any variation in the C-Q slopes (with all $R^2 < 0.08$, i.e., <8% of the variation explained). In contrast, the climate-specific models generally offer up to 20% increase in the variance explained for C-Q slopes, except for EC and SRP, for which performance is equally low regardless of whether the effects of baseflow contribution are separated for individual climates. The low performances for EC and SRP are likely attributed to the smaller magnitudes of C-Q slopes as highlighted in the lower median C-Q slope in Table 1, making it statistically more difficult to explain variations across catchments for these two water quality variables. These results further emphasise that in general, the impacts* *of catchment baseflow contribution on C-Q slopes are better defined within individual climate zones, which confirms the validity of our BFI-based C-Q models (Eqn. 3).”*

In addition, we acknowledge that our Bayesian Hierarchical Model (BHM) predicts across different catchments and is thus different to the catchment-specific WRTDS model. BHM is fitted to all catchments simultaneously and thus has a huge advantage of ‘borrowing power’ – to inform parameter estimation for one group of data by information from other groups (with ‘group’ being climate zone in our model). The hierarchical model architecture is ideally suited to grouped datasets, which enables data within the same group to share common features. The authors’ previous studies illustrated the effectiveness of this model for simulating water quality temporal variability across multiple catchments (Liu et al., 2021; Guo et al., 2020; Guo et al., 2019). Therefore, BHM is useful to conceptualize the nation-wide C-Q dataset across multiple catchments and climate zones. As detailed in our response to your Comment #1, we have added the sections below to help justify our choice of BHM:

- *L96: “We answer our research questions and test our hypotheses with a Bayesian hierarchical model (BHM) (Gelman et al., 2013), which is an integrated framework that enables sharing information across catchments to strengthen the statistical power of explaining variation in individual catchments. The model is a powerful approach to capture water quality variability across catchments of varying conditions and record lengths, which is the case for Australian water quality data (Guo et al., 2019, 2020; Liu et al., 2021).”*
- *L168: “The key reason for choosing this model is the high heterogeneity in the national C-Q dataset in both the record period and the representation of individual climate zones, as illustrated in Section 2.1.1. BHM is effective in handling data-limited situations via its ‘information sharing’ or ‘borrowing power’ across space (Gelman et al., 2013; Webb & King, 2009), which has been shown to be highly effective in explaining variability in spatial-temporal data under data-limited situations. This has been highlighted in several recent studies in modelling water quality over large regions in Australia (Guo et al., 2019, 2020; Liu et al., 2021). Another advantage of BHM is the ability to account for uncertainty, which is especially important for analysing water quality data, as these data are often associated with high uncertainty due to sparse sampling of the natural variability of chemical species in river flow (Guo et al., 2020; Liu et al., 2021).”*

3. Figure 3b: It is not a strictly positive relationship for the entire range of BFI_m. The variability continues to increase with BFI_m up to ~ 0.5 and then starts to decrease with BFI_m. The latter part of the curve seems largely ignored in the manuscript, including Discussion. The same observation holds true for the individual constituents (Figure S5).

We agree with your observation. We have further investigated this and updated Figure 3b as below, along with the interpretation in text:

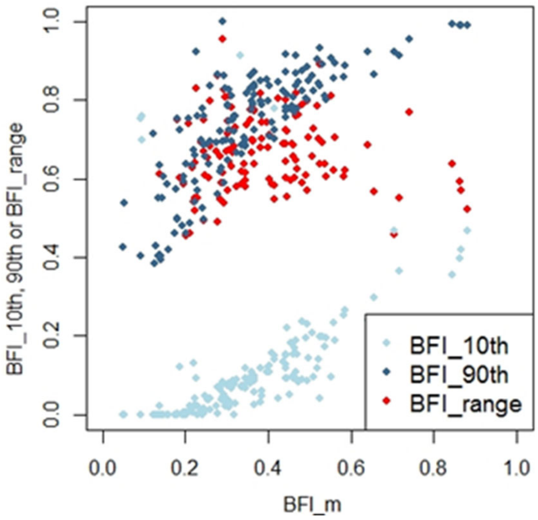


Figure 3 b). the 10th and 90th percentiles of daily BFI (BFI_10th and BFI_90th), and *BFI_range* (BFI_90th – BFI_10th) versus *BFI_m*.

- *L245: "Generally, temperate catchments have the highest BFI_m, while similar BFI_m values are seen across the other four climate zones. BFI_10th and BFI_90th have distributions consistent with BFI_m in all climate zones. As different catchments were analysed for each water quality variable, the same BFI metrics were also generated for each water quality variable, and their distributions are generally consistent across different variables (Figure S4, Supplementary Materials)."*
- *L256: "In general, catchments with high median BFI are likely to have a greater range of variation of daily BFI, as highlighted by the generally increasing BFI_range with higher BFI_m (Figure 3b), Spearman's ρ = 0.33). The link between BFI_m and BFI_range suggests that catchments with higher BFI_m values are more likely driven by highly variable flow pathways. Specifically, a catchment with a low BFI_m tends to be associated with a small range of daily BFI (low BFI_range); thus, the catchment is likely to always have constantly low contributions of baseflow and higher contributions of quickflow, during both dry and wet conditions. In contrast, a catchment with a high BFI_m generally has a large range of daily BFIs (high BFI_range). This means that the catchment is more likely to switch between groundwater contributions in dry conditions (high daily BFI) and surface water contributions during wet conditions (low daily BFI). However, we also note that a small proportion of catchments (9 catchments) with the highest BFI_m (>0.6) actually have smaller BFI_range compared to other catchments with mid-range BFI values (0.4-0.6). This is a result of BFI_10th and BFI_90th both increasing with BFI_m, while the increase in BFI_90th plateaus at high BFI_m. This nonlinearity suggests that the full distribution of catchment baseflow contributions might not be sufficiently represented by either the BFI_m or BFI_range alone, providing further justification for the need to explicitly consider both the overall condition and the variation in catchment baseflow contributions when studying their effects on C-Q relationships."*

4. It would be interesting to investigate the effects of seasons and antecedent discharge conditions (wet vs. dry), both of which may change the response of C-Q slope to the BFI_m metric. There may be strong contrast among, for example, growing vs. non-growing seasons. Toward the end of manuscript, the authors have briefly pointed out the possibility of season effects. I think it is probably beyond your scope to look into these effects in this paper, but I encourage the authors to provide a brief discussion to point out that the response of C-Q slope to the BFI_m metric can vary among different seasons, among different antecedent discharge conditions, and even among different periods. In the latter regard, it is reported that anthropogenic disturbances

and/or management actions occurred in the catchment can cause the C-Q relationship to change. For example, Zhang (2018) provides an investigation of C-Q relationship for different river flows and years: https://doi.org/10.1016/j.scitotenv.2017.09.221.

Thank you for the excellent suggestion. We have added the following discussions on the existing literature on the temporal changes of BFI with respect to existing studies, from which future directions of research is highlighted.

- *L435: "…future studies should also consider more broadly the temporal variations in flow regime and baseflow condition and their influences on C-Q relationships. For example, seasonality can play a big role in shaping the C-Q relationships for nutrients, as these relationships over time during the build-up of pollutant sources, and during the flushing of readily available sources at the onset of high flow periods (Bende-Michl et al., 2013). Besides, anthropogenic disturbances and/or management actions in the catchment can cause changes in C-Q relationships over time (Zhang, 2018). Flow flashiness is also shown to influence the C-Q relationships, which differ across particulates and solutes, and across natural and highly regulated catchments (Moatar et al., 2020)."*

5. The term BFI_m (median BFI) is not self-evident in the Abstract. Given the importance of this metric, I encourage the authors to define it more clearly in the Abstract.

We added the following clarification to the Abstract:

- *L19: "The study aims to assess how baseflow contributions, as defined by the median and the range of daily baseflow indices within individual catchments (BFI_m and BFI_range, respectively), influence C-Q slopes across 157 catchments in Australia spanning five climate zones."*

6.
   a) The authors have used BFI_l and BFI_h to represent the variability of BFI, which makes sense to me. I may have used 2.5% and 97.5% instead but 10% and 90% are fine.
   b) By the way, have you considered using standard deviation to capture the spread, which may help shorten the manuscript in terms of text and figures presented? I think an argument can be added to the end of Section 2.1, which favors the use of BFI_l and BFI_h, that these are percentile based and hence are more robust to outliers.

a) Using 2.5% and 97.5% quantiles have potential risk to capture some outliers considering our catchment selection criteria (Section 2.1). Specifically, catchments with a minimum of 300 C-Q-pairs can be included in our analysis, which means that if 2.5% and 97.5% quantiles are used to calculated *BFI_l* and *BFI_h*, each index can be calculated with <10 data points at the most data-scarce catchments. Therefore, we intend to keep the original decision of having *BFI_l* and *BFI_h* as the 10% and 90% quantiles of BFI for each catchment. Note that in the revised manuscript, the terms *BFI_l* and *BFI_h* were revised as *BFI_10th* and *BFI_90th* to be more intuitive.

b) We thank you for suggesting an additional justification for the use of *BFI_l* and *BFI_h* and we have added this to the revised manuscript as:

- *L162: "BFI_m takes the median of all daily BFIs, which represents the overall baseflow contribution of the catchment. BFI_range is the difference between the 10th and 90th percentiles of daily BFIs (BFI_10th and BFI_90th). These quantile-based metrics were preferred over the mean and standard deviation as they are more robust against outliers."*

A relevant change is that we added a new model with the same structure as our original model based on *BFI_m*, which is based on *BFI_range* (the difference between the 10th and 90th percentiles of daily BFI). We revised Figures 5 and 6 (see below) to show results from both models, which illustrated that the effects of *BFI_range* on C-Q slopes are highly consistent with the modelled effects of *BFI_m* on C-Q slopes. These new results provide more concrete evidence on our previous 'speculation' on how the BFI range in a catchment can impact its C-Q slope. According to these new results, we have substantially revised the discussions after Figures 6 and 7 and its associated discussions in Section 3.3.

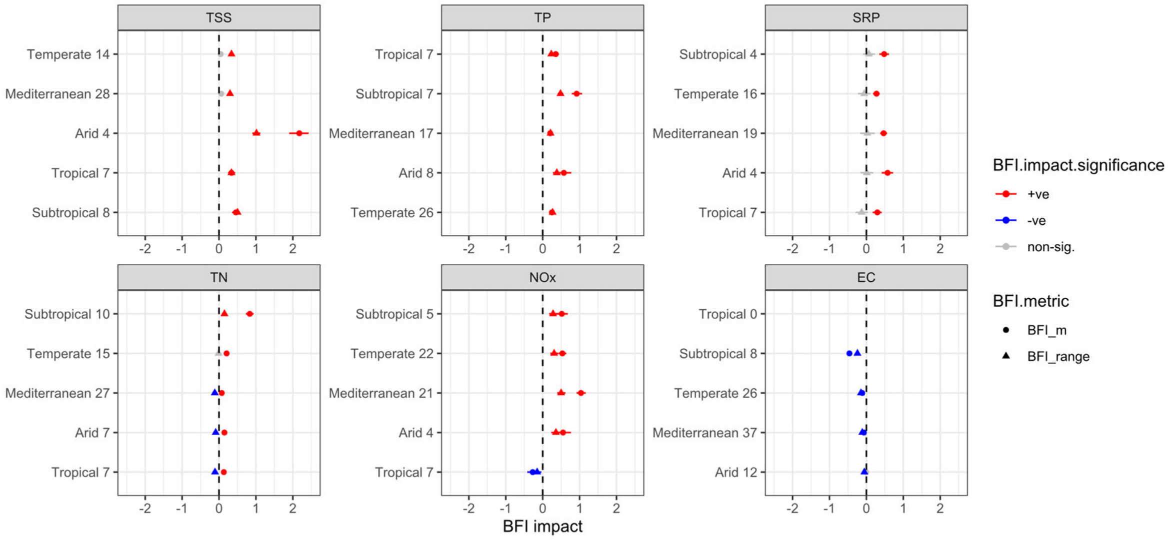


Figure 5. Modelled effects of *BFI_m* and *BFI_range* on catchment C-Q slopes for each climate zone ($\delta_{BFI\_climate}$) for each water quality parameter. The bars show the 95% credible intervals (the range between 2.5th to 97.5th percentiles of Bayesian posterior distribution) of the modelled effects, and the dots indicate the corresponding median levels. The colours indicate whether an effect is significantly positive (red), significantly negative (blue), or non-significant (grey); a positive effect means that the C-Q slope increases with a higher catchment *BFI_m* or *BFI_range*, and vice versa. Black dashed lines show the zero-effect i.e., no effect at all. The plot includes results from models with each of *BFI_m* and *BFI_range* as the key predictor, which are differentiated by marker shapes.

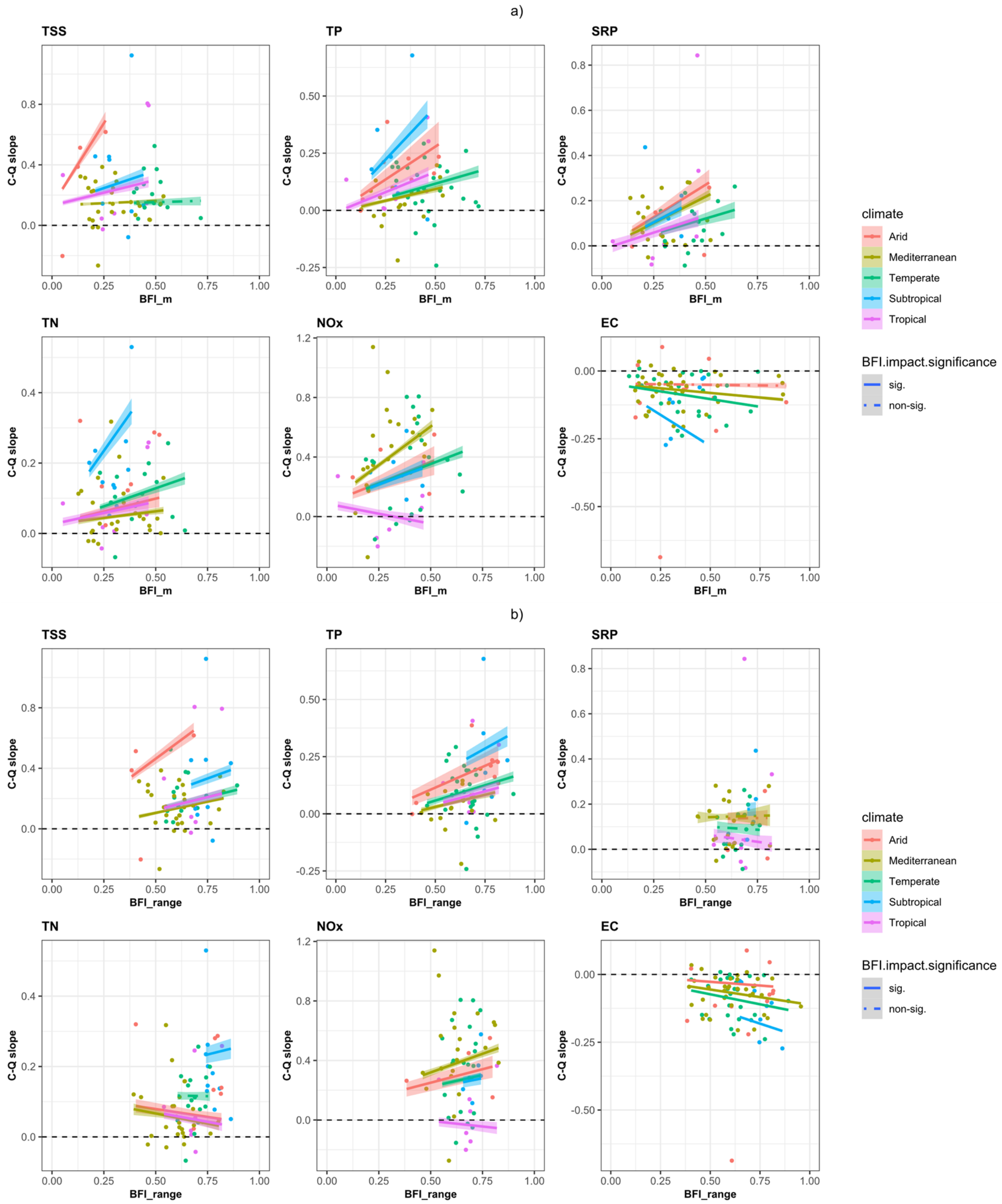


Figure 6. a) Catchment C-Q slope vs. *BFI_m* and b) Catchment C-Q slope vs. *BFI_range*, coloured by climate zones. The lines represent the modelled C-Q slope~*BFI_m* or C-Q slope~*BFI_range* regression lines for individual climate zones. The bands represent the 95% credible interval (the range between 2.5th to 97.5th percentiles of Bayesian posterior distribution) of the modelled C-Q slopes. The dots represent the 'true' C-Q slopes estimated with C-Q observations at individual catchments. The black dashed lines mark a zero C-Q slope which differentiate mobilisation (C-Q slope>0) from dilution (C-Q slope<0).

7. For days with multiple samples, is it necessary to pre-calculate the average concentration? Why not keeping all the samples in the analysis? In addition, it may be helpful to provide a table that quantifies the fraction of such days in the record.

Streamflow records are generally in daily timestep (instead of sub-daily) across the nation, which limits us to use all high-frequency water quality samples to perform C-Q analysis. To clarify this, we added a sentence to justify this averaging process in Section 2.1.1 (data) as:

- *L117: "A daily average is taken if more than one water quality sample was collected on any day at any site – see the percentage of records where more than one samples were taken in one day individual catchments in Table S2 (Supplementary Materials). This is because that streamflow in Australia is largely recorded at a daily timestep, which limits our ability to analyse all high-frequency water quality."*

We added Table S2 to the Supplementary Information to specify the fraction of days with multiple samples – this generally occurs rarely except for some EC sites where high-frequency samples were collected.

8. BFI calculation: I am curious about the use of 0.98 for alpha in the baseflow filter. Did Ladson et al. (2013) recommend this value? What is the rationale?

Yes, this was a recommendation in Ladson et al. (2013). The study stated: "recent comparisons of modelled and measured baseflow values in the Murray Darling Basin suggest a value of 0.98 produces more reasonable results.". Murray Darling Basin is a region in which a large proportion of our study catchments are located (Figure 1), and we already clarified this in L130 (now L152 in the revised manuscript):

- *L156: "The daily BFIs were estimated using a Lynne-Hollick baseflow filter with Alpha = 0.98 … as recommended for Murray-Darling Basin in the south-eastern Australia (Ladson et al., 2013), within which a large number of the study catchments are located."*

9. Figure 2: Please add numbers and units (even if hypothetical) on the y-axis for panels a and b.

We have revised Figure 2 as suggested.

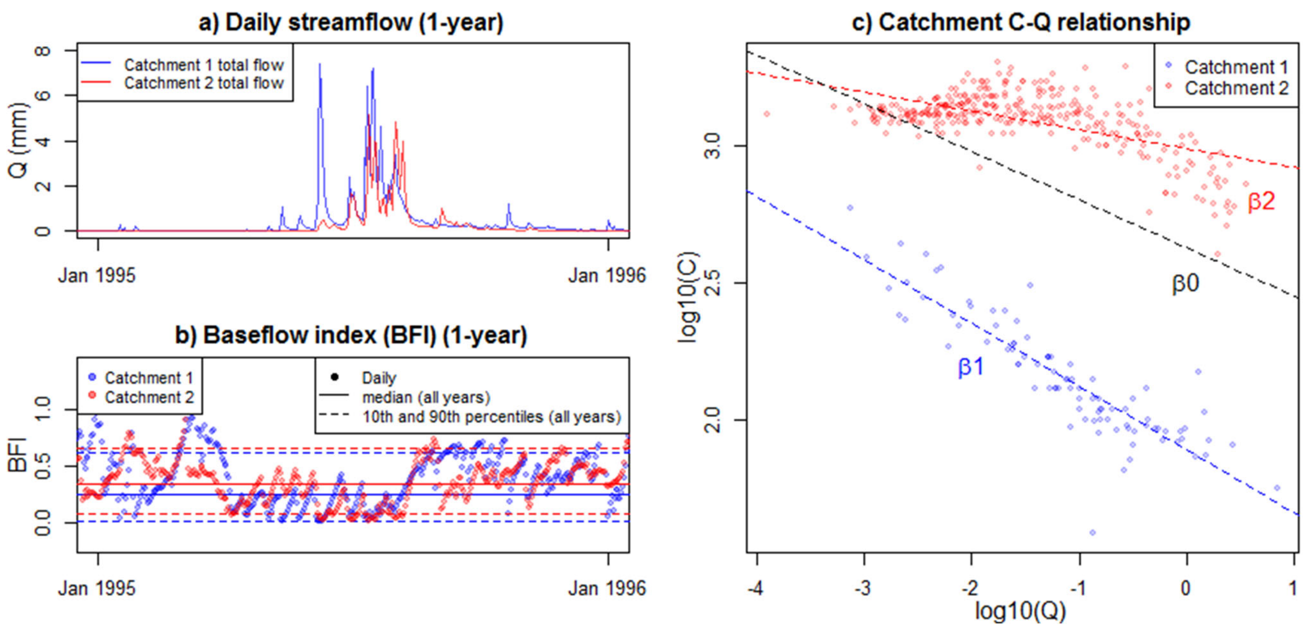


Figure 2. Illustration of conceptualisation of the BFI-based C-Q models (Eqn. 2) with the flow and EC data from two catchments. The catchment median BFI (*BFI_m*) is used as the main predictor of C-Q slope. a) daily flow time-series; b) daily BFI time-series and the corresponding median (*BFI_m*) and the 10th and 90th percentiles. c) C-Q relationships for the two catchments, where the shift in C-Q slope (β_1, β_2) away from the grand mean β_0 is determined by *BFI_m*. Both time-series for the daily flow (a) and BFI (b) are only shown for one year for visualisation.

10. Equations 2-3: Consider changing $\delta BFIclimate$ to δBFI_$climate$. (Move "BFI" to the subscript.) At first glance, I thought this is the product of two variables (δ and BFIclimate).

We will revise this term as suggested (to $\delta_{BFI\ climate}$) throughout the paper.

11. Section 3.3.1, including Table 1: I would like to refer back to my comment above. The NSE values do not seem to be comparable to more established approaches such as WRTDS. What is the value of showing these results? Should the baseline model or the BFI-based model be used for predicting concentrations? Why not those other established approaches?

As detailed in our response to your Comment #2, we have removed the current results in Section 3.3.1 (now as part of Section 3.1) and instead we have included new results to compare the performance of models with i) climate-specific impact of BFI; and ii) a single impact of BFI across all climate zones. We believe this update can help remove the previous confusion on our communication of the model performance.

Furthermore, since the model is fitted to all data across multiple catchments, this model performance essentially describes how well the model explained the total spatio-temporal variability in water quality within the national dataset. Predicting spatio-temporal variability is a much more difficult task than predicting the temporal variability alone – as achieved by WRTDS for individual catchments (Zhang et al., 2021; Sprague et al., 2019). Thus we believe the model performance of BHM is not comparable with that of WRTDS. However, we believe that any related confusion have been removed with the proposed revision of Section 3.3.1.

References:

Zhang, Q., Webber, J. S., Moyer, D. L., & Chanat, J. G. (2021). An approach for decomposing river water-quality trends into different flow classes. Science of The Total Environment, 755, 143562. doi:https://doi.org/10.1016/j.scitotenv.2020.143562

Sprague, L. A., Mitchell, R. M., Pollard, A. I., & Falcone, J. A. (2019). Assessing water-quality changes in US rivers at multiple geographic scales using results from probabilistic and targeted monitoring. Environmental Monitoring and Assessment, 191(6), 348. doi:10.1007/s10661-019-7481-5

12. Section 3.3.2: According to published literature on many catchments around the world, SRP is a minor component of TP, whereas NOx is a major component of TN. It is quite interesting that in these Australian catchments, NOx/TN is quite small. This presents a strong contrast to many regions and may be discussed with a couple of sentences.

Thank you for highlighting this point. We have added some discussion on the contrasting NOx:TN pattern in Australia compared with other regions.

- *L414: "A large proportion of TN in Australia is present in particulate forms (Figure S9) with most catchments having NOx:TN ratios lower than 0.25, which contrasts with many other catchments in the United States and Europe (Ator et al., 2011; Durand et al., 2011)."*