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A Bayesian Approach to Incorporating Spatiotemporal Variation and Uncertainty Limits into Modeling of Predicted Environmental Concentrations from Chemical Monitoring Campaigns

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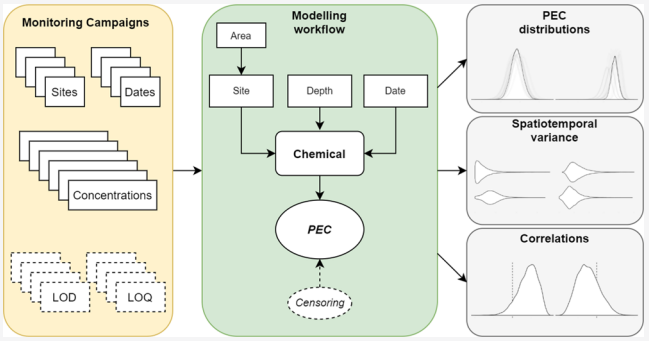
ABSTRACT: Environmental monitoring studies provide key information to assess ecosystem health. Results of chemical monitoring campaigns can be used to identify the exposure scenarios of regulatory concern. In environmental risk assessment (ERA), measured concentrations of chemicals can be used to model predicted environmental concentrations (PECs). As the PEC is, by definition, a predicted variable, it is highly dependent on the underlying modeling approach from which it is derived. We demonstrate the use of Bayesian distributional regression models to derive PECs by incorporating spatiotemporal conditional variances, and limits of quantification (LOQ) and detection (LOD) as *de facto* data censoring. Model accuracies increase when incorporating spatiotemporal conditional variances, and the inclusion of LOQ and LOD results in potentially more robust PEC distributions. The methodology is flexible, credibly quantifies uncertainty, and can be adjusted to different scientific and regulatory needs. Posterior sampling allows to express PECs as distributions, which makes this modeling procedure directly compatible with other Bayesian ERA approaches. We recommend the use of Bayesian modeling approaches with chemical monitoring data to make realistic and robust PEC estimations and encourage the scientific debate about the benefits and challenges of Bayesian methodologies in the context of ERA.



KEYWORDS: *Bayesian methodologies, chemical monitoring, environmental risk assessment, LOD, LOQ*

1. INTRODUCTION

Environmental monitoring is a key component in the assessment of ecosystem health.[1] The possibilities of Bayesian distributional regression models are explored to model the predicted environmental concentrations of a selection of Norwegian monitoring campaigns. It is used to characterize the status of different aspects of the natural environment, such as species abundance or abiotic physical and chemical properties. Environmental monitoring also considers the anthropogenic influence on natural ecosystems, for example, chemical pollution or climate change.[2] This is acknowledged by international legislatives, such as the European Union's Water Framework Directive (WFD) and Marine Strategy Framework Directive, the United States' Clean Water Act, or Japan's Water Pollution Control Law.[3−6] Water bodies like rivers, lakes, streams, and ponds, and also coastal areas and fjord systems, are crucial habitats for ecosystem functioning and hold additional economic and recreational value.[7] An important part of environmental monitoring campaigns, such as those performed to support the WFD, is therefore the chemical monitoring of water bodies and subsequent assessment of ecosystem health.[8]

Data collected in chemical monitoring campaigns can be used to model the predicted environmental concentration (PEC) of chemicals; when such information is not available, PECs are often derived from environmental fate modeling. PECs can be used in environmental risk assessment (ERA), where they are compared to predicted no-effect concentrations (PNECs). This is usually done by dividing the PEC by the PNEC, which results in a hazard quotient; after the application of assessment and safety factors, this becomes a risk quotient (RQ).[9] RQ values above 1 can be considered environmental risks, as the PEC surpasses the PNEC. Based on this, mitigation measures can be put in place, for example, remediation plans or quiescing of local pollutant sources.

A good estimation of both PEC and PNEC is therefore crucial for high-quality ERAs. For the PEC, the European

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Chemicals Agency (ECHA) explicitly encourages modeling approaches, also when measured environmental concentrations are available.[10] The European Food Safety Authority (EFSA) echoes the same sentiments.[11] This is partially motivated by the fact that environmental monitoring campaigns in the same area can span across various dates and multiple sites. The spatiotemporal variability of measured concentrations, for example, fluctuations and peaks of these measured concentrations, needs to be addressed in any modeling approach; otherwise, ambiguous derivations of PECs could lead to wrong decision-making.[11] Depending on the study or monitoring campaign, measurements can take place at several sites and depths within the same area and across time. Up to hundreds of substances can be measured simultaneously. In common practice, measured environmental concentrations are often summarized by taking a deterministic summary statistic, for example, the median concentration and 95% quantile of a substance across all samples. This collapses the information available per substance into a single data point, the measured environmental concentration (MEC) value, which can be heavily influenced by site- and date-specific variations.

Additionally, the measured chemical concentrations of most aquatic monitoring studies come in three categories: concentrations above the limit of quantification (LOQ), concentrations below the LOQ, but above the limit of detection (LOD), and concentrations below the LOD. Data <LOQ and <LOD can be interpreted as the so-called censored data: although the true concentration value is unknown, an upper boundary for the concentration is known (the LOQ and LOD, respectively). Various recommendations and traditions exist for how to handle, especially, data <LOD and also <LOQ. In practice, simplifications are often made when dealing with data below the detection limits.[12−14] For example, data <LOD are often removed, and half the LOQ value assumed for all data <LOQ, thus resulting in artificially fabricated concentration values.[15−17] Although statistical methods exist to handle detection limits in modeling procedures, their use is not widespread, and they do not provide the flexibility to incorporate additional spatiotemporal information. This is the benefit of the statistical programming language Stan, which provides specialized probability density functions that can be used for handling censored data.[18]

One approach to modeling PECs of monitoring campaigns is by using the measured concentrations in distributional regression models, that is, regression models resulting in a distribution of a response variable and not a point estimate. These regression models, especially as hierarchical generalized linear models (HGLMs, also referred to as mixed-effect models), have several desirable properties that help to address the spatiotemporal sources of variation. It is possible to define a distributional family, from which the measured concentrations are assumed to stem from. For example, environmental concentrations of chemicals are usually thought to have originated from processes that can be described by a lognormal distribution.[19] Additionally, the spatiotemporal variations of the measured environmental concentrations can be interpreted as conditional variances of these measured concentrations: for every value of a temporal and spatial variable (*i.e.*, dates and sites), a different variance on the overall concentration is assumed. This allows to distinguish between the "static" background concentrations in the water body and the influence of spatiotemporal variations, thus making predictions more robust. Information on the LOQ and LOD can be incorporated as *de facto* censoring of the measured concentrations, leading to a potentially more realistic uncertainty assessment in PEC modeling.[20]

Recently, Bayesian methods have gained interest in the field of ERA, not only from the scientific community but also from industrial and legislative stakeholders.[21−23] These methods are intuitively appealing, as all outcomes can be interpreted in the context of probability, for example, how likely is an environmental risk given a PEC and a PNEC? Bayesian methods often use sampling algorithms, for example, Markov chain Monte Carlo simulations, to draw equally likely samples from a posterior distribution; this also means that uncertainty can be directly quantified.[24] The posterior distribution is proportional to the product of the prior distribution, based on *a priori* assumptions or knowledge of the system, and the likelihood of the data. The benefits of such procedures are explicit, relatively common sense statistical assumptions, in contrast to the inexplicit and often-ignored assumptions of frequentist methods.[25] Because of the posterior sampling, Bayesian methods allow to derive proxies of values, such as average, standard error, or 95% intervals, directly from the sampled posterior values; they do not need to be specifically constructed. HGLMs can be expressed as Bayesian models and can incorporate conditional variances for spatiotemporal information, as well as information on the censoring of data, that is, LOQs and LODs. Additionally, modern algorithms allow to model spatiotemporal correlation patterns of different substances across sites and dates.[26]

In the present work, we explore the possibilities of Bayesian distributional regression models to model the PEC distributions of a selection of Norwegian monitoring campaigns. Based on three aquatic chemical monitoring campaigns covering 8−145 chemicals, we demonstrate an example workflow for PEC derivation. By introducing model complexity to incorporate as much available information as possible, the principal model output is kept simple, understandable, and interpretable. We further reflect on the benefits and challenges of the proposed methodology and encourage the discussion about scientific and regulatory suitability.

2. MATERIALS AND METHODS

2.1. Description of the Monitoring Campaigns.

Environmental exposure data were taken from three fjord-associated Norwegian monitoring campaigns, representing different pollution scenarios: the southwestern Sørfjord, the southern Kaldvellfjord, and the southeastern Oslofjord. The three monitoring campaigns represent the diverse complexity associated with the chemical monitoring campaigns (Table 1). The Sørfjord campaign contains routine monitoring data for

Table 1. Overview of the Number of Measured Chemicals, Sites, Dates, and Data Points for the Three Monitoring Campaigns[a]

campaign	chemicals	sites	dates	values	<LOQ (%)	<LOD (%)
Sørfjord	8	4	14	220	36	30
Kaldvellfjord	29	4[b]	4	486	17	27
Oslofjord	145	4	1	589	2	74

[a]Additionally, the frequency of concentrations <LOQ and <LOD is given. [b]The four sites are spread across two areas (inner and outer fjord). Additionally, the two sites in the inner fjord contain measurements at two depths.

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eight metals, the Kaldvellfjord campaign contains monitoring data for 29 metals, and the Oslofjord campaign contains data for 145 chemicals, divided between 11 metals and 134 organic compounds. For all the three campaigns, water samples were taken, and chemical concentrations were determined using established extraction and analytical methods, as detailed in their respective reports.[27−29] A complete list of all chemicals for each campaign is given in Supporting Information 1. A summary of the number of chemicals, sampling dates, sampling sites, and frequencies <LOQ and <LOD is given in Table 1. All chemical concentrations are expressed in mol L^{-1} (M) to provide accurate measures of the biologically active elements.

2.2. Predicted Environmental Concentration Distribution Models. Bayesian distributional regression models were fit to the measured chemical concentration data of the three campaigns, resulting in PEC distributions for all chemicals. The reported chemical concentrations were treated as response variables with lognormal distribution, as reasoned for in previous works.[19,30,31] We thus specified four regression models as *de facto* HGLMs.[32,33] In short, models 1 and 2 are based on a subset of data without LOD data and assuming half the LOQ for all LOQ data, and models 3 and 4 are based on the full data sets including LOD and LOQ data. Models 1 and 3 demonstrate the modeling procedure without spatiotemporal conditional variances, whereas models 2 and 4 include all possible spatiotemporal conditional variances. A visual overview of the modeling relations is given in Supporting Information Figure S1. All conditional variables were treated as categorical.

2.2.1. Chemical-Specific PEC (Models 1−4). All models assumed that the measured concentrations differ between chemicals. This was described as

$$y_i \sim \text{lognormal}(\mu_i, \sigma)$$
$$\mu_i \sim \beta_i \quad (1)$$

where $y_i \in \mathbb{R}^+$ is the measured concentration of every chemical i, $\mu_i \in \mathbb{R}$ is the average of a lognormal distribution for each chemical i, $\sigma \in \mathbb{R}^+$ is the shared standard deviation of the lognormal distribution for all chemicals, and $\beta_i \in \mathbb{R}$ is the concentration parameter for each chemical i, which is expressed as a normal distribution on a natural logarithmic scale. Note that an intercept (α) is excluded from this and all other models.

2.2.2. Spatiotemporal Conditional Variances of the PEC (Models 2 and 4). All campaigns contain additional information about the monitoring program that can be incorporated in the modeling procedure as conditional variances. Variables for conditional variances were date and site for the Sørfjord campaign; area, site, depth, and date for the Kaldvellfjord campaign; and site for the Oslofjord campaign. The assumption was that chemical concentrations differ between these variables, such that every chemical can have different concentrations according to these conditional variables. Equation 1 was therefore extended as follows

$$\mu_i \sim \beta_i + \beta_{i,j_1} + \ldots + \beta_{i,j_n} \quad (2a)$$

where $\beta_{i,j} \in \mathbb{R}$ is the conditional variance for each spatiotemporal conditional variable j on chemical i, expressed as normal distribution with average 0. For the Kaldvellfjord campaign, the different sites were divided between two areas. This was expressed by the following equation

$$\mu_i \sim \beta_i + \beta_{i,j_a|j_b} + \beta_{i,j_b} \quad (2b)$$

where $\beta_{i,j_a|j_b} \in \mathbb{R}$ is the conditional variance of each site j_a given a common area j_b on each chemical i, and $\beta_{i,j_b} \in \mathbb{R}$ is the conditional variance of each area j_b on each chemical i; both are expressed as normal distributions with an average of 0.

2.2.3. Spatiotemporal Chemical Correlations (Models 2 and 4). The use of conditional variances allowed to model the correlations of measured concentrations across these conditional variances.[26] The correlations were quantified as Cholesky correlation factors during the modeling procedure.[34] For every possible binary combination of chemicals of every spatiotemporal conditional variance (for example, cadmium−copper for date-specific conditional variance in model 4 of the Sørfjord campaign), this resulted in a distribution of Cholesky correlation factors.

2.2.4. LOQ and LOD as Censoring (Models 3 and 4). Environmental monitoring data routinely come in three different quality categories. Most chemical concentrations can be reliably measured because they are above the machine- and procedure-dependent LOQ. There is also the possibility that chemicals may not be measurable because they either are absent or below the LOD. Finally, some chemicals can be measured above the LOD but below the LOQ. These three categories can be treated as censored data.[35,36]

For models 1 and 2, data <LOD were excluded, and data flagged as <LOQ were assigned to be half the LOQ value.[15−17] These data were modeled using a standard log probability density function (LPDF). For models 3 and 4, no data were removed or altered, and censoring was applied. Concentrations >LOQ were considered uncensored; concentrations <LOQ but >LOD were considered interval-censored; and concentrations <LOD were considered left-censored. Uncensored data were modeled using an LPDF; left-censored data were modeled using a log cumulative distribution function (LCDF); and interval-censored data were modeled as the logarithm of the difference of the exponentiation of the LCDFs for the LOQ and LOD, respectively.[37]

In other words, for uncensored data, the LPDF tries to maximize the probability for observing a (new) value identical to the measured value; for left-censored data, the LCDF tries to maximize the probability for observing a (new) value smaller than the LOD; and for interval-censored data, the LCDF tries to maximize the probability for observing a (new) value in the interval between the LOQ and the LOD.

2.3. Prior Specifications. For all the three campaigns, prior distributions were expressed on a natural logarithmic scale. Per default, normally distributed priors were used. Informative prior values were calculated from the respective data sets, which considerably reduced the sampling time and improved the model convergence. This is sometimes referred to as empirical Bayes.[38] For the Cholesky correlation factors, a Lewandoski−Kurowicka−Joe distribution prior was used.[26] Detailed descriptions of the empirical Bayes prior determination are given in the Supporting Information.

2.4. Model Validation and Comparison. For all models, prior distributions were sampled along the posterior distributions and compared to both the measured concentrations and the posterior distributions in prior predictive checks to verify the suitability of the empirical prior

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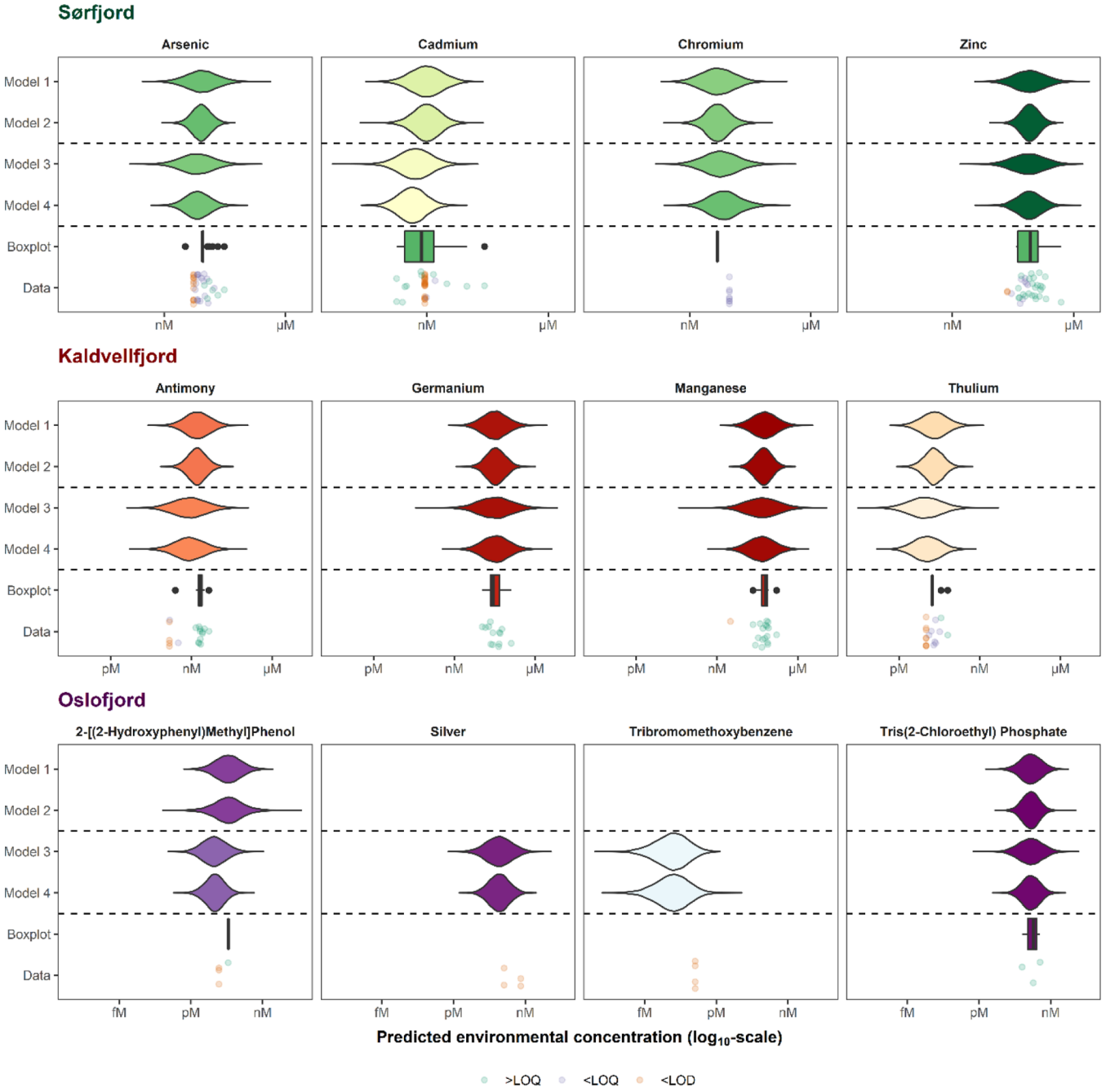

Figure 1. Overview of the PEC distributions of models 1−4 for four random chemicals from each monitoring campaign. The densities are based on 10,000 posterior samples for each model, without considering the conditional variances for models 2 and 4. The color gradients of the densities indicate the mode of the PEC, from lighter hues (lower concentrations) to darker hues (higher concentrations). The reported concentrations are given as jitter points, with the color indicating the measurement (un)certainty. The boxplots are based on the reported data excluding concentrations <LOD and assuming half the LOQ concentration for concentrations <LOQ; the boxplots are directly comparable to the data used for models 1 and 2. Note that the PEC distributions for two chemicals of the Oslofjord campaign were only possible for models 3 and 4 because all reported concentrations were <LOD.

distribution choices. Posterior predictions were compared to the measured concentrations in the posterior predictive checks.[39]

To compare the models, expected log-pointwise predicted densities (ELPDs) were calculated. ELPDs are a measure of predictive accuracy, where higher values indicate a higher accuracy and better model fit.[40] ELPD values were calculated for each model based on leave-one-out (LOO) cross-validation (CV).[41,42] In case of problematic observations (*e.g.*, Pareto shape parameter ≥0.5), other options for ELPD calculations were disregarded because of the steep increase in computational expenses, compared to the expected small change in ELPD values.[43]

Another more traditional approach to assess the goodness of fit of the different regression models is the use of R^2 values.[44] Bayesian R^2 values were calculated for each of the 10,000 posterior draws for every model of each campaign, following a

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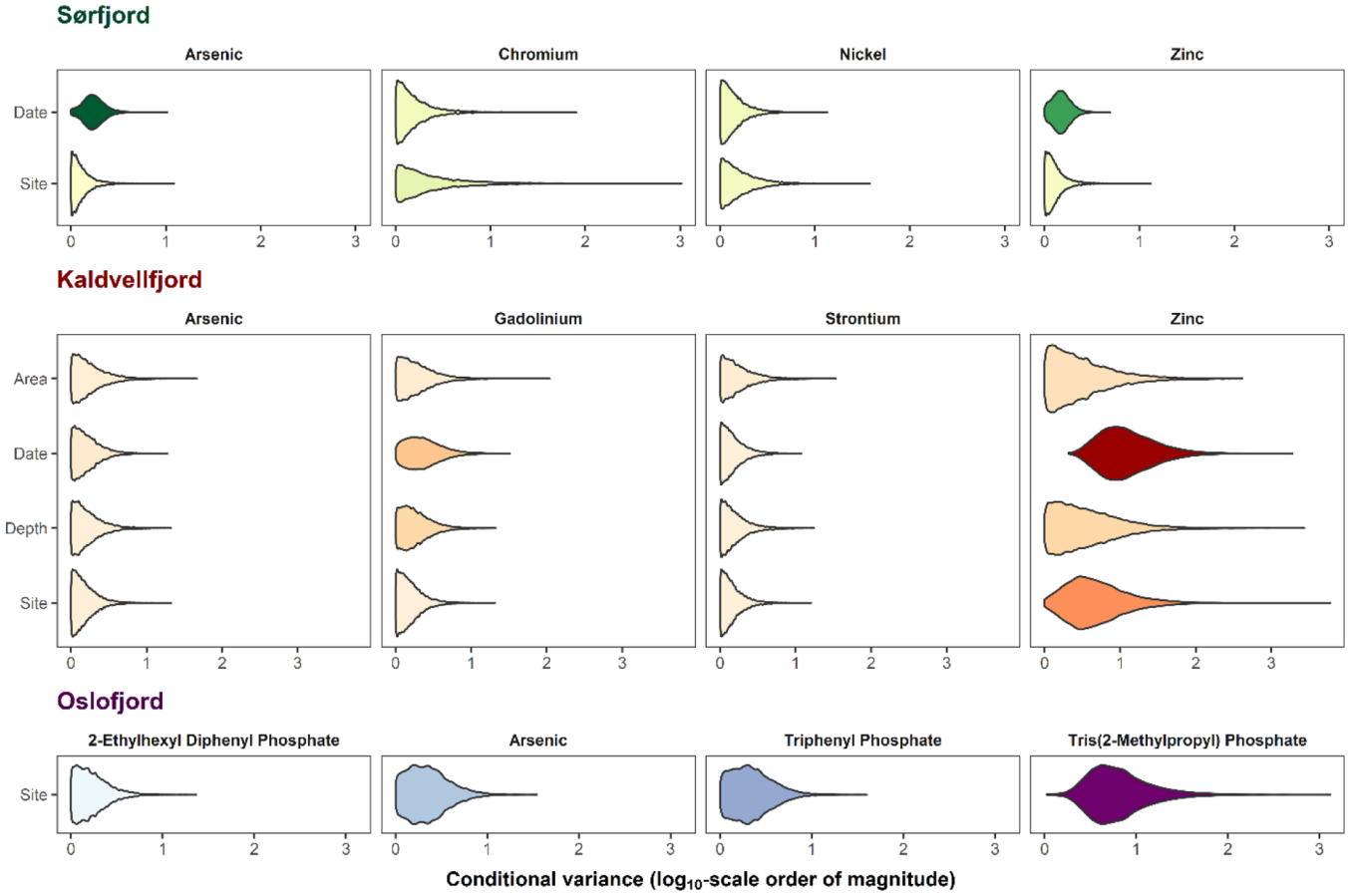

Figure 2. Overview of the spatial and temporal conditional variances (expressed in orders of magnitude of the logarithmic scale) of model 4 for four random chemicals of each campaign. The densities are based on 10,000 posterior samples for each model. The color gradients indicate the mode of the conditional variances, from lighter hues (lower values) to darker hues (higher values). A conditional variance value of 1 would indicate that the conditional variance has the same order of magnitude (on logarithmic scale) as the predicted concentration itself.

proposed Bayesian methodology.[45] This resulted in a distribution of Bayesian R^2 values for every model.

Because both ELPD and R^2 values are dependent on each model's input data, models 1 and 2 are not directly comparable to models 3 and 4.

2.5. Model Results. For all models, 10,000 posterior samples of the linear predictors without spatiotemporal conditional variances (*i.e.*, β_i and σ only) were used to describe the PEC. The posterior samples were characterized in terms of probability densities. As proxy for centrality, the highest maximum probability density estimate of the posterior, that is, the mode,[46] was used in favor of more common proxies, for example, the average or the median. The mode can be visualized as the "peak" of the density distribution of the posterior samples, that is, the concentration with the highest probability. Uncertainty was characterized in credible intervals using the highest density interval (HDI) of the posterior samples. Posterior samples inside an HDI have a higher probability than the posterior samples outside the interval. Because of the relatively high posterior sample size of 10,000, a 95% HDI was chosen, which can be interpreted as 95% credible interval.[47]

2.6. Comparison to Data Quantiles. As a comparison of the Bayesian distributional regression approach and a quantile representation of the measured concentrations, the posterior density distributions of models 1−4 (*i.e.*, PEC distributions from β_i and σ) were visually compared against the boxplot summaries of the data for models 1 and 2 (*i.e.*, without LOD and half the LOQ), as well as the raw data itself. This allowed to visualize the differences in the ways data can be described, especially when chemical concentrations <LOD are abundant.

2.7. Data Analysis. All data were analyzed using open-source statistical software R (version 4.0.3) and its add-on package brms (version 2.14.0).[48,49] The brms package is a high-level interface for the probabilistic statistical inference language Stan (version 2.25.0),[18] accessed through the CmdStanR (version 0.1.3) interface.[50] All Bayesian models were fit using Stan's integrated modified Hamiltonian Monte Carlo sampler with Markov chains. Five independent chains were run for every model with 4000 draws each, whereof half were used for warm-up, resulting in 10,000 posterior samples in total. To simplify Figures 1−3, only four random chemicals were visualized for each monitoring campaign.

2.8. Open Science. The data used in this publication and all scripts for the data analyses are publicly available on the corresponding author's GitHub repository (https://github.com/RaoulWolf/Bayesian-PEC-Modeling).

3. RESULTS AND DISCUSSION

3.1. Technical Aspects and Prior Choices. Bayesian distributional regression models were successfully fit to the data in each of the three chemical monitoring campaigns, with no warnings about divergent transitions. Bulk and tail effective sample sizes of all models were satisfactory, that is, larger than

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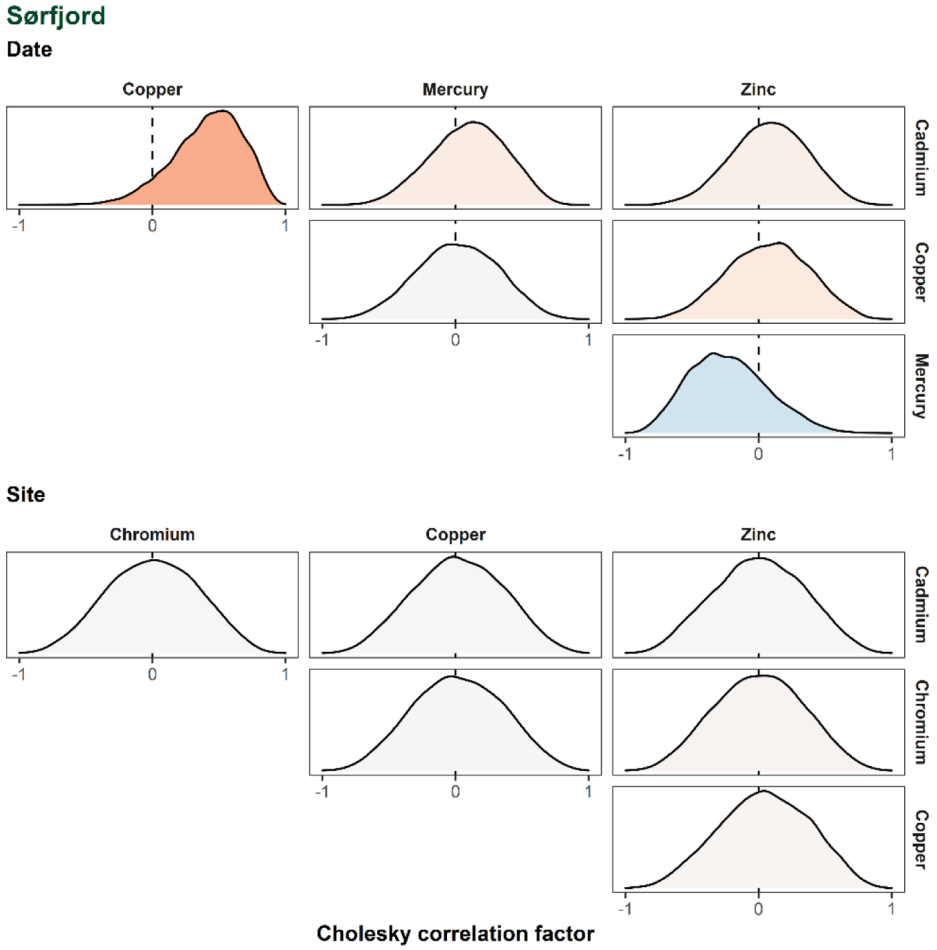

Figure 3. Example of the temporal and spatial correlation patterns for four random chemicals of the Sørfjord monitoring campaign. The densities are based on 10,000 posterior draws of the Cholesky correlation factors. The color gradients indicate the mode of the correlation distribution, from blue (−1; complete negative correlation) through gray (0; no correlation) to red (1; complete positive correlation).

100 times the number of chains,[51] and the MCMCs converged well, with all $\hat{R}$ values <1.01 (see Supporting Information 3).[52] Detailed model summaries are given in Supporting Information 4, and trace plots of the individual Markov chains for up to 12 random model parameters are given in Supporting Information 5.

Prior predictive distributions, that is, predictive distributions of all parameters sampling only from the prior and not including the likelihood of the data, showed excellent overlap with the posterior model parameter distributions for all models (see Supporting Information 5). Additionally, the posterior distributions of both models covered >90% of the measured concentration data range (Supporting Information Table S1).

We conclude that the use of informative priors was appropriate for our modeling approach, but uninformative or flat priors may be used in situations where the empirical Bayes method is either impossible or undesirable.

3.2. PEC Distributions. A realistic derivation of PECs is of crucial importance in ERA. Bad estimations of the PEC can lead to wrong downstream decision-making.[11] This is especially true if the PEC is derived as a single number, for example, as MEC, without accounting for uncertainty. By expressing PECs as samples from a lognormal distribution, we acknowledge and incorporate the inherent uncertainty to make the derivation of PECs more realistic.

All four models resulted in lognormal PEC distributions (Figure 1). Most of the PEC distributions were symmetrical on the logarithmic scale (models 1 and 3). The influence of spatiotemporal conditional variances was largely dependent on the chemicals, with some chemicals showing relatively little variation and others showing clear differences (Figure 1, Supporting Information Figure S2). PEC distributions including conditional variances were also symmetrical on the logarithmic scale (models 2 and 4). The censored PEC distributions (models 3 and 4) of chemicals occurring <LOD showed a left skew (see, for example, PEC distributions for the Oslofjord campaign in Figure 1 and Supporting Information Figure S2).

3.3. Spatiotemporal Conditional Variances. Most spatiotemporal conditional variances had values in orders of magnitude below the measured concentrations (that is, values <1; Figure 2). This can be interpreted as the weak influence of the spatiotemporal variables on the PEC. However, for some chemicals, the conditional variances have higher values, for example, for the chemicals in the Oslofjord campaign. In such situations, the interpretation would be that the variable (in this case, only spatial) has a noticeable influence on the PEC. This enables to critically evaluate the monitoring campaign itself, for example, in cases where the conditional variances display higher orders of magnitude than the measured concentrations; then, the prediction of the PEC is more correlated with the

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Table 2. Exemplary Overview of the Derived PEC Estimates (in mol L^{-1}) for Four Random Chemicals of Each Campaign[a]

	Sørfjord											
	Cadmium			Lead			Mercury			Nickel		
	lower	*central*	*upper*	*lower*	*central*	*upper*	*lower*	*central*	*upper*	*lower*	*central*	*upper*
Data	1.78×10^{-10}	7.12×10^{-10}	2.67×10^{-8}	5.90×10^{-10}	1.20×10^{-9}	1.17×10^{-8}	2.49×10^{-12}	2.99×10^{-11}	4.69×10^{-10}	2.69×10^{-9}	3.46×10^{-9}	4.55×10^{-9}
Model 1	1.64×10^{-10}	8.37×10^{-10}	5.75×10^{-9}	4.18×10^{-10}	2.65×10^{-9}	1.42×10^{-8}	5.01×10^{-12}	2.55×10^{-11}	1.73×10^{-10}	5.94×10^{-10}	4.30×10^{-9}	2.29×10^{-8}
Model 2	2.14×10^{-10}	1.09×10^{-9}	4.65×10^{-9}	5.82×10^{-10}	2.15×10^{-9}	6.44×10^{-9}	6.61×10^{-12}	3.26×10^{-11}	1.80×10^{-10}	1.02×10^{-9}	3.80×10^{-9}	1.33×10^{-8}
Model 3	8.09×10^{-11}	4.92×10^{-10}	3.20×10^{-9}	3.29×10^{-10}	1.99×10^{-9}	1.31×10^{-8}	3.43×10^{-12}	2.04×10^{-11}	1.38×10^{-10}	5.59×10^{-10}	3.82×10^{-9}	2.57×10^{-8}
Model 4	9.17×10^{-11}	4.37×10^{-10}	2.06×10^{-9}	4.94×10^{-10}	2.29×10^{-9}	9.63×10^{-9}	3.17×10^{-12}	1.93×10^{-11}	1.16×10^{-10}	7.88×10^{-10}	4.24×10^{-9}	1.85×10^{-8}

	Kaldvellfjord											
	Cobalt			Lutetium			Vanadium			Zinc		
	lower	*central*	*upper*	*lower*	*central*	*upper*	*lower*	*central*	*upper*	*lower*	*central*	*upper*
Data	2.11×10^{-9}	2.74×10^{-9}	1.25×10^{-4}	1.66×10^{-11}	1.05×10^{-11}	2.08×10^{-11}	1.01×10^{-8}	1.97×10^{-8}	7.20×10^{-8}	4.12×10^{-9}	1.77×10^{-7}	1.20×10^{-6}
Model 1	5.45×10^{-9}	5.13×10^{-9}	5.44×10^{-7}	2.34×10^{-12}	1.95×10^{-11}	2.04×10^{-10}	2.39×10^{-9}	1.87×10^{-8}	1.75×10^{-7}	1.16×10^{-8}	9.68×10^{-8}	7.88×10^{-7}
Model 2	3.93×10^{-10}	7.42×10^{-8}	1.40×10^{-5}	2.31×10^{-13}	1.64×10^{-11}	1.27×10^{-9}	4.50×10^{-9}	2.11×10^{-8}	8.76×10^{-8}	1.48×10^{-8}	9.87×10^{-8}	7.06×10^{-7}
Model 3	1.22×10^{-10}	2.13×10^{-9}	3.51×10^{-8}	3.60×10^{-13}	6.06×10^{-12}	1.07×10^{-10}	1.23×10^{-9}	1.90×10^{-8}	3.38×10^{-7}	2.28×10^{-9}	4.74×10^{-8}	6.33×10^{-7}
Model 4	3.11×10^{-10}	3.42×10^{-9}	6.87×10^{-8}	8.29×10^{-13}	6.96×10^{-12}	8.09×10^{-11}	2.29×10^{-9}	1.71×10^{-8}	1.83×10^{-7}	2.78×10^{-9}	3.47×10^{-8}	5.08×10^{-7}

	Oslofjord											
	1,2,3,4,5,6-Hexachlorobenzene			Arsenic			Triethyl Phosphate			Tris(2-Methylpropyl) Phosphate		
	lower	*central*	*upper*	*lower*	*central*	*upper*	*lower*	*central*	*upper*	*lower*	*central*	*upper*
Data	—	—	—	2.01×10^{-8}	3.97×10^{-8}	8.48×10^{-8}	1.12×10^{-9}	1.23×10^{-9}	3.15×10^{-9}	5.56×10^{-11}	1.90×10^{-10}	2.77×10^{-9}
Model 1	—	—	—	5.47×10^{-9}	3.77×10^{-8}	3.01×10^{-7}	2.27×10^{-10}	1.74×10^{-9}	1.26×10^{-8}	3.43×10^{-11}	2.51×10^{-10}	1.70×10^{-9}
Model 2	—	—	—	8.27×10^{-9}	3.86×10^{-8}	1.88×10^{-7}	3.70×10^{-10}	1.76×10^{-9}	8.39×10^{-9}	4.28×10^{-11}	2.53×10^{-10}	1.60×10^{-9}
Model 3	2.66×10^{-14}	6.44×10^{-13}	1.13×10^{-11}	2.94×10^{-9}	2.35×10^{-8}	2.61×10^{-7}	1.79×10^{-10}	1.76×10^{-9}	1.77×10^{-8}	2.38×10^{-11}	2.25×10^{-10}	2.31×10^{-9}
Model 4	2.57×10^{-14}	8.79×10^{-13}	1.53×10^{-11}	5.38×10^{-9}	3.28×10^{-8}	1.67×10^{-7}	3.27×10^{-10}	1.82×10^{-9}	9.50×10^{-9}	3.53×10^{-11}	3.12×10^{-10}	1.94×10^{-9}

[a]For every chemical, a lower and an upper limit, as well as a centrality proxy, are given. For the data itself, these are 2.5, 50, and 97.5% quantiles, that is, the median and 95% confidence interval. For models 1−4, these are the lower and upper bounds of the 95% highest density interval (HDI), that is, the 95% credible interval, and the mode, that is, the "peak" of the density distribution. For the data quantiles, the reported concentrations <LOD were removed, and the reported concentrations <LOQ were assumed to be half the LOQ; they are equivalent to the boxplots in Figure 1. Note that for 1,2,3,4,5,6-hexachlorobenzene of the Oslofjord monitoring campaign, only reported concentrations <LOD were available; thus, quantiles for the data and models 1 and 2 were not possible. For each descriptor of every chemical in models 1−4, the lowest value is highlighted in blue and the highest value is highlighted in red.

information on dates and sites than on the identity of the chemical itself. Also, the quantification of conditional variances makes it possible to distinguish whether the concentration of a chemical is mainly influenced across dates or sites. For example, the date-specific conditional variance for zinc concentrations in the Kaldvellfjord campaign is considerably higher than the corresponding area-specific conditional variance (Figure 2). This can be interpreted such that zinc concentrations vary more across time than across locations.

Considering conditional variances enables more specialized assessments and potentially more detailed further analyses; for example, spatiotemporal conditional variances allow to inspect special exposure scenarios directly. For example, it is possible to determine how each chemical concentration varies across each site and date. This way, it is possible to quantify, for example, which sites have a higher pollution load. This information could become useful for prioritization of actions, both in terms of chemicals and locations.

3.4. Spatiotemporal Correlations. The use of Cholesky correlation factor distributions can provide further insights into the spatiotemporal correlations of the PECs. An example with four random chemicals from model 4 of the Sørfjord campaign can be seen in Figure 3. Although most chemical−chemical pairs show weak temporal correlations, that is, the modes of the Cholesky correlation distributions are close to zero, there appears to be a positive temporal correlation between cadmium and copper and a negative temporal correlation between mercury and zinc. This could indicate that cadmium

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Table 3. ELPDs (Standard Error in Parentheses) for all Models of Each Campaign, Based on LOO-CV[a]

	Campaign					
	Sørfjord		Kaldvellfjord		Oslofjord	
ELPD	*abs*	*diff*	*abs*	*diff*	*abs*	*diff*
model 1	2763 ± 28	−59.6 ± 13.6	6136 ± 85	−141.2 ± 42.2	3216 ± 57	−12.2 ± 8.0
model 2	**2823±31**	**0.0±0.0**	**6278±71**	**0.0±0.0**	**3228±56**	**0.0±0.0**
model 3	1165 ± 130	−25.1 ± 12.3	4240 ± 209	−66.4 ± 33.5	2751 ± 223	−30.3 ± 16.9
model 4	**1191±130**	**0.0±0.0**	**4306±211**	**0.0±0.0**	**2781±224**	**0.0±0.0**

[a]The left column contains the absolute ELPD values (*abs*), the right column gives the difference in relation to the most accurate model (*diff*). Note that models 1 and 2 were not comparable to models 3 and 4 because of the different data basis. See Materials and Methods for details. The models with the highest ELPD, that is, the highest accuracy, are denoted in bold.

and copper concentrations are influenced by the same temporal process, with no distinct spatial patterns. Information on the spatial or temporal correlation of chemical concentrations can be a powerful addition to the PEC distributions and could eventually help to prioritize regulatory decisions, for example, mitigation measures.

3.5. Measurement Uncertainties. Models including LOQ and LOD information generally resulted in PEC distributions with lower concentrations (Figure 1 and Supporting Information Figure S2). This is not surprising, as these models explicitly take the LOQ and LOD into account as censoring. This likely gives a more credible description of the uncertainties associated with environmental concentrations, overcoming technology-associated hurdles, that is, quantification and detection limits. The practice of fabricating artificial concentrations, for example, based on half the LOQ, is in contrast a relatively arbitrary approach to addressing limits of quantification and detection. Still, censoring of data introduces ambiguity, and therefore the "gold standard" should be to have all measured concentrations >LOQ. However, such an ideal scenario would likely come with substantial analytical efforts; so, the implementation of the LOQ and LOD values into PEC derivations could be a pragmatic alternative solution. The methodology shown in this article can be easily adjusted to include or exclude censoring of data and is flexible to cater for scientific and regulatory demands and needs.

It is important to remember that in case of censoring, the predictions are directly driven by the LOQ and LOD values. The reason for this lies in the empirical Bayes estimation of the prior distributions. As the only reliable information available for censored data are the LOQ and LOD values, they are used for the estimation of the prior distribution. This resembles a "worst-case" assumption, that is, the actual concentrations could be the LOQ or LOD, respectively.

If chemical concentrations are repeatedly <LOD, this can be interpreted in two ways: either the chemical is absent from the monitoring system or the chemical is only present in low concentrations <LOD. The latter could be assumed for ubiquitous legacy pollutants, such as POPs or PAHs, and also metals.[53,54] If technical limits are considerably low and chemicals still occur <LOD, there will still be an increased uncertainty, but in subsequent ERA procedures, PEC distributions with higher uncertainty, but still very low concentration ranges, are not likely to drive an overall risk assessment.

Even though the PEC distributions cover a wider concentration range than the 95% quantile summaries, they provide a more realistic and robust estimation of the actual concentration of the chemicals in the environment, irrespective of the site- and date-related variances. This is especially visible when considering the PEC distributions of chemicals occurring strictly <LOD, as shown in the Oslofjord campaign in Supporting Information Figure S2.

3.6. Bayesian PEC Derivation. The approach of modeling PECs as Bayesian distributional regression models allowed us to separate site- and date-specific variances from the latent background concentrations and incorporate the information of the LOQ and LOD. The 10,000 posterior draws of each model are for the β_i and σ parameters and do not include the conditional variances of sites or dates (Figure 1). This makes the PEC derivation robust to spatial and temporal variations, which is explicitly encouraged within the WFD and by EFSA.[3,11,55,56] Still, it is possible to include this information in other investigations, for example, to model potential worst-case scenarios, to identify pollution hotspots, or to analyze the seasonal cycles of pollution (see Supporting Information Figure S2). The success of such inferences depends largely on the available data for model fitting: the more data available, the higher the accuracies of the model predictions will be.

A comparison of the data's 95% confidence intervals and the models' 95% credible intervals is given in Table 2 for four random chemicals of each monitoring campaign. The PEC distributions of models 1−4 showed a good overlap with the underlying data, with the 95% CI of all models covering >90% of the measured concentrations (Supporting Information Table S1). More visually, the censored PEC distributions of model 4 cover the measured concentrations, while showing a left skew relative to the boxplot summaries, with the modes of the censored PEC distributions being in equal range or lower than the median of the raw data (Figure 1). The PEC distributions also include concentration ranges not covered in the raw data, and the PEC distributions of models 3 and 4 were in the same range or lower than the PEC distributions of models 1 and 2 (Figure 1 and Supporting Information Figure S2). Differences in terms of 95% credible intervals were largely minor, and estimations were usually on the same order of magnitude, except when the percentage of data <LOD was high, for example, for 1,2,3,4,5,6-hexachlorobenzene in the Oslofjord campaign.

3.7. Model Comparisons. Based on the ELPD results (Table 3), the inclusion of spatiotemporal conditional variances generally increased model accuracies. It is therefore beneficial to include spatiotemporal information in PEC modeling to increase robustness to seasonal and local sources of variability. However, higher accuracies do not necessarily mean that the ranges of the PEC distributions become narrower, but they more accurately reflect natural conditions without spatiotemporal variations. As the censoring of models 3 and 4 means they are not based on the same data basis (see

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Materials and Methods), the ELPD estimates of models 3 and 4 cannot be directly compared to models 1 and 2.

The Bayesian R^2 values also increased with the increasing model complexity, that is, models with conditional variances had higher R^2 values (Supporting Information Figure S3). Like the ELPD, the Bayesian R^2 is evaluated in the context of the response variable, and thus the R^2 distributions of models 3 and 4 cannot be directly compared to those of models 1 and 2.

3.8. Reflections. The Bayesian methodology presented in this article has resulted in PEC distributions and credible uncertainty estimations. The methodology is highly flexible and can be adjusted to individual requirements of monitoring studies, for example, regarding spatiotemporal structures. This approach also provides a direct way to quantify uncertainty and to express the PEC as a distribution. This distributional PEC can be utilized in downstream probabilistic analyses, such es probabilistic ERA.

In our proposed methodology, all model predictions cover more than 90% of the original measured concentrations. Such high posterior coverages have been raised as critically important quality assurance parameters by EFSA in other modeling contexts, but are in principle also applicable here.[22] Also, the probability-based uncertainty of the estimated PECs is explicitly desirable in regulatory modeling procedures.[57]

As shown above, the inclusion of spatiotemporal conditional variances generally improved model accuracies. Even in cases where the inclusion of conditional variances made only a minor difference in the predicted PEC distributions, these differences could potentiate when being used further in ERA procedures. We therefore recommend including spatiotemporal conditional variances.

Furthermore, we encourage testing the use of data <LOQ and <LOD in censored models. Although deterministic summary approaches for the PEC can provide similar results when chemicals are reliably measured (that is, >LOQ), the LOQ and LOD can influence the PEC estimations when chemicals mainly occur <LOQ or <LOD. We have provided models based on "classical" data (>LOQ and <LOQ; models 1 and 2) and all available data (>LOQ, <LOQ, and <LOD; models 3 and 4) but acknowledge the need for extensive trials and frameworks before the widespread consideration of censoring in PEC modeling, especially for regulatory purposes.[58] The presented modeling approach is flexible enough to work both with and without data censoring.

Our proposed methodology comes with an increased computational cost and a potential statistical hurdle; although the developments in environmental modeling in recent years certainly point toward more acceptance of Bayesian methods, not the least in regulatory contexts.

Even though we applied our methodology to chemical concentration data from aqueous monitoring studies, the methodology is equally applicable for chemical concentration data from other environmental matrices, for example, sediment or air. In the context of ERA, this methodology is fully compatible with other Bayesian ERA approaches, if the PNEC can also be expressed as distribution of posterior draws.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c06268.

Detailed description of monitoring campaigns, detailed descriptions of empirical prior distribution calculations, posterior predictive checks based on 95% HDI, schematic modeling workflow, comparison of PEC predictions with and without conditional variances, and Bayesian R^2 comparisons (PDF)

Raw data (ZIP)

Scripts for data analyses (ZIP)

List of all chemicals; overview of the $\hat{R}$ values; summaries of model parameters; posterior trace plots; and prior−posterior comparisons (ZIP)

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Notes

The authors declare no competing financial interest.

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