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Bayesian change point quantile regression approach to enhance the understanding of shifting phytoplankton-dimethyl sulfide relationships in aquatic ecosystems

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ABSTRACT

Dimethyl sulfide (DMS) serves as an anti-greenhouse gas, plays multiple roles in aquatic ecosystems, and contributes to the global sulfur cycle. The chlorophyll *a* (CHL, an indicator of phytoplankton biomass)-DMS relationship is critical for estimating DMS emissions from aquatic ecosystems. Importantly, recent research has identified that the CHL-DMS relationship has a breakpoint, where the relationship is positive below a CHL threshold and negative at higher CHL concentrations. Conventionally, mean regression methods are employed to characterize the CHL-DMS relationship. However, these approaches focus on the response of mean conditions and cannot illustrate responses of other parts of the DMS distribution, which could be important in order to obtain a complete view of the CHL-DMS relationship. In this study, for the first time, we proposed a novel Bayesian change point quantile regression (BCPQR) model that integrates and inherits advantages of Bayesian change point models and Bayesian quantile regression models. Our objective was to examine whether or not the BCPQR approach could enhance the understanding of shifting CHL-DMS relationships in aquatic ecosystems. We fitted BCPQR models at five regression quantiles for freshwater lakes and for seas. We found that BCPQR models could provide a relatively complete view on the CHL-DMS relationship. In particular, it quantified the upper boundary of the relationship, representing the limiting effect of CHL on DMS. Based on the results of paired parameter comparisons, we revealed the inequality of regression slopes in BCPQR models for seas, indicating that applying the mean regression method to develop the CHL-DMS relationship in seas might not be appropriate. We also confirmed relationship differences between lakes and seas at multiple regression quantiles. Further, by introducing the concept of DMS emission potential, we found that pH was not likely a key factor leading to the change of the CHL-DMS relationship in lakes. These findings cannot be revealed using piecewise linear regression. We thereby concluded that the BCPQR model does indeed enhance the understanding of shifting CHL-DMS relationships in aquatic ecosystems and is expected to benefit efforts aimed at estimating DMS emissions. Considering that shifting (threshold) relationships are not rare and that the BCPQR model can easily be adapted to different systems, the BCPQR approach is expected to have great potential for generalization in other environmental and ecological studies.

1. Introduction

Dimethyl sulfide (DMS) was recognized as an anti-greenhouse gas because its oxidized products acted as cloud condensation nuclei, which reflected solar irradiation and thereby contributed to the reduction of earth temperature (i.e., the CLAW hypothesis,

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Charlson et al., 1987). Despite recent debates or rejection of the CLAW hypothesis (Cropp et al., 2018; Quinn and Bates, 2011), the hypothesis is likely relevant in some regions like the Southern and Arctic Oceans (Krüger and Graßl, 2011; Levasseur, 2013). DMS also plays multiple essential roles in aquatic ecosystems, such as serving as an antioxidant for phytoplankton (Sunda et al., 2002) and facilitating a tritrophic mutualism between primary producers and top predators (Savoca and Nevitt, 2014). In addition, DMS is important to the global sulfur cycle (Eyice et al., 2015), accounting for about 80% of global biogenic sulfur emissions to the atmosphere (Kettle and Andreae, 2000). Phytoplankton, indicated by chlorophyll *a* (CHL) (Bates et al., 1994; Zhang et al., 2008), is the major producer of DMS (Charlson et al., 1987; Gondwe et al., 2003). Therefore, understanding the CHL-DMS relationship is critical for estimating regional or global DMS emissions from aquatic ecosystems (Anderson et al., 2001; Galí et al., 2015; Simó and Dachs, 2002).

Correlation analysis and ordinary linear regression have been the most widely used methods to explore the CHL-DMS relationship. Most studies revealed a positive effect of CHL on DMS (e.g., a significantly positive correlation coefficient or regression slope) (Gao et al., 2017; Iverson et al., 1989; Lana et al., 2011; Law et al., 2017; Lizotte et al., 2020; Tan et al., 2017; Tortell et al., 2011; Walker et al., 2000; Yang, 1999; 2000; Yang and Tsunogai, 2005; Yang et al., 2009; 2011; Zhang et al., 2014), while several studies reported a negative relationship (Froelichd et al., 1985) or no relationship at all (Nemcek et al., 2008; Watanabe et al., 1995). We note that maximum CHL concentrations in studies deducing positive CHL-DMS relationships were always much lower than those deducing negative or no relationships. For example, CHL concentrations in a series of studies on Chinese seas (Yang, 1999; 2000; Yang and Tsunogai, 2005; Yang et al., 2009; 2011) were all lower than 4 μg/L. In contrast, the CHL concentration can reach approximate 60 μg/L in Froelichd et al. (1985).

A recent study examining the CHL-DMS relationship across a broad range of CHL concentrations implemented a change point model to capture the ascending and descending limbs of this relationship (Deng et al., 2020). The change point model aims to determine one or more unknown change points at which the stressor-response relationship changes. In Deng et al. (2020), the authors used 246 paired observations of CHL and DMS from 100 Chinese lakes and collected 426 paired observations from global oceans. They applied a piecewise linear regression model (Muggeo, 2003) to detect thresholds of CHL concentration, at which CHL-DMS relationships significantly changed. Benefiting from the novel application of piecewise regression, the authors revealed hump-shaped CHL-DMS relationships in both lakes and seas, which were expected to increase the estimation accuracy of global DMS emissions from aquatic ecosystems (Deng et al., 2020). The hump-shaped relationship also seemed to resolve the contradiction of the sign of the CHL-DMS relationship in previous studies, whose deductions might have been constrained by a relatively smaller sample size, a narrow range of sampled CHL concentration, or an application of a overly simplified linear regression model.

Although many informative studies have investigated the CHL-DMS relationship, we note that those studies mainly used mean regression methods (e.g., the ordinary linear regression or piecewise regression), by which the relationship between CHL and the *mean* of DMS distribution was estimated. A practically important alternative to classical mean regression methods is quantile regression (QR) (Koenker and Bassett, 1978). To the best of our knowledge, QR has not been used to examine CHL-DMS relationships.

QR explores the effect of one or more predictors on any quantile of the response variable distribution (Das et al., 2019; Koenker and Bassett, 1978). Compared with mean regression methods, QR can provide a more complete view of possible causal relationships and can reveal useful predictive relationships at some parts of the response variable distribution, even when there is a weak or no predictive relationship between the predictor(s) and the mean of the response variable distribution (Cade and Noon, 2003). In addition, QR appears more robust to outliers of the response variable (Scharf et al., 1998) and is not constrained by the equal variance assumption (Cade and Noon, 2003; Das et al., 2019). QR has been successfully applied to environmental and ecological studies. QR has been used to 1) illustrate a relatively complete view of stressor-response relationships at multiple regression quantiles (Cade et al., 2008; Liang et al., 2021; Muller et al., 2018; Niinemets and Valladares, 2006; Simkin et al., 2016; Xu et al., 2015), 2) obtain reliable prediction intervals of the response variable (Heiskary and Bouchard, 2015; Kampichler and Sierdsema, 2018), and 3) reveal the limiting effect of the stressor on the response variable via the upper boundary of the stressor-response relationship (Fornaroli et al., 2016; Keeley et al., 2012; Youngflesh et al., 2017). The upper boundary of a stressor-response relationship illustrates the behavior of response variable when the stressor is the limiting factor (Cade et al., 1999; Sankaran et al., 2005).

Because the CHL-DMS relationship represents a stressor-response relationship (McDowell et al., 2018), QR seems applicable and helpful to enhance the understanding of the CHL-DMS relationship. Considering the recent finding on the shifting nature of CHL-DMS relationships (Deng et al., 2020), a simple linear QR might not be adequate. A QR method with the ability to detect a change point is required but has rarely been explored (an exploration of this approach could be found in Zhou et al. (2015) who proposed a sequential change point detection method for linear QR).

In this study, we propose a novel Bayesian change point quantile regression (BCPQR) approach to investigate the CHL-DMS relationship in aquatic ecosystems. The BCPQR model integrates two well-developed Bayesian models: a Bayesian change point (BCP) model (Barry and Hartigan, 1993; Erdman and Emerson, 2007) and a Bayesian quantile regression (BQR) model (Benoit and Van den Poel, 2017; Yu and Moyeed, 2001). Both the BCP model (Beckage et al., 2007; Liang et al., 2019; Thomson et al., 2010) and the BQR model (Barneche et al., 2016; Uranchimeg et al., 2018; Yu et al., 2019; Zou and Shi, 2020) have been recently introduced and applied to develop a stressor-response relationship in environmental and ecological fields. However, to our knowledge, this is the first proposal of BCPQR model in environmental and ecological studies.

There are several features of the BCPQR model that makes it desirable for ecological investigations. First, the BCPQR model inherits advantages of the BQR model and the BCP model. It is expected to be able to provide a complete view on the stressor-response relationship (Muller et al., 2018; Xu et al., 2015). The detection of any change point in the regression intercept, slopes, and/or variance of residuals is possible (Beckage et al., 2007; Liang et al., 2019). Second, the Bayesian framework would provide the convenience for parameters estimation. We can straightforwardly incorporate the change point into the BQR model structure. Parameter estimation of BCPQR model could then be achieved using Markov-chain Monte Carlo (MCMC) methods (Qian et al., 2003). Moreover, the parameter estimation framework would allow for the calculation of probability densities representing the uncertainty of parameters (including the change point and the other model parameters) (Ellison, 2004; Gende et al., 2011; Underwood et al., 2017). In addition, based on posterior distributions of parameters, comparing parameters is straightforward (Alameddine et al., 2011; Qian et al., 2009). Finally, prior information – if available – could be used during model development (Ellison, 1996; 2004).

Our objective was to examine whether or not the BCPQR approach can enhance the understanding of CHL-DMS relationships in lakes and seas. We applied the proposed BCPQR model to reevaluate shifting CHL-DMS relationships revealed in Deng et al. (2020). We separately fitted BCPQR models at five regression quantiles. To avoid over confidence in the change point model, a common practice is comparing the change point model with a model without any change point (Cahill et al., 2015; Liang et al., 2019). Therefore, we also fitted a BQR model at each regression quantile and compared performances of the two models as a means to select the best model for characterizing the CHL-DMS relationship.

2. Materials and methods

2.1. Data source

Observations of CHL, DMS, and pH in lakes were directly obtained from Deng et al. (2020), in which the authors sampled 246 sites from 100 shallow lakes in China. Locations of these lakes range from 111°E to 122°E in longitude and from 28°N to 39°N in latitude. CHL concentrations varied widely, ranging from 0.55 μg/L to 58 μg/L, with an average of 11.87 μg/L and a standard deviation of 10.95 μg/L. The average DMS concentration was 175 ng/L, with a standard deviation of 189 ng/L.

In seas, Deng et al. (2020) complied 426 paired observations of CHL and DMS from 20 contributors of the Global Surface Seawater DMS Database (https://saga.pmel.noaa.gov/dms/). The sampling occurred during 1981 and 2012. According to the contributors table (Supplementary Table 2 in Deng et al. (2020)), we found 25, rather than 20, contributors meeting the data filtering requirements. In our analysis, all the data from these 25 contributors were included. We aggregated data by the sampling year and location (longitude and latitude), so that yearly site-specific averages of CHL and DMS were used to develop the CHL-DMS relationship in seas. The final sample size was 497 rather than the 426 in Deng et al. (2020). CHL concentrations ranged from 0.04 μg/L to 57.71 μg/L, with an average of 3.11 μg/L and a standard deviation of 6.11 μg/L. The average DMS concentration was 378 ng/L, with a standard deviation of 540 ng/L. Detailed information on the 25 contributors, code for data aggregation, and the aggregated CHL and DMS data can be found in the supplementary materials.

2.2. Model development

We fitted separate CHL-DMS relationship for lakes and seas at five regression quantiles (0.1, 0.3, 0.5, 0.7, 0.9). We did not explore more extreme regression quantiles (e.g., 0.01, 0.05, 0.95 or 0.99) because the uncertainty of estimated parameters increases (Scharf et al., 1998) with less data available at the tails of distribution of the response variable. For each regression quantile of a certain ecosystem type, we first developed a BQR model and a BCPQR model, respectively. Then, we compared performances of these two models based on the deviance information criterion (DIC) and selected the model with better performance as our top-ranked model. Consideration of both models is helpful for the development of a more reliable relationship – compared to relying on either model independently (Cahill et al., 2015). In the following subsections we introduce the BQR model and the BCPQR model. Because the estimation of the change point can be straightforwardly incorporated into the modeling framework of the BQR model, we did not separately introduce the BCP model. For detailed information on the BCP model, please refer to Beckage et al. (2007) or Erdman and Emerson (2007).

2.2.1. Bayesian quantile regression

A simple linear QR model to estimate the CHL-DMS relationship at a specific regression quantile (p, $0 < p < 1$) can be expressed by Eq. 1:

$$y_i = \alpha_0^p + \alpha_1^p x_i + \varepsilon_i^p, \tag{1}$$

where y represents the $\log_{10}$ transformed DMS concentration (ng/L), x represents the $\log_{10}$ transformed CHL concentration μg/L), and i is the order of observations. α_0^p and α_1^p are the regression intercept and slope at the p quantile, respectively. ε^p represents the error. The parameter estimation is based on the minimization of weighted sums of absolute deviations (Koenker and Bassett, 1978; Muller et al., 2018), which can be expressed by Eq. 2 in our case:

$$\min_{\alpha_0,\,\alpha_1} \sum_i \rho_p\left(\varepsilon_i^p\right) = \min_{\alpha_0,\,\alpha_1} \sum_i \rho_p\left(y_i - \alpha_0^p - \alpha_1^p x_i\right), \tag{2}$$

where ρ_p is the loss function and can be expressed by Eq. 3 for a given value u:

$$\rho_p(u) = u(p - I(u < 0)), \tag{3}$$

where $I(u < 0)$ means the value will be one if $u < 0$, and zero otherwise.

The BQR was first proposed by Yu and Moyeed (2001), in which the loss function (Eq. 3) was revealed to be equivalent to the maximization of a likelihood function formed by combining independently distributed asymmetric Laplace densities. Since then, asymmetric Laplace distribution (ALD) has been widely used for the BQR model (Alhamzawi, 2018; Benoit and Van den Poel, 2017). Benefiting from the attractive feature of ALD, the error can be expressed by Eq. 4 (Kotz et al., 2001; Wang et al., 2016; Zou and Shi, 2020):

$$\varepsilon_i^p = \frac{1-2p}{p(1-p)} W_i + \sqrt{\frac{2W_i}{\delta^p p(1-p)}} Z_i, \tag{4}$$

where δ^p represents the precision parameter of the ALD, W is an exponentially distributed random variable with a rate of δ^p, and Z is a random variable with a standard normal distribution. It is wroth noting that the two random variables, W and Z, are independent (Kotz et al., 2001; Zou and Shi, 2020).

As such, according to Eqs. 1 and 4, the distribution of y can be expressed by Eq. 5:

$$y_i \sim N\left(\alpha_0^p + \alpha_1^p x_i + \frac{(1-2p)W_i}{p(1-p)},\ \frac{2W_i}{\delta^p p(1-p)}\right). \tag{5}$$

Then, posterior distributions of all the parameters can be deduced by Eq. 6:

$$\pi\left(\alpha_0^p, \alpha_1^p, \delta^p\right) \propto \begin{array}{l} \prod_{i=1}^n N\left(y_i;\ \alpha_0^p + \alpha_1^p x_i + \frac{(1-2p)W_i}{p(1-p)},\ \frac{2W_i}{\delta^p p(1-p)}\right) \\ \times\pi\left(\alpha_0^p\right) \times \pi\left(\alpha_1^p\right) \times \pi\left(W_i \mid \delta^p\right) \times \pi\left(\delta^p\right), \end{array} \tag{6}$$

where n is the sample size. As noted by Yu and Moyeed (2001), standard conjugate prior distributions might not be available. However, according to Eq. 6, MCMC methods can be easily applied to calculate posterior distributions of unknown parameters (Chernozhukov and Hong, 2003; Kozumi and Kobayashi, 2011; Lee and Neocleous, 2010; Yu and Moyeed, 2001). For more detailed introductions on the derivation of the BQR model, please refer to Yu and Moyeed (2001), Lancaster and Jun (2009), or Zou and Shi (2020).

2.2.2. Bayesian change point quantile regression

The main functions of BCPQR model used in the CHL-DMS relationship development can be expressed by Eq. 7 and Eq. 8:

$$y_i = \beta_0^p + \beta_{1,\,\phi^p[i]}^p (x_i - cp^p) + \varepsilon_i^p, \tag{7}$$

$$\phi^p[i] = \begin{cases} 1, & x_i \leqslant cp^p \\ 2, & x_i > cp^p \end{cases}, \tag{8}$$

where cp^p represent the change point of $\log_{10}$ transformed CHL, at which the CHL-DMS relationship changes. $\beta_{1,1}$ and $\beta_{1,2}$ are regression slopes before and after the change point, respectively. $\phi^p[i]$ is the step function controlling the index of regression slope. β_0 is the estimated value of $\log_{10}$ transformed DMS at the change point. We did not seek to estimate two different regression intercepts, but used Eq. 7 to make sure the continuity of relationship at the change point.

Comparing functions of the BCP model with those of BCPQR model, we can find that the major difference is the addition of a change point in the BCPQR model structure. Benefiting from the flexibility of Bayesian framework, it is straightforward to incorporate the estimation of a change point into the parameter estimation process of the BQR model. Specifically, we can modify the parameter estimation process for a Bayesian linear regression model to that of a BCP model (Cahill et al., 2015; Liang et al., 2019). Following similar steps of Eq. 2 – 4, we can easily obtain the distribution expression of y in the BCPQR model (Eq. 9):

$$y_i \sim N\left(\beta_0^p + \beta_{1,\phi^p[i]}^p (x_i - cp^p) + \frac{(1-2p)W_{\phi^p[i],i}}{p(1-p)},\ \frac{2W_{\phi^p[i],i}}{\delta_{\phi^p[i]}^p p(1-p)}\right). \tag{9}$$

Accordingly, posterior distributions of all the parameters in the BCPQR model can be expressed by Eq. 10, which can be further deduced using MCMC methods (Wang et al., 2016):

$$\begin{aligned} & \pi\left(\beta_0^p, \beta_{1,1}^p, \beta_{1,2}^p, \delta^p, cp^p\right) \\ \propto\ & \prod_{i=1}^n N\left(y_i;\ \beta_0^p + \beta_{1,\phi^p[i]}^p (x_i - cp^p) + \frac{(1-2p)W_{\phi^p[i],i}}{p(1-p)},\ \frac{2W_{\phi^p[i],i}}{\delta^p p(1-p)}\right) \\ & \times \pi\left(\beta_0^p\right) \times \pi\left(\beta_{1,1}^p\right) \times \pi\left(\beta_{1,2}^p\right) \times \pi\left(W_{\phi^p[i],i} \mid \delta_{\phi^p[i]}^p\right) \times \pi \\ & \left(\delta_{\phi^p[i]}^p\right) \times \pi\left(cp^p\right). \end{aligned} \tag{10}$$

2.2.3. Model selection

To conduct the model selection between the BQR model and the BCPQR model, we calculated DIC values for each pair of models. DIC combines a measure of goodness-of-fit and a measure of model complexity (Spiegelhalter et al., 2002). DIC is a reliable criterion and has been widely used for Bayesian model selection (Cahill et al., 2015; Liang et al., 2019; Meyer, 2016). A smaller DIC indicates a better model. Generally, the model with a smaller DIC is strongly supported if the difference of DIC values between the two moddels is larger than 10 (Ribatet, 2020).

2.2.4. Parameter comparisons

Parameter comparison aims to test whether or not a significant difference exists between two estimated parameters. For Bayesian models, posterior distributions of parameters can be conveniently used to calculate the posterior distribution of the difference between two parameters (Qian et al., 2009). In this study, to compare the difference of paired parameters, we used 3000 samples from the posterior distributions of the parameters of interest and derived the posterior distribution of the difference between the parameters being compared. We then calculated the 95% credible interval of the difference and if the 95% credible interval covered zero, then we deduced that the two parameters were not significantly different. Otherwise, the two parameters were considered significantly different.

We used parameter comparisons to examine the equality of regression slopes at multiple regression quantiles. Classical mean regression methods, including piecewise regression, should meet the assumption of equal variance (Cade and Noon, 2003). Conventionally, comparing regression slopes of QR models at a range of regression quantiles is an effective tool to test the equal variance assumption (Das et al., 2019). If all the test results were not significant, then the equal variance assumption was assured. Otherwise, the assumption was violated and applications of mean regression methods were deemed inappropriate.

We also compared parameters of BCPQR models for lakes with those for seas. At each regression quantile, we tested differences of four regression parameters, including β_0, $\beta_{1,1}$, $\beta_{1,2}$, and cp in Eq. 7. In addition to indicating parameter differences, the comparison can also reveal differences between relationships in lakes and those in seas. If any test result was significant, then the model for lakes and that for seas would be considered significantly different.

Based on the results of BCPQR models, we can examine whether or not the BCPQR approach can enhance the understanding of CHL-DMS relationships. Based on previous studies utilizing Bayesian analysis (Cahill et al., 2015; Yu and Moyeed, 2001) and examining CHL-DMS relationships (Deng et al., 2020), we expected that the BCPQR model would provide new findings on three aspects of the CHL-DMS relationship. Specifically, the BCPQR model would 1) advance the statistical modeling of CHL-DMS relationships and therefore the inferences that can be made about the ecological implications of this relationship, 2) elucidate relationship differences between lakes and seas, and 3) identify factors that might mediate the CHL-DMS relationship. The modeling of CHL-DMS relationship includes providing a full view of the relationship at multiple regression quantiles, reflecting the limiting effect of CHL on DMS, and testing the violation of equal variance assumption. Specifically, based on the results of BCPQR models, we expected to be able to:

- show a relatively complete view of CHL-DMS relationships at multiple regression quantiles, reflecting responses across much of the DMS distribution;
- illustrate the limiting effect of CHL on DMS using the upper boundary of relationship;
- test the equality of regression slopes at multiple regression quantiles to determine whether or not a mean regression method is adequate to develop a reliable CHL-DMS relationship;
- detect relationships differences between lakes and seas at multiple regression quantiles;
- explore whether or not another variable (i.e. pH) influences the limiting effect of CHL on DMS in lakes.

All computations were conducted using the R software (version 4.0.2, R Core Team, 2020). We used the JAGS software (version 4.3.0, Plummer, 2017) for parameter estimation of all the BQR and BCPQR models. We called JAGS from R using the `rjags` (Plummer, 2019) and `R2jags` (Su and Yajima, 2020) packages. Diffuse priors were used for all the parameters. We used four chains with random initial values and ran 1,000,000 iterations for each chain. The first half of the iterations were used for the burn-in process, while the second half were used for summarizing the posterior distribution. Model convergence was evaluated by the Gelman-Rubin convergence statistic ($\hat{R}$) (Brooks and Gelman, 1998) and assured by $\hat{R} < 1.10$ (Filstrup et al., 2017). Code for model development and parameters estimation are available in the supplementary materials.

Table 1
Model comparison results using DIC to compare a Bayesian quantile regression (BQR) model and a Bayesian change point quantile regression (BCPQR) model for describing CHL-DMS relationships in lakes and seas. DIC values are provided for each model and quantile. The DIC difference ("BQR - BCPQR") at each regression quantile was calculated by subtracting the DIC value of the BQR model from the DIC value of the BCPQR model.

Waterbody	Model	Regression quantile				
		0.1	0.3	0.5	0.7	0.9
Lakes	BQR	386.8	348.6	313.2	332.1	335.0
	BCPQR	175.2	226.3	233.1	267.2	275.0
	BQR - BCPQR	211.6	122.3	80.2	64.8	60.0
Seas	BQR	827.6	759.1	712.3	753.1	770.2
	BCPQR	681.3	637.0	624.0	683.6	722.5
	BQR - BCPQR	146.3	122.1	88.4	69.5	47.7

3. Results

3.1. Model selection

DIC values for each pair of BQR and BCPQR models at each regression quantile in lakes or seas are summarized in Table 1. The DIC difference was calculated by subtracting the DIC value of the BQR model from the DIC value of BCPQR model. For all model pairs, the BCPQR model had a much smaller DIC value compared with the BQR model (DIC differences $>$ 47 for all comparisons; Table 1). According to Ribatet (2020), a DIC difference larger than 10 indicates that the model with a larger DIC value has no support relative to the model with the lowest DIC value. Therefore, DIC results support the BCPQR model for describing the CHL-DMS relationship at all regression quantiles and in both lakes and seas.

3.2. Parameter estimation

Because the BCPQR model was the top-ranked model based on DIC, we only reported parameter estimates for the BCPQR models. Posterior distributions of the four key parameters are shown in Fig. 1. Because regression slopes were all positive before the change point and post-change point slopes were all negative, the regression parameter, β_0, can be interpreted as the maximum of $\log_{10}$DMS at the corresponding regression quantile according to Eq. 7, as noted in Fig. 1a. The parameter β_0 increased with increasing regression quantile, which is expected because the observed DMS should be greater at higher quantiles. From the 0.1 quantile to the 0.9 quantile, the posterior mean of β_0 increases from 2.08 to 2.98 for lakes and from 2.16 to 3.26 for seas, corresponding to an average increase of DMS from 120 ng/L to 955 ng/L for lakes and from 145 ng/L to 1820 ng/L for seas.

As for the estimated change points of $\log_{10}$CHL (*cp*, Fig. 1b), posterior means ranged from 0.656 to 0.828. This corresponds to CHL concentrations on the raw scale of 4.53 μg/L and 6.73 μg/L, which is a relatively narrow range. Comparing change points of BCPQR models for lakes and those for seas, there are large overlaps in posterior distributions at all the regression quantiles and the posterior means of the change points are very similar, except for those at the 0.1 regression quantile.

Posterior means of the slopes before the change point for lakes varied substantially with regression quantile, while those for seas were relatively stable (Fig. 1c). Estimates for lakes are also more uncertain compared to estimates for seas. However, for the slopes after the change point, the opposite is true. That is, the posterior means of slopes after the change point for lakes were relatively stable, while those for seas varied with regression quantile and are more uncertain (Fig. 1d). In addition, for seas, the absolute value of slopes increases before the change point with increasing regression quantile (Fig. 1c) and decreases after the change point (Fig. 1d). For lakes, before the change point, the regression slope decreases except at the 0.9 quantile (Fig. 1c). After the change point, the regression slopes show no apparent pattern (Fig. 1d).

4. Discussion

4.1. Relationships at multiple regression quantiles

While a mean regression method, e.g., the piecewise linear regression used in Deng et al. (2020), focuses on the mean of DMS distribution, the BCPQR model revealed a more complete view of the CHL-DMS relationships at multiple regression quantiles (Fig. 2), thus allowing for a more thorough understanding of the response of DMS to changes in CHL across the DMS distribution.

In practice, it is difficult to obtain the *true* upper boundary of the relationship due to the lack of data at the tail ends of the distribution. However, the model at an upper regression quantile provides a reasonable approximation of the *true* upper boundary (Cade et al., 1999). Therefore, if we are interested in the upper boundary of the CHL-DMS relationship, which generally represents the limiting effect of CHL on DMS, we can set the regression quantile to be 0.9 (top curves in Fig. 2a & b). Similarly, we could explore the relationship at the 0.1 quantile if we want to explore the relationship when DMS is mainly limited by factors other than CHL. Furthermore, combining prediction results at the 0.1 and 0.9 quantiles results in the 80% prediction interval of the DMS distribution, if the range of the DMS distribution is of interest.

It is also convenient to conduct uncertainty analysis for regression parameters (Fig. 1) and for predicted DMS values (shaded regions in Fig. 2). In addition, the results provide a visual comparison of relationship differences at multiple regression quantiles. For the relationship in seas, DMS increases faster before the change point, but reduces slower after the change point with increasing regression quantile (Fig. 2b, also revealed by the parameter estimation results in Fig. 1). The CHL-DMS relationship in lakes varies but shows no monotonic trend with the change of regression quantile (Fig. 2a). The variations among regression slopes (Fig. 1) and the relationship differences at multiple regression quantiles (Fig. 2) reflect varied responses to CHL among different parts of DMS distribution. These observed differences in CHL-DMS relationships at different quantiles and before and after change points highlights the fact that multiple mechanisms and processes are likely involved in governing CHL-DMS dynamics and that these drivers may differ between seas and lakes. Currently, we do not have specific hypotheses regarding what drivers might be involved; however, identifying primary drivers and if these drivers vary in importance as a function of CHL or DMS concentrations represents an area of meaningful future research.

4.2. Limiting effect of CHL on DMS

The theoretical basis of limiting effect comes from Liebigs law of the minimum, in which a limiting factor is the one least available among those factors affecting the response variable (Cade et al., 1999). In our analysis, the limiting effect is when CHL is the limiting factor of DMS – which can be illustrated by the upper quantile of the CHL-DMS relationship (Fig. 3). In contrast, piecewise linear regression results (solid lines in Fig. 3) cannot quantify such a limiting effect (Cade et al., 1999).

Following common practice of QR (Joseph et al., 2016; Keeley et al., 2012; Mönkkönen et al., 2017), we employed the relationship deduced by the BCPQR model at the 0.9 regression quantile as the upper boundary of CHL-DMS relationship to reflect the limiting effect of CHL on DMS. We back-transformed $\log_{10}$CHL and

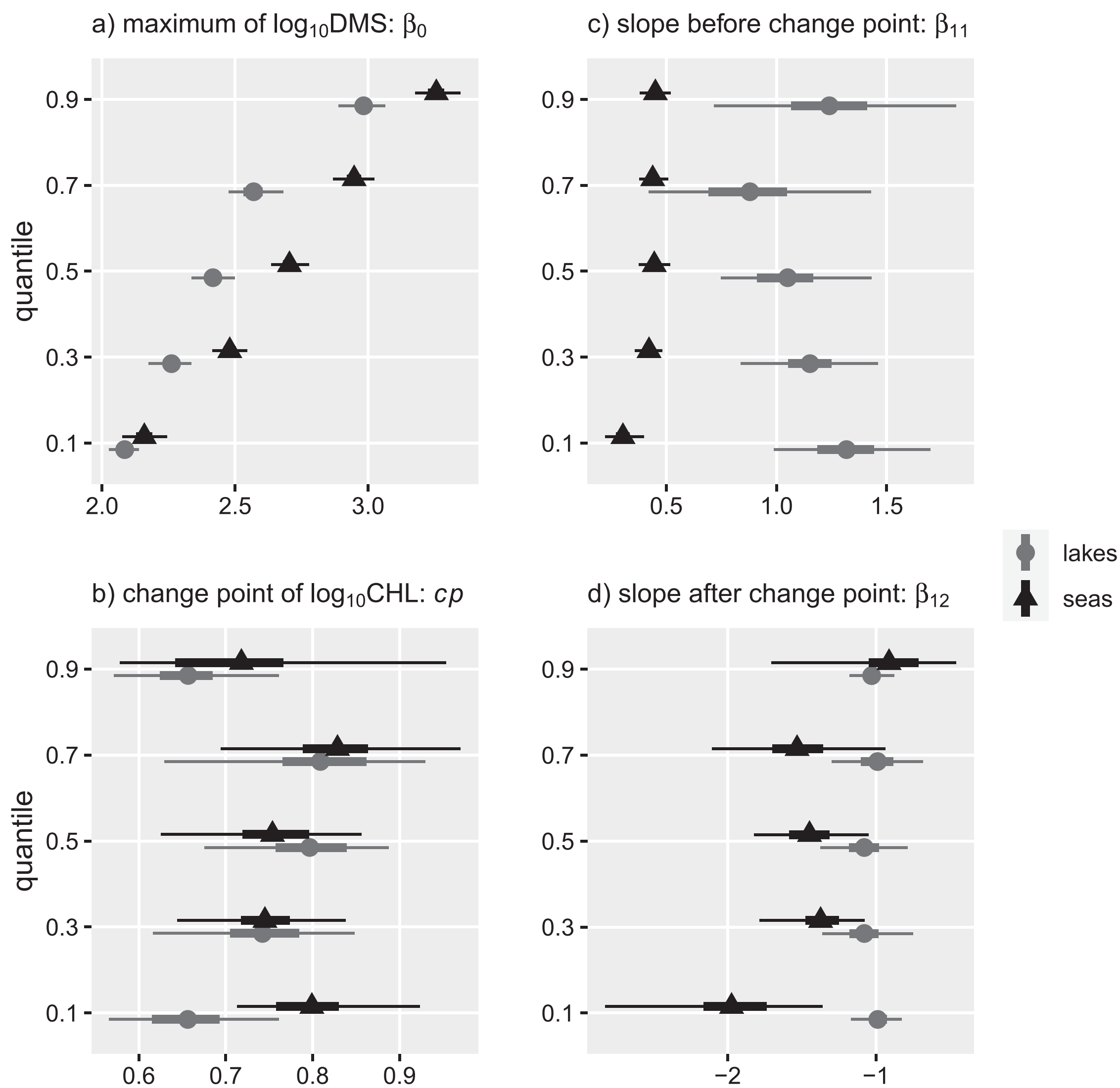

Fig. 1. Posterior summaries of BCPQR model parameters at multiple regression quantiles for lakes (grey symbols) and seas (black symbols). Solid points represent posterior means, while the thin and thick lines represent the 50% and 95% credible intervals, respectively.

$\log_{10}$DMS to their raw scales (Fig. 3). On the raw scales, the difference between the upper boundary of CHL-DMS relationship in lakes and that in seas is apparent. The upper boundary in seas is much greater than that in lakes. In addition, before the change point, DMS increases more sharply with an increase in CHL in seas when compared to that in lakes. Both upper boundaries are right skewed, which means with the increase of CHL, DMS increases faster before the change point and then decreases slower after the change point.

4.3. Equality of regression slopes

The examination of equality of regression slopes can determine whether or not a mean regression method violates the equal variance assumption (Das et al., 2019). Parameter differences of paired slopes are shown in Fig. 4. For slope differences of BCPQR models in seas, 95% credible intervals of several paired slopes (e.g., "0.5–0.1" in Fig. 4a and "0.9–0.1" in Fig. 4b) do not cover zero, indicating significant differences. Therefore, the equal variance assumption is violated. That is, mean regression methods are not appropriate for developing the CHL-DMS relationship in seas. Although some mean regression methods could be used to deduce some quantiles of the DMS distribution (Borsuk et al., 2002), it can be problematic when trying to predict some quantiles of DMS distribution using a mean regression method because of the violation of the equal variance assumption (Cade and Noon, 2003; Das et al., 2019).

For the lake model, there were no significant differences between paired slopes (Fig. 4). However, the sample size is relatively small and uncertainties of regression slopes were large (Fig. 1c & d), which contributed to the insignificance of parameter differences. Before additional data are included to further confirm results of parameter differences, piecewise linear regression methods might be used with caution for the development of CHL-DMS relationship in lakes.

While the piecewise linear regression method might not be appropriate for the development of CHL-DMS relationship in seas, the BCPQR model is not constrained by the equal variance assumption (Cade and Noon, 2003) and thereby can be used as a reliable and robust tool for DMS prediction. Because the median regression is a robust alternative to ordinary linear regression (Koenker and Hallock, 2001), the BCPQR model at the 0.5 regression quantile can act as an effective alternative to the piecewise linear regression. In addition, because of the violation of equal variance assumption, the piecewise linear regression should not be used to deduce predicted quantiles of the DMS distribution (Das et al., 2019); whereas, re-

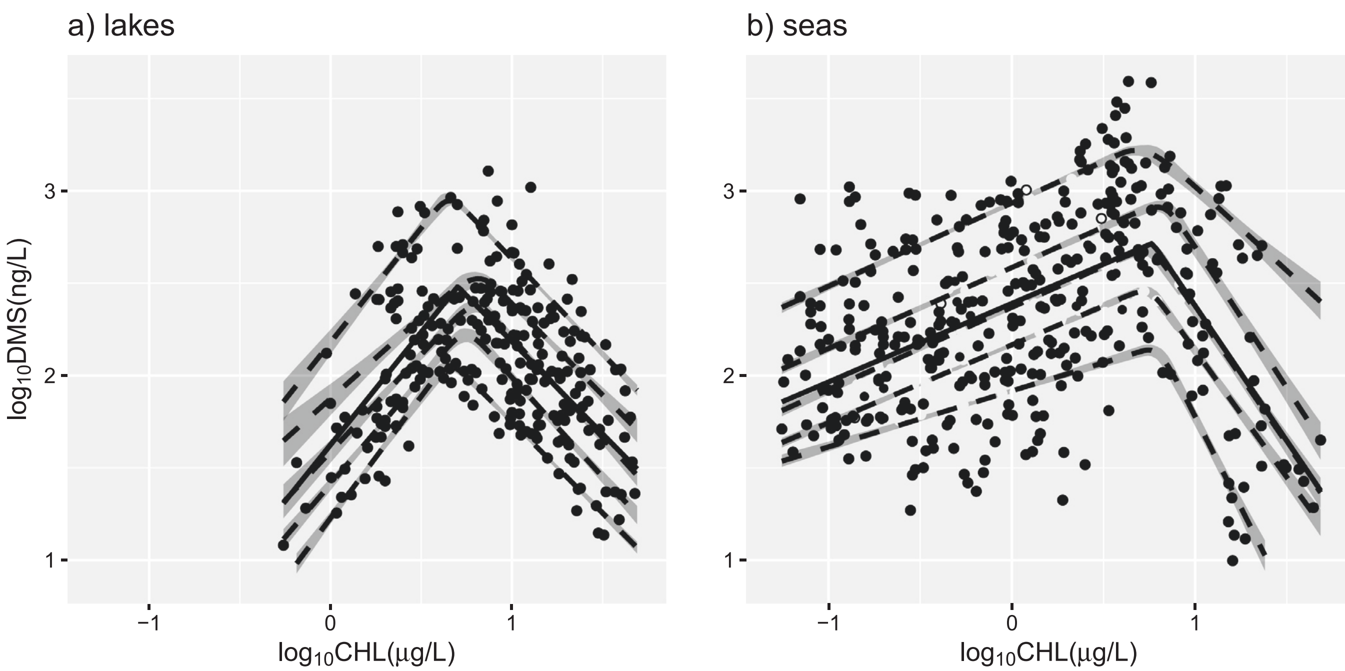

Fig. 2. Estimated fitted lines of the CHL-DMS relationship from the BCPQR model for lakes (a) and for seas (b). From bottom to top, dashed lines represent posterior means of predicted $\log_{10}$DMS at the 0.1, 0.3, 0.5, 0.7, and 0.9 quantiles. The corresponding shaded region represent the 50% credible intervals. We show the 50% credible intervals to improve visual interpretation. For the 95% credible intervals, please refer to Fig. S1. Solid lines are results of piecewise regression (obtained using the `segmented` package in R (Muggeo, 2008)), which are close to coresponding lines of BCPQR models at the 0.5 regression quantile.

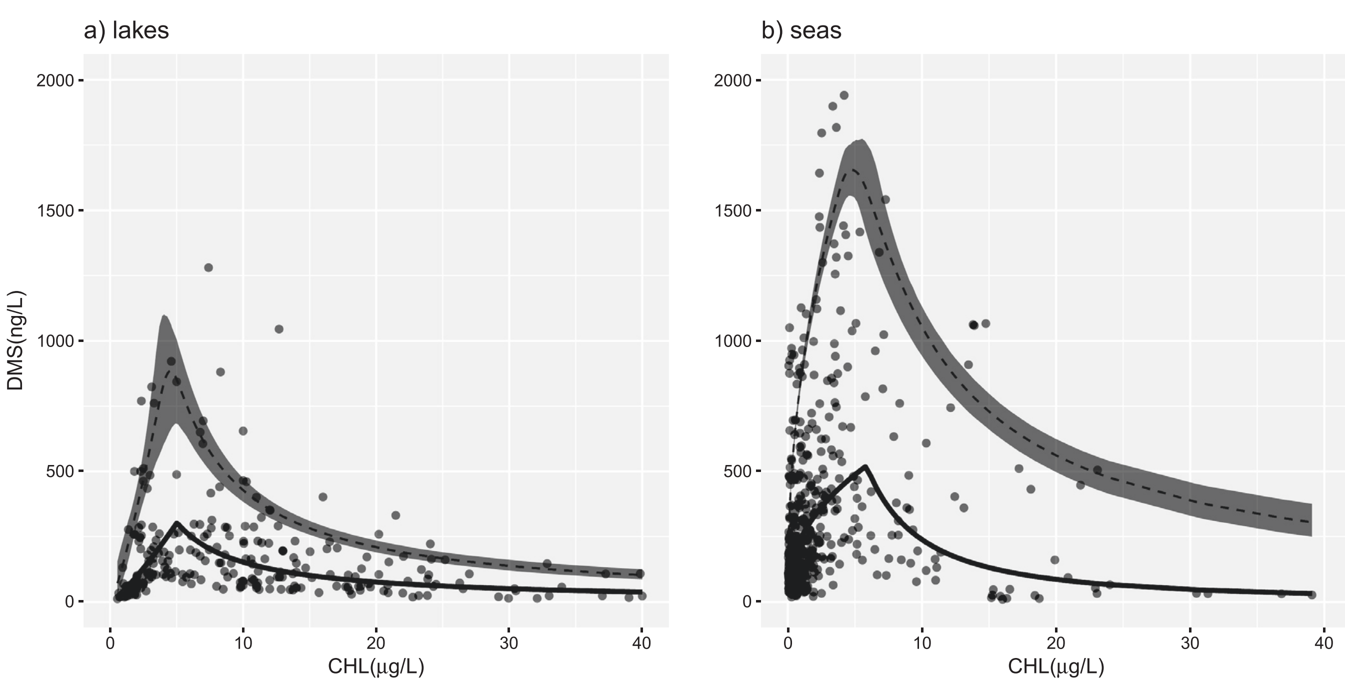

Fig. 3. The limiting effect of CHL on DMS estimated by the CHL-DMS relationship at the 0.9 regression quantile in lakes (a) and in seas (b). Lines represent means of predicted DMS concentrations. The corresponding shaded regions represent the 95% credible intervals of predicted DMS concentrations. For comparison, solid lines are piecewise linear regression results of the $\log_{10}$CHL − $\log_{10}$DMS relationship obtained using the `segmented` package in R (Muggeo, 2008).

liable intervals of predicted DMS distributions are possible using the BCPQR model (Heiskary and Bouchard, 2015; Kampichler and Sierdsema, 2018).

4.4. Relationship differences between lakes and seas

We compared the CHL-DMS relationships in lakes to those in seas by comparing the parameters of the BCPQR models at multiple regression quantiles (Fig. 5). The parameter comparisons provide statistical evidence that the CHL-DMS relationship does in fact differ between lakes and seas. Considering that β_0 in the BCPQR model represents a $\log_{10}$ transformed DMS concentration (Eq. 7, Fig. 1a), when calculating the difference, we retransformed β_0 to the raw unit of DMS (ng/L, Fig. 5a). With increasing regression quantile, the difference of maximum DMS increases. Specifically, at the 0.7 and 0.9 quantiles, posterior means of the differences are

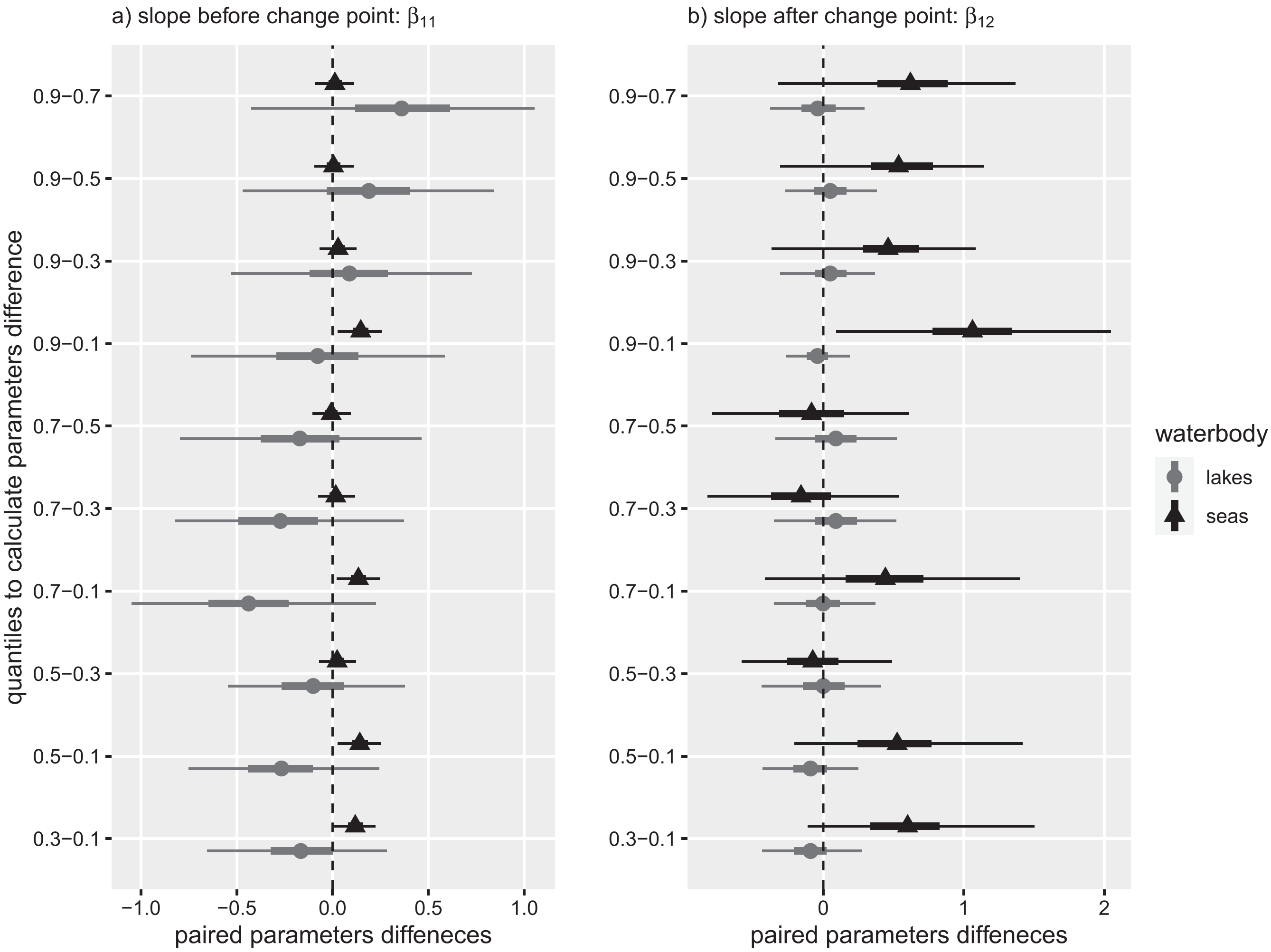

Fig. 4. Posterior summaries of differences between slopes estimated at different quantiles for lakes (grey symbols) and seas (black symbols). Labels on the *y*-axis represent corresponding regression quantiles of BCPQR models, whose slopes are compared. For example, "0.9–0.7" means the posterior difference is calculated by subtracting the slope at the 0.7 quantile from the slope at the 0.9 quantile. A dashed line at zero is added to aid interpretation. Solid points represent posterior means of the differences. The thin and thick lines represent the 50% and 95% credible intervals, respectively. Differences in slopes with 95% credible intervals that do not overlap with zero are considered significantly different.

more than 500 ng/L (DMS concentratons in seas are higher than in lakes), which are large considering the average DMS concentrations for lakes and seas are only 175 ng/L and 378 ng/L, respectively.

We also retransformed the *cp* parameter into the raw units of CHL concentration μg/L) to calculate differences between change points for the lake and seas models (Fig. 5b). We found no significant differences. Particularly, posterior means of differences were close to zero, ranging from -1.78 μg/L to 0.59 μg/L. Also, for each ecosystem type, tests on paired *cp* values showed no significant differences for all combinations of regression quantiles (Fig. S2). These results indicate that the change point of CHL at which the CHL-DMS relationship changes is consistent at multiple regression quantiles and for different ecosystem types. While Deng et al. (2020) revealed that change points were very close for lakes and seas using the piecewise linear regression method, we confirmed this conclusions at multiple regression quantiles based on results from the BCPQR models. It is worth emphasizing, as previously mentioned, that despite the relatively consistent location of CHL change points, corresponding DMS concentrations are substantially different at some regression quantiles (Fig. 5a).

Before the change point, regression slopes of BCPQR models for lakes are significantly larger than those for seas, except for those at the 0.7 regression quantile (Fig. 5c). While Deng et al. (2020) found that the slope before the change point for lakes were almost twice as large as the value for seas (1.22 and 0.62, respectively), our results show that the ratio is not as simple as two at varied quantiles. According to the results of BCPQR models (Fig. 1c), the ratio would be 4.3, 2.7, 2.4, 2.0, and 2.8 for the five regression quantiles from 0.1 to 0.9, respectively. As for regression slopes after the change point, the only significant difference was at the 0.1 regression quantile (Fig. 5d).

Considering significant differences of some regression parameters between models for lakes and those for seas, we conclude that the CHL-DMS relationship in lakes and that in seas are different at each regression quantile, despite the consistency of CHL change points. Obviously, the piecewise linear regression method cannot reveal the consistency of CHL change points for lakes and seas and cannot examine parameter and relationship differences at multiple regression quantiles.

The CHL-DMS relationship differences between lakes and seas indicates that some drivers or processes controlling the emission of DMS are likely to be different in lakes and seas. Deng et al. (2020) revealed the effect of pH on the CHL-DMS relationship in lakes and also extended the finding to seas. However, based on our results, currently, transferring any potential mechanisms controlling DMS in either lake or sea ecosystems

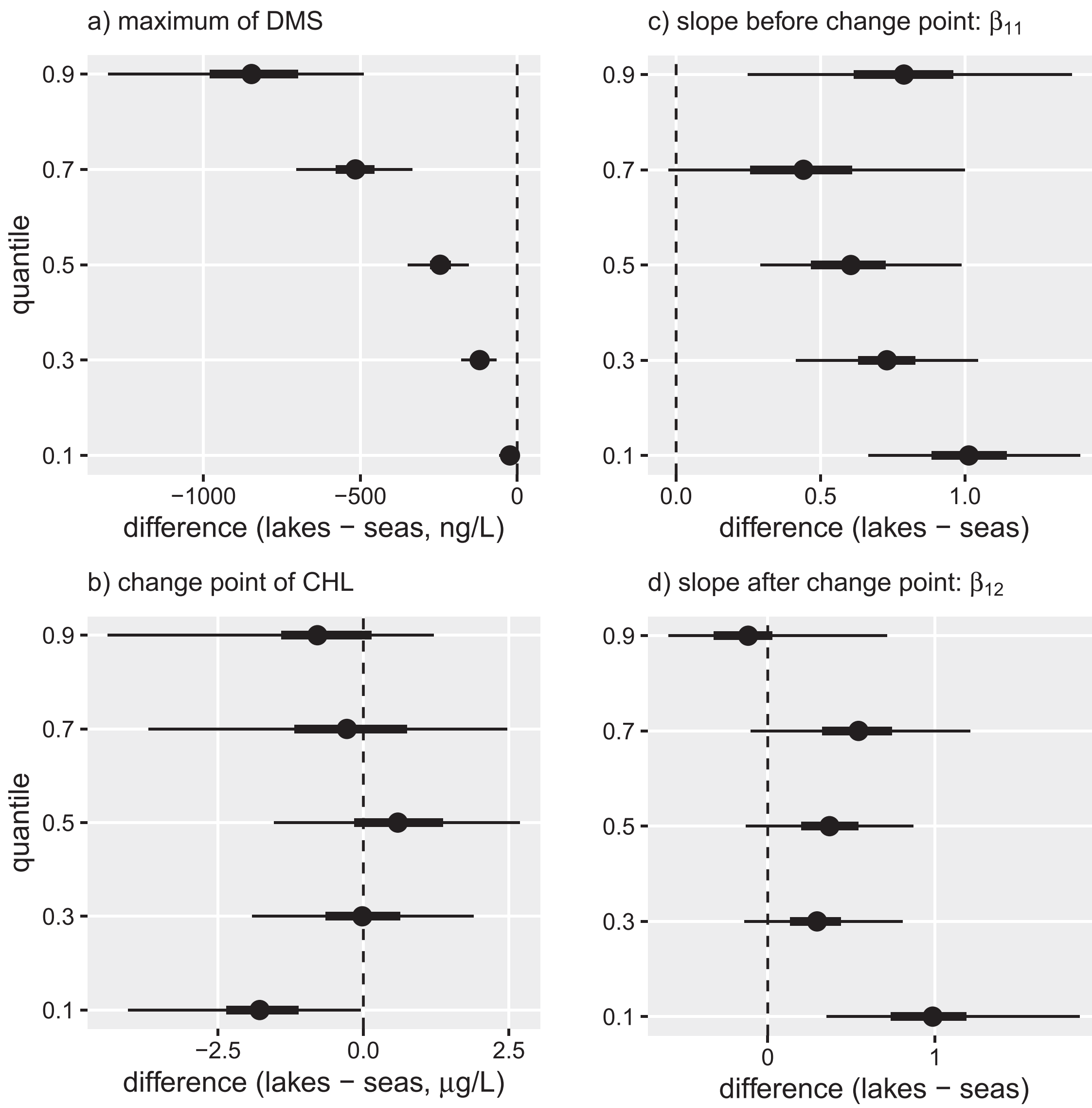

Fig. 5. Posterior summaries of differences between parameter estimates for lakes and seas from BCPQR models. The posterior difference was calculated by subtracting the parameter estimate for seas from the parameter estimate for lakes. A dashed line at zero is added to aid interpretation. Solid points represent posterior means of the difference between lakes and seas. The thin and thick lines represent the 50% and 95% credible intervals, respectively. Differences in parameters with 95% credible intervals that do not overlap with zero are considered significantly different.

into the other ecosystem may be misleading. A possible explanation for the ecosystem-dependent relationship might be the difference in salinity between lakes and seas, with DMS emission likely increasing with increasing salinity concentrations (Gibson et al., 1991; Taalba et al., 2013) – concentrations that promote the production of dimethylsulfoniopropionate (major precursor for DMS) (Curson et al., 2017) Importantly, however, additonal efforts are needed to explore drivers and processes leading to the relationship differences between lakes and seas.

4.5. Impact of pH on CHL-DMS relationship

The pH was emphasized to be the key factor impacting the CHL-DMS relationship in Deng et al. (2020). Specifically, the emission of DMS would increase with increasing CHL when pH $<$ 8.1, and would decease when pH 8.1 (Deng et al., 2020). We reevaluated the effect of pH on the limiting effect of CHL on DMS. We first defined a term, emission potential, to represent the difference between the maximum emission (when DMS was solely limited by CHL) and observed DMS emission. The emission potential can be calculated by the predicted upper boundary minus observed DMS values. Next, assuming that pH is the key factor impacting DMS emission, pH should explain a large proportion of variation of emission potential. Specifically, since the pH threshold value favoring DMS emission is 8.1, the emission potential of DMS should decrease with increasing pH when pH $<$ 8.1, and should increase with additional increases in pH when pH 8.1. That is, the observed DMS should become closer to the upper boundary and the emission potential should become smaller when pH approaches 8.1.

To examine whether or not the above assumption was reasonable, we used the predicted DMS emissions deduced from the BCPQR model for lakes at the 0.9 regression quantile minus the observed DMS concentrations to reflect the emission potential of DMS (Fig. 6). Surprisingly, we found that pH does not explain significant variation in the DMS emission potential. The assumption that pH is a key factor impacting the CHL-DMS relationship is not supported by our results.

Although there is a significant change point of pH in the pH-DMS relationship (refer to Fig. 1c in Deng et al. (2020)), consid-

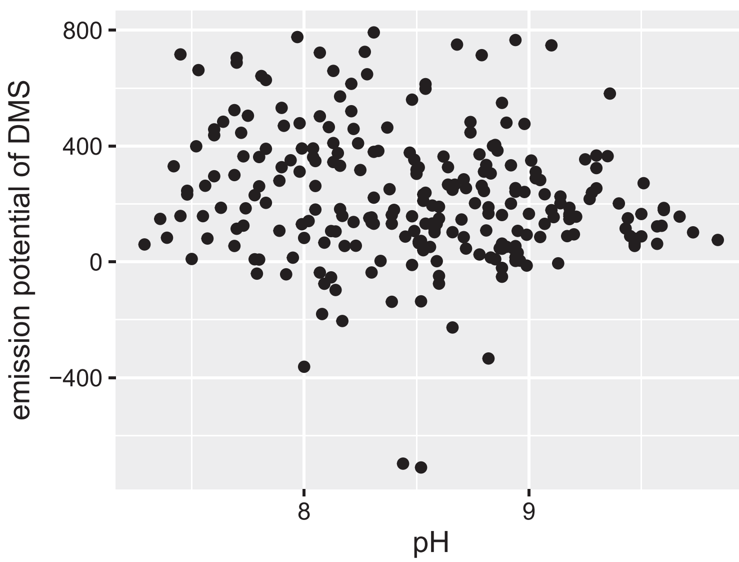

Fig. 6. The effect of pH on the emission potential of DMS in lakes. The emission potential is calculated by the predicted upper boundary (relationship deduced by the BCPQR model at the 0.9 regression quantile) minus the observed DMS value. Observed pH values were not available for seas, so the corresponding analysis was not performed.

ering the obvious positive pH-CHL correlation (refer to Fig. 1d in Deng et al. (2020)) and that CHL can be a causal variable impacting both pH and DMS (Charlson et al., 1987; Nakano and Watanabe, 2005), the causal effect of pH on the CHL-DMS relationship change point is likely a result of a statistical correlation resulting from a shared environmental driver (i.e., CHL drives DMS and pH). However, it is hard to distinguish the causal effect from correlation using a mean regression method. By contrast, the BCPQR model is a useful tool for such an analysis by introducing the emission potential and linking it to the possible driver. Note that the calculation of emission potential relies on the upper boundary deduced from the BCPQR model.

4.6. Implications for understanding CHL-DMS relationships

In this study, we proposed a novel BCPQR model to quantify the CHL-DMS relationship in aquatic ecosystems. Here, we summarize how the BCPQR approach enhanced the understanding of shifting CHL-DMS relationships by comparing it with the application of piecewise regression method:

- BCPQR models provided a relatively complete view of shifting CHL-DMS relationships at multiple regression quantiles, rather than only focusing on the mean of the DMS distribution, as done with piecewise linear regression.
- The upper boundary of the relationship was quantified by the BCPQR model at the 0.9 regression quantile, while the piecewise regression cannot show such an effect.
- We revealed that the equal variance assumption was violated when applying piecewise linear regression in seas, while the BCPQR model could be employed to predict different parts (e.g., the median or an interval) of the DMS distribution without being constrained by this assumption.
- We found consistency in change points of the CHL-DMS relationships for lakes and seas. However, CHL-DMS relationships in lakes and seas were substantially different at all regression quantiles. As such, we caution against transferring results and inferences from one ecoystem type to another.
- According to the upper boundary of CHL-DMS relationship, we introduced and calculated the emission potential of DMS. Using this metric, we showed that pH was not likely to be the key factor impacting the CHL-DMS relationship in lakes. While piecewise linear regression cannot distinguish causality from correlation, the application of BCPQR model might be helpful for causality detection.

4.7. Generalizations of the proposed approach

In practice, shifting stressor-response relationsships, e.g., the hump-shaped stressor-response relationship or other shapes of stressor-response relationship with a change point (e.g., the sign remains the same, but the magnitude of regression slope changes), are not rare in ecology (Keeley et al., 2012; Liang et al., 2019; Wagner and Midway, 2014). Most commonly, mean regression methods, such as the piecewise regression, have been applied to detect the change point and develop the shifting stressor-response relationship (Massicotte et al., 2017; Wang et al., 2017). In this study, we made a step forward in modeling shifting stressor-response relationships in environmental and ecological research. We have demonstrated that the BCPQR model is an effective tool to understand a shifting stressor-response relationship by identifying the change point, providing a complete view on the relationship across the distribution of the response variable, examining significance of parameter differences, making robust predictions, and conducting uncertainty analysis.

Moreover, benefiting from the flexibility of Bayesian framework, the BCPQR model can be easily modified, which will allow for it to be broadly applied in ecological investigations. Potential modifications to the BCPQR model to address other ecological questions include:

- Detecting change points over time. Time could be added as the predictor to estimate a temporal change point, similar to what Cahill et al. (2015) did for a BCP model.
- Multivariate regression. Adding other predictor variables and modeling change points is straightforward under a Bayesian framework (Liang et al., 2019).
- Detecting multiple change points. The model can be modified to accommodate multiple change points, which has been done using other modeling approaches (Beaulieu et al., 2009; Jochner and Menzel, 2015).
- Binary or discrete response variables. The CHL-DMS relationship is a continuous response case. In environmental and ecological studies, the response variable is often binary or discrete (e.g., count data) (Wagner and Midway, 2014). Since the BQR model can accommodate binary or discrete response variables (Benoit

and Van den Poel, 2017; Lee and Neocleous, 2010), the BCPQR model can as well.
- Hierarchical modeling. The hierarchical structure of regression parameters (e.g., regression slopes before and after the change point) can be incorporated into the modeling, as in a hierarchical BCP model (Authier et al., 2011; Thomson et al., 2010).

Therefore, the proposed BCPQR model is expected to have great potential for generalizing to other ecological questions, which will help to better understand shifting stressor-response relationships in environmental and ecological studies. Note that several other QR methods, such as polynomial regression (Fornaroli et al., 2016; Youngflesh et al., 2017), spline smoothing (Keeley et al., 2012), quantile regression forest (Kampichler and Sierdsema, 2018; Veronesi and Schillaci, 2019), and quantile regression neural networks (Cannon, 2011) can be used to fit a nonlinear stressor-response relationship and thereby might be applicable to the development of a shifting CHL-DMS relationship. Compared with these methods, the BCPQR model is parameterized to have fewer parameters, which can effectively avoid over fitting.

The novelty of our research is due to two primary factors: the proposal of the novel BCPQR model and the enhanced understanding of CHL-DMS relationships. In this study, for the first time, we integrated the BCP model and the BQR model to reveal shifting stressor-response relationships at multiple regression quantiles. The proposal of the novel approach thereby contributes to the research field of environmental and ecological modeling. We applied our novel model to reevaluate published CHL-DMS relationships in aquatic ecosystems, by which we found several meaningful and insightful findings regarding the CHL-DMS relationship insight that would not be possible using a traditional piecewise linear regression approach. The insight derived from the proposed approach may also help improve overall estimates of DMS emission from aquatic ecosystems.

5. Conclusions

Integrating the BCP model and the BQR model, we proposed a novel BCPQR model that was able to detect a change point in the QR. We employed the proposed approach to investigate the CHL-DMS relationship in aquatic ecosystems. We revealed new findings in the CHL-DMS relationship modeling, relationship differences between lakes and seas, and factors impacting the CHL-DMS relationship. We thereby concluded that the BCPQR model could indeed enhance the understanding of shifting CHL-DMS relationship. We believe that the BCPQR approach can be generalized to other cases where shifting stressor-response relationships are observed.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary material

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2021.117287

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