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Long-term effects of thin layer capping in the Grenland fjords, Norway: Reduced uptake of dioxins in passive samplers and sediment-dwelling organisms

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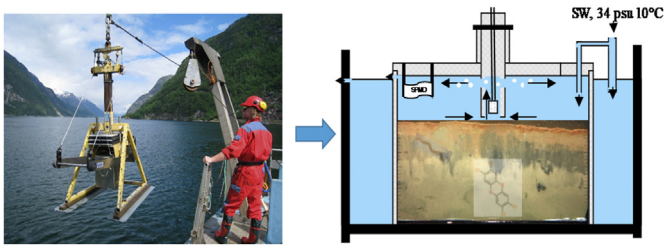
HIGHLIGHTS

- Long-term monitoring of fjord sediments capped with clay and activated carbon.
- Up to 90% reduced uptake of dioxins in passive samplers, gastropods and polychaetes.
- Despite severe recontamination, reduction was 54–61% nine years after capping.
- Deep-burrowing polychaetes showed less uptake reduction than the two other indicators.
- Thin caps without activated carbon had only short-term effects.

GRAPHICAL ABSTRACT



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ABSTRACT

The Grenlandfjords in South East Norway are severely contaminated with dioxins from a magnesium smelter operated between 1950 and 2001. In 2009, the proposal of thin-layer capping as a potential mitigation method to reduce spreading of dioxins from the fjord sediments, resulted in the set-up of a large-scale field experiment in two fjord areas at 30 and 100 m depth. After capping, several investigations have been carried out to determine effects on benthic communities and bioavailability of dioxins. In this paper we present the results on uptake of dioxins and furans (PCDD/F) in passive samplers and two sediment-dwelling species exposed in boxcores collected from the test plots during four surveys between 2009 (after cap placement) and 2018. Sediment profile images (SPI) and analyses of dioxins revealed that the thin (1–5 cm) cap layers became buried beneath several centimeters of sediments resuspended from adjacent bottoms and deposited on the test plots after capping. Uptake reduction ratios (R) were calculated as dioxins accumulated in cores collected from capped sediments divided by dioxins accumulated in cores collected from uncapped reference sediments. Cap layers with dredged clay or crushed limestone had only short-term positive effect with R-values increasing to about 1.0 (no effect) 1–4 years after capping. In spite of the recontamination, cap layers with clay and activated carbon had significant long-term effects with R-values slowly increasing from 0.12–0.33 during the first

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three years to 0.39–0.46 in 2018, showing 54–61% reduced uptake of dioxins (PCDD/F-TE) nine years after capping with AC.

1. Introduction

Polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzo-furans (PCDFs), commonly referred to as dioxins, are a group of hydrophobic, persistent compounds of major environmental concern. Dioxins may be formed in nature by volcanic activity, forest fires and microbial activity, but widespread distribution in natural environments originate from side-products of anthropogenic activities such as refuse incineration, herbicide production and chlor-alkali processes (Kulkarni et al., 2008; Tuomisto, 2019). Recycling from historically contaminated soils and sediments occurs by dissolution to pore water and overlying water, uptake in organisms at low trophic levels and biomagnification in food webs. Eventually, human consumers of fish and other edible organisms caught from wild populations may be put at risk. In European and Norwegian legislation, dioxins and dioxin like compounds are classified as priority pollutants with low environmental regulatory limits (EU, 2013/39, Norwegian Guideline 2:2018).

Sediments and biota in the Grenland fjords in SE Norway have elevated levels of dioxins due to emissions from a magnesium smelter located in the inner fjord area (Oehme et al., 1989). Emissions peaked in the early 1970's with estimated annual discharge of more than 1000 kg, decreasing to less than one kg after major technical revisions at the plant in 1976 and early 1990's (Musdalslien et al., 1991; Persson et al., 2006). The emission reductions and final close-down in 2001 have led to a 10x-100x reduction of dioxin levels in seafood organisms such as crabs, cod and blue mussels (Knutzen and Oehme, 1989; Ruus et al., 2007). However, because of remobilization from fjord sediments, another 20–50 years of natural attenuation were required before modelled dioxin levels in seafood would be compatible with safe consumption (Saloranta et al., 2008). To shorten this period of attenuation to five years or less, the model required (theoretical) elimination of the sediment source over 47% or more of the fjord area. With a fjord area of several km^2 and depths up to 70 m, traditional remediation by dredging or isolation capping was ruled out due to high costs and risk of severe ecosystem impacts. In order to investigate the potential of alternative remediation methods a large-scale field experiment was set up in 2009 (Eek et al., 2009, 2014).

At the time of set-up, evidence was rapidly increasing that release and bioaccumulation of hydrophobic compounds such as polycyclic aromatic hydrocarbons (PAHs) and polycyclic biphenyls (PCBs) could be significantly reduced by the addition of activated carbon (AC) to contaminated sediments (McLeod et al., 2004; Zimmermann et al., 2004; Millward et al., 2005; Cornelissen et al., 2011). The efficiency of AC-amendment has later been confirmed in several mesocosm and field experiments (Rakowska et al., 2011; Josefsson et al., 2012; Kupryianchyk et al., 2013; Samuelsson et al., 2015) and with significant cost reduction compared to conventional dredging (Gosh et al., 2011; Patmont et al., 2014). Potential side effects of the amendment with AC on the benthic ecosystem (Kupryianchyk et al., 2012; Nybom et al., 2015) were essentially unknown at the time. Therefore, in addition to gaining experience on the technical feasibility of thin layer capping at large water depth, the dual objectives of the field experiment were to determine the long-term effects on contaminant fluxes and bioaccumulation as well as to evaluate possible unintended effects on the benthic communities.

Three types of capping materials: 1) crushed limestone, 2) clay and 3) clay mixed with AC were added as a thin-layer cap on the sediment surface in experimental field plots at 30 m and 100 m depth. The results obtained during the first year after capping showed that thin 1–5 cm cap layers had been successfully applied and were causing significant reduction of contaminant fluxes and bioaccumulation, but also severe disturbance of the benthic community structure (Schaanning et al., 2011; Cornelissen et al., 2012; Schaanning and Allan, 2012; Samuelsson et al., 2017). Follow-up investigations in 2012/13 and 2018/19 showed sustained long-term effects at both plots treated with clay and AC (Eek et al., 2014; Schaanning et al., 2014, 2019; Cornelissen et al., 2015; Raymond et al., in prep.; Trannum et al., in prep.). Here we report for the first time a complete nine-year time-series of fluxes and bioaccumulation of dioxins determined in large intact sediment cores (boxcores) collected from experimental field plots capped with various materials (limestone, clay, AC). The results are supported by 1) analyses of sediment profile images (SPI) from frequent surveys throughout the test period and 2) chemical analyses of sediment samples from sectioned cores drawn during the final survey in 2018.

2. Material and methods

2.1. Set up of field experiment

The capping was done in September 2009 as previously described in Cornelissen et al. (2012), and Samuelsson et al. (2017). Four test sites with different treatments (Table 1) were established in two adjacent fjord areas, approximately 4 km apart and connected through several narrow sounds with typical sill depths of 10–25 m (Supplementary, figure S1). At 95 m depth, 12 km from the location of the old factory, 200 × 200 m of the seabed was capped with a mixture of activated carbon (Jacobi Carbons BP2 fine powder; average particle size of 20 μm, 80% smaller than 45 μm) and dredged clay (AC-100). Measurements at this test plot are compared with measurements at an uncapped reference plot (REF-100) located 200–300 m to the north of AC-100. In the second area located 4 km further away from the old factory site, three plots of 100 × 100 m were established at 30 m depth and treated with 1) a similar mixture of AC and clay as the one applied at AC-100 (AC-30), 2) dredged clay without any added AC (CLAY) and 3) crushed limestone without any added AC (LMS). As shown in ch. 3.1 and 3.2 below, the sediments at the 95 m site had higher concentrations of organic carbon and dioxins than the 30 m site. Within each site,

Table 1
Cap design. Last column shows mean cap thickness ± one standard deviation determined in 9–16 SPI-images taken in October 2009; 1 month after capping.

	Plot area	Clay/LMS	AC-powder	Cap thickness (mm)		
	m^2	m^3	t	kg m^{-2}	nominal	measured
LMS	10 000	500	0	–	50	21 ± 12
CLAY	10 000	463	0	–	42	37 ± 11
AC-30	10 000	220	20	2.0	20	11 ± 6
AC-100	40 000	938	60	1.5	21	12 ± 3

differences in sediment quality were relatively small.

2.2. Sampling strategy

2.2.1. SPI

During eight surveys between May 2009 (before capping) and October 2018, the test fields were investigated with SPI (Sediment Profile Imagery) to describe both physical and biological habitat (Nilsson and Rosenberg, 1997). At each survey pictures were taken in a grid with 9 points/plot at AC-30, CLAY and LMS and 16 points/plot at AC-100 (Figure S1). In this paper data from the SPI-investigation is used to determine initial coverage and successive development. More images and all results on biological interpretations of the images have been presented in Schaanning et al. (2019).

2.2.2. Boxcores

Investigations using boxcore sampling, transfer and long-time maintenance in the laboratory were done according to a standard set up used in many previous experiments (Schaanning et al., 2008; Trannum et al., 2018). The technique provides undisturbed sediment cores which are transferred to a mesocosm laboratory where each core is maintained at ambient conditions corresponding to those prevailing at the sampling site (Supplementary, figure S2).

Boxcores were collected for the first time in September 2009, one month after capping, and then again in 2010, 2012 and 2018 (Supplementary, table S1). Results from the first three surveys showed that the two treatments LMS and CLAY only caused short-time effects on fluxes and bioaccumulation. Therefore, these locations were essentially omitted during the last survey. All activities undertaken throughout this field experiment have been described in Schaanning et al. (2011, 2014, 2019); Schaanning and Allan (2012); Eek et al. (2014).

In addition to the cores sampled at the experimental plots and contaminated reference locations in the Grenlandfjord area, control boxes (CON) with mixed sediments from a non-contaminated location in the Oslofjord were included in the set-ups in 2012 and 2018.

2.2.3. Gravity corer

During the survey in 2018, sediment cores were collected from the center of all six plots using a Gemini gravity-corer with double tubes (80 cm long, 8 cm in diameter). The top 10 cm of both cores were sectioned on deck, in 1 or 2 cm thick slices, placed in polycarbonate containers with snap lids and stored frozen at −20 °C until further analyses of dioxins (PCDD/F) and sediment characteristics (C, organic C, N, and water content).

2.3. Boxcore maintenance and subsampling

Immediately after arrival at the research station at Solbergstrand (Berge et al., 1986), the boxcores fitted with transparent lids were submersed to the rim in a large tray continuously flushed with seawater supplied from 60 m depth in the adjacent Oslofjord (OSW60). The overlying water of each core was slowly exchanged with a separate flow of about 1.0 ml min^{-1} of the same source water supplied via a multichannel peristaltic pump. Each core was also continuously stirred and aerated with an air-lift system (Supplementary, Figure S3 and Widdicombe et al., 2004). Thus, the boxcores were maintained at ambient *in situ* conditions with respect to major environmental parameters, i.e. light, O_2 saturation 80 ± 10%, salinity 34 ± 1 PSU (Point Salinity Units) and temperature 10 ± 20º 20 °C.

Bioaccumulation of dioxins were measured using two test species: a gastropod *Tritia reticulata* (former name *Hinia reticulata*) and a polychaete *Hediste diversicolor* (former name *Nereis diversicolor*). Both were collected from wild populations in the Oslofjord and then added to the boxcores. The gastropod has most of the body protected by a shell and resides just below the sediment surface, i.e. above or within the layer affected by cap placement. This species accumulates dioxins mainly via selective uptake of nutrient-rich material. The omnivorous polychaete resides in burrows in and below the cap layer. The worm is exposed both through dermal exposure to the pore water and through ingestion of contaminated sediment particles into its gut. Both species are common test species used in bioaccumulation experiments (Ruus et al., 2005; Samuelsson et al., 2015).

The test organisms were exposed in the sediments for at least 2 months, which is considered suitable to obtain a steady state concentration in the organism (Lee et al., 1991). In 2010 and 2012, only polychaetes were available from the wild populations at the time of set-up and in order to obtain simultaneous retrieval of both organisms, longer exposure periods had to be implemented for the polychaetes (Supplementary Table S1). Bias of the bioaccumulation data due to prolonged exposure periods was not expected and the data did not provide any evidence for occurrence of such bias.

After exposure, all organisms were collected from the sediment and carefully rinsed in a flow of OSW60 seawater. The hard shell of the snails was cracked and removed using pincers and careful rinsing with clean seawater. The polychaetes were depurated overnight in 500 ml of seawater (OSW60). Both organisms were blotted dry before transfer to glass containers for storage at −20º CC until chemical analyses. Control samples (CON) with organisms only exposed to the sediments from their natural habitat in the Oslofjord, either as subsamples of the wild population collected before addition to the boxcores (2009, 2010) or after exposure in the control box, were included in the set-up in 2012 and 2018.

Dioxin fluxes from sediment to water were determined with two types of passive samplers LDPEs (Low Density Polyethylene membranes, 180 × 2.5 cm, average membrane thickness 80 μm) (Allan et al., 2010) or SPMDs (Semi Permeable Membrane Device, 91.4 × 2.5 cm, wall thickness 70–95 μm, filled with 1 ml triolein) purchased from ExposMeter AB (Sweden) The samplers were mounted on steel holders attached under the lid in each core and exposed in the overlying water for 3 months before retrieval. In order to avoid pulse release of dioxins from the pore water, organisms were never added to or removed from the sediments during SPMD-exposure.

2.4. Chemical analyses

Polychlorinated dibenzo-*p*-dioxins and furans PCDD/F were analyzed at Umeå University (2009 and 2010) and Ökometric, Bayreuth Institute for Environmental Research (now Eurofins Ökometric) (2012 and 2018). Basically, all samples were spiked with ^{13}C labelled PCDD/Fs to control for any loss of analyte through extraction, cleanup and analysis. The biological samples were treated with sodium sulphate, then homogenized, dried and extracted in columns successively flushed with 300 ml aliquots of acetone/hexane (5/2) and hexane/diethyl (9/1) mixtures. The passive samplers (SPMDs and LDPEs) were soaked in 150 ml cyclopentane for at least 3 d with 4x exchange of the solvent. Sediments were extracted 15 h with toluene in a soxhlet extractor. The extracts were cleaned up using a multilayer silica gel column conditioned with respectively sulfuric acid, potassium hydroxide and water, and one column with activated carbon. Sulfur was removed using activated copper. Six dioxins (PCDDs) and eleven furans (PCDFs) were finally quantified on a high-resolution gas chromatography/mass spectrometry (HRGC/HRMS) system.

The 17 components quantified are shown in figures below and

Supplementary Table S4. Toxicity equivalents (TE) were calculated using toxicity factors recommended by World Health Organization (Van den Berg et al., 2006). Sum concentrations (PCDD/F) and sum toxicity equivalents (PCDD/F-TE) were calculated assuming zero contribution for components below detection limits.

Sediment samples collected by core sectioning in 2018 were analyzed for water content, total carbon (TC), total organic carbon (TOC) and total nitrogen (TN) content using a Thermo Scientific Flash 2000 elemental analyzer at the department of ecology, environment and plant sciences (DEEP), Stockholm University. For the TOC samples, the sediment was pre-treated with hydrochloric acid to eliminate carbonates according to Hedges and Stern (1984). Water content was determined by drying wet sediment to constant weight at 70 °C. Additional weight loss after drying to 110 °C was less than 0.5%.

2.5. Calculations

The flux (F; pg PCDD/F-TE m^{-2} d^{-1}) was calculated as

$$F = M / (t \times A), \quad (1)$$

where M (pg $sample^{-1}$) is the mass of PCDD/F-TE accumulated in each sampler, t is the time of exposure in days and A is the sediment area in the core liners (0.1 m^2). The method assumes that the mass of dioxins accumulated in the passive samplers is large compared to the gain or loss imposed by concentration changes in the overlying water. If e.g. the flux from the sediment is larger than the uptake rate in the passive sampler, some of the released dioxins will be lost to the drain and the flux correspondingly underestimated. Because time of exposure (t) and sediment area (A) was always the same in the cores from capped and reference plots, this potential error in flux estimates will not affect the uptake ratios (R) (calculated as shown below) which only use the variations in dioxins accumulated either by an organism or a passive sampler.

Capping efficiencies were calculated as uptake ratios

$$R = Mcap/Mref \quad (2)$$

where M is the mass of PCDD/F-TE accumulated in the passive sampler or in the biological sample (pg g^{-1} ww) from, respectively, capped (M_{cap}) and uncapped reference sediments (M_{ref}). If replicate measurements were available, R was calculated by dividing each M_{cap} by the mean M_{ref}. Uptake ratios for the passive samplers will be referred to as flux ratios, and the notations R_f, R_g and R_p will be used for the ratios calculated from the passive samplers, gastropods and polychaetes, respectively.[1] In total, 58 uptake ratios were determined, 22 for accumulation in gastropods, 18 for accumulation in polychaetes and 18 for accumulation in passive samplers (Supplementary Table S2).

3. Results and discussion

3.1. Impact of capping on sediment biogeochemistry

The vertical profiles of carbon, nitrogen and water content measured in the sediments in 2018 (Fig. 1), showed that the capping materials had caused long-term impacts, still observable nine years after treatment. At the 30 m test site, the water content at the three capping treatments (LMS, CLAY and AC-30) was larger than at the reference plot (REF-30). Capping with clay and crushed limestone will entrain some water, which appears to remain trapped in the sediment for long time after placement. This effect was also seen at 100 m depth. At this depth the effect was less clear than at 30 m, probably because of a softer sediment with a higher initial water content. Water content determined in the first slice (0–1 cm) was always higher than in the core below, but frequently biased by incomplete draining of the overlying water during core sectioning.

Inorganic carbon was generally low (<0.5%) at the 30 m site, however, elevated to about 1% at LMS due to carbonates from the added limestone. Organic carbon (OC) was clearly elevated at both AC fields compared to the reference fields with a slight maximum at ca 4 cm depth, corresponding to the position where the cap was originally added in 2009. However, elevated concentrations of OC were also observed at LMS and CLAY where no carbonaceous material had been added, and the nitrogen content was elevated relative to the corresponding reference plots at all capped plots. The added clay was dredged from the 10–400 cm depth-interval in silty clay sediment with a similar concentration of OC as those at the test plots in Ormerfjorden (Cornelissen et al., 2012). Also, the crushed rock from the limestone quarry had very low content of organic matter (0.1%). Therefore, the increased concentration of organic matter (OC and TN) at CLAY and LMS indicated a production or accumulation of organic matter *in situ* after capping. The OC:N ratio at CLAY and LMS was relatively high (10–15) and not different from REF-30 (and REF-100) indicating that the excess organic matter was nitrogen depleted and partially degraded compared to average marine and microbial tissues. Thus, the excess nitrogen at the capped plots most likely represents refractory remnants of tissues produced by a short-term boost of bacterial activity triggered by the capping event, most likely due to perished animals. The occurrence of such an event was supported by investigations of the macrobenthic fauna which showed short-term disturbances at all capped plots and a major loss of biomass 1 month after capping at LMS (Samuelsson et al., 2017). The follow-up investigations after 1, 4 and 9 years have revealed that long-term effects on the benthic fauna occurred at the AC-treatments only (Schaanning et al., 2019; Trannum et al. in prep.).

3.2. Burial and recontamination

Low nitrogen concentrations in activated carbon provide elevated C:N ratios in sediments spiked with AC (Pallares et al., 2018; Bonaglio et al., 2020). If the C:N ratio is used as tracer for the added AC-clay layers, the maxima at 4–5 cm depth at AC-30 and AC-100 (Fig. 1) indicated that the added cap layers had been covered by new sediment at a rate of about 5 mm y^{-1}. This was confirmed by the SPI-images which showed remnants of AC present as black bands below a more brownish surface layer with thickness increasing with increasing time after cap placement (Fig. 2A). Careful analyses of all of the 46 SPI-images taken in the pre-defined grids during the 2018 survey, showed that the boundary between the black band and the brownish surface layer was present at an average depth of 2.0 ± 0.6 cm at AC-30 (mean ± 1 standard deviation, n = 17) and 4.6 ± 0.9 cm at AC-100 (n = 29). This corresponded to sediment growth rates of 2.2 ± 0.7 mm y^{-1} at AC-30 and 5.1 ± 1.0 mm y^{-1} at AC-100.

The sediment growth rate estimated at AC-100 was further supported by a major increase of dioxin concentrations from 699 pg g^{-1} TE at 8 cm to 1351 pg g^{-1} TE at 10 cm (Fig. 2B). At a growth rate of 5.1 mm y^{-1}, as indicated by the SPI-images, this showed a decrease of the sedimentation of dioxins 17 years before the cores were sampled. This agreed well with the closure of the magnesium-smelter and end of direct emissions of dioxins in 2001

[1] An uptake ratio or capping efficiency of e.g. 0.2 means that the flux or tissue concentration at the capped plot is 20% of the flux or tissue concentration at the uncapped reference plot, or 80% lower in capped than in uncapped sediments. Ratios <1.0 mean that capping had a positive effect and ratios ≥1.0 mean that capping had no or negative effect.

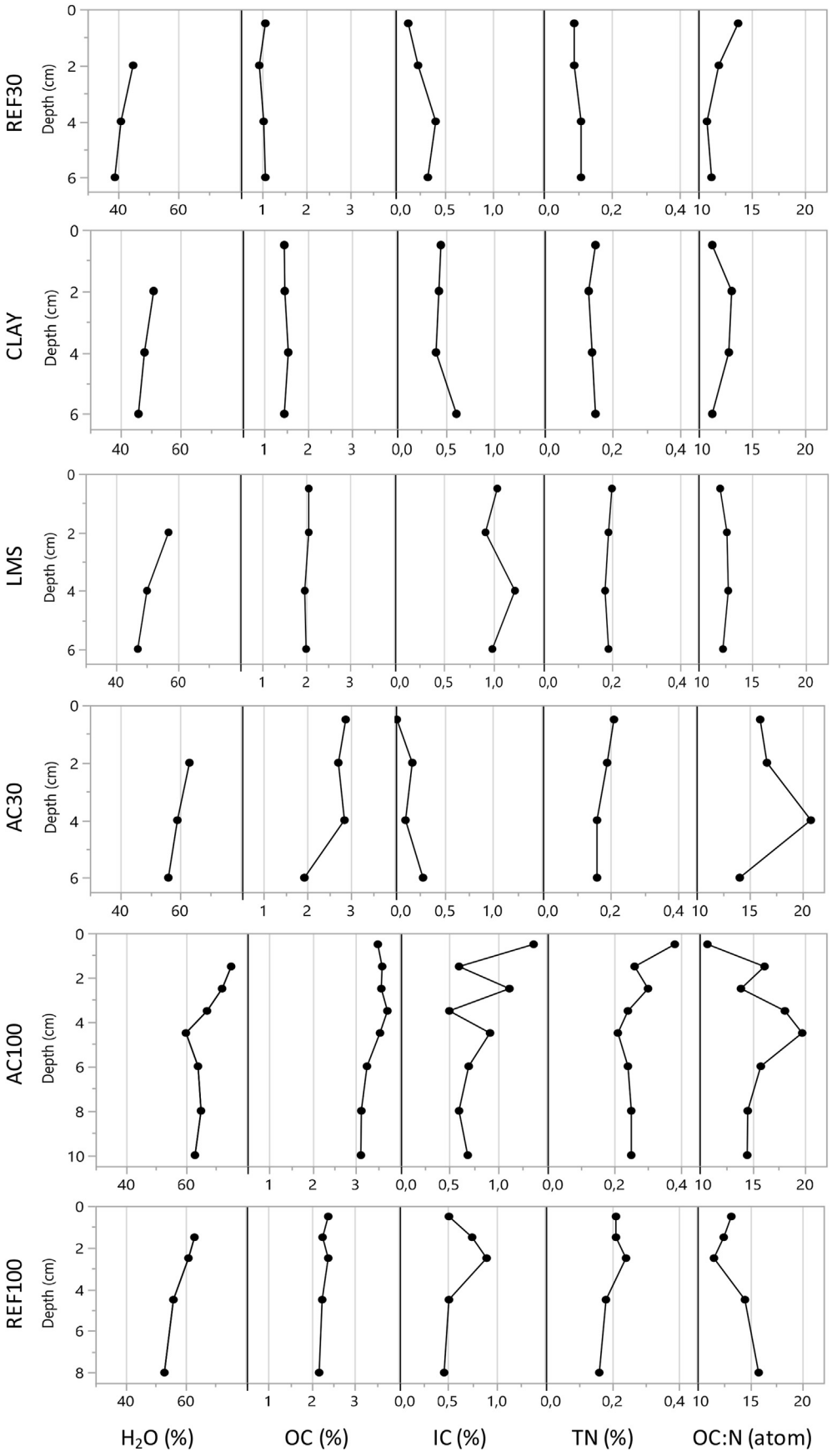
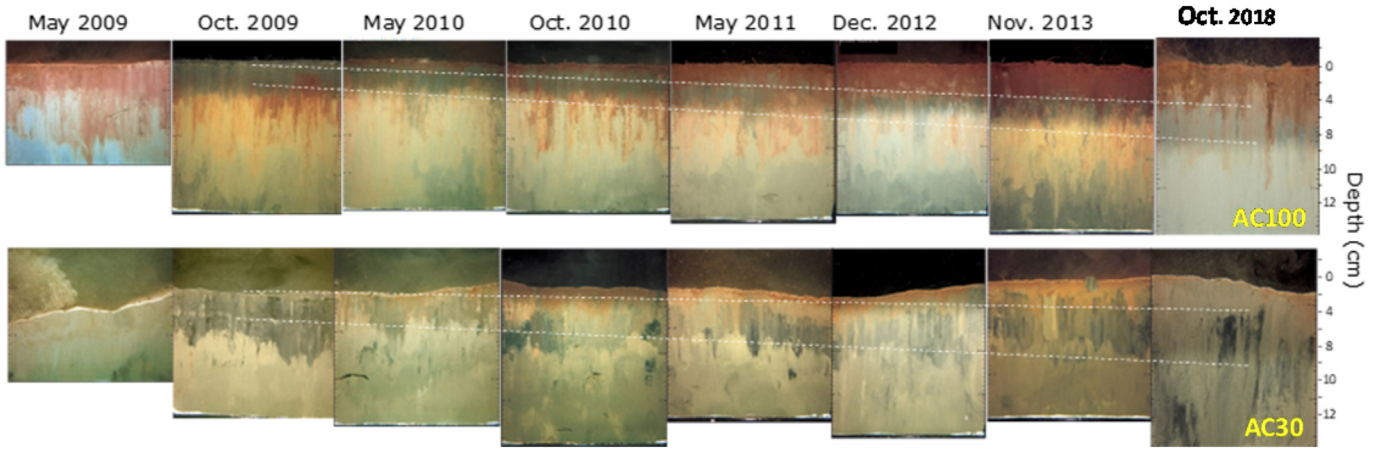


Fig. 1. Water content, organic carbon (OC), inorganic carbon (IC), nitrogen (TN) and carbon:nitrogen ratios in sediment cores sampled at the test fields in October 2018, 9 years after cap treatment.



B) AC and REF nine years after capping

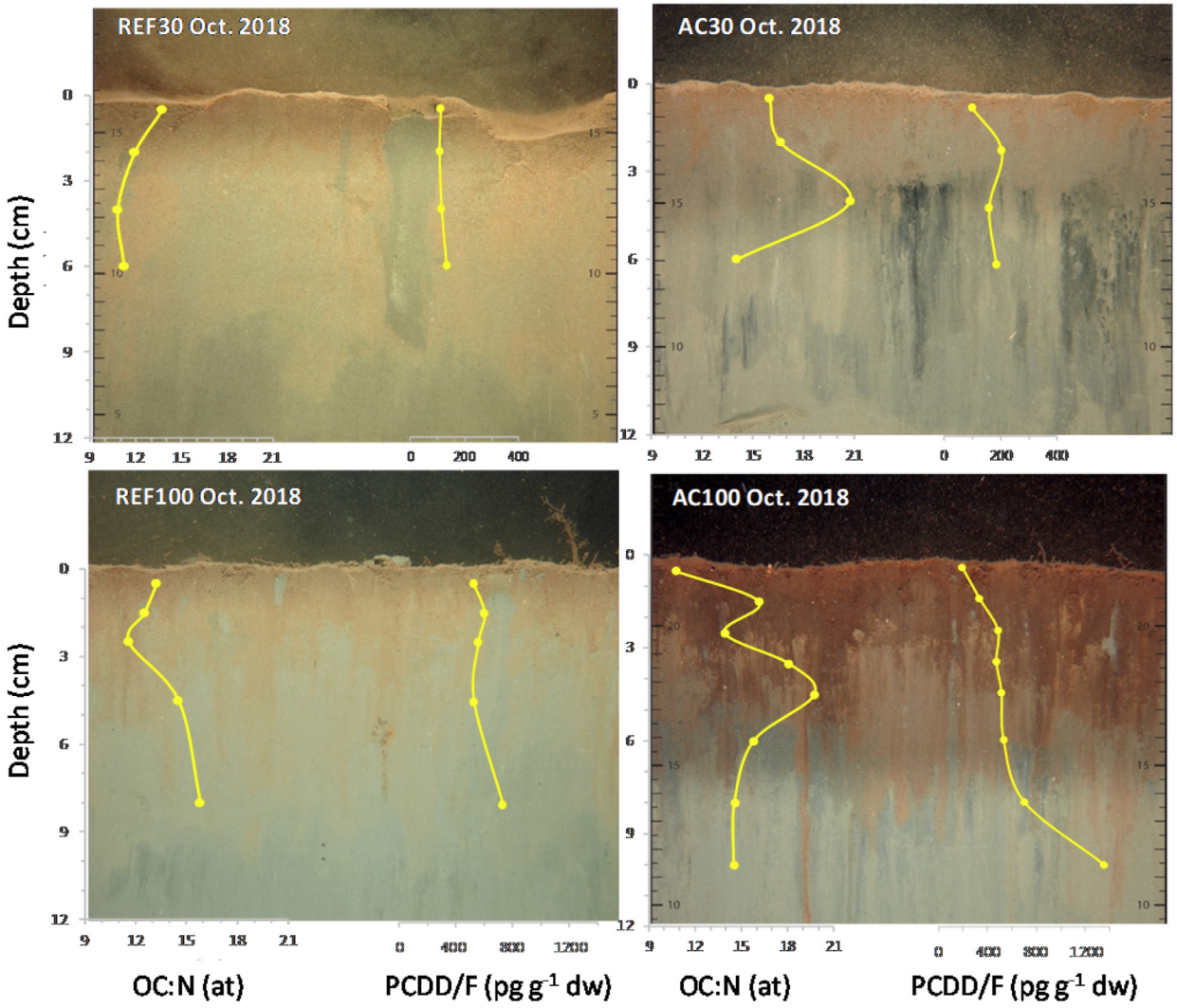


Fig. 2. Sediment Profile Images (SPI). A) Time series at AC-30 and AC-100. Top and bottom of cap layer is indicated by white lines. B) Vertical profiles of C:N ratios and concentration of dioxins (pg g^{-1}) superimposed on SPI-images from 2018; nine years after cap placement.

(Persson et al., 2006).

The vertical profiles showed that concentrations of dioxins were not much lower above than below the cap layers (Fig. 2B). This was consistent with the findings of Allan et al. (2011) who showed a concentration of 447 pg g^{-1} PCDD/F-TE in suspended particles collected above the AC-100 plot at a water depth of 89 m. This was not much different from the concentrations found in the sediments and 4–9 times higher than the concentrations found in suspended particles collected at 5 m depth. Furthermore, a principal component analyses (PCA) showed that the distribution patterns for the 17 PCDD/F components in the suspended matter resembled the pattern found in the bottom sediments (Allan et al., 2011). They explained these findings with internal sediment cycling. i.e. resuspension from basin slopes combined with lateral transport and focusing of sinking particles in a deep and narrow fjord (Colin et al., 1986; Isla et al., 2004; Katz et al., 2012). The importance of biological and other drivers of resuspension from basin slopes are not well understood, but important to consider when planning remediation in a deep fjord environment.

3.3. Dioxin contaminant levels and congener distribution

The concentrations of dioxins were high in both fjord areas, but lower at the 30 m location (99–204 pg g^{-1}) than at the 100 m location (192–1350 pg g^{-1}) (Fig. 2B). The difference between the two locations most likely results partly from different distance to the source (Cornelissen et al., 2012, 2015) and partly from internal recycling and accumulation on the deep fjord bottom (Allan et al., 2011). Compared to the environmental quality standards (EQS) given in the European and Norwegian regulatory framework, the maximum admissible concentration (MAC-EQS) of dioxins (PCDD/F-TE) in coastal sediments was exceeded by a factor of 30–375 (Norwegian Guideline 2:2018).

The relative congener distribution was similar at both experimental sites. Most of the toxicity of the 17 dioxin and furan components was accounted for by the five lighter furans 2378TeCDF to 123678HxCDF with some smaller contributions from 12378PeCDD and 1234678HpCDF (Supplementary, Figure S4, S5). The same compounds accounting for most of the toxicity equivalents in the sediments, also accounted for most of the toxicity equivalents found in passive samplers, gastropods and polychaetes (Supplementary, Figure S6).

3.4. Fluxes and bioaccumulation of PCDD/F-TE

3.4.1. Fluxes

The highest fluxes were measured in sediment cores from the uncapped plots (REF-30 and REF-100) and the two plots capped with clay (CLAY) and crushed limestone (LMS) without added AC (Fig. 3). At the two plots treated with AC (AC-30 and AC-100) the release of dioxins was lower than at the other four plots, and significantly lower than the release from CLAY. The lowest flux of PCDD/F-TE was measured in the clean control (CON). The general magnitude of the fluxes determined here compared well with the fluxes of 1–8 pg m^{-2} d^{-1} measured in benthic chambers deployed in the field in 2009–2011 (Cornelissen et al., 2012). Considering that O_2 deficiency will eliminate bioturbation and hence underestimate fluxes in the closed, bell-jar type of benthic chambers applied in the field measurements (Mustajärvi et al., 2017). On the other hand, as discussed in section 2.5, the flux determined in the flow-through chambers may have been similarly underestimated due to loss of dioxins to the drain.

3.4.2. Bioaccumulation

Bioaccumulation of dioxins by gastropods was highest in organisms exposed in the same sediment cores as those providing the highest release fluxes, i.e. CLAY, LMS, REF-30 and REF-100 (Fig. 3, middle frame). Median concentrations in gastropods from these cores ranged from 2.1 to 4.0 pg g^{-1} compared to 1.2 pg g^{-1} at AC-100, 0.89 pg g^{-1} at AC-30 and 0.32 pg g^{-1} in CON (all on wet weight basis). The statistical comparison showed significantly lower bioaccumulation in gastropods at the AC-plots than at the respective reference plots (Fig. 3).

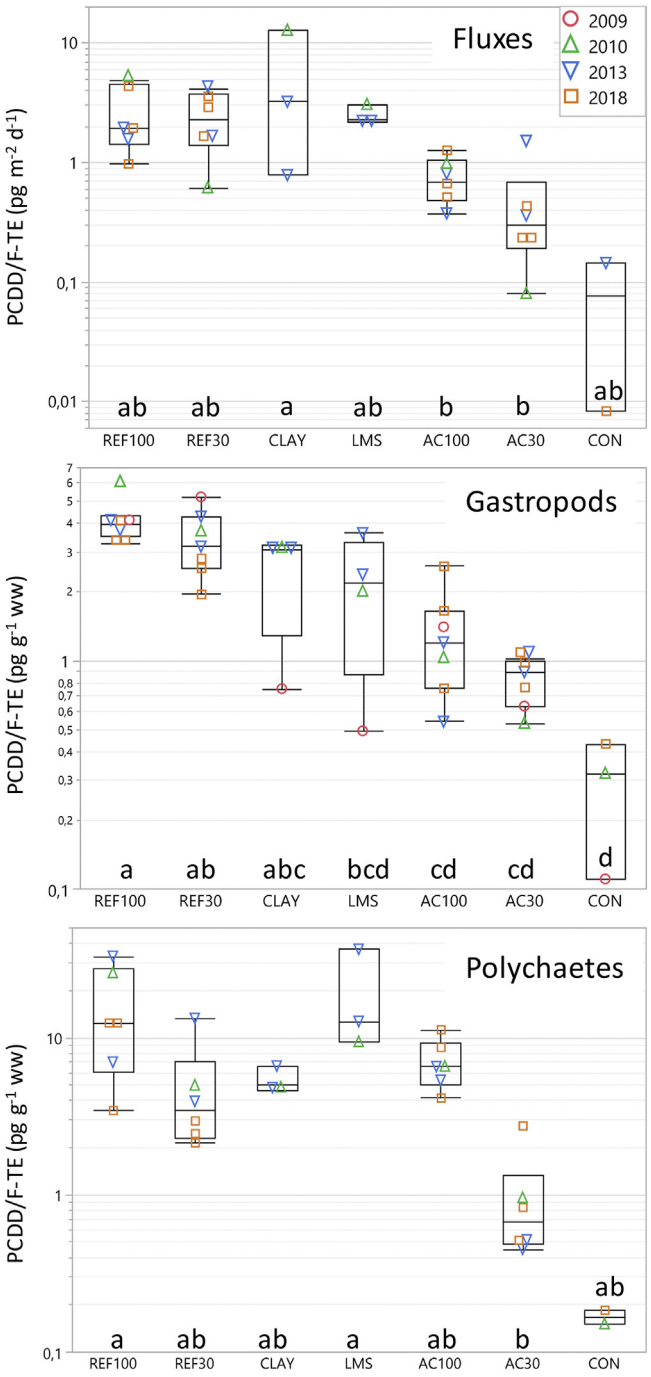


Fig. 3. Fluxes and bioaccumulation determined in boxcore samples from capped and uncapped test plots in the Grenlandfjord area and core liners with mixed control sediments (CON) from the Oslofjord. The box plots show 1st quartile, median and 3rd quartile. Outliers are shown as single points above or below end of whiskers. Letters are taken from connecting letters report in Tukey-Kramer analyses of variance (Supplementary Table S5).

Both species had similarly low (<0.5 pg g^{-1} ww) tissue concentrations in the clean control sediments (CON) (Fig. 3, middle and lower frame). In REF-30 and REF-100, however, median and maximum concentrations were higher in the polychaetes (4.45 and 32.9 pg g^{-1}) than in the gastropods (3.51 and 6.8 pg g^{-1}). This showed that both species had similarly low levels when exposed in non-contaminated sediments, but that the polychaetes accumulated more PCDD/F than the gastropods when transferred to exposures in dioxin-contaminated sediments from the Grenlandfjords.

The two organisms have different physiology and different life strategies (Ruus et al., 2013). The polychaetes have a large unprotected body surface and reside in burrows down to 15–20 cm depth at which dioxin concentrations may be higher than in the more surficial habitat of the gastropods. Considering the increase of the sediment concentrations observed below 9 cm depth at AC-100 (Fig. 2B), the burying activity of the polychaetes suitably explains the large span of concentrations in this organism, ranging from 4 to 11 pg g^{-1} ww in replicate samples from AC-100 in 2018 (Fig. 3). The burying activity of the polychaetes was also suitable to explain the small difference between dioxin concentrations in gastropods and polychaetes exposed in control sediments, which unlike the other sediments were mixed before exposure. Thus, no vertical gradients were present to increase accumulation by the polychaetes exposed in this sediment.

In the capped sediments, accumulation by polychaetes from CLAY and LMS was not significantly different from accumulation at the REF locations. Compared to REF-30, median accumulation was rather higher at the two fields capped with materials without AC added (Fig. 3, lower frame). Thus, neither fluxes nor bioaccumulation by any of the species provided any evidence for long-term effects of thin caps with these materials. Only the surface-dwelling gastropods exposed in 2009 shortly after capping, showed clear reduction compared to the REF locations.

Also, AC-100 provided frequently high levels of dioxins in the polychaetes. However, the median concentration of 6.5 pg g^{-1} ww was about half the concentration in polychaetes from REF-100 (12.5 pg g^{-1} ww). At AC-30, the bioaccumulation in polychaetes was significantly lower than at LMS and REF-100.

The higher sediment concentrations at the deep site compared to the shallower site, suitably explains the higher bioaccumulation in polychaetes from REF-100 compared to REF-30 and in AC-100 compared to AC-30. However, the difference was less clear for fluxes and bioaccumulation in gastropods which tend to suggest that the burrowing potential of the polychaetes may be particularly important at the deep location where the sediment was softer and more nutritious with twice as much OC and N below 5 cm depth (Fig. 1).

3.5. Cap efficiency

The ultimate objective of the capping operation was to reduce the bioavailability of the contaminants present in the sediment. Bioavailability can be measured as uptake in an organism or in a passive sampler (Peven et al., 1996). In this work, bioavailability was measured using passive samplers to indicate uptake by diffusion of truly dissolved dioxins from the overlying water and two different organisms to indicate uptake at different depths and via different pathways and mechanisms (Weston et al., 2000). Variations due to differences in sediment properties, exposure times, season, condition of test organisms etc. will be eliminated when dividing by the corresponding result obtained in the reference sample. By pooling uptake ratios measured with three different indicators on bioavailability (R_f, R_g and R_p) we can compare more data points and obtain a broader and more robust measure on the reduction of bioavailability of the contaminants present in the sediments.

Comparing all uptake ratios for each matrix (Fig. 4), median capping efficiencies were 0.27 for the flux (R_f), 0.32 for the gastropods (R_g) and 0.60 for the polychaetes (R_p). Thus, the polychaete ratios (R_p) were clearly higher than R_f and R_g. Omitting the three outlier ratios shown in the Fig. 4, statistical comparison showed a high degree of consistency between R_f and R_g ($p = 0.92$). R_p was, however, significantly higher than R_f ($p = 0.037$), and close to significantly higher than R_g ($p = 0.058$) (SupplementaryTable S6). The higher R_p values found here, were consistent with previous findings of higher uptake in deep-burying polychaetes (*Marenzelleria* spp, *Hediste diversicolor*) compared to surface-dwelling species (*Monoporeia affinis, Abra alba*) reported to result from subsurface contaminant reservoirs being more accessible to the polychaetes (Josefsson et al., 2011; Samuelsson et al., 2015).

Comparing the different treatments (Table 2, first 3 rows), median uptake ratios of 0.12–0.51 showed that for the entire nine years period after capping the bioavailability had been reduced by 49–88% at plots treated with AC. The ratios of 0.59–1.89 at CLAY and LMS, showed little or no effect at the two plots treated with clay or crushed limestone without activated carbon. As shown in Fig. 3, none of the three indicators provided any significant reduction of dioxins in LMS or CLAY compared to REF-30.

Time trends (Tables 2 and 4 last rows) for the different treatments showed that the low ratios obtained in 2009 were not maintained in any of the later surveys at CLAY and LMS when uptake ratios were close to or higher than 1.0 indicating no long-term effects on bioavailability of dioxins, neither in gastropods, polychaetes or fluxes. This led to the decision of focusing mostly on the AC treatments in 2018 and limit the investigations at CLAY and LMS

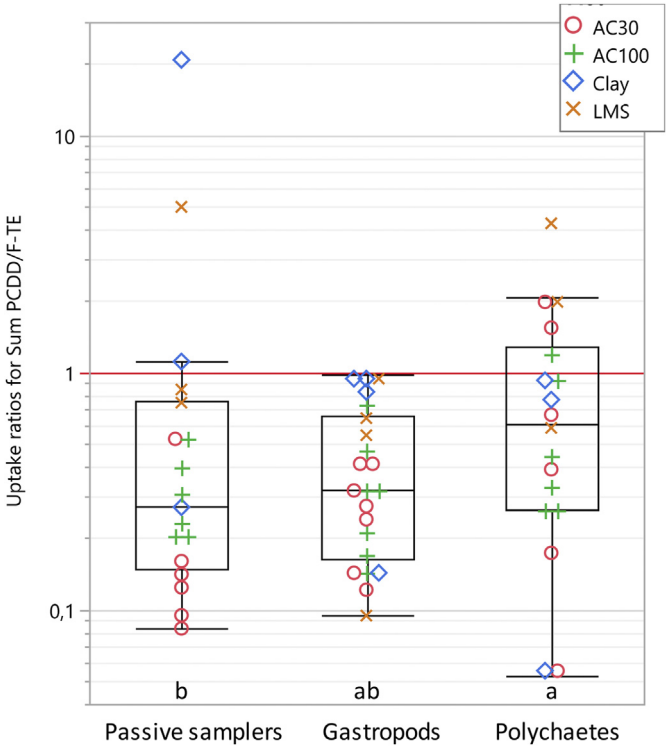


Fig. 4. Uptake ratios for passive samplers (flux), gastropods and polychaetes. The box plots show 1st quartile, median and 3rd quartile. Three outliers are shown as single points above end of whiskers. All measurements 2009–2018 (N = 58). Letters from Tukey-Kramer analyses of variance, Connecting letters report (Supplementary table S6) shows significant difference ($p < 0.05$) between polychaetes and passive samplers.

Table 2
Median uptake ratios for dioxins (PCDD/F-TE) in passive samplers (R_f), gastropods (R_g) and polychaetes (R_p) for different thin cap treatments. The last 4 rows show time trends based on pooling the three matrixes. (Details in Supplementary, figure S5 & S6).

R	Year	N	AC100	AC30	CLAY	LMS
R_f	all	18	0,246	0128	1110	0,789
R_g	all	22	0,312	0266	0,829	0592
R_p	all	18	0,383	0509	0,767	1891
All[a]	2009	4	0,326	0121	0,144	0094
All	2010	12	0,198	0143	0,929	1891
All	2012	24	0,291	0253	0,817	0767
All	2018	18	0,463	0390	–	–

[a] Only $R_g$measured.

to SPI-photography and core sampling for analyses of biogeochemical parameters.

At AC-30 and AC-100 the uptake ratios remained low throughout the investigation period. At the last survey, the ratios had increased slightly from 0.12 to 0.33 throughout 2009–2012 to 0.39–0.46 in 2018.

The uptake ratios observed here compared well with corresponding ratios determined for uptake of PAHs and PCBs in clams ($R = 0.20$) and polychaetes ($R = 0.31$) exposed in sediment cores collected shortly after thin layer capping with bentonite clay and AC in Trondheim harbor (Samuelsson et al., 2015). They also compared well with uptake ratios of 0.27 found for PCBs in passive samplers five years after AC-amendment at tidal flats in San Fransisco Bay (Cho et al., 2012).

The long-term maintenance of more than 50% reduced bioavailability for at least nine years after capping, has not been shown before and occurred in spite of burial of the added caps below 2–5 cm new sediments contaminated with dioxins at concentrations similar to the concentrations in the sediments deposited before capping. The maintenance of high capping efficiencies requires that dioxins from the new material deposited on top of the cap layers are brought into physical contact with available sorption sites on the activated carbon (Werner et al., 2006). In natural environments, bioturbation is probably the most important mediator of this contact (Lin et al., 2018). Bioturbation may act by mixing AC added on top of the sediments downwards to bind contaminants throughout the bioturbated layer. Bioturbation may also act by mixing the added AC upwards to bind contaminants in material settling after the capping operation (Cornelissen et al., 2015; Abel and Akkanen, 2018). Bioturbation is most active within the upper few mm of the sediments and works slowly but continuously to mix AC upwards into the settling material. If sedimentation rates are too high or bioturbation rates are too low, this mechanism is not powerful enough to immobilize the added contaminants. Thus, inappropriate process rates may explain inefficient binding of post-capping added PCBs, as observed in previous field and mesocosm experiments (Abel and Akkannen, 2018; Gidley et al., 2019). Short time of exposure between contaminant and sorbent may also reduce sorbent efficiencies measured in experimental work.

In the Grenlandfjord experiment, input of resuspended contaminants occurred at moderate rates and the time intervals between measurements were sufficiently long to ensure proper binding between contaminant and sorbent. Furthermore, viable benthic communities were present to impel sufficient upwards transport of AC-sorbent to the sediment surface. Eventually the number of available AC-sorption sites will, however, be exhausted both by decreased concentration of AC and by fouling (blocking of pores in the aged AC-material) (Amstaetter et al., 2012; Cho et al., 2012). In 2018, increased uptake ratios at both AC-fields indicated that these processes were beginning to reduce cap efficiencies (Table 2). Nevertheless, the maintenance of better than 54 and 61% uptake reduction for at least nine years demonstrated the long-term potential of activated carbon to reduce bioavailability of dioxins in contaminated sediments.

4. Conclusions

Uptake of dioxins and furans (PCDD/F-TE) was determined in two sediment-dwelling species and passive samplers exposed in boxcore samples transferred from test fields capped with thin (<5 cm) layers of mineral material with and without added activated carbon. Caps of clay mixed with AC reduced dioxin uptake by 69–88% during the first two years, decreasing to 54–61% nine years after cap placement. These efficiencies compared well with efficiencies reported from other field and mesocosm studies performed over much shorter periods of time. The study has also shown, for the first time, that activated carbon added on the sediment surface has a remarkable capability to mitigate recontamination from dioxins in sediments deposited after cap placement. Uptake reduction determined with the deep-burrowing polychaete *Hediste diversicolor* were generally lower than the reduction determined with the surface-dwelling gastropod *Tritia reticulata* and passive samples exposed in the overlying water. Thin caps without added AC had no significant effects on bioavailability one year and more after cap placement. Unfortunately, these positive effects of AC on reducing contaminant release and bioavailability are accompanied with negative side effects on the benthic communities as reported by Samuelsson et al. (2017); Raymond et al. (in prep.) and Trannum et al. (in prep.). More research on finding optimal AC type, dose and application method is needed, but the method should be considered suitable for cost-efficient remediation in many areas where the inherent communities are severely disturbed by anthropogenic activities.

Credit author statement

Morten Schaanning, project manager at NIVA 2009–2014, project coordinator in 2018, Data curation, Funding acquisition, Investigation, Methodology, Project administration, Validation, Visualization, Writing - original draft, review & editing. Jonas Gunnarsson, project manager at Stockholm University, Data curation, Funding acquisition, Investigation, Project administration, Validation, Writing - review & editing. Bjørnar Beylich, responsible for SPI-data, Data curation, Investigation, Methodology, Validation, Visualization, Writing - review & editing. Espen Eek, project coordinator 2009–2014, project advisor at NGI in 2018, Funding acquisition, Project administration, Writing - review & editing

Declaration of competing interest

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

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Appendix A. Supplementary data

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