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Remediation/Restoration

Ecotoxicological Risk of Trace Element Mobility in Coastal Semiartificial Depositional Areas Near the Mouth of the River Rhine, The Netherlands

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Abstract: Artificial sand replenishments are globally used as innovative coastal protection measures. In these replenishments elevated porewater concentrations of trace elements are found. The present study investigated possible ecotoxicological risks at 2 intertidal depositional sites, the Sand Engine as a recent innovative Dutch coastal management project and a semiartificial tidal flat. Using the sediment quality triad approach, we considered 3 major lines of evidence: geochemical characterization, toxicity characterization using bioassays with the estuarine amphipod *Corophium volutator*, and ecological field survey. In both depositional areas *C. volutator* is at risk: moderate (Sand Engine) and low (tidal flat). For tidal flat, the bioavailability of trace elements differs between the field site and the laboratory. Contamination from arsenic and copper is present, but the low survival rate of *C. volutator* from the bioassay suggests the presence of additional contaminations. The highly morphological dynamic environment of Sand Engine creates a less favorable habitat for *C. volutator*, where local spots with stagnant water can temporarily create hypoxic conditions and sulfate becomes reduced. The dynamic system mobilizes especially arsenic, triggering adverse ecotoxic effects at low original sediment concentrations. To conclude, the sediment quality triad approach shows that a semiartificial tidal flat is preferred over a highly dynamic coastal management project like the Sand Engine. The Sand Engine concept does not provide suitable conditions for macrobenthos species like *C. volutator*; therefore, limiting the nature development goal set together with the coastal protection goal. Assessing each line of evidence from the approach together with additional measurements established more precise and realistic conclusions, showing that evaluating the contributions of this method is necessary to understand the causes of risk in a site-specific manner. *Environ Toxicol Chem* 2018;37:2933–2946.

Keywords: Ecotoxicology; Risk assessment; Trace metals; Intertidal flat; *Corophium volutator*; Sediment quality triad

INTRODUCTION

Coastal and estuarine waters mostly receive contaminants via onshore anthropogenic activities. These, after transport via rivers or air, can induce toxic effects on benthic fauna and contribute to the degradation of ecosystem function (Eggleton and Thomas 2004; Cantwell et al. 2008; Roberts 2012). In coastal areas, sediments are very important in ecosystem functioning but also act as a sink for anthropogenic contaminants, thereby becoming a chemical stressor on benthic fauna and associated species (Burton and Johnston 2010; Roberts 2012). Intertidal flats are usually one of the sedimentary environments that can be distinguished in coastal areas. Estuarine intertidal flats show a high biological activity (Herman et al. 2001). These sites harbor primary sources of organic matter, the food base for benthic biota, and as a result contain considerable biomass (Heip et al. 1995).

Environmental studies of intertidal flats generally focus on chemistry, both organic (Benlahcen et al. 1997; Moreira et al. 2016) and inorganic (Alshahri 2017; Vetrimurugan et al. 2017). From an ecological perspective, toxic substances such as trace

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elements, polycyclic aromatic hydrocarbons, and polychlorinated biphenyls are regarded as an important quality criterion (Speybroeck et al. 2006). Heavy metal binding on solid compounds is known to be a critical factor for mobility and bioavailability (Peijnenburg et al. 1999) and thus toxicity. Concentrations of trace elements in sediments can exceed those of the overlying water by 3 to 5 orders of magnitude (Bryan and Langston 1992). Resuspension may result in them becoming toxic, especially to burrowing organisms that are in close contact with porewater. Invertebrates such as amphipods bioaccumulate contaminants through their burrowing and feeding activities (Goodyear and McNeill 1999).

In 2011, a new depositional environment, the Sand Engine (SE), was created along the Dutch coast by an innovative coastal management project (Stive et al. 2013). This mega nourishment contains 21.5 million m^3 of sand and rises up to 6 m above mean sea level. For the Sand Engine, sand was dredged 10 km offshore to a maximum depth of 6 m below the sea floor (Fiselier 2010). The Sand Engine was shaped like a large hook, with an initial length of 2.4 km and a width of 1 km that extends offshore (Supplemental Data, Figure S1). It is designed to enable sand transport by natural processes to feed coastal parts in a 20-km stretch within 20 to 30 yr that are vulnerable to erosion. Besides coastal protection, the Sand Engine has recreation and nature development as additional goals. Because of its distinctive shape, a lagoon is present, sheltered from the waves and with variable tidal velocities. As a result, deposition is taking place, and many benthic microalgae are present (I.R. Pit and E.M. van Egmond, personal observation).

The Rhine flows into the sea approximately 10 km from the Sand Engine and is a source of estuarine and marine contamination (e.g., Laane et al. 2013; Ruff et al. 2015). Numerous physical, chemical, and biological processes affect the fate of particles and micropollutants (Schwarzenbach et al. 2017). The sediment grain size distribution affects the environmental fate of micropollutants because the silt–clay fraction increases the sorptive capacity of the contaminants (Villaescusa-Celaya et al. 2000; Cantwell et al. 2008). Sedimentation of silt/clay material in the lagoon of the Sand Engine might therefore reduce the bioavailability of toxic trace elements. In addition, sorption by organic matter and Fe and Mn oxyhydroxides can lower dissolved metal concentrations in the water column (Cantwell et al. 2008), of which organic matter is correlated with the fine fraction because of its high affinity with clay particles (Oades 1984). However, because the morphological dynamics at the Sand Engine governed by waves and vertical tide are high (De Schipper et al. 2016; Luijendijk et al. 2017), remobilization might occur because of resuspension (Zoumis et al. 2001). A previous study found elevated concentrations of trace elements in porewater at Dutch beach nourishment sites, including the Sand Engine, which had resulted from oxidation processes (Pit et al. 2017a). Thus far it is unclear whether these elevated trace element concentrations are also present where local deposition occurs and whether they bear ecotoxicological risks.

The present study assessed sediment quality at the intertidal area of the engineered Sand Engine and a semiartificial depositional area by comparing the ecological, chemical, and physical characterizations of the 2 depositional areas. Both sites show a natural sedimentation because conditions became favorable after coastal management measures. The intertidal areas are near (the Sand Engine) or at (the semiartificial depositional area) the mouth of the river Rhine, which brings anthropogenic contaminants into the coastal environment. We will also reflect on the nature development goal of the Sand Engine concept and discuss whether trace elements will hamper its attainment. We used the sediment quality triad approach (Chapman 1990; Linkov et al. 2009), considering 3 major lines of evidence (LoE): 1) geochemical characterization, 2) toxicity characterization using bioassays, and 3) ecological field survey. The focus was on the estuarine amphipod *Corophium volutator* as a relevant macrobenthic species often used for risk assessment of contaminated estuarine and marine sediments in large parts of northern Europe (Peters and Ahlf 2005). In addition, toxic trace elements present in the sediment were examined, as well as the bioaccumulation in *C. volutator* collected from the 2 field sites.

MATERIALS AND METHODS

Site selection and characterization

Site selection. We compared 2 intertidal areas (Figure 1). The first site was the lagoon of the Sand Engine, located close to Ter Heijde at the Dutch coast (Stive et al. 2013). The Sand Engine is morphologically dynamic because of its development by erosion enhanced by wind, waves, and currents. The lagoon is on the northern side of the peninsula. Since construction, the entrance to the lagoon has developed enormously, changing the width and depth of the tidal channels and, as a result, the tidal flows entering and leaving the lagoon (Luijendijk et al. 2017). The second site was a semiartificial depositional area at Oostvoorne (tidal flat [TF]), located in the northern part of the estuary of the river Rhine (Noest 1991), which is also known as Slikken van Voorne. At Slikken van Voorne, natural sedimentation occurs because conditions became favorable after coastal management measures, that is, shelter from waves and low tidal flow velocities (De Brouwer et al. 2001). Sediment, porewater, and *C. volutator* were sampled in August and September 2016. Per site, 10 locations were sampled (Figure 1) at low tide, along the low waterline where the intertidal flat was accessible and nearly saturated.

Porewater characterization. Rhizons were used to extract porewater from the soil following Seeberg-Elverfeldt et al. (2005). The rhizons are made of a hydrophilic porous polymer tube, with a pore diameter of 0.1 μm, which ensures the extraction of microbe- and colloid-free, ready-to-analyze solution. The outer diameter of a rhizon is 2.4 mm, and the filter section has a length of 10 cm. Porewater samples were taken at depths of 1.5 and 7.5 cm by inserting a plastic platform with 2 holes at these depths into the sediment. Rhizons inserted through the holes extracted sufficient amounts of porewater horizontally from the sediment, allowing minimum disturbance of the sediment. Porewater was stored and taken to the laboratory in a glass vial for the ion chromatography and a polyethylene vial with 0.2 M HNO3 (10 μL HNO_3/10 mL

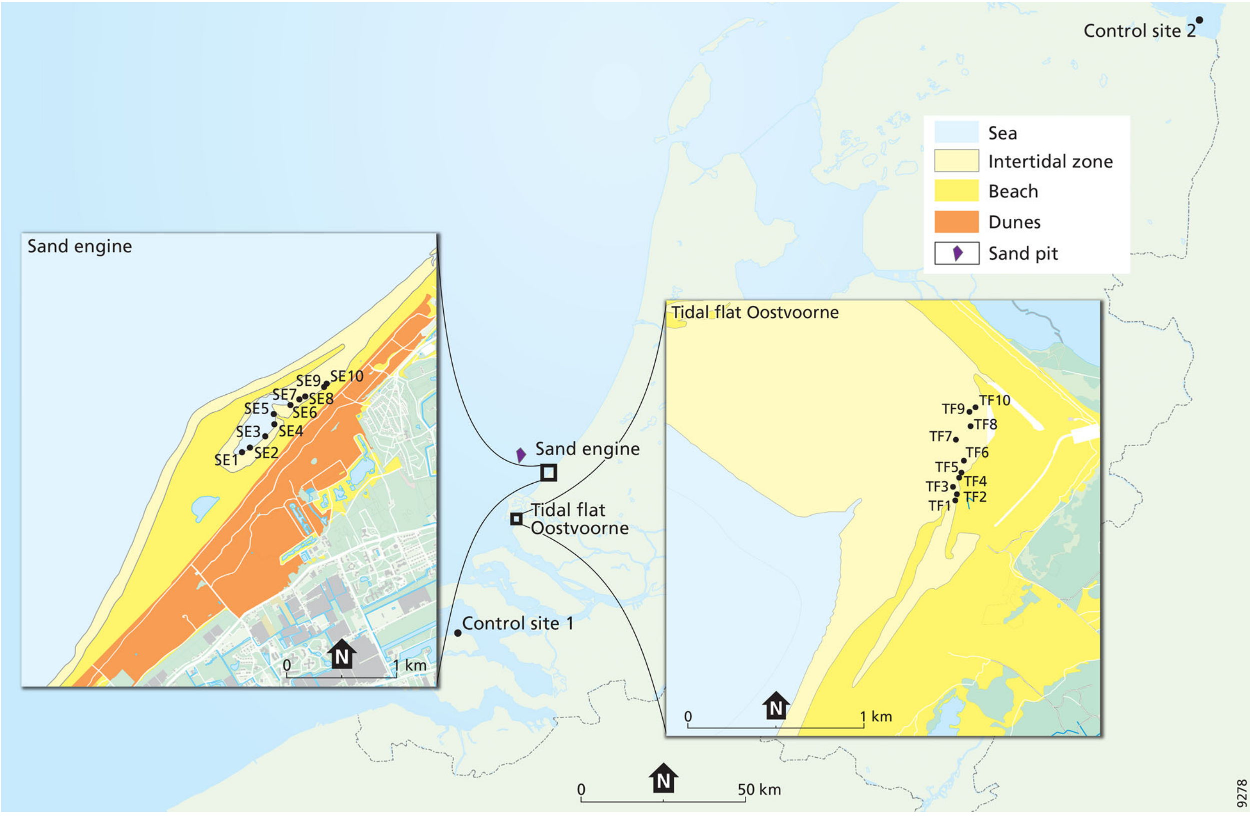

FIGURE 1: Locations of field sites including sample locations: dynamic artificial sand replenishment (Sand Engine) and seminatural tidal flat at Oostvoorne, control site 1, and control site 2. SE = Sand Engine; TF = tidal flat.

porewater sample) for inductively coupled plasma mass spectrometry (ICP-MS).

General characterization of the porewater was done both directly in the field and in the laboratory. In the field, we immediately measured O_2 (Hach Intellical LDO10103), electric conductivity (EC; Hach Intellical CDC401), pH (Hach Intellical PHC101), and redox potential (Pt4805-DPA-SC-S8/225). In addition, porewater was taken to the laboratory in a glass vial and analyzed for total organic carbon (TOC) using a Shimadzu TOC-5050A analyzer. Within 24 h after sampling, alkalinity as HCO_3^- was measured according to Sarazin et al. (1999).

The porewater was analyzed for major ions and trace elements, including rare earth elements using ion chromatography (type 930 Compact IC flex) for (sequenced according to limits of detection [LOD], shown in parentheses) Cl (0.2 mg/L), PO_4 (0.07 mg/L), NO_3 (0.06 mg/L), SO_4 (0.04 mg/L), F (0.03 mg/L), NO_2 (0.02 mg/L), Br (0.002 mg/L), and ICP-MS (type Agilent 7500cx collision cell) for (sequenced according to LOD) Ca (0.15 mg/L); K (0.1 mg/L); Mg (0.05 mg/L); Fe and P (0.04 mg/L); Al (0.01 mg/L); Mn (5 μg/L); Zn (4 μg/L); Sr (1.5 μg/L); Ag and Ba (1 μg/L); Ni (0.8 μg/L); Se and Sn (0.6 μg/L); Cr and Mo (0.5 μg/L); As and Cu (0.4 μg/L); Eu, Gd, Tl, and V (0.3 μg/L); Ce, Co, Cs, Dy, Er, Ga, Ho, In, La, Lu, Nd, Pb, Pr, Rb, Sb, Sm, Tm, U, and Yb (0.2 μg/L); and Be and Cd (0.1 μg/L).

To elucidate whether oxidation or reduction processes influenced the porewater, the measured SO_4 concentration was compared with the SO_4 concentration of conservative mixing of freshwater and seawater. First, the fraction of seawater (f_{sea}) was calculated from the Cl concentration:

$$f_{sea} = \frac{R_{Cl,sample} - R_{Cl,fresh}}{R_{Cl,sea} - R_{Cl,fresh}} \tag{1}$$

In Equation 1 $R_{Cl,sample}$ is the Cl concentration of the sample; $R_{Cl,fresh}$ is the Cl concentration in rainwater, which is assumed to be similar to water from the river Rhine; and $R_{Cl,sea}$ is the Cl concentration in seawater. Then, the conservative mixing concentration ($R_{SO_4,mix}$) is calculated as follows:

$$R_{SO_4,mix} = f_{sea} \cdot R_{SO_4,sea} + (1 - f_{sea}) \cdot R_{SO_4,fresh} \tag{2}$$

In Equation 2 $R_{SO_4,sea}$ and $R_{SO_4,fresh}$ are the SO_4 concentrations in seawater and freshwater, respectively. The rainwater composition with 2.4 ppm SO_4 was derived from Stolk (2001) and that of seawater from Hem (1985) with 2700 ppm SO_4. Finally, the enrichment or depletion of SO_4 (ΔR_{SO_4}) was obtained as follows:

$$\triangle R_{SO_4} = R_{SO_4,sample} - R_{SO_4,mix} \tag{3}$$

A positive ΔR_{SO_4} indicates that porewater is enriched with SO_4, and this enrichment is most likely associated with oxidation of sulfide minerals such as pyrite; a negative ΔR_{SO_4} indicates that

SO_4 has been depleted, which is normally the result of SO_4 reduction in the environments studied.

Sediment characterization. Approximately 500 g of surface sediment was sampled by pooling 3 subsamples: one taken at the center of the sample point and 2 taken 0.5 m from the center. The sediment was sampled with a plastic scoop, and each subsample was taken down to a depth of 10 cm. Sediment samples were oven-dried at 105 °C for at least 24 h. Grain size distribution was measured with a laser diffraction analyzer (Malvern Instruments) in accordance with NEN 5753 (1990). The samples were then dry-sieved in a plastic sieve with nylon cloth to obtain 2 fractions <63 μm (>250 mesh) and 63 to 2000 μm (10–100 mesh). The large fraction was ground into particles <2 μm with a Herzog HP-PA grinding machine. This resulted in contamination with Co, and consequently, the ground fraction could not be used for interpretation on Co. For quantitative determination of mineral compounds, including organic matter, both fractions were subjected to thermogravimetric analysis (TGA) by heating them from 25 to 1000 °C at a rate of 1 °C/min, using a Leco TGA-601. The method used to estimate organic material and carbonate minerals from the TGA data is described in Pit et al. (2017b). The aqua regia digestion method (Houba et al. 1995) was used to obtain the pseudo-total elemental concentrations of the sediment (Chen and Ma 2001), which were then analyzed with inductively coupled plasma optical emission spectrometry (ICP-OES) and ICP-MS to obtain main and trace elements similar to those in the porewater.

Bioassay characterization. Sediment for the bioassay was sampled from 2 locations at tidal flat and Sand Engine. A control was performed with sediment from a control site (CS1), an intertidal mudflat (Oesterput) in the Eastern Scheldt in the southwest Netherlands (Figure 1), which is commonly used as a source of control sediment in toxicity tests (e.g., Stronkhorst et al. 2004; Schipper et al. 2008). Because the sampling locations at tidal flat did not significantly differ in terms of numbers of *C. volutator*, sediment from the first and last transects (TF1 and TF10) was chosen for the bioassay. For Sand Engine, we used sediment from SE5, which contained the highest number of *C. volutator*, and from SE10, which contained a high clay/silt fraction without the presence of *C. volutator*. At each location, a 10-L bucket was filled with sediment and overlying water, closed with a lid, and transported to the laboratory the same or the following day. In September 2016 *C. volutator* individuals needed for the bioassay were sampled at CS1 by sieving the first 2 cm of sediment over a 1-mm sieve and separating *C. volutator* from the other species.

Prior to the experiments the sediment was homogenized and wet-sieved over a 500-μm sieve, except for the control sediment, which contained finer material and was therefore sieved over a 250-μm sieve. Artificial seawater containing 15‰ salt was used for sieving. In total 5 sediments (SE5, SE10, TF1, TF10, and CS1) were sieved and included in the bioassay, which was carried out in a climate chamber at 17 °C in 2 phases over a period of 7 wk.

Ecology characterization. The ecological field survey consisted of *C. volutator* field density and species richness. To estimate *C. volutator* field density, we sampled the population at SE5, SE10, TF1, and TF10. Each sample was taken with a stainless steel rectangular corer of 27 by 37 cm (surface area 999 cm^2) up to a depth of 15 cm. Sediment was sieved over a sieve with 1 mm mesh size, and the macroinvertebrates that remained were placed in a plastic container with 70% ethanol. In the laboratory, the *C. volutator* individuals were counted.

The species richness values of Sand Engine and tidal flat were obtained from parallel studies: that for Sand Engine was obtained from a study within the same project (van Egmond et al. 2018) and that for tidal flat was obtained from van der Zee et al. (2018).

As a control needed for the lines of evidence output, previously published information on *C. volutator* field density and species richness (Dekker and Waasdorp 2006) at Heringplaat (CS2) was used because of a similar grain size distribution. Control site 2 is relatively clean intertidal mudflat with 99% survival rates of *C. volutator* as a bioassay result (Van den Brink and Kater 2006).

Sediment quality triad

The sediment quality triad (from now on referred to as the triad) approach considers 3 lines of site-specific evidence: chemical characterization (e.g., total concentration, bioavailable concentration, bioaccumulation), toxicology (e.g., bioassay of the sediment, biomarkers), and in situ parameters (e.g., benthic community structure, field observations of vegetation; Chapman 1990; Jensen and Mesman 2006; Swartjes et al. 2012). To integrate the results of the different lines of evidences and obtain a triad value for each site, we used the following (Ribé et al. 2012):

$$Triad\ effect\ value = 1 - ((1 - LoE_{CHEM})\cdot (1 - LoE_{BIO})\ (1 - LoE_{ECO}))^{\frac{1}{3}} \tag{4}$$

The triad effect value is a result on an effect scale from 0 to 1, which corresponds to no effect up to maximum effect (Mesman et al. 2006). For each line of evidence, a tool is selected as input for the triad, of which the outcome is projected on the effect scale. Here, the LoE_{CHEM} consists of estimating the bioavailable concentration by quantifying the local toxic pressure of a mixture of compounds. This is done by using the multiple substance potentially affected fraction of species (msPAF; Posthuma and de Zwart 2012). The msPAF value is estimated using the species sensitivity distribution concept. The LoE_{BIO} is a chronic sediment toxicity test consisting of a 49-d exposure with *C. volutator*. A chronic bioassay was chosen instead of a short-term bioassay because of its higher sensitivity to pollution and its environmentally realistic exposure scenarios (van den Heuvel-Greve et al. 2007). The LoE_{ECO} consists of 2 different parameters: the field density of *C. volutator* and the species richness. Two in situ parameters were chosen to have a more realistic overview of the ecosystem where the focus is not just on one species. Following is an elaborated description of each lines of evidence.

LoE$_{CHEM}$. The first lines of evidence is geochemical characterization (LoE$_{CHEM}$) using the chemical elements As, Ba, Be, Cd, Co, Cr, Cu, Mo, Ni, Pb, Tl, Sb, Sn, V, and Zn. The msPAF was used to quantify the ecotoxicological risk based on porewater concentrations (de Zwart and Posthuma 2005; Posthuma et al. 2016). The PAF was calculated using SEDIAS (SEDIment ASsistant; see Hin et al. 2010) from the chronic effect concentrations, using the following equation (Mesman et al. 2006):

$$\mathrm{PAF} = \frac{1}{1 + e^{\frac{\log EC50 - \log C_{pw}}{\beta}}} \tag{5}$$

The EC50 value is the effect concentration for 50% of the macrobenthos species (which includes *C. volutator*), C_{pw} is the porewater concentration (Supplemental Data, Table S1), and β is the standard deviation of the average log effect EC50 concentration. Subsequently, msPAF values were derived via the following equation (Mesman et al. 2006):

$$\mathrm{LoE_{CHEM}} = \mathrm{msPAF} = 1 - \left[\left(1 - PAF\right)_1 \cdot \left(1 - PAF\right)_2 \cdot \left(1 - PAF\right)_n\right] \tag{6}$$

Only PAF values >0.03 were included for the calculation of msPAF because of uncertainties with low PAF values. The input for the triad was the average msPAF value between the 2 depths. A threshold of 0.05 is often used for deriving environmental quality standards (e.g., van Wezel et al. 2000; Maltby et al. 2005) at which 5% of the species will exhibit >50% effects (Posthuma and de Zwart 2012; Posthuma et al. 2016).

LoE$_{BIO}$. The second line of evidence consists of a bioassay (LoE$_{BIO}$) to test chronic sediment toxicity with the *C. volutator* (van den Heuvel-Greve et al. 2007). A total of 800 individuals between 0.5 and 1 mm in size were selected and kept in two 6-L aquaria with a substrate of 4 cm clean silt and artificial seawater (20‰ salt; Instant Ocean) and aerated with compressed air. The aquaria were placed in a climate chamber at 17 °C with a 8:16-h light: dark regime, and the animals were fed powdered fish food ad libitum every other day. After 3 wk, all female *C. volutator* with a brood sac filled with eggs were selected and kept in the aquarium for another week, until juveniles were born. Juveniles aged between 0 and 7 d were used for the bioassay.

In the first phase, 150 randomly selected *C. volutator* individuals were grown in a 6-L aquarium with a 3-cm-thick substrate of one of the test sediments overlain by approximately 10 cm of artificial seawater (20‰ salt; Instant Ocean). The aquaria were covered with transparent plexiglass and aerated with compressed air. After an incubation period of 21 d, mortality was determined. For the second phase, the surviving *C. volutator* individuals were placed in smaller aquaria with fresh sediment that had undergone the same preparation as for the first phase. Approximately 250 to 300 mL of sediment was divided over four 1000-mL glass beakers, and 700 to 750 mL of artificial seawater (20‰ salt; Instant Ocean) was added. Because of the low survival of *C. volutator* in the first phase for some sediments, there were either 2 (TF1, TF10, and SE10) or 4 (CS1 and SE5) replicates, each containing 20 randomly selected surviving juveniles from the first phase. After 4 wk, the surviving adults were collected to determine survival percentage, gender, the number of eggs carried by the females, and body length. The value used for the weight of evidence is the growth inhibition (percentage divided by 100) vis-à-vis the control sample (CS1; Ribé et al. 2012):

$$\mathrm{LoE_{BIO}} = \frac{R_{sample} - R_{control}}{1 - R_{control}} \tag{7}$$

After phases 1 and 2, the *C. volutator* were placed on a Petri dish for 12 to 24 h to empty guts and then frozen until further analyses (see above, *LoE$_{CHEM}$*).

LoE$_{ECO}$. The ecological field survey for the third line of evidence (LoE$_{ECO}$) consists of determining the field density of *C. volutator* and the species richness at each field site. The ecological field survey was scaled in accordance with Ribé et al. (2012) and with CS2 as $R_{control}$:

$$R_1 = \left|\log\left(\frac{avg(R_{sample})}{avg(R_{control})}\right)\right| \tag{8}$$

$$R_2 = 1 - 10^{(-R_1)} \tag{9}$$

Equations 7 and 8 were used for the species richness as well as for the *C. volutator* density. The results were integrated into one line of evidence using Equation 10:

$$\mathrm{LoE_{ECO}} = 1 - [(1 - R_{2,density\ C.\ vol})(1 - R_{2,\ species\ richness})]^{1/2} \tag{10}$$

Bioaccumulation

Individual specimens of *C. volutator* were collected in the field together with the chemical characterization of the fine (<63 μm) fraction to calculate the sediment biota sediment accumulation factor (BSAF) as a field-based measurement endpoint to help reconcile potential differences between the results of the different lines of evidences. Because of low population density at Sand Engine, 20 to 28 adult *C. volutator* individuals were taken per sampling location, compared with 38 to 45 individuals at tidal flat. Individuals were placed in a glass vial and stored in 70% ethanol. Both the individuals stored in ethanol in the field and the frozen individuals from the bioassay were placed in a clean glass vial and dried in the oven for 72 h at 70 °C. They were then subjected to aqua regia digestion and analyzed by ICP-OES and ICP-MS as described for the sediment samples. Because of a low input weight, the results were corrected for the blanco results to prevent any increased concentrations from contamination. When samples contained an input weight of <0.0010 g, the results were not used because of the errors for each variable, which were increased by the high dilution factor. In addition, when the values were below detection limit, the results were not included in the analyses of BSAF because of the high uncertainty. The results of the frozen *C. volutator* individuals from the bioassay contained 2 to 4 replicas and were averaged to obtain one value for each sample location (Bio_SE5, Bio_SE10, Bio_TF1, Bio_TF10, and CS1). The BSAF value was calculated using the following equation:

$$\mathrm{BSAF} = \frac{C_b}{C_s} \tag{11}$$

In Equation 11 C_b is the concentration of the trace metals in biota (milligrams per gram), in this case *C. volutator*, and C_s is the concentration in sediment in the fine fraction (milligrams per gram).

Statistical analyses

Statistical analyses were performed using SPSS (Ver 24) for significant differences between sites or sediment fractions. Prior to the statistical analysis, values below detection limit were set at half the detection limit (Reimann et al. 2008). When >25% of the values of a variable for the chemical characterization of the sediment were below the detection limit, the variable was excluded from the statistical analysis. For the chemical characterization of the porewater, this was not needed because of the possibility to exclude the values which were below detection limit to calculate the msPAF.

All chemical variables were transformed using the isometric log-ratio (ilr) transformation (Egozcue et al. 2003), preferred over a normal log-ratio transformation because it generates a robust estimation of the covariance matrix (Filzmoser et al. 2009). The ilr-transformed data were then back-transformed to centered log-ratio space to maintain the links to the original variables and enable direct interpretation. However, for the triad, the results of each line of evidence were not transformed prior to statistical analyses because of the scaling of the results between 0 and 1.

For significance tests comparing 2 data sets, the Mann-Whitney U test or independent *t* test was used, depending on whether or not the data are normally distributed. When multiple groups of normally distributed data were compared, an independent analysis of variance was performed with a post hoc test. The homogeneity of variances varied significantly per variable, and therefore a Games-Howell post hoc test was chosen (Field 2009). When a normal distribution was not found, an independent nonparametric Kruskal-Wallis test was performed.

Before integrating the lines of evidence results of the chemical characterization per site (msPAF) with the sediment toxicity test (growth inhibition of *C. volutator*) and ecological field survey (density determination of *C. volutator* and species richness), a bootstrap was performed (Efron and Tibshirani 1993). Bootstrapping estimates the properties of the sampling distribution from the sample data, resulting in a more realistic standard deviation when the original sample data comprises a small amount. The sample data are treated as a population (Field 2009), where a total of 100 samples had been randomly drawn from lines of evidence results. However, bootstrapping is only possible when the lines of evidence results are $n > 1$ with nonequal values. From the bootstrapping, a mean and standard deviation are calculated and used for triad input.

RESULTS

General characterization

First, the chemical characteristics of the sediment for the total sediment and the fine fraction as well as the grain size distribution are shown of the 2 intertidal areas at Sand Engine and tidal flat (Table 1). The intervention value is given to compare it with the chemical characterization of the sand. To do this, the intervention value is corrected from a standard soil with 10% organic material and 25% clay (soil particles <2 μm) to a soil most comparable to sand, with 2% organic material and 2% clay (Ministerie van Volkshuisvesting Ruimtelijke Ordening en Milieu 2013). The sediment contents are below the intervention value, especially the concentrations in the coarse fraction. The differences between tidal flat and Sand Engine are large, both for properties like $CaCO_3$, S, and the median diameter of particle size distribution and for the trace metals. Differences are even more pronounced for the fine fraction.

Information on the porewater quality is presented in Table 2, with averages on field measurements and TOC. Differences between the 2 intertidal areas are significant for TOC, EC, and pH (Mann-Whitney *U*, $p < 0.05$). Overall, porewater at tidal flat is more mixed with freshwater, the pH is slightly lower, and TOC is higher compared to Sand Engine. Redox conditions are comparable; but the highest mean value is at Sand Engine for 1.5 cm depth, and the same trend is seen for the alkalinity. Oxygen concentrations are relatively low at both 1.5 and 7.5 cm depths, with Sand Engine having an average of 1.3 ± 0.5 and tidal flat 1.7 ± 0.4 mg/L at 1.5 cm depth; and at 7.5 cm depth it is a bit higher, with Sand Engine having 1.6 ± 0.4 and tidal flat 1.9 ± 0.6 mg/L. From 2 locations at both Sand Engine and tidal flat, the oxygen concentrations of the surface water were measured, showing an average of 9.3 ± 0.4 mg/L, which is close to saturation with air at equilibrium (Stumm and Morgan 1996), illustrating the accuracy of the measurements.

Figure 2 confirms that tidal flat is more fresh compared to Sand Engine, where the concentrations of SO_4 were plotted against Cl together with a mixing line of rainwater and seawater. At tidal flat an average of 6904 ± 758 mg Cl/L was observed compared to $13\,077 \pm 1840$ mg Cl /L at Sand Engine. The SO_4 concentrations are mainly below the mixing line, which indicates reduction processes. However, the relative distance from the mixing line is small; and with ΔR_{SO_4} showing an average of −90 mg/L for Sand Engine and −44 mg/L for tidal flat, reduction may be dominant, but oxidation processes are likely to play a role as well. To get more information on the dominance of anoxic or oxic processes present at the 2 study sites, Fe and Mn in

TABLE 1: Chemical characteristics of the sediment at Sand Engine (SE) and tidal flat (TF) for total sediment and the <63-μm fraction[a]

		Total sediment					<63 μm				
		SE		TF			SE		TF		
	Intervention value[b]	Mean (SD)	Max	Mean (SD)	Max	Sig.[c]	Mean (SD)	Max	Mean (SD)	Max	Sig.[c]
Organic C (%)		0.9 (0.3)	1.6	1.8 (0.56)	2.8		7.7 (1.1)	9.21	5.4 (1)	6.5	**
$CaCO_3$ (%)		4.1 (1.5)	7.3	5.6 (1.7)	9.5	*	12 (3.1)	20.76	7.6 (1.6)	9.1	*
S (%)		0.07 (0.03)	0.1	0.06 (0.02)	0.1	*	0.9 (0.06)	1.0	0.3 (0.1)	0.5	**
Fe (%)		0.4 (0.08)	0.5	0.5 (0.2)	0.8	*	1.7 (0.2)	1.9	1.5 (0.1)	1.7	**
Fraction <63 μm (%)		1.4 (1.1)	2.8	4.4 (3.1)	9.9						
D50		349 (31)	389	210 (40)	259	**					
As	44	2.8 (0.8)	4.2	5.0 (1.1)	7.6		11 (2.1)	14	15 (4.0)	21	**
Ba	237	22 (2.2)	25	24 (3.1)	31	**	35 (3.5)	38	49 (7.4)	61	**
Be	10	0.17 (0.04)	0.25	0.2 (0.07)	0.3	*	0.6 (0.08)	0.7	0.6 (0.05)	0.7	
Cd	8	0.02 (8.9)	0.04	0.08 (0.04)	0.1		0.2 (0.07)	0.3	0.3 (0.03)	0.3	**
Co	54	—		—			7.3 (1.4)	9.8	5.0 (0.5)	5.7	**
Cr	42	9.1 (1.8)	11.8	10.6 (3.7)	17	**	34 (2.3)	37.8	29 (1.7)	32	
Cu	92	2.6 (1.0)	4.2	3.3 (1.0)	5.4	*	11 (1.7)	14	10 (2.9)	18	
Mo	190	0.12 (0.08)	0.29	0.12 (0.02)	0.15	*	1.1 (0.36)	1.7	0.4 (0.2)	0.7	**
Ni	34	5.6 (0.8)	7.1	4.7 (1.2)	6.5	**	20 (2.7)	23	13 (2.0)	16	**
Pb	337	3.1 (0.9)	4.9	7.5 (2.0)	11		20 (2.4)	23	23 (4.3)	29	**
Tl	15	0.04 (0.07)	0.21	0.2 (0.05)	0.2	*	<d.l.		<d.l.		
Sb	22	0.03 (0.04)	0.14	0.08 (0.03)	0.1		<d.l.		<d.l.		
Sn	246	0.1 (0.2)	0.44	0.4 (0.1)	0.6		<d.l.		<d.l.		
V	86	6.9 (2.1)	10	11 (4.0)	17	*	37 (5.4)	43	31 (2.3)	35	**
Zn	303	10 (2.9)	17	27 (6.7)	40		65 (11)	84	81 (11)	98	**

[a]Concentrations of trace elements are in milligrams per kilogram.
[b]The intervention value is corrected from a standard soil (10% organic material and 25% lutum) to a soil most comparable to sand (2% organic material and 2% clay).
[c]Significance estimated for differences between SE and TF with a Mann-Whitney U.
*$p < 0.05$, **$p < 0.001$.
D50 = median diameter of particle size distribution; d.l. = detection limit; Max = maximum; Sig. = significance.

porewater as well as those of the sediment are plotted in Figure 3. Iron concentrations at tidal flat are on average higher compared to Sand Engine for 1.5 cm depth: 4.3 ± 3.4 versus 2.0 ± 2.0 mg/L, although the difference is not significant (Mann-Whitney *U* test, $p < 0.05$). For 7.5 cm depth, Fe concentrations are slightly higher at Sand Engine compared to at tidal flat (3.5 ± 2.4 vs 3.1 ± 1.9 mg/L) and not significant. Manganese concentrations in the porewater are for both 1.5 as well as 7.5 cm depths higher at tidal flat (1.1 ± 0.8 and 1.1 ± 0.6 mg/L) compared to Sand Engine (0.7 ± 0.6 and 0.6 ± 0.4 mg/L) and not significant. The mutual occurrence of oxygen and dissolved Fe and Mn indicates that the sediments are not in redox equilibrium at the sample scale, suggesting ongoing redox reactions. Iron sediment content at Sand Engine with an average of 3926 ± 841 mg/kg is significantly lower compared to tidal flat, with an average of 4907 ± 1851 mg/kg (Mann-Whitney *U* test, $p < 0.05$). For Mn, tidal flat has significantly higher concentrations as well, with an average of 179 ± 97 compared to 84 ± 24 mg/kg at Sand Engine (Mann-Whitney *U* test, $p < 0.05$). Both the Fe and Mn sediment contents for tidal flat are increasing with sample location number, coinciding with distance inland (Figure 1).

Sediment quality triad

The LoE_{CHEM} with the chemical characterization of the porewater at depths of 1.5 and 7.5 cm (Table 3) shows that overall the msPAF values for these depths were higher at Sand Engine than at tidal flat (the actual porewater concentrations are presented in Supplemental Data, Table S1). The difference between the 2 sites is significant for the 1.5 cm depth (independent *t* test, $p < 0.05$) but not for the 7.5 cm depth. Arsenic is the main contributor for each msPAF, followed by Cu, Ni, Zn, and Cr (Table 2). The msPAF per study site averaged over

TABLE 2: Means (standard deviation) of different variables measured in the field (O2, EC, pH, and redox) and in the laboratory (total organic carbon [TOC] and alkalinity)[a]

	TOC (mg/L)	O_2 (mg/L)		EC (ms/cm)		pH		Redox (mV)		Alkalinity (mg/L)	
	0–10 cm	1.5 cm	7.5 cm	1.5 cm	7.5 cm	1.5 cm	7.5 cm	1.5 cm	7.5 cm	1.5 cm	7.5 cm
SE	4.4 (1.2)	1.3 (0.5)	1.6 (0.4)	42 (5.8)	39 (5.1)	7.7 (0.6)	7.9 (0.3)	–153 (88)	–128 (22)	251 (121)	233 (62)
TF	5.9 (2.0)	1.7 (0.4)	1.9 (0.6)	22 (3.3)	22 (3.0)	7.6 (0.1)	7.6 (0.3)	–133 (24)	–122 (41)	173 (15)	197 (44)
Sig.	*			*	*	*	*				

[a]Significance was tested using the Mann-Whitney U, *$p < 0.05$.
EC = electric conductivity; SE = Sand Engine; Sig. = significance; TF = tidal flat.

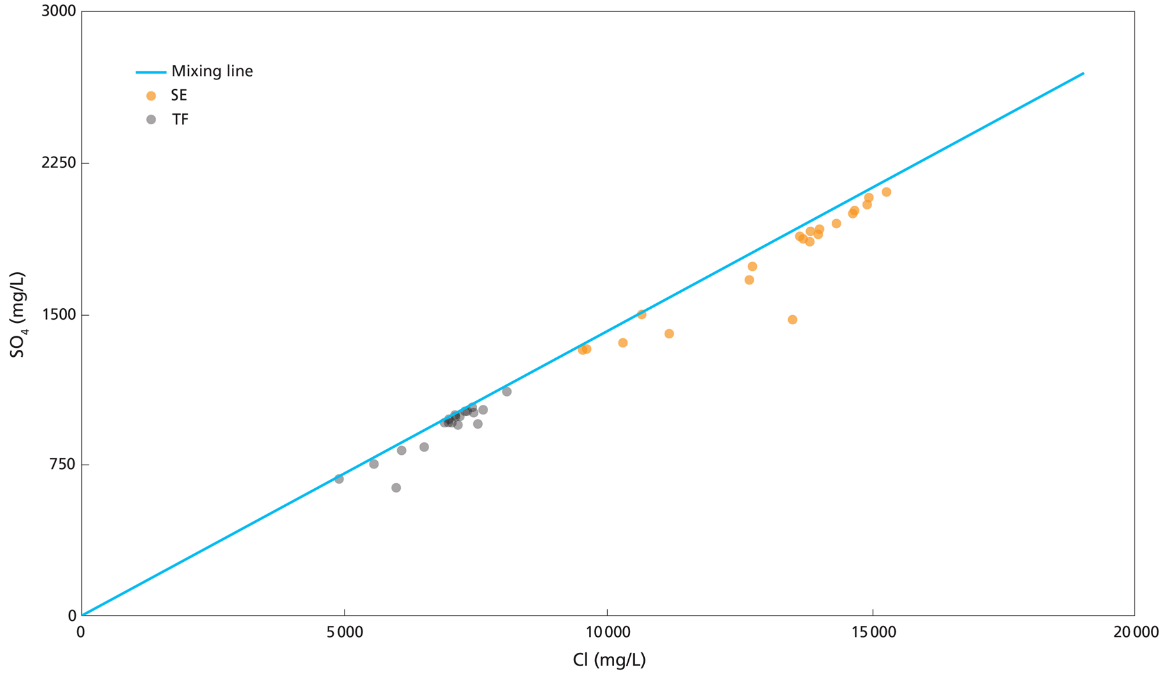

FIGURE 2: Concentrations of SO4 versus Cl for the porewater samples, with the mixing line of rainwater and seawater. SE = Sand Engine; TF = tidal flat.

both depths is 0.22 ± 0.03 for Sand Engine and 0.18 ± 0.04 for tidal flat, with Sand Engine being significantly higher compared to tidal flat (independent *t* test, $p < 0.05$). These averaged msPAF values were used as input values for the triad.

The results of the bioassay (LoE_{BIO}) are given in Table 4 and Supplemental Data, Table S2. In the first part of the bioassay, 21-d population growth inhibition is shown for *C. volutator*. Subsequently for the second generation, growth of the

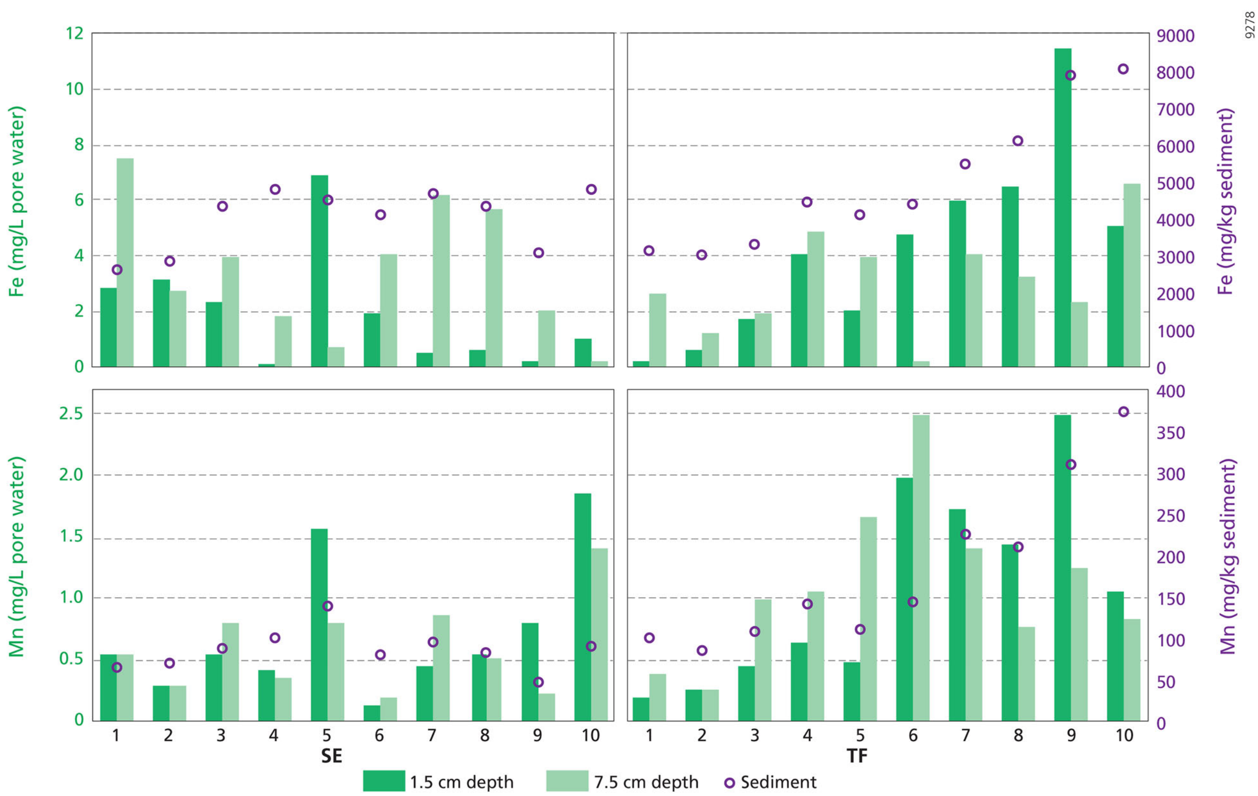

FIGURE 3: Concentrations of Fe (top graphs) and Mn (bottom graphs) with bars for the porewater concentrations, colored dark for the 1.5 cm depth concentration and light gray for the 7.5 cm depth concentration. Numbers represent the different sample locations. Sediment concentrations are shown by open circles. SE = Sand Engine; TF = tidal flat.

TABLE 3: Chemical characterization of the porewater at 1.5 and 7.5 cm depth and potentially affected fraction values of species per trace element and for multiple substances

	PAF												msPAF		msPAF
	As		Cu		Ni		Zn		Cr		Be	V			
	1.5 cm	7.5 cm	1.5 cm	7.5 cm	1.5 cm	7.5 cm	1.5 cm	7.5 cm	1.5 cm	7.5 cm			1.5 cm	7.5 cm	Average
SE1	0.25	0.26											0.25	0.26	0.26
SE2	0.23	0.22											0.23	0.22	0.23
SE3	0.19	0.20		0.04									0.19	0.23	0.21
SE4	0.16	0.19											0.16	0.19	0.18
SE5	0.20	0.11	0.04	0.03		0.05							0.23	0.20	0.22
SE6	0.16	0.18					0.07						0.22	0.18	0.20
SE7	0.17	0.20	0.05	0.05									0.21	0.24	0.23
SE8	0.19	0.21			0.12								0.29	0.21	0.25
SE9	0.21	0.16											0.21	0.16	0.19
SE10	0.29	0.19	0.05						0.03				0.35	0.19	0.27
TF1	0.12	0.21	0.08										0.19	0.21	0.20
TF2	0.11	0.13											0.11	0.13	0.12
TF3	0.14	0.17											0.14	0.17	0.16
TF4	0.18	0.18	0.06	0.07									0.23	0.23	0.23
TF5	0.17	0.18		0.07									0.17	0.23	0.20
TF6	0.16	0.10											0.16	0.10	0.13
TF7	0.18	0.16											0.18	0.16	0.17
TF8	0.17	0.13											0.17	0.13	0.15
TF9	0.23	0.15	0.07	0.04									0.29	0.19	0.24
TF10	0.20	0.22											0.20	0.22	0.21

msPAF = multiple substance potentially affected fraction; PAF = potentially affected fraction; SE = Sand Engine; TF = tidal flat.

individuals and population growth inhibition are given. For CS1 and SE5 enough individuals were present after phase 1 for 4 replicas in phase 2; for the other sites 2 replicas were possible. Control site 1 shows no population growth inhibition and the highest individual growth in length, followed by Sand Engine and then tidal flat. The differences for the population growth inhibition between the 2 phases are large. As input for the triad the location averages of population growth inhibition of phase 2 were used, reflecting a long exposure period. The average values in phase 2 are 0.00 ± 0.00 (Sand Engine) and 0.58 ± 0.18 (tidal flat), which significantly differ (Games-Howell, $p < 0.05$).

The results of the ecological field survey (LoE_{ECO}), including numbers of *C. volutator* per square meter and species richness,

TABLE 4: Results of the bioassay with the population growth inhibition for phases 1 and 2 and individual growth in length for *Corophium volutator* for phase 2

	Population growth inhibition, % (SD)	Growth in length of *C. volutator* including antennae, mm (SD)
Phase 1 (21 d)		
SE5	19.3	
SE10	58.7	
TF1	70.7	
TF10	67.3	
CS1	9.3	
Phase 2 (49 d)		
SE5 ($n = 4$)	0 (0)	5.4 (1.4)
SE10 ($n = 2$)	0 (0)	9.6 (1.9)
TF1 ($n = 2$)	70 (5)	2.8 (1.2)
TF10 ($n = 2$)	45 (0)	2.8 (0.4)
CS1 ($n = 4$)	0 (0)	13.1 (2.8)

CS = control site; SD = standard deviation; SE = Sand Engine; TF = tidal flat.

are presented in Table 5. The average number of *C. volutator* is lower for Sand Engine (215 ± 304), which significantly differs from the high numbers for CS2 ($12\,535 \pm 2496$; Games-Howell, $p < 0.05$). For the species richness, tidal flat and Sand Engine are more similar (3.3 ± 1.1 vs 3.6 ± 3.4) and CS2 has the greatest species richness (17 ± 1.0), with CS2 being significantly different from tidal flat (Games-Howell, $p < 0.05$). Both input values were first scaled vis-à-vis CS2, which gives 0.98 (Sand Engine) and 0.29 (tidal flat) for the number of *C. volutator* and 0.75 (Sand Engine) and 0.82 (tidal flat) for the species richness. Then, the 2 values for each site were integrated as one value for the triad, the LoE_{ECO}.

Integrated results for the triad effect value are given in Table 6. Bootstrapping was done for LoE_{CHEM} and LoE_{ECO} but could not be performed for LoE_{BIO} because Sand Engine input values equaled 0 and bootstrapping of the LoE_{BIO} of tidal flat would create spurious comparisons between the 2 sites. The calculated triad effect value is significantly higher for Sand Engine (0.60 ± 0.17) compared to tidal flat (0.50 ± 0.12; Games-Howell, $p < 0.001$).

Bioaccumulation

To understand whether *C. volutator* is affected by the fine fraction, BSAF values are calculated (Table 7). For Se, Sb, Sn, and Tl BSAF was not calculated because the majority of the values from both sediment and *C. volutator* were below the detection limit. Also, for TF10 the *C. volutator* numbers were too low to obtain accurate results, and for Sand Engine only 5 locations are visible because no *C. volutator* was found at the other locations. For many elements BSAF values are higher at tidal flat compared to Sand Engine, indicating higher bioavailability for uptake of the trace elements at tidal flat. However for As, which drives msPAF,

TABLE 5: Results of the ecological field study, with means standard deviation (SD)[a]

	No. Corophium volutator per square meter				Species richness per square meter			
	n	Mean (SD)	Sig.	Reference	*n*	Mean (SD)	Sig.	Reference
SE	2	215 (304)	B		4	4.1 (3.9)	AB	van Egmond et al. (2018)
TF	2	8960 (2517)	AB		20	3.0 (1.0)	A	van der Zee et al. (2018)
CS2	3	12 535 (2496)	A	Dekker and Waasdorp (2006)	3	17 (1.0)	B	Dekker and Waasdorp (2006)

[a]Sites that show a significant difference are listed with a different letter (Games-Howell, $p < 0.05$).
CS = control site; SE = Sand Engine; Sig. = significance; TF = tidal flat.

BSAF is higher at Sand Engine. The BSAF values from the bioassays are for several elements (As, Ba, Ni, Zn) higher compared to the field sites but certainly not for all chemicals.

DISCUSSION

The present study investigated trace element contamination and ecotoxicological risks at 2 semiartificial depositional sites along the Dutch coast, a recent artificial nourishment (Sand Engine) and a semiartificial tidal flat. Using the sediment quality triad approach (Chapman 1990; Linkov et al. 2009), we considered 3 major lines of evidences: 1) chemical characterization of porewater, 2) toxicity characterization of the sediment using bioassays, and 3) ecological field survey. The results showed a higher triad effect value at Sand Engine (0.60 ± 0.17) than at tidal flat (0.50 ± 0.12) and can be quantified as a moderate risk and a low risk, respectively (Jensen and Mesman 2006). Combining different lines of evidences in one number might destroy distinct information (Suter and Cormier 2011). Therefore, each line of evidence will be discussed in detail.

For LoE_{CHEM}, the msPAF averages of 0.22 ± 0.03 for Sand Engine and 0.18 ± 0.04 exceed 0.05, the threshold. For both Sand Engine and tidal flat, the main and often only contributor to the msPAF values is As in the porewater. If, instead of using directly measured porewater as input for the msPAF, the msPAF was based on measured sediment contents that were standardized and translated to porewater concentrations assuming equilibrium partitioning using K_d, this would result in lower msPAF values of 0.04 ± 0.01 for Sand Engine and 0.05 ± 0.01 for tidal flat. This points to the relevance that equilibrium partitioning is not a cautious assumption in this case. A physical process that would enhance mobilization is resuspension, which results in the oxidation of the sediment and therefore remobilization of the trace elements (Cantwell et al. 2008). Besides aerobic conditions, reducing conditions were observed based on the SO_4 measurements compared to the composition of conservative mixing of freshwater and seawater and the dissolved Fe and Mn concentrations. The presence of anoxic microniches with sulfidic conditions within suboxic surface sediment may explain the presence of both aerobic and anaerobic conditions (Van Cappellen and Wang 1996; Stockdale et al. 2010), which can be a consequence of bioturbation (Bertics and Ziebis 2010). As a result, the msPAF values show the mobilized trace elements at the time of sampling, which are influenced by bioturbation and probably resuspension. Redox conditions are an important factor to control the bioavailability of trace elements. A low redox potential leads to a low mobilization of cationic metals like Cd, Cu, Ni, and Zn but promotes the mobility of oxyanion As (Frohne et al. 2011), which is the case for both sites, especially at Sand Engine. Increased As can be attributed to the reduction of Fe oxyhydroxides and the reduction of arsenate to arsenite (Mitsunobu et al. 2006; Frohne et al. 2011).

TABLE 6: Results of the 3 lines of evidence (LoE) for Sand Engine (SE) and tidal flat (TF)

Scaled/combined LoE	SE	TF
LoE_{CHEM}	0.22 (0.03)	0.19 (0.04)
LoE_{BIO}	0.00 (0.00)	0.58 (0.15)
LoE_{ECO}	0.87 (0.11)	0.56 (0.27)
Triad effect value	0.60 (0.17)	0.50 (0.12)

It can be questioned whether influences of the Rhine are apparent, when marine sediments containing sulfide minerals like pyrite are common (Johnston et al. 2010). To estimate whether Rhine water influences occur at Sand Engine and tidal flat, rare earth elements were analyzed (Supplemental Data, Table S3) because the Rhine has high concentrations of La and Gd (Moermond et al. 2001; Kulaksiz and Bau 2011). It is clear that the sediments along the Dutch coast and in the Wadden Sea show influences from La and Gd (Moermond et al. 2001; Van den Brink and Kater 2006). However, it is not known whether the influence of the Rhine has increased after the construction of the Sand Engine because of similar concentrations 10 km out from shore (Supplemental Data, Table S3), the location of which is equal to the distance from where the sand from the Sand Engine was collected out of a sand pit (Pit et al. 2017b). Further research and correction to background values of La and Gd (Klaver et al. 2014) is needed to estimate the influence from the suspended solids and freshwater originated from the Rhine. In addition, organic pollutants in sediments from 2 locations (one in front and one behind the low waterline) at both Sand Engine and tidal flat were all found to be below their respective detection limits (Supplemental Data, Table S4). As a result, pollution originating from the Rhine seems apparent at both sites, but the intensity at the time of sampling was most likely low.

The LoE_{BIO} yielded results contradicting those of the third line of evidence, the ecological field survey. Although the field-estimated number of *C. volutator* at tidal flat is relatively high, the bioassay results indicate that the environment for this species is contaminated because survival is low. The opposite is the case for Sand Engine, where low numbers of *C. volutator* were found in the field; but the bioassay results were similar to those for the control

TABLE 7: Average biota-sediment accumulation factor values for trace elements, with the means (standard deviation [SD]) for Sand Engine (SE), tidal flat (TF), and bioassay[a]

Corophium volutator and sediment	As	Ba	Be	Cd	Cr	Co	Cu	Mo	Ni	Pb	V	Zn
SE	0.3 (0.1) A	0.6 (0.3) A	0.1 (0.1) AB	0.3 (0.1) A	0.1 (0.1) A	0.1 (0.02) A	3 (0.1) A	0.3 (0.05) A	0.1 (0.04) A	0.1 (0.04) A	0.1 (0.04) A	1.0 (0.1) A
TF	0.2 (0.03) A	1.1 (0.2) B	0.2 (0.1) B	1.4 (0.5) B	0.2 (0.04) B	0.2 (0.05) B	5.7 (1.3) B	1.5 (0.6) B	0.2 (0.1) B	0.1 (0.1) A	0.2 (0.05) B	0.9 (0.1) A
Bioassay	1.1 (0.2) B	7.7 (3.4) AB	0.05 (0.04) A	0.7 (0.6) A	0.02 (–)	0.1 (0.1) AB	1.1 (–)	0.3 (–)	0.4 (0.4) AB	0.1 (0.1) A	0.1 (0.03) A	1.5 (0.4) A

[a]Significance was tested using the Games-Howell post hoc, $p < 0.05$, except when $n = 1$ and no SD is visible.

site. Therefore, we compared differences in the laboratory and field environmental settings: the salinity of the water used in the laboratory (15 and 20‰) coincides with an EC of 21 to 27 mS/cm, and similar EC values were found at tidal flat (~22 mS/cm) but not at Sand Engine (~40.5 mS/cm). Salinities between 15 and 20‰ are preferred for optimum growth and moulting (Mclusky 1967). This factor might explain why Sand Engine sediment in the laboratory was more favorable than sediment in the field but does not explain why tidal flat sediment resulted in such low survival rates during the bioassay. Temperature can also be a factor (Kater et al. 2001), but during sampling in September the field conditions and the laboratory environment were similar and between 15 and 20 °C, a range that does not affect the growth rate of *C. volutator* (Kater et al. 2008). The discrepancy between the bioassay and ecological field survey is likely attributable to differences in bioavailability between the laboratory and the field, as also shown by the different BSAF values for elements driving msPAF (Table 7). A possible explanation for the disparity between the field and laboratory is the sulfide oxidation environment in the laboratory, where absence of binding phases may encourage mobilization of trace elements (Cantwell et al. 2008). The oxidative environment in the laboratory and the preparations prior to the bioassay, like sieving, may have mobilized specific trace elements in the sediment for tidal flat, causing a low survival rate of *C. volutator*.

The reason that field abundance of *C. volutator* was greater at tidal flat than at Sand Engine, which is included in LoE_{ECO}, might be the different histories of the sites. Although both field sites are semiartificial depositional areas, tidal flat has developed more gradually during several decades and the intertidal flat comprises a larger area. Given this, plus the overall eroding conditions at Sand Engine (De Schipper et al. 2016; Luijendijk et al. 2017), it can be assumed that at tidal flat the environment is more stable, favoring higher production by the early summer broods (Gratto and Thomas 1983). The eroding conditions at Sand Engine are desired: the mega nourishment has to be implemented into the beach and dunes within 20 to 30 yr (Stive et al. 2013). Consequently, the dynamic environment at Sand Engine creates spots where refreshing of seawater is absent and stagnant water may occur. Although *C. volutator* might not respond to stagnant water (Kater et al. 2001), it is likely that this species reacts to a hypoxic environment, where its survival rate drastically decreases with time, especially when sulfidic conditions occur (Meadows et al. 1981; Gamenick et al. 1996; Marsden and Rainbow 2004). Nonetheless, *C. volutator* is an opportunistic species, and recovery can be as fast as 2 wk, although responses are successively greater at larger spatial scales (Norkko et al. 2006). However, with the intertidal flat at the Sand Engine having many benthic micro algae (I.R. Pit and E.M. van Egmond, personal observation), hypoxic conditions are more likely to occur and the recovery of *C. volutator* might be slow (Gamenick et al. 1996). Therefore, the stability of the field situation and the large spatial scale at tidal flat may account for the differences in numbers of *C. volutator* between Sand Engine and tidal flat. However, compared to CS2, tidal flat shows lower numbers of *C. volutator*, which, in light of the low survival rates from the bioassay results, may be because of another potential contamination (Suter and Cormier 2011).

Besides the number of *C. volutator*, species richness is also included in the third line of evidence. For both Sand Engine and tidal flat, the species richness was much lower than for CS2, resulting in a high (Sand Engine) or moderately high (tidal flat) value for LoE_{ECO}. Given the very dynamic physical conditions and possibly temporary hypoxic conditions at the Sand Engine site, there might not have been enough time for a high species richness to develop. When the Sand Engine was created in 2011, time was needed to develop mudflat-like conditions and a habitat for species like *C. volutator*. This situation occurred many years ago for tidal flat, and the low numbers for species richness might be caused by another factor, as seen from the bioassay results. Less opportunistic species may not recover as quickly as *C. volutator*, potentially resulting in a lower macrobenthos diversity (Norkko et al. 2006).

In view of the foregoing, it seems that the sediment quality can be evaluated with the triad effect value, but it gives more precise and realistic conclusions when assessing each line of evidence individually. Additional measurements can be included more easily, and a more flexible evaluation is possible. The triad effect value reveals that at the Sand Engine field site the ecotoxicological risk of the sediment to *C. volutator* is higher than at tidal flat. In more detail, it seems that for both field sites another variable is involved, the dynamic environment at Sand Engine and a potentially different contamination at tidal flat, which were not included in the present triad.

With the Sand Engine having recreation and nature development as additional goals to coastal protection, the present study reveals that nature development at the intertidal flat of this mega beach nourishment is restricted because of its dynamic environment. Stagnant water can occur temporarily, where reducing conditions will create a hypoxic environment and macrobenthos species like *C. volutator* may avoid sediments, especially when a sulfidic redox state is present (Meadows et al. 1981). The dominant reducing conditions are causing cationic metals to become less mobilized in the porewater, but the opposite is seen for the oxyanion As. Even though this trace element may create a decreasing water quality, external and internal factors affect bioaccumulation, for example, adaptation, uptake via porewater or sediment, short- and long-term exposure, and the behavior of *C. volutator* (Bat et al. 1998; Marsden and Rainbow 2004). To obtain 3 different goals as a new coastal management technique, the Sand Engine concept has not provided suitable conditions thus far for macrobenthos species like *C. volutator* to establish a stable population. However, when this mega nourishment is compared to a traditional nourishment that needs to be replenished periodically, the Sand Engine does create more opportunities, for example, mudflat conditions, which would otherwise not be a question because traditional beach nourishments show generally a coarse grain size distribution (Pit et al. 2017a) and do not contain intertidal flats as sedimentary subenvironments. One may, however, wonder whether a beach that must be replenished for coastal protection reasons should be partially transformed into an intertidal flat by means of these sand replenishments: is there a need or a benefit for landscape diversification at the small scale in addition to a desire?

CONCLUSIONS

The sediment quality triad method, considering 3 major lines of evidences, was used to investigate trace element contamination and possible ecotoxicological effects for the macrobenthos species *C. volutator* at 2 depositional sites along the Dutch coast: a recent innovative Dutch coastal management project, the Sand Engine, and a semiartificial tidal flat at Oostvoorne. The sediment quality was evaluated with the triad effect value but was more precise and realistic when assessing each lines of evidence individually. Additional measurements can be included easily, and a more flexible evaluation is possible. The triad effect value reveals that the ecotoxicological risk of the sediment to *C. volutator* is higher at the Sand Engine field site than at tidal flat. Evaluation of each line of evidence from the triad method together with additional measurements established that at both sites As is bioavailable. For the semiartificial tidal flat, the sediment quality triad method shows that the bioavailability of trace elements seems to differ between the field site and the laboratory. Contamination from the trace elements As and Cu is present, but the extremely low survival rate of *C. volutator* from the bioassay suggests that additional potential contaminations are present at this tidal flat, which may be why the sediment BSAF was higher here than at the Sand Engine. The highly diverse and dynamic environment of the Sand Engine creates a less favorable habitat for *C. volutator*, where stagnant water can temporarily create hypoxic conditions and sulfate becomes reduced. Because of its shape and location, the Sand Engine has been designed to erode, so it is unlikely that the intertidal flat at the Sand Engine will become stable in time. The dynamic system brings along the mobilization of especially As, which can trigger adverse ecotoxicological effects at low original sediment contents. The Sand Engine site does not provide suitable conditions for macrobenthos species like *C. volutator* to create a stable population. Besides coastal protection, the Sand Engine concept also has recreation and nature development as additional goals. We conclude that the dynamics at the intertidal flat of Sand Engine creates ecotoxicological risks and, as a result, limits the nature development goal of the Sand Engine concept. However, when a beach must be replenished for coastal protection reasons it is questionable whether there is a need or a benefit for landscape diversification at the small scale.

Supplemental Data—The Supplemental Data are available on the Wiley Online Library at DOI:10.1002/etc.4262.

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