



# The role of passive sampling in monitoring the environmental impacts of produced water discharges from the Norwegian oil and gas industry

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## ABSTRACT

Stringent and periodic iteration of regulations related to the monitoring of chemical releases from the offshore oil and gas industry requires the use of ever changing, rapidly developing and technologically advancing techniques. Passive samplers play an important role in water column monitoring of produced water (PW) discharge to seawater under Norwegian regulation, where they are used to; i) measure aqueous concentrations of pollutants, ii) quantify the exposure of caged organisms and investigate PW dispersal, and iii) validate dispersal models. This article summarises current Norwegian water column monitoring practice and identifies research and methodological gaps for the use of passive samplers in monitoring. The main gaps are; i) the range of passive samplers used should be extended, ii) differences observed in absolute concentrations accumulated by passive samplers and organisms should be understood, and iii) the link between PW discharge concentrations and observed acute and sub-lethal ecotoxicological end points in organisms should be investigated.

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## 1. Introduction

Current worldwide oil and gas exploration taking place offshore in sensitive coastal environments (with regards to the habitat for organisms) puts pressure on the ecosystems in these areas. Therefore operational management in the offshore oil and gas industry plays an important role in safeguarding the environment. Produced water (PW) represents the largest volume waste stream in oil and gas production operations from most offshore platforms. Around 30% of the PW discharged into the entire North Sea results from activities carried out by the Norwegian sector (Durell et al., 2006). Stringent regulations related to permissible releases from the oil and gas industry (with regards to drill cuttings, drilling fluids and produced water) exist. Owing to the large input, the North Sea is likely the most impacted and studied recipient of diffuse chemical releases from the oil and gas industry (Bakke et al., 2013). Monitoring programs have been carried out since the 1980s in order to obtain knowledge about the impact of pollutant releases, to identify problems that may arise compromising the quality of the environment, and to define measures to avoid such unintended side effects. Through monitoring programmes, the oil and gas industry obtains

a wealth of descriptive data related to the environmental occurrence (distribution and fate) of pollutants as well as the ecotoxicological effects that these pollutants pose. However, the direct link between these parameters within monitoring programs is currently not comprehensively addressed and understanding of the effects that PW discharge can have on the marine ecosystem still remains challenging.

This article explores how chemical occurrence data could be better linked to ecotoxicological effects data within the current legal monitoring and reporting frameworks the Norwegian oil and gas industry must comply to. Using Norwegian legislation, water column monitoring requirements, and case studies, specific focus is given to the effects of PW discharges to seawater. Current monitoring practice, progressive monitoring methods and research needs are addressed. Norway is used as a case study and as of 2012, Norway was the world's third largest gas exporter and the tenth largest exporter of oil, producing 226 million cubic meters of oil equivalent (Sm<sup>3</sup>) (Ministry of Petroleum and Energy and Norwegian Petroleum Directorate, 2013). Additionally, crude oil, natural gas and pipeline services represented slightly more than half of Norway's export value, with the export of petroleum products amounting to almost 100 billion USD, i.e. nearly 10 times the export value of fish (Ministry of Petroleum and Energy and Norwegian Petroleum Directorate, 2013). Monitoring practice in Norway follows a guideline document issued by the Norwegian Environment Agency

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which was revised and updated in 2015 (Norwegian Environment Agency, 2015). A critical focus will be placed on this guideline document.

## 2. Composition of produced water (PW)

In 2012, around 130 million cubic meters of PW was discharged by the Norwegian offshore oil and gas industry (Norsk olje og gass, 2013), a volume which has been increasing due to aging of wells and the rising number of producing fields. PW includes both formation water (seawater or freshwater trapped with oil and gas in a geological reservoir) and injected water (seawater, freshwater and brine water, as well as added chemicals that are injected to enhance recovery of oil and gas, and to heighten operational safety) and as such contains components such as dispersed oil, aromatic hydrocarbons, alkylphenols, organic acids, heavy metals, radioactive materials and inorganic salts. The exact composition of a particular PW is closely coupled to the geological characteristics of the reservoir under exploration (Bakke et al., 2013; Utvik, 1999). The composition of different PWs have been well characterised in the literature (Utvik, 1999; Røe and Johnsen, 1996; Thomas et al., 2004). In addition to the chemicals found in PW from the formation water, several other chemicals of varying toxicity are added during operation together with the injected water (Norsk olje og gass, 2013).

The petroleum hydrocarbons (total petroleum hydrocarbons, TPH) contained within PW are the chemicals of greatest environmental concern and comprise a complex mixture of thousands of individual chemicals, including BTEX (benzene, toluene, ethylbenzene and xylenes) and PAHs (polycyclic aromatic hydrocarbons). BTEX, which represent the most abundant group of hydrocarbons, are also those that are the most volatile and owing to their rapid evaporation, their environmental effects are limited (Lee and Neff, 2011). In contrast, the higher persistence and toxicity of PAHs implicates them as chemicals of environmental concern (Meador et al., 1995). The 2, 3 and 4 ring PAHs are often most environmentally abundant due to their relatively high aqueous solubility, while the higher molecular weight PAHs are generally associated with dispersed oil droplets (Faksness et al., 2004). Typically the composition of alkylphenols (APs) in PW is dominated by less alkylated C1 to C3 alkylphenols, and whilst these compounds are able to exert environmental effects, they are less toxic than the higher molecular weight and branched para alkylated compounds (Beyer et al., 2012). Naphthenic acids, when abundant in the respective crude oil are also present in the resulting PW and represent a mixture of alkyl substituted acyclic and cycloaliphatic carboxylic acids that can pose an environmental threat (Clemente and Fedorak, 2005).

## 3. Current environmental legislation for monitoring marine, coastal and transitional waters

The overarching legislations that are relevant to the monitoring of marine, coastal and transitional waters in the North-East Atlantic Ocean and its adjacent seas are: (1) the OSPAR Joint Assessment & Monitoring Programme (JAMP) (OSPAR Commission, 2015), (2) the EU Marine Strategy Framework Directive (MSFD; Directive 2008/56/EC applying to coastal and marine waters) (European Parliament and Council of the European Union, 2008), and (3) the Water Framework Directive (WFD 2000/60/EC.) (European Parliament and Council of the European Union, 2000) and its daughter directive on environmental quality standards (EQS values for transitional, coastal, and territorial waters). The OSPAR Commission's strategic objective with regard to offshore oil and gas activities is to prevent and eliminate pollution caused by the offshore industry and to protect the maritime area against adverse effects, thus safeguarding human health and conserving marine ecosystems. In addition, when possible, marine areas which have already been adversely affected should be restored (OSPAR Commission, 2000). OSPAR recognises the importance of achieving synergy between

the activities outlined in the JAMP and the equivalent requirements of the MSFD and WFD. Specific to the monitoring of PW, OSPAR adopted a Recommendation for a Risk-based Approach to the Management of Produced Water Discharges from Offshore Installations (RBA Recommendation) and associated Guidelines. Full implementation of these guidelines will be in 2018. The MSFD states that strategies must be developed and implemented that protect and preserve the marine environment, prevent deterioration or, where practicable, restore marine ecosystems in areas where they have been adversely affected. Strategies to prevent and reduce inputs in the marine environment, with a view to phasing out pollution must be developed. Member states are required to determine the environmental status of their water bodies, establish environmental targets and carry out monitoring programmes. The WFD applies to waters within one nautical mile from land; therefore it is less relevant in the context of PW releases.

Within these regulations, pollutant threshold concentrations that are deemed acceptable to protect marine systems (so called Environmental Assessment Criteria by OSPAR) are provided and are primarily based on aqueous toxicity data that has been recalculated to concentrations in sediments and biota by using equilibrium partitioning models (OSPAR Commission, 2004) which may lead to erroneous overestimations of risk. OSPAR does recognise the relevance of freely dissolved aqueous concentrations of nonpolar compounds for toxicity assessment based on the weight of scientific evidence showing that such concentrations represent the direct negative effect pollutants can have on organisms (Hawthorne et al., 2007; Kraaij et al., 2003). However, freely dissolved concentrations are difficult to measure, as they are often very low, because of rapid PW dilution. Therefore OSPAR requires that these pollutant concentrations be measured in biota and sediment (Commission., O., OSPAR, 2008).

## 4. Current environmental monitoring in the Norwegian water column

The Norwegian Environment Agency is responsible for developing guidelines for monitoring the Norwegian oil and gas industry. In 2015 they published the guideline document "Environmental monitoring of petroleum activities on the Norwegian continental shelf, M-408/2015" (Norwegian Environment Agency, 2015), replacing the guideline from 2011. Thus far, monitoring data from the implementation of this guideline is not available. Monitoring is intended to indicate whether the environmental status of the Norwegian continental shelf is stable, deteriorating or improving, due to offshore operators' activities. Monitoring of both the water column and the native benthic habitat should be carried out in accordance with this guideline. Further discussion is given here about the requirements for water column monitoring since the discharge of PW to seawater is expected to affect the water column most.

Water column monitoring should be carried out every three years and surveys should include: hydrographical measurements, chemical analyses, investigations of field transplanted organisms held in cages and investigations of wild caught organisms. Hydrographic parameters that should be measured include: conductivity, temperature, density, current direction and speed. Chemical analyses that should be carried out using passive samplers includes measuring the content of total hydrocarbons, PAHs, which includes naphthalenes, phenanthrenes and dibenzothiophenes (and these are collectively referred to as NPDs) and APs. The field transplanted organisms that should be placed in cages should be mussels, dominated by *Mytilus edulis*. Passive samplers should also be placed inside the cages in the surveyed areas and be used to quantify the exposure of the mussels. Biological parameters that should be monitored for the mussels include: size, spawning status, health, PAH and metal concentrations, chromosome damage, lysosomal membrane stability and acetylcholinesterase inhibition. Wild caught organisms should include pelagic and demersal species that live in the surveyed area and should be representative of the most important

naturally occurring fish species in the region(s). Biological parameters that should be analysed for fish include: size, gender, liver somatic index, gonad somatic index, condition index, PAH, NPD and PAH metabolite concentrations, tissue changes, DNA damage and chromosome damage. In addition to these measurements, passive samplers should also be used to monitor the dispersal of pollution/discharges to the water environment and to validate dispersal models.

## 5. The role of passive sampling in water column monitoring: current status

Passive sampling involves deploying polymeric materials that can accumulate chemicals from the water phase. Passive samplers are generally classified as equilibrium or kinetic samplers differing in the operational region that they operate. Equilibrium passive samplers assume that the concentration of contaminants in the water phase is constant during the deployment period, while kinetic passive samplers give the time-weighted average contaminant concentration during deployment. Via the use of a pre-determined passive sampler-water partitioning coefficient for the chemicals of interest, the freely dissolved, risk-relevant aqueous concentrations can be determined (Huckins et al., 1990; Mayer et al., 2003) and allowing a way to use passive samplers to quantify the exposure of organisms. Passive sampling has been applied since the 1990s (Huckins et al., 1990) and over the last 25-years, method development has led to successful water column deployments in estuarine waters (Booij et al., 2002; Sacks and Lohmann, 2011; Booij et al., 2006; Monteyne et al., 2013), coastal waters and continental shelves (Munaron et al., 2012; Allan et al., 2012; Fernandez et al., 2012; Zeng et al., 2004), open seas and oceans (Cornelissen et al., 2008a; Benskin et al., 2012), lagoons (David et al., 2010), harbour waters (Adams et al., 2007), lakes (Carls et al., 2004) and bays (Friedman and Lohmann, 2014; Bao et al., 2012; Perron et al., 2013a, 2013b; Xing et al., 2009). It has resulted in the quantification of freely dissolved aqueous concentrations of a great number of organic pollutants. The development of passive sampling methods have improved the detection of hydrophobic organic chemicals to the ng/m<sup>3</sup>, and in some cases to the pg/m<sup>3</sup> concentration range (Cornelissen et al., 2008a; Lydy et al., 2014).

Several different passive samplers are able to provide a time integrated average environmental concentration when they are deployed over a period of weeks to months. Time integrated concentrations are often beneficial in environmental monitoring when compared to traditional water sampling (in which only a snap shot of information, i.e. at one particular place or time is obtained). However, the oil and gas industry is also susceptible to accidental pollutant releases resulting in spikes in aqueous concentrations. Such spikes must be captured in order to correctly assess environmental damage. Passive sampling is not able to capture such point source releases and will limit the ability of a comparison of the spikes in chemical concentrations to observable ecotoxicological effects, in particular for chemicals for which effect thresholds are exceeded following an accidental spill.

The unique environmental conditions occurring during oil and gas exploration imply that factors such as water salinity and temperature (Jonker et al., 2015), depth of monitoring and concurrent pressure, and passive sampler biofouling (Reitsma et al., 2013; Harman et al., 2009a) must be considered. The wider passive sampling literature, as well as a limited number of studies directed at the oil and gas industry have made progress with the issues related to biofouling (Harman et al., 2009a) and salinity (Jonker et al., 2015; Reitsma et al., 2013), concluding these variables can have little to a factor 3–4 effect on the passive samplers performance. The use of performance reference compounds (PRCs), which is a well-tested and documented method for several passive sampler types, allows for the degree of equilibrium of such samplers under such environmental conditions to be determined (Harman et al., 2011; Apell and Gschwend, 2014).

## 5.1. Passive sampling for determining chemical parameters in water column monitoring: current status

The role of passive sampling in water column monitoring according to the guideline document (Norwegian Environment Agency, 2015) is clearly defined and is three fold: i) aqueous concentrations of THC, PAHs, NPD and APs must be determined with the use of passive samplers, ii) passive samplers should quantify the exposure of caged organisms and iii) passive samplers should monitor the dispersal of discharges. The scientific literature provides examples of passive samplers used in offshore deployments to measure the concentration of APs and naphthenic acids using polar organic chemical integrative samplers (POCIS) (Harman et al., 2009b, 2010, 2011, 2014a), as well as NPDs and PAHs using semipermeable membrane devices (SPMD) (Durell et al., 2006; Harman et al., 2009b, 2010, 2011; Utvik and Johnsen, 1999; Utvik et al., 1999). Both types of passive samplers were deployed in the vicinity of the Statfjord B platform (UK-Norwegian boundary of the North Sea) and a concentration gradient of decreasing AP and NPD concentrations with distance from the platform was observed (Harman et al., 2010). In this study, the POCIS was only able to accumulate the most hydrophilic APs and a very large variation between replicate measurements was observed. The SPMDs also suffered the disadvantage of highly variable concentrations for larger PAHs and the authors ascribed this to accumulated concentrations being close to the quantification limits. The same two passive samplers were also deployed in the vicinity of the Ekofisk oil production platform (Norwegian sector of the North Sea, south west of Stavanger) (Harman et al., 2009b, 2011).

## 5.2. Passive sampling for quantifying the exposure of caged organisms: current status

Passive samplers should be used to quantify the exposure of caged mussels dominated by *Mytilus edulis*. Passive samplers provide a measure of the freely dissolved concentrations, and thus the chemical activity of a pollutant in (pore)water and this activity can be used in order to evaluate the exposure of organisms to the pollutants, usually expressed as bioaccumulation (Lydy et al., 2014). Owing to the similarities in uptake mechanisms for pollutants by passive samplers and organisms, a generally good correlation between concentrations of pollutants accumulated by passive samplers and organisms is observed (Lydy et al., 2014). However it is also important to consider processes that affect organisms such as bioaccumulative processes, digestive and active processes and biotransformation cannot be replicated by passive samplers. Passive samplers also provide a measure of the bioavailable concentration of pollutants and are thus more risk-relevant than a determination of total concentrations (Hawthorne et al., 2007).

There are several examples of studies in which passive samplers and caged organisms have been deployed in parallel for offshore water column monitoring. Harman et al. (2011) deployed SPMDs and caged mussels in the vicinity of the Ekofisk oil production platform noting differences in the accumulation patterns for the phases. The concentrations of PAHs and naphthalenes accumulated by the SPMDs and the mussels varied both with the sampling tool used and spatially, although overall trends were similar. For the least hydrophobic compounds this was ascribed to differences in the time taken for these compounds to reach equilibrium in the SPMD and the mussels, whereby the least hydrophobic compounds may not have been significantly accumulated by the mussels. The authors also considered the possibility that filter feeding mussels are exposed to a fraction of particle bound PAHs that may not have been sampled by the SPMDs. Durell et al. (2006) and Utvik et al. (1999) reported monitoring results of campaigns from the Tampen and Ekofisk regions, where concentrations in SPMD passive samplers were compared to accumulation in mussels (Durell et al., 2006; Utvik et al., 1999). These results showed a good agreement between the contaminant profiles observed in the SPMDs and the mussels



deployed near the platform discharges (within 1 km), and at intermediate locations. However, the actual composition of the PAH assemblage was different in the SPMDs and mussel tissue and again the authors ascribed this to differences in partitioning behaviour of different hydrocarbons between ambient water and either the mussel tissues or the SPMDs.

### 5.3. Passive sampling for monitoring the dispersal of pollution/discharges to the water environment and to validate dispersal models: current status

Within the oil and gas industry, modelling of concentrations of chemicals in PW and effects on the environment are most often carried out using the Dose Related Effects risk and Assessment Model (DREAM) (Neff et al., 2006). DREAM uses input data in the form of determined total aqueous concentrations in combination with PW volume, discharge depth, physicochemical compound data, hydrodynamic and meteorological data. Predicted aqueous concentrations using DREAM were compared to measured water concentration measured during monitoring in 2002 and 2006 in the Tampen and Ekofisk regions (Neff et al., 2006). A generally good correlation between modelled and measured concentrations was observed, except for some of the 2/3 ring PAHs at sampling stations near the source release. The authors noted that one of the short-comings of the DREAM model was that it could not distinguish between dissolved and adsorbed contaminant concentrations and thus predictions of water column concentrations are solely based on total concentrations in PW. Passive samplers have been used to monitor release from a marine deep water contaminated sediment disposal site where passive sampler-measured available concentrations (activities), turbidity and flow direction were well correlated (Cornelissen et al., 2008b). At the monitoring site downstream and for the prevalent flow direction, elevated turbidity measurements (0.1–0.2 NTU to 4–5 NTU) were accompanied by elevated PAH chemical activities (e.g. for pyrene from 1 ng/L background to 37 ng/L, and for BaP from 20 pg/L background to 900 pg/L). At the same site a single phase polyoxymethylene (POM) passive sampler resulted in similar spatial and temporal trends as compared to measurements made with the SPMD even though absolute chemical activities varied by a factor of 2 to 5 for different compounds (Schaanning et al., 2007).

## 6. The role of passive sampling in the water column: potential for development

The development and improvement of future monitoring campaigns will be driven by both the need to protect the marine environment through the implementation of legislation, and through a need for an enhanced understanding of the effects of PW on the marine environment, i.e. the goal of the monitoring program itself. These aspects will be achieved through technological and chemical advances and their enforced implementation within monitoring requirement documents. The guideline “Environmental monitoring of petroleum activities on the Norwegian continental shelf, M-408/2015” (Norwegian Environment Agency, 2015) states that “there is still a need to develop methods for conducting water column monitoring” and that “the period between two field surveys shall be used for further development and qualification of methodology. Monitoring results from previous surveys should as far as possible be used as a building block for further development and qualification of methodology”. Operators are also obliged to contribute to the development stages of new methods and when submitting reports are asked to include suggestions for such methodological developments. Such developments will also compliment the OSPAR adopted a Recommendation for a Risk-based Approach to the Management of Produced Water Discharges from Offshore Installations (RBA Recommendation). This guideline is not based on field campaigns, however as knowledge is furthered this can be used in the RBA approach.

### 6.1. Chemical parameters in water column monitoring: potential for development

In addition to the few passive sampler types that have been used to monitor the concentration of chemicals in PW discharges to seawater, several other types of passive samplers have been used in other monitoring programs or experiments to measure concentrations of chemicals that may also be found in PW, example studies are given in Table 1.

These examples highlight the potential for an extension of the current role passive sampling (SPMD and POCIS) has played as a monitoring tool for the oil and gas industry. The SPMD is a double phase passive sampler with a PE outer membrane and a triolien liquid internal receiving phase (Huckins et al., 1990). As compared to the SPMD, single phase passive samplers such as PE, POM and PDMS, generally exhibit faster uptake kinetics, and are easier to process and extract and can (thus) be less expensive. The monitoring document requires that the concentrations of THC, PAHs, NPD and AP are measured, however this neglects many other compounds present in PW discharges. One group of compounds falling in to this category is naphthenic acids and one recent study has taken steps forward with regard to their qualitative, however not quantitative monitoring in PW discharge (Harman et al., 2014a).

### 6.2. Passive sampling for quantifying the exposure of caged organisms: potential for development

There are several examples of studies in which passive samplers and organisms have been deployed in parallel in sediment environments, both in the laboratory and in the field. In-situ deployment of SPME and *Lumbriculus variegatus* in a PAH contaminated sediment resulted in a close to 1:1 correlation between SPME and *Lumbriculus variegatus* body burdens when SPME-water partitioning coefficients and biota concentration factors were used in calculations (van der Heijden and Jonker, 2009). A good correlation was also observed between passive sampler concentrations and tissue concentrations of *Macoma nasuta* when they were exposed to sediment contaminated with organochlorines, shown below in Fig. 1. However an inferior correlation was observed following exposure to a PAH impacted sediment, most likely due to biotransformation of the PAHs (Maruya et al., 2015). Freely available concentrations measured using SPME passive samplers were also able to provide a direct measure of pyrethroid activity experienced by *C. tentans* in sediment, shown below in Fig. 2 (Xu et al., 2007). Similar results exist for laboratory and field trials in which activated carbon has been amended to contaminated sediment and both passive samplers and organisms were exposed. The percent reduction concentrations measured using POM passive samplers were similar to bioaccumulation reductions measured for *Lumbriculus variegatus* (Beckingham and Ghosh, 2013) and *Neanthes arenaceodentata* (Janssen et al., 2011). A thorough review of studies that report such concentrations is given in Lydy et al. (2014). These examples, although not related to oil and gas monitoring, provide further evidence of the potential for development in this area.

It is important to realise that while these studies reported varying degrees of relationships between concentrations observed in the passive samplers and the organisms, often responses are not of the same magnitude, but are proportional (Vinturella et al., 2004). The explanations for these observations have been speculated to include the following; bioaccumulation processes such as digestion and active processes including biotransformation that can affect caged organisms, cannot be investigated by using passive samplers. However in the context of using passive samplers to quantify the exposure of caged organisms, explanations for differences observed lie in the fact that the passive samplers and organisms have different inherent sorption capacities. Only by using the ratio between the biota concentration factor and the passive sampler-water partitioning coefficient, can this be corrected for. In addition, kinetic factors such as lack of equilibration of organisms in

**Table 1**

Examples of studies in which different passive samplers have been used to measure concentrations of chemicals found in produced water.

Compound class	Passive sampler type used	Reference
TPH	POM	Muijs and Jonker (2011)
	PDMS coated glass fibers	
Alkanes	PDMS	Poerschmann et al. (2000)
Alkylated PAHs	POM	Hawthorne et al. (2011)
	PE	Reitsma et al. (2013), Choi et al. (2013)
PAHs	PE	Adams et al. (2007), Hale and Werner (2010), Lohmann et al. (2009)
	POM	Cornelissen et al. (2008b), Jonker and Koelmans (2001)
	Silicone rubber	Smedes et al. (2009)
	PDMS	Thomas et al. (2014)
	SPMD	Durell et al. (2006), Harman et al. (2009b, 2010, 2011), Utvik and Johnsen (1999), Utvik et al. (1999)
Steranes and hopanes	SPMD	Luellen and Shea (2003)
Alkylphenols	PE	Sacks and Lohmann (2011)
	ChemcatcherA	Ahkola et al. (2013), Isobe and Takada (2004)
	POCIS	Munaron et al. (2012), Harman et al. (2009b, 2010, 2011, 2014a), Vallejo et al. (2013), Arditoglou and Voutsas (2008)
Naphthenic acids	POCIS	Harman et al. (2014b)

systems where the passive sampler has reached equilibrium may contribute (based on compound hydrophobicity (Harman et al., 2011) and type of organism). Whilst it is necessary to deploy passive samplers and caged organisms alongside each other in order to measure exposure concentrations, such issues represent short comings in the current use of this method. A greater understanding of how these factors affect the use of passive samplers to quantify the exposure of caged organisms is needed. By deploying passive samplers every three years in monitoring campaigns, passive sampling data can be more easily compared over time than data of caged organisms because passive samplers are not affected by death, species shifts or other similar environmental variables.

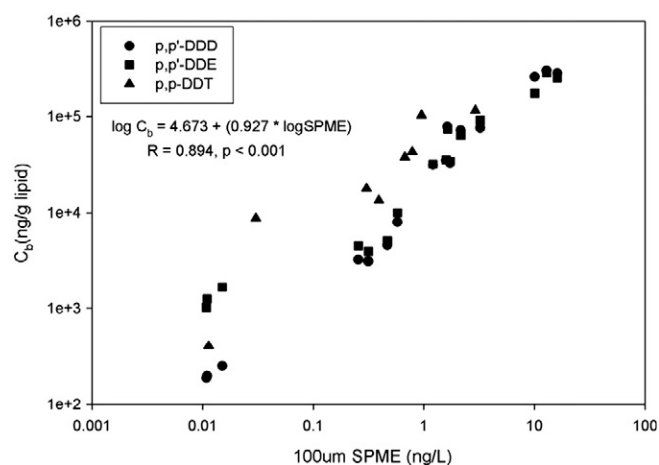
### 6.3. Monitoring the dispersal of pollution/discharges to the water environment: potential for development

When monitoring dispersal of pollutant/discharge, an important prerequisite for the method being used is that very low concentrations can be detected in order to track dispersal. When PW is discharged, the rapid dilution by the receiving water and the volatility of the chemicals in the PW present methodological challenges. Concentrations can reach background levels not far from the point of release and reliable methods able to capture these concentrations must be used in order to quantify environmental impacts. Currently, the lack of detection of a compound, if carried out with a method with a high limit of

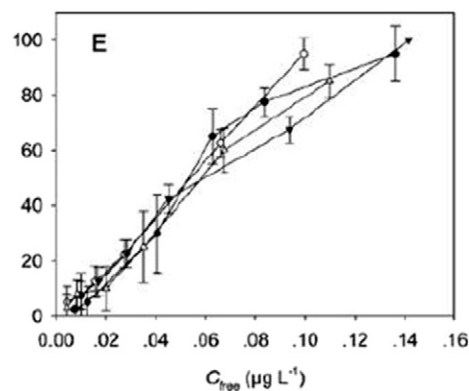
detection, can lead to false negatives and an underestimation of environmental exposure and risk. Passive sampling generally has lower detection limits than currently-used grab sampling methods and thus could extend the concentration range in which pollutants can be detected. For a number of passive samplers (POM, PE) used to sample hydrophobic compounds with large sorption capacities to environmental matrices, very low detection limits can be achieved. For example dioxins can be detected down to pg/m<sup>3</sup> levels (Cornelissen et al., 2008a). Detection limits for less hydrophobic chemicals that are for example quantified using the POCIS are generally higher (Morin et al., 2012). This would yield a greater amount of improved input data for predictive models (Bakke et al., 2013), which would benefit correlations between observed ecotoxicological effects and chemical concentrations (Grung et al., 2009; Skadsheim et al., 2009). DREAM is based on the assumption of a several 1000 fold dilution of recipient concentrations, but confirming this by applying passive sampling to capture extremely low (diluted) concentrations would allow for more accurate predictions (Carlsson et al., 2014). The DREAM model is also based on deployed mussels and currently a data gap exists with regard to the distribution and accumulation of certain compounds in the marine food web and their accumulation in fish (Chowdhury et al., 2009).

### 7. Missing link: coupling passive sampler measurements to ecotoxicological end points

A clear data, knowledge and understanding gap exists regarding the way in which discharge concentrations can be linked to observed ecotoxicological end points. Going beyond the simple relationship between



**Fig. 1.** Concentration of DDTs in *Macoma nasuta* ( $C_b$ ) vs. freely dissolved ( $C_{free}$ ) concentration in sediment porewater measured using SPME after 28 days exposure to spiked sediment. Regression analysis showed a highly significant, 1:1 log–log relationship between  $C_b$  and  $C_{free}$ . Figure kindly provided by Keith Maruya and Elsevier.



**Fig. 2.** *Chronmus tentans* mortality curves in bifenthrin-spiked sediments, with bifenthrin concentration expressed as freely dissolved pore water concentrations  $C_{free}$ . Figure kindly provided by Frank Spurlock and The American Chemical Society.

concentrations of contaminants taken up by passive samplers and organisms is necessary in order to couple the measured concentrations with acute and sub-lethal effects on organisms. Only by achieving this can the real impacts of the discharge of PW on the marine environment be understood. This data gap is clearly expressed in the Norwegian Environment Agency's new guideline document for monitoring offshore activities (Norwegian Environment Agency, 2015). Specific reference is given to the fact that for biological endpoints, quantification should involve exposure to real or man-made PW, specific components in the PW, or to extracts of real PW. By addressing this link, the chemical and ecotoxicological requirements outlined in the Recommendation for a Risk-based Approach to the Management of Produced Water Discharges from Offshore Installations (RBA Recommendation) will also be probed.

There are several examples in the literature that illustrate that toxicity assessments using passive samplers holds promise as monitoring tools via the correlation of concentrations measured with passive samplers to expected or observed ecotoxicological effects, rather than just to observed pollutant concentrations in organisms. The simplest of such approaches is a direct comparison of concentrations measured with passive samplers to water quality criteria put in to place to be protective of the ecosystem and thus based on ecotoxicological data (Maruya et al., 2012; Booij et al., 2016). Reports of comparisons between concentrations accumulated in passive samplers and resulting effects on organisms have been published, although not for offshore deployments. For example, concentrations of PAHs measured with SPME passive samplers were able to accurately predict the chronic toxicity of PAH containing sediments to *Hyalella azteca*. In this approach the toxic response was compared with the number of toxic units calculated from the SPME data and based on the US Environmental Protection Agency (USEPA) sum toxic unit model (Kreitinger et al., 2007). A similar approach was adopted by Harwood et al. (2013) in which the toxicity of bifenthrin to *Hyalella azteca* and *Chironomus dilutus* in three sediments was correlated to concentrations measured with SPME passive samplers (Harwood et al., 2013). More complex approaches have also been taken in which methods have been developed that allow a direct use of concentrations measured in passive samplers and inferred ecotoxicological endpoint response. This has been demonstrated for exposure to spiked water samples where acute water toxicity tests were carried out using *Chironomus dilutus* and *Hyalella azteca* for water samples containing a wide range of hydrophobic organic compounds. SPME passive samplers were used as a dose metric for toxicity and lethal and sublethal concentrations were expressed both on a water concentration and on an equilibrium SPME concentration basis. A significant dose–response relationship was found between the concentration measured using the SPME and the organism mortality (Ding et al., 2012).

Adopting these principles and applying them to PW samples in order to quantify the ecotoxicological impacts that actually arise following exposure, will provide a huge step forward in obtaining a greater understanding of the effects of PW releases on the marine environment. The scientific literature contains a few examples in which the ecotoxicological effects of PAHs and APs in PW on different organisms have been investigated (Beyer et al., 2012; Skadsheim et al., 2009; Brooks et al., 2011; Rastall et al., 2006; Meier et al., 2007; Sundt and Bjorkblom, 2011). Mussels were exposed to 0%, 0.01%, 0.1%, 0.5% and 1% dilutions of PW samples and significant sublethal responses in acyl-CoA oxidase activity, neutral lipid accumulation, catalase activity, micronuclei formation, lysosomal membrane stability in digestive cells and haemocytes, cell-type composition in digestive gland epithelium, and the integrity of the digestive gland tissue were observed (Brooks et al., 2011). Negative environmental endocrine disrupting responses caused by alkylphenols via a binding to and activation of fish estrogen receptors in vitro and the presence of metabolites in bile have been noted following the exposure of fish to PW (Beyer et al., 2012; Rastall et al., 2006; Meier et al., 2007; Sundt and Bjorkblom, 2011). Atlantic cod exposed to PW sampled from the Oseberg C

platform exhibited abnormalities in vitellogenin production, size and development of the fish (Tollefsen et al., 2011).

Current literature is also lacking with regard to the relationship between passive samplers exposed to PW and ecotoxicological effects to native organisms exposed to the same PW. Carlsson et al. (2014) obtained PW samples from several locations in the North Sea and then both extracted them using solid phase extraction (a method that allows the quantification of the freely dissolved concentration, although not a strict deployment of a passive sampler in the PW) and exposed them to zebrafish. Results showed that all PW samples resulted in adverse effects (increased mortality and sublethal effects) on zebrafish and that the freely dissolved concentration was in general, 10 times more toxic to the zebrafish than the particulate bound fraction. However, there was no clear correlation between the freely dissolved concentrations of PAHs and APs and the no-observed-effect-concentration for the zebrafish (Carlsson et al., 2014).

## 8. Research gaps and recommendations

Passive sampling as a monitoring tool for the oil and gas industry operating on the Norwegian continental shelf is a legal requirement. Monitoring of the water column is carried out every three years and between campaigns monitoring methods should be further developed in order to progress our understanding of the negative effects of the release of PW on the water column. The following themes are identified as areas for further research and advances in these areas should feed directly in to legislative requirements in order to improve monitoring campaigns:

- The calibration, optimisation and development of passive sampling methods for other pollutants of concern contained in PW (for example alkylated PAHs, a broader range of alkylphenols, naphthenic acids). In order to achieve this, the composition of PW samples should first be mapped and understood to identify other compounds of concern.
- The calibration, optimisation and development of passive sampling methods for real world exposure conditions (temperature, salinity, water current).
- In line with extending the range of pollutants that should be sampled, the range of different passive sampler types should also be extended in order to identify the most optimal sampler for a particular group of chemicals. Widening the scope from the currently used SPMD and POCIS will allow the most optimal (in terms of detection, ease of deployment and cost) to be identified. Several single membrane passive samplers (PE, POM, silicone rubber) may prove advantageous as compared to SPMD.
- Passive samplers that are able to mimic the exposure of PAHs, APs, THC and NPD in PW to caged mussels should be identified. Differences in the magnitude of sampled compounds by passive samplers and organisms should be better understood.
- Further attempts to validate dispersal models based on the use of passive samplers are needed. This could be addressed by increasing the spatial resolution of passive sampling investigations, including more sampling depths and more sampling locations.
- The link between PW discharge concentrations and observed acute and sub-lethal ecotoxicological effects in organisms needs to be established. Experiments should be carried out in which representative organisms as well as passive samplers are exposed to PW and observed effects are correlated with passive sampler concentrations.

Future research will try to address the above topics, aiming to advance monitoring strategies needed to control the environmental impacts of the offshore oil and gas industry.

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