Original Article **Integrated Environmental Assessment and Management**
DOI 10.1002/ieam.1617

Simulating the uncertain effect of active carbon capping of a dioxin polluted Norwegian fjord[††]

Running head: Uncertain Remediation

Jostein Starrfelt *†‡ & Tuomo Saloranta §

† Norwegian Institute for Water Research, Gaustadalléen 21, NO-0349 OSLO, Norway.
‡ Centre for Ecological and Evolutionary Synthesis, Department of Biosciences, University of Oslo, P.O. Box 1066 Blindern, NO-0316 Oslo, Norway. Phone: +47 22 85 44 00. Fax: +47 22 85 44 01. Email: jostein.starrfelt@ibv.uio.no
§ Norwegian Water Resources and Energy Directorate, Middelthunsgate 29, Postboks 5091 Majorstua, NO-0301 OSLO. Email: tus@nve.no

* To whom correspondence may be addressed.

[††]This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the Version of Record. Please cite this article as doi: [10.1002/ieam.1617]

All Supplemental Data may be found in the online version of this article at the publisher's website.

Submitted 18 September 2014; Returned for Revision 12 December 2014; Accepted 30 December 2014

Abstract

Process-based multimedia models are frequently utilized to simulate the long-term impacts of pollutants and to evaluate potential remediation actions that can be put in place to improve or manage polluted marine environments. Many such models are detailed enough to encapsulate the different scales and processes relevant for various contaminants, yet still are tractable enough for analysis through established methods for uncertainty assessment. Inclusion and quantification of the uncertainty associated with local efficacy of remediation actions is of importance when the desired outcome in terms of human health concerns or environmental classification shows a non-linear relationship with remediation effort. Here we present an updated fugacity based environmental fate model set up to simulate the historical fate of polychlorinated dibenzo-*p*-dioxins and dibenzo-furans (PCDD/Fs) in the Grenland fjords, Norway. The model is parameterized using Bayesian inference and is then used to simulate the effect of capping parts of the polluted sediments with active carbon. Great care is taken in quantifying the uncertainty regarding the efficacy of the activated carbon cap to reduce the leaching of contaminants from the sediments. The model predicts that by capping selected parts of the fjord biota will be classified as *Moderately polluted* approximately a decade earlier than a natural remediation scenario. Our approach also illustrates the importance of incorporating uncertainty in local remediation efforts, as the biotic concentrations scale non-linearly with remediation effort.

Keywords: PCDD/F, remediation, fugacity model, MCMC, uncertainty, nonlinearity Jensen's inequality.

INTRODUCTION

Industrial discharge and pollution is a pervasive problem for numerous Norwegian fjords (Breedveld and others 2010). Though emissions have been greatly reduced with increased knowledge and improved technology for cleaning, there are still several locations where contaminants now are leaching from fjord sediments leading to elevated levels of persistent organic pollutants (POPs) in biota. In some cases there are guidelines in place to restrict the human consumption of the polluted biota, as is the case for the Grenland fjords (Breedveld and others 2010). The Grenland fjords exhibit some of Norways highest concentrations of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in sediments and efforts have been devoted to both to monitor the area (e.g. Bakke and others 2012; Green and others 2013) and to investigate the possibilities and efficacies of suggested remediation strategies (e.g. Saloranta and others 2007). Health advisories have been in place in the area for decades, initially due to mercury pollution, but now due to the concentrations of PCDD/Fs in biota (Økland 2005). Historically, the main source of PCDD/Fs in the Grenland fjords has been a magnesium plant at Herøya in Frierfjorden (see Figure 2), in operation from 1951 to 2002. Though the emissions were greatly reduced in the mid 70's and more so around 1990 and completely halted in 2002 (see SI), the fjord sediments now constitute a considerable secondary source of PCDD/Fs.

In addition to monitoring the area, several model applications have been developed to investigate the distribution and dynamics of PCDD/Fs in the fjords both with focus on abiotic fate (Persson and others 2006; Saloranta and others 2006b; Saloranta and others 2007) and accumulation in the food web (Saloranta and others 2006a), These models have also, in line with the scientific knowledge and technology, developed from fairly simple deterministic applications (Persson and others 2006) to dynamic fugacity based models using advanced uncertainty assessment of parameterization to better capture the uncertainty associated with predicting the future of the fjords and the potential effects of remediation strategies (Saloranta and others 2006a; Saloranta and others 2007). Uncertainty in modelling exercises can be delineated into several categories of uncertainty, arising from different aspects of the link between the real world and the model. There are several proposed frameworks for differentiating between different aspects of uncertainty in environmental modelling (e.g. Klauer and Brown 2004; Refsgaard and Henriksen 2004; Refsgaard and others 2007; Walker and others 2003). Here we adopt distinctions similar to Walker and others (2003) concerning three different types of uncertainty, namely model, parametric and input data uncertainty. Structural or model uncertainty denotes the potential problem of differences in structure and process, i.e. to which degree a mathematical model is able to be an accurate representation of the relevant processes and scales. A model must by definition simplify the system it is intended to model, and this entails the identification of relevant processes and patterns in the system. Inclusion of the relevant processes also imply the *exclusion* of processes deemed unimportant, and researchers can never be certain as to which should be included or not. Structural uncertainty is hard to address, since one of the few ways to properly quantify it needs comparisons of several models of various complexity and with different structures.

Parametric uncertainty (Walker and others 2003) relates to the choice of the parameterization of the relevant processes and patterns; examples include parameters for resuspension and deposition velocity of particles, through parameters of advection to physico-chemical properties of the contaminants being modelled. Uncertainty in parameterizations can be taken into account and quantified, for instance, in a Bayesian framework (e.g. Gelman and others 2004) where selected important parameters are estimated using the model combined with observed data, and parametric distributions are used instead of single parameter values. In such an approach, models are used to generate a range of quantifiable outcomes inherently including the uncertainty in the parameters.

In process-based models of contaminant fate and transport, the goal is often to perform scenario simulations, e.g. to model the implementation of a proposed remediation scenario (Saloranta and others 2007) to predict the global effects or to model the potential impact of changing drivers of the system, for instance climate change (e.g. Borgå and others 2010). The main rationale behind such exercises is that with a sufficiently accurate representation of the processes in the system, one can use the model to simulate alternative futures. Though earlier applications of fugacity based models of contaminant fate in the Grenland fjords have taken great care to include the parametric uncertainty of the model, remediation scenarios have been simplistic and the effect of uncertain effects of the remediation *per se* has not been included (i.e. remediation scenarios such as capping has assumed 100 % removal of contaminants in the sediments of selected boxes (Saloranta and others 2007)). This is essentially an example of not taking input uncertainty into account (Walker and others 2003).

Suggested remediation measures for contaminated fjords include removal of polluted sediments (see e.g. Kulkarni and others 2008) and various techniques for capping the sediments, thereby trapping the contaminants and hindering further leaching from sediment to water and biota. In the Grenland fjords, field scale experiments have been performed to estimate the potential for various capping materials and techniques to limit the transport of contaminants from the sediments to the water (Josefsson and others 2012; Schaanning and Allan 2011). Process based models is the appropriate tool for evaluating the effect of such remediation measures on contaminant concentrations in a larger area and in biota, and for capturing the uncertainty in local effect (i.e. reduction of contaminant flux at parts of the fjord) and how this influences the global endpoint (e.g. concentrations in biota at a larger spatial scale). Capturing the uncertainty in the local effect is pertinent for the correct quantification of global endpoint effects due to the potential for non-linearities in the mapping from local effect (or effort) to global endpoint.

Figure 1 shows possible relationships between local effect (e.g. removal of polluted sediments from a part of a larger system) and resulting effect for a global endpoint (in the whole or a different part of the system). When there is a simple linear relationship (as in Figure 1A), the potential uncertainty (or variability) around a mean local effect (x-axis) will not affect the mean global effect (y-axis). In other words, an expected global effect can be predicted using the mean of a local effect. However, a non-linear relationship between local remediation effect and global endpoints (Figure 1B) complicates the issue. Due to Jensen's inequality ($F(E(e)) < E(F(e))$, F being the function from local (e) to global effect and E denoting the average) non-linear relationships will give rise to incorrect predictions of global effects when only using a point estimate of local effect. As an example, say that a remediation strategy including capping will reduce the flux of contaminants to, on average, 70 %, and with a confidence interval of +/- 20 % (blue lines in Fig 1B). If the relationship between local effect and global endpoint is concave (as in Fig 1B), using the point estimate of local remediation effect at 70 % flux reduction will give a different value for the global endpoint, compared to when the whole distribution of local effects is taken into account (compare dotted horizontal line on the y-axis in Figure 1B, the global effect given a 70 % reduction locally, and the full line denoting the mean global effect given the full distribution of local effect). Since remediation actions usually are targeted at a specific process (e.g. flux from sediment store to water column) on part of the whole system, but global endpoints (e.g. biota concentrations) are what we are really interested in changing, effort needs to be put into quantifying both the uncertainty in local effect *and* potential non-linearity in the relationship between local and global effect.

In this article we present a revised Grenland fjord model application, where the fugacity model (Mackay 2001) is improved in three ways. Firstly the spatial division of the fjords (i.e. the *structure*) is further increased in resolution, to improve the similarity of the model with the real system and to better represent areas which have been identified for possible large-scale capping. Secondly, to calibrate the model and estimate model parameters we now include newer data from biota, sediment and water samples taken from the recent monitoring efforts (e.g. Bakke and others 2012) and projects in the Grenland fjords (Allan and others 2012; Allan and others 2011; Bradshaw and others 2012; Cornelissen and others 2012; Cornelissen and others 2010). Thirdly, we include the uncertainty surrounding the effect of a relevant and proposed remediation action, namely capping parts of the polluted fjords with cleaner sediments infused with active carbon (Josefsson and others 2012; Schaanning and Allan 2011). The local efficacy of such remediation measures have been experimentally tested in pilot studies in the area (Cornelissen and others 2012; Josefsson and others 2012; Schaanning and Allan 2011). By including the uncertainty in local effects of remediation, we can explicitly test for potential effects of Jensen's inequality in scenario simulations. The effect of local remediation action is modelled in a probabilistic manner, incorporating the uncertainty in such actions.

THEORY, METHODS AND MATERIAL

Data

Observations of concentrations of PCDD/Fs were compiled with a total of 97 samples from sediments, 25 samples from particulate matter in the water column, 97 individual or pooled cod samples and 38 pooled crab samples. To represent the background concentrations 4 pooled samples of cod liver from Skagerrak were included. The data comes from various projects and monitoring efforts (Allan and others 2012; Allan and others 2011; Bakke and others 2012; Bradshaw and others 2012; Eek and others 2011; Naes and others 2009; Saloranta and others 2007 for more details see Supporting Information). The concentrations were assumed to be log-normally distributed, reflected in the error model (likelihood function) used in the parameter estimation phase. Yearly averages (within each box or species) were used as to calibrate the model. The variances of the log-normally distributed concentrations were also estimated as part of the parameterization, similar to (Saloranta and others 2007). The model was set up to simulate three of the PCDD/F congeners (23478-PeCDF, 123478/123479-HxCDF and 123678-HxCDF), and the estimated bioaccumulation factors (for cod and crab independently) were assumed to capture the scaling from exposure to these three congeners to toxic equivalency (TEQ) for all PCDD/Fs congeners.

Fugacity Model: Grenland Fjord Application

The dynamic fugacity model (SF-tool) set up has been described in more detail previously (Saloranta and others 2006b; Saloranta and others 2007). The SF-tool consists of a water-sediment fugacity model code able to simulate sources, sinks and transports of contaminants and a bioaccumulation rate constant model code to simulate the uptake and accumulation of POPs in biota. In the application here we applied a simplified bioaccumulation model in which bioaccumulation factors (BAFs) are estimated for cod and crab respectively (see below). The fjord system was divided into 7 areas with 2-3 compartments representing different depths. These water compartments are all associated with a sediment compartment. Compared to earlier applications, the spatial resolution was increased for the outer fjords, due to the considerations to select parts of the outer fjords as possible remediation sites (areas 3, 4 and 5 in Fig 2 were previously modelled as one area). A total of 25 parameters, including error variances for biotic and abiotic observations were estimated in a Markov Chain Monte Carlo approach, using Bayesian inference. This autocalibration scheme has been detailed elsewhere (Saloranta and others 2007), and a full list of all parameters used is given in the Supplementary Information.

Calculation and Classification of Contaminated Biota.

Concentrations of PCDD/Fs in biota (C_b) was modelled by estimating bioaccumulation factors (K_{BAF}) and parameters scaling the fraction of time cod and crabs are exposed to concentrations in the pore water (β) and in the water column $\left((1-\beta)\right)$:

$$C_b = K_{BAF}\left(\beta C_{pw} + (1-\beta)C_w\right)$$

The concentrations in the water compartments (C_w) and porewater (C_{pw}) in the sediment compartments used to predict the biotic concentrations were calculated as a volume weighted average of the two habitats defined; inner fjords (composed of area 1 and 2, see Fig 2) and outer fjords (composed of area 3 and 4, Fig 2). A sill at Breivik (where area 2 and 3 meet) gives credence to the assumption of two distinct populations for each species. Four parameters scaling the maximum habitat depth for cod and crab, in inner and outer areas were also estimated, and were used to calculate the volume weighted average of the exposure through water and sediment (see SI). Finally a response time (t_r) was estimated, which was used to "delay" the concentrations in biota, i.e. concentrations in biota at time t was calculated using the relevant abiotic concentrations at time t - t_r.

Norwegian classification of polluted biota in coastal waters and fjords consists of 5 classes from insignificantly polluted (Class I) to very highly polluted (Class V) with limits for PCDD/Fs in cod liver at 15 / 50 / 100 / 300 ng TEQ / kg w.w. and 10 / 40 / 100 / 250 ng TEQ / kg w.w. for crab hepatopancreas (Molvær and others 1997).

Uncertainty of Remediation Effects

After parameterization of the model, the posterior distributions were used to simulate the effect of capping the sediment compartments in Eidangerfjord (Fig 2, area 3) using active carbon as an additive. This remediation scenario is deemed to be most realistic both in terms of feasibility and monetary valuations (Olsen 2012). To be able to include the uncertainty associated with the local effect, i.e. the reduction in flux of contaminants from sediments to water, a uniform distribution of concentration reductions were used to simulate the effect of the remediation action in the whole system. The remediation scenarios were implemented by reducing the abiotic concentrations of PCDD/Fs in the intermediate and deep sediment compartment in Eidangerfjord on simulated date 15th of August 2015. The model simulated local reductions spanning from 0 to 1, i.e. we implemented the whole range of possible local effecacies.. Essentially, the model is run simulating the global effect of reducing the concentrations in the two deep Eidangerfjord compartments with anything from 0 to 100 %. This allows for comparing any efficacy of a remediation action with concentrations in biota and can be used to generate a plot similar to Figure 1 in order to evaluate possible non-linearities in the relationship between local and global effect.

The estimated local efficacy of a large-scale capping was taken from a pilot study initiated in the fjords in September 2009 with subsequent monitoring of both capped and uncapped reference locations. This study is detailed elsewhere (Cornelissen and others 2012). The field from which the data used here were taken were two 40,000 m^2 large fields in the Eidangerfjord where one was capped with relatively clean dredged clay mixed with active carbon (nominal 2 kg/m^2) yielding an average thickness measured after deployment of about 1.2 cm of the mixture. At 14 and 39 months after cap placement, box-core samples were transferred from these fields to the lab in order to estimate the uptake of dioxins in passive samplers placed in the overlying water

(Cornelissen and others 2012; Josefsson and others 2012; Schaanning and Allan 2011; Schaanning and others 2014). The ratio between uptake in samplers exposed in box-cores from capped and uncapped reference fields were assumed to represent the local efficacy of the capping.

To arrive at an estimate of the local effect of remediation of the contaminated sediments, including the measurement uncertainty in the experimental protocol, we derive a probability distribution of effects (P(*r*), *r* being the reduction in flux or concentration of contaminant that can be transported from the sediment compartments after remediation). This distribution will be used to subsample the remediation simulated in the main model. Measurements of single congeners are reported as having a measurement uncertainty given as a relative error of + / - 26%, i.e. that with 95% confidence the true value of the sample is within this range of the reported value (see e.g. appendix A in Schaannning and Allan (2011)). Values for the samples in 2010 and two replicate samples in 2013 were used to calculate three averages across congeners (i.e. $r = E\left(\frac{C_{water,treatment}}{C_{water,control}}\right)$). Since the efficacy is a ratio of variables, some care needs to be taken to include the uncertainty properly. Here we assume that the uncertainty in the ratio *r* can be expressed as the square root of the sum of the squared relative errors of the two variables, a fairly standard approach to error propagation; $\frac{dr}{r} = \sqrt{\left(\frac{dC}{C}\right)^2 + \left(\frac{dC}{C}\right)^2}$, where $\frac{dr}{r}$ is the relative error of the remediation effect and $\frac{dC}{C}$ is the relative error of the measurements (given as 26%). For simplicity we ignored uncertainty relating to the actual sampling method, i.e. the uncertainty we quantify is only related to detection and measurement uncertainty in the membranes and not the sampling rates in the mesocosms.

To arrive at a probability distribution for the effect of capping the sediments with active carbon we added the three normal probability distributions (one from the 2010 data and 2 replicates from 2013 (Schaanning and others 2014)) for the different samples (see Figure 3). The distribution of local effects thereby captures uncertainty both with regard to measurement (*in* the three different distributions) and variability in the efficacy of the capping (between the three distributions). This summed distribution was cut at 0 and 1, and was discretized to 100 bins for the subsampling of the remediation simulations.

RESULTS AND DISCUSSION

The fugacity model was successfully calibrated with estimates of 23 parameters as well as two error variances defining the likelihood function used (one for abiotic observations and one for biotic observations). The Markov Chain Monte Carlo (MCMC) algorithm used 120.000 iterations of which the last 50.000 were used for predicting future reductions in contaminant concentrations under both a natural remediation scenario and a scenario including capping of Eidangerfjord (area 3). Though the observed data falls within the predictive width of the simulations, concentrations in sediments are in general underestimated (median model bias MB_{sed} = 0.47) and concentrations in the water phase are slightly overestimated (median MB_{wat} = 1.12). However, the model manages to simulate the concentrations in biota rather well (see Figure 4, median $MB_{Inner\ fjords}$ = 1.01, median $MB_{Outer\ \mathrm{fjords}}$ = 0.91, for all predictions and model bias see the Supporting Information (SI)). Under a natural remediation scenario, the median model prediction is that Frierfjorden (area 1 and 2) will be classified as *Moderately polluted* (Class II) in 2039 for cod liver and 2050 for crab, respectively. For the outer fjords this classification is to be reached in years 2040 and 2062, indicating that as the concentrations in the sediments are being washed out a longer time for clearance is expected in the outer fjords (see Figure 4 and Table 1). This illustrates the temporal aspect of such persistent organic pollutants; effectively the fjords are predicted to show marked signs of pollution in biota relevant for human health concerns in close to three human generations after emission halt.

Remediation scenarios were implemented with probabilistic reductions of contaminant concentrations in Eidangerfjord (area 3, see Figure 5) in year 2015. Though there is a substantial variability in the effect on the concentrations in biota (Fig 5 C and D), there is a clear relationship between contaminant reduction in Eidangerfjord and predicted time to fall into pollution class II. The relationship between local remediation efficacy and global endpoint is indeed non-linear (see dotted black lines in Figure 5), showing that in this case Jensen's inequality can affect the results if ignoring uncertainty in local remediation efforts. When subsampling the scenarios to reflect the uncertainty in local remediation effect given a capping of the sediments in Eidangerfjord (Fig 5 E), both cod and crab are expected to fall into Class II earlier than under natural remediation (compare probability distributions in Fig 5 A and B; black distribution shows the expected year for classification II under no remediation and grey under capping). If a large scale capping of the sediments in Eidangerfjord is performed, the model predicts that Class II will be reached approximately 10 years earlier than under natural remediation (see Table 1). Though there is a nonlinear relationship between local remediation (% reduction in concentration) and the global endpoint (biota concentrations), we did not find a large effect on the median predictions of taking the uncertainty in local remediation efforts into account (Table 1), though the confidence intervals are slightly reduced when remediation is modelled as a fixed reduction in flux at 74%. Despite the non-linearity in linking local remediation effort to global endpoint, the importance of Jensen's inequality was only minor for the remediation scenario.

The model assumed that the distribution of concentrations of PCDD/Fs in biota was log normally distributed. The importance of this assumption is evident in the different predictions of classification of biota when using median and mean concentrations (Table 2), in general the median of a log-normal distribution is lower than the mean. Compared to Saloranta and others (2007) our simulations indicate a slight improvement in terms of the inner fjords being classified as *Insignificantly polluted* (Class I, see table 2). Earlier applications of the model has reported median concentrations, but we would argue that classification based on the population mean concentration is more in-line with monitoring guidelines and efforts. As medians of log-normally distributed variables are always lower than the mean, reporting median values can serve to give an impression of a cleaner biota than mean values (see SI). Samples used in monitoring for classification is most often using a mixture of individuals and such a mixture will therefore be an estimator of the mean concentration in the population. The observations used to calibrate the model application here were all mixtures of several individuals (usually 20), and therefore precludes the determination of how dioxins are distributed within a population. The current regulation of monitoring of dioxins in biota in fjords and coastal waters for Norway recommends using the arithmetic mean of samples collected to be compared to the classification limits (Molvær and others 1997).

Bioaccumulation in biota is the result of a complex interplay of physical and biochemical processes; partitioning between different phases, uptake via different routes, biotransformation, growth dilution and faecal elimination. To aid in future modelling of contaminant fate and bioaccumulation we suggest there is a need to investigate how contaminants are distributed in a population, though there have been some attempts at this from risk-analysis in a dietary perspective (e.g. Sioen and others 2007). Knowledge on how contaminants are distributed in natural populations, would not only give credence or not to our assumption of log-normally distributed contaminant concentrations, but could also affect how classification schemes should be implemented. If contaminant concentrations are log-normally distributed in exposed population, different statistics should be used to present them (e.g. means and standard deviations on a regular scale are misleading), and performing classification schemes could better be evaluated in terms of percentiles, especially when biota is consumed on an individual basis. Note that our assumption of log-normally distributed contaminant concentrations in all compartments is also used when defining the likelihood and a change in this assumption would require a full re-calibration of the model with potentially different outputs.

Multimedia models of contaminant fate and transport continue to be a valuable tool for environmental managers and stakeholders, and the output from our model provides valuable input to discussions on how to best deal with the PCDD/F polluted Grenland fjords. To remedy the polluted fjords using capping with active carbon on the scale implemented in the model here has a cost on the order of several hundred MNOK, and decisions regarding the implementation of such measures should be done with the model presented as input to a larger risk assessment of the future of the fjords.

Acknowledgements:

This study was funded by Norsk Hydro. Trine Bekkby is acknowledged for help in calculating areas and volumes for the model application as well as in producing figure 2. Additional support through the "COCO"-project (RCN Gr. no. 234388).

Supporting information available

Additional details on parameters used in the model, as well as predictions and observations for all compartments, model bias and differences in classification between median and mean concentrations in biota are presented in the Supporting Information.

References

Allan IJ, Nilsson HC, Tjensvoll I, Bradshaw C, Næs K. 2012. PCDD/F release during benthic trawler ‐ induced sediment resuspension. Environmental Toxicology and Chemistry 31(12):2780-2787.

Allan IJ, Schaanning MT, Beylich BA. 2011. Dioxins associated with suspended particulate matter in the Grenlandfjords (Norway). Norwegian Institute for Water Research. No. 6144-2011.

Bakke T, Ruus A, Bjerkeng B, Knutsen H. 2012. Monitoring of contaminants in fish and shellfish from the Grenland fjords, 2011 (in Norwegian). Norwegian Environment Agency.

Borgå K, Saloranta TM, Ruus A. 2010. Simulating climate change ‐ induced alterations in bioaccumulation of organic contaminants in an Arctic marine food web. Environmental Toxicology and Chemistry 29(6):1349-1357.

Bradshaw C, Tjensvoll I, Sköld M, Allan I, Molvaer J, Magnusson J, Naes K, Nilsson H. 2012. Bottom trawling resuspends sediment and releases bioavailable contaminants in a polluted fjord. Environmental Pollution 170:232-241.

Breedveld GD, Skei J, Hauge A. 2010. Contaminants in Norwegian fjord sediments: industrial history or future source? Journal of Soils and Sediments 10(2):151-154.

Cornelissen G, Amstaetter K, Hauge A, Schaanning M, Beylich B, Gunnarsson JS, Breedveld GD, Oen AM, Eek E. 2012. Large-Scale Field Study on Thin-Layer Capping of Marine PCDD/F-Contaminated Sediments in Grenlandfjords, Norway: Physicochemical Effects. Environmental science & technology 46(21):12030-12037.

Cornelissen G, Broman D, Næs K. 2010. Freely dissolved PCDD/F concentrations in the Frierfjord, Norway: comparing equilibrium passive sampling with "active" water sampling. Journal of Soils and Sediments 10(2):162-171.

Eek E, Cornelissen G, Schaanning MT, Beylich BA, Storholt P. 2011. Evaluation of performing thin-layered capping in Eidangerfjord and Ormerfjord (in Norwegian). Norwegian Geotechnical Institute.

Gelman A, Carlin JB, Stern HS, Rubin DB. 2004. Bayesian data analysis. Boca Raton, Fla.: Chapman & Hall/CRC.

Green NW, Schøyen M, Øxnevad S, Ruus A, Allan I, Høgåsen T, Beylich B, Håvardstun J, Gudmundson Rogne ÅK, Tveiten L. 2013. Contaminants in coastal waters of Norway 2012 (in Norwegian). Norwegian Institute for Water Research.

Josefsson S, Schaanning M, Samuelsson GrS, Gunnarsson JS, Olofsson I, Eek E, Wiberg K. 2012. Capping efficiency of various carbonaceous and mineral materials for in situ remediation of polychlorinated dibenzo-p-dioxin and dibenzofuran contaminated marine sediments: Sediment-to-water fluxes and bioaccumulation in Boxcosm Tests. Environmental science & technology 46(6):3343-3351.

Klauer B, Brown J. 2004. Conceptualising imperfect knowledge in public decision-making: ignorance, uncertainty, error and risk situations. Environmental Research, Engineering and Management 1(27):124-128.

Kulkarni PS, Crespo JG, Afonso CA. 2008. Dioxins sources and current remediation technologies—a review. Environment international 34(1):139-153.

Mackay D. 2001. Multimedia Environmental Models: The fugacity approach.

Molvær J, Knutzen J, Magnusson J, Rygg B, Skei J, Sørensen J. 1997. Classification of environmental quality in fjords and coastal waters. A guide. Norwegian Environmental Directorate (previously SFT).

Naes K, Saloranta T, Nilsson HC, Cornelissen G, Broman D. 2009. Undersøkelser for å styrke modeller knyttet til beslutningsstøtte for tiltak mot forurensede sedimenter i Grenlandsfjordene (in Norwegian). Norwegian Institute for Water Research.

Olsen M. 2012. En route to a clean fjord in Grenland - BEST final report (in Norwegian). County Governor of Telemark: County Governor of Telemark.

Persson NJ, Cousins IT, Molvær J, Broman D, Næs K. 2006. Modelling the long-term fate of polychlorinated dibenzo-p-dioxins and furans (PCDD/Fs) in the Grenland Fjords, Norway. Science of the total environment 369(1):188-202.

Refsgaard JC, Henriksen HJ. 2004. Modelling guidelines—terminology and guiding principles. Advances in Water Resources 27(1):71-82.

Refsgaard JC, van der Sluijs JP, Højberg AL, Vanrolleghem PA. 2007. Uncertainty in the environmental modelling process–a framework and guidance. Environmental modelling & software 22(11):1543-1556.

Saloranta TM, Andersen T, Naes K. 2006a. Flows of dioxins and furans in coastal food webs: Inverse modeling, sensitivity analysis, and applications of linear system theory. Environmental toxicology and Chemistry 25(1):253-264.

Saloranta TM, Armitage J, Næs K, Cousins I, Barton DN. 2006b. SF-Tool Multimedia Model Package: Model Code Description and Application Examples from the Grenland Fjords. NIVA-report 5216.

Saloranta TM, Armitage JM, Haario H, Næs K, Cousins IT, Barton DN. 2007. Modeling the Effects and Uncertainties of Contaminated Sediment Remediation Scenarios in a Norwegian Fjord by Markov Chain Monte Carlo Simulation. Environmental Science & Technology 42(1):200-206.

Schaanning MT, Allan I. 2011. Field experiment on thin-layer capping in Ormefjorden and Eidangerfjorden, Telemark. Functional response and bioavailability of dioxins 2009-2011. NIVA. No. 6285-2011.

Schaanning MT, Beylich B, Raymond C, Samuelsson GS, Gunnarsson JS. 2014. Thin layer capping of fjord sediments in Grenland: Chemical and biological monitoring 2009-2013.: NIVA. No. 6724-2014.

Sioen I, De Henauw S, Verdonck F, Van Thuyne N, Van Camp J. 2007. Development of a nutrient database and distributions for use in a probabilistic risk–benefit analysis of human seafood consumption. Journal of Food Composition and Analysis 20(8):662-670.

Walker WE, Harremoës P, Rotmans J, van der Sluijs JP, van Asselt MB, Janssen P, Krayer von Krauss M. 2003. Defining uncertainty: a conceptual basis for uncertainty management in model-based decision support. Integrated assessment 4(1):5-17.

Økland TE. 2005. Dietary restrictions i Norwegian harbours and fjords (in Norwegian).

Table 1. Predicted years for the biota to classify as *Moderately* or *Insignificantly polluted* in the Outer fjords. Listed are percentiles of years under no remediation, under uncertain remediation and certain local remediation effect. The importance of taking uncertainty in remediation efficacy into account is here only minor, with both a slightly deviance in the median prediction for crab and a reduced prediction interval when local remediation effect is assumed not to vary. The numbers for transition to class I (*Insignificantly polluted*, 15 and 10 ng/kg ww), particularly for crab, should be interpreted with care, since the simulation period ended in 2130, with many runs not reaching the limit value for class I.

	Cod liver class II (class I)			Crab hepatopancreas class II (class I)		
	No remediation	Remediation with uncertainty	Remediation without uncertainty	No remediation	Remediation with uncertainty	Remediation without uncertainty
2.5 %	2030 (2051)	2022 (2043)	2023 (2043)	2047 (2076)	2037 (2066)	2038 (2067)
50 % / median	2040 (2069)	2031 (2058)	2031 (2058)	2062 (2105)	2052 (2093)	2051 (2092)
97.5 %	2061 (2102)	2048 (2088)	2046 (2084)	2092 (2130)	2077 (2130)	2074 (2130)

Table 2: Comparison of when cod liver concentrations are expected to reach Class I in this and in (Saloranta and others 2007, marked with S2007). Our study indicates that the inner fjords will be cleared of contaminants slightly earlier than previous applications. Note, however, that we argue for the use of *mean* concentrations in biota for classification (lower row), and not *median* which was used by (Saloranta and others 2007). For a larger comparison see the SI.

Percentile	2.5	50	97.5
Year when cod liver reaches Class I in Frierfjord using median concentrations in biota.	2014 (S2007)	2026 (S2007)	2039 (S2007)
	2012 (this study)	2024 (this study)	2035 (this study)
Year when cod liver reaches Class I in Frierfjord using mean concentrations in biota	2052 (this study)	2062 (this study)	2078 (this study)

Figure 1

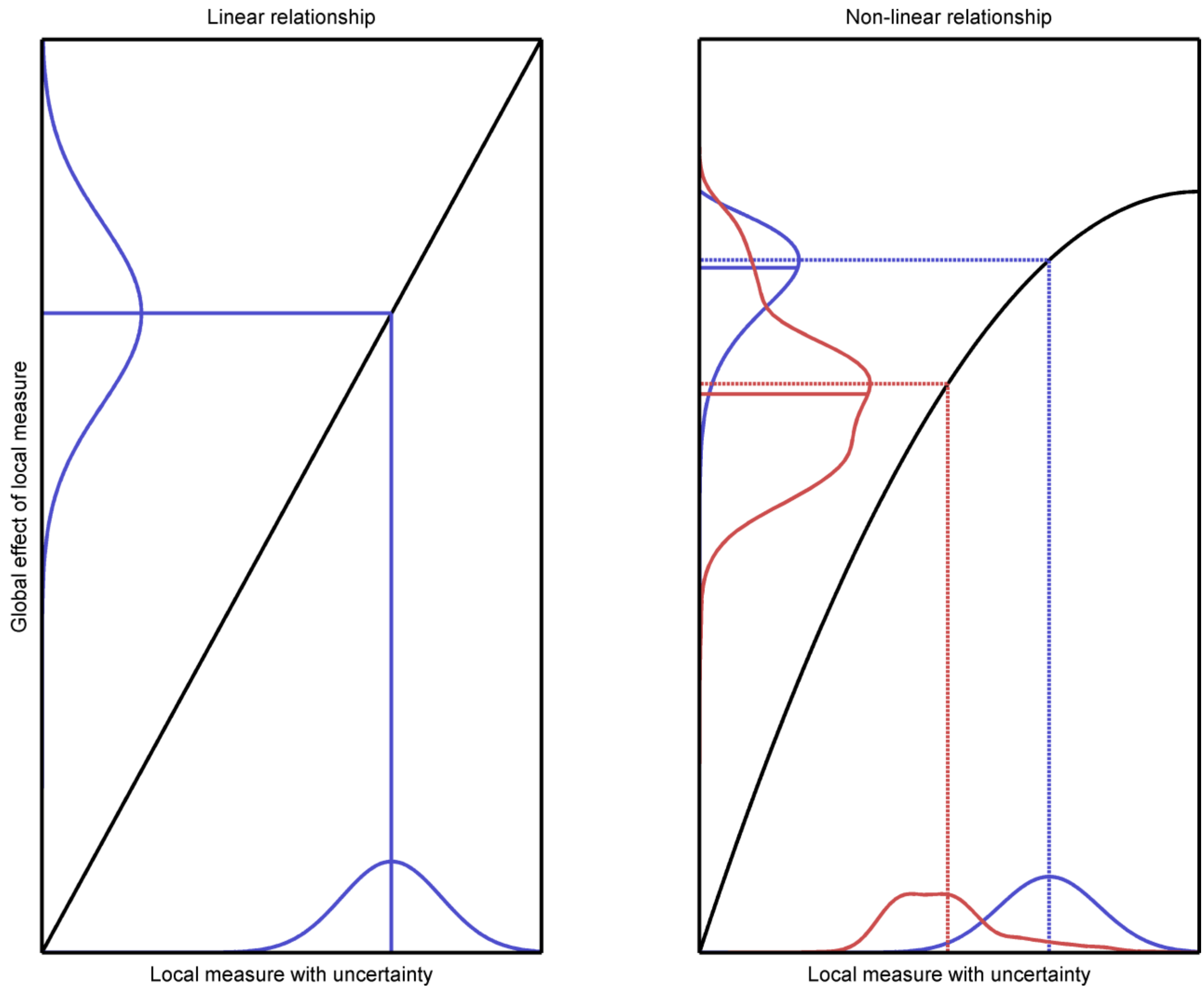
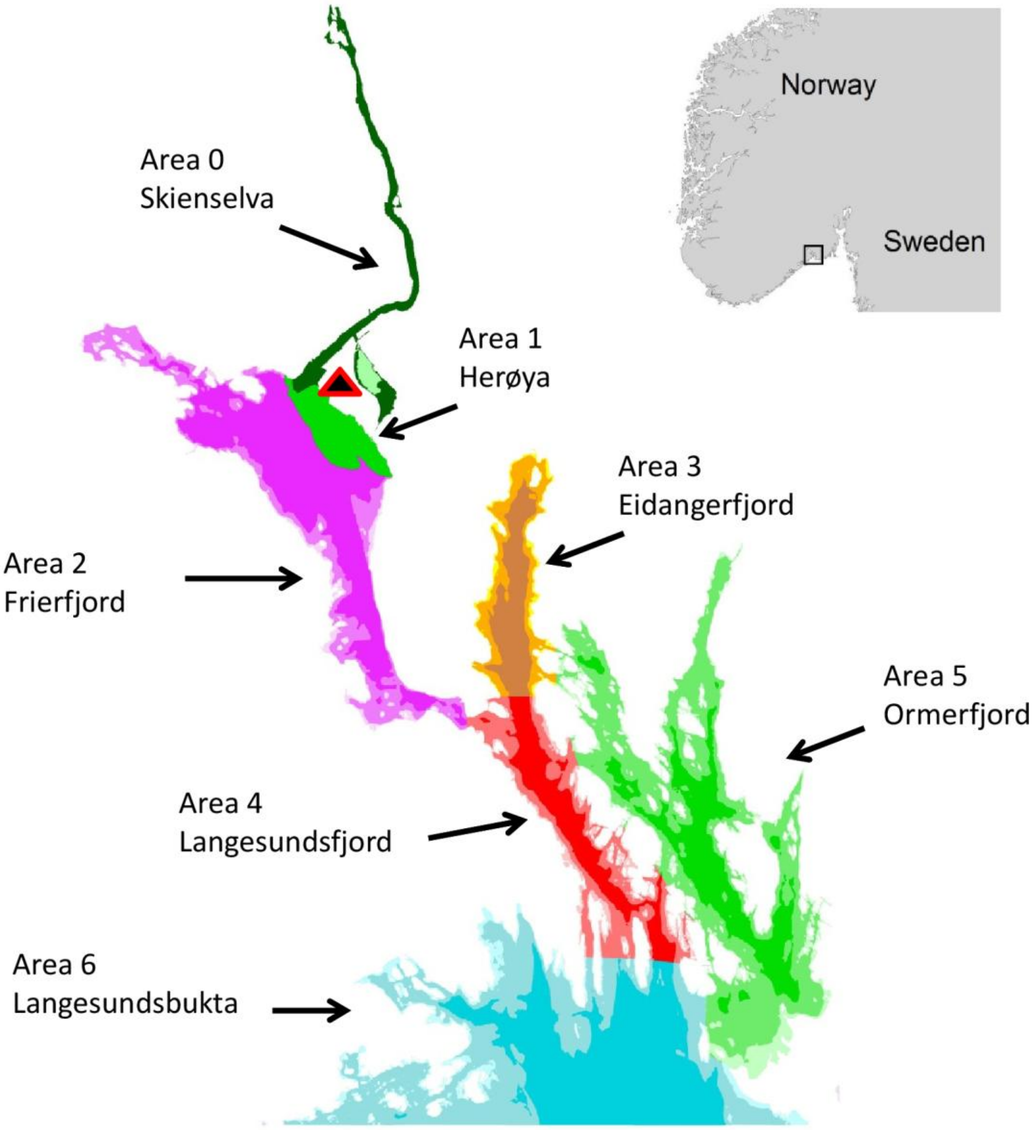

Linear relationship
Non-linear relationship
Global effect of local measure
Local measure with uncertainty
Local measure with uncertainty

Figure 2



Area 0
Skienselva
Area 1
Herøya
Area 2
Frierfjord
Area 3
Eidangerfjord
Area 4
Langesundsfjord
Area 5
Ormerfjord
Area 6
Langesundsbukta
Norway
Sweden

Figure 3

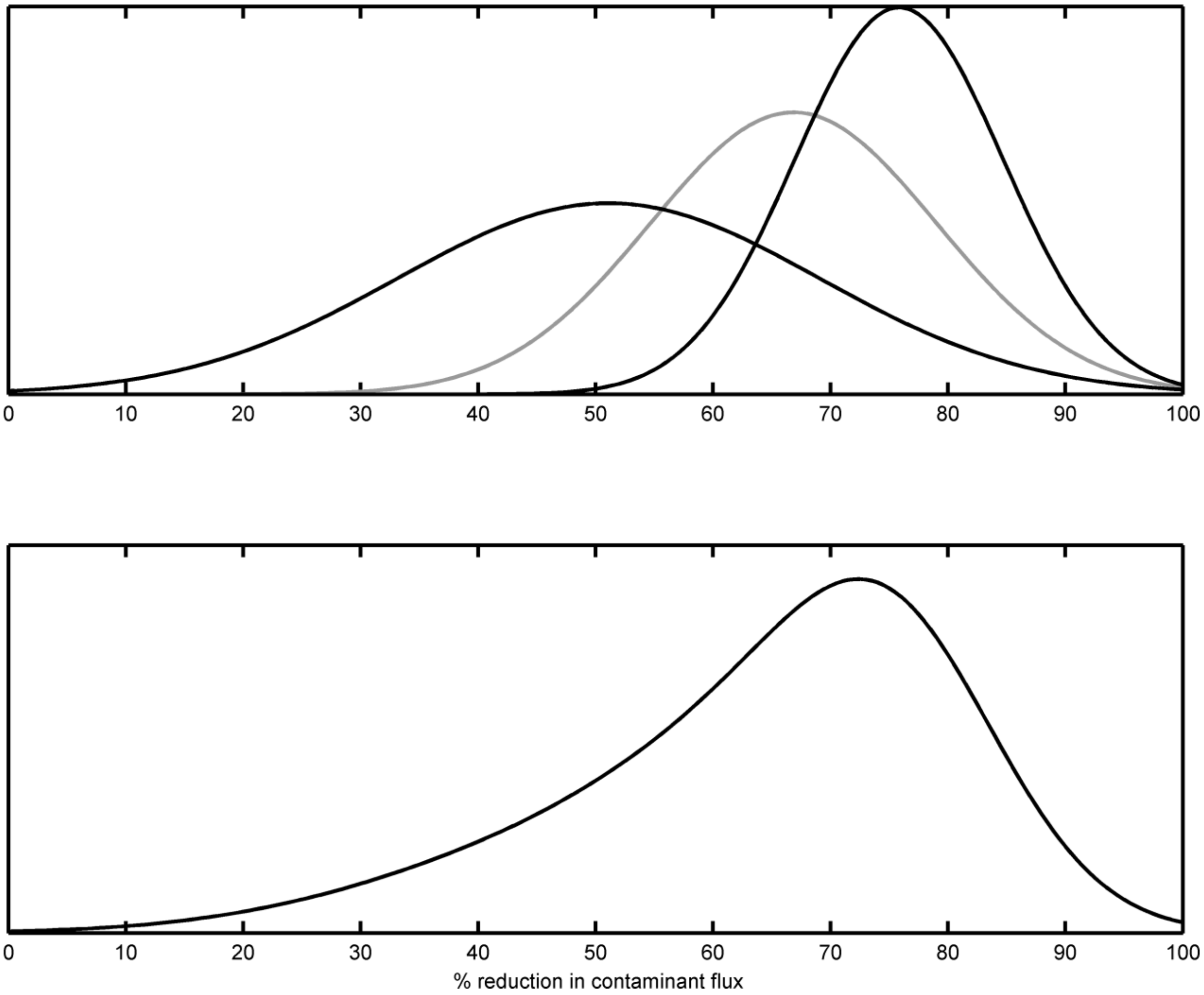


Figure 4

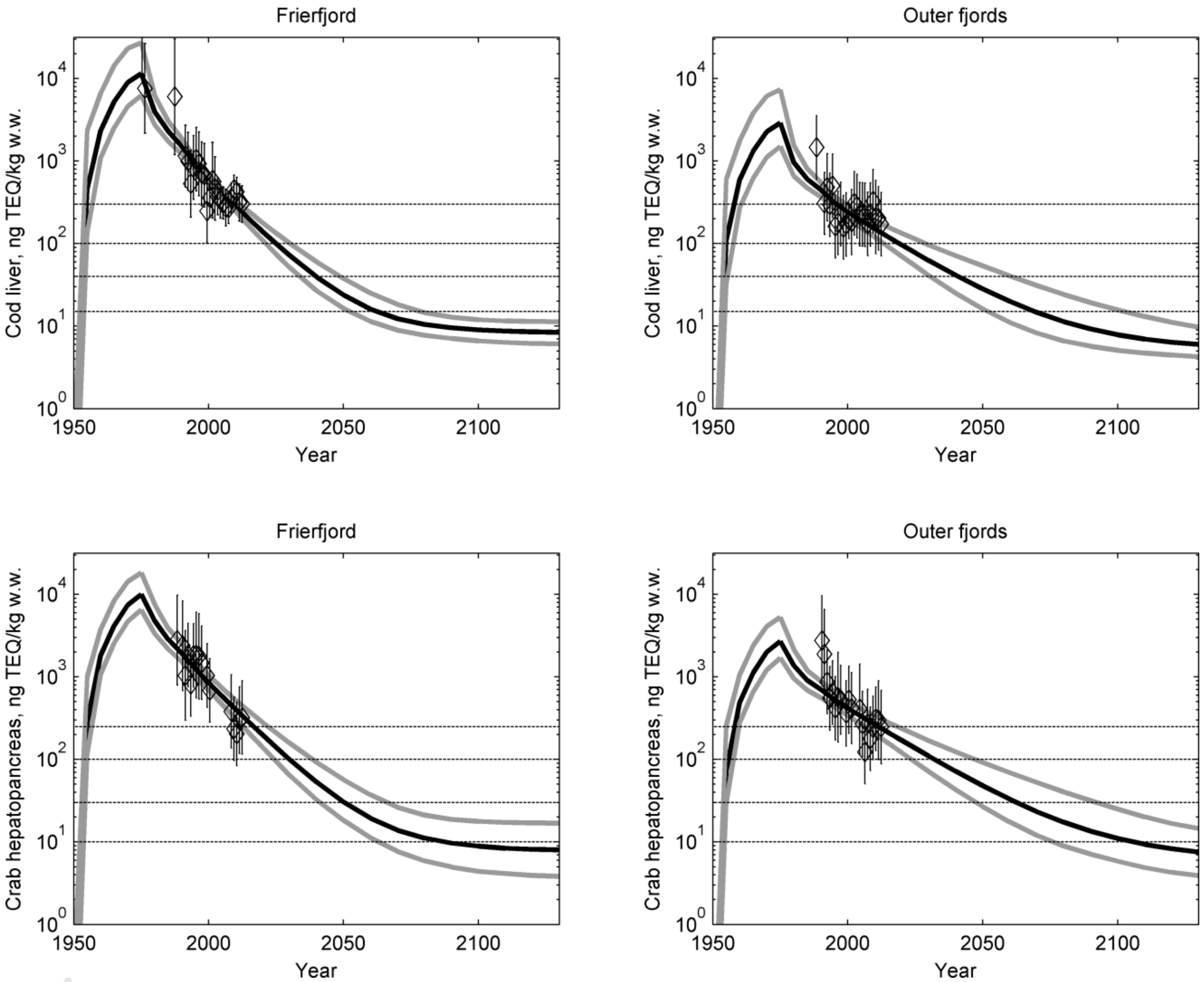
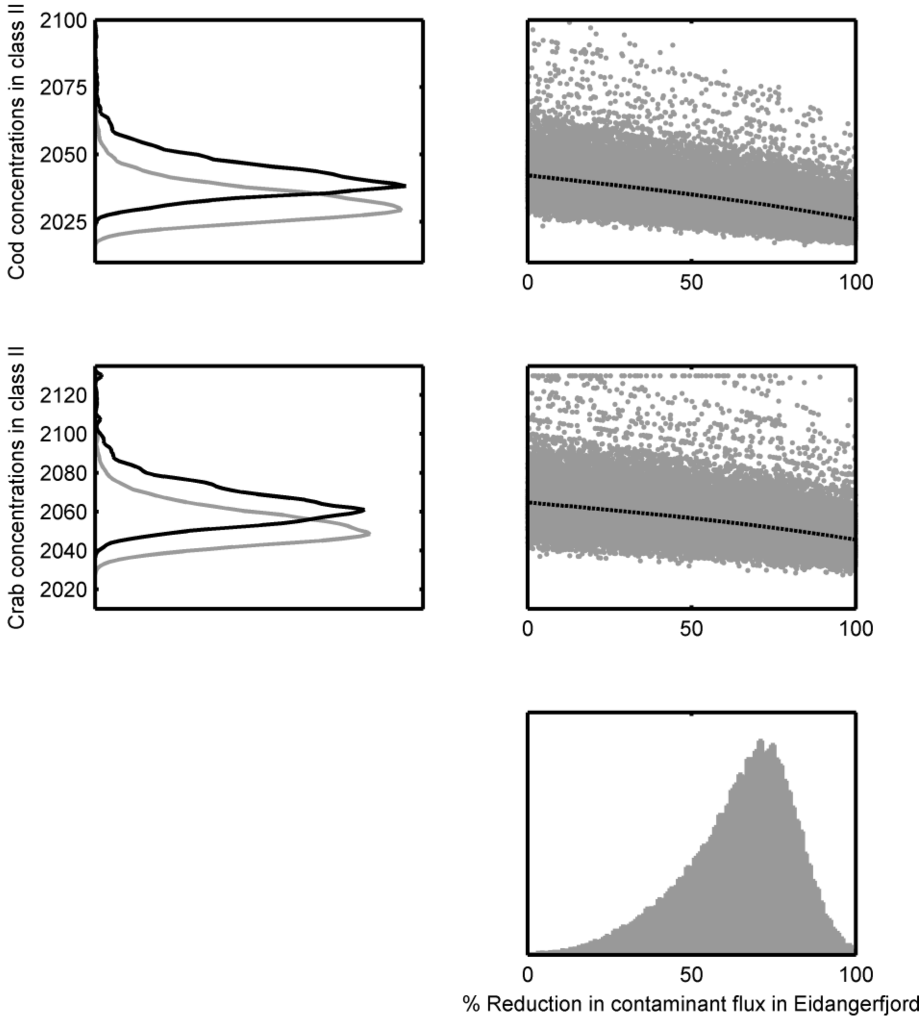

Frierfjord
Outer fjords
Cod liver, ng TEQ/kg w.w.
Crab hepatopancreas, ng TEQ/kg w.w.
Year
1950
2000
2050
2100
10^0
10^1
10^2
10^3
10^4

Figure 5


Cod concentrations in class II
2100
2075
2050
2025
0
50
100
Crab concentrations in class II
2120
2100
2080
2060
2040
2020
0
50
100
0
50
100
% Reduction in contaminant flux in Eidangerfjord